Thermopower near a critical point*

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Previous calculations of the effect of a critical point on the thermopower of metallic ferromagnets are extended to include inelastic scattering processes. Denoting the (temperature-dependent) localized spin-spin correlation function by $g_{\vec{k}}(\omega)$, where \vec{k} is the momentum transfer, and ω the frequency transfer, the thermopower is shown to be proportional to frequency integrals of $g_{2k_F}(\omega)$ and frequency and wave-vector integrals of $kg_{\vec{k}}(\omega)$. Only if the spin-spin correlation function exhibits critical "slowing down" for all frequencies at all \vec{k} 's (an unlikely prospect) does our result reduce to those found previously.

I. INTRODUCTION

The critical scattering of conduction electrons by localized moments undergoing a transition from the paramagnetic to the ferromagnetic or antiferromagnetic state was first examined by de Gennes and Friedel.¹ They computed the impact of critical fluctuations on the electrical resistivity ρ_c using an Ornstein-Zernike form for the localized spin-spin correlation function $g_{\vec{k}}(t=0)$, where \vec{k} is the momentum transfer. The conduction electrons are assumed to interact with the localized spins via isotropic exchange. Subsequently, Fisher and Langer² corrected their treatment for ferromagnets by using a more accurate form for $g_{k}(t=0)$ at large momentum transfer. They found ρ_c $(T > T_c)$ to vary as $t^{1-\alpha} [t = (T - T_c)/T_c]$, in general, smoothly varying at T_c . A number of authors³ have treated antiferromagnetic metals with similar results.

More recently, Thomas *et al.*⁴ and Zorić *et al.*⁵ considered the behavior of the thermoelectric power in the vicinity of T_c . They predicted that⁵

$$\rho Q/T = A\rho_n + B\rho_c + C\Gamma(2k_F, T), \qquad (1)$$

where ρ is the resistivity, Q is the thermopower (which we designate by S), ρ_n is the normal resistivity, $\Gamma(\vec{k}, T)$ is the (localized) spin-spin correlation function [which we designate by $g_{\vec{k}}(t=0)$], with A, B, and C constants. These authors used an expression for S (e.g., Mott and Jones⁶) which is appropriate only to elastic scattering. Because $g_{\vec{k}}(t=0) = \int g_{\vec{k}}(\omega) d\omega$, this would require $g_{\vec{k}}(\omega)$ $= g_{\vec{k}}(0)\delta(\omega)$ for all \vec{k} at the critical point. We do not think there are many (if any) examples of systems where this limiting condition is applicable, and have therefore extended their calculation to include inelastic scattering processes. We have utilized the effective relaxation rate approach of Kubo, Yokota, and Nakajima.⁷ Although this method is approximate,⁸ the form of the solution suggests that it is sufficiently accurate for this level of investigation into the thermopower problem. In sum, the method of Ref. 7 generates an expression for S of the form

$$S_{\mu\nu} = \frac{1}{\sigma_{\mu\nu}T} \int_0^\infty dt \int_0^\beta d\lambda \left\langle Q_\nu J_\mu \left(t + i\hbar\lambda\right) \right\rangle, \tag{2}$$

where $\sigma_{\mu\nu}$ is the μ , ν component of the conductivity tensor; $\beta = 1/k_BT$; Q_{ν} and J_{μ} are the ν and μ component of the heat and electrical currents, respectively; and $\langle \cdots \rangle$ denotes thermal average. We find the double integral in (2) to be proportional to

$$(k_B T)^2 k_F^3 \int_{-\infty}^{\infty} d\omega (\beta \hbar \omega) (e^{\beta \hbar \omega} - 1)^{-1} \\ \times \left(\frac{k_F}{E_F} g_{2k_F}(\omega) + \frac{3(\beta \hbar \omega)^2}{8\pi^2 k_F E_F} \int_{0}^{2k_F} dk \, kg_k(\omega) \right).$$
(3)

