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Multiphoton ionization of dissociating $D_2{}^+$ molecules

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We report the observation of probe pulse multiphoton ionization (MPI) of dissociating $D_2{}^+$ molecular ions. $D_2{}^+$ is formed by a pump light pulse through MPI of D_2 at the equilibrium internuclear separation. The ions then dissociate in a Floquet one- and two-photon channel. MPI of $D_2{}^+$ is observed as a pump-probe delay time-dependent structure in high-resolution D^+ ion kinetic energy distributions. The dependence on delay time is caused by Coulomb repulsion of the two bare deuterons after $D_2{}^+$ MPI which changes with the internuclear separation where the ions are hit by the probe light pulse. A model calculation shows that in the experiment we probably only observe those ions dissociating in the two-photon channel. [S1050-2947(99)01304-9]

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I. INTRODUCTION

In recent experimental investigations Coulomb explosion imaging of nuclear wave functions based on photoionization by ultrashort light pulses was introduced by Stapelfeldt and co-workers [1,2]. This technique is valuable to directly monitor nuclear wave packet dynamics via ion kinetic energy distributions. For example, in a pump-probe arrangement a pump light pulse initiates dissociation of a molecular system (M_1-M_2). By delayed removal of at least two electrons from the dissociating molecule the nuclear wave packet formed by the pump pulse is then projected onto a repulsive Coulomb potential curve of the dissociating system ($M_1{}^{q+}$-$M_2{}^{p+}$). For a short enough probe light pulse photoionization is instantaneous with respect to nuclear motion. In this case the kinetic energy distribution of the ions is an image of the nuclear wave function at the instant of photoionization [1,2].

This imaging method relies on dissociation of the system on a purely repulsive Coulomb potential energy curve after photoionization. Depending on the molecule this may be in question especially at short internuclear separations where the Coulomb potential curve may be distorted by the presence of non-Coulomb interactions [2]. A system with a pure Coulomb interaction potential after removal of two electrons is investigated in this work. We are looking into Coulomb explosion of $D_2{}^+$ molecular ions dissociating in a high-intensity pump light pulse after removal of the electron of this ion by a delayed probe pulse. A schematic view of preparation dissociation and subsequent ionization is given in Fig. 1. We have chosen deuterium instead of hydrogen for the experiment since the higher nuclear mass gives rise to a lower dissociation velocity at similar excess energy in the molecule.

II. EXPERIMENT

In this experiment we used pulses of a Kerr-lens mode locked Ti:sapphire laser at 790 nm amplified to a pulse energy of up to 2 mJ at 10 Hz repetition rate [3]. The full width at half maximum of the pulses was ≈ 80 fsec. Their shape can best be approximated by a Lorentzian intensity distribution. The laser beam was focused to a focal spot diameter of ≈ 25 μm within the interaction region with the D_2 molecules. To adjust the light intensity in the focal spot we used a combination of a $\lambda/2$-wave plate and a polarizer. The necessary pump and probe beams were formed in a Michelson-interferometer-like setup with one arm continuously variable in length. Both beams were linearly polarized with the polarization parallel to each other and pump and probe pulses were of nearly equal intensity.

Deuterium molecules were supplied to the interaction region in a well collimated pulsed supersonic beam. The D_2 velocity in the beam is (1750 ± 100) m/sec. From measurements a speed ratio [4] larger than 20 can be estimated for the beam, which gives rise to a velocity spread of less than

