

## Electron Thermal Conductivity of Superconducting Films of Indium-Gadolinium and Lead-Gadolinium Alloys\*†

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(Received 2 January 1973)

We have measured the thermal conductivity of quench-condensed superconducting lead and indium films containing small amounts of gadolinium. The data are in good agreement with the theory of Ambegaokar and Griffin and of Bennemann (modified for strong-coupling effects), except at the lowest temperatures, where problems in the interpretation of the data cause deviations. We have found that strong-coupling effects do not entirely account for the discrepancy between weak-coupling theory and the data of Bjerkaas *et al.* for the thermal conductivity of quench-condensed films of indium containing small amounts of manganese. Thus, our results support evidence from tunneling and high-frequency electromagnetic experiments that the properties of superconductors with rare-earth impurities are understood, but those with transition-element impurities are not.

### I. INTRODUCTION

Small quantities of a paramagnetic impurity dissolved in a superconductor can sharply depress its superconducting transition temperature.<sup>1</sup> A theory of this effect which is in good agreement with many experimental observations has been given by Abrikosov and Gor'kov.<sup>2</sup> Several theories have been built on this foundation to calculate various properties of superconductors containing paramagnetic impurities.<sup>3-5</sup> Two of these theories, by Ambegaokar and Griffin<sup>4</sup> and by Bennemann,<sup>5</sup> predict the temperature dependence of the electron thermal conductivity as a function of impurity concentration. These two theories are equivalent. The accuracy of the predicted function has been tested by several experimental studies.

Cappelletti and Finnemore<sup>6</sup> studied  $\text{Th}_{1-x}\text{Gd}_x$ , and Williams *et al.*<sup>7</sup> studied  $\text{La}_{98}\text{Lu}_{2-x}\text{Tb}_x$ . In those experiments, which were performed on bulk samples, the measured values of the thermal conductivity included that contributed by the phonons. In order to test the theory, the phonon contribution had to be estimated and subtracted from the measured thermal conductivity; this procedure inevitably introduced significant uncertainties into the desired results. The anisotropy of the energy gap which one frequently finds in bulk samples (including thorium<sup>8</sup>) also affects the temperature dependence of the thermal conductivity, introducing additional uncertainty into the interpretation of the data. The results obtained by these workers were found to be in reasonable agreement with theoretical predictions when these corrections were made.

Bjerkaas *et al.*<sup>9</sup> measured the thermal conductivity of thin  $\text{In}_{1-x}\text{Mn}_x$  films which were formed by condensation onto a substrate held at liquid-helium temperature to promote sample homogeneity and uniformity.<sup>10</sup> The films were not warmed up until all the thermal-conductance measurements had

been completed. Because the phonon mean free path in such films was very short, the phonon contribution to the thermal conductivity<sup>11-13</sup> was negligible, and because the electron mean free path was much shorter than the ideal coherence length, the energy gap in these films was presumably isotropic.<sup>14</sup> Some of the uncertainties in the interpretation of measurements on bulk samples were therefore avoided.

Although Mn had previously been shown to exhibit a localized magnetic moment in quench-condensed In films (so the theory<sup>4,5</sup> should have been valid), Bjerkaas *et al.* found a systematic deviation from the theoretical prediction (see Fig. 1) which had not been seen in the results of the previous workers.<sup>6,7</sup> A possible explanation for this deviation was that strong-coupling effects were not included in the theory; these effects were not understood then, but they are now.<sup>15</sup> In Sec. II we adjust the theoretical curves to include strong-coupling effects. We find that their inclusion does not entirely remove the disagreement between the data of Bjerkaas *et al.* and the theoretical predictions.

Other properties of superconductors with transition-element impurities also disagree with present theories. This has been shown by measurements of electron tunneling<sup>16</sup> and of high-frequency electromagnetic absorption.<sup>17</sup> However, rare-earth impurities have an effect on these other properties which is in reasonable agreement with theory.<sup>16,17</sup> We have therefore measured the thermal conductivity of quench-condensed lead films and indium films containing the rare-earth element gadolinium, expecting that the results would probably agree with the theory.

