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2 Beyond the Runge-Gross Theorem

R. van Leeuwen

2.1 Introduction

The Runge-Gross theorem [Runge 1984] states that for a given initial state the time-dependent density is a unique functional of the external potential. Let us elaborate a bit further on this point. Suppose we could solve the time-dependent Schrödinger equation (TDSE) for a given many-body system, i.e., we specify an initial state $|\Psi_0\rangle$ at $t = t_0$ and evolve the wave function in time using the Hamiltonian $\hat{H}(t)$. Then, from the wave function, we can calculate the time-dependent density $n(\boldsymbol{r}, t)$. We can then ask the question whether exactly the same density $n(\boldsymbol{r}, t)$ can be reproduced by an external potential $v'_{\text{ext}}(\boldsymbol{r}, t)$ in a system with a different given initial state and a different two-particle interaction, and if so, whether this potential is unique (modulo a purely time-dependent function). The answer to this question is obviously of great importance for the construction of the time-dependent Kohn-Sham equations. The Kohn-Sham system has no two-particle interaction and differs in this respect from the fully interacting system. It has, in general, also a different initial state. This state is usually a Slater determinant rather than a fully interacting initial state. A time-dependent Kohn-Sham system therefore only exists if the question posed above is answered affirmatively. Note that this is a v-representability question: Is a density belonging to an interacting system also noninteracting v-representable? We will show in this chapter that, with some restrictions on the initial states and potentials, this question can indeed be answered affirmatively [van Leeuwen 1999, van Leeuwen 2001, Giuliani 2005]. We stress that we demonstrate here that the interacting-v-representable densities are also noninteracting-v-representable rather than aiming at characterizing the set of v-representable densities. The latter question has inspired much work in ground state density functional theory (for extensive discussion see [van Leeuwen 2003]) and has only been answered satisfactorily for quantum lattice systems [Chayes 1985].

2.2 The Extended Runge-Gross Theorem: Different Interactions and Initial States

We start by considering the Hamiltonian

$$\hat{H}(t) = \hat{T} + \hat{V}_{\text{ext}}(t) + \hat{V}_{\text{ee}} \,, \tag{2.1}$$

where $\hat{T}$ is the kinetic energy, $\hat{V}_{\text{ext}}(t)$ the (in general time-dependent) external potential, and $\hat{V}_{\text{ee}}$ the two-particle interaction. In second quantization the constituent terms are, as usual, written as

$$\hat{T} = -\frac{1}{2} \sum_{\sigma} \int \mathrm{d}^3 r \, \hat{\psi}_\sigma^\dagger(\boldsymbol{r}) \nabla^2 \hat{\psi}_\sigma(\boldsymbol{r}) \,, \tag{2.2a}$$

$$\hat{V}_{\text{ext}}(t) = \sum_{\sigma} \int \mathrm{d}^3 r \, v_{\text{ext}}(\boldsymbol{r}, t) \hat{\psi}_\sigma^\dagger(\boldsymbol{r}) \hat{\psi}_\sigma(\boldsymbol{r}) \,, \tag{2.2b}$$

$$\hat{V}_{\text{ee}} = \frac{1}{2} \sum_{\sigma\sigma'} \int \mathrm{d}^3 r \int \mathrm{d}^3 r' \, v_{\text{ee}}(|\boldsymbol{r} - \boldsymbol{r}'|) \hat{\psi}_\sigma^\dagger(\boldsymbol{r}) \hat{\psi}_{\sigma'}^\dagger(\boldsymbol{r}') \hat{\psi}_{\sigma'}(\boldsymbol{r}') \hat{\psi}_\sigma(\boldsymbol{r}) \,. \tag{2.2c}$$

where σ and σ' are spin variables. For the readers not used to second quantization we note that the first few basic steps in this chapter can also be derived in first quantization. For details we refer to [Giuliani 2005, Vignale 2004]. However, good understanding of second quantization is indispensable to understand the next chapter. Good introductions to second quantization are found in [Fetter 1971, Runge 1991].

The two-particle potential $v_{\text{ee}}(|\boldsymbol{r} - \boldsymbol{r}'|)$ in (2.2c) can be arbitrary, but will in practice almost always be equal to the repulsive Coulomb potential. We then consider some basic relations satisfied by the density and the current density. The time-dependent density is given as the expectation value of the density operator

$$\hat{n}(\boldsymbol{r}) = \sum_{\sigma} \hat{\psi}_\sigma^\dagger(\boldsymbol{r}) \hat{\psi}_\sigma(\boldsymbol{r}) \,, \tag{2.3}$$

with the time-dependent many-body wavefunction, $n(\boldsymbol{r}, t) = \langle \Psi(t)|\hat{n}(\boldsymbol{r})|\Psi(t)\rangle$. In the following we consider two continuity equations. If $|\Psi(t)\rangle$ is the state evolving from $|\Psi_0\rangle$ under the influence of Hamiltonian $\hat{H}(t)$ we have the usual continuity equation

$$\frac{\partial}{\partial t} n(\boldsymbol{r}, t) = -\mathrm{i}\langle \Psi(t)| \, [\hat{n}(\boldsymbol{r}), \hat{H}(t)] \, |\Psi(t)\rangle = -\nabla \cdot \boldsymbol{j}(\boldsymbol{r}, t) \,, \tag{2.4}$$

where the current operator is defined as

$$\hat{\boldsymbol{j}}(\boldsymbol{r}) = \frac{1}{2\mathrm{i}} \sum_{\sigma} \left\{ \hat{\psi}_\sigma^\dagger(\boldsymbol{r}) \nabla \hat{\psi}_\sigma(\boldsymbol{r}) - [\nabla \hat{\psi}_\sigma^\dagger(\boldsymbol{r})] \hat{\psi}_\sigma(\boldsymbol{r}) \right\} \,, \tag{2.5}$$

and has expectation value $\boldsymbol{j}(\boldsymbol{r}, t) = \langle \Psi(t)|\hat{\boldsymbol{j}}(\boldsymbol{r})|\Psi(t)\rangle$. This continuity equation expresses, in a local form, the conservation of particle number. Using Gauss' law the continuity equation says that the change of the number of particles within some volume can simply be measured by calculating the flux of the current through the surface of this volume.

