Possibility of Observation of the Critical Paramagnetic Longitudinal Spin Fluctuations in Gadolinium by Muon Spin Rotation Spectroscopy

P. Dalmas de Réotier and A. Yaouanc
Centre d'Etudes Nucleaire/Département de Recherche Fondamentale sur la Matière Condensée, 85X, F-38041 Grenoble, France
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The theory described here shows that the muon spin depolarization rates measured on gadolinium in the critical paramagnetic region give direct information on the longitudinal (along the wave vector $\mathbf{q}$) spin fluctuations. In the Ising regime it is possible to distinguish the parallel (along the c axis) from the perpendicular fluctuations.

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Gadolinium is a rare earth metal which crystallizes in the hexagonal close packed structure (hcp) and exhibits simple ferromagnetic ordering along the c axis at temperatures near the Curie temperature, $T_C \approx 293$ K. Its ferromagnetism is due to the Ruderman-Kittel-Kasuya-Yosida interaction between the Gd^{3+} ions which are in the ${}^8S_{7/2}$ state. This ionic electronic structure suggests that the magnetocrystalline anisotropy is very small. Therefore a simple Heisenberg behavior was expected for the magnetic properties of gadolinium near T_C. Contrary to these expectations, measurements have shown that these properties are quite complicated. It is only recently that a consistent experimental picture of the *static properties* has emerged [1, 2]. It seems that all the complications are due to the magnetic dipole-dipole interactions. These long range interactions cause a crossover as the temperature is reduced in the paramagnetic region from an isotropic Heisenberg regime to a dipolar Heisenberg regime with a crossover temperature of approximately 4 K above T_C. As the temperature is reduced further, there is a second crossover temperature to Ising behavior approximately 0.5–1 K above T_C [3].

The *dynamics properties* of gadolinium have been studied mainly by two hyperfine techniques: the perturbed angular correlation method [4] and the positive muon spin relaxation and rotation spectroscopy (μSR) [5, 6]. There is little information from neutron experiments [7]. μSR measurements performed on a single crystal seem to show that the magnetic fluctuations have a pronounced anisotropy when the temperature is approached to within ≈ 0.5 K from T_C [5]. The μSR data recorded on a polycrystalline sample indicate that the magnetic fluctuations are almost temperature independent for $1 \text{ K} \lesssim T - T_C \lesssim 10$ K [6]. Because μSR measurements can be carried out in a wide temperature range which can extend from T_C up to very high temperature, they should allow us to study in detail the different crossovers. Up to now no quantitative information has been extracted from these measurements mainly because the relation between the data and the correlation functions which characterize the fluctuations of the Gd^{3+} total moments has not been available. Recently it has been shown that it is possible to understand quantitatively μSR data recorded in cubic paramagnets [8]. The purpose of this Letter is to provide a theoretical framework for the interpretation of the available μSR data on gadolinium and to initiate further and more accurate experiments. For an anisotropic crystal structure, the dipolar part of the Hamiltonian is the sum, near the Brillouin zone center, of two terms : one which is also present in an isotropic compound and an extra one which is an Ising-like term in the case of gadolinium. This latter term strongly influences the dynamical behavior near T_C.

Before describing our work in detail we stress that, following the usual terminology, the adjectives "longitudinal" and "transverse" are used overall in the text in two meanings: (1) to designate the orientation of the muon polarization relative to the *applied magnetic field* $\mathbf{B}_{\text{ext}}$, and (2) to indicate the character of a spin fluctuation relative to its *wave vector* $\mathbf{q}$. The meaning becomes clear in the context. In addition we use the words "parallel" or "perpendicular" to describe the direction of the fluctuations relative to the c axis.

μSR experiments [9, 10] are usually performed either with the longitudinal or transverse geometry. These geometries differ in the direction of the initial muon beam polarization, $\mathbf{P}(t = 0)$, relative to $\mathbf{B}_{\text{ext}}$. In a longitudinal (transverse) experiment $\mathbf{P}(t = 0)$ is parallel (perpendicular) to $\mathbf{B}_{\text{ext}}$. Note that a longitudinal experiment can be performed without any applied magnetic field. We take $\mathbf{P}(t = 0)$ parallel to the Z axis of an orthogonal reference frame, $(\mathbf{X}, \mathbf{Y}, \mathbf{Z})$ where $\mathbf{X}, \mathbf{Y}$, and $\mathbf{Z}$ are unit vectors. In a longitudinal (transverse) geometry experiment the $P_Z(t)$ $(P_X(t))$ depolarization function is measured. Each of these functions is an exponential function which is therefore characterized by a damping rate, λ_Z and λ_X, respectively. We shall describe the crystal structure of gadolinium in the orthohexagonal lattice structure $(\mathbf{a}, \mathbf{b}, \mathbf{c})$ with $\mathbf{a} = \mathbf{a}_h$, $\mathbf{b} = \mathbf{a}_h + 2\mathbf{b}_h$, and $\mathbf{c} = \mathbf{c}_h$ where $(\mathbf{a}_h, \mathbf{b}_h, \mathbf{c}_h)$ are the basis vectors of the hexagonal lattice structure. Although gadolinium is described by four interpenetrating identical Bravais lattices in the orthohexagonal structure instead of two in the hexagonal structure, the advantage

