A high-efficiency positive (negative) surface ionization source for RIB generation

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Abstract

A versatile, new concept, spherical-geometry, positive (negative) surface-ionization source has been designed and fabricated which will have the capability of generating both positive- and negative-ion beams without mechanical changes to the source. The source utilizes a highly permeable, high-work-function Ir ionizer ($\phi = 5.29$ eV) for ionizing highly electropositive atoms/molecules; while for negative-surface ionization, the work function is lowered to $\phi = 1.43$ eV by continually feeding cesium vapor through the ionizer matrix. The use of this technique for negative ion beam generation has the potential of overcoming the chronic poisoning effects experienced with LaB$_6$ while enhancing considerably the efficiency for negative surface ionization of atomic and molecules with intermediate electron affinities. The flexibility of operation in either mode makes it especially attractive for RIB applications and, therefore, the source will be used as a complementary replacement for the high-temperature electron impact ionization sources presently in use at the HRIBF. The design features and operational principles of the source will be described in this report.

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1.0 Introduction

Ion sources based on the surface-ionization principle are generally characterized by a high degree of ion beam purity, low energy spread, and limited range of species capability. These sources can be categorized according to the means by which the atomic or molecular vapor is fed onto the ionizing surface and the method used to extract the ions. Ionization can be effected by impinging vapor onto a hot metal surface or allowing the vapor to diffuse through a porous matrix. Surface ionization efficiency can be very high or low, depending on the electrochemical character of the species in relation to the work function of the ionizing surface. The energy spread of the ion beam is characteristically very low and is of the order of thermal energies $\sim 2kT$ ($\ll 1$ eV).

For efficient positive ionization, the work function of the metal should be high. Positive surface ionization is utilized for a number of applications, including: sputter-type negative-ion sources, the generation of electropositive ion beams for sputtering and effecting low-work-function surfaces, for enhancing negative-ion yields for Secondary Ion Mass Spectrometry (SIMS) applications, for microfocused ion beam applications, and for space propulsion applications. Several examples of this type of source are described in Ref. [1]. The surface-ionization process is also highly chemically selective and, therefore, can be used to great advantage for RIB applications where isobaric contamination problems may be important. Therefore, the surface ionization source has also been utilized to generate low-energy positive RIB beams at ISOL facilities (see for example, Refs. 2-4).

For efficient negative ion beam generation the work function of the surface must be low. Negative surface ionization has not been utilized frequently as a means for practical production of ion beams – principally due to the lack of chemically stable low-work-function materials. In contrast, several metals can be chosen for use as positive surface ionizers. LaB$_6$ is the most frequently used low-work-function surface ionizer, having a work function $\phi \equiv 2.7$ eV for polycrystalline and $\phi \equiv 2.36$ eV for single-crystalline material. A few sources based on the use of LaB$_6$ ionizer have been described in the literature (see for example, Refs. 5-7). Unfortunately, LaB$_6$ is easily poisoned and stable operation with the material is difficult [6]. Despite this problem,
LaB$_6$ has also been used at ISOL facilities for negative-ion generation of high-electron-affinity radioactive species with some success (see for example, Refs. 2 and 7). In principle, the negative surface complement of the ionization source described in this report, overcomes the handicap of being easily poisoned because of the continuously renewable supply of the work-function-lowering agent Cs. However, the concept must be evaluated before a final assessment may be made as to its viability for negative ion formation.

2.0 Theory of positive ion formation

Whenever an atom or molecule is near to or adsorbed on a hot metal surface, the valence band is broadened. If the level is broadened to the extent that it overlaps the Fermi level of the metal, electrons can move from the atom or molecule to the metal or from the metal to the atom or molecule, depending on the electropositive/electronegative character of the atom or molecule in relation to the work function of the metal surface. Thus, atoms or molecules may be emitted from the surface as neutral atoms or molecules or in ionic form. The process whereby ions are formed is referred to as surface ionization and obeys the appropriate positive or negative form of the Langmuir-Saha equation. Therefore, the technique can be used effectively to produce either positive- or negative-ion beams from certain members of the periodic chart.

