therefore that a considerable number of the $\sim 120^{\circ}$ K neutrons should have energies greater than 0.023 ev, which is the point at which $0\rightarrow 1$ scattering begins to take place and the *para* cross section begins to increase rapidly. It seems probable therefore that the ratio of *para* cross sections for 0.037 and 0.012 ev neutrons is actually much larger than 29.0/17.6. A layer of parahydrogen of suitable thickness should therefore transmit neutrons in the low energy range of a Maxwell distribution, and cut off rather sharply at an energy of 0.023 ev. Such a low energy beam should be quite useful in experimental work.

In the actual transmission measurements and calculations of cross sections, the chief errors involved are in cell thickness, composition of the hydrogen, and statistical fluctuations of the counts. Of these the largest is probably in the determination of cell thicknesses. Although this will affect the absolute values of the cross sections it will have very little influence on the relative values for different ortho-para compositions. Boiling may reduce the effective thickness of material in a cell. An estimate of the rate of heat transfer to the cells by radiation indicated that the amount of gas produced by evaporation in one second would not fill more than 2 percent of the cell volumes. We have observed in other experiments that the bubbles in liquid hydrogen are small and rise rapidly. Errors due to the presence of bubbles should be of the order of 1 percent. Uncertainty in the measurement of the composition is 1 percent. The number of counts was sufficient to make the probable error due to statistical fluctuations less than 2 percent. The combined error due to all the above factors certainly cannot be greater than 10 percent.

The results of the measurements on liquid N₂, O₂, and H₂O are in substantial agreement with previous values.^{8, 9, 27} Log transmission has been plotted against scattering thickness in Fig. 4. In obtaining proton cross sections from the water measurements the value 4.0×10^{-24} cm² for the cross section of oxygen was taken from Table II. The same value was used for both the ~300°K and ~120°K neutrons since no information on the temperature dependence was available. The ratio of the proton cross sections in water thus obtained is $\sigma_{120}/\sigma_{300}=1.29$. Errors in these measurements are substantially the same as in the hydrogen measurements.

The authors wish to express their appreciation of the cooperation of Director Lyman J. Briggs of the National Bureau of Standards and Dean George B. Pegram of Columbia University which made these experiments possible. They are also indebted to Professor Edward Teller of the George Washington University and to Mr. Julian Schwinger of Columbia University for valuable discussion and advice.

²⁷ M. Goldhaber and G. H. Briggs, Proc. Roy. Soc. A162, 127 (1937).

AUGUST 15, 1938

PHYSICAL REVIEW

VOLUME 5

The Isotopic Constitution of Strontium, Barium, Bismuth, Thallium and Mercury

Alfred O. Nier*

Research Laboratory of Physics, Harvard University, Cambridge, Massachusetts (Received June 18, 1938)

A mass-spectrographic study of the isotopic constitution of Sr, Ba, Bi, and Tl has been made. Although the sensitivity and resolving power of the apparatus were sufficient to detect extremely rare isotopes, no new ones were found. Relative abundances of the isotopes of Sr, Ba, and Tl were determined. It was possible to show that if Hg^{203} exists it must be with an abundance less than one-tenth of that at one time assigned to it by Aston.

A LTHOUGH a knowledge of the existence or nonexistence of rare stable isotopes is of great value in the study of nuclear physics, * National Research Fellow. relatively few elements have been investigated with apparatus capable of detecting isotopes present in only small amounts. The present paper is a report on the continuation of a systematic program, the aim of which is to establish the existence or nonexistence of possible rare isotopes, as well as to determine the relative abundances of the accepted stable ones. Because of the extremely high sensitivity and resolving power of the apparatus it is possible to extend the search for rare isotopes beyond previously attainable limits.

Apparatus

A description of the mass spectrometer employed has already been given.¹ It may be described briefly as an apparatus in which positive ions are formed by the collision of a controlled beam of low energy electrons with the gas or vapor of the elements to be studied. The positive ions are drawn from the ionizing region by suitable electric fields and the m/e analysis is accomplished by means of a 180° magnetic analyzer. Inasmuch as the analyzed ion current is measured by means of an electrometer tube it is relatively easy to obtain accurate determinations of relative abundances. Because of the fact that the metal parts of the mass spectrometer tube are constructed entirely of Nichrome, copper, and tungsten, and are surrounded by a Pyrex envelope without grease or wax joints, the tube may be thoroughly baked, and hence, little difficulty is encountered with impurities. In the study of relatively nonvolatile substances such as Sr, Ba, Bi, and Tl, the element is placed in a small Nichrome furnace inside of the spectrometer tube. The furnace may be heated to a temperature as high as 1200°C and the stream of vapor emitted passes into the electron beam where ions then are formed. For the investigation of volatile substances the furnace is, of course, unnecessary, and the gas or vapor can be admitted to the tube through a capillary leak or by some other means.

In the present work the slits shown in Fig. 1 of reference 1 had the following widths: $S_1 = 0.007''$, $S_2 = 0.006''$, and S_3 , the exit slit of the 180° analyzer (not shown in the figure) = 0.006'' for Hg, Tl, and Bi; 0.018" for Sr and Ba.

