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Probing magnetic excitations, fluctuations and correlation lengths by muon spin relaxation and rotation techniques

P Dalmas de Réotier[1], **P C M Gubbens**[2] **and A Yaouanc**[1]

[1] Commissariat à l'Energie Atomique, Département de Recherche Fondamentale sur la Matière Condensée, F-38054 Grenoble cedex 9, France
[2] Interfacultair Reactor Instituut, 2629 JB Delft, The Netherlands

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Abstract

We first discuss magnetic excitation and relaxation processes which have already been recognized experimentally by muon spin relaxation experiments. Our examples are taken from experiments performed on strongly correlated electronic systems, magnetic metals and geometrically frustrated magnetic compounds. In another part, focusing on the flux-line lattice in the Bragg-glass phase of superconductors, we show that a muon spin experiment can provide information on the in-plane correlation length of the lattice.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

In this article we have chosen to focus on some mechanisms already recognized experimentally which may induce a relaxation of the muon spin in magnetic materials as observed by positive muon spin (μSR) experiments. This relaxation is studied in the best way by the longitudinal field technique which allows us to measure the so-called longitudinal polarization function, $P_Z(t)$ (see e.g. Schenck and Gygax 1995, Karlsson 1995). Z is the axis of the initial muon beam polarization and also the direction of the applied external field, $\mathbf{B}_{\rm ext}$, if any. In fact, here we shall assume $\mathbf{B}_{\rm ext} = \mathbf{0}$ for the measurement of $P_Z(t)$. In addition, we report on a recent theoretical development which indicates the possibility of measuring the in-plane correlation length of the Bragg-glass phase of the flux-line lattice of a superconductor. In this case the transverse-field μSR technique is used (see e.g. Sonier *et al* 2000), in which $\mathbf{B}_{\rm ext}$ is still applied along the Z axis but now it is the polarization function $P_X(t)$ which is measured. The X axis is perpendicular to Z.

In the next section we provide a theoretical background to understand the relation between the muon spin–lattice relaxation rate, λ_Z, and the magnetic correlation tensor or the generalized susceptibility tensor. We restrict ourselves to homogeneous magnetic systems for which $P_Z(t)$

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is basically expected to be an exponential function characterized by λ_Z, if the dynamics is sufficiently fast. Some experimental examples are presented in the third section. This is followed by a discussion of the field distribution in superconductors. In the last section we have gathered some conclusions.

A large number of μSR reviews on magnetism have been written. We mention three of them which are easily accessible: Dalmas de Réotier and Yaouanc (1997), Amato (1997) and Sonier *et al* (2000). The first review covers magnetic materials in general, whereas the last two are specialized: on strongly correlated electronic systems and on vortex matter in superconductors, respectively. We mention that a monograph discussing the applications of the μSR techniques to condensed matter science by Yaouanc and Dalmas de Réotier is in preparation.

2. Excitations and fluctuations; theoretical background

Since our main purpose here is only to give a feeling of the physical meaning of λ_Z, for simplicity we shall assume a single-crystal sample with one magnetic ion per unit cell and crystallographically oriented such that the relevant axes of the crystal and of the laboratory are parallel. As for nuclear magnetic resonance (NMR), we can write λ_Z as an integral over the first Brillouin zone (see e.g. Dalmas de Réotier and Yaouanc 1997):

$$\lambda_Z = \frac{\mathcal{D}}{2}\frac{1}{V}\int \sum_{\beta,\gamma} \mathcal{A}^{\beta\gamma}(\mathbf{q})\, \Lambda^{\beta\gamma}(\mathbf{q}, \omega_\mu = 0)\frac{\mathrm{d}^3\mathbf{q}}{(2\pi)^3}, \tag{1}$$

where we have defined for convenience

$$\mathcal{A}^{\beta\gamma}(\mathbf{q}) = G^{X\beta}(\mathbf{q})G^{X\gamma}(-\mathbf{q}) + G^{Y\beta}(\mathbf{q})G^{Y\gamma}(-\mathbf{q}). \tag{2}$$

By definition $\{\beta, \gamma\} = \{X, Y, Z\}$ and $\mathcal{D} = (\mu_0/4\pi)^2\, \gamma_\mu^2\, (g\mu_\mathrm{B})^2$ where g is the spectroscopic splitting factor, e.g. the Landé factor for a rare earth, μ_B is the electronic Bohr magneton and γ_μ the muon gyromagnetic ratio ($\gamma_\mu = 851.6$ Mrad s^{-1} T^{-1}). V is the volume of the sample. We use the international system of units. λ_Z depends basically on two quantities:

(i) the coupling between the muon spin and the spins of the compound encoded in $\boldsymbol{G}$, and
(ii) the correlation tensor $\boldsymbol{\Lambda}$ (of spins $\mathbf{J}$) we are interested in.

That tensor is probed at $\omega_\mu = \gamma_\mu B_\mathrm{loc}$ where $\mathbf{B}_\mathrm{loc}$ is the local field at the muon site. In this work we assume $\omega_\mu = 0$. This is strictly correct for zero-field measurements performed outside the ordered phases of a magnetic material and only an approximation when an ordered state is probed. Interpreting λ_Z as the muon spin–lattice relaxation rate, equation (1) is a well known NMR expression (see e.g. Moriya 1962).

We shall need $G^{\alpha\beta}(\mathbf{q})$. These tensor elements are not analytical near $\mathbf{q} = \mathbf{0}$. This is due to the long range nature of the dipole interaction between the muon spin and the unpaired electron spins of the compound (Yaouanc *et al* 1993). From this reference we write

$$G^{\alpha\beta}(\mathbf{q} \longrightarrow \mathbf{0}) = -4\pi\left[P_L^{\alpha\beta}(\mathbf{q}) - C^{\alpha\beta}(\mathbf{q} = \mathbf{0}) - \frac{r_\mu H}{4\pi}\delta^{\alpha\beta}\right], \tag{3}$$

where $P_L^{\alpha\beta}(\mathbf{q}) = q^\alpha q^\beta/q^2$ is an element of the longitudinal projection operator, $C^{\alpha\beta}(\mathbf{q})$ is analytical for all values of $\mathbf{q}$, r_μ the number of nearest neighbour magnetic ions to the muon localization site and H the hyperfine constant. It is $\boldsymbol{P}_\mathrm{L}(\mathbf{q})$ which takes care of the non-analyticity of $\boldsymbol{G}(\mathbf{q})$ near the zone centre since it is only piecewise continuous as $\mathbf{q} \longrightarrow \mathbf{0}$. $\boldsymbol{C}(\mathbf{q} = \mathbf{0})$ can be a scalar, i.e. $C^{\alpha\beta}(\mathbf{q} = \mathbf{0}) = (1/3)\delta^{\alpha\beta}$. This particular form holds for example for a muon in a tetrahedral or octahedral site of a face centred cubic crystal.

A theorem, known as the fluctuation–dissipation (Nyquist's) theorem, links thermal fluctuations of the spins described by a correlation function to a related generalized susceptibility function. Since here we focus on $\omega_\mu = 0$, the expression of the theorem is relatively simple (see e.g. Lovesey 1986):

$$\Lambda^{\alpha\beta}(\mathbf{q}, \omega_\mu = 0) = \frac{2V}{\mu_0 g^2 \mu_B^2} k_B T \lim_{\omega_\mu \longrightarrow 0} \frac{\mathrm{Im}\{\chi^{\alpha\beta}(\mathbf{q}, \omega_\mu)\}}{\omega_\mu}. \tag{4}$$

Taking $\mathbf{\Lambda}$ to be a scalar and assuming the correlation to decay exponentially in time,

$$\Lambda^{\alpha\alpha}(\mathbf{q}, \omega_\mu = 0) = \frac{2V}{\mu_0 g^2 \mu_B^2} k_B T \frac{\chi^{\alpha\alpha}(\mathbf{q})}{\Gamma^\alpha(\mathbf{q})}. \tag{5}$$

The time-integrated correlation function of mode $\mathbf{q}$ is therefore proportional to the wavevector-dependent static susceptibility of that mode and inversely proportional to the relaxation rate of the same mode. That rate, also called the linewidth, is the half width at half maximum of the quasi-elastic excitation. The reader has probably noticed that we are using the language of scientists involved in scattering experiments. It is also suitable for describing results from local probe techniques such as the μSR ones. This has long been recognized by people practicing solid state NMR.

We have chosen to start our investigation of processes which can induce a relaxation of the muon spin by discussing the magnon excitation process.

