Branch-imbalance relaxation times in superconductors

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The interrelationship of τ_Q, τ_Q^*, and τ_R, all of which have been used by various authors to describe the relaxation of branch imbalance, is critically reviewed with emphasis on the tractable case of disequilibrium generated by low-voltage tunnel injection and on the physical basis for the differences among the various times.

A tunneling current from a normal metal into a superconducting film generates a potential difference V between pairs and quasiparticles in the superconductor.[1] The quasiparticle potential can be sensed by a second normal-metal film coupled via a tunnel barrier to the reverse side of the superconductor. Since the pair chemical potential remains constant throughout the superconductor, V is conveniently measured relative to a second probe (normal or superconducting, tunneling or metallic) coupled to the superconductor at a point far from the injection region. Tinkham and Clarke[2] (TC) and Tinkham[3] showed that

$$V = Q^*/2eN(0)g_{NS}(T) . \qquad (1)$$

Here, $g_{NS}(T)$ is the *measured* normalized conductance of the probe junction in the limit $eV \ll k_B T$, and $N(0)$ is the density of states for electrons of one spin. The quantity eQ^* is the net quasiparticle charge per unit volume, with

$$Q^* = \sum_k f_k q_k$$
$$= 2N(0) \int_\Delta^\infty (f_{k_>} - f_{k_<}) \, dE_k , \qquad (2)$$

where f_k is the occupation number of the state k, $k_>$ and $k_<$ refer to states of energy $E_k = (\Delta^2 + \epsilon_k^2)^{1/2}$ with $k > k_F$ and $k < k_F$, respectively, and

$$q_k \equiv \epsilon_k/E_K = u_k^2 - v_k^2$$
$$= \pm (E_k^2 - \Delta^2)^{1/2}/E_k = \pm N_s^{-1}(E_k) \qquad (3)$$

is the effective charge of a quasiparticle in the state k. In Eq. (3),

$$u_k^2(\epsilon_k) = v_k^2(-\epsilon_k) = \tfrac{1}{2}(1 + \epsilon_k/E_k) ,$$

and $N_s(E_k)$ is the normalized BCS density of states. (Here, and throughout the paper, we neglect all rounding of the BCS density of states.) Thus, Eq. (1) implies that Q^* can be considered to be a directly measurable quantity, apart from the usual material parameter $N(0)$.

One wishes to interpret measured values of $Q^*(I_{inj}, V_{inj}, T)$ in terms of an injection rate and an appropriate relaxation time. In their original work,

TC considered an injection rate $\dot{Q}_{inj}$ and relaxation time τ_Q of the quasiparticle number branch imbalance Q defined by

$$Q = \sum_{k_>} f_{k_>} - \sum_{k_<} f_{k_<}$$
$$= 2N(0) \int_\Delta^\infty N_s(E_k)(f_{k_>} - f_{k_<}) \, dE_k , \qquad (4)$$

which differs from Q^* in that it does not take account of the fractional effective charges. Subsequently, Schmid and Schön[4] (SS), and, later still, Eckern and Schön[5] (ES) calculated these (and other) effects using a description that involves electrons rather than quasiparticles. In this picture, the total injected current (without regard to its quasiparticle composition) is taken as the source of the effect, and the characteristic time τ_R is proportional to the ratio of V to this total current. More recently, Pethick and Smith[6] (PS) introduced an alternative approach using the quasiparticle description in which they consider the injection rate $\dot{Q}_{inj}^*$ and relaxation time $\tau_q = \tau_{Q^*}$ of the charge imbalance Q^*.

The purpose of this Comment is to make completely explicit the interrelationship among τ_Q, τ_{Q^*}, and τ_R, and their relation to experimentally measurable quantities. This is done by drawing together results from the literature, and discussing in parallel the physical basis for the different approaches. We conclude, in agreement with PS, that τ_{Q^*} is a more appropriate time than τ_Q and should supersede it, whereas τ_R reflects a different conceptual point of view. For clarity, we confine almost the entire discussion to the analytically tractable case of low-voltage injection, where $eV_{inj} \ll k_B T$. We also comment on the relation between the measured relaxation times and the quasiparticle lifetime.

