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# Magnetic Flux Flow and Fluctuation Effects in Thin-Film Superconductors*†

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Resistance measurements have been used to investigate magnetic flux pinning, flux motion, and fluctuation effects in thin films of superconducting indium, thallium, and aluminum which were condensed onto cold substrates and maintained below 20 °K throughout the measurements. The depinning threshold of vortex motion was studied as a function of transport-current density, applied perpendicular magnetic field, and temperature. The absence of the peak effect in films is explained in terms of Pearl's theory of long-range electromagnetic interactions between vortices in films. Our critical-state data do not satisfy the empirical equation of Kim *et al.* which relates the current density to the critical depinning field in bulk samples. Vortex guiding has been observed, and Hall-effect measurements are also reported. Measurements on superimposed films of indium and thallium suggest that the surfaces of these films play an important role in flux pinning. Measurements of the resistive transition in most of our samples showed a peak in resistance near the transition temperature, similar to resistance peaks noticed by some other investigators. This peak disappeared as a perpendicular magnetic field was applied. An explanation of the peak is suggested; it involves small-angle electron scattering from regions in which superconducting fluctuations are occuring.

## I. INTRODUCTION

In recent years, a considerable amount of work has been devoted to the structure of quantized magnetic vortices in superconducting films. Tinkham[1] showed theoretically that a sufficiently thin superconducting film in a perpendicular magnetic field should exhibit a vortex structure (and hence a magnetic flux structure) similar to that found in Abrikosov's mixed state[2] for a bulk type-II superconductor. This has been verified indirectly by experiments,[3-12] and Tinkham's theory has been extended and made more rigorous.[13-16]

The motion of these magnetic vortices (flux flow) in thin superconducting films has also received attention. Giaever has shown experimentally that the motion of vortices in one superconducting film can induce similar motion in another, and that this generates an electric field.[17] Sherrill[18] and Deltour and Tinkham[19] have performed further experiments to investigate this phenomenon. Magnetic vortices can be pinned to imperfections in the film. Deltour and Tinkham have calculated and measured the magnetic field dependence of this pinning as a function of angle.[20] Huebener and Seher have determined the minimum temperature gradient and current density required to depin vortices.[21] Tholfsen and Meissner have made resistance measurements of flux flow in thin films of tin.[22] The study of vortex motion in bulk type-II superconductors had meanwhile been pursued intensively.[23,24] Fundamental to the study of vortex motion in films and in bulk samples have been the theory of Friedel *et al.*[25] of the forces exerted on vortices, the theory of Anderson[26] of the thermal activation of vortex motion, and Bean's concept[27] of the critical state, in which the force exerted on each vortex by the externally applied field and by the other vortices is just canceled by pinning forces. Some of the important details of vortex motion remain topics of theoretical contention. This is particularly true of the Hall angle.[28,29]

We have studied vortex pinning and vortex motion as a function of temperature, transport-current density, and magnetic field in thin films of indium, thallium, and aluminum. The magnetic field was always perpendicular to the sample's surface. We have observed the influence of the proximity effect[30] on the motion of vortices. We have measured voltages both longitudinal and transverse to the current flow. The Hall angle has been determined, and guided vortex motion[31] has been observed. For most of our films the resistive transition to the superconducting state exhibited an interesting phenomenon which we attribute tentatively to fluctuations, and we have investigated its sensitivity to a perpendicular magnetic field. We have noted no magnetic hysteresis in any of the variables which we have measured. In thicker samples, such hysteresis is frequently observed for a perpendicularly oriented magnetic field.[9] In Sec. II we describe the experimental apparatus and techniques. In Sec. III our results are presented and discussed.

## II. APPARATUS AND TECHNIQUES

The evaporator cryostat which was built for this experiment fitted into a liquid-helium Dewar, and its moving parts were controlled by rotating shafts from the top of the cryostat.[32] To achieve uniform film thickness[33] and to prevent the diffusion of one film into another, each film was condensed onto a carefully cleaned, very smooth glass substrate[34] cooled by liquid helium, and was maintained below 20 °K throughout our measurements. Gold electrodes had previously been deposited onto the substrate to provide electrical connections. Figure 1(a) shows the arrangement of these electrodes.

