Citation:

H. Kamerlingh Onnes & Perrier, A., Magnetic researches. X. Apparatus for the general cryomagnetic investigation of substances of small susceptibility, in:
KNAW, Proceedings, 16 II, 1913-1914, Amsterdam, 1914, pp. 689-699

Physics. — *"Magnetic researches. X. Apparatus for the genèral cryomagnetic investigation of substances of small susceptibility."* By Prof. H. Kamerlingh Onnes and Dr. Albert Perrier. Communication N°. 139*a* from the Physical Laboratory at Leiden. (Communicated by Prof. H. Kamerlingh Onnes).

(Communicated in the meeting of October 25, 1913).

§ 1. *Introduction.* This paper contains the full description of the apparatus used in the investigation of Communication III (Comm. N°. 122*a*, continued in IV, Comm. N°. 124*a*) of this series (Proceedings of May 1911). Various circumstances have retarded the extensive description which was promised there instead of the rough sketch.

The construction of the apparatus to be described forms part of a more general scheme to gradually obtain the necessary appliances for the investigation of weak magnetisation at low temperature. In doing this we did not confine our attention to special measurements, but intended to enlarge with as many appliances as possible the almost completely unknown "technique" of investigations in this field.

On the one hand the measurement of magnetic forces, on the other that of magnetic couples suggest themselves. The ballistic method (measurement of flux) is only applied in the study of ferromagnetism [1]).

The method of couples is specially suitable for crystals and for isotropic bodies, which by their shape are seemingly magnetically anisotropic (e. g. ellipsoids). The apparatus with which our first measurements on the susceptibility of liquid and solid oxygen were made (Comm. N°. 116, Proceedings April 1910) is based on this method.[2]) In a modified form this piece of apparatus will, we hope, be soon utilized in the cryomagnetic investigation of crystals.

In measuring forces a non-homogeneous field is used. Two cases have to be distinguished here.

For an object of small dimensions (the volume of which is v and the volume-susceptibility K) placed in the plane of symmetry between the poles of a magnet the force in the direction of the middle of the interferrum is given by

$$F = vKH\frac{\partial H}{\partial y}$$

where H indicates the intensity of the field and y the coordinate at right angles to the field.

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[1]) In some cases which we will not dwell upon here this method might be resorted to.

[2]) The apparatus used by Weiss and Kamerlingh Onnes for the investigation of ferromagnetism at low temperature (Comm. N°. 114) belongs to the same type.

For an object in the shape of a rod of uniform section s, the axis of which is in the plane of symmetry of the poles and passes through the middle of the interferrum, the relation is

$$F = \frac{K}{2} s \left(H''^2 - H'^2 \right)$$

if H'' and H' are the values of the field-strength at the ends of the rod. When dealing with bodies of small dimensions by the method of FARADAY, the spherical object is placed where $H \frac{\partial H}{\partial y}$, therefore the force is a maximum. This is the method of procedure specially used by CURIE in his classical researches.

The rod-method, though applied long ago for measuring the susceptibility of liquids by QUINCKE's method, was hardly used at all in investigations on solids until 1910, when PASCAL adopted it in his important series of magneto-chemical researches [1]).

This is certainly curious, as the principle of the method is very simple and direct, but even more so as the disposition itself offers important advantages over the other methods. If one end of the rod is placed in the middle of the interferrum and care is taken that the other end is as far removed from it as possible, H'' obtains a maximum value and H' remains a quantity which may be neglected or need only be taken into account as a correction.

The susceptibility is thus given by a single field-strength which is much more easily determined than the product $H \frac{\partial H}{dy}$, which has to be derived from several values of H, not to mention the fact, that the measurement itself of H in the middle of the interferrum, where the field is most nearly uniform, can be carried out much more accurately than at the point where the field is least uniform.

An *absolute* measurement by this method can therefore lead to a much more trustworthy result. Moreover in using a rod a much higher *sensibility* can be obtained, on the one hand because a larger quantity of the substance can be utilized, on the other hand because the intensity of the field in the middle of the interferrum can be raised to a much higher value without any objection, which is not by any means the case in the other method. Finally, as the field near the middle of the interferrum can usually be made approximately homogeneous over a space of 1 cc., it is of no great importance at what point exactly within that space the end of the rod

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[1]) P. PASCAL, C. R. 150, p. 1054. 1910. The priority of this application belongs to GOUY. C.R 109, p. 935. 1889.

under investigation is placed, so that as regards this a rough adjustment will be sufficient; the exact opposite holds when it is desired to place a body at the place of maximum action.

