Spin-wave contributions to nuclear magnetic relaxation in magnetic metals

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The longitudinal and transverse nuclear magnetic relaxation rates $1/T_1(T)$ and $1/T_2(T)$ are calculated for three-dimensional (3D) and two-dimensional metallic ferromagnets (FM) and antiferromagnets (AFM) with localized magnetic moments in the spin-wave temperature region. The contribution of the one-magnon decay processes is strongly enhanced in comparison with the standard T-linear Korringa term, especially for the FM case. For the 3D AFM case this contribution diverges logarithmically, the divergence being cut at the magnon gap ω_0 due to magnetic anisotropy, and for the 2D AFM case as ω_0^{-1}. The electron-magnon scattering processes yield $T^2\ln(T/\omega_0)$ and $T^2/\omega_0^{1/2}$ terms in $1/T_1$ for the 3D AFM and 2D FM cases, respectively. The two-magnon (''Raman'') contributions are investigated and demonstrated to be large in the 2D FM case. [S0163-1829(99)09141-9]

Nuclear magnetic resonance (NMR), which is one of most powerful tools for investigating various physical properties, has a number of peculiarities for magnetically ordered materials. Last time, a number of new classes of magnets have been studied by this method, e.g., heavy-fermion compounds, ferromagnetic films and monolayers, low-dimensional systems including copper-oxide perovskites, etc. Thus the problem of theoretical description of various NMR characteristics of magnets is topical again. This problem was already a subject of great interest since the 1950's and 1960's when the interaction of nuclear magnetic moments with spin waves in the localized-spin Heisenberg model was studied.[1,2] However, this model is inadequate to describe the most interesting systems mentioned above where the role of conduction electrons is essential in magnetic properties. Usually the data on the longitudinal nuclear magnetic relaxation rate $1/T_1$ are discussed within itinerant-electron models such as Hubbard model or phenomenological spin-fluctuation theories.[3–6] On the other hand, in a number of systems (e.g., in most rare-earth compounds which are also a subject of NMR investigations, see, e.g., Ref. 7) the s-$d(f)$ exchange model with well-separated localized and itinerant magnetic subsystems is more adequate. Magnetic properties in such a situation differ essentially from those in the paramagnon regime (see, e.g., discussion in Refs. 8,9). At the same time, the contributions to nuclear magnetic relaxation rate owing to electron-magnon interaction were not investigated in detail. In the present work we obtain the dependences of $1/T_1(T)$ and the linewidth $1/T_2(T)$ in the spin-wave region for three-dimensional (3D) and two-dimensional metallic magnets with well-defined local magnetic moments.

We start with the standard Hamiltonian of the hyperfine interaction[10] $H_{\rm hf}=\mathbf{hI}$, where $h_\alpha=A_{\alpha\beta}S_\beta$ and $\hat{A}$ is the hyperfine interaction matrix. This Hamiltonian contains the Fermi (contact) and dipole-dipole contributions, $A_{\alpha\beta}=A^F\delta_{\alpha\beta}+A^{dip}_{\alpha\beta}$. According to Ref. 10 we have

$$h^-=\left(A^F+\frac{1}{3}aF^{(0)}\right)S^-+aF^{(2)}S^++2aF^{(1)}S^z, \qquad (1)$$

$$h^z=\left(A^F-\frac{2}{3}aF^{(0)}\right)S^z+a(F^{(1)}S^++aF^{(1)*}S^-), \qquad (2)$$

where

$$F^{(0)}=\langle(1-3\cos^2\theta)/r^3\rangle,$$

$$F^{(1)}=\langle\sin\theta\cos\theta\exp(-i\phi)/r^3\rangle,$$

$$F^{(2)}=\langle\sin^2\theta\exp(-2i\phi)/r^3\rangle,\quad a=-\frac{3}{2}\gamma_e\gamma_n, \qquad (3)$$

$\langle\cdots\rangle$ is the average over the electron subsystem states, γ_e and γ_n are gyromagnetic ratios for electron and nuclear moments, respectively. In the case of the *local* cubic symmetry we have $F^{(a)}=0$. The Fermi hyperfine interaction is proportional to the electron density at the nucleus and therefore only s states participate in it, the contribution of core s states (which are polarized due to local magnetic moments) being much larger than of conduction electrons. It is just the consequence of considerably smaller localization area (and therefore higher density on nuclei) for the core states. It is obvious that magnetic f or d electrons dominate also in dipole interactions because of large spin polarization. Hence the direct interaction of nuclear spins with that of conduction electrons can be neglected in magnets with well-defined local magnetic moments. Nevertheless, conduction electrons do effect nuclear relaxation via their influence on the local-moment system; besides that, as we shall see below, such contributions possess large exchange enhancement factors.

