

In general the work of a surface is determined experimentally by the use of electrons with a phase wavelength of the order of a few tens of angstroms, while at the same time diffraction patterns are measured by using electrons with wavelengths of only a few angstroms. Because of this, surfaces with the same average work function may give different diffraction patterns, depending upon their degree of homogeneity. Hence it must be concluded that the change $\Delta\phi$ in work function is not by any means a single-valued function of the state of a composite monatomic surface.

In conclusion, I wish to express my thanks to Prof. N. D. Morgulis for suggesting the subject, and for his constant interest in the work.

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Translated by D. C. West

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SOVIET PHYSICS JETP

VOLUME 34 (7), NUMBER 1

JULY, 1958

MEASUREMENT OF THE SURFACE IMPEDANCE OF SUPERCONDUCTORS AT 9,400 MCS

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Received by JETP editor July 17, 1957

J. Exptl. Theoret. Phys. (U.S.S.R.) **34**, 14-22 (January, 1958)

The surface impedance of several superconductors has been measured at a frequency of 9400 Mcs. The temperature dependence of the surface impedance of thin films of Sn and Hg has been studied. The penetration depth was investigated for these films and found to be in accord with the critical magnetic field measurements. The effective conductivity of the films in the superconducting state increases as the temperature is reduced. An investigation was also made of the relation between the impedance of thin Sn and Hg films and the magnetic field at $T < T_0$. The dependence of penetration depth on field strength is in satisfactory agreement with the Landau-Ginzburg theory.

1. A knowledge of the complex impedance of a superconductor at frequencies on the order of 10^{10} cps makes it possible to find the dielectric permittivity of the metal:

$$\epsilon = \epsilon_0 - c^2 / \omega^2 \delta_0^2 \quad (1)$$

Determining the penetration depth of the magnetic field δ_0 from static measurements, it is then possible to estimate the value of ϵ_0 due to the presence of bound electrons in the superconductor.^{1,2} On the other hand, if $\epsilon_0 \ll c^2 / \omega^2 \delta_0^2$, it is possible, using these measurements, to find the penetration

depth δ_0 of the static magnetic field into the superconductor. Since in a high-frequency field the superconducting electrons do not completely screen the effect of the electromagnetic field on the normal electrons, impedance measurements also furnish data on the normal conductivity of a superconducting current. On the basis of the data reported by Pippard and Simon,^{3,4} who measured the total surface resistance of lead at 9400 Mcs, it may be assumed that $\epsilon_0 = 2 \times 10^8$ cgs esu at $T = 3^\circ\text{K}$ and that below T_c the ratio of normal-electron conductivity to free path length σ_N/l falls off as the temperature is reduced. Khikin has investigated the surface resistance of thin, superconducting tin layers at the same frequency.^{5,6} However, the accuracy of these experiments was not sufficient to justify the conclusion which was reached, namely, that ϵ_0 is large ($\sim 10^9$).

The present work is an extension of experiments in this same subject based on a new and more comprehensive method proposed by Khaikin.

2. The method of investigating the surface impedance $Z = R + iX$ is based on the study of the properties of a hollow, copper, cylindrical resonator, the bottom of which is made from the material being investigated. Since the cavity is excited in the TE_{011} mode, no current flows across the joint between the bottom and the walls of the resonator; thus the nature of the contact between the sample and the copper walls of the resonator is unimportant.

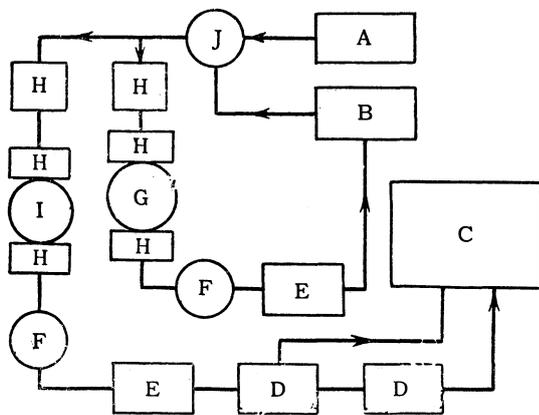


FIG. 1. Block diagram of the system used for measuring the frequency width of the resonator and shift in resonant frequency. A) GSS-6; B) Sawtooth generator; C) EPP-09; D) Outputs I and II; E) Amplifier; F) Detector; G) Standard Cavity Resonator; H) Attenuator; I) Cavity Resonator; J) K-19.

