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Relaxation of nuclear spin due to long-range orbital currents

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We consider a model of metallic layers where transverse fluctuations in the currents in each layer produce a fluctuating magnetic field. This field relaxes a nuclear spin located anywhere in the crystal. We apply this model to an ordinary Fermi liquid, to a model of strongly correlated electrons that may be applicable to oxide superconductors, and to a metal just above its superconducting temperature.

The nuclear-spin-relaxation rate T_1^{-1} in metals is the sum of several contributions: (a) The unpaired electron spins at the Fermi surface relax the nuclear spin via either the contact interaction, or via core polarization in the case of an s orbital. (b) The dipolar interaction between electron and nuclear spins. (c) The orbital effect of the conduction electron, which gives rise to a magnetic field at the nucleus of the form $\mathbf{h}=\mathbf{L}/r^3$ where $\mathbf{L}$ is the angular momentum operator.

Effect (a) dominates in simple metals, whereas in some transition metals, like vanadium, (c) is the dominant mechanism.[1] The theory for the orbital effect was worked out by Obata,[2] based on a tight-binding electronic wave function and the linear superposition of atomic orbitals. The assumption was made that only the orbitals on the nucleus site couple significantly to the nuclear spin. The resulting T_1^{-1} is proportional to $\langle r^{-3}\rangle^2$, which is essentially given by the free-ionic value. Furthermore, this mechanism operates only if angular momentum matrix elements of the different atomic orbitals do not vanish. For example, for copper oxide the orbitals which make up the state at the Fermi surface are predominantly $d_{x^2-y^2}$ with some admixture of d_{z^2}. Since $\langle z^2|\mathbf{L}|x^2-y^2\rangle=0$, there is no orbital effect in this case.[3] Obata also compared the orbital effect with the dipolar coupling [effect (b)]. Since both couplings scale as r^{-3} their contributions are parametrically similar. Obata concluded that except in cases where the angular momentum matrix elements vanish, as in the example given above, the orbital effect is more important.

In this paper we re-examine the orbital contribution to T_1^{-1}. Obata has focused on the orbital current in the atom which contains the nuclear spin. Instead we consider the general question of the magnetic field at any point in the space generated by fluctuating current loops in the metal. To be specific, we consider a layered material which is modeled by a stack of conducting planes a distance d apart. Our first step is to derive a general relation between T_1 and the current-current correlation function in the metallic sheets. Next we apply our result to two cases. The first is where the metallic sheets are described by Fermi-liquid theory. We find that current fluctuations on all length scales contribute equally to T_1^{-1}. To our knowledge, the orbital effect of these larger current loops have not been discussed before. The second application is to the oxide superconductors as modeled by the strongly correlated Hubbard model. This system was in fact the original motivation for our study, because the possibility of spin and charge separation has been suggested for this system[4] and we would like to know whether T_1^{-1} can provide a probe for charge fluctuations. In particular, we have recently worked out the normal-state properties of one particular model, the uniform resonating-valence-bond (RVB) state.[5] We found that the important low-energy fluctuations are described by gauge fields, which represent fluctuations in the spin chirality $\mathbf{S}_1\cdot\mathbf{S}_2\times\mathbf{S}_3$. The spin chirality is in turn coupled to the transverse current in the plane, which should relax the nuclear spin. If the nucleus is located in an atom which lies between the conducting planes and is weakly coupled to the planes electronically, the usual Korringa relaxation [mechanism (a)] may become very small; there is a chance that the orbital effect due to charge current loops in the plane may dominate the relaxation rate. In this case T_1^{-1} will provide valuable information on the current and chirality fluctuation in the planes. We give an explicit estimate of the relaxation rate, which unfortunately turns out to be very small. We also discuss the possibility of enhancing this effect by approaching the superconducting temperature from above.

The relaxation rate when the external field is in the z direction is given by

$$\frac{1}{T_{1z}}=\lim_{\omega\to 0}\left[\frac{kT}{\omega}\gamma_n^2\,\mathrm{Im}\langle H_+(R_n,\omega)H_-(R_n,\omega)\rangle\right], \qquad (1)$$

where γ_n is the nuclear moment, and $H_\pm=H_x\pm iH_y$, where $\mathbf{H}(R_n)$ is the fluctuating magnetic field at the nuclear site R_n. In the orbital mechanism, the field is generated by current loops in the metal. We assume that the currents are confined to the layers,

$$\vec{\mathrm{j}}(\mathbf{R})=\sum_n \vec{\mathrm{j}}_n(\vec{\mathrm{r}})\delta(z-nd)$$

