Determination of the Universality Class of Gadolinium

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We resolve a long-standing puzzle for the static and dynamic critical behavior of gadolinium by a combined theoretical and experimental investigation. It is shown that the spin dynamics of a ferromagnet with hcp lattice structure and a spin-spin interaction given by both exchange and dipole-dipole interaction belongs to a new dynamic universality class, model J^*. Comparing results from mode coupling theory with results from three different hyperfine interaction probes we find quantitative agreement. [S0031-9007(97)04823-0]

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The spin dynamics of simple ferromagnets in the vicinity of their Curie point T_c are archetypical examples of dynamic critical phenomena near second-order phase transitions. Much experimental and theoretical effort has been put into identifying the dynamic universality classes and assigning them to magnetic substances. Nevertheless, experimental observations on Gd [1–6] remained a puzzle up to now. Because of its large localized magnetic moment, and the fact that it is an S-state ion, Gd should have a very small magnetocrystalline anisotropy and therefore be much better a model system for an isotropic Heisenberg magnet than either Fe, Ni, or EuO. As a consequence it should belong to the model J dynamic universality class in the classification scheme of Ref. [7]. The measured static and especially the dynamic critical exponents are, however, not at all compatible with model J. The objective of this paper is to resolve this longstanding seemingly contradictory situation by a combined theoretical and experimental study.

Early experimental observations [8] clearly demonstrate that Gd has an easy axis which coincides with the hexagonal axis of its hcp lattice. The origin of such an easy axis cannot be understood from the magnetocrystalline anisotropy. But, based on a mean field theory [9], it has been argued that a combined effect of the lattice structure and dipolar interactions favors the c axis as the easy direction. This view is supported by measurements [6] of the c axis and basal-plane susceptibility on a single crystal of Gd. It is found that the basal-plane susceptibility crosses over from a singular behavior to a constant on a characteristic temperature scale which can be accounted for by dipolar effects. The analysis of the static critical behavior [6] is, however, complicated by the fact that all experiments are done in the nonasymptotic regime where superposed crossover lead to complex temperature dependences. This may not be easily interpreted in terms of one or the other universality class. This is even more so, as the static critical exponents for the various universality classes are of comparable magnitude.

A surprising and yet unexplained observation was made by a measurement of the critical dynamics using hyperfine methods [1,2]. The critical exponent w for the autocorrelation time τ_c, which should scale as $\tau_c \propto (T - T_c)^{-w}$ in the asymptotic regime, was found to be $w \approx 0.5$. The observed value is not consistent with either Heisenberg or Ising models, but considerably lower.

The purpose of this Letter is twofold. First, we give a theoretical description for a spin system with both exchange and dipolar interaction on a hcp lattice using mode coupling theory. Next, we calculate the relaxation rates observed in various hyperfine interaction measurements, where we account for the details of the coupling tensor in each of these methods. We also report on measurements of the muon spin relation (μSR) rate in high purity single crystal samples of Gd. A comparison of the theoretical predictions with these and earlier [3,5] μSR measurements as well as perturbed angular correlation (PAC) and Mössbauer data [1,2] gives a coherent picture of the dynamic and static critical behavior of Gd and resolves the puzzling situation described above.

Taking into account magnetocrystalline anisotropy as well as the dipolar interaction the spin system is described by the Hamiltonian

$$H = -\sum_{i \neq j} [J_{ij}^{\perp}(S_i^x S_j^x + S_i^y S_j^y) + J_{ij}^{\parallel} S_i^z S_j^z + D_{ij}^{\alpha\beta} S_i^\alpha S_j^\beta].$$

