Charge state distribution of $^{86}$Kr in hydrogen and helium gas charge strippers at 2.7 MeV/nucleon

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The charge state distributions of krypton ($^{86}$Kr) with an energy of 2.7 MeV/nucleon were measured using hydrogen (H$_2$) and helium (He) gas charge strippers. A differential pumping system was constructed to confine H$_2$ and He gases to a thickness sufficient for the charge state distributions to attain equilibrium. The mean charge states of $^{86}$Kr in H$_2$ and He gases attained equilibrium at 25.1 and 23.2, respectively, whereas the mean charge state in N$_2$ gas at equilibrium was estimated to be less than 20. The charge distributions are successfully reproduced by the cross sections of ionization and electron capture processes optimized by a fitting procedure.

I. INTRODUCTION

The charge states of heavy-ion beams play an important role in the performance of heavy-ion accelerators [1–5]. Figure 1 shows the acceleration scheme of medium-mass ions such as calcium and krypton (Kr) at the RIKEN RI Beam Factory (RIBF) [6,7]. Kr ions are extracted from the RIKEN 18-GHz electron cyclotron resonance (ECR) ion source [8], and they are successively accelerated up to the final energy of 345 MeV/nucleon using the RIKEN heavy-ion linac (RILAC) [9], the RIKEN ring cyclotron (RRC) [10,11], an intermediate-stage ring cyclotron (IRC) [12], and a superconducting ring cyclotron (SRC) [13,14].

Two charge stripper sections are placed downstream of the RILAC and RRC. The initial charge states of the Kr ions extracted from the ECR ion source are 18+ to 20+ depending on the mass numbers of the accelerated Kr ions (78–86).

The Kr beams are accelerated up to 2.7 MeV/nucleon using the RILAC, and they are transported to the first charge stripper located at the exit of the RILAC. Carbon foils (C-foils) with thicknesses in the range of 40–80 μg/cm$^2$ [15] are generally utilized as the first charge stripper to strip the charge of the $^{86}$Kr beam to 26+, which is the lowest acceptable charge state for $^{86}$Kr acceleration as determined by the $K$-value of the subsequent cyclotron RRC ($K = 540$ MeV). The charge states of heavy ions in solid materials have been intensively studied, and a large amount of data for Kr over the energy range of 0.01–100 MeV/nucleon is available in Refs. [16–22].

The stripping energy $E$ at the second charge stripper is $E = 46$ MeV/nucleon. The charge state used for subsequent acceleration has not been determined explicitly, but it should be higher than 32+, which is the lowest acceptable charge state of the SRC ($K = 2600$ MeV). Practically, the charge state for acceleration is determined considering the fraction of the desired charge state and emittance growth after the beam passes through the stripper. The charge state of 34+ is one of the target charge states that can be obtained by using a C-foil charge stripper with a thickness in the range of 0.5–1 mg/cm$^2$ [24]. The fraction of Kr$^{34+}$ is 60–65% depending on the stripper thickness. As mentioned in Ref. [24], the fraction of 36+ attains 80% upon using C-foils with thicknesses over 10 mg/cm$^2$. However, the emittance growth of the stripped beam is estimated to be considerably large, and beam loss would increase beyond the permissible limits.

We focus on the first charge stripper since it locates far upstream from the final cyclotron (SRC) and its lifetime greatly affects the beam availability. Inherently, C-foil

FIG. 1. Acceleration scheme of Kr-ion beams at RIBF. The $^{88}$Kr beams are accelerated by means of a linear accelerator (RILAC) and three cyclotrons (RRC, IRC, and SRC). The first and second charge strippers are located downstream of the RILAC and RRC, respectively.

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Gas charge strippers have been suggested as a possible candidate in the light of their durability. However, as is commonly known, the charge states in gas media attain far lower values than with C-foils because of the density effect [16,25–27]. Figure 2(a) shows the plots of the semi-empirical formulas to predict the mean charge states of $^{86}$Kr in gas media at equilibrium ($Q_{eq}$) divided by the projectile atomic number $Z$ ($Q_{eq}/Z$). The predicted values are plotted as a function of the projectile energy $E$. The figure includes the empirical formulas for predicting charge states in gas media as proposed by Sayer [28] (solid line), Betz [29] (dotted line), Strehl [30] (dashed line), Decrock [31] (dash-dotted line), and Schiwietz [32] (dash-two-dotted line). The predicted values of $Q_{eq}$ vary from 19.7 to 22.5, which are far less than the target value of 26. The data of Kr in gases with $Z \geq 7$, such as nitrogen ($N_2$), neon, or argon, are also plotted using open circles [18,19,33].

Recently, charge strippers using gases with small atomic numbers (low-$Z$) such as hydrogen (H$_2$) or helium (He) have been found to provide considerably higher charge states among gases [34,38]. A datum of the equilibrium charge state $Q_{eq}$ of 26. The data of Kr in gas media as predicted by Sayer [28], Betz [29], Strehl [30], Decrock [31], and Schiwietz [32] respectively. (b) Solid and open diamonds represent the $Q_{eq}$ data of U in low-$Z$ (He) [34] and heavier ($Z \geq 7$) [16,18,19,22,35–37] gas media, respectively.

In the present study, we measured the charge state distributions of $^{86}$Kr at 2.7 MeV/nucleon using H$_2$ and He gas strippers with different thicknesses. Additional data were obtained for the charge state distribution of N$_2$ gas to evaluate the difference between low-$Z$ gases and other gases.

