Effect of inelastic scattering on resistive anomalies at magnetic critical points*

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Theoretical treatments of resistive anomalies at magnetic critical points have been based on the quasielastic approximation according to which the scattering system of localized spins is essentially static on the relevant time scale. This approximation is justified by thermodynamic slowing down of spin fluctuations near the critical point. In the case of ferromagnets, for example, this view is most obviously valid for long-wavelength spin fluctuations. These, however, are irrelevant for the resistivity. In this paper, we give an exact evaluation of the lowest-order corrections to the resistivity due to inelastic scattering for both ferromagnets and antiferromagnets. These corrections are numerically significant only for very low spin (e.g., $\sim 30\%$ for S = 1/2) and vary as 1/S(S + 1). Their temperature dependence reflects directly that of the internal energy of the spin system so that static critical properties will continue to be a useful guide in interpreting resistive anomalies.

I. INTRODUCTION

The study of electronic-transport properties (such as the electrical resistivity) at magnetic phase transitions has proven to be quite instructive. If we assume that the contribution $\rho_s(T)$ to the resistivity due to electrons being weakly scattering from essentially localized spins $\{\tilde{\mathbf{S}}_{R}\}$ located at lattice sites $\{\tilde{R}\}$, can be extracted from the total resistivity by some means such as Matthiessen's rule, then a study of the temperature dependence of $\rho_s(T)$ can yield fundamental information concerning the temperature dependence of correlations between spin fluctuations in the critical temperature regime.

Considerable progress has already been made in interpreting the wide variety of resistive "anomalies" observed in both ferromagnets and antiferromagnets (including binary alloys at their order-disorder transition).¹⁻¹³ For the most part, this work has been based on the simple model indicated above in which itinerant electrons are weakly coupled (by s-f exchange) to an array of localized spins and the scattering is treated in the first Born approximation.² Among the various approximations made (see also the following section) is the assumption that the electron-spin scattering is quasielastic. That is, the time scale of spin fluctuations is sufficiently long (relative to other relevant time scales) that correlation functions such as $\langle \vec{S}_{\vec{0}}(0) \cdot \vec{S}_{\vec{R}}(t) \rangle$, which enter the scattering cross section, may be regarded as stationary in time and replaced by $\langle \vec{S}_{\vec{0}}(0) \cdot \vec{S}_{\vec{R}}(t=0) \rangle$. This approximation is, of course, related to the thermodynamic slowing down of spin fluctuations near the critical point and its validity is most obvious in the case of long-wavelength spin fluctuations (this applies to ferromagnets, but analogous considerations hold in the case of antiferromagnets). However, the dominant contributions to the resistivity involve scattering with large momentum transfers⁵ (i.e., $q \sim 2k_F$) and it may not be obvious that the corresponding wavelengths are sufficiently small to invoke thermodynamic slowing down. Since the quasielastic approximation has played a central role in relating the temperature dependence of $\rho_s(T)$ to static critical properties of the spin system,^{3,5-13} it is necessary to investigate more closely its validity and it is this question which is addressed in the present work. The outline of the paper is as follows.

In Sec. II, we formulate an expression for $\rho_s(T)$ in which all inelastic effects are included and some previous work on the problem is discussed. In Sec. III, an exact result is given for the lowestorder inelastic correction and its temperature dependence is analyzed. Section IV consists of a summary of the conclusions.

II. SPIN-FLUCTUATION RESISTIVITY INCLUDING INELASTIC EFFECTS

In order to focus attention directly on the role of inelasticity, we shall make a number of simplifying assumptions [these assumptions are also made in most treatments of the quasielastic approximation to $\rho_s(T)$] and shall restrict attention to the paramagnetic state $(T \ge T_c)$. (i) The current carriers are described by a single isotropic band; the corresponding energies $(\epsilon_{\tilde{k}} = \hbar^2 \tilde{k}^2/2m^*)$ and velocities $(\tilde{v}_{\tilde{k}} = \hbar \tilde{k}/m^*)$ are spin independent. (ii) The coupling between itinerant and localized electrons is of very short range so that matrix elements for scattering are independent of wave number; i.e., $j_{s-f}(\tilde{k}, \tilde{k}') \approx \text{const.}$ (iii) Matthiessen's rule is assumed to be valid and the effects

