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Magnetic Transitions of Superconducting Thin Films and Foils. III. Pb, Sn, and In

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New measurements are reported for the superconducting-to-normal transition of Pb films to reduced temperatures (T/T_c) as high as 0.995. A comparison is made of the Ginzburg-Landau parameter, κ, for Pb, Sn, and In, derived from thin-film measurements (κ_F) and values of κ derived from supercooling and superheating studies (κ_{sc}) by Smith, Baratoff, and Cardona. The comparison reveals large discrepancies between κ_F and κ_{sc}, which is supported by similar measurements on Al by Maloney and de la Cruz. The discrepancy can be associated with a unique sensitivity of the thin-film transitions to nonlocality, but there is at present no rigorous theory.

INTRODUCTION

A basic parameter of a superconductor is the Ginzburg-Landau (GL) parameter κ. For type-II superconductors κ and the thermodynamic critical field $H_c(T)$ determine the magnitude of the various transition fields of a type-II superconductor in the vicinity of T_c: in particular, $H_{c2}(T)$, the transition field between the normal and mixed state, and $H_{c3}(T)$, the parallel transition field for the formation of the surface superconducting sheath. At low temperatures, details of the microscopic theory affect the temperature dependence of both $H_{c2}(T)$ and $H_{c3}(T)$, [1,2] as well as other factors such as spin paramagnetism, [3] the strength of the phonon-electron interaction, [4] and gap anisotropy. [5] However, at T_c, the following simple relationships hold:

$$\left(\frac{dH_{c2}}{dT}\right)_{T_c} = \sqrt{2}\,\kappa\left(\frac{dH_c}{dT}\right)_{T_c},$$

$$\left(\frac{dH_{c3}}{dT}\right)_{T_c} = (1.695)(\sqrt{2}\,\kappa\left(\frac{dH_c}{dT}\right)_{T_c},$$

where κ is simply given in terms of the London penetration depth λ_L, the Pippard coherence distance ξ_0, and the mean free path l, by the relation

$$\kappa = 0.96(\lambda_L/\xi_0) + 0.7\,(\lambda_L/l)\,. \tag{1}$$

As is well known, type-II superconductivity is observed for $\kappa > 1/\sqrt{2}$ and type-I superconductivity for $\kappa < 1/\sqrt{2}$.

The measurement of κ and its effective temperature dependence,

$$H_{c2}(T) \equiv \sqrt{2}\,\kappa(T)\,H_c(T)\,, \tag{2}$$

is a major task of superconducting research since experimental data for $\kappa(T)$ are a definitive test of the microscopic theory, as well as a source of fundamental parameters for a particular metal. Since 1964 a large body of experimental work has verified the theory for type-II superconductors where $l \ll \xi_0$. Departures from the theory have been observed for clean elemental type-II superconductors such as Nb and V, [6] where $l \gg \xi_0$, but these departures have been successfully ascribed to gap anisotropy. In connection with the present paper, it is important to note that for cubic materials such anisotropic effects vanish as T_c, as is suggested by Eq. (1).

For type-I materials where $\kappa < 1/\sqrt{2}$, $H_{c2}(T) < H_c(T)$ and the experimental and theoretical situation is not as well defined. For these materials, the critical fields H_{c2} and H_{c3} represent the supercooling fields. $H_{c2}(T)$ is the supercooling field for the bulk of the specimen and $H_{c3}(T)$ is the supercooling field for that part of the surface parallel to the field. In a supercooling experiment, the supercooling field is observed as the minimum field that can support the normal state either in the bulk or on the surface. Care has thus to be taken so that effect is not masked by inhomogeneities which might lead to premature nucleation of the superconducting state. A convenient sample for these measurements is a collection of widely dispersed small spheres, and this technique has been used to measure the supercooling field for a variety of type-I superconductors. [7] A major problem in the interpretation of the data is that the quantity of prime interest is $H_{c2}(T)$ [Eq. (2)]. However, for the usual geometries, the supercooling field is necessarily $H_{c3}(T)$. In general, we have

$$H_{c3}(T) = C(T)H_{c2}(T)\,, \tag{3}$$

where, close to T_c, $C(T)$ is given by the de Gennes–St. James[8] value of $C = 1.695$. At lower temperatures $C(T)$ can be derived from the theory[2] if sufficiently detailed microscopic data are available for both volume and surface parameters of the metal. However, $C(T)$ is not, in general, a strong function of temperature.

Supercooling studies of a variety of type-I superconductors have been recently utilized[7] to obtain $\kappa(T)$ and κ. These results are supported by elegant single-sphere measurements of Feder and McLachlan, [9] as well as by earlier data of Faber[10] on

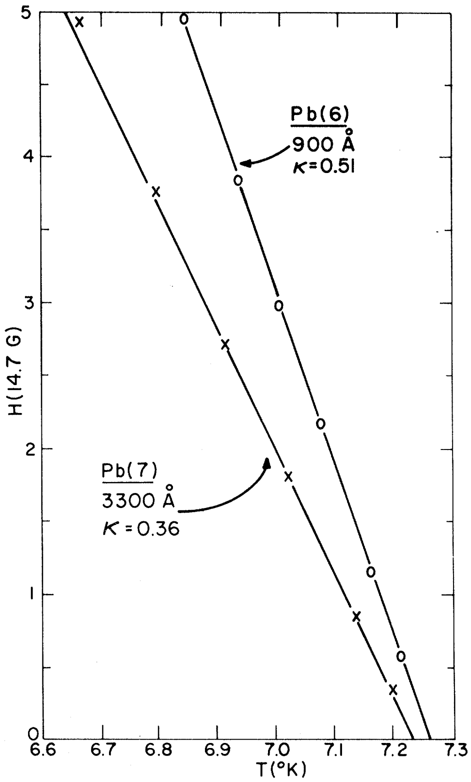

FIG. 1. Perpendicular critical field of a 900-Å Pb film and a 3300-Å Pb film, as a function of temperature close to T_c.

cylindrical specimens. In addition superheating experiments can be used to determine κ and this approach has also been used by Cardona and coworkers.[7] The excellent agreement of these different experiments supports the validity of supercooling for the measurement of $\kappa(T)$ for type-I superconductors. Unfortunately, our ignorance of the microscopic properties of Sn, In, Pb, etc., prevents a direct comparison of experimental values of κ with theory.