### II. THEORY

The temperature dependence of the reduced thermal conductivity  $k = K_{es}/K_{en}$ , where  $K_{es}$  and  $K_{en}$  are the electron contribution to the thermal con-

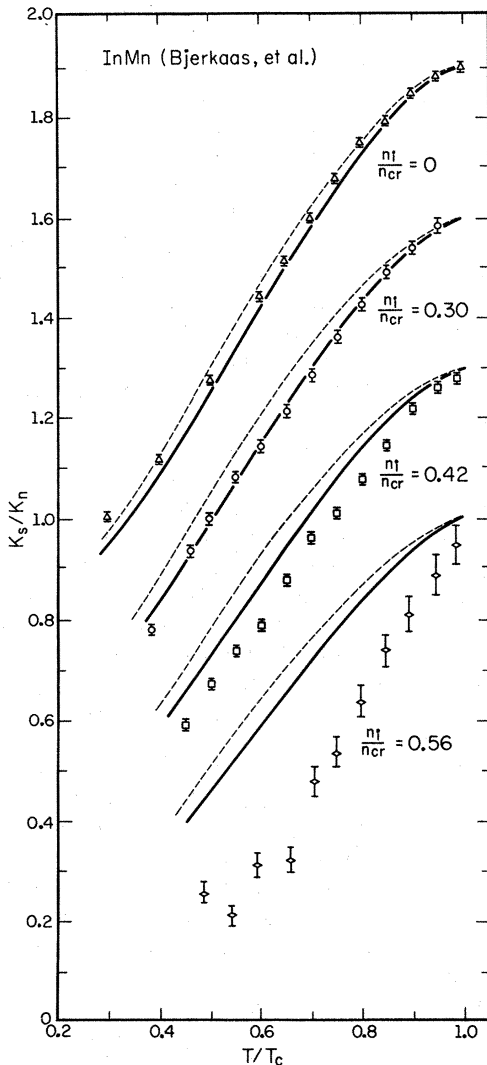


FIG. 1. Experimental values of the reduced thermal conductivity as a function of the reduced temperature obtained by Bjerkaas *et al.*<sup>9</sup> on quench-condensed films of In-Mn alloys. The dashed lines show the theoretical values<sup>4,5</sup> for a weak-coupling superconductor, and the solid lines show the values obtained when the theory is modified by strong-coupling corrections. The zero has been shifted up by 0.3 between each set of data to avoid overlap. The indicated impurity concentrations are those determined from the measured transition temperatures, with strong-coupling correction.

ductivity  $K_s$  and  $K_n$  in the superconducting and normal states, respectively, has been studied previously for pure, strong-coupling superconductors with a short electron mean free path.<sup>15</sup> It was shown that the strong-coupling nature of the electron-phonon interaction affects the results only by increasing the size of the energy-gap parameter  $\Delta$  at the gap edge. We assume that this is also the case for superconductors containing paramagnetic impurities, so the theory<sup>4,5</sup> can be used to deter-

mine the reduced thermal conductivity for strong-coupling superconducting alloys by simply scaling the value of the energy-gap parameter in each alloy by the appropriate amount. Rather than numerically solving the rather complicated expression obtained by Ambegaokar and Griffin,<sup>4</sup> a simple, graphical method of transforming their curves can be used to find the reduced thermal conductivity with strong-coupling corrections.

Since the reduced thermal conductivity is a function of  $\Delta(T)/k_B T$ , where  $k_B$  is the Boltzmann constant and  $T$  is the temperature, the strong-coupling value for the reduced thermal conductivity  $k_s(T)$  can be related to the weak-coupling value  $k_w(T)$  by

$$k_s(T) = k_w(T'), \quad (1)$$

where the temperatures  $T$  and  $T'$  are related by

$$\Delta(T)/k_B T = \Delta_w(T')/k_B T' \quad (2)$$

and  $\Delta_w$  is the weak-coupling value of the energy-gap parameter. For any value of  $T$ , Eq. (2) can be easily solved for  $T'$  by a graphical technique.<sup>18</sup> To do this, we have used the theoretical curves<sup>4</sup> which show  $\Delta(T)/\Delta_p(0)$  as a function of the reduced temperature  $t = T/T_c$  and the reduced impurity concentration  $n_i/n_{cr}$ . Here  $\Delta_p(0)$  is the zero-temperature value of the energy-gap parameter in the pure material,  $T_c$  is the transition temperature,  $n_i$  is the impurity-atom concentration, and  $n_{cr}$  is the critical impurity-atom concentration, which is required to reduce  $T_c$  to 0.

