Spin correlations in rare-earth paramagnetic systems; neutron linewidths and $\mu$SR spin-lattice relaxation rates.

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We consider the spin correlation functions for rare-earth compounds in their paramagnetic state with the aim to understand the temperature dependence of the quasi-elastic and crystal-field linewidths and the spin-lattice relaxation rates measured by the muon-spin-relaxation technique. Both the conduction electron and phonon relaxation mechanisms are treated. First the single-ion dynamics is described using an iterative method introduced by P. M. Richards. Then the case of a regular lattice of rare-earth ions is studied using the random-phase approximation (RPA). Applications to simple crystal-field level schemes are given with particular emphasis on the phonon relaxation mechanism. This allows us to investigate the domain of validity of our results. In order to account for data recorded on insulators, a phenomenological modification is suggested.

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The study of the spin dynamics of a regular lattice of rare-earth ions in the paramagnetic phase of an insulator is not a subject of much interest in itself. However, exotic dynamics may have been seen at low temperature in geometrically frustrated magnetic rare-earth compounds, see Ref. [1] for an example and Ref. [2] for a general reference. Therefore we find useful to establish the expected thermal behaviour of the crystal-field linewidth and the positive muon ($\mu$SR) spin-lattice relaxation rate arising from the phonon-induced relaxation.

A theory for the linewidth of crystal-field excitations in metallic rare-earth systems has been given a long time ago\(^3\). The description of the single-ion dynamics was based on the memory function technique. Later on, the same problem was considered by Richards using a more conventional iterative method akin to the Redfield's, the derived system of equations being solved by Laplace transform\(^4\).

Here we examine the contribution of the conduction electrons and phonons to the spin dynamics in the paramagnetic phase of compounds with a rare-earth sublattice. The crystal electric field acting on the magnetic ions will be dealt with. The interaction between the ions will be accounted for.

Two experimental methods are commonly used to investigate spin dynamics in magnetic materials: the neutron scattering and $\mu$SR techniques. While the relationship between the measured intensity and the physical properties of the spin correlation functions is well documented for the former, see for example Refs. [5,6], it is not the case for the latter, at least for a lattice of rare-earth ions for which crystal-field effects are expected to be important. This is done in this paper, building on previous works dealing with the critical spin dynamics and magnon-induced relaxation as probed by $\mu$SR, see Ref. [7] and references therein.

The organization of this paper is as follows. In Sec. [I] we investigate some basic properties of the spin-correlation tensor. In Sec. [II] we write the $\mu$SR spin-lattice relaxation rate, $\lambda_Z$, in terms of spin-correlation functions. In Sec. [III] these functions are described for the case of a single rare-earth ion, following the method of Richards. In the following section, Sec. [IV], we give the expressions for the conduction electrons and phonon correlation functions. The theory develops in the previous sections is applied for simple crystal-field level schemes in Sec. [V]. The interaction between the magnetic ions is taken into account in Sec. [VI]. We start by using the random-phase approximation (RPA). Since this approximation is found to breakdown for a limit of experimental interest, a phenomenological modification is proposed.

I. PROPERTIES OF THE SPIN-CORRELATION TENSOR

In this section, we study some basic properties, mostly resulting from the point symmetry at the rare-earth site, of the symmetrised spin-correlation function $\Lambda_{\alpha\beta}(\omega) = \int_{-\infty}^{\infty} \langle \{J_\alpha(t)J_\beta(t)\} \rangle \exp(i\omega t)dt$, where $2 \langle \{J_\alpha(t)J_\beta(t)\} \rangle = \langle J_\alpha(t)J_\beta(t) \rangle + \langle J_\beta J_\alpha(t) \rangle$. $J_\alpha$ is the $\alpha$ component of the total angular momentum of a magnetic ion in the compound of interest. We explicitly assume $J$ to be a good quantum number. This is true for a rare-earth ion.

Our first task is to examine the structure of $\hat{\Lambda}(\omega)$, the tensor elements of which are the functions $\Lambda_{\alpha\beta}(\omega)$. This tensor is written in the reference frame $\{x,y,z\}$ attached to the crystal lattice. In fact, because the crystal electric field potential is expressed in terms of Stevens operators which are polynomials of $J_+ = J_x + iJ_y$, $J_- = J_x - iJ_y$ and $J_z$, we shall work with the reference frame $\{+,+,-,z\}$. It is only in this representation that the Stevens operators display nicely their symmetry properties.

In this study we shall split the total Hamiltonian of the system under study into two parts. We write $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1$ where $\mathcal{H}_0$ describes the perturbation of the system relative to $\mathcal{H}_0$. In addition, we take $\mathcal{H}_0$ as the sum of two commutating operators, $\mathcal{H}_0 = \mathcal{H}_{\text{CF}} + \mathcal{H}_L$, where the first
term is modelling the crystal-field energy levels of the rare-earth ions and the second the lattice. We restrict ourselves to the analysis of zero-field measurements. We shall assume in this paper $\mathcal{H}_1 = 0$.

Physically the relaxation of a spin correlation function of a single rare-earth ion is driven by the crystal-field transitions induced by the coupling of the crystal field with the lattice. Neglecting this coupling, i.e. assuming $\mathcal{H}_1 = 0$, we determine which spin correlations are compatible with the point symmetry at the rare-earth site. If a correlation vanishes when the coupling is neglected, it cannot be large even if it is taken into account. Hence, we study the symmetry properties of $\langle J_\alpha(t)J_\beta \rangle_0$, where

$$\langle J_\alpha(t)J_\beta \rangle_0 = \text{Tr} \{ \rho_0 \exp(i\mathcal{H}_0t/\hbar) J_\alpha \exp(-i\mathcal{H}_0t/\hbar) J_\beta \}.$$

We have introduced the density operator $\rho_0 = \exp(-\beta \mathcal{H}_0)/Z_0$ where $Z_0$ is the partition function of $\mathcal{H}_0$, $\beta = 1/(k_b T)$ and Tr$\{A\}$ stands for the trace of $A$ over the quantum states of $\mathcal{H}_0$. Since $[\mathcal{H}_L, J_\alpha(t)] = 0$, $[\mathcal{H}_L, J_\beta] = 0$ and thus

$$\langle J_\alpha(t)J_\beta \rangle_0 = \text{Tr} \{ \rho \exp(i\mathcal{H}_{CF}t/\hbar) J_\alpha \exp(-i\mathcal{H}_{CF}t/\hbar) J_\beta \}.$$

$\rho$ is the density operator for the crystal-field levels. We use the simplified notations $\rho$ and $\langle \cdot \cdot \cdot \rangle$ rather than $\rho_{CF}$ and $\langle \cdot \cdot \cdot \rangle_{CF}$ because there is no risk of confusion.

