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# A Mass-Spectrograph Study of Ba, Sr, In, Ga, Li, and Na

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A mass spectrograph of the 180° type of 22 cm radius has been constructed. The magnetic field of 1400 gauss is supplied by permanent magnets and the accelerating potential by a stabilized rectifier. A mass analysis has been made for a number of elements of the ions obtainable from coated filaments. A peak of Sr at mass 84 comprising 0.5 percent of the total Sr current, and one of Ba at mass 134 comprising 1.8 percent of the total Ba current are concluded to be new isotopes of Sr and Ba, respectively. A search for a third isotope of indium was made but none found. The relative abundances of $Ga^{69}$ and $Ga^{71}$ were determined to be 38.8 percent and 61.2 percent respectively. The existence of $Li^5$ and $Na^{22}$ recently reported by Brewer was not confirmed. Lower limits were placed on other possible isotopes of these elements.

## INTRODUCTION

At the present time all of the known stable elements have been subjected to analysis by the mass spectrograph. In many cases the spectra were obtained with difficulty and the results are correspondingly inaccurate. Recently Blewett and Jones[1] have succeeded in producing very satisfactory sources of ions from such elements as Ba, Sr, and In and it therefore seemed worth while to investigate the isotopic constitution of these elements in the hope of improving the existing data. Indium is especially interesting since it, like bromine displays three different rates of decay after activation by slow neutrons although only two stable isotopes have been found. A careful search for the stable isotopes $Na^{22}$ and $Li^5$ was also undertaken with the object of confirming the recent announcement of their discovery by Brewer.[2]

## APPARATUS AND PROCEDURE

A mass spectrograph of an unusual type was tried initially but proved unsuccessful. The essential idea was to use crossed electric and magnetic fields over a straight path length of 1 meter. The magnetic field was produced by permanent magnets and the electric field by a potential applied to parallel brass plates. The potentials applied to the condenser plates bore a constant ratio to the field accelerating the ions. The ions followed a trochoidal path, a focus being obtained for ions of a given $e/m$ having the same velocity and entering the analyzer in a nearly parallel beam. Very good resolution was obtained with this arrangement but the peaks were often unsymmetrical or split into several components. The collector current was very unsteady being subject to violent fluctuations. The difficulties were ascribed in part to reflections of the ion beam from the long condenser plates and to wax vapor contaminations causing the surfaces of the plates to charge up to varying potentials. The fluctuation of the current seemed to be related to the intensity of the ion beam striking the deflecting plates. After considerable work with this arrangement it was decided to give it up in favor of the apparatus about to be described.

A mass spectrograph of the 180° focusing type was then constructed with the object of obtaining moderately high resolving power. The magnetic field is supplied by permanent magnets and the accelerating fields are obtained from a stabilized rectifier circuit operating on a.c., and giving up to four thousand volts, constant to a fraction of a volt.

The arrangement of the analyzing chamber is shown in Fig. 1. The positions of the ion source and the collector are indicated in the diagram. The semicircular pole pieces *A* are pure soft iron, 5 cm wide, 2 cm thick with a mean radius of curvature of 22 cm. The gap between the pole pieces is 9 mm and was made vacuum tight by waxing the two brass strips *B* which are inset into the two sides of the pole pieces as shown. U-shaped permanent magnets of cobalt steel, made by the Weston Electrical Instrument Corporation, were spaced as closely as possible on the inside of the circle, and the spacing of the outside magnets was arranged to give as nearly

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[1] Blewett and Jones, Washington Meeting, Phys. Rev. **49**, 881 (1936).

[2] Brewer, Phys. Rev. **49**, 635, 856 (1936).

