Enhanced ionization of perpendicularly aligned $H_2$ in intense laser fields

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**A R T I C L E   I N F O**

Article history:
Received 20 October 2010
Received in revised form 17 January 2011
Accepted 17 January 2011
Available online 1 February 2011

Keywords:
Double ionization
Internuclear distance
Enhanced ionization

**A B S T R A C T**

Using three-dimensional classical ensembles, we have investigated the enhancement of double ionization of perpendicularly aligned $H_2$ molecules by a 800 nm laser pulse with intensity ranging from $1 \times 10^{14}$ W/cm$^2$ to $6 \times 10^{14}$ W/cm$^2$. The simulated results show that double ionization probability of $H_2$ strongly depends on $R$ and reaches a maximum at an intensity independent critical distance $R_c = 5$ a.u. Furthermore, the enhancement of double ionization is more pronounced in the cases of weaker or stronger fields. These results, a well indication of the influence of molecular structures and laser–molecule interactions on double ionization of diatomic molecules, are analyzed in detail and qualitatively explained based on the field-induced barrier suppression mechanism and back analysis.

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1. Introduction

Ionization of molecules exposed to intense fields strongly depends on internuclear distance $R$. When $R$ is stretched to a critical internuclear distance $R_c$, which is twice as long as or much longer than the equilibrium internuclear distance $R_e$, ionization of molecules is greatly enhanced. This phenomenon, known as charge-resonance-enhanced ionization (CRIE) for symmetric molecules [1], has been experimentally observed for various diatomic and polyatomic molecules [2–4]. For theoretical investigation of enhanced ionization, both quantum [5–7] and classical [8,9] methods have been employed with the help of Born–Oppenheimer approximation, keeping the nuclei fixed. The validation of this approximation has been confirmed in Ref. [9]. Detailed theoretical analyses have presented some explanations for enhanced ionization, such as field-induced two-electron dynamics [10], laser-induced avoided crossing mechanism [11] and field-induced barrier suppression model [12].

As the simplest molecules, molecular hydrogen and deuterium are easily amenable to theoretical simulations and provide many of the basic features found in more complex molecules. It is for this reason that they have often been chosen as targets in the study of laser–molecule interactions in the past two decades. So far, most theoretical and experimental researches on enhanced ionization were concerned with hydrogen molecules in parallel alignment. The relationship between enhanced ionization of $H_2$ and laser intensity has also been investigated. In Ref. [7], the enhanced ionization of $H_2$ is more pronounced in the case of weaker fields and the critical distance for ionization of $H_2$ was shown to decrease with intensity increasing. In Ref. [13], the critical distances for both single and double ionization of $H_2$ in parallel alignment were almost independent of the laser intensity.

In this paper, with the three-dimensional (3D) classical ensemble model, we investigate the enhancement of double ionization of $H_2$ perpendicularly aligned with respect to the laser polarization direction with intensity ranging from $1 \times 10^{14}$ W/cm$^2$ to $6 \times 10^{14}$ W/cm$^2$. We find that the double ionization probability of $H_2$ strongly depends on $R$ for all laser intensities considered in this work. The critical distance is about 5 a.u., which is independent of laser intensity. In addition, the enhancement of double ionization is more pronounced in the cases of weaker or stronger fields. Qualitative analysis based on the field-induced barrier suppression model [12] and back analysis reveals that this $R$-dependence enhancement of double ionization depends on the potential well of the second electron and the $R$-dependent rescattering probability of the first ionized electron.

2. Theoretical model and results

The 3D classical ensemble model has been described in detail in Ref. [14]. This model has been employed to study double ionization of atoms or molecules extensively [15–20]. For $H_2$, the electron–nuclear interaction and the electron–electron interaction are represented by a two-center 3D soft-Coulomb potential (in atomic units)

$$V(\vec{r}_1, \vec{r}_2) = - \frac{1}{\sqrt{(\vec{r}_1 - \vec{R}/2)^2 + a^2}} - \frac{1}{\sqrt{(\vec{r}_1 + \vec{R}/2)^2 + a^2}} - \frac{1}{\sqrt{(\vec{r}_2 - \vec{R}/2)^2 + a^2}} - \frac{1}{\sqrt{(\vec{r}_2 + \vec{R}/2)^2 + a^2}} + \frac{1}{\vec{r}_1^2} + \frac{1}{\vec{r}_2^2} + b^2.$$
The evolution of the two electrons is governed by Newton’s equations of motion: $d^2 \mathbf{r}_i/dt^2 = -\nabla V(\mathbf{r}_1, \mathbf{r}_2)$, with $i = 1, 2$ denoting different electrons. The electric field $\mathbf{E}(t)$ is a 800 nm linearly polarized laser pulse with a polarization direction along the z axis and a total duration of ten optical cycles, two-cycle turn on, six cycles at full strength, and two-cycle turn off. The molecule axis is aligned perpendicular to the laser polarization direction. In present calculations, the two nuclei are fixed along the x axis with coordinates being $(-R, 0, 0)$ and $(R, 0, 0)$, respectively. For each $R$, the ground state energy of $H_2$ is obtained from Ref. [21]. Similar to Ref. [16], $b$ is set to 0.05. In order to avoid autoionization and guarantee a stable initial ensemble, $a$ is set to be 1.25. The starting position of the electron pairs is distributed along the x axis using a random probability distribution. We firstly select out the electron pairs whose available kinetic energy is positive, then the position and momentum distributions of the initial ensemble are obtained by the same method as that described in Ref. [18].

