Characteristics of the Penetration Depth of Superconducting Indium Alloys

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The realtive penetration depths of a series of In-Sn, In-Tl, and In-Pb alloy specimens were measured at 700 kc/sec with and without a longitudinally applied dc magnetic field. In the dilute range the zero-field penetration depth showed a mean free path dependence roughly proportional to the one-half power of the resistivity, in accordance with previous work, but for samples with resistivities exceeding approximately 1.3 $\mu\Omega$ cm the effective penetration depth became anomalously large, indicating filamentary behavior. Measurements of the magnetic field dependence failed to show the decrease in penetration depth with applied field found by experimenters working at microwave frequencies; instead there was reasonable agreement with the predictions of the Ginzburg-Landau theory. The experimentally estimated values of the parameter κ at which filamentary characteristics appeared were of the order of 0.8 ± 0.05 as compared to the theoretically derived critical value of $1/\sqrt{2}$.

INTRODUCTION

RECENTLY, there has been cause for interest in the various properties of superconducting alloys, both in regions where they closely resemble elemental superconductors in their properties, and where they behave in rather unique ways. This paper considers some of the characteristics of a group of indium solid solutions, concentrating particularly on the effective penetration depth of a magnetic field into the alloy.

The initial work in this area was conducted by Pippard,[1] who studied the effect of the addition of indium to tin on the microwave surface impedance of the material, and concluded from his results that there was a pronounced mean free path effect (subsequently confirmed by Chambers[2] in studies at 10 Mc/sec). This, in turn, led to the concept of a coherence length and pointed the way to the more sophisticated theories of the present. Theoretical support for the mean free path dependence of the penetration depth came from Miller,[3] who derived expressions for the shift in λ as a function of the ratio of coherence length to the mean free path in terms of the BCS formulation.[4] Similar calculations were made independently by Abrikosov and Gor'kov.[5,6]

Pippard also pioneered in measuring a field dependence of the penetration depth of tin.[7] Work in this area did not proceed rapidly because the effect was small, the technique difficult, and the theory inadequate. It was not until 1958, eight years after the initial experiments, that Spiewak[8] extended the technique to examine the effect at 1 kMc/sec in some detail on tin single crystals. The results in this case were extremely complex and indicated a structure which was unexpected in the general picture of superconductivity. Attempts have been made by Dresselhaus[9] to account for some of these observations in terms of galvanomagnetic effects occurring for the normal electrons in the superconducting state, but the total situation remains unclear.

Sharvin and Gantmakher[10] recently completed some very sensitive measurements on pure tin at much lower frequencies (2 Mc/sec). Their results once again show a small and relatively uncomplicated field dependence of the penetration depth, agreeing rather closely with what would be predicted from the Ginzburg-Landau[11] theory. This rather wide discrepancy in the experimental results was taken to indicate that low-frequency measurements were more characteristic of the static properties of superconductors, whereas the observed microwave effects may arise from nonequilibrium phenomena requiring something more than a simple thermodynamic analysis, This is the approach most recently taken by Richards.[12] In a series of measurements at 3 kMc/sec on single-crystal Sn and dilute Sn-In alloys, he found fluctuations fully as complicated as those observed by Spiewak. In the interpretation of his data, however, he felt that the Dresselhaus model could not adequately explain certain rather basic features, and that perhaps some sort of resonance phenomena in the penetration layer must be invoked.

The one thing on which all researchers agree is the need for extending the lower range of frequencies in which measurements are made, and to more thoroughly map out the part played by the addition of impurities.

The present work was conceived with the idea of establishing the composition dependence of the pene-

[1] A. B. Pippard, Proc. Roy. Soc. (London) **A216**, 547 (1953.)
[2] R. G. Chambers, Proc. Cambridge Phil. Soc. **52**, (1956).
[3] P. B. Miller, Phys. Rev. **113**, 1209 (1959).
[4] J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. **108**, 1175 (1957). (Referred to as BCS.)
[5] A. A. Abrikosov and L. P. Gor'kov, J. Exptl. Theoret. Phys. (U.S.S.R.) **35**, 1558 (1958) [translation: Soviet Phys.—JETP **8**, 1090 (1959)].
[6] A. A. Abrikosov and L. P. Gor'kov, J. Exptl. Theoret. Phys. (U.S.S.R.) **36**, 319 (1959) [translation: Soviet Phys.—JETP **9**, 220 (1959)].
[7] A. B. Pippard, Proc. Roy. Soc. (London) **A203**, 210 (1950).
[8] M. Spiewak, Phys. Rev. Letters **1**, 136 (1958); Phys. Rev. **113**, 1479 (1959).
[9] G. Dresselhaus and M. Spiewak Dresselhaus, Phys. Rev. **118**, 77 (1960); Phys. Rev. Letters **4**, 401 (1960).
[10] Y. V. Sharvin and V. F. Gantmakher, J. Exptl. Theoret. Phys. (U.S.S.R.) **39**, 1242 (1960) [translation: Soviet Phys.—JETP **12**, 866 (1961)].
[11] V. L. Ginzburg and L. D. Landau, J. Exptl. Theoret. Phys. (U.S.S.R.) **20**, 1064 (1950). (Referred to as GL.)
[12] P. L. Richards, Phys. Rev. **126**, 912 (1962).

