DYNAMICS OF DISSOCIATION VERSUS IONIZATION IN STRONG LASER FIELDS

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MAR 16 1993

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INTRODUCTION

The behavior of isolated atoms in strong radiation fields has been a topic of intensive theoretical and experimental interest over the last decade and has resulted in a cumulative literature rich in new phenomena, unresolved issues, and exciting new predictions for future studies.¹ Recently, some investigations have begun to examine the general behavior of molecules in intense fields, with special relevance placed on strong-field photodissociation dynamics. The fundamental motivation for such studies derives its interest from the ability to control chemical dynamics by external variation of a laser field. The traditional chemical physics approach relied upon the laser frequency as the only external field parameter for achieving state-selectivity but has met with little success. However, current efforts² have focused on examining all aspects of the laser field for achieving control. Although premature in realization, an understanding of the influence of intensity, coherence, phase, and pulse duration and shaping in concert with frequency may ultimately lead to our ability or inability to control chemical dynamics.

The special challenge encountered in studying molecules in strong fields results from the difficulty of sorting out the general behavior of field-induced effects from the details or specifics of the molecular structure. Investigations on atoms^{3,4} clearly demonstrate the importance of the atomic structure in describing the ionization dynamics in the multiphoton regime. Consequently, it is not surprising to expect enhanced structural effects in molecules due to the many internal degrees of freedom as compared to atoms. However, the anticipated complications posed by molecules are in practice quite tractable. In fact, the field-molecule interaction can lead to some surprising insights into the molecular structure.^{5,6} Furthermore, molecular strong-field studies raise some interesting and unique questions concerning the role and interplay of ionization and dissociation, and can result in studies of atoms in unusual circumstances.⁷

In this paper, experimental results are presented which clearly demonstrate the effectiveness that an external field has in altering the dissociation dynamics. The experiment examines the strong-field dissociation dynamics of molecular hydrogen ions and its deuterated isotopes. These studies involve multiphoton excitation in the intensity regime of 10^{11-14} W/cm² with the fundamental and second harmonic of a Nd:YAG or Nd:YLF laser system. Measurements include energy resolved electron and mass spectroscopy which provide useful probes in elucidating the interaction dynamics predicted by existing models. The example discussed in this paper, examines the strong-field dissociation of H_2^+ , HD^+ , and D_2^+ at green (0.5 μ m) and infrared (1 μ m) frequencies. The diatomic

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ions are formed via multiphoton ionization of the neutral precursor which is physically separable from the dissociation process. This study provides the first observation of the dynamics associated with the above threshold dissociation (ATD) process and analogies will be made with the more familiar above threshold ionization (ATI) phenomenon.

EXPERIMENTAL

The photon sources used are well characterized Nd:YAG (1.06 μm , 10 nsec, 10 Hz) and Nd:YLF (1.05 μm , 50 psec, 1 kHz) lasers. Details of these laser systems have been described previously.⁸ The second harmonic is generated using standard nonlinear techniques either in BBO or KD*P crystals. The intensity ranges for 0.532 and 0.527 μm radiations are 8×10^{11} to 9.7×10^{12} W/cm² and 5×10^{12} to 4×10^{13} W/cm², respectively. The apparatus consists of an ultrahigh vacuum chamber equipped with time of flight electron and mass spectrometers. The field-free photoelectron spectrometer has an energy resolution of about 50 meV for 1 eV electrons and an acceptance angle of 1×10^{-3} sr. It is calibrated using the well known ATI spectrum of xenon atoms. The mass spectrometer consists of a 42 cm long drift tube with a series of electric field acceleration plates. Fragment kinetic energy analysis is achieved by applying an uniform extraction field across the laser focus with the laser polarization parallel to the flight tube axis. This produces two nearly symmetric peaks in the time of flight spectrum corresponding to the two velocity components of the same fragment initially directed towards and away from the detector. The kinetic energy of the fragment is simply determined by measuring the arrival time difference between the peaks. The mass spectrometer energy calibration was performed by recording the weak-field photodissociation of chlorine⁹ molecules. The resolution was estimated to be about 100 meV for 1 eV protons. The background pressure in the ultrahigh vacuum system was 1×10^{-9} torr. The data collection system is always operated in the pulse counting regime with count rates much less than one event per laser shot.

