Precision Measurement of Strong Field Double Ionization of Helium

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The production of He^+ and He^{2+} by a 160 fs, 780 nm laser has been measured over an unprecedented 12 orders of magnitude in counting range. Enhanced double electron emission, called nonsequential (NS) ionization, was observed over an intensity range where the single ionization dynamics is evolving from multiphoton to pure tunneling. The NS yield is found to scale with the ac-tunneling rate for the neutral, even when tunneling is not the dominant ionization pathway. A rescattering mechanism fails to predict the observed NS threshold or magnitude.

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The multiphoton ionization (MPI) of noble gases by strong laser fields can be well described by time-dependent quantum mechanical calculations within the single active electron (SAE) approximation [1,2]. The approximation's success derives from the fact that for long laser wavelengths the ionization dynamics are dominated by single-electron excitations, leading to the *sequential* production of ion charge states. In recent experiments, however, enhanced production of doubly charged ions in both He and Ne [3–5] has been observed with yields that defy a kinetic description based upon sequential rates (see Fig. 1). Given the high intensities and small photon energies involved, it has been argued [3,6] that this enhancement must result from a direct, *nonsequential* (NS) ionization process rather than a resonant process [4]. Unfortunately the current quantitative knowledge of strong field multielectron ionization is so limited that even the basic mechanism for NS double ejection has not been established. In this Letter, a high precision measurement of NS ionization yield in He is reported which extends the sensitivity of the previous experiments by a factor of 10^6. We are able to observe the NS production of He^{2+} over a large enough intensity range to directly test current two-electron ionization models.

We find that NS double ionization is clearly linked to the dynamics of emission of the first electron. Within the intensity range over which the NS process has been observed, the ionization of He evolves from being predominantly multiphoton in character to being purely tunneling. This evolution in the ionization dynamics is corroborated by changes in the photoelectron energy distributions. Most importantly, analysis of the intensity dependence of the NS yield indicates that the two-electron ejection scales with the tunneling component of neutral helium, even when multiphoton ionization dominates.

Two simple mechanisms which invoke features of a two-step, quasistatic picture of strong field ionization have been proposed to explain NS ionization [3,6]. In the first step an electron is released near the peak of the oscillating field amplitude. The electron either passes over or tunnels through the effective barrier created by the Coulombic attraction of the ion core and the laser's instantaneous electric field. At that point (second step), the electron's evolution is dominated by its interaction with the laser field since its motion is essentially removed from the

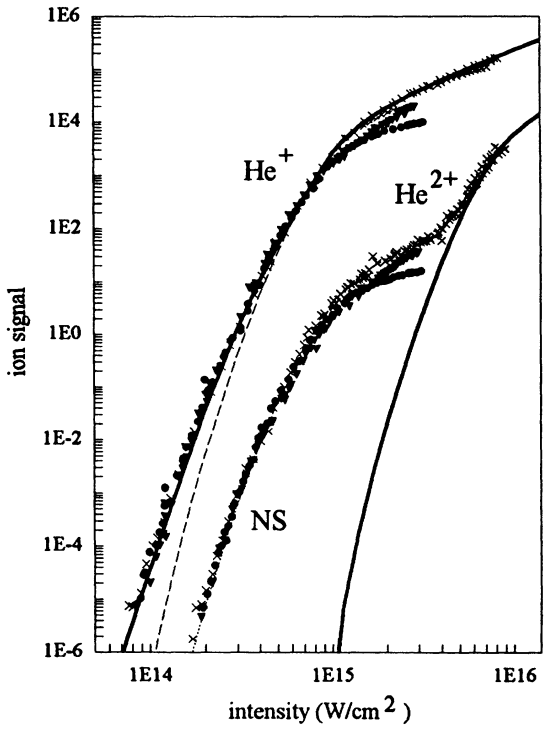

FIG. 1. Measured He ion yields for linear polarized, 100 fsec, 780 nm light. Calculations are shown as solid (SAE) and dashed (ac-tunneling) lines. The measured intensities are multiplied by 1.15. The solid curve on right is the calculated sequential He^{2+} yield.

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influence of the core. The field first accelerates the freed electron away from the ion, then, as the field changes sign during the next half cycle, the electron, if it is not moving too fast, is pushed back toward the core.

