μSR investigations of gadolinium in the paramagnetic regime near the ferromagnetic transition

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We report on the measurements of the muon relaxation rate in gadolinium (Gd) in the critical regime down to $T - T_C = 0.1$ K. The results are compared to the predictions of mode-coupling calculations for a uniaxial and dipolar ferromagnet. One observes that even if the uniaxiality in Gd is quite small, it should not be neglected in order to get a consistent description of our data. Most striking is the fact that these investigations support the assumption that above T_C the muon resides on the tetrahedral interstitial sites and not on the octahedral sites which are chosen in the low temperature phase.

1. Introduction

The rare earth metal gadolinium crystallizes in a hexagonal close packed (hcp) structure. At a Curie temperature of $T_C = 292.8$ K, it shows a ferromagnetic phase transition with the easy axis of the magnetization parallel to the c-axis. The phase transition originates in an RKKY coupling between Gd^{3+} ions with a magnetic moment $\mu_{\text{eff}}^{\text{exp}} = 7.98\mu_B$ which is in good agreement with the magnetic moment of an $^8S_{7/2}$ state. Due to their half-filled shell the Gd ions possess no orbital momentum and do not – in contrast to the other rare earth elements – show a single-ion anisotropy. Therefore one should expect Gd to behave like the prototype of a simple Heisenberg ferromagnet. On the other hand, the existence of an easy axis of magnetization below T_C would suggest an Ising-like behaviour. In contradiction to this experimental measurements of the critical exponents for static properties as well as for the dynamics are neither in

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agreement with an isotropic Heisenberg ferromagent nor with an Ising behaviour [3]. There are even problems to verify the scaling laws and to find the proper universality class for Gd. As has been shown by Frey and Schwabl [1,2] in cubic magnetic systems, problems in the explanation of ferromagnetic phase transitions may be solved, if one takes into account that near T_{C} the dipole–dipole coupling may not be neglected. In a non-cubic crystal, like the hcp structure, the dipole–dipole interaction results in an effective anisotropic coupling between the magnetic dipoles [4], which may be described in a generalized Heisenberg model with anisotropic exchange interaction. Therefore a consistent description of the critical dynamics of Gd has to include the dipole–dipole interaction as well as an anisotropic exchange coupling.

2. Mode-coupling theory

Some of the most successful theoretical approaches in critical dynamics are mode-coupling theories. In this section we will give a short summary of the mode-coupling theories for a uniaxial and dipolar ferromaget in the critical regime above T_{C}. For a detailed description we refer to [5]. Including uniaxiality and dipolar interaction, one starts up with the Hamiltonian

$$\bar{H} = -\sum_{ij} \left[J_{ij}^{\parallel} S_z^i S_z^j + J_{ij}^{\perp} S_x^i S_x^j + J_{ij}^{\perp} S_y^i S_y^j \right] \tag{1}$$

$$+ (g_{\mathrm{L}}\, \mu_{\mathrm{B}})^2 \sum_{i \neq j} \sum_{\alpha\beta} \frac{\partial^2}{\partial X_i^\alpha \partial X_j^\beta} \left(\frac{1}{|\boldsymbol{X}_i - \boldsymbol{X}_j|} \right) S_i^\alpha\, S_j^\beta \tag{2}$$

for the spin system. Retaining only terms relevant in the sense of renormalization-group theory in k-space, one gets for the inverse of the static susceptibility matrix the result

$$(\boldsymbol{\chi}^{-1})^{\alpha\beta}(\boldsymbol{k}) = J \left[\left(\xi^{-2} + m_\alpha + k^2 \right) \delta_{\alpha\beta} + g\, \frac{k_\alpha k_\beta}{k^2} \right], \tag{3}$$

$$m_1 = m_2 = q_A^2, \quad m_3 = 0, \quad g = q_{\mathrm{D}}^2,$$

with two new length scales q_{A}^{-1} and q_{D}^{-1} in addition to the correlation length ξ. Diagonalizing this matrix leads to the physical relevant eigenvalues and eigenvectors of the spin system. The eigenvector basis $\boldsymbol{w}_1(\boldsymbol{k}), \boldsymbol{w}_2(\boldsymbol{k}), \boldsymbol{w}_3(\boldsymbol{k})$ may be seen as an intermediate state between the basis for pure uniaxial,