Only if $g_{\overline{k}}(\omega) = g_{\overline{k}}(0) \delta(\omega)$, does (3) reduce to (1). This amounts to a requirement that all frequencies are critically "slowed down" in the vicinity of the critical point. This approximation is made in the original Letter by Fisher and Langer,² but with the proviso, "Owing to the 'thermodynamic slowing down' of critical fluctuations this is plausible for low wave numbers \vec{k} , but it may bear further investigation for higher values of \vec{k} which we claim are also important." While it is true that little information is available concerning the behavior of $g_{\mathbf{k}}^{\star}(\omega)$ for large momentum transfers, and all of this stems from experiment,⁹ what little there is does not point to such a limiting condition. Indeed, it appears that the large-momentum-transfer "characteristic frequencies" are little affected as one passes through T_c .⁹ We are therefore not in agreement with the thrust of Refs. 4 and 5 that thermopower measurements of metallic ferro-

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magnets near T_c can yield information concerning the static correlation function $g_{2k_F}(\omega=0)$.

Inserting (3) into (2), and including "background" contributions to both the thermopower S and the resistivity, we find

$$S \cong S_n(1 - \tilde{\tau}_n/\tilde{\tau}_c). \tag{4}$$

Here, S_n is the thermopower arising from noncritical scattering related processes (e.g., the phonon drag contribution to S), $\tilde{\tau}_n$ the "effective lifetime" for background processes which one would use in (2) (see Sec. II), and $\tilde{\tau}_c$ the effective lifetime to be used in (2), proportional to the inverse of (3). For spherical Fermi surfaces, we find $\tilde{\tau}_n = 6\tau$, where the conductivity $\sigma = Ne^2\tau/m$. With these identifications, our expression (4) correctly reduces to (1) if we force $g_k^-(\omega) = g_k^-(0) \delta(\omega)$.

According to Refs. 4 and 5, the measured thermopower does reasonable "track" τ (including, of course, the critical scattering contributions to ρ) as T passes through T_c . This may imply, therefore, that the frequency integrals in (3) vary smoothly through T_c . While we do agree with Refs. 4 and 5 that the departure of S from the behavior of τ through T_c gives information about $g_{\bar{k}}(\omega)$, this occurs only through the integral relationship (3). The detailed behavior of $g_{\bar{k}}(\omega)$ may therefore be rather more difficult to extract from measurements of S than suggested in Refs. 4 and 5.

We evaluated (2) in Sec. II using the approximation scheme of Kubo *et al.*⁷ adapted to the critical scattering problem. Our results are discussed in Sec. III.

II. CALCULATION OF THE THERMOPOWER

According to Mott and Jones,⁶ Wilson,¹⁰ and Ziman,¹¹ the thermopower S is given by

$$S_{\mu\nu} = -g_{\mu\nu}/e = (-\rho_{\mu\nu}g^{(2)}_{\mu\nu} + E_F/e)/T, \qquad (5)$$

where *e* is the electronic charge, and $\mathcal{G}^{(2)}_{\mu\nu}$ is a transport integral. A convenient form for $\mathcal{G}^{(2)}_{\mu\nu}$ is given by Kubo *et al.*⁷

$$\mathcal{G}_{\mu\nu}^{(2)} = \int_0^\infty dt \, \int_0^\beta d\lambda \left\langle Q_\nu J_\mu \left(t + i\hbar\lambda\right)\right\rangle,\tag{6}$$

where the symbols are defined after (2). In the same notation,

$$\frac{1}{\rho_{\mu\nu}} = \sigma_{\mu\nu} = \int_0^\infty dt \int_0^\beta d\lambda \langle J_\nu J_\mu (t + i\hbar\lambda) \rangle.$$
 (7)