II. GAS CHARGE STRIPPING SYSTEM

We constructed a prototype of a windowless gas charge stripping system in a beam line based on the basic design of the differential pumping system [34,39,40]. Figures 3 and 4 show the overview of the beam line and its detailed diagram, respectively. The gas charge stripper was installed between the X51 section and the X-Rebuncher (XReb). Figure 5 shows the schematic of this system along with differential pumping speeds. The target region was designated as stage 1 and its physical length was 100 cm. The target gas was injected into stage 1, and its two neighboring stages were prepared for differential pumping. The two differentially pumped stages were located both upstream and downstream of stage 1, and they were designated as stages U2 and U3 for upstream and D2 and D3 for downstream, respectively. Stages U3 and D3 were connected to the beam-line chambers X51a and X51b, respectively. A 10-cm-long tube with 4-mm inner diameter was installed between each stage for reducing the gas
throughput outside of the gas target region. Three tubes were located upstream and downstream of stage 1, i.e., the Kr beam passed through a total of six tubes.

The pressure at stage 1 ($P_1$) was measured by two gauges located upstream ($P_{U1}$) and downstream ($P_{D1}$). The pressures at stages U2 (D2) and U3 (D3) are indicated by $P_{U2}$ ($P_{D2}$) and $P_{U3}$ ($P_{D3}$), respectively. We also measured the gas pressures at sections X51, e42, XReb, and J28 (see Fig. 4) and their corresponding pressures were designated $P_{X51}$, $P_{e42}$, $P_{XReb}$, and $P_{J28}$, respectively. The sections X51 and e42 were 1.0 m and 8.0 m upstream from the center of stage 1, respectively, while the sections XReb and J28 were 3.1 m and 7.5 m downstream, respectively.

Since the gas charge stripper is operated at a beam line, the pressure in stages other than stage 1 are expected to fall rapidly when the pressure $P_1$ is maintained constant. The stages U2 and D2 were evacuated by a pair of mechanical booster pumps (MBPs, Edwards, EH500) with a total evacuation speed of 730 m$^3$/h, i.e., 365 m$^3$/h for each stage. The stages U3 and D3 were evacuated by a turbomolecular pump (TMP) with a pumping speed of 1980 m$^3$/h (SHIMADZU EMIT CO., TMP-550L). The chambers X51a and X51b were evacuated by a TMP with a pumping speed of 792 m$^3$/h (Osaka Vacuum, TG220FCAB). The back pumps for the MBPs and TMPs were rotary pumps with an evacuation speed of 4.5 m$^3$/h (Alcatel 2004A).

The target gas was injected through a mass flow controller (MKS, Type 1579A) and gas flow was tuned so that the pressures $P_{U1}$ and $P_{D1}$ reached desired values. $P_{U1}$ and $P_{D1}$ were measured by a baratron transducer (MKS, 627B) and a sapphire capacitance diaphragm gauge (Tem-Tech Lab., SCM2100), respectively. The pressures $P_{U2}$ and $P_{D2}$ were measured by means of MicroPirani™/Piezo loadlock transducers (MKS, 901P). The pressures $P_{U3}$, $P_{D3}$, $P_{X51}$, $P_{e42}$, $P_{XReb}$, and $P_{J28}$ were measured using cold cathode gauges (Pfeifer IKR060) in the low-pressure range (< 0.1 Pa) or measured by constant-temperature-type Pirani gauges (Pfeifer, TPR010) in the higher-pressure region (> 0.1 Pa). Since all pressure values indicated by gauges except for $P_{U1}$ and $P_{D1}$ depended on the gas species, their values were adequately corrected [41].

### A. Offline pressure test

We performed an offline test to evaluate the maximum possible value of $P_1$ with H$_2$ and He gas injection. The results are shown in Figs. 6 and 7 for H$_2$ and He gases, respectively. Pressures $P_{U2}$ ($P_{D2}$), $P_{U3}$ ($P_{D3}$), $P_{X51}$ ($P_{XReb}$), and $P_{e42}$ ($P_{J28}$) were plotted as functions of $P_{U1}$ ($P_{D1}$) in Fig. 6(a) [Fig. 6(b)] and indicated by open circles (open triangles), solid circles (solid triangles), crosses (x-marks), and open squares (solid squares), respectively. The required maximum pressure at the target region was determined by the gas thickness required for the charge states to attain equilibrium. In the case of a C-foil, the most probable...
charge state attains equilibrium at a thickness of 80 μg/cm² [23]. On the assumption that the gas thickness required for equilibrium is the same as that required for equilibrium using carbon, the required maximum pressure for H₂ and He are 1 kPa and 0.5 kPa, respectively, considering the physical length of the gas target region (100 cm). On the other hand, the maximum pressure was limited by the permissible value of the safety interlock for vacuum that is applied during a beam transport operation. The permissible value was 4×10⁻³ Pa, i.e., no beam can be operational for pressures greater than this value. In the case of H₂, P₄₂ and P₂₈ were below 4×10⁻³ Pa when P₁ (=P₁,U₁ ≈ P₁,D₁) was maintained lower than 1.1 kPa. The interlock was applied by the values measured at P₄₂ and P₂₈ in the case of H₂ gas operation. With regard to the case of He, P₂₂ and P₃₃ were sufficiently lower than the admissible value up to P₁ = 1.8 kPa, the interlock was applied by P₅₁ and PXReb and the curves corresponding to P₄₂ and P₂₈ are not plotted in Fig. 7. 

FIG. 5. Schematic of the gas charge stripper with the differential pumping system. Gases are injected into the target region (stage 1) located at the center. The length of the target region is 100 cm. Other stages are also shown along with the pumping speeds of their respective attached pumps. Three tubes with an inner aperture of 4 mm in diameter and length of 10 cm are placed at the entrance and exit of the target region.

