Measurement of the Initial Condition of Electrons Ionized by a Linearly Polarized, High-Intensity Laser

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We have measured the angular spectrum of electrons born in a linearly polarized, high-intensity laser focus. The spectrum is directly related to the initial conditions of the ionized electrons in the plane perpendicular to the propagation direction. Our measurements for high charge states of neon are in good agreement with predictions of quasistatic tunneling models and limit the initial electron kinetic energy to approximately 0.5% of its average quiver energy. This measurement is important for studies of high-order harmonic generation and direct double ionization. [S0031-9007(96)02268-5]

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Over the past several years, a simple two-stage model of ionization, referred to as the "quasistatic" or "simpleman's" model [1–4], has succeeded in explaining a wide variety of experimental aspects of intense field-atom interactions in the tunneling regime. These include many characteristics of the electron energy spectra [5], direct double ionization in the tunneling regime [6], and the cutoff energy of the high-harmonic plateau [7]. The basic assumption of the model is that ionized electrons can return to collide with the nucleus with an energy that depends on both their initial momentum and the phase within the optical cycle at which they were born. The initial phase, in turn, depends on the ionization model. For classical ionization, or barrier-suppression ionization (BSI) [8], the electrons are born only at the peak of the field and hence have zero initial phase. In ac tunneling models [9,10], the electrons are born over a range in phase within the optical cycle near the peak of the field. After ionization, the trajectory is calculated classically, neglecting the Coulomb force of the residual ions, as it is much weaker than the force due to the laser field [3]. The distributions of electrons as a function of their initial momentum and initial phase determine the rates of several of the abovementioned processes.

This Letter reports a measurement of the initial conditions of electrons ionized in the long-pulse tunneling regime. The Keldysh adiabaticity parameter [11] $\gamma \equiv (I_p/2U_p)^{1/2} < 0.1$, where I_p is the ionization potential of the atom and U_p is the ponderomotive energy [12]. Unlike some previous studies of above-threshold ionization electrons [5,13] in which the effect of the ponderomotive potential was minimized, our intent was for the freed electrons to gain the full energy provided by the field gradient. For electrons born at the peak of the field and with zero initial momentum perpendicular to the propagation direction, the ponderomotive potential leads to an axisymmetric electron distribution in the plane perpendicular to the propagation direction [14]. A small initial momentum perpendicular to the propagation direction or ionization of the electron at an off-peak phase of the optical cycle breaks the symmetry of the ponderomotive potential. We used a high-intensity laser to ionize electrons from neon gas at low density and measured the electron spectra as a function of the forward angle θ (relative to $\boldsymbol{k}$) and the azimuthal angle φ (relative to the polarization direction in the plane perpendicular to the propagation direction). We observed a significant asymmetry in the electron number distribution in the plane perpendicular to the direction of propagation, with more electrons detected along the direction of polarization than perpendicular to it. The observed asymmetry is due to the nonzero initial conditions and is consistent with quasistatic tunneling models.

Earlier work utilizing the retarding-potential grids attempted to measure the initial drift momentum by calculating the difference between the electron *energy* spectra parallel and perpendicular to the polarization direction [3,15]. The direction of the initial momentum along the polarization direction implies that the number, as well as the energy, of electrons seen along the polarization direction will be larger than the number seen perpendicular to it. The azimuthal distribution of the *number* of electrons is a more sensitive measure of the initial conditions than the azimuthal distribution of the *energy* of the electrons. In addition, at high electron energies, the forward electron momentum [16] complicates the retarding potential method.

The distribution of electrons as a function of energy E, forward angle θ, and azimuthal angle φ was measured for electrons produced during the creation of Ne^{6+}, Ne^{7+}, and Ne^{8+} with linearly polarized laser light. Electrons with energies up to 30 keV were observed using the magnetic spectrometer described below [16]. Previous measurements with circularly polarized laser pulses showed photon momentum transfer to the ejected electrons [16] and effects of the mass shift of electrons oscillating in the laser field [17]. The measurement of the electron distributions as a function of both energy and θ was necessary to resolve the closely spaced energy peaks of the upper charge states of neon in the linearly polarized field. Measurement of the electron distributions

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perpendicular to the $\boldsymbol{k}$ vector exclusively would neglect the forward momentum gained by the electrons [16]. The relation between the electron's final kinetic energy E and forward scattering angle θ is given to good approximation by [16,18–20]

$$\theta = \tan^{-1}\sqrt{\frac{2mc^2}{E}}\,. \qquad (1)$$

This relation remains valid regardless of the electron's initial momentum. Electron energy distributions were measured both along and perpendicular to the polarization vector by leaving the spectrometer stationary and rotating the laser field polarization $\pm 45°$ using an AR-coated half-wave plate.