Once the initial ensemble is obtained, the laser is turned on and all trajectories are evolved in the combined Coulomb and laser fields. At the end of the laser pulse, we examine the total DI yield. We define that the molecule is doubly ionized if the energies of both electrons are positive and singly ionized (SI) if either electron firstly possesses positive energy.

In Fig. 1, the total DI probability is shown versus the value of $R$ for six different intensities. As seen from Fig. 1, the enhancement of double ionization can be observed in all cases: the total double ionization probabilities increase as $R$ increases and at some critical point $R_c$, a maximum is reached; the probabilities then decrease at larger $R$. There are two intriguing aspects in the results of Fig. 1. Firstly, the critical distance $R_c$ for double ionization probability of perpendicularly aligned $H_2$ is intensity independent, which is around 5 a.u. for all laser intensities. This is the same as Ref. [13] but different from Ref. [7], where the critical distance $R_c$ for ionization rate of $H_2$ in parallel alignment shifted to smaller values of $R$ for increasing field strength. Secondly, the enhancement of double ionization is more pronounced in the cases with small or large field strengths compared with those with intermediate field strengths. When $I$ is $3 \times 10^{14}$ W/cm$^2$ or $4 \times 10^{14}$ W/cm$^2$, after the double ionization probability increases to its maximum value, the double ionization probabilities are almost unchanged in the molecular region ($R>R_c$), resulting in the less pronounced enhancement of double ionization. Note, since DI probability increases rapidly with $I$ increasing, when $I$ is $1 \times 10^{14}$ W/cm$^2$, DI probabilities are much smaller than those with larger intensities, the variation of DI probabilities between different $R$ is not obvious in Fig. 1 and so for the enhancement of double ionization. This condition is also suitable for $R$ dependent SDI probability with the same intensity stated below.

In order to understand the obtained result of Fig. 1 in detail, we trace back all the DI trajectories and classify DI into nonsequential double ionization (NSDI) and sequential double ionization (SDI) based on whether recollisions occur in the DI process. We define recollision if the distance between the two electrons is less than $d=5$ a.u. after the ionization of the first electron. The statistical results change very little when $d$ ranges from 4 to 7 a.u. If recollision occurs, DI is classified into NSDI. Otherwise DI is classified into SDI.

Fig. 2(a) and (b) shows the statistical NSDI and SDI probabilities versus $R$ for different field strengths respectively. In the region of $R>R_c$, both NSDI and SDI probabilities increase as $R$ increases, as a result the total DI probability also increases with $R$ increasing in this region for all laser intensities. By comparison, the increment speed of SDI probability is far larger than that of NSDI probability and both speeds increase with laser intensity increasing. When $R$ is larger than $R_c$, the NSDI probability decreases as $R$ increases for all laser intensities while the variation of SDI probability shows difference for different laser intensities. As seen from Fig. 2(b) and 2(c), when $I$ is small ($I=1 \times 10^{14}$ W/cm$^2$ or $2 \times 10^{14}$ W/cm$^2$), the NSDI contribution for the total DI is larger than that from SDI, and SDI probabilities also decrease with $R$ increasing and so for the total DI probability for certain. As laser intensity increases, the DI process is increasingly
dominated by SDI. When \( I \) is intermediate (\( I = 3 \times 10^{14} \, \text{W/cm}^2 \) or \( 4 \times 10^{14} \, \text{W/cm}^2 \)), SDI probability is insensitive to \( R \). Since the total DI is dominated by SDI, the total DI probabilities also change little with \( R \) increasing, resulting in much less pronounced enhancement of double ionization. When \( I \) is large (\( I = 5 \times 10^{14} \, \text{W/cm}^2 \) or \( 6 \times 10^{14} \, \text{W/cm}^2 \)), the total DI contribution from NSDI is negligible compared with that from SDI, and SDI probabilities decrease again with \( R \) increasing. Consequently, the total DI probabilities also abide the same variation regularity.

Further, we qualitatively discuss the explanation for above \( R \) and \( I \) dependent NSDI, and SDI probabilities. As is known, SDI is a DI process in which two electrons are independently emitted via tunneling ionization while NSDI is resulted from the recollision between the returning electron and the parent ion. Therefore, for a given \( I \) and a fixed \( R \), the SDI probability is determined by the SI probability, the ionization potential of the second electron (\( I_{p2} \)) and the double potential well distorted by the external laser field. Apart from these factors, the NSDI probability is also determined by the rescattering probability and the returning energy of the first ionized electron.

