

## Coulomb explosion imaging of a dissociating nuclear wavepacket

C. R. Calvert, J. McKenna and I. D. Williams

*Department of Physics, Queen's University Belfast, University Road, Belfast, BT7 1NN, UK*

J. Wood, E. M. L. English and W. R. Newell

*Department of Physics and Astronomy, University College London, Gower Street, London, WC1E 6BT, UK*

W. A. Bryan, I. C. E. Turcu, J. M. Smith, E. J. Divall, K. Ertel, O. Chekhlov, C. J. Hooker, A. J. Langley and J. L. Collier

*Central Laser Facility, CCLRC Rutherford Appleton Laboratory, Chilton, Didcot, Oxon., OX11 0QX, UK*

**Main contact email address** c.calvert@qub.ac.uk

### Introduction

Modern advances in ultrashort laser technology have provided many exciting new possibilities for temporally imaging previously inaccessible ultrafast processes.

Consequently the temporal mapping of bound and dissociative nuclear wave packet motion is currently within reach, and realisation of this goal will help provide fundamental elucidation of the dynamic nature of molecular motion itself, critical to the control of chemical and biological reactions. Moreover, such experimental advancements also provide an unprecedented platform for testing fundamental concepts in quantum mechanical molecular models.

The traditional approach to imaging temporal evolution with laser light is to optically pump some transition and subsequently probe the system by either observing or inducing a radiative transition using precise spectroscopic techniques. Such 'pump-probe' experiments can also be implemented with ultrashort intense field pulses and are in fact much more adaptable as they do not require precise resonant photon transitions. In the present work, the molecules are pumped in the strong field regime where the coupling between populated and unpopulated states is via an intense non-perturbative interaction.

Furthermore, ultrashort pulse durations enable significantly improved temporal resolution permitting observations to be made on extremely short timescales; which is not possible when using the narrow linewidth necessary for spectroscopic measurements. As such, the recent application of ultrashort intense laser pulses in pump probe experiments has enabled the initiation and high resolution observation of temporally evolving dissociative wavepacket motion<sup>[1,2]</sup>. Few-cycle laser pulses enable these detailed experiments to be performed in even the fastest oscillating molecules, such as the hydrogenic systems  $H_2^+$ ,  $D_2^+$  and  $HD^+$ . These fundamental three body quantum systems are ideal candidates for theoretical simulations as their dissociation and ionisation mechanisms in intense laser fields are well documented and understood, as summarised in a recent review article<sup>[3]</sup>. Of these three, the deuterium molecular ion,  $D_2^+$ , has the largest reduced mass and thus has reduced wave packet momentum (vibrational period  $\sim 25$  fs), making it the preferred experimental candidate.

The experiment detailed in this report has utilised Coulomb explosion (CE) imaging<sup>[4]</sup> to probe the time-dependent nature of the  $D_2^+$  nuclear wave packet. Observations have been made of temporally evolving dissociative channels and clear evidence of a dephasing bound wavepacket has also been obtained.

Complementary results from similar experiments have been recently reported<sup>[1,2,5,6,7]</sup> showing good agreement with the present study.

### Experiment

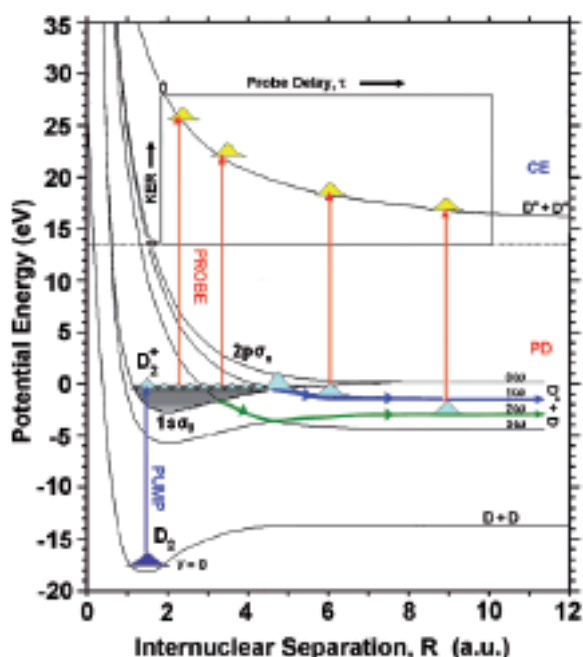
The experiment was carried out on the ASTRA laser, Target Area 1, at the Central Laser Facility, RAL (UK). The Ti: Sapphire oscillator in this system ( $\lambda = 800$  nm, bandwidth 40 nm, kHz repetition rate) was used to generate 30 fs, linearly polarised, laser pulses which were subsequently compressed to 10 fs ( $\sim 100$  nm bandwidth) using a hollow fibre-chirped mirror compressor system. A full discussion on this compression technique has been documented by I C E Turcu *et al.*<sup>[8]</sup> and can be found in CLF Annual Report 2004-2005.

