Breakdown of Eliashberg Theory for Two-Dimensional Superconductivity in the Presence of Disorder

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We have performed tunneling measurements on ultrathin, homogeneous, strong-coupled superconducting Pb films in the regime where T_c varies with sheet resistance. The energy gap Δ and the strong-coupling corrections to the density of states have been measured as functions of the sheet resistance $R_\Box$. We conclude that the standard picture of enhanced Coulomb repulsion with increasing sheet resistance is too naive and that, in the presence of disorder, an analysis more sophisticated than Eliashberg theory is necessary.

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Superconductivity in the presence of disorder in two and three dimensions is of considerable intrinsic interest and offers the opportunity to investigate the nature of transport and Coulomb interactions in this limit.[1-9] If we write the superconducting pair wave function $\psi = \Delta e^{-i\phi}$, where Δ is the amplitude of the order parameter and ϕ is the phase, it is apparent that we can destroy superconductivity in two ways, either by breaking the phase coherence (ϕ) or by suppressing the amplitude Δ. The sample morphology plays an important role in determining which of these two regimes dominates. With use of thin-film deposition techniques, the two limiting cases can be studied: pure phase breaking[4] (the film consists of small ~30–50-Å grains weakly connected) or amplitude suppression (the film is microscopically homogeneous). In this Letter we report a study of the limit in which Δ is varied. This is accomplished via the deposition of ultrathin homogeneous films which are uniform down to thicknesses of a few atomic layers. In such films it has previously been demonstrated[5,6,8] that the superconducting transition temperature T_c decreases with increasing $R_\Box$ in the weakly localized regime for reasons that are still not clear.

Conventional superconductivity results from a balance between attractive binding via the electron-phonon interaction λ and the repulsive Coulomb term μ^*. The value of $\lambda - \mu^*$ determines T_c and Δ through the Eliashberg equations. Ordinarily, λ is obtained from the integral $\lambda = \int dE\, \alpha^2(E) F(E)/E$, where $F(E)$ is the phonon density of states and $\alpha^2(E)$ is the strength of the electron-phonon coupling. This function $\alpha^2(E) \times F(E)$ can be obtained from a tunneling measurement in the strong-coupling case by careful determination of the density of states over the energy range appropriate to the phonons of the material.[10] These effects show up as deviations from the BCS form of the electronic density of states $N_S(E)$, and the strength of these deviations reflects the coupling strength λ. Through a well-established inversion process,[10] these measurements yield the resultant $\alpha^2(E)F(E)$ along with λ and μ^*. Our expectation was that, with increasing disorder, the enhancement of the Coulomb interaction would be reflected by an increase of the Coulomb pseudopotential μ^*.

Using electron tunneling, we have studied the variation of T_c, the superconducting energy gap Δ, the density of states $N(E)$, and the parameters μ^* and λ which measure the strength of the electron-electron repulsion and the electron-phonon coupling. Lead was chosen because, in the low-$R_\Box$ limit, these parameters are all well known.[10] From our measurements, we conclude that (1) the ratio $2(\Delta/kT_c)$ remains constant (at the bulk value) and strong coupled over a range of T_c from 2 to 7 K, (2) $N(E)$ retains its (BCS-like) form with no indication of a distortion of the excitation spectrum or states appearing below the energy gap, and (3) an analysis of the tunneling data using the Eliashberg equations[10] breaks down with increasing $R_\Box$, indicating that it is not proper to think of the electron-phonon and electron-electron interactions as separable in this dirty limit. We find that the electron-phonon renormalizations of $N(E)$ decrease rapidly with increasing $R_\Box$.

The experiments were performed in an ultrahigh-vacuum (UHV), low-temperature thin-film evaporator described previously.[11,12] The tunnel junctions were fabricated by first evaporating an Al electrode on a glass substrate in a conventional evaporator, allowing it to oxidize in air, and then transferring it to the low-temperature evaporator. The apparatus was cooled to 1.6 K and then the counterelectrode of Pb was deposited. The uniform thickness of the Pb was achieved by first depositing ~1 monolayer of Ge immediately prior to the first Pb deposition.[6] Using this procedure, which presumably reduced the surface mobility of the Pb, we found that we could reproducibly deposit ultrathin films (< 10 Å) with $R_\Box \approx 2000$–$3000\ \Omega$. The T_c of the resultant film was measured, and tunneling characteristics in the superconducting and normal state were obtained. Following this, an additional few angstroms of Pb was deposited and the measurement

