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Phase control of molecular ionization: $H_2{}^+$ and $H_3{}^{2+}$ in intense two-color laser fields

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Charge resonance-enhanced ionization is examined for combinations of intense, linearly polarized laser fields, and their second harmonic (1ω-2ω) for both a diatomic molecular ion ($H_2{}^+$) and an asymmetric linear triatomic molecular ion ($H_3{}^{2+}$). Ionization is found to be strongly dependent on the *relative phase* between the two colors in both cases. In particular for $H_2{}^+$ one can achieve nearly complete ionization of electrons at one of the nuclei only. For the asymmetric linear triatomic molecular ion ($H_3{}^{2+}$) enhanced ionization is shown also to occur at large critical distances. In the presence of two color fields, the ionization rate of the asymmetric $H_3{}^{2+}$ can be controlled again by tuning the relative phase between the driving fields. The phase control of the enhanced ionization can be rationalized in terms of field-induced over-the-barrier ionization and laser-induced electron localization or equivalently electron tunneling suppression. [S1050-2947(96)05110-4]

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I. INTRODUCTION

Recently it has been shown by three-dimensional (3D) simulations that intense-laser-field ionization of a molecular system can be dramatically different from that of an atomic system [1–3]. As an example, one-electron diatomic molecular ions such as $H_2{}^+$, when subjected to an intense, linearly polarized laser field parallel to the molecular orientation, ionize with a rate one order of magnitude greater than the H atom when stretched to a critical, large internuclear distance, R_c. This laser-induced-molecular enhancement effect can be attributed to electron tunneling suppression by the dynamic Stark effect of the laser and over-the-barrier ionization induced by the instantaneous static laser field (for details, see [1,2]). The latter can also be viewed as an intense static-field tunneling ionization of quantum systems aided by adjacent ion as originally suggested by Codling, Frasinski, and Hatherly [4], and since then confirmed by numerical simulations on 3D [2] and 1D models [3]. We have called this combined effect CREI, charge resonance-enhanced ionization, and it serves now to shed new light on intense-field Coulomb explosion experiments for small molecules [4–6]. Clear signature of this effect has been recently obtained by Corkum and co-workers [7] experimentally in I_2 molecules and Chelkowski *et al.* [8] in fully 3D computer simulations of Coulomb explosion of $H_2{}^+$. Some indication of the manifestation of this effect in laser-cluster interactions [9] and in multielectron diatomics [10] has been obtained in recent classical numerical simulations.

The purpose of this work is to extend previous studies of CREI in diatomic molecular ions [1,2,8] and to triatomic molecular ion systems for combinations of two coherent laser fields with *controllable* relative phase between them. Using the fact that the total field combined from two laser fields of different colors depends on the *relative phase*, one can achieve controllability of atomic and molecular processes as in recent electron current control experiments [11]. Nonlinear optical processes are therefore expected to be sensitive to this phase and should lead to coherent control [12]. In this work, we examine the effect of combinations of a linearly polarized laser field (ω) with its second harmonic field (2ω), the 1ω-2ω combination, on ionization control. This combination is known to break explicitly the reflection symmetry with respect to the midplane perpendicular to the internuclear axis of a diatomic molecule. It leads to the generation and control of even harmonics for molecules interacting with such mixed fields [13]. In the applications of [13] the intensities of the second harmonic (2ω) field were taken to be three orders of magnitude less than the fundamental (1ω) field in order to illustrate the enormous sensitivity of the even harmonic generation on the ''bias'' (the asymmetry) introduced by the 1ω-2ω mixing along the laser polarization axis. In this work the intensities of the two fields are taken to be comparable. This results in a larger ''bias'' in the electric field along the laser polarization axis that is necessary in order to significantly influence the process of tunneling ionization. We will see that ionization of $H_2{}^+$ in such fields strongly depends on the relative phase between the two colors. In fact, for certain phases ionization can basically occur only at one side of the molecular ion. This provides an efficient approach to prepare asymmetric molecular states.

Another goal of this work is to examine whether intense-field ionization of triatomic molecular systems is also strongly structure and field dependent. We will see in this work that ionization is again greatly enhanced when one bond is stretched to a critical large distance along the laser polarization axis. Such an asymmetric linear $H_3{}^{2+}$ system can be viewed equivalently as a H^+-$H_2{}^+$ system, in collision under the influence of a laser. Thus for the 1ω-2ω field combination, we found that the ionization of the H^+-$H_2{}^+$ system, namely, the removal of electron probability from the equilibrium $H_2{}^+$ in the neighborhood of the H^+ ion, is also strongly dependent on the relative phase. This suggests that the cross sections of the field-assisted ion impact ionization of $H_2{}^+$, i.e.,

$$H^+ + H_2{}^+ \rightarrow 3H^+ + e, \qquad (1)$$

can be controlled by varying the relative phase between the colors.

