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Journal

Journal of applied physics, 66(11)

Authors

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Publication Date

2017-12-11



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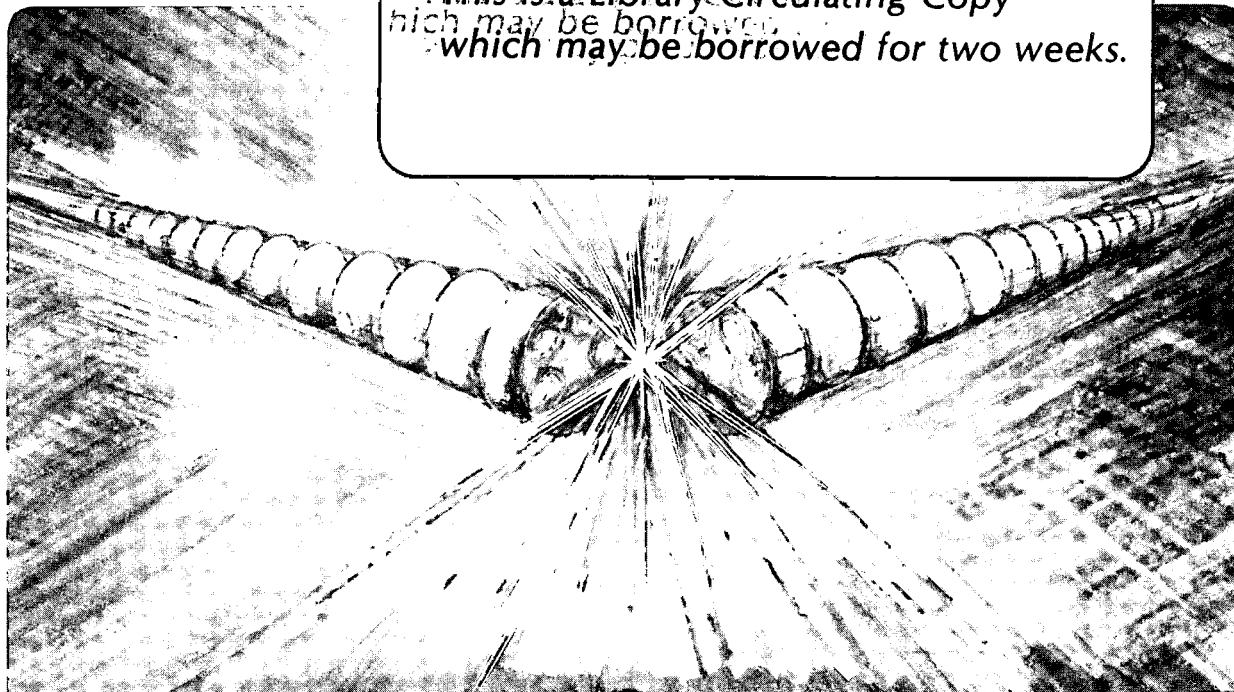
Submitted to Journal of Applied Physics

Ion Spectra of Vacuum Arc Plasma with Compound and Alloy Cathodes

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March 1989

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**ION SPECTRA OF VACUUM ARC PLASMA WITH
COMPOUND AND ALLOY CATHODES***

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* This work was supported by the U.S. Department of Energy under Contract No. DE-AC03-76SF00098, and by the Army Research Office and the Office of Naval Research under Contract No. N00014-88-F-0093.

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ABSTRACT

We have carried out an experimental investigation of the charge state distribution of ions produced in the vacuum arc plasma for the case when the cathode is a compound or alloy. The plasma was generated in a metal vapor vacuum arc ion source, and the charge state spectra were measured using a time-of-flight method. We have compared these spectra to the spectra obtained from cathodes of the constituent elements. The cathode materials used and reported on here were the following groups: SiC/Si/C, (ie, we have compared the spectra obtained using a cathode of silicon carbide with the spectra obtained using (a) a silicon cathode, and (b) a carbon cathode), TiC/TiN/TiO₂/Ti/C, WC/W/C, (UC-ZrC)/UN/U/Zr/C, brass/Cu/Zn, and stainless steel/Fe/Cr/Ni. The arc current employed throughout was approximately 100 Amperes. We find that the charge state distributions change depending on the elemental composition of the alloy or compound of which the cathode is fabricated.

I. INTRODUCTION

The metal vapor vacuum arc is a plasma discharge that occurs between conducting electrodes in vacuum. The fundamental phenomenon which drives the vacuum arc is that of cathode spot formation - minute regions of intense current concentration which reside on the surface of the cathode and at which the solid cathode material is vaporized, ionized, and injected into the interelectrode arc region. The current density at the cathode spots is of order 10^6 A/cm² or more over a spot size of order microns. A typical vacuum arc discharge might consist of from one to many dozens of such spots. It is within the cathode spots that the plasma constituents of the arc are formed - the parameters of the arc are in large part determined by the plasma physics of the spots.

The vacuum arc discharge has been studied experimentally for many decades. An historical survey of the field, pre-1960s, has been given by Cobine,¹ and more recently a very complete review of the entire field of metal vapor arc discharges has been given by Lafferty.² A review of cathode spot behavior has been given by Lyubimov and Rakhovskii.³

Ion generation in the vacuum arc has been investigated by a number of authors.⁴⁻²⁰ Measurements of ion charge state distributions have been reported by several workers^{4-6,21,22} and it is well recognized that the distributions in general contain a high fraction of multiply stripped species. All these prior studies of the ion composition of the plasma have been for the case of a cathode which is a single, pure element.

The metal vapor vacuum arc ion source (called the MEVVA ion source, as an acronym) has been developed at LBL for the generation of high current beams of metal ions.²³⁻²⁸ Beams have been produced at voltages up to 100 kV and with pulsed ion currents up to 1 Ampere. The source works well with a wide range of ion species, spanning the Periodic Table from lithium to uranium. This device, in conjunction with a time-of-flight (TOF) diagnostic for charge-to-mass (Q/A) analysis, provides a most convenient means for the investigation of the ion charge state spectra of the vacuum arc plasma. The charge state distributions (CSD) obtained from a wide range of elemental cathode materials have been investigated and reported in a prior publication.²² In the work described here, we have used the MEVVA ion source and TOF charge state analysis diagnostic to investigate the ion spectra produced when the cathode is a compound or an alloy.

II. EXPERIMENTAL SET-UP

In the MEVVA ion source, the intense plume of highly ionized metal plasma that is created at the cathode spots of a metal vapor vacuum arc discharge is used to provide the "plasma feedstock" from which the ion beam is extracted. The quasi-neutral plasma plumes away from the cathode toward the anode and persists for the duration of the arc current drive. The anode of the discharge is located on-axis with respect to the cylindrical cathode and has a central hole through which a part of the plasma plume streams; it is this component of the plasma that forms the medium from which the ions are extracted. The plasma plume drifts through the post-anode region to the set of grids that comprise the extractor - a three grid, accel-decel, multi-aperture design. The MEVVA ion source has been described in detail in several prior publications.²³⁻²⁸

For the present work we used the MEVVA IV source version. In this source 16 separate cathodes are mounted in a single cathode assembly, allowing the operational cathode to be changed simply by rotating a knob so as to position the desired cathode in line with the anode and extractor of the device. Thus many different cathode materials can be compared in a relatively short experimental run and with confidence in maintaining the same experimental conditions. A photograph of the MEVVA IV source, partially-disassembled to show the multiple cathode feature, is shown in Figure 1.

