ment factor, as calculated using the parameters determined by the over-all surface-resistance calculation of Ref. 3, are such as to account for the size and shape of the correlation-produced peak. Comparing this peak with the experimental peak observed at $\omega_c/\omega = 1.025^1$ gives the value $A_2 = -0.03$, in agreement with the value deduced by Platzman, Walsh, and Foo.[9]

ACKNOWLEDGMENTS

I should like to express thanks to Dr. J. M. Rowell, Dr. G. Dunifer, and Dr. C. C. Grimes for comments on an earlier version of this work.

---

[1] G. A. Baraff, C. C. Grimes, and P. M. Platzman, Phys. Rev. Letters 22, 590 (1969).

[2] C. C. Grimes (private communication).

[3] G. A. Baraff, Phys. Rev. 187, 851 (1969).

[4] G. A. Baraff, Phys. Rev. B 1, 4307 (1970).

[5] Y. C. Cheng, J. S. Clarke, and N. D. Mermin, Phys. Rev. Letters 20, 1486 (1968).

[6] G. A. Baraff, J. Math. Phys. 11, 1938 (1970).

[7] R. G. Chambers, in *The Physics of Metals, I, Electrons*, edited by J. M. Ziman (Cambridge U. P., Cambridge, England, 1969).

[8] The treatment given in Ref. 7, based on replacing $\sigma(k, \omega)$ by its large k form, can also be used in the diffuse case.

[9] P. M. Platzman, W. M. Walsh, Jr., and E. Ni Foo, Phys. Rev. 172, 689 (1968).

PHYSICAL REVIEW B VOLUME 2, NUMBER 3 1 AUGUST 1970

Influence of the Phonon Spectra of In-Tl Alloys on the Superconducting Transition Temperatures

R. C. Dynes
Bell Telephone Laboratories, Murray Hill, New Jersey 07974

(Received 3 February 1970)

A tunneling investigation of the effective phonon density of states through the four distinct crystal structures of the In-Tl alloy series has been performed. The resultant $\alpha^2(\omega)F(\omega)$'s extracted from these studies reflect the characteristics of each individual structure. The data obtained from these experiments serve as a test of existing theoretical expressions for determining T_c of a superconductor from normal-state parameters. It is found that the agreement is somewhat disappointing. In addition, a study of the phonon distribution, or more explicitly, an average phonon energy $\langle\omega\rangle$ in the region of the face-centered cubic to face-centered tetragonal phase transition, suggests an instability in at least one of the phonon modes. This result appears to support the suggestion that this transition is second-order.

I. INTRODUCTION

The ability to predict from various easily determined normal-state parameters whether a material will superconduct and at what temperature has been an elusive dream for many years. Until recently, the only method displaying any success was based on the empirical rules of Matthias,[1] in which, from a previous knowledge of T_c for a wide variety of superconducting elements, compounds, and alloys, and with some experience, one could determine in most cases a considerably better than order-of-magnitude estimate of T_c for a new material. Although this method has survived successfully through several eras in the development of our understanding of superconductivity, it is by no means an accurate determination of T_c. Unfortunately, it gives most people little feel for what physical parameters of the material are the important ones in determining superconductivity. The trends and correlations only hint at the microscopic mechanism or mechanisms responsible for the phenomenon, whereas a detailed study of the pertinent interactions should, in principle, predict accurately the value of T_c.

The BCS theory of superconductivity,[2] suggesting the electron-phonon interaction as the dominant mechanism, predicted that the transition temperature should be given by

