Effect of finite mean free path on spin-flip scattering rates near the critical point of magnetically ordered systems

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The finite electron mean free path of conduction electrons is found to play an important role in determining spin-flip scattering rates, $1/\tau_{sf}$, near a magnetic critical point. Small-momentum-transfer processes are suppressed and the temperature dependence of $1/\tau_{sf}$ is dominated by large-momentum scattering. For $T \gtrsim T_c$, $1/\tau_{sf}$ is monotonically increasing and its leading singular temperature dependence varies as $t^{1-\alpha}$, where $t=(T-T_c)/T_c$. This temperature dependence is also present for $T \lesssim T_c$, but for Heisenberg systems, of present interest, the *leading* singular term varies as $(-t)^{2-\alpha-\phi}$, where ϕ is the crossover exponent.

There has been interest recently in superconducting tunneling in proximity-effect tunnel junctions between a superconductor and various types of magnetically ordered materials, including ferromagnets, antiferromagnets, spin glasses, and magnetic superconductors.[1-7] The zero-bias conductance of these sandwich junctions, when corrected for thermal smearing of the density of states, is interpreted in terms of the spin-flip scattering time of the itinerant electrons in the magnetically ordered material.[8,9] Thus, these experiments offer the possibility, at least in principle, of a new approach to the study of spin correlations near magnetic critical points and in the ordered state of such materials. Thus far, however, only spin glasses and magnetic superconductors have been studied experimentally by this technique.[3,7] The standard theoretical model used to describe the required spin-flip relaxation time is that of de Gennes and Friedel,[10] in which the itinerant electrons of interest are coupled by s-d (or s-f) exchange interactions to localized spins. This model has also been widely used to discuss transport properties, such as electrical resistivity, at various levels of approximation.[10-20]

The first detailed theoretical discussion of itinerant electron relaxation times, as opposed to transport times, near the critical points of materials undergoing standard second-order phase transitions was given by Entin-Wohlman, Deutscher, and Orbach.[1] These authors calculated the spin-flip relaxation rate $1/\tau_{sf}$ to second order in the exchange coupling[21] and drew a number of conclusions concerning the expected temperature dependence of $1/\tau_{sf}$ near the critical point $t=(T-T_c)/T_c=0$. We have extended the work of Ref. 1 by taking into account the fact that the electrons have a finite mean free path, even in the absence of exchange coupling. This feature, which was emphasized by Fisher and Langer,[11] and extended by Richard and Geldart[14] in transport problems, turns out to have dramatic consequences for the scattering rate. The essential point is that even though electron scattering from phonons and static defects does not cause spin flips (in the usual model), all non-spin-flip scattering enters spin-flip transition rates even if the latter is treated in the Born approximation. The simplest way to take this into account is to write a Dyson equation for the electron self-energy

$$\Sigma(\mathbf{k},i\omega_n)=\Sigma_0(\mathbf{k},i\omega_n)+\Sigma_{sf}(\mathbf{k},i\omega_n) \ ,$$

where Σ_0 is due to all non-spin-flip processes, including scattering from static defects and phonons, Σ_{sf} is the spin-flip component of exchange scattering, and $\omega_n=(2n+1)\pi k_B T$ is the usual Matsubara frequency of finite-temperature perturbation theory. The spin-flip component of the self-energy is

$$\Sigma_{sf}(\mathbf{k},i\omega_n)=k_B T\sum_m \int \frac{d^3p}{(2\pi)^3}|j(\mathbf{k}-\mathbf{p})|^2 D_{\mathbf{k}-\mathbf{p}} \times (i\omega_n - i\omega_m) S^0_{\mathbf{p}}(i\omega_m) \ , \quad (1)$$

where j denotes Fourier components of the exchange interaction, D is the exact transverse (spin-flip) spin-correlation propagator,[21] and S^0 is the self-consistent electron propagator which includes Σ_0 fully but, in the spirit of a perturbation expansion in j, is independent of the exchange interaction. For initial simplicity, restrict attention to the paramagnetic state ($T>T_c$) and use the quasielastic approximation.[22] That is, the time scale for localized spin fluctuations near T_c is much larger than any other (electronic) time scales, so that $D_{\mathbf{k}-\mathbf{p}}(i\omega_n - i\omega_m)$ is sharply peaked about $\omega_m=\omega_n$. Then only $S^0_{\mathbf{p}}(i\omega_n)$ is important in Eq. (1) and, after analytic continuation to real frequencies, the spin-flip scattering rate is obtained from the imaginary part of the corresponding self-energy for electrons on the Fermi surface[23] ($p=k_F$, $\omega \approx 0$):