RESULTS AND DISCUSSION

Diatomic hydrogen ion represents the "model" molecular system to study strong-field behavior because of its simple and well-known one-electron structure and as a result should provide the foundations for future molecule-field studies. A physically distinct motivation for studying the strong-field behavior of molecules instead of atoms is the presence of dissociative channels. Questions addressing the validity of extending our knowledge on intense field atomic ionization to describe molecular dissociation seem warranted. For example, does high field dissociation proceed in an analogous manner to ATI, that is, via absorption of additional photons beyond the minimum needed to break a molecular bond and are the dynamics similar. We will address these issues through out this paper and contrast the differences between dissociation and ionization.

Dressed-State Model

Photodissociation of a molecule in an intense field can lead to a phenomenon which is analogous to above threshold ionization (ATI) observed in the photoionization of atoms and molecules. The general effect results in the absorption of additional photons beyond the minimum necessary to break the chemical bond and has been dubbed above threshold dissociation (ATD). The traditional weak-field representation of the photodissociation process is shown in Fig. 1(a) for the first two potential curves of the H_2^+ molecule. The vertical arrows show the internuclear separation for the Franck-Condon (FC) favored 1- and 3-photon absorption. Thus, the absorption of additional photons results in the production of a series of hydrogen atoms and protons with kinetic energies differing by $(1/2)\hbar\omega$ or $\hbar\omega$ total energy. Energetically this representation is equivalent to the more standard ATI picture. However, this picture offers no intuitive understanding of the dynamics of the dissociation process nor potential distortion or vibrational shifts in the strong field limit.

Figure 1. Potential diagrams for (a) traditional photodissociation and (b) dressed-state model for 0.5 μm excitation.

In order to obtain a more intuitive feel for this physics it is advantageous to reformulate the problem in a dressed state basis. In this model, the laser field is easily incorporated as a strong perturber to the molecular potentials. A number of papers^{10,11,12} have been published concerning this approach and the readers are referred to them for more details. A finite number of molecule-field states $|d\rangle$ are constructed as the product of the molecular state $|\phi\rangle$ and the photon-number state $|N-n\rangle$, where $\phi \equiv 1s\sigma_g$ or $2p\sigma_u$, $n = 0, \pm 1, \pm 2, \dots$ and N is the total photon number. Therefore, the weak-field two state picture shown in Fig. 1(a) can be transformed into a infinite series of paired dressed states, as illustrated in Fig. 1(b) for excitation with 2.3 eV photons. The photodissociation is represented by passage along the potential curves to large internuclear separation at the limits corresponding to 1-photon ($|u,1\rangle$), 2-photon ($|g,2\rangle$), and 3-photon ($|u,3\rangle$) absorption (indicated by the horizontal arrows). The potential curve crossings occur at internuclear separations corresponding to multiples of the photon energy (dotted diabatic curves). Since the ground and repulsive ionic state have gerade and ungerade symmetry, only level crossings with an odd difference in photon number can interact because of parity conservation. Note, that the crossings in Fig. 1(b) occur at the odd photon absorption in Fig. 1(a) at the FC favored internuclear separation. Consequently, the molecular potential couple and distort (solid curves in Fig. 1(b)) via the strong dipole interaction. Any populated vibrational levels within the gap will become unstable and predissociate. The number of unstable vibrational states increases with larger field strength since the avoid-crossing gap continues to open. Bond-softening occurs when the field-induced dissociation is at the 1-photon crossing. Furthermore, ATD can occur due to level crossings at higher order couplings. For example, 3-photon dissociation can diabatically dissociate along curve $|u,3\rangle$ or adiabatic 2-photon dissociation along path $|g,2\rangle$. Consequently, a *net even number* of photon dissociation can also occur due to the absorption and stimulated emission of photons. Thus, we see that the dressed-state picture renders an intuitive feel for predicting the dissociation dynamics and we will examine this connection throughout the experimental results.

