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Observation of a nearly isotropic, high-energy Coulomb explosion group in the fragmentation of D_2 by short laser pulses

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We have used momentum spectroscopy to examine the production of deuteron pairs in the double ionization of D_2 molecules by a fast (<200 fs), moderately intense $[(0.65-8.0)\times10^{14}$ W/cm$^2]$ laser pulse. By measuring in coincidence the momentum vectors of both deuterons, we have isolated, at the lowest laser power density, a weak ionization channel characterized by Coulomb-explosion deuterons with a sum energy near 10 eV. This is much higher than has been seen in previous studies. The angular distribution of these deuterons is nearly isotropic, in contrast to previous observations that the deuterons are strongly peaked along the laser polarization. These new observations suggest a previously unobserved ionization mechanism.

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The ionization and fragmentation of simple molecules by intense optical fields has been an active area of study, both experimentally and theoretically, during the past 15 years or so, advancing in step with the improvements in short-pulse laser technology (see, e.g., [1–3]). Because of its comparative simplicity and experimental accessibility, the hydrogen (or deuterium) molecule has played an important role in understanding the behavior of diatomic systems in a high optical field environment. Studies of the single ionization and fragmentation of H_2 by intense laser pulses gave rise to the bond softening model [4,5] for the fragmentation dynamics of field-dressed molecular states. The concept of charge resonance enhanced ionization (CREI) at internuclear separations larger than the equilibrium value was inspired by observations in heavier diatomic systems [5–7], but has received substantial support and verification from measurements and calculations of the double ionization of H_2 and D_2 targets [1–3,8–11]. It is widely assumed that the double ionization of H_2 or D_2 in the laser pulse occurs in a two-step process whereby the $H_2{}^+$ molecule, produced by multiphoton ionization of the neutral molecule, begins to dissociate along one of several routes in the molecular-orbital energy-level diagram of the field-dressed molecule. During the dissociation process, the molecular ion is further ionized through the CREI process at internuclear separations near 6 a.u., where the ionization rate reaches a peak value orders of magnitude larger than that for the ground-state separation of the ion (2 a.u.). The resulting Coulomb explosion energy (3–6 eV sum energy) is then substantially less than the 13.6 eV (sum energy) expected for two ions separated by 2 a.u. Since the fragmentation of $H_2{}^+$ (or $D_2{}^+$) is anisotropic, favoring the linear laser polarization direction, the two protons or deuterons are also observed to have peak intensities narrowly spread about the polarization axis.

In this work, we have used coincident ion momentum spectroscopy to study the double ionization of D_2 in 780 nm laser fields. This approach reveals that, with a small but significant probability, there is a mode of double ionization of D_2 whereby the two ion fragments have much higher energies and a far more nearly isotropic distribution than has been seen in previous studies. The observation of this channel was made possible through the coincident measurement, on an event-by-event basis, of the full momentum vectors of both fragments. Except for covariance-mapping experiments [2,12–14], previous fragment energies and channel identifications have been deduced primarily from time-of-flight measurements of single ions from the two-heavy-fragment pairs, thus missing the full information available from coincidence measurement of the momenta of both ions.

Our approach is similar to that used in momentum spectroscopy as applied to single ion or photon collisions with atomic or molecular targets (see, e.g., [15]). A combination of time- and position-sensitive measurement of the impact of each ion on a microchannel-plate detector located at the end of a uniform electric-field region determines the vector momentum of each fragment at the instant of double ionization (see Fig. 1). The target density was kept low to limit the number of molecules ionized per laser pulse. The laser used was a self-mode-locked Ti:sapphire system producing 780 nm pulses at a 1000 Hz repetition rate, with energy per pulse up to approximately 0.4 mJ; we have used pulses with 200 and 100 fs widths. The laser beam was focused to a point inside the uniform electric field such that the ions created were projected onto the face of the detector. Their impact positions were determined utilizing a two-dimensional delay-line anode. Pulses from the ends of the x and y delay lines and a time pickup signal from the microchannel plate were registered by a multihit time-to-digital converter (TDC) and recorded in event-mode by a microcomputer system. From these data, off-line analysis provided the vector momentum of each ion fragment and preparation of appropriate histograms. The deuterium target molecules were introduced into the chamber as a uniform ambient density.

