Wave-vector region probed by zero-field muon-spin-relaxation measurements in paramagnets near the Curie temperature

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We show that zero-field muon-spin-relaxation studies of the critical paramagnetic fluctuations in ferromagnets are mostly sensitive to fluctuation modes at small wave vectors. The transverse-fluctuation modes can be probed very close to the zone center when the temperature approaches the Curie temperature. A quantitative determination of the zone probed in Ni and Fe is performed.

I. INTRODUCTION

Recently, it has been shown that the muon-spin-depolarization rate measured in a Heisenberg paramagnet near the Curie temperature is a weighted sum of two functions which describe respectively the longitudinal (parallel to the wave vector **q** of the fluctuation mode) and transverse (perpendicular to **q**) fluctuations.[1,2] It is necessary to distinguish these two types of fluctuations because the dipolar interaction between the ion magnetic moments induces an anisotropy which has a strong influence on the dynamics near the zone center of the Brillouin zone. Note that the dipolar interaction always exists in real ferromagnets. Using a mode coupling approximation for the computation of the two functions,[3,4] a quantitative interpretation of the experimental muon-spin-depolarization data recorded on Ni and Fe has been given.[1,2]

Besides the anisotropy between longitudinal and transverse fluctuations, it is known that the dipolar interaction induces, in anisotropic lattice structures such as the hexagonal crystal structure of metallic Gd, an Ising anisotropy.[5] A recent analysis of the meaning of the muon-spin-relaxation rate in Gd shows that measurements of this rate in a single crystal should allow to probe the effect of the Ising anisotropy on the critical paramagnetic fluctuations.[6]

Zero-field muon-spin-relaxation measurements consist to measure the spin lattice relaxation rate of the spin of the muons which are spin 1/2 particles.[7,8] These particles are implanted in an interstitial site of the crystal under study. It is important to note that at the time of implantation they are polarized. This experimental technique, similar to other hyperfine techniques, does not probe the magnetic fluctuations at a given **q** but in a range of **q**. Because it has been shown that the mode coupling approximation provides a quantitative description of the fluctuations in Ni and Fe, it is possible to investigate in details the range of **q** probed in these magnets. This is the purpose of this paper.

The organization of this paper is as follows. In Sec. II we summarize the main results of our previous work. Our purpose is mainly to define our notations. In Sec. III we investigate the **q** region probed by zero-field muon-spin-relaxation measurements in cubic dipolar magnets near the Curie temperature. In the last section we apply the results of the previous section to analyze the Ni and Fe data. A conclusion of our work is also given in this section.

II. ZERO-FIELD MUON-SPIN-RELAXATION RATE IN DIPOLAR CUBIC PARAMAGNETS NEAR THE CURIE TEMPERATURE

A detailed presentation of the results summarized in this section can be found in Ref. 1.

We take the z axis to be parallel to the incoming muon beam polarization. A zero-field muon-spin-relaxation (μSR) measurement consists of measuring the time-depolarization function $P_z(t)$.[7,8] In the cases of interest here this function is an exponential function. We write

$$P_z(t) = \exp(-\lambda_z t)\,, \tag{1a}$$

where we have defined the relaxation rate λ_z:

$$\lambda_z = \frac{\gamma_\mu^2}{2}\int_{-\infty}^{\infty} d\tau\,[\Phi_{xx}(\tau)+\Phi_{yy}(\tau)]\,. \tag{1b}$$