The results presented in this work were obtained by solving the full 3D time-dependent Schrödinger equation for the single-electron molecular ion systems with varying internuclear separation. This is justified when ionization is considered a fast process compared with the concomitant dissociation of the molecules. This approximation is expected to be valid for molecules in intense laser fields (intensity $I \geqslant 10^{14}$ W/cm^2). The numerical techniques employed in this work are described in our previous work [13,14].

II. H_2^+ IN 1ω-2ω FIELDS

We study first the H_2^+ molecular ion with its molecular orientation parallel to the polarization axis of a linearly polarized 1ω-2ω laser field combination with total electric field

$$E(t)=E_0[\cos(\omega t)+\tfrac{1}{4}\cos(2\omega t+\phi)]. \tag{2}$$

The fundamental laser intensity is chosen to be $I_0=6.4\times10^{13}$ W/cm^2 and the second-harmonic laser intensity is $I_2=4\times10^{12}$ W/cm^2. These are so chosen that the effective intensity for the field by Eq. (2) is $I_{\text{eff}}=10^{14}$ W/cm^2 in order to compare with previous single-field calculations [1,2]. The effective intensity I_{eff} for the field of Eq. (2) is defined in this paper to be

$$I_{\text{eff}}(\text{W/cm}^2)=3.52\times10^{16}(5E_0/4)^2 \quad (\text{a.u.}), \tag{2$'$}$$

where $5E_0/4$ is the maximum electric field of Eq. (2) (when, e.g., $\phi=0$ and $\omega t=0$). Laser pulses of such intensities have been shown in our previous calculations to induce CREI through over-the-barrier ionization and dynamic localization (see below). Since the H_2^+ system has cylindrical symmetry in a linearly polarized field collinear with the internuclear axis, the time-dependent Hamiltonian of H_2^+ in a laser field $E(t)\hat{z}$ is given in cylindrical coordinates (ρ-z-θ) as

$$H_t=-\frac{1}{2}\left(\frac{\partial^2}{\partial\rho^2}+\frac{1}{\rho}\frac{\partial}{\partial\rho}+\frac{\partial^2}{\partial z^2}+\frac{\partial^2}{\partial\theta^2}\right)+V_c+zE(t). \tag{3a}$$

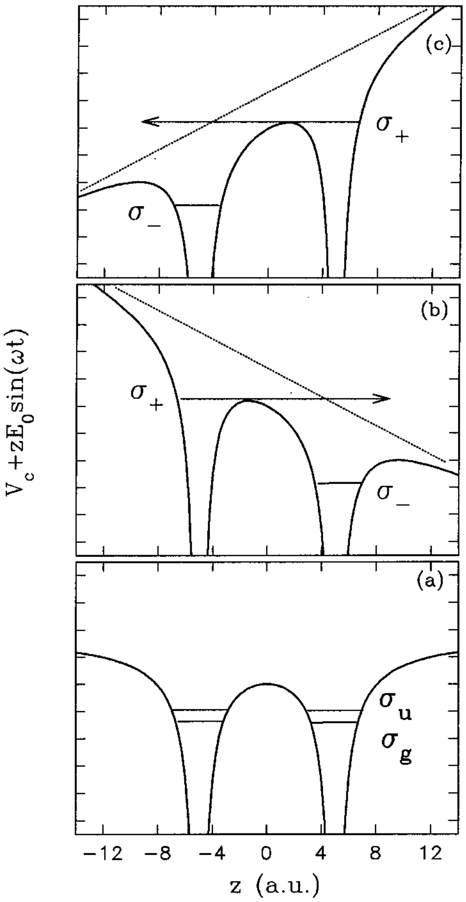

FIG. 1. Section ($\rho=0$) of Coulomb plus external potential for H_2^+ at $R=10$ a.u. in linearly polarized $I=10^{14}$ W/cm^2 fields in cylindrical coordinates highlighting the mechanism of CREI. (a) $\omega t=0$; (b) $\omega t=-\pi/2$; (c) $\omega t=\pi/2$. Solid horizontal lines are the instantaneous field-dressed levels (σ_+, σ_-) evolved from H_2^+ $1\sigma_g$ and $1\sigma_u$ levels. Arrows marks the occurrence of over-the-barrier ionization.

