

DESTRUCTION OF SUPERCONDUCTIVITY IN THIN FILMS BY FIELD AND CURRENT

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Measurements were made on the critical magnetic fields and currents required to destroy superconductivity in thin cylindrical tin films. Qualitative agreement with the Ginzburg-Landau theory was obtained. The structure of the films studied is discussed.

EXPERIMENTAL data on the properties of superconductors with small dimensions, whilst at the time substantially assisting the development of the London and Ginzburg-Landau theories, have not lost their significance in the new microscopic theory.

The aim of the present work was to try to establish the conditions for the destruction of superconductivity in thin tin films by magnetic fields and currents. Although a large number of works on thin films have appeared, we know of no attempts to study systematically a series of identical specimens fabricated simultaneously under identical conditions.

We studied films of various thicknesses which were made in the form of thin cylinders with a large ratio of length to diameter. As is known, when superconductivity in thin films is destroyed by a current, there must be no edges, since the magnetic fields at them attain the critical values for very small currents. The intermediate state arising at the film edges causes Joule heating, which increases avalanche-like and leads to a rapid transition into the normal state for the entire film. The only specimen geometries for which there are no edges are discs and cylinders. Disc specimens were used by Alekseevskii and Mikheeva;¹ however, during their fabrication a number of difficulties arose which from our point of view make work with them rather inconvenient. An attempt to use cylindrical films was made some years ago² and gave encouraging results. Now we have greatly improved this method.

Although, as will be seen from what follows, we did not succeed in completely avoiding the region of anomalously small destructive currents as occurred also when studying discs, nevertheless the method we chose was certainly suitable for solving the problem posed.

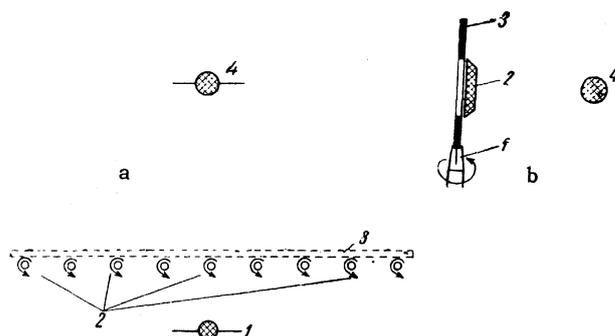


FIG. 1. a) 1—specimen evaporator, 2—specimens, 3—copper screen, 4—evaporator for contacts. b) 1—holder, 2—copper screen, 3—specimen, 4—evaporator for contacts.

EXPERIMENTAL METHOD

The tin films studied (purity of the original tin 99.998%) were deposited on glass rods measured to an accuracy of 0.01 mm which were 0.6 to 0.8 mm in diameter and 24 mm long. The carefully washed rods (nine at a time) were fixed in vertical holders on a special revolving apparatus under a vacuum bell-jar and were continuously rotated at a rate of 8 rpm while the films and contacts were condensed. The process of making the films lasted 5 to 8 minutes and the subsequent deposition of contacts approximately 30 minutes. The disposition of evaporators and substrates is clear from Fig. 1a.; the fixing of the glass rods and the position of the cooled copper screen are shown in Fig. 1b. The screen cross section chosen ensured an adequately sharp transition in thickness from the contact layer to the specimen layer, and also minimized the chance of depositing on to the specimen layer any additional material during contact evaporation. During evaporation (from a previously used tungsten evaporator) the pressure was less than 10^{-6} mm of mercury, and the mean surface temperature of the glass rods while the

specimen layer was being deposited did not apparently exceed $40 - 60^\circ\text{C}$. The tungsten wire crucible for depositing the contact layers was repeatedly fed with tin wire, so that the amount of tin evaporated from it exceeded the amount of tin used to make the specimen layer by a large factor (50 to 100). The amount of substance evaporated on to the specimen was calculated originally from the change in weight of the evaporator and the geometry. To check this calculation we made experiments to determine directly the amount of substance deposited on the specimen, using radioactive tracers.* To do this, strips of thin high-purity aluminum foil were fixed on the screen behind the specimens, alternating with the series of films; the strips, together with a quantity of the original tin were irradiated in a pile and then allowed to stand for several days. After standing one could take the measured activity, after allowing for the background from the foil, to be determined only by the gamma radiation from the Sn^{117} isotope. From several similar experiments, it was found that the relative distribution on the specimens agreed very satisfactorily with the distribution calculated from purely geometrical considerations, but that quantitatively from experiment to experiment the amount exceeded that calculated by a factor of 1.5 to 2. Such a discrepancy destroys the validity of the method and was caused by uneven evaporation of the tin from the surface of the tungsten crucible. Our results were referred to the quantity of substance deposited on the specimen as determined from the experiments with active tin.