All of our indium films were 2.55×0.90 cm, and all of our thallium and aluminum films were 3.20 ×1.05 cm. The edges of the film with the lower transition temperature always overlapped those of the other film so a short circuit would not occur. The indium and thallium (both 99.999% pure) were evaporated from resistively heated molybdenum boats. The aluminum (also 99.999% pure) was evaporated from a stranded tungsten wire.[33] In order to drive off any surface contamination, the production of the films was begun by partially evaporating the metals onto a rotating mask positioned just above the boat or filament. The mask was then rotated out of the way, allowing the metal to deposit on the substrate while the rotating mask and a fixed partition protected the other evaporation sources from contamination. The film thickness was controlled by monitoring the electrical resistance as the film condensed on the substrate. The evaporation rates ranged from 20 to 50 Å/sec. During the film evaporations, the pressure at the upper end of the evaporator cryostat rose from $1\times10^{-7}$ to about $1\times10^{-6}$ Torr and the substrate warmed from 4.2 to about 18 °K. Figures 1(b) and 1(c) show the appearance of the substrate after evaporation of the first and

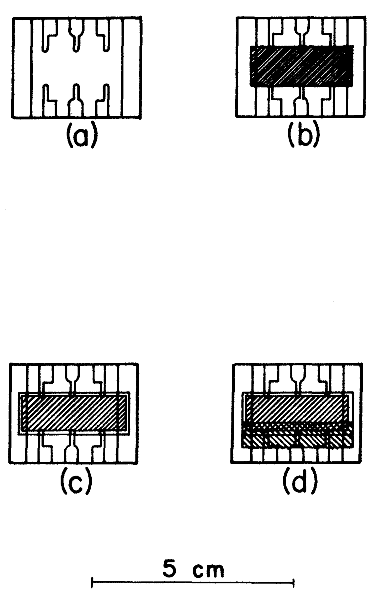


FIG. 1. Appearance of the substrate after deposition of (a) the gold electrodes; (b) the first film; (c) the second film; (d) the indium overlay.

second films. At the conclusion of a run, and before warming the system,[35] another indium film was condensed onto the substrate as shown in Fig. 1(d) to enable us to make optical measurements of the film thickness. A three-position sliding mask was used to produce films of the desired shape, and to cover the films at all times except during their production, thereby protecting the films from some of the gas molecules which would otherwise condense onto them. The substrate was thermally bonded to a copper block by a very thin layer of vacuum grease.[36]

The sample temperature was measured by an encapsulated germanium resistance thermometer, and was controlled by an automatic helium-bath pressure regulator and a carbon resistance thermometer and electronically controlled heater.[37,38] The thermometers were calibrated against the vapor of liquid helium according to the 1958 scale of temperatures.

A highly uniform dc magnetic field, always oriented perpendicular to the sample surface, was generated by a superconducting solenoid which was constructed according to the sixth-order design of Garrett.[39] The field strength was determined from the solenoid current by using the calculated field-current ratio. This ratio was subsequently verified by means of a calibrated rotating-coil gaussmeter.[40] A voltage proportional to the field strength was displayed on the horizontal axis of an $X$-$Y$ recorder. The other voltages of interest were displayed on the vertical axis.

Our ac measurements were made by using an ac transport current through the film and measuring the ac voltage longitudinal or transverse to the transport current with a low-noise preamplifier and lock-in amplifier. The voltage sensitivity was 0.1 $\mu$V. Since most measurements of flux and flux motion reported in the literature were made with a dc transport current and a dc voltage, we also employed a dc technique to check for any possible differences between these two methods. Results obtained with the two methods were compared at frequencies up to 1000 Hz. Separate runs were made on three films to do this. Spot checks were also made on other samples from time to time. There was no difference between the results of the ac and the dc measurements of the critical depinning magnetic field. We have, however, noted in one case small differences (about 5%) in the shape of the resistive magnetic transition. We have also noted in two cases differences (about 10%) in the transverse voltage. In most of the films studied, the longitudinal and transverse voltages were the same with the two methods, within the experimental uncertainty. Most of our measurements were made at a frequency of 1000 Hz.