The magnitude of the magnetocrystalline anisotropy of the system is characterized by $\Delta = J^{\parallel}/J^{\perp}$. The dipolar interaction is characterized by the tensor

$$D_{ij}^{\alpha\beta} = -\frac{(g_L \mu_B)^2}{2} \left(\frac{\delta_{\alpha\beta}}{|\mathbf{x}_{ij}|^3} - \frac{3 x_{ij}^\alpha x_{ij}^\beta}{|\mathbf{x}_{ij}|^5} \right), \qquad (1)$$

with $\mathbf{x}_{ij} = \mathbf{x}_i - \mathbf{x}_j$, g_L is the Landé factor, and μ_B the Bohr magneton. Dipole lattice sums, $D_{\mathbf{q}}^{\alpha\beta} = \sum_{i \neq j} \times D_{ij}^{\alpha\beta} e^{i\mathbf{q}\cdot\mathbf{x}_i}$, can be evaluated by using Ewald's method. For infinite three-dimensional *cubic lattices* the results may be found in Refs. [10,11]. For Bravais lattices with a

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hexagonal-closed packed (hcp) structure the dipole tensor to leading order in $\mathbf{q}$ becomes [9]

$$D_{\mathbf{q}}^{\alpha\beta} = \frac{(g_L\mu_B)^2}{2v_a}\left[\beta_4^\alpha \delta_{\alpha\beta} - 4\pi \frac{q_\alpha q_\beta}{q^2} + \mathcal{O}(q^2)\right],$$

where v_a is the volume of the primitive unit cell with lattice constant a, and the parameters are $\beta_4^x = 4.12$ and $\beta_4^x = 4.32$. Upon expanding the Fourier transform of the exchange interaction $J_{\mathbf{q}}^\alpha = \sum_i' J_{i0}^\alpha e^{i\mathbf{q}\cdot\mathbf{x}_i} \approx J_0^\alpha - Jq^2a^2 + \mathcal{O}(q^4)$, and keeping only those terms which are relevant in the spirit of the renormalization group theory, one finds

$$H = J\sum_q \left[\left(m^\alpha - \Delta_0^\alpha + q^2a^2\right)\delta_{\alpha\beta} + g\,\frac{q_\alpha q_\beta}{q^2}\right] S_{-\mathbf{q}}^\alpha S_{\mathbf{q}}^\beta . \tag{2}$$

There are two sources of uniaxial anisotropy, magnetocrystalline anisotropy, $\Delta_0^\alpha = J_0^\alpha/J$, and dipolar interaction, $m^\alpha = (g_L\mu_B)^2\beta_4^\alpha/2Jv_a$. In addition, the dipolar interaction introduces an anisotropy of the spin fluctuations with respect to the wave vector $\mathbf{q}$ which is reflected by the term proportional to $q_\alpha q_\beta/q^2$. The magnitude g of this anisotropy is given by $g = 4\pi(g_L\mu_B)^2/2Jv_a$. We define a dimensionless quantity $m = (g_L\mu_B)^2(\beta_4^\parallel - \beta_4^\perp)/2Jv_a$ proportional to the ratio between the anisotropy energy and the exchange energy. Putting in values for Gd the ratio of the dipolar contribution to the term $q_\alpha q_\beta/q^2$ and to the uniaxial anisotropy is $\sqrt{g/m} = 7.8738$. In the following, we will show that all available data for Gd can be explained by assuming that the uniaxial anisotropy is solely due to the dipolar interaction. Therefore, we will neglect the magnetocrystalline anisotropy in the following.

Now we turn to an analysis of the critical statics. In the Ornstein-Zernike approximation the susceptibility reads

$$\chi_{\alpha\beta}^{-1}(\mathbf{q}) = J\left[(r_\alpha + q^2)\delta_{\alpha\beta} + q_D^2\,\frac{q_\alpha q_\beta}{q^2}\right], \tag{3}$$

