

temperature along the superconductor is small, the value of θ is estimated from formula (3.8) of [11]. At temperatures close to the transition point, the coefficient η is in this case not a smooth function and it cannot be taken outside the integral with respect to T . Using (22) and the expression for the number of superconducting electrons near T_c , we obtain

$$0 \approx \frac{m T_c}{eh N_0} \eta_{12} \left\{ \ln \frac{T_c - T_1}{T_c - T_2} + 2.2 \frac{(1+d)}{(1+c)f} \left[\frac{2}{3} \left(\left(\frac{T_c - T_1}{T_c} \right)^2 - \left(\frac{T_c - T_2}{T_c} \right)^2 \right) \right] \right. \\ \times \ln \sigma^{-1} + \frac{2}{9} \left(\left(\frac{T_c - T_2}{T_c} \right)^2 \ln \frac{T_c - T_2}{T_c} - \left(\frac{T_c - T_1}{T_c} \right)^2 \ln \frac{T_c - T_1}{T_c} \right. \\ \left. \left. - \frac{2}{3} \left(\frac{T_c - T_2}{T_c} \right)^2 + \frac{2}{3} \left(\frac{T_c - T_1}{T_c} \right)^2 \right) \right\}. \quad (25)$$

Substituting in the last expression $T_2 \sim 0.95 T_c$, $T_1 \sim 0.93 T_c$, $N_0 \sim 10^{23} \text{ cm}^{-3}$, $\eta_{12}^n = \alpha/\rho$, $\alpha = 0.32 \text{ } \mu\text{V/deg}^2$, $\rho/100 n_{\text{imp}} = 0.9 \text{ } \mu\Omega\text{-cm/at.}\%$, $\varepsilon_d \sim 0.1 \text{ eV}$, $\Gamma \sim 10^{-2} \text{ eV}$, and $T_c \sim 1.3 \text{ }^\circ\text{K}$ (this corresponds to the data for $\text{ThCe}^{[9]}$), $N(0) \sim 0.1 \text{ eV}^{-1}$, and $d \sim 5$, we find that at concentrations $n_{\text{imp}} \sim 10^{-5} = 10^{-3} \text{ at.}\%$ we have $\theta \sim 10^{-2}$, which is experimentally feasible. [12] Allowance for the second term in the curly bracket leads to an increase of θ by an approximate factor of 3. With decreasing impurity concentration, θ increases in this case not only because of the decrease of ρ , but also because of the decrease of σ .

Measurements of the thermoelectric effect in superconductors containing a nonmagnetic impurity with a localized level can yield additional data on the nature of these impurities.

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Current dependence of the microwave admittance of thin superconducting films

V. P. Andratskiĭ, V. N. Gubankov, K. K. Likharev, and N. B. Pavlov

Institute of Radio Engineering and Electronics, USSR Academy of Sciences

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The dependence of the microwave ($\omega/2\pi = 9300 \text{ MHz}$) admittance of thin (300-600 Å) superconducting Sn films on the temperature and on the direct current I is investigated. The films are 1-2 μ wide and 30-100 μ long. The measurements are performed at temperatures $T \approx (1.0-0.9) T_c$ and currents I of the order of the pair-breaking current I_c^{GL} . The experimental $\sigma_1(T)$ plot is in good quantitative agreement with the Mattis-Bardin theory; the slope of the plot is slightly less than the theoretical one. The $\sigma_2(I)$ plot at $I \leq I_c^{\text{GL}}$ agrees with that predicted by the Ginzburg-Landau theory for $\omega\tau_{\Delta} \gg 1$ (τ_{Δ} is the time of relaxation of the superconducting condensate to the equilibrium state). An estimate of the lower bound of τ_{Δ} is obtained ($\tau_{\Delta} \gg 10^{-11} \text{ sec}$ at $T \gtrsim 0.9 T_c$).

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INTRODUCTION

The behavior of superconducting films in a weak high-frequency field is customarily described by the admit-

tance $\sigma_s = \sigma_1 - i\sigma_2$, which depends on the frequency ω . In the next higher approximation (in the absence of transport current) the admittance of the films is described by the Mattis-Barden theory. [1] If, however, the direct

current in the film is comparable with the critical pair-breaking current I_c^{GL} ,^[2] then an important role in the admittance is assumed by processes connected with the change of the order parameter Δ , due to the breaking of the Cooper pairs by the current. Under these conditions the high-frequency admittance is a function of the transport current I , and the character of this dependence depends essentially on the ratio of ω to the time τ_Δ of the relaxation of the order parameter to the equilibrium value.

