Anisotropy of the Critical Magnetic Susceptibility of Gadolinium

D. J. W. Geldart, P. Hargraves,[a] N. M. Fujiki, and R. A. Dunlap
Department of Physics, Dalhousie University, Halifax, Nova Scotia, Canada B3H 3J5
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The magnetic susceptibility along the c axis (χ_c) and in the basal plane (χ_b) has been measured on a single crystal of Gd in the reduced temperature range $4\times10^{-4} < t < 1.3\times10^{-2}$. Uniaxial anisotropy is observed. Magnetic dipole-dipole interactions are shown to account for the magnitude of the uniaxial anisotropy and also to lead to complex crossover effects. The observed temperature dependence of $\chi_c(T)$ and $\chi_b(T)$ is described by effective exponents $\gamma_{\rm eff}=1.23\pm0.02$ and $1-\alpha_{\rm eff}=1.01\pm0.03$, respectively.

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The magnetic properties of gadolinium near the Curie temperature have been the subject of considerable debate recently. Even the proper static universality class has not been clearly determined. On the one hand, the picture of S-state ions coupled by isotropic Ruderman-Kittel-Kasuya-Yosida interactions implies Heisenberg critical behavior near T_c. On the other hand, the unique easy (c axis) direction of magnetization implies uniaxial anisotropy which suggests Ising critical behavior. The experimental situation has been ambiguous although critical magnetic properties of Gd have been reported using various methods. While the exponents associated with the magnetization and the specific heat, β and α, respectively, typically yield values near the theoretical Heisenberg values,[1-6] the exponent for the magnetic susceptibility, γ, is typically found to be near the Ising value.[2,3,7-9] This contradictory behavior leads to striking disagreements between theory and experiment, including the violation of scaling laws such as $\alpha+2\beta+\gamma=2$. Such violations are all the more strange since Gd is widely considered to be a prototype of a "simple" ferromagnet. We report here the resolution of this paradox based on a careful experimental study of the paramagnetic susceptibility which we find to exhibit uniaxial anisotropy in the critical regime. The previously unexpected anisotropy is interpreted in terms of magnetic dipole-dipole interactions which are not only surprisingly strong in Gd but are also anisotropic with respect to crystal directions.

The question of magnetic anisotropy is clearly of crucial importance in establishing the universality class of Gd, but it has rarely been dealt with directly. Two potentially relevant experiments have been reported, with contradictory conclusions. Belov *et al.*[10] found the apparent ordering temperature for magnetization in hard directions (in the basal plane), T_b, to be 1.5 ± 0.1 K below the Curie temperature, T_c, for ordering along the easy direction (along the c axis). This difference should be reflected in the corresponding paramagnetic Curie-Weiss temperatures. However, Nigh, Legvold, and Spedding[11] observed no anisotropy and concluded that $\theta_c=\theta_b=317\pm3$ K. Unfortunately, these experimental results have limited implications for the critical regime close to T_c, since, in both cases, the required magnetizations were determined by extrapolation from high fields using the relation $AM+BM^3=H$. This procedure is strictly valid only in a mean-field regime (Landau theory), where fluctuations can be neglected, but is not sufficiently reliable to allow firm conclusions in the true critical regime.

We report here the first direct observation of magnetic anisotropy valid in the critical region above T_c, as determined from low-field magnetic susceptibilities, $\chi_b(T)$ and $\chi_c(T)$, for applied fields in the basal plane and along the c axis, respectively. The sample consisted of a single crystal of high-purity Gd cut into the form of a cube with a mass of 0.1217 g and an edge length of 0.249 ± 0.005 cm. The c axis was oriented perpendicular to two opposite faces of the cube. The original crystal from which this sample was cut was prepared by the Ames Laboratory, Energy and Mineral Resources Research Institute, and prior to measurements was electropolished following the procedure described by Beaudry and Gschneidner.[12]

The magnetic susceptibility above T_c was measured using an ac spectrometer of the Hartshorn bridge type, as modified by Brobeck, Burkey, and Hoeksema.[13] The primary ac drive field has an amplitude of 1.6 $\mathrm{A\,m^{-1}}$ at a frequency of ~3000 Hz. Measurements were made both with the drive field along the c axis and with the drive field in the basal plane. The real part of the intrinsic susceptibility, $\chi(T)$, has been obtained from the measured demagnetization-limited susceptibility, $\chi_{\rm ext}(T)$, using the demagnetization factor N as[14]

