PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

Investigations on Paramagnetism at Low Temperatures. Part II

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III. *Investigations on Paramagnetism at Low Temperatures.—Part II.*

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Introduction.

In a series of papers* the author, partly in collaboration with the late Prof. H. Kamerlingh Onnes, has recently published the results of some investigations on paramagnetism at low temperatures. Included in this work were the measurements of the principal susceptibilities of two crystals (cobalt ammonium sulphate and nickel sulphate) at temperatures ranging from about 300° K. (atmospheric temperature) down to the lowest temperature obtainable with liquid hydrogen, 14° K. These data, with the exception of those of Foëx† for siderose, are the only ones yet obtained for the principal susceptibilities of paramagnetic crystals at low temperatures over any extended range of temperature. The results showed that, at not too low temperatures, the principal susceptibilities χ_1, χ_2, χ_3 follow the law $\chi_n (T + \Delta_n) = C$, $(n = 1, 2, 3)$, in which T = absolute temperature, Δ_n is a constant, and C is the Curie constant which has the *same* value for each of the principal magnetic axes of the crystal. The constants Δ_n are intimately connected with the structure of the crystal, being a function of the "spacing" of the paramagnetic atoms in the corresponding directions in the crystal. The precise connection between these quantities could not, however, be deduced, firstly, because of the present scantiness of the data, and, secondly, because the accuracy with which the Δ's could be determined was small. The susceptibilities themselves were determined with an accuracy of about 1 per cent., but, since Δ is only an additive constant, the error in its determination is greater than that of the determination of the susceptibility. Other interesting points were raised by the results, and a continuation of the research seemed likely to give results of considerable theoretical importance and interest.

The present work was undertaken in continuation of that just mentioned. It was decided to carry out the measurements over a range of temperature of from atmospheric

* 'Phil. Trans.,' A, vol. 224, p. 1 (1923), 'Roy. Soc. Proc.,' A, vol. 104, p. 671 (1923), Comptes Rendus, t. 177, p. 154 (1923).

† 'Ann. de Physique,' vol. 16, p. 174 (1921).

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temperature down to the lowest obtainable with the aid of liquid air, and to aim at reaching an accuracy of one part in a thousand in the measurements. For this purpose the apparatus described in the following pages was designed and constructed. The desired accuracy was not attained in the first measurements given later in the present paper, the accuracy of these being about 1 per cent. As experience was gained with the apparatus, it was seen that the desired accuracy was probably attainable when special attention was paid to the working conditions, in particular the constancy of the temperature.

General Description of Apparatus.

It was decided to employ the Faraday method for the determination of the susceptibilities. A section ground in a known direction from a crystal is suspended in a non-homogeneous magnetic field and the force on it is measured, the susceptibility in the direction of this force being then given by the expression $F = \chi m H \ dH/dx$, in which F = force exerted, χ = specific susceptibility in the direction x, m = mass of the specimen, H = magnetic field.

The first point requiring consideration in designing the apparatus was the means to be employed of obtaining and maintaining the low temperatures. It was not possible to employ the method of boiling certain pure liquefied gases under variable but accurately determined pressures, as has been developed very successfully at the Physical Laboratory of the University of Leiden, because of the non-availability of the very special accessories required and of the need of very special vacuum vessels if an electro-magnet is to be used for producing the magnetic fields. Some cryostat, preferably employing liquid air as the cooling agent, was therefore necessary. It was decided to use the type of cryostat described by Keyes and Young,* as this seemed the most suitable for the present purpose. In this cryostat a bath of pentane is cooled by liquid air in a surrounding vacuum vessel and the temperature is maintained by making and breaking the current in a heating coil, a constancy of temperature of one-hundredth of a degree being obtainable with the modified arrangement here used. Since the outer diameter of the outer vacuum vessel of the cryostat actually constructed for the present work is about 12 cms., it was out of the question to employ an electromagnet to produce the magnetic fields. A large water-cooled coil capable of carrying a current of about 20 amps. was therefore made for this purpose. A definite lower limit was set for the inner diameter of the coil by the fact that the coil had to pass up round the cryostat, and the question of ease of handling of the coil set a limit to the weight and hence to the outer diameter and axial length of the coil. A design for the coil was therefore got out in the light of these limitations, taking also into account the necessity for $H \ dH/dx$ being as great as possible. The coil was very carefully and accurately constructed and its dimensions

* 'Journ. Math., Phys. Mass. Inst. Tech.,' vol. 1, p. 191 (1922).

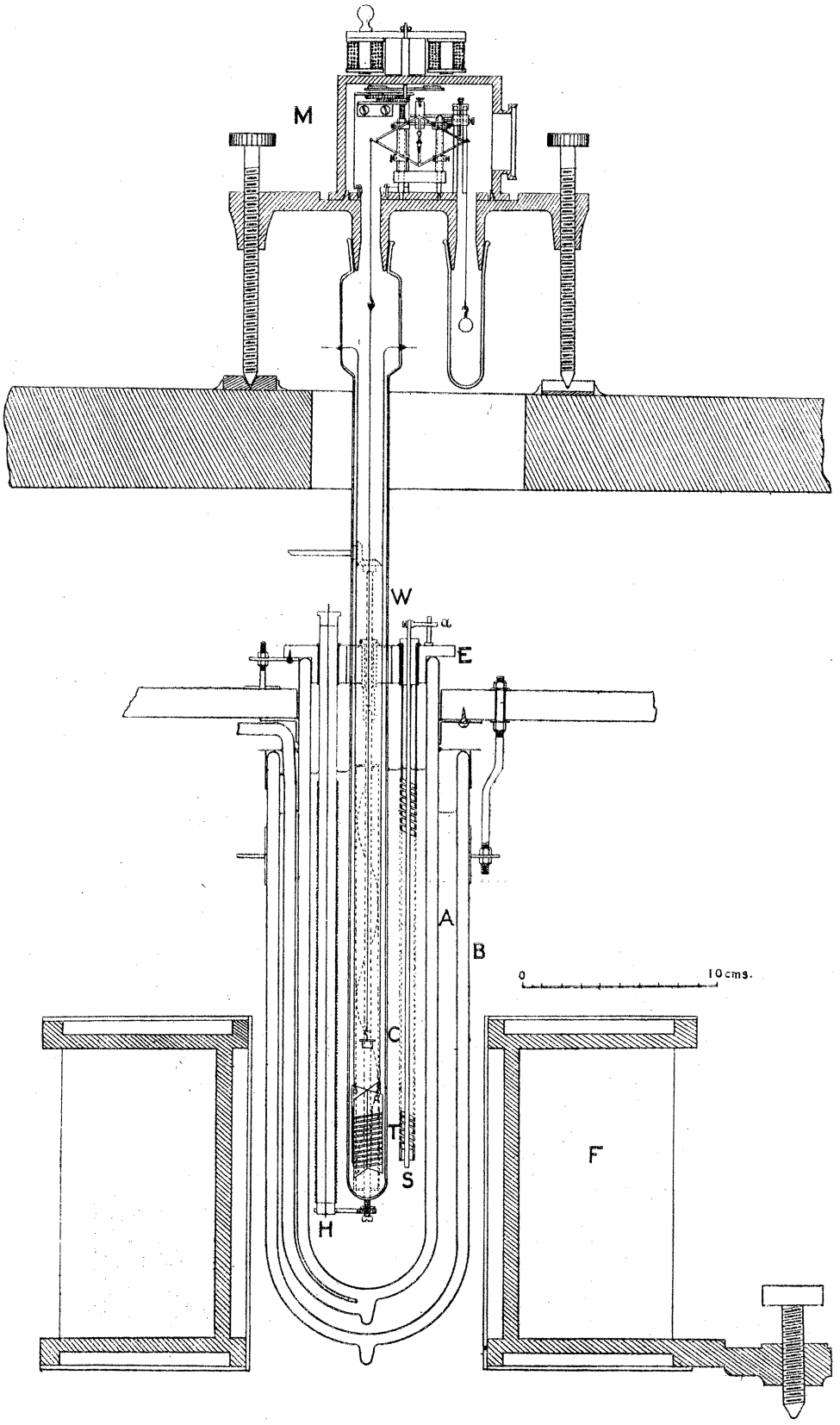

Fig. 1.

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determined, so that the value of H dH/dx could be obtained by calculation from a knowledge of the current flowing and the dimensions of the coil. By this means one of the chief sources of error in this method of the determination of the susceptibility, viz., the *measurement* of H dH/dx, could be avoided. With the coil as actually constructed, it was found that a value of about 40,000 c.g.s. units for H dH/dx could be obtained with a current of 20 amperes.

This value is, however, small compared with that obtainable with an electromagnet for a small distance between the poles. As a consequence the forces exerted on the crystal are correspondingly smaller. A calculation shows that with a crystal weighing 100 milligrams and having a specific susceptibility of 10^{-5} (the order of magnitude usually obtained at atmospheric temperatures) the force to be measured would amount to only about one-hundredth of a milligram weight for a value of H dH/dx of 10,000. Since it was desired to obtain an accuracy of about one part in a thousand in the final result, the means used to measure the forces had to be very sensitive. For this purpose a Pettersson quartz microbalance capable of carrying a load of 200 milligrams and sensitive to one millionth of a milligram was employed. The crystal was suspended from the beam of the balance and the force exerted on it measured directly as a weight. A general idea of the apparatus can be obtained from fig. 1. M is the microbalance from the beam of which the crystal C is suspended by a quartz fibre inside the long weight-tube W. At the bottom of the tube just below the crystal will be seen the platinum resistance thermometer T used to determine the temperatures. Surrounding the weight-tube will be seen the two coaxial vacuum vessels of the cryostat, with the heating coil, stirrer and control mechanism in the pentane bath in the inner vessel. The coil is seen surrounding the outer vessel. Fig. 2 is a photograph of this part of the apparatus as actually set up.

