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ENERGY-GAP ANISOTROPY IN In-DOPED Sn

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One of the fundamental problems yet to be solved in the physics of type-I superconductivity is that of determining the relation between the anisotropy of the superconducting energy gap, the band structure, and the phonon density of states of a real metal (cf. Douglass and Falicov[1]). The most detailed energy gap measurements on a given material are those on Sn made by the ultrasonic absorption[2-4] and electron tunneling[5] techniques. It is the purpose of this Letter to report the following: (1) the direct observation by improved ultrasonic[6] techniques of the variation with impurity concentration of the anisotropy of the superconducting energy gap in a series of In-doped Sn single crystals (<1 ppm to 0.1 at.% In); and (2) the comparison of two models for the temperature dependence of the compressional-wave attenuation in the superconducting state.

The Bardeen-Cooper-Schreiffer (BCS)[7] theory predicts that for an isotropic superconductor the ratio of the compressional-wave attenuation in the superconducting state to that in the normal state is given by

$$\alpha_s/\alpha_n = 2f(\Delta) = 2\{\exp[\Delta(T)/kT]+1\}^{-1}, \qquad (1)$$

where $\Delta(T)$ is the temperature-dependent superconducting energy gap. This result has been shown to be independent of sound frequency and electron mean free path for impurity-limited scattering.[8] Morse, Olsen, and Gavenda[2] and Leibowitz[3] have assumed that the form of Eq. (1) is correct for anisotropic superconductors, but that for a given crystal direction the value of the limiting energy gap $\Delta(0)$ can assume a value different from the BCS value of $1.76kT_c$, where T_c is the transition temperature. The $\Delta(0)$ for a given crystallographic direction is obtained by measuring the slope of a plot of $\ln(\alpha_s/\alpha_n)$ vs T_c/T over the low-temperature range, for which $f(\Delta)\sim e^{-\Delta/kT}$ and $\Delta(T)\sim\Delta(0)$. The measurements[2] on pure Sn indicate an anisotropy of ~20% for compressional-wave propagation along the [001] and [110] directions. Tunneling data[5] indicate that in addition to a possible anisotropy associated with one piece of Fermi surface there are several gaps observed for some crystalline directions in Sn which can be related to the multiple energy surfaces found in the higher Brillouin zones. That there are multiple energy gaps in a real metal such as Sn is not surprising and in fact had been predicted previously by an extension of the BCS theory to a multiband model.[9] At present there is no detailed theory for the effect of multiple gaps on the ultrasonic attenuation. It is not at all clear what the effect of interband pairing and scattering will be on the quasiparticle phonon scattering matrix. The simplest assumption is that of a complete neglect of interband effects, i.e.,

$$\frac{\alpha_s}{\alpha_n} = \sum_i \frac{A_i}{\exp[\Delta_i(T)/kT]}, \qquad (2)$$

where the summation is over all bands, and the constants A_i are subject to the condition $\sum_i A_i = 2$. This form has been used for a two-energy-gap fit for Sn with some success.[10] There is only one model for anisotropic superconductors which has been used to predict the ultrasonic attenuation.[11] The result does not give quantitative agreement with the data for Sn.

It is well established that small amounts of impurity added to a pure superconductor tend to reduce the transition temperature.[12] The reduction is linear in l_e^{-1}, where l_e is the electron mean free path, and according to the current theories[13,14] of dirty superconductivity, the decrease in transition temperature is simultaneous with a smearing out of the energy-gap anisotropy. The energy gap should be approximately isotropic when $\Delta(0) < \hbar\tau^{-1}$, where τ is the impurity-limited relaxation time. For

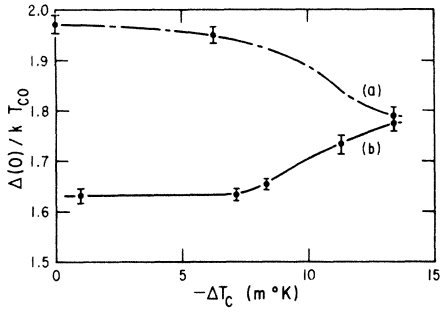

FIG. 1. Superconducting energy gap in units of kT_{c0} given as a function of $(-\Delta T_c)$, determined by single-gap estimates for (a) $q \parallel [110]$ and (b) $q \parallel [001]$.

larger concentrations, the relative charge and size of the impurity ions result in different effects on the concentration dependence of both the energy gap and transition temperature.[15-17] Measurements of infrared absorption[18] and specific heat[19] indicate that the energy-gap anisotropy is reduced by doping; however, these measurements yield less detailed information about the variation of the anisotropy about the Fermi surface than can be obtained from ultrasonic attenuation measurements.

In making measurements on the alloys, it is necessary to maintain the inequality $ql_e \geq 1$, where q is the magnitude of the sound-propagation vector, in order that the magnitude of the normal electronic attenuation be measurably large and that the selectivity of the attenuation mechanism be sufficient to reveal the anisotropic effects. Further, one would like to compare data for various samples at approximately the same value of ql_e to maintain the same degree of selectivity. In order to achieve this condition, measurements were made in a frequency range from 50 MHz for pure Sn to 460 MHz for the Sn-0.1 at.% In alloy. The total electronic attenuation ranged from 28 to 15 dB cm^{-1} indicating that the ql variation was less than a factor of 2. The noise level was reproducibly less than ~±0.02 dB cm^{-1}.

