

Elusive enhanced ionization structure for H_2^+ in intense ultrashort laser pulses

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(Received 21 August 2008; published 19 December 2008)

Enhanced ionization of H_2^+ in intense ultrashort laser pulses was measured using molecular dissociation imaging of an H_2^+ beam and with a pump-probe technique using cold target recoil ion momentum spectroscopy on a D_2 target. Both measurements show a broad, single-peak distribution for enhanced ionization, in contrast to the double-peak structure predicted by “frozen” nuclei theory. This calculated double-peak structure is most likely washed out by nuclear motion and to a lesser extent by intensity averaging.

DOI: [10.1103/PhysRevA.78.063419](https://doi.org/10.1103/PhysRevA.78.063419)

PACS number(s): 33.80.-b, 32.80.Fb

I. INTRODUCTION

The ionization of H_2^+ by an intense ultrashort laser pulse is enhanced for internuclear distances larger than the equilibrium internuclear distance of this molecular ion. This phenomenon is commonly invoked in order to explain the low kinetic energy release (KER) of the “Coulomb exploding” protons (see, for example, the reviews [1,2]).

Zuo and Bandrauk [3] suggested that this enhancement in ionization is due to charge resonance enhanced ionization (CREI) around some critical internuclear distances that are larger than the bond length of H_2^+ . Their calculations predicted two prominent peaks in the ionization rate centered about an internuclear distance, R , of 7 and 10 a.u. (atomic units are used hereafter unless otherwise specified), which initiated further theoretical work on the structure of CREI (e.g., [4–6]). Furthermore, this intriguing structure motivated experimental work trying to reveal it. In a simplified two Coulomb-well model [3], CREI occurs when the bound state in the upper well is above the potential barrier between the two wells. Therefore, one would expect CREI to be independent of the laser color and shift slowly to smaller R with increasing laser intensity.

Gibson *et al.* [7], for example, tailored the experimental conditions to enable the study of CREI of the transient H_2^+ formed early in the laser pulse from H_2 . Williams *et al.* [8], on the other hand, directly studied the ionization of an H_2^+

molecular ion beam. These measurements are in good agreement with each other, and with the theoretical prediction that ionization is enhanced for stretched molecules. However, as shown in Fig. 1, the data does not support the prediction of a large second maximum around $R=10$. It is important to note that in both experiments the internuclear distance was evaluated from the measured KER in order to compare the experimental data to “frozen”-nuclei calculations. To perform this transformation one must adopt some model, and doing so will affect the results as discussed later in this paper. Explicitly, the latter group used the Coulomb explosion model (i.e., $KER=1/R$), while the former subtracted the average dissociation energy of the H_2^+ from the measured KER before employing the Coulomb explosion model.

It has been suggested that the second enhanced ionization peak is hard to observe because of depletion by ionization at the first enhanced ionization. Furthermore, the second enhanced ionization peak yields low KER, therefore making it hard to distinguish from the fragments of dissociation in these experiments [8].

Recently, Ergler *et al.* [9] reported that they found a clear signature for the second enhanced ionization maximum using a pump-probe technique, that is, ionizing H_2 and launching a nuclear wave packet with the pump pulse and ionizing the stretching H_2^+ with a second short pulse. The measured ionization rate as a function of time delay between the two pulses and KER allows for the identification of the dissocia-

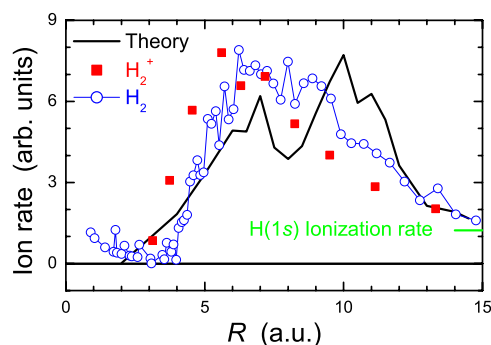


FIG. 1. (Color online) Ionization rate as a function of the H_2^+ internuclear distance, R (H_2^+ data for $I_0=3 \times 10^{15}$ W/cm² from Ref. [8], H_2 data for $I_0=3.2 \times 10^{14}$ W/cm² from Ref. [7], theory [3]).

