Manipulating nonsequential double ionization via alignment of asymmetric molecules

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Abstract: Using classical three-dimensional ensembles, we demonstrate that nonsequential double ionization of HeH+ molecules in an intense laser field can be manipulated by controlling the alignment of the molecular axis relative to the laser field. Both the symmetry of the correlated electron momentum spectrum in the direction parallel to the laser field and the total double ionization yield strongly depend on the angle between the molecular axis and the laser field. When the molecular axis is aligned parallel to the laser field, double ionization is most probable and the correlated electron momentum spectrum parallel to the laser field from nonsequential double ionization exhibits the most asymmetry with respect to the minor diagonal.

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References and links


Nowadays, it is well known that the physical mechanism for nonsequential double ionization (NSDI) of atoms by strong laser fields for infrared wavelengths is based on the classical “recollision model” [1]. The electron-electron correlation momentum spectra have revealed rich recollision dynamics in NSDI process [2, 3, 4, 5, 6, 7, 8]. In the recollision model, the first ionized electron is driven back to its parent ion by the oscillating laser field, causing the ionization of the second electron in a direct (e, 2e)-like encounter or indirectly via recollision-induced excitation of the ion plus subsequent field ionization (RESI) [3]. At lower laser intensities, RESI mechanism dominates NSDI and the two electrons primarily acquire longitudinal momenta (along the laser polarization axis) near zero. While at higher laser intensities (e, 2e) mechanism dominates NSDI and the two electrons preferentially acquire large longitudinal momenta.

Experiments have provided evidence that recollision mechanism is also responsible for NSDI of isonuclear diatomic molecules [9, 10, 11, 12]. Moreover, the experiments on NSDI of N₂ and O₂ molecules [12] have shown that the molecular structure decisively influences the electron momentum correlation pattern. Both calculations based on quantum mechanical S matrix [12] and a semiclassical model [13] have predicted a strong influence of the molecular alignment on double ionization. Controlling attosecond double ionization dynamics has been experimentally demonstrated in NSDI of N₂ by changing the molecular alignment relative to the ionizing laser field [14]. It was found that the two electrons involved in NSDI more likely exit the molecule in the same direction for parallel molecules than for perpendicular molecules. For heteronuclear
diatomic molecules, because of the presence of a permanent dipole moment in such system, strong-field single ionization [15, 16], as well as strong-field double ionization [17], is very different from that for atoms and isonuclear diatomic molecules. The one-dimensional quantum-mechanical calculations predicted that the two electrons involved in NSDI of HeH\textsuperscript{+} by intense laser pulses tend to be simultaneously ejected in the direction of its permanent dipole when the molecular axis is parallel to the laser polarization direction [17]. Thus, an approach to manipulate the emission of the two electrons from NSDI was proposed by controlling the alignment of asymmetric diatomic molecules relative to the laser field.

In this paper, with the classical three-dimensional (3D) ensemble model, we firstly investigate the dynamics of nonsequential double ionization of HeH\textsuperscript{+} in an intense laser field at different angles between the molecular axis and the laser field. We find that both the electron momentum correlation pattern and the total double ionization yield are strongly dependent on the angle between the molecular axis and the laser field.

The 3D classical ensemble model has been described in detail in [18]. The electron-nuclear interaction and the electron-electron interaction for HeH\textsuperscript{+} are represented by a two-center 3D soft-Coulomb potential (in atomic units) \( V(\mathbf{r}_1, \mathbf{r}_2) = -2/\sqrt{(\mathbf{r}_1 + \mathbf{R}/2)^2 + a_1^2 - 1/\sqrt{(\mathbf{r}_1 - \mathbf{R}/2)^2 + a_2^2 - 2/\sqrt{(\mathbf{r}_2 + \mathbf{R}/2)^2 + a_1^2 - 1/\sqrt{(\mathbf{r}_2 - \mathbf{R}/2)^2 + a_2^2 + 1/\sqrt{(\mathbf{r}_2 - \mathbf{R}/2)^2 + b^2}}}} \)

