

## Effect of nuclear motion on tunneling ionization rates of molecules

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We show that the observable rate of tunneling ionization of a molecule in an intense low-frequency laser field is affected by nuclear motion and can essentially differ from a bare electronic characteristic calculated for fixed nuclei. Both the absolute value of the rate and the shape of its orientation dependence are affected. The effect is significant for  $I \sim 10^{14}$  W/cm<sup>2</sup> and becomes more pronounced at lower intensities. An isotope effect in tunneling ionization of H<sub>2</sub> and D<sub>2</sub> is predicted. The results are compared with available experiments.

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The interaction of molecules with intense low-frequency laser pulses is a rapidly developing field attracting much theoretical and experimental interest. One of the main goals is to establish techniques for retrieving the molecular structure information from experimental observables. The retrieving procedure requires the knowledge of the tunneling ionization rate of a target molecule. This important characteristic of the first step of the interaction process depends on the internuclear configuration and the symmetry of an ionizing orbital which are revealed in its dependence on the orientation of a molecule with respect to the laser field. Thus even relatively simple measurements of orientation-resolved total ionization yields already give valuable structure information [1]. More detailed information can be obtained from photoelectron [2–5] and harmonic [2,6,7] spectra, provided that accurate ionization rates needed for their analysis are available. The recently developed weak-field asymptotic theory (WFAT) of tunneling ionization of molecules [8] and its implementation on the basis of quantum chemistry codes [9,10] enable one to reliably evaluate the rates. So far, this theory is restricted to the single-active-electron and frozen-nuclei approximations. In this Rapid Communication, we consider the effect of nuclear motion on the observable tunneling ionization rate of an electron. The effect is shown to be strong, both for absolute values of the rate and the shape of its orientation dependence. The incorporation of nuclear motion on the basis of earlier tunneling theories was discussed in Ref. [11]. It was taken into account in the analysis of the harmonic generation process in Refs. [12,13]. Recent *ab initio* studies [14,15] aim at treating nuclear and electronic motions on equal footing.

We consider tunneling ionization in a static electric field  $\mathbf{F} = F\mathbf{e}_z$ ; the results apply also to time-dependent laser fields in an adiabatic regime specified below. We still employ the single-active-electron approximation, but nuclei can now move. For simplicity, we consider a diatomic molecule. The stationary Schrödinger equation reads (atomic units are used throughout)

$$\left[ -\frac{\Delta_{\mathbf{R}}}{2\mu_a} - \frac{\Delta_{\mathbf{r}}}{2\mu_e} + U(R) + V(\mathbf{r}; \mathbf{R}) - \mathbf{D}(\mathbf{R})\mathbf{F} + \mathbf{r}\mathbf{F} - E \right] \times \Psi(\mathbf{r}, \mathbf{R}) = 0. \quad (1)$$

The heavy subsystem is described by the internuclear vector  $\mathbf{R}$ , the reduced mass of the atoms  $\mu_a$ , the interatomic interaction potential in the molecular ion  $U(R)$ , and its dipole moment  $\mathbf{D}(\mathbf{R}) = D(R)\mathbf{N}$ , where  $\mathbf{N} = \mathbf{R}/R$ . The electron's coordinate  $\mathbf{r}$  is measured from the center of mass of the atoms,  $\mu_e$  is its reduced mass, and  $V(\mathbf{r}; \mathbf{R})$  describes its interaction with the molecular ion. The electronic and nuclear motion can be separated in the Born-Oppenheimer approximation. In the leading order in  $1/\mu_a$ ,  $\mu_e = 1$  and the solution to Eq. (1) takes the form  $\Psi(\mathbf{r}, \mathbf{R}) = \psi(\mathbf{r}; \mathbf{R})\Psi(\mathbf{R})$ , where the electronic and nuclear wave functions satisfy

$$\left[ -\frac{\Delta_{\mathbf{r}}}{2} + V(\mathbf{r}; \mathbf{R}) + \mathbf{r}\mathbf{F} - E(\mathbf{R}) \right] \psi(\mathbf{r}; \mathbf{R}) = 0 \quad (2)$$

and

$$\left[ -\frac{\Delta_{\mathbf{R}}}{2\mu_a} + U(R) + E(\mathbf{R}) - \mathbf{D}(\mathbf{R})\mathbf{F} - E \right] \Psi(\mathbf{R}) = 0. \quad (3)$$

