is questionable if such a shape could be detected by the methods of scintillation spectrometry in a low-intensity transition.

The logft value was calculated to be 7.4 for the 1.87-Mev beta transition in Sb^{129}. This is in the range for first forbidden transitions with $\Delta I=0$ or ± 1 and a change in parity. The ground state of Sb^{129} has an assigned level of $g_{7/2}$ and the 34-day and 72-minute isomers of Te^{129} appear to occupy $h_{11/2}$ and $d_{3/2}$ levels respectively.[8] This would indicate that the 1.87-Mev beta ray of Sb^{129} does not go directly to either of these two levels, but proceeds through a higher excited level and a gamma transition to one or the other of the tellurium isomers.

Since the complete disintegration schemes of these isotopes could not be obtained without many additional measurements, it is not possible at this time to make any definite proposals about the nature of the lower-energy beta transitions or to assign spins to the various levels involved in the complex decay processes.

PHYSICAL REVIEW VOLUME 101, NUMBER 6 MARCH 15, 1956

Search for Possible Naturally Occurring Isotopes of Low Abundance

F. A. White, T. L. Collins, and F. M. Rourke
Knolls Atomic Power Laboratory, Schenectady, New York*

(Received July 8, 1955; revised manuscript received December 27, 1955)

A systematic search has been made for the existence of previously unobserved naturally occurring isotopes of low abundance. A two-stage mass analyzer was employed. With the exception of Ta^{180}, none were found. The improved abundance sensitivity of the two-stage spectrometer, however, allowed determinations of lower experimental "upper limits" to be placed on a large number of isotopes. The elements, sodium, aluminum, scandium, niobium, and cesium were confirmed to be monoisotopic. A comparison has also been made of abundance measurements obtained with this instrument to those obtained in previous investigations.

INTRODUCTION

THERE are at least two reasons for continued interest in isotopic abundance measurements of the naturally occurring elements. First, certain hypotheses concerning nuclear structure can be tested for consistency relative to the experimentally determined isotopic constitution of matter. Second, and of increasing importance, is the interest in measuring the radiative capture of many elements for various neutron spectra. In many instances, it is desirable to measure the buildup of a very small number of nuclei. It is thus necessary that very low experimental "upper limits" be placed on the existence of these nuclei occurring in nature.

Accordingly, we have made such measurements as could be readily made on stable isotopes with the instrumentation which has been developed for nuclear work in this laboratory during the past year.

APPARATUS

The instrument used for this survey was a two-stage mass spectrometer with two 90-degree, 12-inch radius of curvature magnetic sectors arranged in tandem. It is important to note that for such an instrument, abundance sensitivity, rather than resolution alone, is a figure of merit. The abundance sensitivity is defined as the reciprocal of the fractional number of ions of mass M which arrive and are detected at the $M+1$ mass position in the image focal plane. In order for an instrument to have a high abundance sensitivity, e.g., 10^6, it is of course necessary for the detector to have a range of 10^6.

A unique feature of this instrument is that only particles of a single mass at a time are allowed to pass through an intermediate slit, thus minimizing the effect of small-angle elastic scattering within the analyzer tube. For an intermediate slit which is small compared to the dispersion, the fractional number F of ions of mass M, which are scattered to the mass $M+1$ position for a single analyzer, is reduced to F^2. Details of this instrument, which is diagrammatically shown in Fig. 1, are discussed elsewhere.[1]

It was necessary, for some of the measurements reported here, to observe exceedingly small beam currents due to limited positive ion emission. A twenty-stage Allen-type multiplier was employed, not as a dc amplifier, but as a counter[2] of discrete positive ions. A normal background counting rate, due to thermionic electrons emitted from the first dynode only, corresponds to about 5×10^{-20} ampere. Currents in the 10^{-19} ampere range were thus observable.

SAMPLE PREPARATION AND ION PRODUCTION

In an investigation of this type where the primary purpose is to determine new limits on isotopic ex-

---

* Operated for the U. S. Atomic Energy Commission by the General Electric Company.

[1] F. A. White and T. L. Collins, Appl. Spectroscopy 8, No. 4, 169 (1954).

[2] F. A. White and T. L. Collins, Appl. Spectroscopy 8, No. 1, (1954).

