

THE DEVELOPMENT OF A HIGH INTENSITY
MAGNETIC LENS TYPE OF MASS SPECTROMETER

by

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I. INTRODUCTION

The identification and study of isotopes constitute one of the interesting and important branches of contemporary physics. In the cases of the three radioactive series, nature furnishes us with pure isotopic species, but, for the non-radioactive elements, the isotopes occur always in a mixture of constant proportions. Many attempts have been made to separate quantities of such isotopes. The dependence of diffusion and evaporation rates upon mass has been used with partial success. By the first of these methods Aston⁽¹⁾ showed neon must be composed of at least two isotopes. By repeated applications of the second method Bronsted and Hemesy⁽²⁾ obtained samples of mercury differing from each other in weight by about 0.05 per cent. Numerous other attempts, both successful and unsuccessful, are reported in the various scientific journals. A long list of ingenious but unsuccessful methods depending on centrifuging, photo-chemical effects, ionic migrations and the like is given in Aston's "Isotopes"⁽¹⁾. None of the above mentioned devices can give more than extremely incomplete separation.

The use of positively charged atoms and molecules in mass spectrometers is the only available method by which isotopes can be separated in the pure state and their number and mass in a given chemical element determined. Various types of this instrument have been designed by J. J. Thomson, Aston, Dempster, Smythe, Bainbridge, and a number of other investigators⁽³⁾.

All mass spectrometers developed heretofore must collimate the initial ion beam by means of narrow slits and in consequence the amount of isotopic material received at the detector is extremely minute. For

this reason, only measurements of mass and relative abundance are performed on individual isotopes and very faint isotopes may escape detection entirely.

The obvious extension to obtain higher intensities requires a mass spectrometer of such construction that wide beams of ions may be utilized. This is accomplished by the magnetic lens type of instrument which will be discussed in the following pages.

Two of many possible kinds of magnetic lenses have been devised by Dr. W. R. Smythe, and, for convenience in discussion, will be referred to as Type I and Type II. A Type I instrument was designed and constructed at the California Institute of Technology by Dr. Smythe, who used it during 1924 and 1925. He was assisted for a time by Dr. Dinsmore Alter. Dr. Smythe then began investigation of another problem and experiments with the Type I spectrometer were continued by Dr. H. M. Evjen from 1926 to 1929. He made little progress beyond that attained by Dr. Smythe. The author began work with the apparatus in 1929. This thesis is devoted to the results of studies made with the Type I instrument, and to the construction and operation of a successful spectrometer with a Type II magnetic lens.

II. THE THEORY OF THE MAGNETIC LENS.

A wide parallel beam of ions in which individual masses, m_1 , have a common velocity, v_1 , may be obtained by accelerating ions in a uniform electric field. Let one boundary of this field be a plane, the equipotential ion source, $y = V_0$. The other boundary may be the plane $y = V$. Then all ions of mass m_1 and charge e , emerging at the upper boundary of the electric field have a common velocity, $v_1 = \frac{2e(V_0 - V)}{m_1}$

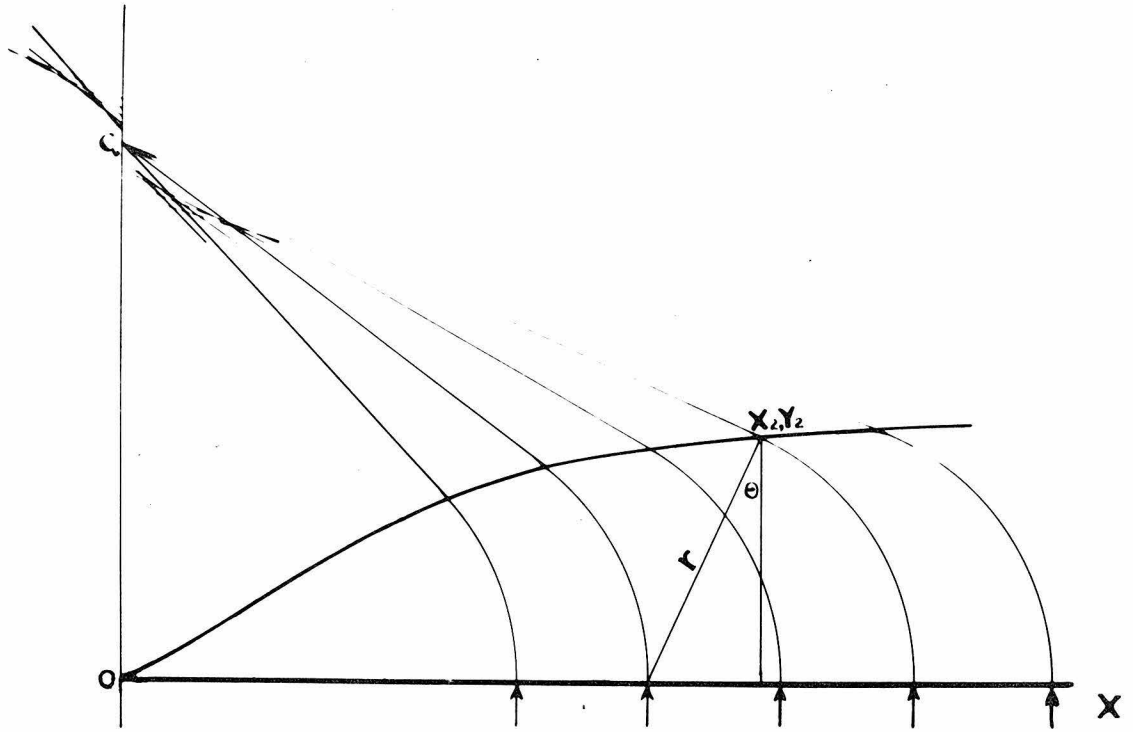


FIG.1

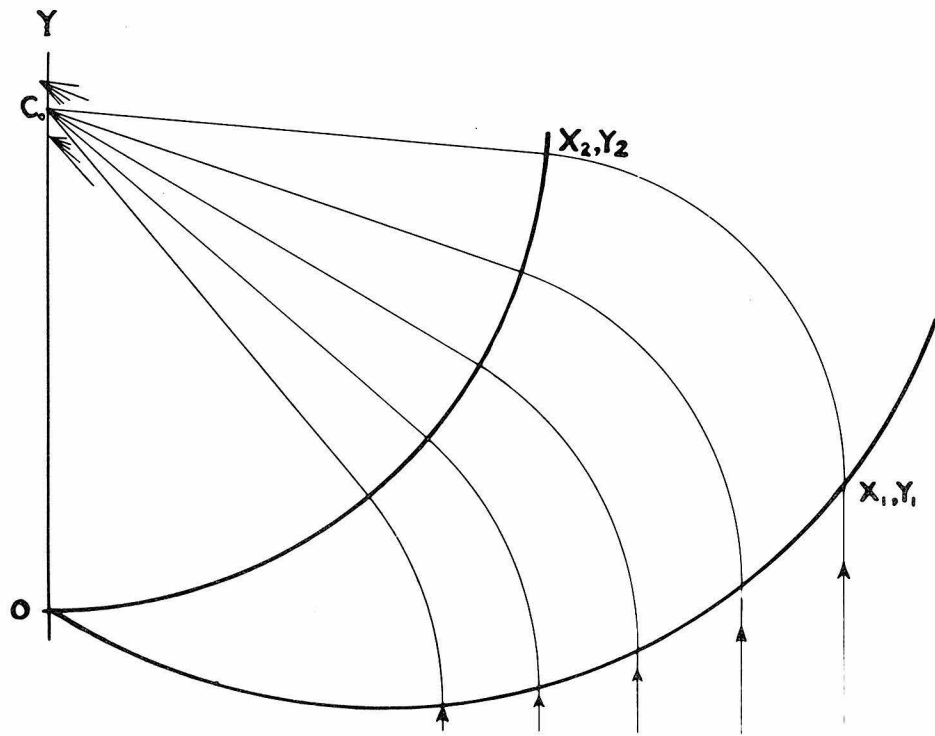


FIG.2

and their trajectories are the straight lines $x = \text{a constant}$.

Let these ions enter a uniform magnetic field whose direction is perpendicular to the plane of the ion beam. Consider the boundaries such a field may have if all ions of mass m_1 are to be brought to a focus at $x = 0, y = C_0$. Two of the sets of boundaries by which this may be accomplished will be considered.

Type I Magnetic Lens

If the plane $y_1 = 0$ is chosen for the first boundary of a uniform magnetic field, the equation of the second boundary surface may be written by inspection of Fig. 1:

$$\tan \theta = \frac{\sqrt{r^2 - y_2^2}}{y_2} = \frac{C_0 - y_2}{x_2}, \text{ or } x_2 = \frac{(C_0 - y_2)y_2}{\sqrt{r^2 - y_2^2}}$$

where r is the radius of the family of circles which the ions of mass m_1 and velocity v_1 follow in the magnetic field.

If ions of mass m_2 also are present in the beam, they follow circles of radius r_2 in the magnetic field and hence their paths do not pass through the principal focus, $x = 0, y = C_0$.

The distinguishing feature of the Type I magnetic lens is that all ion paths are perpendicular to the first boundary of the magnetic field.

