

The effective susceptibility of a paramagnetic powder

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The effective susceptibility (that is, the magnetic moment divided by the external magnetic field) of a paramagnetic powder loosely packed into an ellipsoidal container has been measured against the vapour pressure of liquid helium, and hence, using the results of the preceding paper, against the susceptibility of the compact salt. The definition of the 'Curie' temperature scale is based on the latter, and it is found that the correction to be applied in calculating the Curie temperature from the effective susceptibility of the powder agrees with that given by a theoretical expression derived from Breit's calculation of the demagnetizing field of a powder. This is of importance both in experiments using a loosely packed powder and in experiments using a paramagnetic powder mixed with a non-magnetic material.

The determination of the susceptibility of paramagnetic salts has become of exceptional importance in the temperature region below 1° K, and also in the liquid helium range itself, especially as the temperature scale is intimately connected with it. The correct procedure to be adopted for evaluating the susceptibility from measurements at these temperatures requires some consideration since, owing to the very high intensity of magnetization, the true field inside the sample (which enters into the definition of susceptibility) can differ considerably from the external field, i.e. the field present before the introduction of the sample. For an ellipsoidal sample of compact material this true field can be obtained simply by subtracting from the external field the demagnetizing field $N(M/V)$, N being the demagnetizing factor which can be calculated from the shape of the sample using Maxwell's formulae, and M/V the magnetic moment per unit volume. If, however, the sample is not homogeneous, e.g. a powdered material or mixture, the expression for the demagnetizing field will be more complicated and the theory has not yet been worked out for the general case.

The demagnetizing field has been calculated by Breit (1922) for a number of special cases, such as a space lattice of spheres or of spherical holes. The calculation showed that the average value of the field within the powder

particles (H_2) is less than the average field (H_1) within the sample as a whole by an amount $\frac{4\pi}{3} \frac{1-f}{f} \frac{M}{V}$ ‡, i.e.

$$H_2 = H_1 - \frac{4\pi}{3} \frac{1-f}{f} \frac{M}{V},$$

where f = filling factor = sample density/crystal density. Owing to the demagnetizing effect of the external shape we have

$$H_1 = H - N (M/V),$$

where H is the external field in the absence of the sample. Combining these equations we have

$$H_2 - H = -N (M/V) - \frac{4\pi}{3} \frac{1-f}{f} \frac{M}{V}. \quad (1)$$

The quantity on the right of this equation may be called the demagnetizing field of the powder.

The quantities in which we are interested are (a) the ratio of the intensity of magnetization M/V to the external field H ; this ratio will be called the 'effective susceptibility' κ , since it is the quantity usually measured; (b) the ratio of the intensity of magnetization to the field H_0 acting on the magnetic dipoles; this ratio, denoted by κ_0 , enters into the definition of the 'Curie scale' of temperature (see, for instance, Kurti and Simon 1938*a*). For it is generally assumed that Curie's law will hold if it is the quantity κ_0 that is taken to be inversely proportional to the absolute temperature rather than the susceptibility as defined in electromagnetic theory; and at temperatures where Curie's law is no longer true it is usual to define a new scale (the Curie or T^* scale) by extrapolation of Curie's law, i.e.

$$T^* = c/\kappa_0,$$

c being the Curie constant per c.c.

Now the fields H_2 and H_0 are connected by the well-known relation of Lorentz

$$H_0 = H_2 + \frac{4\pi}{3} \frac{M}{V}.$$

‡ This follows from equation (1') of Breit, which, written with the above notation, becomes

$$\frac{4\pi \left(\frac{M}{V} / H_1 \right)}{f 4\pi \left(\frac{M}{fV} / H_2 \right)} = \frac{1}{1 + \frac{1-f}{3} 4\pi \left(\frac{M}{fV} / H_2 \right)}.$$

This reduces to the above expression.

Here the factor $1/f$ enters because (M/V) is the average intensity of magnetization of the powder, and the Lorentz field naturally depends on the intensity of magnetization inside the crystalline particles. Hence from equation (1) we have

$$\begin{aligned} H &= H_0 + N \frac{M}{V} + \frac{4\pi}{3} \frac{1-f}{f} \frac{M}{V} - \frac{4\pi}{3} \frac{1}{f} \frac{M}{V} \\ &= H_0 - (4\pi/3 - N)(M/V), \end{aligned}$$

$$\text{i.e.} \quad \frac{1}{\kappa} = \frac{H}{\frac{1}{f} \frac{M}{V}} = \frac{1}{\kappa_0} - f \left(\frac{4\pi}{3} - N \right) = \frac{1}{c} (T^* - \Delta), \quad (2)$$

where

$$\Delta = cf(4\pi/3 - N).$$

Hence the 'effective susceptibility' κ is inversely proportional to $(T^* - \Delta)$.