For thermodynamic equilibrium processes, the ratio of ions to neutrals that leave an ideal surface can be predicted from Langmuir-Saha surface ionization theory. For atoms or molecules leaving a hot surface at temperature $T$ and work function $\phi$, the probability of positive ion formation $P_i$ is given by

$$P_i = \frac{\omega_+ (1-r_+) \exp \left( \frac{\phi - l_p}{kT} \right)}{\omega_0 (1-r_0)} \times \left[ 1 + \frac{\omega_+ (1-r_+)}{\omega_0 (1-r_0)} \exp \left( \frac{\phi - l_p}{kT} \right) \right]^{-1}$$

where $r_+$ and $r_0$ are the reflection coefficients of the positive and neutral particles at the surface, $\omega_+$ and $\omega_0$ are statistical weighting factors, and $k$ is Boltzmann’s constant. $\omega_+$ and $\omega_0$ are related to the total spin of the respective species given by
where \( s_i \) is the spin on the \( i \)th electron. Optimum ionization efficiencies are obtained for high-work-function materials and low-ionization-potential atomic species. For elements for which \( I_p > \phi \), the process is much less efficient. For example, the work function for clean tungsten is about 4.6 eV and the ionization potential for indium is 5.8 eV. Thus, in this case, the exponential term \((\phi - I_p)\) in the Langmuir-Saha relation is negative and, therefore, the probability of ionization is low. The efficiency is relatively high for group IA elements, but low \((10^{-2} \text{ or } 10^{-3})\) for elements such as In, Ca, Al, Ga, and Ti. The incident particles which are not ionized are evaporated as neutral atoms. Surface ionization sources can be highly element selective due to the fact that neighboring elements often have very different ionization potentials or electron affinities.

Equation 1 applies to an idealized situation in which the surface properties are isotropic and there are no surface contaminants. The work function of \( \phi \) varies with crystalline orientation, and the presence of minute amounts of certain contaminant materials on the surface can significantly alter the local work function.

### 3.0 Theory of negative surface ionization.

Analogously for negative-surface ionization, the probability \( \eta_i \) for arrival at a position far from the metal as a negative ion or neutral particle depends on the magnitude of the difference between the electron affinity \( E_A \) and the surface work function \( \phi \) of the atom or molecule, i.e., \((E_A - \phi)\). For thermodynamic equilibrium processes, the ratio of ions to neutrals which leave an ideal surface can be predicted from Langmuir-Saha surface ionization theory appropriate for negative-ion formation. The form of the Langmuir-Saha equation for the probability of negative-ion formation for neutral particles of electron affinity \( E_A \) interacting with a hot metal surface at temperature \( T \) and constant work function \( \phi \) is given by
where \( r_\perp \) and \( r_0 \) are the reflection coefficients of the particle at the surface and \( \omega_\perp \) and \( \omega_0 \) are statistical weighting factors for the negative ion and neutral atom, respectively. \( \omega_\perp \) and \( \omega_0 \) are related to the total spin of the respective species given by

\[
\omega = 2 \sum s_i + 1,
\]

where \( s_i \) is the spin on the \( i \)th electron.

From Eq. 2, it is evident that negative-ion yields could be enhanced by lowering the work function \( \phi \) or increasing the surface temperature \( T \) for elements where \( E_A \leq \phi \). The former can be effected by surface adsorption of minute amounts of low-work-function materials such as the group IIA and IIA elements. Analogously, the adsorption of minute amounts of highly electronegative atoms or molecules such as oxygen or the halogens can deleteriously affect the negative-surface ionization efficiency by raising the work function.

### 4.0 Electropositive adsorbate-induced work function changes

It is well known that atomic adsorption of a dissimilar element on a clean surface affects the surface work function. 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. These phenomena are exploited in SIMS as a means for enhancing both positive and negative secondary ion yields.

