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# Intensity dependence of the $H_2^+$ ionization rates in Ti:sapphire laser fields above the Coulomb-explosion threshold

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Ionization rates of the hydrogen molecular ion $H_2^+$ under linearly polarized pulse of intense laser fields are simulated by direct solution of the fixed-nuclei time-dependent Schrödinger equation for the Ti:sapphire laser lines $\lambda = 790$ and 800 nm at high intensities starting from just above the Coulomb explosion threshold (i.e., $6.0 \times 10^{13}$, $1.0 \times 10^{14}$, $3.2 \times 10^{14}$, and $1.4 \times 10^{15}$ W cm$^{-2}$). Results obtained in this research exhibit a high degree of complexity for the *R*-dependent enhanced ionization rates for the $H_2^+$ system in these intense laser fields. The *R*-dependent ionization peaks move towards small internuclear distances and their structure becomes simpler and smoother with the increase in the intensity of the laser pulse, i.e., with the decrease in the Keldysh parameter. Results obtained in this research are comparable to and even more reliable than the results of other theoretical calculations reported recently and successfully simulate the experimental ionization data.

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## I. INTRODUCTION

In the past decade, technology of the production of ultrashort laser pulses has been extensively developed and ultrashort high power laser pulses with durations about a few optical cycles are now available to extend the science of atomic, molecular, and optical physics [1]. Hydrogen atom and hydrogen molecular ion are two fundamental and prototypical systems which can be used to understand and extend these fields of physics. In the intense laser field, these two apparently simple systems exhibit various complex phenomena [2–4]. An interesting and complicated effect is the enhancement of the ionization rates of $H_2^+$ as a function of internuclear separation which results in maxima at some critical separations. Indirect [5,6] and direct [7] evidence has been observed for this behavior in more complex molecules. Following extensive experimental and theoretical evidence [8–16] for the existence of critical internuclear distances larger than the equilibrium separation at which ionization of the molecule is strongly enhanced, recent theoretical studies have concentrated on the calculation of the ionization rates as a function of internuclear distance *R* [17–25]. Alternatively, the experimental energy spectra of the fragment species have been used to derive the *R*-dependent ionization rates [14–16].

In this study, ionization of the hydrogen molecular ion $H_2^+$ under intense linearly polarized pulse of laser fields is simulated by direct solution of the fixed-nuclei time-dependent Schrödinger equation for the Ti:sapphire $\lambda = 790$ and 800 nm lines at four intensities from just above the Coulomb explosion threshold ($5.0 \times 10^{13}$) up to the intensity $\sim$23 times higher, i.e., $6.0 \times 10^{13}$, $1.0 \times 10^{14}$, $3.2 \times 10^{14}$, and $1.4 \times 10^{15}$ W cm$^{-2}$. The Keldysh parameter [26], $\gamma = \sqrt{|E_0|/2U_p}$, for these laser pulses with $\lambda = 800$ nm are calculated and listed in Table I. This parameter is a useful index to determine the effects of different combinations of frequency and field strength on the ionization rate spectra and to determine which regime a particular pulse and system may belong to. In this parameter, $E_0$ is the binding energy of the electron to the atomic or molecular system and $U_p = (E/2\omega)^2$ is the pondermotive energy in atomic units, where $E$ and $\omega$ are, respectively, the electric-field strength and the angular frequency of laser field, both in atomic units. The $\gamma \ll 1$ and $\gamma \gg 1$ ranges correspond, respectively, to the tunneling and the multiphoton regimes, while the $\gamma \approx 1$ range is considered as an intermediate regime. Therefore, for the $H_2^+$ system and the Ti:sapphire laser pulse, the intensity $6.0 \times 10^{13}$ W cm$^{-2}$ belongs to the multiphoton regime, the intensity $1.0 \times 10^{14}$ W cm$^{-2}$ belongs to the intermediate regime, and finally intensities $3.2 \times 10^{14}$ and $1.4 \times 10^{15}$ W cm$^{-2}$ belong to the tunneling regime. The magnitude of the Keldysh parameter is decreased with increasing intensity of the laser field, and thus it is expected that the complexity of the structure of the ionization rates be reduced by increasing intensity.

