Physics. — The law of magnetisation of solid crystals. Resolution of the FARADAY-effect into two effects of different origin. Diamagnetic and paramagnetic rotation of the plane of polarisation. By JEAN BECQUEREL and W. J. DE HAAS. (Communication N<sup>0</sup>. 193a from the Physical Laboratory at Leiden.)

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## I. Introduction.

In a summary of earlier researches, twenty years ago, one of us 1) drew attention to the following fact : the rotatory power of some minerals that contain rare earths (erbium, neodymium, praeseodymium) increases considerably when the temperature falls and is inversely proportional to the absolute temperature. Such a remarkable correspondence with the law of CURIE led the first author to prove the existence of a rotatory power that is intimately connected with paramagnetism. In paramagnetic bodies this rotatory power is superposed on the diamagnetic rotatory power, which is connected with the ZEEMAN-effect and is just as general as this.

We now give the decisive proof of the existence of the paramagnetic rotation of the plane of polarisation. But before describing our experiments we will give a historical review of the problem. Long ago we find the idea of a connexion between the FARADAY-effect and the magnetic properties of a body. Defended by some physicists, disputed by others, this hypothesis has given rise to may discussions.

Four years after the discovery of FARADAY EDMOND BECQUEREL<sup>2</sup>) showed, that the salts of iron in aqueous solution considerably diminish the magnetic rotatory power of the liquid. This induced VERDET<sup>3</sup>) to ascribe to the salts of iron a rotatory power opposite to that found in all experiments of FARADAY. VERDET distinguished these two directions as positive<sup>4</sup>) (water, glass, etc.) and negative.

An obvious hypothesis is, that the negative sign given by iron salts is caused by their magnetic properties, that as a rule diamagnetic substances

<sup>&</sup>lt;sup>1</sup>) JEAN BECQUEREL, le Radium 5, 16, 17, 1908; Rapports présentés au premier congrès intern. du froid 2, 43, 44, 1908.

JEAN BECQUEREL et H. KAMERLINGH ONNES, Comm. Leiden n<sup>0</sup>. 103; le Radium 5, 238, 1908.

<sup>&</sup>lt;sup>2</sup>) EDMOND BECQUEREL, Ann. de Chimie et de Physique, 3e série, 28, 334, 1850.

<sup>3)</sup> VERDET, Ann. de Ch. et de Phys. 3e série, 52, 144, 1858.

<sup>&</sup>lt;sup>4</sup>) The rotation is called positive, when to the observer receiving the light that is propagated in the direction of the lines of force, the plane of polarisation is rotated anti-clockwise.

give a positive rotation and paramagnetic bodies a negative one. Titanium tetrachloride, although diamagnetic, gives a negative rotatory power. In VERDET's opinion this fact proved the independence of the sign of rotation upon the magnetic properties. HENRI BECQUEREL <sup>1</sup>) studied the magnetic substances. He found, that "the magnetic rotation of the bodies is connected with their refractive index and with another function that varies with their specific magnetism". For the first time he showed the connexion between the rotatory power and the magnetic properties of matter.

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Unfortunately at that time (1876) the knowledge of the origin of magnetic phenomena was practically nil, so that the causes of the FARADAY-effect could not be found out. HENRI BECQUEREL ascribed the negative rotation to the production of an internal magnetic field with sign opposite to that of the external field, an hypothesis which has been taken up again by several physicists. Though the idea was plausible, our investigations lead the problem in another direction in connexion with the actual theories of magnetism.

We must now remember an important phenomenon discovered in 1884 by KUNDT<sup>2</sup>): Ferromagnetic metals in leaves so thin as to let through enough light (some thousandths of a millimeter) possess an enormous magnetic rotatory power. The rotation is not proportional to the external field : it asymptotically approaches a limit. According to DU BOIS<sup>3</sup>) the rotatory power is proportional to the *magnetisation* and is *positive*.

One of the principal reasons of DRUDE for advancing his theory called : "the hypothesis of the molecular currents" <sup>4</sup>) was the explanation of the KUNDT-effect. DRUDE assumes in paramagnetic bodies the existence of special currents which are partially or wholly directed in the magnetic field. In diamagnetic bodies these currents would not previously exist, but they would only be induced by the external field. In the most simple case this theory gives a rotatory power proportional to the magnetisation and leads to variations of the rotatory power of opposite sign at the two sides of an absorption band, both for diamagnetic and for paramagnetic bodies.

