Growth of Ionization Balance from F-like to Bare Ions of Heavy Atoms in an Electron Beam Ion Trap

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Ionization balance from F-like to bare ions of In and I is investigated as a function of trapping time by measuring extracted ions from an electron beam ion trap, the Tokyo-EBIT. The present experiment is the first clear demonstration of the temporal behavior of the very highly charged ions that have been produced in the EBIT. The growth rate of extracted H-like ions is compared with that of the intensity of the x-ray emission resulting from radiative recombination into the K-shell of H-like ions. The absolute number density of trapped ions was estimated from the intensity of the x-ray emission.

Keywords: electron beam ion trap, highly charged ion, growth rate, charge state distribution, radiative recombination, x-ray

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1. Introduction

The electron beam ion trap (EBIT) is a powerful instrument for the study of highly charged ions (HCIs). The EBIT was originally developed for trapping HCIs for an extended time in x-ray spectroscopy and fundamental atomic physics studies [1, 2]. Recently, ion extraction systems have been added to EBIT machines to investigate HCI-collision phenomena, due to the rapid growth of interest in the application of HCIs to nano-science on solid surfaces [3,4]. The ion extraction system also makes it possible to understand HCI-production in the trap through direct measurements of the charge-state (q) distribution and find better operation conditions for the EBIT.

The Tokyo EBIT was constructed at the University of Electro-Communications to study spectroscopy and interactions of HCIs having very high charge states [5]. The production of high-q ions was made possible by using high-energy electron beams [6, 7]. In this paper, we report the measurements of q-distributions of extracted ions, and show the growth of these charge-states using different elements and under different operation conditions. Although several experiments for investigating the growth rate of HCIs produced in the trap by measuring the extracted ions have been performed previously [8–10], the present study is the first definitive demonstration of the time-dependent behavior of very high-q HCI-production in the EBIT.

2. Experimental

The experimental arrangement for ion extraction is shown schematically in Fig. 1. A recent experiment using this arrangement has been described in detail elsewhere [11,12]. Briefly speaking, the EBIT consists of an electron gun, ion trap, and electron collector. The electron beam is compressed to about 3×10^{-5} m in radius by a strong magnetic field (4 T). It then passes through the ion-trap region and is collected. HCIs are produced and trapped

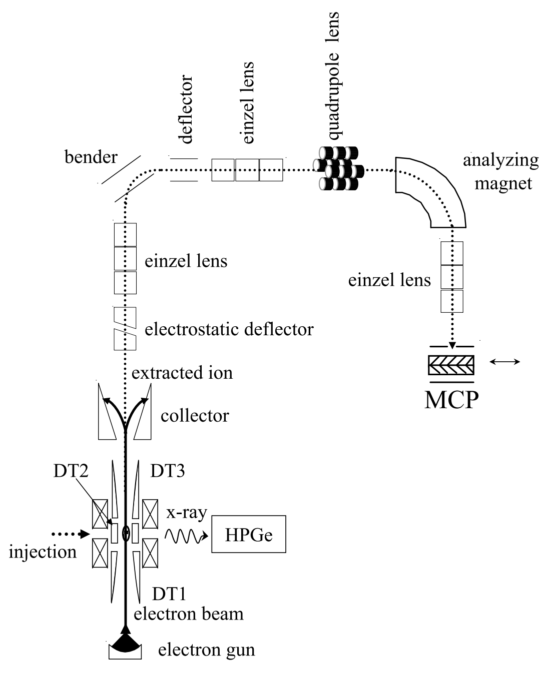

Fig. 1 Schematic view of the Tokyo EBIT and the extraction beam line. DT: drift tube, HPGe: high purity Germanium, MCP: micro-channel plate.

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inside the ion-trap region that consists of three drift tubes (DTs), DT1, DT2, and DT3, from the bottom to the top. The actual trapping occurs in the central tube DT2, which has a length of 0.03 m. In the normal operation for spectroscopic observation of HCIs in the trap, the two end drift tubes, DT1 and DT3, are biased with respect to the central DT2 to form a potential well. Axial trapping of the ions is performed by this potential well, whose depth is usually set at 100 - 300 V, while radial trapping (at the typical radial potential of about 40 V) is performed by a combination of the space-charge field of the electron beam and the applied magnetic field.