where $r_2 = r = \xi^{-2}$, $r_{x,y} = \xi^{-2} + q_A^2$, and we have measured all length scales in units of the lattice constant a. The analysis of the critical behavior resulting from the Hamiltonian is complicated by the fact that besides the correlation length $\xi = \xi_0(T/T_c - 1)^{-\nu}$ there are two anisotropy length scales $q_A^{-1} = a/\sqrt{m}$ and $q_D^{-1} = a/\sqrt{g}$, both resulting from the dipolar interaction. The eigenvalues of the inverse susceptibility matrix are given by $\lambda_1(\mathbf{q}) = q^2 + \xi^{-2} + q_A^2$ and $\lambda_{2,3}(\mathbf{q}) = q^2 + \xi^{-2} + [q_D^2 + q_A^2 \pm W]/2$ where $W = [(q_D^2 + q_A^2)^2 - 4q_D^2q_A^2q_z^2/q^2]^{1/2}$; the eigenvectors $e_i(\mathbf{q})$ are given in a forthcoming publication [12]. It is interesting to note that, due to the combined effect of the dipolar interaction and the uniaxial anisotropy of the lattice, the eigenvalues of the susceptibility matrix remain finite in the limit $q \to 0$ and upon approaching the critical temperature. Only if the angle ϑ between the easy axis (z axis) of magnetization and the wave vector is $\vartheta = 90°$ the third eigenvalue becomes critical. To a good approximation further static crossover effects can be incorporated in an effective exponent of the correlation length ξ [13].

Mode coupling theory is a theoretical method which has been shown to give highly accurate results for the critical dynamics of cubic ferromagnets [11]. Here we generalize this method to noncubic magnets. Starting from the equations of motion for the components $s_{\mathbf{q}}^\alpha$ of the spin $\mathbf{S_q}$ in the eigenvector basis $s_{\mathbf{q}}^\alpha = \sum_i S_{\mathbf{q}}^i e_{\alpha i}(\mathbf{q})$, one can derive the following set of coupled integral equations [12] for the half-sided Fourier transform $\Phi_\alpha(\mathbf{q},\omega) = i\chi_\alpha(\mathbf{q})/[\omega + i\Gamma_\alpha(\mathbf{q},\omega)]$, of the Kubo relaxation function $\Phi_{\alpha\beta}(\mathbf{q},t)$.

$$\Gamma_\alpha(\mathbf{q},\omega) = \frac{4k_BTJ^2}{\chi_\alpha(\mathbf{q})}\int_{\mathbf{k},\omega'}\sum_{\beta\gamma} K_\alpha^{\beta\gamma}(\mathbf{k},\mathbf{q}) \times \Phi_\beta(\mathbf{k},\omega)\Phi_\gamma(\mathbf{q}-\mathbf{k},\omega-\omega'), \tag{4}$$

where $\int_{\mathbf{k},\omega} = \int d^3k/(2\pi)^3\int(d\omega/2\pi)$. The vertex functions $K_\alpha^{\beta\gamma}(\mathbf{k},\mathbf{q})$ for the decay of the mode α into the modes β and γ are given by [12]

$$K_\alpha^{\beta\beta}(\mathbf{k},\mathbf{q}) = T_\alpha^{\beta\beta}(\mathbf{k},\mathbf{q})U_{\alpha\beta}^\beta(\mathbf{k},\mathbf{q})[\lambda_\beta(\mathbf{k}) - \lambda_\beta(\mathbf{q}-\mathbf{k})],$$
$$K_\alpha^{\beta\gamma}(\mathbf{k},\mathbf{q}) = T_\alpha^{\beta\gamma}(\mathbf{k},\mathbf{q})T_\alpha^{\beta\gamma}(\mathbf{k},\mathbf{q}), \quad \beta \neq \gamma, \tag{5}$$