In the experiment, direct measurements are made of the widths of the frequency characteristics and of the change in the resonant frequency of the resonator (Fig. 1). The frequency of the K-19

klystron is modulated periodically by a sawtooth voltage which is applied to the repeller. When the klystron frequency passes through the cavity resonance, a signal is observed at the detector; this signal is proportional to the power transfer coefficient, i.e. it corresponds to the resonator frequency characteristic. The same oscillator excites a standard resonator made of lead and maintained at liquid helium temperature (4.2°K). Since lead is a superconductor below 7°K , this resonator has an extremely narrow pass band (approximately 1 kcs).

The signal from the standard resonator is used as a reference, relative to which the frequency shift of the test cavity is measured. To obtain an absolute frequency scale, a radio-frequency voltage (500 — 5,000 kcs) from a standard signal generator is applied to the repeller of the klystron in addition to the sawtooth voltage. Because the klystron is frequency modulated, its spectrum contains side bands and the signal from the standard resonator has three narrow peaks. The distance between these peaks is determined by the frequency of the standard signal generator and serves as an absolute frequency scale with which the shift of the resonant frequency and the width of the frequency characteristic of the resonator can be measured.

A more precise determination of these quantities is obtained by the use of a special circuit with which the frequency characteristic of the resonator is differentiated twice, producing the following: (a) a voltage proportional to the spacing between the reference point and the point at which the first derivative changes sign, characterizing the change of the resonance frequency of the resonator; (b) a voltage proportional to the distance between the points at which the second derivative of the frequency characteristic change sign, yielding the width of the frequency characteristic of the resonator. Both voltages are recorded by an EPP-09 recording potentiometer. The total accuracy of the measurement circuit is one per cent. To find R and X it is necessary to determine two different quantities: the shift of the resonance frequency of the resonator as the sample makes the transition into the superconducting state and the change in half-width of the resonance curve due to the losses in the sample. The error in the determination of these quantities is about 2 per cent and depends on the proximity to the transition temperature. The probable errors in the determination of ϵ , σ_N/l and σ_{eff} are shown in the appropriate figures.

The resonator is a copper cylinder with an inner diameter of 43 mm. The resonator is coupled to the K-19 oscillator and the detector through two

circular irises 5 mm in diameter cut out of the upper (copper) cover of the resonator; the irises are located symmetrically along a diameter, at a distance of one-half the radius from the center. The irises are terminated by two lengths of circular wave guide with diameter smaller than the critical value; these are used as limiting attenuators between the resonator and the outer coaxial lines. To avoid effects due to motion of the coupling loop on the resonator frequency, the lengths of the attenuators are not changed during the experiment; for this reason the coupling coefficient varies with changing impedance of the sample. The coupling coefficient is never greater than 0.3. The bottom of the resonator is an optically polished plane-parallel slab of smooth quartz, 46 mm in diameter and 3–5 mm in thickness, to which the sample is clamped from the inside. The mercury films being investigated are condensed on the outside of this same slab. The quartz slab is used because the amplitude of the field which acts on the sample is considerably greater than the amplitude of the field acting at the other walls of the resonator because the quartz tends to "concentrate" the field (the dielectric constant of quartz is 3.6); the net result is an increase in the relative fraction of high-frequency resistance of the resonator in the end wall, i.e., the sample being investigated, compared with the rest of the cavity.

The sensitivity is increased by a factor of 3.1 when the quartz slab is 3 mm thick and the resonator length is 39 mm and by a factor of 30 when the thickness of the slab is 5 mm and the length of the resonator 20 mm (in the latter case it is necessary to introduce a correction for the thermal expansion of the quartz in the helium temperature region).

