RECENT ADVANCEMENTS IN HIGH INTENSITY HEAVY NEGATIVE ION
SOURCES BASED ON THE SPUTTER PRINCIPLE*

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ABSTRACT

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Significant advancements have been recently made in the technology of forming high intensity heavy negative ion beams. Due to their ease of operation, simplicity, long lifetime, and wide range of species capabilities, negative ion sources predicated on the sputter principle are being used for an increasing number of diverse applications, including standard tandem accelerator-based research, high-energy ion implantation, tandem accelerator mass spectrometry, and low-energy atomic physics research. Sources have been developed which utilize either (1) direct surface ionization, or (2) a plasma to form the positive ion beam used to effect sputtering of samples containing the material of interest. This article will include a brief review of the fundamental processes underlying negative ion formation in these sources. Emphasis will be placed on the descriptions and performance characteristics of specific sources based on each of the positive ion formation techniques.

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1.0 INTRODUCTION

The technique of sputtering a surface covered with a fractional layer of a highly electropositive adsorbate material such as cesium has proved to be a universal method for generating atomic and molecular negative ion beams from most chemically active elements. In addition to being versatile in terms of species, sources based on this concept are simple in design, easy to operate, and generally have long lifetimes. Because of these factors, such sources are utilized extensively in most tandem electrostatic accelerator heavy ion physics research laboratories, as well as for use in a growing number of other applications, including high-energy ion implantation and tandem accelerator mass spectrometry.

Although the physio-chemical processes responsible for negative ion formation during sputtering of a low-work-function surface are complicated, the practical application of the technique is quite simple. Positive ion beams, usually formed by either direct surface ionization of a Group IA element or in a heavy noble gas (Ar, Kr, or Xe) plasma discharge seeded with alkali metal vapor, are accelerated to energies between a few hundred eV and several keV where they sputter a sample containing the element of interest. The presence of a fractional layer of a highly electropositive adsorbate material is critically important to the enhancement of negative ion formation during the sputtering process. A small fraction of the sputter ejected particles leave the adsorbate covered surface as negative ions and are accelerated through an extraction aperture in the source. Although other negative ion formation processes may be responsible for negative ion formation, particularly during the sputtering of compound or alloyed materials, the mechanism of formation during sputtering from a low-work-function surface is believed to be a form of or closely allied with surface ionization. Several sources predicated on this principle have been developed, a few of which are described in Refs. 1-5.

In this report, we shall assume the validity of the surface ionization mechanism as proposed by Nørskov and Lundqvist [6] and use it in conjunction with the neutral particle energy-angular distribution function of Thompson [7] to form a composite model for estimating the probability for negative ion formation and to illustrate the sensitivity of the sputter negative ion generation process to changes in surface work function ϕ . A brief description of a scaling technique for calculating sputter ratios from the Sigmund theory [8] will also be provided. Several negative ion sources (e.g., those reported in Refs. 1-5) which exemplify the practical realization of this technology will be discussed in terms of their performance characteristics. The list of sources described in this review includes a recently developed, high-brightness, pulsed-mode, plasma sputter negative ion source [5] which exhibits mA intensity capabilities. This source shows considerable promise for tandem electrostatic accelerator/synchrotron injection applications.

2.0 FUNDAMENTALS OF THE SPUTTER NEGATIVE ION GENERATION PROCESS

The adsorption of less than a monolayer of a highly electropositive adsorbate material on the surface of a sample undergoing particle bombardment greatly enhances the probability for secondary negative ion formation [9]. While it is possible that other independent and distinct negative ion formation processes may coexist during sputtering, there is convincing evidence that the mechanism of ion formation during sputtering of a metal surface covered with a partial layer of a material such as cesium is a form of surface ionization. Experimental evidence in support of this mechanism has been provided by Yu [10], who was able to show a direct correlation between change in work function during adsorption of cesium on clean Mo and the yields of Mo^- ions ejected from the surface during bombardment with a Ne^+ beam.