$$\chi^{-1}(T)=\chi_{\rm ext}^{-1}(T)-N\,. \tag{1}$$

The cubic sample geometry was chosen to minimize systematic experimental errors which might interfere with direct comparison of basal-plane and c-axis data and also to minimize errors in estimates of demagnetization factors for the two configurations. The validity of Eq. (1) for taking account of demagnetization effects can be assessed by comparing the present results with those which we previously reported for a high-purity single-crystal sample of considerably different geometry.[9] In both cases, the average demagnetization correction for the c-axis susceptibility in Eq. (1) is determined solely on the basis of geometrical considerations. The corrected susceptibilities thus determined are the same in the two cases, to within the expected uncertainties in measuring $\chi_{ext}(T)$, indicating a consistent treatment of average demagnetization effects. In this previous work[9] we also presented a detailed analysis of our ac susceptibility measurements in the context of previous ac and low-field dc susceptibility experiments. These investigations led us to conclude that our present measurements are made in a regime where applied-field- and frequency-dependent effects are not important. The temperature was measured using a Chromel-Constantan thermocouple to a relative accuracy of about 10 mK. A second drive field at a frequency different from the primary drive frequency was applied in order to detect the temperature at which the measurements were affected by domain-wall nucleation.[15,16] If χ_{ext} is compromised by the presence of domain-wall pinning then the second field will, at least partially, unpin the domain walls and hence increase the measured χ.

The results for the inverse of the measured susceptibility, corrected for demagnetization, are shown in Fig. 1 as a function of temperature for the two configurations. χ_c^{-1} and χ_b^{-1} correspond to crystal orientation with the drive field along the c axis and with the drive field in the basal plane, respectively. Domain-wall nucleation was observed for the c-axis configuration at approximately 293.70 K. In order to analyze the two sets of data over the same range of temperature, we have used a common minimum temperature of $T_{\min} \approx 293.70$ K and a common maximum temperature of $T_{\max} \approx 297.40$ K. For an initial appraisal, we consider nonlinear least-squares fits by simple power laws. We shall see that this is appropriate, in fact, and that the exponents are to be understood as effective exponents influenced by crossover effects (see

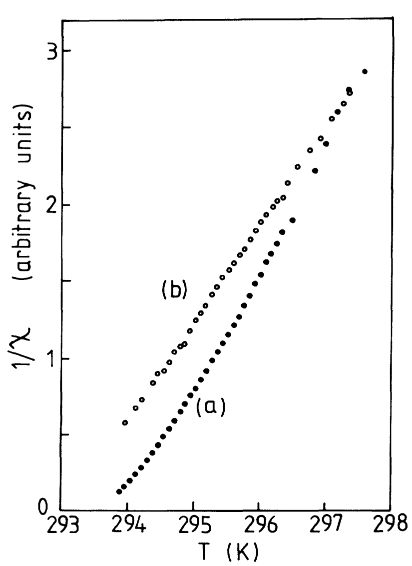

FIG. 1. Inverse magnetic susceptibility $(1/\chi)$ as a function of temperature for a single crystal of Gd measured along the c axis (plot a) and in the basal plane (plot b).

Refs. 17–19 for reviews of such effects). The c-axis data are represented by

$$\chi_c^{-1}(T) = At^{\gamma}, \tag{2}$$

where A, γ, and T_c, which enters $t=(T-T_c)/T_c$, are free parameters. In contrast to $\chi_c^{-1}(T)$ which is expected to vanish at T_c, $\chi_b^{-1}(T)$ is expected to remain finite at T_c, in the presence of anisotropy.[20,21] Consequently, the basal-plane data are represented by

$$\chi_b^{-1}(T) = B + Ct^{y}, \tag{3}$$

where B, C, and y are free parameters. Since $\chi_b^{-1}(T)$ is almost linear in T (as is already clear from Fig. 1), we have chosen to remove unphysical correlation of parameters by fixing the T_c in Eq. (3) at the value determined from the c-axis data. The parameters resulting from these fits are given in Table I. Finally, range-of-fit analyses were made in temperature intervals between $T_{\min}$ and $T_{\max}$ but revealed no anomalous systematic trends in the values determined for the various critical parameters.

The fact that the basal-plane (hard directions) susceptibility remains finite at T_c is the signature of uniaxial anisotropy. Before turning to the question of the physical origin of this anisotropy, we give a direct experimental estimate of its magnitude. Since all experimental conditions for the c-axis and basal-plane susceptibility measurements have been maintained as close to each other as possible to facilitate comparison of data, we may define a reduced-temperature scale for the anisotropy by $\chi_b^{-1}(T_c) = \chi_c^{-1}(T_c + \Delta T_{\text{anis}})$, i.e., $B = At_{\text{anis}}^{\gamma}$ which gives $t_{\text{anis}} = 0.195 \times 10^{-2}$ and $\Delta T_{\text{anis}} = 0.57$ K. Based on the statistical uncertainties of the data in Table I and also on comparison of results of a variety of different runs (not given here), we expect the uncertainty in these direct estimates of anisotropy temperature scales to be of order 15%. An independent estimate of this anisotropy temperature scale is given by observing that $\chi_b^{-1}(T)$ extrapolates to zero at a temperature which is below T_c by $\Delta T'_{\text{anis}} = 0.52 \pm 0.05$ K. A further clear indication of anisotropy is provided by the work of Collins, Chowdhury,

TABLE I. Fitted parameters for c-axis and basal-plane χ^{-1} for gadolinium. T_c was fixed to the c-axis value for the fit to the basal-plane data. Errors in the least significant digits are indicated in parentheses. T_c is measured in K and the parameters A, B, and C are in arbitrary units.

