Quantum many-body dynamics in a Lagrangian frame: II. Geometric formulation of time-dependent density functional theory

I. V. Tokatly[1,*]

[1]*Lehrstuhl für Theoretische Festkörperphysik, Universität Erlangen-Nürnberg, Staudtstrasse 7/B2, 91058 Erlangen, Germany and Moscow Institute of Electronic Technology, Zelenograd 124498, Russia*
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We formulate equations of time-dependent density functional theory (TDDFT) in the comoving Lagrangian reference frame. The main advantage of the Lagrangian description of many-body dynamics is that in the comoving frame the current density vanishes, while the density of particles becomes independent of time. Therefore a comoving observer will see a picture which is very similar to that seen in the equilibrium system from the laboratory frame. It is shown that the most natural set of basic variables in TDDFT includes the Lagrangian coordinate, ξ, a symmetric deformation tensor $g_{\mu\nu}$, and a skew-symmetric vorticity tensor, $F_{\mu\nu}$. These three quantities, respectively, describe the translation, deformation, and the rotation of an infinitesimal fluid element. Reformulation of TDDFT in terms of new basic variables resolves the problem of nonlocality and thus allows us to regularly derive a local nonadiabatic approximation for exchange correlation (xc) potential. Stationarity of the density in the comoving frame makes the derivation to a large extent similar to the derivation of the standard static local density approximation. We present a few explicit examples of nonlinear nonadiabatic xc functionals in a form convenient for practical applications.

I. INTRODUCTION

Traditionally, the physical understanding of various many-body phenomena is based on Landau's intuitive concept of quasiparticles, which relates the behavior of a strongly interacting quantum system to the properties of a gas of noninteracting (or weakly interacting) elementary excitations.[1] The common field-theoretical formulation of the many-body problem[2–4] allows us to rigorously justify this very appealing point of view, provided the system is in a weakly excited many-body state. Unfortunately, practical applications of traditional many-body methods to real systems, even in the equilibrium or linear response regime, is computationally very demanding. In strong nonequilibrium systems the situation is much worse. The simple intuitive picture of elementary excitations breaks down, while the direct application of the standard many-body theory becomes increasingly difficult even for model systems.

An alternative approach to the quantum many-body problem is offered by the density functional theory [5–7] (DFT). DFT represents a mathematically rigorous realization of another famous idea in theoretical physics which is a concept of collective variables theory.[8] Indeed, DFT opens a possibility to formulate the many-body problem in the form of a closed theory that contains only a restricted set of basic variables, such as density in the static DFT (Ref. 5) or density and current in the time-dependent DFT (TDDFT).[6,7,9] In classical physics a theory of this type has been known for more than 250 years. This is classical hydrodynamics. In fact, the Runge-Gross mapping theorem in TDDFT (Ref. 9) proves the existence of an exact quantum hydrodynamics. An analogy of TDDFT to hydrodynamics has been already noted in the original paper by Runge and Gross,[9] (see also Tokatly and Pankratov[10]). In this respect the static DFT should be viewed as an exact quantum hydrostatic. It is indeed known

that the condition of energy minimum is equivalent to the condition for a local compensation of the external and internal stress forces exerted on every infinitesimal element of an equilibrium system.[11] Thus DFT not only introduces an alternative formalism in the quantum many-body problem, but it also naturally suggests an alternative way of thinking that refers to the physical intuition, developed over hundreds of years of experience in classical continuum mechanics. Interestingly, equations of TDDFT in the hydrodynamic formulation can be also considered as force balance conditions, but in a local noninertial reference frame moving with the flow. In the time-dependent case there is a local compensation of the external, inertial, and internal stress forces. This demonstrates a close similarity of the static DFT (which is currently a well developed theory) to TDDFT (which is still under construction) in the comoving frame. The above similarity was the main motivation to reconsider the formulation of TDDFT from the point of view of a local observer in the comoving Lagrangian reference frame. In this paper we present the results of such a reconsideration, based on our recent formulation of the many-body theory in the Lagrangian frame.[12] (In what follows the paper in Ref. 12 is referred to as I.)

Practical applications of any DFT rely on the Kohn-Sham (KS) construction,[5,13] which maps the calculation of basic observables in the interacting systems to the solution of an auxiliary, noninteracting KS problem. Noninteracting KS particles move in a self-consistent exchange correlation (xc) potential that is adjusted to reproduce the correct values of basic variables, i.e., density and current in TDDFT. From the hydrodynamical point of view the KS construction allows one to compute exactly the kinetic part of the internal stress force, while treating the xc contribution to the stress in an approximate fashion. Thus the central problem of any practical DFT reduces to the construction of adequate approxi-

mations for xc potentials. In the static DFT a good starting point is provided by the local density approximation (LDA). On the one hand, the static LDA, by itself, gives quite reasonable results, and, on the other hand, it allows for further modifications and refinements. The construction of a similar basic local approximation in TDDFT turned out to be problematic. The reason for these problems is the inherent nonlocality of the nonadiabatic, nonequilibrium DFT. General arguments,[14,15] based on the harmonic potential theorem,[16] require that any consistent nonadiabatic xc potential must be a strongly nonlocal functional of the density. Otherwise the harmonic potential theorem is violated.

In 1996 Vignale and Kohn[17] (VK) realized that, at least in the linear response regime, the problem of nonlocality can be resolved by changing the basic variable and by using the xc vector potential $\mathbf{A}^{xc}$ instead of the scalar one. Namely, VK showed that if one considers the current $\mathbf{j}$ (instead of the density n) as a basic variable, a consistent linear local approximation for $\mathbf{A}^{xc}$ can be regularly derived.[17] Shortly after that, Vignale, Ullrich, and Conti[18] (VUC) found that the further switching of variables from the current $\mathbf{j}$ to the velocity $\mathbf{v} = \mathbf{j}/n$ allows us to represent the complicated VK expression in a physically transparent viscoelastic form. VUC (Ref. 18) also phenomenologically constructed a nonlinear, nonadiabatic xc functional. In this construction they formally adopted the linear VK form, but with coefficients taken at a "retarded position."[19] The meaning is that the phenomenological construction is (i) similar, (ii), linear, and (iii) based on the Keldysh contour action functional, has been recently proposed by Kurzweil and Baer.[20] An attempt to regularly derive a nonlinear, nonadiabatic local approximation in TDDFT was made in Tokatly and Pankratov.[10] Noting that the applicability conditions for the linear VK approximation exactly coincided with those for the "collisionless hydrodynamics" of Refs. 21–23, we extended the hydrodynamics formalism to TDDFT. Using Landau Fermi-liquid theory, we derived a nonadiabatic, nonlinear approximation for the xc stress tensor that defines the xc potential.[10] It has been shown that the stress tensor is a local functional of new basic variables: the Lagrangian coordinate and a second-rank metric-like tensor. An appearance of the Lagrangian coordinate as a basic variable is quite remarkable, if we recall the above-mentioned [see also I (Ref. 12)] staticlike force balance in the co-moving Lagrangian frame.

In this paper we reformulate TDDFT using the exact equations of quantum many-body dynamics in the Lagrangian frame.[12] We show that possibly the most natural complete set of basic variables in TDDFT consists of the Lagrangian coordinate $\boldsymbol{\xi}$, the symmetric Green's deformation tensor $g_{\mu\nu}$, and the skew-symmetric vorticity tensor $\widetilde{F}_{\mu\nu}$. These three quantities, one vector, one symmetric, and one skew-symmetric tensor, contain 12 numbers that are required for the complete local characterization of a deformed state of any continuum media.[24] Namely, $\boldsymbol{\xi}$, $g_{\mu\nu}$, and $\widetilde{F}_{\mu\nu}$, respectively, describe the translation, deformation, and the rotation of an infinitesimal fluid element. On the other hand, tensors $g_{\mu\nu}$ and $\widetilde{F}_{\mu\nu}$ describe generalized inertia forces in the Lagrangian frame (see I). All three quantities are functionals of velocity in accordance with the Runge-Gross mapping

theorem.[9] The new formulation of TDDFT relates the local stress in the system to the dynamic deformations, which is very natural physically. The main practical advantage is that the reformulation of TDDFT in terms of new basic variables resolves the problem of nonlocality on the most fundamental level. We show that the dynamic xc stress-deformation relation can be made local. This allows us to derive a local nonadiabatic approximation in a regular way that is similar to the derivation of the static LDA. The whole history of nonadiabatic approximations in TDDFT can be viewed as a staircase of successive transformations of basic variables, from the density, via the current and the velocity, to the general geometric characteristics of deformed media. The first step was made by VK (Ref. 17) in 1996. Hopefully the present formulation of TDDFT corresponds to the last step on this staircase.

The structure of the paper is the following. In Sec. II we consider the hydrodynamic formulation of TDDFT. Using this formulation, we introduce the KS system and define xc potentials in terms of stress tensors. In Sec. III we discuss local approximations in DFT. We derive the common static LDA and explain physical reasons for the nonlocality in TDDFT. Sections IV and V are the central parts of the present paper. In Sec. IV we introduce basic variables and develop a complete geometric formulation of TDDFT. We also formulate a universal many-body problem which determines the xc stress tensor and the xc potential as functionals of basic variables. In Sec. V we discuss an approximate solution of the universal many-body problem in the lowest order of the gradient expansion. Explicit examples of nonadiabatic local xc functionals are presented in Secs. V B–V D. In the concluding part of Sec. V D we formulate the full set of time-dependent KS equations in the nonlinear time-dependent local deformation approximation. In the equilibrium case these equations reduce to the common equations of DFT with the static LDA xc potential, while in the linear response regime we recover the results of VK approximation. In Sec. VI we summarize our results. Three appendixes contain technical details of the calculations.

II. DEFINITION OF EXCHANGE CORRELATION POTENTIALS VIA STRESS DENSITY

In this section we discuss a hydrodynamic formulation of DFT and introduce a definition of xc potentials in terms of local stress forces. Let us consider a system of N interacting fermions in the presence of a time-dependent external potential $U_{ext}(\mathbf{x},t)$. The system is described by the following Hamiltonian:

$$H = \hat{T} + \hat{W} + \hat{U}, \qquad (1)$$

where $\hat{T}$, $\hat{W}$, and $\hat{U}$ correspond to the operators of the kinetic energy, interparticle interaction energy, and the energy of interaction with the external field, respectively,

$$\hat{T} = -\int d\mathbf{x}\,\psi^{\dagger}(\mathbf{x})\frac{\nabla^2}{2m}\psi(\mathbf{x}), \qquad (2)$$

$$\hat{W} = \frac{1}{2}\int d\mathbf{x}\,d\mathbf{x}'\,w(|\mathbf{x}-\mathbf{x}'|)\psi^{\dagger}(\mathbf{x})\psi^{\dagger}(\mathbf{x}')\psi(\mathbf{x}')\psi(\mathbf{x}), \quad (3)$$

$$\hat{U} = \int d\mathbf{x}\, U_{\mathrm{ext}}(\mathbf{x},t)\psi^{\dagger}(\mathbf{x})\psi(\mathbf{x}). \qquad (4)$$

Here $w(|\mathbf{x}|)$ is the potential of a pairwise interparticle interaction. Field operators $\psi^{\dagger}$ and ψ satisfy the fermionic commutation relation,

$$\{\psi^{\dagger}(\mathbf{x}),\psi(\mathbf{x}')\} = \delta(\mathbf{x}-\mathbf{x}'). \qquad (5)$$

For definiteness we consider a Fermi system, although most of the results of this paper are independent of statistics.

The hydrodynamic formulation of DFT follows the Heisenberg equations of motion for the density and for the current operators

$$\frac{\partial}{\partial t}n(\mathbf{x},t) - i\langle [H,\hat{n}(\mathbf{x})]\rangle = 0, \qquad (6)$$

$$\frac{\partial}{\partial t}\mathbf{j}(\mathbf{x},t) - i\langle [H,\hat{\mathbf{j}}(\mathbf{x})]\rangle = 0, \qquad (7)$$

where $n(\mathbf{x})=\langle \hat{n}(\mathbf{x})\rangle$ and $\mathbf{j}(\mathbf{x})=\langle \hat{\mathbf{j}}(\mathbf{x})\rangle$ are the density of particles and the current density. The corresponding operators are defined by the standard expressions,

$$\hat{n}(\mathbf{x},t) = \psi^{\dagger}(\mathbf{x},t)\psi(\mathbf{x},t), \qquad (8)$$

$$\hat{j}_{\mu}(\mathbf{x},t) = -\frac{i}{2m}\left(\psi^{\dagger}\frac{\partial \psi}{\partial x^{\mu}} - \frac{\partial \psi^{\dagger}}{\partial x^{\mu}}\psi\right). \qquad (9)$$

Angle brackets in the above formulas denote averaging with the exact density matrix $\hat{\rho}$, i.e., $\langle A\rangle=\mathrm{Tr}\hat{\rho}A$. Equations (6) and (7) can be represented in a form of the hydrodynamics balance equations. (Details of the derivation can be found, for example, in I),

$$D_t n + n\frac{\partial}{\partial x^{\mu}}v_{\mu} = 0, \qquad (10)$$

$$mnD_t v_{\mu} + \frac{\partial}{\partial x^{\nu}}P_{\mu\nu} + n\frac{\partial}{\partial x^{\mu}}U = 0. \qquad (11)$$

Equation (10) is the common continuity equation, while Eq. (11) corresponds to the local momentum conservation law. In these equations $\mathbf{v}=\mathbf{j}/n$ is the velocity of the flow, $D_t = (\partial/\partial t)+\mathbf{v}\nabla$ is the convective derivative and $U=U_{\mathrm{ext}}+U_{\mathbf{H}}$ is the sum of the external and the Hartree potentials,

$$U_{\mathbf{H}}(\mathbf{x},t) = \int w(|\mathbf{x}-\mathbf{x}'|)n(\mathbf{x}',t)d\mathbf{x}'. \qquad (12)$$

The exact stress tensor in Eq. (11),

$$P_{\mu\nu}(\mathbf{x},t) = T_{\mu\nu}(\mathbf{x},t) + W_{\mu\nu}(\mathbf{x},t), \qquad (13)$$

contains the kinetic $T_{\mu\nu}$ and the interaction $W_{\mu\nu}$ contributions. Divergences of the tensors $T_{\mu\nu}$ and $W_{\mu\nu}$ in Eq. (11) come from the commutators of the current operator with operators $\hat{T}$, Eq. (2), and $\hat{W}$, Eq. (3), respectively. In I we have derived the following explicit representations for the stress tensors (see also Refs. 2 and 25–27):

$$T_{\mu\nu}(\mathbf{x}) = \frac{1}{2m}\left\langle (\hat{q}_{\mu}\psi)^{\dagger}\hat{q}_{\nu}\psi + (\hat{q}_{\nu}\psi)^{\dagger}\hat{q}_{\mu}\psi - \frac{\delta_{\mu\nu}}{2}\nabla^2\hat{n}\right\rangle, \qquad (14)$$

$$W_{\mu\nu}(\mathbf{x}) = -\frac{1}{2}\int d\mathbf{x}'\frac{x'^{\mu}x'^{\nu}}{|\mathbf{x}'|}\frac{\partial w(|\mathbf{x}'|)}{\partial |\mathbf{x}'|}$$
$$\times \int_0^1 G_2(\mathbf{x}+\lambda\mathbf{x}',\mathbf{x}-(1-\lambda)\mathbf{x}')d\lambda, \qquad (15)$$

where $\hat{\mathbf{q}}=-i\nabla-m\mathbf{v}$ is the operator of "relative" momentum, and $G_2(\mathbf{x},\mathbf{x}')=\langle\psi^{\dagger}(\mathbf{x})\hat{n}(\mathbf{x}')\psi(\mathbf{x})\rangle-n(\mathbf{x})n(\mathbf{x}')$ is the pair correlation function.[28] It is worth mentioning that the representability of the stress force in a form of a divergence of a tensor is a direct consequence of the Newton's third law (see Appendix A in I).

