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MAGNETIC PROPERTIES OF THIN SUPERCONDUCTING TIN AND INDIUM FILMS

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The conditions under which there is no normal phase in a finite size superconducting film placed at a small angle to a uniform magnetic field are determined. The torques acting on such a film are measured. The dependence of the transverse component of the magnetic moment M on temperature and thickness is determined for tin and indium films 4×10^{-3} to 2×10^{-6} cm thick. The critical field values derived from these measurements are analyzed.

INTRODUCTION

ALTHOUGH the magnetic properties of thin superconducting layers are of considerable interest in the theory of superconductivity (see, for example, references 1—3), they have not been investigated experimentally to any extent. There is only one paper[4] reporting the measurements of the magnetic moments of a stack of superconducting films, the thinnest of which were 3.7×10^{-5} cm thick. We know of no study of the magnetic moments of thinner films.

One of the main difficulties in setting up such experiments lies in the following. The magnetic moment investigated in the theory is that of an infinite layer in a parallel field. In the experiment, however, the measurements must be carried out on films of finite dimensions, which are only approximately parallel. A case close to that considered theoretically can be realized by measuring the longitudinal component $M_{\parallel}$ of the magnetic moment in a field parallel to the surface of the film. It is practically impossible, however, to align the film perfectly parallel to the field, and therefore the magnetic moment $M_{\perp}$ normal to the surface of the layer will be different from zero and consequently, there will always exist the probability that the presence of $M_{\perp}$ affects the measurement results. Nor is it possible to calculate $M_{\perp}$, since its dependence on the temperature and on the thickness of the layer has been determined neither theoretically nor experimentally. A determination of this dependence is of interest in itself, since it will permit a considerable simplification in the experimental determination of the magnetic moments of superconducting films, for the difficult measurement of $M_{\parallel}$ is replaced by a measurement of $M_{\perp}$.

To determine $M_{\perp}$ we measured the torque acting on a layer placed at a small angle, $\varphi \approx 10^{-3} - 10^{-4}$ rad with the direction of the uniform magnetic field. The torque K is given by

$$K = \tfrac{1}{2}(\chi_{\parallel} - \chi_{\perp})\, vH^2 \sin 2\varphi, \tag{1}$$

where $\chi_{\parallel}$ and $\chi_{\perp}$ are the magnetic susceptibilities of the film in a parallel and a perpendicular field, respectively; the susceptibility is defined as $\chi = M/H$, H is the external field, and v is the volume of the layer. For a "bulky" ($d \gg \delta$) superconducting layer of thickness d (δ is the depth of penetration of the magnetic field in the superconductor) the inequality $\chi_{\perp} \gg \chi_{\parallel}$ holds true. In this case the torque is determined only by the transverse component of the magnetic moment $M_{\perp} = \chi_{\perp} vH \sin \varphi$. Consequently, apart from the sign, we have

$$M_{\perp} = 2K / H \cos \varphi \tag{2}$$

We can expect the dependence of $M_{\perp}$ on the temperature and on the thickness to be substantially different for bulky layers and thin ($d \leq \delta$) films. If a bulky layer is in the form of a round disc of diameter D, the vanishing of the field in the layer causes the expression for $M_{\perp}$ to be independent of d, namely[5]

$$M_{\perp} = (1/12\pi)\, HD^3 \sin \varphi. \tag{3}$$

On the other hand, the field in a sufficiently thin film should be different from zero (see, for example, reference 6). Its intensity is determined by the ratio δ/d, and $M_{\perp}$ can be expected to depend on the thickness and on the film temperature. Although the truth of the inequality $\chi_{\perp} \gg \chi_{\parallel}$ is not as evident for thin films as it is for bulky superconductors, this inequality apparently remains valid in a certain thickness range. We therefore determined the value of $M_{\perp}$ for films, as for bulky layers, from formula (2).

In the investigation of the magnetic properties of superconducting films, there is always the danger that the film contains domains of the normal phase. If it is assumed that the film can be approximated

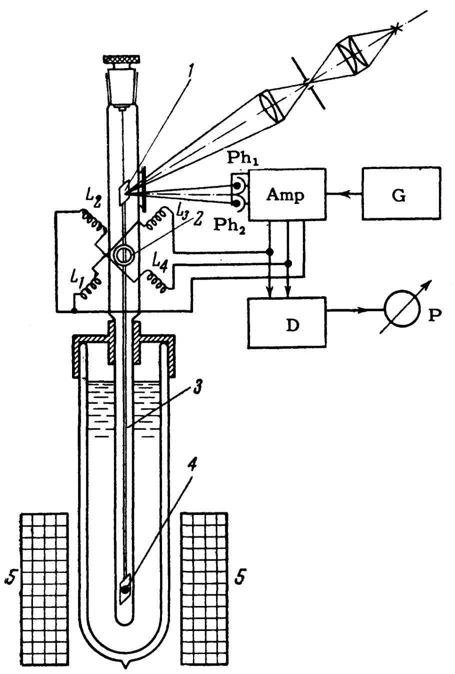

FIG. 1. Diagram of the apparatus: 1 – mirror, 2 – aluminum ring, 3 – glass rod, 4 – sample, 5 – magnet, Amp – amplifier, G – generator, D – detector, P – type KL-48 potentiometer with type M25/3 galvanometer.

