Discrepancies in determinations of the Ginzburg-Landau parameter

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Long-standing discrepancies within determinations of the Ginzburg-Landau parameter κ from supercritical field measurements on superconducting microspheres are reexamined. The discrepancy in tin is shown to result from differing methods of analyses, whereas the discrepancy in indium is a consequence of significantly differing experimental results. The reanalyses, however, confirm the lower κ determinations to within experimental uncertainties.

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I. INTRODUCTION

The Ginzburg-Landau parameter of a superconductor, κ, is generally defined as the ratio of its magnetic penetration depth (λ) to its coherence length (ξ) at the thermodynamical critical temperature (T_c). The parameter relates the two fundamental length scales of the material's superconducting phase, distinguishes between type-I and type-II superconductors, and characterizes the material's response to applied magnetic fields in the superconducting state. Theoretically, for clean materials, it is given by the BCS $\kappa=0.96\lambda_L(0)/\xi_0$, where $\lambda_L(0)$ is the London penetration depth at zero temperature and ξ_0 is the Pippard coherence length.[1,2]

The parameter, although often referred to, is infrequently measured and little tabulated owing to its dependence on sample purity and structure. Where measured, κ has been determined[2] from independent measurements of λ and the thermodynamic critical field H_c, as well as from magnetization measurements on thin films and foils of various materials.[3,4] For type-I materials ($\kappa<1/\sqrt{2}$), another way to determine κ is from measurements of the supercritical fields of microspheres. This technique has been used in determining the κ of aluminum,[5,6] cadmium,[7] gallium,[8] mercury,[5,9,10] indium,[5,11–13] lead,[5,9] tin,[5,11,14–16] thallium,[5] and zinc,[6] as well as a series of alloys.[5] Inexplicably, the results are generally $\sim$ factor 2 smaller than the thin film and/or foil determinations as well as the BCS-defined κ, as shown in Table I. More recent measurements, using a fast pulse induction technique with thin foils, provide results in better agreement with those from the microspheres.[17]

The discrepancy is further complicated by severe disagreements between the microsphere reports themselves. This is shown in Fig. 1 for tin and indium ($\kappa\sim 0.1$), two of the materials most studied with this technique, where the closed (open) symbols of each figure refer to the effective κ_{sh} (κ_{sc}) from the references indicated, defined by

$$\sqrt{2}\kappa_{sh}(t)=h_{sh}^{-2}(t),$$

$$\sqrt{2}\kappa_{sc}(t)=h_{sc}(t), \qquad (1)$$

where $h_{sh}=H_{sh}/H_c$ is the reduced superheating field, $h_{sc}=\eta h_{c2}$ is the reduced supercooling field, $\eta(t=1)=1.695$, and $t=T/T_c$ is the reduced temperature. Although there is no complete theoretical description of the temperature behavior of the supercritical fields which spans the entire T-H phase space, the parameters, in principle, converge to $\kappa\equiv\kappa(1)$ provided the microspheres are sufficiently large to prevent size effects which arise when $\xi(t)$ becomes comparable to the dimensions of the sample.

Whereas the respective $\kappa_{sc}(t)$ are seen in generally good agreement, the $\kappa_{sh}(t)$ disagree significantly, corresponding to differences in H_{sh} of as much as 55 G in tin and 90 G in indium. The $\kappa_{sh}(t)$ manifests the parabolic behavior of $h_c(t)$; the near-linear behavior of the $\kappa_{sc}(t)$, at least above $t\sim 0.4$, is consistent with magnetization measurements,[3,18] with the different curvatures reflecting the difference in dynamics between the flux penetration and expulsion processes.

Equation (1) implies the possibility that the higher $\kappa_{sh}(t)$ of Ref. 5 were obtained with microspheres which failed to achieve a full metastability, i.e., which manifested lower superheating fields. This could result from the presence of material defects which effectively lower the transition field. The apparent convergence of the different $\kappa_{sh}(t)$, and $\kappa_{sh}(t)$ and $\kappa_{sc}(t)$ at $t=1$, despite significant differences at $t\ll 1$ might then be the result of the temperature dependences of both λ and ξ, which diverge as $t\rightarrow 1$. The higher $\kappa_{sh}(t)$ of Ref. 5 might also arise from diamagnetic interactions between the microspheres employed in the suspensions, absent in the single sphere measurements of Ref. 11, which would raise the local fields so that the transitions appear at the lower (applied) fields. Curiously, however, the more recent suspension results[14] are in agreement with those of the single sphere.