γ_μ is the muon gyromagnetic ratio: $\gamma_\mu = 851.6$ Mrad s^{-1} T^{-1}. Here $\Phi_{\alpha\alpha}(\tau)$ is the symmetrized time correlation function of the α component of the local magnetic field at the muon site. It is possible to express this magnetic field as a function of, on the 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, λ_z can be written as a linear combination of spin-spin correlation functions of the magnet. The coefficients of this linear combination are expressed in terms of the tensor mentioned just above. Because we are interested in describing the spin dynamics near the Curie temperature, we have to consider the $\mathbf{q}$ dependence of the tensor and the spin-spin correlation function near the Brillouin zone center, i.e., near $\mathbf{q} = \mathbf{0}$. It occurs that near $\mathbf{q} = \mathbf{0}$ these two quantities have quite simple symmetry properties. They can be expressed in terms of the transverse and longitudinal projection operators. Using the orthogonality property of these two operators it is possible to show that the relaxation rate is given as a weighted sum of two terms: The first one describes the contribution to λ_z of the fluctuation modes longitudinal to $\mathbf{q}$, the second the contribution of the modes transverse to $\mathbf{q}$. As mentioned in the Introduction it is necessary to distinguish these two types of modes because of the dipolar interaction between the magnetic ions. λ_z can then be written as a linear combination of the integrals $\breve{\lambda}^L$ and $\breve{\lambda}^T$ defined by

$$\breve{\lambda}^{L,T} = \int_0^{q_R = q_{\rm BZ}} dq\, q^2 \frac{\chi^{L,T}(q)}{\Gamma^{L,T}(q)}. \tag{2}$$

$\chi^L(q)$ $[\chi^T(q)]$ and $\Gamma^L(q)$ $[\Gamma^T(q)]$ are respectively the longitudinal [transverse] susceptibility and linewidth functions. The boundaries of the integrals defined in Eq. (2) are such that the computation is performed inside a sphere whose center and radius q_R are respectively the center and the radius of the Brillouin zone ($q_R = q_{\rm BZ}$). The aim of this paper is to study the sensitivity of the integral values to the upper boundary q_R. Using the susceptibility and linewidth scaling functions $\hat{\chi}^{L,T}(r,\varphi)$ and $\hat{\Gamma}^{L,T}(r,\varphi)$ we rewrite the expressions given in Eq. (2) changing the integration variable. It is then practical to define new functions $I^{L,T}(\hat{q}_R,\varphi)$ proportional to $\breve{\lambda}^{L,T}$. We set

$$I^{L,T}(\hat{q}_R,\varphi) = \left[1 + \frac{1}{\tan^2\varphi}\right]^{-3/4} \times \int_{b_{\rm min}}^{\infty} dr\, r^{1/2} \frac{\hat{\chi}^{L,T}(r,\varphi)}{\hat{\Gamma}^{L,T}(r,\varphi)}, \tag{3}$$

where $\varphi = \arctan(q_D\xi)$, $\hat{q}_R = q_R/q_D$, and $b_{\rm min} = (1+\tan^{-2}\varphi)^{1/2}/\hat{q}_R$. The angle φ is a measure for the temperature through its dependence on the correlation length ξ. q_D is the dipolar wave vector determined by the relative strengths of the dipolar and exchange interactions. We use the Ornstein-Zernike forms for the susceptibilities and the linewidth functions are calculated by a mode-coupling approximation as explained in Refs. 3 and 4. The lower cutoff of the integrals in Eq. (3) is taken as 0 in the critical region (q_D is very small compared to $q_{\rm BZ}$). For simplicity, from now on we will use the definition $I^{L,T}(\varphi) \equiv I^{L,T}(\hat{q}_R = \infty, \varphi)$. For completeness, in Fig. 1 we present the φ dependence of these functions.

The λ_z expression finally writes

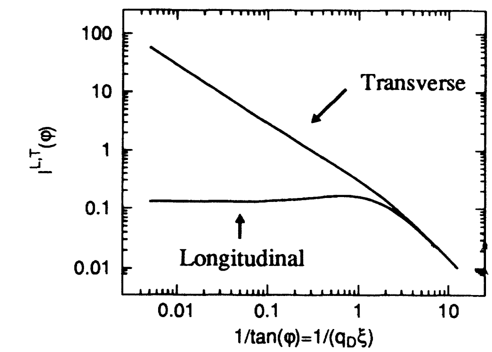

FIG. 1. φ (temperature) dependence of the functions $I^{L,T}(\varphi)$ which describe respectively the contribution to the relaxation rate of the longitudinal and transverse fluctuations.

$$\lambda_z = \mathcal{W}\left[a_L I^L(\varphi) + a_T I^T(\varphi)\right], \tag{4}$$

where $\mathcal{W}$ is a nonuniversal constant, a_L and a_T are material parameters, and $I^L(\varphi)$ and $I^T(\varphi)$ simply describe the contribution to λ_z of respectively the longitudinal- and transverse-fluctuation modes.

