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**Physics.** — "*Researches on the magnetization of liquid and solid oxygen.*" By H. KAMERLINGH ONNES and ALBERT PERRIER.  
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§ 1. *Introduction.* It is scarcely necessary to remark that the investigation of the magnetic properties of oxygen at low temperatures has long occupied a position on the programme of the cryogenic laboratory, or that this has been considered one of the most important items on the programme since the investigation of both liquid and solid oxygen has been made possible by the perfecting of the methods<sup>1)</sup> of obtaining detailed series of measurements at constant temperatures in the region of liquid hydrogen. Indeed, while the strongly magnetic properties of oxygen of themselves select it from all other substances as especially suitable for the study of para-magnetism, we can in the meantime for no other substance obtain the *magnetic equation of state*<sup>2)</sup>, which gives a representation of the magnetic properties of a substance in the three states of aggregation at as many successive temperatures and pressures as possible.

The investigation of oxygen at very low temperatures and at pressures that can easily be realised was expected to give at once results of much importance.

CURIE<sup>3)</sup> found for gaseous oxygen between 20° C. and 450° C. that the *specific susceptibility* (magnetization per gram for  $H=1$ ) was inversely proportional to the absolute temperature, and FLEMING and DEWAR<sup>4)</sup> concluded from their latest measurement of the susceptibility of liquid oxygen at its boiling point that CURIE's law was obeyed down to  $-183^{\circ}$  C.

Does the specific susceptibility continue to increase so strongly at low temperatures or does it approach a limiting value? Is oxygen in the solid state ferro-magnetic? Does the magnetization finally at extremely low temperatures perhaps begin to decrease and disappear completely at the absolute zero?<sup>5)</sup>

<sup>1)</sup> H. KAMERLINGH ONNES, These Proc. Sept. 1906, Comm. from the Leyden labor. no. 94f (1906).

<sup>2)</sup> H. KAMERLINGH ONNES, Commun. from the Leyden labor. Suppl. no. 9 p. 28.

<sup>3)</sup> P. CURIE, Ann. chim. phys. (7) 5 (1895) p. 289.

<sup>4)</sup> FLEMING and DEWAR Proc. Royal Soc. London 63, p. 311, 1898.

<sup>5)</sup> It has since appeared that the magnetization of ferro-magnetic substances does not yet give any justification when the temperature is lowered to the melting point of hydrogen for the assumption that the electrons whose motion causes magnetization are frozen fast to the atoms and that therefore this disappearance at the absolute zero may be expected. (P. WEISS and H. KAMERLINGH ONNES, These Proc. Jan./Febr. 1910, Comm. from the Leyden Labor no. 114 p. 9).

These are questions which, considering the possibility of obtaining important contributions to the knowledge of the influence of density upon susceptibility by high pressures in the region where the gaseous state of aggregation changes continuously into the liquid make it a very attractive work to realise, even from a purely experimental point of view, the representation to which we have just referred.

The work was commenced though only when Prof. WEISS extended his magnetical researches to very low temperatures and the measurements on the magnetization of ferro-magnetic and cognate substances at very low temperatures, which were communicated to the February Meeting<sup>1)</sup>, were undertaken. With that investigation which was carried out at the same time, the present one is very closely related, and for part of them we made use of the same appliances. In our present investigation we have also in various ways made use of Prof. WEISS's method<sup>2)</sup> of determining the magnetization by means of the maximum couple exerted by a magnetic field of variable direction upon an ellipsoid of the experimental substance, a method which had been entirely successful in the other research. We must also express the great advantage we derived from the continued support given us by Prof. WEISS, and we take this opportunity of gratefully acknowledging our indebtedness to him.

The change with temperature of the specific susceptibility of oxygen, the investigation of which was our first object, is of particular importance seeing that CURIE's law follows from LANGEVIN's kinetic theory of magnetism<sup>3)</sup>. It was soon apparent to us that this law was not valid for oxygen, as was thought, down to  $-183^{\circ}$  C., but that it would have to be replaced by another. According to the important paper of DU BOIS and HONDA communicated to the January Meeting — our experiments had already been completed at that time — various elements were found for which CURIE's law did not hold at temperatures above  $0^{\circ}$  C. This at once increases the importance of the further investigation of oxygen, for which over a definite region of temperature CURIE's law is valid, while over another region it obeys a second law, viz.: *that of inverse proportionality to the square root of the absolute temperature*. The results concerning this law and also concerning the probability of a sudden change in the value of the specific susceptibility on solidification will be discussed in § 5.

<sup>1)</sup> P. WEISS and H. KAMERLINGH ONNES. These Proc. Jan./Febr. 1910. Comm. fr. the Leyden labor. no. 114 (1910).

<sup>2)</sup> P. WEISS. Journ. de phys. 4e série t. VI, p. 661; 1907.

<sup>3)</sup> LANGEVIN. Ann. chim. phys. (8) 5, p. 70; 1905.

We have been occupied with another question besides the change of specific susceptibility with temperature, which was suggested both by the experimental results obtained by FLEMING and DEWAR and by the theories of LANGEVIN and WEISS.

In the experiments of the first-named there appears sufficient evidence for the conclusion that there is a decided diminution of the susceptibility as the strength of the field increases (the diminution is of the order of 10% in a field of 2500 gauss). Now, according to the theory of LANGEVIN para-magnetic substances must, it is true, exhibit this phenomenon, but calculation from his formulae limits the magnitude of this change to less than 0.1% in the case of liquid oxygen at its boiling point. Should a higher value than this be obtained, then one would be led to assume the existence of a WEISS molecular field<sup>1)</sup>. We arranged our experiments so that the liquid and the solid oxygen could be subjected to a field of 16000 gauss, a field very much stronger (about six times) than that used by FLEMING and DEWAR, so that we might expect the phenomenon which appeared in the course of their experiments to be exhibited to a much greater degree in ours even at the same temperatures as were used by them. If what was observed by FLEMING and DEWAR could really be ascribed to the beginning of saturation then the theory would further lead us to expect that as the temperature sank the change would strongly increase (becoming infinite at  $T=0$ ), and that in our experiments with liquid hydrogen it would become very striking. We have, however, observed only small deviations, which we shall discuss further in § 5.

As regards the experimental methods employed by us in our investigation, two completely different schemes were adopted: on the one hand was measured the magnetic attraction exerted upon a column of the liquid, and on the other the maximum couple exerted by a homogeneous field upon an ellipsoid. The second method is more especially suitable for comparative measurements and can also be used for frozen oxygen; the first can be used only for the liquid phase, but on the other hand it makes very trustworthy absolute measurements possible; we have therefore adopted it as the basis of our other measurements. In the carrying-out of each method further precautions are still desirable, so that while we are busy pushing on the investigation, we propose at the same time to repeat it in part in order to increase the accuracy of the results obtained by taking such further precautions as have appeared possible in the course of the work.

<sup>1)</sup> Weiss, L'hyp. du champ moléc. loc. cit.

*Liquid oxygen I.*

§ 2. *Method of the magnetic rise.* As mentioned above, we have rendered the method of the magnetic rise employed by QUINCKE, DU BOIS and other observers suitable for use at low temperatures.

One limb of a vertical O-shaped tube, the upper portion of which contains the gaseous, and the lower the liquid phase of the experimental liquefied gas was placed between the poles of a magnet whose field was horizontal.

Let  $H$  be the field, ( $H'$  the field in the other limb is supposed to be so small that  $(H'/H)^2$  is negligible),  $g$  the acceleration due to gravity,  $z$  the difference in height of the levels of the liquid under the influence of  $H$ ,  $\rho$  and  $\rho_0$  the densities of the liquid and of the gaseous phases respectively,  $K$  and  $K_0$  their respective volume susceptibilities, then

$$(K-K_0) H^2 = 2z (\rho - \rho_0) g \quad . \quad . \quad . \quad (1)$$

or, by introducing the absolute specific susceptibility  $\chi$

$$(\chi\rho - \chi_0\rho_0) H^2 = 2z (\rho - \rho_0) g$$

If  $\chi = \chi_0$  then the equation becomes simply

$$\chi = \frac{2zg}{H^2}, \quad . \quad . \quad . \quad . \quad . \quad . \quad (2)$$

which is the formula we have used for our calculations.<sup>1)</sup>

So there are striking advantages offered by this method particularly for an absolute measurement, on account of its applicability to the case of a liquid in equilibrium with its own vapour. There are only two magnitudes to be determined, the distance  $z$ , which can be measured very accurately with a cathetometer, and the field  $H$ ; nor have we to know the density of the liquid in order to be able to find the specific susceptibility.

*Magnetic rise apparatus.* It is a very easy matter to cause an ordinary liquid to ascend under the influence of magnetic attraction, but the experiment is attended by serious difficulties when one has to deal with a liquefied gas. Boiling must be completely avoided, and care must be taken that the vaporization is unnoticeable. The first precaution is necessary because the motion of the liquid or of its surface would render adjustment quite impossible, and the second

<sup>1)</sup> In § 5 we shall give the reasons why we think that  $\chi = \chi_0$ , and should it be possible that this is not the case there is still the greatest probability that  $\chi_0 < 1.5 \chi$ ; in the most unfavourable case at the boiling-point the correction remains below 0.002 in value, while at lower temperatures it is quite negligible on account of the small value of  $\rho_0$ .

is necessary that the total quantity of liquid may not appreciably alter during the measurement of one rise. Moreover magnetic action itself increases the difficulties; it is easy to see that it can occasion the formation of gas-bubbles which divide the column of liquid into two parts, so that the one portion remains suspended between the poles, while the other falls back again. In that case measurement of the ascent is out of the question.

Starting from the thermodynamic potential it appears that in every case the relation

$$2gy\rho > K(H^2 - H_y^2)$$

must hold, where  $H$  is the field at the surface of the liquid, and  $H_y$  the field at a distance  $y$  below the surface of the liquid. These conditions shew that there is a limit to the intensity of the fields in which measurements may be made, for they necessitate a range of extended fields (in this case in a vertical direction). Conical pole-pieces are thus as a matter of fact barred.

