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Nonsequential triple ionization of argon atoms in a high-intensity laser field

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We investigate direct, nonsequential ionization of argon atoms using linearly polarized laser light at 1053 nm with a 600-fs pulse length. We report the production of triply charged argon ions at intensities below the saturation intensity of the first charge state.

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The possibility of nonsequential (NS) double ionization of atoms that are exposed to intense laser fields has been discussed for over a decade [1]. Here "NS double ionization" refers to the simultaneous removal of two electrons rather than a sequential process of removing one electron then removing another a short time later. Recently interest in this topic has been renewed and a number of experimental results have been published along with the proposal of various theoretical models to explain this behavior [2–11]. Two widely discussed models are the "shake-off" model [2] and an electron rescattering model [3].

The "shake-off" model describes the NS process as a mechanism where one electron is ionized by the laser field and the departure of this electron is so rapid that the remaining electrons are "shaken up" enough to free an additional electron. The model is not wavelength or polarization dependent, in principle; however, Walker *et al.* [7] have mentioned that the angular momentum that is absorbed by the electron with circularly polarized light could inhibit the transfer of energy to additional electrons in the shake-off process. This would make the ionization enhancement less noticeable for circularly polarized light than for linearly polarized light.

The rescattering model proposed by Corkum is a model that has not only been used for NS ionization but has also had a large degree of success in describing high harmonic generation and above-threshold ionization (ATI). This model describes the NS ionization as a process whereby an electron is tunnel ionized. The electron then interacts with the laser field where it is accelerated away from the nuclear core. If the electron has been ionized at an appropriate phase of the field, it will pass by the position of the remaining ion half a cycle later, where it can free additional electrons by electron impact. Only half of the electrons are released with the appropriate phase and the other half will never return to the nuclear one.

The electron-impact ionization cross section is highly dependent on electron kinetic energy, and drops dramatically for electron energies below the ionization potential of the bound electrons. The maximum (and most probable) kinetic energy that the returning electrons can have is 3.17 times the ponderomotive potential (U_p) of the laser. This places a cutoff limit on the minimum intensity (intensity is proportional to U_p) where ionization due to rescattering can occur. The production of ions at intensities lower than this cutoff intensity must be due to some process other than electron rescattering. This provides a useful check on the applicability of the model in the low-intensity region. Another important parameter that can shed light upon the applicability of this model is the dependence on laser polarization. A strong polarization dependence is predicted with the rescattering model. With increasing ellipticity the freed electron will return to the core with an ever-increasing impact parameter, thereby causing a rapid decrease in the efficiency of ionization by e-$2e$ scattering.

We have studied the NS ionization of argon using an Nd:glass–Ti:sapphire laser operating at 1053 nm with a pulse length of approximately 600 fs. The laser was focused using $f/30$ optics into an ultrahigh-vacuum chamber having a background pressure of 2×10^{-9} Torr. A large f-number focusing lens was used to maximize the focal volume of the interaction region in order to obtain ion signals for very low ionization rates. Ion species were separated with a time-of-flight spectrometer having a 60-cm-long drift tube and detected with an electron multiplier tube having a gain of approximately 10^6. The repetition rate of the laser was one shot every 45 s, which made it necessary to use single-shot data rather than averaged data. Ion curves were produced by combining a series of intensity scans, each having a different fill pressure in the interaction chamber. The argon pressure in the interaction chamber was controlled by a precision leak valve and ranged from 5×10^{-8} to 1×10^{-4} Torr. Higher pressures could only be used for low ion number, since space-charge effects and detector saturation had to be avoided at all times. We were not able to obtain quantitative data for the fourth charge state due to impurity peaks in the ion spectra.