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tion pathway before ionization by the second pulse [9,10]. This measured time delay is then converted to internuclear distance information, again assuming some model.

Questions have been raised about this recent evidence for the second enhanced ionization peak, however, as it appears where the ionization of the one-photon and two-photon dissociating processes overlap, i.e., at the time delay where H_2^+ undergoing bond softening (BS) or above-threshold dissociation (ATD) is ionized. To make the situation even more interesting, recent preliminary experimental efforts to uncover this structure [11,12] have reached nearly opposite conclusions about its existence.

In this paper we present these pump-probe measurements together with measurements of the ionization of an H_2^+ beam for which ionization was directly separated from dissociation, thus enabling the observation of the low KER contribution to ionization. The results of both measurements are consistent with each other, revealing no evidence for the elusive second enhanced ionization peak, but rather a single broad KER peak. We further suggest that the predicted structure in the ionization probability for frozen nuclei is washed out by the motion of the nuclei during the laser-molecule interaction and by intensity averaging.

II. EXPERIMENT

The ionization of H_2^+ by an intense ultrashort laser pulse was measured using two experimental techniques described briefly below. One involved interrogating an H_2^+ ion beam by intense laser pulses while in the other these ions were generated by a pump pulse, which also initiated their dissociation. The ionization in the latter experiment was caused by a second probe pulse.

Ionization of a 5 keV H_2^+ beam originating from an ion source was measured using three-dimensional (3D) coincidence imaging of both molecular fragments as described in more detail elsewhere [13,14]. Briefly, the H_2^+ beam, having a vibrational population given approximately by a Franck-Condon distribution [15], was ionized by a 40 fs, 790 nm, linearly polarized laser pulse with a peak intensity of up to about 1.7×10^{15} W/cm². The resulting protons were measured in coincidence by an imaging setup providing the momentum vector of each fragment. Random coincidences between protons originating from two H_2^+ ions dissociated by the same laser pulse were discriminated by imposing momentum conservation conditions on the measured momenta. A weak constant electric field in the beam direction was used to completely separate the ionization and dissociation channels [13]. Thus, the KER distribution of the ionized H_2^+ is measured over the whole energy range of interest, including where it overlapped with the high-KER tail of the dissociating H_2^+ (i.e., the $\text{H}^+ + \text{H}$ channel) in previous measurements. As noted before [8], the second enhanced ionization peak is expected to yield KER values exactly in this overlap range.

The pump-probe measurements were conducted using a cold target recoil ion momentum spectroscopy (COLTRIMS) technique described in detail elsewhere [10]. This technique is represented schematically on the potential energy curve diagram in Fig. 2. Briefly, a cold supersonic jet of molecular

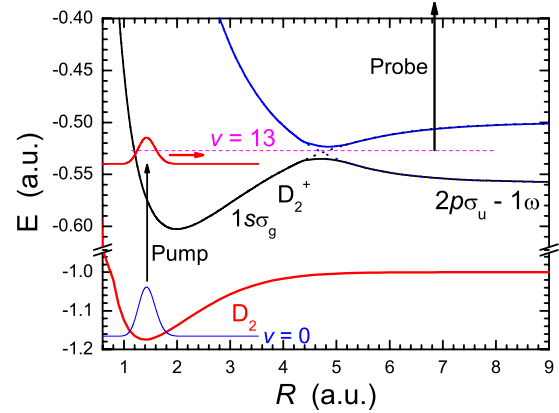


FIG. 2. (Color online) A schematic potential energy curve (PEC) diagram for the D_2 double ionization and dissociation in pump-probe experiments. The time delay between the pump and probe allows the D_2^+ created by the pump pulse to stretch to a larger internuclear distance, R , where the probe pulse ionizes it.