Setting,

$$Q_{\nu} = \frac{\hbar}{m} \sum_{\substack{\mathbf{q},\sigma}} E_{\mathbf{q}} q_{\nu} a_{\mathbf{q}\sigma}^{\dagger} a_{\mathbf{q}\sigma}^{\dagger},$$

$$J_{\nu} = \frac{-e\hbar}{m} \sum_{\substack{\mathbf{q},\sigma}} q_{\nu} a_{\mathbf{q}\sigma}^{\dagger} a_{\mathbf{q}\sigma}^{\dagger};$$
(8)

we are able to simplify (3) as

$$S = \frac{1}{\sigma_{\mu\nu}T} \int_{0}^{\infty} dt \int_{0}^{\beta} d\lambda \left\langle -\frac{\hbar}{m} \sum_{\dot{q},\sigma} \left(E_{\dot{q}}^{\star} - E_{F} \right) \right. \\ \left. \times q_{\nu} a_{\dot{q},\sigma}^{\dagger} a_{\dot{q},\sigma} J_{\mu} \left(t + i\hbar \lambda \right) \right\rangle.$$
(9)

This formal treatment is exact. To proceed further, we approximate the correlation function in (6). We follow Ref. 7, and set

$$S_{\mu\nu}\sigma_{\mu\nu}T = \int_{0}^{\beta} d\lambda \langle Q_{\nu}J_{\mu}(i\hbar\lambda)\rangle \int_{0}^{\infty} dt \,\phi_{\mu\nu}(t), \qquad (10)$$

where

$$\phi_{\mu\nu}(t) = -\left[\phi_{\mu\nu}(0)\right]^{-1} \int_{0}^{B} d\lambda \left\langle \tilde{Q}_{\nu} J_{\mu}(t+i\hbar\lambda) \right\rangle$$
(11)

and

$$\phi_{\mu\nu}(0) = \int_0^\beta d\lambda \langle Q_\nu J_\mu(i\hbar\lambda) \rangle.$$
 (12)

Here, \bar{Q}_{ν} follows from (8) upon replacing $E_{\bar{q}}^{\star}$ by $E_{\bar{q}}^{\star} - E_F$. The integral in (11) is evaluated in second order in the perturbing Hamiltonian

$$H_{s-a} = -\frac{1}{N} \sum_{\dot{q}, \dot{q}'} I_{\dot{q}'-\dot{q}} \left[(a_{\dot{q}'+}^{\dagger} a_{\dot{q}+}^{-} - a_{\dot{q}'+}^{\dagger} a_{\dot{q}+}^{-}) S_{\dot{q}-\dot{q}'}^{\sharp} + a_{\dot{q}'+}^{\dagger} a_{\dot{q}+}^{-} S_{\dot{q}-\dot{q}'}^{\dagger} + a_{\dot{q}+}^{\dagger} S_{\dot{q}-\dot{q}'}^{-} + a_{\dot{q}'+}^{\dagger} a_{\dot{q}+}^{-} S_{\dot{q}-\dot{q}'}^{-} \right],$$
(13)

where $I_{\mathbf{q}'-\mathbf{q}}^{\star}$ is the Fourier transform of the usual J_{s-d} exchange coupling integral. The coefficient $\varphi_{\mu\nu}(0)$ contributes to zeroth order in (13). We use as the zeroth-order Hamiltonian $H_0 = H_s + H_d$, where

$$H_{s} = \sum_{\bar{q},\sigma} E_{\bar{q}} a_{\bar{q}\sigma}^{\dagger} a_{\bar{q}\sigma}^{\dagger} a_{\bar{q}\sigma} \left(E_{\bar{q}} = \frac{\hbar^{2} q^{2}}{2m} \right),$$

$$H_{d} = -\frac{1}{N} \sum_{\bar{k}} J_{\bar{k}} \vec{S}_{\bar{k}} \cdot \vec{S}_{-\bar{k}} \quad \left(\vec{S}_{\bar{k}} = \sum_{j} e^{i\vec{k}\cdot\vec{R}_{j}} \vec{S}_{j} \right).$$
(14)

