Experimental Study of the Tricritical "Wings" in Dysprosium Aluminum Garnet

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Light-scattering techniques have been used to study the behavior of the antiferromagnet dysprosium aluminum garnet near its tricritical "wings." The crossover from tricritical to critical behavior near the wing critical lines has been observed, and the shape of the wing critical lines has been determined. The results are in good agreement with the present theory.

A symmetric tricritical point is a point in a space of three thermodynamic fields at which three lines of critical points intersect.[1] Associated with the critical lines are three first-order or coexistence surfaces, two of which are commonly referred to as "wings" because of the symmetric appearance of the phase diagram. In simple antiferromagnets the three fields are the temperature, T, the uniform internal magnetic field, $\vec{H}_i$, and the staggered magnetic field, $\vec{H}_s$. The latter is a field which couples to (i.e., is the thermodynamic conjugate to) the staggered magnetization. At present, it is not possible to apply a staggered field in the laboratory, and it is therefore not possible to observe the wings in the phase diagram. For other systems with symmetric tricritical points the three fields are different, but in nearly all cases, the field analogous to the staggered field in simple antiferromagnets is also not accessible in the laboratory.[2] One notable exception occurs in certain ferroelectrics, for which the appropriate fields are temperature, pressure, and electric field; and the first experiments on such a system have been reported recently.[3] However, the experiments are difficult, and aside from some preliminary results for one tricritical exponent, it has not yet been possible to test any of the quantitative predictions of the theory. In this Letter we describe a study of a different type of system in which the analog of a staggered field is readily accessible, and we present the first quantitative measurements of a phase diagram as a function of this field.

The system we have studied is the Ising-like antiferromagnet dysprosium aluminum garnet (DAlG).[4] Although, as noted above, it is not at present possible to apply a real staggered field, it has recently been shown[5] that in DAlG it is possible to apply what has been termed an induced staggered field, "$\vec{H}_s$." The effect of "$\vec{H}_s$" is completely analogous to the effect of a real staggered field, in that for "H_s" $=0$, one finds in the H_i-T plane a line of first-order transitions (i.e., a first-order line) which ends at a tricritical point, while for "H_s" $\neq 0$ one finds a first-order line which ends at a critical point. Using symmetry arguments, it has been shown that to lowest order[5,6] "$\vec{H}_s$" $\sim H_x H_y H_z$, where H_x is the component of $\vec{H}_i$ along the [100] crystal axis, etc. Thus, by simply varying the direction of $\vec{H}_i$ relative to the crystal axes, one can vary "H_s," and it is therefore possible in DAlG to explore the staggered dimension of the phase diagram.

We have used light-scattering techniques to measure the H_0-T phase diagram of DAlG as a function of "H_s," where H_0 is the applied or external magnetic field. Demagnetizing effects cause a first-order line in the H_i-T plane to be split open into two lines in the H_0-T plane, and these two phase boundaries enclose a mixed phase region in which domains of the antiferromagnetic and paramagnetic phases coexist.[7] The domains scatter light strongly while the homogeneous phases do not, and thus light-scattering measurements can be used to locate the phase boundaries.[8] The extent of the mixed phase region at constant temperature, ΔH_0, is proportional to the difference of the magnetizations of the two coexisting phases, ΔM.[7] Data for ΔH_0 can therefore be used to determine the exponents associated with ΔM.

Our experiments were performed on a platelet of DAlG with the normal to the plate parallel to a [110] direction. The field was directed approximately normal to the plate, and the angle between $\vec{H}_0$ and the [100] axis, θ, was varied by rotating the crystal about the [1$\bar{1}$2] axis. Since "H_s" $\sim H_x H_y H_z$, its magnitude is proportional to θ for small angles, and in the following we will use θ as a measure of "H_s."[9,10]

Some typical results for the H_0-T phase diagram are shown in Fig. 1. $\theta=0$ corresponds to "H_s" $=0$ ($\vec{H}_0 \| [110]$) and thus the point where the two first-

