Dynamic Core Polarization in Strong-Field Ionization of CO Molecules

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The orientation-dependent strong-field ionization of CO molecules is investigated using the fully propagated three-dimensional time-dependent Hartree-Fock theory. The full ionization results are in good agreement with recent experiments. The comparisons between the full method and the single active orbital method show that although the core electrons are generally more tightly bound and contribute little to the total ionization yields, their dynamics cannot be ignored, which effectively modifies the behavior of electrons in the highest occupied molecular orbital. By incorporating it into the single active orbital method, we identify that the dynamic core polarization plays an important role in the strong-field tunneling ionization of CO molecules, which is helpful for the future development of the tunneling ionization theory of molecules beyond the single active electron approximation.

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Tunneling ionization (TI) is one of the most prominent processes for atoms and molecules subjected to strong laser fields. It ignites various atomic dynamics such as high-order harmonic generation, which forms the basis for attosecond science [1–3]. Recent advances in shaping and tailoring laser pulses as well as aligning molecules provide a more controlled manner to explore the fundamental concepts involved in TI, e.g., the tunneling time and tunneling wave packet [4–7]. In general, the molecular ionization dynamics in strong fields can be successfully described by the quasistatic theories [8–11], which assume that the molecular core is frozen and the laser field is not varying during the TI process. Studies based on these single active electron (SAE) models concluded that the orientation-dependent ionization rate maps the asymptotic electron density distribution, leading to the imaging of the ionizing orbitals [8–12].

However, it has been questioned recently when the observed ionization yields deviate from the expectation based on the shape of the highest occupied molecular orbital (HOMO) [13–20]. Among the various proposed models, the linear Stark effect [13–15] has been incorporated into the molecular Ammosov-Delone-Krainov (MO-ADK) theory [8–10] to explain the orientation-dependent ionization of the OCS molecule [13]. But recent measurements [21] on the orientation-dependent ionization of the CO molecule deviate apparently from the Stark-corrected MO-ADK. The Stark-corrected molecular strong-field approximation [14,22] predicts a result close to experiment, but this method still suffers from the long-standing gauge problem while no consensus has been reached [23]. Other experiments [22,24] also indicate that the linear Stark effect plays a minor role, and the ionization rate is dominated by the orbital profile. The numerical study based on SAE potentials [25] does not solve this puzzle on CO, which triggers an even more challenging need for nonperturbative treatment of the multielectron dynamics of molecules in intense laser pulses.

It has been evidenced that multiorbital and multipole effects come into play for strong-field physics [2,3,18,26–29]. However, the direct numerical integration of the time-dependent Schrödinger equation is computationally prohibitive for systems with more than two electrons [30–32]. The popular approximations include the time-dependent density functional theory [33–35] and the multiconfiguration time-dependent Hartree-Fock theory [36–38], but they both suffer from some disadvantages: the accuracy of the former depends critically on the formulation of the exchange-correlation potential, while the latter is computationally far more expensive, and at present the implementation is limited to either 1D approximations or very small molecules.

In this Letter, we investigate the orientation-dependent strong-field ionization of CO in intense laser fields by the fully propagated three-dimensional time-dependent Hartree-Fock (TDHF) theory [39] within the Born-Oppenheimer approximation. TDHF goes beyond the SAE approach and includes the response to the field of all electrons [39], which helps to identify the multipole effects from the molecular core in strong-field TI. TDHF ignores correlation effects, which play a minor role in the single ionization processes. The full ionization results are compared with the experiment [21] and good agreements are reached. Furthermore, we have performed the calculations using the single active orbital (SAO) approximation; i.e., we propagate the HOMO electrons while freezing the others. The comparisons between the SAO method and the full method show that although the core electrons are generally more tightly bound and contribute little to the total ionization yields, their dynamics cannot be ignored, which effectively modify the behaviors of HOMO electrons. We demonstrate that the dynamic core polarization plays an important role in the strong-field tunneling ionization of CO molecules.