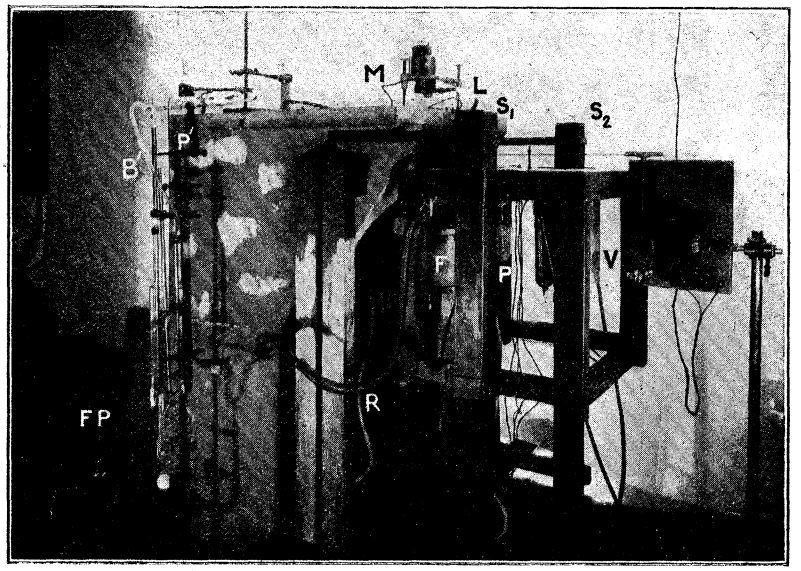

Fig. 2.

Detailed Description of the Apparatus.

(1) *The Microbalance.*

The microbalance used is of the Pettersson type.* It was supplied by the firm of Lyth, of Stockholm. The beam, which is made entirely of quartz, has a length of 5 cm. It is suspended by two quartz fibres instead of resting on knife edges, and the loads are suspended from quartz fibres attached to the ends of the balance beam. These fibres, at their thinnest part, that nearest to the beam, have a thickness of about 1·8 μ. The beam also carries a small mirror, by means of which the position of the beam is read on a scale placed at a distance of 4 metres with the aid of a spot of light reflected from it. The sensitivity of the balance can be altered by turning a small aluminium screw attached to the cross bar of the beam. The load and suspension fibres are detachable from the balance beam, an improvement introduced by Strömberg, whereby one of the greatest difficulties of the manipulation of such a microbalance is avoided. A broken fibre can thereby be replaced relatively easily in a few minutes by anyone possessing a fair degree of manipulative skill. The balance is surrounded by a thick metal case which acts in practice as an almost isothermal surface, thereby aiding in ensuring that the whole of the air in the balance case is at the same temperature. The cover of the balance is provided with a window through which the position of the beam can be observed. A thermometer placed in a receptacle in the removable case gives the temperature of the air in the case. The joints between the cover and the base of the case and between the weight-tubes and the plugs over which they fit are made tight with tap-grease. By turning the small electromagnet attached to the cover, the balance can be freed from the arrestment without disturbing the vacuum in the case, through an armature which is caused by the magnetic forces to move with the electromagnet and which through reduction-gearing rotates the screw moving the arrestment hooks up or down. An inspection of fig. 1 will serve to elucidate the arrangements of the balance and its case.

As is usual, the weights consist of small quartz hooks of various sizes, ranging from about 30 milligrams down to 0·2 milligram, and a hollow sealed bulb, the "air-weight." The rough balancing is performed by hanging the silica weights on the cross-piece attached to one of the load fibres and the fine balancing by altering the pressure of the air (free from carbon dioxide and moisture) in the case and hence the buoyancy of the "air-weight." With this balance it is possible to reach a sensitivity of one ten-millionth of a milligram per millimetre deflection on the scale 4 metres distant. In the present experiments the sensitivity was not pushed to its limit as this was not necessary. The balance was generally used with a sensitivity of about 10^{-6} milligram per millimetre on the scale.

* A description of the latest form of the Pettersson microbalance will be found in 'Phys. Soc. Proc.,' vol. 32, p. 209 (1920).

The pressure of the air in the balance case is read by means of an open mercury manometer P′ and a cathetometer, the height of the barometer B′ placed near the manometer being observed with the same instrument.

So sensitive an instrument as the microbalance requires to be supported on a very stable and steady foundation. To meet this requirement the work was carried out in a room in the basement of the Davy Faraday Laboratory, and a large brick and cement pier was built up to carry the balance. The pier was carried down into the foundations of the building, and brought up through the floor of the room without being in actual contact with it. A large stone slab was fixed to the top of the pier, and the balance was placed upon it. The pier and the stone slab were cut away where necessary to allow the rest of the apparatus to be put in place. Fig. 2 will give an idea of the size and shape of the pier.

(2) *The Cryostat.*

The cryostat was of the type described by Keyes and Young,* modified in certain directions, partly to meet the special circumstances of the present case and partly to increase the accuracy of the automatic control of the temperature. The description which follows is of the present modified design.

Pentane is contained in an unsilvered vacuum glass A (fig. 1), the annular space of which can be evacuated through a side tube sealed on to the lower end of the vessel, as shown. The vacuum is produced by means of a mercury-vapour pump with a Hyvac pump as the fore-pump. A liquid air trap is, of course, included between the pump and the vacuum vessel of the cryostat. The pressure in the annular space of the vessel is measured by means of a Pirani gauge† inserted in the system between the liquid air trap and vacuum vessel. The annular space is filled with hydrogen when liquid air is employed as the cooling agent. Surrounding this vacuum vessel is a larger silvered one B, containing liquid air or, in special circumstances, some other cooling agent.

The pentane bath in the inner vessel contains, in addition to the lower part of the long weight-tube in which the crystal and the platinum resistance thermometer occur, a heating coil, a stirrer and a bimetallic strip wound into a vertical coil, which by twisting and untwisting, as the temperature rises or falls, makes and breaks the heating current, and so controls the temperature.

The heating coil H is wound with fine manganin wire on a thin german-silver tube, the wire being insulated from the metal tube by means of a thin sheet of mica. The wire is wound on double, and the free ends are taken through holes in a talc bush closing the lower end of the tube, and are brought up through the latter parallel to one another. They are then fastened to small terminals on an ebonite bush in the upper end of the tube. The coil is wound double, so that it may not produce any magnetic field, since that would be very undesirable on account of the close proximity of the crystal.

* *Loc. cit.*

† Supplied by the Research Laboratory of the General Electric Company.

Several types of stirrer were tested before a satisfactory one was obtained, the one finally adopted being of the propeller type, and having a large number of small blades arranged in a spiral fashion above each other on a vertical spindle.* The spindle was interrupted near the top with an ebonite rod to minimise conduction along the spindle. This gave a thorough mixing of the liquid of the bath, as it lifted it as well as rotated it. The main difficulty with the stirrer was to obtain one which would give sufficient stirring for a not too rapid rate of revolution, and yet was of such a size as to go into the small space available in the cryostat.

For the control element S a strip of "thermostatic"† metal was wound into a coil of about 20 cm. long and 8 mm. diameter. The upper end of this coil is fastened to a piece of german-silver tubing, while the lower end is free and carries a small cylindrical block of metal. To this latter is fixed the lower end of a light steel rod, which passes up through the coiled strip, and through a long bearing above the tube to which the upper end of the coil is attached. It carries a small arm a, which can be clamped to it by means of a screw. This is tipped with platinum, and moves between two platinum-tipped screws, so serving to make electrical contact between the rod and either the one or the other of the screws. The contact screws and the bearing for the rod are attached to an ebonite cover E, which serves to close the upper end of the inner vacuum vessel. The heating coil and the upper bearing of the stirrer (the lower end of the stirrer spindle turns on a screw point passing through a small bracket attached to the lower end of the tube, on which the heating coil is wound) are also attached to this cover, and the internal mechanism of the cryostat can be removed as a whole by removing the cover. A central hole in the ebonite cover permits the entrance of the long weight-tube into the cryostat.

The cryostat, with its various accessories, is fastened to a frame which can be raised or lowered at will. This consists of a framework of wood in the form of a cube, on the top of which is fastened a wooden board covering the whole. Inside the framework itself hang the Pirani gauge P, the liquid air trap L, and the mercury vapour pump V (fig. 2). By means of a flexible rubber connection R, the system can be evacuated by means of the fore-pump F.P. The upper end of the inner vessel of the cryostat passes through a hole in a projection from the board and is held in place by a metal collar cemented to the vessel and screwed on to the underside of the board. The outer vacuum vessel is supported by means of three bolts attached to the board and which pass through a brass ring cemented to the vessel.

The cryostat thus moves as a whole as the frame is raised or lowered. To permit of this movement two sash-boxes, S_1 and S_2, are fixed vertically in the correct positions, their upper ends being clamped to the pier and their lower ones being let into the floor of the room. Four iron collars attached to the frame pass round the sash-boxes and slide up and down on them. The weight of the frame and the cryostat is balanced by

* The stirrer shown dotted behind the long weight-tube in fig. 1 was that originally used but later replaced.

† Supplied by the British Thomson-Houston Co., Ltd.

means of weights with ropes passing over pulleys in the sash-boxes in the usual way. The frame can be clamped in any position within the limits of its travel by means of four thumb-screws on the iron collars.

