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TEMPERATURE DEPENDENCE OF THE RELAXATION TIME OF THE SUPERCONDUCTING ORDER PARAMETER*

I. Schuller

Argonne National Laboratory, Argonne, IL 60439, U.S.A.

and

University of California, Los Angeles, CA 90024, U.S.A.

and

K.E. Gray

Argonne National Laboratory, Argonne, IL 60439, U.S.A.

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We have measured the temperature dependences of the relaxation time of the superconducting order parameter and of the equilibrium energy gap close to the transition temperature in very clean films of aluminum. The results are only consistent with the temperature and energy gap dependence predicted by Schmid and Schön. We also show that the magnitude and mean free path dependence of the electron inelastic collision time is in good agreement with calculations.

RECENTLY we have shown [1] that the relaxation time of the superconducting energy gap, Δ, diverges close to the transition temperature, T_c, in accordance with the theoretical prediction of several authors [2–6]. However, in those previous experiments, it was not possible to choose between the various theoretical models because the superconducting aluminum films had an anomalous temperature dependence of the equilibrium energy gap in the region of divergence. The equilibrium energy gap (i.e. order parameter) was proportional to $T_c - T$ in the region of divergence instead of the $(T_c - T)^{1/2}$ prediction of the BCS theory. As a result, the relaxation times agreed with the theory of Landau and Khalatnikov [2] and Woo and Abrahams [3] predicting a $(T_c - T)^{-1}$ divergence as well as the theory of Schmid and Schön [6] predicting a Δ^{-1} divergence.

We later found [7] that cleaner superconducting aluminum films obeyed the BCS temperature dependence for the energy gap much closer to T_c, so that a definitive test of the theories was possible. These very clean aluminum films (1000 Å) were evaporated by electron beam from 99.999% pure aluminum slugs. A liquid nitrogen cooled titanium sublimation pump and ion pump provided sufficient pumping to maintain a pressure of about 3×10^{-7} torr while evaporating at a rate of 150–200 Å sec^{-1}. Pre-evaporation at this rate, before opening a shutter provided additional getter pumping of residual oxygen in the system. The electron mean free path l in films made this way was size limited even at 1 μm film thickness, and can be obtained from the normal state resistivity ρ_N using $\rho_N l \sim 1.2 \times 10^{-11}$ Ω-cm, appropriate for polycrystalline aluminum films [8]. Note that these samples are still in the dirty limit required by the Schmid–Schön theory, i.e. $kT_c l \ll \hbar v_F$, where v_F is the Fermi velocity. The tunnel junction was completed by exposing the aluminum film to air and evaporating a tin (2500 Å) counter electrode.

The temperature dependence of the equilibrium energy gap was measured by tunneling, and the deviation from the prediction of the BCS theory was restricted to 1–2 mK below T_c as opposed to $\sim$ 7 mK in the previous experiments [1]. The relaxation time was measured in the same fashion as described in reference [1], except for the use of a different pulse amplifier which eliminated the small overshoot of the signal pulse. The signal to noise ratio was 5 in the worst case. The region of divergence is about 0.5–20 mK as observed in our earlier experiments.

Figure 1 shows a graph of log (τ) vs log $(T_c - T)$ as obtained from the present experiment. The dashed curve is the theoretical prediction of Schmid and Schön. If we ignore the 3 points at lowest temperatures, the experimental data in the region of divergence (0.5–20 mK) are in excellent agreement with predicted *temperature* dependence of Schmid and Schön and are not consistent with the theories of Landau–Khalatnikov and

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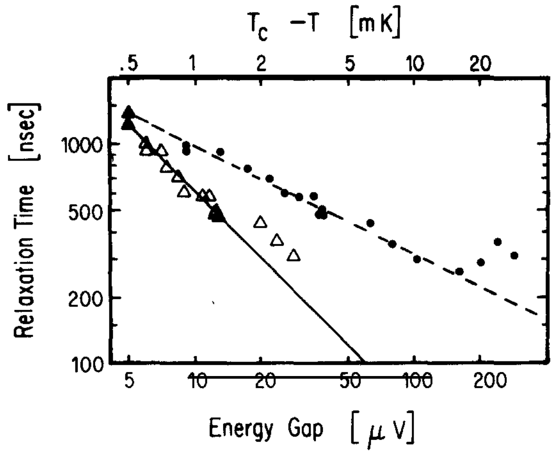

Fig. 1. Solid circles: relaxation time vs $T_c - T$. The dashed line of slope $-1/2$ shows the prediction of the Schmid–Schön theory. Open triangles: relaxation time vs experimentally measured energy gap. The solid line of slope -1 shows the theoretical prediction of Schmid–Schön. $T_c = 1.211$ K.

Woo–Abrahams. We have also included in Fig. 1 our data for log (τ) vs log (Δ), along with the theoretical prediction of Schmid and Schön. Like in our earlier experiments, the Δ^{-1} dependence of τ is obeyed even in the region of non-BCS behavior of the gap.

From the divergence of τ near T_c, we determine the parameter τ_E of the Schmid–Schön theory to be 16 nsec in these very clean films (electron mean free path $l \sim 1800$ Å), whereas we previously found [1] τ_E to be 7 nsec in films having $l \sim 700$ Å. Schmid and Schön define τ_E^{-1} to be the total inelastic scattering rate for electrons at the Fermi surface at T_c, and we will now show that our results are in excellent agreement with the calculation outlined in the following.

