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Spin-wave contributions to nuclear magnetic relaxation in magnetic metals

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Abstract. The longitudinal and transverse nuclear magnetic relaxation rates $1/T_1(T)$ and $1/T_2(T)$ are calculated for three- and two-dimensional (3D and 2D) metallic ferro- and antiferromagnets (FM and 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. Peculiarities of the isotropic 2D limit (where the correlation length is very large) are analyzed.

PACS. 76.60.-k Nuclear magnetic resonance and relaxation – 75.30.Ds Spin waves

1 Introduction

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 [1], ferromagnetic films and monolayers [2], low-dimensional systems including copper-oxide perovskites [3], 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 50-60s when the interaction of nuclear magnetic moments with spin waves in localized-spin Heisenberg model was studied [4,5]. 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 $1/T_1$ (this NMR characteristic is probably most convenient to compare experimental results with theoretical predictions) are discussed within itinerant-electron models such as Hubbard model or phenomenological spin-fluctuation theories. Ueda and Moriya [6,7] calculated the dependences $1/T_1(T)$ for weak itinerant magnets, especial attention being paid to the paramagnetic region, and obtained strong temperature effects. Later this approach was extended to the two-dimensional case and extensively developed in connection with high-T_c superconductors and related compounds [8,9]. 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.*, Refs. [10]) 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. [11,12]). At the same time, the contributions to nuclear magnetic relaxation rate owing to electron-magnon interaction are 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- and two-dimensional (3D and 2D) metallic magnets with well-defined local magnetic moments. In Section 2 we discuss the general formalism and physical picture of hyperfine interactions. In Sections 3 and 4 we calculate various contributions to the relaxation rates in metallic ferro- and antiferromagnets. In Section 5 we analyze the isotropic 2D case where at finite temperatures the long-range order is absent, but the correlation length is very large.

2 Hyperfine interactions

We start from the standard Hamiltonian of the hyperfine interaction [13]

$$H_{\rm hf} = \mathbf{hI}, \;\; h_\alpha = A_{\alpha\beta}S_\beta \tag{1}$$

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$\widehat{A}$ being the hyperfine interaction matrix, which contains the Fermi (contact) and dipole-dipole contributions,

$$A_{\alpha\beta} = A^{\rm F}\delta_{\alpha\beta} + A^{\rm dip}_{\alpha\beta}. \quad (2)$$

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.

The dipole contribution to $H_{\rm hf}$ can be represented as [13]

$$H^{\rm dip}_{\rm hf} = \frac{a}{2}\Bigg(\left[\frac{1}{6}(I^+S^- + I^-S^+) - \frac{1}{3}I^zS^z\right]F^{(0)} + I^+S^+F^{(2)} + 2(I^zS^+ + I^+S^z)F^{(1)}\Bigg) + \text{h.c.} \quad (3)$$

where

$$F^{(0)} = \langle(1-3\cos^2\theta)/r^3\rangle,$$
$$F^{(1)} = \langle\sin\theta\cos\theta\exp(-\mathrm{i}\phi)/r^3\rangle,$$
$$F^{(2)} = \langle\sin^2\theta\exp(-2\mathrm{i}\phi)/r^3\rangle, \quad a = -\frac{3}{2}\gamma_{\rm e}\gamma_{\rm n}. \quad (4)$$

Here $\langle ...\rangle$ is the average over the electron subsystem states, $\gamma_{\rm e}$ and $\gamma_{\rm n}$ are gyromagnetic ratios for electron and nuclear moments, respectively. In the case of the *local* cubic symmetry we have $F^{(a)} = 0$. 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. The investigation of these effects is one of the main aims of this work.

A general way to consider all these contributions is using the Green's function method which leads to the following expression for the longitudinal nuclear magnetic relaxation rate [14]

$$\frac{1}{T_1} = -\frac{T}{2\pi}\mathrm{Im}\sum_{\mathbf{q}}\langle\langle h^+_{\mathbf{q}}|h^-_{-\mathbf{q}}\rangle\rangle_{\omega_{\rm n}}/\omega_{\rm n}, \quad (5)$$

$$\frac{1}{T_2} = \frac{1}{2T_1} - \frac{T}{2\pi}\lim_{\omega\to 0}\mathrm{Im}\sum_{\mathbf{q}}\langle\langle h^z_{\mathbf{q}}|h^z_{-\mathbf{q}}\rangle\rangle_{\omega}/\omega \quad (6)$$

($\omega_{\rm n} = \langle h^z\rangle \ll T$ is the NMR frequency). As follows from (3),

$$h^- = (A^{\rm F} + \frac{1}{3}aF^{(0)})S^- + aF^{(2)}S^+ + 2aF^{(1)}S^z, \quad (7)$$

$$h^z = (A^{\rm F} - \frac{2}{3}aF^{(0)})S^z + a(F^{(1)}S^+ + aF^{(1)*}S^-). \quad (8)$$