TDHF is a single determinant theory and may therefore be applied to quite large systems. Although it includes no correlation, in strong-field cases, the question of how much and under what conditions correlation beyond the Hartree-Fock model is important still remains to be discussed [40–42]. For the numerical implementation, we use the prolate spheroidal coordinates [43], which is almost a natural choice for two-center systems. Our approach is also based on the discrete-variable representation and the finite-elements method [32]. Discrete-variable representation offers distinct advantages in the representation of local potential operators, while finite-elements method provides more flexibility in the design of a numerical grid and increases the sparseness of the kinetics matrix. For the temporal propagation, we use the efficient short iterative Lanczos algorithm [44].

The numerical parameters are as follows. The internuclear distance of CO is fixed at experimental equilibrium of 2.132 a.u. [45]. As the ground electronic state is $^1\Sigma$, the spin-restricted form of TDHF is adopted here. The ground state is determined by relaxing the system in imaginary time from a guess wave function. The electronic configuration of the ground state of CO from the relaxation calculation is $(1\sigma)^2(2\sigma)^2(3\sigma)^2(4\sigma)^2(1\pi)^4(5\sigma)^2$, with the total (HOMO) energy of $-112.790\,911\,8$ ($-0.554\,923\,304$) a.u., in good agreement with the literature values [46]. The electric field $\mathbf{E}(t)$ is linearly polarized (in the xz plane, with β denoting the orientation angle with respect to the molecular axis, see the inset in Fig. 1), $E(t) = E_0 \sin^2(\pi t/\tau) \sin(\omega t + \phi)$, where E_0 is the peak field amplitude, ω is the carrier frequency, τ is the pulse duration, and ϕ is the carrier envelope phase. The laser intensities of interest are in units of $I_0 = 10^{14}$ W/cm^2. After the time propagation T (long enough to allow all the ionization flux to pass by the absorbing boundary), we yield the total (orbital) wave function $\Psi(T)$ [$\psi_i(T)$]. The ionization probability from the orbital i is calculated as $p_i = 1 - \langle \psi_i(T) | \psi_i(T) \rangle$. The total ionization probability is $P = 1 - \langle \Psi(T) | \Psi(T) \rangle = 1 - \prod_i (1 - p_i)$, which can be approximated as $P \approx \sum_i p_i$ for small ionizations ($p_i \ll 1$).

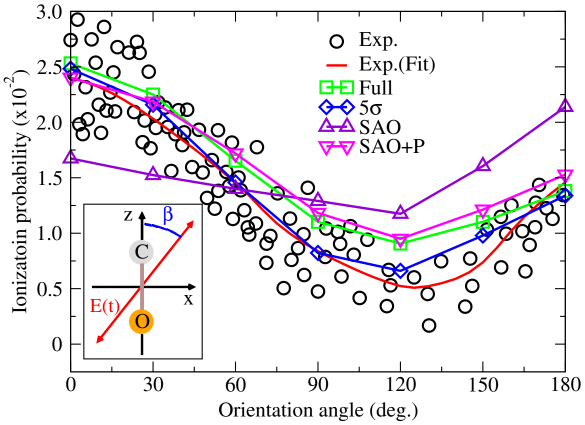

FIG. 1 (color online). The calculated ionization yields of CO versus the orientation angle β: total (open square) and 5σ (open diamond) (from the full method), SAO method (open upward triangle), and SAO + P method (open downward triangle). The experiment data (scattered circle) are taken from [21] (matched to the full calculation at $\beta = 0°$), where a circularly polarized laser field is used with estimated intensity of 4×10^{14} W/cm^2 and pulse duration of 35 fs. The fitted curve of the experimental data is shown in the red line. The inset shows the definition of the orientation angle β.