2.1. Magnon excitations

We first consider a simple ferromagnet with isotropic exchange interactions limited to nearest neighbour magnetic ions. The Hamiltonian for that system is written as

$$\mathcal{H}_{\mathrm{sys}} = -\mathcal{J} \sum_{i,i'} \mathbf{J}_i \cdot \mathbf{J}_{i'} + g\mu_B B_{\mathrm{ani}} \sum_i J_i^Z \qquad \text{with } \mathcal{J} > 0. \tag{6}$$

For a rare-earth ion the exchange constant $\mathcal{J}$ has to be replaced by $\mathcal{J}(g-1)$. The anisotropy of the system is accounted for by the anisotropy field B_{ani} assumed to be parallel to the Z axis. We shall find important to include that term to get a mathematically sound result. It is justified physically since a ferromagnet is always anisotropic. An obvious source of anisotropy is the dipole interaction between the magnetic ions. The form introduced for the anisotropy field is not always mathematically justified, in particular for the dipole interaction. However, it is simple enough that a reliable physical result is easily obtained. Since the energy of an excitation is minimum at the zone centre, the sum over $\mathbf{q}$ in (1) is dominated by the small $\mathbf{q}$ region. Hence, it is justified to use the small $\mathbf{q}$ limit of $\boldsymbol{G}(\mathbf{q})$. In the same limit and for a cubic lattice of cube edge a, the magnon dispersion relation has a well known expression (see e.g. Lovesey 1986):

$$\hbar\omega_{\mathbf{q}} = g\mu_B B_{\mathrm{ani}} + D_{\mathrm{FM}} q^2 \qquad \text{with } D_{\mathrm{FM}} = 2\mathcal{J} J a^2. \tag{7}$$

We use the linear spin-wave approximation.

Because of energy conservation, two magnons are required to relax the muon spin (Yaouanc and Dalmas de Réotier 1991). One magnon is annihilated and a magnon is created by the magnon–muon spin scattering. We are therefore dealing with a Raman scattering process. This means that only $\Lambda^{ZZ}(\mathbf{q}, 0)$ contributes to the relaxation at the level of the linear spin-wave approximation. As a consequence, the hyperfine coupling which is usually isotropic is not relevant to the magnon-induced relaxation. This is derived from (2) since only the non-diagonal terms of $\boldsymbol{G}$ contribute to the relaxation as $\beta = \gamma = Z$. The following formula is

derived:

$$\lambda_Z = 2(2\pi)^3\hbar\mathcal{D}\left\{\tfrac{2}{15} + \left[C^{XZ}(\mathbf{q}=\mathbf{0})\right]^2 + \left[C^{YZ}(\mathbf{q}=\mathbf{0})\right]^2\right\} \times \frac{1}{V^2}\int n\left(\frac{E}{k_\mathrm{B}T}\right)\left[n\left(\frac{E}{k_\mathrm{B}T}\right)+1\right] g_\mathrm{m}^2(E)\,\mathrm{d}E. \tag{8}$$

We have introduced the Bose function

$$n\left(\frac{E}{k_\mathrm{B}T}\right) = \frac{1}{\exp\left(\frac{E}{k_\mathrm{B}T}\right)-1}, \tag{9}$$

and the density of magnetic excitations $g_\mathrm{m}(E)$. We display explicitly the temperature dependence of the Bose function to indicate the origin of the temperature dependence of the spin–lattice relaxation. This result is written in a physically appealing form: the two Bose functions stand for the creation and annihilation of the two excitations and a density of states is associated with each of them. $g_\mathrm{m}(E)$ can be computed for the dispersion relation given at (7). Finally we get

$$\lambda_Z = \frac{\hbar\mathcal{D}k_\mathrm{B}^2}{\pi}\left\{\frac{2}{15} + \left[C^{XZ}(\mathbf{q}=\mathbf{0})\right]^2 + \left[C^{YZ}(\mathbf{q}=\mathbf{0})\right]^2\right\}\frac{T^2}{D_\mathrm{FM}^3}\ln\left(\frac{k_\mathrm{B}T}{g\mu_\mathrm{B}B_\mathrm{ani}}\right), \tag{10}$$

for temperatures such that $g\mu_\mathrm{B}B_\mathrm{ani} \ll k_\mathrm{B}T \ll E_\mathrm{max}$ where E_max is the maximum energy of the magnons (Dalmas de Réotier and Yaouanc 1995); for temperatures such that $g\mu_\mathrm{B}B_\mathrm{ani} \geqslant k_\mathrm{B}T$ one finds that there is no magnon-induced relaxation, the anisotropy energy being so large relative to the thermal energy that the system is frozen in its ground state. λ_Z is found to be approximately quadratic in temperature as already discovered by Mitchell in 1957 for NMR. From the analysis of λ_Z, the value of the magnon stiffness constant can be extracted.

A similar formula to that written at (8) is also valid for antiferromagnets. The main difference is that $g_\mathrm{m}^2(E)$ must be substituted with $g_\mathrm{m}(E)h_\mathrm{m}(E)$ where $h_\mathrm{m}(E)$ is a weighted density of states. The fact that such a density appears instead of a simple density is not unexpected. For a now famous example in the context of NMR, see Mila and Rice (1989).

Obviously, magnons are also basic excitations in itinerant magnetic systems. The framework we have presented can be used to describe their effect on λ_Z.

2.2. Localized spin fluctuations

We now shift our interest to the paramagnetic state, focusing first on systems characterized by localized magnetic properties. A key quantity is the static wavevector-dependent susceptibility $\chi_{\mathbf{Q}_0}(\mathbf{q})$. $\mathbf{Q}_0$ denotes the wavevector of the magnetic structure; $\mathbf{Q}_0 = \mathbf{0}$ for a ferromagnet. Explicitly, in the molecular field approximation

$$\chi_{\mathbf{Q}_0}(\mathbf{q}) = \frac{\mu_0}{v_0}\frac{g^2\mu_\mathrm{B}^2 J(J+1)}{3k_\mathrm{B}T_\mathrm{c}}\frac{1}{\delta + \left[1 - \frac{\mathcal{J}(\mathbf{q})}{\mathcal{J}_{\mathbf{Q}_0}}\right]}, \tag{11}$$

with $\delta = (T - T_\mathrm{c})/T_\mathrm{c}$. T_c is the Curie (Néel) temperature for an (anti-)ferromagnet. v_0 is the volume per magnetic ion and $\mathcal{J}(\mathbf{q})$ the Fourier transform of the exchange interaction,

$$\mathcal{J}(\mathbf{q}) = \mathcal{J}\sum_{\mathbf{i}}\exp\left(-\mathrm{i}\mathbf{q}\cdot\mathbf{i}\right). \tag{12}$$

We define $\mathcal{J}_{\mathbf{Q}_0} = \mathcal{J}(\mathbf{Q}_0)$. If $\delta \gg 1$, i.e. if the temperature is much higher than T_c, $\chi_{\mathbf{Q}_0}(\mathbf{q})$ becomes wavevector independent and is given by the well known Curie–Weiss law:

$$\chi_0 = \frac{\mu_0}{v_0}\frac{g^2\mu_\mathrm{B}^2 J(J+1)}{3k_\mathrm{B}\left(T - T_\mathrm{c}\right)}. \tag{13}$$

χ_0 is the uniform susceptibility. In the same limit the linewidth is also temperature independent; see e.g. Lovesey (1986). Hence, λ_Z becomes temperature independent at high temperature. Neglecting the hyperfine interaction,

$$\lambda_Z = 2\Delta_{\mathrm{ZF}}^2 \tau_{\mathrm{ex}} \qquad \text{with } \Delta_{\mathrm{ZF}}^2 = \left(\frac{\mu_0}{4\pi}\right)^2 \gamma_\mu^2 g^2 \mu_{\mathrm{B}}^2 \frac{J(J+1)}{3} \sum_{i=1}^{N} \frac{1}{r_i^3}\left(\frac{5-3\cos^2\theta_i}{2}\right). \tag{14}$$

θ_i is the polar angle of the radius vector $\mathbf{r}_i$ connecting the muon to ion i. The exchange fluctuation time is $\tau_{\mathrm{ex}} = \sqrt{\pi/2}/\omega_{\mathrm{ex}}$ with

$$\omega_{\mathrm{ex}} = \sqrt{\frac{8\mathcal{J}^2 z J(J+1)}{3\hbar^2}}, \tag{15}$$

where z is the number of nearest magnetic ions surrounding each magnetic ion. This is a result published by Moriya (1956a and 1956b).

As a matter of fact, the predicted temperature independence of λ_Z at high temperature is not always observed because the localized magnetic moments may also be relaxed by the phonons or the conduction electrons of the specimen. These relaxation channels usually involve the crystal field energy levels of the ions, in particular in compounds containing rare-earth ions. Here, following Hartmann *et al* (1986), we discuss the relaxation of a spin-1/2 ion arising from its interaction with the conduction electron density. This is the Korringa relaxation mechanism. We can still write $\lambda_Z = 2\Delta_{\mathrm{ZF}}^2 \tau_{\mathrm{c}}$, but with the correlation time τ_{c} given by the formula

$$\frac{1}{\tau_{\mathrm{c}}} = \frac{1}{\tau_{\mathrm{ex}}} + \frac{1}{\tau_{\mathrm{f-e}}}, \tag{16}$$

where $\tau_{\mathrm{f-e}}^{-1}$ is the ion–electron relaxation rate. As it is well known,

$$\frac{1}{\tau_{\mathrm{f-e}}} = \frac{4\pi}{\hbar}\left[(g-1)\,\mathcal{J}_{\mathrm{f-e}}\, n(E_{\mathrm{F}})\right]^2 k_{\mathrm{B}} T. \tag{17}$$

Here, $n(E_{\mathrm{F}})$ is the conduction-electron density of states per spin direction and $\mathcal{J}_{\mathrm{f-e}}$ the exchange interaction between the ion spin, $\mathbf{J}_i$, and the conduction-electron spin at the ion site so that the Hamiltonian is written

$$\mathcal{H}_{\mathrm{f-e}} = -\mathcal{J}_{\mathrm{f-e}} \mathbf{J}_i \cdot \boldsymbol{\sigma} \tag{18}$$

where $\boldsymbol{\sigma}$ is the Pauli vector operator for the conduction-electron spin. A stable f electronic configuration is assumed, i.e. there is no Kondo effect.

