Momentum Distributions of Ne^{n+} Ions Created by an Intense Ultrashort Laser Pulse

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Vector momentum distributions of Ne^{n+} ($n = 1, 2, 3$) ions created by 30 fs, ≈ 1 PW/cm^2 laser pulses at 795 nm have been measured using recoil-ion momentum spectroscopy. Distinct maxima along the light polarization axis are observed at 4.0 and 7.5 a.u. for Ne^{2+} and Ne^{3+} production, respectively. Hence, mechanisms based on an instantaneous release of two (or more) electrons can be ruled out as a dominant contribution to nonsequential strong-field multiple ionization. The positions of the maxima are in accord with kinematical constraints set by the classical "rescattering model."

In the past, a profound understanding of nonlinear physics leading to single ionization and high harmonic generation has been achieved within a single active electron (SAE) model. Contrarily, the observation of surprisingly large yields of multiply charged ions emerging from the interaction of intense laser fields with single atoms has been the subject of controversial debate since the first experimental evidence was reported about 15 years ago [1]. Even recent precision measurements (see, e.g., [2–4]) could not unambiguously clarify the mechanisms of enhanced multiple ionization. The prediction of multiple ionization yields within SAE, assuming sequential ionization of independent electrons, fails by many orders of magnitude (for an overview, see [5]). While there is common agreement that the correlated electron-electron dynamics is responsible for the observed discrepancy [6], the "nonsequential" (NS) mechanisms leading to the enhanced yields remained unclear and are under lively discussion (see, e.g., [7,8]). Up to now, this debate was solely based on total multiple ionization rate measurements.

In essence, three dynamic mechanisms have been proposed for NS double ionization: First, when one electron is rapidly removed in the laser field the remaining electron cannot adiabatically readjust to the ionic potential and is instantaneously "shaken off" into the continuum [2]. This mechanism is known to dominate double ionization of helium after absorption of single photons with energies beyond 1 keV [9]. Second, a "rescattering" process was proposed [10] within a semiclassical model where the second electron is ionized ($e, 2e$) in a collision with the first electron hitting its parent ion after free propagation during about half an optical cycle in the external laser field. This process is also immanent to many-body intense-field S-matrix calculations [11] which are in excellent agreement with experimental data. Rescattering in a generalized view has been shown to play an important role for NS ionization in SAE calculations where the e-e Coulomb in-

teraction has been approximately incorporated [7]. Third, and most recently, collective multielectron tunneling has been considered in detail but found to be quantitatively too weak. A semiempirical expression resembling a tunneling formula has been found that is in good overall agreement with measured multiple ionization rates [12]. A conclusion that "tunneling is responsible for NS double ionization" has been drawn from the measured ratios of double to single ionization yields over a large range of laser intensities [3].

In this Letter, we present first differential experimental data on double and triple ionization of neon in ultrashort (30 fs FWHM) laser pulses at intensities (≈ 1 PW/cm^2) where NS multiple ionization dominates but saturation is not yet reached. Cold target recoil-ion momentum spectroscopy (COLTRIMS) [13] has been used to measure the momentum vectors $\vec{p}_{\text{ion}}$ of ejected Ne$^{(1-3)+}$ ions. In the limit of negligibly small momentum transfer by the absorbed photons ($p_\gamma < 0.05$ a.u.) and for short laser pulses (no ponderomotive acceleration of electrons) $\vec{p}_{\text{ion}}$ reflects the sum momentum of emitted electrons $\vec{p}_{\text{ion}} = -\sum \vec{p}_e$. Thus, the ion momentum distributions are a sensitive measure of the many-electron dynamics. We demonstrate that each proposed multiple ionization mechanism leads to distinct momentum patterns of the ions. The comparison with the observed distributions gives, for the first time, decisive information about the importance of different mechanisms.

The experiment was performed at the Max Born Institute using light pulses of a Kerr-lens mode locked Ti:sapphire laser at 795 nm wavelength amplified to pulse energies of up to 600 μJ at 1 kHz repetition rate. The light pulses of 30 fs (FWHM) were focused by a lens ($f = 260$ mm) to a spot of 30 μm (FWHM) diameter into an ultrahigh vacuum chamber (5×10^{-11} torr) reaching pulse peak intensities up to 1.8 PW/cm^2. Intensity fluctuations monitored during the experiment

were kept below 5%. To verify the intensity calculated from the beam parameters we measured Ne^{1+} momentum distributions for circularly polarized light at different intensities. An annular momentum distribution was observed with a diameter which is in accordance with previous measurements of electron energy distributions and theoretical models [14].

