Critical magnetic susceptibility of gadolinium

P. Hargraves

Department of Physics, Dalhousie University, Halifax, Nova Scotia, Canada B3H 3J5

R. A. Dunlap*

*Department of Materials Science and Engineering, Massachusetts Institute of Technology,
Cambridge, Massachusetts 02139*

D. J. W. Geldart and S. P. Ritcey

Department of Physics, Dalhousie University, Halifax, Nova Scotia, Canada B3H 3J5

The magnetic suspectibility of a single crystal of high-purity gadolinium has been measured in the critical region with use of ac susceptibility methods. For reduced temperatures $t \gtrsim 10^{-3}$, the data are described by a simple power law of the form $\chi = At^{-\gamma}$ with an exponent $\gamma = 1.22(2)$ and a Curie temperature $T_c = 293.51(3)$ K. For reduced temperatures $t \lesssim 10^{-3}$, the data cannot be described by a simple power law and indicate the possibility of a crossover to different critical behavior.

I. INTRODUCTION

The universality principle states that the critical exponents associated with a phase transition are determined by the universality class. In the case of static behavior, the universality class is implied by the dimensionality of the lattice (d) and the dimensionality of the order parameter (n). For dynamic behavior, each static universality class divides into separate dynamic subclasses which are differentiated by the details of the magnetic interactions. It should, therefore, be possible to determine the universality class and hence understand the physics of the magnetic interactions present on the basis of measured critical exponents. While this is a successful approach for many materials, gadolinium presents certain problems. Some measured exponents seem to clearly indicate Heisenberg behavior $(n = 3)$ while other measured exponents seem to indicate Ising behavior $(n = 1)$. An analysis of the physical properties of Gd does not help to resolve the question. On one hand, the S-state nature of the Gd moments suggests that the behavior should be isotropic and Heisenberg-like; on the other hand, the crystalline anisotropy implies the possibility of Ising-like behavior close to T_c. Many of the published critical exponent values for Gd have resulted from data which are not in the asymptotic regime, data which have been analyzed in a manner that does not take into account possibilities of nonasymptotic behavior, or data obtained in relatively low-purity polycrystalline samples. The recent investigation of the exponent β by Chowdhury, Collins, and Hohenemser[1] has emphasized the necessity of proper data analysis. In the present work we have performed ac susceptibility measurements on a high-purity single crystal of Gd in order to determine the critical behavior of the magnetic suspectibility.

II. EXPERIMENTAL METHODS

The sample of Gd used in the present study was an electrotransport-purified single crystal of approximate dimensions $2 \times 2 \times 9$ mm^3 and was oriented with the c axis along the long dimension. This sample was prepared by the Ames Laboratory, Energy and Mineral Resources Research Institute, and has been characterized by previous resistivity studies.[2] It shows a resistivity ratio of $R(295 \text{ K})/R(4.2 \text{ K}) = 156$, and, prior to the susceptibility measurements, was electrochemically etched.[3]

The ac susceptibility measurements were performed using a modified Hartshorn bridge-type susceptometer.[4,5] The drive field was 1.6 A/m at a frequency of 3000 Hz. The intrinsic susceptibility was obtained from the real part of the demagnetization-limited susceptibility, as described previously.[6]

The temperature was measured and controlled by a Chromel-Constanton thermocouple with relative accuracy and stability over the measurement time of about 20 mK.

The possibility of effects due to domain-wall pinning is investigated by applying a second (larger) ac magnetic field at a different frequency.[7] If the measured χ is compromised because of domain-wall pinning, the application of the second field will partially unpin the walls and an enhancement of χ will be observed. This effect has been reported by Wantenaar et al.[7] in polycrystalline Gd beginning at about $T_c + 2$ K. We have seen similar behavior in amorphous alloys.[5] In addition to the present measurements, we have investigated a sample of Gd powder of fairly low purity. In this case, the domain-wall nucleation was observed beginning at about $T_c + 3$ K. For a high-purity single crystal studied in the present work, domain-wall pinning was observed, beginning at about $T_c + 1.5$ K before the crystal was electropolished. However, after

electropolishing, no enhancement of χ in a second applied field was observed at any measured temperature. This suggests that domain walls were pinned by surface features or impurities prior to electropolishing, but that these effects were not a factor in the polished crystal.