H_2^+ dissociation: 0.53 μm excitation

Figure 2 shows a typical time of flight mass spectrum resulting from the 0.532 μm nonresonant MPI of H_2 and D_2 molecules at an intensity of $9.7 \times 10^{12} \text{ W/cm}^2$. The molecular ions formed by the

Figure 2: Time of flight mass spectrum resulting from 0.5 μm excitation of (a) D_2^+ and (b) H_2^+ at $9.7 \times 10^{12} \text{ W/cm}^2$. Peak labels 1, 2, and 3 correspond to the number of absorbed photons leading to dissociation.

MPI of the neutral molecule produces the peak at long arrival times in the spectra of Fig. 2. The peaks at the short arrival times are the atomic fragments (protons and deuterons) formed via dissociation of the molecular ion. The symmetric substructure results from the forward and backward directed fragments and the splitting is proportional to their initial kinetic energy, as discussed in Experimental Section. The three fragment peaks are labeled according to the number of photons being absorbed by molecular ions leading to dissociation. Figure 3 shows the proton spectrum converted to total kinetic energy. The tick marks indicate the proton's kinetic energy for different initial unperturbed vibrational energy. Accordingly, at low intensity (Fig. 3(b)) most H_2^+ ions are dissociated from the $v_{\text{avg}}^+ = 5$ vibrational level following the absorption of a single photon via bond-softening. The maximum of this peak is observed to shift from $v^+ = 5$ to $v^+ = 4$ at higher intensities, as seen in Fig. 3(a). At higher intensity another peak emerges corresponding to two photon dissociation (peak at $2h\nu$ in Fig. 3) via $v_{\text{avg}}^+ = 2$. Consequently, the spacing of the 1- and 2-photon peak is less than a photon total energy due to differing initial vibrational states and is intensity dependent. *Thus, an immediate difference is apparent between ATD and ATI which is related to the fact that different order odd photon processes have their origin in different initial states or in the dressed state language, at different internuclear separation.* At still higher intensities a third peak appears one photon total energy away from the second peak corresponding to a three photon dissociation. It is clear from the dressed state model that the peak separation for any pair of Floquet states is one photon total energy since dissociation occurred from the same initial vibrational state. The same general behavior is observed for D_2 and HD molecules¹³. Similar kinetic energy shifts are observed in the 0.527 μm (psec) spectra over an intensity range of 5×10^{12} to $4 \times 10^{13} \text{ W/cm}^2$. Shown in Fig. 4(b) is the dissociation fraction F^{H} and F^{D} as a function of laser intensity for H_2^+ and D_2^+ , respectively and where $F^{\text{H}} = [\text{H}^+]/(\text{H}^+ + \text{H}_2^+)$ and $F^{\text{D}} = [\text{D}^+]/(\text{D}^+ + \text{D}_2^+)$. Note, that the dissociation fraction for each molecule increases with intensity and that F^{D} is larger than F^{H} at all intensities shown.

Figure 3: Total kinetic energy (twice the measured proton kinetic energy) spectrum of H^+ fragments resulting from $0.532 \mu m$ dissociation of H_2^+ at (a) 9.7 TW/cm^2 and (b) 2.7 TW/cm^2 . The tick marks show the position of the unperturbed ionic ground state's vibrational levels in one (left) and two (right) photon dissociation processes.

Figure 4: A plot of the (a) ATD branching ratios, R , and (b) dissociation fraction, F , for H_2^+ and D_2^+ as a function of $0.53 \mu m$ intensity.