The test cavity is enclosed in a metal jacket in which gaseous helium at a pressure of 20μ is admitted (at $T = 4.2^\circ\text{K}$) to provide thermal contact between the resonator and the helium bath. The temperature of the resonator is measured with a thermometer made from a carbon resistor. The voltage drop across the carbon thermometer is recorded by the EPP-09 recording potentiometer together with the resonator characteristics. The error in the temperature determination is less than 0.005°K . In special control experiments with a copper resonator it was established that the high-frequency resistance of the carbon remains constant within the limits of the experimental accuracy. Since the experiments are always carried out with the coupling loop fixed (fixed length for the attenuators between the resonators and the external lines) all changes in the resonator characteristics can be ascribed to changes in the proper-

ties of the sample which makes the transition from the normal to the superconducting state. Knowing the change in the width Δf of the frequency characteristic of the resonator produced by losses in the sample and the shift df in the resonance frequency of the resonator, and knowing the geometry of the resonator, it is possible to determine the real part R of the sample impedance and the change in the reactive part dX . To determine the total reactance of the sample one must also know the reactance in the normal state X_n . The total reactance is given by $X = X_n + dX$.

In large samples, where there is an anomalous skin effect, $X_n = \sqrt{3}R_n$, where R_n is the resistance of the sample in the normal state, known from experiment. For thin films, the thickness of which is considerably less than the skin depth, $X_n \ll R_n$ to that dX is comparable with R_n ; hence, the change in reactance measured in the experiment is essentially the total reactance of the thin layer in the superconducting state.⁵

3. To test the method, the surface impedance of a single-crystal sample of tin of high purity ($\rho_{300}/\rho_{4.2} = 4.10^4$) was measured. The temperature dependence of the dielectric permittivity ϵ of the sample and the ratio of the normal-electron conductivity to free path length σ_N/l , as computed by Abriksov,¹ are shown in Fig. 2. In this same figure are shown the limits of accuracy for the

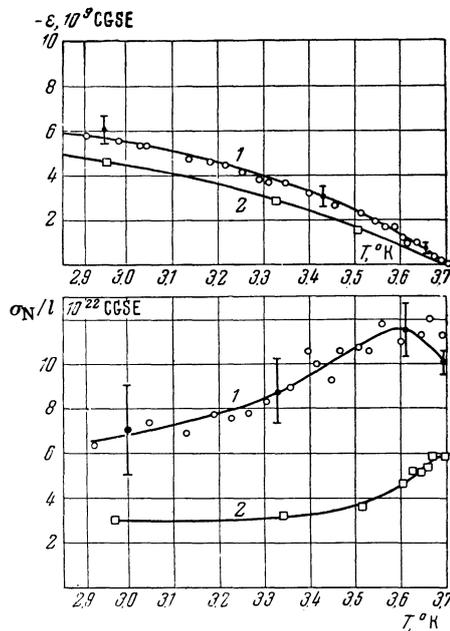


FIG. 2. Dielectric permittivity and the ratio of normal conductivity to free path length in bulk tin. \circ — results of the present work; \square — results obtained by analysis of the data of Ref. 3 at $\theta = 62^\circ\text{K}$.

measurements of ϵ and σ_N/l at various temperatures. Assuming $\epsilon_0 \ll c^2/\omega^2\delta_0^2$, we can estimate the depth of penetration of the magnetic field into the superconductor δ_0 . In the present experiments this quantity obeys the well-known relation between static depth of penetration of the magnetic field and temperature

$$\delta_0 = \delta_{00}/\sqrt{1 - (T/T_K)^4}. \quad (2)$$

The value of δ_{00} is in good agreement with the data obtained from dc measurements.^{7,8} It follows that ϵ_0 is smaller than the accuracy of the present measurements, i.e., virtually the entire dielectric permittivity of the superconductor is determined by the superconducting electrons; for $T = 3^\circ\text{K}$, $\epsilon_0 < 3 \cdot 10^8$. Directly below T_C there is a systematic increase in σ_N/l , the magnitude of which is approximately the same as the limits of accuracy of the experiment; as the temperature is reduced beyond this point, σ_N/l is reduced. Similar results are obtained for ϵ and σ_N/l if one analyzes the data of Refs. 3 and 4 using the Abrikosov formula.¹ The discrepancy in the absolute values of ϵ and σ_N/l is probably a result of anisotropic effects in tin.