Equations (10) and (11) represent the exact local conservation laws which must be satisfied for an arbitrary evolution of the system. Let us apply them to TDDFT. The first, less restrictive part of the TDDFT mapping theorem[9,29] states the existence of a unique and invertible map: $\mathbf{j}\rightarrow U_{\mathrm{ext}}$ or, equivalently, $\mathbf{v}\rightarrow U_{\mathrm{ext}}$. This implies that the exact many-body density matrix $\hat{\rho}(t)$ for a given initial condition, $\hat{\rho}(0)=\hat{\rho}_0$, is a functional of the velocity $\mathbf{v}$. Hence the stress tensor of Eq. (13) is a functional of $\mathbf{v}$ and of the initial density matrix: $P_{\mu\nu}[\hat{\rho}_0,\mathbf{v}]$. Therefore Eqs. (10) and (11) constitute a formally closed set of the exact quantum hydrodynamics equations with the memory of initial many-body correlations. It is interesting to note that the common classical hydrodynamics can be viewed as a particular limiting form of TDDFT. In this limiting case the stress tensor functional is known explicitly—it takes the usual Navier-Stokes form.[30]

In the equilibrium system Eq. (11) reduces to the static force balance equation,

$$\frac{\partial}{\partial x^{\nu}}P_{\mu\nu} + n\frac{\partial}{\partial x^{\mu}}U = 0. \qquad (16)$$

This equation shows that the force produced by the external and the Hartree potentials is compensated by the force of internal stresses. The net force, exerted on every infinitesimal fluid element is zero, which results in zero current density and a stationary particles' density distribution. According to the Hohenberg-Kohn theorem[5,31] any equilibrium observable, in particular the stress tensor, is a functional of the density n: $P_{\mu\nu}=P_{\mu\nu}[n]$. Hence Eq. (16) is, in fact, the equation of the exact quantum hydrostatics that uniquely determines the density distribution in the equilibrium system. In the semiclassical limit Eq. (16) reduces to the common hydrostatics equation[30] (for a degenerate high-density Fermi gas we recover the Thomas-Fermi theory).

Practical applications of DFT always rely on the KS construction that is a particular consequence of the mapping theorems. The current and the density in the interacting system can be reproduced in a system of noninteracting KS particles, moving in a properly chosen self-consistent potential $U_{\mathrm{S}}=U+U_{\mathrm{xc}}$.[32] The hydrodynamic formulation of TD-

DFT and/or DFT allows us to relate the xc potential $U_{\rm xc}$ to the stress density. Hydrodynamics balance equations for the KS system take the form

$$D_t n + n \frac{\partial}{\partial x^\mu} v_\mu = 0, \tag{17}$$

$$mn D_t v_\mu + \frac{\partial}{\partial x^\nu} T^{\rm S}_{\mu\nu} + n \frac{\partial}{\partial x^\mu} U_{\rm xc} + n \frac{\partial}{\partial x^\mu} U = 0, \tag{18}$$

where the kinetic stress tensor of KS system, $T^{\rm S}_{\mu\nu}$ is given by Eq. (14), but with the averaging over the state of noninteracting particles. Comparing Eqs. (10) and (11) with Eqs. (17) and (18) we find that the velocity **v** and the density n of the noninteracting and the interacting systems coincide if the xc potential $U_{\rm xc}(\mathbf{x}, t)$ satisfies the equation,

$$\frac{\partial U_{\rm xc}}{\partial x^\mu} = \frac{1}{n} \frac{\partial P^{\rm xc}_{\mu\nu}}{\partial x^\nu} \tag{19}$$

where $P^{\rm xc}_{\mu\nu}$ is the xc stress tensor that equals the difference of the stress tensors in the interacting and noninteracting systems with the same velocity distribution,

$$P^{\rm xc}_{\mu\nu} = P_{\mu\nu} - T^{\rm S}_{\mu\nu}. \tag{20}$$

Equations (19) and (20) demonstrate the physical significance of $U_{\rm xc}$. The xc potential should produce a force which compensates for the difference of the internal stress forces in the real interacting system and in the auxiliary noninteracting KS system. By the continuity equation, the density n is a functional of the velocity. Therefore Eq. (19) defines $U_{\rm xc}$ (up to a constant) as a functional of **v**. Equation (19) shows that $n^{-1} \partial_\nu P^{\rm xc}_{\mu\nu}$ is a potential vector. This does not mean that vectors $n^{-1} \partial_\nu P_{\mu\nu}$ and $n^{-1} \partial_\nu T^{\rm S}_{\mu\nu}$, separately, have no rotational components. However, according to the balance equations of Eqs. (11) and (18) the rotational components of these vectors coincide, both being equal to the rotational part of the vector $m D_t \mathbf{v}$.

It is also possible to construct the proper KS system using the xc vector potential $\mathbf{A}^{\rm xc}$ or, in the most general case, a combination of the xc vector and scalar potentials.[10,17,18,33] In this case the exact local conservation laws require that the total xc force, $\boldsymbol{\mathcal{F}}^{\rm xc}$, should compensate for the difference of the stress forces in the interacting and the KS systems,

$$\boldsymbol{\mathcal{F}}^{\rm xc}_\mu = \frac{\partial A^{\rm xc}_\mu}{\partial t} - [\mathbf{v} \times (\nabla \times \mathbf{A}^{\rm xc})]_\mu + \frac{\partial U_{\rm xc}}{\partial x^\mu} = \frac{1}{n} \frac{\partial P^{\rm xc}_{\mu\nu}}{\partial x^\nu}. \tag{21}$$

This equation determines the xc potentials $\mathbf{A}^{\rm xc}$ and $U_{\rm xc}$ up to a gauge transform. Equation (21) represents a very important *exact* property of xc potentials: they produce a force that must be a divergence of a second-rank tensor. This requirement automatically implies the well known zero-force and zero-torque sum rules,[6]

$$\int n \boldsymbol{\mathcal{F}}^{\rm xc} d\mathbf{x} = 0, \quad \int n (\mathbf{x} \times \boldsymbol{\mathcal{F}}^{\rm xc}) d\mathbf{x} = 0. \tag{22}$$

We would like to outline that the exact local condition of Eq. (21) is much stronger than the common integral requirements of Eq. (22). Apparently the above definition of xc potentials

equally well applies both to TDDFT and to the static DFT. It should be mentioned that in the equilibrium case the stress forces in the interacting and KS systems, separately, are potential vectors.

Let us briefly discuss the role of the xc vector potential in DFT. Apparently an appearance of $\mathbf{A}^{\rm xc}$ is unavoidable in the presence of an external magnetic field.[33] Independent of the character of external fields, the formulation in terms of $\mathbf{A}^{\rm xc}$ is convenient in the linear response regime.[17,18] Indeed, in the linearized theory we can perform the Fourier transform in the time domain, which makes $\mathbf{A}_{\rm xc}$ completely local, provided the xc stress tensor is a local functional of some basic variables. In the nonlinear regime this advantage clearly disappears. For a nonlinear evolution the description of xc effects in terms of the scalar potential, defined by Eqs. (23) and (24), looks at least as convenient as the formulation in terms of $\mathbf{A}^{\rm xc}$. Below, for definiteness, we assume the noninteracting v representability of the velocity, which allows us to construct the KS system using only the scalar xc potential. Reformulation of the theory for the xc vector potential is straightforward (see the Conclusion).

For the practical applications it is possibly more convenient to represent the force definition of $U_{\rm xc}$, Eq. (19), in a familiar form of the Poisson equation,

$$\nabla^2 U_{\rm xc}(\mathbf{x}, t) = 4\pi \rho_{\rm xc}(\mathbf{x}, t), \tag{23}$$

where the quantity $\rho_{\rm xc}(\mathbf{x}, t)$,

$$\rho_{\rm xc} = \frac{1}{4\pi} \frac{\partial}{\partial x^\mu} \left(\frac{1}{n} \frac{\partial}{\partial x^\nu} P^{\rm xc}_{\mu\nu} \right), \tag{24}$$

can be interpreted as an xc "charge" density. In this context the xc stress force, $n^{-1} \partial_\nu P^{\rm xc}_{\mu\nu}$, has a clear meaning of an xc "polarization" density. The additional differentiation in Eq. (23) requires an additional boundary condition. The most natural condition, which we should impose on the solution to Eq. (23), is the requirement of boundness at infinity.

Equation (19) or, equivalently, Eqs. (23) and (24) reduce the problem of approximations for $U_{\rm xc}$ to the construction of approximations for the xc stress tensor $P^{\rm xc}_{\mu\nu}$. Since the stress density has a clear physical and microscopic meaning there is a hope that the latter problem is more tractable.

III. STATIC LDA VS TIME-DEPENDENT LDA

Let us first derive the standard static LDA from the force definition of $U_{\rm xc}$, Eq. (19). Formally the static $U^{\rm LDA}_{\rm xc}(\mathbf{x})$ is the solution to Eq. (19) in the lowest order of the gradient expansion. This solution is obtained by inserting $P^{\rm xc}_{\mu\nu}$ for a homogeneous system of the density $n(\mathbf{x})$ into the right-hand side of Eq. (19). In the homogeneous system the stress tensors $P_{\mu\nu}$ and $T^S_{\mu\nu}$ are diagonal,

$$P_{\mu\nu}[n] = \delta_{\mu\nu} P(n), \quad T^S_{\mu\nu}[n] = \delta_{\mu\nu} P_0(n),$$

where P and P_0 are the pressure of the interacting system and the pressure of an ideal gas, respectively. Therefore to the lowest order in the density gradients we get

$$P_{\mu\nu}^{xc}[n](\mathbf{x}) = \delta_{\mu\nu}P_{xc}[n(\mathbf{x})], \qquad (25)$$

where $P_{xc}=P-P_0$ is the xc pressure of the homogeneous system. Substituting Eq. (25) into Eq. (19), and using the common thermodynamic relations, $dP=nd\mu$, $\mu=\partial F/\partial n$, we find the following result for the xc potential:

$$U_{xc}[n](\mathbf{x}) = U_{xc}^{LDA}[n(\mathbf{x})], \quad U_{xc}^{LDA}(n) = \frac{\partial F_{xc}}{\partial n}. \qquad (26)$$

Here F_{xc} is the xc free energy of the homogeneous system. The result of Eq. (26) recovers the standard static LDA.[5]

Physically the above derivation of the static LDA means the following: If the density distribution is a semiclassically slow function in space, every small volume element can be formally considered as an independent, homogeneous many-body system. The density in this homogeneous system equals the density at the location of the element. By solving the homogeneous many-body problem we find the stress tensor, which, after the substitution into Eq. (19) provides us with U_{xc}^{LDA}.

The situation in the time-dependent theory is much more complicated. Even if at any instant t the density distribution $n(\mathbf{x},t)$ is a slow function in space, a small volume element, located at some point $\mathbf{x}$, cannot be considered as a system that is independent of surrounding space. For nonadiabatic dynamics, particles, arriving at the point $\mathbf{x}$ from other regions, bring information about other spatial points. This is the physical reason for the well known nonlocality, immanent to any nonadiabatic TDDFT.[14,15] It is straightforward to demonstrate the failure of any plain attempt to extend the above derivation of the static LDA to the time-dependent case. Indeed, a homogeneous many-body problem, which we would get by formally separating a small volume element, corresponds to an infinite system with a strongly nonconserved number of particles. Apparently this problem is meaningless.

In the rest of this paper we show that the nonlocality problem in TDDFT is resolved by changing a "point of view" on the nonequilibrium many-body system. Any flow in the system can be considered as a collection of small fluid elements moving along their own trajectories. It is possible to divide the system into elements in such a way that the number of particles in every element will be conserved. Indeed, by the proper deformation and rotation of a fluid element one can always adjust its shape to the motion of particles and thus prevent flow through its surface. Let us attach a reference frame to one of those moving elements. The motion of the origin of this frame compensates the translational motion of the fluid element. By properly changing scales and directions of coordinate axes we can also compensate for both the deformations and the rotation. This means that an observer in the new frame will see no currents in the system and a stationary density distribution. Thus from the point of view of the comoving observer the nonequilibrium system looks very similar to the equilibrium one, as it is seen by a stationary observer in the laboratory reference frame. This similarity is of course not complete since particles in the described comoving frame should experience inertia forces. However, the inertia forces are determined only by local geo-

metric characteristics of the frame. The locality of inertia forces and the stationarity of the density allow us to consider a small volume element in the comoving frame as an independent many-body system. Therefore we can extend the derivation of the static LDA to the time-dependent case.

The description of a flow in terms of trajectories of small liquid element is the main idea behind the Lagrangian formulation of classical continuum mechanics.[24] One can show that the transformation to the Lagrangian coordinates exactly corresponds to the transformation to the comoving reference frame. In Sec. IV we apply the general description of quantum many-body dynamics in the Lagrangian frame[12] to the corresponding reformulation of TDDFT.

