



Exact Single-Electron Approach to the Dynamics of Molecules in Strong Laser Fields

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We present an exact single-electron picture that describes the correlated electron dynamics in strong laser fields. Our approach is based on the factorization of the electronic wave function as a product of a marginal and a conditional amplitude. The marginal amplitude, which depends only on one electronic coordinate and yields the exact one-electron density and current density, obeys a time-dependent Schrödinger equation with an effective time-dependent potential. The exact equations are used to derive an approximation that is a step towards general and feasible *ab initio* single-electron calculations for molecules. The derivation also sheds new light on the usual interpretation of the single-active electron approximation. From the study of model systems, we find that the exact and approximate single-electron potentials for processes with negligible two-electron ionization lead to qualitatively similar dynamics, but that the ionization barrier in the exact single-electron potential may be explicitly time dependent.

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Molecules in strong laser fields are a fascinating field of research. In such laser fields, it becomes possible to experimentally monitor the electron dynamics in chemical reactions on its natural time scale and in concert with the nuclear dynamics. Recent experimental studies show that parts of the electron dynamics can already be measured [1–4].

With progress in experimental techniques comes the necessity to develop and to improve theoretical tools to analyze the experiments. Single-electron pictures have an important role in this respect. The three-step model [5,6] and its quantum version, the strong field approximation [7], are single-electron models that describe the main mechanism which is responsible for many of the observed effects. Based on the success of these models, a single-active electron (SAE) approximation [8] is often the basis for quantum theories of strong field processes [9,10]. From such investigations, general phenomena that may occur in experiments can be deduced. However, there is no clear understanding of why the SAE works and what its limitations are, or even if it can be derived [11]. Thus, it is highly desirable to investigate how far we can get with a single-electron model.

Typically, in the SAE approximation, a time-dependent Schrödinger equation

$$i\partial_t\chi^{\text{SAE}}(r_1, t) = \left(-\frac{\partial^2}{2} + e^{\text{SAE}}(r_1) + r_1F(t) \right) \chi^{\text{SAE}}(r_1, t), \quad (1)$$

is solved for a single-electron wave function χ^{SAE} , and observables are computed from this wave function [10,11]. Many-electron effects are approximated by an effective time-independent potential $e^{\text{SAE}}(r_1)$, while the interaction with the laser field may, e.g., be described in the dipole

approximation in the length gauge, as is done in (1). The crucial information for the SAE is the effective potential e^{SAE} . While it may be possible to guess a model potential for atoms, this is much harder for molecules [9,12–14]. Nevertheless, hints that more general model potentials can describe effects that seem to be beyond the applicability of the SAE approximation have already existed for a while [15–17].

In this Letter, we present an exact single-electron description of a many-electron system in a laser field, the exact electron factorization (EEF). The EEF is then used to derive an approximation, the time-independent conditional amplitude (TICA) approximation, that is formally close to the SAE approximation (1) but has a different interpretation: The SAE approximation assumes that the processes to be described are essentially single-electron processes and seems to treat all but one electron as “frozen.” Hence, it is often assumed that it cannot describe multielectron effects [18]. In contrast, the EEF and, also, the TICA approximation represent the dynamics of all electrons with an effective potential.

The EEF is a generalization of the exact (electron-nuclear) factorization [19], which separates the nuclear from the electronic system of a molecular wave function exactly, to the case of electrons only. The idea of the EEF was already given for static systems some time ago [20,21], and aspects of it are also known in the field of density functional theory [22–25]. With this Letter, we generalize the EEF to time-dependent problems and show that it is useful for developing the theory of attosecond experiments. The derivation of a TICA approximation from the EEF shows the assumptions that are made when an equation such as (1) is used to represent the dynamics of all electrons. Also, it yields a general procedure for obtaining the TICA potential for any system.

To make the ideas that follow as clear as possible, we write the general equations using a simplified notation with only two spatial coordinates, r_1 and r_2 . In an n -electron system, r_1 and r_2 stand for an arbitrary partitioning of the coordinates of the electrons into two sets. The most important case for our application is the case where r_1 contains the coordinates of one electron and r_2 contains the coordinates of the remaining $n - 1$ electrons. This case is assumed below. Also, we use atomic units and we do not use explicit vector notation. The general equations are given in the Supplemental Material [26].