As first pointed out by Silbernagel *et al* in 1968 (see also Hartmann *et al* 1986), the sign of the exchange interaction may have a profound effect on $\lambda_Z(T)$. An expansion in $1/T$ for the Heisenberg Hamiltonian leads to

$$\lambda_Z(T) = \lambda_Z^{(\infty)}\left(1 + \alpha \frac{\theta_{\mathrm{CW}}}{T}\right), \tag{19}$$

where $\lambda_Z^{(\infty)}$ is the value of the spin–lattice relaxation rate computed from (14) and α is a constant which, within an order of magnitude, is equal to one. θ_{CW} is the Curie–Weiss temperature which is positive for ferromagnets and negative for antiferromagnets. Hence, as the sample is cooled, λ_Z increases or decreases if the muons probe ferromagnetic or antiferromagnetic pair correlations among its neighbours, respectively. The weight of the second term on the right-hand side of (19) depends drastically on the spin configuration around the muon and may even vanish. For the sake of simplicity, let us assume the muon spin to interact only with its nearest neighbour spins in the lattice. If there is only one such atom nearby the muon, pair correlations cannot influence the muon spin and $\alpha = 0$. On the other hand, if two magnetic atoms interact with the muon, $\alpha \neq 0$.

2.3. Crystal field excitations

As just pointed out, in f electron systems crystal electric field (CEF) effects are expected to be important. A framework to deal with them is sketched elsewhere (Dalmas de Réotier *et al* 1996). Here, we assume a system of paramagnetic ions at high temperature with a CEF energy level located at an energy much higher than the thermal energy of the system. The f magnetic moments may fluctuate through a two excitation process. The two excitations involved are usually phonons and then we are dealing with a so-called Orbach process (Orbach 1961). In this simple case $\lambda_Z^{-1} = A + B_{\rm me}\exp[-\Delta_{\rm e}/(k_{\rm B}T)]$. $\Delta_{\rm e}$ is the energy of the excited CEF level involved. $A \simeq \lambda_Z^{-1}$ if the temperature is not too high. At higher temperature λ_Z decreases. The constant $B_{\rm me}$ models the magneto-elastic coupling of the f moments with the phonon bath.

2.4. Itinerant spin fluctuations

There is a relationship between λ_Z and the muon Knight shift, K_μ, derived a long time ago by Korringa (1950):

$$\lambda_Z = \frac{4\pi k_{\rm B} T K_\mu^2}{\hbar}\left(\frac{\gamma_\mu}{\gamma_{\rm e}}\right)^2. \tag{20}$$

This law written originally for NMR is approximately obeyed experimentally. However, this relaxation mechanism is usually not strong enough to induce a visible muon-spin relaxation. This can be understood because the muon Knight shift is usually small relative to the shift observed in NMR.

There is a family of ferromagnetic and antiferromagnetic metallic compounds, the so-called weak itinerant magnets, characterized by relatively low ordering temperatures and small magnetic moments. Their magnetic properties arise from their conduction electrons and their long-range order is lost due to the vanishing amplitude of the magnetic moments at the ordering point. This behaviour is in marked contrast to a compound with localized electrons for which the long-range order is destroyed by thermal excitations, i.e. by spin waves. Originally, these metals comprised only compounds based on 3d elements. However, recently intermetallics made of f elements have been found to exhibit some similar magnetic properties. The description of these itinerant magnets rests on the recognition of the importance of the low-energy spin fluctuations, using either a self-consistent renormalization theory (SCR, Moriya 1985) or a phenomelogical Landau–Ginzburg expansion (Lonzarich and Taillefer 1985). We shall assume isotropic fluctuations.

Since only long-wavelength fluctuations matter, the Ornstein–Zernike approximation for the susceptibility, i.e. the long-wavelength limit of (11), is believed to be adequate in the whole temperature range above the ordering point:

$$\chi_{\mathbf{Q}_0}(\mathbf{q}) = \frac{\chi_0}{1+(q/\kappa)^2} \qquad \text{with} \quad \kappa = \kappa_0\sqrt{\frac{T-T_{\rm c}}{T_{\rm c}}}. \tag{21}$$

$\xi = 1/\kappa$ is the magnetic correlation length. Amazingly, χ_0 is still given by the Curie–Weiss law. The magnetization arising from the conduction electrons can be viewed as a stochastic variable with a variance $\langle(\delta\mathcal{M})^2\rangle$. From the fluctuation–dissipation theorem, $\langle(\delta\mathcal{M})^2\rangle$ obeys the sum rule

$$\langle(\delta\mathcal{M})^2\rangle = \frac{3\,k_{\rm B}T}{2\pi^2\mu_0}\int_0^{q_u}\chi_{\mathbf{Q}_0}(\mathbf{q})q^2\,{\rm d}q. \tag{22}$$

It is assumed that the energy of the magnetic fluctuations is smaller than the thermal energy. We have introduced the upper cut-off q_u. Obviously, q_u cannot be greater than the radius

of the Brillouin zone. Interestingly, the previous formula gives a relation between the magnetization, and therefore the magnetic moment per unit volume $v_0\sqrt{\langle(\delta\mathcal{M})^2\rangle}$, and the wavevector-dependent static susceptibility. v_0 is the volume per magnetic atom.

The linewidth of the paramagnetic fluctuations depends on their nature. For ferromagnetic fluctuations

$$\hbar\Gamma_{\mathbf{Q}_0}(q) = \mathcal{F}q\left[\chi_{\mathbf{Q}_0}(q)\right]^{-1}, \tag{23}$$

where $\mathcal{F}$ is a constant which sets the scale of the linewidth. Using the Ornstein–Zernike form,

$$\hbar\Gamma_{\mathbf{Q}_0}(q) = \frac{\mathcal{F}}{\chi_0}q\left[1+\left(\frac{q}{\kappa}\right)^2\right]. \tag{24}$$

This linear decay at small q, known as the Landau damping, assumes a ballistic motion of the quasi-particles at the Fermi level. It may break down in the presence of strong disorder. The spin–orbit interaction is assumed to be negligible. Otherwise the total magnetization would not be conserved and the expression we give for the linewidth would not be justified. For an antiferromagnet

$$\hbar\Gamma_{\mathbf{Q}_0}(q) = \mathcal{A}(q^2+\kappa^2). \tag{25}$$

Because the staggered magnetization is not a conserved quantity, $\Gamma_{\mathbf{Q}_0}(q)$ does not vanish for antiferromagnetic fluctuations at small wavevectors. The scaling constants $\mathcal{F}$ and $\mathcal{A}$ can be computed from the band structure near the Fermi surface.

From (1), we generally expect for isotropic long-wavelength fluctuations

$$\lambda_Z \propto k_B T\int_{q_\ell}^{q_u}\frac{\chi_{\mathbf{Q}_0}(q)}{\Gamma_{\mathbf{Q}_0}(q)}q^2\,\mathrm{d}q. \tag{26}$$

We have introduced the lower cut-off q_ℓ. This will be justified hereafter.

With this material at hand, it can be shown for a ferromagnet that $\lambda_Z \propto T/(T-T_C)$ and $\lambda_Z \propto T/(T-T_N)^{1/2}$ for an antiferromagnet, both valid in the limit $\kappa \ll q_u$ (Moriya and Ueda 1974).

