Nonsequential double ionization of the aligned hydrogen molecule in strong field*

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This paper studies the nonsequential double ionization (NSDI) process of diatomic molecules aligned parallel and perpendicular to an intense linearly polarized laser field by using a three-dimensional semiclassical model. With this model, it achieves insight into the ion momentum distribution under the combined influence of a two-centre Coulomb potential and an intense laser field, and this result shows the significant influence of molecular alignment on the ratio between double and single ionization rate. Careful investigations show that the NSDI process for different alignment molecules has a close relation to the laser intensity and the different bounding electron distribution has a significant influence on the final ion momentum distribution.

Keywords: nonsequential double ionization, ionization probability, alignment molecule

PACC: 3380K

1. Introduction

Since the experimental realization of the generation of ultrafast laser pulses, the interaction of atoms and molecules with such intense light sources has gained much attention in recent years.[1−12] Studies of the ionization of molecules indicate that molecules are very different from atoms due to their additional nuclear degrees of freedom, orbital symmetries, many-centre interference effects, orientation of the molecule and vibrations.[13] Diatomic molecules show a much higher double ionization yield than the prediction of the single-active-electron model, which is a typical nonsequential double ionization (NSDI) process. However, theory of the molecular NSDI is still a challenging task. The double ionization (DI) yields and the ionized-electron momentum distribution exhibit a strong dependence on molecular structure and alignment. But the details of the pivot remain unknown.

Nowadays some approximate approaches have been developed, such as the time-dependent Schrödinger equation, S-matrix calculation, classical trajectory Monte Carlo calculations and semiclassical model (rescattering model) to describe the NSDI processes.[14−21] Now, it is commonly believed that rescattering is the dominant mechanism for NSDI process and it is responsible for many unusual observations and can give some explanations for many interesting phenomena. A rescattering process can be understood from a simple quasiclassical notion: once an electron in a strong field has undergone a transition into continuum from its initial bound state, its motion is dominated by its interaction with the laser field. In the case of a linearly polarized field, a majority of these electrons will be driven back to the vicinity of the ion core and undergo elastic or inelastic scattering, or be recaptured into the ground state by emitting a high-energy photon.

In this paper, the NSDI process of the molecule H_2 when it is aligned parallel and perpendicular to the polarization direction of the laser field is studied by using a semiclassical rescattering model developed recently.[10] The main purpose of this work is to investigate how the molecular alignment affects the NSDI dynamics and also show the laser intensity dependence of the NSDI process of H_2. The investigations show the bounding electron distribution's effect on the final ion momentum distribution.

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2. Model

The semiclassical rescattering model adopted in our calculations is described by the following two steps as stated above. Firstly, the ionization of the first electron is treated by tunnelling ionization theory considering the two-centre characteristic of the molecule. Then the subsequent evolution of the two electrons is determined by the classical Newtonian equations of motion[10]

$$\frac{\mathrm{d}^2\boldsymbol{r}_i}{\mathrm{d}t^2} = \boldsymbol{E}(t) - \nabla(V_{\mathrm{ne}}^i + V_{\mathrm{ee}}), \tag{1}$$

where $\boldsymbol{E}(t) = (\varepsilon(t)\sin\theta, 0, \varepsilon(t)\cos\theta)$, θ is the angle between the external field and the internuclear axis and $\varepsilon(t) = \varepsilon\cos(\omega t)$. V_{ne}^i and V_{ee} are the Coulomb interactions between nuclei and electrons and between two electrons, respectively. $V_{\mathrm{ne}}^i = -Z_{\mathrm{eff}}/r_{\mathrm{a}i} - Z_{\mathrm{eff}}/r_{\mathrm{b}i}$, $V_{\mathrm{ee}} = 1/|\,\boldsymbol{r}_1 - \boldsymbol{r}_2\,|$, where the radii $r_{\mathrm{a}i}$ and $r_{\mathrm{b}i}$ are the distances between the i^{th} electron and nuclei a and b, respectively.