Type II Magnetic Lens

Suppose the first field boundary is the cylinder, $x_1^2 - 2rx_1 + y_1^2 - 2C_0y_1 = 0$, (Fig. 2). Consider an ion whose path is $x = k$ entering the uniform magnetic field at (x_1, y_1) on this boundary. In the field the ion travels a circular path whose equation is

$$\left[x' - (x_1 - r) \right]^2 + \left[y' - y_1 \right]^2 = r^2,$$

and leaves the field at $(x', y') \equiv (x_2, y_2)$. In the field-free space its path must be the straight line,

$$y - \theta_0 = \frac{y_2 - \theta_0}{x_2} x,$$

if it is to pass through the principal focus, $x = 0, y = \theta_0$. These three equations may be solved for the equation of the second boundary of the field, which is

$$x_2^2 + y_2^2 - 2\theta_0 y_2 = 0.$$

This is the equation of a circular cylinder whose axis passes through the principal focus. Hence the distinguishing feature of the Type II magnetic lens is that all ion paths converging to the principal focus are perpendicular to the second boundary of the magnetic field.

III. FACTORS AFFECTING FOCAL SHARPNESS.

The above equations for the boundaries of the magnetic field were derived from a special set of assumptions; that the magnetic field is uniform and sharply bounded; that the constituents of the ion beam have parallel trajectories; that all ions of the same mass have common velocities upon entering the magnetic field. Under such conditions there exists a sharp focus - we have called it the principal focus - to which all ions of a certain mass, m_1 , converge. These ideal conditions cannot be realized in practice and focal defects result. Since it is obvious that focal defects can originate only in the magnetic lens and in the ion beam, let us consider first those inherent in a magnetic lens.

FOCAL DEFECTS ORIGINATING IN THE MAGNETIC SYSTEM.

All possible causes of focal defects which can be traced to the magnetic system may be grouped into four classes: (1) lack of focal sharpness for ion beams converging toward regions other than the principal focus; (2) fringing fields outside a gap between pole pieces; (3) irregularities in the pole pieces; (4) variations in the total magnetic flux. We shall consider these classes in the order named.

Suppose there are ions of masses different from m_1 present in the beam. If the paths of these ions - say of masses $m_2 > m_1$ and $m_3 < m_1$ - are computed, they will be found to pass above and below the principal focus respectively. The focal regions for these ions are found to be not sharp. If we use the terminology ordinarily associated with optical systems, we may say that something analogous to coma exists for those beams converging off the principal axis. In the Type I magnetic lens these focal regions are diffuse and occur at distances from the lens which are considerably different. (See Fig. 1). This factor materially decreases the linear separation between the various components of the ion beam and, if the ratio m_3/m_1 or m_2/m_1 approaches unity, the separation becomes so small that complete resolution may be accomplished only for a beam of limited breadth. Furthermore, if two or more isotopes are to be collected simultaneously, only one can be in definite focus. Should the collector not be located fortuitously on the principal axis, even this single, definite focus will never be realized.

Fortunately, this defect is removed almost entirely in the Type II lens, (Fig. 2). The focal regions over considerable distances on

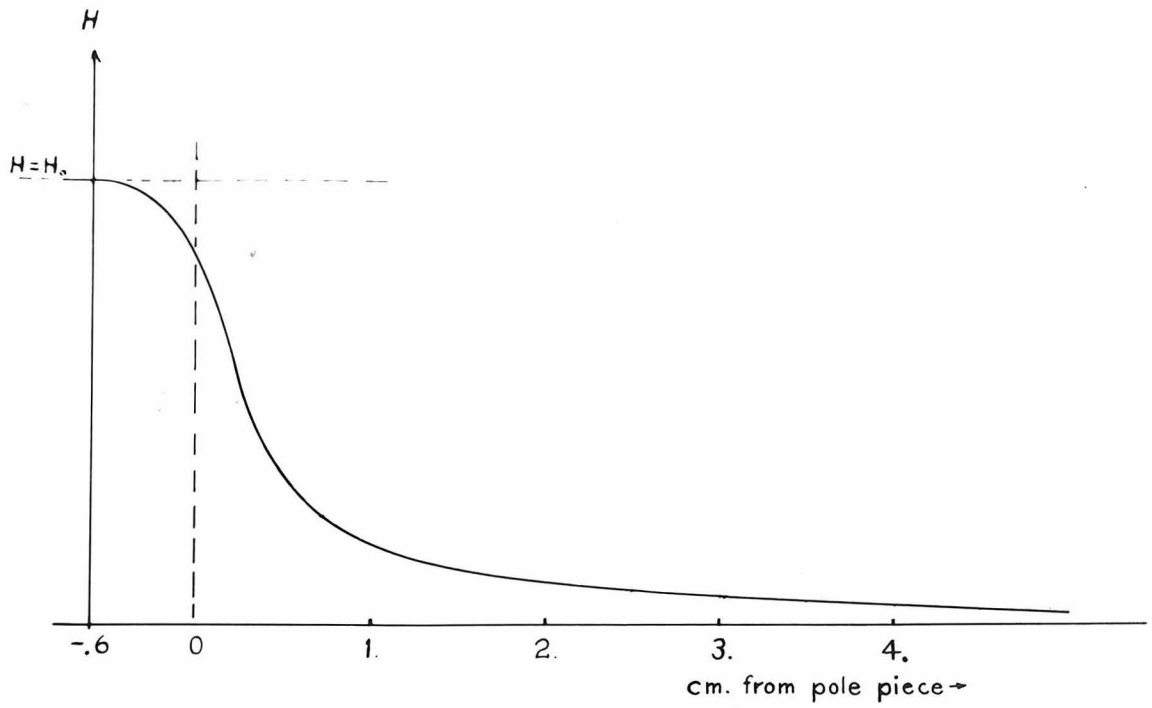


FIG. 3

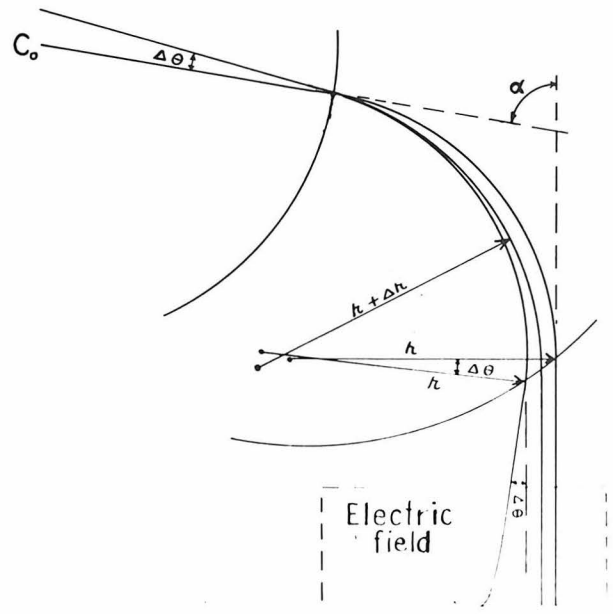


FIG. 4

both sides of the principal focus are comparatively sharp. Moreover, these regions are approximately equidistant from the lens. Consequently, collectors located off the principal axis can be utilized and several isotopes may be collected simultaneously.

It is impossible to realize a sharp boundary for the magnetic field across a gap between pole pieces. The central distribution of field outside a gap 6 millimeters wide between plane pole pieces, as computed from a Schwarz transformation, is shown in Fig. 3. The effect of such distributions must be investigated and an edge correction applied to the pole pieces. In the Type I lens the fringing field scarcely can be calculated, for the upper pole piece boundary is not a symmetrical surface. The Type II lens has circular boundaries so that the field outside the gap may be represented by a radial function, $H(r)$; i.e., the field distribution can be a function of the perpendicular distance to the pole piece boundary only.

The distribution function, $H(r)$, may be obtained in two ways: by constructing pole pieces similar to those to be used and measuring the field at many points; by computations utilizing a Schwarz transformation. The first method is expensive and cannot give the true field at a particular point because measurements must be made with a test coil of finite area. The second method assumes the pole pieces to have infinite permeability and is rigorously applicable only to plane pole piece boundaries. In neither case does it seem possible to express $H(r)$ in a finite number of integrable terms. In consequence, a rigorous solution for the path of an ion in the field $H(r)$ is extremely difficult, probably impossible to obtain.

It is possible to approximate a circular pole piece boundary by circumscribed and inscribed regular polygons. The magnetic field near the pole piece boundaries now is divided into regions which are regular trapezoids. In these regions the distribution of field is given in Cartesian coordinates as a function of a single variable, the distance from one of the sides of the polygon. This reduces the problem to the calculation of successive short ion paths from region to region. Furthermore, by suitable orientation of the circumscribed and inscribed polygons, upper and lower limits may be computed for the circular case.

Thus, suppose a particle of charge e , mass m , and velocity v_0 in an $x y$ plane enters a magnetic field, $H(x)$, whose direction is parallel to the z axis. Let the initial angle between the path of the particle and the x axis be ϕ_0 when the particle is at x_0 . Then the magnetic field imparts a radius of curvature to the ion path given by

$$r = \frac{ds}{d\phi} = \frac{\sec \phi}{d\phi} dx = \frac{mv_0}{He}$$

Hence,

$$\sin \phi_0 - \sin \phi = \frac{e}{mv_0} \int_{x_0}^x H(x) dx$$

Successive applications of this equation may be used to calculate the path through the magnetic field.

Since the computation depends more critically upon the relative values of the field at the boundaries of the trapezoidal regions, it seems advisable to obtain the distribution function, $H(x)$, from a Schwarz transformation. This distribution function is corrected to approximate the circular case, and curves similar to Fig. 3 are used to evaluate the various integrals by mechanical methods.

Similar, but rather involved, calculations may be made for the

effect of the change in direction of the fringing field for ions entering near one side of a pole piece gap. The important point is that the sum of all edge effects may be reduced to such form that the corrected pole piece boundaries may be turned in a lathe. This property of the Type II lens is preferable to that of Type I which must be cut point by point in a milling machine.

