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# Recoil-ion momentum spectroscopy of multiply charged argon ions produced by intense ($\sim 10^{16}$ W cm$^{-2}$) laser light

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**Abstract**

Multiple ionization processes of argon atoms by a linearly polarized laser light with the maximum peak intensity of about $5 \times 10^{15}$ W cm$^{-2}$ were investigated by the recoil-ion momentum spectroscopy. Momentum distributions of argon ions with charge states 3+ and 4+ were measured. The experimental results were compared with calculations based on the sequential ionization model and the ADK ionization rate [M.V. Ammosov et al., Zh. Eksp. Teor. Fiz. 91 (1986) 2008 (Sov. Phys. JETP 64 (1986) 1191)]. The calculated widths of the distributions agreed with the experimental results to within 30%.


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*Keywords:* Barrier suppression ionization; Recoil momentum spectroscopy

## 1. Introduction

There have been several experimental studies about the multiple ionization processes of an atom in a intense light field, such as yield measurements [1] and ion momentum measurements [2,3]. In a low light intensity regime, an unexpectedly large yield of multiply charged ions was reported [4]. The process leading to the large discrepancy from the prediction of the sequential ionization model is called non-sequential ionization and has attracted much attention. Recently, by measuring the momentum of recoil ions, the mechanism of the non-sequential ionization has been clearly identified as the re-scattering process [2] in the low light intensity regime. On the other hand, in a high intensity regime, the multiple ionization is believed

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to be dominated by the sequential ionization. However, experimental evidences supporting the sequential ionization model have been limited to yield measurements. Ion momentum measurement in the high intensity regime will shed new light on the multiple ionization mechanism in this regime.

In this paper, we report the recoil-ion momentum spectroscopy experiment on multiply charged argon atoms at the maximum laser peak intensity of about $5 \times 10^{15}$ W cm$^{-2}$. Using a linearly polarized laser light, the ion momentum parallel to the light polarization was measured in this high intensity region. To our knowledge, this is the first measurement of the momentum distributions of multiply charged ions as high as 4+. A slit was used in this experiment in order to eliminate ions produced from the region distant from the focal plane of the laser beam [5]. Model calculations based on the sequential ionization reproduced the experimental result reasonably.

## 2. Experimental setup

The laser system is based on the chirped pulse amplification (CPA) technique [6]. A light pulse (~1 mJ, 775 nm) from a seed laser system (CPA 2001, Clark-MXR) was amplified up to ~300 mJ by a Ti:S four-pass amplifier pumped with a ~1.5 J pulse from a frequency-doubled Nd:YAG laser (Continuum) operated at 10 Hz. The amplified light pulse was compressed by a pair of parallel gratings. After compression, the duration of the light pulse was ~200 fs and the energy was decreased to ~120 mJ.

The light was sent to an experimental chamber through a fused-silica window. The residual gas pressure of the chamber was lower than $4 \times 10^{-10}$ Torr. In the vacuum chamber, the light was focused onto an effusive argon gas beam of argon with an off-axis parabolic mirror with a focal length of 300 mm. The laser beam profile at the focal plane was elliptical, with the vertical and horizontal widths of 80 μm and 170 μm (FWHM), respectively. The density of the gas target was about $10^4$–$10^5$ cm$^{-3}$, one or two orders of magnitude lower than the residual gas.

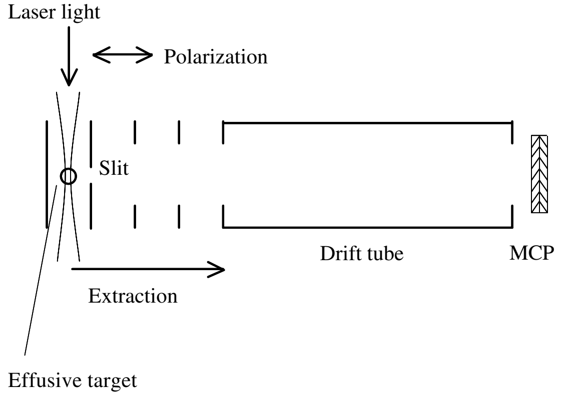


Fig. 1. A schematic drawing of the time-of-flight (TOF) analyzer.

Fig. 1 shows a schematic drawing of the experimental setup around the target. The charge state of the ions, as well as the momentum component parallel to the light polarization, was measured using the time-of-flight (TOF) technique. Multiply charged ions produced by the focused laser light were extracted by a uniform static electric field of about 3 V cm$^{-1}$. The direction of the electric field was set parallel to the light polarization. The extraction region (75 mm) was followed by a drift tube (150 mm), and the extracted ions were detected by a stack of multi-channel plates (MCPs) placed on the other side of the tube. The TOF of the ions was recorded by a multi-hit time-to-digital converter.

