Structure and Dynamics of Atoms, Ions, Molecules, and Surfaces: Molecular Dynamics with Ion and Laser Beams

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The goal of this part of the JRML program is to study the different mechanisms for molecular dissociation initiated by ultrashort intense laser pulses or following fast or slow collisions. To that end we typically use molecular ion beams as the subject of our studies.

Below we give a couple of examples of our recent work[1].

Above threshold Coulomb explosion (ATCE) of H_2^+ in intense fs laser pulses, *Itzik Ben-Itzhak, Pengqian Wang, A. Max Sayler, Kevin D. Carnes, and Brett D. Esry*

This work is the direct result of close collaboration between theory and experiment. Theory suggested a model that was simpler to test experimentally than from first principles. As a consequence, a new mechanism of molecular dissociation was uncovered: above threshold Coulomb explosion.

In talks on the intense field ionization of H_2^+, it is common to see a plot that shows the lowest two H_2^+ Born-Oppenheimer potentials, $1s\sigma_g$ and $2p\sigma_u$, along with the ionization potential $1/R$. The $2p\sigma_u$ state is shifted downward by the photon energy in accord with the Floquet picture [1], but ionizing transitions are indicated by a stack of arrows representing photons. Not liking this mixture of representations — the Floquet picture already includes the photons, after all — we asked ourselves how to describe dissociation and ionization within the same conceptual framework while retaining the benefits of the Floquet picture for dissociation. The simplest answer was to simply include the ionization potentials in the Floquet picture, shifting $1/R$ by integer multiples of the photon energy. These curves, along with the dissociation curves, are shown in the left hand side of Fig. 1(b). Such curves really represent the lower edge of the ionization continuum and assume the ionized electron has zero total energy. But, where they cross the appropriately shifted $1s\sigma_g$ and $2p\sigma_u$ potentials, that ionization channel opens. At low laser intensities near the ionization appearance intensity, these channel openings should translate into structure in the nuclear kinetic energy release (KER) spectrum. No such structure had been reported experimentally, however.

So, with this new picture in mind, we re-analyzed the low-intensity ionization KER data available from our application of the intensity difference spectrum (IDS) method [Pub. #12]. Typical data is shown in the right hand side of Fig. 1 in (a) and (b) [Pub. #19]. The bars at the top of each panel show the peak positions expected from our model and the fit produced by placing a Gaussian at these positions allowing for intensity-induced shifts. Panels (a) and (b) were fit simultaneously using constraints suggested by our model that reduced the total number of free parameters by a factor of two.

Our model also allows us to predict the angular distribution, since we know the number of photons leading to each peak. The idea is that each photon absorbed will contribute a factor of $\cos^2\theta$, since primarily parallel transitions are involved. Thus, for the 800 nm data shown in panels (a) and (b) that require 12-13 photons to ionize, the angular distribution should be $\cos^{24}\theta$ or $\cos^{26}\theta$. Panel (d) shows a $\cos^{2n}\theta$ fit to the data in (a) and (b) with the result that n=12.5±1.0, in good agreement with our model.

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[1] Some of our studies are done in collaboration with *Z. Chang's* group, *C.W. Fehrenbach,* and others.

It occurred to us that a good test of our model would be to change the laser wavelength. Doubling the frequency was a convenient choice and would give a spectrum clearly distinct from the 800 nm result. We then performed the measurement, and the data is shown in panel (c) with the new predictions for the peak positions. Again, our model gives a good fit of the data. The more convincing test of the model, though, is the angular distribution. For 400 nm light, our model predicts ionization primarily with 6-7 photons. The $\cos^{2n}\theta$ fit gave n=6.5±1.0 in good agreement as before.

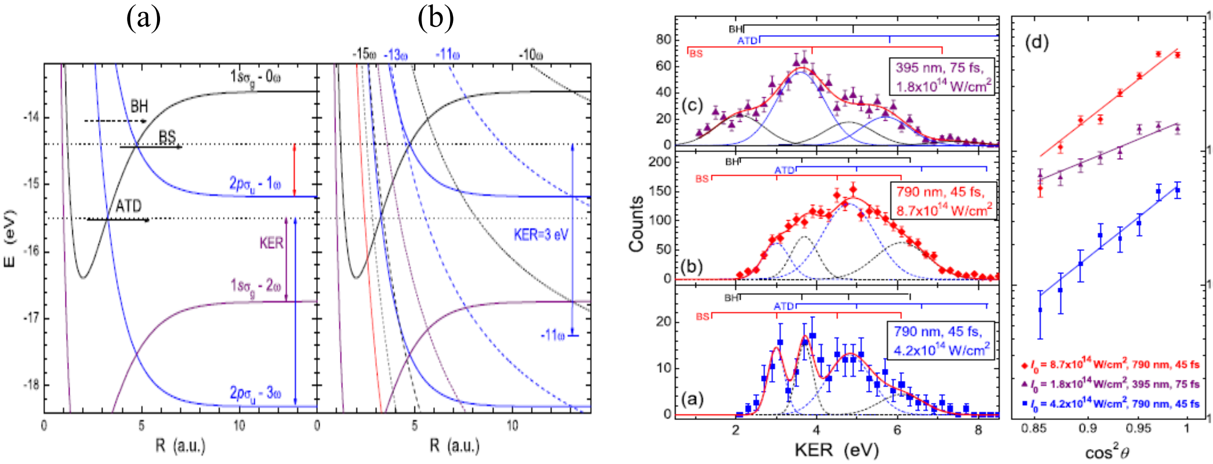

Figure 1. *Left*: (a) The diabatic Floquet potentials for H_2^+. Besides the molecular quantum numbers, each curve carries a photon number label. (b) The same as (a), but including the ionization threshold potentials. *Right*: (a)–(c) Experimental ionization KER spectra. The vertical bars indicate the predicted KER peak locations, grouped by initiating mechanism. (d) Experimental log-log angular distributions corresponding to the spectra (a)–(c).

