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ISOTOPIC CONSTITUTION OF PLUTONIUM. I

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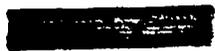
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ABSTRACT

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A mass-spectrographic examination of plutonium has revealed a peak at the 240 mass position, presumably showing presence of  $^{240}\text{Pu}$ . Evidence as to the reality of this peak is not yet complete.



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ISOTOPIC CONSTITUTION OF PLUTONIUM. I

I. INTRODUCTION

Fermi has suggested that the anomalously high spontaneous-fission rates observed in  $49$  may be due to  $94^{240}$ . Experimental substantiation of this hypothesis has been obtained by Chamberlain, Farwell, and Segre<sup>1</sup>).

Heretofore, only one method was available for estimating the amount of  $94^{240}$  present in a given sample of element  $94$ ; this method was based on knowledge of the irradiation,  $nvt$ , the sample had received and on the value of  $\alpha$ . Bacher has pointed out that a mass-spectrographic identification of  $94^{240}$  would give a desirable check on the qualitative nature of the fundamental processes involved; and that in combination with other obtainable data, a quantitative mass-spectrographic determination of  $94^{240}$  could be used to derive the absolute value of the spontaneous-fission constant  $\lambda_p$  and the value of  $\alpha$  as a function of energy.

The  $94$  mass-spectrometer program was undertaken with the following objectives;

- 1) Positive identification of the isotope  $94^{240}$ .
- 2) Quantitative determination of the amount of  $94^{240}$  present in a given sample of  $94$ .
- 3) Tentatively, the isolation of a small sample (e.g. a fraction of a microgram) of  $94^{240}$ .

II. EXPERIMENTAL

A. The Mass Spectrometer

The previous experience of one of the authors with the Nier hexafluoride mass spectrometer led us to believe that it should be possible to detect and measure

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- 1) O. Chamberlain, G. W. Farwell, and E. Segre; LAMS - 131

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$94^{240}$  to one part in  $10^4$  of plutonium. Accordingly, one of the standard Nier instruments was procured and set up with slight modifications for this application. The principal modification and the only one to be mentioned in the present preliminary report, is that of the ion source.

No compounds of plutonium are known at the present time with volatilities comparable to that of uranium hexafluoride. In fact, chemical knowledge at the time of this report indicates that the most volatile compound of which the chemistry is known is plutonium trichloride, which, according to Weinstock<sup>2)</sup>, has a vapor pressure of about  $4 \times 10^{-4}$  mm at  $700^\circ$  C. On the advice of Weinstock and Garner, we did not undertake to use this compound in an ion source as it has been found to be exceedingly hygroscopic and would, therefore, undoubtedly present a serious vacuum problem in the mass spectrometer unless very exacting handling techniques were developed. On the other hand, the results obtained by Perlman, Lipkin, and Weissman have shown that under the proper conditions and with the proper techniques, plutonium metal itself can be readily vaporized in high vacua. Accordingly, the ion source in the standard Nier spectrometer was modified to include a micro-furnace of tantalum from which the metal was vaporized. Fig. 1 is a scale drawing of the modified arrangement. The tantalum crucible (Fig. 1A) is heated to 1100 to  $1300^\circ$  C by radiation from a tungsten filament (Fig. 1B) which is surrounded by a tantalum radiation shield, (Fig. 1C). The vapor pressures<sup>3)</sup> of plutonium at these temperatures are high enough to give sufficient ion currents to permit accurate measurements. Certain vapor shields which were found necessary to prevent electrical conduction on the glass insulators are not shown in the figure. Other details of the spectrometer have already been published by