A calculation of the electron–phonon inelastic collision rate is given by Kaplan *et al.* [10], who use $\alpha^2(\omega)F(\omega)$ to estimate the electron–phonon coupling strength. They find that the total electron–phonon scattering rate for an electron at the Fermi energy at T_c is given by $7\Gamma(3)\zeta(3)/2\tau_0 \cong 8.4\tau_0^{-1}$, where τ_0 is a characteristic of the material [10] and equals 438 nsec for clean bulk aluminum ($l = \infty$ and $T_c = 1.19$ K). The gamma and zeta functions are $\Gamma(3)$ and $\zeta(3)$.

For aluminum, Schmid [9] has calculated the *relative* size of contributions to τ_E^{-1} from electron–phonon and electron–electron inelastic collisions and their dependence on electron mean free path. Knowing the electron–phonon scattering rate for infinite mean free path, we can evaluate the expected relaxation time τ_E^{calc} for each of our experiments using Fig. 4 of reference [9] and the measured mean free paths. We use the free electron Fermi momentum $k_F = 1.75 \times 10^8$ cm^{-1} and include the T_c^3 and T_c^2 dependences of the

Table 1. Relaxation time parameters for aluminum

	l (A)	$k_F l$	T_c (K)	$\tau_E^{\text{calc.}}$ (n sec)	$\tau_E^{\text{meas.}}$ (n sec)
Bulk	∞	∞	1.190	35	
Present work	1800	3150	1.211	10	16
Reference [1]	700	1225	1.321	5	7

electron–phonon and electron–electron coupling. The results are summarized in Table 1. (The effect of different T_c increases the calculated rate by 1/3 in our previous experiment [1] and by 5% in the present one.) We consider the approximate quantitative agreement to be very good, but even more encouraging is the dependence on electron mean free path.

The only other measurement of τ_E in aluminum comes from surface Landau-level resonance experiments [11]. These show values of 19, 145 and 150 nsec for three discrete points on the Fermi surface whereas the above calculation gives 35 nsec (Table 1). Since it is not clear how to average these times and the measurements reflect surface properties, perhaps not characteristic of bulk, a closer comparison may not be relevant.

The calculations of Kaplan *et al.* [10], indicate that the quasiparticle recombination time can also be related to τ_E. However, in this calculation, τ_r is the quasiparticle scattering time *across* the Fermi surface, and corresponds to quasiparticle recombination only at low temperatures. Near T_c, only those quasiparticles scattered into momentum states k, whose single particle energy $(\hbar k)^2/2m$ is within about Δ of the Fermi energy, can contribute to the condensation energy (however, even then, only if the opposite momentum state is already occupied). Hence, scattering across the Fermi surface is irrelevant and the recombination time is approximately [6] $\tau_E kT/\Delta$, since the average scattering time is roughly τ_E, and a fraction of only about Δ/kT of these events can form bound pairs contributing to the condensation energy. However, the calculations at low temperatures are valid and relate the measured τ to τ_E.

The laser pulse results (see also reference [1]) at temperatures below the region of divergence ($T/T_c \sim 0.95$) are a factor of 2.5 greater in the clean films, i.e. they scale with roughly the same factor as τ_E derived from the divergence (see Table 1). Furthermore, these results are in reasonable agreement with the magnitude of earlier steady state recombination time measurements [12]. Unfortunately, τ_E predicted from the low temperature steady state results [12] ($T/T_c \sim 0.4$) is about 30 times larger than that determined from the divergence near T_c in a film with comparable mean free path [1]. In the past it has been popular to blame phonon trapping [10, 12] for enhanced quasiparticle lifetime, however, a trapping factor of 30 is much larger than expected for aluminum [12, 13]. For example, the phonon escape probability α at the boundary with the liquid helium or substrate ($\alpha = 2 \times$ film thickness $\times$ phonon lifetime for pair breaking $\div$ trapping factor $\times$ sound speed) must be less than 0.05 for a trapping factor of 30.

Unfortunately, near T_c, the simple phonon trapping model [14] breaks down since phonon and quasiparticle states with energies many times 2Δ are occupied. However, near T_c, the number of excess phonons in these experiments (e.g. due to the laser pulse) is a small percentage of the number in thermal equilibrium and will not greatly affect the scattering time τ_E. For the same reason, phonon trapping should not influence the recombination time $(\tau_E T/\Delta)$ near T_c, so that the divergent part of the pulse laser experiment measures τ_E without enhancement. The abrupt transition between the divergent region with no phonon trapping and the non-divergent region apparently with large phonon trapping is not understood.

A relaxation time can also be derived from measurements of phase slip centers [15–17]. Although it is an unsettled question as to whether these experiments measure branch imbalance relaxation [15, 18] or quasiparticle recombination [19], reported values of about 100–200 nsec for aluminum [16, 17] are close to the τ (not τ_E) measured in the present experiments. Measurements near T_c have failed to reflect the divergence [15–17] and at present this is not understood.

It is interesting to note that liquid helium [20, 21] gapless superconductors [22] and superconductors above the transition temperature [23] show a temperature dependent relaxation time consistent with $(T_c - T)^{-1}$. The difference between superconductors with an energy gap and gapless systems is that there is an additional coupling of the excitations to the energy gap (order parameter), through changes in the density of states with Δ. This is accounted for in the Schmid–Schön theory but not in the Landau–Khalatnikov theory.

In conclusion, we feel that our results provide strong evidence for the temperature dependence of the relaxation time in superconductors predicted by Schmid–Schön [6]. We also present, we believe for the first time, a measurement of the magnitude and the mean free path dependence of the inelastic electron collision time τ_E, which agrees with theoretical predictions.

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Note added in proof: Leibowitz and Wilt [24] claim to have measured this relaxation time in indium and likewise find a $(T_c - T)^{-1/2}$ divergence.