At low field frequencies, when $\omega\tau_\Delta \ll 1$, the order parameter will follow the field adiabatically, so that to determine Δ at each instant of time we can use the equations of the Ginzburg-Landau (GL) theory.^[3] A generalization of the Ginzburg-Landau theory to the case $\omega\tau_\Delta \sim 1$ was correctly carried only for gapless superconductors.^[4,5] At $\omega\tau_\Delta \gg 1$, the order parameter will respond to the time-averaged value of the square of the field.^[6,7]

The dependence of the high-frequency admittance σ_2 on the transport current is of considerable interest since it permits, on the one hand, to observe the suppression of the order parameter by the current in the region $I < I_c^{GL}$, and on the other to estimate the relaxation time τ_Δ from the character of this dependence.

The first attempt to estimate the relaxation time of the order parameter from measurements of the $\sigma_2(I)$ dependence for Sn was undertaken at 23 GHz by Gittleman *et al.*^[8] They reached the conclusion that for the investigated films, in the temperature region $T/T_c \lesssim 0.9$ (T_c is the critical temperature), the condition $\omega\tau_\Delta \ll 1$ is satisfied. This conclusion disagrees strongly with the later measurements of Peters and Meissner,^[9] who obtained for tin films at $0.95 \leq T/T_c \leq 0.995$ the value $\tau_\Delta = 2.6 \cdot 10^{-10} (1 - T/T_c)^{-1/2}$ sec. It must be noted that the authors of the cited papers^[8,9] present no data with which to estimate the degree of uniformity of the current distribution in the sample in the working temperature range.

Rose^[10] investigated the dependence of $\sigma_2(I)$ at $I \ll I_c^{GL}$ in Al films at a frequency 140 kHz. At such low frequencies, in the entire working temperature range, the condition $\omega\tau_\Delta \ll 1$ was naturally satisfied. Churilov *et al.*^[11] investigated the $\sigma_2(I)$ dependence in Sn films at a frequency 10 GHz. The large scatter of the experimental points in this study does not make it possible to draw any conclusions concerning the value of τ_Δ .

Thus, correct measurements of the nonlinear high-frequency properties of superconductors were made apparently only in^[8,9], where contradictory estimates of τ_Δ were obtained. We have therefore undertaken careful investigations of the dependence of the microwave conductivity of thin tin films on the transport current and on the temperature.

1. EXPERIMENTAL PROCEDURE

A. Film parameters

The investigated samples were narrow (of width $w=1$ to 2μ and length $L=20$ to 100μ) strips of Sn that go over into broad "shore" parts ($w_{sh}=2$ mm), to which the

Sample No.	w, μ	$d, \text{\AA}$	T_c, K	$(R_{300})_{\square}, \Omega$	$(R_N)_{\square}, \Omega$	$l, \text{\AA}$	$\xi(0), \text{\AA}$	$\delta_{\perp}(0), \text{\AA}$
7	1.3	600	3.70	2.00	0.18	950	980	400
10	2.3	300	3.67	5.00	1.00	380	700	2700
11	2.2	550	3.70	2.14	0.14	1330	1100	320

electric conductors were connected. The width w was chosen from considerations of preservation of a uniform distribution of the current over the cross section near T_c . The method used to produce the films and to measure their principal parameters was described earlier.^[12] The measurements were made in the interval $T=(4.2-3)$ K. The temperature was determined accurate to 0.002 K. The critical current I_c was determined from the current-voltage characteristics (CVC) of the films, as the current at which the voltage on the film reached $0.1 \mu\text{V}$. The parameters of certain films are given in the table.

The table lists the values of T_c obtained by extrapolating the function $I_c(T)$ to zero. From the measured values of the room-temperature resistance $(R_{300})_{\square}$ and the residual resistance $(R_N)_{\square}$ per square of surface^[12,13] we estimated the film thickness d , the mean free path of the electrons l , and the depth of penetration δ_{\perp} of the perpendicular magnetic field in the film.

The thickness d was estimated also from optical measurements using an interference microscope. For all the films, the relations $l \sim d < \xi_0$, $d \ll \lambda(T)$, and $d \ll \xi(T)$ were satisfied (where ξ_0 is the coherence length in the pure superconductor, $\lambda(T)$ is the depth of penetration of the weak magnetic field, and $\xi(T)$ is the coherence length in the film). The table lists the values of $\xi(0)$ and $\delta_{\perp}(0)$, which are formally obtained when the value $T=0$ is substituted in the formulas of the GL theory. The measured critical currents of the films I_c , compared with theory in the same manner as done by us earlier,^[12] coincided with the critical pair-breaking current with accuracy $\pm 10\%$.