Equation (2) should be supplemented by the outgoing-wave boundary condition accounting for tunneling ionization of the electron, which turns it into a Siegert eigenvalue problem [16]. The electronic energy eigenvalue  $E(\mathbf{R})$  is complex for  $F \neq 0$ . The WFAT [8] gives the asymptotic solution to this problem for  $F \ll F_c$ , where  $F_c$  is a boundary between tunneling and over-the-barrier regimes of ionization. In the present formulation, the potential in Eq. (2), and hence the solution, are dependent on  $\mathbf{R}$  as a parameter. For neutral molecules in electronic states which correlate with two neutral atoms, the solution to Eq. (3) must vanish as  $R \rightarrow \infty$ . Then the imaginary part of the total energy eigenvalue  $E$  defines the ionization rate. For molecular ions  $E(\mathbf{R}) - \mathbf{D}(\mathbf{R})\mathbf{F} \sim R$  as  $R \rightarrow \infty$ , so Eq. (3) also should be supplemented by the outgoing-wave boundary condition accounting for nuclear tunneling. In this case, the imaginary part of  $E$  acquires a contribution corresponding to dissociation.

Equations (2) and (3) present a rich theoretical model for treating the electronic and nuclear dynamics in an external electric field. In the following, we adopt a number of rather crude approximations which, however, are commonly used along with the Born-Oppenheimer approximation in molecular physics. This is sufficient for a simple analysis of the problem; a more elaborate theory can be developed later.

Let, for  $F = 0$ , Eqs. (2) and (3) have bound-state solutions  $E_v(R)$ ,  $\psi_v(\mathbf{r}; \mathbf{R})$ ,  $E_{vJ}$ , and  $\Psi_{vJM}(\mathbf{R}) = R^{-1}\chi_{vJ}(R)Y_{JM}(\mathbf{N})$ ,

where the subscript  $e$  identifies the electronic state and the nuclear radial function satisfies

$$\left[ -\frac{1}{2\mu_a} \frac{d^2}{dR^2} + \frac{J(J+1)}{2\mu_a R^2} + U(R) + E_e(R) - E_{vJ} \right] \chi_{vJ}(R) = 0. \quad (4)$$

For low-lying vibrational and rotational states one can approximately set  $\chi_{vJ}(R) \approx \chi_{v0}(R)$  and  $E_{vJ} \approx E_{v0} + B(R_0)J(J+1)$ , where  $B(R_0) = (2\mu_a R_0^2)^{-1}$  is the rotational constant and  $R_0$  is the equilibrium internuclear distance at which the function  $U(R) + E_e(R)$  attains its minimum. The effect we are interested in is more pronounced in weak fields. In this case, the eigenvalue of Eq. (2) is given by [8]

$$E(\mathbf{R}) = E_e(R) - \mathbf{d}_e(\mathbf{R})\mathbf{F} + O(F^2) - \frac{i}{2} \Gamma_e(\mathbf{R}), \quad (5)$$

where  $\mathbf{d}_e(\mathbf{R}) = -\langle \psi_e | \mathbf{r} | \psi_e \rangle = d_e(R)\mathbf{N}$  is the dipole moment of the electron in the state  $\psi_e(\mathbf{r}; \mathbf{R})$  and  $\Gamma_e(\mathbf{R})$  is its tunneling ionization rate. The second-order Stark shift can be included into Eq. (5), but this does not qualitatively change the results. The rate  $\Gamma_e(\mathbf{R})$  is exponentially small in  $F$ , so the interaction with field in Eq. (3) is represented by  $-\mu(R)\mathbf{N}\mathbf{F}$ , where  $\mu(R) = D(R) + d_e(R)$  is the total molecular dipole. This interaction preserves the projection  $M$  of the nuclear angular momentum  $\mathbf{J}$  onto the laboratory  $z$  axis. Let  $\Delta E_{nM}$  and  $X_{nM}(\mathbf{N})$ ,  $n = 1, 2, \dots$ , be the pendulum states defined by the eigenvalue problem [17]