RESULTS

A clean sample of strontium metal obtained from Eimer and Amend, labeled 99.9 percent

¹ Nier, Phys. Rev. 53, 282 (1938).

Strontium



FIG. 1. Mass spectrum showing isotopes of strontium. The experimental points are obtained by plotting positive ion current passing through the exit slit of the analyzer as a function of the energy of the ions while the magnetic field is held constant. As there is a definite relation between the energy and the mass of the ions collected, namely, energy \times mass = a constant, the voltage scale can be readily changed to a mass scale, as was done in the figure. The width of the peaks is, of course, due to the finite width of the slits used and the imperfections in focusing.

pure, was placed in the small furnace, shown in Fig. 1 of reference 1. After thorough baking of the tube to remove impurities, work was begun. It was found that a furnace temperature of the order of 700°C gave sufficiently large positive ion currents without too great a rate of consumption of strontium. A typical mass spectrum is shown in Fig. 1. The isotopes observed were the same as those reported by other investigators.²⁻⁵ In Table I are given the relative abundances found in the present work as well as those given by Sampson and Bleakney.⁴ The results are seen to be in good agreement. The writer believes his relative abundances as measured for Sr⁸⁷ and Sr⁸⁶ relative to Sr⁸⁸ to be correct within one percent, while that given for Sr⁸⁴ may be in error by two percent. A possible source of error not taken into account in these measurements is the effect of the free evaporation of Sr atoms from the small furnace. In the case of *ideal* distillation this would mean that the Sr⁸⁴/Sr⁸⁸ ratio as measured would be $(88/84)^{\frac{1}{2}} = 1.023$ times greater than the true ratio. However, as the Sr atoms are evaporated from a solid surface

 ² Blewett and Sampson, Phys. Rev. 49, 778 (1936).
³ Dempster, Phys. Rev. 50, 186 (1936).
⁴ Sampson and Bleakney, Phys. Rev. 50, 456 (1936).
⁵ Mattauch, Akad. Wiss. Wien Ber. 145, 461 (1936).

and not from a liquid (where mixing of particles on the surface with those inside can take place) it does not seem likely that one would have appreciable discrimination.

If one assumes the packing fraction of strontium to be that found by Mattauch⁶ for Sr⁸⁶, namely, -9.0 ± 0.5 , and if one uses a conversion factor 1.000275 in going from the physical to the chemical scale, an atomic weight 85.61 is computed for strontium from the writer's as well as from Sampson and Bleakney's relative abundances. On the other hand, if one assumes the packing fraction of Sr to be the same as that found by Aston⁷ for Kr⁸⁶, namely -7.40, the atomic weight 85.62 is computed. From the packing fraction -6.3, deduced from Dempster's⁸ packing fraction curve, one obtains the atomic weight 85.63. The chemically determined value is accepted as 85.63.

The real reason for studying strontium was for the purpose of searching for possible rare isotopes. By analogy to most of the heavier even elements one might expect to find strontium to consist of a sequence of isotopes having successive mass numbers flanked by a light isotope two units removed from the lightest isotope in the sequence and a heavier isotope two units above the heaviest isotope in the sequence. Thus, on this basis, one should expect to find a stable isotope, Sr⁹⁰. However, a careful investigation of this position as well as the other vacant positions around the known stable isotopes failed to show even the faintest trace of any new isotope. It was possible to set the following upper limits for abundances in these positions relative to Sr⁸⁸: Sr⁹², Sr⁹¹, and Sr⁹⁰, 1/300,000; Sr^{89} , 1/100,000; Sr^{85} , 1/50,000; Sr^{83} , 1/100,000; Sr⁸², Sr⁸¹, and Sr⁸⁰, 1/200,000.

Barium

Barium metal obtained from Eimer and Amend, labeled 99 percent pure, was employed

TABLE I. Relative abundances of isotopes of strontium.

	88	87	86	84	Mean Mass Number
Abundance	100	8.50	11.94	0.68	87.710
S. & B. values	100	9.10	11.64	0.61	87.713

⁶ Mattauch, Naturwiss. 25, 170 (1937).

⁷ Aston, Nature **140**, 149 (1937).

⁸ Dempster, Phys. Rev. 53, 869 (1938).

as a source of barium. The procedure was the same as that used for strontium. Fig. 2 shows a typical mass spectrum obtained. An idea of the sensitivity of the apparatus is gained when one considers that the points for the rare isotopes, Ba¹³² and Ba¹³⁰, are plotted to a scale 100 times that for the other isotopes. The isotopes found are the same as those reported by Dempster⁹ and Sampson and Bleakney.⁴ Table II gives the relative abundances found in the present work,



FIG. 2. Mass spectrum showing the isotopes of barium. Current scale in region of Ba¹³² and Ba¹³⁰ is multiplied by a factor of one hundred.

together with the abundances as given by Sampson and Bleakney. The writer believes his abundances relative to Ba¹³⁸ all to be correct within two percent except the ones for Ba¹³² and Ba¹³⁰, which may be in error by four percent. As in strontium the evaporation of the atoms takes places from the surface of a solid, and hence, one should expect little, if any, separation to take place in the evaporation process.