$$k_B T_c = 1.14\langle\omega\rangle e^{-1/N(0)V} ,$$

where $\langle\omega\rangle$ is an average phonon energy, $N(0)$ is the electronic density of states at the Fermi surface, and V is the pairing potential due to this interaction. More understanding of the electron-phonon and electron-electron interactions followed from the BCS theory, and it is now generally believed[3] that the current theory, as summarized by the gap equation due to Eliashberg,[4] is accurate to a few percent. That is to say, if one knows all the required normal-state parameters for a metal, T_c can be calculated to this accuracy. Such calculations were performed by Swihart, Scalapino, and Wada,[5] Carbotte and Dynes,[6] and more recently by Allen and Cohen.[7] In general such calculations are tedious and have been performed mainly as a confirmation of the expected accuracy of the theory. It is obviously easier, in most cases, to measure T_c than to perform such a calculation. However, for Mg and Li a prediction of T_c has been made,[7] and in the other alkali metals it has been shown that the phonon coupling is too weak to overcome the Coulomb term down to unattainable temperatures. In view of the unwieldy nature of the above calculations, McMillan[8] saw that it was desirable to obtain a simple expression for T_c which contained only parameters which were either known or easily calculable, for the majority of metals. His development of such an expression, the McMillan equation, also illustrates rather simply the important physical properties of a metal which determine superconductivity. More explicitly he showed that, for purposes of determining the critical temperature, materials can be categorized by three parameters: the Coulomb term μ^*, an average phonon energy $\langle\omega\rangle$ and an electron-phonon coupling term λ, where

$$\lambda = 2\int_0^\infty [\alpha^2(\omega)F(\omega)/\omega]\, d\omega \quad .$$

Here $F(\omega)$ is the phonon density of states of the material and $\alpha^2(\omega)$ is a measure of the electron-phonon interaction.

It had been shown earlier by Morel and Anderson[9] that μ^* does not vary strongly over many materials and reasonable guesses can be made of its value. The average phonon frequency $\langle\omega\rangle$ can be equated most simply with the Debye temperature, or preferably calculated from $F(\omega)$ if this is known from neutron scattering. Thus, the only remaining unknown parameter is λ.

This parameter can be determined from an experimental measure of the electronic mass enhancement due to the electron-phonon interaction.[10] It is easily seen[8] that the ratio of the measured electronic effective mass m^* to the band-structure effective mass m is given by

$$m^*/m = 1 + \lambda \quad .$$

Recently, the expression for T_c developed by McMillan has been tested and modified by other investigators[11] studying amorphous systems,[12,13] and the concepts and ideas pointed out in McMillan's work have been used to explain the enhancement of T_c's of various superconductors[14] under extreme conditions.

An alternative single method for determining all three parameters λ, $\langle\omega\rangle$, and μ^* is that of a tunneling experiment. The technique of single-particle tunneling has proven to be an extremely fruitful method for the study of elements,[15] alloys,[16,17] and more recently, amorphous[12,13] materials in the superconducting state. Various investigators have employed the unique properties of a superconducting tunnel junction to investigate not only the superconducting characteristics of these various solid states but also some of the normal-state parameters. McMillan and Rowell have developed an iterative inversion of the Eliashberg gap equations,[4] based on the tunneling density of states.[3] Deviations from the BCS-theory behavior of the conductance of a superconducting tunnel junction are explained in terms of strong electron-phonon coupling and finite quasiparticle lifetimes. They are able to extract the product function $\alpha^2(\omega)F(\omega)$ and μ^*[18] directly from this inversion. The product function $\alpha^2(\omega)F(\omega)$, in turn, has been very useful in determining both superconducting and normal-state properties of the material dependent upon the electron-phonon interaction, such as electron-phonon coupling strength, phonon renormalized effective electron mass, and superconducting critical temperatures[19] under various conditions, for various metals and alloys. This tunneling technique is restricted to the more strongly coupled superconductors with $T_c \geq 2.5$ °K, but yields detailed information about λ, μ^*, and the frequency distribution. Thus, all the information required to calculate T_c is at hand, but it is again obvious that a direct determination of T_c is trivial compared to the tunneling investigation. Thus, "predictions" of T_c will be confined to materials for which tunneling experiments can be performed, and the available tunneling results can be used to check the applicability and accuracy of the theoretical expressions.