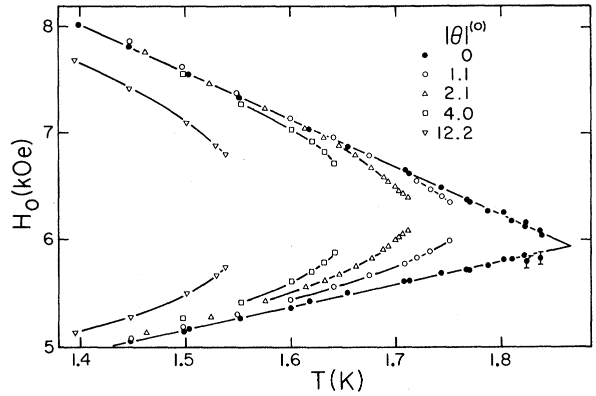

FIG. 1. H_0-T phase diagram for DAlG for several different values of θ.

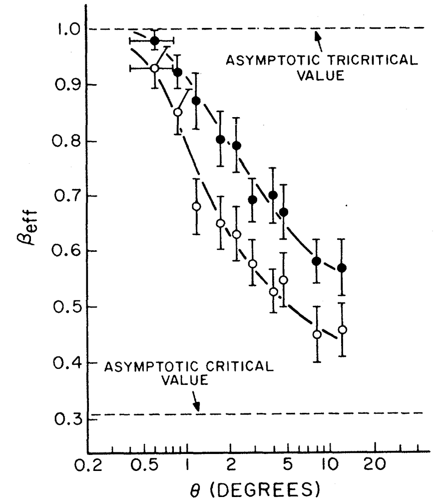

FIG. 2. β_{eff} as a function of θ for $t=0.1$ (closed symbols) and $t=0.03$ (open symbols).

order lines meet is the tricritical point. The theory[11] predicts for this case that

$$\Delta M \sim \Delta H_0 \sim (T_t - T)^{\beta_u}, \qquad (1)$$

where T_t is the tricritical temperature, and the tricritical exponent[12] $\beta_u = 1$. Thus, ΔH_0 should go to zero linearly with the temperature, and it can be seen that the results are in good agreement with this prediction. A least-squares fit to the data gives $\beta_u = 0.97 \pm 0.04$.

For $\theta \neq 0$, the point at which the first-order lines meet is a critical point which is often referred to as a wing critical point. The theory[13] predicts that each wing critical point is Ising-like. Thus, for $\theta \neq 0$, ΔM, and hence ΔH_0, is predicted to behave as

$$\Delta M \sim \Delta H_0 \sim (T_c - T)^{\beta}, \qquad (2)$$

where T_c is the critical temperature and $\beta \simeq 0.31$.[14] The predicted value of β is very different from the value of β_u, and it is interesting to consider the manner in which the exponent "crosses over" from one type of behavior to the other. The theory[15] predicts that the exponent changes discontinuously as one moves from the critical line to the tricritical point. However, it also predicts that the range over which the critical behavior is observed will be a function of location along the critical line. Far from the tricritical point (i.e., for θ large) the asymptotic critical region will be relatively large. As the tricritical point is approached (i.e., as $\theta \to 0$) the critical region will shrink, and outside this region the behavior will be tricriticallike. The size of the critical region shrinks to zero at the tricritical point, leaving behind an asymptotic tricritical region. From Fig. 1 we can see that our results are in at least qualitative accord with these predictions. At low temperatures, far from the wing critical line, the results for $\theta \neq 0$ approach the $\theta = 0$ results quite closely. As the wing critical line is approached, i.e., as θ increases at fixed T, or as T increases at fixed θ, the curvature of the phase boundaries increases, as expected from Eq. (2).