During measurements of transverse voltages, the small misalignment of the voltage probes was corrected by potentiometrically balancing to zero the transverse voltage in the sample's normal state by introducing a small fraction ($\sim 10^{-4}$) of the longitudinal voltage.

The film thicknesses were measured optically with multiple-beam interferometry.[41,42] The uncertainty of the measurements ranged from ± 20 Å for films 100 Å thick to ± 75 Å for films 1000 Å thick. Forty samples have been examined by us in the course of this work.[32]

## III. RESULTS AND DISCUSSION

### A. Critical-State Measurements on Single Films

The critical state[27] of a superconductor can be defined as that in which vortex motion is just experimentally detectable. The critical threshold of vortex motion is a function of transport current, magnetic field, temperature, and material structure.[24] Previous workers on the critical state fixed the magnetic field and swept the transport current until some predetermined detectable longitudinal voltage appeared. It has been our experience that a sharper transition occurs when one fixes the transport current and sweeps the magnetic field slowly, and we have usually proceeded in this manner. (Of course, excessive heating from eddy currents must be avoided.) We have used 0.1 $\mu$V as our critical-state criterion.

In presenting our data, the transport-current density $J_T$ has been conveniently calculated by assuming it to be uniform throughout the film, although it is known that $J_T$ is actually peaked near the edges of the films.[43,44] In the case of ac currents, the peak values are given. Figures 2 and 3 show our critical-state data for an indium film of 150 Å thickness. The data shown are typical of all of our sin-

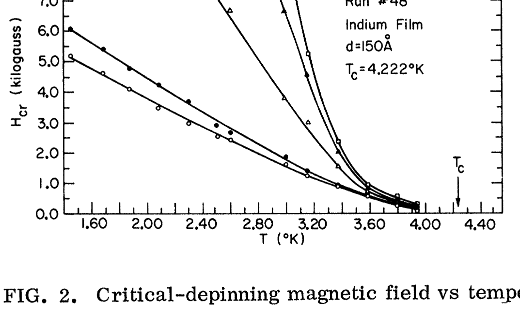


FIG. 2. Critical-depinning magnetic field vs temperature at constant transport-current density for an indium film. Open circle: $J_T = 1.5\times10^3$ A/cm$^2$; solid circle: $J_T = 7.4\times10^2$ A/cm$^2$; open triangle: $J_T = 7.4\times10^0$ A/cm$^2$; solid triangle: $J_T = 7.4\times10^{-1}$ A/cm$^2$; open square: $J_T = 7.4\times10^{-2}$ A/cm$^2$.

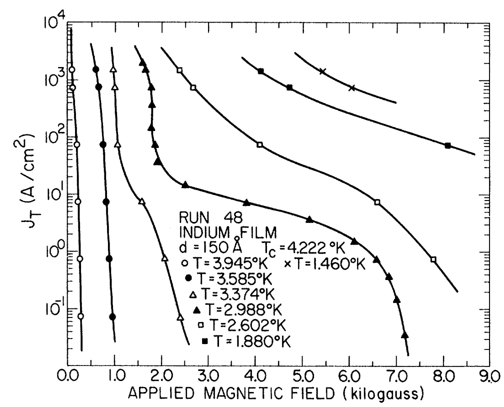


FIG. 3. Transport-current density $J_T$ vs critical depinning magnetic field $H_{cr}$ at constant temperature for an indium film.

gle films of indium, thallium, and aluminum for which such data were obtained (approximately 20 films in all).