with $U_{\alpha\beta}^\gamma(\mathbf{k},\mathbf{q}) = \sum_{ijk}\varepsilon_{ijk}e_{\alpha i}(\mathbf{k})e_{\beta j}(\mathbf{q})e_{\gamma k}(\mathbf{q}-\mathbf{k})$, where ε_{ijk} is the Levi-Cevita symbol, and $T_\alpha^{ii}(\mathbf{k},\mathbf{q}) = \lambda_i(\mathbf{k})U_{i\alpha}^i(\mathbf{k},\mathbf{q})$, $T_\alpha^{ij}(\mathbf{k},\mathbf{q}) = [\lambda_i(\mathbf{k}) - \lambda_j(\mathbf{q}-\mathbf{k})]U_{i\alpha}^j \times (\mathbf{k},\mathbf{q})$ for $i < j$ and $T_\alpha^{ij}(\mathbf{k},\mathbf{q}) = 0$ for $i > j$. For $q_D \to 0$ or $q_A \to 0$ these equations reduce either to the uniaxial or to the isotropic dipolar ferromagnet [11,14]. We have solved the above mode coupling equations in Lorentzian approximation [12]. It is found that the mode coupling equations obey a generalized dynamic scaling law where the linewidth depend on three scaling variables, $x_1 = 1/q\xi$, $x_2 = q_D/q$, and $x_3 = q_A/q$, and the angle ϑ. The explicit functional form, which is implicitly contained in the damping rates for the hyperfine interaction probes discussed below, is too complicated to be presented in this Letter, and we refer the reader to a forthcoming publication [12].

Now we compare the above theoretical results with experiments. We have performed zero field μSR experiments on Gd [15] using a high momentum muon beam at the facility of the Paul Scherrer Institut. Spin polarized muons were implanted in order to measure the distribution and the dynamics of the internal magnetic field at the muon site via the temporal loss of the initial spin polarization. The sample was a spherical Gd single crystal with diameter 2.5 cm. The temperature could be stabilized better than ± 0.05 K. We could describe the temporal loss of the initial muon spin polarization by an exponential decay function $P(t) = \exp(-\lambda_z t)$. Taking into account anisotropic dipolar fields as well as the isotropic Fermi contact field to the local field at the muon site the muon damping rate can be written as [12,16,17],

$$\lambda_{\hat{z}} = \frac{\pi\mathcal{D}}{V^2}\int_{\mathbf{q}}\sum_{\hat\beta\hat\gamma}[G_{\mathbf{q}}^{\hat x\hat\beta}G_{-\mathbf{q}}^{\hat x\hat\gamma} + G_{\mathbf{q}}^{\hat y\hat\beta}G_{-\mathbf{q}}^{\hat y\hat\gamma}]\Phi^{\hat\beta\hat\gamma}(\mathbf{q},0),$$