In the rest of this report, at first, the numerical method used for the modeling of the $H_2^+$ system under laser field is introduced and the details of simulations are presented. Next, the results obtained at various intensities of the laser field are presented and compared with the results of other calculations reported recently and the available experimental data. Finally, the conclusions of this research are presented. Unless otherwise stated, atomic units have been used throughout.

## II. NUMERICAL SOLUTION OF THE TDSE

The time-dependent Schrödinger equation (TDSE) in the cylindrical polar coordinate system for the $H_2^+$ molecular ion located in the laser field of $E(t)$ parallel to the $z$ axis (internuclear axis) reads as

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TABLE I. Values of Keldysh parameter, $\gamma=\sqrt{|E_0|/2U_p}$, calculated for $H_2^+$ molecular ion in an intense laser field with $\lambda=800$ nm. $E_0$ is the binding energy of the electron in the $H_2^+$ $1s\sigma_g^+$ orbital.

| $R$ (a.u.) | $I$ (W cm$^{-2}$) | | | |
|---|---|---|---|---|
| | $6.0\times10^{13}$ | $1\times10^{14}$ | $3.2\times10^{14}$ | $1.4\times10^{15}$ |
| 2 | 1.51 | 1.17 | 0.65 | 0.31 |
| 4 | 1.44 | 1.12 | 0.62 | 0.30 |
| 6 | 1.39 | 1.08 | 0.60 | 0.29 |
| 8 | 1.38 | 1.07 | 0.60 | 0.29 |
| 14 | 1.38 | 1.07 | 0.60 | 0.29 |

$$i\frac{\partial\psi(z,\rho,t)}{\partial t} = H(z,\rho,t)\psi(z,\rho,t), \tag{1}$$

where the total three-dimensional (3D) electronic Hamiltonian is given by [27,28]

$$H(z,\rho,t) = -\frac{2m_p+m_e}{4m_pm_e}\left[\frac{\partial^2}{\partial\rho^2}+\frac{1}{\rho}\frac{\partial}{\partial\rho}+\frac{\partial^2}{\partial z^2}\right]+V_C(z,\rho,t) \tag{2}$$

in which

$$V_C(z,\rho,t) = \frac{-1}{[(z\pm R/2)^2+\rho^2]^{1/2}} + \left(\frac{2m_p+2m_e}{2m_p+m_e}\right)zE_0f(t)\cos(\omega t), \tag{3}$$

where $E_0$ is the laser peak amplitude, $\omega=2\pi\upsilon$ is the angular frequency, and $f(t)$ is the laser pulse envelope which is set as

$$f(t) = \begin{cases} \frac{1}{2}\left[1-\cos\left(\frac{\pi t}{\tau_1}\right)\right] & \text{for } 0\leqslant t\leqslant\tau_1 \\ 1 & \text{for } \tau_1\leqslant t\leqslant\tau_1+\tau_2 \end{cases}, \tag{4}$$

with $\tau_1$ and $\tau_2$ being the rising time and duration of the laser pulse at its full-scale amplitude, respectively.

To solve the above TDSE numerically, we adopted a general nonlinear coordinate transformation. The differentiation operators in Eq. (2) are discretized by 11-point difference formulas which have tenth-order accuracies. For the spatial discretization, we have constructed a finite difference scheme with a nonuniform (adaptive) grid which is finest near the nuclei and coarsest at the border regions of the simulation box. The use of a fine grid near the nuclei improves the treatment of the wave function near the origins (the Coulomb singularities), while the use of a coarse grid near the borders improves the speed of calculations. For the time discretization of the TDSE, a propagator derived from the split-operator method has been used. The ionization rate $\Gamma$ is obtained by calculating the time-dependent norm of the wave function, i.e.,

$$N(t) = \|\Psi(t)\|^2 = \|\Psi(0)\|^2\exp(-\Gamma t), \quad \text{with}$$

$$\Gamma = \frac{-d\ln[N(t)/\|\Psi(0)\|^2]}{dt}. \tag{5}$$

Absorber regions are introduced by using fourth-order optical potentials at the $z$ and $\rho$ boundaries, in order to capture the photoelectrons and prevent the reflection of the outgoing wave packets at the borders of the grid. More details of our calculations are described in our previous report [25].