FIG. 6. The pressure at each stage is plotted as a function of P₁ for H₂ gas. (a) The pressures at the stages upstream of the gas stripper (P₂₂, P₃₃, P₅₁, and PXReb) are plotted as functions of P₁,U₁ in Fig. 7(a) and indicated by open circles (open triangles), solid circles (solid triangles), and crosses (x-marks), respectively. Since P₅₁ and PXReb were sufficiently lower than the admissible value up to P₁ = 1.8 kPa, the interlock was applied by P₅₁ and PXReb and the curves corresponding to P₄₂ and P₂₈ are not plotted in Fig. 7. (b) The pressures downstream of the gas stripper (P₂₂, P₃₃, PXReb, and P₂₈) as a function of P₁,U₁.
The intensities of the stripped beams were measured using a corrected efficiency of 3%. The magnetic field of the DMJ3 was collimated by these six narrow tubes with a total length of 2 m, which corresponded to a beam transmission efficiency of 3%.

The charge state was analyzed by means of the dipole magnet DMJ3. The magnetic field of the DMJ3 was corrected by considering the energy loss in the gas. The intensities of the stripped beams were measured using a Faraday cup (FC-J33) downstream of DMJ3.

A pair of plastic scintillation counters were used to measure the energy of the beam via the time-of-flight (TOF) technique [42]. They were located at sections J41 (SC-J41) and S42 (SC-S42), as shown in Fig. 4. The flight path length between SC-J41 and SC-S42 was 7.0212 m.

**III. EXPERIMENTS**

**A. Krypton beam acceleration and transport**

A series of experiments was performed at the RIKEN RIBF in February 2012 and February 2013 by accelerating $^{86}$Kr beams up to 2.7 MeV/nucleon. The $^{86}$Kr beams were accelerated using the RILAC at a rf-frequency of 36.5 MHz. The $^{86}$Kr$^{20+}$ ions with an intensity of 11–12 eμA were provided by the 18-GHz ECRIS and were transported to the gas charge stripper. The incident beam intensities were measured to be 650–750 eμA (33–38 pA) with an attenuation of 1/10 using a Faraday cup (FC-X51) placed upstream of the gas charge stripper. The beam spot size was 5 mm in diameter at the just upstream of the system. Since this gas stripping system was a prototype, we had no focusing element such as quadrupole or solenoid magnet to transport the beam adequately through the system. Moreover, as written in Sec. II, six tubes with an inner diameter of 4 mm were used for reduction of gas throughput to enhance the differential pumping efficiency in a limited space. Therefore, the beam was collimated by these six narrow tubes with a total length of 2 m, which corresponded to a beam transmission efficiency of 3%.

The charge state was analyzed by means of the dipole magnet DMJ3. The magnetic field of the DMJ3 was corrected by considering the energy loss in the gas. The intensities of the stripped beams were measured using a Faraday cup (FC-J33) downstream of DMJ3.

**B. Gas thickness calibration**

In order to determine the effective thickness of the gas target region, the relationship between $P_1$ and the energy loss of the beams was determined by means of the TOF technique using the pair of scintillation counters SC-J41 and SC-S42. The time differences between the signals of one scintillation counter and rf signals sampled by 1/6 were measured at SC-J41 and SC-S42. Figures 8(a) and (b) show the typical time spectra of the SC-J41 and SC-S42, respectively, when using a C-foil with a thickness of 10 μg/cm². Each spectrum has six peaks, and the time difference between adjacent peaks $\tau_{rf}$ is 27.4 ns (= 1/f, where $f$ denotes the rf-frequency $f = 36.5$ MHz). The error bars in the spectra represent the statistical uncertainty. Each peak was fitted by a Gaussian function (solid curve) to determine the center of the peak. The arrival times at each scintillation counter were defined as these central values. The arrival times at the SC-J41 and SC-S42 are denoted by $t_{J41}$ and $t_{S42}$, respectively. The TOF of the beam $t_{TOF}$ is written as

$$t_{TOF} = k \tau_{rf} + (t_{J41} - t_{S42}).$$

where $k = 11$ denotes the wave number of the bunch clock. The velocity of the beam is calculated from the expression $v = L/t_{TOF}$, where $v$ and $L$ denote the velocity of the beam and the flight path length between SC-J41 and SC-S42 (7.0212m), respectively. The kinetic energy $E$ of the beam is calculated from the relation $\gamma = 1 + E/(muc^2)$, where $\gamma$,

![FIG. 7. The pressure at each stage is plotted as a function of $P_1$ for He gas. (a) The pressures at the stages upstream of the gas stripper ($P_{U1}$, $P_{D1}$, and $P_{S1}$) as a function of $P_{U1}$. (b) The pressures downstream of the gas stripper ($P_{D2}$, $P_{D3}$, and $P_{XReb}$) as a function of $P_{D1}$. Those at $P_{D2}$ and $P_{D3}$ are not plotted since the pressures $P_{S1}$ and $P_{XReb}$ are below the permissible value of $4 \times 10^{-3}$ Pa when maintaining $P_1$ up to 1.8 kPa.](image)

![FIG. 8. Typical TOF spectra of (a) SC-J41 and (b) SC-S42 for a C-foil with a thickness of 10 μg/cm². Gaussian functions are fitted to each peak to determine the center (arrival time).](image)
$m_u$ and $c$ denote the Lorentz factor, atomic mass unit of 931.5 MeV/c$^2$, and light velocity, respectively.

First, the TOF spectra were measured using C-foils with different thicknesses to determine the energy of the incident beam. The maximum magnetic rigidity of the magnet DMJ3 was 0.96 T m (1.2 T × 0.8 m), which meant that a charge state of 21+ or higher was required to bend $^{86}$Kr beams at 2.7 MeV/nucleon. Therefore, the incident $^{86}$Kr$^{20+}$ beam could not be transported beyond the DMJ3, and consequently, $^{86}$Kr$^{24+}$ beams were transported to SC-J41 and SC-S42 after stripping by a C-foil with known thickness values of 10, 40, and 80 $\mu$g/cm$^2$. Figure 9 shows the beam energies after exiting the C-foils as a function of the C-foil thickness. The error bars represent the uncertainties of the energy and the errors are attributed to the uncertainty of the arrival time obtained in the fitting procedure. The incident beam energy was estimated to be 2.672 ± 0.019 MeV/nucleon by extrapolation of the fitted linear function to zero thickness.

