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DISSERTATION

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Analysis of the MultiConfiguration Time-Dependent Hartree-Fock Equations

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à ma mère Chedlia, à mon père Mustapha,
à mon frère Wahid et mes soeurs Imen,
Ines et Zeineb.
à mon oncle Fethi.

To my mother Chedlia, to my father Mustapha,
to my brother Wahid and my sisters Imen,
Ines and Zeineb.
To my uncle Fethi.

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Abstract

The multiconfiguration methods are a natural improvement of well-known simple models for approximating the linear N body Schrödinger equation for atomic and molecular systems with binary - Coulomb in realistic situations- interactions, like the Hartree and the Hartree-Fock equation. Models like MCTDHF are intensively used for numerical simulations in quantum physics/chemistry. However, from the mathematical point of view, these equations are yet poorly understood. The present contribution gives the first rigorous mathematical foundation of the MCTHDH(F) equations with the singular Coulomb interaction. In particular, we formulate in a convenient way for the mathematical analysis the associated initial value problem for which we obtain well-posedness results depending on the regularity of the initial data, with and without an assumption on the rank of the associated density matrix. Also, numerical simulations of a toy model are presented with particular interest to the so called *correlation* which is one of the main motivations and advantage of the multiconfiguration methods compared to Hartree-Fock models.

Zusammenfassung

Die Multiconfigurationsmethoden sind eine natürliche Verbesserung der wohlbekannten einfachen Näherungen der linearen N-Teilchen Schrödinger Gleichung für Atom- und Molekül-systeme mit binärer Wechselwirkung, die realistischerweise mit dem Coulomb Potential modelliert wird. Modelle wie MCTDHF werden intensiv verwendet für numerische Simulationen in der Quantenphysik/chemie. Die vorliegende Arbeit legt die ersten mathematisch rigorosen Fundamente der MCTDH(F) Gleichungen mit singulärer Coulomb-Wechselwirkung. Wir formulieren in einer für die mathematische Analysis besonders geeigneten Weise das assoziierte Anfangswertproblem, für das wir Wohlgestelltheitsresultate zeigen abhängig von der Regularität der Anfangsdaten, mit und ohne Annahmen über den Rang der assoziierten Dichtematrix. Weiters präsentieren wir numerische Simulationen eines vereinfachten Models, mit besonderem Interesse an der so genannten *Korrelation*, welche eine der wesentlichen Motivationen und Vorteile der Multikonfigurationsmethoden ist im Vergleich zu Hartree-Fock Modellen.

Résumé

Les méthodes de multi-configuration sont une amélioration naturelle des modèles simples d' approximation bien connus de l'équation de Schrödinger linéaire à N corps pour les systèmes moléculaires sous interactions binaires -Coulombiennes dans les situations réelles-, tels que les modèles de Hartree et de Hartree-Fock. les modèles telles que MCTDHF sont intensivement utilisés pour des simulations numériques en chimie/physique quantique. Cependant, les équations associées à ces modèles sont encore mal compris d'un point de vue mathématique. La présente contribution apporte la première fondation mathématique rigoureuse aux équations associées à la MCTDH(F) avec interaction singulière de Coulomb . En particulier, on formule le problème d'évolution d'une façon qui convient à l'analyse mathématique et on obtient des résultats d'existence et d'uncité dépendants de la régularité de la donnée initiale avec et sans hypothèse sur le rang de la matrice densité associée. La simulation numérique d'un modèle simplifié est aussi présentée avec un intérêt particulier à ce qu'on appelle *correlation* qui représente à elle seule une des principales motivations et avantages des méthodes de type multiconfiguration comparées aux méthodes de Hartree-Fock.

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Chapter 1

Introduction

Trouver quelque chose en mathématiques,
c'est vaincre une inhibition et une tradition.
Laurent Schwartz

"Analysis of the MultiConfiguration Time-Dependent Hartree-Fock equations"

This thesis consists of 4 publications, appeared or submitted, plus 2 additional sections, plus an introduction, grouped into 6 chapters :

1. "Introduction"

2. "Local-in-time existence of MCTDHF"

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5. "Setting and Analysis of the MCTDHF model"

   C. Bardos, I. Catto, N.J. Mauser and S. Trabelsi,
   Setting and analysis of the multiconfiguration time-dependent Hartree-Fock equations
   Preprint

6. "Numerical illustration of the MCTDHF hierarchy"

The new contribution is to give and present in a unified manner the first mathematical results ever in the existence and uniqueness analysis of the MCTDHF equations with Coulomb interaction. To this end, a new formulation of these quite complicated equations (in comparison to wave equation, NLS,...) is given. We use and adapt a large variety of mathematical tools of PDE analysis, like variational methods, ..., Strichartz estimates as applied by F. Castella for the L^2 analysis of the simple "Schrödinger-Poisson" equation.

Notably the publication of chapter 5 of this thesis is a comprehensive and exhaustive monograph on the "state of the art" of the analysis of MCTDHF equations.

1.1 The linear many particle time-dependent Schrödinger equation

The time-dependent Schrödinger equation (1.1) is a fundamental concept of quantum mechanics to obtain the time evolution of a system that is described by a complex valued function in L^2 called the *"wave function"*:

$$i\frac{\partial}{\partial t}\Psi(t) = \mathscr{H}(t)\,\Psi(t), \tag{1.1}$$

where $\mathscr{H}(t)$ is the "Hamilton operator" associated to the energy of the system. In this thesis we will deal with approximations of the linear many particle time-dependent Schrödinger equation by non-linear "one particle" equations. The system of equations that form the Multi-Configuration Time-Dependent Hartree-Fock method allows for very good approximations of fermions (electrons).

Consider N electrons of mass m_e and charge z_e and M nuclei with respective mass $m_1,\dots,m_M$ and charges $Z_1,\dots,Z_M$. These particles being “somewhere" in the physical space, thus, one associates a position to each one, say $x_k \in \mathbb{R}^3$, $k = 1,\dots,N$ for electrons and $R_k \in \mathbb{R}^3$, $k = 1,\dots,M$ for nuclei. Moreover, for quantum particles, there exist additional independent variables called *spin*, a discrete variable that characterizes the behavior of a particle under the effect of the symmetry of space rotation, denoted by σ_k, $k = 1,\dots,N$ and $\overline{\sigma}_k$, $k = 1,\dots,M$ be, respectively, for the spin of the electrons and nuclei. In most of the mathematical analysis, the spin does not change the analysis and is often omitted for notational simplicity.

The wavefunction depends on time, space and spin variables:

$$\Psi(t,R_1,\overline{\sigma}_1,\dots,R_M,\overline{\sigma}_M,x_1,\sigma_1,\dots,x_N,\sigma_N). \tag{1.2}$$

The density of probability to find the system in a given "state" $(R_1, \overline{\sigma}_1, \ldots, , x_1, \sigma_1, \ldots,)$ at time t is then extracted from the wavefunction Ψ by the square of the modulus of the complex valued wavefunction :

$$|\Psi(t, R_1, \overline{\sigma}_1, \ldots, R_M, \overline{\sigma}_M, x_1, \sigma_1, \ldots, x_M, \sigma_M)|^2. \tag{1.3}$$

It is the obviously nonnegative probability density that the nucleus k be in the position R_k with spin variable $\overline{\sigma}_k$ and the electron p be in the position x_p with spin σ_p at time t for $k = 1, \ldots, M$ and $p = 1, \ldots, N$. In particular, this interpretation requires that the wavefunction Ψ be normalized to one

$$\begin{aligned} \|\Psi\|^2_{L^2(\mathbb{R}^{3N})} = & \sum_{\sigma_1, \ldots, \sigma_N} \sum_{\overline{\sigma}_1, \ldots, \overline{\sigma}_M} \int_{\mathbb{R}^{3N}} dx_1, \ldots, dx_N \int_{\mathbb{R}^{3M}} dR_1, \ldots, dR_M \times \\ \times \quad & |\Psi(t, R_1, \overline{\sigma}_1, \ldots, R_M, \overline{\sigma}_M, x_1, \sigma_1, \ldots, x_N, \sigma_N)|^2 = 1 \end{aligned}$$

An additional important property of a many body wavefunction is its symmetry behaviour that groups quantum particles in two different classes :

- *fermions* : skew-symmetric with respect to permutations of space and spin coordinates. (e.g. electrons or nuclei composed of odd number of nucleons are fermions.) That is

$$\begin{aligned} & \Psi(t, R_1, \overline{\sigma}_1, \ldots, R_M, \overline{\sigma}_M, x_{\varepsilon(1)}, \sigma_{\varepsilon(1)}, \ldots, x_{\varepsilon(N)}, \sigma_{\varepsilon(N)}) \\ & \quad = \quad (-1)^{\varepsilon} \Psi(t, R_1, \overline{\sigma}_1, \ldots, R_M, \overline{\sigma}_M, x_1, \sigma_1, \ldots, x_N, \sigma_N). \end{aligned} \tag{1.4}$$

  The mapping ε is the permutation of $\{1, \ldots, N\}$ and $(-1)^{\varepsilon}$ denotes its parity. In particular, the skew-symmetry property of Ψ expresses the *Pauli exclusion principle* that two electrons with the same spin cannot be in the same space position at the same instant t. We have obviously from (1.4)

$$\Psi(t, R_1, \overline{\sigma}_1, \ldots, R_M, \overline{\sigma}_M, x_1, \sigma_1, \ldots, x_N, \sigma_N) = 0,$$

  if there exists $1 \leq i \neq j \leq N$ such that $x_i = x_j$ and $\sigma_i = \sigma_j$.

- *bosons* : symmetric with respect to permutations of space and spin coordinates. (e.g. photons, nuclei composed of even number of nucleons).
  Note that a N particle wavefunction that is a direct product of N times the same "one particle" wavefunction evaluated at N different positions is trivially symmetric and corresponds to bosons in a very peculiar state, a so called Bose-Einstein condensate.

The equation (1.1) contains the system's self-adjoint Hamilton operator (= "Hamiltonian") $\mathscr{H}(t)$ acting on Ψ in $L^2(\mathbb{R}^{3M}) \otimes L^2(\mathbb{R}^{3N})$. In this work we will be interested in time-independent Hamiltonians $\mathscr{H}$ which correspond to *isolated systems* without exterior interactions. Time-dependent Hamiltonians are of interest e.g. when a system of electrons is exposed to externals electric fields.

The time-independent Hamiltonian $\mathscr{H}$ we consider is

$$\begin{aligned}\mathscr{H} &= -\frac{\hbar^2}{2}\sum_{i=1}^{N}\frac{1}{m_e}\Delta_{x_i} - \sum_{i=1}^{M}\sum_{j=1}^{N}\frac{Z_i Z_e}{4\pi\varepsilon_0}\frac{1}{|R_i - x_j|} + \sum_{1\le i<j\le N}\frac{Z_e^2}{4\pi\varepsilon_0}\frac{1}{|x_i - x_j|} \\ &- \frac{\hbar^2}{2}\sum_{i=1}^{M}\frac{1}{m_i}\Delta_{R_i} + \sum_{1\le i<j\le M}\frac{Z_i Z_j}{4\pi\varepsilon_0}\frac{1}{|R_i - R_j|}. \end{aligned} \tag{1.5}$$

In this formula, $\hbar$ denotes the Planck constant and ε_0 the dielectric constant of the vacuum. The Δ is the Laplace operator. The notation Δ_{x_i} means that it acts on the space variable x_i. The expression of $\mathscr{H}$ is simpler when using the atomic units which are the most widespread in quantum mechanics. This means a scaling where the mass of the electron, the elementary charge and the factor $\frac{1}{4\pi\varepsilon_0}$ are equal to one. The expression of $\mathscr{H}$ is then

$$\begin{aligned}\mathscr{H} &= -\frac{1}{2}\sum_{i=1}^{N}\Delta_{x_i} - \sum_{i=1}^{M}\sum_{j=1}^{N}\frac{Z_i}{|R_i - x_j|} + \sum_{1\le i<j\le N}\frac{1}{|x_i - x_j|} \\ &- \sum_{i=1}^{M}\frac{1}{2\,m_i}\Delta_{R_i} + \sum_{1\le i<j\le M}\frac{Z_i Z_j}{|R_i - R_j|}. \end{aligned}$$

This Hamilton operator corresponds to the classical mechanics Hamiltonian via the correspondence rules $x \to x$ and $p_x \to -i\nabla_x$. Hence the expression of the Hamiltonian (1.5) is composed of terms corresponding to the "kinetic energy" p_x^2 of electrons and nuclei which are the ones involving the Laplacian and the other terms correspond to the "potential energy" of the electrostatic interaction nuclei-electrons, electrons-electrons and nuclei-nuclei.

The Cauchy problem associated to the linear Schrödinger equation reads

$$\mathscr{S}: \quad \begin{cases} i\frac{\partial}{\partial t}\Psi(t) = \mathscr{H}\Psi(t), \\ \\ \Psi(t=0) = \Psi^0 \in \mathscr{X}. \end{cases}$$

where $\mathscr{X}$ denotes the space of physical states. The operator $\mathscr{H}$ being time-independent, a remarkable solution to the system $\mathscr{S}$ is given explicitly by

$$\Psi(t) = e^{-iEt}\Psi(0), \quad \mathscr{H}\Psi^0 = E\Psi^0, \quad \|\Psi\|_{\mathscr{X}} = 1.$$

That is Ψ^0 is the eigenvector of $\mathscr{H}$ corresponding to the eigenvalue E. Moreover, if E is the smallest eigenvalue of $\mathscr{H}$, then Ψ^0 is the so called *the ground state* of the system.

From the point of view of mathematical analysis of existence and uniqueness of solutions, the Cauchy problem $\mathscr{S}$ is well-posed by the Stone's Theorem. That is the evolution of the system is governed by a propagator $(\mathscr{U}(t,s))_{(t,s)\in\mathbb{R}^2}$ on $\mathscr{X}$ satisfying

$$\Psi(t) = \mathscr{U}(t,s)\Psi(s), \quad \text{for all} \quad (t,s) \in \mathbb{R}^2.$$

Moreover,

- $\mathscr{U}(t,r)\mathscr{U}(r,s) = \mathscr{U}(t,s)$ for all $(t,r,s) \in \mathbb{R}^3$.

- $\mathscr{U}(t,s)$ is unitary on $\mathscr{X}$ for all $(t,s) \in \mathbb{R}^2$ and $(t,s) \mapsto \mathscr{U}(t,s)$ is strongly continuous from $\mathbb{R}^2$ to $\mathscr{L}(\mathscr{X})$.

- $\mathscr{U}(t,s) \in \mathscr{L}(D)$ for all$(t,s) \in \mathbb{R}^2$ and $(t,s) \mapsto \mathscr{U}(t,s)$ is strongly continuous from $\mathbb{R}^2$ to $\mathscr{L}(D)$ where D denotes the domain of the operator $\mathscr{H}$.

- The following equalities hold in a strong sense between operators from D to $\mathscr{X}$:

$$i\frac{\partial}{\partial t}\mathscr{U}(t,s) = \mathscr{H}\,\mathscr{U}(t,s) \quad ; \quad i\frac{\partial}{\partial s}\mathscr{U}(t,s) = -\mathscr{U}(t,s)\,\mathscr{H}.$$

Also, notice that the solution of the Cauchy problem $\mathscr{S}$ preserves the normalization constraint on Ψ and this a direct consequence of the unitarity of the propagator $\mathscr{U}$. Moreover, since the Hamiltonian $\mathscr{H}$ is symmetric with respect to the space coordinates of two identical particles, the solution keeps then the symmetry or skew-symmetry property of Ψ^0.

From numerical point of view, the Schrödinger equation is too much of a challenge even for small systems and even if the most powerfull parallel machines are used - already storing the state is "mission impossible". For instance, consider the molecule of water H_20, it is then composed of 3 nuclei and 10 electrons. Thus, one has to work in the huge space (neglecting the spin)

$$L^2(\mathbb{R}^9,\mathbb{C}) \otimes \left(\bigwedge_{i=1}^{10} L^2(\mathbb{R}^3,\mathbb{C}) \right),$$

which is obviously out of the range of todays and mid-futures computers.

The challenge is then to reduce the size of the space and reduce the cost of such reduction on the precision of the result. However, this is extremely demanding: for instance, assume that one has to compute the *formation's enthalpy* of a CO_2 molecule which is known to be of order 0.08% of the total relativistic energy of the system. Thus if one commits an error of order 1%, the result will be that the carbon dioxide is an excellent fuel or a powerful explosive ! (example from [8])

A first step of simplification is the Born-Oppenheimer approximation, based on the observation that the nuclei are much more heavier than electrons and their dynamics can be decoupled and neglected on the electron's time scale. Essentially, this allows to solve the Schrödinger equation for a prescribed configuration of nuclei. Typically, the variables $R_1,\ldots,R_M$ become only parameters and the Hamiltonian is then

$$\begin{aligned} \mathscr{H} &= -\frac{1}{2}\sum_{i=1}^{N}\Delta_{x_i} - \sum_{j=1}^{N}\sum_{i=1}^{M}\frac{Z_i}{|R_i - x_j|} + \sum_{1\le i<j\le N}\frac{1}{|x_i - x_j|}, \\ &:= -\frac{1}{2}\sum_{i=1}^{M}\Delta_{x_i} - \sum_{j=1}^{N}U(x_j) + \sum_{1\le i<j\le N}V(x_i,x_j). \end{aligned}$$

We refer the reader to the excellent introduction of Cancè's Thesis [8] and [10]. Obviously, the problem is then reduced and the space to approximate is now smaller that the initial one. More precisely (again neglecting the spin)

$$\bigwedge_{i=1}^{10} L^2(\mathbb{R}^3, \mathbb{C}),$$

which is still very large for numerical simulations and (iterative) direct solvers are seldom attempted. More drastic approximations allow for the reduction of the many particle wavefunction to systems of "one particle wavefunctions", for the prize of transforming a linear equation into non-linear equations :

1.2 Approximations of the Schrödinger Equations

In this section we shall present two well-known approximations, namely the time-dependent Hartree equation (TDH) for bose condensates and the time-dependent Hartee-Fock (TDHF) for fermions.

In such approximations, the N particles Schrödinger equation is replaced by one or more one particle partial differential equations. Generically, these equations are non-linear and coupled in the case of systems. These equation have, in general, Hamiltonian structure. More precisely, they have a variational structure in the sense that they satisfy variational principles (VP).

The trick is to make a particular "ansatz" for the many particle wave-function in terms of "one particle" wavefunctions (sometimes called "orbitals", "enveloppe functions" etc.).

The simplest possible such ansatz is a product of N times the same wavefunction evaluated at the N different positions : the Hartree-ansatz for boson condensates, explained below. The simplest realistic ansatz for fermions is to take a single Slater determinant of N orthonormed "orbitals" : this leads to the Hartree-Fock equations.

The *multiconfiguration time-dependent Hartree-Fock* (MCTDHF) system, however, represents a sophisticated hierarchy of approximations. Such "multiconfiguration" methods are widely used in quantum chemistry, as a recipy to perform numerical calculations, but mathematically they are very poorly understood. In fact, this thesis is the first exhaustive work on the analysis of the time-dependent case. The MCTDHF system is composed of $K \geq N$ non-linear Schrödinger-type evolution equations (for "the orbitals", as a dynamic basis for an expansion by "Slater determinants") coupled with $\binom{K}{N}$ ordinary differential equations (for " the coefficients"). The many particle wavefunction $\Psi^N(t, x_1, ..., x_N)$ can be well approximated by such linear combinations of Slater determinants that catch also "correlations", in contrast to the simple time-dependent Hartree-Fock (TDHF) method that corresponds to the special case $K = N$.

The concept of "correlation" of many particle systems is as fundamental as mysterious : indeed, there is no common consens how to exactly define it - only that "correlation is zero for Hartree-Fock wavefunctions". A good mathematically oriented discussion and relevant definition of correlation can be found in [19, 20]. We have used this definition to calculate correlation in our numerical illustration of the MCDTHF method, with systematic study of correlation with varying K for fixed N.

In principle, the many particle wavefunction constructed from the solution of MCTDHF converges towards the exact solution Ψ^N with increasing K; however, especially in the time-dependent case there is no proof for this seemingly "obvious" property of MCTDHF. A short and readable introduction to the multiconfiguration time-dependent Hartree-Fock (MCTDHF) system is given in the next chapters of this thesis (or, more exhaustive, [24, 31]).

1.2.1 Variational Principle

From now on $\mathscr{X}$ will denote $L^2(\mathbb{R}^{3N},\mathbb{C})$. Let $\mathscr{M} \subset \mathscr{X}$ be a manifold on which the approximate wavefunction Ψ_{app} of Ψ lies. Also, let $\mathbf{T}_{\Psi_{\text{app}}}\mathscr{M}$ be the tangent space of $\mathscr{M}$ at the point Ψ_{app}. It is the subspace of $\mathscr{X}$ on which lie the derivative of all paths passing through Ψ_{app} and belonging to $\mathscr{M}$. Physicists call it *the space of admissible variations*. Elements of $\mathscr{T}_{\Psi_{\text{app}}}\mathscr{M}$ will be denoted $\delta\Psi_{\text{app}}$. The Dirac-Frenkel VP reads then

$$\left\langle \Psi_{\text{app}} \,\middle|\, i\frac{\partial}{\partial t} - \mathscr{H} \,\middle|\, \delta\Psi_{\text{app}} \right\rangle_{\mathscr{H}} = 0, \quad \text{for all} \quad \delta\Psi_{\text{app}} \in \mathbf{T}_{\Psi_{\text{app}}}\mathscr{M}. \tag{1.6}$$

This principle, then, characterizes the approximate wavefunction $t \mapsto \Psi_{\text{app}}(t) \in \mathscr{M}$ by a projection procedure of $\mathscr{H}\Psi_{\text{app}}$ onto the tangent space $\mathbf{T}_{\Psi_{\text{app}}}\mathscr{M}$. We refer to [16, 17, 25] for details and a short and much more readable explication in [22, 23].

Observe that for a given initial data $\Psi_{\text{app}}(t=0) = \Psi^0_{\text{app}}$, then, approximations satisfying this VP for all $t > 0$ obeys formally,

- $$\|\Psi_{\text{app}}(t)\|_{\mathscr{X}} = \|\Psi_{\text{app}}(t=0)\|_{\mathscr{X}}, \tag{1.7}$$
- $$\left\langle \Psi_{\text{app}}(t) \,\middle|\, \mathscr{H} \,\middle|\, \Psi_{\text{app}}(t) \right\rangle_{\mathscr{X}} = \left\langle \Psi_{\text{app}}(t=0) \,\middle|\, \mathscr{H} \,\middle|\, \Psi_{\text{app}}(t=0) \right\rangle_{\mathscr{X}}. \tag{1.8}$$

This observation is obvious, in fact $\Psi_{\text{app}} \in \mathbf{T}_{\Psi_{\text{app}}}\mathscr{M}$, then

$$\begin{aligned}
\frac{d}{dt}\left\langle \Psi_{\text{app}}(t) \,\middle|\, \Psi_{\text{app}}(t) \right\rangle_{\mathscr{X}} &= 2\,\Re\left\langle \frac{\partial}{\partial t}\Psi_{\text{app}}(t) \,\middle|\, \Psi_{\text{app}}(t) \right\rangle_{\mathscr{X}}, \\
&= 2\,\Im\left\langle \Psi_{\text{app}}(t) \,\middle|\, \mathscr{H} \,\middle|\, \Psi_{\text{app}}(t) \right\rangle_{\mathscr{X}}, \\
&= 0.
\end{aligned}$$

The symbols $\Re$ and $\Im$ denote respectively the real and imaginary parts of a complex number. This proves the conservation law (1.7). The proof of the second assertion (2.6) is in the same spirit. In fact, $\frac{\partial}{\partial t}\Psi_{\text{app}}(t) \in \mathbf{T}_{\Psi_{\text{app}}}\mathscr{M}$ and the Hamiltonian $\mathscr{H}$ being time-independent, then

$$\begin{aligned}
\frac{d}{dt}\left\langle \Psi_{\text{app}} \,\middle|\, \mathscr{H} \,\middle|\, \Psi_{\text{app}} \right\rangle_{\mathscr{X}} &= 2\,\Re\left\langle \frac{\partial}{\partial t}\Psi_{\text{app}} \,\middle|\, \mathscr{H} \,\middle|\, \Psi_{\text{app}} \right\rangle_{\mathscr{X}}, \\
&= 2\,\Im\,\| \frac{\partial}{\partial t}\Psi_{\text{app}} \|^2_{\mathscr{X}}, \\
&= 0.
\end{aligned}$$

From physical point of view, the property (1.7) and (2.6) shows that the system conserves respectively its mass and total energy.

1.2.2 Density Operators

This paragraph will be devoted to the introduction of the so called *density operators*. For that purpose we introduce the notation

$$X_n = (x_1, \dots, x_n), \qquad X_n^N = (x_{n+1}, \dots, x_N),$$

$$dX_n = dx_1 \dots dx_n, \qquad dX_n^N = dx_{n+1} \dots dx_N,$$

and similarly for other capital letters. Now, for $n = 1, \dots, N$, we define the n^{th} density operator *via* its kernel as follows

$$\left[\Psi \otimes \Psi\right]_{:n}(t, X_n, Y_n) \quad = \quad \binom{N}{n} \int_{\mathbb{R}^{3(N-n)}} \Psi(t, X_n, Z_n^N)\, \overline{\Psi}(t, Y_n, Z_n^N)\, dZ_n^N. \tag{1.9}$$

The $\binom{N}{n}$ are normalization factors which are sometimes omitted in the literature and the bar denotes the conjugate of complex number. In the definition (5.9), we used the convention that for $n = N$, we have

$$\left[\Psi \otimes \Psi\right]_{:N}(t, X_N, Y_N) = \Psi(t, X_N)\, \overline{\Psi}(t, Y_N).$$

Hence, $\left[\Psi \otimes \Psi\right]_{:n}(t, X_N, X_N)$ is nothing but the density of probability to find the N particles in the configuration X_N at the instant t. These operators are well-known to be Trace-Class Operators with trace being given by

$$\mathrm{Tr}\left[\Psi \otimes \Psi\right]_{:n} \quad = \quad \binom{N}{n} \int_{\mathbb{R}^{3N}} |\Psi(X_n, X_n^N)|^2 dX_n dX_n^N = \binom{N}{n}.$$

Moreover, the Schrödinger equation (1.1) is the equivalent to the following operator equation

$$i\frac{\partial}{\partial t}\left[\Psi \otimes \Psi\right]_{:1}(t) = \left[H\,,\, \left[\Psi \otimes \Psi\right]_{:1}(t)\right]. \tag{1.10}$$

The $[\cdot, \cdot]$ denotes the commutator of operators, that is $[A, B] = AB - BA$. Also, observe the trivial relation

$$\left[\Psi \otimes \Psi\right]_{:n}(X_n, Y_n) = \frac{n+1}{N-n} \int_{\mathbb{R}^3} \left[\Psi \otimes \Psi\right]_{:n+1}(X_n, z, Y_n, z)\, dz.$$

1.2.3 TDHF approximation

The Hartree-Fock approximation is a very simple adequate method for systems of fermions where a skew-symmetry is imposed on the wavefunction Ψ_{HF}.

Now, given two functions $f, g \in L^2(\mathbb{R}^3)$, then the simplest skew-symmetric function that one can build from f and g in $L^2(\mathbb{R}^6, \mathbb{C})$ is

$$f \otimes g - g \otimes f.$$

That is a determinant. In the same way, given $\phi_1, \ldots, \phi_N \in L^2(\mathbb{R}^3)$, one can construct the determinant from the ϕ_i's as follows

$$\mathrm{Det}_{\phi_1,\ldots,\phi_N}(x_1,\ldots,x_N) = \begin{vmatrix} \phi_1(x_1) & \ldots & \phi_1(x_N) \\ \vdots & & \vdots \\ \phi_N(x_1) & \ldots & \phi_N(x_N) \end{vmatrix}.$$

Moreover if the ϕ_i's form an orthonormal family, then a factor of $\frac{1}{\sqrt{N!}}$ is introduced in order to normalize the determinant to one in $L^2(\mathbb{R}^{3N}, \mathbb{C})$.

After, this simple observation, the TDHF approximation consists in forcing the wavefunction to evolve on the manifold

$$\mathscr{M}_{HF} = \left\{ \Psi_{\mathrm{HF}} = \frac{1}{\sqrt{N!}} \mathrm{Det}_{\phi_1,\ldots,\phi_N}, : \phi_i \in L^2(\mathbb{R}^3), \int_{\mathbb{R}^3} \phi_i \, \overline{\phi}_j \, dx = \delta_{i,j}, \, 1 \le i, j \le N \right\}.$$

The associated system is known to be the following

$$\begin{aligned} i\frac{\partial}{\partial t}\phi_i(t,x) &= \mathbf{H}_x \, \phi(t,x) + \sum_{j=1}^{N} \int_{\mathbb{R}^3} V(x,y) \, |\phi_j|^2(t,y) \, dy \, \phi_i(t,x) \\ &- \sum_{j=1}^{N} \int_{\mathbb{R}^3} \phi_j(t,y) \, V(x,y) \, \overline{\phi}_i(t,y) \, dy \, \phi_j(t,x), \\ &:= \mathbf{H}_x \, \phi_i(t,x) + \mathbf{F}_\phi \, \phi_i(t,x). \end{aligned} \tag{1.11}$$

The operator $\mathbf{H}$ is the self-adjoint time-independent operator acting on $L^2(\mathbb{R}^3)$ and defined as follows

$$\mathbf{H}_x := -\frac{1}{2}\Delta_x + U(x).$$

Also, it is easy to see that the operator $\mathbf{F}_\phi$ is self-adjoint. Now, given an initial data $\phi_i(t=0) = \phi_i^0$, then the TDHF system enjoys

$$\int_{\mathbb{R}^3} \phi_i(t,x) \, \overline{\phi}_j(t,x) \, dx = \int_{\mathbb{R}^3} \phi_i^0(x) \, \overline{\phi}_j^0(t,x) \, dx. \tag{1.12}$$

In fact, multiply (5.72) by $\overline{\phi}_j$ and integrate over $\mathbb{R}^3$. We get

$$i \int_{\mathbb{R}^3} \frac{\partial}{\partial t} \phi_i(t,x) \, \overline{\phi}_j(t,x) \, dx = \int_{\mathbb{R}^3} \left(\mathbf{H} + \mathbf{F}_\phi \right) \phi_i(t,x) \, \overline{\phi}_j(t,x) \, dx$$

Next, conjugate the expression above, swap i and j, use the fact that $\mathbf{H} + \mathbf{F}_\phi$ is a self-adjoint

operator and one get easily the property (1.12). Now, introduce the following functional

$$\begin{aligned}\mathscr{E}_\phi(t) &= \frac{1}{2}\sum_{i=1}^{N}\int_{\mathbb{R}^3}\left(|\nabla\phi_i(t,x)|^2+U(x)\,|\phi_i(t,x)|^2\right)dx\\ &+ \frac{1}{4}\sum_{i,j=1}^{N}\int_{\mathbb{R}^6}\phi_i(t,x)\overline{\phi}_i(t,x)\,V(x,y)\,\phi_j(t,y)\overline{\phi}_j(t,y)\,dx\,dy\\ &- \frac{1}{4}\sum_{i,j=1}^{N}\int_{\mathbb{R}^6}\phi_i(t,x)\overline{\phi}_j(t,x)\,V(x,y)\,\phi_j(t,y)\overline{\phi}_i(t,y)\,dx\,dy.\end{aligned}$$

It is known that the TDHF system preserves the total energy $\mathscr{E}_\phi(t)$. That is

$$\mathscr{E}_\phi(t)=\mathscr{E}_{\phi^0}(t=0),\quad \text{for all}\quad t>0.$$

We refer to the proof of Chadam and Glassey in [15]. However we warn the reader that the proof is laborious calculation and is far from being the simplest way to prove it. In fact, we shall prove formally in [12] that the TDHF system satisfies the Dirac-Frenkel VP (2.5), thus it conserves automatically the total energy $\mathscr{E}_\phi$.

It is of interest to compute the first and the second order kernels. From the one side

$$\begin{aligned}\left[\Psi_{HF}\otimes\Psi_{HF}\right]_{:1}(t,x,y) &= N\sum_{\varepsilon,\kappa\in S_N}(-1)^{\varepsilon+\kappa}\,\phi_{\varepsilon(1)}(x)\,\overline{\phi}_{\kappa(1)}(y)\prod_{k=2}^{N}\int_{\mathbb{R}^3}\phi_{\varepsilon(k)}(x_k)\,\overline{\phi}_{\kappa(k)}(x_k)\,dx_k,\\ &= N\sum_{\substack{\varepsilon,\kappa\in S_N\ s.t.\\ \varepsilon(l)=\kappa(l),\,l=2,\ldots,N}}(-1)^{\varepsilon+\kappa}\,\phi_{\varepsilon(1)}(x)\,\overline{\phi}_{\kappa(1)}(y)\end{aligned}$$

The condition on the permutations ε and κ implies obviously that $\varepsilon\equiv\kappa$. Thus

$$\left[\Psi_{HF}\otimes\Psi_{HF}\right]_{:1}(t,x,y)=\sum_{p=1}^{K}\phi_p(t,x)\,\overline{\phi}_p(t,y). \tag{1.13}$$

From the opposite side

$$\left[\Psi_{HF}\otimes\Psi_{HF}\right]_{:1}(t,x,y,x',y')=\frac{N(N-1)}{2}\sum_{\substack{\varepsilon,\kappa\in S_N\ s.t.\\ \varepsilon(l)=\kappa(l),\,l=3,\ldots,N}}(-1)^{\varepsilon+\kappa}\,\phi_{\varepsilon(1)}(x)\,\phi_{\varepsilon(2)}(y)\,\overline{\phi}_{\kappa(1)}(x')\,\overline{\phi}_{\kappa(1)}(y').$$

In this configuration, the conditions on the permutations implies

$$\varepsilon(1)=\kappa(1),\,\varepsilon(2)=\kappa(2)\quad\text{or}\quad\varepsilon(1)=\kappa(2),\,\varepsilon(2)=\kappa(1).$$

Following this remark, it is obvious to obtain

$$\begin{aligned}\left[\Psi_{HF}\otimes\Psi_{HF}\right]_{:2}(t,x,y,x',y') &= \frac{1}{2}\sum_{p,q=1}^{N}\phi_p(t,x)\,\phi_q(t,y)\,\overline{\phi}_p(t,x')\,\overline{\phi}_q(t,y')\\ &- \frac{1}{2}\sum_{p,q=1}^{N}\phi_p(t,x)\,\phi_q(t,y)\,\overline{\phi}_q(t,x')\,\overline{\phi}_p(t,y').\end{aligned}\tag{1.14}$$

A fascinating thing about the Hartree-Fock approximation, more generally, about determinants is that

$$\begin{aligned}\left[\Psi_{HF}\otimes\Psi_{HF}\right]_{:2}(t,x,y,x',y') &= \frac{1}{2}\left[\Psi_{HF}\otimes\Psi_{HF}\right]_{:1}(t,x,x')\left[\Psi_{HF}\otimes\Psi_{HF}\right]_{:1}(t,y,y') \\ &- \frac{1}{2}\left[\Psi_{HF}\otimes\Psi_{HF}\right]_{:1}(t,x,y')\left[\Psi_{HF}\otimes\Psi_{HF}\right]_{:1}(t,y,x').\end{aligned}$$

That is

$$\left[\Psi_{HF}\otimes\Psi_{HF}\right]_{:2}(t,x,y,x',y') = \frac{1}{2}\begin{vmatrix} \left[\Psi_{HF}\otimes\Psi_{HF}\right]_{:1}(t,x,x') & \left[\Psi_{HF}\otimes\Psi_{HF}\right]_{:1}(t,y,x') \\ \left[\Psi_{HF}\otimes\Psi_{HF}\right]_{:1}(t,x,y') & \left[\Psi_{HF}\otimes\Psi_{HF}\right]_{:1}(t,y,y') \end{vmatrix}.$$

This remark allows in particular to close the Von-Neuman type equation (1.10) for the TDHF methods:

$$i\frac{\partial}{\partial t}\left[\Psi_{\mathrm{HF}}\otimes\Psi_{\mathrm{HF}}\right]_{:1}(t) = \left[\hat{H}\,,\,\left[\Psi_{\mathrm{HF}}\otimes\Psi_{\mathrm{HF}}\right]_{:1}(t)\right].$$

Where the operator $\hat{H}$ is as follows

$$\hat{H} = \mathbf{H} + \hat{V}_{\mathrm{H}} + \hat{V}_{\mathrm{HF}},$$

with

$$\hat{V}_{\mathrm{H}} = \hat{V}_{\mathrm{H}}\left(\left[\Psi_{\mathrm{HF}}\otimes\Psi_{\mathrm{HF}}\right]_{:1}\right) = \frac{1}{4\pi}\int_{\mathbb{R}^3} V(x,z)\left[\Psi_{\mathrm{HF}}\otimes\Psi_{\mathrm{HF}}\right]_{:1}(t,z,z)\,dz.$$

Finally,

$$\hat{V}_{\mathrm{HF}} = \hat{V}_{\mathrm{HF}}\left(\left[\Psi_{\mathrm{HF}}\otimes\Psi_{\mathrm{HF}}\right]_{:1}\right) = \frac{1}{4\pi}V(x,z)\left[\Psi_{\mathrm{HF}}\otimes\Psi_{\mathrm{HF}}\right]_{:1}(t,x,z).$$

This formulation is of particular interest when dealing with infinite system of particles (see [3, 1]). It is also important to notice that starting with initial data $\Psi_{\mathrm{HF}}(t=0)$ factorized as a *Slater determinant*, then the TDHF dynamics broke this *ansatz* in the sense that the solution at some time t is not a *Slater determinant*. This is because of the exchange term

$$\sum_{j=1}^{N}\int_{\mathbb{R}^3}\phi_j(t,y)\,V(x,y)\,\overline{\phi}_i(t,y)\,dy\,\phi_j(t,x).$$

Finally, recall that there exists a variants of the TDHF model, namely the restricted and unrestricted Hartree-Fock methods and refer the reader to [10], for instance, for more details.

1.2.4 TDH approximation for boson condensates

Quantum particles with integer spin, the *bosons*, are undistinguishable and have to obey fundamentally different properties than particles with spin that is a "half of an integer", the *fermions*. Many boson wavefunctions have to be "symmetrized", i.e. to be invariant under permutations of particles.

The simplest possible such ansatz is a product of N times the same wavefunction evaluated at the N different positions : the Hartree-ansatz for boson condensates ("Bose-Einstein Condensates

BEC, an old theoretical idea of Bose and Einstein of a very peculiar state of matter that is now realized in routine experiments). We warn the reader that the term "Hartree ansatz", which should be reserved to the more general ansatz: $\phi_1 \otimes, \dots, \phi_N$ with $\phi_1, \dots, \phi_N$ being an orthonormed basis of $L^2(\mathbb{R}^3)$, is often used for the "Hartree ansatz for BEC" as we state now. Note that the "Schrödinger-Poisson" system, frequently used for the modeling of electrons, is actually obtained from this Hartree ansatz for bosons.

The Hartree approximation consists then in forcing the approximate wavefunction to evolves on the manifold $\mathscr{M}_H$ where

$$\mathscr{M}_{\mathrm{H}} = \left\{ \Psi_{\mathrm{H}}(t,x_1,\dots,x_N) = \phi(t,x_1)\dots\phi(t,x_N), \quad \phi \in L^2(\mathbb{R}^3), \quad \| \phi \|_{L^2(\mathbb{R}^3)} = 1 \right\}.$$

The normalization constraint on ϕ implies obviously $\| \Psi_{\mathrm{H}} \|_{\mathscr{H}} = 1$. Moreover, we have

$$\left[\Psi_H \otimes \Psi_H\right]_{:n}(t,X_n,Y_n) = \binom{N}{n} \prod_{k=1}^{n} \phi(t,x_k)\,\overline{\phi}(t,y_k).$$

In particular,

$$\left[\Psi_H \otimes \Psi_H\right]_{:1}(t,x,y) = N\,\phi(t,x)\,\overline{\phi}(t,y),$$

$$\begin{aligned}\left[\Psi_H \otimes \Psi_H\right]_{:2}(t,x_1,x_2,y_1,y_2) &= \frac{N(N-1)}{2}\,\phi(t,x_1)\,\phi(t,x_2)\,\overline{\phi}(t,y_1)\,\overline{\phi}(t,y_2),\\ &= \frac{N-1}{2N}\left[\Psi_H \otimes \Psi_H\right]_{:1}(t,x_1,y_1)\left[\Psi_H \otimes \Psi_H\right]_{:1}(t,x_2,y_2).\end{aligned}$$

The TDH system of equations reads

$$i\frac{\partial}{\partial t}\phi(t,x) = \mathbf{H}_x\,\phi(t,x) + \int_{\mathbb{R}^3} |\phi|^2(t,y)\,V(x,y)\,dy\,\phi(t,x). \tag{1.15}$$

Recall that, with a different scaling of the interaction, the authors of [2] derived **rigorously** the TDH as a weak coupling limit of the N particles Schrödinger equation as $N \to +\infty$. As for the TDHF case, a Von-Neuman type evolution equation on the first order density matrix is possible. The equation reads

$$i\frac{\partial}{\partial t}\left[\Psi_{\mathrm{H}} \otimes \Psi_{\mathrm{H}}\right]_{:1}(t) = \left[\hat{H}\,,\,\left[\Psi_{\mathrm{HF}} \otimes \Psi_{\mathrm{H}}\right]_{:1}(t)\right].$$

Where the operator $\hat{H}$ is as follows

$$\hat{H} = \mathbf{H} + \hat{V}_{\mathrm{H}},$$

with

$$\hat{V}_{\mathrm{H}} = \hat{V}_{\mathrm{H}}\left(\left[\Psi_{\mathrm{H}} \otimes \Psi_{\mathrm{H}}\right]_{:1}\right) = \frac{1}{4\,\pi}\int_{\mathbb{R}^3} V(x,z)\left[\Psi_{\mathrm{H}} \otimes \Psi_{\mathrm{H}}\right]_{:1}(t,z,z)\,dz.$$

From a purely algebraic point of view, contrary to the TDHF approximation, the TDH conserves the form of the initial *ansatz*. That is, if one consider an initial data Ψ_{H} to be a Hartree product, then the solution is also a Hartree Product for all time.

1.2.5 Mathematical tools for the analysis of TDH and TDHF models

From the mathematical point of view, the TDH and the TDHF models are well-known. They have a huge mathematical literature. In this section, we will present only basic well-posedness results of the associated Cauchy problems. Also, we will give the main tools that allow to obtain such results. Moreover, we shall consider only the Singular Coulomb interaction V and U for simplicity. Given $\phi^0 \in L^2(\mathbb{R}^3)$ such that $\|\phi\|_{L^2(\mathbb{R}^3)}$ and a family of N functions $\phi_1^0, \ldots, \phi_N^0 \in L^2(\mathbb{R}^3)$ such that $\langle \phi_i, \phi_j \rangle_{L^2(\mathbb{R}^3)} = \delta_{i,j}$ for all $1 \leq i,j \leq N$, then the Cauchy problems associated to the TDHF and TDH read respectively

$$\mathscr{S}_{\mathrm{HF}}: \quad \left\{ \begin{array}{rcll} i\frac{\partial}{\partial t}\,\phi_i(t,x) & = & \mathbf{H}_x\,\phi(t,x) + \sum_{j=1}^N \int_{\mathbb{R}^3} \frac{|\phi_j(t,y)|^2}{|x-y|}\,dy\,\phi_i(t,x) & \\ & & -\sum_{j=1}^N \int_{\mathbb{R}^3} \frac{\phi_j(t,y)\,\overline{\phi}_i(t,y)}{|x-y|}\,dy\,\phi_j(t,x) & 1 \leq i \leq N, \\ \phi_i(t=0) & = & \phi_i^0, \quad 1 \leq i \leq N. & \end{array} \right.$$

and

$$\mathscr{S}_{\mathrm{H}}: \quad \left\{ \begin{array}{rcl} i\frac{\partial}{\partial t}\,\phi(t,x) & = & \mathbf{H}_x\,\phi(t,x) + \int_{\mathbb{R}^3} \frac{|\phi(t,y)|^2}{|x-y|}\,dy\,\phi(t,x) \\ \phi(t=0) & = & \phi^0. \end{array} \right.$$

The analysis of the two problems $\mathscr{S}_{\mathrm{HF}}$ and $\mathscr{S}_{\mathrm{H}}$ is based on the same arguments.

Energy space analysis

The "energy space" is a name commonly used in the mathematical and physical literature. This space depends obviously on the nature of the equations under study since it is nothing but the Hilbert space in which the functional energy is well-defined - a typical example is H^1 since the gradient of the wavefunction corresponds to the momentum and kinetic energy is the square of the momentum.

First of all, recall the Hardy inequality which is one of the main tools when dealing with the Singular Coulomb potential. Let $\phi \in H^1(\mathbb{R}^3,\mathbb{C})$ where

$$H^1(\mathbb{R}^3,\mathbb{C}) = \left\{ \phi \in L^2(\mathbb{R}^3), \quad \nabla\phi \in L^2(\mathbb{R}^3) \right\}.$$

Then,

$$\left\| \frac{\phi}{|x|} \right\|_{L^2(\mathbb{R}^3)} \leq 2\,\|\nabla\phi\|_{L^2(\mathbb{R}^3)}. \tag{1.16}$$

Now, recall the HF energy

$$\begin{array}{rcl} \mathscr{E}_{\mathrm{HF}}(t) & = & \frac{1}{2}\sum_{i=1}^N \int_{\mathbb{R}^3} \left(|\nabla\phi_i(t,x)|^2 - \sum_{k=1}^M \frac{Z_k\,|\phi_i(t,x)|^2}{|x-R_k|} \right) dx \\ & + & \frac{1}{4}\sum_{i,j=1}^N \int_{\mathbb{R}^6} \frac{\phi_i(t,x)\overline{\phi}_i(t,x)\,\phi_j(t,y)\overline{\phi}_j(t,y)}{|x-y|}\,dx\,dy \\ & - & \frac{1}{4}\sum_{i,j=1}^N \int_{\mathbb{R}^6} \frac{\phi_i(t,x)\overline{\phi}_j(t,x)\,\phi_j(t,y)\overline{\phi}_i(t,y)}{|x-y|}\,dx\,dy, \end{array}$$

and the Hartree energy

$$\begin{aligned}\mathscr{E}_{\mathrm{H}}(t) &= \frac{1}{2}\sum_{i=1}^{N}\int_{\mathbb{R}^3}\left(|\nabla\phi_i(t,x)|^2-\sum_{k=1}^{M}\frac{Z_k\,|\phi_i(t,x)|^2}{|x-R_k|}\right)dx\\ &+ \frac{1}{4}\sum_{i,j=1}^{N}\int_{\mathbb{R}^6}\frac{\phi_i(t,x)\overline{\phi}_i(t,x)\,\phi_j(t,y)\overline{\phi}_j(t,y)}{|x-y|}\,dx\,dy.\end{aligned}$$

Next, let us see in any space theses quantities are well-defined, finite in other words. First, obviously $\int_{\mathbb{R}^3}|\nabla\phi(t,x)|^2\,dx$ and $\int_{\mathbb{R}^3}|\nabla\phi_i(t,x)|^2\,dx$ are well-defined for $\nabla\phi,\nabla\phi_1,\ldots,\nabla\phi_N\in L^2(\mathbb{R}^3,\mathbb{C})$. Next, for a fixed $1\leq j\leq M$, we have

$$\begin{aligned}\int_{\mathbb{R}^3}\frac{|\phi_i(t,x)|^2}{|x-R_j|}dx &\leq \|\frac{\phi_i}{|x-R_j|}\|_{L^2(\mathbb{R}^3)}\,\|\phi_i\|_{L^2(\mathbb{R}^3)}\\ &\leq 2\,\|\nabla\phi_i\|_{L^2(\mathbb{R}^3)}\,\|\phi_i\|_{L^2(\mathbb{R}^3)}.\end{aligned}\tag{1.17}$$

The right hand side is finite for $\phi_i\in H^1(\mathbb{R}^3,\mathbb{C})$. Finally, it remains to estimate the so called *bielectronic integrals*. Let $1\leq i,j,p,q\leq N$, then using (1.17), we get

$$\left|\int_{\mathbb{R}^6}\frac{\phi_i(t,x)\overline{\phi}_j(t,x)\,\phi_p(t,y)\overline{\phi}_q(t,y)}{|x-y|}\,dx\,dy\right| \leq 2\,\|\nabla\phi_i\|_{L^2(\mathbb{R}^3)}\,\|\phi_j\|_{L^2(\mathbb{R}^3)}\,\|\phi_p\|_{L^2(\mathbb{R}^3)}\,\|\phi_q\|_{L^2(\mathbb{R}^3)}.$$

Again, the right hand side, hence the left hand side, is finite if and only if $\nabla\phi_i\in L^2(\mathbb{R}^3)$ and $\phi_j,\phi_p,\phi_q\in L^2(\mathbb{R}^3)$. We deduce then that $\mathscr{E}_{\mathrm{H}}$ and $\mathscr{E}_{\mathrm{HF}}$ are respectively finite if and only if ϕ and the ϕ_i's are in $H^1(\mathbb{R}^3,\mathbb{C})$.

The system $\mathscr{S}_{\mathrm{HF}}$ is known to enjoy

Theorem 1.1. *Let $\phi_i^0\in H^1\big(\mathbb{R}^3\big)$ for all $i=1,\ldots,N$. Then the Cauchy problem $\mathscr{S}_{\mathrm{HF}}$ has a unique mild solution $\phi_1(t,x),\ldots,\phi_N(t,x)$ satisfying*

$$\phi_i\in C^1\big([0,+\infty);H^{-1}(\mathbb{R}^3)\big)\cap C^0\big([0,+\infty);H^1(\mathbb{R}^3)\big),\quad \textit{for all}\quad 1\leq i\leq N.$$

In the same way, the system $\mathscr{S}_{\mathrm{HF}}$ is known to enjoy

Theorem 1.2. *Let $\phi^0\in H^1\big(\mathbb{R}^3\big)$ for all $i=1,\ldots,N$. Then the Cauchy problem $\mathscr{S}_{\mathrm{H}}$ has a unique mild solution $\phi(t,x)$ satisfying*

$$\phi\in C^1\big([0,+\infty);H^{-1}(\mathbb{R}^3)\big)\cap C^0\big([0,+\infty);H^1(\mathbb{R}^3)\big).$$

The theorems 1.1 and 1.2 provide then, respectively, with a global-in-time existence of mild solutions to the Cauchy problems associated to the TDHF approximation $\mathscr{S}_{\mathrm{HF}}$ and the TDH one $\mathscr{S}_{\mathrm{HF}}$. Both results were proved by Chadam and Glassey in [15]. The main lines of the proofs are the same. First of all, consider the free Schrödinger equation

$$i\frac{\partial}{\partial t}\,\psi(t,x)=-\frac{1}{2}\Delta_x\psi(t,x)-\sum_{k=1}^{M}\frac{Z_k}{|x-R_k|}\,\psi(t,x),\quad \psi(t=0)=\psi^0.$$

We claim that if $\psi^0 \in H^1\left(\mathbb{R}^3\right)$, then the equation above admits a unique mild solution in $H^1\left(\mathbb{R}^3\right)$. This a direct consequence of the fact that the operator U is $\Delta-$bounded (Hardy inequality) and that the operator $i\Delta$ generates a group of isometries on $L^2(\mathbb{R}^3)$. Next, one use Hardy inequality (1.16) in order to conclude. Briefly speaking, on can associate a uniformly bounded propagator to the self-adjoint operator $-\frac{1}{2}\Delta_x - \sum_{k=1}^{M} \frac{Z_k}{|x-R_k|}$ that we denote $\mathscr{T}(t)$. In particular, $\|\mathscr{T}\,\psi\|_{H^1\left(\mathbb{R}^3\right)} \leq$ const. $\|\psi\|_{H^1\left(\mathbb{R}^3\right)}$. The second step consists in showing that the non-linearities are locally lipschitz. Typically, recall that we deal with Coulomb interactions, the main ingredients are the Cauchy-Schwarz and Hardy inequalities (1.16). For illustration, we estimate one term. Thus, given $\phi_1,\ldots,\phi_N,\psi_1,\ldots,\psi_N \in H^1\left(\mathbb{R}^3\right)$, we have

$$
\begin{aligned}
\| \int_{\mathbb{R}^3} \frac{|\phi_j|^2(t,y)}{|x-y|}\,dy\,\phi_i(t,x) \;-\; & \int_{\mathbb{R}^3} \frac{|\psi_j|^2(t,y)}{|x-y|}\,dy\,\psi_i(t,x)\,\|_{L^2(\mathbb{R}^3)} \\
\leq\; & \int_{\mathbb{R}^3} \frac{|\phi_j|^2(t,y)}{|x-y|}\,dy\,\|\phi_i-\psi_i\|_{L^2(\mathbb{R}^3)} \\
+\; & \int_{\mathbb{R}^3} \frac{|\phi_j|^2(t,y)-|\psi_j|^2(t,y)}{|x-y|}\,dy\,\|\psi_i\|_{L^2(\mathbb{R}^3)}, \\
\leq\; & 2\,\|\nabla\phi_j\|_{L^2(\mathbb{R}^3)}\,\|\phi_j\|_{L^2(\mathbb{R}^3)}\,\|\phi_i-\psi_i\|_{L^2(\mathbb{R}^3)} \\
+\; & 2\,\|\nabla\phi_j\|_{L^2(\mathbb{R}^3)}\,\|\phi_j-\psi_j\|_{L^2(\mathbb{R}^3)}\,\|\psi_i\|_{L^2(\mathbb{R}^3)} \\
+\; & 2\,\|\phi_j-\psi_j\|_{L^2(\mathbb{R}^3)}\,\|\nabla\psi_j\|_{L^2(\mathbb{R}^3)}\,\|\psi_i\|_{L^2(\mathbb{R}^3)}, \\
\leq\; & \text{const.}_1\,\|\phi-\psi\|_{L^2(\mathbb{R}^3)}.
\end{aligned}
$$

In the last line above, const.$_1$ depends obviously on $\|\phi\|_{H^1\left(\mathbb{R}^3\right)}$ and $\|\psi\|_{H^1\left(\mathbb{R}^3\right)}$ where, ϕ denotes the N-component vector $(\phi_1,\ldots,\phi_N)$ and equivalently for ψ and $\|\phi\|_{H^1\left(\mathbb{R}^3\right)} = \sqrt{\sum_{i=1}^{N} \|\phi_i\|^2_{H^1\left(\mathbb{R}^3\right)}}$ for instance. Next, the gradient part is handled by the same argument as the one above with the

following estimate

$$\begin{aligned}
\|\nabla\left[\int_{\mathbb{R}^3}\frac{|\phi_j|^2(t,y)}{|x-y|}\,dy\,\phi_i(t,x)\right]\|_{L^2(\mathbb{R}^3)} &\leq \left|\int_{\mathbb{R}^3}\frac{|\phi_j|^2(t,y)(x-y)}{|x-y|^3}\,dy\right|\,\|\phi_i\|_{L^2(\mathbb{R}^3)}\\
&+ \int_{\mathbb{R}^3}\frac{|\phi_j|^2(t,y)}{|x-y|}\,dy\,\|\nabla\phi_i\|_{L^2(\mathbb{R}^3)},\\
&\leq \|\frac{\phi_j}{|x-y|}\|^2_{L^2(\mathbb{R}^3)}\,\|\phi_i\|_{L^2(\mathbb{R}^3)}\\
&+ 2\,\|\nabla\phi_j\|_{L^2(\mathbb{R}^3)}\,\|\phi_j\|_{L^2(\mathbb{R}^3)}\,\|\nabla\phi_i\|_{L^2(\mathbb{R}^3)},\\
&\leq 4\,\|\nabla\phi_j\|^2_{L^2(\mathbb{R}^3)}\,\|\nabla\phi_i\|_{L^2(\mathbb{R}^3)}\\
&+ 2\,\|\nabla\phi_j\|_{L^2(\mathbb{R}^3)}\,\|\phi_j\|_{L^2(\mathbb{R}^3)}\,\|\nabla\phi_i\|_{L^2(\mathbb{R}^3)}.
\end{aligned}$$

Thus, one obtain

$$\begin{aligned}
\|\nabla\left(\int_{\mathbb{R}^3}\frac{|\phi_j|^2(t,y)}{|x-y|}\,dy\,\phi_i(t,x)\right. &- \left.\int_{\mathbb{R}^3}\frac{|\psi_j|^2(t,y)}{|x-y|}\,dy\,\psi_i(t,x)\right)\|_{L^2(\mathbb{R}^3)}\\
&\leq \text{const.}_2\,\|\nabla(\phi-\psi)\|_{L^2(\mathbb{R}^3)}.
\end{aligned}$$

Finally,

$$\begin{aligned}
\|\int_{\mathbb{R}^3}\frac{|\phi_j|^2(t,y)}{|x-y|}\,dy\,\phi_i(t,x) &- \int_{\mathbb{R}^3}\frac{|\psi_j|^2(t,y)}{|x-y|}\,dy\,\psi_i(t,x)\|_{H^1\left(\mathbb{R}^3\right)}\\
&\leq (\text{const.}_1+\text{const.}_2)\,\|\phi-\psi\|_{H^1\left(\mathbb{R}^3\right)}.
\end{aligned}$$

Above, we proved that the TDH non-linearity is locally lipschitz from $H^1\left(\mathbb{R}^3\right)$ into itself. This term is the first one of the TDHF non-linearity, the second term can be estimated in the same spirit using the same arguments. This allows to apply a fixed point Theorem and obtain a local-in-time existence and uniqueness of solutions to the Cauchy problems $\mathscr{S}_{\mathrm{H}}$ and $\mathscr{S}_{\mathrm{H}}$. More precisely, in the class

$$C^1\left([0,\tau];H^{-1}(\mathbb{R}^3)\right)\cap C^0\left([0,\tau];H^1(\mathbb{R}^3)\right)$$

for a time $\tau>0$ small enough. The existence of solution behind the time τ is equivalent to the existence of uniform *a priori* estimates (see [27]). In the case of TDH and TDHF, these estimates are obtained from the conservation laws satisfied by the dynamics of the systems. In the TDH case, we have the conservation of the *mass* translated by the formula (1.12). This proves, in particular, that the L^2 norm of the solutions is conserved during time propagation. However, this is not enough since we aim to prove space energy solutions. Thus, we need an estimate on the H^1 norm. This will be a consequence of the energy conservation for which we refer to [15] fo both cases, TDH and TDHF. However, again, we warn the reader that the proof is far from

being optimal and we will be improved in [12] by taking advantage from the variational nature of these problems. Typically, we shall prove that the TDH and TDHF satisfy the variational principle (2.5) and automatically conserve their total energy following (2.6). Then, recall the so called *Kato inequality*. This inequality, tell us that if $V \in L^{3/2}(\mathbb{R}^3,\mathbb{R}) + L^{\infty}(\mathbb{R}^3,\mathbb{R})$, then for all $\varepsilon > 0$, there exists a constant $C_\varepsilon > 0$ such that

$$\left\langle \Psi \left| \mathscr{H} \right| \Psi \right\rangle_{L^2(\mathbb{R}^{3N},\mathbb{C})} \geq \frac{1-\varepsilon}{2} \int_{\mathbb{R}^{3N}} |\nabla \Psi|^2 dX_N - C_\varepsilon \int_{\mathbb{R}^{3N}} |\Psi|^2 dX_N.$$

Then, replacing Ψ by Ψ_{HF} or Ψ_{H} and use the fact that these two wavefunctions are normalized to one in $L^2(\mathbb{R}^{3N},\mathbb{C})$, one obtain a bound on $\|\nabla\Phi\|_{L^2(\mathbb{R}^3)}$, thus on the $H^1\left(\mathbb{R}^3\right)$ norm. Observe that the sign of U is in the convenient sense here. Finally, it is also easy to prove propagation of regularity, this is also a standard result and we refer for instance to [11] for the Schrödinger-Poisson case which is nothing but the Hartree model with Coulomb interactions. We will also improve this result. Observe that we considered only Coulomb type interactions, however, well-posdness result exist for more general potential. Actually, we will consider potentials in $L^p(\mathbb{R}^3,\mathbb{R}) + L^q(\mathbb{R}^3,\mathbb{R})$ with some assumptions on p and q. From now, in all this introduction, we shall denote $U,V \in L^p$ with $p > \frac{3}{2}$ instead of $U,V \in L^p(\mathbb{R}^3,\mathbb{R}) + L^q(\mathbb{R}^3,\mathbb{R})$ with the same assumption on p and q.

For more details about well-posdness results for the TDH and TDHF we refer to the main paper of Chadam and Glassey [15], but also to [28, 18, 9] and references therein. Moreover, the authors of [1] tackled the study of the Von-Newman type equation associated to the TDHF in the framework of open quantum systems. Finally, we refer to [5] for the analysis of the accuracy of the TDHF approximation and [4] for a rigorous derivation of the TDHF system with bounded interactions.

Finally, we want to emphasize the fact that in this section we presented some formals tools that allows to conclude. However, the robust and rigorous way to prove the well-posedeness in the energy space is to start first by proving the existence of Classical solutions in H^2 with initial data with the same regularity. This is due to the domain of the Laplacian. The result is also obtained by a fixed point Theorem. Next, on can construct the unique mild solution in the energy space as the limit of a sequence of solutions in H^2.

L^2 analysis

By L^2 analysis, we mean analysis of problems starting with initial data with regularity, only, L^2. With such regularity, quantum systems has finite mass but infinite energy. The main ingredients are the properties of the propagator associated to $\frac{i}{2}\Delta$, we denote it $\mathscr{U}(t)$ and Strichartz type estimates. First of all, we denote for all $T \geq 0$

$$L^{p,q}_T := L^p([0,T],L^q(\mathbb{R}^3,\mathbb{C})).$$

Moreover, p' will denote the conjugate of $1 \leq p \leq \infty$, that is $\frac{1}{p} + \frac{1}{p'} = 1$. Finally, for a given $2 \leq q < 6$, we say that the pair of reals (p,q) is admissible, we denote $(p,q) \in \mathscr{A}$, if and only if

$$\frac{2}{3p} = \frac{1}{2} - \frac{}{q}.$$

Then, for all $T>0$, $\phi\in L^2(\mathbb{R}^3),(p,q)\in\mathscr{A}$ and $\psi\in L_T^{a',b'}$ such that $(a,b)\in\mathscr{A}$, we have

$$\|\,\mathscr{U}(t)\,\phi\,\|_{L_T^{p,q}}\leq\rho(p)\,\|\phi\|_{L^2},\tag{1.18}$$

and

$$\|\int_0^t\mathscr{U}(t-s)\,\psi(s)ds\,\|_{L_T^{p,q}}\leq\rho(p,a)\,\|\psi\|_{L_T^{a',b'}},\tag{1.19}$$

with ρ being a constant with its dependency indicated between the parenthesis. Moreover

$$\mathscr{U}(t-s)\,\psi(s)ds\in C([0,T],L^2(\mathbb{R}^3)).$$

The inequality (1.18) describes a remarkable smoothing effect. In particular it tell us that for all $t\in\mathbb{R}$, we have $\mathscr{U}(t)\,L^2(\mathbb{R}^3)=L^2(\mathbb{R}^3)$ and that for all $\phi\in L^2(\mathbb{R}^3)$, we have obviously $\mathscr{U}(t)\,\phi\in L^p(\mathbb{R}^3,\mathbb{C})$. (4.7) is crucial when dealing with non-linearities in the framework of Schrödinger-type equations. Indeed, without loss of generality we write the following Duhamel formula for a Schrödinger-type equation

$$\psi(t)=\mathscr{U}(t)\,\phi-i\int_0^t\mathscr{U}(t-s)\,f(\psi(s)),$$

for a given functional f. Then, (1.18) allows to control the $L^2(\mathbb{R}^3)$ norm of $\mathscr{U}(t)\,\phi$ in terms of the $L^2(\mathbb{R}^3)$ one of ϕ. However, it is merely impossible to control the $L^2(\mathbb{R}^3)$ norm of $\int_0^t\mathbf{U}(t-s)\,f(\psi(s))\,ds$ in terms of the one of ψ for a general non-linearities f. (4.7) will, then, give us the possibility to control the $L_T^{p,q}$ norm of $\int_0^t\mathbf{U}(t-s)\,f(\psi(s))\,ds$ for a given $T>0$ and a couple of reals $(p,q)\in\mathscr{A}$ in terms of the $L_T^{a',b'}$ norm of f for any admissible pair $(a,b)\in\mathscr{A}$ which is in general enough for us to conclude. We shall need also *Young* inequality that we recall for $\phi\in L^p(\mathbb{R}^3,\mathbb{C})$ and $\psi\in L^p(\mathbb{R}^3,\mathbb{C})$

$$\|\phi\star\psi\|_{L^r(\mathbb{R}^3,\mathbb{C})}\leq\|\phi\|_{L^p(\mathbb{R}^3,\mathbb{C})}\,\|\psi\|_{L^q(\mathbb{R}^3,\mathbb{C})},\tag{1.20}$$

where the $\star$ denotes the convolution operator.