In this paper we report the results of an investigation of the superconducting properties of the Tl-In alloy system using the tunneling technique; the results serve as a test for the predictions of existing T_c calculations. This system is convenient for an investigation of this type for several reasons. Firstly, Tl and In are in the same column in the periodic table; as one is alloyed into the other, the electron concentration remains fixed at three electrons/atom. This feature reduces the extreme variation of T_c found for other alloy systems where this is not the case, and results in a system measurable using conventional tunneling techniques throughout the entire concentration range. This allows a comprehensive study of the phonon spectra of these alloys. Secondly, progressing through the entire range of concentration, there are four distinct crystal structures that can be stabilized and studied.[20] In Fig. 1 is illustrated the phase diagram for this binary system. Starting with indium, which is face-centered tetragonal (fct), on increasing the Tl concentration at low temperatures there is a phase transition at 31 at.% into a fcc system. At approximately 55% Tl, a mixed phase of fcc and bcc begins. We have been able to isolate a pure bcc structure at approximately 70%, but this phase is short-lived with respect to concentration, as on proceeding to higher Tl concentration the system transforms to the hcp structure of pure Tl. Hence, in this investigation one can study the phonon spectra and test the expressions for T_c under conditions of various crystal structures for both alloys and pure materials. As it is observed that each particular crystal structure has its own distinctively shaped phonon distribution, this study serves to critically examine these expressions for T_c (which were derived employing well-defined model phonon distributions), under the conditions of varying shapes of the $\alpha^2(\omega)F(\omega)$ functions.

In addition to this study of T_c and $\alpha^2(\omega)F(\omega)$ throughout the alloy series, we have looked more closely at the phase transition, face-centered tetragonal to fcc, at ≈ 31% Tl concentration. It has been suggested[21] that this transition is reminiscent of the transition in V_3Si, in that it looks very much second order. If such were the case, it would be necessary that the internal symmetry change by more than is implied from the macroscopic strain. It has been proposed that this reduction in symmetry could be brought about by a phonon instability. The peak in T_c versus concentration at the point of the transition was cited as possible evidence for this instability. In this paper, we report the results of a search for this proposed instability in the phonon spectrum.

The plan of the paper is as follows. In Sec. II, we outline the current status of the McMillan expression for T_c, how it was derived, and the implications of McMillan's results; also, the modifications to this expression and reasons for them are described. In Sec. III, an outline of the experimental details of this investigation, where they differ from standard techniques, is presented, and a description of the measurement of T_c is included. Section IV contains the results of the experiments and a comparison of the experimental and calculated values of T_c. Section V reports the results of a more careful look at the fcc to fct transition and in Sec. VI conclusions are drawn.

II. THEORY

Since the BCS theory,[2] much progress has been made in the understanding of the electron-phonon interaction and its manifestations in normal and superconducting metals. The Eliashberg theory[4] takes into account the retarded nature of the electron-phonon-electron interaction which is responsible for superconductivity in simple metals and treats the strength of this interaction to accuracy $(m/M)^{1/2}$, where m equals the electronic mass and

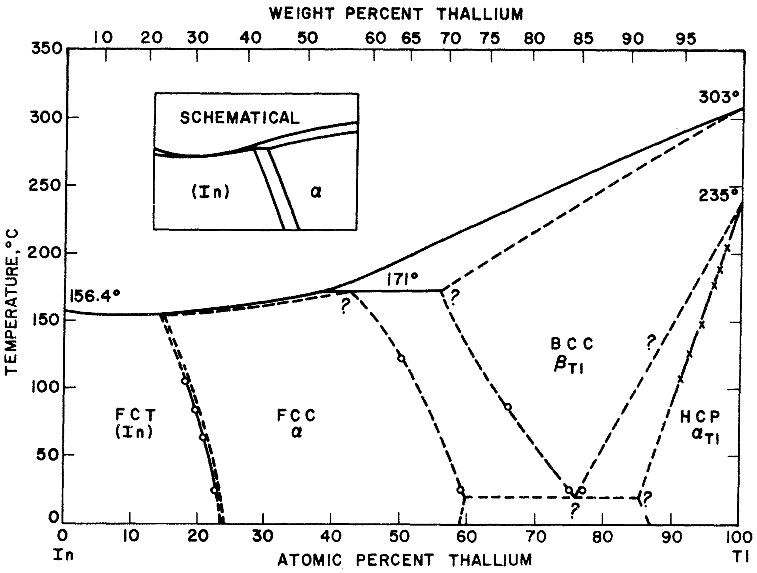

FIG. 1. Concentration-phase diagram at higher temperatures for the In-Tl system (Ref. 20).

