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Mass Spectrum Analysis

1. The Mass Spectrograph. 2. The Existence of Isobars of Adjacent Elements[*]

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(1) The simultaneous fulfillment of the conditions for high resolving power, sensitivity and dispersion have been obtained in an instrument for the investigation of isotopes. Important features of the design are the linearity of the mass scale over a broad region of the recording plate, the elimination of the troublesome effects of surface charges on the electric deflection plates, and the use of relatively wide collimating slits at small separation. The ions diverging from the second collimating slit S_2 are deflected through $\pi/\sqrt{2}$ radians, using the radial field method of electrostatic focusing introduced by Hughes and Rojansky, which produces an energy spectrum in front of the third slit S_3. Beams of ions of all m/e values comprising up to 3 percent of this energy spectrum, traversing S_3, are further deflected through a mean angle of $\pi/3$ radians by a magnetic field which focuses the ions and produces a mass spectrum on the recording plate. The mean dispersion is 5 mm for one percent mass difference, the mean radius in the electric and magnetic fields is 25.4 cm, and a resolving power $M/\Delta M$ of *ca.* 10,000 is attained in routine work. Over 140 mm on the plate, the maximum divergences from linearity are $\pm 1/7000$. (2) Mattauch and Sitte have suggested that isobars differing by one unit in atomic number are unstable or do not exist at all. To test for the existence of isobars of this class, singly and in some cases doubly ionized germanium, cadmium, indium, tin, tellurium, mercury, lead and bismuth were examined, using hydrogen free ion sources. Definite proof was obtained for the existence of three isobaric pairs $Cd^{113}In^{113}$, $In^{115}Sn^{115}$ and $Sb^{123}Te^{123}$, which apparently are stable. The isotopes Cd^{115}, Sn^{121}, Hg^{197}, Pb^{209}, which have been reported as members of isobaric pairs, cannot be present to greater than 1/8, 1/60, 1/2, 1/10, respectively, of the amounts published by Aston. The existence of Pb^{205} is also doubtful. In accordance with Beck's suggestion, developed by Bethe, and by Yukawa and Sakata, both members of these isobaric pairs could be stable for a neutrino mass $m_n > 0$. This hypothesis would necessitate modifications in the theory of the emission of β-rays. Alternatively, a difference in the nuclear spins of the members of the isobaric pairs might result in a condition of effective stability for these nuclei.

1. THE MASS SPECTROGRAPH

VARIOUS combinations of electric and magnetic fields, which act to separate out beams of ions of the same m/e values, are used for the photographic recording of mass spectra. By these methods different degrees of velocity and direction focusing are attained, depending on the ability of the instrument to focus on the recording plate ion beams which are heterogeneous in energy or which diverge from the source. No review of the subject will be undertaken here, as there is an excellent discussion and comparison of mass spectrographs of various types in a recent work by Brueche and Scherzer.[1]

In practice no one instrument has been able to attain simultaneous fulfillment of the conditions for velocity and direction focusing, together with a linear mass scale, one or another having been sacrificed to obtain high resolving power, freedom from "polarization" phenomena or other undesirable effects. The first part of this paper is a brief description of a mass spectrograph of high resolving power, high sensitivity and high dispersion which has been constructed for the study of the masses and the relative abundance of atomic species. The instrument conforms to a new design in which velocity and direction focusing have been secured to a useful extent, and at the same time the undesirable effects of surface charges on the deflecting plates which occasionally might act to change the shape and position of the lines on the recording plate, have been eliminated. An important feature of the design and one which is useful in the comparison of the masses of isotopes to the highest degree of accuracy, is the linearity of the mass scale over a broad region of the recording plate.

The second part of the paper reports the results of the analyses of certain elements, undertaken to secure experimental evidence concerning the

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[†] Sometime Fellow of the John Simon Guggenheim Memorial Foundation. The mass spectrograph was designed during the tenure of this Fellowship.

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[1] E. Brueche and O. Scherzer, *Geometrische Elektronenoptik* (J. Springer, 1934).

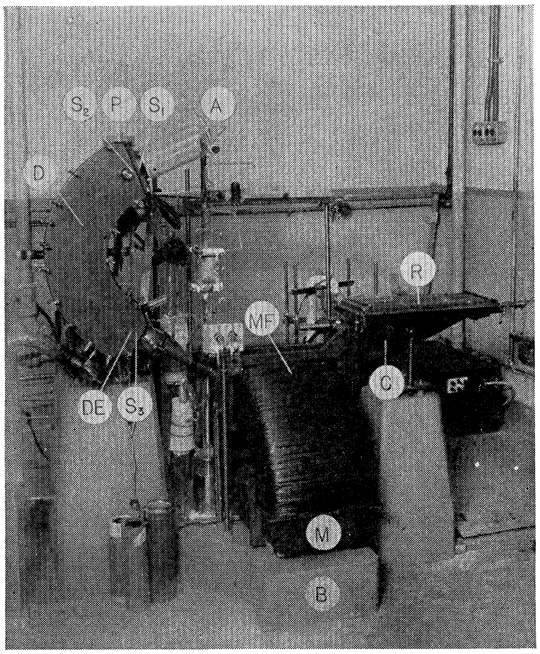

FIG. 1. The mass spectrograph. A, the discharge tube. S_1, adjustable steel slit under the cathode. P, port for adjusting S_2, the second slit. D, deflection chamber containing radial electrostatic field deflecting plates. DE, adjustable diaphragm in front of the third slit S_3, MF, the magnetic field. M, the magnet yoke. B, reinforced concrete base. C, camera chamber. R, cover of the plate-holder compartment.

existence of isobars of elements differing by one unit in atomic number. Mattauch[2] has suggested that isobars of this class either are unstable or do not exist at all and a similar conclusion follows from Heisenberg's discussion of nuclear stability.[3]

The mass spectrograph

Fig. 1 is a photograph and Fig. 2 is a scale drawing of the mass spectrograph. The ions from the discharge tube A are collimated by slits S_1 and S_2. They are then deflected through $\pi/\sqrt{2}$ radians, using the radial field method of electrostatic focusing introduced by Hughes and Rojansky[4] to produce an energy spectrum in front of the third slit S_3. Beams of ions of all m/e values comprising up to 3 percent of this energy spectrum, diverging from the focal plane of the electrical deflector, are further deflected through a mean angle of $\pi/3$ radians by the magnetic field, MF, which forms a mass spectrum of focused ion beams on the recording plate.

Velocity focusing of the ions

The focusing of ions of different velocities is obtained as a result of the geometrical arrangement of the electric and magnetic deflecting fields which are so placed that the dispersion of the ions produced by one field is cancelled by the dispersion produced by the other field for a given velocity difference. Under these conditions ions of the same m/e values having all velocities within a certain range are focused on the recording plate.

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[2] J. Mattauch. Zeits. f. Physik **91**, 361 (1934).

[3] W. Heisenberg. Zeits. f. Physik **78**, 156 (1932).

[4] A. L. Hughes and V. Rojansky, Phys. Rev. **34**, 284 (1929).

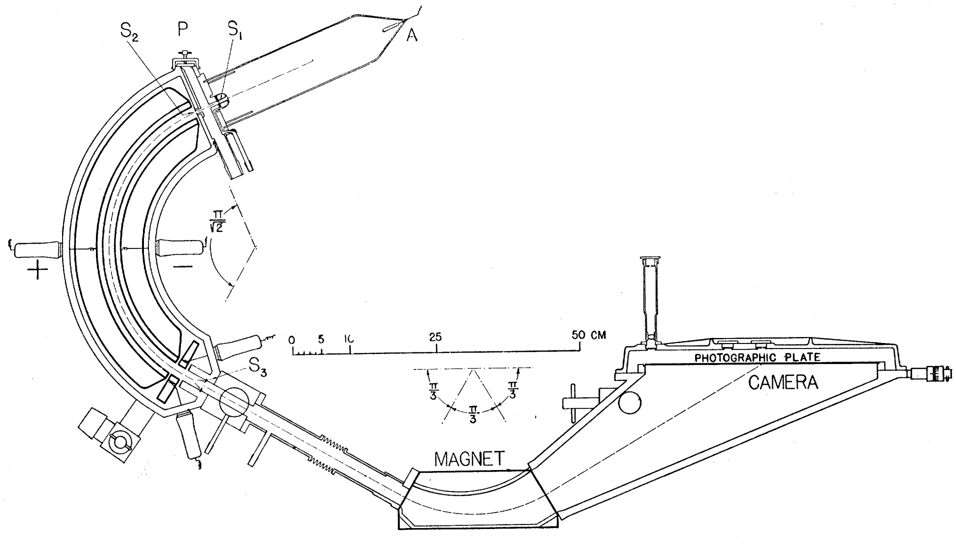

FIG. 2. Scale drawing of the mass spectrograph.

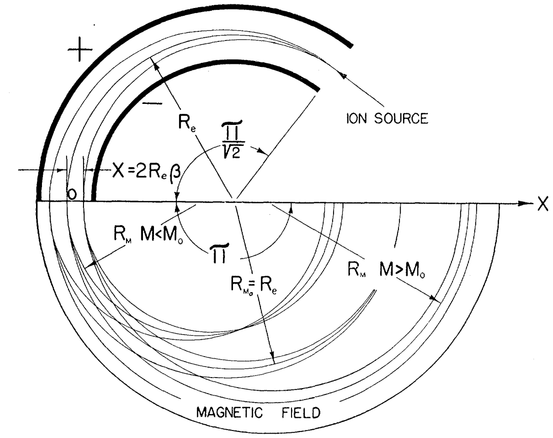

FIG. 3. $\pi/\sqrt{2}$ electric and π magnetic deflections combined. Not to scale.