In what follows we provisionally describe the separate specimens not by the amounts of substance deposited on them, but by the thicknesses (the density of the condensate was taken to be 7.0 g/cc). Special attention was paid to mounting the finished films in the apparatus and to making reliable current and potential contacts to them.

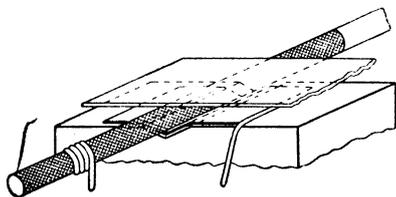


FIG. 2

The specimen mounting and the method of attaching contacts are shown in Fig. 2. The potential contacts were mechanical springs of thin tin foil pressed between the jaws of the holder and the

*We thank I. S. Shapiro and I. A. Antonova for help with these measurements.

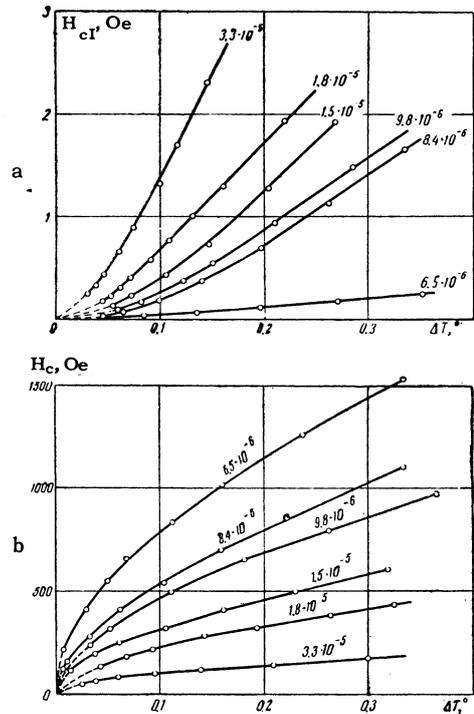


FIG. 3

thick contact layer of tin on the specimen. The current leads were made from three or four close turns of soft tinned copper wire (dia. 50μ) — previously soldered over with a tin-lead solder (melting point 140°C) on a glass rod of the same diameter as the specimen. The short cylinders thus made were tightly put on to the mounted specimen and soldered to it without the use of flux. Several specimens with films of different thickness were mounted parallel to one another in the bottom of a Dewar filled with liquid helium and placed between the poles of an electromagnet. The Dewar was accurately set up relative to the direction of the electromagnet field, using screws and a turning device. The resistance measurements on the films were made using a potentiometer system; to determine the critical currents the ballast resistance in the film circuit was smoothly changed, and with the aid of a high resistance millivoltmeter the instant at which resistance appears was determined.

The temperature range in which the measurements were made was determined at one extreme, by the fact that when measuring the currents needed to destroy superconductivity, I_c , it was impossible to approach closer to the critical temperature than two or three hundredths of a degree, owing to the inadequate sensitivity of the measuring circuit; at the other extreme it was determined by the fact that for large departures from the critical temperature ($\Delta T \approx 0.4$) the Joule heat

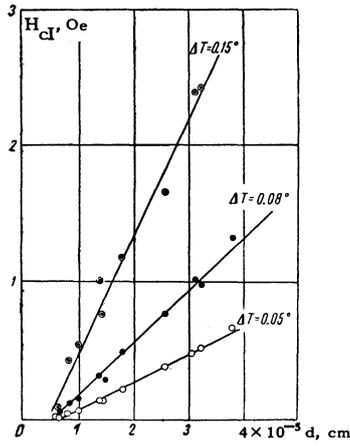


FIG. 4

evolved during the transition increases substantially, causing irreversible changes in the film. The critical magnetic fields (for which these limitations did not apply) were determined in the same temperature interval.

The axes of the films were set parallel to the field by turning the apparatus relative to the electromagnet and finding the minimum resistance of the films for a sub-critical temperature (the setting accuracy was a fraction of a degree).

The results of simultaneous measurements on the critical currents and fields are given for one of the series of films on Figs. 3a and b. Figures 4 and 5 show the variation of critical field with effective film thickness for various departures from the critical temperature.