The noncoincident time-of-flight spectra for three pulse intensities are shown in Fig. 2. At the highest intensity [Fig. 2(c)], three well-known peaks are seen in each half of the time-of-flight spectrum containing D^+ ions (the short time ‘‘half’’ from ions launched in the direction of the extraction

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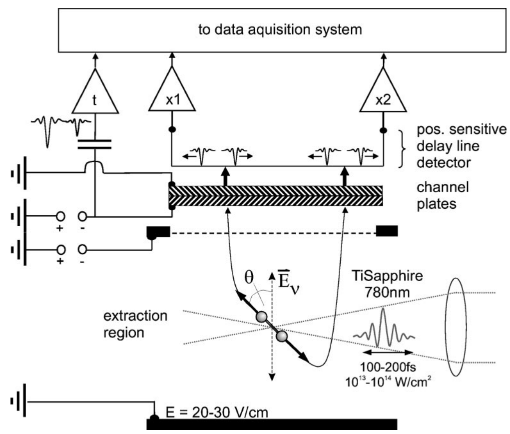

FIG. 1. Sketch of the experimental arrangement.

electric field, the long time ‘‘half’’ from those launched opposite the field). The two dissociation peaks, referred to as bond-softening and ATI peaks, appear at fragment energies near 0.3 and 0.8 eV, respectively; the Coulomb explosion peak from the CREI process appears near 2.2 eV. As expected (see, e.g., Refs. [9,12,16]), as the laser intensity is lowered, the last two of these features become weaker. For the lowest power in Fig. 2, the spectrum is dominated by the bond-softening peak. In this spectrum, no sign of any Coulomb explosion peak is visible, and one would be tempted to conclude that the ionization of the D_2^+ molecule cannot be observed at such a low laser power. This paper demonstrates that a Coulomb feature is still very much present at this laser power, but it requires coincident measurements to reveal it.

Two-ion coincident events are shown in Fig. 3, where two-dimensional histograms of the sum of the energies of the two D^+ ions vs the magnitude of the sum their momenta, $|\mathbf{p}_1+\mathbf{p}_2|$ (where $\mathbf{p}_1$ and $\mathbf{p}_2$ are the vector momenta of the individual D^+ fragments), are plotted. If the two D^+ fragments come from the same molecule, their momentum vectors have equal magnitude and opposite directions in the molecular center of mass, hence $|\mathbf{p}_1+\mathbf{p}_2|$ will be near zero for such events. Thus the true coincidence events lie in a narrow band near $|\mathbf{p}_1+\mathbf{p}_2|=\mathbf{0}$ (to the left of the vertical dashed line in Fig. 3). To the right of the true-coincidence band lie random events which result from coincidences between single ions emitted from different fragmenting molecules. These random coincidences arise primarily from ions produced in the dissociation of D_2^+ molecular ions into a neutral and an ion fragment. To confirm the identification of the true coincidence events, and their separation from the randoms, we purposely generated randoms spectra by choosing, *a posteriori*, $\mathbf{p}_1$ and $\mathbf{p}_2$ from different laser pulses. The panels on the right of Fig. 3 are made from the same data as those on the left, but show only random coincidences generated in this way. The true coincidence events have vanished; the randoms remain.

The projections of the sum fragment energies from the true coincidence band are shown on the horizontal axis of Fig. 3. These spectra represent fragment energy distributions with experimental isolation of the D^+-D^+ decay channel. At the highest laser power, the spectrum is dominated by the well-known broad Coulomb explosion peak centered at a sum energy near 5 eV. As the power is lowered, this peak decreases, and a weak high-energy tail appears. At the lowest power, a remarkably localized peak appears at a sum energy near 10 eV. It is on this feature which we wish to focus our attention in this paper.

