CONSTRUCTION OF A NEW COMBINED VACUUM FURNACE AND MASS SPECTROMETER FOR UNIVERSAL USE.

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1. Introduction.

The vacuum graphite furnace constructed by Saha and Tandon (1936) has been used till now in studying the Lattice energies of alkali halides and electron affinities of halogens (Saha and Tandon 1937, Tandon 1937*a*, 1937*b*, Srivastava 1938, 1939), the thermal ionisation of Ba, Sr, (Srivastava 1940*a*, 1949*b*) Na and K (Bhatnagar 1943), Li (Srivastava and Bhatnagar 1946), Ca and Al (Bhatnagar 1947*a*, 1947*b*). The thermionic work function of graphite (Bhatnagar 1944), molybdenum and iron (Mathur†) and the effect of space charge on electronic currents (Srivastava and Bhatnagar 1944) have also been studied with the help of the furnace.

The method consisted essentially in vaporizing the substance in an auxiliary furnace and then bringing the vapour to an electrically heated graphite furnace of much higher temperature where the molecules suffer thermal dissociation into atoms and ions. The products of dissociation effuse out through an orifice and are collected by a Faraday cylinder, the current being measured by a sensitive galvanometer. The currents due to positive and negative ions are separately measured by applying a suitable negative or positive accelerating potential to the Faraday cylinder.

The first attempt to distinguish between the current due to negative ions and electrons was made by Srivastava (1938) by inserting an electromagnet in such a position that the magnetic field was perpendicular to the effusing ion beam. On applying the magnetic field the electrons were completely deflected off and the negative ion current alone could be measured.

The method was successful in eliminating only the electrons from the ion beam because the mass of the electrons is about 2,000 times less. There may be present in the beam many types of ions of like charge but not differing so much in mass, as is the case with the ions of various elements, and these could not be separated by this method. Further the masses of the ions could not be determined. A method was later developed from the space charge theory by Srivastava and Bhatnagar (1944*a*,1944*b*) and Srivastava (1946), for finding the masses of these ions in case of unipolar unicomponent beams, but this is not applicable to a bipolar or bicomponent mixture (Srivastava and Bhatnagar 1946). Further multiply charged ions would create a difficulty of their own. It is clear therefore that for an accurate quantitative analysis of the different types of ions in the effusing beam we require some sort of mass spectrometer arrangement which should collect all particles of the same mass at one place, so that these can be measured separately from others.

With this end in view the apparatus described in this paper has been designed.

2. Principles of Mass Spectrograph design.

Since the time of Aston's first mass spectrograph various types of mass spectrographs and spectrometers have been designed with a view to obtain high dispersion,

* Now in Lucknow University. † Not yet published.

high resolution and sharp focussing. In the earlier mass spectrographs the beam of mass rays was made homogeneous in some respect before the focussing device was applied. In Dempster's first apparatus (1918) an ion beam of constant energy was deflected and focussed geometrically in a uniform magnetic field. In the Bainbridge spectrograph (1930) and that of Smythe and Mattauch (1926, 1932) a particular velocity is selected by a velocity selector and then deflected and focussed geometrically in the first case by a uniform magnetic field and in the second by a radial electric field.

In all the foregoing apparatus a very sharp focus could not be obtained unless the incident beam was carefully collimated to a perfect linear beam. In actual experiments, however, the beam is somewhat divergent and therefore for a proper design of modern high power mass spectrograph it was essential to investigate the conditions of double focussing. This was completely worked out theoretically by Hertzog, and Mattauch and Hertzog (1934) for all arrangements of a radial electric field and a homogeneous magnetic field for producing both direction and velocity focussing. The double focussing mass spectrographs of Bainbridge and Jordon (1936), of Mattauch (1936) and of Jordon (1940) were constructed fully satisfying the foregoing conditions of double focussing. The double focussing mass spectrograph of Dempster (1935), however, does not fully satisfy these conditions of double focussing. The incident beam employed in all these spectrographs is either of constant energy or of constant velocity.

