Strong Laser Field Fragmentation of H_2: Coulomb Explosion without Double Ionization

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We observe fragmentation of H_2 molecules exposed to strong laser fields into excited neutral atoms. The measured excited neutral fragment spectrum resembles the ionic fragmentation spectrum including peaks due to bond softening and Coulomb explosion. To explain the occurrence of excited neutral fragments and their high kinetic energy, we argue that the recently investigated phenomenon of frustrated tunnel ionization is also at work in the neutralization of H^+ ions into excited H^* atoms. In this process the tunneled electron does not gain enough drift energy from the laser field to escape the Coulomb potential and is recaptured. Calculation of classical trajectories as well as a correlated detection measurement of neutral excited H^* and H^+ ions support the mechanism.

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The hydrogen molecule is one of the most fundamental few-body quantum systems. Nevertheless, when exposed to ultrastrong laser fields it exhibits a wealth of specific ionization and dissociation phenomena [1], resulting from the nontrivial, time dependent interplay of nuclear wave packet motion and strong field electron dynamics [2–9]. In previous studies fragments were mostly found to escape from the strong field interaction either ionized or in the strongly bound ground state, as one might expect. In contrast, we here report on the observation of fast and fragile (i.e., highly excited) neutral dissociation products, immediately raising questions about how they gain their kinetic energy, and how they survive such high laser fields. In this Letter we show that a delicate cooperation between molecular dissociation dynamics on the one hand and the recently observed phenomenon of ''frustrated tunnel ionization'' [10] on the other can account for the observations. The process involves a previously disregarded feature of electronic ionization and rescattering in strong laser fields and is expected to be generally present in strong field molecular dissociation, particularly in cases where singly charged fragments are involved. This work lies in the context of ionization and fragmentation dynamics of small diatomic molecules in strong laser fields which have been extensively studied over the past years. Processes such as above threshold dissociation [2], charge resonance enhanced ionization [3], Coulomb explosion [4], and bond softening [5] in H_2 have been identified as characteristic and unique phenomena. Correlated measurements of ionic fragments and of ionic and electronic fragments have been decisively fruitful in unraveling underlying physical mechanisms [11,12] which arise from the combined electronic and nuclear motion in the presence of a strong external laser field.

Neutral fragments after strong field dissociation have been observed using a fast H_2^+ beam [13], without being able to distinguish between excited and ground states. More recently, Lyman-α radiation has been found to be emitted after exposing H_2 molecules to strong 800 nm laser fields [14]. It was attributed to the excitation of a neutral fragment at large nuclear separations ($R \sim 10$ a.u.), a secondary process which is not immediately related to strong field induced fragmentation. In the present experiments we detect long lived excited neutral H^* fragments directly, using the fact that the excitation energy of $\sim$10 eV or more is sufficient to obtain a signal from a microchannel plate detector (MCP) [10]. This allows us to distinguish excited from ground state fragments and, at the same time, to detect their kinetic energies through time-of-flight techniques.

An answer to the question of how excited neutral fragments can evolve from strong laser fields may be obtained from a closer look at the well-known rescattering model of electron dynamics in strong fields [15]. Generally, in this model an electron tunnels into the continuum, returns to the core, and initiates a wealth of new phenomena such as high harmonic generation (attosecond pulse generation) [16], nonsequential double ionization [17,18], above threshold ionization [19], or electron diffraction [20]. Somewhat surprisingly it has been observed that the laser field, albeit well in the tunneling regime for tightly bound states, may even leave the electron in highly excited Rydberg states [10]. In a classical picture the ionized electron would be gently decelerated over many laser cycles and eventually recaptured when the field has disappeared. This process has been dubbed ''frustrated tunnel ionization'' (FTI) and adds to the portfolio of phenomena occurring within the rescattering model. We will show in the following that, in fact, a delicate interplay between molecular dissociation dynamics on the one hand and FTI on the other accounts for the fragmentation involving excited neutral atoms with high kinetic energy.

