Attosecond stereo time discernible dynamics for strong-field ionization of HeH$^+$

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We study the ionization dynamics of oriented HeH$^+$ in strong linearly-polarized laser fields by numerically solving the time-dependent Schrödinger equation. The calculated photoelectron momentum distributions for parallel orientation show a striking asymmetric structure which, however, can not be described by present models. With a developed model pertinent to polar molecules, we trace the electron motion in real time. We show that this asymmetric structure arises from the interplay of the Coulomb effect and the permanent dipole play a non-negligible role. This structure can be used to probe the degree of orientation which is important in ultrafast experiments for polar molecules.

Introduction.—Above threshold ionization (ATI) is a basic process in strong-laser-matter interactions $^{1, 2}$, which has wide applications in attosecond science $^{3, 4}$. One of the important observables in ATI is the photoelectron momentum distribution (PMD), which encodes rich dynamical information of ATI and the structural information of the target $^{7, 8}$. Many experimental efforts and theoretical efforts including numerical solution of the time-dependent Schrödinger equation (TDSE) and analytical treatments based on strong-field approximations (SFA) $^{2, 3}$, have been devoted to this issue. It is shown that for the simple case of an atom interacting with a linearly-polarized single-color (LPS) laser field, the PMD is symmetric with respect to the axis perpendicular to the laser polarization $^{9}$. The phenomenon also holds for a symmetric linear molecule aligned parallel or perpendicular to the laser polarization $^{10, 11}$. This symmetry is easily understood with considering the time-domain symmetry of the laser pulse and the symmetric geometry of the target. The situation is different for an asymmetric molecule even it is oriented along the laser polarization. For example, numerical studies have shown that the PMD for HeH$^{2+}$ is not symmetric $^{12}$. Naturally, one can think that this asymmetry, a typical characteristic of PMD from polar molecules, is a result of the asymmetric geometry of the target. However, the underlying physical mechanism is unclear due to the lack of an appropriate analytical model.

Here, we study ATI from oriented HeH$^+$ $^{13, 14}$ with a permanent dipole (PD) numerically and analytically. The HeH$^+$ molecule (the first product of chemical reaction in the cosmos) has a stable ground state which can be manipulated in present experiments $^{15, 16}$. The TDSE is first solved with Born-Oppenheimer (BO) approximations, then it is extended to non-BO cases.

The TDSE predictions of PMD of HeH$^+$ show a striking asymmetry. This asymmetry, however, can not be described by the SFA with considering the PD effect (SFA-PD). With further considering the Coulomb modification in SFA-PD (MSFA-PD), this asymmetry is reproduced in our simulations. By contrast, this asymmetry also disappears when neglecting the PD effect in MSFA treatments (see Fig. 1). The comparisons provide deep insights into the complex origin of the asymmetric PMD. We show that the interplay of the Coulomb effect and the PD effect is mainly responsible for this asymmetric structure. When the PD effect destroys the symmetry of the ionization in two consecutive half-cycles, the Coulomb effect further destroys the symmetry of the ionization in the half laser cycle where the ionization mainly occurs. The stereo time dynamic is memorized by photoelectrons, resulting in the asymmetry of PMD.

