Time-Dependent Multicomponent Density Functional Theory for Coupled Electron-Positron Dynamics

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Electron-positron interactions have been utilized in various fields of science. Here we develop time-dependent multicomponent density functional theory to study the coupled electron-positron dynamics from first principles. We prove that there are coupled time-dependent single-particle equations that can provide the electron and positron density dynamics, and derive the formally exact expression for their effective potentials. Introducing the adiabatic local density approximation to time-dependent electron-positron correlation, we apply the theory to the dynamics of a positronic lithium hydride molecule under a laser field. We demonstrate the significance of the coupling between electronic and positronic motion by revealing the complex positron detachment mechanism and the suppression of electronic resonant excitation by the screening effect of the positron.

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Positron physics has been extensively studied [1] since the positron was discovered in 1932 [2]. When a positron beam with an energy of up to a few keV is directed to a material, the incident positron diffuses inside the bulk and is finally annihilated with electrons and γ rays are emitted [3]. An analysis of these γ rays provides various information related to the surface structures, lattice defects, and electronic structures of the material [3–7]. The positron-material interaction plays a key role in these experiments [8]. The interaction between a positron and atom or molecule has also been widely studied to better understand how positrons interact with electrons and are bound to them [9–11]. An experiment that measured positron-atom binding energies through the study of positron-atom recombination under a laser field was recently reported [12].

Theoretical approaches to study these positron physics have been extensively developed [13–17]. Among them, two-component density functional theory (2C-DFT) [4,18–20], which is an extension of DFT [21] to the coupled electron-positron system, has been a powerful first-principles tool to calculate the ground-state electron and positron densities and their properties. 2C-DFT has been successfully applied to studies of positron interaction with atoms, solids, and surfaces to determine the electron-positron momentum distributions [22], positron annihilation lifetimes [23], and positron binding energies [24], to name a few. Another powerful method is the wave-function-based approach, such as the multicomponent molecular orbital method [25] and quantum Monte Carlo method [26,27], which have also revealed much of positron physics with high accuracy. However, the dynamic interaction mechanism between positrons and electrons, especially under a laser field [10–12], has not yet been clarified, because there has been no first-principles method that can simulate the coupled dynamics of positrons and electrons for realistic systems.

In this study, we develop *time-dependent multicomponent density functional theory* (TD-MCDFT) [28,29] to study the coupled electron-positron dynamics. Time-dependent density functional theory (TDDFT) [30–32] has enabled us to simulate real-time, many-electron dynamics, by mapping it to the dynamics of the noninteracting [also called Kohn-Sham (KS)] system evolving in a single-particle potential by virtue of Runge-Gross [30] and van Leeuwen [33] theorems. TD-MCDFT is an extension of TDDFT to multicomponent (MC) systems, and it has been expected to provide a first-principles simulation tool for the dynamics of a system that consists of different types of quantum particles. Li and Tong proved that one-to-one mapping between external potentials and time-dependent (TD) densities is established also in MC systems [28]. Gross *et al.* then applied the Li-Tong theorem to the coupled electron-nuclear systems and developed time-dependent multicomponent Kohn-Sham (TD-MCKS) equations for a one-dimensional model of the H_2^+ molecule [29]. However, there are some difficulties when applying their method to real (three-dimensional) systems. To define electron and nuclear densities that are not constant in space, a body-fixed frame transformation [34], which makes electron density reflect the internal symmetry of the system, must be conducted. Only a Hartree approximation has been tested for a TD electron-nuclear correlation potential, and it did not give satisfactory results [29]. It has been a challenging issue to develop electron-nuclear correlation potential functionals, and there have been ongoing studies on it [35,36].

Here, our purpose is to develop TD-MCDFT for electron-*positron* dynamics. For this, we can circumvent the problem of the body-fixed frame transformation by treating nuclei as classical particles, because classical nuclei serve as external

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potentials for electrons and positrons, and the Hamiltonian of the system is no longer translationally and rotationally invariant [37]. We define TD density and current density both for electrons and positrons, and prove noninteracting v representability [33] for these quantities. The formally exact expression for the TD electron-positron correlation functional is then derived by extending the TDDFT action principle [38] to the MC system. Furthermore, we introduce the adiabatic local density approximation (ALDA) to the TD electron-positron correlation. Finally, the TD-MCDFT method is applied to the dynamics of a positronic lithium hydride (e^+-LiH) molecule under a laser field and elucidate the significant role of the coupling between electronic and positronic motion.