For illustration, we consider the TDH case with Coulomb interaction and set $U\equiv0$ for simplification. However the result is obviously true for Coulomb U. We are the interested in the following system

$$\mathscr{S}_{\mathrm{H}}:\quad\begin{cases}i\frac{\partial}{\partial t}\,\phi(t,x) &= -\frac{1}{2}\,\Delta_x\,\phi(t,x)+\int_{\mathbb{R}^3}\frac{|\phi(t,y)|^2}{|x-y|}\,dy\,\phi(t,x)\\ \\ \phi(t=0) &= \phi^0\in L^2(\mathbb{R}^3).\end{cases}$$

The first thing to observe is that the Coulomb potential lives in the class

$$L^a(\mathbb{R}^3,\mathbb{R})+L^b(\mathbb{R}^3,\mathbb{R}),$$

for, at least, a pair of reals a,b such that $a\in[\frac{3}{2},3[$ and $b\in]3,+\infty[$. That is $\frac{1}{r}=V_1+V_2$ with $V_1\in L^a(\mathbb{R}^3,\mathbb{R})$ and $V_2\in L^b(\mathbb{R}^3,\mathbb{R})$. This can be proved by a simple cut-off and we refer to the fourth chapter of this manuscript for instance. From this point onward we set $\frac{3}{2}<a=d<3$ and $b=\infty$. Thus, the TDH non-linearity splits into two parts, $f_1(\phi)$ associated with V_1 and $f_2(\phi)$ associated with V_2.

Next, we shall prove the following theorem

Theorem 1.3. *The Cauchy problem $\mathscr{S}_{\mathrm{H}}$ has a unique mild solution*

$$\phi \in C(\mathbb{R}, L^2(\mathbb{R}^3)) \cap L^{p,q}_{loc}, \quad (p,q) \in \mathscr{A} \quad s.t. \quad 3 < p < 6.$$

In order to sketch the proof, we need the following

Lemma 1.1. *Let $T > 0$ small enough, $p = \frac{2d}{d-1}$,q such that $(p,q) \in \mathscr{A}$. Moreover, denote $M := \max_{[0,T]}(\|\phi\|_{L^2(\mathbb{R}^3)}, \|\psi\|_{L^2(\mathbb{R}^3)})$. Then, for all $\psi, \phi \in C([0,T], L^2(\mathbb{R}^3)) \cap L^{p,q}_T$ we have*

$$\begin{aligned}
\|f_1(\phi) - f_1(\psi)\|_{L^{p',q'}_T} &\leq \rho(p)\, M^2\, T^{1-\frac{2}{p}}\, \|\phi - \psi\|_{L^{p,q}_T} \\
\|f_2(\phi) - f_2(\psi)\|_{L^{p',q'}_T} &\leq \rho\, M^2\, T\, \|\phi - \psi\|_{L^{\infty,2}_T} \\
&\leq \rho\, M^2\, T^{1-\frac{2}{p}}\, \|\phi - \psi\|_{L^{\infty,2}_T}
\end{aligned}$$

The proof is nothing but Young an Hölder inequalities in space and Hölder inequality in time and we refer the reader to [11, 30] for a detailed proof.

Now, consider a sequence of initial data $\phi^0_n \in H^1(\mathbb{R}^3)$ such that

$$\phi^0_n \overset{n\to\infty}{\longrightarrow} \phi^0 \in H^1(\mathbb{R}^3), \quad \|\phi^0_n\|_{H^1(\mathbb{R}^3)} = \|\phi^0\|_{H^1(\mathbb{R}^3)}.$$

Hence, thanks to Theorem 1.2, to every $n \in \mathbb{N}$, the TDHF cauchy problem $\mathscr{S}_{\mathrm{H}}$ with initial data ϕ^0_n has a unique solution ϕ_n in the class $C([0,+\infty), H^1(\mathbb{R}^3))$ such that $\|\phi_n(t)\|_{L^2(\mathbb{R}^3)} = \|\phi^0_n\|_{L^2(\mathbb{R}^3)}$ for all time $t \geq 0$. Next, let $n \neq k \in \mathbb{N}$, an write the difference of the associated Duhamel formulae

$$\begin{aligned}
\phi_n(t) - \phi_k(t) = \mathscr{U}(t)(\phi^0_n - \phi^0_k) \quad & -i \int_0^t \mathscr{U}(t-s)\,(f_1(\phi_n(s)) - f_1(\phi_k(s)))\, ds \\
& -i \int_0^t \mathscr{U}(t-s)\,(f_2(\phi_n(s)) - f_2(\phi_k(s)))\, ds.
\end{aligned}$$

Using (1.18), we have

$$\|\mathscr{U}(t)(\phi^0_n - \phi^0_k)\|_{L^{a,b}_T} \leq \rho(a)\, \|\phi^0_n - \phi^0_k\|_{L^2(\mathbb{R}^3)}.$$

Next, using (4.7) and the Lemma 1.1, we get for all $(a,b) \in \mathscr{A}$

$$\begin{aligned}
\Big\| \int_0^t \mathscr{U}(t-s)\,(f_1(\phi_n(s)) - f_1(\phi_k(s)))\, ds \Big\|_{L^{a,b}_T} &\leq \rho(a,p) \|f_1(\phi_n) - f_1(\phi_k)\|_{L^{p',q'}_T} \\
&\leq \rho(a,p)\, M^2\, T^{1-\frac{2}{p}} \|\phi_n - \phi_k\|_{L^{p,q}_T}.
\end{aligned}$$

Also,

$$\Big\| \int_0^t \mathscr{U}(t-s)\,(f_1(\phi_n(s)) - f_1(\phi_k(s)))\, ds \Big\|_{L^{a,b}_T} \leq \rho(a)\, M^2\, T^{1-\frac{2}{p}} \|\phi_n - \phi_k\|_{L^{\infty,2}_T}.$$

The inequalities above being valid for all admissible pair $(a,b) \in \mathscr{A}$, one can write them from the one hand for $(a,b) = (p,q) \in \mathscr{A}$ and from the other hand for $(a,b) = (\infty,2)$ and sum up. This

leads to

$$\|\phi_n(t)-\phi_k(t)\|_{L_T^{p,q}}+\|\phi_n(t)-\phi_k(t)\|_{L_T^{\infty,2}}\leq \rho(p)\|\phi_n^0-\phi_k^0\|_{L^2(\mathbb{R}^3)} \\ +\rho(p)\,M^2\,T^{1-\frac{2}{p}}\left(\|\phi_n(t)-\phi_k(t)\|_{L_T^{p,q}}+\|\phi_n(t)-\phi_k(t)\|_{L_T^{\infty,2}}\right)$$

In particular, there exists a time $T'=T'(p,M)$ such that

$$\|\phi_n(t)-\phi_k(t)\|_{L_{T'}^{p,q}}+\|\phi_n(t)-\phi_k(t)\|_{L_{T'}^{\infty,2}}\leq \rho(p,M)\|\phi_n^0-\phi_k^0\|_{L^2(\mathbb{R}^3)} \tag{1.21}$$

Thus, on can deduce the following properties

- $\{\phi_n\}$ is a Cauchy sequence in $L_{T'}^{p,q}\cap L_{T'}^{\infty,2}$.
- $\{\phi_n\}\in C([0,T'],L^2(\mathbb{R}^3))$.
- $\phi_n\overset{n\longrightarrow\infty}{\longrightarrow}\phi$ in $L_{T'}^{p,q}\cap C([0,T'],L^2(\mathbb{R}^3))$.
- $\|\phi_n(t)\|_{L^2(\mathbb{R}^3)}=\|\phi_n^0\|_{L^2(\mathbb{R}^3)}=\|\phi^0\|_{L^2(\mathbb{R}^3)}$ for all $n\in\mathbb{N}$ and $t>0$.

Next, the easy observation that T' depends only on conserved quantities, namely, M allows to reiterate the argument and cover the hole real line $T',2T',3T',\ldots$. Thus obtain the fact that $\phi\in C([0,+\infty),L^2(\mathbb{R}^3))\cap L_{loc}^{p,q}$. The uniqueness point and the equation satisfied by the limit ϕ are obvious to obtain in view of the estimate (4.37) and the ones of the Lemma 1.1 respectively.

Castella in [11] tackled the L^2 analysis TDH case with Coulomb interactions. However, the author considered a system of infinitely many coupled equations. He introduced a scaled functional spaces in order to remediate to this point. Also, Zagatti in [30], considered a system of infinite TDHF type equations on which he established an L^2 existence theory. The second Chapter of the present manuscript generalizes, in some sense, its result.

1.3 The Multiconfiguration time-dependent approximations

This section is devoted to the presentation of the so called *Multiconfiguration* methods which are the main object of the present contribution. The idea behind these models is simple and, depending on the nature of the system under study, is based on the following

$$L_{\mathrm{s}}^2(\mathbb{R}^{3N})\simeq\bigotimes_{i=1}^{N}L^2(\mathbb{R}^3),\quad L_{\mathrm{sk}}^2(\mathbb{R}^{3N})\simeq\bigwedge_{i=1}^{N}L^2(\mathbb{R}^3).$$

with subscript s and sk standing respectively for symmetric and skew-symmetric. These models are know to be the generalization of the Hartree and the Hartree-Fock approximations.

1.3.1 The multiconfiguration time-dependent Hartree-Fock method

The MCTDHF approximation is the adequate method for approximating the time evolution of a quantum system composed of N fermions, electrons in particular. In opposition to the TDHF case where one approximate the wavefunction by a single Slater determinant, the MCTDHF uses a combination of Slater determinants in order to approximate the wavefunction. Intuitively, the more determinants involved in the *ansatz*, the better is the approximation. Let us now, introduce more precisely the MCTDHF.

Let $K \geq N$ be an integer and $\Sigma_{N,K}$ denotes the range of the family of increasing mappings

$$\sigma : \quad \{1,\dots,N\} \longrightarrow \{1,\dots,K\}, \quad \text{for} \quad N,K \in \mathbb{N}^\star.$$

In other words,

$$\Sigma_{N,K} = \Big\{ \sigma = \{\sigma(1) < \ldots < \sigma(N)\} \subset \{1,\dots,K\} \Big\}, \qquad |\Sigma_{N,K}| = \binom{K}{N} := r.$$

For simplicity, we shall use the same notation for the mapping σ and its signature $\{\sigma(1) < \ldots < \sigma(N)\}$. Moreover, for $i \in \sigma$, $\sigma^{-1}(i)$ will denotes the postion (the rank) of the integer i in the set σ. Next, introduce

$$\mathscr{O}_{L^2(\mathbb{R}^3)^K} = \left\{ \Phi = (\phi_1,\dots,\phi_K) \in L^2(\mathbb{R}^3)^K, \quad \int_{\mathbb{R}^3} \phi_i\, \bar{\phi}_j\, dx = \delta_{i,j} \right\}. \tag{1.22}$$

Also we introduce the unit sphere

$$\mathscr{S}^{r-1} = \left\{ C = (C_\sigma)_{\sigma\in\Sigma_{N,K}} \in \mathbb{C}^r, \quad \sum_{\sigma\in\Sigma_{N,K}} |c_\sigma|^2 = 1 \right\}. \tag{1.23}$$

The notation $\sum_{\sigma\in\Sigma_{N,K}}$ means that the sum runs over all the mappings σ of $\Sigma_{N,K}$. Now, given $\sigma \in \Sigma_{N,K}$, we define the associated *Slater determinant* as follows

$$\Phi_\sigma(x_1,\dots,x_N) = \frac{1}{\sqrt{N!}} \begin{vmatrix} \phi_{\sigma(1)}(x_1) & \dots & \phi_{\sigma(1)}(x_N) \\ \vdots & & \vdots \\ \phi_{\sigma(N)}(x_1) & \dots & \phi_{\sigma(N)}(x_N) \end{vmatrix}.$$

That is, the determinant built from the ϕ_i's such that $i \in \sigma$. The vector Φ being in $\mathscr{O}_{L^2(\mathbb{R}^3)^K}$, the factor $\frac{1}{\sqrt{N!}}$ ensures then the normalization $\|\Phi_\sigma\|_{L^2(\mathbb{R}^{3N},\mathbb{C})} = 1$. Also, it is straightforward that for $\sigma,\tau \in \Sigma_{N,K}$, we have

$$\int_{\mathbb{R}^{3N}} \Phi_\sigma(x_1,\dots,x_N)\, \overline{\Phi}_\tau(x_1,\dots,x_N)\, dx_1,\dots,dx_N = \delta_{\sigma,\tau}.$$

The MCTDHF method consists, then, in forcing the wavefunction to evolve on the following set

$$\mathscr{B}_{N,K} = \left\{ \Psi(X_N) = \sum_{\sigma\in\Sigma_{N,K}} C_\sigma\, \Phi_\sigma(X_N), \quad (C,\Phi) \in \mathscr{F}_{N,K} \right\} \subset L^2_{\mathrm{sk}}(\mathbb{R}^{3N},\mathbb{C}), \tag{1.24}$$

where

$$\mathscr{F}_{N,K} := \mathscr{S}^{r-1} \times \mathscr{O}_{L^2(\mathbb{R}^3)^K}.$$

Now, following the *ansatz* in (1.24), we have from the one side

$$[\Psi \otimes \Psi]_{:1}(x,y) \quad = \quad \sum_{i,j=1}^{K} \sum_{\substack{\sigma,\tau \subset \Sigma_{N,K},\, i\in\sigma,\, j\in\tau; \\ \text{s.t. } \sigma\setminus\{i\}=\tau\setminus\{j\}}} (-1)^{\sigma^{-1}(i)+\tau^{-1}(j)}\, C_\sigma\, \overline{C}_\tau\, \phi_i(x)\, \overline{\phi}_j(y).$$

Thus, if one introduce the $K \times K$ Hermitian matrix

$$\mathbf{\Gamma}[C]_{i,j} = \sum_{\substack{\sigma,\tau\in\Sigma_{N,K},\, i\in\sigma, j\in\tau; \\ \{\sigma\setminus i\}=\{\tau\setminus j\}}} (-1)^{\sigma^{-1}(i)}\, \overline{C}_\sigma \quad (-1)^{\tau^{-1}(j)}\, C_\tau, \tag{1.25}$$

one has

$$[\Psi \otimes \Psi]_{:1}(x,y) = \sum_{i,j=1}^{K} \overline{\mathbf{\Gamma}[C]}_{j,i}\, \phi_i(x)\, \overline{\phi}_j(y).$$

>From the opposite side

$$\big[\Psi \otimes \Psi\big]_{:2}(x,y,x',y') \quad = \quad \sum_{i,p,j,q=1}^{K} \gamma_{i,p,j,q}\, \phi_i(x)\, \phi_p(y)\, \overline{\phi}_j(x')\, \overline{\phi}_q(y'),$$

with

$$\gamma_{i,p,j,q} = \frac{1}{2}(1-\delta_{i,p})(1-\delta_{j,q}) \sum_{\substack{\sigma,\tau\, s.t.\, i\neq p\in\sigma\ j\neq q\in\tau \\ \sigma\setminus\{i,p\}=\tau\setminus\{j,q\}}} (-1)^{\sigma}_{i,p}(-1)^{\tau}_{j,q}\, C_\sigma\, \overline{C}_\tau, \tag{1.26}$$

and

$$(-1)^{\sigma}_{i,p} = \begin{cases} (-1)^{\{\sigma^{-1}(i)+\sigma^{-1}(p)+1\}} & \text{if} \quad p < i \\ \\ (-1)^{\{\sigma^{-1}(i)+\sigma^{-1}(p)\}} & \text{if} \quad p > i \end{cases}$$

Now, using a variational procedure, typically the Dirac-Frenkel variational principle and assuming that an approximate solution satisfying this principle exists, one obtain the following system for the MCTDHF

$$\mathscr{S}_{\text{MCHF}}: \quad \begin{cases} i\frac{d}{dt}\, C(t) = \mathscr{M}[\Phi](t)\, C(t), \\ \\ i\,\mathbf{\Gamma}[C(t)]\, \frac{\partial}{\partial t}\Phi(t,x) = \mathbf{\Gamma}[C(t)]\, \mathbf{H}_x\, \Phi(t,x) + (I - \mathbf{P}_\Phi)\, \mathbb{W}[C,\Phi](t,x)\, \Phi(t,x). \\ \\ (C(t=0),\Phi(t=0)) = (C^0,\Phi^0) \in \mathscr{F}_{N,K}. \end{cases}$$

The ODEs system above involve the $r \times r$ Hermitian matrix $\mathscr{M}[\Phi]$ depending only on the ϕ_i's as follows

$$\mathscr{M}[\Phi]_{\sigma,\tau}(t) = \frac{1}{2} \sum_{i,j\in\tau,\, k,l\in\sigma} \qquad (1-\delta_{i,j})(1-\delta_{k,l})\, \delta_{\tau\setminus\{i,j\},\sigma\setminus\{k,l\}} (-1)^{\tau}_{i,j}\, (-1)^{\sigma}_{k,l} \times$$

$$\times \quad \int_{\mathbb{R}^3\times\mathbb{R}^3} \overline{\phi}_j(t,y)\phi_i(t,y)\, V(|x-y|)\, \overline{\phi}_q(t,x)\, \phi_p(t,x)\, dx\, dy,$$

observe that $\mathscr{M}[\Phi]_{\sigma,\tau}$ doesn't depend on space variables for all $\sigma,\tau\in\Sigma_{N,K}$. The matrix $\mathbf{\Gamma}[C(t)]$ being defined in (1.25). The last matrix involved in the system $\mathscr{S}_{\mathrm{MCHF}}$ is $\mathbb{W}[C,\Phi](t,x)$. It is a $K\times K$ Hermitian matrix, however it depends on time and space. It's entries are

$$\mathbb{W}[C,\Phi]_{ij}(t,x)=2\sum_{k,l=1}^{K}\gamma_{i,j,k,l}\,\overline{\phi}_k(t,y)\,V(|x-y|)\,\phi_l(t,y)\,dy, \tag{1.27}$$

with the coefficients $\gamma_{\dots}$ as in (1.26). The operator $\mathbf{H}_x$ is, as before, the *one particle*, self-adjoint operator, that is acting on $L^2(\mathbb{R}^3)$ and by $\mathbf{H}_x:=-\frac{1}{2}\Delta_x+U(x)$. The notation $\mathbf{H}_x\,\Phi(t,x)$ has to be understood as the vector $(\mathbf{H}_x\,\phi_1(t,x),\dots\mathbf{H}_x\,\phi_K(t,x))$ and equivalently for $\frac{\partial}{\partial t}\Phi(t,x)$. Finally, the operator $\mathbf{P}_\Phi$ is nothing but the orthogonal projector onto the space spanned by the ϕ_i's. That is

$$\mathbf{P}_\Phi(\cdot)=\sum_{k=1}^{K}\langle\,\cdot\,,\,\phi_k\,\rangle_{L^2(\mathbb{R}^3)}\,\phi_k.$$

Observe that we have to deal with a strongly coupled non-linear system of K Schrödinger-type PDEs and r first order differential equations.

The main difficulty of the MCTDHF is the matrix $\mathbf{\Gamma}[C]$. In fact, for the time being there is no rigorous mathematical argument that guarantees the invertibility of this matrix. Moreover, there exist some algebraic cases when this matrix is trivially degenerate. Indeed, consider a 2 particles system, thus K has to be even otherwise the matrix $\mathbf{\Gamma}[C]$ is degenerate. The proof is easy and we refer to [13, 14] for a proof (see also [31]). Moreover, Lewin proved in [31] the dual property, more precisely he proved that for the case $K=N+1$ doesn't allow for an invertible density matrix $\mathbf{\Gamma}[C]$. In the present manuscript, a space energy analysis the MCTDHF system $\mathscr{S}_{\mathrm{MCHF}}$ is performed with initial data with functional part in $H^1\big(\mathbb{R}^3\big)$. Because of the difficulty quoted above, the first result we obtained is valid in *short in time*, however it is also possibly global. In order to state our result, we have to introduce

$$\partial\mathscr{F}_{N,K}=\big\{(C,\Phi)\in\mathscr{F}_{N,K}\quad:\quad\mathrm{rank}(\mathbf{\Gamma}[C])=K\big\}.$$

Then,

Theorem 1.4. *Let $U(x),V(|x|)\in L^d(\mathbb{R}^3,\mathbb{R})$ with $d>\frac{3}{2}$ and $(C^0,\Phi^0)\in\partial\mathscr{F}_{N,K}\cap H^1\big(\mathbb{R}^3\big)^K$. Then, there exists a maximal existence time $T^\star>0$ (possibly $+\infty$) such that:*

i) The MCTDHF system $\mathscr{S}_{\mathrm{MCHF}}$ admits a unique solution (C,Φ) with

$$C\in C^1\big([0,T^\star);\mathbb{C}\big)^r,\qquad\Phi\in C^0\big([0,T^\star);H^1(\mathbb{R}^3)\big)^K\cap C^1\big([0,T^\star);H^{-1}(\mathbb{R}^3)\big)^K.$$

that depends continuously on the initial data (C^0,Φ^0).

ii) For every $0\le t<T^\star$, $\big(C(t),\Phi(t)\big)\in\partial\mathscr{F}_{N,K}$.

iii) For every $0\le t<T^\star$,

$$\Big\langle\Psi_{\mathrm{MC}}(t)\Big|\,H\Big|\Psi_{\mathrm{MC}}(t)\Big\rangle_{L^2(\mathbb{R}^{3N})}=\Big\langle\Psi^0_{\mathrm{MC}}\Big|\,H\Big|\Psi^0_{\mathrm{MC}}\Big\rangle_{L^2(\mathbb{R}^{3N})}.$$

iv) Either $T^\star = +\infty$ *or* $T^\star < +\infty$ *and* $\lim_{t \nearrow T^\star} \|\mathbf{\Gamma}[C(t)]^{-1}\| = +\infty$ *or even*

$$\int_0^{T^\star} \|\mathbf{\Gamma}[C(t)]^{-1}\|^{3/2}\, dt = +\infty.$$

This Theorem then provides with a *short in time* well-posedness to the MCTDHF system. However, observe that the result is possibly global in time. In fact, for the time being there is no result going in the sense that starting with an invertible matrix $\mathbf{\Gamma}[C^0]$, a loss of rank possibly occurs. We refer the reader to [13, 14, 31, 21] for remarks on the rank of the first order density operators, that is the rank of the matrix $\mathbf{\Gamma}[C]$. The proof is postponed until [12] and follows the main lines of [15]. That is we use semi-groupe and contraction arguments for *local in time*, say up to a certain $\tau > 0$ existence and uniqueness. Following [22], we start with a density matrix $\mathbf{\Gamma}[C^0]$ of full rank, that is K, thus, this property will be propagated by the dynamics of $\mathscr{S}_{\mathrm{MCHF}}$ at least for an infinitesimal time $T^\star$. After-what, we extend the solution behind τ thanks to some *a priori* estimates, more precisely mass and energy conservation. We will present two proofs fro the energy conservation, the difference is slight. The first one, presented in the second Chapter, is explicit and obtained by tackling directly the MCTDHF equations of motion. The second way, see [12], is by proving that these equations satisfy the VP, thus conserve the total energy. In particular, we prove that if the couple $(C(t),\Phi(t))$ satisfies $\mathscr{S}_{\mathrm{MCHF}}$, then it satisfies obviously the Dirac-Frenkel VP. This well-posedness result with only Coulomb intercation were announced in [29].

Moreover, we were able to propagate the regularity of the solution as follows

Corollary 1.1. *Let* $U(x), V(|x|) \in L^d(\mathbb{R}^3,\mathbb{R})$ *with* $d > \frac{3}{2}$ *and* $(C^0,\Phi^0) \in \partial\mathscr{F}_{N,K} \cap H^s(\mathbb{R}^3,\mathbb{C})^K$ *with* $s \geq 1$. *Then, there exists a maximal existence time* $T^\star > 0$ *(possibly* $+\infty$*) such that the MCTDHF system* $\mathscr{S}_{\mathrm{MCHF}}$ *admits a unique solution* (C,Φ) *with* $C \in C^1\big([0,T^\star);\mathbb{C}\big)^r$ *and* $\Phi \in C\big([0,T^\star);H^s(\mathbb{R}^3,\mathbb{C})\big)^K$ *that depends continuously on the initial data* (C^0,Φ^0). *Morever the solution satisfies* $ii-iv)$ *of Theorem 4.1.*

The proof is easy, in fact one prove that the non-linearities are still locally Lipschitz from $\mathbb{C}^r \times H^s(\mathbb{R}^3,\mathbb{C})$ into itself and get the necessary *a priori* estimate on the $H^s(\mathbb{R}^3,\mathbb{C})^K$ by induction starting from the one of $H^1\big(\mathbb{R}^3\big)$.

The Theorem 4.1 being only possibly global when starting with *regular* initial data. However the result is certainly global if one add an extra assumption on the energy of the initial data. Such assumption is shown to ensure that the matrix $\mathbf{\Gamma}[C(t)]$ remains of rank K during the time evolution. In order to state this result, we introduce

$$\mathscr{E}(C,\Phi) = \langle\, \Psi(C,\Phi)\,|\mathscr{H}|\,\Psi(C,\Phi)\rangle_{L^2(\mathbb{R}^{3N})},$$

$$\mathscr{E}(K) = \min_{(C,\Phi)\in\mathscr{F}_{N,K}} \mathscr{E}(C,\Phi).$$

Now, we have

Theorem 1.5. *Let* $U(x), V(|x|) \in L^d(\mathbb{R}^3,\mathbb{R})$ *with* $d > \frac{3}{2}$ *and* $(C^0,\Phi^0) \in \mathscr{F}_{N,K} \cap H^1\big(\mathbb{R}^3\big)^K$ *such that*

$$\mathscr{E}(C^0,\Phi^0) < \mathscr{E}(K-1).$$

Then, the Cauchy problem $\mathscr{S}_{\mathrm{MCHF}}$ *has a unique global-in-time solution* $(C(t),\Phi(t))$ *satisfying*

- $C \in C^1\big([0,+\infty);\mathbb{C}\big)^r$ *and* $\Phi \in C\big([0,+\infty);H^1\big(\mathbb{R}^3\big)\big)^K$ *that depends continuously on the initial data* (C^0,Φ^0).

ii) For every $t>0$, $\big(C(t),\Phi(t)\big) \in \partial\mathscr{F}_{N,K}$.

iii) For every $t>0$,

$$\mathscr{E}(C(t),\Phi(t)) = \mathscr{E}(C^0,\Phi^0).$$

This provides with the global-in-time well-posedness to the MCTDHF approximation under a suitably assumption on the initial data. The proof is based on a contradiction argument. Obviously, the propagation of the regularity in this global set up holds.

Following [30, 11], we were able to set up an L^2 theory to the MCTDHF. Note, however, that in our case we do not consider infinitely many "mixed states" as they do, but consider the pure states of N particles, a finite set of *orbitals*, that is with K and N fixed. The result is the following

Theorem 1.6. *Let* $U(x),V(|x|) \in L^d(\mathbb{R}^3,\mathbb{R})$ *with* $d>\frac{3}{2}$ *and* $(C^0,\Phi^0) \in \partial\mathscr{F}_{N,K}$ *be an initial data. Then, there exists a time* $T^\star>0$ *(possibly* $=+\infty$*) such that the MCTDHF system* $\mathscr{S}_{\text{MCHF}}$ *admits solutions* $(C(t),\Phi(t))$ *satisfying*

- $C \in C^1([0,T^\star),\mathbb{C})^r \quad$ *and* $\quad \Phi \in C^0([0,T^\star),L^2(\mathbb{R}^3))^K$.

Moreover,

i) $\Phi(t) \in L^{\frac{4}{3}\frac{q}{q-2}}([0,T],L^q(\mathbb{R}^3))^K$ *for all* $2\le q<6$ *and* $0\le T<T^\star$.

ii) *The solution* $(C(t),\Phi(t))$ *is unique in the class*

$$L^\infty([0,T^\star),\mathbb{C})^r \times L^\infty([0,T],L^2(\mathbb{R}^3))^K \cap L^{\frac{4}{3}\frac{q}{q-2}}([0,T],L^q(\mathbb{R}^3))^K,$$

for all $2\le q<6$ *and* $T<T^\star$.

- *Also,* $(C(t),\Phi(t)) \in \partial\mathscr{F}_{N,K}$ *for all* $t\in[0,T^\star)$.

Observe that as in the case of the Theorem 4.1, the result is possibly global. However, in this case the assumption on the initial data of the Theorem 1.5 cannot be used. This is due to the fact that we are dealing with system with infinite energy.

In practical calculations, physicists and chemists sometimes add a small perturbation to the matrix $\mathbf{\Gamma}[C]$ in order to ensure its invertiblity. The nature of the perturbation is, in general, chosen with respect to the numerical scheme used and we refer the reader for instance to [6] in the framework of the MCTDH which is the adequate model for bosons and will be presented below.

Without loss of generality, we replace $\mathbf{\Gamma}[C]$ in $\mathscr{S}_{\text{MCHF}}$ by $\mathbf{\Gamma}[C]+\varepsilon\, I_K$ where I_K denotes the $K\times K$ identity matrix. The system becomes then

$$\mathscr{S}^{\varepsilon}_{\text{MCHF}}:\quad \begin{cases} i\frac{d}{dt}\,C(t)=\mathscr{M}[\Phi](t)\,C(t),\\ \\ i\,\frac{\partial}{\partial t}\Phi(t,x)=\mathbf{H}_x\,\Phi(t,x)+[\mathbf{\Gamma}[C]+\varepsilon\, I_K]^{-1}\;(I-\mathbf{P}_\Phi)\,\mathbb{W}[C,\Phi](t,x)\,\Phi(t,x).\\ \\ (C(t=0),\Phi(t=0))=(C^0,\Phi^0)\in\mathscr{F}_{N,K}. \end{cases}$$

This system conserves the orthogonality of the orbitals and the normalization of the coefficients. This is due to the fact that $\mathbf{\Gamma}[C(t)]+\varepsilon\, I_K$ is obviously invertible and Hermitian for all time and its inverse permutes with the projector $(I-\mathbf{P}_\Phi)$. A straightforward corollary of the Theorem 4.2 provides with a global L^2 theory for such system.

Notice that if one set in the MCTDHF $K=N$, then $|\Sigma_{N,N}|=1$ and Ψ_{MC} turns to be a single Slater determinant multiplied by a phase factor. It is also easy to see that in this configuration $\mathbf{\Gamma}[C]$ coincides with I_N. Thus $T^\star=+\infty$ in Theorem 4.1. This improves the result of Chadam and Glassey in [15]. In the fifth chapter, we show how one can obtain the TDHF system from the MCTDHF one by setting $N=K$ and performing a Gauge transform. Also, $T^\star=+\infty$ in Theorem 4.2 and our result is then compatible with the one of Zagatti in [30]. However we recall that Zagatti consider a system of infinite orbitals and the model he is studying cannot be really called TDHF. Moreover, Castella proved in [11] the same type of result on the Schrödinger poisson model.

1.3.2 The multiconfiguration time-dependent Hartree method

The multiconfiguration time-dependent Hartree (MCTDH in short) is the approximation to use when dealing with particles with integer spin, that is bosons. >From a purely mathematical point of view, MCTDH has only an algebraic difference compared to MCTDHF. In this section, we shall briefly present the MCTDH equations of motion in order to observe the similarity of the structure and be convinced that the results we obtained on MCTDHF allows *mutatis mutandis* also to conclude in the MCTDH framework.

The MCH approximates the wavefunction as follows Ψ by a linear combinations of *Hartree products* , that is

$$\Psi_{\text{MCH}}(x_1,\dots,x_N)=\sum_{i_1,\dots,i_N=1}^{K}\beta_{i_1,\dots,i_N}\,\phi_{i_1}(x_1)\dots\phi_{i_N}(x_N) \tag{1.28}$$

where the β_I's are complex coefficients such that $\sum_{i_1,\dots,i_N=1}^{K}|\beta_{i_1,\dots,i_N}|^2=1$ and $\{\phi_k\}_{1\le k\le K}$ is an orthonormal family in $L^2(\mathbb{R}^3)$. The subscript MCH stands for MC Hartree. Note that the above ansatz is the general version of the ansatz for bose condensates, where only one wavefunction is used.
The loss of the skew-symmetry will complicates only the algebraic aspect of the associated equations. The system of equations associated to the MCTDH is the following

$$\mathscr{S}_{\text{MCH}}:\quad \begin{cases} i\,\frac{d}{dt}\beta_{i_1,\dots,i_N}(t)=\sum_{i_1,\dots,i_N=1}^{K}\langle V\,\prod_{k=1}^{N}\phi_{j_k},\prod_{l=1}^{N}\phi_{i_l}\rangle_{L^2(\mathbf{R}^{3N})}\,\beta_{j_1,\dots,j_N}(t),\\ \\ i\mathbf{\Gamma}^k(t)\,\frac{\partial}{\partial t}\Phi(t,x_k)=\mathbf{\Gamma}(t)^k\,\mathbf{H}_{x_k}\,\Phi(t,x_k)+\,(I-\mathbf{P}_\Phi)\,\mathbb{W}^k_{\beta,\Phi(x_k)}\,\Phi(t,x_k), \end{cases}$$

where $\mathbf{H}_{x_k} = -\Delta_{x_k} + \mathscr{V}(x_k)$ and

$$\mathbf{\Gamma}^k_{i,j}(t) = \sum_{i_1,\ldots,i_{k-1},i_{k+1},\ldots,i_N=1}^{K} \bar{\beta}_{i_1,\ldots,i_{k-1},i,i_{k+1},\ldots,i_N} \beta_{i_1,\ldots,i_{k-1},j,i_{k+1},\ldots,i_N}.$$

Observe that, in contrast to the MCTDHF, here we have N density matrices, each one is associated to a space degree of freedom and we refer the reader to [6, 23] where the MCTDH equations were "derived" first for numerical simulation end. The entries of the matrix $\mathbb{W}^k_{\beta,\Phi(x,k)}$ are defined as follow

$$(\mathbb{W}^k_{\beta,\Phi(x_k)})_{j_k,i_k} = \sum_{n=1,n\neq k}^{N} \sum_{i_1,\ldots,i_{k-1},i_{k+1},\ldots,i_N,j_n} \bar{\beta}_{i_1,\ldots,i_{n-1},j_n,i_{n+1},\ldots,i_{k-1},j_k,i_{k+1},\ldots,i_N} \beta_{i_1,\ldots,i_N} \left(\phi_{i_n}\bar{\phi}_{j_n} \star \frac{1}{r}\right)(t,x_k)$$

where for the notation we assumed that $n<k$ and Φ denotes the $K-$component vector $(\phi_1,\ldots,\phi_K)^T$. Finally the $\mathbf{P}_\Phi$ is again the projector on the space spanned by the $\phi_i's$. In order to observe the similarity with the MCTDHF, let us write ΨMCHF as follows

$$\Psi_{\text{MCHF}}(t,X_N) = \sum_{i_1,\ldots,i_N=1}^{K} \alpha_{i_1,\ldots,i_N}(t)\,\phi_{i_1}(t,x_1)\ldots\phi_{i_1}(t,x_N),$$

where we used the convention introduced by Lewin in [31]

$$\alpha_{i_1,\ldots,i_N} := \begin{cases} 0 & if \quad |\{i_1,\ldots,i_N\}| < N \\ \frac{(-1)^\varepsilon}{\sqrt{N!}}\, c_\sigma, & with \quad \sigma = \varepsilon(\{i_1,\ldots,i_N\}). \end{cases}$$

The coefficients $\alpha_{\ldots}$ are then clearly skew-symmetric with respect to their indexes. Using these coefficients, we have

$$\mathbf{\Gamma}[C]_{i,j}(t) = N \sum_{i_2,\ldots,i_N=1}^{K} \overline{\alpha}_{i,i_2,\ldots,i_N}(t)\,\alpha_{j,i_2,\ldots,i_N}(t),$$

$$\mathbb{W}[C,\Phi](t,x) = N(N-1) \sum_{i_3,\ldots,i_N=1}^{K} \sum_{p,q=1}^{K} \overline{\alpha}_{i,p,i_3,\ldots,i_N}(t)\,\alpha_{j,q,i_2,\ldots,i_N}(t) \times \\ \times \int_{\mathbb{R}^3} \overline{\phi}_p(t,y)\,V(|x-y|)\,\phi_q(t,y)\,dy.$$

Now, recall the TDH approximation to the Schrödinger equation and observe that this method can be seen as a limiting case of the MCTDH. In fact, Let $K=1$ in the *ansatz* (1.28), then the wavefunction turn to be a Hartree product multiplied by a phase factor. Again, we refer the reader to the fifth chapter for more detail on how one get the TDH equation as a limiting case of the MCTDH. In particular, this tell us that our results are valid for the TDH. Moreover, as in the TDHF case, our Theorems are global-in-time since in this configuration the matrix $\mathbf{\Gamma}$ turns to be a scalar.

1.4 Simulation

The multiconfiguration methods are used with success for numerical simulations and we refer the reader to [6, 7] and references therein. The last chapter of the present manuscript is dedicated to numerical simulation of a "Toy model" of the MCTDHF, we consider a system composed of 2 particles, thus an even number of orbitals K. This calculation is based on a standard scheme that suffer from a non-conservation of the the normalization constraints and the total energy from the numerical analysis point of view. However, in some cases, the energy seems to be quasi-conserved but not in other, in particular when the effort of the computation becomes important. To our knowledge, there is no publication concerning the implementation of scheme conserving these relevant quantities. We investigated recently this point and the development of such scheme being in advance.

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Remark

The first part of the next Chapter consists in an appeared note in ”Comptes rendus de l’Académie des sciences“

Trabelsi, S.: Solutions of the Multiconfiguration Time-Dependent Equations in Quantum Chemistry. C. R. Acad. Sci. Paris, Ser. I 345 (2007).

The second part present explicit calculation on which the result of the note is based. We warn the reader that the notation of this chapter is not the same as in the next chapters for fidelity reason to the appeared text of the note.

Chapter 2

Local-in-time existence of solutions to the MCTDHF

2.1 CRAS Note

Abstract

The multiconfiguration methods are a natural generalization of well-known simple models for approximating the linear N body Schrödinger equation for atomic and molecular systems with binary (Coulomb) interactions, like the Hartree and the Hartree–Fock theories. This paper discusses the case of the multiconfiguration time-dependent Hartree–Fock (MCTDHF in short) method which consists in approximating the high-dimensional wavefunction by a time-dependent linear combination of Slater determinants. We formulate the system of equations of motion and we establish the well-posedness of this system in a convenient Hilbert space framework, at least as long as the associated one-particle density matrix keeps the same rank. Our proof covers and simplifies previous well-posedness results of the Cauchy problems associated to the time-dependent Hartree and the time-dependent Hartree-Fock approximations obtained in [3, 4, 6, 7].

Solutions des équations de multi-configurations dépendant du temps en Chimie Quantique.

Résumé

Les méthodes de multi-configurations améliorent des modèles simples d'approximation bien connus de l'équation de Schrödinger linéaire à N corps pour les systèmes moléculaires sous interactions Coulombiennes, tels que les modèles de Hartree et de Hartree–Fock. Dans cette Note, nous étudions le cas de la méthode dite de Multi-Configurations Hartree–Fock dépendante du temps, qui consiste à approcher les fonctions d'onde antisymétriques d'un espace de Hilbert de dimension infinie par une combinaison linéaire dépendante du temps de déterminants de Slater. Nous écrivons le système d'équations d'évolution et nous établissons que ce système est bien posé dans un cadre fonctionnel adéquat, et ceci tant que la matrice densité associée ne change pas de rang. Notre preuve recouvre et simplifie les résultats d'existence et unicité de solutions des problèmes de Cauchy associés aux approximations de Hartree et de Hartree-Fock obtenus dans [3, 4, 6, 7].

Version française abrégée

Cette Note présente un résultat d'existence et d'unicité d'une solution classique pour le problème de Cauchy associé aux équations de multiconfigurations Hartree-Fock dépendante du temps. Elles

fournissent une approximation de l'équation linéaire de Schrödinger à N particules (2.1) (voir la version anglaise). Le Hamiltonien H_N est composé, dans l'ordre, d'un terme d'énergie cinétique, d'un terme d'interaction noyau-électron $V(x) = -\sum_{1\le i\le M} \frac{z_i}{|x-\bar{x}_i|}$ où on a considéré M noyaux fixés aux points $\bar{x}_1,\dots,\bar{x}_M$, enfin d'un terme d'interaction Coulombienne électron-électron. La fonction $\Psi = \Psi(x_1,\dots,x_N) \in \mathscr{H} = L^2(\mathbb{R}^{3N};\mathbb{C})$ est appelée fonction d'onde. Une contrainte d'antisymétrie lui est imposée pour satisfaire au principe d'exclusion de Pauli pour les fermions. L'exemple le plus simple de telles fonctions est le déterminant de Slater

$$\Psi(x_1,\dots,x_N) = \frac{1}{\sqrt{N!}} \det(\phi_i(x_j))_{1\le i,j\le N}$$

où les N fonctions ϕ_i forment une famille orthonormale de $L^2(\mathbb{R}^3,\mathbb{C})$. Le facteur $\frac{1}{\sqrt{N!}}$ est introduit pour normaliser Ψ dans $\mathscr{H}$. La restriction à un seul déterminant de Slater correspond à l'approximation bien connue de Hartree-Fock. L'approximation multiconfigurations Hartree-Fock consiste à considérer une combinaison linéaire finie de tels déterminants. Pour $N \le K$, on définit l'ensemble $\mathscr{A}_N^K = \left\{I = \{i_1 < \ldots < i_N\} \subset \{1,\dots,K\} : |I| = N \le K\right\}$ de cardinal $\binom{K}{N}$, puis l'espace $\mathscr{M}_N^K$ comme dans (2.2). La fonction d'onde est alors formée de $\binom{K}{N}$ déterminants, et décrite par $\binom{K}{N}$ coefficients $C_I(t)$ et K fonctions "mono-électroniques" $\phi_i(t,x)$. Les équations qui régissent le mouvement du système sont données par (6.3a-2.4c). Elles satisfont le principe variationnel (2.5) et s'expriment comme un système couplé de $\binom{K}{N}$ équations différentielles d'ordre 1 pour les coefficients et de K équations aux dérivées partielles non-linéaires pour les fonctions de base. En multipliant (6.3a) par $\bar{C}_I$ et en sommant, on obtient que $\sum_{I\in\mathscr{A}_N^K} |C_I(t)|^2 = 1$ comme conséquence directe du fait que $\mathbf{W}$ est réel. Dans cette note ainsi que dans la publication [1], on s'intérresse au système d'équations (6.3a-2.4c). On démontre l'existence et l'unicité des solutions qui, en particulier, conservent l'énergie totale. Ceci est prouvé directement à partir des équations d'évolution, sans faire appel à un principe variationnel. Techniquement, le Théorème 5.3 est démontré, dans un premier temps, dans un voisinage de (C^0,Φ^0) en supposant que $\mathbf{\Gamma}^0$ est inversible, et en utilisant un argument de point fixe de Picard. Les estimations *a priori* nous permettent de prolonger le résultat jusqu'à un certain temps $T^\star$ au delà duquel la matrice densité peut éventuellement dégénèrer.

2.1.1 General setting and the MCTDHF equations

The aim of this work is to present some results concerning the multiconfiguration (MC) Time-Dependent (TD) Hartree-Fock (HF) equations (=MCTDHF). These methods are used in quantum physics/chemistry to approximate the solutions of the time-dependent N particle linear Schrödinger equation with binary interactions. Using the appropriate scaling ("atomic units" with electron mass equal to 2 etc.) this equation reads

$$i\partial_t\Psi = H_N\Psi := \left(\sum_{1\le i\le N} (-\Delta_{x_i} + V(x_i)) + \sum_{1\le i<j\le N} \frac{1}{|x_i - x_j|}\right)\Psi, \tag{2.1}$$

where $\Psi = \Psi(x_1,\dots,x_N) \in \mathscr{H} = L^2(\mathbb{R}^{3N};\mathbb{C})$ is the so-called wavefunction which we normalize to one in $\mathscr{H}$ for $|\Psi|^2$ is interpreted as the probability density of the N electrons. The N-body Hamiltonian of the system is then the self-adjoint operator H_N acting on the Hilbert space $\mathscr{X}$. The subscript x_i of $-\Delta_{x_i}$ means derivation with respect to the i^{th} variable of the function Ψ, $V(x) =$

$-\sum_{1\le i\le M}\frac{z_i}{|x-\bar{x}_i|}$ is the Coulomb potential created by the nuclei localized at $\bar{x}_i$ with charge $z_i > 0$, and the last term of H_N is the Coulomb repulsive potential between the electrons. Finally ∂_t denotes the partial time derivative. When dealing with electrons (fermions in general), the wavefunction Ψ has to satisfy $\Psi(x_1,\dots,x_N)=\varepsilon(\sigma)\Psi(x_\sigma(1),\dots,x_\sigma(N))$ for all permutation $\sigma\in S_N$ with $\varepsilon(\sigma)$ being its parity. The simplest skew-symmetric elements of $\mathscr{H}$ are the so-called *Slater determinants* $,\Psi(x_1,\dots,x_N)=\frac{1}{\sqrt{N!}}\det(\phi_i(x_j))_{1\le i,j\le N}$ with the ϕ_i's being an orthonormal family of $L^2(\mathbb{R}^3,\mathbb{C})$. The basic idea of the MC method is then to approximate the space of skew-symmetric wavefunctions by subsets of *finite* linear combinations of Slater determinants. More precisely we introduce

$$\mathscr{M}_N^K:=\left\{\Psi\,:\,\Psi=\sum_{I\in\mathscr{A}_N^K}\frac{C_I}{\sqrt{N!}}\,Det_\Phi(I),\ (C,\Phi)\in\mathscr{S}_N^K\right\} \tag{2.2}$$

where $\mathscr{A}_N^K=\big\{I=\{i_1<\ldots<i_N\}\subset\{1,\dots,K\}\,:\,|I|=N\le K\big\}$; that is, Ψ is a linear combination of $|\mathscr{A}_N^K|=\binom{K}{N}$ determinants $Det_\Phi(I)$ built from the (ϕ_{i_k}) for all $i_k\in I$. $\mathscr{S}_N^K$ is the "unit sphere" of the $\binom{K}{N}$ vectors of coefficients (C_I) and the K vectors of functions $(\phi_1,\cdots,\phi_K)^T:=\Phi$, that is

$$\mathscr{S}_N^K:=\left\{(C,\Phi)\in\mathbb{C}^{\binom{K}{N}}\times L^2(\mathbb{R}^3,\mathbb{C})^K,\ \sum_{I\in\mathscr{A}_N^K}|C_I|^2=1,\ \int_{\mathbb{R}^3}\phi_i\,\bar{\phi}_j\,dx=\delta_{i,j}\right\} \tag{2.3}$$

where the bar stands for the complex conjugate. Notice that an intrinsic definition of $\mathscr{M}_N^K$ can be given in terms of the so-called *one-particle density matrix* as follows. To every wavefunction Ψ in $\mathscr{H}$ one can associate a trace-class self-adjoint operator Γ_Ψ acting on $L^2(\mathbb{R}^3;\mathbb{C})$ with kernel $\gamma_\Psi(x,y)=N\int_{\mathbb{R}^{3(N-1)}}\Psi(x,x_2,\dots,x_N)\overline{\Psi}(y,x_2,\dots,x_N)\,dx_2\dots dx_N$. Density matrices of *finite* rank are the ones that precisely correspond to wavefunctions Ψ which can be written as finite linear combinations of Slater determinants. This result is due to Löwdin's expansion theorem [11]. We may now define the increasing sequence of subsets

$$\mathscr{M}_N^K=\left\{\Psi\in\bigwedge_{i=1}^N L^2(\mathbb{R}^3)\,:\,\|\Psi\|_{L^2(\mathbb{R}^{3N})}=1,\,\mathrm{rank}(\Gamma_\Psi)\le K\right\},$$

where $K\ge N$. Then in definitions (2.2) and (2.3), K corresponds to the rank of (Γ_Ψ) and the family $\phi_1,\dots,\phi_K$ to an orthonormal basis of the range, $\mathrm{Ran}(\Gamma_\Psi)$. When $K=N$, we have $|\mathscr{A}_N^N|=1$. Thus Ψ reduces to a single *Slater determinant* and Γ_Ψ is the projector of finite rank N on $\mathrm{Span}\{\phi_i\}_{1\le i\le K}$. This precisely corresponds to the Hartree-Fock (HF) approximation. The corresponding Cauchy problem was investigated first in [3] for bounded interactions. The Coulomb case was solved first in [7] and improved in [6, 4, 5]. On the other side, when $K=+\infty$ we recover, at least formally, the full space $\bigwedge_{i=1}^N L^2(\mathbb{R}^3)$.
From now on we consider time-dependent coefficients $C_I(t)$ and time-dependent functions $\phi_i(t,x)$.

The MCTDHF equations constitute a system of coupled ODEs and non-linear PDEs as follows

$$i\frac{d}{dt}C_I(t) = \sum_{J\in\mathscr{A}_N^K} \langle \mathbf{W}Det_\Phi(J), Det_\Phi(I)\rangle\, C_J(t) \qquad \forall I\in\mathscr{A}_N^K, \tag{2.4a}$$

$$i\mathbf{\Gamma}(t)\,\partial_t\Phi(t) = \mathbf{\Gamma}(t)\,\mathbf{H}\,\Phi(t) + (I-\mathbf{P}_\Phi)\mathbb{W}(C,\Phi)\,\Phi(t) \tag{2.4b}$$

$$C(0) = C^0, \quad \Phi(0) = \Phi^0. \tag{2.4c}$$

Equations (6.3a) form a system of $\binom{K}{N}$ first-order ordinary differential equations where

$$\langle \mathbf{W}Det_\Phi(J), Det_\Phi(I)\rangle = \sum_{1\le i<j\le N}\int_{\mathbb{R}^{3N}} \frac{Det_\Phi(J)(t,x_1,\dots,x_N)\,\overline{Det_\Phi(I)}(t,x_1,\dots,x_N)}{|x_i-x_j|}\,dx_1\dots dx_N$$

denotes the Coulomb interactions between the Slater determinants $Det_\Phi(I)$ and $Det_\Phi(J)$. Equation (6.3b) provides a system of K PDEs on the ϕ_i's, where $\mathbf{H} = -\Delta + V$, and $\mathbf{P}_\Phi$ denotes the orthogonal projector onto $\mathrm{Span}\{\phi_i\}_{1\le i\le K}$, $\mathbf{P}_\Phi = \sum_{i=1}^K \langle\cdot,\phi_i\rangle_{L^2(\mathbb{R}^3,\mathbb{C})}\phi_i$. The $K\times K$ Hermitian matrices $\mathbf{\Gamma}(t)$ and $\mathbb{W}(C,\Phi)(t,x)$ are given by

$$\mathbf{\Gamma}_{i,j}(t) = \sum_{\substack{I,J\in\mathscr{A}_N^K\setminus i\in I, j\in J\\ I\setminus\{i\}=J\setminus\{j\}}} (-1)^{\varsigma(i,I)+\varsigma(j,J)}\overline{C}_I C_J, \quad \text{with} \quad \varsigma(l,I)=k \quad \text{when} \quad i_k = l,$$

$$(\mathbb{W}_{C,\Phi})_{ij}(t,x) = 2\sum_{k,l=1}^K \sum_{\substack{I,J\in\mathscr{A}_N^K \text{ s.t.}\{i,k\}\in I,\{j,l\}\in J\\ I\setminus\{i,k\}=J\setminus\{j,l\}, i\neq k, j\neq l}} (-1)^{\varsigma'(i,k,I)+\varsigma'(j,l,I)}\bar{C}_I C_J\left[(\overline{\phi}_k\,\phi_l) * \frac{1}{|x|}\right]$$

with the convention $\varsigma'(i,k,I) = p+q$ if $i = i_p < k = i_q$ and $\varsigma'(i,k,I) = p+q+1$ if $k = i_q < i = i_p$. Notice that $\mathbf{\Gamma}$ is the representation of the density matrix $\mathbf{\Gamma}_\Psi$ in $\mathrm{Span}\{\phi_i\}_{1\le i\le K}$. Conversely, any "regular" solution of the Cauchy problem (6.3a)-(2.4c) satisfies the so-called Dirac-Frenkel variational principle, more precisely

$$\langle \delta\Psi, (i\partial_t - H_N)\Psi\rangle = 0, \quad \forall\delta\Psi\in\mathscr{T}_\Psi\mathscr{M}_N^K, \tag{2.5}$$

where $\mathscr{T}_\Psi\mathscr{M}_N^K$ denotes the tangent space of $\mathscr{M}_N^K$ at Ψ. However, in [1], we work directly on the equations, without recours to a variational principle.

A crucial question is the following : does the time evolution by the MCTDHF equations conserve the rank of the initial density matrix or might a loss of rank occur after a time $T^\star > 0$? For the time being, there is no definite answer to this question (see however the paper of Lewin [10] and Friesecke [8] for such questions in the stationary case) and, like in [9], we can only state a local in time result with a non-degeneracy assumption.

2.1.2 Main result and sketch of the proof

We first state an existence and uniqueness result on the system of equations (6.3a)-(2.4c) under some assumption on the initial data which ensures that the density matrix $\mathbf{\Gamma}(t)$ does not become

degenerate.

Theorem 2.1. *Given $(C^0,\Phi^0)\in\mathscr{S}_N^K$ (as defined in (2.3)) such that $\mathbf{\Gamma}_{\Phi^0}$ is invertible, that is* $\mathrm{rank}(\mathbf{\Gamma}_{\Phi^0})=K$. *Assume in addition that $\Phi^0\in(H^1(\mathbb{R}^3,\mathbb{C}))^K$. Then, there exists $T^\star>0$ such that the system (6.3a)–(2.4c) admits a unique solution (C,Φ) in $\mathscr{S}_N^K$ for all $t\in[0,T^\star)$ which satisfies*

$$(C,\Phi)\in\mathscr{X}_{T^\star}:=C^1\big([0,T^\star);\mathbb{C}^{\binom{N}{K}}\big)\times\Big(C^1\big([0,T^\star);H^{-1}(\mathbb{R}^3,\mathbb{C}))^K\big)\cap C^0\big([0,T^\star);H^1(\mathbb{R}^3,\mathbb{C})^K\big)\Big).$$

In addition, for every $0\le t<T^\star$,

$$\mathrm{rank}(\mathbf{\Gamma}_{\Phi(t)})=\mathrm{rank}(\mathbf{\Gamma}_{\Phi^0}),\quad \textit{more precisely}\quad T^\star=\inf\{t>0\ :\ \mathrm{rank}(\mathbf{\Gamma}_{\Phi(t)})\le K-1\}.$$

Sketch of proof (see [1] for details): Observe that in a neighborhood of (C^0,Φ^0), the application $\mathbf{\Gamma}\mapsto\mathbf{\Gamma}^{-1}$ is well-defined since we assume $\mathbf{\Gamma}^0$ invertible. By Cauchy-Schwarz and Hardy inequalities, the non-linear operator

$$\Omega:(C,\Phi)\mapsto\Big((\langle\mathbf{W}Det_\Phi(J),Det_\Phi(I)\rangle)_{I,J}C,\mathbf{\Gamma}^{-1}(I-\mathbf{P}_\Phi)\mathbb{W}(C,\Phi)\Phi\Big)$$

is locally Lipschitz in $l^2(\mathbb{C})^{\binom{K}{N}}\times(H^1(\mathbb{R}^3,\mathbb{C}))^K$. Then, the local well-posedness is obtained from the integral formulation of the system by a Picard fixed point argument. Then, we prove that this solution satisfies the system (6.3a)-(2.4c) in a strong sense and is unique by Gronwall Lemma in the class $\mathscr{X}_\tau$ for $\tau>0$ small enough. Finally, the existence and uniqueness of solution up to the time $T^\star:=\inf\{t>0\ :\ \mathrm{rank}(\mathbf{\Gamma}_{\Phi(t)})\le K-1\}$ in the class $\mathscr{X}_{T^\star}$ is equivalent to the existence of uniform in time estimates which allow us to extend the solution beyond the time τ, [12]. The estimates $\sum_{I\subset\mathscr{A}_N^K}|C_I(t)|^2=1$ and $\|\Phi\|_{(L^2(\mathbb{R}^3,\mathbb{C}))^K}=1$ are directly verified respectively from (6.3a) and (6.3b) using the self-adjointness of $\mathbf{W},\mathbf{H}$ and $I-\mathbf{P}_\Phi$. Further necessary estimate on Φ in $(H^1(\mathbb{R}^3))^K$ arises from the following

Theorem 2.2. *Given $(C^0,\Phi^0)\in\mathscr{M}_N^K$ as in Theorem 5.3. Then, for any solution in the class $\mathscr{X}_\tau$ for τ small enough, one has*

$$\mathscr{E}_N^K(t)\ :=\ \langle\mathbf{H\Gamma}(t)\Phi(t),\Phi(t)\rangle_{L^2(\mathbb{R}^{3N})}+\langle\mathbb{W}(C,\Phi)\Phi(t),\Phi(t)\rangle_{L^2(\mathbb{R}^{3N})}=\mathscr{E}_N^K(0)\qquad(2.6)$$

Moreover, there exist positive constants κ and $\tilde\gamma$ such that

$$\tilde\gamma\|\nabla\Phi\|^2_{(L^2(\mathbb{R}^3,\mathbb{C}))^K}\le\kappa\mathscr{E}_N^K(0).$$

The first part of the equality (2.6) is rather a simple computation (see also [10]). Since the solution satisfies the variational principle (2.5), setting $\delta\Psi=\partial_t\Psi\in\mathscr{T}_\Psi\mathscr{M}_N^K$ leads immediately to $\Re\Big(\langle\partial_t\Psi,H\Psi\rangle_{L^2(\mathbb{R}^{3N})}\Big)=0$ where $\Re$ denotes the real part, hence $\mathscr{E}_N^K(t)=\mathscr{E}_N^K(0)$. Notice that setting $K=N$ in this proof leads to the conservation of the energy of the Hartree-Fock system. Finally,

$$\begin{aligned}\tilde\gamma\|\sqrt{\mathbf{H}}\Phi\|^2_{(L^2(\mathbb{R}^3,\mathbb{C}))^K}&\le\|\sqrt{\mathbf{H\Gamma}}\Phi\|^2_{(L^2(\mathbb{R}^3,\mathbb{C}))^K}\\&\le\langle\mathbf{H\Gamma}(t)\Phi(t),\Phi(t)\rangle_{L^2(\mathbb{R}^{3N})}+\langle\mathbb{W}(C,\Phi)\Phi(t),\Phi(t)\rangle_{L^2(\mathbb{R}^{3N})}\\&=\mathscr{E}_N^K(0)\end{aligned}$$

The equivalence of the norms $\|\sqrt{\mathbf{H}}\Phi\|_{(L^2(\mathbb{R}^3,\mathbb{C}))^K}$ and $\|\Phi\|_{(H^1(\mathbb{R}^3,\mathbb{C}))^K}$ allows to conclude the proof. Since $\mathbf{\Gamma}(t)$ is invertible for $t \in [0,T^\star)$, we are able to bound $\tilde{\gamma}(t) := \inf_{|\zeta|^2=1} \langle \mathbf{\Gamma}(t)\zeta,\zeta\rangle$ for all $\zeta \in \mathbb{C}^K$ from below for $t \in [0,T^\star)$. This leads to the desired estimate and proves Theorem 5.3.

In [1], we prove that one can iterate on the H^1 bound and obtain higher regularity for the MCTDHF solution following the regularity of the initial data. We also show how to take advantage from the gauge invariance of the MCTDHF system in order to obtain the Hartree-Fock system and its energy conservation as limiting case only by setting $N = K$. Moreover in this case the matrix $\mathbf{\Gamma}$ becomes the $N \times N$ identity matrix, hence $T^\star = +\infty$. A corollary yielding the global well-posedness of the Hartree-Fock system follows immediately, considerably simplifying the proofs of [6, 7]. In this Note we presented our result on the MCTDHF for brevity, but our method and result apply also to the MCTDH (the Multiconfigurations Time-Dependent Hartree) system which is the appropriate model for bosons, hence to its limiting case, namely the time-dependent Hartree approximation. In order to emphasize the algebraic similarity between the two models, let us write the MCTDHF wavefunction as follows $\Psi(x_1,\dots,x_N) = \sum_{1\le i_1,\dots,i_N\le K} \alpha_{i_1,\dots,i_N}\phi_{i_1}(x_1)\cdots\phi_{i_N}(x_N)$ where we used the convention introduced by Lewin [10], $\alpha_{i_1,\dots,i_N} = \frac{\varepsilon(\sigma)}{\sqrt{N!}} C_{\{i_{\sigma(1)}<\cdots<i_{\sigma(N)}\}}$ with $\sigma \in S_N$ such that $i_{\sigma(1)} < \cdots < i_{\sigma(N)}$. The coefficients $\alpha_{i_1,\dots,i_N}$ are then skew-symmetric with respect to permutations of their indexes. Now, the MCTDH wavefunction is written as a linear combination of Hartree products, that is $\Psi(x_1,\dots,x_N) = \sum_{i_1,\dots,i_N=1}^K \beta_{i_1,\dots,i_N}\prod_{k=1}^N \phi_{i_k}(x_k)$ where the β_I's are complex coefficients. The only difference then is the skew-symmetry of the combination coefficients. The MCTDH equations of motion are similar to MCTDHF but only more complicated from the algebraic point of view in the sense that we have different $\mathbf{\Gamma}^k$ and $\mathbb{W}^k(C,\Phi)$ for each space degree of freedom x_k. The MCTDH equations are widely used in quantum chemistry (see e.g. [2]). To our knowledge, the only mathematical result obtained for the MCTD methods has been given by Lubich and Koch [9] who dealt with the MCTDH equations in parallel to our work. Their existence and uniqueness result uses not only the assumption of the invertibility of $\mathbf{\Gamma}^k(t)$ for $1 \le k \le K$ and $t \le T$ for an arbitrary T, but also the technical assumption on the interactions potential between the electrons to be bounded and twice continuously differentiable, with bounded first and second derivatives. The functional part of the solution is proved to be in H^2. Their result uses Lie commutator method. We are able to deal with the singular Coulomb interactions both for MCTDH and MCTDHF - of course, the case of bounded / regular potential follows immediately.

2.2 Energy conservation for MCTDHF without recourse to a variational principle

The result outlined in the previous section are based, in particular, on the energy conservation. That is

Theorem 2.3. *Let $(C^0,\Phi^0) \in \mathscr{S}_N^K \cap H^1(\mathbb{R}^3)^K$. If there exists $(C(t),\Phi(t))$ satisfying the MCTDHF system* (6.3a)–(2.4c) *for all $t \in [0,T]$ for some $T > 0$, then we have*

$$\langle \Psi(t)|H_N|\Psi(t)\rangle_{L^2(\mathbb{R}^{3N})} = \langle \Psi(t=0)|H_N|\Psi(t=0)\rangle_{L^2(\mathbb{R}^{3N})},$$

for all $t \in [0,T]$.