In contrast to compounds with localized fluctuations, magnetic fluctuations are known to exist in the ordered state of weak itinerant metals, and so may contribute to the spin–lattice relaxation. The perpendicular (to the easy axis taken as the Z axis) response is much larger than the parallel one, resembling the result for the Heisenberg model where the tilting of the localized spins, which induces a perpendicular spin component, is the driving mechanism for spin dynamics. Therefore, to a good approximation,

$$\lambda_Z = \frac{\mathcal{D}}{2}\frac{1}{V}\int\left[\mathcal{A}^{XX}(\mathbf{q})+\mathcal{A}^{YY}(\mathbf{q})\right]\Lambda^{\perp}(q,\omega_\mu)\frac{\mathrm{d}^3\mathbf{q}}{(2\pi)^3}. \tag{27}$$

In the ordered state of a magnet (see e.g. Chaikin and Lubensky 1995)

$$\chi^{\perp}_{\mathbf{Q}_0}(q) = \frac{\chi_{\mathbf{Q}_0}}{\left(\frac{q}{\kappa}\right)^2} = \frac{\chi_{\mathbf{Q}_0}\kappa^2}{q^2}. \tag{28}$$

With this expression for the susceptibility, (24) and (27) and the susceptibility dissipation theorem, we get $\lambda_Z \propto T/{q_\ell}^2$ for a ferromagnet far below the Curie temperature. We have assumed $q_\ell \ll q_u$. If we had taken $q_\ell = 0$, λ_Z would be infinite. This is clearly unphysical and justifies a finite value for q_ℓ. How do we chose this cut-off? We notice that a given energy band near the Fermi energy may split under the action of the spontaneous field into two bands. A simple choice for q_ℓ is the minimum momentum for a fluctuation to occur taken as $q_\ell = k_{F\uparrow} - k_{F\downarrow}$, where $k_{F\uparrow}$ ($k_{F\downarrow}$) is the momentum of the majority (minority) spin electrons near the Fermi level (Lonzarich and Taillefer 1985). Assuming this momentum difference to

be proportional to the spontaneous magnetization, $\mathcal{M}_0$, we deduce that $\lambda_Z \propto T/\mathcal{M}_0^2$. This is the thermal behaviour which was predicted by Moriya and Ueda in 1974. Hence, we do not expect λ_Z to be proportional to the temperature at relatively high temperature because of the thermal dependence of $\mathcal{M}_0$. The same model, but applied to an antiferromagnet, predicts $\lambda_Z \propto T/\mathcal{M}_{\mathbf{Q}_0}$ where $\mathcal{M}_{\mathbf{Q}_0}$ is now the staggered magnetization (Moriya and Ueda 1974). It has been assumed that $q_\ell \propto \mathcal{M}_{\mathbf{Q}_0}$.

2.5. Critical spin fluctuations

When a system is at or close to a critical point, anomalies occur in a wide variety of static and dynamical properties, most commonly discussed within the theory of critical phenomena. This theory concerns continuous phase transitions and therefore excludes first-order phase transitions characterized, for example, by magnetic hysteresis or latent heat at the ordering point.

A central concept of the theory of critical phenomena is the universality class. Since the study of critical phenomena concerns the behaviour of a system whose correlation length is very large compared with interatomic spacings, the many details of the microscopic Hamiltonian is unimportant. It follows that systems near a critical point can be divided into broad groups known as universality classes, such that all members of a given class have identical critical properties. It is found experimentally that static and dynamic physical properties usually follow simple power laws near critical points. A critical exponent is associated with each law. A universality class is characterized by a set of critical exponents (see e.g. Collins 1989).

Here, we analyse the behaviour of λ_Z near a second-order phase transition. That quantity probes the dynamics. The universality concept for dynamical properties is valid if the universality classes defined for static properties are expanded to specify conservation laws (see e.g. Halperin and Hohenberg 1977). For example, this means that two classes are needed for the description of the physics of the Heisenberg model in three dimensions. The ferromagnets, for which the order parameter is a conserved quantity (it is the uniform magnetization) form one class. In the other class are found all the collinear antiferromagnets for which the order parameter (the staggered magnetization) is not conserved. To account for the critical dynamics, an exponent usually denoted z is introduced. The critical exponents are listed, for example, by Hohenemser *et al* (1989).

As the critical temperature is approached, some fluctuation modes slow down, and the response time tends to infinity at the critical point. Because of the slowing down process, λ_Z is usually expected to display a maximum at the ordering point. The analysis of its behaviour in the vicinity of T_c provides information on the universality class of the compound under study.

We shall first focus on the critical dynamics of ferromagnets; see Yaouanc *et al* (1993) and the review of Frey and Schwabl published in 1994. The Hamiltonian of the system is the sum of the Heisenberg and dipole contributions. Although the dipole interaction between the magnetic ions is weak relative to the Heisenberg interaction, it gains importance near T_C due to its long-range nature. This is so because only the long wavelengths are of importance in the critical regime. Mathematically, it means that it is sufficient to keep the terms in the system Hamiltonian up to second order in wavevectors, that is, for cubic systems with lattice parameter a,

$$\mathcal{H}_{\rm sys} \simeq v_0 \int \sum_{\alpha,\beta} \left\{ \left[-\mathcal{J}_0 + \mathcal{J}_1 q^2 a^2 \right] P_{\rm T_1}^{\alpha\beta}(\mathbf{q}) + \left[-\mathcal{J}_0 + \mathcal{J}_1 q^2 a^2 + \mathcal{J}_1 q_{\rm D}^2 a^2 \right] P_{\rm L}^{\alpha\beta}(\mathbf{q}) \right\} \times J^\alpha(\mathbf{q}) J^\beta(-\mathbf{q}) \frac{{\rm d}^3\mathbf{q}}{(2\pi)^3}. \tag{29}$$

$\mathcal{J}_0$ and $\mathcal{J}_1$ are exchange parameters and $P_{\rm T_1}^{\alpha\beta}(\mathbf{q}) = \delta^{\alpha\beta} - P_{\rm L}^{\alpha\beta}(\mathbf{q})$. The ratio of the dipole to

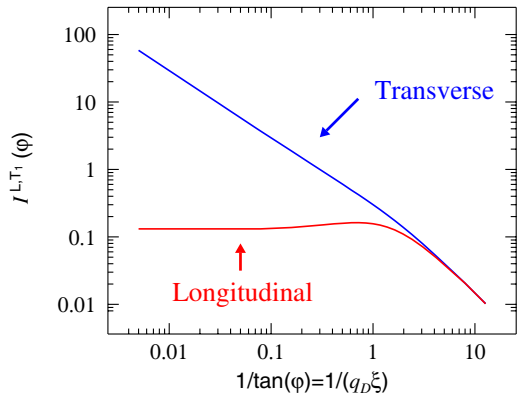

Figure 1. The two functions $I^{\mathrm{L},\mathrm{T}_1}(\varphi)$ versus $1/\tan(\varphi) = 1/(q_\mathrm{D}\xi)$. The functions $I^{\mathrm{L}}(\varphi)$ and $I^{\mathrm{T}_1}(\varphi)$ account, respectively, for the contributions of the longitudinal and transverse (to the wavevector $\mathbf{q}$) fluctuations to the muon spin–lattice relaxation. q_D is the dipole wavevector and ξ the correlation length. Only ξ depends on the temperature. The slope of the two functions for $q_\mathrm{D}\xi \ll 1$ is 3/2. The slope of $I^{\mathrm{T}_1}(\varphi)$ in the low-temperature regime ($q_\mathrm{D}\xi \gg 1$) is unity. From Frey and Schwabl (1994).

exchange interaction is characterized by the dimensionless parameter

$$(q_\mathrm{D}a)^2 = \frac{\mu_0}{v_0}\frac{(g\mu_\mathrm{B})^2}{2\mathcal{J}_1}. \tag{30}$$

q_D is the dipole wavevector. The structure of (29) is remarkable. If the strength of the dipole interaction is neglected, i.e. $q_\mathrm{D} = 0$, the terms in the curly bracket sum up to $(-\mathcal{J}_0+\mathcal{J}_1\mathbf{q}^2a^2)\delta^{\alpha\beta}$. We recover the usual long-wavelength limit of the Heisenberg Hamiltonian with its single length scale ξ such that $\xi^2 \propto \mathcal{J}_1$. The dipole interaction introduces a second length scale proportional to $1/q_\mathrm{D}$ and the Hamiltonian is no longer scalar in $\mathbf{q}$ space. Since the Hamiltonian is a weighted sum of $P^{\alpha\beta}_{\mathrm{T}_1}(\mathbf{q})$ and $P^{\alpha\beta}_{\mathrm{L}}(\mathbf{q})$ for each fluctuation mode, we expect λ_Z to be given also as a weighted sum of two terms:

$$\lambda_Z = \mathcal{W}\left[a_{\mathrm{T}_1}I^{\mathrm{T}_1}(\varphi) + a_\mathrm{L}I^{\mathrm{L}}(\varphi)\right]. \tag{31}$$

φ is a measure for the temperature through the relation $\tan\varphi = q_\mathrm{D}\xi(T)$. The weighting factors a_{T_1} and a_L depend only on the characteristics of the dipole and hyperfine fields at the muon site. In practice a_{T_1} is found to be much smaller than a_L, resulting in a spin dynamics in the vicinity of T_C driven mainly by the longitudinal fluctuation modes as first noticed by Yushankhai in 1989. The two functions $I^{\mathrm{L},\mathrm{T}_1}(\varphi)$ are displayed in figure 1. While $I^{\mathrm{T}_1}(\varphi)$ exhibits a strong temperature dependence in the whole temperature range of the critical region, $I^{\mathrm{L}}(\varphi)$ is temperature independent for $q_\mathrm{D}\xi \gg 1$. A numerical investigation of the region in $\mathbf{q}$ space contributing to λ_Z shows that it is mostly sensitive to fluctuation modes near the zone centre in a region of typical size of a few units of q_D (Dalmas de Réotier *et al* 1994). For example, in the case of metallic nickel at a temperature such that $T - T_\mathrm{C} = 0.056$ K, 90% of λ_Z arises from fluctuation modes with wavevectors smaller than 0.02 Å^{-1}.