III. RESULTS AND DISCUSSION

Data were analyzed in terms of a simple power law of the form

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

where the reduced temperature $t = (T - T_c)/T_c > 0$. Subranges of the data containing ten data points were fitted to expression (1) in order to obtain an effective exponent γ_{eff} as a function of median t value. γ_{eff} is shown for different choices of T_c in Fig. 1. In all cases, the values of γ_{eff} show a sharp decrease for $t \lesssim 10^{-3}$. This point corresponds to data with $T \lesssim 293.77$ K. From this analysis it is reasonable to assume that data for $T \gtrsim 293.77$ K can be fitted to a single power law. Using Eq. (1) for data obtained here with 293.77 K $\le T \le 295.93$ K, we have performed a nonlinear least-squares fit allowing A, T_c, and γ to be free parameters. This fit yields $T_c = 293.51 \pm 0.02$ K and $\gamma = 1.22 \pm 0.01$. Plots of residuals (measured χ-fitted χ) appeared to show a random distribution of deviations. Previously reported values of the exponent γ obtained from fits to a simple power law are given in Table I. The present result is in good agreement with previous values. We note that the previous data are reported for minimum reduced temperatures $t > 10^{-3}$ and it is only for reduced temperatures smaller than this, as indicated in Fig. 1, that our data deviate from this power law. We have made χ measurements on a single crystal of lower purity Gd and have found a deviation from power-law behavior occurring at the same value of $T - T_c$. This indicates that the observed effect is not a result of impurities.

Chowdhury et al.[1] have suggested, on the basis of measurements of the exponent β, that corrections to scaling may be important in the description of the critical properties of Gd. We have analyzed the present data with $T \ge 293.77$ K with an expression of the form
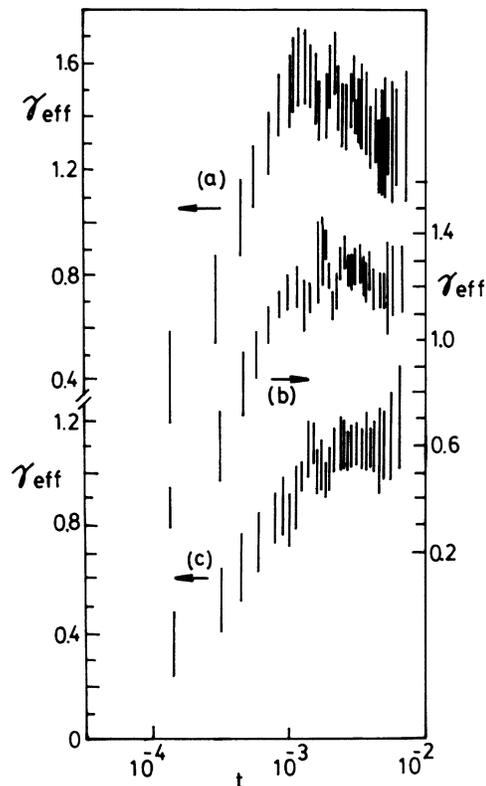
$$\chi = At^{-\gamma}(1 + bt^{\Delta}) \tag{2}$$

FIG. 1. The effective component γ_{eff} plotted as a function of t for (a) $T_c = 293.41$ K, (b) $T_c = 293.51$ K, and (c) $T_c = 293.61$ K.

in order to investigate the possibility of these effects. No appreciable improvement in the goodness of fit (χ^2) could be obtained using Eq. (2), with Δ fixed to the theoretical value of 0.54,[11] over that obtained with the power law of Eq. (1).

The deviation of the critical behavior of χ from $t \lesssim 10^{-3}$ as illustrated in Fig. 1, can indicate the possibility of a crossover in the critical properties. It has been suggested by Geldart, De'Bell, Cook, and Laubitz,[2] on the basis of resistivity studies of Gd, that the critical behavior close to T_c may be dominated by dipolar interactions. Hohenemser and co-workers[12-14] have observed similar effects in perturbed angular correlation and Mössbauer

TABLE I. Selected values of the exponent γ from Gd taken from the literature. Values in ( ) are the errors in the least significant digit(s).

Sample	Method	T_c (K)	γ	Range of t	Reference
Single crystal	Magnetization + Eq. (1)	292.5(5)	1.33	0.0014–0.2	8
Single crystal	Magnetization + Eq. (1)	293.3(1)	1.196(3)	0.0045–0.058	9
Single crystal	Magnetization + scaling equation of state	292.05(15)	1.25(10)	0.0033–0.2	10
Polycrystal	ac susceptibility + Kouvel-Fisher	291.1(1)	1.24(3)	0.0099–0.037	7

effect experiments. The appropriate form of χ for a $d=3$ uniaxial dipolar system, in the limit of small t $(t>0)$ is[15,16]

$$\chi = Ct^{-1}|\ln t|^{1/3}[1+O((\ln|\ln t|)/(\ln t))] , \qquad (3)$$

or in a modified form which includes approximately higher-order logarithmic corrections[17]

$$\chi = At^{-1}|\ln(t/t_0)|^{-1/3} , \qquad (4)$$

where t_0 is a dimensionless parameter of order unity.