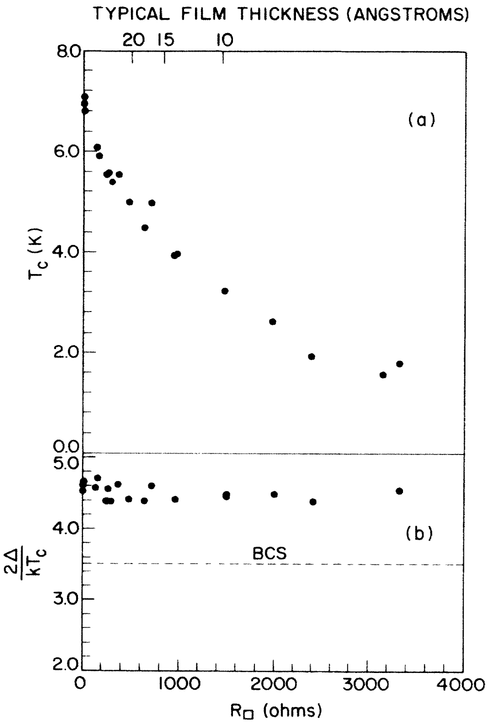

FIG. 1. (a) Superconducting transition temperature T_c of Pb films as a function of the sheet resistance $R_\square$. A monolayer of Ge has been deposited in advance. Typical film thicknesses are plotted at the top of the figure. (b) The ratio $2\Delta/kT_c$ of Pb films as a function of sheet resistance $R_\square$. The BCS weak-coupling value of 3.53 is indicated.

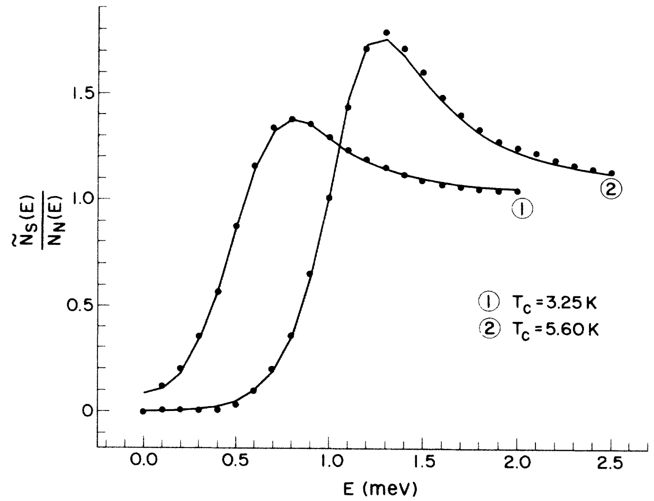

FIG. 2. The measured normalized density of states $\tilde N(E)$ at 1.6 K for two typical film thicknesses. The solid lines are fits by Eq. (2) using the BCS density of states and varying the energy gap Δ.

procedure was repeated. This sequence was iterated until $R_\square$ was reduced to a few ohms at which point T_c and the energy gap Δ reached their respective bulk values. The advantages of this technique are the lack of contamination during measurement (due to UHV conditions) and the ability to use the same tunnel barrier for all values of $R_\square$ in the sequence.

In Fig. 1(a) we show a plot of the variation of T_c (measured resistively at the midpoint) as a function of $R_\square$ for several experiments. These data are in agreement with earlier measurements[6] and illustrate the experimental reproducibility. The film thickness is indicated at the top of the figure. It is worth noting that the resistive transition widths remain narrow for all films (increasing from 0.03 to 0.15 K for the thinnest case). This is quite different from the case where superconductivity is destroyed by breaking the phase coherence of the wave function.[4] There T_c (onset) does not change, but the transition gradually broadens until such time as the material no longer superconducts.