For the experiments reported on here, the arc was driven by a simple LCR circuit with an effective pulse length of several hundred microseconds. The 240 μF capacitance is charged to a voltage of up to several hundred volts with a small, isolated, dc power supply. A high voltage pulse applied to a trigger electrode initiates a surface spark discharge between the trigger electrode and the cathode, which in turn causes the main anode-cathode circuit to close due to the spark plasma, and the vacuum arc proceeds. The source was operated at a repetition rate of several pulses per second, and the charge state spectral measurements were made at the time of peak arc current, approximately 100 A. Beam extraction voltage was 40 kV. The vacuum pressure was $(1-2) \times 10^{-6}$ Torr.

The charge state distribution (CSD) of the extracted ion beam was measured using a time-of-flight (TOF) diagnostic. In this device, an annular set of deflection plates is located in the beam path and biased so as to deflect the beam aside except for a short pulse of about 0.15 microseconds in length; in this way a short sample of the beam is obtained.²⁹ This short pulse is allowed to drift down a 1.25 m long region, during which drift time the different charge-to-mass (Q/A) components of the beam separate out, since they have been accelerated through the same potential drop in the ion source extractor and thus have flight times proportional to $(Q/A)^{-1/2}$. A detector, a well-shielded Faraday cup with magnetic suppression of secondary electrons, at the end of the drift chamber measures the arrival time of the different Q/A components of the beam. The detector is prevented from viewing the intense visible light and UV generated by the vacuum arc by blocking the direct path with a metal plate; the beam is steered onto the detector by the annular deflection plates. The detector measures the electrical current in the different Q/A states and provides a good measurement of the ion composition and CSD of the extracted ion beam. The measured flight times for the various charge states are well fitted by the calculated values, usually to better than the measurement uncertainty of about 1%. The time-of-flight system has been described in more detail in reference 29.

A schematic of the experimental configuration is shown in Figure 2.

III. RESULTS

The TOF spectra measured here are shown in Figures 3 through 9. Note that the first peak in all oscillograms, at 1 cm from the left hand side of the oscilloscope screen, is the TOF gate pulse; this provides a reference from which the flight times corresponding to the different spectral peaks can be measured. These data were all taken for the same arc current, $I_{\text{arc}} = 100$ A, and for a beam extraction voltage $V_{\text{ext}} = 40$ kV; the oscillogram sweep speed is either 0.5 or 1 $\mu\text{s}/\text{cm}$. The spectra were obtained as ion current collected by a Faraday cup, and the amplitudes of the charge state peaks in the oscillograms are then proportional to electrical current; the electrical current is greater than particle current by the charge state Q , $i_{\text{elec}} = Qi_{\text{part}}$. In order to obtain spectral data that would be visually intercomparable, the oscilloscope gain was adjusted for each cathode material, and the vertical current scale in the oscillograms is not the same for different materials; nonetheless, the current scale is always within a factor of several of 400 $\mu\text{A}/\text{cm}$.

Figure 3 shows the spectra obtained from cathodes of SiC, Si, and C. In these spectra it can be seen that the carbon ions produced from a graphite cathode are in the form of singly ionized C^+ only, with no trace of C^{2+} , whereas the carbon from SiC shows a small amount of C^{2+} . Also, the SiC ion spectrum does not appear to reflect the stoichiometry of the SiC solid. When the measured spectral peaks are converted into particle current ($i_p = i_e/Q$) and the charge state components summed, the spectrum indicates that there is approximately twice as much Si in the beam as C.

Figure 4(a) shows the spectra obtained from cathodes of TiC, Ti, and C. Here, as well as the O^+ contaminant in the carbon, we can also see Co^+ and Co^{2+} in the TiC spectrum; cobalt is used as a cement in the preparation of solid TiC, with a concentration of typically about 7%. Similarly to the case for SiC as shown in Figure 3 and described above, here again there is an enhancement in C^{2+} in the TiC spectrum over that in the pure carbon spectrum; additionally, the Ti^+ is increased and the Ti^{3+} decreases in the TiC over that in the titanium spectrum. That is, in the compound TiC cathode, the carbon CSD has been increased and the Ti CSD has been decreased. The TiC spectrum once again does not show the Ti:C fractions expected from the TiC stoichiometry.

The TiN spectrum is shown in Figure 4(b). The Ti spectrum has been decreased to lower charge states as compared to that obtained with a titanium cathode. Only the singly ionized N^+ charge state of the non-metallic nitrogen component is formed. The Ti to N particle fraction ratio in the beam is about 75%:25%, compared to the stoichiometric ratio of 50:50.

In Figure 4(c) the spectrum obtained from a specially prepared TiO_2 cathode is shown. This is particularly interesting in that TiO_2 is an insulator. This cathode material was formed by evaporating a small percentage of copper onto the micron-sized grains of SiO_2 so as to allow the powder to be pressed into the form of a solid conductor; this technique could be of use in a variety of applications. The small fraction (less than 5%) of copper used shows up in the spectrum, and Cu^+ and Cu^{2+} peaks can be seen. As in the SiC, TiC, and TiN spectra, note that here again the Ti CSD has been severely decreased to lower charge state over that from a pure titanium cathode, with now Ti^{3+} having disappeared completely and Ti^+ having become the dominant charge state component. The Ti:O ratio is seen to be well away from the TiO_2 stoichiometry, with the non-metallic component, oxygen, constituting only about 15% of the total ion particle current.

The spectrum of WC, W, and C are shown in Figure 5. Tungsten is notable for the quite high charge states that are produced for the elemental cathode - up to W^{6+} in significant percentage of the distribution. The compound spectrum shows a lot of contaminants - N^+ , O^+ , what is interpretable as Co (used as a cement in the manufacture of WC), plus some unidentified lines. Nevertheless the carbon component of the compound spectrum displays a high fraction of C^{2+} ; the formation of the doubly-ionized C^{2+} in the compound spectrum, compared to the purely singly ionized C^+ in the elemental carbon spectrum is particularly dramatic. At the same time as the carbon mean charge state has been increased in the compound cathode spectrum, the tungsten mean charge state has been severely decreased.

Figure 6 shows some spectra for uranium compounds. In Figure 6(a) are shown spectra obtained for the case of a cathode made of a sintered mixture of the compound UC with the compound ZrC, and the elemental spectra of the components U, Zr, and C. The picture here follows the same trend as for the cases above - the carbon charge states are increased and the uranium and zirconium charge states are decreased. In Figure 6(b) UN and U spectra are compared; again the uranium charge state is decreased.

The spectra obtained from cathodes of brass, copper, and zinc are compared in Figure (7). One cannot distinguish the Cu and Zn peaks in the alloy spectrum, but it is clearly seen that diminution in the Cu spectrum is dramatic - the Cu^{3+} and Cu^{4+} peaks are removed completely from the alloy spectrum.

Spectra from type 304 stainless steel and its component elements Fe, Cr, and Ni are shown in Figure 8. The situation in this case is quite similar to that in the brass spectrum. Although the lines for Fe, Cr, and Ni lie too close together to be distinguishable, none the less it is clear that all of the high charge states are lost from the alloy.

Finally, the spectrum obtained from a lanthanum hexaboride cathode, LaB_6 , is shown in Figure 9. Although we do not have spectra from cathodes of La or B with which to compare the LaB_6 data, this measurement is of interest by itself in that it shows a high fraction of the doubly ionized B^{2+} in the spectrum. The particle fraction ratio determined from the spectrum shown is $\text{La}:\text{B} = 25\%:75\%$.

IV. DISCUSSION

We have found that a vacuum arc plasma can be formed from a cathode which is a compound or alloy material, and that the plasma constituents as determined from the ion beam composition are composed of the cathode constituents. Thus it is possible to form intense beams of mixed ionic species using the MEVVA high current metal ion source; this could be convenient for a number of basic and applied applications. It is particularly interesting that an arc, containing all the elemental cathode constituents, forms not only for the case when the cathode constituents are metallic but also when some of the components are non-metallic, as for example N and O. Although the cathode itself must necessarily be electrically conducting in order for the discharge to occur, it is not a necessary condition that all of the elemental constituents of the cathode material be metallic.