## III. RESULTS AND DISCUSSION

Attempts to directly measure the ionization rate of $H_2^+$ as a function of $R$ [14–16,29] has been recently reported. For interpretation of the experimental ionization rates data, one must take into account a number of parameters. If the duration of irradiation is long and the intensity of laser field is sufficiently low (close to the Coulomb channel threshold, $0.5\times10^{14}$ W cm$^{-2}$ for the $H_2^+$), the initial distribution pattern of the system over the vibrational states will be altered during the laser pulse. When the duration of laser pulse is long and the intensity is relatively higher than the Coulomb channel threshold, the system will be ionized during all cycles of the laser pulse and the nuclear dynamics and dissociation both change the population of different vibrational states. In this situation, the ionization signal depends on the nuclear dynamics as well as on the ionization rates. The vibrational levels, as observed in the dissociation channel [30], are simply shifted by the Coulomb repulsion energy. The ionization signal thus maps two features: the observed vibrational states in the dissociation channel and the ionization rates. In other words, in this situation we can image the dissociation channel by analyzing the peaks of ionization signal which are sufficiently intense and broad.

When the duration of ionization is long (the laser field intensity is low), the system can dissociate and evolve to larger $R$ prior to ionization. Therefore, the ionization signal for large $R$ can be detected. In this condition, the nuclear dynamics can change the population of the system over $R$ variable. When the duration of ionization is short (the laser field intensity is relatively high), the motion of nuclei are slow during the course of ionization process and the system has a little chance to evolve to large values of $R$, and therefore the ionization signal for larger values of $R$ will be weak or even null. It can be concluded that the ionization rate and initial distribution of the system over vibrational states determine the $R$-dependent structure of the ionization signal. In contrast, if the laser field is very intense, the system will be ionized even during the first half cycle of the laser pulse and thus the nuclear dynamics can be totally neglected [31]. This implies that the distribution pattern of the system over the vibrational states will not be altered during the course of the ionization process and it is possible to determine the initial nuclear distribution of the system by determining the $R$-dependent ionization signal. In this condition, the ionization rates are high and the ionization signal depends only on the initial or instantaneous distribution of vibrational state of the system and therefore the ionization signal maps the initial or instantaneous distribution of the system over vibrational states as happens in the Coulomb explosion imaging spectroscopy [31,32].

When the $H_2^+$ molecular ion is exposed to very intense femtosecond laser pulses, two channels of fragmentation are

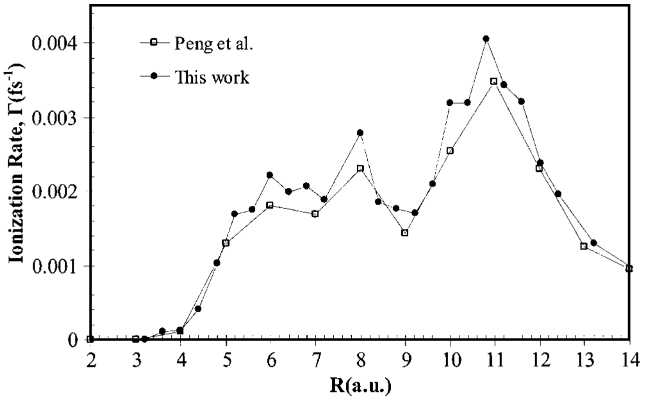

FIG. 1. The ionization rates Γ of $H_2^+$ as functions of $R$ in the linearly polarized field of $I=6.0\times10^{13}$-W/cm$^2$ intensity and $\lambda=790$ nm wavelength calculated in this work (●) compared with the calculated ionization rates reported by Peng *et al.* (□) [23].