The spectrometer [16,21] consists of an electromagnet for selecting electron energy and a detector composed of a scintillator coupled to a photomultiplier tube. The energy window of the spectrometer was varied by changing the magnetic field in the 2-mm gap of the steering magnet. A calibration using a beam of electrons of known energy showed an energy resolution of $\Delta E/E = 0.3$ FWHM. The distribution of electrons as a function of θ was measured by rotating the entire spectrometer about the cylindrical axis that passes through the laser focus at 90° to the laser axis. The angular resolution of the spectrometer is $\Delta\theta = \pm 1.5°$, due to the 1° geometric angular resolution of the spectrometer and a 0.5° uncertainty in the position of the spectrometer about the cylindrical axis. The angular acceptance in the plane perpendicular to the $\boldsymbol{k}$ vector was $\Delta\varphi = 3.3°$. Peak signal-to-noise ratios of 1000 to 1 were obtained with this setup [16].

The laser system used for the experiments was a 1.054-μm, 2-ps laser using chirped-pulse amplification described elsewhere [22]. The laser was focused to a 5-μm ($1/e^2$ radius) focal spot and a peak laser intensity of approximately 1×10^{18} W/cm^2. Neon gas at a pressure of 1×10^{-3} Torr in a background pressure of 1×10^{-8} Torr was used to minimize space charge effects. Laser ionization provided electrons whose interaction time with the focus was shorter than the pulse length or collision time. The signal obtained at each (E, θ, φ) point was normalized to the focal volume expected at 1×10^{18} W/cm^2 to correct for a 10% shot-to-shot fluctuation in laser energy. Imaging of the focus onto a CCD camera revealed a stigmatic focus with a 0.9 ± 0.2 aspect ratio independent of the direction of polarization determined by the position of the half-wave plate. The shot-to-shot variation in the laser's pointing accuracy was ± 1.2 μm and did not increase if the wave plate was rotated between shots.

The number of electrons seen parallel and perpendicular to the polarization direction for each charge state was determined through the contribution of each state to the energy spectrum by making a multiple-Gaussian least-squares fit to each spectrum measured at forward angles θ of 78° through 90° and azimuthal angles of 0° and 90° [21]. Gaussian fits were then made to the number of electrons seen at $\varphi = 0°$ and $\varphi = 90°$ as a function of θ for each charge state. The forward angle θ of peak electron number agreed with Eq. (1) for all charge states observed. Figure 1 shows the measured distributions of Ne^{6+} and Ne^{8+} electrons as a function of forward angle θ, both parallel and perpendicular to the direction of polarization. The ratio of the peak number of electrons seen along the polarization axis, N_{para}, to the peak number seen perpendicular to the polarization axis, N_{perp}, was found to be 1.9 ± 0.4, 2.3 ± 0.3, and 1.8 ± 0.2 for Ne^{6+}, Ne^{7+}, and Ne^{8+}, respectively.

As stated above, ionization of the electron at a nonzero phase of the optical cycle or with a nonzero initial kinetic momentum perpendicular to the propagation direction can give an asymmetric angular spectrum. Both contributions can be described on equal ground as the initial transverse canonical momentum. This quantity can be written as

$$\boldsymbol{P}_{\perp}(t_0) = \boldsymbol{p}_{\perp}(t_0) + \frac{e}{c}\boldsymbol{A}_{\perp}(\Delta\eta)\,, \qquad (2)$$