$$\Gamma_{sf}(\mathbf{k},\omega)=\tfrac{2}{3}\int \frac{d^3p}{(2\pi)^3}|j(\mathbf{k}-\mathbf{p})|^2 G(\mathbf{k}-\mathbf{p},T) A_0(\mathbf{p},\omega) \ , \quad (2)$$

where

$$G(\mathbf{q},T)=\sum_{\mathbf{R}} e^{-i\mathbf{q}\cdot\mathbf{R}}\langle \mathbf{S}_{\mathbf{R}}\cdot\mathbf{S}_0\rangle \quad (3)$$

is the equal-time (static) localized spin-correlation function.[21] The required itinerant electron spectral density is

$$A_0(\mathbf{p},\omega=0)=\frac{1}{\pi}\mathrm{Im}[S_0(\mathbf{p},\omega=0)] = \frac{1}{\pi}\frac{\Gamma_0(\mathbf{p})}{(E^0_{\mathbf{p}}-\mu)^2+[\Gamma_0(\mathbf{p})]^2} \ , \quad (4)$$

where $E_0(\mathbf{p})=\hbar p^2/2m+\Delta_0(\mathbf{p})$ and $\Delta_0(\mathbf{p})$ and $\Gamma_0(\mathbf{p})$ are the real and imaginary parts, respectively, of the non-spin-flip energy Σ_0, for $\omega=0$. At the present level of discussion we are not interested in details of quasiparticle renormaliza-

tion or dispersion relations, so we adopt the simplest approximation which exhibits finite lifetime properties. Thus, we replace $\Delta_0(\mathbf{p})$ and $\Gamma_0(\mathbf{p})$ by constants Δ_0 and Γ_0, taken to be equal to their respective values of the Fermi surface ($p \approx k_F$, $\omega \approx 0$); Δ_0 can be absorbed into the chemical potential μ and then forgotten, but the fact that Γ_0 is finite, due to phonon and impurity scattering, plays an important role in determining the overall structure of the temperature dependence of the spin-flip scattering rate, given by

$$1/\tau_{sf} = \Gamma_{sf}(k_F, 0)/\hbar \ ,$$

near the critical point. It is convenient to isolate the angular integration in Eq. (2) by introducing a factor $\langle \int d^3q\, \delta \times (\mathbf{k}_F - \mathbf{p} - \mathbf{q}) \rangle$, resulting in

$$1/\tau_{sf} = \frac{2}{3\hbar} \int \frac{d^3q}{(2\pi)^3} G(\mathbf{q}, T) |j(\mathbf{q})|^2 F(\mathbf{q}, \Gamma_0) \ , \qquad (5)$$

where, with $\epsilon_{\mathbf{p}} = \hbar^2\mathbf{p}^2/2m$, and $\langle \cdots \rangle$ denoting an angular average,

$$F(q, \Gamma_0) = \left\langle \int d^3p\, \delta(\mathbf{k}_F - \mathbf{q} - \mathbf{p}) \frac{\Gamma_0/\pi}{(\epsilon_{\mathbf{p}} - \mu)^2 + \Gamma_0^2} \right\rangle \ . \qquad (6)$$

As usual, we still replace $j(\mathbf{q})$ by a constant, as would be appropriate for very-short-range exchange interactions and take the Fermi surface (inside the first Brillouin zone) to be spherical.

The temperature dependence of $1/\tau_{sf}$ is a reflection of that of $G(q,t)$, which can be written in the various scaling forms such as $G(q,T) = t^{-\gamma} D(q/\kappa)$, where $\kappa = 1/\xi = \kappa_0 t^{\nu}$ is the inverse correlation length, which exhibits the susceptibility singularity since $D(0) = \text{const} \neq 0$. The q dependence of $F(\mathbf{q}, \Gamma_0)$ for small q is also important, as this also influences the relative importance of different regions of $\mathbf{q}$ in Eq. (5).

Consider first the limiting case of $\Gamma_0 \to 0$, which was studied in Ref. 1. Then Eq. (6) is just the elementary integral of a product of two delta functions and

$$F(q, \Gamma_0 = 0) = m/(2\hbar^2 k_F q)\Theta(2k_F - q) \ ,$$

where $\Theta(x) = 1$ (0) for $x > 0$ ($x < 0$) is the usual unit step function giving the Fermi-surface cutoff. The small q structure of Eq. (5) is then $\propto \int dq\, q^2 [t^{-\gamma} D(q/\kappa)]/q$, from which the temperature dependence is extracted as in Ref. 1 by scaling the q integration by κ to obtain a contibution to

$$1/\tau_{sf} \propto \kappa^2 t^{-\gamma} = t^{2\nu - \gamma} = t^{\eta\nu} \ ,$$