M equals the ion mass. Treating the electron-electron Coulomb repulsions in a pseudopotential manner (following Morel and Anderson[9]) completes the picture and, given the necessary normal-state properties with sufficient accuracy, the superconductor can be described through a solution of the Eliashberg gap equations. This yields a determination of the energy gap function $\Delta(\omega)$; through a finite-temperature solution of these equations one can obtain the temperature dependence of the gap parameter $\Delta(\omega, T)$ and hence, the critical temperature T_c.[19] By a suitable linearization of the gap equations at T_c McMillan has shown[8] that the equation for T_c will assume the general form

$$T_c = \langle\omega\rangle \exp\left[-(1+\lambda)/(A\lambda - B\mu^* - C(\langle\omega\rangle/\omega_0)\lambda\mu^*)\right], \quad (1)$$

where $\langle\omega\rangle$ is an average phonon energy of the material, λ is the electron-phonon coupling strength defined by[3]

$$\lambda = 2\int_0^\infty [\alpha^2(\omega)F(\omega)/\omega]\, d\omega \quad , \qquad (2)$$

and μ^* is the Coulomb repulsion pseudopotential term given by

$$\mu^* = N(0)\, U_c = N(0)\, V_c/[1 + N(0)\, V_c \ln(E_F/\omega_c)] \quad .$$

E_F is the Fermi energy, V_c is the spherical average of the screened Coulomb potential between electrons, $N(0)$ is the electronic density of states at the Fermi level, and ω_0 is a Coulomb cutoff energy greater than $\langle\omega\rangle$ but less than E_F. $A, B,$ and C are parameters of order unity, and ω_0 is the upper cutoff energy of the effective phonon distribution $\alpha^2(\omega)F(\omega)$. Choosing the Debye temperature as a characteristic phonon energy, the gap equations were solved numerically and the parameters determined. However, it was necessary, to obtain this solution, to assume a model shape for the phonon distribution $F(\omega)$. A simplified niobium spectrum was taken. The limitations of this choice will be discussed below. Using this technique the expression derived for T_c was[8]

$$T_c = (\Theta_D/1.45) \times \exp\{-[1.04(1+\lambda)/(\lambda - \mu^* - 0.62\lambda\mu^*)]\} \quad . \qquad (3)$$

One could just as easily choose, instead of Θ_D, an average phonon energy given by

$$\langle\omega\rangle = \int_0^\infty \alpha^2(\omega)F(\omega)\, d\omega \Big/ \int_0^\infty \frac{\alpha^2(\omega)F(\omega)\, d\omega}{\omega} \quad . \qquad (4)$$

In as much as we are probing the phonon distributions and $\alpha^2(\omega)F(\omega)$ is determined by tunneling, this $\langle\omega\rangle$ is more readily obtained than Θ_D. The result of renormalizing for $\langle\omega\rangle$ rather than Θ_D yields for Eq. (3)

$$T_c = (\langle\omega\rangle/1.20) \times \exp\{-[1.04(1+\lambda)/(\lambda - \mu^* - 0.62\lambda\mu^*)]\} \qquad (5)$$

The central quantity of interest in this work, the product function $\alpha^2(\omega)F(\omega)$, is defined by[3]

$$\alpha^2(\omega)F(\omega) = \int_{S_F} d^2p \int_{S'_F} d^2p' \times \sum\nolimits_\lambda \left[\,|g_{(\vec{p};\vec{p}';\lambda)}|^2/(2\pi)^3 v_F\right] \times \delta(\omega - \omega'(\vec{p} - \vec{p}';\lambda)) \Big/ \int_{S_F} d^2p \quad , \qquad (6)$$

where the two integrals in p and p' are taken over the Fermi surface S_F, v_F is the Fermi velocity, $g_{(\vec{p}\vec{p}';\lambda)}$ is the electron-phonon coupling term from state $\vec{p}$ to state $\vec{p}'$ via a phonon $\vec{p} - \vec{p}'$ of branch λ. For some of the more simple materials and alloys that are known to have almost spherical Fermi surfaces and where inelastic neutron scattering results describing the phonons of the system are available, this quantity has been calculated with good success.[6] However, the number of materials for which the data are available and one can expect good results is quite limited. For Tl and In and their alloys, tunneling appears to be the easiest method of obtaining the frequency distributions.