The two proposed NS mechanisms differ on when and how the second electron escapes. Corkum [6] has proposed that when the tunnel-ionized electron revisits the core it can collisionally ionize (e-$2e$) the second electron (rescattering). Thus at the time of rescattering the first electron must have enough energy ($\geq$ 54.4 eV) to free the second electron. Since the maximum energy that can be gained from the field is proportional to the intensity [2,6], an abrupt threshold in the NS yield is expected at low intensity. Fittinghoff *et al.* [3] suggested that the second electron might ionize due to the rearrangement caused by the sudden loss of screening of the bare nucleus by the rapid removal of the first electron (shakeoff). This process is responsible for double ionization by a single high energy photon [7]. Thus, the essential difference is that in shakeoff both electrons become unbound simultaneously, while the rescattering mechanism requires a delay (at least a half optical cycle) between the electrons' escape times. Despite the obvious mechanistic differences, both NS models have produced reasonable agreement with previous experiments [3–5]. Our experiments address this ambiguity by providing sufficient sensitivity to test the model dependent observables.

In our experiment 160 $\pm$ 30 fsec, 780 nm light pulses are focused by a 10 cm focal length doublet lens into an ultrahigh vacuum chamber (10^{-10} torr). The maximum pulse energy at a 1 kHz repetition rate is 1.3 mJ with typical pulse to pulse fluctuations $< 1.7\%$ for 10^6 laser shots. The laser polarization is measured to have an extinction ratio > 100 over the entire intensity range. The laser intensity is calibrated by measuring the short pulse resonant photoelectron spectrum of xenon as a function of pulse energy and concurrently recording the xenon and helium ion yields. The calibration is corroborated by a spot size measurement and accurate to approximately 30%. The sample gas is 99.999% helium which is further scrubbed to < 0.1 ppm for O_2, H_2, H_2O, CO_2, and hydrocarbon impurities. The electron energies are analyzed using time-of-flight (TOF) spectrometers. A 30 cm TOF mass spectrometer provides sufficient resolution ($m/\delta m > 300$) to easily separate H_2^+ and He^{2+} mass peaks. Data collection uses 1 ns binning of discriminated electron and ion events operating at low event probabilities ($\leq$ 0.25/shot). The e/m detection sensitivity is estimated to be 1:1.25 (He^+ : He^{2+}) based on saturated yields.

Figure 1 shows the helium ion yield curves for 160 fsec, 780 nm excitation. Each data point contains $\geq$60 000 laser shots. Results from five separate scans with three different spot sizes are plotted. The He^+ yield increases nonperturbatively up to a measured saturation intensity of 8×10^{14} W/cm^2. Beyond this point the yield increases as $I^{3/2}$ consistent with an expanding Gaussian focal volume. The He^{2+} curve shows the characteristics of two rate kinetics; NS production at low intensities [$(1.5 - 8) \times 10^{14}$ W/cm^2], a saturated regime [$(0.8 - 3) \times 10^{15}$ W/cm^2], sequential production above 3 PW/cm^2 ($He^+ \rightarrow He^{2+}$) and saturation at 8 PW/cm^2. Note that the He^+ and NS He^{2+} yields follow each other over ten orders of magnitude in signal, saturating simultaneously. To firmly establish this fact we varied the confocal spot size relative to ion spectrometer's detection image using different f-number optics. As the spot size exceeds the detection image a reduction from the $I^{3/2}$ scaling is observed above saturation. This purely spatial effect clearly demonstrates that the NS He^{2+} production is *exactly linked* to the saturation of the He^+ yield; the singly and doubly charged ion production are connected with the depletion of neutral helium atoms.

The He^+ yields calculated using the SAE approximation [1] (solid line) and ac-tunneling (ADK) rates [8] (dashed line) also are shown in Fig. 1. SAE provides the total He ionization rate, including both the multiphoton and tunneling pathways. Both calculations agree with the data at high intensity but the ADK curve falls below the measured yield at low intensity. By contrast, the SAE results are in agreement over the full dynamic range of the experiment showing the multiphoton contribution becoming increasingly important below 0.5 PW/cm^2.

Further evidence for the evolution from multiphoton to tunneling can be seen in the photoelectron spectra (PES) of Fig. 2. Figure 2(a) shows that at low intensity

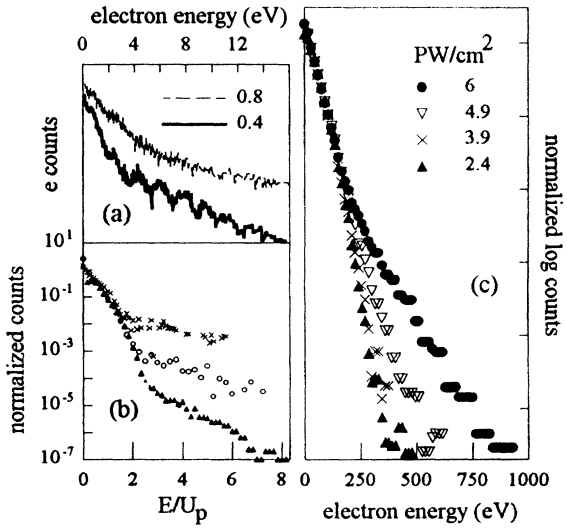

FIG. 2. Helium photoelectron spectra for 780 nm excitation. Low energy electrons plotted in (a) for 0.4 and 0.8 PW/cm^2. Scaled energy, E/U_p, plot at (b) 0.2 (crosses), 0.4 (circles), and 1.0 (triangles) PW/cm^2. The value of U_p is defined by the laser's peak intensity. Plot (c) shows the high energy electron tail (solid circles) resulting from the sequential formation of He^{2+}.