The rising and falling frame is a necessity when the crystal has to be changed and the long weight-tube removed. Since the balance, on account of its delicate nature, must of necessity be a fixture, the cryostat had to be capable of being lowered sufficiently far to permit the removal of the weight-tube when a long fibre was to be put on or removed, or the crystal changed. When this is required the frame rests on the floor, and sufficient room is then available for the necessary operations. When the apparatus is actually being used, the frame is raised to its highest point, where it is shown in fig. 2.

At first the stirrer of the cryostat was driven by means of an electric motor bolted on to the upper side of the board of the frame. Its speed was reduced by means of a worm and pinion gear, and the coupling to the stirrer was made through bevel gearing. This arrangement was, however, rejected on account of the inevitable vibration of the motor and the gearing. This vibration was transmitted to the frame and cryostat and so to the balance, with the result that the latter was always swinging erratically. To avoid this disturbing factor, the motor was removed from the frame and attached to a stand screwed to the floor near the frame. Fig. 2 shows the stand, to which is clamped a board, to which are fastened the motor, its reduction-gearing and a lamp resistance. The drive to the stirrer is now made by means of a large pulley on the motor system and a small pulley on the stirrer, connected by a belt. By adopting this arrangement the difficulty of the vibrations of the motor being transmitted to the balance was avoided, and the position of the balance can now be read accurately while the stirrer is running.

As very great difficulty was experienced in attempting to obtain an inner vacuum vessel made of pyrex glass, one of ordinary glass had to be used. To prevent the disastrous consequences of the pentane falling into the liquid air by the inner vessel cracking accidentally, certain safety devices were adopted. A thin metal liner was made for the inner vessel and was later sealed to the ebonite top of the latter. The pentane was placed in this liner and did not come into contact with the glass walls of the vessel. Various liners were tried, the most satisfactory arrangement being one the greater part of which was of seamless copper and the upper two inches of nickel silver. The good conduction in the copper greatly assisted the stirrer in maintaining a uniformity of temperature in the vertical direction in the pentane bath. The upper part of nickel silver was necessary to prevent the stirrer and control contacts becoming covered with frost by too great conduction of the cold from below.

The upper end of the liner was sealed to the ebonite top of the cryostat by means of de Khotinsky cement to prevent the pentane distilling over the top and condensing between the liner and the wall of the Dewar vessel.

The mode of action of the cryostat is as follows:—The vacuum inside the annular space between the walls of the inner vessel is adjusted approximately to some desired value. As a result, there will be a definite rate of cooling of the pentane due to conduction

across the vacuum from the liquid air in the outer vessel. When the temperature has fallen to about the value desired, the current in the heater is switched on and the arm of the control clamped to the rod so that it moves between the contact screws. The movement is reduced to the least possible by altering the distance between the screws. The stirrer is also started. Then when the temperature of the pentane bath has risen by the heating effect of the current, more than balancing the leak of cold in, the bimetallic strip untwists and makes contact with the contact screw, so cutting out the heating current. The bath then cools and the bimetallic strip twists and later switches on the heating current once more. The process is then repeated, and so the temperature of the bath oscillates between narrow limits. In the actual cryostat described by KEYES and YOUNG, the bimetallic strip, by making or breaking contact with a contact screw, switched the heating current in or out directly, the current passing through the contact on the control element. With this arrangement the oscillations of the temperature were maintained at about one-tenth of a degree. To obtain a better constancy of the temperature, a resistance in the heater circuit was altered by hand in response to the indications of the galvanometer in the circuit of the platinum resistance thermometer used to measure the temperature. The reason for the lack of sensitivity in the automatic control was not in the sensitivity of the bimetallic strip control, for that was ample. It actually lay in the quite considerable pressure required between the contacts for the whole of the heating current to pass. It was stated by KEYES and YOUNG that if this difficulty of the large pressure required between the arm and the contact screw could be overcome, the sensitivity of the temperature control would be greatly increased. To accomplish this, and in view of the fact that the temperature control had to be as good as possible for the present experiments, and also completely automatic, the following arrangement was devised* :—As stated in the previous pages, the arm of the control in the present cryostat moves between two contact screws. These are connected so that when the arm makes contact with either the one or the other, a potential of + 12 volts or — 6 volts is applied to the grid of a triode valve. In the plate circuit of the valve is a small electromagnetic contact breaker, which makes or breaks the circuit of the heating current. When the arm touches the — 6-volt contact screw, no current flows in the plate circuit of the valve and the heating current is " on." When, however, the arm touches the + 12-volt screw, the saturation valve of the plate current flows through the valve and energises the circuit breaker, which switches off the heating current. Actually, several valves connected in parallel are used, so that a fairly large plate current, about 20 milliamperes, is obtained with ordinary commercial apparatus and low grid voltages. By this means the difficulty of the large pressure between the contacts is completely avoided, as the heating current can be switched in and out with the lightest touch between the control contacts. The contacts can be put very close together without risk of sparking on account of the very large resistance in

* A description of the device has been published elsewhere. JACKSON, 'Journ. Sci. Inst.,' vol. 2, p. 158 (1925).

the grid circuit and the low voltages applied. By means of this device the temperature of the cryostat can be controlled to about $+0{\cdot}01^\circ$ C., when everything is in perfect order. The average constancy lies between this value and $0{\cdot}1^\circ$ C.

(3) *The Coil.*

A number of factors had to be taken into account in designing the field coil. Firstly, the diameter of the outer vacuum vessel of the cryostat fixed a certain minimum value for the inner diameter of the coil. The necessity for the magnetic effect of the coil being as large as possible would, however, require the inner diameter of the coil to be as small as possible. Secondly, the fact that $H\,dH/dx$ and not H had to be as large as possible made it necessary to use a more or less flat coil rather than one of any great axial length. The possibility of lifting and manipulating the coil without the aid of any very special appliances set a limit to the permissible weight and hence to the outer diameter and the axial length of the coil. The current for which the coil was to be designed required a certain minimum thickness of the wire used for winding, and as many turns of this wire as possible had to be got into the available winding space. In an endeavour to meet these conflicting requirements, the coil was constructed as follows :—

A large brass bobbin, shown in section in fig. 1 at F, was first made. The inner and outer diameters of the winding space are 13·8 cm. and 34 cm. respectively. The axial length is 15 cm. Provision is made for water-cooling on the upper and lower flat surfaces and the inner cylindrical surface of the bobbin. The bobbin was accurately turned to shape on the inner faces of the winding space. These surfaces were then covered with a layer of "Empire" cloth for insulation, and the dimensions, axial length and diameter of the inner cylindrical surface were determined as accurately as possible. The coil was then mounted in a large lathe and the winding commenced.

The wire, which was No. 14 enamel covered, was contained on two bobbins. A wire from each bobbin was passed through a short fibre tube arranged to swivel slightly, next together over a pulley possessing a small possible traverse in the direction parallel to the axial length of the coil bobbin, then under a jockey pulley carrying a weight, then over another pulley capable of traversing the whole length of the main bobbin, and finally to the coil bobbin itself. The ends of the wire were passed through an ebonite bush let into the upper side of the brass bobbin, and were fastened to two terminals on an ebonite block attached to the bobbin. Then, by guiding the wires with one hand and turning the lathe slowly with the other, the two wires were wound uniformly on to the bobbin side by side under sufficient tension to keep them taut. The wire was wound on double so that the two windings could be used in series or parallel as desired and so that the insulation between the windings could be tested. When a layer of wire had been wound on, the diameter of the layer and its axial length were measured carefully at a large number of places along it. These measurements were carried out with instruments kindly lent

to me for the purpose by Dr. P. E. Shaw, Professor of Physics and Director of the Department of Metrology, at University College, Nottingham, and I wish to express my best thanks to him for the loan. The internal micrometer and the calipers used were first carefully checked against a series of end-standards whose lengths were guaranteed accurate to 1/10,000 of an inch and which were also kindly lent for the purpose by Dr. Shaw. The temperature of the room in which the winding and the measurements were carried out was determined each time the measurements of the dimensions of a layer were made. The temperature did not vary by more than about one degree from day to day.

After the dimensions of the layer had been determined, the triangular gaps left at both ends of the layer between the commencement and the finish of the last turn were filled in up to the level of the top of the layer with melted Faraday wax. When this had set hard another layer of wire could be wound on without the wire sinking into the gaps, the diameters of the layers thus remaining constant along their lengths. The diameter of any layer was found to be satisfactorily constant for various points round the circumference for various positions along the length of the layer. Before the next layer of wire was wound on, the lower layer was covered with " Empire " cloth for insulation. Each layer was thus insulated from the next one. In this way thirty layers of wire were wound on and the dimensions of each layer determined.

The outer layer was covered with "Empire" cloth, wrapped with tape, and the whole coated with shellac varnish. The outer ends of the wires were brought to two terminals fastened to an ebonite block attached to the upper side of the bobbin.

The coil, when complete, weighed rather more than one hundred-weight and contained more than 1,500 yards of wire.

