Muon-spin-relaxation study of the critical longitudinal spin dynamics in a dipolar Heisenberg ferromagnet

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We present zero-field muon-spin-relaxation data on the ferromagnetic fluctuations near the Curie temperature of the intermetallic $GdNi_5$, which is a Heisenberg magnet with strong dipolar interaction. Our data show that the critical longitudinal (along the wave vector **q**) spin fluctuations above and below the Curie temperature are similar. They are an experimental proof of this similarity. This result is explained using the dynamical scaling theory of Halperin and Hohenberg.

Although the transition from the paramagnetic to the ordered state of a simple ferromagnet is an archetypical second-order phase transition, the dramatic effect of the dipolar interaction on the nature of the critical paramagnetic spin fluctuations has been fully understood only recently. An almost quantitative agreement has been achieved between the experimental data from magnetization,[1] neutron,[2] and local probe[3,4] techniques and mode coupling theory.[5] In contrasts below the Curie temperature T_C the experimental data are scarce[6,7] and a complete theory is still lacking.[8] This report presents a detailed study on crystals of the spin dynamics for a dipolar Heisenberg ferromagnet by a local probe technique.

Our zero-field muon-spin-relaxation (μSR) (for an introduction to this technique see Ref. 9) measurements have been performed on the intermetallic ferromagnet $GdNi_5$ which crystallizes in the hexagonal $CaCu_5$ crystal structure (space group $P6/mmm$). It exhibits a ferromagnetic phase transition at $T_C \simeq 32$ K.[10] Nickel itself does not carry a spontaneous magnetic moment but has an induced moment of 0.16 μ_B (Ref. 11) that we will neglect. The Gd^{3+} ions are in the $^8S_{7/2}$ state. This suggests that the magnetocrystalline anisotropy is small.

Recently we have reported preliminary zero-field μSR experimental data[12] which indicates that, contrary to the conclusion of Gignoux *et al.*[10] derived from bulk magnetization measurements, $GdNi_5$ is an axial magnet. In order to understand the discrepancy between bulk and microscopic measurements we have carried out magnetization measurements on a $GdNi_5$ sphere. They show that the easy axis is the c axis and that the anisotropy field is $B_a(T=5\text{ K}) \simeq 0.21$ T. A lattice sum computation of B_a due to the dipolar interaction between the Gd^{3+} ions gives $B_a(T=5\text{ K}) = 0.22$ T. This computation shows also that the lowest energy magnetic moment configuration is obtained for the magnetic moments oriented along the c axis. Therefore $GdNi_5$ is a dipolar Heisenberg ferromagnet, i.e., its magnetic anisotropy is only due to the dipolar interaction.

The reported μSR measurements[13] have been performed at the EMU spectrometer of the ISIS surface muon beam facility[14] (UK) on two single crystals which differ by the orientation of the c axis relative to the initial muon beam polarization $\mathbf{S}_\mu$ which is either parallel or perpendicular to the c axis.[12] We define the Z axis as the axis parallel to $\mathbf{S}_\mu$. We have used the flow cryostat and temperature controller (ITC503 from Oxford Instruments) of the spectrometer. In this cryostat the sample is in contact with a low pressure helium exchange gas of ~ 15 mbar. During the recording of a spectrum we have kept the temperature stable within 0.005 K.

At all temperatures the measured depolarization function is well fitted by a sum of an exponential function which describes the depolarization from the sample and a constant term which takes into account the muons stopped in the silver backing plate and cryostat walls. A typical spectrum is displayed in Fig. 1. The depolarization function relative to the compound is characterized by an initial asymmetry a_i and a damping rate λ_Z. The constant term is easily determined because λ_Z is large in most cases. As expected $a_i(T)$ is temperature independent except below T_C for the sample with $\mathbf{S}_\mu$ perpendicular to the c axis where $a_i = 0$:[12] the spontaneous muon-spin rotation is then too fast to be resolved on a pulsed source such as ISIS.

