Physics. — "*Magnetic Researches.* XXII. *On the determination of the magnetisation at very low temperatures and on the susceptibility of gadolinium sulphate in the region of temperatures obtainable with liquid hydrogen*". By H. R. WOLTJER. (Communication N°. 167*b* from the Physical Laboratory at Leiden). (Communicated by Prof. H. KAMERLINGH ONNES).

(Communicated at the meeting of September 29, 1923).

§ 1. *Introduction.* The significance of extending the investigation on the magnetisation of paramagnetic substances to the temperatures obtainable with liquid helium, that might be expected a priori, has been confirmed in a convincing way by the preliminary research on the magnetisation of gadolinium sulphate in liquid helium carried out by KAMERLINGH ONNES in 1914 [1]). The results then obtained showed the interest of continuing the research on gadolinium sulphate and completing the preliminary qualitative results by more accurate quantitative ones. Other substances, such as the paramagnetic chlorides [2]) presented themselves also for investigation in helium. However, closer inspection of the work of 1914 showed, that it was of little use repeating the work without detailed investigation

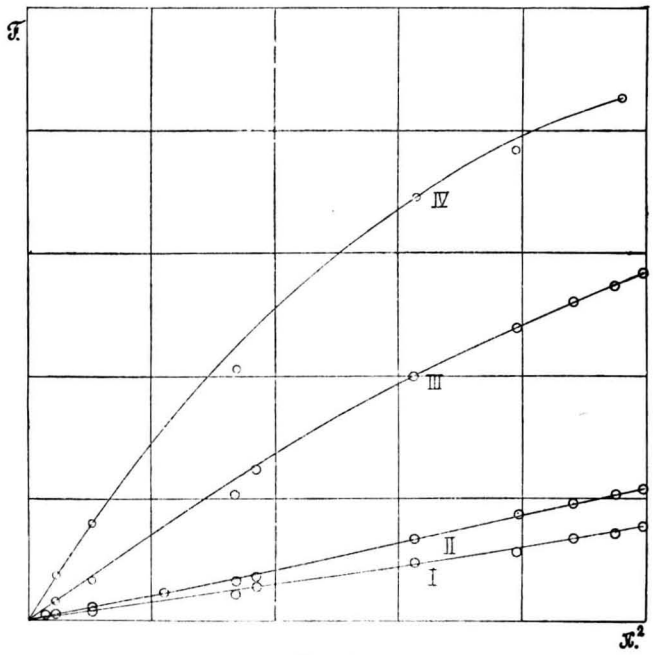

Fig. 1.

[1]) H. KAMERLINGH ONNES, these Proceedings, 17 p. 283; Leiden Comm. N°. 140d. Cf. also idem, Rapport Solvay 1921, p. 131. Leiden Comm. Suppl. N°. 44a.

[2]) l. c. p. 154, resp. p. 25.

of the method, more accurate calibrations and study of the corrections. E. g. concerning the direct results of the observations the considerable deviations [1]) from the proportionality between the force (F) and the square of the magnetic force between the poles of the electromagnet occurring at hydrogen temperatures (cf. fig. 1, taken from the paper mentioned) are particularly striking and, if the results are given in terms of LANGEVIN's theory of paramagnetic gases (cf. fig. 2, taken from Leiden Comm. Suppl. N°. 44a) it may be asked whether no systematic errors occurred.

