Impurity Effects on the Superconductive Critical Temperature of Indium and Aluminum*†

G. CHANIN, E. A. LYNTON, AND B. SERIN
Department of Physics, Rutgers University, New Brunswick, New Jersey
(Received December 10, 1958)

We find that the effects of 0.01 to 1.0 atomic percent of various impurities on the superconductive critical temperature of indium and aluminum are strikingly similar to those previously reported for tin. For sufficiently small amounts of all solutes there is an electronic mean free path effect on the critical temperature such that T_c decreases linearly with increasing reciprocal free path. For larger impurity concentrations the curves of critical temperature *versus* concentration fall into groups according to the valence difference between solvent and solute. T_c has a sharp upward trend for higher valence solutes; for those with lower valence the curves of T_c tend to flatten out. The similarity of these impurity effects for three such different superconductors suggests that they are a fundamental property independent of the detailed nature of the superconductor.

1. INTRODUCTION

IN a recent paper[1] (hereafter referred to as LSZ) measurements were presented of the effect of alloying on the superconductive critical temperature of tin. They showed that for sufficiently small amounts of all solutes there is an electronic mean free path effect on the critical temperature such that T_c decreases linearly with increasing reciprocal free path. For larger impurity concentrations the behavior of T_c becomes more complicated; the curves of critical temperature *versus* concentration fall into groups according to the valence difference between solvent and solute. For higher valence solutes T_c has a sharp upward trend; for those with lower valence the curves of T_c tend to flatten out. We have now completed similar measurements on a series of dilute binary alloys with indium and aluminum as the solvents. We have found that the impurity effect on the critical temperature of these substances is strikingly similar to that reported for tin, as will be demonstrated in this paper.

As in LSZ, T_c was calculated from magnetic threshold field measurements in the temperature region $0.9T_c<T<T_c$. In this range the threshold field H_c is, to a very good approximation, a linear function of the temperature. T_c can therefore be found by extrapolating H_c as a function of T to $H_c=0$, using the method of least squares. The critical temperature for indium is almost as high as that for tin, so that the required temperature range for the indium alloys could be obtained with the conventional cryogenic installation described in LSZ. In order to obtain the much lower temperature range needed for the aluminum samples, an entirely different cryostat was designed and built. A description of this apparatus and of the techniques used to measure the low temperatures is given in Sec. 3.

An indication of the homogeneity of the specimens and a measure of their electronic mean free path in the normal phase was obtained by measuring their electrical resistance both at room temperature and at 4.2°K. As discussed in LSZ, from the measured resistance values we calculate for each specimen the ratio

$$\rho=R_0/(R-R_0)(1-\alpha t_c),$$

which we call the residual resistance ratio. R_0 is the resistance measured at 4.2°K, R, that at room temperature, $T=273°+t_c$, and α is the known temperature coefficient of resistance for the pure material at 273°K. To the degree of approximation to which Matthiessen's rule applies, this ratio is equal to the ratio of the ideal absolute conductivity at 273°K, $\sigma_{\text{id }273°}$, to the conductivity at 4.2°K, $\sigma_{4.2°}$. For any material the ratio of electronic mean free path l to absolute conductivity σ is a constant independent of temperature. The best way of finding its value is from measurements of the ac surface conductivity in the limit of the anomalous skin effect.[2] If in addition one knows the value of $\sigma_{\text{id }273°}$, one can find for every material a unique relation between the reciprocal of the low-temperature electronic mean free path and the resistance ratio. Table I lists the values we have used for l/σ; the references indicate the sources from which we have taken the rele-

TABLE I. Values of l/σ and of $\sigma_{\text{id }273°}$ for indium, aluminum, and tin, and the expression for $1/l$ in terms of ρ calculated from these values.

Material	l/σ (ohm-cm²)	$\sigma_{\text{id }273°}$ (ohm⁻¹-cm⁻¹)	$1/l$ (cm⁻¹)
Indium	0.89×10^{-11}[a]	13×10^{4}[b]	$(0.87\times10^{6})\rho$
Aluminum	0.40[c]	40[d,e]	$(0.62\times10^{6})\rho$
Tin	1.03[c]	10[e]	$(0.97\times10^{6})\rho$

[a] T. E. Faber, Proc. Roy. Soc. (London) **A241**, 531 (1957).
[b] G. K. White and S. B. Woods, Rev. Sci. Instr. **28**, 638 (1957).
[c] T. E. Faber and A. B. Pippard, Proc. Roy. Soc. (London) **A231**, 336 (1955).
[d] P. Alley, thesis, Rutgers University, 1958 (unpublished).
[e] A. N. Gerritsen, *Encyclopedia of Physics*, edited by S. Flügge (Springer-Verlag, Berlin, 1956), Vol. 19, Chap. 2.