For the lead alloy films, this type of analysis is straightforward, because neither  $T_c$  nor the reduced gap  $2\Delta(0)/k_B T_c$  depend significantly on the structure of the film; our quench-condensed films of lead have transition temperatures very close to 7.189 K<sup>19</sup> and reduced energy gaps<sup>15,20</sup> of 4.3–4.5. However, Bergmann<sup>21,22</sup> has shown that electron scattering caused by disorder in indium films increases both the reduced energy gap and  $T_c$ , indicating an increase in the electron-phonon coupling.<sup>23</sup> He has related these effects to the resistivity ratio  $\rho_n/\rho_{273}$ , where  $\rho_n$  is the residual resistivity of the film and  $\rho_{273}$  is 8.2  $\mu\Omega$  cm, which he takes to be the electrical resistivity of bulk indium at 273 K. Figure 2 shows Bergmann's data<sup>21</sup> for pure indium films condensed at various temperatures as a function of  $\ln(\rho_n/\rho_{273})$ . Also shown in Fig. 2 are data for higher resistivity ratios, which have been calculated from other measurements.<sup>9,24</sup> Figure 3 shows a similar plot of Bergmann's data for the reduced energy gap in pure indium films, determined by electron tunneling measurements. In Fig. 3, we also show a value obtained by Bjerkaas *et al.* from thermal-conductivity measurements performed on a pure, quench-condensed film of indium.<sup>9</sup> Evidently the two methods of determining the energy gap are in reasonable agreement.

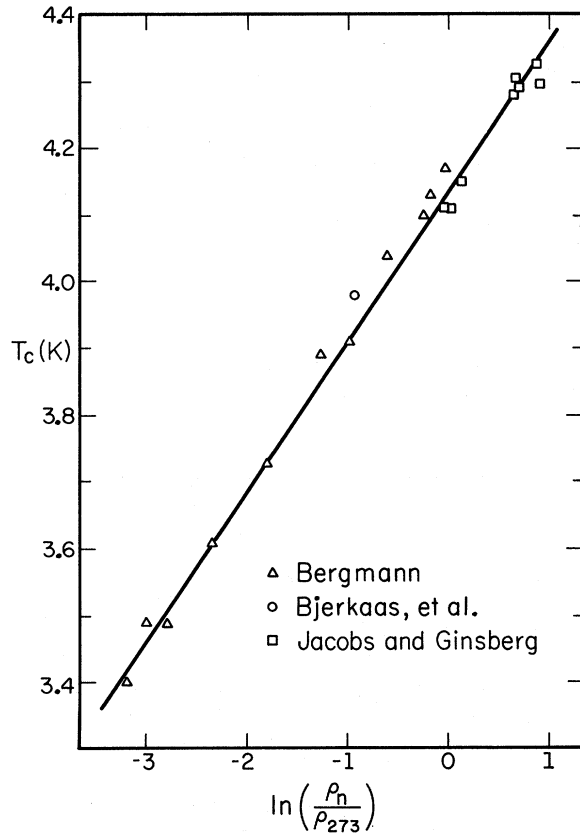


FIG. 2. Transition temperature of quench-condensed pure-indium films as a function of the reduced electrical resistivity.

In order to compare the data of Bjerkaas *et al.* with theory, the theoretical curves were adjusted for strong-coupling effects as follows. From the experimental values of  $K_n(T)/T$ ,  $\rho_n$  was determined from the relation  $\rho_n = LT/K_n$ , where  $L$  is the Lorenz number. We then used the ratio  $\rho_n/\rho_{273}$  with Figs. 2 and 3 to determine the transition temperature  $T_{cp}$  and the reduced energy gap  $2\Delta_p(0)/k_B T_{cp}$  which would be expected for a film of pure indium with the same resistivity ratio. Using the measured value of  $T_c$ , we then calculated  $T_c/T_{cp}$  and the corresponding<sup>2</sup> value of  $n_i/n_{cr}$ . (This value of  $n_i/n_{cr}$  was actually affected only slightly by the strong-coupling correction to  $T_{cp}$ .) From the ratio  $n_i/n_{cr}$ , we determined the theoretical<sup>4</sup> values of  $\Delta(T)/\Delta_p(0)$ . We could then employ the procedure described above to modify the theoretical curves for  $K_{es}/K_{en}$ . In our samples,  $K_{es} = K_s$  and  $K_{en} = K_n$ , because the phonons conduct much less heat than the electrons, as mentioned above. These modified curves are shown in Fig. 1. It is clear that the corrections for strong coupling have improved the agreement between theory and experiment somewhat, but there are still increasing deviations for increasing impurity concentration.

