BRIEF REPORTS

Brief Reports are accounts of completed research which do not warrant regular articles or the priority handling given to Rapid Communications; however, the same standards of scientific quality apply. (Addenda are included in Brief Reports.) A Brief Report may be no longer than 4 printed pages and must be accompanied by an abstract. The same publication schedule as for regular articles is followed, and page proofs are sent to authors.

Polarization dependence of tunneling ionization of helium and neon by 120-fs pulses at 614 nm

D. N. Fittinghoff, P. R. Bolton, B. Chang, and K. C. Kulander
Lawrence Livermore National Laboratory, Livermore, California 94550
(Received 24 May 1993)

We report on the polarization dependence of the ionization of helium and neon by 120-fs pulses at 614 nm. For linearly polarized pulses the data for He^{2+} and Ne^{2+} show enhancements over sequential tunneling. For circularly polarized pulses the data show no distinct enhancements. Dynamic resonances, which produce similar enhancements in the multiphoton regime, do not explain the data in the tunneling regime. Two direct-ionization models are considered: a shake-off model, and a semiclassical model for rescattering of an electron by the ion core. The exhibited polarization dependence of the ion yield enhancement is consistent with the semiclassical rescattering mechanism. Definitive isolation of a direct, nonsequential ionization mechanism in the optical tunneling regime requires continued scaling experiments on the laser wavelength, pulse width and ellipticity, and the target species.

PACS number(s): 32.90.+a, 32.80.Rm, 42.50.Hz

Earlier [1], we observed nonsequential double electron ejection in helium ion yields for 120-fs, linearly polarized 614-nm laser pulses. The nonsequential ionization appears as a feature in the He^{2+} yield that saturates in the same intensity interval as the He^{+} yield and is an observation of direct double ionization in the optical tunneling regime. In this nonperturbative regime, a few neutral atoms survive to the intensities that produce double ionization; to explain the feature, about 1% of those surviving neutral atoms must directly ionize to the doubly charged state. Here we present results for linear polarization in neon and circular polarization in helium and neon.

Reference [1] describes the experimental setup for the linear polarization measurements in detail. Briefly, a colliding-pulse mode-locked dye laser operates at 614 nm and produces peak energies of 2 mJ per pulse. We focus the 120-fs pulses into a low-density collisionless gas target. In the 4.5-μm full-width-at-half-maximum focal spot, the peak intensities reach 10^{16} W/cm^2, and these high laser fields (of the order of an atomic unit) ionize the neutrals. A time-of-flight mass spectrometer then collects and counts the ions. The 1-m spectrometer has enough resolution to distinguish He^{2+} from singly ionized H_2. For the circular polarization measurements, we insert a one-quarter wave plate into the beam. Optical measurements show that the circularly polarized beam has greater than 0.95 ellipticity. To ensure that the data for different charge states are consistent, we acquire all the data for each gas during successive data runs.

Figure 1 shows the measured He^{+}, He^{2+}, Ne^{+}, and Ne^{2+} ion yields versus laser intensity for linear polarization. Also plotted are the ion yields predicted for sequential ionization by linearly polarized light using the theory of Perelomov, Popov, and Terent'ev with the coefficients of Ammosov, Delone, and Krainov (ADK) [2,3]. For comparison with theory, we divide all measured intensities in this paper by 1.25. This factor is within the absolute uncertainty in the measured intensity. We note that we have made no relative shifts between the various

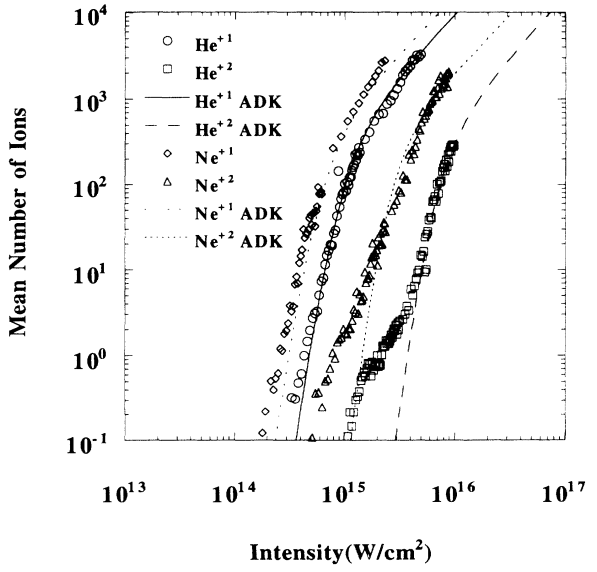

FIG. 1. Helium and neon ion yields for linear polarization and the sequential ADK theory. The intensities for the data are divided by 1.25 for comparison with theory.

