MAGNETIC MOMENT OF SUPERCONDUCTING TIN FILMS

B. K. SEVAST'YANOV and V. A. SOKOLINA

Institute of Crystallography, Academy of Sciences, U.S.S.R.; Moscow State University

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The temperature dependence of the magnetic moment component perpendicular to the surface of a superconducting layer, $M_\perp$, is investigated in tin films of $(3\times3)\times10^{-6}$ and $(5\times5)\times10^{-6}$ cm area and 10^{-5}—10^{-6} cm thickness. It is shown that the value of the moment $M_\perp$ is determined not only by the thickness of the film, but also by its area. Satisfactory agreement is found between the experimental results and the theoretical formula of Zharkov.[2]

In a work published previously,[1] the results were given of measurements of the magnetic moment component, $M_\perp$, perpendicular to the surface of the superconducting layer (the transverse component), in films in the form of circular discs of diameter from 0.6 to 0.06 cm. Here, even in the thinnest of the films studied, the thickness of which was less than the penetration depth δ (usually taken to be of the order of 10^{-5} cm for tin), the magnetic moment $M_\perp$ in weak fields ($H < H_{cr}$) was shown to be the same as for thick layers, and not to depend on either the temperature or the film thickness. Only in two cases was it possible to observe a temperature dependence of the initial slope of the curves $M_\perp(H)$, viz., in a film of thickness 2×10^{-6} cm with dimensions 0.06×0.06 cm and on granular films.

This result, which was previously unexpected, can be understood in terms of the work of Zharkov.[2] It was shown by Zharkov that if the film is approximated by an oblate ellipsoid of revolution, then its moment $M_\perp$ depends on a certain parameter

$$x = \pi dD/16\delta^2. \qquad (1)$$

Here D is the diameter of the film disc; d is the thickness of the disc; δ is the penetration depth of the magnetic field in the superconducting film, which, near the critical temperature of the superconducting transition T_{cr}, is expressed by the well-known formula (see, for example, [3])

$$\delta = \tfrac{1}{2}\delta_0\sqrt{T_{cr}/\Delta T} \qquad (2)$$

(we take δ_0 to be equal to 8.5×10^{-6} cm;[1] $\Delta T = T_{cr} - T$). At sufficiently small values of the parameter x, one should expect a dependence of $M_\perp$ on the temperature and on the thickness of the film, while for large values of x there is no such dependence.

We report here results of an investigation of the temperature dependence of the transverse component of the magnetic moment $M_\perp$ of superconducting films of different thicknesses.

EXPERIMENTAL METHODS AND SPECIMENS

For the determination of $M_\perp$, we measured the torque K acting on a film placed at a small angle φ (10^{-3}—10^{-4} rad) to the magnetic field. The value of $M_\perp$ is determined by the formula*

$$M_\perp = K/H\cos\varphi. \qquad (3)$$

The measurements were carried out on the magnetic torque balance described previously, with inductive electrodynamic self-balancing[4] and with a sensitivity of $\sim10^{-5}$ dyne-cm.

In order that the films satisfy the condition x = 3 for $\Delta T = T_{cr} - T \approx 1°$K, the value of the product Dd should be of the order of 10^{-9} cm^{-2}. The thickness of the layer in these measurements could not be less than 1×10^{-6} cm, since granularity may occur at lower thicknesses and distort the temperature dependence of the initial slope of the curves $M_\perp(H)$ (see, for example, Fig. 10 in [1]). Therefore, it was required that the linear dimension of the film L be of the order of 10^{-3} cm.

Inasmuch as the moment $M_\perp$ is proportional to D^3, the value of $M_\perp$ falls off sharply with decrease of the diameter of the film disc. In order to guarantee a value of the moment sufficient for the measurements, it was required to condense about 10^5 discs of diameter 10^{-3} cm on a single substrate. It was not possible for us to obtain a film in the form of circles of this diameter, but

*In [1] there is an error in the writing of this formula. However, it does not affect the treatment of the results used in the research, since the quantity $M_\perp$ is expressed everywhere in relative units.

we were able to prepare squares with side L equal to 5×10^{-3} and 3×10^{-3} cm.* In order to compare the results obtained on these films with the formula (4) of Zharkov,[2] a certain effective diameter was assigned to the film, D_{eff}. Comparison of the magnetic moments $M_{\perp}$ of bulky superconducting layers in the form of squares with the $M_{\perp}$ of circular layers has shown that $D_{eff} = 2\sqrt{S/\pi}$, where S is the area of the layer. Thus the effective diameters for film squares with sides 3×10^{-3} cm and 5×10^{-3} are 3.4×10^{-3} and 5.6×10^{-3} cm, respectively.

