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Destruction of superconductivity in quench-condensed two-dimensional films

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We have performed systematic tunneling measurements on two-dimensional quench-condensed films of Sn and Pb to investigate the destruction of superconductivity by localization effects. With increasing sheet resistance of the films, the energy gaps and T_c's decrease only slightly, but the gap edges broaden until the width becomes comparable to the gap. Associated with this broadening are low-temperature finite-resistance tails in the resistive transitions.

Near the metal-insulator transition, both localization and interaction effects have a strong influence on the nature of conduction.[1] These effects become even more important as the dimensionality of the system is reduced. Extensive transport, magnetotransport, and tunneling measurements in a variety of three-dimensional (3D), 2D, and 1D samples have resulted in a comprehensive description of these phenomena in the normal state. Recently, there has been a great deal of interest in the role these effects play in the destruction of the *superconducting* state of a disordered metal. Tunneling studies in 3D granular aluminum samples close to the metal-insulator transition[2] showed that the superconducting energy-gap edge broadens, possibly due to lifetime effects, as the resistivity of the sample increases. When the broadening is comparable to the gap, superconductivity disappears. In this paper, we report our results on a corresponding 2D experiment using very thin quench-condensed films of tin and lead.

Two-dimensional systems are particularly appealing for studying localization and interaction effects because it is now understood that all the electronic states are localized and that these effects are somewhat independent of the material studied,[3] depending only on the sheet resistance ($R_\square$) of the sample. However, this assumption does not seem to hold for studies of the competition between these effects and superconductivity. Differences between our results on quench-condensed films and those of other experimental investigations on several diverse systems[4] have led us to recognize the distinction between reducing the pair amplitude and reducing the phase coherence in the destruction of superconductivity. This experiment addresses the latter.

Our earlier tunneling measurements[5] on quench-condensed tin films concentrated on the modification of the normal-metal density of states due to Coulomb interactions. In this experiment, we have focused on the changes in the superconducting excitation spectrum. The films were fabricated as previously described: A film of 99.99% Sn or Pb was evaporated onto fire-polished glass substrates held between 1.5 and 8 K. This film straddled four gold contacts and the aluminum (Al) counterelectrode which was previously deposited at room temperature and oxidized in air. Since we were interested in studying films of very high $R_\square$, it was necessary to work with relatively high-resistance junctions. This was accomplished by oxidizing the Al in air for 20 min to obtain resistances $\gtrsim 10\,000\ \Omega$ for a 0.25 mm^2 junction area. The films were deposited in a low-temperature evaporator that was immersed in liquid helium. Cryopumping on the walls of the chamber is expected to reduce the pressure inside to less than 10^{-10} torr. These films become "continuous" at about 50 Å, so the high $R_\square$ is probably due to weak coupling between small particles of clean metal. The sample geometry (inset to Fig. 2) was chosen to allow four-terminal measurements of both the junction conductance and the film resistance.

The advantage of this technique is that we were able to use the *same* tunnel junction for an entire series of films. This was possible because the evaporator was designed to allow *in situ* conductance measurements. As a result, after evaporating and measuring a film, additional material could be evaporated without formation of an intermediary oxide. In this way, we studied films that varied over 3 orders of magnitude in $R_\square$.

It is important to consider the significance of the granular structure for the nature of this investigation. From numerous localization studies, we now understand that the relevant length scale is given by the electron inelastic diffusion length, $L_i = \sqrt{D\tau_i}$, where D is the electron diffusivity and τ_i is the inelastic scattering time. L_i is the length an electron diffuses before experiencing an inelastic scattering process and, based on previous magnetoresistance measurements,[6] we estimate this length to range from several hundred to about a thousand angstroms for the films under study. As a result, the electrons actually sample many of the small particles comprising the film between inelastic events and average over them. In this sense, the inhomogeneities, which are on a scale $\lesssim 50$ Å, should not affect the properties of interest here. Furthermore, it is the inelastic length which determines the dimensionality of the sample. Since our films are $\lesssim 100$ Å thick, they are clearly two-dimensional under these criteria, as confirmed by magnetoresistance.[6]