The \( R \) dependent \( I_{p2} \) is shown in Fig. 3(a), which is irrelevant with laser intensity. As \( R \) increases, \( I_{p2} \) decreases monotonically. Fig. 3(b) shows \( R \) dependent SI probabilities for different laser intensities. For intermediate or large laser intensities, SI probability is nearly unchangeable with \( R \) and thus makes negligible influence on NSDI and SDI probabilities in this intensity range. Based on the same statistical criterion as that for NSDI, the rescattering probability of the first ionized electron as a function of \( R \) for different laser intensities is plotted in Fig. 3(c), the variation tendency is monotonically decreased and similar for different laser intensities. For perpendicularly aligned diatomic molecules, the stereo images of the field distorted potential wells (the laser electric field reaches the maximum) with \( I = 3 \times 10^{14} \, \text{W/cm}^2 \) are shown in Fig. 4 for example, in which the \( R = 2, 5, \) and 9 a.u. respectively. When \( R \) is given a fixed value, the potential wells in Fig. 4 are projected into a one-dimensional potential curve in Fig. 5, where the solid green lines correspond to \( x = \pm R/2 \) while the dashed red lines correspond to \( x = 0 \). In Fig. 5(a), two lines are nearly coincident when \( R = 2 \) a.u. By careful comparison, the increase of the outer potential with \( R \) increasing is visible. This variation is the same for other laser intensities but with difference in increase amplitude. It is easy to see that electrons are most probable to ionize by moving through potential well along \( x = \pm R/2 \). As for the returning energies of the first electron, they are similar for different \( R \) with the same laser intensity and thus make negligible influence on recollisions when \( R \) varies. Thus, the NSDI probability is mainly determined by \( I_{p2} \) and the rescattering probability.

The explanation for \( R \) dependent NSDI probability is discussed as a whole for all laser intensities while that for SDI probability is separately discussed for different intensity range. For NSDI, the increase of NSDI probability in the region of \( R < R_c \) is mainly originated from the rapid decrease in \( I_{p2} \). When \( I \) is small, though rescattering probability decreases with \( R \) increasing, the increase of SI probability made the increase in the number of rescattering electrons, which also makes contribution to the increment of NSDI probability. As for \( R > R_c \), the decrease of NSDI probability is mainly because of the increase of the outer potential and the decrease of the rescattering probability.

For SDI probability in the region of \( R < R_c \), the increase of SDI probabilities is also mainly originated from the rapid decrease in \( I_{p2} \). For the region of \( R > R_c \), \( I_{p2} \) decreases slowly to increase SDI probability. 

![Fig. 3.](image1) (a) \( R \)-dependent ionization potential of the second electron, (b) \( R \)-dependent SI probability, and (c) \( R \)-dependent rescattering probability of the first ionized electron. The laser intensity for every curve in (b) and (c) is in correspondence with Fig. 1 through line style.

![Fig. 4.](image2) The stereo images of the field distorted potential wells (the laser electric field reaches the maximum) with \( I = 3 \times 10^{14} \, \text{W/cm}^2 \), the polarization of the laser field is along the z axis. (a) \( R = 2 \) a.u., (b) \( R = 5 \) a.u., and (c) \( R = 9 \) a.u.
while the outer potential increases slowly to decrease this probability. The comprehensive effect of these two factors is the origin of the difference in $R$ dependent SDI probabilities for different laser intensities. When $I$ is small or large, the increase in the outer potential plays a major role and thus results in the decrease in SDI probability. When $I$ is intermediate, the roles of these two factors are almost equivalent and thus the SDI probability is insensitive to $R$ for these laser intensities.

3. Conclusion

In summary, we have theoretically investigated the laser intensity-dependence of enhanced double ionization of perpendicularly aligned $H_2$ molecules by intense linearly polarized laser fields with the classical 3D ensemble model. The enhancement of double ionization is visible for all laser intensities but more pronounced in the cases of weaker or stronger fields. The critical distance for double ionization of $H_2$ is about 5 a.u., which is independent of laser intensity. These results provide an insight into the important role of the structure of diatomic molecules and laser–molecule interactions in double ionization.

Acknowledgements

This work was supported by the National Natural Science Foundation of China under Grant No. 10774054, the National Science Fund for Distinguished Young Scholars under Grant No. 60925021, and the 973 Program of China under Grant No. 2006CB806006.

References


Fig. 5. One dimensional potential curve when x in Fig. 4 is set to a fixed value. (a) $R=2$ a.u., (b) $R=5$ a.u., and (c) $R=9$ a.u. Solid green lines: $x=\pm R/2$, dashed red lines: $x=0$. (When $R$ is 2 a.u., two lines are nearly coincident.)