* This work has been supported in part by the Office of Naval Research and by the Rutgers University Research Council.
† This work formed part of a dissertation submitted by G. Chanin to the Graduate Faculty of Rutgers University in partial fulfillment of the requirements for the Ph.D. degree.
[1] Lynton, Serin, and Zucker, J. Phys. Chem. Solids **3**, 165 (1957).
[2] R. G. Chambers, Proc. Roy. Soc. (London) **A215**, 481 (1952).

vant surface resistance. Also listed are $\sigma_{\text{id } 273°}$, and the resulting expression for $1/l$ in terms of ρ. For comparison we list values for tin as well as for indium and aluminum.

2. EXPERIMENTAL DETAILS—INDIUM

The experimental procedure used in preparing and mounting the indium specimens and in determining their critical temperature was in most details identical to that used with the tin specimens and fully described in LSZ. The basic melts were made in the same way and with similar precautions by adding the solute to 99.999% pure indium obtained from the Indium Corporation, Utica, New York. However, because of the tendency of indium to stick to glass, the final samples were cast in reactor-grade graphite. The resulting specimens were mounted in the sample holder previously used for tin, and the same method was applied to determine threshold field values over a range of temperatures near T_c. Similarly, for the resistance measurements the same apparatus and method were used as in LSZ.

3. EXPERIMENTAL DETAILS—ALUMINUM

The preparation of the aluminum samples and the apparatus and method used to determine their critical temperature were quite different from the procedures described in LSZ. Both will, therefore, be described here in detail.

(a) Preparation of Samples

To obtain samples containing a certain concentration of solute, a known amount of nominally 99.99% pure, cleaned aluminum and the appropriate amount of solute were melted together in air in an electrically heated crucible of reactor-grade graphite. Homogeneity of the melt was aided by stirring it with a graphite paddle. This was also used to skim off any surface oxide before pouring the melt into a graphite mold preheated to about 350°C. After cooling, the casting was pulled out of the mold and a sample of the required length cut from it. The final samples were a little over 3 mm in diameter and about 7 cm long. Before being measured, the samples were again cleaned and then annealed in vacuum at a temperature of 525°C for seven days. As the superconducting transitions of all samples were very sharp and some of them even showed supercooling despite being impure, we are fairly sure that they were reasonably homogeneous.

(b) Low-Temperature Apparatus

The critical temperature of aluminum is about 1.2°K, and it was therefore necessary to measure the threshold field at temperatures near 1.0°K. In addition to meeting the problem of reaching such temperatures by pumping, it was necessary for the apparatus to satisfy other requirements. At least two and preferably more samples had to be measured simultaneously in order always to have a pure aluminum sample as a thermometer. The samples had to be reasonably large so as to make the change of their magnetic moment appreciable, and they had to be closely surrounded by search coils so that this change could be detected with little loss in sensitivity. Furthermore, a whole series of measurements would be much simplified by a design which allowed us to change samples without having to break and seal vacuum-tight joints in the low-temperature portions of the apparatus.

We fulfilled these requirements by the design shown schematically in Fig. 1. The principal helium bath is the lower one, L. It has a volume of about 50 cm^3, and is suspended from an upper helium bath U by one inch of a 0.125-in. diameter, 0.010-in. wall thickness stainless steel tube. The film flow into this tube is restricted by an orifice O, 0.25 mm in diameter. The heat leak into L is further restricted by an aluminum radiation shield A, and by radiation baffles in the $\frac{5}{8}$-inch pumping tube P.

L has a tellurium-copper bottom with three sample clamps and a hole into which a 110-ohm Speer carbon resistor R is fixed with Araldite cement. With these clamps, the thermal contact with the samples S is sufficiently good so as to make them follow changes in bath temperature without noticeable time lag. The three search coils C closely surround but do not touch the samples; these three and a fourth dummy coil are attached to the removable bottom of the radiation shield, and remain at or above 4.2°K. This means that the leads to these coils do not constitute a heat leak into the lower bath L.

