J Mol Model (2012) 18:5097–5106
DOI 10.1007/s00894-012-1511-3

ORIGINAL PAPER

Intensity enhancement in the molecular ionization and dissociation dynamics in the presence of noise

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Received: 11 February 2012 / Accepted: 14 June 2012 / Published online: 3 July 2012

Abstract The ionization and the dissociation of the diatomic molecular ion H_2^+ have been investigated within a scheme where a noise field is added to an intense infrared laser field. The results show that both the ionization and the dissociation probabilities are enhanced with the introduction of the additional noise (the Gaussian white noise or the color noise) field. Further, by tuning the noise intensity and the delay time between the laser and the noise, a stochastic resonancelike curve is observed for the ionization or the dissociation dynamics, showing the existence of an optimal noise intensity and delay time for the given laser field.

Keywords Ionization probability · Dissociation probability · Gaussian white noise · Color noise

Introduction

With the rapid developments of the femtosecond (fs) and the attosecond (as) laser technologies, the dynamics of molecules or molecular ions exposed to the ultrashort intense laser pulse has been a subject of great current interest [1–4], especially for that of the simplest two-center one-electron molecular system such as the hydrogen molecular ion (H_2^+). In this area, many interesting phenomena have been revealed, including the bond softening and hardening [5, 6], the electron localization in dissociation channels [7, 8], the above threshold Coulomb explosion (CE) [9, 10], the high-order harmonic generation (HHG) [11, 12], the above-threshold ionization (ATI) and dissociation (ATD) [13, 14] etc. Usually, the ionization probabilities (IPs) and/or the dissociation probabilities (DPs) are critical to gain high signals or high yields for these photochemical and photophysical processes. Up to date, the enhancement of IPs and DPs can be achieved by using very high intense field [15], by the two or three-color field scheme [16], by varying the phase angle between the laser direction and the molecular axis [17], or by using the linearly or circularly polarized chirped laser pulses [18, 19].

Recently, an approach [20–23] which introduces a noise to laser field and is completely different from the above-mentioned ones has been proposed to enhance the photoionization or the photodissociation of molecular systems in laser fields. Most of the relevant investigations have chosen the Gaussian white noise as the added noise field and there are no reports on the color noise source so far. Moreover, all these theoretical investigations have employed Born-Oppenheimer (BO) approximation that separates the slow nuclear motion (fs scale) from the fast electronic motion (as scale) [24, 25]. However, it is hard to get more accurate results when these separated treatments for the electronic and nuclear degrees of freedom are implemented in practice. Therefore, a full-dimensional simulation that simultaneously considers the nuclear and electronic dynamics without BO approximation is preferred for more accuracy. However, as we illuminated in Sec. 2, it is too expensive to apply the full-dimensional calculation to do the investigation in the present paper. Thus, we use physical reasoning and intuition to reduce the dimensionality to a manageable scale that also retains the essential physics of the problem.

Thus, in this paper, we applied a "1+1" dimensional (1+1D) non-Born-Oppenheimer (NBO) model to explore

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simultaneously the nuclear and electronic dynamics and thus the phenomena of molecular ionization and dissociation. This 1+1D model has been tested by others [26–31] and by ourselves [32], showing a qualitatively agreement with the experimental measurements. Using this model, we carried out a detailed investigation when the diatomic molecular ion (H_2^+) is irradiated by an intense infrared laser field in combination with a noise field where either a Gaussian white noise or a color noise is considered.

Theoretical methods

In our calculations, the laser-induced molecular dynamics can be investigated by solving the 1+1D-NBO time-dependent Schrödinger equation (TDSE) [30]. The polarization direction of the laser pulse coincides with the nuclear axis of the molecular ion H_2^+ as well as with the restricted electronic motion. In the dipole approximation and the length gauge, the total molecular Hamiltonian for the diatomic molecular ion under the time-dependent external field is given by (atomic units are used throughout this paper unless stated otherwise.)

$$i\frac{\partial\phi(z,R,t)}{\partial t} = \left[-\frac{1}{2u_e}\frac{\partial^2}{\partial z^2} - \frac{1}{2u_N}\frac{\partial^2}{\partial R^2} + V(z,R) + \kappa zE(t) + \xi RE(t)\right]\phi(z,R,t), \quad (1)$$

where $\kappa=1+1/(m_a+m_b+1)$, $\xi=(m_a-m_b)/(m_a+m_b)$ (here m_a and m_b are the mass of the two nuclei). z (-100 a.u.<z< 100 a.u.) and R (0<R<30 a.u.) are the electronic and the nuclear coordinates, respectively. $u_N=m_am_b/(m_a+m_b)$ and $u_e=(m_a+m_b)/(m_a+m_b+1)$ are the reduced mass of the nucleus and electrons, respectively. $V(z,R) = 1/\sqrt{R^2+a} - 1/\sqrt{(z-R/2)^2+b} - 1/\sqrt{(z+R/2)^2+b}$ is the soft Coulomb potential and a, b (a=0.03 and b=1.0 [33] in this paper) represent the softening parameters to remove the Coulomb singularity [26, 34], the use of which provides an efficient way to obtain realistic numerical results for multiphoton processes

Table 1 A comparison of the computational time between the 1+1D and the 3D models for a single calculation. The laser field is chosen to be 800 nm/10 fs which is used in the present paper (h: hours; m: minutes)

model	ΔR (a.u.)	Δz (a.u.)	Δρ (a.u.)	Δt_e (a.u.)	CPU	T_{CPU}
1+1D	0.1	0.2		0.1	2.4 GHz	35 h 12 m
3D	0.05	0.2	1.0	0.05	2.4 GHz	140 h 36 m

