

## Zero-field muon spin lattice relaxation rate in a Heisenberg ferromagnet at low temperature

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We provide a theoretical framework to compute the zero-field muon spin relaxation rate of a Heisenberg ferromagnet at low temperature. We use the linear spin wave approximation. The rate, which is a measure of the spin lattice relaxation induced by the magnetic fluctuations along the easy axis, allows one to estimate the magnon stiffness constant.

### I. INTRODUCTION

Recently, the measured temperature behavior of the zero-field muon spin relaxation ( $\mu$ SR) rate of some Heisenberg ferromagnets has been analyzed quantitatively in the paramagnetic<sup>1-3</sup> and ferromagnetic<sup>4</sup> critical regimes. This analysis has given information on the anisotropy of the spin dynamics in the reciprocal and direct spaces. It has shown that the anisotropy in the reciprocal space, which is induced by the dipolar interaction, has a strong influence on the dynamics near the zone center of the Brillouin zone. Note that this interaction always exists in real ferromagnets. The experimental data in the isotropic dipolar Heisenberg regime are well explained by the mode coupling approximation of Frey and Schwabl.<sup>5</sup> On the other hand, up to now, the low-temperature behavior of the relaxation rate has only been considered qualitatively<sup>6</sup> or in restricted physical cases.<sup>7</sup> The main purpose of this work is to provide a theoretical framework to analyze  $\mu$ SR relaxation data recorded on Heisenberg ferromagnets at low temperature.

The organization of this paper is as follows. In Sec. II, we summarize the relation between the measured depolarization function and the spin-spin correlation function of the magnet. In Sec. III we compute the muon spin lattice relaxation rate in the linear spin-wave approximation for a Heisenberg Hamiltonian with an energy gap. In Sec. IV we apply the result of the previous section to describe the spin lattice relaxation of a dipolar Heisenberg magnet in the spin-wave temperature region. In this last section we present the conclusion of our work.

### II. MUON SPIN RELAXATION AND SPIN-SPIN CORRELATION FUNCTIONS

We take the  $Z$ -axis parallel to the incoming muon beam polarization. A zero-field muon spin relaxation measurement consists of measuring the time depolarization function  $P_Z(t)$ .<sup>8,9</sup> For simplicity we take the easy magnetic axis  $z$  parallel to  $Z$ . We write<sup>10,11</sup>

$$P_Z(t) = \exp[-\psi_z(t)], \quad (1a)$$

with

$$\psi_z(t) = \gamma_\mu^2 \int_0^t d\tau (t - \tau) [\Phi_{xx}(\tau) + \Phi_{yy}(\tau)]. \quad (1b)$$

$\gamma_\mu$  is the muon gyromagnetic ratio:  $\gamma_\mu = 851.6$  Mrad s<sup>-1</sup> T<sup>-1</sup>.  $\psi_z(t)$  does not depend on the muon pulsation frequency  $\omega_\mu$  because the associated energy  $\hbar\omega_\mu$  is negligible.<sup>6</sup> In Eq. (1b)  $\Phi_{\alpha\alpha}(\tau)$  is the symmetrized time correlation function of the  $\alpha$  fluctuating component of the local magnetic field at the muon site and  $\{x, y, z\}$  is an orthogonal frame.

Experimentally it is usually observed that  $P_Z(t)$  is an exponential function. Equation (1) predicts such a result if  $\tau$  is very small relative to  $t$ , i.e., if the characteristic decay time of  $\Phi_{\alpha\alpha}$  is much smaller than  $t$  which is  $\sim 2 \mu$ s. Taking this hypothesis which is justified and the fact that the field correlation functions are even functions of  $\tau$ , we derive that  $P_Z(t)$  is an exponential function characterized by a damping rate  $\lambda_z$  which can be written in terms of the time Fourier transform of field correlation functions at  $\omega = 0$

$$\lambda_z = \pi\gamma_\mu^2 [\Phi_{xx}(\omega = 0) + \Phi_{yy}(\omega = 0)]. \quad (2)$$

