1st Draft

CURRENT-DEPENDENT ENERGY SPECTRA OF CLUSTER IONS FROM A LIQUID GOLD ION SOURCE.

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Abstract:-

The energy spectra of ions emitted from a liquid-gold point emitter have been measured using a time-of-flight technique. The energy distributions are found to depend strongly on the total current drawn from the ion source. The Au^+ distribution is narrow (~50 eV at 20μA) but broadens and develops a pronounced low-energy tail at high currents and at high source temperatures. Au_2^+, Au_3^+, and Au_4^+ cluster ions have a double-peaked energy distribution at intermediate currents: at low currents the high-energy peak is small or absent, but becomes dominant as the current is increased. The Au^{++} distribution is narrow at all currents.

Introduction.

Liquid metal point ion sources have considerable potential use for semiconductor microfabrication and for ion microprobes because of their high brightness and very small source area. In these ion sources an electrostatic field is used to draw the meniscus of a liquid metal into a sharp cone (Krohn 1961: Taylor and McEwan 1965) and ionization occurs in the intense field at the tip of this cone, giving ion currents in the range 10-100 μA from a source region of 1 μm or smaller diameter.

The energy distribution of the resulting ions is of practical importance, in that chromatic lens aberrations may limit the minimum size of the region into which the ions can be focussed (Seliger 1978). The energy distribution also gives information on the mechanism of ion formation, which is at present incompletely understood. Previous workers have obtained energy distributions using retarding-potential techniques and gallium or caesium sources, finding distributions with FWHM in the range 12-25 eV (Krohn and Ringo 1975, Mair and von Engel 1979, Swanson et al. 1979), while Sudraud and coworkers (1979) have measured energy distributions from a gold source, using a mass-spectrometer to separate the constituents of the ion beam & found a FWHM of 65eV for Au^+. Culbertson et al. (1980) have performed a similar analysis for a gallium ion source. The results obtained by Sudraud and coworkers are of particular interest in that the energy distributions of the cluster ions Au_n^+ (n = 2-7) were found to have two peaks, with the lower-energy peak becoming more pronounced with increasing n. In contrast, Culbertson and coworkers found only single peaks for Ga_2^+ and Ga_3^+ .

Sudraud FWHM =

In this letter results are presented which show that the energy distributions of the constituents of the emission from a gold ion source are strongly dependent on the total current drawn from the source: it is also shown that operation of the source above the minimum temperature needed to liquefy the gold also leads to a broadening of the Au^+ energy distribution.

Experimental.

The ion source used in the present work was provided by UKAEA Culham, and will be described in more detail elsewhere (Prewett et al. 1980): the gold cone was supported on a tungsten needle which was in contact with a reservoir of liquid gold. The mass-analysis and energy measurements were made using a time-of-flight technique. A narrow beam of ions was selected by an aperture in a collector facing the ion source. A pair of electrostatic deflector plates were used to sweep the beam across a small aperture, allowing a pulse of ions to enter a drift tube 850 mm long: the deflector plates were short ($<$1mm) so that the time of origin of ions entering the drift tube was well defined. Individual ions were detected at the end of the drift tube with a Bendix M306 electron multiplier. The flight times of the ions were measured using a purpose-built timer counter operating at 100 MHz. The apparatus will be described in more detail elsewhere.

The time of flight t of ions of mass m and charge ne is related to their energy E or accelerating voltage V by

$$E = neV = \tfrac{1}{2}m\,(L/t)^2 ,$$

where L is the drift-tube length. Differentiating, we obtain

$$\Delta E/E = -\,2\Delta t/t ,$$

where ΔE is the shift in energy which results in a given change in flight time Δt.

Results.

Histograms showing the number of ions with a given flight time are presented in figures 1 and 2, for Au^+ and Au_3^+ respectively, for a range of currents. For Au^+ it is seen that the long tail at low energies (corresponding here to long flight times) which Sudraud and coworkers (1979) observed,is in fact current-dependent. At 20 μA the Au^+ peak is narrow (FWHM 50 eV) and the low-energy tail is very small, while at 90 μA the Au^+ peak has broadened to 110 eV and there is a considerable tail extending to deficits of 1 keV or more. The energy resolution of the apparatus has not been measured, but is calculated to be in the range 20-30 eV: the true FWHM for Au^+ at 20 μA may be as low as 40 eV.

