# LOW-TEMPERATURE MAGNETIC PROPERTIES OF $DyPO_4$: AN IDEAL THREE-DIMENSIONAL ISING ANTIFERROMAGNET

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We have measured the magnetic susceptibility and heat capacity of $DyPO_4$ at low temperatures and compared our results with the exact series expansions based on the three-dimensional Ising model for the diamond lattice. There is excellent agreement between experiment and theory.

Recently it has been shown[1,2] that $DyPO_4$ orders antiferromagnetically at approximately 3.5 K. The optical data indicate that $DyPO_4$ is an ideal three-dimensional Ising system with very small net interaction with neighbors other than nearest neighbors. $DyPO_4$, which has the zircon structure[3] in which the magnetic ions lie on a slightly distorted diamond lattice, is a particularly appropriate material with which to test the theoretical results based on the Ising model because this three-dimensional lattice, to our knowledge, is the only one for which the exact low-temperature series expansions are convergent up to the critical point. In particular, the exact low-temperature series expansions for the energy, heat capacity, and reduced magnetic susceptibility have been obtained.[4,5] In this Letter we report the results of heat-capacity and magnetic-susceptibility measurements of $DyPO_4$ and compare them with those obtained from the exact expansions.

The heat-capacity and the magnetic-susceptibility data are shown in Fig. 1. The Néel temperature determined by the heat-capacity peak is $3.391 \pm 0.001$ K. The susceptibility results indicate that $T_N = 3.40 \pm 0.01$ K, which, within the experimental error, is in agreement with that obtained from the heat capacity.

Essam and Sykes[4] have been able to estimate the energy and entropy within narrow limits and they obtain for the energy and the entropy at the critical point the values $E_c/kT_c = 0.418$ and $S_c/k = 0.511$. The corresponding experimental values, 0.408 and 0.505, respectively, which were obtained from the heat-capacity results, are in excellent agreement with these estimates.

The interaction constant, exchange plus dipolar, can be obtained from the low-temperature asymptotic form for the heat capacity,

$$C/R = (2qJ/kT)^2 \exp(-2qJ/kT).$$

Here $J$ is the interaction constant defined by the spin Hamiltonian

$$\mathcal{H} = \sum_{i>j} \frac{J}{S^2} S_{iz} S_{jz} + \beta \vec{S} \cdot g \cdot \vec{B}$$

for effective spin $S = \frac{1}{2}$ and where the summation is over nearest neighbors only. $g_\parallel = 19.4$ and $g_\perp = 0.51$. When $\log(CT^2/R)$ is plotted versus $1/T$, then a given value of $J$ uniquely determines both the slope and the intercept. The dot-dash line in

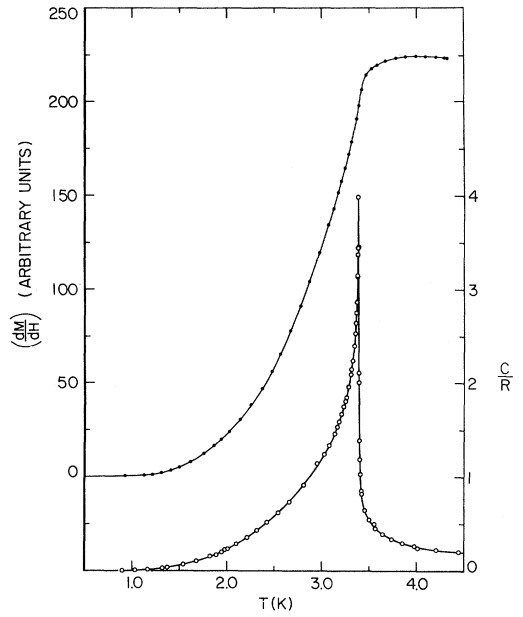


FIG. 1. The heat capacity $C/R$ of $DyPO_4$, in its ordering region, is shown as a function of temperature by the lower curve. The open circles represent the data and the solid line is a smooth line through the data. The magnetic susceptibility $(\partial M/\partial H)$ of $DyPO_4$, as measured along the symmetry axis, is shown for the ordering region by the upper curve. The closed circles represent the data and the solid line is a smooth line through the data.