Using the expressions for these contributions in terms of the Green's functions[11]

$$\frac{1}{T_1}=-\frac{T}{2\pi}\mathrm{Im}\sum_{\mathbf{q}}\langle\langle h^+_{\mathbf{q}}|h^-_{-\mathbf{q}}\rangle\rangle_{\omega_n}/\omega_n, \qquad (4)$$

$$\frac{1}{T_2}=\frac{1}{2T_1}-\frac{T}{2\pi}\lim_{\omega\to0}\mathrm{Im}\sum_{\mathbf{q}}\langle\langle h^z_{\mathbf{q}}|h^z_{-\mathbf{q}}\rangle\rangle_{\omega}/\omega \qquad (5)$$

($\omega_n=\langle h^z\rangle\ll T$ is the NMR frequency) we derive

$$\frac{1}{T_1}=\frac{T}{2}\left\{\left[\left(A^F+\frac{1}{3}aF^{(0)}\right)^2+a^2|F^{(2)}|^2\right]K^{+-}+2a\left(A^F+\frac{1}{3}aF^{(0)}\right)\mathrm{Re}\,F^{(2)}K^{++}+4a^2|F^{(1)}|^2K^{zz}\right\}, \tag{6}$$

$$\frac{1}{T_2}=\frac{1}{2T_1}+\frac{T}{2}\left\{\left(A^F-\frac{2}{3}aF^{(0)}\right)^2K^{zz}+a^2[2|F^{(1)}|^2K^{+-}+(F^{(1)})^2K^{++}+(F^{(1)*})^2K^{--}]\right\}, \tag{7}$$

$$K^{\alpha\beta}=-(1/\pi)\lim_{\omega\to 0}\mathrm{Im}\sum_{\mathbf{q}}\langle\langle S^{+}_{\mathbf{q}}|S^{-}_{-\mathbf{q}}\rangle\rangle_{\omega}/\omega. \tag{8}$$

We proceed with the s-$d(f)$ exchange model Hamiltonian

$$H=\sum_{\mathbf{k}\sigma}t_{\mathbf{k}}c^{\dagger}_{\mathbf{k}\sigma}c_{\mathbf{k}\sigma}-I\sum_{i\alpha\beta}\mathbf{S}_i\boldsymbol{\sigma}_{\alpha\beta}c^{\dagger}_{i\alpha}c_{i\beta}+\sum_{\mathbf{q}}J_{\mathbf{q}}\mathbf{S}_{-\mathbf{q}}\mathbf{S}_{\mathbf{q}}+H_a,$$

where $t_{\mathbf{k}}$ is the band energy, $\mathbf{S}_i$ and $\mathbf{S}_{\mathbf{q}}$ are spin-density operators and their Fourier transforms, σ are the Pauli matrices, H_a is the anisotropy Hamiltonian which results in occurrence of the gap ω_0 in the spin-wave spectrum. It should be noted that similar results may be reproduced for the localized-moment Hubbard magnets (see Refs. 8,12).

First we consider the ferromagnetic (FM) case. Then $K^{++}=0$ and the relaxation rates (6),(7) are the sums of transverse ($\propto K^{+-}$) and longitudinal ($\propto K^{zz}$) terms. Passing to the magnon representation we obtain

$$\langle\langle S^{+}_{\mathbf{q}}|S^{-}_{-\mathbf{q}}\rangle\rangle_{\omega}=2S/[\omega-\omega_{\mathbf{q}}+i\gamma_{\mathbf{q}}(\omega)], \tag{9}$$

where $\omega_{\mathbf{q}}=2S(J_{\mathbf{q}}-J_0)+\omega_0$ is the magnon frequency, $\gamma_{\mathbf{q}}(\omega)\propto\omega$ is the magnon damping. Then we have

$$K^{+-}=\frac{2S}{\pi\omega_n}\sum_{\mathbf{q}}\frac{\gamma_{\mathbf{q}}(\omega_n)}{\omega^2_{\mathbf{q}}} \tag{10}$$