### III. EXPERIMENTAL TECHNIQUES

We made alloys of about 3.1 at. % Gd in Pb and 0.90 at. % Gd in In by placing appropriate quantities of each metal (99.999% pure In and Pb and 99.9% pure Gd) in a covered tantalum crucible which had been previously outgassed at 1300 K, and which had a thermocouple attached to it. The crucible was then placed in a bell jar which was pumped down to a pressure of about  $5 \times 10^{-6}$  Torr, and the metals were heated inductively to a temperature of about 1000 K for 20 min to outgas them. We then admitted helium gas into the bell jar and heated the metals to a temperature of about 1300 K. The induction furnace power supply was then turned off, and helium gas was immediately blown past the crucible to rapidly cool the sample, which solidified within about 50 sec. The resulting ingot was then repeatedly rolled and folded to ensure its homogeneity. Less concentrated alloys were made by mechanically mixing pieces of these alloys with either pure lead or pure indium by repeated rolling and folding. Chemical analysis determined the concentration of these alloys.<sup>19</sup>

To ensure that the films were homogenous, we used a conveyor belt<sup>9</sup> for flash evaporation. About 50 pellets were cut from the ingot and loaded onto this conveyor belt when the cryostat was assembled. To form the sample film later, we moved this conveyor belt so that it dropped pellets one at a time into a resistively heated tungsten-filament boat. After each pellet was dropped into it, the filament was quickly heated to a temperature of about 1800 K to thoroughly evaporate the pellet. The filament was then cooled so that another pellet could be dropped into it without popping out. We chose tungsten as the filament material because hot tungsten reduces oxides.<sup>25</sup> The cryostat's background pressure was less than  $10^{-6}$  Torr

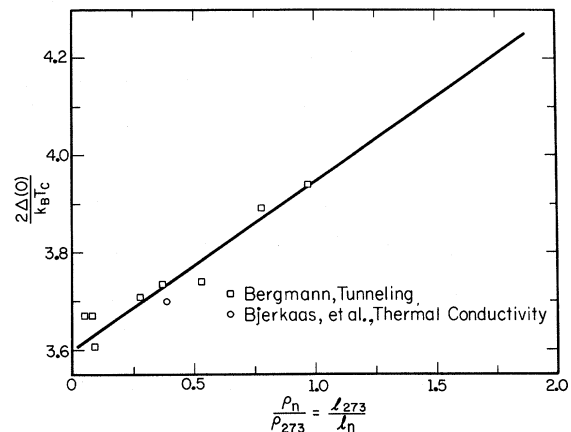


FIG. 3. Reduced energy gap of quench-condensed pure-indium films as a function of the reduced electrical resistivity.

during the production of the film. The evaporation of all the pellets for each sample required about 2 min.

The equipment and procedure used to measure the thermal conductivity in the present investigation were nearly identical to those used previously.<sup>9,15</sup> As in the previous investigation,<sup>15</sup> we first measured the thermal conductance of the circular Kapton-H film substrate,<sup>26</sup> 0.005 cm thick, which formed the bottom of a sample-holder tank. This sample holder was then cooled to about 1.1 K by liquid helium in another tank, and helium gas was condensed in the space above the substrate so the evaporation of the film onto the substrate did not raise the substrate temperature above a temperature of about 3 K. We then pumped the helium gas out of the sample holder, heated the substrate to 9 K, and pumped on it for 10 h to remove adsorbed helium from the substrate. We then re-measured the thermal conductance and determined the thermal conductance of the film by subtraction. To find the thermal conductivity of the film, its conductance was divided by its thickness. No cracking of these films occurred when the cryostat was warmed up to room temperature, so the film thicknesses could be determined by multiple-beam interferometry, as described previously.<sup>9</sup>

In this project, helium was condensed into the sample holder rather than being transferred directly into it, as was done previously. In this way, the pressure difference across the substrate could be held below 50 Torr, so the film was stressed much less. (This procedure also allowed one of the sample-holder inlet tubes to be removed, and this conserved liquid helium.) In addition, we

evaporated a circular area of silver onto the center of the substrate at the same time as we produced the silver isotherms. Resistance leads were then soldered directly to this silver area. This modification allowed us to make a much more accurate determination of the film's electrical resistance than previously. As before, we had helium gas at a pressure of about 50 Torr in the sample holder during the resistance measurements.