We have discussed in some detail the critical dynamics of cubic ferromagnets because quantitative predictions are available. We now briefly discuss the case of antiferromagnets. Since only one length scale is at play, the following power law is derived:

$$\lambda_Z \propto |\delta|^{-\varpi} \qquad \text{with} \quad \varpi = \nu\,(z + 2 - d - \eta)\,. \tag{32}$$

ν, z and η are critical exponents; see e.g. Hohenemser *et al* (1989). d is the dimensionality of the system, i.e. $d = 3$ in three dimensions.

3. Excitations and fluctuations: experimental examples

The intermetallic compound $GdNi_5$ crystallizes in the hexagonal $CaCu_5$ crystal structure and exhibits a ferromagnetic phase transition at $T_C \simeq 32$ K characterized by a small dipole anisotropy field as determined by magnetization measurements: $B_{ani}\,(T = 5\ \text{K}) \simeq 0.21$ T. This compound is a good model for a Heisenberg ferromagnet. The Ni magnetism is not believed to play a role for the spin dynamics. Below about two thirds of T_C, the predicted $\lambda_Z \propto T^2 \ln T$ law is observed with a magnon stiffness constant D_{FM} in the proper range (Dalmas de Réotier and Yaouanc 1997). The expected saturation of λ_Z as T_C is approached is also found experimentally (Yaouanc *et al* 1996). We recall that this saturation is the signature of longitudinal fluctuations at play. As a matter of fact, their effect on the spin dynamics has only been seen clearly by μSR experiments (Frey and Schwabl 1994). From ∼45 K up to the highest temperature where λ_Z has been measured (340 K), λ_Z is found to be temperature independent. Therefore, the density of electronic states at the Fermi level is relatively small. This is in agreement with the conclusions derived from the analysis of μSR data taken for the RNi_5 series where R is a rare-earth element (Mulders *et al* 2003). Therefore, the spin dynamics of isotropic localized magnetic moments which order in a ferromagnetic manner is reasonably well understood in the whole temperature range. A detailed discussion has already been given (Dalmas de Réotier and Yaouanc 1997).

Referring to the general idea that critical dynamics does not depend on the details of the system, but only on its symmetry and the conservation law it obeys, we would expect to observe a similar critical dynamics for localized and itinerant magnetic systems of the same universality class. To test that idea and in relation to the work on $GdNi_5$, a key compound is MnSi because it is a weak isotropic magnet. It crystallizes in a cubic crystal structure ($B20$ lattice type) and orders magnetically in a long-range helical structure with $T_c = 29.5$ K. That compound has been the subject of many studies, including μSR works. However, the expected saturation of λ_Z near $T_c = 29.5$ K has not been reported. In fact, the reason is that the critical region has never been really probed. We have recently detected the saturation within the expected temperature range. A longitudinal field of 5 mT was applied to quench the depolarization from the ^{55}Mn nuclear magnetic moments. The measured behaviour of λ_Z near T_c is therefore not in agreement with the theory sketched in section 2.4. Even more disturbing, our data indicate that the available theory is not able to explain the results of the measurements deep in the paramagnetic region. Below about two thirds of T_C we observe $\lambda_Z \propto T$ instead of the predicted $\lambda_Z \propto T/\mathcal{M}_{\mathbf{Q}_0}$; see figure 2. This is tentatively interpreted assuming that the cut-off q_ℓ is given by a wavevector q_D which is temperature independent. A detailed report of these measurements is in preparation.

In conclusion, the muon spin–lattice relaxation rate in the critical regime of ferromagnets is driven by the dipole interaction. This is probably independent of the nature of magnetism, localized or itinerant. On the other hand, the mechanism of the relaxation in the ferromagnetic state is qualitatively different: it arises from magnons in a localized ferromagnet such as $GdNi_5$ and from magnetic fluctuations in the weak itinerant magnet MnSi.

We now focus our attention on the superconducting ferromagnet UGe_2. It crystallizes in the orthorhombic $ZrGa_2$ crystal structure and has a Curie temperature $T_C \simeq 52$ K. Magnetic measurements indicate a very strong magnetocrystalline anisotropy with an easy magnetization axis along the **a** axis. The discovery of superconductivity below 1 K within a limited pressure range provides an unanticipated example of coexistence of superconductivity and strong ferromagnetism (Saxena *et al* 2000). The electronic pairing mechanism needed for superconductivity is believed to be magnetic in origin. However, it is amazing that ferromagnetically ordered uranium magnetic moments with such a large magnitude ($m_U \simeq$

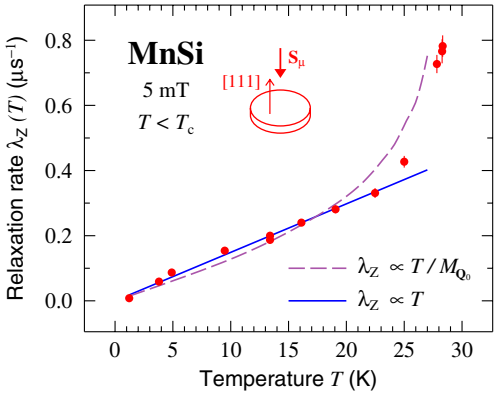

Figure 2. Spin–lattice relaxation rate λ_Z versus temperature measured in the ordered phase of MnSi. A longitudinal field of 5 mT was applied along the [111] crystal axis. The solid line is the result of a linear fit with a slope of 14.9 (1) ms^{-1} K^{-1}. The SCR prediction for the antiferromagnetic case is also shown: $\lambda_Z \propto T/M_{\mathbf{Q}_0}$. The two fits are done for $T \leqslant 22.5$ K and result in chi-squares of 1.05 and 6.75, respectively. The SCR curve has been computed using $M_{\mathbf{Q}_0}$ obtained from neutron scattering (Fåk 2004 private communication). The misfit for the ferromagnetic SCR model is obviously expected to be even worse than for the antiferromagnetic case. That the linear fit breaks down at 25 K and above is not surprising since we are entering the temperature region where the critical spin fluctuations should drive the muon spin relaxation. From Yaouanc *et al*, in preparation.

1.4 μ_B at ambient pressure as deduced from magnetization measurements) are directly at play. Since the pairing must involve the conduction electrons, it is important to characterize their magnetic properties. As the muons localize in interstitial sites, they have the potentiality to yield information on these electrons.

The measurements have been performed at ambient pressure on two single-crystal samples. They differ by the orientation, either parallel or perpendicular, of the **a** axis relative to the initial muon beam polarization. As expected, all the μSR spectra are well described by an exponential relaxation function characterized by λ_Z. A summary of $\lambda_Z(T)$ for the two samples is presented in figure 3. Of most interest is $\lambda_Z(T)$ in the vicinity of the Curie temperature. Qualitatively, it is quite similar to the behaviour found for $GdNi_5$ for which the spin dynamics stems only from the Gd^{3+} magnetic moments (Yaouanc *et al* 1996). The observed saturation of λ_Z as T_C is approached arises from the dipole interaction demagnetization effect on the spin dynamics as already explained.

However, measurements under a longitudinal field show the relaxation to be suppressed by a field as small as $B_{ext} = 2$ mT. This is clearly unexpected. Yaouanc *et al* (2002) use this feature to infer the size of the magnetic moments involved using the fluctuation–dissipation theorem; see section 2.4. Here, we first note that this field corresponds to a Zeeman splitting $\hbar\gamma_\mu B_{ext}$. As usual in NMR, from this splitting we estimate $\tau_c = 1/(\gamma_\mu B_{ext})$. We get τ_c in the microsecond range. This is too long to be relevant to the dynamics of the m_U magnetic moments. Since we have determined a value for τ_c, we can estimate the size of the magnetic moments m_0 at the origin of the relaxation from the level of the relaxation. This can be done recalling from the relaxation theory in the motional narrowing limit that $\lambda_Z = 2\Delta^2\tau_c$ where $\Delta \simeq (\mu_0/4\pi)(m_0^2/r^6)$ where r is the distance between the muon and the nearest neighbour atoms. We get $m_0 \simeq 0.02\,\mu_B$ at T_C. This is really small and cannot be due to the 5f electrons contributing to m_U. The absence of effect of m_U on the measured relaxation is surprising. It simply means that the relaxation associated with the m_U moments is too small to be measured.