We are going to show that the only tensor elements which may not be zero in most physical cases are $\Lambda_{zz}(\omega)$, $\Lambda_{+}(\omega)$ and $\Lambda_{-}(\omega)$. To prove, for example, that $\Lambda_{zz}(\omega)$ is usually incompatible with the point symmetry at the rare-earth site, we need to show $\langle J_- (t) J_z \rangle = 0$. We have

$$\langle J_- (t) J_z \rangle = \text{Tr} \{ \rho \exp(i\mathcal{H}_{CF}t/\hbar) J_- \exp(-i\mathcal{H}_{CF}t/\hbar) J_z \} = \text{Tr} \{ \rho \exp(i\mathcal{H}_{CF}t/\hbar) \mathcal{I} J_- \exp(-i\mathcal{H}_{CF}t/\hbar) J_z \}$$

We have inserted the unit tensor $\mathcal{I}$ twice. $\mathcal{H}_{CF}$ is invariant under the operations of the point symmetry group at the ion site. Let us assume that one of the symmetry operation of the group is a rotation of angle $\varphi = 2\pi/n$ about the $z$ axis, i.e. the $z$ axis is a $n$-fold symmetry axis. Mathematically this invariance means

$$[\mathcal{H}_{CF}, \mathcal{R}_z(\varphi)] = 0,$$

where $\mathcal{R}_z(\varphi) = \exp(-i\varphi J_z)$ stands for the rotation operator about the $z$ axis of angle $\varphi$; see for example Ref. [8]. Recalling the relation

$$\mathcal{R}_z(\varphi) J_- \mathcal{R}_z^{-1}(\varphi) = \exp(-i\varphi) J_-,$$

and Eq. [2] starting from Eq. [1] we derive

$$\langle J_- (t) J_z \rangle = \exp(-i\varphi) \langle J_- (t) J_z \rangle.$$

We have used $\mathcal{I} = \mathcal{R}_z^{-1}(\varphi) \mathcal{R}_z(\varphi)$. The only solution of this equation with $\varphi = 2\pi/n$ and $n \geq 2$ is $\langle J_- (t) J_z \rangle = 0$. In the same way we find $\langle J_z J_- (t) \rangle = 0$. Our conclusion is that the existence of the symmetrised correlation function $\{ \langle J_- (t) J_z \rangle \}$ is inconsistent with the $z$ axis being two-fold or of higher symmetry. Using the same procedure we find five other elements of $\Lambda(\omega)$ to be zero when $n \geq 3$, because $\{ \langle J_- (t) J_z \rangle \} = 0$ only if $n \geq 3$. However, this tensor is not diagonal in $\{ +, -, z \}$ since the three non vanishing functions are $\Lambda_+ (\omega)$, $\Lambda_+ (\omega)$ and $\Lambda_{zz} (\omega)$.

Before diagonalizing $\Lambda(\omega)$, we first establish some general properties of the symmetrised correlation functions. As $\langle J_\alpha(t)J_\beta \rangle^* = \langle J_\beta(t)J_\alpha \rangle$ and $\langle J_\alpha(t)J_\beta \rangle^* = \langle J_\beta(t)J_\alpha \rangle$, from the definition of $\Lambda_{zz} (\omega)$, $\Lambda_+ (\omega)$ and $\Lambda_{-} (\omega)$ we infer these functions to be real. In our notations $A^\dagger$ is the adjoint of operator $A$. Since in addition $\langle J_\alpha(t)J_\beta \rangle^* = \langle J_\beta(t)J_\alpha \rangle$ and $\langle J_\alpha(t)J_\beta \rangle^* = \langle J_\beta(t)J_\alpha \rangle$, we derive $\Lambda_+ (\omega) = \Lambda_+ (\omega)$. Therefore the present discussion leads to restrict the number of correlation functions to be computed to two: $\Lambda_{zz} (\omega)$ and, e.g., $\Lambda_{-} (\omega)$. $\Lambda(\omega)$ is diagonal in a frame denoted by $\{ +, -, z \}$. We shall need to specify explicitly only the eigenvalues. They are $\Lambda_{zz} (\omega) = \Lambda_{zz} (\omega) = \sqrt{\Lambda_{zz} (\omega)}\Lambda_{zz} (\omega) = -\Lambda_{zz} (\omega)$ and $\Lambda_{zz} (\omega) = \Lambda_{zz} (\omega)$. One easily shows that $\Lambda_+ (\omega)\Lambda_+ (\omega) \geq 0$.

Let us introduce the auxiliary function

$$\Omega_{\alpha\beta}(\omega) = \int_{-\infty}^{\infty} \langle J_\alpha(t) J_\beta \rangle \exp(\text{i}\omega t) \text{dt}.$$

The condition of detailed balance gives the relationship

$$\Omega_{\alpha\beta}(\omega) = \exp(\text{i}\omega t) \Omega_{\alpha\beta}(\omega).$$

This implies

$$\Lambda_{zz} (\omega) = \left[ 1 + \exp(\text{i}\omega t) \right] \Omega_{zz} (\omega)/2,$$

$$\Lambda_{zz} (\omega) = \left[ 1 + \exp(\text{i}\omega t) \right] \Omega_{zz} (\omega)/2.$$ 

Hence, once $\Omega_{zz} (\omega)$ and $\Omega_{zz} (\omega)$ are computed, $\Lambda_{zz} (\omega)$ and $\Lambda_{zz} (\omega)$ can be evaluated.

Note that the inelastic neutron scattered intensity is directly proportional to $\Omega_{\alpha\beta}(\omega)$; see for example Refs. [5, 6]. The so-called scattering function $S_{\alpha\beta}(\omega)$ is within a constant of proportionality equal to $\Omega_{\alpha\beta}(\omega)$.

II. μSR SPIN-LATTICE RELAXATION RATE AND SPIN-CORRELATION FUNCTIONS

The $\mu$SR techniques are based on the detection of the positron produced during the muon decay. Of particular interest is the longitudinal field technique for which the initial muon beam polarization, the external field $B_{ext}$ if any, and the axis of the positron detectors are all parallel to the same axis denoted as $Z$. Therefore that axis is of special importance. We define it by its polar angles $\theta$ and $\varphi$ in the reference frame $\{x, y, z\}$ introduced in the previous section. We note that the $\mu$SR techniques probe only the magnetic field at the muon site and therefore
the magnetic field correlation tensor $\Phi(\omega_{\mu})$ at an energy $\hbar\omega_{\mu}$. $\omega_{\mu}$ is proportional to the local field at the muon site. Since the difference between that field and $B_{\text{ext}}$ is usually very smallootnote{In this context, $B_{\text{ext}}$ is the external magnetic field.}, we write $\omega_{\mu} = \gamma_{\mu}B_{\text{ext}}$ where $\gamma_{\mu}$ is the muon gyromagnetic ratio ($\gamma_{\mu} = 2.8156$ Mrad s$^{-1}$ T$^{-1}$). $\Phi(\omega_{\mu})$ is also defined in $\{+',-',z\}$.