The principle of the method used to secure magnetic focusing of ions which differ in velocity may be explained by considering first the elementary special case of 180° magnetic deflection applied to ions which enter the field with the dispersion in velocities produced by the Hughes and Rojansky electrostatic analyzer. For the purpose of this illustration the electric and magnetic fields are assumed to terminate at the x axis.

The expression for the displacement of ions of different velocities which have emerged from the inverse first power electrostatic field between the plates in Fig. 3 is given by $x=\Delta Re=2R_e(\beta-\beta^2)$ where $\beta=\Delta V/V_0=(V_0-V)/V_0$, V is the ion velocity and V_0 is the velocity of the ions in the central path of radius R_e. $\Delta R_e=2R_e\beta$ to a sufficient approximation. In the magnetic field $\Delta R=-R\beta$ as β is negative for $V>V_0$.

Ions of masses M greater or less than that, M_0, of the central beam for which $R_{\text{mag}}=R_{M_0}=R_e$ have radii of curvature $R_M=R_{M_0}(M/M_0)^{\frac{1}{2}}$.[5]

The condition for optimum focus of ions of mass M of different velocities is $R_{M_0}=R_e$. In this case the trajectories of all the ions in the magnetic field intercept the x axis at the same point after a deflection of 180°. The general expression for the distance from the origin to the point of intersection with the x axis is given by $x=2R_{M_0}(M/M_0)^{\frac{1}{2}}(1-\beta)+2R_e\beta$, and if $R_{M_0}=R_e$ and $M=M_0$, x is the same for all values of β which may extend over a range of 2 percent, corresponding to a 4 percent energy band in a practical apparatus.

For ions of mass differing from M_0 the breadth of focus increases on each side of the position $x=2R_{M_0}$. The width of the traces, considering velocity focusing only, is given by $4\beta R_{M_0}((M/M_0)^{\frac{1}{2}}-1)$.

In Table I the width of the traces which is due to imperfect velocity focusing is listed for line positions corresponding to masses of values from $0.6M_0$ to $1.4M_0$, somewhat more than an octave of mass. In the case of 180° magnetic focusing the closest approximation to a linear mass scale over the recording plate is secured if M_0 is at the center of the plate at a distance from the origin $x=2R_{M_0}$. Table I was compiled on this basis.

Various alternative arrangements of the electric and magnetic fields were considered to improve the velocity focusing, to obtain a closer approximation to a linear scale, to avoid the overlapping of stray fields and to obviate the necessity of using a magnet with large pole area to obtain spectra of high dispersion.[6]

The geometry of the fields shown in Figs. 2 and 4 was finally selected as a satisfactory arrangement because it allowed the conditions for velocity and angle focusing to be simultaneously satisfied. With this disposition of the fields, an electric deflection of $\pi/\sqrt{2}$ and a mean magnetic deflection of $\pi/3$ radians, a good approximation to a linear mass scale is secured over the whole of the plate. Over a region 14 cm long the maximum divergences from linearity as determined experimentally are ±1/5000 to 1/7000 depending on the particular slit arrangements used and the position of the recording plate.

It was not found possible to obtain simple equations to express the dispersion and trace

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[5] The Hughes and Rojansky focusing method produces an energy spectrum of focused ions of all m/e values after a deflection of $\pi/\sqrt{2}$ radians in the radial electric field. Any ion of mass M which followed the central path in the electric field, $R=R_e$, describes in the magnetic field a path with a radius of curvature $R_M=R_{M_0}(M/M_0)^{\frac{1}{2}}$, where $R_{M_0}=R_e$, and M_0 is the mass of the ions which are incident on the center of the recording plate. The trajectories in the electric and magnetic fields of ions of different energies can be more simply expressed as a function of $\beta=\Delta V/V_0$, the fractional deviation in velocity, than as a function of their fractional energy difference.

[6] The interesting papers of R. Herzog, and J. Mattauch and R. Herzog, Zeits. f. Physik **89**, 447, 786 (1934), in which entirely different field arrangements are discussed were not published until some months after the present design had been completed and construction of the instrument had been started in the Harvard University instrument shops.

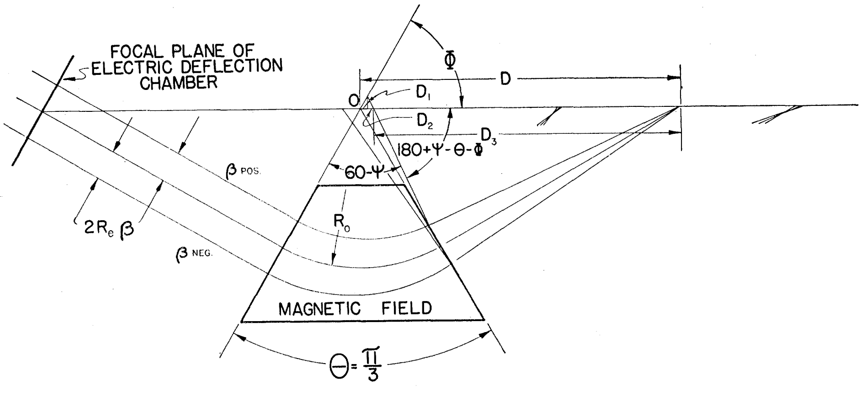

FIG. 4. $\pi/\sqrt{2}$ electric and $\pi/3$ magnetic deflections combined. Not to scale.

width to the desired accuracy. An exact expression for these quantities, however, is easily written down. Using the designations given in Fig. 4 the distance D from the origin O to the point of intersection on the recording plate has been calculated for ions of different masses using values of β equal to $+0.005$, 0, and -0.005 corresponding to a 2 percent range in energy of the focused ions.

$$D=D_1+D_2+D_3=R_0\{(M/M_0)^{\frac{1}{2}}(1-\beta)-1+2\beta\}\times\cos\Phi+\frac{R_0\{(M/M_0)^{\frac{1}{2}}(1-\beta)-1+2\beta\}\sin\Phi}{\tan(180°+\psi-\theta-\Phi)}+\frac{R_0}{\cos(180°+\psi-\theta-\Phi)}\left[(M/M_0)^{\frac{1}{2}}(1-\beta)-\frac{\{(M/M_0)^{\frac{1}{2}}(1-\beta)-1+2\beta\}\sin\Phi}{\sin(180°+\psi-\theta-\Phi)}\right]\quad(1)$$

$$\text{and }\sin\psi=\frac{\{(M/M_0)^{\frac{1}{2}}(1-\beta)-1+2\beta\}\sin\theta}{(M/M_0)^{\frac{1}{2}}(1-\beta)}.\quad(2)$$

These equations are for the general case and reduce to those already given for the restricted 180° case when the appropriate values of Φ and θ are substituted.

Table I gives the values of D when the appropriate values $\beta=\pm0.005$, $\Phi=\theta=60°$, and $M=0.6$ to $1.4M_0$ are substituted in Eqs. (1) and (2). When the degree of velocity focusing and linearity of scale attained are compared with those obtainable for 180° magnetic deflection, the immediate advantages of a magnetic deflection of less than π radians are apparent. Also, if the ions are deflected through a mean angle of only $\pi/3$ radians, the radius of curvature for a given dispersion is only one-half that for 180° focusing and a magnet of very much smaller dimensions can be used.

TABLE I. *Trace widths and line positions for 180° and 60° magnetic deflection compared for equal mass dispersion at the center of the recording plate. Energy band width 2 percent.*

		180° Magnetic Focusing		60° Magnetic Focusing			
Mass	Position on Linear Mass Scale	Position $\beta=0$	Total Trace Width $\beta(\pm0.005)$	Total Trace Width $\pm\beta$	Position $V>V_0$ $\beta=-0.005$	$V=V_0$ $\beta=0$	$V<V_0$ $\beta=0.005$
$0.6M_0$	$1.2R_0$	$1.0984R_0$	$0.009R_0$	$0.0019R_0$	$1.1876R_0$	$1.1885R_0$	$1.1895R_0$
0.65	1.3	1.2249	.0078	.0012	1.2934	1.2939	1.2946
0.7	1.4	1.3466	.0065	.0008	1.3970	1.3974	1.3978
0.8	1.6	1.5777	.0042	.00026	1.6005	1.6003	1.6003
0.9	1.8	1.7947	.0021	.00006	1.8004	1.8004	1.8004
1.0	2.0	2.0000	.0000	.0007	2.0007	2.0000	2.0007
1.1	2.2	2.1952	.0019	.0004	2.2008	2.2009	2.2012
1.2	2.4	2.3818	.0038	.0012	2.4039	2.4043	2.4050
1.3	2.6	2.5607	.0056	.0023	2.6106	2.6116	2.6129
1.35	2.7	2.6476	.0064	.0028	2.7157	2.7169	2.7185
1.4	2.8	2.7329	.0073	.0038	2.8219	2.8236	2.8257

Magnetic focusing of divergent beams

The ions entering the electric deflection chamber from S_2 as a divergent beam are brought to convergence at the focal plane after a deflection through $\pi/\sqrt{2}$ radians. The width of the image of S_2 in the focal plane, considering solely the breadth produced by the divergence of the ions, is equal to $4/3\alpha^2R_e$. α is the half-angle of divergence equal to the sum of the widths of slits S_1 and S_2 divided by twice their separation.