Then we derive

$$\frac{1}{T_1} = \frac{T}{2}\Bigg(\left[(A^{\rm F} + \frac{1}{3}aF^{(0)})^2 + a^2|F^{(2)}|^2\right]K^{+-} + 2a(A^{\rm F} + \frac{1}{3}aF^{(0)})\mathrm{Re}F^{(2)}K^{++} + 4a^2|F^{(1)}|^2K^{zz}\Bigg) \quad (9)$$

$$\frac{1}{T_2} = \frac{1}{2T_1} + \frac{T}{2}\Bigg((A^{\rm F} - \frac{2}{3}aF^{(0)})^2K^{zz} + a^2[2|F^{(1)}|^2K^{+-} + (F^{(1)})^2K^{++} + (F^{(1)*})^2K^{--}]\Bigg) \quad (10)$$

where the quantities $K^{\alpha\beta}$ are defined by

$$K^{\alpha\beta} = -\frac{1}{\pi}\lim_{\omega\to 0}\mathrm{Im}\sum_{\mathbf{q}}\langle\langle S^\alpha_{\mathbf{q}}|S^\beta_{-\mathbf{q}}\rangle\rangle_\omega/\omega. \quad (11)$$

As we shall see below in Section 5, $\omega_{\rm n} \neq 0$ which enters (9) (but not the second term of (10)) may become important in the case of very small magnetic anisotropy.

Formula (5) has a rather general character. On the other hand, the problem of calculating the NMR linewidth is much more complicated. The Moriya formula (6) is in fact applicable only in the case where the line has the Lorentz form (*i.e.*, characteristic frequency of hyperfine field fluctuations is large in comparison with their amplitude) [13]. In insulating crystals the latter condition is usually violated, the lineform being close to Gaussian with the width determined by dipole interactions of nuclear spins. At the same time, in metals the Korringa relaxation described by the formula (6) usually dominates, so that we will use this. A peculiar case is provided by conducting systems which are on the borderline of ferro- or antiferromagnetic instability (*i.e.* with large correlation length ξ), *e.g.*, copper-oxide superconductors [15]. Under this condition, the anisotropic Ruderman-Kittel interaction between nuclear spins turns out to be greatly enhanced and dominates over the dipole interaction. The lineform turns out to be Gaussian with the width being estimated as

$$\frac{1}{T_2} \propto A^2\sum_{\mathbf{q}}\chi^2(\mathbf{q},\omega=0) \quad (12)$$

where $\chi(\mathbf{q},\omega)$ is the dynamical spin susceptibility of electron system. We will use this result in Section 5 when discussing 2D systems which do possess large correlation length.

3 Ferromagnetic metals

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

$$H = \sum_{\mathbf{k}\sigma} t_{\mathbf{k}}c^\dagger_{\mathbf{k}\sigma}c_{\mathbf{k}\sigma} - I\sum_{i\alpha\beta}\mathbf{S}_i\boldsymbol{\sigma}_{\alpha\beta}c^\dagger_{i\alpha}c_{i\beta} + \sum_{\mathbf{q}}J_{\mathbf{q}}\mathbf{S}_{-\mathbf{q}}\mathbf{S}_{\mathbf{q}} + H_{\rm a} \quad (13)$$

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, $H_{\rm a}$ is the anisotropy Hamiltonian which results in occurrence of the gap ω_0 in the spin-wave spectrum. For convenience we include explicitly in the Hamiltonian the Heisenberg exchange interaction with the parameters $J_{\mathbf{q}}$, although really this may be, *e.g.*, the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction. It should be noted that similar results may be reproduced for the localized-moment Hubbard magnets (*cf.* [11,16]).

First we consider the ferromagnetic (FM) case. Then $K^{++} = 0$ and the relaxation rates (9, 10) are the sums of transverse ($\propto K^{+-}$) and longitudinal ($\propto K^{zz}$) terms. Passing to the magnon representation we obtain

$$\langle\langle S^+_{\mathbf{q}} | S^-_{-\mathbf{q}} \rangle\rangle_\omega = 2S/[\omega - \omega_{\mathbf{q}} + {\rm i}\gamma_{\mathbf{q}}(\omega)] \tag{14}$$

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

$$K^{+-} = 2S \sum_{\mathbf{q}} \frac{\gamma_{\mathbf{q}}(\omega_{\rm n})}{\pi \omega_{\rm n} \omega^2_{\mathbf{q}}} \tag{15}$$

(*cf.* Refs. [11,17]). The damping in the denominator of (15) can be neglected for both localized-moment and itinerant-electron magnets (in the latter case the expression (14) corresponds to the RPA structure, see Ref. [11]) due to smallness of $\omega_{\rm n}$. 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 [7].