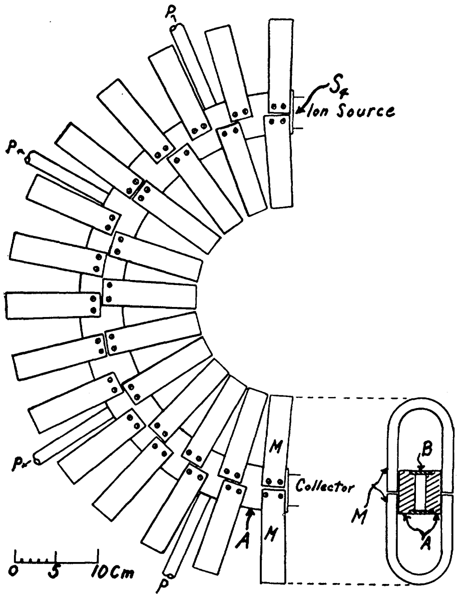

FIG. 1. Magnetic analyzing chamber.

uniform flux density as possible over the pole pieces. The field obtained in the gap was about 1400 gauss and was uniform insofar as could be ascertained by rough measurements with a search coil. In the case of lithium a third of the magnets were removed in order to bring the ions into the voltage range available. Sharp peaks were still observed with the wider spacing of the magnets. Three diaphragms were placed at equal intervals in the analyzing chamber in order to limit the angular spread of the ion beam to 2°. The central diaphragm was made adjustable from the outside and could be used to define the beam still more closely. The chamber was evacuated by two mercury pumps connected to the four pump leads $P$. The mercury vapor was trapped out with liquid air. A separate pump was connected to the tube containing the filament and accelerating slit assembly. The waxed joints were all made with Apiezon-W wax and the pressure reading on an ionization gauge was always between 1 and 2 times $10^{-6}$ mm of Hg.

Fig. 2 shows the arrangement of the ion source and the manner in which the output of the voltage regulator circuit is applied to the accelerating slits. The three slits $S_1$, $S_2$, $S_3$, and the two pairs of deflecting plates $D_1$ and $D_2$ were fastened to tungsten leads which were sealed into a Pyrex tube, the unshielded parts of which were coated with Aquadag and grounded to $S_1$. The filament was mounted on a ground glass joint and could be quickly removed for recoating. The filament $F$ was of platinum foil 2 mm wide by 5 mm long, placed across the slit $S_1$ to make the voltage distribution of the ions as small as possible. $G$ is a guard ring placed in the plane of the filament and connected to one side of it. It was found that the use of this guard ring increased the fraction of the total current that passed through $S_1$ considerably. The slits $S_1$, $S_2$, and $S_3$ were 1.5 mm by 1 cm. The two pairs of deflecting plates $D_1$ and $D_2$ were 1.5 cm apart and had the correct voltage applied to them to counteract the stray magnetic field at the end of the magnets. These two sets of deflecting plates permit the adjustment of both the position and direction of the ion beam striking the slit $S_4$.

The primary circuit of the transformer supplying the full wave rectifier contained an Acme-Delta voltage regulator to help stabilize line fluctuations. The rectified current from the two R.C.A. 879 tubes was passed through a voltage regulating circuit of the type described by Evans.[3] In this case however a 2A5 tube was used with a plate current of from 1 to 4 milliamperes. This was sufficiently large to prevent the very small fluctuations of the positive ion current (i.e., the load) from producing a perceptible change in the voltage. Large voltage changes

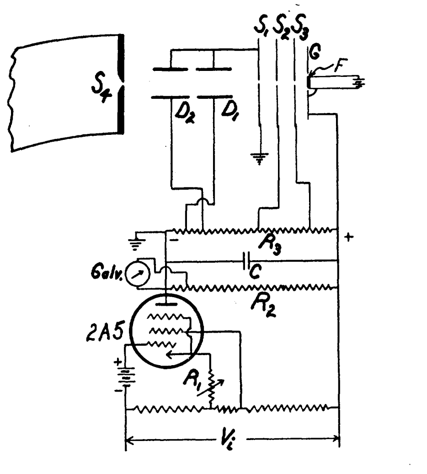

FIG. 2. Diagram showing arrangement of ion gun and connection to the voltage regulator. $V_i$ is the filtered output of the rectifier unit.

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[3] R. D. Evans, Rev. Sci. Inst. 5, 371 (1934).

were obtained by changing the taps on $R_3$ while continuous changes were made by varying $R_1$. The changes in the total accelerating voltage were measured with a galvanometer connected across a small section of the ten megohm wire wound resistor $R_2$. Considerable trouble was experienced with high frequency oscillations in the output of the regulator circuit. Placing a capacity of 2.5 microfarads across the resistor $R_3$ served to eliminate this difficulty.