If one assumes the packing fraction of barium to be the same as that given recently by Aston⁷ for Xe¹³², -4.4, and if one uses a conversion factor 1.000275 in going from the physical to the chemical scale, an atomic weight 137.33 is obtained from the writer's data. With a packing fraction -4.0, as deduced from Dempster's⁸ curve, the value 137.33 is also found. The chemically determined value is given as 137.36.

As in the case of strontium, the main purpose in investigating barium was to search for possible

⁹ Dempster, Phys. Rev. 49, 947 (1936).

TABLE II. Relative abundances of isotopes of barium.

	138	137	136	135	134	132	130	Mean Mass No.
Abundance	100	15.8	10.9	9.2	3.37	0.136	0.141	$137.42 \\ 137.47$
S. & B. values	100	14.9	11.7	7.8	2.35	0.022	0.22	

rare isotopes. However, none was found, and it was possible to set the following upper limits for abundances of hypothetical isotopes relative to Ba¹³⁸: Ba¹⁴², 1/200,000; Ba¹⁴¹ and Ba¹⁴⁰, 1/100,000; Ba¹³⁹, 1/35,000; Ba¹³³, Ba¹³¹, Ba¹²⁹, and Ba¹²⁸, 1/100,000.

Bismuth

Aston¹⁰ found bismuth to consist of a single isotope of mass 209. This fact was verified by Bainbridge and Jordan¹¹ who showed that no other isotope existed with an abundance greater than two percent. From the present study one can set the following upper limits of abundance for the existence of hypothetical isotopes relative to Bi209: Bi213, Bi212, and Bi211, 1/100,000; Bi210 and Bi208, 1/50,000; Bi207, Bi206, and Bi205, 1/100,000. Bismuth is thus seen to be single to a very high degree, indeed.

Thallium

Thallium, an element having an odd atomic number, was found by Schüler and Keyston¹² to consist of two isotopes having the masses 203 and 205. The abundance ratio 203/205 was given as 0.435. The existence of these two isotopes was verified by Aston.¹³ As it appears to be a general rule that odd elements do not have more than two stable isotopes it did not seem likely that any additional ones would be found. However, very few elements have been investigated with apparatus capable of detecting truly rare isotopes, and hence a study of thallium is of interest if for no other reason than to better establish the validity of the rule.

Chemically pure thallium metal, obtained from Eimer and Amend, was used as a source of thallium. The 203/205 ratio was found to be 0.410 ± 2 percent, a number in excellent agreement with the ratio 0.417 found by Aston. As the evaporation of the metal takes place from the liquid phase the abundance ratio as measured may be high by a factor, $(205/203)^{\frac{1}{2}} = 1.005$, due to the effect of ideal distillation. Account was taken of this factor in arriving at the number 0.410.

The abundance ratio found corresponds to a mean mass number 204.42 ± 0.01 . With a packing fraction of +2.0 derived from Dempster's¹⁴ data and a conversion factor 1.000275 in going from the physical to the chemical scale, an atomic weight 204.40 is computed from the writer's data. This is to be compared with a chemically determined value 204.39.

No new isotopes were found, and it was possible to set the following upper limits of abundance for the existence of possible rare ones relative to T1205: T1209, T1208, T1207, and T1206, 1/50,000; Tl²⁰⁴ and Tl²⁰², 1/30,000; Tl²⁰¹, Tl²⁰⁰, and Tl¹⁹⁹, 1/50,000.

Mercury

Hg²⁰³ was at one time reported by Aston¹⁵ to exist as a stable isotope with an abundance approximately 1/5000 that of Hg²⁰². Recently the writer¹⁶ undertook a search for this isotope, but because of limited resolving power was able to conclude only that if it existed at all it must be with an abundance less than that assigned to it by Aston. In the present work on Bi and Tl the resolving power of the apparatus was somewhat higher than was the case in the earlier work, mainly because of the narrower slits used. With a pressure of around 10^{-5} mm of Hg in the tube. the sensitivity was sufficient to show definitely that if Hg²⁰³ existed it must be with an abundance less than 1/50,000 that of Hg²⁰².

¹⁰ Aston, Phil. Mag. **49**, 1197 (1925). ¹¹ Bainbridge and Jordan, Phys. Rev. **50**, 282 (1936). ¹² Schüler and Keyston, Zeits. f. Physik **70**, 1–2, 1 (1931). ¹³ Astor, Proc. Proc. **A134**, 571 (1932).

¹³ Aston, Proc. Roy. Soc. A134, 571 (1932).

¹⁴ Dempster, Phys. Rev. 53, 64 (1938).

 ¹⁵ Aston, Nature 130, 847 (1932).
¹⁶ Nier, Phys. Rev. 52, 933 (1937).