observed [29]. At lower intensities up to $5\times10^{13}$ W cm$^{-2}$, only dissociation into H atoms and $H^+$ (protons) with kinetic energies up to 0.8 eV per fragment occurs. At intensities higher than $5\times10^{13}$ W cm$^{-2}$, the Coulomb explosion channel is opened, producing two protons, with translational energies between 1 and 3.5 eV, and an electron as fragments [29].

The $R$-dependent ionization rate of $H_2^+$ obtained from numerical solution of TDSE for the laser pulse with the wavelength $\lambda=790$ nm and the intensity $I=6.0\times10^{13}$ W cm$^{-2}$, which is just above the Coulomb explosion threshold (at $I=0.5\times10^{14}$ W cm$^{-2}$) is shown in Fig. 1. Our results (●) obtained with the grid size of (170, 64) for the $(z,\rho)$ coordinates, are very good comparable with Peng *et al.*'s results (□) which are obtained via a different approach of the direct solution of the fixed-nuclei TDSE. Our calculations predict ionization rates which are generally higher than those reported by Peng *et al.* However, there is a good correspondence between the two sets of results; both present several peaks at $R\sim6.0$, 6.8, 8, and 10.8. It is evident from Fig. 1 that our results show more details of the $R$-dependent ionization rates. Note that the inner baseline peak, spanning over $R\sim5.2$–8.4, is broad. The higher (outer) baseline peak centered at $R\sim10.8$ is very important in the Coulomb explosion kinetic energy spectra of $H_2^+$. Bandrauk and Lu [33] reported similar calculations for a linearly polarized laser field of $I=1\times10^{14}$ W/cm$^2$ intensity, but with $\lambda=800$ nm wavelength and $\tau_1=5$ cycle rising time.

The calculated ionization rates of $H_2^+$ in a laser field with wavelength $\lambda=790$ nm and intensity $I=1.0\times10^{14}$ W cm$^{-2}$ are presented in Fig. 2. Results of these calculations (●), which are carried out with the grid size (200, 64) for the $(z,\rho)$ coordinates (AG6 in Ref. [25]), exhibit a structure for the inner baseline peak that is a feature of the potential barrier. Results obtained by Peng *et al.* (□) exhibit more or less the same structure. In the $R$-dependent window of this baseline peak, several peaks at $R\sim4.4$, 5.2 and at $R\sim6.8$ (a strong peak) can be observed. The outer baseline peak that is the charge resonance peak [9,21] is similar for both $6.0\times10^{13}$ and $1.0\times10^{14}$ W cm$^{-2}$ intensities. The system and laser pulses for both intensities have Keldysh parameters larger or close to 1 (Table I). Therefore it is naturally expected that their corresponding inner baseline peaks be structured. A comparison between Figs. 1 and 2 shows that the ionization peaks obtained with the $1.0\times10^{14}$ W cm$^{-2}$ intensity are shifted downward by 1.2 as compared to those obtained with the $6.0\times10^{13}$ W cm$^{-2}$ intensity. Furthermore, the ionization rates at the higher intensity are almost four to five times larger.

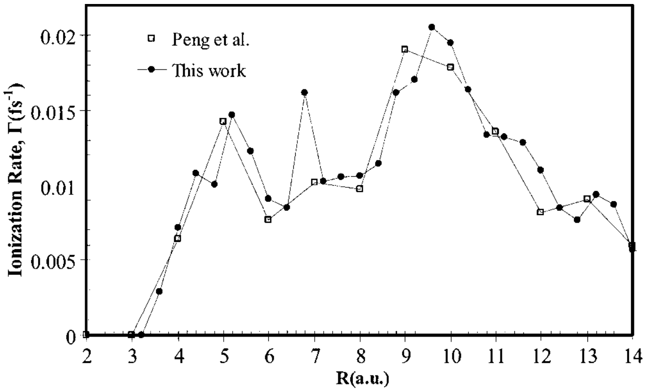

FIG. 2. The ionization rates Γ of $H_2^+$ as functions of $R$ in the linearly polarized field of $I=1.0\times10^{14}$-W/cm$^2$ intensity and $\lambda=790$ nm wavelength calculated in our previous work (●) [25] compared with the calculated ionization rates reported by Peng *et al.* (□) [23].