We have carefully examined the photodissociation of H_2^+ molecules by recording the proton kinetic energy spectra and the H_2 photoelectron spectra simultaneously as a function of laser intensity. Our results verify that the mechanism responsible for H^+ and D^+ formation is the photodissociation of the parent molecular ion. It appears that the amount of H^+ produced is proportional to that of photoelectrons due to (7+1) molecular ionization. In addition, energy conservation implies that seven second harmonic photons of YAG(YLF) can populate only up to $v^+ = 3(4)$ vibrational levels in the ionic ground state, whereas the lowest intensity data have proton energies peaked at $v^+ = 5$. Thus, we conclude that the H^+ peak labeled 1 in Fig. 2 (0.7 eV total energy) is due to the eight photon ionization of H_2 followed by one photon dissociation of H_2^+ .

The dressed-state model predicts that as the field strength increases the fragments kinetic energy will decrease as lower vibrational levels become unstable in the field. Physically, the distort potential corresponds to a softened potential at the outer turning points which decreases the vibrational frequency with increasing field strength. In the current experiment, we observed apparent negative shifts in proton kinetic energies with increasing laser intensity with both 532 nm (ns), as shown in Fig. 3, and 527 nm (ps) pulses. The maximum population of the 1-photon proton kinetic energy peak shifts by ~ 200 meV over the entire intensity range. Likewise, the maximum of the ionic vibrational state distribution in the photoelectron spectroscopy (PES) changes from high to low vibrational levels in a similar intensity range, although the shift in the amplitude of the vibrational distributions is independent of the molecule studied. Thus, the bond-softening mechanism to first order is predominantly an *electronic coupling* and not vibrational. However, we could not observe any commensurate positive energy shifts in the PES peak positions with increasing intensity although a broadening is clearly evident. An asymmetric broadening in our PES could result from spatial or temporal averaging in our experiment masking a "pure" energy shift but the sign of the observed broadening is opposite to that predicted by a bond-softening model.¹³ This inconsistency could imply a shortcoming in our numerical method or an indication that the PES probes the potential at a low intensity before significant potential distortion has occurred. Another contributing factor to the peak width could result via strong-field rotational pumping with increasing laser intensity, causing the photoelectron peaks to broaden towards lower energies.

Referring to Fig. 4(a), the ratio ($R_{2+3,1}$) of dissociation, labeled ATD/1, via the sum of the 2- and 3-photon (ATD) versus the 1-photon channels (peaks labeled 1, 2, and 3 in Fig. 2) for H_2^+ (D_2^+) changes from 7% to 25% (18% to 42%) as the intensity is increased to 1.3×10^{13} W/cm². Thus, the total amount of ATD increases with intensity in a way analogous to the intensity dependence of ATI. The difference observed in the dissociation fraction, F , for H_2^+ and D_2^+ at any given intensity is a vibrational effect which results in a better FC match in the unperturbed vibrational levels for D_2^+ at low intensity. Consequently, D_2^+ dissociates more readily than H_2^+ at this wavelength.

Further insight into the physically distinct dynamics between strong-field dissociation and ionization becomes evident by examining the ATD branching ratio R_{32} (labeled 3/2) for 3- versus 2-photon dissociation of H_2^+ in Fig. 4(a). This ratio decreases from 16.5 to 6.5% over the entire increasing intensity range. Thus, the branching ratio among the high energy fragment channels favors the production of low energy protons with increasing intensity, in sharp contrast to the behavior of ATI. Furthermore, the D_2^+ ratio R_{32} is smaller than H_2^+ and relatively constant ($\sim 6\%$) over the entire intensity range. These branching ratio effects can be all understood with the dressed-state model. Physically, the increasing laser intensity corresponds to a larger avoided-crossing gap resulting in a decrease in the 3-photon diabatic transition rate while favoring 2-photon adiabatic passage (3-photon absorption, 1-photon emission). Since H_2^+ and D_2^+ have a relatively large difference in vibrational frequencies due to their different masses, therefore at low intensities where the gap is not large enough to completely shut off the 3-photon diabatic path as compared to H_2^+ due to the fact that a smaller vibrational frequency implies more adiabatic motion. Specifically, $R_{32}(H) > R_{32}(D)$ at low intensity. As the gap continues to widen, one should also observe the ratio R_{32} to decrease for both H_2^+ and D_2^+ molecules. Such behavior is clearly demonstrated in our experiments. Figure 4(a) also shows for comparison the results of the calculated fragment R_{32}^{LZ} ratios (labeled LZ) predicted by a simple Landau-Zener (LZ) theory.¹⁴ The theory predicts well the general behavior of the ratios as the light intensity changes. However, the calculated ratios are approximately three times larger than the experimental values which could imply that the degree of deformation of

