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## Production of Group IIA atomic and molecular negative ion beams in a cesium-sputter negative ion source

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### Abstract

The results of an investigation on the production of Group IIA atomic and molecular negative ion beams formed in a cesium-sputter negative ion source are presented. The sputtering material was formed by pressing pellets of stoichiometric mixtures of the Group IIA element carbonates and 10% copper powder. Negative ions of several alkaline-earth elements and their oxides have been observed. Beam intensities as high as 180 pA have been observed for  $\text{Sr}^-$  and 20 nA for  $\text{SrO}^-$ .

Negative ion sources have been an invaluable asset for tandem accelerator-based nuclear and high energy atomic physics research programs. They have also been valuable in tandem accelerator mass spectrometry, an experimental technique that is useful in eliminating isobaric contamination in mass spectra [1,2]. Typically, negative ion beams of the order of 1  $\mu\text{A}$  and above are required for tandem accelerators. A method for producing bright beams of negative ions is required for these applications.

Examples of negative ion sources include the cold-cathode Penning source [1,3] and the sequential charge exchange source [1,4]. The cold-cathode Penning source is an electron attachment source based in a plasma environment. The material to be ionized can be introduced into a discharge chamber as a gas through direct vaporization from an oven or through sublimation of a solid rod. The lifetime of the cathode limits the length of source operation. In a sequential charge exchange source, negative ions are produced when a positive ion beam sequentially captures two electrons as it interacts with a low ionization potential vapor target. Production efficiencies depend on the positive ion beam energy, the ionization energy of the donor atom, and the electron affinity of the atom under consideration. The highest production efficiencies occur when projectile-target combinations have a minimum energy defect in both charge exchange processes. For elements with very low electron affinities, production efficiencies are  $\sim 1\%$  or less.

The recent development of sputter-type negative ion sources [5] has provided a means of producing a large

variety of intense negative ion beams. Other benefits of sputter-type sources include their long lifetimes, low emittance, and ease of operation. The most prolific of the sputter-type sources is the cesium-sputter negative ion source. The ion source's operating principle is straightforward. Cesium atoms are thermally ionized and accelerated toward a coated target (pellet). The cesium ion beam forms a thin coating on the target and sputters matter from the target. Particles that are sputtered from the surface of the target produce low velocity atoms that can become negatively charged as they leave the cesiated surface of the pellet. Negative ions that are produced in the source, exit through an aperture and are further accelerated to ground potential as they travel toward an extraction electrode. Among other factors, the beam intensity depends on the electron affinity and the mass of the parent atom. Further details of the design and operation of the source are given in references by Alton [1,6].

The cesium-sputter negative ion source can produce negative ion beams of carbon and copper with intensities as large as 250 and 200  $\mu\text{A}$ , respectively [7]. However, intense negative ion beams consisting of Group IA and Group IIA elements have been difficult to produce using this ion source [4]. Such elements are considered as good candidates for tandem accelerator applications because of their atomic and nuclear properties. Because of the recent discovery of the stable ground states of  $\text{Ca}^-$ ,  $\text{Sr}^-$ , and  $\text{Ba}^-$  [8–10], the cesium-sputter negative ion source could also be used as an ion source for experiments designed to probe the atomic structure of these negative ions and study their atomic collision properties with atoms, molecules, electrons, and photons. Recently, Alton [11] showed that negative ion beams of Group IA elements can be produced

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with sputtering targets consisting of the alkali carbonates. The technique involved pressing target pellets from a stoichiometric mixture of the Group IA element carbonates with 10% metallic powder. The addition of metal powders into the pellet assists in dissipating heat produced during the sputtering process to an external cooling system. Ion beams of the order of  $1\ \mu\text{A}$  were produced with source lifetimes of the order of 24 hr. With one recent exception [9], the production yields of alkaline-earth negative ion beams formed in a cesium-sputter-source have not been reported. Reasons for this include the inability of Be and Mg to form stable negative ions, and the very small binding energies of Ca, Sr, and Ba [8–10]. In addition, the physical and chemical properties of these elements make target preparation difficult. This report describes an investigation on the feasibility of using a cesium-sputter negative ion source for producing beams of Group IIA negative ions for applications in tandem electrostatic accelerators. Since bound negative ions of Mg and Be only exist in doubly excited, metastable states [12,13], the focus of this report will be on the production of  $\text{Ca}^-$ ,  $\text{Sr}^-$ , and  $\text{Ba}^-$  ion beams and their oxides.