B. Measurements at microwave frequencies

The sample was placed at the center of a three-centimeter waveguide perpendicular to its broad walls at the maximum (over the waveguide section) of the electric field parallel to the plane of the film. Behind the sample was located an adjustable short-circuiting plunger. The internal volume of the waveguide, containing the film, was evacuated. The fact that in this case the critical current agreed with the values measured when the sample was placed directly in the liquid helium indicates that good thermal contact existed between the film and the helium bath. The temperature dependences of the components of the admittance of the film $Y=G(T) - iB(T)$ were determined from measurements of the voltage standing wave ratio (VSWR) K (with accuracy ± 0.25) and the position of the minimum of the standing wave (with accuracy ± 0.07 mm) in the waveguide in accordance with the standard procedure.^[14] The power entering the waveguide with the film through the probe of the measuring line from the 9300-MHz klystron oscillator was not more than 10^{-9} W, so that the micro-

wave current in the film was much less than the critical value even at $T/T_c = 0.990$. Control experiments have demonstrated the absence of parasitic reactances capable of causing the minimum of the standing wave to drift when the temperature changed in the interval $T = (4.2$ to $2.2)$ K.

The current measurements were made by a synchronous detection method, where the registered signal was due to modulation of the reflection coefficient of the film (and of the short-circuiting plunger behind it) by means of rectangular current pulses of frequency 1 kHz.^[8]

C. Data reduction method

The equivalent circuit of a superconducting film at microwave frequencies^[15] is a series connection of a certain constant "geometric" inductance \mathcal{L}_g with a kinetic inductance \mathcal{L}_k , the latter being connected in parallel with the loss resistance \mathcal{R} . The values of \mathcal{L}_k and \mathcal{R} are given by

$$\mathcal{L}_k = \frac{1}{\sigma_0 \omega} \frac{L}{S} \frac{1}{J}, \quad \mathcal{R} = \frac{1}{\sigma_1} \frac{L}{S} \frac{1}{J}. \quad (1)$$

where J is the integral that depends on the distribution of the microwave current over the cross section S of the sample: $J = 1$ in the case of a uniform distribution of the current, and $J \approx 4\pi\lambda^2/S2 \ln 2$ in the case of a highly nonuniform distribution. The total impedance of the film is a series circuit consisting of the impedance of the narrow working strips and of the "shore" parts. For the shores we have $R \gg \omega \mathcal{L}_k$ and $\mathcal{L}_k \approx L2 \ln 2/c^2$, i. e., a broad film constitutes at microwave frequencies a constant inductance $\mathcal{L}_0 = \mathcal{L}_g + (L/c^2)2 \ln 2$, and the losses in it are small. This conclusion was confirmed by a control experiment carried out with a wide film.

The total admittance of the short-circuited waveguide segment with the film is given by $Y = G(T) - i[\beta(T) + B_0 + B_{sc}]$, where $B_0 = 1/\omega(\mathcal{L}_0 + \mathcal{L}_1)$, \mathcal{L}_1 is the geometric inductance of the narrow part of the film ($\mathcal{L}_1 \ll \mathcal{L}_0$), and B_{sc} is the susceptance of the short-circuited segment. In our experiments, B_{sc} was made equal to $-B_0$. To this end, the short-circuiting plunger was mounted at a distance $\lambda_w/4$ (λ_w is the wavelength in the waveguide) from the plane where the film was connected, and the position of the minimum of the standing wave was measured in its absence ($G = B = 0$), i. e., we established the reference plane at which $B_{sc} = 0$. Then at $T \ll T_c$, when $G \approx 0$ and $\beta \approx 0$, the same position of the minimum was re-established in the waveguide with the film by using the plunger, thus ensuring the equality $B_0 + B_{sc} = 0$ with accuracy of $\pm 5\%$ of B_0 .