The damping owing to the one-magnon decay processes is given by the well-known expression

$$\begin{aligned} \gamma^{(1)}_{\mathbf{q}}(\omega) &= -2\pi I^2 S \sum_{\mathbf{k}} (n_{\mathbf{k}\uparrow} - n_{\mathbf{k}-\mathbf{q}\downarrow}) \\ &\quad \times \delta(\omega + t_{\mathbf{k}\uparrow} - t_{\mathbf{k}-\mathbf{q}\downarrow}) \\ &\simeq 2\pi I^2 S \omega \lambda_{\mathbf{q}} \end{aligned} \tag{16}$$

where $t_{\mathbf{k}\sigma} = t_{\mathbf{k}} - \sigma IS$, $t_{\mathbf{k}}$ is referred to the Fermi level, $n_{\mathbf{k}\sigma} = n(t_{\mathbf{k}\sigma})$ is the Fermi function,

$$\lambda_{\mathbf{q}} = \sum_{\mathbf{k}} \delta(t_{\mathbf{k}\uparrow}) \delta(t_{\mathbf{k}-\mathbf{q}\downarrow}). \tag{17}$$

The linearity of spin fluctuation damping in ω is the characteristic property of metals. According to (9) this leads to T-linear contributions to $1/T_1$ which is the Korringa law [18]. Note that the simplest expression for the Korringa relaxation

$$1/T_1 \simeq 1/T_2 \simeq A^2 \rho_\uparrow \rho_\downarrow T, \tag{18}$$

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 $1/T_1$ [7,17]). Accurate expression for the "Korringa" contribution in the case under consideration can be derived by the substitution (15) and (16) into (9).

The damping (16) has the threshold value of q, which is determined by the spin splitting $\Delta = 2|I|S$, $q^* = \Delta/v_{\rm F}$ ($v_{\rm F}$ is the electron velocity at the Fermi level). The quantity q^* determines a characteristic temperature and energy scale

$$\omega^* = \omega(q^*) = \mathcal{D}(q^*)^2 \sim (\Delta/v_{\rm F})^2 T_{\rm C} \tag{19}$$

with $\mathcal{D}$ the spin-wave stiffness.

Besides 3D magnets, consideration of the 2D case is of interest (this may be relevant, *e.g.*, for layered magnets and ferromagnetic films; for more details see Sect. 5). We have

$$\lambda_{\mathbf{q}} = \theta(q - q^*) \times \begin{cases} (q v_{\rm F})^{-1}, & {\rm D}{=}\,3 \\ \frac{1}{\pi}(q^2 v_F^2 - \Delta^2)^{-1/2}, & {\rm D}{=}\,2. \end{cases} \tag{20}$$

After integration for the parabolic electron spectrum (q^* plays the role of the lower cutoff), the one-magnon damping contribution to (15) takes the form

$$\delta^{(1)} K^{+-} = \frac{\rho_\uparrow \rho_\downarrow}{\mathcal{D}^2 m^2} \times \begin{cases} 1/4, & {\rm D}{=}\,3 \\ 1/(\pi q^*), & {\rm D}{=}\,2 \end{cases} \tag{21}$$

with

$$\rho_\sigma = \frac{m\Omega_0}{2\pi} \times \begin{cases} k_{{\rm F}\sigma}/\pi, & {\rm D}{=}\,3 \\ 1, & {\rm D}{=}\,2 \end{cases} \tag{22}$$

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 and 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 (18): 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 [19] and Moriya [20] 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 one-magnon decay contribution (21) is absent for so-called half-metallic ferromagnets, *e.g.*, some Heusler alloys, where electron states with one spin projection only are presented at the Fermi surface [17,21]. In such a situation we have to consider two-magnon scattering processes. In this connection, 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 $\omega_0 > \omega_{\rm n}$ and therefore one-magnon processes contribute to the relaxation rate due

to magnon damping only (*cf.* discussion of the phonon-induced relaxation processes in Ref. [13]). However, the mechanisms of magnon damping in magnetic dielectrics (magnon-magnon interactions) are different from those in magnetic metals and degenerate semiconductors [12,22].

The damping in a conducting ferromagnet owing to electron-magnon (two-magnon) scattering processes is calculated in references [11,22] and has the form

$$\frac{\gamma_{\mathbf{q}}^{(2)}(\omega)}{\omega} = \pi I^2 \sum_{\mathbf{kp}\sigma} \left(\frac{t_{\mathbf{k+q}} - t_{\mathbf{k}}}{t_{\mathbf{k+q}} - t_{\mathbf{k}} + 2\sigma IS} \right)^2 \times (\omega_{\mathbf{p}} - \omega) \frac{\partial n_{\mathbf{k}\sigma}}{\partial t_{\mathbf{k}}} \frac{\partial N_{\mathbf{p}}}{\partial \omega_{\mathbf{p}}} \delta(t_{\mathbf{k}} - t_{\mathbf{k-p+q}}) \quad (23)$$

where $N_{\mathbf{p}} = N(\omega_{\mathbf{p}})$ is the Bose function. Substituting this into (15) and performing integration we obtain for D= 3

$$\delta^{(2)} K^{+-} = \frac{\Omega_0 T^{1/2}}{128\pi^2 S m^2 \mathcal{D}^{7/2}} \sum_{\sigma} \rho_{\sigma}^2 \times \begin{cases} 3\pi^{1/2} \zeta(\frac{3}{2}) T, & T \ll \omega^* \\ 8 M_3 \omega^*, & T \gg \omega^* \end{cases} \quad (24)$$

where $\zeta(z)$ is the Riemann function,

$$M_3 = \int_0^{\infty} \mathrm{d}x \left[\frac{1}{x^2} - \frac{x^2 \exp x^2}{(\exp x^2 - 1)^2} \right] \simeq 0.65. \quad (25)$$

