Dynamic imaging of a dissociative $\text{D}_2^+$ nuclear wavepacket in intense laser fields

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Abstract.

Ultrashort intense laser pulses have been used to initiate and image a dissociating $\text{D}_2^+$ nuclear wavepacket. Using a pump probe technique, the temporal evolution of the dissociation process has been observed as well as interferometric effects and signatures of bound wavepacket motion.

1. Introduction

Coherent light sources have long been a primary tool for manipulating and studying complex quantum dynamics. Finely tuned spectroscopic techniques have proven crucial in elucidating fundamental molecular structure and the ongoing advancement of laser technology continues to provide new possibilities in the observation and control of molecular motion. The recent development of few-cycle laser pulses has enabled temporally evolving processes to be initiated and observed on ultrashort femtosecond timescales and thus substantial research in intense field molecular physics has recently been geared towards real-time mapping and control of quantal nuclear wavepackets [1, 2, 3]. The application of this technology to fundamental molecular systems is a vital first step towards manipulation and control of larger, more complex molecules. An ideal primary candidate for such studies is the deuterium molecular ion, $\text{D}_2^+$, which offers the theoretical accessibility of a simple hydrogenic system and has slower wavepacket motion than its isotopic counterparts $\text{H}_2^+$ and $\text{HD}^+$. The experiment detailed in this article incorporates a pump probe technique to initiate a dissociating $\text{D}_2^+$ nuclear wavepacket [4] with subsequent imaging via the Coulomb explosion (CE) channel [5]. Observations have been made of temporally evolving dissociative channels together with interferometric effects and signatures of bound wavepacket motion. The present study complements other recent investigations which have utilised ultrashort laser pulses on hydrogenic systems [2, 6, 7, 8, 9].

2. Experimental method

The experiment was carried out on the ASTRA laser at the Central Laser Facility, Rutherford Appleton Laboratory (UK). The 30 fs output pulses from this Ti: Sapphire laser (central $\lambda = 800$ nm, $\Delta \lambda = 40$ nm, 1 kHz repetition rate) were spectrally broadened ($\Delta \lambda \sim 150$ nm) by self phase modulation through propagation in a hollow core fibre, containing differentially pumped Argon
Figure 1. (Colour online) Schematic diagram of the principle mechanisms involved in the pump probe experiment. The pump pulse initiates a dynamic $D_2^+$ wavepacket, which may dissociate by $1\omega$ or $2\omega$ PD channels and the probe pulse images the wavepacket by projection onto Coulomb potential. The resulting $D^+$ appearance energy is experimentally obtained from the deuteron flight time.

The linearly polarised pump and probe pulses (beam diameter = 10 mm) were aligned into a time of flight mass spectrometer (TOFMS) interaction chamber and focussed onto a spectrally pure $D_2$ gas target using a spherical mirror ($f = 50$ mm). The resulting photodissociation (PD) via ‘bond softening’ [11] and CE products from the high intensity interaction were extracted by a uniform electric field through a 250 $\mu$m aperture, which serves to limit the ion sampling to the highest intensity point of the laser focus and restricts the angular acceptance of the system to fragments projected parallel to the detection axis. After extraction the fragments passed through an acceleration region and field free drift tube prior to collection at a microchannel plate detector, with time of flight (TOF) recorded by a fast digital storage oscilloscope. The detected spectra from 2000 laser shots were averaged to give a single TOF spectrum for each time delay, $\tau$. The operation of the TOFMS in Wiley McClaren mode [10] guaranteed good separation of the ‘forward’ and ‘backward’ dissociation peaks, and thus fragment energies could be readily calculated from the ion time of flight. The polarisation direction of the laser pulses was orientated along the detector axis to ensure efficient collection of the dissociation products.

A schematic of the principle mechanisms in the pump probe experiment is shown in figure 1. The pump pulse ionizes the $D_2$ target molecule and initiates dissociation of the resulting $D_2^+$ ion by net absorption of one ($1\omega$) or two ($2\omega$) photons. In the strong field regime the PD processes may be considered in the Floquet light induced potentials (dressed states) model [12]. These potential surfaces, arising from the coupling of $1s\sigma_g$ and $2p\sigma_u$ states of the $D_2^+$ ion, become distorted as the laser intensity increases, opening up ‘avoided crossings’ wherein the dissociation process is no longer restricted to a resonant $1\omega$ or $2\omega$ process. The dissociating fragments may therefore have a range of energies (0.1–1.2 eV), depending on how many photons are absorbed and which vibrational states are accessed. After some time delay, $\tau$, the probe pulse removes...
the final electron from the system and the deuteron fragments mutually repel each other. In figure 1 this is expressed as a projection of the wavepacket onto the Coulomb potential (energy $\sim 1/R$). The final appearance energy of fragments at a given $\tau$ value, thus relates information about the internuclear separation, R, at which the CE event occurs.