the spectrum [9] has a clear above-threshold ionization structure, indicative of MPI, which disappears at higher intensity. In the experiment, spatial averaging washes out the "single" atom ATI structure in the tunneling regime due to the large ponderomotive shift of the ionization potential and the relatively weak dependence of the ionization rate. In Fig. 2(b) the PES are plotted using an electron energy scaled by the ponderomotive energy [10]. At different intensities, the overall shape of the electron distributions between zero and $2U_p$ [10] is consistent with the classical prediction [11]. More than 99% of the electrons emitted are within this energy range while the fraction with energies $> 2U_p$ decrease at a rate $\propto I^{-2.6}$. Our theoretical analysis shows that over this intensity range, the fraction of tunneling in the total SAE rate increases from 0.05 to 1 with increasing intensity. Therefore the data suggest that tunneling ionization results in fewer electrons with energies $> 2U_p$.

A sensitive measure of the NS dynamics is provided by plotting the intensity dependence of the He^{2+}/He^{+} ratio, shown in Fig. 3. To ensure accuracy, the two ions were concurrently collected at a fixed intensity and averaged for at least 10^6 laser shots. Although the ion curves in Fig. 1 show a strong intensity dependence varying by 7 orders of magnitude between 0.15 and 5.0 PW/cm^2, the ratio exhibits a gentle slope of only $I^{1.3}$. The ratio is constant (0.0020 [3]) from about 5 PW/cm^2 until the sequential production of He^{2+} becomes significant. The onset of sequential He^{2+} production is corroborated in Fig. 2(c) by the unambiguous appearance of a high energy electron tail beyond 3 PW/cm^2. Assuming that the NS rate is given by the pure ac-tunneling rate times a constant, which is defined by the measured ratio of He^{2+}/He^{+} at saturation, the dotted NS yield curve in Fig. 1 results. Furthermore the ratio of this curve to the SAE He^{+} yield curve produces the solid line in Fig. 3. The striking agreement with the data implies that tunneling is responsible for NS double ionization. We emphasize two differences

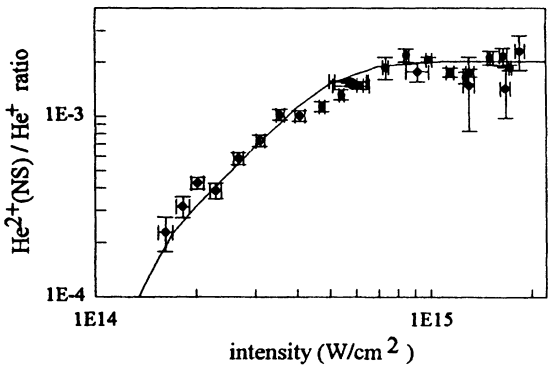

FIG. 3. Intensity dependence of He^{2+} (NS)$/He^{+}$ ratio. Error bars indicate 1 standard deviation. Solid line is calculated; see text for details. The measured intensities are multiplied by 1.15.

between tunneling and MPI. First, in tunneling, electrons are emitted in bursts near the maxima in the oscillating electric field while the multiphoton excitation is constant throughout the optical cycle. Second, the multiphoton ionized electrons appear in the continuum near the nucleus whereas tunneling electrons originate at the outer turning point of the instantaneous potential barrier, $(6-10)a_0$ from the nucleus. These differences mean that the dynamics of the electrons, after reaching the continuum by these separate pathways, can be significantly different.

The above discussion establishes the importance of the tunneling in the NS production of He^{2+} but does not define a mechanism. In the rescattering model [6] the electron revisits the core after a free propagation period of at least half an optical cycle with a maximum kinetic energy of $3.2U_p$. To knock off the second electron, the return energy must exceed the He^{+} binding energy, that is, $3.2U_p \geq 54.4$ eV, resulting in a threshold for NS production at 0.3 PW/cm^2. Allowing for the possibilities that NS ionization would result from collisional excitation of the second electron to excited states (41 eV) which then rapidly field ionize and that the core potential is suppressed by the field at the rescattering time, this threshold is reduced to 0.23 PW/cm^2. *No such threshold is observed in the experiment* which extends down to 0.14 PW/cm^2. Also as the intensity approaches threshold, the NS rate should drop steeply, not with the $I^{1.3}$ NS scaling extracted from the ratio data of Fig. 3. Thus, the rescattering mechanism does not predict the observed low intensity behavior of the He^{2+} yield.