For Au_3^+ (figure 2) it is found that the distribution obtained at ~65μA corresponds closely to that observed by Sudraud et al (1979). However, at low currents the high-energy peak is very small or absent, while at higher currents a sharp high-energy peak develops and finally dominates the low-energy peak. At high currents the distributions show sharp high-energy edges, but somewhat stepped low-energy edges, suggesting the existence of barely-resolved fine structure, with a spacing between steps of some 40 eV.

The relative amplitudes of the low-energy and high-energy peaks of Au_2^+ and Au_4^+ show similar changes with current: for Au_2^+ the peaks are of equal amplitude at ~30 μA, while for Au_4^+ the high-energy peak begins to develop at 60 μA and equals the amplitude of the lower-energy peak at 80 μA.

Au^{++} and Au_3^{++} energy spectra have been measured as a function of current. They are found to resemble closely those published by Sudraud and coworkers (1979) with only a small increase in peak width and tail length at high currents.

The half-width of the main Au^+ peak has been plotted as a function of total source current in figure 3, for two different source temperatures. The lower line shows the half-widths measured at the minimum heater current required to maintain the gold as a liquid, while the higher line shows the half-width measured with an increased heater current , with an estimated rise in reservoir temperature of 110 K. It is seen that the FWHM rises as the current is increasd for both reservoir temperatures. .. Raising the source temperature increases the half-widths by some 25 20 eV at any given current.

Discussion.

The results presented here show that where chromatic aberrations of a subsequent lens system are of importance, the ion source should be operated at as low a current and as low a temperature as is consistent with stable operation. The ion source used in the present work will operate stably at 20μA, and for short periods at 5 μA. Similar low current operation has been reported by Wagner and Hall, and it seems that Sudraud's source ($i_{minimum}$ = 100 μA) may be atypical in this respect.

The origins of the details of the energy spectra for gold ions have been discussed by Sudraud et al (1979) and by Dixon (1979). Dixon concluded that charge exchange collisions in the dense vapour surrounding the hot liquid emitter could account for the differences between the Au^{++} and Au^{+} energy distributions, since the cross-section for the collision

$$\underset{fast}{Au^{+}} + \underset{slow}{Au} \rightarrow \underset{fast}{Au} + \underset{slow}{Au^{+}}$$

is much higher than that for the collision

$$\underset{fast}{Au^{++}} + \underset{slow}{Au} \rightarrow \underset{fast}{Au} + \underset{slow}{Au^{++}}$$

The results presented here would therefore imply that the density of neutral gold vapour surrounding the emitter must increase rapidly as the ion current increases. This seems reasonable, as the surface temperature of the gold will be raised by electron impact and by other processes (Gomer 1979): against this, Mair and von Engel/(1979) found that a gallium source lost a considerable amount of gallium (∼65 %) as vapour even at low currents.

The behaviour of the double-peaked energy-distributions of the gold cluster ions may perhaps be explained by the model of Gomer(1979) who suggested that at low currents field-ionization of metal vapour should be the dominant source of ions: at higher currents the ionization of vapour by secondary electron impact may become important. Gomer further showed that a considerable surface area of the liquid gold cone must contribute vapour to the ionization region in order to account for the observed currents. On this basis the narrow Au_3^+ peak observed at 20 μA would result from field-ionization of Au_3 at some distance from the surface, the neutral clusters originating from vapour generated from the shank of the Taylor cone, and attracted to the ionization region by polarization forces. As the current is increased, the impact of secondary electrons would progressively heat the tip of the cone and enhance the direct supply of vapour from the tip of the cone to the ionization zone: this vapour would be ionized close to the surface, either by electron impact or by field-ionization, resulting in the narrow high-energy peak. Space-charge effects are likely to broaden the observed peaks considerably (Gomer 1979).

Acknowledgements.

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References.