An alternative method for determining $\kappa(T)$ for type-I superconductors is based on the theory of the magnetic transition of thin films in parallel and perpendicular magnetic fields. The parallel critical fields of thin films were first obtained by GL[11] and later extended by de Gennes and St. James[8] to a wider range of film thicknesses. These authors extended the theory to include the existence of surface superconductivity in parallel fields. Tinkham[12] first made the calculation of the perpendicular magnetic transition of thin films and showed that for thin type-I films, a vortex *mixed state* was energetically favored over the usual *intermediate state*. He predicted that the transition field $H_\perp(T, d)$ was given by

$$H_\perp(T, d) = \sqrt{2}\,\kappa(T, d) H_c(T), \qquad (4)$$

where $\kappa(T, d)$ is an effective GL parameter for the film of thickness d at temperature T. In this theory, $H_\perp(T)$ thus corresponds to $H_{c2}(T)$. In the limit $T \to T_c$

$$\kappa(T, d) \to \kappa(d).$$

The quantity $\kappa(d)$ is thus the GL parameter of the element at T_c, where the retained thickness dependence in $\kappa(d)$ includes the possible thickness dependence of the parameters in Eq. (1). In the double limit $T \to T_c$, $d \to \infty$, one anticipates

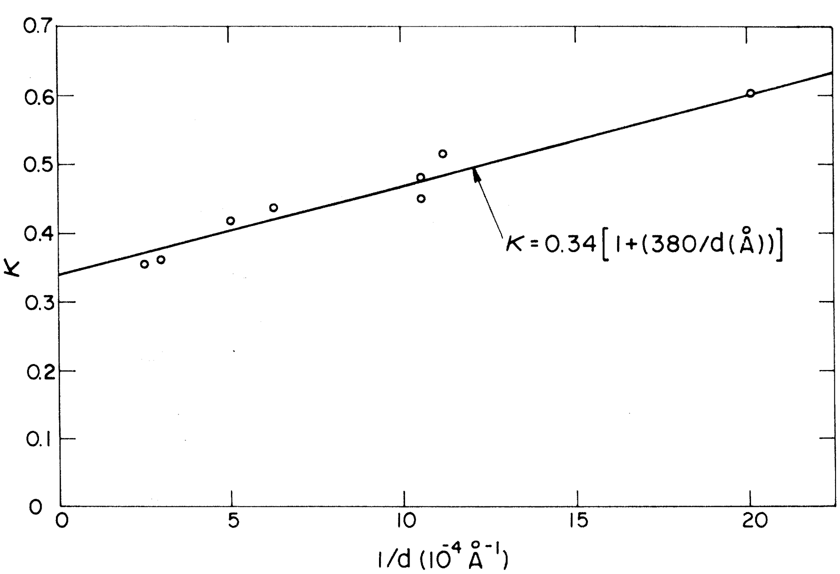

FIG. 2. κ at T_c for Pb films as a function of the inverse film thickness.

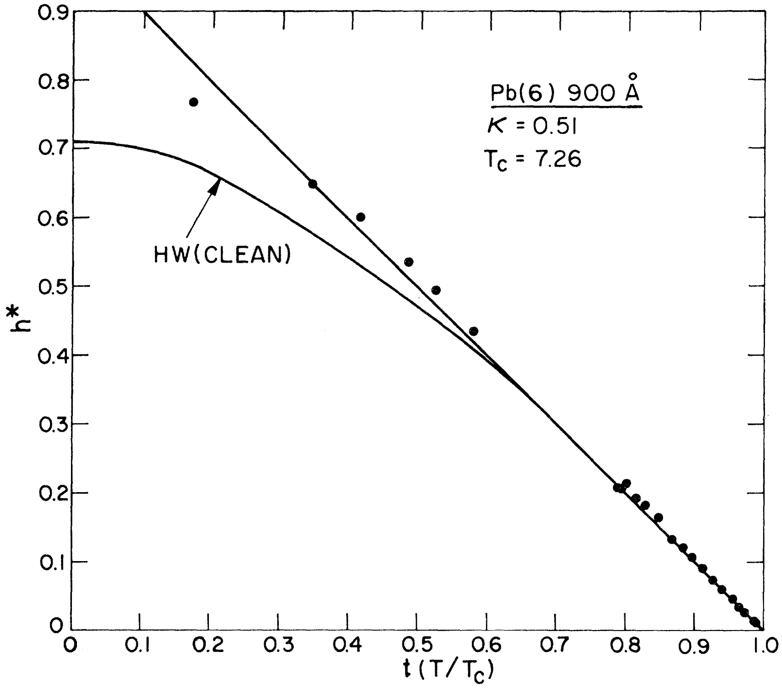

FIG. 3. The quantity h^* [Eq. (5)] as a function of reduced temperature for a 900-Å Pb film. The curve labeled HW is from Ref. 1.

$\kappa(T, d) \to \kappa$.