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of using the former one is that the spin dipolar interactions (between the muon and the lattice on one hand and within the lattice on the other) are better described. We introduce two orthogonal reference frames: (a) frame $(\hat{\mathbf{a}},\hat{\mathbf{b}},\hat{\mathbf{c}})$ in which $\hat{\mathbf{a}}$, $\hat{\mathbf{b}}$, and $\hat{\mathbf{c}}$ are unit vectors collinear to **a**, **b**, and **c**; (b) frame $(\mathbf{x},\mathbf{y},\mathbf{z})$ which is chosen according to the magnetic symmetry of the compound. In our case, because gadolinium orders with its magnetic moments along the c axis, we identify $(\mathbf{x},\mathbf{y},\mathbf{z})$ with $(\hat{\mathbf{a}},\hat{\mathbf{b}},\hat{\mathbf{c}})$. We denote θ the angle between the Z and z axes. We are first going to derive an expression for $\lambda_Z(\theta=0)$.

We generalize the method of Yaouanc *et al.* [8], taking into account the hexagonal and non-Bravais crystal structure of gadolinium. We derive the following formula valid in zero magnetic field:

$$\lambda_Z(\theta=0) = \frac{\pi\mathcal{D}}{V}\int\frac{d^3\mathbf{q}}{(2\pi)^3}\sum_{\beta\gamma}\left[G^{x\beta}(\mathbf{q})G^{x\gamma}(-\mathbf{q}) + G^{y\beta}(\mathbf{q})G^{y\gamma}(-\mathbf{q})\right]\tilde{\Lambda}^{\beta\gamma}(\mathbf{q}). \quad (1)$$

The integral extends over the first Brillouin zone and β and γ stand for x, y, or z. We have defined $\mathcal{D} = (\mu_0/4\pi)^2\gamma_\mu^2(g_L\mu_B)^2$. V is the volume of the sample, μ_0 the permeability of free space, μ_B the Bohr magneton, γ_μ the muon gyromagnetic ratio ($\gamma_\mu = 851.6$ Mrad s^{-1} T^{-1}), and g_L the Landé factor. This equation shows that the damping rate depends on the coupling between the muon spin and the spins of the magnet through the coupling tensor $G(\mathbf{q})$ and on the spin correlation tensor of the magnet itself, $\Lambda(\mathbf{q},\omega)$, taken at $\omega=0$, i.e., $\tilde{\Lambda}(\mathbf{q}) \equiv \Lambda(\mathbf{q},\omega=0)$. We have $G(\mathbf{q}) = 1/n_d\sum_d G_d(\mathbf{q})$ where the index d runs over each of the $n_d = 4$ gadolinium sites belonging to an orthohexagonal unit cell and $G_d(\mathbf{q}) = \sum_i \exp[i\mathbf{q}\cdot(\mathbf{i}+\mathbf{d})]G_{\mathbf{r}_{i+d}}$. i runs over the orthohexagonal cells of the crystal lattice and $G_{\mathbf{r}_{i+d}}$ is a dimensionless tensor which is the sum of the classical dipolar and Fermi contact couplings between the muon spin and the Gd^{3+} ion located at distance vector $\mathbf{r}_{i+d}$ from the muon. $\tilde{\Lambda}(\mathbf{q})$ writes $\sum_{d,d'}\tilde{\Lambda}_{d,d'}(\mathbf{q})$ where $\tilde{\Lambda}_{d,d'}(\mathbf{q})$ is the correlation tensor between spins belonging to sublattices d and d'.

