Anomalous critical spin dynamics in Gd: A revision

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We report a correction of the Curie temperature, T_c, for our Mössbauer study of critical spin dynamics in *Gd* ^{161}Dy. The revised T_c value, which is 0.4 K lower than previously reported, leads to spin-correlation times that diverge with an exponent $w=0.49(5)$. Though based on a well-defined power law over reduced temperatures $10^{-3}<t<10^{-1}$, this result remains anomalous: It agrees neither with the predictions for the Heisenberg model nor with those for the Ising model.

In 1984 we published a Mössbauer study of critical slowing down in *Gd* ^{161}Dy in which we asked the following:[1] Does Gd exhibit order-parameter-nonconserving spin dynamics such as the isotropic ferromagnets Fe, Ni, EuO, and EuS? Because of its large localized magnetic moment, and the fact that it is an *S*-state ion, Gd should be a better Heisenberg system than either Fe or Ni, both of which are partly itinerant. On the other hand, since Gd is noncubic, with uniaxial spin alignment along the *c* axis below T_c, it is possible that it exhibits Ising critical behavior. As noted in our earlier paper[1] experimental values of static critical exponents in Gd do not provide a clear-cut distinction between Ising and Heisenberg behavior.

To characterize the spin dynamics of Gd we converted measurements of the critical component of the Mössbauer linewidth to the wave-vector averaged spin autocorrelation time τ_c using the "motional narrowing" form

$$\Delta\Gamma_c=(hc/E_\gamma)C_{\rm hf}^{\rm ME}\tau_c=(8.01\times10^{12}\ {\rm mm/s^2})\tau_c\ , \quad (1)$$

where E_γ is the gamma-ray energy and $C_{\rm hf}^{\rm ME}$ is the hyperfine coupling parameter derivable from Mössbauer linewidth theory.[1] By recourse to the dynamic scaling form of the dynamic structure factor, $S_c(\mathbf{q},\omega)$, we expressed τ_c in terms of the power law

$$\tau_c=D(T/T_c-1)^{-w}\ , \quad (2)$$

where the critical exponent w is given by the scaling law[2]

$$w=\nu(z+2-d-\eta)\ , \quad (3)$$

and where d is the lattice dimensionality and z, ν, and η are critical exponents defined in the usual manner.

Measurements of τ_c versus T could not be fitted with a single power law, but yielded $w=0.28(2)$ and $0.21(3)$, depending on whether the reduced temperature was unrestricted or limited to $t=(T/T_c-1)<10^{-2}$. These values of w, or corresponding values of z obtained via the scaling law of Eq. (3), were recognized as distinctly anomalous because they cannot be explained by either the $d=3$ iso-

TABLE I. Critical exponent predictions for $d=3$ ferromagnets. Values of β, γ, ν, and η were taken from Ref. 3 and represent the most accurate predictions of renormalization-group theory. Values of α were derived via the scaling law $\alpha+2\beta+\gamma=2$. Values of z are based on the predictions $z=\frac{1}{2}(5-\eta)$, $z=2-\eta/2$, and $z=2+\alpha/\nu$ for the three columns left to right, as given in Ref. 4. Values of w were derived via the scaling law $w=\nu(z+2-d-\eta)$.

	Heisenberg model			
Exponent	Spin conserved		Spin nonconserved	Ising model
β		0.3645(25)		0.3250(20)
γ		1.386(4)		1.2410(20)
ν		0.705(3)		0.6300(15)
η		0.033(4)		0.031(4)
α		−0.115(5)		+0.109(5)
z	2.484(2)		1.984(2)	2.173(5)
w	1.023(5)		0.670(5)	0.718(95)

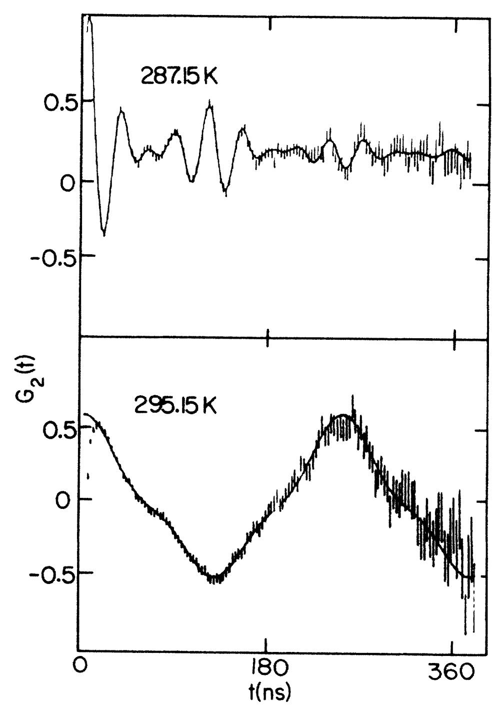

FIG. 1. Typical PAC spectra below (top) and above (bottom) the Curie temperature. Below T_c the spectra may be fitted by a combined magnetic-quadrupole interaction; above T_c the spectra are described by a pure quadrupole interaction. Fitting forms are discussed in Ref. 8.

