



Linear Continuum Mechanics for Quantum Many-Body Systems

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We develop the continuum mechanics of quantum many-body systems in the linear response regime. The basic variable of the theory is the displacement field, for which we derive a closed equation of motion under the assumption that the time-dependent wave function in a locally comoving reference frame can be described as a geometric deformation of the ground-state wave function. We show that this equation of motion is exact for systems consisting of a single particle, and for all systems at sufficiently high frequency, and that it leads to an excitation spectrum that has the correct integrated strength. The theory is illustrated by simple model applications to one- and two-electron systems.

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The dynamics of quantum many-particle systems, as displayed in electromagnetic transitions, quasiparticle relaxation, chemical reactions, ionization, and collision processes, poses a major challenge to computational physicists and chemists. Whereas the calculation of ground-state properties can be tackled by powerful computational methods such as the quantum Monte Carlo method [1], the development of similar methods for time-dependent properties has been slow. One of the most successful methods to date is the time-dependent density functional theory (TDDFT), or its more recent version—time-dependent *current* density functional theory (TDCDFT) [2]. In the common Kohn-Sham (KS) implementation of this method [3,4] the formidable problem of solving the time-dependent Schrödinger equation for the many-body wave function is replaced by the much simpler problem of determining N single-particle orbitals. However, even this simplified problem is quite complex, and furthermore there are features such as multiparticle excitations [5] and dispersion forces [6] that are very difficult to treat within the conventional approximation schemes.

An alternative approach, which actually dates back to the early days of quantum mechanics [7–9], attempts to calculate directly the collective variables of interest—density and current. We call this approach “quantum continuum mechanics” (QCM) because, in analogy with classical theories of continuous media, it attempts to describe the quantum many-body system without explicit reference to the individual particles [10].

The possibility of a QCM formulation of the quantum many-body problem is guaranteed by the very same theorems that lay the foundation of TDDFT and TDCDFT [11,12]. Let us consider a system of particles described by the time-dependent Hamiltonian

$$\hat{H}(t) = \hat{H}_0 + \int d\mathbf{r} \hat{n}(\mathbf{r}) V_1(\mathbf{r}, t), \quad (1)$$

where $\hat{H}_0 = \hat{T} + \hat{W} + \hat{V}_0$ is the sum of kinetic energy (\hat{T}), interaction potential energy (\hat{W}), and the potential energy associated with an external *static* potential (\hat{V}_0). $\hat{n}(\mathbf{r})$ is the particle density operator and $V_1(\mathbf{r}, t)$ is an external time-dependent potential. The exact Heisenberg equation of motion for the current density operator, averaged over the quantum state, leads to the Euler equation

$$m \partial_t j_{\mu}(\mathbf{r}, t) = -n(\mathbf{r}, t) \partial_{\mu} [V_0(\mathbf{r}) + V_1(\mathbf{r}, t)] - \partial_{\nu} P_{\mu\nu}(\mathbf{r}, t). \quad (2)$$

Here m is the mass of the particles and repeated indices are summed over. The key quantity on the right-hand side of Eq. (2) is the *stress tensor* $P_{\mu\nu}(\mathbf{r}, t)$ —a symmetric tensor whose divergence yields the *force density* arising from quantum-kinetic and interaction effects. Now the Runge-Gross theorem of TDDFT guarantees that the stress tensor, like every observable of the system, is a functional of the current density and of the initial quantum state. Thus, Eq. (2) is in principle a closed equation of motion for \mathbf{j} —the only missing piece being the *explicit* expression for $P_{\mu\nu}$ in terms of the current density.

In recent years much effort has been devoted to constructing an approximate QCM [13–18], and several applications have appeared in the literature (see Ref. [19] for some representative examples). All approximation schemes so far have been based on the local density approximation and generalizations thereof. In this Letter we derive a new approximate expression for $P_{\mu\nu}(\mathbf{r}, t)$ as a functional of the current density for systems that perform small amplitude oscillations about the ground state. The new formula is nonlocal, is expressed in terms of ground-state properties that can be calculated without recourse to DFT, and becomes exact in the high-frequency limit.