IV. MANY-BODY THEORY IN THE LAGRANGIAN FRAME AND GEOMETRIC FORMULATION OF TDDFT

A. Many-body problem in the comoving frame

First we briefly review the key results of the many-body theory in the Lagrangian frame (all details and derivations can be found in I). The comoving Lagrangian reference frame is defined as follows: Let $\mathbf{v}(\mathbf{x},t)=\mathbf{j}(\mathbf{x},t)/n(\mathbf{x},t)$ be the velocity of the flow. By solving the following initial value problem:

$$\frac{\partial \mathbf{x}(\boldsymbol{\xi},t)}{\partial t} = \mathbf{v}(\mathbf{x}(\boldsymbol{\xi},t),t), \quad \mathbf{x}(\boldsymbol{\xi},0) = \boldsymbol{\xi}, \qquad (27)$$

we find the function $\mathbf{x}(\boldsymbol{\xi},t)$, which describes the trajectory of a fluid element. The initial point, $\boldsymbol{\xi}$, of the trajectory can be used as a unique label of the element. This initial position of an infinitesimal fluid element is called the Lagrangian coordinate. The transformation from the original $\mathbf{x}$ space to the $\boldsymbol{\xi}$ space of the initial positions is the transformation from the Eulerian to the Lagrangian description of a fluid.[24] On the other hand, the equation $\mathbf{x}=\mathbf{x}(\boldsymbol{\xi},t)$, which maps $\mathbf{x}$ to $\boldsymbol{\xi}$, exactly corresponds to the transformation to the frame, attached to a fluid element. One of the most important characteristics of the Lagrangian frame is Green's deformation tensor,[24] $g_{\mu\nu}(\boldsymbol{\xi},t)$,

$$g_{\mu\nu} = \frac{\partial x^\alpha}{\partial \xi^\mu}\frac{\partial x^\alpha}{\partial \xi^\nu}, \quad g^{\mu\nu} = \frac{\partial \xi^\mu}{\partial x^\alpha}\frac{\partial \xi^\nu}{\partial x^\alpha}. \qquad (28)$$

Tensor $g_{\mu\nu}$ plays the role of metric in the Lagrangian $\boldsymbol{\xi}$ space. (We will use the notation g for the determinant of $g_{\mu\nu}$.) It has been shown in I that the field operators $\widetilde{\psi}(\boldsymbol{\xi},t)$ in the Lagrangian frame are related to the field operators $\psi(\mathbf{x},t)$ in the laboratory frame as follows:

$$\widetilde{\psi}(\boldsymbol{\xi},t) = g^{1/4}\psi(\mathbf{x}(\boldsymbol{\xi},t),t).$$

Apparently the operators $\widetilde{\psi}(\boldsymbol{\xi},t)$ satisfy the common equal-time commutation relations, which is guaranteed by the factor $g^{1/4}$ in their definition. The current operator $\widetilde{j}^\mu(\boldsymbol{\xi},t)$ and the density operator $\hat{\tilde{n}}(\boldsymbol{\xi},t)$ in the Lagrangian frame are defined by the following expressions:

$$\hat{\tilde{n}}(\boldsymbol{\xi},t) = \widetilde{\psi}^\dagger(\boldsymbol{\xi},t)\widetilde{\psi}(\boldsymbol{\xi},t), \qquad (29)$$

$$\widetilde{\widehat{j}^{\mu}}(\boldsymbol{\xi},t) = g^{\mu\nu}\left[\frac{-i}{2m}\left(\widetilde{\psi}^{\dagger}\frac{\partial\widetilde{\psi}}{\partial\xi^{\nu}} - \frac{\partial\widetilde{\psi}^{\dagger}}{\partial\xi^{\nu}}\widetilde{\psi}\right) - \widetilde{v}_{\nu}\widetilde{\psi}^{\dagger}\widetilde{\psi}\right], \quad (30)$$

where $\widetilde{v}_{\nu} = g_{\nu\mu}\widetilde{v}^{\mu}$ is the covariant component of the velocity vector **v**, transformed to the new frame,

$$\widetilde{v}^{\mu}(\boldsymbol{\xi},t) = \frac{\partial\xi^{\mu}}{\partial x^{\nu}}v^{\nu}(\mathbf{x}(\boldsymbol{\xi},t),t).$$

The Heisenberg equation of motion for the density operator of Eq. (29) takes a form of the operator continuity equation,

$$\frac{\partial\widehat{\widetilde{n}}}{\partial t} + \frac{\partial\widetilde{\widehat{j}^{\mu}}}{\partial\xi^{\mu}} = 0. \quad (31)$$

On the level of expectation values Eq. (31) is trivially satisfied. One can check by explicit calculations that the expectation value of the current operator, Eq. (30), is zero, while the expectation value of the density operator, Eq. (29), is time independent,

$$\widetilde{j}^{\mu}(\boldsymbol{\xi},t) = \langle\widetilde{\widehat{j}^{\mu}}(\boldsymbol{\xi},t)\rangle = 0, \quad (32)$$

$$\widetilde{n}(\boldsymbol{\xi},t) = \langle\widehat{\widetilde{n}}(\boldsymbol{\xi},t)\rangle = \widetilde{n}(\boldsymbol{\xi},0) = n_0(\boldsymbol{\xi}), \quad (33)$$

where $n_0(\mathbf{x})$ is the initial density distribution. Equations (32) and (33) are in complete agreement with the qualitative discussion in Sec. III.

According to the results of the paper I, the vector $m\widetilde{v}_{\mu}(\boldsymbol{\xi},t)$ plays the role of an effective vector potential in the equation of motion for the field operator, $\widetilde{\psi}$. In general the velocity vector $\widetilde{v}_{\mu}(\boldsymbol{\xi},t)$ has both potential (longitudinal) and rotational (transverse) parts. The potential part of a vector potential can be always removed from the kinetic energy operator by the gauge transformation. Therefore it is convenient to separate explicitly the potential part, $\widetilde{v}_{L\mu} = \partial_{\mu}\varphi$, of the vector $\widetilde{v}_{\mu}$,

$$\widetilde{v}_{\mu} = \frac{\partial\varphi}{\partial\xi^{\mu}} + \widetilde{v}_{T\mu}, \quad (34)$$

where $\widetilde{v}_{T\mu}$ is the transverse part of $\widetilde{v}_{\mu}$. Performing the gauge transformation $\widetilde{\psi} = e^{im\,\varphi}\widetilde{\psi}'$ in the equation of motion for $\widetilde{\psi}$ [see Eq. (34) in I] we obtain the following equation of motion for the transformed operator $\widetilde{\psi}'$:

$$i\frac{\partial\widetilde{\psi}'(\boldsymbol{\xi})}{\partial t} = g^{-1/4}\frac{\hat{K}_{\mu}\sqrt{g}\hat{K}^{\mu}}{2m}g^{-1/4}\widetilde{\psi}'(\boldsymbol{\xi})$$

$$+ \int d\boldsymbol{\xi}'w(l_{\boldsymbol{\xi},\boldsymbol{\xi}'})\Delta\widehat{\widetilde{n}}(\boldsymbol{\xi}')\widetilde{\psi}'(\boldsymbol{\xi})$$

$$+ \left(m\frac{\partial\varphi}{\partial t} + U - m\frac{\widetilde{v}_{\mu}\widetilde{v}^{\mu}}{2}\right)\widetilde{\psi}'(\boldsymbol{\xi}), \quad (35)$$

where $\Delta\widehat{\widetilde{n}}(\boldsymbol{\xi},t) = \widehat{\widetilde{n}}(\boldsymbol{\xi},t) - \widetilde{n}(\boldsymbol{\xi},t)$. (The Hartree term is included in $U = U_{\text{ext}} + U_{\text{H}}$.) Other notations in Eq. (35) are the same as in I,

$$\hat{K}_{\mu} = -i\frac{\partial}{\partial\xi^{\mu}} - m\widetilde{v}_{T\mu}$$

is the operator of kinematic momentum, and $l_{\boldsymbol{\xi},\boldsymbol{\xi}'}$ is the length of geodesic connecting points $\boldsymbol{\xi}$ and $\boldsymbol{\xi}'$. (Everywhere the rising and lowering of tensor indexes are performed according to the standard rules, i.e., $A_{\mu} = g_{\mu\nu}A^{\nu}$, etc.) The deformation tensor and the velocity vector in Eq. (35) describe generalized inertial forces in the local noninertial reference frame. Tensor $g_{\mu\nu}$ in the kinetic energy term produces the "geodesic" force. This inertia force is responsible for the motion of a free particle along the geodesic in $\boldsymbol{\xi}$ space. The velocity $\widetilde{v}_{\mu}$, which acts as a vector potential in Eq. (35), produces the Coriolis force (an effective Lorentz force) and the linear acceleration force (an effective electric field). The last term in the brackets in Eq. (35) is responsible for the inertia force that is related to the kinetic energy of the frame (an analog of the centrifugal force).

Equation (35) is the equation of motion in a reference frame moving with some velocity **v**. In fact, the form of Eq. (35) is covariant under an arbitrary transformation of coordinates, which is generated by a continuous, vector-valued function $\mathbf{v}(\mathbf{x},t)$ via Eq. (27). The function is (i) continuous, and (ii) vector valued (or vector-valued). To specify a particular reference frame we need to impose an additional "gauge" condition. The gauge condition assigns a particular value to the generating function $\mathbf{v}(\mathbf{x},t)$. There are a few formal possibilities to specify the comoving Lagrangian frame (see I). For example, since the expectation value of the current operator in the Lagrangian frame should be zero, we can impose the condition of Eq. (32) on the solutions to the equation of motion, Eq. (35). In the present paper we prefer to use another gauge-fixing condition. Namely, we require that the solution to the equation of motion, Eq. (35), be consistent with the equation of the local force balance in the Lagrangian frame,

$$m\frac{\partial\widetilde{v}_{T\mu}}{\partial t} + \frac{\partial}{\partial\xi^{\mu}}\left(m\frac{\partial\varphi}{\partial t} + U - m\frac{\widetilde{v}_{\nu}\widetilde{v}^{\nu}}{2}\right) + \frac{\sqrt{g}}{n_0}\widetilde{P}^{\nu}_{\mu;\nu} = 0, \quad (36)$$

where $\widetilde{P}^{\nu}_{\mu;\nu}$ is the covariant divergence[34,35] of the stress tensor,

$$\widetilde{P}^{\nu}_{\mu;\nu} = \frac{1}{\sqrt{g}}\frac{\partial\sqrt{g}\widetilde{P}^{\nu}_{\mu}}{\partial\xi^{\nu}} - \frac{1}{2}\frac{\partial g_{\alpha\beta}}{\partial\xi^{\mu}}\widetilde{P}^{\alpha\beta}. \quad (37)$$

The force-balance equation of Eq. (36) corresponds to the local momentum conservation law, Eq. (11), transformed to the Lagrangian frame. The stress tensor in the Lagrangian frame $\widetilde{P}_{\mu\nu}$ which enters Eq. (36), is a linear functional of the one-particle density matrix $\widetilde{\rho}_1$ and of the pair correlation function $\widetilde{G}_2$,

$$\widetilde{P}_{\mu\nu} = \widetilde{P}_{\mu\nu}[\widetilde{\rho}_1,\widetilde{G}_2](\boldsymbol{\xi},t). \quad (38)$$

The explicit microscopic form of the functional $\widetilde{P}_{\mu\nu}[\widetilde{\rho}_1,\widetilde{G}_2]$ is presented in Appendix A [see Eqs. (A1), (A4), and (A5)].

Equation (36) has precisely the same physical significance as the static force balance equation of Eq. (16). It shows that the inertia forces exactly compensate for the external force, $(\partial/\partial\xi^\mu)U$, and the force of internal stress, $(\sqrt{g}/n_0)\widetilde{P}^\nu_{\mu;\nu}$. The result of this compensation is the absence of the current and stationary density distributions in the Lagrangian frame.

Equations (35) and (36) constitute the full set of equations of quantum many-body theory in the Lagrangian frame.

B. TDDFT in the Lagrangian frame: Stress tensor as a universal functional of the dynamic deformation

Now we are ready to resume the discussion of TDDFT. The complete description of many-body dynamics in the Lagrangian frame corresponds to the solution of the equation of motion, Eq. (35), supplemented by the frame-fixing condition of Eq. (36). Let us note that both Eqs. (35) and (36) contain the same effective potential [the term in the parentheses in Eqs. (35) and (36)]. Using this simple property we can formulate the following two-step procedure for solving the system of Eqs. (35) and (36). In the first step we solve a universal nonlinear many-body problem of the form,

$$i\frac{\partial\widetilde{\psi}'(\boldsymbol{\xi})}{\partial t} = g^{-1/4}\frac{\hat{K}_\mu\sqrt{g}\hat{K}^\mu}{2m}g^{-1/4}\widetilde{\psi}'(\boldsymbol{\xi}) + \left[\int d\boldsymbol{\xi}'w(l_{\boldsymbol{\xi},\boldsymbol{\xi}'})\Delta\hat{\tilde{n}}(\boldsymbol{\xi}')\right.$$
$$\left. + U_{\text{s-c}}(\boldsymbol{\xi},t)\right]\widetilde{\psi}'(\boldsymbol{\xi}), \tag{39}$$

where the effective potential $U_{\text{s-c}}(\boldsymbol{\xi},t)$ is the solution to the following self-consistency equation:

$$-\frac{\partial}{\partial\xi^\mu}U_{\text{s-c}}(\boldsymbol{\xi},t) = \frac{\sqrt{g}}{n_0}\widetilde{P}^\nu_{\mu;\nu}[\widetilde{\rho}_1,\widetilde{G}_2] + m\frac{\partial\widetilde{v}_{T\mu}}{\partial t}. \tag{40}$$

The initial conditions for Eqs. (39) and (40) are the same as in the original physical many-body problem, Eqs. (35) and (36). The special form of the self-consistency equation, Eq. (40), ensures the stationarity of the particle density and zero-current density. Indeed, using Eqs. (39) and (40) we obtain the following equations of motion for the density $\widetilde{n}$ and for the current $\widetilde{\mathbf{j}}$:

$$\frac{\partial\widetilde{n}}{\partial t} + \frac{\partial\widetilde{j}^\mu}{\partial\xi^\mu} = 0, \tag{41}$$

$$\frac{\partial\widetilde{j}_\mu}{\partial t} + \widetilde{F}_{\mu\nu}\widetilde{j}^\nu = 0, \tag{42}$$

where $\widetilde{F}_{\mu\nu}$ is the skew-symmetric vorticity tensor that plays the role of an effective magnetic field,

$$\widetilde{F}_{\mu\nu} = \frac{\partial\widetilde{v}_{T\mu}}{\partial\xi^\nu} - \frac{\partial\widetilde{v}_{T\nu}}{\partial\xi^\mu} = \frac{\partial\widetilde{v}_\mu}{\partial\xi^\nu} - \frac{\partial\widetilde{v}_\nu}{\partial\xi^\mu}. \tag{43}$$

The cancellation of the "external" force $\nabla U_{\text{s-c}}$ and the inertial and the stress forces in Eq. (42) is a consequence of the self-consistency equation, Eq. (40). By solving Eqs. (41) and (42) with the initial conditions $\widetilde{\mathbf{j}}(\boldsymbol{\xi},0)=0$ and $\widetilde{n}(\boldsymbol{\xi},0)=n_0(\boldsymbol{\xi})$,

we indeed confirm that for all $t>0$ $\widetilde{\mathbf{j}}(\boldsymbol{\xi},t)=0$ and $\widetilde{n}(\boldsymbol{\xi},t)=n_0(\boldsymbol{\xi})$.

The self-consistent nonlinear problem of Eqs. (39) and (40) is universal in the sense that no external potential enters the equations. The only external variables in Eqs. (39) and (40) are the deformation tensor $g_{\mu\nu}(\boldsymbol{\xi},t)$ (an effective metric) and the transverse part of the velocity, $\widetilde{v}_{T\mu}(\boldsymbol{\xi},t)$ (an effective vector potential). The vector $\widetilde{v}_{T\mu}$ is uniquely determined by the skew-symmetric vorticity tensor $\widetilde{F}_{\mu\nu}$ (an effective magnetic field), Eq. (43). Therefore by solving the nonlinear problem of Eqs. (39) and (40) we find the many-body density matrix to be a functional of two basic variables, $g_{\mu\nu}(\boldsymbol{\xi},t)$ and $\widetilde{F}_{\mu\nu}(\boldsymbol{\xi},t)$.[36] Inserting this density matrix into the microscopic expression for the stress tensor, Eq. (38), we obtain the universal functional,

$$\widetilde{P}_{\mu\nu} = \widetilde{P}_{\mu\nu}[g_{\mu\nu},\widetilde{F}_{\mu\nu}](\boldsymbol{\xi},t). \tag{44}$$

Calculation of the stress tensor functional, Eq. (44), completes the first step in the solution of the original many-body problem.