Nevertheless, it would appear that the discrepancy has been attributed to such extrinsic effects, and thusly disregarded: the determinations of κ have customarily proceeded by ignoring the $\kappa_{sh}(t)$ to extract κ from the $\kappa_{sc}(t)$ as $t\rightarrow 1$ on the basis of Eq. (1) alone, even though defects and diamagnetic interactions would also have impact on the h_{sc} measurements.

Although in evidence some 30 years ago, these discrepancies have never been addressed in the literature insofar as we are aware. They cast doubt on the determinations of κ in Ref. 5, hence also on the results of Refs. 6, 7, 9, 13, 15, and 16, many of which are the only measurements existing for the given material. They furthermore prevent the straightforward use of such measurements in elaborating the behavior of h_{sh} for $t\ll 1$, the precise temperature dependence of which remains a question of some theoretical interest.[19]

We here reexamine the analyses of the data and clarify at least a part of the problem. The data base and its reanalyses

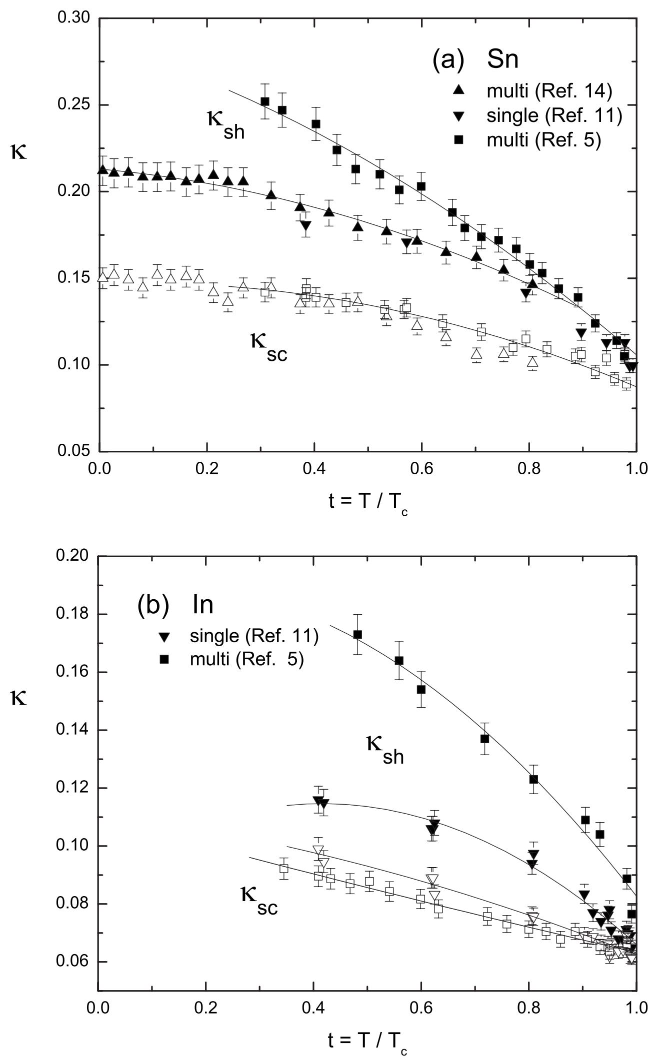

FIG. 1. (a) Compilation of $\kappa_{sh}(t)$ and $\kappa_{sc}(t)$ extracted from measurements of the supercritical fields in tin microspheres. (b) The same as (a) but for indium. Closed symbols represent superheating, open symbols represent supercooling, and lines represent polynomial fits to the respective data sets.

