Observation of Nonsequential Double Ionization of Helium with Optical Tunneling

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We have measured the ion yields for helium ionized by 120 fs, 614 nm laser pulses for intensities near 10^{16} W/cm^2. We have found that for these ultrashort pulses the He^{+2} data exhibit a feature which saturates in parallel with the He^+ signal indicating that the ionization may proceed nonsequentially. We propose a new mechanism, which can exist only in the tunneling regime, for such nonsequential ionization.

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In the study of strong optical field ionization of atoms and ions field effects which are synonymous with tunneling (including possible over-the-barrier effects) can be isolated from conventional multiphoton ionization by using intense, ultrashort pulses. New developments in laser technology have made such studies possible. Focused field amplitudes of 1 a.u. or more (1 a.u. $=5.14\times10^9$ V/cm) and pulse lengths on the order of 100 fs are becoming routinely available. A complete understanding of field ionization using intense ultrashort laser pulses is fundamental to the study of tunnel-ionized plasmas and to the determination of peak laser intensities. The well known Keldysh adiabaticity parameter [1] γ is often used to distinguish tunnel ionization from conventional multiphoton ionization. Tunneling is favored over the multiphoton ionization for the case

$$\gamma=(E_{\text{ion}}/2U_p)^{1/2}\ll 1\,, \tag{1}$$

where E_{ion} is the ionization potential of the atom or ion being ionized and U_p is the ponderomotive potential from the laser field. While Sauer *et al.* [2] correctly point out that the condition $\gamma<1$ cannot strictly be considered to define the tunneling regime, tunneling models have been shown to be generally predictive in this regime to within intensity uncertainties inherent in typical ultrashort pulse laser ionization experiments [3–5]. In the work reported here we have studied the ionization, single and double, of helium by an intense ultrashort laser pulse. In this experiment γ varies from 0.4 to 1.0 Based on the simultaneous saturation of the He^+ ion yield and a feature in the He^{+2} ion yield we have obtained experimental evidence for nonsequential ionization of helium.

We record the number of ions produced versus the peak laser intensity when we focus a short laser pulse into a very low density collisionless gas target. The laser system used is a colliding-pulse mode-locked dye laser which operates at 10 pulses per second producing 1 mJ pulses at 614 nm. In a vacuum chamber, an $f/5$ off-axis parabola focuses the beam in the source region of a time-of-flight mass spectrometer. We calculate the peak intensity from the energy, pulse width, and focal profile. The energy of the laser pulse is measured on every laser shot using the reflection from a beam splitter, and single shot autocorrelations agree with a 120 fs hyperbolic secant-squared temporal profile. Imaging the focus shows a 1.5 times diffraction limited central peak with a full width at half maximum of 4.5 μm. Surrounding the central peak is a large region of low intensity (greater than an order of magnitude down from the peak intensity) containing roughly 50% of the total energy of the beam. The peak intensity is obtained from a numerical integration of the beam profile. The uncertainty in the peak intensity is less than a factor of 2. Peak intensities near 5×10^{15} W/cm^2 corresponding to peak fields above 10^9 V/cm are achieved.

The background pressure in the vacuum chamber is below 10^{-6} Pa. We uniformly backfill the chamber with helium gas to a typical pressure of 10^{-3} Pa. A 600 V/cm extraction field applied across the interaction volume sweeps the ions produced by the photoionization into the 1 m ion time-of-flight mass spectrometer. The spectrometer separates the ions in time according to the square root of the ratio of the ion mass to charge. The ions are detected by microchannel plates at the end of the flight tube and this ion signal is recorded as a function of time using a digitizing oscilloscope. The very small difference in mass to charge ratios between He^{+2} and H_2^+ raises the question of hydrogen contamination; however, the