All θ dependence is absent for linearly polarized light parallel to internuclear axis and an initial σ state. In Eq. (3a)

$$V_c=-[\rho^2+(z-R/2)^2]^{-1/2}-[\rho^2+(z+R/2)^2]^{-1/2}, \tag{3b}$$

is the H_2^+ Coulomb potential with R the internuclear distance (atomic units are used throughout this paper unless otherwise indicated). As described in detail in [2], CREI in diatomic molecular ions can be understood in terms of (a) field-induced over-the-barrier ionization along the laser polarization axis (see Fig. 1); (b) tunneling suppression of the electron between the nuclear wells at critical distances R_c. For H_2^+ in a linearly polarized, $I=10^{14}$ W/cm^2, $\lambda=1064$ nm field, our previous calculations showed that the most significant (fastest) ionization occurs for critical distances around $R_c=7$ and 10 a.u. [1,2] with ionization rates at least one order of magnitude larger than that of equilibrium H_2^+ or the separated atom H. In 1ω-2ω fields, the phase ϕ dependence of ionization is of central interest. We therefore concentrate on the new critical R_c region where CREI is expected to occur. Figure 2 shows the results of a series of phase-dependent calculations for the H_2^+ molecular ion in the field given by Eq. (2) with effective intensity $I_{\text{eff}}=10^{14}$ W/cm^2 and wavelength $\lambda=1064$ nm and at $R_c=9$ a.u., where ionization is found to be maximum. In Fig. 2 we define the ''left'' and ''right'' probabilities as follows:

$$P_L=\int_0^{\rho_{\max}}\rho\, d\rho\int_{-z_{\max}}^{0}dz|\psi(\rho,z,t)|^2, \tag{4a}$$

$$P_R=\int_0^{\rho_{\max}}\rho\, d\rho\int_0^{z_{\max}}dz|\psi(\rho,z,t)|^2, \tag{4b}$$

where $\rho_{\max}=16$ a.u. and $z_{\max}=128$ a.u. define the numerical grid used in our 3D calculations and $\psi(\rho,z,t)$ is the total time-dependent wave function. We see that before the turn-on of the field the probabilities of an electron at both the left well (P_L) and at the right well (P_R) are equal (50%). The total probability and the ground-state population are both 100%. In the presence of the field as given by Eq. (2), the total probability starts to decrease due to ionization. Ionized electron probability is removed away in our calculations by absorbing boundaries [13,14], causing the norm of the wave function to decay. Also decaying is the ground-state population. Fast oscillation of the ground-state population is also seen for all phases. These are Rabi oscillations due to

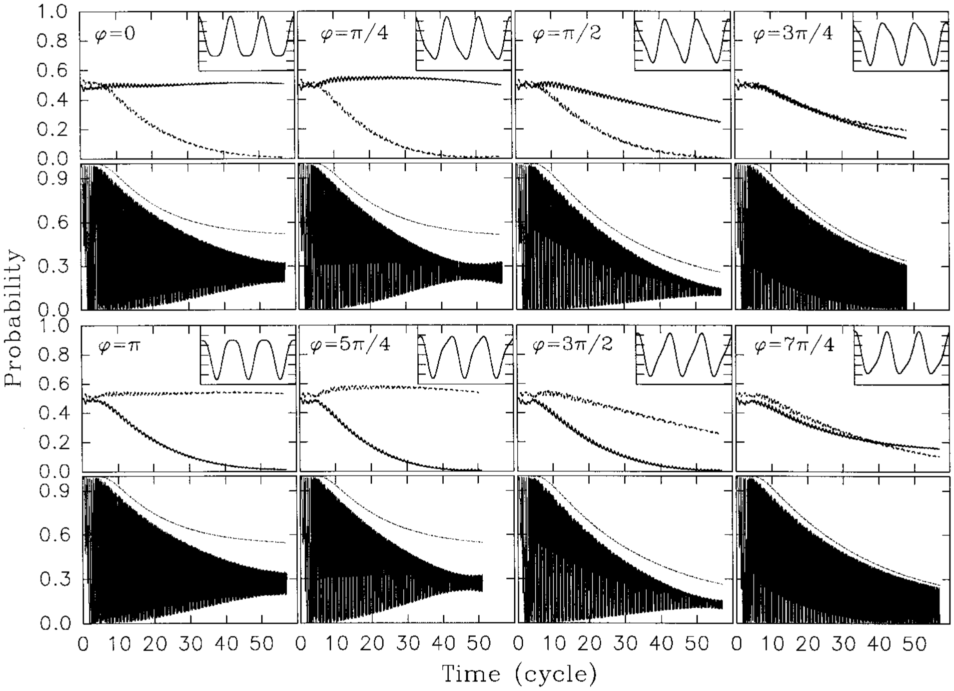

FIG. 2. Ionization of H_2^+ in 1ω-2ω fields given by Eq. (2). Field is turned on linearly in five cycles and is kept constant for 55 cycles (effective $I=10^{14}$ W/cm^2, $E_0=0.0426$ a.u.). Upper plot: solid line, left probability (Eq. 4a); dashed line, right probability (Eq. 4b). Lower plot: solid line, ground-state population; dashed line, total probability (norm of the total time-dependent wave function). Inset plots: $\cos(x)+0.25\cos(2x+\phi)$.