Fig. 1 displays a schematic diagram of the cesium-sputter negative ion source that was utilized in the present experiment. It is a commercially manufactured source, based on Alton's axially geometric design [6,7]. Cesium ions are produced by thermally ionizing cesium atoms with a cylindrical geometry tungsten ionizer heater. Typically, the current through the ionizer was 21.5 A. The acceleration energy (sputter probe voltage) of the cesium ions ranged from 1.4–4.4 keV. Using Alton's approach [11],

sample targets were prepared by pressing a stoichiometric mixture of the carbonates of Ca, Sr, and Ba with 10% Cu powder into pellets with a 6 mm diameter and 4 mm thickness. One advantage of using the Group IIA carbonates is that they are somewhat less hygroscopic and deliquescent than their oxides, and are less reactive than their pure elemental forms. Ions produced in the source were accelerated to energies ranging from 2–25 keV. After traveling approximately 2 m, the beam entered a  $90^\circ$  bending magnet for mass analysis. The resolution of the mass-analyzer was approximately 1%. The intensity of the mass-analyzed negative ion beam was measured with an insertable Faraday cup placed 2 m from the exit aperture of the magnet. The ion beamlines preceding and following the magnet included electrostatic steering and focusing elements, in order to maximize the ion beam current in the insertable Faraday cup.

Representative samples illustrating the measured yields of  $^{40}\text{Ca}^-$  and  $^{88}\text{Sr}^-$  as a function of the sputter probe voltage are presented in Fig. 2. The sputter probe voltage was limited to a maximum of 4.4 kV. Operation of the sputter probe above 4.4 kV led to high voltage arcing in the source with no apparent increase in beam. Since the bending magnet could not resolve the mass peaks of  $\text{Ba}^-$  and  $\text{BaH}^-$ , data illustrating the dependence of the  $\text{Ba}^-$  production as a function of sputter probe voltage have been omitted in Fig. 2. However, negative ion beams with intensities up to 70 pA were observed when the field of the bending magnet was set to pass negative ion with an  $m/q$  of 138.

The mass-analyzer was able to resolve the mass 86 and

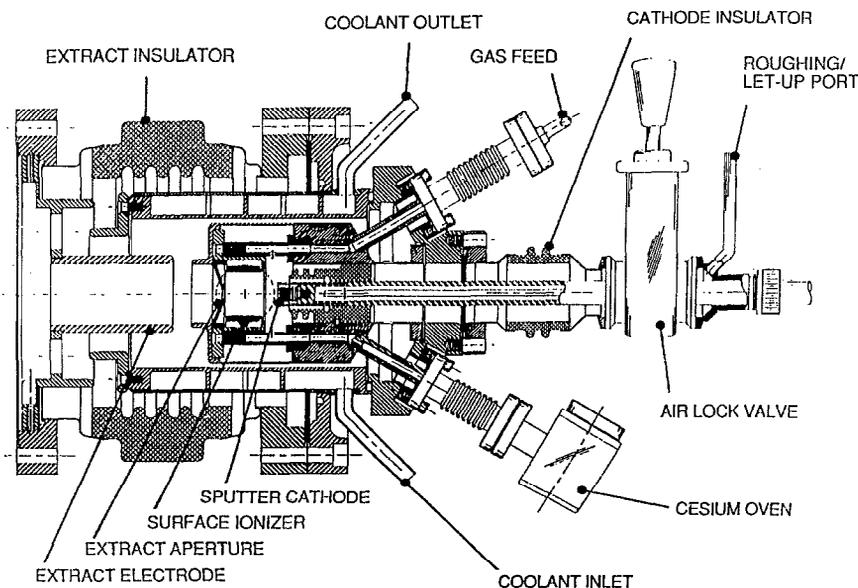


Fig. 1. Cross sectional view of the Kingston Scientific cesium-sputter negative ion source used for producing negative ions containing alkaline-earth elements.

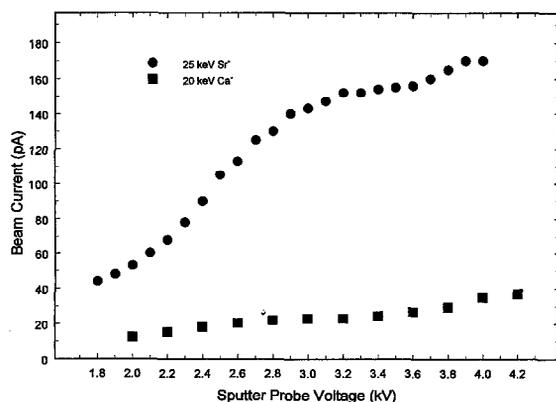


Fig. 2. Measured  $^{40}\text{Ca}^-$  and  $^{88}\text{Sr}^-$  beam intensities as a function of sputter probe voltage. The shaded squares represent 20 keV  $\text{Ca}^-$  yields that were produced from a sputtering target consisting of a  $\text{CaCO}_3$  and Cu mixture. The shaded circles represent 25 keV  $^{88}\text{Sr}^-$  yields that were produced from a sputtering target consisting of a  $\text{SrCO}_3$  and Cu mixture.

