Time-Dependent Density Functional Theory beyond Linear Response: An Exchange-Correlation Potential with Memory

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We propose a memory form of exchange-correlation potential $v_{\mathrm{XC}}(\mathbf{r}, t)$ for time-dependent interacting many-particle systems. Unlike previous memory-XC potentials, our v_{XC} is not limited to the linear response regime. The proposed form of v_{XC} is a generalized local-density approximation chosen so as to satisfy the nonlinear harmonic potential theorem and Newton's third law. For the case of the inhomogeneous electron gas, we give an explicit prescription for v_{XC} based solely on an existing parametrization of the linear XC response kernel $f_{\mathrm{XC},0}^{\mathrm{hom}}(n, \omega)$ of the uniform gas. Application to quantum wells seems promising. [S0031-9007(97)03984-7]

Widespread interest in nonlinear dynamic phenomena leads us to seek an efficient description of time-dependent exchange and correlation effects in strongly perturbed interacting many-body systems. These effects exhibit a memory in general, and recent work [1,2] on *linear* response suggests that a spatially local memory approximation will only be available if one works in terms of currents and vector potentials, rather than densities and scalar potentials. Here we will show that, at least in the case of one-dimensional spatial variation, a very simple reinterpretation of the local-density approximation, at the *nonlinear* level, can lead to the correct *linear* theory as expounded in Ref. [2].

We start from time-dependent density functional theory [3–5]. With suitable representability conditions, it gives the *exact* density as

$$n(\mathbf{r}, t) = \sum_{k=1}^{N} |\psi_k(\mathbf{r}, t)|^2, \tag{1}$$

where the orbitals $\{\psi_k\}$ obey single-particle-like time-dependent Kohn-Sham (TDKS) equations

$$i \frac{\partial}{\partial t} \psi_k(\mathbf{r}, t) = \left(-\frac{\nabla^2}{2} + v_s[n](\mathbf{r}, t) \right) \psi_k(\mathbf{r}, t), \tag{2}$$

$$v_s[n](\mathbf{r}, t) = v(\mathbf{r}, t) + \int n(\mathbf{r}', t) U(\mathbf{r}, \mathbf{r}') d^3 r'$$
$$+ v_{\mathrm{XC}}[n](\mathbf{r}, t). \tag{3}$$

U is the interparticle interaction, $v(\mathbf{r}, t)$ is the external one-particle potential, and $v_{\mathrm{XC}}(\mathbf{r}, t)$ is the exchange-correlation (XC) potential. For a given interaction U and a given initial wave function, $v_{\mathrm{XC}}(\mathbf{r}, t)$ has a unique delayed, nonlocal dependence on the density $n(\mathbf{r}', t')$. We assume that the initial many-body wave function $\Psi(\mathbf{r}_1, \ldots, \mathbf{r}_N, t_0)$ is the nondegenerate, current-free ground state $\Psi_0(\mathbf{r}_1, \ldots, \mathbf{r}_N)$ of the initial potential $v(\mathbf{r}, t_0) \equiv v_0(r)$ and that the density $n(\mathbf{r}, t \le t_0)$ is the ground-state density $n_0(\mathbf{r})$. The initial density can then

be represented in terms of the ordinary ground-state KS orbitals $\psi_{k,0}(\mathbf{r})$, which therefore also serve as initial orbitals $\psi_k(\mathbf{r}, t_0)$ for (1)–(3).

A common approximation for $v_{\mathrm{XC}}(\mathbf{r}, t)$ is the adiabatic local-density approximation (ALDA), involving locality both in space and in time [6,7],

$$v_{\mathrm{XC}}^{\mathrm{ALDA}}[n](\mathbf{r}, t) = v_{\mathrm{XC}}^{\mathrm{hom}}(n(\mathbf{r}, t)) = \left. \frac{d(n \epsilon_{\mathrm{XC}}^{\mathrm{hom}})}{dn} \right|_{n(\mathbf{r},t)}. \tag{4}$$

