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Time slicing in 3D momentum imaging of the hydrogen molecular ion photo-fragmentation

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Photo-fragmentation of the hydrogen molecular ion was investigated with 800 nm, 50 fs laser pulses by employing a time slicing 3D imaging technique that enables the simultaneous measurement of all three momentum components which are linearly related with the pixel position and slicing time. This is done for each individual product particle arriving at the detector. This mode of detection allows us to directly measure the three-dimensional fragment momentum vector distribution without having to rely on mathematical reconstruction methods, which additionally require the investigated system to be cylindrically symmetric. We experimentally reconstruct the laser-induced photo-fragmentation of the hydrogen molecular ion. In previous experiments, neutral molecules were used as a target, but in this work, performed with molecular ions, the initial vibrational level populations are well-defined after electron bombardment, which facilitates the interpretation. We show that the employed time-slicing technique allows us to register the fragment momentum distribution that reflects the initial molecular states with greater detail, revealing features that were concealed in the full time-integrated distribution on the detector. Published by AIP Publishing. [http://dx.doi.org/10.1063/1.4974743]

I. INTRODUCTION

New opportunities and interest in understanding problems in molecular dynamics such as photo-ionization and photo-dissociation processes have been generated with both fast timing and 2-dimensional position determination abilities in new ion detector systems. High resolution and efficiency are achieved by combining micro-channel plate (MCP) detectors with delay-line anodes and make it possible to measure the three components of the momentum vector for each charged fragment from the dissociation processes.1,2 Coincidence measurements involving two or more charged fragments are improved by the performance of the detectors with better spatial and temporal detection resolution, efficiency, and reliability, as well as a significantly decreased dead time. Simultaneously, having available delay-line anodes with a three-layer hexagonal design, a better precision of the time digitizers and advanced signal-reconstruction algorithms was obtained.3 Most notably, unless the difference between their times of impact and spatial separation is very small, it is possible to separate explicitly the signals produced from the two charged fragments that hit the detector at the same time, but at different positions, or the same position, but at different times.4

Our experiment contains such delay-line detectors and builds on Figg’s groundbreaking work ranging from spectroscopy of helium hydride5 to strong-field dissociation of the hydrogen molecular ion.6–12 The apparatus can produce an intense beam of many kinds of molecular ions based on the duo-plasmatron principle, where the electrons from the cathode produce a plasma in the gas introduced at low pressure. The positive ions are then extracted. We have measured current densities of more than 0.35 μA/mm² for the hydrogen molecular ions. The ion beam apparatus now features a MCP detector (HEX 80, RoentDek) with <100 μm position resolution and ~1 ns time resolution.

Despite the unique information that can be gained by using the hydrogen molecular ion targets, these studies are rare due to the additional experimental complexity of using an ion beam as a target. Although theoretical studies commonly use the hydrogen molecular ion for model calculations due to its simplicity (e.g., Refs. 13 and 14), experimentalists study the more accessible neutral hydrogen molecule (e.g., Refs. 15 and 16). In some experiments with the hydrogen molecule, the experimental conditions are tailored to enable the study of the transient hydrogen molecular ions that are formed early in the interaction with a laser pulse (e.g., Refs. 17 and 18). However, the physics of the hydrogen molecular ion in an intense laser beam is more clearly understood by directly using the hydrogen molecular ion as a target. We note that the result of the action of the same laser pulse on a molecular ion in an ion beam can be expected to differ from that on a neutral hydrogen molecule. Indeed, for the former, the dissociation can be affected by excited vibrational states, while for the latter the ground state before ionization has lower energy, and therefore, it is less likely to dissociate.

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We obtain results on the photo-fragmentation of the hydrogen molecular ion with ultrashort laser pulses by employing a time-sliced 3D imaging technique. Time slicing techniques have been successfully used in other experiments such as time-of-flight mass spectrometers to avoid the effects due to intensity averaging. We experimentally reconstruct the laser-induced photo-fragmentation of the hydrogen molecular ions prepared by electron impact ionization, while the neutral molecules are used as a target in most previous experiments. We describe our ion beam apparatus and its potential for future applications.

