

Defining the coherence and controlling the motion of a nuclear wavepacket

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

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

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

Department of Pure and Applied Physics, Queen's University Belfast, Belfast, BT7 1NN, UK

W. A. Bryan*, I. C. E. Turcu, K. G. Ertel, J. M. Smith, E. J. Divall and O. Chekhlov

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

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

Main contact email address w.a.bryan@rl.ac.uk

Introduction

Sub-vibrational-period imaging of the nuclear motion of the D_2^+ ion is achieved using a few-cycle laser pulse pump-probe configuration and time-of-flight mass spectroscopy. Through independent variation of the pump and probe pulse duration, the extent of the temporal and spatial coherence of a nuclear wavepacket is investigated.

The recent surge in few cycle laser pulse technology has lead to a renewed interest in using pump-probe techniques^[1-3] as a means of tracking nuclear motion. Now pulse durations are on a comparable and in some cases even shorter time scale than the nuclear motion of even the smallest molecules, the imaging of their dynamics has become feasible. Recent experiments on many small molecules including D_2^+ , which we report on here, are potentially the first steps to realizing full quantum control of molecular systems in an ultra-short intense laser field. Interest in using vibrational wavepacket control for quantum computation^[4-5] and reaction dynamics are also flourishing, underlining the relevance and impact of such studies as ours upon the wider scientific community.

Experimental Configuration

The experiments were conducted in TA1 of the Astra laser facility at the CLF. The laser pulses are produced from a Ti: Sapphire oscillator and amplified by CPA techniques after being stretched. The pulses (1mJ, 7ps, 5mm dia, 1kHz) are then compressed in a prism compressor before being spectrally broadened by self-phase modulation in a 1m long hollow optical fibre[6] containing 0.3bar of pure Argon gas. This allows further compression by reflection off a series of ten chirped mirrors. The resulting pulses (0.25mJ, 10fs, 7mm dia, 1kHz, 0.4W) are then split in a Mach-Zender interferometer to give pump and probe pulses (see figure 1). The delay between which is controlled by altering the length of one of the interferometer arms by a computer controlled high precision translation stage offering a delay time resolution of 1/3fs.

A $\lambda/2$ plate is placed in the fixed arm of the interferometer to make the polarization of the pump pulse perpendicular to that of the probe pulse and the axis of the time-of-flight spectrometer which are parallel to one another. The beamsplitters (BS) employed are four micron thick pellicles to avoid stretching the pulses unnecessarily; although on leaving the interferometer the pulses from each arm where measured to be 13fs in duration. Additional control over the pulse length is obtained by the insertion of microscope coverslips and glass slides of different thickness into the

beam path to induce positive dispersion which stretches the temporal profile as required (see figure 2). The turning mirror immediately after the interferometer is mounted in a “flip-mount” to allow the pulse durations to be monitored immediately before and after each period of data collection.

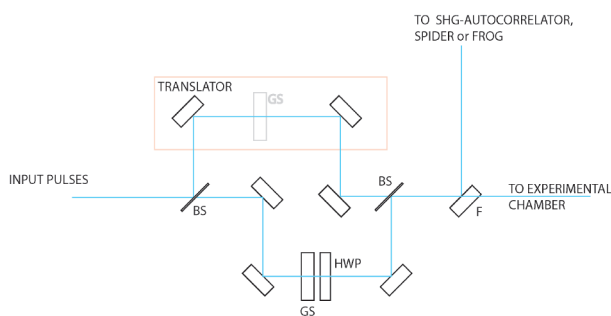


Figure 1. A schematic of the Mach-Zehnder type interferometer employed in the current work. The fixed arm containing a half waveplate (HWP) is at the bottom of the figure and the variable arm at the top. Glass slides (GS) are placed into either arm of the interferometer to stretch the pulses independently of one another. After the interferometer the pulses can be sent for diagnostic tests or directly to the experimental chamber via use of a “flip mirror”.