Tinkham's results can be physically understood from the depression of the intermediate-state critical field ($H_\perp^D$) for type-I superconductors due to the positive surface energy occurring between normal and superconducting domains. Indeed the theory of the intermediate state suggests that $H_\perp^D \to 0$ as $d \to \xi_0 l/(\xi_0/ + l)$. Clearly a mixed state with $H_\perp$ given by Eq. (4) becomes favored under these conditions. Subsequent theoretical work by Maki[13] and Lascher[14] verified Eq. (4) and the more physical argument

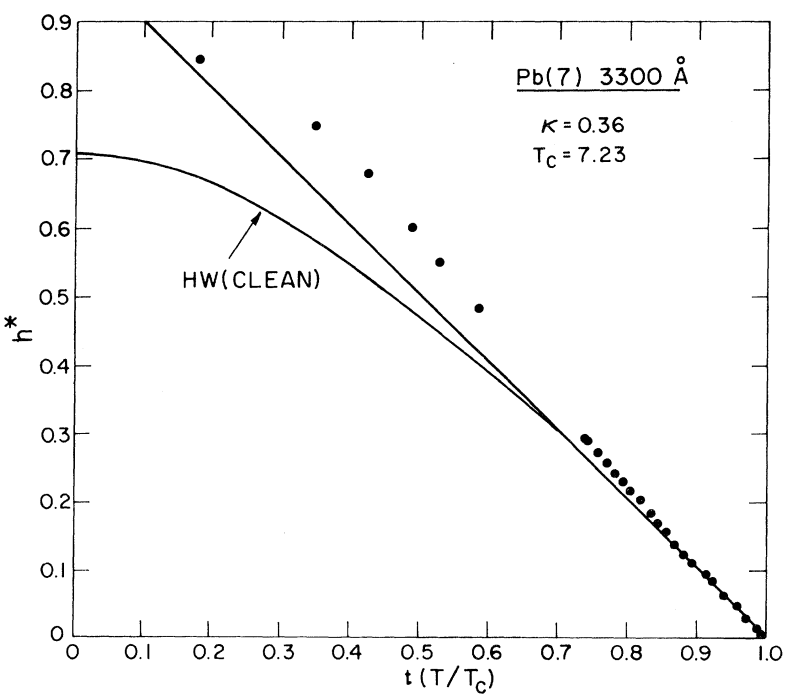

FIG. 4. The quantity h^* [Eq. (5)] as a function of reduced temperature for a 3300-Å Pb film. The curve labeled HW is from Ref. 1.

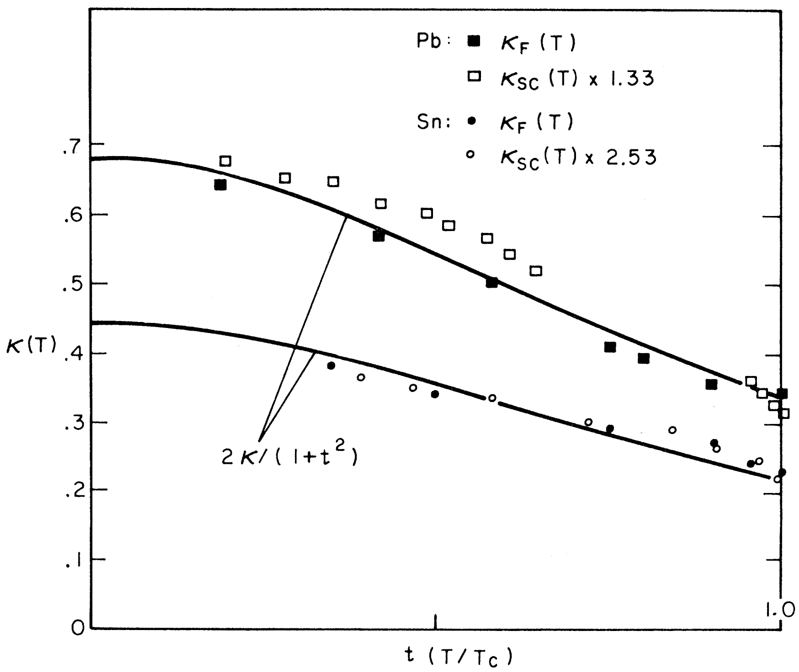

FIG. 5. $\kappa_F(T)$ for bulk Pb and Sn extrapolated from thin-film magnetic transitions as a function of reduced temperature. $\kappa_{sc}(T)$ derived from supercooling (Ref. 7) is shown multiplied by 1.33 for Pb and 2.53 for Sn.

of Tinkham.[12]

One advantage of thin-film measurements taken as a function of field, temperature, thickness, and orientation is that a set of independent data is generated for the superconducting parameters for the same films. The method of analysis and the internal consistency of the results are most clearly demonstrated by Miller and Cody[15] for tin films. The present paper extends previous measurements of Cody and Miller[16] for lead to temperatures