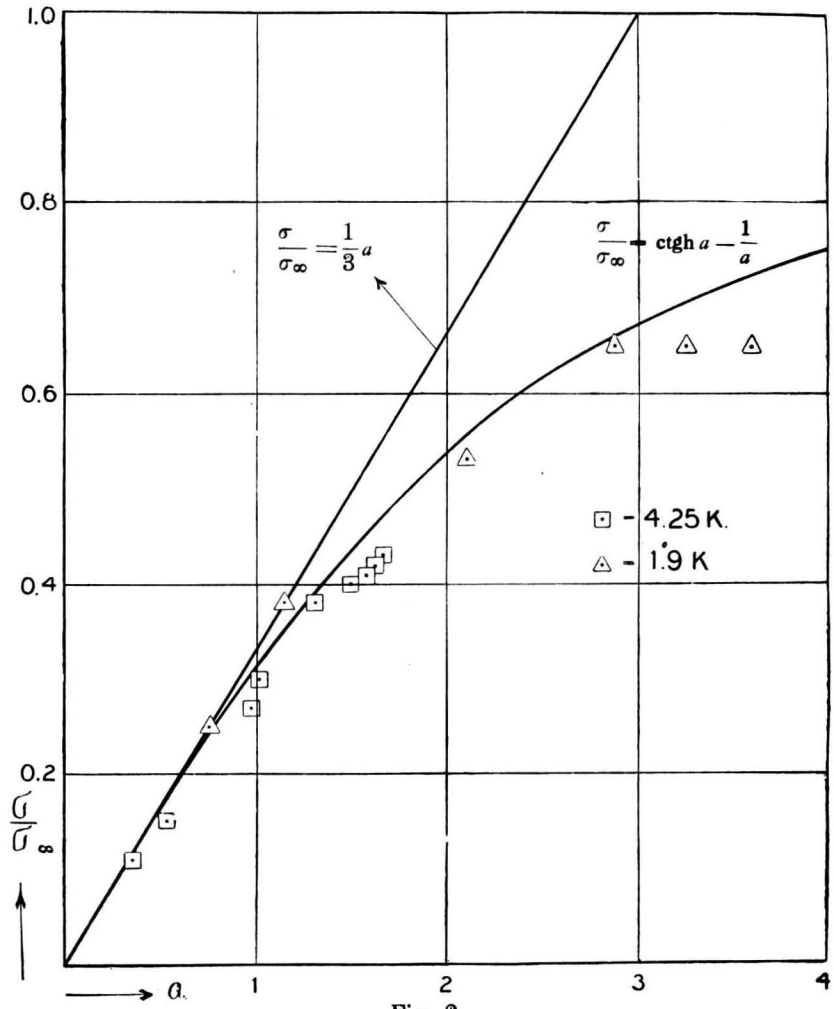

Fig. 2.

The method for measuring the magnetisation, it sources of errors

[1]) In the paper these deviations have been mentioned as probably due to inaccuracy in the topography.

and its corrections form the subject of the following paragraphs; we will consider more especially the topographic calibration of the electromagnet. It was carried out partially by means of the investigation of gadolinium sulphate in liquid hydrogen and so it furnished new material for the knowledge of the susceptibility of this substance, confirming old results. This new material will be communicated at the same time.

§ 2. *Apparatus and method.* The magnetisation was calculated from the force exerted by an inhomogeneous magnetic field on a small quantity of the material. For the measurement of the force the same apparatus was used as in the investigation of gadolinium sulphate in 1914, except a small alteration in connecting the tube containing the substance under consideration. At that time no description was given, so now some details may be mentioned. The apparatus was constructed by Mr. G. J. FLIM, chief of the Technical Department of the Cryogenic Laboratory, mainly on the same principles as the apparatus of KAMERLINGH ONNES and PERRIER [1]) for the investigation of paramagnetic substances. The substance to be investigated is placed at the bottom part of a long rod, the "carrier". This carrier is suspended to one or two floats swimming on mercury. The force exerted by the magnetic field on the substance is compensated by a known force and the compensation is checked by means of a telescope and a scale attached to the carrier (*Sc.* fig. 3). Some modifications were required with a view to the special circumstances. The apparatus is introduced at the top of the helium cryostat (*C*) and is supported by the rim *R*. It is counterbalanced by weights acting on the connecting tube between cryostat and liquefactor. The weight of the apparatus has been minimised. Partially for this purpose the ringshaped trough of the apparatus of KAMERLINGH ONNES and PERRIER has been replaced by a small glass reservoir (*G*) with only one float (*Dr*). The comparatively large forces occurring in the experiments (up to about 200 gr.) induced to prefer magnetic compensation instead of electrodynamic compensation by two coils, though the accuracy was diminished thereby. The compensating force comes from the attraction exerted by a current of suitable intensity passing through a coil *D* at the top of the apparatus on a weak iron rod *S* at the top of the carrier; by putting rings (*Ri*) under the coil *D* its height can be taken such as to exert upward or downward forces, as appears convenient. The distance of the weak iron rod to the interferrum of the electro-

[1]) These Proceedings 16, p. 689 and 786. Leiden Comm. N⁰. 139a.

magnet has been chosen such that the action of the latter on the former may be neglected.