We begin by considering the full Hamiltonian of a system that consists of N^- electrons and N^+ positrons

$$\hat{H} = \hat{T}(\underline{\underline{\mathbf{r}}}^-, \underline{\underline{\mathbf{r}}}^+) + \hat{W}(\underline{\underline{\mathbf{r}}}^-, \underline{\underline{\mathbf{r}}}^+) + \hat{V}(\underline{\underline{\mathbf{r}}}^-, \underline{\underline{\mathbf{r}}}^+, t), \tag{1}$$

where the kinetic energy operator is $\hat{T} = -\sum_{i=1}^{N^-} \frac{1}{2}\nabla^2_{\mathbf{r}_i^-} - \sum_{\alpha=1}^{N^+} \frac{1}{2}\nabla^2_{\mathbf{r}_\alpha^+}$, the interaction operator is $\hat{W} = \sum_{i<j}^{N^-}(1/|\mathbf{r}_i^- - \mathbf{r}_j^-|) + \sum_{\alpha<\beta}^{N^+}(1/|\mathbf{r}_\alpha^+ - \mathbf{r}_\beta^+|) - \sum_{i=1}^{N^-}\sum_{\alpha=1}^{N^+} \times (1/|\mathbf{r}_i^- - \mathbf{r}_\alpha^+|)$, the potential operator due to the interaction with classical nuclei and TD external field is $\hat{V} = \sum_{i=1}^{N^-} v_{\mathrm{ext}}^-(\mathbf{r}_i^-, t) + \sum_{\alpha=1}^{N^+} v_{\mathrm{ext}}^+(\mathbf{r}_\alpha^+, t)$, and $\underline{\underline{\mathbf{r}}}^-$ and $\underline{\underline{\mathbf{r}}}^+$ are the sets of electronic and positronic laboratory coordinates, respectively. Throughout this Letter, the sign $-$ $(+)$ indicates an electron (positron), and atomic units are used unless stated otherwise. The TD electron-positron wave function $\Psi(\underline{\underline{\mathbf{r}}}^-, \underline{\underline{\mathbf{r}}}^+, t)$ obeys the full TD Shrödinger equation $i\partial_t\Psi = \hat{H}\Psi$. By treating nuclei as classical charges that determine the laboratory coordinate system, the TD electron (positron) density $n^{-(+)}$ and current density $\mathbf{j}^{-(+)}$ can be defined as follows:

$$n^\mp(\mathbf{r}^\mp, t) = N^\mp \int d^{N^\pm}\mathbf{r}^\pm \int d^{N^\mp - 1}\mathbf{r}^\mp |\Psi|^2, \tag{2}$$

$$\mathbf{j}^\mp(\mathbf{r}^\mp, t) = \mathrm{Re}\left(N^\mp \int d^{N^\pm}\mathbf{r}^\pm \int d^{N^\mp - 1}\mathbf{r}^\mp \Psi^*(-i\nabla_{\mathbf{r}^\mp}\Psi)\right). \tag{3}$$

The equations of motions for these quantities are as follows:

$$\frac{\partial}{\partial t} n^\mp = -\nabla_{\mathbf{r}^\mp} \cdot \mathbf{j}^\mp, \tag{4}$$

$$\frac{\partial}{\partial t}\mathbf{j}^\mp = -n^\mp(\nabla_{\mathbf{r}^\mp} v_{\mathrm{ext}}^\mp) - i\langle\Psi|[\hat{\mathbf{j}}^\mp, \hat{T} + \hat{W}]|\Psi\rangle, \tag{5}$$

where $\hat{\mathbf{j}}^\mp = \frac{1}{2i}\sum_{l=1}^{N^\mp}(\nabla_{\mathbf{r}_l^\mp}\delta(\mathbf{r}^\mp - \mathbf{r}_l^\mp) + \delta(\mathbf{r}^\mp - \mathbf{r}_l^\mp)\nabla_{\mathbf{r}_l^\mp})$.