III. *Q*-SPACE REGION PROBED BY THE LONGITUDINAL- AND TRANSVERSE-FLUCTUATION FUNCTIONS

The $I^{L,T}(\varphi)$ functions, which we will call fluctuation functions, are given in terms of integrals over the whole Brillouin zone. The integrands depend on the susceptibilities which are functions with dominant values near the zone center. Therefore we suspect that $I^{L,T}(\varphi)$ are mainly determined by the fluctuation modes near the zone center.

One can qualitatively determine the width of the dominant region in the integrals given in Eq. (2). If we suppose that the linewidth functions are proportional to q^2 (which is strictly valid only in the asymptotic dipolar regime of the transverse fluctuations), we have

$$\breve{\lambda}^i \propto \int_0^{q_{\rm BZ}} \frac{dq}{q^2 + \delta_{L,i} q_D^2 + \xi^{-2}}, \tag{5}$$

where we have used the Ornstein-Zernike forms for the static susceptibilities. It is easily seen in Eq. (5) that the width of the dominant region in the integrals is proportional to $(q_D^2 + \xi^{-2})^{1/2}$ and ξ^{-1} for the longitudinal and transverse fluctuations respectively. Hence this contribution would imply that the dominant wave-vector region becomes closer and closer to $q = 0$ with T approaching T_C for the transverse relaxation rate and saturates at a finite size proportional to q_D for the longitudinal relaxation rate. Actually one has to take into account the wave-vector dependence of the longitudinal and transverse linewidth $\Gamma^{L,T}$ to determine the dominant wave-vector region precisely. In order to do so we define the ratio

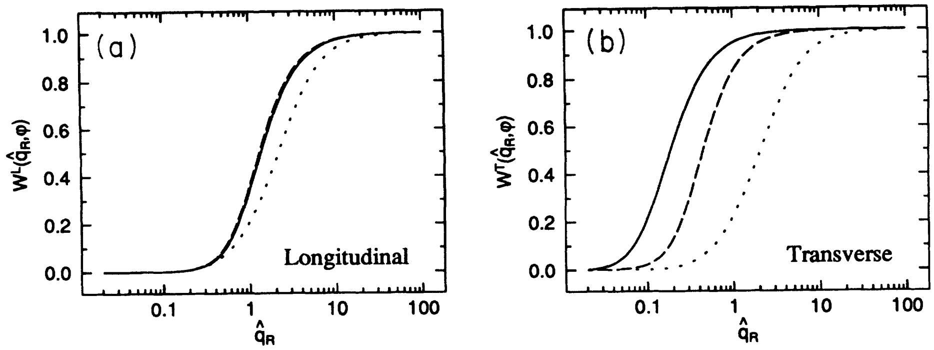

FIG. 2. Relative contribution, as measured by the $W^L(\hat{q}_R,\varphi)$ and $W^T(\hat{q}_R,\varphi)$ functions, of the longitudinal (a) and transverse (b) fluctuation modes to the longitudinal- and transverse-fluctuation functions respectively. The integration in the Brillouin zone is performed from the zone center to $\hat{q}_R$. $1/\tan\varphi$ is inversely proportional to the correlation length ξ; i.e., it increases with the relative temperature $(T-T_C)/T_C$ where T_C is the Curie temperature. The solid, dashed, and dotted lines are calculated for $1/\tan\varphi = 0.1$, 0.3, and 3 respectively.