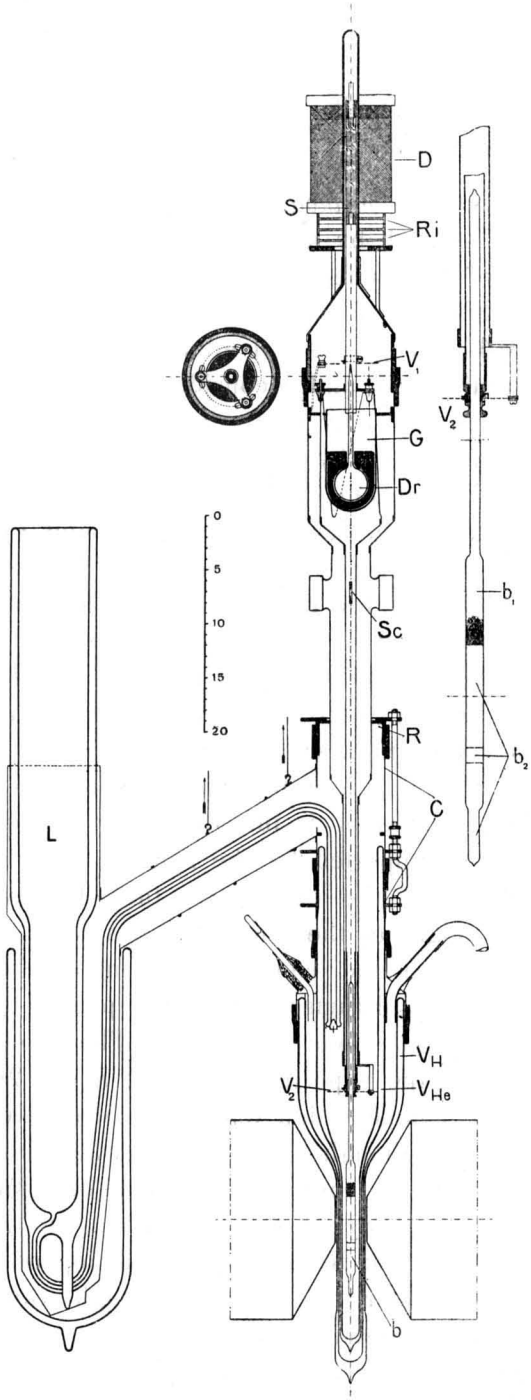

Fig. 3.

The tube (b, cf. the diagram of this detail in fig. 3) containing the magnetic substance, has been made and placed to obtain a symmetrical distribution of glass with respect to a horizontal plane passing through the centres of the poles of the electromagnet. In this way the attraction exerted by the magnet on the glass has been minimised and may be neglected. The dimensions have been chosen such that the sample is at the place of maximum $\frac{\partial H}{\partial z}$ [1], if the tube has been placed symmetrically in the field. The lower part (b_2) of the tube has been evacuated, in the upper part (b_1) a small quantity of helium gas has been introduced in order to improve the temperature equilibrium of the powder and the surroundings and of the particles of the powder mutually. The substance is enclosed between two glass disks, one of which has been melted on the tube, the other is free but is kept in its place by a small plug of cotton wool. Two flattened spiral springs, V_1, V_2 prevent a lateral displacement of the carrier. The lower one has been attached to the carrier and not, as in previous work, to the tube, so that the tubes may be replaced without changing the position of the carrier.

The end faces of the large size Weiss magnet have a diameter of 4 cm and are 26,5 mm apart. The semi-angle of the coneshaped boundary faces is 60°.

The compensating force as function of the intensity of the current in the coil D has often been determined as carefully as possible by suspending weights to the tube. Notwithstanding all precautions unexplained differences subsisted between the different calibrations. The extreme ones differ about 2 °/₀. In calculating a series of observations use was made of the mean of the calibrations "before" and "after".

The specific magnetisation, σ, is calculated from the force measured by means of the relation

$$F g = m \sigma \frac{\partial H}{\partial z}, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (1)$$

where F represents the force (in grammes) exerted on the mass m. The z-coördinate is measured along a vertical from the middle of the interferrum; H is the magnetic force at the point indicated by z; $g = 981.3$.

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[1]) If the susceptibility does not depend on the field strength, the maximum of $\frac{\partial H^2}{\partial z}$ is preferable. [Note added in the translation].

In every set, i. e. every measurement of the force corresponding to a definite value of the magnetic field and a definite temperature, the intensity of the current in the coil D necessary to bring the carrier into a chosen zero position was read the magnetic field being "of" and "on". These readings were taken for both directions of the currents in the coil and in the magnet.

