Double ionization of molecule $H_2$ in intense ultrashort laser fields

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Abstract. By solving numerically the time-dependent Schrödinger equation (TDSE), we have calculated the double ionization probability when a vibrating hydrogen molecule interacts with intense ultrashort laser pulses. The results show that in the case of vibrating nuclei the double ionization probability is higher than that of the fixed nuclei. Additionally, the double ionization probability is larger if the molecule is vibrating in a higher level. This is due to the decreasing of ionization potential when the inter-nuclei separation increases.

1. Introduction
Since intense ultrashort laser fields were produced, interaction between them and atoms, molecules has attracted much interests. When an atom or molecule is exposed to an intense laser field, several non-linear phenomena may occur, e.g. ionization, high-order harmonic generation. Single ionization has been studied for one-electron atoms [1, 2] but with many-electron atoms or molecules effects due to the other electrons or molecular structure need to be involved.

With many-electron atoms, ionization process becomes more complex. In this paper, we focus on double ionization of hydrogen molecule. There are two mechanisms: sequential double ionization and non-sequential double ionization. Sequential double ionization is a process of formation of doubly-charged ions consisting of two single-electron ionization events: the first electron is removed from a neutral atom or molecule followed by detachment of the second electron from the ion. Non-sequential double ionization is a process whose mechanism differs from the sequential one. For example, both the electrons leave the system simultaneously or the second electron's liberation is assisted by the first electron [3, 4].

Investigation of double ionization has been carried out widely with many different methods. In [5], Chen et al. calculated momentum distribution of recoil ions He$^{2+}$ from laser-induced non-sequential double ionization of helium by a semi-classical rescattering model. Moreover, Kopold et al. also developed a method for calculation of the S matrix applied to the distribution for the total electronic momentum. They claimed that the result well agreed with measurement for neon but there was no comparable agreement for helium and argon [6]. In [7] Winger transformation is used to study the electronic center-of-mass motion in phase space for double ionization in a strong laser field. Continuously, in 2006 Parker et al. measured electron energy distributions of singly and doubly ionized helium in an intense 390 nm laser field at two intensities 0.8 PW/cm$^2$ and 1.1 PW/cm$^2$. Authors also confirmed that their numerical solution from solving the time-dependent Schrödinger equation (TDSE) was consistent with measurement [8]. In addition, in [9] Denys...
I. Bondar et al. developed an analytical method for correlated two-electron in strong infrared laser fields. Authors pointed out that if two electrons are ejected nearly simultaneously, they are driven by the laser field in the same direction. If two electron are ejected with a substantial delay, the quarter-cycle or more, they end up going in the opposite directions.

Double ionization of H$_2$, the simplest molecule has been studied, is researched by many groups [10–13]. In [10] Volkova et al. calculated the ratio of single and double ionization of the hydrogen molecule to find out the mechanism of double ionization including tunneling and multiphoton. Furthermore, Sagout et al. set up experiment to measure and calculated the proton spectrum as a function of the pulse laser, the carrier-envelope and the intensity [11]. The dependence of double ionization yield of H$_2$ on carrier-envelope phase was also reported in [12]. Li Yan et al. in addition, studied probabilities of double ionization of H$_2$ for parallel and perpendicular alignment. They concluded that the probability is higher if the molecule is aligned parallel [13].

However, in above mentioned papers, calculations are only carried out with frozen nuclei. Nuclear vibration, in our point of view, should be taken into account. Here, we solve numerically the TDSE to investigate the influence of vibrational levels on double ionization probabilities with different laser parameters. The rest of the paper is arranged as follows. In Sec. 2, we introduce our results of calculation for the probabilities with fixed and vibrating nuclei. The effects of different vibration levels are also analysed in this section. Sec. 4 is the conclusions.

2. Model and basic formalism

In this work, one-dimensional model H$_2$ is used and we solve numerically the TDSE to obtain the double ionization probability. Atomic units are used throughout the paper unless stated.

The TDSE for a H$_2$ exposed to an intense laser field can be written as

$$ i \frac{\partial}{\partial t} \Psi(x_1, x_2, R, t) = \left( -\frac{\partial^2}{2\partial x_1^2} - \frac{\partial^2}{2\partial x_2^2} - \frac{\partial^2}{2\mu \partial R^2} + V_C(x_1, x_2 R) + V_L(x_1, x_2, t) \right) \Psi(x_1, x_2, R, t) $$

where $x_1, x_2$ are electrons coordinate with respect to nuclear center of mass; $R$ is the internuclear separation; $\mu$ is the reduced mass of two nuclei; $V_C(x_1, x_2, R)$ is a soft-core Coulomb potential and $V_L(x_1, x_2, t)$ is the laser-induced potential. In this model, the soft-core Coulomb potential in equation (1) can be written as

$$ V_C(x_1, x_2, R) = \frac{1}{\sqrt{(x_1 - \frac{R}{2})^2 + a}} - \frac{1}{\sqrt{(x_2 + \frac{R}{2})^2 + a}} - \frac{1}{\sqrt{(x_1 - x_2)^2 + a}} - \frac{1}{\sqrt{(x_2 - x_1)^2 + a}} $$

(2)

Constant $a$ called soft-core parameter is added to avoid the singularity of the Coulomb potential and to mimic the real potential energy curve (PEC) of H$_2$. In this paper $a$ is considered to depend parametrically in the internuclear separation. The laser-induced potential is given by

$$ V_L(x_1, x_2, t) = (x_1 + x_2)E_0 f(t) \cos(\omega_0 t) $$

(3)

where $E_0, f(t), \omega_0$ are the electric field amplitude, envelop function and frequency of lase pulse, respectively. The solution for equation (1) can be written as follows

$$ \Psi(x_1, x_2, R, t) = \exp \left[ -i \int_0^t \hat{H} dt \right] \Psi(x_1, x_2, R, 0) $$