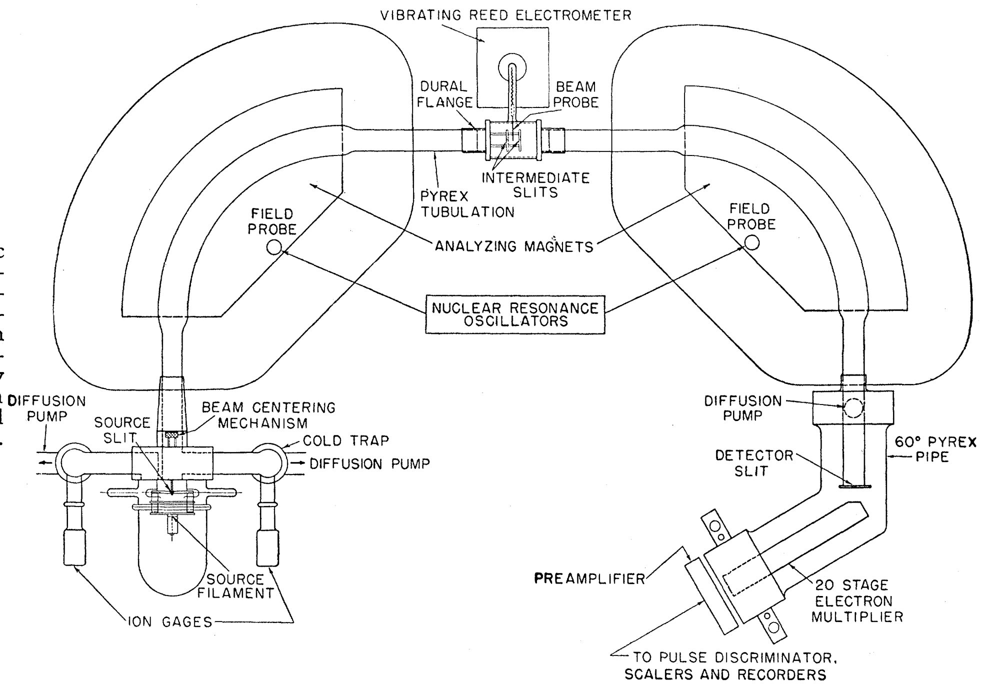

FIG. 1. Diagrammatic sketch of a two-stage magnetic analyzer utilizing 90-degree, 12-inch radius-of-curvature sectors in tandem. Individual positive ions are detected by a twenty-stage electron multiplier and associated high-speed scaling circuits.

istence, rather than precision ratio measurements, certain limitations are immediately apparent. These include positive ion current intensity of the desired element, interference from complex ions having nearly the same *e/m* ratio, superposition of ions of isobars, and range of detector sensitivity. In some instances, and actual filament impurity was utilized as the source of ions of the element, e.g., sodium, from a tungsten filament. No doubly charged ions were observed. In other cases of elements of high ionization potentials, even with large samples (milligrams), the production of suitably intense ion beams was exceedingly difficult.

A surface ionization source was used throughout and unless otherwise noted tungsten was used as a filament material. A 40-mil wide tungsten ribbon was folded in the shape of a *V* along its length and welded to 15-mil diameter supporting legs. This method of supporting prevented buckling at high temperatures. The *V* configuration allowed a fraction of the neutral atoms which were evaporated from the hot tungsten a second chance to become ionized before reaching the first slit of the ion gun. Beam maxima were obtained by adjusting the ion gun in azimuth through a vacuum-tight mechansim.

For the very large ratios which are reported, it was necessary to circumvent the limitation imposed by counting-rate losses of the electron multiplier and its associated high-speed scaling circuits. For positive ion counting rates up to 10^7 counts per minute ($\sim 10^{-14}$ ampere), corrections were made by observing changes in known isotopic ratios. Ratios of 1 to 10^8 were obtained by intermediate measurements with respect to the isotopes of lesser abundance of the given element.

The results of this survey are compiled in Table I together with those of other investigators.

Lithium

Previous experience at this laboratory, with the production of lithium ions by surface ionization, has indicated that the best results are obtained if the sample size is small. Accordingly, the total weight of sample used for this analysis was 10^{-7} gram of Li_2SO_4. Adequate lithium ion current was produced at 600-degrees Centigrade.