[26–28, 35]. The most important feature of this potential is that at large z it falls off like the true Coulomb potential (a=b=0 is the true Coulomb potential), and it was proved previously that the physical characteristics of this model have allowed realistic investigations of the behavior of a H_2^+ ion in a laser field [26, 29]. But as demonstrated in Ref.[26], this

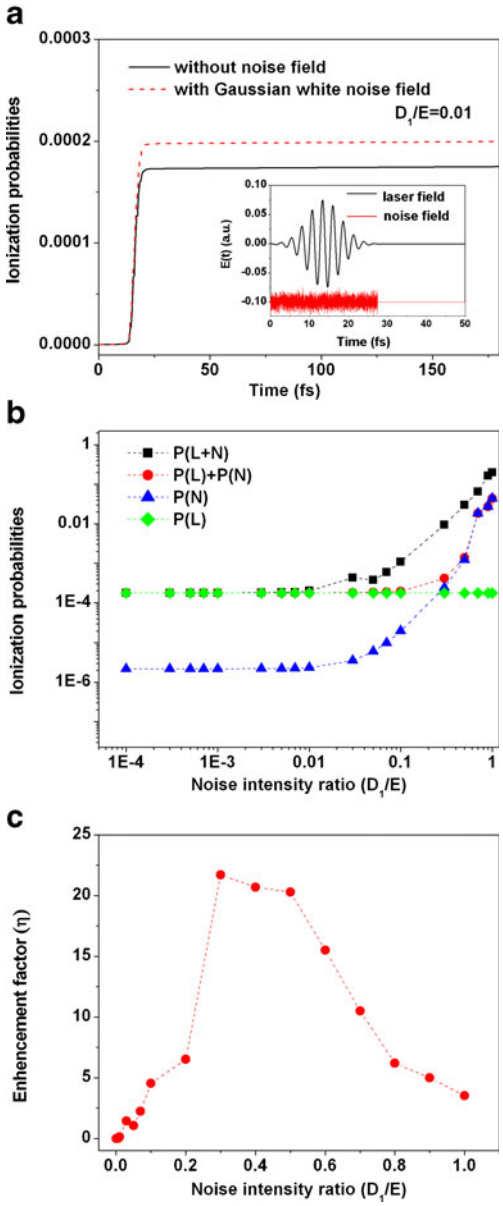

Fig. 1 **a** The IPs of the single laser field (solid black line) and the combined laser-Gaussian white noise field with D_1/E=0.01 (dash red line). The laser field (solid black line) and the noise field (solid red line) are shown in the inset of Fig. 1a. **b** The IPs as a function of the intensity ratio between the Gaussian white noise and the laser field. P(L)−the single laser field, P(N)−the single noise field, P(L)+P(N)−the simple sum of the laser field and noise field, P(L+N)−the combined laser field. **c** The corresponding enhancement factor of the IP

potential model also has some limitations that might influence the molecular dynamics, such as the asymptotic ionization potential (18.24 eV) from the model is higher than the real system (13.6 eV), (ii) the equilibrium distance (2.6 a.u.) is larger than the real system (2.0 a.u.) etc.

The time-dependent wave function $\phi(z,R,t)$can be advanced using the standard second-order split-operator method [36, 37].

$$\phi(t+\delta t) = e^{-iT_R\delta t/2}(e^{-iT_z\delta t/(2N)}e^{-iV\delta t/N}e^{-iT_z\delta t/(2N)})^N e^{-iT_R\delta t/2}\phi(t) + O(\delta t^3), \quad (2)$$

where T_R, T_z and V are the nuclear, the electronic kinetic-energy operators and the interaction potential, respectively. N is the time step ratio between the nucleus and the electron, and N=10 is found to be appropriate for the present investigation. For details of the numerical solution of the time-dependent Schrödinger equation, please refer to [30, 38–41].

The laser field can be expressed as,

$$E(t) = Ee^{-4\ln(2)t^2/\tau^2}\cos(\omega t+\phi) + N(t-\tau_{delay}), \quad (3)$$

where E , ω, τ and φ are the pulse intensity, the frequency, the pulse duration and the carrier-envelope phase (CEP) of the 800 nm fundamental field. N(t) represents the additional noise field, which has the same duration as the 800 nm laser field. τ_{delay} is the delay time between the laser pulse and the noise field. In this paper, two kinds of noises are considered, that is, the Gaussian white noise with the properties of [20–23],

$$\langle N_1(t)\rangle = 0, \quad (4)$$

$$\langle N_1(t)N_1(s)\rangle = 2D_1^2\delta(t-s), \quad (5)$$

where D_1 is the intensity of the Gaussian white noise; and the color noise with the properties of [42]

