Kinetics of nuclear magnetization in a disordered conductor

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Large-scale fluctuations in the Korringa relaxation rate in a dirty conductor cause a nonexponential decay of the nuclear magnetization. The spin diffusion of the nuclei, which completely determines the asymptotic behavior after a long time, plays an important role.

1. The predominant mechanism for the restoration of the equilibrium value of the nuclear magnetization in a metal is the interaction with thermal fluctuations of the spin density of the conduction electrons (Korringa relaxation). In the presence of nonmagnetic impurities, defects, etc., the relaxation rate $T_1^{-1}(r)$, determined by the

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local contact interaction, will clearly become a random function of the coordinate of the nucleus, so a sample will have a spectrum of relaxation times.[1] The implications for the kinetics of nuclear magnetization are not obvious, since the situation is complicated by a diffusion of nuclear spins. It is usually assumed that a fast spin diffusion, determined by the dipole and indirect interactions between nuclei (which, in terms of energy, exceed the reciprocals of the Korringa relaxation time by several orders of magnitude), leads to an averaging of the rates $T_1^{-1}(r)$ and to an exponential decay law for the nuclear magnetization, $m(t)$. The observed deviations from this law are attributed to uncontrolled magnetic impurities in the sample.[2]

In the present letter we show that the spin diffusion actually averages out only the short-wavelength variations in the magnetization, and the fluctuations in $T_1^{-1}(r)$ at large distances (on the order of the phase coherence length of the electrons) lead to a relaxation which is slower than exponential:

$$\frac{\delta m(t)}{m_0} = \exp\left(-\frac{t}{T_1} + \varphi(t)\right). \tag{1}$$

Here T_1^{-1} is the relaxation rate averaged over the positions of the impurities,[3] and the function $\varphi(t)$ takes the following form over the typical experimental times ($t < \tau$, with τ on the order of a few times T_1) for films and bulk metals:

$$\begin{aligned} \varphi(t) &= \left(\frac{t}{T_1}\right)^2 \frac{1}{3\pi(k_F l)^2} \ln^2\left(\frac{2t^*}{\pi t}\right) && d=2, \\ \varphi(t) &= \frac{t}{T_1} \frac{3\pi}{4D_s T_1 (k_F^2 l)^2} \ln\frac{D_s t}{l^2} && d=3. \end{aligned} \tag{2}$$

The parameter $t^* = D/4D_s T$ is the time required for the diffusion of the nuclear excitation over a distance on the order of the thermal coherence length of the electrons; D_s is the spin-diffusion coefficient of the nuclei; D is the electron diffusion coefficient; k_F is the Fermi wave vector; and l is the mean free path of the electrons.

2. We know quite well[4] that the clearly expressed hierarchy of fluctuation times for the electron and nuclear spins means that the relaxation of the longitudinal nuclear magnetization can be described by a Bloch equation

$$\frac{\partial m(rt)}{\partial t} = D_s \nabla^2 m(rt) - \frac{m(rt)}{T_1(r)} . \tag{3}$$

The Korringa relation rate $T_1^{-1}(r,\omega) \sim \langle \sigma_r^+(t)\sigma_r^- \rangle_\omega$ is indeed determined by the correlation function of the transverse spin of the conduction electrons; the frequency ω must be on the order of the reciprocals of the nuclear time scales T_n. At such low frequencies, however, this correlation function no longer depends on ω, since the time scale for its decay due to the spin-orbit coupling, τ_{so}, is many orders of magnitude shorter than the time scales for the nuclear spin. Under the condition $\omega < \tau_{so}^{-1}$ we have $\langle \sigma_r^+(t)\sigma_r^- \rangle_\omega = f(N_r(\epsilon F), \tau_{so}^{(r)})$, where $N(\epsilon_F)$ is the state density, so there is a temporally local Eq. (3) with a random relaxation $T_1^{-1}(r)$.