The three schemes outlined above (respectively SS, PS, and TC) can be expressed as follows:

$$Q^* = (I_{inj}/e\Omega)\tau_R , \qquad (5)$$

$$Q^* = \dot{Q}_{inj}^* \tau_{Q^*} , \qquad (6)$$

and

$$Q^* = (Q^*/Q)\dot{Q}_{inj}\tau_Q , \qquad (7)$$

where Ω is the volume of the nonequilibrium region, and we have identified the $\delta Q_n^{\text{l.e.}}$ of PS with Q^*.

In Eq. (5), I_{inj} is the total electric current injected

$$I_{\text{inj}} = \frac{G_{NN}}{e} \int_\Delta^\infty N_s(E) [f(E - eV_{\text{inj}})$$

$$- f(E + eV_{\text{inj}})] \, dE \qquad (8a)$$

$$\rightarrow G_{NN} V_{\text{inj}} \left[2 \int_\Delta^\infty \left(-\frac{\partial f}{\partial E} \right) N_s(E) \, dE \right]$$

$$\equiv G_{NN} V_{\text{inj}} Y(T) \qquad (eV_{\text{inj}} \ll k_B T) \ . \qquad (8b)$$

Here, G_{NN} is the tunnel conductance with both metals in the normal state. Note that the Yosida[7] function $Y(T)$ is the same as g_{NS} for the ideal BCS case. We use the notation Y to avoid confusion with the *measured* g_{NS} of the probe junction which appears in Eq. (1).

In Eq. (6), $\dot{Q}_{\text{inj}}^*$ is the rate injection of net quasiparticle charge, shown by Pethick and Smith to be

$$\dot{Q}_{\text{inj}}^* = \frac{G_{NN}}{e^2 \Omega} \int_\Delta^\infty N_s^{-1}(E) [f(E - eV_{\text{inj}})$$

$$- f(E + eV_{\text{inj}})] \, dE \qquad (9a)$$

$$\rightarrow \frac{G_{NN} V_{\text{inj}}}{e \Omega} \left[2 \int_\Delta^\infty \left(-\frac{\partial f}{\partial E} \right) N_s^{-1}(E) \, dE \right]$$

$$\equiv \frac{G_{NN} V_{\text{inj}}}{e \Omega} Z(T) = \frac{Z}{Y} \frac{I_{\text{inj}}}{e \Omega} \qquad (9b)$$

$$(eV_{\text{inj}} \ll k_B T) \ .$$

Note that this $Z(T)$ is identical with the $f(T)$ of ^{3}He literature,[8] but we prefer a different notation to avoid confusion with the Fermi function.

In Eq. (7), $\dot{Q}_{\text{inj}}$ is the rate of injection of branch imbalance, found by Tinkham and Clarke[2] to be

$$\dot{Q}_{\text{inj}} = \frac{g_{NN}}{e^2 \Omega} \int_\Delta^\infty [f(E - eV_{\text{inj}})$$

$$- f(E + eV_{\text{inj}})] \, dE \qquad (10a)$$

$$\rightarrow \frac{G_{NN} V_{\text{inj}}}{e \Omega} \left[2 \int_\Delta^\infty \left(-\frac{\partial f}{\partial E} \right) dE \right]$$

$$\equiv \frac{G_{NN} V_{\text{inj}}}{e \Omega} [2 f(\Delta)]$$

$$= \frac{2 f(\Delta)}{Y} \frac{I_{\text{inj}}}{e \Omega} \qquad (eV_{\text{inj}} \ll k_B T) \ . \qquad (10b)$$

The reasons for considering the several schemes can be summarized as follows. Equation (5) has the advantage of defining τ_R in terms of a strictly measurable quantity $\zeta \equiv \Omega g_{NS} V / I_{\text{inj}}$. Equation (6) deals most straightforwardly with the central quantity Q^*. Equation (7) is appropriate if one focuses on the branch imbalance Q rather than the fractional-charge-weighted imbalance Q^*. Historically, Eq. (7) was the first approach used,[1-3] but here we support the proposal of PS that it be superseded by Eq. (6).