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2) B. Weinstock; LA - 122

3) T. E. Phipps, G. Sears, L. S. Phipps, and G. C. Simpson; CK - 1763, p. 31 ff

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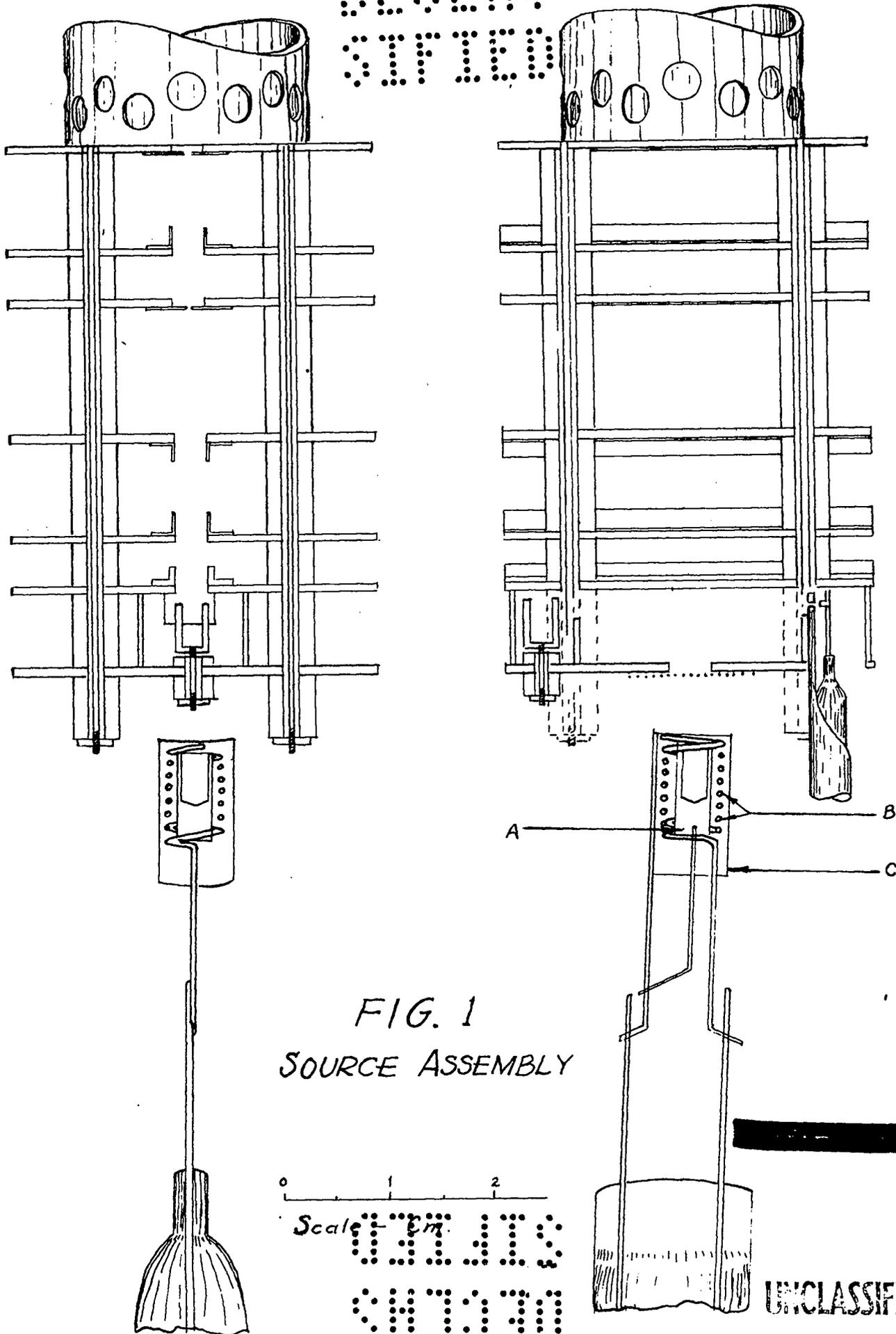


FIG. 1  
SOURCE ASSEMBLY

Scale - 2 cm.

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Nier<sup>4)</sup>. It should be mentioned that in this application the spectrometer tube was inverted and mounted on a separate open metal frame to facilitate operations on the tube and its accessories.