Progress has been made over the past few years toward a quantitative understanding of the secondary negative ion formation process. The process can be formulated in terms of a composite, idealistic, but heuristically useful model. The negative ion current I^- generated by sputtering can be expressed by the following simple expression:

$$I^- = 2\pi I^+ (E_1) S(E_1, \theta_i) \int_{V_i}^{E'} \int_0^{\pi/2} P^-(E_2, \theta) f_o(E_2, \theta) \sin\theta d\theta dE_2. \quad (1)$$

The model does not account for the destructive effects at high positive ion current densities or interference to sputtering at high adsorbate surface coverages and assumes that a uniform and constant adsorbate coverage can be maintained during the sputtering process. In Eq. 1, I^+ is the incident positive ion current of energy E_1 , $S(E_1, \theta_i)$ is the sputter ratio of the target at projectile energy E_1 and angle of incidence with respect to the surface normal θ_i , $f_o(E_2, \theta)$ is the neutral atom energy-angular distribution function and $P^-(E_2, \theta)$ is the probability for negative ion formation of atoms ejected at energy E_2 and polar angle θ with respect to the surface normal. The limits of integration for energy E_2 are V_i , the image potential induced in the surface by the departing negative ion, and E' , the maximum energy transfer that can occur between a projectile of energy E_1 and mass M_1 and a target atom of mass M_2 . E' is given by the relation $E' = 4M_1M_2E_1 / (M_1 + M_2)^2$. The positive ion current I^+ can, in principle, be measured or estimated and the sputter ratio S can be readily calculated by scaling Sigmund theory [8] into agreement with experiment.

2.1 The Positive Ion Current I^+

Several sources, such as those described in Refs. 1-5, utilize either direct surface ionization of cesium vapor as it comes in contact with a hot, high-work-function surface [1-3] or a plasma [4-5] to form positive ions which are then accelerated against a negatively biased probe containing the material of interest. In the latter type of source, the plasma is usually that of a heavy noble gas discharge (Ar, Kr, or Xe) seeded with cesium vapor.

The positive ion currents achievable in sources which utilize direct surface ionization for positive ion formation or extract positive ions from a plasma can attain values up to the space charge limit of the particular electrode configuration. For such sources, the perveance P of the electrode configuration can be measured or computationally determined by solving Poisson's equation by use of computer codes such as described in Refs. 11 and 12. The perveance P is defined by the following relationship between the positive ion current I^+ and the potential difference V between the sample and surface ionizer or plasma:

$$I^+ = PV^{3/2}. \quad (2)$$

The perveance is a function of the geometry of the electrode system or plasma/sputter probe interface and mass M of the bombarding ion species. The geometry of the electrode system, of course, determines the optical properties of the particular source. From the standpoint of beam quality in sources which are predicated on cesium surface ionization, a focusing system is desirable. That is, the emittance of this type of source will be affected by the physical size of the negative ion generation region which is determined by the positive ion current density distribution on the sample surface.

Figure 1 illustrates the relationship between the positive cesium ion current I^+ and the perveance P in cesium surface ionization type negative ion sources equipped with various ionizer geometries. The perveances were computed by the use of the computer code described in Ref. 11. Since we assume that the negative ion current increases in proportion to the positive ion current, the perveance P serves as convenient measure for comparing ion sources which operate up to the space charge positive ion beam limit.

2.2 Estimation of Target Sputtering

The Sigmund theoretical model [8] is the most widely accepted and used model for estimating the number of atoms ejected per incident ion from an amorphous surface during energetic particle bombardment. While the theory predicts shapes of sputter ratio S versus projectile energy E_1 in good agreement with experimental data, calculated values deviate from experimental observation by substantial amounts. In order to utilize the excellent features of the model and to be able to calculate sputter ratios which are in close agreement with experiment, we have developed a computer code for scaling Sigmund theory into agreement with experiment. The scaled Sigmund theoretical formulas for S become

$$S(E_1, \theta_i) = \frac{A_1 3\lambda \alpha E_1}{4\pi^2 U_0 (\cos \theta_i)^F} \quad (\text{for } E_1 \leq 1000 \text{ eV}), \quad (3)$$

$$S(E_1, \theta_i) = \frac{A_2 0.042 \alpha S_n(E_1)}{U_0 (\cos \theta_i)^F} \quad (\text{for } E_1 \geq 1000 \text{ eV}), \quad (4)$$

where α is a tabulated function of the ratio of the target mass M_2 , to projectile mass M_1 , U_0 is the heat of sublimation of the target, θ_i is the angle between the incident projectile and the surface normal. F usually has values between 1 and 1.7, depending on the ratio of target to projectile mass. The remaining terms in the first equation are defined as follows:

$$\lambda = 4M_1M_2/(M_1 + M_2)^2,$$

$$S_n(E_1) = 4\pi Z_1 Z_2 e^2 a_{12} [M_1/(M_1 + M_2)] S_n(\epsilon),$$

where Z_1, Z_2 are the respective projectile and target atomic numbers,

$$a_{12} = 0.8853 a_0 \left(Z_1^{2/3} + Z_2^{2/3} \right)^{-1/2},$$

a_0 is the radius of the first Bohr orbit, $S_n(\epsilon)$ is the projectile stopping power in the target with

$$\epsilon = \frac{M_2 E_1 (M_1 + M_2)}{Z_1 Z_2 e^2 / a_{12}},$$

and A_1, A_2 are scaling parameters which are determined from algorithms derived from experimental data. No provisions have been made in the scaling procedure to account for the effect of the presence of surface adsorbates on the sputtering process.

Sputter ratios versus cesium projectile energies, estimated by use of a computer code based on a modified form of Eqs. 3 and 4 are shown in Fig. 2 for the Group IB elements Cu, Ag, and Au. This scaling procedure has proved to be an

accurate and useful means for correlating theory and experiment and will be the subject of a future report.

2.3 The Probability for Negative Ion Formation

2.3.1 Negative Ion Probability Distribution Function. In the prescription of Nørskov and Lundqvist [6], the probability for negative ion formation during sputtering can be represented by the following simple energy-dependent relation:

$$P^-(E_2, \theta) = \frac{2}{\pi} e^{-\beta \sqrt{M_2} \{ \phi(\sigma) - E_A + V_i \} / \sqrt{2 E_2} \cos \theta} \quad (5)$$

In Eq. 5, ϕ is the work function of the surface which is a function of the relative adsorbate coverage σ , E_A is the electron affinity of the ejected particle of mass M_2 and energy E_2 , V_i is the image potential induced in the surface by the escaping ion, θ is the polar angle of the sputtered ion with respect to the surface normal and β is a constant. In Eq. 5, $\sqrt{2 E_2 / M_2} \cos \theta = v_{\perp}$ is the component of the velocity of the escaping particle perpendicular to the metal surface. Experimental evidence in support of the velocity dependence of the secondary ion formation process has been provided by Yu for negative ions [13].

2.3.2 Neutral Particle Energy-Angular Distribution Function. Under the assumption of isotropic collision cascades and a planar surface potential E_b , Thompson [7] derived the following energy-angular distribution function for sputter ejected atoms:

$$f_0(E_2, \theta) = \frac{A_0 E_2 \cos \theta}{(E_2 + E_b)^3} \quad (6)$$

In Eq. 6, E_2 is the energy of the ejected particle and θ is the polar angle with respect to the normal of the surface. The maximum in the distribution occurs at $E_{2\max} = E_b/2$ and is independent of emission angle θ . The normalization constant A_0 , is determined by requiring that the integral $2\pi \int_{V_i}^E \int_0^{\pi/2} f(E_2, \theta) \sin\theta d\theta dE_2 = 1$ where V_i is the image potential induced in the surface by the departing negative ion and E' is given by $E' = 4M_1M_2E_1/(M_1 + M_2)^2$.

2.3.3 Electropositive Adsorbate Effects. Atomic adsorption of a dissimilar element on a clean surface affects the surface work function [14]. The magnitude and sign of the change depends on the chemical properties of the adsorbed atom (adsorbate) and those of the host material (adsorbent). Electropositive atoms decrease the work function while electronegative atoms tend to increase the work function. Simple analytical expressions have been derived by Alton [15] which can be used to predict, with good accuracy, the value of the work function at its minimum and over the complete range ($\sigma = 0$ to $\sigma = 1$) of adsorbate coverage for many alkali and alkaline earth-metal adsorbate-adsorbent combinations. The relationship of Ref. 15 is given by

$$\phi(\sigma) = \phi_0 + \frac{6\Delta\phi_m \sigma}{(3-\sigma_m)\sigma_m} - \frac{3\Delta\phi_m(\sigma_m+1)\sigma^2}{(3-\sigma_m)\sigma_m^2} + \frac{2\Delta\phi_m\sigma^3}{(3-\sigma_m)\sigma_m^2} [V], \quad (7)$$