There are cases, however, in which only the method of maximum attraction can be applied, e.g. when the susceptibility depends on the field or when the available quantity of the substance is limited (e.g. on account of its rarity).

On the ground of the above considerations we have made it our object to construct a piece of apparatus *which in the first place is suitable for measurements with objects in the shape of an elongated cylinder, which may further, without important change, be adapted to the study of small objects placed at the point of maximum-attraction and finally, in addition to being suitable for solids, may also be used for the investigation of liquids, either by enclosing them in the movable part of the apparatus or by surrounding it as a bath.*

The ease with which our apparatus may be adapted to the various requirements has shown itself a great advantage in our experiments [1]).

§ 2. *General arrangement of the apparatus* (comp. figure and perspective drawing). The main part of the apparatus is a *carrier* movably suspended along the axis of an enclosure which has the shape of a body of revolution. This enclosure is closed airtight, seeing that it must be capable of being exhausted and that it must be possible to maintain throughout the apparatus any pressure below atmospheric. This requirement from the side of cryogenics has its influence on the choice of most other parts.

The carrier the motion of which is guided along the vertical, carries at its lower end the experimental object which is placed between the poles of an electro-magnet with horizontal axis. The magnetic attraction or repulsion acts along the vertical and is *measured* by compensating it by means of the electromagnetic attraction of two co-axial magnetic coils, one of which is attached to the carrier while the other one is fixed. The force between the two coils is given by

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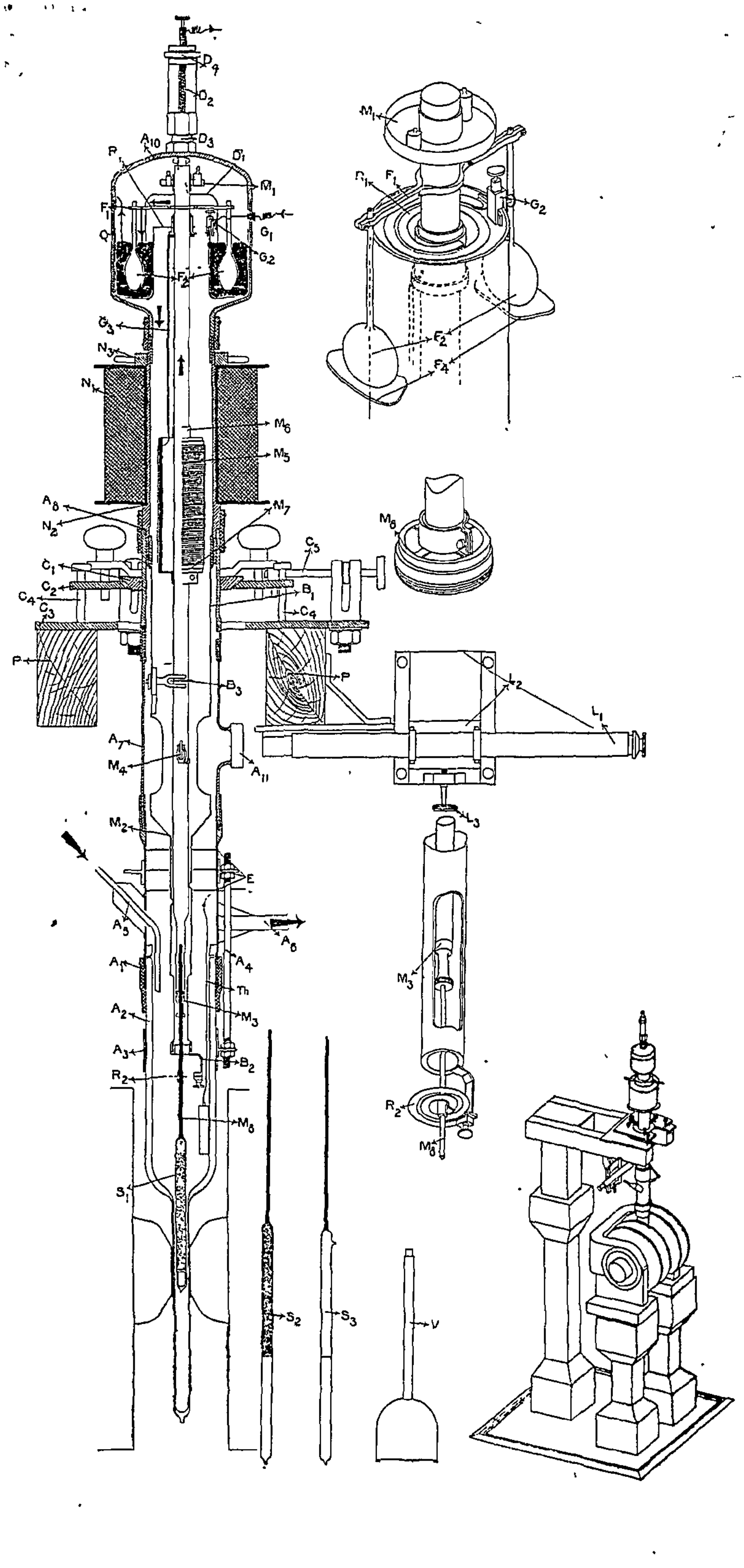
[1]) We may here recapitulate the various apparatus which in the mean time form the complete scheme planned by us:

depending on the use of couples	*a.* apparatus with ellipsoid (Comm. N⁰. 116) *b.* „ for crystals (to be constructed)
depending on the use of forces	*c.* hydrostatic apparatus (Comm. N⁰. 116) *d.* apparatus for objects in the shape of spheres or cylinders (this Comm.)

a. has been used for liquid and solid oxygen, *c.* for liquid gases, *d.* for liquified or solidified gases and various solids.


D_4
D_2
A_{10}
D_3
D_1
P_1
M_1
F_1
G_1
O
G_2
F_2
O_3
N_3
N_1
M_6
M_5
A_8
N_2
M_7
C_1
C_3
C_2
B_1
C_4
C_4
C_3
P
B_3
P
L_2
L_1
A_7
M_4
A_{11}
L_3
M_2
E
A_5
A_6
A_4
A_1
Th
A_2
M_3
A_3
B_2
R_2
M_8
S_1
S_2
S_3
V
M_1
D_1
F_1
G_2
F_2
F_4
M_6
M_3
R_2
M_8

$F = c i_m i_f$, where i_m and i_f represent the currents in the movable and fixed coils and c is a constant which is determined once for all by using known forces (weights)[1].

The electromagnetic compensation has for its sole object making a balance with the forces to be measured: the weight of the carrier itself is balanced *hydrostatically* by means of two floats immersed in mercury; the principle is therefore similar to that of a constant-volume hydrometer.

§ 3. *The various parts of the apparatus.*

a. The enclosure of the cryostat. The space inside the enclosure A of the apparatus is divided by screens E, which prevent exchange of heat between the two parts. The cryogenic part below the screens contains everything connected with the establishment of low temperatures, in the chamber above the screens which remains practically at constant temperature, all the delicate parts for the measurement of the forces are brought together.

The wall of the cryogenic space below E is of german silver. It is joined airtight by means of the india-rubber ring A_1 to the vacuum glass A_2 which contains the bath of liquid gas. The liquid gas is supplied by the german silver tube A_5, the vapours are carried off by A_6. The steel capillary of a heliumthermometer Th is soldered through the wall of the cap.

The upper part of the vacuum-glass is comparatively wide (6 cms), so that the liquid level falls very slowly during the evaporation, which as we shall see is of importance. By means of the copper ring A_3 and the rods A_4 the vacuumglass is firmly connected to the cap, that the considerable forces arising from changes of internal pressure may not change its position.

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[1] For keeping in equilibrium an apparatus of the general type under consideration any kind of force may be used which can be changed gradually without touching the carrier. We can thus work equally well with a given compensating force (definite weights) and changing field (regulating the current through the electromagnet) as with a given field and changing electromagnetic compensating force. When our apparatus was first constructed we did not possess the necessary appliances for accurate field-measurements, and in examining the dependence of the phenomena on the temperature we had to make ourselves independent of the change of the field by confining ourselves to the investigation of the magnetisation at different temperatures at a few field-stengths chosen beforehand and kept constant each time during the experiments

For a modification in which the electromagnetic compensation is replaced by a compensation by weights we refer to a forthcoming description of the apparatus which was used in their investigations by KAMERLINGH ONNES and OOSTERHUIS (Comm. n⁰. 129*b* etc.).