where $\mathbf{R}=(\vec{\mathrm{r}},z)$ and arrows and boldface denote two-dimensional (2D) and 3D vectors, respectively. We consider only transverse currents so that $\vec{\nabla}\cdot\vec{\mathrm{j}}=0$. Thus the Fourier components of $\vec{\mathrm{j}}$ can be parametrized by $j_x(\mathbf{Q})=i(q_y/q)j_\perp(\mathbf{Q})$ and $j_y(\mathbf{Q})=-i(q_x/q)j_\perp(\mathbf{Q})$ where $\mathbf{Q}=(\vec{\mathrm{q}},q_z)$. The magnetic field $\mathbf{H}(\mathbf{R})$ is determined by the 3D Maxwell equations, $\nabla\cdot\mathbf{H}=(4\pi/c)\mathbf{j}$ and $\nabla\cdot\mathbf{H}=0$, where $\mathbf{j}=(\vec{\mathrm{j}},0)$. Since $\mathbf{j}_z=0$, we have $q_xH_y(\mathbf{Q})-q_yH_x(\mathbf{Q})=0$ so that we can parametrize $H_x(\mathbf{Q})=(q_x/q)H_\perp(\mathbf{Q})$ and $H_y(\mathbf{Q})=(q_y/q)H_\perp(\mathbf{Q})$. The Max-

well equations are readily solved to give $H_\perp(\mathbf{Q},\omega)=(4\pi i/c)(q_z/Q^2)j_\perp(\mathbf{Q},\omega)$ and $H_z(\mathbf{Q},\omega)=(4\pi i/c)(q/Q^2)j_\perp(\mathbf{Q},\omega)$. This allows us to relate the frequency spectrum of the magnetic field at a point $\mathbf{R}$ to the current-fluctuation spectrum. By assuming that currents in different layers are uncorrelated, we obtain after performing the q_z integration

$$\langle|H_z(\mathbf{R},\omega)|^2\rangle=\left(\frac{4\pi}{c}\right)^2\frac{1}{d}\int\frac{d^2q}{(2\pi)^2}\sum_m\frac{e^{iG_mz}q}{4q^2+G_m^2}\langle j(\vec{q},\omega)j(-\vec{q},\omega)\rangle_{2d}\,, \tag{2}$$

where $G_m=2\pi m/d$ and $\langle|H_\perp(\mathbf{R},\omega)|^2\rangle=\langle|H_z|^2\rangle$ so that

$$\langle|H_x(\mathbf{R},\omega)|^2\rangle=\langle|H_y(\mathbf{R},\omega)|^2=\tfrac{1}{2}\langle|H_z(\mathbf{R},\omega)|^2\rangle\,. \tag{3}$$

Since only the component normal to the external field can relax the nuclear spin, we conclude that

$$\frac{1}{T_{1x}}=\frac{1}{T_{1y}}=\frac{3}{2}\frac{1}{T_{1z}}\,. \tag{4}$$

Furthermore, provided that the dominant contributions come from $q<\pi/d$, we can restrict the sum over m to the term $m=0$, in which case the relaxation rate is independent of the location of the nucleus. This feature, together with the unusual anisotropy given by Eq. (4), are useful signatures to distinguish the present mechanism from other contributions.

We next apply Eq. (2) to a model where each layer is described by Fermi-liquid theory with mass m_F. The transverse current-current response function $\Pi_\perp^F(q,\omega)$, the imaginary part of which is proportional to the correlation function $\langle j(q,\omega)j(-q,-\omega)\rangle_{2d}$, takes the form[6]

$$\Pi_\perp^F(q,\omega)=\chi_Fq^2+i\omega\sigma_\perp(q,\omega) \tag{5}$$

for $\omega<v_Fq$ and $q<k_F$, where $\chi_F=\hbar/(24\pi m_F)$ is the Landau diamagnetic susceptibility and $\sigma_\perp$ is the transverse conductivity. In the presence of scattering mechanisms giving rise to a mean free path l, $\sigma_\perp$ can be replaced by the unusual static conductivity σ_0 for $ql<l$, where $\sigma_0\approx(e^2/h)k_Fl$ in $2d$. For $ql>1$, $\sigma_\perp$ is given by the anomalous-skin-effect expression which in $2d$ reads $\sigma_\perp(q,0)=e^2k_F/2\pi q$. Substituting Eq. (5) into Eqs. (1) and (2) and keeping only the $m=0$ term, we obtain

$$T_1^{-1}=\gamma_n^2kT\left(\frac{4\pi}{c}\right)^2\frac{1}{d}\int\frac{d^2q}{(2\pi)^2}\frac{1}{4q}\sigma_\perp(q,0)\,. \tag{6}$$

We readily see that for $q>l^{-1}$, all q scales contribute equally, leading to a logarithmic singularity. The contribution from this regime dominates that from $q<l^{-1}$ and we can approximate the integral over q by a lower cutoff at $q=l^{-1}$ and an upper cutoff at k_F, so that

$$\frac{1}{T_1}=kT\gamma_n^2\left(\frac{e}{c}\right)^2\frac{\hbar k_F}{d}\ln(k_fl)\,. \tag{7}$$

