Effect of Specimen Thickness on the Superconducting Critical Temperature of Indium†

W. D. Gregory, M. A. Superata, and P. J. Carroll
Department of Physics, Georgetown University, Washington, D. C. 20007
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The superconducting critical temperature (T_c) is found to decrease inversely proportional to the boundary-limited mean free path (mfp) for thin single-crystal and polycrystal In plates. Since the boundary-limited mfp is not linearly related to specimen thickness for the sample thicknesses used (5–250 μm), these results are a stronger confirmation of the Markowitz-Kadanoff calculations than previously reported in the literature. The In results contrast with similar experiments using Ga single crystals reported by Gregory in which no mfp effect was observed. Possible reasons for this difference are discussed. The possible use of measurements such as these to determine the anisotropic-pairing interaction is discussed.

Previously, one of us reported a null result[1] in a search for the "mean free path" (mfp) effect, produced by boundary scattering, using thin Ga single crystals. (The mfp effect is a depression of T_c proportional to $1/l$, where l is the mfp. This effect is caused by the averaging of the anisotropic-pairing interaction during the scattering process.[2]) The present paper reports a contrasting positive result obtained from a similar experiment using In single crystals and polycrystals.

The methods of making specimens[3] and measuring their critical temperatures relative to a bulk single crystal[4] in zero magnetic field are essentially the same as reported previously. Twelve randomly oriented 99.999% pure In single crystals, grown from seed in molds as flat plates 6 mm wide, 25 mm long, and ranging in thickness from 23 to 250 μ, were used in this work. Figure 1 shows the difference of the T_c of the thin plates and that of a bulk sample (called ΔT_c) plotted versus $1/d$ where d is the specimen thickness. The previous results for Ga single crystals are also plotted for comparison. While no shift of T_c within the scatter in the data is seen for Ga, the In data fit an expression given by

$$\frac{\Delta T_c}{1/d} = -(2.81 \pm 0.15) \times 10^{-6} \text{ K cm} .$$

Lynton and McLachlan[5] and Gregory[1] previously measured a finite shift of T_c in pounded and annealed polycrystalline In foils 5–6 μ thick. If these data are plotted along with the single-crystal In data (see Fig. 2), all of the data can be fit to the expression

$$\frac{\Delta T_c}{1/l} = -(2.94 \pm 0.15) \times 10^{-6} \text{ K cm},$$

where l is the mfp due to boundary scattering,[6] i.e.,

$$l = \tfrac{1}{2} d \ln(l_0/d) , \qquad (1)$$

where l_0 is the bulk mfp, taken as 350 μ for all of the data. (Residual-resistivity measurements made during this work and that of Ref. 5 confirm this value of l_0.) Although the polycrystalline specimens exhibited slightly broader superconducting transitions than the single crystals, it would seem that, since the bulk mfp l_0 were the same for all specimens, the reduced values of ΔT_c seen for the polycrystalline samples are due to the logarithmic factor in l and are not the result of an accidental compensation of part of the mfp effect by residual strain or damage left after annealing. (The fact that T_c and l_0 are the same for the polycrystalline and single-crystal samples of comparable thickness, while the transition width is larger for the polycrystalline foils, is not inconsistent. For example, the transition width is known to be more seriously affected by imperfections than T_c.[7])

Thus, the use of both polycrystalline and single-crystal data allows an evaluation of ΔT_c over a sufficiently large range of specimen thicknesses to distinguish between a true mfp effect and a "$1/d$" effect. Comparison of these data to the Markowitz-Kadanoff theory[2] for the mfp effect shows that the free parameter in this theory λ^i (the ratio of the characteristic time to destroy anisotropy to the transport-collision time, i.e., $\lambda^i = \tau_{\text{anis}}/\tau_{\text{coll}}$) is very nearly unity ($\lambda^i \approx 1$), implying that the scat-

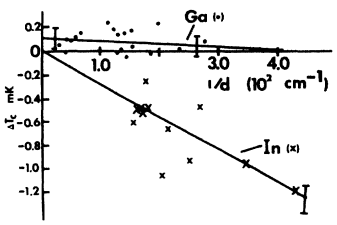

FIG. 1. ΔT_c versus $1/d$ for single crystals of Ga and In where $\Delta T_c \equiv T_c$ (thin plate) $- T_c$ (bulk sample) and d is the specimen thickness. The Ga data are taken from Ref. 1.

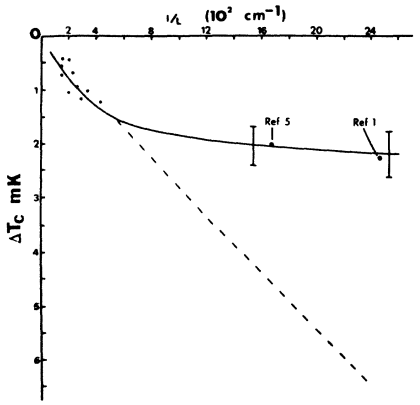

FIG. 2. Solid line $-\Delta T_c$ versus $1/l$ for In where l is the boundary-limited mfp given by Eq. (1). All of the data were taken from this work except the two points marked that were taken from Refs. 1 and 5. Dashed line – the fit of ΔT_c to $1/d$ from Fig. 1, showing the difference between a "$1/l$" and "$1/d$" effect.

tering process is "diffuse," according to the theory of Ref. 1. Using this same theory, we concluded[1] that the null result for Ga could be explained, within the mfp theory, by assuming that boundary scattering in Ga is "specular" or correlated, at least for those electrons affecting T_c. Several possible reasons why correlated scattering would occur were proposed in Ref. 1, with the contrast to In somewhat uncertain because of a concern about the origin of a shift in T_c in the polycrystalline samples. Now that the shift in T_c in In is confirmed with single-crystal samples, one of these postulates deserves further consideration. That is, Ga has a highly anisotropic Fermi surface while In has an almost isotropic Fermi surface. Price[8] and Ham and Mattis[9] have shown that for materials with highly anisotropic Fermi surfaces, the conditions of conservation of energy and momentum during the scattering at a boundary would severely limit the range of scattering angles (producing correlated or specular scattering) while a material with an isotropic Fermi surface would have many allowed scattering angles (producing uncorrelated or diffuse scattering). Our results for Ga and In would agree with this observation.