From now on, we reserve the notation V to the multiplicative electronic interaction operator $\sum_{1\le i<j\le N}\frac{1}{|x_i-x_j|}$. Thus, the operator $\mathbf{H}_x$ will be then reserved to $-\Delta-\sum_{i=1}^{M}\frac{z_i}{|x-\bar{x}_i|}$. Also, we introduce some notation.

$$\begin{aligned}\Psi_i^{(k)}(x_1,\dots,x_{k-1},x_{k+1},\dots,x_N) &= \int_{\mathbb{R}^3}\Psi(x_1,\dots,x_N)\,\bar{\phi}_i(x_k)\,dx_k,\\ &:= \Big\langle \Psi\,,\,\phi_i\Big\rangle_{-k}.\end{aligned}$$

Notice that $\Psi_i^{(k)}$ is a skew-symmetric function that depends on time and on all space variables except x_k, and, for every $1\le k\le K$,

$$\Psi(x_1,\dots,x_N)=\sum_{i=1}^{K}\phi_i(x_k)\,\Psi_i^{(k)}(x_1,\dots,x_{k-1},x_{k+1},\dots,x_N). \tag{2.7}$$

Now, from the one hand, for $t\in[0,T]$, we have

$$\begin{aligned}\langle\Psi_j^{(k)}\big|\Psi_i^{(k)}\rangle_{L^2(\mathbb{R}^{3(N-1)})} &= \int_{\mathbb{R}^{3(N-1)}}\Big\langle\Psi\,|\,\phi_j\Big\rangle_{-k}\Big\langle\,\phi_i\,|\Psi\,\Big\rangle_{-k}dx_1\dots dx_{k-1}dx_{k+1}\dots dx_N,\\ &= N^{-1}\int_{\mathbb{R}^6}\gamma_\Psi(z',z)\,\phi_i(z)\,\overline{\phi}_j(z')\,dz\,dz',\\ &= N^{-1}\,\mathbf{\Gamma}_{i,j}.\end{aligned}$$

The calculation above is justified bt the fact that if $(C(t),\Phi(t))$ satisfies the MCTDHF system (6.3a)–(2.4c), then it belongs to $\mathscr{S}_N^K$ for all $t\in[0,T]$. Now, introduce the notation

$$[\Psi\otimes\Psi]_{:2}(x,y,x',y')=\frac{N(N-1)}{2}\int_{\mathbb{R}^{3(N-2)}}\Psi(x,y,x_3,\dots,x_N)\,\overline{\Psi}(x',y',x_3,\dots,x_N)\,dx_3\dots dx_N.$$

Thus, from the other hand, we have

$$\begin{aligned}\Big\langle\Psi_j^{(k)}\big|\frac{1}{|x_k-x_l|}\big|\Psi_i^{(k)}\Big\rangle_{L^2(\mathbb{R}^{3(N-1)})} &= \Big\langle\,\big\langle\,\Psi\,,\,\phi_j\big\rangle_k\big|\frac{1}{|x_k-x_l|}\big|\,\langle\,\Psi\,,\,\phi_i\rangle_k\,\Big\rangle_{L^2(\mathbb{R}^{3(N-1)})},\\ &= \frac{2}{N(N-1)}\int_{\mathbb{R}^3}\int_{\mathbb{R}^6}[\Psi\otimes\Psi]_{:2}(x,x_l,y,x_l)\,\overline{\phi}_i(x)\,\phi_j(y)\,dx\,dy\,V_{k,l}\,dx_l,\\ &= \frac{1}{N(N-1)}\,\mathbb{W}(C,\Phi)_{i,j}(t,x_k).\end{aligned}$$

We now show that

$$i\frac{\partial\Psi}{\partial t}-H_N\Psi=-(I-\mathscr{P}_\Phi)[V\Psi]+\sum_{k=1}^{N}\sum_{i=1}^{K}\Psi_i^{(k)}\,(\mathbf{I}-\mathbf{P}_\Phi)\left[\mathbf{\Gamma}^{-1}\mathbb{W}(C,\Phi)\,\Phi\right]_i:=\mathscr{R}(\Psi), \tag{2.8}$$

where we have denoted by $\mathscr{P}_\Phi$ the projector on the space spanned by all Slater determinants that may be formed from the ϕ_i's; that is

$$\mathscr{P}_\Phi=\sum_{I\in\mathscr{A}_N^K}\Big\langle\,\cdot\,\Big|Det_\Phi(I)\Big\rangle_{L^2(\mathbb{R}^{3N})}Det_\Phi(I).$$

Note that, on the space of skew-symmetric functions $\bigwedge_{i=1}^{N} L^2(\mathbb{R}^3)$, $\mathscr{P}_\Phi$ coincide with the projector on the Hartree products

$$\mathscr{P}_\Phi = \sum_{l_1,\dots,l_N=1}^{K} \Big\langle \cdot, \prod_{k=1}^{N} \phi_{l_k} \Big\rangle_{L^2(\mathbb{R}^{3N})} \prod_{k=1}^{N} \phi_{l_k}.$$

Notice that we invert the matrix $\mathbf{\Gamma}$, this justified by the fact that we assumed that a solution to the MCTDHF system exists. This is sufficient to implies the invertibility of $\mathbf{\Gamma}(t)$ for all $t \in [0,T]$.

In order to prove (2.8), we start from (2.7) to get

$$\frac{\partial}{\partial t}\Psi = \sum_{I\in\mathscr{A}_N^K} \left[\frac{d}{dt} C_I\right] Det_\Phi(I) + \sum_{k=1}^{N}\sum_{i=1}^{K} \frac{\partial \phi_i(x_k)}{\partial t}\, \Psi_i^{(k)}. \tag{2.9}$$

Then, we insert the MCTDHF equations that compose $\mathscr{S}_{\text{MCHF}}$,(6.3a)–(2.4c), in (2.9) and observe that, by construction,

$$\sum_{k=1}^{N}\sum_{i=1}^{K} \mathbf{H}_{x_k}\, \phi_i(x_k)\, \Psi_i^{(k)} = \left(\sum_{k=1}^{N} \mathbf{H}_{x_k}\right)\Psi, \tag{2.10}$$

whereas, by the ODE part of the system and the skew-symmetry,

$$i \sum_{I\in\mathscr{A}_N^K} \left[\frac{d}{dt} C_I\right] Det_\Phi(I) = \sum_{I\in\mathscr{A}_N^K} \Big\langle \Psi \Big| V \Big| \Phi_\sigma \Big\rangle_{L^2(\mathbb{R}^{3N})} Det_\Phi(I) := \mathscr{P}_\Phi[V\Psi]. \tag{2.11}$$

We conclude by using the definition of the Hamiltonian H_N. Notice the presence of the projectors $(I-\mathscr{P}_\Phi)$ and $(\mathbf{I}-\mathbf{P}_\Phi)$ in the R.H.S of (2.8). In some sens, this shows formally that when the $\phi_i's$ form a complete orthonormal basis of $L^2(\mathbb{R}^3,\mathbb{C})$, $K=\infty$, the projectors above vanishes and (2.8) turn to be the exact Shrödinger equation.

Now, if we test (formally) (2.8) against $\frac{\partial}{\partial t}\Psi$ in $L^2(\mathbb{R}^{3N})$ and take the real part, we obtain

$$\begin{aligned}
\Re\Big\langle \mathscr{R}(\Psi) \Big| \frac{\partial}{\partial t}\Psi \Big\rangle_{L^2(\mathbb{R}^{3N})} &= \Re\Big\langle i\frac{\partial}{\partial t}\Psi - H_N\Psi \Big| \frac{\partial}{\partial t}\Psi \Big\rangle_{L^2(\mathbb{R}^{3N})} = \\
&= \Re\Big(i \Big\| \frac{\partial}{\partial t}\Psi \Big\|^2_{L^2(\mathbb{R}^{3N})}\Big) - \Re\Big\langle \Psi \Big| H_N \Big| \frac{\partial}{\partial t}\Psi \Big\rangle_{L^2(\mathbb{R}^{3N})} = -\frac{1}{2}\frac{d}{dt}\Big\langle \Psi \Big| H_N \Big| \Psi \Big\rangle_{L^2(\mathbb{R}^{3N})},
\end{aligned}$$

since H is self-adjoint. Then the proof of the energy conservation amounts to proving that

$$\Re\Big\langle \mathscr{R}(\Psi), \frac{\partial}{\partial t}\Psi \Big\rangle_{L^2(\mathbb{R}^{3N})} = 0.$$

According to (2.8), $\mathscr{R}(\Psi)$ is a difference of two terms, that we now treat separately. First, accord-

ing to (2.9) and to the definition of $\mathscr{P}_\Phi$, we clearly have

$$\begin{aligned}
\Re\Big\langle \big(I-\mathscr{P}_\Phi\big)[V\Psi]\Big|\frac{\partial\Psi}{\partial t}\Big\rangle_{L^2(\mathbb{R}^{3N})} &= \sum_{k=1}^{N}\sum_{i=1}^{K}\Re\Big\langle\Psi\Big|V\Big|\big(I-\mathscr{P}_\Phi\big)[\frac{\partial\phi_i(x_k)}{\partial t}\,\Psi_i^{(k)}]\Big\rangle = \\
&= \sum_{k=1}^{N}\sum_{i,j=1}^{K}\Re\Big\langle\Psi_j^{(k)}\,\phi_j\Big|V\Big|\Psi_i^{(k)}\,(\mathbf{I}-\mathbf{P}_\Phi)\frac{\partial\phi_i}{\partial t}\Big\rangle_{L^2(\mathbb{R}^{3N})}, \qquad (2.12)\\
&= \sum_{k=1}^{N}\sum_{\substack{l=1\\l\neq k}}^{N}\sum_{i,j=1}^{K}\Re\Big\langle\Psi_j^{(k)}\,\phi_j\Big|\frac{1}{|x_k-x_l|}\Big|\Psi_i^{(k)}\,(\mathbf{I}-\mathbf{P}_\Phi)\frac{\partial\phi_i}{\partial t}\Big\rangle_{L^2(\mathbb{R}^{3N})}, \qquad (2.13)\\
&= \sum_{k=1}^{N}\sum_{\substack{l=1\\l\neq k}}^{N}\sum_{i,j=1}^{K}\frac{1}{N(N-1)}\Re\Big\langle\mathbb{W}(C,\Phi)_{i,j}\phi_j\Big|(\mathbf{I}-\mathbf{P}_\Phi)\frac{\partial\phi_i}{\partial t}\Big\rangle_{L^2(\mathbb{R}^3)}, \qquad (2.14)\\
&= \sum_{i,j=1}^{K}\Re\Big\langle\mathbb{W}(C,\Phi)_{i,j}\phi_j\Big|(\mathbf{I}-\mathbf{P}_\Phi)\frac{\partial\phi_i}{\partial t}\Big\rangle_{L^2(\mathbb{R}^3)},
\end{aligned}$$

where we have used $\mathscr{P}_\Phi[\Psi_i^{(k)}\frac{\partial\phi_i}{\partial t}] = \Psi_i^{(k)}\mathbf{P}_\Phi[\frac{\partial\phi_i}{\partial t}]$ and (2.7) in (2.12), the orthogonality conditions on the ϕ_i's in (2.13) and finally (2.8) in (2.14). Finally, we have obtained

$$\begin{aligned}
\Re\Big\langle \big(I-\mathscr{P}_\Phi\big)[V\Psi]\Big|\frac{\partial\Psi}{\partial t}\Big\rangle_{L^2(\mathbb{R}^{3N})} &= \sum_{i=1}^{K}\Re\Big\langle\big[\mathbb{W}(C,\Phi)\,\Phi\big]_i\Big|(\mathbf{I}-\mathbf{P}_\Phi)\frac{\partial\phi_i}{\partial t}\Big\rangle_{L^2(\mathbb{R}^3)}, \qquad (2.15)\\
&= \Re\Big\langle\mathbb{W}(C,\Phi)\,\Phi\Big|(\mathbf{I}-\mathbf{P}_\Phi)\frac{\partial\Phi}{\partial t}\Big\rangle_{L^2(\mathbb{R}^{3N})}.
\end{aligned}$$

For the second term we proceed as follows

$$\begin{aligned}
&\sum_{k=1}^{N}\sum_{i=1}^{K}\Re\Big\langle\Psi_i^{(k)}\,(\mathbf{I}-\mathbf{P}_\Phi)[\mathbf{\Gamma}^{-1}\mathbb{W}(C,\Phi)\Phi]_i\Big|\frac{\partial\Psi}{\partial t}\Big\rangle_{L^2(\mathbb{R}^{3N})}\\
&= \sum_{k=1}^{N}\sum_{i,j=1}^{K}\Re\Big\langle\Psi_i^{(k)}\,(\mathbf{I}-\mathbf{P}_\Phi)[\mathbf{\Gamma}^{-1}\mathbb{W}(C,\Phi)\Phi]_i\Big|\Psi_j^{(k)}\frac{\partial\phi_j}{\partial t}\Big\rangle_{L^2(\mathbb{R}^{3N})}, \qquad (2.16)\\
&= \sum_{i,j=1}^{K}\Re\left(\overline{\mathbf{\Gamma}}_{i,j}\Big\langle[\mathbf{\Gamma}^{-1}\mathbb{W}(C,\Phi\Phi]_i\Big|(\mathbf{I}-\mathbf{P}_\Phi)\frac{\partial\phi_j}{\partial t}\Big\rangle_{L^2(\mathbb{R}^3)}\right), \qquad (2.17)\\
&= \sum_{i=1}^{K}\Re\Big\langle[\mathbf{\Gamma}^{-1}\mathbb{W}(C,\Phi)\Phi]_i\Big|(\mathbf{I}-\mathbf{P}_\Phi)[\mathbf{\Gamma}\frac{\partial}{\partial t}\Phi]_i\Big\rangle_{L^2(\mathbb{R}^3)},\\
&= \sum_{k,l=1}^{K}\Re\left(\big[\mathbf{\Gamma}\mathbf{\Gamma}^{-1}\big]_{l,k}\Big\langle[\mathbb{W}(C,\Phi)\Phi]_k\Big|(\mathbf{I}-\mathbf{P}_\Phi)\frac{\partial\phi_l}{\partial t}\Big\rangle_{L^2(\mathbb{R}^3)}\right).
\end{aligned}$$

In (2.16) we used (2.9),the equation of the MCTDHF system and the definition $(\mathbf{I}-\mathbf{P}_\Phi)$, and in (2.17) we used (2.8). Therefore

$$\sum_{k=1}^{N}\sum_{i=1}^{K}\Re\Big\langle\Psi_i^{(k)}\,(\mathbf{I}-\mathbf{P}_\Phi)[\mathbf{\Gamma}^{-1}\mathbb{W}(C,\Phi)\Phi]_i\Big|\frac{\partial}{\partial t}\Psi\Big\rangle_{L^2(\mathbb{R}^{3N})} = \sum_{i=1}^{K}\Re\Big\langle[\mathbb{W}(C,\Phi)\Phi]_i\Big|(\mathbf{I}-\mathbf{P}_\Phi)\frac{\partial\phi_i}{\partial t}\Big\rangle_{L^2(\mathbb{R}^3)}. \qquad (2.18)$$

If we now compare (2.15) and (2.18) with the definition of $\mathscr{R}(\Psi)$ in (2.8), we infer that

$$\left\langle \mathscr{R}(\Psi), \frac{\partial}{\partial t}\Psi \right\rangle_{L^2(\mathbb{R}^{3N})} = 0.$$

This ends the proof of the Theorem.[1]

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Remark

The next chapter is a letter to appear (accepted) in ”Applied Mathematics Letters“ with Isabelle Catto, Claude Bardos and Norbert J. Mauser.

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Chapter 3

Global-in-time existence of solutions to the MCTDHF equations: A sufficient condition

Abstract

The multiconfiguration time-dependent Hartree-Fock (MCTDHF in short) system is an approximation of the linear many particle Schrödinger equation with a binary interaction potential by non-linear "one particle" equations. MCTDHF methods are widely used for numerical calculations of the dynamics of few electron systems in quantum physics and quantum chemistry, but the time-dependent case still poses serious open problems for the analysis, e.g. in the sense that global-in-time existence of solutions is not proved yet. In this letter we present the first result ever, where global existence is proved under a condition on the initial data that has to be somewhat close to the "ground state".

3.1 Introduction

The multiconfiguration time-dependent Hartree-Fock (MCTDHF in short) system is an approximation of the linear N particle Schrödinger equation with a binary interaction potential V. MCTDHF methods are widely used for numerical calculations of the dynamics of few electron systems in quantum physics and quantum chemistry, but the time-dependent case still poses serious open problems for the analysis in the sense that global-in-time existence of solutions is not proved yet. In this letter we present the first result ever, where global existence is proved under a condition on the initial data that has to be somewhat close to the "ground state".

The MCTDHF system is composed of $K \geq N$ non-linear Schrödinger-type evolution equations (for "the orbitals", as a dynamic basis for an expansion by "Slater determinants") coupled with $\binom{K}{N}$ ordinary differential equations (for "the coefficients"). The many particle wavefunction $\Psi^N(t,x_1,...,x_N)$ can be well approximated by such linear combinations of Slater determinants that catch also "correlations", in contrast to the simple time-dependent Hartree-Fock (TDHF) method that corresponds to the special case $K = N$. In principle, the many particle wavefunction constructed from the solution of MCTDHF converges towards the exact solution Ψ^N with increasing K; however, especially in the time-dependent case there is no proof for this seemingly "obvious" property of MCTDHF. For a short and readable introduction to the multiconfiguration time-dependent Hartree-Fock (MCTDHF) system we refer the reader to [12] or, more exhaustive, [8].

The existence and uniqueness of solutions of has been established in [11] for bounded and smooth interaction potentials V. The case of the singular Coulomb potential has recently been

stated and solved in [12, 5]. All these results, however, are local-in-time in the sense that the existence, uniqueness and regularity persist only as long as the first order density operator associated to the system remains of maximal rank. In case of a "loss of rank" at a certain time T^*, the well-posedness holds only locally-in-time until T^*.

In this letter, we show how the global-in-time existence can be assured under an assumption on the energy of the initial state Ψ_I^N that will usually be in the "energy space" $H^1(\Omega)^N$, where Ω is the spatial domain.

We prove our result in two somewhat simplified situations : for the finite "discrete case" (i.e. Ω bounded and equipped with the Dirac measure) and for the case where Ω is a bounded subset of $\mathbb{R}^3$ with binary interactions as singular as the Coulomb potential, but strictly positive. The general case in $\Omega = \mathbb{R}^3$ is more technical and will be considered in a forthcoming publication [1].

Let Ω be a measured domain and $\mathscr{H}$ a bounded (or unbounded) self-adjoint operator acting on $L^2_s(\Omega^N)$, the space of skew-symmetric functions $\Psi^N(x_1,x_2,\ldots,x_N)$ (i.e. these functions are invariant under any permutation of the variables $(x_1,x_2,\ldots,x_N)$). The purpose of the MCTDHF method is to approximate the evolution of the "exact" solution $\Psi^N(t,x_1,...,x_N)$ of the linear N particle Schrödinger equation :

$$i\,\frac{\partial}{\partial t}\Psi^N = \mathscr{H}\,\Psi^N\,, \tag{3.1}$$

with a given an initial data $\Psi_I^N(x_1,...,x_N)$ in $L^2_s(\Omega^N)$.

Take an integer $K \geq N$ (the number K of "orbitals" larger than the number of particles) and take the set $\Sigma_{N,K}$ of strictly increasing maps σ from $\{1,2\ldots,N\}$ to $\{1,2\ldots,K\}$ ($\#\{\Sigma_{N,K}\} = \binom{K}{N} := r$). The same symbol σ will be used to denote the image of any such map. Then one introduces the set $\mathscr{F}_{N,K}$ of coefficients and orbitals (with $\langle .,. \rangle$ denoting the scalar product in $L^2(\Omega)$) as follows :

$$\begin{aligned}\mathscr{F}_{N,K}(\Omega) \quad = \quad & \left\{ C = (c_\sigma)_{\sigma\in\Sigma_N^K} \in \mathbb{C}^r \,:\, \sum_{\sigma\in\Sigma_{N,K}} |c_\sigma|^2 = 1 \right\} \times \\ & \left\{ \Phi = (\phi_1,\ldots,\phi_K) \in L^2(\Omega)^K \,:\, \left\langle \phi_i\,,\,\phi_j \right\rangle = \delta_{i,j} \right\},\end{aligned}$$

equipped with the usual norm of $\ell^2(\mathbb{C}^r)\times L^2(\Omega)^K$. With the maps σ and the vector $\Phi = (\phi_1,\phi_2,\ldots,\phi_K)$ one constructs the normalized *Slater determinants*:

$$\Phi_\sigma(t,x_1,\ldots,x_N) = \frac{1}{\sqrt{N!}}\det(\phi_{\sigma(i)}(t,x_j))_{1\leq i,j\leq N}, \tag{3.2}$$

The MCTDHF *ansatz* then consists in taking linear combinations of Slater determinants :

$$\pi(C,\Phi) := \Psi = \sum_{\sigma\in\Sigma_N^K} c_\sigma(t)\,\Phi_\sigma(t,x_1,\ldots,x_N)\,. \tag{3.3}$$

Observe that for any K, K' one has

$$\forall\, K' \leq K \leq \infty \Rightarrow \pi(\mathscr{F}_{N,K'}) \subseteq \pi(\mathscr{F}_{N,K}) \subseteq L^2_s(\Omega^N)\,. \tag{3.4}$$

For all N-particle wavefunctions $\Psi \in L^2(\Omega^N)$ we define the corresponding following "1-particle" density matrix via the trace

$$[\Psi \otimes \Psi]_{:1}(t,x,y) = \int_{\Omega^{N-1}} \Psi(t,x,z_2,\ldots,z_N)\overline{\Psi}(t,y,z_2,\ldots,z_N)dz_2\ldots dz_N.$$

Then the "first order density operator" or "one particle density operator", which is a self-adjoint non negative operator on $L^2(\Omega)$, associated to $\pi(C,\phi)$ for $(C,\Phi) \in \mathscr{F}_{N,K}$ is defined *via* its normalized kernel

$$D_1 := N\,[\pi(C,\Phi)\otimes\pi(C,\Phi)]_{:1}(t,x,y) \quad = \quad \sum_{i,j=1}^{K} \gamma_{i,j}(t)\,\phi_i(t,x)\,\overline{\phi}_j(t,y) := [\mathbf{\Gamma}\,\Phi]\otimes\overline{\Phi}, \quad (3.5)$$

with $\Gamma = (\gamma_{ij})^T$ a positive $K\times K$ Hermitian matrix of trace N. D_1 is then of operator norm less or equal to 1 (cf. [10], [3, Lemma 5.2]). Moreover $\gamma_{i,j}$ is given by the formula:

$$\gamma_{i,j} = \sum_{i\in\sigma, j\in\tau, \sigma\setminus i=\tau\setminus j} (-1)^{\sigma^{-1}(i)}(-1)^{\tau^{-1}(j)}\, c_\sigma\, \overline{c_\tau}\,. \quad (3.6)$$

Its eigenvalues will be denoted by $0 \le \gamma_K \le \gamma_{K-1} \le \ldots \le \gamma_1 \le 1$ and the *"maximal rank hypothesis"* corresponds to the following equivalent statements:

- The rank of the operator D_1 is equal to K.
- The matrix Γ is invertible.
- The smallest eigenvalue γ_K of Γ is strictly positive.

The above construction has been widely used to approximate the "ground state" energy:

$$\mathscr{E}(\mathscr{H}) = \min_{\Psi\in L^2_s(\Omega^N)} \frac{\langle \mathscr{H}\Psi \,|\, \Psi\rangle}{|\Psi|^2} \quad (3.7)$$

by

$$\mathscr{E}(K) = \min_{(C,\Phi)\in\mathscr{F}_{N,K}(\Omega)} \mathscr{E}(\pi(C,\Phi))\,, \text{ with } \mathscr{E}(\pi(C,\Phi)) = \langle \mathscr{H}(\pi(C,\Phi)), \pi(C,\Phi)\rangle. \quad (3.8)$$

Of course we have the following inequality for the energy :

$$\forall K' \le K \le \infty \Rightarrow \mathscr{E}(\mathscr{H}) \le \mathscr{E}(K) \le \mathscr{E}(K'). \quad (3.9)$$

Finally we recall that in the "physical case" namely in $L^2(\mathbb{R}^{3N})$ with $\mathscr{H}$ being the sum of the "kinetic energy" Laplacian plus the binary interaction V being the Coulomb potential :

$$\mathscr{H} = -\sum_{1\le i\le N} \frac{1}{2}\Delta_{x_i} + \sum_{1\le i<j\le N} V(|x_i - x_j|)\,, \quad (3.10)$$

the above energies are finite and one has always $\mathscr{E}(K) < \mathscr{E}(K-2)$ [8, 7].

With the formula (3.6) one observes that the set $\{(C,\Phi)|(C,\Phi)\subset\mathscr{F}_{N,K}(\Omega)$ such that rank of $(D_1)=K\}$ is open. It will be denoted by $\mathscr{F}^0_{N,K}$. Finally the MCTDHF dynamic is the flow $\mathscr{S}$ defined on this open subset by the equations :

$$i\frac{d}{dt}c_\sigma(t)=\left\langle \mathscr{H}\,\Psi\,|\,\Phi_\sigma\right\rangle, \tag{3.11}$$

$$i\,\mathbf{\Gamma}(t)\,\frac{\partial}{\partial t}\Phi(t,x)=(I-\mathscr{P}_\Phi)\left[[\nabla_\Phi\Psi]^\star\,\mathscr{H}\,\Psi\right], \tag{3.12}$$

with $\mathscr{P}_\Phi$ denoting the projector onto the space spanned by the ϕ_i's (cf. [4, 12, 1]).

3.2 Results and proofs

As it can immediately seen in equation (3.12), the invertibility of the matrix $\Gamma(t)$ is an essential issue for the global existence of solutions to the flow $\mathscr{S}$ and this is the very object of the present letter.

First the diagonalisation of the matrice Γ is used:

Lemma 3.1. *For all $0\le t<T$, let the mapping $t\mapsto(C,\ \Phi)(t)$ be continuous in $\mathscr{F}^0_{N,K}$ (i.e. with $\Gamma(t)$ of rank K for $0\le t<T$). Then there exists a unique unitary transformation which maps $(C,\ \Phi)(t)\mapsto(C',\Phi')(t)$ and which diagonalizes D_1 (hence the matrix $\Gamma(t)$). That is,*

$$\pi((C,\Phi))=\sum_\sigma c_\sigma\Phi_\sigma=\sum_\sigma c'_\sigma\Phi'_\sigma=\pi((C',\Phi')),\quad D_1=\sum_{i,j=1}^K\gamma_{i,j}\,\phi_i\otimes\overline{\phi_j}=\sum_{i=1}^K\gamma_i\,\phi_i'\otimes\overline{\phi_i'}. \tag{3.13}$$

The crucial non trivial point in the above statement is the fact that with the full rank hypothesis the unitary transform is uniquely determined.

Now, we claim

Lemma 3.2. *Let $(C^n,\Phi^n)\in\mathscr{F}^0_{N,K}$ be a sequence that converges weakly towards $(C^\star,\Phi^\star)$ in $\mathbb{C}^r\times L^2(\Omega)^K$.*
Assume that , for fixed $1\le m\le K$ and $\beta>0$

- *The eigenvalues of $\mathbf{\Gamma}^n$ (associated with C^n) satisfy*

$$\begin{cases}\gamma^n_K\le\ldots\le\gamma^n_m\le\ldots\le\gamma^n_1,\quad \lim_{n\to+\infty}\gamma^n_m=0,\\ \\ \text{for}\quad 1\le p\le m-1,\quad \lim_{n\to+\infty}\gamma^n_p=\gamma^\star_p\ge\beta>0\,.\end{cases} \tag{3.14}$$

- *After diagonalization as in* (3.13)*, the sequence ${\phi'}_i^n$ satisfies*

$$1\le p\le m-1\ \Rightarrow\lim_{n\to+\infty}\|{\phi'}_p^n-{\phi'}_p^*\|_{L^2(\Omega)}=0\,.$$

Then, the associated sequence of wavefunctions $\Psi^n\in L^2(\Omega^N)=\pi(C'^n,\Phi'^n)$ converge towards

$$\Psi^*:=\sum_{\sigma\cap\{m,\ldots,K\}=\emptyset}\left[\lim_{n\to+\infty}c'^n_\sigma\right]\Phi'^*_\sigma.$$

Proof. Using the unitary transformation, we have

$$\Psi^n = \pi((C^n,\Phi^n)) = \pi((C'^n,\Phi'^n)) = \sum_{\sigma\cap\{m,\dots,K\}\neq\emptyset} c'^n_\sigma\,\Phi'^n_\sigma \quad + \sum_{\sigma\cap\{m,\dots,K\}=\emptyset} c'_\sigma\,\Phi'^n_\sigma\,. \tag{3.15}$$

On the one hand, the first sum in (5.110) converges strongly towards 0 by (5.101). Indeed,

$$\Big\| \sum_{\sigma\cap\{m,\dots,K\}\neq\emptyset} c'^n_\sigma\,\Phi'^n_\sigma \Big\|_{L^2(\Omega^N)} = \sum_{\sigma\cap\{m,\dots,K\}\neq\emptyset} |c'_\sigma|^2 \le \sum_{m\le p\le K}\ \sum_{p\in\sigma} |c'_\sigma|^2 = \sum_{m\le p\le K} \gamma'^n_p \to 0\,.$$

On the other hand, the second sum in (5.110) consists of terms of the form

$$c'^n_\sigma \prod_{1,\dots,m\notin\sigma} \phi'^n_{\sigma(k)}\,m$$

in other words, it ia a tensor products of strongly convergent functions in $L^2(\Omega)$. Thus the second sum converges strongly in $L^2(\Omega^N)$ towards $\Psi^\star$. □

Following the Lemma 3.2, we have

Corollary 3.1. *With the assumptions of the Lemma 3.2 the wavefunction $\Psi^\star$, which is a priori in $\pi(\mathscr{F}_{N,K})$, is in fact in $\pi(\mathscr{F}_{N,m-1})$.*

To explain our results, first a discrete model is considered, then the extension to the classical "physical" problems in a bounded domain is given.

There are several good reasons to consider a discrete problem. First it may be by itself a model for binary interactions. Second it corresponds to the system which is obtained by any kind of discretisation of the continuous problem, in particular when spectral or Galerkin methods are involved. Finally explaining first the discrete model and then the continuous one allows to clearly see separately the issues which are related to analysis from the ones related to the algebraic structure of the problem.

A "discrete case" This situation corresponds to the case when Ω is a finite set of points of cardinal $\#\Omega = L$. We identify the functions ϕ_k with the vectors $\phi_k(l) \in \mathbb{C}, 1 \le l \le L$. $L^2(\Omega)$ is equipped with the Euclidian scalar product. This provides a natural definition for the projector $\mathscr{P}_\Phi$ and the adjoint of $[\nabla_\Phi \Psi]$. Next, Let H_0 be a self adjoint operator in $L^2(\Omega)$ and a potential V represented by a symmetric matrix $(V_{lm})_{1\le l,m\le L}$. The discrete Hamiltonian $\mathscr{H}$ is given by the formula:

$$(\mathscr{H}\,u)(l_1\dots l_N) = \left(\sum_{1\le i\le N} H_{0,i}\,u\right)(l_1\dots l_N) + \left(\sum_{1\le i<j\le N} V_{l_i,l_j}\right) u(l_1\dots l_N)\,. \tag{3.16}$$

Then, we have

Theorem 3.1. *Let the initial data $(C^0,\Phi^0) \in \mathscr{F}_{N,K}$ such that $\mathscr{E}(\pi(C^0,\Phi^0)) < \mathscr{E}(K-1)$.*
Then the discrete flow $\mathscr{S}$ defined by the discrete version of the system (3.11),(3.12) *admits a unique global-in-time solution.*

Proof. In this configuration the equations (3.11) and (3.12) are purely algebraic. Moreover, the matrix $\Gamma(t=0)$ is of rank K otherwise the operator D_1 would be of rank less than K and by the "Löwdin theorem" [10] D_1 would then belong to $\pi(\mathscr{F}_{N,K-1})$ which is incompatible with the hypothesis

$$\mathscr{E}(\pi(C^0,\Phi^0)) < \mathscr{E}(K-1)\,. \tag{3.17}$$

Therefore, the system has a unique local-in-time solution. Moreover, this solution exists as long as $\mathbf{\Gamma}(t)$ is invertible. In the sequel, we shall prove by contradiction the non-existence of a time $T^\star$ such that:

$$0 \le t < T^* \Rightarrow \gamma_p(t) > 0 \quad \text{and} \quad \lim_{t<T^*,\, t\to T^*} \gamma_p(t) = 0 \quad \text{for some} \quad 1 \le p \le K\,.$$

In fact, with the compactness of $\mathscr{F}_{N,K}$, up to extraction of sub-sequences, one can find an integer $1 \le m \le K$, a positive real β and a sequence $t_n < T^\star$ and $\lim_{n\to+\infty} t_n = T^\star$ such that

$$\lim_{n\to+\infty} \gamma_m(t_n) = 0\,,\ 0 < \beta \le \gamma_{m-1}(t_n) \text{ and } \lim_{n\to+\infty} (C(t_n),\Phi(t_n)) = (C^*,\Phi^*) \in \mathscr{F}_{N,K}\,.$$

Then, in the situation of the Lemma 3.2, $\phi_p(t_n)$ converge toward $\phi_p^\star$ for all p, in particular for $1 \le p \le m-1$. Since, the model is discrete, the strong convergence of the $\phi_p(t_n)$ for all $1 \le p \le m-1$ is obvious. Hence, from the corollary 3.1, one deduces that $\Psi^\star = \pi(C^\star,\Phi^\star)$ belongs in fact to $\pi(\mathscr{F}_{N,m-1})$. Now, by the conservation of the energy, the continuity, and (5.98) one obtains the following contradiction

$$\mathscr{E}(K-1) > \mathscr{E}(\Psi(t=0)) = \mathscr{E}(\Psi^n) = \mathscr{E}(\Psi^\star) \ge \mathscr{E}(m-1) \ge \mathscr{E}(K-1)\,.$$

□

The continous "physical case" In the setting of a continous space variable $x \in \Omega$, we now write the potential as follows

$$V(|x|) = \frac{a}{|x|} + V_{\text{reg}}(|x|) \quad \text{with} \quad a \ge 0 \quad \text{and } V_{\text{reg}} \in L^\infty, \tag{3.18}$$

and we take again (3.10) for the Hamiltonian $\mathscr{H}$ in $L^2_s(\Omega^N)$

The following local-in-time result holds :

Theorem 3.2. *Let $(C^0,\Phi^0) \in \mathscr{F}^0_{N,K}(\Omega)$ being an initial data such that $\mathscr{E}\big(\pi(C^0,\Phi^0)\big) < \infty$. Then, the flow $\mathscr{S}$ defined by* (3.10),(3.11),(3.12) *has a unique local-in-time solution for $t \in (-T^*,T^*)$ with $T^* > 0$. In particular, the matrix $\Gamma(t)$ is invertible on this time interval and the system preserves its total energy*

$$\mathscr{E}\big(\pi(C(t),\Phi(t)\big) = \mathscr{E}\big(\pi(C^0,\Phi^0)\big). \tag{3.19}$$

Furthermore, this solution can be uniquely extended as long as this matrix $\Gamma(t)$ remains invertible.

This theorem has been already proven for a bounded and smooth binary interacting potential ($a = 0$ in (3.18)) (cf. [11]) and for the Coulomb potential ($(0 < a)$ cf. [12, 5]). In a simplified setting we now take Ω be an open and bounded domain of $\mathbb{R}^3$ with boundary $\partial\Omega$ and impose homogeneous Dirichlet boundary condition:

$$\Psi = 0 \text{ on } \partial(\Omega)^N.$$

Then for a global-in-time result we have now the following:

Theorem 3.3. *Let $\mathscr{H}$ as in* (3.10) *and let the initial data $(C^0,\Phi^0) \in \mathscr{F}_{N,K}$ and $\Phi^0 \in H^1(\Omega)^K$ be such that $\mathscr{E}(\pi(C^0,\Phi^0)) < \mathscr{E}(K-1)$. Then, the flow $\mathscr{S}$ defined by* (3.10),(3.11),(3.12) *has a unique global-in-time solution in $\ell^2(\mathbb{C}^r) \times H^1(\Omega)^K$.*

Proof. We can conclude by contradiction (as in the discrete case), using the conservation of energy and the fact that the injection of $H^1(\Omega)$ in $L^2(\Omega)$ is compact - which is precisely the point where the simplification of bounded Ω is used.

If $\mathbf{\Gamma}(t)$ becomes degenerate at $T^\star$, then there exists a sequence t_n and an integer $1 \leq m \leq K$ such that

$$\lim_{n\to\infty} \gamma_m(t_n) = 0 \quad \text{and} \quad 0 < \beta \leq \gamma_{m-1}(t_n)\,.$$

Now, in order to adapt the proof done in the discrete case, it is enough to show that

- $\lim_{n\to+\infty} \mathscr{E}(\pi(C^n,\Phi^n)) \geq \mathscr{E}(\pi(C^*,\Phi^*))$.

- For $1 \leq p \leq m-1$, the sequence $\phi_p^n = \phi_p(t_n)$ stays in a relatively compact set of $L^2(\Omega)$.

The first point is a consequence of the lower semi-continuity of the energy (see [9] and add V_{reg}). We shall use the conservation of the energy in order to prove the second point. In fact, for $0 \leq t_n < T^\star$, we have

$$\begin{aligned}\mathscr{E}(\pi((C^0,\Phi^0))) &= \mathscr{E}(\pi(C^n,\Phi^n)) \\ &= \int_{\Omega^N} \left[\frac{1}{2}|\nabla\Psi(t_n)|^2 + \left(\sum_{1\leq i<j\leq N} V(|x_i-x_j|)\right)|\Psi(t_n))|^2\right] dx_1,\ldots,dx_N.\end{aligned}$$

Now, the Coulomb constant a in (3.18) being positive and $V_{reg} \in L^\infty$, one deduce the existence of a non-negative constant $b < \infty$ such that

$$b \geq \frac{1}{2}\int_{\Omega^N} |\nabla\Psi(t_n)|^2 dx_1,\ldots,dx_N = \frac{1}{2}\int_\Omega (\Gamma(t_n)\,\nabla\Phi(t_n),\nabla\Phi(t_n))\,dx\,.$$

Now, using the diagonalization procedure, on obtains (omitting the $'$)

$$b \geq \frac{1}{2}\sum_{1\leq p\leq K} \gamma_p(t_n)\int_\Omega |\nabla\phi_p(t_n)|^2 dx \geq \beta \sum_{1\leq p\leq m-1}\int_\Omega |\nabla\phi_p(t_n)|^2 dx.$$

The compactness of the $\phi_p(t_n)$ for $1 \leq p \leq m-1$ comes, with the hypothesis Ω bounded, from the Rellich Theorem. □

3.3 Conclusion

We have shown that the global-in-time well-posedness of the MCTDHF system (3.10),(3.11),(3.12) follows from the sufficient condition on the initial data $(C^0,\Phi^0) \in \mathscr{F}_{N,K}$ that it's energy is close to the "ground state" energy (3.7) in the sense that

$$\mathscr{E}(\pi(C^0,\Phi^0)) < \mathscr{E}(K-1). \tag{3.20}$$

For sake of simplicity, the electrons-nuclei interaction has been omitted in (3.10) and the domain Ω is assumed bounded. The extension of our results to these situations will be described in a forthcoming publication [1], e.g. the case $\Omega = \mathbb{R}^3$.

The hypothesis (3.20) can be viewed as a stability constraint in the sense of numerical analysis. Given any initial $\Psi_I^N \in L_s^2(\Omega^N)$ for the Schrödinger dynamic (3.1) and any $\varepsilon > 0$. Then, there exists, for K large enough, an element $(C^0, \Phi^0) \in \mathscr{F}_{N,K}^0$ such that

$$\|\Psi_I^N - \pi(C^0, \Phi^0)\|_{L^2(\Omega^N)} \leq \varepsilon \tag{3.21}$$

However the meaning of our result is that, to ensure global existence, the number of orbitals K in the MCTDHF approximation has to be chosen in such a way that the condition (3.20) on the initial energy is satisfied.[1]

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remark

The next chapter is a work with Norbert J. Mauser. Preprint to be submitted.

Mauser, N.J. and Trabelsi, S.: L^2 Analysis of the Multiconfiguration Time-dependent Hartree-Fock Equations

Chapter 4

L^2 Analysis of the Multiconfiguration Time-dependent Hartree-Fock Equations

Abstract

The multiconfiguration methods are widely used by quantum physicists and chemists for numerical approximations of the many electron Schrödinger equation. Recently, some mathematical rigorous results were obtained on the time-dependent MCTDHF equations : short-in-time wellposedness in the Sobolev space H^2 for bounded interactions, in the energy space for singular Coulomb interactions, as well as global wellposedness under a sufficient condition on the energy of the initial data. The present contribution extends the analysis in the setting of an L^2 theory to the MCTDHF, for general interaction potentials that include the singular Coulomb case. This kind of results is also the theoretical foundation of ad-hoc methods used in calculations when modifications ("regularization") of the density matrix destroy the conservation of energy (but keep invariant the L^2 norm).

4.1 Introduction

The Multi-Configuration Time-Dependent Hartree-Fock (MCTDHF) system represents a hierarchy of approximations of the linear N particle Schrödinger equation.

$$i\frac{\partial}{\partial t}\Psi(t,x_1,\ldots,x_N) = \mathscr{H}\,\Psi(t,x_1,\ldots,x_N), \tag{4.1}$$

where $\Psi \in \mathscr{X} = L^2(\mathbb{R}^{3N};\mathbb{C})$ is the N-particle wavefunction which we normalize to one in $\mathscr{X}$ for $|\Psi|^2$ being interpreted as a probability density (see the introduction of this manuscript or any textbook of quantum mechanics). The N-body Hamiltonian of the system is then the self-adjoint operator $\mathscr{H}$ acting on the Hilbert space $\mathscr{H}$ and given by

$$\mathscr{H} = \sum_{1\le i\le N}\left(-\frac{1}{2}\Delta_{x_i} + U(x_i)\right) + \sum_{1\le i<j\le N} V(|x_i - x_j|). \tag{4.2}$$

The subscript x_i of $-\frac{1}{2}\Delta_{x_i}$ means derivation with respect to the i^{th} space variable of the wavefunction Ψ and $U(x)$ is the "external potential" created by the nuclei localized at fixed positions R_m with charges $z_m > 0$ for $1 \le m \le M$. The last term of $\mathscr{H}$ is the interaction potential between the electrons, which is fundamentally given by the Couloumb interaction. Hence we have

$$U(x) = -\sum_{m=1}^{M}\frac{z_m}{|x-R_m|} \quad \text{and} \quad V(|x-y|) = \frac{1}{|x-y|}. \tag{4.3}$$

For a good introduction to MDTDHF the mathematically oriented reader is referred to [16, 17]. A particular advantage over the simple Time-Dependent Hartree-Fock (TDHF) method (see e.g. [2, 3] is that MCTDHF can catch "correlation", an essential concept of many electron systems that vanishes (by definition) for TDHF. We refer the reader to [13, 14] for more details.

MCTDHF methods are widely used for numerical calculations of the dynamics of few electron systems in quantum physics and quantum chemistry (see e.g. [18] and references therein, also [4] for the MCTDH case), but the time-dependent case still poses serious open problems for the analysis in the sense that global-in-time existence of solutions is not proved yet.

The MCTDHF system is composed of $K \geq N$ non-linear Schrödinger-type evolution equations (for "the orbitals", as a dynamic basis for an expansion by "Slater determinants") coupled with $r := \binom{K}{N}$ first order differential equations (for " the coefficients" C). The many particle approximate wavefunction $\Psi_{\mathrm{MC}}(t,x_1,...,x_N)$ can be well approximated by such linear combinations of Slater determinants much better than by the simple TDHF method that corresponds to the special case $K = N$. In principle, for fixed N the MCTDHF equation yields a hierarchy of models with increasing accuracy with increasing K, in the sense that many particle wavefunction constructed from the solution of MCTDHF converges (in some sense) towards the exact solution Ψ^N with increasing K. However, especially in the time-dependent case, there is no proof for this seemingly "obvious" property of MCTDHF.

Let us now formulate the MCTDHF equations which are principially more complicated to write down than the "usual NLS" like cubic NLS or "Schrödinger-Poisson". For a short and readable introduction to the multiconfiguration time-dependent Hartree-Fock (MCTDHF) system, we refer the reader to [15, 20, 1], or [16, 10, 17] focussed on the stationary case. First of all, we introduce the set $\mathscr{F}_{N,K}$ of "coefficents and orbitals" (C,Φ)

$$\mathscr{F}_{N,K} = \left\{ C = (C_\sigma)_{\sigma\in\Sigma_{N,K}} \in \mathbb{C}^r, \quad \sum_{\sigma\in\Sigma_{N,K}} |C_\sigma|^2 = 1 \right\} \times$$
$$\left\{ \Phi = (\phi_1,\ldots,\phi_K) \in L^2(\mathbb{R}^3)^K, \quad \int_{\mathbb{R}^3} \phi_i\,\bar{\phi}_j\,dx = \delta_{i,j} \right\},$$

with $\delta_{i,j}$ being the Kronecker delta, the bar denotes the complex conjugation and

$$\Sigma_{N,K} = \Big\{ \sigma = \{\sigma(1) < \ldots < \sigma(N)\} \subset \{1,\ldots,K\} \Big\}, \qquad |\Sigma_{N,K}| = \binom{K}{N} := r.$$

That is, the range of the family of increasing mappings $\sigma : \quad \{1,\ldots,N\} \longrightarrow \{1,\ldots,K\}$ for $1 \leq N \leq K$, $N,K \in \mathbb{N}^\star$. Now, given $\sigma \in \Sigma_{N,K}$, we define the associated *Slater determinant* as follows

$$\Phi_\sigma(x_1,\ldots,x_N) = \frac{1}{\sqrt{N!}} \begin{vmatrix} \phi_{\sigma(1)}(x_1) & \ldots & \phi_{\sigma(1)}(x_N) \\ \vdots & & \vdots \\ \phi_{\sigma(N)}(x_1) & \ldots & \phi_{\sigma(N)}(x_N) \end{vmatrix}.$$

That is, the determinant built from the ϕ_i's such that $i \in \sigma$. The factor $\frac{1}{\sqrt{N!}}$ ensures then the normalization $\|\Phi_\sigma\|_{L^2(\mathbb{R}^{3N})} = 1$. The MCTDHF wavefunction reads

$$\Psi_{\mathrm{MC}}(t,x_1,\ldots,x_N) \quad := \quad \Psi_{\mathrm{MC}}(C(t),\Phi(t)) \quad := \quad \sum_{\sigma\in\Sigma_{N,K}} C_\sigma(t)\,\Phi_\sigma(t,x_1,\ldots,x_N).$$

In the following, we use the convention that bold letters will be reserved for "one particle" operators, that is operators acting on $L^2(\mathbb{R}^3)$, while "black board" letters stand for matrices. Moreover, by abuse of notation, we use the same notation for operators acting on $L^2(\mathbb{R}^3)$ and diagonal matrix operators acting on vectors in $L^2(\mathbb{R}^3)^K$. The equations of motion associated to the *ansatz* (4.4) correspond to a variational procedure and some gauge algebra for which we refer to [4, 15] and particulary to [1]. In our formulation we obtain the following coupled system :

$$\mathscr{S}: \quad \begin{cases} i\frac{d}{dt}\,C(t) = \mathbb{K}[\Phi](t)\,C(t), \\ \\ i\,\mathbf{\Gamma}[C(t)]\,\frac{\partial}{\partial t}\,\Phi(t,x) = \mathbf{\Gamma}[C(t)]\,\left[-\frac{1}{2}\Delta_x + U(x)\right]\,\Phi(t,x) + (I-\mathbf{P}_\Phi)\,\mathbb{W}[C,\Phi](t,x)\,\Phi(t,x). \end{cases}$$

Let us now define the different operators and matrices involved in the system $\mathscr{S}$. First, $\mathbb{K}[\Phi]$ denotes an $r\times r$ Hermitian matrix depending only on the ϕ_i's and indexed by the sets $\sigma,\tau\in\Sigma_{N,K}$

$$\begin{aligned}\mathbb{K}[\Phi]_{\sigma,\tau}(t) = \frac{1}{2}\sum_{i,j\in\tau,\,k,l\in\sigma} \quad & (1-\delta_{i,j})(1-\delta_{k,l})\,\delta_{\tau\setminus\{i,j\},\sigma\setminus\{k,l\}}(-1)^{\tau}_{i,j}\,(-1)^{\sigma}_{k,l}\times \\ \times \quad & \int_{\mathbb{R}^3\times\mathbb{R}^3}\overline{\phi}_j(t,y)\phi_i(t,y)\,V(|x-y|)\,\overline{\phi}_k(t,x)\,\phi_l(t,x)\,dx\,dy,\end{aligned}$$

where the symbols $(-1)^{\sigma}_{k,l}=\pm 1$, and are not relevant for our analysis, given by

$$(-1)^{\sigma}_{k,l} = \begin{cases} (-1)^{\sigma^{-1}(k)+\sigma^{-1}(l)+1} & \text{if } k<l, \\ \\ (-1)^{\sigma^{-1}(k)+\sigma^{-1}(l)} & \text{if } k>l. \end{cases} \tag{4.4}$$

where $\sigma^{-1}(i)$ is the position of the entry i in the set σ. The matrix $\mathbf{\Gamma}[C(t)]$ is the $K\times K$ Hermitian "density matrix" depending only on the coefficients C_σ's as follow

$$\mathbf{\Gamma}[C(t)]_{i,j} = \sum_{\substack{\sigma,\tau\in\Sigma_{N,K}\\ \sigma\setminus\{i\}=\tau\setminus\{j\}}} (-1)^{\sigma^{-1}(i)+\tau^{-1}(j)}\,\overline{C}_\sigma(t)\,C_\tau(t). \tag{4.5}$$

The operator $\mathbf{P}_\Phi$ is given by

$$\mathbf{P}_\Phi(\cdot) = \sum_{i=1}^{K}\int_{\mathbb{R}^3}\cdot\,\overline{\phi}_i(t,x)\,dx\,\phi_i(t,x).$$

That is the orthogonal projector onto the space spanned by the ϕ_i's. Finally, the $K\times K$ Hermitian matrix $\mathbb{W}[C,\Phi](t,x)$ is the so called *mean field matrix* in the terminology of physics and is given by the entries

$$\mathbb{W}[C,\Phi]_{ij}(x) = 2\sum_{k,l=1}^{K}\gamma_{jkil}\int_{\mathbb{R}^3}\phi_k(y)\,V(|x-y|)\,\overline{\phi}_l(y)\,dy,$$

with

$$\gamma_{ijkl} = \frac{1}{2}\,(1-\delta_{i,j})(1-\delta_{k,l})\sum_{\substack{\sigma,\tau\,|\,i,j\in\sigma,k,l\in\tau\\ \sigma\setminus\{i,j\}=\tau\setminus\{k,l\}}}(-1)^{\sigma}_{i,j}(-1)^{\tau}_{k,l}\,C_\sigma\,\overline{C}_\tau.$$

Observe that the potential V appears in the definition of $\mathbb{W}[C,\Phi]$ and $\mathbb{K}[\Phi]$, but that the kinetic energy operator does not appear in $\mathbb{K}[\Phi]$ and hence in the equation for the $C(t)$. We refer the reader to [1] for more details on this formulation.
Hence we have to deal with a strongly non-linear coupled system of $r = \binom{K}{N}$ first order ODEs and K Schrödinger-type PDEs.

A preliminary result on the existence and uniqueness of solutions to the MCTDH system $\mathscr{S}$ (i.e. the version of the MCTDHF system valid for bosons, particles with integer spin) has been established in [15] for the drastic simplification of bounded and smooth interaction potential V (and $U \equiv 0$), where H^2 regularity for a solution associated to initial data in the Sobolev space H^2 was shown to hold. We believe that in such configuration, it is possible to get L^2 solutions with initial data in L^2.

The case of the singular Coulomb potential has recently been settled in [20, 1] with less regularity of initial data, namely in H^1. However, all these results are local-in-time in the sense that the existence, uniqueness and regularity persist only as long as the density operator associated to the system remains of maximal rank. That is, the matrix $\mathbf{\Gamma}[C(t)]$ remains invertible. In case of a "loss of rank" at a certain time T^*, the well-posedness holds only locally-in-time, until T^*. However, in [9, 1] it is shown how the global-in-time existence can be assured under an assumption on the energy of the initial state $\Psi_{\mathrm{MC}}(C(t=0),\Phi(t=0))$.

The purpose of this Chapter is, essentially, to establish an existence and uniqueness result of solutions for the Cauchy problem associated to the MCTDHF equations, the system $\mathscr{S}$ under suitably chosen conditions on the potentials U and V. More precisely, we request that the potentials satisfy

$$U,V \in L^{p_1}(\mathbb{R}^3,\mathbb{R}) + L^{p_2}(\mathbb{R}^3,\mathbb{R}), \qquad p_1,p_2 > \frac{3}{2}, \quad V \text{ even.}$$

For the sake of notational simplicity, we shall focus from now on $U,V \in L^p(\mathbb{R}^3,\mathbb{R})$ with $p > \frac{3}{2}$ omitting the decomposition in two parts. One can easily see that the same proof holds *mutatis mutandis* in the decomposed case and the challenge is nothing but a much more heavier and longer formulation of the estimates. One more simplification we shall adopt is that we shall usually write $U,V \in L^p(\mathbb{R}^3)$ with $p > \frac{3}{2}$, however, this have to be understood in the sense that $U \in L^p(\mathbb{R}^3,\mathbb{R})$ and $V \in L^q(\mathbb{R}^3,\mathbb{R})$ with different p and q satisfying $p,q > 3/2$.

Observe that the assumptions above hold true in the singular Coulomb case (4.3). Indeed, one use the following cut-off

$$\chi(r)\begin{cases} = 1 & for \quad 0 \le r \le 1 \\ \in [0,1] & for \quad 1 \le r \le 2 \\ = 0 & for \quad 2 \le r \end{cases} \quad \text{and set} \quad \begin{cases} V_1(|x|) = \frac{\chi(|x|)}{|x|} \in L^p(\mathbb{R}^3) \\ \\ V_2(|x|) = \frac{1-\chi(|x|)}{|x|} L^\infty(\mathbb{R}^3) \end{cases}$$

with

$$\frac{3}{2} \le p < 3.$$

Now, let us introduce

$$\partial\mathscr{F}_{N,K} = \Big\{ (C,\Phi) \in \mathscr{F}_{N,K} \quad : \quad \mathrm{rank}(\mathbf{\Gamma}[C]) = K \Big\}. \tag{4.6}$$

Thus, we are able to recall a result from [20, 1] that will be useful for next.

Theorem 4.1. *Let $(C^0,\Phi^0)\in\partial\mathscr{F}_{N,K}\cap H^1(\mathbb{R}^3)^K$ be a "full rank initial data", $U,V\in L^d(\mathbb{R}^3,\mathbb{R})$ and $d>\frac{3}{2}$. Then, there exists a unique maximal solution $(C(t),\Phi(t))$ to the MCTDHF system $\mathscr{S}$ in*

$$C^1\big([0,T^\star);\mathbb{C}\big)^r\times\Big(C^1\big([0,T^\star);H^{-1}(\mathbb{R}^3)\big)^K\cap C^0\big([0,T^\star);H^1(\mathbb{R}^3)\big)^K\Big).$$

Moreover, for all $0\leq t<T^\star$, we have

- *$(C(t),\Phi(t))\in\partial\mathscr{F}_{N,K}$ for all $t\in[0,T^\star)$. That is,*

$$\|C(t)\|_{\mathbb{C}^r}=\|C^0\|_{\mathbb{C}^r},\quad \|\Phi(t)\|_{L^2}=\|\Phi^0\|_{L^2},\quad \textit{and}\quad \operatorname{rank}(\mathbf{\Gamma}[C(t)])=\operatorname{rank}(\mathbf{\Gamma}[C^0])=K.$$

- $\langle\Psi_{\mathrm{MC}}(C(t),\Phi(t))|H_N|\Psi_{\mathrm{MC}}(C(t),\Phi(t))\rangle_{L^2(\mathbb{R}^{3N})}=\big\langle\Psi_{\mathrm{MC}}(C^0,\Phi^0)|H_N|\Psi_{\mathrm{MC}}(C^0,\Phi^0)\big\rangle_{L^2(\mathbb{R}^{3N})}$.

Finally, either $T^\star=+\infty$ or $T^\star<+\infty$ and $\int_0^{T^\star}\|\mathbf{\Gamma}[C(t)]^{-1}\|^{3/2}dt=+\infty$. In particular

$$\limsup_{t\to T^\star}\|\mathbf{\Gamma}[C(t)]^{-1}\|^{\frac{3}{2}}=+\infty.$$

This Theorem then yields a local well-posedness result to the MCTDHF system. However, observe that the result is possibly global since, for the time being, there is no indication that $T^\star$ is necessarily finite [1, 20]. As claimed before, the special case $N=K$ corresponds to a single Slater determinant. In particular, the set $\mathscr{F}_{N,N}$ coincides with $\partial\mathscr{F}_{N,N}$ and thus it is a smooth manifold actually. In other words, the matrix $\mathbf{\Gamma}[C]$ reduces to a globally invertible matrix since it becomes the $N\times N$ identity matrix. Therefore in that case Theorem 4.1 ensures the global existence of a unique solution of the TDHF approximation, that is $T^\star=+\infty$ and it improves then previous results obtained in [8]. Actually, the proof of the energy conservation that we gave is much more readable then the one there.

Finally, recall that this result (and the one we shall present in the next section) is valid in the case of the MCTDH which is the adequate model when dealing with bosons. The so-called Schrödinger-Poisson system (SPS), which coincides with the Hartree system in the special case of "Bose Einstein condensation" when Coulombic interactions are used, can be also obtained as a limiting case of the MCTDH and our results apply obviously to this model.

4.2 Main result

In [23], an L^2-Theory to a model is proved that is called TDHF. However, that model is a peculiar mixed state formulation that does not correspond to physically meaningful Hartree-Fock equations. Indeed, the Hartree-Fock model is characterized by N equations with *occupation numbers* equal to one which is a completely different paradigm than a mixed state for a one particle model. That is, the eigenvalues of $\mathbf{\Gamma}[C(t)]$ are one for all time $t\geq 0$. However, the result obtained in [23] can obviously be adapted to the TDHF case. In our work this TDHF result is improved.

Independently, Castella established an L^2 theory of the mixed state Schrödinger-Poisson system. More precisely, he studied a system of infinitely coupled Schrödinger equations with self-consistent Coulomb potential. The initial data needs only an L^2 regularity, so the initial kinetic

energy can possibly be infinite. Moreover, he obtained a blow-up (resp. decay) estimates for the solution as time goes to zero (resp. infinity).

Our work is inspired by Castella's results and Strichartz techniques. Our result on MCTDHF applies also to the TDHF and the TDH "pure state" case. We prove the following

Theorem 4.2. *Let $U,V \in L^d(\mathbb{R}^3,\mathbb{R})$ with $d > \frac{3}{2}$ and $(C^0,\Phi^0) \in \partial\mathscr{F}_{N,K}$ be a "full rank initial data". Then, there exists a time $T^\star > 0$ (possibly $= +\infty$) such that the MCTDHF system $\mathscr{S}$ admits solutions $(C(t),\Phi(t))$ satisfying*

- $C \in C^1([0,T^\star),\mathbb{C})^r \quad$ *and* $\quad \Phi \in C^0([0,T^\star),L^2(\mathbb{R}^3))^K$.

Moreover,

i) $\Phi(t) \in L^{\frac{4q}{3(q-2)}}([0,T],L^q(\mathbb{R}^3))^K$ *for all* $2 \leq q < 6$

ii) *The solution $(C(t),\Phi(t))$ is unique in the class*

$$L^\infty([0,T],\mathbb{C})^r \times L^\infty([0,T],L^2(\mathbb{R}^3))^K \cap L^{\frac{4q}{3(q-2)}}([0,T],L^q(\mathbb{R}^3))^K,$$

for all $2 \leq q < 6$ and $T < T^\star$.

iii) $(C(t),\Phi(t)) \in \partial\mathscr{F}_{N,K}$ *for all* $t \in [0,T^\star)$.

Hence, this result yields an L^2 theory to the MCTDHF equations as long as the density matrix $\mathbf{\Gamma}[C(t)]$ remains of full rank. Of course, this result applies also to the case of the MCTDH equations. Note that our theorem yields a global L^2 theory for the TDHF and the TDH models (the "pure state" versions of the models studied in [23, 5] as mentioned above).

The paper is structured as follow. In the next section, we collect some well-known tools, like Strichartz estimates and the properties associated to the semigroup generated by $i\frac{1}{2}\Delta$ on $L^2(\mathbb{R}^3)$. Moreover, we prove a local existence result using a standard contraction argument in an adequate space $\mathscr{X}_{T'}^{p,q}$ for a given reals p,q and a nonnegative time T'. In section 3, we shall prove that this local solution satisfies an *a priori* estimate which will be crucial in order to prolongate the solution beyond T'. Then section 4 is dedicated to the proof of the main result 4.2. Finally, we finish with some comments on this Theorem.

4.3 A few technical Lemmata

First of all, let us precise the notation we adopt throughout this chapter and recall some well known tools. The real p' will be the conjugate of p with $1 \leq p \leq \infty$, that is $\frac{1}{p} + \frac{1}{p'} = 1$. By abuse of notation, we denote $L^p = L^p(\mathbb{R}^3,\mathbb{C})$ but also $L^p = L^p(\mathbb{R}^3,\mathbb{R})$ when there is no confusion. The same notation will be used for $L^p(\mathbb{R}^3,\mathbb{C})^K$ and will be specified explicitly when necessary. The associated norms will be denoted $\|\cdot\|_{L^p}$. The same conventions are adopted for the Sobolev space $H^1\mathrm{T}$. The notation $\langle f,g\rangle$ will be reserved for the scalar product $\int_{\mathbb{R}^3} f(x)\,\overline{g}(x)\,dx$ for $f,g \in L^2(\mathbb{R}^3,\mathbb{C})$. $(\mathbf{U}(t))_{t\in\mathbb{R}}$ is the group of isometries $(e^{\frac{i}{2}t\Delta})_{t\in\mathbb{R}}$ generated by $\frac{i}{2}\Delta$ on $L^2(\mathbb{R}^3,\mathbb{C})$. Finally,

κ will be an auxiliary positive constant depending on N and K. Also const. will denotes generic constants depending on quantities that will be indicated explicitly when necessary. Next, for a given real and even potential V, we denote the operator

$$\mathbb{V}[\Phi]_{i,j}(t,x) := \int_{\mathbb{R}^3} \phi_i(t,y)\, V(|x-y|)\, \overline{\phi}_j(t,y)\, dy, \quad 1 \leq i,j \leq K.$$

Moreover, let

$$D_V[f,\overline{g}](t) = \int_{\mathbb{R}^3\times\mathbb{R}^3} f(t,x)\, V(|x-y|)\, \overline{g}(t,y)\, dx\, dy.$$

Notice that D_V depends on the potential V. We shall omit the dependency on t and x when the context make the sense clear. For a given $T>0$ we denote

$$L^{p,q}_T = L^p([0,T],L^q).$$

Now, we use the following definition

Definition 4.1. The pair of reals (p,q) is said to be *admissible*, we denote $(p,q)\in\mathscr{A}$, if and only if the following relation holds true.

$$\frac{2}{3\,p} = (\frac{1}{2}-\frac{1}{q}) \quad \text{and} \quad 2\leq q<6.$$

Thus, we are able to recall the following Lemma

Lemma 4.1. *Let $T>0$, then for all $(a,b),(p,q)\in\mathscr{A}$, $\phi\in L^2$ and $\varphi\in L^{a',b'}_T$, there exists $\rho(a)$ and $\rho(a,p)$ such that*

$$\|\mathbf{U}(t)\,\phi\|_{L^{p,q}_T} \leq \rho(p)\,\|\phi\|_{L^2} \quad \textit{and} \quad \|\int_0^t \mathbf{U}(t-s)\,\varphi(s)ds\|_{L^{p,q}_T} \leq \rho(p,a)\,\|\varphi\|_{L^{a',b'}_T}. \tag{4.7}$$

Proof. This is a classical Lemma and we refer the reader to [11, 12, 19, 6]. □

The first inequality appearing in (4.7) describes a notable smoothing effect. In particular it tell us that for all $t\in\mathbb{R}$, we have $\mathbf{U}(t)L^2=L^2$ and that for all $\phi\in L^2$, we have obviously $\mathbf{U}(t)\,\phi\in L^p$. The second inequality is crucial when dealing with non-linearities in the framework of Schrödinger type equations. Indeed, without loss of generality we write the following generic Duhamel formula

$$\psi(t) = \mathbf{U}(t)\,\phi - i\int_0^t \mathbf{U}(t-s)\,f(\psi(s)),$$

for a given functional f. Then, the first inequality of (4.7) allows to control the L^2 norm of $\mathbf{U}(t)\,\phi$ in terms of the L^2 norm of ϕ. However, it is merely impossible to control the L^2 norm of $\int_0^t \mathbf{U}(t-s)\,f(\psi(s))\,ds$ in terms of the one of ψ for general non-linearities f. We shall see that in our case it is possible if one consider a bounded real potential V. However, this is not the more relevant and interesting case. The second inequality of (4.7) will, then, give us the possibility to control the $L^{p,q}_T$ norm of $\int_0^t \mathbf{U}(t-s)\,f(\psi(s))\,ds$ for a given $T>0$ and a couple of reals $(p,q)\in\mathscr{A}$ in terms of the $L^{p',q'}_T$ norm of f which will be enough for us to conclude.

The Duhamel Formula associated to the MCTDHF system $\mathscr{S}$ for a given initial data (C^0,Φ^0) is written as follows for all time t such that $\mathbf{\Gamma}[C(t)]$ is invertible,

$$\begin{bmatrix} C(t) \\ \\ \Phi(t) \end{bmatrix} = \begin{bmatrix} C^0 \\ \\ \mathbf{U}(t)\,\Phi^0 \end{bmatrix} - i\int_0^t \begin{bmatrix} \mathbb{K}[\Phi(s)]\,C(s) \\ \\ \mathbf{U}(t-s)\left[U\,\Phi(s)+\mathbf{\Gamma}[C(s)]^{-1}\,(I-\mathbf{P}_\Phi)\,\mathbb{W}[C(s),\Phi(s)]\,\Phi(s)\right] \end{bmatrix} ds. \tag{4.8}$$

Remark 4.1. The potential U being time-independent, we chose for simplicity to add it to the non-linear part. But a different way to proceed is to consider the linear PDE $i\frac{\partial}{\partial t}u(t,x) = -\frac{1}{2}\Delta u(t,x) + U(x)\,u(t,x)$ and find adequate reals p for $U \in L^p$ such that one can associate to this flow a propagator that satisfies Strichartz-type estimates (4.7). We refer the reader to [22] for instance.