As a next step, we can consider an analogous continuity equation for the current itself. We have

$$\frac{\partial}{\partial t}\boldsymbol{j}(\boldsymbol{r},t) = -\mathrm{i}\langle\Psi(t)|\,[\hat{\boldsymbol{j}}(\boldsymbol{r}),\hat{H}(t)]\,|\Psi(t)\rangle\ . \qquad (2.6)$$

If we work out the commutator in more detail, we find the expression [Martin 1959]

$$\frac{\partial}{\partial t}j_\alpha(\boldsymbol{r},t) = -n(\boldsymbol{r},t)\frac{\partial}{\partial r_\alpha}v_{\mathrm{ext}}(\boldsymbol{r},t) - \sum_\beta\frac{\partial}{\partial r_\beta}T_{\beta\alpha}(\boldsymbol{r},t) - V_{\mathrm{ee}\,\alpha}(\boldsymbol{r},t)\ . \quad (2.7)$$

Here we have defined the momentum-stress tensor $\hat{T}_{\beta\alpha}$ (part of the energy-momentum tensor)

$$\hat{T}_{\beta\alpha}(\boldsymbol{r}) = \frac{1}{2}\sum_\sigma\Big\{\frac{\partial}{\partial r_\beta}\hat{\psi}_\sigma^\dagger(\boldsymbol{r})\frac{\partial}{\partial r_\alpha}\hat{\psi}_\sigma(\boldsymbol{r}) + \frac{\partial}{\partial r_\alpha}\hat{\psi}_\sigma^\dagger(\boldsymbol{r})\frac{\partial}{\partial r_\beta}\hat{\psi}_\sigma(\boldsymbol{r})$$
$$- \frac{1}{2}\frac{\partial^2}{\partial r_\beta\partial r_\alpha}[\hat{\psi}_\sigma^\dagger(\boldsymbol{r})\hat{\psi}_\sigma(\boldsymbol{r})]\Big\}\ , \quad (2.8)$$

and the quantity $\hat{V}_{\mathrm{ee}\,\alpha}$ as

$$\hat{V}_{\mathrm{ee}\,\alpha}(\boldsymbol{r}) = \sum_{\sigma,\sigma'}\int\mathrm{d}^3r'\,\hat{\psi}_\sigma^\dagger(\boldsymbol{r})\hat{\psi}_{\sigma'}^\dagger(\boldsymbol{r}')\frac{\partial}{\partial r_\alpha}v_{\mathrm{ee}}(|\boldsymbol{r}-\boldsymbol{r}'|)\hat{\psi}_{\sigma'}(\boldsymbol{r}')\hat{\psi}_\sigma(\boldsymbol{r})\ . \qquad (2.9)$$

The expectation values that appear in (2.7) are defined as $T_{\beta\alpha}(\boldsymbol{r},t) = \langle\Psi(t)|\hat{T}_{\beta\alpha}(\boldsymbol{r})|\Psi(t)\rangle$ and $V_{\mathrm{ee}\,\alpha}(\boldsymbol{r},t) = \langle\Psi(t)|\hat{V}_{\mathrm{ee}\,\alpha}(\boldsymbol{r})|\Psi(t)\rangle$. The continuity equation (2.7) is a local quantum version of Newton's third law. Taking the divergence of (2.7) and using the continuity (2.4) we find

$$\frac{\partial^2}{\partial t^2}n(\boldsymbol{r},t) = \nabla\cdot[n(\boldsymbol{r},t)\nabla v_{\mathrm{ext}}(\boldsymbol{r},t)] + q(\boldsymbol{r},t)\ , \qquad (2.10)$$

with $\hat{q}$ and $q(\boldsymbol{r},t)$ being defined as

$$\hat{q}(\boldsymbol{r}) = \sum_{\alpha,\beta}\frac{\partial^2}{\partial r_\beta\partial r_\alpha}\hat{T}_{\beta\alpha}(\boldsymbol{r}) + \sum_\alpha\frac{\partial}{\partial r_\alpha}\hat{V}_{\mathrm{ee}\,\alpha}(\boldsymbol{r})\ , \qquad (2.11\mathrm{a})$$

$$q(\boldsymbol{r},t) = \langle\Psi(t)|\hat{q}(\boldsymbol{r})|\Psi(t)\rangle\ . \qquad (2.11\mathrm{b})$$

Equation (2.10) will play a central role in our discussion of the relation between the density and the potential. This is because it represents an equation which directly relates the external potential and the electron density. From (2.10) we further see that $q(\boldsymbol{r},t)$ decays exponentially at infinity when $n(\boldsymbol{r},t)$ does, unless $v_{\mathrm{ext}}(\boldsymbol{r},t)$ grows exponentially at infinity. In the following we will, however, only consider finite systems with external potentials that are

bounded at infinity (for a discussion of the set of allowed external potentials in ground state DFT we refer to [Lieb 1983, van Leeuwen 2003]).

Let us now assume that we have solved the time-dependent Schrödinger equation for the many-body system described by the Hamiltonian $\hat{H}(t)$ of (2.1) and initial state $|\Psi_0\rangle$ at $t = t_0$. We have thus obtained a many-body wavefunction $|\Psi(t)\rangle$ and density $n(\boldsymbol{r}, t)$. We further assume that $n(\boldsymbol{r}, t)$ is analytic at $t = t_0$. For our system, (2.10) is satisfied. We now consider a second system with Hamiltonian

$$\hat{H}'(t) = \hat{T} + \hat{V}'_{\text{ext}}(t) + \hat{V}'_{\text{ee}} \; . \tag{2.12}$$

The terms $\hat{V}'_{\text{ext}}(t)$ and $\hat{V}'_{\text{ee}}$ represent again the one- and two-body potentials. We denote the initial state by $|\Psi'_0\rangle$ at $t = t_0$ and the time-evolved state by $|\Psi'(t)\rangle$. The form of $\hat{V}'_{\text{ee}}$ is assumed to be such that its expectation value and its derivatives are finite. For the system described by the Hamiltonian $\hat{H}'$ we have an equation analogous to (2.10).

$$\frac{\partial^2}{\partial t^2} n'(\boldsymbol{r}, t) = \nabla \cdot [n'(\boldsymbol{r}, t) \nabla v'_{\text{ext}}(\boldsymbol{r}, t)] + q'(\boldsymbol{r}, t) \; , \tag{2.13}$$

where $q'(\boldsymbol{r}, t)$ is the expectation value

$$q'(\boldsymbol{r}, t) = \langle \Psi'(t) | \hat{q}'(\boldsymbol{r}) | \Psi'(t) \rangle \; , \tag{2.14}$$

for which we defined

$$\hat{q}' = \sum_{\beta,\alpha} \frac{\partial^2}{\partial r_\beta \partial r_\alpha} \hat{T}_{\beta\alpha}(\boldsymbol{r}) + \sum_k \frac{\partial}{\partial r_\alpha} \hat{V}'_{\text{ee}\,\alpha}(\boldsymbol{r}) \; . \tag{2.15}$$