To estimate the importance of rescattering at higher intensities we must consider the evolution of the tunneling wave packets. Before rescattering, the wave packet propagates outside the influence of the ion core for most of the half cycle so its transverse dimension should be comparable to that of a freely spreading Gaussian packet. Thus the width is given in atomic units by $\alpha_t = \sqrt{\alpha_0^2 + (2t/\alpha_0)^2}$, where α_0 is the width at $t = 0$. At time t, α_t can be no smaller than $\sqrt{4t}$ corresponding to an initial width of $\alpha_0 = \sqrt{2t}$. This minimum width after 1/2 cycle at our wavelength is $14.5a_0$, but only for an unphysically broad initial distribution of $10.4a_0$. For a more realistic α_0 of $(3-4)a_0$, α_t is approximately $30a_0$. An analysis of our calculated tunneling wave packet recrossing the core also gives a width of $30a_0$. This is an order of magnitude greater than that inferred by Corkum [6] in fitting earlier data. Knowing α_t, we can place an upper bound on NS production by rescattering assuming the slowly varying field does not appreciably alter the inelastic scattering cross sections. The He^{+} ionization cross section [12] peaks at 5×10^{-18} cm^2 at 130 eV. The excitation cross section to the $n = 2$ states is comparable to this value at threshold (41 eV) and gradually decreases with increasing impact energy while higher excited states have much smaller cross sections [13]. Therefore, taking

the total excitation/ionization cross section to be 5×10^{-18} cm^2 means only electrons with impact parameters less than $0.26a_0$ ($b_{\max} = \sqrt{\sigma_{\max}/\pi}$) will be effective in freeing the second electron. Using a $30a_0$ wave packet width at the first return and assuming all electrons return and have an energy ≥ 41 eV, we obtain an upper limit for the ratio of 1.5×10^{-4}, more than an order of magnitude smaller than the experimental ratio. Clearly, NS ionization due to rescattering is much smaller than our measured yield over the entire dynamic range of the experiment.

The shake-off mechanism [3] focuses on the behavior of the second electron at the instant the first electron enters the continuum. This *simultaneous* two-electron ejection cannot be tested as thoroughly as rescattering without a realistic two-electron calculation but qualitative comparison is possible. As the first electron tunnels free, the second electron is assumed to be shaken up (excited) or off (ionization) with an intensity independent probability. Assuming that MPI results in the gradually removal of electrons from the vicinity of the nucleus, either due to resonant ionization or just because they are promoted into the continuum much nearer the nucleus, then the abruptness required by the shakeoff mechanism is lost. This would explain why only the tunneling component is effective in NS double ionization. Further experimental evidence shows that the NS yield drops significantly as the polarization of the incident laser has some ellipticity [4,14]. In elliptically or circularly polarized fields electrons obtain a substantial amount of angular momentum which could inhibit the transfer of energy to the second electron in the shake-off process, consistent with the observed polarization dependence. We note that the rescattering mechanism can explain an ellipticity dependence since fewer *e*-2*e* collisions are possible as the electrons are pushed towards large impact parameters by the field ellipticity [6]. However, considering the above arguments for transverse wave packet spread the polarization dependence would not be as strong as anticipated since the center of the wave packet could be quite far from the nucleus and still, because of its large width, rescatter with equal efficiency.

In conclusion, we find that the $He^{2+}(NS)/He^+$ ratio *scales with the tunneling fraction of the ionization*, achieving a saturated value of 0.0020. The absence of a threshold in the NS yield, the reduction of high energy electrons in the PES with increasing intensity, and simple physical arguments about the amount of transverse spreading that is possible after 1/2 of an optical cycle lead us to conclude that rescattering mechanism cannot explain the observed NS ionization. These spreading arguments also explain the reduction in high energy electrons and may account for the observed decrease in high-order harmonic efficiency as one approaches the pure tunneling regime. We propose that NS ionization occurs via a simultaneous two-electron ejection either through a shakeoff or threshold mechanism involving some form of electron correlation. Unfortunately, the underlying mechanism for this process remains uncertain and requires a theoretical treatment using appropriate two-electron wave functions. Further experimental insight may be obtained using coincidence techniques. However, we emphasize that the data we have presented provide much more stringent limits on any future proposed NS mechanisms.

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