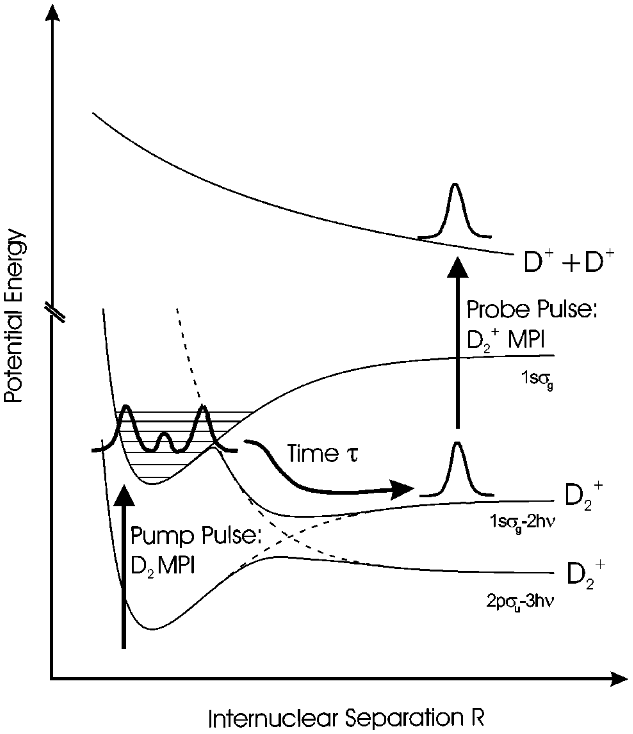

FIG. 1. Schematic view of the pump-probe experiment. Shown are adiabatic (full lines) and diabatic (dashed lines) dressed $D_2{}^+$ potential energy curves for the $1s\sigma_g$ electronic ground and $2p\sigma_u$ first excited state. For further details see the text.

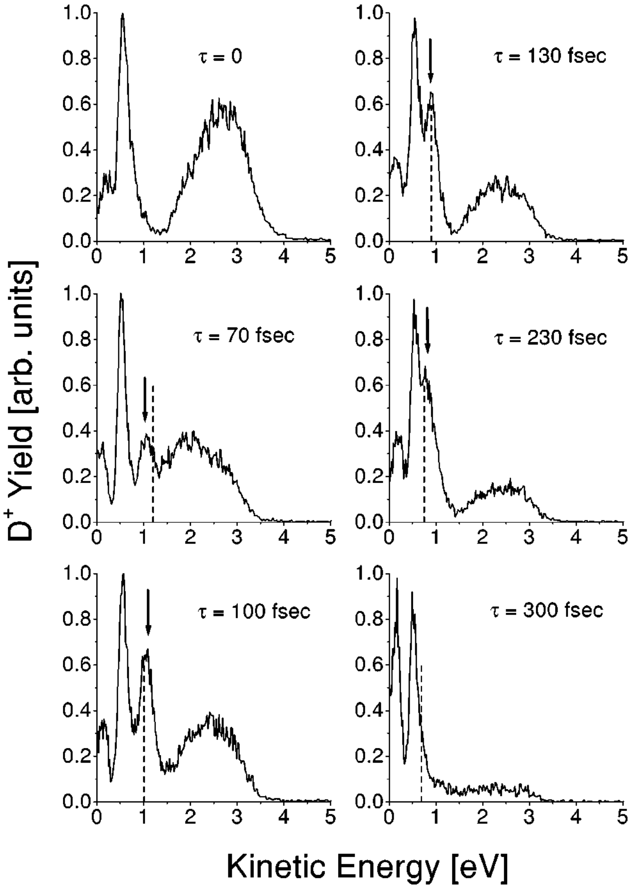

FIG. 2. D^+ ion kinetic energy distributions measured at several delay times τ between a pump and probe light pulse. The respective delay times are given in the spectra. The pump and probe pulse wavelengths are 790 nm and the pulse widths 80 fsec. An observed delay-time-dependent structure is indicated by an arrow in each spectrum where it is visible. The dashed lines give the kinetic energy where we expect to find deuterons from Coulomb explosion after photoionization of D_2^+ by the probe light pulse according to Eq. (4).

±45m/sec along the beam axis. Transverse to the beam axis the velocity spread is determined by the beam collimation to less than ±3 m/sec. The molecular beam setup eliminates the thermal velocity spread present in a bulk gas sample, which we found mainly limits the resolution in kinetic energy distributions of dissociation products, especially in the case of fragments with equal mass [5].

We used an electric-field-free time of flight technique to measure angle resolved kinetic energy distributions of D^+ fragment photoions. The acceptance angle of the flight tube was ±2.3° with respect to the spectrometer axis. The geometry of the spectrometer setup presently limits the achievable kinetic energy resolution to $\Delta E/E = 0.05$ in an energy range $E \leqslant 10$ eV which is relevant for this experiment. In the focal spot of the laser beam the particle density was always kept low enough to eliminate space charge effects on the ion kinetic energy distribution.