To obtain the value of the magnetic field produced by the coil at a particular point on the axis of the coil for a definite value of the current flowing through it in terms of the known dimensions of the coil, recourse could not be had directly to the well-known formula—

$$\mathrm{H} = 2\pi n\mathrm{C}\left\{(x+b)\log\left(\frac{a+d+\sqrt{(x+b)^2+(a+d)^2}}{a-d+\sqrt{(x+b)^2+(a-d)^2}}\right) - (x-b)\log\left(\frac{a+d+\sqrt{(x-b)^2+(a+d)^2}}{a-d+\sqrt{(x-b)^2+(a-d)^2}}\right)\right\}, \quad (1)$$

in which H = field strength at a point distant x from the centre of the coil, n = number of turns per square centimetre of the cross section, C = current in absolute units, $2b$ = breadth in the direction of the axis of the cross section by a plane through the axis, $2d$ = radial depth of the coil, because the coil could not be assumed to be uniform to the required degree of accuracy. The field at any point due to each layer of the coil separately was therefore calculated by means of the formula

$$\mathrm{H} = 2\pi n\mathrm{C}\left\{\frac{x+l}{r_1} - \frac{x-l}{r_2}\right\}, \quad (2)$$

in which n = number of turns per cm., $2l$ = length of the layer, r_1 and r_2 are the distances from the point under consideration to the far and near ends of the layer respectively. To obtain the field for the whole coil the fields due to each layer were then added together. The dimensions used in calculating the field due to each layer were the mean axial length and mean diameter obtained by measurement.

In this way the field of the coil for unit current flowing through it was calculated for points on the axis of the coil from 7·5 cm. to 9·5 cm. distant from the centre of the coil by steps of 1 mm. This range was selected for calculation as it is that in which the crystal is situated in the actual measurements and as it is that in which the maximum value of $H\,dH/dx$ occurs.

To obtain the value of dH/dx at the corresponding points, the values of the field strength were used to calculate the value of n in formula (1), which best fits the calculated values, using the known dimensions of the coil as the other quantities. With this value for the number of turns per square centimetre of the cross section of the coil, the values of dH/dx for the required points could be obtained from the expression obtained from (1) by differentiation with respect to x. The calculations were simplified, as in the region under consideration the change of the field strength with distance along the axis is very nearly linear. The product of the two quantities so calculated gives the values of $H\,dH/dx$ for steps of 1 mm. over the range of x of from 7·5 cm. to 9·5 cm. For positions involving fractions of a millimetre, the value of $H\,dH/dx$ can be obtained from the table by interpolation.

The effects on the value of the field strength of the thickness of the enamel insulation and of the obliquity of the windings were considered in the light of a paper by Rosa.* The length overall, including the insulation, must be used as the length of the equivalent current-sheet. The value of the correction due to the obliquity of the windings was not actually calculated, but it was seen that it was highly probable that the correction was too small to need inclusion in the present case.

Two other sources of error require investigation, the effect of the finite size of the crystal specimen actually used in the observations and deviations of the position of the specimen from the axis of the coil. The effects of the two possible sources of error were calculated at the same time as follows :—The ratio of the values of the field for a point on the axis and one a definite distance from the axis was calculated for the innermost layer of the coil and also for the outermost layer. The effect for the whole coil then lies between the values so obtained.

The value of the field for general points due to a single layer of wire is as follows :—

$$H = 2\pi nC\left[\frac{x+l}{r_1} - \frac{x-l}{r_2} + \frac{y^2}{2^2}3a^2\left\{\frac{x+l}{r_1^5} - \frac{x-l}{r_2^5}\right\}\right.$$
$$\left. + \frac{y^4}{2^2 \cdot 4^2}3 \cdot 5a\left\{\frac{x+l}{r_1^9}(3a^2 - 4(x+l)^2) - \frac{x-l}{r_2^9}(3a^2 - 4(x-l)^2)\right\} + \ldots\right],$$

* 'Bulletin Bureau of Standards,' vol. 2, p. 71.

in which the symbols have the same significance as before and y is equal to the perpendicular distance from the axis of the point under consideration. It will be seen that the first two quantities in the expression give the value of the field for a point on the axis and hence that the remainder of the expression may be regarded as giving the value of the correction due to the point not being on the axis.

The ratio of this correcting factor to the value on the axis was calculated for the innermost and outermost layers of the coil as stated for a point distant 2 mm. from the axis and 7·5 cm. from the centre of the coil. These figures were chosen because the specimen always had a radius of 2-3 mm., and it was situated usually at about 7·5 cm. from the centre of the coil during a measurement. It was found that the ratio for the innermost layer (radius = 13·8 cm.) was $1{\cdot}9 \times 10^{-5}$, and for the outermost layer was $3{\cdot}3 \times 10^{-5}$. It was thus seen that no correction need be applied because of the finite size of the specimen, as the value of $\mathrm{H}\, d\mathrm{H}/dx$ is constant over the transverse section of the specimen to an amount less than the experimental error.

The long weight-tube of the balance case was carefully adjusted once and for all so that it was accurately on the axis of the coil when the latter was mounted in position. The accidental departures from the axial position of the specimen are then quite small, and in general no correction has to be applied to the calculated value of the field. The correction, if necessary, can be obtained by calculations similar to the above.

When in use the coil stands on three teak pillars, the tops of which are provided with steel plates. The coil bobbin has three equidistant feet through which pass levelling screws. These screws serve to fix the position of the coil in the hole, slot and plane formed by the steel caps of the pillars.

Lifting the coil from the floor and mounting it in its required position on the pillars and the reverse process of dismounting present a problem on account of the weight of the coil (about one hundredweight), the constricted nature of the space in which it has to be lifted, the height through which it must be raised, and the small difference in size between the inner hole of the coil and the outer diameter of the vacuum glass of the cryostat (not more than $\frac{1}{4}$ in.). The last condition requires that the coil shall be lifted vertically and accurately concentric with the outer vacuum vessel if the coil is to be lifted without risk of breaking the vessel.

To meet these requirements as simply as possible, the following arrangement was devised and constructed for mounting the coil:—A kind of stretcher was made of two beams of wood 2 in. by 3 in. cross-section and 5 ft. long, with the necessary cross bars, and provided also with handles at the ends for lifting. In the centre of the bars is a thick piece of teak which carries a turn-table. For this latter a piece of wood was turned up with an outer diameter equal to that of the coil bobbin and having a central hole sufficiently large to allow the entry of the outer vacuum vessel. The underside of this circular piece of wood is provided with three large " domes of silence " on which it can turn. Any lateral motion is prevented by two curved guides which embrace the greater

part of the circumference. The coil rests on this turn-table and by means of it can be turned easily about a definite vertical axis. To mount the coil in position on the pillars it is first placed on the frame and arranged so that its feet are just clear of and are at the side of the pillars. Two assistants, one at each end of the frame, then lift the coil carefully in a vertical direction. When it has reached the required height, the weight is supported on two trestles and the coil on the turn-table is turned about a vertical axis until the levelling screws in the feet are over the hole, slot and plane of the pillars. The screws are then turned to take the weight of the coil and the frame is lowered to the ground, where it remains during the experiments. To dismount the coil the process is reversed. To ensure that the frame shall be lifted accurately vertically and always so that the coil is coaxial with the outer vessel of the cryostat, two guide posts attached to either side of the stone pier are provided. Blocks carried on the frame bear against the guide posts and prevent any motion either transversely or longitudinally to the frame. The guide posts are, of course, fixtures, and the frame, when not actually in use for raising or lowering the coil, remains in place on the floor below the cryostat. One of the guide posts is seen in fig. 2, the turn-table and part of the frame being also visible. By means of the frame, in spite of its fairly large weight the coil can be mounted or dismounted with relative ease and without risk of breaking the vacuum vessels by contact with the coil.

When the coil has been mounted on the pillars, levelled and adjusted for height, its position is determined by the aid of a cathetometer focussed on the upper rim of the bobbin.

(4) *The Platinum Resistance Thermometer.*

The platinum resistance thermometer of the four lead type, which is to be found at the lower end of the long weight-tube, was made specially for the purpose. A mica cross was constructed and a double set of deep slots cut in it, arranged spirally in pairs. The cross was then filled up to a cylindrical surface with wax so that the cylinder came half-way along the slots. The fine platinum wire was then wound on double and attached to the necessary leads. By this means* the wire was wound to a cylindrical shape without any sharp corners or kinks and lay half-way along the slots. The wax was then dissolved away in a solvent and the wire thoroughly annealed. A thermometer was thus obtained specially suitable for use at low temperatures. The wire was not strained in any way, and on cooling it could slip farther into the slots without straining. The thermometer was calibrated at the usual points by comparing the potential across it with that across a 10-ohms standard resistance (calibrated by the P.T.R.) in series with it. The comparison was made with the aid of a Tinsley's vernier potentiometer, the apparatus being used later with the same connections to determine the temperatures.

* This method of winding thermometers, etc., has been described by Roebuck, 'Journ. Optical Soc. Amer.,' vol. 6, p. 865 (1922).

The resistance of the thermometer at 0° was about 12 ohms. As it was required for use at low temperatures at which the Callander formula no longer holds, it was also calibrated at the boiling point of liquid oxygen. Then by employing HENNING'S data,* the temperatures can be obtained from a table given by him in terms of the resistance of the thermometer and a single constant determined by the calibration at the boiling point of oxygen. After the calibration had been completed and the thermometer sealed into the long weight-tube, a paper appeared by HENNING and HEUSE† on the calibration (comparison with the gas thermometer) of the platinum resistance thermometer between 0° and — 180°. The authors showed the temperature can be obtained in terms of the resistance of the thermometer from a formula, the constants of which can be obtained by calibrations at the boiling point of sulphur, in steam, in ice, at the sublimation point of solid carbon dioxide, and at the boiling point of liquid oxygen. As, however, the thermometer had been fixed in position in the weight-tube, it was not thought really necessary to disturb it to calibrate it further by the later method. The temperatures were therefore determined with the aid of HENNING'S 1914 data, as already mentioned. The temperatures so obtained are quite sufficiently accurate for the present purpose.