$$\langle N_2(t)\rangle = 0, \quad (6)$$

$$\langle N_2(t)N_2(s)\rangle = D_2^2\lambda e^{(-\lambda|t-s|)}, \quad (7)$$

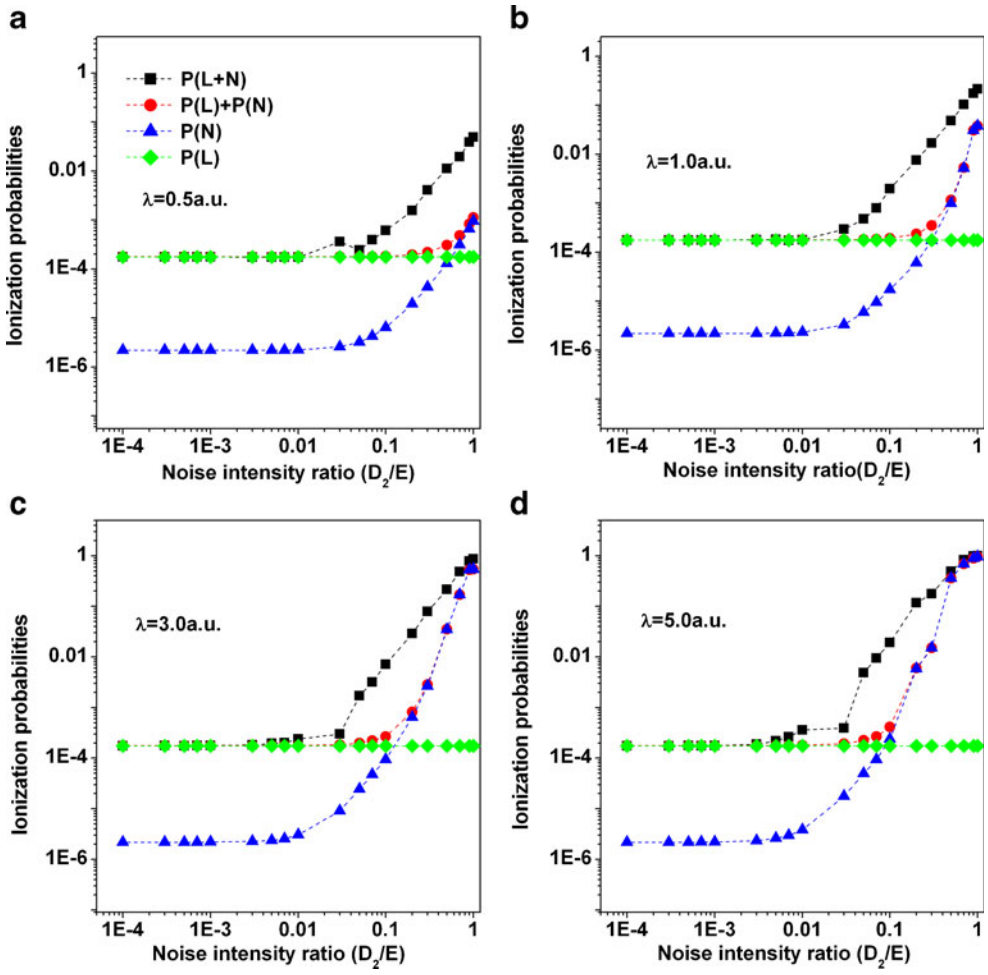

Fig. 2 The IPs as a function of the intensity ratio between the color noise and the laser field **a** for λ=0.5 a.u., **b** for λ=1.0 a.u., **c** for λ=3.0 a.u., and **d** for λ=5.0 a.u

where D_2 and λ are the intensity and the correlation time of the color noise.

The IPs and the DPs are calculated by the flux operator method,

$$P^i(t) = \int_0^t dt' \int_0^{R_s} dRj(R, z_s, t'), \tag{8}$$

$$P^d(t) = \int_0^t dt' \int_{-z_s}^{z_s} dzj(R_s, z, t'), \tag{9}$$

where

$$j = \frac{1}{m_s} \mathrm{Im}[\phi^* \delta(s - s_0) \frac{\partial}{\partial s} \phi], \tag{10}$$

and $m_s=u_N$, s=R, $s_0=R_s$ (R_s=25 a.u. in the present calculation) for dissociation, $m_s=u_e$, s=z, $s_0=z_s$ (z_s=25 a.u. in the present calculation) for ionization.

The enhancement factor, showing the degree of the enhancement during ionization and dissociation, is defined as,

$$\eta = \frac{P(L+N) - (P(L) + P(N))}{(P(L) + P(N))}. \tag{11}$$

Here, the P(L+N), P(L), and P(N) denote the IPs (or the DPs) for the combined laser-noise field, the single laser field, and the single noise field, respectively. We note here that the positive values of this enhancement factor denote the acceleration of the discussed enhancement while the negative values denote the deceleration of the discussed enhancement.

We note that the enhancement phenomena occur under the condition of P(L+N)>P(L), while the enhancement factor defined above is to describe the speed (or the degree) of the enhancement phenomena in relative to P(L)+P(N).

