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The temperature dependence of the energy distribution of the beam emitted by an $Au_{82}Si_{18}$ liquid metal field-ion emitter

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Abstract

The energy distribution of the monomer ion species is studied as a function of emitter temperature for an $Au_{82}Si_{18}$ eutectic liquid metal alloy ion source (LMAIS). Secondary peaks, or shoulders, in the distribution of some of the species are explained in terms of Hornsey's model, which involves the fast movement of the jet in conjunction with a space–charge energy broadening mechanism within the beam. Moreover, the results confirm an earlier conclusion that whereas Au^+, Si^+ and Si^{++} are emitted by direct field-evaporation from the liquid surface, Au^{++} forms by the post ionization of Au^+.
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1. Introduction

Many aspects of liquid metal ion sources (LMISs) have been studied in the last 30 years; yet, few measurements exist on the temperature dependence of their characteristics and even fewer on the behaviour of their beam energy distribution with temperature. This work is concerned with the latter property of an $Au_{82}Si_{18}$ liquid metal alloy ion source. The melting point of the $Au_{82}Si_{18}$ eutectic is 365 °C, but the source was operated at elevated temperatures for better stability. In fact, LMISs are almost always more stable at temperatures above the melting point of the metal, so that a study such as this is of considerable use. Needless to comment on the importance of the energy spread of the beam, which, through the chromatic aberrations of the system, determines the final spot size at the target.

Si beams are of importance for direct write applications, since they would not contaminate a Si target. Moreover, Si is emitted mainly in the form of Si^{++} and these ions require half the

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accelerating voltage to reach a certain energy compared to Si^+ ions.

Some metals are not suitable for LMIS manufacture. In such cases a suitable alloy is produced and the desired species is mass-separated from the beam, e.g. by an $E \times B$ filter, as in the present case. Si is such an element for it reacts chemically with the liquid cone–anchoring needle substrate and it dissolves it. Thus, the need for an alloy, normally of the Au–Si type. The results to be discussed below were all taken at an emission current of 5 μA. The energy distributions were obtained using a retarding field energy analyzer.

2. Experimental results and discussion

Fig. 1 shows the energy distribution of Si^+ for 841, 1054 and 1267 °C. A shoulder can be seen on the slow side of the distribution. This, however, was expected ever since Swanson [1] first observed it with an $Au_{90}Si_{10}$ alloy LMIS under melting point (660 °C) operating conditions. At high temperatures the shoulder tends to merge with the main peak (Fig. 1(c)), just as it does with increasing current [2]. The distribution for Au^+, apart from being asymmetric, shows a slight distortion (Fig. 2) which persists at higher temperatures, becoming somewhat more pronounced. The asymmetry arises from charge transfer collisions between Au^+ ions and the neutral atom cloud that exists in front of the tip; this process results in slow ions and fast atoms [3]. The existence of atoms near the tip is known from the luminous spot that always accompanies emission, the result of excitation of atoms by ion impact [3,4].

The slight distortion in the distribution seen in the case of Au^+ becomes clearly visible as a secondary peak in the case of Au^{++}, as the source temperature is raised (Fig. 3(b)). At higher temperatures the two peaks tend to merge, so that the energy distribution displays a shoulder on the low energy side.