III. RESULTS AND DISCUSSION

Figure 2 shows pump-probe D^+-ion kinetic energy distributions. They were taken by focusing the pump and probe light beams onto the deuterium molecular beam with several delay times between the light pulses chosen in a range between 0 and 300 fsec. The pump-probe delay τ is indicated in each spectrum. The peak intensity of the light pulses was kept fixed at a value larger than $\approx 10^{14}$ W/cm^2 for all delay times. Depending on count rate the spectra shown were accumulated over 10 000–55 000 laser shots. The D^+ kinetic energy given on the horizontal axis in Fig. 2 refers to the center of mass frame of the D_2 molecules in the molecular beam.

The ion kinetic energy distributions consist of two types of structures, one with positions which do not depend on the pump-probe delay, and a single line which moves with the delay. The moving line is indicated by an arrow in the spectra in Fig. 2. For small delay times it emerges from the broad structure centered between 2 eV and 3 eV. With increasing delay it shifts to smaller kinetic energies and finally merges with a line at 530 meV for $\tau \geqslant 300$ fsec. Besides shifting this line becomes narrower with increasing pump-probe delay. The delay-time-independent features consist of two narrow lines below a kinetic energy of 1 eV and the broad stucture extending from ≈ 1.5 eV to 4 eV.

The structures which do not depend on the pump-probe delay are known from different earlier investigations [5–12]. In the high-intensity light wave first D_2 is photoionized near the equilibrium internuclear separation. At ≈ 800 nm excitation wavelength the molecular ion formed starts dissociating via a Floquet one-photon channel (the 140 meV line in Fig. 2) or via a Floquet two-photon channel (the 530 meV line in Fig. 2) [7–10]. The dissociating ions pass through a range of internuclear separations where charge resonance enhanced ionization (CREI) in the strong electric field of the light wave gives rise to locally enhanced photoionization [13–16,10]. This initiates Coulomb explosion of the bare nuclei which is responsible for the broad structure above 1.5 eV kinetic energy.

The mechanism giving rise to the delay-time-dependent line in the ion kinetic energy distributions can be identified through its dependence on probe pulse delay for large τ. It does not pass the line at 530 meV kinetic energy but merges with it. This suggests that the D^+ ions in the delay-dependent structure originate from those molecular ions which dissociate via the Floquet two-photon channel. Through further excitation of these dissociating D_2^+ ion by the probe light pulse the D^+ ions gain kinetic energy.

The final states reached after excitation by the probe pulse can be identified via the internuclear separation R where the dissociating molecular ions are excited. We can estimate R by the following argument. In the Floquet two-photon dissociation channel the D^+ product ion has a final kinetic energy of 530 meV (see Fig. 2). This corresponds to an excess dissociation energy $E_{\mathrm{ex}} = 1.06$ eV:

$$D_2^+ + \text{``}2\hbar\omega\text{''} \rightarrow D^+ + D(1s) + E_{\mathrm{ex}}. \tag{1}$$

The final velocity v_r (for $R \rightarrow \infty$) of the D^+ ion relative to the $D(1s)$ product atom is then given by

$$v_r = \sqrt{2E_{\mathrm{ex}}/\mu}, \tag{2}$$

with μ the reduced mass of the system consisting of the two deuterons. The internuclear separation R_p where the probe pulse hits the molecule can be estimated by assuming that after formation of $D_2{}^+$ by photoionization of the neutral molecule near the D_2 equilibrium internuclear separation $R_e = 1.4$ a.u. [17] dissociation in the Floquet two-photon channel proceeds completely without any acceleration with the final measured velocity v_r. R_p is then given by $R_p = R_e + \tau v_r$ with τ the probe delay. With this relation we get, for example, $R_p \approx 28$ a.u. at a pump-probe delay time of $\tau = 100$ fsec.

The $D_2{}^+$ molecular ion dissociates on the potentials formed by the electronic states $1s\sigma_g$ and $2p\sigma_u$. At intermediate internuclear separations they are strongly coupled by the laser radiation since they form a pair of charge resonance states [14]. For R larger than ≈ 10 a.u. these potentials are virtually flat and degenerate [18]. For $R > 10$ a.u. the molecular ion therefore dissociates practically without acceleration at the final velocity v_r given by Eq. (2). The approximation made above to estimate the internuclear distance where the probe pulse hits the dissociating system is therefore correct for most of the nuclear trajectory if $\tau \geqslant 100$ fsec.