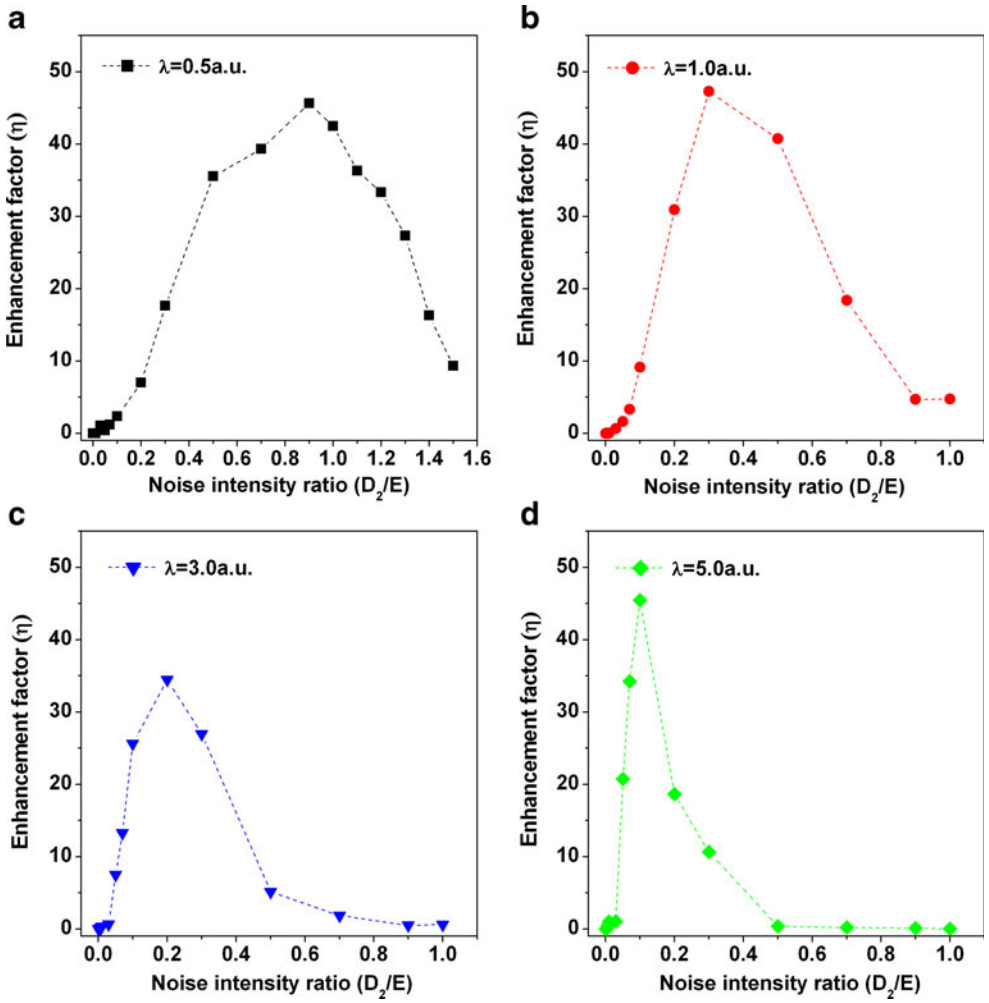

Fig. 3 The enhancement factors of the IPs **a** for λ= 0.5 a.u., **b** for λ=1.0 a.u., **c** for λ=3.0 a.u., and **d** for λ=5.0 a.u

Before we move to the next section, in Table 1, we compare the computational time between the 1+1D model and the 3D model [40, 41] for a single calculation, in order to strengthen the motivation of using the particular-reduced-dimensionality model of 1+1D in the present investigation. The theory for the 3D model can be found in refs [40, 41]. Clearly, for a single calculation, the difference in the computational time is not very large but is still notable. However, when there are lots of conditions to be considered and each of these conditions has averaged over 30-50 different realizations for capturing the statistical feature of the physical quantities, which is just the case of the present study, the difference in total computational time will surely become huge. We estimate that if the 3D model is to be applied, it will cost at least 6-7 months to accomplish the present investigation.

Results and discussion

Enhancement in photoionization

We begin our discussions of the photoionization by first considering the combined laser-Gaussian white noise field. The laser field is chosen to be 800 nm/10 fs (Here, T_{total}=10o.c. with o.c. being the optical cycle of the 800 nm pulse), E=0.0755 a.u., τ_{delay}=0 and $\varphi=0^0$, as shown in the inset of Fig. 1a. Clearly, with the presence of the Gaussian white noise, the resulting IP is enhanced as illustrated by an example case of D_1/E=0.01 in Fig. 1a. The physical origin of this enhancement can be understood by a stochastic resonance two-step model, where the electron is first excited to the excited state by the noise, and it is then ionized from this excited state by the laser field [20, 21]. Figure 1b plotted the IPs as a function of the intensity ratio between the Gaussian white noise field and the laser pulse for the four cases of the combined, the single noise, the single laser fields and the sum over the two single fields. As seen, P(L+N), P(L), P(N) and P(L)+P(N) change very little with the intensity ratio up to D_1/E=0.01, and P(L+N), P(L) and P(L)+P(N) are nearly identical to each other because of the weak contribution from the single noise field over this range (i.e., there is about a factor of two smaller in orders of magnitude of P(N)). However, the continued increase in the intensity ratio can lead to the remarkable enhancement of both P(L+N) and P(N) (and thus P(N)+P(L)) by several orders in magnitude. The enhanced speed is much quicker under the single Gaussian white noise field than the combined field, suggesting that the noise field will gradually play a dominant role in the molecular ionization [22, 23]. Thus, there should have an optimal intensity ratio leading to a maximum ionization

