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Abstract

Using the Mori formalism, a general theory is developed for the calculation of paramagnetic relaxation effects in a general crystal field doublet. For systems with extreme magnetic anisotropy, the relaxation rates show a strong angular dependence, being severely peaked in a direction perpendicular to the magnetic easy axis. Application to the RRh_4B_4 (R = rare-earth) compounds is discussed.

While a very substantial body of literature exists describing experimental results and theoretical interpretations for paramagnetic relaxation data, relatively little attention has been paid to situations where the system being investigated shows extreme magnetic anisotropy. Recent experimental work on RRh_4B_4 (R = rare-earth) systems$^{(1)}$ have shown a number of anomalous features which available theoretical formulations are not able to describe. In the following we will discuss the general problem of spin relaxation in two-level systems, with application to highly anisotropic, Ising-like ground states.

We consider a system of magnetic ions having angular momentum J in a crystalline electric field (CEF) with a large axial anisotropy along a fixed (z) axis. If the sign of the CEF terms causes the large J_z states to be low-lying, and no off-diagonal CEF terms appear, then we generate a set of magnetic doublets where the ground state doublet is purely $J_z = \pm J$, the next doublet has $J_z = \pm(J-1)$ with energy separation Δ, etc. If there are, in addition, small off-diagonal CEF terms and/or the system is in an applied field H ($\Delta >> g_J\mu_B H$) oriented at an angle θ with the z axis, then each CEF doublet will have small admixtures of other angular momentum values. At temperatures small compared to the CEF splittings, this produces a system having very large magnetic anisotropies. It should be noted that relaxation cannot take place in the ground state doublet through normal $\Delta J_z = \pm 1$ processes without the admixture terms described above, if $J > 1$. Thus the nature of the ground state relaxation is different from that of simple $S = 1/2$ doublets. We also note that many such CEF situations are known in rare-earth compounds, although detailed spin relaxation data are relatively infrequent.

In the following we consider temperatures small compared to the CEF splittings, i.e. we focus on the ground state doublet and consider an effective two level system. We begin with a complete set of angular momentum states $\{|m\rangle\}$ for each magnetic site defined by

$$J_z |m\rangle = m|m\rangle; \qquad (-J < m < J), \qquad (1)$$

and a Hamiltonian

$$\mathcal{H}_o = \mathcal{H}_{axial} + \mathcal{H}' - g_J \mu_B \vec{H} \cdot \vec{J}. \qquad (2)$$

The diagonal CEF operator $\mathcal{H}_{axial}$ is a function of J_z^2, $\mathcal{H}'$ contains the off-diagonal CEF terms involving J_x and J_y, and H is the magnetic field applied at an arbitrary angle θ to the z axis. The eigenstates of this Hamiltonian are written

$$\mathcal{H}_o|M\rangle = E_M|M\rangle \; ; \quad |M\rangle = \sum_{m=-J}^{J} \lambda_{Mm} \; |m\rangle . \qquad (3)$$

We consider the CEF parameters to be known. Therefore, the solution of Eq. (2) gives the E_M and λ_{Mm} as known functions of those CEF parameters, as well as of H and θ.

Transitions between the states $|M\rangle$ which lead to relaxation are caused by some coupling to an external lattice, designated by V. We have considered the on-site exchange with the conduction electron system (Korringa mechanism), as well as a general dipolar-like coupling between the magnetic ions which leads to concentration dependent effects in the relaxation. Both these interactions contain the operators $\vec{J}$ for the various sites, which we express at each site through the exact relation

$$J_\alpha = \sum_{M,M'} \langle M \,|J_\alpha|\, M'\rangle \; |M\rangle\langle M'| \quad ; \quad \alpha=x,y,z \qquad (4)$$

where the $\langle M|\, J_\alpha|M'\rangle$ are easily obtained from the known wave-fuctions $|M\rangle$. For our two level system, we label the states as $|1\rangle$ and $|\bar{1}\rangle$, with $E_{\bar{1}} > E_1$. Using Eqs. (3) and (4) and the completeness relation $|1\rangle \langle 1| + |\bar{1}\rangle \langle \bar{1}| = 1$, we obtain

$$J_\alpha = \frac{1}{2}\,[\langle 1|J_\alpha|1\rangle + \langle\bar{1}|J_\alpha|\bar{1}\rangle] + \langle 1|J_\alpha|\bar{1}\rangle\,|1\rangle\langle\bar{1}| + \langle\bar{1}|J_\alpha|1\rangle\,|\bar{1}\rangle\langle 1|$$

$$+ \frac{1}{2}\,[\langle 1|J_\alpha|1\rangle - \langle\bar{1}|J_\alpha|\bar{1}\rangle]\,[\,|1\rangle\langle 1| - |\bar{1}\rangle\,\langle\bar{1}1|\,|]. \qquad (5)$$

From (5), it is clear that the ability of the lattice to induce transitions between the states $|1\rangle$ and $|\bar{1}\rangle$ depends on the coefficients $\langle 1|J_\alpha|\bar{1}\rangle = \langle\bar{1}|J_\alpha|1\rangle^*$ which connect them.

