Exact nonadiabatic part of the Kohn-Sham potential and its fluidic approximation

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(Received 15 November 2019; accepted 25 February 2020; published 16 March 2020)

We present a simple geometrical "fluidic" approximation to the nonadiabatic part of the Kohn-Sham potential, $v_{\rm KS}$, of time-dependent density-functional theory (DFT). This part of $v_{\rm KS}$ is often crucial, but most practical functionals utilize an adiabatic approach based on ground-state DFT, limiting their accuracy in many situations. For a variety of model systems, we calculate the exact time-dependent electron density and find that the fluidic approximation corrects a large part of the error arising from the "exact adiabatic" approach, even when the system is evolving far from adiabatically.

DOI: 10.1103/PhysRevMaterials.4.035002

I. INTRODUCTION

Time-dependent Kohn-Sham (KS) density-functional theory [1–3] (TDDFT) is in principle an exact and efficient theory of the dynamics of systems of interacting electrons. In practical applications, while performing well in some cases, its validity is often restricted by the limitations of available approximate functionals for electron exchange and correlation (xc). Typically, an adiabatic approximation to the xc potential is used, in which the instantaneous electron density is implicitly assumed to be in its ground state, thereby neglecting all "memory effects." While these ground-state approximations have steadily improved [3–14], by definition they cannot approach the exact TDDFT potential: It is necessary to address the nonadiabatic contributions for TDDFT to be capable of predictive accuracy in relation to a multitude of applications to diverse fields such as the determination of electronic excitation energies, including those of a charge-transfer nature [15], electron dynamics [16] including nonperturbative charge transfer dynamics [17], time-resolved spectroscopy [18], and electron scattering [19].

In this paper, to clearly distinguish between adiabatic and nonadiabatic contributions, we consider the purest application of the concept of the adiabatic functional to the *complete* KS potential, $v_{\rm KS}$: at each instant, the DFT KS potential whose *ground-state* density is equal to the exact time-dependent density. The remainder of the exact $v_{\rm KS}$ constitutes the unambiguously nonadiabatic part, to which we also propose an approximation.

We work in the Runge-Gross formalism [1] of TDDFT, in which the exact xc potential, $v_{\rm xc}$, at time t [20] depends on the density at all points in space and all nonfuture times. It has been argued [21–24] that the exact nonadiabatic functional often requires strong nonlocal temporal and spatial dependence on the density. A number of properties of the exact functional, such as the harmonic potential theorem (HPT) [21] and zero-force theorem (ZFT) [22], have been used to identify limitations of previous approximate TDDFT functionals. Adiabatic functionals trivially satisfy many of these exact conditions through their complete lack of memory dependence, yet prove inadequate in many applications [15–19,25–35]. The development of nonadiabatic functionals that continue to satisfy these exact properties is nontrivial. For example, it was shown that modifying the adiabatic local density approximation by introducing time nonlocality, such as in the Gross-Kohn [36] approximation, is inappropriate [21,22].

The best-known approximate nonadiabatic functional is that developed by Vignale and Kohn [24,37,38] (VK). This was constructed by studying the responses to slowly varying perturbations of the homogeneous electron gas, and they found a time-dependent xc vector potential as a functional of the local current and charge densities j and n, thereby implicitly obtaining a scalar potential which depends nonlocally on the density. While the VK formalism has proved promising [39–49], not least through it obeying the HPT and ZFT, its validity is limited [50–54], owing to the constraints under which it was derived.

II. CALCULATIONS

Our calculations employ the iDEA code [55] which solves the many-electron Schrödinger equation exactly for small, one-dimensional prototype systems of spinless electrons [56,57]. This gives us access to the exact electron density $n(x,t)$. We then determine the exact $v_{\rm KS}(x,t)$ through reverse engineering [58]. We also obtain the *exact adiabatic* KS potential [26,34,59] $v_{\rm KS}^{\rm A}$ by applying *ground-state* reverse engineering to the instantaneous density at each time [60]. The *exact nonadiabatic* component $\Delta v_{\rm KS}$ is then $v_{\rm KS} - v_{\rm KS}^{\rm A}$.