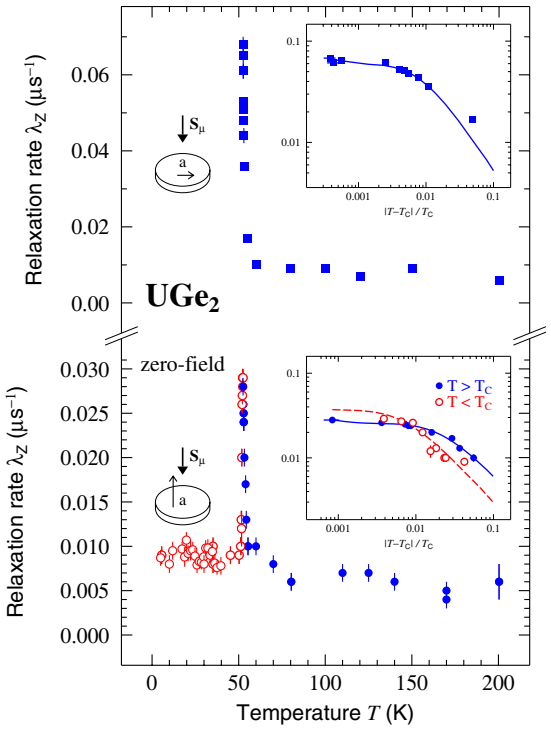

Figure 3. Temperature dependence of λ_Z measured for UGe_2 in zero field for $\mathbf{S}_\mu \perp \mathbf{a}$ and $\mathbf{S}_\mu \parallel \mathbf{a}$ in the upper and lower panels, respectively. $\mathbf{S}_\mu$ stands for the direction of the initial muon beam polarization. The insets display $\lambda_Z(T)$ near T_C. The solid and dashed curves are the results of fits for a dipolar Heisenberg ferromagnet. Since $\mathbf{B}_{loc} \parallel \mathbf{a}$, we cannot observe a spin–lattice relaxation process for $\mathbf{S}_\mu \perp \mathbf{a}$ in the ordered state. The point for $\mathbf{S}_\mu \perp \mathbf{a}$ at $(T - T_C)/T_C = 0.05$ does not fit the critical description, pointing out that it was recorded outside the critical region for that geometrical configuration. The behaviour of $\lambda_Z(T)$ near T_C is typical for a ferromagnet in its critical regime. From Yaouanc *et al* (2002).

m_U measured for UGe_2 and the cubic metallic ferromagnet US (see for example Kernavanois *et al* 2001a) are about equal. US is believed to be a system for which the itinerant character of the 5f electrons is pronounced. Interestingly, from the magnetic form factor measured by neutron diffraction and magnetization measurements, it is inferred for US that the diffuse component of the uranium magnetic moment, i.e. the non-localized part, is 0.15 (4) μ_B. The same type of work for UGe_2 gives for this moment 0.04 (3) μ_B (Kernavanois *et al* 2001b, Kuwahara *et al* 2002). The detailed analysis of the μSR data shows that m_0 arises from weak itinerant long-range magnetic correlations (Yaouanc *et al* 2002). This is in agreement with the size of the diffuse component which is usually attributed to the itinerant electrons. m_0 is certainly negligible relative to m_U for UGe_2. In addition, m_0 is far larger for US than for UGe_2. These facts strongly suggest a large fraction of the 5f electron density to be localized in UGe_2 at ambient pressure. This conclusion is supported by the result of a positron annihilation experiment performed in the paramagnetic phase (Biasini and Troc 2003). Analysing the shape of the x-ray magnetic circular dichroism (XMCD) spectra recorded at $M_{4,5}$ for a number of uranium heavy-fermion compounds, it seems possible to infer information on the nature of the 5f electron density (Yaresko *et al* 2003). Since XMCD spectra have been recorded on UGe_2 at 12 K (Dalmas de Réotier and Yaouanc 2002), their theoretical

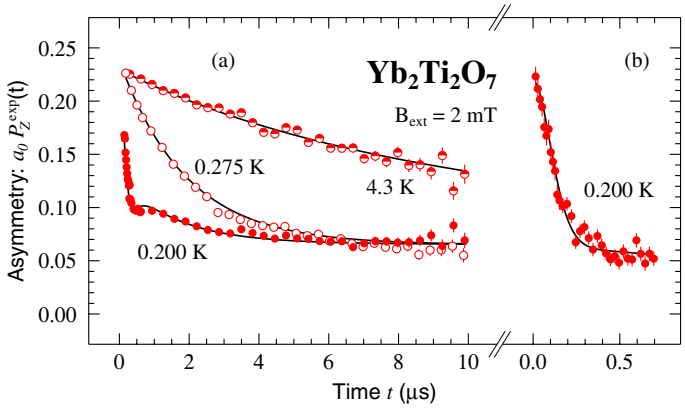

Figure 4. (a) μSR spectra recorded at ISIS for $Yb_2Ti_2O_7$ in a longitudinal field of 2 mT. A marked change occurs on crossing the temperature ($\sim$0.24 K) of the specific heat λ transition. (b) Short-time part of the PSI data at 0.200 K, confirming the absence of short-time oscillations. The slight difference visible between the ISIS and PSI data at 0.200 K is linked with the first-order nature of the transition and the different thermal and magnetic field histories of the two experiments. The solid curves are the result of fits. Adapted from Hodges *et al* (2002).

analysis may provide additional information on the nature of the 5f electron density in this compound.

Since the vast majority of the 5f electron density is localized, the interaction between these electrons located on the uranium atomic sites is probably occurring via the polarized conduction electrons. The measurements indicate the moments of these latter electrons to be approximately 0.02 μ_B. The conventional indirect Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction is therefore at play in UGe_2. In this picture the 5f electrons do not contribute directly to superconductivity. It has been proposed that the superconducting pairing mechanism involves the indirect interaction (Biasini and Troc 2003).

Geometrically derived magnetic frustration in a compound arises when the spatial arrangement of the spins is such that it prevents the simultaneous minimization of all interaction energies; see the review of Ramirez published in 2001. Most of the experimental studies focus nowadays on pyrochlore and gallium garnet compounds, $R_2T_2O_7$ and $R_3Ga_5O_{12}$, respectively. R denotes again a rare-earth atom and T a transition element. The R ions are arranged on a motif of corner sharing tetrahedra for the pyrochlore structure. In the garnet case, the R atoms form two interpenetrating, non-coplanar, corner sharing triangular sublattices.

The classical signature of a magnetic phase transition to a long-range order is a sharp anomaly in the magnetic specific heat at a temperature which corresponds to the phase transition temperature. Such a sharp anomaly is observed for $Yb_2Ti_2O_7$ at $T_\lambda = 0.24$ K. Therefore, it comes as a surprise to observe by μSR that the Yb^{3+} magnetic moments display only dynamical short-range correlations below T_λ (Hodges *et al* 2002). Some spectra are shown in figure 4. Whereas, as expected, exponential relaxation spectra are detected above T_λ, below that temperature spectra typical for a disordered system are found. If the Yb^{3+} magnetic moments were ordering in a coherent fashion at low temperature, we would have observed an oscillation of the spectrum. The lack of magnetic ordering below T_λ has been confirmed by neutron diffraction measurements. Only short-range magnetic correlations are found with a correlation length of $\sim$1.5 nm.

From the analysis of the μSR spectra, we have estimated the correlation time of the Yb^{3+} magnetic moments (Yaouanc *et al* 2003). This can also be done from ^{170}Yb Mössbauer

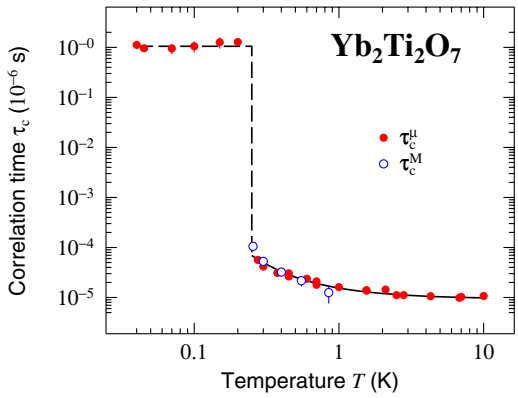

Figure 5. Estimate of the Yb^{3+} fluctuation times in $Yb_2Ti_2O_7$ as obtained from ^{170}Yb Mössbauer (τ_c^M) and μSR (τ_c^μ) measurements. The first-order change in the fluctuation times takes place at the specific heat λ transition. Above ∼0.24 K, the fluctuation times deduced from μSR and Mössbauer spectroscopy match for a Yb^{3+}-muon spin suitable coupling constant. Below ∼0.24 K, the fluctuation time is independent of temperature and has dropped below the lowest value which is measurable with the Mössbauer method (dashed line). The solid curve follows a thermal excitation law. Adapted from Yaouanc *et al* (2003).

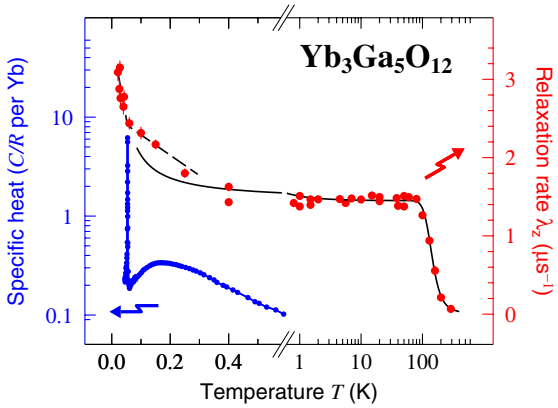

Figure 6. Zero-field muon spin–lattice relaxation rate, λ_Z, measured for $Yb_3Ga_5O_{12}$. The solid curve is the result of a fit. An Orbach process is at play above ∼100 K. Between 1 and 100 K, λ_Z is temperature independent, reflecting the Yb self-correlation in the high-temperature limit. We also display the prediction for the pair correlation in the small-$1/T$ limit for temperatures between 0.1 and 1 K. The two straight dashed lines for $T \leqslant 0.3$ K down to 21 mK are guides to the eyes. The specific heat (from Filippi *et al* 1980) is also reproduced for convenience. Note the change of temperature scale near 1 K. Adapted from Dalmas de Réotier *et al* (2003).

absorption spectra if the correlation time is short enough. The results are presented in figure 5. This figure shows that the specific heat anomaly corresponds to a sharp change (first order since hysteresis is observed) in the dynamics of the Yb^{3+} magnetic moments (about four orders of magnitude) instead of a long-range ordering of these moments. Note that their correlation time is temperature independent below T_λ.