Numerical and analytical methods.—In the BO case, the Hamiltonian of the HeH$^+$ system studied here has the form of $H(t) = H_0 + \mathbf{r} \cdot \mathbf{E}(t)$ (in atomic units of $\hbar = e = m_e = 1$). Here, the term $H_0 = \mathbf{p}^2/2 + V(\mathbf{r})$ is the field-free Hamiltonian, and $V(\mathbf{r}) = \sum_{j=1,2} Z_j e^{-R_j |r - \mathbf{r}_j|}$ is the Coulomb potential with $Z_j$ the charge at $\mathbf{r}_j$. The insets indicate the positions of these two nuclei He and H and the unit vector $\mathbf{e}$ along the laser polarization.
\[ \sqrt{(x-x_j)^2+(y-y_j)^2} \] and \( j = 1, 2 \). Here, \( Z_1 \) and \( Z_2 \) are the effective charges for the He and H centers, respectively. The indices \( i \) and \( o \) denote the inner and outer limits of \( Z_1 \) and \( Z_2 \). \( R_1 \) and \( R_2 \) are the positions of the He and H nuclei that have the coordinates of \( (x_1, y_1) \) and \( (x_2, y_2) \) in the \( xy \) plane, with \( x_{1/2} = \pm R_{1/2} \cos \theta \), \( y_{1/2} = \pm R_{1/2} \sin \theta \), \( R_1 = M_H R/(M_He + M_H) \) and \( R_2 = M_He R/(M_He + M_H) \). \( M_H \) and \( M_He \) are masses of He and H nuclei and \( \theta \) is the orientation angle. For HeH\(^+\), we have used the parameters of \( Z_{1i} = 2/3, Z_{1o} = 1/3, Z_{2o} = 4/3, \) and \( Z_{2o} = 2/3 \). \( \xi = 0.5 \) is the softening parameter. \( \rho(R) \) is the screening parameter, which is adjusted such that the ionization potential \( I_p(R) \) of the model molecule at the distance \( R \) reproduced here matches the real one. For example, for the equilibrium separation of \( R = 1.4 \) a.u., \( I_p = 1.65 \) a.u., \( \rho(R) = 0.94 \), and for the stretched case of \( R = 1.8 \) a.u. with \( I_p = 1.5 \) a.u., \( \rho(R) = 1.21 \). For non-BO cases, we follow the procedure introduced in detail in [17].

The term \( E(t) \) is the electric field which has the form of \( E(t) = \mathbf{E}(t) \) of \( E_x(t) = f(t) E_0 \sin(\omega_0 t) \). \( \mathbf{E} \) is the unit vector along the laser polarization which is parallel to the \( x \) axis here. \( E_0 \) is the maximal laser amplitude relating to the peak intensity \( I \) of \( E_x(t) \). \( \omega_0 \) is the laser frequency and \( f(t) \) is the envelope function. We use trapezoidally shaped laser pulses with a total duration of 8 optical cycles and linear ramps of two optical cycles. The details for solving TDSE of \( i \dot{\Psi}(t) = H(t) \Psi(t) \) with spectral method [18] and obtaining the PDM can be found in [19, 20]. Unless mentioned elsewhere, the parameters used here are \( I = 1.5 \times 10^{15} \text{W/cm}^2 \), \( \omega_0 = 0.114 \) a.u., \( R = 1.4 \) a.u. and \( \theta = 0^\circ \) at which the molecular axis is parallel to \( \mathbf{E} \).

To analytically study the ATI of polar molecules, as in [21], we first incorporate the PD effect into SFA. With the saddle-point approximation, the tunneling amplitude for the photoelectron with the drift momentum \( p \) and the complex ionization time \( t_s = t_0 + i t_x \) can be written as

\[ F(p, t_s) = F(p, t_0) \propto \sum \beta |\mathbf{E}(t_4) \cdot \mathbf{d}_1(p + A(t_4)) e^{-i \beta \mathbf{E}(t_4) \cdot \mathbf{d}_1} |^2 \]