Finally, the distribution of Si^{++}, like that of Au^{++} at low temperatures, displays a near Gaussian shape (Fig. 4). The symmetric nature of Au^{++} at sufficiently low temperatures and of Si^{++} is rela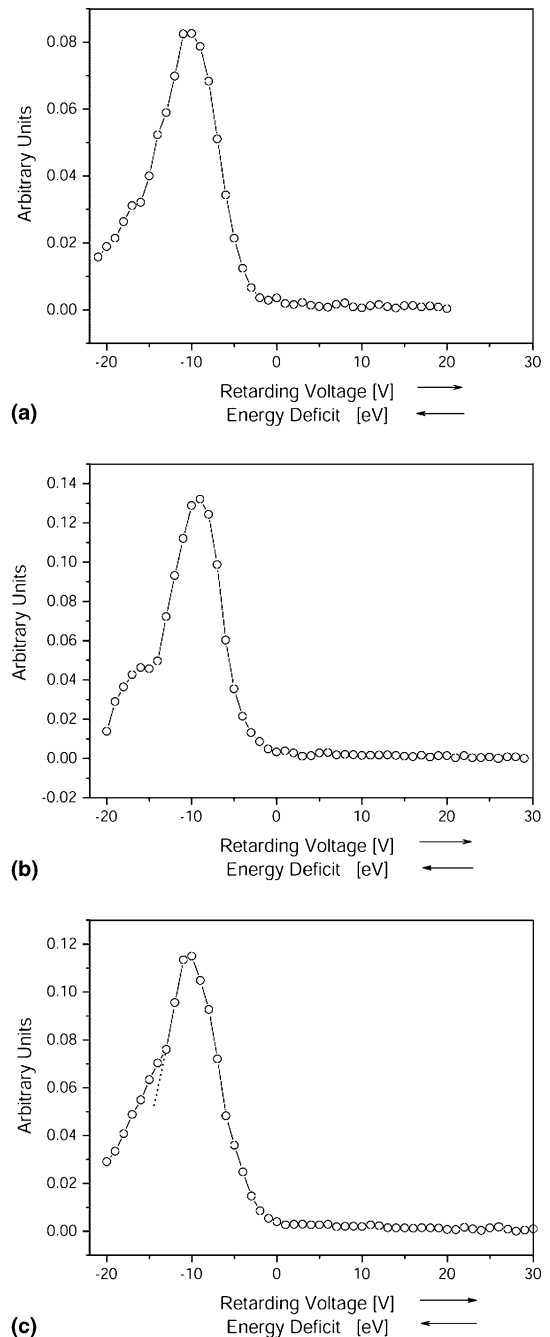ted to the charge transfer collisions mentioned earlier, or the lack of them in the case of doubly charged ions. The atomic cloud in front of the tip is much more transparent to doubly
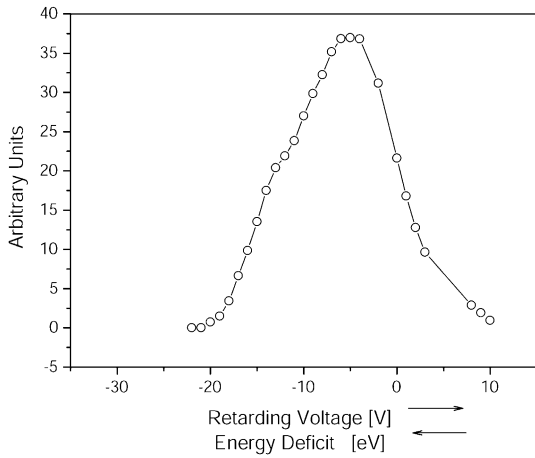

Fig. 1. Energy distribution of Si^+ ions for different emitter temperatures: (a) 841 °C, (b) 1054 °C, (c) 1267 °C.

Fig. 2. Energy distribution of Au$^+$ for $T = 841$ °C.

charged ions than it is for singly charged ones. We shall return to Figs. 1–4 later. For the moment let us concentrate on the (main) peak energy deficit (PED) of the distributions.
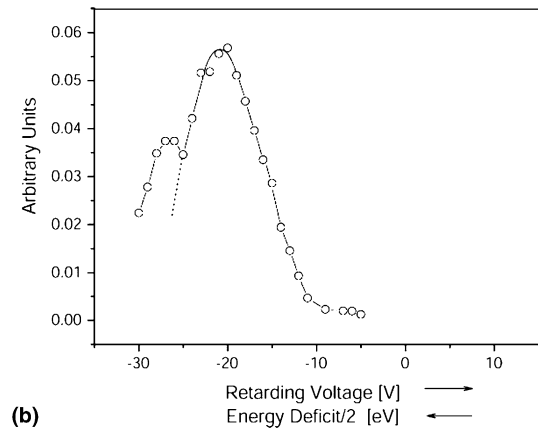
The peak energy deficit of the distribution represents the work expended to turn a bound atom into a free ion in the presence of the electric field. In other words, it represents the work expended by the power supply.