After several preliminary experiments an apparatus was constructed, the most important part of which consisted of two concentric double-walled vacuum tubes, with which we already succeeded in obtaining rather successful measurements. The walls of the double vacuum tube were not silvered, so that we were able to watch how the liquid behaved during the experiments. From the experience thus acquired the improved apparatus which we shall now proceed to describe was designed and constructed.

It will be seen that the construction of the apparatus lays a very heavy tax upon the art of the glass-blower<sup>1)</sup>. As before, the chief part consisted of two independent U-shaped vacuum tubes, the one fitting inside the other. The double walls of each tube are completely silvered on the vacuum side, except in the case of the inner tube, where the distance which the liquid ascends is left free, and in the outer where a sufficient length is left unsilvered to leave a strip of a few millimeters breadth through which the level of the liquid can be read. One of the tubes completely surrounds that portion of the other which contains liquid; this we call the protecting tube. The narrowest portion  $M_1$  (fig. 2) is placed between the poles of the electro-magnet. The narrow limb of the inner tube must of course be perfectly cylindrical. The other limb is enlarged and serves as a reservoir. In order to be able to apply equation (2) all care was taken that the temperature of the liquid and vapour up to a height

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<sup>1)</sup> The double vacuum tube was prepared by Mr. KESSELRING, Laboratory glass-blower, and the remainder by Mr. FLIM, technical assistant at the Laboratory.

somewhat greater than that reached by the column of liquid was everywhere the same both in the wide and in the narrow tube; and further care was taken that where the temperature of the vapour above the liquid in the upper parts of the apparatus changes to ordinary temperature it was as far as possible the same at the same height in the two limbs of the O-shaped space. With this end in view the liquid in the inner tube was, by means of the magnetic field, repeatedly moved up and down under constant vapour pressure, until we might assume that in this tube equilibrium was sufficiently well attained. To make this equilibrium possible the inner tube is surrounded with liquid at the same temperature as that which the liquid in it must attain. In the outer or protecting tube the liquid is kept constantly in motion by means of a stirrer consisting of a brass ring  $S_1$  that can be moved up and down; it is possible to do this and still keep the space closed by utilising the flexible rubber tube  $S_2$ . The vaporization in the inner tube is thus very small (between 0.5 and 1 litre of gas measured under normal atmospheric pressure escapes per hour).

Notwithstanding all these precautions temperature differences must still be encountered. In the liquid, in which the convection currents maintaining heat-equilibrium can be followed by the small particles which they carry along with them, these temperature differences must have been very small. In the gas layer in the upper portion of the O-shaped space there must indeed have been considerable differences; but on account of the small density of the gas, these have but small influence upon the difference of level in the two limbs, and, moreover, that influence may be almost entirely neglected seeing that the observations are simply comparative measurements with and without the magnetic field. Now, care has been taken that the temperature over the distance that the liquid rises can vary but slightly, while in the upper portions of the tube practically the same state of affairs is maintained during both observations. We have therefore omitted the correction that should still have to be applied for possible temperature differences.

Comparing the positions of the liquid in the narrow cylindrical tube with and without the magnetic field also reduces the correction for capillarity to the insignificant differences in form of the menisci, and this correction, too, we have omitted.

The inner and the outer tubes are closed independently of each other by means of the German-silver caps  $P_1, P_4, Q_1, Q_4$  (fig. 1); the junction is made air-tight by the rubber sleeves  $M_0, N_0$ , which at the same time unite the two tubes firmly together. Liquid oxygen

is introduced into the protecting tube through the small tube  $P_s$ , and into the inner tube through  $Q_s$ . The two tubes  $P_s$  and  $Q_s$  lead the vaporized oxygen through the valves  $P_v$  and  $Q_v$  (fig. 2) to two gasometers. Two manometers  $P_m$  and  $Q_m$ , the latter of which is provided with an indicator  $Q_i$ , so that small vapour pressures may be read off accurately, serve at the same time as safety valves. It is not necessary that the oxygen in the protecting tube should be as pure as that in the inner tube; for the latter, with which the observations were made, very pure oxygen was used.

A double sliding movement  $R$  allowed an easy adjustment of the apparatus each time, so that the meniscus in the measuring tube just reached the desired point in the field between the poles, usually in the axis of the pole pieces.

*Course of a series of measurements.* The field is brought to the desired strength and by means of  $R$  the meniscus is made to rise to the desired point, which is read off on a small scale. Then the meniscus is moved up and down several times while care is taken that the field slowly increases. In this way the temperature is made everywhere the same and the walls of the tube are wetted. While the field has the desired value the position of the meniscus is read off; then a reading is made while the field is off; after the meniscus has been three times allowed to rise somewhat higher than the desired position, another reading is made while the field is on; once more a reading is made with the field off and so on several times. In this manner the error arising from vaporization of the liquid during the adjustment of the cathetometer is eliminated<sup>1)</sup>. It is not essential to know the position of the level in the other limb of the tube; so as to be able to take account of this, we ascertained the ratio of the cross-sections of the two limbs of the tube.

We have further made sure that the residual magnetism exerted no appreciable influence upon the position of the meniscus after the current was cut off. For this purpose a feeble current was sent through the coils in the direction opposite to that which had just been broken. Had the residual field exerted any appreciable influence we should have seen first a further sinking of the level, and then a rise as the current was slowly increased. This has not been observed.

We used the same electro-magnet as was used for the cryogenic investigation of the ferro-magnetic metals<sup>2)</sup>, to which we must refer

<sup>1)</sup> To control the position of the meniscus without the magnetic field, we measured the quantity of gas vaporized (cf. preceding page).

<sup>2)</sup> P. WEISS and H. KAMERLINGH ONNES, l. c.

for details regarding its construction. It was only necessary to replace the conical pole-pieces by cylinders with flat ends. Their distance apart was micrometrically adjusted to 25 mm. and controlled with an accurate callipers. We may here remark that between the measurement of the ascent and that of the field, the pole-pieces remained clamped tight to the cores, so the adjustment of the distance could give rise to no error.

Since in the subsequent calculation the strength of the field is involved to the second power, and since we are concerned with an absolute measurement, we endeavoured to make our measurement of the field strength as trustworthy as possible with our present appliances. With this end in view we measured the strength of an arbitrarily chosen standard field by two different processes, and we compared the strengths of the fields used in our experiments with this standard by successively withdrawing the same coil attached to a ballistic galvanometer from the standard field and from the various fields which we desired to measure.

The standard field was set up with the same flat pole-pieces at a distance of 9 mm. apart, and with a current of 5 amp. All precautions were taken to ensure the demagnetization of the magnetic cycle beforehand. This field was first measured by means of *COTTON's magnetic balance*<sup>1)</sup>. As is well known this method consists of equilibrating weights of a total mass  $m$  against the ponderomotive power of the field  $H$  on a straight portion of length  $l$  of a conductor through which a current flows of intensity  $i$ ; then we get

$$H = \frac{mg}{il} \cdot 10.$$

For the degree of accuracy, however, which we wish to reach, several corrections must be taken into account. In the first place the various parts of the balance were accurately calibrated. The length  $l$  of the current element was determined micrometrically and on the dividing engine, and so also was the distance between the arcs of the balance which distance ought to be the same throughout seeing that the arcs must be accurately concentric. The very small deviations from this were allowed for by means of a ballistic investigation of the topography of the field. The balance arms of the weights and of the current element were measured with the cathetometer. The topographical study of the field also gave us the

<sup>1)</sup> For this method of measuring the field and for the magnetic balance see: P. WEISS and A. COTTON, *Le phénomène de ZEEMAN pour les trois raies bleues du zinc*, Bull. Séances Soc. franç. de phys. 1907, p. 140, also J. de phys. 1907.

correction necessary for the force exerted upon the second straight element of the balance (i. e. that outside the pole-gap). The sum total of these positive and negative corrections came to some units per thousand.

The greatest care had to be devoted to the absolute value of  $i$ , which was measured by means of an accurate ammeter by SIEMENS and HALSKE. This was calibrated in absolute ampères by comparing on the potentiometer the potential difference between the terminals of an international ohm (or for the stronger currents of 0,1  $\Omega$ ) with the electromotive force of a WESTON cadmium cell. For the requisite accuracy of the measurements the influence of neighbouring instruments or currents upon the ammeter, or of its position in the earth's field were by no means negligible; we got rid of almost all these irregularities by a suitable adjustment of the distances and of the positions of the rheostats, and we eliminated further possible remaining errors by so connecting *all the conductors* that the currents *in all* except the ammeters could be reversed *at the same time*. Finally we always used the ammeters in the same position with respect to the earth's field as that in which they had been calibrated.

When all calculations and corrections had been completed it was found that the strength of the standard field was 9857 gauss according to this method.

The *second method* by which the value of the same field was found consisted of the sudden withdrawal from between the poles of the magnet of a coil of wire of which the area encircled by the current was known. The change thus caused in the number of induction lines embraced by the coil was compared by means of a ballistic galvanometer with the number of induction lines embraced by a solenoid the dimensions of which were accurately known.

The coil consisted of 19 turns of silk-insulated wire, 0.25 mm. thick, wound round a cylinder of ebonite, 20 mm. in diameter. The dimensions were obtained by various measurements with the micrometer screw and the dividing engine, and were repeatedly controlled. At the same time a control coil was constructed by winding bare copper wire in a helical groove cut in the curved surface of a cylinder of ebonite; the area encircled by the current was then measured for this control coil by the same methods and with the same precautions as were adopted in the case of the first. The ratio of the two was in agreement with the ratio of the deflections of the ballistic galvanometer which were obtained by connecting the two coils in series with the galvanometer and then withdrawing them successively from an unchanged magnetic field. We may further

say that we had previously verified the absence of magnetic properties from the ebonite by means of an apparatus after CURIE in which we utilised the attraction in a non-uniform field.