are described in Sec. II, with the results presented in Sec. III. These are discussed in Sec. IV, and conclusions presented in Sec. V. Generally, we find the discrepancy in tin to result from differing methods of analyses, whereas the discrepancy in indium is a consequence of significantly differing experimental results. Combination with the results from $\kappa_{sc}(t)$ in each case yields results for κ differing only slightly from their previously reported values, but with somewhat larger uncertainties. Although we find no simple explanation for the almost factor 2 difference between κ determinations of this technique and those of the thin films and/or foils, we observe that the latter are also obtained from $\kappa_{c2}=2^{-1/2}h_{c2}$ and that the difference with the microsphere results is nearer a factor $\sim$1.7, suggesting a possible misidentification of H_{c3} as H_{c2} in the previous analyses.

II. DATA BASE

The results by Feder and McLachlan (FM) for tin are obtained from supercritical field measurements on single spheres of diameters 8, 21, and 48 μm, over the temperature range $0.4<t<1$.[11] Both the 8 and 48 μm spheres exhibit strong size and defect effects, and were not considered. The FM report on indium is from measurements on single spheres of 8, 16, and 35 μm diameters, and a powder of 10–50 μm spheres in volume concentration of 17%; we omit all but the 35 μm measurements as a result of observed size and defect effects. The more recent tin results of Larrea *et al.*,[14] from suspensions of microspheres of diameter 33–40 μm at a volume filling factor of 25%, are in general agreement with those of FM over $0.005<t<0.81$. The errors in Fig. 1 represent 4% uncertainties, and are for reader convenience only: Larrea *et al.* report errors of roughly this level, and the reports of Smith, Baratoff, and Cardona (SBC) and FM are assumed comparable since no estimate is provided. There is also a report by Feder *et al.*[12] with suspensions of 1–5 μm tin spheres, which we, however, neglect since the estimated diameter above which size effects are unimportant is $\sim 7.5\xi(T)$, or $\sim$5 μm at $0.95T_c$.[11]

SBC reported results for tin from measurements on suspensions of 5–15 μm diameter spheres suspended to a volume filling factor of <5% over $0.3<t<1$, and for indium from measurements on 1–10 μm microspheres with a filling factor of <5% over the same temperature range.[5]

None of the previous reportings provide the actual supercritical field measurements. For spheres, the H_{sc} of Eq. (1) measured is actually H_{c3}, the critical field for surface nucleation of the superconductive state, which is favored over the bulk nucleation field H_{c2}.[1,2] Moreover, the local superheating field at the sphere equator is a demagnetization enhanced 3/2 larger than the laboratory field.[1,2] For FM, Larrea *et al.*, and $\kappa_{sc}(t)$ of SBC, the supercritical fields were regenerated from the reported $\kappa_{sh}(t)$ and $\kappa_{sc}(t)$ using

$$h_{sh}^{lab}(t) = \frac{2}{3}h_{sh}^{local}(t) = (0.561)\kappa_{sh}^{-1/2}(t),$$

TABLE I. Comparison of Ginzburg-Landau parameter determinations; "—" denotes no experimental measurement.

	Lead	Thallium	Tin	Indium	Cadmium	Aluminum
κ_{grains}	0.25	0.076	0.086	0.066	0.012	0.013
$\kappa_{film/\mathrm{foil}}$	0.34	—	0.15	0.13	—	—
κ_{BCS}	0.43	0.12	0.16	0.12	0.003	0.014

