Ultrafast 3D photodissociation imaging (3D-PDI): initial observations of the interaction of 40fs laser pulses with a beam of H_2^+ ions

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Introduction

Position- and time-sensitive detectors are being employed by a number of Ultrafast Spectroscopy research groups internationally. A particularly successful implementation of such technology is Cold Target Recoil Ion Momentum Spectroscopy (COLTRIMS)[1], also known as the "reaction microscope", which relies on two position sensitive detectors to record the arrival of correlated electrons and positively charged ions. The reaction under investigation (caused by a laser pulse or the impact of a highly charged ion) can be precisely quantified by measuring the distribution of momenta and angular distributions of such particles. The core technology in the COLTRIMS system is the "delay line" anode, placed behind a pair of microchannel plates (MCP), essentially a spatially selective ultra-gain particle amplifier. An ion striking the front face of the MCP pair initiates an fast (<100 ns) electron cascade that is detected on both ends of a long conductor in the anode. The position at which the ion strikes the detector is recovered through accurate measurement of the difference in arrival time of the signal at the ends of the conductor. As the positional information relies on an accurate measurement of the electronic pulse from the MCP, such a system requires dedicated timing electronics.

Mizogawa and co-workers (Tokyo Metropolitan University) have developed an alternative method for recovering positional information when used in conjunction with a MCP pair. By dissipating the charge cloud emitted from the back of the MCP over the Modified Backgammon with Weighted Capacitance (MBWC) anode[2], the centre of the cloud of electrons is related to the relative charge measured on four independent outputs. A major advantage of this system as compared to the delay line system is other than the anode and MCP pair, the only hardware required to operate this system is a fast four-channel digital storage oscilloscope (DSO) coupled to a PC. Such a system has been recently developed for use in the UCL-QUB-RAL collaborative experiment, and is the first such implementation of a position- and time-sensitive anode being used to detect energetic neutral particles. We report preliminary measurements of the ultrafast photodissociation of a 2 keV beam of H_2^+ ions by 40 fs laser pulses.

H_2^+ beamline

As discussed in detail in[3,4], we have developed a specialty in creating beams of ions for the study of ultrafast ionization and dissociation. In the present work, a beam of H_2^+ ions is crossed with the focused output of the Astra Laser Facility (40 fs, 790 nm, 10 Hz, 20 mJ), initiating photodissociation. The mechanism of photodissociation is an area of active theoretical investigation[5,6], thus this work is expected to be of particular relevance.

A schematic of the beamline and location of the new position senstive detector (PSD) is presented in Figure 1. H_2^+ ions are created in a plasma discharge ion source, momentum selected and collimated to a 0.6mm FWHM diameter beam. A spherical mirror is employed to generate the intense field necessary to produce photodissociation over a large volume. This mirror is mounted on an *x-y-z* UHV translation stage, allowing maximization of the focal overlap with the ion beam. By translating the mirror in *z* the laser intensity can be readily controlled.

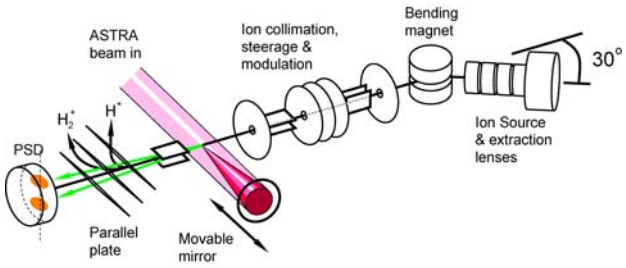

Figure 1. Schematic of the DC plasma discharge ion source, beam steering and focusing ion optics, laser focus and parallel plate analyser. The position-sensitive detector (PSD) is illustrated in Figure 2.

Experiments to date[3,4] have used traditional time of flight technique to measure the kinetic energy of the reaction. The natural progression of this experiment was the introduction of imaging technology to observe the dissociation process in more detail. In the case of a linearly polarized laser pulse photodissociation of H_2^+ is most efficient when the internuclear axis is parallel to the laser polarization. By orienting the polarization perpendicular to both the ion and laser propagation directions, the photodissociation products dissipate as indicated by the green arrows in Figure 1. By measuring the distance travelled, the kinetic energy (or momentum) is retrieved.

Following interaction with the 40 fs laser pulse, the reaction products enter an electrostatic parallel plate analyser, which deflects charged particles into two well-shielded detectors. The main H_2^+ beam is monitored on a Faraday cup, and the H^+ fragments can be detected in an off-axis channeltron. The neutral fragments are undeflected, and exit the parallel plate analyser through a large aperture, entering the PSD.

Recovering positional information

As discussed, we are employing a MBWC (or backgammon) anode after the microchannel plates to measure the arrival of neutral dissociation products. The position at which the H fragment strikes the PSD is encoded by division of charge. Figure 2 illustrates the main elements of the PSD. Despite the parallel plate removing the main beam and charged fragments, energetic ions and electrons can produce noise at the rate of MHz, so electron and ion repeller grids clean the beam, allowing only neutral particles through. The kinetic energy of the neutral particles is sufficient to cause electron cascade in the MCP pair, each event resulting in 10^7 electrons exiting the back of the second MCP. This electron cloud then propagates over a few centimetres, striking the MBWC anode.