Between runs, samples could be changed easily by

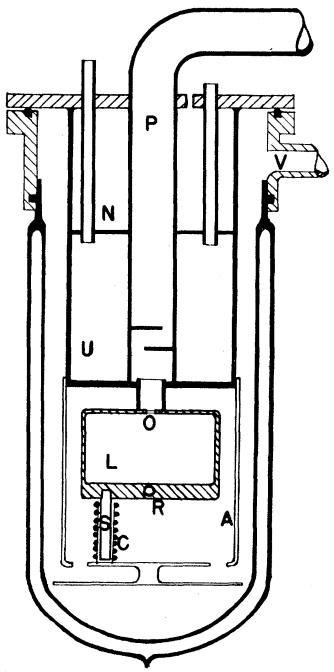

FIG. 1. Schematic diagram of the apparatus used to determine the critical temperature of the aluminum samples.

first removing the bottom of the radiation shield with its search coils. The cylindrical part of the radiation shield could then be unscrewed from the bottom of the upper bath, leaving free access to the clamps holding the three samples.

The entire assembly is sealed with O-rings into a conventional Dewar, as shown in Fig. 1, and this is in turn surrounded by a Dewar filled with liquid nitrogen. The uppermost reservoir N is also filled with liquid nitrogen.

After being precooled to liquid nitrogen temperature, the upper helium vessel is filled in a conventional way by direct transfer of liquid helium. During this filling an atmosphere of helium exchange gas fills all the spaces surrounding the baths. The lower bath L is connected through P to a helium gas holder at a slight overpressure, and as a result liquid helium condenses into L, filling it in about ten minutes. When this has occurred, helium transfer into U is stopped, and the exchange gas is removed through the sidearm V. The very favorable geometry of the apparatus allows us to pump off this gas very quickly, reaching a good vacuum in a few minutes. We are then ready to pump on L.

As long as there is liquid helium in the upper bath U, the heat leak into L is so small that we can reduce its temperature to as low as 0.85°K with a conventional 22-l/sec mechanical pump, in spite of the relatively narrow pumping line P. A single filling of the upper bath lasts about five hours.

(c) Determination of the Critical Temperature

The basic measurement of the threshold field of a given sample at a particular temperature was carried out by the method fully described in LSZ. However, we did not choose to measure accurately the low vapor pressure of the liquid helium in order to determine the temperatures near 1°K, but used instead a different thermometric procedure.

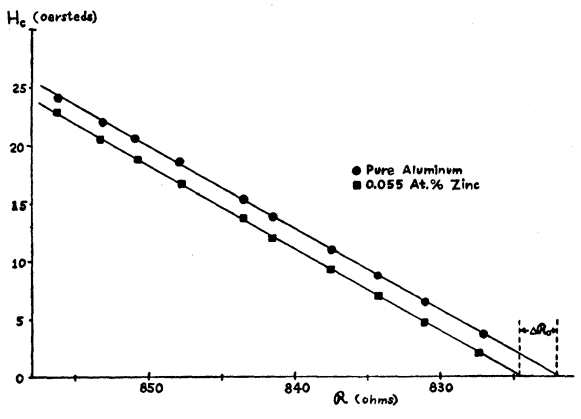

FIG. 2. Typical variation of the magnetic threshold field H_c with the resistance of the Speer resistor. Shown are data for the pure standard and one impure specimen taken during one run; the straight lines through the points are fitted by least squares.

TABLE II. Experimental results for the change in critical temperature, ΔT_c, and the resistance ratio, ρ, for all indium samples, together with the values of the reciprocal mean free path, $1/l$, calculated using the relation in Table I.