The contribution (24) should play the dominant role in the half-metallic ferromagnets [17]. Besides 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 (15). To calculate the integral one can use the high-temperature expression for $N_{\mathbf{p}} = T/\omega_{\mathbf{p}}$. As a result, one gets

$$\delta^{(2)} K^{+-} = \frac{\Omega_0^3 k_{\mathrm{F}} M_2}{8\pi^4 S \mathcal{D}^{5/2} \omega_0^{1/2}} T \quad (26)$$

with

$$M_2 = \int_0^{\infty} \frac{\mathrm{d}x}{1+x^2} \int_0^{\pi/2} \frac{\mathrm{d}\varphi \sin^2 \varphi}{\left(\sin^2 \varphi + x^2\right)^{3/2}} = \int_0^{\infty} \mathrm{d}y \left[1 + \frac{y^2}{\sqrt{1+y^2}} \ln \frac{\sqrt{1+y^2} - 1}{y} \right] \simeq 1.23. \quad (27)$$

Thus in the 2D FM case, in contrast with 3D one, the relaxation rate $1/T_1$ is strongly dependent on the anisotropy gap.

Consider now the second term in the transverse relaxation rate $1/T_2(T)$ (10), which is normally determined by K^{zz}, and the longitudinal contribution to relaxation rate $1/T_1$ in (9), which is due to dipole-dipole interactions with the characteristic constant $\widetilde{A} \sim a|F^{(1)}|$. The simplest calculation from the longitudinal Green's function for the localized-spin subsystem gives

$$\langle\langle S_{\mathbf{q}}^z | S_{-\mathbf{q}}^z \rangle\rangle_{\omega} = \sum_{\mathbf{p}} \frac{N_{\mathbf{p}} - N_{\mathbf{p-q}}}{\omega - \omega_{\mathbf{p-q}} + \omega_{\mathbf{p}}}, \quad (28)$$

$$K^{zz} = \sum_{\mathbf{qp}} \left(-\frac{\partial N_{\mathbf{p}}}{\partial \omega_{\mathbf{p}}} \right) \delta(\omega_{\mathbf{q}} - \omega_{\mathbf{p}}). \quad (29)$$

The quantity (29) has been considered in references [5,23] as a contribution to the NMR line width $1/T_2$. The integration in the 3D case gives the logarithmic singularity

$$K^{zz} = \frac{\Omega_0^2}{16\pi^4 \mathcal{D}^3} T \ln \frac{T}{\omega_0} \cdot \quad (30)$$

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

$$K^{zz} = \left(\frac{\Omega_0}{4\pi\mathcal{D}} \right)^2 N(\omega_0) \simeq \left(\frac{\Omega_0}{4\pi\mathcal{D}} \right)^2 \frac{T}{\omega_0}, \quad T \gg \omega_0. \quad (31)$$

For small enough ω_0 and $\widetilde{A} \sim A$ this contribution can dominate over the "Korringa" contribution (21) in $1/T_1$ at $T > \omega_0/|I\rho|$. The leading contribution to K^{zz} from the *s-d* interaction is determined by

$$\delta \langle\langle S_{\mathbf{q}}^z | S_{-\mathbf{q}}^z \rangle\rangle_{\omega} = 2I^2 S \sum_{\mathbf{kp}\sigma} \frac{1}{(\sigma\omega + \omega_{\mathbf{p-q}} - \omega_{\mathbf{q}})^2} \times \frac{n_{\mathbf{k}\downarrow}(1 - n_{\mathbf{k+p-q}\uparrow}) + N_{\mathbf{p}}(n_{\mathbf{k}\downarrow} - n_{\mathbf{k+p-q}\uparrow})}{t_{\mathbf{k}\downarrow} - t_{\mathbf{k+p-q}\uparrow} + \sigma\omega - \omega_{\mathbf{p}}} \cdot \quad (32)$$

However, it is not singular in ω_0 and practically never important.

4 Antiferromagnetic metals

Now we consider the spiral antiferromagnetic (AFM) structure along the x-axis with the wavevector $\mathbf{Q}$

$$\langle S_i^z \rangle = S \cos \mathbf{QR}_i, \quad \langle S_i^y \rangle = S \sin \mathbf{QR}_i, \quad \langle S_i^x \rangle = 0.$$

We introduce the local coordinate system

$$S_i^z = \hat{S}_i^z \cos \mathbf{QR}_i - \hat{S}_i^y \sin \mathbf{QR}_i,$$
$$S_i^y = \hat{S}_i^y \cos \mathbf{QR}_i + \hat{S}_i^z \sin \mathbf{QR}_i, \quad S_i^x = \hat{S}_i^x.$$

Further we pass from spin operators $\hat{\mathbf{S}}_i$ to the spin deviation operators $b_i^{\dagger}, b_i$ and, by the canonical transformation $b_{\mathbf{q}}^{\dagger} = u_{\mathbf{q}} \beta_{\mathbf{q}}^{\dagger} - v_{\mathbf{q}} \beta_{-\mathbf{q}}$, to the magnon operators. Hereafter we consider for simplicity two-sublattice AFM ordering ($2\mathbf{Q}$ is equal to a reciprocal lattice vector, so that $\cos^2 \mathbf{QR}_i = 1$, $\sin^2 \mathbf{QR}_i = 0$).

