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Electron-electron scattering in dirty three-dimensional aluminum films

T. M. Klapwijk, P. A. van der Plas,* and J. E. Mooij
Department of Applied Physics, Delft University of Technology, Delft, The Netherlands

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Nonequilibrium superconductivity has been used to determine for the first time the electron-electron scattering rate in weakly disordered three-dimensional aluminum films at temperatures of about 1 K. This electron-electron rate strongly exceeds the independently measured electron-phonon rate for resistivity $\rho > 100$ nΩ m and is proportional to $l^{-3/2}$ as predicted by the Schmid theory. The absolute value exceeds the theoretical value by an order of magnitude.

According to the Landau theory of a Fermi liquid, electron-electron scattering in a metal is determined by the available phase space and therefore proportional to $(k_BT)^2/E_F$, with T the temperature, k_B Boltzmann's constant, and E_F the Fermi energy. This general result is strongly modified in the case of an impure metal. Schmid[1] has considered a system of electrons interacting by Coulomb forces and scattered by lattice defects. Assuming a weakly disordered metal ($k_Fl >> 1$), the temperature-dependent[2] electron-electron scattering rate is given by

$$\frac{\hbar}{\tau_{ee}}=\frac{\pi}{8E_F}(\pi k_BT)^2\left[1+0.67\left(\frac{1}{k_Fl}\right)^{3/2}\left(\frac{E_F}{k_BT}\right)^{1/2}\right] , \tag{1}$$

with k_F the Fermi momentum and l the elastic mean free path. For a pure metal ($l \rightarrow \infty$) the T^2 dependence is retained. The dependence on mean free path reflects the violation of strict momentum conservation in a dirty metal. As a consequence the temperature dependence weakens, the second term making a significant contribution for $k_Fl < 100$. For sufficiently short mean free paths the scattering rate is proportional to $T^{3/2}$ and inversely proportional to $l^{3/2}$. Subsequent theoretical work[3,4] has confirmed this result.

So far this theoretical prediction of enhanced electron-electron interaction in impure three-dimensional metals has not been tested experimentally. Information on inelastic scattering rates has been obtained from the magnetoresistance caused by weak localization and from nonequilibrium superconductivity. Analysis has been limited to thin films that are two dimensional from the point of view of electron-electron interaction. As in thin films the Coulomb interaction is screened less effectively than in bulk specimens; the theory must be modified. For films thin compared to the quantum diffusion length $L_T=[(\hbar D/k_BT)]^{1/2}$, with $D=v_Fl/3$ the diffusion constant, one finds[5]

$$\frac{\hbar}{\tau_{ee}}=\frac{e^2k_BTR_{sq}}{2\pi\hbar}\ln\left(\frac{2\pi\hbar}{e^2R_{sq}}\right) , \tag{2}$$

with $\tau=l/v_F$ the elastic scattering time and $R_{sq}=\rho/d$ the resistance per square of the film. Equations (1) and (2) predict a different dependence on mean free path and temperature as well as a difference in absolute value.[6]

It is widely accepted that in a metal inelastic scattering results from two processes: electron-electron and electron-phonon scattering. Various authors[7-9] have reported that for very thin films of aluminum and magnesium at low temperatures the inelastic scattering rate is proportional to T and R_{sq}. This shows, following Eq. (2), that electron-electron scattering dominates in two-dimensional films. In three-dimensional samples inelastic scattering has been studied by Mui, Lindenfeld, and McLean.[10] Their results are obtained for highly resistive samples of 3–60 $\mu\Omega$ m in the limit of strong disorder ($k_Fl << 1$).

In this paper an analysis is presented of inelastic scattering in three-dimensional samples that are weakly disordered ($k_Fl >> 1$). In this regime the theory for electron-electron scattering does apply. The inelastic scattering rate is measured by a method based on nonequilibrium superconductivity. The electron-phonon rate has been independently determined from the thermal impedance. It is found to be much smaller than the total rate, which is therefore attributed to electron-electron processes. This electron-electron scattering rate is proportional to $l^{-3/2}$, as predicted by Eq. (1), but the absolute value is larger by an order of magnitude.

Samples are made by e-gun evaporation of aluminum onto an oxidized silicon substrate. The elastic mean free path is shortened by evaporation in an oxygen atmosphere. Typically the samples are 200 μm long, 1–3 μm wide, and 50–400 nm thick. The mean free path l is determined from the resistivity at 4.2 K and a value of 4×10^{-16} Ω m^2 is used for ρl.[11] Parameters of the samples are given in Table I. Thickness of the samples has been determined with a diamond stylus. The critical temperature T_c is obtained from the resistive transition. Only samples with a narrow (a few mK) and smooth resistive transition have been used. The ratio d/L_T shows that six samples are three dimensional and three are in the transition region $d \simeq L_T$.