In his "Lehrbuch der Optik" DRUDE gives yet a second theory, which he calls the "hypothesis of the HALL-effect". This theory is the same as that given before by W. VOIGT <sup>5</sup>) : the magnetic rotatory power is a consequence of the ZEEMAN-effect and of the dispersion. At both sides of an absorption band the variations in the rotatory power must have the same sign, in contradiction with the other theory.

<sup>&</sup>lt;sup>1</sup>) HENRI BECQUEREL, Ann. de Ch. et de Phys. 5e série 12, 42, 1877. We remember, that in 1897 (C. R. 125, 679) H. BECQUEREL gave a theoretical expression for the magnetic rotation. This expression is identical with that which was afterwards derived from the LARMOR precession; it concerns the diamagnetic rotation.

<sup>&</sup>lt;sup>2</sup>) A. KUNDT, Wied. Ann, 23, 228, 1884; 24, 191, 1886.

<sup>3)</sup> H. DU BOIS, Wied. Ann. 31, 941, 1887.

<sup>&</sup>lt;sup>4</sup>) DRUDE, Lehrbuch d. Optik, 406, 1906.

<sup>5)</sup> W. VOIGT, Wied. Ann. 67, 345, 1899; Ann. d. Phys. 6, 784, 1901; 8, 872, 1902.

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W. VOIGT does not seek the origin of the FARADAY-effect in the magnetisation and he disputes the theory of the "molecular currents". He expresses the opinion that the external field can give rise to internal forces <sup>1</sup>) which for magnetic bodies may be nearly proportional to the magnetisation, so that for thin leaves of ferromagnetic metals a direct influence of the magnetisation on the magnetic rotatory power seems to exist. He comes back to the idea of a field of sign opposite to that of the external field. As to the sign of the KUNDT-effect, which is positive not-withstanding the reversal of the field, this would be explained by the absorption being due to "free electrons".

At the time of the publication of the work of W. VOIGT (1908) the greater part of the physicists kept to the "hypothesis of the HALL-effect", at least for non-conducting bodies. The theory found its origin in the experiments of MACALUSO and CORBINO and of several physicists on the rotatory power of vapours in the neighbourhood of lines that show the ZEEMAN-effect. One of us extended these investigations to solid bodies by experiments on the magnetic rotatory power of crystals of rare earths in the neighbourhood of those absorption bands, for which the absorption of oppositely circular polarised rays had the same value. Even a *quantitative* verification of the theory was given <sup>2</sup>).

On the other hand an important fact was observed in the course of the investigations of crystals of the rare earths. For many bands the opposite circular rays were unequally absorbed under the influence of a magnetic field. A dissymmetry existed for the two circular components. This new phenomenon was directly considered in connexion with paramagnetism <sup>3</sup>).

Here follows, what was written in 1906 :

"In my opinion, we have here to do with an intrinsic manifestation of "the molecular magnetism. The projection of the motion of each electron on "a plane perpendicular to the axis may be resolved into two opposite circular "vibrations. When we suppose, that the paths of the electrons or certain "groups of electrons can be orientated under the influence of the external "field and that the sum of the circular motions in one direction becomes "perceptibly different from the sum of the oppositely directed motions, one "component becomes stronger with respect to the other and a dissymmetry "in the intensities is observed.

".....On the other hand the electrons together will be equivalent to a "magnet directed along the lines of force. So the crystals of xenotime are "magnetic and a cube is orientated, so that the axis becomes parallel to "the field.

"By means of a quarter-wavelength plate combined with a BABINET "compensator it is proved, that the circular double refraction i.e. the

<sup>1)</sup> W. VOIGT, Magneto- und Elektrooptik, 21. 1908.

<sup>7)</sup> JEAN BECQUEREL, le Radium 5, 15, 1908; W. VOIGT, Magneto- und Elektro-optik § 133, 232.

<sup>3)</sup> JEAN BECQUEREL, C. R. de l'Ac. des Sc. 143 (24 déc. 1906).

"magnetic rotation of the plane of polarisation, has opposite signs at the "two sides of the band.....

"The rotatory power varies rapidly in the neighbourhood of the band "notwithstanding its small intensity. The facts may easily be explained "by noticing that, because of the considerable inequality between the two "components, the anormal dispersion for one circular vibration is much "greater than for the other.