Here, H_s is the conduction electron Hamiltonian, and $a_{q\sigma}^{\dagger}$ and $a_{q\sigma}^{\dagger}$ are, respectively, the creation and annihilation operators of conduction electrons with wave vector \bar{q} and spin σ . H_a is the Heisenberg Hamiltonian for the localized spins $\bar{S}_{\bar{k}}$ with wave vector \bar{k} . Using (8) and (14) in (12), the diagonal element of $\phi_{\mu\nu}(0)$, averaged over μ , ν , can be evaluated

$$\overline{\phi}(0) = \frac{1}{3} \sum_{\mu} \phi_{\mu \mu}(0) = \frac{NV_c |e|}{6\hbar^2} k_F (k_B T)^2, \qquad (15)$$

where V_c is the volume of the unit cell, and N the number of electrons.

We now pursue the main thrust of this paper. We need to evaluate (11) to second order in (13). To accomplish this end, we adopt the method of Kubo *et al.*⁷ and calculate $\ddot{\phi}_{\mu\nu}(t)$ to second order. We can then write

$$\phi_{\mu\nu}(t) = \phi_{\mu\nu}(0) + \int_0^t dt_1 \int_0^{t_1} dt_2 \, \ddot{\phi}_{\mu\nu}(t_2). \tag{16}$$

A relaxation-time approximation⁷ sets

$$\frac{1}{2} \int_{-t_1}^{t_1} dt_2 \left(\frac{\ddot{\phi}_{\mu\nu}(t_2)}{\phi_{\mu\nu}(0)} \right) = -\frac{1}{\tilde{\tau}}, \tag{17}$$

independent of t_1 . Then

$$\begin{split} \phi_{\mu\nu}(t) &\cong \phi_{\mu\nu}(0) \left(1 - \int_{0}^{t} \frac{dt_{1}}{\tilde{\tau}} \right) \\ &= \phi_{\mu\nu}(0) (1 - t/\tilde{\tau}) \\ &\approx \phi_{\mu\nu}(0) e^{-t/\tilde{\tau}}. \end{split}$$
(18)

Inserting (18) into (10) completes our calculation. Clearly, many approximations and limiting procedures are involved in this process. These are discussed in Ref. 7 and extensively examined in Ref. 8. A detailed review of the arguments would be beyond the scope of this work. Our results are to some extent a justification of the method, for we find that they reduced to the conventional⁴⁻⁵ elastic scattering result when only zero energy transfers are allowed.

We calculate $\ddot{\phi}_{\mu\nu}(t)$ according to this procedure in Appendix A. Inserting into (17), we find

$$\frac{1}{\tilde{\tau}_{c}} = \frac{1}{3} \sum_{\mu} \delta_{\mu\nu} \frac{1}{\bar{\phi}(0)} \frac{e\beta}{(mN)^{2}} \sum_{\tilde{\mathfrak{q}}_{1}.\tilde{\mathfrak{q}}_{2}} |I_{\tilde{\mathfrak{q}}_{2}-\tilde{\mathfrak{q}}_{1}}|^{2} (q_{2\mu} - q_{1\mu}) [q_{1\nu}(E_{\tilde{\mathfrak{q}}_{1}} - E_{F}) - q_{2\nu}(E_{\tilde{\mathfrak{q}}_{2}} - E_{F})] f_{\tilde{\mathfrak{q}}_{1}}(1 - f_{\tilde{\mathfrak{q}}_{2}}) \\
\times \int_{-\infty}^{\infty} d\omega \ g_{\tilde{\mathfrak{q}}_{2}-\tilde{\mathfrak{q}}_{1}}(\omega) \delta\left(\frac{E\mathfrak{q}_{1} - E\mathfrak{q}_{2} - \hbar\omega}{\hbar}\right),$$
(19)

where we have introduced the subscript c to denote critical scattering. After some simplification,