The ions leaving the focal plane of the first section of the spectrograph again diverge and enter the region of uniform magnetic field intensity between the pole pieces of the magnet. Barber[7] has shown that if the central beam of a divergent bundle of ions enters and leaves the field normal to the edges of the poles, the ions are brought to a focus on the line extended from the source through the center of curvature of the central beam. In the mass spectrograph one beam of ions fulfills this condition exactly, that for which $R_{M_0}=R_{\text{mag}}=R_e$. The width of the trace is $\alpha^2R_{M_0}=2.5\times10^{-4}$ cm where $\alpha=1/360$ for slits of width $S_1=0.13$ mm and $S_2=0.1$ mm separated 4.2 cm. If the divergent ions were not focused at the recording plate the width of the trace would be $1.1\text{ cm}=2\alpha L/\sin\nu$, where L is the path length of the ions from their point of divergence and ν is the angle between the plate and the direction of motion of the incident ions. Figs. 5, 6 and 7 illustrate spectra taken using slits of the width given above. The center of the recording plate is tangent to the locus of the foci of ions of mass greater and less than M_0, the locus lying below the plate.

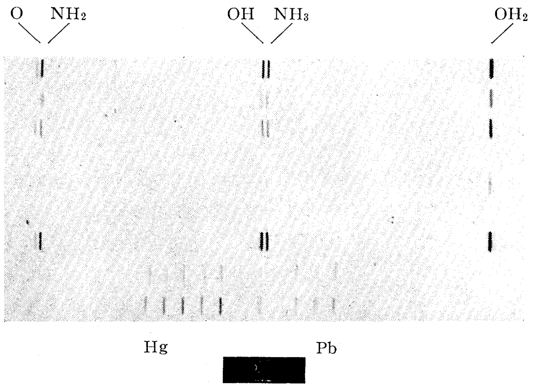

FIG. 5. Natural size. Spectra of matched $OH-NH_3$ doublets and the isotopes of mercury and lead, $S_1=0.13$ mm, $S_2=0.1$ mm, $S_3=5$ mm. The black rectangle represents the width of a single trace for the same slit dimensions but without magnetic refocusing of divergent beams of ions.

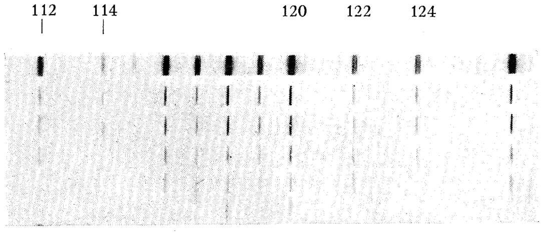

FIG. 6. Natural size. Doubly ionized tin. The magnetic field changed while the top spectrum was being photographed giving the effect of double width lines. On the original plate Sn^{115} appears on the first four spectra but the reported Sn^{121} is not present at all.

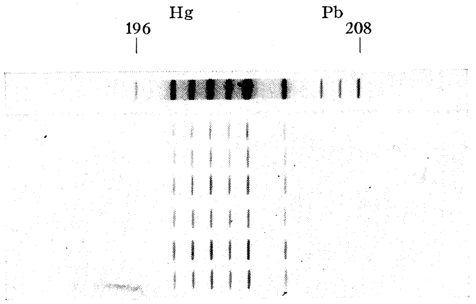

FIG. 7. Natural size. An overexposed spectrum of mercury, and lead, showing the mercury 196 line and comparison spectra from the same plate. An abundance range of 60,000/1 can be recorded and measured.

Good focusing of divergent ion beams is secured over the center of the plate for the high intensity spectra (wide slits close together) used in the analysis of the isotopic constitution of elements and in the search for rare isotopes, Figs. 5, 6, 7. The magnitude of α in the present instrument may appear small, but it is about 5 to 30 times greater than that which has been used in the earlier types of precision photorecording mass spectrographs not utilizing angle focusing.[8, 9, 10] The slit widths are 2 to 5 times greater and the distance between slits has been reduced by one-third to one-tenth. The resolving power $M/\Delta M$ achieved for wide slits is between 1500 and 2000 representing an increase of 3 to 10 times.

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[7] N. F. Barber, Proc. Leeds Phil. Soc. **2**, 427 (1933); W. E. Stephens and A. L. Hughes, Phys. Rev. **45**, 123 (1934); W. E. Stephens, Phys. Rev. **45**, 513 (1934).

[8] J. Costa, Ann. d. Physik **4**, 425 (1925).

[9] F. W. Aston, Proc. Roy. Soc. **A115**, 487 (1927).

[10] K. T. Bainbridge, Phys. Rev. **40**, 132 (1932).

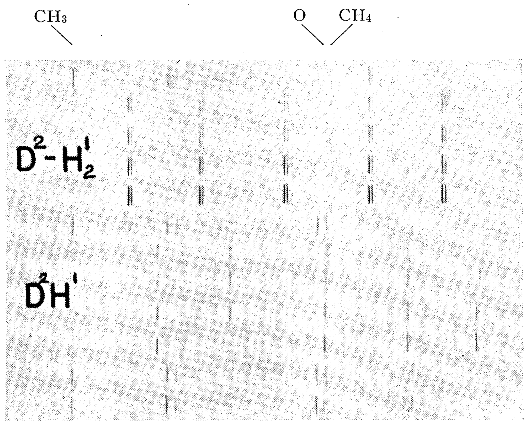

FIG. 8. $O-CH_4$, $D^2-H^1_2$ doublets, and D^2H lines, natural size, for slit widths $S_1=0.06$, $S_2=0.02$, $S_3=1.25$ mm.

As the trace width for magnetic focusing varies as α^2 a reduction of the collimating slit width to values comparable to past usage aids in making the attainment of exceptionally high resolving power entirely practicable.[11]

Resolving power

The factor responsible for a large part of the width of a trace at the recording plate is the nonhomogeneity in energy of the ions which pass through each point along the focal plane of the electric deflection chamber. In the focal plane an energy spectrum of ions of all m/e values is formed of overlapping images of the slit S_2. The width of an image $=S_2+4/3\alpha^2R_e=S_2$, as the second term is negligible in comparison to the slit width. At any point in the focal plane the maximum spread in the velocity of ions of all m/e values passing through the same point is given in terms of $\beta=(V_0-V)/V_0=S_2/2R_e$. When the values of S_2 and R_e are substituted, 0.1 mm and 25.4 cm respectively, the maximum value of β is 1/5000. Consequently ions leaving a single point along the focal plane have all radii of curvature between R and $R(1+\beta)$, so the maximum resolving power $R/\Delta R=M/\Delta M=V/\Delta V=5000$ under the geometrical conditions defined above. When the trace width assignable to imperfect velocity and direction focusing is considered,

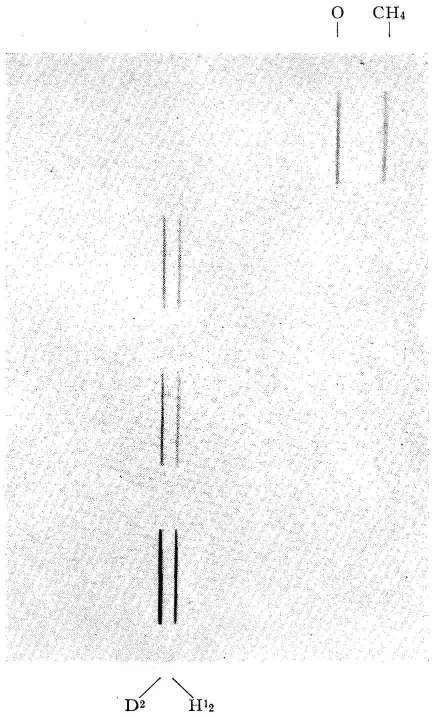

FIG. 9. A portion of Fig. 8 enlarged 5×. $O-CH_4$ and $D^2-H^1_2$ doublets.

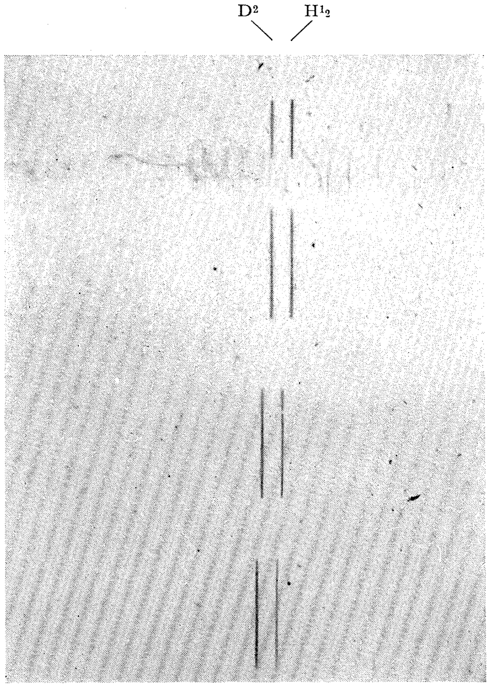

FIG. 10. Some $D^2-H^1_2$ doublets enlarged 5×. $S_1=0.03$, $S_2=0.017$, $S_3=1.0$ mm. Resolving power 10,000 to 15,000.

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[11] The resolving power is defined as the ratio $M/\Delta M$ for *complete* separation of two lines and so is more stringent than the optical definition. A standard value for the density of the traces on Q plates should be included in the definition. The value 0.3 is suggested.

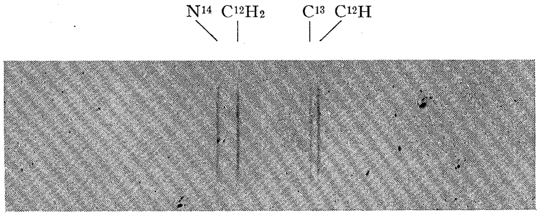

FIG. 11. $N-CH_2$, $C^{13}-C^{12}H$, doublets. Enlarged 5×.

