Rescattering Double Ionization of D_2 and H_2 by Intense Laser Pulses

A. S. Alnaser, T. Osipov, E. P. Benis, A. Wech, C. L. Cocke, X. M. Tong, and C. D. Lin

J. R. Macdonald Laboratory, Physics Department, Kansas State University, Manhattan, Kansas 66506, USA

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We have measured momentum spectra and branching ratios of charged ionic fragments emitted in the double ionization of D_2 (and H_2) molecules by short intense laser pulses. We find high-energy coincident D^+ (and H^+) ion pairs with kinetic energy releases between 8 and 19 eV which appear for linearly polarized light but are absent for circularly polarized light. The dependence on the polarization, the energy distributions of the ions, and the dependence on laser intensity of yield ratios lead us to interpret these ion pairs as due to a rescattering mechanism for the double ionization. A quantitative model is presented which accounts for the major features of the data.

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One of the more intriguing possibilities offered by femtosecond pulses of high intensity light is the use of the outgoing electron wave packet generated in single ionization of a molecular target to probe, in the time domain, the evolution of the associated nuclear wave packet. Because these two wave packets are generated coherently, the revisiting of the singly ionized molecule by the electron at integral multiples of half the optical time period can be used to interrogate the associated nuclear wave packet evolution at controlled later times. Such an idea was recently proposed by Niikura *et al.* [1,2] to interpret their experimental observations of high-energy protons produced from the ionization and dissociation of H_2 by short laser pulses. They proposed that these protons arise from a rescattering excitation of the molecule followed by dissociation of the molecule, and constructed a model for the interpretation of the resulting energy spectra. Staudte *et al.* [3] reported the observation of a similar high-energy D^+ peak in the double ionization of D_2 by femtosecond pulses, but were not able to identify the mechanism responsible. In this Letter we demonstrate that the fast protons (deuterons) seen in Refs. [1–3] are largely due to rescattering-induced ionization, rather than dissociation, of the molecular ion, and show that the multiple returns of the electron wave packet can be seen as fine structure in the energy spectra of the released "fast" protons (H_2) or deuterons (D_2). We find that a correct model interpretation of these return peaks requires the inclusion of not only dissociation but also ionization of the molecular ion.

Rescattering double ionization, whereby the parent ion is further collisionally ionized by the returning electron generated in the first ionization, is well known for atomic targets [4–13] but has only recently been identified for molecules [14]. It is almost certainly the major generator of nonsequential double ionization of atoms. Rescattering multiple ionization of molecular hydrogen has previously escaped detection largely because this molecule can be multiply ionized more easily through charge-resonance-enhanced ionization (CREI) [15–21]. This process, which occurs during the dissociation of the singly ionized molecule, produces well known Coulomb-explosion–like features in the kinetic-energy-release (KER) spectra of the molecular fragments. In the case of D_2 (or H_2) it produces deuterons (protons) with a KER near 5 eV. The identification and clean separation of the rescattering process from the CREI requires some combination of coincidence measurements and energy and angular control of the fragments. In this Letter we use coincident momentum spectroscopy to do this.

The experimental procedure and cold target recoil ion momentum spectroscopy (COLTRIMS) imaging techniques have been previously used by us and others [3,22]. A pulse of 800 nm radiation from a Ti:sapphire laser (pulse length 35 fs, 1 kHz) was focused by an $f/30$ or $f/10$ optical system onto a supersonic jet of D_2 gas. The resulting D_2^+ and D^+ ions were extracted by an electric field of 10–30 V/cm and projected onto the face of a position-sensitive detector equipped with a delay line anode. The positions and flight times for all hits from each laser pulse were measured and analyzed off-line to find the momentum vectors of all detected ions. The D^+ double ionization channel was identified and separated from random coincidences by requiring that the vector sum of the D^+ momenta add to zero. The peak laser intensities were calibrated using the recoil momentum spread of molecules singly ionized with circularly polarized light, as discussed by Litvinyuk *et al.* [23] and checked using single ionization yields of Ar^+ compared to Ammosov-Delone-Krainov (ADK) [24] as well as previously published data [25]. We assign an uncertainty of 30% to our absolute intensity scale.