where $\mathcal{D} = \gamma_\mu^2(\mu_0/4\pi)^2(g_L\mu_B)^2$ and the hatted variables indicate that the corresponding quantities have to be evaluated in the muon reference frame. The coupling of the muon spin and the spins of the host magnet is described in terms of the coupling matrix $G_{\mathbf{q}}^{\hat{\alpha}\hat{\beta}}$, which reflects the particular symmetry of the lattice sites occupied by the muons. The most dominant contribution to the damping rate comes from wave vectors close to the Brillouin zone center [12,17] where one finds $G_{\mathbf{q}\to 0}^{\alpha\beta} = -4\pi[q_\alpha q_\beta/q^2 - p_\alpha]$, with $p_\alpha = d_\alpha + n_\mu H_\mu/4\pi$. With the Fermi contact field $B_{\mathrm{FC}} = -6.98$ kG at $T = 0$ K [18], one gets $n_\mu H_\mu/4\pi = -0.278$ [17], and consequently for octahedral sites $p_x = p_y = 0.0705$ and $p_z = 0.0250$ [17], and for tetrahedral sites $p_x = p_y = 0.0338$ and $p_z = 0.0984$ [12]. The muon relaxation rate λ_z depends on the material parameters $q_A\xi_0$ and $q_D\xi_0$. Since we assume that both anisotropies result from the dipolar interaction the ratio $q_D/q_A = 7.8738$ is known and the number of material parameters is reduced to one. In comparing our theory with μSR experiments at a polarization $\alpha = 90°$ we get the best fit to the data with $q_D\xi_0 = 0.13$ (see Fig. 1). This gives $q_A = 0.0165/\xi_0$ and $q_D = 0.13/\xi_0$. The corresponding crossover temperatures are $T_A = T_c + 0.43$ K and $T_D = T_c + 16.54$ K, suggesting the following crossover scenario. For $T \gg T_D$ we expect critical behavior dominated by the Heisenberg fixed point [19]. The relaxation rate shows power law behavior $\lambda \propto t^{-w}$, with an exponent $w \approx 1$. For temperatures in the interval $T_D > T > T_A$ the dipolar interaction becomes important. But, from the analysis of the uniaxial crossover [12] it turns out that the uniaxial crossovers in dynamics sets in at wave vectors much larger than expected from an analysis of the static quantities. Therefore, even for $T > T_A$ we expect to observe effects from dipolar interaction as well as uniaxial anisotropy. Finally, for $T < T_A$ the critical dynamics is determined by the uniaxial dipolar fixed point. Then the static susceptibilities do no longer diverge for $q \to 0$ and $T \to T_c$ except when the wave vector is perpendicular to the easy axis. Since the relaxation rate λ_z is given by an integral over the whole Bouillon zone, the relative weight of the critical axis along which the susceptibility diverges becomes vanishingly small. As a consequence, the relaxation rate λ_z no longer diverges for $T \to T_c$, i.e., $w \to 0$.

Figure 1 shows a comparison between the theoretical and experimental results for an initial polarization inclined by 90° with respect to the easy axis. The solid and dashed line are the theoretical result for the muon relaxation rate if the muons penetrating the sample are located at tetrahedral and octahedral interstitial sites, respectively. The comparison between theory and experimental favors tetrahedral sites. This is confirmed by μSR experiments with the initial polarization along the easy axis. The ratio $\lambda_z(90°)/\lambda_z(0°)$ for $T \to T_c$ becomes 1.2 and 0.7 for tetrahedral and octahedral sites, respectively [15]. The experiment is closer to the latter, strongly suggesting that muons occupy tetrahedral sites within the Gd lattice.

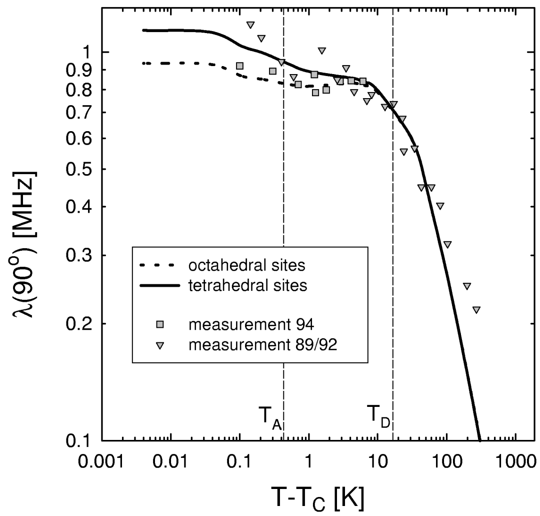

FIG. 1. Experimental and theoretical results of the relaxation rate λ for tetrahedral and octahedral muon sites with $\alpha = 90°$. Data taken from Refs. [3,5] and measured at the μSR facility at the PSI (see inset) [15]. The statistical errors of the relaxation rates are comparable to the size of the symbols.