II. EXPERIMENTAL DETAILS

As it is shown in Fig. 1, our ion beam apparatus has three vacuum chambers, and each chamber is pumped by a turbo molecular pump. The main components are an ion source, a magnet for mass selection, different size apertures for beam collimation, and Faraday cups for measuring the ion current.

In our experiment, a duo-plasmatron ion source with a dc electric discharge was used to generate the molecular ions. Neutral hydrogen gas is sent into the region between an anode and a hollow cathode. The electrons are emitted from the hollow cathode, and a potential difference of 450–600 V accelerates them towards the anode. The intermediate electrode is held at a potential of −100 V with respect to the anode, so it electrostatically concentrates the electron flux. In addition, we have a solenoid magnet with stabilized current (usually in the range of 1 to 1.8 A) surrounding the ion source. The applied magnetic field compresses the plasma and towards the anode, the magnetic field strength increases so that their radial velocity component increases while the axial component decreases when the electrons come close to the anode. Hence, the electrons’ density is getting higher near the anode in order to increase the ionization efficiency of the neutral hydrogen molecules. A high potential difference between the anode and an extraction electrode is applied to extract and accelerate the positive molecular ions from the plasma. In order to extract the positive ions, the whole ion source was held at a positive potential of 12 kV with respect to the rest of the apparatus that was grounded. The ion current is typically around 100 μA with the anode having an aperture of 150 μm. This current of the extracted ions could be optimized by adjusting the solenoid magnet current surrounding the ion source.

We used a set of vertical and horizontal electrostatic deflection plates (DP1) and an Einzel lens (EL1) in order to direct the extracted positive molecular ions into a sector magnet (SM) through the entrance slit A1 (width of 5 mm). Then by changing the magnetic field of the magnet, we deflected the hydrogen molecular ions by 90° to pass through the exit slit A2 (width of 5 mm). The second set of electrostatic deflection plates (DP2) is used to steer the selected ion beam. Then the beam enters Einzel lens EL2. The combination of Einzel lenses EL1 and EL2 is used as an electrostatic telescope to collimate the ion beam and maximize the ion beam current.

To achieve the alignment of the ion beam, the ion current is measured at different points along the beam path. There are two collimating apertures: A4 (300 μm circular aperture) and A7 (a 300 × 25 μm aperture) and the secondary pre-apertures A3 and A6 with diameters of 2 mm and 1 mm, respectively, were placed in front of each collimating aperture to make the alignment easier. All insulated apertures and Faraday cups FC1 and FC2 were connected to a switchable electrometer (Keithley Instruments, model 617). Intermediate FC1 was set on a side and used for adjustments of the ion beam, i.e., H₂⁺ ion beam could be redirected to FC1 by the deflecting plates. The FC2 in front of the MCP detector was positioned in the center to block the unfragmented H₂⁺ ions and to minimize the blocking of the detected fragment (H and H⁺) structures in the outer areas of the MCP.

For the alignment, we first found the best combination of voltages on the Einzel lenses, the deflection plates, and the sector magnet by optimizing the ion current at aperture A4 and its pre-aperture A3. After that, we followed the same procedure for the aperture A7 and its pre-aperture A6. Finally, we maximized the current on the Faraday cup FC2. We repeated the whole procedure several times with similar initial settings to achieve an optimal and sufficient ion current.