tration depth over a relatively broad range for a number of solutes in a given solvent. The operating frequency was such that it might essentially be considered as a measure of the static case, thus establishing a lower limit when compared to microwave experiments. Simultaneously, it was possible to examine the field dependence of the penetration depth of a comparatively large variety of alloys and to explore relationships between them.

EXPERIMENTAL TECHNIQUE

The specimens on which the measurements were made were extruded wires slightly more than 1 mm in diameter and 2 in. long. The materials were mixed, vacuum cast, quenched, extruded, and annealed for several days just below their melting point. Then, about 0.002 in. of surface was removed by electrolytic polishing. All alloys used were dilute substitutional solid solutions, with concentrations well-removed from any known phase boundary.

The specimens now serve as the core of the inductance of a superconducting *LC* circuit, in the manner suggested by Chambers[2] and Schawlow and Devlin.[13] A duplicate empty tank circuit was mounted adjacent to the one containing the specimen to serve as a reference in compensating for extraneous temperature effects, and the two were enclosed in a vacuum-tight copper can filled with 5 mm He exchange gas.

The resonant frequencies of these circuits were in the vicinity of 700 kc/sec, and the difference frequency between the two with no specimen present remained constant to within one cycle in the temperature range from 4.2 to 2.5°K. The unloaded frequencies also remained constant to within 1 part in 10^6 upon the application of an external magnetic field up to 400 Oe.

These tanks were used both as passive networks for transmission measurements of the type performed by Chambers,[2] and more actively to provide the reference frequency in a Franklin oscillator, following the technique employed by Shawlow and Devlin.[13] The first method was employed primarily as a means for checking the Q of the circuit, in the event there were normal metal regions present (see Chiou, Connell, and Seraphim[14]). Since most of the information desired could be deduced from shifts in resonant frequency, the second technique was used for the bulk of the measurements because of the speed and convenience afforded by this method.

The measuring rf field was of the order of 0.1 Oe peak-to-peak for the transmission measurements, and 0.5 Oe for the oscillator measurements. Conventional techniques were employed for controlling temperatures to within 0.001°K during a measurement,[15] and absolute temperatures were determined to ±0.002°K from the He vapor pressure.

Magnetic fields, uniform and stable to 1 part in 10^4, were provided by a Garrett-type coil[16] driven by a current-regulated supply. The field coil was calibrated using a NMR probe. Extraneous transverse fields were compensated for by a pair of Helmholz coils. Measurements of the shifts in resonant frequency of the tank circuit containing the specimen were made with both temperature and applied magnetic field.

Frequency shifts arising from changes in circuit geometry were appropriately compensated for by reference to corresponding shifts in the empty coil oscillator. Deviations from run to run were much more difficult to account for or control, so that all sequential measurements were taken in a single run. Measurements of skin depth in the normal state were ruled out because the consequent decrease in Q required a change in feedback from the oscillator with corresponding effects on frequency. Further, the determination in Q made it impossible to resolve the normal state resonant frequency with sufficient accuracy to be meaningful. This, coupled with frequency variations encountered from run to run and the fact that at 700 kc/sec, $\delta_{\text{normal}}\sim 10\lambda$, ruled out absolute measurement of penetration depths with reference to the normal metal skin depths.

Information concerning relative penetration depths can nevertheless be extracted from the frequency data. Primary deviations in frequency in going from one specimen to another arose from differences in radii and only secondarily to other causes, such as capacitor fluctuations, major temperature and pressure effects having been minimized by the maximum amount of mechanical and electrical isolation, and the remainder approximately compensated for by reference to the empty coil oscillator.