The orientation-dependent ionization yields of CO have been measured in an experiment [21], where it was found that the CO molecule could be more easily ionized at parallel orientation than at antiparallel orientation. In the experiment, a circularly polarized laser field with estimated intensity of $4I_0$ and pulse duration of 35 fs was used. For our linearly polarized laser fields, we use laser pulses of three optical cycles and an equivalent intensity of $2I_0$. The wavelength is 800 nm and carrier envelope phase $\phi = \pi/2$. The O (C) atom is located at the negative (positive) part of the z axis. Thus, for the parallel orientation ($\beta = 0°$), the maximum laser electric field points from C to O, while it points from O to C for the antiparallel orientation ($\beta = 180°$). According to the tunneling theory [8], the ionization rate decreases exponentially with the laser intensity. As a result, the ionization induced by the field with peak intensity dominates the total ionization yields, filtering out the cycle-averaging effects. Also, the ionization potential (IP) of 1π (HOMO-1) is 2.3 eV higher than that of 5σ [46]; thus, a small ionization from inner orbitals is expected. In Fig. 1, the ionization yields are compared with the experiment. The full method yields a ratio of $P(180°)/P(0°) = 0.62$, in good agreement with the averaged ratio extracted from the experiment. Both the calculation and experiment predict a minimum around 120°. Our results show that the total ionization is dominated by 5σ. Because of the orbital symmetry, the ionization from 1π is suppressed for both $\beta = 0°$ and 180°. An observable contribution is available only around perpendicular orientations, where the ionization from 5σ is suppressed. At perpendicular orientations, an ionization from 1π is enhanced, leaving the molecular ion in its excited states. TDHF is insufficient to treat the multiconfiguration processes due to its single configuration nature, resulting in the mild deviation of the full results and the experiment. Thus, we conclude that the combination contribution of multiple orbitals plays a minor role.

In order to identify the core effects, we perform SAO calculations in which only 5σ electrons are active, in contrast to the full method where all the electrons are fully propagated. Note that in our SAO method, the direct and exchange potentials between the core and HOMO electrons are calculated exactly at each time step in the framework of TDHF, which is different from the SAE methods where local model potentials are used [20,25]. The orientation-dependent ionization yields from SAO calculations deviate obviously from the full calculations and the experiment (Fig. 1). The SAO method predicts a larger ionization for $\beta = 180°$, yielding a ratio of $P(180°)/P(0°) = 1.28$, in qualitative disagreement with the experiment. A previous time-dependent Schrödinger equation study [25] based on a SAE potential fails as well, showing that a treatment beyond a single active electron (orbital) is required.

Hinted by the agreement of the experiment with the full method, which includes the responses of core electrons, we attempt to improve the SAO calculation by including the dynamic core polarization induced by the intense laser field [47],

$$V_p(\mathbf{r}, t) = -\frac{\boldsymbol{\alpha}_c \mathbf{E}(t) \cdot \mathbf{r}}{r^3}, \tag{1}$$

where $\boldsymbol{\alpha}_c$ is the total polarizability of core electrons. The SAO method including V_p is noted as SAO + P. We calculate this polarizability in the following way. In the full propagation, we have checked that the induced dipole moments of core electrons were mainly contributed from 1π and 4σ (HOMO-2). Fitting $\boldsymbol{\alpha}\mathbf{E}(t)$ to the numerical induced dipole moment $\mathbf{d}_{\rm ind}(t)$ of each orbital yields $\boldsymbol{\alpha}_{1\pi} = (2.55, 2.55, 4.68)$ a.u. and $\boldsymbol{\alpha}_{4\sigma} = (0.73, 0.73, 0.64)$ a.u. The total polarizability can be approximated as $\boldsymbol{\alpha}_c \approx \boldsymbol{\alpha}_{1\pi} + \boldsymbol{\alpha}_{4\sigma}$. Close to the core, we apply a cutoff for V_p at a point where the polarization field cancels the laser field [47]. This is also necessary to remove the singularity near the core. Taking the z axis, for example, the cutoff point z_c satisfies $\alpha_{zz}E/z_c^2 - z_c E = 0$, which results in $z_c = \alpha_{zz}^{1/3}$. As $\boldsymbol{\alpha}_c$ is anisotropic in general, all the cutoff points constitute an ellipsoidal surface. The SAO + P results are in good agreement with the full calculations and experiment (see Fig. 1).