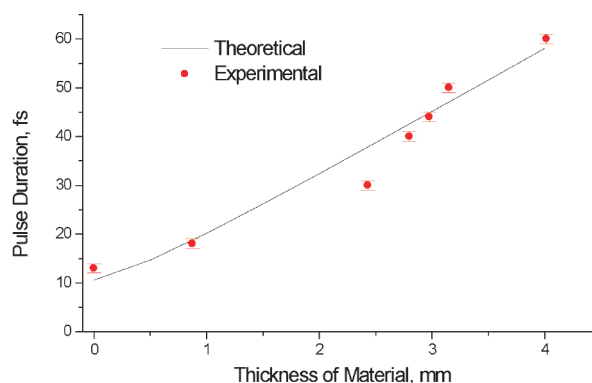


Figure 2. A plot of pulse duration against the thickness of the glass slides inserted into one of the arms of the interferometer in order to stretch the pulses. The theoretical curve^[7] was generated assuming the slides were made of BK7. The experimental values shown are obtained from measurements taken with a SHG-autocorrelator. For clarity only the points shown for the stretching of the probe pulse are shown.

The D_2^+ ions are created in a superposition of vibrational states via laser induced ionization from neutral deuterium gas by the pump pulse. At precisely controlled times the probe pulse then causes these molecular ions to fragment by either Coulomb explosion ($D^+ + D^+$) or photo-dissociation ($D^+ + D$). This happens inside the interaction region of an ultra-high vacuum time-of-flight mass spectrometer operated in a Wiley McLaren mode^[8] with the pressure of the gas kept low enough to avoid space-charge effects. The laser pulses are $f/5$ focused on reflection from a spherical mirror mounted inside the UHV tank to give peak intensities of between 0.5 and 0.8 PW cm^{-2} in the individual pulses.

The fragment ions are extracted through a $1/4 \text{ mm}$ aperture allowing signal from only the most intense region of the laser focus to be imaged. It is from analysis of the energies of these fragments that we catch a glimpse of the nuclear wavepacket dynamics.

Results and Discussion

The D_2^+ vibrational wavepacket is created at the instant of ionization and comprises of a superposition of vibrational states (v -states). On creation, the wavepacket exhibits a high degree of coherence (all v states share a common R value, where R is the internuclear separation). As the wavepacket evolves this coherence de-phases due to the anharmonicity of the $D_2^+ 1s\sigma_g$ potential^[9]. At set time intervals, multiples of what is known as the revival time, this initial degree of coherence is regained^[10]. This behavior is observed experimentally as an oscillation in the photo-dissociation signal with a period equal to the weighted average of the levels encompassed by the original superposition at a delay time of around 550 fs .

In the first series of experiments the duration of the pump pulse was stretched. Figure 3 shows the integrated photo-

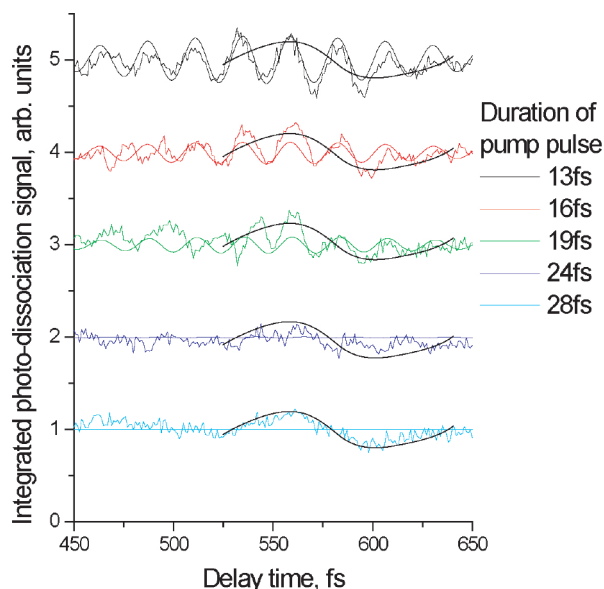


Figure 3. A plot of the integrated photo-dissociation signal against pump-probe time delay for the experiments in which the duration of the pump pulse was varied. As the pump pulse is stretched the signature of the vibrational revival, in the D_2^+ population, recedes to reveal the rotational revival (black curve) feature beneath. For clarity the integrations for each pump pulse length are displaced along the y-axis. Also shown are the theoretical curves generated for each pump pulse width.