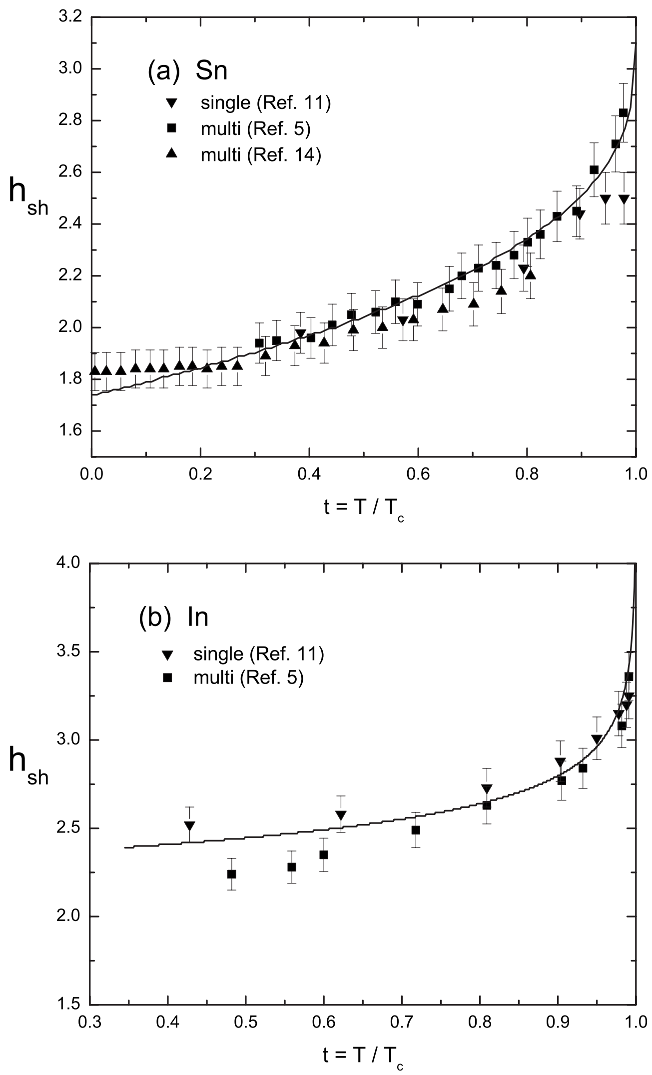

FIG. 2. (a) Comparison of the regenerated SBC $h_{sh}(t)$ in tin with those of FM and Larrea *et al.*, regenerated via Eq. (1). (b) The same as (a) but for indium. Lines are calculated from Eq. (3).

$$h_{sc}^{lab}(t) = \eta h_{c2}^{local} = (2.40)\kappa_{sc}(t), \tag{2}$$

where the prefactor in h_{sh} includes the sphere demagnetization. The factor $\eta(t=1)=1.695$ is valid only for $t\sim 1$ and, in principle, increases with decreasing temperature. A variational lower bound $\eta(t=0)=1.925$ has been obtained from a microscopic analysis of a pure superconductor assuming specular surface reflection,[20] but has not been used in the analyses since Refs. 5, 11, and 14 used only $\eta(1)$.

While the $\kappa_{sc}(t)$ were obtained from Eq. (2), the SBC $\kappa_{sh}(t)$ were obtained from a numerical integration of the Ginzburg-Landau equations following Ginzburg[21] with correction for the demagnetization of the sphere. To retrieve the $h_{sh}(t)$ of SBC, we best fit the upper curve of Fig. 1 of Ref. 5 over the interest range with a seventh order polynomial, and applied the inverse transformation to the full $\kappa_{sh}(t)$ of Fig. 8 in Ref. 5. This reproduces the results of Fig. 8 in Ref. 5 to within 3% over the $0.8 < t < 1$ as indicated in Fig. 2.

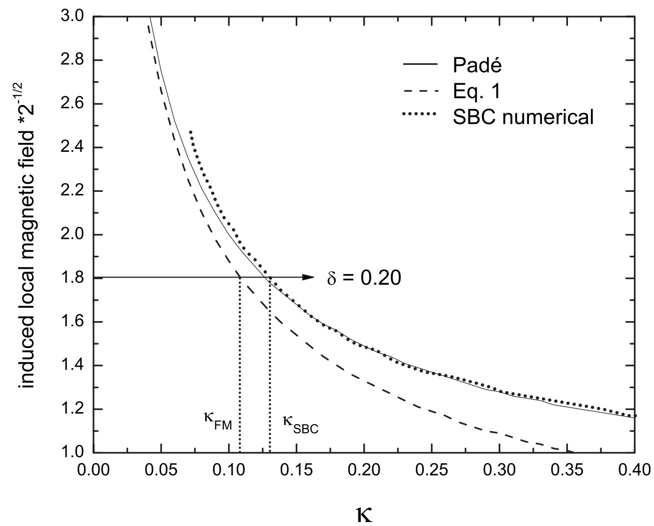

FIG. 3. The upper curve of SBC in Fig. 1 together with Eq. (2) and the Padé [2,2] approximant. Near the midrange of reduced superheating fields, the previous analysis techniques yield a difference δ of $\sim$20% in the determination of κ.