We assume that one can write $L_i=K\pi(r_0^2-r_i^2)$, where r_0 is the effective empty coil radius, and r_i is the shielding radius of the ith specimen. The resonant frequency can be expressed as

$$\omega_i=1/(L_iC)^{1/2}.$$

If we now define a "Gorter-Casimir" penetration depth $\lambda=\lambda_0 z(t)$, where

$$z(t)=[1-(T/T_c)^4]^{-1/2},$$

and then

$$\frac{d\omega_i}{dz}=K\pi C r_i\frac{dr_i}{dz}\frac{1}{(L_iC)^{3/2}}.$$

Furthermore,

$$r_i=r_0^{(i)}+\lambda_0^{(i)}z;\quad dr_i/dz=\lambda_0^{(i)},$$

and

$$\frac{d\omega_i}{dz}=K\pi C r_i\lambda_0^{(i)}\frac{1}{(L_iC)^{3/2}}.$$

[13] A. L. Schawlow and G. E. Devlin, Phys. Rev. **113**, 120 (1959).

[14] C. Chiou, R. A. Connell, and D. P. Seraphim, Phys. Rev. **129**, 1070 (1963).

[15] H. S. Sommers, Rev. Sci. Instr. **25**, 793 (1954).

[16] M. W. Garrett, J. Appl. Phys. **22**, 1091 (1951).

TABLE I. Summary of results on alloy penetration depths.

Sample	$(\rho_{res}/\rho_{id})\times 10^2$	T_c(°K)	$[\lambda_0(\text{alloy})/\lambda_0(\text{In})]$	α_{exp}	β_{exp}	κ_{eff}	α_{theor}	β_{theor}
In-I[a]			1.10±0.05					
In-II		3.404	1.00±0.05	0.08±0.04	0	0.16	0.02	~0
In-III			0.95±0.05	0.07±0.03	0.02±0.02			
In-Sn								
0.23 at.%	0.97	3.395	1.25±0.05	0.05±0.03	0	0.24	0.03	~0
0.66 at.%	2.84	3.411	1.50±0.05	0.16±0.08[b]	0.04±0.02[b]	0.21	0.03	~0
1.48 at.%	6.35	3.484	1.65±0.05	0.07±0.02	0.01±0.01	0.46	0.05	0.02
1.96 at.%	8.42	3.53	1.85±0.05	0.06±0.03	0.02±0.01	0.66	0.07	0.05
3.28 at.%	14.2	3.71	2.00±0.05	0.07±0.03	0.02±0.01	0.67	0.07	0.05
In-Tl								
1.4 at.%	3.4	3.375	1.60±0.05	0.06±0.04	0.03±0.02	0.39	0.05	0.01
3.5 at.%	8.4	3.287	1.90±0.05	...	...	...	...	
5.0 at.%	12.0	3.272	2.10±0.05	0.04±0.02	0.02±0.01	0.67	0.07	0.05
6.2 at.%	14.9	3.268	2.35±0.05	0.05±0.01	0.03±0.01	0.85	0.08	...
7.4 at.%[a]	17.8	3.26	4.25±0.05	0.11±0.03	0.03±0.01	2.71	0.11	...
11.4 at.%[a]	27.4	3.26	4.10±0.05	...	...			
11.4 at.%[a]	27.4	3.26	4.00±0.05	...	...			
In-Pb								
1.03 at.%	6.7	3.409	1.30±0.05	0.12±0.07[b]	0.08±0.02[b]	0.33	0.042	0.01
1.38 at.%	9.0	3.43	1.60±0.05	0.05±0.02	0.01±0.01	0.38	0.047	0.01
2.03 at.%	13.2	3.47	1.65±0.05	0.05±0.03	0.02±0.01	0.35	0.044	0.01
3.60 at.%[a]	23.2	3.60	2.45±0.05	0.11±0.04	0.04±0.01	1.34	0.092	...
Sn		3.720	1.10±0.05	0.04±0.02	0	0.16	0.02	~0
Sn-In								
1.0 at.%	5.8	3.653	1.70±0.05	0.05±0.02	0.01±0.01	0.25	0.03	~0
3.0 at.%[a]	17.4	3.632	4.00	...	...	...	...	...

[a] Exhibited nonideal behavior.
[b] Indications of trapped flux.

Taking a ratio, one then has

$$\frac{(d\omega_2/dz)}{(d\omega_1/dz)}=\frac{r_2}{r_1}\left(\frac{L_1}{L_2}\right)^{3/2}\frac{\lambda_0{}^{(2)}}{\lambda_0{}^{(1)}}=K'\frac{\lambda_0{}^{(2)}}{\lambda_0{}^{(1)}}.$$

The ratio of the slope of the frequency for an alloy specimen, plotted as a function of z, can thus be related to that of the pure base metal, giving

$$\left(\frac{d\nu(\text{alloy})/dz}{d\nu(\text{pure})/dz}\right)=K'\frac{\lambda_0(\text{alloy})}{\lambda_0(\text{pure})}\quad\text{as}\quad T\rightarrow T_c.$$

In the cases with which we are dealing, $r_2\simeq r_1$, and $L_2\simeq L_1$ within the accuracy with which the slope $d\nu/dz$ could be established, i.e., within 5%. Furthermore, by confining our attention primarily to the temperature range close to T_c, other systematic errors in the determination of $d\nu/dz$ tend to vanish, leaving as the major uncertainty the accuracy with which T_c could be determined experimentally.