We consider a nonrelativistic description of a molecule in a laser field. In the dipole approximation in the length gauge, the evolution of the system is described with the time-dependent Schrödinger equation

$$i\partial_t\Psi = \left(-\frac{\partial_1^2}{2} - \frac{\partial_2^2}{2} + V(r_1, r_2) + (r_1 + r_2)F(t)\right)\Psi. \quad (2)$$

Here, $V(r_1, r_2)$ is the Coulomb interaction among the electrons and of the electrons with clamped nuclei, and $F(t)$ is the time-dependent electric field. We note that, by using the reverse factorization [27,28], (2) can be valid without clamping the nuclei. However, then, V would be time dependent and would not be a bare Coulomb potential. The electronic wave function $\Psi(r_1, r_2; s_1, s_2|t)$ depends on spatial coordinates r_j and spin coordinates s_j , which are, in general, not separable, but $\Psi(r_1, r_2; s_1, s_2|t)$ may always be written as a sum of coordinate permutations of a unique spatial wave function $\psi(r_1, r_2|t)$, multiplied by a corresponding spin function $\sigma(s_1, s_2)$ [29]. Below, we work with the spatial wave function $\psi(r_1, r_2|t)$ alone which, for our problem, has the same information content as $\Psi(r_1, r_2; s_1, s_2|t)$ and which has a time evolution given by the Schrödinger equation (2), too.

Next, we make the EEF ansatz

$$\psi(r_1, r_2|t) = \chi(r_1|t)\phi(r_2|r_1, t), \quad (3)$$

with partial normalization condition

$$\langle\phi(r_2|r_1, t)|\phi(r_2|r_1, t)\rangle_2 \stackrel{!}{=} 1 \quad \forall r_1, t, \quad (4)$$

where the notation $\langle\cdot\rangle_2$ represents integration over all coordinates r_2 . It automatically follows that

$$|\chi(r_1|t)|^2 = \langle\psi(r_1, r_2|t)|\psi(r_1, r_2|t)\rangle_2. \quad (5)$$

Given that $|\psi(r_1, r_2|t)|^2$ is normalized to one and has the meaning of a joint probability density (it represents the probability of finding one electron at r_1 and the other electrons at r_2 given we are at time t), χ and ϕ also acquire a special meaning: $|\chi(r_1|t)|^2$ is the one-electron density or marginal density (it represents the probability of finding an electron at r_1 , given time t) and $|\phi(r_2|r_1, t)|^2$ is the

conditional probability of finding $n - 1$ electrons at configuration r_2 , given one electron is at r_1 and given time t . Hence, we call χ the marginal amplitude and ϕ the conditional amplitude. We note that, if the number of spin-up and spin-down electrons is not equal, there are two different factorizations. An example is the three-electron system discussed below.

A variational derivation of the equations of motion for χ and ϕ yields

$$i\partial_t\chi = \left(\frac{[-i\partial_1 + A(r_1, t)]^2}{2} + \epsilon(r_1, t)\right)\chi, \quad (6)$$

for the marginal amplitude. This is a normal time-dependent Schrödinger equation with a vector potential

$$A(r_1, t) = \text{Im}\langle\phi|\partial_1\phi\rangle_2, \quad (7)$$

and a scalar potential

$$\epsilon(r_1, t) = \epsilon_T + \epsilon_V + \epsilon_F + \epsilon_{\text{FS}} + \epsilon_{\text{GD}}, \quad (8)$$

with average kinetic and potential energy of the other electrons

$$\epsilon_T(r_1, t) + \epsilon_V(r_1, t) = \langle\phi|-\frac{\partial_2^2}{2} + V|\phi\rangle_2, \quad (9)$$

the electric field interaction with a modified dipole operator

$$\epsilon_F(r_1, t) = F(t)(r_1 + \langle\phi|r_2|\phi\rangle_2), \quad (10)$$

a Fubini-Study (FS) term

$$\epsilon_{\text{FS}}(r_1, t) = \frac{1}{2}\langle\partial_1\phi|(1 - |\phi\rangle\langle\phi|)|\partial_1\phi\rangle_2, \quad (11)$$

and a gauge-dependent (GD) term

$$\epsilon_{\text{GD}}(r_1, t) = \text{Im}\langle\phi|\partial_t\phi\rangle_2. \quad (12)$$

There is a gauge freedom in the choice of a phase $S(r_1, t)$, because $\tilde{\chi} = e^{-iS(r_1, t)}\chi$ and $\tilde{\phi} = e^{iS(r_1, t)}\phi$ yield the same electronic wave function according to (3), and the equations of motion stay invariant up to the change $\tilde{A} = A + \partial_1 S$, $\tilde{\epsilon}_{\text{GD}} = \epsilon_{\text{GD}} + \partial_t S$. The equation of motion for the conditional amplitude ϕ is not of interest here and can be found in the Supplemental Material [26].