by a flat ellipsoid of revolution, one of the axes of which is small, we can, in analogy with a bulky superconductor, write the condition for the absence of a normal phase in the following form:[6]

$$H_{\perp}/(1-N) \leqslant H_{cr}, \quad (4)$$

where H_{cr} is the critical field and $H_{\perp}$ is the magnetic field component perpendicular to the surface of the film, equal to H sin φ. Taking into account the value of the demagnetizing factor N for a layer in the form of a round disc,[5] we obtain the following condition for the absence of normal phase in the layer

$$\varphi_0 \leqslant \tfrac{1}{2}\pi (d/D)(H_{cr}/H). \quad (5)$$

If the angle between the film and the field is less than φ_0, it can be assumed that the film is wholly in the superconducting state.

EXPERIMENTAL PROCEDURE AND SAMPLES

The torque K was measured with a magnetic torsion balance[1] with feedback, described in detail elsewhere.[8] A block diagram of the apparatus is shown in Fig. 1. A glass rod 3 (diameter ~ 1 mm) carries the sample 4 and a light aluminum ring 2. The latter is located in an alternating magnetic field (10 kc/sec) produced by two pairs of coils $L_1 - L_4$, fed in phase with alternating current. The coils are so arranged that the field vector of one pair is perpendicular to that of the other. The plane of the ring is parallel to the direction of the resultant field. When the amplitude of the field of one of the coil pairs is changed, the vector of the resultant field rotates and causes the ring to turn. The amplitude of the field in each pair of coils is determined by the illumination of photocells Ph_1 and Ph_2, and depends on the position of mirror 1. If no torque acts on the suspension system, the illumination from the diaphragm of the light-source is equally distributed between the photocells; the amplitudes of the magnetic fields are equal and no torque acts on the ring. Once a force couple is applied to the sample, the photocell illumination changes, and the resultant torque on the ring is used to cancel out the torque of the sample. If the system parameters are suitably chosen, the angle between the field and the film remains practically constant during the measurements. The accuracy of the position of the film in the field is 5×10^{-5} rad. What is measured is the feedback current, which depends in such a system linearly on the torsion force acting on the suspension. Such a magnetic balance can measure torques ranging from 5×10^{-5} to 4 dyne-cm.*

The homogeneous magnetic field was produced by air-core coils. The earth's magnetic field was canceled out accurate to approximately 1%. The temperature was determined from the vapor tension of helium.

The samples were films of tin and indium (the purity of the original metal was 99.995 and 99.992%, respectively) obtained by evaporation in vacuo ($\sim 10^{-7}$ mm Hg). A special evaporator was used. The films were condensed on a substrate cooled with liquid nitrogen. The substrates were flat optically polished quartz plates measuring $15 \times 10 \times 1.5$ mm. The substrate was covered with a shield to produce a film in the form of a circular disc. The disc diameters were chosen, in accordance with the film thickness, to satisfy condition (5) and ranged from 0.5 to 0.05 cm.

The magnetic moments of films $\sim 10^{-5}$ cm thick were measured with discs of diameter D = 0.2 – 0.3 cm. Smaller values of D were used for thinner films. Since the torque K decreases with decreasing disc diameter, several discs were condensed on the substrate whenever d was less than 0.2 cm, to obtain measurable torques. The thinnest films measured ($d \approx 3 \times 10^{-6}$ cm, D = 0.05 cm) were condensed through a screen with round holes spaced more than 2D apart.

*This procedure is a further development of the method used by Alekseevskiĭ[7] to measure the critical fields of superconducting films.

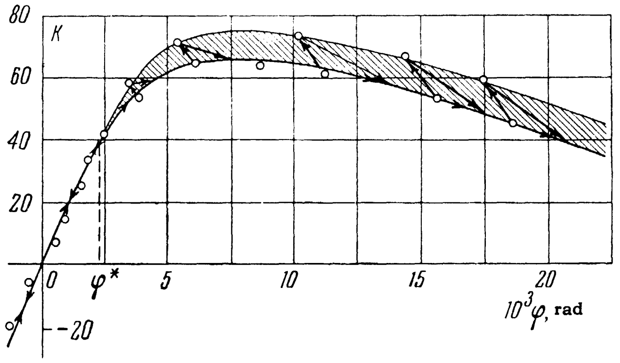

FIG. 2. Dependence of the torque K on the angle φ for a tin film: $d = 1 \times 10^{-5}$ cm, D = 0.06 cm, T = 3.65° K, H_0 = 30 oe. The values of K are given in units proportional to the galvanometer deflection.

