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Production of Multiply Ionized Atoms by Ion-Atom Collisions

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A method is described of extracting and analyzing ionized target atoms induced by ion-atom collisions. The charge-to-mass ratio spectra are measured in a counting mode. The spectra for helium and neon bombarded with protons, alpha particles and nitrogen ions at 0.5−2.5 MeV in energy are presented, which approximately show binomial distributions. These charge spectra are fairly well reproduced by statistically treating the single-electron ionizations.

KEY WORDS: $He^{+,2+}$/$Ne^{+,2+,3+,4+,5+}$/keV energy

INTRODUCTION

Production of multiply charged ions at low energies has recently aroused a keen interest in connection with the problems in fusion plasmas and heavy ion accelerators. The fundamental studies about the collisional interaction of heavy ions with atoms are requested in order to know the behaviors of impurity atoms in plasma.[1] The ionization of impurity atoms is of course endoenergetic and works to degrade the plasma temperature. Beams of heavy ions with high ionicities are needed to construct the economical accelerators, but the production of these ions having low energies are not simple. Quite recently, a pulsed ion source called "EBIS" has been developed, in which a device for confining electrons and ions with a relatively long duration is attained. Unfortunately this EBIS has not been widely used, and at present, the reports using slow multicharged ions[2~4] are not plenty.

On the other hand, there have been numerous studies of inner-shell ionizations by light ion and heavy ion bombardments. The collision mechanisms have been well understood. In particular, when the ionization is caused by the incidence of heavy particles, one can get the deformed spectra of characteristic X-rays[5,6] or Auger electrons[7~9] which are resolved into a number of satellite peaks. The satellite lines are emitted from the target atom having additively ionized outer-shell electrons. This means that target atoms bombarded with heavy ions are multiply ionized and the fractions of high ionicity are far larger than those by electron and photon excitations.[10,11] If one can extract ionized target atoms from the collision region, highly ionized target ions with

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desired energy are easily obtained, but no one has practised such an ion-excitation experiment. Only the electron- and photon-excitation experiments have been reported by Schmidt *et al.*[12]

In this paper, we present a method of extracting target ions of helium and neon bombarded with protons, alpha particles and nitrogen ions and describe an electronic device of analyzing these ions in a counting mode. The obtained charge spectra are processed by a statistical consideration.

EXPERIMENTAL

The experimental arrangement is shown in Fig. 1. An ion beam extracted from a 4 MV Van de Graaff accelerator of Kyoto University passed through a gas charge stripper, in which the beam was split into ions with various charge states. After analyzing these with a 90° magnet and focusing with a pair of Q-magnets, the desired ion beam was made to pass through a collimating system. The well-defined beam of 0.2 mm in width and 1 mm in vertical length traversed the center of a target chamber placed in a large vacuum chamber.

The target chamber was composed of two parallel electrodes and a gas nozzle, and the incident beam was made to collide with a stream of gas ejected from the gas nozzle of 1 mm in diameter, the top of which was located at 3 mm below the beam axis. An extraction voltage V_{ex} was applied to the electrode A, while a half of this voltage was imposed to the nozzle in order to produce a uniform electric field in the chamber. The ionized target atoms were extracted from a hole of 2 mm in diameter bored at the earth electrode B.

The extracted ions were focused by a de-accelerating lens and entered into an E×B spectrometer called "Wien filter".[13,14] If the recoil energy of the target atoms is neglected, the kinetic energy after extraction is given by

$$K=\frac{1}{2}neV_{ex}=\frac{1}{2}mv^2, \tag{1}$$

where n is the charge state, e is the unit charge, m is the ion mass and v is the ion

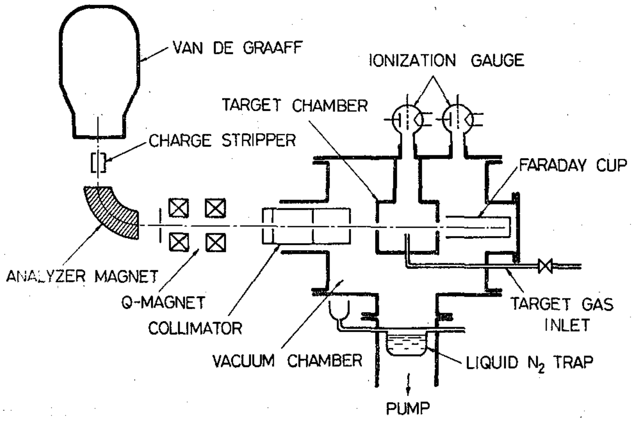


Fig. 1. Experimental arrangement.