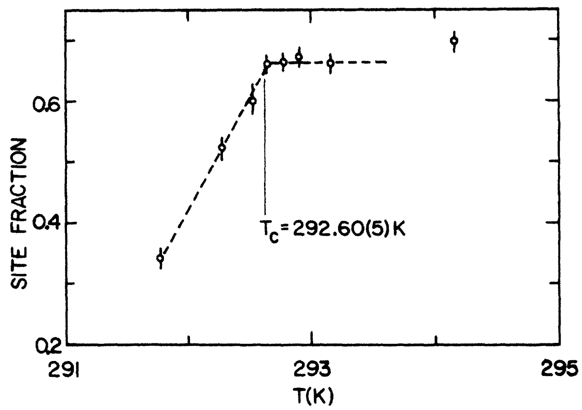

FIG. 2. Root-mean-squared signal amplitudes of PAC spectra for ^{111}In doped, neutron irradiated, and annealed Gd foil used in earlier Mössbauer work (Ref. 1). The sharp break in the amplitudes was used to provide the estimate $T_c = 292.60(5)$ K, independent of Mössbauer experiment.

tropic Heisenberg model or the Ising model (see Table I).[3,4]

In this note we argue that T_c was wrongly fixed in our earlier work, and that a revised value leads to a less puzzling single power law for the divergence of τ_c, with an exponent w that is considerably closer to theoretical expectations.

To understand how the error in T_c was made, and how it can be corrected, consider our methods. The very broad Mössbauer line of *Gd* ^{161}Dy does not provide a reliable way of obtaining T_c, and requires an auxiliary approach. Therefore we doped a small piece of the ^{160}Gd source material with ^{111}In, irradiated and annealed it in the same way as the *Gd* ^{161}Dy Mössbauer source, and conducted perturbed angular correlation (PAC) experiments as a function of temperature. These showed well-defined quadrupole precessions above T_c and a combined magnetic-quadrupole signal below T_c as shown in Fig. 1.[5] Similar results had been obtained earlier by Boström *et al.*[6] We fit all spectra, above and below T_c, with a pure quadrupole signal and noted that the effective site fraction developed a sharp break due to misfitting (Fig. 2), which we interpreted as T_c.

The first indication that this method might be faulty came in recent *Gd* ^{111}In PAC experiments conducted on

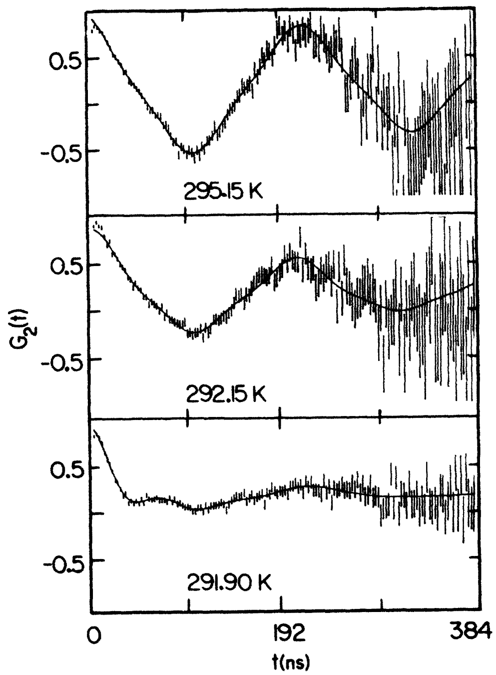

FIG. 4. Typical PAC spectra exhibiting nuclear relaxation above T_c, including least-squares fits used to deduce the nuclear relaxation time.

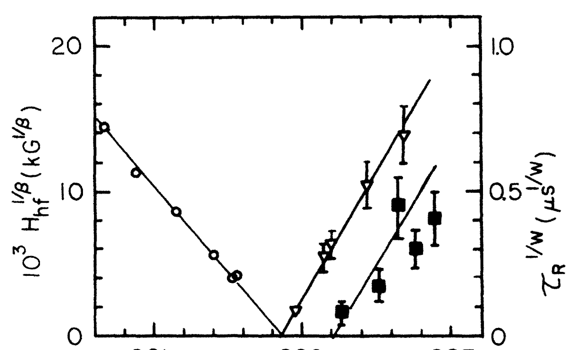

FIG. 3. Determination of T_c via *Gd* ^{111}In PAC data. The data are presented as linearized plots of the hyperfine field below T_c (left scale), and the nuclear relaxation rate above T_c (right scale). The open circles and triangles represent the hyperfine field and nuclear relaxation rate for recently measured single-crystal natural Gd samples, and determine T_c to be 291.85 K by two independent methods. The solid squares represent nuclear relaxation rates obtained for a piece of polycrystalline ^{160}Gd used in the Mössbauer experiments of Ref. 1, and determine T_c to be 292.2(1) K.