\( \mathbf{r}_1, \mathbf{r}_2 \) represent the electronic coordinates, and \( \mathbf{R} \) is the internuclear vector. \( a_1, a_2 \) are shielding parameters for the attraction of He\textsuperscript{2+} and H\textsuperscript{+}, respectively, and \( b \) for the electron-electron interaction. The evolution of the two electrons are governed by Newton’s equations of motion:

\( \frac{d^2 \mathbf{r}_i}{dt^2} = -E(t) - \nabla_x V(\mathbf{r}_1, \mathbf{r}_2), \) with \( i = 1, 2 \) denoting different electrons. \( E(t) \) is a linearly polarized laser field. Without loss of generality the molecule is assumed to lie in the \( x-z \) plane. In present calculations, the He and H nuclei are fixed at \((-2\sin \theta, 0, -2\cos \theta)\) and \((2\sin \theta, 0, 2\cos \theta)\) a.u. respectively, where \( \theta \) is the angle between the molecular axis and the \( z \) axis. Thus, the internuclear distance is 4 a.u. and \( \mathbf{R}=(4\sin \theta, 0, 4\cos \theta) \) a.u. The ground-state energy for HeH\textsuperscript{+} at internuclear distances above 2 a.u. is very close to that for He \((-2.9035 \text{ a.u.})\) [19]. Similar to [20], \( a_1 \) is initially set to 0.825 in order to avoiding autoionization and \( b \) is set to 0.05. As a consequence, \( a_2 = 1.5 \) guarantees a stable initial ensemble.

Our calculations are not expected to reproduce the real double ionization of HeH\textsuperscript{+} molecules, but to study the dependence of the correlated electron momentum spectrum on the molecular alignment relative to the laser field. The classical model of molecules with the approximation that the nuclei are held fixed has been shown to be suitable to describe many features of double ionization of molecules by intense laser field [21].

The initial ensemble is generated by allowing the electron pair to evolve freely without a laser field for a long time. The starting positions of the electron pair are \((0, 0, -1)\) and \((0, 0, +1)\) a.u. The available kinetic energy is distributed between the electrons randomly. The three velocity components of each electron have the same absolute value with sign randomly selected. The positions and momenta of the electron pair in evolution at a time interval 0.5 a.u. give a sufficiently randomized initial ensemble. Changing the starting positions of the electron pair or the starting momentum distribution of each electron does affect details of the distribution of the initial ensemble very little. Figures 1(a) and 1(b) show the position distribution of the initial ensemble of HeH\textsuperscript{+} along the \( x \) and \( z \) axes respectively when the molecular axis is parallel to the \( z \) axis. The position distribution of the initial ensemble along the \( x \) axis is symmetric about \( x_1 = -x_2 \). However, because of asymmetric charge configuration of HeH\textsuperscript{+}, the position distribution of the initial ensemble along the \( z \) axis centralizes on the He nucleus. Once the initial ensemble is obtained, the laser field is turned on and all trajectories are evolved in the combined Coulomb and laser fields. To investigate the influence of the permanent dipole moment of HeH\textsuperscript{2+} on the electron dynamics in NSDI, we reduce the nuclear screening parameters \( a_1, a_2 \) to 0.2 as soon...
Fig. 1. (color online). The density distribution of trajectories of the initial ensemble for \( \theta = 0^\circ \) in position (a) \( x \), and (b) \( z \) space, respectively. The ensemble size is \( 1.6 \times 10^6 \) trajectories.

Fig. 2. (color online). Electron correlation for the double ionization of HeH\(^+\) at laser intensity \( 4 \times 10^{14} \text{ W/cm}^2 \) and wavelength 800 nm for \( \theta = 0^\circ \). Horizontal axis: momentum of electron a parallel to the laser polarization direction. Vertical axis: momentum of electron b parallel to the laser polarization direction. An ensemble size of \( 4 \times 10^5 \) is used.

as one electron achieves \( r = 12 \text{ a.u.} \). To conserve total energy, the radial kinetic energy of each electron is increased [22]. Double ionization is determined if both electrons achieve positive total energies at the end of the laser pulse [23].