A third possible defect in the magnetic field may arise from irregularities in the pole pieces. It is required that the pole piece material shall be homogeneous and that the pole piece faces shall be plane and uniformly separated. The first property is possessed to a very high degree by Armco iron. The lower limit to the second requirement is set by machine shop practice. It may be assumed that no surface irregularities greater than one thousandth of an inch will occur and these may be made smaller if necessary. These irregularities are usually tool marks which are so many and regular that their integrated effect is nil, even if the effect of stresses in the magnetic field in insuring further uniformity is disregarded. The mean separations of successive small areas of the pole piece faces may be made uniform to one ten thousandth of an inch or better, which is ample.

That the total flux in the magnetic system may be held exceedingly constant has been demonstrated by Aston and many other investigators. They obtain sharp lines in mass spectrograms requiring long exposures. We shall assume that variations in total flux may be eliminated by proper experimental technique.

FOCAL DEFECTS ORIGINATING IN THE ION BEAM

The defects in the ion beam must be associated solely with lack of parallelism and lack of common trajectory velocities for ions of

identical mass passing through the magnetic field. It may be assumed that a uniform electric field and an equipotential ion source are obtainable. The remaining factors to be considered are the thermal distribution of ion velocities, magnetic disturbances near the ion source, and collisions and ionic interactions.

The ions in the beam will have thermal velocities given by the Maxwell-Boltzmann distribution law. The thermal velocity component parallel to the accelerating field may be eliminated immediately, for thermal velocity components average only a few hundredths of a volt when the source is as hot as 2,000° K. Such a velocity component becomes negligible when as much as 100 volts accelerating potential is used. Hence the common trajectory velocity criterion is fully satisfied.

The thermal velocity components perpendicular to the electric field are serious since they materially affect the angles at which the ions enter the magnetic field. Suppose an ion has a velocity component η , perpendicular to the accelerating field and parallel to the ion source. Its path in a uniform electric field will be a parabola and it will leave the field at an angle $\Delta\theta$ (Fig. 4) with respect to those ions for which η is zero. The equations of motion give,

$$\tan(\Delta\theta) = \eta \sqrt{\frac{m}{2Ve}} = \Delta\theta$$

Where V is the accelerating potential, e the charge and m the mass of the ion. If $V \gg 1$ e.s.u. of potential, $\Delta\theta$ will be small.

In the magnetic field this ion will travel along a segment of a circle of radius r and hence will be deviated through some angle, α . Upon emerging from the magnetic field the ion will proceed toward the focal plane, making an angle $\Delta\theta$ with the path of an ion of the same mass which has no perpendicular velocity component, η . This latter ion

may be made to follow the same path from the magnetic field to the focal plane if it is accelerated first through a potential $V + \Delta V$ while the magnetic field remains constant.

In the electric field, $\frac{1}{2} mv^2 = (V + \Delta V)e$, and in the magnetic field, $\frac{mv^2}{(r + \Delta r)} = H e v$, where H is the magnetic field strength. Then $\Delta r = \frac{\Delta V}{2V} r$, for Δr is small when $\Delta \theta$ is small. It is easily shown by geometry that $\cos^2(\Delta \theta) = 1 - \left(\frac{r}{r + \Delta r}\right)^2 \sin^2 \alpha$. Hence

$$\Delta \theta = \frac{\Delta r}{r} \sin \alpha = \frac{\Delta V}{V} \frac{\sin \alpha}{2r} = \eta \sqrt{\frac{m}{2Ve}}$$

The number of ions having velocity components between η and $\eta + d\eta$ is given by the Maxwell-Boltzmann law,

$$dN_{\eta} = A e^{-\frac{m \eta^2}{2kT}}$$

$$\therefore (dN_{\eta})_{\alpha} = A_{\alpha} e^{-\frac{e}{4kr^2} \left(\frac{\Delta V}{V}\right)^2 \frac{V}{T} \sin^2 \alpha}$$

The total beam arriving at the principal focus is received after its components have been deviated through all values of α such that $\alpha_1 \leq \alpha \leq \alpha_2$.

Then

$$dN_{\eta} = \int_{\alpha_1}^{\alpha_2} (dN_{\eta})_{\alpha} = B e^{-\beta \left(\frac{\Delta V}{V}\right)^2 \frac{V}{T}}$$

Since r and α depend upon the construction of the magnetic lens, β is a constant of the apparatus.

If η is zero, ΔV is zero. Therefore $B = dN_0$

Let $C_{\Delta V} = \frac{dN}{dN_0}$. Then $C_{\Delta V} = e^{-\beta \left(\frac{\Delta V}{V}\right)^2 \frac{V}{T}}$

is a useful equation giving the shape of the curve measured with an electrometer at the principal focus when the accelerating potential is

varied in steps of $(\Delta V/V)$. If it is desired to resolve two given isotopes of masses m_1 and m_2 , $\Delta V/V$ is the voltage-separation constant; i.e., $\frac{m_2 - m_1}{m_1} = \frac{V_1 - V_2}{V_2} = \frac{\Delta V}{V}$, when the magnetic field is kept constant. Consequently, these isotopes will be resolved only if a certain minimum value of accelerating potential, V , is exceeded. If we consider $V = V_0$ as the potential necessary to bring the most intense part of a given isotopic beam to the principal focus, it is seen that the sharpness of resolution will increase exponentially as V_0 is increased, the magnetic field strength being correspondingly increased to keep the isotopic beam on the principal focus.

Let us consider magnetic effects near the ion source; i.e., in the region of the accelerating electric field, as a possible factor disturbing the parallel condition of the ion beam. The magnetic field in this region may have a number of causes, such as electric currents in the heater used for the ion source, remanent fringing field from the magnetic lens, and so on.

We shall require that deviations in ion paths from the direction of the accelerating electric field shall be small. Hence, magnetic forces and magnetic field components parallel to the electric field may be neglected, and the equations of motion take the simple form,

$$m \frac{d^2 \xi}{dt^2} = H e \frac{dy}{dt}, \quad m \frac{d^2 y}{dt^2} = Y e,$$

where m and e are the charge and mass of the ion, y is the direction of the uniform electric field of strength Y , ξ a direction perpendicular to y , and H the magnetic field component perpendicular to both y and ξ . H will be considered constant for simplicity.

From these equations it follows that an ion initially at rest

will emerge from the electric field making an angle $\Delta\phi$ with the direction of y , given by

$$H = \left(\frac{2 V m}{e} \right)^{\frac{1}{2}} \frac{\Delta\phi}{D},$$

where V is the difference in electric potential across the distance D .

Obviously, the above equation may be generalized for the case in which H is constant only along such surfaces as are electric equipotentials in the cross-section of the ion beam. Then, provided H is nowhere greater than H_{\max} ,

$$H_{\max} \geq \left(\frac{2 V m}{e} \right)^{\frac{1}{2}} \frac{\Delta\phi}{D}$$

Suppose that $H = H_z$ is the magnetic component perpendicular to the plane of the ion beam. It may be shown that the focal regions in the neighborhood of the principal focus, C_0 , will remain comparatively sharp for the Type II lens when $\Delta\phi \leq 0.04$ radians for all ions. Thus if V is 1,000. volts and D is 3 cm., a potassium ion will fulfill this condition of angle if $H_z \geq 400$. gauss. In general, this restriction on H_z is very easily satisfied.

Even were H_z in excess of 400. gauss, it would be necessary only to rotate the direction of the electric field through an angle $\Delta\phi$ to bring the ion paths again tangential to the lines $x = \text{constant}$ upon emerging from the electric field. (See page 3).

In case H is variable in space or time, we can extend the equation for H to obtain the permissible variations. Define a certain value of $\Delta\phi = (\Delta\theta)_0$ as the least angular separation at the focal plane, under ideal conditions, between the path of an ion of mass M , on the chemical scale, and another ion of mass $M + 1$. Now, (by page 10)

$$\Delta\theta = \frac{\Delta r}{r} \sin\alpha,$$

where r is the radius of curvature of an ion path in the field of the magnetic lens and α is the angle of deviation. Then the equations of motion for an ion in the mass spectrometer give

$$(\Delta\theta)_0 = \frac{k}{M^{\frac{1}{2}}} \sin\alpha_0 = \frac{K \sin\alpha_0}{m^{\frac{1}{2}}}$$

To keep the resolution high, require that $\Delta\theta \leq \frac{(\Delta\theta)_0}{2}$. This leads to an equation for the allowable variable magnetic field, H_V , in the electric field space:

$$\frac{[H_V]_{\max}}{(\Delta\theta)_0} = \left(\frac{2}{e}\right)^{\frac{1}{2}} \frac{K \sin\alpha_0}{2 \circ} \frac{V^{\frac{1}{2}}}{D} = \frac{AV^{\frac{1}{2}}}{D} \frac{\text{gauss}}{\text{radian}},$$

where A is a constant which may be computed from the geometry of any given magnetic lens.

In case the minimum value of α is 45 degrees, $A > 10^3$ gauss cm. (radians)⁻¹ (volts)^{- $\frac{1}{2}$} . Then, if $V = 1,000$ volts, $D = 1$ cm., H_V for potassium may be as large as 25 gauss. This may be interpreted as follows: under the above conditions, in the electric field space the magnetic field may be allowed to vary in any manner, so long as the difference of the absolute values of the angular deviations of any two potassium ions, in the plane of the ion beam, is not greater than would be the angular deviation of a single potassium ion acted upon by a constant magnetic field, H_z , of 25 gauss in the same electric field.