A longitudinal field $\mu$SR experiment consists of measuring the relaxation function which is, to a good approximation, an exponential function in the case of interest here, e.g. for paramagnets with a regular lattice of rare-earth ions, and therefore is characterized by a single parameter, the spin-lattice relaxation rate $\lambda_{Z}$. Implicit is the assumption that the motional narrowing limit applies, i.e. the fluctuations of the rare-earth moments are rapid. $\lambda_{Z}$ is directly related to $\Phi(\omega_{\mu})$:

$$\lambda_{Z} = \frac{\gamma^{2}}{2} \sum_{i,j} L_{ij}(\theta, \varphi) \Phi(\omega_{\mu}),$$

where now $\{i,j\} = \{+',-',z\}$, the matrix elements $L_{ij}(\theta, \varphi)$ can be computed from the material given in Appendix C of Ref.\cite{10} and $\Phi(\omega_{\mu})$ is an element of the $\Phi(\omega_{\mu})$. We have introduced the Fourier transform $\Phi(\omega_{\mu}) = \int_{-\infty}^{\infty} \Phi(\omega_{\mu})(t) \exp(i\omega t)dt$, a definition different from the one used previously\cite{11}. The total angular momenta of the magnetic ions interact via the muon spin through the dipole interaction and, in addition, the hyperfine interaction which accounts for the transferred magnetic field for an insulator and the conduction electrons for a metallic compound. This induces a magnetic field at the muon site with correlation functions such that

$$\Phi(\omega_{\mu}) = \left(\frac{\mu_{0}}{4\pi} \right)^{2} g^{2} \mu_{B}^{2} \frac{1}{v^{2}} \sum_{i,j} G_{\alpha\beta}^{ij} \Lambda_{\alpha:j,i}(\omega).$$

$i$ and $j$ run over the magnetic ions. $\Lambda_{\alpha:j,i}(\omega)$ is a spin-correlation function between sites $i$ and $j$. $G_{\alpha\beta}$ is the coupling tensor labeled by the vector $\mathbf{r}_{i}$ which points to site $i$ and the origin of which is the muon site. Explicitly we have

$$G_{\alpha\beta}^{ij} = v \left(3r_{i}\cdot r_{j}\alpha\beta \right) r_{i}^{3} + H_{ij} \delta^{ij}. \quad (12)$$

$H_{ij}$ is the hyperfine constant for the coupling to the ion $i$. For simplicity we assume all the magnetic ions to be equivalent: they sit on a Bravais lattice. $\mu_{0}$ is the magnetic permeability of free space, $g$ the spectroscopic factor, $\mu_{B}$ the electronic Bohr magneton and $v$ the volume per magnetic ion.

Two approaches are now possible. If the interaction between the total angular momenta of the magnetic ions is negligible, we can restrict ourselves to the behaviour of a single momentum. Otherwise their interactions has to be taken into account.

We first consider the case for which $\Lambda_{\alpha:i,j\beta}(\omega) = \Lambda_{\alpha}(\omega) \delta_{i,j}$. The intersite correlations are therefore negligible and thus

$$\Phi(\omega_{\mu}) = \left(\frac{\mu_{0}}{4\pi} \right)^{2} g^{2} \mu_{B}^{2} \frac{1}{v^{2}} \sum_{\alpha} \sum_{i} G_{\alpha}^{ij} G_{\alpha}^{ij} \Lambda_{\alpha}(\omega),$$

Hence, based on our discussion in Sec.\cite{1} $\lambda_{Z}$ is found to be the weighted sum of two spin-correlation functions for a compound with at least a three-fold symmetry axis: they are $\Lambda_{z}(\omega) = \Lambda_{zz}(\omega)$ and, e.g. $\Lambda_{z}(\omega) = \sqrt{\Lambda_{z}(\omega)\Lambda_{z}(-\omega)}$. Obviously, additional symmetry elements of the point group at the rare-earth site may reduce the number of correlations to be computed to only one.

It is convenient to use the space-Fourier transform of $\Lambda_{\alpha}(q, \omega)$ if the intersite correlations are appreciable. Referring to Ref.\cite{12},

$$\Phi(\omega_{\mu}) = \left(\frac{\mu_{0}}{4\pi} \right)^{2} g^{2} \mu_{B}^{2} \frac{1}{v} \int G^{\gamma\alpha}(q) \Lambda_{\alpha}(q, \omega) \frac{d^{3}q}{(2\pi)^{3}},$$

with the definitions $G^{\gamma\alpha}(q) = G^{\alpha\gamma}(q)G^{\alpha\gamma}(-q)$ and $G^{\alpha\gamma}(q) = \sum_{ij} G^{\alpha\gamma}_{ij} \exp(i\mathbf{q} \cdot \mathbf{r}_{ij})$. $V$ is the volume of the sample, i.e. $V = Nv$ where $N$ is the number of magnetic ions in the sample. The integration is over the Brillouin zone. Again, two spin-correlation functions are sufficient to model $\lambda_{Z}$ for a compound with at least a three-fold symmetry axis. As pointed out above, the number may be reduce to one due to the presence of additional symmetries at the rare-earth site.

III. SINGLE-ION CORRELATION FUNCTIONS $\Omega_{\beta\alpha}(\omega)$ IN THE REFERENCE FRAME $\{+',-',z\}$

In this section we give formulae for $\Omega_{\beta\alpha}(\omega)$ and $\Omega_{+\omega}(\omega)$. The interaction between the rare-earth ions is neglected. The correlation functions are evaluated with the Richards’s method\cite{4}. In fact, it is the Laplace transform of $J_{\alpha}(t)$ which is considered there. P. M. Richards has only presented the result of his work, focusing on the electronic relaxation. Here we sketch the derivation and apply the result for two mechanisms which can drive the crystal-field transitions: the conduction electrons and the strain field.

The interaction representation is used. This requires to introduce the two new operators, $J_{\alpha}^{*}(t)$ and $\mathcal{H}_{\alpha}(t)$ (see for example Ref.\cite{13}), such that

$$J_{\alpha}^{*}(t) = \exp(-i\mathcal{H}_{\alpha}t/h)J_{\alpha}(t)\exp(i\mathcal{H}_{\alpha}t/h),$$

$$\mathcal{H}_{\alpha}(t) = \exp(-i\mathcal{H}_{\alpha}t/h)\mathcal{H}_{\alpha}\exp(i\mathcal{H}_{\alpha}t/h). \quad (15)$$

Note that $J_{\alpha}^{*}(t=0) = J_{\alpha}(t=0)$. Using the Heisenberg evolution equation for $J_{\alpha}(t)$,

$$\frac{dJ_{\alpha}(t)}{dt} - \frac{i}{\hbar}[\mathcal{H}_{\alpha}, J_{\alpha}(t)]$$
\begin{align}
&= \exp\left(i\mathcal{H}_0 t/\hbar\right) \frac{dJ_\alpha^*}{dt} \exp\left(-i\mathcal{H}_0 t/\hbar\right), \quad (16) \\
\text{with} \quad \frac{dJ_\alpha^*}{dt} &= \frac{i}{\hbar}\left[\mathcal{H}_\alpha^*(t), J_\alpha^*(t)\right]. \quad (17)
\end{align}

Solving this last equation for \( J_\alpha^*(t) \),
\begin{equation}
J_\alpha^*(t) = J_\alpha + \frac{i}{\hbar} \int_0^t \left[\mathcal{H}_\alpha^*\left(t'\right), J_\alpha^*\left(t'\right)\right] dt'. \quad (18)
\end{equation}

We are basically interested by the matrix elements of Eq.\,(16) For their evaluation, we introduce the expression for \( J_\alpha^*(t) \) in the commutator of Eq.\,(17) dropping the first term on the right hand side which is time independent. We note the kets of \( \mathcal{H}_{\text{CF}} \) as \( \{m, n, \cdots\} \) with, for example, \( \langle \mathcal{H}_{\text{CF}} | m \rangle = \hbar \omega_m | m \rangle \). Denoting \( \omega_{mn} = \omega_m - \omega_n \) and recalling the assumption \( [\mathcal{H}_I, J_\alpha(t)] = 0 \), we derive
\begin{align}
\langle m | \frac{d J_\alpha(t)}{dt} | n \rangle &= -i \omega_{mn} \langle m | J_\alpha(t) | n \rangle \\
&= I + II + III + IV, \quad (19)
\end{align}

where the four terms labelled by \( I, \cdots, IV \) have similar mathematical structures.