Now we prove that there are effective potentials $v_{\mathrm{KS}}^\mp$ in the noninteracting KS system [21,33] that reproduce $n^-(\mathbf{r}^-, t)$ and $n^+(\mathbf{r}^+, t)$ in the interacting system, i.e., TDMC noninteracting v representability, by extending the van Leeuwen theorem [33] to the MC system. Taking the divergence of Eq. (5) and using the continuity Eq. (4) gives:

$$\frac{\partial^2}{\partial t^2} n^\mp = \nabla\cdot[n^\mp(\nabla_{\mathbf{r}^\mp} v^\mp)] + i\nabla\cdot\langle\Psi|[\hat{\mathbf{j}}^\mp, \hat{T} + \hat{W}]|\Psi\rangle, \tag{6}$$

which is valid for both the interacting system and the KS system. Now we impose the condition that the potentials in both systems are Taylor expandable around the initial time t_0, i.e., $v_{\mathrm{ext/KS}}^\mp(\mathbf{r}^\mp, t) = \sum_{k=0}^\infty (1/k!) v_{\mathrm{ext/KS},k}^\mp(\mathbf{r}^\mp, t)(t - t_0)^k$. Furthermore, we impose the initial conditions that the initial state in the interacting system, Ψ_0, and in KS system, Φ_0, yield the same densities and their first time-derivatives, i.e.,

$$n^\mp(\mathbf{r}^\mp, t_0) = n_{\mathrm{KS}}^\mp(\mathbf{r}^\mp, t_0), \tag{7}$$

$$\left.\frac{\partial}{\partial t} n^\mp(\mathbf{r}^\mp, t)\right|_{t=t_0} = \left.\frac{\partial}{\partial t} n_{\mathrm{KS}}^\mp(\mathbf{r}^\mp, t)\right|_{t=t_0}. \tag{8}$$

These initial conditions uniquely determine the solutions of the second-order differential Eq. (6). If $n^\mp(\mathbf{r}^\mp, t) = n_{\mathrm{KS}}^\mp(\mathbf{r}^\mp, t)$ at all times, then subtracting Eq. (6) for the interacting system and the KS system gives the following:

$$\begin{aligned} &\nabla\cdot\{n^\mp[\nabla_{\mathbf{r}^\mp}(v_{\mathrm{ext}}^\mp - v_{\mathrm{KS}}^\mp)]\} \\ &\quad = i\nabla\cdot\langle\Phi|[\hat{\mathbf{j}}^\mp, \hat{T}]|\Phi\rangle - i\nabla\cdot\langle\Psi|[\hat{\mathbf{j}}^\mp, \hat{T} + \hat{W}]|\Psi\rangle, \end{aligned} \tag{9}$$

where Φ is the full wave function of the KS system. Equation (9) has unique solutions for $v_{\mathrm{KS}}^\mp(\mathbf{r}^\mp, t)$ when $n^\mp(\mathbf{r}^\mp, t)$, $\Psi_0(\underline{\underline{\mathbf{r}}}^-, \underline{\underline{\mathbf{r}}}^+)$, $\Phi_0(\underline{\underline{\mathbf{r}}}^-, \underline{\underline{\mathbf{r}}}^+)$, and $v_{\mathrm{ext}}^\mp(\mathbf{r}^\mp, t)$ are given, similar to the procedure described in Ref. [33].

By virtue of the TDMC noninteracting v representability proved above, the following coupled TD-MCKS equations that produce $n^\mp(\mathbf{r}^\mp, t)$ exist as follows:

$$i\frac{\partial}{\partial t}\psi_i^\mp(\mathbf{r}^\mp, t) = \left(-\frac{\nabla^2_{\mathbf{r}^\mp}}{2} + v_{\mathrm{KS}}^\mp(\mathbf{r}^\mp, t)\right)\psi_i^\mp(\mathbf{r}^\mp, t), \tag{10}$$

where $\sum_{l=1}^{N^\mp}|\psi_l^\mp(\mathbf{r}^\mp, t)|^2 = n^\mp(\mathbf{r}^\mp, t)$.