Note that for clean metals where l is dominated by inelastic scattering so that $l\sim T^{-p}$, we obtain $T_1^{-1}\sim T\ln T$, a slight departure from the Korringa law for spin-fluctuation contribution

$$(T_1^{-1})_{\text{Korringa}}=kT\gamma_n^2\gamma_e^2\hbar^3\frac{64\pi^3}{9}\langle|u_k(0)|^2\rangle^2\rho^2\,, \tag{8}$$

where $\gamma_e=e\hbar/2mc$, ρ is the density of states, and $\langle|u_k(0)|^2\rangle$ represents the enhancement of electron density at the nucleus site above the uniform density. It is instructive to rewrite Eq. (7) in a form which facilitates comparison with Eq. (8). For the layered system, $\rho=m_F/2\pi d\hbar^2$ so that

$$T_1^{-1}=kT\gamma_n^2\gamma_e^2\hbar^3\rho^2 4k_Fd(m/m_F)^2\ln(k_Fl)\,. \tag{9}$$

Thus the dimensionless geometrical factor $4k_Fd(m/m_F)^2\ln(k_Fl)$ which is of order unity replaces the density enhancement factor $\langle|u_k(0)|^2\rangle^2$.

The numerical value of the contribution to T_1 due to orbital effect is conveniently evaluated based on Eq. (7). Using $k_F\approx10^8$ cm^{-1}, $d\approx10^{-7}$ cm, $\ln(k_Fl)\approx5$, and taking γ_n to be the nuclear magneton $\mu_n\approx e\hbar/2Mc$, where M is the proton mass, we obtain $T_{1\,\text{Korringa}}\approx2000$ sec at $T=100$ K. In contrast, for a simple metal-like lithium, T_1 is ≈0.1 sec. This is because the enhancement factor $\langle|u_k|^2\rangle^2$ is very large; for lithium it is estimated to be of order 10^3. The long lifetime makes the orbital effect difficult to observe. The only hope may be to use a nucleus with a large γ_n (for example, γ_n for hydrogen is $\approx3\mu_n$, giving a factor of 10 enhancement) and choose a site which is weakly coupled to the electronic states in the layers, so that the orbital effect becomes the dominant mechanism. Another possibility is to observe the effect in the vicinity of the onset of superconductivity, a possibility which we will return to later.

Next, we consider another application of Eq. (2) where the layers are modeled as strongly correlated metals described by the t-J model. We have recently analyzed the normal state of the uniform resonating-valence-bond (RVB) state in terms of decoupling into fermions, which carry spin labels, and slave bosons.[5] We argued that this model is a promising candidate for the description of the anomalous normal-state properties of the oxide superconductors. The physical picture is that the important low-energy fluctuations are fluctuations in the chirality $\mathbf{S}_1\cdot\mathbf{S}_2\times\mathbf{S}_3$. These fluctuations are coupled to fluctuations in the physical transverse current in the layer. For our purpose it suffices to state that the physical current-current response function is given by the composition law[5]

$$\Pi_\perp(q,\omega)=\frac{\Pi_\perp^F(q,\omega)\Pi_\perp^B(q,\omega)}{\Pi_\perp^F(q,\omega)+\Pi_\perp^B(q,\omega)}\,, \tag{10}$$

where $\Pi_\perp^F$ is given by Eq. (5) and $\Pi_\perp^B=\Pi_\perp^{B'}+i\omega\sigma_B(q)$. The real part $\Pi_\perp^{B'}$ is given by χ_Bq^2 for $q<\lambda^{-1}$ where $\chi_B=\hbar T_{BE}^{(0)}/48\pi mT$, $T_{BE}^{(0)}=2\pi x\hbar^2/m$, x is the concentration of holes in the plane and λ is the de Broglie wavelength. For $q>\lambda^{-1}$, we have $\Pi_\perp^{B'}=\hbar\delta/2m$. In Ref. 5 we showed that $\sigma_B\ll\sigma_F$ so that the imaginary part of $\Pi_\perp^B$ can be ignored. With this approximation, we combine Eqs.

(10), (2), and (1) to get

$$(T_1^{-1})_{\rm corr} = \gamma_n^2 kT \left(\frac{4\pi}{c}\right)^2 \frac{1}{d} \int \frac{d^2q}{(2\pi)^2} \frac{1}{4q} \frac{\sigma_\perp^F(q,0)(\Pi_\perp^{B'})^2}{(\chi_F q^2+\Pi_\perp^{B'})^2} . \quad (11)$$

Note that in this formalism the Fermi-liquid theory is recovered if the bosons undergo Bose-Einstein condensation. In this case $\Pi_\perp^{B'}$ diverges and Eq. (11) reduces to Eq. (6). From Eq. (11), it is easy to see that in this model of strongly correlated metals, T_1^{-1} is always less than that from the Fermi-liquid result.

