Time-dependent Treatment of Three-body Systems in Intense Laser Fields

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Program Scope

The primary goal of this program is to understand the behavior of H_2^+ in an intense laser field. Because the system has more degrees of freedom than can be directly treated, past theoretical descriptions have artificially reduced the dimensionality of the problem or excluded one — or more — important physical processes such as electronic excitation, ionization, vibration, or rotation. One component of this work is thus to systematically include these processes in three dimensions and gauge their importance based on actual calculations. Further, as laser pulses get shorter and more intense, approaches that have proven useful in the past may become less so. A second component of this program is thus to develop novel analytical and numerical tools to describe H_2^+. The ultimate goal is to understand the dynamics of these strongly coupled systems in quantum mechanical terms.

Recent progress

The above-stated goal presupposes that the behavior of H_2^+ in an intense field is not already completely understood. Many in this field, though, claim just the opposite — that everything is understood. Some recent measurements, however, belie this claim ([R1], [P14], and see I. Ben-Itzhak's abstract). These two experiments — at NRC in Canada [R1] and JRML at Kansas State [P14] — revealed structure in the relative nuclear kinetic energy spectrum following single and double ionization of H_2^+ and H_2, respectively, that neither was expected nor has an accepted explanation. In fact, there are at least three distinct models being discussed with no resolution imminent.

One of those models was put forward by my collaborators and me in [P14] where we successfully applied it to explain essentially all of the features of our measurements. Our model is described in I. Ben-Itzhak's abstract which also shows its fit of the data, but I will summarize it again here for the clarity of the following discussion. Our model imagines that ionization is initiated by dissociation. At a sequence of internuclear distances, ionization channels for n, $n-1$, ... photon transitions open. The dissociating nuclei pass through these channel-opening distances, leading to the observed peaks. For each dissociation mechanism — bond-softening and above threshold dissociation, for instance — there will thus be a sequence of peaks separated by a photon's energy. In analogy to above threshold ionization and above threshold dissociation, we have labeled this phenomenon above threshold Coulomb explosion.

Our model is most easily visualized using the Floquet potentials [R2] shown in Fig. 1. This figure shows the main non-standard piece of our model: diabatic Floquet ionization-threshold potentials. These potentials are just $1/R - n\omega$ in atomic units and represent n-photon ionization with zero total energy electrons. They are shifted by $n\omega$ in accord with the Floquet representation. One advantage of this picture is that it treats dissociation and ionization on an equal footing. Another advantage is that total energy is conserved within roughly the bandwidth of the laser pulse. The nuclear dynamics in an intense field can thus be completely followed just as for any other set of Born-Oppenheimer potentials. Time-dependent predictions follow from the time the system takes to travel along a given potential.

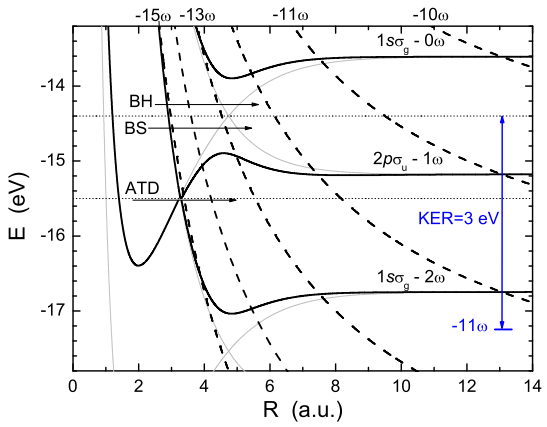

Figure 1: Floquet potentials for H_2^+ at 800 nm: adiabatic dissociation potentials at 10^{13} W/cm^2 (solid lines) and diabatic ionization potentials (dashed lines). The latter are simply $1/R - n\omega$ in atomic units. The approximate total energy available for the main initiating dissociation mechanisms are indicated: bond-softening (BS), bond-hardening (BH), and above threshold dissociation (ATD). The kinetic energy release (KER) expected for BS-initiated, 11ω ionization is indicated at the right (blue). The molecular axis was assumed aligned with the linearly polarized field and fixed in space.

