

## Coordinate scaling in time-dependent current-density-functional theory

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The coupling-constant dependence is derived in time-dependent *current*-density-functional theory. The scaling relation can be used to check approximate functionals and in conjunction with the adiabatic connection formula to obtain the ground-state energy from the exchange-correlation kernel. The result for the uniform gas using the Vignale-Kohn approximation is deduced.

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Time-dependent functional density-functional theory (TDDFT) [1] is developing rapidly as a tool for predicting electronic response to laser fields, both weak and strong [2]. For weak fields, linear response applies, and perhaps the most popular present application of TDDFT is in calculating optical response of molecules, including transition frequencies [3]. For strong fields, TDDFT allows prediction of many properties in response to intense laser pulses, such as high harmonic generation [4]. It seems likely that TDDFT will play a key role in the emerging field of electron quantum control [2].

The time-dependent scheme can also be used for strictly ground-state properties by using the adiabatic connection formula to compute the static exchange-correlation energy [5–7]. Although using TDDFT to calculate ground-state properties might seem an unwarranted complication, the approximate exchange-correlation energy functional within this scheme has many useful properties. For example, it correctly describes the static correlation for bond dissociation [8] and can be used to calculate accurately van der Waals dispersion energies [9–12].

To use TDDFT in the adiabatic connection formula, one must generalize the response functions to arbitrary coupling constant  $\lambda$ . In DFT, this has a very precise meaning, as the density is held fixed while the Coulomb repulsion between electrons is multiplied by  $\lambda$  [5,6]. Coordinate scaling is used to derive the  $\lambda$  dependence of quantities in DFT [13], or *current* DFT [14]. It leads to many useful results, such as the virial theorem for the exchange-correlation energy [13] and exact conditions on that energy [15]. It can be used to check that approximate functionals have the right scaling behavior [16].

Time-dependent *current* density functional theory (TDCDFT) is a more general scheme where the current density is the basic variational parameter instead of the density and it can include arbitrary magnetic fields. Unlike TDDFT, TDCDFT can be approximated with local or semilocal functionals without any conceptual difficulty [17] and is now being used to calculate excitations of quantum wells [18,19], atoms [20], molecules [21–24], and single molecule transport [25,26]. Five years ago, the connection between coordinate scaling and the coupling constant was derived for TDDFT [27]. The present paper generates analogous results within the more general framework of TDCDFT.

TDCDFT [28,29] starts from the Schrödinger equation for  $N$  electrons in a vector potential  $\mathbf{a}_{\text{ext}}$ ,

$$\left\{ \frac{1}{2} \sum_{i=1}^N [\hat{\mathbf{p}}_i + \mathbf{a}_{\text{ext}}(\hat{\mathbf{r}}_i, t)]^2 + \hat{V}_{\text{ext}} + \hat{V}_{\text{ee}} \right\} \Psi = i \frac{\partial}{\partial t} \Psi, \quad (1)$$

where  $\hat{V}_{\text{ext}}$  denotes the one-body potential,  $\hat{V}_{\text{ee}} = \frac{1}{2} \sum_{i,j=1}^N |\hat{\mathbf{r}}_i - \hat{\mathbf{r}}_j|^{-1}$ , and  $\hat{\mathbf{p}}_i = -i\hat{\nabla}_i$ . We use atomic units throughout ( $e^2 = m_e = \hbar = 1$ ), and there is an implicit speed of light constant,  $c$ , included in the vector potential, i.e.,  $\mathbf{B} = c \nabla \times \mathbf{A}$ , where  $\mathbf{B}$  is the usual magnetic field. The physical results from the Schrödinger equation above are invariant under the gauge transformation

$$\bar{v}_{\text{ext}}(\mathbf{r}, t) = v_{\text{ext}}(\mathbf{r}, t) + \frac{\partial \Lambda(\mathbf{r}, t)}{\partial t}, \quad (2)$$

$$\bar{\mathbf{a}}_{\text{ext}}(\mathbf{r}, t) = \mathbf{a}_{\text{ext}}(\mathbf{r}, t) + \nabla \Lambda(\mathbf{r}, t), \quad (3)$$

where  $\Lambda$  is an arbitrary function. The gauge freedom can be used, for example, to remove the scalar potential by choosing  $\partial \Lambda(\mathbf{r}, t) / \partial t = -v_{\text{ext}}(\mathbf{r}, t)$ .