$$[B(R_0)\mathbf{J}^2 - \mu(R_0)\mathbf{N}\mathbf{F} - \Delta E_{nM}]X_{nM}(\mathbf{N}) = 0. \quad (6)$$

The solutions can be expanded as

$$X_{nM}(\mathbf{N}) = \sum_J c_{JM}^n Y_{JM}(\mathbf{N}). \quad (7)$$

Then an approximate solution to Eq. (3) is given by

$$E_{vnM} = E_{v0} + \Delta E_{nM} - \frac{i}{2} \Gamma_{vnM}, \quad (8a)$$

$$\Psi_{vnM}(\mathbf{R}) = R^{-1} \chi_{v0}(R) X_{nM}(\mathbf{N}), \quad (8b)$$

where

$$\Gamma_{vnM} = \int \Gamma_v(\mathbf{N}) |X_{nM}(\mathbf{N})|^2 d\mathbf{N} \quad (9)$$

and

$$\Gamma_v(\mathbf{N}) = \int \Gamma_e(\mathbf{R}) \chi_{v0}^2(R) dR. \quad (10)$$

Here, we have substituted  $\mu(R)$  by its equilibrium value  $\mu(R_0)$ , hence neglecting nuclear tunneling, and have treated the last term in Eq. (5) perturbatively. The observable ionization rate of the molecule in a state  $vnM$  is thus given by Eq. (9).

The bare rate  $\Gamma_e(\mathbf{R})$  is a property of the electronic state for fixed nuclei. This is the most detailed characteristic which explicitly depends on both the internuclear distance  $R$  and orientation  $\mathbf{N}$  of the molecule. The other two rates appearing in the above formulation successively incorporate the effect of nuclear motion. The intermediate internuclear-distance-averaged rate  $\Gamma_v(\mathbf{N})$  explicitly depends only on the orientation and is affected by the probability distribution in the vibrational state  $\chi_{v0}(R)$ . The observable orientation-averaged rate  $\Gamma_{vnM}$  does not bear explicit dependence on the positions of nuclei

and is, in addition, affected by the structure of the rotational state (7). So far,  $\Gamma_e(\mathbf{R})$  has been the main quantity of interest in theoretical calculations [8–10]. It was generally assumed that  $\Gamma_e(R_0\mathbf{N})$  averaged over a distribution of orientations in a given experiment is what should be compared with the experimental results [1]. For typical laser fields  $F \sim 0.1$  ( $I \sim 3.5 \times 10^{14}$  W/cm<sup>2</sup>) of interest for applications, the interaction with field in Eq. (6) exceeds the energy of field-free rotational excitation. The low-lying pendulum states (7) are then sharply oriented along the field to minimize the interaction energy. There exist various techniques to create sharply aligned [18] or oriented [19] rotational wave packets by time-dependent laser fields applicable to polar as well as nonpolar molecules. Our arriving at pendulum states (7) via Eq. (6) in the adiabatic regime is just one of the possible scenarios. Here we do not discuss averaging over orientations in Eq. (9), because the actual contents of the rotational wave packet (7) is determined by particular experimental conditions and may even depend on time. But we discuss the effect of averaging over the internuclear distance in Eq. (10). The main message of this work is that  $\Gamma_v(\mathbf{N})$  may essentially differ from  $\Gamma_e(R_0\mathbf{N})$ .