In terms of the kinetic energy release (KER) spectrum, Fig. 1 allows us to predict the peak positions for each dissociation process. It also allows us to predict which peaks should appear by comparing the pulse duration with the travel times to each crossing. We can further make reasonable arguments about the relative magnitudes of the various peaks. In fact, nearly all of the qualitative trends observed can be explained within our model.

Unknown to us and essentially simultaneously with our work, Andre Staudte in Reinhard Dörner's group was measuring Coulomb explosion of H_2 and D_2 using the intense laser facilities at the NRC in Ottawa in collaboration with Paul Corkum's group. As Paul Corkum later told me, they were very surprised to find structure in their KER spectrum. After careful analysis to eliminate any spurious sources of the structure, they were confident it was real but had no satisfactory physical explanation for it [1]. When we learned of their data, we applied our model to try to explain it.

Their data is shown in Fig. 2(a)-(d) for linear and circular polarization at two different intensities. Our fit, one to a linear polarization spectrum and one to a circular, is also shown in the figure in panels (e) and (f). As with our own experimental data, the model produced an excellent fit. The details of this fitting process are essentially the same as described in [P14]. The key point is that we fit both spectra simultaneously and that the consistency requirements of our model more than halved the number of free parameters.

One key difference between their measurements and ours is that they began from neutral H_2 rather than H_2^+. So, the H_2^+ was created when the laser intensity was already high. One consequence is that there are no peaks from bond-hardening, and above threshold dissociation dominates over bond-softening. The latter effect is consistent with the higher intensities experienced. Our model further explained the differences between the isotopes and the differences between the polarizations.

Future plans

It would seem that our model explains the structure observed for both H_2 and H_2^+ quite well. There are, however, some fundamental assumptions in it that are best tested by comparison with solutions of the time-dependent Schrödinger equation. These tests are one component of the

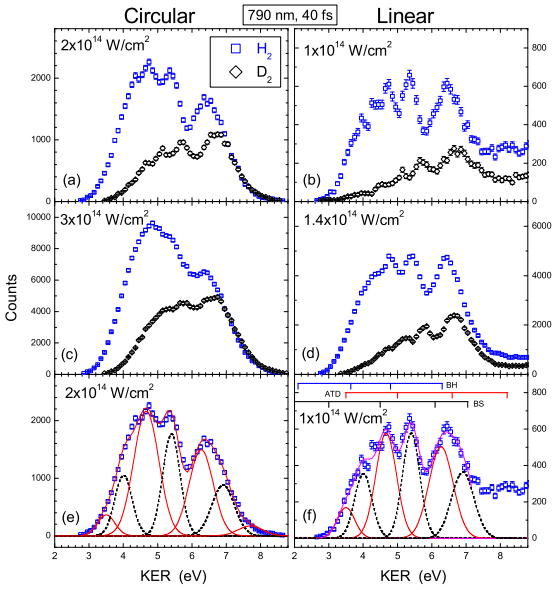

Figure 2: H_2 and D_2 Coulomb explosion KER spectra from Staudte *et al.* [1]. Panels (e) and (f) show our model fit, both for individual peaks and total. The bars in (f) indicate the diabatic peak positions grouped by mechanism. We expect an intensity-dependent shift of the peaks downward from these positions. (Data courtesy of A. Staudte.)

future direction of this work. Whether rigorously supported or not, our model seems to have descriptive and predictive power for this low intensity KER structure where no other explanation has yet proven satisfactory.

My broader future efforts will be to continue to develop the theoretical tools to make quantitative comparisons between theory and experiment. Despite the long history of this problem, very few calculations have been done for the purpose of quantitative comparison (see [R3]). We will also continue to study carrier-envelope phase effects in H_2^+ [P5,P6] and look for other novel effects in these simple systems. I will also expand my group's efforts to effective one-electron molecules and simple two-electron molecules. Our goal in these efforts will be the same as for H_2^+: to make quantitative predictions that can be directly compared with experiment.

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