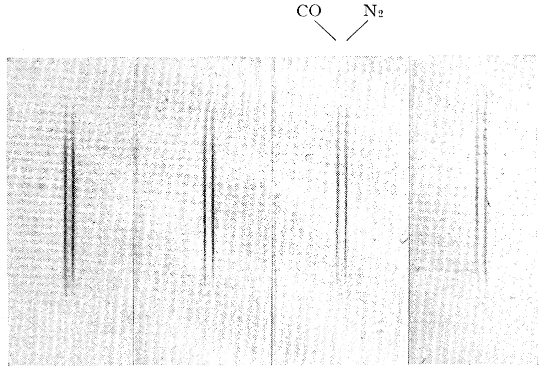

FIG. 12. $CO-N_2$ doublets taken without a diaphragm in front of the recording plate. Enlarged 5×. The ions differ in mass by 1/2500. $S_1=0.04$ mm, $S_2=0.017$ mm, $S_3=1$ mm.

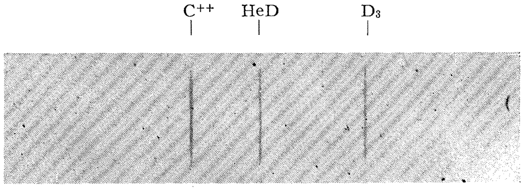

FIG. 13. $C^{++}-HeD^2-D^2_3$ matched triplet. Enlarged 5×.

the maximum realizable resolving power $=1850$ for values $S_1=0.13$ mm, $S_2=0.1$ mm, $S_3=5$ mm. The spectra reproduced in Figs. 5, 6, 7 show that the resolving power deduced from simple theoretical considerations has been attained in the completed instrument. Experimentally it was found that for the optimum focus the photographic plate must be approximately 0.68 cm above the plane indicated by theory. A practical advantage of great value results when the plate is in this position as experimentally one finds that the traces are symmetrical and that the position of the traces is nearly independent of the density. Both of these features are an aid to accurate mass measurements, the primary aim in the construction of this mass spectrograph.

Once the spectrograph had been satisfactorily aligned and adjusted using wide slits and the analysis of a series of elements had been secured,[12] experiments were undertaken with the object of obtaining a resolving power greater than 10,000. The spectra shown in Figs. 8, 9 were taken with $S_1 \leqq 0.06$, $S_2=0.02$, $S_3=1.25$ mm yielding a resolving power of 6000 to 8000. The $D^2-H_2{}^1$ doublets which had never been resolved completely before are well separated even at high trace densities. The duration of the exposures varied from 1 to 5 seconds for the hydrogen spectra. The oxygen-methane doublets are 20 second exposures.

The final slit adjustments now used for routine mass measurements are $S_1=0.03$, $S_2=0.017$, $S_3=1.0$ mm, permitting a resolving power of 10,000 to 15,000 and exposure times of from 2 or 3 seconds for the hydrogen deuterium doublet and 60–180 seconds for the $C^{13}-C^{12}H^1$ doublet. The rare carbon isotope, for several spectra, has been matched in intensity with $C^{12}H^1$ by running a discharge in CO with a trace of methane. Enlarged spectra of various doublets are shown in Figs. 10, 11, 12, 13. Fig. 14 is a densitometer record of a $D^2-H_2{}^1$ doublet. The mass separation of $D^2-H_2{}^1$ is approximately equal to $2\frac{1}{2}$ times the electronic mass and the width of the traces is only 0.00015 mass unit.

The ion source

The ions are produced in the low pressure discharge tube *A* which is usually run at 20,000 volts. The power for the discharge tube is furnished by a full wave rectifier set, generally operated without smoothing condensers. On those occasions when a filtered source of high potential is desired two 0.5 mfd 25 kv condensers in series may be connected. Steadier accelerating potentials for special work are available from the laboratory 100 kv storage battery.

Gas is admitted to the discharge tube under the cathode as shown in Fig. 2. The iron base of the tube supports an aluminum cathode of the Thomson type. This base has been so machined that a spherical discharge tube with a narrow neck which just clears the cathode may also be used.

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[12] Described in the second part of this paper.

Ions of metallic elements are obtained in general by introducing a quantity of the element to be investigated, or a halide salt, into the cathode and running a discharge in neon. Under these conditions a copious supply of singly and doubly ionized metallic ions is formed.[13] In the case of tellurium and indium small beads of these elements were placed in a circular array of holes drilled in an aluminum cathode. Iodides of germanium and tin were used in the same type of cathode. A bismuth cathode was used directly, and lead and cadmium cathodes containing their respective iodides furnished ions of these metals. Zinc, selenium, beryllium, aluminum, and iron have yielded ions by the same general method.[14, 15] Shielding the side of the discharge tube with a glass cylinder as indicated in Fig. 2 is advantageous in preventing the formation of arcs between the cathode and the tube walls.

Experience with another type of mass spectrograph showed that a discharge operated at approximately 15 kilovolts gave a nice balance between high ion current density and the high energy useful in securing satisfactory photographic records.[16] At the same time a discharge at 10–20 kilovolts can be operated at pressures which permit a high concentration of the vapor under investigation in the tube, with a low partial pressure of undesirable contaminating gases.

In the low pressure discharge run at 15–20,000 volts used with the mass spectrograph, the cathode fall space extends for 15 to 25 cm and the positive column is absent or localized just at the anode. The position of the metal or salt which is used to furnish ions is not critical. Substances of high vapor pressure ($P > 10^{-3}$ mm at 200°C) may be placed on the iron base of the discharge tube, as has been done successfully for selenium, beryllium chloride, and the bromide and iodide of cadmium. Tin iodide works well if it is evaporated into the discharge from a side tube. The ions actually recorded on the plate have originated not in the immediate region of the cathode but in the vicinity of the negative glow and along the axis of the tube where the electron density is greatest.

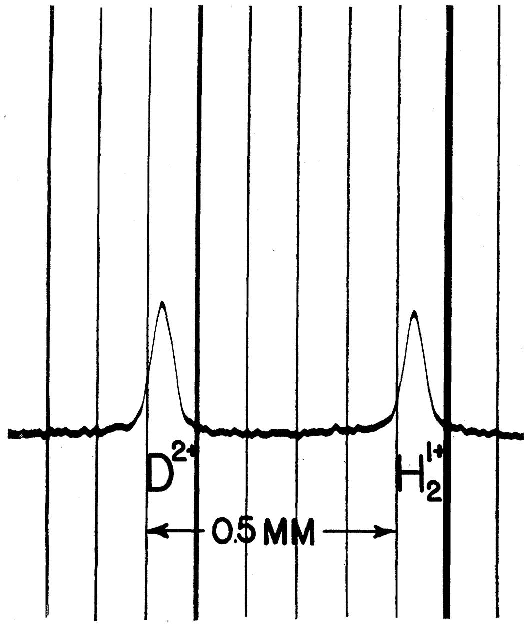

FIG. 14. A matched $D^2-H^1_2$ doublet. Microphotometer slit width 0.025–0.030 mm.

The addition of neon serves to stabilize the discharge and to produce a controlled evaporation of the substance struck by the neon ions. The energy of the incident ions has been sufficient to melt a bismuth cathode and a sheet of cerium placed in poor thermal contact with an aluminum cathode. After a zinc or cadmium cathode has been used for some time the crystalline structure of the metals is brought out as if local evaporation and sputtering had etched the surface in the area of maximum intensity of the incident ions.

The dissociation and ionization of the heterogeneous vapor of the substance evaporated from the cathode probably result from the action of the high speed electrons which have their maximum density along the axis of the tube. The intensity of the central electron beam is so great that unless special care is taken the wall of the pyrex discharge tube may be punctured by local overheating. (A hole one or two millimeters in diameter can be neatly drilled in the end wall by the electrons.) As the ions proceed towards the cathode against this stream of electrons, it is probable that the multiply charged ions are formed in greatest abundance along the axis of

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[13] K. T. Bainbridge, Phys. Rev. **39**, 847, 1021 (1932).

[14] K. T. Bainbridge, Phys. Rev. **43**, 1056 (1933).

[15] H. Lukanow and W. Schutze, Zeits. f. Physik **82**, 610 (1933).

[16] K. T. Bainbridge, J. Frank. Inst. **215**, 509 (1933).

the tube, although some may result from collisions of the second kind with neon ions.

The collimating slits

The ions from the discharge tube are collimated by the slits S_1 and S_2 and enter the chamber D, Fig. 1. The slits, which are 0.1 to 0.15 mm in width for work at high intensities, are separated 4.2 cm. By this arrangement they subtend a larger solid angle than is usual in the collimating system of a photo-recording mass spectrograph, with a corresponding gain in the sensitivity of the instrument.

The first slit, S_1 is formed by a pair of adjustable iron jaws placed in a slot in the discharge tube base directly under the aluminum cathode. S_2 is made of strips of hardened steel screwed to a supporting brass cylinder. The adjustment or removal of the second slit is accomplished through the port, P, Figs. 1 and 2.

The region between the slits is evacuated to isolate the low pressure deflection chamber from the higher pressure region in the discharge tube.

The energy selector

The diverging beam of ions, heterogeneous in energy and m/e values, leaves S_2 and enters the region between two accurately figured cast bronze deflecting plates. The 1.9 cm separation of the plates permits ions differing by 4 percent in energy, diverging in an angle of 1/50, to be focused in front of S_3, after deflection through $\pi/\sqrt{2}$ radians. This wide separation also helps to eliminate the effects of possible surface charges on the plates.

The radius of the central ion path is 25.4 cm.

The magnitude of the deflecting voltage V_d, is given in terms of the accelerating voltage V_a by the equation $V_d=0.16V_a$, as $mv^2/R=2V_ae/R=V_de/R\log b/a$ and the radii of the deflecting plates a and b are 24.45 cm and 26.35 cm respectively. Ions of 15,000 electron-volts energy are deflected by the required amount in the energy selector when the circular plates are connected 1200 volts above and below ground. The auxiliary deflecting plates illustrated in Fig. 2 are used to secure small angle adjustments of the deflected ion beams. It was found experimentally that these plates had to be at three-fourths of the potential of the main deflecting plates in order to have the central beam of ions proceed exactly along the center of the tube connecting the energy selector and the magnet.