the strong coupling between the charge resonant states, the ground and the first excited states ($1\sigma_g$ and $1\sigma_u$) of H_2^+ and the external electromagnetic fields (the transition moment diverges as $R/2$, see [1]). The equally prepared right and left probabilities start to differ from each other as the interacting time increases. Such effect is most prominent for the case of $\phi=0$ and $\phi=\pi$ because of the maximum ''bias'' (the difference between the positive and negative electric-field strengths) which are tabulated in Table I (see also inset plots of Fig. 2). We emphasize that this asymmetry in ionization is consistent with the quasi-dc-field tunneling model of intense laser-field ionization of atoms [15] as applied to diatomic molecular ions [2,4,5] where the tunneling ionization probability is exponentially dependent on the peaks (maximum) strengths of the oscillatory electric field. The quasi-dc-field tunneling model has been shown to work well for the relative high intensity ($I=10^{14}$ W/cm^2) and long wavelength ($\lambda=1064$ nm) fields used in the present studies [2,3]. For the 1ω-2ω combination of Eq. (2), the values of $E(t)$ at the peaks depend on the relative phase ϕ according to Table I. For the case $\phi=0$ ($\phi=\pi$) the positive (negative) maximum is effectively that of a single-color field of $I=10^{14}$ W/cm^2 that is strong enough to induce over-the-barrier ionization (Fig. 1) whereas the negative (positive) minimum is equivalent to that of a single-color field of only $I=3.6\times10^{13}$ W/cm^2 that is insufficient to induce the over-the-barrier ionization. In fact for $\phi=0$ ($\phi=\pi$) the right (left) probability decreases to nearly zero after 60 cycles (210 fs) whereas the left (right) probability is almost unchanged. For phases other than 0 or π, both the left and right probabilities decay at rates essentially determined by the peak strengths list in Table I. Clearly this allows one at the end of the electric-field interaction to obtain a partial or completely asymmetric state for the H_2^+ molecular ions depending on the relative phase between the two colors.

Figure 3 shows a comparison of the ionization rates as a function of R for the above field combinations [$\phi=0$ or π, Eq. (2)] against the single-field case as reported in [2]. In the latter, the two prominent peaks could be identified as due to dynamic field localization [1,2]. For a two-level system with energy difference ω_0, the effective frequency separation ω_e is modified by the field according to the formula [16,17]

$$\omega_e=\omega_0 J_0(2\omega_R/\omega), \tag{5}$$

where $\omega_R=\mu E_0=E_0R/2$ is the Rabi frequency (note that $2\omega_R=E_0R$ is the field-induced potential difference between the two protons), μ is the transition moment, ω is the field frequency, and J_0 is the zeroth-order Bessel function. Thus electron tunneling between the two protons, which is respon-

TABLE I. Maximum strengths of the $1\omega-2\omega$ field given by Eq. (2). Values are in the unit of $5E_0/4$.

Sign	$\phi=0$	$\phi=\pi/4$	$\phi=\pi/2$	$\phi=3\pi/4$	$\phi=\pi$	$\phi=5\pi/4$	$\phi=3\pi/2$	$\phi=7\pi/4$
positive	1	0.96	0.88	0.74	0.6	0.74	0.88	0.96
negative	0.6	0.74	0.88	0.96	1	0.96	0.88	0.74

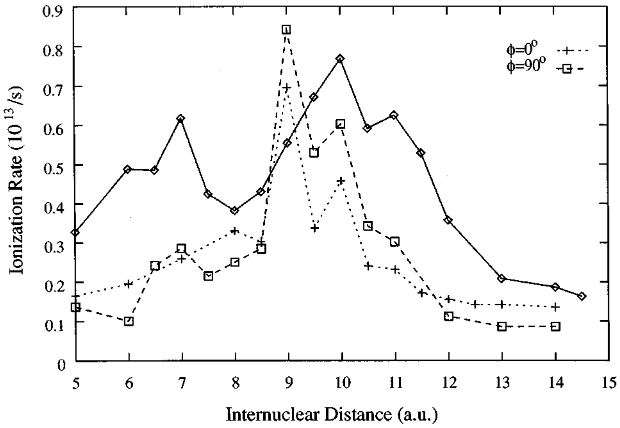

FIG. 3. Ionization rates of H_2^+ as a function of internuclear distance R in laser fields of $I_{\mathrm{eff}}=10^{14}$ W/cm^2, $\lambda=1064$ nm. Solid line: single-color field; dashed line: 1ω-2ω field [Eq. (2)], $\phi=0$ and $\phi=\pi/2$.