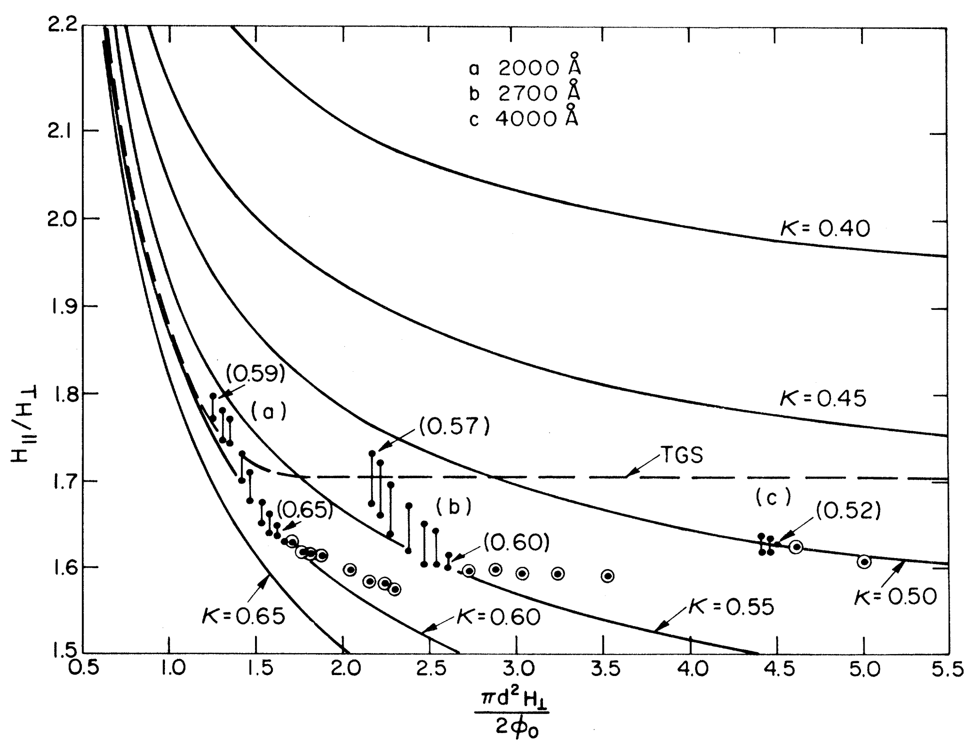

FIG. 6. Ratio of parallel to perpendicular critical fields for Pb films as a function of $\pi d^2 H_\perp / 2\varphi_0$. The second-order-transition curve is labeled TGS (Refs. 8, 15, and 16). Computed first-order transitions are shown for different values of κ from Ref. 11. The closed circles represent first-order transitions for parallel fields. The bars denote the range of hysteresis. The quantities in parenthesis are *measured* values of $\kappa(T, d)$ from $H_\perp(T, d)$.

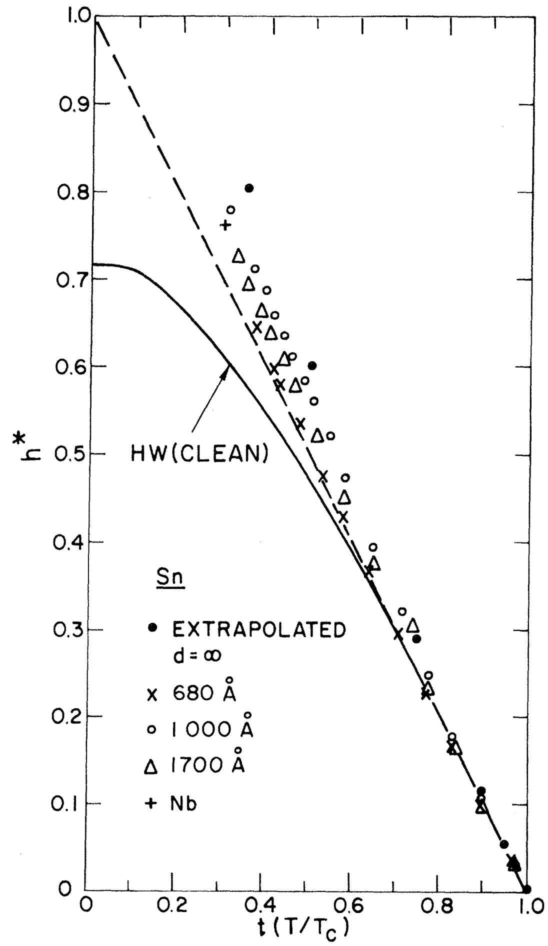

FIG. 7. Quantity h^* for several Sn films, as well as the extrapolated "infinite"-thickness limit. A data point for Nb is shown from Ref. 6. The curve labeled HW is from Ref. 1.

close to T_c. The definitive data for Pb and Sn, to temperatures close to T_c, show clearly that the magnetic transitions of thin films are in excellent agreement with the theory.[8,11–14] However, despite this agreement, large numerical discrepancies exist when the derived values of $\kappa(T)$ and κ are compared with those derived from supercooling experiments made on the same metal. The discrepancies manifest themselves as a constant ratio between $\kappa(T)$ derived from magnetic transitions in thin films $[\kappa_F(T)]$ and $\kappa(T)$ derived from supercooling $[\kappa_{sc}(T)]$ such that $\kappa_F(T) \gtrsim \kappa_{sc}(T)$ and $\kappa_F(T) = A\kappa_{sc}(T)$, where A is a constant for a particular element. Since there is no evidence for systematic experimental error in either measurement, we are confronted with a theoretical gap in our understanding of the magnetic transitions of type-I superconductors. In view of the greater complexity of the thin-film transition compared to supercooling, it is tempting to associate the problem with thin-film transitions. However, experimentally there is no reason to distinguish the measurements.

EXPERIMENTAL RESULTS

The details of the measurements, as well as preparative procedures, are described in previous papers.[14,15] In the following we discuss the individual elements.

Pb

We have recently examined films ranging in thickness from 500 to 4000 Å up to reduced temperatures of $t = 0.995$ $(t = T/T_c)$. Representative data for a 900- and 3300-Å film are shown in Fig. 1. From these and similar plots, one obtains with $(dH_c/dT)_{T_c} = -238.4$ G/°K,[17] the data shown in Fig. 2, where $\kappa(T_c, d)$ is shown plotted as a function of $1/d$. This method of extrapolation is consistent with the parallel data[16] for the same films and leads to $\kappa = 0.34$ for Pb. From the simple theory of Tinkham,[12] the coefficient of the quantity $1/d$ should be $3\xi_0/8$, suggesting that for Pb, $\xi_0 \approx 1000$ Å, and hence from Eq. (1), $\lambda_L \approx 340$ Å. The measured *bulk* mean free path due to impurities is in excess of 40 000 Å and hence can not contribute appreciably to Eq. (1). These results are in good agreement with recent penetration-depth measurements for Pb.[18]