We need to specify \( \mathcal{H}_I \) at this junction. We focus first on the crystal-field fluctuations induced by conduction electrons. The exchange interaction between a rare-earth ion and the conduction electron can be written as
\begin{equation}
\mathcal{H}_I = \mathcal{H}_{\text{el}} = -2I_{\text{ex}}(g - 1) \mathbf{J} \cdot \mathbf{s}, \quad (20)
\end{equation}

where \( I_{\text{ex}} \) is an exchange integral, \( g \) the Landé factor and \( \mathbf{s} \) the electron spin operator. Then, for example,
with \( G_{k\ell,mn}^\text{ph} = \langle k | V | t \rangle \langle m | V | n \rangle / \hbar^2 \) and \( f_{\text{ph}}(t) = \langle \epsilon(t) \rangle_{\text{ph}} \). Here the average is done over the phonon bath.

It is obvious that the \( I \) term for the electronic and phonon relaxation processes have the same mathematical structure. This is also true for the three other terms, that is, \( II, III \) and \( IV \). Therefore we need to solve Eq. \( \text{(19)} \) for \( J_\alpha(t) \) with the four terms on the right hand side having the mathematical structure of Eq. \( \text{(21)} \) (equivalent to Eq. \( \text{(24)} \)). Since each term is expressed as a sum of convolution products, a Laplace transform technique is well suited. That \( \mathcal{H}_1 \) factorises into lattice and crystal-field potentials is seen to be a key ingredient which allows to use the Laplace transform technique. With the notation \( Z_{\alpha,\mn}(s) = \int_0^\infty \exp(-st)(m | J_\alpha(t) | n) dt \),

\[
(s - i\omega_{\mn}) Z_{\alpha,\mn}(s) = -\sum_{p,q} G_{mp,pq} h(\omega) Z_{\alpha,qn}(s) - \sum_{p,q} G_{pq,qn} g(\omega) Z_{\alpha,mp}(s) + \sum_{p,q} G_{mp,qn} [h(\omega) + g(\omega)] Z_{\alpha,pq}(s) + \langle m | J_\alpha | n \rangle. \tag{25}
\]

We denote \( g(\omega) = \int_0^\infty f(t) \exp(i\omega t) dt \) and \( h(\omega) = \int_0^\infty f(-t) \exp(i\omega t) dt \). \( G_{k\ell,mn} \) and \( f(t) \) stand for \( G_{k\ell,mn}^\text{el} \) and \( f_\alpha(t) \) or \( G_{k\ell,mn}^\text{ph} \) and \( f_{\text{ph}}(t) \), depending on the relaxation mechanism under study. Setting \( s = \varepsilon + i\omega \) with \( \varepsilon \to 0^+ \) in the previous system of equations,

\[
i(\omega - \omega_{\mn}) Z_{\alpha,\mn}(i\omega) + \sum_{p,q} G_{mp,pq} h(-\omega + \omega_{\mn}) Z_{\alpha,qn}(i\omega) + \sum_{p,q} G_{pq,qn} g(-\omega + \omega_{\mn}) Z_{\alpha,mp}(i\omega) - \sum_{p,q} G_{mp,qn} [h(-\omega + \omega_{\mn}) + g(-\omega + \omega_{\mn})] Z_{\alpha,pq}(i\omega) = \langle m | J_\alpha | n \rangle - \langle J_\alpha \rangle \delta_{\mn}. \tag{26}
\]

As argued in Sec. \( \text{IV} \) it is a good approximation to neglect the imaginary part of \( h(\omega) \) and \( g(\omega) \). We note that

\[
h(\omega) = g(-\omega) = \exp(-\beta h\omega) g(\omega). \tag{27}
\]

The inclusion of the term \( \langle J_\alpha \rangle \delta_{\mn} \) guarantees that \( \langle J_\alpha(t) \rangle \) relax toward its thermal equilibrium value. This is further discussed for the Redfield theory by, for example, Slichter.

In fact, as explained in Sec. \( \text{III} \) \( \Omega_{+ -}(\omega) \) and \( \Omega_{zz}(\omega) \) are required. From their definitions,

\[
\Omega_{+ -}(\omega) = \frac{1}{Z_{\text{CF}}} \sum_{m,n} \exp(-\beta E_m) Z_{+ ,mn}(-i\omega) \langle n | J_- | m \rangle + \text{c.c.},
\]

\[
\Omega_{zz}(\omega) = \frac{1}{Z_{\text{CF}}} \sum_{m,n} \exp(-\beta E_m) Z_{zz,\mn}(-i\omega) \langle n | J_2 | m \rangle + \text{c.c.}. \tag{28}
\]

IV. CONDUCTION ELECTRON AND PHONON CORRELATION FUNCTIONS

In this section we evaluate the correlation functions for the conduction electrons and phonons.

Turning our attention to the electronic correlation, we
need to compute
\begin{equation}
  g_{el}(\omega) = \int_0^\infty f_{el}(t) \exp(i\omega t) dt.
\end{equation}  
(29)

We shall denote \( c_{k,\sigma}^+ \) and \( c_{k,\sigma} \) the creation and annihilation electron operators of wavevector \( k \) and spin \( \sigma \) and \( \mathcal{H}_e \) the Hamiltonian of the electron bath. The electrons are assumed to be free-like. We use the fermion representation of \( s_x \), i.e.
\begin{equation}
  s_x = \frac{1}{2} \sum_{k' \neq k} \left( c_{k',\downarrow}^+ c_{k,\uparrow} + c_{k,\downarrow}^+ c_{k',\uparrow} \right). \tag{30}
\end{equation}

\( n_q \) is the Fermi distribution function. The second line results after inserting in the first line the identity operator between the first two fermionic operators and using the Heisenberg equation. An Hartree-Fock decoupling is done and the spin dependence of the energy is neglected. We use the fermion representation of \( s_x \), i.e.
\begin{equation}
  s_x = \frac{1}{2} \sum_{k' \neq k} \left( c_{k',\downarrow}^+ c_{k,\uparrow} + c_{k,\downarrow}^+ c_{k',\uparrow} \right). \tag{30}
\end{equation}

Without lost of generality, the ion is taken to sit at the origin of the coordinates. \( f_{el}(t) \) reduces to the sum of two quantities which occur to be equal. Let us focus on one of them:

\[ b_k \text{ is the Bose distribution function. Because of energy conservation during the scattering process, the second term cannot contribute to the relaxation when } \omega > 0. \]

\[ \text{We compute} \]
\begin{equation}
  g_{el}(\omega) = \frac{\pi}{2} \frac{h \epsilon}{N} \left[ N_E \right]^2 \frac{\hbar \omega}{1 - \exp(-\beta \hbar \omega)}. \tag{32}
\end{equation}

\( N \left( E_F \right) \) is the density at the Fermi level per spin.

\[ \text{We now evaluate the phonon correlation function. It is} \]
\[ \text{convenient to express } \epsilon \text{ in terms of its Fourier components. Since the rare-earth ion can} \]
\[ \text{be taken at the origin of the coordinates (see for example Ref. [13]),} \]
\begin{equation}
  \epsilon = i \sum_k \sqrt{\frac{k^2 h}{2 N M \omega_k}} (a_k - a_k^+) \tag{33}
\end{equation}