(4)
where $\Psi(x_1, x_2, R, 0)$ is the initial wave function which is given by the imaginary relaxation method. We also employ the Born-Oppenheimer approximation to obtain the wave function of the molecule. The split operator is used to obtain the time-dependent wave function in (4) [14]. With the time-dependent wave function, we calculate the probability of single and double ionization by

$$P(V, t) = \int_V |\Psi(x_1, x_2, R, t)|^2 dx_1 dx_2 dR.$$  \hspace{1cm} (5)

where $V$ is the integral region. Double ionization occurs in the asymptotic region $V_2 \equiv \{|x_1| \geq x_a, |x_2| \geq x_a\}$. Single ionization region is defined as $V_1 \equiv \{|x_1| < x_a, |x_2| > x_a \cap |x_2| < x_a, |x_1| > x_a\}$, and the neutral molecule is found in $V_0 \equiv \{|x_1|, |x_2| \leq x_a\}$ with $x_a = 20 \text{a.u.}$

3. Results

In this section, we show calculation results of the single and double ionization of $\text{H}_2$ in intense laser fields. First, nuclei are fixed to investigate the dependence of probabilities on laser parameters such as intensity and pulse duration. In Fig.1 the single and double ionization probability of $\text{H}_2$ in a laser field with wave length of 1200nm, intensity of $3 \times 10^{14} \text{W/cm}^2$ and 10-cycle pulse is demonstrated. One can see that from 1st to 3rd cycle, single ionization probability is negligible, from 3rd to 7th the probability increases rapidly and is followed by a plateau region after 7th cycle. In this case, the maximum probability is 36.6%. The double ionization probability also has the similar performance with smaller values. In Fig.1 the double ionization only can be considered to occur from 5th cycle and the maximum value 7.6% is, five times smaller than the single ionization probability.

![Figure 1](image-url)

**Figure 1.** The single ionization (solid line) and double ionization probability (dashed line) of $\text{H}_2$ in a laser field with peak intensity of $3 \times 10^{14} \text{W/cm}^2$, wave length of 1200nm and duration of 40fs (10 cycles).

Thus, single ionization may be considered to occur from 3rd cycle while double ionization happens at 5th cycle. Both processes reach the maximum value at 7th cycle which is followed by a plateau region.

Those results can be explained by using the three-step model. After tunneling, the 1st electron will be accelerated by electric field. When the electric field reverses, the electron will be pulled back and collide with the 2nd electron that leads to double ionization. That is the reason why the double ionization happens nearly two cycles later than single ionization.
In Fig.1, there are some hollows in the curves of ionization probabilities. That is due to being driven back of electrons into neutral molecule region making the ionization probabilities smaller. When releasing electrons balances with returning, the probabilities reach the saturated value.

Next, we calculate the double ionization probability of \( H_2 \) with vibrating nuclei. The results show that the probability of a \( H_2 \) with vibrating nuclei is higher than that with fixed ones. In Fig.2, we plot the probabilities of \( H_2 \) exposed to a laser field with peak intensity of \( 2 \times 10^{14} \text{W/cm}^2 \), wave length of 1200nm and duration of 16fs.

![Figure 2. Double ionization probability of fixed nuclei \( H_2 \) (solid line) and \( H_2 \) in vibrational ground state (dashed line) in a laser field with peak intensity of \( 2 \times 10^{14} \text{W/cm}^2 \), wave length of 1200nm and duration of 16fs (4 cycles).]

With the vibrating molecule, the probability also has a similar behaviour to that of the fixed one but about 1.85 times higher amplitude, i.e., 0.37% and 0.2% respectively. These results may be explained by using terms of ionization potential energy. The energy of a vibrating molecule is always higher than that of a frozen one so the ionization potential is lower. We continue to calculate the probabilities with different laser’s parameters and we also obtain the similar results.

Finally, we investigate the effects of vibration levels on the double ionization probabilities. The vibration levels are limited from \( \nu = 0 \) to \( \nu = 5 \). In Fig.3, we show the double ionization probabilities of a vibrating \( H_2 \) in different levels exposed to a laser pulse with peak intensity of \( 2 \times 10^{14} \text{W/cm}^2 \), wave length of 1200 nm and duration of 16fs.

In Fig. 3, one can see that the probability from a higher vibration level is larger than that from a lower one. The reason is because the ionization potential of the molecule in a lower vibration level is larger than that quantity of a higher one. That leads to a smaller probability to tunnel of electrons. We continue to calculate with different laser’s parameters, the similar phenomena are also obtained. Thus, we conclude that double ionizing atoms in higher vibration levels occurs with larger probabilities.

4. Conclusions

In this paper, we solve numerically the TDSE for 1D \( H_2 \) in intense laser fields to calculate double ionization probabilities. The results point out that the probabilities of the vibrating molecule is always higher than that of a fixed one. In addition, if the molecule is in a higher vibration level, the probability is larger. These results can be explained by using terms of the ionization potential. In next works, extending the method for 2D or 3D molecules need to be investigated.
Figure 3. Double ionization probabilities of H$_2$ with different vibration levels. The laser field with peak intensity of $2 \times 10^{14}$W/cm$^2$, wave length of 1200 nm and duration of 16fs is used.

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