$$\boldsymbol{w}_1(\boldsymbol{k}) = \frac{\mathrm{sgn}(\rho_1)}{\sqrt{\rho_1^2 + \rho_2^2}} \begin{pmatrix} \rho_2 \\ -\rho_1 \\ 0 \end{pmatrix},$$

$$\boldsymbol{w}_2(\boldsymbol{k}) = \frac{\mathrm{sgn}(\rho_1)}{\sqrt{\rho_1^2 + \rho_2^2}} \begin{pmatrix} \rho_1 \\ \rho_2 \\ 0 \end{pmatrix}, \qquad \boldsymbol{w}_3(\boldsymbol{k}) = \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}, \tag{4}$$

and the pure dipolar eigenmodes

$$\boldsymbol{w}_1(\boldsymbol{k}) = \frac{\boldsymbol{k} \times \widehat{\boldsymbol{z}}}{\sqrt{k_1^2 + k_2^2}}, \qquad \boldsymbol{w}_2(\boldsymbol{k}) = \frac{\boldsymbol{k}}{k}, \qquad \boldsymbol{w}_3(\boldsymbol{k}) = \boldsymbol{w}_2(\boldsymbol{k}) \times \boldsymbol{w}_1(\boldsymbol{k}). \tag{5}$$

Using the equations of motion for the three eigenmodes, one can calculate the matrix of the transport coefficients due to the Mori formalism. This matrix is just diagonal. Introducing scale variables

$$x = \xi/q, \qquad y = q_{\mathrm{D}}/q, \qquad z = q_{\mathrm{A}}/q, \qquad \boldsymbol{\rho} = \boldsymbol{k}/q, \tag{6}$$

one verifies that the diagonal elements fulfill the dynamical scaling relation [6],

$$\Gamma_{\alpha\alpha}(\boldsymbol{q}; r, g, m) = A\, q^z\, \gamma_\alpha(\nu; \boldsymbol{R}); \tag{7}$$

$$\begin{aligned} &\gamma_\alpha(\nu; \boldsymbol{R}) && \text{scaling function given in [5],} \\ &A = \sqrt{\frac{J k_{\mathrm{B}} T V^2}{2\pi^3 N^2}} && \text{non-universal scale,} \\ &z = 5/2 && \text{dynamical exponent.} \end{aligned} \tag{8}$$

3. Comparison with the experiment

For the experimental investigation of the critical dynamics of Gd zero-field muon, spin relaxation measurements on a spherical single crystal were continued [7] at the Paul Scherrer Institut. These measurements were performed in a temperature range from $T - T_{\mathrm{C}} = 0.1$ K to $T - T_{\mathrm{C}} = 40$ K, and we were able to stabilize the temperature with a variation smaller than 0.05 K.

Since the muon relaxation rate λ in a uniaxial ferromagnet depends on the angle between the spin of the incoming muons and the easy axis of magnetization, we measured λ at various angles α between the c-axis and the original muon spin direction (fig. 1).

The observed muon depolarisation is given by [9,11]

$$P(t) = \exp(-\lambda\, t), \tag{9}$$

with the relaxation rate given by

$$\lambda = \pi \gamma_\mu^2 \left(\frac{\mu_0}{4\pi}\right)^2 \frac{(g_{\mathrm{L}} \mu_{\mathrm{B}})^2}{V} \int \frac{\mathrm{d}^3\boldsymbol{q}}{(2\,\pi)^3} \left[\sum_{\beta\gamma} G^{\hat{\alpha}\hat{\beta}}(\boldsymbol{q}) G^{\hat{\alpha}\hat{\gamma}}(-\boldsymbol{q}) \Lambda(\boldsymbol{q})^{\hat{\beta}\hat{\gamma}} + \sum_{\beta\gamma} G^{\hat{\alpha}\hat{\beta}}(\boldsymbol{q}) G^{\hat{\alpha}\hat{\gamma}}(-\boldsymbol{q}) \Lambda(\boldsymbol{q})^{\hat{\beta}\hat{\gamma}} \right]. \tag{10}$$