The marginal amplitude $\chi(r_1|t)$ is an interesting quantity. It yields the exact one-electron density, cf. (5), and it obeys a time-dependent Schrödinger equation (6). Additionally, it is straightforward to show that it also yields the exact one-electron current density. From χ , all observables depending on r_1 and ∂_1 , most notably the dipole expectation value (that yields, e.g., the high harmonic generation spectrum)

and the momentum expectation value, can be obtained. Hence, χ may also be called a one-electron wave function, and it gives an exact single-electron picture of the dynamics. However, the full problem does not, by itself, become easier by making ansatz (3). The main problem, now, is to obtain the scalar and vector potentials ϵ and A , which depend on the conditional amplitude ϕ . Nevertheless, the single-electron Schrödinger equation (6) gives us the possibility to derive a single-electron approximation.

One way to derive a single-electron approximation of the form (1) from the exact single-electron equation (6) is to assume a time-independent conditional amplitude, $\phi(r_2|r_1, t) \stackrel{!}{=} \phi_0(r_2|r_1)$. Then, we obtain a time-independent potential

$$\epsilon^{\text{TICA}}(r_1) = \epsilon_T[\phi_0] + \epsilon_V[\phi_0] + \epsilon_{\text{FS}}[\phi_0]. \quad (13)$$

Additionally, we choose the gauge $A(r_1, t) = 0$, although we want to stress that similar to the exact electron-nuclear factorization, cases are conceivable where this choice is not possible and where a Berry phase may occur [30]. With this choice of gauge, the only formal difference between the TICA approximation and the SAE equation (1) is the change of the dipole operator from r_1 to $d(r_1) = r_1 + \langle \phi_0 | r_2 | \phi_0 \rangle_2$; i.e., the equation of motion of the TICA approximation is

$$i\partial_t \chi^{\text{TICA}} = \left(-\frac{\partial_1^2}{2} + \epsilon^{\text{TICA}}(r_1) + d(r_1)F(t) \right) \chi^{\text{TICA}}. \quad (14)$$

The formal similarity between (1) and (14) should not be misunderstood: The TICA approximation does not assume that there is one “active” electron and a frozen core but approximates the exact single-electron density and current density and, hence, the correlated dynamics of all electrons. In contrast, the model potential and initial wave function for a SAE calculation are typically chosen according to the idea of treating only one electron, and observables are calculated with both the active and the frozen part of the electronic wave function.

A typical choice for ϕ_0 is the conditional amplitude of the state at $t = 0$, which usually is an eigenstate of the system. Then, to compute ϵ^{TICA} in practice, it is only necessary to know the one-electron density $\rho(r_1)$ of this state, as $\rho(r_1) = |\chi^{\text{TICA}}|^2$ at $t = 0$, and to solve the time-independent analogue of (14) for ϵ^{TICA} . The modified dipole $d(r_1)$ can be obtained from the initial electronic wave function. While, in our examples, $d(r_1)$ is of minor importance, this may change when more electrons are considered. To obtain the dynamics, it is only necessary to solve the single-electron time-dependent Schrödinger equation (14), independent of the number of electrons in the system. A discussion of a possible practical implementation and its associated numerical cost is given in the Supplemental Material [26].

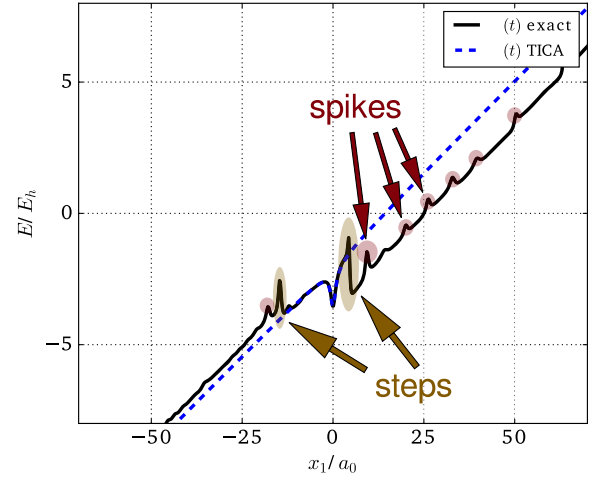


FIG. 1. Exact single-electron potential (8) (solid black line) and TICA potential (13) with laser potential (dashed blue line) at a time where the amplitude of the laser field is maximal, for the one-dimensional two-electron model.