The films were evaporated at a rate $\sim 10^{-5}$ cm^3/min. The layers had a mirror surface down to thicknesses of $\sim 1\mu$, and disclosed no surface defects when viewed under a microscope with magnification × 1500. It should be noted that in individual cases the evaporation rate was $10^{-2} - 10^{-3}$ cm^3/min, in order to study the effect of film structure on the magnetic properties. The surfaces of such films were granular. The condensed film was heated to room temperature and transferred to the measuring instrument. Its stay in air did not exceed 10 or 15 minutes. The bulky layers used were foils 5 — 40 μ thick, produced by rolling.

MEASUREMENT RESULTS

Before starting the measurements of the magnetic moment, the films were oriented in the field. For this purpose the nature of the dependence $K(\varphi)$ was determined (Fig. 2). Its linearity in the range of small angles made it possible to establish a film position parallel to the field with great accuracy, and to maintain an angle $\sim 10^{-3} - 10^{-4}$ rad during the measurements. It is seen from the figure that no hysteresis is observed in the linear portion of the $K(\varphi)$ curve. This proves that there are no domains of the normal phase in the film when $\varphi < \varphi^*$ (Fig. 2). The value of φ^* for all the investigated films was of the same order of magnitude as the values of φ_0 given by formula (5).

The value of $M_\perp$ for bulky superconducting layers* increases linearly with increasing field, up to values close to H_{cr} (see Fig. 3*). The value of $M_\perp$ is determined by the cube of the disc diameter, in accordance with formula (3).† The loss of superconductivity of bulky layers occurs abruptly, so that the curve representing this loss occupies a field interval amounting to a fraction of an oersted. As the field is decreased from $H > H_{cr}$, the magnetic-moment curve is duplicated, that is, the field is crowded out from the layer (Meissner effect).‡

It was noted earlier that in sufficiently thin films one can expect $M_\perp$ to depend on the film temperature and on the thickness. However, as can be seen from Figs. 4 — 6, no dependence of $M_\perp$ on these parameters is observed, although the thickness of the films in Figs. 4 and 5 is comparable with δ, and in the case corresponding to Fig. 6 it is less than δ. The value of $M_\perp$ for thin films in weak fields is found to be the same as that for bulky layers (Fig. 3), and is determined by formula (3).

The dependence of $M_\perp$ on the temperature has been observed only in films only several times 10^{-6} cm thick. Here, apparently, the size of the disc diameter D becomes important. Thus, at D = 0.05 cm, a temperature dependence is observed in the initial slope of the magnetization curves only for films with $d \leq 3 \times 10^{-6}$ cm. For smaller values of D, a temperature dependence of $M_\perp$ is observed also in thicker films. The initial slope of the magnetic moment curves increases with decreasing temperature, in the case of films with $d \approx 3 \times 10^{-6}$ and D = 0.05 cm, in the temperature interval $\Delta T = T_{cr} - T \approx 1°K$, after which $M_\perp$ reaches the same value as for a bulky layer. Figure 7 shows the experimental points obtained in some experiments for a tin film.

The appearance of domains of normal phase in films can be readily noted by controlling the occurrence of hysteresis in the magnetic field. A typical hysteresis picture is shown in Fig. 8. The hysteresis always takes place when the $M_\perp(H)$ curve deviates from linear. As can be seen from Figs. 4 — 7, such a deviation takes place in fields

*The measurements of the magnetization curves at a given temperature were made only after the sample was heated (by light from a special illuminator) to a temperature above critical and was made superconducting in the absence of the field.

*Since the magnetization curves of tin and indium films of equal thickness are similar, $M_\perp(H)$ is plotted for indium films in Fig. 5 only. In all other cases the curves pertaining to indium films have been omitted to save space.

†To facilitate comparison of the magnetic moments of films with different values of D and measured at different angles φ, the quantity $M_\perp^\ddagger = M_\perp/D^3 \sin\varphi$ is shown in Fig. 3, and in Figs. 4–10 later on, in a certain arbitrary scale, which is, however, the same for all plots.

‡The supercooling that takes place on the layers is not considered in the present work.