3. Mass Spectrometers already used for investigating ions.

Several investigators such as Smith (1925), Lawrence (1926), Barton (1927), Hamwell (1927), Hogness and Harkness (1928) have used mass spectrometer of the 180° magnetic focussing type for investigating ions of such substances which exist in the gaseous state or are easily volatilized. The substances studied were hydrogen, iodine, HCl, and the rare gases, and ions were produced by bombarding molecules of these substances by accelerated electrons obtained from heated filaments. The method suffers from several limitations. Firstly, it is very difficult to isolate ions of uniform velocity from the collision space as they will be produced at different places in the chamber and are mixed with electrons. Secondly, only gases and vapours could be studied by this method. Thirdly, the method can only indicate the existence of the various types of ions; the so-called relative abundances observed are of no use in giving us any further information about the quantitative aspects of the reactions involved since the transition probabilities, the precise conditions of reaction, etc., are not fully known or determinable.

We have therefore tried to devise a mass spectrometer which should be capable of studying ions of all substances and be therefore of universal applicability. Further we produce the ions under conditions fully amenable to theoretical treatment. In practice the ions were produced under conditions of perfect thermodynamical equilibrium by heating the vapour inside a graphite furnace under black body conditions.

4. Theory of the magnetic refocussing used in our mass spectrometer.

The refocussing property of magnetic fields has been discussed by W. E. Stephens (1934). A particular case of this is the 60° magnetic focussing used in our mass spectrometer.

Suppose a uniform magnetic field of intensity H in a direction perpendicular to the paper, is limited by boundaries OPQ and OWV (Fig. 1), which make angles θ and γ respectively with the normal to the line AO. Inside the region $QPOWV$ the electron paths will be arcs of circle.

Suppose a homogeneous beam of particles of velocity v enters through a slit A and falls perpendicularly on OPQ at P. If the intensity H of the magnetic field is

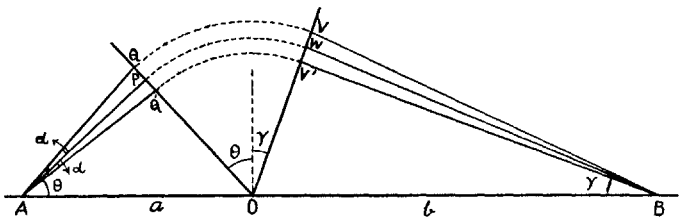

FIG. 1.

such that it bends the beam in an arc of a circle whose radius of curvature $R = a \sin\theta = OP = OW$, then the centre of curvature of the arc will be O and the beam leaves at W perpendicular to OWV meeting the line AO produced at B. The required intensity of the field is given by $H = Re/mvc$. Slightly divergent beams such as AQV and $AQ'V'$ having the same velocity v but making angle $\pm\alpha$ with AP will cross very close to B. It is clear that best refocussing is obtained at a point B such that

$$OB = b = \frac{a \sin\theta}{\sin\gamma}.$$

The spreading S and dispersion D at B are given by

$$S = \frac{a\alpha^2}{2}\left[\frac{\sin^2\theta}{\sin\gamma} + \frac{\sin^2\gamma}{\sin\theta}\right],$$

$$D = a\frac{\sin\theta}{\sin\gamma}\frac{\Delta v}{v}(\sin\theta + \sin\gamma).$$

When $\theta = \gamma$,

$$S = a\alpha^2 \sin\theta,$$

$$D = 2a \sin\theta \frac{\Delta v}{v},$$

and

$$a = b.$$

Considering the space available to us inside the evacuated space and certain other limitations we decided to construct a Dempster type of mass spectrometer in which ions of constant energy are incident. Instead of double focussing, a simple 60° magnetic refocussing as used by Nier (1940) has been employed. We have found $\theta = 30°$ and $a = b = 3$ cm. as convenient for our design. The radius of curvature $R = 1{\cdot}73$ cm. The geometry of the focussing is illustrated in Fig. 2.

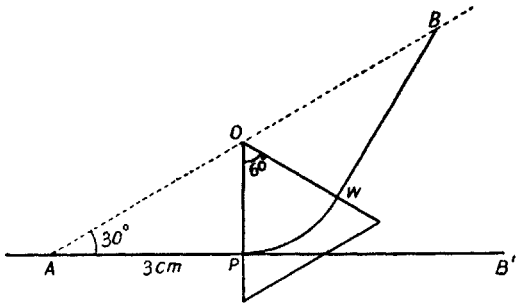

FIG. 2.

$\theta = \gamma = 30°$
$a = b = 3$ cm.
$R = 1{\cdot}73$ cm.

The general arrangement of the assembly is shown in Fig. 3. The positions of the graphite tube G, slit C, magnetic field M and Faraday cylinder F_1, F_2 are indicated in the figure.