We have recently discovered that $Yb_2Ti_2O_7$ is not a unique case: the garnet compound $Yb_3Ga_5O_{12}$ also displays a sharp specific anomaly at low temperature which does not correspond to a magnetic ordering (Dalmas de Réotier *et al* 2003); see figure 6. In this case it

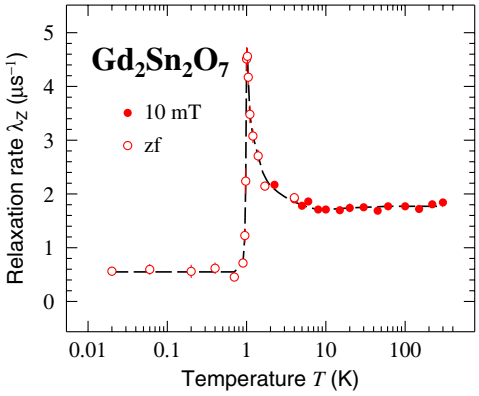

Figure 7. Spin–lattice relaxation rate λ_Z versus temperature for $Gd_2Sn_2O_7$ measured for two values of B_{ext}: 0 and 10 mT. The line is a guide to the eyes.

is the signature of a pronounced building up of dynamical magnetic pair correlations. Besides the sharp anomaly, a broad hump is in fact observed at higher temperature in the specific heat of many geometrically frustrated compounds. Our studies suggest that to observe a conventional long-range ordering below T_λ a sufficiently large magnetic entropy has to be released at the transition.

The physics of frustrated magnets is full of surprises. We have recently discovered that the dynamics of rare-earth moments persists even when they order in a long-range fashion. This is illustrated in figure 7 for the antiferromagnet $Gd_2Sn_2O_7$. We measure an appreciable spin dynamics since the muon spin–lattice relaxation rate is large, even at 20 mK, i.e. far below $T_N \simeq 1$ K. This result is supported by Mössbauer spectroscopy results (Bertin *et al* 2002): the correlated Gd^{3+} moments retain a dynamic character even at extremely low temperature.

The independence of the dynamics on the temperature is also present for $Yb_2Ti_2O_7$ as already noticed. In fact, it seems to be a generic property of geometrically frustrated magnetic materials. It was first reported by Uemura *et al* and collaborators in 1994. An analysis following the material given in section 2.1 shows that the relaxation is induced by a Raman process. Since λ_Z is finite and temperature independent at low temperature, we infer that the dispersion relation of the excitations is characterized by a gap which is proportional to temperature. We believe it is the first time that such a feature for a gap has been recognized experimentally.

As mentioned in the previous section, the magnetic moments which contribute to the relaxation of the muon spin can be relaxed themselves by the phonon or conduction electron density. The drop of λ_Z found for $Yb_3Ga_5O_{12}$ above 100 K, see figure 6, is attributed to an Orbach process in which a phonon induces a transition between the ground state doublet and the CEF excited states located at an energy corresponding to a temperature of about 850 K. Because of the energy conservation requirement, a second phonon is involved. An example of a rare-earth Korringa relaxation has been presented for $Er_5Ir_4Si_{10}$ which displays a charge density wave order at low temperature (Galli *et al* 2002). λ_Z is affected by the formation of the wave via the reduction of the density of states at the Fermi level, induced by the phase transition.

The cleanest signature of antiferromagnetic short-range pair correlations has been found recently for $GdCrO_4$ which orders magnetically at $\sim$22 K (Jimenez *et al* 2004). As predicted by equation (19), as this oxide is cooled from room temperature down to about 50 K, λ_Z decreases monotonically.

4. Flux-line lattices

Here, we focus on the description of the field response of a superconductor in its mixed phase, i.e. for $B_{c1} < B_{ext} < B_{c2}$ where B_{c1} and B_{c2} denote the lower and the upper critical fields, respectively. In fact, we restrict ourselves even more. We shall describe the response for the so-called Bragg-glass phase for which Bragg reflections are observed by small-angle neutron scattering (Giamarchi and Le Doussal 1995). The flux-line lattice (FLL) which diffracts the neutron beam will be assumed to be made of stiff flux tubes running along the Z axis, i.e. the direction of $\mathbf{B}_{ext}$. Hence we neglect the disorder along the Z direction. On the other hand, we account for the in-plane disorder. Therefore, we are dealing with a two-dimensional system for which the induction has only a component along the Z direction.

In the case of interest, the width of the field distribution characterizing the induction field is much smaller than its mean $\langle B \rangle \equiv \langle B^Z(\mathbf{r}) \rangle$. $\mathbf{r}$ is a two-dimensional vector running in a plane perpendicular to the flux tubes. With this condition the transverse-field μSR technique measures the cosine-Fourier transform of the distribution of the component of the induction field along the Z axis. Because the vortex lattice parameter is much larger than the crystallographic lattice parameter, the muons probe uniformly the induction field and thus give access to the distribution of $B^Z(\mathbf{r})$ denoted as $D(B^Z)$.

The simplest description of the FLL follows from the inhomogeneous London equation for the induction. However, it assumes the core radius of each vortex to be negligible. This assumption is at the origin, for example, of the bad description of the high-induction tail of the field distribution. In addition, of special concern to us, it does not lead to an explanation for the observed intrinsic B_{ext} dependence of the amplitude of the Fourier components of the induction. There is no general theory of $B^Z(\mathbf{r})$ valid at arbitrary temperature. The only practical theory to model the core is due to Ginzburg and Landau (see e.g. Kittel (1996) for a simple introduction). It is based on a phenomenological expansion of the free energy in terms of the superconducting order parameter and its gradient. Two length scales are introduced: the London peneration depth, λ_L, and the Ginzburg–Landau coherence length, ξ_{GL}. Minimization of the free energy provides two coupled differential equations: one for the order parameter and one for the currents, that is, for the diamagnetic response of a superconductor. These two Ginzburg–Landau (GL) equations can be written in terms of only a single parameter, the so-called Ginzburg–Landau parameter $\kappa_{GL} \equiv \lambda_L/\xi_{GL}$. The GL equations cannot be solved analytically, even in the limit $\kappa_{GL} \to \infty$, which would be sufficient here. A convenient approximate solution has been given for superconductors with isotropic planar symmetry (Yaouanc *et al* 1997). Recently it has been generalized to superconductors which are anisotropic in the plane perpendicular to B_{ext} (Dalmas de Réotier and Yaouanc, in preparation).

$D(B^Z)$ should exhibit van Hove singularities at the maximum, minimum and saddle points of the field distribution. Although usually observed experimentally, these singularities are always smeared. This arises for instance from the dipole field of the nuclear magnetic moments contained in the specimen. Also, because of the finite lifetime of the muon, the distribution from the measured $P_X(t)$ is broadened in a similar manner as caused by apodization of the data. In addition, in a real material the FLL is disordered by the interaction of the flux tubes with inhomogeneities and defects which act as pinning centres. This leads to a smearing of the singularities of $D(B^Z)$ and to an increase of its width for a stiff FLL (Brandt 1991). The disorder is conventionally accounted for by the convolution of the ideal distribution with a Gaussian function. While this method works quite well numerically for the Bragg-glass phase (see e.g. Sonier *et al* 2000), it is not satisfactory because the physics involved is not clear. Here, we are going to sketch the derivation of a formula for the variance of the FLL

field distribution, $(\Delta_{\rm v}^Z)^2$, which accounts microscopically for the pinning-induced disorder. A complete derivation will be presented elsewhere.