4. Thin mercury films were also investigated. These measurements were made conveniently at liquid helium temperature in the assembled apparatus. The film of mercury was condensed on the optically polished quartz slab which served as the end of the resonator. The evaporator was a copper cup with a nichrome heater in which a drop of mercury was placed. Before evaporation of the film, the heat-exchange gas was completely evacuated from the apparatus by means of an adsorption pump. A power of approximately 0.1 watts was applied to the heater. Under these conditions the temperature of the quartz slab increased at a rate of approximately $0.5^\circ/\text{min}$. In order to avoid temperature rises of more than 2–3 degrees, the precipitation of thick mercury films was carried out in steps. Films with a thickness $d = (5.7 - 36) \times 10^{-6}$ cm were studied. The critical temperature of freshly deposited mercury films was found to be somewhat lower than the critical temperature for bulk mercury (4.15°K), lying in the region $3.9 - 4^\circ\text{K}$. Impedance measurements were made on freshly deposited and recrystallized mercury films; the latter were annealed at boiling nitrogen temperatures for a period of 18–20 hours. The critical temperature of the recrystallized films was $4.15 \pm 0.01^\circ\text{K}$.

The thickness of the films was computed by weighing; the film was weighed by collecting the mercury of the film with the blade of a steel knife.

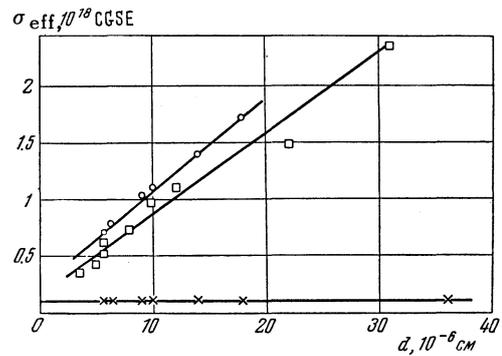


FIG. 3. The effective conductivity of thin films in the normal state as a function of thickness: \times — freshly-deposited mercury films, \circ — recrystallized mercury films, \square — tin films.

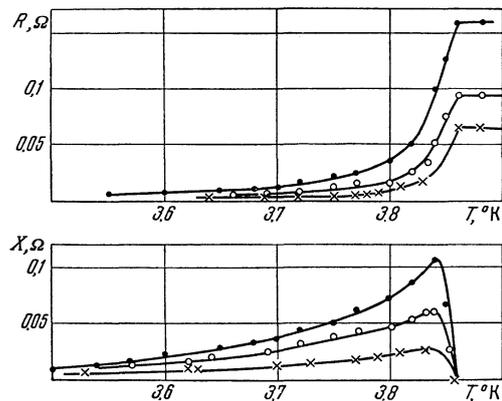


FIG. 4. Resistance and reactance of superconducting thin tin films. The film thickness is given in 10^{-6} cm: \bullet — 5.35; \circ — 7.7; \times — 12.

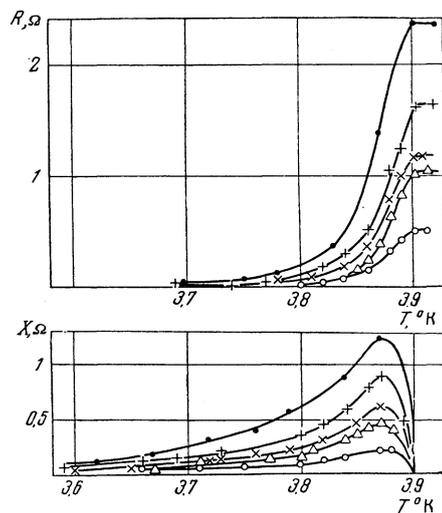


FIG. 5. Resistance and reactance of freshly-deposited superconducting films of mercury. The film thickness is given in 10^{-6} cm: \bullet — 6.4, $+$ — 10, \times — 14, Δ — 18, \circ — 36.

The error in the determination of the thickness was less than 2–3 per cent. The difference in film thickness between the center and the edge was less than 10 per cent.

Thin tin films were made by deposition of spectrally pure tin (99.918) on a glass plate at room temperature. Film with thicknesses of $(3.3 - 33.2) \times 10^{-6}$ cm were studied. The critical temperature of the tin films (3.8–3.9°K) were somewhat above the critical temperature for bulk tin (3.7°K).

