Comment on "Critique of the foundations of time-dependent density-functional theory"

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A recent paper [J. Schirmer and A. Dreuw, Phys. Rev A. **75**, 022513 (2007)] challenges exact time-dependent density-functional theory (TDDFT) on several grounds. We explain why these criticisms are either irrelevant or incorrect, and that TDDFT is both formally exact and predictive.

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Time-dependent density-functional theory (TDDFT) has a rigorous foundation [1–5]. Schirmer and Dreuw (SD) appear to criticize TDDFT on several grounds [6], almost all of which are ultimately conceded by SD themselves. For example, SD state that "… an error is introduced in both the TD and static KS linear response theory if the perturbing (external) potential is given by a nonlocal operator," but this is misleading since such potentials do not exist in DFT. Another example is their long discussion of the problems of the RG action. Such problems were first raised a decade ago [2,7] and resolved shortly after using the Keldysh action [3]. The problems with the action in Ref. [1] have been thoroughly investigated in several works [2–4,8,9]. Finally, SD admit these points, but do not address the current literature.

The sole unresolved criticism of SD is their claim that the Kohn-Sham (KS) equations of TDDFT, even if able to *reproduce* the density evolution of the true interacting system, $n(\mathbf{r},t)$, cannot *predict* that evolution. The KS potential, $v_S(\mathbf{r},t)$, is defined as the unique one-body potential in which noninteracting electrons evolve with density $n(\mathbf{r},t)$. SD claim that their "radical KS" scheme shows that $v_S(\mathbf{r},t)$ functionally depends on the future density, thereby making direct propagation impossible. They further argue that, with neither a variational principle nor some proof of convergence of the TD KS equations for such a potential, TDDFT must be "unfounded." The rest of our Comment addresses this issue: we show where the error lies in SD's arguments and explicitly demonstrate that the KS-TDDFT procedure is indeed predictive.

The flaw in SD's arguments arises from not distinguishing between $v_S(\mathbf{r},t)$ and the exchange-correlation (XC) potential. If $v(\mathbf{r},t)$ is the time-dependent external potential, i.e., the one-body potential applied to the interacting system (which is always known and given), and $v_H(\mathbf{r},t)=\int n(\mathbf{r}',t)/|\mathbf{r}-\mathbf{r}'|d^3r'$ is the Hartree potential, then

$$v_{XC}(\mathbf{r},t)=v_S(\mathbf{r},t)-v_H(\mathbf{r},t)-v(\mathbf{r},t) \tag{1}$$

is known to be a functional of the initial states (both interacting, Ψ_0, and KS, Φ_0) and $n(\mathbf{r},t)$, written as $v_{XC}[n,\Psi_0,\Phi_0](\mathbf{r},t)$. SD argue that, since $v_S(\mathbf{r},t)$ depends on $\partial_t^2 n(\mathbf{r},t)$ (the second time-derivative, which SD denote as "the future"), the possibility of a stringent time propagation is "thwarted"; if the potential functional depends on the "future" of the density, then how could it also possibly predict it? In particular, at the initial time $t=0$, $n(\mathbf{r},0)$, Ψ_0, and Φ_0 are obviously insufficient to determine $\partial_t^2 n(\mathbf{r},0)$, and therefore insufficient to determine $v_S(\mathbf{r},0)$.

What SD miss is that in any TD KS time propagation, $v(\mathbf{r},t)$ is known and prescribed by the physical problem at hand, and it is only $v_{XC}(\mathbf{r},t)$ for which a density-initial-state functional is needed. Herein lies the resolution of the propagation paradox, as the functional dependence of $v_S(\mathbf{r},t)$ generally differs from that of $v_{XC}(\mathbf{r},t)$, in contrast to what is claimed by SD without justification: "A similar temporal nonlocality must be expected for the XC potential…." In particular, $v_{XC}[n,\Psi_0,\Phi_0](\mathbf{r},0)$ depends *purely* on the initial states, as we explicitly show below, and SD's expected dependence on $\partial_t^2 n(\mathbf{r},t=0)$ vanishes. At other times, unlike $v(\mathbf{r},t)$ and $v_S(\mathbf{r},t)$, $v_{XC}(\mathbf{r},t)$ does *not* depend on $\partial_t^2 n(\mathbf{r},t)$: there is a dependence on $\partial_t n(\mathbf{r}t)$ but this can be extracted from $\Phi(t)$, through the continuity equation, and so is available during propagation. (There is also dependence on Ψ_0 and Φ_0 and the earlier history of the density.)