By including the dynamic polarization, we see that V_p enhances the ionization for $\beta = 0°$, while the ionization is suppressed for $\beta = 180°$. This contradicts the prediction of the static theory, which collaborates the polarization effects into an effective IP [13]

$$I_p^{\rm eff}(\mathbf{E}) = I_p(0) + \Delta\boldsymbol{\mu} \cdot \mathbf{E} + \frac{1}{2}\mathbf{E}^T \Delta\boldsymbol{\alpha}\mathbf{E}, \tag{2}$$

where $\Delta\boldsymbol{\mu}$ ($\Delta\boldsymbol{\alpha}$) is the difference of the permanent dipole moment (polarizability) between CO and CO$^+$, and $I_p(0)$ is the IP of CO in the absence of external fields. The linear Stark shift takes into consideration the second term on the rhs of Eq. (2). I_p is raised (reduced) when the laser field is directed parallel (antiparallel) to the orbital dipole. As a result, the linear Stark effect reverses the orientation-dependent ionization rate and indicates a maximum ionization for $\beta = 180°$ [22]. It can be seen that the second-order Stark shift correction [the third term on the rhs of Eq. (2)] is helpless in this situation since I_p is raised for both $\beta = 0°$ and 180°. Therefore, the direct inclusion of the polarizability term (static polarization) in an IP [Eq. (2)] does not improve the MO-ADK theory in the CO case.

A theory of tunneling ionization in complex systems (CS-ADK) [47–49] was proposed a few years ago, where a similar core polarization potential was explicitly taken into account to improve the original MO-ADK theory. We have calculated the ionization probabilities of CO by CS-ADK, but the results were similar to those from MO-ADK, except for a mild suppression of the probabilities for both $\beta = 0°$ and 180°. To investigate the effects of $V_p(\mathbf{r}, t)$, we plot the effective potential $V_{\rm eff}$ felt by the 5σ electron along the molecular axis (Fig. 2). $V_{\rm eff}$ is defined as

$$V_{\rm eff}(\mathbf{r}, t) = V_n(\mathbf{r}) + \int \frac{\rho(\mathbf{r}', t)d^3r'}{|\mathbf{r} - \mathbf{r}'|} + \mathbf{E}(t) \cdot \mathbf{r} + V_p(\mathbf{r}, t), \tag{3}$$

where V_n is the interaction with the nuclei, and $\rho(\mathbf{r}, t)$ is the total electron density without the electron under consideration. Note that V_p is absent in the SAO method. The molecule is propagated from initial time until the field reaches the maximum amplitude of 0.0755 a.u. Thus, electronic dynamics is included in the effective potential. The field-dressed and field-free orbital energy ε of 5σ is presented in Fig. 2 with horizontal marks. $V_{\rm eff}$ is asymmetric for $\beta = 0°$ [Fig. 2(a)] and 180° [Fig. 2(b)], with a higher potential barrier for $\beta = 180°$. If the field-free ε is used, over-the-barrier ionization happens for $\beta = 0°$ and the original MO-ADK predicts a much smaller ionization ratio of $P(180°)/P(0°)$ than unity. The inclusion of polarization potential generally raises the potential barrier for both $\beta = 0°$ and 180°, leading to a suppression of the ionization probabilities in CS-ADK. In our SAO method, ε is lowered (lifted) for $\beta = 0°$ (180°) due to the linear stark effects. As a result, over-the-barrier ionization is almost