sible for the bond in H_2^+, is suppressed whenever $J_0(x)=0$. We have previously correlated even harmonic generation in H_2^+ to such electron tunneling suppression, or equivalently dynamic localization [1,2]. In the presence of 1ω-2ω fields, one sees that the single-field ionization peak at $R_c=7$ for $I=10^{14}$ W/cm^2 is now shifted to large distances $R_c=9$ for phase $\phi=0$ and π. From Eq. (2) we observe that the new critical distances $R_c' \approx 5R_c/4$ if one assumes ionization to occur from E_0 (1ω) only, i.e., the strongest field in the 1ω-2ω combination. This agrees with the results of Fig. 3, and establishes once again both the presence of over-the-barrier ionization and dynamic localization as dominant factors for CREI in single and combinations of fields. We note, however, that for the single-field case, a minimum ionization occurs at $R=8$ a.u. This was shown to be due to residual electron tunneling between the protons. Since the ionization peak at $R=10$ a.u. remains essentially unchanged for 1ω-2ω field excitation (dotted lines) when compared to the single-ω-field excitation (solid line) in Fig. 3, we infer that this peak is mainly a barrier suppression effect as it is frequency independent. On the other hand, the ionization peak that has shifted from 7 to 9 a.u. is obviously frequency and intensity dependent, i.e., the admixture of the 2ω field component has lowered the effective field intensity and the corresponding Rabi frequency ω_R [Eq. (5)] as explained above. This is confirmed in Fig. 3 by the similar phase behavior of the 1ω-2ω field combination at $\phi=0$ and $\pi/2$. The ionization peak at $R=9$ is essentially the same for both phases, reflecting the predominance of the 1ω component. Figure 3 illustrates clearly the main effect of the 1ω-2ω combination: displacement of the $R=7$ a.u. peak to 9 a.u. and essential constancy of the $R=10$ a.u. ionization peak. The first is therefore attributed to dynamic electron localization whereas the latter peak remains a barrier suppression effect.

Furthermore we also note in Fig. 2 that for the maximized field cases $\phi=0$ and $\phi=\pi$ there is a substantial overlap between the final (asymmetric) state $\psi(t_f)$ (t_f=210 fs or 60 optical cycles) and the initial ground state of H_2^+, i.e., $\langle \psi_{1\sigma_g} | \psi(t_f) \rangle \approx 30\%$ (Fig. 2). Since at large internuclear separations the H_2^+ ground $(1\sigma_g)$ and first-excited state $(1\sigma_u)$ states are linear combinations of hydrogenic $1s$ wave functions located at left and right well, respectively (see, e.g., [18]), i.e.,

$$\psi_{1\sigma_{g(u)}} = \frac{1}{\sqrt{2}} [\psi_{1s}(-R/2) + \psi_{1s}(+R/2)], \tag{6a}$$

the final asymmetric state $\psi(t_f)$ thus has a $\sim$40% ($\sqrt{2}\times$30%) overlap with *either* $\psi_{1s}(R/2)$ *or* $\psi_{1s}(-R/2)$. Moreover since the state $\psi_{1s}(R/2)$ or $\psi_{1s}(-R/2)$ is in turn a linear combination (coherent state) of the H_2^+ $1\sigma_g$ and $1\sigma_u$ states, i.e.,

$$\psi_{1s}(\pm R/2) = \frac{1}{\sqrt{2}} (\psi_{1\sigma_g} \pm \psi_{1\sigma_u}), \tag{6b}$$

we thus obtain that for $\phi=0$ or $\phi=\pi$ after 210 fs interaction with the 1ω-2ω pulse, only 50% of H_2^+ is ionized (the norm decreases to 50%, Fig. 2) with the remaining probability primarily in a coherent state of H_2^+ described by Eq. (6b) (40% out of 50%). This provides an efficient way to prepare a coherent state of H_2^+ [Eq. (6b)] via CREI in two-color fields. Such a coherent state has been shown previously to be a unique nonlinear medium for processes such as even harmonic generation [13,17].

The above results are obtained for the homonuclear H_2^+ molecular ions. They should be equally applicable to the heteronuclear HD^+ molecular ions as far as the Coulomb potential and electron-tunneling ionization are concerned. Assuming the left-right selectivity on ionization of Fig. 2 also holds for HD^+ and the molecule disintegrates following the ionization, the $\phi=0$ ($\phi=\pi$) results, i.e., 50% probability survival near the right (left) well and zero near the left (right) well suggest the following dissociative ionization scenario: for $\phi=0$,

$$(50\%)HD^+ \rightarrow H^+ + D^+, \quad (50\%)HD^+ \rightarrow H^+ + D, \tag{7a}$$

$$\Gamma_{H^+/D^+} = 2:1; \tag{7b}$$

for $\phi=\pi$,

$$(50\%)HD^+ \rightarrow H^+ + D^+, \quad (50\%)HD^+ \rightarrow H + D^+, \tag{7c}$$

$$\Gamma_{H^+/D^+} = 1:2. \tag{7d}$$

$\Gamma_{H^+D^+}$ denotes the production ratio of H^+ and D^+. This shows that the photofragments should be strongly influenced and controllable via the relative phase between the two colors of the external fields. Recent experiments by Sheehy, Walker, and DiMauro [20] have shown control on photofragments in multiphoton dissociation of HD^+ in intermediate intense ($I \sim 10^{13}$ W/cm^2) 1ω-2ω field combinations. Our prediction is that the isotope separation ratio should depend on the length of the pulse, i.e., optimum separation will occur after 210 fs for the above laser excitation conditions in fields given by Eq. (2) with $\phi=0$, or π.