The filament source of positive ions was heated by a storage battery and the total emission was generally a few tenths of a microampere. The voltages applied to the accelerating slits, obtained from taps on the resistor $R_3$, were such as to focus the ions into a parallel beam. The potentials for the deflecting plates were taken from the same voltage divider so that a constant ratio to the accelerating voltage was obtained. This arrangement is desired in order that ions emerging through the final slit shall have traversed the same path regardless of the $e/m$ selected.[4]

The entrance and collector slits were made of phosphor-bronze, a nonmagnetic material, and the edges were beveled to prevent reflections of the ions. The entrance slit $S_4$ was set at 0.1 mm, and the collector slit at 0.2 mm. The collector was a Faraday cage connected to the grid of a Western Electric D-96475 electrometer tube. The amplifier circuit was that described by Penick,[5] and was used at a sensitivity of 2 times $10^{-17}$ ampere per millimeter. In the curves shown the positive ion currents to the electrometer tube are plotted against accelerating voltage. The peaks were so sharp that only a few points could be taken on the sides of them. Because of the width of the collector slit with respect to the entrance slit the tops of the peaks were slightly flat and quite accurate readings were obtained for relative abundance measurements as long as the filament emission was low enough to prevent space charge limitation of the positive ion current.

## RESULTS

### Strontium and barium

Figs. 3 and 4 show the mass-spectrograph plots of the strontium and barium isotopes. The ions were produced by the method described by Blewett and Jones.[1] The peaks at mass numbers 84 and 134 for strontium and barium, respectively, are evidence for new isotopes of these elements.[6] The peaks are quite symmetrical and well resolved. The widths of the peaks at the base are probably due to the velocity distribution of the ions and to slight inhomogeneities in the magnetic field due to the gaps in the spacing of the magnets on the pole shoes. These new peaks cannot be attributed to hydrides since they fall on the low mass side of the known isotopes. Neither can they be satisfactorily explained as impurities since the only isotopes in this region belong to krypton and zenon. The heights of the new peaks relative to the element in question were constant under all conditions. It is therefore concluded that they represent true isotopes of strontium and barium respectively. The abundances found for the strontium isotopes are tabulated below together with the upper limits placed on the presence of other possible isotopes.

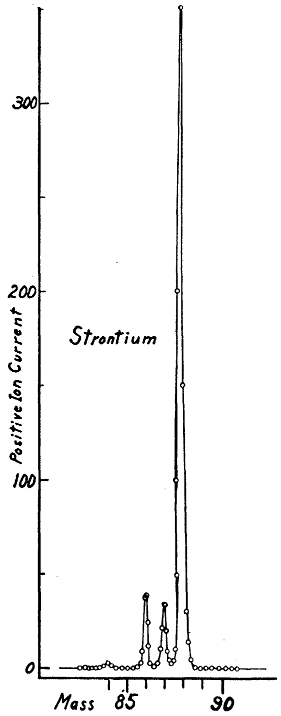

FIG. 3. Mass-spectrum plot of Sr.

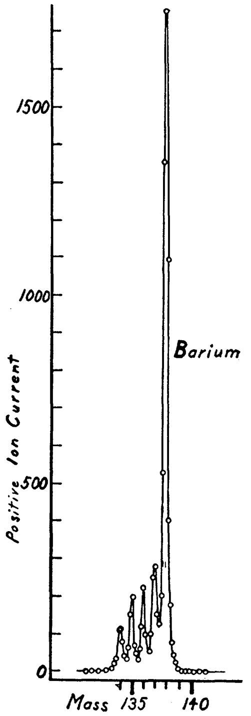

FIG. 4. Mass spectrum of Ba.

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[4] Bleakney, Am. Phys. Teacher **4**, 12 (1936).

[5] D. B. Penick, Rev. Sci. Inst. **6**, 115 (1935).