hydrogen (D_2 was used for practical reasons) was crossed with a linearly polarized laser beam from the same Ti:sapphire chirped-pulse amplification laser system as in the H_2^+ measurement. Shorter laser pulses of about 10 fs (centered around 790 nm) were used to reduce the overlap in time between the pump and the probe. The pump pulse, with a peak intensity of 3×10^{14} W/cm², ionized the D_2 target molecules and also launched the nuclear wave packet of the reaction channel of interest, namely bond softening. This pulse was followed by a more intense probe pulse, with a peak intensity of 9×10^{14} W/cm². The time delay between the two pulses was scanned over a 100 fs range while maintaining a good spatial overlap as explained by Alnaser *et al.* [10].

III. RESULTS AND DISCUSSION

The measured KER at 1.7×10^{15} W/cm², shown in Fig. 3(a), clearly shows a single broad peak around 4.7 eV and not a double-peak structure. This distribution is similar to the one measured previously [8], but the uncertainty at the low KER end has been removed. The KER spectrum measured at a lower intensity of 4.3×10^{14} W/cm², also shown in Fig. 3(a), shows a similar single broad peak that is shifted to somewhat lower energies. This small shift to lower energies with decreasing peak intensity has been attributed to ionization at larger internuclear distances in both H_2^+ [16–18] and H_2 [19–21] targets. The rate of ionization sharply drops with decreasing intensity as indicated by the lower number of counts—all normalized to the same beam current and number of laser pulses.

It is expected that the interaction between the laser field and the H_2^+ will initiate dissociation of the vibrational states within the energy gap at the avoided crossing, as shown in Fig. 3(b). Taking the $v=9$ state, which is expected to be the first state to dissociate, and adding the kinetic energy gained during the dissociation up to $R=7$ (where the first CREI peak is expected) to the energy gain for the Coulomb repulsion at this distance yields 4.6 eV, thus suggesting that the measured KER peak is associated with the first enhanced ionization

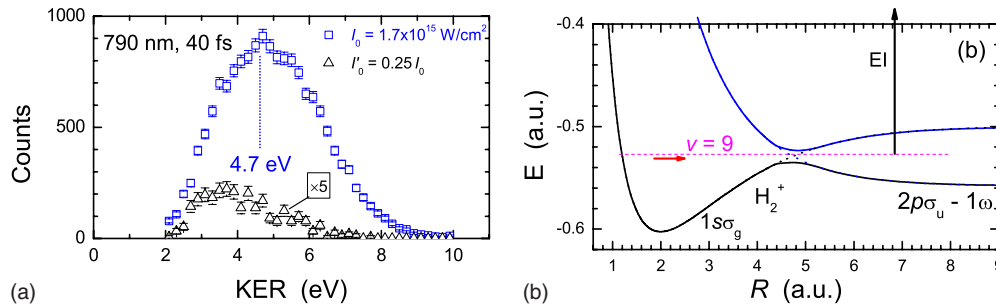


FIG. 3. (Color online) H_2^+ ionization measurements. (a) Measured KER distribution of H_2^+ in 790 nm 40 fs laser pulses: Open blue squares, $I_0 = 1.7 \times 10^{15} \text{ W/cm}^2$; open black triangles, $0.25I_0$. Note that the lower intensity data was multiplied by a factor of 5 in order to present it on the same scale. (b) A schematic PEC diagram for the H_2^+ ionization and dissociation. The arrow labeled EI (enhanced ionization) marks the internuclear distance where ionization is expected to be enhanced.

peak. In contrast, ionization at $R=10$, where the second CREI peak is expected, yields about 3.4 eV, and the data does not show any significant structure around this energy. Neither does it show any contribution at the low KER reported recently by Pavičić *et al.* [16] for enhanced ionization of a D_2^+ beam exposed to $\sim 1 \times 10^{14} \text{ W/cm}^2$, 350 fs laser pulses. The structure they observed around $R=8$, 11, and 15 a.u. might be due to the opening of multiphoton ionization pathways at the respective ionization appearance intensities [22].

