Strong-field double ionization dynamics of vibrating $\text{HeH}^+$ versus $\text{HeT}^+$

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(Dated: November 19, 2019)

We study double ionization (DI) dynamics of vibrating $\text{HeH}^+$ versus its isotopic variant $\text{HeT}^+$ in strong laser fields numerically. Our simulations show that for both cases, these two electrons in DI prefer to release together along the $\text{H(T)}$ side. At the same time, however, the single ionization (SI) is preferred when the first electron escapes along the $\text{He}$ side. This potential mechanism is attributed to the interplay of the rescattering of the first electron and the Coulomb induced large ionization time lag. On the other hand, the nuclear motion increases the contributions of these two electrons releasing together along the $\text{He}$ side. This effect differentiates DI of $\text{HeH}^+$ from $\text{HeT}^+$.

I. INTRODUCTION

Strong-laser-matter interaction leads to many interesting physical processes, such as above-threshold ionization (ATI) [1,2], high-harmonic generation (HHG) [3,4,5], double and multiple ionization [6,11], and laser induced electron diffraction [12,13], etc., which have promising applications in attosecond science [14].

Present studies have revealed the importance of tunneling and rescattering in strong-field processes [15,16]. In comparison with other strong-field processes, double and multiple ionization which involves electron-electron correlation includes richer physical phenomena. As the double ionization (DI) from atoms and symmetric molecules have been studied widely [10], DI from polar molecules with a large permanent dipole [17,18] is less studied. Especially, when the nuclear motion is considered, the situation is more complex. It has been shown that for small polar molecules such as $\text{HeH}^+$, the interaction of the strong laser field and the permanent dipole induces the rapid nuclear motion which has important influences on HHG and ATI of the asymmetric system [19,23]. In addition, the interplay of the Coulomb effect and the permanent-dipole effect also gives rise to a strong asymmetry in photoelectron momentum distributions (PMD) for single ionization (SI) of $\text{HeH}^+$ [24]. This asymmetry is closely related to the permanent dipole induced asymmetric ionization [25] and the Coulomb induced large ionization time delay [26]. Effects of these mechanisms on DI of the asymmetric system are not clear so far.

In this paper, we focus on DI of $\text{HeH}^+$ and its isotopic variant $\text{HeT}^+$ in strong linearly polarized laser fields beyond the Born-Oppenheimer (BO) approximation. The $\text{HeH}^+$ system, the simplest polar heteronuclear molecule, has served theoretically and experimentally as a fundamental model for understanding molecular formation and electron correlation [27]. Numerical solution of the time-dependent Schrödinger equation (TDSE) of the vibrating two-electron system in full dimensions is still not within reach and is easily limited by the existing computing capability. Thus, we use a simplified model where the motion of all particles is restricted to one dimension (1D). It has been shown that such a model can reproduce all important strong-field effects such as multiphoton ionization and HHG [28,30]. This simplified model can also describe qualitatively the correlation effects between electrons and interplay between the electronic and the nuclear motion [31,34].

The calculated PMDs of DI for $\text{HeH}^+$ or $\text{HeT}^+$ show a striking asymmetry with indicating that these two electrons in the DI process prefer to release together along the $\text{H(T)}$ side. This phenomenon in DI differs remarkably from that in SI for $\text{HeH}^+$ or $\text{HeT}^+$, which shows that the first electron in SI prefers to escape along the $\text{He}$ side. This disagreement between DI and SI strongly implies that the rescattering of the first electron plays an important role in DI of the asymmetric system. This rescattering along with the Coulomb induced large ionization time delay remarkably increases the DI yields and results in preferred DI along the $\text{H(T)}$ side. On the other hand, the rapid nuclear motion, which differs remarkably for $\text{HeH}^+$ and $\text{HeT}^+$, increases the contributions of direct ionization (which is preferred along the $\text{He}$ side) to DI.