To learn more about the exact and TICA potentials, we consider one-dimensional models of the helium atom (two electrons) and of the lithium atom (three electrons). All solutions to the involved eigenvalue problems were obtained with help of the linear algebra routines in the SCIPY package [31,32]. The time propagation was performed with the Gonoskov-Marklund propagator [33] or a Runge-Kutta method. Details can be found in the Supplemental Material [26].

For the Helium model, we choose, as the initial state, the spin-singlet ground state and use the parameters of [34]. Our choice for ϕ_0 is the conditional amplitude of the initial state. The dynamics is computed for a 12-cycle laser pulse with a wave length of 580 nm and a maximum intensity of 6.9×10^{14} W/cm². By comparing the dynamics and the high harmonic generation spectra [35], we found very good agreement of the TICA calculations with the exact calculations, and the effect of the modified dipole operator is almost negligible. A representative picture for the comparison of the exact single-electron potential $\epsilon(r_1, t)$ with the TICA potential is shown in Fig. 1, and a movie, as well as the spectra, are given in the Supplemental Material [26].

We see two important features that distinguish the exact potential from the TICA potential: There are time-dependent steps and spikes in the exact potential which are absent in the TICA potential. The steps are similar to those known from time-dependent density functional theory [16,36–38] and are related to those from the exact electron nuclear factorization [39]. They occur only at certain times and positions, develop and disappear rapidly, and can only be found in the gauge-dependent part ϵ_{GD} of the potential. We do not have an intuitive interpretation for the steps in the EEF, but we find that they have negligible effect on the dynamics: They are located at positions with very low electron density and the parts of the potential connected by

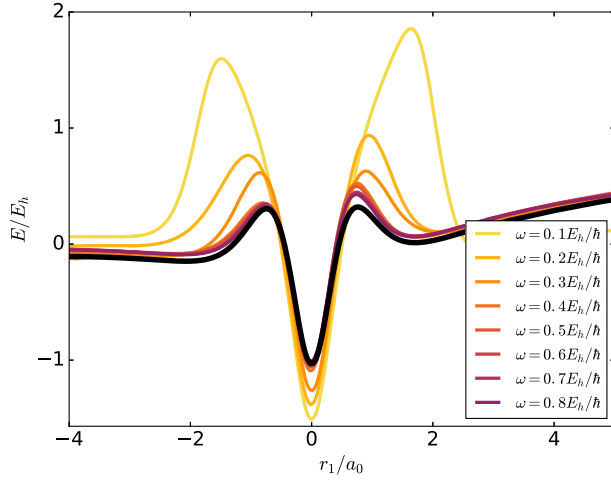


FIG. 2. Exact single-electron potential (8) (colored lines) for different values of the laser frequency, and TICA potential (13) with laser potential (black line) after half of the pulse, where the amplitude of the laser field is maximal, for the one-dimensional three-electron model.

steps are parallel. Hence, they change the momentum of some parts of χ , but this does not result in an important effect for the overall dynamics. The spikes also occur at positions where the one-electron density is minimal. They are a peculiar feature of the factorization ansatz: because $|\chi|^2$ is the one-electron density and this never becomes zero in our systems, we do not have exact nodes unless we impose them by a choice of the phase (which may lead to discontinuities in χ). We found that, in the model propagation, the spikes in the exact potential can be neglected for the propagation of χ , which is equivalent to allowing the marginal amplitude to become zero.

Next, we study a lithium model, a one-dimensional spin-doublet model system with parameters taken from [40], for an eight-cycle laser pulse with several laser frequencies between $0.1 E_h \hbar$ (152 nm) and $1.0 E_h \hbar$ (46 nm), and with a maximum intensity of $8.8 \times 10^{13} \text{ W/cm}^2$. The electronic wave function is given by

$$\Psi = N[\psi(r_1, r_2, r_3)|\uparrow\uparrow\downarrow\rangle + \psi(r_2, r_3, r_1)|\uparrow\downarrow\uparrow\rangle + \psi(r_3, r_1, r_2)|\downarrow\uparrow\uparrow\rangle], \quad (15)$$