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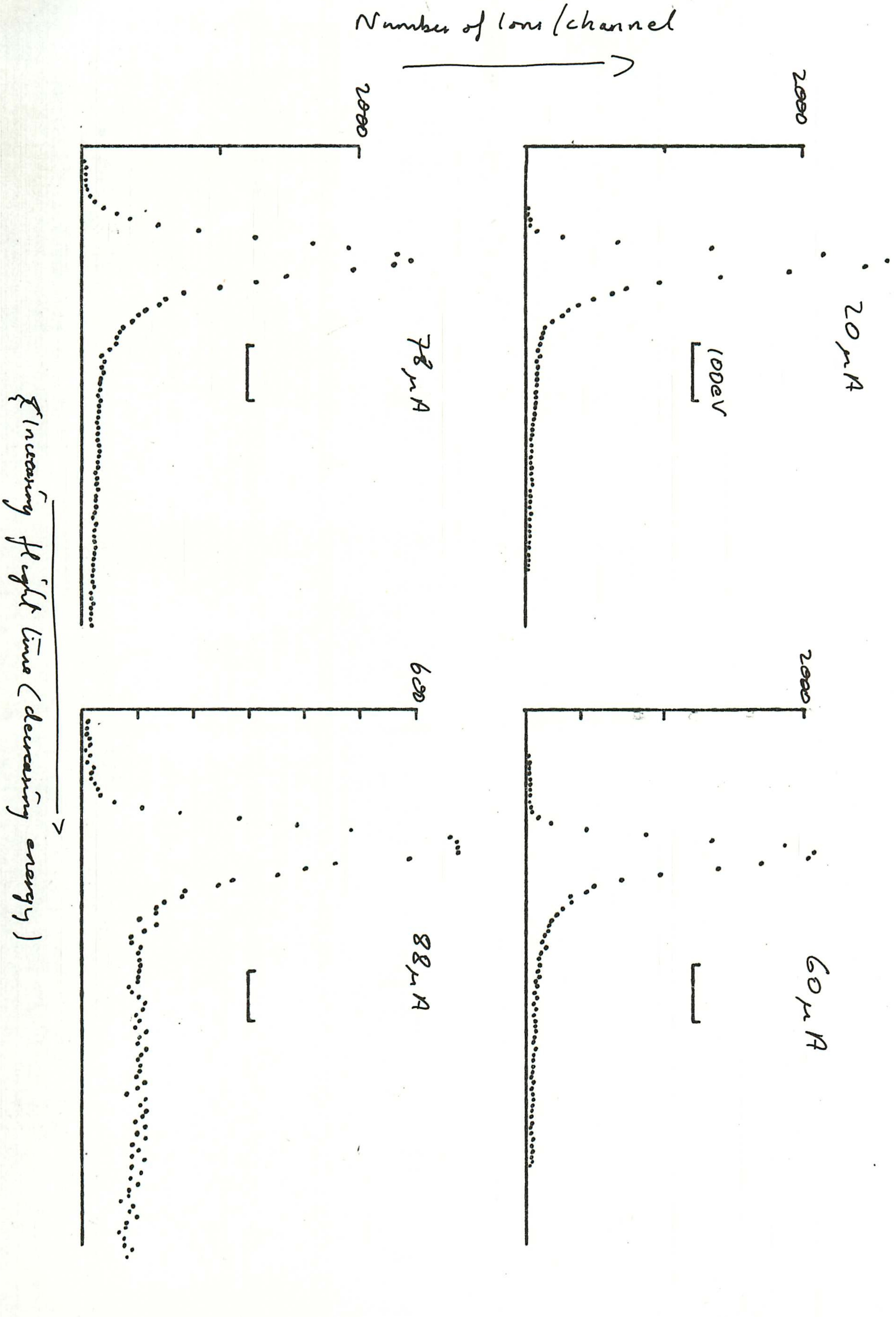
Wagner A and Hall T M

Fig 1.


Number of Ions/channel
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2000
2000
600
20 μA
100eV
78 μA
60 μA
88 μA
Increasing flight time (decreasing energy)