II. NUMERICAL METHODS

The Hamiltonian of the asymmetric molecule $\text{HeH}^+$ studied here has the following form (in atomic units of $\hbar = e = m_e = 1$):

$$\hat{H}(t) = -\frac{\partial^2}{2\mu_N} + \frac{Z_1Z_2}{R} + \frac{1}{\sqrt{(x_1-x_2)^2 + \epsilon}} + \sum_{j=1}^{2} \left[ -\frac{\nabla z_j^2}{2\mu_e} + V_{en}(R, x_j) + x_jE(t) \right].$$

(1)

Here $R$ is the internuclear separation and $x_j$ ($j=1,2$) is the electronic coordinate. $\mu_N = M_HM_T/(M_H+M_T)$ is the nuclear reduced mass and $\mu_e = (M_H+M_T)/(M_H+$
$M_H + 1) \approx 1$ is the electronic reduced mass. $M_He$ and $M_H$ are masses of He and H nuclei. The term $V_{en}$ denotes the interaction between the electron and nuclei and has the following form:

$$V_{en}(R, x) = -\frac{Z_1}{\sqrt{(x - R_1)^2 + \epsilon}} - \frac{Z_2}{\sqrt{(x - R_2)^2 + \epsilon}},$$

(2)

where $Z_1 = 2$ and $Z_2 = 1$ are the charges for He and H centers, respectively. $R_1$ and $R_2$ are positions of He and H nuclei with $R_1 = M_H R/(M_H + M_H)$ and $R_2 = -M_He R/(M_He + M_H)$. $\epsilon = 0.59$ is the smoothening parameter, which is adjusted such that the ground-state energy of the model HeH$^+$ molecule matches the real one of $E_0 = -2.98$ a.u. The equilibrium separation of model HeH$^+$ studied here is $R_e = 2$ a.u., which also holds for model HeT$^+$ and is somewhat larger than the real one of $R_e = 1.4$ a.u.. Here, we have used the length-gauge form of the interaction Hamiltonian. The laser field used here is $E(t) = E_0 f(t) \sin(\omega t)$ with peak amplitude $E_0$, envelope function $f(t)$ and laser frequency $\omega$. In our simulations, we use a seven-cycle laser pulse which is linearly turned on and off for two optical cycles, and kept at a constant intensity for three additional cycles.

We use $\Psi(R, x_1, x_2, t) = \Phi(t)$ on a three-dimensional grid to represent the wave function. The TDSE of $\Psi(t) = H(t) \Phi(t)$ is solved numerically using the spectral method \[32\]. A grid size of $L_{x_1} \times L_{x_2} = 204.8 \times 204.8$ a.u. with the grid step of $\Delta x_1 = \Delta x_2 = 0.4$ a.u. for the electron, a range of $R = 0.6..6.9$ a.u. with the grid step of $\Delta R = 0.1$ a.u. for the internuclear distance, and a time step of $\Delta t = 0.05$ a.u. have proven sufficient convergence for describing the strong field dynamics. In order to avoid the reflection of the electron wave packet from the boundary and obtain the momentum space wave function, the coordinate space is split into the inner and the outer regions with $\Psi(R, x) = \Psi_{in}(R) + \Psi_{out}(R)$, by multiplication using a mask function $F(x_1, x_2, R) = F_1(x_1) F_2(x_2) F_3(R)$. Here, $F_1(x_1) = \cos^{1/2}[\pi[(x_1 - |r_0|)/(L_{x_1} - 2|r_0|)]$ for $|x_1| \geq r_0$ and $F_1(x_1) = 1$ for $|x_1| < r_0$. $r_0 = 3/8 L_{x_1}$ is the critical boundary between the inner and the outer regions for one electron. The relevant electron wave packet passing through this critical boundary will be absorbed by the mask function smoothly. The form of $F_2(x_2)$ is similar to $F_1(x_1)$. A similar absorbing procedure with the mask function $F_3(R)$ is also used for the upper boundary of $R$. In the inner region, the wave function $\Psi_{in}(t)$ is propagated with the complete Hamiltonian $H(t)$. In the outer region, the time evolution of the wave function $\Psi_{out}(t)$ is carried out in momentum space with the Hamiltonian of the free electron in the laser field \[36\]-\[38\]. The mask function is applied at each time interval of 1 a.u. and the obtained new fractions of the outer wave function at the DI condition of $|x_1| \geq r_b$ and $|x_2| \geq r_b$, denoted with $\Psi^{d}_{out}(t)$, are added coherently or non-coherently to the corresponding momentum-space wave function $\Psi^{d}_{out}(t)$. Finally, we obtain PMDs $c_d(p_1, p_2)$ of DI from $\Psi^{d}_{out}(t)$. Similarly, with coherently or non-coherently adding the obtained new fractions of