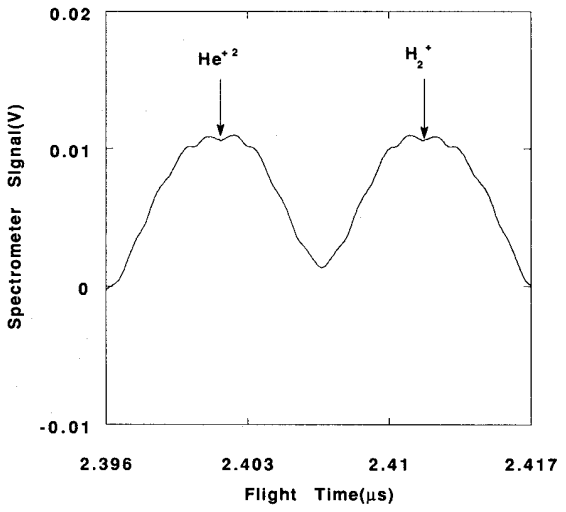

FIG. 1. Resolution of He^{+2} and H_2^+ ions.

spectrometer has a mass resolution sufficient to clearly resolve doubly ionized helium and singly ionized hydrogen molecules as shown in Fig. 1. Thus any hydrogen contamination is easily separated from the helium ionization data. This has been further verified by demonstrating independent pressure scaling of the He^{+2} and H_2^+ peaks. The signal peaks for each ion are integrated and calibrated by the efficiencies of the spectrometer to give the number of ions produced by the laser with an uncertainty near 50%.

Each data run represents 9000 to 11 000 laser shots for a given charge state with energy binning into about 100 intervals. Integrated data are then averaged. Measurements have been repeated several times and show no discernible shift in the relative positions of the He^+ and He^{+2} data. In Fig. 2 these ion yields are plotted versus peak laser intensity. For comparison with theory the data for both charge states has been uniformly shifted down in intensity by a factor of 1.25. This shift is within the experimental uncertainty in the intensity. The He^+ data stop at 2×10^{15} W/cm^2 in order to avoid saturation of the detection system.

Initially, to model our experimental data, we assume multiple ionization to be a "sequential" removal of electrons from atoms and ions with their unperturbed ionization potentials. Superimposed on the data in Fig. 2 are the results predicted by this sequential model which uses the cycle averaged rates Γ given by the tunneling theory of Perelomov, Popov, and Terent'ev [6] with the coefficients given by Ammosov, Delone, and Krainov (ADK) [7],

$$\Gamma=\lim_{\beta\to 1}\gamma_k\left(\frac{2}{\pi}\right)\left(-\frac{\partial}{\partial\beta}\right)^{2n-|m|-1}\frac{1}{\beta}\left(\frac{\beta}{F_k}\right)K_0\left(\frac{\beta}{F_k}\right). \tag{2}$$

Here $F_k=3F_0/2E_k^{3/2}$ where in atomic units F_0 is the laser field strength, E_k is twice the ionization potential of the kth charge state, γ_k is a numerical coefficient multiplying the field dependent terms, and K_0 is the zeroth order modified Bessel function. This is an absolute comparison of the number of ions produced with the intensity scale of the data shifted only by a factor of 1.25.

In implementing ionization models we approximate the beam profile by a Gaussian profile with the same peak intensity and full width at half maximum as the profile measured in the experiment. This approximation is good for ions produced at intensities which are within a factor of 10 of the peak value. The agreement between the sequential ADK model and the data for He^+ is excellent; however, for He^{+2}, the model overestimates the appearance intensity by a factor of about 3. At 5×10^{15} W/cm^2, the He^{+2} data join the sequential curve.

An earlier paper [8] has shown that ion yields for sequential ionization can be described in closed form by standard curves. The shape of the curves depends only on

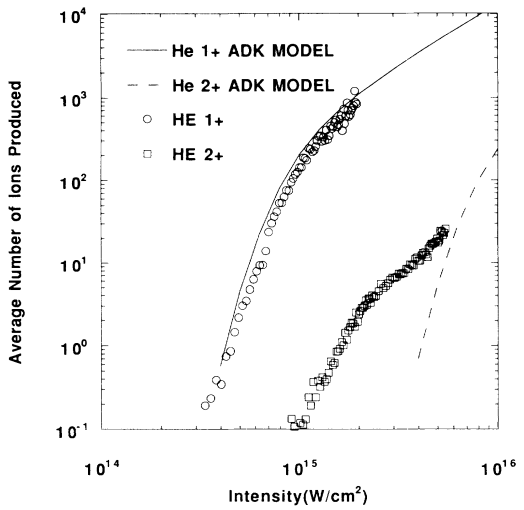

FIG. 2. Ion yield data compared to sequential ADK theory. The data for both charge states have been uniformly shifted down in intensity by a factor of 1.25 for comparison with theory.