For diatomic molecules  $\Gamma_e(\mathbf{R}) = \Gamma_e(R, \beta)$  and  $\Gamma_v(\mathbf{N}) = \Gamma_v(\beta)$ , where  $\beta$  is the angle between  $\mathbf{R}$  and  $\mathbf{F}$ . In the leading-order approximation of the WFAT, the ionization rate of an electron in a  $\sigma$  state is given by [8]

$$\Gamma_e(R, \beta) = |G_{00}(R, \beta)|^2 W_{00}(F; R), \quad (11)$$

where  $G_{00}(R, \beta)$  is the structure factor defined by the asymptotic tail of the unperturbed orbital  $\psi_e(\mathbf{r}; \mathbf{R})$  [9,10] and  $W_{00}(F; R)$  is the field factor,

$$W_{00}(F; R) = \frac{\varkappa}{2} \left( \frac{4\varkappa^2}{F} \right)^{\frac{2}{\varkappa}-1} \exp\left(-\frac{2\varkappa^3}{3F}\right) \Big|_{\varkappa=\varkappa(R)}. \quad (12)$$

Here  $\varkappa(R) = \sqrt{2|E_e(R)|}$  and the subscript refers to the dominant ionization channel with parabolic quantum numbers  $(n_\xi, m) = (0, 0)$ . The origin of the difference between  $\Gamma_v(\beta)$  and  $\Gamma_e(R_0, \beta)$  mentioned above lies in a very strong dependence of the exponential factor in Eq. (12) on the electronic energy  $E_e(R)$ , and hence on  $R$  [20]. While slowly varying functions such as  $B(R)$  and  $\mu(R)$  can be substituted by their equilibrium values upon averaging over  $R$  in low-lying vibrational states, the averaging of  $\Gamma_e(R, \beta)$  in Eq. (10) should be done more carefully. To estimate the effect, let us substitute the structure factor in Eq. (11) and the preexponential factor in Eq. (12) by their values at  $R = R_0$ , but in the exponential factor we set  $E_e(R) \approx E_e(R_0) + \alpha_e \delta R$  and expand  $\varkappa^3(R)$  in  $\delta R = R - R_0$ . The vibrational wave function  $\chi_{v0}(R)$  is approximated by that of a harmonic oscillator with a frequency  $\omega_e$  defined by the expansion  $U(R) + E_e(R) \approx U(R_0) + E_e(R_0) + \frac{1}{2}\mu_a \omega_e^2 \delta R^2$ . Substituting this into Eq. (10), we obtain

$$\frac{\Gamma_v(\beta)}{\Gamma_e(R_0, \beta)} \approx e^\zeta L_v(-2\zeta) \geq 1, \quad \zeta = \frac{\alpha_e^2 \varkappa^2(R_0)}{\mu_a \omega_e F^2}, \quad (13)$$

where  $L_n(x)$  is a Laguerre polynomial. Thus the difference between  $\Gamma_v(\beta)$  and  $\Gamma_e(R_0, \beta)$  is controlled by the parameter  $\zeta$ . The function  $e^\zeta L_v(-2\zeta)$  is equal to 1 for  $\zeta = 0$  and monotonically grows with  $\zeta$ . So the ratio in Eq. (13) is close

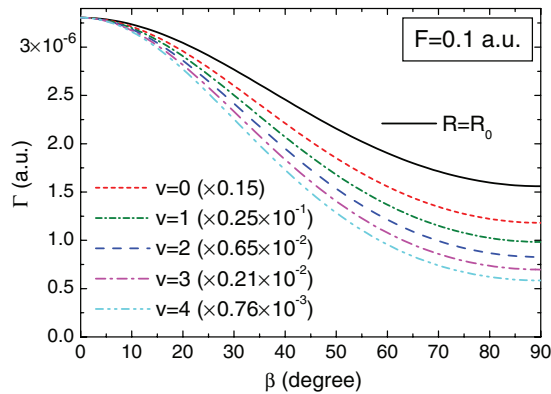


FIG. 1. (Color online) Ionization rates of  $\text{H}_2^+$  for  $F = 0.1$  a.u. Solid line:  $\Gamma_e(R_0, \beta)$ . Dashed lines:  $\Gamma_v(\beta)$  for  $v = 0, \dots, 4$  multiplied by the indicated factors.

to unity for  $\zeta \ll 1$ . However, since  $\zeta$  strongly depends on  $F$ , for sufficiently weak fields this ratio can become large.