McMillan has shown that from (6) it follows easily that

$$\int_0^\infty \alpha^2(\omega)F(\omega)\omega\, d\omega \doteq N(0)\hbar\langle v^2\rangle/2M \quad , \qquad (7)$$

where $\langle v^2\rangle$ is the square of the electron-ion pseudopotential form factor averaged over all scatterings from one point on the Fermi surface to another. From (2), it follows then, that

$$\lambda = N(0)\langle v^2\rangle/M\langle\omega^2\rangle \quad , \qquad (8)$$

where $\langle\omega^2\rangle$, the average phonon energy squared, is

$$\langle\omega^2\rangle = \int_0^\infty d\omega\, \omega\alpha^2(\omega)F(\omega) \Big/ \int_0^\infty \frac{\alpha^2(\omega)F(\omega)}{\omega}\, d\omega \quad . \qquad (9)$$

If the electronic heat-capacity coefficient γ is known, the band-structure density of states at the Fermi surface $N(0)$ can be related to γ and λ by

$$N(0) = 3\gamma/2\pi^2 k_B^2(1+\lambda) \quad , \qquad (10)$$

and this can be applied to (8) to yield an absolute determination of the average electron-ion pseudopotential.

Employing this procedure where T_c, Θ_D, and γ were available, McMillan was able to obtain λ from (3) and hence, determine $N(0)$ with good success for several interesting transition-metal alloy series. Empirically, with the available data, it was found that for a given class of materials the product $N(0)\langle v^2\rangle$ remained relatively constant, although the individual components $N(0)$ and $\langle v^2\rangle$ varied by a factor of 10. The significance of this observation, as was pointed out by McMillan, was the fact that if $N(0)\langle v^2\rangle$ is assumed to be constant for a class of materials, and Eq. (8) is substituted into (5), it is seen that T_c is dominated only by the average phonon energies. This feature has been used by investigators[14] to explain the enhancement of T_c in granular Al films by invoking low-energy surface modes which alter λ and raise T_c.

For simple metals, pseudopotential theory allows us to say even more about the electron-phonon coupling if we can work in the almost-free-electron approximation. In the spirit of this approximation, it has been shown[8] that the coupling constant λ can be given by

$$\lambda = \frac{1.51}{r_s}\,\frac{\langle \bar{v}_q^2 \rangle}{(\langle \omega^2 \rangle / \Omega_p^2)} \quad , \tag{11}$$

where r_s is the radius in atomic units of a sphere containing one electron, Ω_p is the ionic plasma frequency, and $\langle \bar{v}_q^2 \rangle$ is a dimensionless electron-ion matrix element defined by

$$\langle \bar{v}_q^2 \rangle = \frac{\int_0^{2k_F} |\langle \vec{k}+\vec{q}\,|v|\vec{k}\rangle|^2 q^3\, dq}{\int_0^{2k_F} |\langle \vec{k}\,|v|\vec{k}\rangle|^2 q^3\, dq} \quad , \tag{12}$$

where $\langle \vec{k}+\vec{q}\,|v|\vec{k}\rangle$ is the pseudopotential form factor between the states on the Fermi surface $|\vec{k}+\vec{q}\rangle$ and $|\vec{k}\rangle$. Equation (11), although a somewhat simplified form for λ, can be helpful in determining approximately the effect the electron-phonon renormalization has upon the electronic effective mass, the ratio of the effective mass to the band-structure mass being given by

$$m^*/m = 1 + \lambda \quad . \tag{13}$$

In the spirit of this free-electron approximation and calculating $\langle \bar{v}_q^2 \rangle$ from the Heine-Abarenkov form of the pseudopotential, this mass enhancement can be calculated to be $m^*/m = 2.34$ for Tl and 1.99 for In. These values are not in very good agreement with those measured experimentally or the results presented in Sec. IV, and point out the fact that the free-electron assumption, in these simple metals, is not accurate enough for detailed considerations.