In the experiment we use a slightly modified version of an experimental setup which has been described elsewhere

[12]. Briefly, a Ti:sapphire laser beam with a repetition rate of 3 kHz, pulse width of 27 fs, and pulse energies of roughly 100 μJ is focused into a supersonic beam of H_2 molecules. Perpendicular to the molecular beam axis two position sensitive MCP detectors in a COLTRIMS setup, D_1 and D_2, are at distances of 0.61 and 0.22 m from the interaction zone, respectively. The detectors can be configured to detect either ions or electrons. Furthermore, both configurations are also suited for detection of excited neutral species. Excited neutral H^* fragments can be directly detected by a standard microchannel plate detector (MCP) if they have an internal excitation energy of more than $\sim$5 eV. This is certainly fulfilled for the lowest excited states in H. To ensure that we indeed detect neutral atoms we apply electric fields to the interaction zone. We have recorded time-of-flight spectra at different extraction fields to ensure that the spectra remain unshifted as expected for neutrals. The detection scheme for excited neutral atoms relies on the fact that the neutral fragments H^* reach the detector in an excited state, either in a high Rydberg state or in the metastable $H(2s)$ into which excited states may decay. The kinetic energy of the H^* fragments should be considerably higher than that of the supersonic beam in order to reach the detector, which is located perpendicular to the molecular beam axis. That the excited neutral fragments gain sufficient energy from the fragmentation process has been answered by the experiment, see Fig. 1. Under our experimental conditions both the metastable state (lifetime 1/7 s) and high lying Rydberg states (especially high l states) live long enough to reach the detector. Using rate equations and the analytically known decay rates of the H levels [21] one can estimate the survival of excited neutrals on their way to the detector.

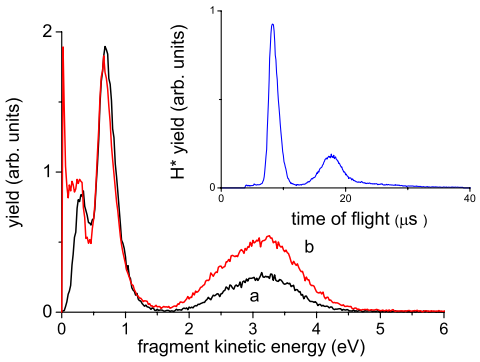

FIG. 1 (color online). Kinetic energy distribution of ionic and excited neutral fragments after strong field interaction at an intensity of $I = 3 \times 10^{14}$ W/cm^2. Shown are the kinetic energy distribution of (a) H^+ (black curve) and (b) H^* (red curve). For comparison it has been assured that the solid angle of detection is the same in both cases. The spectra have been normalized to the maximum of the second low energy peak. The inset shows the H^* time-of-flight spectrum at an extraction voltage of 100 V. Curves taken at other extraction voltages fall on top of this curve as expected for neutral fragments.

We estimate that about 1% of all excited atoms reach the detector in an excited state.

In the inset of Fig. 1 we show a time-of-flight spectrum of H^* atoms after strong field dissociation of H_2 in a linearly polarized laser field with an intensity of $I = 3 \times 10^{14}$ W/cm^2. The time-of-flight (TOF) spectrum can be directly converted into kinetic energy of the fragments by neglecting a small lateral momentum of H^*. We note that the solid angle of detection for the neutral atoms is fixed solely by geometrical considerations and is 0.1 sr. The kinetic energy distribution of H^* together with that of the H^+ ions is shown in Fig. 1. It is striking that the H^* kinetic energy spectrum very much resembles the ionic H^+ fragmentation spectrum. The ion fragment spectrum is well known from previous work [22,23]. The first two low energy peaks originate from bond softening dissociation of H_2^+. The third high energy peak stems from Coulomb explosion of the molecule and is usually explained as a sequential process. First, the strong laser field ionizes H_2 to H_2^+. A nuclear wave packet is induced in H_2^+ at small internuclear separation R that propagates to larger R within a short period of time (7 fs). At intensities around 10^{14} W/cm^2 the mechanism of charge resonance enhanced ionization at internuclear separations R of about 8–10 a.u. leaves behind two bare protons, which repel each other. This results in kinetic energies which depend strongly on the internuclear separation at the instance of the second ionization step. At higher intensities ionization of H_2^+ saturates thus inhibiting the excursion of the nuclear wave packet to large R. Consequently, the second electron is liberated already at small R resulting in higher kinetic energies of the fragments. Correlated measurements of the two ionic fragments have confirmed the Coulomb explosion process [1].