For the measurement of the field there were placed in circuit with the galvanometer the coil on the ebonite cylinder, a manganin resistance to regulate the sensitivity, a secondary coil of 500 turns fitting round the standard solenoid, and finally, an electromagnetic arrangement which could be used as a damper if desired. We also allowed for the very small deviations from the law of proportionality between the deflections of the galvanometer and the quantities of electricity, which had been determined for the galvanometer (one of the DEPREEZ-D'ARSONVAL type) by a previous investigation. The solenoid was constructed with the greatest accuracy by winding bare copper wire on a core of white marble <sup>1)</sup>.

The standardisation of the galvanometer was made by reversing the current in the solenoid; the observations made by withdrawing the coil from the field always took place between two standardisations of the galvanometer; there was, however, no change in the galvanometer constant to be observed. The corrections and precautions necessary in obtaining the strengths of the current are the same as in the case of the balance, and have already been described. The final result of this ballistic method is

9845 gauss.

The relative difference between this and the value given by COTTON'S balance is therefore 0.0012; and this can be neglected especially when one remembers that almost every one of the numerous measurements necessitated by the one method as much as by the other, beginning with the adjustment of the field by means of the ammeter, is accurate only to 0.0005. It may be useful to comment here upon a particular point that increases the difficulty of obtaining this agreement and therefore enables us to rely more upon the correctness of the numbers which we have obtained. The equation for COTTON'S balance involves the strength of the current in the *denominator*, while this magnitude in calculating according to the ballistic method occurs in the *numerator*; a systematic error there-

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<sup>1)</sup> For the dimensions and the description of the solenoid and galvanometer see: P. WRISS, Mesure de l'intensité d'aimantation à saturation en valeur absolue, Arch. Sc. phys. et nat. February 1910, J. de phys. May 1910.

fore in the *absolute* number of ampères would, of necessity, occasion a relative difference *twice as great* between the values of the field obtained by the two methods (the same ammeter was used with the balance and with the solenoid).

We have given the ballistic method a somewhat greater weight than the other on account of the smaller number of corrections it involved, and thus we have finally taken as the value of the standard field

9850 gauss.

Once this standard field was definitely fixed all other measurements could be rapidly made by the ballistic method described above.

For the conical pole-pieces which are employed in experiments according to the maximum couple method, and which give much more powerful but much less uniform fields, we used a coil of 7 to 8 mm. diameter accurately centred on the axis of the pole-pieces. In this case direct comparison with the standard field just mentioned was not possible since the flat polepieces had to be screwed off to make room for the conical poles. To meet this case the area of the small coil encircled by the current was determined once and for all by withdrawing it from the standard field *before* the flat pole-pieces were removed, and comparing the change thus brought about in the number of the induction lines with those of the solenoid by means of the ballistic galvanometer.

All the measurements that we have given up to the present refer to the field in the centre of the space between the poles. For the few exceptional values of the field, and, consequently, of the ascent of the liquid oxygen for which it was necessary to cause it to rise pretty far above the axis of the pole-pieces, the field was determined at those points by simple ballistic comparison with the fields on the axis, and we made use of the cathetometer to adjust the position of the small coil.

*Results of observations and calculations.*

Series of observations with the apparatus with unsilvered walls.

T A B L E Ia.		$t = -183^{\circ}.0 \text{ C. } ^1)$		
Position of meniscus.	Obs. rise $z'$ in cm.	diff. in height with reservoir $z$ in cm.	$H$ in gauss.	$\frac{z}{H^2} \cdot 10^7$
Level of axis	1.032	1.061	2980	1.194
"	1.646	1.690	3727	1.246
"	1.656	1.701	3727	1.224
axis + 2.44 cm.	3.024	3.110	5182	1.158
axis	3.198	3.289	5205	1.244
"	4.16	4.278	5848	1.251
axis + 2.44	4.90	5.050	6570	1.168
axis	5.124	5.270	6600	1.210
axis + 2.44	7.87	8.094	8075	1.242
axis + 2.44	9.20	9.462	9043	1.158*

The difference in height  $z$  was obtained from the observed ascent from  $z = z' (1 + 0.0285)$ . The observation \* was very difficult and is little reliable.

The deviation in the case of the observation in a field of 5182 gauss is probably due to a mistake of 1 in the number of whole millimeters which were read off, but of this we are not certain.

Deviations from proportionality with  $H^2$  are considerable but by no means systematic. If we take the mean of all the measurements with the exception of the last in which special difficulties were encountered we reach the value

$$\frac{z}{H^2} = 1.209 \cdot 10^{-7},$$

and for the specific susceptibility with  $g = 981.3$  for Leiden

$$\chi_{90^{\circ}.1 \text{ K.}} = 237.3 \cdot 10^{-6}.$$

<sup>1)</sup> The boiling point of oxygen according to H. KAMERLINGH ONNES and C. BRAAK These Proc. Oct '08. Comm. fr. th. Leyden labor. N<sup>o</sup>. 107a § 6.

T A B L E Ib.				$t = -201^{\circ}.75 \text{ C.}$
Position of meniscus.	Obs. rise $z'$ in cm.	diff. in height with reservoir $z$ in cm.	$H$ in gauss.	$\frac{z}{H^2} \cdot 10^7$
Level of axis	1.192	1.222	2980	1.376
"	1.893	1.944	3727	1.399
"	1.881	1.935	3727	1.393
"	3.643	3.747	5205	1.383
"	4.623	4.752	5848	1.389
"	5.91	6.078	6600	1.395
axis + 2.5 cm.	7.376	7.586	7421	1.378
axis + 2.5 cm.	8.715	8.963	8069	1.372

$$\text{mean } \frac{z}{H^2} = 1.386 \cdot 10^{-7}$$

whence it follows that  $\chi_{71^{\circ}.35 \text{ K.}} = 272.0 \cdot 10^{-6}$ .

Finally, at  $-209^{\circ}.2 \text{ C.}$  a single observation was made. The rise was 6.115 cm. in a field of 6600 gauss, which with the correction for the sinking in the reservoir gives

$$\frac{z}{H^2} = 1.444 \cdot 10^{-7} \text{ and } \chi_{63^{\circ}.9 \text{ K.}} = 283.4 \cdot 10^{-6}.$$

We shall now give the series of observations made with the silver-walled apparatus which we have already described.

T A B L E IIa.				$t = -183^{\circ}.0 \text{ C.}$
Position of meniscus.	Obs. rise $z$ in cm.	diff. in height with reservoir $z$ in cm.	$H$ in gauss.	$\frac{z}{H^2} \cdot 10^7$
axis	1.060	1.090	2980	1.227
"	1.669	1.746	3727	1.235
"	3.169	3.258	5183	1.213
"	3.220	3.340	5198	1.225
"	4.035	4.148	5807	1.230
"	4.093	4.208	5848	1.230
"	4.101	4.216	5848	1.233
"	5.119	5.262	6578	1.216
axis + 2.5 cm.	7.750	7.967	8075	1.224
"	8.950	9.201	8659	1.227
axis + 3.5 cm.	9.226	9.484	8808	1.222
"	9.266	9.525	8808	1.228

For this apparatus  $z = z' (1 + 0.0280)$ .

The mean value of  $\frac{z}{H^2}$  is  $1.226 \cdot 10^{-7}$ , whence it follows that

$$\chi_{90^{\circ}.1 \text{ K.}} = 240.6 \cdot 10^{-6}.$$

T A B L E IIb.				$t = -201^{\circ}$
Position of meniscus	Obs. rise $z'$ in cm.	diff. in height with reservoir $z$ in cm.	$H$ in gauss	$\frac{z}{H^2} \cdot 10^6$
axis	1.195	1.228	2980	1.38
"	1.879	1.932	3727	1.39
"	3.625	3.726	5205	1.37
"	4.567	4.695	5848	1.37
axis + 3.5 cm.	5.461	5.614	6399	1.37
axis + 2.5 cm.	5.832	5.995	6567	1.39
axis	5.852	6.016	6600	1.38
axis + 3.5 cm.	6.463	6.644	6986	1.36
axis + 2.5 cm.	6.890	7.092	7169	1.38
axis + 3.5 cm.	8.207	8.437	7863	1.36
axis + 2.5 cm.	8.654	8.892	8069	1.36
axis + 3.5 cm.	8.988	9.240	8212	1.37
axis	8.913	9.162	8212	1.35

Mean of all observations is 1.375 whence it follows that

$$\chi_{71^{\circ}.35 \text{ K.}} = 269.9 \cdot 10^{-6}.$$

T A B L E IIc.				$t = -208^{\circ}.2$
Position of meniscus	Obs. rise $z'$ in cm.	diff. in height with reservoir $z$ in cm.	$H$ in gauss	$\frac{z}{H^2} \cdot 10^6$
axis	1.277	1.313	2980	1.478
"	1.996	2.052	3727	1.477
"	3.813	3.920	5205	1.447
"	4.841	4.977	5848	1.461
axis + 2.5 cm.	6.012	6.180	6567	1.438
axis	6.094	6.264	6600	1.438
"	6.113	6.284	6600	1.443
axis + 2.5 cm.	7.146	7.346	7169	1.429
axis + 3.5 cm.	8.579	8.819	7863	1.426

Mean of all observations 1.448, whence it follows that

$$\chi_{61^{\circ}.9 \text{ K.}} = 284.2 \cdot 10^{-6}.$$

Finally for finding the specific susceptibility the density of oxygen was found from the formula<sup>1)</sup>

$$\rho = 1.2489 - 0.00481 (T - 68).$$

From table II we obtain

$$K_{90^{\circ}.1 \text{ K.}} = 275.2 \cdot 10^{-6}$$

$$K_{71^{\circ}.35 \text{ K.}} = 332.8 \cdot 10^{-6}$$

$$K_{61^{\circ}.9 \text{ K.}} = 359.0 \cdot 10^{-6}.$$

Table III gives  $\chi\sqrt{T}$  for each of the temperatures and for each of the series.