As the density is the conjugate variable to the scalar potential  $v_{\text{ext}}(\mathbf{r}, t)$ , the conjugate variable to the vector potential is the current density

$$\hat{\mathbf{j}}(\mathbf{r}, t) = \frac{1}{2} \sum_{i=1}^N \{ \hat{\mathbf{v}}_i(t) \delta(\mathbf{r} - \mathbf{r}_i) + \delta(\mathbf{r} - \mathbf{r}_i) \hat{\mathbf{v}}_i(t) \}, \quad (4)$$

where  $\hat{\mathbf{v}}_i(t) = \hat{\mathbf{p}}_i + \mathbf{a}_{\text{ext}}(\hat{\mathbf{r}}_i, t)$ . The basic theorem of TDCDFT [28,29] states that, for a given initial wave function, a given  $\mathbf{j}(\mathbf{r}, t)$  is generated by at most one  $\mathbf{a}_{\text{ext}}(\mathbf{r}, t)$ , up to a gauge transformation. The density and the current are related through the continuity equation

$$\frac{dn(\mathbf{r}, t)}{dt} + \nabla \cdot \mathbf{j}(\mathbf{r}, t) = 0. \quad (5)$$

To derive the coupling-constant dependence, we first transform the coordinates to  $(\mathbf{r}_i, t) = (\lambda \mathbf{r}'_i, \lambda^2 t')$  in Eq. (1),

$$\left\{ \frac{1}{2} \sum_{i=1}^N \left( \frac{\hat{\mathbf{p}}'_i}{\lambda} + \mathbf{a}_{\text{ext}}(\lambda \hat{\mathbf{r}}'_i, \lambda^2 t') \right)^2 + \hat{V}'_{\text{ext}, \lambda} + \frac{\hat{V}'_{\text{ee}}}{\lambda} \right\} \Psi'_\lambda = \frac{i}{\lambda^2} \frac{\partial}{\partial t'} \Psi'_\lambda, \quad (6)$$

where the prime means that the quantity is evaluated at  $(\mathbf{r}', t')$ , and the scaled normalized wave function is  $\Psi'_\lambda = \lambda^{3N/2} \Psi(\lambda \mathbf{r}'_1, \dots, \lambda \mathbf{r}'_N, \lambda^2 t')$ . Consistent with Ref. [27], we

define the scaled density by  $n_\lambda(\mathbf{r}, t) = \lambda^3 n(\lambda\mathbf{r}, \lambda^2 t)$ . Now we also define the scaled current density

$$\mathbf{j}_\lambda(\mathbf{r}, t) = \lambda^4 \mathbf{j}(\lambda\mathbf{r}, \lambda^2 t). \quad (7)$$

Continuity [Eq. (5)] remains satisfied for all  $\lambda$ . Multiplying Eq. (6) by  $\lambda^2$  and omitting the primes,

$$\left\{ \frac{1}{2} \sum_{i=1}^N [\hat{\mathbf{p}}_i + \lambda \mathbf{a}_{\text{ext}}(\lambda \hat{\mathbf{r}}_i, \lambda^2 t)]^2 + \lambda^2 \hat{V}_{\text{ext}, \lambda} + \lambda \hat{V}_{\text{ee}} \right\} \Psi_\lambda = i \frac{\partial}{\partial t} \Psi_\lambda. \quad (8)$$

We define  $\mathbf{a}_{\text{ext}}^\lambda[\mathbf{j}, \Psi_0]$  as the vector potential for a system with modified coupling constant  $\lambda$ , which gives rise to current  $\mathbf{j}$  starting from wave function  $\Psi_0$ . Thus we identify

$$\mathbf{a}_{\text{ext}}^\lambda[\mathbf{j}, \Psi_0](\mathbf{r}, t) = \lambda \mathbf{a}_{\text{ext}}[\mathbf{j}, \Psi_0](\lambda\mathbf{r}, \lambda^2 t). \quad (9)$$

Although the  $\lambda$  dependence of the external potentials in Eq. (8) is generally complicated, by virtue of the one-to-one correspondence between current and potentials [28,29], the vector potential appearing in Eq. (8) is that *unique* potential producing current density  $\mathbf{j}(\mathbf{r}, t)$  from the initial wave function  $\Psi_0$ , with electron-electron interaction  $\lambda \hat{V}_{\text{ee}}$ .