III. RESULTS

For tin, the $h_{sh}(t)$ of all groups, shown in Fig. 2(a), are in reasonable agreement, although the results of Larrea *et al.* appear flat below $t=0.2$ and there is a general tendency for the SBC results to be higher in the midrange temperatures. The rapid drop in h_{sh} below $t=1$ is consistent with nonlocal electrodynamics, the line indicating the predicted behavior in the extreme nonlocal limit,[22,5]

$$h_{sh}(t) = \sigma\chi^{1/4}\kappa^{-1/3}(1-t)^{-1/12}, \tag{3}$$

where $\sigma=1.35$ (1.42) for diffusive (specular reflective) phonon surface scattering, and $\chi\sim 1$. Equation (3) is, in principle, valid for $1 \gg 1-t \gg \kappa^2$, or about 20% of the available temperature range.

The discrepancy between the SBC and FM/Larrea *et al.* results for tin appears to arise solely from the different analysis techniques. This seems to be not the case for indium, as seen in Fig. 2(b). In this case, the SBC fields do not agree with those of FM, and are generally lower by $\sim$12%. The disagreement increases with decreasing temperature: for $t \sim 0.5$, it amounts to $\sim$60 G. For the same h_{sh}, the two analysis lead to a difference in κ_{sh} of order 20% for $h_{sh}=2.0$ as shown in Fig. 3, in agreement with the 16% stated in Refs. 5 and 11; the difference increases with decreasing h_{sh}, i.e., as $t\to 1$.

The analysis of FM and Larrea *et al.* is the more customary, but that of SBC would appear to be the more correct. FM have, however, argued[11] that the numerical method employed by SBC, which allows for only one-dimensional fluctuations of the order parameter, is too large for $\kappa\sim 0.1$; the SBC results therefore represent only an upper limit. Permitting more than one degree of freedom in the perturbation generally results in lower instability fields. On the other hand, Eq. (1) represents only the first term in a general expansion of κ_{sh}. Dolgert *et al.*[23] have reexamined the relation using the method of matched asymptotic expansion, generating an expression for κ_{sh} through fifth order. The stability of the results was analyzed with respect to both one- and two-dimensional perturbations, and the latter shown not to lead to any additional destabilizing effects in the low limit. For present purposes, it is sufficient to reanalyze the various $h_{sh}(t)$ with the Padé [2,2] approximant,[23] also shown in Fig. 3 and given by

$$h_{sh}=2^{-3/4}\kappa^{-1/2}\left[\frac{1+5.444\,781\kappa+4.218\,101\kappa^2}{1+4.781\,869\kappa+1.365\,523\kappa^2}\right]. \qquad (4)$$

This agrees to within 1% with more recent numerical calculations for $\kappa<1$, which appear to differ only slightly from the SBC curve above $\kappa\sim0.15$. As evident in Fig. 3, neither of the previous analysis appears in particularly good agreement with Padé in the region $\kappa\sim0.08$.

Figure 4(a) summarizes the combined results in tin using Eq. (4), together with the respective $\kappa_{sc}(t)$ determinations from Eq. (2). Here, 4% uncertainties are again shown except where measurements overlap, in which case the standard mean and deviation are given. The κ_{sh} of Refs. 11 and 14 are raised and coincide with those of Ref. 5. In particular, both $\kappa_{sh}(t)$ and $\kappa_{sc}(t)$ are seen not to converge well at $t=1$.