The symmetric Green's deformation tensor $g_{\mu\nu}$ and the skew-symmetric vorticity tensor $\widetilde{F}_{\mu\nu}$ completely characterize the deformed state of a fluid in the Lagrangian description. Therefore Eq. (44) can be interpreted as the exact nonequilibrium "equation of state" that relates the stress tensor to the dynamic deformation in the system. Since $g_{\mu\nu}$ and $\widetilde{F}_{\mu\nu}$ are the functionals of velocity, the stress tensor of Eq. (44) is also a functional of velocity in agreement with the Runge-Gross theorem. However, the present interpretation of $\widetilde{P}_{\mu\nu}$ as a deformation functional looks more natural physically.

Substituting the equation of state, Eq. (44), into the force balance equation of Eq. (36), we get the exact quantum Navier-Stokes equation in the Lagrangian formulation. The full set of exact hydrodynamics equations includes Eq. (36) and the trajectory equation, Eq. (27). The solution of the system of Eqs. (27) and (36) corresponds to the second step in the solution of the original many-body problem. In this step we determine the evolution of velocity for a given external potential. Equations (27) and (36) with the stress tensor of Eq. (44) correspond to the exact TDDFT hydrodynamics in the Lagrangian formulation of continuum mechanics.

The KS formulation of TDDFT requires a knowledge of the xc potential U_{xc}. In Sec. II we have shown that U_{xc} is related to the xc stress tensor $\widetilde{P}^{\text{xc}}_{\mu\nu} = \widetilde{P}_{\mu\nu} - \widetilde{T}^{\text{S}}_{\mu\nu}$, where $\widetilde{T}^{\text{S}}_{\mu\nu}$ is the stress tensor for the noninteracting KS system. Obviously, the KS stress tensor can be found from the solution of a nonlinear noninteracting problem that corresponds to Eqs. (39) and (40) with $w(l_{\boldsymbol{\xi},\boldsymbol{\xi}'})=0$. Hence by solving Eqs. (39) and (40) with and without interaction we compute $\widetilde{P}_{\mu\nu}$ and $\widetilde{T}^{\text{S}}_{\mu\nu}$, respectively. The difference of these tensors gives us the xc stress tensor in the Lagrangian frame as a functional of $g_{\mu\nu}$ and $\widetilde{F}_{\mu\nu}$,

$$\widetilde{P}^{\text{xc}}_{\mu\nu} = \widetilde{P}^{\text{xc}}_{\mu\nu}[g_{\mu\nu},\widetilde{F}_{\mu\nu}](\boldsymbol{\xi},t). \tag{45}$$

Transforming the xc stress tensor of Eq. (45) back to the laboratory frame and substituting the result into Eqs. (23)

and (24), we obtain the equation for the xc potential $U_{xc}(\mathbf{x}, t)$. Another possibility is to determine the xc potential, $\widetilde{U}_{xc}(\boldsymbol{\xi}, t)$, directly in the Lagrangian frame by solving the following equation:

$$\frac{\partial}{\partial \xi^\mu} \widetilde{U}_{xc}(\boldsymbol{\xi}, t) = \frac{\sqrt{g}}{n_0} \widetilde{P}^{xc\nu}_{\mu;\nu}. \qquad (46)$$

The transformation of $\widetilde{U}_{xc}(\boldsymbol{\xi}, t)$ to the laboratory frame corresponds to the following replacement $\boldsymbol{\xi} \to \boldsymbol{\xi}(\mathbf{x}, t)$, i.e.,

$$U_{xc}(\mathbf{x}, t) = \widetilde{U}_{xc}[g_{\mu\nu}, \widetilde{F}_{\mu\nu}][\boldsymbol{\xi}(\mathbf{x}, t), t], \qquad (47)$$

where $\widetilde{U}_{xc}$ is the solution to Eq. (46).

Let us note that the problem of calculation of the equilibrium stress tensor functional, $P_{\mu\nu}$, in the static DFT can be formulated in exactly the same fashion. To calculate $P_{\mu\nu}[n] \times (\mathbf{x})$ we need to find the equilibrium solution to the following universal, nonlinear many-body problem:

$$i \frac{\partial}{\partial t} \psi(\mathbf{x}) = -\frac{\nabla^2}{2m} \psi(\mathbf{x}) + U_{s\text{-}c}(\mathbf{x}) \psi(\mathbf{x})$$
$$+ \int d\mathbf{x}' w(|\mathbf{x} - \mathbf{x}'|) \Delta \hat{n}(\mathbf{x}') \psi(\mathbf{x}), \qquad (48)$$

$$\frac{\partial}{\partial x^\mu} U_{s\text{-}c}(\mathbf{x}) = \frac{1}{n} \frac{\partial}{\partial x^\nu} P_{\mu\nu}[\rho_1, G_2], \qquad (49)$$

where $P_{\mu\nu}[\rho_1, G_2] = T_{\mu\nu}[\rho_1] + W_{\mu\nu}[G_2]$ is defined after Eqs. (14) and (15). For a given density $n(\mathbf{x})$ the equilibrium solution to Eqs. (48) and (49) defines the stress tensor $P_{\mu\nu}$ as a universal functional of n.

The stationarity of the density in the Lagrangian frame makes the dynamic universal problem of Eqs. (39) and (40) to a large extent similar to the equilibrium universal problem of Eqs. (48) and (49). In Sec. V we use this similarity to derive a local nonadiabatic approximation in TDDFT.

V. TIME-DEPENDENT LOCAL DEFORMATION APPROXIMATION

In the Sec. IV we have shown that the calculation of the xc stress tensor, which defines the xc potential, reduces to the solution of the nonlinear, universal many-body problem, Eqs. (39) and (40). Obviously, it is not possible to solve this problem exactly. However, one can try to find an approximate solution by a perturbative expansion in terms of a small parameter. Below we construct a local approximation that corresponds to the lowest order in the gradients of basic variables (i.e., the density in the static DFT and the deformation tensor in TDDFT).

A. General formulation of a nonadiabatic local approximation

1. Preliminaries: Derivation of the static LDA

To illustrate the general procedure we start again with the familiar case of the equilibrium theory. The problem is to find the equilibrium solution to Eqs. (48) and (49), assuming

that the gradients of the density n are vanishingly small. In the limit $\nabla n \to 0$ the spatial derivatives of the stress tensor also vanish. Hence, to the lowest order in the density gradients the solution to the self-consistency Eq. (49) takes a trivial form, $U_{s\text{-}c}(\mathbf{x}) = C$, where C is a constant. Therefore the many-body equation of motion, Eq. (48), simplifies as follows:

$$i \frac{\partial \psi(\mathbf{x})}{\partial t} = -\frac{\nabla^2}{2m} \psi(\mathbf{x}) + \int d\mathbf{x}' w(|\mathbf{x} - \mathbf{x}'|) \hat{n}(\mathbf{x}') \psi(\mathbf{x}). \quad (50)$$

Thus the nonlinear problem of Eqs. (48) and (49) reduces to the usual linear many-body problem for a homogeneous equilibrium system with a given density n. Substituting the equilibrium solution to Eq. (50) into Eqs. (13)–(15), we compute the stress tensor, $P_{\mu\nu}(n) = \delta_{\mu\nu} P(n)$, where P is the pressure of the homogeneous system,

$$P(n) = \frac{2}{d} E_{kin} - \frac{1}{2d} \int \mathbf{x} \frac{\partial w(|\mathbf{x}|)}{\partial \mathbf{x}} G_2^{eq}(|\mathbf{x}|) d\mathbf{x}. \qquad (51)$$

Here d is the number of spatial dimensions, E_{kin} is the kinetic energy per unit volume, and $G_2^{eq}(|\mathbf{x}|)$ is the pair correlation function of the equilibrium homogeneous system. Similarly, by solving the homogeneous noninteracting problem we find the KS stress tensor $T_{\mu\nu}^S(n) = \delta_{\mu\nu} 2/d E_{kin}^{(0)}$, where $E_{kin}^{(0)}$ is the kinetic energy of an ideal Fermi gas. Substituting $P_{\mu\nu}[n(\mathbf{x})]$ and $T_{\mu\nu}^S[n(\mathbf{x})]$ into Eqs. (20) and (19), we recover the common static LDA (see Sec. III).

2. Basic equations of TDLDA: The homogeneous many-body problem

The above procedure allows for a straightforward extension to the time-dependent problem. Let us assume that the characteristic length scale L of the deformation inhomogeneity goes to infinity. In this limit the vector $(\sqrt{g}/n_0)\widetilde{P}^\nu_{\mu;\nu}$ in the right-hand side of Eq. (40) vanishes. Therefore to the lowest order in $1/L \to 0$ the self-consistent solution to Eq. (40) takes the form, $U_{s\text{-}c}(\mathbf{x}, t) = C(t)$ and $\mathbf{v}_T = 0$. Substituting this solution into Eq. (39), we get the equation of motion for the $\widetilde{\psi}$ operator,

$$i \frac{\partial \widetilde{\psi}(\boldsymbol{\xi})}{\partial t} = -\frac{g_{\mu\nu}(t)}{2m} \frac{\partial^2 \widetilde{\psi}(\boldsymbol{\xi})}{\partial \xi^\mu \partial \xi^\nu} + \int d\boldsymbol{\xi}' w(\|\boldsymbol{\xi} - \boldsymbol{\xi}'\|) \hat{n}(\boldsymbol{\xi}') \widetilde{\psi}(\boldsymbol{\xi}),$$
$$(52)$$

where $\|\boldsymbol{\xi} - \boldsymbol{\xi}'\| = l_{\xi, \xi'}$ is the length of the geodesic in a homogeneously deformed Lagrangian space (see Appendix B),

$$\|\boldsymbol{\xi} - \boldsymbol{\xi}'\| = \sqrt{g_{\mu\nu}(t)(\xi^\mu - \xi'^\mu)(\xi^\nu - \xi'^\nu)}. \qquad (53)$$

Equation (52) corresponds to a homogeneous many-body system. It is more natural to reformulate this homogeneous problem using the momentum representation for field operators,

$$\widetilde{\psi}(\boldsymbol{\xi}) = \sum_{\mathbf{k}} e^{ik_\mu \xi^\mu} \widetilde{a}_{\mathbf{k}}. \qquad (54)$$

The equation of motion for annihilation operator, $\widetilde{a}_{\mathbf{k}}$, takes the form,

$$i\frac{\partial \widetilde{a}_{\mathbf{k}}}{\partial t} = g^{\mu\nu}(t)\frac{k_\mu k_\nu}{2m}\widetilde{a}_{\mathbf{k}} + \sum_{\mathbf{p},\mathbf{q}} \frac{\overline{w}(\|\mathbf{q}\|)}{\sqrt{g(t)}}\widetilde{a}_{\mathbf{p}}^\dagger \widetilde{a}_{\mathbf{p}+\mathbf{q}}\widetilde{a}_{\mathbf{k}-\mathbf{q}} \quad (55)$$

where $\overline{w}(q)$ is the Fourier component of the interaction potential, and

$$\|\mathbf{q}\| = \sqrt{g^{\alpha\beta}(t)q_\beta q_\alpha} \quad (56)$$

is the norm of the wave vector in the deformed momentum space. Equation (55) corresponds to the following Hamiltonian:

$$\widetilde{H} = \sum_{\mathbf{k}} g^{\mu\nu}\frac{k_\mu k_\nu}{2m}\widetilde{a}_{\mathbf{k}}^\dagger \widetilde{a}_{\mathbf{k}} + \frac{1}{2\sqrt{g}}\sum_{\mathbf{k},\mathbf{q}} \overline{w}(\|\mathbf{q}\|)\widetilde{a}_{\mathbf{k}}^\dagger \hat{\widetilde{n}}_{\mathbf{q}}\widetilde{a}_{\mathbf{k}-\mathbf{q}}, \quad (57)$$

where $\hat{\widetilde{n}}_{\mathbf{q}} = \sum_{\mathbf{p}}\widetilde{a}_{\mathbf{p}}^\dagger \widetilde{a}_{\mathbf{p}+\mathbf{q}}$ is the density operator in the momentum representation. In Appendix B we show that to the lowest order in the deformation gradients the microscopic expression for the stress tensor $\widetilde{P}_{\mu\nu}$ simplifies as follows:

$$\widetilde{P}_{\mu\nu} = \frac{1}{\sqrt{g}}\sum_{\mathbf{k}} \frac{k_\mu k_\nu}{m}\widetilde{f}(\mathbf{k})$$
$$+ \frac{1}{2g}\sum_{\mathbf{k}}\left[\frac{k_\mu k_\nu}{\|\mathbf{k}\|}\overline{w}'(\|\mathbf{k}\|) + g_{\mu\nu}\overline{w}(\|\mathbf{k}\|)\right]\widetilde{G}_2(\mathbf{k}), \quad (58)$$

where $\widetilde{f}(\mathbf{k}) = \langle \widetilde{a}_{\mathbf{k}}^\dagger \widetilde{a}_{\mathbf{k}}\rangle$ is the Wigner function, $\widetilde{G}_2(\mathbf{k})$ is the Fourier component of the pair correlation function, and $\overline{w}'(x) = d\overline{w}(x)/dx$.

It should be mentioned that Eq. (58) can be derived directly from the "geometric" definition of the stress tensor (see I and Ref. 37),

$$\widetilde{P}_{\mu\nu} = \frac{2}{\sqrt{g}}\left\langle \frac{\delta \widetilde{H}}{\delta g^{\mu\nu}}\right\rangle.$$

Indeed, using the relation $\delta g = -gg_{\mu\nu}\delta g^{\mu\nu}$, and computing the derivative of the Hamiltonian, Eq. (57), with respect to $g^{\mu\nu}$, we immediately recover Eq. (58).

The Hamiltonian of Eq. (57) determines the homogeneous problem which we need to solve for the derivation of a local approximation in TDDFT. This problem corresponds to a system of particles in a small volume located at the point $\boldsymbol{\xi}$ of Lagrangian space. The density of particles is time independent and equal to the initial density, $n_0(\boldsymbol{\xi})$, at that point. (Obviously, the operator of the number of particles commutes with $\widetilde{H}$.) The behavior of the system is governed by the local value of the deformation tensor, $g_{\mu\nu}(\boldsymbol{\xi},t)$. By solving the equations of motion we find the Wigner function $\widetilde{f}(\mathbf{k},t)$ and the pair correlation function $\widetilde{G}_2(\mathbf{k},t)$. Substitution of $\widetilde{f}(\mathbf{k},t)$ and $\widetilde{G}_2(\mathbf{k},t)$ into Eq. (58) yields the stress tensor functional $\widetilde{P}_{\mu\nu}[g_{\mu\nu}(\boldsymbol{\xi},t),n_0(\boldsymbol{\xi})]$. By the repetition of the above procedure for the noninteracting system [Eqs. (57) and (58) with $\overline{w}=0$] we find the KS stress tensor, $\widetilde{T}_{\mu\nu}^S[g_{\mu\nu}(\boldsymbol{\xi},t),n_0(\boldsymbol{\xi})]$, and, finally, the xc stress tensor,

$$\widetilde{P}_{\mu\nu}^{\mathrm{xc}}[g_{\mu\nu}(\boldsymbol{\xi},t),n_0(\boldsymbol{\xi})] = \widetilde{P}_{\mu\nu} - \widetilde{T}_{\mu\nu}^S. \quad (59)$$

Substituting $\widetilde{P}_{\mu\nu}^{\mathrm{xc}}$ of Eq. (59) into Eq. (46) we determine the corresponding xc potential in the Lagrangian frame.