"The question arises, whether the anomalies shown by magnetic bodies "and the great dispersion of the rotation for some of them may not be "connected with a phenomenon of the same nature. The deformations of "the linear vibrations, especially after their passage through the iron leaves, "may be caused by a similar effect."

Thus the different dissymetries in the intensities and the effect of the magnetic rotation of the plane of polarisation have been ascribed long since to a paramagnetic orientation 1). The theory of VOIGT treats the magnetic rotation, but it does not explain the paramagnetic rotation or rather, for this phenomenon the theory must be completed by introduction of the effect of the orientation, which causes an inequality in the absorption of oppositely directed circular vibrations. This was said shortly in one of the sentences of the above quoted passage. Recently R. LADENBURG <sup>2</sup>) did the same. Without knowing the above cited paper, he proceeded from the same ideas and expressed them in mathematical form. R. LADENBURG also did not go further as he too gave a theory of the dispersion of the paramagnetic rotation of the plane of polarisation.

The lower the temperature, the better do the intensities show the dissymetry of paramagnetic origin <sup>3</sup>). We observed that at a temperature of  $4.2^{\circ}$  K. and in a field that need not be strong (10000 Gauss) some bands absorb circular vibrations of only one direction <sup>4</sup>).

Apart from local disturbances caused by absorption bands in the visible spectrum, the great rotatory power of crystals that contain rare earths and its increase with fall of temperature must be due to strong absorption bands in the ultra violet.

In the earlier experiments, which were all made at room temperature or at higher temperatures, only relatively low values were found for the

<sup>&</sup>lt;sup>1</sup>) Of the more recent papers on paramagnetic rotation must be mentioned those of ELIAS on the rare earths (Ann. d. Phys., **35**, 298, 1911); those of ROBERTS, SMITH and RICHARDSON on salts of iron and cobalt in solution (Phil. Mag. **44**, 917, 1922; **49**, 397, 1925); and those of DORTMANN on the dispersion of the magnetic rotation of liquid oxygen and on the solutions of salts of rare earths (Zeitschr. f. Phys. **17**, 93, 1923).

<sup>&</sup>lt;sup>2</sup>) R. LADENBURG, Zeitschr. f. Phys. **34**, 898, 1925; **46**, 168, 1927. The discussion of the work of LADENBURG must be postponed until our next paper. There we shall show that our experiments agree within one thousandth with the dispersion law of LADENBURG.

<sup>&</sup>lt;sup>3</sup>) JEAN BECQUEREL, le Radium 5, 9, 1908; JEAN BECQUEREL and H. KAMERLINGH ONNES, Comm. Leiden n<sup>0</sup>. 103 (§ 10), 1908; JEAN BECQUEREL, le Radium, 6, 330, 1909.

<sup>&</sup>lt;sup>4</sup>) JEAN BECQUEREL, H. KAMERLINGH ONNES and W. J. DE HAAS, Comm. Leiden  $n^0$ . 177, 1925.

paramagnetic rotatory power. According to the circumstances it was the paramagnetic or the diamagnetic effect that predominated. It is evident, that at that time many physicists did not recognize the connexion between the direction of the rotatory power and the magnetic properties. Therefore it is interesting, that HENRI BECQUEREL felt convinced of the existence of such a connexion, because he had observed that a substance with a negative rotatory power (e.g. TiCl<sub>4</sub>) always contained a paramagnetic component.

It was also this rule, which induced one of us to make use of low temperatures for the separation of the paramagnetic from the diamagnetic effects in magneto-optical phenomena 1). We must remark, that the paramagnetic rotation need not be negative 2). In transparent substances however this is the sign which has been found until now at the side of the long wavelengths compared with those of the active bands for which the effect predominates.

## II. The experiments. The cryostat.

We shall omit a description of the spectroscopic arrangement. This is the same as that used in the physical laboratory of Prof. BECQUEREL since 1906, afterwards used also at Leiden in 1908 and 1924. We only mention again, that we used a plane ROWLAND grating with 568 lines per mm., 8 cm. wide and provided with auto-collimation with a lens of 1.3 m. focal length. The spectra observed or photographed were of the first or second order. The source of light was an arc-lamp.

