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Dipole-dipole interactions and the critical resistivity of gadolinium

D. J. W. Geldart and K. De'Bell
Department of Physics, Dalhousie University, Halifax, Nova Scotia, Canada B3H 3J5

J. Cook and M. J. Laubitz
Division of Physics, National Research Council, Ottawa, Ontario, Canada K1A 0R7

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Results are presented for the electrical resistivity of a c-axis single crystal of high-purity gadolinium metal in the immediate vicinity of the Curie temperature. Numerical analysis reveals that the data are not well described by the usual power laws in $(T-T_c)/T_c$ but tend to exhibit a change in effective slope at T_c which can be described by classical Landau theory with logarithmic corrections. This is interpreted in terms of the dipole-dipole interactions, which are shown to become important at about 1 K above T_c. We suggest that gadolinium is a uniaxial dipolar ferromagnet in the asymptotic critical-behavior regime.

The electrical resistivity of gadolinium has unusual behavior in the vicinity of its Curie temperature.[1-3] The c-axis resistivity has a peak at, or very close to, T_c. The apparent negative slope even for $T \approx T_c^+$ is in disagreement with the theory of Fisher and Langer[4] which requires a positive slope at T_c for the spin-fluctuation-induced resistivity $\rho_{sp}(T)$. The contradiction was resolved by Zumsteg, Cadieu, Marcelja, and Parks[2] who attributed the peak at T_c to anomalous thermal expansion in the c direction.[5] After correcting for this thermal expansion, the resulting $\rho_{sp}(T)$ was qualitatively of the form predicted by Fisher and Langer.[4]

The basal-plane resistivity is already qualitatively of the Fisher-Langer form, in that it has positive slope near T_c,[1-3] without invoking thermal expansion effects. A very interesting study was made by Simons and Salamon[3] who measured both the basal-plane resistivity and the specific heat in a range of magnetic fields and found a direct proportionality between the temperature dependence of $C_H(T)-C_{H=0}(T)$ and $\rho'_H(T)-\rho'_{H=0}(T)$ for a range of field strengths H. The fact that these quantities exhibit the same temperature dependence was taken as indirect verification of the Fisher-Langer theory which predicted $\rho'_{sp}(T) \propto |t|^{-\alpha}$, where α is the specific-heat critical exponent and $t=(T-T_c)/T_c$ is the reduced temperature.[6]

The above works support the expected proportionality between $\rho'_{sp}(T)$ and the specific heat. However, they do not determine the specific temperature dependence, e.g., the critical exponent of the power-law behavior. We felt that a detailed study of the temperature dependence is highly desirable for several reasons. An alternative experimental determination of the specific-heat critical exponent would certainly be useful. In addition, there is some interest in exploring a possible crossover from the asymptotic regime of short distance correlations very near T_c to an alternative longer-range description at higher temperature.[7,8]

The objective of this work has therefore been to determine the precise temperature dependence of $\rho_c(T)$ near T_c and to shed light thereby on the nature of the phase transition in Gd. An electrotransport-purified single crystal of Gd of dimensions $2\times2\times9$ mm^3, oriented along the c axis, was made at Ames Laboratory. Its resistance ratio was $R(295\text{ K})/R(4\text{ K})=156$. Similar pieces of lower purity were butt welded to the ends to provide current contacts. Copper wire was pressed onto the end segments for current leads while fine stainless-steel needles served as potential leads for the middle segment of highest purity. The sample holder was placed in a well-stirred oil bath designed for the calibration of thermometers at the National Standards Laboratory of the National Research Council, Ottawa. Its temperature was measured to within 1 mK by a platinum resistance thermometer calibrated to within 1 mK of the International Practical Temperature Scale of 1968 at temperatures of present interest.

