results for a system constrained to maintain $X_s = 1$ and $X_V = 0$. As expected, these agree in the region $kT/\epsilon \lesssim 0.8$ where $X_s - X_V \simeq 1$ for the Ising system, but differ considerably for higher kT/ϵ. This behavior is expected since for the constrained system $E \to \infty$ as $\epsilon/kT \to 0$, while for the Ising system, $E \to 0$ as $T \to T_c$. The result of Fig. 3 may be contrasted with that for a two-dimensional lattice-gas interface, for which E decreases smoothly with temperature to a finite value at $T = T_c$.[14] Our results are similar in form to the classical-theory results[6,7]; i.e., E increases to a maximum at some $T < T_c$, and then decreases to 0 at $T = T_c$. The interfacial free energy σ is proportional to $(1 - T/T_c)^{\mu}$ in the Ising system where the exponent μ is given as $\frac{3}{2}$ by the classical Van der Waals–Cahn-Hilliard theory,[6,7] and as 1.22–1.33 by Fisk and Widom's nonclassical treatment. Since

$$E \propto \frac{d\sigma/kT}{d\epsilon/kT},$$

we obtain from the data in Fig. 3 a value for μ between 1.27 and 1.53, in general agreement with both experiment[9-11] and nonclassical theory.[8]

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Critical Thicknesses in Superconducting Thin Films*

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Direct observations of the magnetic-field distribution in thin films of Pb, Sn, and In indicate that the critical thicknesses separating intermediate-state and mixed-state behavior in these systems are accurately given by theoretical calculations and are much lower than has been believed on the basis of critical-field measurements.

A fundamental characteristic of the equilibrium state of a superconductor is the magnetic structure which arises in an applied magnetic field H_0. Depending on the material and geometrical factors and on H_0, the equilibrium state can be perfectly diamagnetic or only paritally diamagnetic as in Abrikosov's "mixed state" of single quantum fluxoids (type-II superconductors) and in the coarser "intermediate state" found in type-I superconductors of finite dimension. The differences between the mixed and intermediate states is fundamental to the distinction between type-I and type-II superconductors, but, as Tinkham[1] pointed out, sufficiently thin films of any material should exist in the mixed state even if thicker specimens of the same material exhibit intermediate-state behavior. One expects, therefore, that thin films made of a type-I material and having a Ginzburg-Landau parameter $\kappa < 1/\sqrt{2}$ will be type-II superconductors for thicknesses below a critical value d_c. Experimental studies of this interesting change in magnetic behavior in thin-film systems have involved macroscopic properties of these systems and have been difficult to relate quantitatively to theoretical results and to data on bulk specimens. In particular, the experimental values for critical thicknesses have disagreed with theoretical predictions. We recently reported[2] direct observations of the mixed state in films of pure Pb, a type-I superconductor, using a high-resolution Bitter pattern technique which gives pictorial information on the microscopic magnetic state of a superconductor. We present here data on Pb, Sn, and In films, which show that d_c for these systems is given accurately by the Ginzburg-Landau (GL) claculations of Lasher[3] and Maki[4] and is much lower than earlier experimental values.[5,6]

The films were prepared by evaporation at a pressure less than 2×10^{-6} Torr during evaporation. The Sn and In films were grown at 77 K on air-cleaved mica substrates which were precoated with a thin layer of carbon. The Pb films were grown at room temperature on clean, cleaved mica and simultaneously on glass substrates. The evaporation rates exceeded 100 Å/sec in all cases.

To determine $H_\perp$ the critical field in perpendicularly applied magnetic fields, we recorded the resistive (dc) transitions in varying magnetic field. In accordance with the findings of Cody and Miller,[5] we have taken as $H_\perp$ the value of H_0 at which resistance is first observed in the superconducting specimen and have supposed this value to be within about 5% of the actual value for the thinner films, although $H_\perp$ is probably underestimated for films above the critical thickness.

For films sufficiently thin to show mixed-state behavior, $\kappa_1(T)=(1/\sqrt{2})H_\perp(T)/H_{cb}(T)$,[1,4] where $H_{cb}(T)$ is the bulk thermodynamic critical field and $\kappa_1(T)$ is the temperature-dependent GL parameter introduced by Maki.[7] Maki's parameter $\kappa_2(T)$, defined from the slope of the reversible magnetization curve near the critical field, is experimentally inaccessible for thin films because of their great irreversibility.[8] We therefore take $\kappa_2(T)\sim\kappa_1(T)$ for the present, but we will return to this point later.