Fifty-four large capacity 45-volt radio batteries furnish the potential for the deflecting plates.

The slit S_3 is a brass plate with a 10 mm slot cut in it. In practice 7.5 mm was found to be the maximum useful width of S_3, since any increase beyond 7.5 mm adds little to the intensity and brings about an objectionable decrease in the resolving power.[17] For adjusting S_3, two tapered ground joints which hold thin brass flags, Fig. 2, either of which may be turned to cover the slit completely, have been added since the photograph Fig. 1 was taken. By adjusting both of the flags it is possible without breaking the vacuum to select ions of a band of energies of any width up to the maximum spread of 4 percent incident on S_3.

Since the photograph Fig. 1 was taken a window has been provided at DE, Fig. 1, so that S_3 may be inspected visually. The plate in the vicinity of the slit has been coated with willemite, which fluoresces brightly under the impact of the incident positive ions and provides a convenient check on the proper adjustment of the discharge tube.

Freedom from surface charge effects

A definite practical advantage results from the use of deflector plates which are relatively far apart and are maintained at high potentials compared to those of the deflecting plates in other types of mass spectrographs.

Under intense ion bombardment surface films attaining a potential of 1 volt on the deflecting plates have been reported. With small separations the charging of surface films on these plates acts to distort the shape and alter the positions of the traces of the ion beams which are incident on the recording plate.[18, 19] The presence of such effects not only decreases the theoretically attainable resolving power of an instrument, but also makes it difficult to compare accurately the relative positions of the lines. It was partly to overcome these difficulties that the present instrument was designed with deflector plates separated by 1.9

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[17] The cadmium spectra, one of which was used for Fig. 16, were taken with $S_1=S_2=0.25$ mm, $S_3=10$ mm.

[18] F. W. Aston, Proc. Roy. Soc. **A115**, 495 (1927).

[19] A. J. Dempster, Proc. Am. Phil. Soc. **75**, 764 (1935).

cm, to be maintained at a high difference of potential.

With the resolving power adjusted to 2000, observations were made through a 10 power microscope of the fluorescence produced by the ion beams, H, H_2, CH_4, on a willemite screen placed in the same position as the usual recording plate. Under these conditions fluctuations of one part in 2000 in the positions of the lines, caused by changes in the magnetic or electric fields, would have appeared in the microscope as a 2.5 mm shift of the ion beam. No motion of the ion beam could be detected. Spectra taken under different conditions with varying lengths of exposure at resolving powers from 400 to 12,000 show definite sharp edges to the traces.

The potential of semiconducting surface films can be lowered if the charging current per unit area is reduced. One reason for the freedom from surface charges on the deflector plates of this mass spectrograph is that with the large separation and area of the plates, even though the beam issuing from S_2 is very intense, the density of the ions striking the deflector plates is low.

The excellent vacuum conditions in the spectrograph also hinder the formation of surface films on the deflector plates. The gas pressure during a series of exposures is 10^{-6} mm, as measured by an ionization gauge. The sharpness of the lines formed on the recording plate by ions which have traveled more than a meter and a half through the mass spectrograph demonstrates the high vacuum maintained for routine work.

If 0.1 volt surface potentials should appear on the plates a shift in the positions of the recorded lines of only one part in 25,000 would result. No distortion of the shape of the lines can occur when the slits S_1, S_2 and S_3 are adjusted for high resolving power, as the field distribution in the central region between the deflector plates remains uniform even if the area struck by ions incident on the plates becomes charged.

The magnet

The magnet is constructed of two quadrants and a straight section of Armco iron which after machining were properly annealed to insure high permeability at high flux densities. The magnet is wound with approximately 5000 turns of number 12 B. & S. D.C.C. enameled wire, the resistance of the winding being 24 ohms. The heaviest ions examined are brought to the center of the plate with an expenditure of only six watts in the coils. Four 90 ampere-hour six volt automobile batteries connected in series-parallel supply the exciting current for the magnet.

The pole pieces are separated by 0.32 cm brass strips shown in Fig. 2. The top area of the pole faces is open to a depth of 3.5 cm for convenience in measuring the field with a search coil and Grassot fluxmeter.

Allowance for the deflection of the ions in the stray field of the magnet was made from calculations of the integrated effect of the field.[20] The edges of the poles were cut back approximately $1\frac{1}{2}$ gap widths. A still higher order correction can be obtained by a small angular rotation of the electric deflection chamber and the recording plate, but the excellent performance of the mass spectrograph does not require it at this time.

The recording plates and camera

All of the mass spectra secured have been recorded on "*Q*" plates which were developed by the Ilford Laboratories.[21, 22] Two types of the *Q* emulsion have been used.[23] There is available one of exceptionally high sensitivity to positive rays which is particularly suited for the detection of isotopes of low abundance and for the photometry of mass spectra. Isotopes with an abundance ratio of 60,000/1 can be recorded without any indication of solarization of the denser trace. Fig. 15 shows a graph of the density-abundance

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[20] The senior author is indebted to Dr. L. H. Rumbaugh of the Bartol Research Foundation for helpful suggestions concerning the stray magnetic field correction.

[21] F. W. Aston, Proc. Roy. Soc. **A132**, 487 (1931).

[22] Ilford Ltd., J. Sci. Inst. **12**, 333 (1935).

[23] An investigation was made of the sensitivity of various plates by means of a parabola apparatus (J. J. Thomson, Phil. Mag. **13**, 561 (1907); R. Conrad, Physik. Zeits. **31**, 888 (1930)) in which five plates mounted on a hexagon could be brought rapidly into the correct position for exposure so that each plate might receive the same total exposure and fluctuations in the discharge would cancel out. Exposures were for twenty periods of 15 seconds each taken in rotation. Plates sensitized to the infrared by dyes did not respond to positive ions. The Eastman Kodak Process and ultraviolet sensitized plates were more sensitive than the Paget halftone plates previously favored for recording positive rays. None of these plates, however, could compare with the *Q* plates in sensitivity. Samples of these plates have been tested in the far ultraviolet at this laboratory by Dr. H. M. O'Bryan and at the M. I. T. spectroscopy laboratory by Professor J. C. Boyce. The plates are sensitive over the whole of the region tested from 140A to 3000A.

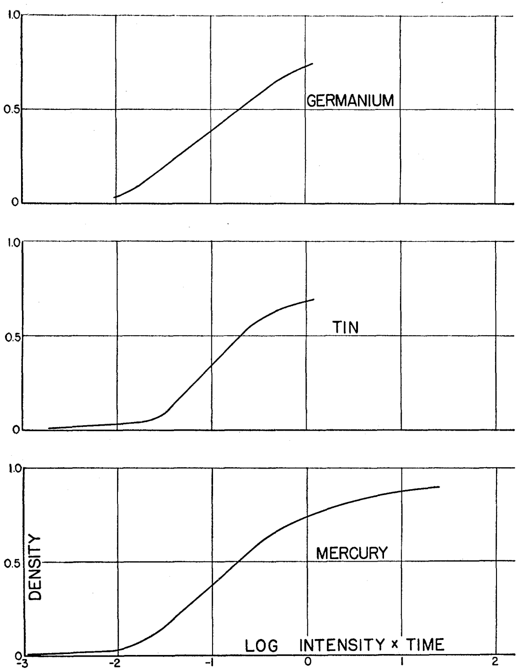

FIG. 15. Characteristic curves of *Q* plates for germanium, tin and mercury ions.

characteristic of the emulsion for 20,000 electron volt tin ions incident on the plate, and 10,000 electron volt germanium and mercury ions. With Paget halftone plates the density at the peak of the linear position of the curve seldom exceeds the value 0.2. With Schumann plates values up to 3.0 are easily attained but the slope of the density intensity graph is very steep.

An exceptionally fine grain *Q* emulsion of good sensitivity is also available which is particularly suited for the mass spectra including close doublets. Figs. 8, 9, 10, 11, are reproductions of spectra taken with this type of emulsion and Fig. 14 is a densitometer record of a $D^2-H_2^1$ doublet. The $CO-N_2$ doublets, Fig. 12, were recorded on the more sensitive larger grained *Q* plates.

The *Q* plates are particularly pleasing to work with because of their uniformity, and they are free from chemical fog even when developed two or three times the normal period in a hydroquinone-caustic soda developer (E. K. Co., D–9).

In the mass spectrograph the recording plate, 2 inches wide and 16 inches long, is supported on a cast bronze carriage. Contact with a lapped plane surface is made over the entire length of the ends and edges of the plate, which is held in position by eight springs. The carriage may be moved parallel to itself by a rack and pinion arrangement, permitting twelve exposures of spectral lines about 3 mm in height. The position of the plate relative to the mass spectrograph is marked by projecting the reduced image of a slit on the plate as illustrated in Fig. 2. The fiducial marker returns to the same position after a new plate is inserted in the spectrograph, as the cover supporting it is accurately aligned by two hardened steel pins.

The large brass stopcocks illustrated in Fig. 2 permit plates to be changed without letting air into the electric deflection chamber and discharge tube or into the mercury diffusion pump which evacuates the camera chamber.

2. THE EXISTENCE OF ISOBARS OF ADJACENT ELEMENTS

Of the 250 known nuclear species[24] there are some of the same mass number but different atomic number which are called isobars.[25]

The members of one class of these isobaric pairs differ by two units in nuclear charge. Heisenberg,[26] Gamow,[27] and Goeppert-Mayer[28] have presented theoretical arguments which support the experimentally observed fact that both members of some 40 of these pairs are stable or, in other words, that no capture or loss of two electrons occurs which would reduce the pairs to a single isotope.