In 1997, Gibson *et al.* succeeded to obtain experimentally the ionization signal of $H_2^+$ as a function of internuclear distance [14], and thus made a direct comparison with and the evaluation of the theoretical results possible. They performed their experiment with 40-fs (full width at half maximum of a Gaussian envelope), 0.5-mJ laser pulses operating at 1-kHz repetition rate [14] and derived valuable ionization signals as functions of internuclear separation for $\lambda=800$ nm at two laser intensities $I=3.2\times10^{14}$ W cm$^{-2}$ and $1.4\times10^{15}$ W cm$^{-2}$. Peng *et al.* made an attempt to compare the theoretical calculations with the experimental ionization signals reported by Gibson *et al.* and Williams *et al.* [23,24]. Here, in this work, we present our computational results compared with previous experimental and theoretical calculations.

Figure 3 compares our theoretical calculations of the $H_2^+$ ionization rates obtained with the grid size (140, 64) for the $(z,\rho)$ coordinates for a laser pulse of $\lambda=800$ nm wavelength and $I=3.2\times10^{14}$ W cm$^{-2}$ intensity, ●, with Peng *et al.* results [23], □, and the experimental ionization signal obtained by Gibson *et al.* [14], ○, which are scaled to the theoretical data. At high intensity, it is expected that the ionization signal can relatively map the ionization rates for small $R$ only. As found previously in the comparison of Figs. 1 and 2, increasing intensity has resulted in a shift of the ionization peaks towards smaller values of $R$.

It can be seen from Fig. 3 that for small nuclear distances up to $R\sim10$, there is a good agreement between the two sets of theoretical calculations and they are nearly compatible with the scaled experimental ionization signal.

The same set of calculations has been carried out for the laser pulse with $\lambda=800$ nm at intensity $I$

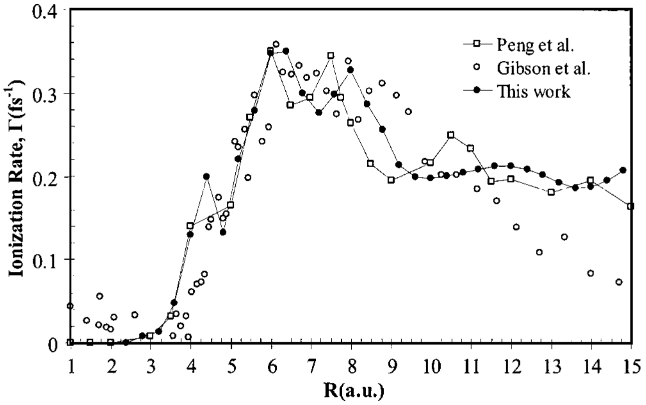

FIG. 3. The ionization rates calculated in this work with a (140, 64) grid size for $(z,\rho)$ coordinates, ●, compared with the experimental results of Gibson *et al.* [14], ○, at $\lambda = 800$ nm and $I = 3.2\times10^{14}$ W/cm$^2$ scaled to the theoretical results. Peng *et al.*'s theoretical results, □, have been given for comparison [23].

$= 3.2\times10^{14}$ W cm$^{-2}$ (as presented in Fig. 3) but with the grid size (740, 120) for the $(z,\rho)$ coordinates. The experimental data of Gibson *et al.* were rescaled to the calculated ionization rates and the results are presented in Fig. 4. By comparing Figs. 3 and 4, we can see that with the increase in the grid size, the calculated ionization rates of $H_2^+$ decrease considerably. Both Figs. 3 and 4(a) show that the experimental data are shifted with respect to the theoretical calculations towards larger values of $R$. Such a comparative trend has also been observed for other theoretical and experimental results [23,24].