It is possible to express the magnetic field at the muon site as a function of, on one hand, a tensor which describes the coupling between the localized spins of the metal and the muon spin and on the other hand of the localized spins components themselves. Because  $\Phi_{\alpha\alpha}(\tau)$  is quadratic in fields,  $\lambda_z$  can be written as a linear combination of spin-spin correlation functions of the magnet. In general the calculation of these correlation functions is performed in  $\mathbf{q}$  space, i.e., in the first Brillouin zone. Therefore the spatial Fourier transform of these functions has to be introduced. This is done in detail in Ref. 1. Using the result given at Eq. (2) we find

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

The integral extends over the first Brillouin zone. This equation shows that  $\lambda_z$  depends on the coupling between the muon spin and the spins of the magnet through the coupling tensor  $G(\mathbf{q})$  and the spin correlation tensor of the magnet,  $\Lambda(\mathbf{q}, \omega)$ , taken at  $\omega = 0$ , i.e.,  $\tilde{\Lambda}(\mathbf{q}) \equiv \Lambda(\mathbf{q}, \omega = 0)$ . The information on the muon localization site is contained in  $G(\mathbf{q})$ . We have defined  $\mathcal{D} = (\mu_0/4\pi)^2 \gamma_\mu^2 (g_L \mu_B)^2$  where  $\mu_0$  is the permeability of free space,  $g_L$  the Landé factor, and  $\mu_B$  the Bohr magneton.  $V$  is the volume of the sample. The formula given at Eq. (3) allows one to deal with Bravais and at least some non-Bravais lattices, for instance the hexagonal close packed lattice. In the latter case  $G(\mathbf{q}) = 1/n_d \sum_d G_d(\mathbf{q})$  where the index  $d$  runs over the  $n_d$  nonequivalent sites of the magnet and  $G_d(\mathbf{q}) = \sum_i \exp[i\mathbf{q} \cdot (\mathbf{i} + \mathbf{d})] G_{\mathbf{r}_{i+d}}$ .  $i$  runs over each cell of the crystal lattice and  $G_{\mathbf{r}_{i+d}}$  is a dimensionless tensor which accounts for the classical dipolar and

Fermi contact couplings between the muon spin and the magnet spins located at distance vector  $\mathbf{r}_{i+d}$  from the muon.  $\tilde{\Lambda}(\mathbf{q})$  writes  $\sum_{d,d'} \tilde{\Lambda}_{dd'}(\mathbf{q})$  where  $\tilde{\Lambda}_{dd'}(\mathbf{q})$  is the correlation tensor between spins belonging to cell sites  $d$  and  $d'$ . Note that the case of Bravais lattice is included in the definition we have just given:  $d$  takes then only one value ( $n_d = 1$ ) and the sums reduce to a single element.

The tensor  $G(\mathbf{q})$  is the sum of dipolar and hyperfine tensors which we denote  $D(\mathbf{q})$  and  $H(\mathbf{q})$ , respectively. In general terms, the sublattice  $d$  contribution to  $D(\mathbf{q})$  writes<sup>1</sup>

$$D_d^{\alpha\beta}(\mathbf{q}) = -4\pi \left[ P_L^{\alpha\beta}(\mathbf{q}) - C_d^{\alpha\beta}(\mathbf{q}) \right], \quad (4a)$$

where  $P_L^{\alpha\beta}(\mathbf{q})$  is the longitudinal projection operator and  $C_d^{\alpha\beta}(\mathbf{q})$  a symmetric tensor:

$$C_d^{\alpha\beta}(\mathbf{q}) = \frac{q_\alpha q_\beta}{q^2} \left[ 1 - \exp\left(\frac{-q^2}{4\rho^2}\right) \right] - \frac{1}{4\rho^2} \sum_{\mathbf{K} \neq 0} (K_\alpha + q_\alpha)(K_\beta + q_\beta) \varphi_0\left(\frac{(\mathbf{q} + \mathbf{K})^2}{4\rho^2}\right) \exp(-i\mathbf{K} \cdot \mathbf{r}_{0+d}) \\ + \frac{v\rho^3}{2(\pi)^{3/2}} \sum_i [2\rho^2 r_{i+d,\alpha} r_{i+d,\beta} \varphi_{3/2}(\rho^2 r_{i+d}^2) - \delta^{\alpha\beta} \varphi_{1/2}(\rho^2 r_{i+d}^2)] \exp(i\mathbf{q} \cdot \mathbf{r}_{i+d}). \quad (4b)$$

The  $D(\mathbf{q})$  expression is derived using an Ewald transformation.<sup>12</sup> The  $\varphi_m(x)$  functions are defined in Ref. 1.  $\mathbf{K}$  is a vector of the reciprocal lattice and  $\mathbf{r}_{i+d}$  (respectively  $\mathbf{r}_{0+d}$ ) is the vector that links the muon site to the ion belonging to sublattice  $d$  and located in lattice cell  $i$  (respectively origin) of the crystal lattice. Expression (4b) gives the same result for all values of the Ewald parameter  $\rho$ , but for numerical applications a value of  $\rho$  is chosen which ensures that both series of Eq. (4b) converge rapidly. The  $C_d(\mathbf{q})$  tensor, the trace of which is 1, reveals the symmetry of the point group at the muon site. Whereas the elements of the  $C_d(\mathbf{q})$  tensor are analytical functions of  $\mathbf{q}$  for all  $\mathbf{q}$  values, the elements of  $P_L(\mathbf{q})$  are only piecewise continuous at  $\mathbf{q} = 0$ .

The hyperfine interaction is short range and usually isotropic. In the lowest order in  $\mathbf{q}$  we have

$$H^{\alpha\beta}(\mathbf{q} = 0) \equiv 1/n_d \sum_d H_d^{\alpha\beta}(\mathbf{q} = 0) = r_\mu H \delta^{\alpha\beta}, \quad (5)$$

where  $r_\mu$  is the number of nearest-neighbor magnetic ions to the muon site and  $H$  a constant which can be deduced from the muon spin rotation frequency at low temperature. Equation (5) is derived under the hypothesis that the muon site is a center of symmetry.

In order to proceed further, in this paper we will only consider the behavior of  $G(\mathbf{q})$  near the zone center. From the previous results, using  $C^{\alpha\beta}(\mathbf{q}) \equiv 1/n_d \sum_d C_d^{\alpha\beta}(\mathbf{q})$ , we derive in the limit  $\mathbf{q} \rightarrow 0$

$$G^{\alpha\beta}(\mathbf{q}) = -4\pi \left[ P_L^{\alpha\beta}(\mathbf{q}) - C^{\alpha\beta}(0) - \frac{r_\mu H \delta^{\alpha\beta}}{4\pi} \right]. \quad (6)$$

### III. MUON SPIN RELAXATION RATE IN A HEISENBERG FERROMAGNET WITH A SMALL ENERGY GAP

For the computation of the damping rate we need an expression for the correlation functions of the magnet. We first have to specify its Hamiltonian. As we have done before<sup>6</sup> we suppose that the magnet is described by a Heisenberg interaction with a small energy gap. Because the minimum magnon energy is much larger than  $\hbar\omega_\mu$ , the energy conservation principle tells us that only the parallel (to the easy axis  $z$ ) fluctuations contribute to the depolarization,<sup>6</sup> i.e., the measurements probe only the correlation function  $\tilde{\Lambda}^{zz}(\mathbf{q})$ . Then a close look at Eq. (3) indicates that the diagonal terms of  $G(\mathbf{q})$  do not contribute to the depolarization. This means that an isotropic hyperfine interaction does not influence the measured damping rate. For many possible muon localization sites in crystals we have  $C^{zx}(\mathbf{q} = 0) = C^{zy}(\mathbf{q} = 0) = 0$ . Using this hypothesis we deduce the simple result  $G^{z\beta}(\mathbf{q} \rightarrow 0) = -4\pi P_L^{z\beta}(\mathbf{q})$  for  $\beta = x, y$ . This leads to the important fact that, within our hypothesis, the muon spin relaxation rate is independent of the muon localization site. The previous analysis leads to the following simple result:

$$\lambda_z = \frac{2\mathcal{D}}{V} \int d^3\mathbf{q} \frac{q_z^2}{q^2} \left( 1 - \frac{q_z^2}{q^2} \right) \tilde{\Lambda}^{zz}(\mathbf{q}). \quad (7)$$

A generalization of this result when  $C^{zx}(\mathbf{q} = 0)$  and  $C^{zy}(\mathbf{q} = 0)$  are not zero is not difficult: it results in a modified prefactor that depends on  $C^{zx}(\mathbf{q} = 0)$  and  $C^{zy}(\mathbf{q} = 0)$ . We compute  $\tilde{\Lambda}^{zz}(\mathbf{q})$  in the linear spin-wave approximation.<sup>13</sup> In this approximation the fluctuating

part of the  $z$  component of the total angular momentum of the magnetic ion,  $\delta J_{\mathbf{q}}^z$ , is expressed in terms of a sum of products of boson operators  $a_{\mathbf{k}}^+$  and  $a_{\mathbf{k}}^-$ :

$$\delta J_{\mathbf{q}}^z = -\frac{1}{N} \sum_{\mathbf{k}, \mathbf{k}_1} \delta(-\mathbf{q} + \mathbf{k} - \mathbf{k}_1, \mathbf{K}) a_{\mathbf{k}}^+ a_{\mathbf{k}_1}^-, \quad (8)$$

where  $\mathbf{K}$  is a vector of the reciprocal lattice. Each of the  $\mathbf{k}$  and  $\mathbf{k}_1$  sums extends over  $N$  vectors of the first Brillouin zone. Because  $\delta J_{\mathbf{q}}^z$  is given in terms of products of two operators, the computation of  $\tilde{\Lambda}^{zz}(q)$  requires the evaluation of four operator products. We decouple these products. We set

$$\langle a_{\mathbf{k}}^+ a_{\mathbf{k}_1}^- a_{\mathbf{q}}^+ a_{\mathbf{q}_1}^- \rangle = N^2 \delta_{\mathbf{k}_1, \mathbf{q}} \delta_{\mathbf{k}, \mathbf{q}_1} n_{\mathbf{k}} (n_{\mathbf{q}} + 1), \quad (9)$$

where  $n_{\mathbf{q}}$  is the standard Bose occupation factor. With these results it is easy to derive an expression for the parallel spin-spin correlation function. We find

$$\tilde{\Lambda}^{zz}(\mathbf{q}) = \sum_{\mathbf{p}} \frac{\exp(\hbar\omega_{\mathbf{p}}/k_B T)}{[\exp(\hbar\omega_{\mathbf{p}}/k_B T) - 1]^2} \delta(\omega_{\mathbf{p}+\mathbf{q}} - \omega_{\mathbf{p}}). \quad (10)$$

In order to proceed further we need an expression for the magnon dispersion relation  $\hbar\omega_{\mathbf{q}}$ . The simplest choice is to write

$$\hbar\omega_{\mathbf{q}} = D_m q^2 + \Delta, \quad (11)$$

where  $D_m$  is the magnon stiffness constant and  $\Delta$  is the energy gap of the magnon dispersion relation. We have supposed that the dispersion relation is isotropic at small  $\mathbf{q}$ . The sum in Eq. (10) can be converted to an integral and can be computed analytically using Eq. (11). We obtain