After the thermometer had been calibrated, it had to be fixed in position in the long weight-tube. Four thin copper leads of suitable length had been attached to the thermometer before calibration, and these were retained for use in the actual experiments. A number of rings of mica were next prepared. The outer diameter of the rings was such that they would just slide easily into the weight-tube, while they had a central hole cut in them of about 1·5 cm. diameter. Four small radial slots equally spaced round the circumference were cut in the outer edge of each ring. The leads were then fitted into these slots, and were fastened in position with a little hard wax. Ten rings were spaced fairly equally along the leads, so that the thermometer rested on the bottom of the weight-tube; the uppermost ring was just within the straight narrow part of the tube. When the rings were in position on the leads, the whole was slid into the long weight-tube so that the thermometer was at the lower end of the tube. The upper ends of the leads were then passed through small holes in the wider part of the tube (see fig. 1), and were sealed vacuum-tight with hard wax. To the ends of the leads were soldered short flexible wires provided with "spade" connectors at their ends. When the weight-tube is in place on the balance, these connectors are clamped under four terminals on a small ebonite block attached to the pier. Permanent leads (just visible at L in fig. 2) are taken from the terminal block to the measuring apparatus.

By means of the mica rings the thermometer leads inside the weight-tube are thus held in position, are insulated from each other, and a clear space of about 1·5 cm. in diameter is left for the entrance of the long fibre, crystal and carrier. They are not shown in fig. 1 to avoid unnecessarily complicating the diagram.

* 'Ann. der Phys.,' vol. 40, p. 635 (1914).

† 'Z. für Phys.,' vol. 23, p. 95 (1924).

(5) *Measurement of the Current in the Field Coil.*

The current for the field coil is derived from the accumulator battery of the laboratory, 48 or 64 volts being usually applied to the circuit. The current passes first through the main switch, then through an ammeter giving a rough reading of the current strength, then through the windings of the field coil connected in parallel, through a Tinsley's 0·1 ohm standard resistance capable of carrying currents up to 22 amperes, through a regulating resistance, and so back to the accumulators. On account of the very large self-inductance of the field coil, any but small currents cannot be switched off directly without risk of damage to the circuit. It was therefore arranged so that a large non-inductive resistance is switched in in parallel with the field coil before the main switch is opened. The inductive current produced on breaking the main current is then taken by the non-inductive resistance, and no damage is done to the coil. The current is, however, always reduced to a fairly small value before switching off as a further precaution.

A rough estimate of the current strength is furnished by the ammeter, so that the current can be adjusted approximately by means of the regulating resistance to any desired value. The exact value of the current strength is obtained by measuring the potential across the terminals of the 0·1 ohm standard resistance. This is done with the aid of the Tinsley's vernier potentiometer, also used for the measurements with the platinum resistance thermometer. As the instrument has a range of 1·9 volts, currents up to 19 amperes can be measured accurately in this way to within 0·0001 ampere, provided that the current remains steady to this degree of accuracy, so that the full sensitivity of the instrument can be made use of. For the larger current strengths such a degree of steadiness is not obtained, in spite of the water-cooling provided on the coil, but the current can still be measured with an accuracy quite sufficient for the present purpose.

Experimental Procedure.

The Crystals.

The substances used in the present investigation belong to the class of monoclinic double sulphates. They were chosen for investigation because they crystallise well, are stable, and because one member of this class has already been investigated.* Fairly large crystals were grown and sections prepared from those which were well formed and quite transparent. A Thomas Campbell-Smith crystal grinding apparatus was used to grind the sections. These were then ground to the shape of small cylinders with the axis of the cylinder perpendicular to the plans of the section. The cylinders were as far as possible prepared of the same weight, generally about 80 milligrams. The masses of the sections were determined with the aid of a Kuhlmann microbalance, sensitive to one-hundredth of a milligram.

* Jackson, 'Phil. Trans.,' vol. 224, p. 1 (1923).

Mounting of Crystal and Preparation of Apparatus for Measurement.

To mount a crystal on the balance for an experiment, it is first attached with a very small amount of shellac to a small carrier. This latter consists of a thin aluminium disc provided with a hook perpendicular to its plane. The crystal section is attached to the underside of the carrier, and when the whole hangs from the long fibre of the balance by means of the hook of the carrier, the plane of the carrier disc is horizontal, and the axis of the crystal section vertical.

As the mounting of the crystal and carrier on the hook of the long quartz fibre of the balance and the bringing up of the long weight-tube into position were tasks of some delicacy and required considerable steadiness and accuracy for their successful accomplishment, two special accessories were constructed to facilitate the work.

A mechanism similar to that used to adjust the position of the rider on the beam of a balance was constructed, the rider hook being replaced by a small platform. The crystal section is placed on this platform and is brought into position near the lower hook of the long fibre. The crystal is then attached to the fibre by carefully moving the platform and afterwards lowering it so as to engage the hook of the carrier with that of the fibre and so leave the crystal hanging therefrom. The operation can, by this means, be carried out easily and rapidly without the risk of breaking the fibre, which is present if the work is done entirely by hand.

On account of the presence of the mica rings in the long weight-tube, it is a matter of some difficulty to raise the latter by hand over its whole length so accurately vertically that the crystal carrier hanging from the long fibre never comes in contact with the mica rings. As this last occurrence would be very likely to break the fine fibres, either the very fine load-fibre on the balance itself or the long fibre hanging from it, an arrangement on the principle of the geometrical slide was therefore constructed. The long weight-tube can be clamped to it and raised slowly by hand, so that it moves always accurately vertically, the slide having been previously adjusted to verticality and the weight-tube to parallelism with it. The weight-tube can thus be placed in position or removed slowly, but with ease and certainty, without risk of damaging the delicate apparatus.

After the crystal has been mounted on the balance and the long weight-tube put in position, weights must be added on the other load fibre to balance the crystal and carrier. A small quartz hook provided with two cross-pieces is kept permanently suspended from the other side of the balance. From this is hung the small quartz bulb or "air-weight." To balance the load of the crystal and carrier, small quartz weights in the form of hooks are hung from the cross-pieces, various combinations of weights being tried until the total load is equal to that on the other arm of the balance to within about 0·01 milligram. This load must be slightly lighter than that of the crystal, etc., the additional weight required being obtained by reducing the pressure of the air in the balance case, so reducing the buoyancy of the "air-weight." This adjusting of the

balance to equilibrium is naturally a somewhat tedious process on account of the number of operations to be performed each time. A set of quartz weights of approximately the required total weight are first suspended from the cross-pieces, the small weight-tube is then replaced, and the arrestment of the balance is lowered by turning the small electromagnet attached to the balance case. If the load is then slightly too light, as is required, the pressure in the balance case is slowly reduced to see if the balance will swing freely in equilibrium at some pressure lower than atmospheric. If this is not the case, the pressure must be brought back to atmospheric once more, the arrestment raised, the small weight-tube removed, and one of the quartz weights exchanged for a slightly heavier one. The operations must then be repeated as before until the balance is in equilibrium at some not too low pressure. (This latter is required in the present case, as the pressure has to be further reduced to balance the pull on the crystal due to the magnetic field.)

The sensitivity of the balance is next determined. To do this the pressure in the balance case is adjusted until the spot of light reflected from the mirror on the beam comes to rest at some definite reading on the scale. This reading and that of the manometer attached to the balance case are then taken. The pressure is then altered by a small known amount and the change in position of the light spot is noted. The deflections on the scale are then known in terms of the change in the air pressure and hence also in terms of actual weights if the temperature of the air in the balance case is known. If the sensitivity of the balance is then not sufficient for the purpose in hand, the centre of gravity of the beam is then raised by slightly turning a tiny aluminium screw attached to the beam. In the present experiments this was usually such that a deflection of one millimetre on the scale corresponded to a change in weight of about one-millionth of a milligram.

After the crystal has been balanced and the sensitivity adjusted to the required amount, the apparatus can be prepared for the actual measurements. The frame carrying the cryostat is next raised and clamped in position. The coil is lifted on to the pillars with the aid of the frame and turn-table as described. By means of the levelling screws the coil is then adjusted for height and levelled. Its height is determined by means of a cathetometer. The cryostat is next filled and adjusted. With a suitable pressure in the vacuum space of the inner vessel, the outer vessel is next filled with liquid air. The stirrer is started and the cooling of the pentane bath allowed to proceed until the temperature has fallen to somewhere near the required value. The arm is then clamped to the rod of the regulator and the heating current switched on. After a little time the temperature will have become steady and observations can be commenced.

Observations.

The pressure in the balance case is then adjusted until the light-spot stands at some convenient reading on the scale. This reading and those of the pressure and temperature of the air in the balance case are then taken. The current in the field coil is then

switched on and adjusted to some definite value. The pressure of the air in the balance case is then altered until the balance is again in equilibrium at some position near the original. The system must then be left for some time to allow the air in the various parts to come once more to equilibrium with the surroundings. When this is the case, the readings of the pressure and temperature of the air in the case must be taken. The position of the light-spot on the scale is observed, and the temperature of the crystal and the strength of the current in the field coil are determined with the aid of the potentiometer. The process is then repeated for some other value of the current in the coil until sufficient number of observations have been obtained at the particular temperature.

Calculation of Observations.

In calculating the susceptibilities of the crystal, use is made of the expression

$$\mathrm{F} = \chi \,.\, m\mathrm{H} \,.\, d\mathrm{H}/dx,$$

in which

F = force acting on crystal,
χ = specific susceptibility in the direction of the axis of the crystal section,
m = mass of crystal section,
H = strength of the magnetic field.

The determination of m presents no difficulties and its value can be measured to one part in a thousand. It remains to be shown how F and H $d\mathrm{H}/dx$ are obtained from the observations.

Force acting on the Crystals.

