THE SUPERCONDUCTING PROPERTIES OF THIN ALUMINUM FILMS

I. S. KHUKHAREVA

Moscow State University

Submitted to JETP editor, May 9, 1962

J. Exptl. Theoret. Phys. (U.S.S.R.) **43**, 1173-1178 (October, 1962)

Studies have been made on aluminum films obtained by condensation in high vacuum at room temperature or on a substrate cooled to liquid helium temperature. The constant factor δ_{00} involved in the temperature variation of the penetration depth close to T_c was determined from the experimental data on the critical magnetic fields of the films. The value of δ_{00} obtained is compared with that calculated from the normal conductivity of the films according to the new theory of superconducting alloys.

In the light of the new microscopic theory, pure superconductors are divided into two groups which differ in their properties in a weak magnetic field. [1,2] The nonlocalization parameter introduced in the theory and describing the size of the associated electron pairs, $\xi_0 = 0.18\hbar v/kT_c$ (where v is the velocity at the Fermi surface), is different in these groups. For superconductors of the "London" type, ξ_0 is substantially smaller than the penetration depth of the magnetic field δ_0 throughout the entire range of temperatures. The "Pippard" type of superconductors correspond to the other limiting case, when $\xi_0 \gg \delta_0$ in the entire range of temperatures, with the exception of the immediate vicinity of T_c. The majority of superconductors belong to the Pippard and intermediate types, differing in the size of the London region. Thus, for tin, this region amounts to $\Delta T = T_c - T \approx 0.1$—$0.15$ K, and for aluminum it is $\Delta T \lesssim (10^{-3})$°K. [3,4]

It was shown by Gor'kov [5] that in the London region the behavior of superconductors is well described by the Ginzburg-Landau equations. For pure superconductors of the Pippard type the region of applicability of the local equations is very small, but it can be much increased by the presence of foreign impurities and various lattice defects, since the scattering of electrons at these inhomogeneities diminishes the spatial correlation of the electrons in the superconducting state. For a sufficiently large concentration of impurities, the role of correlation length is transferred from the dimension of the pair, ξ_0, to the mean free path of the electrons. In the case of thin films of pure superconductors the boundaries of the individual crystallites act as additional scattering centers, which diminish the mean free path of electrons.

FABRICATION OF SPECIMENS AND PROVISION OF LOW TEMPERATURES

The aluminum films were condensed on plain-polished glass substrates with platinum leads fused through them. The films were produced and studied in an apparatus similar to one previously described. [6]

The necessity of obtaining very pure specimens led to a number of difficulties due to the great chemical activity of aluminum. The usual method of obtaining aluminum mirrors by evaporating aluminum from a tungsten heater is not applicable in our case. The best results were obtained by evaporating the aluminum from tungsten wires covered by a protective layer of the ceramic Al_2O_3, and for these the cathode heaters of vacuum tubes were used. The heaters had the form of loops on which were fixed the aluminum charges. The charge was previously melted and conditioned in high vacuum, after which the prepared evaporator was mounted in the apparatus. The power given out by the evaporator did not exceed 1 w.

Before fabricating the films, the apparatus was evacuated by a diffusion pump to 10^{-6} mm Hg and heated in an oven at ~ 300°C for 3—4 hours; after the conditioning, the vacuum in the apparatus was ~ 5—7×10^{-7} mmHg. The evaporation rate was 2—6×10^{-4} g/min. After condensation was complete the apparatus was filled with helium and sealed off.

The assemblies for low temperature condensation were sealed off from the apparatus after outgassing, and were mounted in a helium Dewar. The thicknesses of the films were calculated from the weights of evaporated aluminum. The change of film thickness with condensation time was monitored by the resistance. Films of thicknesses of

0.2×10^{-5} to 14×10^{-5} cm were obtained and studied.

To obtain temperatures of the order of 1°K, a well-known method was used (see [7]). Inside the helium Dewar was placed a second Dewar of somewhat smaller dimensions (dia 30 mm, length 250 mm), which served as the working space. Pumping was by a mercury diffusion pump DRN-50 with a speed of 30 l/sec through connecting tubes of large cross section. A general view of the helium apparatus is given in Fig. 1.