It was not the purpose of this analysis to measure the absolute abundance ratio of the known lithium isotopes, but rather to place upper limits on the possible existence of other isotopes of this element. The Li^7/Li^6 ratio measured in this investigation was 11.32. Upper limits obtained for the existence of Li^5 and Li^8 relative to Li^7 were: Li^5—1/1 500 000 of Li^7; Li^8—1/1 000 000 of Li^7.

Sodium

Sodium is present as an impurity in all filament materials. Because of its relatively high ionization efficiency, prolific beams of Na^+ ions were easily obtained from the filament itself. No sodium isotopes other than Na^{23} were detected. However, the presence of magnesium isotopes at mass 24, 25, and 26 was noticed and consequently no upper limits were obtained for the higher mass isotopes of sodium.

Magnesium

Magnesium ions were produced by thermal ionization of a magnesium nitrate sample. Only the Mg^+ ion was observed. The abundance values obtained in this investigation for the three known isotopes of Mg are given in Table I, together with the previous values obtained by White and Cameron.[3]

A search for other possible isotopes of magnesium showed that Mg^{22} could not exist in excess of 1/300 000 of Mg^{24} while Mg^{21} did not appear to exist in excess of 1/1 000 000 of Mg^{24}.

No other limits could be obtained because of the presence of impurity peaks adjacent to the magnesium spectra, notably Na^{23} and Al^{27}.

Aluminum

A coating of aluminum nitrate on a tungsten filament was used as a source of thermal ions. Beam currents of adequate intensity were observed even at temperatures as low as 900-degrees Centigrade. Al^{27} was found to be the only isotope.

Potassium

Potassium nitrate on a tungsten filament was used as a source of thermal ions. The K^+ ions were accelerated at 4 kilovolts. Voltage discrimination between K^{39} and K^{41} was found to be about 3 percent and the results reported here have been corrected for this effect. Table I also shows the previous values obtained by Nier[4] using electron bombardment of potassium vapor.

No new isotopes of potassium were found, but extremely low upper limits were placed on the possible existence of such isotopes.

Scandium

Scandium oxide was used as a source material. Sc^{45} appeared to be simple, since no other isotope was found

TABLE I. Isotopic abundances.

Element	Mass No.	Abundance This investigation	 Other observers
Li	5	<0.00007	<0.0005[a]
	8	<0.0001	<0.001[b]
Na	21	<0.000001	<0.002[c]
	22	<0.000003	<0.002[d]
	23	100.0	...
Mg	21	<0.00008	...
	22	<0.00025	...
	24	78.8±0.2	78.6±0.13[b]
	25	10.15±0.1	10.11±0.05[b]
	26	11.06±0.1	11.29±0.08[b]
Al	25	<0.00015	...
	26	<0.00015	...
	27	100.0	...
	28	<0.00005	...
	29	<0.00002	...
	30	<0.00005	...
K	36	<0.000008	...
	37	<0.000008	...
	38	<0.000008	..
	39	93.23±0.05	93.08±0.04[e]
	40	0.0118±0.0002	0.0119±0.0001[e]
	41	6.76±0.05	6.91±0.04[e]
	42	<0.000004	<0.00062[f]
	43	<0.000002	<0.00062[g]
Sc	42	<0.010	<0.0002[h]
	43	<0.010	<0.0002[h]
	44	<0.010	<0.0005[h]
	45	100.0	...
	46	<0.010	<0.002[h]
V	48	<0.0001	...
	49	<0.00005	...
	50	0.25±0.01	0.24±0.01[i]
	51	99.75±0.01	99.76±0.01[i]
Mn	53	<0.0004	<0.0066[j]
	54	<0.0003	...
	55	100.0	...
Nb	89	<0.00012	...
	90	<0.00012	...
	91	<0.00019	<0.25[j]
	92	<0.00019	...
	93	100.0	...
	94	<0.00019	...
	95	<0.00010	<0.025[k]
Nb	96	<0.00010	...
	97	<0.00010	...
	98	<0.00010	...
Ru	94	<0.010	...
	95	<0.010	...
	96	5.57±0.08	5.50[l]
	97	<0.010	...
	98	1.86±0.04	1.91[l]
	99	12.7±0.1	12.70[l]
	100	12.6±0.1	12.69[l]
	101	17.1±0.1	17.01[l]
	102	31.6±0.2	31.52[l]
	103	<0.0001	...
	104	18.5±0.1	18.67[l]
	105	<0.0007	...
	106	< 0.0010	...
In	108	<0.000002	...
	109	<0.000002	...
	110	<0.000002	<0.01[d]
	111	<0.000002	<0.01[d]
	112	<0.000004	<0.02[d]
	113	4.33±0.04	4.23±0.03[b]
	114	<0.00002	<0.5[d]
	115	95.67	95.77±0.03[b]
	116	<0.00001	<0.02[d]
	117	<0.000008	<0.02[d]
	118	<0.000008	<0.12[d]
Cs	130	<0.000002	<0.001[m]
	131	<0.000010	<0.005[m]
	132	<0.000030	<0.025[m]
	133	100.0	...
	134	<0.00005	<0.017[m]
	135	<0.00003	<0.002[m]
	136	<0.00001	<0.001[m]
La	134	<0.000007	(all other <0.002)[n]
	135	<0.000007	<0.002
	136	<0.000007	<0.002
	137	<0.000014	<0.002
	138	0.089±0.0015	0.089±0.002[n]
	139	99.911±0.0015	99.911
	140	<0.001	<0.002
	141	<0.002	<0.002
Nd	139	<0.0003	<0.02[o]