Figure 1 shows density plots of the magnitude of the sum of the vector momenta, $\mathbf{p}_1$ and $\mathbf{p}_2$, of the D^+ ions plotted versus the KER. Events lying near $|\mathbf{p}_1 + \mathbf{p}_2| = 0$ represent true double ionization events; events lying off this line are due to the random detection of D^+ ions originating from different molecules within the same laser pulse. Events near a KER of 5 eV are due to the CREI process. Events appearing between approximately 8 and

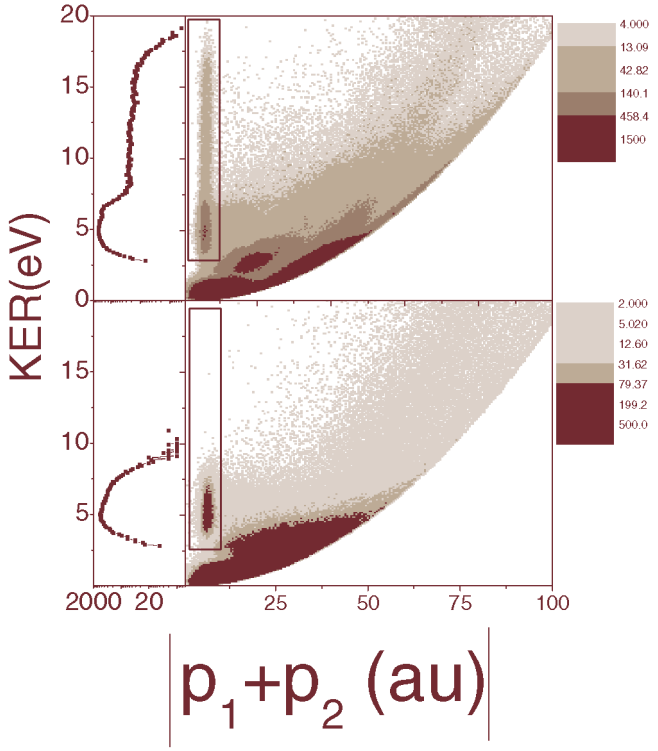

FIG. 1 (color online). Density plot of the KER versus the vector sum of the momenta of D^+ ion pairs . Real coincidences are marked by boxes. Upper panel: linear polarization; lower panel, circular polarization.

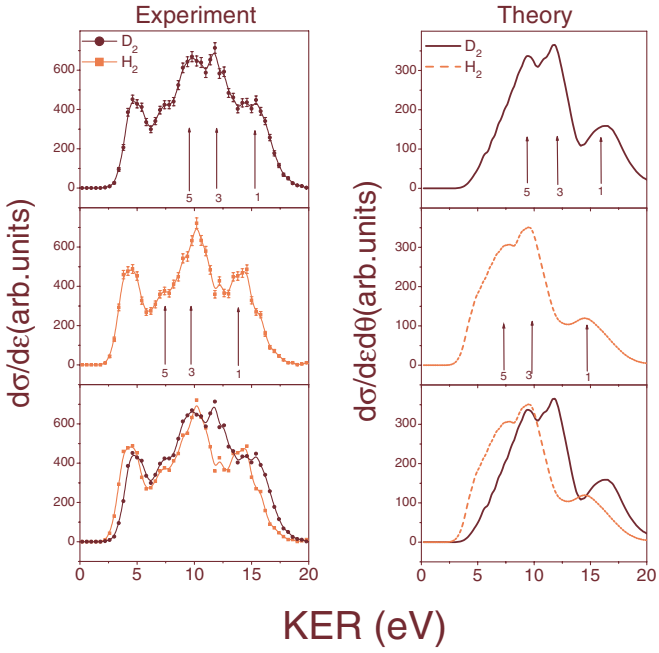

FIG. 2 (color online). KER spectra. Left-hand column: experiment (2.8×10^{14} W/cm^2); right-hand column: model (2×10^{14} W/cm^2). Top panel: D_2 target; middle panel: H_2 target; lower panel: superposition of D_2 and H_2 spectra for comparison. The arrows indicate electron return times (see text for further detail).