Our goal is now to choose v'_{ext} in (2.13) so that $n'(\boldsymbol{r}, t) = n(\boldsymbol{r}, t)$. We will do this by constructing v'_{ext} in such a way that for the k-th derivatives of the density at $t = t_0$ we have $\frac{\partial^k}{\partial t^k} n'(\boldsymbol{r}, t)|_{t=t_0} = \frac{\partial^k}{\partial t^k} n(\boldsymbol{r}, t)|_{t=t_0}$. First we need to discuss some initial conditions. As a necessary condition for the potential v'_{ext} to exist, we have to require that the initial states $|\Psi_0\rangle$ and $|\Psi'_0\rangle$ yield the same initial density, i.e.,

$$n'(\boldsymbol{r}, t_0) = \langle \Psi'_0 | \hat{n}(\boldsymbol{r}) | \Psi'_0 \rangle = \langle \Psi_0 | \hat{n}(\boldsymbol{r}) | \Psi_0 \rangle = n(\boldsymbol{r}, t_0) \; . \tag{2.16}$$

We now note that the basic equation (2.10) is a second order differential equation in time for $n(\boldsymbol{r}, t)$. This means, as we will see soon, that we still need as additional requirement that $\frac{\partial}{\partial t} n'(\boldsymbol{r}, t) = \frac{\partial}{\partial t} n(\boldsymbol{r}, t)$ at $t = t_0$. With the help of the continuity equation (2.4) this yields the condition

$$\frac{\partial}{\partial t} n'(\boldsymbol{r}, t)\Big|_{t=t_0} = \langle \Psi'_0 | \nabla \cdot \hat{\boldsymbol{j}}(\boldsymbol{r}) | \Psi'_0 \rangle = \langle \Psi_0 | \nabla \cdot \hat{\boldsymbol{j}}(\boldsymbol{r}) | \Psi_0 \rangle = \frac{\partial}{\partial t} n(\boldsymbol{r}, t)\Big|_{t=t_0} \; . \tag{2.17}$$

This constraint also implies the weaker requirement that the initial state $|\Psi'_0\rangle$ must be chosen such that the initial momenta $\boldsymbol{P}(t_0)$ of both systems are the

same. This follows directly from the fact that the momentum of the system is given by

$$\boldsymbol{P}(t) = \int \mathrm{d}^3 r \, \boldsymbol{j}(\boldsymbol{r}, t) = \int \mathrm{d}^3 r \, \boldsymbol{r} \, \frac{\partial}{\partial t} n(\boldsymbol{r}, t) \,. \qquad (2.18)$$

The equality of the last two terms in this equation follows directly from the continuity equation (2.4) and the fact that we are dealing with finite systems for which, barring pathological examples [van Leeuwen 2001, Maitra 2001], currents and densities are zero at infinity. For notational convenience we first introduce the following notation for the k-th time-derivative at $t = t_0$ of a function f:

$$f^{(k)}(\boldsymbol{r}) = \left.\frac{\partial^k}{\partial t^k} f(\boldsymbol{r}, t)\right|_{t=t_0} \,. \qquad (2.19)$$

Then our goal is to choose v'_{ext} in such a way that $n'^{(k)} = n^{(k)}$ for all k. Let us see how we can use (2.13) to do this. If we first evaluate (2.13) at $t = t_0$ we obtain, using the notation of (2.19), the expression

$$n'^{(2)}(\boldsymbol{r}) = \nabla \cdot [n'^{(0)}(\boldsymbol{r})\nabla v'^{(0)}_{\mathrm{ext}}(\boldsymbol{r})] + q'^{(0)}(\boldsymbol{r}) \,. \qquad (2.20)$$

Since we want that $n'^{(2)} = n^{(2)}$ and have chosen the initial state $|\Psi'_0\rangle$ in such a way that $n'^{(0)} = n^{(0)}$ we obtain the following determining equation for $v'^{(0)}_{\mathrm{ext}}$:

$$\nabla \cdot [n^{(0)}(\boldsymbol{r})\nabla v'^{(0)}_{\mathrm{ext}}(\boldsymbol{r})] = n^{(2)}(\boldsymbol{r}) - q'^{(0)}(\boldsymbol{r}) \,. \qquad (2.21)$$

The right hand side is determined since $n^{(0)}$ and $n^{(2)}$ are given and $q'^{(0)}$ is calculated from the given initial state $|\Psi'_0\rangle$ as $q'^{(0)}(\boldsymbol{r}) = \langle\Psi'_0|\hat{q}'(\boldsymbol{r})|\Psi'_0\rangle$. Equation (2.21) is of Sturm-Liouville type and has a unique solution for v'_0 provided we specify a boundary condition. We will specify the boundary condition that $v'^{(0)}_{\mathrm{ext}}(\boldsymbol{r}) \to 0$ for $\boldsymbol{r} \to \infty$. With this boundary condition we also fix the gauge of the potential. Having obtained $v'^{(0)}_{\mathrm{ext}}$ let us now go on to determine $v'^{(1)}_{\mathrm{ext}}$. To do this we differentiate (2.13) with respect to time and evaluate the resulting expression in $t = t_0$. Then we obtain the expression:

$$n'^{(3)}(\boldsymbol{r}) = \nabla \cdot [n'^{(0)}(\boldsymbol{r})\nabla v'^{(1)}_{\mathrm{ext}}(\boldsymbol{r})] + \nabla \cdot [n'^{(1)}(\boldsymbol{r})\nabla v'^{(0)}_{\mathrm{ext}}(\boldsymbol{r})] + q'^{(1)}(\boldsymbol{r}) \,. \quad (2.22)$$

Since we want to determine $v'^{(1)}$ such that $n'^{(3)} = n^{(3)}$ and the conditions on the initial states are such that $n'^{(0)} = n^{(0)}$ and $n'^{(1)} = n^{(1)}$, we obtain the following equation for $v'^{(1)}_{\mathrm{ext}}$:

$$\nabla \cdot [n^{(0)}(\boldsymbol{r})\nabla v'^{(1)}_{\mathrm{ext}}(\boldsymbol{r})] = n^{(3)}(\boldsymbol{r}) - q'^{(1)}(\boldsymbol{r}) - \nabla \cdot [n^{(1)}(\boldsymbol{r})\nabla v'^{(0)}_{\mathrm{ext}}(\boldsymbol{r})] \,. \quad (2.23)$$