Several methods for ion or neutral gas injection into the trap have been used in the Tokyo EBIT activity [13]. For ion injection, a metal vapor vacuum arc (MEVVA) ion source [14] can be used to inject low-charged metallic ions into the trap from the top of the EBIT. The most widely used method is neutral injection through one of the side ports at the DT2. In addition to normal gaseous elements such as Kr or Xe, volatile molecular compounds such as CH_3I or $W(CO)_6$ have been injected from a liquid cell and metallic elements such as Mn or Bi from a Knudsen effusion cell (K-cell) [15].

In general, the charge (q) balance in EBIT is determined by electron-ion (ionization and recombination) and charge-exchange collisions with neutral atoms. When the neutral density increases, the mean charge-state $\bar{q}$ of HCIs produced shifts to lower q. Therefore, the injected neutral density should be controlled so as to be of the order of 10^{15} m^{-3} in the trap.

In order to avoid the influence of contamination in the trap due to evaporation of the cathode materials such as Ba and W, the trapped ions must be dumped periodically and re-accumulated for highly effective HCI-production for the source elements. There are two modes of the ion-extraction method, continuous (leaky) and pulsed modes. In the pulsed extraction mode, the trap is emptied by raising the potential of DT2 above that of DT3 in a short period of time. Then, almost all the trapped ions can be dumped toward the detection system. On the other hand, in the continuous extraction mode, ions with enough energy obtained through collisional heating by the electron beam surmount the potential well and escape from the trap region to the detector.

The growth behavior of the ionization balance for HCIs produced in the trap can be studied for both extraction modes. In the pulsed mode, the growth can be observed by varying the time at which the trapped ions are dumped and the corresponding intensities are measured in the detector system. The intensity at each dumping period after a given trapping duration is recorded as a point in the measured growth curve. Therefore, the entire system including HCI-production and -extraction must be strictly stable and well-controlled during the measurement to obtain the growth curve. In the pioneering work by Donets and Ovsyannikov [16], the evolution of q-distribution was thoroughly studied for C, N, O, and Ar using a time-of-flight spectrometer in the pulsed mode.

In the present study, the growth of HCI-production is measured in the continuous mode, in which all DT potentials applied are kept constant; the potential bias of DT1 is 100 V and that of DT3 is equal to the DT2 potential. Although the axial potential well was negligibly small, the radial potential resulting from the space charge of the electron beam can trap the HCIs produced, which is about 40 V at the beam energy of $E_e = 58$ keV and the current $I_e = 151$ mA (the current density of 5.2×10^7 A/m^2).

An extraction beam line is located above the main body of the Tokyo-EBIT (Fig. 1). The HCI-beam transport system includes electrostatic (einzel and quadrupole) lenses, deflectors, and a 90° bender. After selecting the charge-state (q) using an analyzing magnet, ions are detected by a micro-channel plate (MCP). In addition to the observation of extracted ions, the production of high-q HCIs can be monitored by measuring x-rays emitted from the trapped ions through a Be window at 90° to the electron beam using a high purity Germanium (HPGe) detector.

To investigate the growth of charge-state q in the HCI-production, the electron beam is turned on periodically by the fast square-wave control of an applied voltage to the anode (0 V to 5 kV with respect to the cathode). The intensity of the leaked-out HCIs with q is measured as a function of the trapping time-length passed from the beginning of electron beam-on and recorded using a multi-channel scaler (MCS) to observe the growth curve.

The extraction potential is determined by an applied voltage to the DT2 which is 3 kV in the present experiment. The intensity of extracted HCIs is dependent on the operating conditions of the electron beam and gas injection system; the typical intensity of extracted He-like Xe^{52+} ions is over 10^3 ions per second.

3. Charge-State (q) Distribution

Figure 2 shows the charge-state distribution of extracted Mn^{q+} as a function of the mass to charge ratio (m/q), which is measured in the continuous extraction mode. Mn (atomic number $Z = 25$) is injected as a gas nozzle beam into the trap from the K-cell (heated at about 850°C). HCIs of Mn^{q+} are produced here with the EBIT operating at $E_e = 58$ keV and $I_e = 125$ mA. Bare (Mn^{25+}) to He-like (Mn^{23+}) ions are dominant in the extracted ions with the present operation conditions (Fig. 2).