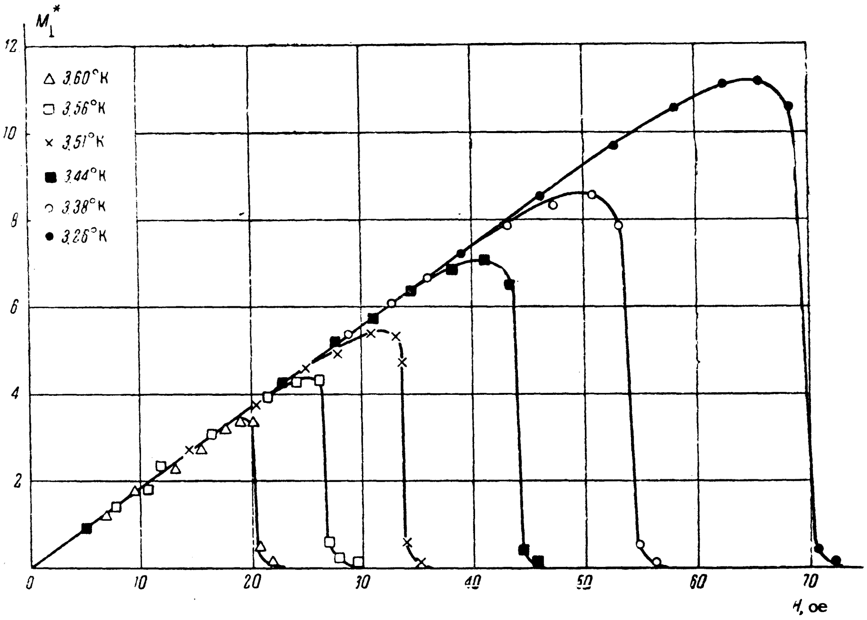

FIG. 3. Magnetization curves of tin foil: $d = 4 \times 10^{-3}$ cm, $D = 0.5$ cm, $\varphi = 1 \times 10^{-3}$ rad, $T_{cr} = 3.73°$ K. Here and elsewhere $M_\perp^* = M_\perp/D^3 \sin\varphi$.

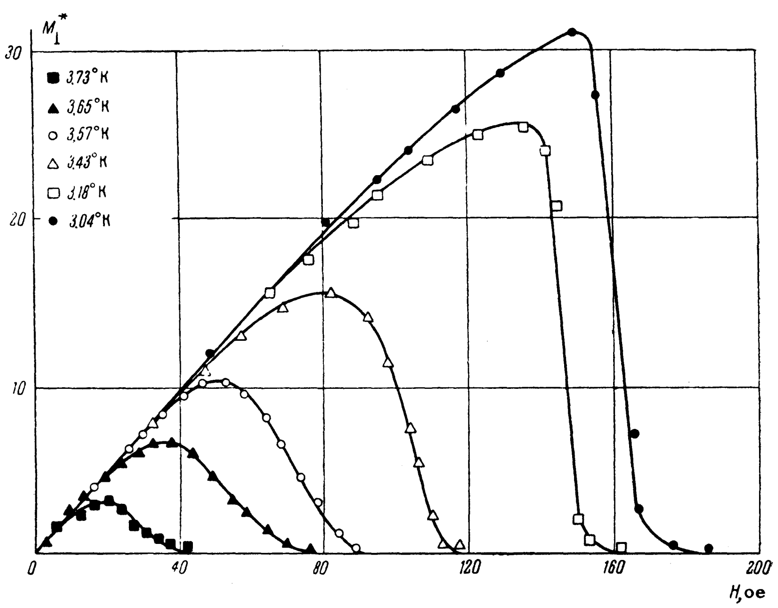

FIG. 4. Magnetization curves of tin film: $d = 3 \times 10^{-5}$ cm, $D = 0.1$ cm, $T_{cr} = 3.81°$ K, $\varphi = 1 \times 10^{-3}$ rad.

considerably below H_{cr}, even when the ratio D/d does not exceed 10^3 and the film makes an angle less than 10^{-3} rad with the field. It is found here that the external field in which the normal phase is produced in the film is of the same order of magnitude as given by (5).

If the geometrical dimensions of the film and the angle φ are such that double connectivity takes place even in weak fields, the magnetic-moment curves are as shown in Fig. 9. Their initial slope has in this case a temperature dependence. However, unlike the films of Figs. 4 — 7, hysteresis takes place even on the linear portions of the curves. This shows that the temperature dependence of the initial slope of the magnetic-moment curve is due in this case only to the temperature variation of the geometric parameters of the superconducting loop produced in the film, and is not connected with the depth of penetration δ. Thus, the presence of domains of normal phase in the film is always accompanied by hysteresis. The absence of hysteresis on the initial portion of the curves of the magnetic moment $M_\perp$ was therefore watched with particular care in all the measurements.