From this and other data, the quantity

$$h^* = -H_\perp(T, d) \Big/ T_c \left(\frac{dH_\perp(T, d)}{dT}\right)_{T_c} \tag{5}$$

can be compared with the predictions of the microscopic theory in the clean limit.[1] Such a comparison is shown in Figs. 3 and 4 for a 900- and 3300-Å film and departures from the theory are apparent. In Fig. 5 we show as well data for $\kappa(T, \infty) \equiv \kappa(T)$ and one notes that the temperature dependence of h^* suggested in Figs. 3 and 4 leads to $\kappa(T)$, which has a temperature dependence given by the "two-fluid" model, i.e.,

$$\kappa(t) = 2\kappa/(1 + t^2) . \tag{6}$$

In Fig. 5 we also show the supercooling data for Pb,[7] multiplied by a *constant* ratio of 1.33. The agreement between these two sets of measurements suggests a simple temperature-independent scaling between $\kappa_F(T)$ and $\kappa_{sc}(T)$. As will be seen in Fig. 5 a similar result holds for Sn.

We have examined the thin-film data of Maldy[19] and of Koepke and Bergmann[20] and find that they are in substantial agreement with our data. In the microwave experiments of Fischer[21] values of $\kappa(T)$ were inferred from $H_{c3}(T)$ measurements, by utilizing theoretical values for $C(T)$ [Eq. (2)]. From these experiments, Fischer derived data for

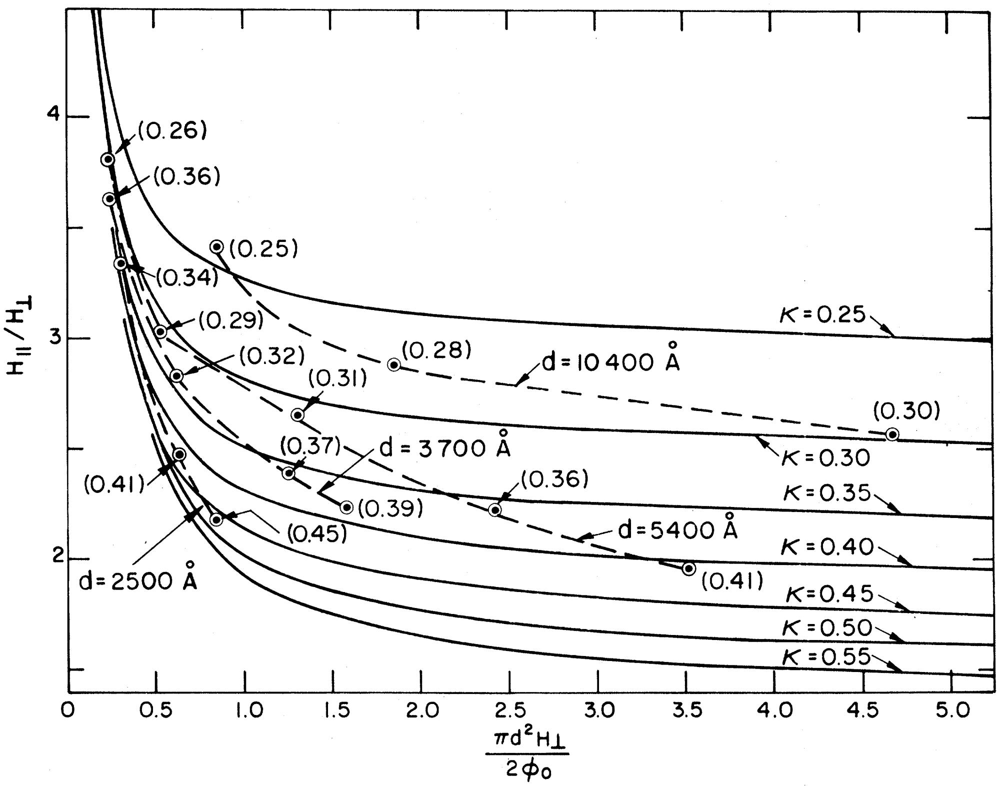

FIG. 8. Ratio of parallel to perpendicular critical fields for Sn films as a function of $\pi d^2 H_{\perp}/\varphi_0$. All parallel transitions are first order. Hysteresis is not shown. Computed first-order transitions are shown for different values of κ from Ref. 11. The quantities shown in parentheses are measured values of $\kappa(T, d)$ from $H_{\perp}(T, d)$.

$H_{c2}(T)$ in agreement with the clean limit of Helfand and Werthamer[1] (Figs. 3 and 4). Our measurements of $H_{c3}(T)$ on a film of the same thickness as Fischer's gave identical results, but in addition $H_{c2}(T) \equiv H_{\perp}(T)$ measurements were made on the same film. Unlike Fischer, we saw no evidence to suggest that the quantity $C(T)$ for Pb was substantially different from its value of 1.695 at T_c. Figure 6 amplifies this point. In Fig. 6, we show $H_{\parallel}/H_{\perp}$ as a function of $\pi d^2 H_{\perp}/2\varphi_0$, where φ_0 is the flux quantum ($\varphi_0 = hc/2e = 2\times 10^{-7}$ G cm^2). On such a plot the surface superconducting critical field is shown by the curve labeled TGS which represents a second-order transition (no hysteresis).[8] The first-order-transition curves are labeled by the parameter $\kappa(T)$ and are obtained in the $\kappa = 0$ limit of the GL theory.[11] Detailed calculations of Arp *et al.*[22] show that this is an excellent approximation. In Fig. 6 data for three films are shown for a variety of temperatures. For each film the value in parentheses denotes