A better correspondence between theoretical and experimental ionization data is presented in Fig. 4(b) in which we have not subtracted the average kinetic energy of the dissociation components (which is known to be 0.5 eV [14]) from the kinetic energy of the Coulomb explosion components in the derivation of the experimental ionization signal of Gibson *et al.* The motif of this treatment is that the dissociation kinetic energy is $R$ dependent. We think that its value for small values of $R$ is smaller and becomes larger with $R$. This is while in the subtraction of 0.5 eV, it is assumed that this kinetic energy is independent of $R$ as is done by Gibson *et al.* Behavior of the ionization signal at large $R$ strongly depends on the kinetic energy of dissociation, whereas the kinetic energy of dissociation slightly influences the ionization signal for small $R$. Therefore the $R$ dependency of the kinetic energy of dissociation should be taken into account in the calculation of the ionization signal from experimental data, especially for large $R$.

Gibson *et al.* obtained another important set of experimental data with a laser pulse having a high intensity $I = 1.4\times10^{15}$ W cm$^{-2}$ at wavelength $\lambda = 800$ nm. In order to reproduce these experimental data by the solution of TDSE, we chose a huge grid size of (1240, 240) for the $(z,\rho)$ coordinates to ensure unrestricted evolution of the electron wave packet. When the hydrogen molecular ion with fixed nuclear distance is exposed to this laser pulse, after a few cycles (about two cycles) in the $\tau_2$ domain, the ionization rates deviate from linear behavior. In this condition, accurate evaluation of the ionization rates by Eq. (5) is difficult.

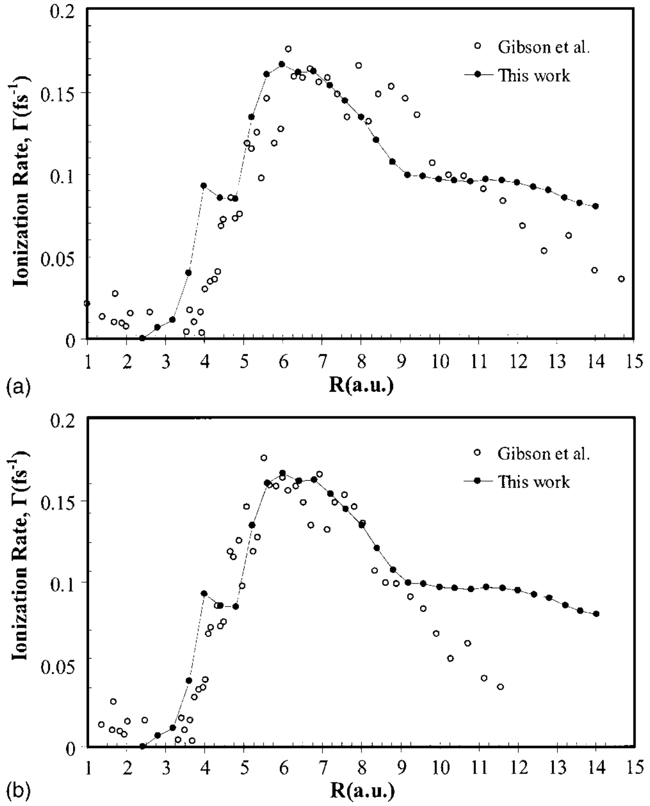

FIG. 4. (a) The ionization rates calculated in this work with a (740, 120) grid size for $(z,\rho)$ coordinates, ●, at $\lambda = 800$ nm compared with the experimental results of Gibson *et al.* [14], ○, at $\lambda = 800$ nm and $I = 3.2\times10^{14}$ W/cm$^2$ scaled to our theoretical results. (b) Same as (a), but without subtracting the average kinetic energy of the dissociation (0.5 eV) from the kinetic energy of the Coulomb explosion components in the derivation of the experimental ionization signal.

