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A Mass-Spectrographic Study of the Isotopes of Hg, Xe, Kr, Be, I, As, and Cs

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A new mass spectrometer is described which has been constructed for the purpose of making accurate determinations of relative abundances of isotopes. The high intensity and high resolving power of the apparatus allow the search for rare isotopes to be extended beyond previously attainable limits. The elements Hg, Xe, Kr, Be, I, As, and Cs have been investigated. Measurements of the relative abundances of the Hg, Xe, and Kr isotopes were made. A search was made for other isotopes than those already known, but none was found. The nonexistence of Hg^{197} is confirmed, and the existence of Hg^{203} is shown to be extremely doubtful. Cs, As, and I are shown to be single to a very high degree. A search for Be^8 revealed that if it existed it must be less than 1/100,000 of Be^9.

THE growth of nuclear physics has made necessary a more exact knowledge of the isotopic constitution of the elements. The existence or nonexistence of rare isotopes must be established, and more should be known about the relative abundances. In this paper will be described a mass spectrometer of extremely high resolving power and sensitivity, especially designed for searching for rare isotopes and measuring relative abundances.

For the elements studied here, a search for possible rare isotopes was made in all the mass number positions in the immediate neighborhood of the known isotopes. In a number of these positions one would hardly expect to find stable isotopes as radioactive ones have already been reported for them. In these cases, the low upper limits for abundances given here may be interpreted as additional evidence for the instability of the positions. In a systematic study of the sort undertaken here, it seems worth while to record all such facts.

APPARATUS AND PROCEDURE

A diagram of the mass spectrometer is shown in Fig. 1. In principle it differs little from that previously described.[1] The present apparatus is considerably larger, and is designed to operate between the poles of an electromagnet instead of inside a solenoid as was previously the case. Because of the larger geometry and greater magnetic field the resolving power has been increased considerably without a loss in sensitivity

The metal parts were constructed entirely of Nichrome V, except for the analyzer *H*, which was made of copper. A Pyrex glass housing covered with electric furnaces enveloped the metal parts.

Electrons from the tungsten filament *F* are accelerated by a voltage connected between *F* and *A*. They follow the magnetic lines of force and are caught on the plate *E* in the box attached to *A*. On their way across the tube they produce positive ions which are drawn through the slit S_1 by a potential applied between *D* and *B*. The beam consisted of a ribbon of electrons about 2 mm wide and 0.2 mm thick.

Those ions which pass through the slit S_1 are further accelerated by a large potential between *B* and *C*. A certain fraction of these then passes through slit S_2 into the 180° analyzer. In the analyzer all ions travel in circular paths; those

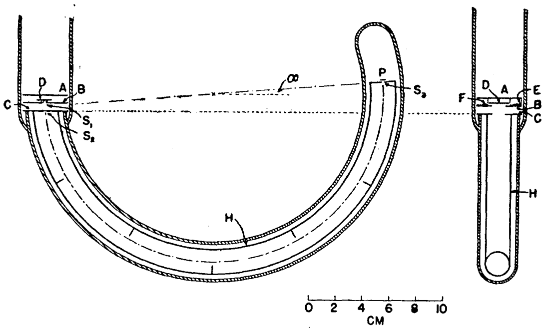

FIG. 1. Diagram of mass spectrometer. Slits S_1 and $S_2=0.2$ mm, Slit $S_3=0.3$ mm. The magnetic field is perpendicular to the paper in the cross-sectional view shown at the left.

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[1] Nier, Phys. Rev. **50**, 1041 (1936).

