Narrowing of the magnetic-resonance line by fluctuations of the exchange interaction in disordered metals

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A kinetic equation is derived for the magnetization of localized spins in disordered metals. The exchange interaction is taken into account. If the correlation function of the exchange interaction, $\omega_{ex}^2 = \frac{1}{8}\Sigma_j < J_{ij}^2 - \langle J_{ij}\rangle_{imp}^2$, is larger than the width (δ) of the local-field distribution, the line acquires a Lorentzian shape and a width $\sim\delta^3/\omega_{ex}^2$.

1. The "exchange narrowing" of a magnetic-resonance line, whose nature was determined in research by Anderson,[1,2] consists of an averaging of the local fields (dipole fields, nonuniform fields, and hyperfine fields) during the rapid exchange motion of the magnetization. In pure metals the primary exchange mechanism is the long-range indirect Ruderman–Kittel (RKKY) exchange, which falls off as $r^{-3}\cos(2k_F r)$ with distance (k_F is the Fermi momentum). Since the work by De Gennes,[3] it has become the general belief that the RKKY interaction falls off exponentially over the mean free path of the conduction electrons, l, in impure metals.

Let us imagine a disordered metal with a mean free path several times the lattice constant a. The metal contains a small number of magnetic impurities, which are separated by an average distance $r_{av} \approx a c^{-1/3} > 1$ ($c \lesssim 0.01$ is the relative concentration of these magnetic impurities). In this case the overwhelming majority of the spins of the magnetic ions are not coupled with each other by exchange. Obviously, there can be no exchange narrowing of the magnetic-resonance line.

In the present letter we make use of an idea from Refs. 4 and 5 which suggests that at distances $r > l$ the RKKY potential does not fall off exponentially. It becomes a random function of the coordinates of the spins. We will show that the exchange narrowing of the magnetic-resonance line reaches its full potential in a disordered magnetically dilute metal.

2. We assume that the primary source of the linewidth is a spread in local fields. The Hamiltonian of the problem is then

$$H = \sum_i \omega_i S_i^z + \frac{1}{2}\sum_{i\neq j} J_{ij}\vec{S}_i\vec{S}_j + \omega_1(t)\sum_i (S_i^+ + S_i^-), \tag{1}$$

where $\omega_i = \omega_s + \Omega_i$ is the local precession frequency of spin i, ω_s is the Larmor frequency, and we set $S = 1/2$ to simplify the calculations. We write the exchange interaction in (1) in the form

$$J_{ij}\vec{S}_i\vec{S}_j = (\delta J_{ij})\vec{S}_i\vec{S}_j + < J_{ij} > \text{imp}\ \vec{S}_i\vec{S}_j, \tag{2}$$

where $\langle ... \rangle_{\text{imp}}$ means an average over the positions of the impurities. We will omit the last term in (2) in the calculations below, since we have $\langle J_{ij} \rangle \text{imp} \sim \exp(-r/l)^{3-5}$. We write an equation of motion for the average transverse magnetization of spin i, $\sigma_i(t) = \langle S_i^+(t) \rangle$

$$i\frac{\partial \sigma_i}{\partial t} = < [S_i^+(t), H] > = -\omega_i\sigma_i - \sum_{j\neq i}(\delta J_{ij}) < S_i^+(t)S_j^z(t) - S_i^z(t)S_j^+(t) > . \tag{3}$$

We take the coupling of the ith and jth magnetic moments into account in the approximation linear in the concentration of spins. We thus write

$$i\frac{\partial}{\partial t} < S_i^+(t)S_j^z(t) > = -\omega_i < S_i^+(t)S_j^z(t) > + \frac{1}{4}(\delta J_{ij})(\sigma_j(t) - \sigma_i(t)). \tag{4}$$

Substituting the solution of (4) into (3), we find an integrodifferential equation for the magnetization of spin i, with a kernel which depends on a difference between arguments:

$$\frac{\partial \sigma_i}{\partial t} = i\omega_i\sigma_i + \frac{1}{4}\sum_{j\neq i}(\delta J_{ij})^2 \int_{t_0}^{t} dt_1(\sigma_j(t_1) - \sigma_i(t_1))(e^{i\omega_i(t-t_1)} + e^{i\omega_j(t-t_1)}). \tag{5}$$

For a steady-state magnetic resonance the lower limit on this integral is $t_0 = -\infty$; for the regime of a decay of free induction it is instead $t_0 = 0$. We find a solution of (5) by taking Fourier (Laplace) transforms:

$$\sigma_i(\omega) = \frac{1}{\Delta}\sigma_i(t=0) + \frac{1}{4\Delta}\sum_{j\neq i}(\delta J_{ij})^2(K_\omega^i + K_\omega^j)\sigma_j(\omega), \tag{6}$$

$$\Delta = i(\omega - \omega_i) + \frac{1}{4}\sum_{j\neq i}(\delta J_{ij})^2(K_\omega^i + K_\omega^j), \quad K_\omega^i = \frac{1}{i(\omega - \omega_i)}.$$

