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DIVERGENT FLUCTUATIONS IN SUPERCONDUCTING FILMS*

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We have observed anomalously broad resistive transitions in superconducting films and have shown that the widths gradually reduce to the usual Aslamazov-Larkin value when a magnetic field is applied parallel to the film. These results are explained by adding to the Aslamazov-Larkin theory another fluctuation contribution to the conductivity of the type first proposed by Maki, which for thin films is logarithmically divergent with a cutoff proportional to the pair-breaking interactions.

Theoretically, fluctuations in thin superconducting films tend to diverge. In fact if one assumes that long-range superconducting order exists, then the fluctuations would actually diverge, thereby destroying such order.[1,2] Naturally one would also expect to see some evidence of this divergence in the fluctuations above the mean-field critical temperature T_c, although perhaps not in the leading order. However, the first successful theory of the rounding of the resistive transition, that of Aslamazov and Larkin (AL),[3] contained no divergence above T_c and was in excellent agreement with the experimental results of Glover[4] on very dirty films of strong-coupling superconductors. In contrast, the early experimental work of Strongin et al.[5] followed by the definitive experiments of Masker and Parks[6] showed that the resistive transitions in aluminum could be anomalously large, even more than an order of magnitude larger than the AL value for low-resistance films. A second theory of the resistive transition by Maki,[7] which appeared shortly after AL, was divergent when applied to films. In the form first proposed by one of us[8] the divergence is cut off by a pair-breaking interaction such as electron-phonon scattering or magnetic fields. The extra term of Maki was predicted to decrease relative to the AL term as stronger magnetic fields are applied parallel to the film. We now wish to report experimental confirmation of this theory, the first experimental evidence identifying the tendency of fluctuations to diverge in superconducting films away from T_c. Furthermore we have observed the same tendency in lead films although the enhancement of the transition width is smaller. The AL result was recovered near the transitions with no significant reduction due to strong coupling.

The AL theory for weak-coupling superconducting films of thickness d less than the temperature-dependent coherence length $\xi(T)=\xi(0)/\tau^{1/2}$, where $\tau=(T-T_c)/T_c$, gives a simple result for the extra conductivity σ' due to fluctuations relative to the normal-state conductivity σ:

$$\sigma_{\rm AL}'/\sigma=\tau_0/\tau, \tag{1}$$

where $\tau_0=(1.52\times10^{-5}\ \Omega^{-1})R_\square$. $R_\square$ is the normal-state film resistance per square area. This contribution to σ' is the extra conductivity of the pairs of electrons which are temporarily formed by fluctuations.

For the same films the additional Maki term may be written

$$\sigma_{\rm M}'/\sigma=[2\tau_0/(\tau-\delta)]\ln(\tau/\delta), \tag{2}$$

where δ is the relative shift of T_c from some original value T_{c0} due to a pair-breaking interaction, $\delta=(T_{c0}-T_c)/T_c$. This additional contribution to σ' comes from the interaction of the normal electrons with the fluctuation pairs.

The total σ' is the sum of $\sigma_{\rm AL}'$ and $\sigma_{\rm M}'$. In the region near the transition where $\tau\ll\delta$ the AL result is obtained, $\sigma_{\rm AL}\gg\sigma_{\rm M}'$. However, far away from the transition, $\sigma_{\rm M}'\gg\sigma_{\rm AL}'$ and a width much broader than τ_0 is observed. The value of δ due to electron-phonon scattering should be $\sim(T/\text{Debye temperature})^2$, and due to electron-electron scattering $\sim(T/\text{Fermi temperature})$. For aluminum both estimates give $\sim10^{-5}$, and for lead the first is larger, $\sim10^{-2}$. δ can be increased most simply by applying an external magnetic field, although one could also add paramagnetic impurities, etc. A field parallel to the film gives an increase of $\delta=\frac{1}{3}[eH_\parallel d\xi(0)]^2$. In a perpendicular field σ' does not follow (1) and (2) due to a quantization of the fluctuation spectrum in vortex states.[9]

Aluminum evaporated from a wetted tungsten filament was deposited on a Pyrex substrate held at room temperature in a vacuum of $\sim10^{-6}$ Torr, whereas the lead was evaporated from a Ta boat with the substrate cooled to liquid-nitrogen temperatures. After the films were exposed to air and annealed at room temperature for several days, a zigzag pattern was scribed within an area less than 1 cm^2 using a precision micromanipulator. The films were then mounted on a variable-temperature surface within an evacuatable chamber. The film temperature was monitored using a carbon resistor calibrated during each run against the vapor pressure of liquid helium and/or a calibrated Ge resistor also attached to the film. For zero-magnetic-field measurements the earth's field was reduced to $\sim10^{-3}$ G using Mumetal shielding, and for nonzero-field measurements the field was provided by a movable room-temperature electromagnet which could be set parallel to the film with an accuracy of one part in 10^4 by monitoring the resistance in the transition region.