Impurity	Concentr. (at.%)	ΔT_c (°K)	ρ	$1/l$ (10^6 cm^{-1})
Pure A	...	...	<0.0001	<0.0001
Pure B	...	+0.0008	...	...
Pure C	...	−0.0002	...	...
Pure D	...	+0.0006	...	...
Bismuth	0.012	−0.0045	0.0034	0.0030
	0.020	−0.0087	0.0047	0.0041
	0.044	−0.0128	0.0095	0.0083
	0.080	−0.0158	0.0180	0.0157
	0.157	−0.0129	0.0346	0.0301
	0.314	+0.0119	0.0725	0.0631
Lead	0.042	−0.0053	0.0033	0.0029
	0.078	−0.0085	0.0062	0.0054
	0.145	−0.0118	0.0114	0.0099
	0.292	−0.0147	0.0220	0.0192
Thallium	0.045	−0.0039	0.0013	0.0011
	0.081	−0.0035	0.0021	0.0018
	0.153	−0.0063	0.0039	0.0034
	0.307	−0.0114	0.0074	0.0064
	0.529	−0.0186	0.0136	0.0118
	0.827	−0.0252	0.0198	0.0172
Tin	0.031	−0.0032	0.0016	0.0014
	0.041	−0.0035	0.0021	0.0018
	0.054	−0.0073	0.0027	0.0023
	0.072	−0.0059	0.0034	0.0030
	0.105	−0.0080	0.0051	0.0044
	0.110	−0.0083	0.0051	0.0044
	0.149	−0.0096	0.0074	0.0064
	0.179	−0.0097	0.0083	0.0072
	0.365	−0.0061	...	...
	0.533	−0.0004	...	...
Cadmium	0.037	−0.0045	0.0014	0.0012
	0.065	−0.0077	0.0027	0.0023
	0.089	−0.0078	0.0036	0.0031
	0.091	−0.0114	0.0040	0.0035
	0.155	−0.0151	0.0062	0.0054
	0.239	−0.0194	0.0097	0.0084
	0.451	−0.0302	...	...
	0.911	−0.0535	...	...
Gallium	0.042	+0.0009	0.0010	0.0009
	0.071	+0.0017	0.0015	0.0013
	0.103	−0.0005	0.0022	0.0019
	0.143	+0.0021	0.0028	0.0024
	0.145	+0.0012	0.0030	0.0026
	0.274	+0.0048	0.0050	0.0043
	0.485	+0.0134	...	...
	0.914	+0.0325	...	...

We restricted our measurements to temperatures less than 0.15°K below the critical temperature of pure aluminum, a range of temperatures over which its threshold field varies linearly with temperature to very high accuracy. We observed that in this region the threshold field also varies linearly with the resistance of the Speer resistor, indicating that this resistance in turn varies linearly with temperature in this restricted range. The method of determining the critical temperatures was therefore as follows:

In any given run, with the same pure sample and two impure ones in the apparatus, we determined the

TABLE III. Experimental results for the change in critical temperature, ΔT_c, relative to the actual "pure" specimen used, as well as the corrected change, ΔT_c^*, relative to an ideally pure specimen, for all aluminum samples. In addition experimental values of the resistance ratio, ρ, and the values of the reciprocal mean free path, $1/l$, calculated using the relation in Table I.

Impurity	Concentr. (at.%)	ΔT_c (°K)	ΔT_c^* (°K)	ρ	$1/l$ (10^6 cm^{-1})
Pure A	···	···	−0.0043	0.0026	0.0016
Pure B	···	−0.0003	···	···	···
Zinc	0.021	−0.0056	−0.0099	0.0044	0.0027
	0.036	−0.0080	−0.0123	0.0056	0.0035
	0.055	−0.0127	−0.0170	0.0082	0.0051
	0.109	−0.0190	−0.0233	0.0129	0.0080
	0.246	−0.0281	−0.0324	0.0250	0.0155
	0.495	−0.0366	−0.0409	0.051[a]	0.032
	1.006	−0.0444	−0.0487	0.079[a]	0.049
Magnesium	0.022	−0.0021	−0.0064	0.0035	0.0022
	0.060	−0.0051	−0.0094	0.0050	0.0031
	0.102	−0.0074	−0.0117	0.0078	0.0048
	0.248	−0.013	−0.017	0.0127	0.0079
	0.499	−0.0445	−0.0488	0.0725	0.0450
	1.10	−0.0478	−0.0521	0.108	0.067
Silver	0.012	−0.0102	−0.0145	0.0075	0.0046
	0.025	−0.0168	−0.0211	0.0137	0.0085
	0.050	−0.0303	−0.0346	0.0245	0.0152
	0.112	−0.0423	−0.0466	0.0535	0.0332
	0.213	−0.0543	−0.0586	0.0948	0.0588
Germanium	0.012	−0.0030	−0.0073	0.0064	0.0040
	0.024	−0.0055	−0.0098	0.0103	0.0064
	0.039	−0.0057	−0.0100	0.0199	0.0123
	0.108	−0.0026	−0.0069	0.0378	0.0234
	0.160	+0.0024	−0.0019	0.0544	0.0337
Silicon	0.026	−0.0035	−0.0078	0.0086	0.0054
	0.054	−0.0048	−0.0091	0.0155	0.0096