The xc stress tensor $\widetilde{P}_{\mu\nu}^{\mathrm{xc}}$, Eq. (59), is a local in the space functional of the deformation tensor. (It should be noted that in general this functional is nonlocal in time.) In what follows, the approximation of Eq. (59) will be referred to as a time-dependent local deformation approximation (TDLDA).

The construction of TDLDA reduces to the solution of the homogeneous many-body problem. In this respect the situation is similar to the static case. However, the homogeneous time-dependent problem, defined by the Hamiltonian of Eq. (57), is still too complicated to be solved exactly. Indeed the operator equation of motion, Eq. (55), generates an infinite set of coupled evolution equations [Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) hierarchy[38]] for correlation functions. The first equation of this hierarchy is the equation of motion for the Wigner function,

$$i\frac{\partial \widetilde{f}(\mathbf{k})}{\partial t} = \sum_{\mathbf{p},\mathbf{q}} \frac{\overline{w}(\|\mathbf{q}\|)}{\sqrt{g}}\langle \widetilde{a}_{\mathbf{p}}^\dagger(\widetilde{a}_{\mathbf{k}}^\dagger \widetilde{a}_{\mathbf{k}-\mathbf{q}} - \widetilde{a}_{\mathbf{k}+\mathbf{q}}^\dagger \widetilde{a}_{\mathbf{k}})\widetilde{a}_{\mathbf{p}+\mathbf{q}}\rangle. \quad (60)$$

An equation for the four-fermion correlator, entering the right-hand side in Eq. (60), couples to the six-fermion correlation functions, etc. However, the homogeneity of the problem and a very specific form of the "driving force" in the equations of motion allow us to construct reasonable approximate xc functionals (see the next sections).

3. Stress tensor of the noninteracting KS system

A necessary step in the derivation of TDLDA is to compute the stress tensor, $\widetilde{T}_{\mu\nu}^S$, in the noninteracting system. This problem can be solved exactly. In the noninteracting case ($\overline{w}=0$) Eqs. (58) and (60) reduce to the following simple forms:

$$\widetilde{T}_{\mu\nu}^S = \frac{1}{\sqrt{g}}\sum_{\mathbf{k}} \frac{k_\mu k_\nu}{m}\widetilde{f}(\mathbf{k},t), \quad (61)$$

$$\frac{\partial}{\partial t}\widetilde{f}(\mathbf{k},t) = 0. \quad (62)$$

Equation (62) shows that the distribution function of noninteracting particles in the Lagrangian frame is time independent. Let us assume for definiteness that the system evolves from the equilibrium state, i.e., $\widetilde{f}(\mathbf{k},0) = n_{\mathbf{k}}^F$, where $n_{\mathbf{k}}^F$ is the Fermi function. In this case the solution to Eq. (62) takes the form

$$\widetilde{f}(\mathbf{k},t) = \widetilde{f}(\mathbf{k},0) = n_{\mathbf{k}}^F. \quad (63)$$

Substituting Eq. (63) into Eq. (61) we get the kinetic stress tensor of the KS system in the Lagrangian frame,

$$\widetilde{T}_{\mu\nu}^S(\boldsymbol{\xi},t) = \frac{\delta_{\mu\nu}}{\sqrt{g(\boldsymbol{\xi},t)}}P_0[n_0(\boldsymbol{\xi})], \quad (64)$$

where the function $P_0(n) = (2/d)E_{\mathrm{kin}}^{(0)}(n)$ is the equilibrium kinetic pressure of a noninteracting homogeneous Fermi gas.

For the practical calculation of xc potential in TDLDA [see Eqs. (19) and (20)] we need the stress tensor $T^S_{\mu\nu}(\mathbf{x},t)$ in the laboratory frame. Application of the common tensor transformation rules,[34]

$$P_{\mu\nu}(\mathbf{x},t) = \frac{\partial\xi^\alpha}{\partial x^\mu}\frac{\partial\xi^\beta}{\partial x^\nu}\widetilde{P}_{\alpha\beta}(\boldsymbol{\xi}(\mathbf{x},t),t), \qquad (65)$$

to the stress tensor of Eq. (64) yields the result,

$$T^S_{\mu\nu}(\mathbf{x},t) = \bar{g}_{\mu\nu}(\mathbf{x},t)\sqrt{\bar{g}(\mathbf{x},t)}P_0[n_0(\boldsymbol{\xi}(\mathbf{x},t))], \qquad (66)$$

where $\bar{g}_{\mu\nu}(\mathbf{x},t)$ is the Cauchy's deformation tensor[24]

$$\bar{g}_{\mu\nu}(\mathbf{x},t) = \frac{\partial\xi^\alpha}{\partial x^\mu}\frac{\partial\xi^\alpha}{\partial x^\nu}. \qquad (67)$$

The determinant $\bar{g}(\mathbf{x},t)$ of Cauchy's deformation tensor, Eq. (67), is related to the determinant $g(\boldsymbol{\xi},t)$ of Green's deformation tensor, Eq. (28), as follows:

$$\bar{g}(\mathbf{x},t) = g^{-1}(\boldsymbol{\xi}(\mathbf{x},t),t). \qquad (68)$$

Equation (66) clearly demonstrates an extreme nonlocality which is related to the memory effects. The stress tensor $T^S_{\mu\nu}(\mathbf{x},t)$ at a given point $\mathbf{x}$ depends on the initial density at the point $\boldsymbol{\xi}(\mathbf{x},t)$ that is the initial position of a fluid element presently at $\mathbf{x}$. Let us show that this dependence on the delayed position can be represented in a local form. By definition of the Lagrangian coordinate, the density $n(\mathbf{x},t)$ in the laboratory frame can be expressed in terms of the initial density distribution (the density in the Lagrangian frame),

$$n(\mathbf{x},t) = \frac{n_0(\boldsymbol{\xi}(\mathbf{x},t))}{\sqrt{g(\boldsymbol{\xi}(\mathbf{x},t),t)}}. \qquad (69)$$

Using the relation of Eq. (68) we can represent the nonlocal quantity $n_0(\boldsymbol{\xi}(\mathbf{x},t))$ in the following form:

$$n_0(\boldsymbol{\xi}(\mathbf{x},t)) = \frac{n(\mathbf{x},t)}{\sqrt{\bar{g}(\mathbf{x},t)}}. \qquad (70)$$

Substituting Eq. (70) into Eq. (66), we obtain a completely local representation for the KS kinetic stress tensor,

$$T^S_{\mu\nu}(\mathbf{x},t) = \bar{g}_{\mu\nu}(\mathbf{x},t)\sqrt{\bar{g}(\mathbf{x},t)}P_0\left(\frac{n(\mathbf{x},t)}{\sqrt{\bar{g}(\mathbf{x},t)}}\right). \qquad (71)$$

The nonlocality of the stress tensor in the form of Eq. (66) is now hidden in the space-time dependence of the function $\bar{g}(\mathbf{x},t)$.

B. Exchange-only TDLDA

The most difficult part in the derivation of an explicit TDLDA is the solution of the interacting problem defined by the Hamiltonian of Eq. (57). In this section we find the exact solution of this problem in the exchange approximation, which provides us with the x-only TDLDA. In the x-only case the pair correlation function $\widetilde{G}_2(\mathbf{k},t)$ is completely determined by the one-article distribution function $\widetilde{f}(\mathbf{k},t)$,

$$\widetilde{G}_2(\mathbf{k},t) = -\sum_{\mathbf{p}}\widetilde{f}(\mathbf{k}+\mathbf{p},t)\widetilde{f}(\mathbf{p},t). \qquad (72)$$

Performing the mean field decoupling of the four-fermion terms in Eq. (60), we find that the right-hand side in this equation vanishes. Therefore the equation of motion for the function $\widetilde{f}(\mathbf{k},t)$ takes the form,

$$\frac{\partial}{\partial t}\widetilde{f}(\mathbf{k},t) = 0. \qquad (73)$$

Equation (73) coincides with the corresponding equation of motion for the noninteracting system, Eq. (62). Hence in the x-only approximation both the Wigner function and the pair correlation function in the Lagrangian frame preserve their initial form,

$$\widetilde{f}(\mathbf{k},t) = n^F_{\mathbf{k}}, \qquad (74)$$

$$\widetilde{G}_2(\mathbf{k},t) = G^x_2(n_0;k) = -\sum_{\mathbf{p}}n^F_{\mathbf{k}+\mathbf{p}}n^F_{\mathbf{p}}. \qquad (75)$$

Here $k=|\mathbf{k}|=\sqrt{k_\mu k_\mu}$ is the usual modulus of $\mathbf{k}$, and $G^x_2(n;k)$ is the exchange pair correlation function in the equilibrium Fermi gas of the density n. Substituting Eqs. (74) and (75) into Eq. (58) we obtain the following stress tensor in the interacting system:

$$\widetilde{P}_{\mu\nu} = \frac{\delta_{\mu\nu}}{\sqrt{g(\boldsymbol{\xi},t)}}P_0[n_0(\boldsymbol{\xi})] + \widetilde{P}^x_{\mu\nu}[n_0(\boldsymbol{\xi}),g_{\mu\nu}(\boldsymbol{\xi},t)]. \qquad (76)$$

The first term in the right-hand side of Eq. (76) is the kinetic stress tensor of the noninteracting system, while the second term $\widetilde{P}^x_{\mu\nu}$, corresponds to the exchange contribution to the local stress density,

$$\widetilde{P}^x_{\mu\nu} = \frac{1}{2g}\sum_{\mathbf{k}}\left[\frac{k_\mu k_\nu}{\|\mathbf{k}\|}\bar{w}'(\|\mathbf{k}\|) + g_{\mu\nu}\bar{w}(\|\mathbf{k}\|)\right]G^x_2(n_0;k). \qquad (77)$$

Using the transformation rules of Eq. (65) we get the following expression for the exchange stress tensor in the laboratory frame,

$$P^x_{\mu\nu}(n,\bar{g}_{\alpha\beta}) = \frac{\sqrt{\bar{g}}}{2}\sum_{\mathbf{p}}\left[\frac{p_\mu p_\nu}{p}\bar{w}'_p + \delta_{\mu\nu}\bar{w}_p\right] \times G^x_2\left(\frac{n}{\sqrt{\bar{g}}};\sqrt{\bar{g}^{\alpha\beta}p_\alpha p_\beta}\right), \qquad (78)$$

where we introduced a shortcut notation $\bar{w}_p=\bar{w}(p)$. Equations (78), (24), and (23) uniquely determine the local potential $U_x(\mathbf{x},t)$ in x-only TDLDA. Apparently the exchange potential $U_x(\mathbf{x},t)$ is a local (both in space and in time) functional of the density $n(\mathbf{x},t)$ and Cauchy's deformation tensor $\bar{g}_{\mu\nu}(\mathbf{x},t)$. In the equilibrium system ($\bar{g}_{\mu\nu}=\delta_{\mu\nu}$) the potential, defined by Eqs. (78), (24), and (23), reduces to that in the common static local exchange approximation.

C. Linear response TDLDA

In the linear response regime the deformation tensor $g_{\mu\nu}$ slightly deviates from the Kronecker symbol,

$$g^{\mu\nu}(\boldsymbol{\xi},t) \approx \delta_{\mu\nu} + \delta g^{\mu\nu}(\boldsymbol{\xi},t), \quad \delta g^{\mu\nu} = -\frac{\partial u_\mu}{\partial \xi^\nu} - \frac{\partial u_\nu}{\partial \xi^\mu},$$

$$(79)$$

where $\mathbf{u}=\mathbf{x}-\boldsymbol{\xi}$ is the displacement vector, which is assumed to be small. In the linearized theory the trajectory equation of Eq. (27) reduces to the common linear relation of the velocity to the displacement,

$$\frac{\partial \mathbf{u}(\boldsymbol{\xi},t)}{\partial t} = \mathbf{v}(\boldsymbol{\xi},t).$$

$$(80)$$

Substituting $g^{\mu\nu}$ of Eq. (79) into Eq. (57) and keeping only the term that are linear in $\delta g^{\mu\nu}$ terms, we obtain the following linearized Hamiltonian:

$$\tilde{H} = H + \hat{P}_{\mu\nu} \delta g^{\mu\nu},$$

$$(81)$$

where H is the standard Hamiltonian for the homogeneous system, and $\hat{P}_{\mu\nu}$ is the stress tensor operator,

$$\hat{P}_{\mu\nu} = \sum_{\mathbf{k}} \left[\frac{k_\mu k_\nu}{m} \tilde{a}_{\mathbf{k}}^\dagger \tilde{a}_{\mathbf{k}} + \frac{1}{2} \left(\frac{k_\mu k_\nu}{k} \bar{w}_k' + \delta_{\mu\nu} \bar{w}_k \right) \hat{\tilde{n}}_{\mathbf{k}} \hat{\tilde{n}}_{-\mathbf{k}} \right].$$

$$(82)$$

First we need to compute the stress tensor $\tilde{P}_{\mu\nu}$ in the Lagrangian frame, Eq. (58). In the linear regime Eq. (58) takes the form,

$$\tilde{P}_{\mu\nu} = \delta_{\mu\nu} P(n_0) + \tilde{Q}_{\mu\nu\alpha\beta}(\omega) \delta g^{\alpha\beta}(\omega),$$

$$(83)$$

where the linear response kernel $\tilde{Q}_{\mu\nu\alpha\beta}(\omega)$ can be represented as follows:

$$\tilde{Q}_{\mu\nu\alpha\beta}(\omega) = \tilde{Q}_{\mu\nu\alpha\beta}^\infty + \Delta\tilde{Q}_{\mu\nu\alpha\beta}(\omega).$$

$$(84)$$

The first frequency-independent term $\tilde{Q}_{\mu\nu\alpha\beta}^\infty$ in Eq. (84) comes from the explicit local-in-time dependence of the integrals in Eq. (58) on the deformation tensor $g_{\mu\nu}(t)$. Namely, the fourth-rank tensor $\tilde{Q}_{\mu\nu\alpha\beta}^\infty$ is defined by the following derivative:

$$\tilde{Q}_{\mu\nu\alpha\beta}^\infty = \left(\frac{\partial \tilde{P}_{\mu\nu}[f^{eq}, G_2^{eq}]}{\partial g^{\alpha\beta}} \right)_{g_{\mu\nu}=\delta_{\mu\nu}},$$

$$(85)$$

where $\tilde{P}_{\mu\nu}[f^{eq}, G_2^{eq}]$ is the stress tensor of Eq. (58), calculated with the equilibrium Wigner function, $f^{eq}(\mathbf{k})$, and the equilibrium pair correlation function, $G_2^{eq}(\mathbf{k})$. The perturbation (the second term) in the linearized Hamiltonian of Eq. (81) induces deviations of the Wigner function and the pair correlation function from their equilibrium values. These deviations are responsible for the second, nonlocal-in-time term in Eq. (84),

$$\Delta\tilde{Q}_{\mu\nu\alpha\beta} = \sum_{\mathbf{k}} \frac{k_\mu k_\nu}{2m} \frac{\delta\tilde{f}(\mathbf{k},t)}{\delta g^{\alpha\beta}(t')}$$

$$+ \frac{1}{2} \sum_{\mathbf{k}} \left(\frac{k_\mu k_\nu}{k} \bar{w}_k' + \delta_{\mu\nu} \bar{w}_k \right) \frac{\delta\tilde{G}_2(\mathbf{k},t)}{\delta g^{\alpha\beta}(t')}. \quad (86)$$