The resistive transitions for a series of Sn films with $R_\square$'s ranging from 98.0 $\Omega/\square$ to 116.4 k$\Omega/\square$ are shown in Fig. 1. In all these measurements, we have measured several regions along each film at several different currents to ensure that the films are homogeneous over the 1 cm length of the film and that the resistances are not current dependent. We choose to present the data on a logarithmic scale to emphasize the sharp onset (which is at approximately the same T for all except the 116.4-k$\Omega/\square$ sample), the initial exponential decrease in resistance, and the appearance of a long tail which grows dramatically with $R_\square$. This means that the T_c of the tin films which do superconduct (defined as the midpoint) changes by less than 10% over more than 2 orders of magnitude in $R_\square$.

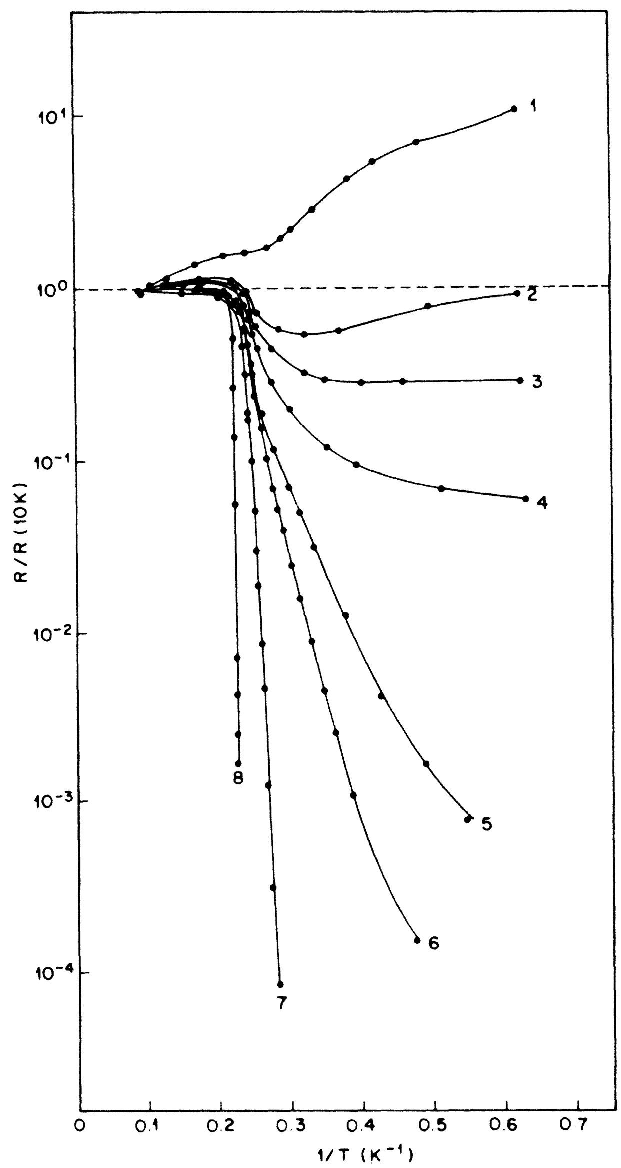

FIG. 1. Resistive transitions for a series of tin films with sheet resistances: 1, 116400 $\Omega/\square$; 2, 47500 $\Omega/\square$; 3, 38000 $\Omega/\square$; 4, 27700 $\Omega/\square$; 5, 16000 $\Omega/\square$; 6, 10500 $\Omega/\square$; 7, 3080 $\Omega/\square$; 8, 98.0 $\Omega/\square$. Note that T_c hardly changes.