This simple estimate is supported by accurate calculations. We have calculated  $\Gamma_v(\beta)$  for a hydrogen molecular ion  $\text{H}_2^+$  in the  $1s\sigma$  state by solving Eq. (4) and using Eqs. (10)–(12) without any additional approximations. Figure 1 shows  $\Gamma_v(\beta)$  for several lowest vibrational states at  $F = 0.1$ . Figure 2 illustrates the dependence of  $\Gamma_v(\beta)$  on  $F$ . The results are compared with  $\Gamma_e(R_0, \beta)$ . In all the cases, there is a large difference between the absolute values of  $\Gamma_v(\beta)$  and  $\Gamma_e(R_0, \beta)$  characterized by numerical factors indicated in the figures. This difference qualitatively agrees with Eq. (13). Indeed, for  $\text{H}_2^+$  we have  $\zeta = 0.0142/F^2$ . Then, for example, for  $v = 0$  and  $F = 0.1$ , the ratio  $\Gamma_v(\beta)/\Gamma_e(R_0, \beta)$  estimated from Eq. (13) is 4.1, while its value obtained from accurate calculations varies from 6.7, for  $\beta = 0^\circ$ , to 5.0, for  $\beta = 90^\circ$ . The dependence of the ratio on  $\beta$  reflects a difference between the shapes of  $\Gamma_v(\beta)$  and  $\Gamma_e(R_0, \beta)$  as functions of the orientation angle  $\beta$ , which is not accounted for by Eq. (13). This difference is also appreciable and would be observable in an experiment of the type reported in Ref. [1].

The parameter  $\zeta$  depends on the reduced mass  $\mu_a$ , so Eq. (13) predicts an isotope effect. To illustrate this, we have calculated  $\Gamma_{v=0}(\beta)$  for hydrogen  $\text{H}_2$  and deuterium  $\text{D}_2$  molecules in the ground electronic state treated in the

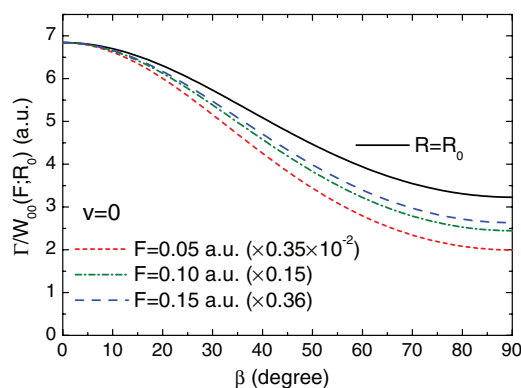


FIG. 2. (Color online) Normalized ionization rates of  $\text{H}_2^+$  [see Eq. (12)]. Solid line:  $\Gamma_e(R_0, \beta)$ . Dashed lines:  $\Gamma_{v=0}(\beta)$  for three values of  $F$  multiplied by the indicated factors.