\( N \) is the number of rare-earth ions in the compound and \( N M \) its mass. \( a_k^+ \) and \( a_k \) are the creation and annihilation phonon operators. Note that only a single branch of phonons is accounted for. It is a simple matter to derive
\begin{equation}
  f_{ph}(t) = \frac{1}{N} \sum_k \frac{k^2 h}{2 M \omega_k} \times \left[ \exp(-i\omega_k t) (b_k + 1) + \exp(i\omega_k t) b_k \right]. \tag{34}
\end{equation}

\[ \text{The imaginary part of } g_{ph}(\omega) \text{ is again neglected. The factor 3 is added to account for the three phonons branches. The } k \text{ sum, which extends over the Brillouin zone, is performed assuming } \omega_k = v_k k. \quad \rho \text{ is the crystal density and } v_s \text{ the sound velocity.} \]

\[ \text{V. MODEL COMPUTATIONS OF THE CORRELATION FUNCTIONS AND THE } \mu \text{SR SPIN-LATTICE RELAXATION RATE} \]

\[ \text{In this section we present analytical results obtained for simple rare-earth level schemes, our purpose being to understand the implications, on the correlation functions and } \mu \text{SR relaxation rate, of the crystal-field-induced relaxation by electrons and phonons. Here we shall focus our analysis on } \Omega_{zz}(\omega) \text{. As mentioned in Sec. [11] } \lambda_Z \text{ is the weighted sum of two spin-correlation functions for a compound with at least a three-fold symmetry axis. We shall assume, for simplicity, } \lambda_Z \text{ to be proportional only to } \Lambda_{zz}(\omega = 0) = \Omega_{zz}(\omega = 0). \]

\[ \text{Without lost of generality, the ion is taken to sit at the origin of the coordinates. } f_{el}(t) \text{ reduces to the sum of two quantities which occur to be equal. Let us focus on one of them:} \]

\[ \text{We now evaluate the phonon correlation function. It is} \]
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  \epsilon = i \sum_k \sqrt{\frac{k^2 h}{2 N M \omega_k}} (a_k - a_k^+) \tag{33}
\end{equation}

\( N \) is the number of rare-earth ions in the compound and \( N M \) its mass. \( a_k^+ \) and \( a_k \) are the creation and annihilation phonon operators. Note that only a single branch of phonons is accounted for. It is a simple matter to derive
\begin{equation}
  f_{ph}(t) = \frac{1}{N} \sum_k \frac{k^2 h}{2 M \omega_k} \times \left[ \exp(-i\omega_k t) (b_k + 1) + \exp(i\omega_k t) b_k \right]. \tag{34}
\end{equation}
We first discussed a result obtained from the iterative method detailed here against a published one derived from the memory function method that was applied to the electronic relaxation of a Ce$^{3+}$ in cubic symmetry. The six crystal-field levels of Ce$^{3+}$ split into a doublet $\Gamma_7$ and a quartet $\Gamma_8$ located at such a high energy that it can be neglected for our purpose. We label the two states of $\Gamma_7$ as $|1\rangle$ and $|2\rangle$. They are $|1\rangle = (1/6)^{1/2} |5/2\rangle - (5/6)^{1/2} |-3/2\rangle$ and $|2\rangle = (1/6)^{1/2} |-5/2\rangle - (5/6)^{1/2} |3/2\rangle$. We compute $\langle 1|J_z|2\rangle = (1/2)J_z|1\rangle = 0$ and $\langle 2|J_z|2\rangle = -(1/2)J_z|1\rangle$. It is easily shown that $Z_{z,12} = Z_{z,21} = 0$ and $Z_{z,11} = -Z_{z,22}$. Therefore we only need to study Eq. 26 for $m = n = 1$. We expect $\Omega_{zz}(\omega)$ to be a Lorentzian function. This is obtained setting $\omega = 0$ in the arguments of the $h$ and $g$ functions. This low-energy approximation will be always done in this section for simplicity. Obviously, it can be easily overcome.

$$i\omega Z_{z,11} + 4h_{el}(\omega = 0)G_{12,21}^{el}Z_{z,11} = \langle 1|J_z|2\rangle.$$  \hspace{1cm} (36)

The relaxation arises from a $G_{kk,mm}$ term that satisfies, as expected, the secular condition $\omega_{kk} = \omega_{mm}$. We compute

$$\Omega_{zz}(\omega) = \frac{g_T^2}{2\omega^2 + \Gamma_{zz}^2},$$ \hspace{1cm} (37)

with the Korringa law$^{13}$

$$\Gamma_{zz} = 4\pi g_T^2 [I_{ex}(g - 1)N(E_F)]^2 k_B T. \hspace{1cm} (38)$$

$g_T = 2\langle 2|J_z|2\rangle = 5/3$ is the spectroscopic factor of $\Gamma_7$. This result was published by Becker and collaborators$^{10}$. From a straightforward computation,

$$\Lambda_{zz}(\omega = 0) = \frac{h}{8\pi [I_{ex}(g - 1)N(E_F)]^2 k_B T} \hspace{1cm} (39)$$

This model predicts $\lambda_Z \propto 1/T$. Such a behaviour was first reported for ErAl$^{19}$. According to Eq. 36, the linewidth is proportional to $h_{el}(\omega = 0)$. If we had considered the phonon relaxation mechanism, we would have found the linewidth to be proportional to $h_{ph}(\omega = 0)$. Hence, $\Gamma_{zz,ph} = 0$, i.e. in contrast to the electronic case the coupling of a doublet to the strain field does not broaden the quasi-static scattering.

The work reported in this paper has been originally motivated by the observation for some geometrically frustrated magnetic materials of a strong temperature dependence of $\lambda_Z$ at high temperature and sometimes a plateau at low temperature for measurements performed in zero field$^{19,20,21,22}$. These materials are insulators, and therefore only the phonon mechanism can play a role.

The physics at high temperature can be understood with a simple crystal-field scheme of three levels at energies $(2|H_{CF}|2) = \delta$, $(3|H_{CF}|3) = \Delta$ with $\Delta \gg \delta$ and zero. The ground state is the $|1\rangle$ state with energy equal to zero. Since our interest is on the low-energy physics, we study Eq. 26 with $m = 1$ and $n = 2$. In addition, assuming $\delta$ negligible,

$$\left(i\omega + \Gamma_{zz}^{(12)}\right)Z_{z,12} = \langle 1|J_z|2\rangle, \hspace{1cm} (40)$$

where

$$\Gamma_{zz}^{(12)} = h_{ph}(\Delta/h)\left(G_{13,31}^{ph} + G_{23,32}^{ph}\right). \hspace{1cm} (41)$$