dissociation signal as a function of wavepacket evolution time. Also plotted are the theoretical curves obtained from a quantum-mechanical simulation of the vibrational wavepacket motion^[11] (figure 4) allowing for a variation in the pumps temporal width. A curve showing the characteristic shape of the full rotation revival is also shown on top of the data points. This is thought to arise from a rotational wavepacket, comprising of a superposition of different rotational states (J -states), that is created through impulsive alignment^[12] by the laser field of the pump pulse. For a more detailed discussion of the rotational dynamics see, English *et al.*^[13].

As the pump pulse is stretched the initial coherence in the vibrational wavepacket is diminished until at a pulse duration equivalent to the vibrational period of the $v=0$ level of D_2^+ it disappears completely. This is observed as a washing out of the fringe pattern in the photo-dissociation signal with increasing pulse duration. This is clearly seen in figure 4 where the structure of the wavepacket disappears for pump pulse duration greater than the $v=0$ time period. As the vibrational revival pattern fades out the rotational revival pattern becomes apparent (figure 3). The theoretical treatment employed only considers the vibrational dynamics of the D_2^+ ensemble; subsequently the rotational behavior of is unaccounted for either in the theory curves on figure 3 or the probability density plots of figure 4.

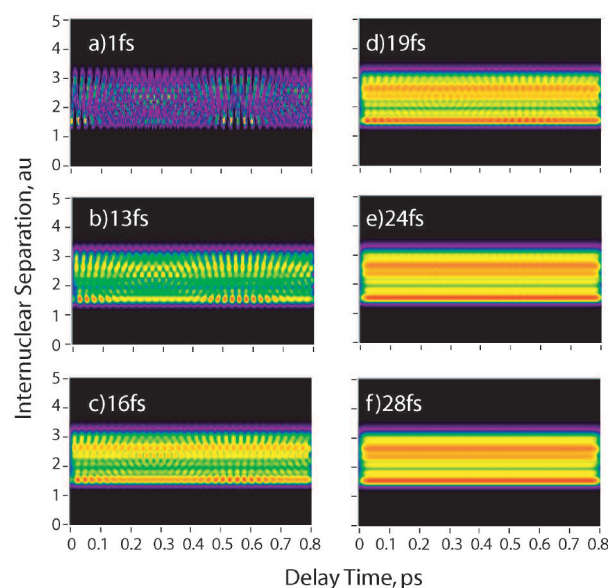


Figure 4. (a)-(f) Probability density plots showing the spread of the vibrational wavepacket across the internuclear coordinate R for the D_2^+ potential well as a function of time delay from production. Each panel is labeled with the FWHM duration of the pump pulse responsible for its initiation. In each panel the probability density is highest in regions coloured red and lowest in those coloured black.

A more subtle effect, however, is that the period of the revival pattern also changes, with the period of the oscillation decreasing as the pump width is increased. This occurs because the higher vibrational levels get washed out at the longer pump durations, but the lower lying levels can still revive due to their period being less than the pump duration. Essentially the pump pulse acts as a temporal filter.

In the second series of experiments the duration of the pump pulse was kept constant at 13fs and the probe duration was varied. In our pump-probe technique the pump pulse can be considered to initiate the dynamics and the probe pulse images them. Therefore within a photographic analogy the stretching of the probe pulse can be thought of as increasing our “exposure time” to the nuclear motion. Therefore if the motion being imaged is quicker than the exposure time a “blurring” or averaging out of the image occurs. From figure 5 this seems to occur abruptly at a pulse duration of around twice the period of the $v=0$ vibrational level. This is twice the duration needed to kill off the revival pattern when stretching the pump pulse.