where ϕ_0 is the intrinsic work function of the surface at $\sigma = 0$ and $\Delta\phi_m$ is the maximum change in work function which occurs at a relative adsorbate coverage value of $\sigma = \sigma_m$. The maximum change $\Delta\phi_m$ in the work function at σ_m is given by

$$\Delta\phi_m \equiv -1.24 \left[\phi_0 - \frac{1}{2} (I_A + E_A) \right] [V], \quad (8)$$

where I_A is the first ionization potential and E_A the electron affinity of the adsorbate material. Expression 8 reproduces, with good accuracy, maximum work function changes observed for alkali and alkaline earth metal adsorbates. The model described by Eq. 7 is practically useful for estimating work function changes induced in surfaces by sputtering of electropositive adsorbate materials and subsequently in work-function-dependent models, such as that of Nørskov and Lundqvist [6].

2.3.4 The Energy Distributions of Neutral Particles and Negative Ions. Figure 3 shows the energy distributions of neutral and negative ions sputtered from a Mo surface at four surface coverages σ as determined by integrating, respectively, the Thompson [7] relation (Eq. 6) and the product of the Thompson relation and the Nørskov and Lundqvist [6] relation (Eq. 5) over the complete range of polar angle $0 \leq \theta \leq \pi/2$. We use Eq. 7 to simulate the work function ϕ versus relative cesium coverage σ . The parameter β was obtained from the fits to the Yu data of Ref. 10 with $V_i = 1.4$ eV; the electron affinity E_A and intrinsic work function ϕ_0 for Mo were used in Eq. 7 for calculating the work function ϕ at a particular cesium surface coverage σ . We note that the peaks in the negative ion energy distribution functions shift toward lower energies as the work function ϕ is lowered and reach the left-most position whenever $\sigma = 0.5$; at higher relative coverages, the distributions shift toward the right-hand side of the energy scale. The maximum probability for negative ion formation occurs for a cesium adsorbate coverage of $\sigma = 0.5$, in agreement with the observations of Yu. Thus, the energy distributions of secondary negative ions in the energy-dependent prescription of Nørskov and Lundqvist are seen to be sensitive in shape and position to surface work function ϕ and adsorbate coverage σ .

2.3.5 Total Probability for Negative Ion Formation. In order to determine the total probability for negative ion formation at a particular surface coverage σ , one must integrate the product function formed with Eq. 5 and Eq. 6 over the complete range of particle ejection energy E_2 and over all polar angle θ . Figure 4 displays total probability P_T^- versus cesium surface coverage σ as predicted for Mo^- . The results displayed in Fig. 4 should reflect the correct shape of the total probabilities versus relative cesium adsorbate coverage σ , if not absolute values.

3.0 SOURCES BASED ON CESIUM SURFACE IONIZATION

Many sources utilize direct surface ionization of cesium vapor as it comes in contact with a hot, high-work-function surface to form positive ions which are then accelerated against a negatively biased probe containing the material of interest. In this source type, the cesium acts both to sputter the sample, as well as, along with the neutral adsorbed cesium, to effect changes in the surface work function. The positive ion currents achievable in such sources can attain values up to the space charge limit of the particular electrode configuration as given by Eq. 2. The geometry of the ionizer/sputter sample system dictates the focal properties of the cesium ion beam at impact. From the standpoint of low emittance, a small negative ion generation region is important. Therefore, a focusing geometry electrode system is desirable. Several negative ion sources based on direct surface ionization of cesium are described below. All of the sources were designed and developed at the Oak Ridge National Laboratory.

Computer simulation has proved to be a valuable and cost effective tool for assisting in the design of ion sources, as well as evaluating the optical properties of

existing source designs. The computer codes described in Refs. 11 and 12 are well suited for such applications. This technique is illustrated through Figs. 5, 6, and 7, which display, respectively, the positive cesium ion optics, positive cesium ion current density distribution at the sample surface, and the negative ion optics of a prototype source under design at the Oak Ridge National Laboratory. The source is equipped with a spherical geometry ionizer and provisions for remotely selecting any one of sixty samples. The negative ion current density distribution at the sample surface is taken to mimic that of the corresponding positive cesium ion current.

The performance characteristics and emittances of sources equipped with spherical, ellipsoidal, and cylindrical geometry ionizers are described below.