#### IV. RESULTS

Some of the data are listed in Table I. Two transition temperatures are indicated for each Pb-Gd alloy;  $T_c$  is the transition temperature implied by the rapid drop of the thermal conductivity just below  $T_c$  (which could thereby be determined to within about 0.04 K), and  $T_R$  is the transition temperature determined by measuring the film's electrical resistance. (For the In-Gd films, these transition temperatures were identical.) For each Pb-Gd alloy film, the difference between these two values of the transition temperature may have resulted from film-thickness variations. Schwidtal has observed a dependence of transition temperature on film thickness for Pb-Gd films.<sup>25</sup> The resistive transition temperature should then occur at the highest temperature at which a continuous superconducting path across the film exists, whereas the decrease in the thermal conductivity should more nearly indicate an average transition temperature of the film. (Incidentally, we made some alloy films which were considerably thicker than the ones reported here. They were unsatisfactory, because they had broad resistive transitions, up to about 1 K wide.) Our data show that the normal-

TABLE I. Chemically determined gadolinium or manganese concentration, film thickness, transition temperature  $T_c$  determined by thermal conductivity, transition temperature  $T_R$  determined by electrical resistance,<sup>a</sup> resistive transition width,<sup>b</sup> resistivity ratio defined in Sec. II, <sup>c</sup> reduced relative impurity concentration determined from the transition-temperature data, and calculated values of the reduced energy gap and the transition temperature for a pure film with the same resistivity.

Sample	$n_i$ (at. %)	$D$ (Å)	$T_c$ (K)	$T_R$ (K)	$\Delta T$ (mK)	$\rho_n/\rho_{273}$	$n_i/n_{cr}$	$2\Delta_P(0)/k_B T_\phi$	$T_\phi$ (K)
PbGd 1	1.02	1800 ± 50	5.30	5.86	45	1.47	0.36	4.29	7.189
PbGd 2	1.46	620 ± 50	3.35	3.63	65	1.46	0.69	4.29	7.189
InGd 1	0.16	2400 ± 60	3.512	3.512	80	1.17	0.23	3.97	4.18
InGd 2	0.29	1390 ± 50	3.263	3.263	45	1.58	0.32	4.05	4.25
InGd 3	0.40	1180 ± 50	2.028	2.028	23	1.60	0.67	4.05	4.25
InGd 4	0.61	1720 ± 40	...	1.236	44	0.85	0.85	3.89	4.14
InMn 1	0.00	2515 ± 30	3.98	3.980	47.5	0.38	0.00	3.75	3.98
InMn 2	0.0160	1150 ± 42	3.198	3.198	33.5	0.55	0.30	3.80	4.05
InMn 3	0.0256	1423 ± 48	2.829	2.829	42.0	0.65	0.42	3.83	4.08
InMn 4	0.0315	877 ± 49	2.366	2.366	37.0	0.69	0.56	3.84	4.10

<sup>a</sup> $T_R$  is defined as the temperature at which the film's resistance is 50% of its low-temperature normal-state value.

<sup>b</sup> $\Delta T$  is defined as the width of the temperature region in which the film's resistance drops from 90 to 10% of its residual resistance as the film is cooled.

<sup>c</sup>For the Pb-Gd and In-Mn films,  $\rho_n$  was not measured; it was calculated from the measured thermal conductivity and the Wiedemann-Franz law.