$$W^{L,T}(\hat{q}_R,\varphi) = \frac{I^{L,T}(\hat{q}_R,\varphi)}{I^{L,T}(\varphi)} \tag{6}$$

and its derivative

$$S^{L,T}(\hat{q}_R,\varphi) = \frac{\partial W^{L,T}(\hat{q}_R,\varphi)}{\partial \hat{q}_R}. \tag{7}$$

$W^L(\hat{q}_R,\varphi)$ $[W^T(\hat{q}_R,\varphi)]$ represents the relative contribution to the longitudinal- [transverse-] fluctuation function of the longitudinal- [transverse-] fluctuation modes with q comprised between 0 and $\hat{q}_R$. The $W^{L,T}(\hat{q}_R,\varphi)$ and $S^{L,T}(\hat{q}_R,\varphi)$ functions are presented in Figs. 2(a), 2(b) and Figs. 3(a), 3(b) respectively for different values of the angle φ, i.e., different values of the temperature relative to the Curie temperature. Closer to T_C, smaller is $1/\tan\varphi$. The $W^{L,T}(\hat{q}_R,\varphi)$ and $S^{L,T}(\hat{q}_R,\varphi)$ functions give the same type of information. Choosing one of the two functions to investigate the q region probed by a fluctuation function is somewhat a question of convenience.

From Figs. 2 and 3 we deduce that the fluctuation functions probe a small wave-vector region. Figure 2 gives information on the wave-vector range needed in the integration to reach a certain percentage of the integral full value. Together with Fig. 1, it will be useful when determining the q region probed for a given material as shown in Sec. IV.

Figure 3 nicely indicates which region is probed by the fluctuation functions. As displayed by Fig. 3(a) the most important contribution to the longitudinal relaxation rate I^L comes from the wave-vector region near $q \simeq q_D$ $(\hat{q}_R \simeq 1)$. On the other hand Fig. 3(b) shows that the region which mostly contributes to the transverse fluctuations is for wave vectors close to the zone center if the temperature is close to T_C, but shifts to larger wave vectors as one goes away from criticality (i.e., with increasing temperature).

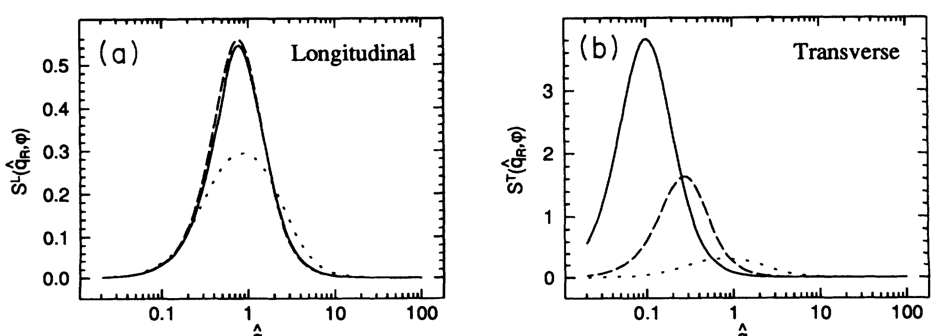

FIG. 3. Relative contribution, as measured by the $S^L(\hat{q}_R,\varphi)$ and $S^T(\hat{q}_R,\varphi)$ functions, of the longitudinal (a) and transverse (b) fluctuation modes to the longitudinal- and transverse-fluctuation functions respectively. $S^{L,T}(\hat{q}_R,\varphi)$ is the derivative of $W^{L,T}(\hat{q}_R,\varphi)$ with respect to $\hat{q}_R$. $W^{L,T}(\hat{q}_R,\varphi)$ are displayed in Fig. 2. The meaning of the different lines is explained in the caption of Fig. 2.

TABLE I. Radius of the q-space region probed by zero-field muon-spin-relaxation measurements in Ni. When the integration of the longitudinal- and transverse-fluctuation modes is performed within a sphere whose radius is given in column "50% of λ_z" (respectively "90% of λ_z") we get 50% (90%) of the λ_z value.