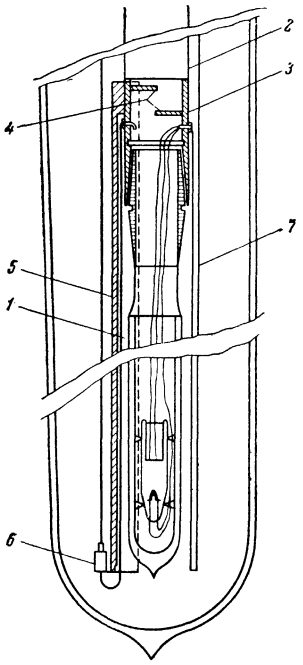

FIG. 1. General view of the apparatus.

The assembly was placed in the internal Dewar 1 and held by a cork collar. The join between the Dewar and the pumping tube 2 was made with a copper union 3, coated with "Ramsay" vacuum putty, which provided a reliable seal down to very low temperatures. To decrease the heat flux, the pumping tube proceeding upwards was made of stainless steel, and inside the transitional coupling were soldered, at some distance from one another, two screens 4, which cut across the entire cross section of the tube. The temperature of the coupling was maintained at 4.2°K with the aid of a thick copper plate 5 dipping into the liquid helium. The inner Dewar was filled with liquid helium to the desired level through the valve 6 fastened to the lower end of the plate. The electrical leads were brought out through a thin German silver tube 7, the lower portion of which dipped into liquid helium.

In the apparatus it was possible to attain and maintain temperatures from 4.2°K to 0.9°K. The temperature of the specimens was measured with a carbon thermometer. The calibration of the thermometer changed somewhat from experiment to experiment, and was monitored by the superconducting transition of massive aluminum.

MEASUREMENTS AND RESULTS

The resistances of the films were measured with a conventional potentiometer circuit. The specimens obtained by condensation in liquid helium were studied both directly after fabrication and after an anneal up to room temperature or 300°C for 3—4 hours.

1. The critical temperature T_c for the superconducting transition of a film ($R/R_0 = 0.5$, where R_0 is the resistance in the normal state) was determined from a graph of the variation of resistance with temperature in zero magnetic field.

Directly after condensation on a cold substrate the films displayed anomalously high values of T_c —up to 2.6°K. For individual specimens (as a rule, those deposited rapidly) the critical transition temperature even attained 3—3.2°K, while T_c measured on a specimen fabricated from the original aluminum was 1.160°K. The anomalous increase of T_c for freshly-condensed aluminum films has also been established by Hilsch. [8]

After a long anneal (about one day) at room temperature, the value of T_c for the films fell to ~ 1.5°K. Subsequent anneal at 300°C for 3—4 hours reduced the critical temperature to values coinciding with T_c for films fabricated at room temperature. Increasing the annealing temperature to 400°C did not effect a change in T_c. As is seen from Fig. 2, the critical temperatures of annealed films and of films deposited at room temperature exceed the transition temperature of bulk aluminum by 0.1°K, and display a pronounced variation on specimen thickness; this is in distinction from mercury films, for which the value of T_c after anneal coincided with the transition temperature of a bulky specimen, and did not depend on film thickness. [6]

2. The absolute value of the film conductivity σ was determined. To do this, the resistances of films of various thicknesses in the form of strips of dimensions 0.09×0.71 cm were measured in the normal state. The conductivity of freshly-deposited films directly after condensation was 0.35×10^{17} cgs esu, but the conductivity of very thick recrystallized specimens was $\sigma = 4.3 \times 10^{17}$ cgs esu. Thus, after the anneal the conductivity increased more than 10 times. However, not only

