Characterization of Molecular Breakup by Super-Intense, Femtosecond XUV Laser Pulses

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We study the breakup of H_2^+ exposed to super-intense, femtosecond laser pulses with frequencies greater than that corresponding to the ionization potential. By solving the time-dependent Schrödinger equation in an extensive field parameter range, it is revealed that highly nonresonant dissociation channels can dominate over ionization. By considering field-dressed Born-Oppenheimer potential energy curves in the reference frame following a free electron in the field, we propose a simple physical model that characterizes this dissociation mechanism. The model is used to predict control of vibrational excitation, magnitude of the dissociation yields, and nuclear kinetic energy release spectra. Finally, the joint energy spectrum for the ionization process illustrates the energy sharing between the electron and the nuclei and the correlation between ionization and dissociation processes.

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Molecular breakup processes induced by strong light-matter interactions are of fundamental interest. Molecular-specific ionization phenomena include charge-resonance-enhanced ionization [1], subcycle multiple ionization bursts [2], electron-nuclei energy-sharing in above-threshold dissociative ionization [3], above-threshold Coulomb explosion [4], and interplay between multiphoton and tunneling ionization [5]. Dissociation phenomena include above-threshold dissociation [6], bond-softening [7], bond-hardening [8, 9], and rescattering induced dissociation [10]. All these processes were discovered in the low-frequency regime where absorption of several photons is needed to reach a breakup channel. Depending on the laser parameters, either dissociation or ionization dominates [11–13].

With advancements in light-source technology, extreme-ultraviolet (XUV) laser pulses of femto- and subfemtosecond duration are now produced from high-order harmonic [14–17] or free-electron lasers [18, 19]. New focusing techniques [20–22] led to XUV femtosecond (fs) pulses with peak intensities \(I \geq 4 \times 10^{17} \text{W/cm}^2\) [23]. Intense XUV pulses were, e.g., applied on rare gases to study sequential versus non-sequential multiple ionization [24, 25], and creation of charge states up to 21 in Xe was observed [21]. For molecules, experiments on HeH^+ [26] and N_2 [27] provided benchmark data for theory. The high-frequency regime is defined in this work as the regime where one-photon ionization is allowed. Since the photon resonance is much closer to the threshold for ionization than for dissociation, ionization is at first glance expected to dominate.

Our goal in this work is to characterize some generic effects of breakup of molecules in the regime of super-intense, high-frequency, fs pulses, supplementing the phenomena known from the low-frequency regime, and adding new insight to the general field of strong laser-matter interaction. The characteristics we find include (a) even with full inclusion of nuclear dynamics, stabilization against ionization occurs, i.e., the ionization yield does not necessarily increase with intensity [28, 29]; (b) a new general mechanism by which dissociation, in contrast to the expectation from energy considerations, completely dominates over ionization; (c) control over the vibrational distribution, dissociation yield, and nuclear kinetic energy release (NKER) spectra by the parameters of the laser pulse; (d) insight into the energy sharing between electronic and nuclear degrees of freedom, as displayed by the joint energy spectrum (JES).

For calculational convenience we consider H_2^+. Indeed, many of the mentioned low-frequency processes were first discovered in H_2^+ and later observed in more complex molecules (see, e.g., [3, 30]). The model includes the dimension aligned with the linearly polarized laser pulse. The TDSE reads (atomic units are used throughout),

\[i \frac{\partial}{\partial t} \Psi(x, R, t) = [T_e + T_N + V_{en}(x, R) + V_I(t)] \Psi(x, R, t)\]  

with \(x\) the electronic coordinate measured with respect to the center of mass of the nuclei, \(R\) the internuclear distance, \(T_e = -(1/2\mu)\nabla^2/\nabla x^2\), \(T_N = -(1/m_p)\nabla^2/\nabla R^2\), \(V_{en}(x, R) = -1/\sqrt{(x - R/2)^2 + a(R)} - 1/\sqrt{(x + R/2)^2 + a(R)} + 1/R\), and \(V_I(t) = -i\beta A(t) / \nabla x\), with \(m_p\) the proton mass, \(\mu = 2m_p/(2m_p + 1)\), \(\beta = (m_p + 1)/m_p\), and \(a(R)\) the softening parameter producing the exact 1s\(\sigma_g\) Born-Oppenheimer (BO) curve [3]. We use vector potentials \(A(t) = (F_0/\omega) g(t) \cos \omega t\), with the field amplitude \(F_0\) related to the peak intensity by \(I = F_0^2\). The envelope is \(g(t) = \exp(-4 \ln(2) t^2/\tau^2)\), with \(\tau\) the full width at half maximum, and the number of cycles \(N_c\) defined by \(\tau = 4\pi \sqrt{2N_c}/\omega\). All pulses considered satisfy the non-relativistic criteria \(2U_p/c^2 \ll 1\) [31, 32] and the dipole condition \(U_p/2\omega c \ll 1\) [33], with \(U_p = F_0^2/4\omega^2\) the ponderomotive potential.
We propagate (1) with a time step of $\Delta t = 0.005$ [34]. The box size is defined as $|x| \leq 100$ and $R \leq 80$ with grid spacings $\Delta x = 0.391$ and $\Delta R = 0.0781$. By imaginary-time propagation we obtain the ground state $\Psi_0(x, R)$ with energy $E_0 = -0.5973$, equilibrium internuclear distance $R_0 = \langle \Psi_0 | R | \Psi_0 \rangle = 2.064$, dissociation limit $E_d = -0.5$, and ionization potential 1.1 at $R_0$. For the real-time propagation, complex absorbing potentials remove the outgoing flux.