3.1 The Source Equipped with a Spherical Geometry Ionizer

The space charge limited optics for cesium ion flow in a source equipped with a spherical geometry ionizer is shown schematically in Fig. 8. This source has been described previously [3] and the emittance of the source is reported in Refs. 16-18. The positive cesium ion beam current density distribution at impact with the sample surface is typically ~ 0.75 mm full diameter when the sample is positioned at the focal point of the system. The computed perveance for cesium in this electrode configuration is $P = 2 \times 10^{-9} [A/V^{3/2}]$.

3.2 The Source Equipped with an Ellipsoidal Geometry Ionizer

This source is briefly described in Refs. 17 and 18, along with the emittance data for the source. Figure 9 illustrates the optics for space charge limited cesium flow in this highly converging electrode system. When placed at the focal point of the

electrode system, the sample wear pattern has a diameter $\phi \cong 1.25$ mm. This electrode configuration has a high perveance in relation to other focusing systems. The computationally determined perveance for the electrode system is found to be $P = 17 \times 10^{-9} [A/V^{3/2}]$.

3.3 The Source Equipped with a Cylindrical Geometry Ionizer

The cylindrical geometry ionizer source configuration, shown in Fig. 10, has been described in detail in Refs. 2 and 3 and the emittance and brightness characteristics of the source have been reported in Refs. 16-18. The observed wear pattern from this source is composed of two parts: a region of concentrated wear with full diameter of ~ 0.75 mm, and a low-density, uniform-wear region with a diameter of ~ 4.5 mm. Because of the greater size of the region of negative ion generation, the emittance of this source is expected to be larger than those of the previously described sources equipped with spherical and ellipsoidal geometry ionizers. The computed perveance of this ionizer geometry for cesium is $P \cong 57 \times 10^{-9} [A/V^{3/2}]$.

3.4 Negative Ion Beam Intensity Data

The versatility of the sources described above is reflected by the wide spectrum of momentum analyzed negative ion beams that have been observed during their operation. A partial list of species and negative ion beam intensities realized from these sources under a variety of operating conditions are the following: 50 μA H^- , 8 μA Li^- , 3 μA , 10 μA B^- , 270 μA C^- , 280 μA O^- , 30 μA F^- , 3 μA Na^- , 1 μA MgH^- , 2.8 μA Al^- , 100 μA Si^- , 1 μA P^- , 100 μA S^- , 30 μA Cl^- , 0.5 μA K^- , 0.4 μA CaH^- , 2 μA ScH^- , 15 μA TiH_3^- , 4 μA CrH_2^- , 1 μA MnO^- , 0.6 μA FeO^- , 1 μA Co^- , 6 μA CoO^- , 150 μA Ni^- , 200 μA Cu^- , 1 μA ZnO^- , 4 μA GaO^- , 2 μA Ge^- , 40 μA As^- , 75 μA Se^- ,

25 $\mu\text{A Br}^-$, 0.5 $\mu\text{A Rb}^-$, 0.6 $\mu\text{A YO}^-$, 18 $\mu\text{A ZrH}$, 0.4 $\mu\text{A Nb}^-$, 1.0 $\mu\text{A NbO}^-$, 1.0 $\mu\text{A MoO}^-$,
 0.5 $\mu\text{A Pd}^-$, 18 $\mu\text{A Ag}^-$, 0.5 $\mu\text{A CdO}^-$, 0.5 $\mu\text{A Sn}^-$, 0.9 $\mu\text{A Sb}^-$, 30 $\mu\text{A I}^-$, 0.2 $\mu\text{A Cs}^-$,
 4 $\mu\text{A TaO}^-$, 0.7 $\mu\text{A WO}^-$, 1 $\mu\text{A ReO}^-$, 0.3 $\mu\text{A Os}^-$, 5 $\mu\text{A Ir}^-$, 75 $\mu\text{A Pt}^-$, 170 $\mu\text{A Au}^-$,
 0.5 $\mu\text{A TlO}^-$, 0.4 $\mu\text{A PbO}^-$, 1 $\mu\text{A PbO}_2^-$, 0.6 $\mu\text{A Bi}^-$, 0.6 $\mu\text{A UC}_2^-$.