$$\frac{1}{\bar{\tau}_{c}} = \frac{1}{\bar{\phi}(0)} \frac{e}{(mN)^{2}} \frac{1}{3} \sum_{\mu,\nu} \delta_{\mu\nu} \sum_{\bar{\mathfrak{q}}_{1},\bar{\mathfrak{q}}_{2}} |I_{\bar{\mathfrak{q}}_{2}-\bar{\mathfrak{q}}_{1}}|^{2} (q_{2\mu} - q_{1\mu}) [q_{1\nu}(E_{\bar{\mathfrak{q}}_{1}} - E_{F}) - q_{2\nu}(E_{\bar{\mathfrak{q}}_{2}} - E_{F})] f_{\bar{\mathfrak{q}}_{1}} (1 - f_{\bar{\mathfrak{q}}_{2}}) \\
\times \frac{1}{2} \int_{0}^{\infty} d\omega \left[g_{\bar{\mathfrak{q}}_{2}-\bar{\mathfrak{q}}_{1}}(\omega) \delta \left(\frac{E_{\bar{\mathfrak{q}}_{1}} - E_{\bar{\mathfrak{q}}_{2}} - \hbar\omega}{\hbar} \right) + g_{\bar{\mathfrak{q}}_{2}-\bar{\mathfrak{q}}_{1}}(-\omega) \delta \left(\frac{E_{\bar{\mathfrak{q}}_{1}} - E_{\bar{\mathfrak{q}}_{2}} + \hbar\omega}{\hbar} \right) \right]. \quad (20)$$

The inelasticity of the scattering process is clearly exhibited. The first term in the large square brackets describes the process of absorption of energy $\hbar\omega$ by the localized spin system from the conduction electrons, while the second corresponds to emission to the conduction electrons. Only when $g_{\mathfrak{q}_2-\mathfrak{q}_1}(\omega) \simeq \delta(\omega)$ will elastic scattering alone contribute to (20) [and thence result in (1)].

After considerable algebra, taking $|I_{\mathfrak{q}_2-\mathfrak{q}_1}|^2$ independent of momentum transfer, (20) can be simplified to

$$\frac{1}{\tilde{\tau}_{c}} = \frac{V_{c}^{2}}{\bar{\phi}(0)} \frac{|e|}{6\hbar^{3}} \frac{I^{2}}{4\pi^{4}} \int_{-\infty}^{\infty} d\omega \left(\beta\hbar\omega\right) (e^{\beta\hbar\omega} - 1)^{-1} \left(\frac{4\pi^{2}}{3} \frac{(k_{B}T)^{2}}{E_{F}} k_{F}^{4} g_{2k_{F}}(\omega) + \frac{1}{2} \frac{(\hbar\omega)^{2}}{E_{F}} k_{F}^{2} \int_{0}^{2k_{F}} dk \, kg_{k}(\omega) \right).$$
(21)

We now clearly see the contribution of $g_{\bar{k}}(\omega)$ at $|\bar{k}| = 2k_F$, as well as an integral of $g_{\bar{k}}(\omega)$ over all \bar{k} , to $1/\bar{\tau}_c$. The coefficient of the second term vanishes as $\omega \to 0$, and the first yields (1) directly in the limit that the frequencies for which $g_{2k_F}(\omega)$ is finite approach zero. Therefore, the elasticity condition is "applied" directly by $g_{\bar{k}}(\omega)$, and requires that all frequencies "slow down" for all \bar{k} for $T \to T_c$. We find such a requirement difficult to accept. It will be discussed in the next section.

Simplifying (21), and using (15), we find as our principal result,

$$\frac{1}{\overline{\tau}_{c}} = \frac{2V_{c} m k_{F} I^{2}}{3\pi^{2} N \overline{\hbar}^{3}} \int_{-\infty}^{\infty} d\omega \, (\beta \overline{\hbar} \omega) (e^{\beta \overline{\hbar} \omega} - 1)^{-1} \left(g_{2k_{F}}(\omega) + \frac{3}{8\pi^{2} k_{F}^{2}} (\beta \overline{\hbar} \omega)^{2} \int_{0}^{2k_{F}} dk \, k \, g_{k}(\omega) \right). \tag{22}$$

so that (23) becomes

The diagonal element of the thermopower tensor, averaged over μ , ν , is given by [see Eq. (10)],