The spectra were obtained in the cases of H$_2$, He, and N$_2$ gas injections at different values of $P_1$. The energy loss was calculated from the difference between the case of no gas injection and the case of gas injection. The thickness of the gas medium was estimated by energy loss calculations using the ATIMA [43] package installed in LISE++. The relationships between the gas thickness and the gas pressure $P_1$ are shown in Fig. 10. The data for H$_2$, He, and N$_2$ gas injections are represented by solid circles, open circles, and solid triangles, respectively. The fitting functions are represented by solid, dotted, and dashed lines for H$_2$, He, and N$_2$ gas injections, respectively.

The uncertainties in thickness include the errors of the beam energy with gas injections along with the uncertainties of the incident beam energy. The data were fitted by linear functions represented by solid, dotted, and dashed lines for H$_2$, He, and N$_2$ gas injections, respectively. The thickness of the gas charge stripper was calculated from these functions. The background pressure in the case of no gas injection was 3.6 Pa, which corresponded to an uncertainty of $3 \times 10^{-3}$ MeV/nucleon at most in comparison with the pressures of H$_2$ or He. This background-pressure contribution is considered negligible in this study.

**IV. RESULTS**

**A. Charge state distribution**

The charge state distributions of $^{86}$Kr measured using H$_2$ and He gas charge strippers with different thicknesses are shown in Fig. 11. The data of the fractions calculated for (a) H$_2$, (b) He, and (c) N$_2$ are plotted in the figure. The data for H$_2$ gas with thicknesses of 10.3, 22.7, 45.6, 67.7, and 106.8 $\mu$g/cm$^2$ are denoted by asterisks, open triangles, open circles, open squares, and open diamonds, respectively. The data for He gas with thicknesses of 15.7, 29.3, 59.2, 124.0, and 247.1 $\mu$g/cm$^2$ are denoted by asterisks, open triangles, open circles, open squares, and open diamonds, respectively. The data for N$_2$ gas with thicknesses of ~10, ~40, ~800, and ~1200 $\mu$g/cm$^2$ are denoted by asterisks, open triangles, open circles, open squares, and open diamonds, respectively. As shown in Fig. 11(c), the complete range of charge state distributions in N$_2$ gas was not obtained because of the magnetic rigidity of the analyzing DMJ3 magnet. Charge state distributions at energies considerably lower than 2.7 MeV/nucleon were obtained below 20+, for which the corresponding N$_2$ gas thickness values were more than 800 $\mu$g/cm$^2$. However, these thickness values had a large uncertainty of more than 30%. Therefore, the charge distribution in N$_2$ gas at equilibrium was not obtained correctly.
The charge state distribution of $^{86}$Kr in H$_2$, He, and N$_2$ gas is plotted in Fig. 11. The charge fraction of H$_2$, He, and N$_2$ gases are denoted by asterisks, open triangles, and open circles, respectively. The data for H$_2$ gas with thicknesses of 10.3, 22.7, 45.6, 67.7, and 106.8 $\mu$g/cm$^2$ are denoted by asterisks, open triangles, open circles, open squares, and open diamonds, respectively. The data for He gas with thicknesses of 15.7, 29.3, 59.2, 124.0, and 247.1 $\mu$g/cm$^2$ are denoted by asterisks, open triangles, open circles, open squares, and open diamonds, respectively. The data for N$_2$ gas with thicknesses of ~10, ~40, ~800, and ~1200 $\mu$g/cm$^2$ are denoted by asterisks, open triangles, open circles, and open squares, respectively. The charge distribution in N$_2$ gas at equilibrium was not obtained accurately.

The fraction of the charge state $q_i$ was calculated as described in a previous study [36]; this charge state fraction is defined as

$$ F(q_i) = \frac{1}{N} \frac{I_{333}/q_i}{I_{351}/q_{ini}}, $$

where $N$ denotes a normalization constant, $I_{351}$ and $I_{333}$ denote the beam intensities measured at FC-X51 and FC-J33, respectively, and $q_{ini}$ denotes the incident charge state of 20+. The index $i$ extends over the range of possible charge states of 0–36. In order to determine $N$, an unnormalized fraction $f(q_i) = \frac{I_{333}/q_i}{I_{351}/q_{ini}}$ was first calculated. The data set of $f(q_i)$ is fitted by a Gaussian function. However, the beam transmission efficiency was only 3% in this study, and the beam intensity measured with each Faraday cup includes its own offset value; hence, the sum $\sum f(q_i)$ over all $i$ does not equal unity. Therefore, the area of the fitted Gaussian function is calculated and introduced as the normalization constant $N$ in order to obtain the sum of $F(q_i)$ as unity. Subsequently, the set of $F(q_i)$ is fitted by a Gaussian function again. The error bars in the fraction data $F(q_i)$ in Fig. 11 are attributed to the uncertainty of the normalization constant $N$, which is arising from the errors of the fitting parameters.