We consider in detail only the case of low injection voltage, $eV_{\text{inj}} \ll k_B T$, where the response Q^* (i.e., V) is linear in I_{inj}, and can be calculated explicitly. Then, the consistency of Eqs. (5), (6), and (7) requires the following relations between the various relaxation times and the measured quantity ζ:

$$\tau_R = \frac{Z}{Y} \tau_{Q^*} = \frac{Q^*}{Q} \frac{2 f(\Delta)}{Y} \tau_Q$$

$$= \frac{e \Omega Q^*}{I_{\text{inj}}} = 2 e^2 N(0) \left[\frac{\Omega g_{NS} V}{I_{\text{inj}}} \right] \qquad (11)$$

$$\equiv 2 e^2 N(0) \zeta \qquad (eV_{\text{inj}} \ll k_B T) \ .$$

[At higher injection voltages, the factors relating the times would have to be replaced by the more general forms from which they were derived, namely Z/Y by $e \Omega \dot{Q}_{\text{inj}}^* / I_{\text{inj}}$ and $2 f(\Delta)/Y$ by $e \Omega \dot{Q}_{\text{inj}}/I_{\text{inj}}$.] The ratio Z/Y, which determines τ_R/τ_{Q^*} for this low-voltage injection case, is a well-defined function of T, which reduces to 1 at T_c and to $k_B T/\Delta$ at low temperatures (see Appendix). It is plotted in Fig. 1.

To find the relation of τ_Q to τ_{Q^*} and τ_R, one must obtain a value for Q^*/Q, which depends on the distribution of nonequilibrium quasiparticles. For the present case of low-voltage injection, and especially near T_c where $\tau_R \approx \tau_{Q^*} \approx \tau_Q \gg \tau_E$, it seems plausi-
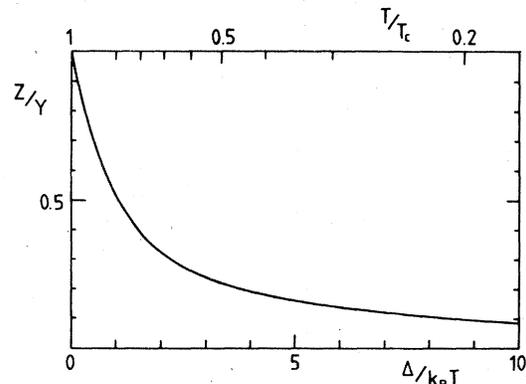

FIG. 1. Ratio Z/Y vs $\Delta/k_B T$. From Eq. (11), $Z/Y = \tau_R/\tau_{Q^*}$, for low injection voltages. The quantities Y and Z are defined in Eqs. (8b) and (9b), and discussed further in the Appendix.

ble that the energy distributions should be characterized by Fermi distributions with shifted chemical potentials. In the original Tinkham-Clarke work, it was assumed that the two branches of the quasiparticle spectrum were described by independent chemical potentials, such that E_k was measured relative to $\mu_>$ and $\mu_<$ for $k > k_F$ and $k < k_F$, respectively. In that case, it follows that

$$\delta f_> - \delta f_< = \left[-\frac{\partial f}{\partial E} \right] (\mu_> - \mu_<) \quad . \tag{12}$$

With this form for $(\delta f_> - \delta f_<)$, it is easy to show that $Q^*/Q = 2f(\Delta)/Y$. Thus for $eV_{inj} \ll k_B T$, one finds $\tau_Q/\tau_{Q^*} = Y(T)Z(T)/[2f(\Delta)]^2$, which ranges from 1 at $T = T_c$ to $\frac{1}{2}\pi$ at $T = 0$. However, in the more recent work of PS, it is argued that it is more physically reasonable to measure ϵ_k (rather than E_k) for both $k_>$ and $k_<$ states relative to a single shifted chemical potential μ, so that $E = [\Delta^2 + (\epsilon - \delta\mu)^2]^{1/2}$, where $\delta\mu$ is the change of μ relative to the pair chemical potential μ_s. This causes a change in occupation number

$$\delta f = \frac{\partial f}{\partial E} \frac{\partial E}{\partial \mu} \delta\mu = \left[-\frac{\partial f}{\partial E} \right] \frac{\epsilon}{E} \delta\mu \quad ,$$

so that

$$\delta f_> - \delta f_< = 2\left[-\frac{\partial f}{\partial E} \right] \frac{|\epsilon|}{E} \delta\mu \quad . \tag{13}$$