Since T_c is apparently not changing with $R_\square$, it is interesting to observe how superconductivity disappears. As $R_\square$ increases, $R(T)$ below T_c develops a long tail where finite resistance persists. At higher $R_\square$, the tail continues to low temperatures and, with increasing $R_\square$, turns around as localization triumphs. Even in the highest-$R_\square$ film shown, there is a hesitation in the monotonic increase in R, indicating some remnant of superconductivity. Our tunneling measurements show the presence of an energy gap over the entire range of $R_\square$. The tunneling data were taken at ~ 1.7 K with the aluminum (Al) in the normal state (a normal-metal–superconductor tunnel junction) considerably simplifying the analysis. Conductance traces were qualitatively similar to those already published, except that we investigated the superconducting gap region with more resolution. In order to do a quantitative analysis, it is necessary to determine a normalized superconducting density of states $N_s(E)/N_n(E)$. This ratio is given by the ratio of the junction conductances in the superconducting and normal states with appropriate thermal factors. Since the logarithmic corrections to the normal-state density of states due to Coulomb interactions are significant, especially at low voltages, for most of the range of $R_\square$ studied, we also measured the conductance at 6 K. To deconvolve the superconducting density of states, we assumed that the measured current could be written in the form[7]

$$I_{ns}(V) = C_n(V)\int_{-\infty}^{\infty} N_s(E)[f(E)-f(E+V)]dE ,$$

where $C_n(V)$ is the normal-state conductance, which is voltage dependent (logarithmic) in this case, $N_s(E)$ is the superconducting density of states under study, and $f(E)$ is the usual Fermi function. For a Bardeen-Cooper-Schrieffer (BCS) superconductor, $N_s(E)$ assumes the form

$$N_s(E) = \frac{E}{\sqrt{E^2-\Delta^2}} ,$$

where Δ is the energy gap. The deconvolution is then accomplished by dividing the conductance measured at ~ 1.7 K (well below the superconducting T_c) by the conductance measured at 6 K. At low voltages, where thermal effects at 6 K truncate the logarithmic divergence of the corrections to the conductance, we extrapolated the high-voltage data and used these values for the division. We end up with a thermally smeared superconducting density of states of the form

$$\tilde{N}_s(E) = \int_{-\infty}^{\infty} N_s(E')[f(E')-f(E+E')]dE' . \qquad (1)$$

The results of such an analysis are shown in Fig. 2, where we plot $\tilde{N}_s(E)$ for (a) a low-$R_\square$ film and (b) a high-$R_\square$ film. Although Δ appears to remain constant, the shape of the curve changes as $R_\square$ increases. Following the results of the 3D work, we used a BCS density of states broadened with a linewidth Γ[2,8] to fit the data:

$$N(E,\Gamma) = \mathrm{Re}\left|\frac{E}{\sqrt{E^2-\Delta^2}}\right|, \quad E = E' - i\Gamma . \qquad (2)$$

To do the fit, we allowed Γ and Δ to vary, but observed that the value of Δ did not change more than $\pm 1.5\%$ over the entire $R_\square$ range studied. Typical best fits are shown as solid lines in Fig. 2. As expected, at low $R_\square$, (1) and (2) provide an excellent description of the data and, although the quality of the fit deteriorates at higher $R_\square$, there is still good