19 eV form the rescattering feature on which this Letter focuses. The spectrum in the upper panel of Fig. 1 was obtained with linearly polarized light; the lower one, with circularly polarized light. The total disappearance of the high KER feature for circular polarization immediately indicates that it is due to a rescattering mechanism which is turned off when the electron does not return to the parent atom, as is the case for circularly polarized light.

Our KER spectra and angular distributions for linear polarization confirm those of Staudte *et al.* [3] and are therefore not presented again here: at the lowest intensity for which the rescattering can be observed, the KER distribution shows a single peak near 9 eV and the angular distribution is nearly isotropic. At higher intensities, the KER distribution expands to fill a band of energies between about 8 and 19 eV and the angular distribution becomes less isotropic. Some representative KER spectra from the present experiment are shown in Fig. 2.

The rescattering can be isolated either by selecting coincident D^+ ions for large θ or by subtracting data taken with circularly polarized light from that taken at the same peak field with linearly polarized light, summing over all angles. The results of these two procedures agree almost perfectly if θ is restricted to very near 90°. The yield of rescattering events per D_2^+ parent ion is found to rise smoothly from an effective threshold around 0.8×10^{14} W/cm^2 to a value of 4% around 3×10^{14} W/cm^2 and to remain nearly constant thereafter, in sharp contrast to the behavior of the corresponding ratio for the CREI which rises exponentially with intensity in this region.

We interpret the rescattering as due to inelastic rescattering from the D_2^+ ion by the electron released from the tunneling ionization of D_2 (see Fig. 3). The overall process is similar to that proposed by Niikura *et al.* [1,2], except that it requires the population of the double ionization channel, which turns out to be very important. The electron generated in the tunneling ionization of the D_2 molecule returns and excites the D_2^+ molecule to an electronically excited state from which it can be ionized further into the double ionization continuum by the laser field. This process was followed quantitatively using a model which is described in detail elsewhere [26] and summarized here. The D_2 molecule was initially presumed to be ionized according to the ADK theory [28], simultaneously emitting an electron and populating a vibrational wave packet having the form of the ground vibrational state of D_2 but propagating on the $1s\sigma_g$ potential curve of the D_2^+ ion. The electrons were followed using classical trajectories, including both laser and ionic fields, while the propagation and spread of the vibrational wave packet was calculated numerically. The probability that the returning electron excited the D_2^+ molecule out of the $1s\sigma_g$ state was calculated using theoretical cross sections [27] for free electrons on H_2^+, adjusted for

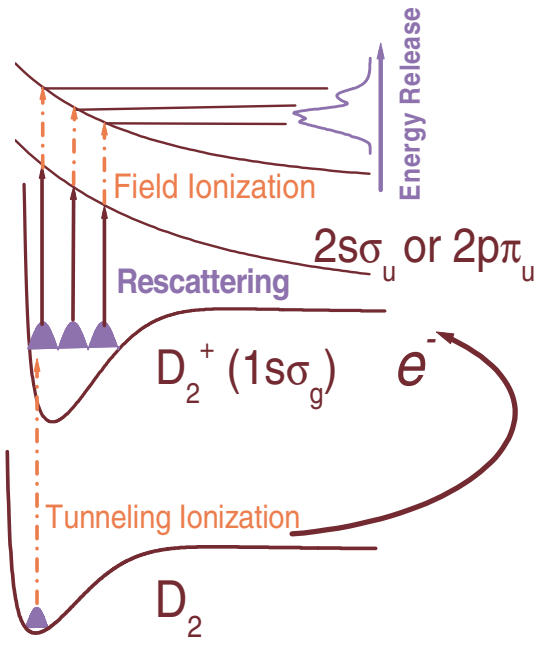

FIG. 3 (color online). Potential curve schematic.

