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Critical magnetic susceptibility of gadolinium

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An extensive analysis of ac magnetic susceptibility measurements of single-crystal gadolinium is presented. The demagnetization-corrected c-axis data are analyzed on the basis of a power law of the form $\chi_c=At^{-\gamma}$ where t is the reduced temperature above T_c. Our results yield effective values of T_c=293.57 K and γ=1.327. The basal plane susceptibility is expressed as a parametric equation in terms of the c-axis susceptibility $\chi_{ab}^{-1}=B'+C'\chi_c^{-y/\gamma}$. The exponent y, which is related to the critical exponent associated with the specific heat α as $y=1-\alpha$, is determined by our analysis to be y=1.01(2). This gives a temperature scale associated with the anisotropy as $t_{\text{anis}}=2.0\times10^{-3}$. These results are interpreted in the context of the effects of dipolar interactions in the critical region.

I. INTRODUCTION

The critical magnetic properties of gadolinium have recently been reported by several authors.[1–8] Although early measurements on Gd[9–12] led to certain ambiguities concerning the proper universality class for this ferromagnet, recent studies using a variety of experimental techniques have provided a consistent picture of the critical behavior which has a firm foundation on modern theoretical predictions. This behavior has been shown to be complex and to be dominated in the critical region by the presence of magnetic dipole–dipole interactions. The recent analysis of basal plane ac susceptibility by Stetter *et al.*[8] is contrary to this interpretation and these authors have suggested the Gd exhibits three-dimensional Ising behavior and that sample imperfections limit the asymptotic critical behavior. It is the purpose of the present investigation to consider the reasons for these apparent inconsistencies, to provide a new analysis of experimental results and to interpret these data in the context of recent theoretical predictions in order to provide a consistent and comprehensive understanding of the static critical magnetic behavior of gadolinium.

II. EXPERIMENTAL METHODS AND DATA ANALYSIS

A single crystal of high purity gadolinium with a resistivity ratio of ρ(295 K/ρ(4.2 K)=156 was cut into the form of a cube with an edge length 0.249±0.005 cm and the c-axis oriented along one of the cube edges. Further details of the sample preparation have been reported in Ref. 6. Measurements of the ac susceptibility have been performed[6,7] for two crystallographic orientations; with the c axis along the applied ac field direction and with one of the basal plane axes along the field direction.

The c-axis susceptibility in the critical region is defined for $t\to0^+$ by the power law of the form

$$\chi_c=At^{-\gamma}, \tag{1}$$

where the reduced temperature t is defined in terms if the Curie temperature T_c as

$$t=\left|\frac{T_c-T}{T_c}\right|. \tag{2}$$

In contrast to the c-axis susceptibility which diverges as T_c is approached from above, the basal plane susceptibility remains finite and is described by

$$\chi_{ab}^{-1}=B+Ct^y, \tag{3}$$

where the exponent y is related to the critical exponent for the specific heat α by[7]

$$y=1-\alpha. \tag{4}$$

The susceptibilities given in the above expressions are intrinsic quantities and are independent of sample geometry. The actual measured susceptibilities are the demagnetization limited external quantities and are related to the intrinsic quantities by the demagnetization factor N as, e.g.,

$$\frac{1}{\chi_{c,\text{ext}}}=\frac{1}{\chi_{c,\text{int}}}+N \tag{5}$$

and similarly for the basal plane susceptibility. Since, in our experimental measurements the sample geometry was chosen to be cubic, the demagnetization factors for the c-axis and the basal plane susceptibility were identical. Equation (5) may be used in conjunction with Eqs. (1) and (3) to express the extrinsic quantities as

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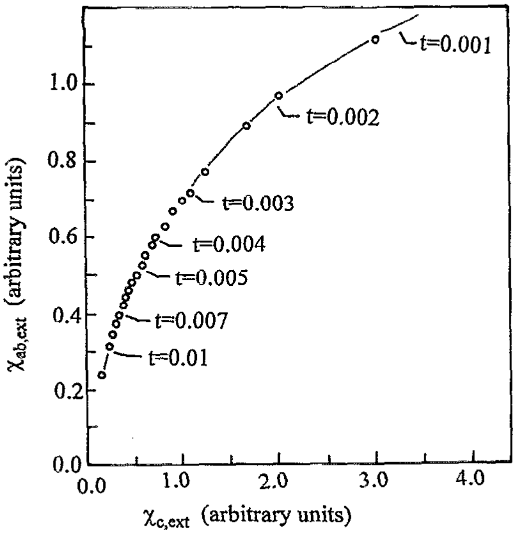

FIG. 1. Measured basal plane susceptibility as a function of measured c-axis susceptibility. Each point represents measurements of the susceptibility along the two directions at the same temperature. The reduced temperature scale as obtained from a fit to T_c for the c-axis data is indicated in the figure.