A standard four-probe method was used to measure the resistivity with a current density of the order of 100 A/cm^2 and a potential of the order of 1 mV/cm. σ' was observed to be independent of current when the current was reduced, as expected from theory. Current dependence should appear when the current approaches the critical current calculated from the Ginzburg-Landau theory for the same absolute value of $T-T_c$, $eE_c\approx|\tau|^{3/2}\pi T/\xi(0)$.[10, 11] The depairing effect should be proportional to $(E/E_c)^2$, typically 10^{-5} in our case.

A most revealing way of presenting the data is to plot the inverse of σ'/σ vs temperature.[6] Data plotted in this way are very sensitive to the value of the normal-state σ used when σ' is very small. We measured the normal σ by applying a strong perpendicular magnetic field to quench the fluctuations. For aluminum σ(normal) was independent of temperature over the range of the experimental data with no magnetoresistance effects. For lead a determination of σ(normal) as a function of temperature was necessary and corrections $\sim10^{-4}$ were made to compensate for the observed magnetoresistance. Far from the transition σ(normal) varies more with temperature than σ'.

According to the AL theory, the inverse of σ'/σ should decrease linearly with slope $(\tau_0T_c)^{-1}$ as the temperature is lowered and intersect the T axis at T_c. Our theory does not predict strictly linear behavior but rather curves of slowly varying slopes depending on the ratio of τ to δ.

The data for two aluminum films are shown in Figs. 1 and 2 in comparison with the theory. In both cases the width of the transition decreases by an order of magnitude as the field is increased and finally reaches the AL value τ_0, in agreement with the theoretical prediction. Our zero-field widths for these and several other films[12] agree with those found by Masker and Parks. As the films become dirtier the deviation from AL in zero field decreases and the value of δT_c necessary to fit the data increases. The smallest δ we have observed was 10^{-4} for another clean film with $R_\square=0.88\ \Omega$. However, the values of δT_c deduced in this way should not be

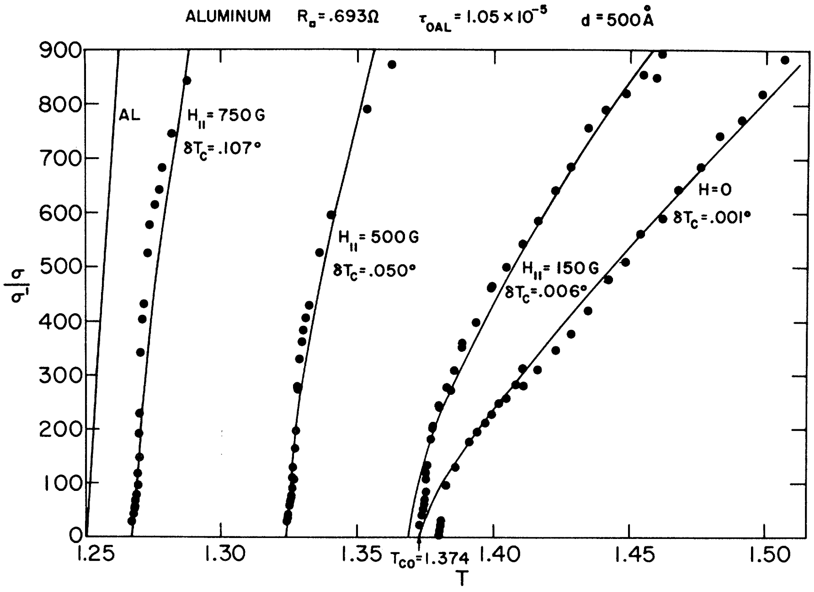

FIG. 1. The inverse of the measured pair conductivity σ' normalized to the normal-state conductivity σ plotted as solid points versus temperature. The curves through the data are calculated from the sum of Eqs. (1) and (2). The value of δT_c, the shift of T_c due to pair breaking, is adjusted to fit the slope of the data in zero field and then increased proportionally to $H_\parallel{}^2$ to obtain the other curves. The straight line labled AL has the slope predicted by the AL theory, Eq. (1). The inverse slope of the data in zero field is 11 times the AL prediction and decreases to the AL value as stronger fields are applied.