The one-particle mechanism for the relaxation of the transverse magnetization, which generates the homogeneous width, is introduced in the standard way: $\omega_i \to \omega_i + i/T_2$. We will understand ω_i below to mean specifically this expression. For σ_i, we then find an iterative expansion from (6). We average each term of the series over the positions of the impurities. The averaging is dominated by the ring diagrams shown in Fig. 1; the loops correspond to the RKKY interaction $J_{ij} = J^2T\Sigma_\omega\sigma_\omega(\vec{r}_i\vec{r}_j)\sigma_\omega(\vec{r}_i\vec{r}_j)$, $\sigma_\omega(\vec{r}_i\vec{r}_j)$ is the electron temperature Green's function,

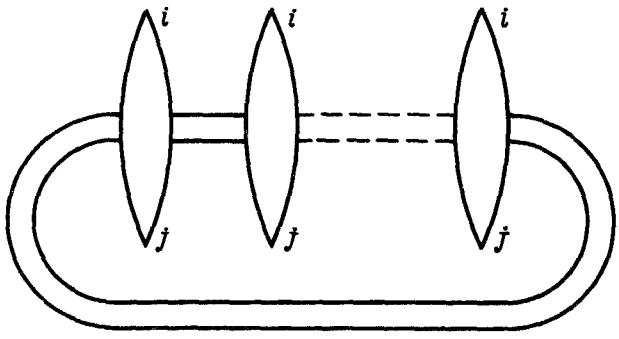

FIG. 1.

and J is the integral of the interaction of a localized spin with the spin of a conduction electron. The double lines connecting the electron loops correspond to a diffusion or a cooperon, depending on the direction of the arrows. These diagrams are proportional to r^{-3k} (k is the number of loops) for $l<r<\xi$, where ξ is the length over which the phase of the conduction electrons relaxes. Diagrams with unequal indices generate a factor $\exp(-r_{ij}/l)$. Diagrams with different numbers of diffusions and coopersons and also diagrams with identical indices but different architectures are small quantities on the order of $(k_F l)^{-1}$. A summation of the ring diagrams leads to an equation for $\overline{\sigma_i(t)}$, whose form is the same as that of Eq. (5), with $\sigma_i(t)$ replaced by $\overline{\sigma_i(t)}$ and $(\delta J_{ij})^2$ replaced by $1/2\ \overline{(\delta J_{ij})^2}$:

$$\frac{\partial\bar{\sigma}_i}{\partial t} = i\omega_i\bar{\sigma}_i + \frac{1}{8}\sum_{j\neq i}\overline{(\delta J_{ij})^2}\int_{t_0}^{t} dt_1(\bar{\sigma}_j(t_1) - \bar{\sigma}_i(t_1))(e^{i\omega_i(t-t_1)} + e^{i\omega_j(t-t_1)}). \quad (7)$$

The quantity $\overline{(\delta J_{ij})^2}$ in (7) depends on only $|\mathbf{r}_i - \mathbf{r}_j|$, so we can find a closed equation for the transverse magnetization of a spin packet,

$$M_n(t) = \sum_{i\in\{n\}}\bar{\sigma}_i(t),$$

i.e., for a set of spins which have a identical precession frequency $\omega_i = \omega_n$. After we go over to a continuous distribution of packet frequencies, with a distribution function $g(\Omega)$, the kinetic equation for the transverse component of the magnetization spectral density $M_\Omega(t)$ becomes

$$\frac{\partial M_\Omega(t)}{\partial t} - i(\omega_s + \Omega + \frac{i}{T_2})M_\Omega(t) = \omega_{ex}^2\int_{t_0}^{t} dt_1 g(\Omega)e^{i\tilde{\Omega}(t-t_1)}\int d\Omega' M_{\Omega'}(t_1)$$

$$+\omega_{ex}^2\int_{t_0}^{t} dt_1 g(\Omega)\int d\Omega' M_{\Omega'}(t_1)e^{i\tilde{\Omega}(t-t_1)} - \omega_{ex}^2\int_{t_0}^{t} dt_1 M_\Omega(t_1)e^{i\tilde{\Omega}(t-t_1)} \quad (8)$$

$$-\omega_{ex}^2\int_{t_0}^{t} dt_1 M_\Omega(t_1)\int d\Omega' e^{i\tilde{\Omega}'(t-t_1)}g(\Omega'),$$

$$\omega_{ex}^2 = \frac{n\rho_F^2 J^4}{8\pi l^3}, \tag{9}$$

where $\tilde{\Omega} = \omega_s + \Omega + i/T_2$, ρ_F is the density of electron states at the Fermi level, and n is the number of spins per unit volume. Integrating Eq. (8) over the frequency Ω, we verify that its right side vanishes, as it should in the case of scalar exchange, which commutes with any component of the total magnetization. A solution of Eq. (8) is

