EFFECT OF MAGNETIC FIELD ON THERMAL CONDUCTIVITY AND ENERGY GAP OF SUPERCONDUCTING FILMS*

D. E. Morris and M. Tinkham†
Department of Physics, University of California, Berkeley, California
(Received April 27, 1961)

We have measured the change in the thermal conductivity of superconducting tin and indium films upon application of a magnetic field in the plane of the film. These experiments were undertaken to explore the dependence of the energy gap and the penetration depth upon magnetic field, and to determine the thermodynamic order of the field-induced superconducting transition in films.

Sample films are evaporated onto glass substrates held at 77°K, after ion-bombardment cleaning of the substrate surface. During an experiment, a typical temperature drop along the film is 0.1°K with ~10^{-7} watt total heat transport along the sample film and substrate. Because of its relatively large thickness (~0.01 cm), the substrate carries most of the heat despite its very low thermal conductivity. The change from superconducting to normal alters the temperature drop by ~0.001°K. Changes of ~2 microdegrees in the temperature drop along the film are detectable. This extreme differential temperature sensitivity is achieved by use of a carbon resistor heater-thermometer at one end of the film, connected in a bridge circuit with an identical reference thermometer located at the opposite end of the film. A copper rod connects the latter end to a helium bath whose temperature is controlled to ~0.001°K by a vapor pressure regulator. Balance of the thermometer bridge is measured by a dc microvoltmeter. Fields up to 1000 oersteds are produced by a water-cooled Helmholtz pair, and the angle of the field with respect to the surface of the film can be adjusted to within 0.02° by auxiliary coils. In practice, field tilts of ±0.4° produce negligible changes in the experimental results. To reach the high critical fields of very thin films, we use an iron-core electromagnet. The film thickness is estimated from the measured critical field and published data[1] relating the critical field to thickness.

Experimental results for films of tin and indium of thickness ~700 A are shown in Fig. 1. For both metals the thermal conductivity increases nearly as H^2 up to the critical field. At the critical field, the thermal conductivity in the superconducting state joins smoothly onto the field-independent conductivity of the normal metal. This indicates that the superconducting transition in a magnetic field is second order for films of this thickness, in contrast to the first-order transition of bulk superconductors in a field.

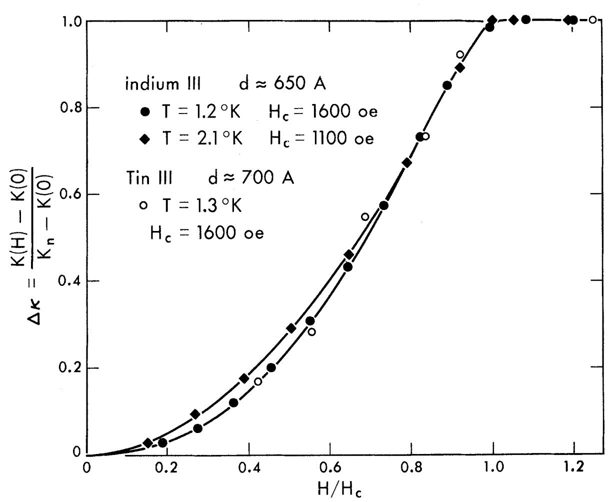

FIG. 1. Change of thermal conductivity of thin superconducting films with magnetic field.

Orientation of the field parallel and perpendicular to the direction of heat flow in the film produces the same effect upon the thermal conductivity to within the experimental accuracy, for films ranging between 700 and 2800 A. Since the induced diamagnetic currents in the superconductor are perpendicular to the field, our data also indicate that the thermal conductivity does not depend upon the angle between the thermal gradient and the diamagnetic current. Thus, if the observed variation of the thermal conductivity is explained in terms of a decrease of the energy gap of the BCS theory[2] with increasing field, the modified gap remains essentially isotropic. This result is interesting in view of the theoretical prediction[3] of a $\vec{p}_F \cdot \vec{v}_{drift}$ term in the excitation spectrum of a superconductor carrying a uniform current. The absence of an observable effect of this sort might be due to the fact that in these films the two equal and opposite surface currents are only slightly separated in space. Alternately, the short-mean-free path in these "dirty" samples may upset the simple prediction. The lack of anisotropy is important in evaluating the results of Giaever and Megerle[4] concerning the magnetic field dependence of the tunnel effect between superconductors, since their method can only determine the energy gap normal to the surface of the film, which is perpendicular to the directions of both the applied magnetic field and induced current.