Here $\epsilon_{\mathrm{XC}}^{\mathrm{hom}}$ is the XC energy per particle of the homogeneous gas. To date Eqs. (1)–(4) have usually [6–11] been solved for the *linear* density response n_1 to a small external scalar potential v_1. In the linear regime the TDKS equations self-consistently determine the density perturbation n_1 in terms of the external and Hartree potentials plus a linearized XC potential

$$v_{\mathrm{XC},1}(\mathbf{r}, t) = \int f_{\mathrm{XC}}(\mathbf{r}t, \mathbf{r}', t') n_1(\mathbf{r}', t') d^3 r' dt', \tag{5}$$

$$f_{\mathrm{XC}}(\mathbf{r}t, \mathbf{r}'t') = [\delta v_{\mathrm{XC}}(\mathbf{r}t)/\delta n(\mathbf{r}'t')]_{|n_0}. \tag{6}$$

v_{XC} and f_{XC} are not known exactly but Gross and Kohn [12] generalized (4) via a spatially local *memory* approximation,

$$v_{\mathrm{XC},1}^{\mathrm{GK}}(\mathbf{r}, t) = \int_{-\infty}^{\infty} f_{XCL}^{\mathrm{hom}}(n_0(\mathbf{r}), t - t') n_1(\mathbf{r}, t') dt', \tag{7}$$

where $f_{XCL}^{\mathrm{hom}}(n, \tau)$ is the Fourier transform, with respect to frequency, of the longitudinal XC kernel $f_{XCL}^{\mathrm{hom}}(n, \omega)$ of the *homogeneous* gas. For Coulomb systems f_{XCL}^{hom} is related to the local-field-correction factor G by $f_{XCL}^{\mathrm{hom}}(n, \omega) = \lim_{q \to 0}[-(4\pi/q^2)G(n, q, \omega)]$. Gross, Kohn, and Iwamoto [12,13] further derived a useful parametrization of $f_{XCL}^{\mathrm{hom}}(n, \omega)$ which has been used in a number of numerical calculations [11,14]. By contrast, the ALDA yields, via (4) and (6), a frequency-independent f_{XCL}^{hom}. A further analysis of $f_{XCL}^{\mathrm{hom}}(n, \omega)$ has now become available [15].

Recently it was shown [1] that the approximation (7), with spatial locality but time nonlocality, violates an exact condition, the harmonic potential theorem (HPT). (This theorem [1], an extension of the generalized Kohn theorem [16,17], applies to interacting harmonically confined systems. It guarantees existence of a mode involving rigid oscillation, at arbitrarily large amplitude, of the charge density *and many-body wave function*.) In Ref. [1] a modification to the Gross-Kohn theory, based on the current rather than the density alone, was suggested in order to remedy this difficulty. Further work by Vignale and Kohn [2] ("VK") involved a current dependence, but also a vector rather than a scalar XC potential, in order to achieve a straightforwardly local linear theory in the limit of slow spatial variations. In VK the tensor connection between $\mathbf{a}_{XC}$ and $\mathbf{j}$ was determined by explicit enforcement of five different constraints including Newton's third law and the HPT. The resulting expression contains longitudinal and transverse terms depending on the longitudinal and transverse XC kernels f_{XCL}^{hom} and f_{XCT}^{hom} of the uniform electron gas [12,13,15]. The transverse part of VK vanishes for the case of one-dimensional inhomogeneities and currents.

The purpose of the present work is to exhibit and discuss a simple, physically motivated *nonlinear* functional which contains memory and yields the longitudinal part of the linear Vignale-Kohn theory in the limit of small density perturbations. Thus one has both (a) a new physical picture which exhibits the somewhat formal linear VK theory as a modified local-density approximation for the XC force, and (b) a first attempt at the inclusion of memory-XC effects in the nonlinear response regime.