Figure 2

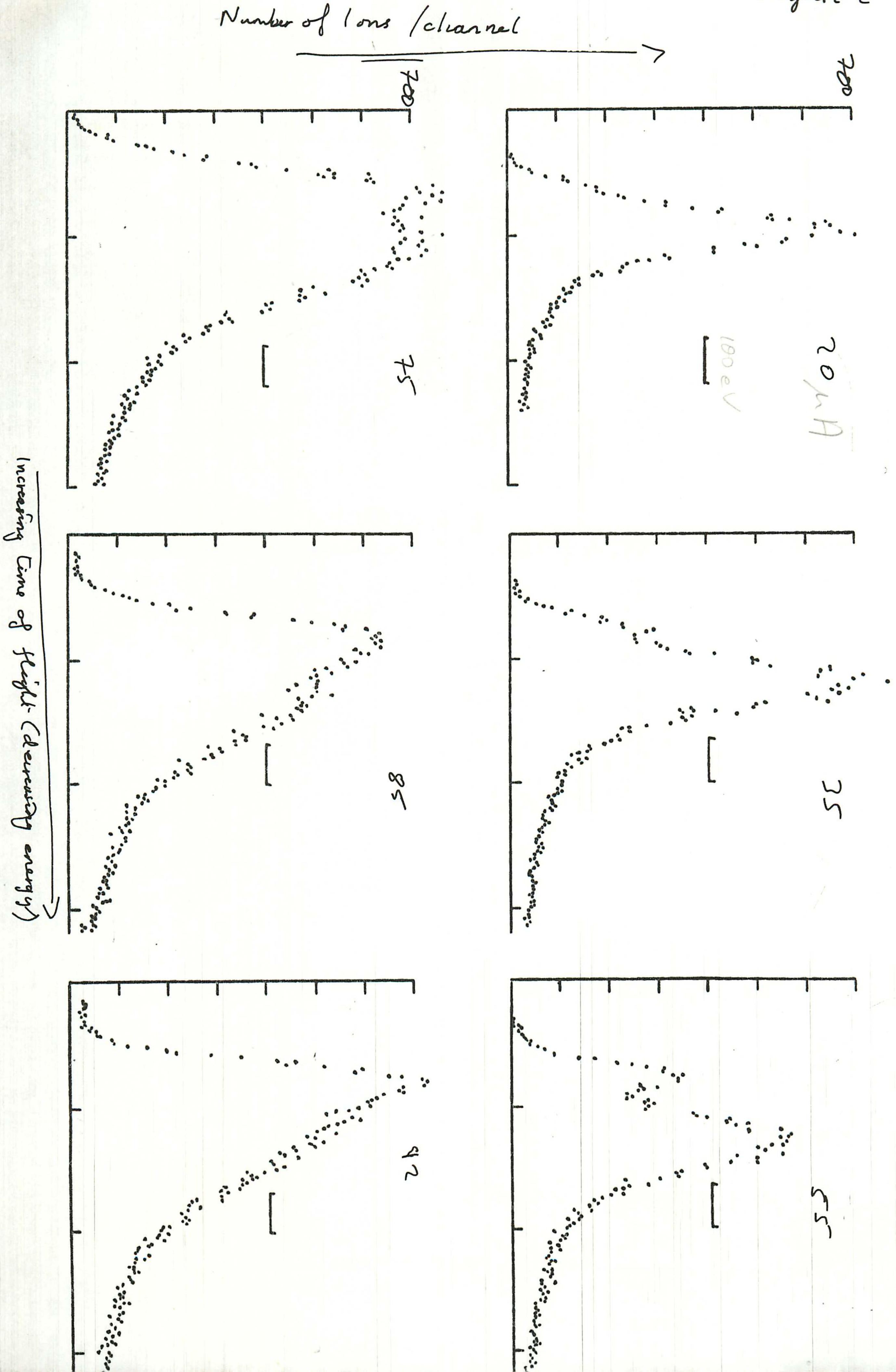

Number of Ions /channel
700
700
Increasing time of flight (decreasing energy)
20 μA
100 eV
35
55
75
85
42