Calculating the Green's functions to second order in I (to second order in the formal quasiclassical parameter $1/2S$, *cf.* Refs. [12,24]) we derive

$$\langle\langle b_{\mathbf{q}} | b_{\mathbf{q}}^{\dagger} \rangle\rangle_{\omega} = \frac{\omega + C_{\mathbf{q}-\omega}}{(\omega - C_{\mathbf{q}\omega})(\omega + C_{\mathbf{q}-\omega}) + D_{\mathbf{q}\omega}^2} \quad (33)$$

$$\langle\langle b_{-\mathbf{q}}^{\dagger} | b_{\mathbf{q}}^{\dagger} \rangle\rangle_{\omega} = \frac{D_{\mathbf{q}\omega}}{(\omega - C_{\mathbf{q}\omega})(\omega + C_{\mathbf{q}-\omega}) + D_{\mathbf{q}\omega}^2} \quad (34)$$

with

$$C_{\mathbf{q}\omega} = S(J^{\rm tot}_{\mathbf{Q}+\mathbf{q},\omega} + J^{\rm tot}_{\mathbf{q}\omega} - 2J^{\rm tot}_{\mathbf{Q}0}) + \sum_{\mathbf{p}}[C_{\mathbf{p}}\Phi_{\mathbf{pq}\omega} - (C_{\mathbf{p}} - D_{\mathbf{p}})\Phi_{\mathbf{p}00} + \phi^{+}_{\mathbf{pq}\omega} + \phi^{-}_{\mathbf{pq}\omega}] + g_{\mathbf{q}} \quad (35)$$

$$D_{\mathbf{q}\omega} = D_{\mathbf{q}-\omega} = S(J^{\rm tot}_{\mathbf{q}\omega} - J^{\rm tot}_{\mathbf{Q}+\mathbf{q},\omega}) + \sum_{\mathbf{p}} D_{\mathbf{p}}\Phi_{\mathbf{pq}\omega} + h_{\mathbf{q}}.$$

The s-d exchange contributions of the first order in $1/2S$ correspond to the RKKY approximation

$$J^{\rm tot}_{\mathbf{q}\omega} = J_{\mathbf{q}} + I^2 \sum_{\mathbf{k}} \frac{n_{\mathbf{k}} - n_{\mathbf{k}-\mathbf{q}}}{\omega + t_{\mathbf{k}} - t_{\mathbf{k}-\mathbf{q}}} \quad (36)$$

($n_{\mathbf{k}} = n(t_{\mathbf{k}})$ is the Fermi function), the second summand in (36) being the ω-dependent RKKY indirect exchange interaction. The function Φ, which determines the second-order corrections, is given by

$$\Phi_{\mathbf{pq}\omega} = (\phi^{+}_{\mathbf{pq}\omega} - \phi^{-}_{\mathbf{pq}\omega})/\omega_{\mathbf{p}}, \quad (37)$$

$$\phi^{\pm}_{\mathbf{pq}\omega} = I^2 \sum_{\mathbf{k}} \frac{n_{\mathbf{k}}(1 - n_{\mathbf{k}+\mathbf{p}-\mathbf{q}}) + N(\pm\omega_{\mathbf{p}})(n_{\mathbf{k}} - n_{\mathbf{k}+\mathbf{p}-\mathbf{q}})}{\omega + t_{\mathbf{k}} - t_{\mathbf{k}+\mathbf{p}-\mathbf{q}} \mp \omega_{\mathbf{p}}}$$

where

$$\omega_{\mathbf{p}} = (C^2_{\mathbf{p}} - D^2_{\mathbf{p}})^{1/2} = [4S^2(J_{\mathbf{p}} - J_{\mathbf{Q}})(J_{\mathbf{Q}+\mathbf{p}} - J_{\mathbf{Q}}) + \omega_0^2]^{1/2}$$

is the magnon frequency to zeroth order in I and $1/2S$. The ω-independent corrections $g_{\mathbf{q}}, h_{\mathbf{q}}$ that describe the "direct" magnon-magnon interaction are written down in references [12,24].