The contributions to this equation come only from the two terms that satisfy the secular condition $\omega_{kk} = \omega_{mm}$ for $G_{kk,mm}^{ph}$. The other terms oscillate with an angular frequency $\omega_\Delta = \Delta/h \approx 10^{15}\text{rad s}^{-1}$ for $\Delta/k_B = 100$ K, and therefore average out to zero in the time scale of the measurements. This simple model predicts a quasi-elastic scattering. Assuming further $\beta\Delta \gg 1$ gives $\Gamma_{zz,ph}^{(12)} \propto \exp(-\beta\Delta)$ and therefore an activated behaviour for $\lambda_Z(T)$ is found. Components such as $Z_{z,31}$ are not relevant for the quasi-elastic response and $\lambda_Z$ since they are strongly inelastic. This argument is valid if the temperature is not too large so that $Z_{z,31}$ is a Lorentzian function of $\omega$ and so the contribution of $Z_{z,31}$ at $\omega = 0$ is negligible. The relaxation we are discussing is Orbach-like. It involves a real two-phonon scattering: one phonon induces a transition between $|1\rangle$ and $|3\rangle$ and the other between $|3\rangle$ and $|2\rangle$. It has been identified experimentally in $\mu$SR data$^{22}$. This scattering occurs only if phonons of the proper energy are available. For a compound with a Debye phonon spectrum, this means that the excitation energy $\Delta$ cannot be larger than $k_B \Theta_D$ where $\Theta_D$ is the Debye temperature. To complete our analysis of the high temperature regime, we note that $\Gamma_{zz,ph}^{(12)}$ is found to be proportional to $T$ when $\beta\Delta \ll 1$. Therefore $\lambda_Z \propto 1/T$ when the thermal energy exceeds the crystal-field energy. Amusingly, the Korringa relaxation gives an identical temperature dependence.

The same crystal-field scheme can be used to study the physics at low temperature. The high-energy level is now neglected. Focusing again on the low-energy sector, $Z_{z,11}$ and $Z_{z,22}$ are the relevant components because $\delta$ is no more negligible. They obey a system of two coupled linear equations:

$$i\omega Z_{z,11} + 2h_{ph}(\delta/h)G_{12,21}^{ph} (Z_{z,11} - Z_{z,22}) = \langle 1|J_z|1\rangle - \langle J_z \rangle,$$

$$i\omega Z_{z,22} - 2h_{ph}(\delta/h)G_{22,21}^{ph} (Z_{z,11} - Z_{z,22}) = (2|J_z|2) - \langle J_z \rangle. \hspace{1cm} (42)$$

This system can be solved for $Z_{z,11}$ and $Z_{z,22}$ and $\Omega_{zz}(\omega)$ evaluated. Here we simply consider two limits. If $\beta\Delta \gg 1$, $Z_{z,11}$ controls the quasi-elastic linewidth. Since $h_{ph}$ and $(\langle 1|J_z|1\rangle - \langle J_z \rangle)$ vanish, the width of the quasi-elastic line vanishes also. Hence, the coupling of the strain field to the crystal field does not induce a linewidth. In the limit $\beta\Delta \ll 1$, $Z_{z,11}$ and $Z_{z,22}$ have an equal weight in the expression for $\Omega_{zz}(\omega)$. Since they are of opposite sign,
\( \Omega_z(\omega) = 0 \). Hence, in the high-temperature limit of the two-level system the magneto-elastic coupling does not induce a linewidth. However, as our study of the three-level system has shown, the energy levels located at energy above \( \delta \) may contribute to the relaxation.

In summary, in this section we have found that the magneto-elastic coupling can effectively explain the activated behaviour of \( \Lambda_z \) found at relatively high temperature for some insulators. The same coupling gives a vanishing linewidth at low temperature. However, other mechanisms can contribute to it. Such a mechanism is unveiled in the next section.

### VI. Spin-Correlation for a Regular Lattice of Paramagnetic Ions

Here we study the effect of the magnetic interactions between the ions located in a regular crystal lattice. Hence, rather than \( \mathcal{H}_0 = \mathcal{H}_{CF} + \mathcal{H}_L \) with \( \mathcal{H}_L = \mathcal{H}_{el} \) or \( \mathcal{H}_L = \mathcal{H}_{ph} \), we write \( \mathcal{H}_0 = \mathcal{H}_{CF} + \mathcal{H}_L + \mathcal{H}_H \) where \( \mathcal{H}_H \) describes the ion-ion magnetic interaction. We shall limit ourselves to the two-ion Heisenberg Hamiltonian, i.e.

\[
\mathcal{H}_H = - \sum_{i,j} \mathcal{J}(ij) \mathbf{J}_i \cdot \mathbf{J}_j,
\]

where \( \mathcal{J}(ij) = 0 \) when \( i = j \). Introducing the following relation in the previous equation,

\[
\mathcal{J}(ij) = \frac{1}{N} \sum_{\mathbf{q}} \mathcal{J}_q \exp \left[ i \mathbf{q} (i - j) \right],
\]

we derive

\[
\mathcal{H}_H = - \frac{1}{N} \sum_{\mathbf{q}} \mathcal{J}_q \mathbf{J}_q \cdot \mathbf{J}_{-\mathbf{q}}.
\]

We have used the relation

\[
\mathbf{J}_q = \sum_i \mathbf{J}_i \exp (-i \mathbf{q} \cdot \mathbf{i}).
\]

We recall that the magnetic ions are taken to sit on a Bravais lattice for simplicity. As in Sec. III it is usually convenient to work with continuous rather than discrete wavevectors. This leads to

\[
\mathcal{H}_H = -\nu \int \mathcal{J}(\mathbf{q}) \mathbf{J}(\mathbf{q}) \cdot \mathbf{J}(-\mathbf{q}) \frac{d^3 \mathbf{q}}{(2\pi)^3},
\]

with

\[
\mathcal{J}(\mathbf{q}) = \sum_i \mathcal{J}(ij) \exp [-i \mathbf{q} \cdot (i - j)].
\]

From now on we shall work with the reference frame \( \{+^\prime, -^\prime, \mathbf{z}\} \) where the spin-correlation tensors are diagonal. To simplify the notation and because \( \Lambda_z(\omega) \) is simply \( \Lambda_z(\omega) \) (see Sec. III, we shall only consider that correlation. Rather than \( \Lambda_z(\omega) \) we have to evaluate the wavevector dependent symmetrized correlation function

\[
\Lambda_z(\mathbf{q}, \omega) = \int_{-\infty}^{\infty} \langle \{J_z(\mathbf{q}, t)J_z(-\mathbf{q}, 0)\}\rangle \exp(i\omega t)dt,
\]

where \( 2 \langle \{J_z(\mathbf{q}, t)J_z(-\mathbf{q}, 0)\}\rangle = \langle J_z(\mathbf{q}, t)J_z(-\mathbf{q}, 0)\rangle + \langle J_z(-\mathbf{q}, 0)J_z(\mathbf{q}, t)\rangle \).

In general the available theories does not consider the correlation tensor but rather the generalized susceptibility tensor. The fluctuation-dissipation theorem provides a relation between the elements of the two tensors, see for example Ref. [23]. Denoting the imaginary part of \( \chi_z(\mathbf{q}, \omega) \) as \( \text{Im}\{\chi_z(\mathbf{q}, \omega)\} \),

\[
\Lambda_z(\mathbf{q}, \omega) = \frac{\hbar V}{\mu_0 g^2 \mu_B^2} \coth \left( \frac{\beta \hbar \omega}{2} \right) \text{Im}\{\chi_z(\mathbf{q}, \omega)\}.
\]

Therefore we need to compute \( \text{Im}\{\chi_z(\mathbf{q}, \omega)\} \).