Now all quantities on the right hand side of (2.23) are known. The initial potential $v'^{(0)}_{\mathrm{ext}}$ was already determined from (2.21) whereas the quantity $q'^{(1)}$ can be calculated from

$$q'^{(1)}(\boldsymbol{r}) = \left.\frac{\partial}{\partial t} q'(\boldsymbol{r}, t)\right|_{t=t_0} = -\mathrm{i}\langle\Psi'_0| [\hat{q}'(\boldsymbol{r}), \hat{H}'(t_0)] |\Psi'_0\rangle \,. \qquad (2.24)$$

From this expression we see that $q'^{(1)}$ can be calculated from the knowledge of the initial state and the initial potential $v_{\text{ext}}'^{(0)}$ which occurs in $\hat{H}'(t_0)$. Therefore, (2.23) uniquely determines $v_{\text{ext}}'^{(1)}$ (again with boundary conditions $v_{\text{ext}}'^{(1)} \to 0$ for $\boldsymbol{r} \to \infty$). We note that in order to obtain (2.23) from (2.22) we indeed needed both conditions of (2.16) and (2.17). It is now clear how our procedure can be extended. If we take the k-th time-derivative of (2.13) we obtain the expression

$$n'^{(k+2)}(\boldsymbol{r}) = q'^{(k)}(\boldsymbol{r}) + \sum_{l=0}^{k} \binom{k}{l} \nabla \cdot [n'^{(k-l)}(\boldsymbol{r})\nabla v_{\text{ext}}'^{(l)}(\boldsymbol{r})] \,. \qquad (2.25)$$

Demanding that $n'^{(k)} = n^{(k)}$ then yields

$$\nabla \cdot [n^{(0)}(\boldsymbol{r})\nabla v_{\text{ext}}'^{(k)}(\boldsymbol{r})] = n^{(k+2)}(\boldsymbol{r}) - q'^{(k)}(\boldsymbol{r}) - \sum_{l=0}^{k-1} \binom{k}{l} \nabla \cdot [n^{(k-l)}(\boldsymbol{r})\nabla v_{\text{ext}}'^{(l)}(\boldsymbol{r})] \,. \qquad (2.26)$$

The right hand side of this equation is completely determined since it only involves the potentials $v_{\text{ext}}'^{(l)}$ for $l = 1 \ldots k-1$ which were already determined. Similarly the quantities $q'^{(k)}$ can be calculated from multiple commutators of the operator $\hat{q}'$ and time-derivatives of the Hamiltonian $\hat{H}'(t_0)$ up to order $k-1$ and therefore only involves knowledge of the initial state and $v_{\text{ext}}'^{(l)}$ for $l = 1 \ldots k-1$. We can therefore uniquely determine all functions $v_{\text{ext}}'^{(k)}$ from (2.26) (again taking into account the boundary conditions) and construct the potential $v_{\text{ext}}'(\boldsymbol{r},t)$ from its Taylor series as

$$v_{\text{ext}}'(\boldsymbol{r},t) = \sum_{k=0}^{\infty} \frac{1}{k!} \, v_{\text{ext}}'^{(k)}(\boldsymbol{r}) \, (t - t_0)^k \,. \qquad (2.27)$$

This determines $v_{\text{ext}}'(\boldsymbol{r},t)$ completely within the convergence radius of the Taylor expansion. There is, of course, the possibility that the convergence radius is zero. However, this would mean that $v_{\text{ext}}'(\boldsymbol{r},t)$ and hence $n(\boldsymbol{r},t)$ and $v_{\text{ext}}(\boldsymbol{r},t)$ are nonanalytic at $t = t_0$. Since the density of our reference system was supposed to be analytic we can disregard this possibility. If the convergence radius is non-zero but finite, we can propagate $|\Psi_0'\rangle$ to $|\Psi'(t_1)\rangle$ until a finite time $t_1 > t_0$ within the convergence radius and repeat the whole procedure above from $t = t_1$ by regarding $|\Psi'(t_1)\rangle$ as the initial state. This amounts to analytic continuation along the whole real time-axis and the complete determination of $v_{\text{ext}}'(\boldsymbol{r},t)$ at all times. This completes the constructive proof of $v_{\text{ext}}'(\boldsymbol{r},t)$.

Let us now summarize what we proved. We specify a given density $n(\boldsymbol{r},t)$ obtained from a many-particle system with Hamiltonian $\hat{H}$ and initial state $|\Psi_0\rangle$. If one chooses an initial state $|\Psi_0'\rangle$ of a second many-particle system with two-particle interaction $\hat{V}_{\text{ee}}'$ in such a way that it yields the correct

initial density and initial time-derivative of the density, then, for this system, there is a unique external potential $v'_{\text{ext}}(\boldsymbol{r}, t)$ [determined up to a purely time-dependent function $c(t)$] that reproduces the given density $n(\boldsymbol{r}, t)$.

Let us now specify some special cases. If we take $\hat{V}'_{\text{ee}} = 0$ we can conclude that, for a given initial state $|\Psi'_0\rangle = |\Phi_0\rangle$ with the correct initial density and initial time derivative of the density, there is a unique potential $v_{\text{KS}}(\boldsymbol{r}, t)$ [modulo $c(t)$] for a noninteracting system that produces the given density $n(\boldsymbol{r}, t)$ at all times. This solves the noninteracting v-representability problem, provided we can find an initial state with the required properties. If the many-body system described by the Hamiltonian $\hat{H}$ is stationary for times $t < t_0$, the initial state $|\Psi_0\rangle$ at t_0 leads to a density with zero time-derivative at $t = t_0$. In that case, a noninteracting state with the required initial density and initial time-derivative of the density (namely zero) can be obtained via the so-called Harriman construction [Harriman 1981, Lieb 1983]. Therefore a Kohn-Sham potential always exists for this kind of switch-on processes. The additional question whether this initial state can be chosen as a ground state of a noninteracting system is equivalent to the currently unresolved noninteracting v-representability question for stationary systems [Kohn 1983a, Ullrich 2002a, Dreizler 1990] (for an extensive discussion see [van Leeuwen 2003]).