---

[3] J. R. White and A. E. Cameron, Phys. Rev. **74**, 991 (1948).
[4] A. O. C. Nier, Phys. Rev. **77**, 789 (1950).

TABLE I.—*Continued.*

Element	Mass No.	Abundance	
		This investigation	Other observers
Nd	140	<0.0001	<0.004[o]
	142	27.3±0.2	27.13±0.2[o]
	143	12.32±0.09	12.20±0.1[o]
	144	23.8±0.18	23.87±0.2[o]
	145	8.29±0.06	8.30±0.05[o]
	146	17.10±0.14	17.18±0.2[o]
	147	<0.0002	<0.01[o]
	148	5.67±0.05	5.72±0.06[o]
	149	<0.0001	<0.005[o]
	150	5.56±0.05	5.60±0.06[o]
	151	<0.0001	...
	152	<0.0002	...
	153	<0.0001	...
Hf	172	<0.00002	...
	173	<0.00002	...
	174	0.163±0.002	0.18±0.01[p]
	175	<0.00005	...
	176	5.21±0.02	5.15±0.02[p]
	177	18.56±0.06	18.39±0.01[p]
	178	27.10±0.10	27.08±0.04[p]
	179	13.75±0.05	13.78±0.02[p]
	180	35.22±0.10	35.44±0.06[p]
	181	<0.00003	...
	182	<0.00005	...
	183	<0.00005	...
	184	<0.00005	...
	185	<0.00002	...
Ta	177	<0.0003	(all other <0.01)[b]
	178	<0.0003	<0.01
	179	<0.0003	<0.01
	180	0.0123±0.0003	<0.01
	181	99.9877±0.0003	100.0[b]
	182	<0.0002	<0.01
	183	<0.0002	<0.01
Pt	188	<0.0013	...
	189	<0.0005	...
	190	0.0127±0.0005	0.012[q]
	191	<0.0005	...
	192	0.78±0.01	0.78[q]
	193	<0.001	...
	194	32.9±0.1	32.8[q]
	195	33.8±0.1	33.7[q]
	196	25.2±0.1	25.4[q]
	197	<0.001	...
	198	7.19±0.04	7.23[q]
	199	<0.001	...
	200	<0.001	...
Pb	202	<0.003	...
	203	<0.0004	<0.002[r]
	204	1.40±0.02	1.48[r]
	205	<0.001	<0.01[r]
	206	25.2±0.1	23.6[r]
	207	21.7±0.1	22.6[r]
	208	51.7±0.2	52.3[r]
	209	<0.00005	<0.001
	210	<0.000007	<0.001
	211	<0.000007	...
	212	<0.000007	...
	213	<0.000005	...
U	232	<0.00006	<0.0029[s]
	233	<0.00006	<0.0029[s]
	234	0.0056±0.0001	0.0058[t]
	235	0.718±0.005	0.715[s]
	236	<0.0002	<0.0029[s]
	237	<0.0001	<0.0083[s]
	238	99.276±0.005	99.28[s]
	239	<0.0001	<0.0083[s]
	240	< 0.00005	<0.0015[s]

[a] J. P. Blewelt, Phys. Rev. **49**, 900 (1936).