The values of T_c used in this work were determined both from ac data taken *in situ* during a run (i.e., the temperature at which the circuit commenced to resonate during cool-down, and extrapolation of critical field measurements where the external field sufficient to quench oscillations was noted), and from extensive dc resistance and susceptibility measurements on samples prepared from the same melts. Where discrepancies arose (never exceeding 10 mdeg), best values were chosen in terms of minimum curvature in the plot of ν vs $z(T/T_c)$.

EXPERIMENTAL RESULTS

The findings from measurements on a series of indium-alloy specimens and two tin specimens, which were run for comparison, are compiled in Table I, which gives the alloy composition, critical temperature T_c, and the residual resistivity ratio ρ_{res}/ρ_{id}. Also listed are the field coefficients α and β as defined by the expansion

$$\lambda(H,T)=\lambda(0,T)[1+\alpha(H/H_c)^2+\beta(H/H_c)^4\cdots]. \quad (1)$$

Figure 1 illustrates the measured change in frequency with temperature as noted for an indium sample containing 3.65% Pb. The shift in frequency is closely linear in $z(t)$ over an unexpectedly wide range in temperature. This is of particular interest since it is in the class of rich alloy solutions which, in many other respects, depart rather drastically from the behavior of more dilute solutions. Samples of this particular composition, for instance, showed blurred and broadened temperature and field transitions, along with other characteristics typical of nonideal superconductors. Apparently the functional dependence of the macroscopically averaged penetration depth on temperature in zero-magnetic field is not materially affected.

This is not true for some other aspects of field penetration in alloy superconductors, as can be seen in Fig. 2. Here is plotted the ratio of the alloy penetration depth to that of high-purity indium ($\rho_{res}/\rho_{id}<2\times 10^{-4}$) as a function of the half-power of residual resistivity ratio normalized to T_c. This manner of presenting the data is suggested from the theoretical relations derived by Miller,[3] who predicted, assuming $l/\lambda_L(T)\ll 1$, a dependence of the form

$$\lambda(0)=\frac{\lambda_L(0)(\xi_0/l)^{1/2}}{[1-(4l/\pi^2\xi_0)ln(\pi\xi_0/l)]^{1/2}} \quad \text{as} \quad T\rightarrow 0. \quad (2)$$

Under these conditions the denominator will be slowly varying, and corrections arising from it would be beyond the accuracy of the experimental data. Using also the BCS relation $\xi_0=0.18\hbar v_0/kT_c$, we have

$$\lambda(0)\propto(\rho_{\text{res}}/T_c)^{1/2}.$$

Despite the fact that the conditions for validity of this expression are not generally met, i.e., $l>2\times 10^{-6}$ cm [the value for the London penetration depth $\lambda_L(0)$ given by Dheer[17]], the present results are not inconsistent with a dependence of this form.

Two things must be pointed out concerning the plots shown in Fig. 2. The first concerns the apparent depression of the points for all the lead-doped specimens below those of the tin- and thallium-doped samples. It may be that the lead-doped specimens do not electropolish with complete uniformity, although no signs of structure could be seen optically. Alternately, the annealing time for these specimens may have been insufficient, although dc measurements indicated the specimens were quite homogeneous. If inhomogeneities exist, they must involve only a small fraction of the active surface area. Magnetic transitions are sharp for all the Pb-doped samples, except the specimen containing 3.65 at.% Pb, and the general temperature and field dependence of the resonant frequencies of these alloys resemble that of the other indium alloys. Consequently, it is probably *equally* valid to assume a real difference with solute, despite the absence of theoretical arguments in support of this view.

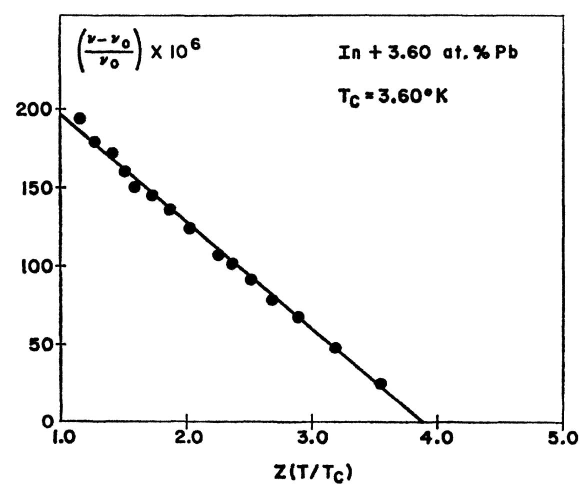

FIG. 1. Dependence of the oscillator resonant frequency on the Gorter-Casimir function $z(T/T_c)=[1-(T/T_c)^4]^{-1/2}$.