The Euler equation (2) is conveniently expressed in terms of the displacement field $\mathbf{u}(\mathbf{r}, t)$, which in the linear regime is defined by $\mathbf{j}(\mathbf{r}, t) = n_0(\mathbf{r}) \partial_t \mathbf{u}(\mathbf{r}, t)$, where $n_0(\mathbf{r})$ is

the ground-state density. It is also convenient to write the density and the stress tensor as the sum of a large ground-state component and a small time-dependent part, i.e., $n(\mathbf{r}, t) = n_0(\mathbf{r}) + n_1(\mathbf{r}, t)$ and $P_{\mu\nu}(\mathbf{r}, t) = P_{\mu\nu,0}(\mathbf{r}) + P_{\mu\nu,1}(\mathbf{r}, t)$. Then the linearized form of the Euler equation (2) takes the form

$$mn_0(\mathbf{r})\partial_t^2\mathbf{u} = -n_0(\mathbf{r})\nabla V_1(\mathbf{r}, t) + \mathcal{F}_1(\mathbf{r}, t), \quad (3)$$

where the total force density,

$$\mathcal{F}_{\mu,1}(\mathbf{r}, t) \equiv -n_1(\mathbf{r}, t)\partial_\mu V_0(\mathbf{r}) - \partial_\nu P_{\mu\nu,1}(\mathbf{r}, t), \quad (4)$$

is a linear functional of $\mathbf{u}(\mathbf{r}, t)$. Our approximate expression for $\mathcal{F}_{\mu,1}$ will be presented in terms of the functional

$$E[\mathbf{u}] \equiv \langle \psi_0[\mathbf{u}] | \hat{H}_0 | \psi_0[\mathbf{u}] \rangle, \quad (5)$$

which is the energy of the deformed ground state $|\psi_0[\mathbf{u}]\rangle$, obtained from the undistorted ground state $|\psi_0\rangle$ by displacing the volume element located at \mathbf{r} to a new position $\mathbf{r} + \mathbf{u}(\mathbf{r}, t)$. More precisely, we will argue that the force density can be represented as

$$\mathcal{F}_{\mu,1}(\mathbf{r}, t) = - \int d\mathbf{r}' \frac{\delta^2 E[\mathbf{u}]}{\delta u_\mu(\mathbf{r}) \delta u_\nu(\mathbf{r}')} \Big|_{\mathbf{u}=0} u_\nu(\mathbf{r}', t), \quad (6)$$

where the second variational derivative of $E[\mathbf{u}]$, evaluated at the ground state ($\mathbf{u} = 0$) has an exact expression in terms of the one- and two-particle density matrices of the *ground state*. We will show that the representation (6) is exact for all one-particle systems and also for many-particle systems at sufficiently high frequency.

Equation (6) can be derived by performing a transformation to the ‘‘comoving reference frame’’ [17,18]—a non-inertial frame in which the density is constant and equal to the ground-state density and the current density is zero—and assuming that the wave function in this frame is independent of time. This assumption is correct in one-particle systems, where the constancy of the density and the vanishing of the current density completely determine the wave function. It is also generally valid on very short time scales, or for frequencies higher than the characteristic energy of single-particle excitations, because on these time scales it is not possible for the particles to ‘‘forget’’ the correlations built into the initial ground-state wave function. In all other cases our approximation replaces the exact ‘‘normal modes’’ of the system by a smaller set of approximate normal modes, in such a way that the total spectral weight is conserved. Rather than delving into the sophisticated mathematics of the transformation to the comoving reference frame, in this Letter we present a more elementary and direct derivation of Eq. (6), which clearly demonstrates the satisfaction of the spectral sum rules.