The plasma composition for some of the cathode materials investigated here is given in Table I. In this Table, the plasma fraction is the percentage of the plasma that is composed of the elemental species, summed over all charge states, obtained from the TOF data of the previous section, and expressed as a particle fraction. The particle fraction has been obtained from the current fraction (raw oscillogram data); thus for example the un-normalized particle fraction for a given element has been obtained from $f_p = \sum i_e/Q$, where i_e is the amplitude of the Q'th charge state peak. The molecular components are all represented in the plasma from the compound cathode, but the elemental fractions in the plasma do not appear to reflect the stoichiometric fractions of the cathode materials with any strong correlation.

The CSD's of the ions produced from the elemental constituents of the compound or alloy cathode materials are different from the CSD's produced from the purely elemental cathode materials. Those elemental components that by themselves produce relatively high charge states suffer a reduction in the higher charge state fractions when incorporated into a compound cathode, and conversely, those elements that by themselves produce relatively low charge states experience an increase to somewhat higher charge states. For example, the higher charge states normally present in U or W are diminished in the compound case; C, on the other hand, normally shows only the singly ionized C^+ component, but in the compound case the doubly-ionized C^{2+} component is present also. Thus the parameters of the cathode spot plasma, where the ionization takes place and the charge state distribution is formed, must be different in the two different cases.

It is interesting to refer the brass/Cu/Zn spectra shown in Figure 8 to the ionization potentials of the elements involved. The presence of Cu^{4+} in the Cu spectrum implies the existence of a component of plasma electrons with energy above 55.2 eV, this being the ionization energy needed to produce the quadruply-ionized state.³⁰ In the Zn spectrum, on the other hand, the fraction of electrons with energy greater than 39.7 eV is small (ionization energy for the production of triply-ionized Zn).³⁰ The vapor pressure of zinc is much higher than that of copper, and the zinc plasma must be highly collisional due to the presence of a high fraction of neutral zinc atoms, leading to electron cooling and consequently lower charge states. A similar consideration may play a role in the case of the brass plasma, and in the apparent lowering of the copper charge states by the presence of the zinc.

V. CONCLUSION

Measurements have been made of the ion spectra and charge state distributions of the vacuum arc plasma produced from a number of compound and alloy cathode materials. These spectra have been compared to those obtained from cathodes composed of single elements.

We find that the plasmas formed from compound and alloy cathodes contain species from all of the cathode elemental constituents, although the plasma fractions can depart significantly from the stoichiometric fractions. Even for the case when the cathode material molecular composition contains non-metallic atomic components, such as nitrogen or oxygen, these non-metallic components are also formed into the plasma state. It is a requirement, however, that the cathode itself be macroscopically electrically conducting, in order for the vacuum arc to take place at all.

The charge state distributions of the component ionic species in the plasma from compound or alloy cathodes can be quite different from the charge state distributions produced in plasma from elemental cathodes. Those species which in the elemental cathode case have higher mean charge state suffer a reduction in mean charge state in the compound case, and conversely the lower mean charge state species see an increase in mean charge state - there is an averaging effect in the compound cathode case.

ACKNOWLEDGMENTS

We are indebted to Bob MacGill and Mark West for their support of the mechanical systems of the experimental facilities, and to Jim Galvin for support of the electrical and electronics systems.

This work was supported by the U.S. Department of Energy under Contract No. DE-AC03-76SF00098, and by the Army Research Office and the Office of Naval Research under Contract No. N00014-88-F-0093.

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TABLE I

Cathode Material	Element	Stoichiometric Fraction (%)	Plasma Fraction (%)
SiC	Si	50	67
	C	50	33
TiC	Ti	50	60
	C	50	40
TiN	Ti	50	75
	N	50	25
TiO ₂	Ti	33	85
	O	67	15
UN	U	50	50
	N	50	50
UC-ZrC	U	25	15
	Zr	25	45
	C	50	40
LaB ₆	La	14	25
	B	86	75

Comparison of the cathode composition with the composition of the ion beam obtained from the cathode. "Stoichiometric fraction" is the atomic fraction of the given element in the cathode, and "Plasma fraction" is the measured fraction of particle current (all charge states) of the given element in the ion beam.

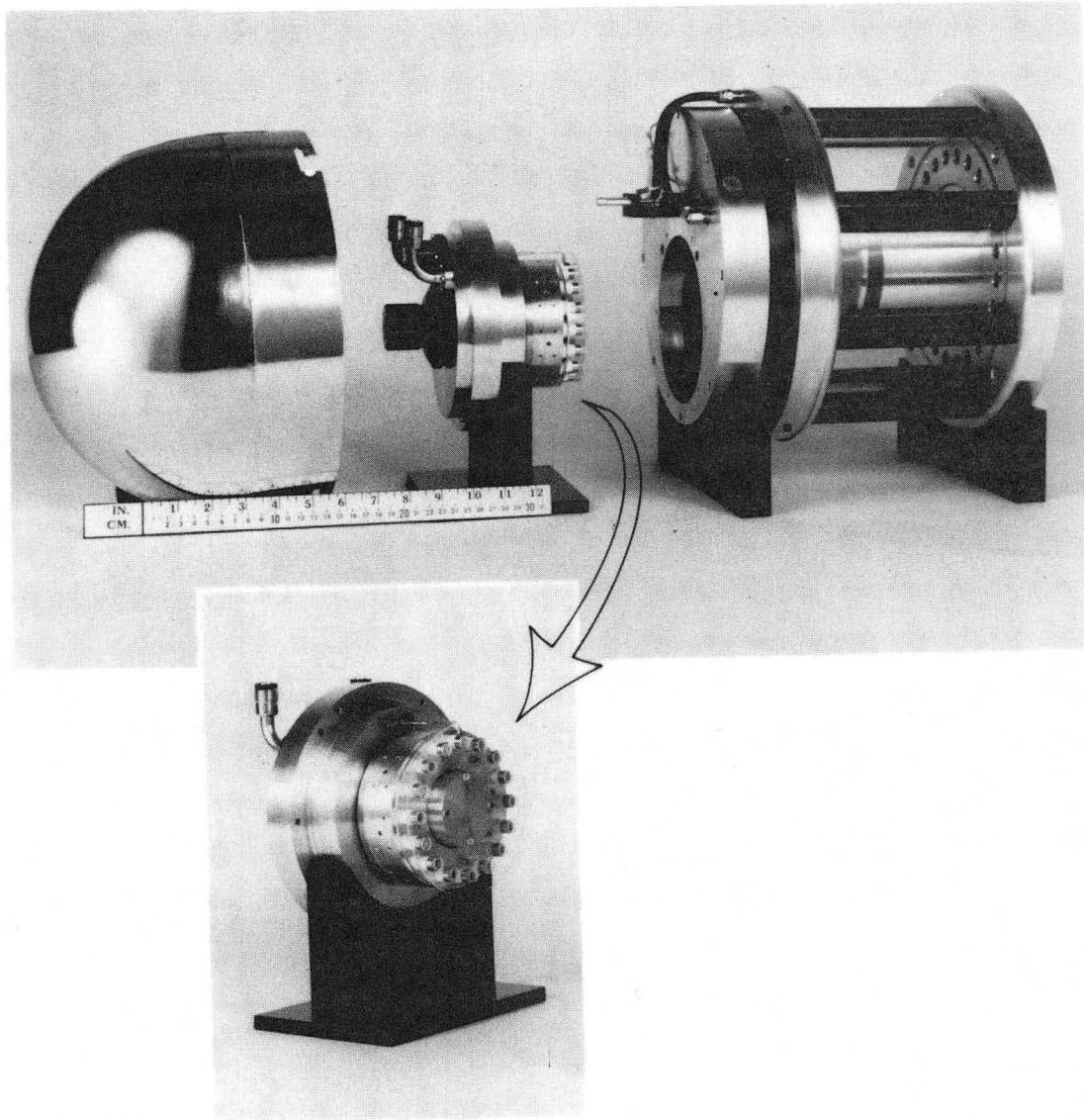


Figure 1 The MEVVA IV ion source, showing the multiple cathode configuration, CBB860-9309A.