III. H_3^{2+} IN 1ω-2ω FIELDS

We now consider the simplest triatomic system, the asymmetrically stretched linear H_3^{2+} in intense laser fields. This can be considered as a H^+-H_2^+ collinear collision in a laser field. Linear H_3^{2+} is not a stable molecule [19] but the non-

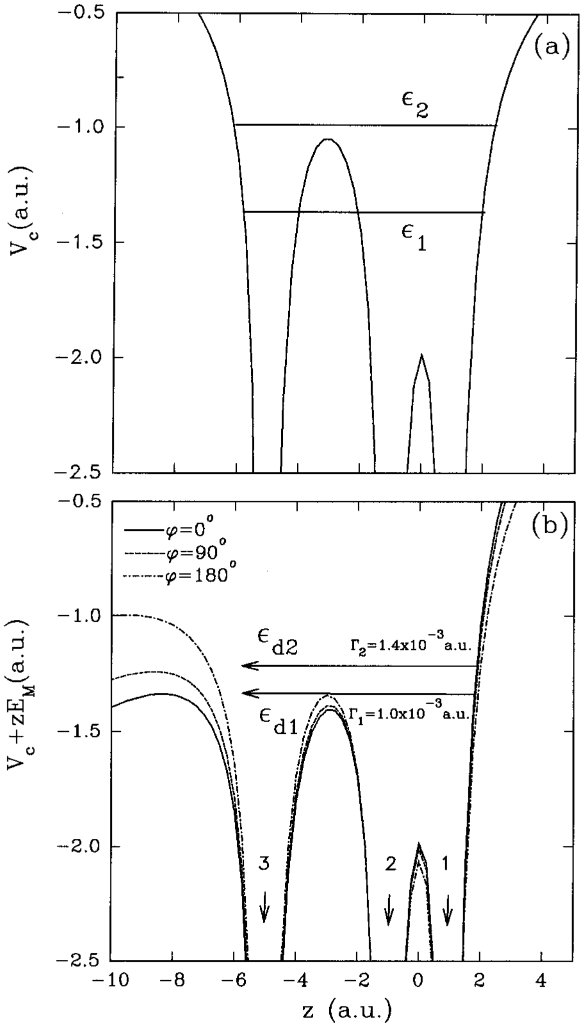

FIG. 4. Section (ρ=0) of potentials for asymmetric H_3^{2+} at L_B=5 a.u. and R=2 a.u. [Eq. (7)] in cylindrical coordinates. Numbered arrows mark the positions of nuclei. The horizontal bars are the lowest energy levels ϵ. (a) The field-free Coulomb potential V_c; (b) the Coulomb plus external electric potential, $V_c+zE_M(\phi)$, where E_M is the maximum positive strength of Eq. (2) given in Table I for effective intensity 5×10^{14} W/cm^2 (E_0=0.095 a.u.). The Γ's are the autoionizing rates associated with the levels (1 a.u.$=4.134\times10^{16}$/s) for ϕ=0.

symmetric H_3^{2+} system is clearly existent during the H^+-H_2^+ collision. Such collision processes have been recently studied in plasma physics [21] and our calculations will emphasize laser-induced effects. In cylindrical coordinates (ρ-z-θ), the time-dependent Hamiltonian of such a 3D system in a laser field $E(t)\hat{z}$ is of the form of Eq. (3a) with the Coulomb potential V_c now given by

$$V_c=-[\rho^2+(z-R/2)^2]^{-1/2}-[\rho^2+(z+R/2)^2]^{-1/2}-[\rho^2+(z+L_B)^2]^{-1/2}, \tag{8}$$

where R is the internuclear distance for the equilibrium H_2^+ and the external bond length L_B is the distance from the H^+ to the *center* of the remaining H_2^+ (see Fig. 4). The process of losing an electron from H_2^+ in the external field obviously depends on the position of the third proton (the H^+ ion).

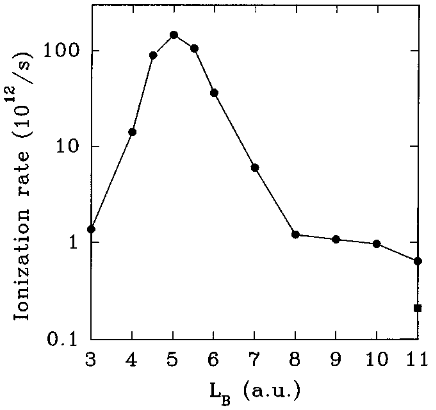

FIG. 5. Ionization rate of the linearly asymmetric stretched H_3^{2+} system in a $I=5\times10^{14}$ W/cm^2, λ=1064 nm single-laser field as a function of the stretched bond length L_B. H_2^+ is taken to be equilibrium (R=2 a.u.). The square on the right vertical axis marks the ionization rate of H_2^+.