In the region distant from the focal plane, enormous number of low charge state ions are produced, since the laser beam has a larger cross section and the peak intensity is low. In order to detect only ions produced near the focal plane, a slit with a width of 0.5 mm was placed 5 mm downstream from the target as shown in Fig. 1. In addition, the slit greatly simplifies the treatment of the position dependent peak intensity variation in the calculation described below.

## 3. Model calculations of the ion momentum distributions

For comparison with experimental results, calculations of the ion momentum distributions were performed. Such calculations for ions with charge state as high as 4+ have not been reported previously, to our knowledge. An analytic formula is proposed, which gives an approximate momentum distribution of multiply charged ions. The formula needs only the ionization potentials and the light frequency, convenient for a rough estimation of the ion momentum distribution. However, the accuracy of this formula is limited because of several simplifications. A more accurate numerical simulation based on a rate equation is also done.

In the calculation of the momentum distribution, we take only the sequential ionization process into consideration for simplicity. It is assumed that the ion momentum is expressed as the sum of the momenta of emitted electrons. The momentum distribution of an ionized electrons is given as [7],

$$f(p_\parallel) = A\exp\left[-\left(\frac{p_\parallel}{\delta p_\parallel}\right)^2\right], \quad \delta p_\parallel^2 = \frac{3F^3}{\omega^2(2\mathrm{IP})^{3/2}}, \tag{1}$$

where $F$ is the field strength, $\omega$ is the frequency of the light and IP is the ionization potential.

The approximate formula for the ion momentum distribution is derived from the following considerations. As the light intensity increases with time at the front of the pulse, the electrons are sequentially ionized. We expect that the dominant part of ionization takes place when the intensity reaches the classical threshold value given by $I_{\mathrm{BSI}} = \mathrm{IP}^4/16Z^2$, where $Z$ represents the charge of the ion core. Therefore, in the calculation of the electron momentum distribution, corresponding field strength $F_{\mathrm{BSI}} = \mathrm{IP}^2/4Z$ is substituted into (1). Assuming there is no momentum correlation among the emitted electrons, the ion momentum distribution is given by convolution of the electron momentum distributions. The final expression for the momentum distribution of the ions with charge state $q^+$ is

$$g(p_\parallel;q^+) = A\exp\left[-\left(\frac{p_\parallel}{\Delta p_\parallel}\right)^2\right], \quad \Delta p_\parallel^2 = \frac{3}{2^{15/2}\omega^2}\sum_{j=1}^{q}\frac{(\mathrm{IP}_j)^{\frac{9}{2}}}{j^3}, \tag{2}$$

where $\mathrm{IP}_j$ represents the $j$th ionization potential. We refer to this model as 'BSI', since the model is based on the barrier-suppression ionization (BSI) model [8].

We have performed another calculation of momentum distribution which is expected to be more accurate. This calculation is based on a rate equation describing the following charge state evolution,

$$\mathrm{Ar} \xrightarrow{\Gamma_1} \mathrm{Ar}^+ \xrightarrow{\Gamma_2} \cdots \xrightarrow{\Gamma_4} \mathrm{Ar}^{4+} \xrightarrow{\Gamma_5} \cdots \tag{3}$$

The non-sequential processes is not incorporated. The cycle-averaged ADK rates are used for the ionization rates $\Gamma_q$ in (3) [9]. The rates $\Gamma_q$ in (3) are implicit functions of position and time through the light intensity. The dependence of the light intensity on the position and the time is assumed to be Gaussian, given by

$$I(r,t) = I(r)\exp[-(t/t_0)^2], \quad I(r) = I_0\exp[-(r/r_0)^2]. \tag{4}$$

($t_0$: the duration of the pulse, $I_0$: the maximum peak intensity, $r_0$: the beam width at the focal plane).[1] Here the intensity variation along the laser beam is assumed to be negligibly small within the narrow slit. In the following, the integration on time is performed first, and then the integration on position is done.

The integration of the rate equation (3) is done for each position $r$, using a Monte Carlo method. For each particle, the intensity at which ionization occurs is evaluated. Then the momentum of the emitted electron is sampled from the distribution (1). The ion momentum is calculated by summing the momenta of all the emitted electrons. This procedure is repeated for many particles and the ion momentum distribution is obtained as a function of the position. Finally, the integration on the position coordinate is done.

We note an advantage of the narrow slit. The volume element in the cylindrical coordinate

[1] More precisely, I($r$) should be replaced by an elliptical Gaussian distribution. However, it can be shown that the difference between the circular beam and the elliptic beam does not affect the calculated momentum distribution.

system is proportional to d(ln $I$), derivative of the logarithm of the peak intensity $I(r)$, as

$$dV = 2\pi \Delta w r\, dr = C dI/I = C d(\ln I) \quad (I < I_0), \tag{5}$$

where $\Delta w$ represents the width of the slit and $C$ an appropriate constant. This property greatly simplifies the treatment of the integral on space.