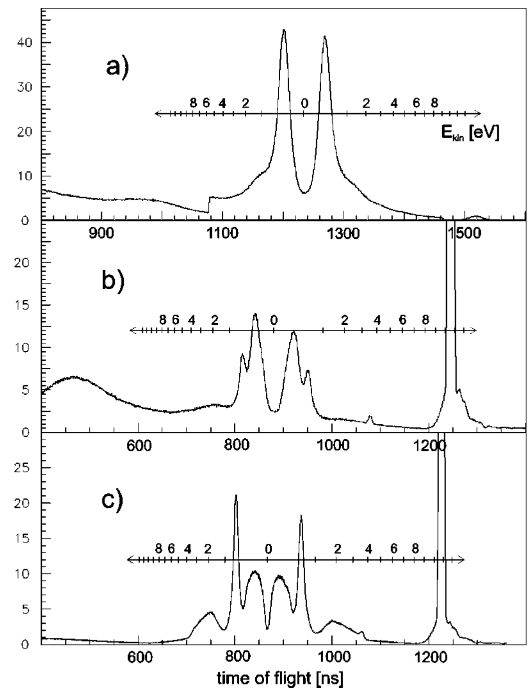

FIG. 2. D^+ time-of-flight distributions at intensities: (a) 0.65, (b) 1.5, and (c) 8.0×10^{14} W/cm^2. The scale inset in each panel shows the conversion from time-of-flight to D^+ kinetic energy per ion, E_{kin}. Differences in the time-of-flight scale reflect the use of different extraction fields. The large off-scale peak shown in (b) and (c) arises from undissociated D_2^+ ions. The small peak between 1150 and 1200 nsec in (b) and (c) is due to HD^+. There is no correlation between the vertical scales on the different panels.

The unusual nature of this peak is confirmed by the angular distribution of the momenta of the two ion fragments. Figure 4 shows the sum ion energy vs the cosine of the polar angle, θ, of emission of the D^+ ions with respect to the polarization of the laser field for events in the true-coincidence band. At the two highest laser powers [Figs. 4(b) and 4(c)], a strong peaking of events along the laser polarization is seen for the CREI peak (sum energy near 5 eV), as is expected and well documented experimentally (see, e.g., [9,12,16]). However, at the lowest laser power [Fig. 3(a)], the angular distribution of the 10 eV peak is nearly isotropic, making it quite obvious when observed in this way even though it is weak in total intensity. The gap in the data near $\cos(\theta)=0$ results from our inability to resolve ion hits which are less than 15 ns apart.

We believe that the 10 eV peak results from a previously unobserved ionization process. This peak has the following characteristics. (i) It is quite localized in explosion energy, indicating that it is generated by an ionization process localized near an internuclear distance of 2.7 a.u. (ii) Ions contributing to it have a nearly isotropic distribution. (iii) It is a