§ 3. *Corrections, auxiliary measurements and sources of errors.*

a. Forces on the carrier without sample. These forces appeared to be not quite negligible and they increased with decreasing temperature. Investigation of the different parts of the apparatus showed that those forces were caused especially by a small screw at the bottom of the carrier (near V_2). The comparatively large increase of these forces when the temperature falls from 20° to 14° K. is very striking, e. g. 70 amp. passing through the electromagnet the attraction amounts to

0,259	gr.	at atmospheric temp.
0,326	,,	20° K.
0,350	,,	14° ,,

This is not what would be expected if the brass of the screw mentioned contained iron as an impurity. Further, such a comparatively very large increase in the liquid hydrogen region would give reason of suspecting much larger forces in the range of helium temperatures. However, they are then not large as appears from there being no systematic difference between the observations in which the mentioned parts of the carrier were certainly below and those in which they were certainly at some distance above the liquid helium level [1]). Particular circumstances prevented determining those forces (whose comparatively large increase in the hydrogen region appeared firstly afterwards) at helium temperatures and in the light of the foregoing remark it seemed not absolutely necessary. In the following observations the correction for the forces on the carrier without sample has been applied for the hydrogen temperatures only.

b. Correction for demagnetisation. This correction may attain considerable values at the temperatures of liquid helium. In the case of a sphere of a homogeneous substance of density d in a homogeneous field the demagnetising field is $-\frac{4}{3}\pi\sigma d$. In our experiments the cirumstances did not correspond exactly to these conditions. The sample is a *powder* in the shape of a small *cylinder*

[1]) Cf. the following communication § 3 note.

and is placed in an inhomogeneous field. Dr. BREIT [1]) has made a careful investigation in the case of a powder. According to him a first approximation for the demagnetisation is obtained if the formula mentioned is applied, taking for d not the density of the powder itself, but of the substance. If necessary this correction has been applied in that manner.

c. Topographical corrections. $\frac{\partial H}{\partial z}$ is in first approximation proportional to the field strength in the middle of the interferrum: H_0. The factor of proportionality was calculated from a ballistic topographical calibration of the magnet [2]). At currents of 10 and 20 amp. no appreciable difference in the topography was stated and for $z = 2.45$ cm. ($\frac{\partial H}{\partial z}$ being there a maximum) was found:

$$H = 0.815 \,.\, H_0, \qquad \frac{\partial H}{\partial z} = 0.199 \,.\, H_0 \quad . \quad . \quad . \quad (2,\ 3)$$

If however for gadolinium sulphate [3]) the force F' is calculated as a function of H_0, no proportionality of F to H_0^2 is found, as might be expected on account of previous measurements [4]) (apart from small corrections if LANGEVIN's formula is followed) but deviations occur up to 20 %. This appears from table I and fig. 4. To the observed value of F, given in the third column now first a correction for the demagnetisation is applied: F is multiplied by $1 + \frac{4}{3}\pi d_0 \chi$; according to the remark b (see above), d_0 is taken equal to 3 [5]), for χ, the specific susceptibility, the value following from the un-corrected measurements has been taken. At 20°.42 K. this correction is 1.2 %, at 13°.98 K. 1.8%. In the column headed L the corrections for the deviations according to LANGEVIN's formula have been given. With those two corrections an apparent Curie-constant $C' = \chi T$ has been calculated.

The values found for C' appear to be strongly dependent on the field strength (cf. fig. 5). This may not be due to errors in the

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1) These Proceedings 25, p. 293; Leiden Comm. Suppl. N⁰. 46.

2) The calibration really refers to a pole distance of 26 mm., not to 26.5 mm., the distance occurring in the experiments described.

The parameters of this field do not belong to those for which FORRER has given so much and such important data (J. FORRER, thesis Zürich, 1919).

3) The gadolinium sulphate, $Gd_2(SO_4)_3 \,.\, 8H_2O$, originated from the supply previously kindly sent by Prof. URBAIN. Two tubes have been filled with it, Gd I and Gd II, containing resp. 0.4735 en 0.4414 gr. of gadolinium sulphate.

4) H. KAMERLINGH ONNES and E. OOSTERHUIS, these Proceedings 15, p. 322 § 6, Leiden Comm. N⁰. 129*b*, § 6.

5) P. GROTH, Chem. Krystallographie II (1908), p. 460.

calibration of the magnetic field. This calibration may be estimated to be accurate to a few thousands. The deviations must be caused by the circumstance that at large and at small values of H_0 the proportionality mentioned may not be expected to hold [1]).