Semiempirical relations have been developed which relate the work function change \( \Delta \phi \) to the surface coverages [8]. These relations can be used to predict, with good accuracy, the value of the work function \( \phi \) over the complete range of adsorbate coverage (\( \sigma = 0 \) to \( \sigma = 1 \)). The equation which expresses the functional dependence of \( f \) can be written as follows:
where $\phi_0$ is the intrinsic work function of the sample and $\Delta \phi_m$ is the maximum change in surface work function induced by the adsorbate at optimum coverage $\sigma_m$. The approximate relation for computing maximum work function changes $\Delta \phi$ that occur at optimum fractional coverages $\sigma_m$ is given by

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

where $\phi_0$ is the intrinsic work function of the surface before the adsorbate is added and $I_A$ and $E_A$ are, respectively, the first ionization potential and electron affinity of the adsorbate material. The empirical relation is found to reproduce, with good accuracy, maximum work function changes for alkali metal and alkaline earth metal adsorption. From this expression, we note that for maximum changes in work function $\Delta \phi_m$, the term $1/2 (I_A + E_A)$ should be small. Thus, the choice of adsorbate material is extremely important. Of the stable elements available, Cs is the most effective element in the periodic chart for this application. The expression is valid for single crystalline as well as polycrystalline materials and also reproduces maximum changes due to group IIA adsorbates as well as other adsorbate materials. More details on the model and the accuracy of Eq. 4 for predicting maximum work functions for a number of adsorbate/adsorbent combinations are given in Ref. 8.

5.0 Ion Source design

The design features of the positive (negative) surface ionization source now under evaluation for potential use at the HRIBF [9] are schematically illustrated in Fig. 1. The source is designed to be used as a complementary replacement of the electron impact ionization sources presently in use at the HRIBF [10,11] for pending RIB generation
and therefore, is designed to mount in the same vacuum envelope. The principles of operation and design details of these sources are described below.

For RIB generation, collimated \(^1\text{H}, ^2\text{D}, ^3\text{He}, \) and \(^4\text{He}\) ion beams from the Oak Ridge Isochronous Cyclotron (ORIC) will pass through a thin window in the target material reservoir where they will interact with the refractory target materials chosen for the production of the desired radioactive beam. The species of interest are diffused from the target material which is maintained at high temperature during the production process. The target material reservoir is positioned within the inner diameter of a series-connected, resistively heated, triaxial Ta tube. The reservoir can be heated to temperatures exceeding 2000°C by passing a current through the tubular structure. The power required to heat the assembly to 2000°C is \(\sim 2.25\) kW (4.5 V at 500 A). The Ta target material reservoir is coated with Re metal as are beam transport tubing and internal surfaces of the source to reduce the residence times of chemically active elements during adsorption. Radioactive species diffused from the high temperature target will be transported through the resistively heated vapor transport tube and through the highly permeable Ir ionizer. Ionization occurs as the particles are evaporated from the spherical-geometry surface. The ionizer and vapor transport tube are heated resistively to \(\sim 1100°C\), the critical temperature required for evaporation of the ion species. The power required to achieve the critical temperature will be 0.35 kW. The total power required to heat the target and ionizer to the temperatures given above will be of the order of 2.6 kW. The ionizer is made of a highly permeable, carbon-bonded-fiber (CBCF) matrix coated with \(\sim 10\) μm of Ir. The conversion to the positive (negative) mode can be made by not applying (applying) power to the cesium reservoir; ion extraction, in the respective mode, is effected by applying \(-5\) kV(+5 kV) potential to the extraction electrode, depending on the species of interest. The ion extraction electrode system for the source was iteratively designed by use of the computer code described in Ref. 12, which includes space charge. A display of the positive (negative) ion optics of the source are shown in Fig. 2. The spherical-geometry extraction electrode system is designed to focus the particular ion beam through the ion exit aperture as shown, the beam is then further accelerated up to energies as high as 50 keV. The post accelerator gap can be varied in order to
optimally accelerate ion beams over a wide energy range as required for beam transport through the first stage isotope separator system of the HRIBF [9].