In Fig. 2 we present $\lambda_Z(T)$ for the two orientations of $\mathbf{S}_\mu$ relative to the c axis. Whereas λ_Z is temperature independent for $T \gtrsim 50$ K, it displays a weak increase when approaching T_C from above as shown in Fig. 3. On the other hand, below T_C, $\lambda_Z(T)$ exhibits a more pronounced temperature dependence, except near T_C where it saturates (Fig. 4). A description of the temperature behavior of λ_Z in the critical ferromagnetic region is the main subject of this paper. Before discussing it in detail we analyze the data recorded for $T \ll T_C$ and in the paramagnetic critical region in order to characterize the magnetic fluctuations in these two temperature regions.

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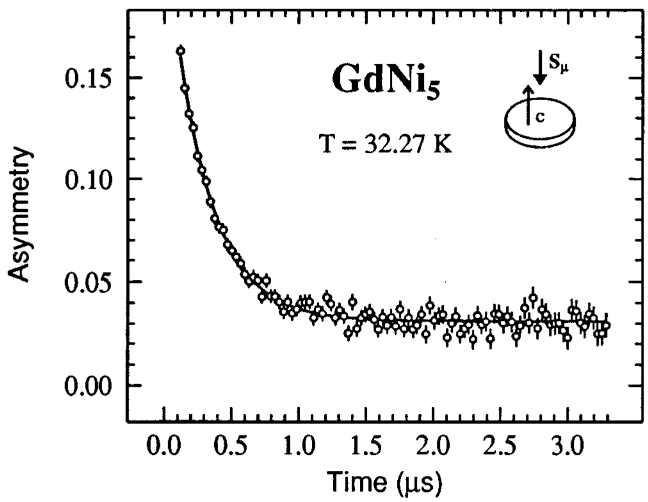

FIG. 1. A typical μSR spectrum recorded with the initial muon polarization $\mathbf{S}_\mu$ parallel to the c axis. This spectrum contains eight million events.

The T_C value has been determined from the $\lambda_Z(T)$ data recorded with $\mathbf{S}_\mu$ parallel to the c axis. We have considered the measurement for which $\lambda_Z(T)$ is maximum and the closest measurements on each side. T_C has been taken as the average of these three points and the error bar the distance between the average and the extreme points. In Figs. 3 and 4 we do not display the data points in the interval used for the T_C determination. We have found $T_C = 31.832$ (16) K. This is consistent with the initial asymmetry and damping rate data recorded on the sample with the c axis perpendicular to $\mathbf{S}_\mu$ and with the published value.[10,11] Note that the conclusions of this work do not depend critically on the precision of the T_C determination.

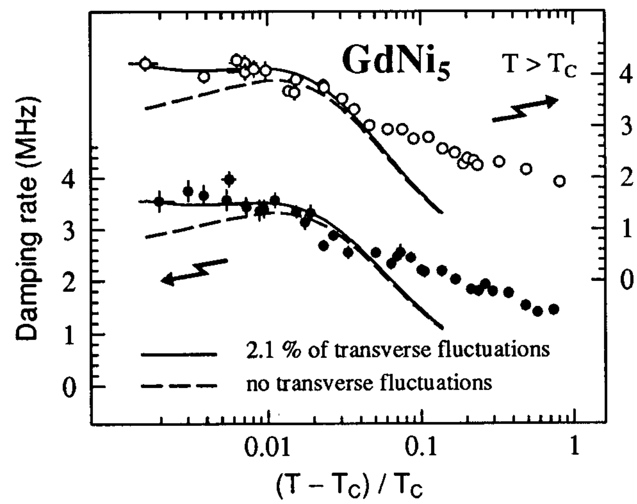

FIG. 3. Zero-field μSR damping rate λ_z measured in the critical paramagnetic state given as a function of the temperature relative to the Curie temperature and the orientation of $\mathbf{S}_\mu$ relative to the c axis (same symbol convention as in Fig. 2). The full and dashed lines are predictions of the mode coupling theory for the paramagnetic critical behavior of λ_Z(T) in a dipolar Heisenberg ferromagnet.