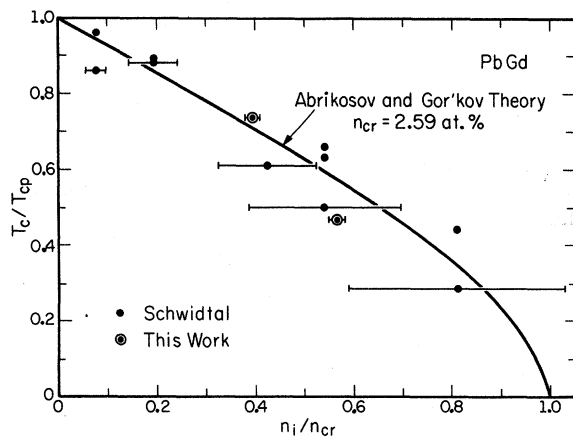


FIG. 4. Experimental and theoretical (Ref. 2) reduced transition temperature as a function of the reduced impurity concentration for the Pb-Gd films. The indicated impurity concentrations are those determined by chemical analysis of the ingots from which the films were made.

state thermal conductivity in the temperature range from  $T_c$  to  $T_R$  is proportional to  $T$  as expected, however, so we believe that the postulated nonuniformity of the film thickness had very little effect on the thermal conductivity below  $T_c$ . Each of the films in this investigation had a thermal conductivity which was proportional to  $T$  above  $T_c$ <sup>19</sup>; this indicates that phonon conduction was negligible, as desired.

We display in Figs. 4 and 5 the ratio  $T_c/T_{cp}$  ( $T_{cp}$  is defined in Sec. II) as a function of the impurity concentration (determined by chemical analysis) for the two alloy systems, along with the theoretical curves. To obtain the best fit, a value  $n_{cr}$

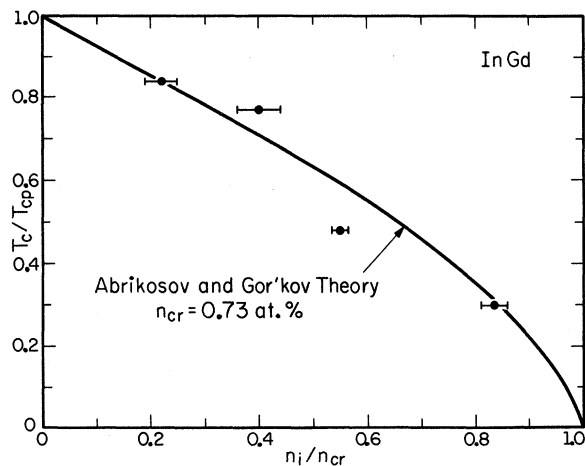


FIG. 5. Experimental and theoretical (Ref. 2) reduced transition temperature as a function of the reduced impurity concentration for the In-Gd films. The indicated impurity concentrations are those determined by chemical analysis of the ingots from which the films were made.

= 2.59 at. % was used for the Pb-Gd data, and a value  $n_{cr} = 0.73$  at. % was used for the In-Gd data. The scatter in the Pb-Gd data is thought to result partly from the apparent dependence of the transition temperature on the film thickness,<sup>25</sup> and scatter in the In-Gd data is thought to result largely from inexact chemical analysis of the samples. (Very little ingot material was available for analysis of these samples.<sup>19</sup>)

We observe no obvious deviation in Figs. 4 and 5 of the type predicted to occur if the impurity spins order,<sup>27,28</sup> so the theory of Ambegaokar and Griffin<sup>4</sup> and of Bennemann<sup>5</sup> may be applicable. For the reasons just mentioned for the scatter in Figs. 4 and 5, we have assumed the theory of Abrikosov and Gor'kov<sup>2</sup> to be valid, and we have calculated the value of the reduced impurity concentrations  $n_i/n_{cr}$  from the measured transition temperatures. We use these values, which are shown in Table I, throughout the rest of this discussion.

Figures 6 and 7 show the reduced thermal-conductivity data of the two alloy systems, along with