We did not attempt nonlinear least-squares fits of data to Eq. (4), since it is clear at the outset that satisfactory fits will not be found. It is easy to see from simple numerical tests that the $\gamma_{\text{eff}}(t)$ for data described by Eq. (4) must approach unity for small t; i.e., $\gamma_{\text{eff}}(t \to 0)=1$, as expected. This fails to occur only if T_c has been wrongly determined. It is clear from Fig. 1 that $\gamma_{\text{eff}}(t)$ does not approach unity at small t for the indicated choices of T_c. Moreover, it has not been found possible to bring about $\gamma_{\text{eff}}(t \to 0)=1$ by varying T_c over a reasonable range. Of course, there are corrections to Eq. (4) and certainly complications due to crossovers, so it is difficult to draw completely firm conclusions.[18]

While the exponent β as measured in Gd is consistent with the usual Heisenberg value,[11] the commonly reported values of γ are anomalously low, e.g., near 1.20–1.24. The present measurements for $t \lesssim 10^{-3}$ seem to suggest

that what may appear in susceptibility data to be an asymptotic critical region for $t \gtrsim 10^{-3}$ with $\gamma \sim 1.2$ may, in fact, be indicative of very complex nonasymptotic behavior.

In conclusion, we observe a distinction in the critical susceptibility of Gd for reduced temperatures above and below $\sim 10^{-3}$. Above 10^{-3} the data may follow a single power law with an effective exponent $\gamma = 1.22(1)$. This is consistent with previously reported values of γ for similar reduced temperature ranges, but we suggest that this effective exponent value should not be interpreted as an asymptotic value. For $t \lesssim 10^{-3}$, the behavior of χ cannot be described by a power law and is also not well described by the asymptotic temperature dependence expected for a uniaxial dipolar behavior.

ACKNOWLEDGMENTS

*On leave from Department of Physics, Dalhousie University, Halifax, Nova Scotia, Canada B3H 3J5.

[1] A. R. Chowdhury, G. S. Collins, and C. Hohenemser, Phys. Rev. B **33**, 6231 (1986).

[2] D. J. W. Geldart, K. De'Bell, J. Cook, and M. J. Laubitz, Phys. Rev. B **35**, 8876 (1987).

[3] B. Beaudry and K. A. Gschneidner, Jr., in *Handbook on the Physics and Chemistry of Rare Earths*, edited by K. A. Gscheidner and L. Eyring (North-Holland, New York, 1978), Vol. 1, p. 173.

[4] C. M. Brobeck, R. R. Burkey, and J. T. Hoeksema, Rev. Sci. Instrum. **49**, 1279 (1978).

[5] P. Hargraves and R. A. Dunlap, J. Phys. F **18**, 553 (1988).

[6] R. A. Dunlap and A. M. Gottlieb, Phys. Rev. B **22**, 3422 (1980).

[7] G. H. J. Wantenaar, S. J. Campbell, D. H. Chaplin, T. J. McKenna, and G. V. H. Wilson, J. Phys. C **13**, L863 (1980).

[8] C. D. Graham, J. Appl. Phys. **36**, 1135 (1965).

[9] M. N. Deschizeaux and G. Develey, J. Phys. (Paris) **32**, 319 (1971).

[10] M. Vincentini-Missoni, R. I. Joseph, M. S. Green, and J. M. L. Sengers, Phys. Rev. B **1**, 2312 (1970).

[11] J. C. LeGuillou and J. Zinn-Justin, Phys. Rev. Lett. **39**, 95 (1977).

[12] G. S. Collins, A. R. Chowdhury, and C. Hohenemser, Phys. Rev. B **33**, 4747 (1986).

[13] A. R. Chowdhury, G. S. Collins, and C. Hohenemser, Phys. Rev. B **30**, 6277 (1984).

[14] A. R. Chowdhury, G. S. Collins, and C. Hohenemser, Phys. Rev. B **33**, 5070 (1986).

[15] A. I. Larkin and D. E. Khmel'nitskii, Zh. Eksp. Teor. Fiz. **57**, 2087 (1969) [Sov. Phys. JETP **29**, 1123 (1969)].

[16] E. Brézin and J. Zinn-Justin, Phys. Rev. B **13**, 251 (1976).

[17] A. Aharony and B. I. Halperin, Phys. Rev. Lett. **35**, 1308 (1975).

[18] See the review by A. D. Bruce, Adv. Phys. **29**, 111 (1980).