In Eq. (13), we have replaced $|\epsilon - \delta\mu|$ by $|\epsilon|$, since $\delta\mu$ is very small. A distribution of nonequilibrium electrons similar to Eq. (13) is also found in the work of Schmid and Schön, and in the numerical solution to the kinetic equation reported by Chi and Clarke[9] for the case of high-voltage injection. [Note that δf_E of SS is $(\epsilon/E)\delta f$ in our notation.] With the form given in Eq. (13), one finds $Q^*/Q = Z(T)/2f(\Delta)$, so that, from Eq. (11), at least in this limit of low-voltage injection, $\tau_Q = \tau_{Q^*}$ for all temperatures.[10]

In the more general case of $eV_{inj} \gtrsim k_B T$ and $\Delta \gtrsim k_B T$, there is no simple expression for Q^*/Q, although the computer result of Chi and Clarke supports the notion that $Z(T)/2f(\Delta)$ may be a good approximation quite generally. In view of this uncertainty, and because the case for using Eq. (13) seems persuasive, it seems preferable to simply abandon the τ_Q scheme in favor of the τ_{Q^*} scheme, as was suggested by Pethick and Smith, and hereafter to concentrate on a comparison of the τ_{Q^*} and τ_R approaches, where a real difference of viewpoint exists.

We have already seen that near T_c, where $\Delta \ll k_B T$, the difference between $I_{inj}/e\,\Omega$ and $\dot{Q}^*_{inj}$ and hence between τ_R and τ_{Q^*} is small. At lower temperatures the factor ϵ^2/E^2, which is different in the expressions for $\dot{Q}^*_{inj}$ [Eq.(9b)] and $I_{inj}/e\,\Omega$ [Eq.

(8b)], has the effect that $\dot{Q}^*_{inj}$ falls below $I_{inj}/e\,\Omega$ by the factor Z/Y plotted in Fig. 1. At low temperatures this ratio becomes $\approx k_B T/\Delta$ (see Appendix). As a result, the inferred value of τ_R will be a factor Z/Y lower than the inferred value of τ_{Q^*} for the same experimental data. This difference in definiton of τ_R and τ_{Q^*} stems from the conceptual difference in the description used by TC and PS compared to that used by SS. In the SS formulation, the *entire* injection current I_{inj} appears as a source for a quasiparticle charge (no factor ϵ^2/E^2). However, near the gap edge the charge is very rapidly converted into a supercurrent at a rate [Eq. (28) of Ref. 5]

$$\frac{1}{\tau_{conv.}} = \frac{2\Delta N_2}{N_1} \approx \frac{\Delta^2}{\epsilon^2} \frac{1}{\tau_E} \quad , \tag{14}$$

which diverges at the gap. (The notation here follows SS and ES. N_1 is the density of states and N_2 describes the quasiparticle-pair conversion as indicated by this equation. For further details, the reader should consult the original papers.[4,5,11]] Adding to this term the "scattering out" rate $\approx 1/\tau_E$, we find an energy-dependent rate $(E^2/\epsilon^2)/\tau_E$. Thus, while in the TC-PS picture the injection rate is reduced by a factor ϵ^2/E^2, in the SS approach the conversion rate is enhanced over the electron-phonon scattering rate by a factor E^2/ϵ^2. The resulting stationary quasiparticle distribution functions are the same in both approaches [Eq. (13)]. The difference seems to be mainly semantic, at least in the steady-state situation.

In practical experiments at low temperatures, the injection voltage is normally large compared with Δ/e in order that sufficient current be injected to give measurable branch imbalance voltages. In that case, both I_{inj} and $\dot{Q}^*_{inj}$ become nonlinear functions of V_{inj} as well as of T, and the inferred relation between τ_R and τ_{Q^*} also becomes a function of V_{inj} as well as T. A particularly illuminating example is found in the work of Chi and Clarke,[9] in which the injected current is provided by an S'-S tunnel junction. In that case, there is a step increase in I_{inj} when V_{inj} passes through $(\Delta + \Delta')/e$ but no corresponding change is observed in the measured probe voltage. Because the entire increase in injected current occurs at the gap edge, where the effective ϵ_k/E_k is zero, there is no change in $\dot{Q}^*_{inj}$ at the step, and hence the experimental result implies that τ_{Q^*} also holds constant through the step. By contrast, τ_R must decrease abruptly to reflect the constant observed V in the face of an abrupt increase in I_{inj}. This example shows that some gain in insight can be obtained by decomposing the Schmid-Schön τ_R into two factors as $\tau_R = F^* \tau_{Q^*} \equiv (e\,\Omega\,\dot{Q}^*_{inj}/I_{inj})\tau_{Q^*}$, since the first factor is readily calculated and leaves a value for τ_{Q^*} which is apparently relatively stable with respect to changes in injection conditions.