The films were obtained by condensation of tin vapor in a vacuum no worse than 2×10^{-6} mm Hg on quartz plates of optical flatness, cooled by liquid nitrogen. The substrate was covered by a square grid of the given dimensions. A standard electrolytic screen used in electron microscopy for specimen preparation was used for this purpose. The linear dimension of the cells in these screens amounted to 5×10^{-3} cm. We succeeded in decreasing the dimension of the cell to 3×10^{-3} cm by rolling the screen on special rollers which had optically smooth surfaces. Microphotographs of parts of the films studied are shown in Fig. 1.

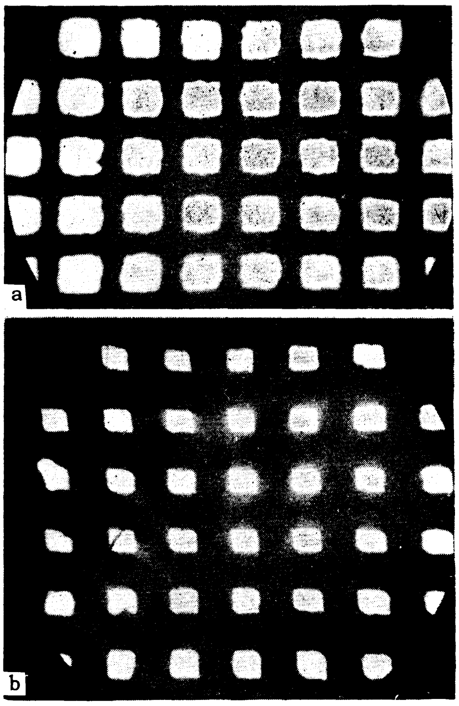

FIG. 1. Microphotographs of parts of the films investigated: a—film with side of the square $L = 5 \times 10^{-3}$; b—film with $L = 3 \times 10^{-3}$ cm.

†We shall call these "films of small dimension," keeping it in mind that the layers usually employed for measurements of the magnetic moment of a layer have a dimension on the order of a centimeter.

It is seen in the figure that the scatter of the dimensions of the cells in each such grid is small; therefore, we can assume that the magnetic properties of the films are equivalent to the magnetic properties of the individual squares.

The thickness of the layers was measured by a Linnik microinterferometer (MII-4). In the case of thin films ($\sim 10^{-6}$ cm), when the error in the measurement of the thickness exceeds twenty percent, the thickness of the layer was determined from the weight of the vaporized tin and the geometric parameters of the evaporator. A correction was used, obtained by measuring the thickness of a sufficiently thick film and comparing it with the computed quantity.

Figure 2 is a photograph of the interference picture in one of the squares of film, 1.4×10^{-5} cm thick. By analysis of the interference bands one can determine not only the thickness of the film but also the profile of its cross section. As is seen from Fig. 2, the film, condensed through a grid of $(3-5) \times 10^{-3}$ cm mesh, is a plane layer with sharply terminated edges. However, it should be noted that such a profile of the cross section of the film squares is obtained only in the case in which the substrate and the grid are annealed for 2—3 hours in vacuum at a temperature of 300—350°C. If the annealing is not carried out, then, inasmuch as the grid does not adhere closely to the surface of the substrate, the squares usually have diffuse contours and may even overlap one another.

The position at which the film is parallel to the field was determined as previously[1] from the dependence of the torque on the angle. The measurement of this dependence is carried out in the following manner: The film is set at some angle to the field and the field is turned off. Next, the field is applied and the value of the torque is recorded. Then the field is turned off, the specimen is heated to $T > T_{cr}$ and the superconducting state

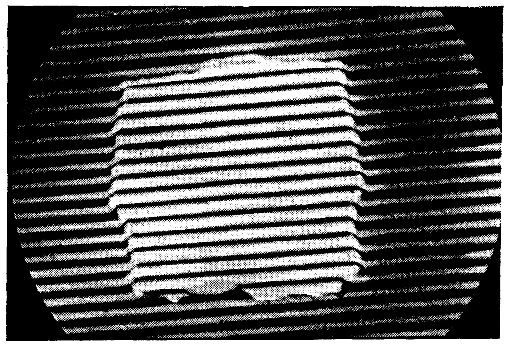

FIG. 2. Interference picture on the surface of the film. Thickness of the film $d = 1.4 \times 10^{-5}$ cm, $L = 5 \times 10^{-3}$ cm.

is restored in the absence of the field. After this, the procedure is repeated for another value of φ. The dependence of the torque on the angle thus obtained is represented in Fig. 3. The linear character of this dependence in the region of small angles makes it possible to obtain the angle $\varphi = 0$ with an accuracy of $\sim 10^{-5}$ rad.

