Production of high-charge-state thorium and uranium ions in an electron-beam ion trap

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High-charge-state thorium (up to 80+) and uranium (up to 70+) ions have been extracted from an electron-beam ion trap (EBIT). The ions were produced after injecting low-charge-state thorium and uranium ions initially from a metallic vapor vacuum arc ion source into the trap. The extracted ions from the EBIT provide microsecond-wide ion-beam pulses of about 10^4 ions per second. The production and loss rates are estimated from modeling calculations and are compared to the measured extracted ion rates.

I. INTRODUCTION

Much effort has been devoted to the research and development of ion sources capable of producing very high ionic charge states in high-Z atoms. One type that has demonstrated great success is the electron-beam ion source (EBIS), a concept that originated with Donets [1]. Much interest in the EBIS developed when Donets and Ovsyannikov [2] reported measurements of ionization cross sections for xenon up to Xe^{47+} . The EBIS offered the possibility of building a laboratory-size alternative to the large accelerators for studies of highly charged ions. To further exploit this possibility, the electron-beam ion trap (EBIT) was built at Lawrence Livermore National Laboratory [3,4].

The EBIT is, in essence, a variant of the EBIS, differing in emphasis rather than kind. The main structural difference is the length of the electron-beamion interaction path; it is generally about 1 m in the EBIS and about 2 cm in the EBIT. The EBIT produces higher charge states and can maintain a given charge state much longer than was possible in an EBIS. The most highly charged ion studied in the EBIT to date is neonlike uranium U⁸²⁺, and trapping times in excess of 5 h have been observed [5]. In operation, the EBIT is proving to be a versatile and powerful tool for investigating atomic structure and electron-ion interactions [6–12].

The capability of the EBIT has recently been extended by adding an efficient ion extraction system that allows for the transport of the highest-charge-state ions to an external target and detector system [13]. In addition to doing high-resolution x-ray spectroscopy, electron-ion collisional processes can now also be studied independently by directly measuring the charge-state distributions of the extracted ions. Furthermore, the high efficiency of the extraction, i.e., the number of extracted ions per pulse available for experiments outside the EBIT, opens up avenues for new physics to be studied. The extracted ions can be used as a beam of slow (few eV of kinetic energy), very highly charged (more than 500keV potential energy) ions for atomic collision experiments.

The objectives of this paper are (1) to demonstrate that the EBIT can be used as a source of high-quality, highcharge-state, high-Z ions, (2) to discuss quantitatively the processes that limit the highest charge state and the maximum ion yield from the EBIT, and (3) to provide recommendations on how the EBIT can be easily enhanced to provide highly charged ions in quantities that are competitive to other ion sources.

II. EXPERIMENTAL SETUP AND PARAMETERS

The operation of the EBIT and the ion extraction system are described in detail by Levine *et al.* [3,4] and Schneider *et al.* [13], respectively. Only the details relevant to the production and extraction of highly charged Th and U ions are given here.

The major requirements of an EBIS or an EBIT are relatively well understood. These are (1) a high-quality vacuum, (2) a well-aligned axial magnetic field, and (3) a high-energy, high-current-density electron beam. The EBIT's vacuum system is designed to provide a vacuum of down to 10^{-12} Torr inside the drift tubes. Residual gas from the drift tube walls, contaminants from the cathode, and cooling gas injected intentionally into the trap raise the background pressure. In a later section we show that the pressure is still less than 10^{-10} Torr, which is low enough to provide sufficient cooling for the high length of the high length.

The typical operating electron-beam current in the EBIT is 100 mA. The present cathode can have an output of up to 200 mA. As the beam enters the drift tube region, a 3-T axial magnetic field from two superconducting Helmholtz coils compresses the beam to current densities of up to 6000 A/cm². The axial magnetic field varies by less than 0.02% over the 2-cm length of the ion trap. The drift tube assembly accelerates the beam electrons to their full interaction energy and provides an axial electrostatic trap for the positive ions. The present apparatus is capable of producing electron energies up to 30 keV, and an upgrade currently in construction will have electron energies up to 150 keV.