The most probable charge states are defined as the central values obtained via the fitting procedure. However, as the gas thickness increases, the discrepancy between $F(26+)$ and $F(27+)$ is significant, as clearly shown in Fig. 11(a). It is known as the shell effect, first observed by Mouk et al. [45], and also observed in the case of uranium stripping [34]. This discrepancy is caused by the difficulty of ionizing from the L shell of the Kr ion. Ionization potentials to remove one M-shell electron from Kr$^{25+}$ and one L-shell electron from Kr$^{26+}$ are 1.2 keV and 2.9 keV, respectively [46]. The electron traveling with projectile with kinetic energy of 2.7 MeV/nucleon ($\beta = 0.076$) has a kinetic energy of 1.5 keV. It is difficult to strip electrons with removal energies higher than 1.5 keV [47,48]. This fact also implies that excited states have little contribution in ionization process in gases [45]. Thus, the standard Gaussian function cannot reproduce the charge distribution. Consequently, an asymmetric parameter $e_a$ is introduced to improve the fitting results, as described in Ref. [28]. The corresponding asymmetric Gaussian function is written as

$$ F_a(q) = F_{mp} \exp \left[ \frac{q^2}{2(1 + e_a \sigma^2)} \right], $$

$$ t = \frac{q - q_{mp}}{\sigma}, $$

where $q$, $q_{mp}$, $F_{mp}$, and $\sigma$ denote the charge state, the most probable charge state, fraction at the most probable charge state, and distribution width, respectively. Fitting by both standard and asymmetric Gaussian functions were applied in the case that the fraction values $F(26+)$ and $F(27+)$ were both greater than zero. The asymmetric Gaussian was adopted when the reduced chi-square value became smaller than the result of the standard Gaussian. The fitted parameters for the H$_2$ and He charge strippers are listed in Table I along with their uncertainties. The thickness is calculated for H$_2$, He, and N$_2$ gases by the linear functions obtained in Sec. III B. The errors in thickness are attributed to the uncertainties of the fitting parameters. The most probable charge states correspond to the mean charge states in the case of the standard Gaussian function for which the asymmetric factor $e_a = 0$. The mean charge states are slightly less than the most probable charge states if the asymmetric factor is less than zero. We found that the mean charge states in H$_2$ and He gases attain equilibrium at 25.1 and 23.2, respectively. The charge state of $^{86}$Kr in N$_2$ at equilibrium was estimated to be less than 20+. Further, we could obtain a Kr$^{26+}$ beam with a charge fraction of 31% in H$_2$ for a thickness of 68 $\mu$g/cm$^2$; thus, H$_2$ gas is a good candidate for Kr-ion acceleration. For practical usage, the amount of the fraction $F(26+)$ is sufficient at the H$_2$ gas thickness of 70 $\mu$g/cm$^2$, which corresponds to the required P$_1$ pressure of 0.7 kPa. On the assumption that the aperture of 10 mm in diameter can be applied to practical operation,
the required pumping speeds should be greatly enhanced to maintain the pressure at each stage as same as this study. Especially, the required pumping speed for the second stage is estimated to be over 10000 m$^3$/h. As a realistic solution, the second stage would be divided into two stages (stage 2 and 3) evacuated by MBPs, and hence the next stage (stage 4) would be evacuated by TMP. The required pumping speeds for stages 2, 3, and 4 are 4000 m$^3$/h (MBP), 2000 m$^3$/h (MBP), and 5400 m$^3$/h (TMP), respectively. However, since the full length of the system becomes long, the suitable design of a beam transport is also needed.

The equilibrium charge states in H$_2$ and He are plotted in Fig. 12 along with the data shown in Fig. 2. The solid and open squares represent the $Q_{eq}/Z$ values of H$_2$ and He, respectively, as obtained in this study. The x-mark represents the $Q_{eq}/Z$ value for N$_2$ on the assumption that $Q_{eq}$ in N$_2$ equals 20+. The $Q_{eq}/Z$ values were enhanced by 0.14 and 0.09 in H$_2$ and He gases, respectively, when compared with the case of N$_2$ gas. Their enhancements of $Q_{eq}$ can be larger since $Q_{eq}$ in N$_2$ is actually estimated to be smaller (i.e., $Q_{eq}/Z$ for N$_2$ will shift along the direction of the arrow in Fig. 12).

**V. CHARGE STATE CALCULATIONS**

In general, the reactions causing a change in the charge are divided into two categories: ionization and electron capture. The equilibrium charge state is determined by the competing processes of ionization and electron capture. Ionization can result from collisions between projectile electrons and the target nuclei or bound electrons. Electron capture can occur by the transfer of electrons from the target atoms to the projectile. The details regarding the calculations of the ionization and electron capture cross sections are summarized in Ref. [49], and further, the consistency between the theoretical and experimental cross sections is also demonstrated. In this section, we use the CGS system of units following the description in Ref. [49].

**A. Ionization reactions**

A semi-classical approach to Born-based calculations that is classified under the set of binary encounter model (BEM) has been developed for simplified application to ionization calculations. These models were proposed first by Gryzinski [50] and Garcia [51]. Additional work toward their development was performed by Vriens [52]. Calculations based on this model employ a simplified two-body Coulomb scattering cross section for the target nuclear charge collision with the projectile electrons. The cross section for the ionization of an electron in the $nl$ shell ($nl$-electron) in the projectile is defined as
\[ \sigma_{nl} = 4\pi a_0^2 \left( \frac{Ry}{I_{nl}} \right)^2 (Z_t^2 + Z_t) G(V), \]  
\[ G(V) = \frac{\alpha^{3/2}}{V^2} (1 - \beta)(1 - \beta^4 + V^2) \left\{ \alpha + \frac{2}{3} (1 + \beta \ln(2.7 + V)) \right\}, \]  
\[ \alpha = \frac{V^2}{1 + V^2}, \quad \beta = \frac{1}{4V(1 + V)}; \]  
for \( V < 0.206 \), \( G(V) \) is simply \( G(V) = \frac{4V^4}{15} \).  