Collisions and ionic interactions can have little effect on an ion beam if the vacuum is good. Collisions of ions with walls and gas molecules are proportional in number to the density of the beam, and so must give a background of about the same relative intensity for any type

of mass spectrometer, under the same vacuum conditions. This background can be made small, in percentage of the total ion beam in a magnetic lens type instrument, by the same standard methods used successfully in other mass spectrometers. The possibility of electrical repulsions between ions in the beam as it converges to the focal point may be ruled out by elementary calculations. These ions are not closely spaced and are in the converging part of the beam for so short a time that interaction is negligible.

SUMMARY

The object of the above discussion is to make clear that certain very definite conditions must be satisfied if any magnetic lens type of mass spectrometer is to operate successfully. The most rigid of these conditions is set by the Maxwellian distribution of ion velocities. Application of the equation for $C \Delta V$ (page 10) will show that isotopes differing by 5 % in mass cannot be resolved unless an accelerating potential of at least 1,000 volts is used, it being assumed that the ion source is heated to about 900° K. and that all other conditions are ideal. The Maxwellian angular spread in the ion beam is independent of the mass of the ion and, in a given apparatus, depends only on the temperature of the ion source and the accelerating potential. The uniformity and steadiness of the magnetic and electric fields are conditions only slightly less rigid, since the angular separation between isotopic beams is not large, even for small mass numbers. Pole piece edge corrections cannot be made rigorously, so far as I am aware. Considerable absolute error is permissible in a Type II lens, which is partially self-correcting, but relative errors, in correcting from region to region, must be kept small. The other factors discussed are important

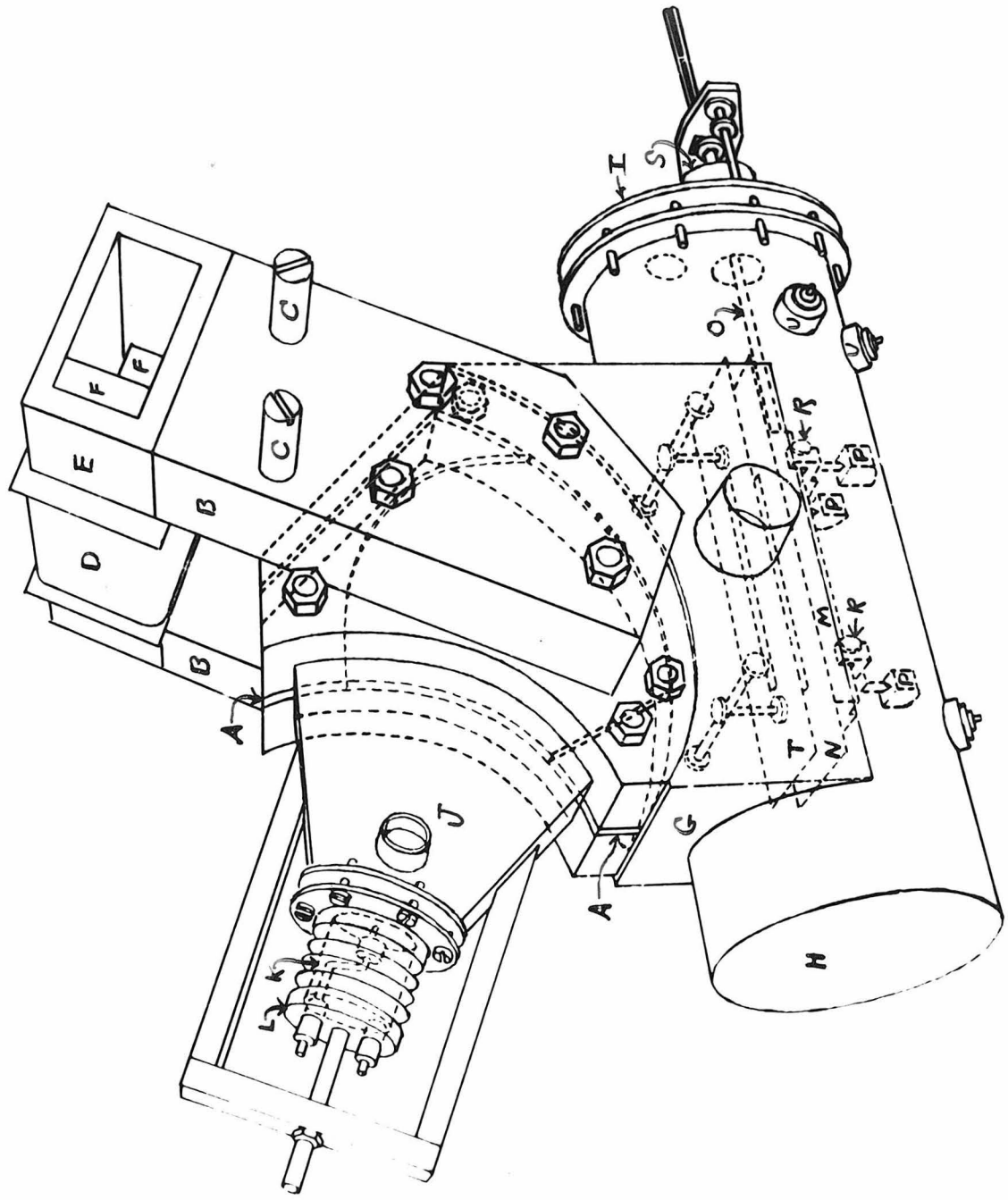
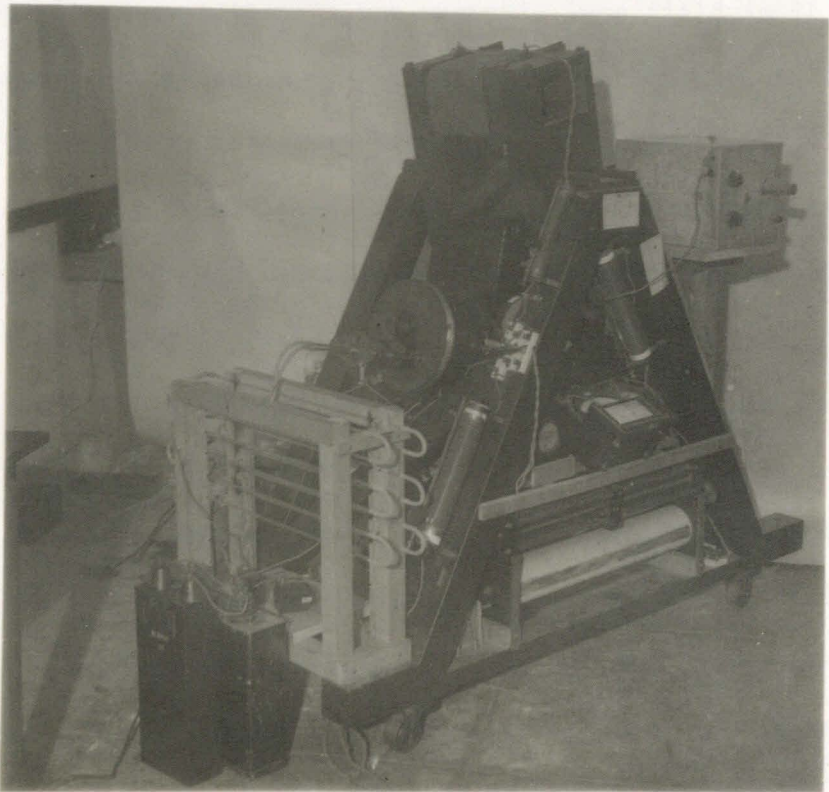
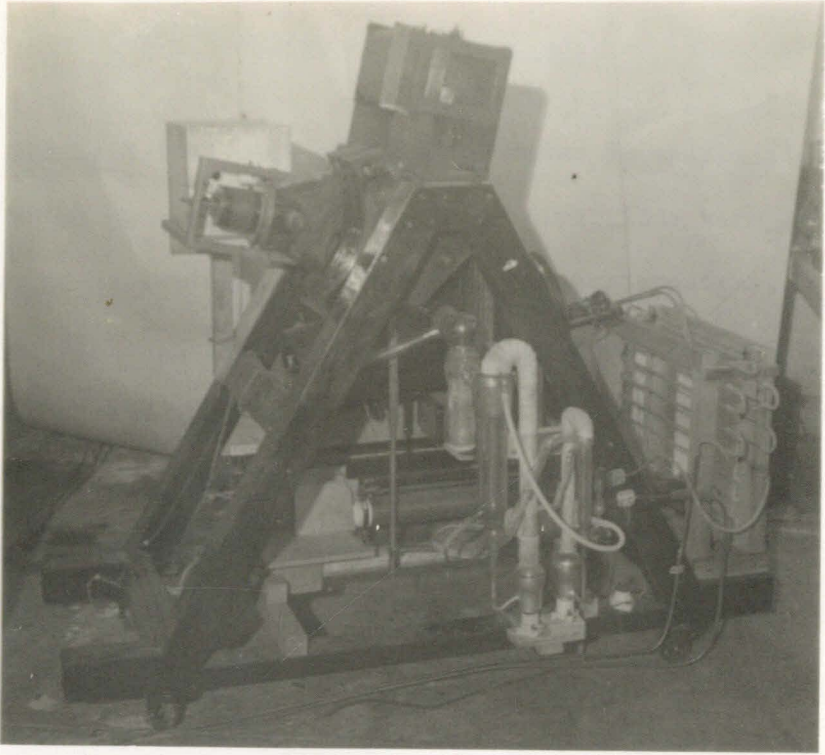


Fig. 5A



when refinements in resolution are sought.