$^{88}$  isotopes of the  $\text{Sr}^-$  beam, and was also able to separate the  $\text{Ca}^-$  and  $\text{CaH}^-$  mass peaks. Contributions from  $^{86}\text{SrH}_2^-$ , and  $^{87}\text{SrH}^-$  to the  $m/q = 88$  peak were assumed to be small, since the sputtering pellets are neither reactive nor hygroscopic. Negative ion beams of Sr and Ca were produced at energies as low as 2 keV and 10 keV, respectively. Fig. 2 shows that the  $\text{Ca}^-$  and  $^{88}\text{Sr}^-$  yields were quite low. As a comparison, Heinicke et al. [14,15] and Kaiser et al. [16] have used a cold-cathode Penning ion source to produce  $\text{Ca}^-$  beams ranging from 1–20 nA. Kaiser et al. [14] also reported a yield of 20 nA for  $\text{Sr}^-$ . Alton et al. [3] have produced  $\text{Ca}^-$  through sequential charge exchange with Li vapor. In their investigation,  $\text{Ca}^-$  production efficiencies of less than 1% were measured, but beam intensities were not reported. Recently, Berkovits et al. [9] produced 2 pA of 115 keV  $^{88}\text{Sr}^-$  beam with a cesium-sputter source and pure Sr target. Their apparatus was able to distinguish between  $^{80}\text{SrH}_2^-$ ,  $^{88}\text{Sr}^-$ , and  $^{87}\text{SrH}^-$ , using a tandem accelerator mass spectrometry technique. Although the yields shown in Fig. 2 are sufficient for atomic and molecular physics investigations, they are still quite small for tandem electrostatic accelerator applications.

For most applications, molecular negative ion beams are less desirable than those composed of atomic negative ion beams. This is a result of energy partitioning among the fragments produced upon collisional dissociation in the stripping process. In some circumstances, however, negative molecular ions containing the elements of interest have larger electron affinities than their atomic counterparts. As a result, the molecular ions are formed with a much higher probability than the elemental species of

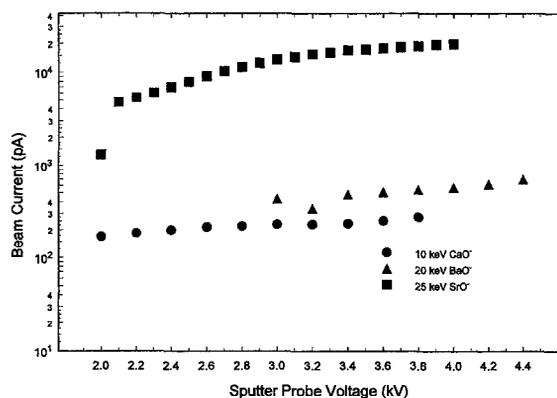


Fig. 3. Measured  $^{40}\text{CaO}^-$ ,  $^{88}\text{SrO}^-$ , and  $^{138}\text{BaO}^-$  beam intensities as a function of sputter probe voltage. The shaded circles represent 10 keV  $^{40}\text{CaO}^-$  yields that were produced from a sputtering target consisting of a  $\text{CaCO}_3$  and Cu mixture. The shaded squares represent 25 keV  $^{88}\text{SrO}^-$  yields that were produced from a sputtering target consisting of a  $\text{SrCO}_3$  and Cu mixture. The shaded triangles represent 20 keV  $^{138}\text{BaO}^-$  yields that were produced from a sputtering target consisting of a  $\text{BaCO}_3$  and Cu mixture.

interest. An attempt has been made to produce negative ion beams of the Ca, Sr, and Ba oxides from target pellets consisting of the heavier Group IIA carbonates. The results are shown in Fig. 3. In each case, the oxide negative ion yields are significantly higher than the atomic ions. It may be possible to improve these yields by using pellets consisting of the Group IIA oxides. However, their physical and chemical properties make target preparation and handling difficult. Negative ion beams consisting of the carbides of Ca and Sr were also observed, but were found to be significantly smaller than the oxide beams.

In summary, the production yields of the Group IIA atomic and molecular ions have been investigated with  $\text{CaCO}_3$ ,  $\text{SrCO}_3$ , and  $\text{BaCO}_3$  sputter probes. Although the yields are quite sufficient to study their atomic structure and interactions with photons and particles, the beam yields are still not adequate for most other applications. Other sputter probes consisting of the atomic species of interest could prove to be viable alternatives for producing intense beams of  $\text{Ca}^-$ ,  $\text{Sr}^-$ , and  $\text{Ba}^-$  or intense molecular negative ion beams with  $\text{Ca}^-$ ,  $\text{Sr}^-$ , or  $\text{Ba}^-$  constituents.

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