Since our purpose is to describe the critical spin dynamics of gadolinium which is a ferromagnet, we are only interested in the behavior of the tensor $G(\mathbf{q})$ near the zone center. We find

$$G^{\alpha\gamma}(\mathbf{q}\longrightarrow\mathbf{0}) = -4\pi\left[P_L^{\alpha\gamma}(\mathbf{q}) - p_\alpha\delta^{\alpha\gamma}\right], \quad (2)$$

with $p_\alpha \equiv (d^\alpha + r_\mu H/4\pi)$ where d^α is a contribution from the dipolar coupling. $P_L^{\alpha\gamma}(\mathbf{q}) = q^\alpha q^\beta/q^2$ is the longitudinal projector operator. In addition we define the transverse projector operator $P_T^{\alpha\gamma}(\mathbf{q}) = \delta^{\alpha\gamma} - P_L^{\alpha\gamma}(\mathbf{q})$ which will be used below. Using the fact that the muon in gadolinium localizes in an interstitial octahedral site [11] we compute $d^x = d^y = 0.3485$ and $d^z = 1 - 2d^x = 0.3030$. r_μ is the number of nearest neighbor magnetic ions to the muon localization site and H the hyperfine constant which is proportional to the Fermi contact field, $B_{\rm cont}$, at the muon site:

$$B_{\rm cont} = \frac{\mu_0}{4\pi}g_L J\mu_B\frac{n_d}{v}r_\mu H, \quad (3)$$

where v is the volume of the orthohexagonal cell. From the experimental values $B_{\rm cont} = -0.698$ T [11], $g_L J = 7.1$, and $v = 132$ Å^3 [12] we compute $r_\mu H/4\pi = -0.278$. Therefore $p_\alpha \simeq p = 1/3 - 0.278 = 0.055$, i.e., $p_\alpha \ll 1$. This means that this parameter can be safely neglected in the temperature range investigated up to now. We shall prove it below.

For the computation of $\lambda_Z(\theta=0)$ we need to specify $\tilde{\Lambda}(\mathbf{q})$, the properties of which are controlled by the Hamiltonian $\mathcal{H}$ of the magnet which we take as the sum of two terms : the usual Heisenberg Hamiltonian and the dipolar Hamiltonian which describe the exchange and dipolar interactions between the Gd^{3+} ions, respectively. The critical dynamics is determined by the small $\mathbf{q}$ behavior of these interactions. In this limit the dipolar interaction depends on the tensor [3]

$$A^{\alpha\beta}(\mathbf{q}\longrightarrow\mathbf{0}) = \alpha_1 P_L^{\alpha\beta}(\mathbf{q}) - \alpha_3^\alpha\delta^{\alpha\beta}, \quad (4)$$

where $\alpha_3^x = \alpha_3^y \neq \alpha_3^z$. We have retained in Eq. (4) only the terms relevant in the sense of the renormalization group theory. The first term describes the usual dipolar coupling between the ions which is always present. The second term introduces an Ising component in $\mathcal{H}$ which is due to the anisotropy of the hcp crystal structure. Therefore, in addition to the necessity to distinguish the fluctuations transverse and longitudinal to $\mathbf{q}$ as needed for the cubic compounds, we have to consider whether these fluctuations are perpendicular or parallel to the easy axis [13]. The symmetry properties of $\mathcal{H}$ in real and $\mathbf{q}$ space dictate the following functional dependence for the spin correlation functions:

$$\tilde{\Lambda}^{\beta\gamma}(\mathbf{q}) = \tilde{\Lambda}^{\beta,T}(q)P_T^{\beta\gamma}(\mathbf{q}) + \tilde{\Lambda}^{\beta,L}(q)P_L^{\beta\gamma}(\mathbf{q}). \quad (5)$$

$\tilde{\Lambda}(\mathbf{q})$ is a symmetric tensor. $\tilde{\Lambda}^{\beta,T}(q)$ and $\tilde{\Lambda}^{\beta,L}(q)$ are respectively the transverse and longitudinal correlation functions for the fluctuations which are either perpendicular ($\beta = x,y \equiv \perp$) or parallel ($\beta = z \equiv \parallel$) to the c axis. If the fluctuations perpendicular and parallel to the c axis are identical, the $\tilde{\Lambda}^{\beta\gamma}(\mathbf{q})$ expression is, as expected, the same as the one used for cubic compounds. The $\tilde{\Lambda}^{\beta,T}(q)$ and $\tilde{\Lambda}^{\beta,L}(q)$ functions are related by the fluctuation-dissipation theorem to the static wave vector dependent susceptibilities, $\chi^{\beta,T}(q)$ and $\chi^{\beta,L}(q)$, respectively, and linewidths of the spectral weight functions at $\omega=0$, $\Gamma^{\beta,T}(q)$ and $\Gamma^{\beta,L}(q)$, respectively,

$$\tilde{\Lambda}^{\beta,T;L}(q) = \frac{2N}{\mu_0(g_L\mu_B)^2}k_BT\frac{\chi^{\beta,T;L}(q)}{\Gamma^{\beta,T;L}(q)}. \quad (6)$$

N is the number of Gd^{3+} ions in the crystal and k_B the Boltzmann constant.