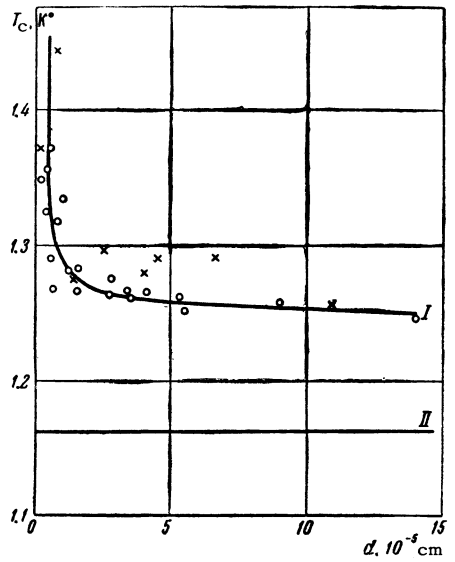

FIG. 2. The variation of T_c on film thickness d. Curve I: o – films evaporated at room temperature; x – films evaporated in liquid helium and annealed to 300° C. Curve II: bulk aluminum.

the absolute values of the conductivity, but also the characters of the variation of σ on film thicknesses d are different. As is seen from Fig. 3, the conductivity of freshly-deposited films remains constant down to the smallest studied thicknesses (0.2×10^{-5} cm), whilst for recrystallized films and films deposited at room temperature σ was constant only down to 1.0×10^{-5} cm, after which a sharp decrease was observed.

Such behavior of the normal conductivity is easily explained when the difference in the structure of the specimens studied is taken into account. As is well known, condensation at liquid helium temperature leads to the formation of a very dispersed structure, as a result of which the mean

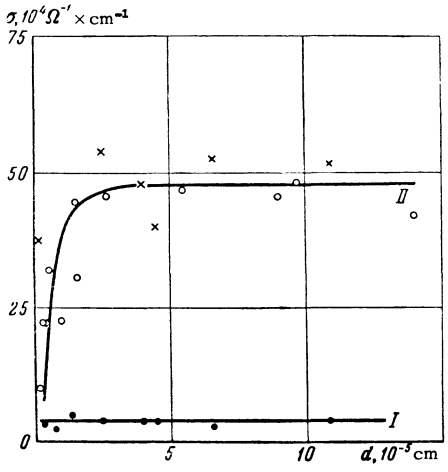

FIG. 3. The variation of film conductivity σ on thickness d. • – freshly deposited films; x – films annealed to 300° C; o – films evaporated at room temperature.

free path of electrons in freshly-deposited films is substantially less then their thickness, and the scattering of electrons at the boundaries of the film plays no part. In the process of annealing the crystallites expand, so that for sufficiently thin films they become of the same dimensions as the film thickness. The mean free path now starts to depend significantly on the film thickness. In the range of thicknesses for which the conductivity in the normal state is constant (for freshly-deposited specimens this is true in all cases), it is, of course, possible to consider the films as massive metal, and, for estimating the mean free path, it is possible to use the value of σ/l measured in massive aluminum; $\sigma/l = 20.4 \pm 2.0 \times 10^{-11}$ Ω^{-1} cm^{-2}.[9] From the values of σ obtained, the following estimates were made: $l = 1.9 \times 10^{-7}$ cm for freshly-deposited specimens; $l = 2.3 \times 10^{-6}$ cm for specimens recrystallized or obtained at room temperature. These values agree well with the variation shown in Fig. 3, and do not contradict the data of other workers. Thus, for freshly-deposited mercury films, the value $l \approx 5 \times 10^{-7}$ cm has been obtained,[10] and, according to Faber's data,[11] for a bulky polycrystalline aluminum specimen of 99.89% purity $l = 40 \times 10^{-6}$ cm.