with \( S = S(p, t_4) \) and \( \beta \equiv (1/\det(t_4))^{1/2} \). Here, \( \mathbf{d}_1(v) = \langle v|\mathbf{j}|0\rangle \) is the bound-free transition matrix element, \( S(p, t_4) = \int_0^t (p + A(t_4')/2 + p + E(t_4') \cdot \mathbf{D}) dt'd'' \), and \( \mathbf{A}(t) \) is the vector potential of the electric field \( \mathbf{E}(t) \) and \( \mathbf{D} \) is the permanent dipole. Generally, the value of \( \mathbf{D} \) used in the expression of \( F(p, t_s) \) should be the one related to the ground state of the polar molecule. However, the value of \( \mathbf{D} \) depends on the choice of the coordinate origin. Here, we assume that \( \mathbf{D} = \mathbf{D}_1 - \mathbf{D}_2 \) where \( \mathbf{D}_1 (\mathbf{D}_2) \) is the PD related to the ground state (the first excited state) of the asymmetric system. Our simulations show that with the above definition, the value of \( \mathbf{D} \) is invariant when varying the coordinate origin. In this paper, we will call the above SFA with considering the PD effect the SFA-PD. In our TDSE cases, for \( R = 1.4 \) a.u., \( D = -0.43 \) a.u. and for \( R = 1.8 \) a.u., \( D = -0.83 \) a.u. These values will also be used in our model treatments.

Then as in [22, 23], we solve the Newton equation

\[ \mathbf{r}(p, t) = -\mathbf{E}(t) - \nabla_{\mathbf{r}} V(\mathbf{r}) \] for each SFA electron trajectory \( (p, t_s) \), with initial conditions [24] \( \mathbf{r}(p, t_0) = p + A(t_0) \) (the exiting momentum) and \( \mathbf{r}(p, t_0) = Re(\int_{t_0}^{t_1} p + A(t') dt') \) (the exiting position). Here, the real part \( t_0 \) of \( t_s \) is considered as the exiting time. The final Coulomb-modulated drift momentum is obtained with \( p_f = \mathbf{r}(p, t \to \infty) \), which is relating to the amplitude \( F(p, t_0) \). In this paper, we will call the above Coulomb-modified SFA-PD the MSFA-PD. According to the SFA, the exiting time \( t_0 \) agrees with the ionization time \( t_i \) at which the instantaneous energy \( E_{in}(t) = |\mathbf{r}(p, t)|^2/2 + V(\mathbf{r}) \) becomes larger than zero. However, as discussed in [25], the MSFA predicts a time lag \( t_d = t_i - t_0 \) with \( t_d > 0 \). This lag influences remarkably on dynamics of the laser-driven system. It can change the direction of the drift momentum, e.g., from \( p_x < 0 \) to \( p_x > 0 \) in our cases, and importantly increases the contributions of long trajectory to ATI. As a result, the PDM of SFA is modified remarkably. Below, we will show that for HeH\(^+\), the time lag \( t_d \) which is related to electrons exiting the potential along the H side plays an important role in the PDM of the polar molecule. Accordingly, we term the direction-dependent time lag \( t_d \) the stereo one.

**Asymmetric PMD.**—In Fig. 1(a), we show the TDSE photoelectron momentum distribution for HeH\(^+\) in BO cases. This distribution shows a remarkable asymmetry with respect to the axis of \( p_x = 0 \). The SFA-PD result, as shown in Fig. 1(b), however, shows a symmetric distribution, implying that the PD effect itself does not induce this asymmetry. With considering the Coulomb effect, as seen in Fig. 1(c), the MSFA-PD clearly reproduces this asymmetry. In addition, the interference structures of the distribution in Fig. 1(c) are also comparable to the TDSE ones in Fig. 1(a). By contrast, when assuming \( D = 0 \) in the MSFA-PD simulations, a symmetric distribution is observed, as seen in Fig. 1(d). This result implies that the asymmetric Coulomb potential itself also does not induce the asymmetric distribution. From these comparisons, one can conclude that the asymmetric PDM of polar molecules is closely related to the interaction of PD and Coulomb effects.