[17] P. N. Dheer, Proc. Soc. (London) **A260**, 333 (1961).

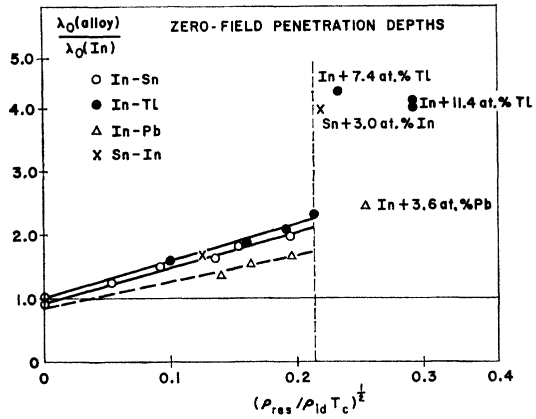

FIG. 2. The relative penetration depth of a group of indium-base alloys as a function of the square root of the reduced resistivity, normalized to T_c. Three tin-base samples are also included for purposes of comparison.

The most striking feature of Fig. 2 is the definite appearance of nonideal or filamentary superconductivity when the solute concentrations exceed certain values. For the thallium system this breakoff point was found to be rather abrupt and dramatic, occurring between 6.2 at.% Tl and 7.4 at.% Tl. At that point the apparent penetration depth shows a sharp increase, accompanied by a simultaneous deterioration in other superconducting characteristics, such as magnetic transition width, indicating the onset of filamentary effects. Similar behavior was observed in samples containing 11.4% Tl and 3.6% Pb. The jump in magnitude for the penetration depth was not so great for the latter, but its other properties were no less nonideal. The fact that the measured value was low by comparison with Tl indicates, not too surprisingly, that filamentary characteristics may indeed be more sensitive to the solute material than are the "ideal" superconducting properties of more dilute solutions. Whether or not one has a critical *resistivity*, as was suggested by Chiou *et al.*, or a critical value of λ (alloy) is not resolved. The Ginzburg-Landau theory gives the criterion that if

$$\kappa=\sqrt{2}e_{\text{eff}}H_0\lambda^2/\hbar c>1/\sqrt{2},$$

the interphase surface energy becomes negative, and one has Abrikosov's "superconductor of the second kind."[18] It was from this relationship that Gor'kov[19] derived an expression for the critical resistivity, assuming a Pippard-type dependence of λ on mean free path. Certainly, a more comprehensive investigation would be needed to resolve this rather fine point. Measurements on In-Ga, where T_c rises almost immediately, would provide a contrast to the In-Tl system, where T_c is depressed out to appreciable concentrations (see Table I).

[18] A. A. Abrikosov, J. Exptl. Theoret. Phys. (U.S.S.R.) **32**, 1442 (1957) [translation: Soviet Phys.—JETP **5**, 1174 (1957)].
[19] L. P. Gor'kov, J. Exptl. Theoret. Phys. (U.S.S.R.) **37**, 1407 (1959) [translation: Soviet Phys.—JETP **10**, 998 (1960)].

The results for a pure polycrystalline tin sample and two Sn-In samples are included for comparison, although perhaps it is not quite legitimate to present them on the same plot. The first point to be noted is that the ratio of the penetration depths for tin-to-indium is not out of line with currently accepted values for their respective penetration depths,[17,20] i.e., $\lambda(\mathrm{Sn})/\lambda(\mathrm{In})\simeq 500$ A/430 A$=1.1$. Secondly, the Sn+3.0 at.% In was very emphatically nonideal, especially at lower reduced temperatures. It tends to support the idea of a critical resistivity (or penetration depth) in agreement with the results on the indium-base samples. It may be, however, that the annealing time of three weeks at 220°C was not adequate for complete homogeneity, as compared with indium-base alloys which show substantially higher diffusion rates.