[b] J. R. White and A. E. Cameron, Atomic Energy Commission Report MDDC-1195 (unpublished); Phys. Rev. **74**, 991 (1948).

[c] A. K. Brewer, Phys. Rev. **49**, 856 (1936).

[d] M. S. Sampson and W. Bleakney, Phys. Rev. **50**, 456 (1936).

[e] A. O. C. Nier, Phys. Rev. **77**, 789 (1950).

[f] A. O. C. Nier, Phys. Rev. **48**, 283L (1935).

[g] A. O. C. Nier, Phys. Rev. **50**, 1041 (1936).

[h] W. T. Leland, Phys. Rev. **77**, 634 (1950).

[i] D. C. Hess, Jr., and M. G. Inghram, Phys. Rev. **76**, 1717 (1949).

[j] M. B. Sampson and W. Bleakney, Phys. Rev. **50**, 732 (1936).

[k] A. A. Cohen, Phys. Rev. **63**, 219(A) (1943).

[l] L. Friedman and A. P. Irsa, J. Am. Chem. Soc. **75**, 5741 (1953).

[m] A. O. C. Nier, Phys. Rev. **52**, 933 (1937).

[n] Inghram, Hayden, and Hess, Phys. Rev. **72**, 967 (1947).

[o] Inghram, Hess, and Hayden, Phys. Rev. **74**, 98 (1948).

[p] R. F. Hibbs, Atomic Energy Commission Report AECU-556, August 1949 (unpublished).

[q] Inghram, Hess, and Hayden, Plutonium Project Report, Argonne National Laboratory ANL-4012, July, 1947, (unpublished), p. 7, also W. T. Leland, Phys. Rev. **76**, 992 (1949).

[r] A. O. C. Nier, J. Am. Chem. Soc. **60**, 1571 (1938).

[s] A. O. C. Nier, Phys. Rev. **55**, 150 (1939).

[t] Goldin, Knight, Macklin, and Macklin, Phys. Rev. **76**, 1561 (1949), also C. A. Keinberger, Phys. Rev. **76**, 1561 (1949).

greater than 1/10 000 of Sc^{45}. The thermal ions observed were the Sc^+ metallic ions. No appreciable ScO^+ ions were noticed. Approximately 10^{-14} ampere of Sc^{45} ions was obtained at 1200°C.

Vanadium

A solution of vanadyl chloride containing 0.2 milligram of vanadium was evaporated to dryness on a tungsten filament. Interest in this element was centered around the possible existence of an isotope at mass 48. Minute impurity peaks from calcium, chromium, and titanium made it difficult to place very low upper limits on this and other isotopes of vanadium. The results listed below are the result of correcting for the presence of the known impurities. In all cases, the calculated correction would account for the entire peak, but a 5 percent uncertainty in the correction allows only the following upper limits to be placed for other isotopes relative to V^{51}: V^{48}—1/1 000 000 of V^{51}; V^{49}—1/2 000 000 of V^{51}.

The V^{51}/V^{50} ratio, after correcting the V^{50} peak for slight contributions from titanium and chromium, was found to be 398±15.

Manganese

Manganese chloride was used as a source of Mn^+ ions. No isotopes other than Mn^{55} were found in this investigation. Impurity peaks due to chromium caused interference in the manganese region. By using Nier's[5] values for the abundance of the chromium isotopes, corrections were made on the observed peak height at mass 53 and 54. The remaining impurity peaks at mass 56 and 57 were not identified, thus preventing limits from being obtained at these positions.

---

[5] A. O. C. Nier, Phys. Rev. **55**, 1143(A) (1939).

Niobium

A niobium filament was used as a source of Nb^+ ions. Impurity peaks due to molybdenum were noticed. By subtracting the molybdenum contribution according to the abundances given by Williams and Yuster,[6] upper limits were placed on all other possible isotopes of niobium from mass 89 to mass 98.