The absence of a second CREI peak is a central result of this paper, so we critically explore possible explanations for not seeing it.

One may argue that the pulse intensity might drop before the dissociating H_2^+ stretches to $R=10$, thus suppressing the second enhanced ionization peak. However, given that dissociation of the $v=9$ state begins at intensities well below 10^{13} W/cm^2 [14], we expect the intensity to still be increasing during the passage through $R=10$ for a 40 fs laser pulse as it takes about 13 and 22 fs for the H_2^+ to stretch from the curve crossing to $R=7$ and 10, respectively.

Another possible concern is that the population of the $v=9$ state was depleted by the enhanced ionization around $R=7$. However, our data suggests the contrary, as the measured rate of dissociation of this vibrational state is not negligible [18], thus indicating that a significant fraction of the H_2^+ molecules passed through both enhanced ionization regions without being ionized.

To further clarify the two issues discussed above, we studied the time evolution of the dissociating nuclear wave packet of each vibrational state of H_2^+ using the standard approach of solving the time-dependent Schrödinger equation in the Born-Oppenheimer representation. In Fig. 4 we show snapshots of these dissociating wave packets at the time the (40 fs, 790 nm, $1 \times 10^{14} \text{ W/cm}^2$) laser pulse reaches its peak intensity, where ionization is most likely. It is clear from the figure that the $v=8$ and 9 states cannot participate in enhanced ionization as they pass through the whole CREI region, between 7–10 a.u., long before the laser pulse reaches a high enough intensity to ionize the molecule. This result agrees with our observations [18] of a large dissociating component of H_2^+ even for peak intensities limited to the range [14] between 8.7×10^{14} and $1.7 \times 10^{15} \text{ W/cm}^2$. Also, the low-lying vibrational states, $v \leq 2$, do not contrib-

ute to enhanced ionization as they never stretch far enough. More importantly, these calculations reveal the intricate nature of the competition between ionization and dissociation as both depend on dynamics driven by the specific laser pulse.

The main states that contribute to enhanced ionization at the peak intensity of the laser pulse, shown in Fig. 4, are $v=6$ and 7—following their bond softening, and the $v=3$ —following its above-threshold dissociation. It would appear that the vibrationally trapped part of the higher vibrational states, i.e., $v \geq 10$, contributes to enhanced ionization, but it is known that this simple aligned-nuclei model for H_2^+ grossly overestimates the role of vibrational trapping [23]. In any case, the vibrational population of our H_2^+ beam falls off rapidly with increasing v above $v=3$, thereby limiting the contribution to enhanced ionization from the vibrationally trapped states.

To elucidate the fact that the H_2^+ target spans a wide range of internuclear distances when ionization becomes possible we show the total distribution of the dissociating nuclear wave packet as a function of internuclear distance, $P(R)$, in Fig. 5. This distribution was computed by adding the contributions from all vibrational states incoherently with weights given by the Franck-Condon population of the H_2^+ beam. Clearly, there is population available for enhanced ionization over a wide range of internuclear distances. It is important to note that for the calculated pulses, the population at $R=10$ a.u. is smaller by about a factor of 2 relative to the population at $R=7$ a.u. However, this should not prevent the observation of the predicted CREI structure, shown in Fig. 1, but rather make the two peaks comparable to each other. This indicates that depletion at the first CREI peak does not occur for an H_2^+ beam target, though it might in studies using an H_2 target where the H_2^+ is created at a relatively high intensity. Furthermore, the exact $P(R)$ distribution may change with laser pulse parameters, in particular the pulse duration. As shown in Fig. 5, reducing the peak intensity only reduces the overall population, while the relative population between $R=7$ and 10 a.u. changes slowly. Therefore, intensity averaging is not expected to change dramatically the ability to observe the predicted CREI peaks.