Series with the first apparatus			Series with the improved apparatus		
$T$	$\chi \cdot 10^6$	$\chi\sqrt{T} \cdot 10^3$	$T$	$\chi \cdot 10^6$	$\chi\sqrt{T} \cdot 10^3$
90.1	237.3	2.25	90.1	240.6	2.283
71.35	272.0	2.29	71.35	269.9	2.279
63.9	283.4	2.26	64.9	284.2	2.289
mean		2.27			2.284

There is no systematic change to be noticed in the product  $\chi\sqrt{T}$ ; the greatest deviation from the mean is 1% with the first apparatus, and only  $\frac{1}{4}$  % with the second; moreover the deviations in the two series at corresponding temperatures are in opposite directions. Hence within the limits of accuracy of the observations the specific susceptibility can be represented by the formula

$$\chi = \frac{2284}{\sqrt{T}} \cdot 10^{-6}.$$

In the comparative measurements which we shall describe in the sequel we shall find the same law, at least as far as its form is regarded. For the discussion of this point we refer to § 5.

The differences between the various values of the ratio  $\frac{\chi}{H^2}$  are greater than we should be led to expect from the accuracy obtained

<sup>1)</sup> BALY and DONNAN. J. Chem. Soc. 81 (1902) p. 907.

(0,05%) in the measurements with the cathetometer of the displacements of the level, and from the accuracy of the measurements of the field-strengths, of which a discussion is given above. It is certain that the cause of these deviations must arise from a source other than the measurement of these two data, though we cannot with certainty indicate what this may be.

We may in the meantime remark that, at least in the case of the first series, the unsteadiness of the apparatus in the vertical direction in the not quite homogeneous field, and the slight inconstancy of the temperature have certainly been contributory causes of these deviations, since the second apparatus which was improved exclusively in these directions gave much more regular results. This remark, however, does not seem to account sufficiently for certain appreciable changes that occurred without any noticeable corresponding irregularity in the pressure or in the convection current of the liquid, while there was also no noticeable change in the shape of the meniscus.

### *Liquid oxygen II.*

§ 3. *Measurements by the method of the maximum couple exerted upon an ellipsoid.* Further comparative measurements for liquid oxygen at various temperatures were obtained by means of the method of the maximum couple exerted by a uniform field upon an ellipsoid. This method has already been described and discussed in connection with the research on ferro-magnetic substances<sup>1)</sup>; it will be sufficient to discuss the modifications which were found to be necessary owing to the particular circumstances under which the method had to be applied to the present research.

In the first place on account of the small value of the susceptibility it was necessary to make the couple to be measured as large as possible; with this end in view we chose an *oblate* ellipsoid of revolution, instead of a prolate; its axis of revolution was placed horizontal in a field which could turn round a vertical axis.

The ratio that is taken between the axes is not a matter of indifference; for a given major axis the couple, which is proportional to  $(N_1 - N_2)v$ , is a maximum for a ratio of the major to the minor axis that is only slightly smaller than 3; we have therefore taken this value of the ratio for the construction of the ellipsoids.

We used the same electromagnet as served for the measurements made by WEISS and KAMERLINGH ONNES (loc. cit.). Two pairs of pole-

<sup>1)</sup> P. WEISS, J. de phys. (4) 6 (1907) p. 655. P. WEISS and H. KAMERLINGH ONNES, Comm. N<sup>o</sup>. 114 These Proc. Jan /Febr 1910.

pieces were used; first the cylindrical pole-pieces with quite flat end surfaces that had been used for the measurement of the magnetic rise, and then truncated conical pole-pieces the end surfaces of which (slightly concave, see in this connection p. 818) were 4 cm. in diameter, and the side surfaces of which were connected by convex surfaces of revolution to the cylinders that formed the cores; these were 9 cm. in diameter. These pole-pieces were constructed to give the strongest possible field when the distance between the poles was taken to be 20 m.m. By this means a field of about 16000 gauss was obtained.

Our observations were made with an ellipsoid that was diamagnetic with respect to the surrounding medium — a solid silver ellipsoid immersed in a bath of liquid oxygen. The ellipsoid was turned by the "Société genevoise pour la construction d'instruments de physique" from a block of very pure MERCK silver. A preliminary experiment showed that it was very slightly diamagnetic with respect to air, and that this was quite negligible with respect to the liquid oxygen. The axes were measured microscopically on the dividing engine; this gave major axis = 1.0973 cm. and axis of revolution = 0.3654 cm.

Furthermore, two intermediate ordinates parallel to the axis of revolution were measured on the dividing engine, and they were found to be 2% greater than the corresponding ordinates of a perfect ellipse with the same axes. This deviation from ellipsoidal shape was confirmed by a direct determination of the volume from the weight and the density, which gave

$$0.2329 \text{ c.c.}$$

while calculation from the dimensions of the axes gave

$$0.2308 \text{ c.c.}$$

In the calculations we made use of the value 0.2329.

The cryogenic apparatus, essentially the same as that used by WEISS and KAMERLINGH ONNES is shown in Pl. I fig. 3. Once more we see the *cover B*, the *adjusting tube f'*, and the *holder b'*. The cover with its various parts: the cap with the stuffing-box *D*, glass tube *C*, window with plane parallel glass plate *C<sub>1</sub>*, the system *BG* for adjusting the whole apparatus, the tension rods *B<sub>1</sub>* for supporting the DEWAR tube, the helium thermometer *θ*, the little screens to protect the upper portions of the apparatus from cooling, etc. is just the same as before. The DEWAR tube is of the same shape, but the lower portion is of greater diameter. The only difference between the adjusting tube *f'* and that which was used in the other investigations is that the lower portion *f'<sub>1</sub>* is of greater diameter.

The *holder* and the *torsion spring* are, on the other hand, completely altered. On account of the smallness of the couple to be measured all foreign magnetic actions had to be eliminated as carefully as possible. Preliminary experiments showed us that a metallic holder could not be used, not only on account of the traces of para- or ferro-magnetic impurities that are never absent from workable metals but also on account of the difficulty of keeping the surface sufficiently clean; this difficulty was encountered repeatedly in the silver ellipsoid that we used in our experiments, and it is probable that the constant contact of the hands with iron tools plays a part in causing it. Glass seemed to be by far the most suitable material both on account of the absence of inherent magnetization and of the fact that the surface on account of its smoothness can be kept quite clean. The holder which we finally adopted was made completely of glass: it consists of a tube  $b'$  5 mm. in diameter that at  $b'_3$  is drawn out to a narrow but thickwalled stem, 0.7 mm. in diameter. To this stem the silver ellipsoid was attached; for this purpose a hole of sufficient width to fit was bored along one of its greater diameters and the ellipsoid was then fixed at the desired height by means of a little wax that completely filled the narrow space between the glass and the metal. The tube was then pumped free from air and sealed off, so that the liquefaction of the air that it would otherwise contain would be prevented. The flat mirror for measuring the angle of torsion and the oil-damper were also attached to the holder.

*The torsion springs.* On account of the smallness of the couples to be measured (the constants of the springs were of the order of 1200 c.g.s. while those used for the investigation of the ferro-magnetic substances were some tens of thousands) it was found more suitable to use a straight instead of a helical spring. We took a strip of phosphor bronze about 5.5 cm. long ( $l'$ ) and  $0.2 \times 0.01$  sq. cm. in cross-section. The upper end was soldered to a spiral spring of three turns made from a much thicker strip than the other; the greatest dimension of this strip was horizontal so that in this way it fulfilled its purpose of being elastic to tension while taking no part in torsion; its presence is essential to prevent the breaking of the thin glass stem or of the platinum-iridium stretching wire that is soldered to the stem. This stretching wire is made from a platinum-iridium wire of 0.1 mm. diameter, which was rolled very thin so as to make its torsion constant extremely small without diminishing to any great degree its resistance to breakage. The stretching wire is fused at  $b'_4$  to the lower end of the glass stem, and at its other extremity it carries a knob  $c'$  which is held fast in a ring  $f''_4$ .

The mounting of the apparatus took place with the same precautions regarding the centring of the whole, the tension of the springs, etc. and by a method similar to that which has been described in the research upon the ferro-magnetic metals.

*The course of the observations* is very simple once everything has been set up in position. First, those azimuths of the electromagnet are tentatively determined for which the couple in both directions is a maximum. It was sufficient to do these experiments two or three times with suitably chosen fields, since the azimuth changes but very little with the field, and for other values of the field one can without danger have recourse to interpolation. After that the series of observations took place in the following manner: Before making a measurement with any particular current this was reversed a certain number of times so as to obtain a well-defined field; we had not here to deal with a value of the saturation-magnetization, which changes but slowly with the field, but in our case the couple was proportional to the square of the field, so that inaccurate values of the field that might be obtained notwithstanding the fact that the iron of the electromagnet was extremely soft would make their influence very strongly felt in our results. Then the electromagnet was adjusted to one of the determined azimuths, the torsion angle was read off for the two directions of the field, the current broken, the electromagnet turned to the opposite azimuth, and so on several times. At the end of a series a measurement with one of the first fields was repeated as a control.

*Sources of error, difficulties, corrections, and controls.*

1. *Inhomogeneity of the magnetic field.* As will be seen from the following discussion this source of error is by far the most important in our case and is indeed the only one that need be taken into account. If we assume that the field near the centre of the pole-gap may be represented by an expression of the form

$$H = H_0 + \frac{r^2}{2} \left( \frac{\partial^2 H}{\partial x^2} \right)_0 (\cos^2 \theta - \frac{1}{2} \sin^2 \theta). \quad . \quad . \quad . \quad (3)$$

where  $H_0$  is the field in the centre, and  $r$  and  $\theta$  polar coordinates of a point in the pole-gap with respect to the centre as origin<sup>1)</sup>. Let us now replace the ellipsoid by a vertical disc whose diameter is equal to the major axis of the ellipsoid; by taking the expression for the energy of the magnetized disc in the field and differentiating it with respect to the angle between the disc and the lines of force, we obtain for the couple caused by the inhomogeneity of the field:

<sup>1)</sup> Cf. P. WEISS and H. KAMERLINGH ONNES l.c.

$$M' = \frac{3}{16} v r^2 I \left( \frac{\partial^2 H}{\partial x^2} \right)_0 \sin \varphi \cos \varphi. \quad (4)$$

( $r$  = radius of the disc).