The experimental systems of interest here are the compounds $Y_{1-c}R_cRh_4B_4$ ($c \ll 1$; R = Dy, Ho). Using the CEF parameters of Dunlap et al.,$^{(2)}$ we find $|\langle 1|J_x|\bar{1}\rangle|$, $|\langle 1|J_y|\bar{1}\rangle| < 10^{-5}$ for both Dy and Ho at all θ for $H < 20$ kOe. Such terms are therefore not effective in generating relaxation. For both Ho and Dy, we find $\langle 1|J_z|\bar{1}\rangle < 10^{-2}$ for $\theta \neq 90°$. If, on the other hand, $\theta \cong 90°$ we find for the non-Kramers ion (Ho), $\langle 1|J_z|\bar{1}\rangle \sim 1$ in weak fields and shows a little field dependence while for the Kramers ion (Dy), $\langle 1|J_z|\bar{1}\rangle$ is small in weak fields, but is ~1 for only $H > 10$ kOe. Physically, the peaking of relaxation around 90° occurs from field-induced admixtures of other angular momentum states as the system moves from J_z quantization to J_x quantization.

From Eq. (5), we see that the angular momentum operators J_α are formed from the three basic excitations, A_ν,

$$A_1 = \frac{1}{2}\,[\,|1\rangle\,\langle 1| - |\bar{1}\rangle\langle\bar{1}|\rangle],\ A_2 = A_3^{\dagger} = |1\rangle\langle\bar{1}|. \qquad (6)$$

Thus the evaluation of the response functions $(J_\alpha(t), J_\alpha(0))$ requires the evaluation of $C_{\nu\nu'}(t) \equiv (A_\nu(t), A_{\nu'}(0))$, where the notation (,) is that in the Mori formalism.(3) From standard Mori theory we find

$$\frac{\partial}{\partial t} C_{\nu\nu'}(t)+i\omega_\nu C_{\nu\nu'}(t) = -\sum_{\nu_1} \int_o^t dt' \Gamma_{\nu\nu_1}(t-t')C_{\nu_1\nu'}(t') \tag{7}$$

where ω_ν is the frequency associated with A_ν and the $\Gamma_{\nu\nu_1}(t)$ are the memory functions associated with the interactions V which cause the decay of $C_{\nu\nu'}(t)$. The Korringa mechanism leads to simple exponential decays of $C_{\nu\nu'}(t)$ with Lorentizian line shapes and no concentration dependence. The pair interactions among the magnetic ions lead to a Gaussian (t^2) time decay, dependent on the concentration.

To solve Eq. (7), we replace $C_{\nu_1\nu'}(t')$ by $C_{\nu_1\nu'}(t)$. As pointed out by Abragam,(4) this gives accurate results for the absorption spectra (related to the Fourier transform of $C_{\nu\nu'}$) in cases where the fluctuations in the local field as given by $\Gamma_{\nu\nu_1}(t)$ are either fast or slow. For slow fluctuations, the approximation is obvious. For fast fluctuations, the error lies far out in the wings of the absorption curve and of no interest.

Within this approximation, we have

$$C_{\nu\nu'}(t) = e^{-i\omega_\nu t} \sum_{\nu_1} [e^{-f(t)}]_{\nu\nu_1} C_{\nu_1\nu'}(0) \quad , \qquad (8)$$

with the decay being determined by the matrix

$$f_{\nu\nu_1}(t) = \int_0^t dt' \exp[i(\omega_\nu - \omega_{\nu_1})t'] \int_0^{t'} dt'' \Gamma_{\nu\nu_1}(t'') \exp[i\omega_{\nu_1} t''] . \qquad (9)$$

In the low frequency limit, we find

$$f(t) = R_K t + G t^2/2 \quad . \qquad (10)$$

The first term is the Korringa contribution, with a relaxation matrix

$$R_K = \frac{\pi}{2} \left[\frac{I_{ex} N(0)}{2}\right]^2 \left(\frac{k_B T}{\hbar}\right) m_K \quad , \qquad (11)$$

where m_K is a well-defined matrix involving matrix elements of the operators J_α, and so is completely determined by the CEF.

The second term of Eq. (10) is related to pair interactions among the magnetic ions, with G depending on the nature of the interaction. For Heisenberg coupling, we find

$$G = W \sum_{\ell_1} \left[\frac{2 V_{\ell\ell_1}}{\hbar}\right]^2 \quad , \qquad (12)$$

where $V_{\ell\ell_1}$ is the exchange coupling between sites ℓ and ℓ_1, and W is again a well-defined matrix related to matrix elements of J_α. If $c << 1$, then a configurational average of this Gaussian like behavior gives a simple exponential decay, as shown by McHenry et al.(5) That is,

$$\langle\langle e^{-Gt^2/2} \rangle\rangle = e^{-R_H t} \qquad (13)$$

where

$$R_H = c\langle V^2 \rangle^{1/2} m_H \quad ; \quad m_H^2 = W \ . \qquad (14)$$

Thus R_H is linear in the concentration, c, and proportional to the average exchange coupling. With Eqs. (10) and (13), the configurationally averaged correlation functions become

$$\langle\langle C_{\nu\nu'}(t) \rangle\rangle = e^{-i\omega_\nu t} \sum_{\nu_1} [e^{-(R_K+R_H)t}]_{\nu\nu_1} C_{\nu_1\nu'}(0) \ . \qquad (15)$$

With this expression, the appropriate relaxation functions can be obtained for arbitrary crystal field interactions.

We have outlined a general theory for the calculation of magnetic relaxation effects for a generalized CEF doublet, including both single ion and pair interaction mechanisms. Details of the calculation, as well as application to the RRh_4B_4 data, will be published separately.

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