The cryostat consists of three vacuum glasses. In the innermost one the liquid helium boils, in the second which surrounds the first, liquid hydrogen boils and in the glass surrounding the first two boils liquid nitrogen. The glasses were blown with great skill. The inner tube of the helium-glass has a diameter of 5 mm., the outer tube of the nitrogen-glass one of 15 mm. The light must pass through twelve glass walls and five layers of liquid. The pressure of the helium which boils in the inner glass is measured by means of a closed mercury manometer. During an experiment the pressure is kept accurately constant by means of a regulating cock.

In the cryostat cap a metal rod can both turn round its longitudinal axis and be moved up and down. To this metal rod, in the cryostat, a glass rod has been glued at the end of which the holder for crystals is glued. By vertical displacement each crystal may be brought into the beam of light and if necessary the crystal axis may be turned in a horizontal plane.

## III. Object of the investigation. Measuring method.

The object of the investigations was the determination, for some wavelengths and for some very low temperatures, of the variation of the magnetic

<sup>1)</sup> JEAN BECQUEREL, le Radium, 5, 6, 1908.

<sup>2)</sup> See LADENBURG, Zeitschr. f. Phys. 34, 903, note; 46, 172 and 176.

rotatory power as a function of the intensity of the field. Among the monoaxial crystals one seems especially suited for this research viz. *tysonite* ( (La, Ce, Nd + Pr)  $F_3$ ) because of the following reasons:

1º. Of the crystals investigated until now tysonite has the greatest magnetic rotatory power.

20. The crystal is beautifully transparent and can therefore be used in thicker plates than other crystals. With a plate of 1.866 mm., which we possess and notwithstanding the twelve glass walls of the cryostat the spectrum is of good intensity.

30. The absorption bands in the visible spectrum are nearly all combined into groups and for the greater part are little active except the band  $\lambda = 5776$ . Moreover they are separated by large intervals in which their influence can be neglected. When the wavelength is chosen within these intervals, only the principal phenomenon is observed, separated from all local disturbances. As the earlier investigations, mentioned in § I, had shown that the magnetic rotatory power must be of paramagnetic origin, we might expect to find at sufficiently low temperatures a deviation from the law of proportionality to the field strength, which law had always been found since FARADAY. It was even to be expected, that the rotation would

be a function of  $\frac{H}{T}$ . We need not point out the importance of the determina-

tion of this function for the theory of magnetism.

For the study of this phenomenon a very accurate measurement is of course required, but before all we must choose a very quick method, if necessary somewhat sacrificing the accuracy.

Now at very low temperatures the magnetic rotatory power of tysonite is enormous, so that a determination of the rotations between 1500° and more thans 5000° to an accuracy of 1°, is not only unnecessary but of imaginary advantage, as the determination of the field with an accuracy of  $1/_{100}$ ° is already difficult.

The measurements must be made quickly in an uninterrupted series under conditions as fixed as possible; otherwise they are not well comparable after reduction to one and the same temperature. We have chosen the most simple method viz. that of the canaliculated spectrum.

The light of an arc-lamp is sent along the lines of force through the poles of an electro-magnet, which are pierced with holes of 1 mm. diameter. This beam is sufficiently parallel. The crystal plate which may be regarded as cut perfectly normal to the axis has been fixed with the greatest care with the aid of a BABINET compensator <sup>1</sup>) in such a way, that the axis is parallel to the incident light. When the compensator and analysor, which were used for the adjustment, are taken away, the incident light is so

<sup>&</sup>lt;sup>1</sup>) This method has been described in the former communication of the Phys. Lab. Leiden N<sup>0</sup>. 191c II § 11. From measurements of the phase differences between the transmitted vibrations with oblique direction of incidence we conclude statistically that the position of the plate is sufficiently perpendicular to the axis.

polarised that the electric vector is vertical. We then put before the slit of the spectroscope a rhombohedrar of iceland spar which forms a doublerefracting analysor. In this way we obtain two spectra in juxtaposition, and when there is no field the spectrum corresponding with the horizontal vibrations is extinct 1).

The light of an iron arc-lamp, standing on one side, is reflected by a mirror and can be thrown on the slit of the spectroscope whenever we want to superpose the iron spectrum on that of the crystal in order to determine the wavelengths.

As soon as the current of the electro-magnet is switched on the magnetic rotation is observed and as the field increases we see that in the two adjacent spectra numerous black lines are displaced from the violet towards the red.