3. For all the films, curves were taken of the destruction of superconductivity by a magnetic field directed parallel to the plane of the film. In Figs. 4 and 5 are given a series of $H_c = f(\Delta T)$ curves for films of various thicknesses deposited, respectively, at room temperature and helium temperature. As is seen from Fig. 4 for thin films ob-

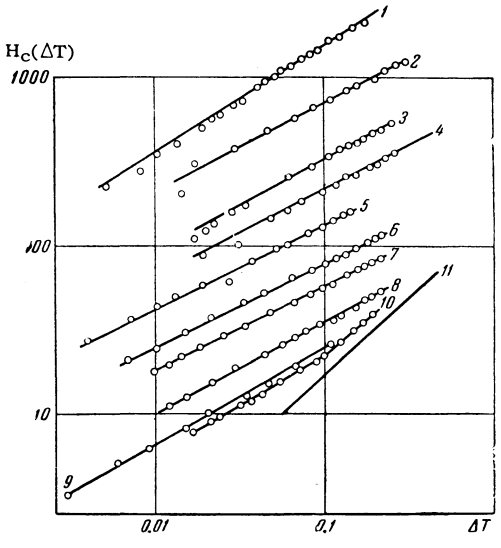

FIG. 4. The variations of H_c with ΔT for films condensed at room temperature. Film thicknesses in 10^{-5} cm: 1 – 0.2; 2 – 0.37; 3 – 0.57; 4 – 1.2; 5 – 1.6; 6 – 2.8; 7 – 3.5; 8 – 5.5; 9 – 9.7; 10 – 9.0; 11 – massive aluminum.

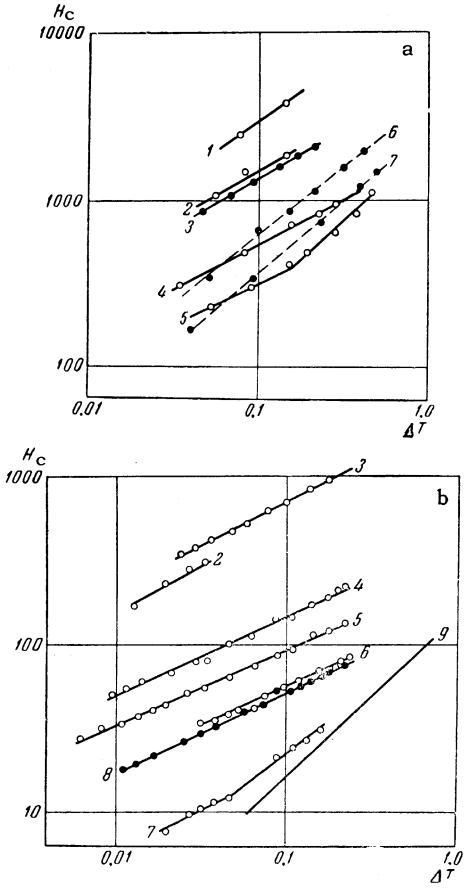

FIG. 5. The variations of H_c with ΔT for films condensed at liquid helium temperature: a – freshly-deposited films; b – films annealed to 300° C. Film thicknesses in 10^{-5} cm: 1 – 0.32; 2 – 0.12; 3 – 0.73; 4 – 1.35; 5 – 2.5; 6 – 6.6; 7 – 10.9; 8 – 4.5; 9 – massive specimen.

tained by condensation at room temperature we have $H_c \sim (\Delta T)^{1/2}$. For a very thick specimen ($d = 9.0 \times 10^{-5}$ cm), a characteristic break was observed in the variation of H_c with ΔT. The temperature corresponding to this change in the character of the dependence $H_c = f(\Delta T)$ coincided with the appearance of hysteresis in the destruction of superconductivity by the field. Thin freshly-deposited films obeyed the law $H_c \sim (\Delta T)^{1/2}$ with sufficient accuracy (Fig. 5a). For one of the thickest specimens a break in the curve $H_c(\Delta T)$ was observed, to the right of which $H_c \sim \Delta T$. Two specimens (6 and 7) failed to obey the general rule; these were obtained at high deposition rates (specimen 6 also displayed the highest value of T_c). After annealing, these specimens were no longer exceptions, and they then followed the usual law; $H_c \sim (\Delta T)^{1/2}$ for thin films. For the film of maximum thickness this relation tended with increasing ΔT to H_c for massive aluminum (Fig. 5b). The absolute values of H_c for recrystallized films are reduced to approximately one-tenth, and in their