Next we apply the same argument to the Kohn-Sham system, where the electrons are noninteracting ( $\hat{V}_{\text{ee}}=0$ ) and  $\mathbf{a}_{\text{ext}}(\mathbf{r}, t)$  is replaced by an effective vector potential,  $\mathbf{a}_s(\mathbf{r}, t)$ , defined to reproduce the same current as the interacting system. Since our previous argument does not depend on the interaction,  $\mathbf{a}_s^\lambda(\mathbf{r}, t)$  also satisfies Eq. (9). And the Hartree vector potential,  $\mathbf{a}_H(\mathbf{r}, t) = -\nabla \int dt' \int d\mathbf{r}' e^2 n(\mathbf{r}', t') / |\mathbf{r} - \mathbf{r}'|$ , satisfies the same scaling. From the definition of the exchange-correlation potential,  $\mathbf{a}_s = \mathbf{a}_{\text{ext}} + \mathbf{a}_H + \mathbf{a}_{\text{XC}}$ , we see that it must obey the same scaling as the other vector potentials,

$$\mathbf{a}_{\text{XC}}^\lambda[\mathbf{j}; \Psi_0, \Phi_0](\mathbf{r}, t) = \lambda \mathbf{a}_{\text{XC}}[\mathbf{j}_{1/\lambda}; \Psi_{0,1/\lambda}, \Phi_{0,1/\lambda}](\lambda\mathbf{r}, \lambda^2 t), \quad (10)$$

where there is also a functional dependence on the initial Kohn-Sham wave function  $\Phi_0$ , from  $\mathbf{a}_s(\mathbf{r}, t)$ . This is the central result of this work.

When the vector potential is irrotational, i.e., can be gauge transformed to a scalar potential, the TDDFT  $\lambda$  dependence of Ref. [27] can be derived from these more general results. From the gauge transformation, Eqs. (2) and (3), one can see that an irrotational vector potential is transformed to a scalar potential through  $\partial \mathbf{a} / \partial t = -\nabla v$ . Inserting Eq. (10), we find

$$\nabla v_{\text{XC}}^\lambda(\mathbf{r}, t) = \lambda \partial \mathbf{a}_{\text{XC}}(\lambda\mathbf{r}, \lambda^2 t) / \partial t = \lambda^3 \nabla_\lambda v_{\text{XC}}(\lambda\mathbf{r}, \lambda^2 t), \quad (11)$$

where  $\nabla_\lambda = \partial / \partial(\lambda\mathbf{r})$ . Requiring the potential to vanish far from the system we recover  $v_{\text{XC}}^\lambda(\mathbf{r}, t) = \lambda^2 v_{\text{XC}}(\lambda\mathbf{r}, \lambda^2 t)$  from Ref. [27]. This relation can also be derived directly from Eq. (8) by the same arguments used for the vector potential.

While Eq. (9) represents the most general form, applicable to all TDCDFT applications, we next look at the special case of the linear response of an electronic system. The susceptibility,  $\chi$ , is usually defined by

$\delta n(\mathbf{r}, t) = \int d\mathbf{r}' dt' \chi(\mathbf{r}, t; \mathbf{r}', t') \delta v(\mathbf{r}', t')$ , where  $\delta n$  is a small change in density due to a small perturbation in the potential,  $\delta v$ . We sometimes represent the previous equation as  $\delta n = \chi * \delta v$ . Since we now have a vector potential, we can generalize the linear response to  $\delta \mathbf{j} = \vec{\chi} * \delta \mathbf{a}$ . We restrict ourselves to applying (time-dependent) perturbations on systems for which the external potentials are static. The response can then be considered as a functional of the ground-state density only, not the current.

The scaling relation for the linear-response exchange-correlation kernel in TDDFT is given in Ref. [30]. In TD-CDFD the tensor analog is defined as  $\vec{f}_{\text{XC}} = \delta \mathbf{a}_{\text{XC}} / \delta \mathbf{j}$  and we can find the scaling relation with the functional differentiation

$$\begin{aligned} \mathbf{a}_{\text{XC}}^\lambda[n + \delta n](\mathbf{r}, t) - \mathbf{a}_{\text{XC}}^\lambda[n](\mathbf{r}, t) \\ = \lambda \{ \mathbf{a}_{\text{XC}}[n_{1/\lambda} + \delta n_{1/\lambda}](\lambda\mathbf{r}, \lambda^2 t) - \mathbf{a}_{\text{XC}}[n_{1/\lambda}](\lambda\mathbf{r}, \lambda^2 t) \} \\ = \lambda \int d\mathbf{r}' dt' \vec{f}_{\text{XC}}[n_{1/\lambda}](\lambda\mathbf{r}, \mathbf{r}', \lambda^2 t - t') \delta \mathbf{j}_{1/\lambda}(\mathbf{r}', t') \\ = \lambda \int (\lambda^3 d\bar{\mathbf{r}})(\lambda^2 d\bar{t}) \vec{f}_{\text{XC}}[n_{1/\lambda}](\lambda\mathbf{r}, \lambda\bar{\mathbf{r}}, \lambda^2(t - \bar{t})) \frac{\delta \mathbf{j}(\bar{\mathbf{r}}, \bar{t})}{\lambda^4}. \end{aligned}$$