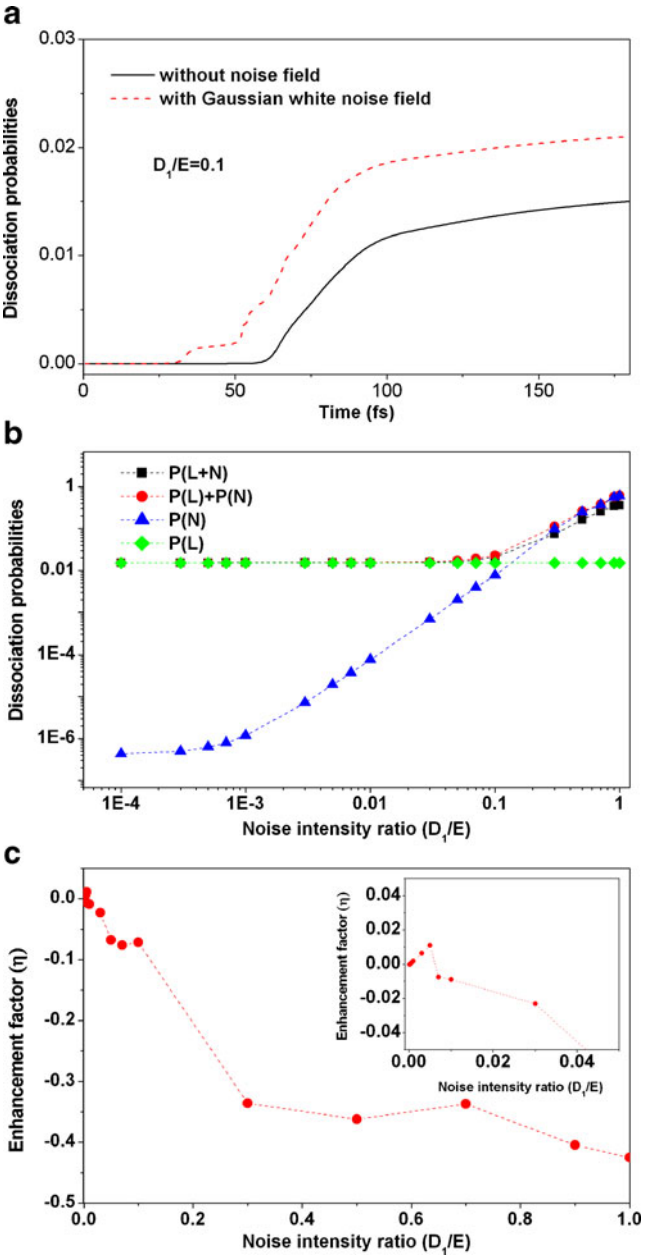

Fig. 4 **a** The DPs of the single laser field (solid black line) and the combined laser-Gaussian white noise field with D_1/E=0.1 (dash red line). **b** The DPs as a function of the intensity ratio between the Gaussian white noise and the laser field. **c** The corresponding enhancement factor of the DP. The inset of Fig. 4c shows a high-resolution figure of Fig. 4c

enhancement. This value can be found in terms of the calculated enhancement factor as a function of the intensity ratio shown in Fig. 1c. The curve starts around zero which indicates the laser field plays a dominant role in the ionization process at the initial stage (P(L)»P(N)). Then, it rapidly increases with a maximum point around $D_1/E=0.3$, and after that it drops down indicating the ionization process is finally dominated by the noise field (P(L)«P(N)).

Figure 2a–d show the IPs as a function of the intensity ratio for the case of adding the color noise source, with the correlation times being 0.5 a.u., 1.0 a.u., 3.0 a.u. and 5.0 a.u., respectively. Similarly, the IPs have been enhanced also, and with the increase in the correlation time, not only the IPs keep enhancing but also the enhanced speed of the single color noise field is accelerated. This means that the color noise field with large correlation time is favorable for dominating the ionization process. From the enhancement factor shown in Fig. 3a–d, we see that the maximum enhancement factor with the color noise field for all correlation times is higher than with the Gaussian white noise field. Moreover, with the increase in the correlation time, the maximum enhancement value moves towards the low intensity ratio, that is, $D_2/E=0.9$, 0.3, 0.2 and 0.1 for the cases of $\lambda=0.5$ a.u., 1.0 a.u., 3.0 a.u. and 5.0 a.u., respectively.

Enhancement in photodissociation

We also begin our discussions of the photodissociation from the combined laser-Gaussian white noise field, and the calculated DP, shown in Fig. 4a, is for a special case of $D_1/E=0.1$. For comparison, DP without the noise field is also presented in this figure. Clearly, with the introduction of the Gaussian white noise, not only the resulting DP is enhanced, but also the initial dissociation time is reduced (changing from the previous 58 fs to the present 31 fs). This