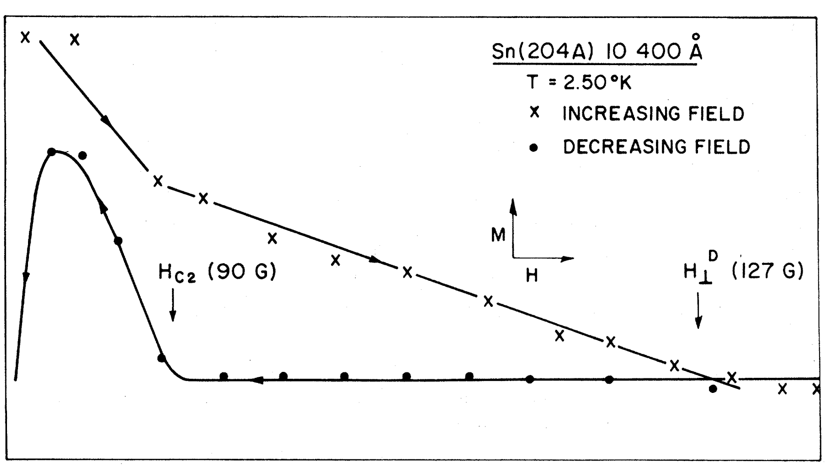

FIG. 9. Hysteretic magnetization of a 10 400-Å Sn film as a function of field close to the transition to the normal state ($H_{\perp}^{D}$). The field denoted H_{c2} denotes the assumed supercooling field. The magnetization scale is arbitrary. Complete magnetization curves are shown in Ref. 15.

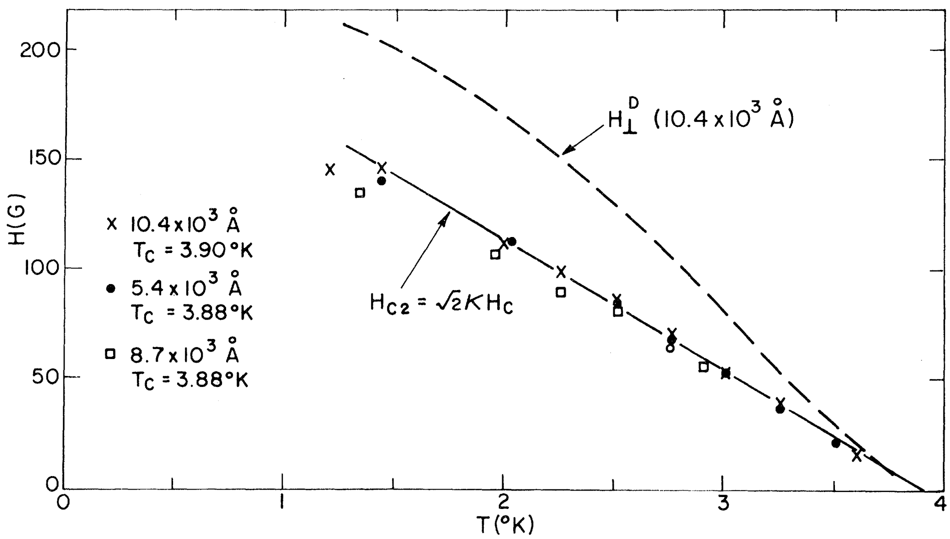

FIG. 10. Quantity $H_\perp^D(T)$ and $H_{c2}(T)$ (Fig. 9) as a function of temperature for several Sn films. The solid curve is calculated from values of κ extrapolated from the thickness domain of second-order transitions, i.e., $\kappa(T,d)$ (Fig. 10 in Ref. 15).

$\kappa(T,d)$, calculated from $H_\perp(T,d)$. As seen in Fig. 6, there is excellent agreement between measured values of $\kappa(T,d)$ [from $H_\perp(T,d)$; Eq. (4)] and computed values of κ obtained from the theory[11] [$H_\parallel(T,d)/H_\perp(T,d)$ and $(\pi d^2 H_\perp/2\varphi_0)$]. Although this method of presentation is not particularly sensitive, it does exhibit the consistency between the theory and experiment for parallel and perpendicular transitions on the same films. Particularly noteworthy in Fig. 6 is the loss of hysteresis at values of $H_\parallel/H_\perp \approx 1.6$ where the second-order transition occurs. This result should be compared with Fischer's assumption[21] that, for Pb, $H_\parallel/H_\perp \sim 1.9$. For Pb our own and other thin-film data lead to $\kappa = 0.34$, while the supercooling data give $\kappa = 0.24$.

Sn

No new results are given in this paper, since previous work presents extensive data up to T_c. However, we would like to focus on the internal consistency of the thin-film data, the significance of the data for the theory of the magnetic transition, and the relation of these data to the supercooling results.[7]

Figure 7 shows a plot of h^* as a function of temperature for several thin films. As previously observed for Pb, the data depart considerably from the theory,[1] and are again reminiscent of departures observed for Nb and V.[6] A measure of the consistency of the data is shown for the transitions exhibited in Fig. 8 ($H_\parallel$ first order, $H_\perp$ second order). The agreement with the theory is excellent, although it must be again noted that this plot is sensitive to κ only for low values of $(\pi d^2 H_\perp/2\varphi_0)$.

Figure 5, which is reproduced in part from Ref. 15, shows $\kappa(T,\infty)$ as a function of reduced temperature t and again the agreement with the "two-fluid" expression [Eq. (6)] is evident. In this figure we show the supercooling data of Ref. 7 for Sn, multiplied by a constant ratio of 2.53, and we note the excellent agreement over the entire temperature range.

Thick tin films exhibit a striking magnetic hysteresis in the transition to the intermediate state at $H_\perp^D$.[15] This hysteresis is suggestive of supercooling of the perpendicular transition and is shown in Fig. 9 for a Sn film at 2.50 °K with thickness $d = 10\,400$ Å. In Fig. 9 we note the characteristic linear variation of the magnetization in increasing fields,[15,16] but for decreasing fields the transition is characteristic of supercooling. Considering the film geometry, we will identify this supercooling field with $H_{c2}(T,d)$ rather than $H_{c3}(T,d)$.[23] In Fig. 10 we show $H_{c2}(T,d)$ as a function of temperature for films of thickness $d = 5400$, 8700, and 10 400 Å. The plot also shows $H_\perp^D$, and calculated values of $H_{c2}(T,d) \equiv \sqrt{2}\,\kappa(T,d) H_c$ obtained by extrapolation from the "Tinkham" regime. The agreement of the supercooling fields with the calculated value is striking. Figure 10 shows that the κ measured for thin Sn films agrees with the supercooling exhibited by thicker Sn films.