Before demonstrating in general the density- and initial-state dependence of $v_{XC}(\mathbf{r},t)$ ("potential functionals" in SD's notation) and why direct propagation works, we give a simple counterexample to SD. Propagate the time-dependent Schrödinger equation for one electron in some potential from some initial state. Applying SD's "radical KS" scheme and logic, this is impossible, because that potential functionally depends on the future density [e.g., Eq. (75) of SD]. This conclusion is incorrect because $v(\mathbf{r},t)$ is *given*. It is never extracted from the density. Instead, what is needed for standard KS TDDFT is a density-initial-state functional only for $v_{XC}(\mathbf{r},t)$. For one electron, $v_{XC}(\mathbf{r},t)=-v_H(\mathbf{r},t)$ (choosing the KS initial state as the true initial state, the only sensible choice [10]), i.e., a functional of the instantaneous density alone. There is no dependence on $\partial_t^2 n(\mathbf{r},t)$, contrary to the "expectations" of SD. More generally, because only the density dependence of $v_{XC}(\mathbf{r},t)$ is needed in any TDDFT KS calculation, their result for $v_S(\mathbf{r},t)$ is irrelevant to the question of propagation.

First, to clarify the question of "future" dependence, we note that a dependence on $\partial_t^2 n(\mathbf{r},t)$ does not violate causality

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at any finite t. For any $0<t<T$, $v[n;\Psi_0](\mathbf{r},t)$ can depend only on the density in the interval $[0,T]$ and not on later times because, by the RG theorem, two potentials that are the same in $[0,T]$ but differ for $t>T$ must give different densities at times $t>T$, i.e., v is a causal functional of the density. The same holds for $v_S[n;\Phi_0](\mathbf{r},t)$. From Eq. (1), $v_{XC}[n;\Psi_0,\Phi_0](t)$ is then also a causal functional of the density and initial states for any finite t, and $\partial_t^2 n(\mathbf{r},t)$ may be evaluated to the left of time t [e.g., using $n(t),n(t-\Delta t),n(t-2\Delta t)$]; but this argument cannot be applied at $t=0$, and so the start of a propagation appears problematic.

To see why this is in fact not a problem, we will use results from Ref. [5] and the notation that a superscript (k) indicates the kth time derivative at $t=0$, e.g., $n^{(k)}(\mathbf{r}) = \partial_t^k n(\mathbf{r}t)|_{t=0}$. Begin by noting that both $n(\mathbf{r},0)$ and $n^{(1)}(\mathbf{r})$ are determined by Ψ_0, the initial wave function of the interacting problem, because of continuity: $\partial_t n(\mathbf{r},t)=-\nabla\cdot\mathbf{j}(\mathbf{r},t)$. This in turn restricts allowed choices of Φ_0 to only those that recover these values. The form of $v(\mathbf{r},t)$ influences only second and higher time derivatives of $n(\mathbf{r},t)$. The TD Schrödinger equation implies [5]

$$\partial_t^2 n(\mathbf{r},t) = \nabla\cdot[n(\mathbf{r},t)\,\nabla\, v(\mathbf{r},t)] + q(\mathbf{r},t), \tag{2}$$

where

$$q(\mathbf{r},t) = \langle\Psi(t)|\hat{\tau}(\mathbf{r}) + \hat{w}(\mathbf{r})|\Psi(t)\rangle, \tag{3}$$

and

$$\hat{\tau}(\mathbf{r}) = \frac{1}{2}\sum_{i,k}\partial_i\partial_k\Big(\partial_i\hat{\psi}^\dagger(\mathbf{r})\partial_k\hat{\psi}(\mathbf{r}) + \partial_k\hat{\psi}^\dagger(\mathbf{r})\partial_i\hat{\psi}(\mathbf{r}) - \frac{1}{2}\partial_i\partial_k[\hat{\psi}^\dagger(\mathbf{r})\hat{\psi}(\mathbf{r})]\Big),$$

$$\hat{w}(\mathbf{r}) = \sum_k \partial_k\int d^3r'\,\hat{\psi}^\dagger(\mathbf{r})\hat{\psi}^\dagger(\mathbf{r}')\partial_k\frac{1}{|\mathbf{r}-\mathbf{r}'|}\hat{\psi}(\mathbf{r}')\hat{\psi}(\mathbf{r}).$$