At its focus the laser beam was crossed by a low-density (10^8 atoms/cm^3) supersonic jet of Ne atoms. The jet was formed by expanding Ne gas at a pressure of 5 bar through a cooled (LN$_2$ temperature) 10 μm nozzle. After expansion, the beam was collimated over a total length of 2 m to a diameter of 2 mm at the interaction point. Ions created in the focus were extracted by a weak homogeneous electric field of 1 V/cm. After acceleration over 10 cm and passing a field free drift tube of 20 cm length they were recorded by a two dimensional position sensitive multichannel plate detector (0.1 mm position resolution). From the time of flight the charge state of the ion and its momentum component in the direction of extraction is obtained, the two other components are calculated from the position on the detector. Thus, COLTRIMS yields the charge state and the momentum vector of each single ion with an actual resolution in any direction of $\Delta p_{x,y,z} \leq 0.2$ a.u. (limited mainly by the temperature of the gas jet) and a solid angle for acceptance of 4π for all ions with momenta below $|\vec{p}_{ion}| \leq$ 10 a.u. Only the salient points of recoil-ion spectroscopy are given here; details can be found in Ref. [13].

In Fig. 1 the momentum distributions of Ne^{1+} ions parallel ($P_\parallel$) and perpendicular ($P_\perp$) to the polarization axis are shown for linearly polarized light. The laser intensity of 1.3 PW/cm^2 corresponds to a Keldysh parameter [15] of $\gamma = 0.35$ indicating that the production of singly
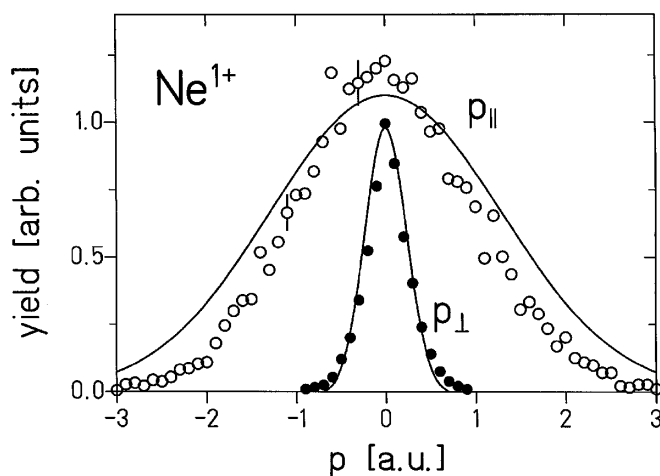
charged ions mainly occurs via tunnel ionization. In this case most of the ions are "born" at a time when the electric field maximizes. For linear polarization the electric field can be written as $\vec{E}(t) = \vec{E}_o(t)\sin(\omega t)$. Assuming that an ion is created with charge state q and zero initial momentum at a time t_o the final ionic drift momentum strongly depends on the phase $\phi = \omega t_0$ as