In this paper, the simplest diatomic molecule is H_2 chosen for the diatomic molecule to be studied both theoretically and experimentally.[22] In our simulation, for H_2, $Z_{\mathrm{eff}} = 1$, the weight of each classical trajectory in the ensemble is $w(t_0, v_{\mathrm{per}}) = w(0)w(1)$.[10] Here $w(0)$ is the tunnelling rate in the quasi-static approximation:

$$w(0) = \frac{16\cdot c_1^2(2|I_{\mathrm{p1}}|)^{5/2}}{\varepsilon^2}\mathrm{e}^{-\kappa R}\int_0^{2\pi}\mathrm{d}\phi\int_0^1\left\{\cosh^2\left(\frac{\kappa R\mu}{2}\right)\times\frac{\mu\mathrm{d}\mu}{[\sin\theta\cos\phi(1-\mu^2)^{1/2}+\mu\cos\theta]^2}\right.$$
$$\left.\times\exp\left[-\frac{2}{3\varepsilon}\frac{(2|I_{\mathrm{p1}}|)^{3/2}}{|\sin\theta\cos\phi(1-\mu^2)^{1/2}+\mu\cos\theta|}\right]\right\}, \tag{2}$$

when the molecular alignment is parallel to the laser polarization direction, $\theta = 0$; when it is perpendicular to the laser polarization direction, $\theta = \pi/2$. $w(1) = (2|I_{\mathrm{p1}}|)^{1/2}/(\varepsilon\pi)\exp[-v_{\mathrm{per}}^2(2|I_{\mathrm{p1}}|)^{1/2}/\varepsilon]$ is the transverse velocity distribution, satisfying a Gaussian distribution.

To solve the Newtonian equations of motion, the initial conditions of the electrons are as follows. The classical-trajectory Monte Carlo method to produce the initial condition of the bound electron is used. The azimuthal symmetry of the system allows us to use the cylindrical coordinates (z, ρ, φ). For each grid point (z_i, ρ_i, φ_i) uniformly distributed in the three-dimensional space; to get the ground-state wave function of the singly-charged molecular ion H_2^+, we use a linear combination of the atomic orbital and molecular orbital approximations. The electron's position and momentum are constrained by the energy requirement that the electron total energy E satisfies $E_{\mathrm{min}} < E < E_{\mathrm{max}}$. Here $E_{\mathrm{min}} = |I_{\mathrm{p2}}| - 0.05$ a.u. and $E_{\mathrm{max}} = |I_{\mathrm{p2}}| + 0.05$ a.u., where I_{p2} is the ionization potential of the singly-charged molecular ion. In our computation, the parameters are chosen as $I_{\mathrm{p1}} = -0.5684$ a.u., $I_{\mathrm{p2}} = -1.1026$ a.u. (for the internuclear distance $R = 2.0$ a.u.) and $\omega = 0.05695$ a.u., corresponding to the wavelength $\lambda = 800$ nm.

3. Nonsequential double ionization for different molecular alignments

In this section we apply our theory to calculate the double ionization of the hydrogen molecule double ionization for alignment of the molecular axis parallel and perpendicular to the polarization axis respectively. About 9000 double ionization cases are obtained in our calculation (both for parallel and perpendicular conditions). In the calculation, the field strength is a constant during $t < 5T$ (T is the period of the laser field) and is turned off in a cosine-squared shape during the last two periods.

Figure 1 shows the single ionization rate for H_2 in Eq. (2). A comparison between the numerical result of Saenz[23] and ours shows that the ionization rate gives a good agreement with Saenz.