IV. THE APPARATUS.

The principal part of this apparatus is a Type II magnetic lens in which $C_0 = r = 8$ inches. These dimensions were chosen because they give an acceptable value to the apparatus constant, β , in the Maxwellian distribution equation for $C_{\Delta V}$, (page 10), without making the magnetic lens abnormally large in any dimension. A Schwarz transformation was utilized in computing the edge corrections for the lens according to the method outlined on pages 6, 7, 8. The concave pole piece boundary is increased and the convex boundary decreased in radius as a result of this correction.

The pole piece gap is $\frac{1}{4}$ inch, the pole piece faces being held apart by brass spacers, A, Fig. 5. These spacers are so placed that the least permitted deviation in the magnetic lens field is 45 degrees and the greatest deviation is 90 degrees. In consequence, an ion source 20 cm. in length is necessary if the whole working section of the magnetic lens is to be utilized. The pole piece boundaries are extended well beyond the working section to insure uniform conditions at the edges of the magnetic field.

Four one half inch brass bolts pass through the spacers and both pole pieces, clamping the whole rigidly together. There are two side pieces, B, in the magnet yoke. They have a cross-section of 32 square inches and are fastened to the pole pieces with stud bolts. Two heavy studs, C, are screwed into each side piece. The entire apparatus is supported on these studs, which rest in V blocks on a frame. The field windings, D, consist of 890 turns of Number 12 copper wire placed over a hollow iron box, E, into which a variable number of iron bars, F, may

be inserted. This part of the magnetic circuit may be saturated at any desired value by changing the number of the bars, thereby facilitating the maintenance of a steady magnetic field across the pole piece gap. The entire magnetic circuit is made of Armco Ingot Iron because of its exceptional homogeneity.

The pole pieces are each 4 inches wide and are used as a base for a box, G, of rectangular cross-section, which is fastened to the sides and ends of the pole pieces with screws. This box fits into a brass tube, H, 24 inches long and 8 inches in internal diameter. One end of this tube is permanently closed. The other has a removable circular cover, I, for which a tongue and groove construction is used. A narrow rubber gasket is fitted into the bottom of the groove to insure a tight joint. A wedge-shaped box, J, fastened to the pole pieces with screws completes the vacuum system. The whole is evacuated through a short, large tube by means of a two stage mercury diffusion pump backed with a Cenco oil pump.

The ion collector is inside a brass cylinder, K, in one end of which the slit system is placed. The whole is carried on a Sylphon bellows, L, to facilitate location of the best focal region.

The source block, M, is a large brass block containing tubes for water-cooling. A semi-cylindrical opening runs the length of the upper surface. This opening receives the ion source, carried below the lower accelerating field plate, N. The cooling system leads out through a tube, O, from which the water passes through a series of glass tubes which form a resistance to ground. The source block is supported on three insulators, P, by leveling screws. Lateral adjustments of the source block are made with the screws, R. The water tubes in O pass out through an insulating block, S, which is carried on a small Sylphon

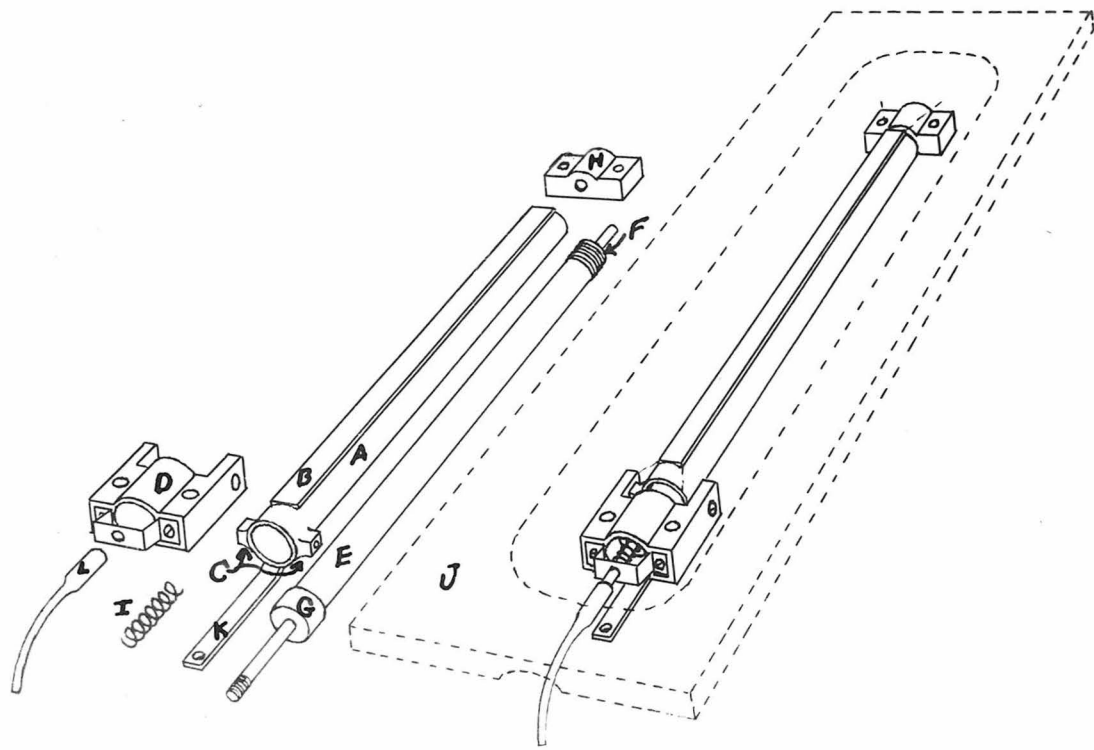


Fig. 6

bellows for flexibility. The upper electric field plate, T, is at the potential of the case and is adjustable both laterally and vertically. Both electric field plates are extended 4 inches beyond each end of the ion source to insure a uniform accelerating field. The potential and current terminals are brought in on the lower side of the large brass tube through a number of insulators, U.

V. THE CONSTRUCTION OF EXTENDED EQUIPOTENTIAL ION SOURCES.

Any ion source to be used with a Type I or Type II magnetic lens should possess the following characteristics: (1) it must be an equipotential surface; (2) it must be straight in the direction of its length, as it is placed in one boundary of a uniform electric field; (3) it must contribute little or nothing to the magnetic field in its neighborhood; (4) it must furnish sufficient heat to cause the source material to emit ions freely; (5) it should be long-lived and rugged; (6) it should be electrically insulated, if possible, from its surroundings.

Ten different types of source construction have been used at various times during the course of this research. Nine of these, which had wire heating elements, will be dismissed without description. In these nine, the objectionable feature was short life, if adequate temperatures were maintained. Long wire elements in general must have a continuous support, especially if the non-magnetic characteristics of the source are to be satisfied. The electrically insulating materials necessarily introduced as supports tend to make the source assembly bulky, hard to out-gas, and wasteful of heat. In consequence, the furnace elements must be overheated and soon disintegrate. This disintegration is hastened by a large number of ceramic materials with which metallic

heating elements interact.

The tenth, and best, of the types of source construction tried is diagramed in Fig. 6. A long iron tube, A, has a shallow trough, B, welded or machined along its surface. At one end, two projections, C, are used to support the tube in a lavite insulating block, D. The electrical heating element is a Globar, E, whose ends are inserted in iron terminal cups, F and G. These cups contain a bit of aluminum which melts and insures good electrical contact between cups and Globar. The cup F is threaded and screws into one end of the iron tube. The rod-like projection of F then forms the other tube support when inserted in a lavite insulating block, H. The cup G slides freely in a hole in the lavite block D, and a tungsten spring, I, holds the terminal cups firmly against the ends of the Globar. The source material is placed in the trough and the assembly is mounted into the lower electric field plate, J, by screws through the lavite blocks. One current terminal is a nickel strip, K, welded to the iron tube. The other current terminal, L, screws over the rod projecting from the terminal cup G. Since the potential of the iron tube is held at a high constant value, the heating current is obtained from an insulating transformer.