We now take $\hat{V}'_{\text{ee}} = \hat{V}_{\text{ee}}$. We therefore consider two many-body systems with the same two-particle interaction. Our proof then implies that for a given v-representable density $n(\boldsymbol{r}, t)$ that corresponds to an initial state $|\Psi_0\rangle$ and potential $v_{\text{ext}}(\boldsymbol{r}, t)$, and for a given initial state $|\Psi'_0\rangle$ with the same initial density and initial time derivative of the density, we find that there is a unique external potential $v'_{\text{ext}}(\boldsymbol{r}, t)$ [modulo $c(t)$] that yields this given density $n(\boldsymbol{r}, t)$. The case $|\Psi_0\rangle = |\Psi'_0\rangle$ (in which the constraints on the initial state $|\Psi'_0\rangle$ are trivially satisfied) corresponds to the well-known Runge-Gross theorem. Our results in this section therefore provide an extension of this important theorem. As a final note we mention that the proof discussed here has recently been extended in an elegant way by Vignale [Vignale 2004] to time-dependent current-density functional theory. In that work it is shown that currents from an interacting system with some vector potential are also representable by a vector potential in a noninteracting system. This is, however, not true anymore if one considers scalar potentials. Interacting-v-representable currents are in general not noninteracting-v-representable [D'Agosta 2005a].

2.3 Invertibility of the Linear Density Response Function

In this section we will address the question if we can recover the potential variation $\delta v_{\text{ext}}(\boldsymbol{r}, t)$ from a given density variation $\delta n(\boldsymbol{r}, t)$ that was produced by it. There is, of course, an obvious non-uniqueness since both $\delta v_{\text{ext}}(\boldsymbol{r}, t)$ and $\delta v_{\text{ext}}(\boldsymbol{r}, t) + c(t)$, where $c(t)$ is an arbitrary time-dependent function, produce

the same density variation. However, this is simply a gauge of the potential and is easily taken care of. Thus, by an inverse we will always mean an inverse modulo a purely time-dependent function $c(t)$ and by different potentials we will always mean that they differ more than a gauge $c(t)$.

From the work of Mearns and Kohn [Mearns 1987] we know that different potentials can yield the same density variations. However, in their examples these potentials are always potentials that exist at all times, i.e. there is no t_0 such that $\delta v_{\text{ext}} = 0$ for times $t < t_0$. On the other hand, we know from the Runge-Gross proof that a potential $\delta v_{\text{ext}}(\boldsymbol{r}, t)$ (not purely time-dependent) that is switched on at $t = t_0$ and is analytic at t_0 always causes a nonzero density variation $\delta n(\boldsymbol{r}, t)$. In this proof, the first nonvanishing time-derivative of δn at t_0 is found to be linear in the corresponding derivative of δv_{ext} and therefore the linear response function is invertible. Note that this conclusion holds even for an arbitrary initial state. The conclusion is therefore true for linear response to an already time-dependent system for which the linear response function depends on both t and t' separately, rather than on the time-difference $t - t'$. In the following we give an explicit proof for the invertibility of the linear response function for which the system is initially in its ground state. However, we will relax the condition that δv_{ext} is an analytic function in time, and we therefore allow for a larger class of external potentials than assumed in the Runge-Gross theorem. For clarification we further mention that it is sometimes assumed that the Dyson-type response equations of TDDFT are based on an adiabatic switch-on of the potential at all times. This is, however, not the case The response functions can simply be derived by first order perturbation theory on the TDSE using a sudden switch-on of the external time-dependent potential [Fetter 1971]. The typical imaginary infinitesimals that occur in the denominator of the response functions result from the Fourier-representation of the causal Heaviside function (written as a complex contour integral) in the retarded density response function rather than from an adiabatically switched-on potential. The linear response equations of TDDFT are therefore in perfect agreement with a sudden switch-on of the potential.

We consider a many-body system in its ground state. At $t = 0$ (since the system is initially described by a time-independent Hamiltonian we can, without loss of generality, put the initial time $t_0 = 0$) we switch on an external field $\delta v_{\text{ext}}(\boldsymbol{r}, t)$ which causes a density response δn. We want to show that the linear response function is invertible for these switch-on processes. From simple first order perturbation theory on the TDSE we know that the linear density response is given by [Fetter 1971]

$$\delta n(\boldsymbol{r}_1, t_1) = \int_0^{t_1} \mathrm{d}t_2 \int \mathrm{d}^3 r_2 \, \chi_R(\boldsymbol{r}_1 t_1, \boldsymbol{r}_2 t_2) \delta v_{\text{ext}}(\boldsymbol{r}_2, t_2) \,, \qquad (2.28)$$

where

$$\chi_R(\boldsymbol{r}_1 t_1, \boldsymbol{r}_2 t_2) = -\mathrm{i}\,\theta(t_1 - t_2)\langle \Psi_0| \left[\Delta \hat{n}_H(\boldsymbol{r}_1, t_1), \Delta \hat{n}_H(\boldsymbol{r}_2, t_2) \right] |\Psi_0\rangle \,, \quad (2.29)$$

is the retarded density response function. Note that here, instead of the density operator $\hat{n}_H$ (in the Heisenberg picture with respect to the ground state Hamiltonian $\hat{H}$), we prefer to use the density fluctuation operator $\Delta\hat{n}_H = \hat{n}_H - \langle\hat{n}_H\rangle$ in the response function, where we use that the commutator of the density operators is equal to the commutator of the density fluctuation operators. Now we go over to a Lehmann representation of the response function and we insert a complete set of eigenstates of $\hat{H}$:

$$\delta n(\boldsymbol{r}_1, t_1) = \mathrm{i} \sum_n \int_0^{t_1} \mathrm{d}t_2 \int \mathrm{d}^3 r_2\, \mathrm{e}^{\mathrm{i}\Omega_n(t_1 - t_2)} f_n^*(\boldsymbol{r}_1) f_n(\boldsymbol{r}_2) \delta v_{\mathrm{ext}}(\boldsymbol{r}_2, t_2) + \mathrm{c.c.}\,,$$

(2.30)

where $\Omega_n = E_n - E_{\mathrm{GS}} > 0$ are the excitation energies of the unperturbed system (we assume the ground state to be nondegenerate) and the functions f_n are defined as

$$f_n(\boldsymbol{r}) = \langle \Psi_{\mathrm{GS}} | \Delta\hat{n}(\boldsymbol{r}) | \Psi_n \rangle\,.$$

(2.31)

The density response can then be rewritten as

$$\delta n(\boldsymbol{r}_1, t_1) = \mathrm{i} \sum_n f_n^*(\boldsymbol{r}_1) \int_0^{t_1} \mathrm{d}t_2\, a_n(t_2) \mathrm{e}^{\mathrm{i}\Omega_n(t_1 - t_2)} + \mathrm{c.c.}\,,$$

