Atomic excitation during recollision-free ultrafast multi-electron tunnel ionization

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Introduction

When atoms are subjected to intensities generated by modern high-power laboratory-scale ultrafast laser systems, ionization readily results: the massive electric field induced by the radiation causes near-instantaneous distortion of the distribution of charge around an atom. However, treating ionization as simply severing electrons from the atom in a classical manner is insufficient. At the opposite end of the intensity scale, optical ionization of an atomic system can proceed via the absorption of multiple photons leading to the creation of a ladder of virtual states. However, at the field strengths present, a purely perturbative multiphoton treatment is also incomplete. The foundations of modern theoretical descriptions of the nonlinear photoionization of atoms exposed to high intensity radiation were laid in 1964 by Keldysh1). The manner in which electrons are liberated from an atom can be summarised in a relatively straightforward manner through the Keldysh adiabaticity parameter, γ, the ratio between the frequency of the laser light and the frequency of an electron tunnelling through a potential barrier. While a large number of experimental and theoretical works continue to investigate these mechanisms2), the original definition of Keldysh still stands: when $\gamma >> 1$, multiphoton ionization (MPI) results, and when $\gamma << 1$, the electron escapes via a tunnel ionization (TI) process.

Method

Given their ease of availability and handling, Noble gas atoms have long been the staple of atom-laser interaction studies. A number of pivotal experimental studies over the past decade have observed multiple ionization as laser intensity is varied, and have revealed much about the complex and nonlinear nature of atomic ionization3). However, the majority of such studies are somewhat hampered by the manner in which the laser intensity is controlled. By changing the energy of the focused laser pulse, the peak intensity may be accurately controlled. This method results in a variation of the geometrical "size" of the laser focus, and given that the processes of interest depend strongly on laser intensity *and* the volume of the laser focus generating this signal, direct comparisons with theory are impossible without including an instrumental distortion.

A novel solution to this so-called "volume variation" problem was suggested by Van Woerkom and co-workers4): by loosely focusing a high energy laser pulse into a tightly apertured spectrometer, only those ionization states generated within a narrow spatial (and thus intensity) window are detected. By translating the focusing optic, the laser intensity may be controlled, thus intensity selectively scanning (ISS) is feasible. Our adaptation of this technique to experimentally realistic nongaussian laser focuses5) is described in detail by J. Wood (UCL) 6). Suffice to say we are now able to recover volume- and diffraction- independent ionization probabilities, directly equivalent to theoretical predictions.

In a linearly polarized laser field, one or more liberated electrons may be driven by the laser field, and in some cases, excitation7) or further ionization8) of the ion can be caused by the recolliding electron if it passes sufficiently close to the ion. This process is investigated in detail by E.M.L. English (UCL) 9). However, in this work, we have made recollision events *negligible* by employing circular polarisation in order to clarify the mechanisms underlying multi-electron tunnel ionization (METI).

We present an experimental investigation of the behaviour of argon exposed to 50fs pulses over the intensity range 10 TW/cm^2 to 100 PW/cm^2 using the ISS technique to carefully detect the probability of tunnel ionization. The experimental study to be presented was carried out at the Astra laser facility at the Rutherford Appleton Laboratory, with typical operating parameters: 790 nm 50fs pulses delivering 30mJ at a repetition rate of 10Hz. Such pulses were transmission focused using f /11 optics creating a macroscopic focus of peak intensity $I_0 \approx 10^{17}$ Wcm^{-2}. The ionization processes under investigation require between 10 TW/cm^2 and 10 PW/cm^2 to saturate thus the usable range of the focused Astra beam extends over tens of mm. The experimental technique will be discussed in detail elsewhere, in summary, the laser was focused into the source region of an ultra-high vacuum (4×10^{-10} mbar) time-of-flight mass spectrometer (TOFMS). Argon was introduced into the TOFMS as a defuse jet, with the number density low enough to negate space-charge effects. Ionized argon atoms were electrostatically extracted through a 250 μm aperture, and detected on a pair of microchannel plates after 11 cm of field-free drift and the ion signal averaged over 10^3 laser shots. The laser focus was translated by a computer controlled motion stage, exposing the detector to different "slices" through the focal volume, where the intensity distribution present in a particular slice is represented by the relative ion charge states.

The partial probability of ionization to Ar^{n+} (n = 1 to 6) is recovered from the ISS data using the technique as discussed5). The distinction "partial" is necessary as the recovered probability distribution is only accurate at intensities below saturation. Above saturation, the deconvolution technique is known to break down, thus the PPI is defined as unity. This limitation does not hinder our understanding of the atomic ionization mechanism: a physically realistic measure may be readily recovered, referred to as the conserved probability of ionization (CPI), as presented in Figure 1.