An analogous equation applies to the KS system, with $v_S(\mathbf{r}t)$ and $q_S(\mathbf{r},t)=\langle\Phi(t)|\hat{\tau}(\mathbf{r})|\Phi(t)\rangle$. Requiring that the density evolutions be the same yields

$$\nabla\cdot[n(\mathbf{r},t)\,\nabla\, v_{HXC}(\mathbf{r},t)] = q(\mathbf{r},t) - q_S(\mathbf{r},t), \tag{4}$$

where $v_{HXC}(\mathbf{r},t)=v_H(\mathbf{r},t)+v_{XC}(\mathbf{r},t)$. This equation is of Sturm-Liouville form: such equations have a unique solution for $v_{HXC}(\mathbf{r},t)$ if $n(\mathbf{r},t)$ and $q(\mathbf{r},t)-q_S(\mathbf{r},t)$ are given, together with the boundary condition that $v_{HXC}(r\to\infty,t)\to 0$. We shall assume this boundary condition for all that follows; any choice of a TD constant for the asymptotic potential does not affect the density [5]. Thus a KS potential can always be found for any density for the interacting system, provided the initial conditions are met.

We now use Eq. (4) to show that at each time step, the functional input to $v_{XC}(\mathbf{r},t)$ consists of the Ψ_0, Φ_0, and the density evolved through previous times. At $t=0$,

$$\nabla\cdot[n(\mathbf{r},0)\,\nabla\, v_{HXC}(\mathbf{r},0)] = q(\mathbf{r},0) - q_S(\mathbf{r},0). \tag{5}$$

Since $q(\mathbf{r},0)$ and $q_S(\mathbf{r},0)$ are determined by Eq. (3), which is known entirely from the initial wave functions, the solution of this equation determines $v_{XC}(\mathbf{r},0)$. [The Hartree potential $v_H(\mathbf{r},0)$ is determined as usual directly from the instantaneous density.] Thus, as mentioned earlier, $v_{XC}(\mathbf{r},0)$ depends on the initial states alone: the dependence on the second time derivative of the density in Eq. (2) and its KS counterpart cancel once their difference is taken in $v_{HXC}[n,\Psi_0,\Phi_0](\mathbf{r},0)$. Together with $v(\mathbf{r},0)$, this evolves Φ_0 forward one time step:

$$\left(-\frac{1}{2}\nabla^2 + v_{HXC}(\mathbf{r},0) + v(\mathbf{r},0)\right)\phi_i(\mathbf{r},0) = i\partial_t\phi_i(\mathbf{r},0). \tag{6}$$

This yields the orbitals at the first time step, $\phi_i(\mathbf{r},\Delta t) = \phi_i(\mathbf{r},0)+\partial_t\phi_i(\mathbf{r},0)\Delta t$, from which one can obtain the density evolved to the first time step, $n(\mathbf{r},\Delta t)=\Sigma_i|\phi_i(\mathbf{r},\Delta t)|^2$, as well as the current at the first time step, $\mathbf{j}(\mathbf{r},\Delta t) = \mathrm{Im}\,\Sigma_i\phi_i^*(\mathbf{r},\Delta t)\nabla\phi_i(\mathbf{r},\Delta t)$, and, through the equation of continuity, also the first time derivative of the density $\dot{n}(\mathbf{r},\Delta t)=-\nabla\cdot\mathbf{j}(\mathbf{r},\Delta t)$.

Next, consider $t=\Delta t$, for which we need to find $\partial_t v_{HXC}(\mathbf{r},0)$. Take a time derivative of Eq. (4), evaluating the terms at $t=0$:

$$\nabla\cdot[n(\mathbf{r},0)\,\nabla\,\partial_t v_{HXC}(\mathbf{r},0)] = -\nabla\cdot[\partial_t n(\mathbf{r},0)\,\nabla\, v_{HXC}(\mathbf{r},0)] + \partial_t[q(\mathbf{r},0) - q_S(\mathbf{r},0)], \tag{7}$$

where

$$\partial_t[q(\mathbf{r},0) - q_S(\mathbf{r},0)] = i\langle\Phi_0|[\hat{\tau}(\mathbf{r}),\hat{H}_S(0)]|\Phi_0\rangle - i\langle\Psi_0|[\hat{q}(\mathbf{r}),\hat{H}(0)]|\Psi_0\rangle. \tag{8}$$