[a] Measured by P. Alley, thesis, Rutgers University, 1958 (unpublished).

threshold field for each specimen at nine or ten temperatures corresponding to different resistance values, $\mathcal{R}$, of the Speer resistor. By extrapolation we then found for each sample $\mathcal{R}_0$, the resistance value at zero field, and from this in turn $\Delta\mathcal{R}_0$, the difference between $\mathcal{R}_0$ for an impure sample and $\mathcal{R}_0$ for the pure one. From the measurements in each run we also obtained the slope of the linear H *vs* $\mathcal{R}$ curve for the pure sample. Multiplying the reciprocal of this by the value for $(dH_c/dT)_{T_c}$ measured by Cochran and Mapother[3] (142 gauss/degree) gave us for the given run the temperature dependence of the resistor, from which the measured $\Delta\mathcal{R}_0$ could be translated into a ΔT_c.

The Speer resistor had a value of 800–900 ohm at the critical temperature. Its temperature dependence in this region was about 200 ohms/degree, and varied by less than 5% from run to run. Typical values for the variation of H_c with $\mathcal{R}$ are shown in Fig. 2.

4. RESULTS AND DISCUSSION

In Tables II and III are listed the results of our measurements for each of our indium and aluminum specimens. ΔT_c is the difference between the critical temperature of the given sample and that of the pure specimen used as standard in each run. The use of the same pure sample in each run and the calculation of ΔT_c from its T_c as obtained in that particular run eliminated most systematic errors due to thermometry, temperature scale, and coil calibration.

The first entries in each table show the results obtained with different pure specimens to check on the reliability of our ΔT_c values. The amount of variation of T_c for these samples is less than the estimate of our uncertainties as outlined in LSZ and supports our belief that we can determine ΔT_c to within two millidegrees or less.

The last column in each table lists the reciprocal of the electronic mean free path at low temperature, calculated from the resistance ratio ρ with the relation listed in Table I. The pure indium we used has a resistance ratio smaller than 10^{-4}, corresponding to a value of $1/l$ less than 10^{+2} cm^{-1}, which is completely negligible. Our "pure" aluminum, on the other hand, has $\rho=0.0026$, so that $1/l\simeq 1600$ cm^{-1}, which is only a little smaller than that of the most dilute impure aluminum specimens. Such a high value of ρ for nominally pure aluminum is typical; e.g., Faber[4] reports similar values.

This means, of course, that the critical temperature of such a specimen is different from that of an ideally pure one with a negligible value of $1/l$. For any quantitative consideration of the impurity effect on T_c one should use the critical temperature of such an ideal sample as a standard of comparison. We shall show presently that the initial portion of a plot of ΔT_c *vs* $1/l$

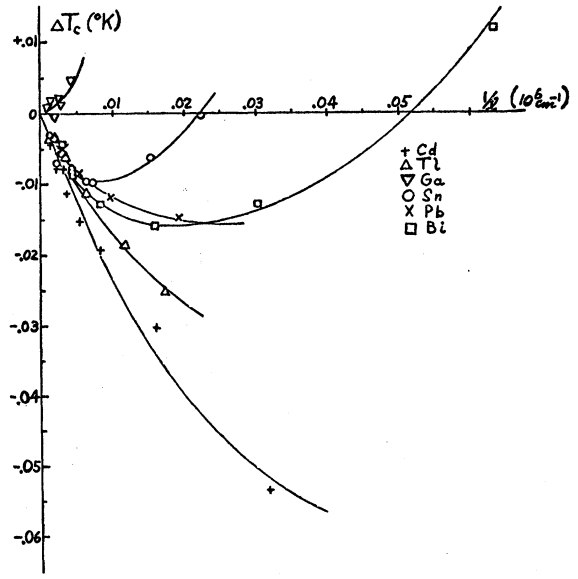

FIG. 3. The variation of the change in the critical temperature, ΔT_c, as a function of the reciprocal electronic free path, $1/l$, for the indium specimens.