We now choose either from the iron spectrum, or from the absorption spectrum<sup>2</sup>) of the crystal the radiation for which we are going to make the measurements and we look at the passage of the black lines through the chosen spectral line. In this way we find the rotations  $n\pi$  in the spec-

trum of the horizontal vibrations and  $\left(n\pi+\frac{\pi}{2}\right)$  in the spectrum of the

vertical vibrations.

At each passage we note the intensity of the current, which gives the intensity of the field, the curve that gives H as a function of the current having been plotted beforehand.

For this determination of the field as a function of the current we made use of the deviation of the components of the line 5221 of xenotine (at the temperature of liquid air). This line is fine and very sensitive, for it gives a deviation  $8\frac{1}{2}$  times that of the normal ZEEMAN-effect <sup>3</sup>).

The individual errors in the measurements can be of the order of 5  $^{0}/_{00}$ in a field of 30000 Gauss, and they are the greater the weaker the field. These errors may be corrected to a large extent by the form of the curve.

We have preferred this method because it gives the field between the pole-pieces, i. e. just at the place where we must know it.

The maximum field that was used (current of 150 Ampère, distance between the pole-pieces 16 mm.) was found to be 26.73 kilogauss.

The measurements were made at the following temperatures, and for the accompanying wavelengths.

<sup>1)</sup> At very low temperatures the extinction is never total; for the residual magnetism of the pole-pieces is sufficient to give a rotation of several degrees.

<sup>2)</sup> We chose an isolated fine line, which is inactive or sufficiently so to cause no appreciable disturbance. We must still mention that the black line almost wholly covers the spectral line in one spectrum, but that this line remains visible in the other spectrum, which then has its maximum intensity. This makes the observations possible.

<sup>3)</sup> It is an acknowledged fact, that the deviation is proportional to the field. For the absolute measurements the calibration was made at Zürich in 1909. The deviation was determined on a cliché in a field, the intensity of which had been measured by Prof. WEISS (7.71 Å for 349000 G).

$$T=4^{\circ},21 \begin{cases} \lambda = 4258,9 \text{ (line of tysonite)} \\ \lambda = 5328,5 \text{ (middle of an iron-doublet)} \\ \lambda = 6249,5 \text{ (line of tysonite)} \end{cases} \begin{cases} \text{thickness of the plate} \\ e = 1.866 \text{ mm.} \end{cases}$$
$$T=1^{\circ},71^{4} \begin{cases} \lambda = 4258,9 \\ \lambda = 5328,5 \end{cases} \text{ with two plates for which:} \begin{cases} e_{1} = 1.866 \text{ mm.} \\ e_{2} = 0.675 \text{ mm.} \end{cases}$$
$$T=1^{\circ},41^{2} \\ \text{and } 1^{\circ},39^{4} \end{cases} \lambda = 5328,5 \end{cases} \text{ thickness of plate} \quad e = 1.866 \text{ mm.}$$

The given temperatures below  $2^{\circ}$  are a little incertain. We shall see further on why the above temperatures have been assumed.

The measured negative rotations are corrected for the positive rotation caused by the cryostat, which may not be neglected (5°, for 5890, 12.5° for 4000 Å).

## IV. Approach of the saturation. Law of the paramagnetic rotation as a function of $\frac{H}{T}$ . The magneton.

First we have plotted the curves of the rotations as a function of the field.

At  $4.21^{\circ}$  it is already evident, that the rotation is no longer proportional to the field 1). At temperatures of  $1.7^{\circ}$  and of  $1.4^{\circ}$  the deviation from proportionality is considerable.

We shall begin with the discussion of the results at  $1.7^{\circ}$ , which we found for two lines and with two plates of different thickness.

We plotted the curves  $\varrho = f(H)$  in such a way that they represented the experimental results as truly as possible and without proconceived opinion i. e. without making use of any theoretical law. For all these different curves we have established that their ordinates bear a fixed ratio to each other viz. that the rotations found for the same wavelength with two different plates are proportional to the thickness of these plates.