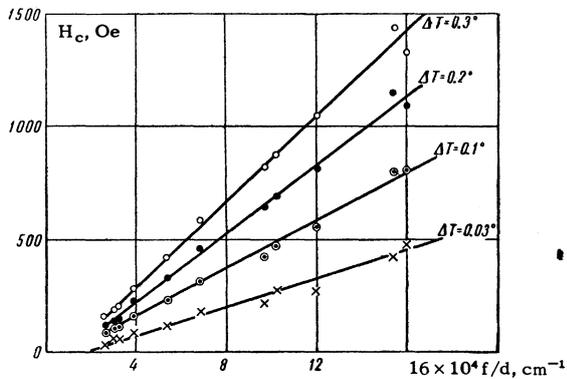


FIG. 5

DISCUSSION OF RESULTS

To treat the experimental results obtained, Ginzburg's work³ on the critical currents in thin films was used; formulae are given there which are applicable when the film thickness is smaller than the penetration depth of the magnetic field, but greater than the parameter, ξ , in the theories of Pippard⁴ and Bardeen, Cooper, and

Schrieffer.⁵ The value of the critical field in this case is given as

$$H_c = \frac{\sqrt{6} \delta_{00} \sqrt{T_c}}{d} \left| \frac{dH_{cm}}{dT} \right| (\Delta T)^{1/2}, \tag{1}$$

and the size of the critical field of a current H_{cI} as

$$H_{cI} = \frac{4\sqrt{2} d}{3\sqrt{3} \delta_{00} \sqrt{T_c}} \left| \frac{dH_{cm}}{dT} \right| (\Delta T)^{1/2}, \tag{2}$$

where d is the film thickness and δ_{00} is the penetration depth at $0^\circ K$ (for tin $|dH_{cm}/dT| = 151$ oe/deg.).

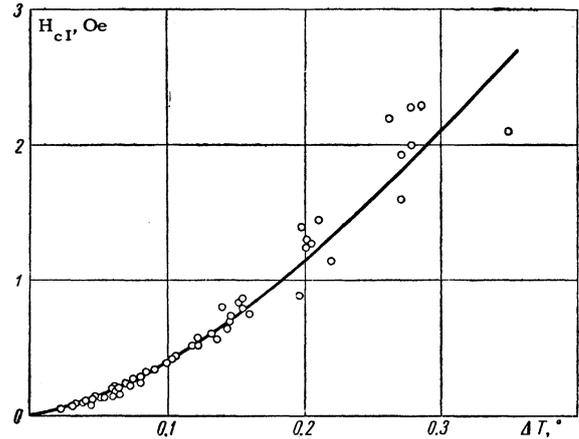


FIG. 6

In Figs. 6 and 7 the variations of H_{cI} and H_c are given for two series of films; it is seen that the experimental points lie satisfactorily on the appropriate curves $A\Delta T^{3/2}$ and $B\Delta T^{1/2}$, fitted to our data at $\Delta T = 0.1^\circ K$ (the scale on the ordinate axis corresponds to a film of 1.5×10^{-5} cm; for other films the scale is appropriately altered).

The ratio of $H_c H_{cI} / \delta_{00}^2 H_{cm}^2$, which, according

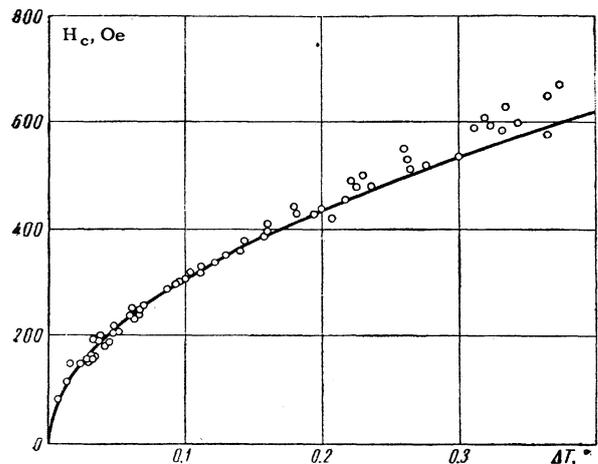


FIG. 7

to reference 3, ought to be constant and numerically equal to unity, was much smaller in our experiments and was 0.22 ± 0.03 ; the penetration depths* δ_{00}^H and δ_{00}^I were, respectively, $1.9 \pm 0.3 \times 10^{-5}$ cm and $9.3 \pm 1.5 \times 10^{-5}$ cm, which are much greater than the value δ_{00} for bulk tin of 5×10^{-6} cm. The small value of the ratio $H_C H_{CI} / \frac{8}{3} H_{Cm}^2$ may be connected with the fact that the critical currents measured in the experiment are greatly reduced by the inevitable presence in the specimens of uncontrolled "weak" spots, which also determine the current which destroys superconductivity.

The data presented in Figs. 4 and 5 show that the inverse and direct proportionalities of the critical field on the film thickness expected from Eqs. (1) and (2) are on the whole obeyed.

The substantial difference (about a factor of three) in the penetration depth δ_{00}^H we have found from the value obtained by Zavaritskii⁶ for thin films can perhaps be explained by the fact that Zavaritskii determined penetration depths which varied systematically with thickness only for the very thickest films he studied. Also an important factor here is the inaccuracy in our determinations of the true thickness of the films. The penetration depths determined from the current data are much bigger because anomalously small currents destroy superconductivity, and these results cannot serve as material for comparison with the theory. In comparatively finely dispersed films Pippard's parameter ξ (see reference 4) is much smaller than in sufficiently good massive specimens. Therefore, it is understandable that our value of δ_{00}^H is substantially larger than δ_{00} for the bulk metal.