 $S = \left[\overline{\phi}(\mathbf{0}) / T\sigma\right] \mathbf{\tilde{\tau}}.$ (23)

The effective scattering time $\bar{\tau}$ in the presence of background scattering is given by

$$1/\tilde{\tau} = 1/\tilde{\tau}_n + 1/\tilde{\tau}_c, \tag{24}$$

$$S = [\overline{\phi}(0)/T\sigma] [1/\overline{\tau}_{n} + 1/\overline{\tau}_{c}]^{-1}$$

$$\approx [\overline{\phi}(0)/T\sigma] \overline{\tau}_{n} (1 - \overline{\tau}_{n}/\overline{\tau}_{c})$$

$$= S_{n} (1 - \overline{\tau}_{n}/\overline{\tau}_{c}). \qquad (25)$$

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III. DISCUSSION

The result (25) for the thermopower of a ferromagnetic metal reduces to that of Refs. 4 and 5 if, as stated earlier, one takes $g_{\vec{k}}(\omega) = g_{\vec{k}}(0) \delta(\omega)$. Then, (25) becomes

$$S = S_n - S_n (4V_c m k_F / N\hbar^3) \tau I^2 g_{2k_T} (\omega = 0).$$
 (26)

When we identify Ref. 4's $K_0 = 3m/(\hbar k_F)^3$, and take $V_c^{-1} = k_F^3/2\pi^2$, Eq. (26) reduces to Eq. (5) of Ref. 4. We therefore have reason to believe that the difference between our principal result, (22), and that of Refs. 4 and 5, lies in the assumption of elasticity implicit in the latter.

The physical situation is not clear. Dynamic scaling is valid only in the small momentum transfer limit.¹² Beyond that regime, one cannot say anything about the correlation function $g_{\overline{k}}(\omega)$ rigorously. Experimentally,⁹ it does not appear that anything happens to the finite \overline{k} modes in the vicinity of T_c , so that we do not believe there is any significant slowing down of the characteristic frequencies associated with these modes. We would then argue that inelastic processes contribute significantly to S, and involve the entire range of frequencies excited for temperatures around T_c . For a strictly three-dimensional situation, this means that frequencies up to the localized spinspin exchange frequency will be important.

In such a situation, the first two terms in the integrand of (22) will be essentially constant over the range of integration. The first term in the large parentheses then integrates to $g_{2k_{\rm P}}(t=0)$, the equal-time spin-spin correlation function for momentum transfer $2k_F$. The temporal character of this result has been cogently argued for by Parks.¹³ The second term in the large parentheses integrates over frequency to $d^2g_{\vec{k}}(t)/dt^2|_{t=0}$, with a subsequent integration over momentum transfer \vec{k} . It is difficult to say any more about this term, except that we expect all momentum transfers to contribute, there being no reason why the small momentum transfers should dominate. The sum and substance of the form of both terms is that there is nothing that dynamic scaling theory can say about them. The momentum transfer is large, and we know of no argument concerning the behavior of the characteristic frequencies for such a momentum regime.

For a system exhibiting three-dimensional ordering at T_c , but composed of weakly interacting twoor one-dimensional layers or chains, the fluctuations of the localized spin system may have a considerably greater range than $k_B T_c$.¹⁴ It is then conceivable that the wave vectors appearing in (22) at T_c may be small, the fraction of excitations limited by $k_B T_c/\hbar\omega_{\rm ex}$, where $\omega_{\rm ex}$ is the exchange frequency. It may then be the case that, for all practical purposes, the scattering can be regarded as static (critical slowing down of the long-wave-length modes), and the result of Refs. 4 and 5 will be relevant. It would be of interest to explore such types of magnetic metals.

