Static universality class implied by the critical exponents of Gd

Ataur R. Chowdhury,* Gary S. Collins,† and Christoph Hohenemser
Department of Physics, Clark University, Worcester, Massachusetts 01610
(Received 4 November 1985)

Structural characteristics and available critical exponent measurements fail to distinguish clearly between Ising and Heisenberg critical behavior in Gd. In this context we present a new perturbed $\gamma\gamma$-angular correlation measurement of the exponent β which appears to resolve the ambiguity. From our result, $\beta=0.399(16)$, we conclude that Gd is described by Heisenberg critical behavior for reduced temperatures $\epsilon > 10^{-3}$. This does not exclude crossover to another value for smaller ϵ.

According to the universality principle, static critical exponents are uniquely determined by lattice dimensionality d and order-parameter dimensionality n. This principle has been repeatedly tested by asking the question: For materials with given d and n, are critical exponents the same, and do their values agree with increasingly accurate theoretical predictions? When care is taken to eliminate nonasymptotic data, agreement between experiment and theory is quite good.[1]

With maturing of the field one may *assume* universality and ask a second question: Given the exponent values for a specific material, what is implied about its universality class? Asking the question in this way can highlight experimental inconsistencies and provide insights into a material's structure that are not apparent from other physical characteristics.

In the present paper we ask this second question for Gd. Specifically, is Gd a $d=3$ Ising or a Heisenberg ferromagnet, i.e., is n 1 or 3? Structural characteristics and past exponent measurements provide inconsistent answers. To help resolve the issue we present a new measurement of the exponent β which is more nearly asymptotic than earlier work.

Consider Gd: On the one hand, it is an S-state ion which should have isotropic spin interactions, and like Fe and Ni, exhibit Heisenberg critical behavior. On the other hand, it is noncubic, with spin alignment along the c axis below T_c, suggesting that, like MnF_2, it will exhibit Ising critical behavior. Ising behavior is also suggested by the observation of Ising-like domain walls near T_c.[2]

Selected experimental exponent values[3–11] (Table I) span both the Heisenberg and Ising model predictions[12] (Table II). Values of α suggest Heisenberg behavior by their sign, but are generally twice as large as predicted. Values of β cluster around 0.37 but also include 0.31 and thus span both predictions. Values of γ are uniformly supportive of Ising behavior, while values of δ are generally too low for either prediction. Inconsistency between the measurements is indicated by failure of the scaling law $\alpha+2\beta+\gamma=2$.

We believe that nonasymptotic data are a principal source of the trouble in Table I. With the exception of one α value, none of the measurements spans a full decade within the reduced temperature range $|\epsilon| = |1-T/T_c| < 10^{-2}$, and none are analyzed in terms of correction to scaling. Because asymptotic critical behavior is generally restricted[1] to $|\epsilon| < 10^{-2}$, this can lead to substantial systematic errors and apparent violation of scaling.

It is possible that for most of the experimentally accessible region Gd may be in a transition region between isotropic Heisenberg and $d=3$ Ising behavior—a case that has been described as the anisotropic Heisenberg model. If one may be guided by high-temperature-series results for classical spins on a fcc lattice,[13] this leads to *effective* critical exponents that are intermediate between Heisenberg and Ising fixed points.

In our new experiment on Gd we employed perturbed $\gamma\gamma$-angular correlations (PAC) to measure the hyperfine field $H_{hf}(T)$ at nuclei of dilute ^{111}In solute atoms. Draw-

TABLE I. Selected static critical exponents for Gd. (Values listed were selected from Ref. 3 on the condition that they be based on significant data in the range $|t| < 10^{-2}$.)

Exponent	$10^3 t_{min}$	Method	Reference
α: $-0.09(5)$ α': $-0.32(5)$	1	Specific heat	4
α: $-0.20(2)$ α': $-0.20(2)$	1	Specific heat	5
β: 0.385[a]	1.7	Barkhausen noise	2
β: 0.31(5)	3	Ferromagnetic transmission	6
β: 0.5–0.39	2.7	Magnetization	7
γ: 1.3(1)	2	Magnetization	8,9
β: 0.37(1)[b] γ: 1.25(10) δ: 4.39(10)	2	Magnetization and scaling equation of state	10
β: 0.381(15) γ: 1.196(3) δ: 3.615(15)	4	Magnetization and scaling equation of state	11

[a]Authors do not state error.
[b]Value of γ calculated from scaling law $\gamma=\beta(\delta+1)$.