where $\boldsymbol{p}_{\perp}(t_0)$ is the kinetic momentum perpendicular to the propagation direction at the time of ionization t_0 and $\Delta\eta$ is the phase displacement from the peak of the field at $\eta = kz - \omega t_0 = n\pi (n = 0, 1, 2\ldots)$. The electric field has the form $\boldsymbol{F}(\eta) = \boldsymbol{F}_0 \cos\eta$ if we choose the vector potential $\boldsymbol{A}(\eta) = (c\boldsymbol{F}_0/\omega)\sin\eta$ and note that $\boldsymbol{F} = -(1/c)\partial\boldsymbol{A}/\partial t$. F_0 is the field strength at the peak of the optical cycle during which ionization occurs, and ω is the laser frequency. The initial kinetic momentum $\boldsymbol{p}_{\perp}(t_0)$ could have components both parallel and perpendicular to the polarization direction. In our calculations of a

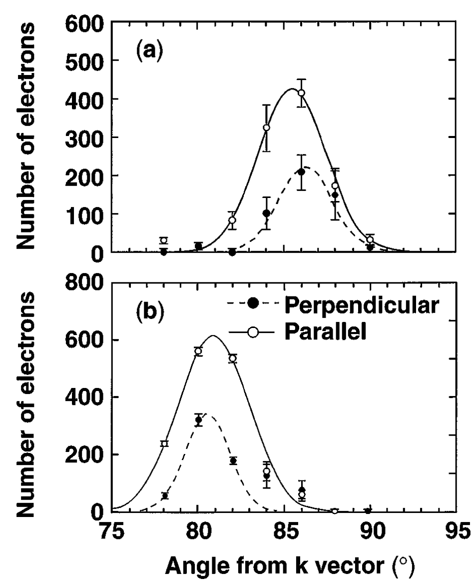

FIG. 1. Angular spectrum (relative to the $\boldsymbol{k}$ vector) of (a) Ne^{6+} and (b) Ne^{8+} electrons observed both parallel (open circles) and perpendicular (closed circles) to the direction of polarization. The solid and dashed lines are Gaussian fits to the data.

classical trajectory, we disregard contributions such as wave-packet spreading, and assume any initial kinetic momentum is small and in the polarization direction.

A fully relativistic Monte Carlo simulation [21] has been used to determine the relation between the number ratio $N_{\mathrm{para}}/N_{\mathrm{perp}}$ and the electron's initial canonical momentum. We performed a series of simulations in which the atoms were ionized according to BSI [8] at the peak of the field ($\Delta\eta = 0$), and the electrons were given a Gaussian distribution of initial canonical momenta with a HWHM width P_{H} around $\boldsymbol{P}_{\perp}(t_0) = 0$. A linearly polarized laser pulse with a Gaussian spatial and temporal profile was propagated over several thousand atoms placed randomly throughout the focal volume. The fully relativistic equations of motion for the trajectories were solved for each electron. The parameters used in the simulation reflect those of our laser system. Data obtained from this simulation were analyzed in the same manner as the experimental data. Figure 2 shows the number ratio $N_{\mathrm{para}}/N_{\mathrm{perp}}$ for electrons released in the creation of the eighth charge state of neon as a function of the HWHM width P_{H} of the initial canonical momentum distribution. P_{H} is given in units of the maximum quiver momentum eF_0/ω. The calculated relationship between $\boldsymbol{P}_{\perp}(t_0)$ and eF_0/ω is similar for Ne^{6+} and Ne^{7+}. From the calculations we can estimate the experimental value of P_{H} to be 24 $\pm$ 9%, 29 $\pm$ 3%, and 25 $\pm$ 3% of eF_0/ω for Ne^{6+}, Ne^{7+}, and Ne^{8+}, respectively. This corresponds to an average initial rms drift energy that is about 10% of the ponderomotive energy U_p for all charge states considered.

The initial canonical momentum distributions inferred from the data are in good agreement with those predicted by the Ammosov-Delone-Krainov (ADK) tunneling ionization model [10] when the electrons have zero initial kinetic momentum. The ADK rate falls off exponentially as the amplitude of the applied electric field and, therefore, gives a significant probability of ionization near, but below, the peak of the field during the optical cycle.