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of the finite-electron mean free path will not be explicitly included. (iv) The scattering is described by a Boltzmann equation and an adequate approximation to the spin resistivity in the *i*th crystallographic direction is assumed to be given by the corresponding variational principle¹⁴ (the subscript s indicating the spin contribution will be dropped in the following for notational convenience)

$$\rho^i(T) \le \rho^i[\phi^i],\tag{1}$$

where

$$\rho^{i}[\phi^{i}] = \frac{\beta}{2} \sum_{\mathbf{k}\mathbf{k}'} Q_{\mathbf{k}\mathbf{k}'} f_{\mathbf{k}}(1 - f_{\mathbf{k}'}) (\phi_{\mathbf{k}}^{i} - \phi_{\mathbf{k}'}^{i})^{2} / \left(2e \sum_{\mathbf{k}} \phi_{\mathbf{k}}^{i} v_{\mathbf{k}}^{i} (\partial f_{\mathbf{k}}' / \partial \epsilon_{\mathbf{k}}) \right)^{2}$$

$$\tag{2}$$

in which the most elementary trial function is used, i.e., $\phi_{\mathbf{f}}^{i} = v_{\mathbf{f}}^{i}$. In Eq. (2), $\beta = 1/k_{B}T$, $f_{\mathbf{f}}$ is the Fermi occupation function and

$$Q_{\vec{\mathbf{k}}\vec{\mathbf{k}}'} = \frac{2\Omega_0 |j_{s-f}|^2}{\hbar^2} \sum_{\vec{\mathbf{k}}} e^{-i(\vec{\mathbf{k}}-\vec{\mathbf{k}}')\cdot\vec{\mathbf{k}}} \int_{-\infty}^{\infty} dt \ e^{i(\vec{\mathbf{k}}-\vec{\mathbf{k}}')t/\hbar} \langle \vec{\mathbf{S}}_{\vec{\mathbf{0}}}(0)\cdot\vec{\mathbf{S}}_{\vec{\mathbf{k}}}(t) \rangle, \tag{3}$$

where Ω_0 is the volume per ion and $\vec{S}_{R}(t)$ is the spin operator at site \vec{R} in the Heisenberg picture. To illustrate the desired points, it will be sufficient to consider only systems having a Bravais lattice and cubic symmetry (the index *i* denoting crystallographic direction can then be dropped).

In the high-temperature limit $[(T_c \ll T \ll T_F),$ where T_c and T_F are the Curie and Fermi temperatures, respectively], Eq. (2) simplifies very considerably and it is convenient to normalize $\rho(T)$ to the corresponding spin disorder resistivity ρ_0 where

$$\rho_0 = 3\pi S(S+1)\Omega_0 |j_{s-f}|^2 / e^2 \hbar v_F^2 .$$
(4)

To evaluate Eq. (2) more generally, introduce $\epsilon_{\vec{k}}$, $\epsilon_{\vec{k}}$, and $\vec{q} = \vec{k} - \vec{k}'$ as variables of integration. It is straightforward to obtain¹⁵

$$\frac{\rho(T)}{\rho_0} = \frac{12/(2k_F)^4}{S(S+1)} \int_0^{2k_F} dq \, q^3 \int \frac{d\hat{\Omega} \, q}{4\pi} G(\vec{q}, T), \quad (5)$$

where

$$G(\mathbf{\bar{q}},T) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{\beta \hbar \omega}{1 - e^{-\beta \hbar \omega}} A_{\omega}(\mathbf{\bar{q}},T) , \qquad (6)$$

with

$$A_{\omega}(\vec{\mathbf{q}},T) = \sum_{\vec{\mathbf{R}}} e^{i\vec{\mathbf{q}}\cdot\vec{\mathbf{R}}} \int_{-\infty}^{\infty} dt \ e^{i\omega t} \langle S_{\vec{\mathbf{p}}}^{s} S_{\vec{\mathbf{R}}}^{s}(t) \rangle.$$
(7)

The factor of $\beta \hbar \omega / (1 - e^{-\beta \hbar \omega})$ in Eq. (6) arises from taking into account the energy transfer $\hbar \omega$ in scattering events and the result is valid for $|\hbar \omega| \ll \epsilon_F$. If this factor is neglected, Eq. (6) reduces to an equal time correlation function and Eq. (5) becomes the usual quasielastic approximation

$$\frac{\rho_{QE}(T)}{\rho_0} = \frac{4}{(2k_F)^4} \int_0^{2k_F} dq \, q^3 \int \frac{d\hat{\Omega} q}{4\pi} \, \Gamma(\vec{q}, T) \,, \quad (8)$$

where $\Gamma(\vec{q}, T)$ is the Fourier (lattice) transform of $\Gamma(\vec{R}, T) = \langle \vec{S}_{\vec{d}} \cdot \vec{S}_{\vec{R}} \rangle / S(S+1)$. We may consider the structure of the static (equal time) spin correlation function to be reasonably well known, provided that we are willing to transcribe results of model calculations¹⁶⁻²⁰ to more complex systems of present interest where indirect exchange coupling may play a role (see Sec. III).

