MICROWAVE CONDUCTIVITY OF GRANULAR ALUMINUM FILMS IN THE SUPERCONDUCTING TRANSITION REGION*†

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Data are presented on the resistivity of granular aluminum films near the superconducting transition temperature T_c for both dc and 20-GHz excitation. Comparison with a recent theory of the effect of fluctuations on the frequency-dependent resistivity above T_c shows much less excess conductivity than predicted. A possible explanation for the discrepancy may lie in the incorrectness of a currently suggested form for the time-dependent Ginsburg-Landau equation.

Several experimental studies[1,2] of the details of the dc superconducting transition for superconductors with a short mean free path have shown that a considerable conductivity exists above the critical temperature. The shape of the dc resistance-versus-temperature curve, $R(T)$, was found to be in good agreement with the theoretical result of Aslamazov and Larkin[3]:

$$R(T)=R_n(1+\tau_0/\tau)^{-1}, \tag{1}$$

where R_n is the normal-state resistance, τ is given in terms of the transition temperature T_c as $\tau=(T-T_c)/T_c$, and τ_0 is a constant given in Eq. (3) below. The effect is attributed to fluctuations of the order parameter, which manifest themselves in an excess conductivity in the transition region.

Recently, Schmidt[4] has calculated the frequency dependence of these fluctuations using a version of the time-dependent Ginzburg-Landau equations given by Abrahams and Tsuneto.[5] Qualitatively, his results predict that the effect should decrease with frequency. For a thin film of thickness d, much less than the Ginzburg-Landau correlation length, the excess conductivity parallel to the film, $\sigma_\parallel(\omega, T)$, is derived to be

$$\sigma_\parallel(\omega,T) = \frac{e^2}{16\hbar d\tau}\left[\frac{\pi}{\tilde\omega}-\frac{2}{\tilde\omega}\tan^{-1}\frac{1}{\tilde\omega}-\frac{1}{\tilde\omega^2}\ln|1+\tilde\omega^2|\right], \tag{2}$$

where $\tilde\omega=\pi\hbar\omega/16k_BT_c\tau$, e is the electronic charge, k_B is Boltzmann's constant, and $\hbar$ is Planck's constant. When $\sigma_\parallel$ is added to the normal-state conductivity σ_n [$=Ne^2l/P_f$, where N is the carrier density, l is the mean free path, and P_f is the Fermi momentum], the total conductivity σ_T in the transition region is obtained. In terms of the experimentally measured resistances, the result becomes

$$R_T=R_N\left[1+\frac{P_f}{16N\hbar dl\tau}(1-\tfrac{1}{6}\tilde\omega^2)\right]^{-1}. \tag{3}$$

At zero frequency Eq. (1) gives the Aslamazov and Larkin result cited above.

In the course of transmission-type experiments at 20 GHz, reported elsewhere,[6] we have obtained data on the microwave conductivity of granular superconducting aluminum films. For temperatures less than T_c we found that the transmission is very well described by the Mattis and Bardeen conductivity functions.[7] For granular aluminum films then, one has a BCS superconductor with short mean free path. In the light of Schmidt's theory, we have reexamined our dc and microwave data above T_c. Qualitatively, we find that our films which have mean free path ranging from 1.6 to 10 Å[8] agree with the theory for dc excitation but show much less excess conductivity than predicted for 20-GHz excitation.

In Fig. 1, we present the dc transition data and the relative amplitude of the transmission in the superconducting state to that in the normal state $|T_s/T_n|$ as a function of temperature for one typical film. This sample was chosen because both the microwave and dc data were taken on the same film. After the microwave data were taken, the film was cut into a four-point configuration with a sharp point and the dc resistive transition measured. For this film both the thickness (275 ± 50 Å) and the effective mean free path[8] (1.65 Å) fall within the limits of the theory. Curve A is a plot of the dc transition, i.e., $\omega=0$, predicted by Eq. (3). The predicted resistance

