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Double ionization in the perturbative and tunneling regimes

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We have studied the doubly charged ion yields and electron energy spectra (normal and coincidence) for double ionization of atoms in two different ionization regimes (perturbative and tunneling). In both cases, the double-ionization rates appear to be anomalously large in some intensity range and strongly reduced by circular polarization. It is argued that these similar behaviors must result from different physical mechanisms.

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The simplest mechanism by which two or more electrons can be ejected by a strong optical field is a sequence of one-electron removals, possibly enhanced by ionic resonances like in the case of alkaline-earth-metal atoms [1]. There are a few cases where this interpretation cannot account for the observations. The first case is the double ionization of xenon by 50-ps, 0.53-μm pulses [2]. The Xe^{2+} ion yield versus intensity displays a characteristic knee that prevents a kinetic description based on a single rate (see Fig. 1). The commonly accepted phenomenological scenario to explain this behavior is that double ionization is achieved through a *direct* two-electron process with an enhanced rate that terminates upon depletion of the neutral ground state. For intensities beyond this neutral saturation, another rate applies that accounts for the (*sequential*) ionization of the ground-state ion. However, no physical reason has been proposed that explains why the direct process is so greatly enhanced. Another example of a nonsequential process has been reported for the double ionization of helium by 100-fs, 0.62-μm pulses [3]. The shape of the He^{2+} ion yield curve is qualitatively identical to the xenon case discussed above, except for one major difference; helium ionizes nonperturbatively while xenon is perturbative.

In this Rapid Communication, we will address the issue of double ionization as it pertains to the situation discussed above, that is, the occurrence of a nonsequential rate or knee structure. The question to be put forth is whether the observation of a knee structure (two rates) qualifies the conclusion of direct two-electron ejection. In other words, do all atoms that manifest such a structure behave the same? This study examines the polarization dependence of the ion yield for the above mentioned cases. These two cases differ by two orders of magnitude in terms of their saturation intensity. Consequently, xenon ionization is consistent with lowest-order perturbation theory (LOPT), while helium behaves nonperturbatively. We also employ electron spectroscopy to obtain more insight into the underlying physics and report the results of electron-electron (EE) and electron-ion (EI) coincidence measurements for the xenon case. The results are discussed in terms of recently proposed models.

The first experiment has been carried out with a 1-kHz repetition rate Nd:YLF (neodymium-doped yttrium lithium fluoride) laser producing 50-ps, 0.527-μm pulses [4]. The ions are charge analyzed in a time-of-flight mass spectrometer. The resulting ion yields as a function of intensity for both polarizations are plotted in Fig. 1. The Xe^{2+} ion yield does indeed show a knee or double rate for linear polarization, as reported by L'Huillier *et al.* [2]. Furthermore, this knee is completely suppressed for circular polarization. The intensity dependence for the production of Xe^+ and Xe^{2+} (CP case) ions is consistent with LOPT, as expected for multiphoton ionization at moderate intensities (10^{12-13} W/cm^2).

In order to extract more information we have carried out EI and EE coincidence measurements, which is possible due to our laser's high repetition rate. The EI coincidences are detected in a collinear electron-mass time-of-flight spectrometer design. After a sufficient time delay for the electrons to leave the interaction region, a voltage is applied to accelerate the ions into the second time-of-flight tube. The ions are discriminated according to their charge and the electron energy spectrum recorded in coincidence with one or the other charge state. The normal photoelectron and EI spectra are shown in Figs. 2(a) and 2(b), respectively. The EI technique enhances the sensitivity to detect electrons resulting from the sequential double ionization (peaks marked with asterisks), which are otherwise masked in the background of the normal spectrum. This measurement reaffirms that the majority of the doubly ionized species are produced by sequential ionization of the Xe^+ ground state, although the sensitivity is inadequate for detecting electrons resulting from the knee region.