The magnetic properties of films depend appreciably on the structure of their surface. Thus, the absence of temperature dependence in the initial slope of the $M_\perp(H)$ curves for layers with $d > 3 \times 10^{-6}$ cm occurs only for films with mirror surface, in which no defects are seen under a microscope with magnification ~ 1500. On the other

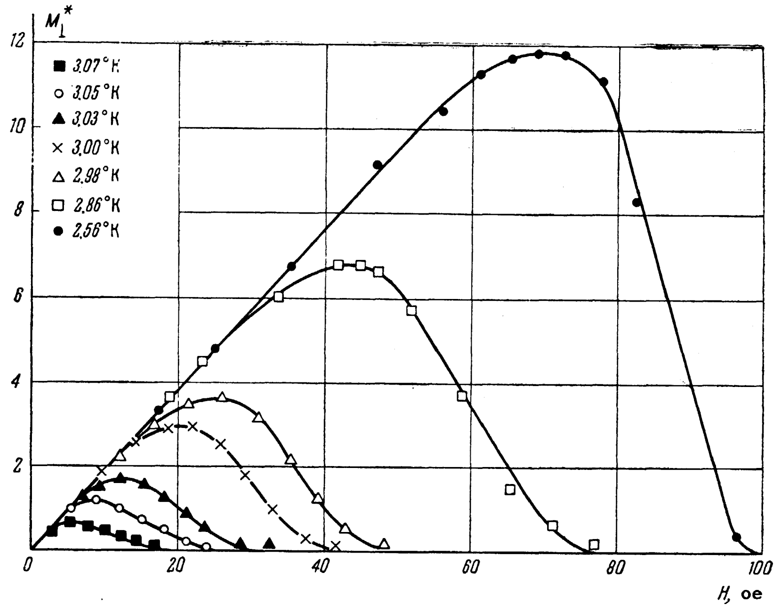

FIG. 5. Magnetization curves of indium films: $d = 3 \times 10^{-5}$ cm, $D = 0.1$ cm, $T_{cr} = 3.10°$ K, $\varphi = 1 \times 10^{-3}$ rad.

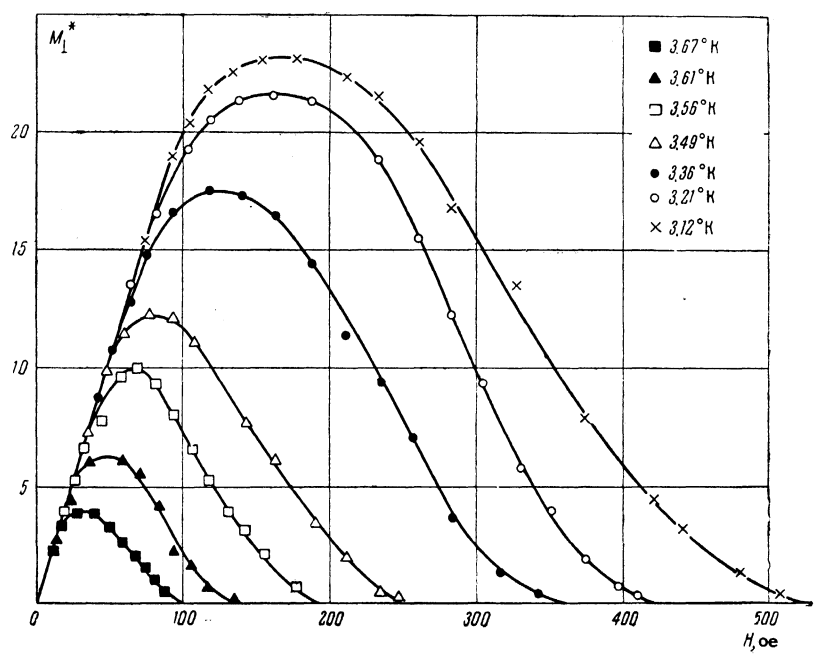

FIG. 6. Magnetization curves of tin films: $d = 1.2 \times 10^{-5}$ cm, $D = 0.06$ cm, $T_{cr} = 3.77°$ K, $\varphi = 4 \times 10^{-4}$ rad.

hand, films with granular surfaces display a temperature dependence of the initial slope of the magnetization curves (Fig. 10) even when $d > 10^{-5}$ cm. In this case there is no hysteresis in the linear portions of the curves. The increase in the moment $M_\perp$ with decreasing temperature takes place in an interval $\Delta T \approx 0.1°$K. The value of $M_\perp$ at the lowest temperature is somewhat lower than would follow from (3).

The form of the magnetic-moment curves (Figs. 3 — 7) depends on the ratio d/d_{cr}, where d_{cr} is the "critical thickness," defined in the Ginzburg-Landau theory as $d_{cr} = \sqrt{5}\,\delta$ (Fig. 11). In the case when $d > d_{cr}$ and the superconducting transition is a first-order phase transition, the loss of superconductivity occurs abruptly. The magnetic-moment curve has in this case a sharp discontinuity at fields close to H_{cr}. On the other hand, when $d < d_{cr}$, there are no sharp breaks in the curve to evidence any supercooling. One can therefore conclude that in films with $d < d_{cr}$ the superconducting transition is apparently a phase transition of second order. It is interesting to note in this connection that the temperature dependence of the slope of the superconductivity-loss curves is observed only in films with thickness $d \approx d_{cr}$ (Figs. 4, 5).

