Measuring the dispersion of a recolliding electron wave packet

E M L English, J Wood, W A Bryan, S L Stebbings, W R Newell
Department of Physics and Astronomy, University College London, Gower Street, London, WC1E 6BT, UK

J McKenna, M Suresh, B Srigengan, I D Williams
Department of Physics, Queen's University Belfast, Belfast, BT7 1NN, UK

I C E Turcu, J M Smith, K G Ertel, E J Divall, C J Hooker, A J Langley
Central Laser Facility, CCLRC Rutherford Appleton Laboratory, Chilton, Didcot, Oxon., OX11 0QX, UK

Main contact email address: *w.bryan@ucl.ac.uk*

Abstract

Presented here is a study of recollision in Kr^{2+} ions, from which a measurement of the size of the returning electron wave packet has been made. The ultrafast (40fs) laser field has been varied from linear polarization to circular, via a series of elliptical polarizations, while maintaining a constant distribution of sequential ionization as a function of intensity.

Introduction

Ultrafast laser-matter interactions have been studied in physics, chemistry and materials research, and have lead to the discovery of interesting multiphoton and nonperturbative effects. These include above threshold ionization[1] and dissociation[2], and the study of high-order harmonic generation[3]. Long-term goals of research include the generation of attosecond laser pulses and coherent control: the ability to control atoms and molecules with light.

The phenomenon of nonsequential double ionization is a continuing area of research. The recollision model has been proposed[4], whereby an electron is initially tunnel ionized but then returns to the core and causes further ionization. It is well documented that recollision can occur when the laser field is linearly polarized, but not when it is circularly polarized[5]. The ionized electron is accelerated away from the core by the electric field, but as the field changes sign for the next half cycle, it can be driven back along its path to the core if the field is linearly polarized. However, if the field is circularly polarized, the electron will be deflected from its path by the perpendicular component of the laser field, and be unable to return to the core. Elliptically polarized light is used in this experiment to control the motion of the electron wave packet in the laser field.

As the laser field controls the recolliding electron, the point at which the electron first enters the continuum is also of great importance. The recollision model requires the electron to be released near the peak of the oscillating field. At the point of recollision, the initial electron must have enough energy to cause secondary ionization. The most likely velocity of an electron returning to the core corresponds to a kinetic energy of 3.17 times the ponderomotive potential[4].

The rate at which the wave packet spreads in the direction perpendicular to the linear laser field also determines the amount of recollision. The size of the spread electron wave packet is estimated from these results.

Experimental Configuration

The laser used was the Ti:Sapphire (40fs, 790nm, 10Hz) at RAL (UK) Astra, with a focused peak intensity of $8\text{x}10^{16}$ Wcm^{-2}. The 22mm diameter beam is f/11 focused into the interaction region of a time-of-flight mass spectrometer (TOFMS). Pulse length is stretched by 10% on entry to the vacuum chamber through a 3mm thick fused silica window. The laser field is linearly polarized, and is altered by rotation of a quarter-wave plate for this experiment.

Neutral Kr atoms were ionized in the interaction region of the TOFMS. Gas pressure inside the TOFMS was $3\text{x}10^{-8}$ mbar, low enough to avoid space-charge effects. Ions were extracted from the interaction region by an applied voltage of 300Vcm^{-1}, and passed through a 250µm aperture. Ions then entered a 110mm field free drift tube, and detected by a pair of micro-channel plates, from which the signal was retrieved.

Intensity Selective Scanning (ISS)[6,7] was used to produce results with high spatial resolution. The 250µm aperture before the TOFMS drift tube limits the spatial acceptance of the spectrometer, so that only a small slice of the laser focus is exposed to the detector. The focusing lens is then moved along the z-axis of the laser beam in sub-mm steps, translating the confocal volume with respect to the aperture. An average is taken from the ion signal detected at each lens position z_f.

This method has been combined with the novel experimental technique of effective intensity matching (EIM-ISS)[8,9]. The theory of EIM-ISS defines a constant ratio, R_{EIM} between the laser intensities of the linearly (I_{lin}) and circularly (I_{circ}) polarized laser beams such that the spatial distribution of the ions detected for each polarization are the same for all z_f values assuming that all nonsequential ionization processes are negligible. Results taken using Neon gas were used to define the ratio R_{EIM}, as it is the least susceptible of the noble gases to nonsequential ionization.

Recollision in Kr^{2+}

The ion signal from Kr^{2+} when using a linear, elliptical and circular laser field can be compared directly as the EIM-ISS technique has been utilized. The results are shown in Figure 1.

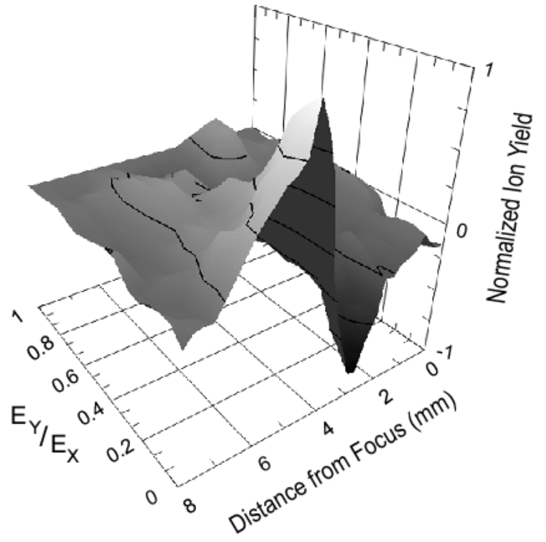

Figure 1. Graph showing only recollision effects in Kr^{2+} as a function of ellipticity and z_f position, which indicates on-axis intensity. 1 is circular, 0 is linear.