ionization potential and laser parameters; however, the standard nature of the ion yield predictions is purely a consequence of the integrated rate equations for the sequential case. What remains are functions of a single variable, the normalized laser field scaled by the ionization potential. For sequential ionization, a charge state can be produced only after the previous charge state begins to volume saturate. In our data for He^{+2} the feature which saturates in parallel with the saturation of He^+ and then rises to join the predicted sequential curve cannot be explained by sequential ionization.

In order to explain the origin of the discrepancy between the He^{+2} data and the sequential prediction, a simple model is considered which includes a nonsequential component. Because the doubly excited states in He are almost 40 eV above the first ionization potential, we believe the enhancement we observe in the production of He^{+2} is unlikely to be due to any resonance process. Therefore for this case we propose that the mechanism is the first electron leaving the atom so quickly, by either tunneling or over-the-barrier escape, that the second electron has a substantial probability of being left in an excited state of the He^+ ion which is then immediately ionized. That is, *during the laser cycle* when the instantaneous laser field strength is large enough that neutral helium (with a binding energy of 24.58 eV) ionizes, no *excited* bound states of the ion exist. By energy arguments alone the laser is able to deplete the excited states of the ion faster than the ground state of the neutral. This double ejection will not happen at lower intensities because at these long wavelengths the first electron leaves slowly enough that the second electron has time to adiabatically adjust to its new, more tightly bound ground state. At

optical photon energies there is negligible coupling to continuum states. We note that double ejection in helium has been observed in single photon ionization for photon energies larger than the double-ionization threshold [9]. At these higher energies the ejection of the second electron is due to the overlap of the initial orbital with the continuum states. Furthermore, in these single photon experiments the ion is left with a population distribution in excited states without further ionization because of the lower intensity of the synchrotron source.

The major unknown in our mechanism is the intensity at which double ejection becomes probable. We anticipate the threshold to be an intensity high enough that the Keldysh parameter is less than 1, but lower than that corresponding to the onset of over-the-barrier ionization of neutral helium. Therefore our model has an adjustable parameter, a critical intensity I_c, which we expect to lie in the approximate range, $(3\text{-}14)\times10^{14}$ W/cm^2. A second parameter in the model, a^2, is the probability that the second electron is left in the ground state of the ion. To estimate this value we consider the measurements of the double-to-single photoionization ratio as a function of the incident photon energy reported in Ref. [9]. The maximum shakeoff probability in the single photon experiments is 3.5%–4%. This is reached when the total energy above the threshold for double ionization is over 30 eV. For total energies of 10 eV the fraction is 1%. In our experiments the ponderomotive energy is 36 eV at 10^{15} W/cm^2. For ultrashort laser pulses we expect the average photoelectron energy to be less than half this value [10]. Therefore for our model we chose $1-a^2$ to be 2%, somewhat higher than the lower value because of the additional contribution from the absence of excited bound states. Our results and conclusions are not very sensitive to this parameter, since a small change in the critical intensity can account for reasonable variations in the shakeoff fraction.