The curves obtained for the magnetic moment enable us to determine the critical fields H_{cr} of the films.[7] The value of H_{cr} was determined by extrapolating the most steeply drooping portion of the superconductivity-loss curve to the field axis. This excludes the possibility of obtaining exaggerated

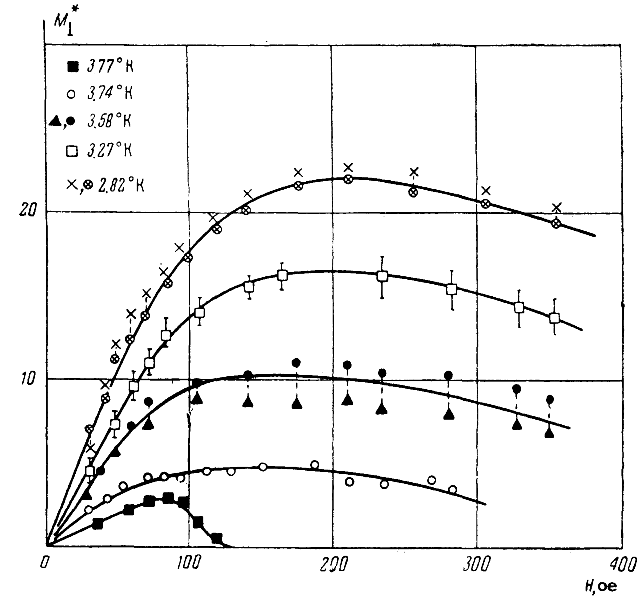

FIG. 7. Magnetization curves of a tin film with temperature dependent susceptibility; d = 2 × 10^{-6} cm, D = 0.05 cm, φ = 2.5 × 10^{-4} rad, T_{cr} = 3.78° K.

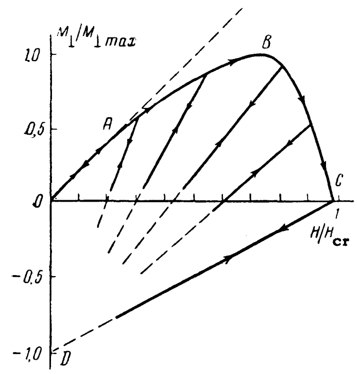

FIG. 8. Typical picture of hysteresis phenomena in thin superconducting films.

values of H_{cr} as a result of stresses in the film. The value of dH_{crb}/dT, determined in this manner for a bulky layer of tin (d = 4 × 10^{-3} cm) amounts to 151 oe/deg, which is in good agreement with the data given in the literature. The temperature dependence of H_{cr} is well described by the Ginzburg-Landau formulas:[9]

$$H_{cr}/H_{crb} = 2\sqrt{6}\ (\delta/d) \text{ for } d \ll \delta, \tag{7}$$

$$H_{cr}/H_{crb} = 1 + \delta/d \text{ for } d \gg \delta, \tag{8}$$

where $\delta = \frac{1}{2}\delta_0\sqrt{T_{cr}/\Delta T}$. As can be seen from Fig. 11, the experimental points fit the theoretical curves quite well. The value of δ_0 for tin films, determined from their critical fields by means of formulas (7) and (8), is found to be (8.5 ± 0.5) × 10^{-6} cm. For indium films, δ_0 = (10 ± 0.5) × 10^{-6} cm.

As is well known,[10] at temperatures near T_{cr} tin is a "London" type metal, and the following relation should therefore be satisfied for tin films[11,12]

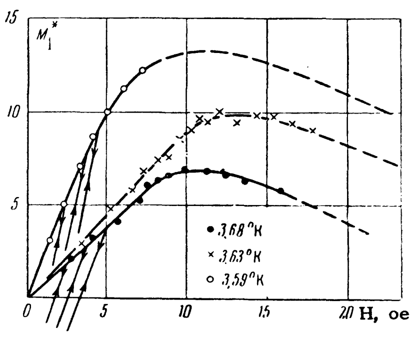

FIG. 9. Magnetization curves of films that do not satisfy condition (5); d = 1.3 × 10^{-5} cm, D = 0.7 cm, φ = 1.5 × 10^{-2} rad.

