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Test of Tricritical Point Scaling in Dysprosium Aluminum Garnet

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The scaling hypothesis for a tricritical point has not received any tests from experimental data for magnetic systems. Here, data for dysprosium aluminum garnet are used to test the tricritical scaling hypothesis in the directions \( \vec{x}_1, \vec{x}_3 \) lying in the \( H-T \) plane. Specifically, \( M-H-T \) data near the tricritical point are found to collapse from a family of curves to a single curve (scaling function), supporting the validity of a tricritical scaling hypothesis.

In metamagnetic materials, and in certain model systems, the "critical line" \( H = H_c(T) \) terminates at a certain nonzero temperature \( T_c \), below which there is a line of first-order phase transitions. Griffiths has introduced the term "tricritical point" (TCP) to describe the point \( [T = T_c, H = H_c(T_c), H_{st} = 0] \), where \( H_{st} \) denotes the staggered magnetic field.

Dysprosium aluminum garnet (DyAlG) has a tricritical point at \( T_c = 1.66 \pm 0.01 \) K, \( H_c = 3.25 \) kOe, as determined by measurements of Landau et al. The TCP scaling hypothesis implies certain "data collapsing," and, conversely, data collapsing implies TCP scaling. The extent to which experimental data near the TCP collapse onto a single "scaling function" therefore provides a measure of the validity of the TCP scaling hypothesis. In this work we test data collapsing for \( M-H-T \) data (for \( H_{st} = 0 \)) near the tricritical point in DyAlG.

We formulate the TCP scaling hypothesis in terms of the Gibbs potential \( G(H, T) \); Near the TCP, \( g = G - G(H_c, T_c) + M(H - H_c) + S(T - T_c) \) is postulated to be a generalized homogeneous function, i.e., there exist three numbers ("scaling powers") \( \bar{a}_1, \bar{a}_2, \bar{a}_3 \) such that for all positive \( \lambda \),

\[
\lambda \bar{a}_1 \bar{x}_1, \lambda \bar{a}_2 \bar{x}_2, \lambda \bar{a}_3 \bar{x}_3 = \lambda g(\bar{x}_1, \bar{x}_2, \bar{x}_3).
\]

Here \( S \) is the entropy and the variables \( \bar{x}_j \) are defined at the TCP.

Equation (1) implies that the derivatives and Legendre transforms of \( g \) are also generalized homogeneous functions with scaling powers related to the original scaling powers \( \bar{a}_j \). In particular, we find the TCP exponents \( \beta_u, \gamma_u, \) and \( \delta_u \) (in the Griffiths notation) are expressible in terms of \( \bar{a}_2, \bar{a}_3 \) only,

\[
\beta_u = \frac{1 - \bar{a}_2}{\bar{a}_3}, \quad \gamma_u = \frac{1 - 2\bar{a}_2}{\bar{a}_3}, \quad \delta_u = \frac{1 - \bar{a}_2}{\bar{a}_3},
\]

where \( \beta_u, \gamma_u, \) and \( \delta_u \) are defined, respectively, by the relations

\[
M^u - M^u \sim |T - T_c|^{\beta_u}, \quad \chi \sim |T - T_c|^{\gamma_u}, \quad M - M_c \sim |H - H_c|^{\delta_u}.
\]

Eliminating \( \bar{a}_2, \bar{a}_3 \) from (2a), we obtain the TCP

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scaling law,

\[ \delta_u = 1 + \gamma_u / \beta_u. \]  

Wolf et al.\(^a\) have determined \( \beta_u, \gamma_u \) for DyAlG near its TCP, with the results \( \beta_u = 0.65 \pm 0.05, \gamma_u = 1.3 \pm 0.1 \). Accordingly, (3) predicts \( \delta_u = 3.0 \pm 0.4 \). As a crude graphical test of the TCP scaling hypothesis, we estimated the value of \( \delta_u \) from the data, and found that it is consistent with (3), but just within experimental error. Unfortunately, data along the isotherm \( T = T_i \) are not available, so visual interpolation was necessary. This process was also made more difficult by uncertainty in the value of the tricritical field \( H_t \).\(^b\) Using log-log plots of \( M - M_i \) versus \( H - H_t \) for several trial values of \( H_t \), and also plots of \( |M - M_i|^\delta_u \) versus \( H \) for several trial values of \( \delta_u \), we conclude that the data are roughly consistent with \( \delta_u = 3 \), and \( H_t = 3.30 - 3.45 \) kOe.