There is a second important class of isobars, those pairs of the same mass number which differ by one unit in nuclear charge. Approximately 16 pairs of this class have been reported, some of which have one member which is definitely known to be unstable. For example K^{40} and Rb,^{87} which are paired with Ca^{40} and Sr^{87} respectively, transmute into the latter elements by the emission of beta-rays.[29] One of the isotopes of

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[24] O. Hahn, Ber. d. D. Chem. Ges. **A69**, 5 (1936).

[25] The term *isobar* originally suggested by Stewart (Phil. Mag. **36**, 326 (1918)) was altered to *isobare* to avoid confusion with the metereological term!

[26] W. Heisenberg, Zeits. f. Physik **78**, 156 (1932) and Scott Lectures, Cambridge Univ. 1934.

[27] G. Gamow, 1934 Int. Conf. Physics, **1**, 60 (1935).

[28] M. Goeppert-Mayer, Phys. Rev. **48**, 512 (1935).

[29] G. von Hevesy and W. Dullenkopf, Zeits. f. anorg. Chemie **221**, 167 (1934); G. P. Baxter and C. M. Alter,

neodymium may also belong in this category.[30] Mattauch[31] and Sitte[32] concluded from a study of the known atomic species that for adjacent elements there are no isobaric pairs of which both members are stable. This conclusion is in agreement with the results of Heisenberg's discussion of the stability of nuclei with respect to beta-ray emission.

The work described below was undertaken to determine experimentally whether stable isobaric pairs do exist for elements differing by one unit in atomic number. The type of ion-source previously described permitted the use of hydrogen-free compounds of the elements under investigation. In this way successful analyses were made of elements which previously had been introduced into the discharge tube only in chemical combination with methyl and ethyl radicals.

The elimination of hydrides in the discharge tube does away with two types of error in isotopic analysis. The addition of hydrogen to an isotope results in another line in the mass spectrum which may be mistaken for an isotope, and the measurement of the relative abundance of actual isotopes may be invalidated by superimposed hydrides.

Isotopic analysis

The first element investigated was tellurium ($z=52$). First and second order spectra gave further proof of the existence of Te^{123} which is isobaric with $Sb^{123}(z=51)$.[33] No Te^{127}, isobaric with $I^{127}(z=53)$, was present to more than 0.8 percent of the total. Tellurium was analyzed before any iodides had been introduced into the discharge tube so that a test could be made for the presence of Te^{127} without the danger of confusion with I^{127}.[33]

Indium ($z=49$) gave two isotopes of mass number 115 and 113 in agreement with the results of Wehrli and Miescher,[34] and Aston.[35] In^{115} is a member of the isobaric triplet reported by Aston[18, 36, 37] in which he includes the isotopes $_{48}Cd^{115}$ and $_{50}Sn^{115}$. In^{113} is isobaric with Cd^{113} which is one of the more abundant isotopes of this element.

Cadmium was analyzed to test the reality of the $In^{115}-Cd^{115}$ and $In^{113}-Cd^{113}$ isobaric pairs. A microphotometer record of cadmium is shown in Fig. 16 where the black line above the figure 115 illustrates the amount of Cd^{115} reported by Aston.[36] No evidence was found for its existence to $\frac{1}{8}$ of this amount. Dempster[18] has also reported an analysis of cadmium in which Cd^{115} does not appear, without giving an upper limit for the abundance. Cd^{118}, found from band spectrum analysis[38] to be more abundant than Cd^{106} and Cd^{108}, does not appear.

Tin was examined for the purpose of investigating the isobaric pairs $_{50}Sn^{115}-{_{49}In^{115}}$ and $_{50}Sn^{121}-{_{51}Sb^{121}}$. Fig. 6 shows a series of four spectra of doubly ionized tin which with some of singly ionized tin gave proof of the existence of Sn^{115}, which appeared on all of the spectra, but failed to show Sn^{121}, although from the photometric records of these spectra 1/60 of the amount reported by Aston[18, 36] would be easily detectable. The appearance of a line of mass 121 which accompanied the tin isotopes in Aston's spectra is difficult to explain. If this line were a hydride of the most abundant isotope Sn^{120} it would indicate 11 percent hydride contamination and the corresponding hydrides of Sn^{122} and Sn^{124} should have been easily detected as their abundance would exceed that of the real isotope Sn^{115}. If the line of mass 121 was not a hydride but a fortuitously occurring molecular ion there arises the difficulty of accounting for the consistency in the measurements of its apparent abundance on successive spectra taken under different conditions.

Mercury was investigated for the purpose of examining Hg^{197} and Hg^{203} to which values have been assigned by their discoverer[39] of *ca.* 0.01 and 0.006 percent abundance. $_{80}Hg^{197}$ would be isobaric with $_{79}Au^{197}$ and $_{80}Hg^{203}$ forms a pair

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J. Am. Chem. Soc. **55**, 3270 (1933); O. Klemperer, Proc. Roy. Soc. **A148**, 638 (1935); A. O. Nier, Phys. Rev. **48**, 283 (1935).

[30] W. F. Libby, Phys. Rev. **45**, 845 (1934).

[31] J. Mattauch, Zeits. f. Physik **91**, 361 (1934).

[32] K. Sitte, Zeits. f. Physik **96**, 512 (1935).

[33] K. T. Bainbridge, Phys. Rev. **39**, 1021 (1932). Te^{127} was previously reported as a doubtful isotope.

[34] M. Wehrli, Naturwiss. **22**, 289 (1934); M. Wehrli and E. Miescher, Helv. Phys. Acta **7**, 298 (1934).

[35] F. W. Aston, Proc. Roy. Soc. **A149**, 396 (1935).

[36] F. W. Aston, Proc. Roy. Soc. **A130**, 302 (1931).

[37] F. W. Aston, Phil. Mag. **49**, 1191 (1925).

[38] E. Svensson, Nature **131**, 28 (1933); G. Stenvinkel and E. Svensson, Nature **135**, 955 (1935).

[39] F. W. Aston, Nature **130**, 847 (1932).

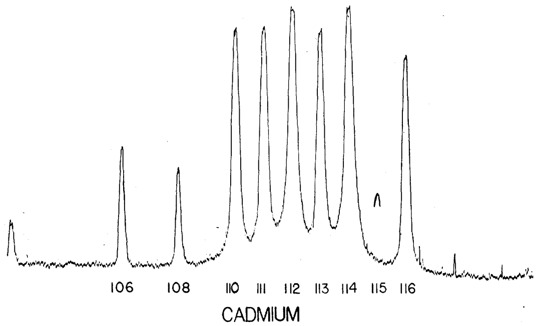

FIG. 16. Densitometer record of a spectrum of cadmium taken with $S_1=0.25$ mm, $S_2=0.25$ mm, $S_3=10$ mm.

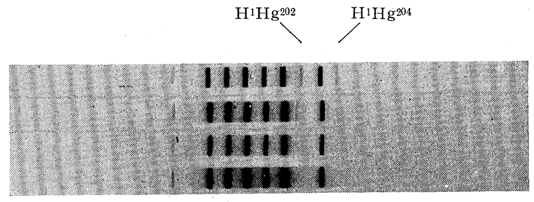

FIG. 17. Spectra of mercury showing hydrides of Hg^{202} and Hg^{204}. Successive spectra were taken with decreasing amounts of water vapor in the discharge tube.

with the $_{81}Tl^{203}$ isotope.[40, 41, 42] No Hg^{197} was found to $\frac{1}{2}$ of the abundance reported although this amount could easily have been detected. The presence of the line of mass 197 previously reported cannot be attributed to a hydride of Hg^{196} as the low abundance of this isotope, 0.1 percent, would require a 10 percent hydride effect which was not apparent for the Hg^{202} or Hg^{204} isotopes. Sitte's[32] suggestion that Hg^{201} does not exist has absolutely no foundation in fact. A definite answer concerning Hg^{203} is not possible at this time as second order spectra of mercury of sufficient intensity to record a line of abundance 1/20,000 or less were not taken and in the first order a hydride $Hg^{202}H^1$, present to even 1/5000 of the abundance of Hg^{202}, invalidates any attempts to examine Hg^{203}. Hydrides of mercury do form and appear to be dependent more on the concentration of water vapor in the discharge tube than upon the partial pressure of any hydrogen which may be present. Fig. 17 shows a series of four spectra of mercury taken with a new discharge tube in which water vapor was freely given off from the glass walls when the tube was first used. The decrease in the intensity of the hydride lines at 197, 203 and 205 corresponding to lower water vapor concentration in the discharge is apparent for successive spectra. The addition of molecular hydrogen directly to a discharge in mercury vapor does not produce hydrides of the same high intensity.

In addition to the most abundant isotopes of lead Pb^{206}, Pb^{207}, and Pb^{208} found by mass spectrum analysis,[43] and Pb^{204} from work on hyperfine structure,[44] Aston[45] has reported small amounts of Pb^{203}, Pb^{205}, Pb^{209} and Pb^{210}. $_{82}Pb^{209}$ which is isobaric with $_{83}Bi^{209}$ is the most abundant of these new isotopes and was reported present to 0.85 percent. No line of mass 209 appeared on our spectra, although 1/10 of the abundance previously reported could easily have been detected. Fig. 7 shows one of the spectra of lead, and another taken with phosphine in the discharge tube is shown in Fig. 5. Aston's figures for the abundance of Pb^{205} and Pb^{209} were obtained from spectra definitely contaminated by lead hydride which by his measurement was present to 2.3 percent. As we could find no line of mass 209 our results indicate that the actual abundance of lead hydride was probably 4 percent (an increase of 1.7 percent) and the lines of mass 205 and 209 present to 0.02 of 204 and 0.017 of 208 on Aston's spectra were hydrides of Pb^{204} and Pb^{208}.