Picking up of kinetic energy by excitation of the dissociating molecular ion at the internuclear distances where we observe a resolvable probe-delay-dependent line in the ion kinetic energy distributions is only possible by repulsion forces between bare nuclei. This repulsion can become efficient only if the system is either photoionized or excited into bound electronic states where the electronic wave function extends over a spatial region comparable to or larger than the internuclear separation; i.e., larger than, for example, ≈ 28 a.u. in diameter at a probe delay of 100 fsec. At the intensity reached in the probe light pulse we expect that the preferentially reached final state will be the electron in the ionization continuum and not in a highly excited bound state.

At $R > 10$ a.u. binding forces in the initial $1s\sigma_g$ and $2p\sigma_u$ states are practically negligible. In this range of internuclear distances the electron is localized at one of the nuclei of $D_2{}^+$. The molecule therefore consists of a D$(1s)$ ground state atom with a nearby deuteron which slightly perturbs the atomic state. Neglecting small corrections the ionization potential of this system is then simply given by

$$I_p(R) = I_{p0} + \frac{e^2}{R}, \tag{3}$$

with I_{p0} the ionization potential of the ground state atom ($I_{p0} = 13.6$ eV) and e^2/R the Coulomb repulsion energy of the bare nuclei at the internuclear separation R. In the case of deuterium $I_p(R)$ varies between ≈ 15 eV (70 fsec probe delay) and ≈ 14 eV (300 fsec probe delay). Photoionization of the dissociating molecular ion will not modify the velocity of the nuclei at the instant of ionization at a certain nuclear separation R (Franck-Condon principle). The D^+ kinetic energy E_{kin} measured in the experiment is therefore given by half of the sum of the excess dissociation kinetic energy $E_{\text{ex}} = 1.06$ eV in the Floquet two-photon dissociation channel and the nuclear Coulomb repulsion energy e^2/R at the instant of photoionization:

$$E_{\text{kin}} = \frac{1}{2}\left(E_{\text{ex}} + \frac{e^2}{R}\right). \tag{4}$$

For the delay times chosen in the experiment we expect to observe D^+ ions from probe pulse photoionization with a kinetic energy in the range between ≈ 1.2 eV ($\tau = 70$ fsec) and ≈ 0.7 eV ($\tau = 300$ fsec). At the individual delays chosen in Fig. 2 the dashed vertical lines indicate the kinetic energy where we expect to find the D^+ ions from probe pulse photoionization according to expression (4) with the internuclear separation determined by $R_p = R_e + \tau v_r$. The agreement between the calculated line positions and the experiment is satisfactory especially for long delay times. Discrepancies found especially for $\tau = 70$ fsec are expected to arise mainly due to the approximations made to calculate R_p. But also the estimated error in determining the delay time τ ($\Delta\tau \approx \pm 15$ fsec) may add. Using a constant relative velocity v_r [Eq. (2)] for the dissociating $D_2{}^+$ ion is expected to give rise to poor agreement for decreasing τ since the range of internuclear separations where this approximation is valid shrinks in comparison to the small R range where it certainly fails.