dependence on internuclear distance, including both $2p\sigma_u$ and $2p\pi_u$ channels and correcting for the difference between "near" and asymptotic electron energies [9]. Once populated, the excited electronic states were allowed to ionize further onto the double ionization potential curve according to ADK ionization rates. This process was followed over the time of a 35 fs long pulse and the population on the dissociative and double ionization curves was followed to evaluate the energies and angles of the emitted particles.

In the top panels of Fig. 2 we show experimental and model results for the KER spectra for the rescattering process for intensities of (2.8 and 2) $\times 10^{14}$ W/cm^2, respectively. The model intensity was chosen to be lower than the experimental one (by an amount not exceeding the experimental uncertainty) because this slightly enhanced the agreement. The experimental data were summed over an angular range of 65° to 85° in θ in order to gain statistics, a procedure which allows a small amount of the CREI , seen as a peak near a KER of 5 eV, to remain in the spectrum. The model was evaluated at 90°. The rescattering fills a band between about 8 and 19 eV, the energies corresponding to the dissociation of the molecule on the double ionization potential from the outer and inner turning points of the molecular wave packet (2.2 and 1.4 a.u., respectively). Structure in the KER spectra is attributed to the quantized times of return of the electron. The vertical arrows in those spectra indicate the KERs which result if the center of the wave packet is excited to the double ionization curve at 2/3 of an optical period after the departure of the electron and at successive full periods later. The observed maxima in the yields correlate well with the first, third, fifth, and higher return times (many returns "pile up" at the outer turning point of the wave packet). The electron return times are in principle integral multiples of half the optical period, but the dominance of the odd return times is expected from previous calculations on atoms [9,12] and confirmed by the present model. The model predicts more dominance of ionization at the outer turning point (KER around 9 eV) than is seen in the data, but otherwise agrees with the data in the general structure.

In order to further investigate whether this structure can indeed be attributed to "clock" structure from the returning electron pulses, we investigated the KER spectrum also for a molecular hydrogen target for which the wave packet evolves more rapidly. The H_2 and D_2 gases were mixed in the source so that both targets saw exactly the same laser intensity. The results are shown as the middle panel in Fig. 2, where the expected differences in the return peak structure is confirmed. The agreement between model and experiment on the location of these peaks is excellent.

The rescattering model provides a natural explanation of several other features seen in the data and confirmed by the model. The model predicts a threshold for the rescattering process around 0.6×10^{14} W/cm^2 because the energy difference between the $1s\sigma_g$ and $2p\sigma_u$ states is never less than 10 eV even at the outer turning point of the nuclear wave packet (2.2 a.u.) and this intensity is required to produce a 10 eV electron return energy. At laser intensities just above this, only excitation near the outer turning point is possible, which produces the rather sharp KER peak near 9 eV seen by Staudte *et al.* [3]. The smooth increase of the yield per D_2^+ ion of d^+ ions from the rescattering process to a nearly constant value around 3×10^{14} W/cm^2 reflects the behavior of the excitation cross section. The near isotropy of the angular distributions reported by Staudte *et al.* [3] we believe is due to the near isotropy of both the first ADK ionization stage (predicted [28] but not previously observed) and of the rescattering excitation.