The part of the enclosure above the screens E is entirely of brass with the exception of tube A_7, which is glass.

At A_8 and at the joint with the upper part of the enclosure above N_3, A_9, (the letter is omitted in the figure,) the parts fit each other with friction, which is of great convenience in the building up of the apparatus.

At the upper end the enclosure A is enlarged to a wide chamber containing the parts which serve to keep the carrier afloat. It is closed by an arched cover A_{10}, which again fits on the wall with friction. To this part of the enclosure is attached the german silver tube B_1B_2 which narrows down towards the bottom and to which is fastened at B_2 the spring which guides the movable carrier in a vertical direction and the two stopping pins B_3, which prevent the carrier from moving too far up or down. [1])

The enclosure further serves to suspend the entire apparatus from two horizontal beams; by means of the ball socket C_1C_2 the direction of the axis of the apparatus may be changed, without altering its height. The plate C_2 bears with three adjusting screws C_4 on the ground plate C_3, which in its turn is fixed to the beams, and may be shifted in a horizontal plane in two directions at right angles to each other by means of the screws C_5. With the adjusting screws the apparatus may be moved 3 cms up or down; this is necessary in using FARADAY's method in order to *find* the maximum force by displacing the apparatus with respect to the electromagnet.

b. The movable carrier with adjuncts. In the figure the carrier is indicated by M. It consists of a long thinwalled tube of brass, at the same time light and firm, lengthened by a narrow german silver tube which at its end carries a thread M_3. The experimental objects are also fitted with german silver top-pieces, which may be screwed on to M_3. They are thus easily attached to and detached from the carrier. At convenient heights the brass tube is provided with the following parts: the springs R_1 and R_2 which guide the motion, the marks M_4 for the purpose of reading the position of the carrier, a stopping ring B_3 for confining the motion between the pins, the electromagnetic coil M_5 moving with the carrier, the carriers F_1 of the floats and a scale M_1. As regards these various parts the following may be added.

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[1]) The german silver tube might without disadvantage have been replaced by a simpler arrangement; in the design experiments were contemplated which were not carried out. If due care is taken, the stopping pins may also be dispensed with.

c. Vertical guidance of the carrier. The space between the experimental objects and the inner wall of the vacuum-glass can sometimes not be more than a few tenths of a millimetre when measurements with strong fields are to be made; with the slightest movement of the axis of the carrier from its original position owing to a small asymmetry in the action of the electromagnet or any other cause the carrier would not be able to move up and down freely. This difficulty was quite satisfactorily overcome by guiding the carrier in its up and down motion by the aid of two flattened spiral springs [1]. The outer end of both is fastened to the stationary part of the apparatus, the inner end to the carrier and the plane of the springs is placed at right angles to the axis of the carrier. By the device of using flat springs a movement of the middle in the plane of each of the spirals is almost completely prevented. Usually the upper spring R_1 attached to the carrier remains the same, while each separate experimental object is provided with its own spring, which is removed from the apparatus with the object.

d. The hydrometric equilibrium. To keep the carrier afloat on mercury the upper chamber of the apparatus is provided with a ring-shaped trough Q (in our experiments of glass, later on of china) which is centred on the axis of the carrier. The latter is fitted with a horizontal arm F_1, in which at both ends are fixed the tubes of the floats F_2, glass bulbs, the shape of which is not unlike a flattened ellipsoid. The tubes of these bulbs which are of *very small section* are the only part that projects above the mercury. The section has to be small in order that the upward pressure of the mercury shall vary very little, if the apparatus is to be sensitive to a very small change of the vertical force acting on it. But the size cannot fall below a certain limit, because the tubes must also serve to compensate the diminution in upward pressure in the bath on the experimental object, owing to evaporation.