We start from the linear response of the current density to an external vector potential of frequency ω

$$j_\mu(\mathbf{r}, \omega) = \int d\mathbf{r}' \chi_{\mu\nu}(\mathbf{r}, \mathbf{r}', \omega) A_{\nu,1}(\mathbf{r}', \omega), \quad (7)$$

where $j_\mu(\mathbf{r}, \omega)$ is the Fourier component of the current at

frequency ω and $\chi_{\mu\nu}(\mathbf{r}, \mathbf{r}', \omega)$ is the current-current response function. At high frequency, $\chi_{\mu\nu}$ has the well-known expansion [20]

$$\chi_{\mu\nu}(\mathbf{r}, \mathbf{r}', \omega) = \frac{n_0(\mathbf{r})}{m} \delta(\mathbf{r} - \mathbf{r}') \delta_{\mu\nu} + \frac{M_{\mu\nu}(\mathbf{r}, \mathbf{r}')}{m^2 \omega^2}, \quad (8)$$

where the first term (diamagnetic) is frequency-independent and

$$M_{\mu\nu}(\mathbf{r}, \mathbf{r}') \equiv -m^2 \langle \Psi_0 | [\hat{H}_0, \hat{j}_\mu(\mathbf{r}), \hat{j}_\nu(\mathbf{r}')] | \Psi_0 \rangle \quad (9)$$

is the first spectral moment of the current-current response function. Now, substituting Eq. (8) in Eq. (7) and noting that $\mathbf{j}(\mathbf{r}, \omega) = -i\omega n_0(\mathbf{r})\mathbf{u}(\mathbf{r}, \omega)$ and that the vector potential is related to the scalar potential by the equation $\mathbf{A}_1(\mathbf{r}, \omega) = \frac{\nabla V_1(\mathbf{r}, \omega)}{i\omega}$, we obtain (to leading order in $1/\omega^2$)

$$-m\omega^2 n_0 u_\mu = -n_0 \partial_\mu V_1 - \int d\mathbf{r}' M_{\mu\nu}(\mathbf{r}, \mathbf{r}') u_\nu(\mathbf{r}', \omega). \quad (10)$$

This is equivalent to our equation of motion (3), with $\mathcal{F}_{\mu,1}$ given by Eq. (6), if and only if

$$M_{\mu\nu}(\mathbf{r}, \mathbf{r}') = \frac{\delta^2 E[\mathbf{u}]}{\delta u_\mu(\mathbf{r}) \delta u_\nu(\mathbf{r}')} \Big|_{\mathbf{u}=0}. \quad (11)$$

To show that this is the case, we observe that the deformed ground state is related to the undeformed ground state by the unitary transformation

$$|\Psi_0[\mathbf{u}]\rangle = e^{-i \int d\mathbf{r} \hat{\mathbf{j}}(\mathbf{r}) \cdot \mathbf{u}(\mathbf{r})} |\Psi_0\rangle. \quad (12)$$

Here we have used the fact that the current density operator $\hat{\mathbf{j}}(\mathbf{r})$ is the generator of a translation of all the particles in an infinitesimal volume located at \mathbf{r} . Thus, the transformation (12) amounts to performing different translations by vectors $\mathbf{u}(\mathbf{r})$ at different points in space, i.e., precisely to deforming the system according to the displacement field $\mathbf{u}(\mathbf{r})$. Substituting the above expression for $|\Psi_0[\mathbf{u}]\rangle$ in the definition of $E[\mathbf{u}]$ and expanding to second order in \mathbf{u} , we can easily verify that

$$E[\mathbf{u}] \simeq E_0 + \frac{1}{2} \int d\mathbf{r} \int d\mathbf{r}' u_\mu(\mathbf{r}) M_{\mu\nu}(\mathbf{r}, \mathbf{r}') u_\nu(\mathbf{r}'), \quad (13)$$

which establishes the validity of Eq. (11) [21].