In summary, we have formulated the problem of the thermoelectric power S of a ferromagnetic metal. We have examined the scattering of the conduction electrons in the vicinity of the critical temperature T_c , and have shown how the inelastic processes contribute to S. We have derived a result for S which should be valid for any temperature. We have compared our result with the previous elastic-scattering-only calculation for these systems, and shown how ours goes over to that limit under the appropriate conditions. Finally, magnetic dimensionality effects have been shown to affect the thermopower, and to affect the character of energy transfer between the electronic and magnetic systems.

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APPENDIX

We calculate $\ddot{\phi}_{\mu\nu}(t)$ in this appendix. By construction,

$$\begin{split} \ddot{\phi}_{\mu\nu}(t) &= \left(\frac{i}{\hbar}\right)^2 \int_0^\beta d\lambda \left\langle Q_\nu [H, [H, J_\mu(t+i\hbar\lambda)]] \right\rangle \\ &= -\left(\frac{i}{\hbar}\right)^2 \int_0^\beta d\lambda \left\langle [H, Q_\nu] [H, J_\mu(t+i\hbar\lambda)] \right\rangle. \end{split}$$
(A1)

This expression will be calculated to second order in H_{s-d} . Each of the commutators will be first order in H_{s-d} , so that the thermal average need only be taken with respect to H_0 , given by (14). It is easy to show that $\ddot{\phi}_{\mu\nu}(t) = \ddot{\phi}^*_{\mu\nu}(t)$ so that $\ddot{\phi}(t)$ is real. One then can write

$$\begin{split} \vec{\phi}_{\mu\nu}(t) &= \frac{1}{2\hbar^2} \int_0^\beta d\lambda \, \langle [H, Q_\nu] [H, J_\nu(t + i\hbar\lambda)] + [H, J_\mu] [H, Q_\nu(-t + i\hbar\lambda)] \rangle \\ &= -\frac{e}{(mN)^2} \int_0^\beta d\lambda \sum_{\mathbf{\tilde{q}}_1, \mathbf{\tilde{q}}_2} |I_{\mathbf{\tilde{q}}_2 - \mathbf{\tilde{q}}_1}|^2 (q_{2\mu} - q_{1\mu}) [q_{1\nu}(E_{\mathbf{\tilde{q}}_1} - E_F) - q_{2\nu}(E_{\mathbf{\tilde{q}}_2} - E_F)] f_{\mathbf{\tilde{q}}_1}(1 - f_{\mathbf{\tilde{q}}_2}) \\ &\times \left[\langle \mathbf{\tilde{S}}_{\mathbf{\tilde{q}}_1 - \mathbf{\tilde{q}}_2} \cdot \mathbf{\tilde{S}}_{\mathbf{\tilde{q}}_2 - \mathbf{\tilde{q}}_1}(t + i\hbar\lambda) \rangle \exp\left(\frac{i}{\hbar} (t + i\hbar\lambda) (E_{\mathbf{\tilde{q}}_1} - E_{\mathbf{\tilde{q}}_2})\right) \right. \\ &+ \langle \mathbf{\tilde{S}}_{\mathbf{\tilde{q}}_1 - \mathbf{\tilde{q}}_2} \cdot \mathbf{\tilde{S}}_{\mathbf{\tilde{q}}_2 - \mathbf{\tilde{q}}_1}(-t + i\hbar\lambda) \rangle \exp\left(\frac{i}{\hbar} (-t + i\hbar\lambda) (E_{\mathbf{\tilde{q}}_1} - E_{\mathbf{\tilde{q}}_2})\right) \right] \,, \end{split}$$
(A2)

so that $\ddot{\phi}_{\mu\nu}(t) = \ddot{\phi}_{\mu\nu}(-t)$. Defining¹⁵

$$g_{\vec{\mathbf{q}}_{2}-\vec{\mathbf{q}}_{1}}(t) = \langle \vec{\mathbf{S}}_{\vec{\mathbf{q}}_{1}-\vec{\mathbf{q}}_{2}} \cdot \vec{\mathbf{S}}_{\vec{\mathbf{q}}_{2}-\vec{\mathbf{q}}_{1}}(t) \rangle , \tag{A3}$$