Next, we state the following Lemma which will be used intensively along this paper.

Lemma 4.2. *Let $U,V \in L^d$, $(p,q),(p_i,q_i) \in \mathscr{A}$ for $i=1,\dots,4$ and $T>0$. Then,*

$$\|U\,\phi_1\|_{L_T^{p',q'}} \leq T^{\frac{3}{2}\left(\frac{1}{q}+\frac{1}{q_1}\right)-\frac{1}{2}}\,\|U\|_{L^d}\,\|\phi_1\|_{L_T^{p_1,q_1}}, \quad \frac{1}{q}+\frac{1}{q_1} = 1-\frac{1}{d}, \tag{4.9}$$

$$\|\mathbb{V}[\Phi]_{1,2}\,\phi_3\|_{L_T^{p',q'}} \leq T^{\frac{3}{2}\left(\frac{1}{q}+\Sigma_{k=1}^3\frac{1}{q_k}\right)-2}\,\|V\|_{L^d}\prod_{i=1}^3\|\phi_i\|_{L_T^{p_i,q_i}}, \quad \frac{1}{q}+\sum_{i=1}^3\frac{1}{q_i} = 2-\frac{1}{d}, \tag{4.10}$$

$$\left|\int_0^T D_V(\phi_1(t)\,\overline{\phi}_2(t),\phi_3(t)\,\overline{\phi}_4(t))\,dt\right| \leq T^{\frac{3}{2}\Sigma_{k=1}^4\frac{1}{q_k}-2}\,\|V\|_{L^d}\prod_{i=1}^4\|\phi_i\|_{L_T^{p_i,q_i}}, \tag{4.11}$$

$$\sum_{i=1}^4\frac{1}{q_i} = 2-\frac{1}{d}. \tag{4.12}$$

Proof. The proof is just a straightforward calculation based on the well-known Hölder and Young inequalities in space and time; we leave it to the reader and refer to [23] for the detailed calculations for a similar estimate. □

A straightforward corollary of the Lemma above is then

Corollary 4.1. *Let $U,V \in L^d, d>\frac{3}{2}$ and $(p,q=\frac{2d}{d-1}) \in \mathscr{A}$. Then, for all $T>0$, we have*

$$\|U\,\Phi\|_{L_T^{p',q'}} \leq \kappa\,T^{\frac{3}{q}-\frac{1}{2}}\,\|U\|_{L^d}\,\|\Phi\|_{L_T^{p,q}}, \tag{4.13}$$

$$\left|\int_0^T \mathbb{K}[\Phi(t)]\,dt\right| \leq \kappa\,T^{\frac{3}{q}-\frac{1}{2}}\,\|V\|_{L^d}\,\|\Phi\|_{L_T^{\infty,2}}^2\,\|\Phi\|_{L_T^{p,q}}^2, \tag{4.14}$$

$$\|\mathbb{W}[C,\Phi]\,\Phi\|_{L_T^{p',q'}} \leq \kappa\,T^{\frac{3}{q}-\frac{1}{2}}\,\|V\|_{L^d}\,\|C\|_{\mathbb{C}^r}^2\,\|\Phi\|_{L_T^{\infty,2}}^2\,\|\Phi\|_{L_T^{p,q}}, \tag{4.15}$$

$$\|\mathbf{P}_\Phi\,\mathbb{W}[C,\Phi]\,\Phi\|_{L_T^{1,2}} \leq \kappa\,T^{\frac{3}{q}-\frac{1}{2}}\,\|V\|_{L^d}\,\|\Phi\|_{L_T^{\infty,2}}^3\,\|\Phi\|_{L_T^{p,q}}^2. \tag{4.16}$$

Proof. The proof is obvious and relies on the estimates (4.10-4.12) of the Lemma 4.2. The assertion (4.13) is easy, in fact one set $(p_1,q_1)=(p,q)\in\mathscr{A}$ and get the result. Next, given a potential $V\in L^d$, the matrix $\mathbb{K}$ involves elements of type $D_V(\phi_i\overline{\phi}_j,\phi_k\overline{\phi}_l)$ for which one use the estimate (4.12) by setting, for instance, $(p_3,q_3)=(p_4,q_4)=(\infty,2)\in\mathscr{A}$ and $(p_1,q_1)=(p_2,q_2)=(p,q)\in\mathscr{A}$. Also, the vector $\mathbb{W}[C,\Phi]\,\Phi$ involves terms of type $\mathbb{V}[\Phi]_{i,j}\phi_k$ that can be handled using the estimate (4.10). In fact, we set for instance $(p_2,q_2)=(p_3,q_3)=(\infty,2)\in\mathscr{A}$ and $(p_3,q_3)=(p,q)\in\mathscr{A}$. Finally, the vector $\mathbf{P}_\Phi\,\mathbb{W}[C,\Phi]\,\Phi$ involves terms of type $D_V(\phi_i\overline{\phi}_j,\phi_k\overline{\phi}_l)\,\phi_l$. In particular we observe that since $D_V(\phi_i\overline{\phi}_j,\phi_p\overline{\phi}_l)$ is only time-dependent scalar. Thus, estimating the left hand side (L.H.S.) in an $L_T^{p',q'}$ leads automatically to an $L_x^{q'}$ norm on ϕ_l in the right hand side (R.H.S.). For convenience, we estimate this term in $L_T^{1,2}$ in order to get an $\|\phi_l\|_{L_T^{\infty,2}}$ in the R.H.S. and use the same choice as in (4.14). □

Remark 4.2. First observe that for $d>\frac{3}{2}$, we have obviously $2\le q=\frac{2d}{d-1}<6$. Moreover, the estimates of the Corollary 4.1 involve T^α with power $\alpha>0$ so that $T^\alpha\to 0$ as $T\to 0$. Indeed, since $2\le q<6$, we have $0<\frac{3}{q}-\frac{1}{2}\le 1$. This observation will be crucial in the sequel. Form the opposite side, assume V bounded, that is $d=\infty$. Then, the estimates of the Corollary 4.1 are valid with $q=2$ and α, the power of T, is equal to 1.

Next, given $T>0$ and $(p,q)\in\mathscr{A}$, we define

$$\mathscr{Z}_T^{p,q}=L_T^{\infty,2}\cap L_T^{p,q},\quad \mathscr{X}_T^{p,q}=\mathbb{C}^r\times\mathscr{Z}_T^{p,q},$$

endowed with the norms

$$\|\phi\|_{\mathscr{Z}_T^{p,q}}=\|\phi\|_{L_T^{2,\infty}}+\|\phi\|_{L_T^{p,q}},\quad \|C,\Phi\|_{\mathscr{X}_T^{p,q}}=\|C\|_{\mathbb{C}^r}+\|\Phi\|_{\mathscr{Z}_T^{p,q}}.$$

A topology on $\mathscr{Z}_T^{p,q}$ and $\mathscr{X}_T^{p,q}$ being defined, we are able to prove the following

Lemma 4.3. *Let $U,V\in L^d, d>\frac{3}{2},(p,q)\in\mathscr{A}$ such that $q=\frac{2d}{d-1}$. Then, for all $T>0$, we have*

$$\|\mathbb{K}[\Phi(t)]\,C(t)-\mathbb{K}[\Phi'(t)]\,C'(t)\|_{L^1([0,T])}\le \mathrm{const}_1\,T^{\frac{3}{q}-\frac{1}{2}}\,\|(C,\Phi)-(C',\Phi')\|_{\mathscr{X}_T^{p,q}},\quad (4.17)$$

$$\|U\,(\Phi-\Phi')\|_{L_T^{p',q'}}\le \kappa\,T^{\frac{3}{q}-\frac{1}{2}}\,\|U\|_{L^d}\,\|\Phi-\Phi'\|_{\mathscr{Z}_T^{p,q}},\quad (4.18)$$

$$\|\mathbb{W}[C,\Phi]\,\Phi-\mathbb{W}[C',\Phi']\,\Phi'\|_{L_T^{p',q'}}\le \mathrm{const}_2\,T^{\frac{3}{q}-\frac{1}{2}}\,\|(C,\Phi)-(C',\Phi')\|_{\mathscr{X}_T^{p,q}},\quad (4.19)$$

$$\|\mathbf{P}_\Phi\,\mathbb{W}[C,\Phi]\,\Phi-\mathbf{P}_{\Phi'}\,\mathbb{W}[C',\Phi']\,\Phi'\|_{L_T^{1,2}}\le \mathrm{const}_3\,T^{\frac{3}{q}-\frac{1}{2}}\,\|(C,\Phi)-(C',\Phi')\|_{\mathscr{X}_T^{p,q}},\quad (4.20)$$

with $\mathrm{const}_1,\mathrm{const}_2$ *and* const_3 *depending on*

$$N,K,\|U\|_{L^d},\|V\|_{L^d},\|\Phi\|_{L_T^{\infty,2}},\|\Phi'\|_{L_T^{\infty,2}},\|C\|_{\mathbb{C}^r},\|C'\|_{\mathbb{C}^r},\|\Phi\|_{L_T^{p,q}}\ \text{and}\ \|\Phi'\|_{L_T^{p,q}}.$$

Proof. For a given real potential V, observe that the difference $\mathbb{K}[\Phi]-\mathbb{K}[\Phi']$ involves terms of type $D_V(\phi_i\,\overline{\phi}_j,\phi_k\,\overline{\phi}_l)-D_V(\phi_i'\,\overline{\phi'}_j,\phi_k'\,\overline{\phi'}_l)$ that we estimate as follows

$$\begin{aligned}\left|D_V(\phi_i\,\overline{\phi}_j,\phi_k\,\overline{\phi}_l)-D_V(\phi_i'\,\overline{\phi'}_j,\phi_k'\,\overline{\phi'}_l)\right| &\le \left|D_V([\phi_i-\phi_i']\,\overline{\phi}_j,\phi_k\,\overline{\phi}_l)\right|+\left|D_V(\phi_i'\,[\overline{\phi}_j-\overline{\phi'}_j],\phi_k\,\overline{\phi}_l)\right|\\ &+ \left|D_V(\phi_i'\,\overline{\phi'}_j,[\phi_k-\phi_k']\,\overline{\phi}_l)\right|+\left|D_V(\phi_i'\,\overline{\phi'}_j,\phi_k'\,[\overline{\phi}_l-\overline{\phi'}_l])\right|.\end{aligned}$$

Following (4.14), we have

$$\begin{aligned}\|\mathbb{K}[\Phi]\,C-\mathbb{K}[\Phi']\,C'\|_{L^1([0,T])} &\leq \|\mathbb{K}[\Phi]\|_{L^1([0,T])}\,\|C-C'\|_{\mathbb{C}^r}+\|\mathbb{K}[\Phi]-\mathbb{K}[\Phi']\|_{L^1([0,T])}\|C'\|_{\mathbb{C}^r},\\ &\leq \kappa T^{\frac{3}{q}-\frac{1}{2}}\,\|V\|_{L^d}\,\|\Phi\|^2_{L_T^{\infty,2}}\,\|\Phi\|^2_{L_T^{p,q}}\,\|C-C'\|_{\mathbb{C}^r}\\ &+\kappa\,T^{\frac{3}{q}-\frac{1}{2}}\,\|V\|_{L^d}\,\|C'\|_{\mathbb{C}^r}\,\left[\|\Phi\|_{L_T^{\infty,2}}\|\Phi\|^2_{L_T^{p,q}}+\|\Phi'\|_{L_T^{\infty,2}}\|\Phi'\|^2_{L_T^{p,q}}\right]\|\Phi-\Phi'\|_{L_T^{\infty,2}}\\ &+\kappa\,T^{\frac{3}{q}-\frac{1}{2}}\,\|V\|_{L^d}\|C'\|_{\mathbb{C}^r}\,\left[\|\Phi\|^2_{L_T^{\infty,2}}\|\Phi'\|_{L_T^{p,q}}+\|\Phi'\|^2_{L_T^{\infty,2}}\|\Phi\|_{L_T^{p,q}}\right]\|\Phi-\Phi'\|^2_{L_T^{p,q}}.\end{aligned}$$

The estimate (4.17) is then proved by setting for istance

$$\begin{aligned}\text{const}_1 &= \kappa\|V\|_{L^d}\|\Phi\|^2_{L_T^{\infty,2}}\,\|\Phi\|^2_{L_T^{p,q}}+\kappa\,\|V\|_{L^d}\,\|C'\|_{\mathbb{C}^r}\,\left[\|\Phi\|_{L_T^{\infty,2}}\|\Phi\|^2_{L_T^{p,q}}+\|\Phi'\|_{L_T^{\infty,2}}\|\Phi'\|^2_{L_T^{p,q}}\right]\\ &+ \kappa\|V\|_{L^d}\|C'\|_{\mathbb{C}^r}\,\left[\|\Phi\|^2_{L_T^{\infty,2}}\|\Phi'\|_{L_T^{p,q}}+\|\Phi'\|^2_{L_T^{\infty,2}}\|\Phi\|_{L_T^{p,q}}\right].\end{aligned}$$

Thanks to (4.13), the inequality (4.18) is straightforward. Now, from the one hand

$$\begin{aligned}\|\mathbb{W}[C,\Phi]\,\Phi-\mathbb{W}[C',\Phi']\,\Phi'\|_{L_T^{p',q'}} &\leq \|\mathbb{W}[C,\Phi]\,(\Phi-\Phi')\|_{L_T^{p',q'}}\\ &+ \|\mathbb{W}[C-C',\Phi]\,\Phi'\|_{L_T^{p',q'}}\\ &+ \|\mathbb{W}[C',\Phi-\Phi']\,\Phi'\|_{L_T^{p',q'}}.\end{aligned}$$

Observe that $\mathbb{W}[C,\Phi]$ is quadratic in C and Φ. Then, by (4.15), we obtain

$$\begin{aligned}\|\mathbb{W}[C,\Phi]\,\Phi-\mathbb{W}[C',\Phi']\,\Phi'\|_{L_T^{p',q'}} &\leq \kappa\,T^{\frac{3}{q}-\frac{1}{2}}\,\|V\|_{L^d}\|C\|^2_{\mathbb{C}^r}\|\Phi\|^2_{L_T^{\infty,2}}\|\Phi-\Phi'\|_{L_T^{p,q}}\\ &+ \kappa\,T^{\frac{3}{q}-\frac{1}{2}}\,\|V\|_{L^d}\left[\|C\|_{\mathbb{C}^r}+\|C'\|_{\mathbb{C}^r}\right]\|\Phi\|^2_{L_T^{\infty,2}}\|\Phi'\|_{L_T^{p,q}}\|C-C'\|_{\mathbb{C}^r}\\ &+ \kappa\,T^{\frac{3}{q}-\frac{1}{2}}\,\|V\|_{L^d}\|C'\|^2_{\mathbb{C}^r}\left[\|\Phi\|_{L_T^{\infty,2}}+\|\Phi'\|_{L_T^{\infty,2}}\right]\|\Phi'\|_{L_T^{p,q}}\|\Phi-\Phi\|_{L_T^{\infty,2}}.\end{aligned}$$

Thus, one set for instance

$$\begin{aligned}\text{const}_2 &= \kappa\|V\|_{L^d}\left[\|C\|^2_{\mathbb{C}^r}\|\Phi\|^2_{L_T^{\infty,2}}+\|C'\|^2_{\mathbb{C}^r}\left[\|\Phi\|_{L_T^{\infty,2}}+\|\Phi'\|_{L_T^{\infty,2}}\right]\|\Phi'\|_{L_T^{p,q}}\right]\\ &+ \kappa\|V\|_{L^d}\left[\|C\|_{\mathbb{C}^r}+\|C'\|_{\mathbb{C}^r}\right]\|\Phi\|^2_{L_T^{\infty,2}}\|\Phi'\|_{L_T^{p,q}}.\end{aligned}$$

It remains to estimate the projection part in $L_T^{1,2}$. For that purpose, we estimate first $[\mathbf{P}_\Phi-\mathbf{P}_{\Phi'}]\,\xi$ in $L_T^{1,2}$ for a given function $\xi(t,x)\in L_T^{p',q'}$ for all $(p,q)\in\mathscr{A}$. This can be achieved thanks to Hölder inequality in space and time as follows

$$\begin{aligned}\|[\mathbf{P}_\Phi-\mathbf{P}_{\Phi'}]\,\xi\|_{L_T^{1,2}} &\leq \sum_{k=1}^{K}\left[\|\langle\xi,\phi_k\rangle\|_{L^1([0,T])}\,\|\phi_k-\phi_k'\|_{L_T^{\infty,2}}+\|\langle\xi,\phi_k-\phi_k'\rangle\|_{L^1([0,T])}\,\|\phi_k'\|_{L_T^{\infty,2}}\right],\\ &\leq \kappa\|\xi\|_{L_T^{p',q'}}\|\Phi\|_{L_T^{p,q}}\,\|\Phi-\Phi'\|_{L_T^{\infty,2}}+\kappa\|\xi\|_{L_T^{p',q'}}\|\Phi'\|_{L_T^{\infty,2}}\,\|\Phi-\Phi'\|_{L_T^{p,q}}. \quad (4.21)\end{aligned}$$

Now, we have

$$\begin{aligned}\|\mathbf{P}_\Phi\,\mathbb{W}[C,\Phi]\,\Phi-\mathbf{P}_{\Phi'}\,\mathbb{W}[C',\Phi']\,\Phi'\|_{L_T^{1,2}} &\leq \|\mathbf{P}_\Phi\,[\mathbb{W}[C,\Phi]\,\Phi-\mathbb{W}[C',\Phi']\,\Phi'\|_{L_T^{1,2}}\\ &+ \|[\mathbf{P}_\Phi-\mathbf{P}_{\Phi'}]\,\mathbb{W}[C',\Phi']\,\Phi'\|_{L_T^{1,2}},\\ &\leq \kappa\|\mathbb{W}[C,\Phi]\,\Phi-\mathbb{W}[C',\Phi']\,\Phi'\|_{L_T^{p',q'}}\|\Phi\|_{L_T^{p,q}}\|\Phi\|_{L_T^{\infty,2}}\\ &+ \kappa\|\mathbb{W}[C',\Phi']\,\Phi'\|_{L_T^{p',q'}}[\|\Phi\|_{L_T^{p,q}}+\|\Phi'\|_{L_T^{\infty,2}}]\|\Phi-\Phi'\|_{\mathscr{Z}_T^{p,q}}.\end{aligned}$$

The estimate above is an application of (4.21), first with $\Phi' \equiv 0$ and $\xi \equiv \mathbb{W}[C,\Phi]\,\Phi - \mathbb{W}[C',\Phi']\,\Phi'$, second with $\xi \equiv \mathbb{W}[C',\Phi']\,\Phi'$. Finally, Let $q = \frac{2d}{d-1}$ and recall that $V \in L^d$ with $d > \frac{3}{2}$. Then following (4.19), we get the desired estimate by setting for instance

$$\text{const}_3 \quad = \quad \text{const}_2\,\|\Phi\|_{L^{p,q}_T}\|\Phi\|_{L^{\infty,2}_T} + \kappa\|V\|_{L^d}\|C'\|^2_{\mathbb{C}^r}\|\Phi'\|_{L^{p,q}_T}\|\Phi'\|^2_{L^{\infty,2}_T}[\|\Phi\|_{L^{p,q}_T} + \|\Phi'\|_{L^{\infty,2}_T}],$$

which achieves the proof. □

Now, let $R,T>0$ be arbitrary reals to be fixed later on and let $(C^0,\Phi^0) \in \mathscr{F}_{N,K}$. Now, introduce the ball

$$\tilde{\mathscr{X}}^{p,q}_T(R) = \Big\{(C,\Phi) \in \mathscr{X}^{p,q}_T : \quad \|C,\Phi\|_{\mathscr{X}^{p,q}_T} \leq R\Big\}. \tag{4.22}$$

This defines a complete metric space equipped with the distance induced by the norm of $\mathscr{X}^{p,q}_T$. Finally, introduce the following mapping

$$\pi_{C^0,\Phi^0} : \begin{bmatrix} C(\cdot) \\ \\ \Phi(\cdot) \end{bmatrix} \mapsto \begin{bmatrix} C^0 \\ \\ \mathbf{U}(\cdot)\,\Phi^0 \end{bmatrix} - i\int_0^{\cdot} \begin{bmatrix} \mathbb{K}[\Phi(s)]\,C(s) \\ \\ \mathbf{U}(\cdot - s)\,\Big[U\,\Phi(s) + \mathbb{L}[C(s),\Phi(s)]\Big] \end{bmatrix} ds, \tag{4.23}$$

with

$$\mathbb{L}[C,\Phi] := \mathbf{\Gamma}[C]^{-1}\,(I - \mathbf{P}_\Phi)\,\mathbb{W}[C,\Phi]\,\Phi.$$

This formulation is then well defined as long as the matrix $\mathbf{\Gamma}[C(t)]$ is invertible. From now on, we will consider initial data $(C^0,\Phi^0) \in \partial\mathscr{F}_{N,K}$, that is the associated first order density operator is of full rank. Thus, the quadratic dependence of $\mathbf{\Gamma}[C]$ on the coefficients C_σ and the continuity of the MCTDHf flow guarantees that this property will be propagated up to a certain time $T^\star$, at least infinitesimal but also possibly infinite. That is, $\mathbf{\Gamma}[C(t)]$ is of rank K for all $t \in [0,T^\star)$, hence invertible and we refer the reader to [1] for more details on this point. From now on we accept the existence of $T^\star$ and we postpone the proof of such fact to [1].

Now, we claim the following

Lemma 4.4. *Let $U,V \in L^d, d > \frac{3}{2}, (p,q = \frac{2d}{d-1}) \in \mathscr{A}$ and $(C^0,\Phi^0) \in \partial\mathscr{F}_{N,K}$ with $\|C^0,\Phi^0\|_{\mathbb{C}^r\times L^2} \leq \beta$. Then, there exist a radius $R>0$ and a time T with $0<T<T^\star$ such that the mapping π is a strict contraction on $\tilde{\mathscr{X}}^{p,q}_T(R)$. Moreover, given $(C'^0,\Phi'^0) \in \partial\mathscr{F}_{N,K}$ with $\|C'^0,\Phi'^0\|_{\mathbb{C}^r\times L^2} \leq \beta$,then*

$$\|(C,\Phi) - (C',\Phi')\|_{\mathscr{X}^{p,q}_T} \leq \text{const}\,\|(C^0,\Phi^0) - (C'^0,\Phi'^0)\|_{\mathbb{C}^r\times L^2}, \tag{4.24}$$

where (C,Φ) and (C',Φ') denote the fixed points of π associated with (C^0,Φ^0) and (C'^0,Φ'^0) respectively.

Proof. The proof is based on the Lemma 4.3. By abuse of notation, $\pi_{C^0,\Phi^0}(C,\Phi)$ will be used instead of the vertical notation (4.23). Moreover for notation's lightness, we set

$$(S\phi)(t) := \int_0^t \mathbf{U}(t-s)\,\phi(s)\,ds.$$

Next, let $(a,b) \in \mathscr{A}$, $(C,\Phi),(C',\Phi') \in \mathscr{X}_T^{p,q}$, $T>0$ to be fixed later on and $t \in [0,T]$. Finally we set $(p=\frac{4d}{3}, q=\frac{2d}{d-1}) \in \mathscr{A}$. Then

$$\begin{aligned}
\|\pi_{C^0,\Phi^0}(C(t),\Phi(t)) \quad - \quad & \pi_{C'^0,\Phi'^0}(C'(t),\Phi'(t))\|_{\mathbb{C}^r \times L_T^{a,b}} \leq \\
& (1+\rho(a))\|(C^0,\Phi^0)-(C'^0,\Phi'^0)\|_{\mathbb{C}^r \times L^2} \qquad (4.25)\\
+ \quad & \| \int_0^t \left[\mathbb{K}[\Phi(s)]\,C(s) - \mathbb{K}[\Phi'(s)]\,C'(s)\right] ds\|_{\mathbb{C}^r} + \|S\left(U\,\Phi - U\,\Phi'\right)(t)\|_{L_T^{a,b}} \\
+ \quad & \|S\left(\mathbb{L}[C,\Phi]-\mathbb{L}[C',\Phi']\right)(t)\|_{L_T^{a,b}} := \mathscr{T}_1 + \ldots + \mathscr{T}_4.
\end{aligned}$$

The term $\mathscr{T}_1$ is due to the Lemma 4.1. More precisely, the first assertion of (4.7). Next, thanks to the inequality (4.17) of Lemma 4.3, we have

$$\mathscr{T}_2 \leq \mathrm{const}_1\, T^{\frac{3}{q}-\frac{1}{2}}\, \|(C,\Phi)-(C',\Phi')\|_{\mathscr{X}_T^{p,q}}. \qquad (4.26)$$

Now, we shall make use of the second assertion of (4.7) in order to estimate $\mathscr{T}_3$ and $\mathscr{T}_4$. We start with

$$\begin{aligned}
\mathscr{T}_3 \quad &\leq \quad \rho(a,p)\,\|U\,\Phi - U\,\Phi'\|_{L_T^{p',q'}}, \\
&\leq \quad \rho(a,p)\,\kappa\, T^{\frac{3}{q}-\frac{1}{2}} \|U\|_{L^d}\, \|(C,\Phi)-(C',\Phi')\|_{\mathscr{X}_T^{p,q}}. \qquad (4.27)
\end{aligned}$$

The second line above is due (4.18) and we upper bounded obviously the $\mathscr{Z}_T^{p,q}$ norm by the $\mathscr{X}_T^{p,q}$one. Finally

$$\begin{aligned}
\mathscr{T}_4 \quad &\leq \quad \|S\left(\mathbf{\Gamma}[C]^{-1}\mathbb{W}[C,\Phi]\,\Phi - \mathbf{\Gamma}[C']^{-1}\mathbb{W}[C',\Phi']\,\Phi'\right)(t)\|_{L_T^{a,b}} \\
&+ \quad \|S\left(\mathbf{P}_\Phi\mathbf{\Gamma}[C]^{-1}\mathbb{W}[C,\Phi]\,\Phi - \mathbf{P}_{\Phi'}\mathbf{\Gamma}[C']^{-1}\mathbb{W}[C',\Phi']\,\Phi'\right)(t)\|_{L_T^{a,b}}, \\
&\leq \quad \rho(a,p)\,\|\mathbf{\Gamma}[C]^{-1}\mathbb{W}[C,\Phi]\,\Phi - \mathbf{\Gamma}[C']^{-1}\mathbb{W}[C',\Phi']\,\Phi'\|_{L_T^{p',q'}} \\
&+ \quad \rho(a,p)\,\|\mathbf{P}_\Phi\mathbf{\Gamma}[C]^{-1}\mathbb{W}[C,\Phi]\,\Phi - \mathbf{P}_{\Phi'}\mathbf{\Gamma}[C']^{-1}\mathbb{W}[C',\Phi']\,\Phi'\|_{L_T^{1,2}}.
\end{aligned}$$

Next, observe the trivial algebraic relation

$$\begin{aligned}
\|\mathbf{\Gamma}[C]^{-1}-\mathbf{\Gamma}[C']^{-1}\| \quad &\leq \quad \|\mathbf{\Gamma}[C]^{-1}(\mathbf{\Gamma}[C']-\mathbf{\Gamma}[C])\mathbf{\Gamma}[C']^{-1}\| \\
&\leq \quad \kappa\,\|\mathbf{\Gamma}[C]^{-1}\|\|\mathbf{\Gamma}[C']^{-1}\|(\|C\|+\|C'\|)\|C-C'\|_{\mathbb{C}^r}.
\end{aligned}$$

Thus, by the mean of (4.15,4.16,4.19,4.20), we get

$$\mathscr{T}_4 \quad \leq \quad \rho(a,p)\,\mathrm{const}_4\, T^{\frac{3}{q}-\frac{1}{2}}\, \|(C,\Phi)-(C',\Phi')\|_{\mathscr{X}_T^{p,q}}. \qquad (4.28)$$

More precisely

$$\begin{aligned}
\mathrm{const}_4 \quad = \quad & \kappa\|V\|_{L^d}\|\mathbf{\Gamma}[C]^{-1}\|\|\mathbf{\Gamma}[C']^{-1}\|\|C'\|_{\mathbb{C}^r}^2[\|C\|_{\mathbb{C}^r}+\|C'\|_{\mathbb{C}^r}][1+\|\Phi'\|_{L_T^{\infty,2}}\|\Phi'\|_{L_T^{p,q}}] \times \\
\times \quad & \|\Phi'\|_{L_T^{\infty,2}}^2\|\Phi'\|_{L_T^{p,q}} + \|\mathbf{\Gamma}[C]^{-1}\|[\mathrm{const}_2+\mathrm{const}_3].
\end{aligned}$$

Summing (4.26-4.28) and add the sum to the first line of (4.25) leads to

$$\|\pi_{C^0,\Phi^0}(C(t),\Phi(t))-\pi_{C'^0,\Phi'^0}(C'(t),\Phi'(t))\|_{\mathbb{C}^r\times L^{a,b}_T}\leq(1+\rho(a))\|(C^0,\Phi^0)-(C'^0,\Phi'^0)\|_{\mathbb{C}^r\times L^2}$$
$$+\quad\left[\text{const}_1+\rho(a,p)\,\text{const}_4+\rho(a,p)\,\kappa\|U\|_{L^d}\right]T^{\frac{3}{q}-\frac{1}{2}}\|(C,\Phi)-(C',\Phi')\|_{\mathscr{X}^{p,q}_T}. \quad (4.29)$$

The inequality (4.29) being true for any admissible pair $(a,b)\in\mathscr{A}$. Therefore, we write it in the special case $(a,b)=(p,q)\in\mathscr{A}$ and then in the case $(a,b)=(\infty,2)\in\mathscr{A}$. Moreover, we set $(C^0,\Phi^0)\equiv(C'^0,\Phi'^0)$ and use $\|C,\Phi\|_{\mathscr{X}^{p,q}_T},\|C',\Phi'\|_{\mathscr{X}^{p,q}_T}\leq R$ since $(C,\Phi),(C',\Phi')\in\tilde{\mathscr{X}}^{p,q}_T(R)$. Recall that $T<T^\star$ since the initial data is in $\partial\mathscr{F}_{N,K}$, that is the matrix $\mathbf{\Gamma}[C]$ and $\mathbf{\Gamma}[C']$ are invertible thus there exists $\theta>0$ such that $\|\mathbf{\Gamma}[C]^{-1}\|,\|\mathbf{\Gamma}[C']^{-1}\|\leq\theta$. This procedure allows to write

$$\begin{aligned}\|\pi_{C^0,\Phi^0}(C(t),\Phi(t))-\pi_{C^0,\Phi^0}(C'(t),\Phi'(t))\|_{\mathscr{X}^{p,q}_T}&\leq\text{const}_5(\kappa,R,\theta,p)\,T^{\frac{3}{q}-\frac{1}{2}}\times\\&\times\quad\|(C,\Phi)-(C',\Phi')\|_{\mathscr{X}^{p,q}_T}. \quad (4.30)\end{aligned}$$

Equivalently, if we set $(C',\Phi')\equiv(0,0)$ and use the fact that $\|C^0,\Phi^0\|_{\mathbb{C}^r\times L^2}\leq\beta$. Again, after summation

$$\|\pi_{C^0,\Phi^0}(C(t),\Phi(t))\|_{\mathscr{X}^{p,q}_T}\leq\text{const}_6(\kappa,R,\theta,\beta,p)\,T^{\frac{3}{q}-\frac{1}{2}}. \quad (4.31)$$

More precisely

$$\begin{aligned}\text{const}_5&=2\kappa R^4\|V\|_{L^d}[5+\rho(p)(2\theta^2R^4+(2\theta+7)\theta R^2+5\theta)]+\rho(p)\kappa\|U\|_{L^d},\\\text{const}_6&=2(1+\rho(p))+2\kappa R^5[\|V\|_{L^d}+\rho(p)\theta\|V\|_{L^d}(1+R^2)]+2R\rho(p)\kappa\|U\|_{L^d}.\end{aligned}$$

Thus, we choose R and T such that

$$T<\inf\left\{T^\star,\left[\frac{R}{\text{const}_6}\right]^{\frac{2q}{6-q}}\right\},\quad R\,\text{const}_5-\text{const}_6<0. \quad (4.32)$$

That is

$$\text{const}_6\,T^{\frac{3}{q}-\frac{1}{2}}<R\quad\text{and}\quad\text{const}_5\,T^{\frac{3}{q}-\frac{1}{2}}<1. \quad (4.33)$$

Thus, by (4.30, 4.31), π_{C^0,Φ^0} is a strict contracting map on $\tilde{\mathscr{X}}^{p,q}_T$.

It remains to prove the continuous dependence on the initial data (4.24). Again, the essence is the inequality (4.29). Let (C,Φ) and (C',Φ') be the fixed points of π_{C^0,Φ^0} and $\pi_{C'^0,\Phi'^0}$ respectively. Let us write again the inequality for $(a,b)=(p,q)$ and then in the case $(a,b)=(\infty,2)$ and sum up. One obtain

$$\begin{aligned}&\|\pi_{C^0,\Phi^0}(C(t),\Phi(t))-\pi_{C'^0,\Phi'^0}(C'(t),\Phi'(t))\|_{\mathscr{X}^{p,q}_T}=\|(C,\Phi)-(C',\Phi')\|_{\mathscr{X}^{p,q}_T}\\&\quad\leq2(1+\rho(p))\|(C^0,\Phi^0)-(C'^0,\Phi'^0)\|_{\mathbb{C}^r\times L^2}+\text{const}_5(\kappa,R,\theta,p)\,T^{\frac{3}{q}-\frac{1}{2}}\|(C,\Phi)-(C',\Phi')\|_{\mathscr{X}^{p,q}_T}.\end{aligned}$$

Finally, the estimate above with (4.33) proves the continuous dependence on the initial data (4.24). □

Now, we are able to claim

Proposition 4.1. *Let $(C^0,\Phi^0)\in\partial\mathscr{F}_{N,K},U,V\in L^d(\mathbb{R}^3)$ with $d>\frac{3}{2}$ and $(p,q=\frac{2d}{d-1})\in\mathscr{A}$. Then there exists a $T(p,\|C\|_{\mathbb{C}^r},\|\Phi\|_{L^2},\|\Phi^0\|_{L^2})>0$ and a unique solution $(C(t),\Phi(t))\in\mathscr{X}_T^{p,q}$ to*

$$\begin{bmatrix}C(t)\\ \\ \Phi(t)\end{bmatrix}=\begin{bmatrix}C^0\\ \\ \mathbf{U}(t)\,\Phi^0\end{bmatrix}-i\int_0^t\begin{bmatrix}\mathbb{K}[\Phi(s)]\,C(s)\\ \\ \mathbf{U}(t-s)\,\Big[U\,\Phi(s)+\mathbb{L}[C(s),\Phi(s)]\Big]\end{bmatrix}ds. \tag{4.34}$$

In particular, the functional part of the solution satisfies

$$\|\Phi\|_{L_T^{p,q}}\leq 2\,\rho(p)\,\|\Phi^0\|_{L^2}. \tag{4.35}$$

Proof. Thanks to the Lemma 4.4, there exists a $T'>0$ for which the integral formulation (4.34) associated to the MCTDHF admits a unique solution $(C(t),\Phi(t))\in\mathscr{X}_{T'}^{p,q}$. The main point then is to characterize $0<T\leq T'$ for which the property (4.35) holds. The functional part of the solution satisfies

$$\Phi(t)=\mathbf{U}(t)\Phi^0-i\int_0^t\mathbf{U}(t-s)\,\mathbb{L}[C(s),\Phi(s)]\,ds$$

Let $0<\tau\leq T\leq T'$ where T is to be fixed later on and recall that $\|\mathbf{\Gamma}[C(t)]^{-1}\|\leq\theta$ for all $0\leq t\leq T^\star$. Next, we estimate the $L_\tau^{p,q}$ of the R.H.S. and get using the estimate (4.7) of Lemma 4.1.

$$\begin{aligned}
\|\Phi\|_{L_\tau^{p,q}} &\leq \rho(p)\,\|\Phi^0\|_{L^2}+\|\int_0^t\mathbf{U}(t-s)U\,\Phi(s)\,ds\|_{L_\tau^{p,q}}\\
&+ \|\int_0^t\mathbf{U}(t-s)\mathbf{\Gamma}[C(s)]^{-1}\,\mathbb{W}[C(s),\Phi(s)]\,\Phi(s)\,ds\|_{L_\tau^{p,q}}\\
&+ \|\int_0^t\mathbf{U}(t-s)\mathbf{\Gamma}[C(s)]^{-1}\,\mathbf{P}_{\Phi(s)}\mathbb{W}[C(s),\Phi(s)]\,\Phi(s)\,ds\|_{L_\tau^{p,q}},\\
&\leq \rho(p)\,\|\Phi^0\|_{L^2}+\|U\,\Phi\|_{L_\tau^{p',q'}}+\|\mathbf{\Gamma}[C]^{-1}\|\,\|\mathbb{W}[C,\Phi]\,\Phi\|_{L_\tau^{p',q'}}\\
&+ \|\mathbf{\Gamma}[C]^{-1}\|\,\|\mathbf{P}_\Phi\,\mathbb{W}[C,\Phi]\,\Phi\|_{L_\tau^{1,2}},\\
&\leq \rho(p)\,\|\Phi^0\|_{L^2}+\kappa\,\theta\,T^{\frac{3}{q}-\frac{1}{2}}\,\|U\|_{L^d}\,\|\Phi\|_{L_\tau^{p,q}}+\kappa\,\theta\,T^{\frac{3}{q}-\frac{1}{2}}\,\|V\|_{L^d}\,\|C\|_{\mathbb{C}^r}^2\,\|\Phi\|_{L_\tau^{\infty,2}}^2\|\Phi\|_{L_\tau^{p,q}}\\
&+ \kappa\,\theta\,T^{\frac{3}{q}-\frac{1}{2}}\,\|V\|_{L^d}\,\|C\|_{\mathbb{C}^r}^2\,\|\Phi\|_{L_\tau^{\infty,2}}^3\,\|\Phi\|_{L_\tau^{p,q}}^2.
\end{aligned}\tag{4.36}$$

Next, we follow the argument of Tsutsumi in [21]. That is, we choose T so small so that there exists a positive number η satisfying

$$\begin{cases}
f(\eta,T):=\rho(p)\,\|\Phi^0\|_{L^2}-\eta+\kappa\,\theta\,\eta\,T^{\frac{3}{q}-\frac{1}{2}}\Big(\|U\|_{L^d}+\|V\|_{L^d}\|C\|_{\mathbb{C}^r}^2\,\|\Phi\|_{L_\tau^{\infty,2}}^2\Big[1+\eta\,\|\Phi\|_{L_\tau^{\infty,2}}\Big]\Big)\\
\qquad\qquad<0,\\
\\
0<\eta\leq 2\,\rho(p)\,\|\Phi^0\|_{L^2}.
\end{cases}$$

For that purpose, it is sufficient ti choose $T>0$ so that

$$T<\inf\left\{\Big(2\,\kappa\,\theta\,\Big[\|U\|_{L^d}+\|V\|_{L^d}\|C\|_{\mathbb{C}^r}^2\,\|\Phi\|_{L_\tau^{\infty,2}}^2\Big[1+2\,\rho(p)\,\|\Phi^0\|_{L^2}\,\|\Phi\|_{L_\tau^{\infty,2}}\Big]\Big]\Big)^{\frac{2q}{q-6}},T'\right\}. \tag{4.37}$$

Then, let

$$\eta_0 = \min_{\eta} \left\{ 0 < \eta \leq 2\,\rho(p)\,\|\Phi^0\|_{L^2} \quad ; \quad f(\eta, T) = 0 \right\}. \tag{4.38}$$

Now, let $\mathscr{Y}(\tau) = \|\Phi\|_{L^{p,q}_\tau}$. Then, by (4.36), we have for $0 \leq \tau \leq T$

$$\begin{cases} \mathscr{Y}(\tau) \leq \rho(p)\,\|\Phi^0\|_{L^2} + \kappa\,\theta\,\mathscr{Y}(\tau)\,T^{\frac{3}{q}-\frac{1}{2}} \left(\|U\|_{L^d} + \|V\|_{L^d} \|C\|^2_{\mathbb{C}^r}\,\|\Phi\|^2_{L^{\infty,2}_\tau} \left[1 + \mathscr{Y}(\tau)\,\|\Phi\|_{L^{\infty,2}_\tau} \right] \right), \\ \\ \mathscr{Y}(\tau = 0) = 0. \end{cases} \tag{4.39}$$

Then, if T is chosen such that (4.37) holds, then by (4.38, 4.39), we get

$$\mathscr{Y}(\tau) \leq \eta_0 \leq 2\,\rho(p)\,\|\Phi^0\|_{L^2}, \quad \text{for all} \quad \tau \in [0, T].$$

Thus, passing to the limit as $\tau \to T$, we get by Fatou's Lemma

$$\|\Phi\|_{L^{p,q}_T} \leq 2\,\rho(p)\,\|\Phi^0\|_{L^2},$$

which is the desired inequality. □

4.4 proof of Theorem 4.2

In this section, we finish the proof of our main result, the Theorem 4.2. The main tools will be the proposition 4.1 and the Theorem 4.1 that ensure the existence and uniqueness of a solution, in short time, to the MCTDHF system in the energy space H^1.

Let $(C^0, \Phi^0) \in \partial\mathscr{F}_{N,K}$. By proposition 4.1, there exists $0 < T_0 < T^\star$ such that the integral formulation (4.34) admits a unique solution in the space $\mathscr{X}^{p,q}_{T_0}$. First of all, observe that for all $(p,q) \in \mathscr{A}$, we have the continuous embedding

$$L^{q'}(\mathbb{R}^3) \hookrightarrow H^{-1}(\mathbb{R}^3).$$

Thus, in view of (4.13,4.15,4.16), we get that $\Phi \in C([0,T_0], H^{-1})$, thus $\Phi \in C_w([0,T_0], L^2)$ where the subscript $_w$ stands for weak. Now, let $(C^{0,n}, \Phi^{0,n}) \in \partial\mathscr{F}_{N,K} \cap H^1(\mathbb{R}^3)^K$ be a sequence of initial data for $n \in \mathbb{N}$ such that

$$\|C^{0,n}, \Phi^{0,n}\|_{\mathbb{C}^r \times L^2} \leq \|C^0, \Phi^0\|_{\mathbb{C}^r \times L^2}, \quad (C^{0,n}, \Phi^{0,n}) \overset{n \to +\infty}{\longrightarrow} (C^0, \Phi^0) \quad \text{in} \quad \mathbb{C}^r \times L^2. \tag{4.40}$$

Consequently, by Theorem 4.1, we get for all $n \in \mathbb{N}$, the existence of a unique solution $(C^n(t), \Phi^n(t))$ for all $t \in [0, T^\star)$ such that

$$C^n \in C^1([0,T^\star), \mathbb{C})^r, \quad \Phi^n \in C^0([0,T^\star), H^1(\mathbb{R}^3))^K. \tag{4.41}$$

Moreover, for all $n \in \mathbb{N}$, we have the following conservation law

$$\|C^n(t)\|_{\mathbb{C}^r} = \|C^{0,n}\|_{\mathbb{C}^r}, \quad \|\Phi^n(t)\|_{L^2} = \|\Phi^{0,n}\|_{L^2}, \quad \forall\, t \in [0, T^\star).$$

Now, by (4.40), (C^0, Φ^0) and $(C^{0,n}, \Phi^{0,n})$ are fixed points of the applications π_{C^0,Φ^0} and $\pi_{C^{0,n},\Phi^{0,n}}$ respectively on the same closed ball of $\mathscr{X}^{p,q}_{T_0}$ for all $(p, 2 \leq q = \frac{2d}{d-1} < 6) \in \mathscr{A}$. Thus, one can

pass to the limit as $n \to +\infty$ using the continuous dependence on the initial data (4.24) and (4.40), we get

$$\|C(t)\|_{\mathbb{C}^r} = \|C^0\|_{\mathbb{C}^r}, \quad \|\Phi(t)\|_{L^2} = \|\Phi^0\|_{L^2}, \quad \text{for all} \quad 0 \leq t \leq T_0. \tag{4.42}$$

Hence, (4.42) with $\Phi \in C_w([0,T_0],L^2)$ leads to $\Phi \in C([0,T_0],L^2)$. Moreover, the fact that $C \in C^1([0,T_0],\mathbb{C})^r$ follows from the continuity of $t \mapsto \mathbb{K}[\Phi(t)]$ for all $t \in [0,T_0]$ which is a consequence of the continuity of the $\phi_i's$.

Now, recall that the functional part of this solution satisfies the *a priori* estimate (4.35). In particular, the time T_0 depends only on the constants $N,K,p,\|C^0\|_{\mathbb{C}^r}$ and $\|\Phi^0\|_{L^2}$. Thus, one can reiterate the argument with initial data $(C(T_0),\Phi(T_0)),(C(2T_0),\Phi(2T_0)),\dots$ up to $T^{\star-}$. We now check the uniqueness of the solution to the integral formulation associated to the MCTDHF, namely (5.79). The uniqueness on $[0,T_0]$ if given for free by the contraction argument. Again, one can reiterate up to $T^{\star-}$ and get the uniqueness on the hole labs $[0,T^\star)$. The points i and ii are straightforward. The last point to make clear before finishing the proof of the Theorem 4.2, is the equivalence between the MCTDHF as an integral formulation and in the distributional sense. For that purpose, we refer, for instance, to [5] (paragraph 4: Proof of Theorem 2.2, more precisely, the uniqueness part of the proof).

Recall that the Coulomb potential satisfies after cut-off $\frac{1}{|x|} \in L^d + L^\infty$ with $d \in \left[\frac{3}{2},3\right[$. Thus, for $p = \frac{2d}{d-1}$, we have obviously (omitting the case $d = \frac{3}{2}$) $3 < q < 6$ which is the result obtained by Castella in [5] for the SPS system. Moreover, notice that the estimates of the Lemma 4.3 hold true for bounded potentials. In particular in such case one can show that the mapping

$$\begin{bmatrix} C \\ \\ \Phi \end{bmatrix} \mapsto \begin{bmatrix} \mathbb{K}[\Phi]C \\ \\ U\,\Phi + \mathbf{\Gamma}[C]^{-1}\,(I-\mathbf{P}_\Phi)\,\mathbb{W}[C,\Phi]\,\Phi(s) \end{bmatrix} ds,$$

is locally lipschitz in $\mathbb{C}^r \times L^2$. This justifies in particular the restriction of the results obtained in [15] since in this configuration we need only to prove the preservation of $\mathscr{F}_{N,K}$ by the MCTDHF flow.

4.5 some comments and conclusion

For the time being, all the well-posedness results on the multiconfiguration models [15, 20, 1] hold up to a certain time $T^\star$, possibly infinite, for which $\operatorname{rank}(\mathbf{\Gamma}[C(T^\star)]) < K$. In [9], we obtained a sufficient condition that ensures the global well-posedness. More precisely, starting with initial data $(C^0,\Phi^0) \in \mathscr{F}_{N,K} \cap H^1(\mathbb{R}^3)^K$ satisfying

$$\left\langle \Psi_{\mathrm{MC}}(C^0,\Phi^0)|\mathscr{H}_N|\Psi_{\mathrm{MC}}(C^0,\Phi^0)\right\rangle_{L^2(\mathbb{R}^{3N})} < \min_{(C,\Phi)\in\mathscr{F}_{N,K-1}} \left\langle \Psi_{\mathrm{MC}}(C,\Phi)|\mathscr{H}_N|\Psi_{\mathrm{MC}}(C,\Phi)\right\rangle_{L^2(\mathbb{R}^{3N})},$$

then, the MCTDHF system $\mathscr{S}$ admits a unique global-in-time solution $(C(t),\Phi(t))$ in the class

$$C^1\big([0,+\infty);\mathbb{C}\big)^r \times \Big(C^1\big([0,+\infty);H^{-1}(\mathbb{R}^3)\big)^K \cap C^0\big([0,+\infty);H^1(\mathbb{R}^3)\big)^K\Big).$$

We refer the reader to [9, 1] for details about the proof. However, such condition is not valid for an L^2 theory since the energy of the initial data is not well-defined for $(C^0,\Phi^0) \in \mathscr{F}_{N,K}$. In order

to remediate to this problem, physicists and chemists perturb the density matrix [4, 18]. In order to illustrate their method we use, without loss of generality, the following perturbation

$$\mathbf{\Gamma}_\varepsilon[C] = \mathbf{\Gamma}[C] + \varepsilon I_K,$$

with I_K being the $K \times K$ identity matrix. The MCTDHF system $\mathscr{S}$ is then $\varepsilon-$dependent and reads

$$\mathscr{S}_\varepsilon \quad \begin{cases} i\frac{d}{dt} C(t) = \mathbb{K}[\Phi](t)\, C(t), \\ \\ i\,\mathbf{\Gamma}_\varepsilon[C(t)]\, \frac{\partial}{\partial t}\, \Phi(t,x) = \mathbf{\Gamma}_\varepsilon[C(t)]\, \left[-\frac{1}{2}\Delta_x + U(x)\right]\, \Phi(t,x) + (I - \mathbf{P}_\Phi)\, \mathbb{W}[C,\Phi](t,x)\, \Phi(t,x). \end{cases}$$

Then, observe that the L^2 theory we proved in the present paper is also global for $\mathscr{S}_\varepsilon$ since the time T of the Proposition 4.1 depends only on conserved quantities and one can iterate and cover the whole real line. Also, observe that $\mathscr{S}_\varepsilon$ conserves the normalization of the coefficient and the orbitals. However it doesn't conserve the total energy because of the perturbation. We are then able to claim the following corollary for $\mathscr{S}_\varepsilon$

Corollary 4.2. *Let $U, V \in L^d(\mathbb{R}^3)$ with $d > \frac{3}{2}$ and $(C^0, \Phi^0) \in \mathscr{F}_{N,K}$ be an initial data. Then, the perturbed MCTDHF system $\mathscr{S}_\varepsilon$ admits solutions $(C_\varepsilon(t), \Phi_\varepsilon(t))$ satisfying*

- $C_\varepsilon \in C^1([0,+\infty), \mathbb{C})^r \quad$ *and* $\quad \Phi_\varepsilon \in C^0([0,+\infty), L^2(\mathbb{R}^3))^K$.

Moreover,

i) $\Phi_\varepsilon(t) \in L^{\frac{4q}{3(q-2)}}_{loc}([0,+\infty), L^q(\mathbb{R}^3))^K$ *for all* $2 \le q < 6$

ii) *The solution $(C_\varepsilon(t), \Phi_\varepsilon(t))$ is unique in the class*

$$L^\infty([0,+\infty), \mathbb{C})^r \times L^\infty([0,+\infty), L^2(\mathbb{R}^3))^K \cap L^{\frac{4q}{3(q-2)}}_{loc}([0,+\infty), L^q(\mathbb{R}^3))^K,$$

for all $2 \le q < 6$.

iii) $(C_\varepsilon(t), \Phi_\varepsilon(t)) \in \mathscr{F}_{N,K}$ *for all* $t > 0$. *In other words,*

$$\|C_\varepsilon(t)\|_{\mathbb{C}^r} = \|C^0\|_{\mathbb{C}^r} \quad \textit{and} \qquad \|\Phi_\varepsilon(t)\|_{L^2} = \|\Phi^0\|_{L^2}.$$

However, a global H^1 theory on the system $\mathscr{S}_\varepsilon$ is not possible since the total energy is far from being conserved. This is due to the fact that $\mathbf{\Gamma}[C]\mathbf{\Gamma}_\varepsilon[C]^{-1} \neq I_K$ and we refer the reader to the first chapter for an explicit proof of the energy conservation. Moreover we don't believe that the perturbed energy

$$\left\langle \left(\left[\sum_{i=1}^N -\frac{1}{2}\Delta_{x_i} + U(x_i) \right] \mathbf{\Gamma}_\varepsilon[C] + \frac{1}{2}\mathbb{W}_{C,\Phi} \right) \Phi\,,\, \Phi \right\rangle$$

is decaying during the time evolution. An alternative strategy will be to pass to the limit $\varepsilon \to 0$, however this depend on wether or not the system $\mathscr{S}_\varepsilon$ satisfies a uniform (in ε) estimate which is related to the non-decay of the perturbed energy. We hope to come back to this point in a forthcoming work.

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remark

The next Chapter is a work with Isabelle Catto, Claude Bardos and Norbert J. Mauser. Preprint to be submitted.

Catto, I., Bardos, C., Mauser, N.J. and Trabelsi, S.: Global Analysis of the Multiconfiguration Time-Dependent Hartree-Fock Equations. Submitted

Chapter 5

Setting and Analysis of the MCTDHF model

Abstract

In this paper we motivate, formulate and analyze time-dependent (TD) *Multiconfiguration (MC) methods* that are extensively used in quantum physics/chemistry for the simulation of interacting many electron systems.

The MultiConfiguration Time-Dependent Hartree-Fock (MCTDHF) equations are a generalization of the Hartree (H) and Hartree-Fock (HF) equations. Our emphasis is on the time-dependent equations where we present the first existence and uniqueness results for the case of a Coulomb interaction.

We present short time well-posedness results of the associated Cauchy problems in H^1 and L^2 under suitable assumption on the rank of the system's "density matrix". Also, we present "global-in-time" results under suitable conditions on the initial data which has to be "somewhat close" to the ground-state.

Also, we present results on TDHF and TDH that are obtained as special cases of the analysis of MCTDHF, significantly simplifying previous proofs.
We emphasize the variational nature which simplifies the mathematical analysis. e.g. the proof of the conservation of the energy.

5.1 Introduction

The aim of this work is to prove mathematically rigorous results concerning the multiconfiguration time dependent Hartree-Fock (MCTDHF) equations. Such models are used in many body quantum physics and quantum chemistry to approximate the solutions of the time-dependent N particle linear Schrödinger equation with binary interaction. The MCTDH(F) models are natural generalizations of the well-known time-dependent Hartree (TDH) approximation and the time-dependent Hartree-Fock (TDHF) approximation, yielding a hierarchy of models that, in principle, should converge to the exact model.
Unless otherwise specified the quantum system under consideration is (a "molecule") composed of a finite number M of nuclei of masses $m_1, \dots, m_M > 0$ with charge $z_1, \dots, z_M > 0$ and a finite number N of electrons.
Under the standard Born–Oppenheimer approximation to separate the dynamic of the nuclei and using a scaling with "atomic units" with $m_e = 2$, the N-body Hamiltonian of the electronic system

subject to the external potential of the nuclei is then the self-adjoint operator

$$\mathscr{H}_N = \sum_{1\le i\le N}\left(-\frac{1}{2}\Delta_{x_i}+U(x_i)\right)+\sum_{1\le i<j\le N}V(x_i-x_j), \tag{5.1}$$

acting on the Hilbert space $L^2(\mathbb{R}^{3N};\mathbb{C})$. The N electrons state is modeled through a so-called *wavefunction* $\Psi=\Psi(x_1,\ldots,x_N)$ living in $L^2(\mathbb{R}^{3N})$ and normalized in order that $\|\Psi\|_{L^2(\mathbb{R}^{3N})}=1$, for $|\Psi|^2$ is interpreted as the probability density of the N electrons. In (5.1) and throughout the paper, the subscript x_i of $-\Delta_{x_i}$ means derivation with respect to the i^{th} variable of the function Ψ. Next,

$$U(x) := -\sum_{m=1}^{M}\frac{z_m}{|x-R_m|},$$

is the Coulomb potential created by M nuclei of respective charge $z_1,\cdots,z_M>0$ located at points $R_1,\cdots,R_M\in\mathbb{R}^3$ and $V(x)=\frac{1}{|x|}$ is the Coulomb repulsive potential between the electrons. Actually our whole analysis carries through to more general potentials such that $U,V\in L^{3/2}(\mathbb{R}^3)+L^\infty(\mathbb{R}^3)$ for the Kato inequality to hold, $V\ge 0$ and U_- goes to 0 at infinity in a weak sense.

In most cases, the domain of the position x will be the whole space $\Omega=\mathbb{R}^3$ or an open set of $\mathbb{R}^3$ endowed with the Lebesgue measure. It will be also useful to consider for Ω a discrete finite or infinite set with a convenient discrete measure.

The time evolution of a quantum system starting from some initial data $\Psi^0\in L^2(\Omega^N)$ is governed by the time-dependent Schrödinger equation

$$\left\{\begin{array}{rcl} i\dfrac{\partial\Psi(t)}{\partial t} &=& \mathscr{H}_N\Psi(t)\,,\\[2ex] \Psi(0) &=& \Psi^0. \end{array}\right. \tag{5.2}$$

For large classes of initial data, the Cauchy problem (5.2) is known to be well-posed, by the Stone theorem which ensures the existence of an unitary group $\mathscr{U}(t)=\exp(-it\mathscr{H}_N)$ such that the unique global solution is given by $\Psi(t)=\mathscr{U}(t)\Psi^0$ for all $t\in\mathbb{R}$.
However, for nearly all applications, the numerical treatment of the exact problem is out of the reach of even the most powerful computers, and approximation algorithms have to be used.

In order to account for the Pauli exclusion principle which features the fermionic nature of the electrons, an antisymmetry condition is imposed to the wave-function Ψ *i.e.*

$$\Psi(x_1,\ldots,x_N)=\varepsilon(\sigma)\,\Psi(x_{\sigma(1)},\ldots,x_{\sigma(N)}),$$

for every permutation σ of $\{1,\ldots,N\}$. The space of antisymmetric wave-functions will be denoted by $\bigwedge_{i=1}^N L^2(\mathbb{R}^3)$. Simplest elements of this space are the so-called *Slater determinants*

$$\Psi(x_1,\ldots,x_N)=\frac{1}{\sqrt{N!}}\det(\phi_i(x_j))_{i,j}\quad\forall i,j\in\{1,\ldots,N\}, \tag{5.3}$$

where the ϕ_i's form an orthonormal family in $L^2(\Omega)$, the factor $\frac{1}{\sqrt{N!}}$ ensures the normalization condition on the wave-function. Such a Slater determinant will be denoted by $\phi_1\wedge\ldots\wedge\phi_N$ in

the following. Löwdin's expansion theorem [26] states that the family of Slater determinants that may be built from an orthonormal Hilbert basis of $L^2(\Omega)$ form an Hilbert basis of $\bigwedge_{i=1}^N L^2(\mathbb{R}^3)$. The basic idea of the MC method is to approximate this huge space by subsets of *finite* linear combinations of Slater determinants.

Hence the *multiconfiguration time-dependent Hartree-Fock* (MCTDHF) system represents a sophisticated hierarchy of approximations. Such "multiconfiguration" methods are widely used in quantum chemistry, as a recipy to perform numerical calculations, but mathematically they are very poorly understood. In fact, the results for the stationary case are very recent [18, 25] and the present paper is the first exhaustive work on the analysis of the time dependent case.

The MCTDHF system is composed of $K \geq N$ non-linear Schrödinger-type evolution equations (for "the orbitals", as a dynamic basis for an expansion by "Slater determinants") coupled with $\binom{K}{N}$ ordinary differential equations (for " the coefficients"). The many particle wavefunction $\Psi^N(t,x_1,...,x_N)$ can be well approximated by such linear combinations of Slater determinants that catch also "correlations", in contrast to the simple time-dependent Hartree-Fock (TDHF) method that corresponds to the special case $K = N$.

The concept of "correlation" of many particle systems is as fundamental as mysterious : indeed, there is no consensus how to exactly define it - only that "correlation is zero for Hartree-Fock wavefunctions". A good mathematically oriented discussion and relevant definition of correlation can be found in [21, 22]. We have used this definition to calculate correlation in our numerical illustration of the MCDTHF method, with a systematic study of correlation with varying K for fixed N.

In principle, the many particle wavefunction constructed from the solution of MCTDHF converges towards the exact solution Ψ^N with increasing K; however, especially in the time-dependent case there is no proof for this seemingly "obvious" property of MCTDHF. A short and readable introduction to the mathematics of time-dependent multiconfiguration Hartree-Fock (MCTDHF) system is given in [29] and [3], or, more exhaustively, in [24], for the time independent case we recommend [25].

In this paper we present the state-of-the-art of the existence and uniqueness analysis of the MCTDHF equation with the singular Coulomb potential as binary interaction : We prove "finite-time" well-posedness in the Sobolev space H^1, the "energy space", under the condition of "full rank" of the "density matrix" of the system. The analysis is extended to the setting of an L^2 theory. Also we prove global well-posedness under a sufficient condition on the energy of the initial data which has to be somewhat "close to the ground state".

Some of the results presented here have been announced in [29] and [3] and the details of the L^2 theory are worked out in [30].

We will use the following notation : $\langle\cdot,\cdot\rangle$ and $\langle\cdot|\cdot\rangle$ denote the usual scalar products in $L^2(\Omega)$ and $L^2(\Omega^N)$ and $a\cdot b$ the complex scalar product of two vectors a and b in $\mathbb{C}^K$ or $\mathbb{C}^r$. The bar denotes complex conjugation.

We set $L^2_\wedge(\Omega^N) := \bigwedge_{k=1}^N L^2(\Omega)$ where the symbol $\wedge$ denotes the skew-symmetric tensorial product.

Throughout the paper bold face letters correspond to one-particle operators on $L^2(\Omega)$, calli-

graphic bold face letters to operators on $L^2(\Omega^N)$, whereas "black board" bold face letters are reserved to matrices of such operators.

5.2 The Stationary Multi-configuration Hartree-Fock Ansatz

5.2.1 The MCHF *ansatz*.

Let (Ω,μ) a given measure space. For positive integers $N \leq K$, let $\Sigma_{N,K}$ denotes the range of the family of increasing mappings $\sigma : \{1,\dots,N\} \longrightarrow \{1,\dots,K\}$. In other words,

$$\Sigma_{N,K} = \Big\{ \sigma = \{\sigma(1) < \dots < \sigma(N)\} \subset \{1,\dots,K\} \Big\}, \qquad |\Sigma_{N,K}| = \binom{K}{N} := r.$$

From now on, we shall use the same notation for the mapping σ and its range $\{\sigma(1) < \dots < \sigma(N)\}$. Next we define

$$\mathscr{F}_{N,K} := S^{r-1} \times \mathscr{O}_{L^2(\Omega)^K}$$

with

$$\mathscr{O}_{L^2(\Omega)^K} = \Big\{ \Phi = (\phi_1,\dots,\phi_K) \in L^2(\Omega)^K \,:\, \int_\Omega \phi_i \bar{\phi}_j \, dx = \delta_{i,j} \Big\}, \tag{5.4}$$

with $\delta_{i,j}$ being the Kronecker delta and with S^{r-1} being the unit sphere in $\mathbb{C}^r$ endowed with the complex euclidean distance

$$S^{r-1} = \Big\{ C = (c_\sigma)_{\sigma\in\Sigma_{N,K}} \in \mathbb{C}^r : \quad \|C\|^2 = \sum_{\sigma\in\Sigma_{N,K}} |c_\sigma|^2 = 1 \Big\}. \tag{5.5}$$

The notation $\sum_{\sigma\in\Sigma_{N,K}}$ means that the sum runs over all the mappings σ of $\Sigma_{N,K}$. We shall use the shorthand $\sum_\sigma$ instead of the cumbersome one above when there is no confusion. Now, given $\sigma \in \Sigma_{N,K}$ and Φ in $\mathscr{O}_{L^2(\Omega)^K}$, we define the associated *Slater determinant* as follows

$$\Phi_\sigma(x_1,\dots,x_N) = \phi_{\sigma(1)} \wedge \dots \wedge \phi_{\sigma(N)} = \frac{1}{\sqrt{N!}} \begin{vmatrix} \phi_{\sigma(1)}(x_1) & \dots & \phi_{\sigma(1)}(x_N) \\ \vdots & & \vdots \\ \phi_{\sigma(N)}(x_1) & \dots & \phi_{\sigma(N)}(x_N) \end{vmatrix};$$

that is, the skew-symmetric function Φ_σ is the determinant built from the ϕ_i's such that $i \in \sigma$. The vector Φ being in $\mathscr{O}_{L^2(\Omega)^K}$, the factor $\frac{1}{\sqrt{N!}}$ ensures the normalization $\|\Phi_\sigma\|_{L^2(\Omega^N)} = 1$. Next, we define the mapping

$$\begin{aligned} \pi_{N,K} : \quad \mathscr{F}_{N,K} \quad &\longrightarrow \quad L^2_\wedge(\Omega^N) \\ (C,\Phi) \quad &\longmapsto \quad \Psi = \pi_{N,K}(C,\Phi) = \textstyle\sum_\sigma c_\sigma \Phi_\sigma. \end{aligned} \tag{5.6}$$

When there is no ambiguity, we simply denote $\pi = \pi_{N,K}$. The application $\pi_{N,K}$ (thereby Ψ) is then a multilinear skew-symmetric mapping from $\mathscr{F}_{N,K}$ into $L^2_\wedge(\Omega^N)$. More precisely, it maps continuously $\mathscr{F}_{N,K}$ equipped with the natural topology of $\mathbb{C}^r \times L^2(\Omega)^K$ into

$$\mathscr{B}_{N,K} = \pi(\mathscr{F}_{N,K}) = \Big\{ \Psi = \sum_\sigma c_\sigma \Phi_\sigma : \quad (C,\Phi) \in \mathscr{F}_{N,K} \Big\}.$$

It is also clearly infinitely differentiable with respect to C and Φ. The wave-function Ψ is a linear combination of r Slater determinants and is therefore skew-symmetric. The set $\mathscr{B}_{N,N}$ is the set of single determinants or *Hartree–Fock states*. Of course $\mathscr{B}_{N,K} \subset \mathscr{B}_{N,K'}$ when $K' \geq K$ and actually

$$L^2_\wedge(\Omega^N) = \lim_{K\to+\infty} \mathscr{B}_{N,K},$$

in the sense of an increasing sequence of sets, since Slater determinants form an Hilbert basis of $L^2_\wedge(\Omega^N)$ (see [26]). In particular, for $\sigma, \tau \in \Sigma_{N,K}$, we have

$$\langle \Phi_\sigma \,|\, \Phi_\tau \rangle = \delta_{\sigma,\tau}. \tag{5.7}$$

More generally, if Φ, $\Xi \in L^2(\Omega)^N$ (not necessarily in $\mathscr{O}_{L^2(\Omega)^N}$), then

$$\langle \phi_1 \wedge \ldots \wedge \phi_N \,|\, \xi_1 \wedge \ldots \wedge \xi_N \rangle = \det\left(\langle \phi_i ; \xi_j \rangle_{1\leq i,j\leq N}\right) \tag{5.8}$$

(see [26]).

