Momentum distribution of highly charged ions formed by strong laser fields

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Abstract. Momentum distributions of highly charged ions produced by a strong laser pulse with duration of ~ 200 fs and maximum intensity of ~ 50-70 PW/cm² are studied. Due to the spatial variation of the laser intensity within the beam focus, different charge states are produced during a single laser shot. The measured widths of the momentum distribution of the product ions exhibit a linear dependence on the ionization potential of the ions for various rare gas atoms. Such a linear dependence is analyzed theoretically using the single active electron model.

1. Introduction
Due to technological advances high-power lasers have become available which are strong enough to multiply ionize atoms and produce highly charged ions [1, 2]. The dynamics of atoms in such strong lasers can be studied by measuring not only the ion yield but also the momentum distribution of the product ions [3, 4, 5]. This provides deeper insights into the ionization dynamics. The measured distributions of the multiply charged ion momentum parallel to the polarization axis of the electric laser field is observed to be sensitive to whether the ionization is sequential or non-sequential [3, 6]. For weak laser intensities non-sequential ionization involving strong electron-electron correlations (rescattering) [7, 8] dominates leading to two distinct peaks with a dip at \( p = 0 \) in the momentum distribution. On the other hand, strong lasers ionize electrons from an atom sequentially and the resulting momentum distribution is Gaussian-like with a peak at \( p = 0 \) [4, 9].

In this paper we explore the regime with a very intense laser field by which up to 8-fold charged ions can be produced via sequential ionization. The driving field considered is a linearly polarized laser field approximated by

\[
F(t) = F_{\text{max}} f(t) \cos(\omega t + \phi) \quad (\tau < t < \tau),
\]

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Figure 1. Measured (a) and calculated (b) width (FWHM) of the ion momentum distribution as a function of ionization potential $I_p$. The atoms are driven by the laser field (50-70 PW/cm$^2$, 775nm, 200fs). The results are fitted to a power law $\propto I_p^{1.1}$ indicated by the dashed line.

where $F_{\text{max}}$ is a peak field strength, $\omega$ a laser frequency, $\phi$ the carrier envelope phase, and $f(t) = \cos^2(\pi t/(2\tau))$ the approximate laser envelope. (For long pulses of about 200 fs (FWHM), the carrier envelope phase $\phi$ is unlikely to be important and neglected ($\phi = 0$) in the following.) Due to the focusing of the laser, $F_{\text{max}}$ depends on the position of atoms relative to the focal point of the laser. The closer the atoms are to the focal point, the larger the maximum strength $F_{\text{max}}$ is. As a consequence, the spatial spread of the atomic beam in the interaction region results in a broad charge state distribution within a single laser shot. The momentum distribution of the produced ion $A^{q+}$ is thus that integrated over a range of laser intensities: from the ionization threshold of $A^{q+}$ to the intensity for which the ion yield of the next charge state becomes dominant. The main contributions result from the laser intensity near the upper boundary for which the yield of $A^{q+}$ is largest. In such a strong field sequential ionization is the dominant process and the momentum distribution is expected to be Gaussian. The width of the distribution is observed to display a nearly linear behavior as a function of the ionization potential of each ion (Ne and Ar, see Fig. 1a). Such a dependence is also observed for other rare gas target atoms [5].

The theoretical investigation of multiple ionization is not an easy task. As the number of electrons involved increases, the ab-initio quantum calculation of the many-body dynamics becomes beyond the reach of currently available computing power. When each electron is ionized sequentially, however, we can simplify the model drastically: the ionization of each electron is analyzed by the single active electron model. In the next section, we briefly review the method to simulate sequential ionization processes which reproduces the observed behavior in the measured width of the momentum distributions [9]. In Sec. 3, we introduce an even simpler model to understand the origin of the nearly linear scaling observed in the measured results. We show that for very strong laser fields with a duration of 200 fs the electron is ionized (detached) even before the laser field reaches its maximum amplitude. While the electron will experience the maximum amplitude of the laser after ionization, its final momentum is determined by the temporal laser amplitude at the time of ionization rather than the maximum laser amplitude $F_{\text{max}}$. This temporal laser amplitude features the linear dependence on $q$ which eventually leads to the linear scaling of the momentum distribution width.

2. Simulation of sequential ionization by the single active electron model
We briefly review the quasi-classical method used to simulate the momentum distribution of highly charged ions by a strong laser field (see details in [9]). In sequential ionization processes,
the detachment of each electron is simulated independently within a single active electron model using the classical Monte Carlo method including tunneling (CTMC-T) \cite{10, 9}. We consider the production of \( A^{q+} \) ion by the laser field with a given peak field strength \( F_{\text{max}} \) [Eq. (1)]. The detachment of the \( i^{\text{th}} \) electron is simulated by the Hamiltonian

\[ H_i = \frac{p_i^2}{2} + V_i(r) + zF(t) \]  