From Eq. (1) and the results given at Eqs. (5), (6) and at Eq. (2) with $p_\alpha = 0$, it is a simple matter to derive an expression for $\lambda_Z(\theta = 0)$:

$$\lambda_Z(\theta = 0) = \mathcal{V} \int_0^{q_{BZ}} dq\, q^2 \left[\frac{4}{5} \frac{\chi^{\perp,L}(q)}{\Gamma^{\perp,L}(q)} + \frac{1}{5} \frac{\chi^{\parallel,L}(q)}{\Gamma^{\parallel,L}(q)} \right]. \quad (7)$$

We have set

$$\mathcal{V} = \frac{8}{3} \frac{\mu_0}{4\pi} \gamma_\mu^2 k_B T \frac{n_d}{v} \simeq \frac{8}{3} \frac{\mu_0}{4\pi} \gamma_\mu^2 k_B T_C \frac{n_d}{v}. \quad (8)$$

Using the same approach, an expression for $\lambda_Z(\theta = \pi/2)$ is easily obtained:

$$\lambda_Z\left(\theta = \frac{\pi}{2}\right) = \mathcal{V} \int_0^{q_{BZ}} dq\, q^2 \left[\frac{3}{5} \frac{\chi^{\perp,L}(q)}{\Gamma^{\perp,L}(q)} + \frac{2}{5} \frac{\chi^{\parallel,L}(q)}{\Gamma^{\parallel,L}(q)} \right]. \quad (9)$$

Note that the damping rates depend only on the longitudinal fluctuations. By simple linear combination, Eqs. (7) and (9) give the possibility to determine separately

$$\lambda^{\beta,L} = \mathcal{V} \int_0^{q_{BZ}} dq\, q^2 \frac{\chi^{\beta,L}(q)}{\Gamma^{\beta,L}(q)} \quad (10)$$

for $\beta = \perp$ and $\beta = \parallel$ if $\lambda_Z(\theta = 0)$ and $\lambda_Z(\theta = \pi/2)$ are measured. Therefore μSR measurements on gadolinium give directly information on the longitudinal spin dynamics. In addition it is possible to distinguish the perpendicular from the parallel fluctuations. If p_α is different from zero, the transverse fluctuations contribute to the damping. With $p_\alpha = 0.055$, if we use the results derived for the cubic dipolar Heisenberg model [14], we find that at $T - T_C = 0.25$ K the contribution of the transverse fluctuations is almost negligible: it represents only $\simeq 20\%$ of the contribution of the longitudinal fluctuations. At $T - T_C = 1$ K it has already decreased down to $\simeq 8\%$.

We have just derived the longitudinal damping rate expressions for $\theta = 0$ and $\theta = \pi/2$. For an arbitrary θ value the following relation holds:

$$\lambda_Z(\theta) = \cos^2\theta\, \lambda_Z(\theta = 0) + \sin^2\theta\, \lambda_Z(\theta = \pi/2). \quad (11)$$

Thus the damping rate $\lambda_Z(\theta)$ is a linear combination of $\lambda_Z(\theta = 0)$ and $\lambda_Z(\theta = \pi/2)$. Equation (11) has been derived using Eqs. (2) and (5). The relation as given in Eq. (11) is valid for the transverse damping rate if Z is replaced by X. Moreover, the following relations between the longitudinal and transverse damping rates can be easily shown : $\lambda_X(\theta = 0) = \lambda_Z(\theta = \pi/2)$ and $\lambda_X(\theta = \pi/2) = [\lambda_Z(\theta = \pi/2) + \lambda_Z(\theta = 0)]/2$. Thus transverse field measurements do not carry more information than zero field measurements (here we neglect the effect of the applied field on the magnetic fluctuations).

For a polycrystalline sample, in a first approximation, we have $P_{Z;X}(t) = \exp\left(-\bar{\lambda}_{Z;X} t\right)$ where $\bar{\lambda}_Z$ and $\bar{\lambda}_X$ are respectively the powder average of $\lambda_Z(\theta)$ and $\lambda_X(\theta)$. The previous relations which are valid for crystals imply $\bar{\lambda}_Z = \bar{\lambda}_X$.