Finally, assuming the main contribution to enhanced ionization in our measurements comes from the $v=6$ and 7 states, the kinetic energy gained upon dissociation is some

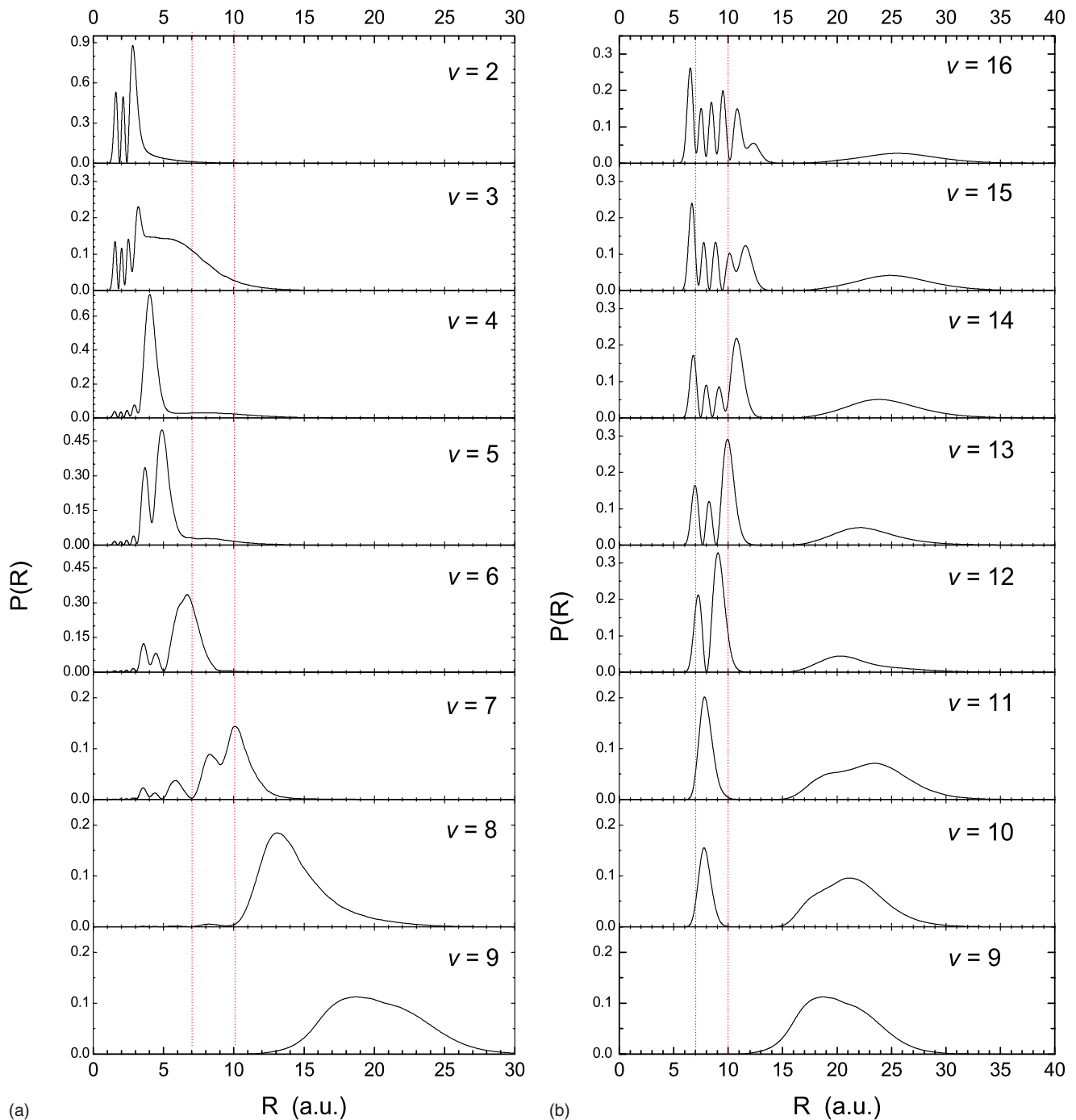


FIG. 4. (Color online) H_2^+ theory. Computed nuclear wave packets at the laser peak intensity for a 790 nm, 40 fs, 1×10^{14} W/cm² pulse. The different panels depict a snapshot of the R distribution of the different vibrational levels.

what lower than the one used previously for the $v=9$ state. We would therefore expect ionization at $R=7$ a.u. to yield 4.3 eV instead of 4.6 eV. The somewhat higher KER may indicate contributions of the $v=8$ state or, more likely, ionization at somewhat smaller R values.