Equation (10) implies

$$f_{\text{XC}}^\lambda[n_0](\mathbf{r}, \mathbf{r}', t - t') = \lambda^2 f_{\text{XC}}[n_{0,1/\lambda}](\lambda\mathbf{r}, \lambda\mathbf{r}', \lambda^2(t - t')), \quad (12)$$

or, in frequency space,

$$f_{\text{XC}}^\lambda[n_0](\mathbf{r}, \mathbf{r}', \omega) = f_{\text{XC}}[n_{0,1/\lambda}](\lambda\mathbf{r}, \lambda\mathbf{r}', \omega/\lambda^2). \quad (13)$$

These results are needed to implement the TDCDFT version of the adiabatic connection formula as shown below.

In the special case of a uniform electron gas,

$$f_{\text{XC}}^\lambda[n_0](\mathbf{q}, \omega) = \frac{1}{\lambda^3} f_{\text{XC}}[n_{0,1/\lambda}]\left(\frac{\mathbf{q}}{\lambda}, \frac{\omega}{\lambda^2}\right). \quad (14)$$

The above relation implies that, for a uniform gas, knowing the exchange-correlation kernel as a functional of the density is the same as knowing the coupling-constant dependence; this was used for the equivalent TDDFT case [30,31].

There have been various approximations proposed for  $f_{\text{XC}}$  [32–34] and  $f_{\text{XC}}^\lambda$  [35,36] since they are such important quantities. The main TDCDFT approximate functional currently in use is the Vignale-Kohn (VK) functional [17]. It is a gradient expansion in the current density, and uses as input the  $q \rightarrow 0$  limit of both the longitudinal exchange-correlation kernel,  $f_{\text{XC}}^L(\omega)$  [which is precisely the scalar  $f_{\text{XC}}(\omega)$  of TDDFT], and the transverse kernel,  $f_{\text{XC}}^T(\omega)$  of the uniform gas. We have checked that the VK functional respects the above scaling relation, Eq. (13), provided that  $f_{\text{XC}}^{L,T}(\omega)$  used in constructing the functional also respects the appropriate scaling. The most recent approximation for these kernel components is that of Qian and Vignale [36]. We verified that it satisfies Eq. (14), assuming the Landau parameters are invariant under simultaneous scaling of the density and the coupling constant.

Just as for the exchange-correlation potential, the scaling relation for the exchange correlation can be derived from TDCDFT. When the vector potential is irrotational, the scaling relation of  $f_{XC}^{\vec{\lambda}}$  reduces to that of the scalar kernel  $f_{XC}$  [30], via

$$\begin{aligned} \vec{\nabla}\vec{\nabla}'f_{XC}^{\vec{\lambda}}[n_0](\mathbf{r},\mathbf{r}',\omega) &= \omega^2 f_{XC}^{\vec{\lambda}}[n_0](\mathbf{r},\mathbf{r}',\omega) \\ &= \omega^2 f_{XC}^{\vec{\lambda}}[n_{0,1/\lambda}](\lambda\mathbf{r},\lambda\mathbf{r}',\omega/\lambda^2) \\ &= \lambda^4 \vec{\nabla}_{\lambda}\vec{\nabla}'_{\lambda} f_{XC}[n_{0,1/\lambda}](\lambda\mathbf{r},\lambda\mathbf{r}',\omega/\lambda^2). \end{aligned} \quad (15)$$

Then, since  $f_{XC} \rightarrow 0$  as  $\mathbf{r} \rightarrow \infty$  for any finite system, integration implies that the scaling of the current kernel reduces to the scaling of the scalar kernel,  $f_{XC}^{\vec{\lambda}}[n_0](\mathbf{r},\mathbf{r}',\omega) = \lambda^2 f_{XC}[n_{0,1/\lambda}](\lambda\mathbf{r},\lambda\mathbf{r}',\omega/\lambda^2)$ , as in Ref. [30].