TABLE II. Theoretical static critical exponents for $d=3$ magnetic systems. [Values of β and γ are from J. C. Le Guillou and J. Zinn-Justin, Phys. Rev. Lett. 39, 95 (1977). Values of α and δ are calculated from β and γ via the scaling relations $(\alpha+2\beta+\gamma)=2$ and $\gamma=\beta(\delta-1)$].

Exponent	Ising model	Heisenberg model
α	+0.110(2)	−0.116(2)
β	+0.325(1)	+0.365(1)
γ	+1.240(1)	+1.387(1)
δ	+4.816(3)	+4.797(3)

ing on evidence that $H_{\rm hf}(T)$ scales like the spontaneous magnetization near T_c (Ref. 14), we determined β via a power-law fit of the data (see below). The advantages of PAC for high-temperature critical phenomena studies in metallic ferromagnets have been extensively noted elsewhere.[14] Chief among them is the possibility for probing the magnetization in zero field without degradation of the signal near T_c.

Our samples were oriented single crystals of Gd (obtained from the Ames Laboratories) into which 10 μ Ci of carrier-free ^{111}In had been diffused. Sources were prepared by depositing ^{111}In dissolved in dilute HCl on the sample surface, evaporating to dryness, and diffusing *in vacuo* for 1–2 hours at 1150 K. Because of the short (2.7d) half life of ^{111}In, its estimated concentration in our samples was $\sim 10^{-9}$.

The sample temperature was controlled by a two-stage thermoelectric module enclosed in a vacuum can. Temperature regulation was achieved via a thermocouple controlled differential voltmeter. Temperature stability was better than ±0.05 K.

PAC spectra were collected with a four counter spectrometer with the sample's c axis perpendicular to the counter plane and were reduced via methods described elsewhere.[15] This leads to a time-dependent perturbation factor $G_2(t)$ which we analyzed in terms of magnetic and electric quadrupole frequencies $\omega_L=\mu H_{\rm hf}/\hbar I$ and $\omega_0=(3\pi/10)e^2Qq_{zz}$, respectively. Here μ, Q, and I are the nuclear magnetic moment, quadrupole moment, and spin, respectively, and q_{zz} is the principal component of the electric field gradient.

As noted in earlier work,[16–18] down to 260 K, the magnetization, $H_{\rm hf}$ and q_{zz}, are approximately colinear along the c axis. This makes possible analysis of the data via the closed form:

$$G_2(t)=S_0+S_{1a}\cos(\omega_0 t)\cos(\omega_L t)+S_{1b}\cos(\omega_0 t)\cos(2\omega_L t)+S_2\cos(2\omega_0 t)\cos(\omega_L t)+S_3\cos(3\omega_0 t)\cos(2\omega_L t)\ . \quad (1)$$

The amplitudes S_0, S_{1a}, S_{1b}, S_2, and S_3 depend on the crystal orientation relative to the detectors. In our experimental geometry, with $H_{\rm hf}$ perpendicular to the counter plane, the terms in $\cos(2\omega_L t)$ dominate,[15] thus increasing the sensitivity near T_c in this geometry.

As can be seen in Fig. 1, the spectra exhibit slow damping. In order to describe this behavior we modified Eq. (1) by introducing separate damping factors for the terms in $\cos(\omega_L t)$ and $\cos(2\omega_L t)$, respectively:

$$G_2(t)=S_0+\exp(-t/\tau_R)\cos(\omega_L t)[S_{1a}\cos(\omega_0 t)+S_2\cos(2\omega_0 t)]+\exp(-2t/\tau_R)\cos(2\omega_L t)[S_{1b}\cos(\omega_0 t)+S_3\cos(3\omega_0 t)]\ , \quad (2)$$

and fitted the data with ω_L, ω_0, τ_R, S_0, S_{1a}, S_2, and S_3 as free parameters. This yielded good fits, but with negative or near-zero amplitudes for terms in $\cos(\omega_L t)$, as expected. We therefore set $S_{1a}=S_2=0$ and obtained even better fits. In the absence of quantitative relaxation theories for the case at hand, the justification of the damping terms in Eq. (2) is entirely empirical: they fit the data.