3.5 Emittance Data

Average normalized emittance data for the sources described above are displayed in Fig. 11 as a function of total percent of negative ion beam intensity. The emittance values are defined by the following relations:

$$\epsilon_{ny} = \pi \int \int dx dx' \sqrt{E} \text{ (x-direction) },$$

$$\epsilon_{ny} = \pi \int \int dy dy' \sqrt{E} \text{ (y-direction) },$$

$$\text{and } \epsilon = (\epsilon_{nx} \cdot \epsilon_{ny})^{1/2}. \quad (9)$$

In Eqs. 9, x, y are position coordinates, x', y' are angular coordinates, and E is the energy of the ion beam. Emittance in this prescription is usually given in units of $\pi \text{ mm.mrad (MeV)}^{1/2}$. The definitions given by Eqs. 9 are used for the emittances of all sources described in this report.

4.0 PLASMA SPUTTER NEGATIVE ION SOURCES

The advantage of the plasma-type source lies in the fact that, when operated in a high-density plasma mode, the negatively biased sputter probe containing the material of interest is uniformly sputtered. This characteristic makes it possible to take

advantage of the large area spherical geometry optics which occur between the spherical sector sputter probe and the plasma sheath surrounding the probe.

Negative ions created in the process are accelerated and focused through the plasma to a common focal point which is usually chosen as the ion exit aperture. Thus, high beam intensities can often be realized while preserving a reasonable emittance value. The plasma discharge in this type of source is usually formed from a heavy noble gas (Ar, Kr, or Xe) seeded with cesium vapor.

4.1 The Radial Geometry Plasma Sputter Source

An example of a plasma-type source is that described in Ref. 4. In this source, which embodies some of the principles associated with the radial geometry source developed by Tykesson and Andersen [19], a weak magnetic field (~ 150 G) is used to collimate the primary electron beam which is thermionically emitted by a tantalum filament located at the end of the ionization chamber. The electron beam produces an approximately uniform plasma by collisional impact with neutral cesium vapor introduced into the chamber from the externally mounted oven. Auxiliary discharge support gas (usually Ar) is introduced into the chamber to supplement the cesium vapor; chemically active gases may also be introduced into the chamber for generation of atomic negative ions from the gas itself or for chemical combination with the sputter probe material in the formation of molecular negative ions. The sputter sample is cylindrical (typically, 10 mm in diameter) with a concave spherical negative ion emission surface machined into the face of the material of radius $\rho = 15$ mm. The sputter probe is maintained at ~ -1000 V relative to the discharge chamber. Fig. 12 displays the negative ion optics of the radial geometry cesium plasma source described in Ref. 4.

4.1.1 Negative ion beam intensity data. Among the ions and intensities that have been reported from this source are the following: 175 $\mu\text{A H}^-$, 0.4 $\mu\text{A Li}^-$, 4 $\mu\text{A BeH}_3^-$, 25 $\mu\text{A BeO}^-$, 0.6 $\mu\text{A B}^-$, 20 $\mu\text{A C}^-$, 20 $\mu\text{A C}_2^-$, 30 $\mu\text{A O}^-$, 20 $\mu\text{A F}^-$, 12 $\mu\text{A S}^-$, 12 $\mu\text{A MgH}_3^-$, 20 $\mu\text{A Si}^-$, 20 $\mu\text{A F}^-$, 2.5 $\mu\text{A Al}^-$, 9 $\mu\text{A I}_2^-$, 2 $\mu\text{A Ca H}_3^-$, 2.5 $\mu\text{A Ti H}_3^-$, 55 $\mu\text{A Ni}^-$, 50 $\mu\text{A Cu}^-$, $\sim 1 \mu\text{A Sr H}_3^-$, 35 $\mu\text{A Ag}^-$, 3.1 $\mu\text{A TaN}^-$, 1.4 $\mu\text{A W}^-$, 80 $\mu\text{A Au}^-$, and 7 $\mu\text{A PbN}^-$.

4.1.2 Emittance data. The normalized emittance (as defined by Eqs. 9) versus percent total negative ion beam current for a 48 $\mu\text{A } ^{197}\text{Au}^-$ ion beam is shown in Fig. 13. More detailed information concerning the emittance of this source is given in Ref. 19.