Bismuth was analyzed and appears to consist of a single isotope 209 with no other present to more than 2 percent. This result is in agreement with the value for the chemical atomic weight.

The superior qualities of Q plates for photometric work made it advisable to repeat the examination of germanium which had been made with another instrument using Schumann plates. The new spectra confirmed the previous results,[46] showing five isotopes 70, 72, 73, 74 and 76.

Photometric measurements

Upper limits were set for the abundance of the rare or non-existent isobars by photometric analysis of the mass spectra.

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[40] A. J. Dempster, Nature **136**, 65 (1935).

[41] S. Imanishi, Inst. Phys. and Chem. Res., Tokyo, Sci. Papers **28**, 129 (1935).

[42] H. Schüler and J. E. Keyston, Naturwiss. **19**, 320 (1931).

[43] F. W. Aston, Nature **120**, 224 (1927).

[44] H. Schüler and E. G. Jones, Naturwiss. **20**, 171 (1932). Zeits. f. Physik **75**, 563 (1932).

[45] F. W. Aston, Proc. Roy. Soc. **A140**, 535 (1933). Nature **129**, 649 (1932).

[46] K. T. Bainbridge, Phys. Rev. **43**, 1056 (1933); J. Frank. Inst. **215**, 509 (1933).

The plates were first calibrated by making a series of exposures of different duration while the discharge tube conditions were maintained as constant as possible, and the plate calibration secured in this way was checked against Aston's photometric measurements of the relative abundance of the definitely attested isotopes of the elements investigated. The agreement was satisfactory for the purpose.

The density-log abundance curve was then extended as far as possible taking care that for the longer exposures the density of the traces of the low abundance isotopes should be at least as great as the density of the traces secured at shorter exposures for the isotopes of higher abundance. In a series of exposures of decreasing duration finally there is a point where only a fraction of the isotopes are recorded, the less abundant ones failing to appear against the background. From the photometric curve and the relative abundance of the isotopes determined initially by the method of exposures of different duration, a limit may be found in this way for the minimum detectable amount of an isotope.

The upper limits given in this report for the abundance of apparently nonexistent isotopes were *three times* the "minimum detectable amount" as found from the calibration curves and photometric analyses of the most intense spectra of the elements in the appropriate region for Sn^{121}, Pb^{209} and other isobars under investigation. The upper limits published are considered amounts which could be detected with certainty were the isobars present to these values.

Discussion of results

Six isotopes were found definitely to constitute isobaric pairs: $Cd^{113}In^{113}$, $In^{115}Sn^{115}$, and $Te^{123}Sb^{123}$. The apparent stability of isobars of this class[47] may be accounted for on the neutrino hypothesis introduced by Pauli and Fermi,[48] if the neutrino mass μ is greater than zero as Beck and Bethe have suggested.[49, 50] Yukawa and Sakata have developed a general theory of the stability of adjacent isobars of atomic numbers z and $z-1$.[51] If the difference in the proper energy ΔW of the isobars, $(M_z-M_{z-1})c^2$, is larger than $(m_\beta+\mu)c^2$ the emission of a positron can take place from the nucleus of charge z. If ΔW is smaller than $-(m_\beta+\mu)c^2$ then the isobar $z-1$ can disintegrate with the emission of an electron. In the region of ΔW between $+(m_\beta+\mu)c^2$ and $-(m_\beta+\mu)c^2$, both nuclei could be stable were it not that the isobar of higher atomic number can change by capture of a K electron if ΔW is larger than $-m_\beta c^2(1-\alpha^2z^2)^{\frac{1}{2}}+\mu c^2$, where α is the fine structure constant. The condition for stability is that ΔW must be less than $-(m_\beta+\mu)c^2$ and greater than $-(m_\beta-\mu)c^2$, considering the energy of orbital electrons as simply $m_\beta c^2$. This condition can be satisfied if the mass of the neutrino is greater than zero. In addition ΔW should be small, which is not impossible as the isotopic weight defect curve has a minimum in the region $z=50$,[52] and the masses of isobars could be nearly equal.

Assuming zero neutrino mass, Yukawa and Sakata have calculated the probability of occurrence of the three modes of transmutation for different values of z and of ΔW, for $\Delta W \ngtr +m_\beta c^2$ or $\nless -(1-\alpha^2z^2)^{\frac{1}{2}}m_\beta c^2$. The values obtained for the mean lifetimes of the nuclei, assuming the same spin values for the isobars of a pair, are extraordinarily short, of the order of several hours to a few days for elements in the region $z=50$. But if the members of the isobaric pairs have different spins in every case, the result is a condition of practical stability for the isobaric nuclear species of a pair. It is significant in this connection that there are large variations in spin magnitudes in the region $z=50$,[53] and that the magnetic moment is negative for the odd mass number isotopes Cd^{111}, Cd^{113}, Sn^{117}, Sn^{119}. No measurements have been made of the spins of both members of an isobaric pair of the class considered, owing to the low abundance of Sn^{115}

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[47] Libby, Phys. Rev. **46**, 196 (1934), reports that tin is not β radioactive. Mattauch, reference 2, refers to some unpublished work of G. Kirsch, Rieder and Kara-Michailowa in which antimony failed to show any β activity. Weak α radioactivity from Te has been looked for by Kirsch (ibid.), and indium was examined by J. Schintlmeister, Akad. Wiss. Wien Ber. **144**, 475 (1935), but it is not known if these experiments could have detected a weak β-radiation. We have compared the activity of indium with that of lead free tin by means of a Geiger-Müller screen counter. There is no indication of any radiation from indium.

[48] E. Fermi, Zeits. f. Physik **88**, 161 (1934).

[49] G. Beck, Zeits. f. Physik **91**, 370 (1934); Phys. Rev. **47**, 202 (1935).

[50] H. Bethe, Rev. Mod. Phys. **8**, 198 (1936).

[51] H. Yukawa and S. Sakata, Proc. Phys. Math. Soc. Japan, **17**, 467 (1935).

[52] F. W. Aston, *Mass Spectra and Isotopes*, p. 167.

[53] S. Tolansky, Proc. Roy. Soc. **A144**, 574 (1934).

and Te^{123}. It is hoped that the examination of In^{113} may be possible as this isotope is present to approximately 5 percent of In^{115} and so is a nucleus more amenable to analysis than Sn^{115} or Te^{123}.

The construction of the mass spectrograph was made possible by a grant to Professor Theodore Lyman and the senior author from the Milton Fund. The authors take great pleasure in acknowledging the interest and encouragement of Professor Lyman, the assistance of Professors H. R. Mimno and J. C. Street, the suggestions and aid of Mr. David Mann, who was responsible for the construction of the instrument, and Mr. Leighton's assistance with the glass parts.

We are indebted to Professor H. Bethe for valuable discussions and to Professor W. A. Noyes, Jr. and Dr. E. A. Flood of Brown University, to Professor A. V. Grosse of the University of Chicago, and to Professor G. P. Baxter and Dr. J. H. Faull of Harvard University for their generous assistance in providing the chemical compounds which were used in the analyses.

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The Transmutation of Sulphur by Th C′ Alpha-Particles

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The protons emitted from sulphur under bombardment of Th C′ alpha-particles have been resolved into three groups corresponding to nuclear energy changes -2.4 ± 0.3, -2.85 ± 0.3 and -3.6 ± 0.3 MEV. The first of these values, together with Bainbridge's value for the mass of Cl^{35}, has been used to deduce a value for the mass of S^{32}, namely 31.9812 ± 0.0016. An excitation curve for the emission of protons has been plotted which obeys the Gamow penetration formula within the limits of error: from this formula the nuclear radius of S^{32} has been derived and has the value $5.4\pm0.3\times10^{-13}$ cm.

The determination of the energy levels of as many nuclei as possible is an essential preliminary to the foundation of a satisfactory nuclear theory. In particular, the binding energy of a nucleus, which depends on its mass defect, is of great importance and the present knowledge of nuclear masses as far as neon has already been used to test various suggested intranuclear forces and constitutions. Beyond neon the information is unsatisfactory and there is clearly need for an experimental determination of as many nuclear masses as possible in this region of heavier nuclei. The element sulphur offers an opportunity for investigation since the isotopic mass of its neighbor chlorine is known and a transmutation to chlorine, with a determination of the energy change involved, would enable the isotopic mass of sulphur to be found. Such a transmutation is feasible according to the reaction:

$$_{16}S^{32}+{_2He^4}\rightarrow{_{17}Cl^{35}}+{_1H^1},$$

in which the only unknown mass is that of sulphur.

The fact that sulphur has the relatively high nuclear charge of 16 means that its potential barrier is high and therefore that the proposed transmutation will not take place unless very energetic particles are used to bombard it. The alpha-particles emitted by Th C′ are the most energetic available, having an energy of nine million electron volts, and are therefore the most likely to effect the required change. We therefore bombarded a sulphur target with Th C′ alpha-particles with intent to detect emitted protons and measure their maximum energy. This has enabled the mass of sulphur to be derived. At the same time transmutations leaving a chlorine nucleus in an excited state occur, giving groups of shorter range protons, and measurement of the ranges of these groups has enabled the values of the excited energy levels to be found.