(see Refs. 8,13). The damping in the denominator of Eq. (10) can be neglected for both localized-moment and itinerant-electron magnets [in the latter case the expression (9) corresponds to the RPA structure, see Ref. 8] due to smallness of ω_n. On the contrary, temperature dependences of magnetization, resistivity, etc., in weak itinerant magnets are just determined by the damping in the denominator, i.e., by paramagnon excitations rather than by spin waves.[4]

The damping owing to the one-magnon decay processes is given by the imaginary part of the RKKY-type polarization operator

$$\gamma^{(1)}_{\mathbf{q}}(\omega)\simeq 2\pi I^2S\omega\lambda_{\mathbf{q}},\quad \lambda_{\mathbf{q}}=\sum_{\mathbf{k}}\delta(t_{\mathbf{k}\uparrow})\delta(t_{\mathbf{k}-\mathbf{q}\downarrow}), \tag{11}$$

where $t_{\mathbf{k}\sigma}=t_{\mathbf{k}}-\sigma IS$, $n_{\mathbf{k}\sigma}=n(t_{\mathbf{k}\sigma})$ is the Fermi function. The linearity of spin fluctuation damping in $\omega_{\mathbf{q}}$ is a characteristic property of metals. According to Eq. (6), this leads to T-linear contributions to $1/T_1$ which is the Korringa law. Note that the simplest expression for the Korringa relaxation $1/T_1\simeq 1/T_2\simeq A^2\rho_{\uparrow}\rho_{\downarrow}T$, where A is an effective hyperfine interaction constant, ρ_{σ} are the partial densities of electron states at the Fermi level, is practically never applicable for magnetic metals [e.g., exchange enhancement factors can change even the order of magnitude of $1/T_1$ (Refs. 4,13)]. Accurate expression for the ''Korringa'' contribution in the case under consideration can be derived by the substitution of Eqs. (10) and (11) into Eq. (6).

The damping (11) has the threshold value of q, which is determined by the spin splitting $\Delta=2|I|S$, $q^*=\Delta/v_F$ (v_F is the electron velocity at the Fermi level). The quantity q^* determines a characteristic temperature and energy scale, $\omega^*=\omega(q^*)=\mathcal{D}(q^*)^2\sim(\Delta/v_F)^2T_C$ with $\mathcal{D}$ the spin-wave stiffness.

In the 2D case (which may be relevant, e.g., for layered magnets) we have

$$\lambda_{\mathbf{q}}=\theta(q-q^*)\times\begin{cases}(qv_F)^{-1}, & D=3,\\ \dfrac{1}{\pi}(q^2v_F^2-\Delta^2)^{-1/2}, & D=2.\end{cases} \tag{12}$$

After integration for the parabolic electron spectrum (q^* plays the role of the lower cutoff), the one-magnon damping contribution to Eq. (10) takes the form

$$\delta^{(1)}K^{+-}=\frac{\rho_{\uparrow}\rho_{\downarrow}}{\mathcal{D}^2m^2}\times\begin{cases}1/4, & D=3,\\ 1/(\pi q^*), & D=2,\end{cases} \tag{13}$$

$$\rho_{\sigma}=\frac{m\Omega_0}{2\pi}\times\begin{cases}k_{F\sigma}/\pi, & D=3,\\ 1, & D=2\end{cases} \tag{14}$$

with m the electron effective mass, Ω_0 the lattice cell volume (area). Thus in the 3D case the factor of I^2 is canceled, and the factor of I^{-1} occurs in the 2D case, so that we obtain a strongly enhanced T-linear Korringa-type term (remember that $\mathcal{D}\sim J\sim I^2\rho$ for the RKKY interaction). This means that the contribution of conduction electrons to T-linear relaxation rate via their interaction with localized spins is indeed much more important than the ''direct'' contribution: perturbation theory in the s-d exchange coupling parameter I turns out to be singular. Earlier such contributions (for the 3D case) were calculated by Weger[14] and Moriya[15] for iron-group metals. However, Moriya has concluded that for these materials they are not important in comparison with orbital current contributions. In the case under consideration (where magnetic subsystem is well separated from the conductivity electrons) the situation is different and the spin-wave contribution in $1/T_1$ is normally the most important.