The preceding discussion shows that probe pulse photoionization of the dissociating molecular ion obviously is responsible for the probe-delay-dependent structure in the D^+ kinetic energy distributions. The whole mechanism is schematically shown in Fig. 1 in a dressed $D_2{}^+$ potential curve diagram. Shown are the diabatic $1s\sigma_g$, $1s\sigma_g$-$2\hbar\omega$, and $2p\sigma_u$-$3\hbar\omega$ potential curves (dashed lines) and the relevant adiabatic potentials (full lines) in the external light electric field. Avoided crossings appear in the adiabatic potentials where the diabatic potentials cross. After pump pulse photoionization of D_2 near the equilibrium internuclear separation $R = 1.4$ a.u. the molecular ion is first excited to the dissociation continuum of the $2p\sigma_u$ state by three-photon absorption near the point where the $1s\sigma_g$ and $2p\sigma_u$-$3\hbar\omega$ diabatic states cross. During dissociation the molecule preferentially stays on the adiabatic potential which connects $2p\sigma_u$-$3\hbar\omega$ to the $1s\sigma_g$-$2\hbar\omega$ diabatic curve at large internuclear separations. This is caused by the large energy gap induced by the strong one-photon coupling of the $1s\sigma_g$-$2\hbar\omega$ to the $2p\sigma_u$-$3\hbar\omega$ diabatic state at the light intensity where efficient three-photon absorption initiates dissociation. The probe pulse hits the molecular ion dissociating in the two-photon Floquet channel (on the large R $1s\sigma_g$-$2\hbar\omega$ diabatic potential) and ''projects'' the nuclear wave function onto the repulsive Coulomb potential curve (see Fig. 1) without modifying the nuclear state of motion (Franck-Condon principle). Concerning the projection one has to keep in mind that at the presently used light pulse width it is not instantaneous with respect to nuclear motion. For example, the relative nuclear dissociation velocity v_r in the asymptotic (large) R regime is $\approx 14\,000$ m/sec. The internuclear separation therefore can change by approximately 21 a.u. within the light pulse width of ≈ 80 fsec. This ''noninstantaneous'' ionization will have an effect on the kinetic energy distribution of the D^+ ions by broadening the probe pulse-dependent structure in the ion kinetic energy distribution. The broadening is expected to decrease with increasing probe delay since the gradient of the repulsive Coulomb interaction between the bare nuclei after photoionization decreases with increasing internuclear distance. This is what is seen in the experimental data (see Fig. 2).

A simple simulation of the pump-probe experiment is able to reproduce the main features of the experimental data. For the simulation we use a rate equation approach to model photoionization of D_2 and $D_2{}^+$ and a classical-mechanics approach for $D_2{}^+$ dissociation. The experiment is characterized by the following set of two equations:

$$\frac{dn}{dt} = -W_1(t)\, n(t),$$
$$\frac{\partial n_+}{\partial t} + v_r(R,t)\frac{\partial n_+}{\partial R} = -W_2(R,t)\, n_+(r,t) + \delta(R-R_0)\, W_1(t)\, n(t). \tag{5}$$

Here $n(t)$ is the number of D_2 ground state atoms and $n_+(R,t)$ the number density of $D_2{}^+$ molecular ions found with internuclear separation R at time t. $W_1(t)$ is the D_2 and $W_2(R,t)$ the $D_2{}^+$ ionization rate, respectively. W_2 explicitly depends on the internuclear separation R since the ionization potential changes with R. The δ function $\delta(R-R_0)$ in the second equation ensures that $D_2{}^+$ is formed by ionization of D_2 at a specific internuclear separation $R=R_0$ (the D_2 equilibrium internuclear separation $R_0 = 1.4$ a.u. [17]). Within the framework of classical mechanics the term $v_r(R,t)\, \partial n_+/\partial R$ in the second partial differential equation accounts for dissociation of the molecular ion at a velocity $v_r(R,t)$ of the nuclei with respect to each other. v_r depends on R and via the time-dependent amplitude of the light pulse possibly also explicitly upon time t. The first equation allows for depletion of D_2 molecules by photoionization.

From a solution $n_+(R,t)$ of the pair of equations (5) the number density $N(R)$ of deuteron pairs which are formed at an internuclear separation R at the end of the light pulse can be determined from

$$N(R) = \int_{-\infty}^{\infty} dt \;\; W_2(R,t)\, n_+(R,t). \tag{6}$$

With $N(R)$ known the deuteron kinetic energy distribution $N_k(E_{\text{kin}})$ is found from

$$N_k(E_{\text{kin}}) = N(R(E_{\text{kin}}))\frac{dR}{dE_{\text{kin}}}, \tag{7}$$

with E_{kin} related to R via expression (4) above.

The velocity $v_r(R,t)$ has to be calculated from classical equations of motion. Since in the intense electromagnetic field it is difficult to determine the nuclear motion at small internuclear separations where the coupling of the potentials is strong we approximated $v_r(R,t)$ by the constant final relative velocity of the nuclei in the Floquet two-photon dissociation channel. $v_r(R,t)$ is then given by expression (2) above with $E_{\text{ex}} = 1.06$ eV. As already discussed above, with this choice we expect to get the best agreement with the experimental data for long delay times of the probe relative to the pump pulse.