charge states or gas species. Each data curve represents 10 000 to 14 000 data shots averaged in bins. The data reproduce the enhancement in He^{2+} ionization observed before. Furthermore, for intensities above 5×10^{15} W/cm^2, the experiment confirms the previously assumed continuation of the He^{2+} curve along the predicted sequential ionization curve. This confirms our earlier assertion that the yield exhibits a new nonperturbative physical process in addition to sequential tunneling and is not a refutation of tunneling ionization theory or an experimental error in the relative intensity between charge states. The new data for Ne^{2+} also exhibit enhanced ionization. As in the case of helium, the yield appears at an intensity lower than the ADK appearance intensity. The yield then rises and saturates at the same intensity as Ne^+. Eventually, again as for helium, the yield rises to follow the sequential ADK prediction for Ne^{2+} yield. The ADK appearance intensities for the singly charged ions of each should exhibit a largely cubic dependence in the ionization potential [4]. The observed ratio of the He^+ and Ne^+ appearance intensities in 3/2 and agrees with this closed-form prediction.

Figure 2 shows the measured He^+ , He^{2+}, Ne^+, and Ne^{2+} ion yields versus laser intensity for circular polarization. The figure also shows the ion yields predicted for circular polarization by sequential ADK theory. The enhanced ion yield features observed in He^{2+} and Ne^{2+} for linear polarization now disappear below the resolution of the experiment. The expected experimental error in relative intensity between the charge states is 25%. Thus we may not interpret the visible deviation from the spacing of the charge states given by the ADK model as an enhancement. The important point, however, is that circular polarization suppresses the enhancement. Also, as in the linear case, the observed ratio of the He^+ and Ne^+ appearance intensities agrees with the ADK prediction.

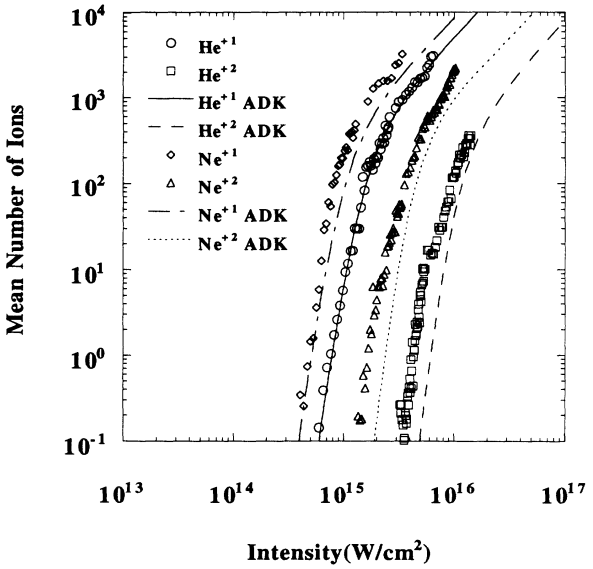

FIG. 2. Helium and neon ion yields for circular polarization and the sequential ADK theory. The intensities for the data are divided by 1.25 for comparison with theory.

We note that for He^+ and Ne^+ the appearance intensities for circular polarization are approximately twice the appearance intensities for linear polarization. Tunneling with circular polarization differs from tunneling with linear polarization in two ways. First, for a given laser intensity, the peak electric field for circular polarization is smaller by the square root of 2. This reduces the barrier suppression that allows the tunneling. Second, during the optical cycle the circular rate is constant because the field amplitude is constant. The linear rate, however, varies with the instantaneous field amplitude. For sequential ionization these differences result in a higher appearance intensity for circular polarization.