We first analyze λ_Z for $T \ll T_C$. We express λ_Z in terms of correlation functions.[4,15] An energy conservation argument tells us that only the parallel (to the easy axis which we denote z; the Z and z axes are parallel) fluctuations contribute to λ_Z,[16] i.e., λ_Z depends on the zz component of the spin correlation tensor of the magnet $\Lambda^{zz}(q,\omega)$ taken at zero energy transfer. Modeling the effect of the dipolar interaction between the Gd^{3+} ions by B_a, we compute $\Lambda^{zz}(q,\omega=0)$ and then derive the following expression:[17]

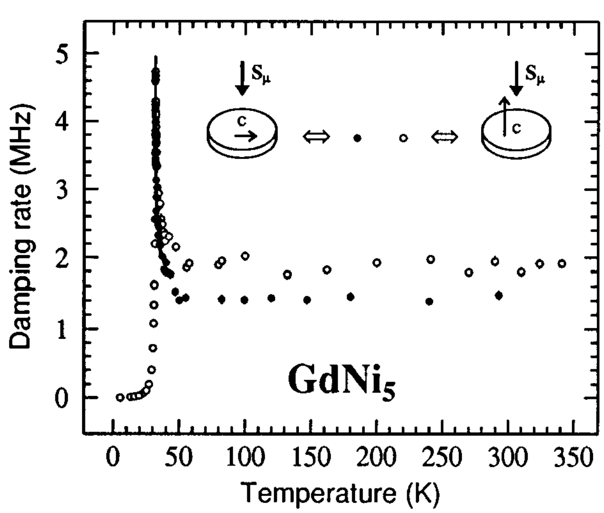

FIG. 2. Temperature dependence of the zero-field μSR damping rate measured on two crystals of $GdNi_5$ which differ by the orientation of $\mathbf{S}_\mu$ relative to the c axis. We do not observe any μSR signal below the Curie temperature when $\mathbf{S}_\mu$ is perpendicular to the c axis because this axis is the easy magnetic axis.

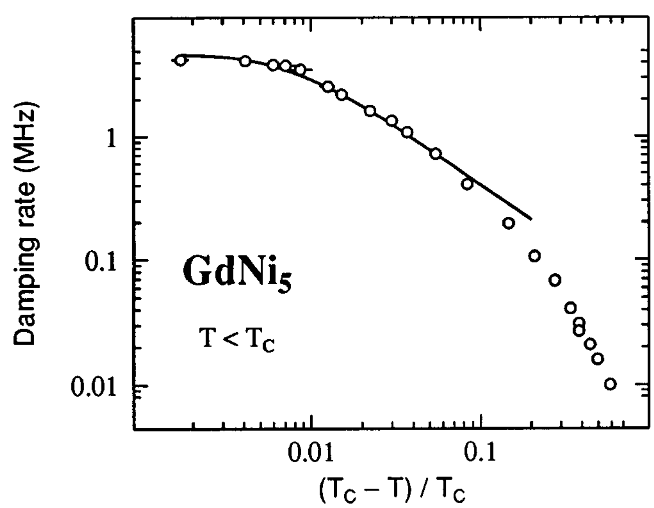

FIG. 4. Zero-field μSR damping rate measured in the ferromagnetic state given as a function of the renormalized temperature relative to the Curie temperature. The *full line* is the prediction for the *critical paramagnetic* fluctuations (Refs. 5 and 19). The relative weight of the longitudinal and transverse fluctuations is taken as given by the analysis of the paramagnetic fluctuations.

$$\lambda_Z = \frac{\mathscr{C} g_L^2 T^2}{D^3} \ln\left[\frac{\exp(T_a/T)}{\exp(T_a/T)-1}\right]. \quad (1)$$