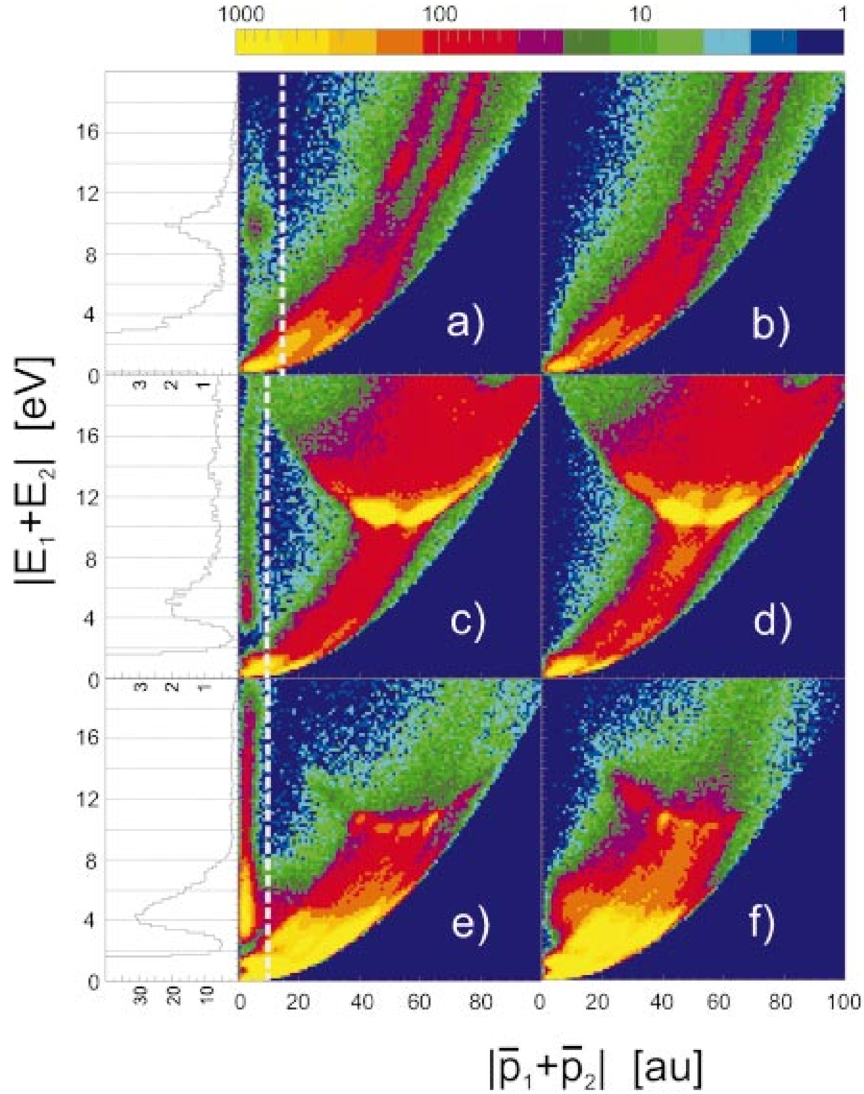

FIG. 3. (Color) Density plots of the sum energy of two coincident D^+ ions versus the magnitude of the vector sum of their momenta. The intensities are (a,b) 0.65, (c,d) 1.5, and (e,f) 8.0×10^{14} W/cm^2. The spectra in the left-hand column (a,c,e) contain both true coincidences and random coincidences. The spectra in the right-hand column (b,d,f) are generated by using first and second D^+ hits from different laser pulses, and thus contain only random coincidences. The projections of sum energies for events lying to the left of the dashed horizontal line in the spectra (a, c, and e) are shown along the energy sum axis.

very weak channel: the total yield in the 10 eV peak is between 4×10^{-4} and 2×10^{-3} of the yield of $D_2{}^+$ ions. (The large spread comes mainly from uncertainties in the D^+ detection efficiency.) Although the data set is not sufficiently continuous to show this, we suspect that, as the laser power is increased, this peak will broaden and evolve into the high-energy fragments seen in Figs. 4(b) and 4(c).

We consider two possible mechanisms which might result in such a feature. The isotropy of the angular distribution shows that, if the precursor is $D_2{}^+$, it is not oriented along the polarization vector at our lowest power level. One possible orientation-independent ionization mechanism would be a rescattering process [19], similar to that seen for atomic targets, whereby the electron liberated in the first ionization event revisits and collisionally ionizes the molecular ion. However, at 6.5×10^{13} W/cm^2 the ponderomotive energy of an electron is only 3.7 eV and its maximum return energy is only 11.8 eV. This is far below the Franck-Condon ionization energy of 26 eV needed to ionize a $D_2{}^+$ ion at an internuclear distance of 2.7 a.u. Thus the experimental conditions seem to exclude the operation of this process in a single step ionization. It is possible, however, that a multirescattering process, involving intermediate excitation of the ion, could be taking place.

A second possibility is that this peak might arise from the small probability that a wave packet produced on the gerade ground-state potential curve of $D_2{}^+$ is further ionized while remaining on this potential curve. In this scenario, the wave packet, created in a Franck-Condon transition from the D_2 potential into the $D_2{}^+$ potential, transits and spreads in this well. This wave packet spends most of its time at the turning points, which lie near 1 and 2.7 a.u. If one reflects this (time-dependent) wave packet from the $v=1$ ground state $D_2{}^+$ potential onto the repulsive D^+-D^+ potential, one obtains a