Our data span a large temperature range above and below T_c. Results for the relaxation rate $1/\tau_R$ versus T, shown in Fig. 2 (top), yield an asymmetrical singularity near T_c which will be considered elsewhere in a paper concerned with spin dynamics.[19] Values for ω_0 versus T are shown in Fig. 2 (middle) and exhibit a transition near T_c which we attribute to a continuous change in the c/a ratio due to magnetostriction.[20] Results for the predominant amplitude S_{1b} are shown in Fig. 2 (bottom) and

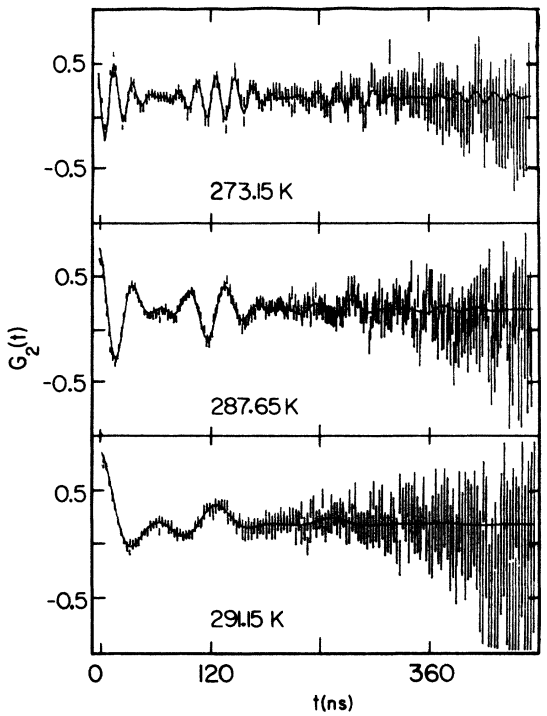

FIG. 1. PAC spectra at selected temperatures below T_c for the sample with its c axis perpendicular to the plane of the counters. Fits to the data were obtained as described in the text.

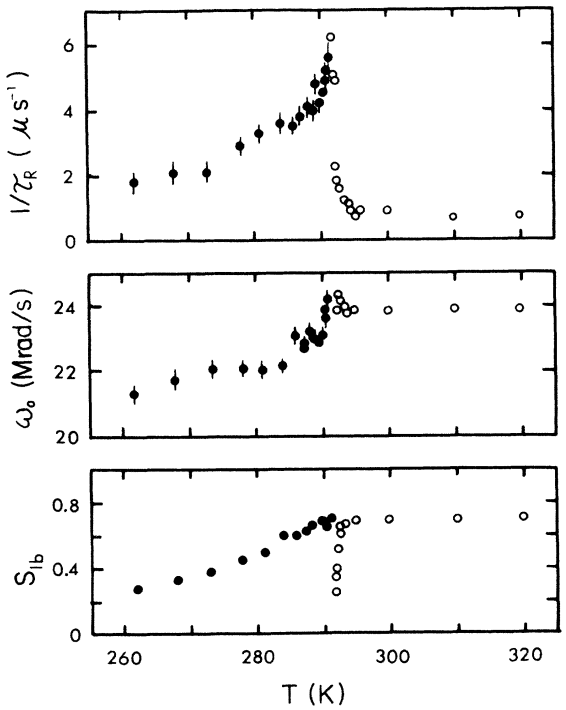

FIG. 2. Temperature dependence of the relaxation rate $1/\tau_R$ (top), the quadrupole frequency ω_0 (middle), and the amplitude S_{1b} (bottom). Interpretations of these parameters are given in the text.

TABLE III. Hyperfine fields.

T^a (K)	ω_L (Mrad/s)	H_{hf} (kG)
291.52	26.2(6)	17.3(4)
291.40	30.7(3)	20.4(2)
291.40	29.9(5)	19.8(2)
291.15	36.7(2)	24.3(1)
291.89	41.0(2)	27.2(1)
290.64	45.3(2)	30.0(1)
290.39	49.9(2)	33.1(1)
290.14	52.9(2)	35.1(1)
289.87	56.0(2)	37.1(1)
289.62	59.3(2)	39.3(1)
289.12	63.6(2)	42.2(1)
288.62	68.8(2)	45.6(1)
287.61	77.5(2)	51.3(1)
287.42	79.4(2)	52.6(1)
286.11	89.1(2)	59.0(1)
284.11	102.2(2)	67.7(1)
281.11	118.5(2)	78.5(1)
277.10	137.3(2)	91.0(2)
273.10	153.6(2)	101.8(2)
268.10	172.5(3)	114.4(2)
262.10	191.8(2)	127.1(2)

[a]Errors in T were ±0.05 K.

show a sharp decline near T_c and a slow decline below T_c. The former defines a region of possible rounding in the transition ($\Delta T_c < 0.2$ K) and has been excluded from further analysis. The latter can be explained by attenuation, which increases as the Larmor period approaches the spectrometer time resolution.