The detection of the *direct* two-electron process could be revealed by EE coincidence measurements. In such a process, the two electrons released by the xenon atom will be strongly energy correlated. Let the photon energy be

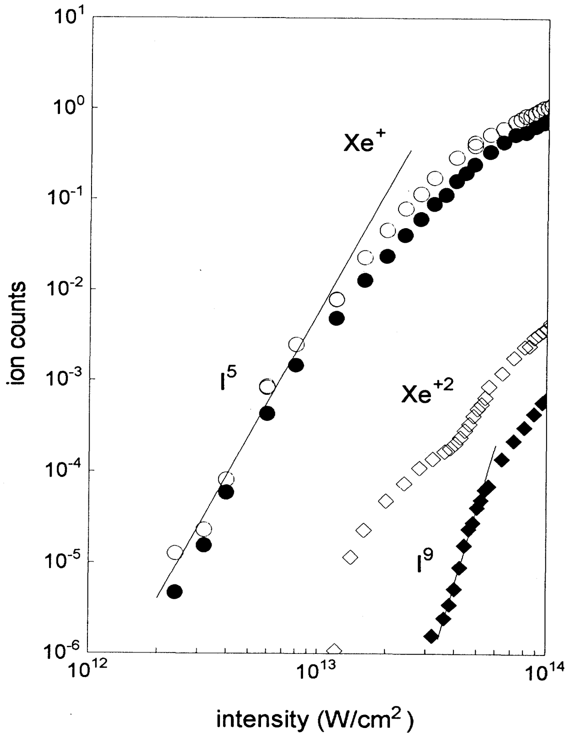

FIG. 1. Intensity dependence of the ion yield curves for Xe^+ and Xe^{2+} resulting from 527-nm, 50-ps excitation of neutral xenon. The open circles and diamonds are taken with linearly polarized (LP) light and the closed circles and diamonds with circularly polarized (CP) light.

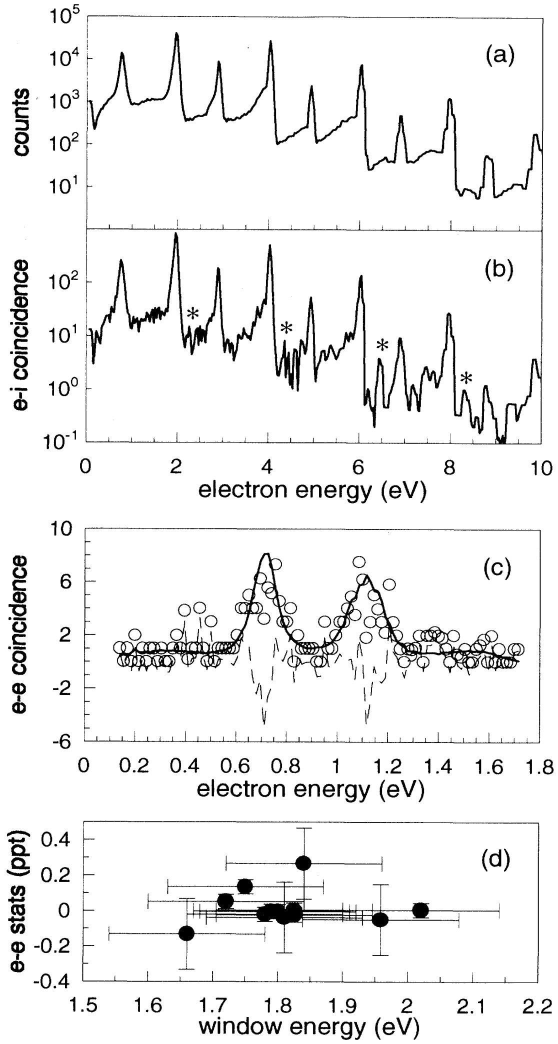

FIG. 2. Electron energy spectra resulting from ionization of xenon by 527-nm, 50-ps pulses. The normal spectrum (a) consists of a series of above-threshold ionization peaks resulting from the sequential ionization of neutral xenon forming the Xe^+ ground $P_{1/2,3/2}$ state at 0.8 and 2.0 eV, respectively. The electron-ion coincidence spectrum (b) shows the enhanced peaks (asterisks) corresponding to the sequential ionization of the Xe^+ ground state. The electrons are detected in this spectrum in coincidence with the Xe^{2+} ion. The electron-electron coincidence spectrum (c) shows the raw coincidence data (open circles), calculated false coincidences (solid line), and the difference (dashed line) for an energy window of (1.75±0.06) eV. A typical EE spectrum is an average over 3.6–7.2 million laser shots. (d) shows the integrated qualifying coincidences vs window energy in units of parts per thousand (ppt).