Ruthenium

Powdered ruthenium sponge was suspended in amyl acetate and evaporated to dryness on a tungsten filament. The intensity of ruthenium ions by surface ionization was extremely high and stable for long periods of time, and over a wide range of temperatures.

The accepted values for the abundance of the isotopes of ruthenium are based on the photographic-plate measurements of Ewald[7] using Nier's old cadmium abundances for calibration purposes. The values measured in this investigation are in good agreement with Ewald's values, except for Ru^{98} where our value is 16 percent lower. Our values are in better agreement with those reported by Friedman and Irsa.[8] In the case of the lower mass ruthenium isotopes, corrections of about 2 percent were made for the molybdenum contribution which was noticed at all temperatures studied. The abundance values are given in Table I together with those reported by Friedman and Irsa.

Indium

Indium metal was used as a source of its own ions. No other isotopes of indium other than In^{113} and In^{115} were detected. Due to the relatively high ion emission current of In, it was possible to obtain very low upper limits on the existence of other isotopes.

Cesium

A solution of cesium nitrate was evaporated to dryness on a niobium filament. Cs^+ ions were obtained at a filament temperature of 500-degrees Centigrade. Cesium was found to be simple, and the extremely low upper limit shown in Table I for the existence of other possible Cs isotopes were obtained by extrapolation of the counting-rate losses at Cs^{133} up to 5×10^8 counts per minute (10^{-12} ampere).

Lanthanum

A solution of lanthanum oxide in dilute HCl was coated on a tantalum filament. Both the La^+ and the $(LaO)^+$ peaks had to be corrected for the presence of barium ions in the lanthanum spectra. The La^{139}/La^{138} ratio was measured to be 1120 ± 20.

Neodymium

Neodymium oxide, "spec pure," was used in this investigation. Both the NdO^+ ions and the Nd^+ ions were observed. SmO^+ ions caused slight contributions in the NdO^+ region, but no interference was observed at the metal region. The abundances measured in this investigation are given in Table I, together with the values obtained by Inghram, Hess, and Hayden in 1948. Other isotopes were searched for, and the upper limits obtained are shown in Table I.

Hafnium

The source of hafnium ions was a sliver of hafnium metal, melted onto the tungsten filament. The thermal ions measured were Hf^+, no indication of HfO^+ ions being observed. The isotopic ratio Hf^{180}/Hf^{179} was corrected for the minute contribution from W^{180}, which amounted to less than 0.1 percent. The abundances measured are given in Table I. Also, included in this table are the values obtained by Hibbs,[9] using electron bombardment of $HfCl_4$ vapor.

Tantalum

A tantalum metal filament was used as a source of its own ions. It has recently been shown[10] that this element consists of two isotopes, Ta^{181} and Ta^{180}, and is not monoisotopic as previously believed. The Ta^{181}/Ta^{180} ratio was measured to be 8120 ± 200. Upper limits measured for the existence of other isotopes relative to Ta^{181} are: Ta^{177}, Ta^{178}, Ta^{179} – 1/330 000 of Ta^{181}; Ta^{182}, Ta^{183} – 1/500 000 of Ta^{181}.

Platinum

A sliver of chemically pure platinum was melted onto a tungsten filament. The Pt^+ beam current from such a source of thermal ions was adequate for an accurate analysis of the Pt isotopes, but the beam currents could not be made large enough to search for new isotopes below 0.001 percent relative abundance. Three samples were used in an attempt to obtain a higher Pt^+ current; but in each case, the filament burned out at approximately 1800-degrees Centigrade, probably due to the formation of a W-Pt alloy of relatively low melting point. The abundances measured in this investigation are given in Table I, along with the previous adopted values of Inghram, Hess, and Hayden[11] and the value for Pt^{190} measured by Leland.[12] By correcting for a small iridium 191 and 193 peak, the upper limits were determined for other isotopes relative to Pt^{195}.

---

[6] D. Williams and P. Yuster, Phys. Rev. **64**, 556 (1948).

[7] H. Ewald, Z. Physik **122**, 686 (1944).

[8] L. Friedman and A. P. Irsa, J. Am. Chem. Soc. **75**, 5741 (1953).