the calculated potential curves is beyond the limits of applicability of simple LZ theory or a short coming of our numerical Floquet analysis.

H_2^+ dissociation: 1.06 μm excitation

Figure 5 is a plot of a typical time of flight mass spectrum resulting from dissociation of H_2 with 1.06 μm radiation. The dissociation fraction F^H for H_2^+ molecules, where $F^H = [H^+/(H^+ + H_2^+)]$, changes from 0.25 to 0.43 for the entire dynamic range of laser intensities $2.3 - 4.6 \times 10^{12} W/cm^2$. The peaks labeled *a* and *b* correspond to a total proton kinetic energy of 0.49 and 2.88 eV, respectively, and are assignable to the dissociation of H_2^+ following absorption of 2- and 4-photons. For both processes, the measured proton kinetic energies give an initial vibrational distribution with an average value of $v_{avg} = 3$. *No odd number photon dissociation processes are observed in the present experiment at all intensities studied.* Furthermore, the ATD ratio, $R_{42}(H)$, for the 4-photon and 2-photon dissociation channels are intensity dependent and increase from 0.11 to 0.14 with increasing intensity.

Figure 5: Time of flight mass spectrum of H_2 resulting from 1.06 μm excitation at an intensity of 4.6 TW/cm^2 . The peak labelled 2 and 4 are the forward velocity components formed via the 2- and 4-photon dissociation of H_2^+ . The insert is an expanded view of the proton distribution.

The photoelectron spectra of H_2 taken at two different laser intensities with 1.06 μm radiation is shown in Fig. 6. The tick marks indicate the number (order) of photons absorbed by the H_2 molecule. Since the IP of H_2 is 15.42 eV, a minimum of 14 photons are required to ionize. The low-intensity photoelectron spectrum plotted in Fig. 6(b) can be characterized as a broad distribution centered at 5 eV which exhibits suppression of low energy electrons up to 2.8 eV and a maximum kinetic energy at 15 eV. However, the spectrum still shows structure which is vibrationally resolvable and assignable to the first four vibrational levels of the H_2^+ ground state. As the laser intensity is increased in Fig. 6(a), the photoelectron distribution tends to become structureless and shifted towards higher kinetic energy (peaked at 7.5 eV). Likewise, the low-energy electron peaks associated with the 16 and 17-photon ionization processes show additional suppression. The MPI and dissociation of H_2 at 1.06 μm have been studied by Zavriyev *et al.*¹³ using 100 ps pulses. Their photoelectron distributions show a similar shape to those shown in Fig. 6 but with no resolvable structure. The difference between the two experiments probably reflects the change in saturation intensity *via* the pulse width dynamics which unfortunately results in an inability to extract any

detailed information from the photoelectron spectrum. Consequently, long pulse studies can provide a useful look at the mapping of the molecule-field interaction. The mechanism resulting in the suppression of low energy electrons in Fig. 6 is not understood. The suppression is analogous in its behavior to "channel closure" observed in rare gas atoms¹⁵ which is caused by the increasing binding energy of the atom in the field and proportional to the ponderomotive potential. However, the ponderomotive potential available at our highest intensity is only 0.5 eV, which is obviously not enough shift to account for our degree of electron suppression.

Figure 6: Photoelectron spectra of H_2 molecule taken at $1.06 \mu\text{m}$ intensities of (a) 2.3 TW/cm^2 and (b) 4.1 TW/cm^2 . The tick marks indicate the number of absorbed photons above the H_2 ground state.