It is further possible to derive information

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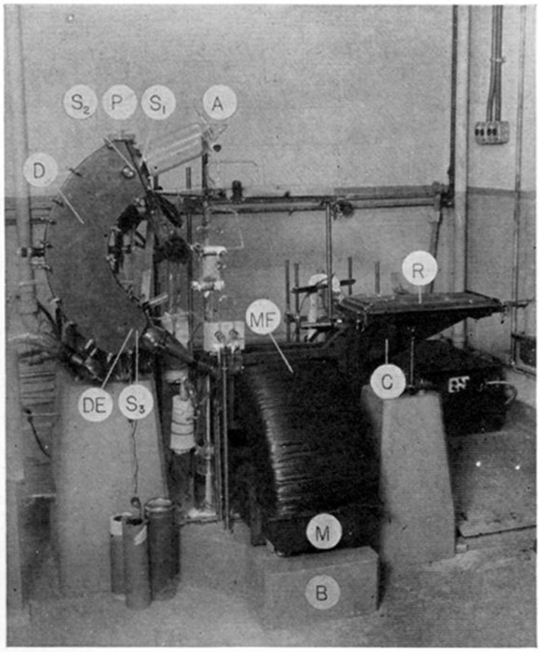

FIG. 1. The mass spectrograph. *A*, the discharge tube. S_1, adjustable steel slit under the cathode. *P*, port for adjusting S_2, the second slit. *D*, deflection chamber containing radial electrostatic field deflecting plates. *DE*, adjustable diaphragm in front of the third slit S_3, *MF*, the magnetic field. *M*, the magnet yoke. *B*, reinforced concrete base. *C*, camera chamber. *R*, cover of the plate-holder compartment.

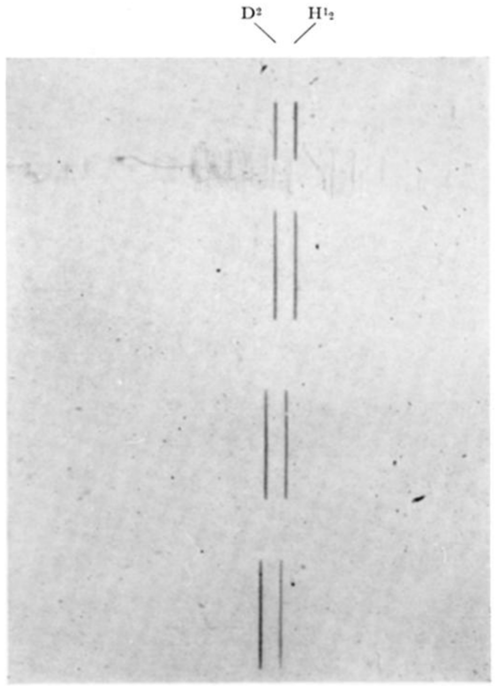

FIG. 10. Some $D^2-H^1_2$ doublets enlarged 5×. $S_1=0.03$, $S_2=0.017$, $S_3=1.0$ mm. Resolving power 10,000 to 15,000.

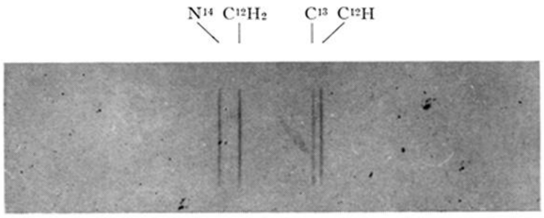

FIG. 11. $N-CH_2$, $C^{13}-C^{12}H$, doublets. Enlarged 5×.

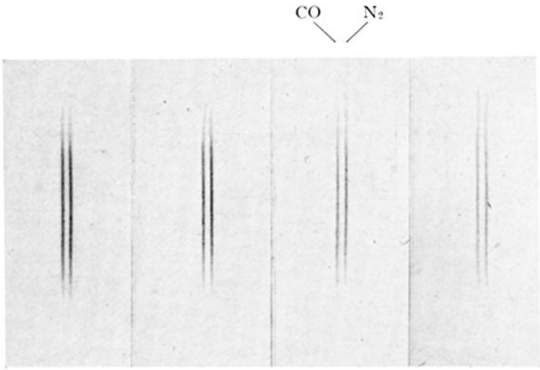

FIG. 12. CO$-$$N_2$ doublets taken without a diaphragm in front of the recording plate. Enlarged 5×. The ions differ in mass by 1/2500. $S_1=0.04$ mm, $S_2=0.017$ mm, $S_3=1$ mm.

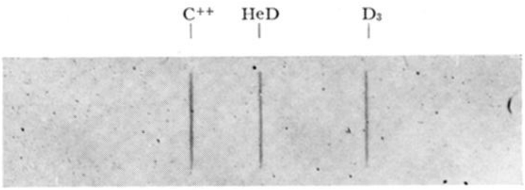

FIG. 13. $C^{++}-HeD^2-D^2_3$ matched triplet. Enlarged 5×.

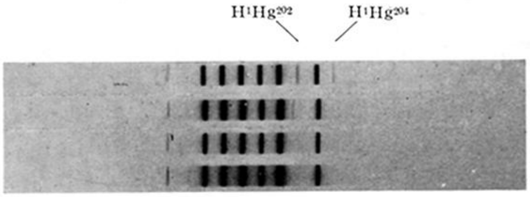

FIG. 17. Spectra of mercury showing hydrides of Hg^{202} and Hg^{204}. Successive spectra were taken with decreasing amounts of water vapor in the discharge tube.

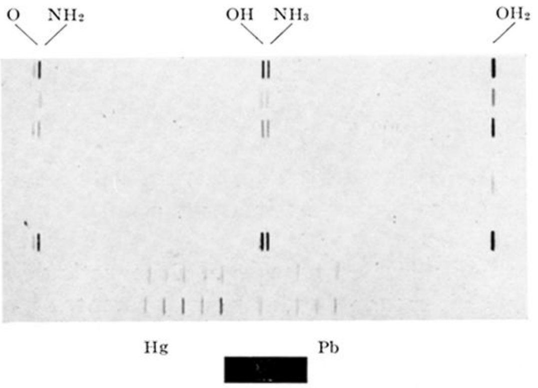

FIG. 5. Natural size. Spectra of matched $OH-NH_3$ doublets and the isotopes of mercury and lead, $S_1=0.13$ mm, $S_2=0.1$ mm, $S_3=5$ mm. The black rectangle represents the width of a single trace for the same slit dimensions but without magnetic refocusing of divergent beams of ions.

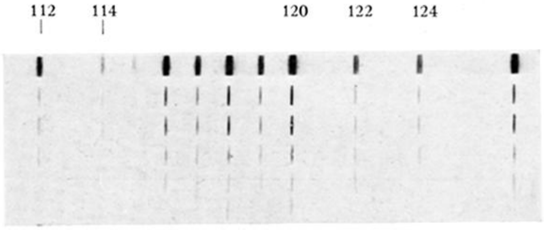

FIG. 6. Natural size. Doubly ionized tin. The magnetic field changed while the top spectrum was being photographed giving the effect of double width lines. On the original plate Sn^{115} appears on the first four spectra but the reported Sn^{121} is not present at all.

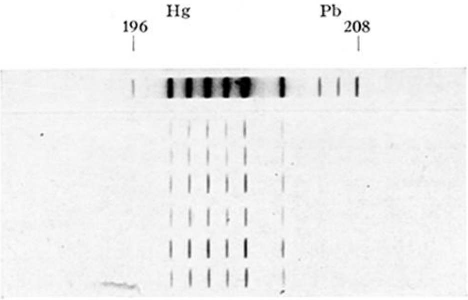

FIG. 7. Natural size. An overexposed spectrum of mercury, and lead, showing the mercury 196 line and comparison spectra from the same plate. An abundance range of 60,000/1 can be recorded and measured.

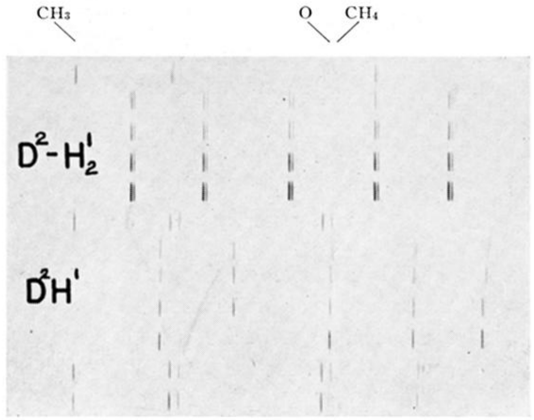

FIG. 8. O$-$CH$_4$, D$^2-$H$^1_2$ doublets, and D^{2}H lines, natural size, for slit widths $S_1=0.06$, $S_2=0.02$, $S_3=1.25$ mm.

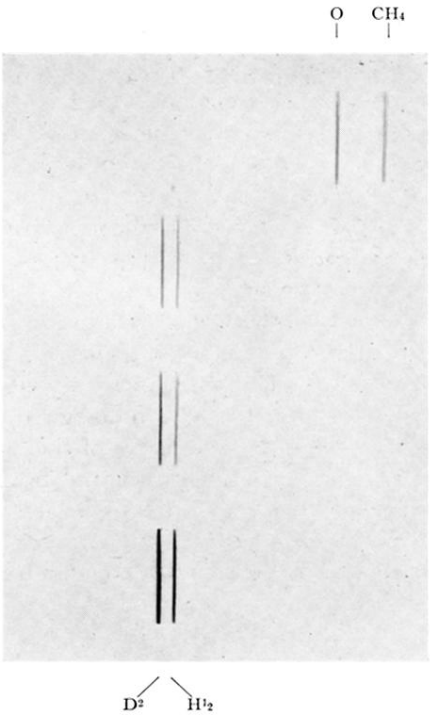

FIG. 9. A portion of Fig. 8 enlarged 5×. O$-$CH$_4$ and D^2$-$H$^1{}_2$ doublets.