The results of a least squares fitting are given in Table II, together with the reported results of SBC and FM. The errors in this work are fitting errors only, using $\kappa(t)=m_1+m_2(1-t)^{\rho}$. As indicated, the $\kappa_{sh}(t)$ and $\kappa_{sc}(t)$ converge at $t=1$ to within 3σ, yielding $\kappa(\mathrm{Sn})=0.094\pm0.006$. Although the difference with the previously quoted results appears insignificant, it occurs at the level of $\sim4\sigma$.

A similar reanalysis of $\kappa_{sh}(t)$ for indium using Eq. (4) shows a continuing discrepancy below $t\sim0.9$. Although the SBC $\kappa_{sh}(t)$ is little affected in the reanalysis, those of FM are significantly increased above their values in Fig. 1. The implied larger superheating fields of the FM determinations suggest that the SBC suspension was in a mixed state and not fully superheated. Nonetheless, both $\kappa_{sh}(t)$ converge toward $\kappa_{sc}(t)$ at $t=1$, as seen in Fig. 4(b), with $\kappa(\mathrm{In})=0.064\pm0.008$ resulting from a $(1-t)^{\rho}$ fit. This is shown in Table II in comparison with the results of SBC and FM. The difference with previous determinations is well within error owing to the fitting uncertainty. The analysis is, however, strongly dependent on the data near $t=1$, with the larger uncertainty in the indium result reflecting both the lack of data below $t\sim0.4$ and measurement differences between the samples.

IV. DISCUSSION

The supercritical field determinations of all reports are obtained from hysteresis curves of the first order tin and indium transitions. Different reports have, however, assumed different definitions of the critical fields within the hysteresis curves, as shown schematically in Fig. 5 where "integrated events" means the cumulative sum of events at each field from the beginning of the up or down field ramp. As evident, these definitions alone can lead to an appreciable variation in the reported $\kappa(t)$. In general, the κ_{sh} and κ_{sc} parameters of FM should be consistently higher than those of SBC, which is not observed in the case of FM κ_{sh}.

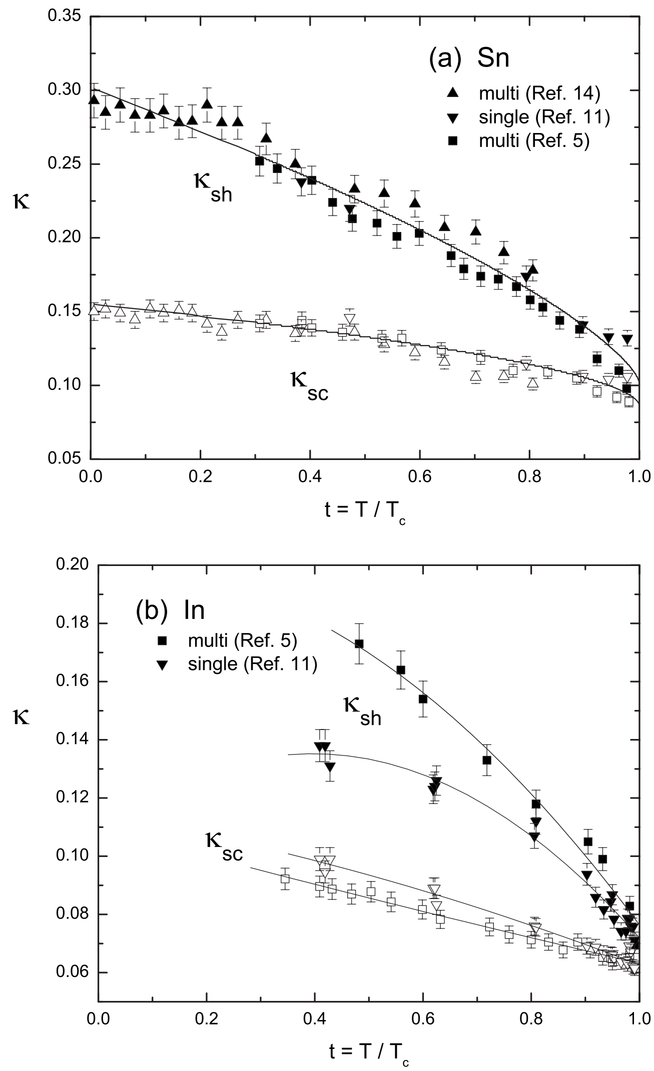

FIG. 4. (a) Reanalysis of the combined κ_{sh} and κ_{sc} in tin via Eqs. (4) and (2). A least squares fit of each yields the values for κ shown in Table II. (b) The same as (a) but for indium. The lower FM κ_{sh} results for indium suggest the grains of SBC not to have been fully metastable as a result of defect presence. Lines are polynomial fits to the respective data sets.