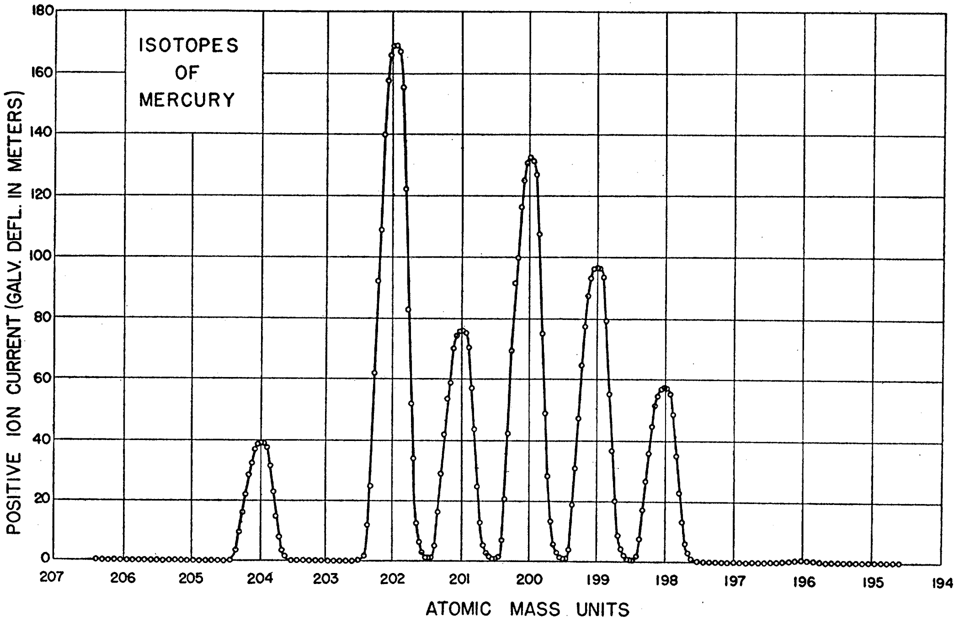

FIG. 2. Mass spectrum showing the isotopes of mercury. Hg^{196} just visible. Positive ion current is expressed in terms of meters deflection of galvanometer at its highest sensitivity. One meter represents about 2.5×10^{-13} amp.

which have the proper ratio of mass to charge emerge through S_3 behind which they are collected by the plate P and measured by an electrometer tube amplifier.

If the energy of the ions is varied, one can sweep over the mass spectrum. The magnetic field was held constant by a special vacuum tube compensator attached to the generator supplying the power for the electromagnet. No difficulty was had in holding the magnetic field constant to one part in 10,000. In practice, the magnetic field employed for a given element was such that the ions needed approximately 1200 volts energy to travel in the required path, a circle of five inch radius. The radius of the pole face of the magnet was seven inches, and the air gap width was two inches.

The accelerating voltage given the ions was applied by means of a voltage divider connected between D, B, and C in such a fashion that the drop between D and B was always a given fraction of that between D and C. As pointed out by Bleakney,[2] this is desirable, as then all ions regardless of m/e value will have traveled the same paths before entering the analyzer.

Because of the trochoidal motions of the ions in going from S_1 to S_2 the plate C does not lie in a radial plane of the analyzer. It is tilted at a small angle α (approximately 3°) as indicated in the diagram. The ions thus enter the analyzer at the proper angle to travel in the center.

As the apparatus is entirely free of wax and grease joints, it can be baked and thoroughly outgassed before the introduction of an unknown element. Only a trace of CO, CO_2 and water vapor are found present after such treatment. No other residual gasses can be detected so that the danger of mistaking a residual gas for a rare isotope is entirely negligible for all the elements studied. Because of the extremely low pressure of the water vapor present and the complete absence of grease vapor, troublesome hydrides are never formed.

For the measurement of relative abundances and the detection of rare isotopes, electrical measurement of ion currents such as is employed

TABLE I.

MASS NUMBER	196	197	198	199	200	201	202	203	204
Abundance	0.50	0	34.2	57.6	78.7	44.6	100	0	22.7
Aston's values	0.34	(0.03)	33.7	56.2	81.1	46.6	100	(0.02)	23.4

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

here has many advantages over photographic recording. The quantitative measurement of intensities by the photographic method is, at its best, tedious, and is subject to many errors. The direct measurement of current with electrical instruments is to be preferred.