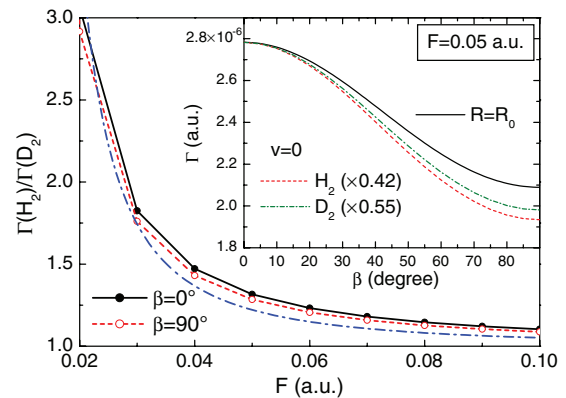


FIG. 3. (Color online) The ratio of the ionization rates  $\Gamma_{v=0}(\beta)$  of  $\text{H}_2$  and  $\text{D}_2$  as a function of  $F$  for  $\beta = 0^\circ$  (solid black line) and  $90^\circ$  (dashed red line). Dashed-dotted blue line: Results from Eq. (14). The inset shows  $\Gamma_e(R_0, \beta)$  (solid line) and  $\Gamma_0(\beta)$  for  $\text{H}_2$  and  $\text{D}_2$  (dashed lines) multiplied by the indicated factors.

Hartree-Fock approximation [21]. The results are shown in Fig. 3. For  $F = 0.05$ , the ionization rate of  $\text{H}_2$  exceeds that of  $\text{D}_2$  by 30%, and this difference grows for weaker fields. For both molecules, the shape of  $\Gamma_0(\beta)$  only slightly differs from that of  $\Gamma_e(R_0, \beta)$ , suggesting that Eq. (13) should work well. From Eq. (13) we obtain

$$\frac{\Gamma_0(\text{H}_2)}{\Gamma_0(\text{D}_2)} \approx e^{(1-1/\sqrt{2})\zeta(\text{H}_2)} \geq 1, \quad (14)$$

where  $\zeta(\text{H}_2) = 0.0017/F^2$ . As can be seen from Fig. 3, Eq. (14) is in good agreement with accurate results. Laser-induced alignment of molecular hydrogen is experimentally very difficult because of the very small polarizability (and its anisotropy) and the large rotational constant. However, the angular dependencies of the tunneling ionization rates of  $\text{H}_2$  [22] and  $\text{D}_2$  [23] have been measured by an ingenious method using the bond-softening channel. In the leading-order approximation for  $F \rightarrow 0$ , the WFAT predicts an anisotropy  $\Gamma_e(R_0, 0^\circ)/\Gamma_e(R_0, 90^\circ) = 1.33$  [9]. The molecular tunneling model of Ref. [24] for the same ratio gives 1.17. In the present calculations we obtain  $\Gamma_0(0^\circ)/\Gamma_0(90^\circ) = 1.44$  and 1.40 for  $\text{H}_2$  and  $\text{D}_2$ , respectively. This is consistent with the value of 1.42 obtained by extrapolating the results of Ref. [22] to zero intensity. Moreover, our prediction that  $\text{H}_2$  has a larger anisotropy than  $\text{D}_2$  for the same intensity agrees with the results of Ref. [23], although the difference between the two isotopes observed in this experiment is within error bars.

The results for a static electric field discussed above remain valid for time-dependent laser fields in the adiabatic regime with respect to electronic motion, that is, for sufficiently low frequency,  $\omega \ll F^2/\mathcal{A}^4(R_0)$ , at a given intensity [25]. In addition, the validity of Eqs. (13) and (14) obtained from Eq. (11) requires  $F \ll F_c$  [8]. The adiabaticity with respect to vibrational motion is not required, as long as transitions to other vibrational states can be neglected. This can be seen from the very structure of the nuclear wave function (8b). The averaging of  $\Gamma_v(\beta)$  over orientations may produce a further departure of observable rates  $\Gamma_{vnM}$  from a bare electronic characteristic  $\Gamma_e(R_0, \beta)$ . This depends on experimental details of creating a rotational wave packet (7) and can be also affected

by statistical distribution in an initial ensemble of molecules. The effect, however, should not be as strong as for averaging over vibrational motion.

To summarize, taking into account nuclear motion can essentially modify the observable tunneling ionization rate of a molecule with respect to a bare electronic rate calculated for fixed nuclei. The difference is controlled by the parameter  $\zeta$  [see Eq. (13)] and becomes more pronounced for weaker fields.

This parameter depends on the reduced mass of the atoms, which results in an isotope effect for the rate of tunneling ionization.

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