Figure 3

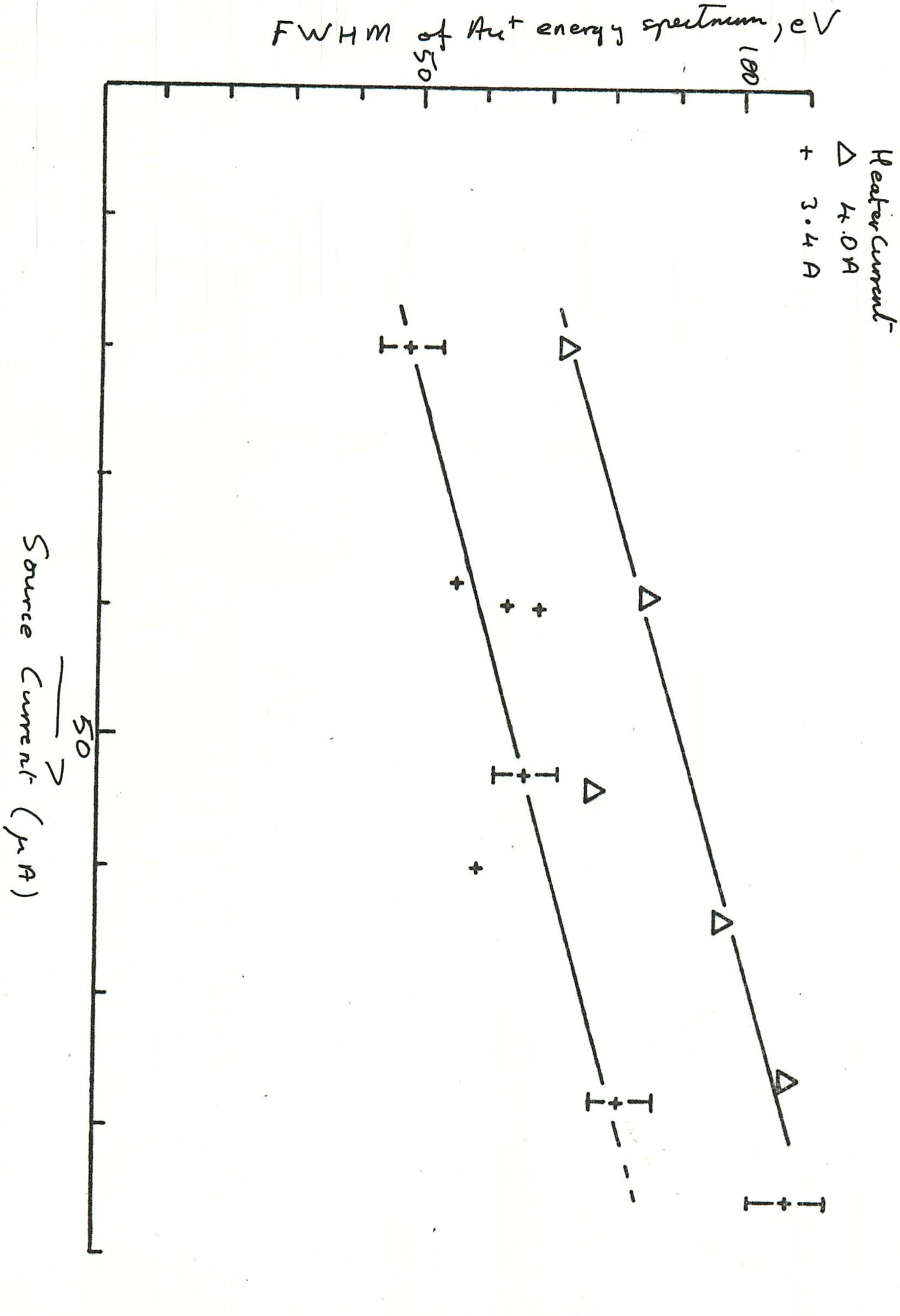
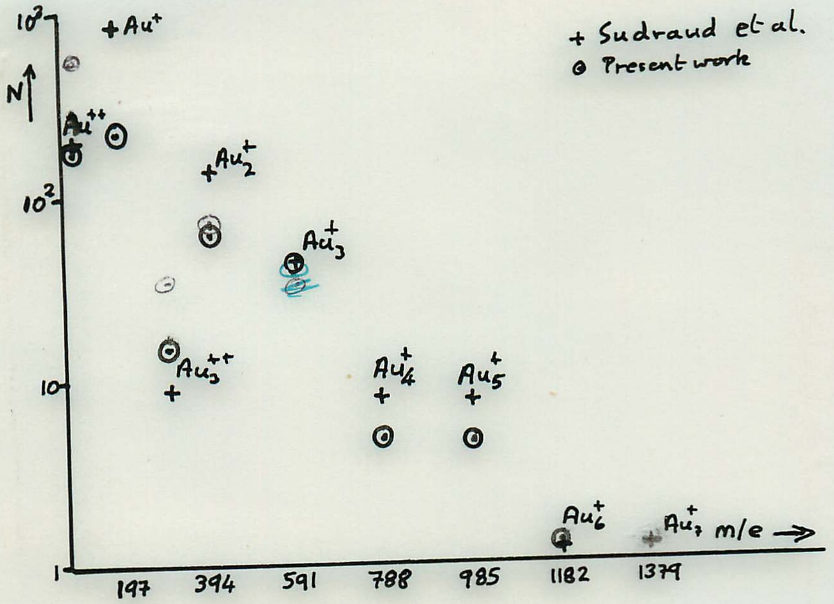