The D_2 ionization rate $W_1(t)$ is approximated for the numerical calculations by the Ammosov-Delone-Krainov (ADK) tunnel rate for multiphoton ionization applied to D_2 [19]. This long wavelength approximation for the rate will be used for simplicity. At the intensity for the numerical simulation this has to be viewed as a lowest-order approximation to the actual rate since the Keldysh parameter γ [20] is slightly larger than one ($\gamma \approx 1.1$). The approximation to the rate is strictly valid only for $\gamma \ll 1$. The $D_2{}^+$ ionization rate $W_2(R,t)$ is also approximated by an ADK tunnel ionization rate. The R dependent ionization potential appearing in the rate is chosen to be the ionization potential of the unperturbed $1s\sigma_g$ electronic ground state of the molecular ion. This is expected to be a reasonable approximation at small $R<3$ a.u. and at large internuclear separations $R>10$ a.u. (see Fig. 1). In the intermediate range the excited ionic state $2p\sigma_u$ is involved in the decay mechanism of $D_2{}^+$ we probe at large internuclear separations. Therefore the choice we made for convenience is probably poor in this R range. In the ADK rate the effective principal quantum number n^* of the $D_2{}^+$ electronic ground state appears. It is defined via $n^* = Z(R)/\sqrt{2\, I_p(R)}$ [19] with $I_p(R)$ the R-dependent ionization potential and Z the effective ionic charge acting on the active electron. Z has to be chosen R dependent since for $R \to 0$ Z is equal to 2 and for $R \to \infty$ to one. In the transition range we arbitrarily choose $Z(R) = 1 + \exp(-R/\tilde{R})$ with $\tilde{R} = 5.8$ a.u., the internuclear separation where the electron in the ground state becomes localized at one or the other nucleus.

For the light pulses we use a Lorentzian intensity distribution which best reproduces measured autocorrelation traces. In the delay time range where pump and probe pulses overlap in time we superimpose the light waves incoherently by adding intensities to arrive at the total intensity the molecules are exposed to. This is reasonable since the Michelson interferometer setup used to subdivide the light beam into a pump and probe part is not interferometrically stable.

With these ingredients we solved the rate equations numerically for pump-probe light pulses of equal intensity (1×10^{14} W/cm^2) and a pulse width of 80 fsec [full width at half maximum (FWHM)]. The light intensity is slightly lower than the intensity for saturation of ionization of D_2 and $D(1s)$ ground state atoms. The result is shown in Fig. 3. In Fig. 3(a) the number density $N(R)$ [see expression (6) above] is shown as a function of the internuclear separation for the pump-probe delays of the experiment (see Fig. 2). In Fig. 3(b) the kinetic energy distribution $N_k(E_{\text{kin}})$ [see expression (7) above] of the D^+ ions is given for the same pump-probe delay times.

The calculated ion kinetic energy distributions [Fig. 3(b)] show a line at a position which is in good agreement with the measured delay-dependent structure in the experiment (Fig. 2) for delay times $\tau \geq 100$ fsec. At $\tau = 70$ fsec an obvious deviation of the calculated from the measured line position is found. The calculated line appears at a higher kinetic energy than the measured one. Supposing the measured delay time is correct (uncertainty in time measurement is ± 15 fsec), the velocity of dissociation v_r is underestimated in the calculation. This is expected since at small internuclear separations the actual relative velocity $v_r(R,t)$ of the nuclei is probably higher than $\sqrt{2\, E_{\text{ex}}/\mu}$, the dissociation velocity used in the calculation. The deviation becomes increasingly unimportant with increasing pump-probe delay.