The force acting on the crystal is given by $\mathrm{F} = \mathrm{M} \,.\, g$, where M is the mass equivalent to the change in pressure required to balance the pull on the crystal and g is the acceleration due to gravity. This force can be calculated from the expression

$$\mathrm{F} = \mathrm{M} \,.\, g = g\frac{(p_1 - p_2)}{760} \,.\, \left\{ d_t \,.\, v_1 - (d_{t'} - d_t)\, v_2 - \left(d_{t'} - \frac{d_2}{d_1} d_t \right) v_3 \right\},$$

in which

g = acceleration due to gravity,
p_1 = initial pressure in millimetres of mercury of the air in the balance case,
p_2 = final pressure,
v_1 = volume of the "air-weight,"
v_2 = volume of hook and carrier,
v_3 = volume of the crystal,
d_t = density of the air in the balance case at temperature t,
$d_{t'}$ = density of air surrounding the crystal (temperature = t'),
d_1 = density of fused quartz,
d_2 = density of crystal.

The first expression in the last bracket corresponds to the change in the buoyancy of the " air-weight " and the second and third expressions to a correcting term required because the density of the crystal is not in general the same as that of the fused quartz from which the balance is constructed, and the temperature of the air surrounding the crystal is not the same as that of the air in the balance case.

Calculation of $\mathrm{H}\,.\,d\mathrm{H}/dx$.

To obtain the value of $\mathrm{H}\,.\,d\mathrm{H}/dx$ in equation (1) from the table of calculated values, the exact position of the crystal with regard to the coil must first be calculated. The position of the middle of the crystal is first of all determined with a cathetometer, when the balance is in equilibrium with the light-spot at a known position on the scale. The height of the top of the coil is afterwards determined with the same cathetometer. The position of the crystal relative to the middle of the coil is then known when the light-spot is at the particular reading on the scale. As now, the balance is not in general brought back exactly to this position when the pressure is altered to equilibriate the pull due to the magnetic field, the exact position of the crystal with regard to the coil requires a further simple calculation. To the original position must be added the difference in the readings on the balance scale for the original and final positions multiplied by the ratio of the length of the arm of the balance beam to the distance from the balance to the scale. Thus, if the balance scale reads S_1, the distance of the middle of the crystal to the middle of the coil is x, as determined by the cathetometer, the final reading on the scale is S_2, the length of the arm of the balance beam is l_1 and l_2 is the distance of the scale from the balance, then the position in the middle of the crystal relative to the coil at the time of the actual reading is given by

$$x \pm (\mathrm{S}_1 - \mathrm{S}_2)\, l_1/l_2.$$

The value of $\mathrm{H}\,.\,d\mathrm{H}/dx$ at this point can then be obtained from the table.

Calculation of the Principal Susceptibilities.

From the values of $\mathrm{H}\,.\,d\mathrm{H}/dx$ at the point occupied by the crystal, the mass of the latter and the force acting on it, the value of χ_x, the specific susceptibility in the direction of x, can be calculated. From the values of χ_x for various definitely orientated sections, the values of the principal susceptibilities of the crystal can be obtained. The crystals at present under investigation, the paramagnetic double sulphates, belong to the monoclinic prismatic class and, in these cases, four orientated sections must be measured to provide the necessary data for the calculation of the three principal susceptibilities, χ_1, χ_2 and χ_3, and the angle ψ which χ_1 makes with the crystallographic c axis. Since, on account of the symmetry of the crystal, χ_3 lies along the b axis, the measurement of a section the axis of which is parallel to the b axis will give the value of χ_3 directly. To determine χ_1, χ_2 and ψ, three sections were prepared, the axes of which were all in the symmetry plane, and were respectively perpendicular to the (100) and (001) planes and parallel to

the a axis. χ_1, χ_2 and ψ can then be obtained from the observations by means of the following method of calculation.* Let the angle between χ_x and χ_1 be ϕ and that between χ_1 and the c axis be ψ. The value of $\Phi = \phi + \psi$ is known from the orientation of the section. Then χ_x is given by the following expressions :—

$$\chi_1 \cos^2 \phi + \chi_2 \sin^2 \phi = \chi_x$$

or

$$\frac{\chi_1 + \chi_2}{2} + \frac{\chi_1 - \chi_2}{2} \cos 2\phi = \chi_x.$$

We may rewrite the latter expression in terms of the known quantities Φ and χ_x :—

$$\frac{\chi_1 + \chi_2}{2} + \frac{\chi_1 - \chi_2}{2}(\cos 2\,\Phi \cos 2\,\psi + \sin 2\,\Phi \sin 2\,\psi) = \chi_x.$$

Putting

$$\frac{\chi_1 + \chi_2}{2} = \mathrm{A}, \qquad \frac{\chi_1 - \chi_2}{2} \cos 2\,\psi = \mathrm{B}, \qquad \frac{\chi_1 - \chi_2}{2} \sin 2\psi = \mathrm{C},$$

we obtain

$$\mathrm{A} + \mathrm{B} \cos 2\,\Phi + \mathrm{C} \sin 2\,\Phi = \chi_x.$$

For the three sections in the symmetry plane we have :—

$$\mathrm{A} + \mathrm{B} \cos 2\,\Phi_{\mathrm{II}} + \mathrm{C} \sin 2\,\Phi_{\mathrm{II}} = \chi_{\mathrm{II}},$$

$$\mathrm{A} + \mathrm{B} \cos 2\,\Phi_{\mathrm{III}} + \mathrm{C} \sin 2\,\Phi_{\mathrm{III}} = \chi_{\mathrm{III}},$$

$$\mathrm{A} + \mathrm{B} \cos 2\,\Phi_{\mathrm{IV}} + \mathrm{C} \sin 2\,\Phi_{\mathrm{IV}} = \chi_{\mathrm{IV}}.$$

These equations are then solved for A, B and C, and we have :—

$$\tan 2\psi = \frac{\mathrm{C}}{\mathrm{B}},$$

$$\chi_1 = \mathrm{A} + \frac{\mathrm{B}}{\cos 2\psi},$$

$$\chi_2 = \mathrm{A} - \frac{\mathrm{B}}{\cos 2\psi}.$$

The values for Φ_{II}, Φ_{III} and Φ_{IV}, for the sections actually measured, are as follows :— $\Phi_{\mathrm{II}} = 90^\circ$, $\Phi_{\mathrm{III}} = (\beta - 90)$, $\Phi_{\mathrm{IV}} = \beta$. ($\beta$ = crystallographic angle.)

In calculating the susceptibilities from the observational data, a small correction must be applied to allow for the force exerted on the carrier itself. This correction was evaluated by carrying out at various temperatures a series of blank experiments in which the apparatus was exactly as in the actual observations, except that no crystal was attached to the carrier.

* Finke, 'Ann. der Phys.,' vol. 31, p. 149 (1910).

Experimental Results.

Cobalt Potassium Sulphate.—The results of the measurements on the four sections of cobalt potassium sulphate are given in the following tables :—

T.	$\chi_3 \times 10^6$.	T.	$\chi_{II} \times 10^6$.
289°·9	24·9	289°·8	23·7
272·6	25·9	267·3	25·4
258·8	26·5	257·6	25·8
252·9	27·4	245·3	26·6
245·2	27·8	228·0	28·8
240·4	28·8	201·6	30·8
230·1	29·9	167·0	35·1
214·0	32·1		
194·1	35·2		
180·0	37·2		

T.	$\chi_{III} \times 10^6$.	T.	$\chi_{IV} \times 10^6$.
290°·3	28·8	286°·5	23·2
266·7	30·8	278·7	23·5
257·2	32·4	252·5	25·3
248·1	34·2	234·9	26·6
242·7	34·7	230·2	27·5
235·3	35·1	219·1	28·0
213·6	41·8	190·7	30·7
197·5	43·8	177·6	32·3

The susceptibilities given in these tables are collected together in fig. 3, in which the

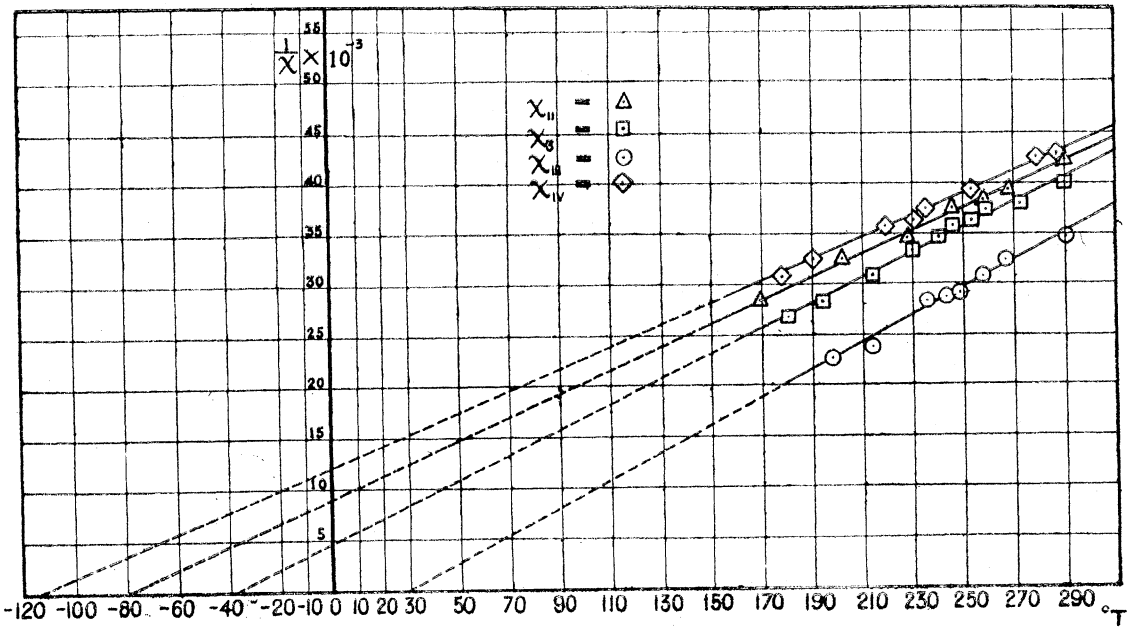

Fig. 3.