In the case of single-grain measurements, variations in the supercritical fields arise mainly from the grain metallurgy. FM performed careful investigations of defects as part of their study. The hysteresis curves were measured as a function of the orientation of the spheres in the applied field, and the κ_{sh} and κ_{sc} taken from the direction yielding the lowest values (i.e., highest h_{sh}, lowest h_{sc}). The results are strongly

TABLE II. Resume of κ determinations in Sn and In from supercritical field measurements of small grains.

	Tin	Indium
SBC	0.087±0.002	0.060±0.002
FM	0.093±0.001	0.062±0.001
This work (combined)	0.094±0.006	0.064±0.008

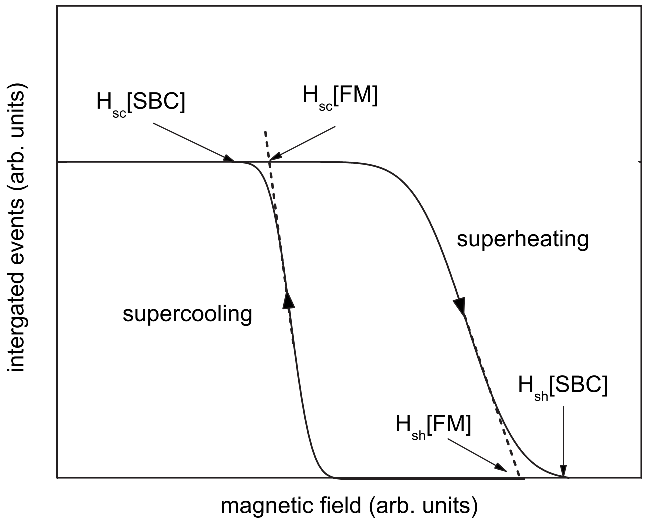

FIG. 5. Hysteresis curve schematic, indicating the various field definitions for h_{sh} and h_{sc} employed by SBC and FM.

dependent on T_c. In principle, for $T \sim T_c$, the coherence length is sufficiently large that these effects are negligible; at lower temperatures, the coherence length is smaller than the defect and the effects are more noticeable. For this reason, FM also employed $\kappa_R(t) \sim (h_{sc}/h_{sh})^{2/3}$ as a comparison for $\kappa_{sc}(t)$, which has the advantage of being insensitive to T_c since it is independent of H_c.

In the case of the suspension measurements, the deformation of the hysteresis from the rectangular single sphere cycle is due largely to the interplay between defects and diamagnetic interactions between the spheres.[24] At lower temperatures, defects in the sphere metallurgy may significantly reduce the superheating capacity of the material, resulting in a lowered transition field such as observed in the indium data of SBC. However, the defects should also serve as nucleation centers for "early" normal (N)$\rightarrow$superconducting (S) transitions at lower temperatures, which is not observed. On the other hand, the advantage of the multisphere measurement lies on the averaging over the defect contribution—the last sphere to transition to the normal state is the most defect-free sphere of the suspension.

The recent work of Peñaranda *et al.*[25] has demonstrated significant diamagnetic interactions even for filling factors as small as 5%. These may vary from one suspension to another of the same material as a result of inhomogeneous distributions. Although the last sphere to transition in the S$\rightarrow$N case is effectively free of diamagnetic interactions, this is not true for the N$\rightarrow$S where diamagnetic interactions are a maximum. As the field decrease continues, the new N$\rightarrow$S transitions enhance the local diamagnetic contribution to the local fields of those spheres still normal. This would suggest that the $\kappa_{sc}(t)$ determinations with suspensions are lower than actual by some amount proportional to the diamagnetic interactions, since the local field is higher. As seen in Fig. 1(a), the SBC $h_{sc}(t)$ for tin are essentially the same as those of the FM single sphere. For indium, however, the SBC $h_{sc}(t)$ are systematically lower than those of FM by ~10% for $t \leq 0.8$.