**Potential mechanism.**—To understand the potential mechanism, first, we introduce the PD effect on ATI of polar molecules. In Figs. 2(a) to 2(c), we present a sketch of the laser-dressed states related to the PD effect and the electric field \( E(t) \) in one laser cycle. As discussed in [20], the PD effect depends strongly on the polarization of the laser field. For the first half cycle with \( E(t) > 0 \) at which electrons escape from the H side, due to the PD effect, the energy of the ground state \( |0\rangle \) (the first excited state \( |1\rangle \)) is dressed up (down), making the ionization easier to occur (see Fig. 2(a)). For the second half cycle with \( E(t) < 0 \) at which electrons tunnel out of the potential along the He side, the situation reverses and the ionization is more difficult to occur (see Fig. 2(b)). This phenomenon has been termed asymmetric ionization of polar molecules [26]. The classical prediction [27, 28] of drift momenta \( p_x(t) = A_x(t) \) is also presented in Fig. 2(c), indicating that the electron born in the time regions
I and IV (II and III) contributes to $p_x < 0$ ($p_x > 0$).

With the introduction of the PD effect, in Fig. 2(d), we further compare time-dependent ionization probabilities in one laser cycle predicted with different methods. The TDSE ones are obtained with evaluating $I(t) = 1 - \sum_m |\langle m | \Psi(t) \rangle|^2$. Here, $|m\rangle$ is the bound eigenstate of $H_0$ obtained through imaginary-time propagation. The model ones are obtained with calculating $I(t) = \sum_{p_x} |F(p_x, t_0)|^2$ at $E_0(t) > 0$. The SFA-PD results give a good illumination on the asymmetric ionization, with showing a remarkable increase just around the time of $t = 4.25T$ in the first half cycle and a small increase just around $t = 4.75T$ in the second half cycle. Here, $T = 2\pi/\omega_0$. The TDSE results, however, show a larger increase around a time remarkably later than $t = 4.25T$ (about 100 attoseconds) in the first half cycle and a smaller increase around a time near $t = 4.75T$ for the second half cycle. These phenomena of asymmetric ionization and striking ionization time lag in two consecutive half-cycles are reproduced by the MSFA-PD. Without considering the PD effect, the MSFA fails for predicting the asymmetric-ionization phenomenon. These results in Fig. 2(d) are in agreement with those in Fig. 1. From Fig. 2, this asymmetry in PMD can be easily understood. For SFA-PD and MSFA without PD, the contributions of regions II and III versus regions I and IV to ionization are almost the same. For TDSE and MSFA-PD, the contributions of regions II and III are remarkably larger than those of regions I and IV. These results indicate that due to both the PD effect which induces the dominating ionization for electrons exiting the potential along the H side, and the Coulomb effect which induces the large stereo ionization time lag related to the H-side ionization, the PMD for $p_x > 0$ has larger amplitudes than that for $p_x < 0$, explaining the asymmetry in PMD observed in Figs. 1(a) and 1(c). This remaining difference between TDSE and MSFA-PD predictions, especially for the second half cycle may be due to the omission of the excited-state effect in MSFA-PD treatments, which is not easy to incorporate into the MSFA at present.

This asymmetric structure also depends on the molecular orientation. In Fig. 3, we show the PMD at $\theta = 180^\circ$, obtained with TDSE-BO and MSFA-PD. Compared to the case of $\theta = 0^\circ$, the He (H) nucleus is located in the left (right) side for $\theta = 180^\circ$. With this inversion of the molecular geometry, one can observe the inversion of the asymmetric structure in PMD for both of numerical and analytical results here. Similar analyses to Fig. 2 for the case of $\theta = 180^\circ$ further support our above conclusions that the Coulomb-induced large ionization time lag, mainly responsible for this asymmetric structure of PMD, is always related to the electron exiting the potential along the side of the lighter nucleus. In other words, the asymmetric PMD of polar molecules is dependent on the exiting position of tunneling electron and encodes attosecond stereo time dynamics of polar molecules in strong laser fields. The meaning of this finding will be discussed later. We mention that for $\theta = 90^\circ$, this asymmetry in PMD disappears in our simulations.