The general shape of the curves showing $J_T$ vs the critical-state magnetic field $H_{cr}$ are similar to those of foils and bulk samples.[45,46] However, for bulk samples, this curve sometimes has a fairly sharp peak just before its decrease at high fields. No evidence for this "peak effect" was noted in any of our $J_T$-vs-$H$ curves. In attempting to explain the peak effect in bulk samples, Pippard[47] has suggested that as the field is increased, the rigidity of the vortex lattice decreases more rapidly than the pinning strength of inhomogeneities. This would allow vortices to adjust their positions relative to the pinning centers and to become more strongly pinned at high fields. This increased pinning would decrease the observed longitudinal voltage.[48] In the case of thin films, we suggest that the longer range[13] of the electromagnetic interaction forces between vortices may maintain a relatively more rigid vortex lattice and thereby destroy the peak effect.

We have tried to fit our critical-state data to an empirical relation of Kim *et al.*[23] which characterizes the critical state in terms of a constant $\alpha$ (independent of $H$ but dependent on $T$):

$$\alpha = J_T (H_{cr} + H_0),$$

where $H_0$ is a constant. If this equation is satisfied, then a plot of $J_T$ vs $J_T H_{cr}$ should be linear with slope $(-H_0)^{-1}$ and intercept $\alpha/H_0$. Such a plot of our critical-state data for each of our films is nonlinear. This is indicated by Fig. 4 for one of our indium films. Similar plots have been reported by Kim *et al.*[23] and by Aron and Ahlgren[49] for bulk samples; they do show a nearly linear behavior.

We have observed a reduction in the amount of vortex pinning after annealing our films (e.g., about 20% reduction for an indium film of 100 Å thickness, evaporated at 17 °K and then annealed to 32 °K for 15 min, at $T/T_c = 0.70$ and $J_T = 1.0 \times 10^3$ A/cm²). Similar effects of annealing on the critical state have been reported for bulk samples.[50]

A significant feature of our critical-state data is that the critical depinning magnetic field depends only weakly on the magnitude of the transport-current density. This is clear from Fig. 2. In our films, an increase in the transport current apparently contributed only weakly to the Lorentz force exerted on vortices. This bears on the still undecided theoretical question of the way in which the current flows near a vortex.[28] Huebener and Seher have drawn the same conclusion from their experimental measurements of the Nernst effect and the critical depinning current density in superconducting lead films.[21]

### B. Resistive Magnetic Transition and Critical-State Measurements on Binary-Film Systems

We have performed a series of experiments on double-film systems in order to study the effect of the proximity of a second superconductor[30] on the mixed state, the resistive magnetic transition, and the critical state of the two-film system.

Table I summarizes our binary-film data. The transition temperatures of our thin films are in good agreement with those in the literature.[51–53] For the indium-thallium system, the change in the transition temperature due to the proximity effect is consistent with the results of Jacobs and Ginsberg.[52]

In Table I, the transition temperature is defined as the temperature at which the film reached half its normal-state resistance. The transport-current density was on the order of 1 A/cm² for measurements of the transition temperature.

Figure 5 shows the dependence of the critical depinning field $H_{cr}$ on temperature for an In-Tl binary-film system. (In designating a system's two films, we always list first the material of the film which was produced first.) Critical-depinning data

TABLE I. Summary of our binary-film data. Film A was evaporated first and B second.

| Run No. | System | $T_c^A$ (°K) | $T_c^{A+B}$ (°K) | $d^A$ (Å) | $d^B$ (Å) |
|---|---|---|---|---|---|
| 33 | In(A) + Tl(B) | 4.145 | 3.880 | 190 | 90 |
| 34 | In(A) + Tl(B) | 4.275 | 3.400 | 190 | 190 |
| 38 | In(A) + Tl(B) | 4.235 | 3.490 | 155 | 325 |
| 39 | In(A) + Tl(B) | 4.270 | 3.148 | 170 | 700 |
| 40 | Tl(A) + In(B) | 2.898 | 3.250 | 550 | 200 |
| 42 | Tl(A) + In(B) | 2.910 | 3.530 | 300 | 180 |
| 43 | Al(A) + In(B) | 2.282 | 3.178 | 280 | 185 |
| 50 | In(A) + In(B) | 4.277 | 4.296 | 200 | 200 |