In the work to be presented here, the abundance of an isotope is assumed to be proportional to the positive ion peak it produces in the mass spectrogram. Sometimes, especially if large space charges are present, it is not correct to assume this, as the apparatus may discriminate. If this is the case, one always finds that an abundance ratio will be a function of some of the conditions in the tube, such as the pressure, electron current, or other factors. Conversely, if the abundance ratio remains constant in spite of large changes in all the possible variables, one is quite certain that the value is correct. In all of the work presented here, this point was carefully checked, and it is believed that the values given are correct within the limits of error imposed.

Results

Mercury[3]

A typical mass spectrum obtained is shown in Fig. 2. The isotopes found and their respective abundances are listed in Table I. Aston's[4] values, converted from percentage to relative abundances are also included.

The mean mass number is 200.61. When allowance is made for the packing fraction of +0.8 and for the conversion factor of 1.000275 in going from the atomic to the chemical scale, a chemical weight of 200.58 is obtained. Aston's figures yield a value 200.60. The present International value is 200.61. The writer believes that his abundances relative to Hg^{202} are all correct to within one percent, except Hg^{196}, which may be correct to only about three percent.

A search for other isotopes was also made. The following upper limits for abundances relative to Hg^{202} were found: Hg^{206}, 1/18,000; Hg^{205}, 1/18,000; Hg^{203}, 1/5000; Hg^{197}, 1/8000; Hg^{195}, 1/20,000; Hg^{194}, 1/20,000.

Bainbridge and Jordan[5] showed that Hg^{197} does not exist to half of the amount attributed to it by Aston. The present investigation shows that it cannot exist to one-fourth the amount attributed to it by Aston. The upper limit set on Hg^{203}, 1/5000 of Hg^{202}, is just the abundance given for it by Aston. While the present investigation may not be conclusive on this point, it does show that Aston's Hg^{203} was probably spurious.

Figure 3 shows a spectrum taken in the region about Hg^{196}. From this spectrogram one can obtain an idea of the resolving power and sensitivity of the apparatus. Hg^{196}, it should be pointed out, is barely visible in Fig. 2.

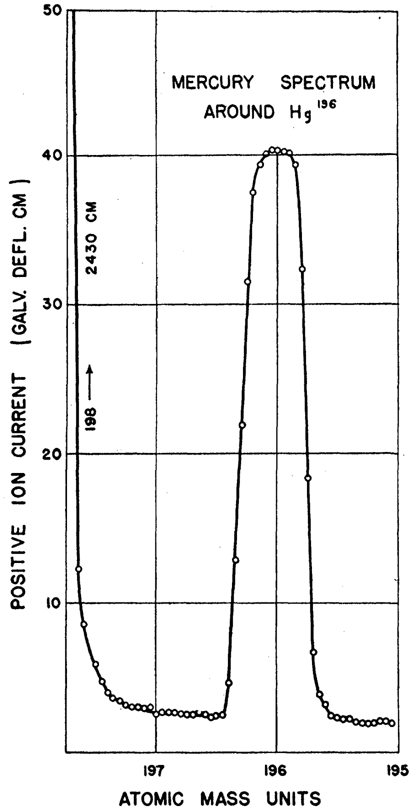

FIG. 3. A spectrum taken in the region about Hg^{196}. Hg^{198} peak 2430 cm high for this run.

Xenon

A typical spectrum obtained for xenon is shown in Fig. 4. The isotopes found were the same as those reported by Aston.[6] Table II

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[3] Presented at the Washington meeting of the Am. Phys. Soc. April 29, 1937. Phys. Rev. **51**, 1007 (1937).

[4] Aston, *Mass Spectra and Isotopes*, p. 120.

[5] Bainbridge and Jordan, Phys. Rev. **50**, 282 (1936).

[6] Aston, *Mass Spectra and Isotopes*, p. 106.