[3] J. F. Cochran and D. E. Mapother, Phys. Rev. **111**, 132 (1958).

[4] T. E. Faber, Proc. Roy. Soc. (London) **A231**, 353 (1955).

is linear. Thus we can extrapolate back to $1/l=0$ and find that our "pure" aluminum has a critical temperature which is 0.0043°K below the ideal value. Adding this to the ΔT_c obtained for each impure sample gives ΔT_c^*, the value of ΔT_c relative to ideally pure aluminum. Note that to do this for indium or tin would require a correction of much less than one millidegree.

In Fig. 3 we have plotted the variation of ΔT_c as a function of $1/l$ for indium, in Fig. 4 that of ΔT_c^* for aluminum. Plotting the same ΔT_c and ΔT_c^* values as functions of the impurity concentration, rather than $1/l$, has the same effect as it did in the case of tin (see Fig. 2, in LSZ): the striking clustering of the initial portions of the curves in Figs. 3 and 4 disappears. From a large scale plot of these portions this initial decrease in ΔT_c is observed to be linearly proportional to $1/l$, with slopes which are listed in Table IV. The value of this slope for tin is given for comparison.

A further inspection of Figs. 3 and 4 shows that the variation of T_c with impurity is strikingly similar for both indium and aluminum throughout the entire range of mean free path. In both cases there is a marked difference between one set of curves which after the initial linear decrease has a sharp upward trend, and another set which also deviates upward from the initial straight line but much more gradually. In the case of indium samples the sharp rise is displayed by the curves for Bi, Pb, and Sn impurities; with aluminum as host this is true for Si and Ge. All these impurities have a valence which is *higher* than that of the solvent. The set of curves which rises only a little from the initial line consists of Cd in the case of indium, and Zn, Mg, and Ag for aluminum; all these have a *lower* valence than their hosts. Furthermore, the curve for indium samples containing thallium, which has the same valence as indium, falls somewhere between these two groups.

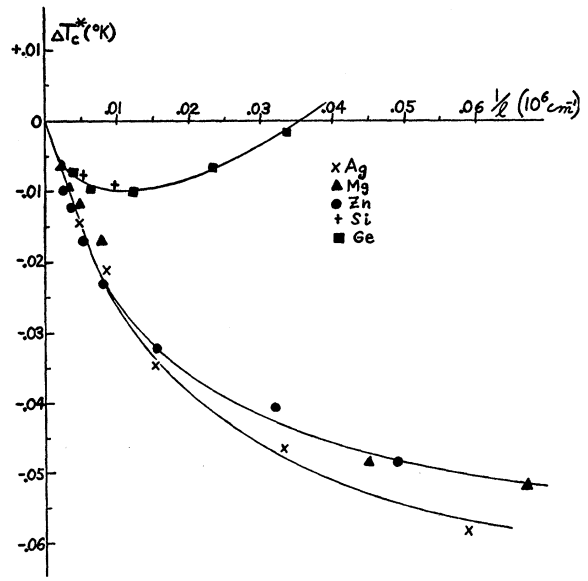

FIG. 4. The variation of the corrected change in the critical temperature, ΔT_c^*, as a function of the reciprocal electronic free path, $1/l$, for the aluminum specimens.

TABLE IV. Initial slopes of the curves of ΔT_c *versus* $1/l$.

Material	Initial slope $\Delta T_c/(1/l)$ (degree-cm)
Indium	$(2.6\pm0.7)\times10^{-6}$
Aluminum	$(2.7\pm0.3)\times10^{-6}$
Tin	$(2.7\pm0.3)\times10^{-6}$

The only exception to this general pattern of behavior is the variation of T_c with the addition of gallium to indium. Gallium and thallium have the same valence as indium, and one would expect their curves to be similar. Figure 3 shows that this is clearly not the case.

The results for indium and aluminum are very similar not only to each other, but also to our previous results for tin. In view of the fact that these three elements are widely dissimilar in almost every respect except for their being superconductors, we are strongly inclined to conclude that all the qualitative features of the impurity effect on T_c found in these investigations are fundamental properties independent of the detailed nature of the superconductor.