Figure 1(a) shows the ionization probability $P_{\text{ion}}$ of $H_2^+$ resulting from laser pulses with $\tau = 11.1$ fs, $\omega = 2.278$, and different $F_0$. $P_{\text{ion}}$ increases with $F_0$ for lower amplitudes until it reaches a maximum at $F_0 \approx 6.2$, whereafter it decreases. The latter behavior indicates stabilization with respect to ionization [29]. Figure 1(b) presents the dissociation probability $P_{\text{dis}}$. For the lower values of $F_0$, we observe no dissociation as expected from perturbation theory. At $F_0 \approx 9$, dissociation sets in, increasing with $F_0$ until it “saturates” at $F_0 \approx 15$.

To obtain a complete picture, Figs. 1(c) and 1(d) present $P_{\text{ion}}$ and $P_{\text{dis}}$, as functions of $F_0$ and $\omega^2$. For fixed $\omega$ the suppression of ionization for large $F_0$ is evident [Fig. 1(c)]. Lobes corresponding to large $P_{\text{ion}}$ are seen emanating from the origin. By using a version of high-frequency Floquet theory (HFFT) [35] here generalized to include nuclear motion, we find that the lobes in Fig. 1(c) are along $\alpha_0 = F_0/\omega^2$ corresponding to peaks in the ionization rates. The dashed line traces the largest HFFT rate, in reasonable agreement with the TDSE result. In the case of $P_{\text{dis}}$ in Fig. 1(d), we observe appreciable dissociation for pulses with $\alpha_0 \geq 2.41$ indicated by the dashed line. In this regime, $P_{\text{ion}} + P_{\text{dis}} = 1$, implying unity probability for breakup. The remnants of the lobes from ionization are present in Fig. 1(d), an indication of the different time scales for ionization and dissociation processes: ionization occurs first, and what is not ionized, dissociates after the pulse. The physics of the dashed line will be explained later.

In order to elucidate the origin of the onset of dissociation in Fig. 1, it is instructive to transform Eq. (1) into the Kramers-Henneberger (KH) frame [36, 37], where the complete laser-molecule interaction is contained in the modified electron-nuclei interaction $V_{\text{eN}}(x + \alpha(t), R)$, with the quiver motion $\alpha(t) = \int_0^t A(t')dt'$. In the case of a monochromatic laser field, the Floquet ansatz for the wave packet (WP) and the Fourier expansion of $V_{\text{eN}}(x + \alpha(t), R)$ results in a coupled set of equations. For large $\omega$, effectively only the zeroth order Fourier component, $V_0(x, R, \alpha_0) = (\omega/2\pi) \int_0^{2\pi/\omega} V_{\text{eN}}(x + \alpha(t), R) dt$, remains, resulting in the structure equation [29, 37]

$$[H_e(x, R; \alpha_0) + T_N] u(x, R; \alpha_0) = W(\alpha_0) u(x, R; \alpha_0),$$

with $H_e(x, R; \alpha_0) = T_e + V_0(x, R; \alpha_0)$ the field-dressed electronic Hamiltonian. For a given $\alpha_0$, Eq. (2) is solved in the BO-approximation, yielding the field-dressed BO curves $E_{\text{fBO}}(R, \alpha_0)$ and the dressed energies $W_{\text{f}}(\alpha_0)$ [38], with the indices $i = 1, 2, \ldots$ and $\nu = 0, 1, \ldots$ denoting the electronic and vibrational states. To treat pulsed laser fields, we let the maximal quiver amplitude vary with the field envelope, $\alpha_0 \rightarrow \alpha_0(t) \equiv \alpha_0 g(t)$. The lowest
BO curve is plotted in Fig. 2 for \( \alpha_0(t) = 0, 1.87 \) and 3.25. With increasing \( \alpha_0(t) \), the BO curve is shifted upwards in energy, towards greater \( R \), and becomes gradually shallower. The latter implies that the dressed vibrational time scale, \( T_v(\alpha_0(t)) \equiv 2\pi/|W_{1,1}(\alpha_0(t)) − W_{1,0}(\alpha_0(t))| \), increases with \( \alpha_0(t) \).