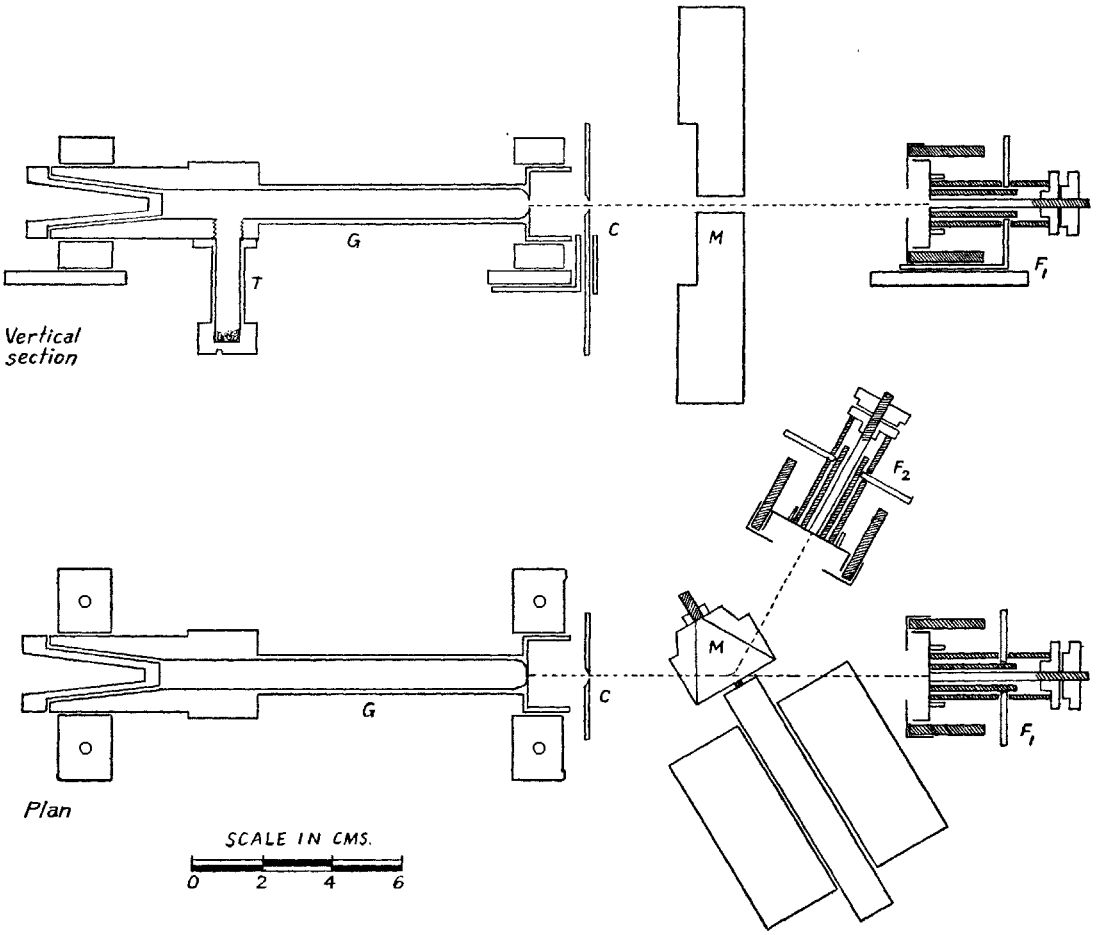

FIG. 3.

5. DESIGN OF THE MASS SPECTROMETER.

The apparatus consists of four parts:—

(i) Ion source.
(ii) Accelerating slit.
(iii) Magnetic field.
(iv) Collector.

(i) *Ion Source.*—This is a graphite tube G which is heated by passing a current of about a thousand amperes from a low tension transformer. The graphite tube is mounted on copper electrodes which carry the heating current and which are themselves attached to water-cooled brass tubes leading outside the evacuated chamber through electrically insulated and vacuum-tight junctions for the purpose of electrical connection. The experimental details of this arrangement, as well

as the enclosing drum and the connections to the pumps were exactly the same as described by Saha and Tandon (1936). This graphite tube, which we call the main furnace, has a side-tube T of graphite or iron attached to it, which was heated by thermal conduction as in the experiments of Srivastava (1940). This side-furnace contained the substance under investigation in the solid or liquid state and the vapour of this substance on entering the main furnace suffers thermal dissociation into atoms, ions and electrons. The products of dissociation effuse out of a narrow orifice of diameter 2·0 mm. producing a composite ion beam consisting of particles of all velocities obeying Maxwell's distribution law.