FWHM of Au$^+$ energy spectrum, eV
50
100
Heater Current
△ 4.0A
+ 3.4A
Source Current (μA)
50

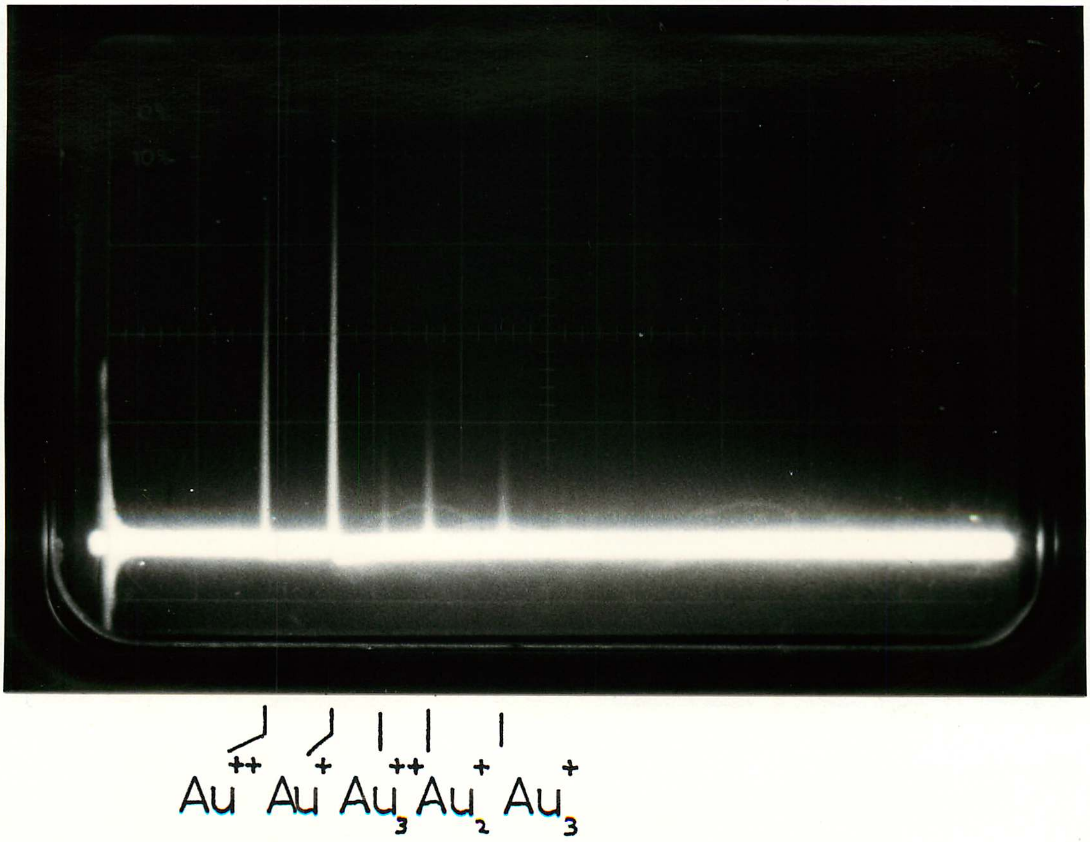
Figure captions

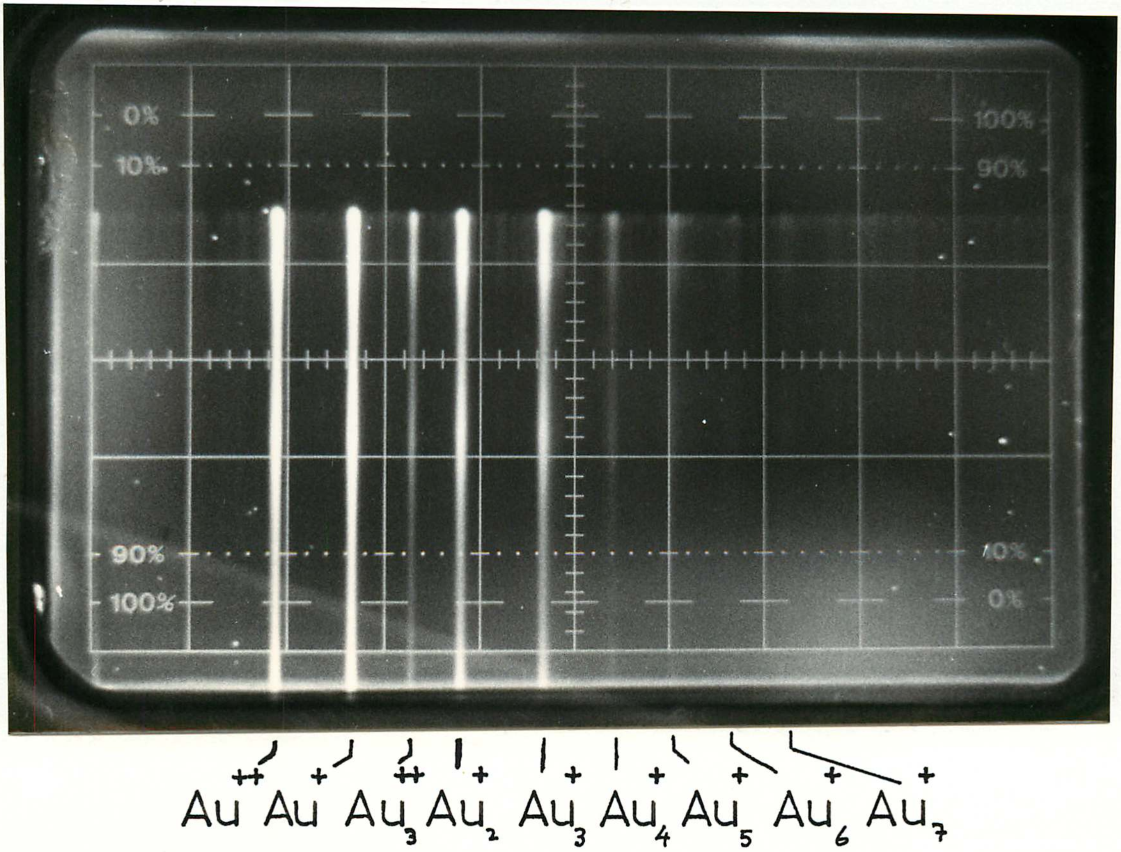
1) Au^+ time-of-flight spectra at different total source currents. ~~[illegible]~~ Arbitrary zero for horizontal axis. Scale bars ~~is~~ are 100eV.

2) Au_2^+ time of flight spectra at different source currents. Arbitrary zero for horizontal axis. Scale bars are 100eV

3) Full width at half-maximum (FWHM) for Au^+ energy spectrum, plotted as a function of source current for two gold reservoir temperatures. Lower line is obtained at minimum temperature to liquefy gold.


10³
N
10²
10
1
+ Au⁺
Au⁺⁺
Au₂⁺
Au₃⁺
Au₃⁺⁺
Au₄⁺
Au₅⁺
Au₆⁺
Au₇⁺
m/e
197
394
591
788
985
1182
1379
+ Sudraud et al.
⊙ Present work