We solve the following set of rate equations for the atoms in the focal volume to predict the ion yields in this model:

$$\begin{aligned} dn_0/dt &= -k_0 n_0 , \\ dn_1/dt &= k_0' n_0 - k_1 n_1 , \\ dn_2/dt &= (k_0 - k_0') n_0 + k_1 n_1 , \end{aligned} \tag{3}$$

where n_i and k_i are the population and ionization rates for charge state i, k_0' is $a^2 k_0$ for $I > I_c$ and is k_0 otherwise. We use the single-active-electron ionization rates calculated for He and He$^+$ [11] which in the sequential regime agree well with the ADK rates discussed above. Therefore up to I_c the ionization is sequential, and above it, a direct, double ejection component is possible. Only those neutrals which survive to I_c can double ionize in this model. Integrating the above rate equations for the spatial-temporal intensity distributions within the focal volume we obtain the ion yields shown in Fig. 3. Again the data have been uniformly shifted down the intensity scale by a factor of 1.25 for comparison with the theory. By varying I_c, the nonsequential ionization signal can be reproduced accurately, including the shape, the intensity shift above the single-ionization threshold, the magnitude relative to the single-ion yield, and its merge with the sequential prediction at highest intensities. The absolute yields are accurate for both charge states. The optimum value of I_c is found to be 1.03×10^{15} W/cm^2, near the middle of the expected range. This seems to indicate that it is actually tunneling rather than over-the-barrier ionization which initiates this process.

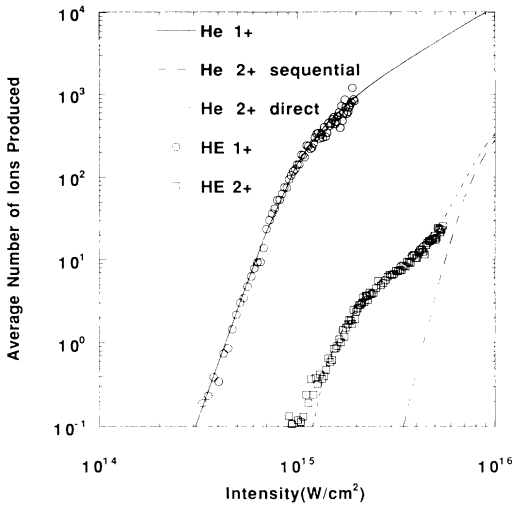

FIG. 3. Ion yield data compared to sequential and nonsequential numerical solution of the Schrödinger wave equation. The data for both charge states have been uniformly shifted down in intensity by a factor of 1.25 for comparison with theory.

The ultrashort pulse used in this work is critical for producing a measurable number of double ejection events. For pulses of 1 ps duration and longer, our calculations show that this process will not be significant because too few neutral atoms survive to I_c. In fact, measurements at 1 μm with 2 ps pulses [12] have shown no evidence of double ejection in helium. Earlier observations of xenon ion yields [13] using lower intensities and much longer pulses (5–200 ps) exhibited an intensity dependent structure which may be attributed to an ion resonance [14]. It is not likely caused by the shakeoff mechanism discussed here.

We have shown our model to be capable of reproducing the observed helium ion yields as functions of the laser intensity, but it also says something about the photoelectron energy, angular distributions, and the effect of using other polarizations and frequencies. When the nonsequential process occurs, the energy distribution for the first electron will be modified because the second electron will be ejected with somewhat arbitrary energies. In ad-

dition to the above-threshold ionization structure seen in single ionization, a weak broad distribution will be present due to the direct process. Therefore very careful coincidence measurements of the electron energy distributions will be necessary to definitively corroborate that the structure in the He^{+2} yield is attributable to direct double ionization. Our model assumes that both electrons escape almost in phase when the electric field of the laser is at its maximum. Therefore, both electrons should be ejected in the same direction which is in contrast to the back-to-back emission expected according to the Wannier threshold law [15]. In addition, this shakeoff mechanism for nonsequential ionization should remain with circular polarization and all optical wavelengths provided the pulse length is short enough.

In summary, we have found the saturation in parallel of the He^{+} ion yield and a feature in the He^{+2} ion yield. This is the first direct experimental evidence for nonsequential double ionization of helium using intense, ultrashort laser irradiation at optical frequencies. We have also proposed a new mechanism for the nonsequential ionization. These data show that one cannot infer peak intensity using the common method of measuring threshold or appearance intensities and comparing them with the predictions of sequential models. Similar investigations with heavier noble gases can be applied to the study of tunnel-ionization-driven recombination lasers.

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