(2.32)

where we defined

$$a_n(t) = \int \mathrm{d}^3 r\, f_n(\boldsymbol{r}) \delta v_{\mathrm{ext}}(\boldsymbol{r}, t)\,.$$

(2.33)

Now note that the time integral in (2.32) has the form of a convolution. This means that we can simplify this equation using Laplace transforms. The Laplace transform and its deconvolution property are given by

$$\hat{\mathcal{L}}f(s) = \int_0^\infty \mathrm{d}t\, \mathrm{e}^{-st} f(t)\,,$$

(2.34a)

$$\hat{\mathcal{L}}(f * g)(s) = \hat{\mathcal{L}}f(s)\hat{\mathcal{L}}g(s)\,.$$

(2.34b)

where the convolution product is defined as

$$(f * g)(t) = \int_0^t \mathrm{d}\tau\, f(\tau) g(t - \tau)\,.$$

(2.35)

If we now take the Laplace transform of δn in (2.32) we obtain the equation:

$$\hat{\mathcal{L}}(\delta n)(\boldsymbol{r}_1, s) = \mathrm{i} \sum_n f_n^*(\boldsymbol{r}_1) \frac{1}{s - \mathrm{i}\Omega_n} \hat{\mathcal{L}} a_n(s) + \mathrm{c.c.}$$

(2.36)

If we multiply both sides with the Laplace transform $\hat{\mathcal{L}}(\delta v_{\mathrm{ext}})$ of δv_{ext} and integrate over $\boldsymbol{r}_1$ we obtain

$$\int d^3 r_1 \, \hat{\mathcal{L}}(\delta v_{\text{ext}})(\boldsymbol{r}_1, s) \hat{\mathcal{L}}(\delta n)(\boldsymbol{r}_1, s) = i \sum_n \frac{1}{s - i\Omega_n} |\hat{\mathcal{L}} a_n(s)|^2 + \text{c.c.}$$

$$= -2 \sum_n \frac{\Omega_n}{s^2 + \Omega_n^2} |\hat{\mathcal{L}} a_n(s)|^2 . \quad (2.37)$$

This is the basic relation that we use to prove invertibility. If we assume that $\delta n = 0$ then also $\hat{\mathcal{L}}(\delta n) = 0$ and we obtain

$$0 = \sum_n \frac{\Omega_n}{s^2 + \Omega_n^2} |\hat{\mathcal{L}} a_n(s)|^2 . \quad (2.38)$$

However, since each prefactor of $|\hat{\mathcal{L}} a_n|^2$ in the summation is positive the sum can only be zero if $\hat{\mathcal{L}} a_n = 0$ for all n. This in its turn implies that $a_n(t)$ must be zero for all n. This means also that

$$\int d^3 r \, \Delta \hat{n}(r) \delta v_{\text{ext}}(\boldsymbol{r}, t) |\Psi_0\rangle = \sum_n |\Psi_n\rangle \int d^3 r \, \langle \Psi_n | \Delta \hat{n}(\boldsymbol{r}) | \Psi_0 \rangle \delta v_{\text{ext}}(\boldsymbol{r}, t)$$

$$= \sum_n a_n(t) |\Psi_n\rangle = 0 . \quad (2.39)$$

Note that $a_0(t)$ is automatically zero since obviously $\langle \Psi_{\text{GS}} | \Delta \hat{n}(x) | \Psi_{\text{GS}} \rangle = 0$. If we write out the above equation in first quantization again we have

$$\sum_{k=1}^{N} \Delta v_{\text{ext}}(\boldsymbol{r}_k, t) |\Psi_{\text{GS}}\rangle = 0 , \quad (2.40)$$

where N is the number of electrons in the system and $\Delta v_{\text{ext}}(\boldsymbol{r}, t)$ is defined as

$$\Delta v_{\text{ext}}(\boldsymbol{r}, t) = \delta v_{\text{ext}}(\boldsymbol{r}, t) - \frac{1}{N} \int d^3 r \, n_{\text{GS}}(\boldsymbol{r}) \delta v_{\text{ext}}(\boldsymbol{r}, t) , \quad (2.41)$$

where n_{GS} is the density of the unperturbed system. Now (2.40) immediately implies that $\Delta v_{\text{ext}} = 0$ and, since the second term on the right hand side of (2.41) is a purely time-dependent function, we obtain

$$\delta v_{\text{ext}}(\boldsymbol{r}, t) = c(t) . \quad (2.42)$$

We have therefore proven that only purely time-dependent potentials yield zero density response. In other words, the response function is, modulo a trivial gauge, invertible for switch-on processes. Note that the only restriction we put on the potential $\delta v_{\text{ext}}(\boldsymbol{r}, t)$ is that it is Laplace-transformable. This is a much weaker restriction on the potential than the constraint that it be an analytic function at $t = t_0$, as required in the Runge-Gross proof. One should, however, be careful with what one means with an inverse response function. The response function defines a mapping $\chi : \delta \mathcal{V}_{\text{ext}} \rightarrow \delta \mathcal{N}$ from the set of potential variations from a nondegenerate ground state, which we call

$\delta\mathcal{V}_{\text{ext}}$, to the set of first order density variations $\delta\mathcal{N}$ that are reproduced by it. We have shown that the inverse $\chi^{-1} : \delta\mathcal{N} \to \delta\mathcal{V}_{\text{ext}}$ is well-defined modulo a purely time-dependent function. However, there are density variations that can never be produced by a finite potential variation and are therefore not in the set $\delta\mathcal{N}$. An example of such a density variation is one which is identically zero on some finite volume.