By making software cuts on the angle between the polarization vector and $\mathbf{p}_1$ (or $\mathbf{p}_2$) for noncoincident deuterons, we can select (noncoincident) high-energy deuterons emitted at right angles to the polarization vector, in the same way they were isolated by hardware by Niikura *et al.* [1,2]. In Fig. 4 we plot the ratio between the yield of coincident high-energy D^+-D^+ pairs (double ionization) and the yield of all "high-energy" (KER 8–20 eV) D^+ ions (dissociation plus double ionization) versus laser intensity. In making this plot, we have taken only deuterons emitted at angles greater than 60° to the polarization vector and have determined the absolute deuteron detection efficiency by comparing observed noncoincident and coincident CREI yields. The figure shows the surprising result that ionization through the rescattering process often results in double ionization rather than dissociation even at low intensities. The model predictions for this ratio, shown in Fig. 4, are in good agreement with the measurements.

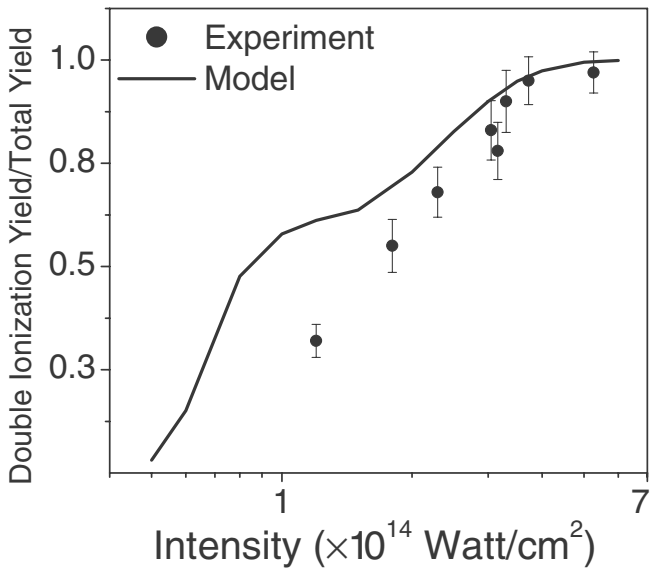

FIG. 4. The ratio of the yield of double ionization D^+ ions with KER between 8 and 20 eV divided by total (noncoincident) yield of D^+ ions in the same energy range, plotted versus peak laser intensity. Data points: experiment ($\theta = 60°$–$80°$); line: model ($\theta = 90°$).

How do our results differ from those of Niikura *et al.* [1,2]? The rescattering mechanism discussed in those works includes conceptually the possibility that, following electronic excitation, the molecular ion can either dissociate or be further ionized after excitation. Indeed, the rescattering process is several times referred to in those works as "double ionization (excitation)." However, the quantitative model used there assumes that, at their intensity of $(1.5 \pm -0.5) \times 10^{14}$ W/cm^2, only the dissociative channel, the $2p\sigma_u$, produces high-energy protons (deuterons). Our data (Fig. 4) show that this is not the case: at this intensity about half of the high KER yield is coming through the double ionization channel. It is very difficult to keep the laser intensity low enough to avoid ionizing the molecular ion. Thus a quantitative model interpretation of the energy spectra should include both excitation of other excited states and ionization of the molecular ion. Failure to do this can lead to incorrect identification of features of the energy spectra. For example, on the basis of our model interpretation we believe that the strong "peak" discussed in Ref. [2] is due more to the third, not the first, return of the electron wave packet.

In summary, we have identified a rescattering mechanism for the double ionization of D_2 (and H_2) by short, intense laser pulses. We find that the production of fast protons (deuterons), with KER above 8 eV, is largely due to this process over a large range of laser intensities. We find experimental structure in the KER spectra which, with the help of a quantitative model, we interpret as due to the quantized return times of the electronic wave packet launched simultaneously with the vibrational wave packet on the $1s\sigma_g$ potential curve of the molecular ion. As predicted by the model, this structure is different for D_2 and H_2. Inclusion of both dissociation and double ionization channels in the model is essential to a correct interpretation of the KER spectra.

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