We now present a qualitative model of the dissociation mechanism. The validity of the model is determined later by TDSE results. Let \( \tau \) be the time scale for the turn-on (and -off) of the laser pulse, and \( \alpha_{0,\text{th}} \) the quiver amplitude satisfying \( W_{1,0}(\alpha_{0,\text{th}}) = E_d \), i.e., when the dressed ground state equals the dissociation limit [inset of Fig. 1(d)]. Provided the pulse satisfies (i) \( \alpha_0 > \alpha_{0,\text{th}} \), (ii) \( T_v(0) \sim \tau \), and (iii) \( \tau < T_v(\alpha_0) \), the dissociation process occurs as follows [Fig. 2(b)]. During the turn-on of the pulse, (ii) ensures the population to follow the field-dressed ground state adiabatically. At the field maximum, (i) implies that the bound WP populates dressed eigenstates with energies greater than \( E_d \). Due to (iii), the turn-off of the pulse can be considered sudden, and the nuclear WP does not feel the fast change of the electronic potential, leaving its position and energy unchanged. After the pulse, the nuclear WP is trapped above \( E_d \), resulting in dissociation via the field-free electronic ground state, with NKER given by \( E_N(\alpha_0) = W_{1,0}(\alpha_0) − E_d \).

The laser field regime for which \( P_{\text{dis}} \) is nonzero in Fig. 1(d) satisfies (i)-(iii). For \( H_2^+ \), we have \( \alpha_{0,\text{th}} = 2.41 \), \( T_v(0) = 15.2 \) fs and \( T_v(\alpha_{0,\text{th}}) = 41.7 \) fs. In Fig. 1(d), the onset of dissociation is indeed around the dashed line corresponding to \( \alpha_0 = \alpha_{0,\text{th}} \). This agreement supports the physical picture of the model. The frequency range \( \omega^2 = 2 - 8 \) in Fig. 1(d) where \( P_{\text{dis}} \) is nonzero corresponds to \( \tau = 9.0 - 17.9 \) fs, which fulfills condition (ii) at least approximately. For \( \alpha_0 > \alpha_{0,\text{th}} \), we have \( \tau < T_v(\alpha_{0,\text{th}}) < T_v(\alpha_0) \), and condition (iii) is satisfied as well. Ionization occurs throughout the whole duration of the pulse due to higher-order Floquet components in \( V_N(x + \alpha(t), R) \). In the stabilization regime, ionization is greatly suppressed, leaving the population trapped above \( E_d \) to dissociate. This explains \( P_{\text{ion}} + P_{\text{dis}} = 1 \) in the parameter regime of Fig. 1(d) where \( P_{\text{dis}} \) is nonzero. The detection of dissociative fragments with energy \( E_N(\alpha_0) \) is thus a direct confirmation of dynamics in the stabilization regime.

Figures 2(a) and (c) illustrate the cases for which (i) is satisfied, but (ii) and (iii) are not. In the case where the pulse duration is short, \( \tau < T_v(0) \), the perturbation can be considered sudden, and the initial population is unaffected, resulting in no dissociation [Fig. 2(a)]. For long pulses \( \tau \gg T_v(\alpha_0) \), the adiabatic approximation is accurate, and the initial state follows the dressed ground state throughout the whole pulse resulting in a final population similar to the initial population [Fig. 2(c)]. For the case where (ii) and (iii) are satisfied, but with \( \alpha_0 < \alpha_{0,\text{th}} \), no dissociation occurs. Instead, an excited vibrational WP is created containing field-free vibrational states with quantum numbers \( \nu \) satisfying \( W_{1,\nu}(0) = W_{1,0}(\alpha_0) \) [Fig. 2(e)].