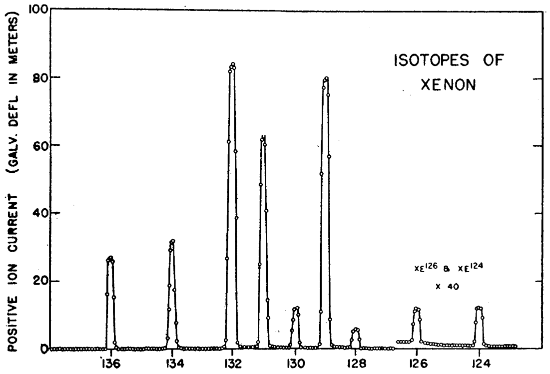

FIG. 4. Mass spectrum showing the isotopes of xenon. Xe^{126} and Xe^{124} drawn to 40 times scale.

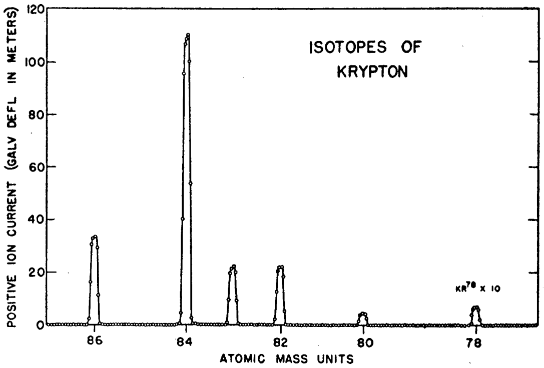

FIG. 5. Mass spectrum showing the isotopes of krypton. Kr^{78} drawn to 10 times scale.

gives the abundances along with those given by Aston. The mean weight obtained is 131.40. When account is taken of the packing fraction of -5.3 and the conversion factor in going from the atomic to the chemical scale, a chemical weight of 131.29 is obtained. Aston's figures yield the weight 131.24. The present International value is 131.3. The writer believes that his abundances relative to Xe^{132} are correct to one percent, except for the values for Xe^{126} and Xe^{124}, which are correct to at least three percent.

A search was made for other isotopes. None was found, and the following upper limits can be set for their abundance relative to Xe^{132}: Xe^{138}, Xe^{137}, Xe^{135}, and Xe^{133}, 1/15,000 Xe^{132}; Xe^{127}, 1/30,000 Xe^{132}; Xe^{125}, Xe^{123}, and Xe^{122}, 1/60,000 Xe^{132}.

Krypton

Krypton was found to consist of six isotopes, as was shown by Aston.[7] A typical spectrum is shown in Fig. 5. A list of the isotopes and their respective abundances is given in Table III. Aston's values, converted from percentage to relative abundances, are also included. The chemical weight, computed from the above figures, is 83.81. Aston's figures give the value 83.77. The present chemical weight is 83.7. The writer believes that his abundances relative to Kr^{84} are correct to one percent, except the Kr^{78} one, which should be within two percent.

TABLE II.

MASS NUMBER	124	126	128	129	130	131	132	134	136
Abundance	0.347	0.327	7.06	97.3	15.1	78.5	100.0	39.1	33.2
Aston's values	0.295	0.295	8.48	100.0	15.4	76.3	97.6	38.0	32.4

As with the other elements, a search was made for other isotopes. None was found, and the following upper limits for their existence relative to Kr^{84} could be set: Kr^{88}, 1/50,000 Kr^{84}; Kr^{87} and Kr^{85}, 1/25,000 Kr^{84}; Kr^{81}, Kr^{79}, Kr^{77}, and Kr^{76}, 1/50,000 Kr^{84}.

Beryllium

Beryllium was introduced into the apparatus as $BeCl_2$ vapor. The reason for studying this element was to search for Be^8. Thus the 78 position $(Be^8Cl^{35}Cl^{35})^+$ and the 43 position $(Be^8Cl^{35})^+$ were investigated. It was concluded that Be^8, if stable, exists to less than one part in 100,000 in Be. Bleakney, Blewitt, Sherr, and Smoluchowski[8] showed that it did not exist to more than one part in 10,000.