We first consider a straightforwardly local ("SL") but nonlinear and time-delayed approximation for the exchange-correlation potential. By this we mean that v_{XC} has the form

$$ v_{XC}^{\text{SL}}(\mathbf{r}, t) = \int_{-\infty}^{\infty} w_{XC}\big(n(\mathbf{r}, t'), t - t'\big)\, dt', \qquad (8) $$

where $w_{XC}(n, \tau)$ is, as yet, an unknown nonlinear function of two variables, with $w_{XC}(n, \tau < 0) = 0$ for causality. Note that the space variable $\mathbf{r}$ is the same on both sides of (8). A potential of the form (8) was proposed previously [18] by Bünner and Gross, who fixed w_{XC} by reference to the XC kernel f_{XC} of the uniform electron gas. This potential was shown in [18] to change its shape when the density is rigidly boosted, a behavior which has since been shown [19] to be forbidden: it also can be shown to violate the HPT.

We now introduce a simple generalization of the notion of spatial locality which will ensure that a modified form of Eq. (8) satisfies the harmonic potential theorem in its nonlinear form: for this it is sufficient [1,19] that the XC potential move rigidly when the density is rigidly accelerated with arbitrary amplitude. To this end, the new physical input required is the notion that *memory resides not with each fixed point* $\mathbf{r}$ *in space, but rather with each*

separate fluid element. Thus the element which arrives at location $\mathbf{r}$ at time t "remembers" what happened to it at earlier times t' when it was at locations $\mathbf{R}' = \mathbf{R}(t' | \mathbf{r}, t)$ different from its present location $\mathbf{r}$. Here the trajectory function $\mathbf{R}(t' | \mathbf{r}, t)$ of a fluid element is its position at time t', given that its position at time t is $\mathbf{r}$. $\mathbf{R}$ can be defined unambiguously by demanding that its time derivative is the fluid velocity $\mathbf{u} = \mathbf{J}/n$ formed from the quantal current density $\mathbf{J}(\mathbf{r}, t)$,

$$ \frac{\partial}{\partial t'} \mathbf{R}(t' | \mathbf{r}, t) = \mathbf{u}(\mathbf{R}, t') \equiv \mathbf{J}(\mathbf{R}, t')/n(\mathbf{R}, t'), \qquad (9) $$

where all occurrences of $\mathbf{R}$ have the same arguments as on the left side of Eq. (9). The boundary condition on (9) is

$$ \mathbf{R}(t | \mathbf{r}, t) = \mathbf{r}. \qquad (10) $$

Note that, despite the classical appearance of the equations defining $\mathbf{R}$, we do not simply have classical hydrodynamics because the current in Eq. (9) is obtained from the KS orbitals,

$$ \mathbf{J}(\mathbf{r}, \mathbf{t}) = \frac{1}{2i} \sum_{k=1}^{N} (\psi_k^* \nabla \psi_k - \psi_k \nabla \psi_k^*). \qquad (11) $$

[The cases we will be concerned with below (e.g., quantum wells with "edge" electromagnetic excitation) involve three-dimensional systems, but all quantities vary in one space direction only, and all currents are in the same direction. Under these conditions (11) follows exactly from (1)–(3) by continuity.]

The simplest way to incorporate the above idea is to replace $n(\mathbf{r}, t')$ in (8) by $n\big(\mathbf{R}(t' | \mathbf{r}, t), t'\big)$, thereby satisfying the HPT. Unfortunately, unlike (8), this theory violates Newton's third law, which requires the total XC force to be zero at any time [5,19,20],

$$ \int n(r, t) \mathbf{F}_{XC}(r, t)\, d^3 r = 0. \qquad (12) $$

Here $\mathbf{F}_{XC} \equiv -\nabla v_{XC}$ is the XC force per particle. Clearly (12) is satisfied if the XC force per unit volume, $n \mathbf{F}_{XC}$, is a space gradient of form $-\nabla P_{XC}$ where P_{XC} vanishes at the boundaries. With this and the HPT in mind, we introduce the following ansatz for the XC force per particle:

$$ \mathbf{F}_{XC}(r, t) = -\frac{1}{n(\mathbf{r}, \mathbf{t})} \nabla \int_{-\infty}^{\infty} \Pi_{XC}\big(n(\mathbf{R}', t'), t - t'\big)\, dt'. \qquad (13) $$