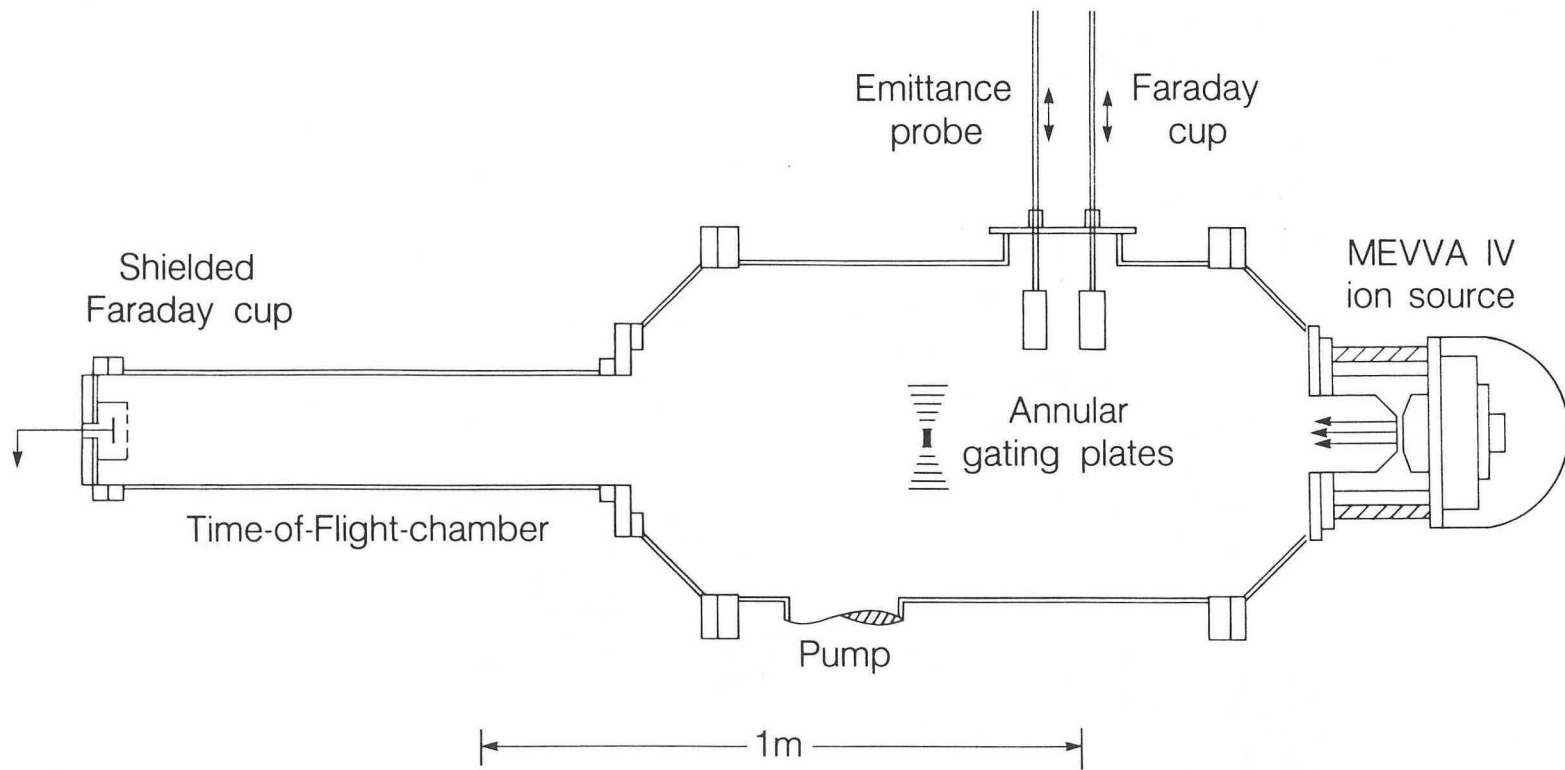


Figure 2. Schematic of the experimental configuration.

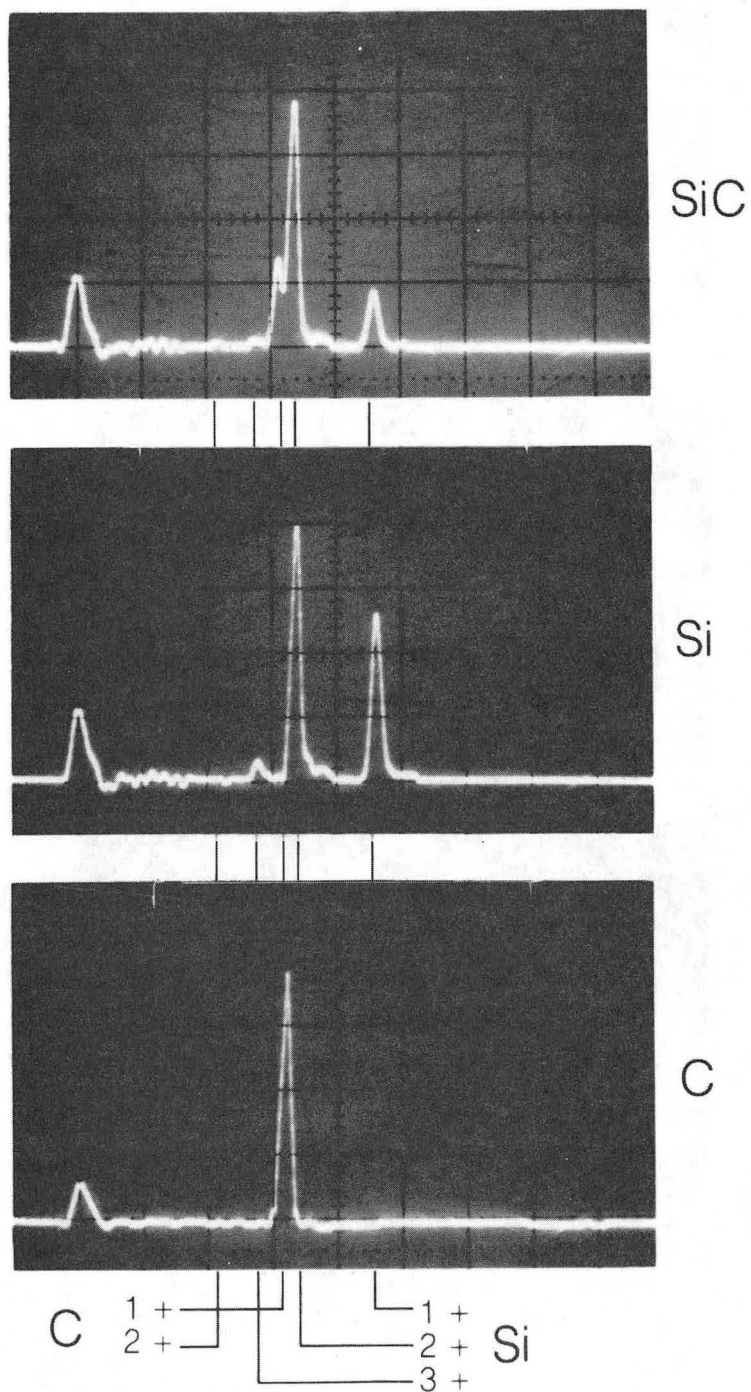


Figure 3 Time-of-flight charge state spectra for SiC, Si and C. Vertical scale: current collected by the Faraday cup, gain approx. 400 $\mu\text{A}/\text{cm}$. Horizontal sweep speed: 0.5 $\mu\text{s}/\text{cm}$, SBB 8810-10411.