Next, we derive the expressions of $v_{\mathrm{KS}}^\mp(\mathbf{r}^\mp, t)$ from an action principle. Action principles in TD-MCDFT can be formulated using the Keldysh time-contour technique [29,39]; however, here we instead extend that in TDDFT formulated by Vignale [38] to the electron-positron system. According to the Li-Tong theorem [28], the full wave function is functional of the TD densities, and thus the quantum mechanical action is also a density functional, as shown here:

$$\begin{aligned}A[n^-, n^+] &= \int_{t_0}^{t_1} dt \langle \Psi[n^-, n^+] | i\partial_t - \hat{H} | \Psi[n^-, n^+] \rangle \\ &= A_0[n^-, n^+] - \int_{t_0}^{t_1} dt \int d\mathbf{r}^- n^-(\mathbf{r}^-, t) v_{\mathrm{ext}}^-(\mathbf{r}^-, t) \\ &\quad - \int_{t_0}^{t_1} dt \int d\mathbf{r}^+ n^+(\mathbf{r}^+, t) v_{\mathrm{ext}}^+(\mathbf{r}^+, t), \qquad (11)\end{aligned}$$

where we define $A_0[n^-, n^+] = \int_{t_0}^{t_1} dt \langle \Psi | i\partial_t - \hat{T} - \hat{W} | \Psi \rangle$. As in Ref. [38], we consider the variations of the densities such that $\delta\Psi[n^-, n^+](t_0) = 0$, and the variational principle $\delta A[n^-,n^+] = i\langle \Psi(t_1) | \delta\Psi(t_1) \rangle$ [31,38] leads to the following:

$$\frac{\delta A_0[n^-, n^+]}{\delta n^\mp} - v_{\mathrm{ext}}^\mp(\mathbf{r}^\mp, t) = i \left\langle \Psi(t_1) \middle| \frac{\delta \Psi(t_1)}{\delta n^\mp} \right\rangle. \qquad (12)$$

Similarly, the variational principle for KS systems leads to

$$\frac{\delta A_0^{\mathrm{KS}}[n^-, n^+]}{\delta n^\mp} - v_{\mathrm{KS}}^\mp(\mathbf{r}^\mp, t) = i \left\langle \Phi(t_1) \middle| \frac{\delta \Phi(t_1)}{\delta n^\mp} \right\rangle, \qquad (13)$$

where $A_0^{\mathrm{KS}}[n^-, n^+] = \int_{t_0}^{t_1} dt \langle \Phi | i\partial_t - \hat{T} | \Phi \rangle$. Now we define the exchange-correlation (xc) action functional for the electron-positron system as follows:

$$A_{\mathrm{xc}}^{-+}[n^-,n^+] = A_0^{\mathrm{KS}}[n^-,n^+] - A_0[n^-,n^+] - A_{\mathrm{H}}[n^-,n^+], \qquad (14)$$

where $A_{\mathrm{H}} = \int dt d\mathbf{r}_1^- d\mathbf{r}_2^- \{[n^-(\mathbf{r}_1^-,t) n^-(\mathbf{r}_2^-,t)]/[2|\mathbf{r}_1^- - \mathbf{r}_2^-||]\} + \int dt d\mathbf{r}^- d\mathbf{r}^+ \{[n^-(\mathbf{r}^-,t) n^+(\mathbf{r}^+,t)]/[|\mathbf{r}^- - \mathbf{r}^+||]\} + \int dt d\mathbf{r}_1^+ d\mathbf{r}_2^+ \times \{[n^+(\mathbf{r}_1^+,t) n^+(\mathbf{r}_2^+,t)]/[2|\mathbf{r}_1^+ - \mathbf{r}_2^+||]\}$ is the Hartree action functional. With these definitions of A_{xc}^{-+}, and Eqs. (12) and (13), we find the expression of the TD-MCKS potentials

$$v_{\mathrm{KS}}^\mp(\mathbf{r}^\mp, t) = v_{\mathrm{ext}}^\mp(\mathbf{r}^\mp, t) + v_{\mathrm{H}}^\mp(\mathbf{r}^\mp, t) + v_{\mathrm{xc}}^\mp(\mathbf{r}^\mp, t), \qquad (15)$$

where $v_{\mathrm{H}}^\mp = (\delta A_{\mathrm{H}} / \delta n^\mp)$ are the Hartree potentials and

$$v_{\mathrm{xc}}^\mp = \frac{\delta A_{\mathrm{xc}}^{-+}}{\delta n^\mp} + i \left\langle \Psi(t_1) \middle| \frac{\delta \Psi(t_1)}{\delta n^\mp} \right\rangle - i \left\langle \Phi(t_1) \middle| \frac{\delta \Phi(t_1)}{\delta n^\mp} \right\rangle \qquad (16)$$

are the exchange-correlation potentials, which incorporate all many-body effects.