From the expressions for $G(T)$ and $\beta(T)$ and $\omega \mathcal{L}_k \ll \omega \mathcal{L}_0$ (this condition was satisfied in the experiment at $1 - T/T_c \gg 10^{-3}$) we obtain

$$\frac{\sigma_1}{\sigma_N} \approx \frac{1}{J} \frac{Z_0 R_N}{X_0^2} \frac{1}{Z_0 G(1 + \beta^2/G^2)}, \quad \frac{\sigma_2}{\sigma_N} \approx \frac{1}{J} \frac{Z_0 R_N}{X_0^2} \frac{\beta/G}{Z_0 G(1 + \beta^2/G^2)} \quad (2)$$

where Z_0 is the characteristic impedance of the wave-

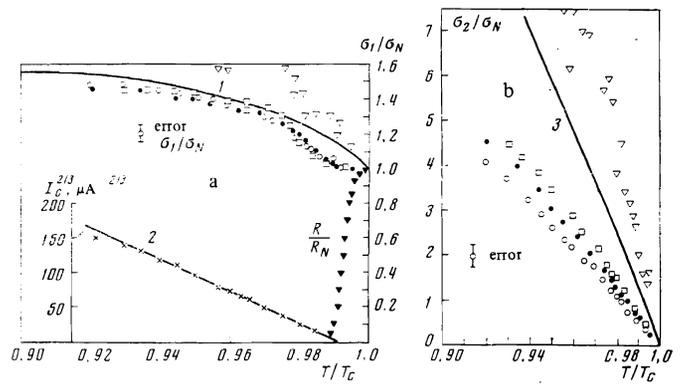


FIG. 1. Temperature dependences of the admittance σ_s of narrow Sn films at 9300 MHz in the absence of transport current: a) real part (σ_1); b) imaginary part (σ_2). \circ —sample No. 7; \bullet —sample No. 10; \square —sample No. 11; ∇ —experimental values from^[11]. Curves 1 and 3—dependences of σ_1 and σ_2 on the temperature in accordance with the Mattis–Bardeen theory. Curve 2—theoretical dependence of I_c^{GL} for sample No. 11, \times —values of I_c for sample No. 11, ∇ —values of $R(T)/R_N$ for sample No. 11.

guide, $X_0 \equiv 1/B_0$, and $R_N = L/S\sigma_N$ is the dc resistance of the sample at $T = 4.2$ K.

We note that at $T > T_c$ the condition $w \ll 2\delta_{sk}^2/d$ (where $\delta_{sk} = (c^2/2\pi\omega\sigma_N)^{1/2}$ is the depth of the skin layer) is satisfied for the investigated films, i. e., the microwave currents were uniformly distributed over the cross section of the film, and consequently $\mathcal{R} = R_N$. Since furthermore the condition $R_N \ll X_0$ is always satisfied, we have $Z_0 G(T_c) \approx Z_0 R_N/X_0^2$ for $T \geq T_c$. Thus, the quantity $Z_0 R_N/X_0^2$ that enters in (2) can be obtained by measuring the VSWR at $T \geq T_c$:

$$(Z_0 R_N/X_0^2) \approx K^{-1}(T_c).$$

When the current measurements were made, the registered signal was proportional to the change of the modulus of the reflection coefficient of the film, i. e., to the change of σ_2 by an amount^[1] $\Delta\sigma_2$. To calibrate the quantity $\sigma_2(I)/\sigma_2(0)$, this signal was compared with the signal measured after the superconductivity of the sample was completely destroyed by the current.

Another calibration method was to compare the measured signal with the signal at 100% modulation of the microwave power. Both calibration methods yielded values $\sigma_2(I)/\sigma_2(0)$ that agreed within 10%.

2. EXPERIMENTAL RESULTS

Figures 1a and 1b show plots of σ_1/σ_N and σ_2/σ_N against the relative temperature for typical samples whose parameters are listed in the table. For comparison, the figures show also the results of^[11]. The solid lines show the corresponding plots resulting from the Mattis–Bardeen theory. In the reduction of the results of the microwave measurements, the critical temperature was determined by linear extrapolation of the function $\sigma_2(T)$ to zero. At the value of T_c determined in this manner, the film resistance was usually $R(T_c) = (0.95 - 1.0)R_N$.

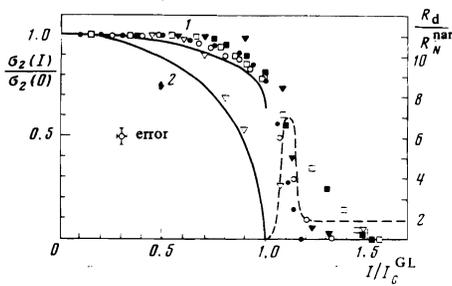


FIG. 2. Imaginary part σ_2 of the admittance of the film vs. the transport current. Numbers of the samples and values of T/T_c : ■—No. 10; 0.945. □—No. 10; 0.910. ●—No. 11; 0.950. ○—No. 11, 0.930. The experimental values from [11] are: ▼— $T/T_c = 0.965$. ▽— $T/T_c = 0.988$. Curve 1—theoretical dependence (5) ($\omega\tau_\Delta \gg 1$), curve 2—theoretical dependence (6) ($\omega\tau_\Delta \ll 1$). Dashed curve—dependence of the differential resistance R_d/R_N^{nar} on the current for sample No. 11.