$1/\tan\varphi$	$T - T_C$ (K)	50% of λ_z (Å^{-1})	90% of λ_z (Å^{-1})
0.1	0.056	0.003	0.022
0.3	0.27	0.009	0.036
3	7.7	0.027	0.094

TABLE II. Same data as Table I but relative to Fe.

$1/\tan\varphi$	$T - T_C$ (K)	50% of λ_z (Å^{-1})	90% of λ_z (Å^{-1})
0.1	0.20	0.007	0.035
0.3	0.97	0.018	0.069
3	27	0.071	0.240

IV. Q-SPACE REGION PROBED BY ZERO-FIELD MUON-SPIN-RELAXATION RATE IN Ni AND Fe

In Refs. 1 and 2 it has been shown that Eq. (4) gives a good description of the available data recorded in Ni and Fe. In this section we investigate the **q** region probed by these data. We will show by an example how we proceed for this estimate. Then we will give the estimates for Ni and Fe at three different temperatures in the form of tables.

We consider a measurement performed on Ni at a temperature for which $1/\tan\varphi = 0.1$. We first compute the corresponding temperature. From the values of q_D (0.013 Å^{-1}) and of the correlation length at $T = 2T_C$ (1.23 Å) listed in Ref. 1 we deduce that $(T - T_C) = 0.056$ K. The Ni data are well described by Eq. (4) with $a_L = 0.603$ and $a_T = 0.100$.[1] From Fig. 1 we deduce that at $1/\tan\varphi = 0.1$ we have $I^L(\varphi) = 0.135$ and $I^T(\varphi) = 3.00$. Thus we find that $a_L I^L(\varphi) = 0.081$ and $a_T I^T(\varphi) = 0.300$ and therefore $a_L I^L(\varphi) + a_T I^T(\varphi) = 0.381$. Note that this value is proportional to the λ_z value which is computed with the q integration performed over the whole Brillouin zone. Now suppose we are looking for the value of $\hat{q}_R$ which gives 50% of the λ_z value. We find that $\hat{q}_R = 0.252$. This is done as follows. For the values of $\hat{q}_R$ and $1/\tan\varphi$ considered we deduce, using Fig. 2(a) and Fig. 2(b), that $W^L(\hat{q}_R, \varphi) = 0.0161$ and $W^T(\hat{q}_R, \varphi) = 0.630$ respectively. From these values we compute that $W^L(\hat{q}_R, \varphi) \times a_L I^L(\varphi) = 0.0013$ and $W^T(\hat{q}_R, \varphi) \times a_T I^T(\varphi) = 0.189$. The sum of these two quantities gives 0.190. This is approximately half of the value obtained when the integration is performed over the whole Brillouin zone.

This example clearly shows that one probes the zone center fluctuations only if the transverse fluctuations dominate. This occurs when the temperature is sufficiently close to T_C (except in the case where $a_T = 0$). Using the method just explained it is possible for different temperatures to compute $\hat{q}_R$ for a given percentage of the λ_z value originated from a sphere of radius q_R. Using the relation $\hat{q}_R = q_R/q_D$ and the value of q_D we can estimate the radius of the q-space region probed by zero-field muon-spin-relaxation measurements. Some results are collected in Tables I and II for Ni and Fe respectively. It is well known that it is possible to obtain information on the q dependence of the fluctuation modes from neutron scattering measurements. At present the smallest q value for which measurements can be performed seems to be 0.01 Å^{-1}.[9] This is comparable to the values given in the tables.

In conclusion we have shown that zero-field muon-spin-relaxation studies of the critical paramagnetic fluctuations in ferromagnets allow one to probe the fluctuation modes at small q vectors. Quantitative estimates have been given for Ni and Fe. The results are summarized in the tables. We note that, as stressed in Ref. 1, these studies are performed in truly zero-field condition. It may even be possible to study directly the longitudinal fluctuations in zero-field.[6]

We note that the available μSR data on Ni and Fe have not been recorded very near the Curie temperature. Such data would be useful to test the validity of the Hamiltonian used in Refs. 1 and 2 to describe the magnetic fluctuations at small wave-vector values.

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