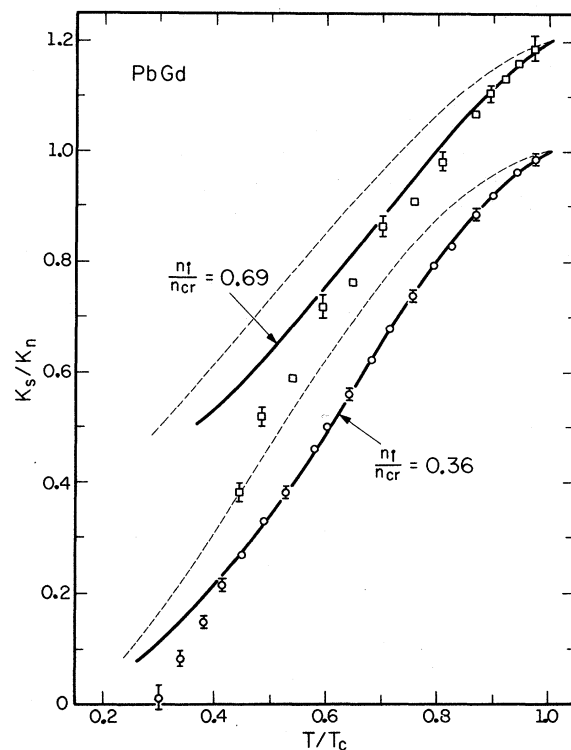


FIG. 6. Reduced thermal conductivity as a function of the reduced temperature for the two Pb-Gd films. The dashed lines show the theoretical values<sup>4,5</sup> for a weak-coupling superconductor, and the solid lines show the values obtained when the theory is modified by strong-coupling corrections for a reduced gap appropriate for pure lead, 4.3. The zero has been shifted up by 0.2 for the higher-concentration alloy to avoid overlap. The indicated impurity concentrations are those determined from the measured transition temperatures.

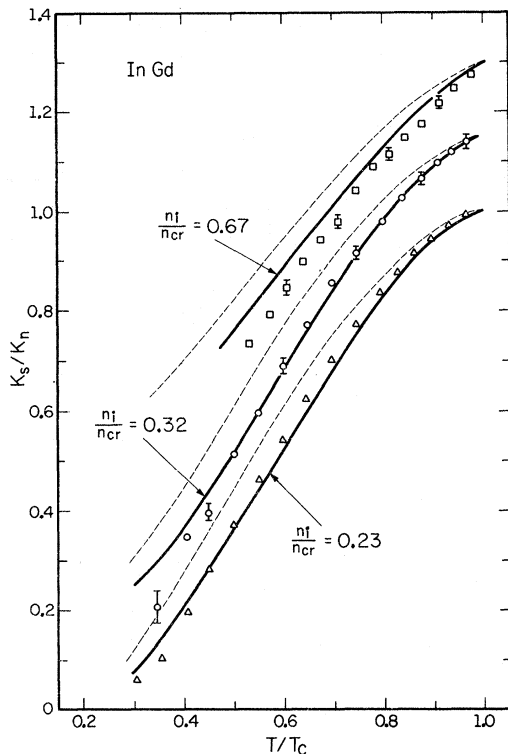


FIG. 7. Reduced thermal conductivity as a function of the reduced temperature for the three In-Gd films. The dashed lines show the theoretical values (Refs. 4 and 5) for a weak-coupling superconductor, and the solid lines show the values obtained when the theory is modified by strong-coupling corrections corresponding to the reduced energy-gap values shown in Table I. The error bars for the bottom set of data cannot be shown because they are approximately the size of the triangular symbols. The zero has been shifted up by 0.15 between each set of data to avoid overlap. The indicated impurity concentrations are those determined from the measured transition temperatures, with strong-coupling correction.

the theoretical curves, which were calculated by applying the strong-coupling corrections discussed in Sec. II. The theoretical curves<sup>4</sup> for a weak-coupling superconductor are also shown. For both alloy systems, the experimental values are in good agreement with the strong-coupling theory except below 2 K, where the measured thermal conductivity is significantly smaller than theory predicts. In fact, the thermal conductivity of the Pb-Gd films appeared to be negative at the lowest temperatures (data not shown). Obviously, experimental difficulties must have caused the low-temperature deviation.

This deviation is understandable if we assume that the substrate itself had a smaller thermal conductance after the sample film was on it than before. This decrease in the substrate's thermal conductance may have resulted from two factors.