Arsenic

Aston[9] found arsenic to consist of a single isotope of mass 75. The present investigation confirms this. The element was introduced into the apparatus in its vapor form. The following upper limits can be set for hypothetical isotopes: As^{79}, As^{78}, and As^{77}, 1/100,000 As^{75}; As^{76},

TABLE III.

MASS NUMBER	78	80	82	83	84	86
Abundance	0.608	3.52	20.2	20.2	100	30.6
Aston's values	0.738	4.31	20.7	20.7	100	29.4

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[7] Aston, *Mass Spectra and Isotopes*, p. 105.

[8] Bleakney, Blewitt, Sherr and Smoluchowski, Phys. Rev. **50**, 545 (1936).

[9] Aston, *Mass Spectra and Isotopes*, p. 139.

1/50,000 As^{75}; As^{74}, 1/20,000 As^{75}; As^{73}, As^{72}, and As^{71}, 1/100,000 As^{75}.

Iodine

Aston[10] showed that iodine consisted of a single isotope of mass 127. This, too, is confirmed by the present investigation. Iodine vapor was admitted into the apparatus and a search was made around the 127 and 254 peaks. It was possible to set the following upper limits for the abundances of other isotopes relative to I^{127}: I^{131}, 1/250,000; I^{130}, 1/120,000; I^{129}, 1/40,000; I^{128}, 1/15,000; I^{126}, 1/25,000; I^{125}, I^{124}, and I^{123}, 1/50,000.

Caesium

Caesium was found to be single by Aston.[11] Bainbridge[12] searched for other isotopes, but was unable to find any. Because of the much higher sensitivity of the present apparatus it seemed worth while to search further. Caesium was introduced in its vapor form into the apparatus. No new isotopes were found. The following upper limits can be set for the abundances of hypothetical isotopes relative to Cs^{133}: Cs^{137} and Cs^{136}, 1/100,000; Cs^{135}, 1/50,000; Cs^{134}, 1/6000; Cs^{132}, 1/4000; Cs^{131}, 1/20,000; Cs^{130} and Cs^{129}, 1/100,000.

The construction of the large electromagnet used in this work was made possible by a grant from the Milton fund. The author wishes to express his appreciation to Professor Kenneth T. Bainbridge for his encouragement and assistance, to Mr. H. L. Leighton for his design and construction of the glass parts of the apparatus, and to Mr. David Mann for his suggestions and aid in the construction of the electromagnet.

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[10] Aston, *Mass Spectra and Isotopes*, p. 154.
[11] Aston, *Mass Spectra and Isotopes*, p. 111.
[12] Bainbridge, Phys. Rev. **36**, 1668 (1930).

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Wide-Angle Interference of Multipole Radiation

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The paper presents a discussion of wide-angle interference phenomena and their dependence upon the nature of the light. Formulas are derived representing the interference pattern for various geometrical arrangements and for arbitrarily composed sources. It is shown that the coherence properties of beams emerging from a point source should depend in a marked manner upon the type of poles which compose it. Special discussion is given for dipole, quadrupole, and octopole sources. The treatment is limited to the use of ideal mirrors; extensions are indicated.

I. Introduction

The coherence properties of light rays emitted from a point source under a finite angle were first investigated to some extent in the well-known experiment of Schrödinger[1] on wide-angle interference. This experiment was originally undertaken to decide between the concepts of unidirectional and spherical emission of light; it was thus primarily concerned with a side of the question which today possesses only historical importance, since the new quantum theory has reinterpreted the apparent conflict between the two old points of view.

Schrödinger observed that the two rays which passed through two holes in a screen produced an interference pattern on the other side. In the *final* interpretation given to his results he remarked that this observation follows alone from the assumption that the interference phenomenon can be described with the help of the Huygens-Kirchhoff principle by the assignment of proper values to the light vectors in the two holes: such a description, according to Schrödinger, leads with necessity to an interference pattern behind the screen.

It is at this point that our opinion differs. We believe that even at the present state of theoretical physics extended importance can be

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[1] E. Schrödinger, Ann. d. Physik **61**, 69 (1920).