Another consequence of the above analysis is the following. Suppose the linear response kernel has eigenfunctions, i.e. there is a λ such that

$$\int \mathrm{d}t_2 \int \mathrm{d}r_2 \, \chi_R(\boldsymbol{r}_1 t_1, \boldsymbol{r}_2 t_2)\zeta(\boldsymbol{r}_2, t_2) = \lambda\zeta(\boldsymbol{r}_1, t_1) \, . \tag{2.43}$$

Laplace transforming this equation yields

$$\int \mathrm{d}^3 r_2 \, \Xi(\boldsymbol{r}_1, \boldsymbol{r}_2, s)\hat{\mathcal{L}}\zeta(\boldsymbol{r}_2, s) = \lambda\hat{\mathcal{L}}\zeta(\boldsymbol{r}_1, s) \, , \tag{2.44}$$

where Ξ is the Laplace transform of χ explicitly given by

$$\Xi(\boldsymbol{r}_1, \boldsymbol{r}_2, s) = \mathrm{i}\sum_n \frac{f_n^*(\boldsymbol{r}_1)f_n(\boldsymbol{r}_2)}{s - \mathrm{i}\Omega_n} + \text{c.c.} \tag{2.45}$$

Since Ξ is a real Hermitian operator, its eigenvalues λ are real and its eigenfunctions $\hat{\mathcal{L}}\zeta$ can be chosen to be real. Then ζ is real as well and (2.37) implies (if we take $\delta v_{\text{ext}} = \zeta$ and $\delta n = \lambda\zeta$)

$$\lambda \int \mathrm{d}^3 r \, [\hat{\mathcal{L}}\zeta(\boldsymbol{r}, s)]^2 < 0 \, , \tag{2.46}$$

which implies $\lambda < 0$. We have therefore proven that if there are density variations that are proportional to the applied potential, then this constant of proportionality is negative. In other words, the eigenvalues of the density response function are negative. In this derivation we made again explicit use of Laplace transforms and therefore of the condition that $\zeta = 0$ for $t < 0$. The work of Mearns and Kohn shows that positive eigenvalues are possible when this restriction is not made. The same is true when one considers response functions for excited states [Gaudoin 2004]. We finally note that similar results are readily obtained for the static density response function [van Leeuwen 2003] in which case the negative eigenvalues of the response function are an immediate consequence of the Hohenberg-Kohn theorem.

Let us now see what our result implies. We considered the density $n[v_{\text{ext}}]$ as a functional of v_{ext} and established that the response kernel $\chi[v_{\text{GS}}] = \delta n/\delta v_{\text{ext}}[v_{\text{GS}}]$ is invertible where v_{GS} is the potential in the ground state and that $\delta n/\delta v_{\text{ext}}[v_{\text{GS}}] < 0$ in the sense that its eigenvalues are all negative definite. We can now apply a fundamental theorem of calculus, the *inverse function theorem*. For functions of real numbers the theorem states that if a continuous function $y(x)$ is differentiable at x_0 and if $\mathrm{d}y/\mathrm{d}x(x_0) \neq 0$ then

locally there exists an inverse $x(y)$ for y close enough to y_0, where $y(x_0) = y_0$. The theorem can be extended to functionals on function spaces (to be precise Banach spaces, for details see [Choquet-Bruhat 1991]). For our case, this theorem implies that if the functional $n[v_{\text{ext}}]$ is differentiable at the ground state potential v_{GS} and the derivative $\chi[v_{\text{GS}}] = \delta n/\delta v_{\text{ext}}[v_{\text{GS}}]$ is an invertible kernel then for potentials v_{ext} close enough to v_{GS} (in Banach norm sense) the inverse map $v_{\text{ext}}[n]$ exists. Since we have shown that the linear response function $\chi[v_{\text{GS}}]$ is invertible this then proves the Runge-Gross theorem for Laplace transformable switch-on potentials for systems initially in the ground state.

2.4 Consequences of v-Representability for the Quantum Mechanical Action

The role that is played by the energy functional in stationary density-functional theory is played by the action functional in time-dependent density functional theory. The correct form of the action appears naturally within the framework of Keldysh theory and is discussed in detail in Chap. 3. However, historically the first action within time-dependent density-functional theory context was defined by Peuckert [Peuckert 1978] (who already made a connection to Keldysh theory) and later in the Runge-Gross paper [Runge 1984]. However, as was discovered later [Gross 1996, Burke 1998b] this form of the action leads to paradoxical results. Rajagopal [Rajagopal 1996] attempted to introduce an action principle in TDDFT using the formalism of Jackiw and Kerman [Jackiw 1979] for deriving time-ordered n-point functions in quantum field theory. However, due to the time-ordering inherent in the work of Jackiw and Kerman the basic variable of Ragagopal's formalism is not the time-dependent density but an transition element of the density operator between a wavefunction evolving from the past and a wavefunction evolving from the future to a certain time t. Moreover, the action functional in this formalism suffers from the same difficulties as the action introduced by Runge and Gross. In this section we will show that these difficulties arise due to a restriction of the variational freedom as a consequence of v-representability constraints. For two other recent discussions of these points we refer to [van Leeuwen 2001, Maitra 2002c].

We start with the following time-dependent action functional

$$A[\Psi] = \int_{t_0}^{t_1} \mathrm{d}t \ \langle \Psi | \mathrm{i}\frac{\partial}{\partial t} - \hat{H}(t) | \Psi \rangle \ . \tag{2.47}$$

The usual approach is to require the action to be stationary under variations $\delta\Psi$ that satisfy $\delta\Psi(t_0) = \delta\Psi(t_1) = 0$. We then find after a partial integration

$$\delta A = \int_{t_0}^{t_1} \mathrm{d}t \ \langle \delta\Psi | \mathrm{i}\frac{\partial}{\partial t} - \hat{H}(t) | \Psi \rangle + \text{c.c.} + \mathrm{i}\langle \Psi | \delta\Psi \rangle \big|_{t_0}^{t_1} \ . \tag{2.48}$$

With the boundary conditions and the fact that the real and imaginary part of $\delta\Psi$ can be varied independently we obtain the result that

$$\left[\mathrm{i}\frac{\partial}{\partial t} - \hat{H}(t)\right]|\Psi\rangle = 0 \;, \tag{2.49}$$

which is just the time-dependent Schrödinger equation. We see that the variational requirement $\delta A = 0$, together with the boundary conditions is equivalent to the time-dependent Schrödinger equation.