To summarize our H_2^+ beam studies, they clearly suggest that enhanced ionization occurs over a wide range of internuclear distances as predicted [3] and observed in many previous studies (see, for example, Ref. [2]). However, our measurements suggest that the predicted structure in the ionization rate as a function of internuclear distance [3] does

not lead to structure in the KER as some would expect, but rather shows a single broad peak.

We complement our ion beam studies with a pump-probe measurement on a D_2 target (the D_2 isotope was used over the H_2 for practical reasons as both are expected to exhibit similar behavior in ionization). In such measurements depletion at a smaller R cannot occur. In this case the time evolution of the dissociating wave packet is initiated by the first ultrashort laser pulse, i.e., a nuclear wave packet is launched. The ionization probability at different internuclear distances is then probed by a delayed ultrashort pulse.

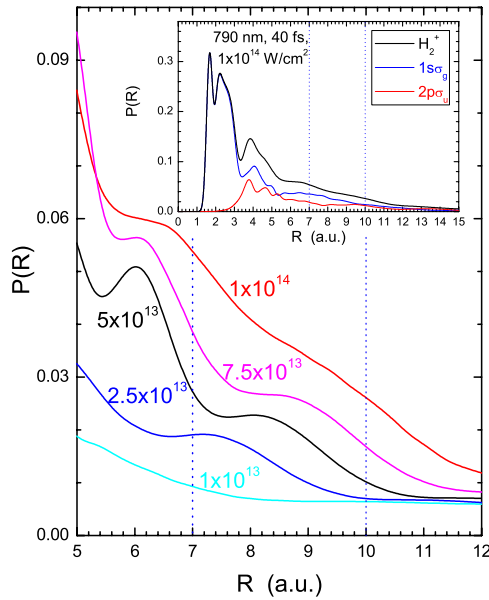


FIG. 5. (Color online) H_2^+ theory. The distribution of internuclear distances for a Franck-Condon averaged vibrational population of H_2^+ at the moment the laser intensity peaks, computed for a 790 nm, 40 fs laser pulse at a few peak intensities. In the inset we show a more complete R range and the fractional population of the $1\sigma_g$ and the $2p\sigma_u$ states out of the total $n=1$ manifold (the contribution of the $n=2$ manifold is small in this case but can play an important role in other cases [24]). Note that the population at $R=7$ a.u. and $R=10$ a.u. is decreasing with decreasing laser intensity, but the relative population does not change significantly.

The peak intensity of the pump pulse was kept low enough to minimize contributions from ATD. The bond softening and above-threshold dissociation mechanisms can be tracked in time and distinguished from each other by the trace they follow on a KER vs time-delay plot (e.g., Refs. [9,10]). It has been shown by Alnaser *et al.* [10] that only bond softening has a significant contribution to the time-delay KER data shown in Fig. 6(b). Furthermore, they showed that this data is well described by a classical time evolution of the dissociating D_2^+ molecule on the $2p\sigma_u$ state starting from the curve crossing with the $1\sigma_g$ state a short time (~ 12 fs, i.e., roughly the time it takes the nuclear wave packet of D_2^+ to reach the crossing) after the pump pulse [see line in Fig. 6(b)]. This model allows the evaluation of the internuclear distance as a function of time and the conversion from time delay to R shown on the top axis of Fig. 6(a). It can clearly be seen from this distribution that ionization is enhanced around $R=7$ and, more importantly, that the second enhanced ionization peak around $R=10$ is missing. Of course, that does not suggest that enhanced ionization does not occur at $R=10$, just that the prominent predicted peak at that internuclear distance, clearly visible in Fig. 1, is replaced by a low ionization rate in the measured data associated with this internuclear distance. More importantly, independent of any model to convert to R , the raw data exhibits a single enhanced ionization peak in agreement with our H_2^+ results shown before.