$$p(t_0) = \frac{q}{\omega} E_0(t_0)\cos(\omega t_0). \tag{1}$$

Thus, the final ion momentum distribution contains information about the phase ϕ when ionization occurs. It follows from Eq. (1) that ions created at $\phi = 90°$ (i.e., at maximum field strengths where the tunneling probability maximizes) gain zero drift momentum and, hence, the final momentum distribution peaks at zero. Our experimental results are in nice agreement with a simple expression for electron momentum distributions derived by Delone and Krainov [14] on the basis of tunnel ionization (Fig. 1). Thus, as expected, the Ne^{1+} momentum spectra are well understood in terms of tunnel ionization at the
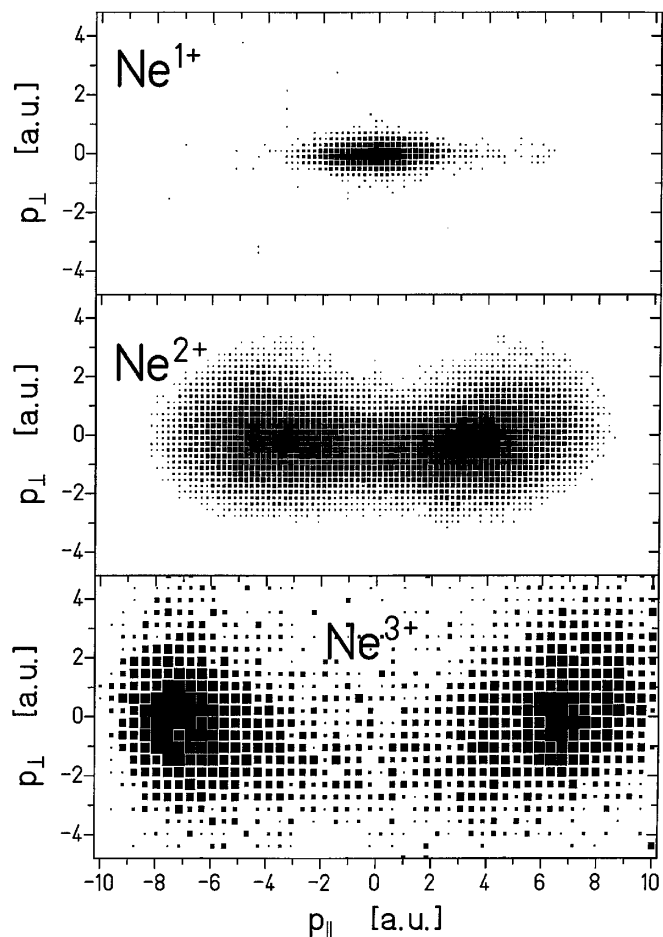

FIG. 1. Momentum distributions parallel ($p_\parallel$) and perpendicular ($p_\perp$) to the polarization axis of Ne^{1+} ions created by 30 fs linearly polarized 795 nm laser pulses at a peak intensity of 1.3 PW/cm^2 (solid line: theoretical results according to [14]).

FIG. 2. Two-dimensional momentum distributions ($p_\parallel$, $p_\perp$) of Ne^{n+} ions at peak intensities of 1.3 PW/cm^2 (Ne^{1+}, Ne^{2+}) and 1.5 PW/cm^2 (Ne^{3+}). The distributions are integrated over the third Cartesian coordinate.
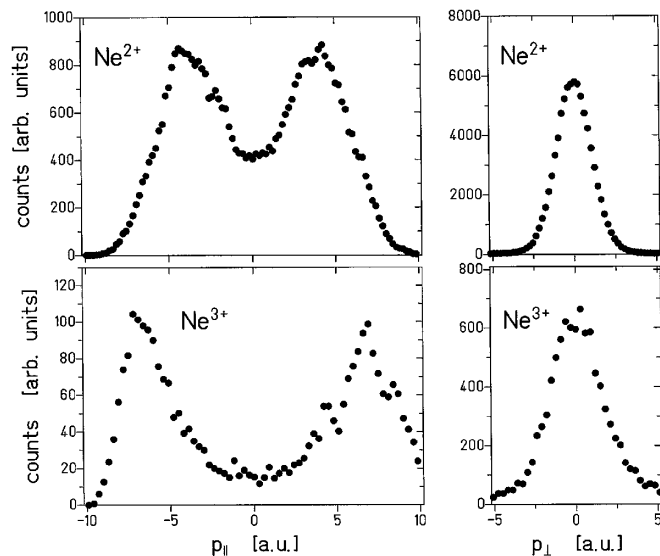
FIG. 3. Projections of the momentum distributions of Fig. 2 onto the axis parallel ($p_\parallel$) and perpendicular ($p_\perp$) to the polarization direction for Ne^{2+} and Ne^{3+}.
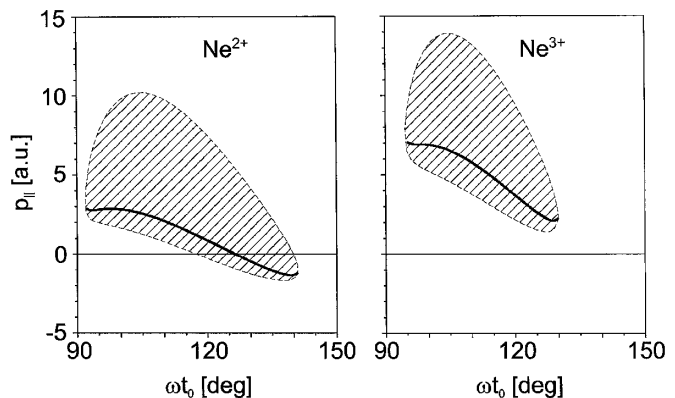

FIG. 4. Within a classical rescattering model kinematically allowed regions of final longitudinal ion momenta (hatched areas) as a function of the phase $\phi = \omega t_0$ where initial single ionization occurs. The solid line is a reasonable likely phase dependence (see text). Intensities are the same as in Fig. 2.

present intensity and the agreement confirms the approximation that the ion balances the electron momenta.