Fig. 2(a) represents the asymptotic form of the heat capacity fitted to the data, and the value of $J/k$ determined from it is $1.25 \pm 0.05$ K. Using the relation $kT_c/qJ = 0.6760$, as given by Essam and Sykes for the diamond lattice, with $T_c = 3.391$ K and $q = 4$, the calculated value of $J/k$ is 1.254 K, identical with that determined from the heat capacity in the low-temperature limit. This value is in excellent agreement with the value measured directly spectroscopically[1] ($J/k = 1.29 \pm 0.03$ K). One can now put this value of $J/k$ into the low-temperature series expansion of Essam and Sykes and compare the fit of the theory with the data. This comparison for the heat capacity is shown in Fig. 2(a) where the circles represent the experimental data and the solid curve is that calculated from theory. At this point perhaps we should point out that once $T_N$, or $J$, has been selected there are no adjustable parameters in their theory. If there is a nuclear hyperfine interaction present, then the form of the heat capacity at low temperature is

$$\frac{C}{R} = \left(\frac{2qJ}{kT}\right)^2 \exp\left[-\frac{2qJ}{kT}\right] + \frac{a}{T^2},$$

rather than the simpler form given above. One can see from Fig. 2(a) that the measured $CT^2/R$ is independent of temperature below approximately 0.6 K, in agreement with this prediction. The dashed line gives the value of this hyperfine contribution as $3.3 \times 10^{-4}$ K$^2$.

Sykes, Essam, and Gaunt[5] have derived the exact low-temperature series expansion for the reduced susceptibility for the three-dimensional Ising antiferromagnet assuming the diamond lattice. Their series expansion for $\chi T$ is represented by the dashed line in Fig. 2(b) where the circles represent the data. The fit is quite poor. Fisher[6] has shown that the magnetic susceptibility and heat capacity are related by the expression $C \simeq A[\partial(\chi T)/\partial T]$. If this relation is valid, then $E \propto \chi T$ and one can compare the fit of the measured $\chi T$ curve with the low-temperature series expansion for the energy derived by Essam and Sykes. This comparison is shown in Fig. 2(b) where the solid line is that calculated from the series expansion for the energy. We should point out that the Sykes, Essam, and Gaunt expression for $\chi T$ is different from that given by Essam and Sykes for the energy for the same lattice and it can be seen that although the slopes of the two curves are the same in the low-temperature limit, the expansion for the energy fits the data much better than does the expression for the reduced susceptibility given by Sykes, Essam, and Gaunt.[5] The expressions for the energy and reduced susceptibility do give the same low-temperature

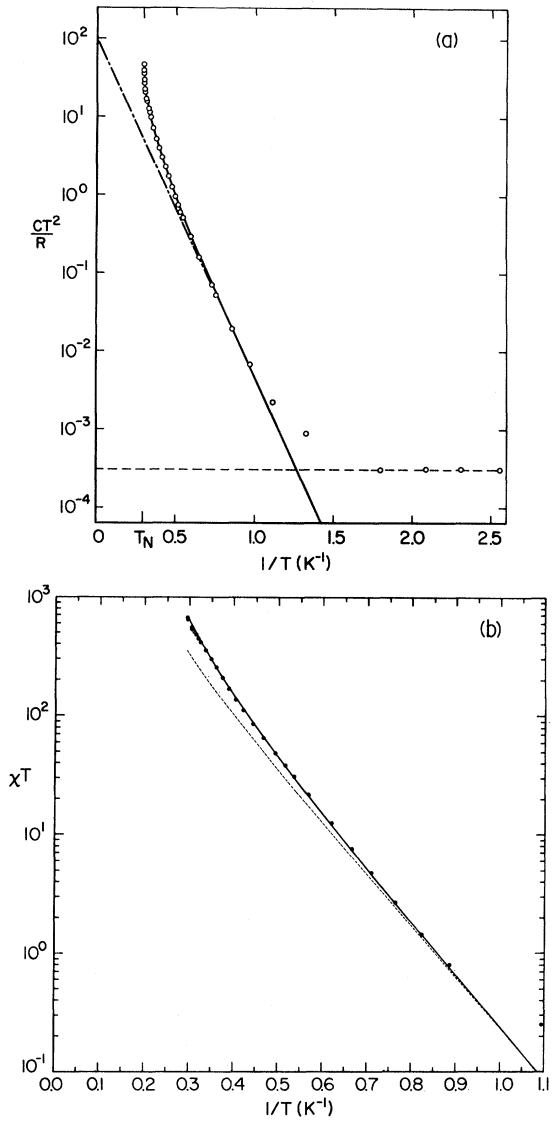


FIG. 2. (a) $\mathrm{Log}_{10}\, CT^2/R$ vs $1/T$ for $DyPO_4$ where the circles represent the data. The dot-dash line gives the low-temperature limiting behavior in the absence of hyperfine interactions. This line was drawn tangent to the low-temperature data such that its intercept and slope determined $J$ uniquely. The dashed line gives the hyperfine contribution to the heat capacity. The solid line is that calculated from the exact series expansion for the heat capacity as given by Essam and Sykes. (b) The logarithm of the reduced magnetic susceptibility versus $1/T$. The solid circles represent the data. The solid line through the data is that calculated from the exact series expansion for the energy as given by Essam and Sykes. The dashed line was calculated from the exact series expansion for $\chi T$ as given by Sykes, Essam, and Gaunt.

limiting slope, however, and the experimentally measured value of $J/k$, determined from the limiting slope, is 1.25 K. This is the same value as that which was determined from heat-capacity results as well as from spectroscopic measurements.