Here $\mathbf{R}' = \mathbf{R}(t' | \mathbf{r}, t)$ and $\Pi_{XC}(n, \tau)$ is a pressurelike scalar memory function of two variables, whose value will be determined below. Equation (13) is a modified local-density assumption, in that for fixed $\mathbf{r}$, t, and t', v_{XC} depends only on the density at one place $\mathbf{R}'$: "locality" is thereby defined relative to a fluid element rather than to a fixed position $\mathbf{r}$. For consistency with the static LDA we require (13) to reproduce the ground-state XC force,

$$ \nabla v_{XC}^{\text{hom}}(n_0(\mathbf{r})) = \frac{1}{n_0(\mathbf{r})} \nabla \Pi_{XC}(n_0(\mathbf{r}), \omega = 0). \qquad (14) $$

Here $v_{XC}^{\text{hom}}(n) = \frac{d}{dn}[n\epsilon_{XC}^{\text{hom}}(n)]$ is the static LDA XC potential. The form (13) guarantees satisfaction of the nonlinear HPT. To see this, note that in HPT motion under harmonic confinement, the ground-state density $n_0(\mathbf{r})$ suffers a rigid displacement $\mathbf{X}(t)$ so that the density is $n(\mathbf{r}, t) = n_0(\mathbf{r} - \mathbf{X}(t))$. Because the many-body wave function also moves rigidly [1], the current density is $\mathbf{J}(\mathbf{r}, t) = n_0(\mathbf{r} - \mathbf{X}(t)) d\mathbf{X}/dt$. The fluid velocity $\mathbf{u} = \mathbf{J}/n$ is then $d\mathbf{X}/dt$, independent of $\mathbf{r}$. Then Eqs. (9) and (10) show $\mathbf{R}' = \mathbf{r} + \mathbf{X}(t') - \mathbf{X}(t)$. Thus (13) and (14) yield

$$\mathbf{F}_{XC}(\mathbf{r}, t) = -\frac{1}{n_0(\mathbf{r} - \mathbf{X}(t))}$$
$$\times \nabla \Pi_{XC}(n_0(\mathbf{r} - \mathbf{X}(t)), \omega = 0)$$
$$= -\nabla v_{XC}^{\text{hom}}(n_0(\mathbf{r} - \mathbf{X}(t))).$$

That is, the XC potential in the case of HPT motion is the rigidly shifted ground-state XC potential, and this guarantees satisfaction of the HPT [1,19,20].

In the case of 1D inhomogeneity one can go straight from the force (13) to a scalar potential by a spatial integration,

$$v_{XC}(r, t) = -\int_{-\infty}^{r} F_{XC}(r', t)\, dr', \qquad (15)$$

resulting in a highly nonlocal XC potential for use in the TDKS equations. For general 3D variations, with a suitable choice of gauge one can derive an equivalent vector potential $\mathbf{A}_{XC}(r, t) = \int_{-\infty}^{t} \mathbf{F}_{XC}(r, t')\, dt'$ which is then used via a minimal gauge substitution in the Kohn-Sham single-particle Schrödinger equations. There are some technical difficulties with this 3D approach, however, so we will restrict attention here to 1D inhomogeneity, in which case one can choose to use either the scalar or the vector potential as just described, with equivalent results.

Equation (13) is the main result of the present work. It remains to identify the function Π_{XC} for arbitrary ω, and to show that (13) reduces to the linear VK theory in the appropriate limit. Consider small motions at frequency ω around a static equilibrium, in the sense that the displacement $\mathbf{x}(\mathbf{r}, t) \equiv \mathbf{R}(t\,|\,\mathbf{r}, t_0) - \mathbf{r}$ of each fluid element from its initial ($t = t_0$) position $\mathbf{r}$ is small. Then $\mathbf{J}$ in (9) is a small quantity (first order in $\mathbf{x}$ at fixed ω), and (9) can be integrated to give

$$\mathbf{R}(t'\,|\,\mathbf{r}, t) = \mathbf{r} + \mathbf{x}(\mathbf{r}, t') - \mathbf{x}(\mathbf{r}, t) + O(\mathbf{x}^2). \qquad (16)$$