We use a 800 nm linearly polarized laser pulse with an intensity of \( 4 \times 10^{14} \text{ W/cm}^2 \) and a total duration of 10 optical cycles, switched on and off linearly over 2 optical cycles. The polarization direction of the laser field is along the \( z \) axis. The angle \( \theta \) between the molecular axis and the laser field is varied in the range from \( 0^\circ \) to \( 90^\circ \). Figure 2 shows the electron momentum correlation spectrum in the direction parallel to the laser field for \( \theta = 0^\circ \). \( k_a^\parallel, k_b^\parallel \) represent the momentum components of the two electrons parallel to the laser field direction, respectively.
The correlation pattern in the first and third quadrants is obviously asymmetric with respect to the diagonal $k^a_{||} = -k^b_{||}$. Both electrons with high momenta most likely exit the molecule with positive final momenta. Evidently, these events originate from the (e, 2e) mechanism because of the fact that both electrons acquire the same large drift momentum in the (e, 2e) process [3] from the laser field after recollision. In addition, the correlation pattern in the first quadrant exhibits a fingerlike structure, which suggests binary and recoil collisions [6] in the (e,2e) process.

In the $k^a_{||}, k^b_{||}$ correlation spectrum the asymmetry is more clear by separating double ionization events according to different mechanisms. Figure 3(a) shows the correlation pattern from events only due to the (e, 2e) mechanism. About 30% of the total double ionization trajectories are due to the (e, 2e) mechanism. Further, both electrons are found to be emitted predominantly with positive final momenta. For the (e, 2e) mechanism, the recollision often occurs close to the zero crossing of the oscillating field and thus both electrons accumulate the same high parallel drift momentum from the laser field [24, 3]. Plot (b) of Fig. 3 shows a histogram of the counts of double ionization trajectories from Fig. 3(a) vs laser phase at the recollision time. The recollision time is defined as the time of closest approach of the two electrons during the (e, 2e) recollision. Schematically, the black solid curve denotes the laser electric field and the red solid curve shows the drift momentum $-A(t_{r})$ that the electrons acquire from the laser field after recollision, where $t_{r}$ is the recollision time and $A(t)$ is the vector potential of the
laser $A(t) = - \int_{-\infty}^{t} E(t') \, dt'$. Most of recollisions occur in the phase range $0.25 - 0.6T$. The two electrons due to these recollisions acquire positive drift momenta from the laser field and thus most of them achieve positive final momenta. Only a few of recollisions occur in the phase range $0.75 - 1T$. These recollisions result in both electrons primarily achieving negative final momenta.

Plot (c) of Fig. 3 shows the counts of recolliding electrons from opposite hemispheres vs kinetic energy when they reach $r = 7 \text{ a.u.}$ before recollision. The blue bands represent recolliding electrons from the positive $z$ axis and the red bands represent electrons from the negative $z$ axis. For single ionization of HeH$^+$, because of the electron cloud of the ground state predominantly localized on the He nucleus, the electrons are mostly ionized in the direction of the negative $z$ axis [17]. However, Fig. 3(c) evidently shows that most of recolliding electrons come from the positive $z$ axis. Moreover, the maximum kinetic energy of electrons from the positive $z$ axis is about 20 eV higher than that from the negative $z$ axis, which is in good agreement with the prediction of a simple one-dimensional classical model including the Coulomb field of the permanent dipole of HeH$^{2+}$ [17]. The asymmetric charge configuration of HeH$^{2+}$ essentially defines a permanent dipole. According to the convention, the dipole moment is directed from the negative charge to the positive charge. The center of negative charge of HeH$^{2+}$ is near the He nucleus and the center of positive charge is $R/3$ right from the He nucleus. Therefore, the dipole moment of HeH$^{2+}$ is directed from the He nucleus to the H nucleus [15]. Figure 3(c) reveals that the permanent dipole attracts the electrons returning from the positive $z$ axis while repulses them from the negative $z$ axis. Thus, the recolliding electrons from the positive $z$ axis dominate recollisions. As a consequence, the two electrons due to the $(e, 2e)$ mechanism are more likely to be ejected in the direction of the positive $z$ axis.