In his monograph on *Magnetism and very low temperatures** Casimir has pointed out that the above calculation cannot be considered absolutely reliable in the general case, as for example with 'salt crystals crushed in a mortar and compressed to a density corresponding to $f = 0.6$ or $f = 0.7$ '. He considers it more advisable to use only very dense powders or solid material (since no doubt arises about the expression for Δ when $f = 1$) rather than to perform special experiments to test the above result. The difficulty cannot be avoided in this way, however, when a mixture of a paramagnetic salt with a non-paramagnetic salt is used; such mixtures are often necessary when it is desired to investigate the properties of substances not in themselves suitable for cooling by adiabatic demagnetization, at temperatures only obtainable by this method (Kurti and Simon 1935). Also, loose powders of paramagnetic salt have to be used in the capsule technique employed by various workers (Kurti, Rollin and Simon 1936; Kurti and Simon 1938*b*; Shire and Allen 1938).

The purpose of the experiment described in this paper is therefore to determine the value of Δ for a loosely packed salt, actually with a filling factor of about one-half. From equation (2) it will be seen that this involves the measurement of the effective susceptibility κ and of T^* , i.e. the true susceptibility. The former is measured by a mutual inductance method; the latter is determined from the vapour pressure of helium, using the results of the preceding paper.

* We are considerably indebted to this monograph for parts of the theoretical exposition above.

EXPERIMENTAL PROCEDURE

Since the value of Δ is a few hundredths of a degree, it is obvious that its determination involves the measurement of temperatures in the liquid helium range, and particularly near 1°K , to an accuracy of 0.1%. Such accuracy requires special apparatus such as that used for the measurement of the vapour pressure of liquid helium using the susceptibility of a compact paramagnetic salt as thermometer (see preceding paper). It is clear that once the vapour-pressure curve was established, the same apparatus could be used for the measurement of the effective susceptibility of a loosely packed powder.

The experimental procedure will only be briefly outlined, since full details are given in the preceding paper. Manganese ammonium sulphate was chosen as the paramagnetic salt since it has a high Curie constant, and the value of Δ is therefore comparatively high. It was roughly powdered in a mortar and loosely packed into a container whose shape was that of a 4:1 ellipsoid. This container was made of 'Perspex', turned as thin as possible and pierced by a number of small holes to ensure good thermal contact between the salt and the liquid helium bath in which it was immersed. The filling factor f was 0.55.

The temperature of the bath was determined from its vapour pressure using the 'scale 1937' down to 1.6°K and the more accurate 'scale 1939' below that point. The pressure was measured by a precision mercury manometer above 3 mm. Hg; lower pressures were measured by a special McLeod gauge, accurate to 1%, corresponding to 0.1% in temperature. The effective susceptibility was measured by a mutual inductance method, observations being made at the boiling-points of oxygen and hydrogen, and from the boiling-point of helium down to 1°K .

RESULTS

The effective susceptibility κ was measured by surrounding the sample by two coils, and measuring the deflexion δ of a ballistic galvanometer in the circuit of the secondary coil when a known current was reversed in the primary coil. The deflexion δ was reduced to the value δ_1 , corresponding to the reversal of unit current. The deflexion in the absence of the sample was not quite balanced out by an external mutual inductance, so that κ was not proportional to δ_1 but to $\delta_1 - a$, where a is a small constant. We have then

$$T^* - \Delta = k/(\delta_1 - a). \quad (3)$$

In the general case when this formula is used for the determination of T^* the value of Δ is known, and the constants k and a are obtained from two or more calibration points. In this experiment Δ is the unknown, and the simplest procedure is to write (3) in the form

$$T^*(\delta_1 - a) = \Delta(\delta_1 - a) + k. \quad (4)$$

This is of the form $y = \Delta x + k$, and hence on plotting $y = T^*(\delta_1 - a)$ against $x = (\delta_1 - a)$, a straight line should be obtained whose slope gives the correct value of Δ . As it is not possible to determine the value of a directly by removing the sample, equation (4) can only be used to determine Δ if the value of a obtained by calculation from two observed points is independent of Δ . To the accuracy required, which is the same (0.01 cm.) as the accuracy of measurement of δ_1 , this is so, for the value of a can be obtained from the values of δ_1 observed at the boiling temperatures of oxygen and hydrogen. Compared with these temperatures Δ is very small and can be neglected.

The values of δ_1 and T at the boiling-points of oxygen and hydrogen are

$$\begin{aligned} \delta_1 &= 0.09 & T &= 90.1^\circ, \\ \delta_1 &= 1.40 & T &= 20.3^\circ. \end{aligned}$$

Substituting these values in equation (3) the value of a is obtained as -0.29 .

The results obtained in the liquid helium range are given in table 1. In the first two columns the values of δ_1 and the vapour pressure are given, the latter being the measured pressure corrected for the thermolecular pressure difference and the creeping film effect as described in the preceding paper. These corrections are negligible except at the last two points.