Direction	Function	Parameters
c axis	At^{γ}	$A=1120(50)$
		$T_c=293.57(2)$
		$\gamma=1.23(2)$
Basal plane	$B+Ct^{y}$	$B=0.52(6)$
		$C=292(37)$
		$T_c=293.57$
		$y=1.01(3)$

and Hohenemser[22] on perturbed γ-γ angular correlation experiments on a single crystal of Gd above T_c. Models of critical dynamics based on isotropic spin fluctuations were found to fail below $t \approx 3\times 10^{-3}$. On the other hand, their experimental results below $t \approx 1\times 10^{-3}$ were well described by an anisotropic spin-fluctuation model. This corresponds to an absolute anisotropy temperature for this dynamical property in the range of 0.29 to 0.88 K, in good correspondence with our results for static properties.

The physical mechanism which causes this anisotropy above T_c is obviously relevant to the question of what picks out the c axis as the unique easy axis of magnetization below T_c. This question was recently discussed by Fujiki, De'Bell, and Geldart[23] in terms of mean-field shifts in transition temperatures induced by magnetic dipole-dipole interactions. By numerical evaluation of the appropriate hcp lattice sums as a function of the c/a ratio, dipolar interactions were found to favor the c axis as the easy direction for values of c/a *less* than the ideal value of $(8/3)^{1/2}=1.633$. To be precise, dipolar interactions raise the mean-field transition temperature above the corresponding isotropic short-range exchange value of T_{c0} for any direction of ordering, but the shift ΔT_{c0} is larger for ordering along the c axis ($\Delta T_{c0}^{c}=1.713$ K) than for ordering in the basal plane ($\Delta T_{c0}^{b}=1.633$ K) for $c/a \approx 1.59$ as appropriate for Gd near T_c. We see that the combined result of dipolar interactions and lattice anisotropy introduces *two* new temperature scales in addition to T_{c0}. The average (essentially isotropic) effect of dipolar interactions carries the mean-field scale $\Delta T_{\rm iso}=(\Delta T_{c0}^{c}+\Delta T_{c0}^{b})/2 \approx 1.673$ K (which is very close to the value of 1.674 K we obtain by the method of Fisher and Aharony[24] assuming full isotropy). The anisotropic effect of the dipolar interactions has the mean-field temperature scale $\Delta T_{\rm anis}=\Delta T_{c0}^{c}-\Delta T_{c0}^{b}\approx 0.080$ K.

In order to include the effects of fluctuations so as to obtain physically correct estimates of these temperature scales, we follow Fisher and Aharony[24] in defining dimensionless coupling constants $g_{\rm anis}=\Delta T_{\rm anis}/T_{c0}$ and $g_{\rm iso}=\Delta T_{\rm iso}/T_{c0}$. Wherever possible, we prefer to use experimental data for Gd so as to reduce model-dependent estimates. We thus estimate T_{c0} by the paramagnetic Curie temperature $\theta_p=317\pm 3$ K.[11] The coupling constants are then $g_{\rm anis}=2.52\times 10^{-4}$ and $g_{\rm iso}=5.28\times 10^{-3}$. Corresponding crossover temperatures from a higher-temperature regime to a lower-temperature regime are then defined by $t_{\rm anis}^{x}=g_{\rm anis}^{1/\phi_D}$ and $t_{\rm iso}^{x}=g_{\rm iso}^{1/\phi_H}$ where, to second order in the ϵ expansion, $\phi_D \approx 1.28$ is the exponent for crossover from the isotropic dipolar regime and $\phi_H \approx 1.365$ is the exponent for crossover from the isotropic regime with only short-range exchange interactions.[17-19] Crossover temperatures are thus $t_{\rm anis}^{x}\approx 1.52\times 10^{-3}$, $t_{\rm iso}^{x}\approx 2.15\times 10^{-2}$ on a reduced scale and $\Delta T_{\rm anis}^{x}\approx 0.45$ K, $\Delta T_{\rm iso}^{x}\approx 6.30$ K on an absolute scale.