This compensation is effected by raising the level of the mercury. For this purpose use is made of a plunger D_1, a small glass flask of a shape corresponding to that of the trough which is moved up and down by means of a rod D_2 with thread and milled head D_4 passing through a stuffing box D_3. This contrivance, which was found

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[1] Springs of that kind are made by cutting on the lathe a spiral groove 0.2 to 0.3 m.m wide in a plate of german silver (comp. perspective figures $R_1 R_2$). By giving different widths to the spiral strip for a given diameter springs may be obtained of any desired degree of sensibility. The inner end is soldered to a small tube, the outer end is fixed in a clamping screw.

very serviceable supplies the advantage that at the beginning the hydrostatic pressure of the bath need only be approximately compensated, which is done by placing a weight about equal to the pressure on the scale M_1; the accurate adjustment is made afterwards by regulating the level of the mercury.

e. The electromagnetic compensation. The fixed coil N_1 consists of 1275 turns of insulated copper wire, wound on a brass frame, sliding closely over the outside of the enclosure; the coil rests on the ring N_2 and is fixed at the top by the screw N_3. The movable coil M_5 has on the one hand to be as light as possible, on the other it has to produce as great a force as possible; account was therefore taken of the fact that for a given weight it is an advantage to make the radius of the coil large and the number of turns small. The coil contains 248 turns ($d = 0.7$ mm.) in two layers, wound on a thin-walled ebonite tube, which is held between two supporting brass rings $M_6 M_7$ in the shape of wheels, which may be clamped to the carrier at the desired height.

The wire which carries the current to the movable coil passes through and is insulated from the cover G_1 and is connected to the clamping screw G_2 of the upper spiral spring; the current passes through this spring to the ring by which it is attached and which is insulated with ebonite, along the wire G_3 to the coil and back through the carrier itself, the rod of the floats, a platinum wire dipping in the mercury, the mercury and finally a second platinum wire, which carries the current to the cover.

The electromagnetic system is calibrated once for all by fixing to the lower end instead of the experimental object a small scale, on which definite weights are placed, and regulating the current until the balance is obtained.

The level at which the carrier floats, is read on a glass plate M_4 with a scale division in tenths of a millimetre, which is focussed with a microscope L_1 magnifying about 40 times. For this purpose a window of thick plane-parallel glass is sealed on to an opening in the glass tube A_7. On the side opposite to the microscope behind the tube an electric glowlamp is placed in such a position that the scale divisions are seen light on a half-dark background: in this manner it may be very sharply determined when the cross fibre of the microscope exactly coincides with the division.

§ 4. *The experimental tubes.* The substances investigated by us (salts, powdered metal) are all enclosed in glass tubes, concerning which the following may be mentioned.

It is desirable, that the upward pressure due to the bath changes as little as possible when the liquid level falls through evaporation; for this reason the tubes end at the top in thin glass rods M_8 of 2to 2.5 mm. diameter. The lower spiral spring R_2 and the thread by means of which the tube is screwed to the carrier are sealed to this rod with some KHOTINSKY glue. As regards the shape of the tube we have used different forms. Tubes as shown at S_1 are used for substances of high susceptibility, for which the magnetic action on the glass or on the bath plays a subordinate part, so that for them it may be entirely neglected or else a correction may be easily applied. The tube is filled with the substance, when it is still open, at the bottom putting in small quantities at the time, which are evenly compressed in order to obtain a tight filling and at the same time a uniform density throughout the whole tube; the substance is then closed in with a small plug of glass wool to prevent its being heated during the sealing of the tube and the tube is sealed off at the air-pump. The smaller the susceptibility of the substance the greater influence the susceptibility of the air would have and the morene cessary it is to be assured of a good vacuum; a high vacuum, however, is obviously unnecessary.

In cases where account has to be taken of the susceptibility of the glass, which may give rise to fairly strong forces [1]), tubes of type S_2 are used, the lower half of which, separated from the upper half by a glass partition, is exhausted. When this partition is placed on the level of the axis of the poles, the correction for the glass disappears, as it is divided symmetrically with respect to the axis; the susceptibility of the substance is in that case directly compared with that of the vacuum. Type S_3, which does not require further explanation is meant for the measurement of the susceptibility of the liquid in the bath.