Substituting (16) into (13), using (14) with $f_{XC}^{\text{hom}}(n, \omega = 0) = \partial v_{XC}^{\text{hom}}/\partial n$ and writing $n(\mathbf{r}, t) = n_0(\mathbf{r}) + n_1(\mathbf{r}, t)$ we obtain a first-order perturbation $\mathbf{F}_{XC1}$ to the XC force,

$$\mathbf{F}_{XC1}(\mathbf{r}, t) = \frac{n_1(\mathbf{r}, t)}{n_0(\mathbf{r})} \nabla n_0(\mathbf{r}) f_{XCL}^{\text{hom}}(n_0(\mathbf{r}), \omega = 0)$$

$$-\frac{1}{n_0(\mathbf{r})} \nabla \int_{-\infty}^{\infty} \frac{\partial}{\partial n_0} \Pi_{XC}(n_0(\mathbf{r}), t - t')\{\nabla n_0(\mathbf{r}) \cdot [\mathbf{x}(\mathbf{r}, t') - \mathbf{x}(\mathbf{r}, t)] + n_1(\mathbf{r}, t')\}\, dt'. \qquad (17)$$

Time-Fourier transforming (17) and applying it to the linear response of a uniform electron gas of density n [for which $\mathbf{F}_{XC1} = -f_{XCL}^{\text{hom}}(n, \omega)\nabla n_1(\mathbf{r}, \omega)$, and $\nabla n_0(\mathbf{r}) = \mathbf{0}$] we find

$$\frac{\partial \Pi_{XC}(n, \omega)}{\partial n} = n f_{XCL}^{\text{hom}}(n, \omega),$$

where $f_{XCL}^{\text{hom}}(n, \omega)$ is the well-known [12] longitudinal dynamic XC kernel of the uniform electron gas. Thus

$$\Pi_{XC}(n, \omega) = \int_0^n \rho f_{XCL}^{\text{hom}}(\rho, \omega)\, d\rho, \qquad (18)$$

since $\Pi_{XC} = 0$ for a gas of zero density. Fourier transforming (18) we obtain (see [13])

$$\Pi_{XC}(n, \tau) = \Pi_\infty(n)\delta(\tau) + \Delta\Pi_{XC}(n, \tau), \qquad (19)$$

where $\Pi_\infty(n) = \int_0^n \rho f_{XCL}^{\text{hom}}(\rho, \infty)\, d\rho$. The $\delta(\tau)$ part of (19) is mandated by the third frequency moment sum rule, and from [13] we obtain $\Pi_\infty(n) = -6.537, -2.557, -1.199$, and -0.3711 (10^{-4} Hartree units) for n such that $r_s = 2, 2.5, 3$, and 4, respectively. Figure 1 shows the memory part $\Delta\Pi_{XC}$ within the parametrization of Ref. [13].

Returning to the linear response of a nonuniform system, Fourier transformation and partial expansion of

the outer gradient in the second term of (17) now give

$$\mathbf{F}_{XC1}(\mathbf{r}, \omega) = -\frac{\nabla n_0(\mathbf{r})}{n_0(\mathbf{r})}[n_1(\mathbf{r}, \omega) + \nabla n_0(\mathbf{r}) \cdot \mathbf{x}(\mathbf{r}, \omega)]\delta f$$

$$-\nabla\{\nabla n_0(\mathbf{r}) \cdot \mathbf{x}(\mathbf{r}, \omega)\delta f + n_1(\mathbf{r}, t')f\},$$
$$(20)$$

where $f = f_{XCL}^{\text{hom}}(n_0(\mathbf{r}), \omega)$ and $\delta f = f_{XCL}^{\text{hom}}(n_0(\mathbf{r}), \omega) - f_{XCL}^{\text{hom}}(n_0(\mathbf{r}), \omega = 0)$.