The ratio  $\frac{M'}{M}$  of this couple for an angle of  $45^\circ$  to the fundamental couple is

$$\frac{M'}{M} = \frac{3}{16} \cdot \frac{\left( \frac{\partial^2 H}{\partial y^2} \right)_0}{(N_2 - N_1) I} \cdot r^2.$$

If then we suppose that the relative change of the field in the space occupied by the ellipsoid is of the order of 1 in 1000, the formula given above shews us that although the disturbing couple is a little smaller than the chief couple, the two are *of the same order of magnitude*. Hence we see the great influence that this source of error can have in the investigation of weakly magnetic substances. (With ferro-magnetic bodies it is quite negligible: see the previous paper).

We have accordingly devoted the greatest attention to this source of error. The *conical* pole-pieces were made slightly concave, during which process we every time determined the inhomogeneity of the field by means of a ballistic galvanometer and a small coil that was slightly displaced. We ascertained that the change in the field in a space of about 1 c.c. was certainly less than 1 in 2000. We have not had time to pursue this investigation further, and, besides, we should have to obtain a much more sensitive ballistic galvanometer. But it will be seen that the homogeneity of the field was sufficient for the comparative measurements we proposed to make. We may further remark that all these precautions refer exclusively to the conical pole-pieces; the experiments with the cylindrical pole-pieces were nearly free from these sources of error.

We allow for these disturbing couples in the following way: Assuming that  $\left( \frac{\partial^2 H}{\partial x^2} \right)_0 = \lambda H$  the expression for the couple due to inhomogeneity given above becomes ( $\varphi = 45^\circ$ ):

$$\frac{3}{32} v r^2 \lambda I H$$

or

$$\frac{3}{32} v r^2 \lambda K H^2,$$

which we shall represent by

$$\beta K H^2.$$

If  $\alpha$  is the angle of torsion of the holder and  $C$  the constant of the spring, then

$$Ca = \frac{v}{2} (N_1 - N_2) K^2 H^2 + \beta K H^2. \quad . \quad . \quad . \quad . \quad . \quad (5)$$

Thus just as if there were no correction for inhomogeneity the second side of the equation remains always proportional to the square of the field. Even without knowing that correction, if  $\beta$  is itself a constant we should be able to deduce from the observations whether  $K$  is a function of the field or not. We see, however, that the constancy of  $\beta$  requires that of  $\lambda$ , i.e. that the field must remain homothetic no matter how great it should be. Now this is not the case as can be seen from the quotients  $\frac{2\sigma}{H^2}$  in tables V, VII, and VIII. Table V shows first an increase, then the quotient reaches a maximum and diminishes considerably; tables VII and VIII shew a change in exactly the opposite direction; this is just what one would expect if  $\beta$  were variable and  $K$  constant, for the tables refer to two practically identical bodies, of which the one is dia- and the other para-magnetic. Now in either case the fundamental couple (uniform field) is in the same direction while the couple due to inhomogeneity changes sign with the susceptibility; should, therefore, the correction in the one case first increase and then decrease, it must in the other case first decrease and then increase. We shall return to this point in § 4.

Since this determination aims only at relative measurements, we have once and for all taken as the value of the susceptibility of oxygen at  $-183^\circ\text{C}$ . the value that was given by the improved apparatus for measuring the magnetic rise. With the help of this value we have calculated the values of  $\beta$  for each field from equation 5): (see tables V and VI). These values fall pretty well on a curve of means. Finally the susceptibility at the lower temperatures is calculated by means of the value of  $\beta$  as a function of the field given by this curve. We shall take the opportunity of the corresponding series of observations to make some remarks upon the influence of the inhomogeneity for each of the three pole-gaps that were used.

2. *The inconstancy of the magnetization as a function of the azimuth.* The general expression for the couple in a uniform field

$$(N_1 - N_2) I^2 v \sin \varphi \cos \varphi$$

only reaches its maximum value just at  $\varphi = 45^\circ$ , and consequently  $\sin \varphi \cos \varphi = 1/2$  since  $I$  remains constant during the torsion. Here again we see a fundamental difference between the application of this method to the investigation of saturation magnetization and to that of a body of constant susceptibility. It is clear that in the first case the condition  $I = \text{constant}$  is, as it were, fulfilled by definition.

In our case the deviation from this is by no means *a priori* negligible: the two limiting values of  $I^2$  ( $\varphi = 0^\circ$  and  $\varphi = 90^\circ$ ) differ in our case by 0.3 %, and since  $I^2$  always changes between these two limits in the same direction the error caused thereby when  $\sin \varphi \cos \varphi = 1/2$  is less than 0.1 %.

In contrast with the two foregoing sources of error, the reaction of the magnetized ellipsoid upon the *distribution of magnetism over the surface of the pole-pieces* can clearly have no effect in the case of a body of small susceptibility while on the other hand, it had to be taken into account in the case of the ferromagnetic bodies. Indeed, with oxygen we have to deal with a magnetization that in the strongest fields of the electromagnet reaches a value of only a few units (in the case of iron it was 1700!).

3. *Influence of the holder.* In this connection we may notice two actions that may go together. In the first place there is the inherent magnetism of the stem, and then there is also an action analogous to that which we wish to measure, for if the stem is not a perfect body of revolution, it is acted upon in the liquid oxygen just as if it were a *supplementary ellipsoid*. We investigated these two sources of error in a blank experiment in liquid oxygen in which the silver ellipsoid was removed, and the surface of the glass was carefully freed from all traces of wax. From this we obtained a maximum of only 1 to 2 % which need not be taken into account.

4. *The concentration of the oxygen.* The oxygen in the bath contained a little nitrogen, the concentration of which constantly decreased during the experiment owing to its faster vaporization. So as to be able to allow for this we analysed the gas at the beginning and at the end of each series of observations. The mean concentration was 1.25 % at the beginning and 0.35 % at the end (at the moment that the DEWAR vessel was almost empty). We allowed for this concentration as far as possible; in this respect there remains an uncertainty of about 0.3 %.

5. *Calibration of the suspension springs.* The main torsion spring described above was calibrated outside the apparatus by observing the time of oscillation of a system suspended from it with and without the addition of a known moment of inertia. For the latter we used a bronze ring of rectangular meridian cross section, the diameters and height of which were measured with the cathetometer. Calculation gave the moment of inertia as

$$582.09 \text{ c.g.s.}$$

Care was taken that the spring was subjected to the same tension during the calibration as it experienced while in the apparatus (by attaching suitable weights to it by a torsion-less wire).

For the constant of the spring we found 1184.5 c.g.s.

The platinum-iridium stretching wire gives a torsion couple as well as the spring; the correction for this was determined by the same method as was used in the analogous case by WEISS and KAMERLINGH ONNES (loc. cit.) and it was found to be 0.0152 times the constant of the spring. The difference between the values of the constant at 18° C. and at -190° C. is smaller than the errors of observation. The calculations were therefore carried out with the constant  $1184.5 (1 + 0.0152) = 1202.5$ .

6. *Oscillations.* The silver ellipsoid should be protected sufficiently from the influence of oscillations arising from external causes by the occurrence of intensive FOUCAULT currents, but the occurrence of these currents, which were unusually strong gave rise to great difficulties in the observations. In the first place the holder was extremely slow to reach its position of equilibrium. Further, the smallest change in the current flowing through the electromagnet occasioned a sudden kick in the whole moveable apparatus, an immediate result of the oblique position of the ellipsoid with respect to the lines of force. Hence the regulation of the current had to be done with the greatest care. We retained the oil-damper but removed the fixed partitions, for the capillary action of these gave rise to couples that, although small, were still not negligible.

*Results of the observations.*

TABLE IVa. Cylindrical pole-pieces 21 mm. apart. $t = -183^{\circ}.0$ C.			
$H$ gauss	double deflection $2\delta$ cm. of the scale	$\frac{2\delta}{H^2} \cdot 10^7$	$K \cdot 10^6$
2250	0.37	0.731	277
4537	1.41	0.685	268.8
6676	3.21	0.7200	275.3
8339	5.10	0.7335	278.1
9387	6.44	0.7307	277.5
10120	7.45	0.7274	277.0
10685	8.26	0.7234	276.1
11130	8.99	0.7258	271.6
11440	9.38	0.7167	274.8
11765	9.90	0.7155	274.6

Mean  $K_{90^{\circ}.1K.} = 275.6$

TABLE IVb. Cylindrical pole-pieces 21 mm. apart. $t = -201^{\circ}.1$ C.			
$H$ gauss	double de- flection $2\delta$ cm. of the scale	$\frac{2\delta}{H^2} \cdot 10^7$	$K \cdot 10^6$
2250	0.50	0.988	324
4537	2.10	1.020	328.0
6676	4.66	1.041	331.1
8339	7.36	1.058	334.0
9387	9.17	1.042	331.4
10120	10.63	1.038	330.8
10685	11.81	1.034	330.2
11130	12.70	1.025	328.6
11440	13.41	1.024	328.4
11765	14.14	1.022	328.0

$$\text{Mean } K_{72^{\circ}.0\text{K.}} = 330.0.$$

Without it being corrected for lack of uniformity in the field the means give the following values:

$$\chi_{90^{\circ}.1\text{K.}} = \frac{275.6}{1.143} \cdot 10^{-6} = 241.1 \cdot 10^{-6}$$

$$\chi_{72^{\circ}.0\text{K.}} = \frac{330.0}{1.230} \cdot 10^{-6} = 268.3 \cdot 10^{-6}.$$

The corresponding results obtained by the method of the magnetic rise were

$$240.6 \cdot 10^{-6} \text{ and } 269.3 \cdot 10^{-6}.$$

The differences between the results as obtained by the two methods are scarcely 0.4 %. This gives us great confidence in the ellipsoid method even for this particularly difficult determination, and it shews that the method is also suitable for absolute measurements if only the necessary care is taken to ensure the uniformity of the field and the correctness of the shape of the ellipsoid.