$$\tilde{\Lambda}^{zz}(q) = \frac{V\hbar k_B T}{16\pi^2 D_m^2 q} \left[ \frac{1}{\exp[(D_m q^2/4 + \Delta)/k_B T] - 1} - \frac{1}{\exp[(D_m q_{BZ}^2 + \Delta)/k_B T] - 1} \right], \quad (12)$$

where  $k_B$  is the Boltzmann constant and  $q_{BZ}$  the radius of the Brillouin zone. We note that usually at low temperature we have  $(D_m q_{BZ}^2 + \Delta) \gg k_B T$ . This is the case, for instance, for GdNi<sub>5</sub> for which  $D_m = 3.2 \text{ meV \AA}^2$  and  $q_{BZ} = 0.9 \text{ \AA}^{-1}$  (see Ref. 4). Then the expression of the correlation function greatly simplifies:

$$\tilde{\Lambda}^{zz}(q) = \frac{V\hbar k_B T}{16\pi^2 D_m^2 q} \times \frac{1}{\exp[(D_m q^2/4 + \Delta)/k_B T] - 1}. \quad (13)$$

Notice that, because of the simple relation we take for the magnon dispersion relation, the correlation function is a function of the magnitude of  $\mathbf{q}$  and not of its orientation.

So far in this section we have not mentioned if we are dealing with Bravais or non-Bravais lattices. In a non-Bravais lattice where  $\tilde{\Lambda}^{zz}(\mathbf{q})$  writes  $\sum_{d,d'} \tilde{\Lambda}_{dd'}^{zz}(\mathbf{q})$ , the existence of more than one atom per unit cell gives rise to acoustic and optical branches in the dispersion relation. Because of the relatively huge gap present in the optical branch(es), the spin correlation functions  $\tilde{\Lambda}_{dd'}^{zz}(\mathbf{q})$  with  $d \neq d'$  are not relevant as seen at Eqs. (10) or (12).

The expression of the muon spin relaxation rate for the model considered in this section [specified by the dispersion relation given at Eq. (11)] is obtained using Eqs. (7) and (13). We derive

$$\lambda_z = \frac{C g_L^2 T^2}{D_m^3} P(q_{BZ}), \quad (14a)$$

with

$$P(q) = \ln \left[ \frac{1 - \exp[-(D_m q^2/4 + \Delta)/k_B T]}{1 - \exp(-\Delta/k_B T)} \right], \quad (14b)$$

where we have defined  $C = (2/15\pi) (\mu_0/4\pi)^2 \gamma_\mu^2 \mu_B^2 \hbar k_B^2 = 129.39 \text{ (meV)}^3 \text{ \AA}^6 \text{ s}^{-1} \text{ K}^{-2}$ . In the limiting case,  $(D_m q_{BZ}^2/4 + \Delta) > D_m q_{BZ}^2/4 \gg k_B T$ , we have

$$\lambda_z = \frac{C g_L^2 T^2}{D_m^3} \ln \left[ \frac{\exp(\Delta/k_B T)}{\exp(\Delta/k_B T) - 1} \right]. \quad (15)$$

A numerical study of the  $P(q)$  function shows that the expression for  $\lambda_z$  given at Eq. (15) is a reasonable approximation of the  $\lambda_z$  expression given at Eq. (14).

In practice we have  $\Delta \ll k_B T$ . In this high-temperature regime we obtain the simple result

$$\lambda_z = \frac{C g_L^2 T^2}{D_m^3} \ln(k_B T/\Delta). \quad (16)$$

The  $T^2 \ln(k_B T/\Delta)$  dependence of  $\lambda_z$  has been predicted in the past for an anisotropic contact interaction which can occur in nuclear magnetic resonance.<sup>14,15</sup> Some years ago we have derived this temperature dependence for muon spin relaxation rate using the model considered in the present work.<sup>6</sup> But the derivation was not sound mathematically: the constant  $G$  [introduced at Eq. (15) of Ref. 6] was undetermined.