4.2 The High-Intensity Plasma Sputter Negative Ion Source

The multi-cusp magnetic field plasma surface source, routinely employed for the production of high-intensity, pulsed H^- ion beams at LAMPF [20] and at the National Laboratory for High Energy Physics [21] has recently been modified for use as a high-intensity pulsed-mode heavy negative ion source [5].

For heavy negative ion generation, a high-density plasma discharge, seeded with cesium vapor, is produced by pulsing the discharge voltage of two series connected LaB_6 cathodes. In order to produce higher heavy negative ion beam intensities by sputter ejection at a given probe voltage, a chemically inert, heavy discharge support gas such as Ar, Kr, or Xe, is utilized. Cesium is introduced into the discharge from an external cesium oven. The sheath surrounding the negatively biased sputter probe (spherical radius, $\rho = 140 \text{ mm}$ and diameter, $\phi = 50 \text{ mm}$) which is made of the material of interest and is maintained at a negative voltage relative to

housing, (typically, -500 to -2000 V) serves as the acceleration gap and lens for focusing the ion beam through the exit aperture (diameter, $\phi = 18$ mm). When operated in the pulsed mode, the source holds considerable promise for use in conjunction with tandem electrostatic accelerator/synchrotron injection applications for heavy ion research. To date, the source has principally been tested in a low-duty-cycle (repetition rate: 1-50 Hz) macropulsed mode (pulse width: 50-300 μ s). The negative ion generation and ion extraction regions of a prototype source under design at the Oak Ridge National Laboratory is shown in Fig. 14. Computational simulation of the optics of this source for heavy negative ion generation using the code described in Ref. 12 is displayed in Fig. 15 for 3.5 mA O^- and 1 mA Au^- ion beams.

4.2.1 Negative ion beam intensity data. Table 1 provides a partial list of total beam intensities, species, and probe materials utilized during operation of the source in pulsed mode. It should be noted that the peak negative ion beam intensities are frequently 100 times or more greater than the cesium surface ionization sources described previously.

4.2.2 Emittance data. Examples of beam emittances versus percent total negative ion beam are shown in Fig. 16 for Ni beams with peak pulse intensities of 2.5 and 6 mA. The emittances are seen to increase in proportion to the ion beam intensity as expected from space charge considerations. At the 80% contour level, the emittance values are comparable to those for the sources described previously when operated in the pulsed mode, yet the beam intensities are often 100 times greater.

5.0 CONCLUSIONS

Considerable progress has been made toward a quantitative understanding of the mechanisms underlying negative ion formation during sputtering of a surface covered with small amounts (< 1 monolayer) of a highly electropositive adsorbate material such as cesium. The development of models such as described in this paper have moved experiment and theory to closer agreement. There is, however, a need for additional experimental data before a proper assessment of the validity of models such as the one proposed by Nørskov and Lundqvist [6] can be made.

The sputter principle has proved to be a simple and almost universal method for efficient formation of negative ion beams from chemically active elements. Heavy negative ion sources predicated on this principle, such as described in this report, continue to be improved in terms of intensity and beam quality. Recent advancements in sputter ion source technology are exemplified by the high intensity pulsed mode plasma sputter source which can produce intensity levels of several mA from a wide spectrum of negative ion species.

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Table 1. A partial list of total heavy negative ion beam intensities (peak) from the high-intensity plasma sputter heavy negative ion source [5].

Sputter Probe Material	Sputter Probe Voltage (V)	Geometry	Total Peak Beam Intensity (mA)	Species (%)
Ag	937	Spherical	6.2	Ag ⁻ (91)
Au	437	Spherical	10.3	Au ⁻ (73)
Bi	937	Spherical	2.7	Bi ⁻ (6); O ⁻ (42)
C	937	Spherical	6.0	C ⁻ (36); C ₂ ⁻ (58)
Co	937	Spherical	6.0	Co ⁻ (85)
Cu	438	Spherical	8.2	Cu ⁻ (77)
CuO	438	Flat	4.5	Cu ⁻ (40); O ⁻ (60)
GaAs	937	Flat	3.7	As ⁻ (20); As ₂ (52)
GaP	937	Flat	1.8	P ⁻ (44)
Mo	438	Spherical	30.0	O ⁻ (67)
Ni	438	Spherical	6.0	Ni ⁻ (87)
Pd	937	Spherical	7.6	Pd ⁻ (69)
Pt	937	Spherical	8.1	Pt ⁻ (71)
Si	937	Spherical	6.0	Si ⁻ (75)
Sn	937	Spherical	3.6	Sn ⁻ (67)