We can now use relations $\mathbf{j}/n_0(\mathbf{r}) = -i\omega\mathbf{x}$ [the linearized form of (9)] and $n_1 = -n_0\nabla \cdot \mathbf{x} - \mathbf{x} \cdot \nabla n_0$ (linearized continuity). By these means we verify that the
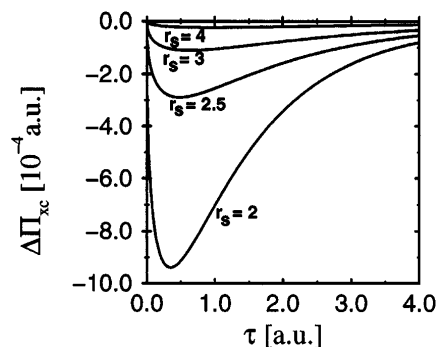

FIG. 1. Memory function $\Delta\Pi_{XC}$ vs delay τ (Hartree units).

linearized form (20) is identical with $-i\omega \mathbf{a}_{1XC}^{\text{long}}(\mathbf{r},\omega)$ where $\mathbf{a}_{1XC}^{\text{long}}(\mathbf{r},\omega)$ denotes the "longitudinal" part of the XC vector potential, obtained from Eq. (19) of Ref. [2] by omission of the transverse terms involving f_{XCT}^{h}. Thus in the case of 1D variations where the transverse terms in Ref. [2] vanish identically, the present nonlinear theory linearizes to give exactly the VK result: this VK form is known to be valid in the linear limit of slow spatial variations.

It may seem surprising that the nonlinear response of an inhomogeneous system is entirely determined, in the present theory, by the linear XC coefficient $f_{XCL}^{\text{hom}}(n,\omega)$ of the uniform gas. This arises because of the bold assumption of the form (13) for the nonlinear XC potential $v_{XC}(\mathbf{r},t)$, depending on the density at only one foreign spacetime point at a time—i.e., no cross dependencies such as $n(\mathbf{r}',t')n(\mathbf{r}'',t'')$ are included. The form (13) can be regarded as the lowest term in a resummed functional Taylor expansion of $v_{XC}(\mathbf{r},t)$ in terms of densities at one foreign spacetime point, two such points, three such points, etc. This notion is discussed in more detail on pp. 127–128 of Ref. [5], where the need to satisfy Eq. (12) was not, however, considered. A consequence of the present form is that the second-order and higher nonlinear terms in an expansion of v_{XC} in powers of the density will contain a restricted form of memory relating to the density at just one previous time, not two, three, . . . , previous times as would be allowed for these expansion coefficients in general. This is a consequence of the very simple "local-with-memory" form of (13). With this restriction, Eq. (13) sums all orders of nonlinear response.

Numerical implementation of the present scheme might appear difficult because of the apparent need to store a two-time quantity $\mathbf{R}(t' \mid \mathbf{r},t)$ along with the TDKS orbitals $\psi_k(\mathbf{r},t)$. In fact, one only needs to use the one-time quantity $\mathbf{X}(\mathbf{r},t) = \mathbf{R}(t \mid \mathbf{r},t_0)$ where t_0 is the initial time. Indeed, it may be more convenient to rewrite the KS equations in terms of KS wave functions ϕ_k relabeled by the initial position of the fluid element,

$$\phi_k(\mathbf{r},t) = \psi_k(\mathbf{X}(\mathbf{r},t),t).$$

Then memory resides separately with each chosen point $\mathbf{r}$.

In summary, we have shown how the concept of a local-density approximation for the XC potential can be extended to the nonlinear time-dependent case with memory, with satisfaction of exact nonlinear constraints, the harmonic potential theorem [1], and Newton's third law. Our scheme follows by postulating that locality should be defined relative to a fluid element rather than to a fixed spatial point $\mathbf{r}$. It is summarized by Eqs. (13),

(18), (9), and (15). It uses as input only the well-known linear longitudinal XC coefficient $f_{XCL}^{\text{hom}}(n,\omega)$ of the uniform gas [12,13,15]. When linearized, it yields the longitudinal part of the Vignale-Kohn form of XC vector potential [2], which is believed to be exact in the limit of slow 1D spatial variations of a 3D system. Immediate applications may arise in the context of recent work [21,22] on nonlinear processes involving 1D motions of the electron gas in semiconductor quantum wells.

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