The ratios between double ionization (DI) and single ionization (SI) of the H_2 molecule for the parallel and perpendicular alignment with respect to the laser field are presented in Fig. 2. It shows that the

ratio ($= [H_2^{2+}]/[H_2^+]$) depends on the intensity for the parallel and perpendicular alignment. The internuclear separation is 2.0 a.u., and the peak laser intensities vary from $I = 1.0 \times 10^{14}$ W/cm^2 to $I = 2.0 \times 10^{14}$ W/cm^2. The dotted line with circles shows the parallel cases and the solid line with squares shows the perpendicular ones. The overall behaviour is almost monotonically raised with increasing intensity. It also shows that there remains a tendency: the ratio between DI and SI yields is greater for parallel aligned molecules than that for perpendicular ones. The difference between the parallel and perpendicular molecules may come from the molecular orbital.[13,24,25]

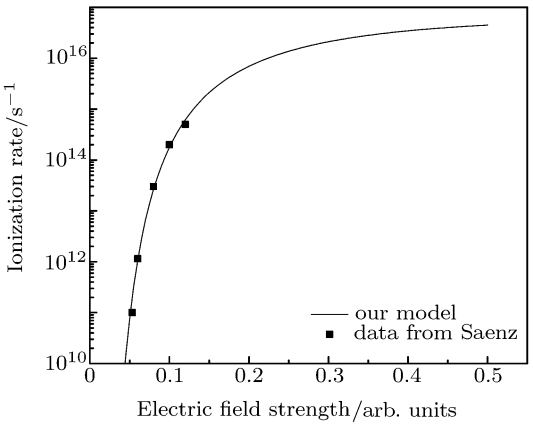


Fig. 1. Comparison of the single ionization rate of H_2 given by the two-centre ADK theory (solid line) and the result of Saenz[23] (squares).

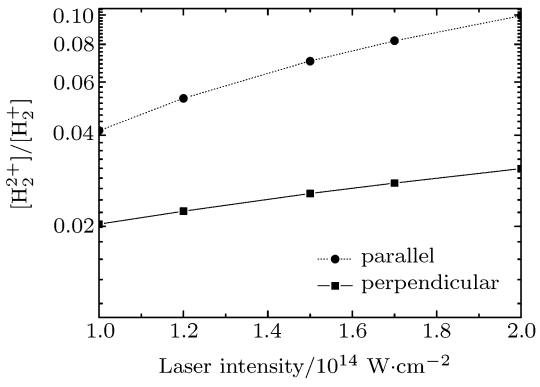


Fig. 2. The dependence of the ratio between DI and SI of the different alignment molecules H_2 on the laser intensity. The internuclear separation is 2.0 a.u.

Figure 3 presents DI probabilities as a function of time at $I = 1.5 \times 10^{14}$ W/cm^2 for both alignment (all in arbitrary units, parallel molecules DI probability: line with circles; perpendicular molecules DI probability: line with hollow circles). The general trend of the curves is quite similar. For both alignments, the DI curves present a ladder-like increase. The DI curves rise steeply around the zero of the field; this is a typical signature of instant ionization. Meanwhile the DI probability also increases around the maxima of the field, which is a signature of excited ionization.[10] From our calculation the DI probabilities are quite similar for parallel and perpendicular molecules; the ionization probability is slightly larger for parallel than for perpendicular cases, this is due to the nature of alignment-dependent ionization of H_2.

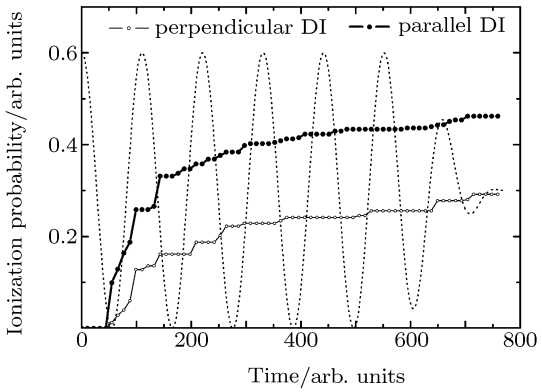


Fig. 3. The DI rate as a function of time. The intensity of the laser pulse is $I = 1.5 \times 10^{14}$ W/cm^2, the internuclear distance is $R = 2.0$ a.u. The dashed line shows the shape of the pulse (see text).