since $\gamma = (2 - \eta)\nu$.[24] This result is obviously sensitive to the evaluation of $F(\mathbf{q}, \Gamma_0)$, and we have emphasized that the finite conduction-electron lifetime, or mean free path, due to non-spin-flip scattering processes should *not* be neglected. To see this most clearly, set $q = 0$ in Eq. (6), which forces $p = k_F$, so the integration is trivial, giving

$$F(q = 0, \Gamma_0) = 1/(\pi\Gamma_0) \ . \qquad (7)$$

Thus the $q \to 0$ limit and the $\Gamma_0 \to 0$ limit cannot be freely interchanged, and for any physical situation with finite Γ_0 the q dependence of $F(q, \Gamma)$ is regular near $q = 0$. In this case, the small q structure of Eq. (5) is

$$\int dq\, q^2 [t^{-\gamma} D(q/\kappa)] 1/\Gamma_0 \ ,$$

but it is incorrect to attempt to extract a temperature-dependent contribution to $1/\tau_{sf}$ by scaling the q integration by κ (and neglecting details of the limit of integration) as before. For example, such a procedure would violate the "equal site" sum rule stating that the integral over the first Brillouin zone of $G(\mathbf{q}, T)$ is

$$\int \frac{d^3q}{(2\pi)^3} G(\mathbf{q}, T) = G(\mathbf{R} = 0, T) = S(S+1) \ , \qquad (8)$$

which is strictly independent of temperature.[14] Thus, the "apparently" singular contributions from the small-q structure of $G(\mathbf{q}, T)$ cancel completely in a correct calculation. Due to Eq. (7), a similar situation exists for the small-q contributions to $1/\tau_{sf}$, as is seen by rewriting Eq. (5) as

$$1/\tau_{sf} = \frac{2}{3\pi\hbar\Gamma_0} \int \frac{d^3q}{(2\pi)^3} G(\mathbf{q}, T) \tilde{F}(q, \Gamma_0) \ , \qquad (9)$$

where

$$\tilde{F}(q, \Gamma_0) = F(q, \Gamma_0)/F(0, \Gamma_0)$$

is exactly unity for $q = 0$ and is regular in its dependence for small q and finite Γ_0, so the apparently singular contributions at small q do not survive in Eqs. (5) or (9). To be explicit, we can split the region of integration in Eq. (9) into $q \lesssim \kappa$ and $q \gtrsim \kappa$, for example, and calculate the corresponding contributions $1/\tau_{sf}^{<}$ and $1/\tau_{sf}^{>}$. Using $\tilde{F}(q, \Gamma_0) = 1 + O(qk_F/\Gamma_0)$, we find

$$1/\tau_{sf}^{<} \propto t^{3\nu - \gamma}[1 + O(t^{\nu})]$$

by scaling the variable of integration. The sum rule, Eq. (8), tells us that the leading term due to $\tilde{F}(0, \Gamma_0) = 1$ must be canceled *exactly* by a corresponding term from $1/\tau_{sf}^{>}$ of opposite sign. The next correction of order $t^{4\nu - \gamma}$ is due to the deviations of $\tilde{F}(q, \Gamma_0)$ from unity at finite q and need not be canceled fully by $1/\tau_{sf}^{>}$. This exponent can be rewritten using $3\nu = 2 - \alpha$ as $1 - \alpha + (1 + \nu - \gamma) > 1 - \alpha$, and constitutes a weaker temperature dependence than the large-q $>> \kappa$ contributions which we now discuss. The contributions from large-q $>> \kappa$ will be different from those of Eq. (8) due to the departures of $\tilde{F}(q, \Gamma_0)$ from unity at large q and to differences in the volume of integration, which also affects large-q contributions. Fortunately, the temperature dependence of the large-q spin correlations is known from the observations of Fisher and Langer[11] and subsequent detailed calculations which give[25-27]

$$G(q, T) = G(q, T_c)[1 + C_1(q)t + C_2(q)t^{1-\alpha} + \cdots] \ , \qquad (10)$$

where

$$C_1(q) = -[1 + (\gamma - 1)/\alpha] X_q$$

and

$$C_2(q) = [(\gamma - 1)/\alpha] X_q^{1-\alpha} \ ,$$

with $X_q t = (\kappa/q)^{1/\nu}$. It follows that the spin-flip scattering rate near T_c is given by the large q (or short distance, $R << \xi$) correlations as

$$1/\tau_{sf} = A_0 + A_1 t + A_2 t^{1-\alpha} + \cdots \ . \qquad (11)$$

Estimates of A_1/A_0 and A_2/A_0 are obtainable from $C_1(q)$ and $C_2(q)$ for large $q \approx 2k_F$ for the material of interest, if desired.[28] However, we note the general features that $A_0 > 0$, while $A_1 \gtrless 0$ and $A_2 \lesssim 0$ for $\alpha \lesssim 0$. Thus, in addi-

tion to a generally increasing background due to non-spin-flip processes, the observed scattering rate due to spin-flip scattering from critical fluctuations is predicted to be monotonically increasing for $T \gtrsim T_c$ (irrespective of the sign of α). This is in contrast to the conclusions of Ref. 1 and is a direct consequence of the finite lifetime due to non-spin-flip scattering events.