Since at sufficiently high temperature compared to T_C the Ising anisotropy does not influence the static magnetic properties [1, 2], one can expect that the dynamical properties should not be influenced by this anisotropy. Therefore in this temperature region the fluctuations perpendicular and parallel to the c axis are identical. This means that $\lambda_Z(\theta)$ and $\lambda_X(\theta)$ are equal and independent of θ. We find

$$\lambda_Z(\theta) = \lambda_X(\theta) = \bar{\lambda}_Z = \bar{\lambda}_X = \mathcal{W} I^L(\varphi), \quad (12)$$

where $\mathcal{W}$ is a nonuniversal constant and $I^L(\varphi)$ a function which has been first defined in Ref. [8]. It can be found by identifying Eq. (12) with Eq. (7) or Eq. (9) and has been computed by Frey and Schwabl [14]. The angle φ is a measure for the temperature through its dependence on the correlation length ξ: $\varphi = \arctan(q_D \xi)$ where q_D is the dipolar wave vector determined by the relative strengths of the dipolar and exchange interactions.

We now consider the two sets of published gadolinium data [5, 6]. In Fig. 1 we present the 10 mT transverse field damping rate data recorded on a polycrystalline sample [6]. Although the previous theoretical results are strictly valid only in zero magnetic field, we may expect that a small magnetic field will not affect them. The full line in Fig. 1 is a fit to the polycrystalline data using Eq. (12) with $\mathcal{W} = 5.82$ MHz and $q_D \xi_0 = 0.105$. ξ_0 is the correlation length at $T = 2T_C$. From the expression of $\mathcal{W}$ given at Eq. (5.10c) of Ref. [8] we deduce $q_D = 0.084$ Å^{-1} and therefore $\xi_0 = 1.25$ Å. The expression for $\mathcal{W}$ which has been derived for cubic compounds should be valid in our case because there is no crystalline structure de-

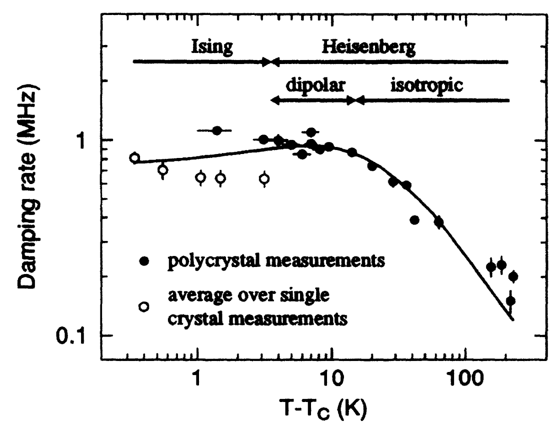

FIG. 1. Temperature dependence of the μSR damping rate for gadolinium. The full line is the prediction of the dipolar Heisenberg model which depends on two parameters: $q_D \xi_0 = 0.105$ which fixes the temperature scale and $\mathcal{W} = 5.82$ MHz which sets the scale of the damping rate. The polycrystal and single crystal data are from Refs. [6] and [5], respectively. The notation is further explained in the main text.

pendent parameter in it. Comparing these values to the corresponding ones for cubic magnets (Table II of Ref. [8]), we see they are within the expected range. We note that the mode-coupling theory of the dipolar Heisenberg model [14] gives a good description of the data down to $T - T_C \simeq 4$ K. The crossover from the isotropic Heisenberg regime to the dipolar Heisenberg regime occurs at $\simeq 15$ K which is to be compared to the $\simeq 4$ K value derived from the susceptibility measurements. In Fig. 1 we have plotted also the powder average of the single crystal zero field damping rate data [5]. There is a disagreement between the two sets of data. We do not understand the origin of the discrepancy on the vertical scale. We suspect that it reflects some experimental problems. Because of this discrepancy we cannot analyze the data in more detail. We simply note that a comparison between the polycrystalline data and the full curve of Fig. 1 seems to indicate that the Ising crossover occurs around 4 K above T_C, i.e., when the theoretical prediction for the dipolar Heisenberg Hamiltonian fails to account for the polycrystalline experimental data.

In conclusion we have shown that μSR measurements have the potentiality to give essential information on the longitudinal fluctuations in gadolinium in *truely zero field*. Before giving any definite statement about their temperature dependence more data are needed, particularly in zero field for which the present theoretical development is directly applicable. The purpose of the analysis of such data would be to check the validity of the Hamiltonian used to describe the magnetic properties of gadolinium at T_C and the reliability of the mode coupling approximation to describe uniaxial magnets [15]. It is worth noting that when the muon site and the contact field are known, it is a relatively simple matter to write the relations between the μSR damping rates and the spin correlation functions. Near the Curie temperature μSR probes mainly the dynamical properties close to the center of the Brillouin zone where only limited experimental data are available from other methods.

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