A different derivation [Löwdin 1972] which does not put any constraints on the variations at the endpoints of the time interval is the following. We consider again a first order change in the action due to changes in the wavefunction and require that the action is stationary. We have the general relation

$$0 = \delta A = \int_{t_0}^{t_1} \mathrm{d}t \, \langle\delta\Psi|\mathrm{i}\frac{\partial}{\partial t} - \hat{H}(t)|\Psi\rangle + \int_{t_0}^{t_1} \mathrm{d}t \, \langle\Psi|\mathrm{i}\frac{\partial}{\partial t} - \hat{H}(t)|\delta\Psi\rangle \;. \tag{2.50}$$

We now choose the variations $\delta\Psi = \delta\tilde{\Psi}$ and $\delta\Psi = \mathrm{i}\delta\tilde{\Psi}$ where $\delta\tilde{\Psi}$ is arbitrary. We thus obtain

$$0 = \delta A = \int_{t_0}^{t_1} \mathrm{d}t \, \langle\delta\tilde{\Psi}|\mathrm{i}\frac{\partial}{\partial t} - \hat{H}(t)|\Psi\rangle + \int_{t_0}^{t_1} \mathrm{d}t \, \langle\Psi|\mathrm{i}\frac{\partial}{\partial t} - \hat{H}(t)|\delta\tilde{\Psi}\rangle \tag{2.51a}$$

$$0 = \delta A = -\mathrm{i}\int_{t_0}^{t_1} \mathrm{d}t \, \langle\delta\tilde{\Psi}|\mathrm{i}\frac{\partial}{\partial t} - \hat{H}(t)|\Psi\rangle + \mathrm{i}\int_{t_0}^{t_1} \mathrm{d}t \, \langle\Psi|\mathrm{i}\frac{\partial}{\partial t} - \hat{H}(t)|\delta\tilde{\Psi}\rangle \;. \tag{2.51b}$$

From (2.51a) and (2.51b) we obtain

$$0 = \int_{t_0}^{t_1} \mathrm{d}t \, \langle\delta\tilde{\Psi}|\mathrm{i}\frac{\partial}{\partial t} - \hat{H}(t)|\Psi\rangle \;. \tag{2.52}$$

Since this must be true for arbitrary $\delta\tilde{\Psi}$ we again obtain the time-dependent Schrödinger equation

$$\left[\mathrm{i}\frac{\partial}{\partial t} - \hat{H}(t)\right]|\Psi\rangle = 0 \;. \tag{2.53}$$

We did not need to put any boundary conditions on the variations at all. We only required that if $\delta\tilde{\Psi}$ is an allowed variation that then also $\mathrm{i}\delta\tilde{\Psi}$ is an allowed variation.

Let us now discuss the problems with the variational principle when one attempts to construct a time-dependent density-functional theory. The obvious definition of a density functional would be [Runge 1984]

$$A[n] = \int_{t_0}^{t_1} \mathrm{d}t \, \langle\Psi[n]|\mathrm{i}\frac{\partial}{\partial t} - \hat{H}(t)|\Psi[n]\rangle \;, \tag{2.54}$$

where $|\Psi[n]\rangle$ is a wavefunction which yields the density $n(\boldsymbol{r},t)$ and evolves from a given initial state $|\Psi_0\rangle$ with initial density $n_0(\boldsymbol{r})$. By the Runge-Gross theorem such a wave function is determined up to a phase factor. In order to define the action uniquely we have to make a choice for this phase factor. An obvious choice would be to choose the $|\Psi[n]\rangle$ that evolves in the external potential $v_{\text{ext}}(\boldsymbol{r},t)$ that vanishes at infinity and yields the density $n(\boldsymbol{r},t)$. This corresponds to choosing a particular kind of gauge. There are of course many more phase conventions possible. The trouble obviously arises from the fact that the density only determines the wavefunction up to an arbitrary time-dependent phase. However, there are more problems. Suppose we avoid the phase problem by defining a functional of the external potential rather than the density

$$A[v] = \int_{t_0}^{t_1} \mathrm{d}t \; \langle \Psi[v]| \mathrm{i}\frac{\partial}{\partial t} - \hat{H}(t)|\Psi[v]\rangle \; . \qquad (2.55)$$

Note that the potential v in the argument of the action is only used to parametrize the set of wavefunctions used in the action principle. This potential v is therefore not the same as the external potential in the Hamiltonian $\hat{H}(t)$ of (2.55) as this Hamiltonian is fixed. The state $|\Psi[v]\rangle$ is a state that evolves from a given initial state $|\Psi_0\rangle$ by solution of a time-dependent Schrödinger equation with potential v as its external potential. As the potential obviously defines $|\Psi[v]\rangle$ uniquely, including its phase, the action is well-defined. The question is now whether one can recover the time-dependent Schrödinger equation by making the action stationary with respect to potential variations δv. It is readily seen that this is not the case. The reason for this is that all variations $\delta\Psi$ of the wave function must now be caused by potential variations δv which leads to variations over a restricted set of wave functions. In other words, the variations $\delta\Psi$ must be v-representable. For instance, when deriving the Schrödinger equation from the variational principle one can not assume the boundary conditions $\delta\Psi(t_0) = \delta\Psi(t_1) = 0$. Since the time-dependent Schrödinger equation is first order in time, the variation $\delta\Psi(t)$ at times $t > t_0$ is completely determined by the boundary condition for $\delta\Psi(t_0)$. We are thus no longer free to specify a second boundary condition at a later time t_1. Moreover, we are not allowed to treat the real and imaginary part of $\delta\Psi$ as independent variations since both are determined simultaneously by the potential variation δv. This means that the first derivation of the TDSE that we presented in this section can not be carried out. It is readily seen that also the second derivation based on (2.51a) and (2.51b) fails. If $\delta\Psi$ is a variation generated by some $\delta\hat{V}(t) = \int \mathrm{d}^3 r \, \hat{n}(\boldsymbol{r})\delta v(\boldsymbol{r},t)$, then $\delta\Psi$ satisfies

$$\left[\mathrm{i}\frac{\partial}{\partial t} - \hat{H}_v(t)\right] |\delta\Psi\rangle = \delta\hat{V}(t)|\Psi\rangle \; , \qquad (2.56)$$

where $\hat{H}_v$ is a Hamiltonian with potential v and we neglected terms of higher order. Multiplication by the imaginary number "i" yields that the variation $\mathrm{i}\delta\Psi$ must be generated by potential $\mathrm{i}\delta v$. This potential variation is however

imaginary and therefore not an allowed variation since all potential variations must be real.

We therefore conclude that time-dependent density-functional theory can not be based on the usual variational principle, and indeed attempts to do so have led to paradoxes. In Chap. 3 we will discuss how an extended type of action functional defined on a Keldysh time-contour [van Leeuwen 2001, van Leeuwen 1998] can be used as a basis from which the time-dependent Kohn-Sham equations can be derived. This also has the immediate advantage that the action functional can then be directly related to the elegant formalism of nonequilibrium Green function theory which offers a systematic way of constructing time-dependent density functionals. Some examples of such functionals can be found in reference [von Barth 2005]. With hindsight it is interesting to see that already the work of Peuckert [Peuckert 1978], which is one of the very first papers in TDDFT, makes a connection to Keldysh Green functions, and in fact several of his results (such as the adiabatic connection formula) are perfectly valid when interpreted in terms of the action formalism of the next chapter.