We must remember that there was a great number of absolute

measurements whose results had to be used (axes and volume of the ellipsoid, constants of the springs, magnetic field, density of the liquid oxygen) and also that the shape of the ellipsoid was not perfect. On the other hand we must remark that the application of the correction for the non-uniformity of the field might conceivably have diminished the correspondence between the results obtained by the two methods. We have, however, both theoretical and experimental grounds for the assumption that this correction remains within the limits of accuracy of not more than 0.5% in the case of cylindrical pole-pieces with flat end surfaces 90 mm. in diameter and at a distance of 21 mm. apart.

T A B L E Va. Conical pole-pieces 20 mm. apart. $t = -183^{\circ}.0$ C. (To determine $\beta$ we assumed $\chi_{90^{\circ}.1} = 240.6 \cdot 10^{-6}$ ).			
$H$ gauss.	Double de- flection $2\delta$ cm. of the scale.	$\frac{2\delta}{H^2} \cdot 10^7$	$10^6 \beta$
3685	1.27	0.935	58.5
4615	1.96	0.920	54.7
6944	4.55	0.9437	60.8
9205	7.96	0.9400	59.7
11280	11.90	0.9348	58.5
12835	15.44	0.9374	59.1
14015	18.26	0.9295	57.0
14900	20.19	0.9098	51.9
15585	21.73	0.8945	48.1
16120	22.87	0.8802	44.5

A graph of  $\beta$  as function of  $H$  was made, which was used for the following table.

T A B L E Vb. Conical pole-pieces 20 mm. apart. $t = -208^{\circ}.2$ C.				
$H$ gauss.	Double de- flection $2\delta$ cm. of the scale.	$\frac{2\delta}{H^2} \cdot 10^7$	$\beta \cdot 10^6$	$K \cdot 10^6$
2296	0.79	1.498	56.0	[357]
4615	2.75	1.291	57.8	[328]
6944	6.82	1.414	58.5	344.2
9205	12.15	1.435	59.8	346.3
11280	18.25	1.434	59.7	346.2
12835	23.67	1.437	59.2	346.9
14015	27.89	1.420	56.5	346.1
14900	30.84	1.389	52.4	345.1
15585	33.24	1.368	48.0	345.0
16120	35.44	1.363	43.8	347.3

The mean with the exception of the two values placed between brackets is 345.9 and it gives

$$\chi_{64^{\circ}.9 \text{ K.}} = 275.0 \cdot 10^{-6}$$

while the method of the magnetic rise gave

$$\chi_{64^{\circ}.9 \text{ K.}} = 283.5 \cdot 10^{-6}$$

The difference is 3%; but in this connection we must remember that the correction for non-uniformity is about 16%, and that the temperature of the liquid becomes very uncertain at the pressure of 11 mm. under which the liquid boils at this temperature.

Finally, we now give two series of measurements which were made with other pole-gaps so as to obtain other deviations in the uniformity of the field. They were hastily made and under unfavourable circumstances, since oscillations and disturbances caused by the running of machines in the neighbourhood interfered with the observations. We give them more as examples of how the method of calculation followed still leads to good results even when the couples due to non-uniformity of the field are extremely large (28% of the chief couple).

T A B L E VIa. Conical pole-pieces 18.2 mm. apart. $t = -183^{\circ}.0$ C. (To determine $\beta$ we assumed $\chi_{90^{\circ}.1} = 240.6 \cdot 10^{-6}$ ).			
$H$ gauss	double de- flection $2\delta$ cm. of the scale	$\frac{2\delta}{H^2} \cdot 10^7$	$\beta \cdot 10^6$
5013	3.34	1 328	159.2
7547	7 31	1.283	147.4
9993	12 64	1.246	137.9
12165	18.33	1 238	136 2
13760	22.39	1.183	121.7
14900	26.26	1 182	121.5
15750	28.83	1.162	116.6
17005	35.58	1.230	133.9

$\beta$  was again graphed as a function of  $H$ , which led to the correction for  $K$  in the following table.

T A B L E VIb. Conical pole-pieces 18.2 mm. apart $t = -208^{\circ}.2$ C.				
$H$ gauss	double de- flection $2\delta$ cm. of the scale	$\frac{2\delta}{H^2} \cdot 10^7$	$\beta \cdot 10^6$	$K \cdot 10^6$
5013	46.8	1.861	152	341
7547	10.71	1.880	146.5	348
9993	18.88	1.862	137.5	351
12165	27.92	1.885	130.7	357
13760	35.35	1.868	125.5	359
14900	41.36	1.861	123.0	360
15750	45.67	1.840	122.5	357
17005	51.81	1.791	127.5	347

The mean 353 gives  $\chi = \frac{K}{1.255} = 279.5 \cdot 10^{-6}$  a value that is not much smaller than  $283.5 \cdot 10^{-6}$ , which was obtained by the method of the magnetic rise.

*Solid oxygen.*

§ 4. *Ellipsoid of solid oxygen.* In this case observations had to be made directly upon an ellipsoid of oxygen. The oxygen therefore had to be frozen in a mould of approximately the same form and dimensions as the solid silver ellipsoid described above. This new condition necessitated the following experimental arrangement.

The cover and the DEWAR tube are the same as for liquid oxygen, with the exception of the cap  $D$ . The adjusting tube is also the same, but it is so arranged that it can be moved as a whole up or down, while the whole apparatus remains closed and in its place. With this end in view it is attached to the tube  $m$ , which moves through the stuffing-box  $D''_2$ ; this corresponds to  $D_2$  of the liquid oxygen apparatus, but in this case the wide glass tube  $C_1$  is lengthened by a rigid brass tube  $M$  that serves to give sufficient play to the vertical movement of the whole adjusting tube. The former stem  $k$  had to be lengthened by the same amount ( $L''_1, L''_2$ ), and is contained in the tube  $m$ .

The holder is also a glass tube  $b''$ ; it is not however closed, but at  $b''_2$  it changes into a very much narrower tube (0.5 mm.) that ends at  $b''_1$  in a glass ellipsoid  $a''$ . To this ellipsoid there is fused a solid stem  $b''$ , that connects it with the stretching wire. The oxygen gets to the ellipsoid through the holding tube which it enters at  $b''_2$ . A rubber tube  $n$  ( $d = 3$  mm.) admits the gas from outside; it is attached to the inlet tube  $n_1$  that passes through the cover and is soldered to it. With this arrangement it is easy to cause the oxygen to solidify inside the ellipsoid. When the apparatus is ready for use the adjusting tube is pulled upwards by the cap  $\Delta$  till the glass ellipsoid reaches the unsilvered part of the vacuum glass. The vacuum glass is then filled with liquid hydrogen. While the ellipsoid is still connected with a reservoir of oxygen, the adjusting tube with the ellipsoid is slowly pushed downwards until it does not quite touch the liquid hydrogen but is in its vapour. The oxygen is then seen to condense slowly, and, if the operation is carefully performed, the whole ellipsoid and supply tube are seen to fill with liquid oxygen. The tube being lowered still further, vapour is reached that is sufficiently cold to cause the oxygen to solidify. On account of the large

contraction of the oxygen on solidification it is seldom that one does not see some empty space in the ellipsoid; the operation must then be repeated several times, since the oxygen that is still liquid at this temperature has a pretty great viscosity and flows with difficulty from the tube; we shall return to this point later. When the ellipsoid is completely filled with solid oxygen the adjusting tube may be lowered right down. A mark is made beforehand, so that the ellipsoid may be accurately adjusted to the centre of the gap when the silvered tube is again in its place.

*Errors, corrections, auxiliary measurements.*

1. *Couples due to inhomogeneity.* As will presently appear, we made measurements not only in liquid hydrogen (solid oxygen), but also keeping everything else the same, at two temperatures in a bath of liquid oxygen (i.e. with the same ellipsoid of liquid oxygen). Since the susceptibility of the liquid oxygen was known, we had therefore two measurements of the couples due to inhomogeneity as a function of the field; they are given in Table VII. As a result of the somewhat smaller dimensions of the ellipsoid, these corrections are comparatively much less important.

2. *Purity of the oxygen.* The oxygen was freed from nitrogen by vaporizing a large quantity of impure liquid oxygen under reduced pressure.

3. *Density of the solid oxygen.* We have already mentioned the difficulty of completely filling the ellipsoid with solid oxygen. On account of the opaqueness of the oxygen that has already solidified one cannot with certainty assert that this condition has been fulfilled<sup>1)</sup>.

Since the specific susceptibility is determined from a known volume this error would have immediate effect upon the result. We tried to eliminate this error as well as possible by determining the density with the same ellipsoid by filling it with solid oxygen under the same circumstances as those obtaining in the experiments and then measuring the quantity of gas formed from it on vaporization. We may assume that the small cavities that may form are pretty much the same in the various cases. Indeed, from two similar measurements the density measured in this way was found to vary by only about 1%. By taking as the mean density that determined by these experiments, the eventual presence of cavities is allowed for. In this way we obtained

$$\rho = 1.41.$$

The *absolute* values of the couples due to inhomogeneity of the

<sup>1)</sup> When there is an empty space of a few mm<sup>3</sup>, however, it can be seen quite well.

field are not modified by a cavity formed in the vertical axis, as was usually the case, for it is clearly those portions towards the surface of the ellipsoid that are the chief contributors to them. On the other hand, they might obtain a greater *relative* influence, but as the observations shew, the sum of the corrections arising from this cause is so small that they may be regarded as independent of the susceptibility within the limits of accuracy of the experiments. In that case this difficulty completely disappears.

4. *Dimensions of the ellipsoid.* The internal volume was obtained by filling the ellipsoid with mercury and weighing it. It was 0.1812 c.c. The change of volume under atmospheric pressure was found to be of no account by pumping the space above the mercury free from air and observing the position of the mercury in the capillary.