The inelastic scattering rate has been determined by a method described in more detail in Ref. 12. It is based on microwave-enhanced superconductivity. By applying microwaves to a thin superconducting film, a nonequilibrium distribution $f(E)$ is generated that is substantially different from the Fermi function $f_0(E)=[\exp(E/k_BT)+1]^{-1}$. The degree of nonequilibrium is inversely proportional to the inelastic scattering rate. The nonequilibrium distribution leads to an enhanced critical current I_c in narrow superconducting strips. Experimentally, I_c is determined from the current-voltage characteristic. The response of the critical current is recorded to application of a small amount of microwave power at a particular frequency. For $\omega < \omega_{min}$ the response is negative; I_c is depressed, whereas for $\omega > \omega_{min}$ the response is positive. The minimum frequency ω_{min} is higher for larger values of the inelastic scattering rate. In Ref. 12 an expression is given valid for tempera-

TABLE I. Parameters of the samples: d, thickness; $\rho_{4.2}$, resistivity at 4.2 K; T_c, critical temperature; L_T, quantum diffusion length; α, heat-transfer coefficient; τ_{in}, inelastic scattering time; $\tau_{e\text{-ph}}$, electron-phonon scattering time.

	Sample no.								
	1	2	3	4	5	6	7	8	9
d (nm)	615	240	334	118	45	64	245	136	305
$\rho_{4.2}$ (nΩ m)	43.7	43.8	73.2	85.9	88.2	88.9	147	209	346
T_c (K)	1.349	1.379	1.439	1.467	1.468	1.578	1.672	1.830	1.958
d/L_T	4.1	1.6	3.0	1.1	0.4	0.7	3.3	2.3	6.9
α (GW/m^{3}K)	...	11.4	12.1	...	16.3	21.0	22.8	22.9	31.7
τ_{in} (ns)	7.6	6.5	6.4	3.3	1.5	2.6	1.9	1.1	0.5
$\tau_{e\text{-ph}}$ (ns)	...	16.3	16.1	...	12.2	10.2	10.0	10.9	8.4

tures $T_c - T \ll T_c$, which relates ω_{min} to τ_{in}:

$$\omega_{min}\tau_{in}\frac{\hbar\omega_{min}}{k_BT_c}=\frac{\pi}{2}\left[\frac{k_BT_c}{3\Delta_c}\ln\left(16.9\frac{\Delta_c}{\Gamma_c}\right)+0.97\left(\frac{T_c-T}{T_c}\right)\frac{k_BT_c}{\Delta_c\Gamma_c}-G_h\right] . \quad (3)$$

Here Δ_c is the value of the energy gap at the critical current, $\sqrt{2/3}$ times the equilibrium gap at the measuring temperature. Γ_c is the pair breaking rate due to the supercurrent at the critical value and to inelastic scattering:

$$\Gamma_c=\frac{4}{3\pi}k_B(T_c-T)+\frac{\hbar}{2\tau_{in}} . \quad (4)$$

The second term is only important for relatively small scattering rates. In Eq. (3) G_h is a heating parameter which takes into account the increase of the average electron temperature above the bath temperature.[13] By measuring ω_{min} at various temperatures below T_c and fitting it to Eq. (3), one finds τ_{in}. Both G_h and τ_{in} are used as fitting parameters. Adjusting the inelastic scattering rate, including Eq. (4) iteratively, brings Eq. (3) in the right frequency range. As can be observed from Eq. (3), τ_{in} is the most important parameter. With the present samples we found that changing G_h by a factor of 2 changes the quality of the fit while changing the value of τ_{in} by less than 20%. Values of G_h are consistent with the heat-transfer coefficients discussed below. The measurements are performed near T_c yielding τ_{in} at one temperature only, $T = T_c$.

As discussed in Ref. 12, in deriving Eq. (3) analytic expressions that approximate results from the microscopic theory are used. On the basis of past experience, we have confidence in the reliability of the method to within a factor of 2. In particular, results on two-dimensional films obtained previously[12] agree rather well with those from weak localization as shown by Santhanam and Prober.[8] As far as the measuring method is concerned, no difference exists between two and three dimensions.

In Fig. 1 results on the scattering rates for three-dimensional samples are shown, as are the older results on two-dimensional samples (for details, see Ref. 12). Circles are two-dimensional films with thickness $d \ll L_T$ (range $0.3L_T$–$0.04L_T$). Squares are thick three-dimensional samples with $d \gg L_T$ (range $1.6L_T$–$6.9L_T$). Data are plotted as a function of resistivity. The two sets of data are clearly distinct. Because at the same resistivity for varying thickness a gradual transition from 3D to 2D behavior is expected, results obtained in the transition region $L_T \simeq d$ are also shown (triangles). With decreasing d/L_T a gradual shift towards 2D results occurs. Qualitatively, these results confirm the predictions of the theory for electron-electron scattering; in particular, the role played by the characteristic length L_T in grouping the data is suggestive. However, to allow an unequivocal identification of the measured inelastic scattering rate the electron-phonon rate must be known.