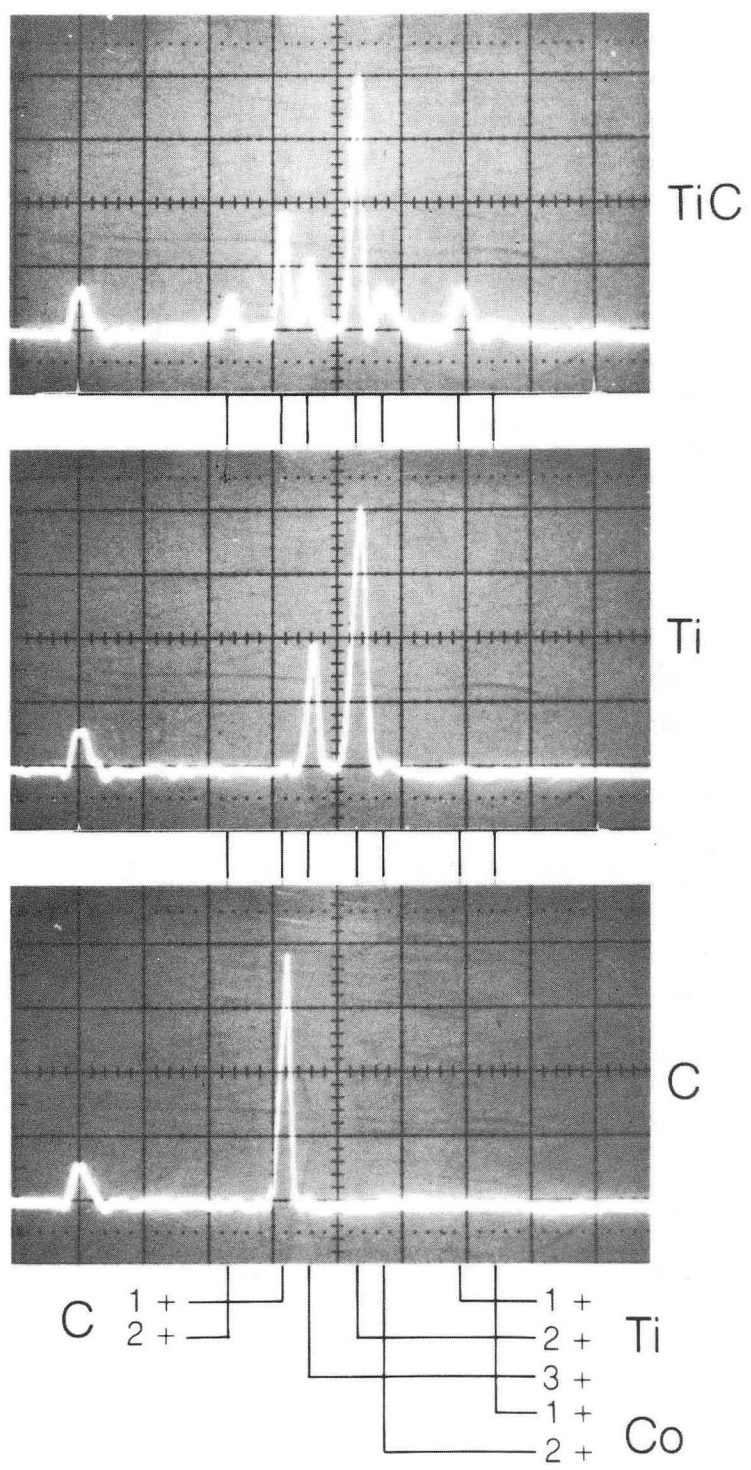


Figure 4(a) Charge state spectra for TiC, Ti and C, XBB 8810-10412.

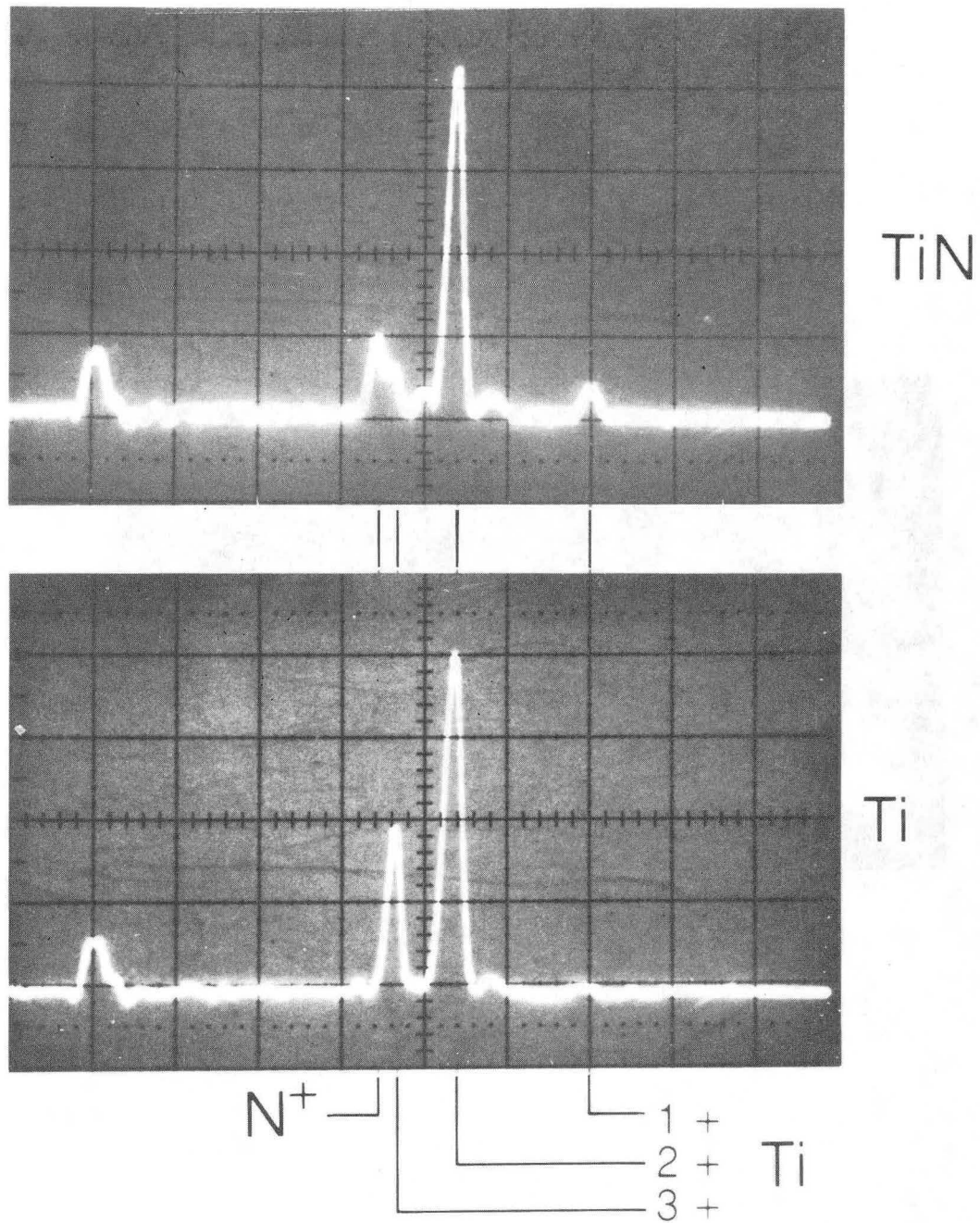
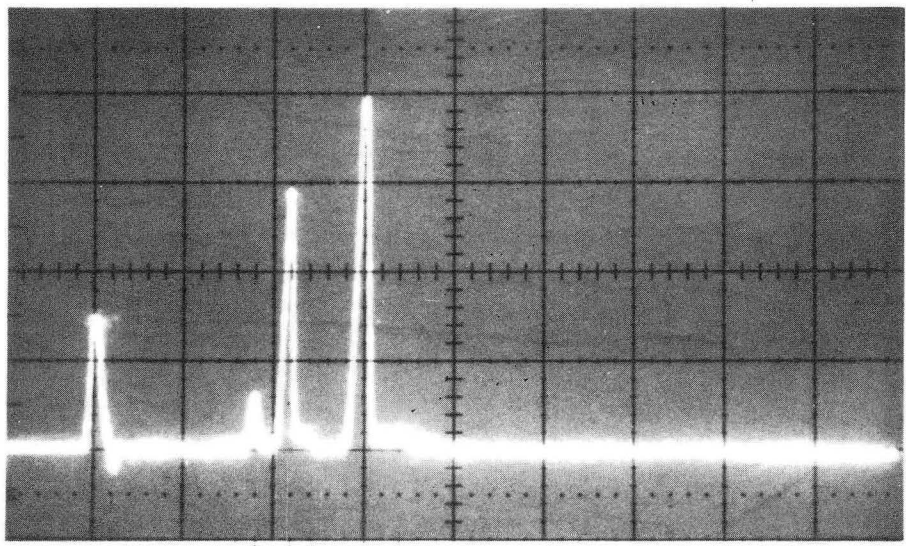