The scaling function predictions for \( m(H, T) = (M - M_i)/M_i \) data with \( \bar{x}_i = H/H_t = 0 \) may be obtained by differentiating Eq. (1) with respect to

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**FIG. 1.** \( M - H - T \) data near the TCP for DyAlG plotted in two scaled forms. Here we have taken \( \beta_u = 0.65, \delta_u = 3, H_t = 3.37 \) kOe, \( T_t = 1.665 \) K; magnetization is measured in units of \( M_i = 250 \) emu/cm\(^3\). In (a), we scale by \( \bar{x}_2 = h + \epsilon \times \tan \delta \); the branch above the abscissa is for \( \bar{x}_2 > 0 \), while that below is for \( \bar{x}_2 < 0 \). In (b), we scale by \( \bar{x}_3 = \epsilon \); the branch through the origin is for \( \bar{x}_3 = (T - T_t)/T_t > 0 \), while the branch for \( \bar{x}_3 < 0 \) exhibits a discontinuity at \( \bar{x}_2 = 0 \). All solid curves are simply eyeball fits to the data.
\( \bar{x}_z \) and setting \( \lambda = (1/|\bar{x}_z|)^{1/2} \). We obtain

\[ m(\pm 1, \bar{x}_z/|\bar{x}_z|^{1/2} \delta_u) = m(\bar{x}_z, \bar{x}_z) / |\bar{x}_z|^{1/2} \delta_u / \delta_u. \]

(4)

Substituting \( 1/\delta_u = (1 - \bar{x}_z)/\bar{x}_z \) and \( 1/\beta_u \delta_u = \bar{x}_z/\bar{x}_z \), from (2a), we find that \( m \) scaled by \( \bar{x}_2^{1/2} \delta_u \) is a function of \( \bar{x}_z \) scaled by \( \bar{x}_2^{1/2} \delta_u \). To use Eq. (4), we transform the coordinates of a point \((H, T)\) to \((\bar{x}_z, \bar{x}_z)\),

\[ \bar{x}_z = h + \epsilon \tan \theta, \]

(5a)

\[ \bar{x}_z = \epsilon. \]

(5b)

Here \( \epsilon \equiv (H - H_1)/H_1 \), \( \epsilon \equiv (T - T_1)/T_1 \), and \( \theta \) is determined from the slope at the TCP of the phase boundary in \( h - \epsilon \) space. We estimate \( \tan \theta = 0.60 \), Were the phase boundary parallel to the \( T \) axis at the TCP, \( \theta \) would be zero, and one would simply have the ordinary "Widom variables," \( \bar{x}_z = h \), \( \bar{x}_z = \epsilon \).

About 100 scaling plots of \( m / |\bar{x}_z|^{1/2} \delta_u \) versus \( |\bar{x}_z| / |\bar{x}_z|^{1/2} \delta_u \) [cf. Eq. (4)] were constructed for different combinations of the four parameters \( T_1, H_1, \beta_u \), and \( \delta_u \), which were varied within the bounds of the experimental uncertainties. All plots include data from a wide range of temperatures (seven isotherms, from 1.35 to 2.3 K) and magnetizations (from 0.5\( M_s < M < 1.5 \)) \( M_s \).

The degree of data collapsing is more sensitive to the choice of \( T_1 \) and \( H_1 \) than \( \beta_u \) and \( \delta_u \). Figure 1(a) is one such plot with \( T_1 = 1.665 \) K, \( H_1 = 3.37 \) kOe, \( \beta_u = 0.65 \), and \( \delta_u = 3 \). This set of parameters is one of several which produce good data collapsing. Renormalization-group calculations \( 11 \) of tricritical exponents predict \( \beta_u = 0.97, \gamma_u = 1.07, \delta_u = 2.1 \) for the He\(^3\)-He\(^4\) system. Using these values, we find the data collapsing for DyAlG is definitely poorer than that shown in Fig. 1. One might conclude that the theoretical treatments, while they may suffice for He\(^3\)-He\(^4\), fail in some way to describe the DyAlG TCP.\(^{12}\)

Figure 1(b) shows the same data scaled a second way; one differentiates with respect to \( \bar{x}_z \), sets \( \lambda \equiv |\bar{x}_z|^{-1/2} \delta_u \), and plots \( m / |\bar{x}_z|^{1/2} \delta_u \) versus \( |\bar{x}_z| / |\bar{x}_z|^{1/2} \delta_u \). Two branches are apparent, one for \( \bar{x}_z = (T - T_1)/T_1 > 0 \) and one for \( \bar{x}_z < 0 \); the discontinuity of the latter branch at \( \bar{x}_z / |\bar{x}_z|^{1/2} \delta_u = 0 \) indicates the first-order transition.

In summary, the DyAlG data appear to reduce from a family of isotherms (Fig. 6 of Ref. 1a) to two distinct branches when plotted in either one of the two scaling forms, Figs. 1(a) or 1(b). This data collapsing occurs over what we feel is a surprisingly wide temperature range, sug-


17. Certain data collapsing predictions of tricritical-point scaling have been considered in the He\(^3\)-He\(^4\) system by G. Goldner and H. Meyer (private communication); see also G. Goldner and H. Meyer, Phys. Rev. Lett. 26, 1554 (1971).