As a matter of fact it seems that all three peaks observed in the neutral spectrum can be linked to those in the H^+ fragment spectrum. Obviously, the high energy peak in the neutral spectrum of Fig. 1 stems from a process similar to Coulomb explosion and the first two low energy peaks suggest that a process similar to bond softening is at work. It is worth mentioning that the height of the ''Coulomb explosion'' peak relative to the second bond softening peak is slightly increased in the neutral fragment spectrum. However, the total amount of excited neutral fragments formed is rather low compared to the number of ion fragments as can be inferred from classical calculations described below ($\sim$5%). To shed light on the physical mechanism behind the kinetic energy distribution of excited neutral fragments we focus on the high energy peak. If it originates from a process similar to the Coulomb explosion one has to find the associated other fragment, which may either be another excited H^* or another H^+ ion. Consequently, we have performed a measurement, where we looked for the correlated appearance of an excited H^* atom and a H^+ ion. To perform the correlated measurement (H^+, H^*), one has to consider

that the rate of excited neutral formation and detection is rather low. Consequently we set the trigger of the data acquisition on the detection of an excited H^* atom on detector D_2. With the second detector D_1 we look for the related H^+ ion. Figure 2 shows the result. Correlated events have to appear along the black line, which indicates equal momentum of the fragments. We find a clear correlation between H^+ and H^* at energies corresponding to the high energy peak in Fig. 1. This corroborates our assumption that a process similar to Coulomb explosion is responsible for the high kinetic energies of the H^* atoms. The energy distribution of H^* found in the correlated measurement agrees very well with the appropriate peak in the measurement, where we detect only H^* atoms. This indicates that the high energy peak in Fig. 1 almost exclusively stems from a process, where at the same time a H^+ ion is formed. At lower energies around 1 eV, where the peaks due to bond softening (BS) are expected, a broad signal is observed, which is not clearly aligned along the black line. Unfortunately, this range of energies is not accessible for a correlated measurement. For technical reasons we were not able to measure the low energy part of the neutral excited atom yield in a correlated measurement. Furthermore, false coincidences, which are unavoidable due to the low production of excited neutral H^* atoms with respect to the large number of ionic fragments overwhelms possible true coincidences. Thus, the false coincidences arise to a large extent from the fact that more than one ion is detected per laser shot meaning that more than one dissociation event happens in the corresponding laser pulse. The strong peak marked with (X) in Fig. 2 indicates exclusively false coincidences.

A possible way to explain the energy distribution for neutral fragmentation is to assume that the H^* fragments gain their energy from a process similar to Coulomb explosion in the first place followed by neutralization.

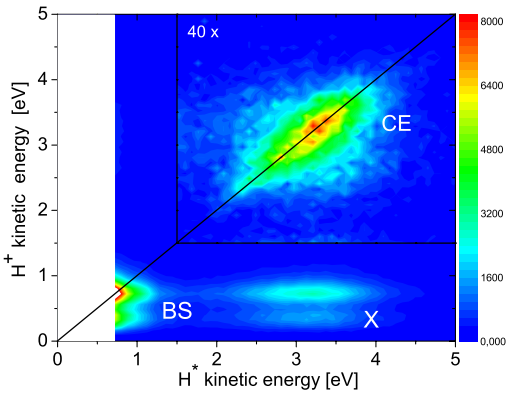

FIG. 2 (color online). From a correlated TOF measurement of neutral excited and ionic fragments a color coded correlated diagram of the kinetic energy of neutral excited H^* and H^+ is extracted. The black line indicates where correlated events should be located. Clearly visible are correlated events originating from the Coulomb explosion (CE). For further explanation, see text.

Neutralization might occur in a process similar to the atomic case, where it has been found that about 10% of the number of ions is converted into excited states of the neutral atom [10,24,25]. The explanation in terms of the FTI process [10] is as follows: although the electron has tunneled out, its final drift energy is not large enough to overcome the attractive Coulomb potential of the ion and is recaptured. This leads to a characteristic distribution of principal quantum numbers n for the final states of the resulting excited neutral atoms. One decisive difference between the atomic and the molecular case is that in the latter, one of the electrons has to be recaptured by a fast H^+ ion. The FTI mechanism predicts that the recapture process vanishes for circularly polarized light. This has been experimentally demonstrated for atoms [10]. In H_2 we also find that the excited neutral fragment signal vanishes for circularly polarized light thus supporting the FTI mechanism.

We have performed Monte Carlo (MC) simulations in analogy to the atomic case [10] by solving the classical equations of motion. First, we show in Fig. 3 that for a specific initial condition one of the H^+ ions is able to capture the tunneled electron in a trajectory with negative total energy. For the calculation we start with a H_2^+ molecule where the two nuclei are initially at rest and separated by $R = 5$ a.u. At the instance of tunnel ionization, which is taken to be close to the field strength maximum, we assume that the electron starts from a position right in between the two nuclei, in accord with the charge resonance enhanced ionization model [3]. Furthermore, we take zero electron momentum along the direction of laser polarization, but allow for a lateral momentum [26], $p_\perp = 0.1$ a.u. The laser intensity was set to 3.19×10^{14} W/cm^2. It is obvious from the trajectories shown that once the electron is tunneled it is then driven by the laser field until the laser pulse is turned off. Eventually, it is recaptured by one of the ions and trapped in a low lying bound state of the hydrogen atom H^*.