It is seen from Fig. 3 that the energy deficit of the main peak does not change with temperature, and the same was found for the other three ionic species. The values deduced from Figs. 1–4 are ∼5, ∼40, ∼10 and ∼20 eV, for Au$^+$, Au^{++}, Si$^+$ and Si^{++}, respectively. However, it is well known that the peak deficit shifts towards lower values with increasing current [5] and this is particularly true for singly charged ions, like e.g. Au$^+$. The shift is related to the asymmetry of the curves, discussed earlier, coupled to the energy broadening mechanism; the latter arises from coulomb interactions within the beam. A full discussion on the matter is given in [6]. Thus, to obtain a true value of the PED one has to measure it at near source extinction currents of the order of 1 μA. We have, in fact, done so, and the results are presented in Table 1, together with theoretical values calculated from [7].
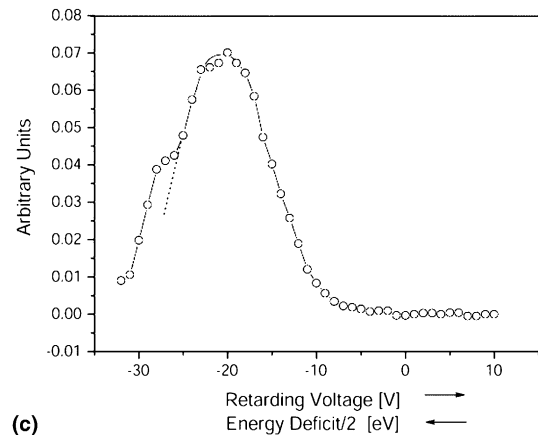
$$\Delta E(n) = \Lambda + \sum_n I_n - n\varphi_{\mathrm{C}} - Q(n). \qquad (1)$$
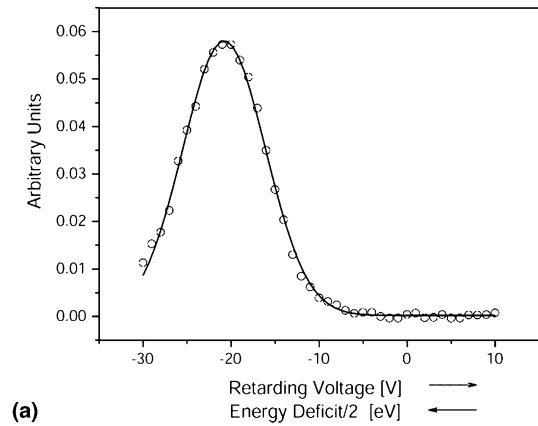
Fig. 3. Energy distribution of Au^{++}: (a) 947 °C, solid curve: Gaussian fit; (b) 1160 °C, (c) 1267 °C.

Here $\Delta E(n)$ is the peak energy deficit, or the primary peak energy deficit; I_n is the nth ionization
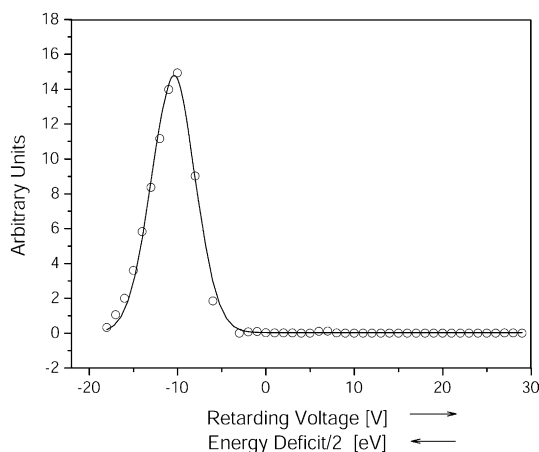
Fig. 4. Energy distribution of Si^{++} for $T = 1054$ °C; solid curve: Gaussian fit.

Table 1
Comparison of experiment with theory for the (primary) PED for Si and Au ions, $\Delta E(n)$, $n = 1, 2$

	Exp. $\Delta E(1)$	Exp. $\Delta E(2)$	Theor. $\Delta E(1)$	Theor. $\Delta E(2)$
Si	8.9	19.6	8.1	19.4
Au	7.4	38.7	7.9	22.9

Values of Λ, I_1, I_2, ϕ_C(Ni) from [8].

potential of the atom, subsequently ion, if the atom is n-fold ionized; φ_C is the work of the retarding electrode; $Q(n)$ is the field-reduced potential barrier seen by a field-evaporated ion. Q is normally taken as ~ 0 [1].

It can be seen that the agreement between experiment and theory is good, in the case of Si^+, Si^{++} and Au^+, strongly indicating that these ions form by field evaporation. In the case of Au^{++}, there is total disagreement, by a factor of almost 2. The disagreement becomes understandable, however, for, as shown recently, Au^{++} does not form by direct field-evaporation from the liquid surface, but by the post-ionization of Au^+ [2]. Post-ionization is the process whereby an ion sheds one or more electrons, as it moves away from the emitter, that tunnel back to the emitter [9]. From potential energy diagrams [10], it is clear that the point of post-ionization is at a distance of a few atomic radii from the point where direct field-evaporation into the doubly charged state takes, or would take place.