All results were moreover obtained by variation of the magnetic field, rather than temperature, which as shown by Chaddah and Roy[26] results in an enhanced $h_{sc}(t)$, apart from any diamagnetic interaction, as a result of the thermal fluctuations induced by the field changes themselves.

The variation of κ with temperature suggests a variation of the transition order with temperature, which has possibly important ramifications since κ is then less a fundamental property of the superconductor than a simple ratio between the two characteristic lengths in the description, both of which vary with temperature and yield results consistent with the observed κ determinations. Variation of the order of the transition with the temperature is predicted in recent renormalization-based reformulations of basic superconductive theory,[27] which include fluctuations in the involved gauge and scalar fields, and result in a dividing line between type-I and type-II behaviors at $\kappa = 0.8/\sqrt{2}$ with a magnetic response which can be varied between type I and type II simply by temperature change. This variation has been seen in magnetization measurement of nitrogen-doped Ta ($\kappa = 0.665$),[18] but is otherwise unconfirmed.

V. CONCLUSIONS

Reexamination of previous Ginzburg-Landau parameter determinations from superheating field measurements of microspheres of tin and indium shows the long-standing discrepancy in reported $\kappa_{sh}(t)$ of tin to derive from the different analysis methods employed, rather than whether single- or multigrain determinations, or grain metallurgy. Although the tin results of SBC can be made to agree with those of FM (and Larrea *et al.*), the customary analysis via Eq. (2) is in disagreement with that performed numerically. This is not the case for indium, where the discrepancy reflects severe differences in the supercritical field measurements themselves.

A similar reexamination of the supercooling field measurements uncovers no divergences capable of explaining the discrepancies with the measurements of Refs. 3 and 4. On the contrary, the variation of h_{sc} with t appears to confirm the latter, raising several questions as to the true significance of κ as a descriptor of superconductors. The resolution of these is, however, beyond the scope of this work.

The available $t < 1$ data are of insufficient precision to permit more than suggestions as to κ. Analysis of all superheating results via the Padé approximation yield a convergence of $\kappa_{sh}(t)$ and $\kappa_{sc}(t)$ to κ, with values for each material only slightly different from those previously reported on the basis of $h_{sc}(t)$ alone. These previous results, however, are likely larger than actual as a result of diamagnetic interactions between the superconducting sphere population. Apart from nonlocal effects, the large κ_{sh} for $t < 0.8$ is likely due to defect presence, which causes nucleation of the normal state to occur at fields below the maximal H_{sh}. These become unimportant as $t \rightarrow 1$, increasing h_{sh} and generating a rapid drop in κ_{sh}.

Whether the proper analysis is by Eq. (2), numerical, or Padé remains a question. Nevertheless, all of the materials analyzed in Ref. 5 (tin, indium, thallium, lead, and mercury) follow from the SBC in Fig. 1; in contrast, the analysis of the remaining references has proceeded via Eq. (2), which is seen to underestimate the respective $\kappa(t)$. In view of the significance of the Ginzburg-Landau parameter, the uncertainties inherent to this technique and the variation in experimentally obtained results, and technological advances over the last 30 years, careful remeasurements of the supercritical fields over the full temperature range and their analysis within a definitive description of the κ-H_{sh} plane would seem to be necessary.

The discrepancy in κ between spheres and thin film and/or foil determinations has been known for some decades but, to the best of our knowledge, remains unexplained. The thin film and/or foil results are, in fact, a factor $\sim$1.7 larger than those from the microspheres; they are also in better agreement with κ derived from h_{c3} via $\kappa(t) = (1.695\sqrt{2})^{-1}h_{c3}(t)$ and agree in general with the lower temperature results of the microspheres.

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