The occurrence of NS double ionization is now well known, and evidence of it can be seen in previously published data [12,13], using nearly the same parameters (ionization of argon at a wavelength of 1053 nm and a pulse length of approximately 1 ps); however, no discussion regarding possible mechanisms for the NS ionization was made on the evidence at that time. The results we present here in Fig. 1 show evidence of NS triple ionization as well. A significant triply charged ion signal is present at laser intensities lower than the saturation intensity of both the second and first charge states. This indicates the appearance of

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triply charged ions at intensities where there are still neutral atoms remaining in the laser focus. There are two possible channels through which the triply charged ions can be created: NS triple ionization of neutral atoms, or NS double ionization of singly charged ions. We conclude that both NS triple and NS double ionization are occurring, as we discuss below. We have also seen the appearance of the fourth charge state at an intensity lower than the saturation intensity of the second charge state. This position is marked by an arrow in Fig. 1. Again, the 4+ ions probably result from both NS double and triple ionization.

Theoretical curves can be calculated for NS ionization models by including a channel for the production of doubly charged ions directly from neutral atoms and for triply charged ions from both the first charge state and from the neutral atoms. The populations of each charge state can be described by the following coupled rate equations, which allow for direct double and triple ionization:

$$\frac{dN_0}{dt}=-N_0(R_{01}+A_{02}+R_{03}),$$

$$\frac{dN_1}{dt}=N_0R_{01}-N_1(R_{12}+R_{13}),$$

$$\frac{dN_2}{dt}=N_0R_{02}+N_1R_{12}-N_2R_{23},$$

$$\frac{dN_3}{dt}=N_0R_{03}+N_1R_{13}+N_2R_{23}-N_3R_{34}. \tag{1}$$

Here N_i is the population of the ith charge state and R_{ij} is the transition rate from the ith charge state to the jth charge state.

These equations are solved using the Ammosov-Delone-Krainov (ADK) tunneling formula [14] for sequential ionization terms ($R_{i,i+1}$), and the remaining R_{ij}'s are estimated by means of the rescattering theory of Corkum [3]. We proceed in a manner similar to that outlined in Refs. [3,7,9].

After the electron has been freed, its motion is determined primarily by its interaction with the laser field where its expansion follows approximately that of a Gaussian wave packet [7]. The spatial extent of the wave packet after half a laser cycle, along with the electron-impact ionization cross section and the kinetic energy of the free electron, determines the amount of ionization that occurs with this process. The evolution of the Gaussian packet width in atomic units can be described as $\alpha_t=\sqrt{\alpha_0^2+(2t/\alpha_0)^2}$, where α_0 is the width at $t=0$. Using $\alpha_0=3.8$ Å for the diameter of neutral atomic argon [15], we find that $\alpha_t=11.4$ Å half a laser cycle later for our laser frequency. The cross section for ionization by electron impact is highly dependent on the electron kinetic energy [16]. We have included this dependence in our calculations, using the field-free inelastic scattering cross section, since it has been shown that the scattering cross section is not appreciably altered by the presence of the laser field [11]. An additional assumption is that 50% of the freed electrons will reencounter the nuclear core with a kinetic energy equal to 3.17 times the ponderomotive potential at the time of ionization. This is by far the most probable kinetic energy for the returning electrons to have [3], but there is still a significant fraction of returning electrons that have a somewhat lower kinetic energy. Consequently our calculation overestimates the contribution to ionization by this rescattering mechanism.