Both the ionization of an H_2^+ ion beam and the pump-probe measurements of a D_2 target yield the same qualitative

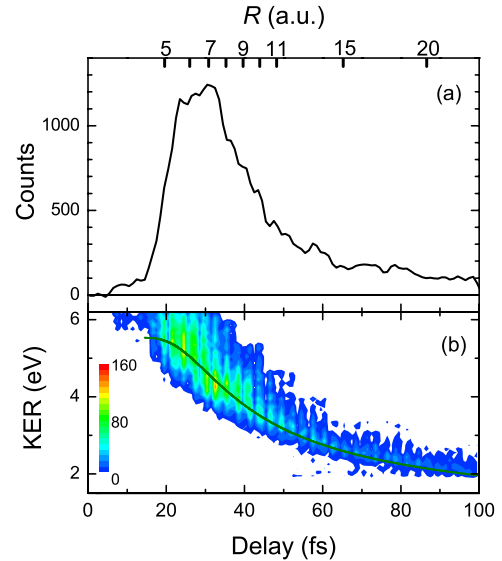


FIG. 6. (Color online) D_2 pump-probe measurements. (a) Measured time-delay distribution (note the associated value of R marked on the top axis), and (b) measured time-delay KER distribution. Note that the data in (b) follows the calculated BS dissociation path—solid line (see text). As stated in the experimental section, the pump and probe pulses were 10 fs long with peak intensities of 3×10^{14} and 9×10^{14} W/cm², respectively.

result. Namely, ionization is enhanced as predicted, but instead of the predicted double-peak structure, only a single broad peak is observed. It is important to note that in both measurements the KER is a measurable, but the internuclear distance R is not. Thus, the best comparison with theory can be accomplished if theory computes the measurable quantities for each experiment directly. In addition, it is important to include the nuclear vibrational motion. Freezing the nuclei for the computation of the ionization probability might simplify the problem, but there is no realistic way to measure the ionization of stretched molecules while keeping the nuclei frozen. Moreover, the dissociation energy is not negligible in comparison with the Coulomb explosion energy in the case of enhanced ionization.

Lacking calculations of the specific observable quantities in our measurements, we are forced to convert the KER data to an internuclear distance in order to compare with theory, in particular with the predicted structure. As a byproduct, this conversion facilitates a direct comparison of the two measurements. Following the success of the classical time evolution model in fitting the time-delay KER distribution of ionization from the bond softening dissociation of D_2 [10], we applied a similar model to the dissociation of the H_2^+ target. As discussed previously, we expect the $v=9$ state to dissociate through the energy gap, shown in Fig. 3(b), early in the laser pulse and gain a kinetic energy $E_D(R)$ with respect to the $2p\sigma_u$ curve. Upon ionization at R the protons gain an additional energy of $1/R$, thus resulting in a measured $KER = E_D(R) + 1/R$, where $E_D(R) = E_{v=9} - E_{2p\sigma_u-1\omega}(R)$. Using this model the measured KER distribution shown in Fig. 3(a) was converted to the R distribution shown in Fig. 7(a), where it is also compared with the distribution derived from the pump-probe measurements. The agreement between

