Probing atomic ionization mechanisms in intense laser fields by calculating geometry and diffraction independent ionization probabilities

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Abstract

Unprecedented insight into the mechanisms responsible for the ionization of xenon in an intense laser field has been achieved. This has been possible by combining the new technique of effective intensity matching with intensity selective scanning and accurate modeling of the energy distribution in a non-Gaussian laser focus.

Introduction

Modern ultrafast high power laser systems typically generate pulses with a Gaussian profile in the far-field, yet, geometric constraints imposed by the optical transport system prevent this perfect profile from reaching experimental chambers. For many practical applications this poses no problem as the confocal volume normally lies within the volume imaged by the detector employed. However, this is not the case with intensity selective scanning (ISS)[1] where the acceptance volume of the detector is physically limited. Therefore it is essential to quantify the spatial distribution of intensity within the laser focus in order to fully understand the laser atom system under investigation.

Starting from the Collins form of the Huygens-Fresnel integral an analytical solution has been deduced for the propagation of a truncated Gaussian laser beam through an arbitrary ABCD optical system. The solutions from which are then employed to remove the geometrical influence of the volume of the laser focus from atomic ionization measurements. The removal of the volume is done in a manner much akin to that described by Walker *et al.*[2] but with the on axis intensity distribution given by our non-Gaussian treatment. Such analysis yields geometry independent ionization probabilities as a function of intensity which in turn allows determination of the dominant ionization mechanism.

Experimental Method

In the experiment laser pulses (τ = 55fs, λ = 790nm, E = 20mJ, dia. = 22mm) of known polarization are focused into an ultra high vacuum chamber by a 25cm lens mounted on a translation stage. The chamber is then filled with xenon to a pressure of 1×10^{-8} mbar. A time of flight mass spectrometer (TOF-MS) with a 250µm entrance aperture is then used in conjunction with the precise movement of this focusing lens to produce a complete ISS-scan of the interaction region.

Linear and circular polarized pulses are used in the experiment in order to investigate recollision ionization. The circularly polarized pulses are used as a control, as freed electrons will not recollide with their parent ions when exposed to such a field[3], thus the ion yields obtained with circularly polarized light are in no part obtained through none sequential ionization processes. To allow such a comparison we employ intensity selective scanning with effective intensity matching (EIM-ISS). This technique works by finding a constant ratio, REIM, between the linear and circular polarization laser intensities; such that the relative ion yield is kept constant regardless of polarization as the detector is translated across the focus. For this experiment EIM was obtained by setting the linear intensity to be 0.65 that of the circular[4]. This value was deduced from extensive tests with neon in a TOF-MS apparatus. Neon was selected for the calibration as it exhibits low levels of non-sequential ionization[5] allowing comparison across the widest possible range of z ≈ 10mm (which is equivalent to an intensity range of 10^{13}-10^{16} Wcm^{-2}).

The non-Gaussian treatment of the laser focus is required to account for the effects of diffraction upon the laser beams intensity profile[6,7]. The main source of this diffraction was found to be the last aperture before the focusing lens.

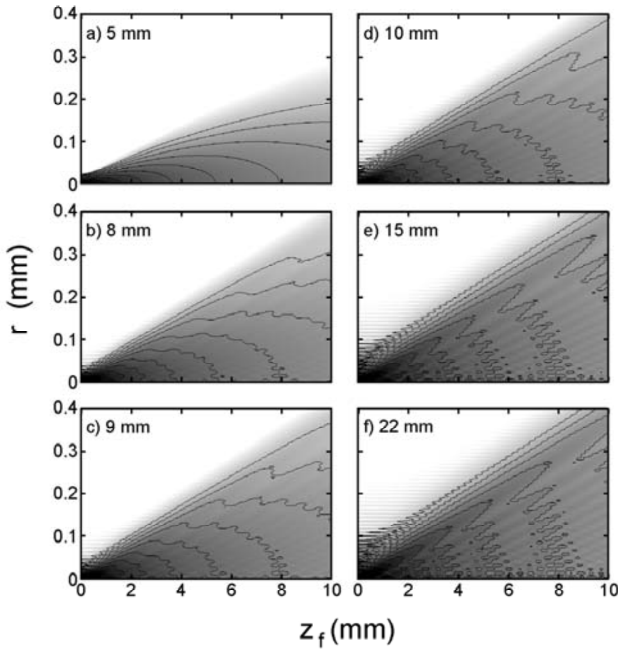

Figure 1. Simulated intensity distributions for a range of beam diameters passing through an aperture of radii 11mm. The plots are shown on a logarithmic grey scale with isointensity contours separated by an order of magnitude.

Figure 1 above shows the effects of diffraction from this aperture on the intensity profile of the laser focus. On such a plot a perfect Gaussian focus would have smooth isointensity contours; diffraction introduces oscillations upon them. As the beam diameter approaches that of the aperture the Gaussian profile get increasingly distorted.

Panel f) in the Figure represents the intensity distribution of the laser focus used in the experiment. Here the diameter of the beam matches that of the aperture and the diffraction effects are most pronounced.

Results and Discussion

At each z position time of flight spectra compiled over 500 laser shots are recorded. From these spectra a complete ISS-scan of the focus is achieved, which can be integrated to give relative ion yields, see Figure 2.