The external axes were measured directly. Then the thickness of the glass at ten different points was determined by focussing a microscope on the image of the outer surface formed on the mercury with which the ellipsoid was filled. It changed but slightly from place to place. The mean was taken and twice that value was subtracted from the external measurements. The results were:

1.044 cm.

and

0.335 cm.

Calculating the volume from these figures we get 0.1925 c.c. which is about 6% greater than the true volume as directly determined. This is accounted for by the special shape of the meridian section which curves somewhat too strongly at the outer ends. For calculating the coefficients of demagnetization we took a mean ellipsoid with the same major axis and the minor axis small enough to give the real volume<sup>1)</sup>. The data for the calculation were therefore:

1.044 cm.

and

0.3173 cm.

5. *Opposing couple.* The suspension spring and the stretching wire were the same as were used for the liquid oxygen. We must, however, allow for the rubber supply tube for the oxygen. This (which was chosen as thin as possible) modified both the zero and the constants of the total opposing couple, as soon as the pressure

<sup>1)</sup> It is clearly not quite right to do this; there are, however, experimental data to support this method of correcting: V. QUITTNER (Diss. Zürich 1908, also Arch. sc. phys. et nat. Genève, Sept.—Nov. 1908) found that this method of treatment was sufficiently accurate even for discs, bodies that deviate far more from an ellipsoid than those we used.

difference between the inside and the outside of the tube appreciably altered (on account of the change in shape of the tube). In all our experiments, therefore, we took care that there was a constant pressure difference of 70 mm. between the pressure inside the cover and that inside the holder (the latter was the smaller of the two). We got a very sensitive indication of the constancy of this difference not only from the manometers but also from the zero position of the holder. Experiments carried out outside the apparatus shewed that the constant of the total couple changed about 10% between the complete flattening of the rubber tube by the atmospheric pressure and equality between the pressures on both sides. This corresponds to a deflection on the reading scale of *more than a metre*. If we assume rough proportionality we find that a displacement of 1 cm. would indicate a change in the opposing couple of only 0.1 %. The zero was kept constant to a few millimetres.

The calibration was made under circumstances exactly the same as in the experiments (pressure difference, etc.).

The total constant with the addition of that of the stretching wire was

$$1503 + 18 = 1521 \text{ cgs.}$$

#### Results.

TABLE VII. Calculation of the corrections for non-uniformity from observations made in a bath of liquid nitrogen. Conical pole-pieces 20 mm apart.							
H gauss	t = - 195° 6 C.			t = - 210° 0 C.			$\beta \cdot 10^4$ (mean). Double weight given 195° 6
	2d cm.	$\frac{2d}{H^2} 10^7$	$\beta \cdot 10^4$	2d cm.	$\frac{2d}{H^2} 10^7$	$\beta \cdot 10^4$	
4615	1.18	0.554	-0.137	1.59	0.746	-0.199	-0.458
6944	2.69	5577	127	3.53	0.7322	232	162
9205	4.73	5580	126	6.21	7330	231	160
11280	7.08	5560	132	9.23	7251	250	171
12835	9.17	5564	131	12 10	7341	228	163
14015	11.14	5670	100	14.49	7378	219	140
14900	12.90	5812	060	16.94	7613	162	094
15585	14.29	5884	039	—	—	—	—
16120	15.67	6031	003	20 21	7781	121	010

It can be seen that the values obtained for  $\beta$  are not the same at the two temperatures. Meanwhile it has to be applied here only as a correction for the susceptibility of solid oxygen which at the most is 3%. A difference of temperature of 1° C. in the bath under reduced pressure gives more than half the difference between the two values, whence we have given the determination under reduced pressure only half the weight accorded the measurement at ordinary pressure.

The uncertainty of the mean has less than 1% influence upon the value of the susceptibility of solid oxygen. The curve for  $\beta$  as well as its sign correspond with what were found for the silver ellipsoid.

TABLE VIII. <i>Susceptibility of solid oxygen.</i> $t = -252^{\circ}.8$ (bath of liquid hydrogen boiling under atmospheric pressure).				
$H$ gauss	$2s$ cm. of the scale	$\frac{2s}{H^2} \cdot 10^7$	$K \cdot 10^6$ uncorrected	$K \cdot 10^6$ corrected according to tab. VII.
2296	0.89	1.60	519	533
4615	3.57	1.676	518.3	532.9
6244	7.92	1.642	512.3	527.3
9205	14.07	1.660	515.0	530.1
11280	21.14	1.661	515.2	530.1
12835	27.92	1.684	518.7	532.8
14015	32.96	1.678	517.6	529.0
14900	37.38	1.683	518.4	527.5
15585	40.77	1.678	517.6	523.7
16120	44.05	1.696	520.0	523.6

Mean 529.0

whence it follows that  $\chi_{20^{\circ}.3 \text{ K.}} = \frac{529.0}{1.41} 10^{-6} = 375.2 \cdot 10^{-6}$ .

T A B L E IX. <i>Susceptibility of solid oxygen.</i> $t = -258.9^{\circ}$ (bath of liquid hydrogen under 70 mm. vapour pressure).				
$H$ gauss	$2d$ cm. of the scale	$\frac{2d}{H^2} \cdot 10^7$	$K \cdot 10^6$ uncorrected	$K \cdot 10^6$ corrected
2296	1.49	2.257	600.5	614.5
4615	4.80	2.253	600.1	614.6
6944	10.86	2.252	600.0	615.0
9205	19.24	2.270	602.2	617.1
11280	28.13	2.210	594.3	609.2
12835	37.09	2.250	599.6	613.7
14015	45.05	2.293	604.7	616.7
14900	51.24	2.306	606.8	615.9
15585	55.73	2.293	605.3	614.1
16120	60.20	2.317	608.5	612.1

From the mean 614.0 follows  $\chi_{sol. 14^{\circ} 2K.} = \frac{614.0}{1.41} = 435.6 \cdot 10^{-6}$ . The products into  $\sqrt{T}$  are

$$- 252^{\circ} 8 \quad 375.2 \cdot 10^{-6} \sqrt{20.3} = 1690 \cdot 10^{-6}$$

$$- 258.9^{\circ} \quad 435.6 \cdot 10^{-6} \sqrt{14.2} = 1641 \cdot 10^{-6}.$$

Hence we can represent the two observations pretty well by

$$\chi_{sol.} = \frac{1690}{\sqrt{T}} \cdot 10^{-6},$$

which is adjusted to the measurement at the higher temperature.

The deviation from this ratio for the lower temperature, however, is somewhat greater than the errors of observation.

§ 5. *Summary and conclusion.* As regards the dependence of specific susceptibility upon temperature our most reliable determination gives

$$\chi_{liq. 90^{\circ} 1 K.} = 240.6 \cdot 10^{-6}.$$

CURIE found  $\chi = \frac{33700}{T} \cdot 10^{-6}$  between  $20^{\circ} C.$  and  $450^{\circ} C.$  whence it

would follow that for  $T = 90^{\circ}.1 \text{ K.}$   $\chi = 374.10^{-6}$  a number that differs essentially from ours<sup>1)</sup>).

There is therefore no possibility of extrapolating CURIE's law to the liquid phase of oxygen. This was also the conclusion reached by FLEMING and DEWAR in their first treatment of the question, but after more careful experiments they rejected their former result<sup>2)</sup>.

The results obtained from the two magnetic rise apparatus at lower temperatures can, within the limits of experimental error, be expressed by a very simple law: *the specific susceptibility is inversely proportional to the square root of the absolute temperature.* From the observations obtained with the more reliable apparatus we deduce the formula

$$\chi_{liq.T} = \frac{2284}{\sqrt{T}} \cdot 10^{-6}$$

which holds to within 5%. None of the results obtained by the method of the maximum couple are in conflict with those deduced from the formula.

The results with solid oxygen approximately follow the relation

$$\chi_{sol.T} = \frac{1690}{\sqrt{T}} \cdot 10^{-6}.$$

At the lowest temperatures there is a small deviation indicating a smaller increase at lower temperatures; it is, however, so small that we may still accept the formula given as approximately correct for the solid state of aggregation below the melting point of oxygen and down to  $14^{\circ}.2 \text{ K.}$

Further experiments at more numerous temperatures must show exactly how far these deductions hold for the liquid and solid states. They shew (see fig. 5) that there is a jump in the value of  $\chi$  at the melting point, since

$$\chi_{liq.T_m} = 1,3 \chi_{sol.T_m}$$

<sup>1)</sup> R. HENNIG's (1893) result should give

$$\chi = \frac{27600}{T} 10^{-6} \text{ and } \chi_{90^{\circ}.1 \text{ K.}} = 307 \cdot 10^{-6}.$$

<sup>2)</sup> FLEMING and DEWAR's results: 1st paper (1896)  $\chi_{90^{\circ}.1 \text{ K.}} = 200 \cdot 10^{-6}$ ; 2nd paper (1898)  $281 \cdot 10^{-6}$ , mean  $243.5 \cdot 10^{-6}$  pretty much the same as our result. According to the mean of the result of FARADAY and BECQUEREL the specific susceptibility for oxygen at  $0^{\circ}$  is  $91 \cdot 10^{-6}$ ; this gives by extrapolation from CURIE's law  $\chi_{90^{\circ}.1 \text{ K.}} = 299 \cdot 10^{-6}$ . The English savants used this number in their second research for the comparison of the susceptibility of liquid oxygen with that of the gas.

We hope to answer the question if this jump really exists by special experiments arranged for the purpose; we may, in the meantime, consider that it does probably exist. What CURIE found in the transformation of  $\gamma$  iron to  $\delta$  iron is analogous to the sudden change which we here assume to exist while the form of the law remains unaltered, and which can occur at the melting point or at a point of transformation to an allotropic modification. WEISS<sup>1)</sup> has shown that this can be accounted for on the assumption that at this particular point di-atomic iron changes into tri-atomic.

On the other hand we consider it probable that the law according to which the specific susceptibility increases with the temperature, viz: inverse proportionality to the square root of the absolute temperature at lower temperatures, gradually transforms into that of inverse proportionality (CURIE's law) at higher temperatures, and that each of these laws, therefore, may be but approximative to the same function over different ranges of values of the independent variable  $T$ .