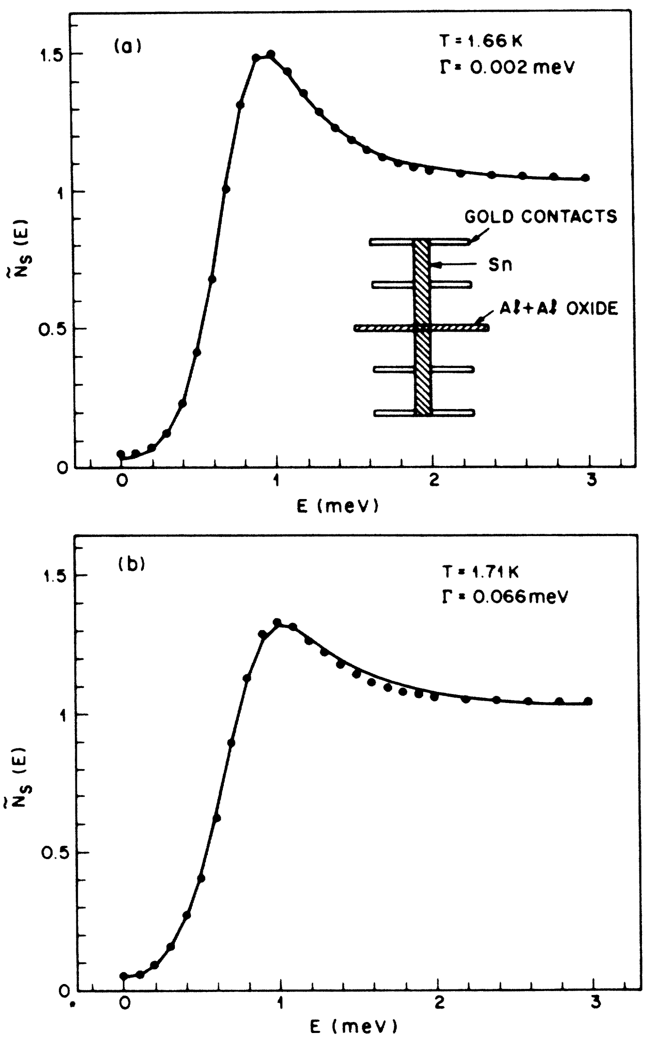

FIG. 2. The density of states deconvoluted from conductance data for (a) a 541.7-Ω/□ tin film and (b) a 9933-Ω/□ tin film. The solid lines are the best fit curves obtained using a BCS density of states broadened by the indicated value of Γ for each case. Inset shows the sample geometry.

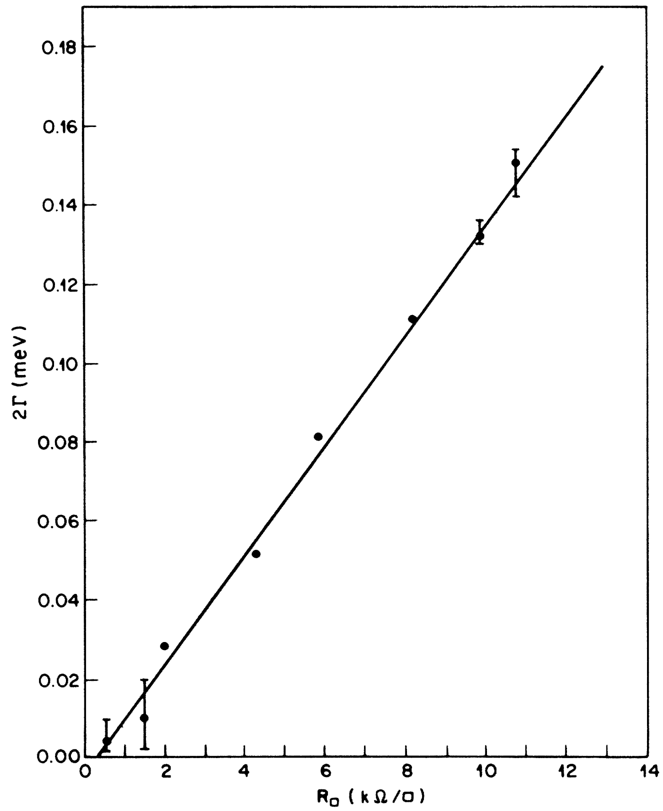

FIG. 3. Plot of the lifetime broadening factor, 2Γ, as a function of sheet resistance for a series of tin films. Solid line is least-squares fit to the data. For comparison, the value of Δ for these films is 0.74 ±0.1 meV.

agreement in the gap region at 9933 Ω/□. We have noticed that the data tend to fall below the fit between 1 and 3 mV and that this deviation becomes more pronounced as $R_\square$ increases. The same effect was present in the 3D data. Although we do not show the data, very similar results were obtained for Pb as well.