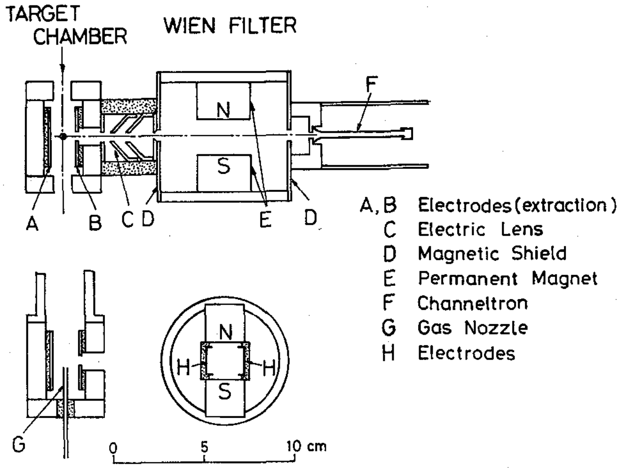


Fig. 2. Target assembly connected with a Wien filter.

velocity. In the Wien filter, the Lorentz force acting on the ion is canceled by an electric force as

$$Bnev = neE, \tag{2}$$

where B and E are the strengths of the magnetic and electric fields which cross perpendicularly to each other in the filter. From Eqs. (1) and (2), we get

$$E = B(neV_{ex}/m)^{1/2}, \tag{3}$$

and this means that the ion charge state can be selected by changing the applied electric field. A permanent magnet was used to produce the B-value of about 0.1 Wb/m^2 (1,000 Gauss). The target assembly connected with the Wien filter is shown in Fig. 2.

In order to get the charge spectrum in a pulse height pattern, two linearly varying electric potentials, $0 \sim +250$ V and $0 \sim -250$ V, were imposed to the electrodes in the Wien filter. The charge-analyzed ions which straightly traverse on the axis of the filter

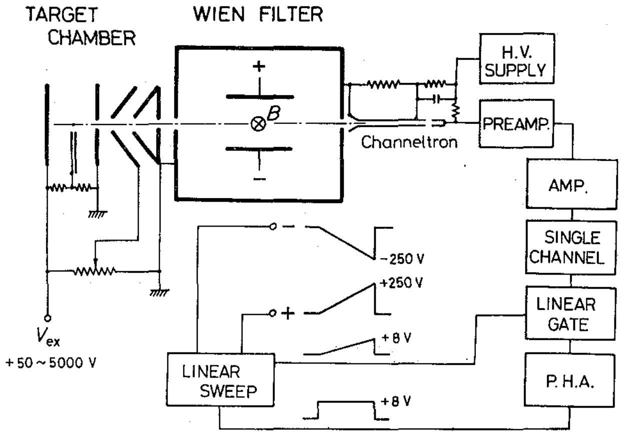


Fig. 3. Block diagram of the electronic circuits for measuring the charge-to-mass ratio spectrum of ionized target atoms in a counting mode.

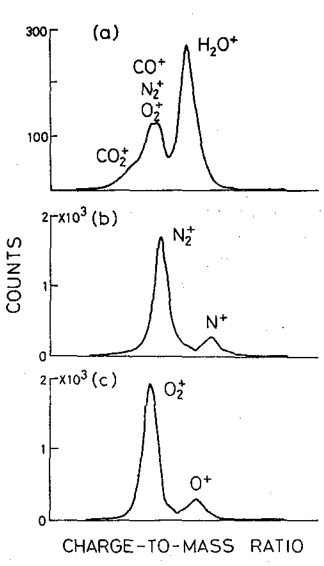


Fig. 4. Charge-to-mass ratio profiles of the residual gas, nitrogen and oxygen molecules excited by 1.0 MeV protons.
(a) residual gas, (b) nitrogen (N_2), (c) oxygen (O_2).
The ordinate is proportional to the square root of charge-to-mass ratio.