(A2) reduces to

$$\ddot{\phi}_{\mu\nu}(t) = -\frac{1}{(mN)^2} \sum_{\mathbf{\tilde{q}}_1, \mathbf{\tilde{q}}_2} |I_{\mathbf{\tilde{q}}_2 - \mathbf{\tilde{q}}_1}|^2 (q_{2\mu} - q_{1\mu}) [q_{1\nu}(E_{\mathbf{\tilde{q}}_1} - E_F) - q_{2\nu}(E_{\mathbf{\tilde{q}}_2} - E_F)] \\ \times g_{\mathbf{\tilde{q}}_2 - \mathbf{\tilde{q}}_1}(t) \exp\left[\frac{i}{\hbar} (E_{\mathbf{\tilde{q}}_1} - E_{\mathbf{\tilde{q}}_2})t\right] [2f_{\mathbf{\tilde{q}}_1}(1 - f_{\mathbf{\tilde{q}}_2})].$$
(A4)

We need to evaluate

$$\frac{1}{2} \int_{-t_1}^{t_1} dt_2 \left[\ddot{\phi}_{\mu\nu}(t_2) / \phi_{\mu\nu}(0) \right], \tag{A5}$$

according to (17). Introducing the Fourier transform of (A3), the time integral generates

$$\frac{\sin[(E_{\tilde{\mathfrak{q}}_1}-E_{\tilde{\mathfrak{q}}_2}-\hbar\omega)t_1/\hbar]}{(E_{\tilde{\mathfrak{q}}_1}-E_{\tilde{\mathfrak{q}}_2}-\hbar\omega)/\hbar}\,.$$

This is to be integrated over t_1 , according to (18), from zero to $t < \tilde{\tau}$. Unless the energy difference $E_{\vec{q}_1} - E_{\vec{q}_2} - \hbar \omega$ is sufficiently small (on a scale of $\hbar/\tilde{\tau}$), the integrand in (18) oscillates so wildly that the integral over t vanishes. If, however, the energy difference vanishes, then (A4) in (A5) is independent of t_1 , and we may let $t_1 \rightarrow \infty$, resulting in

$$\lim_{t_1\to\infty}\frac{\sin[(E_{\mathbf{\tilde{q}}_1}-E_{\mathbf{\tilde{q}}_2}-\hbar\omega)t_1/\hbar]}{(E_{\mathbf{\tilde{q}}_1}-E_{\mathbf{\tilde{q}}_2}-\hbar\omega)/\hbar}=\pi\delta((E_{\mathbf{\tilde{q}}_1}-E_{\mathbf{\tilde{q}}_2}-\hbar\omega)/\hbar).$$

Inserting (A4) into (17), and using this result, we find

$$\frac{1}{\tilde{\tau}_{c}} = \frac{1}{3} \sum_{\mu} \delta_{\mu\nu} \frac{1}{\bar{\phi}(0)} \frac{e\beta}{(mN)^{2}} \sum_{\mathbf{\tilde{q}}_{1}, \mathbf{\tilde{q}}_{2}} |I_{\mathbf{\tilde{q}}_{2}-\mathbf{\tilde{q}}_{1}}|^{2} (q_{2\mu} - q_{1\mu}) [q_{1\nu}(E_{\mathbf{\tilde{q}}_{1}} - E_{F}) - q_{2\nu}(E_{\mathbf{\tilde{q}}_{2}} - E_{F})] f_{\mathbf{\tilde{q}}_{1}}(1 - f_{\mathbf{\tilde{q}}_{2}}) \\
\times \int_{-\infty}^{\infty} d\omega g_{\mathbf{\tilde{q}}_{1}-\mathbf{\tilde{q}}_{2}}(\omega) \delta \left(\frac{E_{\mathbf{\tilde{q}}_{1}} - E_{\mathbf{\tilde{q}}_{2}} - \hbar\omega}{\hbar} \right).$$
(A6)

where we have used the subscript c to denote critical scattering. This result is reproduced as (19) in the text.

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