[6] Blewett and Sampson, Phys. Rev. **49**, 778 (1936).

| Mass number | 82 | 83 | 84 | 85 | 86 | 87 | 88 | 89 | 90 |
|---|---|---|---|---|---|---|---|---|---|
| Percentage | <0.05 | <0.1 | 0.5 | <0.05 | 9.6 | 7.5 | 82.4 | <0.2 | <0.05 |

Assuming a packing fraction of −8.2 the atomic weight for strontium comes out to be 87.62 which is to be compared with the chemical value 87.63. The peak at mass 134 comprises 1.8 percent of the total barium ion current. Our relative abundance measurements on barium were not very accurate but on combining our results with Aston's and taking a packing fraction of −6.1 we get 137.36 for the atomic weight in exact agreement with the chemical value. The limits placed on other possible barium isotopes are given below.§ The percentages given are the upper limits set on the existence of other possible isotopes.

| Mass number | 132 | 133 | 139 | 140 |
|---|---|---|---|---|
| Percentage | <0.05 | <0.1 | <0.25 | <0.1 |

## Indium

Fig. 5 shows the curve for indium which has also been previously reported.[6] A very careful search was made for a possible new isotope since its existence is indicated by the observation of three radioactive periods of decay, induced by bombardment with neutrons.[7] Only two of these periods have been definitely shown to be sensitive to slow neutrons. The abundance of the two known isotopes was found to be 21±1 for the ratio of 115 to 113. The limits placed on other possible indium isotopes are as follows.

| Mass number | 110 | 111 | 112 | 114 | 116 | 117 | 118 | 119 |
|---|---|---|---|---|---|---|---|---|
| Percentage | <0.01 | <0.01 | <0.02 | <0.5 | <0.02 | <0.02 | <0.012 | <0.003 |

## Gallium

Gallium ions were obtained by coating a platinized tungsten spiral with a water paste of gallium sesqui-oxide.[1] Large peaks of sodium and potassium ions were at first observed but on heating the filament to about 1800°C for some time they disappeared and gallium ion peaks at masses 69 and 71 were observed. The ion intensity however was too small to allow any search for new isotopes. The percentage abundances of the two known isotopes were found to be 38.8 percent and 61.2 percent, respectively, with an error of ±1 percent. These values are in agreement with Aston.[8]

## Lithium

Brewer[2] has recently reported evidence for the existence of a lithium isotope of mass 5. An attempt has been made to check his results. Fig. 6 shows the curve obtained for lithium. The peak at mass 7 is 100,000 cm high. The background becomes zero at a small distance from the mass 6 peak as shown in the magnified portion of the curve. A peak 1 cm high should have been observed had it been present so it may be concluded from this experiment that lithium 5 is present to less than 1 part in 100,000 of lithium 7. This is thought to be a conservative estimate. Dr. Blewett of this laboratory has also attempted to find $Li^5$ with an apparatus very similar in size and arrangement to that used by Brewer. Although his resolving power and background were not as favorable as ours, his total intensity was much greater. He believes one part in 200,000 would have been detectable but could find no trace of $Li^5$. We are indebted to Dr. Blewett for allowing us to mention his results in this connection.

## Sodium

Brewer[2] has also reported the existence of an isotope of sodium of mass 22 present to 1 part in 5000 of sodium 23. The possibility of a stable sodium isotope of mass 22 is particularly interesting since it has generally been thought to be unstable, disintegrating with emission of a positron. Meitner[9] and Frisch[10] have obtained evidence for this positron emission. Laslett[11] has also obtained a positron emitter by a different disintegration process, which he attributes to sodium 22. He has determined the half-life to be about 9 months.

Fig. 7 shows one of the curves obtained in a careful search for $Na^{22}$. There is a small back-

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§ *Note added in proof:* After this paper had gone to press a note by Dempster, Phys. Rev. **49**, 947 (1936) appeared in which he confirmed the existence of $Ba^{134}$ and reported two new isotopes at masses 132 and 130. Using a new type of source we also find these new isotopes and have succeeded in measuring their abundances in the third and fourth orders giving the following results:

| Mass number | 130 | 132 | 134 | 135 | 136 | 137 | 138 |
|---|---|---|---|---|---|---|---|
| Percentage | 0.16 | 0.015 | 1.72 | 5.7 | 8.5 | 10.8 | 73.1 |

The atomic weight calculated from these abundances comes out to be 136.35.