By changing the alignment of the permanent dipole of the molecule with respect to the laser polarization direction, we can control the influence of the permanent dipole on the dynamics of the recolliding electron and thus the electron momentum correlation pattern. Figures 4(a) and 4(b) show the $k_a^\parallel k_b^\parallel$ correlation spectrum for $\theta = 45^\circ$ and $90^\circ$, respectively. For $\theta = 45^\circ$, the correlation pattern also exhibits a clear asymmetry with respect to the diagonal $k_a^\parallel = - k_b^\parallel$. However, the asymmetry is reduced compared to that for $\theta = 0^\circ$. In addition, the fingerlike structure disappears in the momentum correlation pattern for $\theta = 45^\circ$, which implies that recoil...
Fig. 5. (color online). The probability of double ionization of HeH$^+$ at laser intensity $4 \times 10^{14}$ W/cm$^2$ and wavelength 800 nm as a function of $\theta$. The blue solid line is drawn to guide the eye.

collisions become less probable as the $\theta$ increases. Because the laser pulse is linearly polarized, the recolliding electron moves mainly in the direction of the laser field. As the angle between the laser field and the permanent dipole increases from $0^\circ$ to $90^\circ$, projection of the permanent dipole on the laser field decreases. Therefore, the influence of the permanent dipole on the recolliding electron reduces as $\theta$ increases, which results in decreasing of the maximum kinetic energy of recolliding electrons from the positive $z$ axis while in increasing from the opposite direction. The trajectory back-analyzing confirms this interpretation. As a consequence, the ratio of the number of double ionization events from the first quadrant versus from the third quadrant reduces as $\theta$ increases.

When the molecular axis is perpendicular to the laser field, the $k_{||}^a, k_{||}^b$ correlation spectrum becomes symmetric with respect to both diagonals, as shown in Fig. 4(b). It has been predicted in [17] because both the charge configuration of HeH$^+$ and the laser field are symmetric with respect to the molecular axis when $\theta = 90^\circ$. Thus, our results show that the symmetry of the electron momentum correlation in the direction parallel to the laser field can be manipulated by changing the alignment of the asymmetric molecule relative to the laser field. Further, the double ionization yield also has a strong dependence on the alignment of HeH$^+$ molecules. Figure 5 shows that the double ionization probability of HeH$^+$ molecules quickly decreases as $\theta$ increases from $0^\circ$ to $90^\circ$.

This dependence results from the influence of the transversal component of the permanent dipole, perpendicular to the laser field, on the returning electrons upon reaching the nuclei. The transversal component of the permanent dipole imposes a transversal net Coulomb force on the rescattering electron as it is close to HeH$^{2+}$. The transversal net Coulomb force leads the returning electron to have a significant transversal momentum before recollision occurs and may miss recollision with the parent ion finally. This net Coulomb force is proportional to the transversal component of the permanent dipole. As a consequence, the number of efficient recollisions quickly decreases as the transversal component of the permanent dipole increases. Thus, the yield of double ionization due to recollision mechanism quickly decreases as $\theta$ increases from $0^\circ$ to $90^\circ$. When the molecular alignment is perpendicular to the laser polarization, the transversal net Coulomb force achieves the maximum and the probability of recollision, as well as the yield of double ionization, is the lowest. Implicit in this explanation is that the single ionization of molecules is insensitive to alignment, which is confirmed in a multiphoton single ionization experiment of aligned N$_2$ [25] and by molecular tunnel ionization theory [26].
When the nuclear shielding parameters $a_1, a_2$ keep unchanged during the whole laser pulse, the key features of NSDI of HeH$^+$ are not altered. Our calculations show the total double ionization yield varies little while the asymmetry of the correlated electron momentum spectrum with respect to the minor diagonal reduces for the same molecular alignment. Since for larger nuclear shielding parameters the permanent dipole of HeH$^2+$ becomes less and thus its influence on the electron dynamics in NSDI decreases.

In summary, we have shown that during nonsequential double ionization the dynamics of the recolliding electrons can be controlled by changing the alignment of asymmetric molecules relative to the laser field. The permanent dipole of the molecule has a strong influence not only on the maximum kinetic energy of the recolliding electrons, but also on their transversal momenta. This provides a strong constraint on possible recollision mechanisms. Thus, controlling the alignment of asymmetric molecules can manipulate the symmetry of the electron momentum correlation spectrum and the double ionization yield at the same time.

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