The coupling tensor in PAC and Mössbauer measurements reduces to a Fermi contact coupling. Hence the observed relaxation rate is a sum over the eigenmodes $\tau_c = (k_B T/3v_a)\sum_\alpha \int_{\mathbf{q}} \chi_\alpha(\mathbf{q})/\Gamma_\alpha(\mathbf{q})$. Important information about the behavior of the autocorrelation time can be gained from a scaling analysis. An effective dynamical exponent z_{eff} may be defined by $\tau_c \propto (T - T_c)^{-w_{\mathrm{eff}}}$ with $w_{\mathrm{eff}} = \nu_{\mathrm{eff}}(z_{\mathrm{eff}} - 1)$, where we have neglected the Fisher exponent η. If dipolar interactions and uniaxial anisotropy were absent, one would expect $w \approx 1.0$. The dipolar interaction is known to be a relevant perturbation. It leads to asymptotic static critical exponents which are only slightly different from the corresponding Heisenberg values. But, since the dipolar interaction implies a nonconserved order parameter, the asymptotic dynamic exponent becomes $z_{\mathrm{D}} \approx 2$ resulting in a crossover from $w \approx 1.0$ to $w_{\mathrm{D}} \approx 0.7$. A uniaxial interaction is also known to be a relevant perturbation. Again, the static critical exponents are not changed very much, e.g., one finds that Ising (I) value $\nu_{\mathrm{I}} = 0.63$, but the dynamic exponent becomes $z_{\mathrm{I}} \approx 4$ if the order parameter is conserved ($z_{\mathrm{I}} \approx 2$ otherwise). The corresponding exponent for the hyperfine relaxation rate would be $w_{\mathrm{I}} \approx 1.89$ and $w_{\mathrm{I}} \approx 0.63$ for conserved and nonconserved order parameter, respectively. According to these scaling arguments it is hard to think of any dynamic universality class which could lead to an effective exponent w_{eff} smaller than about 0.6. Actually, Mössbauer and PAC measurements on Gd show distinctly anomalous low values $w \approx 0.5$, which cannot be explained by either of the above scenarios. This experimental puzzle can be resolved if one considers the combined effect of the dipolar interaction

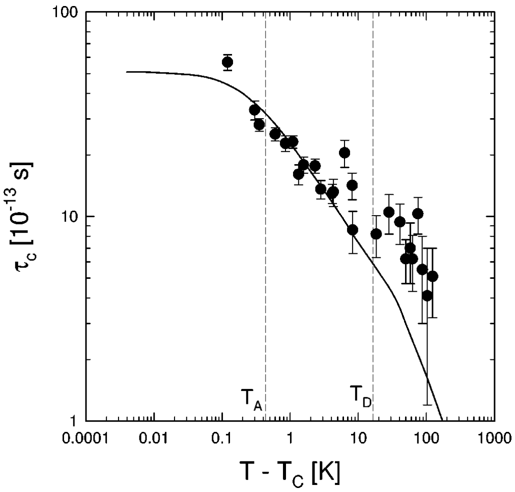

FIG. 2. Experimental and theoretical results of the autocorrelation time τ_c for PAC experiments. Data from [1].

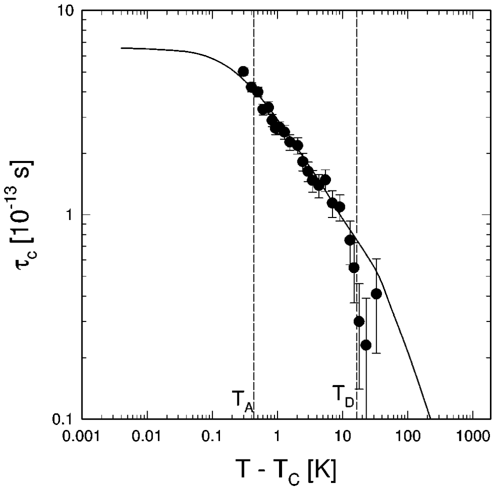

FIG. 3. Experimental and theoretical results of the autocorrelation time τ_c from Mössbauer spectroscopy [2].

and uniaxial anisotropy. As we have seen in the above analysis of the static critical behavior of uniaxial dipolar ferromagnets, *all* eigenvalues of the susceptibility matrix remain finite upon approaching the critical temperature except when the wave vector of the spin fluctuations is perpendicular to the easy axis. Since this is only a region of measure zero in the Brillouin zone one actually expects that the relaxation rate does no longer diverge upon approaching T_c.