TABLE I.

Gadolinium sulphate II ($m = 0{,}4414$ gr.) $T = 20°.42$ K.								
Nr.	I	F	H_0	L	$10^2\ C'$	q	$10^2\ C$	$\frac{(C-C_m)}{C}$
4	5 amp.	0.81 gr.	3295	0.0	2.100	1.018	2.064	+1.7%
5	5	0.80						
6	10	3.10	6605	0.1	2.015	0.997	2.021	—0.45
3	15	6.98	9875	0.2	2.031	1.000	2.031	0.0
7	20	12.00	12940	0.4	2.038	1.005	2.028	—0.1
2	30	20.66	17320	0.8	1.962	0.963	2.037	+0.3
8	30	20.56						
9	45	25.99	20235	1.2	1.820	0.897	2.029	—0.1
1	60	28.17	21600	1.4	1.729	0.856	2.021	—0.45
10	60	28.00						
$T = 13°.98$ K.								
15	4	0.74	2627	0.1	2.093	1.026	2.040	+0.5
16	5	1.13	3295	0.1	2.032	1.018	1.996	—1.7
14	10	4.52	6605	0.2	2.025	0.997	2.031	0.0
13	20	17.41	12940	1.0	2.046	1.005	2.036	+0.3
17	20	17.39						
18	30	29.32	17320	1.9	1.942	0.963	2.017	—0.7
12	45	37.36	20235	2.6	1.826	0.897	2.036	+0.3
11	60	40.33	21600	3.0	1.739	0.856	2.033	+0.1
19	60	40.44						
20	70	41.77	22230	3.2	1.701	0.835	2.037	+0.3

[1]) In fig. 2 the points for the higher field strengths show the same kind of deviation from the LANGEVIN curve at 4°,25 K. as at 1°,9 K. In my opinion this fact is caused by the absence of proportionality mentioned in the text.

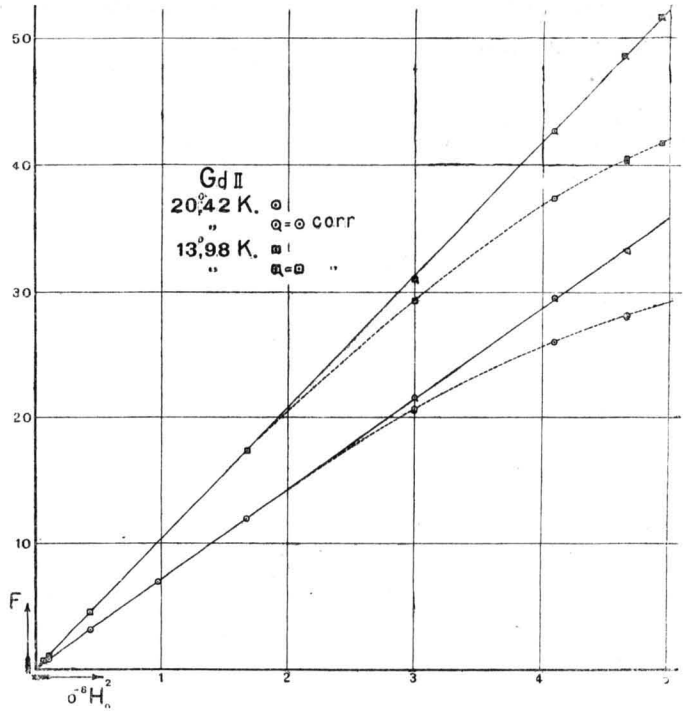

Fig. 4.

We have put:

$$\left.\begin{aligned} H &= s \,.\, 0.815 \,.\, H_0 \\ \frac{\partial H}{\partial z} &= r \,.\, 0{,}199\, H_0 \end{aligned}\right\} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (4)$$

$$q = s \,.\, r \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (5)$$

and for 15 amp. $q = s = r = 1$.

The quantities q, s and r are called the topographical corrections.

The apparent Curie-constant C' is connected to the true Curie-constant C by the formula: $C' = qC$ and does not depend on the temperature. Fig. 5 shows that within the limits of accuracy of the experiments at both hydrogen temperatures [1]) the same values for C' are found. Only at 5 amp. ($H_0 = 3295$) where the forces are small and the measurements less accurate there exists a larger deviation.