Equation (7) is again of Sturm-Liouville form, with a unique solution for $\partial_t v_{HXC}(\mathbf{r},0)$: all the other quantities in the equation are known either from the initial states or the results of previous time propagation. Equation (7) is not an explicit functional of the initial states and density alone, due to the appearance of $v(0)$ in the commutators in Eq. (8); but evaluating Eq. (2) at $t=0$ gives the functional dependence of $v(0)$ on Ψ_0, $n(\mathbf{r},0)$, and $\partial_t^2 n(\mathbf{r},0)$. Since $\partial_t^2 n(0)=[\partial_t n(\Delta t) - \partial_t n(0)]/\Delta t$ (limit $\Delta t\to 0$ understood), the appearance of the second derivative at $t=0$ implies only a dependence on the first derivative at $t=\Delta t$, directly available from $\Phi(\Delta t)$ via the equation of continuity. So

$$v_S(\mathbf{r},\Delta t) = v(\mathbf{r},\Delta t) + v_{HXC}(\mathbf{r},0) + \partial_t v_{HXC}(\mathbf{r},0)\Delta t \tag{9}$$

is determined and predicts the time evolution of the density at $t=2\Delta$.

Each subsequent time derivative of Eq. (4) produces one higher time derivative of the XC potential, such that $v_{HXC}^{(k)}(\mathbf{r})$ is determined solely by the initial states and $n^{(l)}(\mathbf{r})$ with $l \leqslant (k+1)$, all available from propagation to the kth time step:

$$\nabla\cdot[n(\mathbf{r},0)\,\nabla\, v_{HXC}^{(k)}(\mathbf{r})] = Q^{(k)}(\mathbf{r}), \tag{10}$$

$$Q^{(k)}(\mathbf{r}) = q^{(k)}(\mathbf{r}) - q_S^{(k)}(\mathbf{r}) - \sum_{l=0}^{k-1}\binom{k}{l}\nabla\cdot[n^{(k-l)}(\mathbf{r})\,\nabla\, v_{HXC}^{(l)}(\mathbf{r})]. \tag{11}$$

The $q^{(k)}(\mathbf{r})$ and $q_S^{(k)}(\mathbf{r})$ involve multiple commutators of the operator $\hat{q}(\mathbf{r})$ with the true and KS Hamiltonians, respectively, and their time derivatives, sandwiched between the

initial states Ψ_0, Φ_0, respectively. For example, for $k=2$,

$$q^{(2)}(\mathbf{r}) - q_S^{(2)}(\mathbf{r}) = -\langle\Psi_0|[[\hat{q}(\mathbf{r}),\hat{H}(0)],\hat{H}(0)]|\Psi_0\rangle + \langle\Phi_0|[[\hat{\tau}(\mathbf{r}),\hat{H}_S(0)],\hat{H}_S(0)]|\Phi_0\rangle - i\langle\Psi_0|[\hat{q}(\mathbf{r}),d\hat{H}/dt(0)]|\Psi_0\rangle + i\langle\Phi_0|[\hat{\tau}(\mathbf{r}),d\hat{H}_S/dt(0)]|\Phi_0\rangle.$$

The $v(0)$ and $dv(0)/dt$ appearing in the commutators are causal density and initial-state functionals via Eq. (2), so Eqs. (10) and (11) yield the XC potential as a causal implicit functional of purely the density and initial states. We reiterate that a dependence on $\partial_t n(\mathbf{r},t)$ presents no difficulty during propagation. As one propagates, one can evaluate this instantaneously by simply computing the divergence of the current density of $\Phi(t)$. Thus all quantities needed are available from the past propagation.

So we have shown that the propagation can be done in a predictive manner, and that the expressions for $v_{XC}[n,\Psi_0,\Phi_0](\mathbf{r},t)$ that we give are causal functionals of the density and initial state: explicit at $t=0$ [Eq. (5)] and implicit at later times [via $v[n,\Psi_0]$, Eqs. (4), (7), (8), and (11)]. This construction holds for any time-dependent potentials and densities that are equal to their Taylor expansions for $t\geqslant 0$ for a finite period of time [as assumed for $v(\mathbf{r},t)$ in the Runge-Gross one-to-one mapping proof]. Full analyticity is not required: in particular, as nothing is assumed for times earlier than $t=0$, this procedure applies to sudden switch-on potentials.