This source assembly meets all of the requirements enumerated above. The potential drop along the iron tube is only a few thousandths of a volt. Since the heating current follows the iron tube and returns through the concentric Globar, the resultant magnetic field is nearly zero. The lavite blocks stay comparatively cool and are good electrical insulators. Thermal expansions in the Globar are compensated by the tungsten spring, and one support of the iron tube can slide in its lavite block. Lavite is the product formed by firing high grade soapstone. It

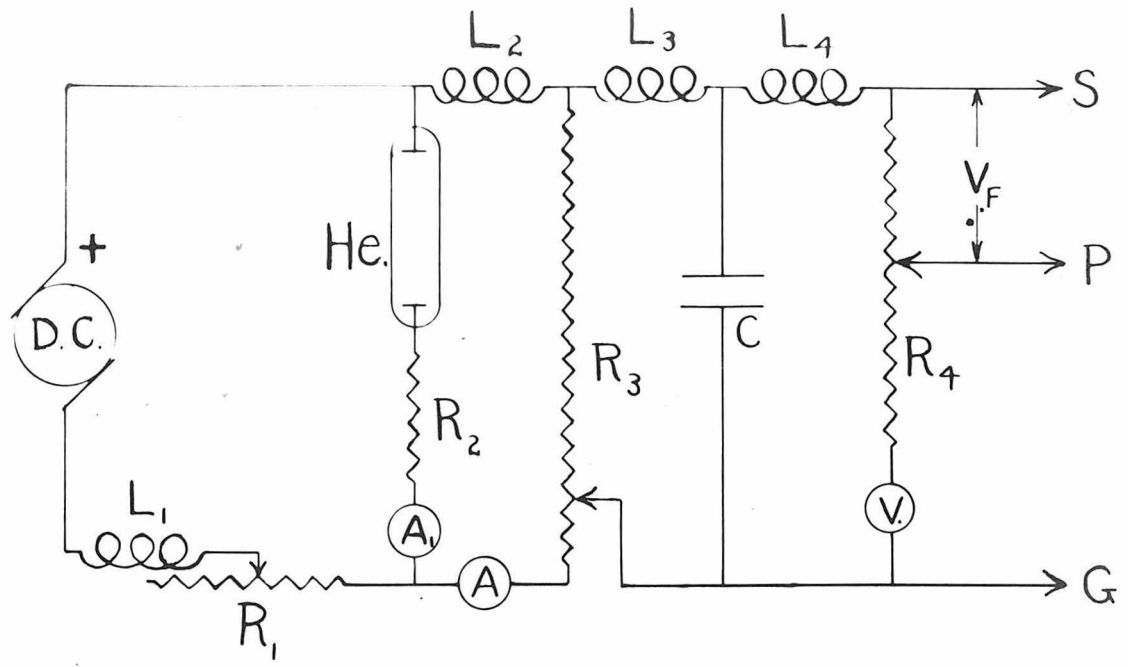


FIG.7

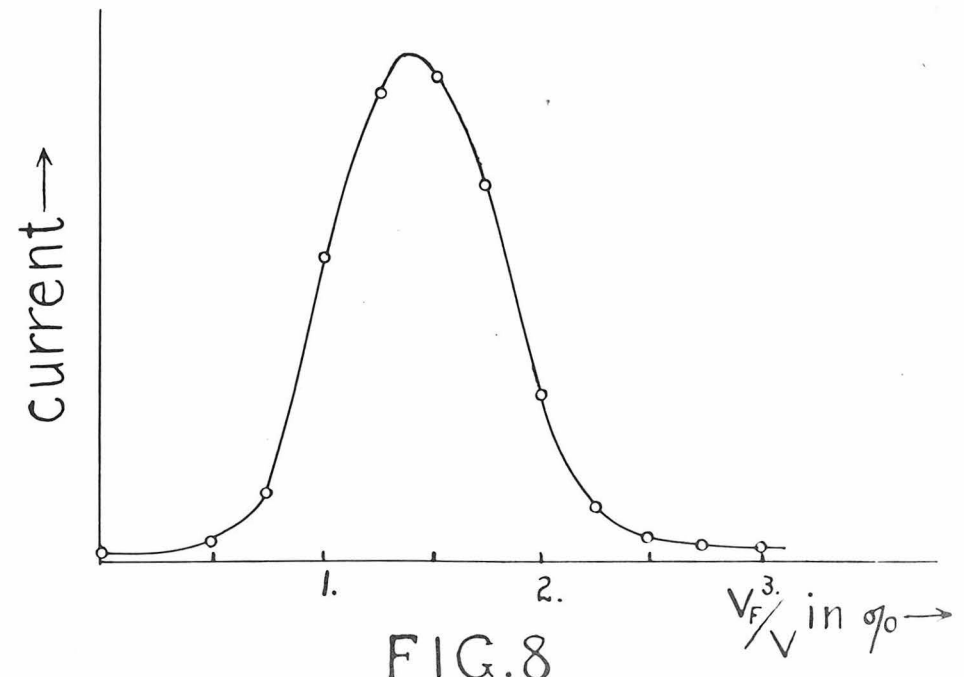


FIG.8

may be cut to any desired shape before firing, which makes it an especially suitable material for these blocks. The Globar heating element is strong and rigid. It is a Carborundum product and may be heated safely to 1800°C . Above this temperature its silicon evaporates. If the temperature of the outside tube is to be above about 1000°C ., the iron parts should be replaced with graphite.

The only source material used has been Kunsman catalyst (4). The catalyst is packed in the trough of the source assembly and reduced for about twenty hours in a hydrogen atmosphere preparatory to use in the apparatus. A water-cooled tube has been built for this purpose. There is little differential thermal expansion between the iron tube and the iron oxide base of the catalyst. This factor is important in an extended ion source, for otherwise the material breaks up and heats poorly. It has been suggested by Dr. Smythe that liquid tin might be used as a base containing the element to be ionized but as yet this possibility has not been tested.

VI. THE CONTROL OF ACCELERATING POTENTIALS.

Obviously, the source of potential for the uniform electric field must be steady to a high degree, since all ions of identical masses must have paths of the same curvature in the magnetic field if good resolution is to result. Some method of control should be introduced by which the accelerating potential can be varied in small steps while the magnetic field is kept constant. Of course, it is possible to hold the voltage constant and vary the magnetic field, but such procedure is not recommended. Voltage measurements are simple, rapid, and accurate compared to measurements of magnetic field strengths. Further-

more, the spectrometer equation for mass number,

$$N = K \frac{H^2}{V},$$

where K is an apparatus constant, makes V a more convenient variable for computations than is H.

The voltage source used to date is a small 2,000 volt direct current generator, (Fig. 7). The voltage drop across the resistance R_3 , in parallel with the generator, is held constant by a helium discharge tube, He, furnished through the courtesy of Mr. Erickson of the Claude Neon Company in Los Angeles. A helium or neon discharge tube of short length may have a zero resistance characteristic over a considerable current range; i.e., $R = \Delta E / \Delta i = 0$. If the discharge tube is lengthened for use at higher potentials, a larger percentage of the total voltage drop occurs in the positive column of the tube and the resistance characteristic becomes negative but remains constant, under proper tube conditions, for a certain current range. A fixed non-inductive resistance such as R_2 is introduced in series with the tube to bring the resultant characteristic back to zero. A rheostat, R_1 , is put in series with one generator line. It is used to vary the current through the helium tube to the proper range.

Any desired voltage may be tapped off on R_3 . With a good generator, such a system will give satisfactory voltage control. Actually, the generator used has so large a voltage ripple, probably due to an open or shorted armature winding, that the helium tube is thrown periodically off the zero characteristic range. In consequence, a number of chokes L_1 , L_2 , L_3 , and L_4 , and a two microfarad condenser, C, have to be used in addition to the helium tube. The voltage between the grounded case

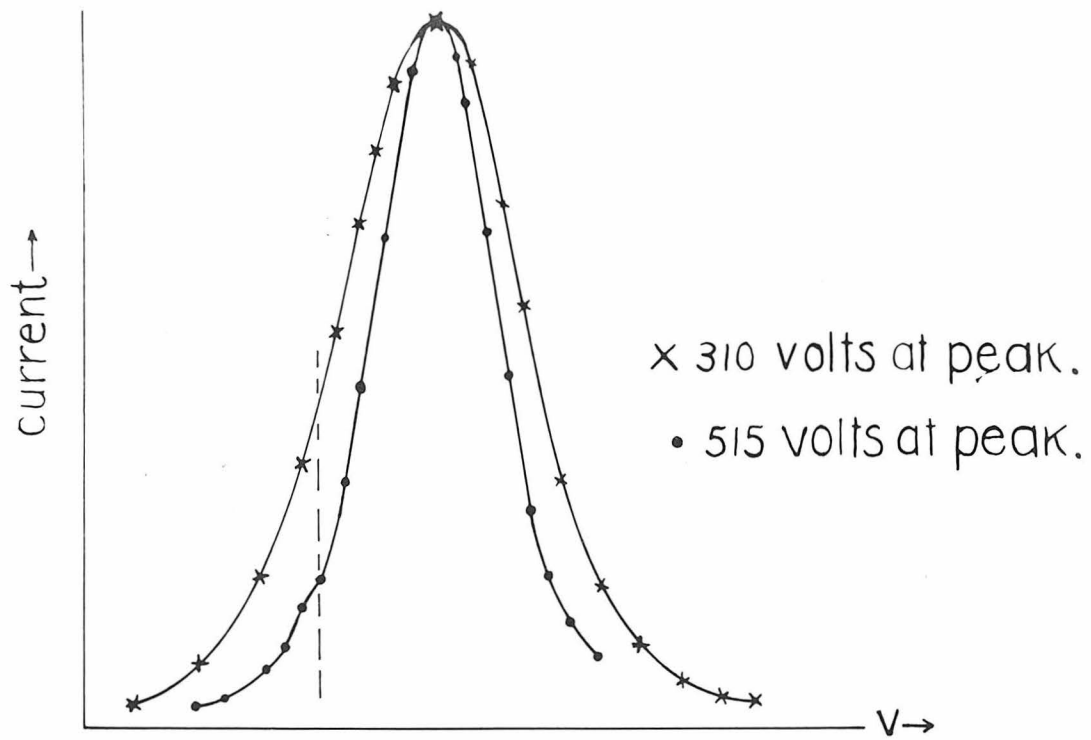


FIG. 9

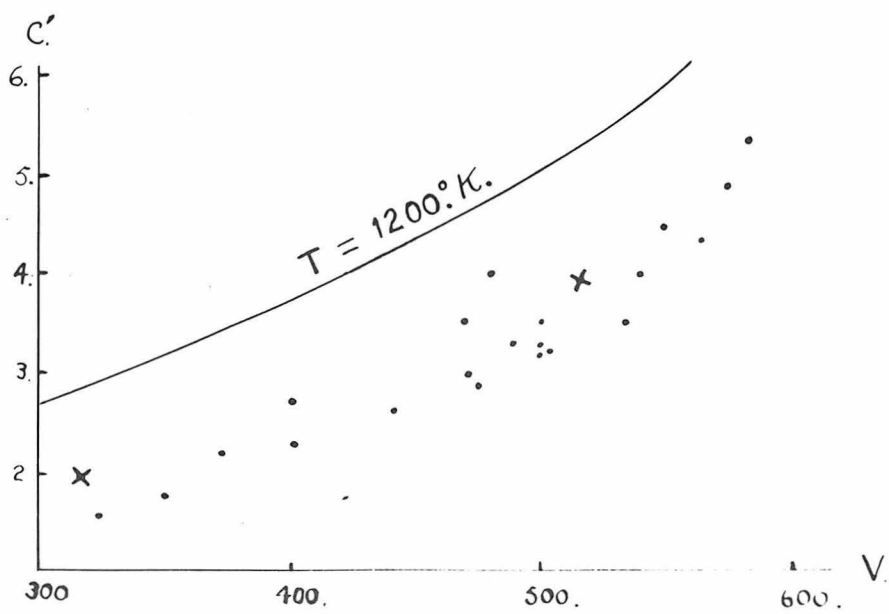


FIG. 10

connection, G, and the source, S, is read from a voltmeter, V, used with a multiplying resistance R_4 . A voltage tap, P, is taken from R_4 . Its use will be explained later - page 25.