Figures 3 and 4 concern themselves with the magnetic field dependence of the penetration depth for representative samples. Here the frequency shift produced by the application of a dc magnetic field, $H_{\mathrm{appl}}<H_c$, is reduced to a shift in the penetration depth, i.e.,

$$[\lambda(H,T)-\lambda(0,T)]/\lambda(0,T) = [\nu(H,T)-\nu(0,T)][d\nu(0,T)/dz]^{-1}.$$

Following the analysis of Sharvin and Gantmakher,[10] when the applied field is the sum of constant and measuring fields, i.e.,

$$H=H_{\mathrm{appl}}+H_1\sin\omega t,\quad H_1<H_{\mathrm{appl}}, \tag{3}$$

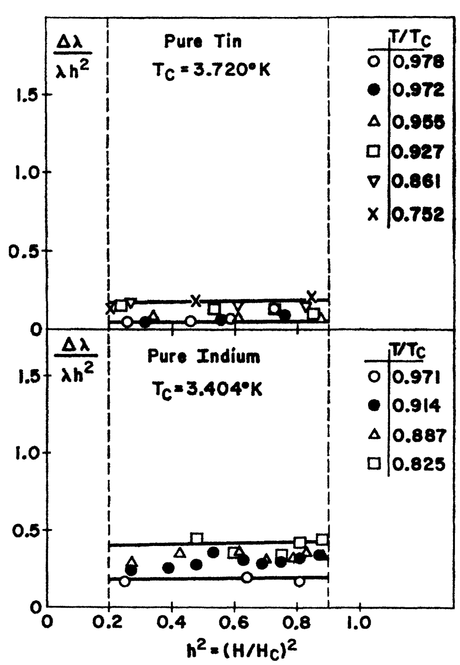

FIG. 3. The field dependence of the penetration depth of high-purity tin and indium, displayed as $[\lambda(H)-\lambda(0)]/[\lambda(0)(H/H_c)^2]$ vs $(H/H_c)^2$.

[20] A. B. Pippard, Proc. Roy. Soc. (London) **A203**, 98 (1950).

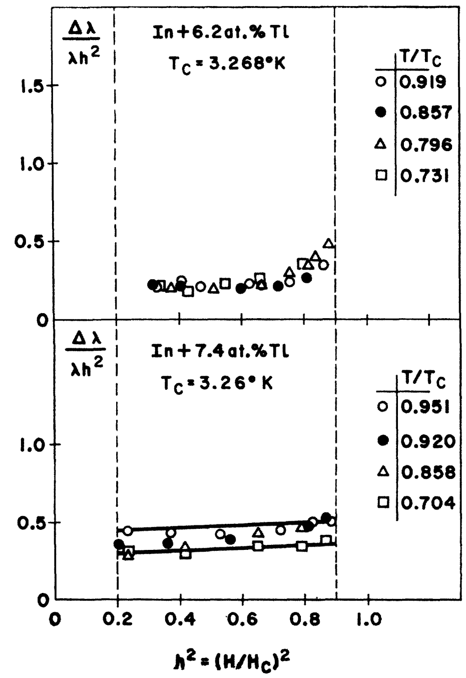

FIG. 4. The field dependence of the penetration depth of In-Tl alloy samples below and above the critical limit indicated in Fig. 2.

the flux linking the surface layer per unit length is

$$\Phi=H(0,T)[(1+\alpha(H/H_c)^2+\beta(H/H_c)^4]. \tag{4}$$

Substituting (3) in (4) and considering only the terms oscillating at a frequency ω and dependent on H_{appl}, we have

$$\Phi_\omega=\lambda(0,T)\{[3\alpha+(15/2)\beta h_1^2]h^2+5\beta h^4\}H_1\sin\omega t, \tag{5}$$

where $h\equiv H_{\mathrm{appl}}/H_c$; $h_1=H_1/H_c$. Assuming $H_{\mathrm{appl}}\gg H_1$, the effective change in the penetration depth, determined from the ac field, is

$$\Delta_{\mathrm{eff}}\lambda/\lambda(0,T)\simeq 3\alpha h^2+5\beta h^4. \tag{6}$$

Thus, if we plot $\Delta\lambda/\lambda(0)h^2$ vs h^2, the resulting curve should have a value of 3α at its vertical intercept and a constant slope of 5β.

Figure 3 compares the results for polycrystalline-tin and polycrystalline-indium. In both cases the quartic term can be seen to be vanishingly small. The quadratic dependence is also relatively small but well defined. There is also a slight shift towards larger values of α at lower temperatures. For the purposes of comparison of field coefficients for different alloys, we shall overlook this temperature dependence and examine only the major variation in going from sample to sample, and return later to the finer details.

Figure 4 shows similar plots for In-Tl specimens immediately above and below the "breakaway point" in Fig. 2. The upturn seen for $h>0.7$ in the case of the In+6.2% Tl is very likely due to the initial appearance of a very small "toe" on the magnetic transition rather than higher-order terms in the field dependence of the superconductor itself. Data for the In+7.4% Tl are remarkably well behaved, considering the fact that in other respects they act in typical nonideal manner, and the effective zero-field penetration depth is anomalously large (see Fig. 2). The field coefficients are also appreciably larger in this case.