We have also successfully applied this model to explain the surprising structure in the double ionization of H_2 reported recently by Staude *et al.* – see the report of Brett Esry for details.

Enhanced ionization of H_2^+ (CREI) – *I. Ben-Itzhak, P.Q. Wang, A.M. Sayler, K.D. Carnes, M. Leonard, B.D. Esry, A.S. Alnaser, B. Ulrich, X.M. Tong, I.V. Litvinyuk, C.M. Maharjan, P. Ranitovic, T. Osipov, S. Ghimire, Z. Chang and C.L. Cocke*

The goal for this JRML "super-group" effort was to reveal the predicted structure in the CREI spectrum of H_2^+. Our data shows no such structure mainly due to the nuclear motion of the stretching molecule and intensity averaging in the interaction region.

The research conducted at JRML on the interaction of intense short-pulse lasers with molecular ion beams and with molecular targets is complementary to each other. In an effort to improve our understanding of enhanced ionization we teamed up. Zuo and Bandrauk suggested that this enhancement in ionization is due to charge resonance enhanced ionization (CREI) around some critical internuclear distances that are larger than the bond length of H_2^+. Their calculations predicted two prominent peaks in the ionization rate centered about an internuclear distance, R, of 7 and 10 a.u. [2], which initiated further theoretical work on the structure of CREI (e.g. Refs. [3-4]). Furthermore, this intriguing structure motivated experimental work trying to reveal it [5-9]. The fact that ionization of H_2^+ is enhanced at some large R is well established. However, evidence for the second CREI peak around 10 a.u. is still in dispute. Using the pump-probe technique we selected the ionization of D_2^+ dissociating on the $2p\sigma_u$-1ω curve and converted the time delay to the internuclear distance where ionization occurred, as shown in Fig. 2(Left). We also converted the KER distribution of an ionized H_2^+ beam, shown in Fig. 2(Right), to *R* using a similar classical model for the propagation. Both measured distributions clearly show a single broad enhanced-ionization peak and do not support the predicted second CREI peak at 10 a.u. We suggested that the CREI structure predicted by "frozen nuclei" calculations [2-4,10] is washed out by nuclear motion during the stretch prior to ionization [10] and by intensity averaging effects. We hope that these recently submitted results will bring to an end the "hunt" for the double peak structure in enhanced ionization.

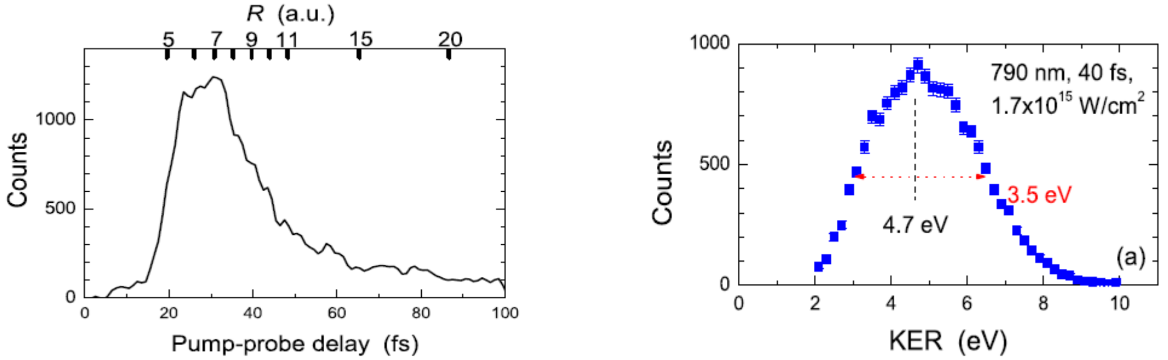

Figure 2. *Left*: measurements. (a) Measured pump-probe time-delay distribution of D_2, the associated value of *R* is marked on the top axis. *Right*: Measured KER distribution for an H_2^+ beam. Note that a KER of about 4.6 is expected for ionization at R=7 a.u. (including the dissociation energy gained to that point), while ionization at R=10 a.u. should yield 3.4 eV.

In addition to the projects described in some detail above, we have studied a few other molecular-ion beams with our short-pulse laser and extended our studies to include few-cycle pulses. Furthermore, we have conducted a few ion-molecule collision experiments [see, for example Pub. #17-18]. In parallel, we are upgrading our molecular dissociation imaging setup for upcoming studies of collisions of a few keV molecular ion beams with atomic targets.

Future plans: We are in the process of analyzing recent measurements of H_2^+, O_2^+ and N_2^+ beams interrogated by intense sub-10 fs FWHM pulses. First attempts to measure the predicted effects of the carrier envelop phase (CEP) on HD^+ laser induced dissociation (see Pub. #7) have recently been carried out. Further improvements are required to bring this project to completion. Progress has been made on the understanding of the dissociation and ionization of more complex diatomic molecules, such as O_2^+ and N_2^+, and we will continue our efforts in this direction. Finally, we hope to finish the upgrade of our new experimental setup, which will enable kinematically complete studies of dissociative capture at keV energies.

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