On the other hand, a more basic reason for a lack of a mfp effect in Ga would be a lack of an energy-gap anisotropy in the first place. Since several authors estimated large values of gap anisotropy for Ga (see Table I), this did not seem a likely argument previously and, therefore, was not presented. In fact, one of us (W. D. G.) had obtained preliminary measurements of the anisotropy[1] using the mfp effect produced by addition of silver impurity to Ga and found a reasonable anisotropy (~ 15%). Recently, however, direct measurements of the gap energy by tunneling into bulk Ga single crystals by two different groups[10,11] has yielded estimates of the gap anisotropy in Ga no larger than 5%. If one now compares all of the methods for measuring gap anisotropy, one finds that surface-type measurements (such as tunneling or the boundary-scattering shift of T_c) produce low estimates of the anisotropy and bulk measurements (such as ultrasonic attenuation, thermal conductivity, etc.) produce high estimates of the anisotropy. This comparison is given in Table I. Although it must be cautioned that all of the anisotropy measurements are subject to some criticism (see Shepelev[12]), the fact remains that a very interesting connection appears to exist between the surface measurements and negligible anisotropy effects. Since Ga is the only superconductor that expands on freezing and is also known to collect impurities at the surface,[13] it is entirely possible that the Ga surface is strained or impure and does, locally, have most of the gap anisotropy removed.

For those single-crystal materials that do exhibit an mfp effect due to boundary scattering

TABLE I. Comparison of the Ga-energy-gap anisotropy measured by various techniques, showing the contrast in estimates obtained from "surface-" and "bulk-type" measurements.

Method measuring bulk properties		Methods measuring surface properties	
Method	$\langle a^2 \rangle$ [a]	Method	$\langle a^2 \rangle$ [a]
Mean-free-path-effect impurity scattering[b]	0.02	Mean-free-path-effect boundary scattering[b]	0.0025
Specific heat[c]	0.02	Tunneling[d]	0.0025
Critical field[e]	0.04	Tunneling[f]	0.0020
Ultrasonic attenuation[g]	0.075		
Thermal conductivity[h]	0.017		
Nuclear-spin relaxation time[i]	0.011		

[a] $\langle a^2 \rangle$ defined as in Ref. 2.
[b] Reference 1.
[c] T. P. Sheahen, J. F. Cochran, and W. D. Gregory (unpublished).
[d] Reference 10.
[e] Reference 4.
[f] Reference 11.
[g] B. W. Roberts and H. R. Hart, Bull. Am. Phys. Soc. 7, 185 (1962); also (private communication).
[h] N. V. Zavaritskii, Zh. Eksperim. i Teor. Fiz. 37, 1506 (1959) [Soviet Phys. JETP 10, 1069 (1960)].
[i] R. H. Hammond and W. D. Knight, Phys. Rev. 120, 762 (1960).

(such as In), we note that application of the theory of Ref. 1 shows that the shift of T_c will depend on the orientation of the crystal with respect to the surface of the plate and that a study of the mfp effect for samples of varying orientation may be used to unfold the anisotropic-pairing interaction. In fact, some of the scatter in the data in Fig. 1 seems to be related to variation in crystal orientation. Using Ref. 1, the angular dependent part of ΔT_c would seem to be about 30% of the total. This part of ΔT_c should be measurable, to 10% accuracy, if the scatter in the data can be reduced by a factor of 3.

Finally, we note that Naugle and Glover[14] have recently reported a $1/d$-type depression of T_c, for thin films of amorphous materials. The total ΔT_c, as well as the slope $(\Delta T_c/d^{-1})$ that they observe, are in good agreement with the Maskowitz-Kadanoff[2] theory, leading to the interesting speculation that they are seeing the mfp effect. However, it is difficult to see how the amorphous materials they use could have any of the gap anisotropy required to observe an mfp effect. Also, they note that the resistances per square ($R_\square$) of their films are proportional to $1/d$ and that the films appear to have no ordered structure beyond tens of angstroms. These results imply that the mfp is much smaller than their film thicknesses so that the sample boundary would not determine l. We must conclude at the present time that, although similar phenomena, our data and those of Naugle and Glover are not necessarily due to the same effect.

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Self-Consistent Hartree-Fock Calculation of the Dielectric Function of an Electron Gas

James W. F. Woo and Sudhanshu S. Jha*

IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598

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The dielectric function of the electron gas has been calculated in the self-consistent Hartree-Fock approximation. Results are presented and compared with that obtained in random-phase-approximation and Hubbard theories.

In the study of the uniform electron gas, knowledge of the wave number and frequency-dependent dielectric function $\epsilon(q, \omega)$ has proved to be extremely useful. Physical properties of the system, such as the dynamic form factor $S(q, \omega)$ and the ground-state energy E_0, are expressible in terms of $\epsilon(q, \omega)$.

The most commonly used expression for the dielectric function is calculated[1] by using the self-consistent Hartree method or the random-phase approximation (RPA). In the diagrammatic perturbation theory,[2] this is equivalent to the summation of bubble diagrams. It gives a good description