Resonantly enhanced ionization rates do not explain our observations. In resonantly enhanced ionization, the ionization rate increases when the energy of an allowed atomic transition nearly equals the energy of an integral number of photons. Changing to circular polarization would significantly decrease any resonance effects—in agreement with the experiments—because of the decreased number of allowed electronic transitions. Since dynamic resonances can occur as the atomic energy levels shift in response to the applied field, resonances do become likely. In addition, experiments in the multiphoton regime show similarly enhanced xenon ion yields for longer pulse widths [5,6]. Resonances, however, are unlikely to influence the results of tunneling experiments that are nonperturbative. Direct solution of the Schrödinger equation for a single active electron (SAE) for helium shows no evidence of resonant enhancements [1]. The SAE calculation would miss two-electron effects. But enhancements due to double excitations are unlikely on energetic grounds. The double excited states of helium are 60 eV above the ground state of the atom—almost 40 eV above the first ionization potential. Thus double ejection in the tunneling regime is unlikely to be due to resonance states.

We explain our data in terms of direct or nonsequential ionization. Ionization of the outer two electrons in a multielectron atom can be represented by the following coupled set of rate equations:

$$\frac{dn_0}{dt} = -k_0 n_0 ,$$

$$\frac{dn_1}{dt} = k_0' n_0 - k_1 n_1 ,$$

$$\frac{dn_2}{dt} = (k_0 - k_0') n_0 + k_1 n_1 .$$

Here n_j and k_j are the population- and intensity-dependent total ionization rates for states of charge j. If the ionization process is purely sequential, k_0' will exactly equal k_0. If there is any process that transfers population directly from the neutral to the doubly charged ion, then $k_0' < k_0$. Examination of the rate equations produces two general conclusions. First, the appearance intensity for the second ion will be lower than that predicted by a purely sequential model. Second, any direct component will volume saturate in parallel with the single-ion signal as the laser intensity increases. Our data agree with these

conclusions. Details of the ionization processes determine the values of the intensity-dependent ionization-rate coefficients. A nonperturbative direct nonsequential component therefore determines the scaling of observed ion yield enhancements with gas species, laser wavelength, ellipticity, and pulse width. Similarly, the character of the photoelectron spectrum is uniquely determined in the direct-ionization regime.

In our earlier paper [1], we proposed a mechanism similar to the shake-off process observed in single-photon double ionization of helium [7]. In this process the first electron absorbs a photon with enough energy to remove both electrons and rapidly leaves the vicinity of the nucleus, causing the second electron to be "shaken up" into the continuum some fraction of the time. For photon energies from 80 to 120 eV, the shake-up probability is a few percent of the single ionization probability. For the multiple-photon case and photon energies of 2 eV, we postulated that this process might also occur in the tunneling regime if the rate at which the first electron leaves the atom becomes large enough. The direct component would be measurable only if the pulse rise time were short enough; neutral population must still exist within the focal volume. The process would then exhibit an intensity and rate threshold in single ionization such that above this critical intensity I_c and rate R_c the process becomes possible. Shake-off would occur within much less than a quarter of an optical cycle. Using these assumptions we were able to fit the observed double ionization yields. In fitting our helium data, we find that the probability is comparable to that in the single-photon case, approximately 0.5%, and $I_c = 10^{15}$ W/cm^2 for helium. The corresponding critical rate is $R_c = 1.2 \times 10^{14}$ sec^{-1}. The shake-off process should be strongly dependent on the pulse width of the laser and independent of its wavelength and ellipticity.

Figure 3 compares the linear data with a shake-off prediction for helium and neon. For the prediction, we use the linear ADK rates and evaluate the direct component using $\alpha \equiv k_0'/k_0 = 0.995$ when $k_0 \geq R_c$ and 1 otherwise. For neon the critical instantaneous rate (intensity) for the onset of a direct ionization component is 4×10^{13} sec^{-1} (5×10^{14} W/cm^2).