On the other hand, the structure of $A_{\omega}(\mathbf{\bar{q}},T)$ is extremely complex and involves both "local" dynamics²¹ as well as large scale critical effects. To isolate (partially) the various energy scales, it is convenient to write²²

$$A_{\omega}(\vec{\mathbf{q}},T) = \frac{\hbar\omega}{e^{\hbar\hbar\omega} - 1} \Phi_{\omega}(\vec{\mathbf{q}},T), \qquad (9)$$

where $\Phi_{\omega}(\mathbf{\tilde{q}},T) = \Phi_{-\omega}(\mathbf{\tilde{q}},T)$ is the Fourier transform of

$$\Phi_t(\vec{\mathbf{q}},T) = \frac{1}{N} \int_0^\beta d\lambda \langle S^{\boldsymbol{z}}_{\vec{\mathbf{q}}}(0) S^{\boldsymbol{z}}_{-\vec{\mathbf{q}}}(t+i\hbar\lambda) \rangle.$$
(10)

At this point, two avenues are suggested. One might introduce an approximation for the "relaxation function" $\Phi_{\omega}(\mathbf{q}, T)$ based on a specific model and then attempt to evaluate $ho(T)/
ho_0$ numerically or otherwise. For example, this was the approach of Mannari⁴ in his early considerations of the effect of inelasticity on $\rho(T)$. Mannari concluded that the inelasticity was rather important and that, when taken into account, $\rho'(T) = \partial \rho(T) / \partial T$ $\approx Ct^{-\lambda}$ with C > 0 and $\lambda \simeq \frac{1}{2}$ for $t = (T - T_c)/T_c \le 10^{-3}$ whereas the quasielastic approximation was thought to lead to an upward cusp in $\rho(T)$ at $T = T_c$ [so that $\rho'_{QE}(T) < 0$ for $T > T_c$]. Without wishing to detract from the value of Mannari's work, this conclusion is not completely correct and omissions can be traced to an inadequate treatment of certain aspects of $\Phi_{\omega}(\mathbf{q}, T)$ which, we emphasize, is a very delicate quantity.

As an alternative to adopting approximations for $\Phi_{\omega}(\vec{q}, T)$, we shall follow a second rather transparent procedure.²³ We assume initially that in-

elastic effects are small relative to the quasielastic contribution and calculate the lowest order inelastic correction exactly. The nature of the expansion parameter is then quite clear and some comments can also be made concerning higherorder inelastic corrections. This is done in Sec. III.

III. LOWEST-ORDER INELASTIC CORRECTIONS TO RESISTIVITY

From the derivation of $\rho_{QE}(T)$ [see Eq. (8)], it is clear that inelastic corrections involve an expansion in powers of frequency or, more precisely, an expansion in powers of the ratio [energy scale of $\Phi_{\omega}(\bar{\mathbf{q}}, T)$] to $k_B T$. To obtain a consistent expansion, combine Eqs. (6) and (9) in

$$G(\mathbf{\bar{q}},T) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{\beta \hbar \omega}{1 - e^{-\beta \hbar \omega}} \frac{\hbar \omega}{e^{\beta \hbar \omega} - 1} \Phi_{\omega}(\mathbf{\bar{q}},T), \quad (11)$$

and use

$$\frac{\beta(\hbar\omega)^2}{(1-e^{-\beta\hbar\omega})(e^{\beta\hbar\omega}-1)}=\frac{1-\frac{1}{12}(\beta\hbar\omega)^2+\cdots}{\beta}$$

to obtain

$$G(\mathbf{\bar{q}},T) = \frac{\Phi_{t=0}(\mathbf{\bar{q}},T)}{\beta} - \frac{1}{12}\beta \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} (\hbar\omega)^2 \Phi_{\omega}(\mathbf{\bar{q}},T) + \cdots$$
(12)

Similarly expanding

$$\frac{\hbar\omega}{e^{\beta\hbar\omega}-1}=\frac{1-\frac{1}{2}\beta\hbar\omega+\frac{1}{12}(\beta\hbar\omega)^2+\cdots}{\beta}$$

shows

$$G_{\mathbf{QE}}(\mathbf{\bar{q}},T) \equiv \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \, \frac{\hbar\omega}{e^{\beta\hbar\omega} - 1} \Phi_{\omega}(\mathbf{\bar{q}},T) \,, \tag{13}$$

to be given by

$$G_{QE}(\mathbf{\bar{q}}, T) = \frac{\Phi_{t=0}(\mathbf{\bar{q}}, T)}{\beta} + \frac{1}{12}\beta$$
$$\times \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} (\hbar\omega)^2 \Phi_{\omega}(\mathbf{\bar{q}}, T) + \cdots .$$
(14)