Finally we want to note that from $H_\parallel$ data of Tilly *et al.*[24] on Sn we infer $\kappa = 0.260 \pm 0.05$, and from the data of Chang, Kinsel, and Serin[25] we infer $\kappa = 0.21 \pm 0.01$. In this last case we exclude alloy specimens and only include data for pure films of varying thickness. We also utilize $(dH_c/dT)T_c = -154$ G/°K.[26] These data should be compared to our results of $\kappa = 0.22 \pm 0.01$,[15] and the supercooling result of $\kappa = 0.087$.[7]

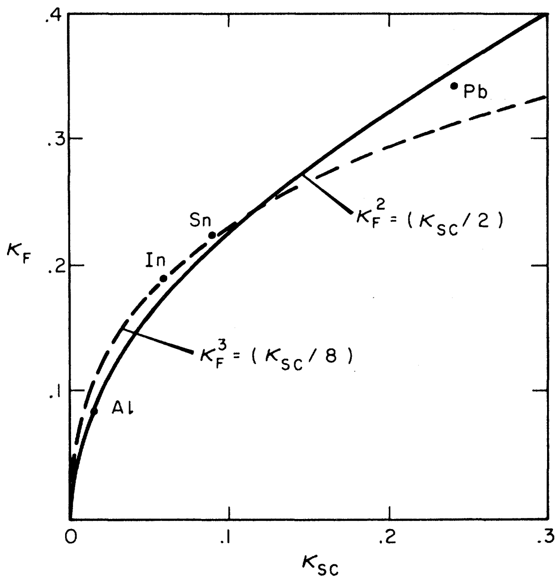

FIG. 11. Quantity κ_F determined by thin-film measurements plotted as a function of κ_{sc} determined by supercooling measurements, for Al, In, Sn, and Pb. The solid curve shown is an empirical fit to the data. The dashed curve is derived from nonlocal considerations and is discussed in the text.

In

We have made an analysis of unpublished magnetization data[27] as well as the microwave measurements of Gittleman, Bozowski, and Rosenblum[28] and thin-film magnetization data of Chang and Serin[29] and of Miller, Kington, and Quinn.[30] From measurements on pure films we find $\kappa = 0.13 \pm 0.03$ for In. We utilized $(dH_c/dT)_{T_c} = -157$ G/°K.[26] This value for κ has considerably more uncertainty than that for Sn and Pb since only limited thicknesses were utilized. It should be compared to that derived from supercooling, $\kappa = 0.060$.[7] More extensive measurements on In films by Brandt *et al.*[31] suggest that $\kappa = 0.19 \pm 0.04$. In the following, we will utilize this result.

Table I summarizes the above results, as well as recent work of Maloney and de la Cruz[32] on Al. In Fig. 11 we show κ_F derived from thin-film data as a function of κ_{sc} derived from supercooling. An empirical fit to the data that pass through the origin is shown by the solid curve. The dashed curve in Fig. 11 is derived from a simple consideration of nonlocality. Its derivation and significance will be discussed later.

DISCUSSION

Table I and Fig. 11 present the main results of this paper; namely, the clear discrepancy between the determination of κ for the type-I superconductors Sn, Pb, In, and Al from thin-film magnetic transitions and from supercooling measurements. For Sn, Pb, and In the impurity mean free path is larger than the coherence distance. For Al films an extrapolation is made to the pure limit.[32]

It would be tempting to ascribe this discrepancy to systematic errors in either of the measurements, but there is no evidence for such. The thin-film data are independent of substrate (e.g., glass, aluminum,[33] and nylon[25] give the same results), whether the edges of the film are scribed,[16] and are independent of measurement technique (microwave surface impedance, magnetization, low-frequency susceptibility, and dc resistance). Furthermore, the striking internal consistency of the thin-film measurements suggests that what is measured is a property of the film. Conversely the agreement of supercooling data for single- and multiple-sphere specimens,[9,7] the agreement with supercooling on macroscopic specimens,[10] and the agreement with κ derived from superheating[7] suggest as well that κ_{sc} is a property of the particular metal.

Since most thin films have a preferred orientation[34] one might ascribe the discrepancy between κ_F and κ_{sc} to anisotropy effects.[6] Such an explanation might account for the noncubic metals, Sn and In, but cannot account for the discrepancy exhibited by the cubic materials, Pb and Al. As is well known, anisotropy effects vanish at T_c, and as

TABLE I. Comparison of thin-film and supercooling experiments.

	κ_F	κ_{sc}	Comment
Al	0.086 (Ref. 32)	0.014 (Ref. 32)	...
In	0.19 ± 0.04 (Ref. 31) 0.13 ± 0.03[a]	0.060 (Ref. 7)	limited data[a]
Sn	0.22 ± 0.01 (Ref. 15)	0.087 (Ref. 7)	ratio independent of T
Pb	0.34 ± 0.02[a]	0.24 (Ref. 7)	ratio independent of T

[a]This work.

shown in Fig. 1, there is no evidence for such a reduction in κ close to T_c for Pb or in the work of Maloney and de la Cruz[32] for Al. Furthermore, there is no experimental evidence to suggest that anisotropy could account for such large charges in $H_{c2}(T)$.[6]

Finally one might suggest that the increase in κ is associated with impurities in the thin film limiting the mean free path in addition to the boundary scattering. Careful analysis of resistivity data for Sn and Pb[15,16] and In[31] shows that the measured films are quite pure, and boundary scattering dominates. A bulk mean free path short enough to have the contribution to κ_{sc} shown in Table I would have effectively eliminated the observed size-dependent resistance in these films.