In principle, there are two possible variants of experiments with thin layers obtained under clean conditions by condensation in vacuum (we ignore all other methods of producing thin layers). The first of these variants consists in the successive manufacture of separate films with a known quantity of substance deposited on unit area under conditions in which one is easily able to vary within wide limits the temperature of the dielectric substrate on which the condensation is carried out. The drawback of this method is the difficulty of accurately repeating the conditions when making the series of specimens required, and also the limitation in the choice of geometrical shape.

The second possibility (used in our work) consists of the simultaneous fabrication of a

series of specimens with different masses (which had been chosen beforehand) deposited on separate substrates. However, this method can be realized in practice only if the temperature of the substrate is close to room temperature.

Of course, the structure of films obtained by condensation depends very much on the temperature of the condensation surface and even the speed of condensation.

The lower the temperature of the substrate relative to room temperature, the denser and more highly dispersed is the deposit. Close to helium temperatures all deposited metals without exception are very homogeneous and are apparently completely amorphous deposits (not possessing a defined crystalline structure) with densities only slightly reduced relative to that of the bulk metal.*

The speed of condensation in these conditions is not important and can affect the structure of the deposited films only if the heat produced by radiation from the evaporator and by the heat of condensation causes the substrate temperature to rise.

On increasing the substrate temperature the conditions under which the deposit is formed are different for different metals. Here one can state the following qualitative considerations.

Refractory and high melting-point metals form deposits, the structure of which is not greatly affected by the substrate temperature.† The deposits are homogeneous and highly dispersed — electrical conductivity appears in them at very small effective thicknesses (of the order of atomic). Relatively fusible and low melting-point metals‡ (which include tin) form, under these condensation conditions, films whose structure depends markedly on the amount of substance deposited on unit area of substrate. In the very first stage of condensation on a clean substrate separate centers grow isolated from one another; this causes metallic conductivity to appear only at effective thicknesses of many hundreds of atomic layers. The density in this "sub-layer" is significantly smaller than the bulk density of the condensed metal. The formation of a comparatively dense layer during further condensation proceeds on the "sub-layer" already created,

*We leave aside the possibility of transformation polymorphs in the film.

†If this temperature is sufficiently far from the melting point of the metal.

‡The specific behavior of alkaline metal films is not considered.

*In these calculations the data for the very thin films were not used.

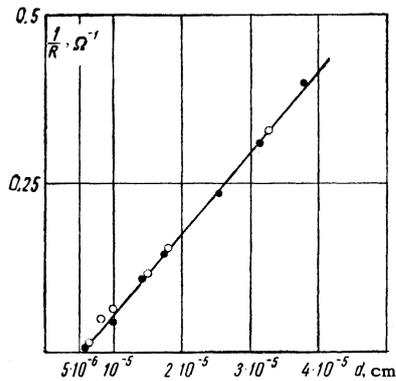


FIG. 8

a fact which is well illustrated by Fig. 8, which shows the variation of the electrical conductivity of tin films on their effective thickness.*

As is seen from this graph, the normal variation of conductivity with thickness starts only after the formation of the "sub-layer" has finished.

Thus, it must be borne in mind that the series of films we made are quite far from ideal, since in the thickest the "sub-layer" can amount to 10% of their thickness. Some compensation for the depth inhomogeneity of films obtained at room temperature is provided by their comparatively highly dispersed state — the crystallite dimen-

sions apparently approach their effective film thicknesses in magnitude. In this sense they are quite different from the highly dispersed "amorphous" films obtained by condensation at low temperatures.

However, all the drawbacks of the method we chose for making the films are balanced by the possibility of making a series of specimens simultaneously and of obtaining these specimens in an ideal shape for the problem posed, i.e., the study of the destruction of superconductivity by current.

We are grateful to V. L. Ginzburg for his interest in the work and valuable discussion and to D. I. Vasil'ev for help with the experiment.

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² L. A. Feigin and A. I. Shal'nikov, *Dokl. Akad. Nauk SSR* **108**, 823 (1956).

³ V. L. Ginzburg, *Dokl. Akad. Nauk SSSR* **118**, 464 (1958), *Soviet Phys.—Doklady* **3**, 102 (1958).

⁴ A. B. Pippard, *Proc. Roy. Soc.* **216**, 547 (1953).

⁵ Bardeen, Cooper, and Schrieffer, *Phys. Rev.* **108**, 1175 (1957).

⁶ N. V. Zavaritskii, *Dokl. Akad. Nauk SSSR* **78**, 665 (1951).

*On cooling to 4.2° the resistance of our films fell by a factor of ten for the thinnest films and by a factor of 30 for the thickest ones.