(ii) *Accelerating slit.*—For producing ions of constant energy out of this non-homogeneous beam it is necessary to accelerate these ions by applying a high potential. Since the thermal velocities are of the order of 0·5 volt, an accelerating voltage of about 30 volts is sufficient to produce a homogeneous beam. This voltage is applied to the slit C with respect to the graphite tube which is earthed. The slit C, which also acts as the defining slit, has to be properly mounted, aligned and insulated. The method of support is illustrated in Fig. 4. The L-shaped iron plate P is fixed to the slotted copper plate supporting the graphite blocks. To the vertical arm of P is attached another iron plate Q which is tightened by two screws x and y, and another iron plate R containing the defining slit (which is in the form of a circular hole of 3·0 mm. diameter), is supported in between these two plates, but is

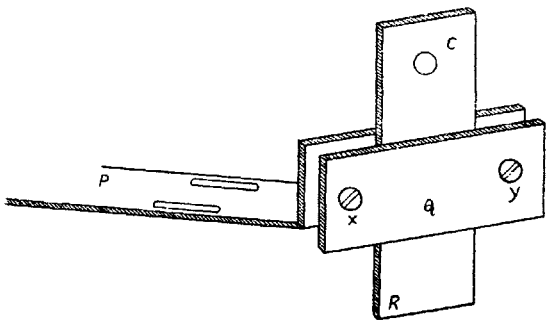

FIG. 4.

carefully insulated from the latter by mica sheets on either side of it. Suitable arrangements are provided for adjusting the distance from the ion source, the height and alignment of the slit.

(iii) *Magnetic field.*—The design and construction of a suitable electromagnet is a major problem in these investigations. For success in these experiments it is essential that the magnetic core should be of such ferromagnetic material which possesses high permeability and low retentivity (small hysteresis loop). The only material available to us satisfying these criteria were transformer stampings. A large number of such U-shaped stampings were rivetted together and two light brass spools, each having 1,000 turns of No. 21 S.W.G. enamel wire, were mounted on either arm of the stamping. The ends of each winding were brought out separately so that the two spools may be connected either in series or in parallels. Mica has been used after every second layer of winding to further improve the insulation.

As already explained the pole-pieces should be so designed as to produce a homogeneous, wedge-shaped magnetic field, the angle between the faces of the wedge being 60°. Further the fringing effect of the magnetic field had to be taken into account to determine the actual extent of the magnetic field in order to locate

correctly the position of the collector. Pole-pieces were made out of an electromagnetic specimen of soft iron and were in the form of a trapezoid of 5·3 cm. in thickness, the four sides measuring 2·5, 2·3, 0·5, 2·3 cm. respectively. Slots in the pole-pieces made it possible to vary the air gap from 0·5 cm. to 1·5 cm. The two cross-sectional views of the electromagnet are shown in Fig. 5.

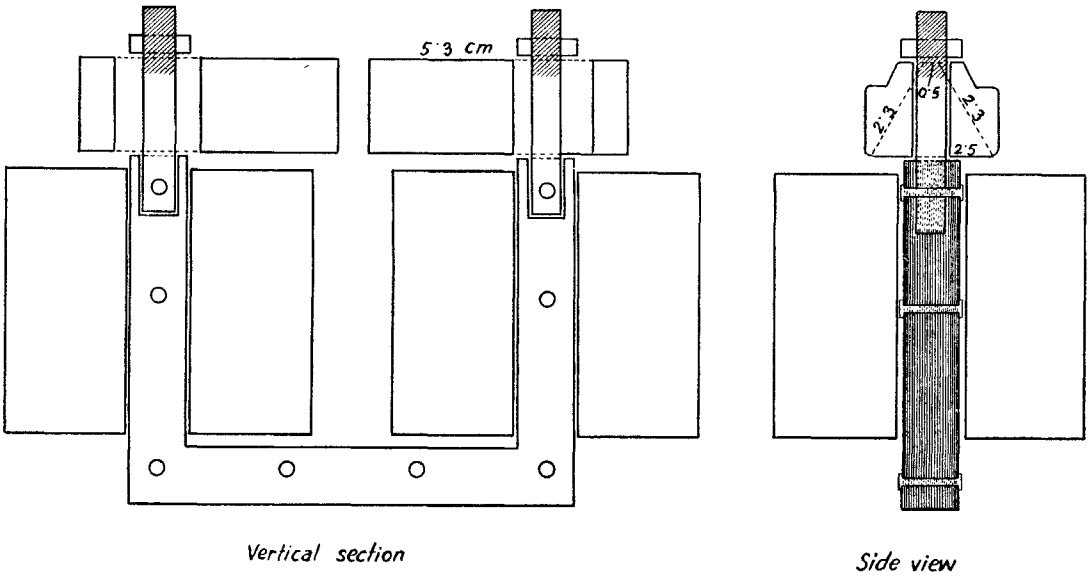

FIG. 5.