with the antisymmetry condition $\psi(r_1, r_2, r_3) = -\psi(r_2, r_1, r_3)$ for the spatial wave function. There are two possible factorizations, one for the spin-up one-electron density $|\chi_\uparrow(r_1|t)|^2 = \langle \psi(r_1, r_2, r_3) | \psi(r_1, r_2, r_3) \rangle_{23}$ and one for the spin-down one-electron density $|\chi_\downarrow(r_3|t)|^2 = \langle \psi(r_1, r_2, r_3) | \psi(r_1, r_2, r_3) \rangle_{12}$. As we aim at describing processes that mainly involve the valence electron, which is a spin-up electron, we only consider the factorization for χ_\uparrow .

Figure 2 shows a snapshot of the exact and TICA potentials for the three-electron model system in the laser

fields, at a time where the electric field is maximal, in the spatial region close to the nucleus. Movies of the dynamics for different frequencies of the laser field are given in the Supplemental Material [26]. Note that the TICA potential is independent of the laser frequency, as it is a sum of the initial exact potential and the laser interaction in dipole form, cf. (14).

The initial potential shows a deep minimum, but also a barrier to the left and to the right followed by shallow minima. This reflects the electronic structure of the problem. It also illustrates that, in contrast to the example of the helium model, a suitable model potential for a SAE calculation of a many-electron system is in general not easy to guess and an *ab initio* treatment like the TICA approximation is recommended.

During the evolution, time-dependent spikes and steps occur in the exact potential as in the two-electron case. What is most striking, however, is the explicit time dependence of the exact potential around the bounding region: At times where the field is strong, the potential well, located around $r_1 = 0$, becomes deeper, and the barriers at its sides become higher. These changes become stronger with a smaller frequency of the laser pulse. Interpreted in terms of the three-step model, the effect of this additional barrier is clear: It suppresses the tunneling out of the bounding well and is an obstacle during the recombination step. In contrast, the TICA potential does not show this time-dependent barrier, which results in a similar qualitative dynamics, but different quantitative results.

Further investigations of the contributions to the exact potential show that this effect originates in the interaction of the two spin-up electrons, while the effect of the contributions of the spin-down electron to the exact potential is small. We expect the time-dependent barrier to be partly a consequence of the antisymmetry condition of the electronic wave function for the two spin-up electrons, which is not met by the TICA: While the EEF is exact and preserves symmetries of the full wave function, it does not reflect those symmetries in the equations of motion. Consequently, approximations to the EEF lead to a symmetry breaking, which may be of minor importance, as in the two-electron spin singlet case, but which we expect to be significant in most cases, as in the three-electron spin doublet case. Hence, it may be worthwhile to include symmetries of the problems explicitly. Extensions like a repeated factorization [41] or more symmetric factorizations need to be explored in the future.

We also compared the TICA dynamics with a SAE calculation based on the exact Kohn-Sham orbitals and Kohn-Sham potential (obtained as explained in [37]). For the model system, the SAE calculations are in relatively good agreement with the exact dynamics and generally give slightly better results than the TICA calculation for small laser frequencies, but worse results for higher frequencies. Details can be found in the Supplemental Material [26].

However, our choice for a SAE calculation is idealized (using the exact Kohn-Sham potential) and modeling more complex systems like molecules with a SAE approach need further assumptions and approximations. Hence, the significance of this comparison is so far limited.

In summary, from our investigations, we learned the following: First, there is an exact single-electron theory, the EEF, and it can be used to obtain a single-electron approximation, the TICA approximation. The TICA approximation is formally close to the typical SAE approach, but it has a well-defined effective potential that contains information of the electronic structure of the system. A TICA calculation is feasible for relatively large molecules, and it may be useful in situations where the alignment of the molecule relative to the laser field matters. Second, we found that a two-electron spin-singlet system behaves similar to a one-electron system and that the shape of the exact potential is simple to model, at least as long as two-electron ionization is negligible. Hence, it is easy to approximate, but its applicability as a model for many-electron effects is limited. Third, we saw that, already for the three-electron model system, an *ab initio* treatment like the TICA approximation is necessary to obtain a potential which includes all relevant features. Finally, it became clear that the TICA approach is useful to compute qualitative effects, again with the restriction of negligible two-electron ionization. It does, however neglect a time-dependent barrier that changes the ionization and recombination step. This barrier, in turn, challenges our view on the three-step model and shows the need for further studies.

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