When $\varphi > \varphi^*$ the plot is no longer linear and the values of K are not reproducible in repeated measurements (shaded part). From the curve shown in Fig. 3, it is possible to conclude that when $\varphi < \varphi^*$ there are no domains of normal phase in the film. When $\varphi > \varphi^*$ they do appear, so that the scatter in the torque values is quite natural.

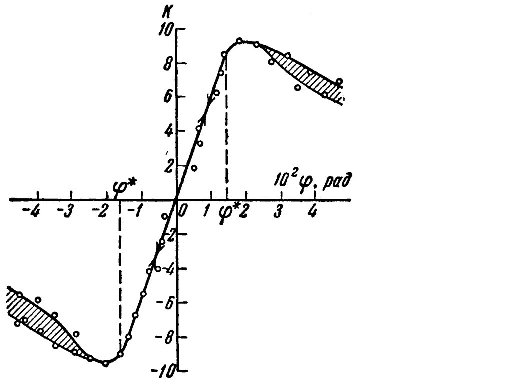

FIG. 3. Dependence of the torque K on the angle φ ($d = 2.7 \times 10^{-6}$ cm, $L = 3 \times 10^{-3}$ cm, $T_{cr} = 3.95$°K, H = 167 Oe, T = 3.42°K).

The angle in the measurements was always chosen much smaller than φ^* and usually amounted to $\sim 10^{-4}$ rad. The magnetic field of the earth was compensated with an accuracy to within ~ 0.01 Oe. The temperature was determined by the helium vapor pressure.

RESULTS OF MEASUREMENTS

The magnetization curves $M_\perp(H)$ for tin films of 1.4×10^{-5} cm thickness and 5×10^{-3} cm dimension, taken at different temperatures, are plotted in Fig. 4. As can be verified from this drawing, the dependence $M_\perp(H)$ differs from the analogous dependence for films with dimensions on the order of 0.1—1 cm.[1] Departures from linearity are not observed up to fields close to the critical. Only in the neighborhood of H_{cr} are the curves rounded off, after which a sharp break appears (destruction curve). One then concludes that the domains of normal phase appear in films of small dimension for fields sufficiently close to the critical, more than for films of dimension on the order of 0.1—1 cm.

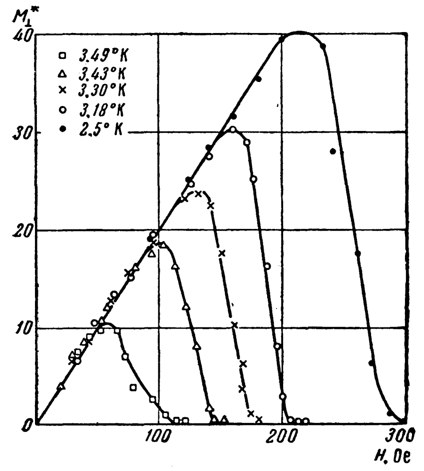

FIG. 4. Magnetization curves of a tin film ($d = 1.4 \times 10^{-5}$ cm, $L = 5 \times 10^{-3}$ cm, $T_{cr} = 3.69$°K, $\varphi \approx 1 \times 10^{-3}$ rad). Here and in Figs. 5, 6 the relative value of $M_\perp^* = M_\perp/D^3 \sin\varphi$ is given.

The hysteresis phenomena in films of small dimension are also different. Upon reduction of the field from $H > H_{cr}$, the curves $M_\perp(H)$ for films with thickness up to 1×10^{-5} cm do not change sign, but reproduce values close to the values of $M_\perp(H)$ in an increasing field (Fig. 5). The small difference ordinarily observed in the value of $M_\perp$ is evidently explained by the fact that some squares of the film become doubly connected, so that $M_\perp$ changes sign in them upon decrease in the field. If the thickness of the films is less than 10^{-5} cm, then even at dimensions from 3×10^{-3} to 5×10^{-3} cm the hysteresis on them is the same as in the case of the layers of ~ 1 cm, investigated earlier.[1]

On the 3×10^{-3} cm films it is possible to observe a temperature dependence of the initial slope of the curves of $M_\perp(H)$ (Fig. 6). Here the hysteresis on the initial linear portion of these curves, which gives information on the double connectedness in the films, is not observed, so