A lengthy calculation allows us to calculate the three components of the force density functional arising from the kinetic, interaction, and external potential parts of the Hamiltonian: $\mathcal{F}_{\mu,1} = \mathcal{F}_{\mu,1}^{\text{kin}} + \mathcal{F}_{\mu,1}^{\text{int}} + \mathcal{F}_{\mu,1}^{\text{pot}}$, where

$$\begin{aligned} \mathcal{F}_{\mu,1}^{\text{kin}} &= \partial_\alpha [2T_{\nu\mu,0} u_{\nu\alpha} + T_{\nu\alpha,0} \partial_\mu u_\nu] - \frac{1}{4m} \partial_\nu \partial_\mu (n_0 \partial_\nu \nabla \cdot \mathbf{u}) \\ &\quad + \frac{1}{4m} \partial_\nu \{2(\nabla^2 n_0) u_{\nu\mu} + (\partial_\nu n_0) \partial_\mu \nabla \cdot \mathbf{u} \\ &\quad + (\partial_\mu n_0) \partial_\nu \nabla \cdot \mathbf{u} - 2\partial_\mu [(\partial_\alpha n_0) u_{\nu\alpha}]\}, \end{aligned} \quad (14)$$

$$[u_{\nu\mu} = (\partial_\nu u_\mu + \partial_\mu u_\nu)/2],$$

$$\mathcal{F}_{\mu,1}^{\text{int}} = \int d\mathbf{r}' K_{\mu\nu}(\mathbf{r}, \mathbf{r}') [u_\nu(\mathbf{r}) - u_\nu(\mathbf{r}')], \quad (15)$$

$$\mathcal{F}_{\mu,1}^{\text{pot}} = -n_0(\mathbf{r})\mathbf{u} \cdot \nabla \partial_{\mu} V_0. \quad (16)$$

Here we have introduced the equilibrium stress tensor

$$T_{\mu\nu,0} = \frac{1}{2m}(\partial_{\mu}\partial'_{\nu} + \partial_{\nu}\partial'_{\mu})\rho^{(1)}(\mathbf{r},\mathbf{r}')|_{\mathbf{r}=\mathbf{r}'} - \frac{1}{4m}\nabla^2 n_0 \delta_{\mu\nu}, \quad (17)$$

where $\rho^{(1)}(\mathbf{r},\mathbf{r}')$ is the one-particle density matrix. The interaction kernel K in Eq. (15) is given by

$$K_{\mu\nu}(\mathbf{r},\mathbf{r}') = \rho_2(\mathbf{r},\mathbf{r}')\partial_{\mu}\partial'_{\nu}w(|\mathbf{r}-\mathbf{r}'|), \quad (18)$$

where $w(|\mathbf{r}-\mathbf{r}'|)$ is the interaction potential and $\rho_2(\mathbf{r},\mathbf{r}') \equiv \rho^{(2)}(\mathbf{r},\mathbf{r}'|\mathbf{r},\mathbf{r}')$, where $\rho^{(2)}$ is the two-particle density matrix.

The excitation energies of the system are obtained from the solution of Eq. (10) after setting $V_1 = 0$. This equation defines a Hermitian eigenvalue problem with positive eigenvalues ω_n^2 —the square of the excitation energies. The positivity follows from the fact that a deformation of the ground-state wave function must necessarily increase the energy. The corresponding eigenfunctions $\mathbf{u}_n(\mathbf{r})$ are mutually orthogonal with respect to the scalar product $(\mathbf{u}_n, \mathbf{u}_m) \equiv \int d\mathbf{r} u_n(\mathbf{r})u_m(\mathbf{r})n_0(\mathbf{r}) = 0$ if $n \neq m$. These eigenfunctions must be regarded as approximations to the matrix elements of the current density operator between the ground state and the excited state in question, i.e., $\mathbf{u}_n(\mathbf{r}) \approx \frac{[\mathbf{j}]_{n0}(\mathbf{r})}{\omega_n n_0(\mathbf{r})}$, where $[\mathbf{j}]_{n0}(\mathbf{r}) \equiv \langle \Psi_n | \hat{\mathbf{j}}(\mathbf{r}) | \Psi_0 \rangle$. It is easy to verify that the sum rule $\sum_n \omega_n [\mathbf{j}_{\mu}]_{n0}(\mathbf{r}) [\mathbf{j}_{\nu}]_{0n}(\mathbf{r}') = m^{-2} M_{\mu\nu}(\mathbf{r},\mathbf{r}')$ is satisfied by the approximate $[\mathbf{j}]_{n0}(\mathbf{r})$. In this sense our approximation preserves the total strength of the spectrum. It is only for one-particle systems that the “approximate” $[\mathbf{j}]_{n0}(\mathbf{r})$ becomes exact. Let us now illustrate the theory with two simple examples.