The supposition that the change of specific susceptibility with density is of no importance lies at the bottom of the assumption of the gradual transformation of CURIE's law into that of  $T^{-1}$ . If, on the other hand, we assume that this change is of importance, that e. g. when the internal pressure is considerable the molecules under its influence undergo not only a compression but also a lessening of their magnetic moments, then a region of great molecular compressibility in which the specific susceptibility should change both with the temperature and with the density should exist between the gaseous phase in which the specific susceptibility would be pretty well independent of the pressure, and the liquid phase at lower temperatures, in which the molecules would not be appreciably affected by an additional external pressure on account of their already great internal pressure, and in which, therefore, the specific susceptibility would also be pretty well independent of the pressure. As regards the difference between the magnetic moment of the elementary magnets in the condition of saturated liquid and vapour and that at normal or smaller density at the same temperatures, it is to be expected according to that representation, that this difference will change with temperature in consequence of the change of density with temperature.

The assumption can also be made that complex molecules are formed in the liquid state, and that these diminish the intensity of the elementary magnets; in that case changes in susceptibility of

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<sup>1)</sup> P. WEISS, loc. cit.

mixtures of liquid oxygen with non-magnetic gases should obey the thermodynamic laws that govern the number of such complexes. But all this must be established by further experiments which we hope to complete; in the meantime the most probable assumption is the old one that the specific susceptibility is independent of the pressure.

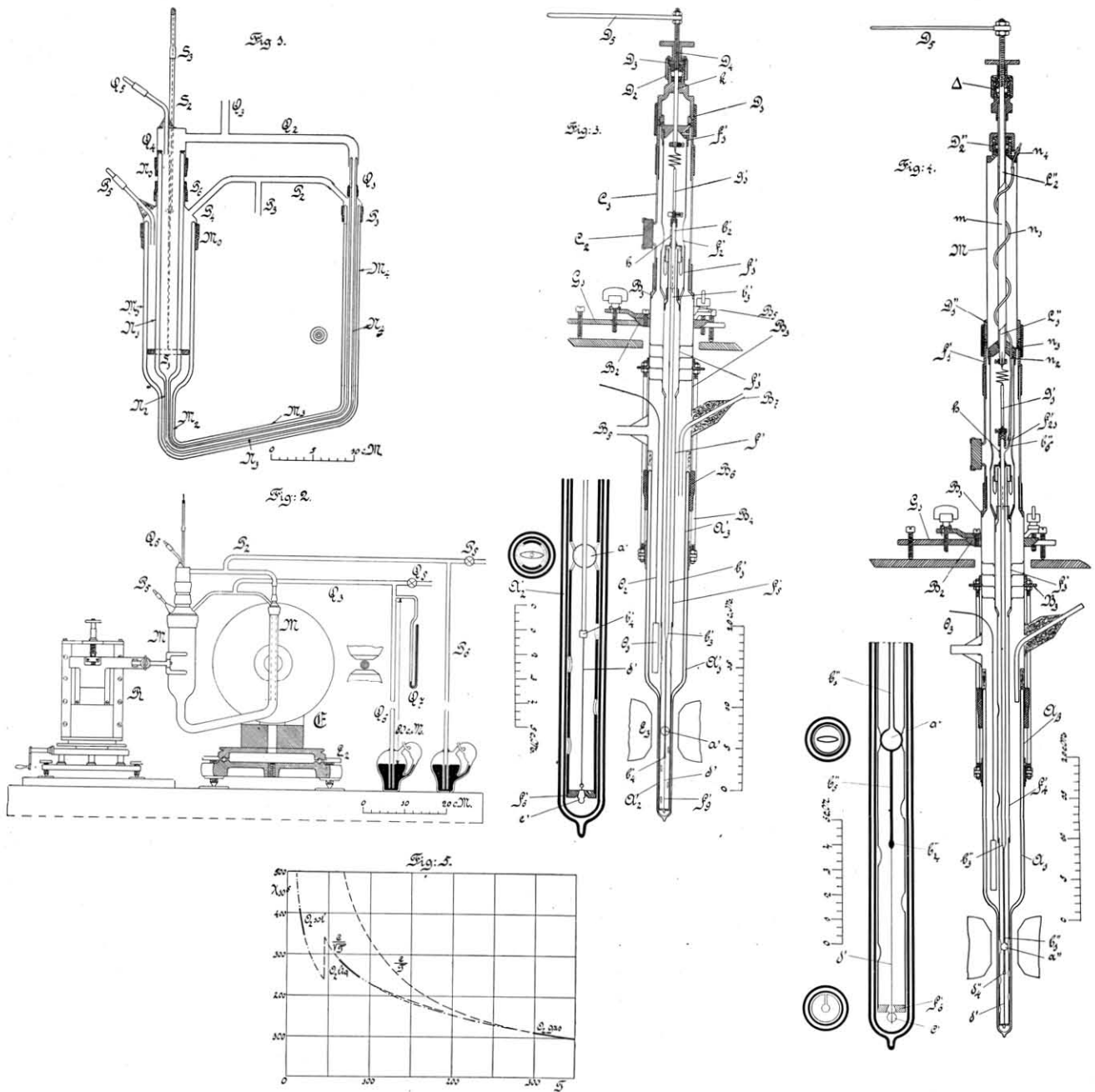
As regards the question as to whether the specific susceptibility at lower temperatures still follows the law of inverse proportionality to the root of the absolute temperature, if the ferro-magnetism with a very low-lying CURIE point according to WEISS's theory of corresponding magnetic states does not exist, then the change to a still slower increase with decreasing temperature and the approximation to a limiting value is, perhaps, more probable.

The law of  $T^{-\frac{1}{2}}$  at once gives rise to the question if instead of the LANGEVIN elementary magnets whose intensity is independent of the temperature, we should assume that their intensity varies directly as  $\sqrt{T}$ ; that is, that we should assume the existence of elementary currents or electrons moving in their paths with speeds proportional to (and, therefore, determined by) the speeds of molecular heat motions. In other words, while LANGEVIN's theory already supposes that the planes in which the electrons move follow the motions of the molecules, but that the areas described in those planes are still independent of heat motion, we should now assume that the electrons undergo the influence of heat motion at their motion in their paths, and, if the radius of their path has also become invariable, revolve while remaining in the same position with respect to the atom; they would be electrons that are frozen fast to the atom, an assumption that has already been made to explain other phenomena.

This addition to LANGEVIN's theory, however, does not lead to a specific susceptibility proportional to  $T^{-\frac{1}{2}}$  as one at first sight would be inclined to think, but to a constant specific susceptibility.

To substantiate that addition it will probably be necessary to proceed to still lower temperatures than those of our experiments. It seems at present that it is not impossible that then the law  $\chi$  proportional to  $T^{-\frac{1}{2}}$  changes to  $\chi = \text{const.}$ : our observations on solid oxygen seem to indicate a change in this direction. The assumption to which this is equivalent: viz, that the magnetic motions of the electrons cease at the absolute zero, and to which our experiments seem to lead, is much more satisfactory than that the magnetic motions of the electrons still persevere even at the absolute zero.

The second question to which we devoted attention — the dependence of susceptibility upon field strength requires no detailed treat-



ment. The method of the magnetic rise seemed in some instances to give a decrease of the order of 1% in a field of 8000 gauss, while the method of the maximum couple gave with the cylindrical pole-pieces up to 12000 gauss only a very small systematic deviation and with the conical pole-pieces (16000 gauss) the deviation was scarcely appreciable.

The solid oxygen ellipsoid with which a much lower temperature was reached seemed to give a small decrease at 16000 gauss; it is possible, however, that a greater deviation is obscured by the correction for the non-uniformity of the field. We consider, however, that, assuming that the experiments were accurate to within 1% the change of the susceptibility with the field up to 16000 gauss remains within the limits of experimental error. This is in agreement with the theory of LANGEVIN, if this, notwithstanding the deviation from CURIE's law, is still applied.

**Physics.** — "*The magneto-optic KERR-Effect in ferromagnetic compounds and alloys*". By STANISLAW LORIA. (Communication from the Bosscha-Laboratory).

It has been shewn by KAZ<sup>1)</sup>, RIGHI<sup>2)</sup>, KUNDT<sup>3)</sup>, SISSINGH<sup>4)</sup>, ZEEMAN<sup>5)</sup> and also by KERR<sup>6)</sup> himself that the phenomenon discovered by the last named in 1876 depends not only on the orientation of the reflecting surface with respect to the magnetic vectors, but also (in a somewhat complicated manner) on the angle of incidence and the position of the plane of polarization of the incident beam. In the simplest and by far the most important case of almost normal incidence of light polarized perpendicularly or parallel to the plane of incidence, the reflected light in general is elliptically polarized according to RIGHI<sup>7)</sup>; the rotation of the major axis of the ellipse depends on the magnetisation and the wave-length.

According to the measurements made by DU BOIS<sup>8)</sup> it is in every case proportional to the former; as regards the variation with the

<sup>1)</sup> P. C. KAZ, Diss., Amsterdam 1884.

<sup>2)</sup> A. RIGHI, Ann. de Chim. et Phys. (6) 4 p. 433, 1885.

<sup>3)</sup> A. KUNDT, Wied. Ann. 23 p. 228, 1884; 27 p. 199, 1886.

<sup>4)</sup> R. SISSINGH, Arch. Néerl. (1) 27 p. 173, 1894.

<sup>5)</sup> P. ZEEMAN, Leiden Comm. no. 15, 1895; no. 29, 1896. Arch. Néerl. 27 p. 252 1894.

<sup>6)</sup> J. KERR, Phil. Mag. (5) 3 p. 389, 1877. Phil. Mag. (5) 5 p. 161, 1878.

<sup>7)</sup> A. RIGHI, Ann. de Chim. et Phys. (1) 9 p. 120, 1886.

<sup>8)</sup> H. DU BOIS, Wied. Ann. 39 p. 25, 1890.