According to the Bardeen, Cooper, and Schrieffer theory of superconductivity,[5] the critical temperature of a superconductor depends on its fundamental parameters by the relation

$$T_c=A\exp(-1/NV),$$

where A is a constant of the material depending only on its Debye temperature, N is the density of electronic energy states at the Fermi surface, and V an average matrix element arising from the electron-phonon and the Coulomb interaction.

The linear decrease of T_c with increasing values of the reciprocal electronic mean free path occurs immediately for amounts of impurity too small to affect either the Debye temperature or the density of states. The effect on T_c must therefore be due to a dependence of V on $1/l$. Pippard has suggested[6] that this matrix element depends on the electronic mean free path in a similar way as does the scattering of phonons by electrons. Our results seem to agree with this view qualitatively.

To explain the more complicated variation of T_c beyond the initial linear region, one must take into account the effect of the impurities on the other quantities affecting T_c. Clearly impurities with different numbers of valence electrons than the solvent may affect the density of states N by changing the number of electrons per atom. This by itself is probably the explanation for the difference between the T_c variation of electropositive and electronegative impurities. However, it does not seem to be sufficient to explain the

[5] Bardeen, Cooper, and Schrieffer, Phys. Rev. **108**, 1175 (1957).
[6] A. B. Pippard, J. Phys. Chem. Solids **3**, 175 (1957).

TABLE V. Slopes of the curves of the resistance ratio *versus* atomic concentration.

System	ρ/at.%
In-Bi	0.23
In-Pb	0.078
In-Sn	0.048
In-Cd	0.040
In-Tl	0.024
In-Ga	0.020

general upward trend of all T_c curves. In an attempt to obtain further information about this question, we are presently extending our threshold field measurements on indium and aluminum samples to much lower temperatures so as to be able to calculate the electronic specific heat in the normal state. We discussed in LSZ how we did this for the impure tin samples, and how we found that the temperature coefficient of the specific heat and hence the density of states appeared to be increased by all solutes, independently of valence. Clearly it is important to determine whether a similar effect exists in indium and aluminum, as this may explain the general upward trend of T_c. As an independent check, work is in progress on calorimetric measurements of the specific heat.

We do not understand the fact that gallium impurity does not seem to depress the critical temperature of indium. It may be that the metallurgy of this solution is more complicated than is thought. In any case, whatever the mechanisms may be which cause the changes in critical temperature in the region beyond the initial linear one, one would expect them to compete more strongly with the mean free path effect, the more slowly the free path varies with impurity concentration. A measure of this variation is obtained from the rate of change of the resistance ratio ρ with impurity concentration. The values we obtain for this rate are listed in Table V for the indium alloys.

Of all the binary systems we have investigated, the mean free path of that containing gallium clearly varies most slowly with increasing impurity concentration. It is possible, therefore, that the anomalous behavior of the critical temperature of the alloys containing gallium is due to the masking of the mean free path effect by the other competing influences which in these, as in all other alloys, tend to increase the critical temperature. However, it must be noted that we seem to be able to observe readily the changes produced in the indium-thallium alloys, for which the slope listed in Table V is almost as small. Gallium atoms differ from thallium in that the former are smaller in size and in mass than indium atoms, whereas the latter are larger and heavier. Thus it may be that the anomalous behavior of the critical temperature of the In-Ga alloys arises from size and mass effects which are not very evident in the other alloy systems.

We do not believe that the peculiar behavior of this one alloy system invalidates our general conclusions.

5. ACKNOWLEDGMENTS

We continue to profit greatly from many discussions with Professor E. Abrahams and Professor P. R. Weiss. We are also grateful to Professor D. E. Mapother for sending us details of his work before its publication.

We would like to thank Dr. P. Alley for his assistance in the preparation of the aluminum samples.

The aluminum used in this work was given to us by the Aluminum Company of America through the kind efforts of Mr. J. C. Dorgan, to whom we would like to express our appreciation.

One of us (G. C.) acknowledges most gratefully the award of a Radio Corporation of America Fellowship for the year 1957–1958.

Mr. A. Siemons assisted in the design of our apparatus and was largely responsible for its construction.