The qualitative physical model is validated by TDSE calculations. Figure 3 shows \( P_{\text{ion}}, P_{\text{dis}} \) and \( P_{\text{ion}} + P_{\text{dis}} \) for \( \omega = 2.278 \) and different pulse durations \( N_c \) as a function of \( F_0 \). While \( P_{\text{ion}} \) in Fig. 3(a) increases with \( N_c \) for a fixed \( F_0 \), \( P_{\text{dis}} \) in Fig. 3(b) behaves differently. For \( N_c = 20 \) \((\tau = 2.2 \text{ fs} < T_v(0) = 15.2 \text{ fs})\), there is minimal dissociation in accordance with the model predictions [Fig. 2(a)]. For a given \( F_0 \), \( P_{\text{dis}} \) increases with \( N_c \) until it reaches a maximal value at \( N_c \approx 60 - 80 \), whereafter \( P_{\text{dis}} \) decreases due to adiabaticity. For pulses in the range \( N_c = 60 - 140 \) and the larger \( F_0 \), the saturation condition \( P_{\text{ion}} + P_{\text{dis}} = 1 \) [Fig. 3(c)] contributes to the decrease in \( P_{\text{dis}} \). For these \( N_c \)’s the onset of dissociation is approximately at the dotted line corresponding to \( \alpha_0 = \alpha_{0,\text{th}} \), consistent with Fig. 2(b). For the pulse with \( N_c = 200 \) \((\tau = 22 \text{ fs})\), \( P_{\text{dis}} \) is less than 0.3, consistent with the picture that the population follows the field-dressed states adiabatically and dissociation is suppressed [Fig. 2(c)].

We have thus validated the physical mechanism for dissociation in Fig. 2, which implies that by varying the two laser parameters, \( \tau \) and \( \alpha_0 \), the vibrational populations, dissociation yields, and NKER can be controlled. This prediction is confirmed by the TDSE results of Fig. 4, which shows the bound vibrational populations at the end of the pulse, and the NKER spectra, for \( \omega = 2.278 \), \( F_0 = 1 - 17.9 \), and \( N_c = 10 - 500 \). For \( N_c = 10 \), the \( \nu = 0 \) level is most populated, except at \( F_0 \gtrsim 14 \) where strong non-adiabatic couplings excite some vibrations. No dissociation occurs, in agreement with Fig. 2(a). For \( N_c = 140 \), the vibrational population and the NKER spectra follow approximately the dotted line corresponding to \( W_{1,0}(\alpha_0) = E_d \). This is in agreement with Figs. 2(b), 2(e), and the accompanying discussions, where we argued that the energy of the final
The JES after the pulse (\(t = \infty\)) shows four distinct peaks, denoted by \(Q_1, \ldots, Q_4\). The physical picture in Fig. 2(b) explains these. At \(t = -6.7\) fs, the Stark shift \(\Delta(t) = W_{1,0}(\alpha_0(t)) - E_0\) is negligible, producing \(Q_1\) in the JES along the line corresponding to the one-photon resonance \(E_\omega + E_N = E_0 + \omega\). At \(t = 0\), according to the model, the nuclear WP has the largest Stark shift \(\Delta(0) = 0.09732\), and \(Q_2\) emerges along the shifted one-photon resonance \(E_\omega + E_N = W_{1,0}(\alpha_0) + \omega\). The internuclear distance of the WP is \(\langle R(t) \rangle = 3.2\), resulting in \(Q_2\) having \(E_N \approx 1/3.2 = 0.32\). During the turn-off of the pulse, the physical model predicts that the population stays above \(E_\omega\), with the possibility of dissociation towards larger \(R\). Indeed, in the JES at \(t = 6.7\), \(Q_3\) and \(Q_4\) emerge along the Stark-shifted resonance, with smaller \(E_N\) corresponding to larger \(R\). These results indicate that dissociation and probing of dissociation by ionization can be achieved using a single pulse, with the nuclear WP being promoted to the field-free dissociation continuum during turn-on (pump) and probed through ionization during turn-off. For \(t > 0\), \(Q_1\) stays constant in magnitude and position consistent with the model prediction that no field-free bound states become populated during pulse turn-off. For this reason the dynamic interference effect [40, 41], where ionized WPs with equal continuum energy created during the rising and falling edges of the pulse interfere, is not observed.

In conclusion, we have highlighted some characteristics of the breakup of molecules by interaction with fs pulses in the super-intense, high-frequency regime. Dissociation not only occurs, but can dominate over ionization. This is surprising as in the overlapping of two continua, the transition probability is expected to be largest for the continuum threshold that is closest to the photon resonance. We explained the dissociation by a mechanism wherein the nuclear WP follows the lowest field-dressed BO curve during the pulse turn-on, but stays in the field-free continuum during the turn-off, taking advantage of the different effective vibrational time-scales at different field amplitudes. It was shown that by varying the pulse parameters, control over the vibrational distribution, the dissociation yield and NKER spectrum could be achieved. The occurrence of dissociation is strongly dependent on the suppression of ionization in the strong field regime, and we presented the JES for the ionization process, and related its structures to dynamics taking place at different instants of time during the pulse.

The described dynamics could be experimentally verified by observing the sharp onset of dissociation at \(\alpha_{0,\text{th}}\) by tuning \(I\) with fixed \(\omega\). The pulses considered have \(F_0\) less than one order of magnitude greater than what is presently experimentally available. The demand on intensity could be relaxed by considering excited initial WPs, which require less energy for dissociation.

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