Some Magnetization Studies of Cr^{+++}, Fe^{+++}, Gd^{+++}, and Cu^{++} at Low Temperatures and in Strong Magnetic Fields

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WE have undertaken a study of magnetic properties of paramagnetic coolants at well-known temperatures in as low a range as possible. First, we studied moderately "well-behaved" paramagnetic substances in the liquid helium range. As is shown in Fig. 1, these substances, containing Cr^{+++} ($^4F_{3/2}$ state for the free ion, but quenched to a free spin system by the crystalline electric field), Fe^{+++} ($^6S_{5/2}$ state for free ion), and Gd^{+++} ($^8S_{7/2}$ state for the free ion), all follow free spin Brillouin functions (except for almost negligible deviations) up to over 99.5 percent saturation[1] for 50 000 gauss and 1.30°K.

In order to extend our knowledge of the possible causes and consequences of small departures which may become important in some paramagnetic coolants at extremely low temperatures, we carried out similar magnetization experiments on copper sulfate pentahydrate[2] in which the departure from a Brillouin function was very pronounced in the helium range. That is, a plot of magnetic moment against H/T gives magnetic isotherms which do not superimpose. In Fig. 2 we note that this dispersion is in qualitative agreement with calculated moment isentropes of Geballe[3] since isothermal and adiabatic moments should approach each other in the limit of zero field. It has been suggested by Geballe that this anomalous behavior may be associated with antiferromagnetism.[4] Since it is inconvenient to work under the Néel (antiferromagnetic) temperature for $CuSO_4 \cdot 5H_2O$ which is considerably under 1°K, we have also started a study of $CuCl_2 \cdot 2H_2O$ for sub-Néel magnetization studies as its Néel temperature[5] is ~4.3°K. We used the same technique for copper chloride dihydrate as for the other salts. This consisted in moving a spherical sample (a powder in this case) from the center of one coil to the center of a duplicate, oppositely wound coil. The deflection of a ballistic galvanometer in series with the coil system was proportional to the moment of the sample. The experimental results which we obtained indicated constant moments for fixed fields in the sub-Néel range studied as is indicated by sample values in Fig. 3 and symbolized by

$$(\partial M(H)/\partial T)_H = 0 \qquad (1)$$

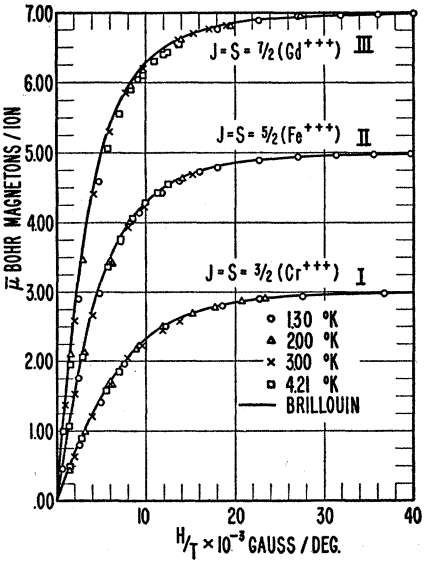

FIG. 1. Plot of magnetic moment *vs* H/T for (I) potassium chromium alum, (II) ferric ammonium alum, and (III) gadolinium sulphate octahydrate.

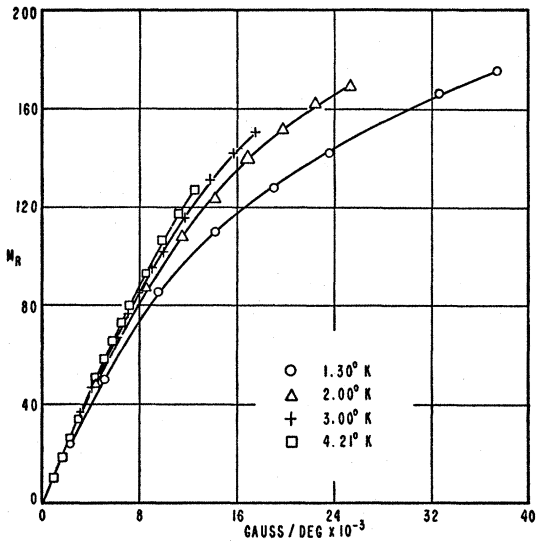

FIG. 2. Plot of relative moment *vs* H/T for copper sulfate pentahydrate.