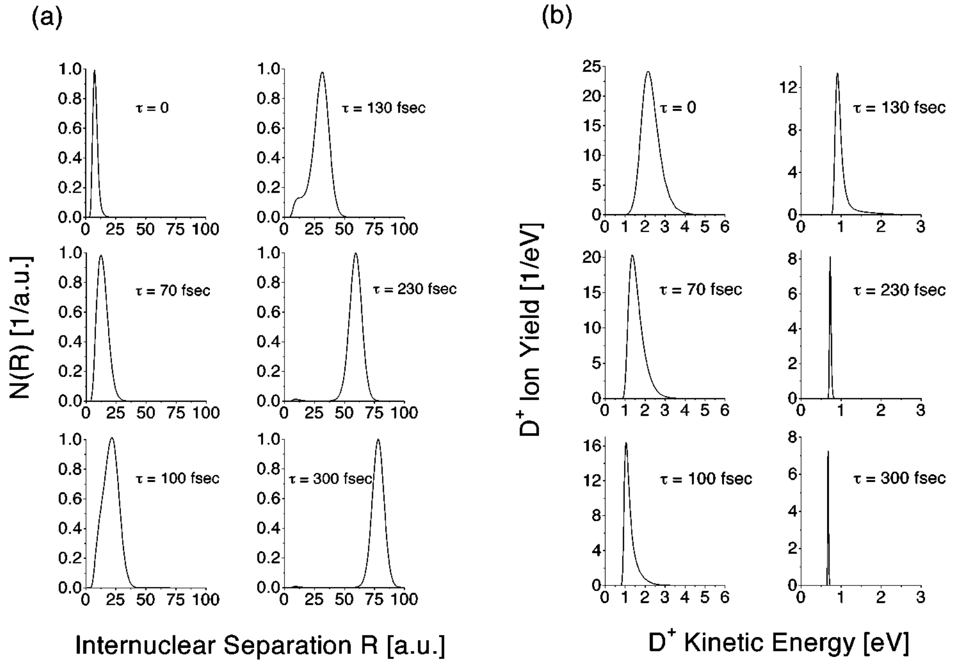

FIG. 3. (a) Calculated number density $N(R)$ of deuteron pairs formed by pump and probe pulses as a function of the internuclear separation for the pump-probe delay times of the experiment [see expression (6) in the text]. (b) The corresponding D^+ ion kinetic energy distributions calculated with expression (7). In the calculation only those $D_2{}^+$ ions were taken into account which dissociate in the Floquet two-photon channel.

The width of the calculated line decreases with increasing pump-probe delay. It is exclusively determined by the slope of the repulsive D^+-D^+ Coulomb potential and the pulse width of the pump and probe pulses. With increasing R the slope of the potential approaches zero. Therefore a zero width of the calculated line is expected at this extreme internuclear separation. In the experiment a decrease in the linewidth was also observed for increasing pump-probe delay (see Fig. 2). But there a lower limit of the width is determined by the energy resolution of the experimental setup. Also the present calculation neglects a possible initial spread in energy of the dissociating molecular ion which may add to the width observed in the experiment.

In the calculation one finds a broad line centered at 2.1 eV for $\tau=0$ pump-probe delay. At this delay where a single pulse with intensity 2×10^{14} W/cm^2 is acting on the D_2 molecule the total D^+ yield is highest [see the D^+ ion yield scale in Fig. 3(b)]. At zero delay dissociation of the molecular ion proceeds after photoionization of D_2 until the internuclear separation reaches $\approx$7.5 a.u. [see Fig. 3(a)]. There the ionization potential of $D_2{}^+$ is reduced to a value where efficient ionization becomes possible in the trailing edge of the light pulse which has a pulse width of 80 fsec. The decrease in ion yield with increasing pump-probe delay time is caused by a decreasing light intensity at the probe pulse maximum due to the slowly decreasing trailing edge of the pump pulse. The rising ionization rate of $D_2{}^+$ due to the ionization potential which decreases with increasing internuclear separation cannot compensate for the decreasing probe peak intensity. This situation changes if the light intensity of the probe pulse is increased to a point where photoionization of $D_2{}^+$ can be saturated beyond a certain internuclear separation.