The schematic of the rescattering process is depicted as: the H_2 is first ionized, releasing an electron into the oscillating laser field. When the electron is driven back to the molecular ion, the internuclear distance may extend. After the collision, the other electron will be excited to a higher excited state or ionize. In this section, the internuclear distances at $R = 1.4$ a.u. and $R = 2.0$ a.u. are considered. Figure 4 shows the ion momentum (along the direction of the field) distributions of the H_2^{2+} molecular ion. Figures 4(a) and 4(b) presents the momentum distribution of the molecules oriented parallel and perpendicular to the polarization direction with the internuclear $R = 1.4$ a.u. and $I_{p2} = -1.28427$ a.u. Figures 4(c) and 4(d) plot the distributions of the momentum of the molecules oriented parallel and perpendicular to the polarization direction with $R = 2.0$ a.u. and $I_{p2} = -1.1026$ a.u. at the intensity $I = 1.5 \times 10^{14}$ W/cm^2. It is interesting to note that whatever the internuclear distance is, the ion momentum distribution shows a triple-hump structure for both alignments. This structure comes from a dis-

tribution peak at the zero momentum (line with circles), and another peak at non-zero momenta (line with squares), which implies that the two electrons escape the binding potential within the same half laser cycle after the first electron–electron collision. The observation tells us that when the internuclear distance is fixed, parallel aligned molecules are more likely to distribute at non-zero momenta. For the same alignment molecule, the final ion momentum distribution will be wider with increasing internuclear distance.

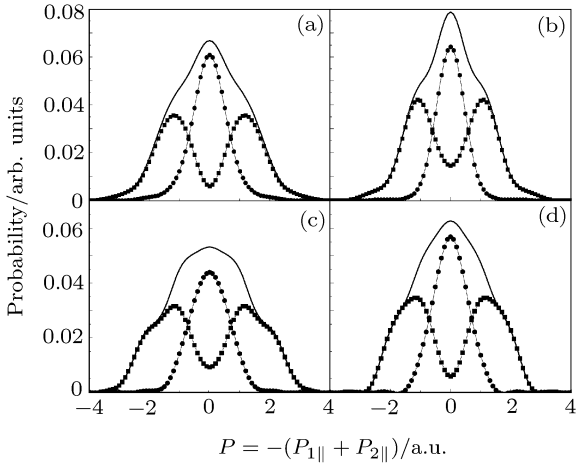


Fig. 4. The ion momentum (along the direction of the field) distributions of the H_2^{2+} molecule. The intensity of the laser pulse is $I = 1.5 \times 10^{14}$ W/cm^2: (a) parallel molecule with $R = 1.4$ a.u.; (b) perpendicular molecule with $R = 1.4$ a.u.; (c) parallel molecule with $R = 2.0$ a.u.; (d) perpendicular molecule with $R = 2.0$ a.u. (see text). Line with solid dots: distribution peaked at zero momentum. Line with squares: distribution peaked at nonzero momentum. Solid line: the final distribution of ion momentum.

The different internuclear distance (different bounding electron distribution) corresponds to the different returning time of the first electron,[24,26,27] and the H_2^+ will be excited to the σ_u or π_u electronic state. The ionization of H_2^+ in a laser field depends sensitively on the potential.[10,25] The H_2^+ with different internuclear distance makes the electron feel the different bounding energy and meanwhile the different alignment makes the H_2^+ ion potential more complex. Further research of more excited states is in preparation.

4. Conclusion

In conclusion, the alignment dependence of NSDI of H_2 has been investigated by using a semiclassical rescattering model. The ratio ($= [H_2^{2+}]/[H_2^+]$) of ionization probabilities of DI and SI increases as the intensity increases. The ionization probability of DI is a function of time, which is quite consistent with the rescattering picture. The results investigating different excited states play a novel role in different alignment molecules. With the different returning time of the first ionized electron, the nuclear wave packet propagates from its mean internuclear distance to some other one, since the H_2^+ ionizes sensitively to its potential, making the final momentum distribution different. With more consideration of the intrinsic properties of the molecule, our model can give more perfect descriptions of the rescattering progress and explanations for the physical picture.

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