

**Physics.** — *The principal susceptibilities of Manganese Ammonium-sulphate crystals at low temperatures.* By L. C. JACKSON and W. J. DE HAAS. (Comm. Number 187c from the Physical Laboratory at Leiden.

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*Introduction.* Crystalline powder of manganese ammonium sulphate is known to follow the law of CURIE down to the lowest temperatures at which it has been examined <sup>1)</sup>. The data available at the time of the research, seemed to indicate that the  $1/\chi \cdot T$ -lines for the principal susceptibilities of paramagnetic crystals (if these lines are straight) are parallel to each other. It therefore seemed important to examine the principal susceptibilities of a crystal of manganese ammonium-sulphate. We might namely expect either the three principal susceptibilities to be equal and consequently the crystal to be magnetically isotropic, though belonging to the monocline system, or, what was more probable both positive and negative values of the constant  $\Delta$  to occur in the formula  $\chi(T + \Delta) = \text{constant}$ .

It seemed however, just as well possible that each of the three principal susceptibilities follows the law of CURIE.

§ 1. *Experimental research.* We grew large well-formed crystals of manganese ammonium sulphate and made sections from these in the shape of small cylinders with the aid of a crystal grinding-goniometer, so that the axes of these cylinders form a definite known angle with the crystallographic axis. In order to determine the three principal susceptibilities and the angle between the susceptibility  $\chi_1$  and the  $c$ -axis three differently orientated disks from the crystal are necessary, while the fourth equation is given by the known value of the susceptibility  $1/3 (\chi_1 + \chi_2 + \chi_3)$ .

The susceptibilities in the directions of the axes of the disks were then determined by measuring the force exercised on the small cylinders in a non-homogeneous field, that is with the method of CURIE. The value of  $H \frac{dH}{dX}$  being not yet known at the place where the disk was put, the apparatus was calibrated with the aid of a thin celluloid capsule exactly of the same form as the little disks. It was fixed in the magnetic field in exactly the same position as the disks and filled with the finely divided material. The force on the empty capsule was proved to be neglectable.

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<sup>1)</sup> L. C. JACKSON and H. KAMERLINGH ONNES, Proc. Roy. Soc. London Oct. 1923. Comm. N<sup>o</sup>. 168a.

Thus the susceptibilities of the crystal disks were determined in the temperature region of liquid hydrogen, boiling under different pressures. The apparatus used (somewhat changed for our purpose) has been described Comm. N<sup>o</sup>. 139*b*.

§ 2. *Method of calculation.* The axes of the three crystal disks were parallel to the „*b*”-axis, being the axis of symmetry of the crystal, perpendicular to the „*c*” (0.0.1) plane and perpendicular to the „*p*” (1.1.0) plane of the crystal respectively. When the susceptibilities in these directions are called *b*, *c* and *p*, respectively while *m* is the mean susceptibility, the main susceptibilities  $\chi_1, \chi_2, \chi_3$  ( $\chi_3$  coincides with the „*b*”-axis,  $\chi_1$  and  $\chi_2$  lie in the plane of symmetry) can be determined from the following equations:

$$\chi_3 = b \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (1)$$

$$\chi_1 \cos^2 \{\psi + (\beta - 90)\} + \chi_2 \sin^2 \{\psi + (\beta - 90)\} = c \quad . \quad . \quad . \quad . \quad (2)$$

$$\chi_3 \cos^2 \alpha + (\chi_1 \sin^2 \psi + \chi_2 \cos^2 \psi) \sin^2 \alpha = p \quad . \quad . \quad . \quad . \quad (3)$$

$$\chi_1 + \chi_2 + \chi_3 = 3m \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (4)$$

where  $\psi$  is the angle between  $\chi_1$  and the „*c*” axis of the crystal and  $\alpha$  the angle between the normal on the „*p*” plane ((1.1.0) plane) and the „*b*” axis, while  $\beta$  represents the angle between the „*a*” and „*c*” axes.

From these equations  $\chi_1, \chi_2, \chi_3$  and  $\psi$  can be solved.

§ 3. *Experimental results.* The following tables give the values of the susceptibilities as they have been observed.