The resulting few-cycle pulses were then prepared for pump probe studies by use of a Mach-Zehnder interferometer. This arrangement enables the beam to be split into two separate components of comparable energy and subsequently recombined collinearly with an adjustable temporal delay ( $\tau$ ) between the pump and probe pulses. This was done using a series of mirrors, beam splitting pellicles and a translation stage permitting the path length of the probe pulse to be altered on a sub-femtosecond ( $\sim 300$  as) timescale. Spatial overlap of the pulses was imaged on a CCD camera and temporal overlap was maximised by cross correlation of the pulses. Pulse profile diagnostics were also obtained using interference effects around the temporal overlap region, for a full discussion see W A Bryan *et al.*<sup>[9]</sup>.

After transmission through the interferometer optics, the resulting pulses ( $\sim 15$  fs) were interacted with a molecular target and the  $\tau$  value at which the probe pulse images the temporally evolving system was varied with significantly high resolution. The laser pulses entered a time of flight mass spectrometer (TOFMS) interaction chamber<sup>[10]</sup> and were focused by an  $f/5$  spherical mirror onto a spectrally pure  $D_2$  gas target. Tunnel ionisation<sup>[11]</sup>, photodissociation, PD, (via bond-softening)<sup>[12]</sup> and Coulomb explosion<sup>[13,14]</sup> products were then extracted through a 250  $\mu\text{m}$  aperture by a DC electric field (2000  $\text{V m}^{-1}$ ). After extraction and acceleration, these products passed through a field free drift tube and were collected at a microchannel plate detector, with flight time recorded via a fast digital storage oscilloscope. The position of the extraction aperture ensured that detected signal arose solely from interactions at the volume of peak intensity of the laser and the finite width of the aperture restricts the angular acceptance of detection to fragments projected along the detection axis. The TOFMS was run in Wiley-McClaren<sup>[15]</sup> mode in order to give good separation of the 'forward' and 'backward'

TOF peaks. As the electrostatic conditions in the source region of the TOFMS are well known, the fragment kinetic energy could be readily calculated from the ion time of flight.

A schematic of the pump probe experiment is presented in figure 1, wherein the two separable pulse interactions with the molecular target are presented as ionisation and dissociation processes involving the pertinent potential energy surfaces. The pump pulse ionises the neutral  $D_2$  target, projecting its ground state vibrational wavefunction onto the bound  $1s\sigma_g D_2^+$  potential surface, launching a non-stationary coherent wavepacket. At sufficient intensities, the pump pulse further couples the population from the bound  $1s\sigma_g$  state onto the dissociative  $2p\sigma_u$  state of the  $D_2^+$  ion, thus initiating dissociation ( $D + D^+$ ) of the wavepacket via the  $1\omega$  and  $2\omega$  channels, displayed in figure 2 in terms of Floquet dressed potentials<sup>[16,17,18]</sup>.



**Figure 1.** Schematic of the pump probe experiment. The pump pulse ionises the  $D_2$  target creating a dynamic  $D_2^+$  wavepacket which is later projected onto the CE curve ( $\sim 1/R$ ) by the probe pulse, giving the system a  $\dot{U}$  dependent kinetic energy release (KER).

Using linearly polarised light, any dissociating PD fragments ( $D$  or  $D^+$ ) are projected along (or close to) the polarisation direction of the pulse. This effect is intrinsically dependent upon the coupling of the  $1s\sigma_g$  and  $2p\sigma_u$  states which is sensitive to the angle between the pulse polarisation and the axis of the molecular bond<sup>[19]</sup>. The pump and probe pulse polarizations were aligned parallel to the detection axis of the TOFMS, ensuring that the initiated dissociating fragments were projected towards the detector.

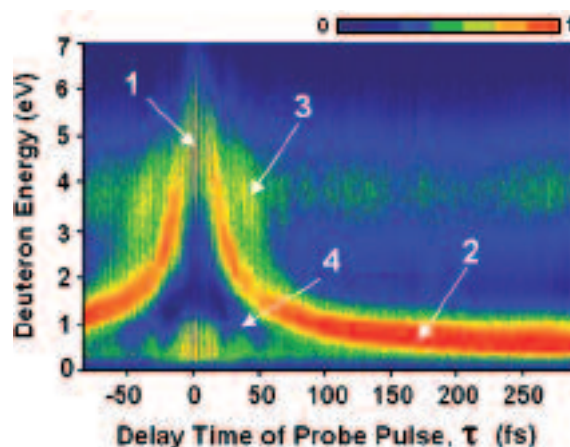
Subsequent to the pump pulse, the system is left to evolve in a field-free state such that the wavepacket may continue to perform dissociative motion. After a time delay,  $\tau$ , the probe pulse images the wavepacket by projecting it onto the repulsive Coulomb potential (i.e. by removing the final

electron from the system). The final appearance energy of the fragment therefore reflects the internuclear separation,  $R$ , at which this probing event occurs.