18. The data upon which Fig. 6 of Ref. 1a was based have been very kindly supplied to us in tabular form by W. P. Wolf and D. P. Landau.

19. Consider the critical line \( L_1 \) in the \( H-T \) plane (Fig. 1 of Ref. 4b). At each point \( P \) on \( L_1 \), we can define three directions. The first direction, \( x_4(P) \), is a direction perpendicular to the coexistence surface and is called the "strong" direction. The second direction, \( x_4(P) \), is parallel to the coexistence surface, but is not parallel to the critical line \( L_1 \); it is called the "weak" direction. The third direction, \( x_4(P) \), is parallel to the critical line and is called the "independent" direction. As the point \( P \) moves toward the tricritical point, these three directions attain limiting orientations, and we define \( F_4 = \lim_{P \to \text{TCP}} x_4(P) \).


21. A value of \( H_1 = 3.25 \) kOe is too low, as evidenced by the fact this isomorph does not pass through the TCP in Fig. 6 of Ref. 1a.

22. In the experiment of Refs. 1, data were taken in a
rough grid to map the region 0.5–8.0 K in temperature and 0–15 kOe in field; note that the isotherm passing nearest the TCP is at $c \approx 0.03$. Unfortunately, there are considerable experimental difficulties associated with measuring magnetizations near the TCP and the first-order line; these are discussed in Refs. 1a, 1b, 15 K. Riedel and F. J. Wegner, Phys. Rev. Lett. 29, 349 (1972).

A second possibility, though, is that the area around the TCP in which true asymptotic behavior could be observed is in fact quite small, so that the present data simply do not penetrate the actual "tricritical region." Of course, this interpretation is possible for all exponent data.

Pressure Variation of the Quadrupole Interaction in Cadmium Metal*

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A theoretical study, for the first time, of the electric field gradient in cadmium metal at two pressures yields a pressure variation in agreement with time-differential perturbed-angular-correlation data. The sign of the net electric field gradient determined by the electronic contribution, which is larger and of opposite sign to the ionic contribution, in conjunction with imminent expected experimental sign of the coupling constant, will provide the sign of the excited Cd$^{111}$ quadrupole moment.

The recent exploitation of the time-differential perturbed-angular-correlation (TDPAC) technique1–4 to study nuclear quadrupole interactions in metals has provided a valuable opportunity to improve our understanding of the origin of the nuclear quadrupole effects in metals. This technique provides a means of studying quadrupole interactions in excited states of those nuclei which have zero quadrupole moment in the ground state and would therefore not have been suitable for study of field gradients at nuclear sites. One example which has been exploited most extensively in this respect is the excited state of Cd$^{111}$ which has a spin of $\frac{5}{2}$. In particular, both the quadrupole coupling constant in cadmium metal and its variation2–3 with pressure and temperature have been studied recently. In the present study, for the first time in a metal, a theoretical analysis is carried out for the pressure variation of the field gradient in cadmium, including both conduction electron and ionic contributions, for comparison with experiment. An equally important and timely result of the present study is the sign of the field gradient, since development of experimental procedures are currently5 under way to determine the sign of the coupling constant $e^{2}qQ$ through the TDPAC technique. A combination of our theoretical sign for $e^{2}qQ$ and the experimental sign for $e^{2}qQ$, when the latter is available, should provide the sign of $Q$ for the excited state of Cd$^{111}$, which should be very useful for the testing of nuclear theories for $Q$.

The study of the conduction electron contribution6–8 to the field gradient $e^{2}qQ$ requires a detailed scanning of the occupied Fermi volume, rather than just the Fermi surface as is the case for magnetic hyperfine properties. The band energy values and wave functions have to be calculated at each of the scanned $k$ points in the Brillouin zone (reduced zone scheme), the energy values being needed to decide which of the energy bands at the point lies below the Fermi energy. The occupied eigenstates are used7–8 to calculate the electronic contribution to the field gradient. In the present work, we have carried out two such calculations for two pressures, namely 1 atm ($\approx$ 1 bar) and 50 kbar.

In common with the earlier successful interpretations9–10 of Fermi-surface properties of cadmium, a pseudopotential approach is utilized here for obtaining the energy bands and band wave functions. However, in our present work, we require wave functions in the entire Fermi volume rather than the vicinity of the Fermi surface. The available nonlocal energy-dependent pseudopotential having been derived for the latter, it has to be modified to apply to all of $k$ space. The procedure for this shall be briefly described.

The general form of the energy-dependent pseudopotential is given by

$$V_{s}(E) = V_{c} + \sum \delta(E - E_{i})P_{i},$$

(1)

where $V_{c}$ is the real potential seen by the conduction electrons, and $P_{i}$ is the projection operator.