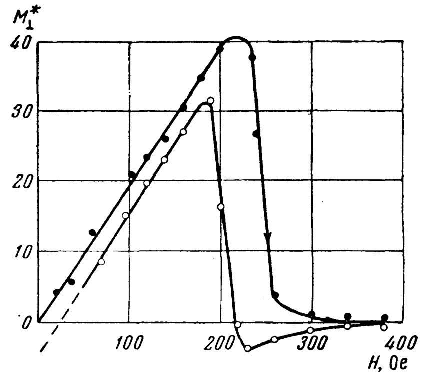

FIG. 5. Hysteresis on films of small size ($d = 1.4 \times 10^{-5}$ cm, $L = 5 \times 10^{-3}$ cm, $T_{cr} = 3.69$°K, T = 3.0°K, $\varphi \approx 5 \times 10^{-4}$ rad): ●— field increasing, ○ — field decreasing.

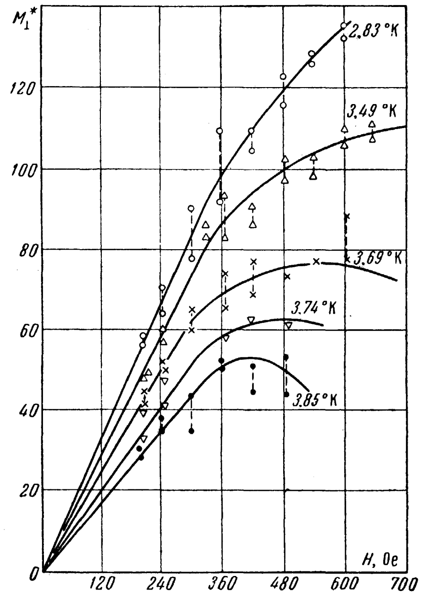

FIG. 6. Initial parts of the magnetization curves of a tin film ($d = 2.7 \times 10^{-6}$ cm, $L = 3 \times 10^{-3}$ rad, $T_{cr} = 3.95°$ K, $\varphi = 6 \times 10^{-4}$ rad) for different temperatures.

that the change of $M_\perp$ with temperature in weak fields is not the consequence of the presence in the film of domains of normal phase. This dependence can be evidently explained by the effect of the penetration depth δ.

The dependence of the value of $M_\perp/M_\perp^0$ (where $M_\perp^0$ is the transverse component of the magnetic moment of the bulky superconducting layer) on the parameter x, determined from the temperature variation of the initial slope of the curves $M_\perp(H)$, which ratio is obtained on 3×10^{-3} cm tin films of thickness 2.7×10^{-6} and 2.8×10^{-6} cm, is given in Fig. 7. The continuous portion of the curves at the origin of the coordinates is a theoretical curve constructed in correspondence with

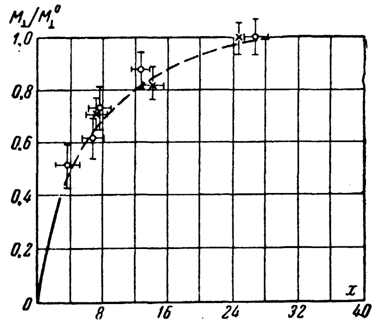

FIG. 7. Dependence of $M_\perp/M_\perp^0$ on the parameter $x = \pi dD/16\delta^2$; o – points corresponding to the magnetization curve shown in Fig. 6; x – points corresponding to the magnetization curves of the film with $d = 2.8 \times 10^{-6}$ cm and $L = 3 \times 10^{-3}$ cm. The continuous part of the curve near the origin is constructed from Eq. (4) of Zharkov.[2] The dashed curve is drawn through the experimental points.

the formula (4) of the work of Zharkov.[2] The dashed curve is drawn through the experimental points. As is evident from Fig. 7, there is qualitative agreement of the experimental and theoretical curves. With increasing parameter x, the transverse component of the magnetic moment $M_\perp$ increases and for $x \approx 30$ reaches the value $M_\perp^0$. If $x > 30$, then the moment $M_\perp$ of the thin film is shown to be equal to the moment of the bulky layer of the same dimension. Here $M_\perp$ is likewise independent of the temperature and of the thickness of the layer, even if the latter is less than the penetration depth δ determined from the critical fields for the film in a parallel field.

This circumstance makes it possible to use as the moment of the bulky layer the moment of the same field at a sufficiently low temperature in the determination of the relative values of $M_\perp/M_\perp^0$.

We note that the penetration depth enters into the expression for the parameter x. We have determined δ from the value of the critical fields of the same films according to the well-known formulas of the Ginzburg-Landau theory.[7] Here the value of δ_0 for tin films was found to be equal to $(8.5 \pm 0.5) \times 10^{-6}$ cm, as before.