6.0 Predicted capabilities of the positive (negative) surface ionization source

Either positive- or negative-ion beams resulting from interactions between highly electropositive or electronegative atoms or molecules and a spherical-sector surface ionizer maintained at \(-1100\)°C can be formed with the source displayed in Fig. 1. The dual-surface-ionization capability greatly extends the number of species for which the source can be utilized. The ionizer is made of highly permeable Ir which has an intrinsically high work function \((\phi = 5.29 \text{ eV})\) and, is, therefore, very efficient for positive surface ionization of the group IA members of the periodic chart (Li, Na, K, Rb, Cs, and Fr) and group IIA elements at lower efficiencies. When operated in the negative-surface-ionization mode, the work function is lowered to a value of approximately \(\phi \approx 1.43 \text{ eV}\) by feeding Cs vapor through the permeable matrix at a rate sufficiently fast so that \(-0.5\) monolayer of Cs is absorbed onto the Ir surface. In principle, this technique can be utilized for the efficient generation of negative-ion beams from high-electron-affinity elements such as members of the group VIIA elements (F, Cl, Br, I, At) and, perhaps, other members of the periodic chart, either in atomic or molecular form. Several molecules have remarkably high electron affinities that should easily be ionized by the low-work-function characteristics of the Ir/Cs surface. Molecules such as CN, BO, CS, SO, and PO, as well, should all ionize with relatively high probabilities if the work function of the surface can be maintained at a low value. The efficiencies \(P_i\) for positive (negative) ion formation, as calculated from Langmuir-Saha theory, are shown in Fig. 3. The negative surface ionization efficiencies are calculated under the assumption that the work function is always maintained at a constant value of \(\phi \approx 1.43 \text{ eV}\).

7.0 Conclusions

The prototype source displayed in Fig. 1 is presently under evaluation for operation in both the positive and negative modes, and therefore, the results of these tests will be reported at a later date. While we are confident that the positive-surface-ionization
mode will perform up to expectation, it is premature to speculate on the stability of the negative-surface-ionization mode of operation. At this time, we do not know whether or not the flow rate of Cs, required in order to efficiently ionize highly electronegative species, can be maintained without compromising the stability of operation. That is, too much Cs in the extraction region of the source may cause sparking. Also, chemical reactions between the Cs metal and highly electronegative species may adversely affect the stability of operation and ionization efficiency of the surface ionization process. The source described in this article, probably will require further development. However, we are cautiously optimistic that the technique will work based on our extensive experience when applied to negative-sputter-type ion sources which operate very stably for extended periods of time. The successful development of a dual-purpose, positive (negative) ion source surface-ionization source will find many applications, including their usage at ISOL and RIB facilities. The technique for negative-ion generation also offers the prospect of a solution to the chronic poisoning problem associated with the use of LaB$_6$ ionizers.
References

Figure Captions

1. ORNL-DWG 95-10131. Schematic drawing of the self-extraction, spherical-geometry, positive- or negative-surface-ionization source equipped with a highly permeable Ir ionizer.

2. ORNL-DWG 95M-8398. Ion optics of the spherical-geometry positive (negative) surface-ionization source in Fig. 1 as calculated by use of the computer code described in Ref. 12.

3. ORNL-DWG 95M-8085B. Theoretical probability for positive surface ionization from a clean Ir surface for selected highly electropositive atoms and for negative surface ionization from a clean Ir surface with ~0.5 monolayer of Cs on the surface to create a low work function surface as required for negative surface ionization of highly electro-negative ion species.
1 Mesh Unit = 0.1 mm

Positive/Negative Ion Beams

0 kV

$0 = \pm 20 \text{ kV}$

$0 = \pm 18 \text{ kV}$

$R$ (mesh units)

$Z$ (mesh units)
Electron Affinity, $E_A$(eV)

1.0 1.5 2.0 2.5 3.0 3.5 4.0

Probability for ionization, $P_i$(%)

Ionizer
- Ir: $\phi = 5.27$ eV
- Ir + Cs: $\phi = 1.43$ eV

Surface Temperature: $T = 1473 \, ^\circ$K

Fig. 3