For comparison, HCI-production of In ($Z = 49$) as a higher Z element is investigated using almost the same EBIT operating conditions ($E_e = 58$ keV, $I_e = 151$ mA). The charge-state distribution of extracted In^{q+} is shown in Fig. 3. While the dominant charge-states in the extracted In^{q+} range from Ne-like ($q = 39$) to He-like ($q = 47$) ions, weak signals for H-like and bare ions can be observed (see the inset of Fig. 3). The intensity ratio of H-like to bare ions is measured to be 11.8 ± 0.8 under the present operat-

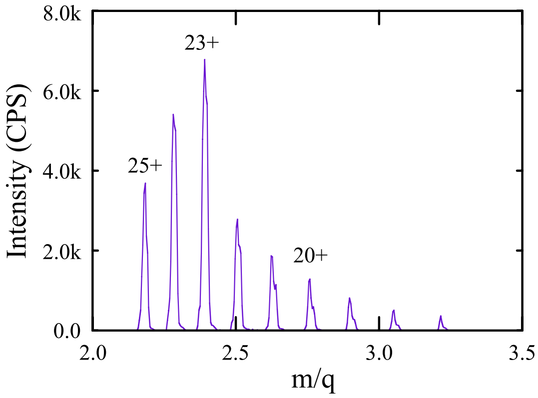

Fig. 2 Charge-state distribution of extracted $^{55}Mn^{q+}$ ($Z = 25$) from the Tokyo EBIT in the continuous extraction mode under the EBIT operation condition of $E_e = 58$ keV and $I_e = 125$ mA.

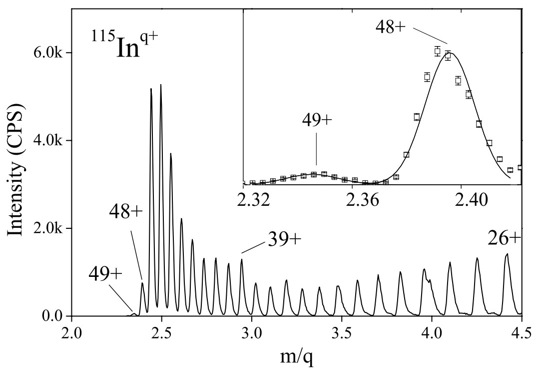

Fig. 3 Charge-state distribution of $^{115}In^{q+}$ ($Z = 49$) ions ($26 \leq q \leq 49$) in the continuous extraction mode under the EBIT operation condition of $E_e = 58$ keV and $I_e = 151$ mA. The insert figure shows extracted H-like In^{48+} and bare In^{49+} ions. The solid line is the Gaussian function fitted to the data.

ing conditions.

We investigated the charge-state distribution of HCIs in the trap for In^{q+} by measuring x-ray emission spectra from the trapped HCIs (Fig. 4). Radiative recombination (RR) of HCIs with the beam electrons produced a series of lines in which the energy was equal to the sum of the electron and binding energies of the vacant shell into which the electron is captured. In Fig. 4, a clear series of x-ray peaks corresponds to x-ray spectra from In^{q+} resulting from RR into $n = 1$ to $n = 4$ shells, (the small peaks correspond to contaminant HCIs of Ba and W from the cathode materials).

H-like and bare In ions produce resolved RR lines for K-shell ($n = 1$) capture (see the insert of Fig. 4). RR spectra from the other high-q ions are incompletely resolved. However, the equilibrium q-distribution can be obtained, in principle, by fitting the RR spectra, because the RR cross-sections are well known. The dominant peak from RR into

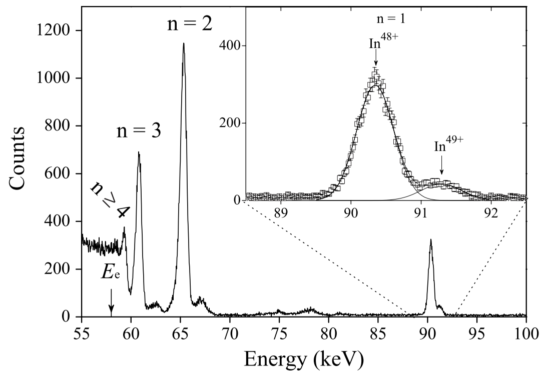

Fig. 4 Radiative Recombination (RR) x-ray spectra emitted from trapped $^{115}In^{q+}$ ions under the EBIT operating condition $E_e = 58$ keV and $I_e = 151$ mA. The insert shows the RR into $n = 1$ in H-like ions (In^{48+}) and bare ions (In^{49+}). The solid line is the Gaussian function fitted to the data.