The EBIT is equipped with a MEVVA (metallic vapor vacuum arc) source [14] which produces the initially low charged (up to 4+) Th and U ions, in pulses of about 10^{11} ions per pulse. Only a fraction of these ions make it into



FIG. 1. Measured charge-state distributions for Th ions and background ions. The electron-beam energy is 10 keV and the confinement time is 2 s.

the trap. Once in the central drift tube trap region, the ions are confined axially by the potential well provided by two end drift tubes. The axial potential well depth is typically around 100-300 V. Radial confinement is provided by a combination of the space-charge field of the electron beam and the applied axial magnetic field. The ions undergo successive ionizations in the electron beam. The highest ionization state present in the trap is determined by the electron-beam energy. In order to inject the ions into the trap, confine and ionize them for a given time, and to extract them, the potentials on the trap have to be varied in a fast switching mode. In the extraction phase the ions are electrostatically deflected out to the MEVVA-einzel lens in the path between the MEVVA and the trap. The same lens provides focusing of the initially injected ions from the MEVVA into the trap. After passing the deflector, the ions are refocused via a second einzel lens and are momentum analyzed in a 90° magnet. A schematic diagram of the EBIT, including the ion extraction system, is shown in Fig. 1 of Schneider et al. [13].

III. ION EXTRACTION MEASUREMENTS

Figure 1 shows a spectrum of mass and chargeanalyzed ions which are extracted from the EBIT, following injection of low-charge-state Th ions into the trap with an electron-beam energy of 10 keV and a confinement time of 2 s. The spectrum shows various contributions from the lighter ions in the trap. These clearly identifiable contributions show the species of the background ions that are formed in the trap following ionization of residual gas atoms. The peaks allow for a calibration of the total spectrum of mass-charge-analyzed ions. The yield of ions from the residual gas in the extraction spectrum appear to be reduced when high-Z ions are injected and ionized in the trap. This observation is consistent with the evaporative cooling mechanism [5,15]. Energy is transferred from the high-Z ions to the low-Z ions through Coulomb collisions. The low-Z ions, because of their lower charge, escape from the trap faster than the high-Z ions.



FIG. 2. Measured charge-state distributions for (a) Th ions at a beam energy of 24 keV, trapping time of 3 s, and axial potential well depth of 100 V; (b) U ions at a beam energy of 9 keV, trapping time of 4 s, and axial potential well depth of 140 V.

Figures 2(a) and 2(b) show the measured charge-state distributions of Th, and U, respectively. For the ions were confined in the trap for 3 s, the electron-beam energy was 24 keV, and the axial potential well depth was 100 V. For U, the ions were confined in the trap for 4 s, the electron-beam energy was 9 keV, and the axial potential well depth was 140 V. These confinement times were long enough to allow the highest charge states for each species to reach steady state.

The vacuum in the beam transport system between the EBIT and the analyzing magnet is about 10^{-7} Torr. Suppose the ions spend about 1 μ s along the extraction path. The charge-exchange rate at 10^{-7} Torr for Th⁷⁷⁺ and U⁶⁶⁺ with O is roughly 10⁸ ions/s. Thus, the number of ions destroyed by charge exchange during the extraction is ~10², which is negligible compared to the number of ions extracted from the trap.

The extraction yield is around 10^4 ions. If we assume an extraction efficiency of 10% and a detection efficiency of 50%, the number of low-charge ions from the MEVVA that reach the trap region and eventually gets ionized is 2×10^5 ions. Since the MEVVA can provide up to 10^{11} ions per pulse, we known that we can inject more ions than can be trapped. In the next section we discuss the processes that determine the maximum number of ions that can be trapped ion the EBIT.