Now we consider the effects of electron-magnon interaction. The intrasubband one-magnon damping (which is absent in the FM case) is finite at arbitrarily small q [25]. 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

$$\omega^* = \omega(q^*) = cq^* \sim (\Delta/v_{\rm F})T_N. \quad (38)$$

We have

$$K^{+-} = -\frac{2S}{\pi} \lim_{\omega\to 0} {\rm Im} \sum_{\mathbf{q}} \omega^{-1} C_{\mathbf{q}\omega}/\omega^2_{\mathbf{q}}, \quad (39)$$

and the term with K^{++} in (9) vanishes due to the property $D_{\mathbf{q}\omega} = -D_{\mathbf{q}+\mathbf{Q}\omega}$. The one-magnon contribution owing to the imaginary part of (36) in the 3D case takes after integration the form

$$\delta^{(1)}K^{+-} = \frac{S^2\Omega_0}{\pi^2 c^2}\left(P_0 \ln\frac{\omega_{\max}}{\omega_0} + P_{\mathbf{Q}} \ln\frac{\omega_{\max}}{\omega_0^*}\right). \quad (40)$$

Here c is the magnon velocity defined by $\omega^2_{\mathbf{p}} = \omega^2_{\mathbf{p}+\mathbf{Q}} = \omega_0^2 + c^2p^2$,

$$P_{\mathbf{p}} = I^2 \lim_{\mathbf{q}\to 0} |\mathbf{q} - \mathbf{p}| \sum_{\mathbf{k}} \delta(t_{\mathbf{k}})\delta(t_{\mathbf{k}-\mathbf{q}+\mathbf{p}}), \quad (41)$$

(the quantity P_0 depends, generally speaking, on the direction of the vector $\mathbf{q}$, see Refs. [7,26,27]), the second logarithm in the brackets of (40) contains the cutoff

$$\omega_0^* = \sqrt{\omega_0^2 + (\omega^*)^2}.$$

The "enhancement" factor in (40) is smaller than in the FM case because of the linear dispersion law of magnons, but this contribution still dominates over the "usual" Korringa term (18). Besides that, a large logarithmic factor occurs (in the isotropic case, this is cut at $\omega_{\rm n}$ only). Note that a similar logarithmic singularity in $1/T_1$ takes place for 3D itinerant-electron antiferromagnets [6]. It is interesting that the intersubband contribution does not lead here to enhancing the singularity, unlike the situation for the magnon damping, magnetic and transport properties [12,28]. Under the "nesting" conditions ($t_{\mathbf{k}+\mathbf{Q}} \simeq -t_{\mathbf{k}}$ in a large part of the Fermi surface) the singularity is not enhanced as well.

The singularity becomes stronger in the 2D case where integration gives

$$\delta^{(1)}K^{+-} = \frac{S^2\Omega_0}{\pi c\omega_0}\left(\frac{\pi}{2}P_0 + P_{\mathbf{Q}} \arctan\frac{\omega_0}{\omega^*}\right). \quad (42)$$

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 (37). After a little manipulation we obtain

$$\delta^{(2)}K^{+-} \simeq 2SL \sum_{\mathbf{p}\to 0,\mathbf{q}} \frac{1}{q\omega^2_{\mathbf{q}+\mathbf{p}}}\left(-\frac{\partial N_{\mathbf{p}}}{\partial\omega_{\mathbf{p}}}\right)[P_0 + P_{\mathbf{Q}}\widetilde{\phi}(q)] \quad (43)$$

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

$$\delta^{(2)}K^{+-} = \frac{SL\Omega_0^2}{8\pi^4 c^4}\left[P_0 f(T,\omega_0) + P_{\mathbf{Q}} f(T,\omega_0^*)\right] \quad (44)$$

where

$$f(T,\omega_0) = \int_{\omega_0}^{\infty} {\rm d}\omega\omega\left(-\frac{\partial N(\omega)}{\partial\omega}\right)\ln\frac{\omega_{\max}}{\omega} \simeq T\ln\frac{T}{\omega_0}\left(\ln\frac{\omega_{\max}}{\omega_0} - \frac{1}{2}\ln\frac{T}{\omega_0}\right), \quad T \gg \omega_0. \quad (45)$$

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

$$\delta^{(2)}K^{+-} \simeq T\frac{SL\Omega_0^2}{4\pi^4 c^4}\left(P_0 \ln^2\frac{T}{\omega_0} + P_{\mathbf{Q}} \ln^2\frac{T}{\omega_0^*}\right), \quad (46)$$

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

$$K^{zz} \simeq \sum_{\mathbf{pq}} \frac{L^2}{2\omega^2_{\mathbf{p}}}\left(-\frac{\partial N_{\mathbf{p}}}{\partial\omega_{\mathbf{p}}}\right)\delta(\omega_{\mathbf{q}} - \omega_{\mathbf{p}}). \quad (47)$$

The corresponding contribution to $1/T_2$ was considered in reference [4]. The term in the longitudinal relaxation rate determined by (47) is estimated as

$$\delta^{(z)}(1/T_1) \propto \widetilde{A}^2 \times \begin{cases} T^3/J^4, & \mathrm{D}=3 \\ T^2/J^3, & \mathrm{D}=2. \end{cases} \quad (48)$$

Provided that the dipole-dipole contributions in (9) are considerable ($\widetilde{A} \sim A$), this term can dominate over the "Korringa" term (40) 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 [13].