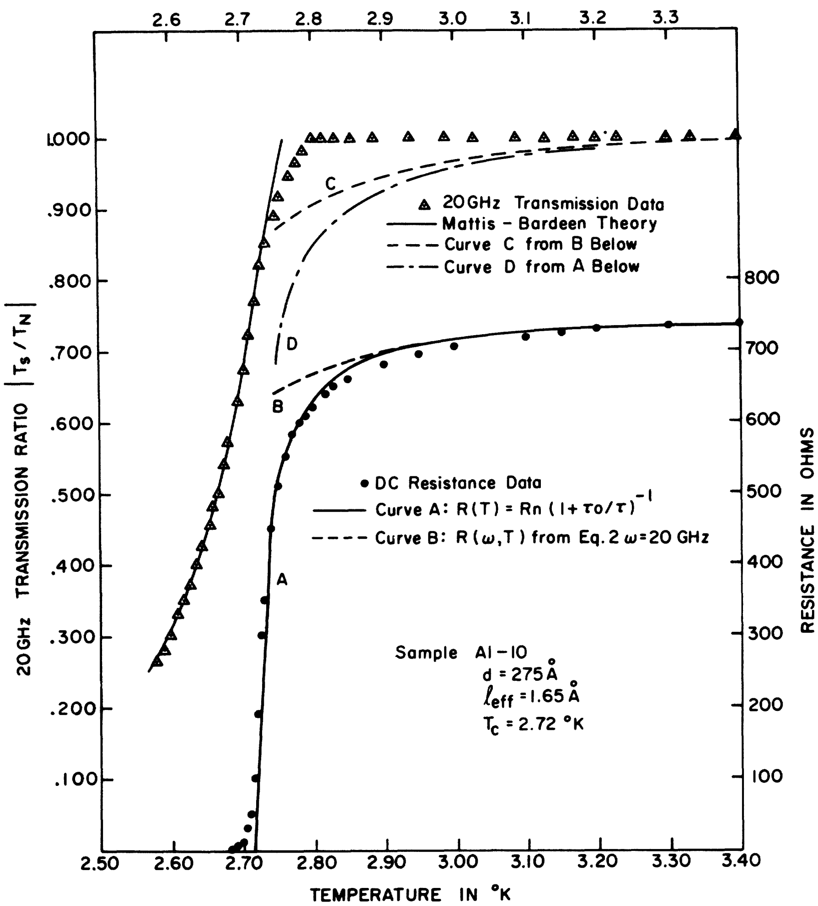

FIG. 1. Transition data for granular aluminum film No. 10.

at 20 GHz is shown in curve B. Curves C and D show the calculated transmission ratio[6,9] using curves A and B, respectively.

It can be seen from Fig. 1 that while the agreement in the dc case is satisfactory, much less excess conductivity than predicted is obtained at 20 GHz.

The data are clearly too incomplete for a detailed discussion but a few remarks would seem to be in order. Schmidt's theory is in some sense a test of the proposed time-dependent Ginzburg-Landau equation. In obtaining the predicted dynamical effects, certain thermal-averaging procedures were used to obtain the frequency spectrum of fluctuations which was then related via the fluctuation-dissipation theorem to the real part of the conductivity. While it is possible to imagine that different averaging procedures would yield quantitative agreement, a distinct alternative possibility remains that the time-dependent equation itself does not adequately describe relaxation phenomena in a BCS superconductor.

Further experiments are now in progress to investigate the frequency dependence of the effect.

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VARIATION OF LONG-RANGE ORDER IN Fe_3Al NEAR ITS TRANSITION TEMPERATURE*

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The long-range order parameter of the fcc ordered Fe_3Al structure, as measured by x-ray scattering, varies as $(T_C-T)^\beta$, with $\beta=0.307$, in the range between 1 and about 10 K below T_C (823 K).

Of the many order-disorder transformations in binary metallic solid solutions, only a handful appear to be other than first order in the thermodynamic sense. These are the transitions from the $L2_0$ structure to $A2$ in CuZn, FeCo, and FeAl, and that from $D0_3$ to $L2_0$ in Fe_3Al. The scanty evidence for this classification has been reviewed[1] by one of us. In the light of a possible connection between the crystal structure and the order of the transformation, it seemed desirable to verify that some order-disorder transformations do take place without any isothermal discontinuity in long-range order and, therefore, presumably without a latent heat. For such transformations, it also seemed desirable to study the behavior of the pair-correlation function in detail near the transition temperature. An x-ray investigation of Fe-Al alloys was begun because the components of this system differ far more in x-ray scattering amplitude than do those of either of the other two systems mentioned above. In this Letter, we are presenting our results on the variation of the long-range order parameter with temperature near the $D0_3 \rightarrow L2_0$ transition ($T_C \cong 823$ K). We have also measured the "critical" diffuse scattering above T_C, but interpretation of these data is strongly dependent on corrections for instrumental resolution, and is reported in the following Letter.[2]

The starting material for the sample was an ingot containing 29.2-at.% aluminum, made by the Hamilton Watch Company. A 5-kg piece of this ingot, after annealing for 40 h just below its melting point, had substantially one orientation. A slab with faces parallel to (111) was cut from this crystal, and annealed at 1375°C for 48 h in hydrogen to reduce composition variations. The final specimen was a disk about 1 cm in diameter and 1 mm thick. The face exposed to the x-ray beam was polished metallographically, last with 1-μ diamond powder, and was lightly etched to remove traces of cold work. The opposite face was brazed to the end of a copper cylinder, on whose lateral surface was wound a Nichrome wire heater insulated with MgO from its stainless steel sheath. The furnace, with surrounding radiation shields, was supported on a goniometer of the "Eulerian cradle" type, in an evacuated chamber mounted on the spindle of a commercial x-ray diffractometer.

Temperatures constant to about ±0.02°C for many hours were achieved by proportional control of the heating power, the controlling signal being the difference between a fixed emf and that from a Chromel-Alumel thermocouple attached to the furnace. A very low-noise, commercial breaker amplifier provided the necessary gain of about 6000 between the error signal and the input to the controller.

The x rays were the chromium $K\alpha$ radiation from a tube operated at 50 kV and 30 mA, selected by diffraction from (200) "planes" of a lithium fluoride crystal plastically bent to a toroidal form. The detection system, using a xenon proportional counter and pulse-height selection, was of standard commercial design, except that the incident beam was continually monitored. For this purpose, the beam passed through a thin, uniform titanium foil, the fluorescence from which was counted and used to drive the electronic timer. All intensity measurements were thus automatically normalized to constant incident flux.

The square of the long-range order parameter of the $D0_3$ structure is proportional to the intensity of any Bragg reflection with all odd indices, referred to the fcc unit cell containing 16 atoms. We measured the first and most intense of these, (111), in several series. The goniometer was set at the orientation giving maximum intensity,