The resistance of the sample was measured using a model No. 9920 Kuster's dc comparator bridge (Guildline Instruments, Smiths Falls, Ontario) in the resistance configuration with a 1 mΩ standard resistance. All measurements were made at 0.5 A using rapid current reversals and a null was sought using the bridge galvanometer. Data were taken in two runs. Run I covered the range of approximately 287 to 298 K. Reproducibility was very good and no hysteresis was observed on thermal cycling. Run II covered the range 293.2 to 293.8 K, where the peak was located, with higher precision. These data points were taken in a single warming cycle at intervals of about 40 mK and with an accuracy of about (3×10^{-4})%.

Since our objective was to study the relative temperature dependence of the resistance very near T_c, rather than to provide absolute values of either the resistivity or the temperature, we ignore the form factor of the sample and discuss our results in terms of the ratio $r(t) = R(T)/R_0=\rho(T)/\rho_0$, where ρ_0 is the resistivity at a point (near the peak) chosen for normalization purposes. Similarly, we also ignore a constant correction to the temperature, determined experimentally to be about 50 mK, due to self-heating. This affects only the specification of the absolute value of T_c and does not influence our discussion of the dependence on the temperature difference $T-T_c$.

In order to determine the precise temperature dependence of $r(T)$, we constructed nonlinear least-squares fits in the usual way. We first considered the standard power law

$$r(T)=A_0+A_1t+B_\pm|t|^x\ , \tag{1}$$

where $B_\pm$ denotes the amplitude for $T \gtrless T_c$ of the singular term, and attempted to obtain fits in various temperature ranges to observe sensitivity to the range of fit; $|T-T_c| \lesssim 1$ K, 1.5 K, 2.5 K, and 5 K for run I and $|T-T_c| \lesssim 0.3$ K for run II. Whenever a fit to the data could be found, systematic structure was present in plots of the residuals (the difference between the measured r and the fit to the data) thus indicating poor quality and an unreliable representation of the data. Usually no fit could be obtained but there was a consistent tendency for the exponent x to approach 1, mimicking a change of slope at $t=0$. We conclude that Eq. (1) does not give a satisfactory fit. Therefore, to test for the possibility of a change of slope at T_c, we considered a fit to

$$r(T)=A_0+A_2t^2+B_\pm|t|^{x\pm}+C_\pm|t| \tag{2}$$

over the full temperature range, to include all data points in view of the large number of free parameters. Different critical exponents $(x_\pm)$ and different slopes $(C_\pm)$ were allowed for $T \gtrless T_c$. Upon iteration of the fitting routine, both the amplitudes $B_\pm$ and the exponents $x_\pm$ converged to essentially zero with estimated uncertainties which were also essentially zero to within the numerical accuracy of the fitting routine (the uncertainties in $B_\pm$ and $x_\pm$ were given as $\sim 10^{-35}$ and $\sim 10^{-14}$, respectively). The probable errors associated with all other parameters were, of course, much larger and of a magnitude consistent with the noise level of the data. This strikingly indicates that the data are much better described by a change of slope rather than by a standard power law. Other parameters in this fit were $A_0=1.00003 \pm (2\times 10^{-5})$, $A_2=-2.21 \pm 0.28$, $C_-=-0.08236 \pm 0.005$, $C_+=-0.2168 \pm 0.005$, $T_c=293.454 \pm 0.028$, and the rms error was 3×10^{-5}. These results were stable upon addition of a further regular A_3t^3 term and plots of residuals appeared to be reasonably random.

A discontinuity in slope at T_c might raise the question of whether the phase transition is of second order. However, we recall that no effects of hysteresis were observed so we adopt the view that the transition is indeed of second order. Consequently, we must provide a fundamental explanation, based on the known interactions in the material, of why the resistivity of Gd should exhibit, to within the accuracy of the data, an apparent change in slope at T_c.