The finite electron mean free path also plays an important role just below the critical point $T \lesssim T_c$ as well. The potential small-q contribution to $1/\tau_{sf}$, which varies as $|t|^{2\nu-\gamma}$, is still removed, of course, due to $\Gamma_0 \neq 0$ and the implications of Eq. (8). The next potential contribution described in Ref. 1 is proportional to the square of the magnetization $M(t) \propto |t|^{\beta}$. This arises if the long-range order is included in the electronic band structure. Then non-spin-flip exchange scattering from spin fluctuations would involve $\langle (S_R^z - \langle S_R^z \rangle)(S_0^z - \langle S_0^z \rangle) \rangle$ and, more importantly for the present discussion, the spin-flip scattering rate would involve $[M(t)]^2$ via its indirect appearance in the band structure [see Eq. (4)]. It has previously been emphasized that this separation of the long-range order in band structure introduces a type of mean-field approximation which should be avoided, as it is unreliable in the critical temperature region,[16,19] and a detailed discussion, including a derivation of the Boltzmann equation which describes the scattering events in the critical regime, has also been given.[20] The net result is that the above separation should not be made for $T \approx T_c$ to describe finite distance correlations and the magnetization squared does not enter $1/\tau_{sf}$. In other words, since the non-spin-flip mean free path is finite, it is again the short-distance ($R \lesssim$ mean free path $\ll \xi$) or large-q spin correlations which dominate $1/\tau_{sf}$. The spin-flip processes which determine $1/\tau_{sf}$ are described by

$$\tfrac{1}{2}\langle S_R^+ S_0^- + S_R^- S_0^+ \rangle = \langle S_R^x S_0^x + S_R^y S_0^y \rangle$$

for a Heisenberg model. These transverse correlations must be distinguished from the longitudinal correlations below T_c and, in addition to terms which carry the internal energy singularity, a new contribution appears which varies as $(-t)^{\psi}$ where $\psi = 2-\alpha-\phi$ and ϕ is the crossover exponent.[27] From the known ϵ expansions for α and ϕ, that for ψ is found to be[29]

$$\psi = 1 - \frac{2\epsilon}{(n+8)} + \frac{2(n^2+6n+14)\epsilon^2}{(n+8)^3} + O(\epsilon^3)$$

and a [1/1] Padé approximant yields $\psi = 0.846$ for $n=2$ (XY model) and 0.864 for $n=3$ (Heisenberg model). The ϵ expansion truncated at second order gives essentially the same results. We conclude that probably $\psi < 1$, in which case it is the *leading* singularity below T_c (recall that $1-\alpha > 1$). It should be noted that this contribution does not appear in the particular combination of longitudinal and transverse correlation functions which enters the electrical resistivity below T_c. These results, based on the quasielastic approximation for spin correlations, are not expected to be seriously modified by consideration of inelastic corrections.[18] Also, these conclusions concerning the dominance of short-range correlations apply to antiferromagnets and to other magnetic-order structures.

In summary, we have extended the work of Ref. 1 by including the effect of finite non-spin-flip scattering beyond the usual Born approximation while including spin-flip processes to lowest order. Short-distance spin correlations dominate the temperature dependence of $1/\tau_{sf}$ in the critical regime, just as they do for the electrical resistivity. The above detailed discussion has been given for magnetic systems which undergo a standard type of second-order phase transition. For such systems, the experimental study of $1/\tau_{sf}$ would be very interesting in view of the possibility of determining the crossover exponent. Although of considerable interest, such experiments would be very difficult and have not yet been carried out. Insofar as spin-glass transtions are certainly less will understood than second-order transitions, the eventual extension of the above theoretical discussion to spin glasses near the spin-glass transition temperature T_{sg} is desirable. It may be very tempting to conjecture that the temperature dependence of $1/\tau_{sf}$ due to quasielastic short-distance correlations will survive the configuration average required to describe spin glasses.[30] This could be consistent, of course, with the observed overall structure of the $1/\tau_{sf}$ which is obtained from dilute AgMn spin glasses in tunnel junctions,[3] but precise prediction of temperature dependence as a function of $T - T_{sg}$ will only be provided by a proper theory of spin glasses. In addition, the effect of inelastic scattering[6] over a broader temperature range, particularly in films with low values of T_{sg}, can also be included, but need not be a dominant effect near T_{sg}. Further experimental and theoretical work would be very useful for the development of this approach as applied to spin glasses and to other systems exhibiting exotic transitions, as well as to simpler magnetic materials.

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