Figure 1a shows also a plot of $R(T)/R_N$ and the dependence of the critical current I_c on the temperature for sample No. 11. The solid line is the theoretical plot of I_c^{GL} :

$$I_c^{GL} = \frac{c\Phi_0}{6.3 \cdot \pi^2 \xi \delta_\perp} w. \quad (3)$$

We note that extrapolation of the $I_c(T)$ plot to zero yields a value of T_c that is approximately 0.03°K lower than the value of T_c determined by the microwave method. This is a reflection of the fact that $\sigma_2 \neq 0$ when superconducting nuclei appear, whereas the appearance of I_c calls for the formation of a superconducting channel in the entire sample.

Figure 2 shows plots of $\sigma_2(I)/\sigma_2(0)$ against the normalized current for samples 10 and 11 at various temperatures in the interval $T/T_c = 0.91$ to 0.95, where the non-uniformity of the current distribution over the film cross section could not exceed 20%. These plots agree well with one another for all the investigated samples in the interval $T/T_c = 0.91$ –0.98. Figure 2 shows also the results of Churilov, Dmitriev, and Svetlov. [11]

Figure 3 shows the current-voltage characteristic of film No. 11 (the current is normalized to I_c^{GL}).

All the investigated films have a common feature—the existence of a resistive region with $R < R_N$ in a rather wide current interval. It can be seen from Figs. 2 and 3 that $\sigma_2(I)$ decreases to zero at currents when the differential resistance R_d of the sample reaches a value equal to the resistance R_N^{nar} of the narrow part of the film. With further increase of the current, the CVC is parallel to the line $V = IR_N^{nar}$ with a certain excess current.

3. DISCUSSION OF RESULTS

We discuss first the behavior of the high-frequency admittance in the absence of transport current (Figs. 1a and 1b). For all the investigated samples good agreement is observed between the experimental plots of σ_1/σ_N and the theoretical relation constructed with the aid of the table in [16].

According to the Mattis–Bardeen theory, the increased energy absorption in comparison with the absorption in the normal phase (i. e. $\sigma_1 > \sigma_N$) at $\hbar\omega \lesssim 2\Delta(T)$ is due to the presence of singularities in the state density at the edges of the gap.

The dependence of the imaginary part of the admittance σ_2 on the temperature at $\hbar\omega \ll 2\Delta(T)$ takes the form [1]

$$\frac{\sigma_2}{\sigma_N} \approx \frac{\pi\Delta}{\hbar\omega} \text{th} \left(\frac{\Delta}{2kT} \right). \quad (4)$$

According to (4), a linear increase of σ_2 should be observed near T_c with increasing T . The relation (4) is represented in Fig. 1b by a solid curve. We see that the character of the temperature dependence of the measured quantities σ_2/σ_N agrees with the predictions of the theory. A discrepancy between experiment and theory with respect to the slope $d\sigma_2/dT$, similar to that obtained by us, was observed before [17] and remains unexplained. It is possible that this discrepancy is due to neglect, in the reduction of the experimental results, of the delay of the electromagnetic field in the film, which can lead to a certain change in the slope [2] $d\sigma_2/dT$.

We proceed now to a discussion of the dependence of the high-frequency susceptance σ_2 on the current. At present there is no microscopic theory capable of describing the dependence of σ_2 on the current in gap-type superconductors at arbitrary values of $\omega\tau_\Delta$. For the limiting cases $\omega\tau_\Delta \gg 1$ and $\omega\tau_\Delta \ll 1$ we can write down [7, 15] relations between σ_2 and the current, inasmuch as in the former case the order parameter depends only on the time-averaged value of the square of the field, and in the latter case the adiabatic regime is realized:

$$\frac{I}{I_c^{GL}} = \frac{3\sqrt{3}}{2} \frac{\sigma_2(I)}{\sigma_2(0)} \sqrt{1 - \frac{\sigma_2(I)}{\sigma_2(0)}} \quad \text{at } \omega\tau_\Delta \gg 1, \quad (5)$$

$$\frac{I}{I_c^{GL}} = \left(1 + \frac{1}{2} \frac{\sigma_2(I)}{\sigma_2(0)} \right) \sqrt{1 - \frac{\sigma_2(I)}{\sigma_2(0)}} \quad \text{at } \omega\tau_\Delta \ll 1. \quad (6)$$

Plots of (5) and (6) are shown in Fig. 2 by solid lines.