Similarly to the adiabatic connection formula used in ground-state DFT [5–7,37], which relates the exchange-correlation energy to the susceptibility, we introduce the adiabatic connection for the ground state of a system with a static scalar potential using *current* DFT susceptibility

$$E_{XC} = -\frac{1}{2} \int_0^1 d\lambda \int_{-\infty}^{\infty} \frac{d\omega}{2\pi i} \text{Tr}([\vec{\chi}^{\vec{\lambda}} - n_0(\mathbf{r})\mathbf{1}] * \vec{T}); \quad (16)$$

the trace is  $\text{Tr}(\vec{a}) = \int d\mathbf{r} \sum_i a_{ii}(\mathbf{r},\mathbf{r})$  and  $\vec{T} = -\vec{\nabla} V_{ee} \vec{\nabla} / \omega^2$ . The symbol  $\mathbf{1}$  stands for  $\delta(\mathbf{r}-\mathbf{r}')\delta_{ij}$ . The tensor susceptibility is related to the exchange-correlation kernel through [38]

$$\vec{\chi}^{\vec{\lambda}} = \vec{\chi}_s + \vec{\chi}_s(\lambda\vec{T} + f_{XC}^{\vec{\lambda}})\vec{\chi}^{\vec{\lambda}}, \quad (17)$$

where  $\vec{\chi}_s$  is the tensor susceptibility for the Kohn-Sham system

$$\begin{aligned} \chi_{s,ij}(\mathbf{r},\mathbf{r}',\omega) &= n_0(\mathbf{r})\mathbf{1} \\ &+ \sum_{\alpha,\beta} (f_{\alpha} - f_{\beta}) \frac{\phi_{\alpha}^*(\mathbf{r})\vec{\nabla}_i\phi_{\beta}(\mathbf{r})\phi_{\beta}^*(\mathbf{r}')\vec{\nabla}_j\phi_{\alpha}(\mathbf{r}')}{\omega - (\epsilon_{\beta} - \epsilon_{\alpha}) + i\eta}, \end{aligned} \quad (18)$$

where  $f$  is the occupation number, i.e., 1 for an occupied state, 0 for an unoccupied one, and  $\eta$  is infinitesimal. The Kohn-Sham wave functions and energies are denoted by  $\phi$  and  $\epsilon$ .

In the special case of a homogeneous gas, the longitudinal and transverse responses decouple, and reordering the terms within the trace of Eq. (16) shows that only the longitudinal

components contribute to  $E_{XC}$ , i.e., it reduces to the usual scalar case. Lein *et al.* [30] tested a variety of approximations to the scalar  $f_{XC}$  for the uniform gas to see how well they reproduced the known correlation energy. To perform the same test for the VK functional, we first note that, although VK is a gradient expansion in the current, yielding terms of order  $q^2$ , these terms are actually zero order in  $q$  when transformed back to the equivalent scalar kernel via Eq. (15). So we find that VK, inserted in the current adiabatic connection formula, reduces to inserting  $f_{XC}^{\vec{\lambda}}(\omega) = f_{XC}^{\text{unif}}(q \rightarrow 0, \omega)$  in the usual scalar adiabatic connection formula. This approximation was already tested by Lein *et al.*, and is labeled “local RA” in their work. (Although they used a different parametrization [34] from QV [36], the results are unlikely to depend strongly on such details.) They found about a factor of 2 reduction in error relative to the adiabatic local-density approximation (ALDA). We have thus demonstrated that, for the special case of the uniform gas, the VK approximation, inserted in the current adiabatic connection formula, improves over ALDA.

Carrying out a calculation of Eq. (16) on molecules or solids is much more computationally demanding than the usual ground-state calculations with approximate exchange-correlation energy functionals, but is probably not much more expensive than the scalar case. Such calculations are presently being performed [37,39] because the use of the adiabatic connection formula correctly describes the dissociation of molecules [8] and dispersion energies [9–12]. The exchange-correlation kernel of TDCDFT being better suited to local or semilocal approximations than the pure density theory [17], we would expect that it would supersede TDDFT when used within the adiabatic connection formula.

To summarize, we have used coordinate scaling to derive the coupling-constant dependence of the exchange-correlation potential in TDCDFT. We have derived the adiabatic connection formula for TDCDFT, and shown how the VK approximation performs for a uniform gas. We have also given explicit formulas relating both potentials and kernels in TDCDFT to their counterparts in TDDFT. Given both the recent use of TDDFT exchange-correlation kernels in the adiabatic connection formula, for calculating bond dissociation curves, and the use and tests of TDCDFT for excitations in which TDDFT has shown limitations, it is clear that an important application of this work is likely to be realized in the near future.

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