[1] W. E. Henry, Phys. Rev. **85**, 487 (1952); Phys. Rev. **88**, 559 (1952).
[2] W. E. Henry, Phys. Rev. **87**, 1133 (1952).
[3] T. H. Geballe and W. Giauque, J. Am. Chem. Soc. **74**, 3513 (1952).
[4] L. Néel, Ann. Physik **X 18**, 5 (1932); **XI 5**, 232 (1936); J. H. Van Vleck, J. Chem. Phys. **9**, 85 (1941).
[5] C. J. Gorter and J. Haantjes, Physica **18**, 285 (1952).

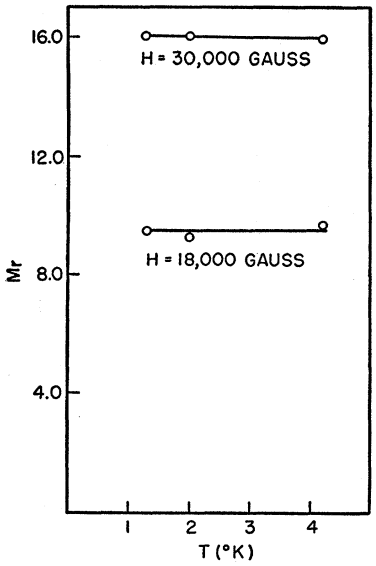

FIG. 3. Plot of relative moment *vs* T for two values of magnetic field for copper chloride dihydrate.

in the moderately high field range at liquid helium temperatures. Our experimental results may be compared qualitatively with announced[6] results of van den Handel, Poulis, and Gijsman[7] in the moderately high field range. According to our results, if one now plots M against H/T (see Fig. 4) as was done for the very closely paramagnetic salts, one obtains, for a given H/T, a moment which decreases linearly with temperature. If the assumption is made that the transition from paramagnetic to antiferromagnetic behavior is not necessarily abrupt, but perhaps gradual for some structures, then for these structures one may expect a departure from a Brillouin function even for a sample considerably above the Néel temperature (see Fig. 2). In the linear range (no saturation effects), the magnetic moment may be represented by

$$M=F(T)H, \tag{2}$$

where $F(T)$ goes from a T^{-1} dependence in the paramagnetic case to a constant in the antiferromagnetic case. $F(T)$ depends, at least in part, on the distance above the Néel temperature and the strength of the forces contributing to the manifestations of antiferromagnetism, as was shown by Van Vleck.[4] Further study on this dependence would be helpful, for, knowing $F(T)$, one could better attack the problems of magnetic and thermodynamic behavior of paramagnetic coolants at extremely low temperatures.

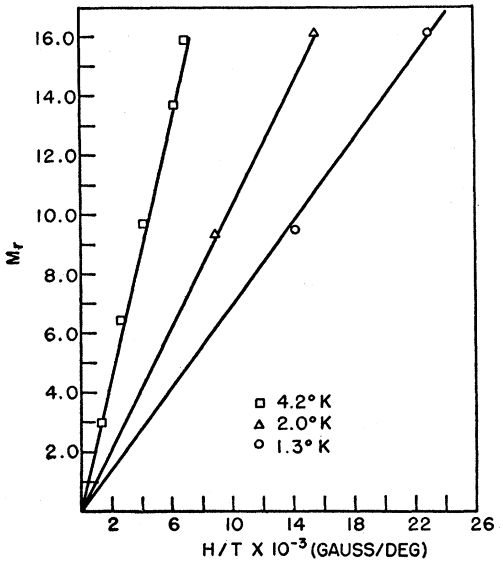

FIG. 4. Plot of relative moment *vs* H/T for copper chloride dihydrate.

[6] N. J. Poulis and G. E. G. Hardeman, Physica **18**, 201 (1952).
[7] van den Handel, Poulis, and Gijsman, Physica (to be published).