![Diagram of the ion beam apparatus](https://example.com/diagram.png)

**FIG. 1.** Layout of the ion beam apparatus. IE: intermediate electrode; DP: deflection plates (13 mm × 22.5 mm × 0.85 mm); EL: Einzel lenses; SM: sector magnet, R = 150, B = 10G, 8A; A1, A2: two perpendicular slits of 5 mm width each; A3: 60 mm circular plate with 2 mm circular hole, A4: 60 mm × 60 mm square plate with 300 μm circular hole, A5: 8 mm circular hole, A6: 60 mm × 60 mm square plate with 3 mm circular hole, A7: 25 μm × 200 μm slit and 1 mm × 1 mm square hole; IA: ion accelerator; S: vertical slit with 0.75 mm gap; IG: ion gate, 92 mm × 40 mm deflection plates with 5 mm separation; L: focusing lens; FC1, 2: Faraday cups; MCP: microchannel plate detector. Inset (a): Interaction volume defined by the crossing of a laser beam (red cylinder) and an ion beam (a blue box). Inset (b): Schematic diagram of the geometry for the beam collimation along the x-axis.
The experimental resolution depends on the width and divergence of the ion beam. They are determined by the sizes of the collimating apertures along the laser polarization axis (z axis) and the positions of the apertures and the detector on the ion beam axis. The collimation geometry in the x-z plane is shown in inset (b) of Fig. 1. The pair of outer lines shows the maximum allowed divergence of the beam, and the inner lines illustrate the collimated ion beam. The widths on the detector are indicated by the letters s and S. The divergence angles \( \alpha = 0.25 \text{ mrad} \) and \( \beta = 0.30 \text{ mrad} \) and the widths of the ion beam \( s = 701 \mu \text{m} \), \( S = 884 \mu \text{m} \) can be easily calculated from the similarity of the triangles. By using a 300 \( \mu \text{m} \) circular aperture A4 and a 300 \( \mu \text{m} \times 25 \mu \text{m} \) aperture size A7 for collimating the ion beam, we could optimize the current of the 12 keV hydrogen molecular ion beam measured on the Faraday cup FC2 up to 3–4 nA.

The collimated, velocity bunched, and therefore nearly monochromatic hydrogen molecular ion beam was crossed with the focused laser beam at right angle. Laser pulses from a chirped pulse amplification laser system with a repetition rate of 1 kHz, pulse energies of 1.0 mJ, duration of 50 fs, and centered at the wavelength of 800 nm were focused by an achromatic lens. The focal position of the femtosecond laser beam was adjusted coarsely before the actual experiment by observing the position of the laser-induced optical breakdown, when the apparatus was filled with air. The overlap of the two beams was assured by using a plate with a 50 \( \mu \text{m} \) pinhole, placed at 45° with respect to the ion beam that passed through it, and then the laser beam was adjusted to pass through it. Our focus diameters were greater than the height of the rectangular molecular ion beam (25 \( \mu \text{m} \)), and the Rayleigh length was large compared to the width of the molecular beam (200 \( \mu \text{m} \)), which reduced the volumetric effect of the intensity variations within the interaction region (Fig. 1, inset (a)). We have a laser beam diameter of 60 \( \mu \text{m} \), a molecular ion velocity of \( 1.5 \times 10^6 \text{ m/s} \), and a typical current of about 80 nA in the interaction region, which corresponds to ~40 molecules within the cross section of the focused laser beam.

Under the action of laser pulses, the hydrogen molecular ion could dissociate into fragments, \( \text{H}_2^+ \rightarrow \text{H}^+ + \text{H} \), which continued moving toward the MCP detector, and this time-of-flight (TOF) part of the apparatus is in essence a TOF spectrometer. The dissociated fragments were detected by the three-dimensional imaging system, namely an arrival time signal at the MCP detector, and the position signals from the anode delay line were recorded allowing the evaluation of the fragment momentum. The distance between the interaction region of the laser radiation with the ions and the detector was 140 cm. All TOF information was measured relative to the start signal from a photodiode triggered by the laser pulse.