$n = 2$ in Fig. 4 suggests that the high-q ions from F-like to He-like In ions with L-shell ($n = 2$) hole(s) are abundantly trapped, which is in agreement with the observation of the q-distribution of extracted In^{q+}.

Since the RR cross-section to $n = 1$ for a bare ion is two times larger than that for a H-like ion corresponding to the number of K-holes, the abundance ratio of H-like to bare In ions is more precisely determined, which is estimated to be 12.8 ± 0.9 from the intensity measurement of RR ($n = 1$) spectra. This is also in agreement with the measured abundance ratio in the extracted ion-intensities. Consequently, the charge-state q distribution of extracted ions is considered to be equivalent to the relative abundance of HCIs in the trap for the very highly charged ions. In other words, under the condition that the extraction, transport, and detection efficiencies are the same for different q-ions with a narrow q-region, the q-distribution of extracted ions may be measured to be equivalent to that of trapped ions.

Based on this equivalency, the absolute number density of trapped ions can be estimated from the measured intensity of RR x-ray. At equilibrium, the x-ray count rate for RR into n-shell is approximately expressed as

$$R_n^{\mathrm{RR}} \approx \epsilon \times T \times \frac{L I_e N_q \sigma_n^{\mathrm{RR}}}{e} \times \frac{\mathrm{d}\Omega}{4\pi}, \tag{1}$$

where ϵ is the detection efficiency of the HPGe for the measured x-ray, T is the transmission of the Be window, L is the trap length, N_q is the density of the trapped HCIs with q, σ_n^{RR} is the the scaled RR cross-section for RR into n [17], e is the electron charge, and $\mathrm{d}\Omega$ is the solid angle subtended by the HPGe. First, from the measurement of RR into $n = 1$ (R_1^{RR}) (Fig. 4), the densities of H-like In^{48+} and bare In^{49+} ions are determined to be $1.0 \pm 0.2 \times 10^{14}$ and $8.2 \pm 1.6 \times 10^{12}$ m^{-3}, respectively. Second, using the relative intensity distribution of extracted HCIs including In^{48+} and In^{49+} (Fig. 3), the number densities of He-like (47+) to

Ne-like (39+) ions, which dominate the ions in the trap, are obtained. By summing up these values, the number density of the trapped ions is approximated to be 3×10^{15} m^{-3}. On the other hand, the number density of electrons was about 2.3×10^{18} m^{-3} in the present operation of the electron beam with E_e = 58 keV, I_e = 151 mA, and the radius of 3×10^{-5} m. Therefore, space-charge neutralization of the electron beam by the trapped ions is estimated to be less than 10%, assuming the averaged charge state $\bar{q}$ of trapped HCIs to be 45.

4. Time Evolution

The growth of the charge-states of the extracted HCIs was observed to investigate HCI-production efficiencies for different ionic species in the trap and the different EBIT-operation conditions. Figure 5 shows the growth curves for In^{q+} with q = 40 (F-like) to 49 (bare). At time (t) = 0, the electron beam begins to interact with the injected In atoms and ionize them, where the bound electrons in outer shells ($n \geq 3$) are stripped very quickly (< 0.1 s) and those in n = 2 are stripped successively over a relatively long time. After 1.5 s, the charge-state distribution reached a steady state.

For the lower electron-energy operation of E_e = 23 keV (Fig. 5 (a)), the most abundant charge-state is q = 45 (Be-like) in the steady state, and H-like and bare ions could not be observed. However, for the E_e = 58 keV operation (Fig. 5 (b)), the growth curve shows a different shape, that is, the most abundant ions are He-like(q = 47), the intensities of higher q ions clearly increase with q (> 43) in the steady state and H-like and bare ions can be observed.