In this figure, the circular data has been subtracted from all results.

Both linear and circular fields are defined so as to produce the same amounts of primary ionization ($h\upsilon + Kr \rightarrow Kr^{+} + e$) and sequential ionization ($h\upsilon + Kr^{+} \rightarrow Kr^{2+} + 2e$), where the resulting electron does not return to the ionic core. Recollision ($e + Kr^{+} \rightarrow Kr^{2+} + 2e$) is not detected with a circular field. By subtracting the circular data, all contributions from both primary and sequential ionization are removed. Only the non-sequential effect of recollision is presented in Figure 1.

The y-axis shows ellipticity E_y/E_x where 1 is circular and 0 is linear. It can be seen that the returning electron becomes increasingly efficient at re-ionizing the nucleus as the field changes from circular to linear. At y = 0 maximum recollision is observed in the peak at $z_f = 4.5$mm. The fact that this peak occurs at relatively high z_f also indicates that the nonsequential production of Kr^{2+} reaches a maximum at an intensity lower than saturation, as ionization of Kr^{2+} is saturated (> 3×10^{14} Wcm^{-2}) in the region $0 < z_f < 3$mm.

The trough at 3mm is due to an increase in Kr^{3+} produced by recollision ($e + Kr^{2+} \rightarrow Kr^{3+} + 3e$), where a corresponding reduction in sequential production of Kr^{2+} is detected.

Deconvolution of a Non-Gaussian Focus

For a quantitative analysis of these results, the geometry dependence of the ionization yields must be removed. The traditional method to perform this deconvolution is described in[6]; however this method removes only the Gaussian focusing. Although the Ti:Sapphire laser generates pulses with a Gaussian profile in the far field, optics transporting the beam into the TOFMS interaction region will cause truncation of the beam, resulting in a non-Gaussian focus. An alternative model has been used[8,9], which accounts for these apertures.

In order to represent a typical experimental system, beam diffraction is quantified before focusing takes place. The analytical method involves propagating a truncated Gaussian beam through an ABCD optical system. Figure 2 shows the data for ellipticities close to linear polarization, deconvoluted using this non-Gaussian model.

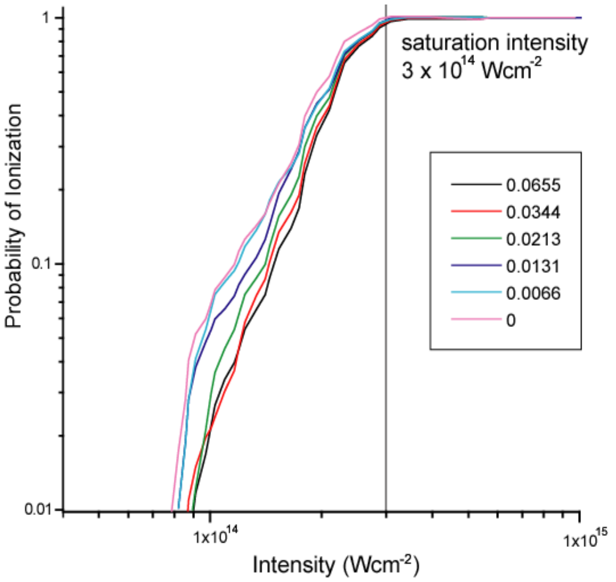

Figure 2. Deconvoluted EIM-ISS results for elliptical and linear polarizations displaying recollision. Only those ellipticities closest to linear polarization are shown for clarity. At an ellipticity of 0.0344 (red line) no recollision effects are discernible.

In Figure 2, the classic signature of recollision is seen in the 'shoulder' feature in the region of 10^{14} Wcm^{-2}. The shoulder is most pronounced for the linear curve (pink line). It can be seen from Figure 2 that recollision appears to occur only with ellipticities very close to linear polarization. This suggests that only a small perpendicular field is required to deflect the returning spread electron wave packet form the core. By measuring the amount of deflection, i.e. the perpendicular displacement of the wave packet, an estimate can be made of the size of the electron wave packet at its point of return.

Measuring the Spread of the Electron Wave Packet

The displacement of the recolliding electron has been calculated using classical equations. The electron's initial position and velocity at the moment of release into the electric field was selected so that a return to the core will occur. The size of the perpendicular component of the electric field was calculated at the ellipticity for which recollision becomes undetectable. It was assumed that the returning electron ionizes the core on the first pass of its trajectory.

The perpendicular displacement of the electron wave packet was calculated to be 2Å. The diameter of the core is of the order of 1Å. For recollision ionization to occur there must be significant overlap between the spread electron wave packet and the core. This indicates that the diameter of the spread wave packet has a radius of approximately 1.5Å. This is in reasonable agreement with other studies on Argon[10].

If the classical electron radius is taken as the size of the wave packet on release, the transverse spread of the electron wave packet is 1.2 Å/fs. The spread is assumed to be linear during a half cycle of the laser field oscillation (1.3 fs). This is in good agreement with an earlier estimate of 1.5 Å/fs[4].

Conclusions

Ion yields from recollision have been used to estimate the dispersion of the returning wave packet. The fact that results are in good agreement with other studies indicates that this is a valid technique, using the well established mechanism of recollision ionization.

Other factors to be considered as a result of electron-ion interaction include elastic scattering of the electron for above threshold ionization. Following collision, defined by the impact parameter, electrons are produced with much greater energy than the ponderomotive potential. This will yield further information about the angular dependence of correlated electron wave packet production.

Control of the returning electron wave packet by the laser field has also been demonstrated. This is of importance for high harmonic generation, leading to attosecond pulse production.

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