Let us now compare the results of our mode coupling theory with hyperfine experiments on Gd mentioned above [1,2]. The autocorrelation time τ_c is shown in Figs. 2 and 3 for PAC experiments and Mössbauer spectroscopy, respectively. Both sets of data are in excellent agreement with the results from mode coupling theory for $T - T_c < 10$ K. Note that besides the overall frequency scale there is no fit-parameter, since we have used the same set of values for the dipolar and uniaxial wave vector as for our comparison with μSR experiments.

In summary, we have outlined a mode coupling theory for uniaxial dipolar ferromagnets, where the uniaxiality solely results from the dipolar interaction. We have also reported measurements on a high purity single crystal sample of Gd in the paramagnetic regime. From the quantitative agreement between this theory and our μSR and previous PAC and Mössbauer measurements the following conclusions can be drawn: (i) The universality class of Gd is the uniaxial dipolar ferromagnet, where both the isotropic dipolar and the uniaxial contribution to the spin Hamiltonian are due to a combined effect of dipolar interaction and noncubic-lattice structure. This corresponds to a new anisotropic model J^*. (ii) The dominant factor for the uniaxial anisotropy in Gd is the dipolar interaction. (iii) Muons in Gd are located at tetrahedral interstitial sites close to T_c.

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[1] G. Collins, A. Chowdhury, and C. Hohenemser, Phys. Rev. B **33**, 4747 (1986).
[2] A. Chowdhury, G. Collins, and C. Hohenemser, Phys. Rev. B **30**, 6277 (1984); **33**, 5070 (1986).
[3] O. Hartmann *et al.*, Hyperfine Interact. **64**, 369 (1990).
[4] E. Wäckelgard *et al.*, Hyperfine Interact. **31**, 325 (1986).
[5] E. Wäckelgard *et al.*, Hyperfine Interact. **50**, 781 (1989).
[6] D. Geldart, P. Hargraves, N. Fujiki, and R. Dunlap, Phys. Rev. Lett. **62**, 2728 (1989).
[7] P. Hohenberg and B. Halperin, Rev. Mod. Phys. **49**, 435 (1977).
[8] J. Cable and E. Wollan, Phys. Rev. **165**, 733 (1968).
[9] N. Fujiki, K. De'Bell, and D. Geldart, Phys. Rev. B **36**, 8512 (1987).
[10] A. Aharony and M. Fisher, Phys. Rev. B **8**, 3323 (1973).
[11] E. Frey and F. Schwabl, Adv. Phys. **43**, 577 (1994).
[12] S. Henneberger, E. Frey, F. Schwabl, and G. Kalvius (unpublished).
[13] K. Ried, Y. Millev, M. Fähnle, and H. Kronmüller, Phys. Rev. B **51**, 15 229 (1995).
[14] C. Bagnuls and C. Joukoff-Piette, Phys. Rev. B **11**, 1986 (1975).
[15] S. Henneberger *et al.*, Hyperfine Interact. **104**, 301 (1997).
[16] A. Yaouanc, P. D. de Réotier, and E. Frey, Phys. Rev. B **50**, 3033 (1994).
[17] P. D. de Réotier and A. Yaouanc, Phys. Rev. Lett. **72**, 290 (1994).
[18] A. Denison, H. Graf, W. Kündig, and P. Meier, Helv. Phys. Acta **52**, 460 (1979).
[19] Actually, the observed relaxation rate seems to show systematic deviations from the theoretical curve for $T \gg T_D$. This may be caused by a crossover into a Gaussian regime with $w \approx 0.75$ which would lead to the slight upwards bend of the data above T_D in Fig. 1.