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Clearly structured fragment kinetic energy spectra discovered in the Coulomb explosion of H_2^+ and D_2^+

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Abstract. In an ion beam experiment the Coulomb explosion of H_2^+ and D_2^+ in intense laser fields was investigated. The kinetic energy spectra of the fragments H^+ and D^+ show strong structure at laser intensities close to the threshold. They can be interpreted by net two-photon dissociation followed by Coulomb explosion at a sharp critical internuclear distance.

PACS. 33.80.Gj Diffuse spectra; predissociation, photodissociation – 33.80.Rv Multiphoton ionization and excitation to highly excited states (e.g., Rydberg states) – 42.50.Hz Strong-field excitation of optical transitions in quantum systems; multiphoton processes; dynamic Stark shift

Numerous experimental and theoretical studies on molecules in intense femtosecond laser light pulses have been performed since this novel kind of light became available more than a decade ago.

H_2^+ and D_2^+ were the preferred molecular species since they are the most elementary molecules in nature consisting of two protons or deuterons bound by one electron. These systems have a relatively simple potential curve system with a bonding ground state and an excited dissociative state immediately above it, which are coupled by a charge resonance transition. The next electronic states are about 11 eV higher in energy resulting in a rather pure two state system.

Exposing these molecules to very intense femtosecond laser pulses two channels of fragmentation were observed. At lower intensities up to 5×10^{13} W/cm^2 only dissociation into H atoms and H^+ (protons) with kinetic energies up to 0.8 eV per fragment occurs. Figure 1 shows the projection of the momenta of the fragments on the 2-dimensional detector with the dissociation channel near the center. In recent ion beam experiment even the H and H^+ fragments from single vibrational levels of H_2^+ and D_2^+ could be discerned in the momentum distributions [1], thereby the special effects predicted by the "light induced potential" (LIP) theory were demonstrated in detail. Some theoretical *ab initio* studies by other groups ([2, 3]) were performed or are underway to interpret the radial and angular structure of the momentum distributions as in Figure 1.

At intensities higher than 5×10^{13} W/cm^2, the Coulomb explosion channel opens up, leading to two protons or deuterons with translational energies between 1 and 3.5 eV and an electron as fragments (Fig. 1). The charge-resonance-enhanced ionisation (CREI) theory [4] explains this effect as follows: at first in the molecular system the electron is located at one proton by charge resonance resulting in a H atom and a proton, which then depart from each other. This dissociating system then remains under the influence of the laser electric field which causes ionisation at a certain distance R_c, the so-called critical distance [4]. Consequently the two unshielded protons repel each other, each gaining half of the Coulomb energy corresponding to the distance R_c, e^2/R_c. In the *ab initio* calculations, Zuo and Bandrauk have found enhanced ionisation probability in the range from $R \approx 5$ to $R \approx 12$ a.u. with two rather sharp maxima [4].

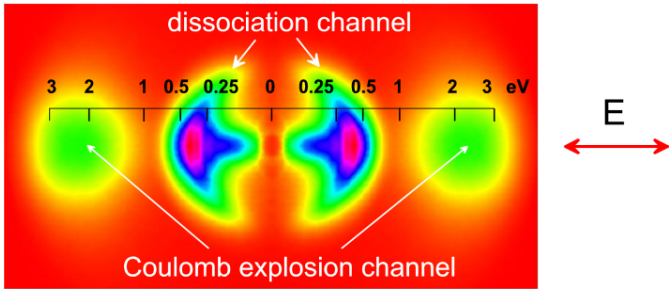

Fig. 1. The momentum distribution of the H_2^+ fragments as projected onto the multichannel plate detector ($I = 1 \times 10^{15}$ W/cm^2). The ion beam was perpendicular to the drawing plane, the direction of the laser polarisation is in the plane. The kinetic energies per fragment in the polarisation direction are inserted. Only one half of the image was really measured, the other half was simulated by mirroring for completeness.

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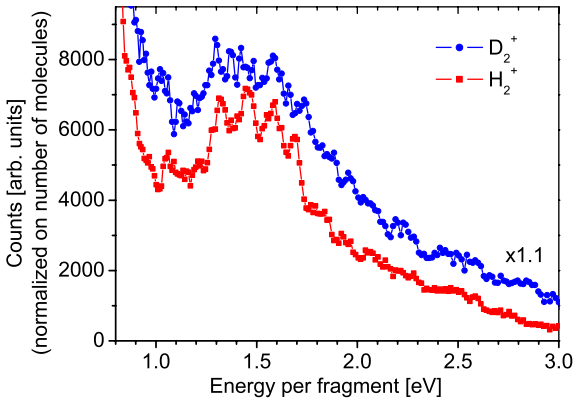

Fig. 2. The number of proton (H^+) and deuteron (D^+) counts from the Coulomb explosion of H_2^+ and D_2^+ as a function of their kinetic energy in comparison. The intensities were $I = 0.6 \times 10^{14}$ W/cm^2 for H_2^+ and 1.0×10^{14} W/cm^2 for D_2^+.