A key point of the time dependent case is that the set of ansatz $\mathscr{B}_{N,K}$ is not invariant by the Schrödinger dynamics. It is even expected (but so far not proved to our knowledge) that the solution of the exact Schrödinger equation (5.2) with initial data in $\mathscr{F}_{N,K}$ for some finite $K \geq N$ features an infinite rank at any positive time as long as many-body potentials are involved (see [19] for related issues on the stationary solutions and Subsection 5.3.6 for the picture for non-interacting electrons). We therefore have to rely on a approximation procedure that forces the solutions to stay on the set of ansatz for all time. In order to write down such a time evolution, we first need to investigate further the algebraic and the geometric structure of this set. The range $\mathscr{B}_{N,K}$ of $\mathscr{F}_{N,K}$ by the mapping π is characterized in Proposition 5.2 below in terms of the so-called first-order density matrix (Subsection 5.2.2). Its geometric structure is investigated in Subsection 5.2.3 below.

5.2.2 Density Operators

In this section, we recall the definition of the so-called n^{th} *order density operator* $\left[\Psi\otimes\Psi\right]_{:n}$ (see *e.g.* [1, 26, 6, 4]). Our convention is the same as Löwdin in [26]. In the particular framework of the multi-configuration Hartree–Fock (MCHF) methods, the explicit expression we shall obtain of the first and second order density matrices in terms of the functions ϕ_i and the coefficients c_σ will be important in the sequel (see also [18]). For $n = 1,\ldots,N$ and $\Psi \in L^2_\wedge(\Omega^N)$, a trace-class self-adjoint operator $\left[\Psi\otimes\Psi\right]_{:n}$ is defined on $L^2_\wedge(\Omega^n)$ through its kernel $\left[\Psi\otimes\Psi\right]_{:n}$

$$\left[\Psi\otimes\Psi\right]_{:n}(X_n, Y_n) \;=\; \binom{N}{n} \int_{\Omega^{N-n}} \Psi(X_n, Z_n^N)\, \overline{\Psi}(Y_n, Z_n^N)\, dZ_n^N, \tag{5.9}$$

for $1 \leq n \leq N$ and

$$\left[\Psi\otimes\Psi\right]_{:N}(X_N, Y_N) = \Psi(X_N)\,\overline{\Psi}(Y_N),$$

where we used the notation

$$X_n = (x_1,\ldots,x_n), \qquad X_n^N = (x_{n+1},\ldots,x_N),$$

$$dX_n = dx_1\ldots dx_n, \qquad dX_n^N = dx_{n+1}\ldots dx_N,$$

and similarly for other capital letters. A simple calculation shows that, for $1 \leq n \leq N-1$,

$$\big[\Psi\otimes\Psi\big]_{:n}(X_n,Y_n) = \frac{n+1}{N-n}\int_\Omega \big[\Psi\otimes\Psi\big]_{:n+1}(X_n,z,Y_n,z)\,dz \tag{5.10}$$

In particular, given $1 \leq n \leq p \leq N-1$, one can deduce the expression of $\big[\Psi\otimes\Psi\big]_{:n}$ from the one of $\big[\Psi\otimes\Psi\big]_{:p}$. These kernels are skew-symmetric within each set of capital variables and they satisfy

Proposition 5.1 ([1, 14, 15])**.** *For every integer $1 \leq n \leq N$, the n-th order density matrix is a trace-class self-adjoint operator on $L^2_\wedge(\Omega^n)$ such that*

$$0 \leq \big[\Psi\otimes\Psi\big]_{:n} \leq 1, \tag{5.11}$$

in the sense of operators, and

$$\mathrm{Tr}_{L^2(\Omega^n)}\big[\Psi\otimes\Psi\big]_{:n} = \binom{N}{n}.$$

First- and second-order density matrices will be of particular interest for us. Indeed, the first-order density matrix allows to characterize the set $\mathscr{B}_{N,K}$ (see Proposition 5.2 below) whereas the second-order density matrix is needed to express expectation values of the energy Hamiltonian as soon as two-bodies interactions are involved.

The first-order (or one-particle) density matrix $\big[\Psi\otimes\Psi\big]_{:1}$ is often denoted by γ_Ψ in the literature and in the course of this paper. According to Proposition 5.1 above it is a non-negative self-adjoint trace-class operator on $L^2(\Omega)$, with trace N and with operator norm less or equal to 1. Therefore it is diagonalizable with eigenvalues $\{\gamma_i\}_{i\geq 1}$ such that $0 \leq \gamma_i \leq 1$, for all $i \geq 1$, and $\sum_{i\geq 1}\gamma_i = N$. In particular, at least N of the γ_i's are not zero, and therefore $\mathrm{rank}\,\gamma_\Psi \geq N$, for any $\Psi \in L^2_\wedge(\Omega^N)$. The case when $\mathrm{rank}\,\gamma_\Psi = N$ corresponds to the Hartree–Fock ansatz where Ψ is reduced to a single determinant. The first-order density matrix is then a projector of rank N

Actually, multi-configuration ansatz correspond to first-order density matrices with *finite rank*, and we actually have the following

Proposition 5.2 (Löwdin's expansion theorem [26]; see also [18, 25])**.** *Let $K \geq N$. If $\Psi = \pi(C,\Phi)$ with $(C,\Phi) \in \mathscr{F}_{N,K}$, then* $\mathrm{rank}\big[\Psi\otimes\Psi\big]_{:1} \leq K$ *and* $\mathrm{Ran}\big[\Psi\otimes\Psi\big]_{:1} \subset \mathrm{Span}(\phi_1;\cdots;\phi_K)$*. Conversely, let*

$$\mathscr{B}_{N,K} = \big\{\Psi \in L^2_\wedge(\Omega^N)\ :\ \|\Psi\| = 1 \quad\text{and}\quad \mathrm{rank}\big[\Psi\otimes\Psi\big]_{:1} \leq K\big\}.$$

If $\Psi \in \mathscr{B}_{N,K}$ and if $\mathrm{rank}\big[\Psi\otimes\Psi\big]_{:1} = K'$ *with $N \leq K' \leq K$ and with $\{\phi_1;\ldots;\phi_{K'}\}$ being an orthonormal basis of* $\mathrm{Ran}\big[\Psi\otimes\Psi\big]_{:1}$*, then Ψ can be expanded as a linear combination of Slater determinants built from $(\phi_1;\cdots;\phi_{K'})$. In particular,*

$$\mathscr{B}_{N,K} = \pi(\mathscr{F}_{N,K}).$$

If $\Psi = \pi(C,\Phi) \in \mathscr{B}_{N,K}$, then the kernel of $[\pi(C,\Phi)\otimes\pi(C,\Phi)]_{:1}$ is given by the formula

$$\gamma_{\pi(C,\Phi)}(x,y) := [\pi(C,\Phi)\otimes\pi(C,\Phi)]_{:1}(x,y) = \sum_{i,j=1}^{K}\gamma_{ij}\,\phi_i(x)\,\overline{\phi}_j(y) \tag{5.12}$$

whereas for the second-order density matrix kernel we have

$$[\Psi\otimes\Psi]_{:2}(x,y,x',y') = \sum_{i,j,k,l=1}^{K}\gamma_{ijkl}\,\phi_i(x)\,\phi_j(y)\,\overline{\phi}_k(x')\,\overline{\phi}_l(y'). \tag{5.13}$$

We denote by $\mathbf{\Gamma}_{\pi(C,\Phi)}$ the $K\times K$ matrix with entries $\bar{\gamma}_{ij}$, $1\le i,j\le K$; that is, up complex conjugation, the matrix representation of the first-order density operator in $\mathrm{Span}\{\Phi\} := \mathrm{Span}\{\phi_1;\ldots;\phi_K\}$. (This convention allows for the formula (5.19) below to hold). According to Proposition 5.1, $\mathbf{\Gamma}_{\pi(C,\Phi)}$ is a positive $K\times K$ Hermitian matrix of trace N with same eigenvalues as γ_Ψ and same rank. In particular, there exists a unitary $K\times K$ matrix U such that $U\,\mathbf{\Gamma}_{\pi(C,\Phi)}\,U^\star = \mathrm{diag}(\gamma_1,\ldots,\gamma_K)$ with $0\le\gamma_k\le 1$ and $\sum_{k=1}^K \gamma_k = N$. Hence, γ_Ψ can be expanded as follows

$$\gamma_\Psi(x,y) = \sum_{i=1}^K \gamma_i\,\phi_i'(x)\,\overline{\phi'}_i(y), \tag{5.14}$$

where $\Phi' = U\cdot\Phi$ with obvious notation and with $\{\phi_1';\cdots;\phi_K'\}$ being an eigenbasis of γ_Ψ.

Explicit expressions for the coefficients of the first- and second- order density operators play an essential role in our analysis. They are recalled in the following

Proposition 5.3 ([18], Appendix 1)**.** *Let $\Psi=\pi(C,\Phi)$ in $\mathscr{B}_{N,K}$. Then,*

$$\gamma_{ijkl} = \frac{1}{2}\,(1-\delta_{i,j})(1-\delta_{k,l}) \sum_{\substack{\sigma,\tau\,|\,i,j\in\sigma,k,l\in\tau\\ \sigma\setminus\{i,j\}=\tau\setminus\{k,l\}}} (-1)^\sigma_{i,j}(-1)^\tau_{k,l}\,c_\sigma\,\overline{c}_\tau, \tag{5.15}$$

with

$$(-1)^\sigma_{i,j} = \begin{cases} (-1)^{\sigma^{-1}(i)+\sigma^{-1}(j)+1} & \text{if } i<j,\\ \\ (-1)^{\sigma^{-1}(i)+\sigma^{-1}(j)} & \text{if } i>j. \end{cases} \tag{5.16}$$

In particular, from (5.10)*,*

$$\gamma_{ij} = \frac{2}{N-1}\sum_{k=1}^K \gamma_{ikjk} = \sum_{\substack{\sigma,\tau\,|\,i\in\sigma,\,j\in\tau\\ \sigma\setminus\{i\}=\tau\setminus\{j\}}} (-1)^{\sigma^{-1}(i)+\tau^{-1}(j)}\,c_\sigma\,\overline{c_\tau} \tag{5.17}$$

and

$$\gamma_{ii} = \sum_{\sigma\,|\,i\in\sigma} |c_\sigma|^2. \tag{5.18}$$

Note that it is easily recovered from (5.18) that $0\le\gamma_i\le 1$ for $C\in S^{r-1}$. Similarly n-th order eigenvalues and orbitals may be defined for n-th order density matrices (see [1] for more details). Since we have restricted the analysis to the case of one-body and two-bodies interactions, only the first- and second- order density matrices play a rôle here, but our results carry through general (symmetric) n-bodies interactions as well.

Remark 5.1. It is worth emphasizing the fact that the coefficients γ_{ij} and γ_{ijkl} only depend on the expansion coefficients C and not on the orbitals, and that this dependency is quadratic. This property actually holds true at any order $1\le p\le N-1$. We shall rely on it in the proof of existence of solutions in Section 5.4.

As a consequence of the above remark, we shall use the shorthand $\mathbf{\Gamma}(C)$ for $\mathbf{\Gamma}_{\pi(C,\Phi)}$ from now on.

Remark 5.2. Of course, when $K = N$ (Hartree–Fock case), γ_Ψ being of trace N must be the projector on $\mathrm{Span}(\phi_1;\cdots;\phi_N)$; that is, $\mathbf{\Gamma}_{\pi(1,\Phi)}$ is the $N\times N$ identity matrix and

$$\gamma_\Psi(x,y) = \sum_{i=1}^{N} \phi_i(x)\,\overline{\phi}_i(y) := \mathbf{P}_\Phi(x,y),$$

with $\mathbf{P}_\Phi$ denoting the projector on $\mathrm{Span}\{\phi_1,\cdots,\phi_N\}$. In this case, (5.17) and (5.15) simply reduce to $\gamma_{ij} = \delta_{i,j}$ and $\gamma_{ijkl} = \frac{1}{2}\big(\delta_{i,k}\delta_{j,l} - \delta_{i,l}\delta_{j,k}\big)$.

In the following the time evolution of a MCTDHF ansatz Ψ that stays in $\mathscr{B}_{N,K}$ is stated and studied in terms of the time evolution of the expansion coefficients C and of the K one-particle functions Φ. It is then necessary to characterize the preimage $\pi^{-1}(\Psi)$ in $\mathscr{F}_{N,K}$ for any $\Psi\in\mathscr{B}_{N,K}$.

The first obvious fact is

Proposition 5.4. *Let (C,Φ) and (C',Φ) in $\mathscr{F}_{N,K}$ such that $\sum_\sigma c_\sigma\,\Phi_\sigma = \sum_\sigma c'_\sigma\,\Phi_\sigma$. Then $C = C'$.*

Proof. The family $\{\phi_i\}_{1\le i\le K}$ being an orthonormal family in $L^2(\Omega)$, the same holds true for the family of determinants $\{\Phi_\sigma\}_{\sigma\in\Sigma_{N,K}}$ in $\bigwedge_{k=1}^N L^2(\Omega)$. The claim follows obviously for $c_\sigma = c'_\sigma = \langle\Psi\,|\,\Phi_\sigma\rangle$. □

Another easy particular case is the Hartree–Fock ansatz ($K = N$). The preimage of $\Psi^{HF}\in\mathscr{B}_{N,N}$ by $\pi_{N,N}$ is well-known. Indeed, $\Psi^{HF} = \phi_1\wedge\cdots\wedge\phi_N = \psi_1\wedge\cdots\wedge\psi_N$ if and only there exists a $N\times N$ unitary transform U such that $(\phi_1,\cdots,\phi_N) = (\psi_1,\cdots,\psi_N)\cdot U$ (see Proposition 5.5 below). However since

$$K\le K' \implies \mathscr{B}_{N,K}\subset\mathscr{B}_{N,K'},$$

any Slater determinant $\Psi^{HF} = \phi_1\wedge\cdots\wedge\phi_N$ belongs to $\mathscr{B}_{N,K}$ for all $K\ge N$. As observed previously, $\pi_{N,N}^{-1}(\Psi^{HF})$ is perfectly identified with the orbit of Φ by $\mathscr{O}_N$, with $\mathscr{O}_\ell$ being the set of $\ell\times\ell$ unitary matrices. On the contrary, when $K > N$, if (C,Φ) and (C',Φ') in $\mathscr{F}_{N,K}$ are such that $\pi(C,\Phi) = \pi(C',\Phi') = \Psi^{HF}$, there is no algebraic relation between the vectors Φ and Φ' in $L^2(\Omega)^K$. Indeed, up to a permutation of the indices, we may assume that $\mathrm{Span}\{\phi_1,\cdots,\phi_N\} = \mathrm{Span}\{\phi'_1,\cdots,\phi'_N\} = \mathrm{Ran}\,\gamma_{\Psi^{HF}}$ and that ϕ_i and ϕ'_i belong to $\mathrm{Ran}\,\gamma_{\Psi^{HF}}{}^\perp$ for all $N+1\le i\le K$. Then, $(\phi_1,\cdots,\phi_N)$ and $(\phi'_1,\cdots,\phi'_N)$ are deduced from each other by a $N\times N$ unitary transform and $\gamma_\Psi = P_{(\phi_1,\cdots,\phi_N)} = P_{(\phi'_1,\cdots,\phi'_N)}$. We deduce from (5.18) that $c_\sigma = c'_\sigma = 0$ for all $\sigma\in\Sigma_{N,K}$ such that $\{N+1,\cdots,K\}\cap\sigma\neq\emptyset$. In other words, $C = C'$ with $c_\sigma = c'_\sigma = 1$ when $\sigma = \{1,\cdots,N\}$, and 0 otherwise, $\Psi_{HF} = \phi_1\wedge\cdots\wedge\phi_N = \phi'_1\wedge\cdots\wedge\phi'_N$ whereas for $N+1\le i\le k$, ϕ_i and ϕ'_i may be taken arbitrary in the infinite dimensional subspace $\mathrm{Ran}\,\gamma_{\Psi^{HF}}^\perp$ of $L^2(\Omega)$.

More generally, when $\Psi\in\mathscr{B}_{N,K}$ and $\mathrm{rank}\gamma_\Psi = K$, the preimage $\pi_{N,K}^{-1}(\Psi)$ has an orbit-like geometric structure that is made precise in Proposition 5.5 below. We therefore introduce

$$\partial\mathscr{B}_{N,K} := \big\{\Psi\in\mathscr{B}_{N,K} : \mathrm{rank}\,\gamma_\Psi = K\big\}$$

and, by analogy,

$$\partial\mathscr{F}_{N,K} = \pi_{N,K}^{-1}(\partial\mathscr{B}_{N,K}) := \big\{(C,\Phi)\in\mathscr{F}_{N,K} : \mathrm{rank}\,\mathbf{\Gamma}(C) = K\big\}.$$

In this latter case, as in [23], we shall say that the associated $K \times K$ matrix $\mathbf{\Gamma}(C)$ satisfies a *full-rank assumption*. In particular it is invertible with positive eigenvalues. Since invertible matrices form an open subset and since the mapping $C \mapsto \mathbf{\Gamma}(C)$ is continuous, it is clear that $\partial\mathscr{F}_{N,K}$ is an open subset of $\mathscr{F}_{N,K}$.

Clearly $\partial\mathscr{B}_{N,N} = \mathscr{B}_{N,N}$ and $\partial\mathscr{F}_{N,N} = \mathscr{F}_{N,N}$; that is, the full-rank assumption is automatically satisfied in the Hartree–Fock setting (see Remark 5.2).

On the opposite, it may happen that $\partial\mathscr{B}_{N,K} = \emptyset$ (in that case $\mathscr{B}_{N,K} = \mathscr{B}_{N,K-1}$). Indeed, not all integers $K \geq N$ are admissible ranks of first-order density matrices as recalled in the following lemma

Lemma 5.1 (Classification of admissible ranks; Lemma 1.1 in [18])**.** *Let $K \geq N$, the set $\partial\mathscr{B}_{N,K}$ is non-empty if and only if:*

$$K \begin{cases} = 1 & N = 1 \\ \geq 2, \text{ even} & N = 2 \\ \geq N, \neq N+1, & N \geq 3. \end{cases}.$$

From now on, unless otherwise specified, we only deal with pairs (N,K) with K admissible. We recall from [26] the following

Proposition 5.5. *Let (C,Φ) and (C',Φ') in $\partial\mathscr{F}_{N,K}$ such that $\pi(C,\Phi) = \pi(C',\Phi')$. Then, there exists a unique unitary matrix $U \in \mathscr{O}_K$ and a unique unitary matrix $d(U) = \overline{\mathbb{U}} \in \mathscr{O}_r$ such that*

$$\Phi' = U \cdot \Phi, \quad C' = d(U) \cdot C$$

where, for every $\sigma \in \Sigma_{N,K}$,

$$\Phi'_\sigma = \sum_\tau \mathbb{U}_{\sigma,\tau}\,\Phi_\tau.$$

Moreover,

$$\mathbf{\Gamma}(C') = U\,\mathbf{\Gamma}(C)\,U^\star. \tag{5.19}$$

Proof. Let (C,Φ) and (C',Φ') in $\partial\mathscr{F}_{N,K}$ such that $\pi(C,\Phi) = \pi(C',\Phi') = \Psi \in \partial\mathscr{B}_{N,K}$. From Proposition 5.2, $\mathrm{Span}\{\Phi\} = \mathrm{Span}\{\Phi'\} = \mathrm{Ran}(\gamma_\Psi)$ with Φ and Φ' in $\mathscr{O}_{L^2(\Omega)^K}$, therefore there exists a unique unitary matrix $U \in \mathscr{O}_K$ such that $\Phi' = U \cdot \Phi$. Eqn. (5.19) follows by definition of $\mathbf{\Gamma}(C)$. Accordingly, there exists a unique unitary matrix $\mathbb{U}$ in $\mathscr{O}_r$ that maps the family $\{\Phi_\sigma\}_{\sigma\in\Sigma_{N,K}}$ to $\{\Phi'_\sigma\}_{\sigma\in\Sigma_{N,K}}$. More precisely, being given $\sigma \in \Sigma_{N,K}$, we have by a direct calculation (see also [26])

$$\Phi'_\sigma \;=\; \sum_\tau \mathbb{U}_{\sigma,\tau}\,\Phi_\tau \tag{5.20}$$

where, for all $\sigma,\tau \in \Sigma_{N,K}$,

$$\begin{aligned} \mathbb{U}_{\sigma,\tau} &= \begin{vmatrix} U_{\sigma(1),\tau(1)} & \cdots & U_{\sigma(N),\tau(1)} \\ \vdots & \vdots & \vdots \\ \vdots & \vdots & \vdots \\ U_{\sigma(1),\tau(N)} & \cdots & U_{\sigma(N),\tau(N)} \end{vmatrix} = \det\big(U_{\sigma(j),\tau(i)}\big)_{1\leq i,j\leq N}, \\ &= \det\Big(\langle \phi'_{\sigma(j)}; \phi_{\tau(i)}\rangle\Big)_{1\leq i,j\leq N}. \end{aligned} \tag{5.21}$$

By construction the $r \times r$ matrix $\mathbb{U}$ with matrix elements $\mathbb{U}_{\sigma,\tau}$ is unitary. By the orthonormality of the determinants, we have

$$c'_\sigma = \Big\langle \pi(C,\Phi) \,|\, \Phi'_\sigma \Big\rangle = \sum_\tau c_\tau \Big\langle \Phi_\tau \,|\, \Phi'_\sigma \Big\rangle = \sum_\tau \overline{\mathbb{U}}_{\sigma,\tau}\, c_\tau, \tag{5.22}$$

whence the lemma with $d(U) = \overline{\mathbb{U}}$. □

5.2.3 Full-rank and fibration

Under the full-rank assumption and given (N,K) admissible, the set $\partial\mathscr{B}_{N,K}$ is a principal fiber bundle. Indeed, within geometry-differential terminology, $\partial\mathscr{B}_{N,K}$ is called the base, and, for any $\Psi \in \partial\mathscr{B}_{N,K}$, the preimage $\pi^{-1}(\Psi)$ is the fiber at Ψ. Proposition 5.2.2 helps defining a group action on $\partial\mathscr{F}_{N,K}$ that is referred to with the following notation

$$\begin{aligned} &(C',\Phi') = \mathscr{U}\cdot(C,\Phi) \quad \Longleftrightarrow \quad C' = d(U)\cdot C \quad \text{and} \quad \Phi' = U\cdot\Phi, \\ &\mathscr{U} := \big(d(U),U\big) \in \mathscr{O}_r \times \mathscr{O}_K. \end{aligned} \tag{5.23}$$

Indeed on the one hand, it is clear from Lemma 5.4 or from the expression for the matrix elements of $d(U)$ that $d(I_K) = I_r$. On the other hand from (5.20) and (5.22) it is easily checked that $d(UV) = d(U)\,d(V)$. Therefore couples of the form $\big(d(U),U\big)$ form a subgroup of $\mathscr{O}_r \times \mathscr{O}_K$ that we denote by $\mathscr{O}_K^r$. The action of $\mathscr{O}_K^r$ is not free on $\mathscr{F}_{N,K}$ itself (as shown before on the examples of Slater determinants in $\mathscr{F}_{N,K}$ with $K > N$), but it is free on $\partial\mathscr{F}_{N,K}$ and transitive on any fiber $\pi^{-1}(\Psi)$ for every $\Psi \in \partial\mathscr{B}_{N,K}$. Therefore, the mapping π defines a principal bundle with fiber given by the group $\mathscr{O}_K^r$. We can define local (cross-)sections as continuous maps $s : \Psi \mapsto (C,\Phi)$ from $\partial\mathscr{B}_{N,K}$ to $\partial\mathscr{F}_{N,K}$ such that $\pi \circ s$ is the identity. In particular, $\partial\mathscr{F}_{N,K}/\mathscr{O}_K^r$ is homeomorphic to $\partial\mathscr{B}_{N,K}$. Since the map π is C^∞, one concludes from the inverse mapping theorem that the above isomorphism is also topological. In the Hartree–Fock case $K = N$ where the full-rank assumption is automatically fulfilled, $\pi_{N,N}^{-1}\big(\mathscr{B}_{N,N}\big)$ is a so-called Stiefel manifold.

Having equipped $\partial\mathscr{B}_{N,K}$ with a manifold structure we can now study the tangent space of $\partial\mathscr{B}_{N,K}$.

Being polynomial in its variables, the application π is clearly differentiable with respect to C and Φ and its derivatives can be written down explicitly. We then define

$$\begin{aligned} \nabla : \qquad \partial\mathscr{B}_{N,K} \qquad &\longrightarrow \quad \mathscr{L}\left(\mathbb{C}^r \times L^2(\Omega)^K; L^2(\Omega^N)\right) \\ \Psi = \pi(C,\Phi) \quad &\longmapsto \qquad \nabla\Psi = (\nabla_C\Psi, \nabla_\Phi\Psi) \end{aligned}$$

where here and below $\mathscr{L}(E;F)$ denotes the set of continuous linear applications from E to F (as usual $\mathscr{L}(E) = \mathscr{L}(E;E)$). For any $\Psi = \pi(C,\Phi) \in \partial\mathscr{B}_{N,K}$

$$\nabla_C\Psi = \Big(\frac{\partial\Psi}{\partial c_{\sigma_1}}, \ldots, \frac{\partial\Psi}{\partial c_{\sigma_r}}\Big) = (\Phi_{\sigma_1}, \ldots, \Phi_{\sigma_r}), \tag{5.24}$$

and for any $\zeta = (\zeta_1, \ldots, \zeta_K) \in L^2(\Omega)^K$,

$$\nabla_\Phi\Psi\,[\zeta] = \Big(\frac{\partial\Psi}{\partial\phi_1}[\zeta_1], \ldots, \frac{\partial\Psi}{\partial\phi_K}[\zeta_K]\Big), \tag{5.25}$$

with $\frac{\partial \Psi}{\partial \phi_k}$ being a linear application from $L^2(\Omega)$ to $L^2_\wedge(\Omega^N)$ that is given by

$$\frac{\partial \Psi}{\partial \phi_k}[\zeta] = \sum_{i=1}^{N} \zeta(x_i) \int_\Omega \Psi(x_1,\dots,x_N)\, \overline{\phi}_k(x_i)\, dx_i, \tag{5.26}$$

for any function $\zeta \in L^2(\Omega)$. Actually, for every $\sigma \in \Sigma_{N,K}$, $1 \le k \le K$, we have

$$\frac{\partial \Phi_\sigma}{\partial \phi_k}[\zeta] = \begin{cases} 0 & \text{if } k \notin \sigma, \\ \\ \phi_{\sigma(1)} \wedge \cdots \wedge \phi_{\sigma(j-1)} \wedge \zeta \wedge \phi_{\sigma(j+1)} \wedge \cdots \wedge \phi_{\sigma(N)} & \text{if } \sigma^{-1}(k) = j. \end{cases} \tag{5.27}$$

From (5.27) we recover the Euler Formula for homogeneous functions, that reads here

$$\Psi = \frac{1}{N} \sum_{k=1}^{K} \frac{\partial \Psi}{\partial \phi_k}[\phi_k] := \frac{1}{N}\, \nabla_\Phi \Psi \cdot \Phi. \tag{5.28}$$

From (5.28), (5.24), (5.25) and (5.26) it is easily checked that the tangent space of $\mathscr{B}_{N,K}$ at $\Psi = \pi(C,\Phi)$ is given by

$$\begin{aligned} \mathbf{T}_\Psi \mathscr{B}_{N,K} = \Big\{ \delta\Psi = \sum_\sigma \Phi_\sigma\, \delta c_\sigma + \frac{1}{N} \sum_\sigma \sum_{k=1}^{K} c_\sigma \frac{\partial \Phi_\sigma}{\partial \phi_k}[\delta\phi_k] \in L^2_\wedge(\Omega^N) \ : \\ \delta C = \big(\delta c_{\sigma_1}, \cdots, \delta c_{\sigma_r}\big) \in \mathbb{C}^r,\ \delta\phi_k \in \mathrm{Span}\{\Phi\}^\perp, \text{ for every } 1 \le k \le K \Big\}. \end{aligned} \tag{5.29}$$

In the following we shall also make use of the adjoint $\nabla_\Phi \Psi^\star$ of the operator $\nabla_\Phi \Psi$ that is defined by

$$\forall \zeta \in L^2(\Omega)^K, \forall\, \Xi \in L^2_\wedge(\Omega_N), \quad \langle \nabla_\Phi \Psi^\star[\Xi]\,;\, \zeta \rangle_{L^2(\Omega)} = \Big\langle \Xi \Big| \nabla_\Phi \Psi \cdot \zeta \Big\rangle_{L^2(\Omega^N)}. \tag{5.30}$$

In other words, for all $1 \le k \le K$, $\dfrac{\partial \Psi^\star}{\partial \phi_k}$ denotes the linear operator in $\mathscr{L}\big(L^2_\wedge(\Omega^N); L^2(\Omega)\big)$ that is defined by

$$\frac{\partial \Psi^\star}{\partial \phi_k}\,[\Xi](x) = N \int_\Omega \phi_k(y) \Big(\int_{\Omega^{N-1}} \Xi(x,x_2,\dots,x_N)\, \overline{\Psi}(y,x_2,\dots,x_N)\, dx_2 \cdots dx_N \Big) dy,$$

for any function Ξ in $L^2_\wedge(\Omega^N)$.

It is also worth emphasizing the fact that changing (C,Φ) to (C',Φ') following the group action (5.23), involves a straightforward change of "variable" in the derivation of Ψ; namely, with a straightforward chain rule,

$$\nabla_C \Psi = \mathbb{U}^\star \cdot \nabla_{C'} \Psi = \nabla_{C'} \Psi \cdot d(U), \quad \nabla_\Phi \Psi = \nabla_{\Phi'} \Psi \cdot U, \tag{5.31}$$

and

$$[\nabla_{\Phi'} \Psi]^\star = U \cdot [\nabla_\Phi \Psi]^\star. \tag{5.32}$$

Finally, we shall make use below of the following further properties of the functional derivatives of Ψ.

Lemma 5.2. *Let $(C,\Phi) \in \mathscr{F}_{N,K}$ with $\Psi = \pi(C,\Phi)$. Then, for all $\zeta \in \mathrm{Span}\{\Phi\}^{\perp}$, $\xi \in L^2(\Omega)$ and $\sigma,\tau \in \Sigma_{N,K}$, we have*

$$\left\langle \frac{\partial \Phi_\tau}{\partial \phi_k}[\zeta] \,\middle|\, \Phi_\sigma \right\rangle = 0,$$

and

$$\left\langle \frac{\partial \Psi}{\partial \phi_k}[\zeta] \,\middle|\, \frac{\partial \Psi}{\partial \phi_l}[\xi] \right\rangle = \mathbf{\Gamma}_{lk} \langle \zeta, \xi \rangle, \tag{5.33}$$

for any $1 \le k,l \le K$.

Proof. The first claim follows immediately in virtue of (5.27) and (5.8). For the second claim we proceed as follows. Thanks to (5.27) again

$$\begin{aligned}
\left\langle \frac{\partial \Psi}{\partial \phi_k}[\zeta] \,\middle|\, \frac{\partial \Psi}{\partial \phi_l}[\xi] \right\rangle &= \sum_{\sigma,\tau \,|\, k\in\sigma, l\in\tau} c_\sigma \overline{c}_\tau \left\langle \frac{\partial \Phi_\sigma}{\partial \phi_k}[\zeta] \,\middle|\, \frac{\partial \Phi_\tau}{\partial \phi_l}[\xi] \right\rangle \\
&= \sum_{\substack{\sigma,\tau \,|\, k\in\sigma, l\in\tau \\ \sigma\setminus\{k\}=\tau\setminus\{l\}}} (-1)^{\sigma^{-1}(k)} (-1)^{\tau^{-1}(l)} c_\sigma \overline{c}_\tau \langle \zeta, \xi \rangle \\
&= \mathbf{\Gamma}_{lk} \left\langle \zeta, \xi \right\rangle.
\end{aligned}$$

We conclude with the help of (5.17). □

5.2.4 Correlation to quantum physics

The wave-function $\Psi \in L^2(\Omega^N)$ with $\|\Psi\| = 1$ is interpreted through the square of its modulus $|\Psi(X_N)|^2$ $(= \left[\Psi \otimes \Psi\right]_{:N}(X_N,X_N))$ that represents the density of probability of presence of the N electrons in Ω^N. More generally, for all $1 \le n \le N$, the positive function $X_n \mapsto \left[\Psi \otimes \Psi\right]_{:n}(X_n,X_n)$ is in $L^1(\Omega^n)$ with L^1 norm equal to $\binom{N}{n}$, and it is interpreted as $\binom{N}{n}$ times the density of probability for finding n electrons located at $X_n \in \Omega^n$. Any set $\{\sigma(1),\ldots,\sigma(N)\}$ for $\sigma \in \Sigma_{N,K}$ is called a *configuration* in quantum chemistry literature and this is where the terminology *multi-configuration* comes from for wave-functions in $\mathscr{B}_{N,K}$. When $\{\phi_k\}_{1\le k\le K}$ is an orthonormal basis of $\mathrm{Ran}\left[\Psi \otimes \Psi\right]_{:1}$ each mono-electronic function ϕ_k is called an *orbital* of Ψ. When the orbitals are also eigenfunctions of $[\pi(C,\Phi) \otimes \pi(C,\Phi)]_{:1}$ according to (5.14) they are referred to as *natural orbitals* in the literature whereas the associated eigenvalues $\{\gamma_i\}_{1\le i\le K}$ are referred to as *occupation numbers*. Under the full-rank assumption, only occupied orbitals are taken into account. The function $\int_\Omega \Psi(x_1,\ldots,x_N)\,\overline{\phi}_k(x_k)\,dx_k$ is known as a *single-hole function* in the literature (see *e.g.* [7, 9]). Finally, the $K \times K$ matrix $\mathbf{\Gamma}(C)$ is called *the charge- and bond matrix* (see Löwdin [26]).

A key concept for many particle system is “correlation" (which is related to “entanglement"). Whereas the “correlation energy" of a many particle wavefunction associated to a many particle Hamiltonian is a relatively precise concept, the intrinsic correlation of a many particle wavefunction as such is a rather vague concept, with several different definitions in the literature. Gottlieb and Mauser recently introduced a new measure for the correlation [21, 22]. This *non-freeness* is

an entropy type functional depending only on the density operator$[\Psi\otimes\Psi]_{:1}$, and defined as follow

$$\begin{aligned}\mathfrak{E}(\Psi) = &- Tr\Big\{[\Psi\otimes\Psi]_{:1}\, log([\Psi\otimes\Psi]_{:1}))\Big\}\\ &- Tr\Big\{(\mathbf{1}-[\Psi\otimes\Psi]_{:1})\, log(\mathbf{1}-[\Psi\otimes\Psi]_{:1})\Big\},\end{aligned} \tag{5.34}$$

Hence it depends on the eigenvalues of $[\Psi\otimes\Psi]_{:1}$ in the following explicit way

$$\mathfrak{E}(\Psi) = - \sum_{i=1}^{K}\Big(\gamma_i\, log(\gamma_i)+(1-\gamma_i)\, log(1-\gamma_i)\Big).$$

It is a concave functional minimized for $\gamma_i = 0$ or 1. In the MCHF case this functional depends implicitly on K and N *via* the dependency on the γ_i's. This definition of correlation has the following basic property

Lemma 5.3. *The correlation vanishes if and only if Ψ is a single Slater determinant.*

Proof. We present the simple but instructive proof which has not been given in [21, 22], based on the *Löwdin expansion Theorem* [26] .
Assume that $\Psi = \phi_1\wedge\ldots\wedge\phi_N$,i.e. factorized as a single Slater determinant. Then, $[\Psi\otimes\Psi]_{:1}(x,y) = \sum_{k=1}^{N}\phi_k(x)\overline{\phi}_k(y)$. That is $\gamma_k = 1$ for all $1\le k\le N$ (see also the third assertion of the Lemma 5.1). Thus $\mathfrak{C}(\phi_1\wedge\ldots\wedge\phi_N) = 0$. Next, $\mathfrak{C}(\Psi) = 0$ if and only if the eigenvalues of $[\Psi\otimes\Psi]_{:1}(x,y)$ are 0 or 1. Thus, there exists $p\in\mathbb{N}^\star$ such that $[\Psi\otimes\Psi]_{:1}(x,y) = \sum_{k=1}^{p}\gamma_k\,\phi_k(x)\overline{\phi}_k(y)$. But $\sum_{k=1}^{N}\gamma_k = N$ and $\gamma_k = 1$ for all $1\le k\le p$. Hence, obviously $p = N$. That is $[\Psi\otimes\Psi]_{:1}(x,y) = \sum_{k=1}^{N}\phi_k(x)\overline{\phi}_k(y)$ and the *Löwdin expansion Theorem* tells us that Ψ is a single Slater determinant. □

It is interesting to notice that the correlation does not vanish in the case of a sum of disjoint Slater determinants. In fact, let

$$\Psi = \sum_{I_p} c_p\Phi_p,\quad I_p = \{Np+1,\ldots,N(p+1)\},\ p = 0,\ldots,k.$$

It is straightforward that

$$\begin{aligned}[\Psi\otimes\Psi]_{:1}(x,y) &= \sum_{I_p}|c_p|^2[\Phi_p\otimes\Phi_p]_{:1},\\ &= \sum_{I_p}|c_p|^2\sum_{i\in I_p}\phi_i(x)\,\overline{\phi}_i(y).\end{aligned}$$

Its eigenvalues are then $|c_p|^2$ and each one is at least of multiplicity N. Next using the fact that $\sum_{p=1}^{k}|c_p|^2 = 1$ and that $\mathfrak{C}$ vanishes only if the eigenvalues are 0 or 1, one can see that $\mathfrak{C}(\Psi) > 0$ for all $k > 1$. The case $k = 1$ being the case of a single Slater determinant treated before which the only case for which the correlation is zero. In addition, we believe that any convex or (concave) function of the eigenvalues of the operator $[\Psi\otimes\Psi]_{:1}$ vanishing if and only if the associated eigenvalues are all 0 or 1 can be considered as a measure for the degree of correlation of the concerned system. For example, only the first or the second part of the functional defined in (5.34).

Lemma 5.3 shows why the single Slater determinant case is usually taken as the definition of uncorrelated (also called “free") wavefunctions and why the Hartree-Fock ansatz is not able to catch “correlation effects". Even if the initial data happens to be a single Slater determinant, the interaction of the particles would immediately create "correlations" in the time evolution - however, the TDHF method forces the dynamic to stay on a manifold where correlation is alwawys zero.
Improving the approximation by adding determinants systematically brings in correlation into the *multiconfiguration ansatz*. Now correlation effects of the many particle wavefunction can be included in the initial data and the effects of dynamical correlation - decorrelation can be caught in the time evolution. This is a very important conceptually advantage of MCTDHF for the modeling and simulation of correlated few electron systems. Such systems, for example in "photonics" where an atom interacting with a laser is measured on the femto- or atto-second scale, are increasingly studied and have given a boost to MCTDHF. For example A. Scrinzi's powerful programme package for numerical simulation of MCTDHF (see e.g. [9] is used by the groups of top physicists in the field of "atto physics" (see e.g. the Nature papers with F. Krausz and nobel laureate T.W. Hänsch [2]).

5.3 Flow on the Fiber Bundle

In this section, we consider an arbitrary self-adjoint operator $\mathscr{H}$ in $L^2(\Omega^N)$. We will come back to the physical case in Section 5.4 below. In order to write down the MCTDHF equations, we need to extend the ansatz (5.6) to the time-dependent case by considering a time-dependent combination coefficients and functions. In other words

$$\Psi(t,x_1,\dots,x_N) = \sum_{\sigma} c_\sigma(t)\,\Phi_\sigma(t,x_1\dots,x_N), \tag{5.35}$$

with $\big(C(t),\Phi(t)\big) \in \mathscr{F}_{N,K}$ for all $t \geq 0$. Our starting point is the following differential coupled system whose derivation will be explained below

$$\mathscr{S}_0 : \quad \begin{cases} i\,\dfrac{dC}{dt} = \big\langle \mathscr{H}\,\Psi \,|\, \nabla_C \Psi \big\rangle, \\[2ex] i\,\mathbf{\Gamma}\big(C(t)\big)\,\dfrac{\partial \Phi}{\partial t} = (\mathbf{I}-\mathbf{P}_\Phi)\,\nabla_\Phi \Psi^\star[\mathscr{H}\,\Psi], \\[2ex] \big(C(t),\Phi(t)\big) = \big(C_0,\Phi_0\big), \end{cases}$$

for given initial data $\big(C_0,\Phi_0\big)$ in $\mathscr{F}_{N,K}$. The operator $\mathbf{P}_\Phi$ in $\mathscr{S}_0$ denotes the projector onto the space spanned by the ϕ_i's. More precisely

$$\mathbf{P}_\Phi(\cdot) = \sum_{i=1}^{K} \big\langle \cdot\,,\,\phi_i \big\rangle\,\phi_i. \tag{5.36}$$

Remark 5.3. Since for every $\sigma \in \Sigma_{N,K}$, $\dfrac{\partial \Psi}{\partial c_\sigma} = \Phi_\sigma$, the system for the c_σ's can also be expressed as

$$i\,\frac{dc_\sigma}{dt} = \sum_{\tau} \big\langle \mathscr{H}\,\Phi_\tau \,|\, \Phi_\sigma \big\rangle\, c_\tau. \tag{5.37}$$

This equation is then obviously linear in these variables. Furthermore when the ϕ_i's (or equivalently the Φ_σ's) are kept constant in time, (5.37) is nothing but a Galerkin approximation for the evolution equation (5.2). The MCTDHF approximation then reveals as an extension (with extra degree of freedom in the basis functions) of the Galerkin approximation. However since the system $\mathscr{S}_0$ is coupled, the Galerkin approximation cannot be seen as a special case of the MCTDHF unless $\nabla_\Phi \Psi^\star[\mathscr{H}\Psi] \in \mathrm{Span}\{\Phi\}^\perp$ and $\mathbf{\Gamma}$ is of rank K .

5.3.1 Conservation Laws

From this point onward, the full-rank assumption $\mathbf{\Gamma}(C)$ in $\mathrm{GL}(K)$ is assumed on the time interval $t \in [0,T)$ with $T > 0$. In this section, we check that the expected conservation laws (propagation of constraints, conservation of the energy) indeed hold for the MCTDHF system. We start with the following

Lemma 5.4 (The dynamics preserves $\mathscr{F}_{N,K}$)**.** *Let $(C_0,\Phi_0) \in \mathscr{F}_{N,K}$ being the initial data. If there exists a solution to the system $\mathscr{S}_0$ on $[0,T]$ with $T>0$ such that $\mathrm{rank}\,\mathbf{\Gamma}\big(C(t)\big) = K$ for all $t \in [0;T]$, then $(C(t),\Phi(t)) \in \partial\mathscr{F}_{N,K}$ for all $t \in [0;T]$; that is*

$$\sum_\sigma |c_\sigma(t)|^2 = 1, \quad \int_\Omega \phi_i(t)\,\bar{\phi}_j(t)\,dx = \delta_{i,j},$$

for all $t \in [0;T]$.

Proof. First we prove that $\sum_\sigma |c_\sigma(t)|^2 = \sum_\sigma |c_\sigma(0)|^2$ for all $t \in [0,T]$. By taking the scalar product of the differential equation satisfied by C in $\mathscr{S}_0$ with C itself, we get

$$\frac{d}{dt}|C(t)|^2 = 2\,\Re\big(\frac{d}{dt}C(t);C(t)\big) = 2\,\Im\sum_\sigma \Big\langle \mathscr{H}\Psi \,|\, c_\sigma\,\Phi_\sigma \Big\rangle = 2\,\Im\Big\langle \mathscr{H}\Psi \,|\, \Psi\Big\rangle = 0,$$

thanks to the self-adjointness of $\mathscr{H}$, where $\Re$ and $\Im$ denote respectively real and imaginary parts of a complex number. From the other hand, the full-rank assumption allows to multiply the system of equations satisfied by Φ in $\mathscr{S}_0$ by $\mathbf{\Gamma}^{-1}$ to obtain

$$i\,\frac{\partial\Phi}{\partial t} = (\mathbf{I}-\mathbf{P}_\Phi)\,\mathbf{\Gamma}(C)^{-1}\,\nabla_\Phi\,\Psi^\star[\mathscr{H}\Psi]. \tag{5.38}$$

(Notice that $\mathbf{P}_\Phi$ commutes with $\mathbf{\Gamma}(C)^{-1}$.) By definition $\mathbf{I}-\mathbf{P}_\Phi$ projects on the orthogonal subspace of $\mathrm{Span}\{\Phi\}$, therefore $\frac{\partial}{\partial t}\phi_i$ lives in $\mathrm{Span}\{\Phi\}^\perp$ for all t. Hence,

$$\Big\langle \frac{\partial\phi_i(t)}{\partial t}\,,\,\phi_j(t)\Big\rangle = 0. \tag{5.39}$$

for all $1 \le i,j \le K$ and for all $t \in [0,T]$. This achieves the proof of the lemma. □

In Physics' literature, the MCTDHF equations are usually derived from the so-called Dirac–Frenkel variational principle (see, among others, [16, 17, 23]) that demands that

$$\Big\langle i\frac{\partial\Psi}{\partial t} - \mathscr{H}\Psi \,\Big|\, \delta\Psi\Big\rangle = 0, \quad \text{for all} \quad \delta\Psi \in \mathbf{T}_\Psi\mathscr{B}_{N,K},$$

where $\mathbf{T}_\Psi\mathscr{B}_{N,K}$ denotes the tangent space to the differentiable manifold $\mathscr{B}_{N,K}$ at Ψ. We indeed check in the forthcoming proposition that, if $(C,\Phi) \in \partial\mathscr{F}_{N,K}$ satisfies $\mathscr{S}_0$ for all $t \in (0,T)$, then it also satisfies the Dirac-Frenkel variational principle.

Proposition 5.6 (Link with the Dirac-Frenkel variational principle)**.** *Let $(C,\Phi) \in \partial\mathscr{F}_{N,K}$ satisfying $\mathscr{S}_0$ on $[0,T]$. Then, $\Psi = \pi(C,\Phi)$ satisfies*

$$\Big\langle i\frac{\partial\Psi}{\partial t} - \mathscr{H}\Psi \,|\, \delta\Psi \Big\rangle = 0, \quad \textit{for all} \quad \delta\Psi \in \mathbf{T}_\Psi\mathscr{B}_{N,K}. \tag{5.40}$$

Proof. We start with the characterization (5.29) of the elements in $\mathbf{T}_\Psi\mathscr{B}_{N,K}$. Since the full-rank assumption is satisfied in $[0,T]$, the orbitals satisfy (5.38), and therefore $\frac{\partial\phi_k}{\partial t} \in \mathrm{Span}\{\Phi\}^\perp$ for all $t \in [0,T]$ and all k's. On the one hand, being given $\sigma \in \Sigma_{N,K}$, we have

$$\begin{aligned}
\Big\langle i\frac{\partial\Psi}{\partial t} - \mathscr{H}\Psi \,\Big|\, \frac{\partial\Psi}{\partial c_\sigma}\Big\rangle &= i\sum_\tau \frac{dc_\tau}{dt}\Big\langle \Phi_\tau \,\Big|\, \Phi_\sigma\Big\rangle - \Big\langle \mathscr{H}\Psi \,\Big|\, \Phi_\sigma\Big\rangle \\
&\quad + i\sum_\tau c_\tau \Big\langle \frac{\partial\Phi_\tau}{\partial t} \,\Big|\, \Phi_\sigma\Big\rangle, \qquad (5.41)\\
&= i\frac{dc_\sigma}{dt} - \Big\langle \mathscr{H}\Psi \,\Big|\, \Phi_\sigma\Big\rangle = 0,
\end{aligned}$$

thanks to the equation satisfied by c_σ. Indeed,

$$\frac{\partial\Phi_\tau}{\partial t} = \sum_{k=1}^K \frac{\partial\Phi_\tau}{\partial\phi_k}\big[\frac{\partial\phi_k}{\partial t}\big],$$

and therefore the sum in (5.41) vanishes thanks to Lemma 5.2. On the other hand, for every $1 \le k \le K$ and for any function ζ in $\mathrm{Span}\{\Phi\}^\perp$, we have

$$\begin{aligned}
&\Big\langle i\frac{\partial\Psi}{\partial t} - \mathscr{H}\Psi\Big|\frac{\partial\Psi}{\partial\phi_k}[\zeta]\Big\rangle \\
&= i\sum_\sigma \frac{dc_\sigma}{dt}\Big\langle \Phi_\sigma \,|\, \frac{\partial\Psi}{\partial\phi_k}[\zeta]\Big\rangle + i\sum_{j=1}^K \Big\langle \frac{\partial\Psi}{\partial\phi_j}\big[\frac{\partial\phi_j}{\partial t}\big] \,\Big|\, \frac{\partial\Psi}{\partial\phi_k}[\zeta]\Big\rangle \qquad (5.42)\\
&\quad -\Big\langle \mathscr{H}\Psi \,\Big|\, \frac{\partial\Psi}{\partial\phi_k}[\zeta]\Big\rangle, \\
&= \Big\langle i\Big((\mathbf{\Gamma}\big(C(t)\big)\cdot\frac{\partial\Phi}{\partial t}\Big)_k - \frac{\partial\Psi^\star}{\partial\phi_k}[\mathscr{H}\Psi]\,,\,\zeta\Big\rangle, \\
&= -\Big\langle \mathbf{P}_\Phi\frac{\partial\Psi^\star}{\partial\phi_k}[\mathscr{H}\Psi]\,,\,\zeta\Big\rangle = 0. \qquad (5.43)
\end{aligned}$$

Indeed, on the one hand, in virtue of Lemma 5.2, the first term in the right-hand side of (5.42) vanishes whereas the second one identifies with $i\sum_{j=1}^K \mathbf{\Gamma}_{kj}(C)\langle\frac{\partial\phi_j}{\partial t},\zeta\rangle = \Big\langle \big(\mathbf{\Gamma}\big(C(t)\big)\cdot\frac{\partial\Phi}{\partial t}\big)_k\,;\,\zeta\Big\rangle$ since $\frac{\partial\phi_j}{\partial t}$ and ζ both belong to $\mathrm{Span}\{\Phi\}^\perp$. On the other hand, the last line (5.43) is obtained using the equation satisfied by Φ in $\mathscr{S}_0$ and by observing that $\mathbf{P}_\Phi\,\zeta = 0$ since $\zeta \in \mathrm{Span}\{\Phi\}^\perp$. The proof is complete. □

Let us now recall the definition of the energy

$$\mathscr{E}(\Psi) = \mathscr{E}\big(\pi(C,\Phi)\big) = \Big\langle \mathscr{H}\Psi \,|\, \Psi\Big\rangle.$$

It is clear that $\mathscr{E}(\Psi)$ depends on time *via* $(C(t),\Phi(t))$. As a corollary to Proposition 5.6 we have the following

Corollary 5.1 (Conservation of Energy by the flow)**.** *Let $T > 0$ and $(C,\Phi) \in \partial\mathscr{F}_{N,K}$ satisfying $\mathscr{S}_0$ on $[0,T]$ and such that $\Phi(t)$ lies in the domain of $\mathscr{H}$ for all t in $[0,T]$. Then,*

$$\mathscr{E}\big(\pi(C(t),\Phi(t))\big) = \mathscr{E}\big(\pi(C^0,\Phi^0)\big) \quad \text{on } [0,T].$$

Proof. We first observe that $\frac{\partial\Psi}{\partial t} \in \mathbf{T}_\Psi\mathscr{B}_{N,K}$, for

$$\frac{\partial\Psi}{\partial t} = \sum_\sigma \frac{dc_\sigma}{dt}\Phi_\sigma + \frac{1}{N}\sum_\sigma\sum_{k=1}^K c_\sigma \frac{\partial\Phi_\sigma}{\partial\phi_k}\Big[\frac{\partial\phi_k}{\partial t}\Big].$$

Then it is indeed of the form of the elements of $\mathbf{T}_\Psi\mathscr{B}_{N,K}$ according to (5.29) since $\frac{\partial\phi_k}{\partial t}$ lies in $\mathrm{Span}\{\Phi\}^\perp$ whenever $\mathbf{\Gamma}(t)$ is invertible. Then, applying Proposition 5.6 with $\delta\Psi = \frac{\partial\Psi}{\partial t}$ one obtains

$$0 = \Re\Big\langle i\frac{\partial\Psi}{\partial t} - \mathscr{H}\Psi \,\Big|\, \frac{\partial\Psi}{\partial t}\Big\rangle = -\Re\Big\langle \mathscr{H}\Psi \,\Big|\, \frac{\partial\Psi}{\partial t}\Big\rangle = -\frac{1}{2}\frac{d}{dt}\Big\langle \mathscr{H}\Psi \,\Big|\, \Psi\Big\rangle. \tag{5.44}$$

Hence the result. □

5.3.2 An *a posteriori* error estimate

In this section, we will establish an $L^2(\Omega)^N$ error bound for the MCTDHF approximation compared with the exact solution to the linear $N-$particle Schrödinger equation (5.2). Let us introduce the projector $\mathscr{P}_{T_\Psi\mathscr{B}}$ onto the tangent space $\mathbf{T}_\Psi\mathscr{B}_{N,K}$ to $\mathscr{B}_{N,K}$ at Ψ. Then, we claim

Lemma 5.5. *Given an initial data $(C^0,\Phi^0) \in \partial\mathscr{F}_{N,K}$ and an exact solution Ψ_E to the N-particle Schrödinger equation* (5.2)*. Then, as long as (C,Φ) is a solution to $\mathscr{S}_0$ in $\partial\mathscr{F}_{N,K}$, we have*

$$\|\Psi_E - \Psi\|_{L^2(\Omega^N)} \le \|\Psi_E^0 - \Psi^0\|_{L^2(\Omega^N)} + \left|\int_0^t (I - \mathscr{P}_{\mathbf{T}_\Psi\mathscr{F}})\,[\mathscr{H}\Psi(s)]\,ds\right|.$$

Proof. First, Proposition 5.6 expresses the fact that $\mathscr{P}_{\mathbf{T}_\Psi\mathscr{B}_{N,K}}\big(i\frac{\partial\Psi}{\partial t} - \mathscr{H}\Psi\big) = 0$. Therefore

$$i\frac{\partial\Psi}{\partial t} - \mathscr{H}\Psi = (I - \mathscr{P}_{\mathbf{T}_\Psi\mathscr{B}_{N,K}})\left[i\frac{\partial\Psi}{\partial t} - \mathscr{H}\Psi\right] = -(I - \mathscr{P}_{\mathbf{T}_\Psi\mathscr{B}_{N,K}})\,[\mathscr{H}\Psi], \tag{5.45}$$

since $\frac{\partial\Psi}{\partial t}$ lives in the tangent space $\mathbf{T}_\Psi\mathscr{B}_{N,K}$. Next, subtracting 5.45 from 5.2, we get

$$i\frac{\partial(\Psi_E - \Psi)}{\partial t} - \mathscr{H}(\Psi_E - \Psi) = -(I - \mathscr{P}_{\mathbf{T}_\Psi\mathscr{B}_{N,K}})\,[\mathscr{H}\Psi]$$

Then, we apply the PDE above to $\Psi_E - \Psi$ and we integrate formally over Ω^N. The result follows by taking the imaginary of both sides and by using the self-adjointness of $\mathscr{H}$. □

Roughly speaking, the above lemma tells that the closer is $\mathscr{H}\Psi$ to the tangent space $T_\Psi\mathscr{B}_{N,K}$, the better is the MCTDHF approximation. Intuitively, this is true for large values of K. Let us mention that this bound was already obtained in [27] and it is probably far from being accurate.

5.3.3 Unitary Group Action on the Flow

As described above a time-dependent ansatz $\Psi(t) \in \mathscr{B}_{N,K}$ is the image of several elements of $\mathscr{F}_{N,K}$ by the projection π. Proposition 5.5 defines a unitary and transitive group action on the set of fibers of $\partial\mathscr{F}_{N,K}$. This action can be also be interpreted in terms of *Gauge transforms*, and we will come back on this important issue later on. There are infinitely many (equivalent) ways for lifting the time evolution of the wave-function on the basis $\mathscr{B}_{N,K}$ onto time evolution equations from one fiber to the over across a given section (that is prescribed by the choice of the initial data (C_0, Φ_0) in $\pi^{-1}(\Psi(0))$). In order to study mathematically the time evolution of the wave-function or to perform numerical codes, it is convenient to have at our disposal several explicit and equivalent representations for the MCTDHF equations over different sections and to understand how they are related to each other. An illustrating example is System $\mathscr{S}_0$ that is well suited for checking energy conservation and constraints propagation over the flow but that is badly adapted for proving existence of solutions for the Cauchy problem or for designing numerical codes. On the contrary, we prove below that this system is unitarily (or gauge-) equivalent to System (5.64) below on which our mathematical analysis will be performed and that we will be referred to as "working equations" in the following.

Before turning to the main theorem of this section, we first state some results on the flow of unitary $K \times K$ matrices whose (obvious) proofs are skipped. In all that follows $T > 0$ is fixed.

Lemma 5.6 (Flow of unitary transforms)**.** *Let $U_0 \in \mathscr{O}_K$ and let $U(t) \in C^1\big([0,T); \mathscr{O}_K\big)$ with $U(0) = U_0$. Then, $t \mapsto M(t) := -i\frac{dU^*}{dt}U$ defines a continuous family of $K \times K$ hermitian matrices, and for all $t > 0$, $U(t)$ is the unique solution to the Cauchy problem*

$$\begin{cases} i\dfrac{dU}{dt} = U(t)M(t), \\ U(0) = U_0. \end{cases} \tag{5.46}$$

Conversely, if $t \mapsto M(t)$ is a continuous family of $K \times K$ Hermitian matrices and if $U_0 \in \mathscr{O}_K$ is given, then (5.46) defines a unique C^1 family of $K \times K$ unitary matrices.

Of particular interest will be for us the following two corollaries

Corollary 5.2. *Let $t \mapsto \mathbf{G}(t)$ be a continuous family of self-adjoint operators on $L^2(\Omega)$ and let $\Phi = (\phi_1(t), \phi_2(t), \ldots, \phi_K(t)) \in \mathscr{O}_{L^2(\Omega)^K}$ such that $\phi_i(t)$ belongs to the domain of $\mathbf{G}(t)$ for every $t \in [0,T)$. Then the $K \times K$ matrix M with entries $M_{ij}(t) = \langle \mathbf{G}(t)\phi_i(t)\,;\,\phi_j(t)\rangle$ is Hermitian and the Cauchy problem* (5.46) *defines a globally well-defined C^1 flow on the set of unitary $K \times K$ matrices.*

Remark 5.4. Actually when $\mathbf{G}$ is a non-negative self-adjoint operator on $L^2(\Omega)$, we rather impose the weaker condition for the ϕ_k's to lie in the form domain of $\mathbf{G}$. This remark will be used without mentioning again in the sequel. When $\mathbf{G}$ is the Laplacian or, more generally a one-body Schrödinger operator, this compatibility condition reduces to $\phi_i \in H^1(\mathbb{R}^3)$ or, say, $\phi_i \in H^1_0(\Omega)$ when Ω is a bounded domain. (Other boundary conditions could of course be considered.)

Corollary 5.3. *Let (N,K) be an admissible pair, let $t \mapsto M(t)$ be a continuous family of $K \times K$ Hermitian matrices and let $U^0 \in \mathscr{O}_K$. Then, if $t \mapsto U(t)$ denotes the unique family of unitary $K \times K$*

matrices that solves (5.46)*, the unitary $r \times r$ matrix $\mathbb{U}$ given by* (5.21) *is the unique solution to the differential equation*

$$\begin{cases} i\dfrac{d\mathbb{U}}{dt} = \mathbb{U}\mathbb{M}, \\ \\ \mathbb{U}(0) = d\big(U^0\big), \end{cases} \tag{5.47}$$

with

$$\mathbb{M}_{\sigma,\tau} = \sum_{\substack{i\in\sigma,\, j\in\tau \\ \sigma\setminus\{i\}=\tau\setminus\{j\}}} (-1)^{\sigma^{-1}(i)+\tau^{-1}(j)} M_{ij}. \tag{5.48}$$

In addition, when M is obtained through a self-adjoint operator $\mathbf{G}$ as in the statement of Corollary 5.2, then (5.48) *turns into*

$$\mathbb{M}_{\sigma,\tau} = \sum_{i=1}^{N} \big\langle \mathbf{G}_{x_i} \Phi_\sigma \big| \Phi_\tau \big\rangle. \tag{5.49}$$

Proof. For σ and τ given it is convenient to denote by $\mathbb{U}_{\sigma,\tau(j)}$ the column vector in $\mathbb{C}^N$ with entries $\big(\mathbb{U}_{\sigma(i),\tau(j)}\big)_{1\le i\le N}$ and by

$$[\mathbb{U}_{\tau(1)}, \mathbb{U}_{\tau(2)}, \dots, \mathbb{U}_{\tau(N)}]_\sigma$$

the determinant composed with these vectors. With this notation (5.46) gives

$$i\frac{d\mathbb{U}_{\sigma,\tau(j)}}{dt} = \sum_{k=1}^{K} M_{k,\tau(j)}\, \mathbb{U}_{\sigma,k} \tag{5.50}$$

Differentiating the relation

$$\mathbb{U}_{\sigma,\tau} = [U_{\tau(1)}, U_{\tau(2)}, \dots, U_{\tau(N)}]_\sigma$$

and using the multilinearity with respect to the column vectors and Eqn. (5.50) one obtains:

$$i\,\frac{d\mathbb{U}_{\sigma,\tau}}{dt} = \sum_{\substack{1\le k\le K \\ 1\le j\le N}} M_{k,\tau(j)} [U_{\tau(1)}, U_{\tau(2)}, \dots, U_{\tau(j-1)}, U_k, U_{\tau(j+1)}, \dots, U_{\tau(N)}]_\sigma. \tag{5.51}$$

On the other hand since $\mathbb{U}(t)$ is a flow of unitary matrices it is solution, of a differential equation of the following type:

$$i\,\frac{d\mathbb{U}_{\sigma,\tau}}{dt} = \sum_{\tau'} [U_{\tau'(1)}, U_{\tau'(2)}, \dots, U_{\tau'(N)}]_\sigma \tilde{\mathbb{M}}_{\tau',\tau} \tag{5.52}$$

Identification of the coefficients of

$$[U_{\tau'(1)}, U_{\tau'(2)}, \dots, U_{\tau'(N)}]_\sigma$$

gives, taking in account the number of permutation needed to change

$$\tau(1), \tau(2), \dots \tau(j-1), k, \tau(j+1), \dots \tau(N) \text{ into } \tau'(1), \tau'(2) \dots, \tau'(N)$$

$$\tilde{\mathbb{M}}_{\tau',\tau} = \sum_{\substack{k\in\tau',\, j\in\tau \\ \tau'\setminus\{k\}=\tau\setminus\{j\}}} M_{k,j} (-1)^{\tau^{-1}(j)+\tau'^{-1}(k)}.$$

Let us now prove (5.49). Let $\sigma, \tau \in \Sigma_{N,K}$. We first observe that

$$\sum_{i=1}^{N} \mathbf{G}_{x_i} \Phi_\sigma = \sum_{i=1}^{N} \phi_{\sigma(1)} \wedge \ldots \wedge \mathbf{G}\,\phi_{\sigma(i)} \wedge \ldots \wedge \phi_{\sigma(N)}. \tag{5.53}$$

Now we use (5.8) and the Laplace method to develop a determinant with respect to the row that contains the terms involving $\mathbf{G}$ to get

$$\begin{aligned} \sum_{i=1}^{N} \big\langle \mathbf{G}_{x_i} \Phi_\sigma \big| \Phi_\tau \big\rangle &= \sum_{i=1}^{N} \big\langle \phi_{\sigma(1)} \wedge \ldots \wedge \mathbf{G}\,\phi_{\sigma(i)} \wedge \ldots \wedge \phi_{\sigma(N)} \big| \Phi_\tau \big\rangle \\ &= \sum_{i,j=1}^{N} (-1)^{i+j} \langle \mathbf{G}\,\phi_{\sigma(i)}; \phi_{\tau(j)} \rangle\, \delta_{\sigma\setminus\{\sigma(i)\},\tau\setminus\{\tau(j)\}}, \end{aligned}$$

in virtue of (5.7). Hence (5.49) using (5.48) and the definition of M. □

We are now able to state the main result of this section.