$B^Z(\mathbf{r})$ is built up from the contribution of all the flux tubes in the FLL. Neglecting the effect on $B^Z(\mathbf{r})$ of the interaction between the fields from the different flux tubes, we write $B^Z(\mathbf{r})$ as a convolution product:

$$B^Z(\mathbf{r}) = \int b^Z(\mathbf{r}-\mathbf{r}')\varrho(\mathbf{r}')\,\mathrm{d}^2\mathbf{r}'. \tag{33}$$

$b^Z(\mathbf{r})$ is the field at the vector position $\mathbf{r}$ for a flux tube centred at the origin of the coordinates and $\varrho(\mathbf{r})$ the flux density. The depression of the superconducting order parameter due to the overlap of the vortex cores will be approximately described by the $(1-b^4)$ factor (we define $b = \langle B\rangle/B_{\rm c2}$) to be introduced later on. It is convenient to work in Fourier space, with the result

$$B^Z(\mathbf{r}) = \int b^Z(\mathbf{q})\varrho(\mathbf{q})\,\exp(\mathrm{i}\mathbf{q}\cdot\mathbf{r})\,\frac{\mathrm{d}^2\mathbf{q}}{(2\pi)^2}. \tag{34}$$

The flux tubes are not centred at the ideal positions $\{\mathbf{i}\}$ but at the effective positions $\{\mathbf{i}+\tilde{\mathbf{u}}(\mathbf{i})\}$ which deviate from the equilibrium positions by the displacement vectors $\{\tilde{\mathbf{u}}(\mathbf{i})\}$. The density is therefore written as $\varrho(\mathbf{r}) = \sum_{\mathbf{i}}\delta\left[\mathbf{r}-\mathbf{i}-\tilde{\mathbf{u}}(\mathbf{i})\right]$. Although this density is not a periodic function, it can be expanded in terms of Fourier components if the displacement field is smooth (Giamarchi and Le Doussal 1995): $\varrho(\mathbf{r}) \simeq s_{\rm c}^{-1}\sum_{\mathbf{G}}\exp\{\mathrm{i}\mathbf{G}\cdot[\mathbf{r}-\mathbf{u}(\mathbf{r})]\}$. $\mathbf{G}$ is a reciprocal lattice vector, $s_{\rm c}$ the area of the unit cell and $\mathbf{u}(\mathbf{r})$ the displacement of a flux tube as a function of its real position. This decomposition of the delta function is strictly valid in the absence of dislocations. With this expression for the density and defining $\mathcal{B}^Z(\mathbf{q}) = b^Z(\mathbf{q})/s_{\rm c}$,

$$B^Z(\mathbf{r}) = \sum_{\mathbf{G}}\int \mathcal{B}^Z(\mathbf{q})\exp(\mathrm{i}\mathbf{q}\cdot\mathbf{r})\int \exp\left[-\mathrm{i}\,(\mathbf{q}-\mathbf{G})\cdot\mathbf{r}'\right]\exp\left[-\mathrm{i}\mathbf{G}\cdot\mathbf{u}(\mathbf{r}')\right]\,\mathrm{d}^2\mathbf{r}'\frac{\mathrm{d}^2\mathbf{q}}{(2\pi)^2}. \tag{35}$$

To proceed further, we need to remember two points. Only a thermal average of a physical quantity can be measured and, because we are dealing with a disordered system, an ensemble average has to be performed. Denoting the thermal average $\langle\cdots\rangle$ and the disorder average $\overline{\cdots}$, we derive

$$\left(\Delta_{\rm v}^Z\right)^2 = \sum_{\mathbf{G}\neq\mathbf{0}}\int\left|\mathcal{B}^Z(\mathbf{q})\right|^2\int\exp\left[-\mathrm{i}\,(\mathbf{q}-\mathbf{G})\cdot\mathbf{r}\right]\mathcal{C}_{\mathbf{G}}(\mathbf{r})\,\mathrm{d}^2\mathbf{r}\,\frac{\mathrm{d}^2\mathbf{q}}{(2\pi)^2}. \tag{36}$$

We have introduced the translational order correlation function

$$\mathcal{C}_{\mathbf{G}}(\mathbf{r}) = \overline{\langle\exp\left[-\mathrm{i}\mathbf{G}\cdot\mathbf{u}\,(\mathbf{r})\right]\exp\left[\mathrm{i}\mathbf{G}\cdot\mathbf{u}(\mathbf{0})\right]\rangle}. \tag{37}$$

The information on the disorder is embedded in $\mathcal{C}_{\mathbf{G}}(\mathbf{r})$. To get a feeling of its meaning we investigate two asymptotic limits. But first we need to specify $\mathcal{B}^Z(\mathbf{q})$. For definitiveness we shall assume the superconductor to be isotropic. Hence, according to the literature (Yaouanc *et al* 1997),

$$\mathcal{B}^Z(\mathbf{q}) = \frac{\Phi_0}{s_{\rm c}}(1-b^4)\frac{wK_1(w)}{1+\lambda_{\rm L}^2q^2} \qquad \text{with} \quad w^2 = \xi_{\rm v}^2q^2. \tag{38}$$

$K_1(x)$ is a modified Bessel function. We have defined the length $\xi_{\rm v} = \sqrt{2}\xi_{\rm GL}(1+b^4)^{1/2}[1-2b(1-b)^2]^{1/2}$. We denote the flux quantum by Φ_0 ($\Phi_0 = 2.07\times10^{-15}\,\mathrm{T\,m^2}$). $wK_1(w)$ acts as a cut-off factor.

Let us first assume a disorder-free FLL. This means that there is no displacement of the flux tube positions. Referring to the definition of $\mathcal{C}_{\mathbf{G}}(\mathbf{r})$, we conclude that $\mathcal{C}_{\mathbf{G}}(\mathbf{r}) = 1$. Hence $\int \exp[-\mathrm{i}(\mathbf{q}-\mathbf{G})\cdot\mathbf{r}]\mathcal{C}_{\mathbf{G}}(\mathbf{r})\,\mathrm{d}^2\mathbf{r} = (2\pi)^2\delta(\mathbf{q}-\mathbf{G})$ and therefore

$$\left(\Delta_{\mathrm{v}}^{Z}\right)^2 = \sum_{\mathbf{G}\neq\mathbf{0}} \left|B_{\mathbf{G}}^{Z}\right|^2 = 3.711\times 10^{-3}\frac{\Phi_0^2}{\lambda_{\mathrm{L}}^4} f_{\mathrm{v}}^2(b). \tag{39}$$

The function $f_{\mathrm{v}}(b)$ is presented elsewhere (Yaouanc *et al* 1997). $f_{\mathrm{v}}(b=0) = 1$ and it decays monotonically with b. In the other extreme limit, i.e. in the case of extremely strong disorder, $\mathcal{C}_{\mathbf{G}}(\mathbf{r})$ is proportional to a delta function and thus $\sum_{\mathbf{G}\neq\mathbf{0}}\int \exp[-\mathrm{i}(\mathbf{q}-\mathbf{G})\cdot\mathbf{r}]\mathcal{C}_{\mathbf{G}}(\mathbf{r})\,\mathrm{d}^2\mathbf{r} = s_{\mathrm{c}}$. The integration over $\mathbf{q}$ can be done analytically only in the London limit, i.e. $wK_1(w) \to 1$. We obtain

$$\left(\Delta_{\mathrm{v}}^{Z}\right)^2 = \frac{\langle B\rangle\Phi_0}{4\pi\lambda_{\mathrm{L}}^2}. \tag{40}$$

This result was first published by Brandt in 1991.

The functional form of the correlation function $\mathcal{C}_{\mathbf{G}}(\mathbf{r})$ is discussed by Giamarchi and Le Doussal (Giamarchi and Le Doussal 1995). It depends on a single length: the in-plane correlation length of the FLL, R_a^{XY}. Hence, if λ_{L} and ξ_{GL} are already known or have been measured by μSR using the usual procedure (see e.g. Sonier *et al* 2000), our result indicates that R_a^{XY} can be determined from the measurement of the variance of the FLL field distribution.

5. Conclusions

The purpose of this article has been to show that the μSR techniques allow us to study quantitatively magnetic excitations, fluctuations and correlation lengths. The framework to interpret the experimental results uses the linear response theory which has been developed for many years by scientists involved in neutron experiments. This points out the interest for the people of the muon and neutron communities to exchange information and even to work together. This can be done in western Europe since the two existing muon and accelerator-based neutron sources share common facilities. Neutron scattering is theoretically able to fully characterize the magnetic property of a compound because it probes its energetic and spatial characteristics. However, experimental resolutions are such that a magnetic mode with a fluctuation time slower than a nanosecond or with wavevector smaller than 0.01 Å cannot be studied. Such modes can be nicely investigated by μSR. Typical new examples discussed here are the critical magnetic fluctuations of the itinerant electrons in the ferromagnetic superconductor UGe_2 and the density of magnetic excitations in frustrated compounds. In addition, we point out the possibility of estimating the in-plane correlation in the Bragg-glass phase of superconductors.

Scheduled technical developments are going to extend the range of applications of the μSR techniques. We mention first the dedicated low-energy muon spectrometer currently being built at the Paul Scherrer Institute in Switzerland. It will give the opportunity to study multilayers from the subnanometre range to the 100 nm region. Secondly, regarding conventional energy muons, a spectrometer with a tenfold increase in counting rate is envisaged at the muon pulsed facility located near Oxford in England. The relatively large dimensions required for a sample to be run at a muon spectrometer have always been a problem. Here also good news is coming: a sample of 3.74 mg has recently been investigated (Sonier *et al* 2003). Concerning measurements under extreme conditions, μSR is well known for the numerous studies performed at extremely low temperature, down to 13 mK. Currently, external fields up to 8 T can be applied and it is possible to apply a pressure up to 15 kbar and down to 0.35 K.

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