The source voltage is varied by shifting the grounded tap on the rheostat R_3 . This rheostat was made by winding several thousand feet of number 33 enameled Advance wire on two glass tubes 26 inches long and 4 inches in diameter. The enamel later was sanded off under the sliding ground tap. Such a rheostat can be used safely for several thousand volts.

VII. EXPERIMENTAL RESULTS.

The usefulness of the magnetic lens type of mass spectrometer is determined entirely by the resolution and the intensity of the ion beam at the collector. The results of experiments on these factors will now be given.

When this research was begun, a Type I instrument already had been built. A water-cooling system was introduced around the source. Potassium ions from Kunsman catalyst were used and a large number of resolution curves were plotted from data taken at various voltages. The dependence of resolution upon the Maxwellian distribution of thermal ion velocities soon became apparent. The peaks sharpened with increasing voltage. In Fig. 9 are shown two representative resolution curves, reduced to the same relative voltage scale, in which collector current intensities are plotted against accelerating voltage. The curve taken at 310 volts is wider than the one taken at 515 volts by about the amount predicted by the expression for $C_{\Delta V}$, page 10. It will be noticed that on the 515 volt curve, the isotope K 41 is just beginning

to appear. Its theoretical position with respect to the center of the broad K 39 peak is marked by the dotted line.

The fundamental dependence of resolution upon accelerating voltage may be made more plain in another way. $C_{\Delta V}$ is defined as the ratio $\frac{dN}{dN_0}$. For convenience in measuring the resolution curves, define

$$C' = \frac{1}{C_{\Delta V} + \frac{1}{19}} = \frac{1}{\frac{dN_{\eta}}{dN_0} + \frac{1}{19}}$$

The factor $\frac{dN_0}{19}$ is added to dN_{η} because the ratio of K 41 to K 39 is 1/19, (5). Evidently C' so defined is the ratio of the intensity at the center of the K 39 peak to the height at which K 41 occurs. C' is equal to 19 for perfect resolution of K 39 and K 41, ($C_{\Delta V} = 0$).

Theoretical values for C' at any voltage may be computed for infinitely narrow collector slits and measured values may be obtained from the resolution curves, although the measured values always must be less than the computed values because of the finite width of the collector slit.

In Fig. 10 the theoretical curve for C' at 1200° K. is plotted for comparison with the measured values for C' taken from over twenty resolution curves at various voltages. The points for the curves in Fig. 9 are marked with crosses. During these runs the source temperatures were kept roughly constant, though considerable variation must have occurred, because wire wound heating elements were being used at the time and had to be rebuilt quite often. The Type I instrument had not been edge-corrected when built and was subject to the coma defect mentioned on page 5. It also was found that the magnetic field in one part of the Type I lens was not homogeneous, probably due to poor annealing of the iron in the pole pieces. These factors, together with the finite width of the collector slit, explain the discrepancies between

the theoretical curve and the experimental points.

The theoretical curve rises rapidly when the voltage is greater than 600 and becomes asymptotic to $C' = 19$ at about 2,000 volts. The magnetic field in the Type I instrument had to be saturated to bring 600 volt potassium ions to the collector, so it was impossible to extend these investigations to higher voltages with the original Type I lens. For a time attempts were made to obtain higher resolution by reducing the temperature of the ion source. In addition, a collimating device, consisting essentially of 100 narrow parallel openings made from one mil copper, was constructed. The device was designed to remove ions having excess thermal velocity components perpendicular to the accelerating field. These methods gave some of the higher values of C' , but such a large decrease in intensities resulted from their use that they were abandoned as impractical.

The information gained by these investigations warranted the construction of a new apparatus. Accordingly, the Type II magnetic lens instrument already described was designed and constructed to specifications adopted after considerable theoretical investigation of edge-corrections and thermal velocities.

The first runs were taken with the new apparatus in January, 1932. The two following months were wasted in attempts to improve the resolution by adjustments on the apparatus. When it was discovered that an enormous voltage ripple, which the helium tube could not eliminate, was present in the generator used as a source of accelerating potential, a number of chokes were obtained or constructed to operate at the correct current values and were used in conjunction with the helium tube and a condenser. The resolution curve shown in Fig. 11 resulted once the accelerating voltage was steadied. This curve and others were taken at

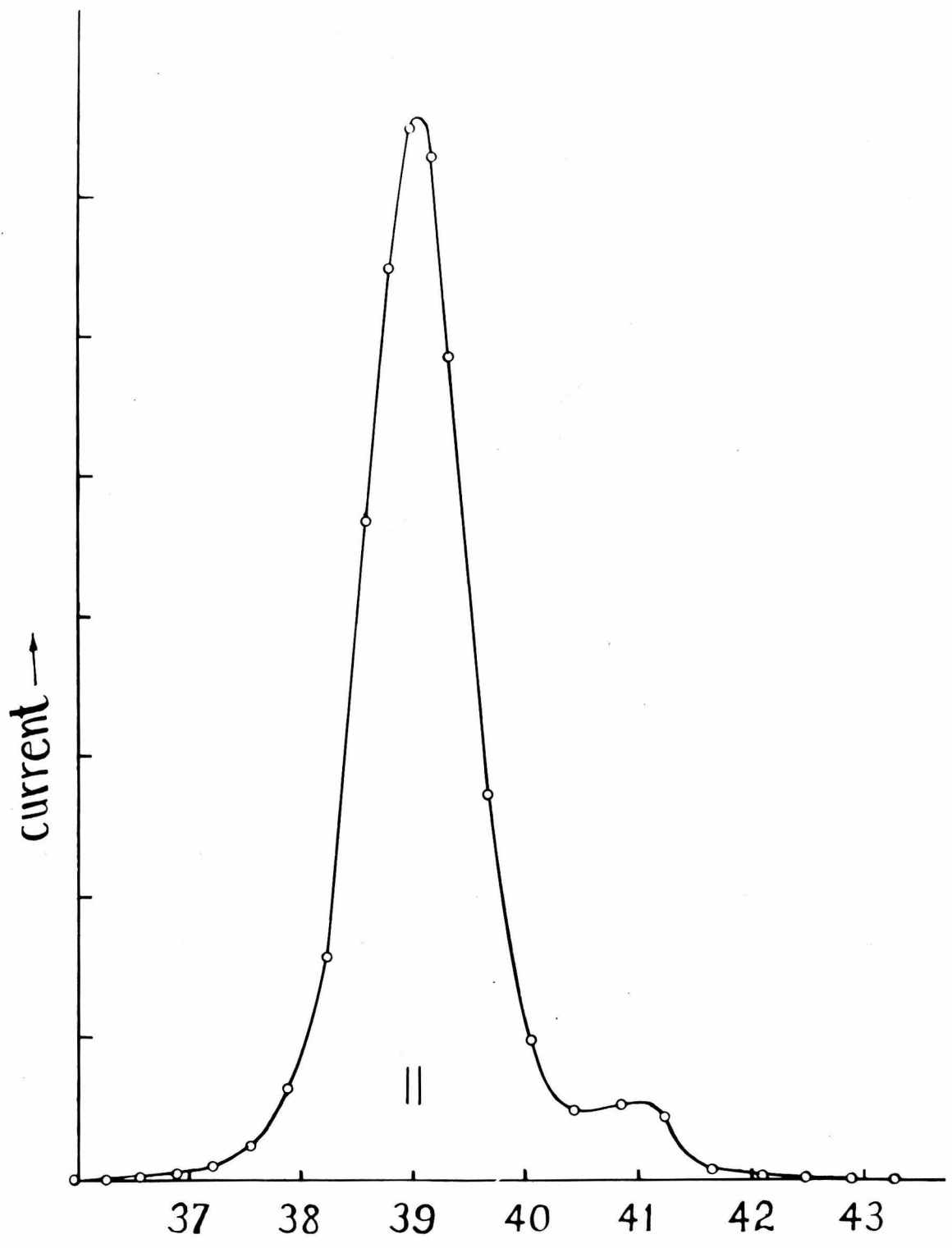


FIG.11

about 1500 volts. This is the approximate upper limit set by the voltage obtainable from the generator and helium tube, minus the voltage drop in the chokes.