Let e.g. a and b the experimental curves for the wavelengths 5328.5 and 4258.9 for the plate with e = 1.866 mm. We take all measurements for the violet line and their ratio to the ordinates of the curve a for the same field intensities. Then we take the observations for the green ray and the ratio's between the points of the curve b and the measured values for the green line. The mean of all these ratio's gives with very good accuracy the ratio between the rotations for the violet and for the green, which ratio is independent of the field (of the 32 values obtained 16 differ less than

<sup>&</sup>lt;sup>1</sup>) This deviation from the proportionality which is not large at  $4.2^{\circ}$  escaped our first observations (Comm. Leiden n<sup>0</sup>. 177) because we had confined ourselves to 3 determinations and the 3 values of the field intensity measured separately were not known with the same accuracy as those which are derived from numerous measurements and thus permit us to lot the field-current-curve.

2  $0/_{00}$  from the common mean and the two largest deviations are only  $+7 0/_{00}$  and  $-5 0/_{00}$ ).

For the two plates used, the ratio of the rotations for the same wavelength proved to be equal within 1/1000 the ratio of their thicknesses as measured with the comparator.

Thus the first result deduced from the experiments is that the law of the variation of the magnetic rotatory power as a function of the field is independent of the wavelength. The problem now is to find this law. After having measured, as has been said, the ratio's of the rotations for the two plates and for the two lines used we reduced all measurement to one curve  $C_1$  which represents all measurements for the temperature considered (1.7°) and can be plotted in a very sure way.

Until now the only investigations that showed an approach to the saturation, were those of WOLTJER and KAMERLINGH ONNES <sup>1</sup>) with gadolinium sulphate. As these authors concluded from their investigations, that the law of magnetisation was that of LANGEVIN, we first tried to identify the curve C with that of LANGEVIN. We found however systematic deviations, which though not large, still exceeded the possible errors. Moreover it is natural, that the law is different from that of LANGEVIN, as the conditions are quite different from those for which he made his theory. We must keep in mind that the investigations of WOLTJER and KAMERLINGH ONNES were made with crystalline powder and not with an orientated crystal.

For the magnetisation in one of the principal directions of a crystal LENZ<sup>2</sup>) and afterwards EHRENFEST<sup>3</sup>) deduced in a theoretical way a more complete theory, in which the function in question contains the tangens hyperbolicus. We have tried this theory and this time with success. The curve C is represented as well as might be expected by the formula :

$$y = A tgh C H$$
.

The value of C that gives the best agreement between the theoretical and experimental curves is :

$$C = 3.923.10^{-5}$$
.

With this value of C the deviations of the theoretical curve from the experimental one are of the order of the width of a pencil-line on a plot of  $80 \times 80$  cm. It is evident that the primary cause of the magnetic rotation is a magnetisation effect and we see that this relation is expressed by a simple law <sup>4</sup>).

The tangens at the origin coïncides with the curve over a considerable length; this is the law of CURIE.

<sup>1)</sup> WOLTJER and KAMFRLINGH ONNES, Comm. Leiden n<sup>0</sup>. 167.

<sup>&</sup>lt;sup>2</sup>) W. LENZ, Phys. Zeitschr. 21, 613, 1920.

<sup>3)</sup> P. EHRENFEST, Comm. Leiden Suppl. n<sup>0</sup>. 44b.

<sup>&</sup>lt;sup>4</sup>) At first view it seems surprising to find a simple law for a crystal containing three paramagnetic ions, but in a following communication we shall show, that the paramagnetic orientation is due to only one of these three ions.

There is no reason for introducing a molecular field worth considering. Nor is it necessary to take into account a demagnetising field, as the rotation is proportional to the thickness of the plate.

Assuming that the rotation is proportional to the magnetisation effect to which it is due (which seems to be true, though it has not been proved 1)), we must identity CH with the variable a of LANGEVIN; just as in the theories of LENZ and of EHRENFEST. We then have :

$$C = \frac{\Sigma_{\infty}}{RT}$$

where R is the gas-constant and  $\Sigma_{\infty}$  the saturation-magnetisation reduced to a gram-molecule and expressed in the magnetic moment of the unit of magnetism the so-called magneton  $\mu$ , which determines the phenomenon. So we can find  $\Sigma_{\infty}$ , when the temperature is known.

For these experiments the pressure of the helium was reduced to 9 mm. The temperature of helium boiling under a pressure of 9 mm. lies between 1.66° and 1.76° K. The temperature cannot be given with certainty, as the temperature-scale of the gasthermometer has not yet been fixed <sup>2</sup>).