The data analysis involves the reconstruction of the momenta of the ions after calibrating all the necessary experimental parameters. The ion detector provides the timing and position information of the detected particles. Once the distance of the detector from the interaction region (b), the value of the electric field (E) over the length of the TOF spectrometer, and the time-of-flight \( t_i \) and the hit position \( (x_i, y_i) \) are known precisely, the three components of momentum gained by an ion in the laser field in three mutually perpendicular directions \( p_{x,i}, p_{y,i}, p_{z,i} \) can be calculated by using simple relations,

\[
p_{x,i} = m_i(x_i - x_{\text{orig}})/t_i, \quad p_{y,i} = m_i(y_i - y_{\text{orig}})/t_i, \quad p_{z,i} = m_i(b/t_i - \text{Eq.} t_i/2),
\]

where \( m_i \) and \( q_i \) are masses and charges of the fragments; \( x_{\text{orig}} \) and \( y_{\text{orig}} \) are the coordinates of the dissociation point. In this equation the timing information of the detected particle is provided by the difference between the “start” and the “stop” signals from the photodiode and the MCP detectors with the correction, which depends on the relative delay of the signals from the photodetector to the MCP extrapolated to zero TOF distance. The two transverse positions, \( x_i \) and \( y_i \), of the ion are obtained from the position sensitive detector.

In a photon-induced dissociative ionization process with two fragments, conservation of linear momentum determines that the fragments have equal values of momenta, but with opposite directions. Using momentum conservation, we obtain the following expressions:

\[
x_{\text{orig}} = \frac{m_1x_1 + m_2x_2}{m_1 + m_2}, \quad y_{\text{orig}} = \frac{m_1y_1 + m_2y_2}{m_1 + m_2}. \tag{2}
\]

In fact, the dissociation point is localized along the x-axis by the focusing spot size (~60 \( \mu \text{m} \)), which is less than the ion beam diameter, and along the y-axis by the smallest of the doubled Rayleigh range (~0.6 mm) and the ion beam extension in the y direction (0.2 mm). Then imposing the conditions of Eq. (2) and the additional conditions

\[
m_1(x_1 - x_{\text{orig}})/t_1 = m_1(x_2 - x_{\text{orig}})/t_2, \tag{3}
\]

only those pairs of arrival events \( (t_1, x_1, y_1), (t_2, x_2, y_2) \) on the detector are selected, which satisfy momentum conservation with the temporal uncertainty defined by the accuracy of time measurement (~1 ns). Such filtering considerably reduces the background of false counts. The sum-momenta distributions then are approaching \( \delta \)-functions: they are essentially nonzero only in the narrow region close to the \( x \) and \( y \) zeros.

Assuming the absence of the field for the kinetic energy release (KER), we obtain similar to Ref. 24,

\[
\text{KER} = \frac{(p_{\text{rel},x}^2 + p_{\text{rel},y}^2 + p_{\text{rel},z}^2)(m_1 + m_2)}{2m_1m_2}, \tag{4}
\]

\[
p_{\text{rel},x} = \frac{(p_{x,2} - p_{x,1})/2}{m_1m_2(x_2 - x_1)/m_1t_2 + m_2t_1},
\]

where

\[
p_{\text{rel},y} = \frac{(p_{y,2} - p_{y,1})/2}{m_1m_2(y_2 - y_1)/m_1t_2 + m_2t_1},
\]

\[
p_{\text{rel},z} = \frac{(p_{z,2} - p_{z,1})/2}{(m_1 + m_2)t_1t_2}.
\]

III. RESULTS AND DISCUSSION

A schematic view of the TOF spectrometer part of the apparatus is shown in Fig. 2(a). The laser field is linearly polarized perpendicular to the ion beam. The dissociated ion fragments, \( \text{H}^+ \) and \( \text{H} \), obtain momentum components of the
FIG. 2. (a). Schematic view of the TOF spectrometer: the laser beam interacts with the ion beam that passes through the acceleration plates resulting in H+ and H fragments; the lower graph shows the voltage distribution on the ion acceleration plates. (b): The neutral and charged fragments resulting from the photo-dissociation of the hydrogen molecular ion after applying ion acceleration voltage with magnitude +750 V for ∆t = 60 ns for the separation of ion and neutral fragments: TOF spectra (upper graphs) and related fragment distributions accumulated for TOF detection intervals 220 ns–340 ns, 220–280 ns, and 280–340 ns (lower graphs), 1 pixel = 25 μm.