(2)

where \( V_i(r) \) is the model potential \cite{11} representing the core potential of the \( A^{i+} \) ion screened by bound electrons. The interactions between the electron to be detached and \( i - 1 \) electrons which are detached earlier than the \( i^{\text{th}} \) electron are neglected. (Atomic units are used throughout unless otherwise stated.) The equations of motion are solved numerically for an ensemble of trajectories (microcanonical ensemble with \( H_i = I_p(i) \)) representing the ground state wavefunction of the \( i^{\text{th}} \) electron with the ionization potential \( I_p(i) \). Each trajectory is allowed to tunnel through the potential barrier formed by the core potential and the laser field, with the tunneling probability estimated semiclassically, so that the tunneling ionization can be properly represented without solving the Schrödinger equation. The final momentum distribution of the electron can be determined by statistically sampling the final momenta of trajectories of the ensemble. Due to momentum conservation, this electron momentum distribution is considered to be identical to the ion momentum distribution. When the simulated momentum distributions are compared with the measured data, they are integrated over a range of laser peak field strength \( F_{\text{max}} \) weighted by the distribution of the laser field strength within the interaction region (\( \propto 1/F_{\text{max}} \)) and the ionization probability.

For the momentum distribution of the \( A^{q+} \) ion, the \( q^{\text{th}} \) detached electron has the largest momentum and provides the main contribution to the overall width of the ion momentum distribution. Considering only the main contribution from the \( q^{\text{th}} \) electron we could well reproduce the measured width \( \Delta p \) for different rare gas atoms. The estimated width depends almost linearly on the ionization potential \( I_p(q) \) (Fig. 1b).

### 3. Analysis using the strong field approximation

When we consider the production of the \( A^{q+} \) ion by the laser with a given peak field \( F_{\text{max}} \), the peak field has to be large enough to detach \( q \) electrons but should not be too strong to detach the \((q + 1)^{\text{th}}\) electron. Figure 2 displays the ionization probability of \( \text{Ne}^{i+} \) ions (Eq. (2) with \( i = q \)) as a function of \( F_{\text{max}} \). In this calculation, the hydrogenic approximation for the core

![Figure 2](image)

**Figure 2.** Ionization probability (solid lines) of Ne ions as a function of peak field strength \( F_{\text{max}} \) of the laser pulse (775nm, 200fs). The vertical dashed lines are the classical ionization thresholds \( F_{\text{th}}(q) \).
potential $V(r) = -q/r$ was used for simplicity. Near the classical ionization threshold

$$F_{th}(q) = I_p(q)^2/(4q) \quad (3)$$

the yield of Ne$^{q+}$ ion saturates while the next charge state starts appearing around its threshold $F_{th}(q+1)$. Thus, the momentum distribution is integrated over a range $F_{th}(q) < F_{max} < F_{th}(q+1)$. While the peak laser amplitude exceeds the ionization threshold $F_{th}(q)$, the potential barrier at the saddle point ($x = y = 0$) is lowered below the binding energy $-I_p$ of the $q^{th}$ electron. However, the potential energy ($E_z = -I_p - (p_x^2 + p_y^2)/2$) of the electron can be below the barrier preventing the ionization over the barrier. Thus it is important to include the tunneling effect into the ionization dynamics even when the peak laser amplitude is above the threshold value. For long ($\sim 200$fs) laser pulses, the electron will be ionized mostly even before the laser reaches its maximum amplitude. The tunneling rates are small but the electron is exposed to weak fields for long times due to the slow ramping of the laser causing the tunneling probability to be non-negligible. Figure 3a displays the Ne$^{1+}$ ion yield by the laser field with a peak field strength of $F = 1.4F_{th}(q = 1)$. The electron is ionized before the laser field reaches the maximum. When the laser amplitude (solid light blue/grey line) is near the threshold (dashed line), the ionization yield reaches the half of the maximum value within a single peak. The corresponding momentum shift $\delta p_{max}$ is the width of the final momentum distribution (see the main text for the details.)
shift becomes nearly zero $\sin(\omega t_{\text{nn}}) \sim 0$ (Fig. 3c). Accordingly, as the ionization yield is non-negligible for a finite duration $\pi/(2\omega) - \delta t < t_{\text{nn}} < \pi/(2\omega) + \delta t$ (Fig. 3b), the electron momentum is shifted at maximum $\delta p_{\text{max}} = F_{\text{max}} f(t_{\text{nn}}) \delta t + O(\delta t^2)$ (Fig. 3c). Considering that $p_z(t_{\text{nn}}) \approx 0$ at tunneling the width of the momentum distribution for the ionized electron is given by $2\delta p_{\text{max}}$ and the width is simply determined by the temporal laser amplitude at tunneling, $F_{\text{nn}} = F_{\text{max}} f(t_{\text{nn}})$. The ionization yield is non-negligible over many optical periods. For the quantitative evaluation of the momentum distribution width, therefore, the average of momentum shift $\delta p_{\text{max}}$ over those optical periods has to be calculated. However, this averaging broadens the distribution similarly for all ions with different $I_p(q)$ without changing the $I_p$-dependence of the distribution width $\delta p_{\text{max}}$. Thus the scaling behavior of the momentum distribution width can be analyzed without this averaging taken into account. As seen in Fig. 3a the electron is ionized with the largest probability when the laser amplitude is around the threshold value $F_{\text{nn}} \approx F_{\text{th}}(q)$. The temporal laser amplitude $F_{\text{nn}}$ at the largest tunneling probability can be more precisely determined as the field strength for which the static ionization rate (of the order of $10^{-2}$ a.u. for Ne with $F = F_{\text{th}}$ with only a small dependence on $q$) multiplied by an optical cycle (of the order of $10^2$ a.u. for $\omega = 0.059$ a.u.) becomes on the order of one. Indeed, for Ne, the laser amplitude $F_{\text{nn}}$ at the maximum tunneling probability slightly increases with increasing $q$ ($F_{\text{nn}} \approx F_{\text{th}}$ for $q = 1$, $1.08 F_{\text{th}}$ for $q = 2$, and $1.15 F_{\text{th}}$ for $q = 3$ (Fig. 4)). Thus an additional $q$ dependence of the temporal laser amplitude at tunneling can be fitted as $F_{\text{nn}} \approx q^{0.1} F_{\text{th}}(q)$. Thus the width of the final momentum distribution can be estimated as $2q^{0.1} F_{\text{th}}(q) \delta t$. We note that in the ionization yields (Fig. 4a-c) the 2nd peak about 2000 a.u after the 1st peak is observed. In the CTMC-T, the trajectories oriented along the laser polarization have larger ionization rates than those oriented perpendicular to the laser polarization. The latter is responsible for the 2nd maximum in the ionization yield. The interval $\delta t$ depends only on the laser frequency, i.e., $\delta t \sim \omega^{-1}$ and is rather independent of the ionization potential $I_p(q)$ or the charge $q$. According to Eq. (3), therefore, the width $\delta p_{\text{max}}$ of the momentum distribution is proportional to $I_p^2$. This scaling appears to contradict the linear behavior observed in Fig. 1. One key factor to resolve this contradiction is the $q$-dependence of the ionization potential. Figure 5 shows an approximately linear dependence of the ionization potential $I_p(q)$ for Ne and Ar, i.e., $I_p(q) \propto q$. This linear dependence is well known, in particular, for the systems, such as C$_{60}$, involving many equivalent electrons [13]. For Ne and Ar, the same applies when $q$ is small. For large values of $q$, slight deviations from the