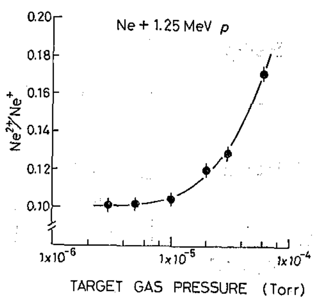


Fig. 5. Yield ratio of Ne^{2+}/Ne^+ as a function of the target gas pressure for neon bombarded with 1.25 MeV protons. *The gas pressure is only a measure to* estimate the pressure at the collision zone.

were detected by a channel electron multiplier (commercially called spiraltron, Bendix SEM-4214), and the signals were fed to a conventional amplifying system. The output signals, after pulse shaping, were mixed with another linear sweep in a linear gate circuit and thus pulses whose amplitudes were proportional to the square root of the charge-to-mass ratio were generated. The block diagram of the whole electronic system is presented in Fig. 3.

A high speed oil diffusion pump equipped with a liquid nitrogen trap was employed to evacuate the vacuum chamber, and the base pressure was less than $\sim 7\times10^{-7}$ Torr. The target gas pressure was monitored by an ionization gauge placed at the top of the target chamber. The beam intensities were 50∼500 nA for light projectile ions and 5∼50 nA for N^{2+} ions, which were measured with a deep Faraday cup arranged just behind the target chamber.

The charge-to-mass profile for the residual gas excited by 1 MeV protons is presented in Fig. 4(a), which indicates a mixture of light molecules if we refer to the nitrogen and oxygen profiles respectively shown in (b) and (c) of the same figure.

RESULTS AND DISCUSSION

A collision-free condition for the created target ions should be kept in the target chamber, otherwise the charge spectrum would be significantly altered before extraction. Therefore, it is necessary to know how the charge profile changes against the target gas

Table. Relative Intensities of Multiply Ionized Atoms for Helium and Neon by the Incidence of Protons, Alpha particles and N^{2+} ions.

Collision	Projectile energy (MeV)	Relative intensity					
		R_1	R_2	R_3	R_4	R_5	R_6
He+p	0. 5	0. 96	0. 041				
	1. 0	0. 97	0. 028				
	1. 5	0. 98	0. 024				
He+α	1. 0	0. 79	0. 21				
	1. 5	0. 78	0. 22				
	2. 0	0. 82	0. 18				
	2. 5	0. 87	0. 13				
He+N^{2+}	1. 0	0. 65	0. 35				
Ne+p	0. 5	0. 81	0. 18	0. 015	—		
	1. 0	0. 87	0. 12	0. 010	—		
	1. 5	0. 90	0. 091	0. 010	—		
Ne+α	1. 0	0. 46	0. 35	0. 16	0. 030	—	
	1. 6	0. 53	0. 32	0. 13	0. 020	—	
	2. 0	0. 59	0. 32	0. 080	0. 011	—	
	2. 4	0. 62	0. 30	0. 069	0. 010	—	
Ne+N^{2+}	1. 0	0. 32	0. 37	0. 23	0. 073	0. 009	—

pressure. Figure 5 shows the intensity ratio Ne^{2+}/Ne^{+} as a function of the gas pressure, where plural collisions clearly increase at more than 1×10^{-5} Torr. Consequently each collision experiment was carried out at below this pressure limit. Note that this pressure corresponds to about 1×10^{-3} Torr at the nozzle exit. The extraction voltage of $V_{ex}=$ 1, 000 V (600 V/cm) was necessary to obtain the saturated charge spectrum, and this implies that ionized target atoms are not fully extracted from the hole of the electrode, if a weak electric field is applied in the target chamber.

The measured charge fractions for helium and neon excited by protons, alpha particles and N^{2+} ions with 0. 5 to 2. 5 MeV in energy are presented in the table, and the representative charge profiles are shown in Figs. 6 and 7, respectively. It is seen that target atoms are more multiply ionized as the projectile mass increases. In the well resolved Ne+N^{2+} spectra of Fig. 7(c), each main peak accompanies a small subpeak, which is attributable to the heavier isotope of neon, that is ^{22}Ne. The yield ratio of the subpeak to main peak is estimated to be 0. 091, and this agrees with the known value of $^{22}Ne/^{20}Ne=0.097$ within the experimental errors.

The relative intensity R_n is defined by

$$R_n=Y_n/\sum Y_n, \tag{4}$$

where Y_n is the yield of target ions with the n-th charge state. The measured R_n values with 10% uncertainty in average are listed in the table. The experimental uncertainties come from the transmission characteristic of the Wien filter and the counting efficiency of the channeltron for different charge and energy of the extracted ions, and the uncertainties become larger for the ions with higher charge state.