Figure 4 similarly compares the calculation with the data for circular polarization. In the rate scaling, we use the circular ADK ionization rates and, for each gas, the same critical instantaneous rate R_c determined in the linear case. For circular polarization the corresponding critical intensities for helium and neon are 2×10^{15} and 1×10^{15} W/cm^2, respectively.

These figures reveal two features. First, as expected for equal critical rates, the circular polarization critical intensity is twice that for the linear case. Secondly, while the circular predictions show a smaller yet persistent enhancement of the doubly charged ion yields, the circular data suggest an extinction of this effect. The linear case by contrast is more predictive.

In contrast, Corkum and co-workers [8,9] have proposed an alternative model for direct, nonsequential ionization that predicts a strong laser ellipticity and wavelength dependence. The model is presented in a unified way as also describing harmonic generation yields and above threshold ionization (ATI) spectra. With respect to direct ionization it is a three-step semiclassical process. First, the field of the laser suppresses the Coulomb barrier and liberates an electron by optical tunneling. Second, the liberated electron interacts with the laser field in the continuum. This produces an approximately 50% probability of accelerating the electron back toward the parent ion within a minimum time integral of one-half period. Here the laser Coulomb fields are additive. Third, this electron removes a second electron from the

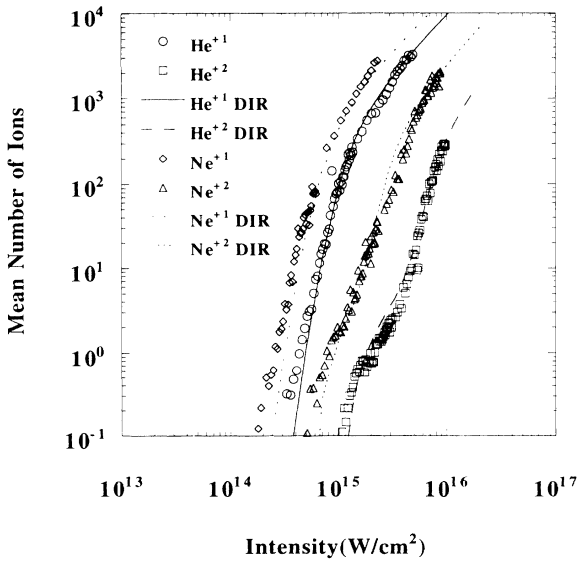

FIG. 3. Helium and neon ion yields for linear polarization and the ADK theory including a direct path for ionization (DIR). The intensities for the data are divided by 1.25 for comparison with theory.

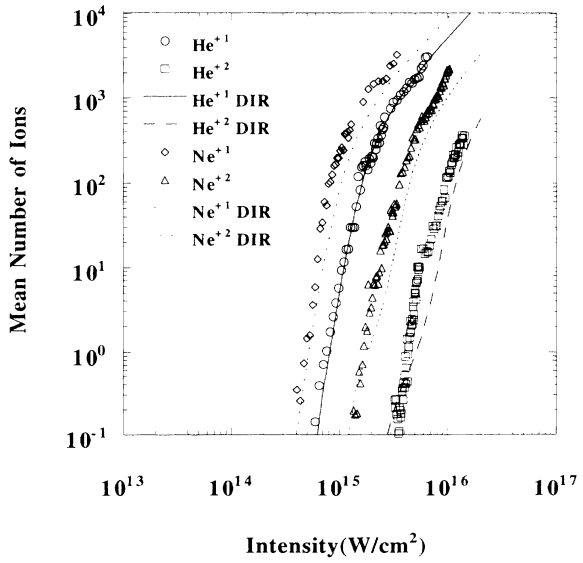

FIG. 4. Helium and neon ion yields for circular polarization and the ADK theory including a direct path for ionization (DIR). The intensities for the data are divided by 1.25 for comparison with theory.

ion by inelastic, e-$2e$ scattering. The scattering probability is determined by the energy-dependent cross section. The maximum electron kinetic energy during the reencounter with the singly charged ion is $3.17I_p$, where I_p is the laser pondermotive energy ($I_p = e^2E_l^2/4m\omega^2$). This energy can exceed 100 eV at 10^{15} W/cm^2 using 614-nm light.