reciprocals of the susceptibilities are plotted against the corresponding absolute temperatures. To obtain the data for the calculation of the principal susceptibilities, points are read off from these curves at intervals of twenty degrees from 290° to 170°. The values of the susceptibilities calculated from these reciprocals are used as the values of χ_{II}, χ_{III}, and χ_{IV}, and are given in the following tables:—

Crystal II.			Crystal III.			Crystal IV.		
T.	$\frac{1}{\chi} \times 10^{-3}$.	$\chi \times 10^{6}$.	T.	$\frac{1}{\chi} \times 10^{-3}$.	$\chi \times 10^{6}$.	T.	$\frac{1}{\chi} \times 10^{-3}$.	$\chi \times 10^{6}$.
°			°			°		
290	42·3	23·6	290	35·2	28·4	290	43·4	23·0
270	40·0	25·0	270	32·5	30·8	270	41·3	24·2
250	37·7	26·5	250	29·8	33·6	250	39·1	25·6
230	35·4	28·2	230	27·1	36·0	230	36·9	27·1
210	33·1	30·2	210	24·4	41·0	210	34·7	28·8
190	30·8	32·5	190	21·7	46·1	190	32·5	30·8
170	28·5	35·1	170	19·0	52·6	170	30·4	32·9

The following data are also required in the calculation of the principal susceptibilities.

$$\beta = 104^{\circ}\ 55'.$$

$$\cos 2\Phi_{II} = -1, \qquad \sin 2\Phi_{II} = 0,$$

$$\cos 2\Phi_{III} = 0{\cdot}8675, \qquad \sin 2\Phi_{III} = 0{\cdot}4974,$$

$$\cos 2\Phi_{IV} = -0{\cdot}8675, \qquad \sin 2\Phi_{IV} = -0{\cdot}4974.$$

The values of the principal susceptibilities and the angle ψ calculated by the above method are given below. The values given under the heading χ_3 are those read off from the graph of $1/\chi_3$ in fig. 3 for the same temperatures as those used in the calculations of χ_1 and χ_2.

T.	$\chi_1 \times 10^{6}$.	$\chi_2 \times 10^{6}$.	$\chi_3 \times 10^{6}$.	ψ.
°				° ′
290	28·4	23·0	24·6	20 2
270	30·8	24·2	26·2	20 34
250	33·6	25·5	28·0	20 11
230	37·0	27·0	30·1	20 43
210	41·1	28·7	32·6	20 26
190	46·2	30·6	35·4	20 1
170	52·7	32·8	38·8	20 5

It will be seen that the values of ψ given in the last column are constant to within the experimental error, the greatest deviation from the mean value, 20° 17′, being 26′.

In the following table are given the values of the principal molecular susceptibilities corrected for the diamagnetic properties of the anion and the water of crystallisation present in the molecule. The reciprocals of these quantities are plotted against the corresponding absolute temperatures in fig. 4, the values of $1/\chi_{3m}$ being, however, the actual observed values.

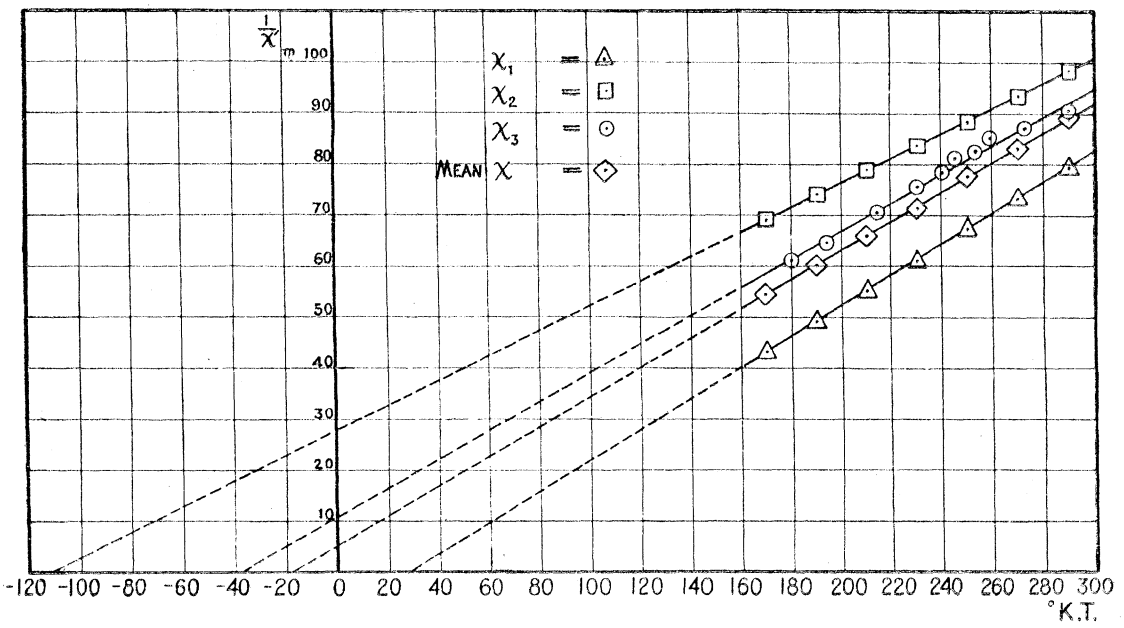

Fig. 4.

T.	χ'_{1m}.	χ'_{2m}.	χ'_{3m}.	Mean χ'_m.
°				
290	0·0126	0·0102	0·0109	0·0112
270	0·0136	0·0107	0·0116	0·0120
250	0·0148	0·0113	0·0124	0·0128
230	0·0163	0·0119	0·0133	0·0140
210	0·0181	0·0127	0·0144	0·0151
190	0·0203	0·0135	0·0156	0·0165
170	0·0232	0·0145	0·0172	0·0183

Nickel Ammonium Sulphate.—The following results have been obtained with a section prepared from a crystal of nickel ammonium sulphate and so orientated (axis of cylinder parallel to b axis of crystal) as to give χ_3 directly. The observations thus permit a calculation of the magneton number and are given here to show what is to be expected

for this crystal. The corrected molecular susceptibilities are given in the third column, the reciprocals being plotted in fig. 5.

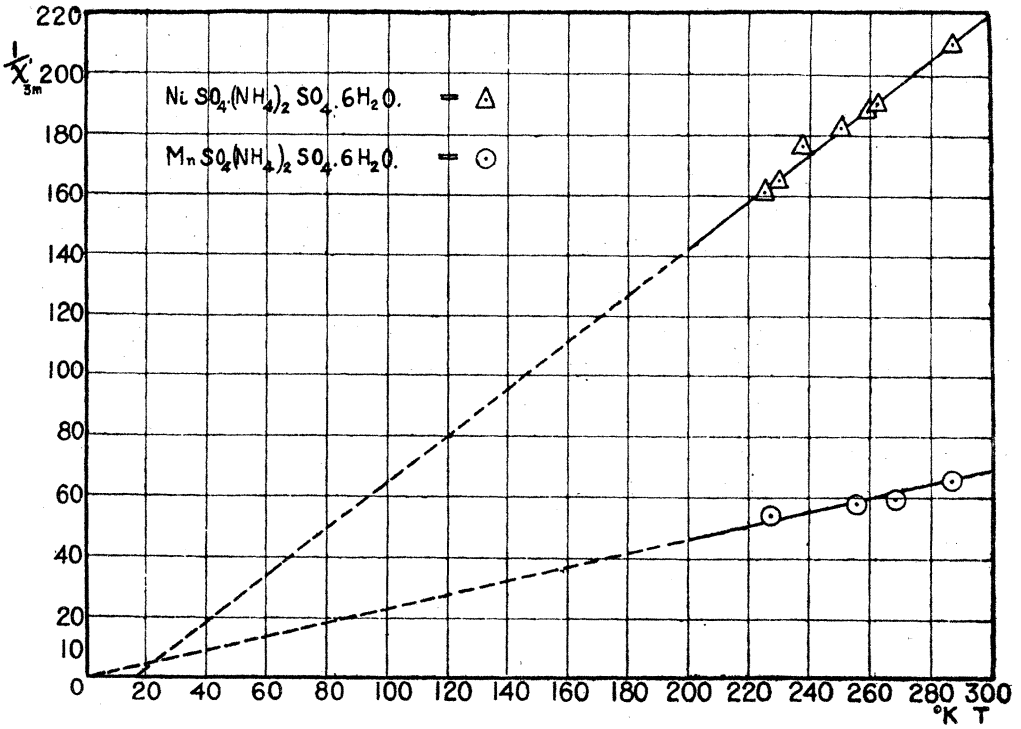

Fig. 5.