The shape of the curve in Fig. 11 closely follows the theoretical shape derived from a Maxwell-Boltzmann distribution law for the thermal velocities of ions at the heated source, and may be taken as an independent proof that this law holds for ions emitted by a heated body.

The peak at 41 is 6.85% of the peak at 39, if the highest part of the K 39 peak is taken to be as drawn in Fig. 11. The accepted ratio of K 41 to K 39 gives 5.26%, so there must be a background of about 1.6% of the K 39 peak present in the K 41 peak. This background is due chiefly to the Maxwellian spread. The theoretical calculation of this spread gives 1.4% as the background, so the agreement is quite satisfactory. It must be remembered that the higher mass numbers come to the collector at the lower voltages; i.e., the mass spectrometer equation is $N = K \frac{H^2}{V}$, so the background from the Maxwellian spread is somewhat heavier on the side of higher mass numbers.

It has been found that intensities at the collector may be increased by a rather simple device. The lower field plate is beveled slightly along the sides of the slot in which the source rests, so that the source sets at the bottom of a shallow V. In consequence, the field immediately above the lower plate will be slightly convergent and the ions leaving the source will tend to be focused into the central plane of the pole piece gap. This initial focusing can be made to offset the diverging action of the electric field along the slot in the upper plate, and will result in an increased number of ions reaching the collector. The amount of this focusing action can be varied at will by applying a

small potential difference, V_F , between the lower plate and the source. A variable tap - P, Fig. 7 - on the multiplying resistance of the voltmeter circuit may be used for this purpose, which allows V_F to be made any desired fractional part of the total accelerating potential, V . Since the ratio V_F/V remains constant for a given setting of the focusing voltage tap, P, the resolution curves will be unaffected and will represent the true distribution of intensities about the collector slit. It scarcely need be said that this would not be the case if V_F alone were given a constant value.

Fig. 8 shows the marked focusing effect which may be gained by proper adjustment of the ratio V_F/V for a particular beveled plate. When this ratio is zero, the ion beam presumably is overfocused and only a small percentage of the ions reaches the collector. A rather broad maximum occurs as the ratio is increased to the optimum value. For still higher values of V_F/V the beam becomes underfocused, which results in it being divergent, and most of it is lost. Doubtless this effect is not entirely due to focusing, as the space charge limitation is more favorable when the initial field becomes more divergent. This may explain why the focusing curve does not come down quite as sharply on the side of high values of V_F/V .

The space charge sets a definite limit to the current which may be drawn from an ion source in an electric field. As the space charge increases, a condition is reached such that the potential gradient at the source becomes zero and the emission rate of the source cannot be increased further. The derivation of the current limit set by space charge in a uniform electric field is a standard one and will not be repeated here. The limiting current, i , is given by:

$$i = \frac{1}{9\pi} \left(\frac{2e}{m} \frac{V^3}{y^4} \right)^{\frac{1}{2}},$$

where y is the distance between two parallel planes whose potential difference is V .

The greatest current so far obtained at the collector has been 2×10^{-6} amperes, when a potential difference of 1500 volts was applied to plates one inch apart. This current is of the magnitude one would expect from the space charge equation and presumably is given by it, for neither increased source temperatures nor the substitution of sources known to be more copious emitters increases the collector current. The dependence on distance also seems to be of the nature given by the space charge equation. Further increases in intensity evidently will require higher voltages or reconstruction of the present electric field system to obtain shorter distances between the plates.

In Fig. 11 the width of the collector slits, in terms of the linear separation between K 39 and K 41, is represented by the parallel lines drawn at the lower center of the K 39 peak. This slit width was made small purposely to give a truer measure of the actual resolution. If isotopes are to be collected, the collector slit width should be increased to the maximum allowable size so as to include as much of the total ion beam as possible.

VIII. POSSIBLE APPLICATIONS OF THE INSTRUMENT

This type of mass spectrometer has certain features which fit it for uses not possible with other instruments. A few of these will be discussed.

The instrument could be used to advantage in the search for faint

isotopes of atomic numbers less than 40. In the present apparatus an extended source 20 cm. in length and 6 mm. in width is used. The integrated emission from such sources is quite steady and many times more intense than that obtainable in a Dempster type apparatus with narrow slits. For smaller mass numbers, the resolution between successive masses is quite complete and very faint isotopes should be detected, even in those elements for which only poor ion sources are at present available. A case in point is the suspected faint isotope Be 8 for whose existence Watson and Parker (6) have found evidence in the band spectra of Be H.

Isotopes of low mass number are of great interest in the problems of cosmic radiation, nuclear structure, etcetera, for it is only when the nuclear structure is simplest, that packing fractions can be measured with most accuracy and transmutation experiments performed.

The instrument could be used for collecting isotopic samples for nuclear studies. Efficient sources should be developed for as many as possible of the isotopic elements lithium, boron, magnesium, sulphur, and silicon, which, together with potassium can be subjected to nuclear disintegration as shown by Rutherford, Chadwick, Kirsch, Petterson and others (7). Of these elements, good sources for lithium and potassium and fair sources for magnesium are known. The separation of the isotopes of these elements is a problem of considerable interest because of the information about nuclear structure and stabilities which would be obtained by transmutation experiments on single isotopes. This is particularly the case for magnesium, which has isotopes 24, 25, and 26. The effect of adding one electron and proton pair to the nucleus might be made clear if samples of the pure isotopes were obtained for disintegration experiments.

The technique of transmutation experiments has been developed to such a point that masses between 10^{-3} and 10^{-4} grams of material, usually deposited as oxides on thin films of gold or platinum, are feasible. To obtain a deposit of 10^{-4} grams of K 39 an exposure of about seven hours to a collector current of 10^{-5} amperes would suffice.

The determination of the isotope or isotopes responsible for the radioactivity of potassium has attracted much interest because this element is far removed from other radioactive elements and falls upon a part of Aston's curve where nuclear energy considerations from packing fractions seem to forbid radioactive disintegration, unless there is a discontinuity of some kind in Aston's curve at this place. In addition, potassium is the heaviest element which all investigators agree can be disintegrated by bombardment with alpha-particles. Kossel suggested K 43, Harkins K 40 or K 41, as the radioactive isotope. More recently, Beck and Fournier have predicted K 43 and K 40 respectively as the disintegrating isotope, basing their conclusions on classifications of all known isotopes for reoccurring regularities in nuclear constitution, (8). Bainbridge (9) has found that potassium isotopes other than K 39 and K 41, if they exist at all, must be extremely faint. Hevesy and Lögstrup (10) and Biltz and Ziegert (11) have measured the radioactivity of potassium in which the heavier isotopes have been slightly concentrated by repeated evaporation and believe the radioactivity to be increased slightly. Obviously, this work does not isolate any particular isotope as the radioactive one.

If we accept Holmes and Lawson's (12) half period of 1.5×10^{12} years for potassium we must have about 3×10^{-3} grams to obtain one β^- particle per second. A 200 hour exposure to collector currents of 10^{-5}

amperes would be necessary to obtain that much potassium. It might be possible to use considerably less potassium, however, if the isotopic samples were placed in a Wilson cloud chamber being used simultaneously for other purposes, since a very few β -ray tracks originating on the potassium would be photographed from time to time.

The instrument in modified forms could find uses outside the field of isotopes. For example, suppose it were desired to make spectroscopic studies of the energy and momentum relationships in inelastic collisions between positive ions of a single element and neutral gas atoms. A modified form of this instrument could be adapted to obtain intense ion beams in which only individual elements would be resolved. Spectroscopic observations could be made on the excitation of resonance lines in the gas as a function of the energy, mass, and charge of the exciting ion. Other uses perhaps will be suggested from time to time.

IX. CONCLUSION

It is the belief of the author that the research outlined in this thesis contributes a new instrument applicable to problems in isotopic study heretofore beyond the reach of available apparatus, and that this instrument may be used successfully in other fields wherever concentrated pure ion beams are demanded.

The author wishes to express his most sincere appreciation to Dr. W. R. Smythe under whose direction this work was done. He desires to thank Messrs. Sherburne and Brower of the Astrophysics Machine Shop for their fine workmanship on the Type II mass spectrometer.

References

- (1) Aston - "Isotopes", Arnold, London, 1924.
- (2) Brönsted and Hevesy - Phil. Mag., 43, 31.
- (3) J. J. Thompson - "Rays of Positive Electricity", Longmans, Green and Co., New York, 1921.
Aston - Phil. Mag., 38, 709, 1919.
Dempster - Phy. Rev., 11, 316, 1918.
Smythe - Phy. Rev., 28, 1275, 1928.
- (4) Kunsman, Harnwell and Barton - Phy. Rev., 27, 737, 1926.
- (5) Dempster - Phy. Rev., 20, 631, 1922.
- (6) Watson and Parker - Phy. Rev., 37, 167, 1931.
- (7) Rutherford, Chadwick and Ellis - "Radiations from Radioactive Substances".
- (8) Harkins - Natl. Acad. of Sci. Proc., 11, 630, 1925.
Kossel - Phys. Zeit., 20, 265, 1919.
Beck - Zeit. f. Physik, 47, 407, 1928.
Fournier - J. de Physique et le Radium, 1, 94, 1930.
- (9) Bainbridge - J. Franklin Inst., 212, 317, 1931.
- (10) Hevesy and Lögstrup - Z. Anorg. Allgem. Chem., 171, 1928.
- (11) Beltz and Giegert - Phys. Zeit., 29, 197, 1928.
- (12) Holmes and Lawson - Phil. Mag., 2, 1218, 1926.