Figure 4(b) Charge state spectra for TiN and Ti, XBB 8810-10417.



TiO₂

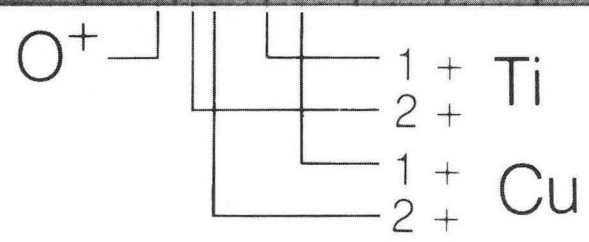


Figure 4(c) Charge state spectra for TiO₂, XBB 8810-10415.

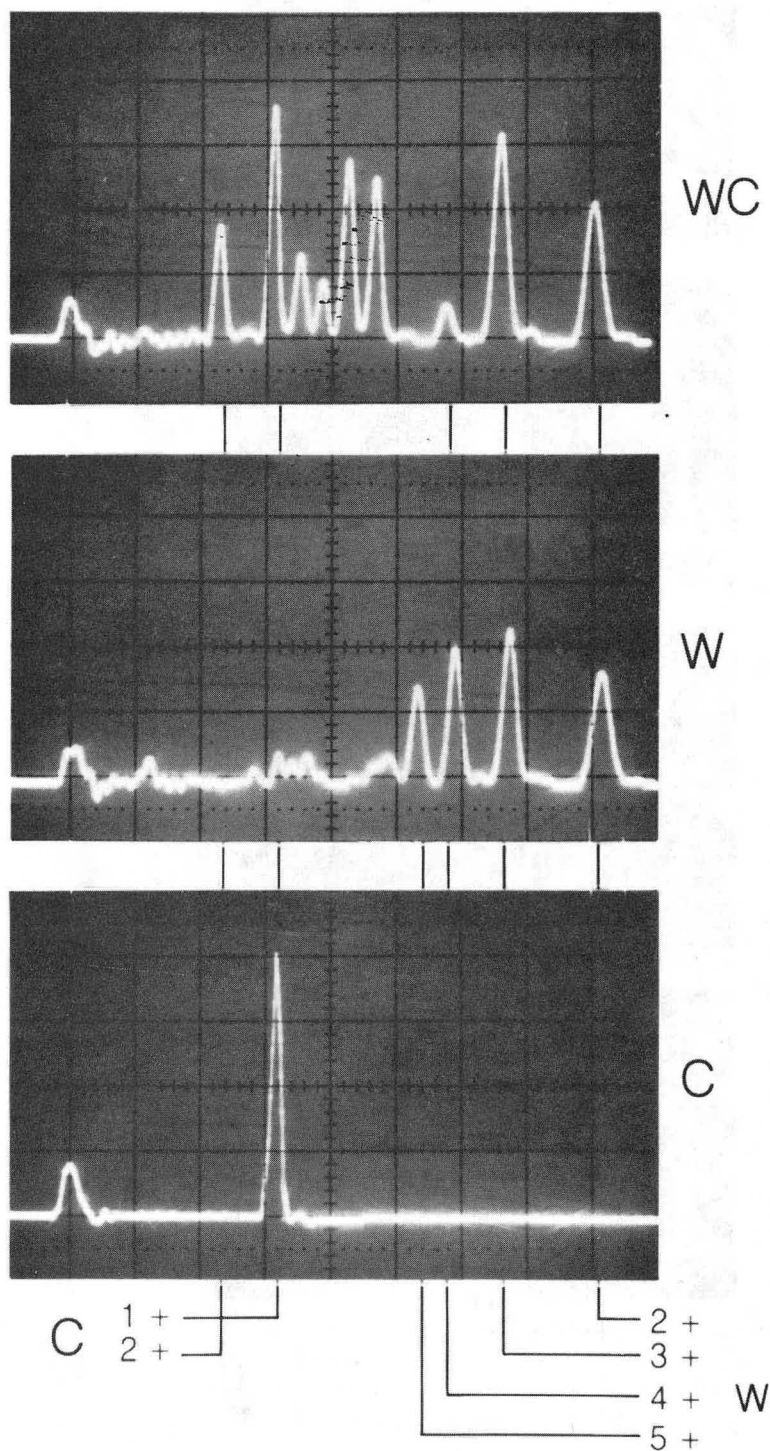


Figure 5 Charge state spectra for WC, W and C. XBB 8810-10413.