3. Results and discussion

The surface plot in figure 2 displays the deuteron kinetic energy as a function of $\tau$, for an experiment carried out over a 300 fs $\tau$-range using 15 fs (0.5 PW cm$^{-2}$) pulses. At small time delays the pump and probe pulses are temporally overlapped and the relative phase difference between their electric field cycles gives rise to constructive and destructive interference effects. At delay step intervals of one optical cycle (2.67 fs) near $\tau = 0$, the two pulses behave effectively as one strong pulse with peak intensity up to 2 PW cm$^{-2}$ giving a large fragment yield. Similarly at half optical cycle steps there is destructive interference with little or no fragment yield observed. Thus ‘interference stripes’ are observed as clearly evident in the inset in figure 2. In the case of constructive interference, the $D_2^+$ wavepacket is created and projected directly onto the Coulomb potential curve at small R, with deuterons appearing with a maximum fragment energy upwards of 6 eV. This overlapped pulse around $\tau = 0$ also gives rise to direct observation of $1\omega$ and $2\omega$ PD products with energies 0–1 eV. The separate $1\omega$ and $2\omega$ channels are not resolved from each other as a range of vibrational states around the avoided crossings are accessed, contributing to overlapping energy distributions from these two channels.

The red band of fragmentation signal which shifts from high energy to low energy as $\tau$ increases, is the CE imaging of the dissociative wavepacket. At small $\tau$ values, the wavepacket is at small internuclear separation when lifted onto the Coulomb curve and thus the CE imaging imposes a high fragmentation energy. For larger values of $\tau$, the wavepacket reaches large internuclear separations prior to being imaged and thus the removal of the final electron releases less kinetic energy as the Coulomb repulsion has become less significant. Hence, as $\tau$ increases, the CE energy tends towards zero and the asymptotic limit of the shifting red band therefore tends to the PD energies seen around $\tau = 0$. The dashed line in figure 2 is a predicted trajectory
for a $1\omega$ dissociation process with final fragment energy set at 0.34 eV, corresponding to the mean $1\omega$ appearance energy experimentally observed for a single pulse process. In this simple simulation the path of a fragment along the dissociative channel has been classically modelled and then projected onto Coulomb curve at each $\tau$ value, resulting in excellent agreement with the experimental data.

The main features in figure 2 are representative of dissociative effects but there are also signatures of bound wavepacket motion. In this case the pump pulse has ionized the D$_2$ target molecule to create a coherent bound wavepacket in the D$_2^+$ 1$s\sigma_g$ potential. At around 34–38 fs the bound wavepacket has executed 1.5 vibrational periods and is in phase near the $1\omega$ and $2\omega$ crossings. Thus at this $\tau$ value an enhanced signal is observed in the PD (0–1 eV) channel and is most clearly seen in the inset of figure 2. 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. A similar enhancement effect is seen in the CE channel (3–5 eV) but the structure disappears at later times as the wavepacket dephases [7]. This dephasing effect is a fundamental property of the quantum nature of the bound wavepacket and recent simulations [3] and experiments [1, 2, 13] have shown fascinating effects wherein this dephasing wavepacket revives at some later time.

4. Conclusion

This work has provided direct time-dependent experimental imaging of a dynamic fundamental molecule on an ultrashort timescale. The dissociative D$_2^+$ wavepacket has been imaged via Coulomb explosion, reflecting information about the D$_2^+$ dissociative potential surface and the temporal evolution of the dissociative wavepacket. Dephasing properties of the bound D$_2^+$ wavepacket have also been observed, showing good consistency with other recent studies. Such pioneering achievements have only been made possible by the remarkable advancements in ultrashort laser pulse technology, with few-cycle pulses now enabling detailed dynamical studies of the most elementary molecular systems, therein providing an important interface between experiment and fundamental quantum theory.

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