Theoretical treatment

While recollision excitation and ionization in ultrafast laser pulses has generated a lively debate between a number of experimental and theoretical groups around the world, little mind has been given to the possibility of excitation during METI, not through the influence of the returning electron, but a behaviour *inherent* to laser - atom interactions. An exception to the previous statement is the recent theoretical work of Zon and co-workers10), who calculated the probability of tunnel ionization in 5 fs and 50 fs circularly polarized laser pulses for Ar and Kr using Keldysh theory. The salient points of the treatment of Kornev *et al.*10) are the following. The Keldysh theory of tunnel ionization of an atom is generalized to account for (i) TI and core excitation where the action of the departing electron excites the one of the remaining electrons in the ion. This process is referred to as inelastic tunnelling, (ii) the

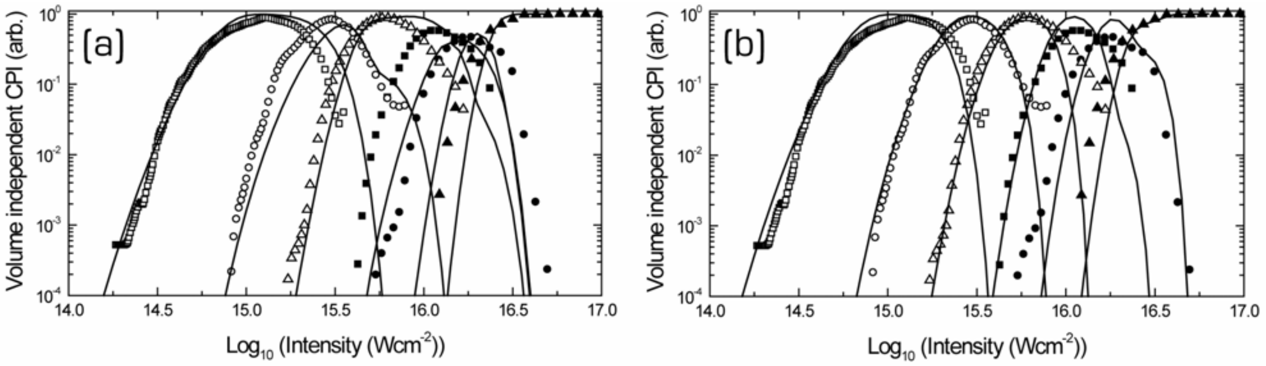

Figure 1. Volume- and diffraction-free conserved probability of ionization (CPI) to Ar^{n+} (where n 1 to 6, left to right) in a 50fs 790 nm circularly polarized laser pulse, where both frames contain the same experimental points. (a) A poor agreement is found in a comparison between the present results (data points) and the theoretical prediction of Kornev *et al.*[10)] when only sequential ionization from the ground state is considered. (b) A remarkable agreement is observed when core excitation, and cascaded multi-electron processes are included.

multiple TI of *N* equivalent electrons (iii) combinations of single- and multi-electron cascaded reactions through combinations of (i) and (ii). Such a generalization required a dramatic expansion of the number of levels involved in predicting METI as compared to traditional so-called ADK treatment. Kornev *et al.* also considered the generalized overlap integrals necessary to describe correctly excitation during METI. The predictions of Zon and co-workers are prime for comparison with our CPI data, as they present their data in terms of a spatially uniform intensity. Such a comparison is presented in Figure 1, where the present experimental data are the data points, the theoretical prediction of ground-to-ground state ionization only are compared in Figure 1a, whereas ground and excited state transitions are compared in Figure 1b. Note the CPI data has been shifted to a slightly higher intensity to improve the agreement with the theoretical curves.

Figure 1a and 1b clearly highlights the necessity to consider multi-electron excitation processes during the laser pulse, even with no recollision present. When pure ground state sequential TI is considered (Figure 1a), an acceptable agreement is found for the lowest charge states. This agreement quickly fails, with the fit to Ar^{4+} and Ar^{5+} being particularly bad. In contrast, when excitation processes are included in the METI prediction, an excellent fit is found for all charge states. A comparison between Figure 1a and 1b demonstrates that METI between Ar^{+} and Ar^{3+} is relatively well described by a ground state only process, with a small contribution due to excitation. However, in the case of Ar^{4+} and Ar^{5+}, the major significance of ionic core excitation is evident.

Conclusion

To conclude, we have recorded for the first time strong evidence for the presence of considerable atomic excitation during tunnel ionization by an ultrafast circularly polarized laser pulse, particularly for intermediate charge states. Given the polarization of the laser pulse, such an observation is made without the need to embrace recollision processes at all. We intend to extend our discussion of this important observation at conference.

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