Note that λ_Z is independent of the characteristics of the muon localization sites in $GdNi_5$. We have defined $T_a = g_L \mu_B B_a / k_B$. g_L is the Landé factor, μ_B the Bohr magneton, k_B the Boltzmann constant, and D the magnon stiffness constant. $\mathscr{C}$ is a universal constant [$\mathscr{C} = 129.39$ (meV)^3 Å^6 s^{-1} K^{-2}]. The dipolar interaction induces the two-magnon process giving a finite exponential damping rate.[16] Using Eq. (1) for $T \lesssim 16$ K we find that $D = 3.2$ (1) meV Å^2. This value is in the expected range when compared to the value for typical Heisenberg magnets such as EuO and EuS for which $D = 11.65$ meV Å^2 and 2.56 meV Å^2, respectively.[18] We deduce the dipolar wave vector q_D which determines the relative strengths of the dipolar and exchange interactions. For a cubic compound we derive $q_D = g_L \mu_B (\mu_0 / 2\mathscr{J} a^2 v_0)^{1/2} = g_L \mu_B (\mu_0 S / D v_0)^{1/2}$ because $D = 2\mathscr{J} S a^2$ (Ref. 4). $S = 7/2$ is the value of the Gd^{3+} spin, μ_0 is the permeability of free space, v_0 the volume per ion ($v_0 = 82.6$ Å^3), $\mathscr{J}$ is an exchange integral, and a the cube edge. Because q_D is an energy ratio, its expression should not strongly depend on the lattice structure. We find $q_D = 0.19$ Å^{-1}.

We now consider $\lambda_Z(T)$ measured in the critical paramagnetic region. The observed temperature independence of λ_Z (see Fig. 3) has already been measured for nickel and gadolinium which are dipolar Heisenberg ferromagnets. It has been explained as an effect of the dipolar interaction.[4,15] Therefore we are led to attribute the behavior of $\lambda_Z(T)$ for $T > T_C$ to that interaction.

This interpretation is consistent with the fact that λ_Z is only weakly dependent on the direction of $\mathbf{S}_\mu$ relative to the c axis in the critical regime (15% difference between the two directions). Using Ref. 19 we determine that the measurements probe fluctuations for $q \sim q_D$. Because of this relatively large q value, the small q approximation for the tensor describing the coupling between the muon spin and the lattice spins may be only approximate. This may explain the 15% anisotropy. The saturation effect observed in this temperature range has a simple explanation:[15] λ_Z probes mainly the longitudinal (along the wave vector $\mathbf{q}$) magnetic fluctuations. It has been shown for nickel, iron and gadolinium that λ_Z can be written as a weighted sum of the contributions from the longitudinal and transverse fluctuation modes:

$$\lambda_Z = \mathscr{W}^+ [a_L I^L(\varphi) + a_T I^T(\varphi)], \quad (2)$$

where $\mathscr{W}^+$ is a nonuniversal constant, $a_{L,T}$ depends only on the muon localization site(s), and $I^{L,T}(\varphi)$ are universal fluctuation functions of the temperature through the angle φ.[4,15,19] We have $\varphi = \arctan(q_D \xi_0^+ t^{-\nu})$ where ξ_0^+ is the correlation length at $T = 2T_C$, $t \equiv |T - T_C|/T_C$, and ν the correlation length critical exponent ($\nu \simeq 0.69$). The superscripts L and T refer to the longitudinal and transverse (relative to $\mathbf{q}$) fluctuations, respectively. The index $+$ $(-)$ on a parameter specifies that we consider this parameter in the paramagnetic (ferromagnetic) state. The observed saturation effect occurs if $a_L I^L(\varphi) \gg a_T I^T(\varphi)$.

We first consider Fig. 3 and the data recorded with $\mathbf{S}_\mu$ and c parallel. The full line is the prediction of Eq. (2) with $\mathscr{W}^+ a_L = 24$ MHz and $\mathscr{W}^+ a_T = 0.50$ MHz. The dashed line is computed with $\mathscr{W}^+ a_L = 24$ MHz and $\mathscr{W}^+ a_T = 0$ MHz. We now describe the data recorded with $\mathbf{S}_\mu$ and c perpendicular. The full line is drawn with $\mathscr{W}^+ a_L = 20.4$ MHz and $\mathscr{W}^+ a_T = 0.425$ MHz and the dashed line with $\mathscr{W}^+ a_L = 20.4$ MHz and $\mathscr{W}^+ a_T = 0$ MHz. Obviously, for the four curves, we have taken the same $q_D \xi_0^+$ value ($q_D \xi_0^+ = 0.065$). Note that the contribution of the transverse fluctuations is small. Using our previously determined q_D value we deduce $\xi_0^+ \sim 0.34$ Å. This is smaller than expected. It simply points out that for $t \gtrsim 0.04$ (i.e., when the theoretical curves do not describe the data) the noncritical short-range magnetic correlations outweigh the critical fluctuations. Therefore the derived $q_D \xi_0^+$ value is not correct. It should be reminded that the available theory only describes the critical fluctuations (characterized by small wave vectors). To end up with the data recorded in the paramagnetic phase, we note that the damping rate value of the points very close to T_C (namely the points which either have been used for the determination of T_C or correspond to $t \lesssim 0.002$) is significatively larger than the saturation value obtained for $0.002 \lesssim t \lesssim 0.02$. This can be seen by comparing the values of the damping rate in Figs. 2 and 3. This increase of the damping rate could be due to the Ising crossover that has been for instance observed beyond the dipolar Heisenberg regime in metallic Gd.[20,15]