Comparing Eqs. (81), (82), and (86) we find that the dynamic kernel $\Delta\tilde{Q}_{\mu\nu\alpha\beta}$ can be related to the following stress autocorrelation function:

$$\Delta\tilde{Q}_{\mu\nu\alpha\beta}(\omega) = -i \int_0^\infty dt e^{i\omega t} \langle [\hat{P}_{\mu\nu}(t), \hat{P}_{\alpha\beta}(0)] \rangle. \quad (87)$$

Substituting the stress tensor in the Lagrangian frame, Eq. (83), into the transformation formula of Eq. (65) we compute the stress tensor in the laboratory frame,

$$P_{\mu\nu}(\mathbf{x},\omega) = \delta_{\mu\nu} P[n_0(\mathbf{x})] + \delta P_{\mu\nu}(\mathbf{x},\omega), \quad (88)$$

where

$$\delta P_{\mu\nu} = -\delta_{\mu\nu} \frac{\partial P}{\partial n_0} \mathbf{u} \nabla n_0 + Q_{\mu\nu\alpha\beta}(\omega) g^{\alpha\beta}(\omega). \quad (89)$$

The kernel $Q_{\mu\nu\alpha\beta}(\omega)$ in the laboratory frame takes the form, below which is similar to that of Eq. (84),

$$Q_{\mu\nu\alpha\beta}(\omega) = Q_{\mu\nu\alpha\beta}^\infty + \Delta\tilde{Q}_{\mu\nu\alpha\beta}(\omega). \quad (90)$$

The frequency-independent term $Q_{\mu\nu\alpha\beta}^\infty$ in Eq. (90) is related to the quantity $\tilde{Q}_{\mu\nu\alpha\beta}^\infty$ of Eq. (85) as follows:

$$Q_{\mu\nu\alpha\beta}^\infty = \frac{1}{2} P(\delta_{\mu\alpha}\delta_{\nu\beta} + \delta_{\nu\alpha}\delta_{\mu\beta}) + \tilde{Q}_{\mu\nu\alpha\beta}^\infty. \quad (91)$$

It is worth mentioning that the first term in the right-hand side in Eq. (89) guarantees that the harmonic potential theorem[16] is satisfied. In fact, only this term survives for the rigid motion of the system. Within the present formalism this term comes from the expansion of the argument of $\tilde{P}_{\mu\nu}$ in the transformation rule of Eq. (65). The correction to the kernel in the laboratory frame [the first term in Eq. (91)] corresponds to the expansion of the tensor prefactor in Eq. (65).

By symmetry the fourth-rank tensor $Q_{\mu\nu\alpha\beta}$ is uniquely representable in the form,

$$Q_{\mu\nu\alpha\beta} = \left(\frac{K}{2} + \frac{\mu}{d} \right) \delta_{\mu\nu}\delta_{\alpha\beta} + \frac{\mu}{2}(\delta_{\mu\alpha}\delta_{\nu\beta} + \delta_{\nu\alpha}\delta_{\mu\beta}). \quad (92)$$

The scalar coefficients, $K(\omega)$ and $\mu(\omega)$, in Eq. (92) are related to the tensor $Q_{\mu\nu\alpha\beta}$ contracted over different pairs of indexes,

$$K(\omega) = \frac{2}{d^2} Q_{\alpha\alpha\beta\beta}(\omega), \quad (93)$$

$$\mu(\omega) = \frac{2}{d^2 + d - 2} \left[Q_{\alpha\beta\alpha\beta}(\omega) - \frac{1}{d} Q_{\alpha\alpha\beta\beta}(\omega) \right]. \quad (94)$$

Substitution of Eq. (92) into Eq. (89) yields the following result for the linear correction to the stress tensor in the laboratory frame,

$$\delta P_{\mu\nu} = -\delta_{\mu\nu}\frac{\partial P}{\partial n_0}\mathbf{u}\,\nabla\,n_0 + \delta_{\mu\nu}K\frac{1}{2}\delta g^{\alpha\alpha} + \mu\left(\delta g^{\mu\nu} - \frac{\delta_{\mu\nu}}{d}\delta g^{\alpha\alpha}\right).$$

(95)

The stress tensor $\delta P_{\mu\nu}$ of Eq. (95) has a clear viscoelastic form, where $K(\omega)$ and $\mu(\omega)$ are the bulk modulus and the shear modulus, respectively. The xc stress tensor, $\delta P_{\mu\nu}^{\text{xc}}$, is the difference of the expressions given by Eq. (95) for the interacting and the noninteracting systems. Apparently $\delta P_{\mu\nu}^{\text{xc}}$ takes a form of Eq. (95) with P, K, and μ being replaced by P_{xc}, K_{xc}, and μ_{xc}, respectively, where

$$P_{\text{xc}} = P - \frac{2}{d}E_{\text{kin}}^{(0)},$$

(96)

$$K_{\text{xc}} = K - K_0, \quad \mu_{\text{xc}} = \mu - \mu_0,$$

(97)

are the xc pressure and the xc viscoelastic moduli. In Eqs. (96) and (97) $E_{\text{kin}}^{(0)}$, K_0, and μ_0 correspond to the kinetic energy, the bulk modulus, and the shear modulus of an ideal Fermi gas. Therefore in the linear response regime our TDLDA naturally reduces to the Vignale-Kohn approximation[17] in the viscoelastic formulation of Ref. 18.

An explicit microscopic representation for the bulk K and the shear μ moduli can be found using Eqs. (94), (93), (91), (90), (87), and (85). Both K and μ take the following general forms:

$$K(\omega) = K^{\infty} + \Delta K(\omega),$$

(98)

$$\mu(\omega) = \mu^{\infty} + \Delta\mu(\omega).$$

(99)

The first terms, K^{∞} and μ^{∞}, in the right-hand sides of Eqs. (98) and (99) are obtained by the substitution of $Q_{\mu\nu\alpha\beta}^{\infty}$ from Eq. (91) into Eqs. (93) and (94). Performing straightforward calculations for the interacting and the noninteracting systems, we arrive at the following results for the high frequency parts of the xc elastic moduli:

$$K_{\text{xc}}^{\infty} = \frac{d+2}{d}\left[\frac{2}{d}E_{\text{kin}}^{\text{xc}} + \sum_{\mathbf{k}}\frac{k^2\bar{w}_k'' + (3d+1)k\bar{w}_k' + 2d^2\bar{w}_k}{2d(d+2)}G_2^{\text{eq}}(k)\right],$$

(100)

$$\mu_{\text{xc}}^{\infty} = \frac{2}{d}E_{\text{kin}}^{\text{xc}} + \sum_{\mathbf{k}}\frac{k^2\bar{w}_k'' + (d+1)k\bar{w}_k'}{2d(d+2)}G_2^{\text{eq}}(k),$$

(101)

where $E_{\text{kin}}^{\text{xc}} = E_{\text{kin}} - E_{\text{kin}}^{(0)}$ is the xc kinetic energy of the equilibrium system. In the special case of Coulomb interaction the momentum integrals in Eqs. (100) and (101) can be expressed in terms of the potential energy per unit volume,

$$E_{\text{pot}} = \frac{1}{2}\sum_{\mathbf{k}}\bar{w}_k G_2^{\text{eq}}(k).$$

In d dimensions the Coulomb potential is proportional to $1/k^{d-1}$. Therefore we get the following identities for the derivatives that enter Eqs. (100) and (101),

$$k\bar{w}_k' = -(d-1)\bar{w}_k \quad \text{and} \quad k^2\bar{w}_k'' = d(d-1)\bar{w}_k.$$

These identities allow us to simplify Eqs. (100) and (101) as follows:

$$K_{\text{xc}}^{\infty} = \frac{2(d+2)}{d^2}E_{\text{kin}}^{\text{xc}} + \frac{d+1}{d^2}E_{\text{pot}},$$

(102)

$$\mu_{\text{xc}}^{\infty} = \frac{2}{d}E_{\text{kin}}^{\text{xc}} - \frac{d-1}{d(d+2)}E_{\text{pot}}.$$

(103)

The high-frequency forms of Eqs. (102) and (103) are well known in the literature.[39–41] Commonly they are derived using the "third moment sum rule." Within our formalism the expressions of Eqs. (100) and (101) for K_{xc}^{∞} and μ_{xc}^{∞} come about almost trivially from the explicit local-in-time dependence of the stress tensor, Eq. (58), on the deformation tensor.

To represent the frequency dependent parts of viscoelastic moduli in the most convenient form we decompose the stress tensor operator $\hat{P}_{\mu\nu}$, Eq. (82), into scalar and traceless parts,

$$\hat{P}_{\mu\nu} = \delta_{\mu\nu}\hat{P} + \hat{\pi}_{\mu\nu},$$

(104)

where $\hat{P} = 1/d\,\text{Tr}\hat{P}_{\mu\nu}$ is the pressure operator,

$$\hat{P} = \frac{1}{d}\sum_{\mathbf{k}}\left[\frac{k^2}{m}\tilde{a}_{\mathbf{k}}^{\dagger}\tilde{a}_{\mathbf{k}} + \frac{1}{2}(k\bar{w}_k' + d\bar{w}_k)\hat{\tilde{n}}_{\mathbf{k}}\hat{\tilde{n}}_{-\mathbf{k}}\right],$$

(105)

and $\hat{\pi}_{\mu\nu}$ is the operator of the traceless part of the stress tensor ($\text{Tr}\hat{\pi}_{\mu\nu}=0$),

$$\hat{\pi}_{\mu\nu} = \sum_{\mathbf{k}}\left(\frac{k_\mu k_\nu}{k^2} - \frac{\delta_{\mu\nu}}{d}\right)\left(\frac{k^2}{m}\tilde{a}_{\mathbf{k}}^{\dagger}\tilde{a}_{\mathbf{k}} + \frac{k\bar{w}_k' + d\bar{w}_k}{2d}\hat{\tilde{n}}_{\mathbf{k}}\hat{\tilde{n}}_{-\mathbf{k}}\right).$$

(106)

Substituting $\Delta\tilde{Q}_{\mu\nu\alpha\beta}(\omega)$ of Eq. (87) into Eqs. (93) and (94) and using Eqs. (104)–(106), we find that $\Delta K_{\text{xc}}(\omega)$ equals the pressure-autocorrelation function,

$$\Delta K_{\text{xc}}(\omega) = -2i\int_0^{\infty}e^{i\omega t}\langle[\hat{P}(t),\hat{P}(0)]\rangle dt,$$

(107)

whereas the dynamic part of the shear modulus, $\Delta\mu_{\text{xc}}(\omega)$, is proportional to the autocorrelation function of the traceless tensor operator $\hat{\pi}_{\mu\nu}$,

$$\Delta\mu_{\text{xc}}(\omega) = \frac{-2i}{d^2+d-2}\int_0^{\infty}e^{i\omega t}\langle[\hat{\pi}_{\mu\nu}(t),\hat{\pi}_{\mu\nu}(0)]\rangle dt.$$

(108)

The coefficient, $2/(d^2+d-2)$, in Eq. (108) is exactly the inverse number of independent components of a second-rank traceless tensor. We would like to outline a very natural form of Eqs. (107) and (108), which is in clear agreement with the physical significance of the quantities K and μ.

The frequency-dependent contributions to the viscoelastic moduli are related to the dynamics of the Wigner function and the pair correlation function [see Eq. (86)]. In Sec. V B we have shown that in the exchange approximation the time-

dependent deformation tensor in the Hamiltonian of Eq. (57) does not induce any dynamics of $\widetilde{f}(\mathbf{k})$ and $\widetilde{G}_2(\mathbf{k})$. Therefore only correlations are responsible for nonvanishing $\Delta K_{xc}(\omega)$ and $\Delta \mu_{xc}(\omega)$. The most important effect of the dynamic correlations, which is described by Eqs. (107) and (108), is the memory loss due to collisions. Since in a zero-temperature Fermi system the collisional dissipation should be suppressed, there is a hope that the frequency-dependent parts of K_{xc} and μ_{xc} do not substantially influence the dynamics. (We note that this is not in general true for steady state transport situations, in which dissipation plays an essential role.) Neglecting $\Delta K_{xc}(\omega)$ and $\Delta \mu_{xc}(\omega)$ we get a purely elastic xc stress tensor with the bulk and the shear moduli defined by Eqs. (100) and (101). Another argument due to Conti and Vignale[39] also shows that for an electron gas the elastic approximation should work reasonably well. Indeed, the dissipation effects are absent in the x-only approximation that is valid in the weak-coupling (high-density) regime. In the strong-coupling (low-density) limit, electrons tend to form a Wigner crystal—the state where the collisional dissipation also vanishes. Therefore one naturally expects that at all intermediate densities the purely elastic approximation should provide a reasonable description of the dynamic stress.

D. Nonlinear elastic TDLDA

The linear VK approximation with a purely elastic bulk modulus K_{xc}^∞, Eq. (100), and shear modulus μ_{xc}^∞, Eq. (101), allows for a simple nonlinear extension. In this section we derive this nonlinear elastic TDLDA and formulate a complete set of self-consistent KS equations in a convenient for practical applications form.

1. Exchange-correlation stress tensor in the elastic TDLDA

Elastic TDLDA is based on the assumption that both the Wigner function and the pair correlation function in the Lagrangian frame preserve their initial forms. This corresponds to the dynamics with extremely pronounced memory that is not destroyed by the effects of collisional relaxation. To get the stress tensor for the system evolving from the equilibrium state we have to substitute $\widetilde{f}(\mathbf{k},t)=f^{eq}(n_0;k)$ and $\widetilde{G}_2(\mathbf{k},t)=G_2^{eq}(n_0;k)$ into Eq. (58). As a result the stress tensor in the Lagrangian frame takes the form,

$$\widetilde{P}_{\mu\nu} = \frac{\delta_{\mu\nu}}{\sqrt{g}}\frac{2}{d}E_{kin}(n_0) + \frac{1}{2g}\sum_{\mathbf{k}}\left[\frac{k_\mu k_\nu}{\|\mathbf{k}\|}\bar{w}'(\|\mathbf{k}\|)\right.$$
$$\left. + g_{\mu\nu}\bar{w}(\|\mathbf{k}\|)\right]G_2^{eq}(n_0;k). \qquad (109)$$

Tensor $\widetilde{P}_{\mu\nu}(\boldsymbol{\xi},t)$ of Eq. (109) locally depends on the density $n_0(\boldsymbol{\xi})$ and Green's deformation tensor $g_{\mu\nu}(\boldsymbol{\xi},t)$ in a given point $\boldsymbol{\xi}$ of the Lagrangian space. Transforming this tensor back to the laboratory frame and subtracting the KS stress tensor of Eq. (71) we obtain the following result for the xc stress tensor in the physical **x** space,

$$P_{\mu\nu}^{xc} = \frac{2\bar{g}_{\mu\nu}}{d}\sqrt{\bar{g}}E_{kin}^{xc}\left(\frac{n}{\sqrt{\bar{g}}}\right) + \frac{\sqrt{\bar{g}}}{2}\sum_{\mathbf{p}}\left[\frac{p_\mu p_\nu}{p}\bar{w}'_p + \delta_{\mu\nu}\bar{w}_p\right]$$
$$\times G_2^{eq}\left(\frac{n}{\sqrt{\bar{g}}}; \sqrt{\bar{g}^{\alpha\beta}p_\alpha p_\beta}\right). \qquad (110)$$

Equation (110) determines the xc stress tensor as a function of the time-dependent density, $n(\mathbf{x},t)$, and Cauchy's deformation tensor $\bar{g}_{\mu\nu}(\mathbf{x},t)$. Let us remember that the memory-related nonlocality of $P_{\mu\nu}^{xc}$, Eq. (110), is hidden in the "local" representation of the function $n_0[\boldsymbol{\xi}(\mathbf{x},t)]$ [see Eq. (70)]. The "elastic" xc potential is the solution to the Poisson Eq. (23), where the xc "charge density" is defined after Eqs. (24) and (110).