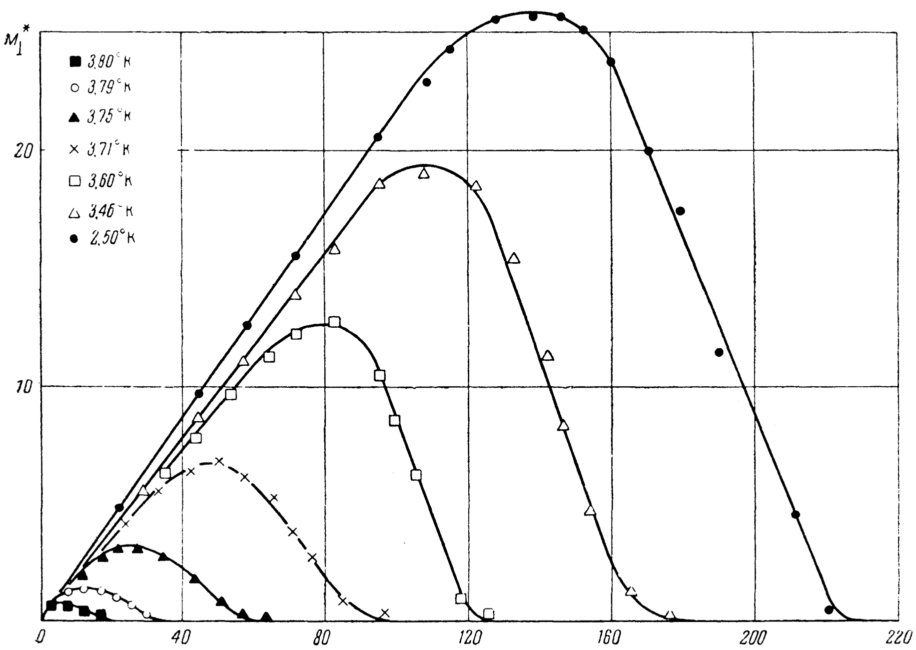

FIG. 10. Magnetization curves of a tin film with granular surface. The initial slope of the curves has a temperature dependence. d = 2.5 × 10^{-5} cm, D = 0.1 cm, φ = 1 × 10^{-3} rad, T_{cr} = 3.86° K.

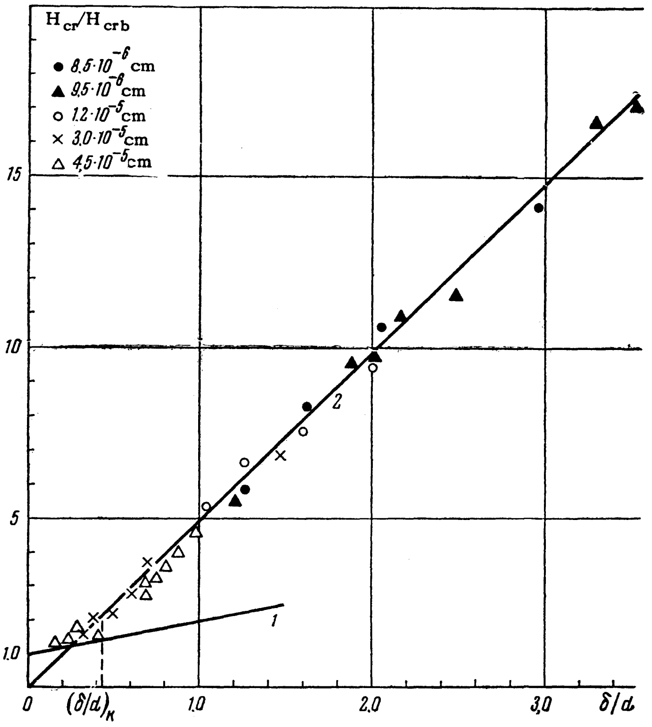

FIG. 11. Dependence of H_{cr}/H_{crb} on the ratio δ/d for tin films: curves 1 and 2 are based on formulas (8) and (7), respectively.

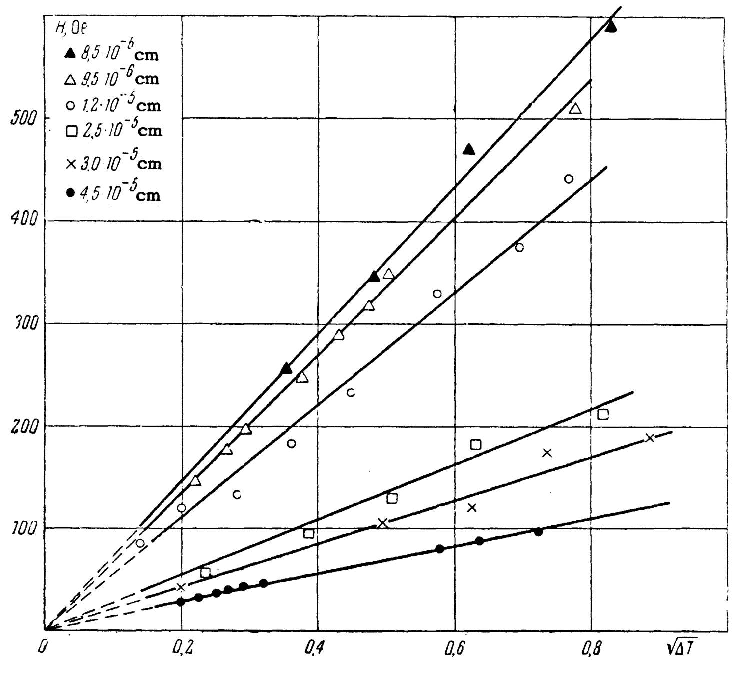

FIG. 12

$$H_{cr} = \sqrt{6}\;(\delta_0/d)\sqrt{T_{cr}}\,|dH_{crb}/dT|\,\sqrt{\Delta T}. \quad (9)$$