The damping in a conducting ferromagnet owing to electron-magnon (two-magnon) scattering processes, calculated in Refs. 8 and 17, can be represented as

$$\frac{\gamma^{(2)}_{\mathbf{q}}(\omega)}{\omega}=\pi I^2\sum_{\mathbf{kp}\sigma}\left(\frac{t_{\mathbf{k}+\mathbf{q}}-t_{\mathbf{k}}}{t_{\mathbf{k}+\mathbf{q}}-t_{\mathbf{k}}+2\sigma IS}\right)^2(\omega_{\mathbf{p}}-\omega)\frac{\partial n_{\mathbf{k}\sigma}}{\partial t_{\mathbf{k}}}\frac{\partial N_{\mathbf{p}}}{\partial\omega_{\mathbf{p}}}\times\delta(t_{\mathbf{k}}-t_{\mathbf{k}-\mathbf{p}+\mathbf{q}}), \tag{15}$$

where $N_{\mathbf{p}}=N(\omega_{\mathbf{p}})$ is the Bose function. Substituting this into Eq. (10) and performing integration we obtain for $D=3$

$$\delta^{(2)}K^{+-}=\frac{\Omega_0 T^{1/2}}{128\pi^2 S m^2 \mathcal{D}^{7/2}}\sum_{\sigma}\rho_\sigma^2 \times\begin{cases}3\pi^{1/2}\zeta\left(\frac{3}{2}\right)T, & T\ll\omega^*,\\ 8M_3\omega^*, & T\gg\omega^*,\end{cases} \tag{16}$$

$$M_3=\int_0^\infty dx\left[\frac{1}{x^2}-\frac{x^2\exp x^2}{(\exp x^2-1)^2}\right]\simeq 0.65, \tag{17}$$

where $\zeta(z)$ is the Riemann function. The contribution (16) should play the dominant role in the half-metallic ferromagnets.[13,16] In addition to that, this contribution may modify considerably the temperature dependence of $1/T_1$ in ''usual'' ferromagnets, a crossover from $T^{5/2}$ to $T^{3/2}$ dependence of the correction taking place.

For $D=2$ at $T,\omega^*\gg\omega_0$ small magnon momenta of order of $(\omega_0/\mathcal{D})^{1/2}$ make the main contribution to Eq. (10). On using the high-temperature expression $N_{\mathbf{p}}=T/\omega_{\mathbf{p}}$ one gets

$$\delta^{(2)}K^{+-}=\frac{\Omega_0^3 k_F M_2}{8\pi^4 S\mathcal{D}^{5/2}\omega_0^{1/2}}T, \tag{18}$$

$$M_2=\int_0^\infty\frac{dx}{1+x^2}\int_0^{\pi/2}\frac{d\varphi\sin^2\varphi}{(\sin^2\varphi+x^2)^{3/2}}\simeq 1.23. \tag{19}$$

Thus in the 2D FM case, in contrast with 3D one, the relaxation rate $1/T_1$ is strongly dependent on the anisotropy gap. It is worthwhile to note an important difference between relaxation processes via phonons and via magnons. The main difference is due to the gap in magnon spectrum. Usually $\omega_0>\omega_n$ and therefore one-magnon processes contribute to the relaxation rate due to magnon damping only (see the discussion of the phonon-induced relaxation processes in Ref. 10). However, the mechanisms of magnon damping in magnetic dielectrics (magnon-magnon interactions) are different from those in magnetic metals and degenerate semiconductors.[17,9]

Consider now the second term in the transverse relaxation rate $1/T_2(T)$, Eq. (7), which is normally determined by K^{zz}, and the longitudinal contribution to the relaxation rate $1/T_1$ in Eq. (6), which is due to dipole-dipole interactions with the characteristic constant $\tilde{A}\sim a|F^{(1)}|$. The simplest calculation from the longitudinal Green's function for the localized-spin subsystem gives

$$K^{zz}=\sum_{\mathbf{qp}}(-\partial N_{\mathbf{p}}/\partial\omega_{\mathbf{p}})\delta(\omega_{\mathbf{q}}-\omega_{\mathbf{p}}). \tag{20}$$