## Results and Discussion

Figure 2 displays the result of measuring the deuteron kinetic energy of fragmentation as a function of probe delay time. The experiment was carried out with two near-identical 15 fs ( $0.5 \text{ PW cm}^{-2}$ ) pulses. For positive delay time the variable path of the interferometer acted as the pump pulse with the fixed path providing the probe; the converse is true for negative delay time. The highly symmetric structure (about  $\tau = 0$ ) in figure 2 thus serves to confirm that the two pulses were near identical.

At  $\tau < 20$  fs (arrow 1) the pulses are spatially overlapped and the relative phase difference between their electric fields gives rise to constructive and destructive interference effects. Thus for  $\tau$  values at multiples of half an optical cycle ( $4/3$  fs) striking destructive interference stripes are observed wherein little or no fragment yield is present. Concordantly at steps of one full optical cycle ( $8/3$  fs) around  $\tau = 0$  there is effectively one strong pulse (constructive interference) with an intensity of up to  $2 \text{ PW cm}^{-2}$ . Here the  $D_2^+$  wavepacket, at small  $R$ , is projected directly onto the Coulomb potential curve and the deuterons appear with fragment energy as high as 7 eV (corresponding to the  $\tau = 0$  point in figure 1). This overlapped pulse around  $\tau = 0$  also gives rise to direct observation of  $1\omega$  and  $2\omega$  PD fragments with 0 - 1 eV energy in figure 2.



**Figure 2.** Deuteron fragmentation energy as a function of pump probe delay time,  $\tau$ . Highlighted features detail interferometric effects (1), CE imaging of dissociating wavepacket (2) and CE/PD imaging of remaining bound wavepacket(3)/(4). The colour scale signifies the fragment yield, given in arbitrary units.

The red band of fragmentation signal which shifts from high energy to low as  $\tau$  increases (indicated by arrow 2) signifies the CE imaging of PD channels. As the dissociating system evolves in time it follows the highlighted paths shown on figure 1 (blue arrow for  $1\omega$  dissociation, green arrow for  $2\omega$  dissociation), with the internuclear separation increasing and thus the final appearance energy from the CE process decreasing ( $E_{\text{frag}} = E_{1\omega,2\omega} + 1/R$ ). Asymptotically the imaging energies of these channels tend to the PD fragmentation energies

(cf:  $1\omega$  and  $2\omega$  at  $\tau = 0$ ) whereby at large  $R$  the removal of the final electron causes no significant effect (Coulomb repulsion is negligible). As discussed, a significant amount of the  $D_2^+$  wavepacket created by the pump pulse is dissociative; however a portion remains in the bound  $1s\sigma_g$   $D_2^+$  potential. At around 34 - 38 fs, having executed  $\sim 1.5$  vibrational periods, this coherent bound wavepacket is in phase near the 1 and 2 photon crossings and consequently enhanced signal is observed in the CE and PD channels as indicated by arrows 3 and 4 respectively.

The PD imaging of the bound wavepacket is not clearly resolved beyond  $\tau = 100$  fs due to the dominant structure of the CE imaged dissociative wavepacket. Moreover, the enhanced structure at the outer turning point of the  $1s\sigma_g$  well is not observed at later times in the CE channel due to dephasing of the bound  $D_2^+$  vibrational wavepacket. This effect is a fundamental property of the quantum nature of the bound wavepacket, and corresponding simulations<sup>[20]</sup> have shown fascinating effects wherein the wavepacket dephases and then revives at some later time.

### Conclusion

This work has provided direct experimental imaging of the path of a molecule as it dissociates and therein has established a well resolved observation of the dissociative potential energy surface of  $D_2^+$ . Dephasing properties of the bound  $D_2^+$  wavepacket have also been directly observed and the results presented in this article show good consistency with other recent studies<sup>[1,2,4,21]</sup>. Such pioneering achievements have only been possible due to the remarkable advancements in ultrashort laser technology, with few-cycle laser pulses now enabling detailed dynamical studies of the most elementary molecular systems, therein providing a useful interface between experiment and fundamental quantum theory.

In addition to the work in this report, further experiments have been carried out in order to obtain detailed information on the dynamical nature of bound wavepacket motion. For in-depth experimental and theoretical details on imaging the bound  $D_2^+$  vibrational wavepacket see J McKenna *et al.*<sup>[22]</sup> and J Wood *et al.*<sup>[23]</sup> and the article by E M L English *et al.*<sup>[24]</sup> investigating rotational wave packet revivals.

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