A. Fluidic approximation

In developing an approximation to $\Delta v_{\rm KS}$, it is helpful to consider the situation in different inertial frames, related through a Galilean transformation, as noted by Tokatly *et al.* [31,61–64]. While $v_{\rm KS}^{\rm A}$ requires zero correction in any inertial frame when the density is fully static in one of these frames, in the more general case the nonadiabatic corrections to $v_{\rm KS}^{\rm A}$ may be expected to be at their smallest in the local, instantaneous rest frame of the density, defined by a transformation velocity of the local velocity field $u(x,t) = j(x,t)/n(x,t)$. In particular, the effects of acceleration ($\dot{u} \neq 0$) and dispersion ($\partial_x u \neq$

0) have the least effect in a frame where u itself is zero [65]. Conveniently, introducing a vector potential $A = -u(x,t)$ in the original frame of reference is (apart from an unimportant temporal phase factor) equivalent to a Galilean transformation to the local instantaneous rest frame [61,62,66]. As described above, the nonadiabatic correction should be minimal in the latter frame, and here we adopt the simple assumption that it is zero. We term this the *fluidic* approximation. The resulting nonadiabatic correction in the original frame is therefore

$$\Delta v_{\mathrm{KS}}(x, t > 0) = -\int_{-\infty}^{x} \frac{\partial}{\partial t} u(x', t > 0)\, dx', \qquad (1)$$

where we have gauge transformed A into a scalar potential. It is evident that the density dependence of this Δv_{KS} is nonlocal in both space and time [24].

B. System 1

As a first test of the fluidic approximation, we consider two interacting electrons in a potential well, which takes the form of an inverted Gaussian function. Initially, in the ground state, a uniform electric field, $-\varepsilon x$, is applied at $t = 0$, driving the electrons to the right and inducing a current [Fig. 1(a)]. The sudden application of the perturbation means that we are well outside of the adiabatic limit, and this can be seen by solving the time-dependent KS equations with the exact adiabatic KS potential, $v_{\mathrm{KS}}(t) = v_{\mathrm{KS}}^{\mathrm{A}}(t)$. By plotting the change in the electron density from the ground state, δn, we find $v_{\mathrm{KS}}^{\mathrm{A}}(t)$ on its own to be wholly inadequate ($\approx$13% error in n [67] at $t = 8$ a.u.), while adding the fluidic approximation substantially reduces this error to less than 1% [Fig. 1(b)].

To understand these results, we analyze the nonadiabatic correction to the KS potential in both its scalar and its vector forms. We find very good agreement between the exact ΔA_{KS} and that obtained using the fluidic approximation $-u(x,t)$ [Fig. 2(a)]. The velocity field u [the negative of the fluidic curve in Fig. 2(a)] quickly becomes strongly nonuniform in both space and time as the electrons explore excited states—far removed from a universal rest frame. Similarly close agreement between the exact and fluidic Δv_{KS} [Fig. 2(b)] is evident when the nonadiabatic correction is cast into its scalar form through Eq. (1).

C. Systems 2A, 2B, 2C

We now consider a set of systems of interacting electrons in atomiclike external potentials which decay much more slowly at large x, $v_{\mathrm{ext}} = -a/(|x| + a)$ with $a = 20$, thereby increasing correlation. At time $t = 0$, a static sinusoidal perturbation of the form $\varepsilon \cos(0.75x)$ is applied, where ε is 0.02 for system 2A (two electrons), 0.02 for system 2B (three electrons), and 0.1 for system 2C (three electrons).

In system 2A, the sudden perturbation at $t = 0$ acts to push the two electrons apart. This results in a velocity field that is varying in both space and time, as in system 1; in this case, even the sign of u is not the same for all x, which takes us even further away from a universal rest frame. Correspondingly, we find the exact adiabatic potential to be insufficient ($\approx$5% error in n at $t = 5$ a.u.), while adding the fluidic approximation reduces this error to $\approx$1%. System 2B contains three interacting electrons in the same v_{ext} as system 2A. The additional electron results in a ground-state density that is much less spatially uniform. We run the simulation for 5 a.u. of time and find similar results: $v_{\mathrm{KS}}^{\mathrm{A}}$ produces an error in n of $\approx$5%, and the fluidic approximation reduces this to $\approx$1%.