The constant \( 4\pi a_0^2 \text{Ry}^2 \) gives \( 4\pi a_0^2 \text{Ry}^2 = 6.51 \times 10^{-14} \) (cm\(^2\) eV\(^2\)), where Ry denotes the Rydberg unit of energy \( \text{Ry} = 13.6 \) (eV) and \( a_0 \) the Bohr radius \( a_0 = 5.29 \times 10^{-10} \) (cm). The parameter \( I_{nl} \) represents the ionization potential experienced by the \( nl \)-electron in the projectile, and \( Z_t \) denotes the target nuclear charge. The values of \( I_{nl} \) are available at the NIST homepage [46]. The term \( Z_t \) denotes the effective screened target nuclear charge as perceived by the individual \( nl \)-electron in the projectile. For the sake of simplicity, \( Z_t \) is temporarily denoted by \( \bar{Z}_t(b_{nl}) \), where \( b_{nl} \) denotes an impact parameter experienced by the projectile \( nl \)-electron. The expression for \( \bar{Z}_t(b_{nl}) \) is given by  
\[ \bar{Z}_t(b_{nl}) = Z_t - \int_0^{b_{nl}} n(r) d^3r, \]  
where \( n(r) \) denotes the electron density about the target nucleus at a given radius \( r \). The simplified approach to obtain \( n(r) \) is discussed in Ref. [49], and from the approach, we have  
\[ n_n(r) = \frac{C_n}{r} \exp \left( \frac{-r}{B_n} \right), \]  
where \( n_n(r) \) denotes the density of electrons in shell \( n \) (\( n \)-electrons), and \( C_n \) and \( B_n \) denote the parameters determined for each shell \( n \). The parameters \( C_n \) and \( B_n \) are expressed using the total number of \( n \)-electrons, \( N_n \), as below  
\[ N_n = 4\pi C_n B_n^2. \]  
\( B_n \) can be interpreted as the median radius of a given shell \( n \), and it is obtained from the definition in Ref. [49],  
\[
\sigma_{nl} = \frac{n^2 a_0^2}{Z_t - S_n},
\]
where \( S_n \) denotes the screening value for the \( n \)-electrons in the target. \( S_n \) is determined as  
\[ S_n \approx f_i N_i + f_n N_n + f_o N_o, \]  
where \( f_i \), \( f_n \), and \( f_o \) denote the fractions of the inner electrons \( N_i \), \( n \)-electrons \( N_n \), and outer electrons \( N_o \) enclosed within \( B_n \), respectively. The values of \( f_i \), \( f_n \), and \( f_o \) applied in Ref. [49] are 0.90, 0.264, and 0.10, respectively. Consequently, here, the total electron density is given by  
\[ n(r) = \sum_n n_n(r). \]  
The relation between the impact parameter and ionization potential is given by  
\[ I_{nl} = \bar{Z}_t(b_{nl}) e^2 \frac{e^2}{b_{nl}}, \]  
where \( e \) denotes the elementary charge. The impact parameter \( b_{nl} \) is obtained by solving Eqs. (9) and (15) for \( b_{nl} \). We derive \( \bar{Z}_t(b_{nl}) \) by substituting the obtained \( b_{nl} \) value in Eq. (9).

Following Bethe [53] and Scheidenberger et al. [54], the contribution of the target-bound electrons to the total ionization cross section can be incorporated by using the term \( \bar{Z}_t^2 + Z_t \) in Eq. (5). The ionization cross sections are functions of the square of the ionizing charge. The term \( \bar{Z}_t^2 \) represents an interaction between the \( nl \)-electron in the projectile and the ionizing charge \( \bar{Z}_t \). In the case of interaction between the \( nl \)-electron in the projectile and an electron in the target atom, the square of the electron charge is unity, but it contributes by a factor of \( Z_t \) to the cross section. The total ionization cross section \( \sigma_{BEM} \) can be calculated by summation over all \( nl \)-electrons in the projectile as  
\[ \sigma_{BEM} = \sum_{nl} \sigma_{nl}. \]  

B. Electron capture reactions

The capture of an electron in a target by a projectile ion can occur either by a radiative or a nonradiative reaction. Radiative capture reactions dominate over nonradiative capture in the energy range higher than 100 MeV/nucleon [54]. In the energy region studied here, the cross sections corresponding to the radiative process contribute less than \( 10^{-2} \) of the total capture cross section. The significant nonradiative process for target electron capture by a projectile is the charge transfer reaction. The first model of the charge transfer reaction is attributed to
Oppenheimer [55], and it was later refined by Brinkman and Kramers [56]. The final form is known as the OBK model. This charge transfer reaction rate depends on the initial energy $E_i$ of the bound electron in the target and its final energy $E_f$ in the projectile ion, where $E_i$ and $E_f$ denote the binding energy in the initial and final states, characterized by the quantum numbers $n_i$ and $n_f$, respectively. The charge transfer cross section is expressed by

$$
\sigma_{\text{OBK}} = 4.1 \times 10^4 \sum_{n_i} \sum_{n_f} N_i a_{\text{eik}}
$$

$$
\times \frac{Q^2 a^4 q e^4}{E_k^2 E_i^2 (E_i + E_f)^2} \left\{ E_k^2 + 2E_i(E_i + E_f) + (E_i - E_f)^2 \right\}^{\frac{3}{2}}
$$

in which $E_k = m_e v_e^2/2$, where $m_e$ and $v_e$ denote the electron mass and velocity, respectively, $Q$ the projectile charge state, and $N_i$ the number of electrons in the original target shell with a quantum number $n_i$. The eikonal factor $a_{\text{eik}}$ accounts for reduction in this reaction with increased deviation from the Born approximation on which it is based. This factor is expressed as

$$
a_{\text{eik}} = \frac{\pi \eta v_i}{\sinh(\pi \eta v_i)} \exp \left[ -2\eta v_i \arctan \left( \frac{\eta}{v_i} \right) \right]
$$

where $\eta = a_0 c/\nu$ ($a_0$: fine-structure constant), $\nu = (E_f - E_i)/\text{Ry}$, and $v_i = \sqrt{E_i/\text{Ry}}$. The total charge transfer reaction rate is obtained by summing over all the initial electron bound states in the target and all final captured states in the projectile.