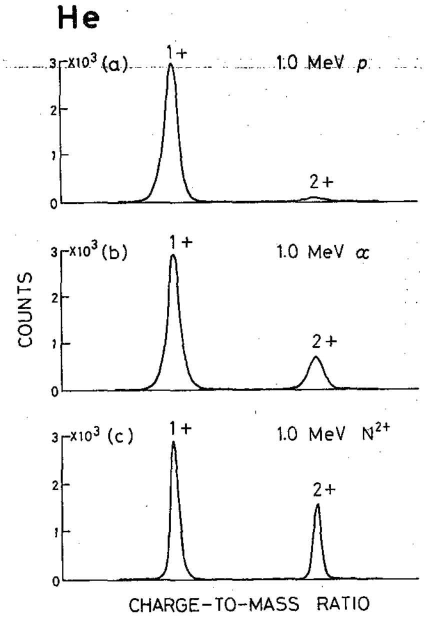


Fig. 6. Representative charge spectra of helium bombarded with protons, alpha particles and N^{2+} ions at 1.0 MeV.
(a) by protons, (b) by alpha particles, (c) by N^{2+} ions.
The ordinate is proportional to the square root of charge-to-mass ratio.

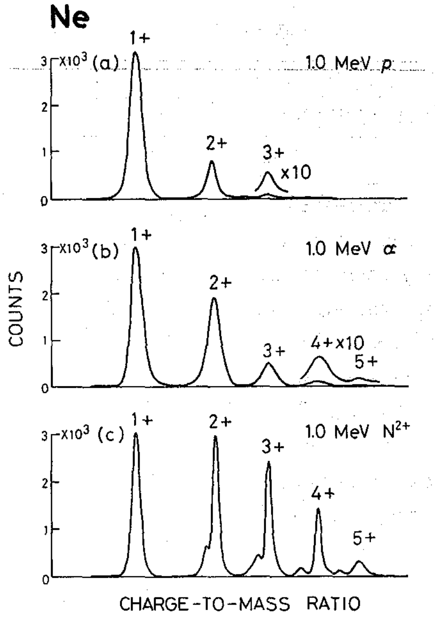


Fig. 7. Representative charge spectra of neon bombarded with protons, alpha particles and N^{2+} ions at 1.0 MeV.
(a) by protons,
(b) by alpha particles,
(c) by N^{2+} ions.

When an electron of the I-th shell of a target atom is ionized by an energetic projectile, the ionization cross section is expressed as

$$\sigma_{\mathrm{I}}=\int 2\pi b\ P_{\mathrm{I}}(d)\,\mathrm{d}b, \tag{5}$$

where b is the impact parameter and $P_{\mathrm{I}}(b)$ is the ionization probability for the I-th shell. If the multiple ionization is treated statistically,[15,16] the cross section for ionizing one of two K-shell electrons and n among eight L-shell electrons is given by

$$\sigma_{1\mathrm{K}n\mathrm{L}}=\int 2\pi b\ {}_2C_1P_{\mathrm{K}}(b)[1-P_{\mathrm{K}}(b)] \times {}_8C_nP_{\mathrm{L}}(b)^n[1-P_{\mathrm{L}}(b)]^{8-n}\mathrm{d}b, \tag{6}$$

where ${}_iC_j$ is the binomial coefficient, $P_{\mathrm{K}}(b)$ and $P_{\mathrm{L}}(b)$ are the single ionization probabilities for the K- and L- shells, respectively. For the ionization of helium, Eq. (6) becomes

$$\sigma_{n\mathrm{K}}=\int 2\pi b\ {}_2C_nP_{\mathrm{K}}(b)[1-P_{\mathrm{K}}(b)]^{2-n}\mathrm{d}b \qquad n=1,\ 2. \tag{7}$$

If the relation $P_{\mathrm{K}}(b)\ll 1$ holds, we have

$${}_2C_2P_{\mathrm{K}}{}^2(1-P_{\mathrm{K}})^0\ll{}_2C_1P_{\mathrm{K}}{}^1(1-P_{\mathrm{K}})^1\ll{}_2C_0P_{\mathrm{K}}{}^0(1-P_{\mathrm{K}})^2\simeq 1,$$

then the expression for neon is

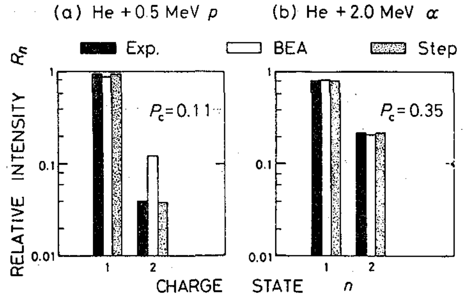
$$\sigma_{nL} \simeq \sigma_{0KnL} \simeq \int 2\pi b \; {}_8C_n P_L(b)^n [1-P_L(b)]^{8-n} db \qquad n=1,\ 2,\ \cdots,\ 8, \tag{8}$$