Crystal disk „ <i>b</i> ”		Crystal disk „ <i>c</i> ”		Crystal disk „ <i>p</i> ”	
<i>T</i>	$\chi \cdot 10^6$	<i>T</i>	$\chi \cdot 10^6$	<i>T</i>	$\chi \cdot 10^6$
20 <sup>o</sup> .3 <sub>7</sub> K.	547	20 <sup>o</sup> .3 <sub>5</sub> K.	534	20 <sup>o</sup> .3 <sub>7</sub> K.	577
18.9 <sub>4</sub>	592	19.1 <sub>4</sub>	583	19.7 <sub>7</sub>	611
16.9 <sub>4</sub>	660	17.8 <sub>2</sub>	613	17.6 <sub>0</sub>	669
15.0 <sub>7</sub>	740	16.4 <sub>0</sub>	662	15.2 <sub>2</sub>	766
		15.0 <sub>0</sub>	738	14.8 <sub>5</sub>	791

In order to make the further calculation a set of values is necessary, which have been reduced to the same temperatures. Consequently graphs have been drawn of the values mentioned above of the  $\chi$ 's for the „*b*”, „*c*” and „*p*” disks as functions of the temperature. The values of the „*b*”, „*c*”, „*p*” are then graphically reduced to  $T = 20^{\circ}.35, 19^{\circ}.0, 17^{\circ}.0$  and  $15^{\circ}.0$  K.

These values follow here :

	Crystal disk			mean susceptibility of the powder „m”
	„b”	„c”	„p”	
T.K.	$\chi \cdot 10^6$			
20.3 <sub>5</sub> K.	548	534	578	542
19.0	589	572	620	580
17.0	662	639	690	649
15.0	748	723	780	735

Moreover the following data <sup>1)</sup> are still required :

$$a : b : c = 0.7360 : 1 : 0.4972$$

$$\beta = 107^{\circ}2'$$

$$p(1.1.0) : p(1.\bar{1}.0) = 70^{\circ}16'$$

The final results of the calculation are :

T	$\cos^2 \psi$		
20.3 <sub>5</sub> K.	0.8518	of	0.1471
19.0	0.7813	..	0.2176
17.0	0.7936	..	0.2063
15.0	0.8366	..	0.1663

T	$\chi_1 \cdot 10^6$	$\chi_2 \cdot 10^6$	$\chi_3 \cdot 10^6$
20.3 <sub>5</sub> K.	754	323	548
19.0	819	333	589
17.0	926	359	662
15.0	1038	420	748

The values in the third column (0.1471 etc.) have been accepted provisionally as the exact ones, as they come nearer to the values, which are known for the other members of the family of the monoclinic double

<sup>1)</sup> See GROTH, Chemische Kristallographie.

sulphates, to which manganese ammoniumsulphate belongs. No great accuracy may be expected however in the determination of  $\psi$ . The values of the principal susceptibilities will be accurate to about two percent. From these values we calculated the molecular susceptibilities, taking into account the diamagnetic property of the anion, the crystallization water and the ammoniumsulphate. The results are given in the following table. The values for  $\chi'_{3m}$  are those, which have been observed with the aid of the crystaldisk „b”.

$T$	$\chi'_{1m}$	$\chi'_{2m}$	$T$	$\chi'_{3m}$
20.3 <sub>5</sub> K.	0.295	0.127	20.3 <sub>7</sub> K.	0.214
19.0	0.321	0.130	18.9 <sub>4</sub>	0.232
17.0	0.362	0.140	16.9 <sub>4</sub>	0.258
15.0	0.406	0.165	15.0 <sub>7</sub>	0.290

§ 4. *Results.* The inverse values of the corrected molecules susceptibilities are plotted against the absolute temperature. In this way the variation of the different susceptibilities with  $T$  has been represented

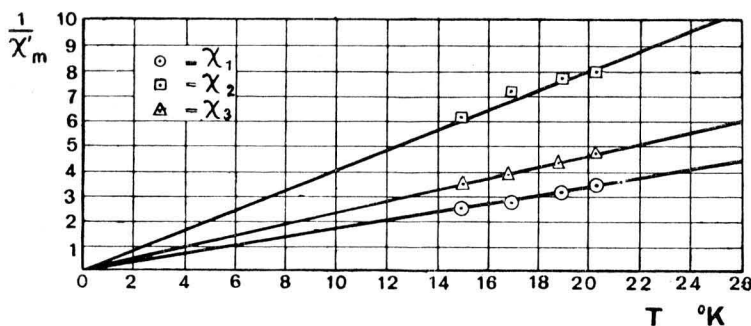


Fig. 1.

graphically. We see from Fig. 1 that the  $\frac{1}{\chi} \cdot T$  lines are neither coinciding, nor parallel. The values are lying as well as possible on three straight lines, which all pass through the origin.

This means that the principal susceptibilities follow the law of CURIE  $\psi \cdot T = C$ , but with different values for the  $C$ .

The mean susceptibility of the crystalpowder of manganese ammoniumsulphate obeys the law of CURIE, because each of the principal susceptibilities follows this law.