The magnetic-field distributions were observed by the deposition of fine ferromagnetic particles on the superconducting surfaces, and the patterns obtained were observed by electron microscopy using a replica technique.[2,9] Examples are shown in Fig. 1 (the dark regions indicate the normal, flux-transmitting portions of the samples). Figure 1(a) shows the field distribution on an 1100-Å-thick In film for $H_0=50$ Oe. This type of pattern, consisting of isolated superconducting domains in a normal matrix is observed for each kind of film for sufficiently high fields and thicknesses. The structure is qualitatively independent of field history and of the decoration conditions (e.g., the amount of ferromagnetic material deposited and the particle size), and no finer structure in the normal regions has ever been detected in such a pattern. We have taken this type of structure to represent an intermediate rather than a mixed state; it is similar to intermediate-state structures reported elsewhere for thicker specimens.[10,11]

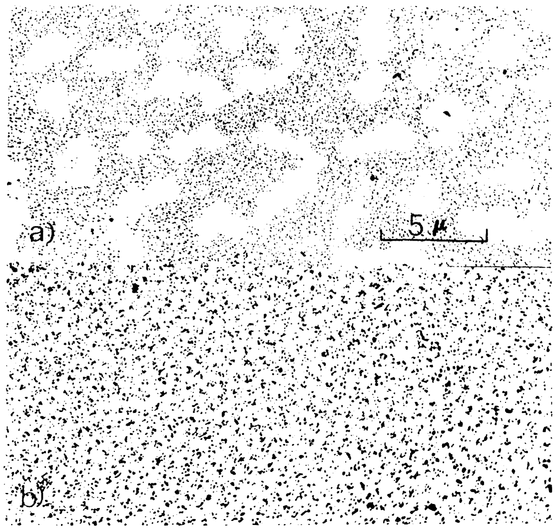

FIG. 1. Magnetic-flux distribution at the same field ($H_0=50$ Oe), temperature (2.1 K), and magnification for two indium films (a) $d=1100$ Å, $H_\perp=65$ Oe, and (b) $d=300$ Å, $H_\perp=123$ Oe (dark regions are normal).

Figure 1(b) shows the single-fluxoid mixed-state pattern on a 300-Å In film at the same magnification and applied field as in Fig. 1(a). No long-range order is present.[2] That the pattern is indeed a single-fluxoid structure is confirmed by checking that $n\varphi_0\approx H_0$, where n is the spot density and φ_0 is the superconducting flux quantum. The limit on resolution in Fig. 1(b) is brought about primarily by the small modulation in the field for the indium specimen at this applied field rather than by the spatial resolution of the decoration technique. The mixed-state pattern on a Pb specimen at 400 Oe applied field can be resolved with an effective resolution similar to that of Fig. 1(b). Even though the reduced field $h=H_0/H_\perp$ is only about 0.45 in Fig. 1(b), patterns could not be observed at very much higher fields for this film, whereas reduced fields of up to

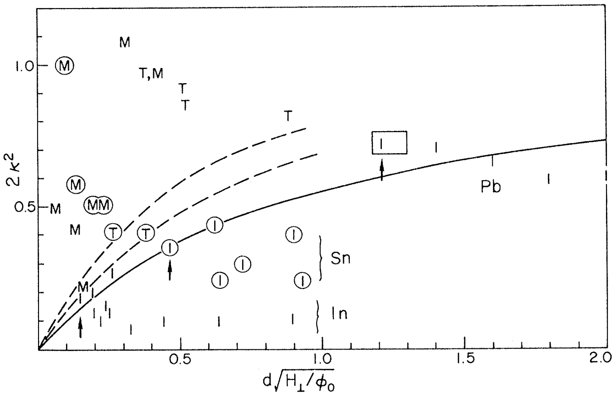

FIG. 2. Observation of a single-fluxoid mixed-state (M), multifluxoid-state (T), or intermediate-state (I) pattern, recorded for each film at the position determined by the measured values of $H_\perp$, d, and $\kappa_2 \cong \kappa_1 \equiv (1/\sqrt{2})H_\perp/H_{cb}$. The values used for the bulk critical fields are H_{cb}(Pb, 4.2 K) = 540 Oe; H_{cb}(Sn, 2.1 K) = 205 Oe; H_{cb}(In, 2.1 K) = 177 Oe. For the tin films, the symbols are enclosed in circles to help distinguish these data from the lead and indium data. The dashed and solid lines are the result of a calculation by Lasher (Ref. 3) and represent the boundaries between the intermediate and mixed states (solid line) and between different kinds of mixed states (dashed lines). We have indicated by arrows the films whose thicknesses are given in Table I, column 1. It is possible to deduce the thicknesses for the other data points from their coordinates; but since the range of thicknesses covered by the data is of general interest, these ranges are 600–4000 Å for Pb, and 300–4000 Å for Sn and In.

0.95 could be studied for thicker films in the intermediate state. Below we have recorded such observations as representing the "high-field" structure for the films although we have found[2] that often a film having a single-fluxoid structure at low fields will have a multifluxoid structure or intermediate-state structure at higher fields.