Theoretically, electron-phonon scattering in dirty metals has a complicated dependence on mean free path and temperature.[14] Unfortunately, an experimental check of these theoretical predictions is not yet available. Therefore we have experimentally determined the electron-phonon rate by an independent method described below.

In the investigated samples the voltage-carrying state for temperatures $T_c - T \gg 30$ mK is dominated by self-heating. After exceeding the critical current a normal hot spot is formed in the strip which balances Joule heating against heat conduction in the metal and heat transfer to the thermal bath.[15] In the current-voltage characteristic a minimum current is observed where with decreasing current the sample switches back into the superconducting state. At this current plateau the temperature of the normal hot spot is lowered to T_c. From a model calculation one finds that

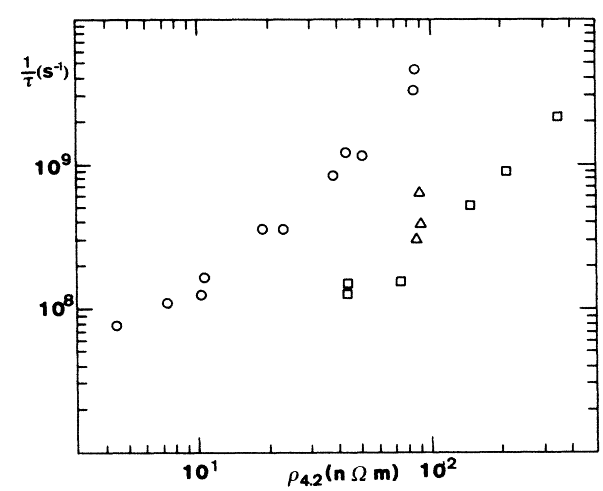

FIG. 1. Inelastic scattering rates as a function of resistivity. Three-dimensional samples (squares), two-dimensional samples from Ref. 7 (circles), and samples in the transition region (triangles).

the minimum current density $j_{\min}$ is given by

$$\rho j_{\min}^2 = K\alpha(T_c - T) \ , \tag{5}$$

independent of the heat conduction. T is the bath temperature and α the heat-transfer coefficient from the electrons to the thermal bath. K is a proportionality constant which depends on specific aspects of the model. In the original model proposed by Skocpol, Beasley, and Tinkham[15] Ohmic dissipation takes place only in a normal hot spot, while the superconducting regions in the strip are assumed dissipationless. In that case K equals $\sqrt{2}$. However, it is known that at a current-carrying normal-superconducting interface dissipation extends into the superconductor over a length of approximately the inelastic scattering length. This aspect has been included in the model by Stuivinga, Klapwijk, Mooij, and Bezuijen.[16] For parameters valid for aluminum films it is found that the minimum current is slightly reduced, equivalent to K about 1. In the following analysis $K=1$ has been used, the resulting error being at most 20%.

The measured heat-transfer coefficient α can be used as a measure of the electron-phonon scattering time. Little[17] has pointed out that the thermal impedance between the electron system and the thermal bath will be limited by electron-phonon relaxation if the temperature is low enough and/or the sample is sufficiently small. The heat transfer per unit volume can be represented by two thermal resistances in series:

$$\frac{1}{\alpha} = \frac{d}{Y_K} + \frac{\tau_{e\text{-ph}}}{c_v} \ , \tag{6}$$

d is the thickness of the film, Y_K^{-1} the Kapitza thermal boundary resistance, c_v the electronic heat capacity, and $\tau_{e\text{-ph}}$ the relaxation time for electron-phonon scattering. The first term represents the thermal resistance between phonons of the metal and phonons of the substrate and liquid helium. It is limited by acoustic mismatch. The second term describes heat transfer from the electrons to the phonons of the metal. The thickness d enters Eq. (6) because the first term is a surface effect, whereas the second term is a volume effect. For aluminum to superfluid helium we find from the literature[18] a Kapitza conductance of 1500 $\mathrm{W/m^2\,K}$, parallel to a conductance from aluminum to the silicon dioxide substrate of the same order of magnitude.[19] Consequently, the thermal resistance of thin films of aluminum is expected to be dominated by the second term of Eq. (6) for thicknesses below 0.5 μm. $j_{\min}$ is measured as a function of temperature and its dependence predicted by Eq. (5) is verified. The slope of $j_{\min}$ against the square root of temperature provides α.