Theorem 5.1 (Flow in different gauge)**.** *Let $U_0 \in \mathscr{O}_K$, $(C_0, \Phi_0) \in \partial \mathscr{F}_{N,K}$ and let $t \mapsto \mathbf{G}(t)$ be a family of self-adjoint operators in $L^2(\Omega)$.*

Assume that there exists a solution $(C, \Phi) \in C^0\big(0, T; \partial \mathscr{F}_{N,K}\big)$ of $\mathscr{S}_0$ with initial data (C_0, Φ_0) such that $t \mapsto \langle \mathbf{G}(t)\phi_i(t); \phi_j(t) \rangle$ is continuous on $[0,T]$ for every $1 \le i, j \le K$. Define the family of unitary transforms $U(t) \in C^1\big([0,T); \mathscr{O}_K\big)$ that satisfy (5.46) *with $M_{ij} = \langle \mathbf{G}\,\phi_i; \phi_j \rangle$ as in Corollary* (5.2)*. Then the couple $(C', \Phi') = \mathscr{U}(t) \cdot \big(C; \Phi\big)$ with $\mathscr{U}(t) = \big(d(U(t)); U(t)\big)$ defined by* (5.23) *and* (5.21) *is solution to the system $\mathscr{S}_{\mathbf{G}}$ defined by*

$$(\mathscr{S}_{\mathbf{G}}) \begin{cases} i\,\dfrac{dC'}{dt} = \Big\langle \mathscr{H}\,\Psi \,|\, \nabla_{C'} \Psi \Big\rangle - \Big\langle \displaystyle\sum_{i=1}^{N} \mathbf{G}_{x_i}\,\Psi \,|\, \nabla_{C'} \Psi \Big\rangle, \\ i\,\mathbf{\Gamma}(C')\,\dfrac{\partial \Phi'}{\partial t} = \mathbf{\Gamma}(C')\,\mathbf{G}\,\Phi' + (\mathbf{I} - \mathbf{P}_{\Phi'})\,\nabla_{\Phi'} \Psi^\star \Big[\mathscr{H}\,\Psi - \displaystyle\sum_{i=1}^{N} \mathbf{G}_{x_i}\,\Psi \Big], \\ \big(C'(0), \Phi'(0)\big) = \mathscr{U}_0 \cdot (C_0, \Phi_0) \end{cases}$$

with $\Psi = \pi(C, \Phi) = \pi(C', \Phi')$, $\mathscr{U}_0 = \big(U_0, d(U_0)\big) \in \mathscr{O}_K^r$ being defined by (5.23) *and with $\mathbb{M}$ being the $r \times r$ hermitian matrix given by* (5.48)*.*

Remark 5.5. Passing from $\mathscr{S}_0$ to $\mathscr{S}_{\mathbf{G}}$ amounts to change the operator $\mathscr{H}$ by $\mathscr{H} - \sum_{i=1}^{N} \mathbf{G}_{x_i}$ in both equations and by adding the linear term $\mathbf{\Gamma}(C')\,\mathbf{G}\,\Phi'$ in the equation satisfied by Φ'. Note that whereas solutions to $\mathscr{S}_0$ in $\partial \mathscr{F}_{N,K}$ satisfy

$$i \big\langle \frac{\partial \phi_i}{\partial t}; \phi_j \big\rangle = 0,$$

for all $1 \le i, j \le K$, we have

$$i \big\langle \frac{\partial \phi_i'}{\partial t}; \phi_j' \big\rangle = \langle \mathbf{G}\,\phi_i'; \phi_j' \rangle. \tag{5.54}$$

This is what the index G (for "gauge) is referring to in physicists language.

The proof of this theorem is postponed until the end of this subsection and we rather state before some corollaries or remarks. Observing that, to every Hermitian matrix M, one can associate a self-adjoint operator $\mathbf{G}$ in $L^2(\Omega)$ such that $M_{ij} = \langle \mathbf{G}\phi_i ; \phi_j \rangle$ by demanding that

$$\mathbf{G}\,\phi_i = \sum_{j=1}^{K} M_{ij}\,\phi_j \text{ for all } 1 \le i \le K.$$

we state below Theorem 5.2 that is an equivalent version of Theorem 5.1. Somehow, whereas Theorem 5.2 adopts the terminology of differential geometry where unitary equivalent equations are defined on sections of the fiber bundle, the previous theorem is rather stated in the language of physicists that choose *a priori* a gauge field on the orbitals (that is, the operator $\mathbf{G}$), and then express the variational principle (5.40) under the constraints $\Psi = \pi(C,\Phi) \in \mathscr{B}_{N,K}$ with Φ satisfying (5.54) for all t. This procedure formally provides with the gauge-dependent system of equations $\mathscr{S}_{\mathbf{G}}$.

Theorem 5.2 (Flow over different sections)**.** *Let $U_0 \in \mathscr{O}_K$ and let $(C_0,\Phi_0) \in \partial\mathscr{F}_{N,K}$.*

(i) Let $t \mapsto M(t)$ be a continuous family of $K\times K$ Hermitian matrices on $[0,T]$ and let $U(t) \in C^1\big([0,T);\mathscr{O}_K\big)$ be the corresponding solution to(5.46)*. Assume that there exists a solution $(C,\Phi) \in C^0\big(0,T;\partial\mathscr{F}_{N,K}\big)$ of $\mathscr{S}_0$ with initial data (C_0,Φ_0). Then, the couple $(C',\Phi') = \mathscr{U}(t)\cdot(C,\Phi)$ with $\mathscr{U} \in \mathscr{O}_K^r$ defined by* (5.23) *and* (5.21) *is solution to the system $\mathscr{S}_M$ defined by*

$$\begin{cases} i\,\dfrac{dC'}{dt} = \Big\langle \mathscr{H}\,\Psi \,|\, \nabla_{C'}\Psi \Big\rangle - \mathbb{M}'\,C', \\[2ex] i\,\mathbf{\Gamma}(C')\,\dfrac{\partial\Phi'}{\partial t} = (\mathbf{I}-\mathbf{P}_{\Phi'})\,\nabla_{\Phi'}\Psi^\star[\mathscr{H}\,\Psi] + \mathbf{\Gamma}(C')\,M'\,\Phi' \\[2ex] \big(C'(0),\Phi'(0)\big) = \mathscr{U}_0\cdot(C_0,\Phi_0) \end{cases} \tag{5.55}$$

with $\Psi = \pi(C,\Phi) = \pi(C',\Phi')$, $\mathscr{U}_0 = \big(U_0, d(U_0)\big) \in \mathscr{O}_K^r$ being defined by (5.23) *and with*

$$M' = UMU^\star, \quad \overline{\mathbb{M}}' = \mathbb{U}\mathbb{M}\mathbb{U}^\star,$$

where $\mathbb{M}$ be the $r\times r$ hermitian matrix with entries given by (5.48)*. Moreover, if $\mathbf{\Gamma}(C') = U\,\mathbf{\Gamma}(C)\,U^\star$ is invertible on $[0,T)$, then for all $1 \le i,j \le K$,*

$$i\,\big\langle \frac{\partial\phi_i'}{\partial t} ; \phi_j' \big\rangle = M_{ij}'.$$

(ii) Conversely, assume that there exists a solution $(C,\Phi) \in C^0\big(0,T;\partial\mathscr{F}_{N,K}\big)$ of $\mathscr{S}_0$ with initial data (C_0,Φ_0) and let $U(t) \in C^1\big([0,T);\mathscr{O}_K\big)$. Then, the couple $(C',\Phi') = \mathscr{U}(t)\cdot(C,\Phi)$ with $\mathscr{U} \in \mathscr{O}_K^r$ defined by (5.23) *and* (5.21) *is solution to the system $\mathscr{S}_M$ with $M(t) = -i\,\frac{dU^*}{dt}\,U$.*

Remark 5.6 (Lagrangian interpretation and gauge theory)**.** The equations can also be derived (at least formally) thanks to the Lagrangian formulation: One writes the stationarity condition for the action

$$\mathscr{A}(\Psi) = \int_0^T \Big\langle \Psi \Big| i\frac{\partial}{\partial t} - H_N \Big| \Psi \Big\rangle\, dt$$

over functions $\Psi = \Psi(t)$ that move on $\mathscr{F}_{N,K}$. The associated time-dependent Euler–Lagrange equations take the form (5.55) with $\Psi = \pi(C,\Phi)$, M an hermitian matrix and with $\mathbb{M}$ be the $r \times r$ hermitian matrix linked to M through Eqn. (5.48) above. As observed already by Cancès and Lebris [10], even if they appear so, the Hermitian matrices M and $\mathbb{M}$ should not be interpreted as time-dependent Lagrange multipliers associated to the constraints $(C,\Phi) \in \mathscr{F}_{N,K}$ since the constraints on the coefficients and the orbitals are automatically propagated by the dynamics (see Lemma 5.4), but rather as degrees of freedom within the fiber at Ψ. This is what physicists call the *gauge invariance* of the equations. In particular, this gauge invariance can be used to set M and $\mathbb{M}$ to zero for all t as observed in Lemma 5.2 and Eqn. 5.56 below, so that the above system can be transformed into the simpler system $(\mathscr{S}_0)$ we started with.

Theorem 5.1 and Corollary 5.2 provide with a differential equation that has to satisfy a unitary matrix $U(t)$ to transform $\mathscr{S}_0$ into $\mathscr{S}_{\mathbf{G}}$. A direct calculation shows that, given two self-adjoint one-particle operators $\mathbf{G}$ and $\mathbf{G}'$, the solution to

$$\begin{cases} i\dfrac{dU(t)}{dt} & = U(t)\, M_{G\to G'}, \\ \\ U(t=0) & = U^0 \end{cases} \tag{5.56}$$

with $\Big(M_{G\to G'}\Big)_{ij} = \big\langle (\mathbf{G}-\mathbf{G}')\,\phi_i\,;\,\phi_j\big\rangle$ maps a solution to $\mathscr{S}_{\mathbf{G}}$ to a solution to $\mathscr{S}_{\mathbf{G}'}$. In particular, if we prove existence of solutions for the system $\mathscr{S}_{\mathbf{G}}$ for some operator $\mathbf{G}$ then we have existence of solutions for any system $\mathscr{S}_{\mathbf{G}'}$. Another immediate though crucial consequence of Theorem 5.1 and Theorem 5.2 is given in Corollary 5.4 below. It states that for whatever choice of gauge the constraints on the expansion coefficients and on the orbitals are propagated by the flow and the energy is kept constant since it is the case for the system $\mathscr{S}_0$. Also the rank of the first-order density matrices does not depend on the gauge.

Corollary 5.4 (Gauge transforms and conservation properties)**.** *Let $T > 0$. Let $\mathbf{G}$ be a self-adjoint (possibly time-dependent) operator acting on $L^2(\Omega)$. Assume that there exists a solution to the system $\mathscr{S}_G$ on $[0,T]$ such that* $\operatorname{rank}\boldsymbol{\Gamma}\big(C(t)\big) = K$ *and such that the matrix $\big\langle \mathbf{G}\phi_i;\phi_j\big\rangle_{1\le i,j\le K}$ is continuous. Then, for all $t \in [0;T]$,*

$$(C(t),\Phi(t)) \in \partial\mathscr{F}_{N,K},$$

and the energy is conserved, that is

$$\mathscr{E}\big(\pi(C(t),\Phi(t))\big) = \mathscr{E}\big(\pi(C(0),\Phi(0))\big).$$

In addition, $\Psi = \pi(C,\Phi)$ satisfies the Dirac-Frenkel variational principle (5.40)*.*

Proof of Corollary 5.4. From the one hand, by Theorem 5.1 and its remark, if (C,Φ) satisfies $\mathscr{S}_{\mathbf{G}}$ with initial data in $\partial\mathscr{F}_{N,K}$, there exists a family of unitary transforms $U \in C^1\big(0,T;\mathscr{O}_K\big)$ such that $(C,\Phi) = \mathscr{U}\cdot(C',\Phi')$ where (C',Φ') satisfies $\mathscr{S}_0$ with same initial data. By Lemma 5.4, $\mathscr{S}_0$ preserves $\mathscr{F}_{N,K}$, hence so does $\mathscr{S}_{\mathbf{G}}$ since U and $\mathbb{U} = d(U)$ are unitary. From the other hand, by Lemma 5.5, we have $\pi(C,\Phi) = \pi(C',\Phi') = \Psi$, thus the energy is conserved by the flow and Eqn. (5.40) is satisfied since $\mathbf{T}_\Psi\mathscr{B}_{N,K}$ only depends on the point Ψ on the basis $\mathscr{B}_{N,K}$ and not on the chosen preimages on the fiber $\pi^{-1}(\Psi)$. □

A natural requirement could be to write down the equations on a section of the fiber bundle on which the Hermitian matrix $\mathbf{\Gamma}$ is kept diagonal for all time. Explicit though complicated form of the corresponding change of gauge is due to [7] and we have the following

Lemma 5.7 (Diagonal density matrix)**.** *Let (C,Φ) satisfying $\mathscr{S}_0$ with initial data (C_0,Φ_0) and let $U_0 \in \mathscr{O}_K$ that diagonalizes $\mathbf{\Gamma}(C_0)$. We assume that for all time the eigenvalues of $\mathbf{\Gamma}(C)$ are simple, that is $\gamma_i \neq \gamma_j$ for $1 \leq i,j \leq K$ and $i \neq j$. Define a $K \times K$ Hermitian matrix by*

$$M_{ij} = \begin{cases} \dfrac{1}{\gamma_j - \gamma_i} \Big\langle \mathscr{H}\,\Psi \,|\, \dfrac{\partial \Psi}{\partial \phi_i}[\phi_j] \Big\rangle - \Big\langle \dfrac{\partial \Psi}{\partial \phi_j}[\phi_i] \,|\, \mathscr{H}\,\Psi \Big\rangle & \text{if } i \neq j, \\ \\ 0 & \text{otherwise}, \end{cases}$$

and consider the family $t \mapsto U(t) \in \mathscr{O}_K$ that satisfies (5.46) *with $U(t=0) = U_0$. Then $(C',\Phi') = \mathscr{U}(t)\cdot(C,\Phi)$ is solution to*

$$\begin{cases} i\,\dfrac{dC'}{dt} = \Big\langle \mathscr{H}\,\Psi \,|\, \nabla_{C'}\Psi \Big\rangle - \mathbb{M}'\,C', \\ \\ i\,\gamma_i\,\dfrac{\partial \phi_i'}{\partial t} = (\mathbf{I} - \mathbf{P}_{\Phi'})\,\dfrac{\partial \Psi^\star}{\partial \phi_i'}\,[\mathscr{H}\,\Psi] + \gamma_i\,M'\,\Phi', \\ \\ \big(C'(0),\Phi'(0)\big) = \mathscr{U}_0 \cdot (C_0,\Phi_0) \end{cases}$$

with the notation of Theorem 5.2. In particular, $\mathbf{\Gamma}(C') = diag\big(\gamma_1(t),\ldots,\gamma_K(t)\big)$ for every t.

Proof. Using the equation for the coefficients in $\mathscr{S}_M$ together with (5.17), the evolution equation for the coefficients of the density matrix writes

$$\begin{aligned} i\,\frac{d\gamma_{ij}}{dt} &= \sum_{\substack{\sigma,\tau\,:\,i\in\sigma,\ j\in\tau \\ \sigma\setminus\{i\}=\tau\setminus\{j\}}} (-1)^{\sigma^{-1}(i)+\tau^{-1}(j)} \big[\langle \mathscr{H}\,\Psi \,|\, c_\sigma\,\Phi_\tau\rangle - \langle c_\tau\,\Phi_\sigma \,|\, \mathscr{H}\,\Psi\rangle\big] \\ &+ \sum_{\substack{\kappa,\sigma,\tau\,:\,i\in\sigma,\ j\in\tau \\ \sigma\setminus\{i\}=\tau\setminus\{j\}}} (-1)^{\sigma^{-1}(i)+\tau^{-1}(j)} \big[\mathbb{M}_{\sigma,\kappa}\,c_\kappa\,\bar{c}_\tau - \mathbb{M}_{\kappa,\tau}\,\bar{c}_\kappa\,c_\sigma\big]. \end{aligned}$$

$$\begin{aligned} i\,\frac{d\gamma_{ij}}{dt} &= \Big\langle \mathscr{H}\,\Psi \,|\, \frac{\partial \Psi}{\partial \phi_i}\,[\phi_j] \Big\rangle - \Big\langle \frac{\partial \Psi}{\partial \phi_j}\,[\phi_i] \,|\, \mathscr{H}\,\Psi \Big\rangle \\ &\quad - \sum_{k=1}^{K} \Big\{ \mathbf{\Gamma}_{ik}\,M_{kj} - M_{ik}\,\mathbf{\Gamma}_{kj} \Big\}. \end{aligned}$$

Next, we require that

$$\gamma_{ij}(t) = \gamma_i\,\delta_{i,j}, \quad \text{that is} \quad \frac{d\mathbf{\Gamma}_{ij}}{dt} = 0 \quad \forall\, 1 \leq i \neq j \leq K.$$

Using the above equation, a sufficient condition is given by

$$M_{i,j} = \frac{1}{\gamma_{i,i} - \gamma_{j,j}} \Big[\Big\langle \mathscr{H}\,\Psi \,|\, \frac{\partial \Psi}{\partial \phi_i}[\phi_j] \Big\rangle - \Big\langle \frac{\partial \Psi}{\partial \phi_j}[\phi_i] \,|\, \mathscr{H}\,\Psi \Big\rangle \Big].$$

This achieves the proof. □

We now turn to the common proof of Theorem 5.1 and Theorem 5.2.

Proof of Theorem 5.1 and Theorem 5.2. Let $(C(t),\Phi(t))$ be a solution to $\mathscr{S}_0$ and let $\mathbf{G}$ be as in the statement of the theorem. With $M_{ij} = \langle \mathbf{G}\,\phi_i\,;\,\phi_j\rangle$ we define the family of unitary transforms $U(t)$ according to Corollary 5.2 and $d(U)(t) = \overline{\mathbb{U}}(t)$ is then given by Corollary 5.3. We set $\mathbb{V} = \overline{\mathbb{U}}$, $C'(t) = \mathbb{V}(t)\,C(t)$ and $\Phi'(t) = U(t)\Phi(t)$. Thanks to (5.47), $\mathbb{V}$ solves

$$\begin{cases} i\dfrac{d\mathbb{V}}{dt} = -\mathbb{V}\,\overline{\mathbb{M}}, \\ \\ \mathbb{V}(0) = d\big(U^0\big). \end{cases} \tag{5.57}$$

Then, for all $\sigma \in \Sigma_{N,K}$,

$$\begin{aligned} i\,\frac{dC'}{dt} &= i\,\frac{d\mathbb{V}}{dt}\,C + \mathbb{V}\,i\frac{dC}{dt} = -\mathbb{V}\,\overline{\mathbb{M}}\,\mathbb{V}^\star\,C' + \mathbb{V}\,\big\langle \mathscr{H}\,\Psi\,\big|\,\nabla_C\Psi\big\rangle, \\ &= -\mathbb{V}\,\overline{\mathbb{M}}\,\mathbb{V}^\star\,C' + \mathbb{V}\,\big\langle \mathscr{H}\,\Psi\,\big|\,\nabla_{C'}\Psi\,\mathbb{V}\big\rangle, \end{aligned}$$

thanks to (5.31) and (5.57). On the one hand, since $\mathbb{V}$ is unitary,

$$\mathbb{V}\,\big\langle \mathscr{H}\,\Psi\,\big|\,\nabla_{C'}\Psi\,\mathbb{V}\big\rangle = \big\langle \mathscr{H}\,\Psi\,\big|\,\nabla_{C'}\Psi\big\rangle.$$

On the other hand, when M is obtained through $\mathbf{G}$, we get by a direct calculation from (5.49)

$$\Big(\mathbb{V}\,\overline{\mathbb{M}}\,\mathbb{V}^\star\,C'\Big)_\sigma = \sum_\tau \Big\langle \sum_{i=1}^N \mathbf{G}_{x_i}\,\Phi'_\tau\Big|\Phi'_\sigma\Big\rangle\,c'_\tau = \Big\langle \sum_{i=1}^N \mathbf{G}_{x_i}\,\Psi\Big|\Phi'_\sigma\Big\rangle.$$

Combining these two facts we get the first equation in $\mathscr{S}_{\mathbf{G}}$, namely

$$i\,\frac{dC'}{dt} = \Big\langle \mathscr{H}\,\Psi\Big|\nabla_{C'}\Psi\Big\rangle - \Big\langle \sum_{i=1}^N \mathbf{G}_{x_i}\,\Psi\Big|\nabla_{C'}\Psi\Big\rangle.$$

We turn now to the equation satisfied by Φ'. To simplify the notation we use the shorthand $\mathbf{\Gamma}$ for $\mathbf{\Gamma}(C)$ and $\mathbf{\Gamma}'$ for $\mathbf{\Gamma}(C')$ respectively. Then, using $\mathbf{\Gamma}' = U\,\mathbf{\Gamma}\,\mathbb{U}^\star$ and (5.46), we have

$$\begin{aligned} i\,\mathbf{\Gamma}'\,\frac{\partial\Phi'}{\partial t} &= \mathbf{\Gamma}'\,i\,\frac{dU}{dt}\,\Phi + \mathbf{\Gamma}'\,U\,i\,\frac{\partial\Phi}{\partial t}, \\ &= \mathbf{\Gamma}'\,UMU^\star\,\Phi' + U\,\mathbf{\Gamma}\,i\,\frac{\partial\Phi}{\partial t}, \\ &= \mathbf{\Gamma}'\,UMU^\star\,\Phi' + (\mathbf{I}-\mathbf{P}_{\Phi'})\,U\,\nabla_\Phi\Psi^\star\big[\mathscr{H}\,\Psi\big], \\ &= \mathbf{\Gamma}'\,UMU^\star\,\Phi' + (\mathbf{I}-\mathbf{P}_{\Phi'})\,\nabla_{\Phi'}\Psi^\star\big[\mathscr{H}\,\Psi\big], \end{aligned} \tag{5.58}$$

thanks to (5.32) and since clearly $\mathbf{P}_{\Phi'} = \mathbf{P}_\Phi$ for $\mathrm{Span}\{\Phi\} = \mathrm{Span}\{\Phi'\}$. It is easily checked that when M is given through $\mathbf{G}$ we have

$$\big(UMU^\star\big)_{ij} = \langle \mathbf{G}\,\phi'_i;\phi'_j\rangle,$$

and therefore

$$UMU^\star\,\Phi' = \mathbf{P}_{\Phi'}\,\mathbf{G}\,\Phi'.$$

Hence (5.58) also writes

$$i\,\mathbf{\Gamma}'\,\frac{\partial \Phi'}{\partial t} \;=\; \mathbf{\Gamma}'\,\mathbf{G}\Phi' + (\mathbf{I}-\mathbf{P}_{\Phi'})\,\nabla_{\Phi'}\Psi^\star\big[\mathscr{H}\,\Psi\big] - (\mathbf{I}-\mathbf{P}_{\Phi'})\,\mathbf{\Gamma}'\,\mathbf{G}\,\Phi'.$$

We now check that, for all $1 \le i \le N$,

$$(\mathbf{I}-\mathbf{P}_\Phi)\,\big(\mathbf{\Gamma}\,\mathbf{G}\,\Phi\big)_i = (\mathbf{I}-\mathbf{P}_\Phi)\,\frac{\partial \Psi^\star}{\partial \phi_i}\,\Big[\sum_{j=1}^N \mathbf{G}_{x_j}\,\Psi\Big].$$

(the primes being suppressed here to simplify notation), thereby proving that

$$i\,\mathbf{\Gamma}'\,\frac{\partial \Phi'}{\partial t} = \mathbf{\Gamma}'\,\mathbf{G}\,\Phi' + (\mathbf{I}-\mathbf{P}_{\Phi'})\,\nabla_{\Phi'}\Psi^\star\Big[\mathscr{H}\,\Psi - \sum_{i=1}^N \mathbf{G}_{x_i}\,\Psi\Big].$$

Indeed, for all $\xi \in L^2(\Omega)$, thanks to (5.33) in Lemma 5.2 in (5.59) and (5.53) in (5.60), we have

$$\begin{aligned}
\Big\langle\,(\mathbf{I}-\mathbf{P}_\Phi)\,\big(\mathbf{\Gamma}\,\mathbf{G}\,\Phi\big)_i;\xi\Big\rangle \;&=\; \sum_{k=1}^K \mathbf{\Gamma}_{ik}\,\big\langle\,\mathbf{G}\,\phi_k;(\mathbf{I}-\mathbf{P}_\Phi)\,\xi\big\rangle,\\
&=\; \sum_{k=1}^K \Big\langle\,\frac{\partial \Psi}{\partial \phi_k}[\mathbf{G}\,\phi_j]\,;\,\frac{\partial \Psi}{\partial \phi_i}[(\mathbf{I}-\mathbf{P}_\Phi)\,\xi]\,\Big\rangle, \qquad (5.59)\\
&=\; \Big\langle\,\sum_{j=1}^N \mathbf{G}_{x_j}\,\Psi\,;\,\frac{\partial \Psi}{\partial \phi_i}[(\mathbf{I}-\mathbf{P}_\Phi)\,\xi]\,\Big\rangle, \qquad (5.60)\\
&=\; \Big\langle\,(\mathbf{I}-\mathbf{P}_\Phi)\,\frac{\partial \Psi^\star}{\partial \phi_i}\,\big[\sum_{j=1}^N \mathbf{G}_{x_j}\,\Psi\big]\,;\,\xi\,\Big\rangle,
\end{aligned}$$

by the definition (5.30) of $\frac{\partial \Psi^\star}{\partial \phi_i}$; whence the result since ξ is arbitrary in $L^2(\Omega)$. □

5.3.4 Standing waves solutions

In this section we search for particular solutions to the system $\mathscr{S}_M$ whose corresponding wavefunction is of the form $\Psi(t,x) = e^{-i\lambda t}\Psi(x)$ with $\lambda \in \mathbb{R}$ and $\Psi \in \mathscr{B}_{N,K}$, by analogy with the definition of standing waves for the exact Schrödinger equation. For $\Psi \in \partial\mathscr{B}_{N,K}$, the corresponding fiber is obtained through $\Psi(t,x) = \pi(C',\Phi')$ with $(C',\Phi') = \mathscr{U}(t)\cdot(e^{-i\lambda t}C,\Phi)$, where $(C,\Phi) \in \mathscr{F}_{N,K}$ is fixed, independent of time, and $\mathscr{U}(t) \in \mathscr{O}_K^r$. Inserting (C',Φ') in $\mathscr{S}_M$ for an arbitrary $K\times K$ Hermitian matrix $M = M(t)$ and using the formulas (5.31) and (5.32) for the changes of variables, we arrive at

$$\begin{cases}
\Big(i\,\dfrac{d\overline{\mathbb{U}}(t)}{dt} + \mathbb{M}\,\overline{\mathbb{U}} + \lambda\,\overline{\mathbb{U}}\Big)\,C = \overline{\mathbb{U}}\,\Big\langle\,\mathscr{H}\,\Psi\,|\,\nabla_C\Psi\Big\rangle,\\[2ex]
\mathbf{\Gamma}(C)\,\Big(i\,U^\star\,\dfrac{dU}{dt} - U^\star M U\Big)\,\Phi = (\mathbf{I}-\mathbf{P}_\Phi)\,\nabla_\Phi\Psi^\star\,[\mathscr{H}\,\Psi],\\[2ex]
U(0) = I_K.
\end{cases}$$

Note that in the above system $\Psi = \pi(C,\Phi)$ and $\mathbf{\Gamma}(C)$ are independent of time, and we may restrict to the case when $\mathbf{\Gamma}(C)$ is invertible (by only considering occupied orbitals). We start with the

equation satisfied by Φ. Observing that the left-hand side lives in $\mathrm{Span}\{\Phi\}$ whereas the right-hand side lives in $\mathrm{Span}\{\Phi\}^{\perp}$, we conclude that there are both equal to zero. Therefore, there exists a $K \times K$ matrix Λ that is independent of t and such that

$$\nabla_{\Phi}\Psi^{\star}[\mathscr{H}\Psi] = \Lambda \cdot \Phi.$$

In the case when the operator $\mathscr{H}$ is given by (5.63) below this equation writes

$$\mathbf{\Gamma}(C)\,\mathbf{H}\,\Phi + \mathbb{W}[C,\Phi]\,\Phi = \Lambda \cdot \Phi \tag{5.61}$$

(see Lemma (5.8) below). Also since the left-hand side has to be independent of t we get

$$i\frac{dU}{dt} = MU.$$

Comparing now with the equation for the coefficients we infer from Corollary 5.3 that

$$i\frac{d\overline{\mathbb{U}}(t)}{dt} = -\mathbb{M}\,\overline{\mathbb{U}},$$

hence

$$\left\langle \mathscr{H}\Psi \,|\, \nabla_C \Psi \right\rangle = \lambda\, C. \tag{5.62}$$

Equations (5.61) and (5.62) are precisely the MCHF equations that are satisfied by critical points of the energy and that were derived by Lewin [25]. The real λ is the Lagrange multiplier corresponding to the constraint $C \in S^{r-1}$ where the Hermitian matrix Λ is the matrix of Lagrange multipliers corresponding to the orthonormality constraints on the orbitals. In [24] Le Bris has proved the existence of ground-states - that is, minima of the energy over the set $\mathscr{F}_{N,K}$ - for the physical Hamiltonian (5.1), on the whole space $\mathbb{R}^3$, and under the assumptions $K = N+2$ and $\sum_{m=1}^{M} z_m > N-1$. Later on Friesecke extended this result to general admissible pairs (N,K), under the same assumption on the nuclear charge and for one- and two- potentials U and V that belong to the Kato class (that contains the Coulomb-type potentials)

$$\begin{aligned} L^{3/2}(\mathbb{R}^3) + L^{\infty}_{\varepsilon}(\mathbb{R}^3) := \{ & v \in L^{3/2}(\mathbb{R}^3) + L^{\infty}(\mathbb{R}^3) \,: \\ & \forall \varepsilon > 0,\, \exists v_1 \in L^{3/2}(\mathbb{R}^3),\, v_2 \in L^{\infty}(\mathbb{R}^3),\, v = v_1 + v_2,\, \|v_2\|_{\infty} \leq \varepsilon. \} \end{aligned}$$

(This is the closure of the C^{∞} functions with compact support for the norm of $L^{3/2}(\mathbb{R}^3) + L^{\infty}(\mathbb{R}^3)$.) Finally Lewin proved the existence of infinitely many critical points of the MCHF energy for any pairs (N,K), hence the existence of infinitely many solutions to the coupled system (5.61) – (5.62) that satisfy the full-rank assumption. All these solutions then give rise to infinitely many standing waves of the MCTDHF system and thereby of particular global-in-time solutions.

5.3.5 N-body Schrödinger type operators with two-body interactions

So far we have considered a generic Hamiltonian $\mathscr{H}$ and we have written down a coupled evolution system of equations for this operator. At this point, we consider more specifically an Hamiltonian in $L^2(\Omega^N)$ of the following form

$$\mathscr{H}_N \Psi = \sum_{i=1}^{N} \mathbf{H}_{x_i} \Psi + \sum_{1 \leq i < j \leq N} V(x_i, x_j)\, \Psi. \tag{5.63}$$

In the definition above, $\mathbf{H}$ is a self-adjoint operator acting on $L^2(\Omega)$ To fix ideas we take $\mathbf{H} = -\frac{1}{2}\Delta + U(x)$. As usual, the subscript x_i means that the operator is acting on the i^{th} space variable and it will be omitted when there is no confusion, and V is a real multiplicative operator acting on $(x,y) \in \Omega^2$ such that

$$V(x,y) = V(y,x), \quad \forall\, x,y \in \Omega.$$

We sometimes use the shorthand $V_{i,j}$ for $V(x_i,x_j)$ and we denote $\mathscr{V} = \sum_{1\le i<j\le N} V_{i,j}$. Now, expanding the expression of $\mathscr{H}$ in the system $\mathscr{S}_0$ and arguing as in the proof of Theorem 5.1 we obtain

$$\mathscr{S}_0 : \quad \begin{cases} i\,\dfrac{dC}{dt} = \Big\langle \sum\limits_{i=1}^{N} \mathbf{H}_{x_i}\,\Psi \,|\, \nabla_C \Psi \Big\rangle + \Big\langle \mathscr{V}\,\Psi \,|\, \nabla_C \Psi \Big\rangle, \\ \\ i\,\mathbf{\Gamma}(C)\,\dfrac{\partial \Phi}{\partial t} = (\mathbf{I} - \mathbf{P}_\Phi)\,\nabla_\Phi \Psi^\star \Big[\mathscr{V}\,\Psi + \sum\limits_{i=1}^{N} \mathbf{H}_{x_i}\,\Psi \Big]. \end{cases}$$

Comparing with System $\mathscr{S}_{\mathbf{G}}$ in Theorem 5.1, one observes that with $\mathbf{G} = \mathbf{H}$ one get an equivalent system of the form

$$\mathscr{S}_{\mathbf{H}} : \quad \begin{cases} i\,\dfrac{dC}{dt} &= \Big\langle \mathscr{V}\,\Psi \,|\, \nabla_C \Psi \Big\rangle, \\ \\ i\,\mathbf{\Gamma}(C)\,\dfrac{\partial \Phi}{\partial t} &= \mathbf{\Gamma}(C)\,\mathbf{H}\,\Phi + (\mathbf{I} - \mathbf{P}_\Phi)\,\nabla_\Phi \Psi^\star [\mathscr{V}\,\Psi], \end{cases} \tag{5.64}$$

(provided $t \mapsto \langle \mathbf{H}\phi_i ; \phi_j \rangle$ makes sense). From Corollary 5.4 we know that if the initial data in (5.64) lies in $\mathscr{F}_{N,K}$ then the solutions of the system remain so for all time. This allows to recast the equations in (5.64) as follows.

Lemma 5.8. *Let $\Psi = \pi(C,\Phi) \in \mathscr{B}_{N,K}$. Then, for all $\sigma \in \Sigma_{N,K}$*

$$\Big\langle \mathscr{V}\,\Psi \,|\, \nabla_C \Psi \Big\rangle = \mathbb{K}[\Phi]\,C,$$

and

$$(\mathbf{I} - \mathbf{P}_\Phi)\,\nabla_\Phi \Psi^\star [\mathscr{V}\,\Psi] = (\mathbf{I} - \mathbf{P}_\Phi)\,\mathbb{W}[C,\Phi]\,\Phi,$$

where $\mathbb{K}[\Phi]$ (resp. $\mathbb{W}[C,\Phi]$) is a $r \times r$ (resp. $K \times K$) Hermitian matrix with entries

$$\mathbb{K}[\Phi]_{\sigma,\tau} = \frac{1}{2} \sum_{i,j\in\tau,\, k,l\in\sigma} \delta_{\tau\setminus\{i,j\},\sigma\setminus\{k,l\}} (-1)^{\tau}_{i,j}\,(-1)^{\sigma}_{k,l}\, D_V\big(\phi_i\,\bar\phi_k\,,\,\bar\phi_j \phi_l\big), \tag{5.65}$$

and

$$\mathbb{W}[C,\Phi]_{ij}(x) = 2 \sum_{k,l=1}^{K} \gamma_{jkil}\,\big(\phi_k\,\bar\phi_l \star_\Omega V\big), \tag{5.66}$$

where here and below we denote

$$D_V(f,g) = \iint_{\Omega\times\Omega} V(x,y)\,f(x)\,\overline{g}(y)\,dxdy,$$

$$f \star_\Omega V = \int_\Omega V(\cdot,y)\,f(y)\,dy,$$

and with the coefficients γ_{ijkl} being defined by (5.15) *in Proposition 5.3 .*

Proof. Let $\sigma,\,\tau\in\Sigma_{N,K}$ and let $1\le p<q\le N$ be given. By developping the determinants Φ_τ and Φ_σ with respect to the columns that contain the variables x_p and x_q, we get

$$\begin{aligned}
&\Phi_\tau(x_1,\dots,x_N)\\
&\quad= \frac{1}{\sqrt{N(N-1)}}\sum_{\substack{i,j=1\\ i\neq j}}^{N}\phi_{\tau(i)}(x_p)\,\phi_{\tau(j)}(x_q)\,(-1)^{i+p+q-1+p_i(j)}\,\Phi^{(p,q)}_{\sigma\setminus\{\sigma(i),\sigma(j)\}},\\
&\quad= \sqrt{\frac{2}{N(N-1)}}\sum_{1\le i<j\le N}\big(\phi_{\tau(i)}\wedge\phi_{\tau(j)}\big)(x_p,x_q)\,(-1)^{i+p+q+j}\,\Phi^{(p,q)}_{\sigma\setminus\{\sigma(i),\sigma(j)\}},
\end{aligned}$$

where $p_i(j)=j$ if $j<i$ or $=j-1$ if $j>i$ and where $\Phi^{(p,q)}_{\sigma\setminus\{\sigma(i),\sigma(j)\}}$ denotes the $(N-2)\times(N-2)$ Slater determinant that is built from $\sigma\setminus\{\sigma(i),\sigma(j)\}$ and with the variables x_p and x_q omitted in $(x_1,\dots,x_N)$. Therefore, using (5.7) for the scalar product of Slater determinants,

$$\begin{aligned}
\Big\langle V(x_p,x_q)\,\Phi_\tau|\Phi_\sigma\Big\rangle \;&=\; \frac{2}{N(N-1)}\sum_{\substack{1\le i<j\le N\\ 1\le k<l\le N}}(-1)^{i+j+k+l}\,\delta_{\sigma\setminus\{\sigma(k),\sigma(l)\},\tau\setminus\{\tau(i),\tau(j)\}}\times\\
&\qquad\times\Big\langle V(x,y)\,\phi_{\tau(i)}\wedge\phi_{\tau(j)}\,\Big|\,\phi_{\sigma(k)}\wedge\phi_{\sigma(l)}\Big\rangle.
\end{aligned}$$

Since it is independent of p and q, the term $\mathbb{K}[\Phi]_{\sigma,\tau}=\sum_{1\le p<q\le N}\Big\langle V(x_p,x_q)\,\Phi_\tau\;|\,\Phi_\sigma\Big\rangle$ equals $N(N-1)/2$ times such a term; hence (5.65).

We now turn to the proof of (5.66). First, for every $1\le i\le K$,

$$\frac{\partial\Psi^\star}{\partial\phi_i}\,[\mathscr{V}\,\Psi]=\sum_{\sigma,\tau}c_\sigma\,\bar c_\tau\,\frac{\partial\Phi^\star_\tau}{\partial\phi_i}[\mathscr{V}\,\Phi_\sigma]=\sum_{\sigma,\tau\,:\,i\in\tau}c_\sigma\,\bar c_\tau\,\frac{\partial\Phi^\star_\tau}{\partial\phi_i}[\mathscr{V}\,\Phi_\sigma],\tag{5.67}$$

in virtue of (5.27). Next, we assume $i\in\tau$, and we argue by duality according to (5.30) by fixing $\xi\in L^2(\Omega)$. Then, arguing as above, we obtain

$$\begin{aligned}
&\Big\langle(\mathbf{I}-\mathbf{P}_\Phi)\,\frac{\partial\Phi^\star_\tau}{\partial\phi_i}[\mathscr{V}\,\Phi_\sigma]\,;\,\xi\Big\rangle=\Big\langle\mathscr{V}\,\Phi_\sigma\,\Big|\,\frac{\partial\Phi_\tau}{\partial\phi_i}\big[(\mathbf{I}-\mathbf{P}_\Phi)\,\xi\big]\Big\rangle=\\
&\quad=\sum_{\substack{1\le k<l\le N\\ 1\le\tau^{-1}(i),j\le N}}(-1)^{k+l+\tau^{-1}(i)+j}\,\delta_{\sigma\setminus\{\sigma(k),\sigma(l)\},\tau\setminus\{i,\tau(j)\}}\times\\
&\qquad\qquad\times\Big\langle V(x,y)\,\phi_{\sigma(k)}\wedge\phi_{\sigma(l)}\,\Big|\,\big[(\mathbf{I}-\mathbf{P}_\Phi)\,\xi\big]\wedge\phi_{\tau(j)}\Big\rangle.
\end{aligned}$$

All other terms for which $(\mathbf{I}-\mathbf{P}_\Phi)\,\xi$ does not act on the variables x_p or x_q vanish by definition of $\mathbf{I}-\mathbf{P}_\Phi$. We now identify in the above formula the terms that are the scalar producted with ξ, and we observe that they appear two times interchanging the rôle played by the variables x and y.

Since ξ is arbitrary in $L^2(\Omega)$ we then get

$$\begin{aligned}(\mathbf{I}-\mathbf{P}_\Phi)\,\frac{\partial\Phi^\star_\tau}{\partial\phi_i}[\mathscr{V}\,\Phi_\sigma] \\ = \quad (\mathbf{I}-\mathbf{P}_\Phi)\sum_{\substack{1\le k,l\le N\\ 1\le\tau^{-1}(i),j\le N}}(-1)^{k+p_k(l)+\tau^{-1}(i)+j}\delta_{\sigma\setminus\{\sigma(k),\sigma(l)\},\tau\setminus\{i,\tau(j)\}}\times \\ \times\phi_{\sigma(k)}\left(\phi_{\sigma(l)}\bar{\phi}_{\tau(j)}\star_\Omega V\right).\end{aligned} \tag{5.68}$$

We now insert (5.68) into (5.67) and we compare with the formulas (5.15) for the coefficients of the second-order density matrix to obtain

$$(\mathbf{I}-\mathbf{P}_\Phi)\,\frac{\partial\Phi^\star_\tau}{\partial\phi_i}[\mathscr{V}\,\Phi_\sigma] = (\mathbf{I}-\mathbf{P}_\Phi)\sum_{j=1}^{K}\mathbb{W}[C,\Phi]_{i,j}\,\phi_j,$$

with $\mathbb{W}[C,\Phi]_{i,j}$ given by (5.66). This achieves the proof of the lemma. □

As a corollary of the above lemma and within this choice of gauge, the system now writes

$$\left\{\begin{array}{rcl} i\dfrac{dC}{dt} &=& \mathbb{K}[\Phi]\,C,\\[2ex] i\,\mathbf{\Gamma}(C)\,\dfrac{\partial\Phi}{\partial t} &=& \mathbf{\Gamma}(C)\,\mathbf{H}\,\Phi+(\mathbf{I}-\mathbf{P}_\Phi)\,\mathbb{W}[C,\Phi]\,\Phi,\\[2ex] \big(C(0),\Phi(0)\big) &=& \big(C^0,\Phi^0\big)\in\mathscr{F}_{N,K},\\[2ex] \Psi &=& \pi(C,\Phi).\end{array}\right. \tag{5.69}$$

It is equivalent to System (5.64) as long as the solution lies in $\mathscr{F}_{N,K}$. This system will be referred to as *working equations* according to [9, 23]. The equations satisfied by the orbitals form a coupled system of non-linear Schrödinger-type equations that is better adapted for well-posedness analysis as will be seen in the forthcoming subsection.

Before that, we first treat apart in the last two subsections the special cases of the *free system* and of the *time-dependent Hartree–Fock equations*(TDHF in short).

5.3.6 Free Systems - interacting Systems

In this section we consider free systems for which the interaction potential V is switched off. Then the system (5.64) becomes

$$\left\{\begin{array}{l} i\,\dfrac{dC}{dt}=0,\\[2ex] i\,\mathbf{\Gamma}(C)\,\dfrac{\partial\Phi}{\partial t}=\mathbf{\Gamma}(C)\,\mathbf{H}\,\Phi.\end{array}\right.$$

From the first equation the coefficients c_σ's are constant during the evolution. In particular the full-rank assumption is satisfied for all time whenever it is satisfied at start. In that case the orbitals satisfy K independent linear Schrödinger equations through

$$i\,\frac{\partial \Phi}{\partial t} = \mathbf{H}\,\Phi, \tag{5.70}$$

and the N-particle wave-function $\Psi = \pi(C,\Phi)$ solves the exact Schrödinger equation

$$\begin{cases} \quad i\dfrac{\partial \Psi}{\partial t} = \displaystyle\sum_{i=1}^{N} \mathbf{H}_{x_i}\,\Psi, \\ \\ \quad \Psi(t=0) = \pi(C_0,\Phi_0). \end{cases} \tag{5.71}$$

Conversely, the unique solution to the Cauchy problem (5.71) with $(C_0,\Phi_0) \in \partial\mathscr{F}_{N,K}$ coincides with $\pi(C(t),\Phi(t)) \in \mathscr{F}_{N,K}$ where $\Phi(t)$ is the solution to (5.70). This is a direct consequence of the fact that the linear structure of (5.71) propagates the factorization of a Slater determinant. In particular, this enlightens the fact that the propagation of the full-rank assumption is intricately related to the non-linearities created by the interaction potential V between particles.

5.3.7 MCTDHF ($K = N$) contains TDHF

In this section we check that the usual time-dependent Hartree-Fock (TDHF) equations are indeed obtained as a special case of the above general setting by simply setting $K = N$. We consider the following *ansatz*

$$\Psi^{HF} = \phi_1 \wedge \ldots \wedge \phi_N,$$

for $\Phi = (\phi_1,\ldots,\phi_N)$ in $\mathscr{O}_{L^2(\Omega)^N}$. The TDHF equations write (up to a unitary transform)

$$i\,\frac{\partial \phi_i}{\partial t} = \mathbf{H}\,\phi_i + \mathscr{F}_\Phi\,\phi_i, \tag{5.72}$$

for $1 \le i \le N$, with $\mathscr{F}_\Phi$ being the self-adjoint operator on $L^2(\Omega)$ that is defined by

$$\mathscr{F}_\Phi\, w \;=\; \Big(\sum_{j=1}^{N}\int_\Omega V(\cdot,y)|\phi_j(y)|^2 dy\Big)\, w - \sum_{j=1}^{N}\Big(\int_\Omega V(\cdot,y)\overline{\phi}_j(y) w(y)\,dy\Big)\,\phi_j.$$

Existence and uniqueness of global-in-time solutions in the energy space $H^1(\Omega^N)$ is due to Chadam and Glassey [13]. They also checked by integrating the equations that the TDHF equations propagate the orthonormality of the orbitals and that the Hartree–Fock energy is preserved by the flow. Derivation of the TDHF equations from the Dirac-Frenkel variational principle may be encountered in standard Physics textbooks (see *e.g.* [28]). Let us also mention the work [10] by Cancès and LeBris who have investigated existence of solutions to TDHF equations including time-dependent electric field and that are coupled with nuclear dynamics.

By simply setting $K = N$ in the MCTDHF formalism one gets

$$\#\Sigma_{N,K} = 1\,,, \quad \mathbf{\Gamma}(t) = I_N, \tag{5.73}$$

and

$$\Psi(t) := C(t)\,\phi_1(t) \wedge \ldots \wedge \phi_N(t), \quad C(t) = e^{-i\theta_\Phi(t)},$$

for some $\theta_\Phi \in \mathbb{R}$. In addition according to Remark 5.2,

$$\gamma_{jkil} = \frac{1}{2}\left(\delta_{i,j}\,\delta_{k,l} - \delta_{i,k}\,\delta_{j,l}\right). \tag{5.74}$$

Therefore with the definitions (5.65) and (5.66)

$$\begin{aligned}
\mathbb{K}[\Phi] &= \left\langle \mathscr{V}\,\phi_1 \wedge \ldots \wedge \phi_N \,|\, \phi_1 \wedge \ldots \wedge \phi_N \right\rangle, \\
&= \sum_{i,j,k,l\,:\,\{i,j\}=\{k,l\}} (-1)^{i+p_i(j)+k+p_k(l)} D_V(\phi_i\,\bar{\phi}_k; \bar{\phi}_j\,\phi_l), \\
&= \sum_{i=1}^{N} \left\langle \mathscr{F}_\Phi \phi_i; \phi_i \right\rangle,
\end{aligned}$$

and

$$\sum_{j=1}^{N} \mathbb{W}[C,\Phi]_{i,j}\,\phi_j = \mathscr{F}_\Phi\,\phi_i.$$

Eventually for $N = K$, according to (5.69), the MCTDHF system in the working form turns out to be

$$\mathscr{S}_{\mathbf{H}}(N=K) \quad \left\{ \begin{array}{rcl}
\dfrac{d\theta_\Phi(t)}{dt} &=& \displaystyle\sum_{i=1}^{N} \left\langle \mathscr{F}_\Phi \phi_i; \phi_i \right\rangle, \\[2ex]
i\,\dfrac{\partial \phi_i}{\partial t} &=& \mathbf{H}\,\phi_i + (\mathbf{I} - \mathbf{P}_\Phi)\,\mathscr{F}_\Phi\,\phi_i \\
&=& \mathbf{H}\,\phi_i + \mathscr{F}_\Phi\,\phi_i - \displaystyle\sum_{j=1}^{N} \langle \mathscr{F}_\Phi\,\phi_i; \phi_j \rangle\,\phi_j, \\[2ex]
\theta_\Phi(0) = 0 \quad , \quad \Phi(0) = (\phi_1^0; \ldots; \phi_N^0),
\end{array} \right.$$

with $(\phi_1^0; \ldots; \phi_N^0) \in \mathscr{O}_{L^2(\Omega)^N}$. Comparing with (5.55), we introduce the $N \times N$ Hermitian matrix M with entries $M_{i,j} = -\langle \mathscr{F}_\Phi\,\phi_i; \phi_j \rangle$. According to Lemma 5.6 there exists a unique unitary matrix $U(t)$ such that

$$\left\{ \begin{array}{rcl}
i\,\dfrac{dU}{dt} &=& -U\,M, \\[2ex]
U(t=0) &=& I_N.
\end{array} \right.$$

In virtue of (5.21) the corresponding unitary matrix that transforms $\phi_1 \wedge \ldots \wedge \phi_N$ into $(U\phi_1) \wedge \ldots \wedge (U\phi_N)$ is then simply the complex number $\mathbb{U} = \det(U)$ that is of modulus 1 and that satisfies

$$\left\{ \begin{array}{rcl}
i\,\dfrac{d\mathbb{U}}{dt} &=& -\mathrm{tr}(M)\,\mathbb{U}, \\[2ex]
\mathbb{U}(t=0) &=& 1.
\end{array} \right. \tag{5.75}$$

Comparing (5.75) with the equation satisfied by $\theta_\Phi(t)$ in $\mathscr{S}_{\mathbf{H}}(N=K)$ it turns out that $\mathbb{U} = e^{i\theta_\Phi(t)}$. In that special case a change of gauge translates into multiplication by a global phase factor.

Applying Theorem 5.2, the functions ϕ_i', $1 \leq i \leq N$, defined by $\Phi' = U\Phi$ satisfy the standard Hartree–Fock equations (5.72) and $C'(t) = \overline{\mathbb{U}}C(t) = 1$ for all time; that is $\Psi = \phi_1' \wedge \ldots \wedge \phi_N'$. Being a special case of the MCTDHF setting we then deduce "for free" that the TDHF equations propagate the orthonormality of the initial data, that they satisfy the Dirac-Frenkel variational principle and that the flow keeps the energy constant.

5.4 Local-in-time analysis of the MCTDHF equations

In this section we focus on the N-body Schrödinger operator defined by (5.1) in the introduction, and we investigate the existence of solutions with finite energy. Recall that the energy is simply given by

$$\mathscr{E}(\Psi) = \Big\langle \mathscr{H}_N \Psi \,|\, \Psi \Big\rangle.$$

For the special Hamiltonian we are considering here and for $\Psi = \pi(C,\Phi)$ in $\mathscr{F}_{N,K}$, it may be recasted in the following equivalent forms [18, 25]

$$\begin{aligned}
\mathscr{E}(\Psi) &= \mathscr{E}\big(\pi(C,\Phi)\big), \\
&= \Big\langle \Big(\mathbf{H}\boldsymbol{\Gamma} + \frac{1}{2}\mathbb{W}[C,\Phi]\Big)\Phi, \Phi \Big\rangle_{L^2}, \qquad (5.76)\\
&= \int_{\mathbb{R}^3} \Big(\boldsymbol{\Gamma}(C)\,\Phi; \mathbf{H}\,\Phi\Big)\,dx + \iint_{\mathbb{R}^3\times\mathbb{R}^3} \frac{[\Psi\otimes\Psi]_{:2}(x,y,x,y)}{|x-y|}\,dxdy, \\
&= \sum_{i,j=1}^{K} \gamma_{ij} \int_{\mathbb{R}^3} \left[\frac{1}{2}\nabla\phi_i \nabla\bar{\phi}_j + U\,\phi_i\bar{\phi}_j\right] dx + \sum_{i,j,k,l=1}^{K} \gamma_{ijkl}\, D\big(\phi_l\,\bar{\phi}_i\,;\,\phi_k\,\bar{\phi}_j\big).
\end{aligned}$$

Everything below carries through more general potentials real-valued U and V in $L^2(\mathbb{R}^3)+L^\infty(\mathbb{R}^3)$ with $V \geq 0$. These conditions ensure that $\mathscr{H}_N$ is self-adjoint in $L^2(\Omega^N)$, that the Kato inequality holds and that the energy space is $\mathbb{C}^r \times H^1(\mathbb{R}^3)^K$ (respectively $\mathbb{C}^r \times H^1_0(\Omega)^K$ when Ω is a bounded domain). In the following section we will impose further conditions on the potentials that ensure that the energy functional is weakly lower semi-continuous on the energy space (although when Ω is not bounded, $\Psi \mapsto \mathscr{E}(\Psi)$ is NOT weakly lower semi-continuous on $H^1(\Omega^N)$; see [18]). Sufficient conditions are (for example) $U \geq 0$ or $U \in L^p(\Omega)$ with $p > 3/2$ when Ω is bounded, or U_- in the Kato class for the whole space (in that latter case the negative part of U has to go to 0 at infinity at least in a weak sense). This assumptions are fulfilled by the potentials in the class $L^p(\mathbb{R}^3)+L^q(\mathbb{R}^3)$ with $3/2 < p,q \leq +\infty$ considered in [30, 34]. The extension to some time-dependent potentials is discussed in Section 5.7. Nevertheless all proofs below are mainly detailed for Coulomb potentials.

From the mathematical point of view, the MCTDHF methods, compared to the TDHF for instance, are far from being known and understood. To our knowledge, the first analytical result has been obtained in [23] in the MCTDH framework for bosons which is similar to the MCTDHF from the analysis point of view although more complicated from the algebraic point of view for several density-matrices have to be considered (see Section 5.8). Under a full-rank assumption on one-body density matrices, the authors proved short-time existence and uniqueness of solutions in the functions space $H^2(\mathbb{R}^3)$ with the help of Lie commutators techniques. Their Hamiltonian

do not include any exterior potential U and features a regular and bounded interaction potential V between the bosons. The analysis that we develop below provides with a H^1-theory for the MCTDHF equations that include Coulomb type interactions. Of course our result remains valid in the case of bounded potentials and for H^2 initial data, and therefore it extends the previous result of [23]. Theorem 5.3 below tells us that starting from an initial data whose associated density matrix $\mathbf{\Gamma}\big(C^0\big)$ is invertible, then the dynamics of $\mathscr{S}_{\mathbf{H}}$ propagates this property, by continuity, at least for a short time. The well-posedness is proved in this lapse of time. There is no mathematical proof nor numerical evidence whether a loss of rank might happen or not in finite time. In the following section we provide with a new sufficient (although strong) condition on the initial data that ensures the full-rank of the density matrix for all time, thereby allowing for global-in-time solutions (in addition to the standing waves).

5.4.1 Local well-posedness of the working equations in the energy space

Our main result in this section is the following

Theorem 5.3. *Let $(C^0,\Phi^0)\in\partial\mathscr{F}_{N,K}$. Let Φ^0 be in $H^1(\mathbb{R}^3)^K$ or in $H^1_0(\Omega)^K$ when $\Omega\neq\mathbb{R}^3$. Then, there exists a maximal existence time $T^\star>0$ (possibly $+\infty$) such that:*

(i) The MCTDHF system (5.69) *admits a unique solution (C,Φ) with*

$$C\in C^1\big([0,T^\star);\mathbb{C}^r\big),\qquad \Phi\in C^0\big([0,T^\star);H^1(\mathbb{R}^3)^K\big)\cap C^1\big([0,T^\star);H^{-1}(\mathbb{R}^3)^K\big).$$

This solution depends continuously on the initial data (C^0,Φ^0).
For every $0\leq t<T^\star$,

(ii) $\big(C(t),\Phi(t)\big)\in\partial\mathscr{F}_{N,K}\quad$ *and* $\quad\mathbf{\Gamma}\big(C(t)\big)\in GL_K(\mathbb{C})$.

(iii) The energy is conserved :

$$\Big\langle\,\mathscr{H}_N\Psi_{\rm MC}(t)\Big|\Psi_{\rm MC}(t)\Big\rangle=\Big\langle\,\mathscr{H}_N\Psi^0\Big|\Psi^0\Big\rangle\quad\text{ with }\Psi_{\rm MC}=\pi(C,\Phi).$$

(iv) Either $T^\star=+\infty$ or $T^\star<+\infty$ and $\limsup_{t\nearrow T^\star}\|\mathbf{\Gamma}(C)(t)^{-1}\|=+\infty$. In this latter case, we actually have

$$\int_0^{T^\star}\|\mathbf{\Gamma}\big(C(t)\big)^{-1}\|^{3/2}dt=+\infty.$$

From now on, we shall use the Euclidian norms for the vectors C and Φ

$$\|C\|^2:=\sum_{\sigma\in\Sigma_{N,K}}|c_\sigma|^2,\quad\|\Phi\|_{L^p}^2:=\sum_{i=1}^K\|\phi_i\|_{L^p(\mathbb{R}^3)}^2,$$

and similarly for other functional spaces. Moreover, for a $m\times m$ matrix M we use the Frobenius norm

$$\|M\|=\sqrt{\sum_{i,j=1}^m|M_{i,j}|^2}.$$

Let $X = \mathbb{C}^r \times \big(H^1(\mathbb{R}^3)^K\big)$ and, for every $T > 0$, $X_T = C^0\big([0,T];X\big)$ endowed with the respective norms

$$\|(C,\Phi)\|_X = \|C\| + \|\Phi\|_{H^1}, \quad \|(C,\Phi)\|_{X_T} = \sup_{t\in[0,T]} \|\big(C(t),\Phi(t)\big)\|_X.$$

The first important remark (that was already pointed out in [23]) is that the matrix elements of $\mathbf{\Gamma}_\Phi$ only depend on the $\binom{K}{N}$-component vector C through the algebraic expression (5.17). Of course, for $(C,\Phi) \in \mathscr{F}_N^K$, $\mathbf{\Gamma}(C) = \mathbf{\Gamma}_\Phi$ (see Remark 5.1). The application $C \mapsto \mathbf{\Gamma}(C)$ is quadratic in C, thus locally Lipschitz continuous. Then, given a vector C^0 such that $\mathbf{\Gamma}\big(C^0\big)$ is invertible, the matrix $\mathbf{\Gamma}(C)$ remains invertible for C in a neighborhood of C^0. Consequently, starting from an initial data $(C^0,\Phi^0) \in \mathscr{F}_{N,K}$ such that $\mathbf{\Gamma}\big(C^0\big)$ is invertible, there exists a positive (possibly infinite) time T (at least $T << 1$) such that if the flow of the MCTDHF system ensures the existence of a solution $(C(t),\Phi(t))$ depending continuously on the initial data for $t \in [0,T]$, then the associated density matrix $\mathbf{\Gamma}\big(C(t)\big)$ will stay in a neighborhood of $\mathbf{\Gamma}\big(C^0\big)$ in the open subset of invertible matrices.

From now on, we assume as in the statement of Theorem 5.3 that the so-called "full-rank condition" holds at start; that is, $\mathbf{\Gamma}\big(C^0\big)$ is invertible. Thus, under the full-rank condition on $\mathbf{\Gamma}\big(C(t)\big)$ for $t \in [0,T)$ and $T > 0$, the Cauchy problem (5.69) is equivalent to

$$\begin{cases} i\dfrac{dC}{dt} = \mathbb{K}[\Phi]\,C, \\ \\ i\dfrac{\partial\Phi}{\partial t} = \mathbf{H}\Phi + \mathbb{L}[C,\Phi], \\ \\ \partial(C;\Phi) \in \mathscr{F}_{N,K}, \\ \\ C(0) = C^0, \quad \Phi(0) = \Phi^0. \end{cases} \tag{5.77}$$

where the non-linear part $\mathbb{L}[C,\Phi]$ is the $K-$component vector given by

$$\mathbb{L}[C,\Phi] = \mathbf{\Gamma}(C)^{-1}\,(\mathbf{I} - \mathbf{P}_\Phi)\mathbb{W}[C,\Phi]\Phi. \tag{5.78}$$

The proof of existence and uniqueness of a local-in-time solution of the Cauchy problem (5.77) is similar to the original proof of Chadam and Glassey for the TDHF model [13] and Koch and Lubich's approach for the MCTDH model [23]. For U being the Coulomb electron-nuclei interaction defined by (5.1), the Coulomb propagator $\exp[it\mathbf{H}]$ is a uniformly bounded one-parameter group of operators on $H^1(\mathbb{R}^3)$ [13]. Then, the MCTDHF initial-value problem may be recasted in the form of integral equations thanks to the Duhamel formula as follows

$$\begin{cases} C(t) &= C^0 - i\displaystyle\int_0^t \mathbb{K}[\Phi(s)]\,C(s)\,ds, \\ \\ \Phi(t) &= \exp[it\mathbf{H}]\Phi^0 - i\displaystyle\int_0^t \exp[i(t-s)\mathbf{H}]\,\mathbb{L}[C(s),\Phi(s)]\,ds. \end{cases} \tag{5.79}$$

Existence and uniqueness of a so-called *mild solution* is then ensured by Segal's Theorem [32] for abstract evolution equations by checking that the non-linearities $(C,\Phi) \mapsto \mathbb{K}[\Phi]\,C$ and $(C,\Phi) \mapsto$

$\mathbb{L}\big[C,\Phi\big]$ are locally bounded and locally Lipschitz continuous in a neighborhood of $\big(C^0,\Phi^0\big)$ in X_T for any $T>0$ small enough such that $\mathbf{\Gamma}\big(C(t)\big)$ is invertible on $[0,T]$. This is the purpose of Subsection 5.4.4 (Lemma 5.12, Lemma 5.13 and their corollaries). The equivalence of the integral formulation (5.79) and the initial value problem (5.77) is standard; see e.g. [31]. Next, thanks to Corollary 5.4, the constraint $\big(C(t),\Phi(t)\big)\in\mathscr{F}_{N,K}$ is satisfied for all $t\in[0,T]$. (Indeed, the proof of Corollary 5.4 heavily relies on the self-adjointness of $\mathbf{H}$ and keeps unchanged when $\mathbf{\Gamma}_\Phi$ is replaced by $\mathbf{\Gamma}(C)$.) Thereby we may identify again the matrix $\mathbf{\Gamma}(C)$ with the matrix representation of $\mathbf{\Gamma}_\Phi$ in $\mathrm{Span}\{\Phi\}$. Next, relying on the conservation of the energy ensured by Corollary 5.4 we prove the existence of the solution over a maximal time interval beyond which the density matrix degenerates (Subsection 5.4.5).

5.4.2 Step 1 - Properties of the operator $\mathbf{H}$

Following Chadam and Glassey [13], we claim

Lemma 5.9. *Let $Z=\sum_{1\le m\le M} z_m$. Then, there exists a Z-dependent positive constant, say γ_Z, such that $\mathbf{H}+\gamma_Z=-\Delta+U+\gamma_Z$ is a positive self-adjoint operator with domain $D(-\Delta)$, thus $(\mathbf{H}+\gamma_Z)^{\frac{1}{2}}$ exists as a positive self-adjoint operator with domain $D((-\Delta)^{\frac{1}{2}})$. Finally we have the following equivalence :*

$$\|\cdot\|_{H^1(\mathbb{R}^3)}\simeq\|\,(\mathbf{H}+\gamma_Z)^{\frac{1}{2}}\cdot\|.$$

Proof. In [13], the authors proved the result for $M=1$. The extension to the case of more nuclei is straightforward. □

Note that the statement remains valid for any potential U in the Kato class. As a corollary we obtain that the Coulomb propagator $\big\{\exp[it\mathbf{H}]\big\}_{t\in\mathbb{R}}$ is a uniformly bounded one-parameter group of operators in $H^1(\mathbb{R}^3)$. In particular, for every $T>0$, there exists a positive constant M_T (that only depends on $\mathscr{V}$) such that

$$\|e^{i(t-s)H}\,\varphi\|_{H^1(\mathbb{R}^3)}\le M_T\,\|\varphi\|_{H^1(\mathbb{R}^3)},\qquad\text{for all }\varphi\in H^1(\mathbb{R}^3)\text{ and }t,s\in[0,T].\tag{5.80}$$

5.4.3 Step 2: Local Lipschitz bounds

The main result in this subsection is the following.