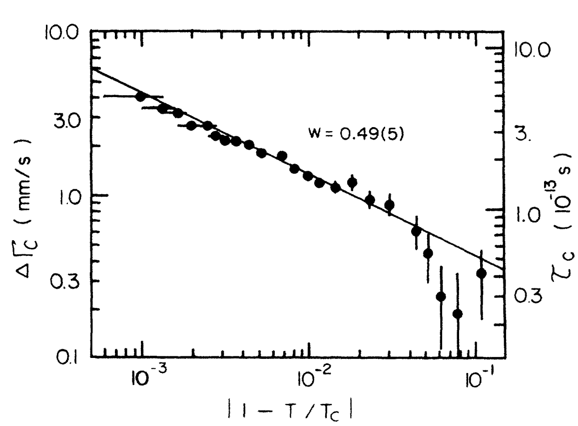

FIG. 5. Revised logarithmic plot of the critical component of the *Gd* ^{161}Dy Mössbauer linewidth as a function of reduced temperature, with T_c fixed at 292.2 K. A least-squares fit to the data yields $w = 0.49(5)$.

TABLE II. Additional points for *Gd* ^{161}Dy.

T (K)	Γ exp (mm/s)	$\Delta\Gamma_c$ (mm/s)	τ_c (10^{-13} s)
292.50	11.24(14)	4.03(17)	5.02(21)
292.60	10.59(15)	3.38(17)	4.21(21)

TABLE III. Sensitivity of w and D to choice of T_c.

T_c (K)	w	D (10^{-13} s)	χ^2
291.10 (fixed)	0.539(21)	0.111(14)	1.02
291.20 (fixed)	0.494(19)	0.133(15)	1.02
291.30 (fixed)	0.441(18)	0.167(18)	1.22
292.15(9)	0.518(47)	0.121(25)	1.05

single crystals below T_c for the purpose of determining the critical exponent β from the variation of the hyperfine field.[7] In analyzing these data via a power law we obtained $T_c=291.85(5)$ K, as shown in Fig. 3, left curve. This is 0.75 K lower than the value obtained via Fig. 2.

A check of this result was obtained through additional PAC measurements above T_c, using the same sample. Here we found that the spectra could be fitted with a temperature-independent quadrupole interaction modulated by a *strongly temperature-dependent relaxation* that had been unnoticed previously (see Fig. 4). We find that the nuclear relaxation time goes to zero at 291.8(1) K, as shown in Fig. 3, middle curve. Assuming that relaxation is caused by critical spin fluctuations, we accept the zero intercept of the relaxation rate as a second, independent way of determining T_c.

Because these results were not directly obtained on the Mössbauer sample used in earlier work, we reexamined the PAC data underlying Fig. 2 and found that these, too, could be fitted with a relaxation rate which goes to zero at 292.2(1) K, as shown in Fig. 3, right curve. We conclude that $T_c=292.2(1)$ K is the correct Curie temperature for the *Gd* ^{161}Dy Mössbauer data.

With the revised T_c value, the table of critical line broadenings given earlier,[1] and two points previously thought to be below T_c (Table II), we obtain a revised fit to Eq. (2), leading to the result $w=0.494(19)$. In contrast to our earlier analysis, the Mössbauer data now exhibit a single power law over the full range of reduced temperature, $10^{-3}<t<10^{-1}$. The quality of the power law is shown in Fig. 5.

To explore the sensitivity of the fitted values of w and D to the choice of T_c we show in Table III results for the full range of uncertainty of T_c. Nearly equivalent results also shown in Table III are obtained when w, D, *and* T_c are left free in fitting to Eq. (3). Successive elimination of points far from T_c produces no statistically significant changes in fitted values of the critical parameters, though it does introduce progressively larger errors.

For all these reasons we quote the final result

$$w=0.49(5),\ 10^{-3}<t<10^{-1}\ . \quad (4)$$

As can be seen from Table I, this is not consistent with either the spin-conserving or spin-nonconserving Heisenberg models, or the three-dimensional Ising model. Though the revised value of w is closer to theoretical predictions than earlier, it remains anomalous. A check on the result can be obtained via nuclear relaxation studies of *Gd* ^{111}In. Details of this work, currently underway in our laboratory, will be reported separately.[8]

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[5] Figure 1 was originally published as Fig. 3 of Ref. 1. In the original version the time scale was inadvertantly expanded by a factor of 10/9. This error has been corrected in the present paper. The same scale error also occured for Fig. 1 of Ref. 1.

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