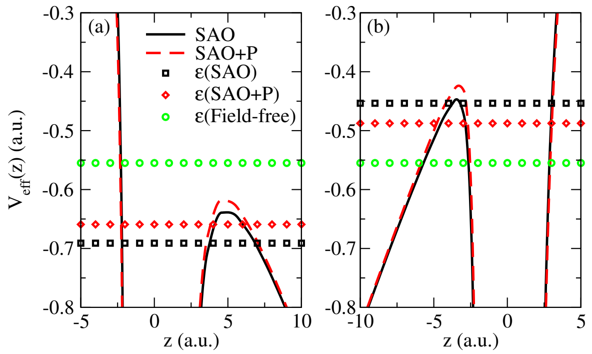

FIG. 2 (color online). Effective potential on the 5σ electrons along the molecular axis predicted by the SAO method (solid lines) and SAO + P method (dashed lines), when the maximum laser electric field ($|E| = 0.0755$ a.u.) points to O (a) and to C (b), respectively. The horizontal marks represent the corresponding field-dressed and field-free orbital energies of 5σ.

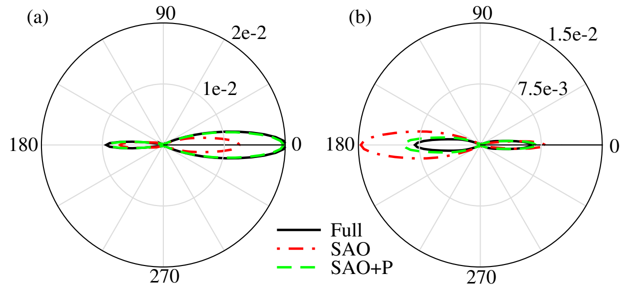

FIG. 3 (color online). Photoelectron angular distributions of CO for (a) parallel and (b) antiparallel orientation computed by the full method (solid lines), the SAO method (dot-dashed lines), and the SAO + P method (dashed lines).

satisfied for $\beta = 180°$ while electrons have to tunnel through a barrier to ionize for $\beta = 0°$, reversing the orientation-dependent ionization rate. The Stark-corrected MO-ADK theory fails to explain the experiments due to the same reason as the SAO method. In the SAO + P method, although the potential barrier is raised for both orientations, ε is shifted towards different directions: ε is lifted for $\beta = 0°$ and lowered for $\beta = 180°$. It reflects the orbital distortion and also the dynamics of 5σ electrons by the dynamic core polarization.

In order to fully characterize the ionization dynamics, we further investigate the photoelectron angular distribution (PAD) for different orientations. The PAD in direction $\hat{\mathbf{r}}$ is calculated as [25]

$$\frac{\partial P}{\partial \Omega} = \int_0^T \hat{\mathbf{r}} \cdot \mathbf{j}(R_b, t) dt, \qquad (4)$$

where $\mathbf{j}(R_b, t)$ is the flux calculated at R_b and time t in direction $\hat{\mathbf{r}} = (\theta, \varphi)$. The time T is chosen long enough to allow all the flux to pass R_b. In Fig. 3, the PADs integrated over the angle φ are computed at the same laser parameters as the total ionization yields for $\beta = 0°$ and $180°$. It can be seen that the emission of photoelectrons is mainly directed along the molecular axis. The major loaf is ionized by the peak electric field of the pulse, while subpeak fields ionize the minor part. The PADs from the SAO method deviate from the full results just as in the case of total ionizations (Fig. 1). The SAO + P method yields good agreement with the full method. Together with Figs. 1 and 3, we identify that the dynamic core polarization does improve the original SAO method and is crucial for the correct description of ionization for multielectron molecules subjected to intense few cycle pulses.

In conclusion, we demonstrated that the orientation dependence of strong-field ionization probabilities of CO is essentially affected by the core electronic states of multielectron molecules. The single active orbital method predicted qualitatively incorrect ionization yields due to the neglect of the core polarization dynamics. By including the polarization potential from the laser polarized molecular core, the results agreed with the experiment, allowing the identification of the importance of the dynamic core polarization. This is expected to have implications for high harmonic generation as well, where the encoded multielectron effects are being actively explored [2,3,28,29]. We conclude a theory beyond the single active electron is in need for the tunneling ionization of multielectron systems, by taking into account the dynamical distortion of the ionizing orbital as a prerequisite.

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