The proton kinetic energy spectrum of Fig. 5 shows no evidence that 1-photon dissociation is occurring at $1 \mu\text{m}$. Thus, bond-softening is not playing any significant role in the dissociation dynamics at this wavelength, although it dominates at $0.53 \mu\text{m}$. This result is not difficult to understand, since the 1-photon curve crossing associated with the dressed-state picture illustrated in Fig. 7, occurs at large enough internuclear separation (~ 5 bohr) which necessitates the need for significant population in the higher vibrational ($v=7$) levels of the molecule. However, our photoelectron spectrum analysis shows that only the lower vibrational states are populated via the MPI process.

As discussed above for $0.53 \mu\text{m}$ excitation, the ATD ratio (R_{32}) decreases with increasing laser intensity, that is, dissociation occurs more readily through the lower-order channels as the laser intensity increases. This behavior is just the opposite at $1 \mu\text{m}$, where the ATD ratio (R_{42}) increases with rising intensity. The $1 \mu\text{m}$ behavior is analogous to the more familiar intensity dependence observed in ATI but with a major difference being the formation of fragments with energy spacing $2\hbar\omega$. Although this strong wavelength dependence results in such different dynamics, the interpretation is all consistent with the dressed-state model discussed above. According to the dressed-molecular states model, illustrated in Fig. 7 for $1 \mu\text{m}$ radiation, the diabatic dressed-states $|\mu,3\rangle$ and $|\mu,5\rangle$ cross the ground state $|g,0\rangle$ at 3.6 and 3.1 bohr, respectively. Furthermore, the intersections are electric-dipole allowed ($g \rightarrow u$) which result in an intensity-dependent avoid-crossing. The gap also occurs in the potential region which are vibrational populated by the MPI process. The same argument applies to the diabatic crossing (dashed line in Fig. 7) between the $|g,2\rangle$ and $|\mu,3\rangle$ curves. The 1-photon interaction results in an avoid-crossing which is more strongly laser dependent than the higher order processes. In addition, the bond-softening gap intensity dependence is greater at $1 \mu\text{m}$ than $0.5 \mu\text{m}$ because of the increasing electric dipole moment at larger internuclear separation. Consequently, the high intensity and longer wavelength causes the 1-photon gap to become so large (solid lines) that adiabatic passage dominates (3-photon absorption, 1-photon emission) resulting in only 2-photon dissociation. The same logic then follows for any dressed-state pairs, *i.e.* $|g,4\rangle$ and $|\mu,5\rangle$, which always result in even-photon dissociation. This is physically manifested in our proton kinetic energy spectrum as the series of peaks separated by the 2-photon

(total) energy. Likewise, the increase in the ATD ratio (R_{42}) results from the fact that each proton peak in the series occurs via a different order photon process. Thus, the dynamics, as manifested by the increasing ATD ratio, results in more efficient production of higher-order protons with increasing intensity. This has similar dynamics compared with production of high energy ATI electrons.

Figure 7: The dressed-state H_2^+ molecular potentials for a $1.06 \mu\text{m}$ laser field. The dotted lines show the diabatic crossings which distort (full lines) due to the molecule-field interaction.

CONCLUSIONS

In conclusion, the experiments presented above clearly demonstrate the effectiveness that an external field has in altering the dissociation dynamics. Furthermore, the strong-field dissociation dynamics manifest very distinctive behavior compared to ionization. The dressed-state picture has proven to be a potent intuitive model for understanding the high-field dynamics and should provide a valuable predictive model for future studies.

ACKNOWLEDGMENTS

We would like to thank Dr. F. Mies and Prof. A. Giusti-Suzor for their useful discussions. This research was carried out at Brookhaven National Laboratory under contract No. DE-AC02-76CH00016 with the U.S. Department of Energy and supported by its Division of Chemical Sciences, Office of Basic Energy Sciences.

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