Au^{++}
Au^{+}
Au_3^{++}
Au_2^{+}
Au_3^{+}


0%
10%
90%
100%
100%
90%
10%
0%
Au^{++}
Au^{+}
Au_3^{++}
Au_2^{+}
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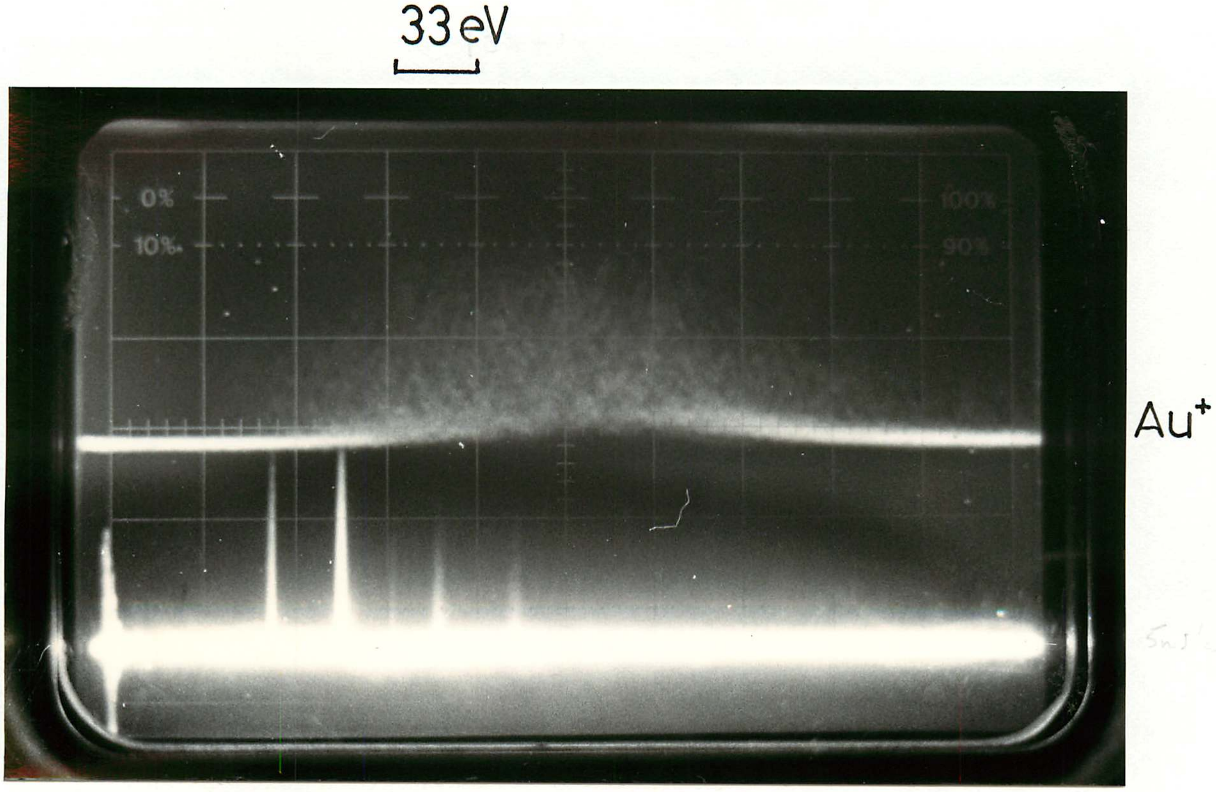
Au_4^{+}
Au_5^{+}
Au_6^{+}
Au_7^{+}



5 μS/cm / 50 nS/cm

Brian

Herewith a copy of the Culham report for last year, & a first draft of paper on gold energy spectra, to be submitted to Journal of ~~Applied~~ Physics D as a letter if Phil Prewett agrees.

Culham have so far supplied 1 Gallium & 1 gold ion source — the gallium source was anomalous in some ways & Prewett is bringing a new one & hopefully a Sn source as well to see if we can get more reproducible results (in particular for energy spreads): some of the anomalies may well result from the temperature effect shown in the paper for gold, which wasn't previously known.

Another area of interest, not mentioned in the enclosed paperwork, but which we have some data on (for fast neutrals see below), is the ~~net~~ neutral content of the ion beam (ie thermal slow neutrals (hard to detect, but probably a lot of them ~50%?, & fast charge-exchange neutrals $Au^+_{fast} + Au_{slow} \to \overset{\sim 100eV}{Au_{fast}} + Au^+_{slow}$ which should increase at higher currents).

More sophisticated energy analysis (retarding-grid or deflection-type analyser) would be useful, especially for multi-isotope sources (Ga, Sn), & I would guess that Culham would be pleased to get data in this area, as it directly affects the practical applications of their sources, as well as the theory of the source operating mechanism.

Bob