Now, the field acting at the tip of our $Au_{82}Si_{18}$ liquid emitter has been estimated at ~ 3.3 V/Å [2]. Thus a potential difference of ~ 10 V must exist between the said points. The former point corresponds to the point of intersection of the potential energy curves of the singly and doubly charged ions; the latter point corresponds to the point where the curve for the doubly charged ion intersects the atomic curve. Translated into energy, for a doubly charged ion, a ~ 10 V potential difference corresponds to ~ 20 eV; i.e. the difference between theory and experiment for the PED of Au^{++}. [1]

Let us now turn our attention back to Figs. 1–4. The most natural explanation that comes to mind for the secondary hump in the Si^+ (Fig. 1) distribution is a field-ionization mechanism of free atoms at some distance from the emitter. However, there are serious difficulties with this explanation due to insufficient heat input to the emitter to account for the necessary atom flux that needs to be evaporated [10]. Another – more plausible – possibility is that the required number of atoms comes from the break-up of large ionic complexes or droplets. However, a field-ionization mechanism, regardless of where the atoms come from, is a highly improbable proposition in the case of Au^{++} (Fig. 3). Free space field-ionization directly into the doubly charged state simply does not happen. The emergence of secondary peaks or shoulders with increasing temperature was first reported by Swanson with Ga^+ [11] and subsequently by Hornsey with both Ga^+ and In^+ [12]. The latter author also put forward a most original hypothesis for the doubly peaked nature of the energy distribution at elevated temperatures, based on the fast movement of the liquid jet [13].

In putting together his model Hornsey was inspired by a similar peak splitting effect observed in radio frequency plasmas. In this case ions arriving at an oscillating sheath are then accelerated onto a target, so that the potential through which an ion

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[1] Swanson finds 16.5 ± 2 eV difference for the PED for Ga^{++}, also believed to form by post-ionization, in this case of Ga^+ [1]. Swanson's results refer to voltage, rather than energy, distribution measurements. Thus, for doubly charged ions all his results have to be multiplied by a factor of 2.

falls depends on the phase of the oscillation at the time of arrival. Assuming that instabilities of the liquid cone–jet system depend on temperature in roughly the same way they depend on current, Hornsey argued that the low energy peaks can arise from the convolution of an energy spectrum that includes space–charge energy broadening. Computer simulations based on assumed values of certain unknown parameters, produced the high temperature energy distribution of LMISs remarkably well. The same program also produced the observed shape and behaviour with increasing current of the low temperature, singly peaked distributions. Hornsey's model requires jet vibration frequencies in the GHz range. Such frequencies are difficult to detect experimentally; in fact attempts to do so have failed (S.P. Thomson, private communication to GLR Mair).

Now, Fig. 5 shows the energy spread (FWHM), $\Delta E_{1/2}$, as a function of emitter temperature (T), for Si^+, Si^{++}, Au^+ and Au^{++}. For Si^+ and Au^{++} at high temperatures, the energy spread of the distribution corresponding to the primary peak is only considered. Our results agree well with those of Swanson's $Au_{90}Si_{10}$ source at low temperatures, the agreement in the case of Si^{++} being almost perfect. Disagreement exists in the case of Si^+, where Swanson's energy spread value for 5 μA is considerably larger than ours. This is understandable, however, since Swanson measured the spread of the distorted distribution, whereas Fig. 5 shows the spread corresponding to primary peak only.

3. Conclusions

In conclusion, the energy spread and PED for the primary peak does not appear to change much with temperature and this is understandable since the energy spread is determined mainly by coulomb interactions within the beam. The doubly peaked nature of the distribution is most naturally explained by the fast movement of the jet, in conjunction with a coulomb energy broadening mechanism. Au^{++} forms by the post-ionization of Au^+, as opposed to Si, Si^+ and Si^{++}, which form by direct field-evaporation.

Fig. 5. Energy spread (primary peak) versus T for Si^+, Si^{++}, Au^+ and Au^{++} ions.

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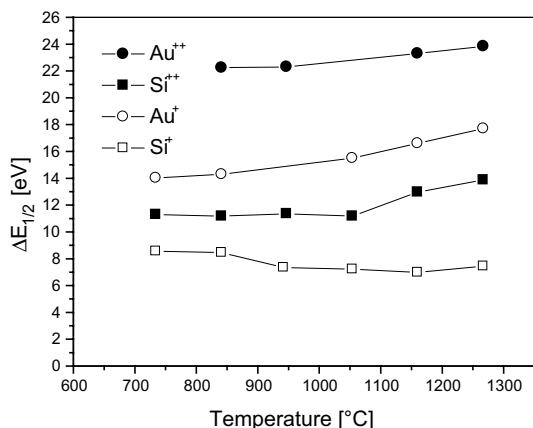[13] R.I. Hornsey, J. de Phys. 50 (1989) C8/197.