Fitted values of $\omega_L(T)$ and deduced values of $H_{hf}(T)$ are listed in Table III. As a first step toward extracting the exponent β from the variation of $H_{hf}(T)$, we fitted to the power law

$$H_{hf}(T)/H_{hf}(0)=B(1-T/T_c)^{\beta}, \quad (3)$$

with B, T_c, and η treated as free parameters, and $H_{hf}(0)$ taken from the work of Boström *et al.*[18] Asymptotic values of these parameters were obtained by varying the range of temperature included in the fitting,[14] as shown in Fig. 3. For reduced temperatures $\epsilon < 10^{-2}$ we obtain

$$\beta=0.41(2),\ B=0.90(6),\ T_c=291.85(5)\ \text{K}. \quad (4)$$

A logarithmic plot of H_{hf} versus ϵ, employing this T_c value is shown in Fig. 4. Both Figs. 3 and 4 indicate significant deviations between the asymptotic line characterized by the parameters of Eq. (4) and the data for $\epsilon > 10^{-2}$. This indicates that correction to scaling terms are important.

To include them we refitted the data to

$$H_{hf}(T)/H_{hf}(0)=B\epsilon^{\beta}(1+a_m\epsilon^{\Delta}), \quad (5)$$

with $\Delta=0.55$ fixed at its theoretical value,[12] $T_c=291.85$ K fixed at the value derived from fitting Eq. (3) for $\epsilon < 10^{-2}$, and B, β, and a_m free. This yields the result

$$\beta=0.399(5),\ B=0.79(3),\ a_m=0.78(9), \quad (6)$$

indicating that correction to scaling terms are capable of producing significant changes in exponent and amplitude values.

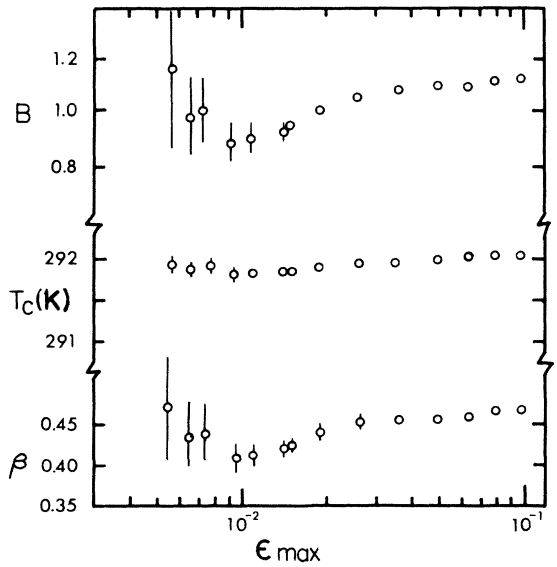

FIG. 3. Range-of-fit analysis for the simple power law given in Eq. (3). Shown are fitted values of B, T_c, and β as the maximum reduced temperature ϵ_{max} included in the fit is varied. Note that as ϵ_{max} is reduced below $\epsilon=10^{-2}$ all three parameters appear to approach asymptotic values.

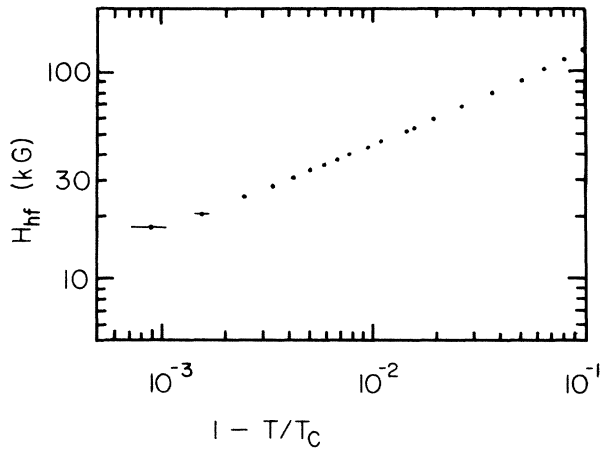

FIG. 4. Logarithmic plot of the hyperfine field as a function of reduced temperature, with $T_c = 291.85(5)$ K.