A practical theoretical method to describe the effect of the intersite correlations on the susceptibility is the RPA which leads to the following simple formula:

\[
\tilde{\chi}(\mathbf{q}, \omega) = \left\{ I - \frac{2V \mathcal{J}(\mathbf{q})}{\mu_0 g^2 \mu_B^2} \tilde{\chi}(\omega) \right\}^{-1} \tilde{\chi}(\omega).
\]

\( \{A\}^{-1} \) stands for the inverse of tensor \( A \). \( \tilde{\chi}(\omega) \) is the susceptibility tensor for a magnetic ion in the lattice, the intersite interactions being switched off. The elements of the tensors \( \tilde{\chi}(\omega) \) and \( \tilde{\Lambda}(\omega) \) are related. Using again the fluctuation-dissipation theorem,

\[
\text{Im}\{\chi_z(\omega)\} = \frac{\mu_0 g^2 \mu_B^2}{\hbar V} \tanh \left( \frac{\beta \hbar \omega}{2} \right) \Lambda_z(\omega).
\]

The real and imaginary parts of \( \chi_z(\omega) \) are linked through the Kramers-Kronig theorem:

\[
\text{Re}\{\chi_z(\omega)\} = -\frac{1}{\pi} \mathcal{P} \int_{-\infty}^{\infty} \frac{\text{Im}\{\chi_z(\omega)\}}{\omega - u} du.
\]

\( \mathcal{P} \) stands for the principal part of the integral. Thus, once tensor elements such as \( \Lambda_z(\omega) \) are known, the elements of \( \tilde{\chi}(\omega) \) can be computed using Eqs. 52 and 53. Then, \( \text{Im}\{\chi_z(\mathbf{q}, \omega)\} \) can be determined from Eq. 51. Finally \( \Lambda_z(\mathbf{q}, \omega) \) is obtained recalling Eq. 50. Within our conventions \( \tilde{\chi}(\mathbf{q}, \omega) \) and \( \tilde{\Lambda}(\omega) \) are dimensionless.

Since the tensors are diagonal in the reference frame \( \{+^\prime, -^\prime, \mathbf{z}\} \), Eq. 51 can be solved easily for \( \tilde{\chi}(\mathbf{q}, \omega) \). Combined with Eq. 50

\[
\tilde{\chi}(\mathbf{q}, \omega) = \left\{ I - \frac{2V \mathcal{J}(\mathbf{q})}{\mu_0 g^2 \mu_B^2} \tilde{\chi}(\omega) \right\}^{-1} \tilde{\chi}(\omega).
\]
Therefore, if \( \Lambda_z(\omega) \) and \( \mathcal{J}(\mathbf{q}) \) are known, it is possible to compute numerically \( \Lambda_z(\mathbf{q}, \omega) \) and \( \Lambda_z \) for a given muon site using the material of Sec. III. We can proceed further analytically for the elastic case, i.e. when \( \omega = 0 \). For definiteness, we assume \( \Lambda_z(\omega) \) to be Lorenzian, i.e.

\[
\Lambda_z(\omega) = \frac{2(J_z^2) \Gamma_z}{\omega^2 + \Gamma_z^2} \quad (55)
\]

We derive

\[
\Lambda_z(\mathbf{q}, \omega) = \Lambda_z(\omega = 0) \left[ 1 - \frac{4(J_z^2)\mathcal{J}(\mathbf{q})}{J_z^2} \int_{-\infty}^{\infty} \frac{1}{x(x^2+1)} \tanh \left( \frac{\beta \hbar \omega}{2} x \right) dx \right]^2 = \frac{2(\mathcal{J}^2)}{\Gamma_z} \left[ 1 - \frac{4(J_z^2)\mathcal{J}(\mathbf{q})}{J_z^2} \int_{-\infty}^{\infty} \frac{1}{x(x^2+1)} \tanh \left( \frac{\beta \hbar \omega}{2} x \right) dx \right]^2. \quad (56)
\]

To understand this expression we consider the consequences of a magnetic phase transition. That transition is characterized, among other properties, by its critical temperature \( T_c \) and a wavevector \( \mathbf{q}_c \) which sets the repetition length of the magnetic structure for \( T < T_c \). At \( T_c \), the generalized susceptibility diverges for \( \mathbf{q} = \mathbf{q}_c \), and so does \( \Lambda_z(\mathbf{q}_c, \omega = 0) \). From the previous equation

\[
\frac{\hbar}{\mathcal{J}(\mathbf{q}_c)} = \frac{2(\mathcal{J}^2)}{\Gamma_z} \frac{1}{\pi} \int_{-\infty}^{\infty} \frac{1}{x(x^2+1)} \tanh \left( \frac{\hbar \Gamma_z}{2k_B T_c} x \right) dx. \quad (57)
\]

Let us study \( I(\alpha) = \int_{-\infty}^{\infty} \frac{1}{x(x^2+1)} \tanh(\alpha x) dx \). We evaluate numerically \( I(1) = 2.08 \). This is clearly smaller than \( \int_{-\infty}^{\infty} \frac{1}{x(x^2+1)} dx = \pi \). However, smaller is \( \alpha \) better is the approximation of linearising the function \( \tanh(\alpha x) \). In our case \( \alpha = (h \Gamma_z)/(2k_B T_c) \). Physically, we expect \( h \Gamma_z \gg k_B T_c \) for a conventional phase transition. Assuming the linearisation to be valid, we get

\[
\mathcal{J}(\mathbf{q}_c) = \frac{k_B T_c}{2(\mathcal{J}^2)}. \quad (58)
\]

This is consistent with the mean-field result.

We known that \( \lambda_c \) in zero field is given as a sum over the Brillouin zone of \( \Lambda_z(\mathbf{q}, \omega = 0) \). Approaching the critical point, the sum is expected to be dominated by \( \Lambda_z(\mathbf{q}_c, \omega = 0) \) since that correlation diverges at \( T_c \). How does it diverge? According to our RPA result of Eq. 59 upon linearising the \( \tanh \) function, we derive

\[
\Lambda_z(\mathbf{q}_c, \omega = 0) = \Lambda_z(\omega = 0) \left( \frac{T}{T - T_c} \right)^2. \quad (59)
\]

While Eq. 59 is obviously not valid in the critical regime since the RPA is a kind of mean-field approximation, it should provide a good description deep in the paramagnetic regime. Note that \( \Lambda_z(\mathbf{q}_c, \omega = 0) \) reaches a finite temperature independent value at high temperature.

Now we study \( \Lambda_z(\mathbf{q}, \omega = 0) \) in the limit \( T \gg T_c \) (\( T = T_C \) for a ferromagnet and \( T_c = T_N \) for an antiferromagnet). We find:

\[
\Lambda_z(\mathbf{q}, \omega = 0) = \Lambda_z(\omega = 0) \left( 1 + 2 \frac{\mathcal{J}(\mathbf{q})}{\mathcal{J}(\mathbf{q}_c)} \frac{T}{T - T_c} \right). \quad (60)
\]

Because \( \mathcal{J}(\mathbf{q}_c) > 0 \), the sign of the thermal correction at high temperature is given by the sign of \( \mathcal{J}(\mathbf{q}) \). Referring to Eq. 27 positive and negative \( \mathcal{J}(\mathbf{q}) \) values correspond to ferromagnetic and antiferromagnetic interactions at wavevector \( \mathbf{q} \), respectively. This means that a ferromagnetic (antiferromagnetic) interaction increases (decreases) the correlation function \( \Lambda_z(\mathbf{q}, \omega = 0) \). This result was derived a long time ago, but the wavevector dependence of the interaction was overlooked. The 1/T dependence of \( \Lambda_z(\mathbf{q}, \omega = 0) \) is a typical pair correlation effect at high temperature.