A number of conclusions are immediate. (1) Dipolar effects are present throughout the range of the present experiment. (2) The theoretical estimate of the scale of anisotropy ($\Delta T_{\rm anis}^{x}\approx 0.45$ K) is in excellent agreement with the experimental values ($\Delta T_{\rm anis}\approx 0.57\pm 0.09$ K or $\Delta T_{\rm anis}'\approx 0.52\pm 0.05$ K). Note that small nondipolar contributions may also be present but that dipolar effects alone are sufficient to account for the experimental results. (3) The asymptotic critical regime is of uniaxial (Ising type) symmetry with dipolar interactions playing an important role. However, it should be noted that logarithmic corrections characteristic of three-dimensional uniaxial dipolar magnets[17-19] have not yet been observed experimentally in Gd. (4) The nonasymptotic critical regime is very complex. We emphasize in this regard that the above crossover temperatures only set the scales of the midpoints of crossover regions and that there are no abrupt effects. In fact, several decades may typically be required to complete a simple crossover.[17-19] It is clear that the range of the present experiment (and most others reported on Gd) is influenced by a pattern of overlapping crossovers. Analysis of data in terms of power laws therefore will generally yield effective exponents and not asymptotic exponents even though corrections to asymptotic scaling may be included in the fitting procedures.

Finally, we turn to the interpretation of the temperature dependence of $\chi_c^{-1}(T)$ and $\chi_b^{-1}(T)$ in terms of effective exponents over the present range $4\times 10^{-4} < t < 1.3\times 10^{-2}$. The value of $\gamma_{\rm eff}=1.23\pm 0.02$ (Table I) is in good agreement with our earlier study of $\chi_c^{-1}(T)$ on a single crystal of comparable quality.[9] It is clear from the above discussion that the closeness of this $\gamma_{\rm eff}$ to the pure (short-range interaction) Ising value is fortuitous. To second order in the ϵ expansion,[17-19] $\gamma_I \approx 1.24$, while a more precise value is $\gamma_I=1.241\pm 0.002$.[25] Turning to $\chi_b^{-1}(T)$, its temperature dependence is expected to reflect that of the internal energy on general grounds.[20,21] Expansions in $1/N$ and ϵ confirm this.[26-29] We thus interpret $y=1.01\pm 0.03$ (Table I) and $1-\alpha_{\rm eff}$ so $\alpha_{\rm eff}=-0.01\pm 0.03$. (In general, a term linear in t is also present but it has coalesced with $t^{1-\alpha_{\rm eff}}$ in view of the smallness of $\alpha_{\rm eff}$.) This $\alpha_{\rm eff}$ is consistent with the results of a recent resistivity measurement on a single crystal of high quality of the same origin in the range $4\times 10^{-5} \lesssim |t| \lesssim 10^{-3}$; the leading temperature dependence was linear (but with a change of slope at T_c).[30] An early specific-heat study (together with references to other work) was made by Simons and Salamon, who found that $\alpha=-0.20\pm 0.02$ provided a consistent description of their data.[6] In a more recent study in the range $8\times 10^{-4} < |t| < 2\times 10^{-2}$ on a single crystal of higher quality (comparable to the sample used in the present work), Lancaster *et al.* considered a number of fitting procedures.[31] Their best fit was obtained for $\alpha \approx -0.30$ (error bars were not specified) but this result was based on assuming a discontinuity at T_c of the specific heat [their Eq. (3)]. If we assume a continuous specific heat and seek an effective exponent, then we should choose results based on their Eq. (1) which gives

$\alpha_{\rm eff}\approx -0.02$.[31] Finally, we note that a rough consistency check on effective critical exponents can be given since it is known that thermodynamic scaling laws for effective exponents are correct to zeroth order in the ϵ expansion so that, in particular, $\alpha_{\rm eff}+2\beta_{\rm eff}+\gamma_{\rm eff}\approx 2$.[32,33] An estimate of $\beta_{\rm eff}$ is available from the work of Chowdhury, Collins, and Hohenemser, who analyzed their hyperfine field data in several interesting ways. Their fits with a single power law (without corrections to scaling) in the range of $1.1\times 10^{-3}\lesssim |t| \lesssim 10^{-2}$ gave $\beta_{\rm eff}=0.41\pm 0.02$ from which we obtain an independent estimate $y=1-\alpha_{\rm eff}=\gamma_{\rm eff}+2\beta_{\rm eff}-1=1.05\pm 0.04$ which is in good agreement with our other determinations.

In summary, we have measured $\chi_c(T)$ and $\chi_b(T)$ on the same high-quality single crystal of Gd and observe uniaxial anisotropy on a temperature scale $\Delta T_{\rm anis}\approx 0.5$ K. Magnetic dipole-dipole interactions have been shown to account for the uniaxial anisotropy and also to lead to a pattern of overlapping crossover regions. The present experiment and virtually all others reported thus far on critical properties of Gd must be interpreted in terms of effective critical exponents for which a consistent assignment has been given here.

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[a]Present address: Department of Physics, University of Ottawa, Ottawa, Ontario, Canada K1N 6N5.

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