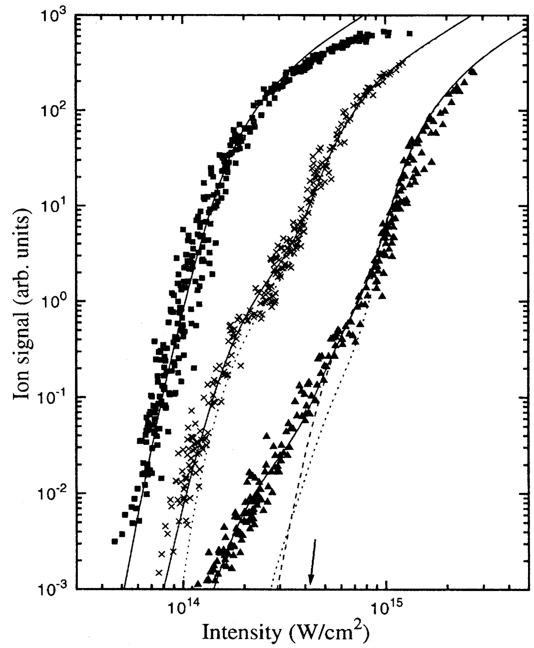

FIG. 1. Argon-ion signal as a function of incident laser intensity for a 600-fs 1053-nm linearly polarized laser pulse. Each point represents a single laser shot. The calculated curves are shown as dotted lines (ADK plus electron rescattering model), solid lines (ADK plus curve fit for NS double and triple ionization), and dashed lines (ADK plus curve fit for NS double ionization only). The experimentally measured intensities are multiplied by 1.2 for comparison to the calculations.

The rescattering model calculations are shown in Fig. 1 as dotted lines. The model produces significantly better agreement with the data than a purely sequential model would; however, it clearly fails at the low-intensity end of the third charge state, and for the second charge state it cannot completely account for the ion production in view of the fact that the calculation is an overestimate. This suggests that, if the electron rescattering process is contributing to the NS ionization, it is not the dominant factor. It is important to note that the discrepancy at the low-intensity end of the third charge state cannot be attributed to uncertainties in the experimentally measured intensities for the following reason. We have an experimental uncertainty in the absolute laser intensity of approximately 50%; however, the relative uncertainty is much smaller, and evidence of this can be seen by the small amount of scatter in the data. The fact that the relative uncertainty in the intensity is small indicates that the spacing between charge states in Fig. 1 is known with a high degree of precision. The spacing of the theoretical charge states is known exactly, and it is a comparison of these spacings that leads to the conclusion that the electron rescattering model fails at the lower end of the second and third charge states.

We have also solved the coupled rate equations (1) by using the ADK formula for sequential ionization and the remaining R_{ij}'s as free parameters in a curve fit. In this case the NS R_{ij}'s are modeled as a fixed (not intensity-dependent) fraction of the sequential $R_{i,i+1}$ terms. Not surprisingly we can produce better agreement than with the rescattering model, but the important point is that the data for the third charge cannot be fit without the inclusion of the term R_{03}, which is the term for NS triple ionization. The dashed line in Fig. 1 shows the curved fit using $R_{03}=0$. The fact that the R_{03} term is required to produce the correct spacing between charge states suggests that NS triple ionization is occurring. The best values found for the curve fit are $R_{02}=R_{01}/110$, $R_{03}=R_{01}/11\,000$, and $R_{13}=R_{12}/167$ for the solid lines; and $R_{02}=R_{01}/110$, $R_{03}=0$, and $R_{13}=R_{12}/125$ for the dashed line.

Due to time limitations on laser time allocated to this experiment it was not possible to obtain quantitative data for circular polarization; however, we have observed with circular polarization that the appearance of the second charge state does not occur until the signal from the first charge state has saturated. Likewise, the third charge state does not appear until the second charge state has saturated. These observations are consistent with the electron rescattering model, any model involving resonant enhancement, and the shake-off model, as discussed in Ref. [7].

Previously published work on the NS ionization of helium atoms using 160-fs pulses at 780 nm has also shown that the electron rescattering model cannot fully account for the NS ionization [7]. There are two arguments for this. First, there is no low-intensity cutoff of the second charge state, as there should be. Second, the high-intensity contribution is estimated to be too small to completely account for the enhanced ion signal. The results we present here also indicate that another mechanism must be present to fully account for the NS ionization. Whether this additional mechanism can be described as a "shake-off" process, a transient resonant enhancement by autoionizing states, or some other process is presently unknown. One such other process that has not been explained before, to our knowledge, is the strong field-enhanced absorption of the high-order harmonics emitted by the atoms or ions during the interaction. We are now actively exploring this idea.

In summary, we report an observation of triply charged argon atoms at laser intensities lower than the saturation intensity for both the first and second charge states. This is the result of direct double and triple ionization from singly charged ions and from neutral atoms. The results are qualitatively consistent with a shake-off model [2], but no quantitative comparison can be made presently with this model. Ionization rates predicted by a rescattering model [3] are too low to fully account for the observed ion production.

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