Having measured τ_{Q^*} or τ_R, one would like to determine a value for the quasiparticle lifetime, $\tau_E(T)$, due to electron-phonon collisions. In general, the inverse lifetime for a quasiparticle of energy E is the sum of the scattering and recombination rates

$$\tau_E^{-1}(T) = \tau_s^{-1}(E,T) + \tau_r^{-1}(E,T) \ . \tag{15}$$

In particular, one is interested in the normal-state lifetime at the Fermi energy and at T_c, $\tau_{E-0}(T_c)$. We can also relate $\tau_{E-0}(T_c)$ to the time τ_0 of Kaplan et al.[12] by using Eq. (18) of Ref. 12 and noting that, since $\tau_s^{-1}(0,T) = \tau_r^{-1}(0,T_c)$,

$$\tau_{E-0}^{-1}(T_c) = 7\zeta(3)\tau_0^{-1} = 8.4\tau_0^{-1} \ . \tag{16}$$

Very close to T_c, the relation between τ_{Q^*} or τ_R and $\tau_{E-0}(T_c)$ is practically independent of injection voltage[4,6,13]

$$(Y/Z)\tau_R = \tau_{Q^*} = 4k_B T \tau_{E-0}(T_c)/\pi\Delta \ . \tag{17}$$

At lower temperatures, the relation of a measured value of τ_{Q^*} or τ_R to $\tau_{E-\Delta}(T)$ depends on temperature and injection voltage, and, in general, must be determined by a computer calculation. Such a calculation[13] indicates that Eq. (17) is accurate to within $\pm 15\%$ for $eV_{inj} \lesssim 3\Delta(T)$ and $\Delta \lesssim k_B T$, an accuracy that is probably adequate for most practical purposes. The computations have also been extended to lower temperatures, but we emphasize that, in general, elastic impurity scattering in an anisotropic superconductor will make a substantial contribution to τ_R^{-1} or τ_Q^{-1}, particularly at low temperatures where it eventually dominates. Also, magnetic impurities or an external magnetic field, if present, can drastically reduce τ_R. Since the results given in this paper pertain to the case of inelastic phonon scattering only, they can be used only in the absence of such magnetic pair-breaking perturbations, and at temperatures high enough that the elastic scattering-anisotropy mechanism is dominated by the inelastic phonon one. This consideration combined with the need for a reliable quantitative theory implies that, in most practical situations, one can obtain reliable estimates of $\tau_{E-0}(T_c)$ only from data taken in the limit $\Delta \lesssim k_B T$.[14] If one bears in mind these limitations, the absence of phonon-trapping effects (which can be uncertain to a factor of ~ 2) gives this type of measurement a substantial advantage over measurements of effective recombination times or energy relaxation times as a

means of determining τ_E. Moreover, these measurements have intrinsic interest as a unique example of a type of transport phenomenon in which the *inelastic* scattering time is dominant, since (apart from gap anisotropy effects) elastic scattering is ineffective for relaxing branch imbalance.

ACKNOWLEDGMENTS

APPENDIX

We define two integrals, and give their asymptotic values:

$$Y(T) = 2 \int_\Delta^\infty N_s(E) \left[-\frac{\partial f}{\partial E} \right] dE$$

$$\rightarrow 1 - \frac{7\zeta(3)}{4\pi^2} \left[\frac{\Delta}{k_B T} \right]^2 \cdots \quad (\Delta \ll k_B T)$$

$$\rightarrow \left[\frac{2\pi\Delta}{k_B T} \right]^{1/2} e^{-\Delta/k_B T} \quad (\Delta \gg k_B T) \ ; \tag{A1}$$

and

$$Z(T) = 2 \int_\Delta^\infty N_s^{-1}(E) \left[-\frac{\partial f}{\partial E} \right] dE$$

$$\rightarrow 1 - \frac{\pi\Delta}{4k_B T} + \frac{7\zeta(3)}{4\pi^2} \left[\frac{\Delta}{k_B T} \right]^2 \cdots \quad (\Delta \ll k_B T)$$

$$\rightarrow \frac{k_B T}{\Delta} Y(T) \quad (\Delta \gg k_B T) \ , \tag{A2}$$

where $\zeta(3) = 1.202 \cdots$. Strictly speaking, Y and Z are functions of Δ/kT. They become functions of T if one takes the BCS form of $\Delta(T)$.