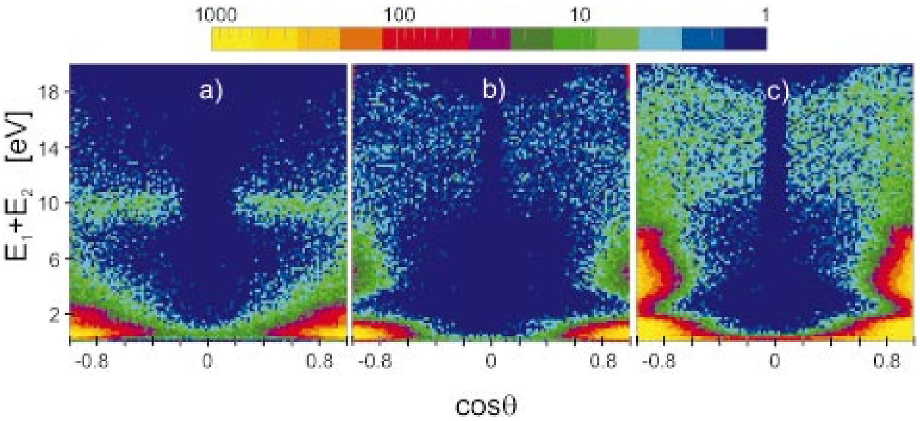

FIG. 4. (Color) Density plots of cos(θ) versus the sum energy of two D^+ ions. Here θ is the angle between the relative momentum vector of the two D^+ ions and the polarization vector. Below a sum energy of about 2 eV, the spectrum has major contributions from random coincidences (see Fig. 3). The intensities are (a) 0.65, (b) 1.5, and (c) 8.0×10^{14} W/cm^2.

sum energy spectrum with a peak near 10 eV, and a long tail extending to higher energies. Ionization from this wave packet is expected to increase rapidly with internuclear separation, R; this dependence would suppress the high energy tail and probably sharpen the 10 eV peak. We note that evidence for direct ionization from the $\upsilon=1$ component of this wave packet has been recently reported by Trump *et al.* [18].

Although the energy from this interpretation of the peak is correct, one must ask whether the expected strength of this channel is consistent with the data. Calculations by Bandrauk and Ruel based on a two-dimensional model for the ${H_2}^+$ molecule [17] give an ionization rate at an internuclear distance of 3 a.u. near 7×10^{10}/s for conditions similar to ours, and show only a weak dependence of the rate on the orientation of the molecule. Over a pulse length of 200 fs, this implies an ionization probability of 1.4×10^{-2}, which is probably consistent with, although somewhat larger than, the observed branching ratio of order 10^{-3}, in view of uncertainties in both numbers. However, a more recent three-dimensional calculation by Chelkowski [20] gives ionization rates between 4×10^6 and 9×10^8/s for power densities of 6.5×10^{13} and 1×10^{14} W/cm^2 and $R=2.7$ to 3 a.u. This result is confirmed by calculations by one of us (U.S.) based on the complex-basis-function method [21]. These rates would yield branching ratios considerably below that observed. Furthermore, the isotropy question remains open in this interpretation. (We note that nonintuitive alignment of ${H_2}^+$ in short laser pulses has been recently reported by Frasinski *et al.* [22]) It is possible that some combination of the two mechanisms discussed above is operating. We consider that the interpretation of this peak remains an open question.

In summary, using coincident measurement of the momenta of both D^+ ion pairs produced from the application of modest-power laser pulses to D_2, we have identified a previously unobserved weak branch in what is presumably the ionization of ${D_2}^+$. This branch is characterized by a rather well-defined Coulomb explosion sum energy of 10 eV, which is consistent with ionization localized near an internuclear distance of 2.7 a.u. The ion pair angular distribution is nearly isotropic, showing that the precursor molecules are not aligned at this power level and that the ionization mechanism must be nearly angle-independent. Our observations may result from a small probability to ionize a vibrational wave packet trapped in the gerade ${D_2}^+$ ground-state potential curve. Although two possible ionization mechanisms are suggested, no fully satisfactory interpretation for either the isotropy of the ions or the experimental branching ratio for this channel is at hand.

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