Tunneling measurements on these samples provide a clear description as to how the reduction in the amplitude of the pair wave function proceeds. We have extracted Δ and the density of states $N(E)$ for these samples. The current-voltage characteristic of a normal-metal–superconductor tunnel junction at finite T is given by

$$I(V)=C_n(V)\int_{-\infty}^{\infty} N_S(E)[f(E)-f(E+V)]\,dE, \tag{1}$$

where $C_n(V)$ is the voltage-dependent normal-state conductance (logarithmic in this case because of electron-electron interactions), $f(E)$ is the usual Fermi function, and $N_S(E)$ is the superconducting density of states. For a BCS superconductor the latter assumes the form $N_S(E)=E/(E^2-\Delta^2)^{1/2}$. From a measure of the conductance of the tunnel junction in the superconducting state, normalized by the conductance in the normal state, one obtains a thermally smeared density of states

$$\tilde N_S(E)=\int_{-\infty}^{\infty} N_S(E')[f(E)-f(E+E')]\,dE'. \tag{2}$$

Using this expression and adjusting Δ, we obtain a very high quality fit to the data. Examples of the fit are shown in Fig. 2. We conclude from this that the density of states continues to be of the BCS form for reduced T_c films with little or no modification of this shape. We do not observe any states appearing in the gap or any additional gap broadening. However, Δ does change with changing T_c and $R_\square$. The extracted value for Δ from these measurements can be compared with the associated T_c and the $2\Delta/kT_c$ for a series of samples is illustrated in Fig. 1(b). This ratio in the weak-coupling limit should be 3.53 and deviations from this give a traditional measure of the electron-phonon coupling strength. Although T_c has decreased by a factor of ~ 4, the ratio $2\Delta/kT_c$ remains ≈ 4.4—well above the weak-coupling limit of 3.53. Taken alone, this result suggests that the strength of the electron-phonon coupling remains unchanged even though T_c is strongly suppressed.

Our expectations following this result were that the reduction in T_c could be simply described by enhanced Coulomb interactions which could, in turn, be described by an enhanced Coulomb pseudopotential μ^*. The existence of this enhancement is suggested by the multitude of evidence that Coulomb interactions become more important with increasing $R_\square$.[13,14] It is now well established that a logarithmic correction to the normal-state density of states results from Coulomb interactions in two dimensions.[14] Indeed, such a correction is clearly seen in the background conductances of these tunnel junctions.

Turning now to the strong-coupling corrections to the BCS density of states, we examine the results of a set of conductance measurements on a series of films as is shown in Fig. 3. The T_c of these films varies from 7.28 down to 3.25 K. The highest-T_c case is for a film of very low $R_\square$, and is consistent with results obtained for bulk disordered Pb. The deviations from the BCS shape for transverse and longitudinal phonons are clearly evident. Using the McMillan inversion program, we obtain values of $\lambda = 1.77$ and $\mu^* = 0.107$ for our thickest film, which are reasonable for bulk-disordered Pb. However, the thinner films in the same experiment show a systematic and marked decrease in the strength of the deviations from BCS. It should be reemphasized that the results presented in Fig. 3 represent one experiment in that the differences observed are solely due to a change in the Pb film thickness (all use the same Al electrode and tunnel barrier). This decrease in the apparent electron-phonon coupling strength is *substantially* larger than would be expected from the T_c decrease alone.

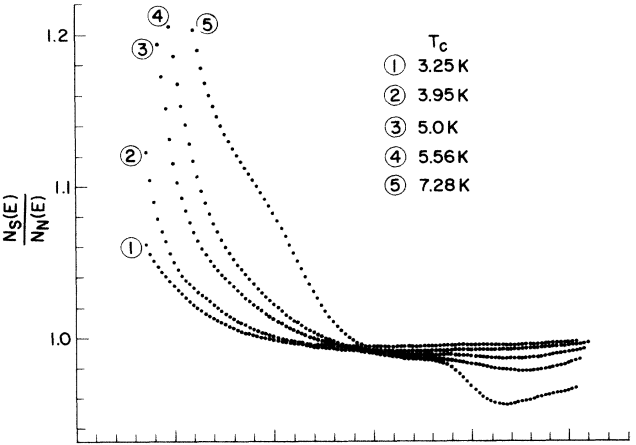

FIG. 3. Normalized density of states in the energy region where renormalizations due to the electron-phonon interaction are seen. This is for five films with the same tunnel junction. The transverse and longitudinal phonon structure at 5 and 10 meV is clear in the highest T_c film but diminishes rapidly with decreasing T_c.

A quantitative analysis of the data of Fig. 3 (and additional data from other measurements) reinforces this qualitative discussion. In fact, the λ extracted from this data decreases so rapidly with decreasing film thickness that the Eliashberg equations do not converge for the data of films with a $T_c \lesssim 4.5$ K. Even those fits to data with 7.2 K $> T_c >$ 4.5 K yield the surprising result that μ^* *decreases* and eventually goes negative with decreasing T_c.