The effective conductivity σ_{eff} in the normal state (cf. Fig. 3) was determined from the high-frequency resistance of the mercury and tin films at temperatures above T_c . For tin and recrystallized mercury films σ_{eff} is in good agreement with the theoretical relation $\sigma_{\text{eff}} \sim \sigma_N d / \ell$ (ℓ is the electron free path in the bulk metal) obtained under the assumption that electron diffusion scattering takes place at the boundaries of the film, i.e., that the electron free path in the metal is limited to the thickness. In the unannealed mercury films σ_{eff} was found to be independent of d . This would seem to mean that films condensed at low temperatures have extremely fine crystal structure and are essentially amorphous. The electron free path in this film is considerably less than the thickness of the film. From a comparison of the absolute magnitudes of σ_{eff} in freshly deposited and recrystallized mercury films it may be assumed that $\ell \approx 5 \times 10^{-7}$ cm in the freshly deposited films.

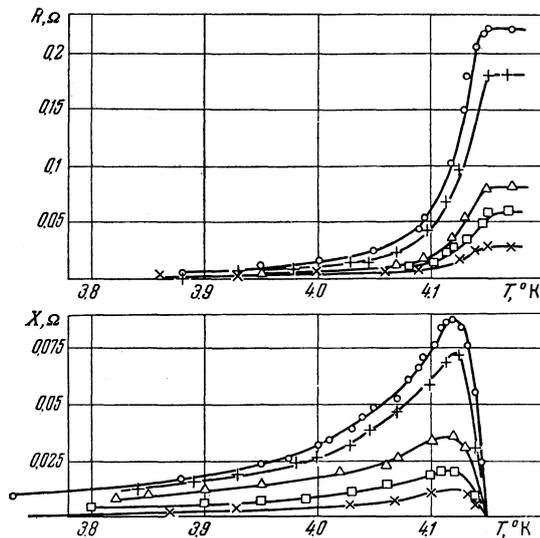


FIG. 6. Resistance and reactance of recrystallized superconducting mercury films. The film thickness is given in 10^{-6} cm: \circ —5.7; $+$ —6.4; Δ —10; \square —14; \times —18.

The change of surface impedance with temperature as the film goes from the normal to the super-

conducting state is shown in Figs. 4, 5, and 6. Using the Ginzburg formula,² we can compute ϵ and σ_{eff}

$$\epsilon = -\frac{4\pi X}{\omega d |Z|^2}, \quad \sigma_{\text{eff}} = \frac{1}{d} \frac{R}{|Z|^2}, \quad (3)$$

this formula is valid when $d^2 \ll \delta_{\text{SC}}$. This condition is satisfied for films in the normal state and at temperatures 0.3–0.4 degrees away from T_c . All calculations were carried out for temperatures close to T_c since Eq. (3) does not apply at lower temperatures; furthermore, there is a significant reduction in the accuracy of the measurements of R and dX .

The results of the calculations of ϵ and σ_{eff} are shown in Figs. 7–12. Assuming that the entire dielectric permittivity of the film is due to the superconducting electrons, i.e., $\epsilon_0 \ll c^2/\omega^2\delta_0^2$, it is possible to compute the depth of penetration of the static magnetic field into the superconductor δ_0 and to estimate δ_{00} , using the relation between δ_0 and temperature (2); this relation is found to be in good agreement with the obtained results.

For freshly deposited films $\delta_{00} = (14.5 \pm 1.5) \times 10^{-6}$ cm; for recrystallized films $\delta_{00} = (5.5 \pm 0.5) \times 10^{-6}$ cm. The latter value agrees with the value $(5.3 \pm 0.5) \times 10^{-6}$ cm obtained by Khukhorova⁹ in measurements of δ_{00} in the same films using the critical magnetic field method. For the