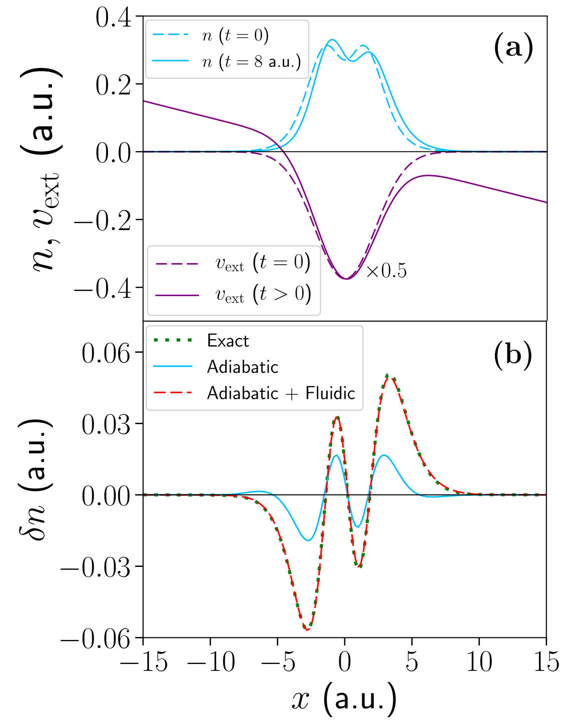

FIG. 1. System 1: Two interacting electrons in a Gaussian potential well, with a uniform electric field applied at $t = 0$, driving the electrons to the right and inducing a current. (a) The ground-state external potential (dashed purple) and exact ground-state electron density (dashed blue), along with the perturbed external potential (solid purple) and exact time-dependent electron density at $t = 8$ a.u. (solid blue). (b) The change in the exact electron density [$\delta n(x,t) = n(x,t) - n(x,0)$] at $t = 8$ a.u. (short-dashed green), along with that obtained when using the exact $v_{\mathrm{KS}}^{\mathrm{A}}$ (solid blue), and when adding the exact $v_{\mathrm{KS}}^{\mathrm{A}}$ with the fluidic approximation $\Delta A_{\mathrm{KS}} = -u$ (dashed red). The exact adiabatic potential is clearly inadequate, but its error is substantially reduced by the fluidic approximation.

As mentioned above, the fluidic approximation assumes that a system remains close to its ground state in the local instantaneous rest frame. To stretch this approximation severely, in system 2C the perturbing potential is much stronger, resulting in a much larger response of the density [Fig. 3(a)]. The fluidic approximation still succeeds in reducing the error in the density, from $\approx$25% where only the exact adiabatic potential is used, to $\approx$6% at $t = 5$ a.u. [Fig. 3(b)]. At later times, the dynamic (time-dependent) xc effects become very significant. To confirm this, we replace the xc component of the exact time-dependent v_{KS} with the fixed ground-state v_{xc}, thereby suppressing the dynamic part, and find this potential to be wholly inadequate ($\approx$62% error in n at $t = 18$ a.u.). Here, the exact adiabatic KS potential is better ($\approx$17% error), while adding the fluidic approximation improves it further ($\approx$15% error) [Fig. 3(c)].

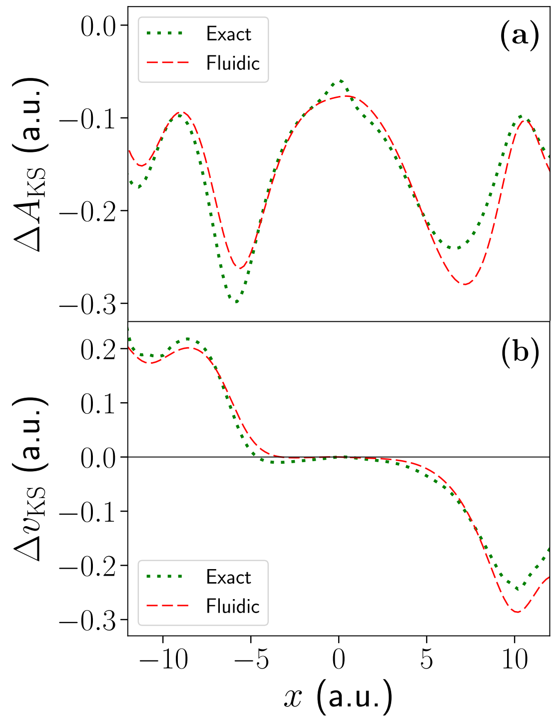

FIG. 2. The nonadiabatic correction to the KS potentials for system 1. (a) The exact $\Delta A_{\rm KS}$ (short-dashed green) and that obtained using the fluidic approximation $\Delta A_{\rm KS} = -u$ (dashed red), at $t = 8$ a.u. (b) The corresponding exact (short-dashed green) and fluidic (dashed red) $\Delta v_{\rm KS}$ in its scalar form. The fluidic approximation performs very well, even though the velocity field is non-uniform in both space and time. (The exact adiabatic approximation, of course, amounts to setting $\Delta A_{\rm KS} = \Delta v_{\rm KS} = 0$.)