We believe that this result signals a breakdown of this Eliashberg analysis. A fundamental assumption in the analysis is that the energy scale of the Coulomb interactions is so much greater than that of the electron-phonon interactions that the two contributions are separable. This is clearly implied in labeling the two interaction parameters λ and μ^* as electron-phonon and electron-electron. It has been pointed out[2,15,16] that in the presence of sufficient disorder the dynamics of the electron-electron interaction becomes important and affects the final pairing so that such a distinction is no longer possible. The fact that we observe Coulomb corrections to the normal-state density of states on an energy scale comparable to the effects due to $\alpha^2(E)F(E)$ further substantiates this. If indeed the two contributions are not separable, thinking of the destruction of superconductivity as being due to either a reduced λ or an enhanced μ^* is too naive. These tunneling results demonstrate that a more sophisticated treatment of the renormalization of the electron propagator with both time-retarded electron-phonon and electron-electron effects is necessary.[15,16] Only then will we be in a position to analyze quantitatively data of the type shown in Fig. 3 to extract the relevant parameters.

It is worth contrasting these results with those in the pure phase-breaking (inhomogeneous) limit.[4] In this limit the resistive T_c onset does not change with increasing $R_\square$, while the transition width broadens until such time as the material ceases to conduct. Furthermore, while the measured energy gap remains constant, substantial lifetime broadening is observed with increasing disorder. This broadening is consistent with enhanced inelastic scattering.

In summary, we have performed superconducting tunneling measurements on ultrathin films of Pb, a strong electron-phonon–coupled superconductor, in the homogeneous limit. With increasing $R_\square$ (decreasing thickness) both T_c and Δ decrease monotonically. Surprisingly, the ratio $2\Delta/kT_c$ remains at the strong-coupled value of 4.4, suggesting no fundamental change in the nature of the electron-phonon coupling. The density of states in the region of the gap retains its BCS shape with no evidence for gap smearing or excess states appearing in the gap. Finally, an analysis of the tunneling-conductance curves indicates that the deviations from BCS at higher energies associated with the electron-phonon function $\alpha^2(E)F(E)$ are substantially weaker than expected. The usual Eliashberg analysis fails for relatively small values of T_c suppression. This implies that an analysis of these superconductors using existing Eliashberg theory is inadequate and the dynamics of the Coulomb interactions must be taken into account.

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[1] P. W. Anderson, K. A. Muttalib, and T. V. Ramakrishnan, Phys. Rev. B **28**, 117 (1983).

[2] S. Maekawa, H. Ebisawa, and H. Fukuyama, J. Phys. Soc. Jpn. **52**, 1352 (1983).

[3] R. C. Dynes, J. P. Garno, G. B. Hertel, and T. P. Orlando, Phys. Rev. Lett. **53**, 2437 (1984).

[4] Alice E. White, R. C. Dynes, and J. P. Garno, Phys. Rev. **33**, 49 (1986).

[5] J. M. Graybeal and M. R. Beasley, Phys. Rev. **29**, 4167 (1984).

[6] M. Strongin, R. S. Thompson, O. F. Kammerer, and J. E. Crow, Phys. Rev. B **2**, 1078 (1971).

[7] A. Kapitulnik and G. Kotliar, Phys. Rev. Lett. **54**, 473 (1985).

[8] A. F. Hebard and M. A. Paalanen, Phys. Rev. B **30**, 4063 (1984).

[9] Michael Ma and Patrick A. Lee, Phys. Rev. B **32**, 5658 (1985).

[10] W. L. McMillan and J. M. Rowell, in *Superconductivity,* edited by R. L. Parks (Marcel Dekker, New York, 1969).

[11] R. C. Dynes, J. P. Garno, and J. M. Rowell, Phys. Rev. Lett. **40**, 479 (1978).

[12] R. C. Dynes and J. M. Rowell, Phys. Rev. B **11**, 1884 (1975).

[13] A. E. White, R. C. Dynes, and J. P. Garno, Phys. Rev. B **31**, 1174 (1985).

[14] B. L. Altshuler, A. G. Aronov, and P. A. Lee, Phys. Rev. Lett. **44**, 1288 (1980).

[15] D. A. Browne, K. Levin, and K. A. Muttalib, to be published.

[16] D. Belitz, to be published.