$\hbar\omega$, and N the minimum number of photons necessary to overcome the total binding energy E_b of the two electrons (sum of the atom and ion ionization potentials). Then the electron kinetic energies E_1 and E_2 are correlated through energy conservation $E_c \equiv E_1 + E_2 = N\hbar\omega - E_b$. Experimentally, an energy window is defined as some center energy plus some bandwidth, $E_c \pm \delta E$, and electrons whose summed energies on a single laser shot fall within this window are recorded. The experiment is then repeated by changing the value of E_c. The electrons were collected over a 2.4π sr solid angle by a parabolic electrostatic mirror and energy-analyzed in a time-of-flight spectrometer [5]. A typical EE coincidence spectrum is shown in Fig. 2(c). The integrated coincidence counts, following subtraction of the false coincidences between background and single-ionization electrons, are displayed in Fig. 2(d) as a function of E_c. The result is zero coincidence events within $\pm 4 \times 10^{-4}$ and sets an upper limit for the relative number of direct events of about two orders of magnitude lower than the Xe^{2+}:Xe^+ ion ratio.

The second set of results concerns double ionization of helium by 100-fs, 0.62-μm pulses [6], as shown in Fig. 3 for both linearly and circularly polarized light. Both the laser and the electron-ion spectrometer have been described elsewhere [5]. Care was taken to insure reduction of artifacts due to space-charge, contact potentials, and background ions, specifically $H_2{}^+$. The absolute shape and intensity scale of the ion yield curves produced with linearly polarized (LP) light reproduce the results of Fittinghoff *et al.* [3] under similar laser pulse conditions. The essential thing to note is that the shape (knee) and polarization dependence (suppression) are qualitatively identical to the xenon case but occur at two orders of magnitude higher intensity. The same polarization dependence has been observed by Fittinghoff and Bolton [8]. Furthermore, the sequential portion of the He^{2+} curve agrees well with the Ammosov-Delone-Krainov (ADK) [7] tunnel rate. Thus, contrary to xenon at 0.5 μm, helium ionizes nonperturbatively and is consistent with a quasistatic description. Although both experiments provide the same signature in the ion yields, the ionization regimes dictate that the underlying physical mechanisms *must be different.*

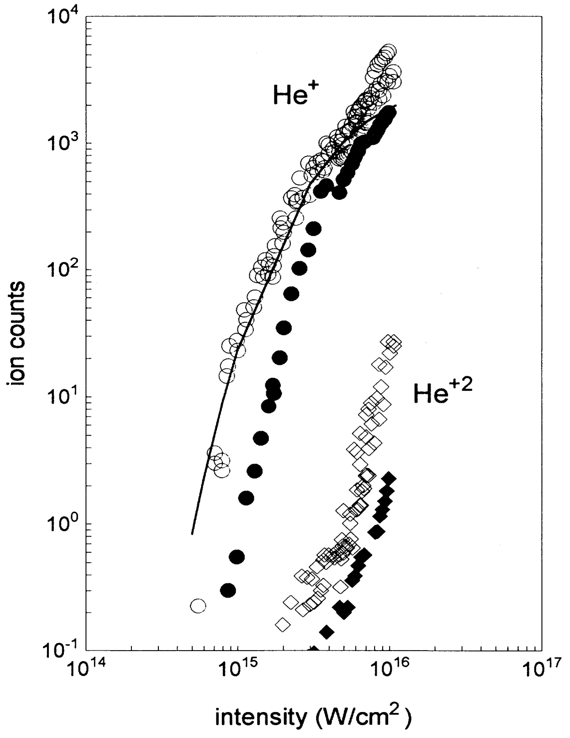

FIG. 3. Intensity dependence of the ion yield curves for He^+ and He^{2+} resulting from 617-nm, 100-fs excitation of neutral helium. The open circles and diamonds are taken with LP light and the closed circles and diamonds with CP light.