We find that this unusual feature can be understood in terms of the dipole-dipole contribution, H_{dd}, to the interactions. Although the usual exchange interaction is the dominant interaction driving the phase transition in Gd, the weak H_{dd} becomes important near T_c because of its long range.[9,10] In addition, H_{dd} couples spin directions to crystallographic directions with interesting consequences even for an S-state ion having otherwise isotropic interactions. To suggest the significance of this fact, we have found by direct numerical evaluation of lattice sums that H_{dd} is minimized if the spins order along the c axis of the hcp lattice. This is obviously consistent with the c axis of Gd being the unique easy axis of magnetization in the ordered state. To obtain a rough estimate of when dipolar interactions become important as the temperature is lowered in the paramagnetic state, we have also evaluated numerically the mean-field shift in transition temperature, $\Delta T_c^{\rm MF}$, induced by H_{dd} and obtained $\Delta T_c^{\rm MF}=1.729$ K. This sets the overall scale for dipolar effects and we thus expect a crossover at about $T-T_c \sim 1.5$ K from the usual Heisenberg behavior at higher T to a dipolar region. Moreover, the asymptotic critical behavior will be that of a uniaxial (Ising) dipolar ferromagnet (due to the unique easy axis) with the basal-plane spin components having only secondary critical behavior.[11] This is in contrast to a dipolar ferromagnet which is isotropic with respect to critical fluctuations along different crystallographic directions.

A uniaxial dipolar ferromagnet has the remarkable property that its critical behavior is described by classical Landau theory apart from (nontrivial) logarithmic corrections, due to the fact that its upper critical dimension is $d_c=3$ as opposed to $d_c=4$ for short-range-interaction systems.[12–14] As a result, the usual power-law behavior of critical phenomena is absent. For example, for T near T_c, the correlation length satisfies[12–14]

$$1/\xi^2=A_\pm|t|\,\big|\ln|t|\big|^{-1/3}\ , \tag{3}$$

where $A_+/A_-=\frac{1}{2}$. Similarly, the specific heat is found to have the leading T dependence given by[12–14]

$$C_p(T)=B_\pm\big|\ln|t|\big|^{1/3} \tag{4}$$

with $B_+/B_-=\frac{1}{4}$.

To appreciate the leading effect of critical fluctuations on the electrical resistivity, we apply the usual theory[4,7,8,15,16] based on the Born approximation, quasielastic scattering, and a simple spherical (or ellipsoidal) Fermi surface (for simplicity),

$$\rho_{\rm sp}(T)\propto\int d^3q\,\Phi(\mathbf{q})G^{zz}(\mathbf{q},T)\ , \tag{5}$$

$\Phi(\mathbf{q})\propto q$ for $q<2k_{\rm FC}$, and $\Phi(\mathbf{q})=0$ for $q>2k_{\rm FC}$, where $2k_{\rm FC}$ is the Fermi-surface caliper in the c direction. The spin correlation function of the c-axis (defined as the z direction) spin fluctuations, at least for $q\xi \lesssim 1$, is

$$G^{zz}(\mathbf{q},T)=1/(\kappa^2+\mathbf{q}^2+g_0q_z^2/\mathbf{q}^2)\ , \tag{6}$$

where $\kappa^2=1/\xi^2$ of Eq. (3) and g_0 is the dipolar coupling constant.[12,13] The integral in Eq. (5) is elementary and the leading term is

$$\rho_{\rm sp}(T)-\rho_{\rm sp}(T_c)\propto-\kappa^2\propto-|t|\,\big|\ln|t|\big|^{-1/3}+\cdots\ . \tag{7}$$

As we will see in the following, these logarithmic corrections are very difficult to detect over a range of one or two decades, since the stronger linear $|t|$ dependence dominates. It is easy to verify that both Eq. (7) and the exact integral Eq. (5) appear to be rather linear in t and, in fact, resemble the experimental results. Of course, Eq. (6) will not apply close enough to T_c where $2k_{\rm FC}\xi<1$ is seriously violated. Estimating parameters appropriate for a simple model of Gd,[8] we find $2k_{\rm FC}\xi \gtrsim 1$ for $t \lesssim 10^{-2}$. However, in contrast to Eq. (7), the temperature dependence of ρ in this range of short-distance correlations near T_c (which also includes the range of crossovers and dipolar domination) can be deduced from $\rho'_{sp} \propto C_p$ which implies, from Eq. (4),

$$\rho_{sp}(T) - \rho_{sp}(T_c) \propto |t| \, |\ln|t|\,|^{1/3} + \cdots , \qquad (8)$$

with an amplitude ratio for this contribution of $-\frac{1}{4}$.