The values for C' have been smoothed graphically and then the topographical correction q has been determined from $q = \frac{C'}{C'_{15\text{ amp.}}}$.

In the column $10^2 C$ the value of $10^2 C'$ corrected with q has been

[1]) The circles refer to 20°,42 K., the squares to 13°,98 K.

given and in the last column the difference (in percents) of $10^2 C$ with the mean value 2,030.

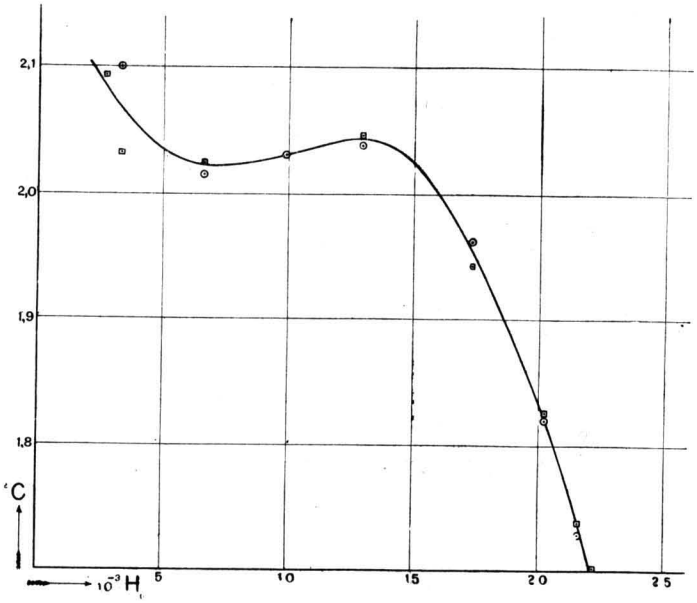

Fig. 5.

r was determined from experiments on the attraction of two small ellipsoids [1]) of Swedish Carbon iron placed as well as possible at the same spot as the substances in the actual experiments. Use was made of the measurements of STEINHAUS und GUMLICH [2]) on the relation between field strength and magnetisation when saturation is nearly reached, the so called law of approach

s was calculated from formula (5). The values found for r and s have also been smoothed graphically [3]).

In these determinations the distribution of magnetism on the pole faces of the magnet has been supposed to be perfectly rigid [4]).

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1) Masses 30.0 and 32.0 mg., major axis 6.2 mm., minor axis 1.1 m.m.

2) Ber. d. Physik Ges. 17 (1915) p. 271.

3) This causes the product of the given r and s to be not exactly equal to q.

4) Cf. P. WEISS, J. de Phys. May 1910 and P. WEISS and H. KAMERLINGH ONNES, Leiden Comm. N⁰. 114, p 16.

Strictly speaking: for a magnet current of 15 amp. the distribution of magnetism on the pole faces of the magnet has been supposed to be perfectly rigid and as regards the other current intensities it has only been supposed to be the same for gadolinium sulphate at hydrogen temperatures and for the S.C. iron ellipsoids. In fact, the magnetic moments are of the same order in both cases (though the volumes on which they are distributed are different); in the case of gadolinium sulphate at *helium* temperatures they are much larger, yet the same values of r and s have been applied (cf. next communication) [Note modified in the translation].

So the values given in table II have been found.

d. Corrections for diamagnetism of the liquid bath and of the anion could be left out of consideration.

e. As regards the *accuracy* and the *sources* of *error* may firstly be pointed out that the *heliumtemperatures* are rather uncertain, especially the lower ones. There was no room for a special stirrer and so the liquid could be stirred only so much as was possible by moving the floating system up and down. Therefore probably the temperature was not always evenly distributed and not perfectly well defined. This is especially important at temperatures below the maximum of density; then the cooling at the surface by evaporation does not give rise to downward convectional currents. However the lower temperatures are not only somewhat indefinite, but the values accepted are not very accurate. They have been determined graphically by means of the total existing material for helium vapour pressures[1]), but this leaves at the temperatures between 1° and 3° K. uncertainties of the order of 0,1 of a degree.

TABLE II.

Pole distance 26.5 mm.; $s = 2.45$ cm.		
I	r	s
3 amp.	0.973	1.062
4	0.983	1.044
5	0.990	1.030
10	0.999	1.003
15	1.000	1.000
20	0.995	1.002
30	0.960	1.010
45	0.893	1.021
60	0.837	1.030
70	0.808	1.035

f. Much care was bestowed on the *adjusting* of the *sample* to the proper place in the magnetic field, or more accurately, of the adjusting of the magnet to the sample, the cryostat not being movable.