The values of Γ obtained from this fitting procedure on data from two different series of films show a linear dependence on $R_\square$ as seen in Fig. 3. If, as suggested in Ref. 2, the energy broadening is caused by inelastic electron-electron scattering processes, which are enhanced at low electron diffusivity (D), we should be able to estimate the scattering rate ($1/\tau_i$) using $2\Gamma = \Delta E \sim \hbar/\tau_i$. At 10 830 Ω/□ this yields a rate of 2.3×10^{11} sec^{-1}. For order-of-magnitude comparison, we also estimated $1/\tau_i$ using the previously measured[9] inelastic *e-e* scattering rate in Mg and the observed linear dependence of τ_i on D and found $1/\tau_i \approx 1.9\times10^{11}$ sec^{-1} at these temperatures. The agreement is surprisingly good since we are comparing a magnetoresistance determination of τ_i in Mg with a tunneling determination of τ_i in Sn, but it gives us added confidence in our picture of inelastic electron-electron scattering causing the lifetime broadening. Furthermore, an extrapolation of the plot in Fig. 3 implies that 2Γ will be equal to the gap at ~ 50 000 Ω/□, about where we observe that the system shows localized behavior below T_c. Additional evidence for this picture comes from measurements of the temperature variation of Γ. Although the results are not yet complete, we find that Γ increases substantially with increasing T up to T_c, where we can no longer measure it using this technique.

Our observation of a constant T_c and Δ is different from some previous investigations[4] of the impact of disorder on 2D superconductivity. In these studies it was found that there was a substantial reduction in T_c and Δ with increasing $R_\square$, and no superconductivity gradually disappeared as T_c and Δ approached zero. Here we believe we are addressing a rather different question: What happens to superconductivity when the microscopic superconducting coupling strength is not changing, but small microscopic regions are being decoupled from each other? It is interesting that, in this situation, superconductivity disappears at the point where strong localization begins to dominate and Coulomb interaction effects become very important. This occurs at 10 000–20 000 Ω/□ regardless of the material studied. Therefore, we think we are truly investigating the details of the competition between superconductivity (when the electron-phonon coupling is unchanged) and localization.

In principle, all 2D states are localized, but only weakly so at low $R_\square$. At finite temperature, inelastic scattering events delocalize the electrons on a length scale given by the inelastic scattering length. We estimate that the localization length and the coherence length are comparable in the region of 10 000–20 000 Ω/□. This gives us further confidence that we are observing the intrinsic struggle[10] between localization and superconductivity.

In summary, we have studied the nature of the destruction of superconductivity in 2D films of Sn and Pb as a function of increasing $R_\square$. This has been accomplished by systematic $R(T)$ and tunneling measurements on quench-condensed films. We find that T_c and Δ remain almost unchanged up to 10 000 Ω/□. The data can be best described by a decreased lifetime of the pairs and quasiparticles due to inelastic scattering. This is manifested as a temperature-dependent broadening of the density of states, similar to that observed in 3D. As the disorder increases, the broadening becomes comparable to the energy gap, localization wins out, and the material no longer superconducts.

[1]P. A. Lee and T. V. Ramakrishnan, Rev. Mod. Phys. **57**, 287 (1985).

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

[3]E. Abrahams, P. W. Anderson, D. C. Licciardello, and T. V. Ramakrishnan, Phys. Rev. Lett. **42**, 673 (1979).

[4]M. Strongin, R. S. Thompson, O. F. Kammerer, and J. E. Crow, Phys. Rev. B **2**, 1078 (1971); D. G. Naugle, R. E. Glover, and W. Moormann, Physica **55**, 250 (1971); R. C. Dynes, J. P. Garno, and J. M. Rowell, Phys. Rev. Lett. **40**, 479 (1978); H. Raffy, R. B. Laibowitz, P. Chaudhari, and S. Maekawa, Phys. Rev. B **28**, 6607 (1983); J. M. Graybeal and M. R. Beasley, *ibid.* **29**, 4167 (1984); A. F. Hebard and M. A. Paalanen, *ibid.* **30**, 4063 (1984).

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

[6]G. Bergmann, Phys. Rev. B **25**, 2932 (1982); A. E. White, R. C. Dynes, and J. P. Garno, *ibid.* **29**, 3694 (1984).

[7]M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1975), p. 46ff.

[8]R. C. Dynes, V. Narayanamurti, and J. P. Garno, Phys. Rev. Lett. **41**, 1509 (1978).

[9]A. E. White, R. C. Dynes, and J. P. Garno, Phys. Rev. B **29**, 3694 (1984).

[10]M. Ma and P. A. Lee (unpublished).