We are using the symmetrized spin correlation functions

$$\Lambda(\boldsymbol{q})^{\beta\gamma} = \frac{1}{2}\left[\left\langle \delta S_{j_1}^{\beta}(\omega = 0) \delta S_{j_2}^{\gamma} \right\rangle + \left\langle \delta S_{j_2}^{\gamma} \delta S_{j_1}^{\beta}(\omega = 0) \right\rangle \right], \tag{11}$$

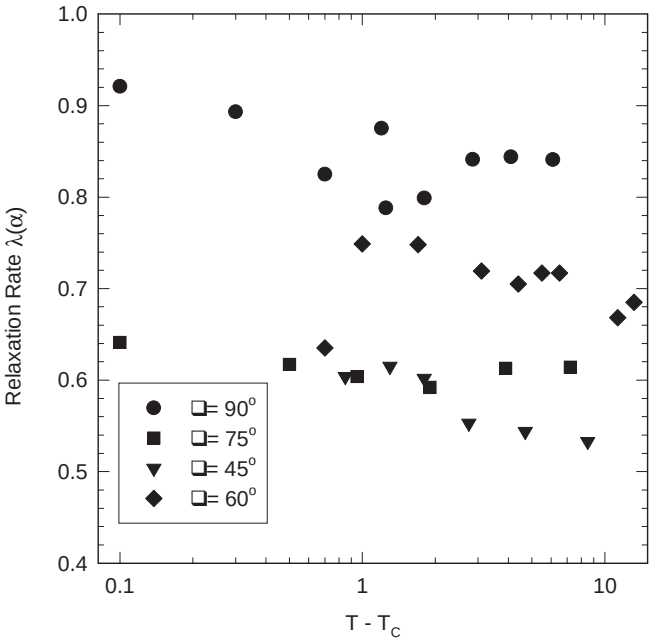

Fig. 1. The measured relaxation rate $\lambda(\alpha)$ as function of $T - T_{\mathrm{C}}$ at various λ.

which may be expressed through the line widths and the eigenvalues of the static susceptibility as follows

$$\Lambda(\boldsymbol{q})^{\beta\gamma} = \frac{2k_{\mathrm{B}}T}{\mu_0(g_{\mathrm{L}}\mu_{\mathrm{B}})^2}\sum_{\alpha} w_{\alpha\beta}(\boldsymbol{q})w_{\alpha\gamma}(\boldsymbol{q})\frac{1}{\lambda_\alpha(\boldsymbol{q})\,\Gamma_{\alpha\alpha}(\boldsymbol{q})}. \tag{12}$$

The coupling tensor $G^{\hat{\alpha}\hat{\beta}}(\boldsymbol{q} \to 0)$ describes the dipole coupling of the muon to the lattice spins and Fermi contact field for the muon. Since the zero-field relaxation measurements in the critical regime are dominated by the paramagnetic fluctuations at small wave vectors [12], the coupling tensor may be approximated with its value at zero wave vector

$$G^{\alpha\beta}(\boldsymbol{q} \to 0) = -4\pi\left[\frac{q_\alpha q_\beta}{q^2} - p_\alpha\right]. \tag{13}$$

In these equations, one has to pay attention to the fact that in eq. (9) the coupling tensor and the spin correlation function are measured in a coordinate system $(\widehat{\boldsymbol{x}}, \widehat{\boldsymbol{y}}, \widehat{\boldsymbol{z}})$ with the spin of the incoming muons parallel to the $\widehat{z}$-axis, but in eqs. (11) and (12) we are using a coordinate system $(\boldsymbol{x}, \boldsymbol{y}, \boldsymbol{z})$ which is rotated by an angle α to $(\widehat{\boldsymbol{x}}, \widehat{\boldsymbol{y}}, \widehat{\boldsymbol{z}})$ and where the c-axis of the crystal is in z-direction. So, one has to perform an orthogonal transformation on eqs. (11) and (12) to get the expressions used in eq. (9).