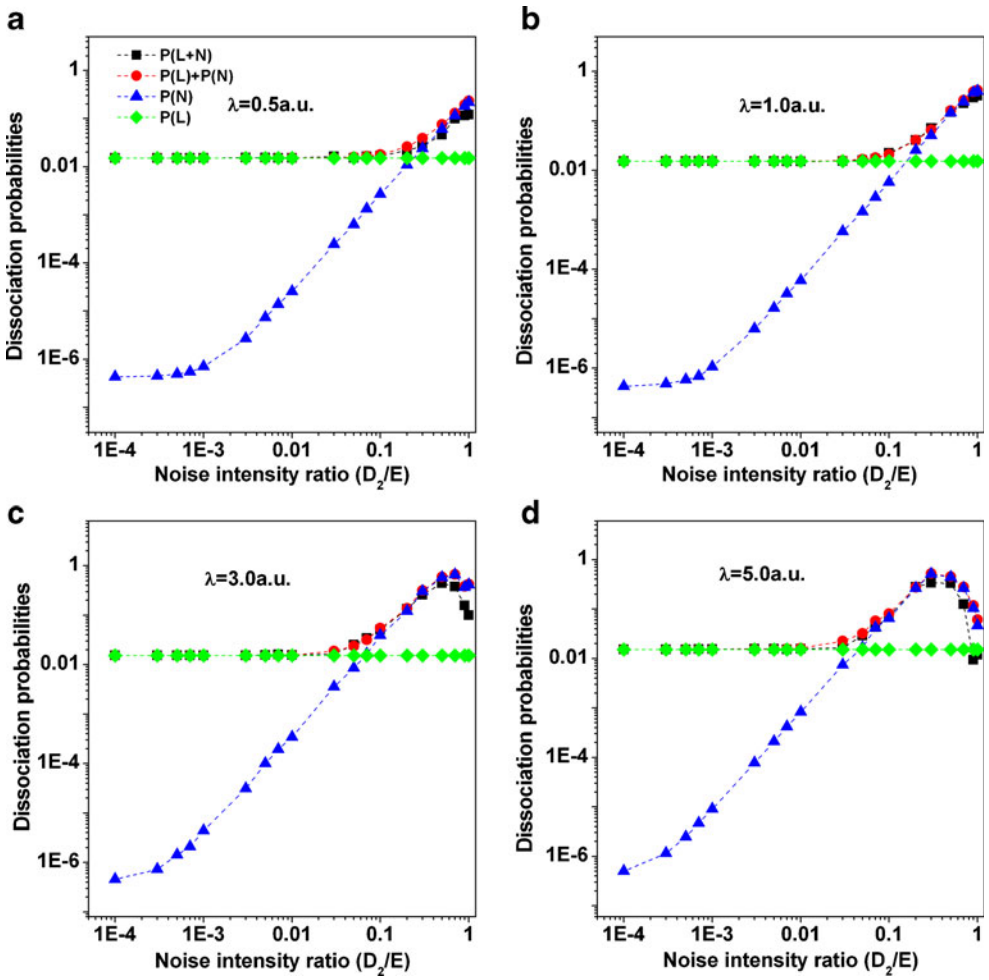

Fig. 5 The DPs as a function of the intensity ratio between the color noise and the laser field **a** for $\lambda=0.5$ a.u., **b** for $\lambda=1.0$ a.u., **c** for $\lambda=3.0$ a.u., and **d** for $\lambda=5.0$ a.u

is understandable, because the noise field will first excite the H_2^+ ion either to its high vibrational state or to the low electronic excited state, then the laser field will further excite it to the dissociative state, leading to an earlier and stronger dissociation. Figure 4b shows the DPs as a function of the intensity ratio, demonstrating that the DPs have almost no changes when $D_1/E<0.07$. Further, as the intensity ratio increases and starting from $D_1/E=0.3$, the contribution from the single Gaussian white noise field begins to exceed that from the combined field, which implies an aggravating role of the noise field in the dissociation dynamics. The rapider drop of the enhancement factor in comparison with the ionization process, as shown in Fig. 4c, indicates that the dissociation process is much quicker to be dominated by the noise field compared with the ionization process shown in Fig. 1c. Moreover, the negative values and the drop trend of the enhancement factor have demonstrated that the degree of the dissociation enhancement has been restrained with the increasing intensity ratio. An exception is that there is a very weak enhancement with $\eta_{max}=0.02$ at low intensity ratio, which can be seen from a high resolution figure shown in the inset of Fig. 4c.

Figure 5a–d show the DPs as a function of the intensity ratio for adding the color noise field with the correlation times being 0.5 a.u., 1.0 a.u., 3.0 a.u. and 5.0 a.u., respectively. The enhancement phenomenon of the DPs has also been observed, and the dissociation is rapidly and dominantly controlled by the color noise. Moreover, with the increasing correlation time, the DPs will keep enhancing. However, for larger correlation time, such as $\lambda=3.0$ a.u. and 5.0 a.u., the DPs have a maximum value around $D_2/E=0.3$, and then they begin to decrease, evidencing the existence of an optimal value for noise intensity in dissociation enhancement. To better understand such dissociation enhancement, the enhancement factors have been shown in Fig. 6a–d. We

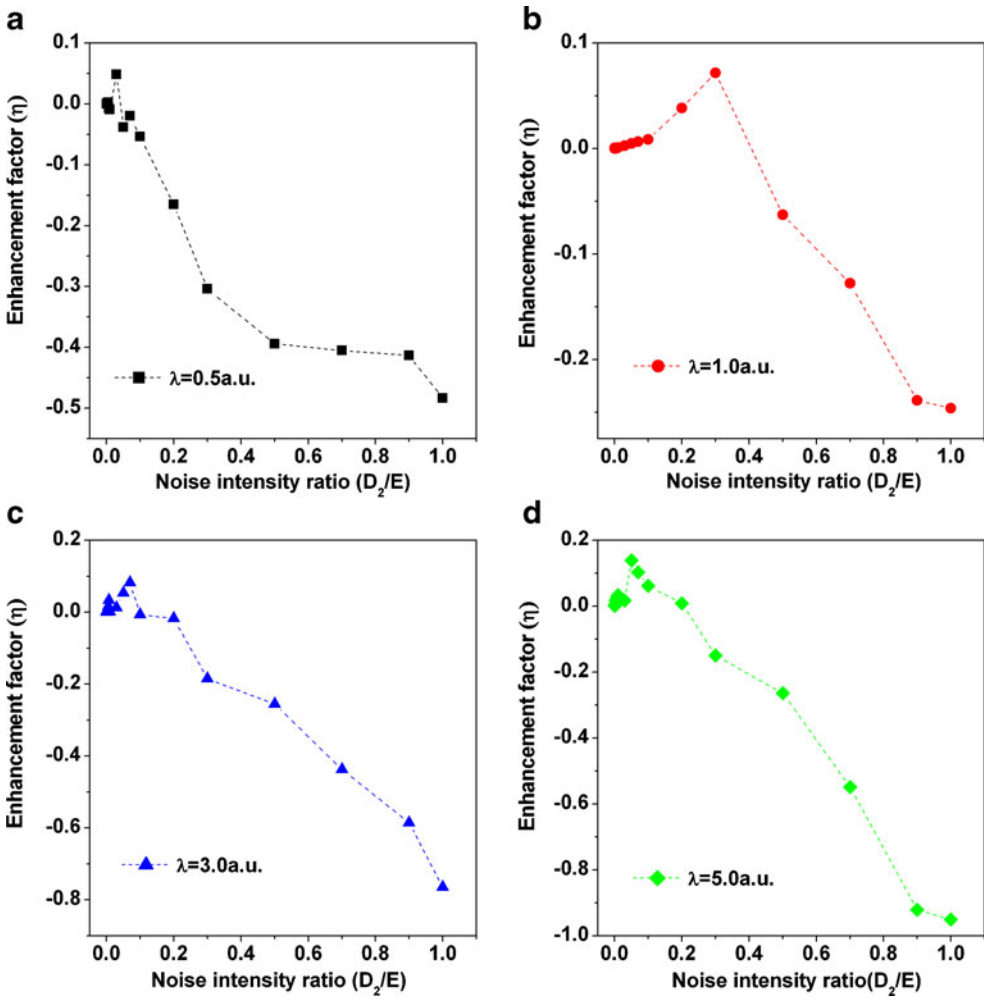

Fig. 6 The enhancement factors of the DPs **a** for λ= 0.5 a.u., **b** for λ=1.0 a.u., **c** for λ=3.0 a.u., and **d** for λ=5.0 a.u