§ 5. *Additional apparatus.* The electromagnet is a copy of WEISS's electromagnet which was used in previous researches of this series. The yoke is, however, placed horizontally this time, in order to leave the space below the apparatus completely free (comp. perspective drawing). Usually poles were used of the shape shown, the flat end-faces of which had a diameter of 40 mms. At a polar distance of 15 to 20 mms. the topographical inequality of the field about the middle of the interferrum was not above 0.1 $^0/_0$ within a

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[1]) The susceptibility of glass at low temperature was determined by us in Comm. No. 124*a*, p. 6.

distance of 1 cm. The field-strengths were measured with a Cotton-balance of the usual pattern by W. C. Weber of Zürich.

The circuits of the fixed and movable coils are entirely independent of each other: each of them contains an accurate ammeter, a commutator and rheostats, in which the current is reversed on commutation, in order to neutralise any magnetic influence on the ammeters. They are within reach of the observer seated in front of the microscope.

The field-strength of the electromagnet is given by the current flowing through it; the field was not adjusted until the magnetising current had been several times reversed.

If the evaporation of the bath in the apparatus as described is too strong, as is the case when liquid hydrogen is used, it is diminished by surrounding the bottom half of the vacuum-glass with a vacuum-glass with liquid air.

§ 6. *Method of observation.* Passing by certain simplifications which were often possible we proceeded as follows.

The enclosure and the carrier (without experimental tube) are first adjusted so that the common axis is vertical and passes through the centre of the interferrum. When this position is arrived at, the apparatus is not moved sideways any more.

The experimental tube is then screwed to the carrier and its spring clamped. By means of the weight on scale M_1 the carrier is made to float on the mercury approximately at the desired level and care is taken that the movable coil has the correct position relatively to the fixed coil. The apparatus as a whole is then moved in a vertical direction until the lower end of the experimental cylinder falls about in the line of the axis of the poles, after which the cylinder is adjusted more accurately by turning the apparatus about the ball socket C_1. When finally the poles have been brought at the right distance, everything is ready for the observations at ordinary temperature.

In changing to low temperatures as much weight is added to the scale as agrees approximately with the upward pressure of the bath to be expected and the cover is fastened to the apparatus air-tight by means of the india-rubber ring; after drawing out the poles, the vacuum-glass is placed carefully round the experimental tube, connected airtight and centred in a manner similar to that used in the apparatus of Weiss and Kamerlingh Onnes. After having made sure that everything is airtight, the liquid gas is admitted to the vacuum-glass, the poles are brought back to their position, and the carrier is adjusted to its zero by means of the plunger; the currents in the large electromagnet i_e and in the fixed coil i_f are adjusted to suitable whole numbers and the current i_m (in the movable coil) regulated by a gradual change of the resistance until the carrier has come back to the zero. The current i_m is then noted down and the operations are repeated for the 4 possible combinations of the currents i_e, i_f and i_m. Before and after each observation the zero-position of the carrier is observed or again adjusted; when the change amounts to only a few tenths of a millimetre, there is no objection to do this, more simply than by means of the plunger, by shifting the microscope a little.

(To be continued).

Chemistry. — *"The application of the theory of allotropy to electromotive equilibria."* By Prof. A. Smits. (Communicated by Prof. J. D. van der Waals.)

1. I communicated already before[1]) that the investigation for testing the theory of allotropy with different elements and anorganic as well as organic compounds was in progress. The investigation of the metals, which had been started with *tin* and *mercury*, was somewhat delayed, because all the time had to be devoted to the study of *phosphorus* and *mercury-iodide*, so that only comparatively shortly ago the metals could be taken in hand again.

As may be supposed as known, the theory of allotropy rests on this *fundamental assumption* that every phase of a system that behaves as a unary one is at the least built up of two kinds of molecules which are in internal equilibrium, and must necessarily be taken as the components of a pseudo-system. This theory comprises, therefore, all possible states of aggregation of a substance, and on account of the importance of its conclusions its principial interest lies in the region that has been least investigated up to now, viz. that of the solid state.

Now it is clear that the experiments which are carried out to test this theory are undertaken in the first place to prove that the different states of aggregation and particularly the solid phases of a substance which presents the phenomenon of allotropy, are really mixtures, and in internal equilibrium, for every time that this succeeds a confirmation of the said theory has been found. In the second place an attempt may be made by a continuation of the in-

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[1]) These Proceedings, April 26, 1912, XIV, p. 1199.