Now the success of the theory is dependent on how to approximate $v_{\mathrm{xc}}^\mp$ so that they can be calculated practically. Here we introduce the adiabatic approximation, i.e., neglecting the boundary terms and approximating A_{xc}^{-+} as this:

$$A_{\mathrm{xc}}^{-+,\mathrm{A}} = \int_{t_0}^{t_1} dt (E_{\mathrm{xc}}^-[n_0^-] + E_{\mathrm{c}}^{-+}[n_0^-, n_0^+])|_{n_0^\mp \to n^\mp(t)}, \qquad (17)$$

where E_{xc}^- is the electron-electron xc energy functional in DFT, and E_{c}^{-+} is the electron-positron correlation energy functional in 2C-DFT. Note that, in many cases, a system that consists of one positron and many electrons is of interest; thus in Eq. (17) and hereafter we omit the positron-positron xc term in A_{xc}^{-+} and the positron-positron Hartree term in A_{H}, which ensures a rigorous self-interaction correction for a single positron [19]. The adiabatic approximation to the xc term has been successfully used in many TDDFT studies [31,40], while its limitations, such as the lack of the memory effect [32,41], have also been extensively discussed [32,41]. Here, as the first application of TD-MCDFT to a realistic molecular system, we use the adiabatic approximation. Specifically, the LDA [42] is used for E_{xc}^-. For E_{c}^{-+}, we also use the LDA revised by Zubiaga *et al.*, which was reported to be suitable for the ground state of positronic atoms [24]. We should note that the LDA lacks the nonlocal correlation effect that leads to the correct asymptotic behavior in finite systems. The weighted-density approximation [43] to correlation would be one of the approach to improve the functional.

We now show the application of TD-MCDFT presented here to the dynamics of a e^+-LiH molecule under a laser field. The LiH molecule has been the target of many previous theoretical studies on positron-molecule interactions [10,27,44,45], and recent experimental studies [10–12] focused on the response of a positron-molecule compound to a laser field.

One LiH molecule is placed in the center of a $30 \times 30 \times 30$ Å^3 cubic unit cell so that the molecular axis is along the x axis. The bond length of LiH is set to the experimental value of 1.60 Å [46]. One positron is then added to the LiH molecule, and the ground-state electron and positron densities are determined by the self-consistent 2C-DFT calculation [4,18–20] using LDA both for E_{xc}^- [42] and E_{c}^{-+} [24] and the plane wave basis set with norm conserving pseudopotentials [47] (cut off energy of 816 eV). As we treat the charge system, a homogeneous compensating background charge is introduced.

The inset of Fig. 1(a) shows plots of the isosurfaces of the ground-state electron (blue, 0.89 e^-/Å^3) and positron (red, 0.22 e^+/Å^3) densities, where Li and H nuclei are plotted as green and gray spheres, respectively. The electron density is localized around the H atom, and the positron density is loosely bound to the electron density. The densities integrated over the yz plane, $n^\mp(x) = \int dy \int dz n^\mp(\mathbf{r})$, are plotted in Fig. 1(a) as black (n^-) and red (n^+) dotted lines. The Li (H) nucleus is located at $x = -0.8$ (0.8) Å. These distributions of the ground-state densities are in good agreement with those calculated by the wave-function-based approach [27,44].