where $P_L(b)$ is the L-shell ionization probability averaged for the 2s- and 2p-subshells. Since σ_n is proportional to Y_n, the present relative intensity R_n can be expressed by

$$R_n = \sigma_n / \sum \sigma_n. \tag{9}$$

Inner-shell ionization of atoms excited by protons and alpha particles has been studied by many workers, and the observed ionization cross sections have been consistently fitted to the theories of Born approximation (PWBA)[17] and binary encounter approximation (BEA).[18] The BEA theory gives the precise ionization probabilities P_K and P_L as the functions of the impact parameter and the impact velocity.[19,20] The forms of P_K and P_L are both gaussian-like.

In Figs. 8 and 9, the calculated relative intensities for helium and neon, respectively, are compared with the observed intensities at the representative proton and alpha energies. The data for the He+α and Ne+p collisions are fairly well reproduced by the theory, but some discrepancies are seen in the He+p and Ne+α collisions. The BEA



Fg. 8. Observed and calculated relative intensities of ionized helium atoms excited by protons and alpha particles against the charge state. (a) by 0.5 MeV protons, (b) by 2.0 MeV alpha particles.

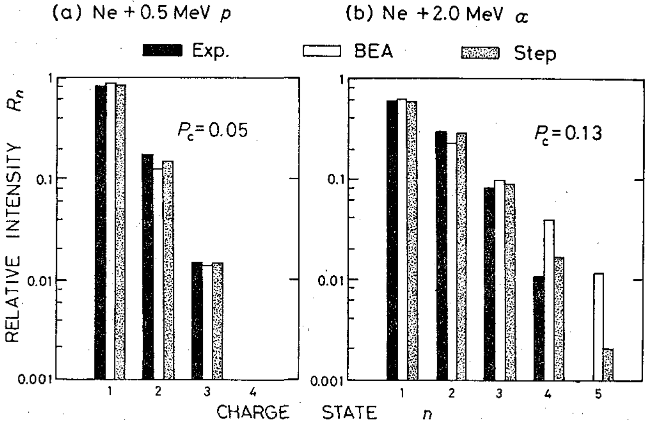


Fig. 9. Observed and calculated relative intensities of ionized neon atoms excited by protons and alpha particles against the charge state. (a) by 0.5 MeV protons, (b) by 2.0 MeV alpha particles.

theory treats the Coulombic ionization of an inner-shell electron whose orbital velocity is comparable with the projectile velocity, that is, $E/\lambda u \sim 1$ where E is the projectile kinetic energy, u is the shell binding energy of the target atom and λ is the projectile mass in units of electron mass. Since the He-K and Ne-L binding energies are a few tens eV, the $E/\lambda u$ values are about 10 for 0.5 MeV proton and 2.0 MeV alpha incidence. Then the practical ionization probabilities would be considerably different from those predicted by the BEA theory. In particular, the He+N^{2+} and Ne+N^{2+} collisions cannot be treated by this theory.

The inner-shell excitation by heavy ion impact has been explained by another theory called "molecular orbital theory" (MO model).[21] As a heavy projectile approaches a target atom, the shell electrons move along the growing molecular orbitals and some electrons are promoted to the higher orbitals leaving inner-shell vacancies after separation. The simplest form of the inner-shell excitation probability in this MO model is a step function,[22] that is, a constant probability within a defined distance of separation as

$$\begin{aligned} P(b) &= P_c \qquad d \leq R \\ &= 0 \qquad d > R, \end{aligned} \tag{10}$$

where R is given by $R \leq \sqrt{2}a$, a being the mean shell radius of the target atom. Then Eqs. (7) and (8) turn to

$$\sigma_K = {}_2C_n P_c^n (1-P_c)^{2-n} \int 2\pi b \, db \qquad n = 1,\ 2 \text{ for helium} \tag{11}$$

$$\sigma_L = {}_8C_n P_c^n (1-P_c)^{8-n} \int 2\pi b \, db \qquad n = 1,\ 2,\ \cdots,\ 8 \text{ for neon.} \tag{12}$$

The relative intensity R_n is simply dependent on only one parameter P_c, which can be estimated by a least squares fitting with the experimental data.