[1]) H. Kamerlingh Onnes and Sophus Weber, these Proceedings 18, p. 493; Leiden Comm N°. 147b; H. Kamerlingh Onnes, Leiden Comm. N°. 159 p. 35.

Once the magnet was adjusted in its place, it was marked by means of two plummets suspended to the cryostat and marking two pointers on the yoke of the magnet, for the magnet had temporarily to be removed to afford opportunity of bringing the DEWAR vessels V_{He} and V_H (fig. 3) into place. The large magnet is very heavy and there was no device for moving the magnet sligthly in horizontal direction, so the horizontal adjustment was accompanied by great difficulties and possibilities for inaccuracy.

During the operations with liquid helium and liquid hydrogen the cryostat, forming one whole with the liquefactor, moved slightly in an irregular way as a consequence of the changing temperature circumstances in the different parts. By means of pulling rods the initial position with respect to the magnet was restored.

As far as the adjustment in vertical direction is concerned, it must be pointed out that the distance (at atmospheric temperature) from the centre of the mass to the centre of the field is considered as "place" of the sample in the magnetic field. This place determines the values of the constants in formulae (2)—(5). In the measurements in liquid hydrogen and in liquid helium this place has changed really by the shortening of the carrier in consequence of its cooling. The influence on $\frac{\partial H}{\partial z}$ will be very small as $\frac{\partial H}{\partial z}$ is maximum, but for the same reason the influence on H has to be taken into consideration. In itself there is reason for a correction. In the (rather unfavorable) case that the carrier up to 20 cm above the sample has the temperature of the boiling point of liquid hydrogen and the other part is at atmospheric temperature, a shortening of 0,3 mm would follow from the data of CH. LINDEMANN.[1]) H would be 0,006 H_0 smaller than corresponds to formula (2) i. e. about 0,7 $^0/_0$. Yet no correction has been applied, because it would have required a accurate determination of the place of the substance during the measurements as the sinking of the liquid level changed the temperature distribution along the carrier and thus the place of the sample. Moreover in the measurements in liquid hydrogen and in liquid helium (and the experiments only refer to these temperatures) the correction is nearly equal when the liquid level is on the same height, as the expansion coefficient at these low temperatures rapidly decreases to zero.

g. Finally it must be mentioned that no trace has been observed of the powder particles getting directed or remaining directed by the magnetic forces.

[1]) Physik. Zs. 13, (1912), p. 737.

§ 4. *The Curie constant of gadolinium sulphate.* In § 3*c* it has been mentioned already that for *Gd* II 2,030 $\times$ 10^{-2} has been found.

For the Curie constant of *Gd* I we find:

$T = 20°,31$ K.	$\chi = 1,0566 \,.\, 10^{-3}$	$C = 2,146 \,.\, 10^{-2}$
$T = 14°,68$	$\chi = 1,4663$ „	$C = 2,152$
	mean:	2,149

The measurements on *Gd* I have been considered as less accurate than those on *Gd* II, because (cf. § 3*f* on the difficulties of the adjustment) the tube appeared afterwards for unknown reasons to be not exactly in the middle between the pole faces, but 1,6 mm out of the center. A previous determination of the Curie-constant of *Gd* I quite independent of the present research had given 2,113 $\times$ 10^{-2}. So it is not very probable that the large difference between the Curie constants of *Gd* I and *Gd* II is due to inaccurate adjustment of the tube only. Besides it must be remarked that different observers have found values differing more still than the values mentioned: from the results of Mlle FEYTIS [1]), KAMERLINGH ONNES and PERRIER [2]), and KAMERLINGH ONNES and OOSTERHUIS [3]) the Curie-constant of gadolinium sulphate is found to be [4]).

Mlle FEYTIS	2,167 . 10^{-2}
K. O. and P.	2,086
K. O. and O.	2,016.

These differences are not yet explained.

Finally, I wish to express my sincere thanks to Professor KAMERLINGH ONNES for his kind interest in my work.

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1) Paris C. R. 153 (1911), p. 668.

2) These Proceedings 14, p. 115; Leiden Comm. N°. 122a.

3) „ „ 15, p. 322; Leiden Comm. N°. 129b.

4) A correction has been applied for the diamagnetism of the crystal water and of the anion. The first correction had been applied already by Mlle FEYTIS.