opposite signs in the plane normal to the initial ion beam propagation direction and thus spatially separate. A longitudinal electric field, typically 20–30 V/mm, was applied to the acceleration plates in the ion-beam direction to accelerate the ion fragments in order to clearly separate the charged and neutral fragments from each other. The original ion beam was initially slowed down, and after dissociation the newly created fragments were accelerated (see the voltage diagram at the bottom of Fig. 2(b)), thus differentiating the dissociation products from the ions in the original beam, since the energy of the latter was not changed in this process. The application of an accelerating voltage with a magnitude of +750 V resulted in two distinct peaks in the TOF spectra for the neutral and charged fragments from the hydrogen molecular ion photo-dissociation, which are shown in the upper graphs of Fig. 2(b). The fragment distributions accumulated in TOF intervals between 220 ns and 340 ns, 220 and 280 ns, and 280 and 340 ns are presented in the graphs of Fig. 2(b).

Figure 3 displays a series of slices through the momentum sphere of the hydrogen molecular ion fragments after photo-dissociation with the laser pulses. The numbers at the bottom of the images indicate the TOF time intervals used to generate the images. The time interval of 230–240 ns corresponds to the center of the distribution (most intense symmetric half-moon shaped curves in Fig. 3). This center slice of the TOF distribution in the inset of Fig. 3 is sharper than the momentum distribution of the full data set (197–274 ns), which allows to resolve the peaks corresponding to the different vibrational states as is indicated. The KER distribution per fragment (H or H+), calculated for the most distinct apex portion (∼10°) of the center slice 230–240 ns of Fig. 3, is presented in Fig. 4. The labels v = 6...10 correspond to the peaks due to the prominent transitions from the vibrational levels of H+2, which depend on the initial excitation distribution in the produced H+2 ions and the Franck–Condon factors, having for v = 9 the highest transition probability due to near-resonance conditions for the used laser wavelength.8

The energy spread of the ions extracted from a duoplasmatron ion source is less than 10 eV.25 In the propagation direction, this leads to a half-width of the velocity distribution ∼200 m/s with spread of the TOF times of only ∼0.4 ns, which results in the fragment KER spread on the MCP of ∼0.2 meV. The energy spread of the ions just before the fragmentation gives equivalent energy spread of ∼less than 10 meV. The observed KER of fragments also causes variation of the TOF, resulting in the energy spread on the MCP less than ∼30 meV.

FIG. 3. Series of time slices (10 ns intervals between 197 ns and 274 ns) through the hydrogen molecular ion momentum distribution. The momentum distribution of the center slice (230 ns–240 ns) is clearly sharper than the momentum distribution of the full data set (197 ns–274 ns).
In order to reconstruct experimentally the laser-induced photo-fragmentation 3D structure of the hydrogen molecular ions, we applied no voltage to the ion accelerator, thereby the distributions of the charged and neutral fragments were not time-shifted on the MCP detector. The TOF spectrum of photo-dissociation of the hydrogen molecular ions is shown in Fig. 5 and a 3D reconstruction of the fragment momentum distribution consisting of separate slices is presented in Fig. 6.

IV. CONCLUSION

Using an ion beam apparatus with a state-of-the-art detector featuring the position and time resolution, we obtained results for the fragmentation of the hydrogen molecular ion with 800 nm, 50 fs laser pulses by employing a time-sliced 3D imaging (a measurement of all the three momentum components). A series of slices through the hydrogen molecular ion momentum sphere after photo-dissociation was obtained. The analysis of these slices indicates that the center slice of the TOF distribution is clearly sharper than the momentum distribution of the full data set, which allows resolving the peaks corresponding to the different vibrational states. We have also demonstrated that by using the time-slicing technique together with an appropriate voltage applied to the ion accelerator, we could selectively study the charged (H$^+$) or neutral (H) fragments. Consequently, the developed detection system is well-suited for 3D imaging of the fragment momentum distribution and is a promising tool for imaging studies of photo-fragmentation of the atomic and molecular systems of interest.

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