When L_B is large, the influence of the H^+ ion on the ionization is negligible and the ionization rate is the normal rate of H_2^+ in the external laser field only. When L_B is small, the attraction force from the third proton (the H^+ ion) will increase the binding force for the electron in H_2^+ and thus prevents the electron from being ionized by the external field. Given the existence of enhanced ionization (CREI) for diatomic molecular ions [1,2], it remains to know whether there exists a similar critical, intermediate-large distance where the ionization rate exceeds the ionization rate of H_2^+. The numerical results for the ionization rates are plotted in Fig. 5 for the above-described system as a function of the external bond length L_B for the laser condition of $I=5\times10^{14}$ W/cm^2, λ=1064 nm. The intensity is chosen to allow appreciable ionization to occur for equilibrium H_2^+ (R=2 a.u.) in the single field alone (the ionization rate is then 2.2×10^{11}/s). We see that the calculated ionization rates indeed show a prominent maximum at L_B=5 a.u. that is *two orders* of magnitude larger than either rate of H_2^+ alone (L_B large) or that of H_3^{2+} where L_B is small. The results shown in Fig. 5 provide a direct numerical confirmation that intense-field ionization of triatomic molecular ion systems is also strongly structure dependent as in diatomic molecular systems [22]. Clearly ionization can be enhanced by orders of magnitude when a bond of the molecule is stretched along the laser polarization axis to a critical, large distance.

This enhancement of ionization can be understood again as field-induced over-the-barrier ionization of the instantaneous field-dressed levels [1–4]. The lowest two bare and field-dressed energy levels for the H^+-H_2^+ system at L_B=5 a.u. and R=2 a.u. are calculated and shown in Fig. 4. The ground state has an energy of $\epsilon_1=-1.3497$ a.u. slightly lower than the H_2^+ ground state at equilibrium internuclear separation (−1.1 a.u.) due to the attraction of the third proton. The first excited state of the H^+-H_2^+ system is relatively far away from the ground state with the energy $\epsilon_2=-0.9792$ a.u. in contrast to the case of H_2^+ at large internuclear separation where the ground state and the first-excited states are almost degenerate. The energy difference between the ground and the first-excited states is $\epsilon_2-\epsilon_1=0.37$ a.u., which is much larger than both the laser frequency ω=0.043 a.u.

TABLE II. The autoionization rates and energies of dc-field-dressed levels. Values are in atomic units. For ionization rates, 1 a.u.$=4.134\times10^{16}$/s.

Phase	$\phi=0$	$\phi=\pi$
Γ_1	0.001 03	0.000 02
Γ_2	0.001 42	0.000 05
ϵ_{d1}	$-1.356\,22$	$-1.211\,00$
ϵ_{d2}	$-1.270\,68$	$-1.178\,95$

and the coupling (Rabi) frequency, $\Omega_R=\mu\mathcal{E}_0=0.098$ a.u. between these two levels (the transition moment μ is calculated to be 0.82 a.u. and $\mathcal{E}_0=0.119$ a.u.). The fact that the energy difference $\epsilon_2-\epsilon_1$ is much larger than the Rabi frequency Ω_R suggests that the energy level of the field-dressed ground state will be close to the bare ground-state level. This is confirmed by numerical calculations [Fig. 4(b)] using a technique described in [2]. In a dc field with strength equalling the maximum field strength of Eq. (2) for an effective intensity 5×10^{14} W/cm^2 at $\phi=0$, the value of the dressed level is calculated to be $\epsilon_{d1}=-1.3556$ a.u., very close to the bare level ϵ_1 (-1.3497 a.u.). This level can therefore be considered to evolve primarily from the ground state of the H^+-H_2^+ system. Also shown in Fig. 4(b) is the second lowest dressed level with energy $\epsilon_{d2}=-1.211$ a.u. The dc-field-dressed levels are, of course, not true bound states. Rather they are autoionizing states in the presence of the dc field as marked by arrows in Fig. 4(b) for the case $\phi=0$. The autoionizing rates $\Gamma_{1,2}$ and energies are also listed in Table II. As in the case of diatomic molecular ions [2] the higher dressed levels are populated by the nonadiabatic effect induced by the oscillatory electric fields [2] (see also Fig. 11 in [23]). The enhancement of ionization at the critical bond length ($L_B=5$ a.u.) can therefore be attributed as a consequence of over-the-barrier ionization of the instantaneous dc-field-dressed levels induced by the laser. When L_B is much less than the critical bond length (5 a.u.) the outer barrier in Fig. 4 (the barrier to the left of proton ''3'') becomes high and confines the dressed levels. The inner barrier (the barrier between proton ''1'' and ''2'') becomes low and so do the levels. This greatly diminishes the ionization probability. In the other limit, namely, when L_B is much greater than the critical bond length, the inner barrier grows such that the H^+ ion is basically isolated from the remaining H_2^+ and has little influence on the ionization. The ionization rate then approaches the much lower normal rate of H_2^+ (2.2×10^{11}/s) in the same fields alone.