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[1]J. Clarke, Phys. Rev. Lett. **28**, 1363 (1972); J. Clarke and J. L. Paterson, J. Low Temp. Phys. **15**, 491 (1974).
[2]M. Tinkham and J. Clarke, Phys. Rev. Lett. **28**, 1366 (1972).
[3]M. Tinkham, Phys. Rev. B **6**, 1747 (1972).

[4]A. Schmid and G. Schön, J. Low Temp. Phys. **20**, 207 (1975).

[5]U. Eckern and G. Schön, J. Low Temp. Phys. **32**, 821 (1978).

[6]C. J. Pethick and H. Smith, Ann. Phys. (N.Y.) **119**, 133 (1979).

[7]K. Yosida, Phys. Rev. **110**, 769 (1958).

[8]R. Combescot and H. Ebisawa, Phys. Rev. Lett. **33**, 810 (1974).

[9]C.C. Chi and J. Clarke, Phys. Rev. B **19**, 4495 (1979).

[10]Near equality of τ_{Q^*} and τ_Q is also found in the numerical calculation of Chi and Clarke. Chang [Phys. Rev. B **19**, 1420 (1979)] shows that near T_c and for energies $E \gg kT$, the asymptotic energy-dependent rate $\tau_{\bar{Q}^*}^{-1}(E)$ exceeds $\tau_{\bar{Q}}^{-1}(E)$ by a factor of $\frac{1}{2}\pi$. However, at low energies he finds that $\tau_{\bar{Q}^*}^{-1}(E)$ is *less* than $\tau_{\bar{Q}}^{-1}(E)$. Averaged over the appropriate nonequilibrium distribution [Eq. (13)], this difference apparently cancels out. In this connection, we also make the following remark on the original calculation of τ_Q by Tinkham (Ref. 3). After correcting the identification of the parameter α by a factor of 2 (as noted earlier by Kaplan *et al.*) so that $\tau_{E-0}(T_c) = (8.4\alpha T_c^3)^{-1}$, the calculation of Ref. 3, which used the asymptotic form of $\tau_{\bar{Q}}^{-1}(E)$ for high E, gives $\tau_Q = (2T_c/\Delta)\tau_{E-0}(T_c)$. Since the asymptotic form of $\tau_{\bar{Q}^*}^{-1}(E)$ is simply larger by a factor of $\frac{1}{2}\pi$, the calculational procedure of Ref. 3 gives $\tau_{Q^*} = (4T_c/\pi\Delta)\tau_{E-0}(T_c)$, in precise agreement with the exact result of SS and others. Although this exact agreement for τ_{Q^*} must be considered fortuitous, the plots of Chi and Clarke comparing the exact and asymptotic form of $\tau_{\bar{Q}}^{-1}$ and $\tau_{\bar{Q}^*}^{-1}$ make it clear that the asymptotic formula should be a good approximation near T_c for $\tau_{\bar{Q}^*}^{-1}(E)$, but should definitely underestimate $\tau_{\bar{Q}}^{-1}(E)$ because of the singular behavior of the exact form near Δ. This singular behavior does not arise with $\tau_{\bar{Q}^*}^{-1}(E)$, since the use of fractional charges in the definition of Q^* eliminates the discontinuous structure of Q at Δ.

[11]The connection between the Green's function technique (SS) and the Boltzmann-equation approach (PS) is also discussed by O. Entin-Wohlman and R. Orbach, Phys. Rev. B **19**, 4510 (1979).

[12]S. B. Kaplan, C. C. Chi, D. N. Langenberg, J. J. Chang, S. Jafarey, and D. J. Scalapino, Phys. Rev. B **14**, 4854 (1976).

[13]C. C. Chi and J. Clarke, Phys. Rev. B (to be published).

[14]However, in the case of Al, where $\tau_{E-0}(T_c)$ is especially long, it is necessary to be much closer to T_c for the inelastic scattering to dominate (Ref. 9).