### B. Adjustment and Tests of the Mass Spectrometer with Mercury

The presence of mercury in the mass spectrometer from the mercury diffusion pumps invariably used in this type of instrument furnishes a useful and convenient source of ions for preliminary adjustment and routine checking of the operation of the instrument. The adjustment of the spectrometer to optimum resolution and intensity was done through out-and-try tests on the resolution of the  $\text{Hg}^{202}$  and  $\text{Hg}^{201}$  ions. The mercury spectrum was found to agree with that of the literature<sup>5)</sup>. Fig. 2 is a plot of the mass spectrum about the  $\text{Hg}^{202}$  and  $\text{Hg}^{201}$  peaks. The two vertical dotted lines at the base of each peak represent the theoretical width of the peak as given by the equation:

$$w = \frac{R \alpha^2 + s_1}{(R/m)\Delta m} \quad (1)$$

where

$w$ = width of peak in mass units	= 0.314
$R$ = radius of curvature	= 15.25 cm
$\alpha$ = half-angle of divergence of ions from source	= 0.0151 r
$s_1$ = source-slit width	= 0.0203 cm

$$(1/m)\Delta m = 1/200$$

### C. Tests with Tin

It is possible to carry out the above adjustments without use of the micro-furnace, since mercury vapor, with  $\text{CO}_2$ -alcohol slush on the cold trap, was present to

4) A.C.C. Nier, M.G. Inghram, and E.P. Ney; A-203

5) Nier, Phys. Rev., 52, 931, (1937)

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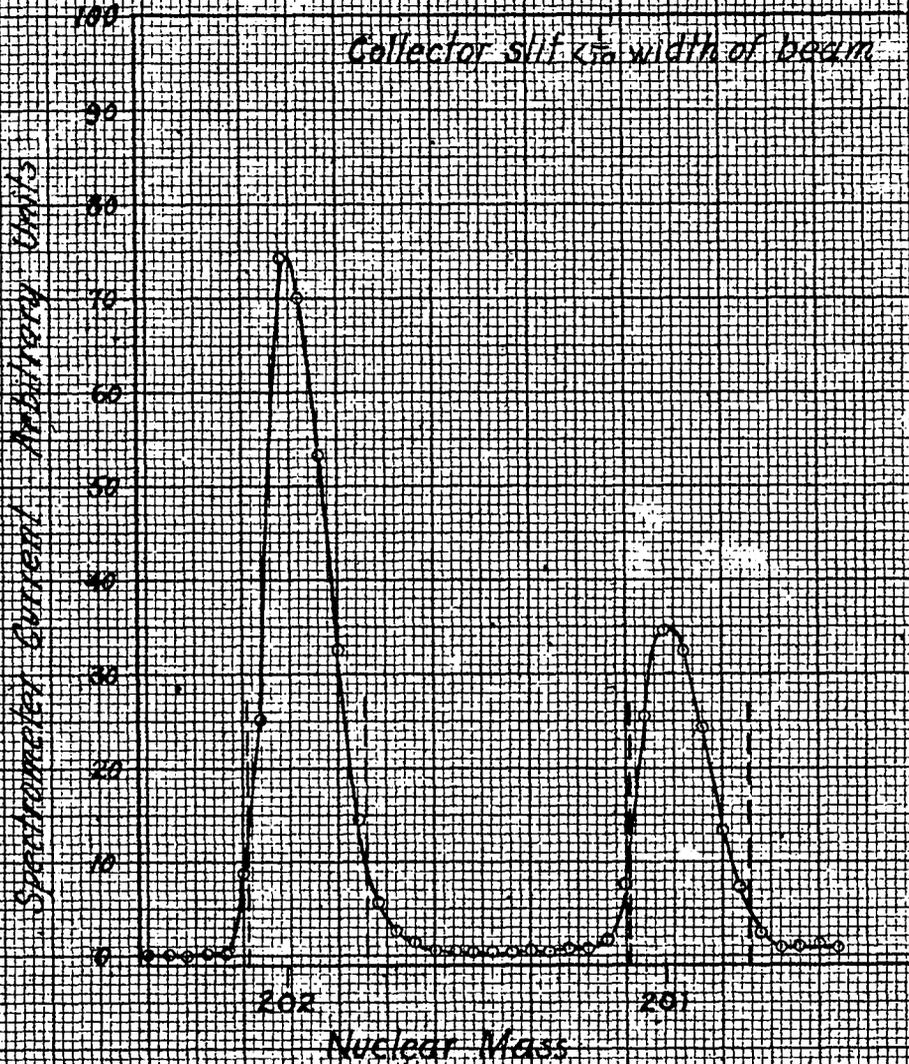