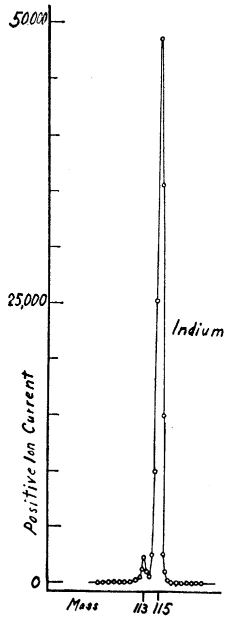
[7] Szilard and Chalmers, Nature **135**, 98 (1935).

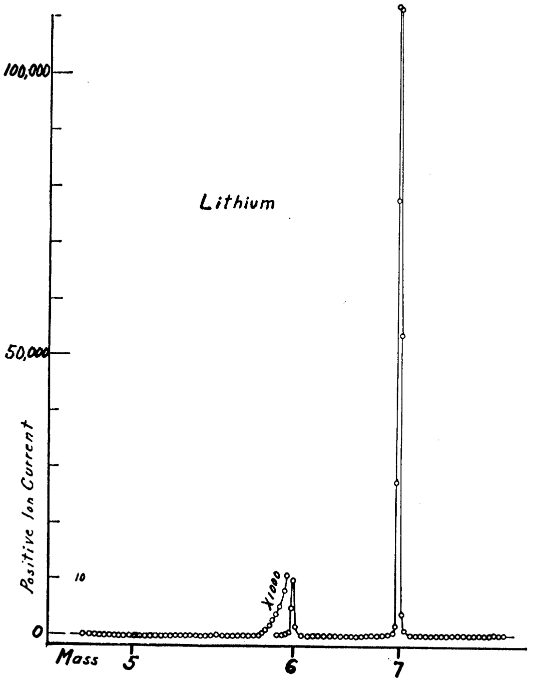
[8] Aston, Proc. Roy. Soc. **A149**, 396 (1935).

[9] Meitner, Naturwiss. 22, 420 (1934).

[10] Frisch, Nature **136**, 220 (1935).

[11] Laslett, Seattle Meeting, Am. Phys. Soc. June, 1936, paper No. 22.





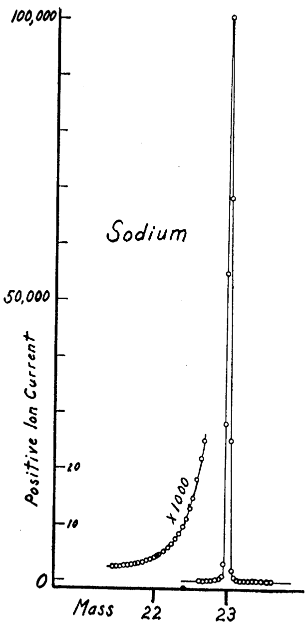


FIG. 5 (left). Mass spectrum of In.
FIG. 6 (center). Mass spectrum of Li. Curve in region of mass 5 is magnified 1000 times.
FIG. 7 (right). Mass spectrum plot of Na. Region of mass 22 magnified 1000 times.

ground of about 2 cm at the mass 22 position. A peak of 2 cm above the background could easily have been detected and it is concluded that $Na^{22}$ is present to less than 1 part in 50,000 of sodium 23. The fact that our apparatus was working properly is indicated by an analysis of potassium which showed the isotope at mass 40, recently reported by Nier,[12] to be clearly resolved and of the right abundance, as measured by Nier[12] and Brewer,[13] in spite of the much higher background than that in the case of either lithium or sodium.

It seems impossible to reconcile our results with those of Brewer unless $Li^5$ and $Na^{22}$ are present to a much smaller degree than reported, or that the peaks he observes are spurious, possibly due to a reflection of some sort. Spurious peaks have very often been found with our mass spectrograph but careful adjustment, and proper shielding and alignment have served to eliminate them. Moreover while many peaks have been found to be only ghosts, in no case have we been able, by improper adjustment, to make a true isotope disappear from the mass spectrum.

We are indebted to Mr. Briggs E. Napier for his help in constructing the rectifier. It is a pleasure also to acknowledge the many helpful discussions and the assistance of Dr. A. Bramley during part of this work. This research was aided by a grant from the American Philosophical Society for which we express our appreciation.

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[12] A. O. Nier, Phys. Rev. **48**, 283 (1935).

[13] Brewer, Phys. Rev. **48**, 640 (1935).