were obtained for the indium film by itself, and then for the binary film. The thallium film apparently reduced the strength of vortex pinning in the temperature range from 2.10 to 2.90 °K as shown in Fig. 5. The superconducting order parameter was expected to be depressed by the proximity of the thallium film.[30,52] This would tend to reduce the vortex pinning as in Deltour and Tinkham's experiment[20] on the dependence of vortex pinning on a parallel magnetic field. Below 2.10 °K, where the order parameter in the thallium film was not so small, the binary-film system exhibited stronger pinning than the single indium film. This was perhaps a result of the pinning sites added by the second film. We have examined four In-Tl systems, and they all showed the type of behavior indicated in Fig. 5.

We have made similar critical-state measurements on two Tl-In binary-film systems, again obtaining critical-depinning data for the first film and for the binary-film system, but producing the thallium film first. The data for one of these two samples are shown in Fig. 6 and the other one displayed a similar behavior. The characteristics of the thallium film referred to in Fig. 6 are consistent with those of our other single films. However, the characteristics of the Tl-In system (measured with

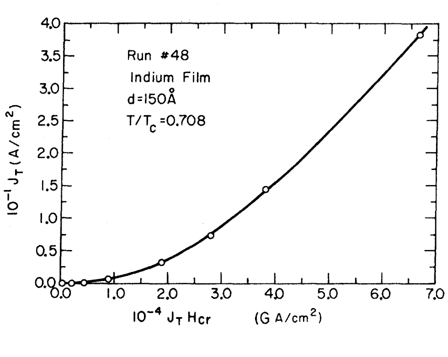


FIG. 4. Transport-current density $J_T$ vs $J_T H_{cr}$ for an indium film.

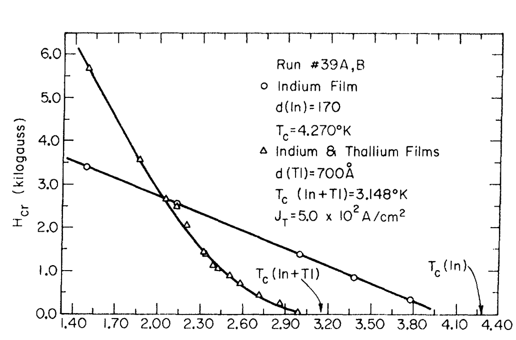


FIG. 5. Critical-depinning magnetic field vs temperature at constant transport-current density for an indium-thallium film system. The indium film was evaporated first.

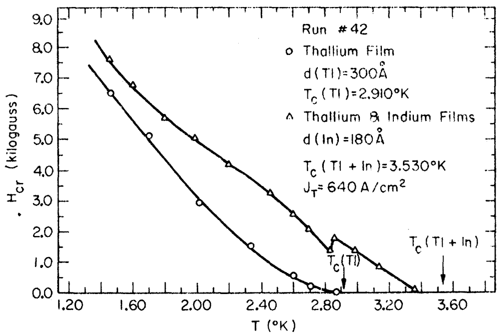


FIG. 6. Critical-depinning magnetic field vs temperature at constant transport-current density for a thallium-indium film system. The thallium film was evaporated first.

the same average transport-current density) show an abrupt reduction in the pinning strength for a temperature just below the transition of the thallium film. We propose that as the thallium film started to become more superconducting the discontinuity in the order parameter near the surface of the Tl-In interface was decreased, and the amount of vortex pinning was therefore diminished. We do not know why the dip in the curve near the transition temperature of the thallium film was never seen in binary-film systems for which the indium film was produced first. We can only suggest that the structure of the interface between the two films could have been quite different in a system for which the indium film was produced first, so the amount of vortex pinning at the interface may therefore have been much smaller when the indium film was made first.