## 4. Results and discussions

The experiment was done for the argon target with 240,000 laser shots. The highest charge state observed was $Ar^{4+}$. The $Ar^{5+}$ signals were not identified because they were masked by $O^{2+}$ ions, which have the same $m/q$ value as $Ar^{5+}$ and come mainly from water molecules in the chamber. The average numbers of the detected argon ions per laser shot were about 0.3 and 0.03 for $Ar^{3+}$ and $Ar^{4+}$ ions, respectively. The total number of detected ions including residual gas ions, which are mainly fragments of $H_2$ and $H_2O$ molecules, was ~18 on average per laser shot.

The solid circles in Fig. 2(a) and (b) represent the measured momentum distributions parallel to the light polarization for $Ar^{3+}$ and $Ar^{4+}$, respectively. The momentum distribution exhibits a single peak without any structure. This result is in contrast to the double-hump structure associated with the non-sequential ionization process [2]. The widths of the momentum distributions parallel to the light polarization are 4.8 a.u. and 6.2 a.u. for $Ar^{3+}$ and $Ar^{4+}$ (FWHM), respectively.

In the momentum spectroscopy experiment, the space charge effect of other charged particles has to be considered. As mentioned above, the main part of the created ions was from the residual gas. We confirmed the variation of the peak width was less than 5% when the residual gas pressure was raised up to $1 \times 10^{-9}$ Torr.

The solid lines and the solid squares in Fig. 2 show the results of the 'BSI' and 'ADK' model calculations, respectively. The 'BSI' model, in spite of its simple form, gives the widths of correct orders of magnitude. However, the 'BSI' results are by a factor of about 2 narrower than the experimental

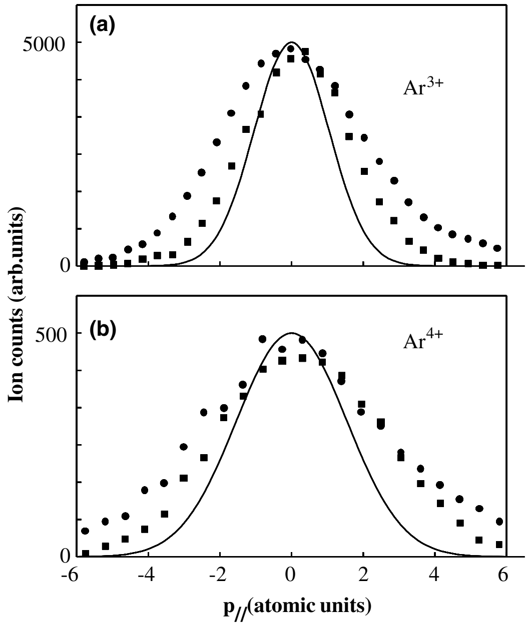


Fig. 2. The momentum distributions of $Ar^{3+}$ and $Ar^{4+}$ parallel to the light polarization. The solid circles represent the experimental distributions. The solid lines and the solid squares represent the calculations based on our 'BSI' and 'ADK' models, respectively (see text).

results. This indicates that most ionization occurred at intensities higher than $I_{BSI}$. The result of the 'ADK' model, in which the time evolution of the charge state is treated in a more realistic way, shows better agreement than the 'BSI' result. The widths of the 'ADK' results are 27% and 12% smaller than the experimental values for $Ar^{3+}$ and $Ar^{4+}$, respectively.

Finally, the validity of the assumptions made in the calculations is discussed. In the calculation, the re-scattering process is neglected. This process is known to produce ions having large momentum. Hence inclusion of the re-scattering process into the calculation would lead to broader momentum distributions. However, it is difficult to properly include this effect and is beyond the scope of this paper. Another subtle point in the calculation to be discussed is the use of the ADK rate. The ADK model describes the tunneling ionization and is valid if intensity is sufficiently low, $I \ll I_{BSI}$. It is known that the ADK model overestimates the ionization rate near and above $I_{BSI}$ [10]. In our simulation with the ADK rate, the dominant part of the ionization events occurs at high intensity $I \gtrsim I_{BSI}$. Hence, the use of more accurate ionization rate at high intensity would improve the accuracy of the calculation.

## 5. Summary

In order to investigate the multiple ionization processes of argon atoms in a linearly polarized strong light field, the ion momentum distribution parallel to the light polarization was measured. By placing a slit between the reaction volume and the ion detector, we selectively observed the ions produced near the focal plane of the laser beam. This slit also simplified the treatment for the spatial dependence of the peak intensity in the calculation.

In the experiment, the momentum distributions of $Ar^{3+}$ and $Ar^{4+}$ ion were investigated. The measured ion momentum distributions were compared with calculations. In the calculations, the sequential ionization model was adopted. Describing the time evolution of the ion charge state with a rate equation using the ADK ionization rate, the calculation reproduced the experimental momentum distribution reasonably.

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