The first inelastic correction is thus

$$\Delta G(\mathbf{\tilde{q}}, T) = G(\mathbf{\tilde{q}}, T) - G_{QE}(\mathbf{\tilde{q}}, T)$$
$$= -\frac{1}{6}\beta (i\hbar)^2 \mathbf{\tilde{\phi}}_{i=0}(\mathbf{\tilde{q}}, T), \qquad (15)$$

since the integral in Eq. (12) or (14) is given by the second time derivative, at t = 0, of $\Phi_t(\bar{q}, T)$. The required derivative may, however, be easily evaluated. Using Eq. (10) and a Kubo identity,²⁴ we find

$$\mathring{\Phi}_{t}(\vec{\mathbf{q}},T) = (1/i\hbar N) \langle [S_{\vec{\mathbf{q}}}^{s}(t), S_{\vec{\mathbf{q}}}^{s}] \rangle, \qquad (16)$$

so that

$$\ddot{\boldsymbol{\Phi}}_{t=0}(\mathbf{\bar{q}},T) = [1/(i\hbar)^2 N] \langle [[S_{-\mathbf{\bar{q}}}^s,H],S_{\mathbf{\bar{q}}}^s] \rangle.$$
(17)

As suggested in Sec. II, we assume that the static critical properties of the spin system may be described by an effective Heisenberg Hamiltonian (having a contribution due to indirect exchange) of the form²⁵

$$H = -\sum_{\vec{n}\vec{n}'} J(\vec{R} - \vec{R}') \vec{S}_{\vec{R}} \cdot \vec{S}_{\vec{R}'} , \qquad (18)$$

with $J(\vec{R} - \vec{R'}) = 0$ for $\vec{R} = \vec{R'}$. The commutators in Eq. (17) are then easily evaluated to give

$$\Delta G(\vec{\mathbf{q}}, T) = -\frac{2}{3}\beta \sum_{\vec{\mathbf{R}}} J(\vec{\mathbf{R}}) \langle S_{\vec{\mathbf{0}}}^{\mathbf{g}} \cdot S_{\vec{\mathbf{R}}}^{\mathbf{g}} \rangle (1 - \cos \vec{\mathbf{q}} \cdot \vec{\mathbf{R}}).$$
(19)

The first inelastic correction to Eq. (8) is thus

$$\frac{\rho_I(T)}{\rho_0} = \frac{\rho(T) - \rho_{\mathbf{QE}}(T)}{\rho_0} = \frac{4}{(2k_F)^4} \int_0^{2k_F} dq \ q^3 \int \frac{d\hat{\Omega} q}{4\pi} \left(-\frac{2}{3}\beta \sum_{\mathbf{\vec{R}}} J(\mathbf{\vec{R}})\Gamma(\mathbf{\vec{R}}, T)(1 - \cos\mathbf{\vec{q}} \cdot \mathbf{\vec{R}}) \right). \tag{20}$$

It should be noted that this result is exact [subject, of course, to the use of the effective Hamiltonian, Eq. (18)] and applies to both ferromagnets and antiferromagnets.

Several points are apparent from Eq. (20). At high temperature, a perturbation expansion for $\Gamma(\vec{R},T)$ is appropriate and it is easy to see that $\rho_I(T)/\rho_0$ is of order $(T_c/T)^2$ and negative. Our present interest lies, of course, in the critical temperature range. For T near T_c , the temperature dependence of $\Gamma(\vec{R},T)$ dominates $\rho_I(T)/\rho_0$ and it is instructive to rewrite Eq. (20) as

$$\frac{\rho_I(T)}{\rho_0} = -\sum_{\vec{R}} J(\vec{R}) \Gamma(\vec{R}, T)_3^2 \beta f(R), \qquad (21)$$

where, with $\alpha = 2k_F R$,

$$f(\mathbf{R}) = \frac{4}{(2k_F)^4} \int_0^{2k_F} dq \, q^3 \int \frac{d\hat{\Omega} \, q}{4\pi} \left(1 - \cos \mathbf{\vec{q}} \cdot \mathbf{\vec{R}}\right)$$
$$= 1 - \frac{4}{\alpha^2} \left(-\cos \alpha + \frac{2}{\alpha} \sin \alpha - \frac{2}{\alpha^2} (1 - \cos \alpha)\right)$$

(22)

is non-negative and bounded. The similarity of this result to the internal energy per spin [see Eq. (18)],

$$\frac{U(T)}{N} = -\sum_{\vec{R}} J(\vec{R}) \Gamma(\vec{R}, T) S(S+1), \qquad (23)$$

makes it evident that the temperature dependence of $\rho_0(T)/\rho_0$ reflects that of the internal energy.