The growth curves of q-distribution for the extracted ions are measured for I^{q+} ions with different Z (= 53) but under the same operating conditions as those for In (Z = 49) (Fig. 6). Although overall structures in the time evolution seem qualitatively similar, the relative ionization balance in the steady state is shifted to the lower stripped ions in the same E_e operation, according to the higher ionization energy of I^{q+} at the same iso-electronic sequence compared with that of In^{q+}.

As shown in Figs. 5 and 6, a distinct peak appears in the respective growth curve of a HCI with relatively lower q such as F-like and O-like ions, where the peak position shifts toward longer interaction (trapping) time with increasing q. According to a simple model, the q-balance is determined by the ionization rate corresponding to the positive and negative growth rates due to radiative recombination and escape from the trap of heated ions by electron impact [9]. The two negative rates become important for ions with higher q and Z. Therefore, at the beginning of the interaction with electrons, the number of ions with q would increase rapidly with the increase in the number of ($q - 1$) ions, reach the peak value quickly, and then de-

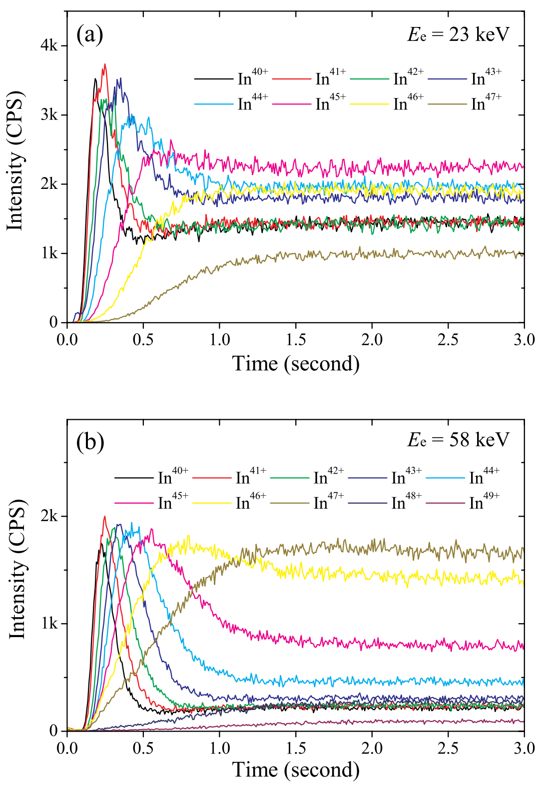

Fig. 5 (Color online) Growth curves of extracted ^{115}In^{q+} ions with the electron beam energy of 23 keV (a) and 58 keV (b) from q = 40 (F-like ion) to q = 49 (bare ion).

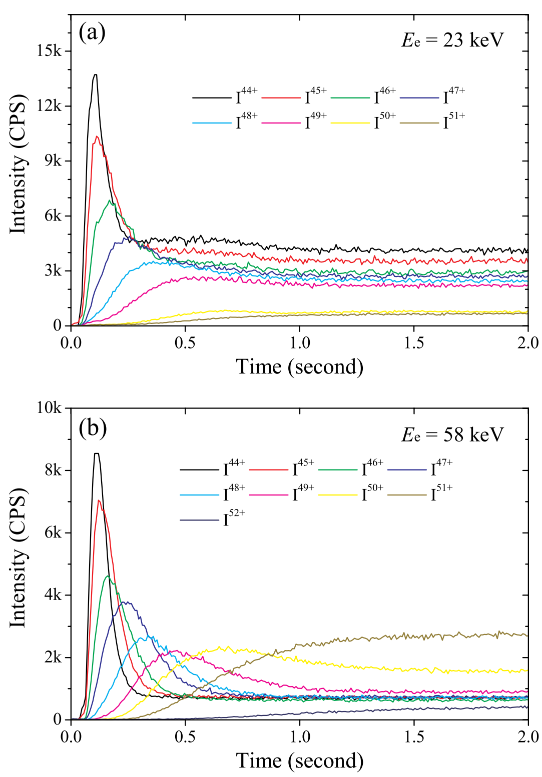

Fig. 6 (Color online) Growth curves of extracted ^{127}I^{q+} ions with the electron beam energy of 23 keV (a) and 58 keV (b) from q = 44 (F-like ion) to q = 52 (H-like ion).