Recently, two *direct* mechanisms have been proposed [3, 9] to explain the nonsequential production of doubly ionized helium by 614-nm, 120-fs LP pulses [3]. The first of these two processes is termed shake-off (SO) by Fittinghoff *et al.* [3]. This model assumes that the ionization of the first or outer electron is impulsive (sudden approximation) and occurs on such a rapid time scale (less than half an optical cycle) that the inner electron cannot *instantaneously* readjust to the new ionic potential and consequently has some probability to ionize (tunnel). A simple uncertainty principle argues that the short time associated with the ionization of the outer electron results in a large enough bandwidth to ionize the inner electron. Corkum [9] has recently analyzed the same helium data [3] using a quasistatic (QS) model [10] of two-electron ejection. In this scenario, electrons excited in the continuum and accelerated by the field have some probability of returning to the vicinity of the core in the next half optical cycle. If the energy of the electron as it passes the core exceeds the e-$2e$ scattering energy, then two-electron ejection can occur. Thus, the essence of this model lies within the electron's initial conditions and the e-$2e$ collisional cross section. Both models differ from the standard Wannier [11] description for two-electron escape since there is no need to invoke electron-electron correlations. Furthermore, both share a common need for large ionization rates, viz., high intensity (neither model applies to the xenon case), in order to validate their ansatz, but differ significantly in physical dynamics. In particular, the shake-off model should be rather sensitive to the pulse duration, while for the quasistatic model the polarization should be the critical parameter.

For xenon, all the spectroscopic evidence validates a perturbative ionization regime. Furthermore, under the conditions of the experiment, the ponderomotive energy, which provides an estimate of the ac-Stark shifts, is at most 0.5 eV, which is too small to support the QS model. Likewise, the long ionization time scale cannot justify a sudden approximation (SO model). These arguments are reinforced by the coincidence measurements, which show *no evidence* for a direct process. What emerges from the measurements is a picture for ionization of xenon at 0.5 μm that *is not direct two-electron ejection* but instead a *higher-order sequential process* involving final-state resonances, i.e., autoionizing. In this *sequential* scenario the increased rate giving rise to the knee structure is due to a continuum two-electron resonance that autoionizes, followed by ionization of the excited ion. The polarization dependence implies that the continuum state must have relatively low angular momentum, which suppresses the resonance in circular polarization by simple selection-rule arguments. One may speculate that its energy is around ten photons above the xenon ground state on the basis of the multiphoton rates and the structure of the Xe I and Xe II spectra. Furthermore, such a scenario requires no unusually large cross sections or inconsistencies with LOPT to explain the existence of the knee structure. However, more investigations are necessary to assign it.

The situation is rather different for helium. Electron energy spectra [5, 12] and ion rates clearly indicate that the ionization regime is very nonperturbative and consistent with a tunneling (or over-the-barrier) description. Thus, the xenon scenario, which invokes resonant states, becomes physically unreasonable, while the direct models become more meaningful. In such a regime, the appearance intensity can be easily predicted by using the ADK rate [7]. In fact, the ADK agreement with the He^+ experimental curve is good, but it severely overestimates the experimentally measured He^{2+} appearance intensity by a factor of about 2.5 (see Fig. 3).

In summary, double ionization of xenon and helium shows the same behavior versus intensity or polarization in spite of occurring in quite different ionization regimes. This similarity seems fortuitous, and care must be exercized in the identification of such features in other atoms. For xenon the behavior results from one-electron *sequential* ionization involving continuum resonances (which remains to be identified), but for helium this is ruled out. In fact, *direct* two-electron ionization appears to be the most logical scenario for helium. Both the SO and QS model predictions are compatible with the observations, and more tests are needed to determine the correct one. Finally, another recent investigation of the double ionization of helium by 1053-nm, 1.5-ps pulses [13] did *not* show the knee structure. However, this negative result may be explainable either by the increased pulse duration in the SO model or by a slightly elliptical polarization in the QS model. EE coincidence measurements should provide a crucial test of these models.

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