We have just established that $GdNi_5$ is an axial dipolar magnet. We have determined its anisotropy magnetic field, its dipolar wave vector and the fact that μSR essentially measures the longitudinal fluctuations in the critical paramagnetic region. We now consider $\lambda_Z(T)$ measured in the *critical ferromagnetic region*. In Fig. 4 we compare $\lambda_Z(T)$ to the prediction of Eq. (2) with the fluctuation functions given by paramagnetic mode coupling theory:[5] the data are well described. The full line is computed with $\mathscr{W}^- a_L = 27$ MHz, $\mathscr{W}^- a_T = 0.56$ MHz, and $q_D \xi_0^- Z_{oz} = 0.020$. Therefore $\xi_0^- Z_{oz} = 0.057$ Å. Z_{oz} is a renormalization factor: it appears for example in the expression of the wave-vector-dependent susceptibility in the critical regime of a simple Heisenberg magnet below T_C;[6] its value has not been computed for a dipolar magnet. We have kept the relative weight of the longitudinal and transverse fluctuations as determined from the analysis above T_C. The fact that the fit works so well suggests that the static wave-vector-dependent susceptibility probed by the measurements[4,5] is of the Ornstein-Zernike form with a renormalized correlation length which we note $\xi_0^- Z_{oz}$. Although this result is not obvious, it is known to be valid near T_C for a Heisenberg ferromagnet with no dipolar interaction.[6] The ferromagnetic and paramagnetic $\mathscr{W} a_L$ values are in reasonable agreement.

The description of the dynamics below T_C is *a priori* more complicated than above T_C. In a dipolar Heisenberg magnet the dipolar interaction has a twofold manifestation. Whereas in the direct space it determines the easy axis, in the reciprocal space it splits the fluctuations into longitudinal and transverse modes relative to $\mathbf{q}$. The observed similarity between the paramagnetic and ferromagnetic longitudinal critical fluctuations is remarkable. It can be understood using the dynamical scaling theory of Halperin and Hohenberg.[21] The basic quantity which distinguishes the different regions in the (q,ξ^{-1}) plot is $q\xi$. Our measurements are mostly sensitive to longitudinal modes with $q \sim q_D$.[19] Therefore the relevant quantity for our data is $q_D\xi$. If we take $\xi_0 = 1$ Å we find $q_D\xi \simeq 5$ at $t = 10^{-2}$. Despite our rough estimate for the correlation lengths we are yet clearly in the critical region of the paramagnetic and ferromagnetic dynamics. Referring to Fig. 1 of Ref. 21 we understand the continuity of the dynamical behavior crossing T_C and therefore the observed similarity. Nevertheless this argument calls for a detailed theoretical justification: in a dipolar magnet two scaling variables are needed[5] instead of one for the model of Ref. 21.

In contrast to our findings above T_C, we do not observe below T_C short-range correlation effects. Remembering that λ_Z is expressed as an average of correlation functions over the Brillouin zone,[19] we deduce that, below T_C, the long-range correlations (small **q**) outweigh the short-range correlations (large **q**) in the average process.

Recently it has been shown that the behavior of the homogeneous magnetization dynamics in the ferromagnetic and paramagnetic states of EuS are similar.[7] We note that the measurements were performed in an applied field. On the opposite, our muon-spin-relaxation measurements have been performed in truly zero field. This is a definitive advantage.

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