taken too seriously since the theory is not very sensitive to the exact value of δT_c chosen, depending on it only logarithmically. The detailed agreement between theory and experiment is worse in the dirtier film, perhaps because the transition is broader (being proportional to $R_\square$) and therefore the values of τ and δ in the measured temperature interval are not very small ($\gtrsim 0.1$), whereas the theory was derived assuming them to be $\ll 1$.

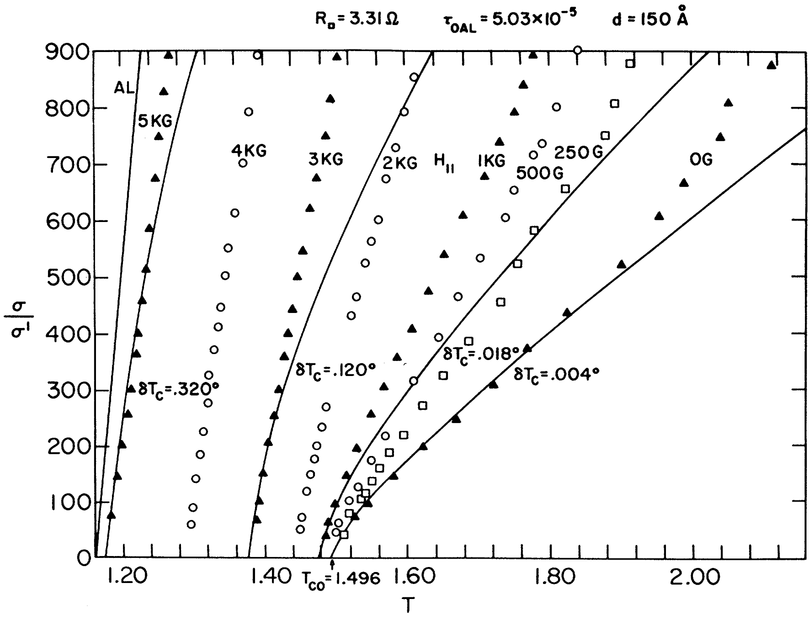

FIG. 2. Data presented as in Fig. 1 for an aluminum film with a resistance 5 times larger. The theoretical curves shown correspond to the data drawn as solid triangles, $H_\parallel = 0$, 1, 3, and 5 kG. The zero-field curve in this case has an inverse slope 10 times AL and is fitted with a larger value of δT_c.

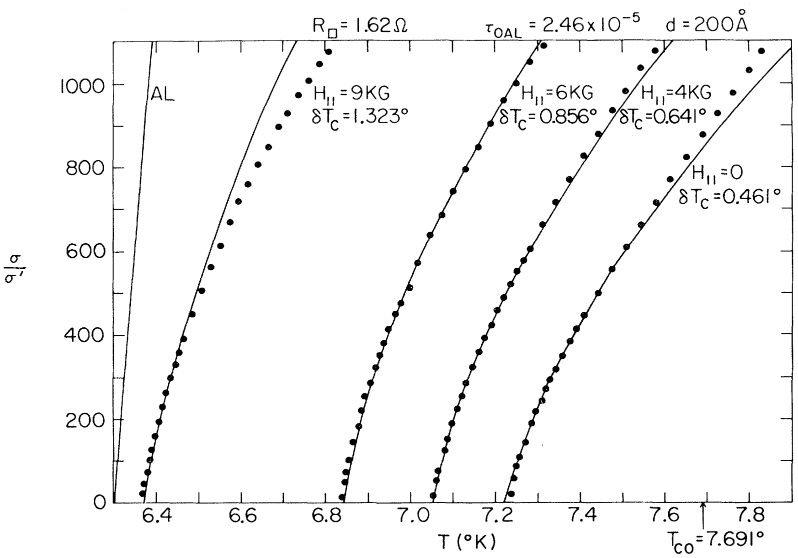

FIG. 3. Data for a lead film presented as for the aluminum films. δT_c in zero field is considerably larger than for the aluminum films, and the inverse slope away from the transition is now only 3 times AL. Near the transitions the AL slope is observed with no significant corrections due to strong coupling.