To determine the initial canonical momentum width produced by the ADK theory, the Monte Carlo simulation was modified to employ this tunneling theory of ionization instead of the simpler BSI theory. For each atom, the ionization rate was calculated several hundred times per laser cycle, and the electron was released with zero initial kinetic momentum at a phase corresponding to this probability rate. Figure 3 shows the distribution of electrons produced by the simulation for the eighth charge state of neon as a function of $\Delta\eta$. The HWHM width of this distribution was found to be about 0.27 rad for each charge state Ne^{6+}, Ne^{7+}, and Ne^{8+}, corresponding to a $\boldsymbol{P}_{\perp}(t_0)$ distribution width of about $0.27eF_0/\omega$, in good agreement with the values derived from the experimental data.

The agreement of our data with the predictions of the ADK model implies that the initial kinetic momentum $\boldsymbol{p}_{\perp}(t_0)$ of the electrons must be small in comparison with the electron's final momentum. The error ranges of our peak electron energy data (parallel and perpendicular to the polarization direction) and our $N_{\mathrm{para}}/N_{\mathrm{perp}}$ data put an upper limit on $\boldsymbol{p}_{\perp}(t_0)$. If we allow for a nonzero initial kinetic momentum in the polarization direction in the Monte Carlo simulation using the ADK theory, our data limits $\boldsymbol{p}_{\perp}(t_0)$ to 5.5 $\pm$ 1.6%, 2.7 $\pm$ 2.1%, and 6.1 $\pm$ 3.0% of eF_0/ω for Ne^{6+}, Ne^{7+}, and Ne^{8+}, respectively. This corresponds to initial kinetic energies of less than 50 eV for high-energy electrons with final energies of about 10 keV. Future experiments using a higher-resolution spectrometer and elliptic polarization could lower this upper limit on the initial kinetic momentum.

In summary, we have made the first distribution measurement of the initial conditions of electrons born in a linearly polarized, high-intensity laser focus in the tunneling regime. Good agreement was achieved between the fully relativistic simulation employing the ADK tunneling model and measurements of the momentum distribution width deduced from the asymmetry measured in the

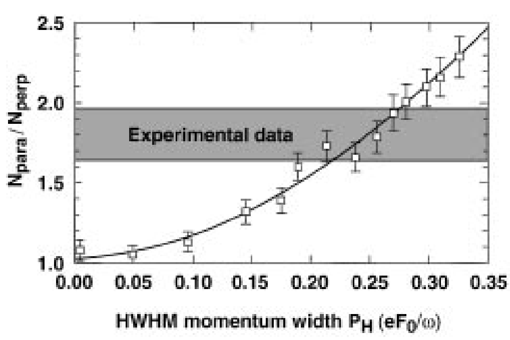

FIG. 2. Comparison of the Ne^{8+} asymmetry ratio $N_{\mathrm{para}}/N_{\mathrm{perp}}$ to the HWHM width P_{H} of the initial canonical momentum distribution. Squares represent values of $N_{\mathrm{para}}/N_{\mathrm{perp}}$ calculated from data generated by a simulation with the momentum distribution width P_{H}. The error bars are statistical. The solid line is a polynomial fit to the data. The limits of the shaded region are those of the experimental data.

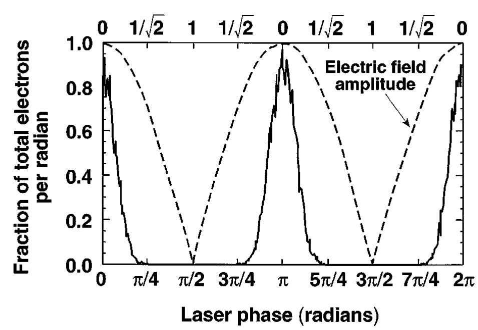

FIG. 3. Distribution of the electrons produced during the creation of Ne^{8+} as a function of the laser phase $\Delta\eta$ at which they were ionized, from the ADK simulation. The amplitude of the laser electric field (dashed line) is overlaid for comparison. The top axis shows the corresponding initial electron momentum in units of eF_0/ω.

angular distribution of electrons. This successful test of the quasistatic tunneling model has important implications for the interpretation of experimental results in high-order harmonic generation and for the development of further-refined models of electron spectra and direct double ionization.

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