As name suggests, the backgammon anode consists of a series of interleaved triangular conductors printed onto an insulating

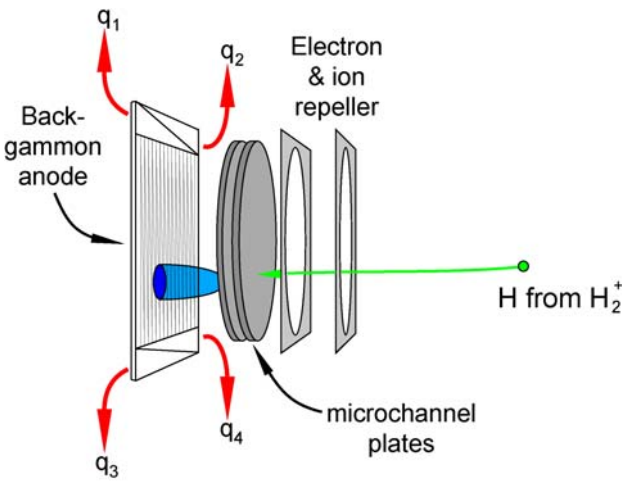

Figure 2. Illustration of the main elements of the PSD. Only neutral photodissociation fragments pass through the two repeller plates (green arrow), causing electron cascade in the MCP pair (blue cone), which dissipates over the conducting elements of the MBWC anode. By measuring the ratio of charges (red), the kinetic energy of photodissociation can be measured.

ceramic plate. The triangular conductors can be visualized as a series of 2mm wide, 50mm long rectangular strips spaced 0.25mm apart with a 0.25mm diagonal insulator across each strip. The separation between the MCPs and anode causes the electron cloud emitted by the MCP to disperse over a number of strips and the position parallel to the 50mm edge is proportional to the ratio of charges measured at the ends of the triangular conductors. So far, the anode has encoded the position of the article impact in one dimension. By dividing the charge again between two pairs of triangular conductors, the position perpendicular to this axis is encoded. Thus when a particle strikes the MCP pair, the position in x and y on the PSD is recovered by measuring the charges q_1 to q_4, and performing the following straightforward relationships:

$$x = \frac{q_1 + q_2}{q_1 + q_2 + q_3 + q_4} \text{ and } y = \frac{q_1 + q_3}{q_1 + q_2 + q_3 + q_4} \qquad (1)$$

As the H_2^+ ions propagate along the beamline, neutralization occurs, thus there is considerable noise from energetic neutral particles created by the passage of the ions. However, the photodissociation events we are interested in can readily be detected despite this noise, as the energetic neutral H fragments from the laser interaction arrive at a very well defined flight time, which depends on the ion beam kinetic energy in the direction of propagation. By setting the laser polarization direction perpendicular to the ion propagation direction, the Doppler broadening caused by the narrow range of beam kinetic energies (typically 50 eV in 2 keV) is removed.

Three-dimensional photodissociation imaging (3D-PDI)

We now present preliminary observations of the photodissociation of H_2^+ by a 40fs laser pulse at an intensity of 10^{13} Wcm^{-2}, recorded using our new PSD system. As the position of arrival of the photofragment is measured in two dimensions, and the time of flight due to the ion beam kinetic energy is used to differentiate the photodissociation signal from the large background, we refer to this new technique as three-dimensional photodissociation imaging (3D-PDI).

In Figure 3, we present the first true output from the 3D-PDI system. While the results show some scatter, such images are strongly encouraging, indicating that the PSD and associated software are operating correctly. Very importantly, if the laser polarization direction is altered (i.e. set parallel to the ion beam

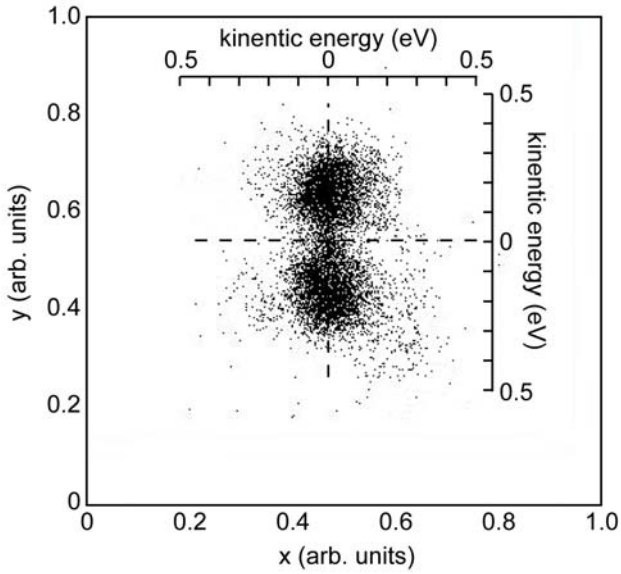

Figure 3. Photodissociation of H_2^+ by 40 fs 790 nm Astra laser pulses, at an intensity of 10^{14} Wcm^{-2}. This 3D-PDI output was recorded with 10^4 laser shots.

propagation direction) the vertically split distribution of laser-correlated neutral H fragments merge into a broad central peak, and the distribution of arrival times is almost identical to our early measurements.

By using a 5 mm wide horizontal mask, we were able to calibrate x and y recovered from Equation (1) to position on the anode. Through knowledge of the beamline and PSD dimensions, we were then able to calibrate the detected kinetic energy of the observed neutral H photofragments, as indicated in Figure 3. The centre of the photodissociation image (at 0 eV) does not correspond to the centre of the detector (x = y = 0.5 units). This is due to a slight mechanical misalignment of the detector to the centre of the ion beam, estimated to be < 1 mm.

Conclusion

We have presented the first results from the recently commissioned position sensitive detector that will be used as part of the successful UCL-QUB-RAL investigation into ultrafast photodissociation reaction dynamics in H_2^+ and D_2^+. By measuring the position and time the neutral H photofragments arrive at the detector, three-dimensional photodissociation imaging is now possible. We intend to improve the resolution of the instrument by optimizing both the detector and associated software.

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