As mentioned before, different NS multiple ionization mechanisms result in specific ion momentum distributions. The "collective tunneling" mechanism [12] should lead to a momentum distribution concentrated around zero as discussed before for single ionization [see Eq. (1)]. This follows also for the proposed "shake-off" mechanism [2]. In both cases, the two electrons are released "simultaneously" close to the maximum field strength, i.e., around $\phi = 90°$ where the tunneling probability maximizes. In contrast to these instantaneous double (multiple) ionization processes a stepwise ionization by inelastic rescattering of the first electron leads to substantially different final momenta. The second or more electrons are ionized when the first electron revisits the parent ion, which occurs most likely at a phase when the electric field strength is close to zero. This results in a large final drift momentum according to Eq. (1). In addition, a certain momentum can be transferred to the ion in this ionizing (e, ne) collision.

The experimental two-dimensional $(p_\parallel, p_\perp)$ momentum distributions for Ne^{n+} ions are shown in Fig. 2. In striking contrast to Ne^+ the distributions for Ne^{2+} and Ne^{3+} exhibit a minimum at $p_\parallel = 0$ and symmetric wings peaking at 4 and 7.5 a.u., respectively. Thus, any instantaneous process leading to double or triple ionization can be excluded as a dominant contribution to multiple ionization under the present conditions. Hence, rescattering appears to be the most appropriate model.

In the following, we will give a qualitative interpretation of our experimental results using a two-step model for double and triple ionization based on inelastic ionizing collisions of the rescattered first electron [10]: First, at a time t_0 single ionization occurs via tunneling. Depend-

ing on the phase $\phi = \omega t_0$ the first electron may return back at a time t_1 leading to doubly or triply charged ion via an $(e, 2e)$ or $(e, 3e)$ process, respectively. Considering the kinematical conditions for the inelastic rescattering the final longitudinal momentum of the ion is given by

$$p_{n+}(t_0) = p_{1+}(t_1) + \Delta p + p_{n+}^{\text{drift}}(t_1), \qquad n = 2, 3, \tag{2}$$

where $p_{1+}(t_1)$ is the momentum of the singly charged ion [which balances the momentum $p_e(t_1)$ of the rescattered electron] at time t_1 (it should be noted that t_1 is a function of the initial phase ωt_0). The second term denotes the momentum Δp transferred to the ion in the (e, ne) process, and $p_{n+}^{\text{drift}}(t_1)$ is the final drift momentum an ion with charge state $q = n$ (created with zero initial momentum) accumulates.

The kinematics of the inelastic (e, ne) collision with an ionization potential I_p sets well-defined constraints for the longitudinal momentum transfer $\Delta p^{\text{min, max}} = p_e(t_1) \mp n \cdot \sqrt{(1/n)[p_e^2(t_1) - 2I_p]}$ and therefore defines via Eq. (2) a lower and upper limit of possible final longitudinal momenta of created ions as a function of the phase $\phi = \omega t_0$ when the first electron was ionized. This kinematically allowed region is displayed in Fig. 4 in addition with a reasonable likely phase dependence (solid line in Fig. 4) assuming minimal energy loss of the first electron scattered in the forward direction. Since single ionization by tunneling preferentially occurs near $\phi = 90°$ we expect favored ion momenta of 3–4 a.u. for Ne^{2+} and of 7–8 a.u. for Ne^{3+}, respectively, in close overall agreement with the experiment. A detailed description of momentum distributions within the classical rescattering model will be published in a forthcoming paper.

In conclusion, as a result of the measured momentum distributions for multiply charged Ne ions we are able to rule out several mechanisms proposed to explain nonsequential ionization in intense light pulses. Only the kinematics of the rescattering mechanism is in accordance with

the present experimental data. Upon inelastic scattering at the first return of the tunnel ionized first electron to the core a second and even two electrons can be removed from Ne^+ by electron impact ionization. In the future, kinematically complete experiments similar to those performed for ion, electron, and single photon impact [13] are planned using combined recoil-ion and many-electron momentum spectroscopy [16].

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