IV. PROCESSES CONTROLLING THE HIGH-CHARGE STATES

The highest ionization state present in the trap is determined by the electron-beam energy. The theoretical limit on the ionization balance is determined by the ratio of two electron-ion interactions: electron-impact ionization and radiative recombination. There is also an unavoidable contribution from charge exchange with neutral atoms and low-charge-state ions. In the absence of cooling, the collisional heating of the ions by the electron beam limits the maximum charge state and the time that the ions can remain trapped. In order to minimize the escape of the highly charged high-Z ions, these ions are cooled by the continuous injection of low-Z atoms into the trap. The amount of low-Z material used to cool the high-Z ions must be chosen to provide adequate cooling while not significantly degrading the charge-state balance through charge exchange. The evaporative cooling process has been experimentally demonstrated by Schneider et al. [5] and discussed in detail by Penetrante et al. [15].

We have developed a code to calculate the evolution of the ion charge-state distribution in an EBIT. The code solves the set of coupled nonlinear rate equations for the ion number and energy balance. Details of the calculations are described by Penetrante *et al.* [16]. The processes controlling the ion number balance are (1) electron-impact ionization, (2) radiative recombination, (3) charge exchange, and (4) radial and axial escape from the trap. Dielectronic recombination can be easily incorporated in the calculation; however, it is a resonant process and is negligible for the electron-beam energies used

in this study. The processes controlling the energy balance are (1) heating of the ions by the electron beam, (2) energy transfer among the individual ions, and (3) energy loss through radial and axial ion escape. Code inputs are the neutral gas density of the coolant, initial density of the injected high-Z ions, electron-beam energy and current, applied axial potential, magnetic field strength, physical dimensions of the trap, and atomic physics data. The calculations are dependent upon the quality of the collision cross sections. We have adopted the use of general and simple formulas for these cross sections. For the ionization cross sections we use the formula derived by Lotz [17]. The radiative recombination cross section derived by Kim and Pratt [18] is used. The chargeexchange cross-section formula is given by Müller and Salzborn [19]. The accuracies of these formulas are not always known for the parameter regions that we are interested in. For example, there is a paucity of ionization cross-section measurements at the energies of interest to check the accuracy of the Lotz formula. The measurements that do exist suggest that the ionization cross sections estimated by the Lotz formula may be low by factors of 2 or more for partially stripped multiply charged ions due to neglect of indirect ionization. Despite the obvious errors, the procedure allows detailed parameter studies to establish how the basic operating parameters affect the charge-state distribution.



FIG. 3. Calculated steady-state ion charge-state distributions for (a) Th and (b) U, as a function of the background gas pressure. The electron-beam energy used is 24 keV for Th and 9 keV for U. The electron-beam current is 100 mA and the axial magnetic field is 3 T.



FIG. 4. Calculated steady-state ion charge-state distribution for (a) Th with a background gas pressure of 7×10^{-11} Torr, and (b) U with a background gas pressure of 4×10^{-11} Torr. The operating parameters are the same as in Figs. 2 and 3.

The maximum extraction yield for a given charge state is determined by three factors: (1) the availability of coolant ions from the background gas, (2) the degradation due to charge exchange with the background gas, and (3) the neutralization of the electron-beam space charge by the highly charged ions.

It is useful to estimate the background gas pressure in the EBIT in order to determine whether or not sufficient cooling is being achieved, and whether or not charge exchange is tolerable. The present ionization gauges are good only down to 10^{-10} Torr and are located in remote positions, so we cannot verify what the actual pressure is in the trap. Furthermore, the gas injection system for the coolant gas, although it provides an adjustable level of cooling while minimizing the gas load on the EBIT's vacuum pumps, does not provide a measure of the gas density that intersects the electron beam. We estimate the background gas pressure by comparing the charge-state distributions from the extracted ion measurements with those from the computer simulations.

Figure 3 shows the calculated charge-state distributions for Th and U as a function of background gas pressure. The pressure dependence of the charge states come about because of charge exchange. The measured distributions in Fig. 2 correspond to pressures of 7×10^{-11} and 4×10^{-11} Torr for Th and U, respectively. The calculated charge-state distributions shown in Fig. 4 are in very



FIG. 5. Calculated ionization, radiative recombination, and charge-exchange rates in (a) Th and (b) U. The parameters are the same as in Fig. 4.