### C. Relation between cross sections and equilibrium charge states

The total ionization $\sigma_{\text{BEM}}$ and electron capture $\sigma_{\text{OBK}}$ cross sections are plotted as functions of the projectile charge state in Fig. 13. The total ionization and electron capture cross sections of Kr in $\text{H}_2$ gas are represented by solid and dash-two-dotted curves, respectively. The ionization and electron capture cross sections in $\text{He}$ gas are represented by dash-dotted and dotted curves, respectively. The charge states at the intersection point for $\text{H}_2$ and $\text{He}$ are 25.01 and 23.43, respectively. Ionization and electron capture cross sections optimized to fit the experimental data of mean charge states ($\sigma_{\text{BEM}}$ and $b\sigma_{\text{OBK}}$) are also plotted as a function of the projectile charge states. The solid and long-dashed curves indicate the cross sections multiplied by $a_{\text{He}} = 0.99$ and $b_{\text{He}} = 0.75$ for $\text{H}_2$. The dash-dotted and dotted curves represent cross sections multiplied by $a_{\text{He}} = 1.00$ and $b_{\text{He}} = 1.08$ for $\text{He}$. The charge states at the intersection point are 25.33 and 23.28 for $\text{H}_2$ and $\text{He}$ gases, respectively.

#### 1. Evolution of charge state distribution

When an ion beam passes with a velocity $\nu$ through a target of thickness $t$, the charge state composition of the beam varies due to ionization and electron capture events. In our study, the fractions of the charge state $q$ were defined as the function $Y_q(t)$ at each thickness value $t$. The fractions $Y_q(t)$ obey a system of linear coupled differential equations,

$$
\frac{dY_q(t)}{dt} = \sum_{q' < q} \sigma(q', q) Y_{q'}(t) - \sigma(q, q') Y_q(t),
$$

where $\sigma(q, q')$ denotes the cross sections for changing the charge state from $q$ to $q'$. The summation in Eq. (21) is extended by $q$ and $q'$ over the range of possible charge
states. The fractions $Y$ are normalized by $\sum_q Y_q = 1$. Here, with the assumption that the charge changing process occurs via single-electron loss or capture, i.e., $|q' - q| = 1$, Eq. (21) yields the following simple relation upon using the notations $\sigma^{\text{ion}}$ and $\sigma^{\text{cap}}$,

$$\frac{dY_q}{dt} = \sigma^{\text{ion}}(q-1,q)Y_{q-1}(t) - \{\sigma^{\text{cap}}(q,q-1) + \sigma^{\text{ion}}(q,q+1)\}Y_q(t) + \sigma^{\text{cap}}(q+1,q)Y_{q+1}(t).$$  \tag{22}$$

Equation (22) is integrated over the thickness $t$ by the Runge-Kutta-Gill method, and the mean charge state $<q(m)>$ is calculated as $<q(m)> = \sum_q q Y_q$ for each thickness listed in Table I. The parameter $\chi^2$ is defined as $\chi^2 \equiv \sum_{i=1}^{m} \left( \frac{\sigma^{\text{cal}}_i - \sigma^{\text{exp}}_i}{\delta_i} \right)^2$, where $\sigma^{\text{cal}}_i$ and $\delta_i$ are the experimentally obtained mean charge states listed in Table I and their uncertainties, respectively, and index $i$ ranges from 1 to the total number of data points ($N_{\text{exp}} = 5$ for both H$_2$ and He gas measurements).

The parameters $a$ and $b$ in Eqs. (19) and (20) were fitted such that the $\chi^2$ value becomes minimum. The obtained values of $a$ and $b$ were 0.99 and 0.75 for H$_2$ and 1.00 and 1.08 for He, respectively. The optimized ionization and electron capture cross sections are shown in Fig. 13. The ionization and electron capture cross sections are multiplied by $a_{\text{H}_2} = 0.99$ and $b_{\text{H}_2} = 0.75$ for H$_2$, and $a_{\text{He}} = 1.00$ and $b_{\text{He}} = 1.08$ for He gas, respectively. The solid and long-dashed curves in Fig. 13 indicate the optimized cross sections for H$_2$. The dash-dotted and dashed curves in Fig. 13 indicate those for He. The charge states at the point of intersection are 25.33 and 23.28 for H$_2$ and He gas, respectively.

Figure 14 shows the mean charge states plotted as a function of the gas thickness. The horizontal error bars represent the thickness uncertainty listed in Table I. The uncertainties in the mean charge state are smaller than the symbols. The calculation results obtained using the optimized parameters $a$ and $b$ are also indicated by solid and dotted curves for H$_2$ and He gases, respectively. The calculation results obtained using the optimized parameter sets ($a_{\text{H}_2}, b_{\text{H}_2}$) and ($a_{\text{He}}, b_{\text{He}}$) are represented by solid and dotted curves for H$_2$ and He gases, respectively. In addition, the data corresponding to the C-foil [23] are also indicated by solid triangles for comparison.

The charge state distributions reproduced by calculations using the optimized cross sections are shown in Figs. 15 and 16 for H$_2$ and He, respectively. The experimental data are also shown, and from the figure, we note that the calculation results successfully reproduce the fraction data. Figures 15(a), (b), (c), (d), and (e) show the charge distributions in H$_2$ for thicknesses of 106.8, 67.7, 45.6, 22.7, and 10.3 $\mu$g/cm$^2$, respectively. The solid circles represent the data shown in Fig. 11(a). The solid lines represent the calculated results with optimized parameters $a_{\text{H}_2} = 0.99$ and $b_{\text{H}_2} = 0.75$. Figures 15(a), (b), (c), (d), and (e) represent the charge distributions in He for thicknesses of 247.1, 124.0, 59.2, 29.3, and 15.7 $\mu$g/cm$^2$, respectively. The solid circles represent the data shown in Fig. 11(b). The solid lines represent the calculated results with the optimized parameters $a_{\text{He}} = 1.00$ and $b_{\text{He}} = 1.08$. 