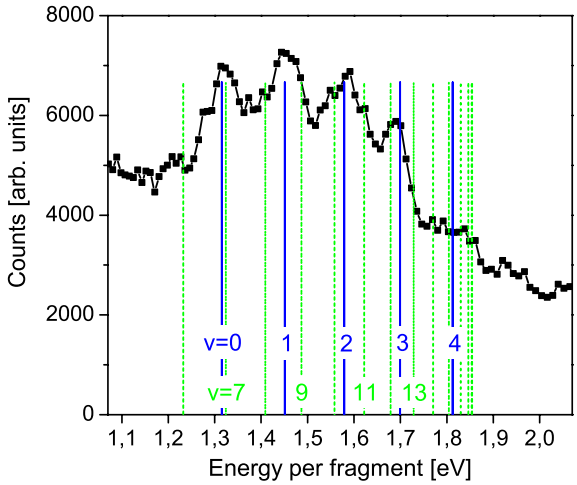

Fig. 3. Coulomb explosion kinetic energy spectra of H_2^+ as in Figure 2 with the assignment of the vibrational excitation of the parent molecular states, using dotted lines if dissociated by one photon, and using solid lines if dissociated by two photons in the first step.

In the experiments reported here the molecular ions were generated in a DC discharge source. This deviates from most preceding experiments done on a H_2-gas probe leaked into high vacuum [5]. An ion beam is drawn out of the source and accelerated to energies of about 11 keV, mass selected and collimated to a very thin beam with a size of about 25 × 300 μm. Femtosecond laser pulses with a maximum energy of 2 mJ at 790 nm and a duration of less than 100 fs were produced in a commercial 1-kHz Ti-sapphire laser system. The laser beam was focused by a 30-cm lens on the ion beam with a current of a few nanoamperes, achieving peak intensities here of up to 10^{15} W/cm^2.

Since the polarisation of the light was perpendicular to the ion beam, the fragments are additionally accelerated in this direction. When coming from one rovibrational state of H_2^+, they lie on an imaginary sphere that expands when moving towards the 2-dimensional multichannel plate detector, which is oriented perpendicular to the ion beam. A rather sharp half circular shaped pattern is generated there (Fig. 1), from which the original spatial 3-dimensional distribution of the momenta can be regained by an inverse Abel transformation.

After the investigation of the dissociation channel [1] ($H_2^+ \rightarrow H+H^+$), some results of our work on the Coulomb channel ($H_2^+ \rightarrow H^+ + H^+ + e^-$) are reported here. The proton fragments in the Coulomb explosion channel appear in Figure 1 as a circularly shaped, only little structured distribution that is very narrow compared to that of the dissociation channel.

Surprisingly, the momentum distribution integrated over one degree along the polarisation axis reveals clear structure for both H_2^+ and D_2^+, as shown in Figure 2. This momentum distribution was measured at intensities close to the threshold of the Coulomb channel at 5×10^{13} W/cm^2. The position of the peaks and relative amplitudes are found to be very sensitive to small changes in intensity. To our knowledge, this kind of structure has not been observed before in the numerous measurements performed on H_2 and H_2^+ at high intensity [5,6], and is also not predicted by theory [5]. However, different type of structure was reported in experiments performed on H_2 gas [7,8].

First we suspected that the structure measured here could be ascribed to the vibrational levels as observed in the dissociation channel [1] simply shifted by the Coulomb repulsion energy of the two protons corresponding to CREI theory. A satisfying assignment of the peaks to the vibrational quantum numbers analogous to that in the dissociation channel [1] using the one-photon absorption energy "comb" failed.

However, including the net two-photon absorption (three-photon absorption followed by one-photon emission) in the dissociative step, as suggested by the dressed state model [5], led to a convincing assignments of the peaks as shown in Figure 3. The centers of the biggest peaks in the spectrum are well fitted by the energies of the vibrational quanta $v = 0$–4 of the net two-photon absorption process, while the smaller features in the spectrum can be explained by the one-photon absorption quanta. In order to match the spectrum, both one- and net two-photon kinetic energy combs were shifted by the same Coulomb repulsion energy of about 1.07 eV. This kinetic energy corresponds to Coulomb repulsion at approximately 12 a.u.

The surprisingly clear vibrational structure in the Coulomb explosion channel can be explained by a closer look at the CREI model: in the leading dissociative process the molecules from different vibrational levels gain different kinetic energies as already observed in the dissociation channel. They all are ionised then at the same critical distance and by the Coulomb repulsion all gain the same additional kinetic energy. So the vibrational structure is preserved.

The strong two-photon absorption in the Coulomb-shifted vibrational spectrum has to be explained by the relatively high intensity used in the Coulomb explosion

experiments since this also leads to a stronger two-photon effect in the preceding dissociative process. The Coulomb explosion spectra of D_2^+ as shown in Figure 2 can be assigned and interpreted in an analogous way and will be reported in a forthcoming paper.

Compared to the dissociation channel, the number of Coulomb explosion fragments in the presented measurements was two orders of magnitude smaller. Due to the much lower signal-to-noise ratio, only some indications of structure can be recognized in the projected two-dimensional momentum distributions. The peaks in the kinetic energy spectra such as in Figure 3 become clear only after summing the counts of a sufficient number of pixels (here about ten) that lie on the same radius. The corresponding angular distributions can be fitted with Gaussian distributions of full widths at half maximum (FWHM) between 30° and 34° (±1°). Such narrow angular distributions are presumably caused by a strong non-linear dependence of the ionisation probability on the angle between the molecular axis and the axis of the laser electric field. To our knowledge, no quantitative theoretical investigation of the angular distribution of the Coulomb explosion fragments is published.

We conclude that our measurements can be qualitatively explained by the CREI theory [4]. In particular the concept of a sharp critical distance at which the ionisation takes place seems to be supported. However, there is only weak support for a second critical distance of similar effectivity leading to another "Coulomb image" of the dissociation spectrum as was postulated by Zuo and Bandrauk [4]. For better comparison with theory, calculations with parameters describing more accurately our experimental conditions would be needed.

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