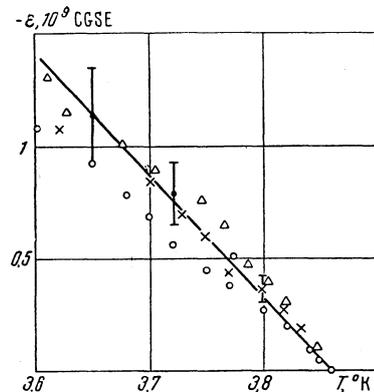


FIG. 7. Dielectric permittivity of superconducting tin films. The quantity d is given in 10^{-6} cm: \circ —5.35; \times —7.7; Δ —12; \bullet —22.

tin films $\delta_{00} = (6.5 \pm 0.5) \times 10^{-6}$ cm, which is in agreement with the data reported by Zavaritskii.¹⁰ In all films which were investigated σ_{eff} increases as the temperature is reduced in the vicinity of T_c . We may note that an increase of σ_N/ℓ with temperature was noted also in bulk tin (Fig. 2) at temperatures close to T_c . This result is not as yet understood.

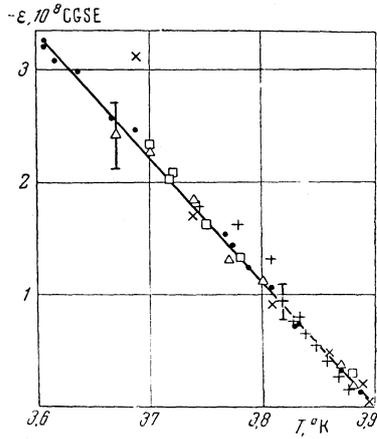


FIG. 8. Dielectric permittivity of freshly-deposited superconducting mercury films. The quantity d is given in 10^{-6} cm: ●—36; +—18; □—14; △—10; ×—7.7.

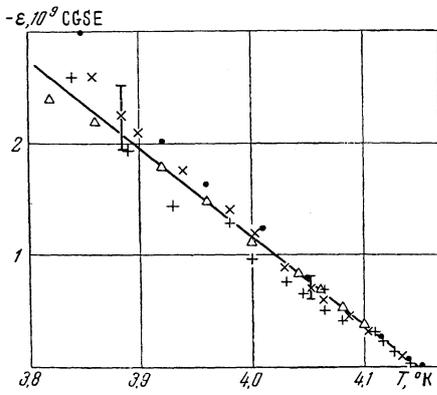


FIG. 9. Dielectric permittivity of recrystallized superconducting mercury films. The quantity d is given in 10^{-6} cm: ●—18; ×—19; +—6.6; △—10.

5. An investigation was also made of the relation between the surface impedance of the superconductor and a fixed magnetic field $H < H_C$. The experiments were carried out with bulk samples and thin films. In order to interpret the results of these experiments it is important that the magnetic field be precisely parallel to the surface of the sample being investigated. In a bulk sample this requirement is automatically satisfied because of the Meissner effect. In thin films the critical magnetic field $H_{C\parallel}$ parallel to the film is several times larger than the field perpendicular to the film $H_{C\perp}$, i.e., when $T < T_C$ the resistance of film is a minimum when the magnetic field is parallel to the plane of the film. This property was used to ensure that the plane of the film was parallel to the magnetic field.

The impedance of the bulk sample of tin remained unchanged, to within an accuracy of 2 per

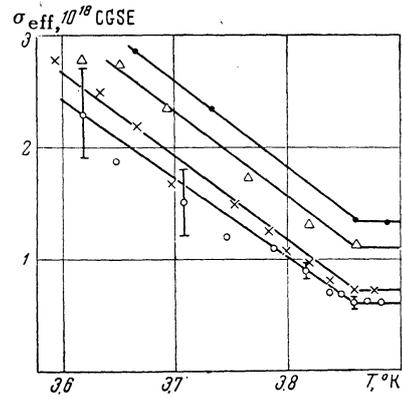


FIG. 10. Effective conductivity of superconducting tin films. The quantity d is given in 10^{-6} cm: ○—5.35; ×—7.7; △—12; ●—22.