In summary, the enhanced ionization of the system of H^+-H_2^+ can be viewed as a process of an adjacent-ion (H^+) aided tunneling ionization of the quantum system (H_2^+) in the intense laser field. The adjacent ion serves to lower in concert with the external electric field the potential barrier that confines the electron to allow the occurrence of over-the-barrier ionization and consequently the enhanced ionization.

We now turn to the control of ionization for the triatomic system, H^+-H_2^+ at the critical L_B distance, in a 1ω-2ω field. Without loss of generality, we have assumed that the adjacent ion is at the left side of H_2^+ (see Fig. 4). We have seen that the enhancement of ionization of the nonstretched H_2^+ in the presence of a neighboring ion occurs because the external ion's Coulomb field ''adds'' up with the external electric field that causes the over-the-barrier ionization when the field acquires its *positive* maximum strengths. This is the essence of the quasi-dc-field tunneling model of intense-field ionization [15]. Clearly in a 1ω-2ω field the maximum positive strength of the the field varies largely depending on the phase (Table I). The adjacent ion-aided over-the-barrier ionization can thus be virtually turned on and off depending on the phases as seen in Fig. 4 (''on'' for $\phi=0$ and ''off'' for $\phi=\pi$). The molecule survival probability [the norm of the wave function, $\int_0^{\rho_{\max}}\rho\, d\rho\int_{-z_{\max}}^{z_{\max}}dz|\psi(\rho,z,t)|^2$] for the H^+-H_2^+ system at $L_B=5$ a.u. and $R=2$ a.u. in an 1ω-2ω external field given by Eq. (2) with effective $I=5\times10^{14}$ W/cm^2 are shown in Fig. 6 as a function of the relative phase ϕ. One sees that the degree for control of the ionization is considerable: the electron survival probability quickly diminishes to nearly zero after the five cycle linear turn-on of the field for phases around 0°, whereas it remains almost unity for phases around ±180°. This sensitivity of ionization on the peak field strength is consistent with the *exponential* dependence of ionization probability on the peak strength of the field predicted by the quasi-dc-field tunneling model [15]. Around 0° the negative (left) peak strength (Table I) reaches maximum (100%) and around ±180° it is at the minimum (60%). Comparing Table II and Fig. 4 one also sees that for $\phi=\pm\pi$ the two levels are bound by the outer barrier whereas for $\phi=0$ both levels are over the outer barrier. This is different from H_2^+ (Sec. II) where $\phi=0$ and $\phi=\pi$ situations are symmetric in terms of the ionization of the upper dressed levels (σ_+, Fig. 1). Clearly one can expect that the dependence of ionization on the relative phase between the two-color field, as seen in the process of laser-assisted H^+-H_2^+ collision in 1ω-2ω fields will apply to other general laser-assisted ion-ion collisions.

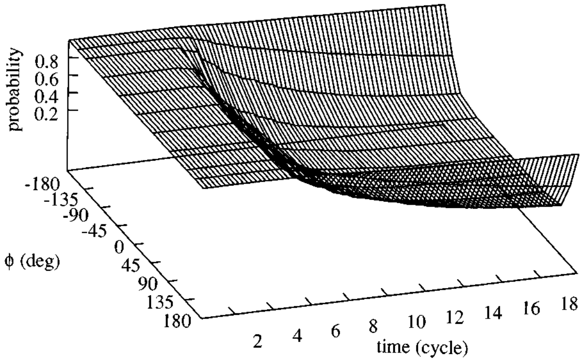

FIG. 6. The electron survival probability of the linearly asymmetric H_3^{2+} system in 1ω-2ω fields. $E_0=0.095$ a.u. (effective $I=5\times10^{14}$ W/cm^2, $L_B=5$ a.u.).

IV. CONCLUSIONS

This work extends our previous studies on enhanced ionization (CREI) of diatomic molecular ions in intense laser fields to combinations of laser fields with their harmonics and to triatomic systems. Such combinations explicitly break the reflection symmetry along the laser polarization axis and cause the ionization to be anisotropic. We have shown that via tuning the relative phase between the two colors, we can turn ‘‘on’’ or ‘‘off’’ the ionization from either side of H_2^+, thus creating an asymmetric coherent electronic state of the molecular ions after the interaction with short laser pulses. For the asymmetric linear H_3^{2+} system, we have shown that enhanced ionization again occurs at a large, critical distance via a so-called adjacent-ion aided ionization. This ionization can be again turned ‘‘on’’ or ‘‘off’’ by tuning the relative phase between the two colors of a combination of 1ω-2ω driving fields. The effect should therefore occur readily in atom-atom or atom-molecule ion-ion collisions.

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