Results on the heat-transfer coefficient α are listed in Table I. In addition, the electron-phonon scattering rate deduced from Eq. (6) is given. The Kapitza resistance has been ignored, because for these samples the second term in Eq. (6) dominates. We estimate that the value of $\tau_{e\text{-ph}}^{-1}$ is accurate to within a factor of 2. In Fig. 2 we present the inelastic scattering rate for the samples with $d/L_T > 1$ (dots) together with the electron-phonon rate (open triangles). For resistivities exceeding 100 nΩ m the electron-phonon rate is an order of magnitude below the total rate. It appears to increase slightly with increasing resistivity. We compare these rates with theoretical predictions for electron-phonon scattering in dirty systems by Keck and Schmid.[14] Since the measurements are restricted to a temperature range close to T_c, $\tau_{e\text{-ph}}$ is effectively determined at a temperature $T=T_c$ which increases with increasing resistivity. If the data are transformed into an isothermal plot at $T=1.35$ K based on the conservative assumption of a temperature dependence proportional to T^3, the results shown as closed triangles are obtained. The electron-phonon scattering rate decreases with increasing resistivity, a result theoretically anticipated in Ref. 14. Quantitatively, at a temperature of $T=1.35$ K, a scattering rate of about $5\times10^7\ \mathrm{s^{-1}}$ is predicted for $k_F l \simeq 100$ ($\rho=70$ nΩ m), which is in reasonably good agreement with the data. Santhanam and Prober[8] find $\tau_{e\text{-ph}}^{-1} = 1.6\times10^7\ \mathrm{T^3}$ in two-dimensional films, also in quite good agreement with the present data.

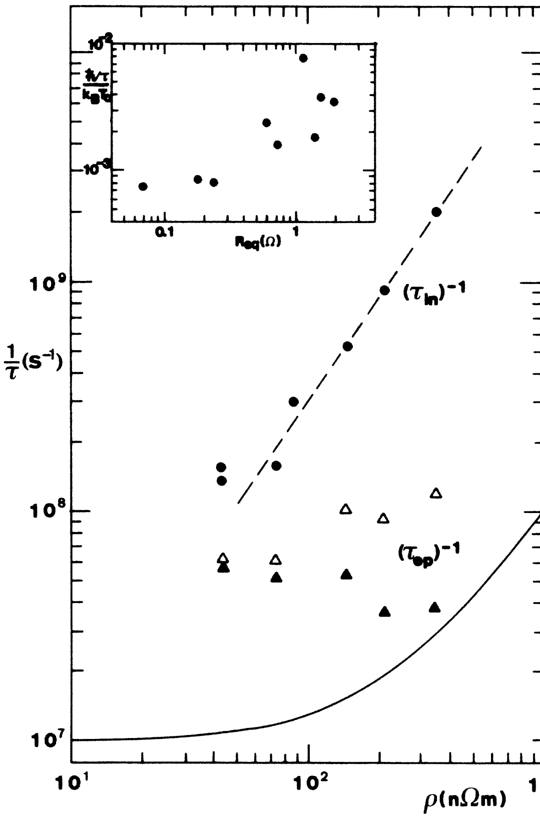

FIG. 2. Comparison of total inelastic scattering rate for three-dimensional samples (dots) with electron-phonon rate (open triangles, as measured; closed triangles, transformed to $T=1.35$ K). Full line, theory for electron-electron scattering, Eq. (1); dashed line, proportional to $\rho^{3/2}$. The inset shows the absence of correlation between the total scattering rate and R_{sq}.

Returning to the total inelastic scattering rate, Fig. 2 clearly demonstrates that electron-electron scattering is dominating. This permits comparison with the theoretical predictions of Eq. (1). In Fig. 2 the full curve represents Eq. (1) for $k_F = 1.75\times10^{10}\ \mathrm{m^{-1}}$, $E_F = 8$ eV, and $T=1.35$ K. Evidently, theory underestimates the scattering rate by more than an order of magnitude. Because Eq. (1) predicts a dependence proportional to $l^{-3/2}$ for shorter mean free paths, a dashed line is drawn proportional to $\rho^{3/2}$, in good agreement with the data. For comparison we have also shown (inset Fig. 2) the dependence on R_{sq} as suggested by Eq. (2). A correlation between $\hbar/\tau k_B T$ and R_{sq} is absent, in contrast to what is usually observed for two-dimensional films.[8,12]

We conclude that a three-dimensional electron-electron scattering rate has been observed and that the rate is much larger than predicted by Eq. (1). Interestingly, the latter conclusion has also been reached in experimental studies of the electron-electron contribution to the resistivity in very clean single-crystal specimens.[20] MacDonald, Taylor, and Geldart[21] have explained this discrepancy by electron-electron interaction mediated by virtual phonons.

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*Present address: Philips Research Laboratories, Eindhoven, The Netherlands.

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