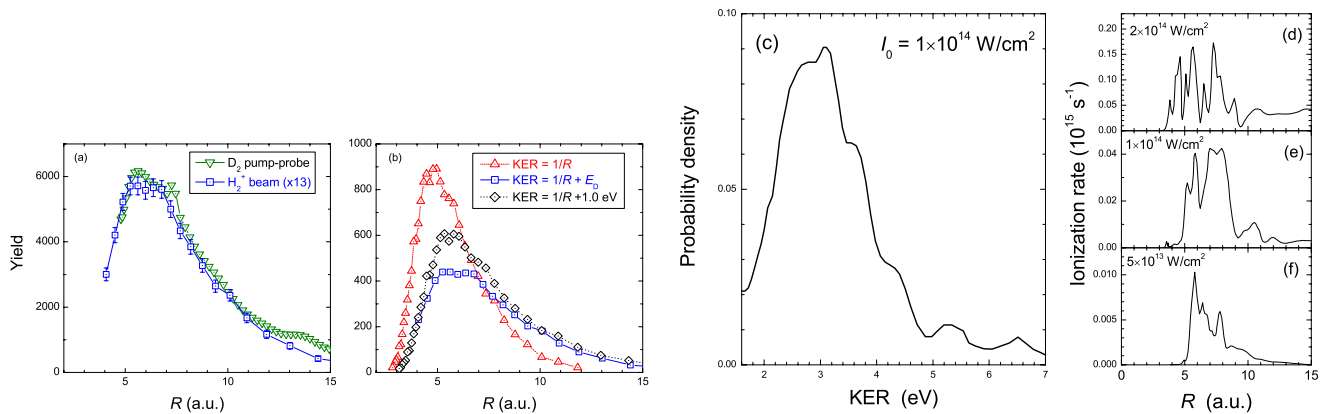


FIG. 7. (Color online) (a) The measured ionization rate as a function of internuclear distance, R , derived from the H_2 pump-probe and H_2^+ molecular-ion beam measurements (see text). (b) Comparison of a few models used to convert the measured KER to R . Explicitly, the Coulomb explosion model for which $KER=1/R$, adding the average dissociation energy (1.0 eV) [7], and our model $KER=1/R+E_D$ (see text). CREI theory from Chelkowski *et al.* [25]: (c) Proton KER spectrum including nuclear motion, and (d)–(f) frozen nuclei calculation for a few peak intensities. Note that the KER range in (c) matches the relevant range of R in (d)–(f).

the two is very good except for the high R tail which is associated with the low KER (or long time-delay) data that is of lesser quality. This conversion, however, is very sensitive to the model used to transform the KER to R , as shown in Fig 7(b), and it should be used with caution.

Furthermore, converting the measured H_2^+ KER distribution using the $E_D(R)$ that is expected for a net two-photon ATD pathway yields a similar result. This reinforces the statement above preferring a direct comparison with theoretical calculations of the measurable quantities. However, in spite of these limitations, the data strongly suggests the existence of a single broad enhanced ionization peak. In other words, no second maximum.

The main question remaining is why not?

There are a couple of reasons which are most likely responsible for the absence of the second peak. First, and most important, the nuclear motion washes out the structure as was demonstrated by Chelkowski *et al.* [25] in calculations for a 40 fs, 600 nm laser pulse interacting with H_2^+ , shown in Figs. 7(c)–7(f). The structure of the ionization rate for frozen nuclei, shown in panel (e), for example, washes out dramatically when the nuclear motion along R is included and the expected KER is computed, as shown for the same peak intensity in panel (c).

In addition, the structure in the ionization rate changes significantly with laser intensity, as seen in Figs. 7(d)–7(f). Experimentally, the spatial distribution of intensities imposes

an intensity averaging on any measurement, further smearing any expected structure. If there is to be any hope of observing such structures, it is crucial to have calculations including these effects to suggest under what conditions they might be observed.

IV. CONCLUSION

In conclusion, we have presented compelling experimental evidence suggesting that H_2^+ ionization for high intensity short laser pulses is enhanced for a wide range of internuclear distances, approximately around $R=7$. This distribution was found to be structureless in contrast to the recent structure found near the ionization appearance intensity [22]. More importantly, it is suggested that the structure predicted by frozen nuclei calculations is washed out by nuclear motion, and that intensity averaging effects should smear any structure further. More complete calculations that closely follow the experimental conditions and directly yield the observable quantities are needed in order to observe this elusive structure.

ACKNOWLEDGMENTS

Supported by the Chemical Sciences, Geosciences and Biosciences Division, Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy.

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