FIG. 6. Calculated evaluation of charge-state distributions of (a) Th and (b) U ions.

good agreement with the measured distributions in Fig. 2.

The estimated pressures correspond to background gas densities greater than 10^6 cm⁻³. If we assume that the space-charge neutralization of the electron beam by the ions is 10%, the charge-state distributions in Fig. 2 (or Fig. 4) correspond to total ion densities of 2.3×10^9 and 2.2×10^9 cm⁻³ for Th and U, respectively. Our calculations show that a continuous supply of 10^6 cm⁻³ O or N in the trap is more than sufficient to completely cool the highly charged Th and U ions.



FIG. 7. Calculated effect of the electron-beam current density on the average charge states of Th and U ions.



FIG. 8. Calculated effect of the electron-beam current density on the total ionization, radiative recombination, and chargeexchange rates of Th and U ions.

The measured charge-state distributions shown in Fig. 2 are remarkably close to the theoretical limit in which the populations of the charge states are determined only by the ratio of the cross sections of electron-impact ionization to radiative recombination. The quality of the charge states attained in the EBIT indicates how well it meets its mechanical requirements.

The calculated ion production and loss rates for Th and U are shown in Fig. 5. These rates assume a spacecharge neutralization of the electron beam of 10%. If the extraction and the detection efficiencies are indeed 10 and 50%, respectively, then the extracted ion rates shown in Fig. 2 indicate that the Th and U ion densities provide around 10% space-charge neutralization of the electron beam.

The duty cycle is determined by the time it takes for the charge-state distribution to reach steady state. The evolution of the Th and U charge-state distributions are shown in Fig. 6. It takes about 1 s for the highly charged Th and U ions to reach their maximum values.

The final limitation on the maximum number or density of trapped ions is determined by the space-charge neutralization of the electron beam by the ions. Cooling and charge exchange are important considerations to take into account; however, we have shown that a continuous supply of 10^6 cm⁻³ of background gas is sufficiently high enough to cool very highly charged Th and U ions with trapping densities of greater than 10^9 cm⁻³, and sufficiently low enough to prevent excessive degradation of the charge states by charge exchange. Thus, the maximum density of trapped ions will always be limited by the density of the electron beam. Provided that a sufficient number of ions from the MEVVA can be guided into the trap region to reach the beam neutralization limit, increasing the electron-beam current density is the easiest way of further increasing the extracted ion yield. Higher current densities will also allow the tolerance of a higher background gas density for cooling. Figure 7 shows the effect of the current density on the average charge state of Th and U ions. The effect of the current density on the total ionization, radiative recombination, and charge-exchange rates is shown in Fig. 8.

V. CONCLUSIONS

The EBIT can be used as an efficient and reliable source of very high-charge-state ions for atomic species up to uranium. This has been demonstrated by producing and extracting ions up to Th^{80+} and U^{70+} with the EBIT. By comparing with the extracted ion measurements, model calculations indicate that the background gas pressure in the trap is less than 10^{-10} Torr. This pressure is low enough to prevent the degration of the high charge states by charge exchange, but high enough to provide sufficient evaporative cooling for the highly charged ions. With an electron-beam current density of about 3000 A/cm², the experiments show that the extracted ions provide μ s-wide ion beam pulses of about 10⁴ ions per s. The duty cycle is determined by the time it takes for the charge-state distribution to reach steady state, which is about 1 s for Th and U. Further enhancements of the trap and the efficiency of the extraction system could eventually result in higher yields of highquality, high-charge, high-Z ions from the EBIT.