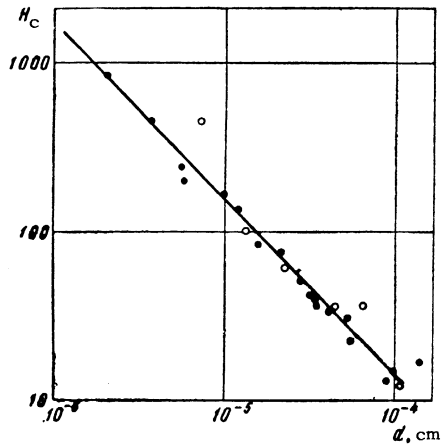

FIG. 6. The variation of H_c with d for $\Delta T = 0.04°$ K: ● – films obtained at room temperature; o – films condensed at 4.2° K and annealed to 300° C.

properties they are not different from specimens made at room temperature. The critical field for these films is inversely proportional to their thickness (Fig. 6).

DISCUSSION OF RESULTS

Pure aluminum belongs to the superconductors of the Pippard type. Therefore in the entire temperature range except the immediate neighborhood of T_c ($\sim 10^{-3}$°K) its behavior in a magnetic field cannot be described by the local equations. However, when films are condensed on a cold substrate, and even one at room temperature, a system of very small crystals is obtained, in which the mean free path of electrons is substantially smaller than in massive aluminum ($l \ll \xi_0$; for Al $\xi_0 = 1.6 \times 10^{-4}$ cm). As in the case of alloys, this allows us to use the local equations for a much wider range of temperatures.

For recrystallized films and films condensed at room temperature, the experimental values of the normal conductivity were used to calculate the constant coefficient in the penetration length, δ_{00}, according to the formula of the new theory of superconducting alloys due to Abrikosov and Gor'kov.[12]

Close to T_c, for $l \ll \delta_0$, we have

$$\delta_0(\Delta T) = (c/3.06\pi)\sqrt{\hbar/2k\sigma\Delta T}.$$

Putting $\delta_0(\Delta T) = \frac{1}{2}\delta_{00}(T_K/\Delta T)^{1/2}$, we obtain the value

$$\delta_{00} = (16.5 \pm 0.7)\cdot 10^{-6} \text{ cm}.$$

To calculate δ_{00} from the critical magnetic fields of the films, a formula from the macroscopic Ginzburg-Landau[13] theory for thin films was used. Close to T_c

$$\delta_{00} = H_c d \left[6T_c \Delta T\right]^{-1/2} \Big/ \left(\frac{dH_{cm}}{dT}\right)_{T_c}.$$

Measurement of H_c for films of various thicknesses showed that $H_c \sim 1/d$ and $H_c \sim (\Delta T)^{1/2}$. Thus, the value $\delta_{00} = (13.5 \pm 0.7) \times 10^{-6}$ cm was obtained, which agrees well with that calculated from the normal conductivity.

To handle the data for the freshly-deposited films, the conclusions of the theory developed by Abrikosov [14] for the case $\kappa > 1/\sqrt{2}$ were used. Consideration of the variation $H_c/H_{cm} = f(1/d\sqrt{T})$ obtained experimentally made it possible to evaluate δ_{00} using this theory, and the result $\delta_{00} = (3.9 \pm 0.3) \times 10^{-5}$ cm was obtained.

From the value of the normal resistance $\delta_{00} = (4.1 \pm 0.2) \times 10^{-5}$ cm. The values of δ_{00} obtained in the present work significantly exceed previously reported measurements of this quantity in bulky aluminum specimens. [15,16] Thus, δ_{00} for freshly-deposited films is approximately 10 times, and for recrystallized films 3 times larger than the corresponding value for massive aluminum. This increase of δ_{00} is possibly due to the decrease in the mean free path of electrons in a film, caused by its finely crystalline structure. This explanation is in qualitative agreement with the dependence of δ_{00} on l predicted by Gor'kov [17] for superconducting alloys.

In conclusion, I express great gratitude to A. I. Shal'nikov for guidance in the work, and to A. A. Abrikosov and N. V. Zavaritskiĭ for discussing the results obtained.

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Translated by K. F. Hulme
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