In the exchange approximation the stress tensor $P_{\mu\nu}^{xc}$, Eq. (110), reduces to the x-only tensor $P_{\mu\nu}^x$ of Eq. (78) that is exact in the weak-coupling limit. In the linear response regime the corrections to the density and to the Cauchy's deformation tensor are proportional to the displacement vector,

$$n = n_0 - \nabla n_0 \mathbf{u}, \quad \bar{g}_{\mu\nu} = \delta_{\mu\nu} + \frac{\partial u_\mu}{\partial x^\nu} + \frac{\partial u_\nu}{\partial x^\mu}. \qquad (111)$$

Linearizing the stress tensor of Eq. (110) and using Eq. (111) we straightforwardly recover the VK approximation[17,18] with the elastic moduli K_{xc}^∞, Eq. (100), and μ_{xc}^∞, Eq. (101).

2. Self-consistent Kohn-Sham equations

Let us formulate the complete set of self-consistent KS equations in the elastic TDLDA. The Kohn-Sham formulation of TDDFT allows to calculate the density $n(\mathbf{x},t)$ and the velocity $\mathbf{v}(\mathbf{x},t)$ in the interacting N-particle system using the ideal gas formulas,

$$n(\mathbf{x},t) = \sum_{j=1}^{N} |\phi_j(\mathbf{x},t)|^2, \qquad (112)$$

$$\mathbf{v}(\mathbf{x},t) = \frac{1}{n}\sum_{j=1}^{N}\frac{i}{2m}[\phi_j \nabla \phi_j^* - \phi_j^* \nabla \phi_j]. \qquad (113)$$

Single particle orbitals $\phi_j(\mathbf{x},t)$ satisfy the time-dependent KS equations,

$$i\frac{\partial \phi_j}{\partial t} = -\frac{\nabla^2}{2m}\phi_j + (U_{ext} + U_{eff}[n,\bar{g}_{\mu\nu}])\phi_j, \qquad (114)$$

where $U_{ext}(\mathbf{x},t)$ is the external potential. For the practically important case of a three-dimensional (3D) system with Coulomb interaction the effective potential $U_{eff}[n,\bar{g}_{\mu\nu}](\mathbf{x},t)$ is the solution to the following Poisson equation:

$$\nabla^2 U_{eff} = 4\pi(e^2 n + \rho_{xc}[n,\bar{g}_{\mu\nu}]). \qquad (115)$$

The first term in the brackets in Eq. (115) generates the Hartree potential U_H while the second term is responsible for the xc potential. The xc charge density ρ_{xc} is the local functional of n and $\bar{g}_{\mu\nu}$,

$$\rho_{xc} = \frac{1}{4\pi}\frac{\partial}{\partial x^\mu}\left[\frac{1}{n}\frac{\partial}{\partial x^\nu}P_{\mu\nu}^{xc}(n,\bar{g}_{\mu\nu})\right], \qquad (116)$$

where $P_{\mu\nu}^{\rm xc}(n, \bar{g}_{\mu\nu})$ is the *function* of $n(\mathbf{x}, t)$ and $\bar{g}_{\mu\nu}(\mathbf{x}, t)$, which is defined by Eq. (110). In Appendix C we show that for a Coulomb system, Eq. (110) simplifies as follows:

$$P_{\mu\nu}^{\rm xc} = \frac{2}{3} \bar{g}_{\mu\nu} \sqrt{\bar{g}} E_{\rm kin}^{\rm xc}\left(\frac{n}{\sqrt{\bar{g}}}\right) + L_{\mu\nu}(\bar{g}_{\alpha\beta}) E_{\rm pot}\left(\frac{n}{\sqrt{\bar{g}}}\right), \quad (117)$$

where $L_{\mu\nu}(\bar{g}_{\alpha\beta})$ is a purely geometric factor that is explicitly defined in Appendix C. Therefore the dependence of $P_{\mu\nu}^{\rm xc}(n, \bar{g}_{\mu\nu})$ on $\bar{g}_{\mu\nu}$ and on $n/\sqrt{\bar{g}}$ is completely factorized, which should significantly simplify practical applications. The kinetic $E_{\rm kin}^{\rm xc}(n)$ and the potential $E_{\rm pot}(n)$ energies of the homogeneous electron gas can be expressed in terms of the xc energy per particle $\epsilon_{\rm xc}(n)$ (see, for example, Ref. 39). For $d=3$ we get,

$$E_{\rm kin}^{\rm xc}(n) = 3n^{7/3}\left(\frac{\epsilon_{\rm xc}}{n^{1/3}}\right)', \quad E_{\rm pot}(n) = -3n^{8/3}\left(\frac{\epsilon_{\rm xc}}{n^{2/3}}\right)'$$

Hence, our nonadiabatic TDLDA requires only a knowledge of the function $\epsilon_{\rm xc}(n)$ for the homogeneous electron gas, exactly as the common static LDA does.

The density n, which enters Eqs. (115)–(117), is related to KS orbitals via Eq. (112). The second basic variable, Cauchy's deformation tensor $\bar{g}_{\mu\nu}$, is uniquely determined by the velocity $\mathbf{v}(\mathbf{x}, t)$, Eq. (113). To compute the deformation tensor we need to solve the trajectory equation of Eq. (27) and then substitute the solution into the definition of $\bar{g}_{\mu\nu}$, Eq. (67). It is, however, more convenient to determine this tensor directly from the solution of an equation of motion for $\bar{g}_{\mu\nu}(\mathbf{x}, t)$.[42] This equation of motion can be derived as follows. Let us consider the contravariant tensor $\bar{g}^{\mu\nu}$ (the inverse of $\bar{g}_{\mu\nu}$),

$$\bar{g}^{\mu\nu} = \frac{\partial x^\mu}{\partial \xi^\alpha} \frac{\partial x^\nu}{\partial \xi^\alpha}. \quad (118)$$

Using the trajectory equation of Eq. (27) we can compute the time derivative of $\bar{g}^{\mu\nu}$, Eq. (118), at constant $\boldsymbol{\xi}$ (i.e., within the Lagrangian description),

$$\left(\frac{\partial \bar{g}^{\mu\nu}}{\partial t}\right)_{\boldsymbol{\xi}} = \frac{\partial v^\mu}{\partial x^\alpha} \bar{g}^{\alpha\nu} + \bar{g}^{\mu\alpha} \frac{\partial v^\nu}{\partial x^\alpha}. \quad (119)$$

The time derivative of $\bar{g}_{\mu\nu}$ can be related to the time derivative of $\bar{g}^{\mu\nu} = (\bar{g}_{\mu\nu})^{-1}$ as follows: $\partial_t \bar{g} = -\bar{g}(\partial_t \bar{g}^{-1})\bar{g}$. Using this relation and taking into account that

$$(\partial_t)_{\boldsymbol{\xi}} = (\partial_t)_{\mathbf{x}} + \mathbf{v}\nabla,$$

we get the final equation of motion for Cauchy's deformation tensor $\bar{g}_{\mu\nu}(\mathbf{x}, t)$,

$$\frac{\partial \bar{g}_{\mu\nu}}{\partial t} = -v^\alpha \frac{\partial \bar{g}_{\mu\nu}}{\partial x^\alpha} - \frac{\partial v^\alpha}{\partial x^\mu} \bar{g}_{\alpha\nu} - \frac{\partial v^\alpha}{\partial x^\nu} \bar{g}_{\alpha\mu}. \quad (120)$$

Equation (120) should be solved with the initial condition $\bar{g}_{\mu\nu}(\mathbf{x}, 0) = \delta_{\mu\nu}$, which follows from the initial condition for the trajectory equation of Eq. (27).

The system of Eqs. (112)–(117) and (120) constitute the complete set of self-consistent KS equations in the nonlinear elastic TDLDA. In the equilibrium situation ($\bar{g}_{\mu\nu} = \delta_{\mu\nu}$) this system reduces to the common static KS equation with the LDA xc potential. In the linear regime it recovers the results of VK approximation with the elastic moduli of Eqs. (102) and (103). The nonadiabatic memory effects are described by Cauchy's deformation tensor, which satisfies Eq. (120). It should be noted that from the computational point of view the solution of this equation does not introduce any addition difficulties. Formally Eq. (120) has the same structure as the time-dependent KS Eq. (114). Hence Eqs. (114) and (120) can be solved simultaneously by the same method.

Very recently VK approximation has been successfully applied to the description of the optical and polarization properties of many different systems, such as atoms, molecules, semiconductors, and polymers.[43–47] Since VK approximation is a linearized version of our theory, we hope that the general TDLDA also will become a useful tool for studying nonlinear time-dependent phenomena.

VI. CONCLUSION

TDDFT extends powerful ideology of the ground state DFT to the domain of nonequilibrium phenomena. However, in contrast to the static DFT, which is currently a common computational tool in many branches of physics, its time-dependent counterpart still suffers from a number of unresolved problems. One of those problems is a lack of a well founded basic local approximation that would play a role similar to the LDA in the static DFT. In this paper we have shown that the local approximation in TDDFT can be regularly derived, but that this derivation requires almost complete reconsideration of the theory. We reformulated TDDFT from the point of view of a local comoving observer. The new formulation of the theory shows that the most natural basic variables in TDDFT are the local geometric characteristics of the deformations in a quantum many-body system.

Throughout this paper we used the analogy of TDDFT to classical continuum mechanics. The importance of the hydrodynamic interpretation, which perfectly fits the very idea of DFT, is one of the messages of the present work. Using the hydrodynamic formulation of TDDFT we were able to relate the xc potentials to the local stress. In particular we proved that the exact xc force must have the form of the divergence of a second-rank tensor. The well known zero-force and zero-torque sum rules are direct consequences of this strong local requirement. The functional dependence of xc potential on the basic variables also acquires a clear physical meaning. It corresponds to the stress-deformation relation, which is very natural from the point of view of continuum mechanics. If spatial derivatives of the deformation tensor are small, the stress-deformation relation becomes local and therefore we get the local approximation for the xc potential in TDDFT. It is natural to abbreviate this approximation as TDLDA, which means time-dependent local deformation approximation. In the linear response regime the general stress-deformation relation (TDLDA) reduces to the linear Hook's law,[30] which exactly coincides with the viscoelastic VK approximation.[17,18] The formal applicability conditions for the general nonlinear TDLDA are the same as for the linear VK approximation. It should be also noted that

neither of previously proposed nonlinear phenomenological constructions[18–20] is in general confirmed by the present regular microscopic consideration.

In Sec. V of this paper we discussed the elastic TDLDA. In this approximation the xc stress tensor is simply a function of the density and of the Cauchy's deformation tensor. For a system with a Coulomb interaction we presented the xc stress tensor and the xc potential in an explicit "ready for implementation" form. We also formulated the full set of self-consistent KS equations in TDLDA. In the equilibrium state the deformation tensor is diagonal and TDLDA reduces to the standard static LDA, while in the linear response regime it recovers VK approximation. To conclude we mention that the self-consistent equations of Sec. V D 2 can be straightforwardly reformulated in terms of xc vector potential. The only difference is that the Poisson equation for U_{xc} should be replaced by Eq. (21) which relates $\mathbf{A}^{xc}$ to the xc stress tensor. This replacement leads to one more evolution equation which should be solved simultaneously with the KS equation and the equation for Cauchy's deformation tensor.

APPENDIX A: STRESS TENSORS IN A GENERAL, NONINERTIAL FRAME

The microscopic representation for the stress tensor $\widetilde{P}_{\mu\nu}(\boldsymbol{\xi},t)$ in a general, local, noninertial frame has been derived in I,

$$\widetilde{P}_{\mu\nu}(\boldsymbol{\xi},t) = \widetilde{T}_{\mu\nu}(\boldsymbol{\xi},t) + \widetilde{W}_{\mu\nu}(\boldsymbol{\xi},t), \tag{A1}$$

where the kinetic stress tensor $\widetilde{T}_{\mu\nu}(\boldsymbol{\xi},t)$ and the interaction stress tensor, $\widetilde{W}_{\mu\nu}(\boldsymbol{\xi},t)$, are obtained by the transformation of $T_{\mu\nu}(\mathbf{x},t)$, Eq. (14), and $W_{\mu\nu}(\mathbf{x},t)$, Eq. (15), to the new frame. Namely,

$$\widetilde{T}_{\mu\nu}(\boldsymbol{\xi},t) = \frac{\partial x^\alpha}{\partial \xi^\mu}\frac{\partial x^\beta}{\partial \xi^\nu} T_{\alpha\beta}(\mathbf{x}(\boldsymbol{\xi},t),t), \tag{A2}$$

$$\widetilde{W}_{\mu\nu}(\boldsymbol{\xi},t) = \frac{\partial x^\alpha}{\partial \xi^\mu}\frac{\partial x^\beta}{\partial \xi^\nu} W_{\alpha\beta}(\mathbf{x}(\boldsymbol{\xi},t),t). \tag{A3}$$