see that the maximum dissociation enhancement factors are all very small in comparison with the ionization cases, and they rapidly drop down to the small and negative values as similar as the Gaussian white noise case.

Through analyzing the DPs, we see that both the Gaussian white noise and the color noise can enhance the photodissociation. And the dissociation process is rapidly and dominantly controlled by the noises at very low intensity in comparison with the ionization process.

Delay time effect

Up to now, we have considered the case of using a pair of simultaneous (laser and noise) fields and observed the ionization and the dissociation enhancement. However, we also wish to know if these enhancements still exist when there is a delay between the laser and the noise. Thus, in Fig. 7, we present the IPs (Fig. 7a and b) and the DPs (Fig. 7c and d) as a function of the delay time in the range [-10o.c., 10o.c.]. Here we use the color noises with the correlation time λ= 3.0 a.u., D_2/E=0.01 (Fig. 7a and c) and 0.05 (Fig. 7b and d) as the examples and the other cases are quite similar to them. Indeed, the enhancement exists in the ionization and the dissociation because of P(L+N)>P(L) or P(N), exhibiting strong dependence on the delay time. For all intensity ratios, the optimal delay time, τ_{delay}=-6o.c.±0.5o.c. for ionization enhancement or τ_{delay}=-1o.c.±0.5o.c. for dissociation enhancement, remains almost the same, illustrating an insensitivity to the noise intensity. And this has been tested and proved for the Gaussian white noise and the color noise with different correlation time. The effect on initial ionization (or dissociation) time of the delay time can be found in Fig. 8, where the time-dependent ionization probability (Fig. 8a–b) and the dissociation probability (Fig. 8c–d) are presented for three special delays of τ_{delay}=-10o.c., 0o.c., and 5o.c. Here, the noise is also chosen to be the color noise with λ=3.0 a.u.. The figure shows that both the initial ionization time (Fig. 8a) and the initial dissociation time (Fig. 8c) change very little with the delay time at the low intensity ratio D_2/E=0.01. However, at higher intensity ratio

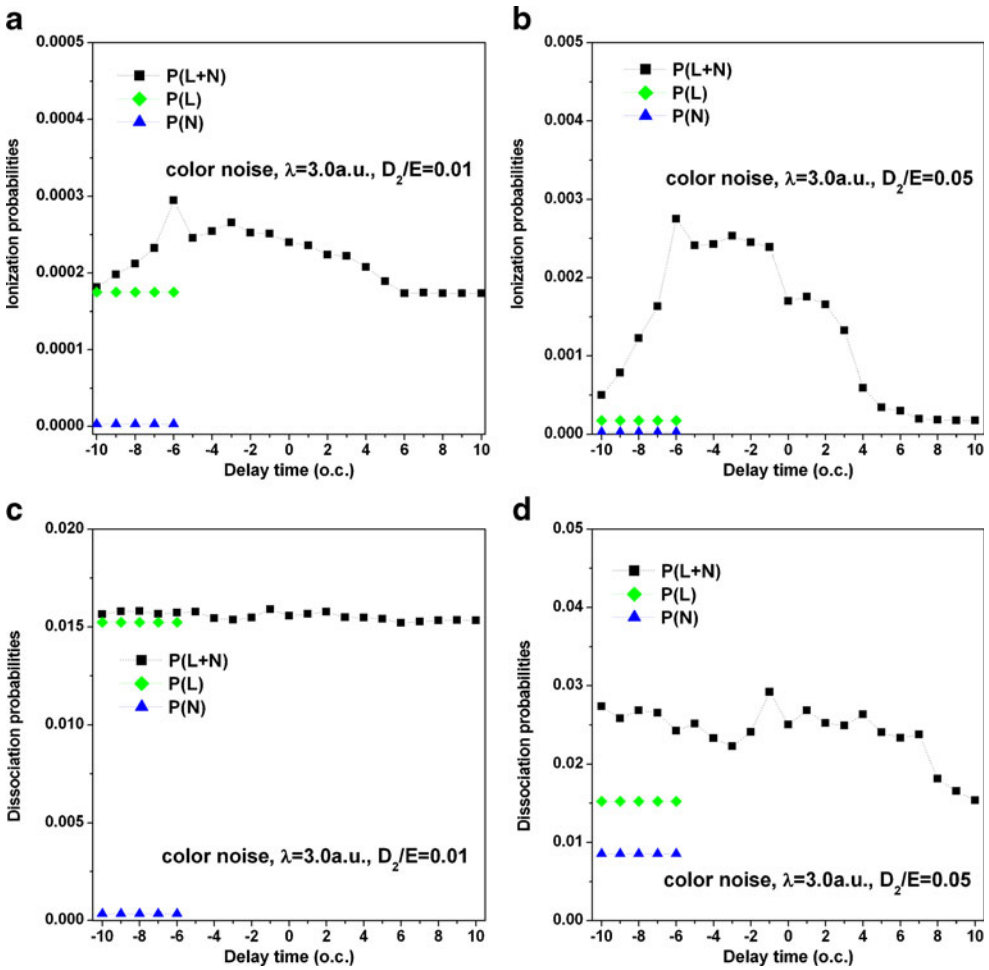

Fig. 7 **a** and **b** The IPs as a function of the delay time between the laser and the noise. The noise is chosen to be the color noise with λ=3.0 a.u., D_2/E=0.01 (Fig. 7a) and λ= 3.0 a.u., D_2/E=0.05 (Fig. 7b). **c** and **d** The DPs a function of the delay time between the laser and the noise. The noises are the same as those in Figs. 7a and b