Since the contribution of the dipole coupling is non-isotropic, the wave vector independent part of the coupling tensor p_α depends on the interstitial site of the muon in a very sensitive way:

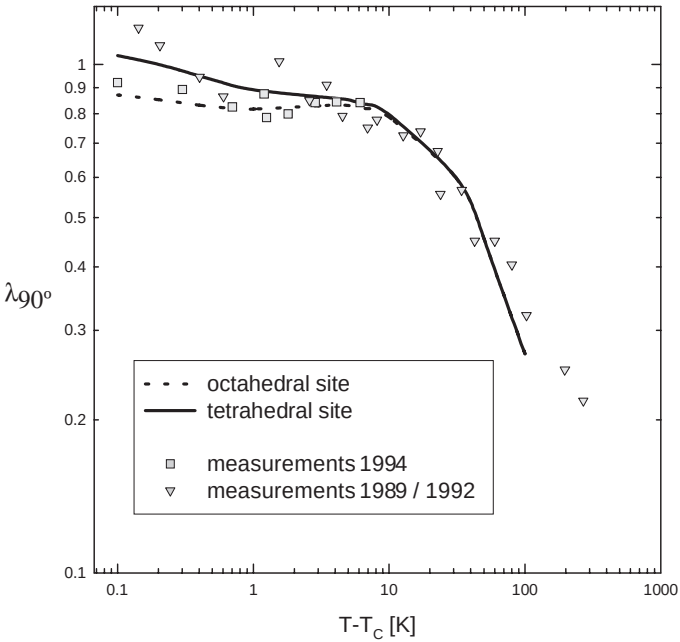

Fig. 2. The measured relaxation rate at $\alpha = 90°$ compared to theoretical calculations for tetrahedral and octahedral sites.

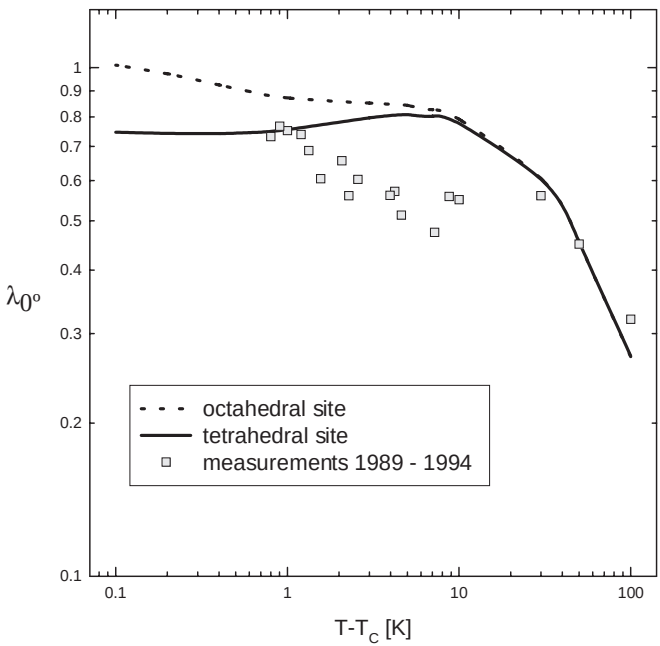

Fig. 3. The measured relaxation rate at $\alpha = 0°$ compared to theoretical calculations for tetrahedral and octahedral sites.

- tetrahedral interstitial site:

$$p_x = 0.0338, \qquad p_y = 0.0338, \qquad p_z = 0.0984; \tag{14}$$

- octahedral interstitial site [11]:

$$p_x = 0.0705, \qquad p_y = 0.0705, \qquad p_z = 0.025. \tag{15}$$

The results for these two possible sites of the relaxation rate with muon spin perpendicular and parallel to the c-axis are shown in figs. 2 and 3 respectively.

4. Discussion

If one compares the experimental results to the theoretical calculations of the relaxation rate, one can see that for the angle $\alpha = 90°$ the tetrahedral as well as the octahedral sites are consistent with the measurements (fig. 2). But since the measurements definitely show that the relaxation rate parallel to the c-axis is always smaller than perpendicular to the c-axis (fig. 2 and fig. 3), the comparison with our calculations excludes the occurrence of the octahedral sites and favours the tetrahedral sites. So our results suggest that although at temperatures below 220 K, there is a strong evidence that muon resides on the octahedral sites [8], at higher temperatures there should be a change in the interstitial site.

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