The solution of (3) can be written in the form of a functional integral

$$\frac{\delta m(t)}{m_0} = \langle\langle \exp\{-\int \frac{d\tau}{T_1[r(\tau)]}\}\rangle_{imp}\rangle_{tr}. \quad (4)$$

Two averages are taken in (4): one over the positions of the nonmagnetic impurities, which are responsible for the spatial fluctuations in the spin density of the electrons, and one over the trajectories $r(t)$ of the diffusive motion of a nuclear excitation through the sample. The latter averaging leads to a "smoothing" of the fluctuations in the random quantity $T_1^{-1}(r)$ over length scales on the order of $\sqrt{D_s t}$. The relaxation rate, however, is not quantity which is directly related to the total charge density of the electrons, so it can fluctuate because of interference effects at large distances. Specifically, these large-scale fluctuations ($\sqrt{D_s^t} \ll r \ll \sqrt{D/T}$) are not averaged out by the spin diffusion, and they determine the deviation from an exponential relaxation.

It is not a simple matter to evaluate (4) for arbitrary times. For a weak localization, however, in which case the fluctuations are small ($\delta T_1^{-1} < T_1^{-1}$), and for a sufficiently wide time interval $t < t_0 \sim 1/\delta T_1^{-1} \gg T_1$, the quantity in (4) can be calculated through an expansion in a series in the fluctuations δT_1^{-1}. The function $m(t)$ then takes the form in (1), while the function $\varphi(t)$ is expressed in terms of the correlation function of the fluctuations in the relaxation rates, $K(r_1 - r_2) = \langle \delta T_1^{-1}(r_1)\delta T_1^{-1}(r_2)\rangle_{imp}$:

$$\varphi(t) = \int_0^t d\tau \int_0^\tau d\tau_1 \int dr P_s(r, \tau_1) K(r), \quad (5)$$

where $P_s(rt) = (4\pi D_s t)^{-d/2} \exp(-r^2/4D_s t)$ is the Green's function of the spin-diffusion equation. The fluctuations in $T_1^{-1}(r)$ are determined primarily by the fluctuations in the electron state density, since we have $T_1^{-1} \sim N^2(\epsilon_F)$. The fluctuations in τ_{s0} are unimportant here, since τ_{s0} appears in the expression for $T_1^{-1}(r)$ only within a logarithm [through the quantum-mechanical corrections of the type $(4\pi^2 DN(\epsilon_F))^{-1} \times \ln(\tau_{s0}/\tau)$]. The correlation function $K(r)$ is then determined by standard diagrams which describe fluctuations in the state density (two electron loops, taken at the points r_1 and r_2 and linked by either two diffusions or cooperons[6]); it is found to be

$$K(r) = \frac{1}{8(\pi^2 DT_1 N(\epsilon_F))^2 T} \int_{-\infty}^{\infty} \frac{d\Omega}{\sinh^2 \Omega/2T} \times \left(\frac{\Omega/2T}{\tanh \Omega/2T} - 1\right)(P_\Omega^D(r)P_\Omega^D(r) + P_\Omega^C(r)P_\Omega^C(r)), \quad (6)$$

where P_Ω^D and P_Ω^C are the diffusion and cooperon propagators, which satisfy the equations

$$(-\Omega - D\nabla^2 + \gamma_{C,D})P_\Omega^{(C,D)}(r, r_1) = \delta(r - r_1) \quad (7)$$

in coordinate space. Calculating them, and taking the integrals in (5) under the condition $t^* > t$, which holds if the temperature is not too high, we find expressions (2).

Expressions (2) hold at times $t < t_0$, where t_0 is the solution of the equation $T_1 \frac{\partial \varphi(t)}{\partial t} = 1$. In the weak-localization region we have $t_0 \gg T_1$, and experimentally we would observe a functional dependence (2) which is quite different from exponential. Corresponding measurements by pulsed NMR methods make it possible to determine the nuclear spin diffusion coefficient D_s and to obtain additional information about the parameters of disordered systems.

The asymptotic behavior of $m(t)$ at long times $t > t^*$ in this problem is controlled not by the wings of the distribution function of the relaxation times, as it would be in the absence of a spin diffusion,[1] but by another mechanism, specifically, the diffusive energy transport from slowly relaxing nuclei to "fast" nuclei over a distance $\sim\sqrt{D/T}$. This process must be accompanied by the onset of a behavior $\exp(-t/t^*)$, which could in principle be observed near a metal-insulator transition, where t^* is not too large in comparison with the characteristic values of T_1.

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