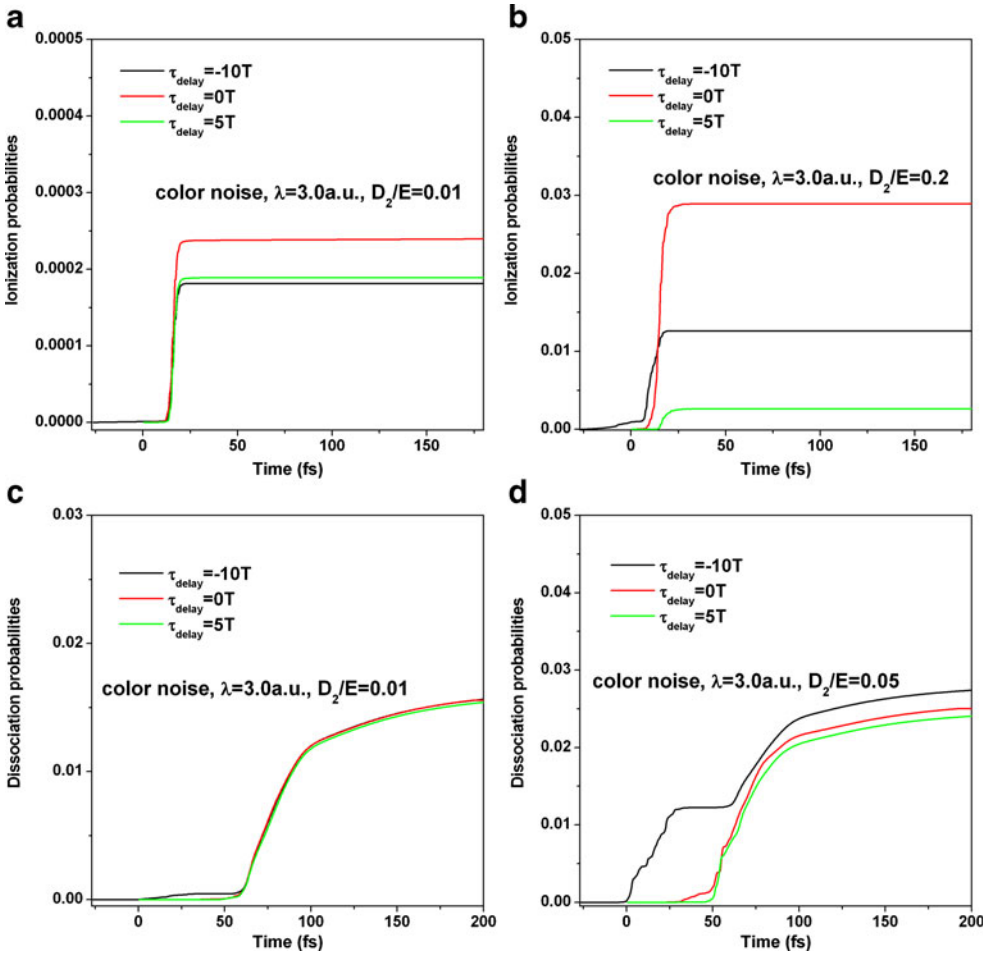

Fig. 8 **a** and **b** The time dependent IPs for three special delay times (τ_{delay}=-10o.c., solid black line; τ_{delay}=0o.c., solid red line; τ_{delay}=5o.c., solid blue line). The noise is chosen to be the color noise with λ=3.0 a.u., D_2/E=0.01 (Fig. 8a) and λ=3.0 a.u., D_2/E=0.2 (Fig. 8b). **c** and **d** The time dependent DPs for the above three special delay times. The noise is chosen to be the color noise with λ=3.0 a.u., D_2/E=0.01 (Fig. 8c) and λ=3.0 a.u., D_2/E=0.05 (Fig. 8d)

(e.g. D_2/E=0.2 for ionization and 0.05 for dissociation as shown in Figs. 8b and d, respectively), the beginning time of the ionization and the dissociation differs at different delay time. This is because at low intensity ratio, the ionization and the dissociation are significantly controlled by the laser field, while as the intensity ratio increases the noises begin to participate and make contribution, causing the differences in the starting time.

Conclusions

In conclusion, using the 1+1D reduced model, we theoretically investigated the intensity enhancement in the photoionization and photodissociation processes when a model H_2^+ ion is exposed to an infrared laser field combined with a noise (the Gaussian white noise or the color noise). It is found that with the introduction of the noise, both the ionization and dissociation probabilities have been enhanced. Through tuning the noise intensity, a stochastic resonancelike curve has appeared for the ionization or dissociation processes, suggesting the existence of an optimal noise intensity for the given laser field. Finally, through analyzing the delay effect between laser and noise, an optimal intensity-independent delay time has been determined for the ionization and dissociation enhancements, respectively.

Acknowledgments This work is supported by National Science Foundation of China (10874096 and 20633070).

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