FIG 2  
Mass Spectrum of Hg<sup>202</sup> and Hg<sup>201</sup>

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a pressure of about  $10^{-9}$  mm. A further check was made on the performance of the instrument before proceeding to studies on element 49. Several metals are known to have vapor pressures comparable with that of plutonium. Of these, tin was selected for use as a stand-in and was introduced in a Mo crucible. Fig. 3 is a mass spectrum showing the isotopes  $\text{Sn}^{120}$  and  $\text{Sn}^{119}$ . This result indicated that modifying the ion-source did not result in loss of resolution.

The ratio of the intensities of the two peaks of Fig. 3 does not give the abundance ratio  $\text{Sn}^{119}/\text{Sn}^{120} = 0.34$  reported by Aston<sup>6</sup>). This is caused by a shift in spectrometer current during the detailed recording of this spectrum. Measurements of the ratio made in rapid alternation confirmed the ratio given by Aston to a few percent.

The background at the 120.5 mass position was found to be less than 1/10,000 of the current due to  $\text{Sn}^{120}$  at the 120.0 mass position. This is indicative of the background to be found at the  $\text{Pu}^{240}$  position in the case of 49.

### III. PRELIMINARY RESULTS ON 49

During the experiment performed on tin, difficulty was encountered in that the metal did not vaporize freely, presumably because of the formation of a surface film during the process of baking. This effect required the use of higher furnace temperatures than were anticipated, and at such temperatures it was found that the glass insulators in the ion source began to give electrical conduction, which conduction disappeared on cooling. On the basis of the experience of Lipkin, Perlman, and Weissman, it was believed that the formation of such a film on 49 would not cause difficulties, since, according to their experience, tantalum is wet readily by molten

6) Aston, Nature, 137, 613 (1936)