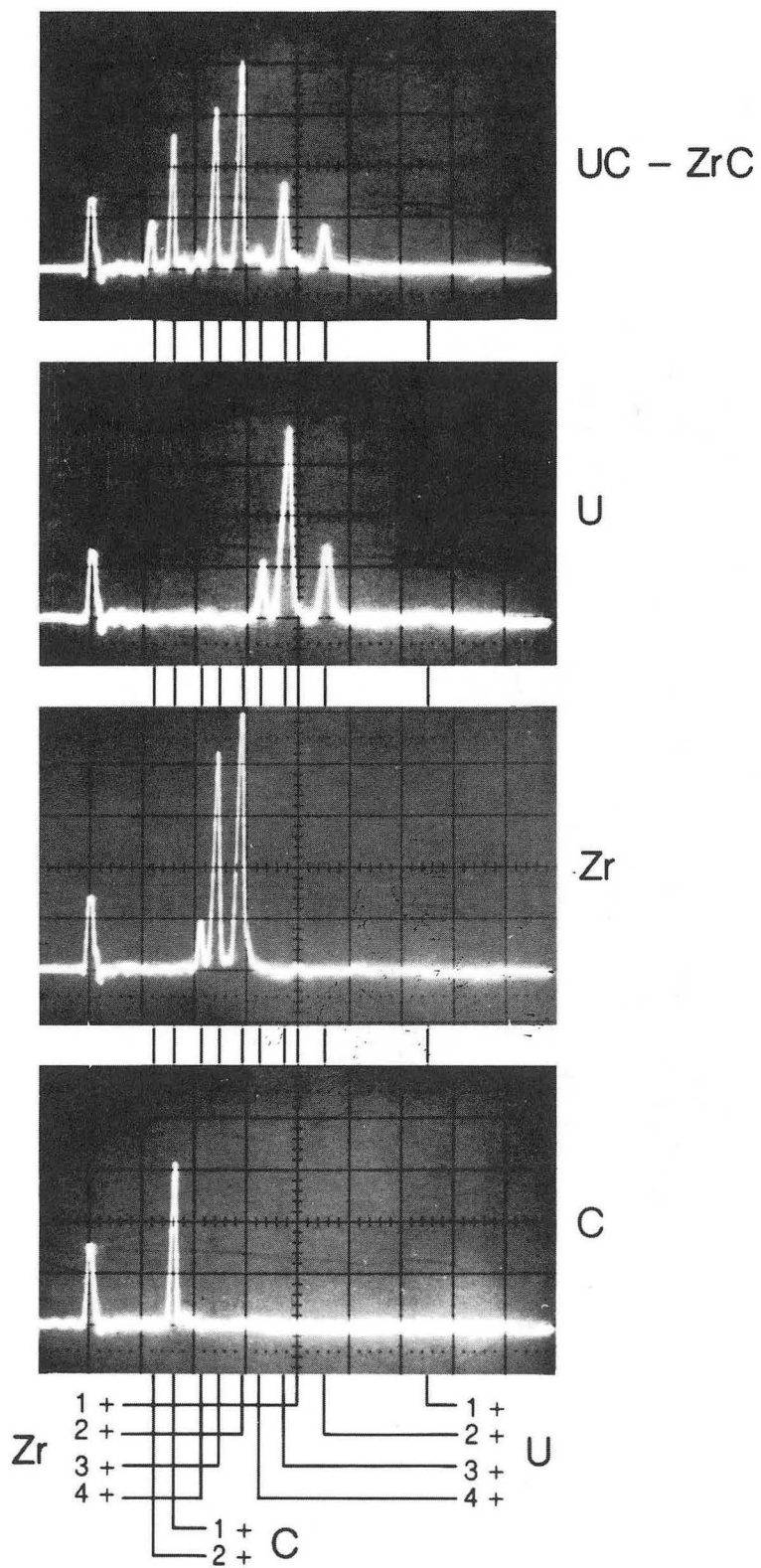


Figure 6(a) Charge state spectra for UC-ZrC (sintered alloy of U, Zr and C, XBB 8810-10421).