The results of Fig. 1 may be used to compute a dependence of the energy gap upon field in a simple way if the electronic term K_e dominates the thermal conductivity, and if K_e is primarily limited by electron scattering by lattice imperfections. Under these conditions the ratio of the thermal conductivity in the superconducting state K_{es} to that in the normal state K_{en} is given by Bardeen, Rickayzen, and Tewordt[5] as their Eq. (3.6). This may be written

$$\frac{K_{es}}{K_{en}} = G(\epsilon_0/kT) = \int_{\epsilon_0}^{\infty} E^2(\partial f/\partial E)dE \Big/ \int_0^{\infty} E^2(\partial f/\partial E)dE,$$

where $f(E/kT)$ is the Fermi function. If we assume that the effect of a field upon the superconducting state can be adequately represented as a change of the ϵ_0 of BCS, then the observed quantity $\Delta\kappa$ plotted in Fig. 1 should be given by

$$\Delta\kappa = \frac{K_{es}(H) - K_{es}(0)}{K_{en} - K_{es}(0)} = \frac{G[\epsilon_0(H)/kT] - G[\epsilon_0(0)/kT]}{1 - G[\epsilon_0(0)/kT]}.$$

This relation was used to invert the experimental data on indium III to yield $\epsilon_0(H)$, with the result shown in Fig. 2. Because $G(\epsilon_0/kT)$ initially drops only quadratically as ϵ_0/kT increases from zero, the gap must drop more steeply to zero near H_c than the approach of $K_{es}(H)$ to K_{en} there. The difficulty of distinguishing between a broadened transition because of film inhomogeneity and a truly rounded approach to K_{en} because of the form of $G(\epsilon_0/kT)$ results in an uncertainty in the choice of H_c. The bars on the plotted experimental points in Fig. 2 indicate the size of this uncertainty.

Our results for the dependence of the energy gap upon magnetic field in a thin film are qualitatively similar to those from the more direct measurements of Giaever and Megerle,[4] although their data were taken on a thicker film at a much higher reduced temperature (T/T_c) and in a different metal (aluminum). From our data in Fig. 2, $\epsilon_0(H)/\epsilon_0(0)$ seems to approach $1 - (H/H_c)^2$ at low temperatures, but it is moderately well fitted at $T/T_c = 0.63$ by $\epsilon_0(H)/\epsilon_0(0) = [1 - (H/H_c)^2]^{1/2}$. The latter form is that given by Douglass[6] based on the Landau-Ginzburg-Gorkov theory, which is expected to hold near T_c.

With thicker films, new features appear as shown in Fig. 3. The thickest film, tin II with

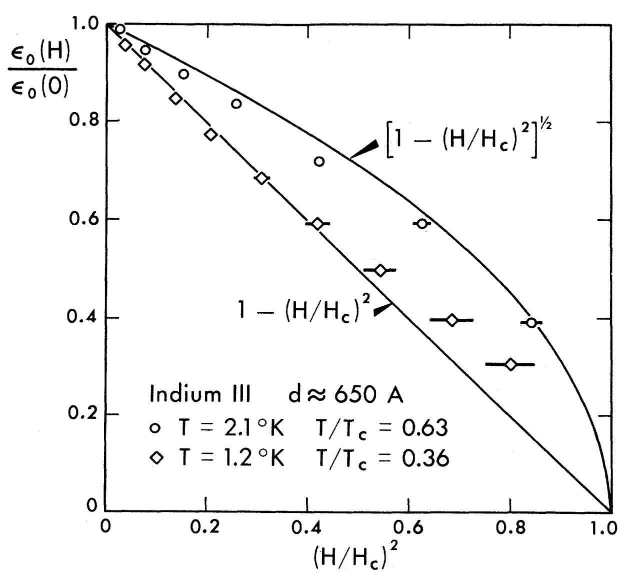

FIG. 2. Magnetic field dependence of superconducting energy gap computed from data of Fig. 1.