$$\chi_{c,\text{ext}}^{-1}=A^{-1}t^{y}+N, \tag{6}$$

and

$$\chi_{ab,\text{ext}}^{-1}=B+Ct^{y}+N. \tag{7}$$

In these expressions the coefficients A, B, and C and the exponents γ and y are fitted parameters. While the Curie temperature may be obtained by independent methods and the demagnetization factor may be estimated on the basis of geometric considerations, it is essential to determine the values of the parameters in Eqs. (6) and (7) in a consistent manner. It is clear from the form of Eq. (7) that the values of B and N cannot be determined independently from the basal plane data. It should also be noted that since $|\alpha| \ll 1$, then $y \approx 1$ and the value of C is highly correlated to the value of $B+N$. Because N is the same in Eqs. (6) and (7) it is possible to combine these two expressions and to fit the c axis and basal plane data simultaneously. Solving both expressions for t gives

$$\chi_{ab,\text{ext}}^{-1}=B'+C'\left[\frac{\chi_{c,\text{ext}}^{-1}-N}{N}\right]^{y/\gamma}, \tag{8}$$

where B' is given by $B+N$ and C' may be expressed in terms of C and A. This expression is valid for a given value of t and requires that experimental measurements for the c axis and basal plane orientations be made for the same values of temperature (as is the case for our measurements). This expression allows B', γ, and y to be determined in a consistent manner for both the c axis and basal plane data without the need to know T_c. Fitted values of these parameters may then be used to obtain T_c from a fit to Eq. (1) for the c-axis data.

TABLE I. Fitted parameter values for the critical susceptibility of gadolinium.

Parameter	Value	Uncertainty
A	1728	1.0
B	0.524	0.005
N	3.84	0.02
γ	1.327	0.002
y	1.01	0.01
T_c	293.57	0.02

III. RESULTS AND DISCUSSION

Measured basal plane susceptibility as a function of c-axis susceptibility measured at the same temperature is illustrated in Fig. 1. Parameters obtained from least-square fits to Eqs. (1) and (8) are given in Table I along with uncertainties based on a statistical analysis of the fits.

Although the use of a cubic sample in our studies results in a large demagnetization correction, it is precisely the use of this geometry which allows for a direct comparison of the c-axis and basal plane behavior in the critical region without the need to apply these corrections. Perhaps the ideal sample geometry in this respect would be a sphere. However, the likelihood of introducing additional defects during the machining is an important consideration in the choice of a cubic sample. The anisotropic nature of the critical behavior is readily apparent from our measurements for reduced temperatures less than about 10^{-2} and this, in itself, provides evidence to support our choice of the model based on dipolar interactions as described above for the analysis of these data. A direct measurement of the importance of the dipolar interactions may be obtained on the basis of the above treatment. A reduced temperature scale for the dipolar interactions is expressed in terms of the coefficients in Eqs. (2) and (3) as[7]

$$t_{\text{anis}}=(B/A)^{1/\gamma}. \tag{9}$$

Using the values of these parameters as given in Table I, t_{anis} is found to be 2.2×10^{-3}. This corresponds to an actual temperature difference between T_{anis} and T_c of 0.65 ± 0.10 K. This is in good agreement with theoretical predictions[7,13,14] for the crossover temperature to anisotropic dipolar behavior in this system; $\Delta T=T_{\text{anis}}-T_c=0.45$ K, and previous investigations involving an independent analysis of c-axis and basal plane susceptibility data.

In a comparison of recent experimental studies of Gd it is essential to consider differences in sample quality and geometry and their effects on measured magnetic properties. Of particular relevance in this respect is the relationship of the sample used in the recent study by Stetter *et al.*[8] and that used for our own investigation. Stetter *et al.*[8] have used a 300 Å film grown with the Gd basal plane parallel to the (110) surface of a tungsten substrate. It is clear from the annealing studies reported by these authors that their as-deposited films show magnetic behavior which is severely influenced by the presence of defects. Even after annealing at 870 K these authors base their data analysis on a model involving the existence of imperfections which cause a spread in Curie temperatures of 1 K. No further improvement in the sharpness of the transition was observed for higher temperature anneals. Stetter *et al.*[8] have suggested that the behavior which we attribute to dipolar effects may actually be the result of sample imperfections as they occur on a temperature scale comparable to the T_c smearing observed in the thin-film experiment. The following evidence would seem to be against this interpretation for the following reasons:

(1) The samples used in our studies and in those of Stetter *et al.*[8] have been prepared by different methods and it would be highly coincidental if the level and type of imperfections in the two samples yielded precisely the same influence on the magnetic properties.

(2) There is no evidence on the basis of our resistivity studies of single-crystal Gd of any smearing of T_c on a reduced temperature scale of about 10^{-4} (see Ref. 5), or in the c-axis ac susceptibility data reported previously.[6,7] This is also the case for other studies of the magnetic properties of bulk Gd samples reported in the literature.[1–4]

Our own studies,[6,7] as well as those of others, e.g., Hohenemser and co-workers,[1–4] indicate that the critical temperature region of Gd exhibits complex crossover behavior over an experimentally observable range of reduced temperatures and it is not likely that measured critical properties will be asymptotic. Instead, critical exponents derived from studies of Gd should be treated as effective exponents for the particular range of reduced temperatures studied and that the values of these exponents will inevitably be dominated by anisotropic effects which result from the presence of dipolar interactions. This behavior of critical properties has been shown to result from a crossover pattern from exchange dominated Heisenberg behavior far above T_c to isotropic dipolar to anisotropic dipolar close to T_c.

The present analysis provides additional evidence for the behavior described above and indicates that, by the proper choice of sample geometry and measurement of data at proper temperature intervals, meaningful results which demonstrate the anisotropic dipolar character of gadolinium can be obtained without the need for an independent knowledge of the Curie temperature or the sample demagnetization factor.

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