Now we propagate these densities under the laser fields by solving the TD-MCKS equations (10). The laser field is applied along the x axis, described within the dipole approximation and length gauge as $v_{\mathrm{laser}}^\mp(\mathbf{r}^\mp, t) = \mp E(t) x$ and $E(t) = E_0 \sin(\omega t) \exp[(t - t_0)^2/\sigma^2]$, where $E_0 = 0.2$ V/Å, $\sigma = 2$ fs, $t_0 = 4$ fs, and we compare the results from three different energies of $\omega = 0.5$, 1.5, and 3.0 eV [$E(t)$ are shown in Fig. 2(b)]. Note that the lasers we used are the pulsed femto second lasers and rather shorter

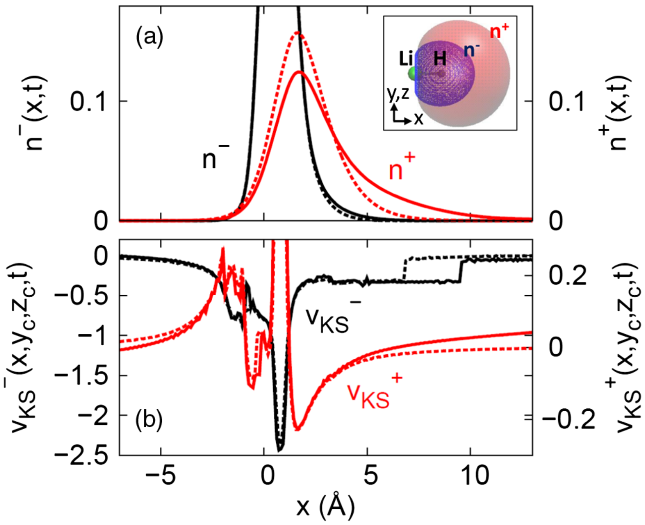

FIG. 1. (a) Snapshots of $n^{-}(x,t)$ (black) and $n^{+}(x,t)$ (red) at $t=0$ (dotted) and $t=4.36$ fs (solid) in the dynamics of e^{+}-LiH under a laser field ($\omega=1.5$ eV), and (b) corresponding $v_{\mathrm{KS}}^{\mp}(x,y_c,z_c,t)$. The inset shows the isosurfaces of the ground-state densities (see text).

than those used in the recent experiments [10–12], while the peak intensity ($I=|E_0|^2=0.5\times10^{12}$ W/cm^2) is larger than those in the experiments. In Fig. 1(a), the snapshot of $n^{-}(x,t)$ [$n^{+}(x,t)$] at $t=4.36$ fs for $\omega=1.5$ eV is plotted as a black (red) solid line [48]. Figure 1(b) shows the corresponding $v_{\mathrm{KS}}^{\mp}(x,y_c,z_c,t)$ [48] [y_c (z_c) is the midpoint of y (z) side of the unit cell]. The discontinuity appeared in v_{KS}^{-} around $x=5$–10 Å corresponds to the cutoff radius introduced by Zubiaga *et al.* [24], and is considered to negligibly affect the electron dynamics as it appears only in the region where electron density is negligibly small. It is evident from this figure and the movies in the Supplemental Material [48] that considerable positron density moves toward the positive x direction at $t=4.36$ fs while oscillating due to the laser field. This indicates the increase of the probability to find the positron that leaves the LiH molecule.

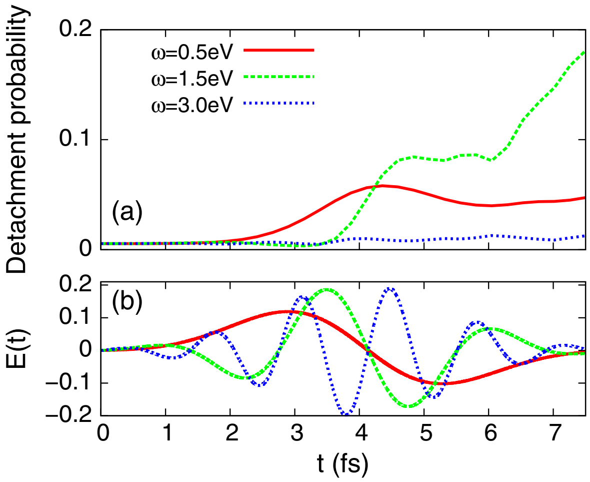

FIG. 2. (a) Time evolution of positron detachment probability for three different laser fields, and (b) corresponding $E(t)$.