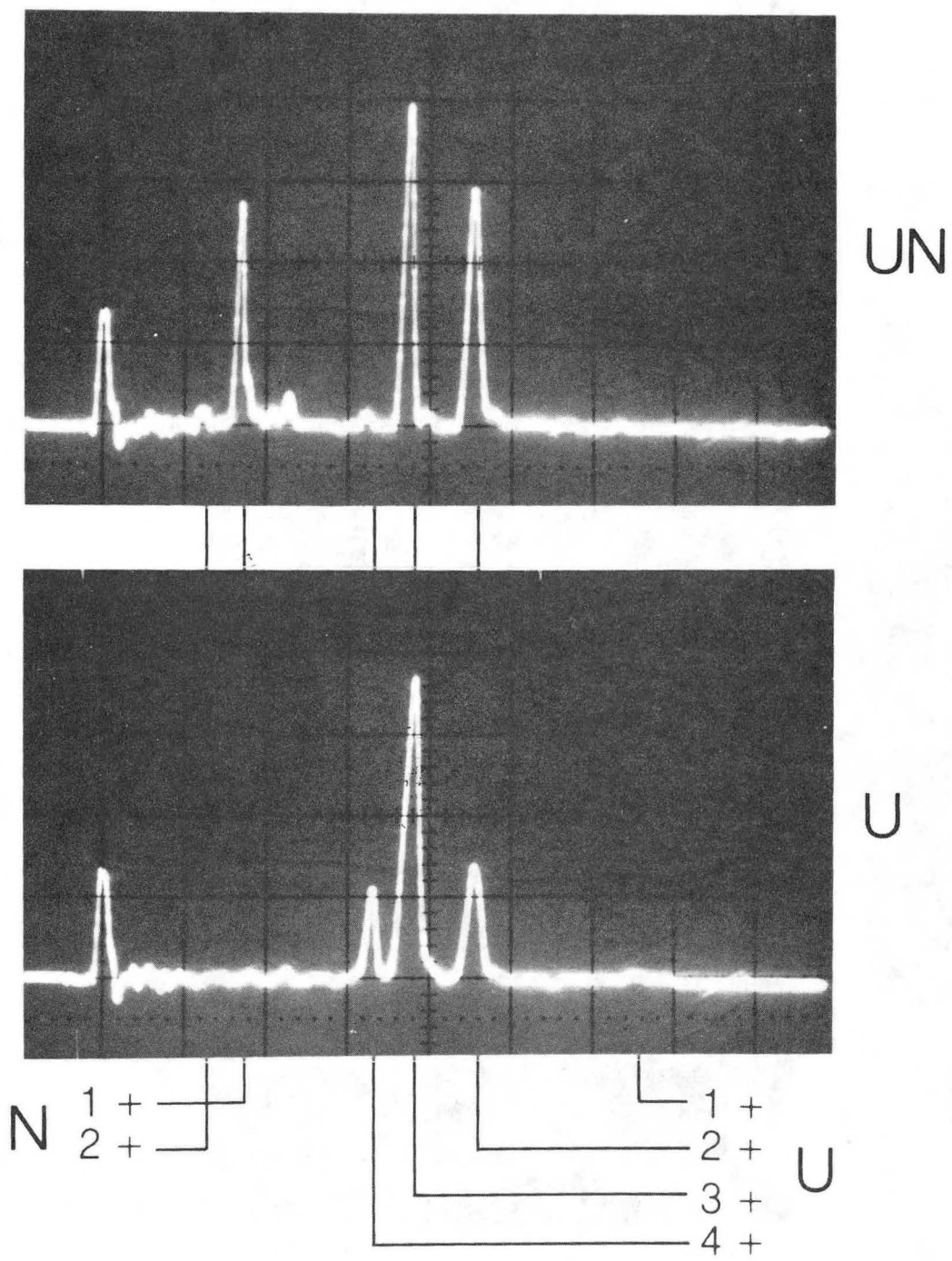


Figure 6(b) Charge state spectra for UN and U, XBB 8810-10421

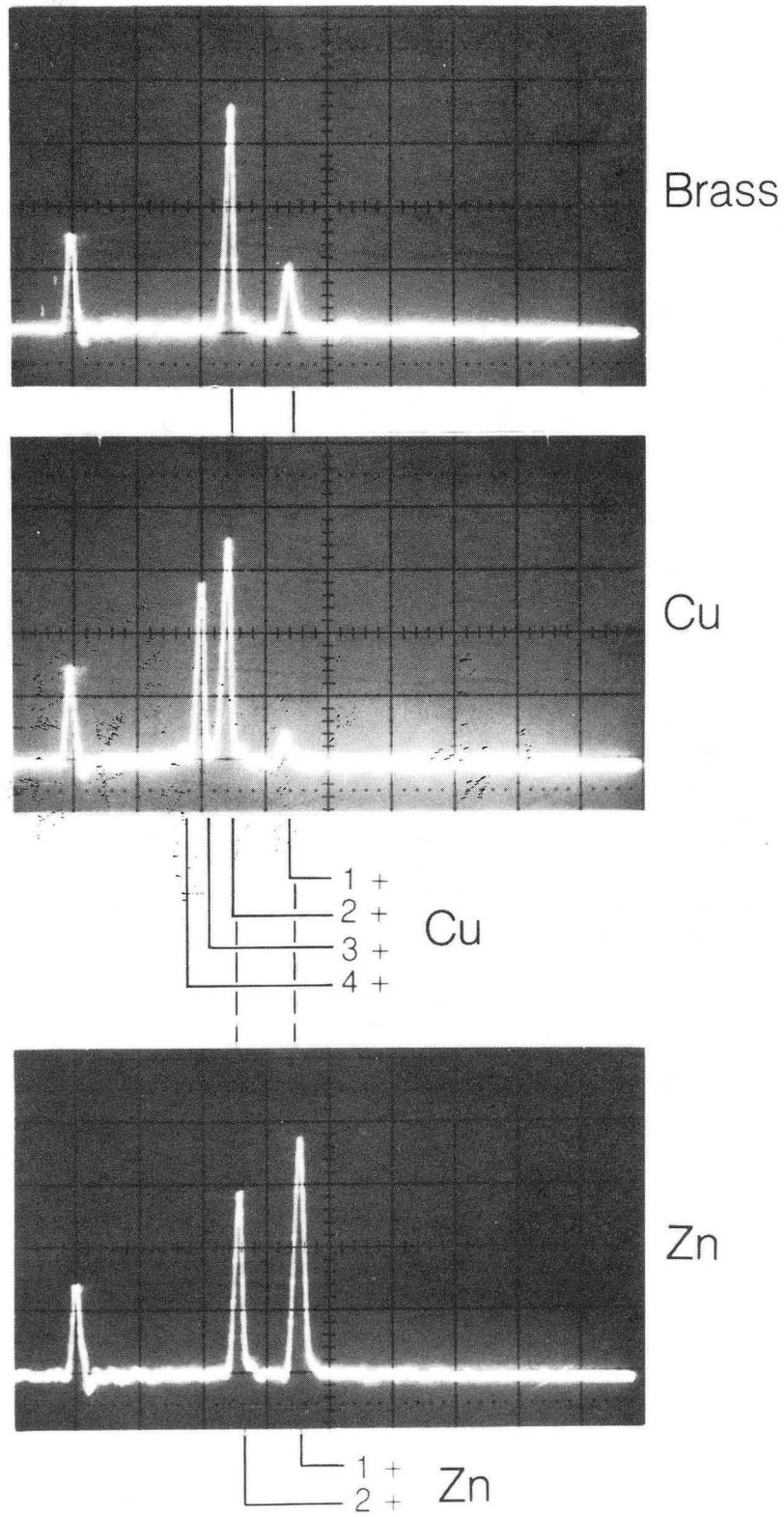
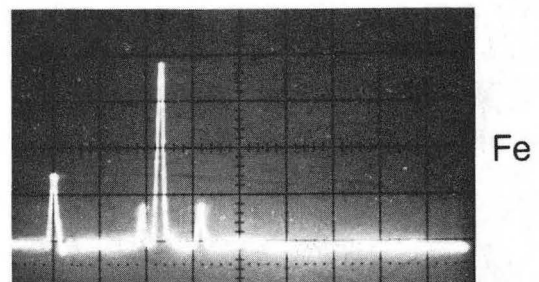
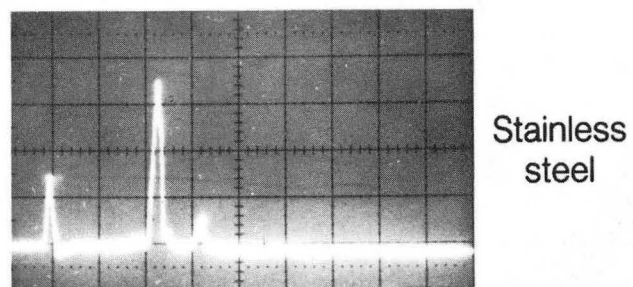
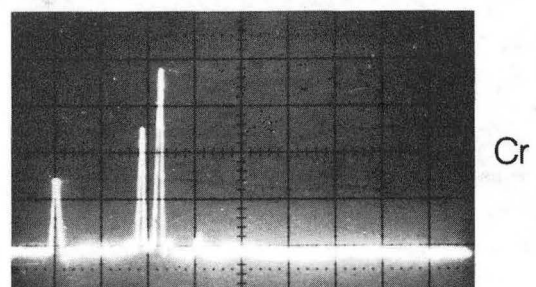


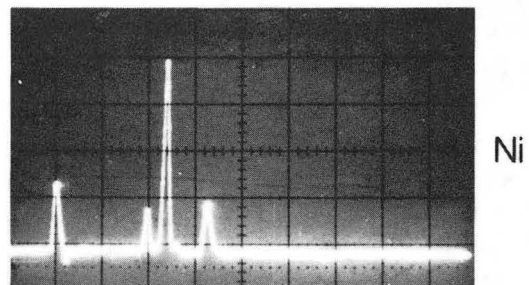
Figure 7 Charge state spectra for brass, Cu and Zn, XBB 8810-10416.



1 +
2 +
3 +



1 +
2 +
3 +



1 +
2 +
3 +

Figure 8 Charge state spectra for 304 stainless steel, Fe, Cr and Ni, XBB 8810-10421.

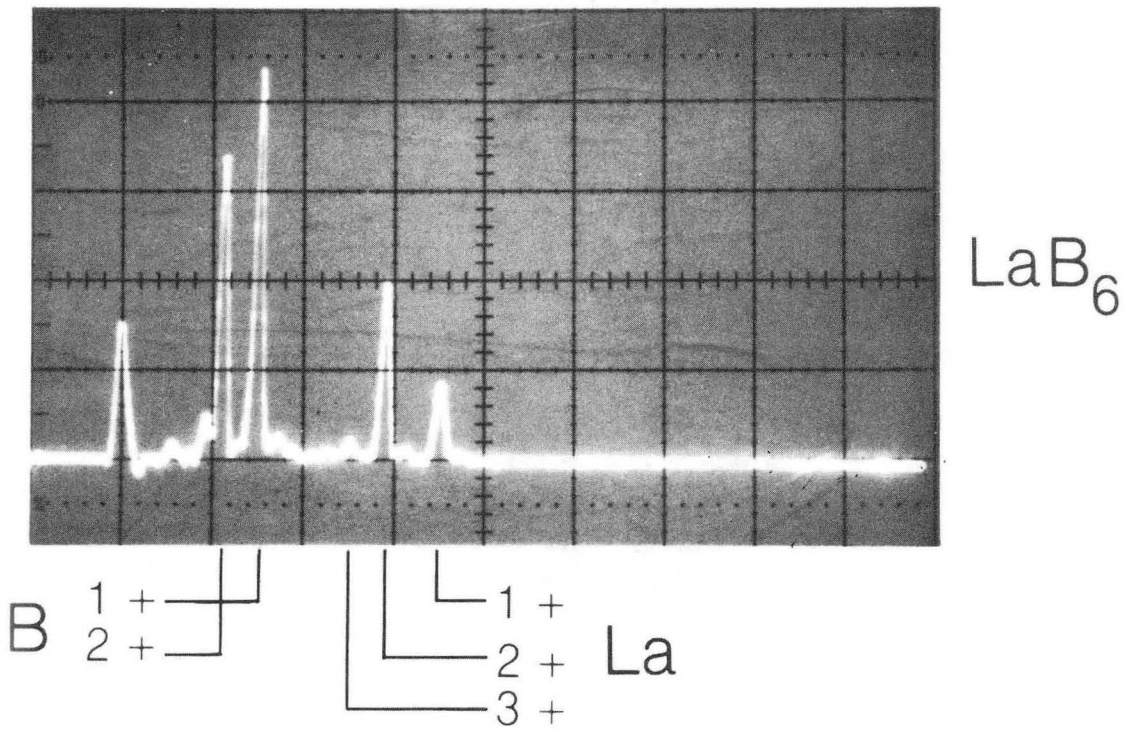


Figure 9 Charge state spectra for LaB_6 , XBB 8810-10414.

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