With 1,66° we find 
$$\Sigma_{\infty} = 0.968 \ \mu_{Bohr} = \mu_B$$
  
, 1,76° ,  $\Sigma_{\infty} = 1.026 \ \mu_{Bohr}$ .

Perhaps it is merely fortuitous, that the value of the BOHR magneton lies just between the values of  $\Sigma_{\infty}$  that are found with the two estimations of the temperature. But it is more probable that  $\Sigma_{\infty}$  is exactly the magneton. We must moreover keep in mind that for this orientated crystal we must consider the quantisation of the paths with respect to the common direction of the axis and of the field.

If we suppose it is established that  $\Sigma_{\infty} = \mu_B$ , then it is the temperature that is determined by our experiments. It is then 1.714°, except for the errors in the determination of the absolute intensities of the field and in the value of the magneton.

If it were possible to proceed with the measurements at 4.21° in the same way as we have just done, it would be possible to identify the experimental rotation curve at the known temperature of liquid helium under normal pressure with the tanh-line and no uncertainty would remain in the value of  $\Sigma_{\infty}$ . At 4.21° however the magnetic moment is too small and the curvature too weak to identify the curve with sufficient certainty. Only the following is sure. If we plot curves of  $\varrho$  not as a function of H but as a function of  $\frac{H}{T}$ , then the curves of the measurements at 4.21° and those which we obtain by taking for the temperature of the above described experiments 1.714° have ordinates that bear in a remarkable way a fixed ratio to each other.

<sup>&</sup>lt;sup>1</sup>) At all events the proportionality has been proved for high temperature, the rotation and the magnetisation being then both proportional with the field.

<sup>2)</sup> Investigations on this subject are proceeding at Leiden.

Thus the law of the hyperbolic tangent represents not only the results for helium under a pressure of 9 mm., but also (but for a factor) the experimental results at  $4.21^{\circ}$  for the three lines used.

We might also assume  $1.66^{\circ}$  of  $1.76^{\circ}$  for the temperature of the reduced helium without appreciably disturbing the proportionality between the ordinates of the curves. The values of  $\frac{H}{4.21}$  remain too small to deviate the tang-hyp. curve much from its tangent at the origin.

Finally the law of the hyperbolic tangent has been confirmed by two other series of measurements which we made after having reduced the pressure with two coupled Siemens pumps. If we start from the hypothesis, that the carrier of the magnetic moment has a magnetic moment equal to the magneton of BOHR, we find for the temperatures of the two series  $1.41^{\circ}$ and  $1.39^{\circ}$ . By application of the vapour tension formula of VERSCHAFFELT (pressure of the order of 2.8 mm.) we find a temperature of about  $1.4^{\circ}$ .

Fig. 1 represents the hyperbolic tangent obtained by reducing all measurements with the two plates for the different temperatures and for the different wavelengths to one curve with the aid of the previously determined ratios between the ordinates of each curve and those of one definite curve. The full curve is the hyperbolic tangent, calculated under the assumption of the BOHR magneton and the temperatures derived from it (see above):  $4.21^{\circ}$ ,  $1.71^{\circ}$ ,  $1.41^{\circ}$ ,  $1.39^{\circ}$ . Two scales of abscissae are given, the scale in  $\frac{H}{T}$ 

and the scale for the variables a, where

$$a=\frac{\mu_B H}{RT}.$$

The ordinates are the rotations expressed in the saturation rotation viz.  $\frac{\varrho}{\rho}$ . The last point corresponds with 85.7 % of the saturation.

The last points, obtained at the very lowest temperatures, are the result of measurements made with special care. At the temperature of  $1.7^{\circ}$  some uncertaintity existed in the determination of the strongest fields as may be seen in the figure (between the values 14300 and 15300 of  $\frac{H}{T}$ ). The cause lies either in the fatigue of the observers towards the end of the series of measurements or in the fact that the method of the canaliculated spectrum becomes less sensitive the closer the black lines are together. The same error in the position of a black line gives then of course an increasing error in the magnitude of the rotation derived from it 1). Perhaps also variations of the temperature play a role.

<sup>&</sup>lt;sup>1</sup>) In weak fields also the sensitiveness decreases but for another reason: the black lines become too wide. The increase in sensitivity by their large displacement for a small change in the field does not compensate for the decrease of the sensitivity by the difficulty in determining the middle of the lines.