This laser energy of $\omega=1.5$ eV gives the largest positron detachment probability, which is calculated by the integration of $n^{+}(x,t)$ over the region outside the bound region (-3.4 Å $< x <$ 7.2 Å) [49], among the three energies as shown in Fig. 2(a). The 3.0 eV laser field leads to the lowest (very few) detachment probability. We note that the 0.81 eV laser energy, which corresponds to the approximate positron binding energy (calculated on the basis of Janak's theorem [50]), leads to the lower detachment probability (not shown here) than the one obtained with 1.5 eV laser. Furthermore, the positron dynamics are not synchronized to $E(t)$ [Fig. 2(b)], but there is a retardation or an even more complex response to the laser field [48]. Electrons and positrons respond to the laser field in the opposite way while attracting each other. This is the cause of the complex dynamics that appeared in Fig. 2. Only the TD-MCDFT calculation can reveal the mechanism as to how the coupled positron and electron respond to the laser field by providing the dynamics of $v_{\mathrm{KS}}^{\mp}$. The movies [48] show that, in the case of $\omega=1.5$ eV, v_{KS}^{+} bends effectively for positron detachment at around $t=4$ fs, while, in the case of $\omega=3.0$ eV, v_{KS}^{+} changes the direction of its gradient before the positron starts to depart. Note that here we propagate the system up to 10 fs while fixing classical nuclear position. However at this timescale the quantum nuclear motion may play a role and its study is an important direction for future work [37].

We present another intriguing result that shows the importance of the coupling between electronic and positronic motion in Fig. 3. There we compare the TD dipole moment of LiH, $d_x^{-}(t)=-\int d\mathbf{r}^{-}xn^{-}(\mathbf{r}^{-},t)$, between the system of e^{+}-LiH (red) and LiH (black) under the laser field with $\omega=3.0$ eV. The linear-response TDDFT

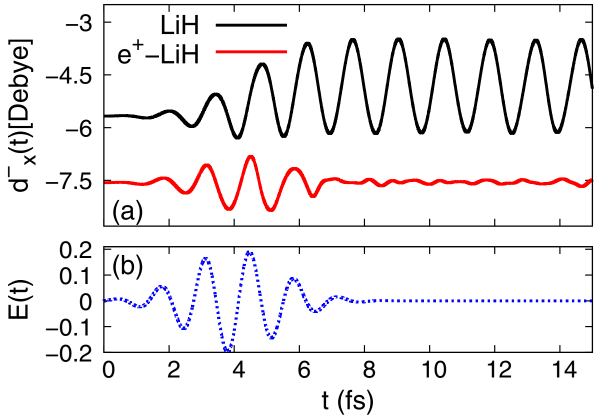

FIG. 3. (a) Time-dependent dipole moment of LiH $d_x^{-}(t)$ of e^{+}-LiH (red) and LiH (black) under the laser field ($\omega=3.0$ eV), and (b) corresponding $E(t)$.

calculation [51] for LiH (not shown here) shows that the 3.0 eV energy laser elevates the electrons to the first excited state. It is evident in Fig. 3 that a charge density oscillation, which arises from the resonant excitation and remains after the laser field fades [52], is produced in LiH (black). The center of the dipole oscillation shifts to around −4.85 Debye from its ground-state value of −5.67 Debye, which indicates the occurrence of charge transfer excitation [53]. Now turning to the e^+-LiH case (red), we see that such a charge oscillation is not induced. Thus, the positron suppresses the electronic excitation, as the result of the coupling between electronic and positronic motion. A positron attracts electrons and responds to the laser field in the opposite way to electrons. As a result, electrons are forced to move toward the direction opposite to the field by the positron. In other words, the response of the electrons to the laser field is screened by the positron. The movies of the dynamics [48] clearly show that the electron density in e^+-LiH moves toward the opposite direction compared to that in LiH.

In conclusion, we have developed TD-MCDFT for coupled electron-positron systems, which is a first-principles method that treats both the electron and positron dynamics quantum mechanically. TD-MCDFT was applied to e^+-LiH under a laser field. There was no simple relationship between the laser energy and the positron detachment probability, which indicates a complex coupling between electronic and positronic motion. Furthermore, it was elucidated that the attached positron significantly suppresses laser-induced electronic excitations, which suggests the possibility that the absorption spectrum and excited-state nuclear dynamics may also be changed. There are many other promising applications of the theory, such as application to the positron migration in the bulk of the material to reveal how it is trapped in a defect or surface, and positron scattering by materials. These are key events in positron beam experiments, and the TD-MCDFT methodology developed here will be valuable for the study of fundamental positron physics.

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