**Asymmetry degree.**—We have also extended our simulations to other laser parameters and to non-BO cases. Relevant results are presented in Fig. 4, where we plot the asymmetry degree of the PMD which is defined as the ratio of the total amplitudes with $p_x > 0$ to those of $p_x < 0$. The TDSE results with $R = 1.4$ a.u. in Fig. 4(a) show that on the whole, this degree is larger for shorter laser wavelengths and lower laser intensities. These parameter-dependent phenomena are basically re-

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**Figure 2:** A sketch of the PD effect and the Coulomb-induced ionization time lag. (a) and (b): the field-free (dotted curves) and laser-dressed (solid curves) asymmetric potentials and these two field-free ($|0\rangle$ and $|1\rangle$) and laser-dressed ($|0'\rangle$ and $|1'\rangle$) lowest states when the laser polarization is antiparallel (a) or parallel (b) to the permanent dipole (which is directing from the He nucleus to the H nucleus); (c): the fields of $E_x(t)$ and $-A_x(t)$; (d): time-dependent ionization probabilities calculated with TDSE-BO, SFA-PD, MSFA-PD and MSFA without considering PD. For comparison, each model curve in (d) is multiplied vertically by a constant factor to match the TDSE one. The vertical lines divide the laser cycle into four parts (I-IV) as shown.

**Figure 3:** Photoelectron momentum distributions of HeH⁺ obtained with different methods at $\theta = 180^\circ$. (a): TDSE-BO; (b): MSFA-PD. The insets are as in Fig. 1.
simulations at distance is around the present laser parameters, the maximal stretching distance $\alpha$ obtained with TDSE-BO at different degrees of orientation.

Potential applications.—One of the potential applications of the asymmetric PMD is to evaluate the degree of orientation $\alpha = (n_u - n_d)/(n_u + n_d)$ achieved in experiments. Here, $n_u$ ($n_d$) is the number of the molecule pointing up (down). In Fig. 5, we show the PMDs of TDSE-BO at different degrees of orientation $\alpha$ with assuming perfect alignment. One can observe that with the decrease of the value of $\alpha$, this asymmetry in PMD diminishes and for $\alpha = 0$ (random orientation), a symmetric PMD is observed. In other words, this asymmetric structure is sensitive to the degree of orientation. Using this phenomenon, one can evaluate the value of $\alpha$. In Fig. 5(c), we plot the asymmetry degree versus $\alpha$, obtained with TDSE-BO simulations and a simple formula of $\gamma(\alpha, \beta) = [(\sqrt{1+\alpha})/2 + \sqrt{(1-\alpha)/(2\beta)})^2]/[\sqrt{(1+\alpha)/(2\beta)} + \sqrt{(1-\alpha)/2}]$. Here, $\gamma$ is the asymmetry degree at $\alpha$, and $\beta$ is that at the perfect orientation obtained with TDSE simulations. This formula can be easily deduced with the assumption that for each $|p_x|$, the PMD of $p_x > 0$ differs from that of $p_x < 0$ only for a constant factor $\gamma$. The TDSE and analytical results agree with each other, implying that this formula can be used to evaluate the degree of orientation.

In summary, we have studied ATI of HeH$^+$ in LPS laser fields. The PMD of HeH$^+$ shows a striking asymmetry beyond the predictions of previous models. With the development of a model which considers both the Coulomb effect and the PD effect, we show that this remarkable asymmetry arises from attosecond stereo time dynamics of polar molecules in strong laser fields. The PD effect induces a preferred ionization when the active electron escapes from a certain side of the molecule. The Coulomb effect further induces a large time lag (about 100 attosecond) between tunneling and ionization, which remarkably changes the symmetry of PMD. This mechanism is essential for polar molecules with a large PD such as CO and is believed to have important influences on other strong-field processes such as high-order ATI and non-sequential double ionization. Besides monitoring the electron tunneling dynamics with attosecond resolution, the asymmetry of PMD for polar molecules can also be used for calibrating the degree of orientation.

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