Again in contrast to when the pump pulse was stretched, the period of the oscillation in the photo-dissociation signal remained constant as the laser pulse duration was increased. This is because as the pump pulse was the same for all the measurements; so was the vibrational wavepacket excited. Conversely when the pump pulse was varied different wavepackets were initiated in each measurement, each one forcing its own characteristic period of oscillation onto the photo-dissociation signal observed.

Figure 5 also shows no signs of the rotational revival seen previously. This is the case for two reasons. When the probe pulse durations are short enough to clearly image the vibrational revival the weaker rotational revival is suppressed by the vibrational structure. With probe pulses of greater than 40fs the vibrational structure is suppressed. However, such durations are similar to those of the rotational revival features (see the black curves on figure 3 and ref^[13]). Hence they are not observed experimentally.

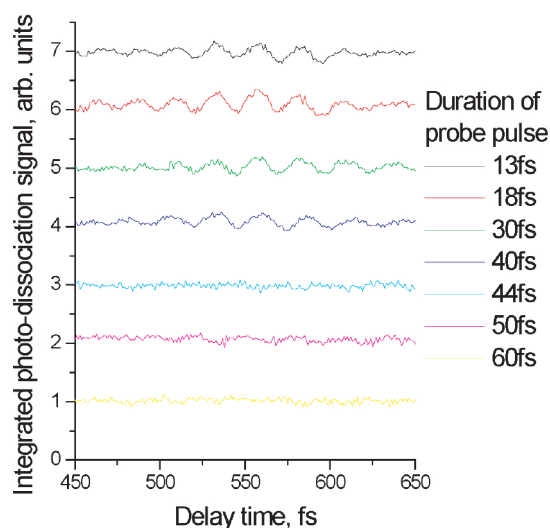


Figure 5. A plot of the normalized integrated photo-dissociation signal against pump-probe delay time for the experiments in which the duration of the probe pulse was varied. As in figure 3 the normalized integrations have been separated out along the y-axis for aesthetic reasons only.

Conclusions

Using few cycle laser pulses the sub-vibrational-period imaging of the nuclear motion of the D_2^+ ion has been achieved. By variation of both the pump and probe pulse durations, the extent of the temporal and spatial coherence of a nuclear wavepacket throughout its evolution has been traced. Additionally rotational revival structures, arising from an impulsive alignment process towards the pump pulse have been isolated in the data. In both instances these experimental observations were found to be in excellent agreement with our own theoretical models^[11] and those of others^[10].

References

1. F. Légaré *et al.*, *Phys. Rev. A* **72** 052717 (2005)
2. A. S. Alnaser *et al.*, *Phys. Rev. A* **72** 030702(R) (2005)
3. Th. Ergler *et al.*, *Phys. Rev. Lett.* **95** 093001 (2005)
4. G. Gollub *et al.*, *New J. Phys.* **8** 48 (2006)
5. U. Troppmann *et al.*, *New J. Phys.* **8** 100 (2006)
6. M. Nisoli *et al.*, *Appl. Phys. Lett.* **68** 2793, (1996)
7. P. Dietrich, “*Refractn*” (2005)
8. W. C. Wiley and I. H. McLauren, *Rev. Sci. Instrum.* **26** 12 1150 (1955)
9. T. E. Sharp, *Atomic Data* **2** 119 (1971)
10. B. Feuerstein and U. Thumm, *Phys. Rev. A* **67** 063408 (2003)
11. J. McKenna *et al.*, *CCLRC CLF Annual Report* (2006)
12. T. Seideman, *J. Chem. Phys.* **115** 5965 (2001)
13. E. M. L. English *et al.*, *CCLRC CLF Annual Report* (2006)