In Fig. 1 the energy gaps, in units of kT_{c0} where T_{c0} is the transition temperature of pure Sn, are given as a function of $-\Delta T_c$; T_c is a good measure of l_e in this concentration range.[12] The curves in Fig. 1 show the limiting energy gaps for $q \parallel [110]$ and $q \parallel [001]$, respectively, as determined by an analytic fit of data from ~1.2 to 2.2°K assuming the single-gap BCS model. The reduction in anisotropy is obvious, and the value of $\Delta(0)$ toward which the gaps tend lies between 1.75 and 1.80, close to the BCS value of 1.76 and in agreement with the infrared-absorption[17] and specific-heat[18] data. The energy gaps found for pure Sn are compared in Table I with other values in the literature. The values measured by the analytic fit are at the upper end of the error brackets for the graphical[2] fit in both cases. Figure 2 is a plot of $\ln(\alpha_S/\alpha_n)$ vs T_c/T for $q \parallel [001]$ in (a) pure Sn and (b) 0.1 at.% In-doped Sn for $\Delta T_c \sim -15$ mdeg K. The change in energy gap is obvious, and in addition the data for (b) fit the BCS function over the entire temperature range within a standard error of ~0.4% of the total electronic attenuation, which is approximately the observed experimental error. This result is an additional verification of the validity of the BCS theory for an isotropic superconductor.

Table I. Comparison of pure Sn energy gap $[2\Delta(0)]$ measurements.

q	Current work	Morse, Olsen, and Gavenda[a]
$\parallel [001]$	$(3.26 \pm 0.02)kT_c$	$(3.2 \pm 0.1)kT_c$
$\parallel [110]$	$(3.94 \pm 0.04)kT_c$	$(3.8 \pm 0.1)kT_c$

[a]See reference 2.

From the tunneling data[5] it is expected that two gaps ($1.5kT_c$ and $1.9kT_c$) will contribute to processes involving electrons in the [001] plane. Using Eq. (2) and assuming a two-gap fit for the pure Sn data, where $q \parallel [001]$, a fit

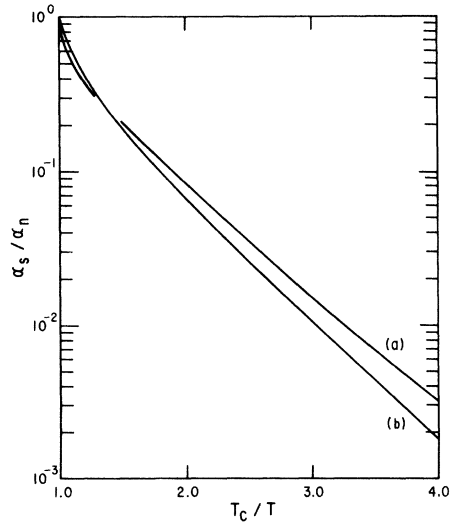

FIG. 2. $\ln(\alpha_S/\alpha_n)$ vs T_c/T for $q \parallel [001]$ in (a) pure Sn and (b) 0.1 at.% In-doped Sn.

within the experimental error is obtained over the entire temperature range for $A_1 \sim 1.5$, $\Delta_1(0) \sim 1.4kT_c$, and $\Delta_2(0) \sim 4kT_c$. Since the lower gap dominates over most of the temperature range, the determination of $\Delta_1(0)$ is much better than that of $\Delta_2(0)$. An energy gap as low as $1.4kT_c$ has been measured by tunneling methods[5]; however, $\Delta_2(0)$ appears to be much too large to be physically meaningful. For the [110] plane a multigap fit to the data is impractical since tunneling data indicate that at least four gaps will contribute. The multigap fits for both [001] and [110] planes reduce to a single-gap fit with A going to 2 for the most heavily doped samples.

A crude check of the agreement between the observed energy-gap variation and the theory of dirty superconductors can be made as follows: Using the BCS expression for ξ_0 and the condition that the anisotropy is removed when $\tau \sim \hbar[\Delta(0)]^{-1}$, it can be shown that $l_e \simeq 3.2\xi_0 = 0.7\times10^{-4}$ cm. The frequency for which $ql_e \sim 1$ for the above condition is $\nu_s \sim 750$ MHz. From the ultrasonic data it is estimated that $ql_e \sim 1$ when $\nu_s \sim 460$ MHz in the 0.1% In sample. This agreement is excellent within the limits of the estimate. These measurements are currently being extended to samples containing various amounts of Sb and Cd as well as to higher impurity concentrations. Since the anisotropic effects are removed in the range of solute concentration studied herein, the requirement on the constancy of ql_e can be relaxed somewhat for higher doping; it is only necessary to obtain a sufficiently large electronic attenuation to permit the measurement of an energy gap. Hence it is possible to compare the effects of various impurities on the energy gap as well as on the transition temperature.

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