We made one run with a binary-film system in which both films were composed of indium and were approximately 200 Å thick. The presence of the second film, which reduced the sample's surface-to-volume ratio significantly, reduced the critical depinning magnetic field (e.g., about 40% at $T/T_c = 0.90$ and $J_T = 56$ A/cm²). This would indicate that the surfaces of our films were strong pinning sites for vortices. Other experimenters[45,54,55] have drawn similar conclusions from their data. If one or both surfaces of a sample are not flat, then the theory of Bean and Livingston[56] would be relevant for this phenomenon.

### C. Transverse Voltages and Hall Angle

We have made measurements of voltage transverse to the transport current in an applied perpendicular magnetic field. All of our films had a relatively large component of transverse voltages that was an even function of the magnetic field and an odd function of the transport current. Transverse voltages of this nature, which would be induced by longitudinal vortex motion,[48] have been commonly observed in bulk samples and have been called non-Hall voltages. They are thought to be due to vortex guiding by the sample's inhomogeneities, such as those related to the direction in which a foil sample was rolled to reduce its thickness.[31]

For our films, the transverse voltage, as a function of applied perpendicular magnetic field at constant transport-current density and temperature, rose sharply to a peak for low fields and then more slowly fell to zero for increasing applied field. The shape of this peak, and even its sign, could be altered by changing the magnitude of the transport current. The magnetic field at which the transverse voltage just fell to zero always corresponded to the field at which the longitudinal voltage just reached the normal-state voltage. The peak at low fields suggests that vortex guiding effects were strongest for relatively low vortex densities.

We observed a small component of the transverse voltage which was an odd function of the applied magnetic field as well as an odd function of the transport current. We take this component to be a true Hall voltage. However, precise measurement of the Hall voltage was made difficult by the much larger non-Hall transverse voltage. (All Hall-voltage measurements were made at $T/T_c = 0.9$ and with $J_T \cong 10^3$ A/cm².) We were able to measure the Hall voltage in three films, two of indium and one of thallium. In those films, we found the Hall voltage to be proportional to the applied perpendicular magnetic field down to a field at which the longitudinal resistance had decreased by a factor of 2 from its normal-state value. Typically, the Hall angle was on the order of $10^{-4}$ rad at the magnetic field required to restore the full normal-state resistance.

### D. Fluctuation Phenomena near Transition Temperature

We have measured the resistive transition into the superconducting state (in zero applied magnetic

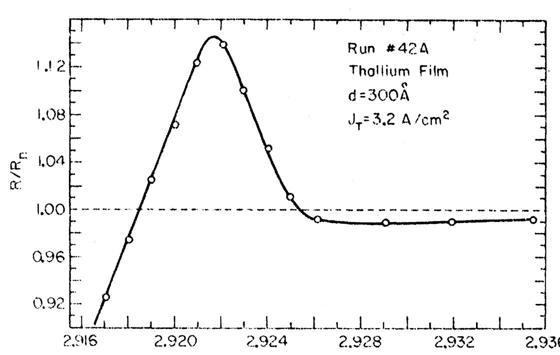


FIG. 7. Ratio of the longitudinal resistance to the normal-state resistance vs temperature for a thallium film. $T_c = 2.910$ °K and $T_c$ (width) = 0.019 °K where $T_c$(width) is the temperature width between $R/R_n = 0.90$ and $R/R_n = 0.10$.

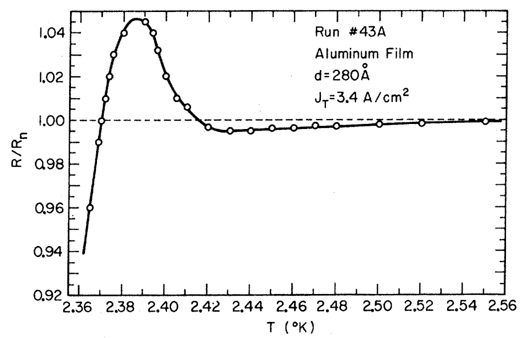


FIG. 8. Ratio of the longitudinal resistance to the normal-state resistance vs temperature for an aluminum film. $T_c = 2.282\,°K$ and $T_c$(width) = 0.132 °K where $T_c$(width) is the temperature width between $R/R_n = 0.90$ and $R/R_n = 0.10$.