The quantity (20) has been considered in Refs. 18,2 as a contribution to the NMR line width $1/T_2$. The integration in the 3D case gives the logarithmic singularity

$$K^{zz}=\frac{\Omega_0^2}{16\pi^4\mathcal{D}^3}T\ln\frac{T}{\omega_0}. \tag{21}$$

For $D=2$ this singular term is inversely proportional to the magnetic anisotropy parameter and very large:

$$K^{zz}=\left(\frac{\Omega_0}{4\pi\mathcal{D}}\right)^2 N(\omega_0)\simeq\left(\frac{\Omega_0}{4\pi\mathcal{D}}\right)^2\frac{T}{\omega_0},\quad T\gg\omega_0. \tag{22}$$

For small enough ω_0 and $\tilde{A}\sim A$ this contribution can dominate over the ''Korringa'' contribution (13) in $1/T_1$ at $T>\omega_0/|I\rho|$. The contributions to K^{zz} from the *s-d* interaction are not singular in ω_0 and practically never important.

Now we consider the two-sublattice antiferromagnetic (AFM) structure with the wave vector $\mathbf{Q}$. The expressions for the spin Green's functions are obtained in Refs. 9,19. We have

$$K^{+-}=-(2S/\pi)\lim_{\omega\to 0}\mathrm{Im}\sum_{\mathbf{q}}\omega^{-1}C_{\mathbf{q}\omega}/\omega_{\mathbf{q}}^2,\quad K^{++}=0 \tag{23}$$

with $\omega+C_{\mathbf{q}\omega}$ being the numerator of the spin-deviation Green's function. The one-magnon damping is given again by the imaginary part of the RKKY-type polarization operator. The intrasubband part (which is absent in the FM case) is finite at arbitrarily small q.[20] Similar to the FM case, the contributions of intersubband transitions (which correspond to small $|\mathbf{q}-\mathbf{Q}|$) are cut at the characteristic temperature and energy scale $\omega^*=\omega(q^*)=cq^*\sim(\Delta/v_F)T_N$, where c is the magnon velocity defined by $\omega_{\mathbf{p}}^2=\omega_{\mathbf{p}+\mathbf{Q}}^2=\omega_0^2+c^2p^2$. In the 3D case takes integration yields

$$\delta^{(1)}K^{+-}=\frac{S^2\Omega_0}{\pi^2c^2}\left(P_0\ln\frac{\omega_{\max}}{\omega_0}+P_{\mathbf{Q}}\ln\frac{\omega_{\max}}{\omega_0^*}\right), \tag{24}$$

$$P_{\mathbf{p}}=I^2\lim_{\mathbf{q}\to 0}|\mathbf{q}-\mathbf{p}|\sum_{\mathbf{k}}\delta(t_{\mathbf{k}})\delta(t_{\mathbf{k}-\mathbf{q}+\mathbf{p}}), \tag{25}$$

the second logarithm in the brackets of Eq. (24) contains the cutoff $\omega_0^*=[\omega_0^2+(\omega^*)^2]^{1/2}$. The ''enhancement'' factor in Eq. (24) is smaller than in the FM case because of linear dispersion law of magnons, but this contribution still dominates over the ''usual'' Korringa term. Besides that, a large logarithmic factor occurs (in the isotropic case, this is cut at ω_n only). Note that a similar logarithmic singularity in $1/T_1$ takes place for 3D itinerant-electron antiferromagnets.[3] It is interesting that the intersubband contribution does not lead here to enhancing the singularity (even in the nesting situation), unlike the results for the magnon damping, magnetic and transport properties.[21,9] The singularity becomes stronger in the 2D case where integration gives

$$\delta^{(1)}K^{+-}=\frac{S^2\Omega_0}{\pi c\omega_0}\left(\frac{\pi}{2}P_0+P_{\mathbf{Q}}\arctan\frac{\omega_0}{\omega^*}\right). \tag{26}$$

This fact may be important when treating experimental data on layered AFM metals.