Critical behavior has been a very active field of theoretical interest in the recent past[7] and the nature of the divergence at the critical point still appears to be an unsettled question. In the critical region for the diamond lattice, Essam and Sykes predict that the heat capacity has a logarithmic singularity below the critical point, but in the region above the critical point the divergence appears to be sharper than logarithmic. We show in Fig. 3 our heat-capacity data for the critical region for both $T<T_N$ and $T>T_N$. It can be seen that the straight line through the data for $T<T_N$ over the region $10^{-1}$ to $8\times10^{-4}$ in $(T_N-T)/T_N\equiv t$ is in excellent agreement with the prediction of Essam and Sykes. Recently Gaunt and Domb[8] have predicted that the critical region exists only for $t<10^{-4}$ and that the heat capacity should diverge as $t^{-1/8}$ rather than logarithmically. Their theoretical values join on to our data perfectly at the low values of $t$. Gaunt and Domb, furthermore, have a second term in the heat capacity proportional to $t^{+7/8}$ and, as stated by them, the combination of this term and the $t^{-1/8}$ term gives a curve which appears to be logarithmic in the region $10^{-1}>t>10^{-4}$. Since our data extend down to only $8\times10^{-4}$ in $t$, we are unable to distinguish experimentally between the logarithmic dependence predicted by Essam and Sykes and the power-law dependence predicted by Gaunt and Domb because both predict a logarithmic dependence in the region of our data. The dashed curve through the data for $T>T_N$ is given by $C/R=0.155t^{-0.35}$. Others[9] have observed a similar dependence on $t$ in other materials for $T>T_N$. The solid curve through the data for $T>T_N$ was drawn such that slope$(T<T_N)=2.5\times$slope$(T>T_N)$ as calculated by Essam and Sykes for the case in which the heat capacity has a logarithmic singularity both above and below $T_N$.

In conclusion, $DyPO_4$ appears to be an ideal three-dimensional Ising antiferromagnet which fits the theory for the three-dimensional Ising model as far as has been tested. The fact that the $Dy^{3+}$ ions in $DyPO_4$ lie almost on the diamond lattice is particularly significant, because for the diamond lattice the low-temperature series expansions are convergent right up to the critical point and theory and experiment can be compared throughout the entire region.

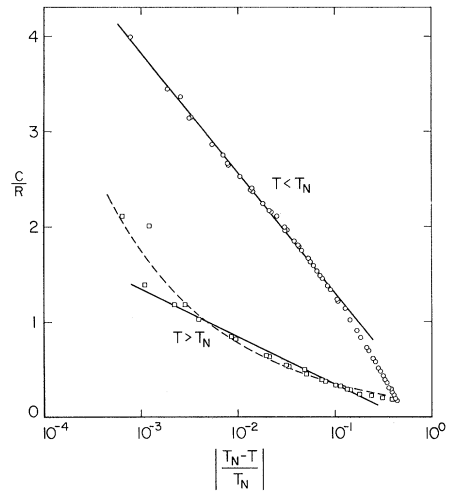


FIG. 3. The comparison of theory and experiment for the heat capacity plotted as a function of $\log_{10}t$ where $t\equiv|(T_N-T)/T_N|$. The open circles represent the data for $T<T_N$ and the squares represent the data for $T>T_N$. The solid line through the data $T<T_N$ shows the logarithmic dependence of the heat capacity in the region $10^{-1}>t>8\times10^{-4}$. The solid line through the data for $T>T_N$ was drawn such that slope$(T<T_N)=2.5\times$slope$(T>T_N)$. The dashed curve gives the power-law dependence.

Further work on $DyPO_4$ is in progress and will be reported in due course.

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[1]J. C. Wright and H. W. Moos, Phys. Letters 29A, 495 (1969).

[2]G. T. Rado, Phys. Rev. Letters 23, 644 (1969).

[3]R. W. G. Wyckoff, Crystal Structures (Interscience Publishers Inc., New York, 1965), 2nd ed., Chap. VIII.

[4]J. W. Essam and M. F. Sykes, Physica 29, 378 (1963).

[5]M. F. Sykes, J. W. Essam, and D. S. Gaunt, J. Math. Phys. 6, 283 (1965).

[6]M. E. Fisher, Phil. Mag. 7, 1731 (1962).

[7]See for example, the review by M. E. Fisher, Rept. Progr. Phys. 30, 615 (1967).

[8]D. S. Gaunt and C. Domb, J. Phys. C: Phys. Soc. (London) Proc. 1, 1038 (1968).

[9]G. S. Dixon and J. E. Rives, Phys. Rev. 177, 871 (1969); J. W. Philp, R. Gonano, and E. D. Adams, J. Appl. Phys. 40, 1275 (1969); B. E. Keen, D. P. Landau, and W. P. Wolf, J. Appl. Phys. 38, 967 (1967).