The calculated relative intensities for the best fit values of P_c are indicated in Figs. 8 and 9 in order to compare with the results of the BEA estimation. Since the

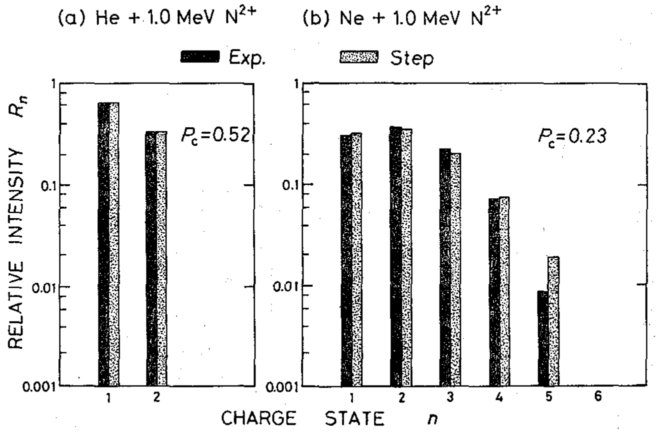


Fig. 10. Experimental and theoretical relative intensities of ionized helium and neon excited by 1.0 MeV N^{2+} ions as a function of the charge state.
(a) helium, (b) neon.

$He+N^{2+}$ collision cannot be fitted to any known ionization theory, the ionization probability is also assumed to be a simple step function. The experimental R_n values for the $He+N^{2+}$ and $Ne+N^{2+}$ collisions at 1 MeV in energy are illustrated in Fig. 10, where the calculated R_n values are compared. Although the observed charge spectra are fairly well reproduced by the BEA and MO theories, one cannot tell at present which theory is preferable.

When a high degree of ionization for neon takes place, a relative contribution of the K-shell ionization increases, which is neglected in the above equations. The K ionized light atoms mostly accompany Auger transition, and in this process one ionicity increases by emitting an Auger electron. This charge state promotion is not included in the equations for neon. However the treatment is quite difficult because highly K ionized atom decays partly via Auger electron emission and partly via X-ray emission and that the decay ratio depends upon the ionicity.

It is important that the magnitude of P_c derived from the fitting procedure with the experiment may not give the correct cross section value if the interaction radius R is fixed. Rather, the R value should be adjusted so as to get the reasonable cross section. Then the interaction radius should be said another parameter in explaining the multiple ionization. A question is that $P_K(b)$, $P_L(b)$, and P_c are always taken invariable when any number of electrons is ionized. Strictly speaking, however, the shell electrons are successively released within about 10^{-16} sec as a projectile passes through a target atom. Then the ionization probability would decrease as the target ionicity increases because the shell binding energy becomes larger for more ionized atom. If such a consideration is introduced into Eqs. (7), (8), (11), and (12), the relative intensities for higher charge states decrease and the experimental profiles in Figs. 9(b) and 10(b) would be better reproduced by the calculations. The quantitative treatment is going on.

From the charge state profiles of Figs. 6 and 7, the initial energy spreads of the target ions are estimated to be within a few eV even in heavy ion-light atom collisions such as the $He+N^{2+}$ case. This implies that the present ionization is caused at a remote distance of internuclear separation where no apperciable recoil energy is given to the target atom. Note that the resolutions are improved in (c) of these figures.

In order to obtain ions of extremely high charge state, it is necessary to employ the collision with a very large P_c value. Figures 8 to 10 tell that very heavy projectiles having at least 0.1 MeV/amu energy are needed for such an ionization, and this means that a new machine able to accelerate argon or krypton ions is desirable. This new accelerator called "METALAC" is under construction at our laboratory.

In conclusion, a simple method is presented of producing multiply ionized heavy ions at very low energies. Various electronic devices to analyze and measure these ions in a counting mode are described. The relative intensities for the charge profiles show approximately binomial distributions, and these are interpreted statistically according to the recent theories in ion-atom collisions. Highly ionized atoms at keV energies are thus extracted by the present experiment, we can carry out the collision studies for these multiply charged ions onto target gases. The preliminary experiment for the $Ar^{3+}+Ar$ collision has shown a successful result.

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In this memorial issue for the retirement of Prof. S. Shimizu, we admire his pioneering works performed in atomic-nuclear fields. More than twenty years ago, he already suggested new experimental studies with the use of low energy accelerators, and one of the ion-atom collision experiments is presented here.

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