In Fig. 2 we reproduce the theoretical results of Lasher[3] for the boundaries separating intermediate-state and mixed-state behavior for a superconducting plate characterized by the GL parameter κ, critical field $H_\perp$, and thickness d. Also reproduced are the boundaries between single-fluxoid and multifluxoid mixed states and between single-fluxoid states with different fluxoid geometries. For $T < T_c$ the ordinate in Fig. 2 should actually be $2\kappa_2^2(T)$. Also in Fig. 2 we present our high-field magnetic structure observations for Sn and In at $T = 2.1$ K and for Pb at 4.2 K. Each film with thickness d (measured by interferometry) and critical field $H_\perp$ is recorded at $2\kappa^2 \cong 2\kappa_1^2(t) \equiv H_\perp^2(T)/H_{cb}^2(T)$, with the symbol M (single-fluxoid mixed state), T (multifluxoid mixed state), or I (intermediate state) indicating the type of structure we observed for the film at the highest field for which we observed a pattern. $H_{cb}(T)$ is obtained from Decker, Mapother, and Shaw[12]; for Sn and In, we have corrected these values slightly (a few percent) by requiring that $H_{cb}(0)/T_c = \text{const}$. A single point is enclosed in a box to indicate typical limits of error. The error in $d(H_\perp/\varphi_0)^{1/2}$ is derived principally from the disagreement between the thickness measured by interferometry and that obtained from a room-temperature resistance measurement. Further errors possibly arise from the difficulty of distinguishing between the different states in some cases and from the low values of the reduced field at which observations on some of the thinner films were made.

For the Sn and In films, the agreement of the observations with Lasher's theory is exact within the accuracy of our measurements. For the Pb films the intermediate state and multifluxoid mixed state are stable for values of $d(H_\perp/\varphi_0)^{1/2}$ significantly below the value predicted by Lasher's result, but this disagreement is small compared to the disagreement previously existing between the predictions of this theory and the values of the critical thickness obtained from macroscopic critical-field measurements. To demonstrate this we list in Table I the values of d_c as obtained from our observations, from Lasher's theory with our critical field data, and from the critical field data alone. Maki's calculation,[4] which is similar to Lasher's yields theoretical values for d_c which are slightly (~10% for these films) higher than the values $d_{c\text{L}}$ in Table I.

We note that the values for d_c listed in Table I are valid only for our particular set of specimens, and we expect that they are only upper bounds for the values one could obtain for more ideal specimens. The extent of the agreement with theory indicated in Fig. 2 is not expected to be specific to our specimens since we have accounted for the variations in the superconducting properties introduced by imperfections by using the measured value of κ in plotting our data.

The disagreement between Lasher's and Maki's results and our Pb-film data occurred for Pb films grown on both glass and mica substrates in this experiment, and the same level of disagreement was obtained in our earlier Pb films,[2] prepared identically to the Sn and In films of this work, so that we feel that the discrepancy is not purely specimen related. Although there

TABLE I. Values for the critical thicknesses obtained from magnetic-structure observations compared to those obtained from critical-field data (d_{cM}) and from Lasher's calculation and our critical-field data (d_{cL}).

Film	d_c [a] (Å)	d_{cL} [b] (Å)	d_{cM} [c] (Å)
Pb	2500 ± 10%	3000	9000 (Ref. 4)
Sn	1800 ± 10%	1800	5200 (Ref. 4)
In	800 ± 20%	900	6200 (Ref. 5)

[a] Thickness of the thinnest film for which we observed the intermediate state (arrows in Fig. 2).

[b] From linear extrapolations of the values obtained for temperatures near the temperatures in our study; $T = 4.2$ K for Pb and $T = 2.1$ K for Sn and In.

[c] The thicknesses determined by the intersection of the solid line in Fig. 2 and lines through our data for each metal.

are several grounds on which the Pb films can be differentiated from the Sn and In films—e.g., Pb is a strong-coupling superconductor and the Pb films were somewhat "cleaner," having larger values for the ratio of the mean free path to the superconducting coherence length—it is clear that a similar discrepancy may exist for the lower-κ region and be simply unresolved by our Sn and In data.

Assuming the validity of our results, the discrepancy may reflect a limit in the applicability of the theoretical calculations to our experiments. The calculations are local calculations based on ideal specimens and are valid close to $H_\perp$. With the qualifications mentioned earlier, we believe our observations gave the high-field magnetic structures for the films, but the films were certainly not ideal. Neither were the films sufficiently impure to be considered local, but, according to Maki,[4] local corrections would slightly increase rather than decrease the theoretical values for d_c. Alternatively, an obvious weakness in our data presentation lies in the necessary substitution of the parameter κ_1 for κ_2 in plotting our data. Usually one has $\kappa_1(T) \lesssim \kappa_2(T)$ but there have been suggestions[13, 14] that the condition $\kappa_1(T) > \kappa_2(T)$ may hold in certain situations if strong-coupling corrections are important.

We conclude that for a rather wide range of values of κ, the GL results of Maki and Lasher give at least an upper bound on the critical thickness even for ordinary, highly imperfect evaporated thin films. In fact, for our Sn and In films, the theories could be used to predict quite precisely which type of magnetic structure a film would have. The often-quoted critical thicknesses inferred from magnetization data are much higher than those we obtained, in agreement with theory. As reported[2] earlier for Pb, no qualitative or sudden quantitative change in magnetic structure is observed at these greater thicknesses, although the thickness dependence of the critical fields for our films agrees with earlier observations.

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