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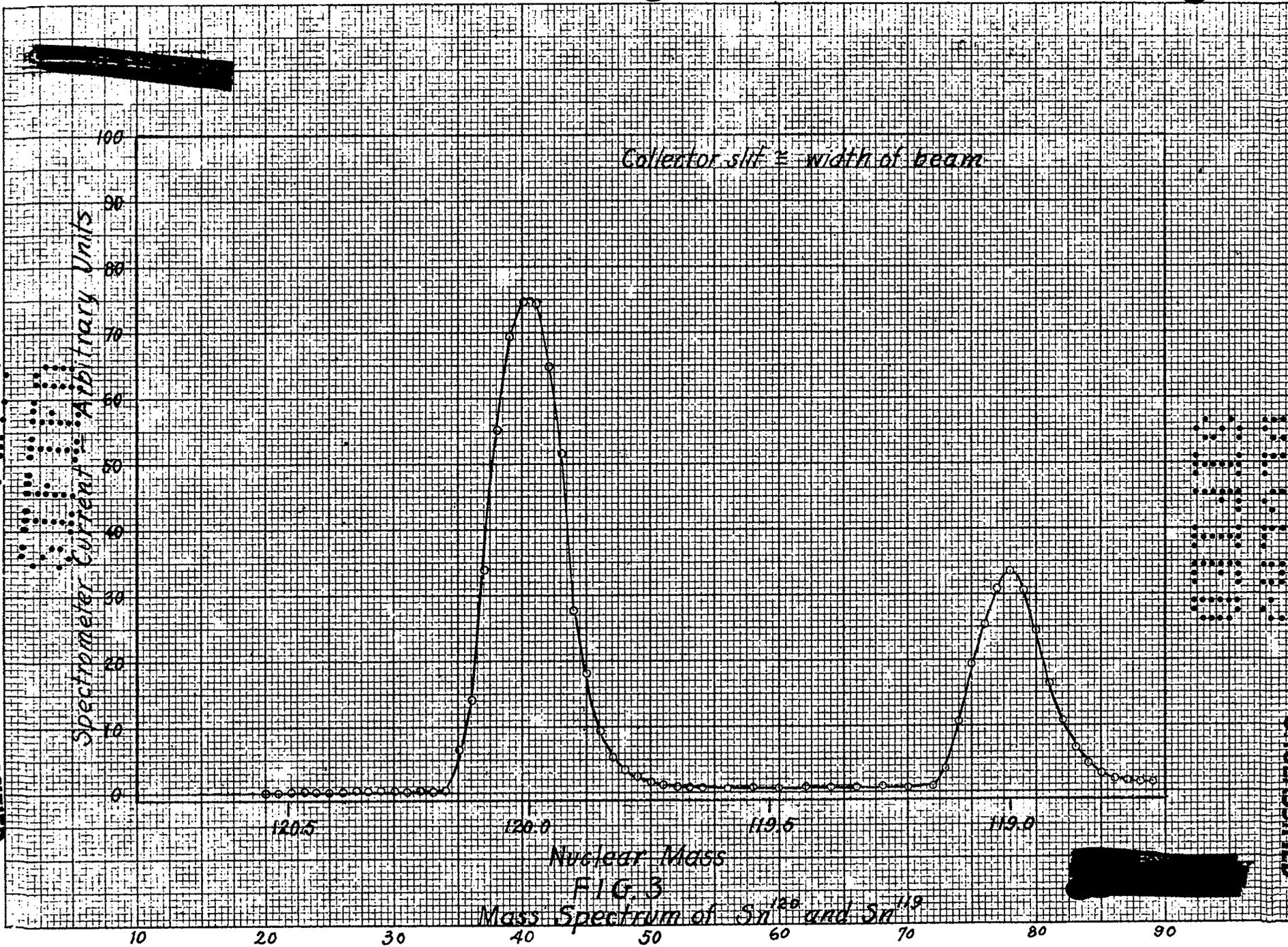


FIG. 3  
 Mass Spectrum of  $Sn^{120}$  and  $Sn^{119}$

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49 at temperatures of 1100° C. One would expect, then, that after the solid charge of 49 melted, surface tension would carry the free metal to adjacent clean walls of the crucible, whence it could vaporize freely.

The sample of metal used in the first run had the following history:

a) Purification by Wahl

TABLE I

Lot No. 209-P

X's Lot No.	Our Lot No.	Date rec'd from X	Grams
34-C	204-X	10-16-44	1.271
34-D	205-X	10-16-44	3.760
26-A	206-X	10-21-44	1.503
36-A	207-R	10-28-44 purif.treat.	1.638

b) Reduction by Baker. Sample Nos. 5414 and 5415.

Reduced from  $\text{PuF}_4$  with Ca, in MgO-lined crucible.

b) Vacuum-Remelting by Litz and Hammel

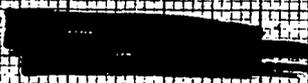
The sample was remelted in vacuum at 1200° C for 30 minutes in a CeS crucible.

Seventy-one mg of this 49 metal was introduced into the spectrometer. The mass spectrum indicating the nature of the results is given in Fig. 4. In addition to the large peak observed, assumed to be at mass 239, a small peak was found at the position then corresponding to mass 240. As indicated by the spread of the points on the 240 peak, the measurement was made near the limit of stability of the FP-54 ampli-

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Collector slit  $\approx$  width of beam