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- [1] E. D. Donets, IEEE Trans. Nucl. Sci. NS-23, 897 (1976).
- [2] E. D. Donets and V. P. Ovsyannikov, Zh. Eksp. Teor. Fiz.
 80, 916 (1981) [Sov. Phys.—JETP 53, 466 (1981)].
- [3] M. A. Levine, R. E. Marrs, J. R. Henderson, D. A. Knapp, and M. B. Schneider, Phys. Scr. **T22**, 157 (1988).
- [4] M. A. Levine, R. E. Marrs, J. N. Bardsley, P. Beiersdorfer,

C. L. Bennett, M. H. Chen, T. Cowan, D. Dietrich, J. R. Henderson, D. A. Knapp, A. Osterheld, B. M. Penetrante, M. B. Schneider, and J. H. Scofield, Nucl. Instrum. Methods Phys. Res. B **43**, 431 (1989).

[5] M. B. Schneider, M. A. Levine, C. L. Bennett, J. R. Henderson, D. A. Knapp, and R. E. Marrs, in *International* Symposium on Electron Beam Ion Sources and Their Applications, Proceedings of the International Symposium on Electron Beam Ion Sources and Their Applications, AIP Conf. Proc. No. 188, edited by A. Hershcovitch (AIP, New York, 1989), p. 158.

- [6] R. E. Marrs, M. A. Levine, D. A. Knapp, and J. R. Henderson, Phys. Rev. Lett. 60, 1715 (1988).
- [7] D. A. Knapp, R. E. Marrs, M. A. Levine, C. L. Bennett, M. H. Chen, J. R. Henderson, M. B. Schneider, and J. H. Scofield, Phys. Rev. Lett. 62, 2104 (1989).
- [8] R. E. Marrs, C. Bennett, M. H. Chen, T. Cowan, D. Dietrich, J. R. Henderson, D.A. Knapp, M. A. Levine, K. J. Reed, M. B. Schneider, and J. H. Scofield, J. Phys. (Paris) Colloq. 50, C1-445 (1989).
- [9] C. M. Brown, U. Feldman, G. A. Doschek, J. F. Seely, R. E. La Villa, V. L. Jacobs, J. R. Henderson, D. A. Knapp, R. E. Marrs, P. Beiersdorfer, and M. A. Levine, Phys. Rev. A 40, 4089 (1989).
- [10] P. Beiersdorfer, M. H. Chen, R. E. Marrs, and M. A. Levine, Phys. Rev. A 41, 3453 (1990).
- [11] P. Beiersdorfer, A. L. Osterheld, M. B. Schneider, W.

Goldstein, J. R. Henderson, D. A. Knapp, M. A. Levine, R. E. Marrs, and D. Vogel, Phys. Rev. Lett. 65, 1995 (1990).

- [12] J. R. Henderson, P. Beiersdorfer, C. L. Bennett, S. Chantrenne, D. A. Knapp, R. E. Marrs, M. B. Schneider, K. L. Wong, G. A. Doschek, J. F. Seely, C. M. Brown, R. E. LaVilla, J. Dubau, and M. A. Levine, Phys. Rev. Lett. 65, 705 (1990).
- [13] D. Schneider, D. DeWitt, M. W. Clark, R. Schuch, C. L. Cocke, R. Schneider, K. J. Reed, M. H. Chen, R. E. Marrs, M. Levine, and R. Fortner, Phys. Rev. A 42, 3889 (1990).
- [14] I. G. Brown, J. E. Glavin, R. A. MacGill, and R. T. Wright, Appl. Phys. Lett. 49, 1019 (1986).
- [15] B. M. Penetrante, J. N. Bardsley, M. A. Levine, D. A. Knapp, and R. E. Marrs, Phys. Rev. A 43, 4873 (1991).
- [16] B. M. Penetrante, J. N. Bardsley, D. DeWitt, M. Clark, and D. Schneider, Phys. Rev. A 43, 4861 (1991).
- [17] W. Lotz, Z. Phys. 206, 205 (1967); 216, 241 (1968).
- [18] Y. S. Kim and P. H. Pratt, Phys. Rev. A 27, 2913 (1983).
- [19] A. Müller and E. Salzborn, Phys. Lett. 62A, 391 (1977).