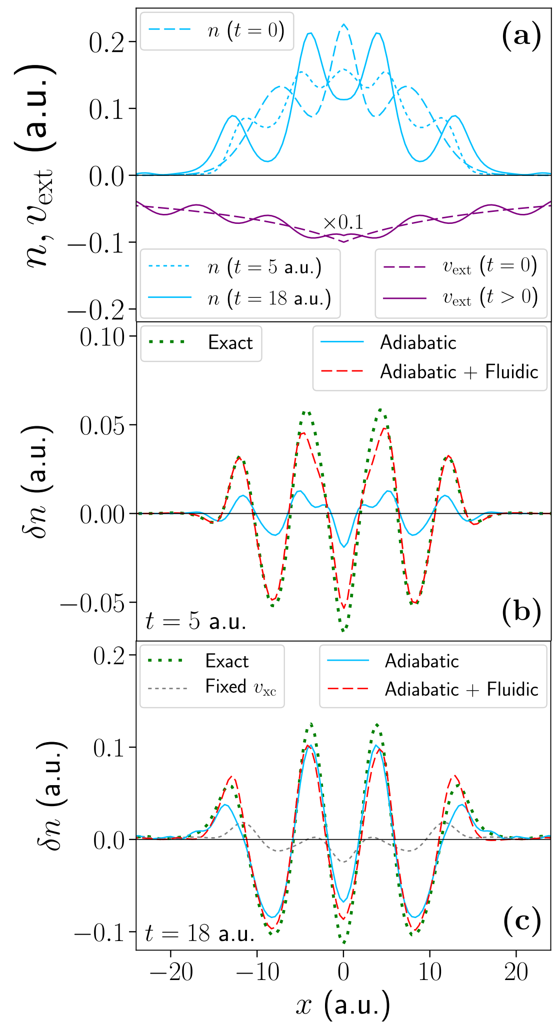

FIG. 3. System 2C: Three interacting electrons in an atomiclike potential, with a static sinusoidal perturbation applied at $t = 0$, pushing the electrons apart. (a) The ground-state external potential (dashed purple) and exact ground-state electron density (dashed blue), along with the perturbed external potential (solid purple) and exact time-dependent electron density at $t = 5$ a.u. (short-dashed blue) and $t = 18$ a.u. (solid blue). (b) The change in the exact electron density at $t = 5$ a.u. (short-dashed green), along with that obtained when using the exact $v^{\rm A}_{\rm KS}$ (solid blue), and when adding the exact $v^{\rm A}_{\rm KS}$ with the fluidic approximation (dashed red). Even though the density is strongly disrupted, the fluidic approximation remains successful. (c) The same as (b) but at $t = 18$ a.u., where the dynamic xc contribution is very significant, evident by the completely inadequate result obtained with the fixed $v_{\rm xc}$ (short-dashed gray) (see main text). Here, the exact $v^{\rm A}_{\rm KS}$ is better, but adding the fluidic approximation improves it further.

D. Exact conditions

A number of properties of the exact xc functional are known, and these are often used to identify the limitations of approximate functionals. We now explore whether the fluidic approximation satisfies these exact conditions.

We begin with the *one-electron limit*, where the exact xc functional, when applied to a one-electron system, reduces to the negative of the Hartree potential $v_{\rm H}$, thereby canceling the spurious self-interaction. This means that $v_{\rm KS}$ is described exactly by a known functional [16,26,34], which has been termed [68] the single orbital approximation—itself capable of capturing features such as steps in the KS potential [16,69]—whose nonadiabatic part is

$$\Delta v_{\rm KS}(x,t) = -\int_{-\infty}^{x} \frac{\partial}{\partial t} u(x',t)\, dx' - \frac{1}{2} u^2(x,t). \quad (2)$$

We note that the first term is the fluidic approximation [Eq. (1)]. We have studied systems of one electron in the external potentials from systems 1, 2A, and 2C, and confirm that the full Eq. (2) yields the exact $v_{\rm KS}$; here, the effect on the density of including the $-u^2/2$ term ranges from <0.1% (potential 2A) to 14% (potential 2C), so the fluidic approximation alone is already satisfactory. Indeed, in our two- and three-electron systems, the effect of adding the additional term to the fluidic approximation is small and typically slightly deleterious.