We have performed a quantitative analysis of the crossover behavior using the concept of an effective exponent,[15] β_{eff}. For our purposes,[10] it is convenient to use a definition for β_{eff} which differs slightly from the usual one.[15] We define $\beta_{eff}(T_0)$ to be the value derived from fitting data for ΔH_0 from an infinitely small range of temperature about the temperature T_0 to the power law

$$\Delta H_0 = A(T_c - T)^{\beta_{eff}(T)}, \qquad (3)$$

with the parameters A, T_c, and $\beta_{eff}(T_0)$ all allowed to vary in order to obtain the best fit. We have used our data to estimate β_{eff} as a function of T_0 and θ by performing a large number of fits to Eq. (3), in which the (necessarily finite) range of temperature used in the fit, T_0, and θ were varied.

The results for β_{eff} as a function of θ at constant reduced temperature, $t \equiv (T_c - T_0)/T_c$, are shown in Fig. 2. Here the value of T_c was obtained from the fit closest to the critical point. From Fig. 2 we see that there is a monotonic variation of β_{eff}, as it crosses over from its tricritical value at small angles, to a value at large angles which is at least qualitatively consistent with the asymptotic critical-line value predicted by the theory. Unfortunately we were unable to approach the critical line close enough to observe the asymptotic critical behavior. Also, we did not attempt to follow the crossover to larger angles since there are complications arising from the presence of another tricritical point which occurs at 50.8°, corresponding to a [021] direction. However, we note that, in a separate experiment with $\vec{H}_0 \| [111]$,[10,16] it was possible to penetrate into the asymptotic critical region, and the exponent β was found to be 0.30 ± 0.02, in good agreement with the theory.

There are at present no quantitative predictions with which to compare the results in Fig. 2. Previous theoretical work[15,17] has considered the crossover behavior of different exponents and, as far as we know, there is no reason to expect that the crossover behavior will be the same for different exponents. The general behavior which we have found does not appear to be unreasonable, but further work on this problem is clearly needed.

We have also used our data to determine the shape of the wing critical lines. This was accomplished by extrapolating phase diagrams like those shown in Fig. 1 to estimate T_c. First, an upper bound on T_c was determined by fitting the ΔH_0-T data near the critical point to Eq. (3) with A, T_c, and β_{eff} all allowed to vary. A lower bound on T_c was then obtained by fitting ΔH_0-T data near the critical point to Eq. (3) with β_{eff} held fixed at the theoretically predicted critical value which is 0.31. This extrapolation procedure should bracket the true value of T_c provided only that β_{eff} varies monotonically from its asymptotic tricritical value to its asymptotic critical value. Although we cannot expect a monotonic variation *a priori*, the results in Fig. 2 are, in fact, completely consistent with this behavior. The results for T_c as a function of θ are shown in Fig. 3. We note that since [110] is an axis with twofold symmetry, these results should be independent of the sign of θ, which is indeed the case. From Fig. 3 we see that the critical line appears to have a cusp at $\theta = 0$, as predicted by the theory. The theory also makes the quantitative prediction that T_c should vary as

$$T_c - T_t \sim H_s^{1/\varphi\Delta_t} \sim |\theta|^{1/\varphi\Delta_t}, \qquad (4)$$

where φ and Δ_t are the usual tricritical exponents.[12] The theory[11] predicts $\varphi = 2$ and $\Delta_t = \frac{5}{4}$, which yields $T_c - T_t \sim |\theta|^{2/5}$. From Fig. 3 we see that the data are quite consistent with this prediction. A least-squares fit to the power law in Eq. (4) gives $1/\varphi\Delta_t = 0.41 \pm 0.03$.