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Figure Captions

1. Cesium ion current I^+ versus perveance P for various cesium sputter negative ion sources designed at the Oak Ridge National Laboratory. Ionizer geometry: x spherical, single sample; O prototype spherical, multiple sample; □ ellipsoidal; ● cylindrical, bore diameter: 25.4 mm; ■ cylindrical, bore diameter: 12.7 mm.
2. Sputter ratio as a function of cesium projectile energy for the Group 1B elements estimated from scaled Sigmund theory.
3. Energy distribution of sputtered Mo neutral atoms and Mo^- ions calculated by integrating Eqs. 5 and 6, respectively. The adsorbate is assumed to be cesium. Three relative coverages were used in the calculations: $\sigma = 0.25$, $\sigma = 0.5$, and $\sigma = 0.75$. The parameters β and V_i were obtained from the fits to the Mo^- versus relative cesium coverage data of Yu (Ref. 10).
4. Total probability P_T^- for Mo^- formation versus relative cesium coverage σ determined by integrating the product of Eqs. 5 and 6 over the complete range in energy and angle.
5. The positive cesium ion optics for space charge limited flow in the prototype cesium sputter negative ion source under design at the Oak Ridge National Laboratory. The source is equipped with a spherical geometry cesium surface ionizer and a multiple sample holder.

6. The cesium positive ion beam current density distribution at the sample surface of the prototype cesium sputter negative ion source shown schematically in Fig. 5.
7. The negative ion optics in the prototype cesium sputter negative ion source under design at the Oak Ridge National Laboratory. The negative ion beam current density distribution at the sample surface is assumed to mimic that of the positive ion current density distributions shown in Fig. 6.
8. Positive cesium ion optics for space charge limited flow in the cesium sputter negative ion source equipped with a spherical geometry cesium ionizer (Ref. 3).
9. Positive cesium ion optics for space charge limited flow in the cesium sputter negative ion source equipped with an ellipsoidal geometry cesium ionizer (Refs. 17 and 18).
10. Positive cesium ion optics for space charge limited flow in the cesium sputter negative ion source equipped with a cylindrical geometry cesium ionizer (Ref. 2).
11. Average normalized emittance ϵ versus percent total negative ion beam for the negative ion sources equipped with spherical (Ref. 3), ellipsoidal (Refs. 17 and 18), and cylindrical (Ref. 2) geometry cesium surface ionizers.
12. Computational simulation of the negative ion optics of the radial geometry cesium plasma source (Ref. 4).

13. Normalized emittance ϵ versus percent total negative ion beam for the radial geometry source described in Ref. 4. The total beam for this measurement was $48 \mu\text{A } ^{197}\text{Au}^-$.
14. The negative ion generation regions of a prototype, axial geometry, pulsed mode, negative ion source based on the principle of the source described in Ref. 5.
15. Computational simulation of the negative ion optics of the prototype, pulsed mode, negative ion source, shown schematically in Fig. 14.
16. Normalized emittance ϵ versus percent total negative ion beam for the high-intensity, plasma-sputter, negative ion source described in Ref. 5.

Fig. 1

Fig. 2

Fig. 3

(X 10⁻³)

TOTAL PROBABILITY P_T

$$P_T = 2\pi \int_0^{\pi/2} \int_{V_i}^{E_i} f_0 P \sin \theta d\theta dE_2$$

RELATIVE CESIUM COVERAGE σ

Fig. 4

Fig. 5

Fig. 6

Fig. 7

Fig. 8

Fig. 9

Fig. 10

Fig. 11

(a) EXISTING ION OPTICS: $j = 0.2 \text{ mA/cm}^2$; $\theta_{\text{rms}} = 2.33^\circ$

(b) PROPOSED LOW ANGULAR DIVERGENCE ION OPTICS: $j = 0.2 \text{ mA/cm}^2$; $\theta_{\text{rms}} = 0.79^\circ$

Fig. 12

Fig. 13

Fig. 14

ION ENERGY: 51 keV; ION BEAM INTENSITY: 0^- : 3.5mA; Au $^+$: 1mA

Fig. 15

Fig. 16