Experimental values for α and β taken from plots such as these are included in Table I, along with effective values for the Ginzburg-Landau κ, and some theoretically estimated values for the field coefficients.

The method by which this was accomplished was to accept the value of κ given by Gor'kov[21] and Faber[22] for pure indium, i.e., $\kappa_{\mathrm{In}}=0.16$ as $T\rightarrow T_c$. Then, according to GL,

$$\alpha=\kappa(\kappa+2\sqrt{2})/8(\kappa+\sqrt{2})^2, \tag{7}$$

and

$$\beta\simeq 4.7\times 10^{-2}\kappa^2(1-5.1\kappa+7\kappa^2), \tag{8}$$

for $\kappa\ll 1$ and $\beta\ll\alpha$, from which we obtain $\alpha_{\mathrm{In}}=0.02$ and $\beta\sim 0$.

One can also deduce a constant of proportionality

$$\kappa_{\mathrm{eff}}(\mathrm{alloy})\simeq 9.2\times 10^{-3}H_c\left(\frac{\lambda(\mathrm{alloy})}{\lambda_0(\mathrm{In})}\right)^2 \quad \text{for} \quad T\sim T_c.$$

Thus, knowing the relative penetration depth and critical field one can calculate the GL predicted values of α and β. For some of the richer alloys the calculated values of β were relatively large, and since the formula given above is invalid for such circumstances, these numbers are not included in the table.

It is to be noted that the agreement between the measured and calculated values of α and β are, with certain exceptions, better than either the data or the theoretical treatment might lead one to expect. Of the alloys, only those particular specimens which exhibited irreproducibility and history dependence, indicative of flux trapping, gave experimental values of α_{exp} out of range of the predicted α_{theor}. The In+0.66 at.% Sn specimen so indicated had a zero-field penetration depth somewhat above what might have been anticipated, while for In+1.03 at.% Pb it was somewhat low, although in neither case were the deviations particularly large. This would indicate that in zero-applied field the associated normal surface area is either extremely small or well removed from a sensitive region in the measuring coil. Only with the application of an appreciable external field does the trapped-flux region commence to play a role.

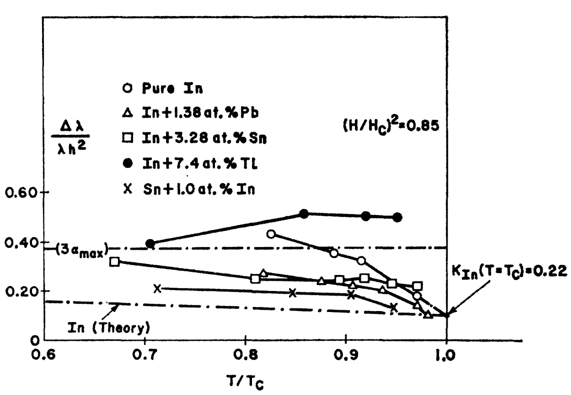

FIG. 5. Temperature dependence of the shift in penetration depth with magnetic field for a fixed value of the reduced field H/H_c.

From these data it would also appear that the In+6.2 at.% Tl is definitely a borderline case falling between ideal and filamentary regions. Here κ_{eff} exceeds the GL limit of $1/\sqrt{2}$. Correspondingly, it gives indication of early field break up, as previously discussed concerning the plot in Fig. 4. It essentially sets the lower limit for break up quite closely.

Let us now address the problem of the temperature dependence of the GL coefficients. Figure 5 shows the variation of $\Delta\lambda/\lambda h^2$ with temperature for various representative alloys in a fixed value of magnetic field. Before discussing the meaning of this plot, it is well to consider the theoretical situation. According to GL,

$$\kappa(t)=\mathrm{const}H_c(t)\lambda^2(t).$$

Assuming

$$H_c\simeq H_0(1-t^2)$$

and

$$\lambda(t)\simeq\lambda_0/(1-t^4)^{1/2},$$

$$\kappa(t)\propto \mathrm{const}H_0\lambda_0^2/(1+t^2),$$

and κ would double in going from $T=T_c$ to $T=0$. The theoretical estimate given by Gor'kov[21] is $\kappa(T\rightarrow T_c)=0.22$. Recalling the theoretical expressions for α and β as a function of κ, Eqs. (7) and (8), we see that these quantities will also increase as the temperature is lowered.