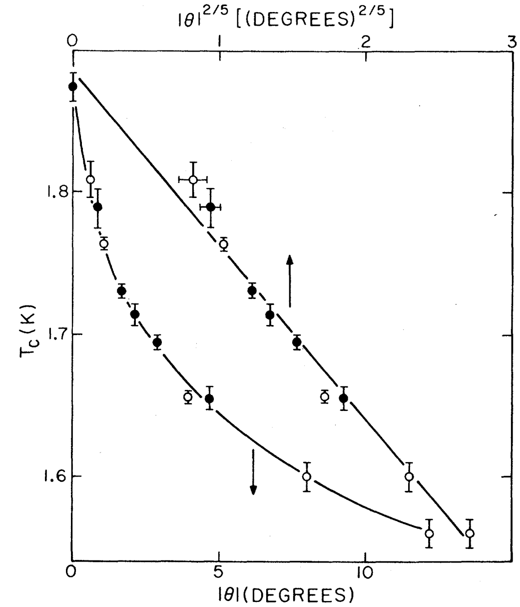

FIG. 3. T_c as a function of $|\theta|$ (lower scale) and $|\theta|^{2/5}$ (upper scale). Open symbols, measurements made with $\theta > 0$; close symbols, $\theta < 0$.

In summary, we have presented the first quantitative results for the shape of the tricritical wings. Our results should provide a useful test of future theories of the crossover behavior near the wing critical line.

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[9]In principle, one should really distinguish between the internal field $\vec{H}_i$ and the applied field $\vec{H}_0$, which are related by $\vec{H}_i = \vec{H}_0 - \overleftrightarrow{N}\cdot\vec{M}$, where $\overleftrightarrow{N}$ is the demagnetizing tensor. For the particular geometry used in this experiment, $N_\parallel \approx 4\pi$ and $N_\perp \approx 0$, where $\parallel$ and $\perp$ denote directions parallel and perpendicular to [110]. For small θ, the component of field perpendicular to [110] is therefore approximately the same for both $\vec{H}_i$ and $\vec{H}_0$ and is given by $H_0 \sin\theta \approx H_0\theta$, while the corresponding parallel components, though different, remain independent of θ to first order. The product $H_x H_y H_z$ is thus proportional to θ referred to either internal or external fields. In practice, it is clearly more convenient to use the angle of the applied field as a measure of the induced staggered field. A further discussion of this point is given in N. Giordano, thesis, Yale University, 1977 (unpublished).

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Theory of Interlayer Exchange for Adsorbed ^{3}He Monolayers

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An explanation of experimental NMR relaxation times, T_2, for ^{3}He adsorbed on Grafoil at coverages just greater than one monolayer is given in terms of the dipolar interaction of first-layer particles modulated by quantum exchange of particles between first and second layers. It is found that a Heisenberg-Hamiltonian description fails and one must use an exchange *operator* formulation which permits transitions among second-layer $\vec{k}$ states during the exchange process.

Helium adsorbed on Grafoil represents a quasi-two-dimensional system which has a number of interesting phases,[1] e.g., gas, liquid, solid, and registered solid. Recently, experimental NMR results[2] (Fig. 1) have shown evidence for three distinct phases at $T = 1$ K: (a) a fluid phase for coverage $x \lesssim 0.7$; (b) a two-dimensional solid for $0.7 \lesssim x \lesssim 1.0$, and (c) a phase for $x \gtrsim 1$ for which T_2 increases linearly with coverage. Theoretical calculations[2] for region (b) showed that the T_2 data could be explained in terms of a dipolar linewidth motionally narrowed by quantum tunneling. Region (c) of the T_2 data was tentatively identified as being caused by the formation of the second layer of helium atoms with motion via exchange with the first layer. Evidence for a registered phase was also reported recently.[3]

In this Letter I report calculations which justify the explanation of region (c) in terms of interlayer exchange. The novel feature of this explanation is the discovery that a calculation based on a Heisenberg-exchange Hamiltonian proves inadequate and one needs an exchange *operator* formulation.[4-8] The exchange operator is usually written in the form[9]

$$H_{\text{exch}} = \textstyle\sum_P \mathcal{J}_P P^{(\sigma)}, \tag{1}$$

where $P^{(\sigma)}$ is a pair permutation operator in spin space and $\mathcal{J}_P$ is an operator having diagonal and off-diagonal elements in position space (usually in a phonon basis). This concept was originally suggested by Thouless,[4] and developed later by