5 Isotropic 2D case and NMR in layered and frustrated magnets

Now we investigate the case of layered magnets, in particular, the isotropic 2D limit. A detailed treatment of the spin correlation functions and corresponding spin-fluctuation contributions to $1/T_1$ in the isotropic 2D Heisenberg antiferromagnets with $\omega_\mathrm{n} \to 0$ was performed in reference [29]. Here we calculate also corrections owing to electron-magnon interaction.

The magnetic ordering temperature is determined by magnetic anisotropy or interlayer coupling,

$$T_\mathrm{M} \sim |J|S^2/\ln(|J|S^2/\max\{\omega_0, |J'|\}) \quad (49)$$

(for more details see, *e.g.*, Refs. [30]). Despite the absence of the long-range ordering (LRO) at finite temperatures, spin-wave description holds even in the pure 2D isotropic case in the broad temperature region up to $T \sim |J|S$ (*i.e.*, $T_\mathrm{M} \to |J|S^2$) owing to strong short-range order (SRO). In a more general case of finite ω_0 and J', this description holds at $T \gg T_\mathrm{M}$. A possibility to describe LRO without introducing anomalous averages (like sublattice magnetization) in terms of singularities of the spin correlation function was demonstrated in reference [31]. Such an approach enables one to obtain an unified description of ordered and disordered phases. In the pure 2D case the magnetization (or sublattice magnetization for the AFM case) $\overline{S}$ is replaced in both magnetic and electronic properties by the square root of the Ornstein-Cernike peak (see, *e.g.*, [32]). The gap in the effective spin-wave spectrum appears at finite temperatures, which is determined by the inverse correlation length. The correlation length in the situation under consideration is estimated as [33]

$$\xi \propto \exp\left(\pi|J|S^2/T\right). \quad (50)$$

As shows the two-loop scaling theory [34], the preexponential factor is temperature independent; quantum effects can renormalize the exchange parameter J [30].

To describe formally NMR in the absence of LRO ($\langle \mathbf{S}_i \rangle = 0$) we follow to reference [31] and consider the autocorrelation function of the nuclear spin $\mathbf{I}$ [35]. Performing calculations with the simplest Hamiltonian $H_\mathrm{hf} = A\mathbf{I}\mathbf{S}_i$ to second order in A we derive

$$(I^+, I^-)^\omega = \frac{2}{3}I(I+1)[-\mathrm{i}\omega + \Sigma(\omega)]$$

with the memory function

$$\Sigma(\omega) = A^2 \int_0^\infty \mathrm{d}t \exp(\mathrm{i}\omega t) \sum_{\mathbf{q}} \langle S^z_{-\mathbf{q}}(t) S^z_{\mathbf{q}} + \frac{1}{2} S^-_{-\mathbf{q}}(t) S^+_{\mathbf{q}} \rangle, \quad (51)$$

$$\langle S^\alpha_{-\mathbf{q}}(t) S^\beta_{\mathbf{q}} \rangle = \int_{-\infty}^\infty \mathrm{d}\omega \exp(\mathrm{i}\omega t) \mathcal{J}^{\alpha\beta}_{\mathbf{q}}(\omega),$$

$$\mathcal{J}^{\alpha\beta}_{\mathbf{q}}(\omega) = -\frac{1}{\pi} N(\omega) \mathrm{Im} \langle\langle S^\beta_{\mathbf{q}} | S^\alpha_{-\mathbf{q}} \rangle\rangle_\omega. \quad (52)$$

As discussed in reference [32], the spectral density $\mathcal{J}^{\alpha\beta}_{\mathbf{q}}(\omega)$ contains an almost singular contribution

$$\delta\mathcal{J}^{\alpha\beta}_{\mathbf{q}}(\omega) \propto \Delta_{\mathbf{q}-\mathbf{Q}} \Delta_\omega \quad (53)$$

where $\Delta_{\mathbf{q}}$ and Δ_ω are delta-like functions smeared at the scales $q \sim \xi^{-1}$ and $\omega \sim \omega_\xi$ with the characteristic spin-fluctuation energy $\omega_\xi \sim \mathcal{D}\xi^{-2}$ (FM), $\omega_\xi \sim c\xi^{-1}$ (AFM). To obtain the singular term in $\Sigma(\omega)$ with the correct factor of S^2 we can introduce a very small magnetic anisotropy (which does not violate time-reversal symmetry), so that the whole singular contribution passes to $K^{zz}_{\mathbf{q}}(\omega)$ (*cf.* Ref. [31]). Then the term $\mathrm{i}A^2S^2/\omega$ occurs at $\omega \gg \omega_\xi$, and we obtain the expression

$$(I^+, I^-)^\omega = \frac{\mathrm{i}}{3} I(I+1) \left(\frac{1}{\omega - AS} + \frac{1}{\omega + AS} \right) \quad (54)$$

which describes precession of the nuclear spin with *both* frequencies $\pm AS$. We see that the resonance picture holds at $\omega_\mathrm{n} \gg \omega_\xi$ only. In the opposite case the NMR line is smeared, but we can calculate the quantity $1/T_1$ according to (5).