[9] R. F. Hibbs, Atomic Energy Commission Report AECU-556, August, 1949 (unpublished).

[10] White, Collins, and Rourke, Phys. Rev. **97**, 556 (1955).

[11] Inghram, Hayden, and Hess, Plutonium Project Report, Argonne National Laboratory Report ANL-4012, 1947 (unpublished).

[12] W. T. Leland, Phys. Rev. **76**, 992 (1949).

Lead

Two milligrams of lead nitrate was used as a source material. Adequate ion beams of Pb^+ were produced by surface ionization. The spectrum obtained is shown in Fig. 2.

The main interest in investigating lead was to look for the possible existence of Pb^{205}. This mass position was found to be occupied by Tl^{205}, as proved by the presence of a mass 203 peak in the ratio of $205/203=2.4$. Upper limits for these isotopes of lead were obtained by correcting for the thallium peaks.

The mass 209 position showed the presence of a peak which, if it were Pb^{209}, had an abundance of 0.0005 percent. The possibility that this peak was due to a trace of impurity of bismuth cannot be excluded. The relative abundance of this peak was constant over a temperature range from 2000°C to 2600°C, and was observed in two samples of lead. A third sample was spiked with 0.93 percent by weight of bismuth. Analysis of this sample showed the 209 peak had an average abundance of 0.5 percent, but was far from constant, varying from 0.1 percent to 2 percent with changes in temperature. Chemically "pure" bismuth itself, when analyzed, was found to give large beam currents of the lead isotopes, indicating the relative ionization efficiency is much higher for lead.

The abundances measured for lead isotopes are given in Table I. The source of the lead sample is unknown.

Uranium

A water solution of uranium nitrate evaporated on a tungsten filament was used as a source of thermal ions. Large beam currents of UO_2^+, UO^+, and U^+ ions were observed. The U^+ ion was the major component at temperatures above 1500-degrees centigrade; consequently, the isotopic abundance measurements were made in the U^+ region. The resolution between U^{234} and U^{235} was such that less than 0.5 percent of the U^{234} peak height was due to the spread of the U^{235} beam. Resolution between the U^{235} and U^{238} peaks was such that less than 0.01 percent of the U^{235} peak could possibly be due to U^{238}. The results are given in Table I together with the present adopted relative isotopic abundances which were obtained from a combination of Nier's[13] mass spectrometric values and specific alpha activity measurements of Kienberger[14] and Goldin *et al.*[15]

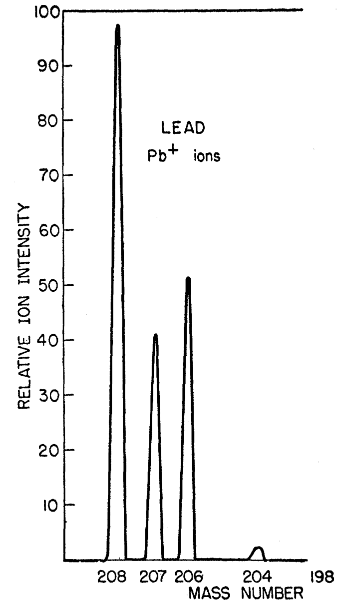

FIG. 2. Mass spectrum of lead.

CONCLUSION

Of the nineteen elements reported here, seven (manganese, ruthenium, cesium, lanthanum, hafnium, platinum, and lead) were suggested by Stehn[16] of our Theoretical Physics Unit, as possibly containing undiscovered stable or long-lived isotopes. This investigation has not reduced appreciably the possibility of Ru^{94}, Pt^{188}, Pb^{202}, or Pb^{205} existing in nature, but the probability that the remaining elements contain new isotopes is considerably reduced.

The elements reported here are those which could be readily analyzed in our present instrument without making substantial changes in our source arrangement. The study will be continued to include the rare earth elements as well as those of the fifth and sixth periods which are not reported here.

---

[13] A. O. C. Nier, Phys. Rev. **55**, 150 (1939).

[14] C. A. Keinberger, Phys. Rev. **76**, 1561 (1949).

[15] Goldin, Knight, Macklin, and Macklin, Phys. Rev. **76**, 336 (1949).

[16] J. R. Stehn (private communication, January, 1954).