The physical origin of the  $T^2$  factor in Eq. (16) is clear: each of the two magnons contributing to the muon spin depolarization process (Raman process) accounts for a factor  $T$ . We note that  $\lambda_z$  given by Eq. (16) diverges if  $\Delta = 0$ , i.e., for a pure Heisenberg magnet. This fact is not disturbing because a real magnet has always a small energy gap.<sup>16</sup>

Although the result of Eq. (16) has not been published before, it has already been used to extract the stiffness constant of the dipolar ferromagnet GdNi<sub>5</sub> from its zero muon spin relaxation rate data.<sup>17</sup>

$\lambda_z$  has been computed by other authors<sup>7</sup> for EuO. In the present work, we have presented a general framework to compute the  $\mu$ SR relaxation rate for a ferromagnet at low temperature. Then for a given magnon dispersion relation the rate has been computed explicitly. In the proposed framework the coupling between the muon spin and the electronic spins is described by the tensor  $G(\mathbf{q})$ . For a given muon localization site in a crystal, its elements are easily computed. One does not have to consider separately the relaxation due to the hyperfine and dipolar interaction.

#### IV. MUON SPIN RELAXATION IN A DIPOLAR HEISENBERG FERROMAGNET AT LOW TEMPERATURE

In the previous section we have computed  $\lambda_z$  for a simple Heisenberg Hamiltonian with an energy gap. In a dipolar magnet the gap is induced by the dipolar interaction between the ions. Recently the neutron scattering function has been computed for a dipolar Heisenberg magnet in the linear spin-wave approximation.<sup>18</sup> From this work an expression for the  $zz$  correlation function can be deduced. Relative to our expression the differences are that a relatively complicated weighting factor exists besides the thermal factor in Eq. (10) and the magnon dispersion relation for a dipolar Heisenberg magnet is used. However, qualitatively this factor does not influence the correlation function  $\tilde{\Lambda}^{zz}(\mathbf{q})$ . The magnon dispersion relation writes<sup>19</sup>

$$\omega_{\mathbf{q}}^2 = \varepsilon_{\mathbf{q}}(\varepsilon_{\mathbf{q}} + \epsilon \sin^2 \theta_{\mathbf{q}}), \quad (17)$$

where  $\theta_{\mathbf{q}}$  is the polar angle of wave vector  $\mathbf{q}$  relative to the easy axis,  $\varepsilon_{\mathbf{q}}$  the dispersion relation without the dipolar interaction, and  $\epsilon = (\mu_0 g_L \mu_B M_0) / \hbar$  a characteristic dipolar energy ( $M_0$  is the saturation magnetization). Therefore a natural choice is to identify the gap  $\Delta$  with  $\hbar\epsilon$ . In this case the effect of  $\sin^2 \theta_{\mathbf{q}}$  is neglected. Another

possibility is to take for the gap value the bulk magnetic anisotropy energy:  $\Delta = g_L \mu_B B_a$  where  $B_a$  is the magnetic anisotropy field. These two choices represent limiting cases. We note that for GdNi<sub>5</sub> we have  $\hbar\epsilon/k_B \sim 1.2$  K and  $(g_L \mu_B B_a)/k_B \sim 0.3$  K, i.e., there is a factor of 4 difference. But because the purpose of the measurement of the relaxation rate is to measure the spin-wave stiffness constant which is proportional to  $[\ln(k_B T/\Delta)]^{1/3}$ , this uncertainty by a factor of 4 will only introduce an error of  $\sim 20\%$  on the extracted  $D_m$  value from the relaxation data (in the case of GdNi<sub>5</sub> the  $D_m$  value is deduced from the relaxation data recorded at about 10–15 K).

In this work we have given a general framework to analyze positive muon longitudinal relaxation data at low temperature for a Heisenberg magnet. Using the simplest possible approximation, we have shown that the measurements allow one to estimate the magnon stiffness constant. The framework which is given here should allow one to deal with other problems in an easy way. We think for instance of antiferromagnets.

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