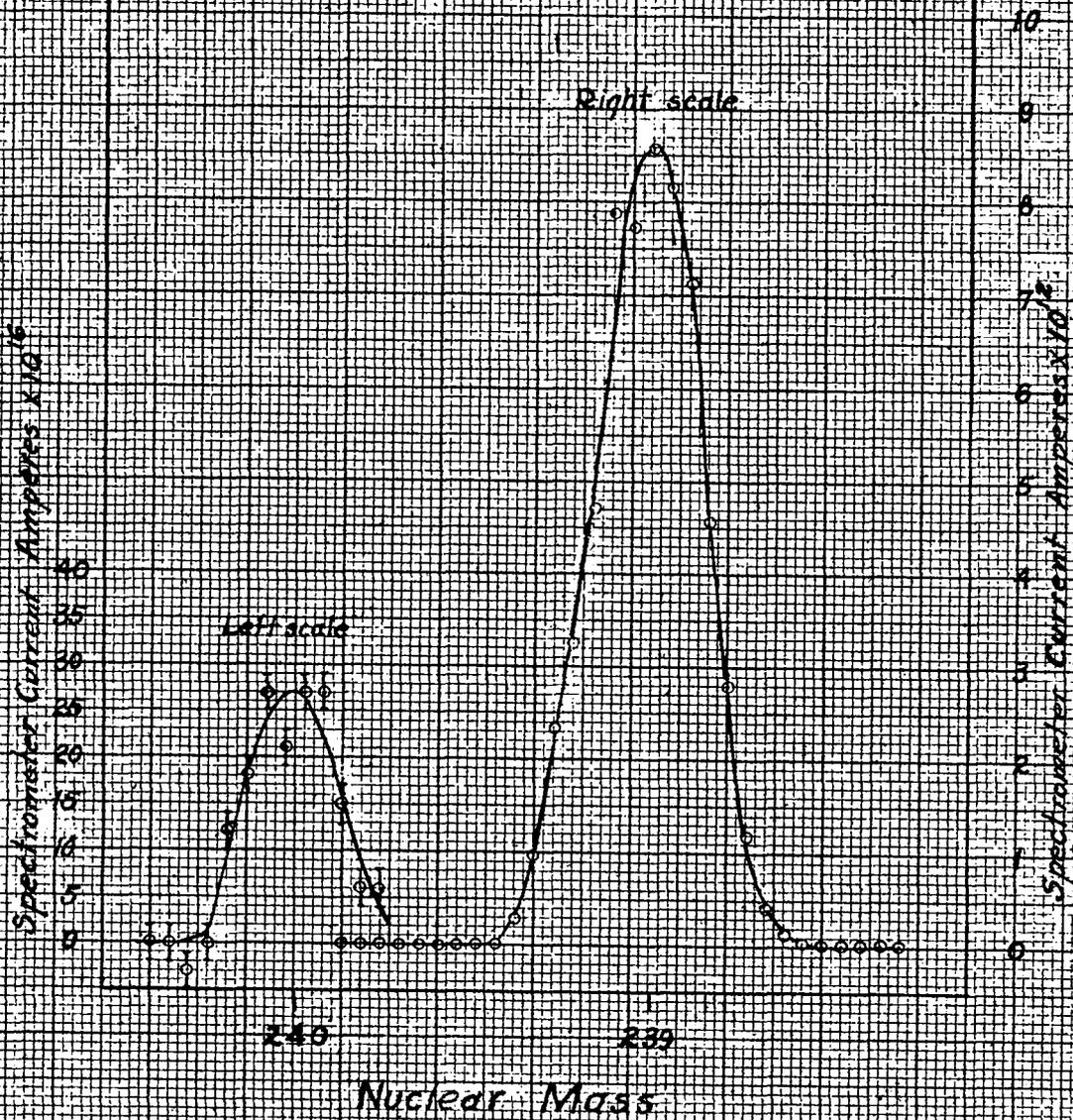
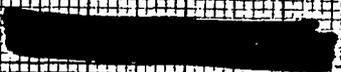


FIG 4  
Mass-Spectrum of Plutonium

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lier used. It is, nevertheless, reasonably certain that such an isotope exists; particularly since during the course of the short run, the currents were increased until the 240 peak was six times more intense in current than illustrated in Fig. 4. From several measurements on the ratio of currents at the 240 and 239 mass positions, we conclude that  $94^{240}$  is present to  $3.3 \pm 0.5$  parts in  $10^4$ .