The dependence of H_{cr} on 1/d and on $\sqrt{\Delta T}$ was found to be linear (Fig. 12) for tin films. The indium films also display a linear dependence of H_{cr} on these parameters. The value of δ_0 determined for tin and indium films from (9) coincides with the values determined from formulas (7) and (8).

DISCUSSION OF THE RESULTS

Proceeding to analyze the results, we note that in our measurements φ amounted to $10^{-2} - 10^{-4}$ rad. Such an angle can readily arise in measurements of the magnetic moment $M_\perp$ in a parallel field. For example, one can assume that in the experiments by Lock[4] and Schawlow[13] the film was parallel to the field not closer than to within 10^{-3} rad, since neither the orientation of the films in the field nor the quality of the substrate surface were specially controlled. Consequently the data on the domains of normal phase in the films and on the character of the superconductivity-loss curves, obtained in the present investigation, can be said to pertain to films measured practically in a parallel field.

The results obtained show that normal-phase domains occur in films in fields considerably lower than H_{cr}, even when the linear dimensions of the film amount to a fraction of a millimeter, and $\varphi \approx 10^{-4}$ rad. Therefore, in investigations of δ (T) and in particular of δ (H) near H it is essential to ensure the absence of no normal-phase domains in the film. We note that in Lock's experiments the films were probably doubly-connected in fields close to H_{cr}, for the magnetization curves displayed hysteresis as the field was decreased from $H > H_{cr}$. However, the instants when the normal-phase domains were produced in the films was not checked in Lock's experiments.

In spite of the fact that the film thickness becomes comparable with the depth of penetration δ even when $d = (5-6)\times 10^{-5}$ cm, a temperature dependence of $M_\perp$ is observed in weak fields only for the thinnest films, of thickness on the order of 10^{-6} cm (Fig. 7). Unfortunately, it is still impossible to make a detailed evaluation of these results, since there is no theoretical expression for $M_\perp$.*

In the discussion of the results, notice must be taken of the fact that the region of the superconductivity-loss curve (BC in Fig. 8) increased as the thickness d was decreased and T approached T_{cr}, (Figs. 3 — 7), although it might have been expected (see, for example, references 6 and 14) that, in view of the decrease in the absolute value of the magnetic susceptibility of a thin film, the

*G. F. Zharkov was kind enough to advise us that he derived formulas for the dependence of $M_\perp$ on the temperature and on the thickness of the layer.

distortion which the susceptibility produces in the configuration of the magnetic field would become smaller, and the region of superconductivity loss should also decrease.

Notice should be taken of the convenience of determining the value of H_{cr} of films with the aid of a magnetic torsion balance with feedback, since it dispenses with the need of using current leads for the film. By keeping the position of the sample fixed during the experiments the difficulties[6] which have arisen in the work of Alekseevskii[7] are eliminated, and it becomes possible to perform the measurements starting with values $\Delta T = 0.02°$ K.

The depth of penetration δ_0, determined from the critical fields, was found to be greater in films than in bulk superconductors. Although the films did disclose previously[12,15] an increased value of δ_0, nevertheless, in the determination of H_{cr} from the electric resistance, this might have been due to the shunting action of the stressed portions of film, which have higher values of H_{cr}. On the other hand, if H_{cr} is determined by magnetic measurements, the influence of the stressed portions is eliminated. Nevertheless, the value of δ_0 obtained for tin and indium films in the present investigation exceeds considerably the values of δ_0 of bulk single-crystal samples. This result is apparently explained by the fact[10] that the parameter ξ of the Bardeen-Schrieffer-Cooper theory is smaller for films than for bulk single-crystal superconductors.

In conclusion, the author is deeply grateful to Professor N. E. Alekseevskiĭ for many valuable hints and for a discussion of the results, to G. F. Zharkov for discussing many theoretical problems, and to Yu. V. Sharvin for useful remarks when the article was prepared for press. The author is grateful to Professor A. I. Shal'nikov for his interest. Great help in the performance of the experiments was rendered by V. A. Sokolin and E. G. Kharakhash'yan, for which the author expresses his thanks.

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