the outer wave function at the SI condition of $|x_1| < r_b$ or $|x_2| < r_b$, denoted with $\Psi^{s}_{out}(t)$, to the corresponding momentum-space wave function $\Psi^{s}_{out}(t)$, one can obtain PMDs $c_s(p_1, p_2)$ of SI from $\Psi^{s}_{out}(t)$. Here, $r_b = 8$ a.u., which defines the spacial region where the electron is considered to be located at bound states \[34\]. Accordingly, the integral of the loss at the DI (SI) grid boundaries over time gives the total DI (SI) probability $P(d(s))$ with $P(d(s)) = \int [1 - |\Psi_{in}(R, x_1, x_2, t)|^2] dRdx \int_0^{\infty} dt = \int \gamma_{d(s)}(R) dR$ at $|x_1| \geq r_b$ and $|x_2| \geq r_b$ ($|x_1| < r_b$ or $|x_2| < r_b$), which includes contributions $\gamma_{d(s)}(R)$ at different $R$. Here, we focus on the main characteristics of PMD from HeH$^+$ versus HeT$^+$. For clarity, we present the non-coherent results, i.e., $c_d(p_1, p_2) = \int |\Psi^{d}_{out}(t)|^2 dR dt = \int \beta_d(R) dR$, and $c_s(p_1, p_2) = \int |\Psi^{s}_{out}(t)|^2 dR dt = \int \beta_s(R) dR$, which also include R-dependent contributions $\beta_d(s)(R)$. This TDSE treatment for HeT$^+$ is similar to that for HeH$^+$ with replacing H by T in relevant expressions.

### III. RESULTS AND DISCUSSIONS

#### A. Asymmetric PMDs of DI

In Fig. 1, we show calculated PMDs of DI for HeH$^+$ and HeT$^+$ at different laser parameters. These distributions indicate momentum correlation between these
To explore the potential mechanism, in Fig. 2 we plot the time-dependent ionization probabilities in one laser cycle, which is approximately evaluated with $P(t) = 1 - \sum_{n=15}^{15} \langle n|\Psi(t)\rangle^2$ and is relating to SI. Here, $|n\rangle$ is the $n$th electronic bound eigenstate of the field-free Hamiltonian $H_0 = \frac{1}{\sqrt{(x_1-x_2)^2 + \epsilon^2}} + \sum_{j=1}^{2} \frac{\psi_j^2}{2\mu} + V_{ee}(R, x_j)\rangle$ at the BO approximation. Excluding more bound-state components from $|\Psi(t)\rangle$, results are similar to $P(t)$. We divide the one-cycle time region into four parts denoted with I-IV. First, for all cases in Fig. 2, the ionization is strong in the first half laser cycle and is weak in the second half laser cycle. This phenomenon has been termed as asymmetric ionization and is identified as arising from the effect of the permanent dipole [25]. Secondly, in the first half laser cycle, the ionization mainly occurs in the region II after the laser field arrives at its peak. The reason has been attributed to the Coulomb induced large ionization time delay. Due to this delay, many electrons which tunnel out of the laser-Coulomb formed barrier near the peak of the laser field in region I are ionized finally in region II [26]. By comparison, in the second half laser cycle, the contributions in region IV after the time of peak intensity also dominate the ionization for the same reason of Coulomb induced delay as in the first half cycle, but the contributions of region III are also non-negligible. In Ref. [39], it has been shown that the contributions in region III arise from effects of excited states. Specifically, some electrons are pumped into the excited states from the ground state around the peak of the laser field in the first half laser cycle and survive the falling part of the laser field of region II. Then the excited-state electrons with lower ionization potentials are ionized mostly in the arising part of the laser field of region III. Thirdly, as increasing the laser intensity and wavelength, the contributions of region III decrease due to the decrease of the excited state effect, as discussed in [20]. Fourthly, when the ionization yields of HeH$^+$ with lighter nuclei are larger than HeT$^+$, a careful analysis tells that the ionization asymmetry in the first and the second half laser cycle is somewhat more remarkable for HeT$^+$ than for HeH$^+$. These SI characteristics will be used to analyze the potential DI mechanisms.