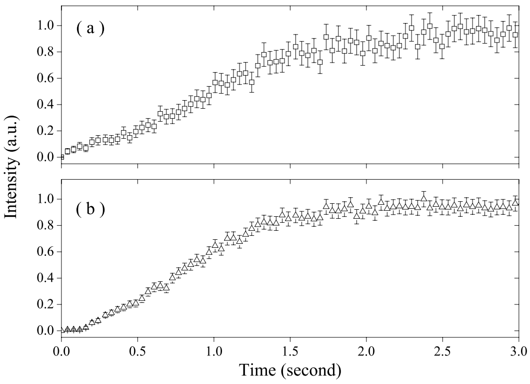

Fig. 7 Relative growth curves of the intensity for RR x-ray into $n = 1$ in H-like In^{48+} ions in the trap (a) and that for the extracted In^{48+} (b).

crease gradually by the above negative growth factors together with decreasing $(q-1)$ ions. Consequently, the peak structure appears in the respective growth curve. In fact, as shown in Figs. 5 (b) and 6 (b) at the $E_e = 58$ keV operation, peaks appear for F- to Li-like ions, whose positions shift to the longer time with increasing q. For He-like ions, which are the most abundant ions in the steady state, there are no clear peaks in the growth curves; however, by using the appropriate coolant gas in the ion-cooling procedure in the trap, the broad peaks might appear. In contrast to the ions mentioned above, numbers of H-like and bare ions are and their growth rates are very low. Since the ionization cross-sections of He-like and H-like ions are significantly smaller than those of other ions with lower q, their ionization time-constants necessarily become very large.

Figure 7 shows the growth curve of RR ($n = 1$) x-ray signals for H-like In^{48+} ions (Fig. 7(a)), which can be meaningfully compared with that of extracted H-like In^{48+} ions (Fig. 7 (b)). The temporal growth behaviors are very similar, which suggests that the growth of HCI-production in the trap could be estimated by observing the intensity variation of the extracted ions. However, the slightly different behavior at the beginning phase just after the electron beam-on could be detected upon careful observation, where the growth curve for extracted H-like In^{48+} ion might have a short delayed-onset. We assume that it takes time for very highly charged ions like In^{48+}, which would exist at the bottom of the potential well, to leak out in the continuous (leaky) mode.

5. Summary

Owing to recent progress of experimental research on highly charged ions (HCIs), the EBIT machines have advanced rapidly as ion sources that can provide slow HCIs with very high charge-states (q's). The Tokyo EBIT has also been developed into a powerful HCI-source for application research on the extracted HCI-beam to nanotechnology [18–21], and also for atomic physics research that investigates the resonant HCI-interaction with electrons in the trap by observing the variation of q-distribution in the extracted ions [22–24].

In the course of our systematic investigation, we observed the q-distributions of extracted HCIs with the different species and the EBIT operations, and also measured the growth behavior of those ions, which is assumed to reflect the growth of charge-states of HCIs produced in the trap.

In the present observations, the measured growth curves (Figs. 5 (b) and 6 (b)) are the first definitive demonstration of the temporal evolution of the ionization balance in the HCI-production for high Z elements. There have been many studies that model HCI-production incorporating various kinds of atomic processes in the EBIT and of escape processes from the trap [25]. Among them the simplest model includes only single-charge-changing processes by electron impact ionization and recombination [26]. Even with this simple model, the calculated growth curve could reproduce qualitatively the experimental observation in the high-q region, as shown in Figs. 5 and 6. However, the quantitative agreements have not been obtained sufficiently well even if a number of possible processes were considered. Therefore, it seems important to accumulate experimental investigations using a wide variety of the HCI-species and operating conditions.