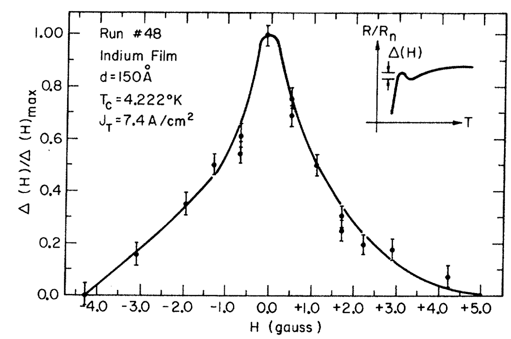


FIG. 9. Effect of an applied perpendicular magnetic field on the peak in resistance. The vertical component of the earth's magnetic field has been added to the applied magnetic field to yield the values shown.

field) for many of our films. All of them showed a gradual decrease in resistance with decreasing temperature well above $T_c$ which is similar to some previous observations.[57,58] However, about 60% of our films showed a peak in resistance just before entering into the superconducting state. These results were the same for ac current as for dc current. Figures 7 and 8 show this peak in resistance for a thallium and an aluminum film, respectively. It was also observed for indium films.

We observed a similar resistance peak for many of our double-film systems. We have noted no systematic factors that would suggest why a particular film or double-film system did or did not show the peak.

An applied perpendicular magnetic field can destroy this peak. Figure 9 shows this for the case of an indium film. We have examined the effect of an applied perpendicular magnetic field on the peak for only one other sample. This was an In-Tl binary-film system in which the In was 170 Å thick and the Tl film was 700 Å thick. For this system, an applied field of 540 G was necessary to destroy the peak.

Schwidtal[59] observed a similar peak in resistance for thin films of Pb and for films of Pb with up to 2.1-at.% Gd. His films were evaporated onto quartz substrates cooled by liquid helium. These films had dimensions of 1.0×10.0 mm. Schwidtal was unable to give an explanation for the resistance peak. More recently, Ogushi *et al.* have observed a related resistance peak for type-I Sn films and for type-II Pb-In alloy films as a function of parallel magnetic field rather than of temperature.[60] Grassie and Green have reported an excess resistance in aluminum films near $T_c$ of up to 10% of the normal-state value in disordered aluminum films.[61] They reported that the film resistance exceeded the normal-state value in the absence of a magnetic field near $T_c$ and in the presence of either a parallel or a perpendicular field below $T_c$.

Ogushi *et al.* have suggested that the increase in resistance may be related to the surface-grain structure of their films. Grassie and Green proposed that the peak is associated with the voltage across junctions arising from a distribution of superconducting grains separated by thin insulating barriers. Masker *et al.*[62] had made the same suggestion to account for a more gradual increase in the resistance of granular aluminum films as the temperature was lowered toward the transition temperature. However, this tunneling mechanism does not explain the temperature dependence of the resistance which we have observed, namely, a sharp peak which is confined to a small temperature range near the transition temperature. We therefore propose the following speculation as another possible mechanism.

It is well known that at low temperature, electrical conduction in a flat film can be dominated by electrons moving nearly parallel to the sample's surface[63] if the electron mean free path $l_0$ is comparable to the film thickness $d$. (In our films, $l_0/d$ ranged approximately from 0.3 to 0.7.) Near $T_c$, electrons may be deflected slightly as they enter or leave local regions which are fluctuating into the superconducting state.[64] This increase in low-angle scattering would give rise to increased surface scattering and hence an added resistance. This would be particularly likely to be displayed by very flat films, such as those deposited on our very smooth substrates.[34]

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