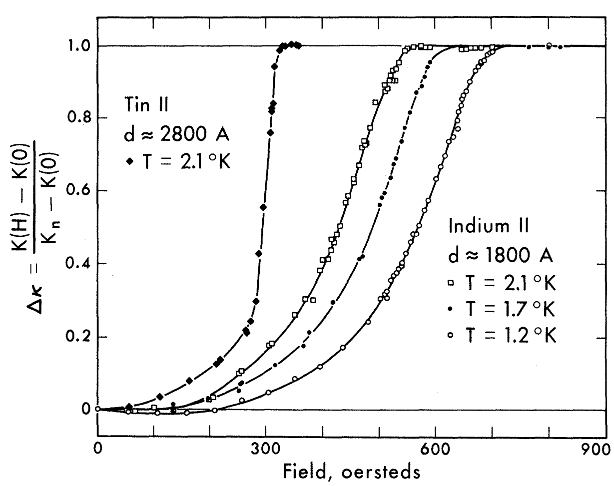

FIG. 3. Change of thermal conductivity of superconducting films of intermediate thickness with magnetic field.

$d \approx 2800$ A, has $\Delta\kappa \sim H^2$ up to the vicinity of H_c, where it reaches only $\sim 25\%$; then K appears to rise discontinuously to K_n, indicating that the transition is first order in films of this thickness. By our method of analysis, we find that $\epsilon_0(H)/\epsilon_0(0)$ drops abruptly from ~ 0.83 to 0. If we apply the theory of reference 6, the observed critical field ratio for this film, $H_c/H_c(\text{bulk}) \approx 1.53$, implies that $d/\lambda \approx 3.6$ and $\epsilon_0(H_c)/\epsilon_0(0) \approx 0.78$. The agreement with the above value of 0.83 is quite good considering the limited accuracy and the fact that the gap will not be completely uniform in a film as thick as this one. The film indium II, of intermediate thickness $d \approx 1800$ A, is still thin enough to display a second-order transition, but the increase of K near H_c is much faster than H^2. This behavior may be explained in terms of an increase of the penetration depth λ as the gap decreases, as would be expected on the basis of sum-rule arguments.[7] Since the film thickness is smaller than a coherence length ξ_0, we expect the energy gap to be nearly independent of position in the film. We may then apply the approach of Pippard,[8] considering the balance between condensation energy and magnetic energy to determine the gap (or order parameter) as a function of field. Taking account of the dependence of film susceptibility on (d/λ) and of the dependence of λ on ϵ_0, one is led to expect a change in the form of $\epsilon_0(H/H_c)$ of the observed sense for films with $d > \lambda$. A similar conclusion follows from the explicit theory of reference 6. Only in a film thin enough ($d \ll \lambda$) so that the penetration is always nearly complete will there be a definite limiting relation between $\epsilon_0(H)/\epsilon_0(0)$ and H/H_c.

In indium II at the lowest temperature we see an apparent initial decrease of thermal conductivity with field. We believe that this represents a decrease in the phonon conductivity K_g, produced by increased electronic scattering of phonons as the energy gap is decreased. This interpretation is reasonable, as the initial decrease is seen at the lowest temperatures and in a relatively thick film, both conditions favoring K_g over K_e, and it is observed in indium, having a lower Debye Θ than tin. The observed K_n indicates an electronic mean free path limited by defect scattering to $l \sim 200$ A. Then K_g, limited only by boundary scattering, should be comparable in magnitude to K_e, as required for the validity of this interpretation.

*Supported in part by the National Science Foundation, the Alfred P. Sloan Foundation, and the Office of Naval Research.

†Member of the Miller Institute for Basic Research in Science.

[1]D. Shoenberg, Superconductivity (Cambridge University Press, New York, 1952), Fig. 71(b) on p. 169.

[2]J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. 108, 1175 (1957).

[3]N. N. Bogoliubov, J. Exptl. Theoret. Phys. (U.S.S.R.) 34, 58 (1958) [translation: Soviet Phys.–JETP 7, 41 (1958)].

[4]I. Giaever and K. Megerle, Phys. Rev. 122, 1101 (1961).

[5]J. Bardeen, G. Rickayzen, and L. Tewordt, Phys. Rev. 113, 982 (1959).

[6]D. H. Douglass, Jr., Phys. Rev. Letters 6, 346 (1961).

[7]M. Tinkham and R. A. Ferrell, Phys. Rev. Letters 2, 331 (1959).

[8]A. B. Pippard, Phil. Mag. 43, 273 (1952).