We have calculated the ionization rates using the time-dependent system energy defined as

$$E = E_r + \frac{i\Gamma}{2}, \tag{6}$$

where $E_r$ is the real part of the system energy and $\Gamma$ is the ionization rate. The average ionization rate for any time interval is obtained by the integration of $\Gamma$ over that interval. In this manner, it becomes possible to calculate ionization rates accurately.

Using Eq. (6), the ionization rates have been calculated for the time interval between the $\tau_1 + \tau_2 = 5$ and $\tau_1 + \tau_2 = 7$ cycles and compared with the experimental ionization signal in Fig. 5. As expected, there is a good agreement between the two experimental and theoretical data sets at small $R$. However, the present calculations overestimate the ionization signals at large $R$. This is because the hydrogen molecular ion cannot evolve and survive to larger nuclear distances under the experimental condition with very intense laser field. Therefore the large-$R$ portion of ionization signal calculated theoretically is missing in the experimentally measured signal.

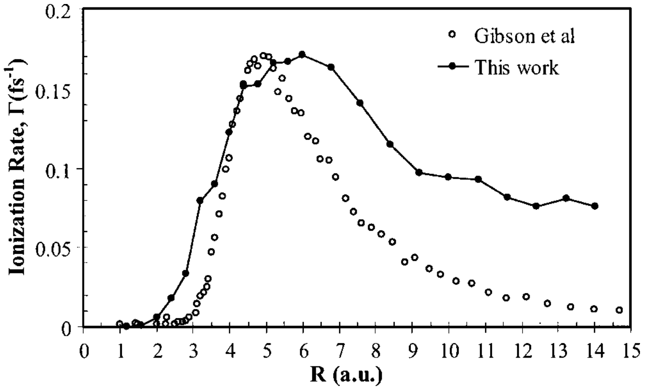

FIG. 5. The ionization rates calculated in this work with a (1240, 240) grid size for $(z,\rho)$ coordinates, ●, compared with the experimental results of Gibson *et al.* [14], ○, at $\lambda = 800$ nm, $I = 1.4 \times 10^{15}$ W/cm$^2$ scaled to the theoretical calculations.

A comparative study shows that the maximum value of the ionization rate is almost unaffected by the increase in the intensity of the laser pulse by going from Fig. 4 to Fig. 5.

## IV. CONCLUSION

The full dimensional electronic TDSE has been solved for the aligned hydrogen molecular ion, $H_2^+$, with fixed nuclei under the Ti:sapphire laser light at $\lambda = 790$ and 800 nm with various intensities, from $I = 6.0 \times 10^{13}$ up to $1.4 \times 10^{15}$ W cm$^{-2}$ using time-efficient adaptive method. This work exhibits the high degree of complexity of the $R$-dependent enhanced ionizations for the $H_2^+$ system in these intense laser fields. For the pulses used in this study, the Keldysh parameter varies from 0.29 up to 1.51, i.e., from the tunneling to the multiphoton regimes. It is found that structure of the enhanced ionization signal for the $H_2^+$ system is shifted to small internuclear distances and becomes simpler and smoother with the decrease in the Keldysh parameter, i.e., with the increase in the intensity of the laser pulse from $I = 6.0 \times 10^{13}$ up to $1.4 \times 10^{15}$ W cm$^{-2}$. The results obtained in this research are in agreement with the theoretical results reported recently in the literature and successfully simulate the experimentally observed ionization signals at short internuclear distances. A study of the intensity dependence of the enhanced ionization carried out previously on a one-dimensional system led to similar regularities [34]. For an accurate and precise prediction of the experimental signals over the full range of $R$, nuclear dynamics (nonfixed $R$) should be included in the time-dependent treatment of the problem. This is an open topic for further studies on this system.

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