The data for a lead film are shown in Fig. 3. The value of δ in zero field, 0.06, is again somewhat larger than the calculated[13] value, 0.010-0.016. Near the transitions, between 0.8 and 1.0 of the AL value is obtained, in agreement with Glover's observations on much dirtier films.[4,14] According to the generalization of the time-dependent Ginzburg-Landau theory by Fulde and Maki[15], strong-coupling effects should result in a width near the transition related to AL by a factor γ/α.[16] α is defined as the amount by which the slope dH_{c2}/dT is enhanced near T_c relative to the weak-coupling value in the strong-coupling theory of Eilenberger and Ambegaokar.[17] We have measured the slope dH_{c2}/dT and using their formulas find an enhancement factor α between 2.0 and 2.3, in agreement with measurements of H_{c3}.[18,19] Hence, the absence of a significant strong-coupling correction to the AL width requires $\gamma \approx 2$ and a discrepancy appears between experiment and the Fulde-Maki estimate $\gamma \approx 1 + T_c/$(Debye temperature) ≈ 1.1.

In conclusion, we have observed anomalous fluctuations in superconducting films and verified their nature by selectively quenching them with magnetic fields in agreement with theory. We wish to thank M. Strongin for his constant interest and encouragement and R. D. Parks for making known to us his experimental results on aluminum films prior to publication.

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PIEZOELECTRIC POLARON-CYCLOTRON RESONANCE IN THE QUANTUM LIMIT IN n-CdS

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The zero-temperature cyclotron resonance of the electron splits into two groups as the temperature is increased. One group moves rapidly toward very small mass, the other toward larger mass. This splitting has not been predicted by previous theories. Moreover, the first member of the small-mass group can be accounted for only qualitatively by these theories.

We have observed a spectacular splitting of the electron-cyclotron-resonance absorption line in n-CdS as a function of temperature. We believe this to be the first observation of the temperature-dependent behavior of the piezoelectric polaron in the quantum limit. The cyclotron resonance was observed at wavelengths of 337 and 195 μm by using an HCN-DCN laser as a source of radiation.[1] At low temperatures and high frequencies where the quantum condition is generously satisfied, namely, $k_BT/\hbar\omega < \frac{1}{4}$, a single sharp absorption line is seen as shown at the lower left of Fig. 1. It appears to be the true cyclotron-resonance electron effective mass in CdS of $(0.159 \pm 0.002)m_0$ at 337 μm and $(0.172 \pm 0.002) \times m_0$ at 195 μm. It is observable only in this limit and is less than the "bare" mass value[2] of $0.2m_0$. We believe that this is due to electron-phonon interaction and, in this case, it corresponds to the emission of phonons. As the temperature is increased to the range $\frac{1}{4} < k_BT/\hbar\omega < \frac{1}{2}$, the single absorption line becomes an unresolved doublet with center near 0.17 m_0 which is the mass previously reported.[3,4] Finally, at higher temperatures, the two lines become two groups of lines. These

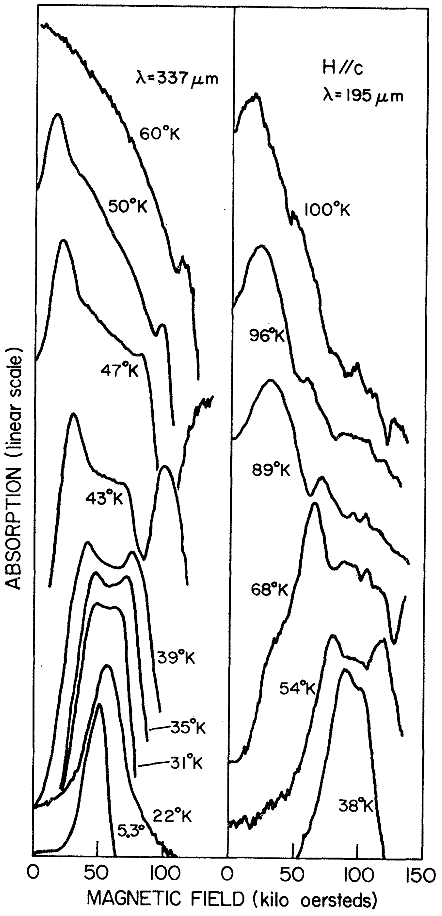

FIG. 1. Tracings of cyclotron-resonance absorption lines in n-CdS for several temperatures and at two different wavelengths.