Lemma 5.10 (A priori bounds, local Lipschitz bounds)**.** *Let $\big(C^0,\Phi^0\big)\in\partial\mathscr{F}_{N,K}$ with Φ in $\big(H^1(\mathbb{R}^3)^K\big)$. Then, the non-linearities $(C,\Phi)\mapsto\mathbb{K}[\Phi]C$ and $(C,\Phi)\mapsto\mathbb{L}\big[C,\Phi\big]$ in (5.79) are locally bounded and locally Lipschitz continuous in $\mathbb{C}^r\times\big(H^1(\mathbb{R}^3)^K\big)$ in a neighborhood of $\big(C^0,\Phi^0\big)$.*

Lemma 5.10 will follow as a corollary of Lemma 5.5 and Lemma 5.6 below.

Before turning to the proof of Lemma 5.10, we start with recalling a few technical lemmas about the Coulomb potential taken from [13]. The proof is a straightforward application of Cauchy–Schwarz' and Hardy's inequalities. For given $\Phi\in H^1(\mathbb{R}^3)^K$, let us introduce the $K\times K$ Hermitian matrix $\mathbb{V}[\Phi]$ with entries

$$\mathbb{V}[\Phi]_{ij}=\big(\phi_i\,\overline{\phi}_j\big)\star\frac{1}{|x|}.$$

Then, we have

Lemma 5.11 ([13], Lemma 2.3)**.** *Let $\phi, \psi \in H^1(\mathbb{R}^3)$, then $(\phi\psi) \star \frac{1}{r} \in W^{1,\infty}(\mathbb{R}^3)$, and we have*

$$\|(\phi\psi) \star \frac{1}{r}\|_{L^\infty} \leq 2\|\nabla\phi\|_{L^2(\Omega)}\|\psi\|_{L^2}, \tag{5.81}$$

and

$$\left\|\nabla\left((\phi\psi) \star \frac{1}{r}\right)\right\|_{L^\infty} \leq 4\|\nabla\phi\|_{L^2(\mathbb{R}^3)}\|\nabla\psi\|_{L^2}. \tag{5.82}$$

In particular, for all $\Phi \in H^1(\mathbb{R}^3)^K$,

$$\|\mathbb{V}[\Phi]\|_{\mathscr{M}_{K\times K}\left(W^{1,\infty}(\mathbb{R}^3)\right)} \lesssim \|\Phi\|_{H^1}^2, \tag{5.83}$$

where here and below $\lesssim$ is a shorthand for any universal bound that only depends on K and N, and the function $\Phi \mapsto \mathbb{V}[\Phi]$ is locally Lipschitz continuous from $(H^1(\mathbb{R}^3))^K$ to $\mathscr{M}_{K\times K}\left(W^{1,\infty}(\mathbb{R}^3)\right)$.

As a by-product of the above lemma the following local Lipschitz bounds hold.

Lemma 5.12. *Let Φ and $\tilde{\Phi}$ in $H^1(\mathbb{R}^3)^K$. Then we have*

$$\|\mathbb{K}[\Phi]\| \lesssim \|\Phi\|_{L^2}^3 \|\Phi\|_{H^1} \tag{5.84}$$

$$\|\mathbb{K}[\Phi] - \mathbb{K}[\tilde{\Phi}]\| \lesssim \left(\|\Phi\|_{L^2}^3 + \|\tilde{\Phi}\|_{L^2}^3\right)\|\Phi - \tilde{\Phi}\|_{H^1}. \tag{5.85}$$

In particular we have the following

Corollary 5.5. *The mapping $\left(C, \Phi\right) \mapsto \mathbb{K}[\Phi]C$ is locally Lipschitz continuous from $\mathbb{C}^r \times \left(H^1(\mathbb{R}^3)^K\right)$ to $\mathbb{C}^r$.*

Proof. First of all, for every $1 \leq i, j, k, l \leq K$,

$$\begin{aligned}\left|D(\phi_j\,\overline{\phi}_i\,,\,\phi_k\overline{\phi}_l)\right| &= \left|\left\langle \mathbb{V}_{j,i}\phi_l, \phi_k\right\rangle_{L^2(\Omega)}\right| \\ &\leq 2\|\nabla\phi_i\|_{L^2(\Omega)}\|\phi_j\|_{L^2(\Omega)}\|\phi_k\|_{L^2(\Omega)}\|\phi_l\|_{L^2(\Omega)}, \\ &\lesssim \|\Phi\|_{L^2}^3\|\Phi\|_{H^1},\end{aligned} \tag{5.86}$$

the last line being a direct consequence of (5.81). In particular this proves (5.84) since according to (5.65), $\mathbb{K}[\Phi]$ is a finite sum of terms of this kind. Next, we have clearly

$$\left|D(\phi_j\phi_i, \phi_l\phi_k) - D(\tilde{\phi}_j\tilde{\phi}_i, \tilde{\phi}_l\tilde{\phi}_k)\right| \lesssim \left(\|\Phi\|_{L^2}^3 + \|\tilde{\Phi}\|_{L^2}^3\right)\|\Phi - \tilde{\Phi}\|_{H^1}.$$

This proves the estimate (5.85) for the same reason. The local Lipschitz continuity of the mapping $\left(C, \Phi\right) \mapsto \mathbb{K}[\Phi]C$ follows immediately. □

We now turn to the proof of the local Lipschitz continuity of the non-linearity $(C, \Phi) \mapsto \mathbb{L}[C, \Phi]$ in a neighborhood of the initial data. We first claim and prove the following

Lemma 5.13. *Let $C^0 \in S^{r-1}$ with $\mathbf{\Gamma}(C^0)$ invertible. Then, there exists a positive constant ρ and a positive constant M depending only on ρ and C^0 such that for any $C \in \mathbb{C}^r$*

$$\|C - C^0\| \leq \rho \quad \Longrightarrow \quad \mathbf{\Gamma}(C) \in GL_K(\mathbb{C}) \text{ and } \quad \|\mathbf{\Gamma}(C)^{-1}\| \leq M. \tag{5.87}$$

For every Φ and $\tilde{\Phi}$ in $H^1(\mathbb{R}^3)^K$ and for every $C, \tilde{C} \in \mathbb{C}^r$ such that $\|C - C^0\| \leq \rho$ and $\|\tilde{C} - C^0\| \leq \rho$, we have

$$\|\mathbf{\Gamma}(C)^{-1} - \mathbf{\Gamma}(\tilde{C})^{-1}\| \lesssim \left(\|C\| + \|\tilde{C}\|\right) \left(\|\mathbf{\Gamma}(C)^{-1}\| + \|\mathbf{\Gamma}(\tilde{C})^{-1}\|\right) \|C - \tilde{C}\|, \tag{5.88}$$

$$\|\mathbf{P}_\Phi\|_{\mathscr{L}(H^1)} \lesssim \|\Phi\|_{L^2} \|\Phi\|_{H^1}, \tag{5.89}$$

$$\|\mathbf{P}_\Phi - \mathbf{P}_{\tilde{\Phi}}\|_{\mathscr{L}(H^1)} \lesssim \left(\|\Phi\|_{H^1} + \|\tilde{\Phi}\|_{H^1}\right) \|\Phi - \tilde{\Phi}\|_{H^1}, \tag{5.90}$$

$$\|\mathbb{W}[C, \Phi]\, \Phi\|_{H^1} \lesssim \|C\|^2 \|\Phi\|_{H^1}^3, \tag{5.91}$$

with $\mathscr{L}(H^1)$ denoting the set of linear operators on $H^1(\mathbb{R}^3)$.

$$\left(C, \Phi\right) \mapsto \mathbb{W}[C, \Phi]\, \Phi \text{ is locally Lipschitz continuous from } \mathbb{C}^r \times \left(H^1(\mathbb{R}^3)^K\right) \text{ to } \left(H^1(\mathbb{R}^3)\right)^K. \tag{5.92}$$

Proof. We first recall that invertible matrices form an open subset of $\mathscr{M}_{K\times K}(\mathbb{C})$ and that the mapping $M \mapsto M^{-1}$ is continuous and even locally Lipschitz continuous since

$$\begin{aligned} \|M^{-1} - \tilde{M}^{-1}\| &= \|M^{-1}\,(\tilde{M} - M)\,\tilde{M}^{-1}\| \\ &\leq \|M^{-1}\|\,\|\tilde{M}^{-1}\|\,\|M - \tilde{M}\|. \end{aligned}$$

In addition, being quadratic, the mapping $C \mapsto \mathbf{\Gamma}(C)$ is locally Lipschitz continuous with respect to C. We immediately deduce (5.87) and (5.88) by composition of locally bounded and locally Lipschitz functions.

Next, the operator $\mathbf{P}_\Phi$ is a sum of K terms of the form $\langle \phi, \cdot \rangle_{L^2}\, \phi$ with ϕ in $H^1(\mathbb{R}^3)$. Hence (5.89) and (5.90) since $\mathbf{P}_\Phi$ is quadratic with respect to Φ. Finally, recall from (5.66), that, for all $1 \leq i, j \leq K$,

$$\mathbb{W}[C, \Phi]_{i,j} = 2 \sum_{i,j,k,l=1}^{K} \gamma_{jkil}\, \mathbb{V}[\Phi]_{k,l},$$

with the coefficients γ_{jkil} depending quadratically on C according to (5.15)They are therefore locally Lipschitz continuous with respect to C, whereas the mapping $\Phi \mapsto \mathbb{V}[\Phi]$ is locally Lipschitz continuous from $H^1(\mathbb{R}^3)^K$ to $\mathscr{M}_{K\times K}\left(W^{1,\infty}(\mathbb{R}^3)\right)$ thanks to Lemma 5.10. (5.91) and (5.92) then follow again since $\mathbb{W}[C, \Phi]\Phi$ is obtained as a finite sum of compositions of locally bounded and locally Lipschitz continuous functions. □

As a direct consequence of the above lemma, we immediately deduce the following corollary, by observing from (5.78) that the mapping $(C, \Phi) \mapsto \mathbb{L}[C, \Phi]$ is obtained as a composition of locally Lipschitz continuous functions.

Corollary 5.6. *The mapping $(C, \Phi) \mapsto \mathbb{L}[C, \Phi]$ is locally Lipschitz continuous in a neighborhood of (C^0, Φ^0) in X with $\mathbf{\Gamma}(C^0)$ invertible.*

5.4.4 Step 3: Local existence and uniqueness of solution to (5.77)

In the previous section, we have proved that the mapping $(C,\Phi) \mapsto \big(\mathbb{K}[\Phi]C\,;\,\mathbb{L}[C,\Phi]\big)$ is locally bounded and locally Lipschitz continuous in a neighborhood of the initial data (C^0,Φ^0) in X. Existence and uniqueness of a solution $(C(t),\Phi(t))$ to the integral equation (5.79) in a neighborhood of (C^0,Φ^0) in X_T for $0<T$ small enough follows by Segal's Theorem [32], which also ensures the continuity with respect to the initial data. The equivalence between the integral formulation (5.79) and the initial value problem (5.77) for $(C^0,\Phi^0) \in \mathscr{F}_{N,K}$ with $\Phi^0 \in H^1\big(\mathbb{R}^3\big)^K$ is deduced by a standard argument and we refer the reader to [31, 12]. Next, thanks to Lemma 5.4 and Corollary 5.4, the constraint $\big(C(t),\Phi(t)\big) \in \mathscr{F}_{N,K}$ is satisfied for all $t \in [0,T]$. (Indeed, the proof of Lemma 5.4 and Corollary 5.4 heavily relies on the self-adjointness of $\mathbf{H}$ and keeps unchanged when $\mathbf{\Gamma}_\Phi$ is replaced by $\mathbf{\Gamma}(C)$.) Then, for all $t \in [0,T)$

$$\sum_{\sigma\in\Sigma_{N,K}} |c_\sigma(t)|^2 = \sum_{\sigma\in\Sigma_{N,K}} |c^0_\sigma|^2 = 1,$$

and, for all $1 \le i,j \le K$,

$$\langle \phi_i(t), \phi_j(t)\rangle = \langle \phi_i(0), \phi_j(0)\rangle = \delta_{i,j}.$$

In particular,

$$\forall t \in [0,T) \quad \|C(t)\| = 1 \quad \text{and} \quad \|\Phi(t)\|_{L^2} = K. \tag{5.93}$$

Thereby we may identify again the matrix $\mathbf{\Gamma}(C)$ with the matrix representation of $\mathbf{\Gamma}_\Phi$ in $\mathrm{Span}\{\Phi\}$. Then $\big(C(t),\Phi(t)\big)$ is also a solution to (5.69) and by the system itself we have the further regularity

$$C(t) \in C^1\big([0,T],\mathbb{C}^r\big) \quad \text{and} \quad \Phi(t) \in \times C^0([0,T],H^1(\mathbb{R}^3)^K) \cap C^1([0,\tau],H^{-1}(\mathbb{R}^3)^K).$$

5.4.5 Step 4: Maximal solution

To simplify notation, from now on we use the shorthand $\mathbf{\Gamma}(t)$ for $\mathbf{\Gamma}\big(C(t)\big)$. Existence of a global-in-time solution requires both to control of the H^1 norm of Φ and of the norm of $\mathbf{\Gamma}^{-1}(t)$. With the conservation of the energy this is equivalent to control only the norm of $\mathbf{\Gamma}^{-1}(t)$. Let T^* denotes the maximal existence time and assume that $T^* < +\infty$. We first show that

$$\limsup_{t\uparrow T^*} \|\mathbf{\Gamma}(t)^{-1}\| = +\infty. \tag{5.94}$$

We argue by contradiction and assume that there exists a positive constant M_0 such that for all $t \in [0,T^*)$, $\|\mathbf{\Gamma}(t)^{-1}\| \le M_0$. Thanks to (5.93), we know that $(C(t),\Phi(t))$ stays in $\mathscr{F}_{N,K}$ for all t in $[0,T^*)$. We now prove that there exists a positive constant K_0 such that

$$\forall t \in [0,T^*), \quad \|\Phi(t)\|_{H^1} \le K_0. \tag{5.95}$$

Thanks to Lemma 5.1 and Corollary 5.4, the energy is preserved by the flow, and therefore using the expression (5.76)

$$\begin{aligned}\Big\langle \mathbf{H}\,\mathbf{\Gamma}(t)\,\Phi(t),\Phi(t)\Big\rangle &\le \Big\langle \mathbf{H}\,\mathbf{\Gamma}(t)\,\Phi(t),\Phi(t)\Big\rangle + \frac{1}{2}\Big\langle \mathbb{W}[C,\Phi]\,\Phi(t),\Phi(t)\Big\rangle,\\ &= \mathscr{E}(\pi(C,\Phi)) = \mathscr{E}(C^0,\Phi^0),\end{aligned}$$

for all $0 \leq t < T^*$ since

$$\Big\langle \mathbb{W}[C,\Phi]\,\Phi(t),\Phi(t)\Big\rangle \geq 0,$$

for $V \geq 0$. As in [25, 18], Kato's inequality then yields that

$$\|\sqrt{\mathbf{\Gamma}}\Phi\|_{H^1} \leq M_1,$$

where M_1 is a positive constant that is independent of $t \geq 0$. Now let $\mu(t) \in (0,1]$ be the smallest eigenvalue of the hermitian matrix $\mathbf{\Gamma}(t)$, then

$$\frac{1}{\mu(t)} \leq \|\mathbf{\Gamma}^{-1}\| \leq \frac{K}{\mu(t)} \quad \text{and} \quad \frac{1}{\sqrt{\mu(t)}} \leq \|\sqrt{\mathbf{\Gamma}}^{-1}\| \leq \frac{K}{\sqrt{\mu(t)}},$$

for all $t \in [0,T^*)$, and therefore

$$\|\Phi\|_{H^1} \leq \frac{K}{\sqrt{\mu(t)}}\,\|\sqrt{\mathbf{\Gamma}}\Phi\|_{H^1} \leq K M_1\,\|\mathbf{\Gamma}^{-1}\|^{1/2}. \tag{5.96}$$

In particular, this shows (5.95) with $K_0 = m M_1 M_0^{1/2}$. Therefore, for any $t \in [0,T^*)$ arguing as in Step 3 above, we may build a solution to the system on $[t,t+T_0]$ for $T_0 > 0$ that only depends on M_0 and K_0. Since t is arbitrary close to T^* we reach the contradiction with the definition of T^*. Hence (5.94) holds.

Now, derivating the expression $\mathbf{\Gamma}\mathbf{\Gamma}^{-1} = \mathbb{I}_K$ with respect to t, we get

$$\frac{d\mathbf{\Gamma}^{-1}}{dt} = -\mathbf{\Gamma}^{-1}\,\frac{d\mathbf{\Gamma}}{dt}\,\mathbf{\Gamma}^{-1}, \tag{5.97}$$

for all $t \in [0,T^*)$. From the expression of $\mathbf{\Gamma}$ in terms of C and since $\|C\| = 1$, it holds

$$\Big\|\frac{d\mathbf{\Gamma}}{dt}\Big\| \lesssim \Big\|\frac{dC}{dt}\Big\| \lesssim \big\|\Phi\big\|_{H^1} \lesssim \|\mathbf{\Gamma}^{-1}\|^{1/2}$$

in virtue of (5.77) and of the bounds (5.84) in Lemma 5.12 using the fact that $\|\Phi\|_{L^2} = K$. Inserting the last bound above in (5.97) and integrating over t yields

$$\|\mathbf{\Gamma}(t)^{-1}\| \leq \|\mathbf{\Gamma}(0)^{-1}\| + \text{const.}\int_0^t \|\mathbf{\Gamma}(s)^{-1}\|^{3/2}\,ds,$$

for all $t \in [0,T^*)$. Because of (5.94), this implies that $\int_0^{T^*} \|\mathbf{\Gamma}(s)^{-1}\|^{3/2}\,ds = +\infty$. This concludes the proof of Theorem 5.3.

5.4.6 Further regularity of the solutions

In the previous section we have established the well-posedness of the MCTDHF system in the energy space. It is of interest, for numerical simulations and mathematical analysis, to verify that the MCTDHF dynamics propagates further regularity of the initial data as the Schrödinger's does. Actually, we will claim

Corollary 5.7. *Let $(C^0,\Phi^0)\in\partial\mathscr{F}_{N,K}$ and let Φ^0 be in $H^m(\mathbb{R}^3)^K$, with integer $m\geq 1$ (respectively $H^m_0(\Omega)^K$ when $\Omega\neq\mathbb{R}^3$). Then, the solution of System* (5.64) *in the energy space whose existence is ensured by Theorem 5.3 is such that*

$$\Phi\in C^0([0,T^\star),H^m(\mathbb{R}^3)^K)\cap C^1([0,T^\star),H^{m-2}(\mathbb{R}^3)^K).$$

Moreover, this solution depends continuously on the initial data and satisfies the assertions ii)$-$iv) of Theorem 5.3.

Proof. The proof is based on standard arguments [11]. One start with initial data $(C^0,\Phi^0)\in\partial\mathscr{F}_{N,K}$ with $\Phi^0\in H^m(\mathbb{R}^3)^K$ and prove that the non-linearities $(C,\Phi)\mapsto\mathbb{K}[\Phi]\,C$ and $(C,\Phi)\mapsto\mathbb{L}[C,\Phi]$ are still locally bounded and locally lipschitz in $\mathbb{C}^r\times H^m(\mathbb{R}^3)^K$ in a neighborhood of (C^0,Φ^0). This follows immediately after the observation that for all $\Phi\in H^m(\mathbb{R}^3)^K$, we have

$$\|\mathbb{K}[\Phi]\|\lesssim\|\Phi\|^4_{H^1}\lesssim\|\Phi\|^4_{H^m},$$

$$\|\mathbf{P}_\Phi\,\mathbb{W}[C,\Phi]\,\Phi\|_{H^m(\mathbb{R}^3)^K}\lesssim\|C\|^2\,\|\Phi\|^2_{H^1}\,\|\Phi\|^3_{H^m},$$

$$\|\mathbb{W}[C,\Phi]\,\Phi\|_{H^m}\lesssim\|C\|^2\,\|\Phi\|^2_{H^{m-1}}\,\|\Phi\|_{H^m}.$$

The first and second assertions are obvious and we refer to [11] for the last one. The local in time existence in $\mathbb{C}^r\times H^m(\mathbb{R}^3)^K$ is then obtained. Next, using the Duhamel formula for the PDEs system, we get for all $t\in[0,A]\subset[0,T^\star)$

$$\|\Phi(t)\|_{H^m}\leq\|\Phi^0\|_{H^m}+\text{const.}\sup_{[0,A]}\big(\|\mathbf{\Gamma}^{-1}(s)\|\,\|C\|^2\,\|\Phi(s)\|^2_{H^{m-1}}\big)\int_0^t\|\Phi(s)\|_{H^m}\,ds.$$

Gronwall's lemma allows to conclude. □

5.5 Sufficient condition for global-in-time existence

For any $K\geq N$, we denote

$$I(K)=\inf\big\{\mathscr{E}(\pi(C,\Phi))\,:\,(C,\Phi)\in\mathscr{F}_{N,K}\big\}.$$

Observe the following trivial relation,

$$\forall K'\leq K\leq\infty,\quad\inf\sigma(H_N)\leq I(K)\leq I(K'),\tag{5.98}$$

with $\inf\sigma(H_N)$ being the bottom of the spectrum of H_N on $L^2_\wedge(\Omega^N)$. Recall that the maximal rank hypothesis corresponds to the following equivalent facts :

(i) The rank of the operator $[\pi(C,\Phi)\otimes\pi(C,\Phi)]_{:1}$ is equal to K;

(ii) The $K\times K$ matrix $\mathbf{\Gamma}(C)$ is invertible;

(iii) The smallest eigenvalue of $\mathbf{\Gamma}(C)$ is strictly positive.

Theorem 5.4. *Let $(C^0,\Phi^0)\in\mathscr{F}_{N,K}$ be an initial data in* (5.69) *with $\mathbf{\Gamma}\big(C^0\big)$ invertible. Assume that $T^\star<+\infty$ then*

$$\mathscr{E}(\pi(C^0,\Phi^0))\geq I(K-1).$$

As an immediate by-product we get a sufficient condition assuring the global-in-time invertibility of the matrix $\mathbf{\Gamma}\big(C(t)\big)$, or equivalently, the non-existence of a finite time T^* such that

$$\lim_{t\to T*}\gamma_i(t)=0\quad\text{ for some }\quad 1\leq i\leq K. \tag{5.99}$$

Corollary 5.8. *If $(C^0,\Phi^0)\in\partial\mathscr{F}_{N,K}$ satisfies*

$$I(K)\leq\mathscr{E}(\pi(C^0,\Phi^0))<I(K-1), \tag{5.100}$$

then $T^\star=+\infty$; that is the solution is global-in-time.

Remark 5.7. Notice that since $I(K)<I(K-2)$ holds true in physical situations [24, 19], existence of initial data fulfilling (5.100) is ensured for infinitely many values of K's up to infinity. Indeed, either $I_K<I_{K-1}$ or $I_K=I_{K-1}$ and then $I_{K-1}<I_{K-2}$. Hence, the global-in-time well-posedness of the associated Cauchy problems for such initial data.

Proof of Theorem 5.4. Let (C,Φ) be the solution of (5.69) with initial data (C^0,Φ^0) whose existence is ensured by Theorem 5.3. We assume that $T^\star<+\infty$ and

$$\limsup_{t\uparrow T^\star}\|\mathbf{\Gamma}\big(C(t)\big)^{-1}\|=+\infty.$$

Equivalently, the eigenvalues of $\mathbf{\Gamma}(C)$ being arranged in decreasing order $0\leq\gamma_K\leq\gamma_{K-1}\leq\ldots\leq\gamma_1\leq 1$, this means

$$\liminf_{t\uparrow T^\star}\gamma_K(t)=0.$$

Then there exists a sequence t_n converging to $T^\star$, a positive number β and an integer $N+1\leq m\leq K$ such that

$$\lim_{n\to+\infty}\gamma_m(t_n)=0\quad\text{ and }\quad 0<\beta\leq\gamma_{m-1}(t_n)\,.$$

Indeed, since $\sum_{k=1}^K\gamma(t)=N$ for all t, then at least N eigenvalues do not go to zero as t goes to $T^\star$. We denote $C^n=C(t_n)$, $\Phi^n=\Phi(t_n)$, $\gamma_i^n=\gamma_i(t_n)$, $\mathbf{\Gamma}^n=\mathbf{\Gamma}\big(C(t^n)\big)$ and so on for other involved quantities. For all $n\geq 1$, $(C^n,\Phi^n)\in\partial\mathscr{F}_{N,K}$. Thus according to Proposition 5.5, there exists a unique sequence of unitary transforms $\mathscr{U}^n\in\mathscr{O}_K^r$ that map $(C^n,\,\Phi^n)$ into (C'^n,Φ'^n) with Φ'^n being an eigenbasis for the operator γ^n. In particular the corresponding matrix $\mathbf{\Gamma}'^n:=\mathbf{\Gamma}(C'^n)$ is diagonal. In other words,

$$\begin{aligned}\Psi^n:=\pi(C^n,\Phi^n)&=\sum_\sigma c_\sigma^n\,\Phi_\sigma^n=\sum_\sigma {c'}_\sigma^{\,n}\,{\Phi'}_\sigma^{\,n}=\pi(C'^n,\Phi'^n),\\ \gamma^n&=\sum_{i,j=1}^K\gamma_{ij}^n\,\phi_i^n\otimes\overline{\phi_j}^n=\sum_{i=1}^K\gamma_i^n\,\phi_i'^n\otimes\overline{\phi_i'}^n.\end{aligned}$$

Since the group of unitary transforms is compact, we may argue equivalently on the sequence (C'^n,Φ'^n) that we keep denoting by (C^n,Φ^n) for simplicity. Now, we claim the following

Lemma 5.14. *Let $N+1 \le m \le K$ and $\beta > 0$ and let (C^n, Φ^n) be a sequence in $\partial \mathscr{F}_{N,K}$ with $\mathbf{\Gamma}^n = \mathbf{\Gamma}(C^n)$ diagonal whose eigenvalues satisfy*

$$\begin{cases} 0 \le \gamma_K^n \le \ldots \le \gamma_{m+1}^n \le \beta \le \gamma_m^n \le \ldots \le \gamma_1^n \le 1, \\ \lim\limits_{n\to+\infty} \gamma_m^n = 0 \text{ and for all } \quad 1 \le i \le m-1, \quad \lim\limits_{n\to+\infty} \gamma_i^n = \gamma_i^\star \ge \beta. \end{cases} \tag{5.101}$$

Then,

$$\text{for all } m \le i \le K, \quad \lim_{n\to+\infty} \gamma_i^n = 0. \tag{5.102}$$

and

$$\text{for all } \sigma \in \Sigma_N^K, \qquad \{m,\ldots,K\} \cap \sigma \neq \emptyset \quad \Longrightarrow \quad \lim_{n\to+\infty} c_\sigma^n = 0. \tag{5.103}$$

In particular,

$$\lim_{n\to+\infty} \sum_{\sigma\subset\{1,\ldots,m-1\}} |c_\sigma^n|^2 = 1, \tag{5.104}$$

$$\text{for all } 1 \le i \le m-1, \quad \lim_{n\to+\infty} \sum_{\sigma\subset\{1,\ldots,m-1\} \,|\, i\in\sigma} |c_\sigma^n|^2 = \gamma_i^*, \tag{5.105}$$

$$\sum_{i=1}^{m-1} \gamma_i^\star = N, \tag{5.106}$$

$$\text{if } \{i,j,k,l\} \cap \{m,\ldots,K\} \neq \emptyset, \quad \lim_{n\to+\infty} \gamma_{ikjl}^n = 0, \tag{5.107}$$

and

$$\lim_{n\to+\infty} \Big\| \Psi^n - \sum_{\sigma\subset\{1,\ldots,m-1\}} c_\sigma^n \, \Phi_\sigma^n \Big\|_{L^2(\mathbb{R}^{3N})} = 0. \tag{5.108}$$

Proof. Claim (5.102) follows immediately from (5.101), whence (5.103) in virtue of (5.18). (5.104), (5.105) and (5.106) follow immediately from (5.101) and (5.18) since the sequence $C^n \in S^{r-1}$ is compact in $\mathbb{C}^r$. From the expression (5.15) for $\gamma_{ikl\,j}^n$, we observe that

$$\begin{aligned} \left|\gamma_{ijkl}^n\right| &\lesssim \min\left(\sqrt{\gamma_i^n}\,;\sqrt{\gamma_k^n}\right) \min\left(\sqrt{\gamma_j^n}\,;\sqrt{\gamma_l^n}\right) \\ &\lesssim \min\left(\gamma_i^n\,;\gamma_k^n\,;\gamma_j^n\,;\gamma_l^n\right)^{1/2}, \end{aligned} \tag{5.109}$$

since $0 \le \gamma_\cdot^n \le 1$. We thus get (5.107) from (5.102).

On the other hand, we have

$$\Psi^n = \pi(C^n, \Phi^n) = \sum_{\sigma\cap\{m,\ldots,K\}\neq\emptyset} c_\sigma^n \, \Phi_\sigma^n + \sum_{\sigma\cap\{m,\ldots,K\}=\emptyset} c_\sigma^n \, \Phi_\sigma^n. \tag{5.110}$$

As a consequence of (5.103)

$$\lim_{n\to+\infty} \Big\| \sum_{\sigma\cap\{m,\ldots,K\}\neq\emptyset} c_\sigma^n \, \Phi_\sigma^n \Big\|_{L^2(\mathbb{R}^{3N})} = 0,$$

since each determinant ${\Phi_\sigma}^n$ is normalized in $L^2(\mathbb{R}^{3N})$. Hence (5.108). □

Since the MCTDHF flow keeps the energy constant, we have

$$\mathscr{E}\big(\pi(C^n,\Phi^n)\big) = cste = \mathscr{E}\big(\pi(C,\Phi)\big),$$

for all $n \geq 1$. This property provides with additional information on the sequence $(C^n;\Phi^n)$ that is collected in the following lemma.

Lemma 5.15. *Let $(C^n;\Phi^n)$ be like in Lemma 5.14. We assume in addition that*

$$\mathscr{E}\big(\pi(C^n;\Phi^n)\big) \leq cste.$$

Then, extracting subsequences if necessary, we have

$$\text{for all } m \leq i \leq K, \quad \sqrt{\gamma_i^n}\,\phi_i^n \text{ converges to } 0 \text{ weakly in } H^1(\mathbb{R}^3) \text{ and strongly in } L^2(\mathbb{R}^3), \tag{5.111}$$

$$\text{for all } 1 \leq i \leq m-1,\ \phi_i^n \text{ is bounded in } H^1(\mathbb{R}^3), \tag{5.112}$$

and

$$\liminf_{n\to+\infty} \mathscr{E}\big(\pi(C^n,\Phi^n)\big) \quad \geq \quad I(m-1). \tag{5.113}$$

Proof. We first recall that, for any $(C;\Phi)$ in $\mathscr{F}_{N,K}$ and for the particular Hamiltonian we are dealing with, the energy functional writes

$$\begin{aligned}
\mathscr{E}\big(\pi(C,\Phi)\big) &= \int_{\mathbb{R}^3} \Big(\mathbf{\Gamma}(C)\,\Phi; (-\frac{1}{2}\Delta + U)\,\Phi\Big)\,dx + \iint_{\mathbb{R}^3\times\mathbb{R}^3} \frac{[\Psi\otimes\Psi]_{:2}(x,y,x,y)}{|x-y|}\,dxdy, \\
&= \sum_{i,j=1}^K \gamma_{ij} \int_{\mathbb{R}^3} \left[\frac{1}{2}\nabla\phi_i\,\nabla\bar{\phi}_j + U\,\phi_i\,\bar{\phi}_j\right] dx + \sum_{i,j,k,l=1}^K \gamma_{ijkl}\, D\big(\phi_l\,\bar{\phi}_i;\,\phi_k\,\bar{\phi}_j\big),
\end{aligned}$$

with $(\cdot;\cdot)$ being the scalar product in $\mathbb{C}^K$. Using the fact that the ϕ_i^n's diagonalize γ^n, we have

$$\begin{aligned}
\mathscr{E}(\pi(C^n,\Phi^n)) &= \sum_{i=1}^K \gamma_i^n \int_{\mathbb{R}^3} \left[\frac{1}{2}|\nabla\phi_i^n|^2 + U\,|\phi_i^n|^2\right] dx + \sum_{i,j,k,l=1}^K \gamma_{ijkl}^n\, D\big(\phi_l^n\,\bar{\phi}_i^n;\,\phi_k^n\,\bar{\phi}_j^n\big), \\
&\geq \sum_{i=1}^K \gamma_i^n \int_{\mathbb{R}^3} \left[\frac{1}{2}|\nabla\phi_i^n|^2 + U\,|\phi_i^n|^2\right] dx. \qquad (5.114)
\end{aligned}$$

where in (5.114) we used the positivity of the two-body interaction potential V. By the Kato inequality, for any $0 < \varepsilon < 1$, there exists $C_\varepsilon > 0$ such that

$$|U| \leq -\varepsilon\,\Delta + C_\varepsilon$$

in the sense of self-adjoint operators. Then,

$$\sum_{i=1}^K \gamma_i^n \int_{\mathbb{R}^3} U\,|\phi_i^n|^2\,dx \geq -\varepsilon\left(\sum_{i=1}^K \gamma_i^n \int_{\mathbb{R}^3} |\nabla\phi_i^n|^2\,dx\right) - C_\varepsilon N.$$

Therefore

$$\sum_{i=1}^K \gamma_i^n \int_{\mathbb{R}^3} |\nabla\phi_i^n|^2\,dx \leq cste.$$

Thus, for all $1 \le i \le K$, $\sqrt{\gamma_i^n}\,\phi_i^n$ is bounded in $H^1(\mathbb{R}^3)$. (5.111) and (5.112) are then deduced from (5.108) and (5.101) respectively. Since, under the hypotheses on U, the map $\varphi \mapsto \int_{\mathbb{R}^3} U\,|\varphi|^2\,dx$ is weakly lower semi-continuous on $H^1(\mathbb{R}^3)$, we deduce from (5.111) that

$$\liminf_{n\to+\infty} \sum_{i=1}^{K} \gamma_i^n \int_{\mathbb{R}^3} \left[\frac{1}{2}|\nabla\phi_i^n|^2 + U\,|\phi_i^n|^2\right] dx \ge \liminf_{n\to+\infty} \sum_{i=1}^{m-1} \gamma_i^n \int_{\mathbb{R}^3} \left[|\nabla\phi_i^n|^2 + U\,|\phi_i^n|^2\right] dx. \tag{5.115}$$

We now check that

$$\liminf_{n\to+\infty} \sum_{i,j,k,l=1}^{K} \gamma_{ijkl}^n D\big(\phi_l^n\,\bar\phi_i^n\,;\,\phi_k^n\,\bar\phi_j^n\big) = \liminf_{n\to+\infty} \sum_{i,j,k,l=1}^{m-1} \gamma_{ijkl}^n D\big(\phi_i^n\,\bar\phi_l^n\,;\,\phi_k^n\,\bar\phi_j^n\big), \tag{5.116}$$

by showing that

$$\liminf_{n\to+\infty} \sum_{\substack{i,j,k,l=1\\ \{i,j,k,l\}\cap\{m,\dots,K\}\neq\emptyset}}^{K} \gamma_{ijkl}^n D\big(\phi_l^n\,\bar\phi_i^n\,;\,\phi_k^n\,\bar\phi_j^n\big) = 0. \tag{5.117}$$

Let $\{i,j,k,l\}\cap\{m,\dots,K\} \neq \emptyset$. We assume without loss of completeness that $i \ge m$. Then thanks to (5.86) and (5.109)

$$\begin{aligned} |\gamma_{ijkl}^n D\big(\phi_l^n\,\bar\phi_i^n\,;\,\phi_k^n\,\bar\phi_j^n\big)| &\lesssim \sqrt{\gamma_i^n}\sqrt{\gamma_j^n}\,\|\phi_i^n\|_{L^2}\,\|\nabla\phi_j^n\|_{L^2}\,\|\phi_k^n\|_{L^2}\,\|\phi_l^n\|_{L^2},\\ &\lesssim \|\sqrt{\gamma_i^n}\,\phi_i^n\|_{L^2}, \end{aligned}$$

since $\|\phi_k^n\|_{L^2} = \|\phi_l^n\|_{L^2} = 1$ and since in any case $\sqrt{\gamma_j^n}\,\nabla\phi_j^n$ is bounded in L^2 independently of n. Therefore each term which appears in the sum in (5.117) converges to 0 as n goes to infinity thanks to (5.111). Claim (5.117) then follows.

Gathering together (5.115) and (5.116) we have

$$\begin{aligned} &\liminf_{n\to+\infty} \mathscr{E}\big(\pi(C^n;\Phi^n)\big)\\ &\quad\ge \liminf_{n\to+\infty} \Big[\sum_{i=1}^{m-1} \gamma_i^n \int_{\mathbb{R}^3} \left[|\nabla\phi_i^n|^2 + U\,|\phi_i^n|^2\right] dx + \sum_{i,j,k,l=1}^{m-1} \gamma_{ijkl}^n D\big(\phi_i^n\,\bar\phi_l^n\,;\,\phi_k^n\,\bar\phi_j^n\big)\Big]. \end{aligned} \tag{5.118}$$

The point now consists in showing that the right-hand side in (5.118) is bounded from below by $\liminf_{n\to+\infty} \mathscr{E}\big(\pi(\tilde C^n,\tilde\Phi^n)\big)$ where $\tilde C^n = (c_\sigma^n)_{\sigma\subset\{1,\dots,m-1\}} \in \mathbb{C}^{m-1}$ and $\tilde\Phi^n = (\phi_1^n,\dots,\phi_{m-1}^n) \in \mathscr{O}_{L^2(\Omega)^{m-1}}$. There is a slight difficulty arising here from the fact that (with obvious notation) $\tilde\gamma_{ij}^n$ is close but different from $\gamma_i^n\,\delta_{ij}^n$ and similarly for $\tilde\gamma_{ijkl}^n$ and γ_{ijkl}^n. (Also $\tilde C^n$ is not normalized in $\mathbb{C}^{m-1}$ (only asymptotically) but this will be dealt with afterwards.)

First we observe that because of (5.17) for every $i,j \in \{1,\cdots,m-1\}$,

$$\gamma_i^n\,\delta_{ij}^n - \tilde\gamma_{ij}^n = \sum_{\substack{(\sigma\cup\tau)\cap\{m,\cdots,K\}\neq\emptyset\\ i\in\sigma,\,j\in\tau,\,\sigma\setminus\{i\}=\tau\setminus\{j\}}} (-1)^{\sigma^{-1}(i)+\tau^{-1}(j)}\,c_\sigma^n\,\bar c_\tau^n,$$

goes to 0 as n goes to infinity thanks to (5.103). In addition, each term of the form $\int_{\mathbb{R}^3}\left[\frac{1}{2}\nabla\phi_i^n\cdot\nabla\bar\phi_j^n+U\,\phi_i^n\cdot\bar\phi_j^n\right]dx$ is bounded independently of n for $i,j\in\{1,\cdots,m-1\}$. Therefore

$$\sum_{i=1}^{m-1}\gamma_i^n\int_{\mathbb{R}^3}\left[\frac{1}{2}|\nabla\phi_i^n|^2+U\,|\phi_i^n|^2\right]dx=\sum_{i,j=1}^{m-1}\tilde\gamma_{ij}^n\int_{\mathbb{R}^3}\left[\frac{1}{2}\nabla\tilde\phi_i^n\,\nabla\bar{\tilde\phi}_j^n+U\,\tilde\phi_i^n\,\bar{\tilde\phi}_j^n\right]dx+o(1). \tag{5.119}$$

For the same reason, and with obvious notation, for all $1\le i,j,k,l\le m-1$,

$$\lim_{n\to+\infty}\left|\gamma_{ijkl}^n-\tilde\gamma_{ijkl}^n\right|=0,$$

since according to (5.15) the extra terms in these differences only involve coefficients c_σ^n with $\sigma\cap\{m,\dots,K\}\neq\emptyset$. Again each term of the form $D\big(\phi_i^n\,\bar\phi_l^n;\phi_k^n\,\bar\phi_j^n\big)$ is bounded independently of n for $i,j,k,l\in\{1,\dots,m-1\}$. Therefore

$$\sum_{i,j,k,l=1}^{m-1}\gamma_{ijkl}^n D\big(\phi_i^n\,\bar\phi_l^n;\phi_k^n\,\bar\phi_j^n\big)=\sum_{i,j,k,l=1}^{m-1}\tilde\gamma_{ijkl}^n D\big(\phi_i^n\,\bar\phi_l^n;\phi_k^n\,\bar\phi_j^n\big)+o(1). \tag{5.120}$$

Therefore, gathering together (5.118), (5.119) and (5.120),

$$\liminf_{n\to+\infty}\mathscr{E}\big(\pi(C^n;\Phi^n)\big)\ge\liminf_{n\to+\infty}\mathscr{E}\big(\pi(\tilde C^n;\tilde\Phi^n)\big). \tag{5.121}$$

Since $\tilde C^n$ is not in S^{m-1} (it is only the case asymptotically), $(\tilde C^n;\tilde\Phi^n)$ is not in $\mathscr{F}_{N,m-1}$, thus we cannot bound immediately $\mathscr{E}\big(\pi(\tilde C^n;\tilde\Phi^n)\big)$ from below by $I(m-1)$. We proceed as follows. We denote $\tilde\Psi^n=\pi(\tilde C^n,\tilde\Phi^n)$. Then,

$$\lim_{n\to+\infty}\|\tilde\Psi^n\|^2=\lim_{n\to+\infty}\sum_{\sigma\subset\{1,\dots,m-1\}}|c_\sigma^n|^2=1, \tag{5.122}$$

in virtue of (5.104). Finally the energy being quadratic with respect to Ψ

$$\mathscr{E}\big(\pi(\tilde C^n;\tilde\Phi^n)\big)=\|\tilde\Psi^n\|^2\,\mathscr{E}\Big(\frac{\tilde\Psi^n}{\|\tilde\Psi^n\|}\Big)\ge\|\tilde\Psi^n\|^2 I(m-1), \tag{5.123}$$

for $\tilde\Psi^n/\|\tilde\Psi^n\|\in\mathscr{F}_{N,m-1}$ for all $n\ge1$. Gathering together (5.121), (5.122) and (5.123) and taking the limit as n goes to infinity we deduce (5.113). □

Remark 5.8. When Ω is a bounded domain of $\mathbb{R}^3$, the proof is much easier and it is detailed in [3]. On the one hand, any sequence in $\mathscr{F}_{N,K}$ is relatively compact in $\mathbb{C}^r\times L^2(\Omega)^K$ thanks to the Rellich theorem. On the other hand, the energy functional $\Psi\mapsto\mathscr{E}(\Psi)$ is weakly lower semi-continuous in $H^1(\Omega^{3N})$ while it is not in $H^1(\mathbb{R}^{3N})$ as already observed by Friesecke [18]. Therefore it is easily checked in that case that

$$\liminf_{n\to+\infty}\mathscr{E}\big(\pi(C^n;\Phi^n)\big)\ge\liminf_{n\to+\infty}\mathscr{E}\big(\pi(C^\star;\Phi^\star)\big)\ge I(m-1),$$

with $(C^\star;\Phi^\star)\in\mathscr{F}_{N,m-1}$ being the weak limit of the sequence $(\tilde C^n;\tilde\Phi^n)$ introduced in the above proof.

Remark 5.9 (Stability, Consistency and invertibility of the density matrix $\mathbf{\Gamma}$)**.** The main factor in the instability of the working equations or any gauge-equivalent system, is the inverse of the density matrix. In the present section, criteria for the global invertibility of $G(C)$ have been given. These criteria do not provide with an uniform estimate for $\|\mathbf{\Gamma}^{-1}\|$, and furthermore increasing the consistency of the MCTDHF approximation leads to the increase of the number K of orbitals. As usual consistency and stability are both necessary and antinomic. Indeed, the most obvious observation is that one always has

$$\|\mathbf{\Gamma}^{-1}\| \geq \frac{K}{N},$$

for $\mathbf{\Gamma}$ has at most K positive eigenvalues whose sum equals N. Therefore the smallest can be at most N/K. These considerations lead either to a limitation on K or to a regularization or a "cut-off " of $\mathbf{\Gamma}^{-1}$.

5.6 L^2 solutions of MCTDHF

We briefly recall an L^2 analysis of the MCTDHF equations without an H^1 estimate, i.e. without restriction to finite energy. This is in line e.g. with the work of Castella on the L^2 analysis of the Hartree case ("Schrödinger-Poisson") which has inspired our Strichartz techniques.

Besides the theoretical interest, there is a very practical interest in a theory without energy conservation : in numerical simulations problems with (nearly) singular density matrix are dealt with by ad-hoc methods like perturbing the density matrix to keep it invertible. E.g. by adding a small identity matrix $\mathbf{\Gamma}_\varepsilon = \mathbf{\Gamma} + \varepsilon Id$ (see e.g. [9] or by taking

$$\mathbf{\Gamma}_\varepsilon = \mathbf{\Gamma} + \varepsilon \exp(-\mathbf{\Gamma}/\varepsilon), \tag{5.124}$$

for small values of ε (see [7]). Note that with this method vanishing eigenvalues are perturbed at order ε while the others are unchanged up to exponentially small errors in terms of ε.

In this perturbed version the energy is no longer preserved by the flow and analysis has to rely on a strategy with mass conservation only. This is achieved by using of Strichartz estimates which only require L^2 control on the data.

We fix $\varepsilon > 0$ and use the system (5.77) with $\mathbf{\Gamma}$ replaced by $\mathbf{\Gamma}_\varepsilon$

$$\begin{cases} i\frac{dC}{dt} &= \mathbb{K}[\Phi]\,C, \\ i\frac{\partial \Phi}{\partial t} &= \mathbf{H}\Phi + \mathbf{\Gamma}_\varepsilon[C]^{-1}\,(\mathbf{I}-\mathbf{P}_\Phi)\,\mathbb{W}[C,\Phi]\Phi, \\ C(0) = C^0, \qquad & \Phi(0) = \Phi^0, \end{cases} \tag{5.125}$$

with $\mathbf{\Gamma}_\varepsilon := \mathbf{\Gamma}_\varepsilon(C)$ being defined by (5.124). In [30] we have established existence and uniqueness of global-in-time solutions of mild solutions to (5.125) that are given by the Duhamel formula

$$\begin{aligned} C(t) &= C^0 + \int_0^t \mathbb{K}[\Phi(s)]\,C(s)\,ds, \\ \Phi(t) &= \exp[-\frac{1}{2}it\Delta]\Phi^0 - i\int_0^t \exp[-\frac{1}{2}i(t-s)\Delta]\,U(s)\Phi(s)\,ds \\ &\quad -i\int_0^t \exp[-\frac{1}{2}i(t-s)\Delta]\,\mathbf{\Gamma}_\varepsilon[C(s)]^{-1}\,(\mathbf{I}-\mathbf{P}_\Phi)\,\mathbb{W}[C,\Phi]\Phi\,ds. \end{aligned}$$

with the help of the Picard fixed point theorem and Strichartz estimates. We introduce the following spaces (cf Zagatti [34] and Castella [11]):

$$X_T = L^\infty(0,T;\mathbb{C}^r) \times \big((L^\infty(0,T;L^2(\mathbb{R}^3)) \cap (L^p(0,T;L^q(\mathbb{R}^3))\big)^K,$$

where $2 \leq q = \frac{2d}{d-1} < 6$ and $\frac{2}{3p} = (\frac{1}{2} - \frac{1}{q})$ (Strichartz admissibility) for U,V being in L^d with $d > \frac{3}{2}$. Then with Holder inequalities interpolation and Strichartz estimates for all $T > 0, \phi \in L^2$ and $\varphi \in L^{a'}(0,T;L^{b'}(\mathbb{R}^3))$

$$\| \exp[-\frac{1}{2} it\Delta]\, \phi \|_{L^p(0,T;L^q(\mathbb{R}^3))} \leq \rho(p)\, \|\phi\|_{L^2},$$
$$\| \int_0^t \exp[-\frac{1}{2} i(t-s)\Delta]\, \varphi(s) ds \|_{L^p(0,T;L^q(\mathbb{R}^3))} \leq \rho(p,a)\, \|\varphi\|_{L^{a'}(0,T;L^{b'}(\mathbb{R}^3))}.$$

with ρ being a universal non negative constant and a', b' the conjugate of a, b respectively with $\frac{2}{3a} = (\frac{1}{2} - \frac{1}{b})$.

Then, we prove the existence of a radius $R > 0$ and a time $T' > 0$ such that the mapping

$$(C,\Phi) \mapsto \Big(\int_0^t \mathbb{K}[\Phi(s)]\, C(s) ds\, ;\, \int_0^t e^{-i(t-s)\frac{1}{2}\Delta} \big(U\Phi(s) + \mathbf{\Gamma}_\varepsilon(s)^{-1} (\mathbf{I} - \mathbf{P}_\Phi) \mathbb{W}[C,\Phi](s)\, \Phi(s) \big)\, ds \Big),$$

maps the ball of radius R of $X_{T'}$ into itself for all $t \in [0,T']$ and it is Lipschitz with a Lipschitz bound depending on

$$\mathbf{\Gamma}_\varepsilon, \|U\|_{L^d}, \|V\|_{L^d} \quad \text{and} \quad \|C,\Phi\|_{X_{T'}},$$

which leads in particular to the local existence and uniqueness of solutions in $X_{T'}$. Moreover, there exists $0 < T_0 \leq T'$ such that the solution fullfills the estimate

$$\|\Phi\|_{L^p(0,T_0;L^q(\mathbb{R}^3))} \lesssim \|\Phi\|_{L^\infty(0,T_0;L^2(\mathbb{R}^3)},$$

for all Strichartz admissible pairs (p,q).

The existence and uniqueness of a solution in X_∞ with no assumption on the H^1 norm follows, from the conservation of $\|C,\Phi\|_{\mathbb{C}^r \times L^2}$. In particular, one can iterate the argument from $0, T_0$ to $T_0, 2T_0$... (see the details in the previous chapter)

5.7 Extension to time-dependent potentials...

Assume now that the Hamiltonian $\mathscr{H}$ depends on time through the exterior potential U in (5.1) (by considering for example moving nuclei). For the sake of simplicity we only restrict ourselves to U and $\frac{\partial U}{\partial t}$ in $L^\infty(\Omega)$. System $\mathscr{S}_0$ with $\mathscr{H}$ replaced by $\mathscr{H}(t)$ keeps on preserving the constraints since Lemma 5.4 only relies on the self-adjointness of the Hamiltonian and its solutions satisfy the Dirac-Frenkel variational principle. However the conservation of the energy is no longer ensured by the flow. Indeed, following the lines of the proof of Corollary 5.1, we have

$$\frac{d}{dt} \mathscr{E}\big(\Psi(t)\big) = \frac{d}{dt} \big\langle \mathscr{H}(t)\, \Psi(t) \big| \Psi(t) \big\rangle = \big\langle \frac{\partial U}{\partial t}\, \Psi(t) \big| \Psi(t) \big\rangle.$$

Then the energy in controlled for any finite time, whence the existence and the H^1-regularity of solutions as long as the matrix $\mathbf{\Gamma}\big(C(t)\big)$ remains invertible. If we assume additionally that, for all time, the solution $\Psi = \pi(C,\Phi) \in \partial\mathscr{B}_{N,K}$ satisfies

$$\langle \frac{\partial U}{\partial t}\,\Psi(t)\big|\Psi(t)\rangle \leq h(t)\,\langle \mathscr{H}(t)\,\Psi(t)\big|\Psi(t)\rangle,$$

for a given function h, then by the Gronwall lemma

$$\langle \mathscr{H}(t)\,\Psi(t)\big|\Psi(t)\rangle - \langle \mathscr{H}\,\Psi_0\big|\Psi_0\rangle \leq \exp\Big(\int_0^t h(s)\,ds\Big).$$

Then concerning the conservation of the global full-rank of the one-particle density matrix, the result of Theorem 5.4 remains true provided

$$\mathscr{E}(\Psi_0) = \langle \mathscr{H}(0)\,\Psi_0\big|\Psi_0\rangle \leq I(K-1) - \exp\Big(\int_0^{+\infty} h(s)\,ds\Big).$$

5.8 Extension to other multi-configuration type models

Given (C,Φ) in $\mathscr{F}_{N,K}$, it is also convenient (see *e.g.* [25]) to use the alternate equivalent expression for the wave function Ψ given by

$$\Psi = \sum_f \alpha_f \Phi_f, \tag{5.126}$$

where f is any injective mapping from $\{1,\cdots,N\}$ onto $\{1,\cdots,K\}$, not necessarily increasing, (we again identify the function itself with its range), and where Φ_f is the Hartree product $\Phi_f(x_1,\cdots,x_N) = \phi_{f(1)}(x_1)\,\phi_{f(2)}(x_2)\cdots\phi_{f(N)}(x_N)$. The above sum runs over all such mappings f. The coefficients α are linked to the C's through the following

$$\alpha_f = \frac{(-1)^\varepsilon}{\sqrt{N!}}\,c_{\varepsilon\circ f}, \tag{5.127}$$

with ε being the permutation of $\mathfrak{S}_N$ such that $\varepsilon\circ f \in \Sigma_{N,K}$. In other words, to every c_σ for $\sigma\in\Sigma_{N,K}$ corresponds $N!$ coefficients α_f that differ only by a $\pm$ sign. The coefficients α_f are skew-symmetric with respect to permutations of their indexes. These coefficients exhibit better than the c'_σs the algebraic similarity between the MCHF method and the MCH one where no skew-symmetry assumption is prescribed for the wave-function. Here we explain how our method include and extend previous works for the MCTDH and TDH models as well.The normalization constraint on the expansion coefficients c_σ translates into $\sum_f |\alpha_f|^2 = 1$. Using this new expression, a straightforward computation shows that

$$\gamma_{ij} = N \sum_{\substack{f,g\,:\,f(1)=i,g(1)=j\\ f\setminus\{i\}=g\setminus\{j\}}} \alpha_f\,\bar{\alpha}_g,$$

and

$$\gamma_{ijkl} = \binom{N}{2} \sum_{\substack{f,g\,:\,f\setminus\{i,j\}=g\setminus\{k,l\}\\ f(1)=i,f(2)=j,g(1)=k,g(2)=l}} \alpha_f\,\overline{\alpha}_g.$$

1

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Chapter 6

Numerical illustrations of the MCTDHF hierarchy

Abstract

This chapter is dedicated to some numerical illustrations of the MCTDHF. The simulations are based on simple numerical schemes that are certainly not optimal for MCTDHF and do not preserve well all physical quantities such as normalization constraints or total energy. Our goal is not to develop or analyse numerical methods, but simply to illustrate the hierarchy of the MCTDHF system with a simple 2 particle system in one space dimension. We compare simulations of the exact 2 particle Schrödinger equation with MCTDHF for K = 6, and also a comparison with K = 4 and K = 2 = N which is the TDHF case.

Despite the simplicity of our approach, the numerical results show quite interesting properties like the good quality of the MCTDHF approximation the non-monotonous behaviour of the "correlation".

6.1 The MCTDHF equation for N = 2 particles

Recall that the MCTDHF approximation consists in forcing the wavefunction to evolve on the following set

$$\mathscr{M}_N^K = \left\{ \Psi \in \bigwedge_{i=1}^{N} L^2(\Omega) \,:\, \Psi(x_1,\ldots,x_N) = \sum_{\sigma\in\Sigma_{N,K}} C_\sigma(t)\,\Phi_\sigma(t,x_1,\ldots,x_N),\ (C,\Phi)\in\mathscr{F}_{N,K} \right\}, \tag{6.1}$$

for all $N \leq K$, where

$$\mathscr{F}_{N,K} = \left\{ (C,\Phi) \in \mathbb{C}^{\binom{K}{N}} \times L^2(\Omega)^K \,:\, C = (C_{\sigma_1},\ldots,C_{\sigma_{\binom{K}{N}}})^T,\ \sum_{\sigma\in\Sigma_{N,K}} |C_\sigma|^2 = 1, \right.$$
$$\left. \Phi = (\phi_1,\cdots,\phi_K)^T,\ \int_\Omega \phi_i\,\bar{\phi}_j\,dx = \delta_{i,j} \right\}, \tag{6.2}$$

with the $\binom{K}{N}-$component vector C being arranged in lexicographical order and

$$\Sigma_{N,K} = \left\{ \sigma = \{\sigma(1) < \ldots < \sigma(N)\} \subset \{1,\ldots,K\} \right\}, \qquad |\Sigma_{N,K}| = \binom{K}{N} := r.$$

In (6.2), Φ_σ denotes the normalized Slater determinant built from $\{\phi_i\}_{i\in\sigma}$. Obviously, we have $|\Sigma_{N,K}| = \binom{K}{N} := r$, that is Ψ is a linear combination of r Determinants. The constraints on the C_σ's and the ϕ_i's in $\mathscr{F}_{N,K}$ guarantee that $\| \Psi \|_{L^2(\Omega^N)} = 1$ since

$$\left\langle \Phi_\sigma \,|\, \Phi_\tau \right\rangle_{L^2(\Omega^N)} = \delta_{\sigma,\tau},$$

where $\left\langle f|g \right\rangle_{L^2(\Omega^N)} = \int_{\Omega^N} f(X_N)\,\overline{g}(X_N)\,dX_1$, $X_N := (x_1,\ldots,x_N)$ and $dX_N := dx_1\ldots dX_N$. $\overline{g}$ denotes the complex conjugate of g, while $\delta_{\sigma,\tau}$ is the Kronecker symbol for sets. The MCTDHF equations are the following

$$i\,\frac{d}{dt}C_\sigma(t) = \langle \Psi\,|V|\,\Phi_\sigma\rangle_{L^2(\Omega^N)}, \quad \sigma\in\Sigma_{N,K}, \tag{6.3a}$$

$$i\,\mathbf{\Gamma}[C(t)]\,\frac{\partial}{\partial t}\Phi(t) = \mathbf{\Gamma}[C(t)]\,\mathbb{H}\Phi + (\mathbf{I}-\mathbf{P}_\Phi)\mathbb{W}[C,\Phi]\,\Phi(t). \tag{6.3b}$$

Equations (6.3a) form a system of r first-order ordinary differential equations where

$$\left\langle \Phi_\sigma\,|V|\,\Phi_\tau \right\rangle_{L^2(\Omega^N)} \;=\; \sum_{1\le i<j\le N}\int_{\Omega^N} \Phi_\sigma(t,X_N)\,\overline{\Phi}_\tau(t,X_N)\,V(|x_i-x_j|)\,dX_N,$$

is the interaction between the Slater Determinants Φ_σ and Φ_τ. Note that in our formulation the free Hamiltonian does not occur in (6.3a) and we refer the reader to the fifth chapter for more details on the reason and the algebraic structure of the MCTDHF in general. Equation (6.3b) provides with a system of K non-linear PDEs on the ϕ_i's, where $\mathbb{H}$ is the operator matrix with entries $\mathbb{H}_{i,j} = \delta_{i,j}\,\mathbf{H}$ for all $1\le i,j\le K$, here

$$\mathbf{H} = -\frac{1}{2}\,\Delta_x + \sum_{i=1}^{M} U(|x-\bar{x}_i|).$$

The operator $\mathbf{P}_\Phi$ denotes the orthogonal projector onto $\mathrm{Span}\{\phi_i\}_{1\le i\le K}$, i.e.

$$\mathbf{P}_\Phi = \sum_{i=1}^{K}\langle\cdot,\phi_i\rangle_{L^2(\Omega)}\phi_i. \tag{6.4}$$

Finally, the $K\times K$ Hermitian matrices $\mathbf{\Gamma}[C]$ and $\mathbb{W}[C,\Phi]$ are given as follows

$$\mathbf{\Gamma}[C(t)]_{i,j} = \sum_{\substack{\sigma,\tau\in\Sigma_{N,K}\\ \sigma\setminus\{i\}=\tau\setminus\{j\}}} (-1)^{\sigma^{-1}(i)+\tau^{-1}(j)}\,\overline{C}_\sigma(t)\,C_\tau(t), \tag{6.5}$$

$$\mathbb{W}[C,\Phi](t,x)_{i,j} = 2\sum_{p,q=1}^{K}\gamma_{j,q,i,p}[C(t)]\int_\Omega \overline{\phi}_p(t,y)\,V(|x-y|)\,\phi_q(t,y)\,dy, \tag{6.6}$$

with $\sigma^{-1}(j)$ being the position of j in the set σ. Also, the coefficients $\gamma_{j,q,i,p}[C(t)]$ are quadratic in the C'_σs as follows

$$\gamma_{ijkl} = \frac{1}{2}\,(1-\delta_{i,j})(1-\delta_{k,l}) \sum_{\substack{\sigma,\tau\,|\,i,j\in\sigma,k,l\in\tau\\ \sigma\setminus\{i,j\}=\tau\setminus\{k,l\}}} (-1)^{\sigma}_{i,j}(-1)^{\tau}_{k,l}\,C_\sigma\,\overline{C}_\tau,$$

where

$$(-1)^{\tau}_{i,p} = \begin{cases} (-1)^{\{\tau^{-1}(i)+\tau^{-1}(p)+1\}} & if \quad i < p, \\ \\ (-1)^{\{\tau^{-1}(i)+\tau^{-1}(p)\}} & if \quad i > p. \end{cases}$$

We refer the reader to the previous chapters for more details.

From this point onward, we shall consider a 2-particle version of MCTHDF, which allows for some simplifications.

Let $N = 2, (C, \Phi) \in \mathscr{F}_{2,K}$ with an arbitrary $K \geq 2$. Then, we have

$$\Psi(t,x,y) = \sum_{\sigma \in \Sigma_{2K}} C_\sigma(t)\, \Phi_\sigma := \sum_{i,j=1}^{K} \alpha_{i,j}(t)\, \phi_i(t,x)\, \phi_j(t,y),$$

where the coefficients $\alpha_{i,j}$ are skew-symmetric with respect to their indices i.e. $\alpha_{i,j} = -\alpha_{j,i}$ and defined as follows

$$\alpha_{i,j} = \frac{1}{\sqrt{2}} C_{\sigma=\{i<j\}} \quad \text{such that} \quad \sum_{i,j=1}^{K} |\alpha_{i,j}|^2 = \sum_{\sigma \in \Sigma_{2,K}} |C_\sigma|^2 = 1.$$

In particular, we obtain that

$$\mathbf{\Gamma}[C]_{i,j}(t) := \mathbf{\Gamma}[\alpha]_{i,j}(t) = 2 \sum_{p=1}^{K} \overline{\alpha}_{i,p}(t)\, \alpha_{j,p}(t),$$

$$\mathbb{W}_{i,j}[C,\Phi](t,x) := \mathbb{W}_{i,j}[\alpha,\Phi](t,x) = 2 \sum_{p,q=1}^{K} \overline{\alpha}_{i,p}(t)\, \alpha_{j,q}(t) \int_\Omega \overline{\phi}_p(t,y)\, V(|x-y|)\, \phi_q(t,y)\, dy.$$

Moreover, if we denote by α the skew-symmetric matrix with entries $\alpha_{i,j}$ for $1 \leq i,j \leq K$, one observe that

$$\mathbf{\Gamma}[\alpha] = -2\,(\alpha\, \alpha^\star)^T. \tag{6.7}$$

Then, from the one hand, we have $\mathrm{rank}(\mathbf{\Gamma}[\alpha]) = \mathrm{rank}(\alpha)$ which is even. In particular the matrix $\mathbf{\Gamma}[\alpha]$ is singular for odd K. Since the invertibility of $\mathbf{\Gamma}[\alpha]$ is crucial for the analysis of the MCTDHF, we shall consider from now on even K and we refer the reader to [5, 6, 7, 9] for remarks and rank classification depending on K of the matrix $\mathbf{\Gamma}$ in the general case. Thus, we deal with the following system

$$\mathscr{S}_2 : \quad \begin{cases} i\, \frac{d}{dt}\, \alpha_{i,j}(t) = \sum_{p,q=1}^{K} D(\phi_p\, \overline{\phi}_i\,,\, \phi_q\, \overline{\phi}_j)\, \alpha_{p,q}(t), \\ \\ i\, \mathbf{\Gamma}[\alpha(t)]\, \frac{\partial}{\partial t}\, \Phi(t) = \mathbf{\Gamma}[\alpha(t)]\, \mathbb{H}\, \Phi + (\mathbf{I} - \mathbf{P}_\Phi)\, \mathbb{W}[\alpha,\Phi]\, \Phi(t), \end{cases}$$

where

$$D(\phi_p\, \overline{\phi}_i\,,\, \phi_q\, \overline{\phi}_j) = \int \int_{\Omega \times \Omega} \phi_p(t,x)\, \overline{\phi}_i(t,x)\, V(|x-y|)\, \phi_q(t,y)\, \overline{\phi}_j(t,y)\, dx\, dy.$$

These are the so called *bielectronic* integrals and from the numerical point of view, the most costly to compute.