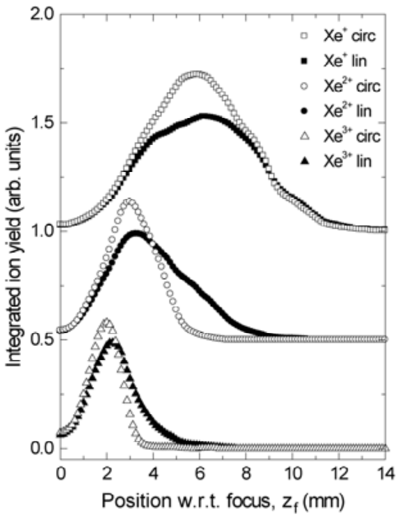

Figure 2. Raw EIM-ISS data for the ionization of xenon to Xe^{n+} (n = 1, 2, 3,). The integrated ion yield is recorded as a function of the focusing lens position, z_f, with respect to the axis of the spectrometer. The presence of recollision ionization is apparent in the Xe^{2+} and Xe^{3+} signal as an enhancement of signal at low intensity in the linear data compared to the circular data[4]. Ripples in the ion yield are a consequence of diffraction.

The integrated ISS-scan is then deconvoluted, using the relevant non-Gaussian model of the laser focus for the experiment. This yields geometry free partial probability of ionization (PPI) as a function of intensity.

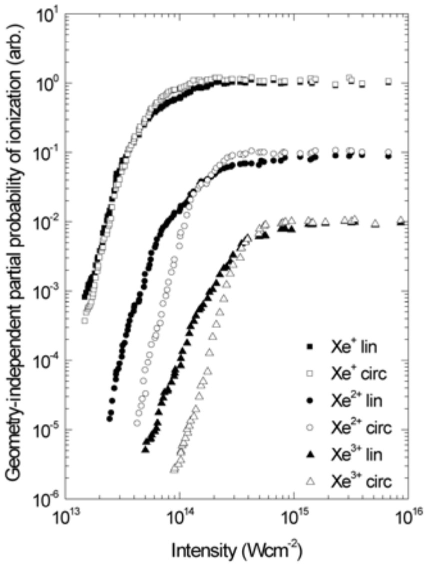

Figure 3. Geometry and diffraction free partial probability of ionization for Xe^{n+} (n = 1, 2, 3,) measured with circular (open symbols) and linear (closed symbols) polarizations as a function of intensity. Increasing charge states are normalized to descending orders of magnitude for clarity.

Doing this for both sets of data, as in Figure 3, allows the decoupling of the non-sequential and sequential ionization. This can be seen in the figure by comparison of the PPI curves in the low intensity regime. For the two higher charge states, where recollision is expected, there is an enhancement of probability of approximately an order of magnitude. The PPI curves for Xe^{+} show no variation with polarization as no recollision ionization is present.

In order to establish how the dominant sequential ionization mechanism changes with intensity, intensity is translated to Keldysh parameter[8].

$$\gamma = \frac{\omega_{laser}}{\omega_{tunneling}} = \sqrt{\frac{E_i}{2U_p}} \qquad (1)$$

Equation (1) defines the Keldysh parameter,γ. It relates the laser frequency, ω_{laser}, to that of the tunneling electron, $\omega_{tunneling}$. E_i is the ionization potential of the atom and U_p is the pondermotive potential of the laser field ($U_p = 9.33\times10^{-14}\ I\ \lambda^2$) both expressed in terms of eV.

When γ >> 1, multiphoton ionization (MPI) dominates as the laser frequency is much greater than the tunneling frequency. When the two frequencies are almost equal, γ ≈ 1, tunneling ionization (TI) dominates. If the laser frequency is much lower than the tunneling frequency, γ << 1 and the most prominent mechanism is field ionization (FI).

In Figure 4 the FI region gives a zero gradient. Where MPI dominates a constant positive gradient is observed, from which, the number of photons involved can be deduced. The remaining section of the graph is the region where TI dominates. This is seen to vary with charge state although the point of saturation of the PPI curves is similar for all the states shown.

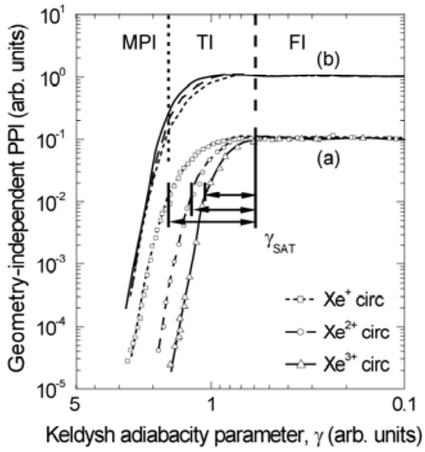

Figure 4. Geometry independent partial probability of ionization (PPI) as a function of Keldysh parameter for Xe^{n+} (n = 1, 2, 3,) for circular polarization. (a) Direct conversion of intensity to Keldysh parameter. (b) Scaled conversion of intensity to Keldysh parameter (× 1.00 for Xe^{+}, × 0.76 for Xe^{2+} and × 0.62 for Xe^{3+}).

Conclusion

The new non-Gaussian treatment of a laser focus combined with effective intensity matching has allowed significant advancements to be made in the understanding of atomic ionization in intense fields. With the increased clarity gained from the new techniques the physics involved can be scrutinized to evermore depth and exciting conclusions drawn.

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