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[1] M.A. Levine, R.E. Marrs, J.R. Henderson, D.A. Knapp and M.B. Schneider, Phys. Scr. T **22**, 157 (1988).
[2] D.H.G. Schneider, J. Steiger, T. Schenkel and J.R. Crespo Lòpez-Urrutia, *Atomic Physics with Heavy Ions* edited by H.F. Beyer and V.P. Shevelko (Springer, Berlin, Heidelberg, 1999) Chap. II.
[3] T. Schenkel, A.V. Hamza, A.V. Barnes and D.H. Schneider, Prog. Surf. Sci. **61**, 23 (1999).
[4] J.D. Gillaspy, J. Phys. B: At. Mol. Opt. Phys. **34**, R93 (2001).
[5] H. Watanabe *et al.*, J. Phys. Soc. Jpn. **66**, 3795 (1997); F.J. Currell *et al.*, J. Phys. Soc. Jpn. **65**, 3186 (1996).
[6] N. Nakamura, F.J. Currell, D. Kato, A.P. Kavanagh, Y.M. Li, S. Ohtani, H.A. Sakaue, M. Sakurai, J. Sun, S. Takahashi, M. Tona, H. Watanabe, C. Yamada and N. Yoshiyasu, Can. J. Phys. **86**, 315 (2007).
[7] S. Ohtani, AIP Conf. Proc. of EBIS/T: 8th Int'l Symposium. **572**, 84 (2001)
[8] E.D. Donets, Phys. Scr. T **3**, 11 (1983).
[9] B.M. Penetrante, J.N. Bardsley, D. DeWitt, M. Clark and

D. Schneider, Phys. Rev. A **43**, 4861 (1991).
[10] E. Sokell, F.J. Currell, H. Shimizu and S. Ohtani, Phys. Scr. T **80**, 289 (1999).
[11] M. Tona and S. Takahashi, J. Phys.: Conf. Ser. **2**, 57 (2004).
[12] M. Tona, H. Watanabe, S. Takahashi, N. Nakamura, N. Yoshiyasu, M. Sakurai, T. Terui, S. Mashiko, C. Yamada and S. Ohtani, Surf. Sci. **601**, 723 (2007).
[13] H. Watanabe, J. Sun, M. Tona, N. Nakamura, M. Sakurai, C. Yamada, N. Yoshiyasu and S. Ohtani, Phys. Rev. A **75**, 062901 (2007).
[14] I.G. Brown, J.E. Galvin, R.A. MacGill and R.T. Wright, Appl. Phys. Lett. **49**, 1019 (1986).
[15] C. Yamada, K. Nagata, N. Nakamura, S. Ohtani, S. Takahashi, T. Tobiyama, M. Tona, H. Watanabe, N. Yoshiyasu, M. Sakurai, A.P. Kavanagh and F.J. Currell, Rev. Sci. Instrum. **77**, 066110 (2006).
[16] E.D. Donets and V.P. Ovsyanniko, Sov. Phys. JETP **53**, 466 (1981).
[17] Y.S. Kim and R.H. Pratt, Phys. Rev. A **27**, 2913 (1983).
[18] H. Watanabe, S. Takahashi, M. Tona, N. Yoshiyasu, N. Nakamura, M. Sakurai, C. Yamada and S. Ohtani, Phys. Rev. A **74**, 042901 (2006).
[19] J. Sun, H. Watanabe, M. Tona, T. Watanabe, N. Nakamura, C. Yamada and S. Ohtani, Phys. Rev. A **77**, 032901 (2008).
[20] M. Tona, Y. Fujita, C. Yamada and S. Ohtani, Phys. Rev. B **77**, 155427 (2008).
[21] M. Tona, T. Abe, H. Watanabe, J. Sun, N. Nakamura, C. Yamada, M. Kotani and S. Ohtani, Phys. Rev. A **77**, 052902 (2008).
[22] N. Nakamura, H. Tobiyama, H. Nohara, D. Kato, H. Watanabe, F.J. Currell and S. Ohtani, Phys. Rev. A **73**, 020705 (2006).
[23] H. Watanabe, H. Tobiyama, A.P. Kavanagh, Y.M. Li, N. Nakamura, H.A. Sakaue, F.J. Currell and S. Ohtani, Phys. Rev. A **75**, 012702 (2007).
[24] N. Nakamura, A.P. Kavanagh, H. Watanabe, H.A. Sakaue, Y.M. Li, D. Kato, F.J. Currell and S. Ohtani, Phys. Rev. Lett. **100**, 073203 (2008).
[25] I.V. Kalagin, D. Küchler, V.P. Ovsyannikov and G. Zschornack, Plasma Sources Sci. Technol. **7**, 441 (1998).
[26] B.M. Penetrante, D. Schneider, R.E. Marrs and J.N. Bardsley, Rev. Sci. Instrum. **63**, 2806 (1992).