SUMMARY

Let us summarize the conclusions of this paper:

(a) There is no evidence to suggest that thin-film measurements are more susceptible to either random or systematic experimental error than are the supercooling or superheating measurements.

(b) The internal consistency of the thin-film results with regard to field orientation suggests that κ_F is a basic parameter for the thin-film magnetic transitions and supports the initial view of Tinkham.[12] Similarly, the agreement of the small-sphere supercooling data of Cardona and associates[7] with results for microscopic specimens suggests that at T_c $\kappa_{sc}=\kappa$.

(c) The approximately constant ratio of $\kappa_F(T)$ to $\kappa_{sc}(T)$ for Sn and Pb permits the following general expression for $\kappa_F(T,d)$:

$$H_\perp(T,d)\equiv\sqrt{2}\,\kappa_F(T,d)H_c\,. \tag{7}$$

From the temperature and thickness dependences shown in Figs. 2 and 5, we have for Sn and Pb[15,16]

$$\kappa_F(T,d)=\kappa_{sc}(T)[\kappa_F/\kappa_{sc}]\,[1+b(t)/d]\,. \tag{8}$$

Table II lists values of $b(t)$ for Sn and Pb. As noted in earlier publications it is of the order of $\frac{3}{8}\xi_0$, as is suggested by a local generalization of the Tinkham model to include boundary scattering.[15,16]

There does not seem to be any theoretical explanation of the discrepancy between κ_F and κ_{sc} and it has largely gone unnoticed in the theoretical literature. The most plausible explanation of these results is not anisotropy, but the high degree of nonlocality of the electrodynamics in these low-κ films even close to T_c. As is well known, the freedom to use local electrodynamics requires that

$$\lambda(T)/\xi_0\gg 1\,. \tag{9}$$

TABLE II. The quantity $b(t)$ for Sn and Pb.

Sn	t	1.00	0.95	0.90	0.75	0.50	0.35
	$b(t)$	570 Å	540 Å	398 Å	403 Å	388 Å	355 Å
Pb	t	1.00	0.58		0.41	0.19	
	$b(t)$	380 Å	260 Å		240 Å	200 Å	

The inequality expressed in Eq. (9) can be rewritten close to T_c as

$$\kappa^2\gg 2(1-t)\,. \tag{10}$$

If we assume the identification of κ_{sc} with κ, it is clear that even for the high reduced temperatures used for Pb and Sn ($t\approx 0.985$), local electrodynamics is not satisfied. Thus the identification of κ with λ_L/ξ_0 is not exact.

A simple modification of Tinkham's expression for $H_\perp(T)$ shows the form of the expected modification of Eq. (1) to include nonlocality.[12] The quantity $\kappa_F(T)$ is given by

$$\kappa_F(T)=2\sqrt{2}\,\pi\lambda^2(T)H_c/\varphi_0\,. \tag{11}$$

If rather than the local limit for $\lambda(T)$,

$$\lambda(T)=\lambda_L/[2(1-t)]^{1/2}\,, \tag{12}$$

we use the Pippard nonlocal limit,[12] we obtain

$$\lambda^2(T)\approx(0.42)\,[\lambda_L^2/2(1-t)]\,(\xi_0/\lambda_L)^{2/3}\,. \tag{13}$$

Thus in the nonlocal limit, close to T_c we find

$$\kappa_F\approx(0.4)(\lambda_L/\xi_0)^{1/3}\,, \tag{14}$$

or, if we identify κ_{sc} with (λ_L/ξ_0),

$$\kappa_F\approx 0.4(\kappa_{sc})^{1/3}. \tag{15}$$

Equation (15) is close in magnitude and functional dependence of κ_F as a function of κ_{sc} as shown by the dashed curve in Fig. 11. However, as shown by the solid curve in Fig. 11, the data are not sufficiently complete to establish the functional relation between κ_F and κ_{sc}.

Clearly the identification of κ_{sc} with λ_L/ξ_0 and κ_F with $(\lambda_L/\xi_0)^{1/3}$ assumes that nonlocal electrodynamics dominates the thin-film transition but not supercooling. A consistent interpretation of these data requires that supercooling be independent of nonlocality in the electrodynamics. In other terms, $H_{c3}(T)$ is independent of the nonlocality of the electrodynamics, whereas $H_{c2}(T)[H_\perp(T)]$ is sensitive to it. The data shown in Figs. 9 and 10 support this conclusion.

These heuristic considerations cannot replace a theory. It is the purpose of the present paper to show that such a theory is required since the experimental situation is well defined.

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[34]Sn films had a strong preferred orientation ⟨001⟩ perpendicular to plane; Pb films had a strong preferred orientation ⟨111⟩ perpendicular to plane.

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Investigation of Microwave-Induced dc Voltages across Unbiased Josephson Tunnel Junctions*

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We have studied the power dependence of the microwave-induced dc voltages across "unbiased" Pb—Pb oxide—Pb Josephson tunnel junctions and found a systematic dependence. We have also found that the Josephson effect in the form of induced quantum voltages was observable in a large dc magnetic field as high as 0.5 kG, about one-half of the H_{c2} of the superconducting Pb film.

I. INTRODUCTION

It is commonly known that when a Josephson junction is exposed to external microwave radiation of frequency ν and dc biased at a voltage $V_n = nh\nu/2e$, a zero-resistance dc current will flow resulting from the interaction between the microwave field and the ac Josephson current.[1] These dc currents appear in its current-voltage (I-V) characteristic as current steps[2] at constant voltages