As seen from Fig. 2, at $I \leq I_c^{GL}$ there is good agreement between the experimental $\sigma_2(I)/\sigma_2(0)$ plots and the theoretical relation (5), which is valid at $\omega\tau_\Delta \gg 1$. Since this agreement took place in the interval $T/T_c = 0.91$ –0.98 we can estimate the lower bound of τ_Δ : at $T/T_c \geq 0.91$ we have $\tau_\Delta \gg 10^{-11}$ sec. This greatly exceeds the “diffusion” relaxation time at the same temperature [5].

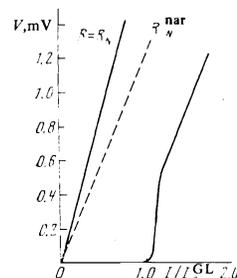


FIG. 3. Current-voltage characteristic for sample No. 11 at $T/T_c = 0.950$. The solid line is the CVC at $T > T_c$. The dashed line corresponds to the resistance of the narrow part of the film, R_N^{nar} .

$$\tau_{\text{diff}} [\text{sec}] = 3 \cdot 10^{-12} / (T_c - T) [\text{K}].$$

On the other hand, it does not contradict the measurement results of Peters and Meissner^[9], $\tau_{\Delta}(0.91) [\text{sec}] = 8.6 \times 10^{-10}$, nor the estimates of the time of homogeneous gap relaxation given in the theoretical papers.^[6,18,19]

At currents $I > I_c^{\text{GL}}$, experiment shows a smooth decrease of σ_2 with increasing current. To explain this behavior of σ_2 it is necessary to understand the details of the process of the transition of the superconducting film to the normal state under the influence of a current larger than the pair-breaking current. From the form of the CVC in Fig. 3 it is seen that there is a current interval where the film is in a resistive state with $R < R_N^{\text{nar}}$. This resistive region is possibly connected with the formation in the film of a one-dimensional layered structure.^[20-22] It should be noted, however, that at $\xi < w$ and $\xi < \delta_1$ there can exist in the film also a two-dimensional dynamically mixed state, inasmuch as at $I \approx I_c^{\text{GL}}$ the edge potential barrier, which hinders the entry of the vortices, is suppressed.^[15]

The absence of hysteresis on the CVC, as noted by Eru *et al.*,^[23] indicates in all probability that thermal effect plays a negligible role in our case.

From a comparison of Figs. 2 and 3 we see that σ_2 vanishes at currents when the differential resistance R_d of the film reaches a value equal to the resistance R_N^{nar} of the narrow part of the film, although a certain excess current does flow.

CONCLUSION

1. In the absence of transport current the temperature dependences σ_1 of Sn films agree with the microscopic Mattis-Bardeen theory in the temperature region where the distribution of the current over the cross section of the film is uniform. At the same time, the slope $d\sigma_2/dT$ of the $\sigma_2(T)$ function deviates somewhat from the theory.

2. In the current region $I \leq I_c^{\text{GL}}$ there is good agreement between the measured values of $\sigma_2(I)/\sigma_2(0)$ and the theory, which is valid at $\omega\tau_{\Delta} \gg 1$, and this yields a lower-bound estimate $\tau_{\Delta} \gg 10^{-11}$ sec in the temperature region $T/T_c \geq 0.91$. This estimate of τ_{Δ} does not contradict the experimental results of Peters and Meissner, nor the theoretical estimates of^[6,18,19]

For a more exact measurement of τ_{Δ} near the critical temperature it is thus necessary to carry out experi-

ments at frequencies lower than ~ 1 GHz.

The authors are grateful to A. F. Volkov for fruitful discussions and to V. A. Smirnov for help with the measurements.

- ¹⁾Inasmuch as in the absence of current there is little difference between σ_1 and σ_N (see below), the dependence of σ_1 on the current, at least at $I < I_c^{\text{GL}}$, can be neglected. The same is evidenced also by additional experiments performed by us.
²⁾Of course, the delay does not affect the measured values of $\sigma_2(I)/\sigma_2(0)$, which are relative.

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