The result of the transformation, Eqs. (A2) and (A3), takes the following form:

$$\widetilde{T}_{\mu\nu}(\boldsymbol{\xi},t) = \frac{1}{2m}\left\langle (\hat{K}_\mu g^{(-1/4)}\widetilde{\psi})^\dagger(\hat{K}_\nu g^{(-1/4)}\widetilde{\psi}) \right.$$
$$+ (\hat{K}_\nu g^{(-1/4)}\widetilde{\psi})^\dagger(\hat{K}_\mu g^{(-1/4)}\widetilde{\psi})$$
$$\left. - \frac{g_{\mu\nu}}{2}\frac{1}{\sqrt{g}}\frac{\partial}{\partial\xi^\alpha}\sqrt{g}g^{\alpha\beta}\frac{\partial}{\partial\xi^\beta}\frac{\widetilde{\psi}^\dagger\widetilde{\psi}}{\sqrt{g}} \right\rangle, \tag{A4}$$

$$\widetilde{W}_{\mu\nu}(\boldsymbol{\xi},t) = -\frac{g_{\mu\alpha}g_{\nu\beta}}{2\sqrt{g}}\int_0^1 d\lambda \int d\boldsymbol{\eta}\, d\boldsymbol{\eta}'\, \delta[\boldsymbol{\xi}$$
$$- \mathbf{z}_{\boldsymbol{\eta},\boldsymbol{\eta}'}(\lambda)]\frac{\dot{z}_{\boldsymbol{\eta},\boldsymbol{\eta}'}^\alpha(\lambda)\dot{z}_{\boldsymbol{\eta},\boldsymbol{\eta}'}^\beta(\lambda)}{l_{\boldsymbol{\eta},\boldsymbol{\eta}'}}\frac{\partial w(l_{\boldsymbol{\eta},\boldsymbol{\eta}'})}{\partial l_{\boldsymbol{\eta},\boldsymbol{\eta}'}}\widetilde{G}_2(\boldsymbol{\eta},\boldsymbol{\eta}'). \tag{A5}$$

Here the function $\mathbf{z}_{\boldsymbol{\eta},\boldsymbol{\eta}'}(\lambda)$ is the geodesic that connects points $\boldsymbol{\eta}$ and $\boldsymbol{\eta}'$, and $l_{\boldsymbol{\eta},\boldsymbol{\eta}'}$ is the length of this geodesic. The curve $\mathbf{z}_{\boldsymbol{\eta},\boldsymbol{\eta}'}(\lambda)$ can be found from the solution of the geodesic equation (see, for example, Ref. 34),

$$\ddot{z}^\mu(\lambda) + \Gamma^\mu_{\alpha\beta}(\mathbf{z})\dot{z}^\alpha(\lambda)\dot{z}^\beta(\lambda) = 0, \tag{A6}$$

supplemented by the boundary conditions $\mathbf{z}(0) = \boldsymbol{\eta}$, $\mathbf{z}(1) = \boldsymbol{\eta}'$. In Eqs. (A5) and (A6) $\dot{\mathbf{z}} = \partial\mathbf{z}/\partial\lambda$, and $\Gamma^\mu_{\alpha\beta}$ is an affine connection,

$$\Gamma^\mu_{\alpha\beta}(\boldsymbol{\xi}) = \frac{1}{2}g^{\mu\nu}\left(\frac{\partial g_{\nu\alpha}}{\partial\xi^\beta} + \frac{\partial g_{\nu\beta}}{\partial\xi^\alpha} - \frac{\partial g_{\alpha\beta}}{\partial\xi^\nu}\right). \tag{A7}$$

Equations (A4) and (A5) define tensors $\widetilde{T}_{\mu\nu}$ and $\widetilde{W}_{\mu\nu}$ as functionals of the microscopic state of the system. Tensor $\widetilde{T}_{\mu\nu}$ is a linear functional of the one particle density matrix, $\widetilde{\rho}_1(\boldsymbol{\xi},\boldsymbol{\xi}') = \langle\widetilde{\psi}^\dagger(\boldsymbol{\xi})\widetilde{\psi}(\boldsymbol{\xi}')\rangle$. Similarly, $\widetilde{W}_{\mu\nu}$ is a linear functional of the pair correlation function $\widetilde{G}_2(\boldsymbol{\xi},\boldsymbol{\xi}') = \langle\widetilde{\psi}^\dagger(\boldsymbol{\xi})\hat{\widetilde{n}}(\boldsymbol{\xi}')\widetilde{\psi}(\boldsymbol{\xi})\rangle - \widetilde{n}(\boldsymbol{\xi})\widetilde{n}(\boldsymbol{\xi}')$. Therefore,

$$\widetilde{P}_{\mu\nu} = \widetilde{T}_{\mu\nu} + \widetilde{W}_{\mu\nu} = \widetilde{P}_{\mu\nu}[\widetilde{\rho}_1,\widetilde{G}_2](\boldsymbol{\xi},t). \tag{A8}$$

Equation (A8) is the result, which we need for the discussion of TDDFT in Sec. IV.

APPENDIX B: STRESS TENSORS FOR A HOMOGENEOUS DEFORMATION

For a homogeneous system with $g_{\mu\nu}(\boldsymbol{\xi},t) = g_{\mu\nu}(t)$ and $\widetilde{v}_{T_\mu} = 0$ the general expressions, Eqs. (A4) and (A5), for the stress tensors simplify as follows. Equation (A4) for the kinetic stress tensor takes the form,

$$\widetilde{T}_{\mu\nu} = \frac{1}{2m\sqrt{g}}\left\langle \frac{\partial\widetilde{\psi}^\dagger}{\partial\xi^\mu}\frac{\partial\widetilde{\psi}}{\partial\xi^\nu} + \frac{\partial\widetilde{\psi}^\dagger}{\partial\xi^\nu}\frac{\partial\widetilde{\psi}}{\partial\xi^\mu} \right\rangle = -\frac{1}{m\sqrt{g}}\frac{\partial^2\widetilde{\rho}_1(\boldsymbol{\xi})}{\partial\xi^\mu\partial\xi^\nu}, \tag{B1}$$

where $\widetilde{\rho}_1(\boldsymbol{\xi}-\boldsymbol{\xi}') = \langle\widetilde{\psi}^\dagger(\boldsymbol{\xi})\widetilde{\psi}(\boldsymbol{\xi}')\rangle$ is the one particle density matrix for the homogeneous system. Introducing the Wigner function,

$$\widetilde{f}(\mathbf{k}) = \int e^{-ik_\mu\xi^\mu}\widetilde{\rho}_1(\boldsymbol{\xi})d\boldsymbol{\xi}, \tag{B2}$$

we obtain the following final representation for $\widetilde{T}_{\mu\nu}$:

$$\widetilde{T}_{\mu\nu} = \frac{1}{\sqrt{g}}\sum_{\mathbf{k}}\frac{k_\mu k_\nu}{m}\widetilde{f}(\mathbf{k}). \tag{B3}$$

To calculate the interaction stress tensor, Eq. (A5), we need to solve the geodesic equation of Eq. (A7). For a ho-

mogeneous metric the solution is a straight line,

$$\mathbf{z}_{\boldsymbol{\eta},\boldsymbol{\eta}'}(\lambda) = \boldsymbol{\eta} + (\boldsymbol{\eta}' - \boldsymbol{\eta})\lambda. \tag{B4}$$

The length of the geodesic, $l_{\boldsymbol{\eta},\boldsymbol{\eta}'}$, can be calculated as follows:

$$l_{\boldsymbol{\eta},\boldsymbol{\eta}'} = \int_0^1 \sqrt{g_{\mu\nu}(\mathbf{z})\dot{z}^\mu(\lambda)\dot{z}^\nu(\lambda)}\, d\lambda = \sqrt{g_{\mu\nu}(\eta^\mu - \eta'^\mu)(\eta^\nu - \eta'^\nu)}$$

$$:= \|\boldsymbol{\eta} - \boldsymbol{\eta}'\|. \tag{B5}$$

Substituting Eqs. (B4) and (B5) into Eq. (A5) and taking into account that for a homogeneous system $\widetilde{G}_2(\boldsymbol{\eta},\boldsymbol{\eta}') = \widetilde{G}_2(\boldsymbol{\eta} - \boldsymbol{\eta}')$, we arrive at the following result:

$$\widetilde{W}_{\mu\nu} = -\frac{g_{\mu\alpha}g_{\nu\beta}}{2\sqrt{g}}\int \frac{\xi^\alpha \xi^\beta}{\|\boldsymbol{\xi}\|}\frac{\partial w(\|\boldsymbol{\xi}\|)}{\partial\|\boldsymbol{\xi}\|}\widetilde{G}_2(\boldsymbol{\xi})d\boldsymbol{\xi}. \tag{B6}$$

Let us expand the pair correlation function, $\widetilde{G}_2(\boldsymbol{\xi})$, in a Fourier series,

$$\widetilde{G}_2(\boldsymbol{\xi}) = \sum_{\mathbf{k}} e^{ik_\mu \xi^\mu}\widetilde{G}_2(\mathbf{k}), \tag{B7}$$

and express $\widetilde{W}_{\mu\nu}$, Eq. (B6), in terms of $\widetilde{G}_2(\mathbf{k})$.

First we note, that the following simple relation holds. Let $\bar{F}(|\mathbf{k}|)$, where $|\mathbf{k}| = \sqrt{k_\mu k_\mu}$, be the Fourier component of a function $F(|\boldsymbol{\xi}|)$, i.e.,

$$\bar{F}(|\mathbf{k}|) = \int e^{-ik_\mu \xi^\mu}F(|\xi|)d\boldsymbol{\xi}. \tag{B8}$$

Then, the Fourier component of the function $F(\|\boldsymbol{\xi}\|)$ can be expressed in terms of $\bar{F}$ as follows:

$$\int e^{-ik_\mu \xi^\mu}F(\|\boldsymbol{\xi}\|)d\boldsymbol{\xi} = \frac{1}{\sqrt{g}}\bar{F}(\|\mathbf{k}\|), \tag{B9}$$

where $\|\boldsymbol{\xi}\| = \sqrt{g_{\mu\nu}\xi^\mu \xi^\nu}$ [see Eq. (B5)], and

$$\|\mathbf{k}\| = \sqrt{g^{\mu\nu}k_\mu k_\nu}. \tag{B10}$$

Substituting the expansion of Eq. (B7) into Eq. (B6) and using Eqs. (B8) and (B9), we get the required representation for the interaction stress tensor,

$$\widetilde{W}_{\mu\nu} = \frac{1}{2g}\sum_{\mathbf{k}}\left[\frac{k_\mu k_\nu}{\|\mathbf{k}\|}\bar{w}'(\|\mathbf{k}\|) + g_{\mu\nu}\bar{w}(\|\mathbf{k}\|)\right]\widetilde{G}_2(\mathbf{k}). \tag{B11}$$

In Eq. (B11) the function $\bar{w}(|\mathbf{k}|)$ is the Fourier component of the interaction potential $w(|\boldsymbol{\xi}|)$ and $\bar{w}'(x) = d\bar{w}(x)/dx$.

The stress tensor $\widetilde{P}_{\mu\nu}$ of Eq. (58) is the sum of $\widetilde{T}_{\mu\nu}$, Eq. (B3), and $\widetilde{W}_{\mu\nu}$, Eq. (B11).

APPENDIX C: ELASTIC STRESS TENSOR IN COULOMB SYSTEMS

The general expression for the xc stress tensor, Eq. (110), can be represented in a much simpler form if the particles

interact via the Coulomb potential, $\bar{w}_p = A_d/p^{d-1}$ (where $A_3 = 4\pi e^2$ and $A_2 = 2\pi e^2$). Below we show that in this case the second term in Eq. (110) can be related to the potential energy, E_{pot}, of a homogeneous electron gas with the density $n/\sqrt{g}$.

Let us represent the momentum integral in Eq. (110) as a sum of two terms,

$$W_{\mu\nu} = W_{\mu\nu}^{(1)} + W_{\mu\nu}^{(2)} \tag{C1}$$

where

$$W_{\mu\nu}^{(1)} = \delta_{\mu\nu}\frac{\sqrt{g}}{2}\sum_{\mathbf{p}}\frac{A_d}{p^{d-1}}G_2^{\text{eq}}(\sqrt{\bar{g}^{\alpha\beta}p_\alpha p_\beta}), \tag{C2}$$

$$W_{\mu\nu}^{(2)} = -(d-1)\frac{\sqrt{g}}{2}\sum_{\mathbf{p}}A_d\frac{p_\mu p_\nu}{p^{d+1}}G_2^{\text{eq}}(\sqrt{\bar{g}^{\alpha\beta}p_\alpha p_\beta}). \tag{C3}$$

To shorten the notations we retain only important momentum dependence in the argument of the pair correlation function G_2^{eq}.

The transformation of $W_{\mu\nu}^{(1)}$, Eq. (C2), is straightforward. By changing the integration variables this equation can be reduced to the form,

$$W_{\mu\nu}^{(1)} = \delta_{\mu\nu}\frac{\bar{g}}{2}\sum_{\mathbf{p}}\frac{A_d}{[\bar{g}_{\alpha\beta}p_\alpha p_\beta]^{(d-1)/2}}G_2^{\text{eq}}(p). \tag{C4}$$

Separating the integration over the modulus and the direction of momentum in Eq. (C4) yields the following result for $W_{\mu\nu}^{(1)}$:

$$W_{\mu\nu}^{(1)} = \delta_{\mu\nu}\bar{g}\left\langle\frac{1}{[\bar{g}_{\alpha\beta}l^\alpha l^\beta]^{(d-1)/2}}\right\rangle_{\mathbf{l}}E_{\text{pot}}, \tag{C5}$$

where $\mathbf{l}$ is a unit vector ($\mathbf{l}^2 = 1$), and the angle brackets, $\langle(\cdots)\rangle_{\mathbf{l}}$, stand for the averaging over the directions of $\mathbf{l}$.

The momentum integral for $W_{\mu\nu}^{(2)}$, Eq. (C3), can be reduced to a similar form. Let us first represent the deformation tensor $\bar{g}_{\mu\nu}$ in terms of its eigenvalues, λ_j^2, and eigenvectors, $\eta_{j\mu}$,

$$\bar{g}_{\mu\nu} = \lambda_j^2 \eta_{j\mu}\eta_{j\nu}. \tag{C6}$$

Here $j = 1,\ldots,d$ labels the eigenvectors $\boldsymbol{\eta}_j$ that satisfy the completeness and the orthonormality conditions,

$$\eta_{j\mu}\eta_{j\nu} = \delta_{\mu\nu}, \quad \eta_{i\mu}\eta_{j\mu} = \delta_{ij}. \tag{C7}$$

Tensor $\bar{g}^{\mu\nu}$ has the same eigenvectors, while its eigenvalues equal to $1/\lambda_j^2$. Substituting the eigenvector expansion of $\bar{g}^{\mu\nu}$ into Eq. (C3), and performing an obvious change of the integration variables, we arrive at the following result for the tensor $W_{\mu\nu}^{(2)}$:

$$W_{\mu\nu}^{(2)} = -\bar{g}\eta_{j\mu}\eta_{j\nu}\left\langle\frac{(d-1)\lambda_j^2(\boldsymbol{\eta}_j\mathbf{l})^2}{[\bar{g}_{\alpha\beta}l^\alpha l^\beta]^{(d+1)/2}}\right\rangle_{\mathbf{l}}E_{\text{pot}}. \tag{C8}$$

Combining Eqs. (C1), (C5), and (C8) we obtain the interaction stress tensor $W_{\mu\nu}$ in the following form:

$$W_{\mu\nu} = L_{\mu\nu}(g_{\alpha\beta})E_{\text{pot}}, \tag{C9}$$

where the calculation of the function $L_{\mu\nu}(g_{\alpha\beta})$ involves only the angle integration. The angle integrals [factors with angle brackets in Eqs. (C5) and (C8)] are the scalar functions which depend only on the eigenvalues of $\bar{g}_{\mu\nu}$. For $d=2,3$ these integrals are reducible to a combination of the standard elliptic integrals.

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*Electronic address: ilya.tokatly@physik.uni-erlangen.de

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