Returning to Fig. 5, we see that qualitatively the more dilute alloys and the pure metals do show this trend, exemplified in the plot by pure indium, In+1.38 at.% Pb, and Sn+1.0 at.% In. The In+3.28 at.% Sn appears to be a borderline case, as are the In+5.0 at.% Tl and 6.2 at.% Tl samples. In these cases, $\Delta\lambda/\lambda h^2$ is essentially temperature independent, while for richest alloys, such as In+7.4 at.% Tl, the shift is in the opposite direction. The theoretical expression for $\alpha(\kappa)$ is in qualitative agreement with the observed decrease in temperature sensitivity for increasing solute content, via the resultant increase in $\kappa(T\rightarrow T_c)$ with alloying, and the decrease in $\Delta\alpha/\Delta\kappa$ with increasing κ. Quantitative comparisons do not fare so well. If we assume that $\Delta\lambda/\lambda h^2=3\alpha(t)+5\beta(t)h^2$ we see that we can derive a theoretical curve, assuming a value for $\kappa(T\rightarrow T_c)$. In Fig. 5 a curve for pure In is plotted,

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using Gor'kov's value for κ at T_c. It is immediately apparent that theory and experiment are in reasonable agreement at $T=T_c$ but the predicted temperature variation is far too small. Furthermore, the experimental values of $\Delta\lambda/\lambda h^2$ exceed the theoretical limit $\alpha_{\max}\equiv\lim_{\kappa\to\infty}\alpha(\kappa)=\frac{1}{8}$. Just what fraction of the latter discrepancy can be accounted for by including contributions from the β term awaits an adequate expression for this quantity in the limit of large κ.

DISCUSSION

In reviewing the evidence brought forth in this investigation, certain points merit some further comment Taken as a whole, the most striking feature is that the results, by and large, are in closer correspondence to the theoretical predictions of Abrikosov and Gor'kov, along with the measurements of Sharvin and Gantmakher, than with the high-frequency measurements of Spiewak *et al.* At *no* time was there any indication of a *decrease* in penetration depth upon application of a magnetic field, as reported by Pippard, Spiewak, and Richards. The general shape of the curves in Fig. 5 are indeed similar to those displayed by Richards for the temperature dependence of the surface reactance of a series of Sn-In alloys. In fact, a corresponding flattening out of the curves with increased solute content can be seen in both cases. Close examination at temperatures as high as 0.97 T_c fail to reveal any indication of a sign change in the field dependence, so that if it occurs at all at these lower frequencies, it must do so only at temperatures extremely close to the transition temperature. It is true that Richards and Spiewak both worked with single-crystal specimens, whereas the present data were taken from polycrystals. In view of the fact that the sign reversal was observed in the microwave measurements for all major crystallographic axes of Sn, however, the crystallinity would not seem to be sufficient reason for the absence of this effect in the present case. Even if the shift were strain sensitive, the fact that the tin sample (the one with which the most direct comparisons can be made) consisted essentially of only a few large crystallites would tend to minimize such an effect. Pure indium is generally stress-relieved by plastic flow[23] but, unfortunately, data on the field dependence of indium at higher frequencies are lacking. Altogether, these results support the contention, already expressed by other authors,[12,24] that at sufficiently low frequencies ($\nu\ll10^9$ pcs) the relatively simple quasi-static interpretations are at least qualitatively correct, whereas some other mechanism must be invoked at higher frequencies.

The other major point to be noted is the way in which the transition from ideal to filamentary superconductivity shows up in the present series of measurements, i.e., the rather dramatic increase in the "zero-field" penetration depth (for complete accuracy, one must remember the presence of the rf measuring field), generally coupled with a corresponding change in the sensitivity to applied magnetic fields. Judging from these results, one either has a critical resistivity of the order of 1.3 $\mu\Omega$ cm or a critical penetration depth $\lambda_0(0)$ of the order of 1000 A [assuming Dheer's[17] value of 430 A for $\lambda_0(0)$ of pure indium].

The over-all correlation of ideal and nonideal behavior with experimental estimates of the GL parameter κ either less or greater than the theoretically derived critical value of $1/\sqrt{2}$ was most encouraging. The present results would indicate that $\kappa_{\text{critical}}\sim0.8$ but unfortunately there is no well-defined criterion for ideal or nonideal behavior and hence the boundary is somewhat arbitrary. The fact that the shift of λ with magnetic field may *exceed* the allowable theoretical limit for the rich alloys is most probably indicative of the mixed structure in superconductors of this type.

Whether there exist differences in penetration depth for alloys of the same resistivity but with different solute material remains a problem. Unfortunately, the present work is not extensive enough to answer that question properly, and further measurements with these and other alloys must be made for the matter to be truly clarified.

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