Linear harmonic oscillator.—For a harmonic oscillator of frequency ω_0 , the eigenvalue problem takes the form

$$\frac{1}{4}u'''' - xu''' + (x^2 - 2)u'' + 3xu' - \frac{\omega^2 - \omega_0^2}{\omega_0^2}u = 0, \quad (19)$$

where the prime denotes differentiation with respect to x . Solving Eq. (19) with the boundary condition $n_0^{1/2}(x)u(x) \rightarrow 0$ for $|x| \rightarrow \infty$, we obtain the exact excitation spectrum $\omega_n = \pm n\omega_0$ ($n = 1, 2, \dots$) and the corresponding eigenfunctions $u_n(x) \propto H_{n-1}(x)$, which are proportional to the matrix elements of the current density operator.

Two-electron system.—Consider a system of two electrons repelling each other with interaction potential $\frac{e^2}{|x_1 - x_2|}$ in a one-dimensional parabolic trap of frequency ω_0 . Because of the separation of center of mass and relative variables this model can easily be solved numerically, and even analytically in the limit of strong correlation. We only focus on the strongly correlated limit ($\omega_0 \rightarrow 0$) and the singlet states. In this limit the two electrons become localized near $x = \pm x_0/2$, where $x_0 = (2e^2/m\omega_0^2)^{1/3}$ is large compared to $l = (m\omega_0)^{-1/2}$ (see Fig. 1). The wave func-

tion for the relative coordinate $x_r = x_1 - x_2$ is symmetric and consists of two identical “blobs” centered at $\pm x_0$, each blob being the wave function of a harmonic oscillator of frequency $\omega_0\sqrt{3}$. The center of mass wave function is that of a harmonic oscillator with frequency ω_0 . The exact eigenstates are characterized by two non-negative integers n (center of mass) and m (relative motion) and are denoted by (n, m) . $(0, 0)$ is the ground state. The excitation energy associated with the state (n, m) is $E_{nm} = (n + m\sqrt{3})\omega_0$. From the wave functions we calculate, without approximations, the displacement field $u_{nm}(x)$. Some of the results are shown in Fig. 1. The displacement field of the $(1, 0)$ excitation, which corresponds to a rigid translation of the center of mass, is uniform in space, while the displacement field of the $(0, 1)$ excitation, which corresponds to the classical breathing mode, changes sign around the origin. The $(1, 0)$ and $(0, 1)$ modes exhaust the classical phonon modes of a system of two localized particles. The remaining excitations are fully quantum mechanical. Examining Fig. 1 one quickly realizes that all the excitations with a given value of $n + m$ and the same parity of m produce the same displacement field, but have different energies.