$$M_\Omega(\omega) = \frac{-ig(\Omega)M_0 + \omega_{ex}^2[g(\Omega)K_\Omega(\omega)\int d\Omega' M_{\Omega'}(\omega) + \int d\Omega' M_{\Omega'}(\omega)K_{\Omega'}(\omega)]}{i(\omega - \omega_s - \Omega) + \frac{1}{T_2} + \omega_{ex}^2[K_\Omega(\omega) + \int d\Omega' g(\Omega')K_{\Omega'}(\omega)]}, \tag{10}$$

where M_0 is the static magnetization. The observed shape of the magnetic-resonance line is proportional to the spectral density of the total magnetization, $M(\omega)$:

$$M(\omega) = \int d\Omega M_\Omega(\omega) = -\frac{iA_0 M_0}{\omega_{ex}^2[(1-A_1)^2 - A_0 A_2]},$$
$$A_n = \omega_{ex}^2 \int d\Omega \frac{g(\Omega)}{\Delta\Omega}(K_\Omega(\omega))^n, \quad K_\Omega(\omega) = \frac{1}{i(\omega - \omega_s - \Omega) + 1/T_2}, \tag{11}$$
$$\Delta_\Omega = i(\omega - \omega_s - \Omega) + \frac{1}{T_2} + \omega_{ex}^2[K_\Omega(\omega) + \int d\Omega' g(\Omega')K_{\Omega'}(\omega)].$$

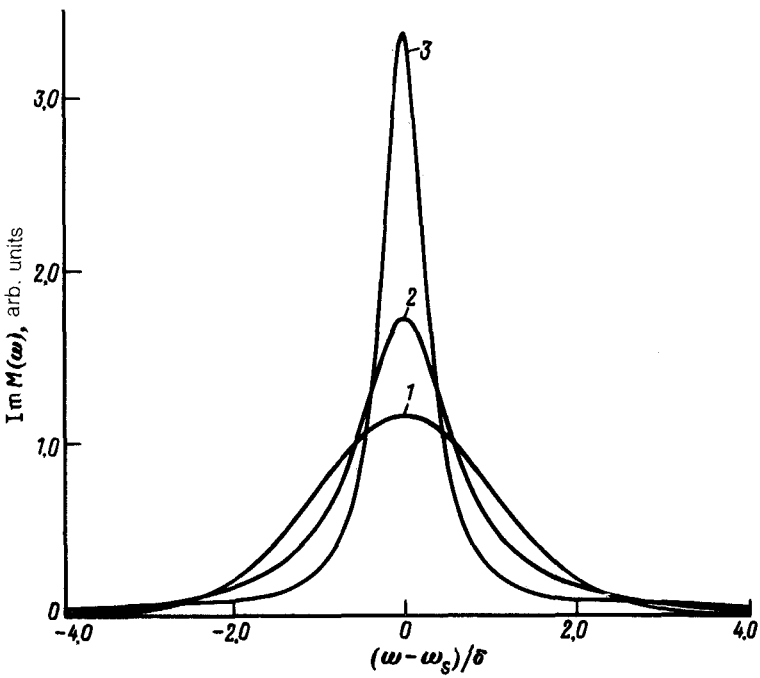

FIG. 2.

We can work from (11) to construct the shape of the resonant-absorption line for an arbitrary distribution of local fields $g(\Omega)$. Figure 2 illustrates the process by which the magnetic-resonance line narrows, and the shape of the line changes, as ω_{ex} increases. As $g(\Omega)$ we have adopted a Gaussian distribution $(2\pi\delta)^{-1/2}\exp[-(\Omega^2/2\delta^2)]$. Curve 1 in this figure corresponds to $\omega_{ex}/\delta = 0$, curve 2 to $\omega_{ex}/\delta = 1$, and curve 3 to $\omega_{ex}/\delta = 2$; for all three curves, we are using $1/T_2 = 0.1\delta$.

An estimate based on the typical parameter values $\rho_F J \approx 10^{-2}$–10^{-1}, $c \approx 10^{-3}$–10^{-2}, and $l \approx 2a$–$5a$ yields $\omega_{ex} \approx 10^9$–10^{12} Hz. If, in the calculation of ω_{ex}^2, we replace $\langle(\delta J_{ij})^2\rangle$ by $\langle J_{ij}\rangle_{imp}^2$, we find a result smaller by a factor[6] $\exp(2r_{cp}/l) \sim 10^2$–$10^3 \sim p$. The condition for exchange narrowing, $\omega_{ex} > \delta$, is essentially impossible to satisfy, at least at standard ESR frequencies, $\sim 10^{10}$ Hz.

In the limit $\omega_{ex} > \delta$, $|\omega - \omega_s|$, the shape of the exchange-narrowed magnetic-resonance line can be found analytically. An evaluation of the integrals in (11) leads to the Lorentzian shape

$$M(\omega) = \frac{\omega_s M_0}{\omega_s - \omega + i\left[\frac{1}{T_2} + \sqrt{\frac{2}{\pi}}\frac{\delta^3}{\omega_{ex}^2}\right]}. \tag{12}$$

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