DISCUSSION OF THE RESULTS

In the analysis of the results obtained, the question first arises as to whether one can compare the results of measurements on films with the theoretical formula obtained for a flat ellipsoid of revolution. In this connection we note that the formula[6]

$$M_\perp^0 = (12\pi)^{-1} D^3 H, \tag{4}$$

which determines the transverse component of the magnetic moment of the bulky superconducting layer, is also obtained under the assumption that a layer of finite extent can be approximated by a flat ellipsoid of revolution. The large number of measurements of the moment $M_\perp^0$, carried out by us on bulky superconducting layers of different diameters, shows that the measured values of $M_\perp^0$ coincide with the $M_\perp^0$ determined by Eq. (4), within the limits of accuracy of the experiment.

So far as the shape of the films is concerned, comparison of the magnetic moments of films having the shape of squares with the magnetic moments of circular films shows that the character of the magnetization curves does not depend on the geometric shape of the film and is determined only by the area of the latter.

Inasmuch as relative values $M_{\perp}/M_{\perp}^{0}$ are considered in the present work, with $M_{\perp}^{0}$ defined as the $M_{\perp}$ of the same film at the lowest temperature, where the parameter $x > 30$, the deviation of the film shape from circular in the given case is inconsequential. Taking into account the arguments set forth, we have applied Eq. (4) of Zharkov [2] for treatment of the results obtained on thin plane layers of square shape.

The dependence of $M_{\perp}/M_{\perp}^{0}$ on the parameter x, given in Fig. 7, is obtained for films of dimension 3×10^{-3} cm. The change of $M_{\perp}$ with temperature for such films with thickness of the order of 2×10^{-6} cm is observed in the temperature range $\Delta T = T_{cr} - T \approx 1°K$. For thicker films, $M_{\perp}$ increases so rapidly upon lowering of the temperature that all the change of $M_{\perp}$ takes place for a ΔT of the order of tenths of a degree, and the observation of the temperature dependence of the initial slope of the curves of $M_{\perp}(H)$ becomes very difficult.

It should be noted that the moment $M_{\perp}$ on 5×10^{-3} cm films increases sharply as the temperature decreases, so that even for a thickness of the film of 2×10^{-6} cm it achieves a value of $M_{\perp}^{0}$ at $\Delta T = 0.1°K$. Here, in view of the smallness of the measuring field, the error of the measurement amounted to 30 — 40 percent. Therefore, although it was seen that $M_{\perp}$ decreases for decrease in the parameter x, it was not possible to obtain reliable results on films of extent of 5×10^{-3} cm.

The results published earlier, [1] and also the results obtained in the present work, show that the magnetic properties of thin superconducting films placed at a small angle ($\sim 10^{-4}$ rad) to the field, are determined not only by the thickness of the layer, but also by its dimension. Here the moment $M_{\perp}$ is equal to the moment of a bulky layer, $M_{\perp}^{0}$, if $x > 30$, in spite of the fact that the thickness of the film can be less than the penetration depth δ determined from the critical field.

The data obtained by us, and also the calculation carried out by Zharkov, [2] testify to the fact that the moment $M_{\perp}$ can exceed $M_{\parallel}$ by several orders of magnitude. Therefore, for measurements on thin films in an approximately parallel field, it is especially necessary to control the contribution made by the transverse component $M_{\perp}$ to the measured quantity.

The temperature dependence of $M_{\perp}$ in weak fields for calibrated films with dimensions on the order of several centimeters can clearly be explained by the fact that in this case the parameter must be referred, not to the whole film, but to each granule. Here, in view of the smallness of the dimensions of the granules, it can be shown that the condition $x < 30$ is satisfied, and the initial slope of the magnetization curves $M_{\perp}(H)$ shows a temperature dependence. In this case, we note that the previously observed dependence of the initial slope of the curves $M_{\perp}(H)$ for a film of thickness 2×10^{-6} cm and diameter 0.06 cm (see Fig. 7 in [1]) is much smoother than follows from Fig. 7. This is evidently the consequence of the fact that the whole film can be shown to be granular in spite of the measures taken.

It should be noted that Eq. (4) of Zharkov [2] is obtained for the case of local interaction between the field and the superconducting flow. The sufficiently satisfactory agreement of the experimental and theoretical curves again demonstrates that tin is a superconductor of the London type near T_{cr}.

In conclusion, the authors express their deep gratitude to Prof. N. E. Alekseevskiĭ for valuable advice and discussion of the results, and to G. F. Zharkov for useful discussions and observations.

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