Before describing nonlinear least-squares fits to data of forms containing Eq. (7) or Eq. (8), we must emphasize that the above $|\ln|t|\,|^{1/3}$ is already an approximation to $|a+\ln|t|\,|^{1/3}$, where a is a nonuniversal constant expected to be of order unity.[12,13] Dropping this constant as well as other next-to-leading correction terms in order to reduce the number of free parameters is a reasonable approximation only if $\ln|t|$ is not too small. Furthermore, over a relatively small temperature range, $|\ln|t|\,|^{1/3}$ is very slowly varying and difficult to detect in fits to data. The same comments apply to $|\ln|t|\,|^{-1/3}$.

To test for a consistent description of the data in terms of the above logarithmic corrections, we attempted fits of the data to

$$r(T) = A_0 + A_1 t + B_{\pm} |t| \, |\ln|t|\,|^{x}$$

in the temperature ranges $\Delta T \lesssim 0.3$ K (run II only), 1, 1.5, and 2.5 K to determine sensitivity to the range of fitting, as before. As expected, no fits could be obtained when all parameters were allowed to vary freely. We therefore constrained x to be $\pm\frac{1}{3}$ and varied T_c while seeking fits consistent with $B_+/B_- \approx -\frac{1}{4}$ for $x = \frac{1}{3}$ and $B_+/B_- \approx \frac{1}{2}$ for $x = -\frac{1}{3}$ as suggested by the previous discussion. Good fits could be obtained for both $x = \pm\frac{1}{3}$ in the range $\Delta T \le 0.3$ K while rms errors increased in the larger temperature ranges as expected. A comparison of the experimental and fitted results in the two cases, $x = \pm\frac{1}{3}$, is given in Fig. 1. It is clear that the fits to the data appear to be very nearly linear and that the logarithmic corrections are extremely slowly varying. The previously noted strong tendency of fits to the data to prefer a change in slope rather than a standard power law is therefore understandable. In the available temperature interval, the fractional powers of logarithms only contribute to the effective slope.

We conclude that the behavior of the c-axis resistivity of Gd is consistent with our suggestion that the asymptotic critical behavior is that of a uniaxial dipolar ferromagnet, described by Landau theory with logarithmic corrections. This clearly also must apply for the basal-plane resistivity, and features of the published results for a-axis resistivity indeed lead us to believe that an analysis of the type which we have given would confirm this (note in this context that the phonon background slope in the basal-plane resistivity is more than twice that of the c-axis resistivity). We observe that although uniaxial dipolar ferromagnetic insulators have previously been studied, with one of the best known examples being $LiTbF_4$,[17,18] to our knowledge Gd is the first example suggested of a uniaxial dipolar ferromagnetic metallic conductor, at least as far as its asymptotic critical behavior is concerned.

In previous experiments on Gd in the critical regime, any detailed analysis has been based on fitting of data to power laws, as is appropriate for three-dimensional systems with short-range interactions. Although a fit to the data will generally be possible, the fit may not be of good quality and the parameters, such as critical exponents, which result, may be difficult to interpret. Consider the example of the spontaneous magnetization $M(t) \propto |t|^{\beta}$.[6] Very careful work by Chowdhury, Collins, and Hohenemser[19] yields the estimate from experiment $\beta_{expt} = 0.399 \pm 0.016$ which is close to the theoretical prediction of $\beta_H = 0.3645 \pm 0.0025$ for the Heisenberg model far from that for Ising systems $\beta_I = 0.3250 \pm 0.0020$.[20] The discrepancy between β_{expt} and β_H was attributed to the fact that the experimental temperature range, $|t| > 10^{-3}$, was not sufficiently asymptotic. We suggest that the data are also influenced by dipolar effects since the crossover from Heisenberg to dipolar critical behavior was estimated above to occur at about 1.5 K from T_c, or at a reduced temperature $t_d \approx 5 \times 10^{-3}$. In the asymptotic regime of uniaxial dipolar critical behavior, the spontaneous magnetization varies as $M(t) \propto |t|^{1/2} |\ln \times |t|\,|^{1/3}$.[12-14] Under these circumstances, it is certainly plausible that the effective exponent should lie between the Heisenberg value and the classical Landau value, as is