First, the phonons in the substrate may be more diffusely reflected from the metal film than from the bare substrate surface. Second, the metal film may increase the rate at which reflections of phonons from the substrate's surface convert longitudinal phonons into transverse phonons, which may be less effective in transporting heat in amorphous polymers.<sup>29</sup>

According to experimental data of Zeller and Pohl,<sup>30</sup> the phonon mean free path in amorphous dielectrics is approximately proportional to  $1/T^2$  between 1 and 2 K. (See their Fig. 9.) If the H-film substrate is assumed to have a phonon mean free path in the same range as those found by Zeller and Pohl, then it could be 25% of the H-film's thickness at 1 K. If the metallic film changes the mean free path of the phonons striking it by only 5%, we would then expect this to decrease the thermal conductance of the H film by about 1.3%. This would explain the observed deviations. They are approximately proportional to  $1/T^2$ , as we would expect.

If one accepts our explanation of the low-temperature deviations of our data, then there would be deviations of this type in the data of Bjerkaas *et al.* for In-Mn alloys, but they would be considerably smaller because their sample films had larger thermal conductances than our films had.

## V. CONCLUSIONS

It has been shown that strong-coupling corrections do not remove the disagreement between the theory and the thermal-conductivity data of Bjerkaas *et al.* on films of quench-condensed In-Mn alloys.<sup>4</sup> On the other hand, our measurements of the thermal conductivity of films of Pb-Gd and In-Gd alloys are in good agreement with theory (except at lowest temperatures, where difficulties of interpretation cause deviations). These results support experimental evidence from other types of experiments<sup>16,17</sup> that calculations based on the theory of Abrikosov and Gor'kov<sup>2</sup> accurately describe the properties of superconductors which contain rare-earth impurities, but not those with transition-element impurities. Clearly, we need a better theoretical understanding of the effect of transition-element impurities on superconductors. Chaba and Nagi<sup>31</sup> have been successful in explaining data on electron tunneling into Pb-Mn films in terms of a strong interaction between the conduction electrons and the magnetic impurity atoms. (This interaction would be weaker for rare-earth impurities than for transition-element impurities, because the  $4f$ -electron shells are buried within the rare-earth atoms.) Perhaps further theoretical work along the same lines will explain the thermal conductivity of superconductors which contain transition-element impurities.

## ACKNOWLEDGMENTS

We are grateful to David Princehouse for valuable help in devising the method that we used to

make the alloy ingots. We also profited from a discussion with William Reese about the low-temperature thermal conductivity of polymers.

\*Research supported in part by the National Science Foundation under Grant Nos. GP-28996 and GH-33634.

<sup>†</sup>Paper based in part on the Ph.D. thesis of B. J. Mrstik, University of Illinois, 1973.

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## Upper Critical Field and the Density of States in Amorphous Strong-Coupling Superconductors

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(Received 24 July 1972)

The upper critical field of the amorphous superconductors  $\text{Bi}_{0.85}\text{Tl}_{0.15}$ , Ga,  $\text{Sn}_{0.86}\text{Cu}_{0.14}$ , and  $\text{Pb}_{0.75}\text{Bi}_{0.25}$  is measured in the temperature range from 1.5°K to the zero-field transition temperature. Films of the amorphous metals about 1500 Å thick are obtained by quenched condensation onto a substrate at He temperature. The initial slopes of the  $B_{c2}(T)$  curves are used to determine the electronic density of states at the Fermi surface. For all the investigated materials these values were enhanced compared with the density of states which one obtains from the free electron model. For Ga,  $\text{Sn}_{0.86}\text{Cu}_{0.14}$ , and  $\text{Pb}_{0.75}\text{Bi}_{0.25}$  the experimental enhancement factor agrees well with the electron-phonon enhancement factor  $1 + \lambda$ , where  $\lambda$  is taken from superconducting-tunneling experiments. The temperature dependence of  $B_{c2}$  deviates from that predicted by Werthamer, Helfand, and Hohenberg, showing  $B_{c2}$  values too large in the low-temperature region. It is suggested that the strong-coupling behavior of the amorphous superconductors is responsible for the deviation at low temperature.

### I. INTRODUCTION

A number of metals can be obtained in the amorphous state by quenched condensation onto a substrate at He temperature. This was first done by Buckel and Hilsch.<sup>1</sup> The residual resistivity of

these amorphous metals is very similar to the resistivity of the corresponding liquid metals. Buckel<sup>2</sup> and Fujime<sup>3,4</sup> examined the amorphous metals by electron diffraction. They found that there is no ordering range longer than a few atomic spacings. Such a short-range ordering is also