The contribution owing to electron-magnon scattering processes is determined by the imaginary part of the function Φ from Ref. 9. After a little manipulation we obtain

$$\delta^{(2)}K^{+-}\simeq 2SL\sum_{\mathbf{p}\to 0,\mathbf{q}}\frac{1}{q\omega_{\mathbf{q}+\mathbf{p}}^2}\left(-\frac{\partial N_{\mathbf{p}}}{\partial\omega_{\mathbf{p}}}\right)[P_0+P_{\mathbf{Q}}\phi(q)],$$

where $L=2S(J_0-J_{\mathbf{Q}})$, $\phi(q<q^*)=0$, $\phi(q\gg q^*)=1$. The integration in the 3D case yields

$$\delta^{(2)}K^{+-}=\frac{SL\Omega_0^2}{8\pi^4c^4}[P_0 f(T,\omega_0)+P_{\mathbf{Q}} f(T,\omega_0^*)], \quad (27)$$

$$f(T,\omega_0)=\int_{\omega_0}^{\infty} d\omega\,\omega\left(-\frac{\partial N(\omega)}{\partial\omega}\right)\ln\frac{\omega_{\max}}{\omega}$$

$$\simeq T\ln\frac{T}{\omega_0}\left(\ln\frac{\omega_{\max}}{\omega_0}-\frac{1}{2}\ln\frac{T}{\omega_0}\right), \quad T\gg\omega_0. \quad (28)$$

Thus we have $1/T_1\propto T^2\ln T$. In the 2D case we derive

$$\delta^{(2)}K^{+-}\simeq T\frac{SL\Omega_0^2}{4\pi^4c^4}\left(P_0\,\ln^2\frac{T}{\omega_0}+P_{\mathbf{Q}}\,\ln^2\frac{T}{\omega_0^*}\right), \quad (29)$$

so that the singularity is not enhanced in comparison with the 3D case.

The contributions owing to longitudinal fluctuations will be estimated for the localized subsystem only. We obtain

$$K^{zz}\simeq\sum_{\mathbf{pq}}\frac{L^2}{2\omega_{\mathbf{p}}^2}\left(-\frac{\partial N_{\mathbf{p}}}{\partial\omega_{\mathbf{p}}}\right)\delta(\omega_{\mathbf{q}}-\omega_{\mathbf{p}}). \quad (30)$$

The corresponding contribution to $1/T_2$ was considered in Ref. 1. The term in the longitudinal relaxation rate determined by Eq. (30) is estimated as

$$\delta^{(z)}(1/T_1)\propto\tilde{A}^2\times\begin{cases}T^3/J^4, & D=3,\\ T^2/J^3, & D=2.\end{cases} \quad (31)$$

Provided that the dipole-dipole contributions in Eq. (6) are considerable ($\tilde{A}\sim A$), this term can dominate over the ''Korringa'' term (24) of order of $A^2I^2\rho^2T\ln|J/\omega_0|/J^2$ at $T/|J|>|I\rho|\ln^{1/2}|J/\omega_0|$ only. Note that this two-magnon contribution is similar to the two-phonon (Raman) contribution in the spin-lattice relaxation. The existence of the gap ω_0 is not important here (at least if it is sufficiently small), but the matrix elements of interaction of nuclear spins with magnons are singular, unlike those for acoustic phonons ($|M_{\mathbf{q}\to 0}|^2\sim 1/q$ instead of q). Therefore we have a T^3 law instead of T^7 one for the phonon scattering.[10]

To conclude, in most cases the main contribution to $1/T_1$ in the localized-moment magnetic metals is of Korringa type, but its physical origin is more complicated than in the paramagnetic case. Formally, it results from the interaction of nuclear magnetic moments with the *localized* electronic subsystem with taking into account the ''Stoner'' (Landau) damping of spin waves via conduction electrons. This contribution is greatly enhanced in comparison with the standard Korringa term by inverse powers of exchange interaction [$s-d(f)$ parameter], especially in ferromagnets. In the AFM case we have $1/T_1\propto\ln\omega_0$ ($D=3$), $1/T_1\propto\omega_0^{-1}$ ($D=2$). Thus the ''Korringa'' relaxation rate in the magnetic metals should be much larger than in paramagnetic ones where the relaxation is determined by direct interaction with conduction electrons (such a term is also present in the magnetically ordered state, but is much smaller than the contribution discussed). More complicated electron-magnon scattering processes may result in considerable deviations of the temperature dependence of $1/T_1$ from the linear Korringa law.

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