A simple evaluation on the drift momentum of the electron with the classical expression $p = -A(t)$ tells that electrons born in regions I and IV (II and III) have minus (plus) momenta. Here, $A(t)$ is the vector potential of $E(t)$. The time-dependent asymmetric ionization results in Fig. 2 thus imply that for SI, electrons...
with plus momenta have large amplitudes. This point has been shown in Ref. [24] for vibrating HeH$^+$ or HeT$^+$ (b-c). In (a), the one-cycle time region is divided into four parts (I-IV), labeled by different colors. The insets in (a) plot the laser-dressed asymmetric Coulomb potential, the laser-dressed electronic states $|0'\rangle$ and $|1'\rangle$ corresponding to the free-field electronic ground state $|0\rangle$ and the first excited state $|1\rangle$ of HeH$^+$ or HeT$^+$, when the laser polarization is antiparallel (regions I and II) or parallel (regions III and IV) to the permanent dipole which is directing from the He nucleus to the H(T) nucleus. For the antiparallel case, the electronic ground state is dressed up and the first excited state is dressed down, making the ionization easier to occur. This situation reverses for the parallel case. As a result, the ionization is strong (weak) in the first (second) half laser cycle for the present cases. These analyses are also applicable for HeH$^{2+}$ or HeT$^{2+}$. In (b) and (c), possible sequential (L1 and L3) and non-sequential (L2 and L4) DI routes associated with the first electron born in regions II (b) and IV (c) are plotted.

C. Mechanisms of DI

In Fig. 4(a), we plot the electric field $E(t)$ in one laser cycle, with dividing the time region into four parts as in Fig. 2. The laser-dressed Coulomb potential and the laser-dressed two lowest electronic states of HeH$^+$ (HeT$^+$) corresponding to the first and the second half laser cycle are also plotted here as the insets. When the SI mainly occurs in regions II and IV, as discussed in Fig. 2 we focus on possible DI routes associated with SI events in these two regions.

First, in Fig. 4(b), we plot DI routes associated with the birth of the first electron in region II. In this case, as the first electron ionizes, the second electron can be ionized directly by the external field in this region with contributing to the first quadrant in PMDs of DI. It should be noted that due to the Coulomb induced large ionization time lag, these two ionized electrons in region II can find their origins in region I. We denote these SDI routes contributing to the first quadrant “L1”. The first electron born in region II can also return to and recollide with the second electron in region III (short NDSI route) and region IV (long one). For the short route, the Coulomb effect will also induce the delay of DI time, just as it does in region I, resulting in the emission of these similar to SI. We therefore anticipate that non-sequential double ionization (NSDI) dominates in present cases. In fact, extended TDSE simulations for 1D vibrating HeH$^{2+}$ (HeT$^{2+}$) show that for the present laser parameters, the ionization probability of HeH$^{2+}$ (HeT$^{2+}$) from its ground state is remarkably lower than the ratio of DI to SI for HeH$^+$ (HeT$^+$), as shown in Fig. 3 suggesting that NSDI dominates here [24]. Next, we discuss possible routes of NSDI in detail.
two electrons in region IV and contributing to DI in the third quadrant. For the long one, both electrons will also contribute to DI in the third quadrant. We denote these NSDI routes contributing to the third quadrant “L2”. For lower laser intensities, probabilities for direct ionization of the second electron by the laser field are small, and the route L2 dominates in DI for the cases in Fig. 5(b).

For DI routes associated with the birth of the first electron in region IV, this situation reverses, as plotted in Fig. 5(c). In this case, NSDI routes associated with the rescattering of the first electron contribute to the first quadrant and SDI routes related to sequential ionization of these two electrons contribute to the third quadrant. We denote these SDI and NSDI routes “L3” and “L4” respectively. Due to that the SI amplitudes are smaller in region IV than those in region II and the second electron is bounded more deeply in region IV than in region II (see the insets in Fig. 2(a)), for a laser cycle, the main contributions to DI come from route L2, with PMDs of DI showing large amplitudes in the third quadrant. When the laser intensity increases, the ionization yields of HeH$^+$ associated with direct ionization by the laser field increase and the contributions of route L1 increase. As a result, PMDs of DI for the asymmetric system become more symmetric. For increasing the laser wavelength, the asymmetric system stretches to a large distance at which direct ionization is usually easier to occur, resulting in somewhat similar results as increasing the laser intensity.