We have therefore repeated the experiments and lowered the temperature still more in order to enter a region where the curve shows a strong curvature. We concentrated our forces principally upon the determination of the last points (means of several observations), so that these last points



may be considered as very certain, especially since we had taken measures to keep the temperature very constant.

The two dotted curves are LANGEVIN curves, which have the same

tangent at the origin as the hyperbolic tangent (the tangent in the origin seems to be well established).

The lowest curve passes through the last point; it is however not good, evidently being too low for the measurements taken as a whole, corresponding to the mean  $\frac{H}{T}$  values. The highest curve represents very well the measurements up to  $\frac{H}{T} = 10000$ , but it is not good for higher values of  $\frac{H}{T}$ .

It is evident that no LANGEVIN curve gives such a strong curvature for the high values of  $\frac{H}{\overline{T}}$  as the experimental curve.

In the case of tysonite the paramagnetic rotation is very well represented by the law:

$$\varrho = (\varrho_{\infty})_{\lambda,\tau} tgh a ; a = \frac{\Sigma_{\infty} H}{RT}$$

and  $\Sigma_{\infty}$  seems to be the BOHR magneton.

It is necessary to investigate the paramagnetic rotation of other crystals. If the law found above is exactly valid for tysonite, which is very probable, then the saturation constant is determined by the ratio  $\frac{\varrho}{tgha}$  which is a constant for a given temperature and a given wavelength. The values obtained are given below expressed in radians. They refer to the thicket plate (e = 1.866 mm. at room temperature).

 $T_{k} = 77,55^{\circ} 20,36^{\circ} 4,21^{\circ} 1,71^{\circ} 1,41^{\circ} 1,39^{\circ}$   $\begin{cases} 6249 \text{ Å} & -14,66\pi \\ 5328,5 \text{ Å} & -24,61\pi - 21,81\pi - 19,61\pi - 18,74\pi - 18,73\pi \\ 4258,9 \text{ Å} & -51,60\pi - 45,21\pi - 40,02\pi - 36,01\pi \end{cases}$ 

The numbers given for the temperatures of  $77.55^{\circ}$  (liquid nitrogen) and of  $20.36^{\circ}$  (liquid hydrogen) are the results of measurements made with different wavelengths in a field of 26.73 kilogauss. The rotations for 5328.5 Å and 4258.9 Å have been read on the full curve and then divided by tanh a.

For one and the same wavelength the saturation rotation decreases with the temperature and therefore the rotation  $\rho$  also decreases with the temperature for a given value of  $\frac{H}{T}$ . For a given value of H however it increases up to the temperature 1.39°.

The decrease of  $\rho_{\infty}$  with the temperature may be easily explained: the paramagnetic rotation is caused by the carriers of the magnetic moment, but the paramagnetic phenomenon manifests itself optically indirectly through the dispersion of the light. In order that a rotatory power can be observed, it is necessary, that one or more *active* absorption bands exist. By active is meant ,,which absorb to a different degree opposite circular polarised rays". The variability of  $\rho_{\infty}$  is a manifestation of the fact, that the active absorption bands are not independent of the temperature, which moreover is evident for all bands in the visible spectrum. This change with the temperature might consist of a displacement in the spectrum, but this could not be great and could have only slight influence. The intensity of the bands in particular must vary and the decrease of  $\rho_{\infty}$  is the proof of a decrease of the absorption. It is impossible to extrapolate and to say before hand whether the intensity of the bands approaches a finite limiting value or that it always decreases as the temperature approaches zero or that it passes perhaps through a minimum.

The variability of  $\rho_{\infty}$  explains why the rotation in weak fields or at less low temperatures does not exactly follow the law of CURIE.

First we have plotted the curves for  $\frac{1}{\varrho}$  as a function of T for a constant H, and it might be thought that the deviation from the rectilinear law, which is accentuated at very low temperatures, is caused by a cryomagnetic anomaly. The explanation however is quite otherwise : the deviation from the rectilinear law is caused 1<sup>st</sup> by the approach of saturation 2<sup>nd</sup> by the decrease of the absorption. In reality, as long as  $\frac{H}{T}$  is not too large, the law of CURIE is obeyed for the magnetisation which is the primary cause of the optical effect (at the origin the tanh.curve coïncides with its tangent) but not of the magnetic rotatory power; the absorption makes the effect more complicated.

Finally we wish to express our thanks to Miss RIETVELD for her valuable assistance in the measurements.