An asymmetry of the calculated ion kinetic energy distri-

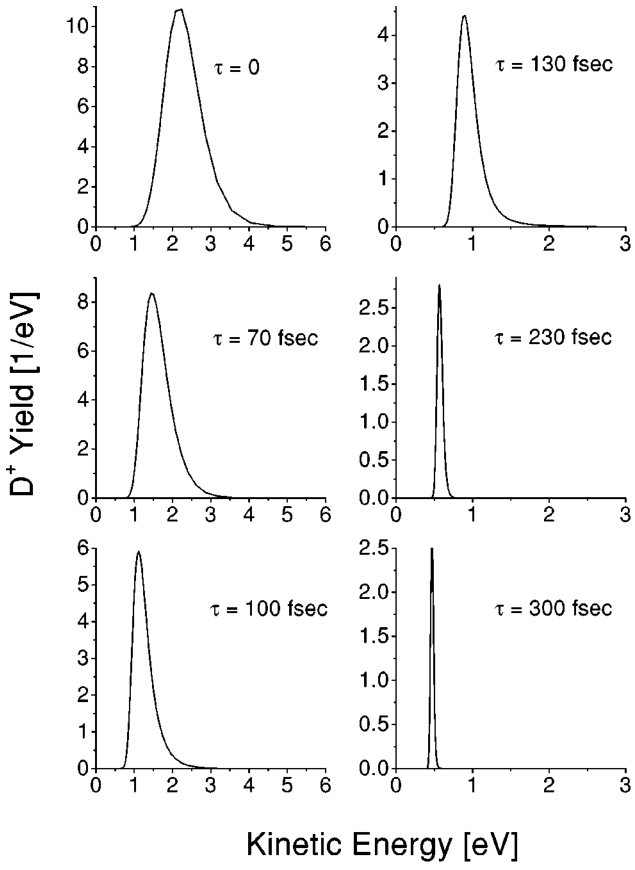

FIG. 4. Calculated D^+ ion kinetic energy distributions formed by photoionization of those $D_2{}^+$ molecules which dissociate in the Floquet one-photon channel. We have chosen the same pump-probe delay times τ and other pulse parameters as for the calculation in Fig. 3.

butions [Fig. 3(b)] with a long high energy tail has two reasons. First, the slope of the repulsive Coulomb potential decreases with increasing internuclear separation. This gives rise to a steep low-energy cutoff of the lines. Second, for delay times smaller than ≈ 130 fsec enhanced photoionization is found near 10 a.u. internuclear separation [see Fig. 3(a)]. This is caused by still overlapping pump and probe pulses which have broad wings since the time dependence of the light intensity has a Lorentzian shape. The significant overlap is certainly responsible for the long high-energy tail at short delay times ($\tau \leqslant 130$ fsec).

In the experiment we did not find an indication of probe pulse photoionization of those molecular ions which dissociate in the Floquet one-photon channel (i.e., by bond softening dissociation [11]). In this channel the dissociation products have a small final kinetic energy of 140 meV (see Fig. 2) and the number of dissociating ions is small. The model calculation easily reveals why a corresponding line is not found in the ion kinetic energy distributions. The result of the numerical evaluation of the rate equations for the $D_2{}^+$ ions dissociating by bond softening is shown in Fig. 4. At probe pulse delay times between zero and 130 fsec the corresponding ion kinetic energy distribution we expect practically coincides with the distribution from photoionization of $D_2{}^+$ dissociating in the Floquet two-photon channel (compare to Fig. 3). The only difference is a slightly larger width of the lines in Fig. 4 at equal delay times. In the experimental ion kinetic energy distributions we expect that the ion yield in the delay-dependent structure originates mainly from the Floquet two-photon dissociation channel ($0 \leqslant \tau \leqslant 130$ fsec). This is suggested by the calculated spectra which at equal delay times give rise to a higher ion yield for dissociation in the Floquet two-photon channel (compare the ion yield axis scales in Figs. 3 and 4) at otherwise equal starting conditions for dissociation. In the measured spectra the real starting conditions are even in favor of the Floquet two-photon channel for delay times smaller than 130 fsec. At 230 fsec and 300 fsec delay time a possible contribution of multiphoton ionization (MPI) of $D_2{}^+$ dissociating in the Floquet one-photon channel can no longer be resolved from the line corresponding to unionized $D_2{}^+$ molecules dissociating in the two-photon channel.

In this paper we presented the observation of delayed probe pulse multiphoton ionization of molecular deuterium ions dissociating in a Floquet two-photon channel in a high-intensity femtosecond pump laser pulse. This photoionization leads to Coulomb explosion of the completely stripped nuclei. The Coulomb repulsion energy appears in D^+ ion kinetic energy spectra and is indicative of the internuclear separation where the probe pulse hits the dissociating molecule. The most interesting perspective of this experiment is nuclear wave function mapping of the dissociating $D_2{}^+$ ion similar to the experiments of Stapelfeldt and co-workers on dissociating molecular iodine [1,2]. This can be done by further reducing the temporal width of the pump and probe pulses into the 10 fsec pulse width regime. This type of experiment may give valuable insight into $D_2{}^+$ nuclear wave function dynamics in a strong field light wave.

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