T.	$\chi_3 \times 10^6$.	χ'_{3m}.
°		
287·3	11·6	0·00476
262·6	12·8	0·00525
259·8	12·9	0·00529
250·7	13·5	0·00548
237·9	13·9	0·00568
230·4	14·9	0·00606
225·3	15·2	0·00620

Manganese Ammonium Sulphate.—In May, 1925, the writer, in collaboration with Prof. W. J. DE HAAS, carried out, at the Physical Laboratory of the University of Leiden, a series of measurements of the principal susceptibilities of manganese ammonium sulphate over the range of temperature obtainable with liquid hydrogen (20° to 14° K.). It is hoped to publish the results shortly. It was found that the principal susceptibilities of this crystal all followed the law $\chi\,(T + \Delta) = \text{constant}$, Δ being positive for one axis, negative for another, and zero for the third, χ_3. The last susceptibility thus follows

CURIE's law over this range of temperature. The magneton number calculated from the observations on χ_3 was found to be 29·4. Measurements were later undertaken with the present apparatus to extend the observations above recorded to higher temperatures. They were commenced on a section of the substance, the axis of which was parallel to the *b* axis of the crystal. After a few observations had been made, further measurements were rendered impossible by the crystal rapidly losing its water of crystallisation in the partial vacuum of the balance case. The rate at which the water was lost by the crystal became so great that the spot of light moved over the balance scale at a speed of several centimetres a minute, so effectively preventing further work with that particular section. Manganese ammonium sulphate is for some reason difficult to obtain in large perfectly transparent crystals, the larger crystals being rendered slightly cloudy by inclusions of white opaque matter. The dehydration of the crystal commenced at one of the small flaws and then spread throughout the mass of the section. The few observations which were obtained before the dehydration became too rapid are given below, as they provide some confirmation of the Leiden results. The accuracy of these observations, on account of the disturbances due to the incipient dehydration, is not very great, and it is hoped to repeat the measurements when perfect crystals can be obtained for the purpose.

T.	$\chi_3 \times 10^6$.	χ'_{3m}.
°		
286·8	39·2	0·0152
268·7	43·6	0·0169
255·7	44·6	0·0173
227·0	47·9	0·0185

Discussion of the Results.

It will be seen that the relation between $1/\chi$ and the absolute temperature is linear for the range of temperature investigated, the susceptibilities following the law $\chi\,(T + \Delta) =$ constant. The values of $1/\chi$ for the principal susceptibilities of cobalt potassium sulphate lie on three approximately parallel straight lines. This result is in agreement with that of previous investigations.* It cannot yet be stated definitely whether the CURIE constants for the three principal axes are in reality equal and the present deviations are due to experimental errors, or whether they are actually only approximately equal. Further experimental data are necessary before the point can be settled definitely.

The values of Δ_1, Δ_2 and Δ_3 in the expression $\chi_n\,(T + \Delta_n) =$ constant for cobalt potassium sulphate are -28, $+111$ and $+37$ respectively. Comparing these values with those obtained previously for the isomorphous compound cobalt ammonium sulphate, viz., 9, 38 and 17 respectively, we see that they follow algebraically the same

* FOËX, 'Ann de Phys.,' vol. 16, p. 174 (1921); JACKSON, 'Phil. Trans.,' vol. 224, p. 1 (1923).

order, but whereas with the latter substance the Δ's are all positive, Δ_1 in the case of cobalt potassium sulphate is negative. The connection between the values of Δ along the principal axes and the structure of the crystal is obviously a complicated one, and no attempt to determine the nature of this connection can profitably be made for these monoclinic crystals at the present stage of the investigation. A further investigation of cobalt potassium sulphate at still lower temperatures would very probably furnish interesting results, especially in the case of χ_1.

The mean of the principal susceptibilities of this crystal follows the law $\chi\,(T + 19) =$ constant with a value of the CURIE constant giving a WEISS magneton number equal to 26·1. A direct measurement of the mean susceptibility with the powdered substance gave the law $\chi\,(T + 21) =$ constant, with a WEISS magneton number of 25·3.* The agreement between the two independent determinations is thus fairly good, but the results for the crystal are higher than those for the powdered substance, the values for the molecular susceptibility at 290° K. being 0·0112 and 0·0104 respectively.

The results may further be compared with the measurements of FINKE.† He gives the values of the principal susceptibilities of cobalt potassium sulphate at atmospheric temperature. His data are as follows :—

$$\chi_1 = 66{\cdot}5_9 \times 10^{-6},\ \chi_2 = 49{\cdot}6_2 \times 10^{-6},\ \chi_3 = 77{\cdot}1_5 \times 10^{-6},\ \psi = -21^\circ\ 20',$$

the susceptibilities being calculated per unit volume. The mean susceptibility per unit mass calculated from these figures is $29{\cdot}1_6 \times 10^{-6}$ compared with $25{\cdot}3 \times 10^{-6}$ obtained in the present determination. The values given by FINKE are almost certainly too high. It will be seen that there is no resemblance between the present results and those of FINKE.

The principal susceptibility of nickel ammonium sulphate also follows the law $\chi\,(T + \Delta) =$ constant with a Δ equal to -16. The WEISS magneton number calculated from the observations is 16·0. Previous measurements on the powdered material gave a magneton number 15·9 and the law $\chi\,(T + 4) =$ constant. The value of the mean susceptibility at atmospheric temperature was $10{\cdot}7 \times 10^{-6}$. For χ_3 we find $11{\cdot}6 \times 10.^{-6}$ FINKE gives the value $17{\cdot}9_3 \times 10^{-6}$ for κ_3 for nickel ammonium sulphate. Re-calculated as a specific susceptibility this value becomes $9{\cdot}36 \times 10^{-6}$.

The above work was carried out in the Davy Faraday Laboratory of the Royal Institution, London, and I wish to express my cordial thanks to the Director of the Laboratory, Sir W. H. BRAGG, for placing at my disposal all the necessary facilities. My thanks are also due to the technical staff for valuable assistance in the construction and assembling of the apparatus.

* JACKSON, 'Roy. Soc. Proc.,' A, vol. 104, p. 671 (1923).

† FINKE, 'Ann. der Phys.,' vol. 31, p. 149 (1910).

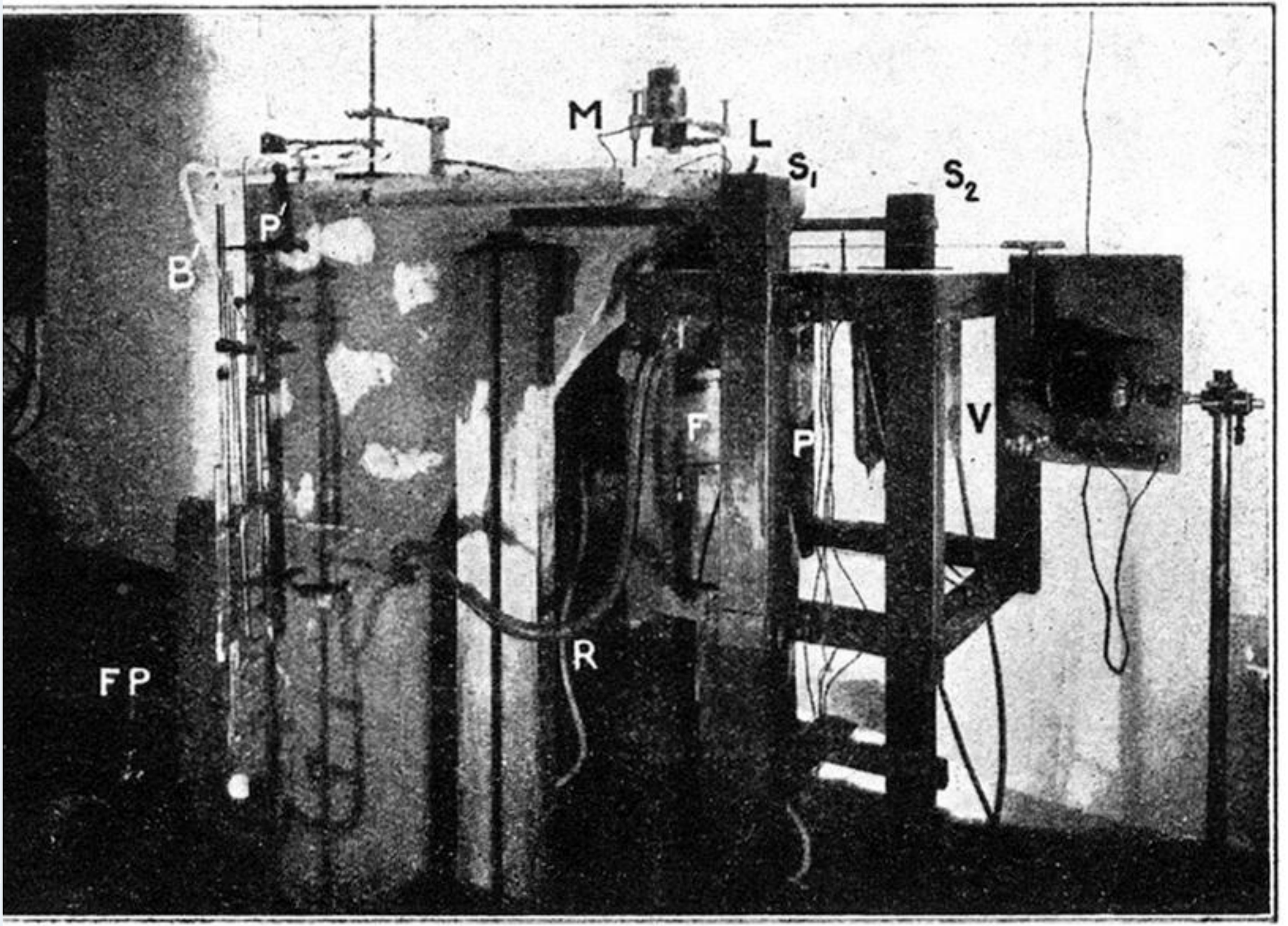

Fig. 2.

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