In a systematic MC simulation, we vary the starting time of the tunneling electron in the laser field weighted by the tunneling ionization probability, the lateral momentum of

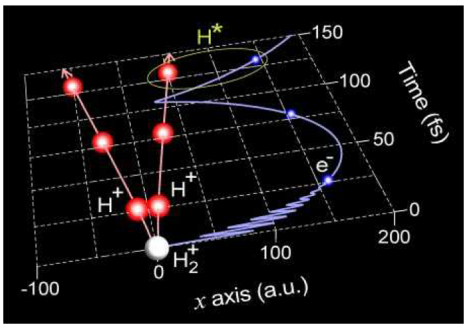

FIG. 3 (color online). Classical trajectory calculation for the Coulomb explosion of H_2^+ after tunnel ionization. Shown are the components of the trajectories of the two nuclei and the electron along the direction of the laser polarization as a function of time.

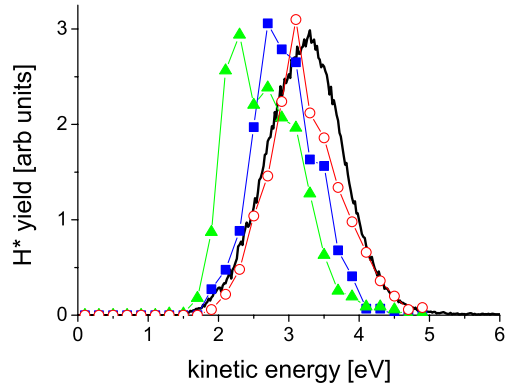

FIG. 4 (color online). Comparison of simulated kinetic energy distributions of excited neutral fragments H^* after Coulomb explosion with experimental data (solid line). Shown are the results from classical Monte Carlo simulations of the Coulomb explosion, where one of the two escaping H^+ recaptures the tunneled electron to form an excited neutral H^* atom. We assumed a Gaussian distribution of nuclear separations centered at $R_0 = 3.5$ a.u. (○), 4 a.u. (■), and 5 a.u. (▲) with a width of 1 a.u.

the electron and chose randomly a nuclear separation R according to a Gaussian distribution centered at R_0. As a result from the calculation we obtain the final momenta of the two fragments, either (H^+, H^+) or $(H^*(n), H^+)$, the number of neutral excited atoms and the final n distribution of the excited fragments. The n distribution is determined from the total energy of the subsystem ion plus electron. At large integration times we have evaluated the momentum of the ion and of the associated neutral excited atom. In Fig. 4 we show the results of three different calculations, in which the Gaussian distribution of the nuclear separations was centered at $R_0 = 3.5$, 4, and 5 a.u. with a width of 1 a.u. (full width half maximum) and zero velocity of the protons at the time of ionization. We plot the number of bound trajectories where an electron has been captured, as a function of the final kinetic energy of the fragment and compare it with the energy distribution of H^* fragments extracted from the correlated measurement shown in Fig. 2. The distributions were normalized to the maximum of the experimental yield. The results show that the electron dynamics in the strong laser field is suitable to explain the origin of the fast H^* fragments in the kinetic energy range of the Coulomb explosion. Best agreement has been found, when the ionization process starts around an nuclear separation centered at 3.5 a.u. Finally, it should be noted that similar calculations can also be performed for the low energy peaks, as will be detailed separately.

In conclusion, the initial question of why we observe excited neutral H^* fragments with high kinetic energies after strong field dissociation of H_2 molecules can be answered in the following way: The strong laser field initially ionizes stepwise both electrons followed by a Coulomb explosion process of the charged fragments. Thereby, one of the electrons does not gain enough energy to fully escape and is recaptured in an atomic Rydberg state of one of the fragment protons, leading to a fast, highly excited hydrogen atom. Consequently, the kinetic energy distribution of the excited neutral fragments is similar to the ionic ones. It can be envisaged that this process is generally present in strong field molecular dissociation, particularly in cases where singly charged fragments are observed. The investigation shows that for about 5% of all Coulomb explosion events the initially tunneled electron further participates in the molecular fragmentation dynamics, which should be accounted for in a complete analysis of the strong field fragmentation of H_2.

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