Figure 4. Ionization yield of Ne$^{q+}$ ion ($q = 1, 2, 3$) as a function of time. Ne atoms are subject to the laser (50-70PW/cm$^2$, 775nm, 200fs). The laser envelope (solid light blue/grey line) and the ionization threshold $F_{\text{th}} = I_p^2/4$ (dashed line) are also scaled by the threshold value $F_{\text{th}} = I_p(q)/4q$.

Figure 5. $q^{\text{th}}$ ionization potential $I_p(q)$ for Ne and Ar plotted as a function of $q$. The ionization potential is fitted for each atom by a linear scaling $I_p(q) \propto q$ indicated as the dashed line.
linear scaling are expected (see the onset in Fig. 5). For the observed ions in Fig. 1 ($q \leq 5$), it is safe to assume that the ionization potential is given by $I_p(q) = qI_p(1)$. The width of the momentum thus becomes proportional to $q^{1.1}$, explaining the linear dependence on the ionization potential with the same spcies of atom (Ne or Ar) seen in Fig. 1.

Within this simplified model there is a small target-atom-dependence of the momentum width $\delta p$ through the 1st ionization potential, i.e., $\delta p \propto I_p(1)^2 q^{1.1}$. This target atom dependence is hardly seen in the measured momentum width. One known problem of the CTMC-T method is that the tunneling probabilities for hydrogenic atoms are underestimated compared to those predicted by the corresponding quantum simulations [10]. Moreover, the tunneling probabilities for rare gas atoms involving multiple electrons may cause additional uncertainties in the estimation of ionization probabilities. More accurate evaluation of the ionization probabilities may modify the momentum distribution width by a different constant factor for each target atom and narrow the small difference between the measured and calculated results. On the other hand, it is interesting to measure the width for the target atoms with relatively large 1st ionization potential, such as He, which would reveal more prominently the target atom dependence if any.

4. Conclusion
We showed that the width of the ion momentum distribution driven by an intense (50-70 PW/cm$^2$) and long (200 fs) laser pulse can be well represented by the single active electron model assuming that sequential ionization dominates the ionization process. The width is determined through the dynamics after the electron is detached from the core ion. In the strong field regime, the effect of the core potential does not play an important role and the temporal laser amplitude at the time when the electron is detached determines the final width of the momentum distribution. The obtained width explains the nearly linear behavior of the distribution width as a function of ionization potential.

Acknowledgments
This research is supported by the FWF-SFB-F016 (Austria) and EU-HITRAP Project number HPR1-CT-2001-50067. JB also acknowledges support from the RIKEN Eminent Scientist program.

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