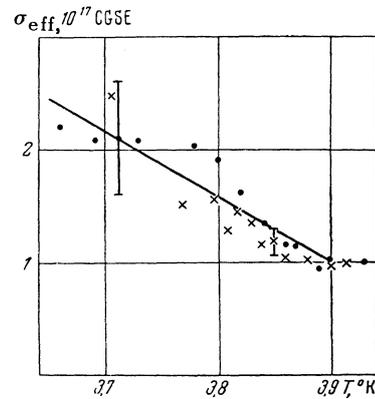


FIG. 11. Effective conductivity of freshly-deposited superconducting mercury films. The quantity d is given in 10^{-6} cm: ●—36; ×—18.

cent, as the field was increased up to $0.91 H_C$. Consequently, the depth of penetration of the field into the superconductor δ_0 is independent of field (within these same limits). This finding is in agreement with the results obtained by Pippard³ and is consistent with the Landau-Ginzburg theory,¹¹ according to which δ_0 in a bulk sample should not change by more than 2 per cent as the magnetic field is varied. In thin samples, the thickness of which is comparable with the depth of penetration at a given temperature, the Landau-Ginzburg theory gives the following dependence of penetration depth on field:

$$\delta_0(H) = \delta_{00} / \sqrt{1 - (H/H_C)^2}. \quad (4)$$

The results of the present experiments with mercury films and tin films are in good agreement with this theoretical relation (Fig. 13). A discrepancy between theory and experiment is found only

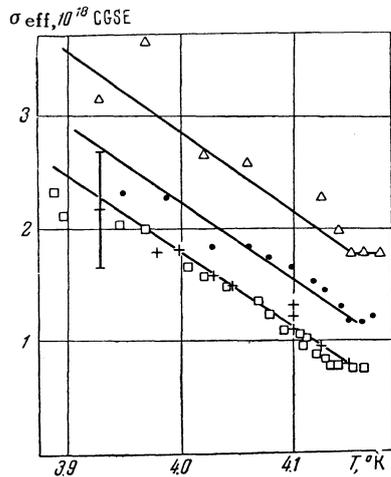


FIG. 12. Effective conductivity of recrystallized superconducting mercury films. The quantity d is given in 10^{-6} cm: Δ —18; \bullet —14; $+$ —6.4; \square —5.3.

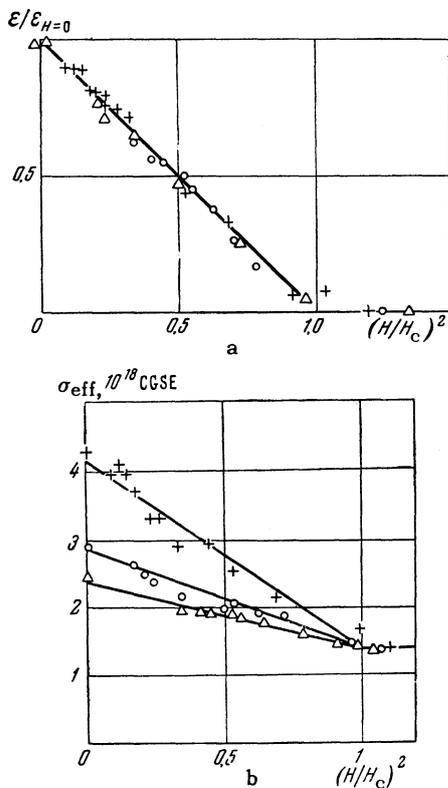


FIG. 13. The relation between dielectric permittivity (a) and effective conductivity (b) and the magnetic field for a tin film of thickness $d = 22 \times 10^{-6}$ cm. Δ — $T = 3.754^\circ\text{K}$, \circ — $T = 3.724^\circ\text{K}$, $+$ — $T = 3.658^\circ\text{K}$ ($T_c = 3.918^\circ\text{K}$).

in the region close to H_c ; this discrepancy is probably due to the fact that in a magnetic field the transition of thin films from the normal state to the superconducting state is not a sharp one.

In all the films which were investigated σ_{eff} is independent of magnetic field in the normal state. In the superconducting state σ_{eff} falls off as the field is increased, approaching the values of σ_{eff} in the normal state at $H = H_c$.

The author is deeply indebted to A. I. Shal'nikov under whose direction the present work was carried out, to M. S. Khaikin for his continued interest and a number of valuable discussions, and to V. L. Ginzburg and B. M. Bolotovskii for a discussion of the results.

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