Provided that $\omega_0 \ll \omega_\xi$ the quantity ω_ξ plays a role of the gap in the magnon spectrum. Therefore at $\omega_\mathrm{n} \ll \omega_\xi$ the cutoffs in the singular contributions to $1/T_1$ described by (42, 46, 31) are determined by very small inverse correlation length, so that they have very large values and possess unusual temperature behavior.

In the 2D FM case the expression (26) is also applicable, but with another expression for $\omega_0, \omega_0 \to \mathcal{D}\xi^{-2}$ which is exponentially small. We have

$$\delta^{(2)}(1/T_1) \propto I^2A^2T^2/\mathcal{D}^{5/2}\omega_\xi^{1/2} \propto I^2\omega_\mathrm{n}^2\xi T^2/\mathcal{D}^3. \quad (55)$$

In the isotropic 2D AFM case we obtain from (42)

$$\delta^{(1)}(1/T_1) \propto I^2A^2T/c\omega_\xi \propto I^2\omega_\mathrm{n}^2\xi T/c^2. \quad (56)$$

As follows from (46, 50),

$$\delta^{(2)}(1/T_1) \propto I^2A^2(T^2/c^4)\ln^2\xi \simeq \mathrm{const}(T). \quad (57)$$

Possibility to observe such dependences experimentally is of great interest. Unfortunately, most experimental data for layered compounds deal with copper-oxide systems which are on the borderline of AFM instability. The latter results in a specific temperature dependence of the spin susceptibility and strong deviations from the Korringa law. This makes separation of one- and two-magnon contributions impossible.

At $\omega_{\rm n} \gg \omega_\xi$ the cutoff in the singular contributions to $1/T_1$ described by (42, 46, 31) is the NMR frequency $\omega_{\rm n}$. However, such a cutoff is absent for the corresponding terms in $1/T_2$ owing to the interaction $\widetilde{A} \sim a|F^{(1)}|$. Provided that $\widetilde{A} \sim A$ we reproduce for these terms the dependences (55–57). However, as discussed in Section 2, for large ξ the lineform turns out to be Gaussian owing to strong Ruderman-Kittel interaction between nuclear spins, the Moriya formula (6) is inapplicable, and one can estimate the linewidth from (12). In the 2D case we obtain $1/T_2 \propto \xi$. A more detailed discussion of this situation and an application to copper-oxide systems is given in reference [15].

We see that NMR investigations can be in principle used to obtain the temperature dependence of the correlation length. When crossing the magnetic ordering point in layered systems the NMR picture should not change radically. Formally, as follows from (49, 50) at $T \sim T_{\rm M}$ we have $\ln \xi \simeq \ln |JS^2/\max\{\omega_0, |J'|\}|$, so that the cutoffs are joined smoothly.

A similar situation can take place for other systems with suppressed LRO and strong SRO, *e.g.*, for frustrated 3D magnetic systems [36] (where ξ is also large, the lineform is Gaussian, and we obtain from (12) $1/T_2 \propto \xi^{1/2}$). This may explain why the problem of detecting long-range magnetic ordering is frustrated systems with small ordered moments with the use of the NMR method so difficult. Indeed, the NMR data for heavy-fermion systems are doubtful and contradict to results of other experiments [1].

6 Conclusions

In the present paper we have investigated in detail various mechanism of nuclear magnetic relaxation in metallic ferro- and antiferromagnets in the spin-wave temperature region. In the most cases the main contribution to $1/T_1$ is of Korringa type, but its physical origin is more complicated than in paramagnetic metals. 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 3D antiferromagnets such a contribution contains the logarithmic singularity which is cut at the gap in the magnon spectrum (magnetic anisotropy energy) ω_0. Thus we can conclude that the "Korringa" relaxation rate in 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). In the 2D AFM case we have $1/T_1 \propto \omega_0^{-1}$. In the isotropic limit the singularity in $1/T_1$ is cut at very small inverse correlation length, so that the one-magnon contribution becomes very large and possesses unusual temperature behavior.

Besides that, we have calculated contributions from more complicated magnon damping processes (electron-magnon scattering). For antiferromagnets and 2D ferromagnets they contain singular logarithmic or (in the 2D FM case) power-law factors which are also cut at ω_0. These contributions may result in considerable deviations of the temperature dependence of $1/T_1$ from the linear Korringa law. In the 3D FM case this contribution is also noticeable and probably can be separated when fitting experimental data. For half-metallic ferromagnets, where the "Stoner" damping is absent, this scattering should be the main nuclear relaxation mechanism (see the discussion of experimental data in the review [17]).

Provided that the "longitudinal" matrix elements of dipole-dipole hyperfine interactions in (9) are not too small, the two-magnon ("Raman") relaxation processes may be also important in $1/T_1$, especially for 2D ferromagnets.

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