The mounting of the electromagnet is shown in Fig. 6. An iron rod *A* has been clamped to one of the electrodes *E* by means of a clamp *C*. The iron rod is insulated from the electrode by placing mica between the clip and electrode. To

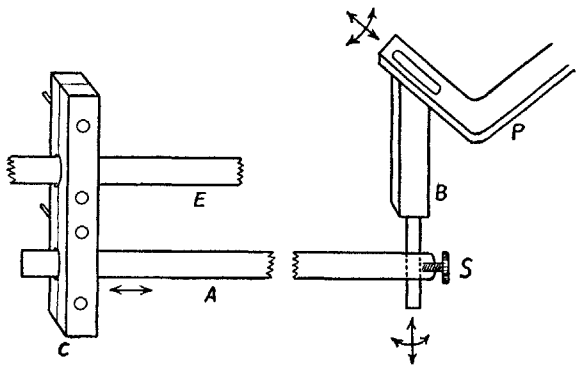

FIG. 6.

this rod is fixed by means of a screw *S* a vertical brass support *B* which carries at its other end a screw and slot arrangement for attaching to it a curved brass strip *P*. The other end of this strip is fixed to the arm of the electromagnet.

The whole arrangement is quite rigid and possesses the advantage of allowing all possible adjustments of electromagnet. The various possible adjustments are indicated by the arrows.

(iv) *Collector.*—There are two Faraday cylinders F_1, F_2, (Fig. 3) to collect the beam of ions, one for the direct, and the other for the deflected beam. They are made of brass and mounted on a L-shaped iron plate L (Fig. 7), but insulated from the latter by means of quartz tubes. The iron plate itself is clamped to a horizontal copper plate fixed to one of the brass electrodes. The iron plate carrying the Faraday cylinder and the supporting copper plate, both are slotted but in perpendicular directions, and hence the Faraday cylinder can be adjusted and

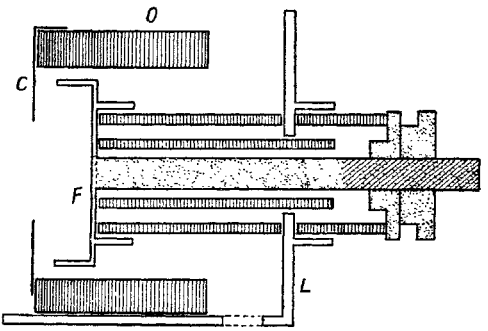

FIG. 7.

fixed in any desired position and direction. For preventing stray ions from reaching the Faraday cylinder, a small wide quartz tube O is mounted over the latter, and a cap C made of brass with a hole in the centre is fitted on this quartz tube.

Seven connecting leads have been brought out of the evacuated chamber as follows: Two for the two Faraday cylinders, two for the electromagnet, two for the thermo-couple (which is Pt,Pt-Rh couple) for measuring the temperature of the side-furnace, and one for the potential on the accelerating slit. The slit and the two caps of the Faraday cylinder were connected together so that the ions may travel in field-free space. The temperature of the main furnace is measured by a Leeds and Northrup's disappearing filament pyrometer.

The whole chamber is evacuated by means of two four-stage mercury diffusion pumps in parallel with a speedy-vac backing pump and is properly cooled by a water jacket. Pressure up to 10^{-4} mm. of mercury can be reached within an hour of starting the pumps.

The adjustment and working of the apparatus will be described in a later paper along with the results obtained.

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SUMMARY.

In this paper, the design and construction of a new combined vacuum graphite furnace and mass spectrometer has been fully described and its advantages over other mass spectrometers discussed. It is very convenient to handle and is of universal applicability since it can be used to produce with ease ions of all substances and of all types under black-body conditions and all these ions can be separately collected and measured. The apparatus is likely to prove of great utility in a variety of measurements with negative and positive ions.

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