The XC potentials used in practical applications may be viewed as approximations to this formally exact construction of the potential functional. Although most of the applications to date have utilized adiabatic approximations, depending on the instantaneous density alone, memory dependence is a well-recognized feature of time-dependent functionals (see, e.g., Refs. [11–16]) and memory-dependent functionals have been successfully applied to real calculations. There are also explicit systematic methods based on many-body perturbation theory to construct approximate XC potentials for practical applications [17]. The equations for these approximate potentials also show the fundamental property that we demonstrated above: the XC potentials at a given time are completely determined by the density evolved up to and including the present time and the initial states.

Last, we give an example to show explicitly the error in SD for an interacting case. We consider two electrons in one dimension, and we need look only at $t=0$. In one dimension, Eq. (5) reduces to

$$v_S(x,0) = v_{ext}(x,0) + \int^x \frac{dx'}{n(x',0)} \int^{x'} dx''[q(x,0) - q_S(x,0)]; \tag{12}$$

but inversion of the time-dependent KS equation [18,19], as in SD, yields here

$$v_S(x,0) = -\frac{1}{2}\left(\frac{\partial_x n}{2n}\right)^2 - \frac{1}{2}\left(\frac{j}{n}\right)^2 + \frac{\partial_x^2 n}{4n} - \int^x \left(\frac{\partial_t j}{n} + \frac{\partial_x j^2}{2n^2}\right)dx', \tag{13}$$

where $j=j(x,0)$ is the initial current density, determined from the initial wave function. (We drop the spatial and $t=0$ indices on the right, for ease of reading.) The alleged dependence on the future arises through the term $\partial_t j$ on the right: this may be equivalently written in terms of $\partial_t^2 n(x,0)$, and is the only term not directly obtainable from the initial states.

However, this future dependence disappears as soon as we relate the KS system to the interacting system via the Heisenberg equation of motion for the current of the interacting system:

$$\partial_t j = -n\partial_x v_{ext} - T_{xx} - W_x, \tag{14}$$

where $\partial_x T_{xx} = \langle\Psi_0|\hat{\tau}|\Psi_0\rangle$ and $\partial_x W_x = \langle\Psi_0|\hat{w}|\Psi_0\rangle$. Substituting this for $\partial_t j$ in the right-hand side of Eq. (13),

$$-\int^x \frac{\partial_t j}{n}dx' = \int^x \frac{dx'}{n(x',0)} \int^{x'} \partial_t^2 n(x'',0)dx'' = v_{ext}(x,0) + \int^x \frac{T_{xx}+W_x}{n}dx', \tag{15}$$

i.e., the "future" dependence is in the external potential and other terms at $t=0$. That is, once the connection with the interacting system is made, the apparent dependence on the future evaporates as $\partial_t^2 n(x,0)$ is determined by initial-state information and by the external potential that the interacting system is subjected to. The dependence is explicit in this two-electron example, but in the general N-electron case, the construction of Ref. [5] implies that this is always true.

Inversion of the KS equations alone yields information only about the KS potential as a functional of the density, but tells nothing about the XC potential. It cannot because it contains no information about any interacting system. This is most easily seen in the ground-state problem. One can trivially invert the single KS orbital equation for any two-electron density and get its KS potential, but this tells nothing about XC unless the corresponding external potential to subtract from it is known; and there is no way to find that without inverting the interacting Schrödinger equation, thereby making the functional dependence as implicit as in the original definition [18,19].

Before concluding, we briefly review SD's discussion of the lack of a proof of numerical convergence of KS propagation when nonadiabatic functionals are used (the "trajectory mode" in SD's notation). SD only claim that such a proof is needed after they incorrectly deduce that the "potential functional" propagation mode fails. There are no proofs of the existence of solutions and their convergence for, e.g., the Navier-Stokes equations, but should they be "abandoned?" (In fact, the KS propagation has been explicitly converged in a recent calculation with a nonadiabatic potential [16].)

To summarize, (i) SD's rejection of the original RG formulation of TDDFT originates in a logical error in their conclusions from their "radical KS" scheme, (ii) this erroneous conclusion led SD to dismiss without argument the constructive proof of the XC potential that has long existed in the literature, in which it is clear that the potential depends only on the past, leading to TD KS equations that are indeed predictive, (iii) finding a general proof of convergence for TDDFT propagation would be interesting, but its foundations do not depend on this, and (iv) the one-to-one density-potential mapping in no way depends on the action functional proposed in RG. Even though a rigorous action principle has been proven within the Keldysh formalism [3], as well as in real time [20], it is not needed to prove that the theory is predictive. Thus the illusion is not TDDFT, as claimed by SD, but the apparent dependence on the future.

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