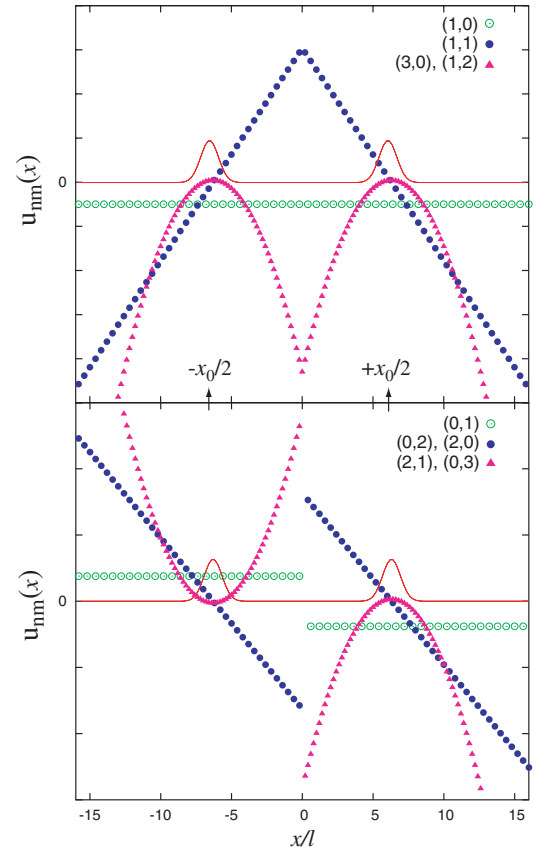


FIG. 1 (color online). Unnormalized displacement fields for a few low-lying excitations of the two-electron system described in the text. The solid line is the ground-state density. Analytically we find $u_{nm}(x) \propto H_{n+m-1}[2(x - x_0/2)]$ for $x \approx x_0/2$, with parity $(-1)^{n-1}$ independent of m . The large value of the displacement field for $x \sim 0$ does not have a physical significance since the density is exponentially small in that region.

TABLE I. Comparison between exact and calculated (appr) excitation energies in the strongly correlated regime. The average frequency $\bar{\omega}$ of a group of excitations is calculated numerically from the sum rule discussed in the text. Analytically one finds $\bar{\omega}^2/\omega_0^2 = 2 + 3\sqrt{3}k + 6k(k-1)(2-\sqrt{3}) - (-1)^m \times (2-\sqrt{3})^k$, where $k \equiv n+m-1$: these exact values are indistinguishable, up to the third decimal digit, from the numerical results listed in the last column.

(n, m)	$\omega_{nm}^{\text{exact}}/\omega_0$	$\omega_{nm}^{\text{appr}}/\omega_0$	$\bar{\omega}/\omega_0$
(1, 0)	1.0	1.00	1.00
(0, 1)	1.732	1.740	1.732
(2, 0)	2.0	2.643	2.632
(0, 2)	3.464		
(1, 1)	2.732	2.736	2.732
(3, 0)	3.0	3.950	3.942
(1, 2)	4.464		
(2, 1)	3.732	3.965	3.960
(0, 3)	5.196		
(4, 0)	4.0	5.224	5.217
(2, 2)	5.464		
(0, 4)	6.928		

Let us now see what our elastic equation of motion predicts for this system. In Table I we present the energies of a few low-lying excitations obtained from the numerical solution of Eq. (10) in the strongly correlated regime. We see that the energies of excitations such as (1, 0), (0, 1), and (1, 1), which do not “share” their displacement field with other excitations, are very well reproduced by our calculation within the accuracy of the numerical work. On the other hand, groups of excitations that share the same displacement field are replaced by a single excitation of average frequency, in such a way that the total spectral strength of the group is preserved. It can be proved that the excitation frequency $\bar{\omega}$ that replaces the frequencies ω_l of the excitations in a given group is given by the sum rule $\bar{\omega}^2 = \sum_l f_l \omega_l^2$, where $f_l = \frac{2m|\int d\mathbf{r} \mathbf{j}_{0l}(\mathbf{r}) \cdot \bar{\mathbf{u}}(\mathbf{r})|^2}{\omega_l}$ is the “oscillator strength” of the l th excitation, $\bar{\mathbf{u}}(\mathbf{r})$ is the normalized solution of the eigenvalue problem with eigenvalue $\bar{\omega}^2$, and the sum runs over all the excitations in the group. In the last column of Table I, we have checked that the sum rule is quite well satisfied by our numerical solution.

The QCM formulation is applicable directly to the Kohn-Sham system, in which case we only need the ground-state KS orbitals and a reasonable approximation for the exchange-correlation field. While the KS method treats the noninteracting kinetic stress tensor exactly, our method should be computationally more agile, since it does not involve time-dependent orbitals and/or the inversion of large linear response matrices. Our theory extends the well-known collective approximation of the homogeneous electron gas to inhomogeneous systems and should therefore be useful in dealing with collective effects such as multiparticle excitations and the dipolar fluctuations that are responsible for van der Waals attraction [22,23].

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