For HeT$^+$ with heavier nuclei than HeH$^+$, at the same laser parameters, the laser-induced stretching for HeT$^+$ is smaller than for HeH$^+$, as shown in Fig. 4. Generally, direct ionization prefers larger R at which the ionization potential of the vibrating system is lower. Accordingly, direct-ionization yields for HeT$^+$ are also smaller than for HeH$^+$. As a result, PMDs of DI for HeT$^+$ usually show a stronger asymmetry than for HeH$^+$.

To validate these above discussions, in Fig. 6 we plot the ratio of amplitudes of PMDs of DI in the first quadrant to those in the third quadrant “L2”. When the laser intensity increases, the ionization yields of HeH$^+$ associated with direct ionization by the laser field are small, and the route L2 dominates in DI for the cases in Fig. 6(b).

Figure 6: Ratio of PMD amplitudes of DI in the 1st quadrant to those in the 3rd quadrant for HeH$^+$ (a) and HeT$^+$ (b) at different laser parameters as shown.

Figure 7: Comparisons between R-dependent ionization probabilities $\gamma_d(R)$ of SI (a) versus $\gamma_d(R)$ of DI (b) for HeH$^+$ and HeT$^+$, obtained with different laser parameters as shown.

D. R-resolved DI and SI

To provide further insight into electron-nucleus coupled DI dynamics of polar molecules, in Fig. 7 we plot R-dependent probabilities $\gamma_d(R)$ of SI and $\gamma_d(R)$ of DI for HeH$^+$ and HeT$^+$, averaged by the corresponding total probabilities $P_d$ and $P_{d2}$, respectively, at different laser parameters.

First, as increasing laser intensities and wavelengths, for SI, the structure of these R-dependent distributions in Fig. 7 changes from a relatively sharp hump to the plane and broad one, but the center of the hump does not change basically. For DI, however, the distributions extend to somewhat larger distances. These different responses of DI and SI on laser parameters revealed here agree with the experimental results in [21]. Secondly, in some cases such as for HeH$^+$, the location of the hump both for SI and DI is remarkably larger than the equilibrium separation $R_e = 2$ a.u.. By comparison, for the symmetric case of model H$_2$ [34], the position of the hump is nearer to the equilibrium distance and the structure of the hump is not sensitive to laser parameters. The results suggest that due to the permanent-dipole induced rapid stretching which remarkably diminishes the ion-
Figure 8: R-dependent PMDs $\beta_d(R)$ of DI for HeH$^+$ (the left column) and HeT$^+$ (right). The laser parameters are as in Figs. 1(e) and 1(f).

IV. CONCLUSION

In conclusion, we have studied the ionization dynamics of vibrating HeH$^+$ with comparing it to HeT$^+$. The photoelectron momentum distributions for both DI and SI show an asymmetric structure but with the contrary trend. As the Coulomb induced large ionization time delay plays an important role in the asymmetry in SI, the rescattering of the first electron along with this delay contributes to the asymmetry in DI. The nuclear motion mainly influences the events of SDI. The contributions of SDI decreases the asymmetry in DI momentum distributions, and this decreases is more remarkable for HeH$^+$ with lighter nuclei and more rapid nuclear motion than for HeT$^+$. This asymmetry in DI is expected to appear for other oriented polar molecules with a large permanent dipole. In addition, the proposed DI mechanism of rescattering followed by Coulomb induced ionization time delay holds for all of atoms and molecules including symmetric and asymmetric ones. For these symmetric cases without a permanent dipole, this asymmetry discussed in the paper does not appear in PMDs of DI. However, effects relating to this mechanism are possible to resolve with using two-dimensional laser fields which have shown the capability in probing sub-cycle strong-field electron dynamics.

Acknowledgement

This work is financially supported by the National Key Research and Development Program of China (Grant No. 2018YFB0504400); the National Natural Science Foundation of China (Grant Nos. 91750111 and 11904072); the Research Team of Quantum Many-body Theory and Quantum Control in Shaanxi Province, China (Grant No. 2017KCT-12); and the Fundamental Research Funds for the Central Universities, China (Grant Nos. 2017TS008 and GK201801009).

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