The value of T^* is not appreciably different from T except at the lowest temperatures, and the value of $T^* - T$ is obtained from the preceding paper.

TABLE 1

δ_1 cm.	vapour pressure mm.	T ° K	T^*	$x = (\delta_1 - a)$	$y = T^*(\delta_1 - a)$
7.99	754	4.209	4.209	8.28	34.85
10.62	245.3	3.216	3.216	10.91	35.08
13.72	78.3	2.510	2.510	14.01	35.17
16.33	33.1	2.125	2.125	16.62	35.32
19.83	10.36	1.753	1.755	20.12	35.31
23.78	3.07	1.470	1.473	24.07	35.46
27.95	0.92	1.258	1.261	28.24	35.61
30.37	0.47 ₃	1.160	1.164	30.66	35.69
35.24	0.130	1.008	1.012	35.53	35.96
35.33	0.129	1.007	1.011	35.62	36.01

There the vapour pressure of helium was compared with the true susceptibility of a compact sample of manganese ammonium sulphate, so that by using those results the true susceptibility of our loosely packed sample is obtained, indirectly, from that of the compact sample. Thus the effect of the small deviation of the salt from Curie's law is eliminated.

Using for a the value -0.29 obtained above, the values of $x = \delta_1 - a$, and of $y = T^*(\delta_1 - a)$ are calculated and shown in figure 1, and from the slope of the straight line drawn through the points the value of Δ is obtained as

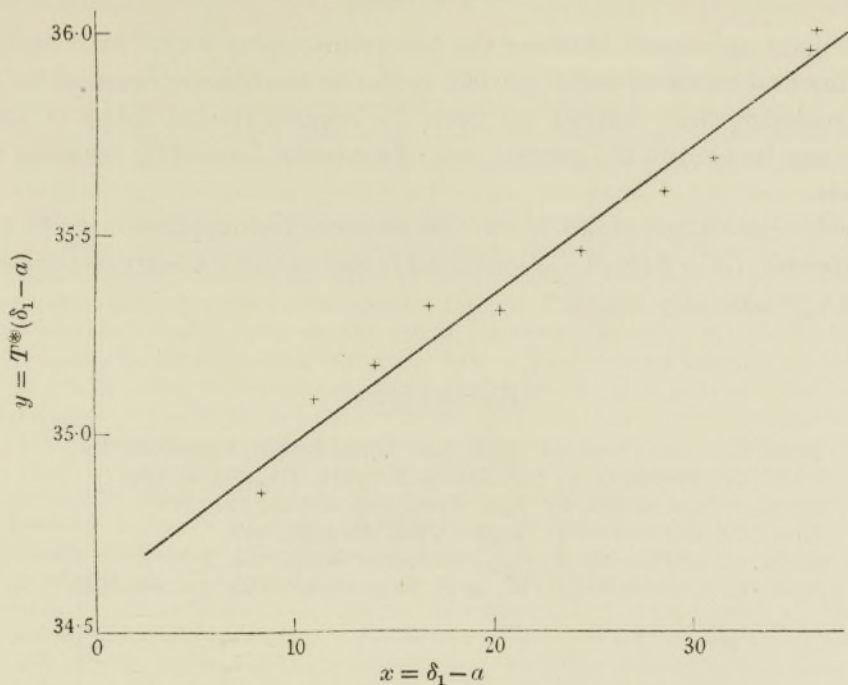


FIGURE 1

0.038° . The values of T^* and of δ_1 should each be accurate to 0.1% , and the error in the value of y should not therefore be more than 0.2% . It will be seen that the maximum deviations of the experimental points from the straight line are of this order. As the range of values of x is about 30, it follows that the accuracy of the determination of Δ , the slope of the straight line, is about $\pm 0.002 \times 35/30 = \pm 0.002^\circ$.

This experimental value of Δ has now to be compared with that given by the theoretical expression $\Delta = fc(4\pi/3 - N)$

where $f = 0.55$, $c = 2.00 \times 10^{-2}$, $N = 0.95$ for a 4 : 1 ellipsoid with major axis parallel to the field.

Inserting these figures, we have $\Delta = 0.037^\circ$. This value is in agreement with the experimental value of $0.038 \pm 0.002^\circ$ and differs considerably from the value 0.066° for a compact sample. It should be noted that at the lowest temperatures which can be attained by adiabatic demagnetization of manganese ammonium sulphate (about 0.1°), the use of the incorrect value of Δ would introduce an error of about 30 % in the temperature.

CONCLUSION

The good agreement between the theoretical value 0.037° of Δ and the experimental value of $0.038^\circ \pm 0.002$ indicates that the expression for the demagnetizing field derived by Breit for certain special forms of space lattice can be used in the general case of a powder formed by crushing salt crystals.

For the calculation of the 'Curie' temperature the experiment shows that the expression $\Delta = fc(4\pi/3 - N)$ is certainly still valid for loosely packed salts with an f -value of one-half.

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