![Figure 14](image_url)  
**FIG. 14.** Mean charge states of $^{86}$Kr plotted as a function of gas thickness. The data for H$_2$ and He gases are represented by solid and open circles, respectively. The data corresponding to the C-foil stripper are also indicated by solid triangles. The calculation results obtained using the optimized parameter sets ($a_{\text{H}_2}, b_{\text{H}_2}$) and ($a_{\text{He}}, b_{\text{He}}$) are represented by solid and dotted curves for H$_2$ and He gases, respectively.

![Figure 15](image_url)  
**FIG. 15.** The charge state distribution of $^{86}$Kr in H$_2$ for various thicknesses. The charge distributions at thicknesses of (a) 106.8, (b) 67.7, (c) 45.6, (d) 22.7, and (e) 10.3 $\mu$g/cm$^2$ are indicated by solid circles along with the calculated fractions (solid lines).
The relation, can be partially presented by exponential form:

\[ F(q_0 + 1) = \frac{\sigma_{\text{ion}}^0}{\sigma_{\text{cap}}^0} \exp(-c_i), \]  

\[ F(q_0 - 1) = \frac{\sigma_{\text{cap}}^0}{\sigma_{\text{ion}}^0} \exp(-c_c). \]  

On the assumption that the charge distribution has standard Gaussian form, the relation \( F(q_0 + 1)/F(q_0) = F(q_0 - 1)/F(q_0) \) can be obtained. We derive

\[ \sigma_{\text{ion}}^0 = \sigma_{\text{cap}}^0 \exp \left[ \frac{1}{2} (-c_i + c_c) \right]. \]  

Substituting Eq. (29) into Eq. (27),

\[ \frac{F(q_0 + 1)}{F(q_0)} = \exp \left[ -\frac{1}{2}(c_i + c_c) \right]. \]  

On the other hand, the fraction of each charge state is calculated by a Gaussian function,

\[ F(q) = F(q_0) \exp \left[ -\frac{(q - q_0)^2}{2\sigma_{\text{cal}}^2} \right], \]  

where \( \sigma_{\text{cal}} \) denotes the calculated width. For \( q = q_0 + 1 \), we obtain the relation

\[ \frac{F(q_0 + 1)}{F(q_0)} = \exp \left[ -\frac{1}{2\sigma_{\text{cal}}^2} \right]. \]  

Comparing Eqs. (30) and (32), we obtain \( \sigma_{\text{cal}} \) as

\[ \sigma_{\text{cal}} = \sqrt{\frac{1}{c_i + c_c}}. \]  

The \( \sigma_{\text{cal}} \) is calculated assuming that each mean charge state listed in Table I is considered as \( q_0 \). Firstly, \( \sigma_{\text{ion}} \) and \( \sigma_{\text{cap}} \) in the forms of Eqs. (23) and (24) are calculated for \( q_0 \) from the cross sections optimized in Sec. V C 1. The parameters \( c_i \) and \( c_c \) are obtained by the fitting to the cross sections around \( q_0 \). Then the \( \sigma_{\text{cal}} \) is calculated from Eq. (33). Figure 17 shows the distribution width \( \sigma \) in Table I along with \( \sigma_{\text{cal}} \) as a function of the gas thickness. The data for H\(_2\) and He gases are represented by solid and open circles, respectively. The calculation results of \( \sigma_{\text{cal}} \) obtained using the optimized parameter sets (\( a_{\text{H}_2}, b_{\text{H}_2} \)) and (\( a_{\text{He}}, b_{\text{He}} \)) are represented by solid and dashed curves with asterisks and x-marks for H\(_2\) and He gases, respectively. The values of \( \sigma_{\text{cal}} \) for H\(_2\) well reproduced the measured widths, although the calculation is based on the assumption that the charge state distribution attains at equilibrium. Those of He gas are in agreement within an accuracy of 0.2 at the thickness sufficient for equilibrium. It is found that the distribution width at the equilibrium can be reproduced by the accurate
The optimized scaling factors were optimized such that the calculation of the cross sections calculated by using the BEM and OBK model were optimized such that the calculation of the charge evolution as described in ionization and electron capture cross sections, without calculations of the charge evolution as described in Sec. V C 1.

VI. SUMMARY AND CONCLUSIONS

We measured the charge state distributions of $^{86}$Kr at 2.7 MeV/nucleon by using H$_2$ and He gas charge strippers. The mean charge states of $^{86}$Kr in H$_2$ and He gases attained equilibrium at 25.1 and 23.2, respectively, whereas the mean charge state in N$_2$ gas at equilibrium was estimated to be below 20. The $Q_{eq}/Z$ values were enhanced by 0.14 and 0.09 in H$_2$ and He gases, respectively. We were able to obtain $^{86}$Kr$^{26+}$ with a fraction of 31% in H$_2$ gas with a thickness of 68 $\mu$g/cm$^2$. The H$_2$ gas stripper can be applied for $^{86}$Kr acceleration. The ionization and electron capture cross sections calculated by using the BEM and OBK model were optimized such that the calculation of the charge state evolution reproduced the experimental data. The optimized scaling factors $a$ and $b$ are $a = 0.99$ and $b = 0.75$ for H$_2$ and $a = 1.00$ and $b = 1.08$ for He.

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