6.2 The numerical methods

We want to solve numerically the system $\mathscr{S}_2$ for a given initial data $(C^0,\Phi^0)\in\mathscr{F}_{2,K}$.

For the sake of simplicity we work in a setting in one space dimension : For $b>a>0$ we use the domain $\Omega=[a,b]\in\mathbb{R}$ for our computation. Further, we impose periodic boundary conditions. Moreover, we use one nucleus, fixed at position $R=0$, so that $U(x)=-\frac{z}{|x|}$. Also we consider smoothed Coulomb interactions in order to avoid the singularities, as frequently done in physics, e.g. in [3].

The potentials are then given by $V_\varepsilon(|x-y|)=\frac{1}{|x-y|+\varepsilon}$ and $U_\varepsilon(x)=-\frac{z}{|x|+\varepsilon}$ with ε being a small constant that we set to 0.7408.

Thus, we want to solve the following system

$$\mathscr{S}_2:\quad\begin{cases} i\,\frac{d}{dt}\,\alpha_{i,j}(t)=\sum_{p,q=1}^K D_\varepsilon(\phi_p\,\overline{\phi}_i\,,\,\phi_q\,\overline{\phi}_j)\,\alpha_{p,q}(t),\\[2ex] i\,\mathbf{\Gamma}[\alpha(t)]\,\frac{\partial}{\partial t}\,\Phi(t)=\mathbf{\Gamma}[\alpha(t)]\,\mathbb{H}\,\Phi+(\mathbf{I}-\mathbf{P}_\Phi)\,\mathbb{W}_\varepsilon[\alpha,\Phi]\,\Phi(t),\\[2ex] (C(t=0),\Phi(t=0))=(C^0,\Phi^0)\in\mathscr{F}_{N,K},\ \phi_i(t,a)=\phi_i(t,b),\ t>0,\ i=1,\dots,K. \end{cases}$$

There are different possible schemes that allow to solve a coupled system of ODEs and PDEs. For instance, for many scientists, the fourth-order Runge-Kutta is the first, but the last as well, on ODE integrators. The choice is usually made depending on the required accuracy and the computation cost. Now, let us introduce the following, respectively, linear and non-linear flows

$$\mathscr{L}^t:\quad\begin{cases} \frac{\partial}{\partial t}\,\phi_p(t,x)=i\left(\frac{d^2}{dx^2}-U_\varepsilon(x)\right)\phi_p(t,x)\\[2ex] \phi_p(t=0)=\varphi_p,\quad p=1,\dots,K \end{cases}$$

and

$$\mathscr{N}^t:\quad\begin{cases} \begin{pmatrix}\frac{d}{dt}\,\alpha_{i,j}(t)\\[2ex]\frac{\partial}{\partial t}\,\phi_p(t,x)\end{pmatrix}=-i\begin{pmatrix}\sum_{p,q=1}^K D_\varepsilon(\phi_p\,\overline{\phi}_i\,,\,\phi_q\,\overline{\phi}_j)\,\alpha_{p,q}(t)\\[2ex]\sum_{q,l=1}^K\mathbf{\Gamma}[\alpha(t)]^{-1}_{p,q}\,(\mathbf{I}-\mathbf{P}_\Phi)\,\mathbb{W}_\varepsilon[\alpha,\Phi]_{q,l}\,\phi_l(t)\end{pmatrix}\\[2ex] \alpha_{i,j}(t=0)=\alpha^0_{i,j},\ \phi_p(t=0,x)=\phi^0_p,\ 1\le i<j\le K,\ 1\le p\le K. \end{cases}$$

Of course, the solutions are ε-dependent and we don't mention that explicitly for notation's lightness. Now, the system $\mathscr{S}_2$ can be solved by operators spiltting method. For instance, a first order splitting is then given by $\mathscr{L}^t\,\mathscr{N}^t$. We choose to solve $\mathscr{S}_2$, using the so called *Strang* splitting which is a splitting of order 2 and is achieved by $\mathscr{L}^{\frac{t}{2}}\,\mathscr{N}^t\,\mathscr{L}^{\frac{t}{2}}$. In explicit words, this scheme advance the solution of $\mathscr{S}_2$ from time t_n up to t_{n+1} as follows

- On $[t_n,t_n+\frac{\Delta t}{2}]$, solve
$$\frac{\partial}{\partial t}\,\phi_p(t,x)=i\left(\frac{d^2}{dx^2}-U_\varepsilon(x)\right)\phi_p(t,x),$$
for $p=1,\dots,K$.

- On $[t_n, t_{n+1}]$, solve

$$\begin{pmatrix} \frac{d}{dt}\,\alpha_{i,j}(t) \\ \\ \frac{\partial}{\partial t}\,\phi_p(t,x) \end{pmatrix} = -i \begin{pmatrix} \sum_{p,q=1}^{K} D_\varepsilon(\phi_p\,\overline{\phi}_i\,,\,\phi_q\,\overline{\phi}_j)\,\alpha_{p,q}(t) \\ \\ \sum_{q,l=1}^{K} \mathbf{\Gamma}[\alpha(t)]^{-1}_{p,q}\,(\mathbf{I}-\mathbf{P}_\Phi)\,\mathbb{W}_\varepsilon[\alpha,\Phi]_{q,l}\,\phi_l(t) \end{pmatrix},$$

- On $[t_n + \frac{\Delta t}{2}, t_{n+1}]$, solve

$$\frac{\partial}{\partial t}\,\phi_p(t,x) = i\left(\frac{d^2}{dx^2} - U_\varepsilon(x)\right)\phi_p(t,x),$$

  for $p = 1,\ldots,K$.

Let us now precise how each part is solved. The linear part is again solved by a *strang* splitting. Let M be an even positive integer, let $\Delta x > 0$ be the spatial mesh size with $\Delta x = \frac{b-a}{M}$. Also denote the step of time by Δt. Then, the grid points and the time step are

$$x_j := a + j\,\Delta x, \quad t_n = n\,\Delta t, \quad j = 1,\ldots,M, \quad \text{and} \quad n = 0,1,2,\ldots.$$

Next, let $\alpha^n_{i,j}$ and $\phi^{p,n}_i$ be respectively the approximation of $\alpha_{i,j}(t_n)$ and $\phi_i(t_n,x_p)$. Now, the linear part, corresponding to the flow $\mathscr{L}^t$ is then solved for given coefficients and for all $p = 1,\ldots,K$ on $[t_n,t_{n+1}]$ as follows

- $\phi^{j,\star}_p = \exp\left(-\dfrac{i\,\Delta t}{2}\,U_\varepsilon(x)\right)\phi^{j,n}_p, \quad j = 0,\ldots,M-1,$
- $\phi^{j,\star\star}_p = \dfrac{1}{M}\displaystyle\sum_{l=-\frac{M}{2}}^{\frac{M}{2}-1} \exp\left(-i\,\frac{\Delta t}{2}\left[\frac{2\,\pi\,l}{b-a}\right]^2\right)\hat{\phi}^{l,\star}_p \exp\left(i\frac{2\pi}{M}\,jl\right), \quad j = 0,\ldots,M-1,$
- $\phi^{j,n+1}_p = \exp\left(-\dfrac{i\,\Delta t}{2}\,U_\varepsilon(x)\right)\phi^{j,\star\star}_p, \quad j = 0,\ldots,M-1,$

where $\hat{\phi}^{l,\star}_p$ are the Fourier Coefficients of $\phi^\star_p$ given by

$$\hat{\phi}^{l,\star}_p = \sum_{j=1}^{M-1} \phi^{j,\star}_p \exp\left(i\frac{2\pi}{M}\,jl\right).$$

It is straightforward that this splitting scheme for the linear part is unconditionally stable. Indeed (see [1]), for $p = 1,\ldots,K$, let $\varphi^\star_p$ denotes the M-component vector $(\phi^{0,\star}_p,\ldots,\phi^{M-1,\star}_p)$ and introduce the $L^2(\Omega)$ discrete norm as follows

$$\|\varphi^\star_p\|_{\ell^2} = \sqrt{\Delta x \sum_{k=0}^{M-1} |\phi^{k,\star}_p|^2}.$$

Thus, one has

$$\begin{aligned}
\|\varphi_p^{n+1}\|_{\ell^2}^2 = \Delta x \sum_{k=0}^{M-1} \left|\phi_p^{k,n+1}\right|^2 &= \Delta x \sum_{k=0}^{M-1} \left|\exp\left(-i\frac{\Delta t}{2}U_\varepsilon(x_k)\right)\phi_p^{k,\star\star}\right|^2 = \Delta x \sum_{k=0}^{M-1} \left|\phi_p^{k,\star\star}\right|^2, \\
&= \Delta x \sum_{k=0}^{M-1} \left|\frac{1}{M}\sum_{l=\frac{M}{2}}^{\frac{M}{2}-1} \exp\left(-i\frac{\Delta t}{2}\left[\frac{2\pi l}{b-a}\right]^2\right)\hat{\phi}_p^{l,\star}\exp\left(i\frac{2\pi}{M}k\,l\right)\right|^2, \\
&= \frac{\Delta x}{M}\sum_{l=\frac{M}{2}}^{\frac{M}{2}-1}\left|\exp\left(-i\frac{\Delta t}{2}\left[\frac{2\pi l}{b-a}\right]^2\right)\hat{\phi}_p^{l,\star}\right|^2 = \frac{\Delta x}{M}\sum_{l=\frac{M}{2}}^{\frac{M}{2}-1}\left|\hat{\phi}_p^{l,\star}\right|^2, \\
&= \frac{\Delta x}{M}\sum_{l=\frac{M}{2}}^{\frac{M}{2}-1}\left|\sum_{k=0}^{M-1}\phi_p^{k,\star}\exp\left(i\frac{2\pi}{M}k\,l\right)\right|^2 = \Delta x\sum_{k=0}^{M-1}\left|\phi_p^{k,\star}\right|^2, \\
&= \Delta x \sum_{k=0}^{M-1}\left|\exp(-i\frac{\Delta t}{2}U_\varepsilon(x_k))\phi_p^{k,n}\right|^2 = \Delta x\sum_{k=0}^{M-1}\left|\phi_p^{k,n}\right|^2, \\
&= \|\varphi_p^n\|_{\ell^2}^2.
\end{aligned}$$

It remains to solve the non-linear part of the system $\mathscr{S}_2$ corresponding to the flow $\mathscr{N}^t$. Observe that the system is coupled now. We will decouple it by solving the ODE system on the coefficients for a given functions and *vice versa*. For simplicity, we use the C formulation, thus, it is easy to transform the ODEs system as follows

$$i\frac{d}{dt}C(t) = \mathscr{M}_\varepsilon(\Phi(t))\,C(t),$$

with $\mathscr{M}$ being the $r\times r$ matrix given by its entries

$$\mathscr{M}_{\varepsilon\,\sigma,\tau}(\Phi(t)) := \left\langle \Phi_\tau(t,x,y)\,|\,\frac{1}{|x-y|+\varepsilon}\,|\,\Phi_\sigma(t,x,y)\right\rangle_{L^2(\Omega^2)}.$$

We use an implicit method to solve this ODE for a given set of functions $\phi_1,\ldots,\phi_K$. That is

$$C^{n+1}-C^n = -i\frac{\Delta t}{2}\mathscr{M}_\varepsilon(\Phi^n)\left(C^{n+1}+C^n\right), \tag{6.8}$$

thus

$$C^{n+1} = \left(I_r + i\frac{\Delta t}{2}\mathscr{M}_\varepsilon(\Phi^n)\right)^{-1}\left(I_r - i\frac{\Delta t}{2}\mathscr{M}_\varepsilon(\Phi^n)\right)^{-1}C^n,$$

with I_r being the $r\times r$ identity matrix. This scheme is easily translated in term of the coefficient $\alpha_{\ldots}$. Its particular advantage is that it conserves the norm of the coefficients. That is for a given initial data $(C^0,\Phi^0)\in\mathscr{F}_{2,K}$. Then, we have for all positive time

$$\sum_{\sigma\in\Sigma_{2,K}}|C_\sigma(t)|^2 = \sum_{\sigma\in\Sigma_{2,K}}|C_\sigma^0|^2.$$

In fact, multiply (ℓ^2 scalar product) the equation (6.8) by $\overline{C}^{n+1} - \overline{C}^n$, we obtain

$$\left|C^{n+1}\right|^2 - |C^n|^2 + 2\,i\,\Im(C^{n+1}\cdot C^n) \quad = \quad -i\,\frac{\Delta t}{2}\,\mathscr{M}_\varepsilon(\Phi^n)\,\left(C^{n+1}+C^n\right)\cdot(\overline{C}^{n+1}+\overline{C}^n).$$

Now, notice that the matrix $\mathscr{M}_\varepsilon(\Phi^n)$ is clearly Hermitian, thus, for all r-component vector $\mathbb{X}$, one has obviously $\mathscr{M}_\varepsilon(\Phi^n)\,\mathbb{X}\cdot\overline{\mathbb{X}} \in \mathbb{R}$. Hence, by taking the real part in the equation above one get

$$\left|C^{n+1}\right|^2 - |C^n|^2 \simeq \Delta t\,\frac{d}{dt}\,|C(t)|^2 = 0.$$

This proves the conservation property. From the practical point of view, the most costly step is the inversion of the matrices $I_r \pm i\,\frac{\Delta t}{2}\,\mathscr{M}_\varepsilon(\Phi^n)$. However since their size is clearly very reasonable, this inversion do not affect so much the computation cost. At this level, the construction of the matrix $\mathscr{M}_\varepsilon(\Phi^n)$ can be restrictive. In fact, the computation of the *bielectronic integrals* $D_\varepsilon(\phi_i\,\overline{\phi}_p\,,\,\phi_j\,\overline{\phi}_q)$ is heavy and this is one of the issues that we will tackle in a future work by using a contracted Gaussian expansion for the orbitals or a fast multi-pole algorithm. However, the integrals computed at this step, will be stored and used again for the first evaluation of the Runge-Kutta method that we will use in order to solve the PDEs part of $\mathscr{N}^t$. We want to solve

$$\frac{\partial}{\partial t}\phi_p(t,x) = -i\,\sum_{q,l=1}^{K}\,\mathbf{\Gamma}[\alpha(t)]^{-1}_{p,q}\,(\mathbf{I}-\mathbf{P}_\Phi)\,\mathbb{W}_\varepsilon[\alpha,\Phi]_{q,l}\,\phi_l(t),$$

using the fourth-order Runge-Kutta method for a given coefficients. After approximation of the time derivative, we get

$$\phi_p^{n+1} = \phi_p^n - i\,\Delta t\,\sum_{q,l=1}^{K}\,\mathbf{\Gamma}[\alpha^{n+1}]^{-1}_{p,q}\,(\mathbf{I}-\mathbf{P}_{\Phi^n})\,\mathbb{W}_\varepsilon[\alpha^{n+1},\Phi^n]_{q,l}\,\phi_l^n.$$

Thus, the method is the following for $1 \le p \le K$

First step t_n
- $\mathscr{I}_1^p = -i\,\Delta t\,\sum_{q,l=1}^{K}\mathbf{\Gamma}[\alpha^{n+1}]^{-1}_{p,q}\,(\mathbf{I}-\mathbf{P}_{\Phi^n})\,\mathbb{W}_\varepsilon[\alpha^{n+1},\Phi^n]_{q,l}\,\phi_l^n,$

Second step $t_n+\frac{\Delta t}{2}$
- $\mathscr{I}_2^p = -i\,\Delta t\,\sum_{q,l=1}^{K}\mathbf{\Gamma}[\alpha^{n+1}]^{-1}_{p,q}\,(\mathbf{I}-\mathbf{P}_{\Phi^n+\frac{1}{2}\mathscr{I}_1})\,\mathbb{W}_\varepsilon[\alpha^{n+1},\Phi^n+\frac{1}{2}\,\mathscr{I}_1]_{q,l}\times \times\left(\phi_l^n+\frac{1}{2}\,\mathscr{I}_1^l\right),$

Third step $t_n+\frac{\Delta t}{2}$
- $\mathscr{I}_3^p = -i\,\Delta t\,\sum_{q,l=1}^{K}\mathbf{\Gamma}[\alpha^{n+1}]^{-1}_{p,q}\,(\mathbf{I}-\mathbf{P}_{\Phi^n+\frac{1}{2}\mathscr{I}_2})\,\mathbb{W}_\varepsilon[\alpha^{n+1},\Phi^n+\frac{1}{2}\,\mathscr{I}_2]_{q,l}\times \times\left(\phi_l^n+\frac{1}{2}\,\mathscr{I}_2^l\right),$

Fourth step t_{n+1}
- $\mathscr{I}_4^p = -i\,\Delta t\,\sum_{q,l=1}^{K}\mathbf{\Gamma}[\alpha^{n+1}]^{-1}_{p,q}\,(\mathbf{I}-\mathbf{P}_{\Phi^n+\mathscr{I}_3})\,\mathbb{W}_\varepsilon[\alpha^{n+1},\Phi^n+\mathscr{I}_3]_{q,l}\times \times\left(\phi_l^n+\mathscr{I}_3^l\right),$

$$\phi_p^{n+1} = \phi_p^n + \tfrac{1}{6}\left(\mathscr{I}_1^p + 2\left[\mathscr{I}_2^p+\mathscr{I}_3^p\right]+\mathscr{I}_4^p\right).$$

Above, we reserved the notation $\mathscr{I}_k$ for $k = 1,2,3$ for the K-component vector $(\mathscr{I}_k^1, \ldots, \mathscr{I}_k^K)^T$. Obviously, this method requires four evaluations of the non-linear part per step of time Δt. However, observe that the *bielectronic integrals* needed for the computation of $\mathscr{I}_1$ are already computed and stored during the resolution of the ODEs system. even, with this restriction, the fourth-order Runge-Kutta seems to be very costly as soon as the number of grid points or orbitals is increased. A second order Runge-Kutta method seems to be the best compromise, it requires only two evaluations of the non-linear part with one is already computed as explained above. This method reads

First step t_n

- $\mathscr{I}_1^p = -i\,\Delta t \sum_{q,l=1}^K [\alpha_{p,q}^{n+1}]^{-1} \left(\mathbf{I} - \mathbf{P}_{\Phi^n}\right) \mathbb{W}_\varepsilon[\alpha^{n+1}, \Phi^n]_{q,l}\, \phi_l^n,$

Second step $t_n + \frac{\Delta t}{2}$

- $\mathscr{I}_2^p = -i\,\Delta t \sum_{q,l=1}^K [\alpha_{p,q}^{n+1}]^{-1} \left(\mathbf{I} - \mathbf{P}_{\Phi^n + \frac{1}{2}\mathscr{I}_1}\right) \mathbb{W}_\varepsilon[\alpha^{n+1}, \Phi^n + \frac{1}{2}\mathscr{I}_1]_{q,l} \times \times \left(\phi_l^n + \frac{1}{2}\mathscr{I}_1^l\right),$

$$\phi_p^{n+1} = \phi_p^n + \mathscr{I}_2^p.$$

An extra gain can be achieved using the symmetry of the *bielectronic integrals*. In fact, they obey obviously to

$$\begin{aligned} D(\phi_i\,\overline{\phi}_p\,,\,\phi_j\,\overline{\phi}_q) &= D(\phi_j\,\overline{\phi}_q\,,\,\phi_i\,\overline{\phi}_p), \\ &= \overline{D}(\phi_p\,\overline{\phi}_i\,,\,\phi_q\,\overline{\phi}_j), \\ &= \overline{D}(\phi_q\,\overline{\phi}_j\,,\,\phi_p\,\overline{\phi}_i). \end{aligned}$$

The integration can be achieved using any standard quadrature formula since we use a smoothed Coulomb interactions. The main weakness of the scheme is that it does not conserve neither the orthogonality of the orbitals neither the energy. Thus, following physicists/chemists [3, 2], an orthogonalization procedure is performed after each Runge-Kutta step. This procedure is done using SVD decomposition since a simple Gram-Schmidt procedure is simply *terrible* from the numerical point of view because of the build-up of roundoff errors.

One of the motivations of the MCTDHF method is the so called *Correlation*. This concept somewhat intuitive but deep is far from being well-known from the mathematical point of view. Different definitions were stated to such entropy function in the physical literature, unfortunately, usually far from taking advantage from the potential of the MCTDHF. In [8], the authors defined the correlation as follows

$$\mathfrak{E} = -[\Psi \otimes \Psi]_{:1} \log\left([\Psi \otimes \Psi]_{:1}\right) - (1 - [\Psi \otimes \Psi]_{:1}) \log\left(1 - [\Psi \otimes \Psi]_{:1}\right),$$

where $[\Psi \otimes \Psi]_{:1}$ denotes the kernel of the *first order density operator* for a given function in $L^2(\Omega^2)$ and is given as follows

$$[\Psi \otimes \Psi]_{:1}(x,y) = 2 \int_a^b \Psi(x,z)\,\overline{\Psi}(y,z)\,dz.$$

Equivalently to the operator formulation above, in the MCTDHF case one can formulate the correlation as follows

$$\mathfrak{E} = -\sum_{i=1}^{K} \left[\gamma_i \log(\gamma_i) + (1-\gamma_i)\log(1-\gamma_i)\right], \tag{6.9}$$

where the γ_i's are the eigenvalues of $[\Psi \otimes \Psi]_{:1}(x,y)$, that is the matrix $\mathbf{\Gamma}[C]$. One can observe easily that $\sum_{i=1}^{K} \gamma_i = 2$, $0 \leq \gamma_i \leq 1$ and we refer the reader to the previous chapters of the present manuscript for more details. In particular a loss of rank can occur when one or more of these eigenvalues vanishes and this is the main difficulty of the MCTDHF. In order to ensure the invertibility of the matrix $\mathbf{\Gamma}[C]$, physicist perturb it by adding a small term. However, in our numerical simulations, we noted that such loss of rank never occurs. Again, this issue is to be confirmed and understood in the future.

In [3] and reference therein, the authors start usually with an initial data coming from a imaginary time propagation which coincides, under certain conditions, with the ground state. Our approach is slightly similar. In fact, we compute the ground state Ψ_{G} for a 2 particle system using the exact Schrödinger equation. Next, we consider an arbitrary orthonormal basis of $L^2(\Omega)$, say $\phi_1^0, \ldots, \phi_L^0$ with $L \geq K \geq 2$. Thus, the tensorial products obtained from this basis form an orthonormal basis of $L^2(\Omega) \otimes L^2(\Omega)$. Next, we expand the ground state in this basis in order to get the coefficients. In other words,

$$\alpha_{i,j}^0 = \int_a^b \int_a^b \Psi_{\mathrm{G}}(x,y)\, \overline{\phi}_i^0(x)\, \overline{\phi}_j^0(y)\, dx\, dy, \quad 1 \leq i < j \leq L.$$

In particular, the larger is L, the better is the approximation of the ground state Ψ_{G} and one recover it exactly for $l = +\infty$ from a purely mathematical point of view. Next, one compute the associated energy using the following formula

$$\begin{aligned} \mathscr{E}_\varepsilon(\Psi_{\mathrm{G}}) &= \sum_{k=1}^{L} \int_a^b \left(\left| \sum_{l=1}^{L} \alpha_{l,k}^0 \nabla \phi_i^0(x) \right|^2 - \frac{2Z}{|x|+\varepsilon} \left| \sum_{l=1}^{L} \alpha_{l,k}^0\, \phi_i^0(x) \right|^2 \right) dx \\ &+ \sum_{i,j,p,q=1}^{L} \alpha_{i,j}^0\, \overline{\alpha}_{p,q}^0\, D_\varepsilon(\phi_i^0\, \overline{\phi}_p^0,\, \phi_j^0\, \overline{\phi}_q^0). \end{aligned} \tag{6.10}$$

The initial K-orbitals wavefunction we need will be chosen from the set of the functions $\phi_1^0, \ldots, \phi_L^0$. This allows for $\binom{L}{K}$ possibility. The selection criterion will be the wavefunction that maximizes the correlation or the one that has the closer energy to the one associated to Ψ_{G}. However, the correlation being, by definition, zero in the Hartree-Fock case, that is $N = K = 2$, we will not be able to choose 2 orbitals from the basis $\phi_1^0, \ldots, \phi_L^0$ following this criterion. Thus, the algorithm for the selection of the initial data is then the following

- Get the ground state Ψ_{G} of 2 particles.

- Expand Ψ_{G} in a chosen orthonormal basis $\phi_1^0, \ldots, \phi_L^0$ for $L \geq K \geq 2$ and compute the energy using the formula (6.10).

- Select a set of K functions from $\phi_1^0, \ldots, \phi_L^0$, compute the coefficients by projection onto Ψ_{G} and compute the associated energy.

- Choose the set of orbitals and coefficients that has the closer energy to $\mathscr{E}_\varepsilon(\Psi_{\mathrm{G}})$.

6.3 Numerical results

Now we present the numerical results. As said before, our preliminary simple scheme does not yield a good conservation of the normalization constraints on the orbitals and of the total energy.

For our simple illustrations, we made another simplification by considering a system of 2 interacting electrons only, that is we switch off the nuclei-electrons interaction by setting $U \equiv 0$. Thus, instead of the function denoted Ψ_G, we shall consider any periodic and skew-symmetric function in $L^2(\Omega^2)$.

6.3.1 Exact Schrödinger

In order to compare our results with the "exact" many body solution, we implemented a code that solves the exact Schrödinger equation (5.2) for 2 electrons. The scheme is based on a 2-dimensional FFT and a second order splitting. Thus, the scheme conserves the total energy and the normalization of the wavefunction $\Psi(t,x,y)$. For instance, let any skew-symmetric periodic wavefunction $\Psi^0(x,y)$ on $[-\pi,\pi]$ be an initial data. Thus, using a 65 grid points with a step of time $\Delta t = 10^{-3}$, one observe after 1500 iterations

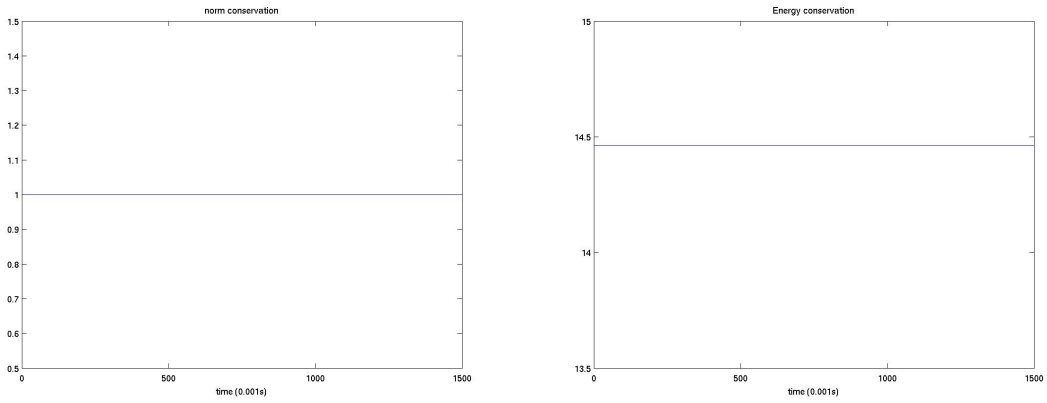

Fig 1: Left: $L^2([-\pi,\pi]^2)$ norm. Right: Exact energy.

For illustration, one compute the associated initial and final density matrices, respectively $[\Psi \otimes \Psi]_{:1}(t = 0x,y)$ and $[\Psi \otimes \Psi]_{:1}(t = 1.5s,x,y)$. The following plots correspond to their complex modulus.

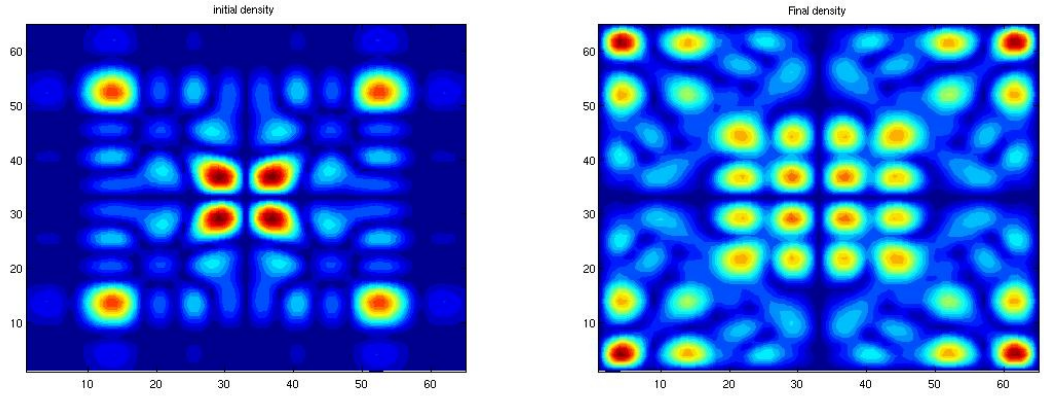

Fig 2: Left: $|[\Psi\otimes\Psi]_{:1}(t=0,x,y)|$. *Right:* $|[\Psi\otimes\Psi]_{:1}(t=1.5s,x,y)|$.

Let us now, see how the correlation behaves when one consider λV instead of V as interaction for a given real $\lambda > 0$.

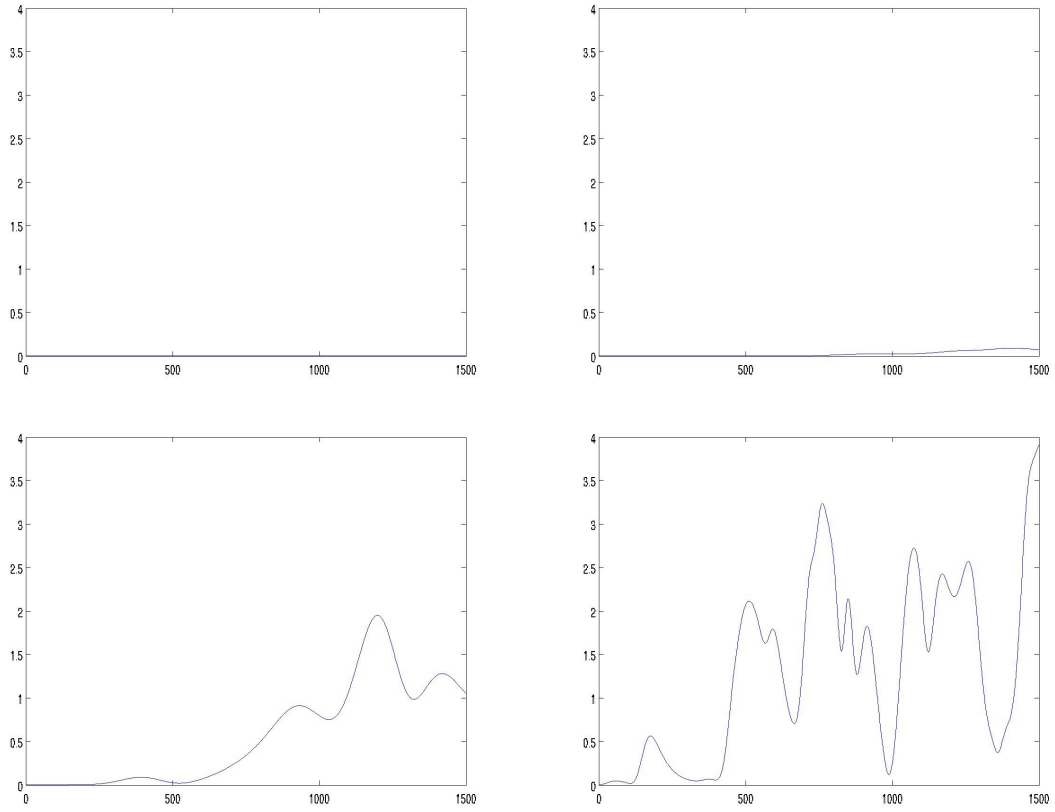

Fig3: Top left: $\lambda = 0$. *Top right:* $\lambda = 1$.
Bottom left:$\lambda = 10$. *Bottom Right:* $\lambda = 100$.

Intuitively, the concept of the correlation depends only on the binary (or more) interactions. Thus, the parts of the equation involving one particle operators don't affect the behaviour of this quantity.

This can be observed by picking first $\lambda = 0$. Moreover, changing the *amplitude* of the interaction induces a change on the correlation.

6.3.2 The multiconfiguration

From this point onward, we fixe an arbitrary initial basis of $L^2([-\pi,\pi])$, for instance, we consider a set of *contracted Gaussians*, that is a Hermit polynomes multiplied by Gaussians. Then, the energy of each configuration for a given even number of orbitals K is then computed and a set of orbitals is selected with respect to the energy criterion. For instance, let $L = 10$ and $K = 4$ with a 65 grid points, the output is as follows

Table 6.1: **Selection of the initial set of orbitals Output**

Set of orbitals				**Associated energy**	**Associated correlation**
1	2	3	4	10.8465272147042	8.468075282035520E-002
1	2	3	5	8.64315754916613	3.499703871743529E-004
1	2	3	6	7.97109497717691	1.485685419474745E-002
1	2	3	7	10.9937537047303	3.628647527529055E-002
1	2	3	8	11.2644436890320	2.489284028909053E-003
1	2	3	9	11.0101659961229	9.455604833788367E-002
1	2	3	10	10.2371271153711	2.685906666840111E-003
1	2	4	5	12.9926918192775	8.363662129343810E-002
1	2	4	6	8.93649031045242	1.206612394342400E-003
1	2	4	7	15.6932778595730	1.099499541717949E-002
1	2	4	8	13.1516089863967	0.130770071821374
1	2	4	9	13.4769555041006	9.211024298227306E-002
1	2	4	10	12.5029875869822	0.247384336921126
1	2	5	6	10.7824267483824	2.692142258957693E-003
1	2	5	7	13.8062221548991	4.546643547967373E-002
1	2	5	8	12.0520810518688	6.364574911489363E-003
1	2	5	9	14.0673345974119	0.127756566665871
1	2	5	10	15.1392837773325	6.788066745206695E-003
1	2	6	7	8.21368805686139	3.009383994043378E-003
1	2	6	8	10.9632841951628	3.000286845879582E-002
1	2	6	9	11.4738570925081	1.957948985476239E-002
1	2	6	10	10.3854238540620	5.875741091162416E-002
1	2	7	8	13.8885881419334	8.381039546144331E-002
1	2	7	9	10.2988391167879	1.709904518750193E-002
1	2	7	10	13.4443692030507	0.160928735999961
1	2	8	9	14.1274272883246	0.243829790262993
1	2	8	10	12.3102818997941	2.333976587877232E-002
1	2	9	10	13.5646127899092	0.451465684721640
1	3	4	5	7.87078782853896	1.636614736840354E-006
1	3	4	6	6.19786262718999	9.099720668817418E-003
1	3	4	7	12.8825946040691	2.431902176354137E-004

...

Set of Orbitals				Associated energy	Associated correlation
1	3	4	8	12.7269742139802	6.622435279690364E-006
1	3	4	9	10.0799510773386	4.349384904557678E-002
1	3	4	10	10.4793289124025	5.617242552433166E-006
1	3	5	6	5.93694018109580	3.393449157576057E-007
1	3	5	7	8.82466210164216	1.061063106583041E-006
1	3	5	8	7.09482770569479	1.288464679459618E-002
1	3	5	9	9.01315447027433	3.457322183583682E-006
1	3	5	10	9.30379213006178	1.183313658130454E-002
1	3	6	7	5.81507572354313	3.280299249948173E-003
1	3	6	8	10.7086276047680	1.293245022885519E-006
1	3	6	9	9.07045358771142	3.823365931843832E-003
1	3	6	10	8.45450837276313	1.431345783396643E-006
1	3	7	8	13.3836525118370	3.938589371434289E-006
1	3	7	9	7.57145154625088	1.607063041566143E-003
1	3	7	10	11.2524058756145	4.780346812434234E-006
1	3	8	9	13.4609965068321	1.265150358359388E-005
1	3	8	10	9.61603789847356	2.769193568713035E-002
1	3	9	10	11.3707541016595	1.590213903198376E-005
1	4	5	6	8.85643238660779	1.168509421374699E-002
1	4	5	7	15.5668749107906	4.055023541994347E-002
1	4	5	8	8.49595043088762	4.384507955009847E-006
1	4	5	9	12.5876711176085	5.422341515115253E-002
1	4	5	10	13.5494753232519	2.067852736664297E-006
1	4	6	7	7.89441544493519	1.559582193344991E-003
1	4	6	8	8.94296884119444	2.206036290746799E-002
1	4	6	9	10.0334019117902	1.601700072570559E-002
1	4	6	10	8.36703016078546	4.540507279828361E-002
1	4	7	8	15.6102689589845	7.849512101152376E-002
1	4	7	9	12.0267360740937	2.965045454491019E-002
1	4	7	10	15.1060537869148	0.160075919993801
1	4	8	9	12.8293357170234	0.102169971511459
1	4	8	10	8.48242505970105	7.196502821311391E-006
1	4	9	10	12.1821021359460	0.209394231122627
1	5	6	7	8.50733942000344	2.342025302734037E-006
1	5	6	8	6.88727279642921	6.700523824595840E-007
1	5	6	9	11.7763276198510	5.495262603709819E-003
1	5	6	10	11.8727720871981	7.291423721593757E-007
1	5	7	8	9.67195600779891	1.880022813928544E-006
1	5	7	9	10.5013505000236	2.765831835455346E-003
1	5	7	10	14.7271885090694	2.864032519968393E-006
1	5	8	9	10.2015972378938	5.994307601600371E-006
1	5	8	10	11.3945857808501	3.354536228103554E-002
1	5	9	10	14.9994662726110	1.086729102000794E-005

...

Set of Orbitals				Associated energy	Associated correlation
1	6	7	8	8.75330566586298	9.768428098352169E-003
1	6	7	9	8.17571638184922	3.816202677673603E-003
1	6	7	10	8.16056693551057	2.108404970400407E-002
1	6	8	9	11.8631140603940	2.154423109887676E-003
1	6	8	10	6.81421616031232	6.552430893747829E-007
1	6	9	10	11.2632998065670	2.684018781304195E-002
1	7	8	9	10.5393932147610	7.092401048708107E-003
1	7	8	10	9.81164253563280	1.267379827428919E-006
1	7	9	10	10.1965209045295	1.699441220688883E-002
1	8	9	10	10.0355800618622	3.796981568126451E-006
2	3	4	5	8.45350209327711	3.188507967604445E-002
2	3	4	6	7.27278078434052	1.609436921190702E-005
2	3	4	7	14.2980991720192	1.880589823389776E-004
2	3	4	8	13.6931627922550	7.848166578892833E-002
2	3	4	9	11.6089364125124	1.755964099993600E-003
2	3	4	10	11.6060367679192	9.523443194851949E-002
2	3	5	6	5.86578998378328	5.787849875005389E-003
2	3	5	7	8.96831233908502	1.481697459178053E-002
2	3	5	8	8.20710047397160	1.829936523337293E-003
2	3	5	9	9.07012797299151	4.013395824311421E-002
2	3	5	10	9.01126813427181	4.982132893947317E-005
2	3	6	7	6.86996980719564	5.222258108572570E-005
2	3	6	8	11.2503169890877	1.531251837379042E-002
2	3	6	9	10.0350635271519	3.941887966760974E-004
2	3	6	10	9.07826650128112	1.785524383930383E-002
2	3	7	8	14.1739115135159	3.928194664234486E-002
2	3	7	9	8.87323941161785	4.479103530938020E-004
2	3	7	10	12.1622088062683	4.411913105367588E-002
2	3	8	9	14.2033731364597	0.105111279539307
2	3	8	10	12.4146459736628	8.128492952582620E-003
2	3	9	10	12.2343005277686	0.112484366635865
2	4	5	6	8.26955229247387	6.064224505371168E-006
2	4	5	7	15.2081040733564	6.703988451698996E-005
2	4	5	8	8.52844638317382	1.470624707062237E-002
2	4	5	9	12.3478760521929	5.718682866351114E-004
2	4	5	10	13.5511410789528	2.207996830826149E-002
2	4	6	7	4.14979030795444	2.443397292079567E-004
2	4	6	8	8.95389702971346	2.987564543987963E-005
2	4	6	9	8.42997210879951	3.525254200131385E-004
2	4	6	10	8.14529915641917	9.630984008018000E-006
2	4	7	8	15.8005402207183	5.550776826126325E-005
2	4	7	9	10.4218500196052	1.044382477267641E-003
2	4	7	10	15.0308240697935	8.953114363977930E-005

...

Set of Orbitals				Associated energy	Associated correlation
2	4	8	9	13.0582449336856	4.004427755319926E-004
2	4	8	10	8.60490532422243	2.810780473073465E-003
2	4	9	10	12.1699673082694	5.643969985649554E-004
2	5	6	7	7.85072889272382	1.824222746756757E-005
2	5	6	8	6.36653948075148	2.954468928607773E-003
2	5	6	9	11.0819801091590	1.368203197607117E-004
2	5	6	10	11.3378385791900	4.410644321197371E-003
2	5	7	8	9.22404180623196	8.038063021317777E-003
2	5	7	9	9.94065712228245	1.755062330073659E-004
2	5	7	10	14.2998230836479	1.179417981693946E-002
2	5	8	9	9.64355931880255	2.297443417871021E-002
2	5	8	10	9.61610717322080	2.692120837543126E-004
2	5	9	10	14.4601052164887	3.266397188035823E-002
2	6	7	8	8.75404921070453	1.461471173546220E-005
2	6	7	9	5.77572159274666	2.776546921782270E-004
2	6	7	10	7.96028094511936	2.277741424793254E-005
2	6	8	9	11.8273037111576	1.000064758337133E-004
2	6	8	10	6.56435984832734	6.038710480622052E-004
2	6	9	10	10.9942569741814	1.434078105933682E-004
2	7	8	9	10.6197120422606	1.415538979449973E-004
2	7	8	10	9.60269891639605	1.783330115872930E-003
2	7	9	10	10.0881440519082	2.131921524854233E-004
2	8	9	10	9.73247843180107	5.446430338983279E-003
3	4	5	6	4.03569293068853	6.208517417507256E-003
3	4	5	7	10.7987449428298	2.208572392113298E-002
3	4	5	8	7.01162071838049	3.280916300435060E-006
3	4	5	9	7.91082213285334	3.192309472329722E-002
3	4	5	10	9.24758660362106	1.064055681593464E-006
3	4	6	7	6.40996319029963	2.089676554235317E-002
3	4	6	8	9.33885676116142	1.745021558392962E-002
3	4	6	9	8.15470113997604	4.602846802081338E-004
3	4	6	10	7.18765740748028	2.158360780268574E-002
3	4	7	8	16.0313426695782	6.459974320423918E-002
3	4	7	9	11.2174774080847	1.055335628714927E-003
3	4	7	10	13.9356089124601	8.029620520589945E-002
3	4	8	9	13.1654326969459	9.442870754569285E-002
3	4	8	10	10.0750878917536	1.162712443951421E-005
3	4	9	10	11.0857969009322	0.122565848735751
3	5	6	7	3.62987422706307	2.499670282531226E-003
3	5	6	8	5.18157254808066	4.103108949813873E-007
3	5	6	9	6.88684615520624	3.339645635283892E-003
3	5	6	10	7.19555579933215	3.528699978103184E-007
3	5	7	8	7.99269582906212	9.945006785970567E-007

...

Set of Orbitals				Associated energy	Associated correlation
3	5	7	9	5.60325620092113	1.832185260913877E-003
3	5	7	10	10.0467336170946	1.342859259744534E-006
3	5	8	9	8.29653843727806	2.497017345913645E-003
3	5	8	10	7.19974351038951	1.323888534906463E-002
3	5	9	10	10.1451562709467	5.134390616992530E-006
3	6	7	8	9.01660762935080	8.461644859699391E-003
3	6	7	9	6.85426115637677	1.447346765746235E-004
3	6	7	10	6.91063040503147	1.082558800784234E-002
3	6	8	9	12.2090643209993	1.161873827752455E-002
3	6	8	10	8.46549004637455	1.008980477770289E-006
3	6	9	10	10.0521669422915	1.565236784989910E-002
3	7	8	9	10.7772205339303	6.969515807343515E-003
3	7	8	10	11.4723032652339	1.625786911506504E-006
3	7	9	10	8.82321728268259	9.721258754979263E-003
3	8	9	10	11.6999261411195	3.457684838040606E-006
4	5	6	7	7.35665581945235	1.421531784695226E-005
4	5	6	8	4.34935498631911	3.675626196200265E-003
4	5	6	9	9.05901208282037	1.718267078019759E-004
4	5	6	10	9.39030878406997	5.889048957449076E-003
4	5	7	8	11.0823051954633	2.206373443428034E-004
4	5	7	9	12.4337116892361	4.440457935702112E-004
4	5	7	10	16.1230722775250	2.431957887807986E-002
4	5	8	9	8.25130876737266	2.307312443990034E-002
4	5	8	10	7.35118222675249	3.918680965887761E-006
4	5	9	10	13.1942713135314	3.844162348429523E-002
4	6	7	8	8.11782576406064	1.172032936095611E-005
4	6	7	9	3.91588423354505	1.188336925021825E-004
4	6	7	10	7.30435423334947	1.742985551753642E-005
4	6	8	9	9.67435946956570	1.328965568574836E-004
4	6	8	10	4.47978685250523	9.225849750950496E-004
4	6	9	10	8.80689767382400	1.889535710246398E-004
4	7	8	9	13.2179692021618	3.690876617915702E-004
4	7	8	10	11.2641257027302	4.050545446737259E-003
4	7	9	10	12.6428458761529	5.513975290700210E-004
4	8	9	10	8.37598804938554	6.796109217578003E-003
5	6	7	8	4.06042083911347	2.180329353976359E-003
5	6	7	9	7.83099757990189	2.692142258893269E-003
5	6	7	10	9.05402538929006	3.550015893639125E-003
5	6	8	9	7.27974615527073	3.113214262685080E-003
5	6	8	10	5.69414983050595	3.918706084576646E-007
5	6	9	10	12.2554152412852	5.450069356297131E-003
5	7	8	9	5.93989559876388	2.025382326181307E-003
5	7	8	10	8.53452259729756	6.145224544579857E-007

...

Set of Orbitals				Associated energy	Associated correlation
5	7	9	10	10.9794368276030	3.807926121634084E-003
5	8	9	10	8.93960367894829	9.094450450198377E-007
6	7	8	9	8.67650379156473	5.929094284109192E-005
6	7	8	10	4.33925693471936	5.922790743586482E-004
6	7	9	10	7.88348279378305	9.345915444972138E-005
6	8	9	10	7.42137010850509	1.043263346220008E-003
7	8	9	10	6.25811520069360	8.890736284802188E-004
Ψ_G				1.40244543700138	3.741835421742382E-002
Selected set of orbitals				**Associated energy**	**Associated correlation**
3	5	6	7	3.62987422706307	2.499670282531226E-003

Observe in particular that the set of orbitals we selected following the energy criterion is not the one that leads to the most correlated wavefunction.

K=2 (TDHF)

The multiconfiguration Hartree-Fock method can be seen as a generalization of the Hartree-Fock method. In fact HF dynamics corresponds exactly to the multiconfiguration dynamics with $N = K$, thus $K = 2$ in our configuration. One of the main feature of the multiconfiguration methods is the correlation that vanishes by definition when the approximate wavefunction is a single Slater determinant. The density matrix turns to be the identity and the eigenvalues are then equals to one which corresponds to $\mathfrak{E} = 0$. In the sequel, we use the same initial data in order to solve the TDHF and the exact Schrödinger equation. That is, one select two orbitals, then construct one of the two possibles Slater determinants, they differs only by sign, and then consider it as the initial data for the Exact Schrödinger equation. First of all, one expect that the correlation remains zero for all time during the TDHF propagation. However, since a binary interaction (coupled) is considered in the exact Schrödinger equation, it is a simple exercise to see that immediately the exact dynamic broke the structure of the determinant. Thus, one expect that, the correlation becomes strictly positive immediately. This is confiremd by the next plot

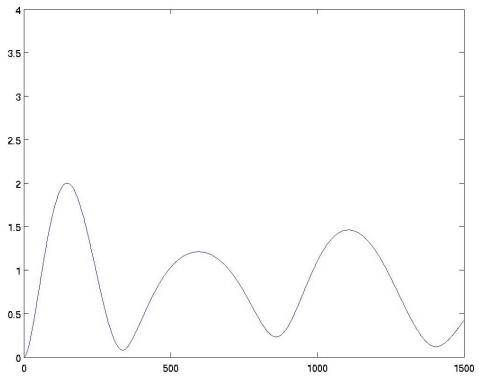

Fig4: TDHF vs Exact correlation

As mentioned above, one of the weakness of the scheme that we are using for the resolution of

the MCTDHF, is that it do not conserve the energy from the numerical analysis point of view.

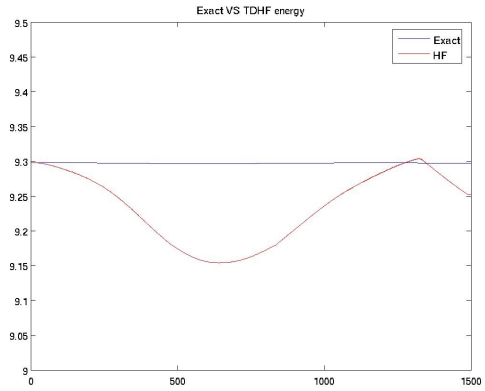

Fig5: TDHF vs Exact energy

The issue of the development of a scheme that conserves exactly the energy is in development. The next plots of the density matrix are of particular interest. In fact, running the exact Schrödinger and the MCTDHF (TDHF here) with the same initial data corresponding to the following density matrix gives

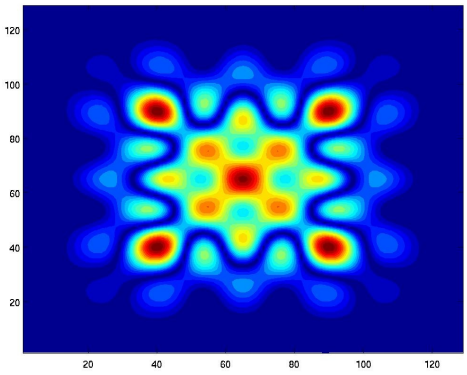

Fig6: $|[\Psi \otimes \Psi]_{:1}(t=0,x,y)|$

Again using a 65 grid points and a step of time of $\Delta t = 10^{-3}$. After 1500 iteration, we get

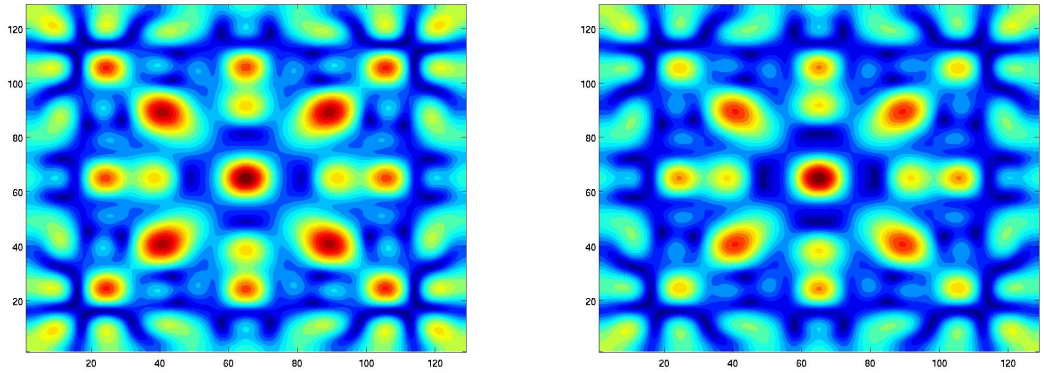

Fig7: Left: $|[\Psi E \otimes \Psi_E]_{:1}(t = 1.5s, x, y)|$. *Right:* $|[\Psi HF \otimes \Psi_{HF}]_{:1}(t = 1.5s, x, y)|$

Clearly, the two solutions are very similar. In particular, this shows the potential of the simpler case of the MCTDHF, that is the TDHF.

K=4

Now, we consider a 4 orbitals expansion instead of 2. One expect that the larger K, the better is the approximation in some sense. However up to now there is no rigorous argument that proves this seemingly obvious fact. One of the evident advantage of expanding the Multiconfiguration wavefunction is that from a purely mathematical point of view, one can compute the degree of correlation of the wavefunction.

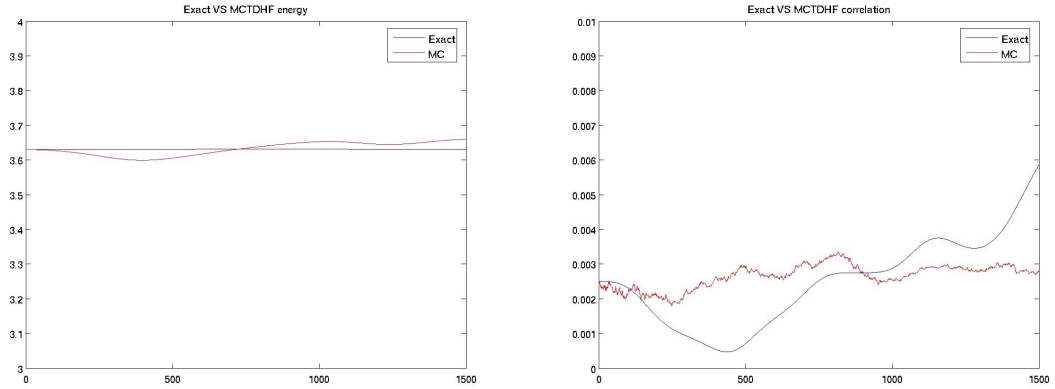

Fig7: Left: Exact VS MCTDHF energy. Right: Exact VS MCTDHF correlation

One observe that even with the non exact conservation of the energy of the MCTDHF, one see small oscillations around the exact one. However, the behavior of the correlation is not well approximated. In fact, one observe that the MCTDHF correlation stays somewhat closed, with small oscillations, to the exact one but up to a very small time, approximatively up to 10^{-1}. Again, we will present a plot of the initial density matrix and then the corresponding solutions respectively of the Exact Schrödinger and the MCTDHF. The initial data is the following one

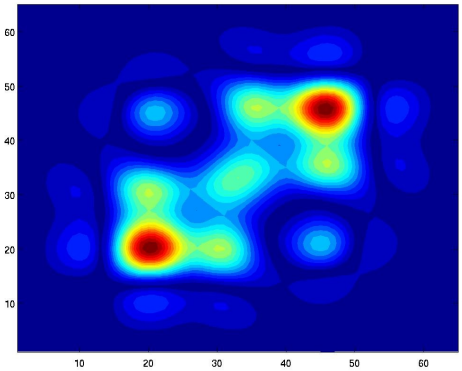

Fig6: $|[\Psi \otimes \Psi]_{:1}(t=0,x,y)|$

The associated solutions are

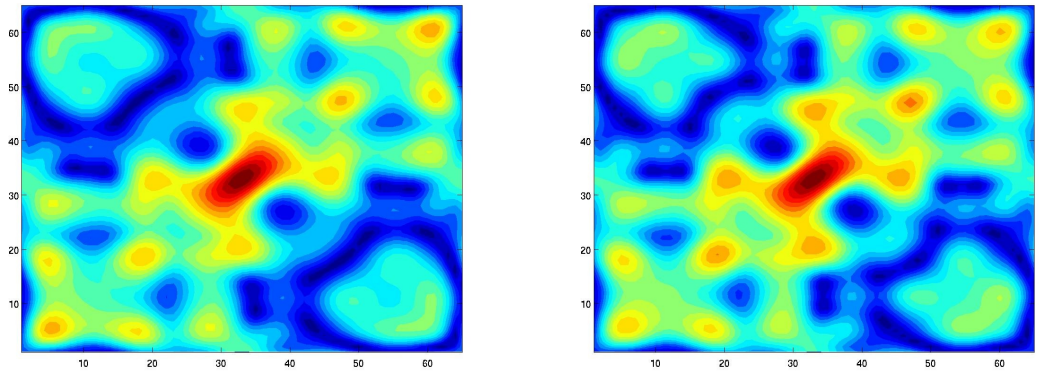

Fig7: Left: $|[\Psi E \otimes \Psi_E]_{:1}(t=1.5s,x,y)|$. *Right:* $|[\Psi HF \otimes \Psi_{HF}]_{:1}(t=1.5s,x,y)|$

In contrast with the the previous section, the second plot is obtained using the MCTDHF with 4 orbitals and again one observe that the solutions have a very closed structure.

K=6

In this section a 6 orbitals simulation is performed. However, in this case, one observe that the energy of the MCTDHf is far from being conserved as one can see in the next plots. This is due to the fact that the scheme is not designed to conserve the energy but also to the build-up of roundoff error since in this configuration, the computation is more heavier than in the TDHF or in the 4 orbitals case. Also, one observe that the correlation is far from being propagated in the same way by the exact and the approximated dynamics. Notice that in this case, the correlation is the same up to a very small time. This is an interesting issue to made clear in the future.

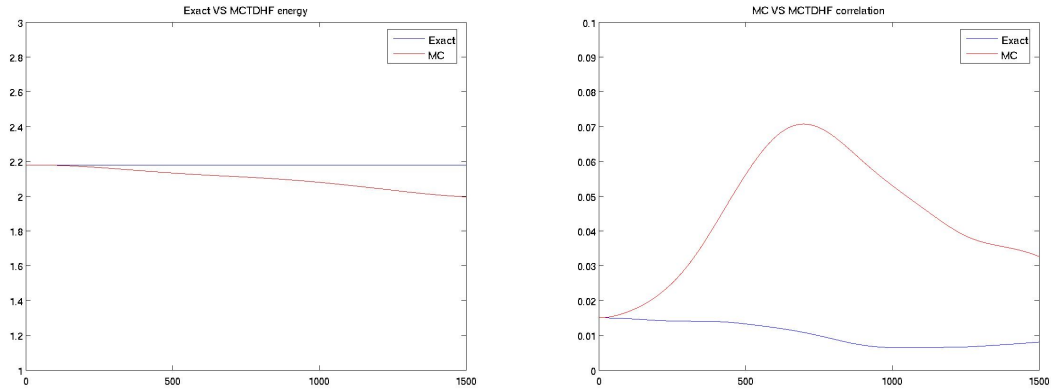

Fig7: Left: Exact VS MCTDHF energy. Right: Exact VS MCTDHF correlation

The initial data is then builded from 6 orbitals and looks as

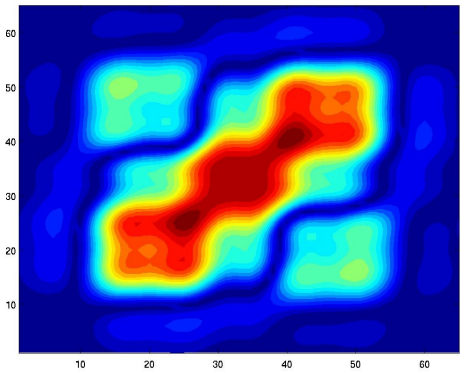

Fig6: $|[\Psi \otimes \Psi]_{:1}(t=0,x,y)|$

The associated solutions with the exact and the MCTDHf dynamics are

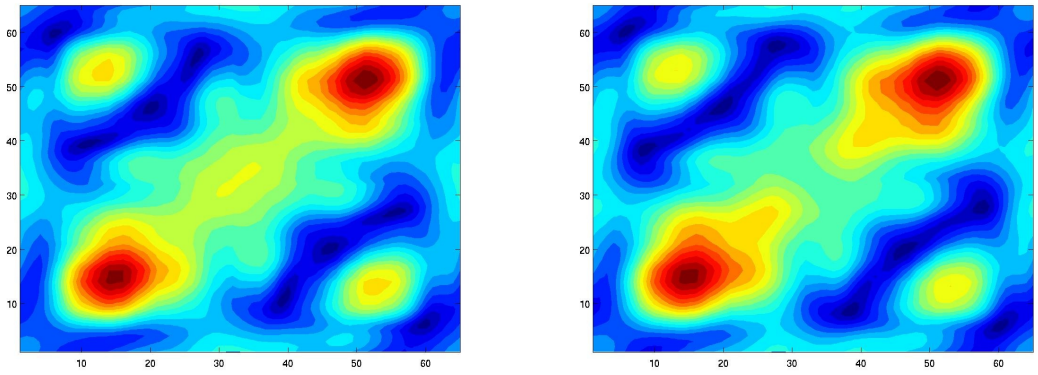

Fig7: Left: $|[\Psi E \otimes \Psi_E]_{:1}(t=1.5s,x,y)|$. *Right:* $|[\Psi HF \otimes \Psi_{HF}]_{:1}(t=1.5s,x,y)|$

Again, one observe, even with the non conservation of the energy, that the solutions has exactly

the same shape.

6.4 conclusion

We have done some very basic numerical simulations of the MCTDHF equations. Our methods are far from being optimal and we hope to obtain better and more accurate results in follow up numerical work. However, already these simple simulations allow interesting observations and an instructive illustrations of the MCTDHF hierarchy.

For example we observe that in all the situations we tested, we have never encountered a case where the density matrix $\mathbf{\Gamma}[C(t)]$ becomes degenerate. Our believe is that this is not due to the selection of the initial data we made, but to the sensible behavior of the coefficients since they change very slowly. We observed a variation of the "correlation" on the setting.

Finally, the basic simulations we presented show that the MCTDHF approximate in a good way the solution of the Schrödinger equation since the density matrices obtained from the exact equation and the approximated one are very similar, with the same quantitative structure. Moreover, we see that with appropriate schemes, the MCTDHF method superior to the simple TDHF.

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Chapter 7

Conclusion and Outlook

In this thesis we have presented the first results on the existence and uniqueness analysis of the MCTDHF equation with potentials as binary interactions being in a large functional class (containing the singular Coulomb potential) in a formulation of the equations particularly appropriate for our methods.

We have proven well-posedness in the Sobolev space H^1, the "energy space" and in L^2. These results trivially extend to analogous results for Sobolev spaces of higher regularity like H^2, which are useful for numerics.

The key assumption of "full rank" of the system's "density matrix" implies that we do not get global results. It is unclear if this is an intrinsic problem of the MCTDHF equations or if we just have not found the optimal methods for the analysis yet.

In order to prove global well-posedness we trade this "full rank condition" for the solution at time t for a sufficient condition on the energy of the initial data which has to be somewhat "close to the ground state". A close look on the proof shows that it is not obvious how to lift this condition. Also, the fact that the global well-posedness for TDHF follows so easily from our proof (since the density matrix is trivially of full rank for all times) indicates that the quite complicated MCTDHF system indeed has a complex structure that is very different from "simple" equations like Schrödinger-Poisson or TDHF. Eventually a geometric point of view is a key to such methods for global wellposedness.

Another important open question is the convergence of the solutions of MCTDHF to the solutions of the linear N particle Schrödinger for $K \longrightarrow \infty$. This "dogma" of quantum chemistry is still on the level of "folklore" from the point of view of rigorous mathematics. There is hope to achieve such results.

However, a rigorous derivation of MCTDHF as a "weak coupling limit" ($N \rightarrow \infty$ with a scaling factor of $1/N$ in front of the interaction potential following the concept of the derivation of the Schrödinger-Poisson system as laid out in [1] seems to be impossible.

We shall continue to work on these analytical questions, assisted by more and more profound numerical studies of the MCTHDF system.

The development of good numerical methods for MCTDHF is an art on it's own and we shall do this in close contact e.g. with A. Scrinzi, eventually cooperating to extend and improve the algorithms of his MCTDHF software package.

For example the question of boundary conditions on finite domains of simulation is highly nontrivial - we shall try to adopt "absorbing boundary layer" techniques that have been developped for

NLS.

In order to study more realistic models we are currently adopting the techniques of Cancèsand Lebris [2] for time-dependent Hartree-Fock with "moving nuclei", more precisely the 2 cases of TDHF coupled to classical dynamics for either one nucleus subject to a general space-time dependent electric field or TDHF coupled to several nuclei and a special space-time dependent electric field. Such results would be the basis for an "ab initio" quantum chemistry theory for the formation of molecules out of their components.

Another line of future work on MCTDHF is to invoke control theory, for example for models of laser interacting with molecules.

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Publications

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2. Global-in-time existence of solutions to the multiconfiguration time- -dependent
Hartree-Fock equations:
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Applied mathematics letters (2008), doi:10.1016/j.aml.2007.12.033

3. Analysis of the MCTDHF equations with Coulomb Interactions:
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4. L^2 Analysis of the MCTDHF equations with Coulomb Interactions:
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Scientific talks

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2. MCTDHF: The multiconfiguration time-dependent Hartree-Fock system:
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3. More About the MCTDHF and MCTDH:
Wissenschaftskolleg Differential Equations Seminar Vienna ĐAustria- Oct. 2006

4. On the MCTDHF approximation:
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Conferences

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