During the publication of this report, data from X giving the grams of product per ton for the components of Lot 209-P were received. These data can be used to calculate the amount of 410 in a given batch of 49 by means of a formula provided by Fermi:

$$q/p = 45 \times (4/3)(\bar{p}/\bar{u}),$$

where

q = grams 410 formed in u

p = grams 49 formed in u

u = grams 28

The data and calculation are presented in Table II:

TABLE II

Lot No. 209-P

X Lot No.	( $\bar{p}/\bar{u}$ ) gms/ton	(q/p) in ppm	Wt factor
34-C	4.714	312	$\times 1.271 = 396$
34-D	4.714	312	$\times 3.760 = 1173$
26-A	2.458	163	$\times 1.503 = 244$
36-A	4.511	298	$\times \frac{1.638}{8.172} = \frac{488}{2301}$

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$$2301/8.172 = 282 \text{ ppm}$$

The agreement with our figure,  $330 \pm 50$  ppm, is good. The agreement within our limit of error is fortuitous.

The following may be said as to the reliability of our observation of  $Cl_2^{240}$ :

1) The spectrometer was operated at a vacuum of the order of  $1 \times 10^{-7}$  mm of mercury. To attain a vacuum of this order of magnitude in an instrument of this kind, prolonged baking and pumping at high temperatures are necessary. No grease or rubber-gasket joints can be used. On the basis of the general behavior of such instruments, it is very unlikely that heavy molecules present as residual background in the apparatus could have been responsible for this peak. Heavy molecule background was not found to be present.

2) The peaks at mass positions 239 and 240 were both found to be very temperature-sensitive. The variation with temperature was of such nature that the ratio of the intensities of the two lines was independent of temperature and time over the period of observation, which was, unfortunately, rather brief.

3) It seems unlikely that the peak at 240 is a "ghost", since as far as can be ascertained from the data, it is of the anticipated shape and occurs at the anticipated mass position. No such "ghosts" appear in either the mercury or tin spectra, although searches were made for them.

It should be stressed that this result is a preliminary one, particularly since the following additional checks on the reality of the 240 peak will be made in the future:

1) The pressure of water vapor in the system can be drastically reduced through the use of a liquid air on the cold trap. If the peak at 240 were caused by a hydride, the ratio of its intensity to the intensity of the 239 peak should be affected.

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2) More accurate measurements of the intensity of the  $^{240}$  peak over a longer period of time and a wider range of temperatures will eliminate the uncertainty mentioned in (2) above.

3) In principle, perhaps the most exhaustive test of the chemical identity of the two peaks observed is that their "appearance potentials" be the same. As the Nier spectrometer is not designed to make measurements of "appearance potential", this test will probably not be made unless it is pointed out to us that it is necessary.

Unfortunately the operation of the spectrometer in this first run was not as simple as desired. The evaporation of 49 from the lips of the micro-furnace resulted in production of metallic films on many of the glass insulators in the ion source. For this reason, it will be necessary to introduce much more complete insulator shields, and although this will be simple to do, the decontamination of the ion source prior to machining of the parts will take some time. Because of this leakage, which developed very quickly, it was not possible to make an exhaustive examination of the surrounding mass positions. We can, however, set an upper limit on the amount of  $^{241}$  present as one part in  $10^4$ .

#### IV. CONCLUSION

On the basis of the results to date it may be said that:

1) A mass spectrometer has been set up which can be used to study the isotopic constitution of element 94. For the present, sensitivity for  $^{240}_{94}$  is of the order of two parts in  $10^5$ . It is quite possible that this sensitivity can be increased by subsequent research.

2) Evidence has been obtained for the existence of the isotope  $^{240}_{94}$ , and one particular sample of 49 metal has been given a preliminary analysis.

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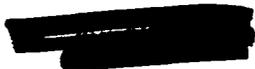
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3) It should now be possible to measure  $\alpha$  as a function of energy.

4) It is now possible to get an absolute value of  $\lambda_p$ .

5) It may become possible, by considerable development in the future, to separate a fraction of a microgram of relatively pure  $94^{240}$ . This is not possible, of course, until  $49$  containing at least several percent of  $410$  becomes available in gram quantities.

It should be emphasized that the present results are exceedingly preliminary and subject to revision at a later date. Subsequent results will be announced in subsequent reports in a series under this title.



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