To gain some feeling for the magnitude of the inelastic corrections, several approaches are available. For example, consider initially the extreme case of nearest-neighbor interactions on a simple cubic lattice [i.e., $J(\vec{R})=J$ if $|\vec{R}|=a$ and zero otherwise]. Then

$$\rho_{T}(T)/\rho_{0} = \frac{2}{3}\beta f(a)U(T)/NS(S+1).$$
(24)

Estimates of the relevant quantities $[\beta U(T)/N \approx \frac{1}{2}$ for $T \approx T_c$,^{17,26} $f(a) \approx \frac{3}{4}$ for $2k_F a \approx \pi$] show that $\rho_I(T_c)/\rho_0 \approx -1/4S(S+1)$ which corresponds to a correction to the quasielastic result of about 2% for $S = \frac{7}{2}$ (as is appropriate for the rare-earthmetal ferromagnet gadolinium) or of some 30% for $S = \frac{1}{2}$.²⁷ For the rare-earth antiferromagnets, one finds $\rho_I(T_N)/\rho_0 \approx -1/4J(J+1)$, where the total ionic angular momentum is J = L + S.

It is also of interest to determine the contribution of inelastic corrections to the slope of the resistivity. As indicated above, it is to be expected that the critical temperature dependence of $\partial \rho_I(T)/\partial T$ reflects that of the heat capacity $C(T) = \partial U(T)/\partial T$. Within the simple-cubic nearestneighbor-interaction model, it follows from Eq. (24) that, apart from regular terms,

 $k_B T[\rho_I'(T)/\rho_0]/[C(T)/N] \approx \frac{2}{3} f(a)/S(S+1)$ $\approx [2S(S+1)]^{-1}.$

The corresponding ratio for the quasielastic contribution, estimated on the basis of a generalization of the Ornstein-Zernike form for $\Gamma(\vec{R}, T)$, is $\frac{2}{3}T_C/T_{C0}$, where T_{C0} is the mean-field estimate of the transition temperature, and differs significantly from the above only in that the $[S(S+1)]^{-1}$ factor is absent.⁹ The fact that the relevant expansion parameter describing inelastic corrections is $[S(S+1)]^{-1}$ is not surprising. It should also be emphasized that this conclusion holds for any reasonable effective spin-spin interaction, $J(\vec{R})$, and is not restricted to nearest-neighbor models. For example, we have explicitly verified, by a more lengthy analysis, that all of the above features, including the semiquantitative numerical estimates, remain valid if $J(\vec{R})$ is taken to be of Ruderman-Kittel-Kasuya-Yosida indirect-exchange origin.

IV. SUMMARY AND DISCUSSION

In Sec. III, it was seen that corrections to the spin-fluctuation resistivity due to inelastic scattering are of order $[S(S+1)]^{-1}$ relative to the corresponding inelastic values near the critical point and that the corrections are numerically significant only for low-spin systems. Also, the temperature dependence of $\rho_I(T)$ reflects that of the internal energy of the spin system just as in the case of the quasielastic contribution to the resistivity.^{5,6} These conclusions are fairly general and apply to both ferromagnets and antiferromagnets subject, of course, to their being adequately described by an effective Heisenberg Hamiltonian such as that given by Eq. (18). Note, in particular, that inelastic corrections to the resistivity of a binary alloy at its order-disorder temperature (the static properties of a binary alloy are formally equivalent to those of a $S = \frac{1}{2}$ Ising antiferromagnet) are not given by the above although they are expected to be minor.

It is particularly important that the temperature dependence of the singular part of $\rho'(T)$ reflects that of the singular part of the specific heat. Physically, this is due to the fact that relatively short-range (on a microscopic scale) spin fluctuations are being sampled, even in the critical temperature range, and that local "energetics" are involved. In view of the above conclusions, we feel that static critical properties will continue to be a useful guide when attempting to interpret the detailed temperature dependence of resistive anomalies in simple magnetic systems and that, where necessary, corrections can be made with some confidence.

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in terms of such "relaxation functions," see, W. Marshall and S. W. Lovesey, *Theory of Thermal Neutron Scattering* (Oxford U.P., Oxford, 1971).

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