The result of the RPA approximation is quite simple, in particular for \( \Lambda_z(\mathbf{q}, \omega = 0) \). When \( \Lambda_z(\omega = 0) \) is determined, \( \Lambda_z(\mathbf{q}, \omega = 0) \) can be evaluated by dividing \( \Lambda_z(\omega = 0) \) by a proper denominator. However, in Sec. IV we have found \( \Lambda_z(\omega = 0) \) to vanish if the thermal energy is much larger than the crystal-field energy \( E_{\text{CF}} \). When the magneto-elastic coupling drives the crystal-field relaxation. In this case \( \lambda_c \) should drop as the compound is warmed up far above \( E_{\text{CF}}/k_B \) since \( \Lambda_z(\mathbf{q}, \omega = 0) \) becomes negligible. This is inconsistent with the experimental finding for the garnet \( \text{Yb}_3\text{Ga}_5\text{O}_{12} \). To understand the origin of this problem, we need first to specify the \( \text{Yb}^{3+} \) level scheme. Because \( \text{Yb}^{3+} \) is a Kramers ion, the states are at least doubly degenerate. The mean distance between the doublet ground state and the three closely spaced excited doublets is \( \Delta/k_B \approx 850 \text{ K} \). Since the ground state is a doublet, \( E_{\text{CF}} = 0 \). In addition, \( \beta \Delta \gg 1 \). Hence, from the RPA approximation we expect an activated behaviour for \( \lambda_c(T) \) outside the critical
paramagnets given in Ref. \[5\]. We get:

The spectral function of the self-correlation to be Gaussian much simpler method to estimate $\Gamma$ the sum can only be done numerically. Here we propose the formulae for because it assumes a non-zero single ion susceptibility cannot be described by the RPA approximation we use itily done. Referring to Eq. 55 for the definition of $\Gamma_z$, we could have gone a step further, keeping the term in powers of the strain and we have kept the linear term. It involves two virtual phonons and it is described by setting $\Lambda_z(\omega = 0)$ equal to the sum over the Brillouin zone of $\Lambda_z(q, \omega)$. However, the formulae for $\chi_z(q)$ and $F_z(q, \omega)$ are approximate and the sum can only be done numerically. Here we propose a much simpler method to estimate $\Gamma_{z,\text{exc}}$. We suppose the spectral function of the self-correlation to be Gaussian and use the expression of the second moment for paramagnets given in Ref. \[5\]. We get:

$$\Gamma_{z,\text{exc}} = \sqrt{\frac{16J_z^2 z(J+1)}{3\pi\hbar^2}}. \quad (61)$$

Interestingly, $\Gamma_{z,\text{exc}}$ depends on the two-ion exchange interaction $J$ (see Eq. 53, $J = J(ij)$) that we have limited to the $z$ nearest neighbour ions. Therefore it reflects the environment of the ions under study. $\Gamma_{z,\text{exc}}$ is temperature independent in the high-temperature limit we take and for the conventional magnets we consider, i.e. we do not discuss complicated compounds such as the frustrated ones.

The inclusion of the phonon-induced relaxation is easily done. Referring to Eq. 55 for the definition of $\Gamma_z$,

$$\Gamma_z = \Gamma_{z,\text{exc}} + \Gamma_{z,\text{ph}}. \quad (62)$$

In the same way, for the electronic relaxation we have

$$\Gamma_z = \Gamma_{z,\text{exc}} + \Gamma_{z,\text{el}}. \quad (63)$$

$\Gamma_{z,\text{el}}$ and $\Gamma_{z,\text{ph}}$ are obviously obtained from the study of $\Omega_{z}(\omega)$. Eq. 62 has been proven to provide a good fit of $\lambda_z(T)$ observed for Yb$_3$Ga$_5$O$_{12}$\[20\] but only for $T > 0.4$ K. The increase of $\lambda_z$ for $T_c < T < 0.4$ K arises from the intersite correlations. The RPA provides a model for these correlations, but outside of the critical regime.

In conclusion, the analysis of the neutron quasi-elastic linewidth and $\mu$SR spin-lattice relaxation rate should be done, at least in a first approximation, with Eq. 53 or Eq. 60 and using for the single ion linewidth the expression given at Eq. 66 or Eq. 63.

## VII. Discussion and Conclusion

We have studied the symmetrised correlation functions as measured by neutron scattering and the $\mu$SR spin-lattice relaxation rate. We have focused our work on the paramagnetic state of magnetic materials made of a regular lattice of rare-earth ions. The relaxation of the crystal-field levels has been assumed to be driven by the conduction electrons or the lattice vibrations. Mathematically, we have used the iterative Laplace transform method due to P. M. Richards. The interaction between the ions has been modelled with the random-phase approximation. A phenomenological modification of this approximation has been proposed to cure its breakdown which comes into light when the crystal-field levels are not closely packed. Thanks to the point symmetry at the rare-earth site, for many compounds of interest, we have shown that at most two symmetrised correlation functions are relevant.

Through the study of simple crystal-field models, the Orbach relaxation mechanism has been uncovered for the linewidth and $\lambda_z$. We have argued for a single ion that the linewidth should vanish at low temperature if the relaxation is driven by the lattice vibrations. However, physically this limit cannot be reached because of the on-site fluctuations described by $\Gamma_{z,\text{exc}}$ in Sec. VI which set a lower bound on the linewidth and therefore an upper bound on $\lambda_z$.

As clearly seen from its derivation, the Richard’s system of linear equations is obtained if the Hamiltonian describing the interaction of the lattice field with the crystal field can be factorised into these two fields. This sets limits on the relaxation mechanisms which can be handled. For example, the Raman mechanism cannot be included. It involves two virtual phonons and it is described by treating the interaction Hamiltonian in second order.\[16\] It leads to a linewidth $\Gamma_{\text{ph}} \propto T^{-n}$ with a large value for the exponent $n$, typically $n = 5$ or $n = 7$. According to Electron Paramagnetic Resonance data, this mechanism can appear only at low temperatures.\[20\]

The modulation of the crystal field has been expanded in powers of the strain and we have kept the linear term. We could have gone a step further, keeping the term quadratic in strain. This would have introduced at least one new free parameter. Unless really justified for a particular physical problem, we do not believe it is worthwhile.

Although our interest in this paper has been on the paramagnetic phase of a magnetic material, the method
we have discussed can be extended to its ordered magnetic state. One has to account for the molecular field acting on the rare-earth ion by adding a Zeeman term to $H_{\text{CF}}$ as done in Ref. \cite{27}. The RPA models the magnetic fluctuations due to the two-ion interaction.

In conclusion, we have proposed a framework to analyze the linewidth as measured by neutron scattering and the $\mu$SR relaxation rate for compounds with rare-earth sublattices. We take into account the electronic and phonon-induced relaxation of the crystal-field levels. We have proposed a phenomenological modification of the result of random-phase approximation which appears for some particular crystal-field level schemes.

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