It is readily seen that for $q<\lambda^{-1}$, the contribution to the integral in Eq. (11) is again logarithmically divergent and dominates over the contribution from $q>\lambda^{-1}$. Keeping this contribution alone, we obtain

$$(T_1^{-1})_{\rm corr} = T_{10}^{-1}[\chi_B^2(T)/\chi_d^2(T)]\ln(l/\lambda)/\ln(k_F l) , \quad (12)$$

where T_{10}^{-1} is the relaxation rate for Fermi liquid given by Eq. (6) and $\chi_d=\chi_F+\chi_B$. Thus we see that apart from logarithmic terms, the reduction from the Fermi-liquid behavior is proportional to $[\chi_B(T)/\chi_d(T)]^2$, which is an important parameter in the strong-correlation theory. It will be valuable to determine this parameter experimentally, even though as we saw from our estimates, the rate is so small that the prospect of its measurement is not promising.

We next consider the nuclear-spin relaxation rate due to the orbital current when the superconducting transition temperature is approached from above.[7] We expect an enhancement of the orbital current fluctuation as the metal is tending towards a perfect diamagnet. This problem is easily treated using the time-dependent Ginsburg-Landau (TDGL) phenomenological theory. The free energy is given by

$$F=\sum_q \left(\frac{\hbar^2 q^2}{2m}+|\alpha|\right)\psi_q^*\psi_q , \quad (13)$$

where ψ_q is the Fourier component of the superconducting order parameter, $\alpha=\alpha'(T-T_c)$, and the coherence length $\xi^2=\hbar^2/2m|\alpha|$. The fluctuation spectrum is given according to TDGL by

$$\langle\psi_q^*(0)\psi_q(\tau)\rangle = \frac{2m}{\hbar^2}\frac{kT}{q^2+\xi^{-2}} e^{-|\tau|/\tau_k} ,$$

where $\tau_k^{-1}=(\hbar^2/m)(k^2+\xi^{-2})\gamma$ and $\gamma=\hbar\pi/8kT_c$. The current-current correlation function is computed as

$$\langle j_q^\mu(t) j_{-q}^\nu(0)\rangle = \left(\frac{e\hbar}{m}\right)^2 \sum_k (2k+q)_\mu (2k+q)_\nu \langle\psi_k^*(t)\psi_k(0)\rangle\langle\psi_{k+q}(t)\psi_{k+q}^*(0)\rangle . \quad (14)$$

Computing Eq. (14) in the limit of $q=0$, $\omega\to 0$ yields the well-known expression for the Aslamasov-Larkin contribution to the fluctuation conductivity. Here we need instead the opposite limit, $\omega\ll v_F q$, and then we substitute Eq. (14) into Eq. (2). After some computation, we obtain the result

$$T_{1\,\rm fluc}^{-1}/T_{10}^{-1} = \frac{2\pi^3 A(\xi/\xi_0)}{(k_F\xi_0)\ln(k_F l)} , \quad (15)$$

where

$$A=\int \frac{d^2q}{(2\pi)^2}\frac{d^2k}{(2\pi)^2}\frac{1}{q}\frac{(2\vec{k}+\vec{q})^2-[(2\vec{k}+\vec{q})\cdot\vec{q}]^2/q^2}{(\vec{k}^2+1)[(\vec{k}+\vec{q})^2+1][\vec{k}^2+(\vec{k}+\vec{q})^2+2]}$$

is a numerical coefficient of order unity. Thus we see that T_1^{-1} is enhanced near T_c as $(T-T_c)^{-1/2}$. Furthermore, the effect is largest when T_c is large and when $k_F\xi_0$ is small, which suggests that the oxide superconductors are the best candidate to observe this effect. However, we must keep in mind that the Gaussian fluctuation theory used here is valid only outside the critical region, which becomes large when $\xi_0 k_F$ is small, so that this calculation should be taken only as a qualitative indication of possible enhancement.

Finally, we remark that dipolar coupling with local electron-spin moments also decays as r^{-3}. The range is the same as the contribution of the orbital current, so that its contribution is parametrically similar. For ordinary orbital effects, Obata[2] concluded that dipolar effects are numerically smaller, and a similar conclusion probably holds for the present case. For strongly correlated metals where there exist local moments, the antiferromagnetic correlation between the moments will lead to additional cancellations for the dipolar contribution. Thus it is likely that if the nucleus is sufficiently isolated from the conduction-electron spin in the metallic layers, the long-range orbital current considered in this paper may be the dominant relaxation mechanism.

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