The errors in (6) are underestimates because they do not reflect variability in T_c. Changing T_c by ± 0.05 K, its estimated uncertainty in the fit to Eq. (3), produces a $\pm 5\%$ change in β and corresponding changes in the other parameters. This leads us to quote the final result:

$$\beta = 0.399(16), \quad B = 0.79(7), \quad a_m = 0.78(20) . \qquad (7)$$

Because it is based on a closer approach to T_c than all previous magnetic measurements, an explicit search for the asymptotic region, and a final fit that includes corrections to scaling, we believe this result is the most reliable magnetic exponent for Gd.

Given the assignment of generous error limits, we can rule out Ising behavior ($\beta = 0.325$) in the temperature region we have sampled. Our result, which is the highest recorded thus far for Gd, is also not consistent with the isotropic Heisenberg model ($\beta = 0.365$). The most plausible explanation is that the presence of large correction to scaling terms have reduced the region of asymptotic Heisenberg behavior so much that our data do not overlap it strongly.

The tradeoff between the value of β and corrections to scaling is illustrated by fixing T_c at 291.75 K, two standard deviations below our best estimate. With this, fitting to Eq. (5) yields $\beta = 0.362(8)$, $B = 0.65(3)$, and $a_m = 1.29(15)$, in excellent agreement with the prediction $\beta = 0.365(1)$. Unfortunately, only substantial data in the region $10^{-4} < \epsilon < 10^{-3}$ could justify this choice of T_c.

In short, we conclude that Gd is not an Ising system in the temperature region we have studied, but exhibits critical behavior that is close to Heisenberg-model predictions. Physically this indicates that, despite uniaxial spin alignment below T_c, isotropic spin fluctuations should prevail in the region of reduced temperatures we have investigated. This is an hypothesis which can be tested by a study of spin dynamics above T_c. Such work has been completed and will be published separately.[19] Neutron scattering experiments probing critical fluctuations would also be of interest.

Help in source preparation and data analysis was received from Carl Allard, Reinhardt Schuhmann, and Nicholas Rosov. Useful critical comments were received from Alfred Kleinhammes. Research support was received from the National Science Foundation under Grant No. DMR 83-03611.

*Present address: Department of Physics, University of Central Florida, Orlando, FL 32816.

†Present address: Department of Physics, Washington State University, Pullman, WA 99164.

[1]R. M. Suter and C. Hohenemser, J. Appl. Phys. **50**, 1814 (1979).

[2]P. Molho and J. L. Porteseil, J. Phys. (Paris) **44**, 83 (1983).

[3]We compiled Table I from K. Stierstad, R. Anders, and W. von Horsten, *Experimental Values of Critical Exponents and Amplitude Ratios at Magnetic Phase Transitions,* No. 20-1 of *Physics Data* (Fach-Informations Zentrum Karlsruhe, Federal Republic of Germany, 1984).

[4]E. A. S. Lewis, Phys. Rev. B **1**, 4368 (1970).

[5]D. S. Simons and M. B. Salamon, Phys. Rev. B **10**, 4680 (1974).

[6]P. Sheng, C. B. Manikopoulos, and T. R. Carver, Phys. Rev. Lett. **30**, 234 (1973).

[7]A. G. A. M. Saleh and N. H. Saunders, J. Magn. Magn. Mater. **29**, 197 (1982).

[8]P. Heller, Rep. Prog. Phys. **30**, 731 (1967).

[9]C. D. Graham, Jr., J. Appl. Phys. **36**, 135 (1965).

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

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

[12]J. C. Le Guillou and J. Zinn-Justin, Phys. Rev. Lett. **39**, 95 (1977). See also J. C. Le Guillou and J. Zinn-Justin, J. Phys. (Paris) Lett. **46**, L137 (1985).

[13]D. Jasnow and M. Wortis, Phys. Rev. **176**, 739 (1968).

[14]C. Hohenemser, T. Kachnowski, and T. K. Bergstresser, Phys. Rev. B **13**, 3154 (1976).

[15]A. R. Arends, C. Hohenemser, F. Pleiter, H. de Waard, L. Chow, and R. M. Suter, Hyperfine Interactions **8**, 191 (1980).

[16]J. W. Cable and E. O. Wollan, Phys. Rev. **165**, 733 (1965).

[17]F. Milstein and L. B. Robinson, Phys. Rev. **177**, 904 (1969).

[18]G. Boström, B. Liljegren, B. Jonsson, and E. Karlson, Phys. Scr. **3**, 175 (1971).

[19]Gary Scott Collins, Ataur R. Chowdhury, and Christoph Hohenemser, Phys. Rev. B **33**, 4747 (1986).

[20]F. J. Darnell, Phys. Rev. **130**, 1825 (1963).