The wavelength dependence of this model is mainly due to the quadratic scaling of the pondermotive energy with λ. The polarization dependence stems from the possible transverse component of an electron trajectory induced by a perpendicular component of the laser field. In the circular case the amplitudes of the two perpendicular field orientations are equal, and the initially liberated electron can be steered away from the ion after tunneling. Quantum mechanically, the transverse spread of the electron wave packet results in an effectively larger impact parameter. This reduces scattering and thus suppresses nonsequential ionization.

The Corkum model has been shown to fit our linear data and to predict extinction of a direct nonsequential ionization component for circular polarization. We have also recently been informed that this strong ellipticity dependence has been confirmed by Walker *et al.* [10] and Kondo *et al.* [11] in helium and by Dietrich *et al.* [12] in neon. For linear polarization Kondo *et al.* have also shown that the nonsequential ionization feature seen in ^{4}He using 745-nm light is not observed in ^{3}He at 248 nm. Although the pulse width at 248 nm was 440 fs they attribute this result to the quadratic reduction of the "reencounter" energy (relevant to the e-$2e$ scattering) below the 54-eV cross-section threshold in helium.

Although our data and these other recent observations are consistent with Corkum's semiclassical description, the absence of the direct ionization feature in ^{3}He using 1.5-ps pulses at 1053 nm [13] is still a puzzle in that it appears consistent with a shake-off picture. Furthermore, both proposed processes suggest that such ionization is likely to occur with several target species.

In summary, we have identified in helium and neon a nonperturbative direct nonsequential ionization process that exhibits a strong ellipticity dependence in contrast to sequential tunneling ionization. It is also clear that continued experiments that examine dependence of this phenomenon on ellipticity, pulse width, wavelength, and target species are necessary to isolate definitively the nonsequential mechanism. Our current examination of ion yields at 800 nm with variable ellipticity will be the subject of a subsequent report.

We thank L. DiMauro and P. Corkum for many useful communications. This work was performed under the auspices of the U.S. Department of Energy at Lawrence Livermore National Laboratory under Contract No. W-7405-Eng-48.

[1] D. N. Fittinghoff, P. R. Bolton, B. Chang, and K. C. Kulander, Phys. Rev. Lett. **62**, 2642 (1992).
[2] M. V. Ammosov, N. B. Delone, and V. P. Krainov, Zh. Eksp. Teor. Fiz. **91**, 2008 (1986) [Sov. Phys. JETP **64**, 1191 (1986)].
[3] A. M. Perelomov, V. S. Popov, and M. V. Terent'ev, Zh. Eksp. Teor. Fiz. **50**, 1393 (1966) [Sov. Phys. JETP **23**, 924 (1966)].
[4] B. Chang, P. R. Bolton, and D. N. Fittinghoff, Phys. Rev. A **47**, 4193 (1993).
[5] A. l'Huillier, L. A. Lompré, G. Mainfray, and C. Manus, J. Phys. B **16**, 1363 (1983).
[6] A. l'Huillier, L. A. Lompré, G. Mainfray, and C. Manus, Phys. Rev. A **27**, 2503 (1983).
[7] R. Wehlitz, F. Heiser, O. Hemmers, B. Langer, A. Menzel, and B. Becker, Phys. Rev. Lett. **67**, 3764 (1991).
[8] P. B. Corkum, Phys. Rev. Lett **71**, 1994 (1993).
[9] K. J. Schafer, B. Yang, L. F. DiMauro, and K. C. Kulander, Phys. Rev. Lett. **70**, 1599 (1993).
[10] B. Walker, E. Mevel, B. Yang, P. Breger, J. P. Chambaret, A. Antonetti, L. F. DiMauro, and P. Agostini, Phys. Rev. A **48**, 894 (1993).
[11] K. Kondo, A. Sagisaka, T. Tamida, Y. Nabekawa, and S. Watanabe, Phys. Rev. A **48**, 2531 (1993).
[12] P. Dietrich, N. H. Burnett, M. Ivanov, and P. B. Corkum (unpublished).
[13] J. Peatross, B. Buerke, and D. Meyerhofer, Phys. Rev. A **47**, 1517 (1993).