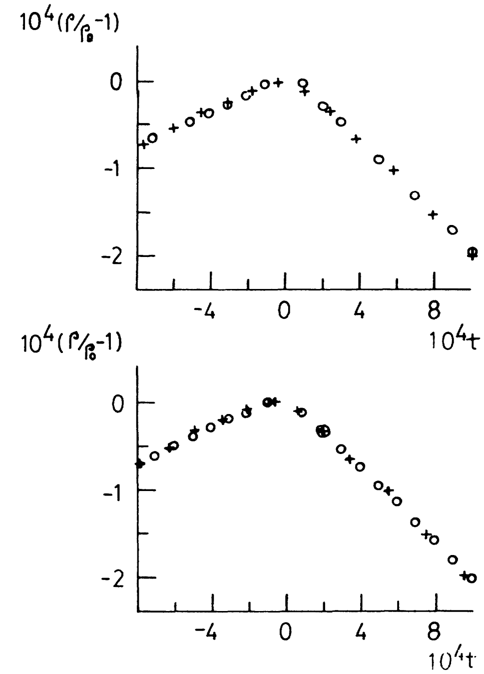

FIG. 1. Comparison of experimental data, represented by crosses (+), and fits to the data, based on logarithmic corrections due to dipole-dipole interactions in an Ising-type system as described by Eq. (9), represented by circles (○). The parameters in the upper plot, for $x = \frac{1}{3}$, are $A_0 = 1.00$, $A_1 = -0.32$, $B_+ - 0.06$, $B_- = 0.22$, and $T_c = 293.461$ K with rms error $= 0.4 \times 10^{-5}$. The parameters in the lower plot, for $x = -\frac{1}{3}$, are $A_0 = 1.00$, $A_1 = -0.12$, $B_+ = -0.18$, $B_- = -0.41$, and $T_c = 293.471$ K with rms error $= 0.3 \times 10^{-5}$. The slowly varying fractional power of the logarithm cannot be distinguished from a contribution to the effective slope in this temperature interval. (The number of figures reported for T_c represents "fine tuning" to obtain the desired B_+/B_- ratio—see text.)

observed. Similar situations exist for other critical exponents, both static and dynamic.[21] In this context, note the recent study of spin autocorrelation times in Gd above T_c by perturbed angular correlations.[22] The spin fluctuations were isotropic for $t > 3\times10^{-3}$, but the critical exponent, w, describing the wave-number-averaged autocorrelation time did not agree with either the predicted Ising or Heisenberg value, and a crossover to uniaxial spin fluctuation was found for $t < 3\times10^{-3}$.[22]

We must emphasize that it is not at all trivial to detect convincingly multiplicative logarithmic corrections to power laws even if the additional problem of a crossover to Heisenberg behavior were not present. For this reason, a new precise specific-heat experiment in the reduced temperature range $10^{-5} \lesssim |t| \lesssim 10^{-3}$ would be particularly useful as the logarithmic corrections, Eq. (4), are the leading singular terms and are not masked by a power law, e.g., as in Eq. (7). A detailed numerical analysis of data should then be feasible. It should be pointed out that the possibility of a discontinuity in addition to a power law has been suggested for both specific-heat[23] and thermal expansion[24] data on Gd near T_c. It is clear that additional experimental and theoretical work will be useful to elucidate the suggested role of dipolar interactions on the interesting critical behavior of Gd.

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