The ZFT [22] follows from Newton's third law and requires the net force exerted on the system by $v_{\rm H}$ and $v_{\rm xc}$ to vanish. At the level of the KS potential, $\int n(x,t) \partial_x \Delta v_{\rm KS}(x,t)\, dx = \int n(x,t) \partial_x v_{\rm ext}(x,t)\, dx$, since the exact $v^{\rm A}_{\rm KS}$ satisfies the theorem in its own right. In the fluidic approximation for system 1 [70], the left- and right-hand sides of this equation are within 11% of one another so the theorem appears to be approximately obeyed.

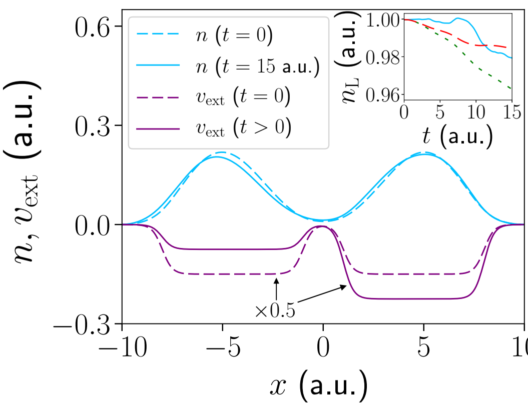

FIG. 4. System 3: Two interacting electrons in a tunneling system. Inset: The exact total electron number on the left-hand side ($x < 0$) (short-dashed green); also the exact adiabatic (solid blue) and fluidic approximation (dashed red).

The HPT [21] shows that in a system of interacting electrons in a harmonic potential, subject to a uniform electric field at $t = 0$, the density rigidly moves in the manner of the underlying classical harmonic oscillator. We have shown that the fluidic approximation adds exactly the nonadiabatic correction required [71] by the HPT. We have also confirmed this numerically for two interacting electrons in a harmonic potential.

A constraint that can be challenging for nonadiabatic functionals is the *memory condition* [72], which notes that $v_{xc}(t)$ and hence $v_{KS}(t)$ must be independent of which previous instant in the evolution of the system is to be used to designate the "initial state." This is violated by the VK functional [34]. Equation (1) demonstrates that the fluidic approximation satisfies this memory condition by virtue of its dependence only on the instantaneous rate of change of u, and not its full history.

E. System 3

As a challenging test of the fluidic approximation, we finally consider two interacting electrons in a tunneling system. Initially, v_{ext} is a symmetric double-well potential, with one electron localized in each well. At $t = 0$, the left-hand well is raised and the right-hand well lowered, initiating tunneling through the barrier [Fig. 4]. A tunneling electron has an imaginary momentum, meaning that the (real) velocity field is of less physical significance. Correspondingly, the fluidic approximation recovers less of the adiabatic density error, but nevertheless reduces it from ≈8% to ≈4%, at $t = 15$ a.u. Accordingly, the tunneling rate from the left-hand side to the right-hand side is initially improved, but this is not the case at later times [inset of Fig. 4].

III. CONCLUSIONS

In summary, we have calculated the *exact* adiabatic and nonadiabatic parts of the KS potential, v_{KS}^{A} and Δv_{KS}, for a variety of model systems. Δv_{KS} is precisely defined by our procedure, and represents the part of the time-dependent KS potential that is *intrinsically unobtainable from a ground-state functional*. Our key finding is that a simple geometrical approximation to this nonadiabatic KS potential—making use of a Galilean transformation to the local instantaneous rest frame—recovers most of the density error attributable to the exact adiabatic approach: typically 80–95% in the ballistic systems studied. Studies of additional systems should further illuminate this decomposition of the KS potential of TDDFT in highly nonadiabatic situations, with the fluidic approximation providing a solid foundation for a hierarchy of approximations to Δv_{KS}.

Data created during this research is available from the York Research Database [73].

ACKNOWLEDGMENTS

We thank Jack Wetherell and Nick Woods for recent developments in the iDEA code, and Matt Hodgson and Carsten Ullrich for helpful comments.

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[66] The stated A causes the wave function in the original frame to become the wave function in the instantaneous rest frame multiplied by $\exp(-iu^2t/2)$.

[67] The integrated absolute error, $\int dx|n_1(x,t) - n_2(x,t)|$, expressed as a percentage of the total number of electrons.

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