Recently, Garland and Allen[11] have argued that the work of McMillan is not sufficiently general and does not take into account variations in the shape of the phonon spectrum from the model of niobium used in the original calculations. These variations can result from two effects: a different crystal structure from Nb resulting in a differently shaped phonon distribution, or lattice disorder, resulting in a smearing out of the spectrum, causing deviations in Θ_D or $\langle \omega \rangle$. Following the method of McMillan via the computer, a two-Lorentzian model of the phonon distribution was used to simulate various tunneling results on crystalline and amorphous materials, and an expression similar to (3) was derived, given by

$$T_c = (0.25 + 0.67x)\,\omega_0 \times \exp\left(\frac{1+\lambda}{(0.53 + 0.62x^{1/2})\lambda - \mu^* - (0.25+0.67x)\lambda\mu^*}\right) , \tag{14}$$

where the additional parameter x is a measure of the weighting of the frequency distribution relative to its cutoff energy ω_0, and is defined by

$$x = \langle \omega^2 \rangle / \omega_0^2 \; . \tag{15}$$

From the results of tunneling into various types of superconductors it has become clear, since the work of McMillan, that this factor will vary for different crystal structures and degrees of disorder. Any distribution with very strong low-energy modes, such as that of amorphous materials or of Hg, will have a lower x than those with a single very peaked distribution near ω_0.

Using this modification of McMillan's work, Garland and Allen have more accurately described the critical temperatures of amorphous Ga and Bi [12,13] condensed on low-temperature substrates. It is argued that this expression is much more generally applicable in extreme cases than the McMillan expression, which was derived for a single-phonon distribution. This is achieved at the expense of an additional parameter x which is somewhat difficult to determine unless detailed knowledge of the phonon distribution is obtainable.

With the introduction of still another parameter (which is difficult to measure short of determining a complete frequency distribution) it should be emphasized that it is still much easier to simply measure T_c of most materials than to determine $\alpha^2(\omega)F(\omega)$ from tunneling or $F(\omega)$ from neutron scattering.

III. EXPERIMENTAL

Many of the experimental aspects of this work are by now standard and have been extensively described elsewhere.[18] These techniques will only be outlined briefly in this section. Only details that are new or slightly different will be treated in any depth.

The thin-film tunnel junctions were of the Al-insulator-alloy type, and the experiments were performed at 1.0°K so that the aluminum film was superconducting. The alloys were prepared by a flash evaporation technique in which tiny pellets of the previously prepared alloy were individually dropped onto a resistively heated Mo boat. Each film consisted of approximately 100–200 pellets so that any inhomogeneities were, hopefully, of short range. For materials of very similar vapor pressures, this method results in a very high level of reproducibility[16,22] while the reliability necessarily decreases with increasing difference in vapor pressures. For this alloy series it was found that the resultant film concentrations were within a few percent of the pellet concentrations in spite of the moderate differences in the vapor pressures. The deviations, where they existed, were always on the Tl-rich side, as expected, but from sample to sample these deviations were fairly consistent, of the order of a few percent, and each alloy reported here represents several junctions prepared as a check for reliability.

The values of T_c of the alloys were measured using a method particularly suitable for a tunneling experiment.[23] The zero-bias resistance of the alloy junction was monitored as a function of temperature and at T_c a sharp discontinuity in the slope of this curve appeared as the energy gap in the excitation spectrum of the superconductor began to open. This measurement was easily effected using the identical first derivative techniques which were later employed in measuring the phonon effects in these alloys. A small (~ 20 nA) ac signal was applied across the junction at zero bias and the fundamental ac voltage detected through a lock-in amplifier. The result of a typical measurement of resistance versus T for the alloy $\mathrm{Tl}_{0.5}\mathrm{In}_{0.5}$ is shown in Fig. 2, where the discontinuity in slope of this curve is clearly displayed. We choose to define T_c as that point where the extrapolated resistance intersects the baseline for $T > T_c$.

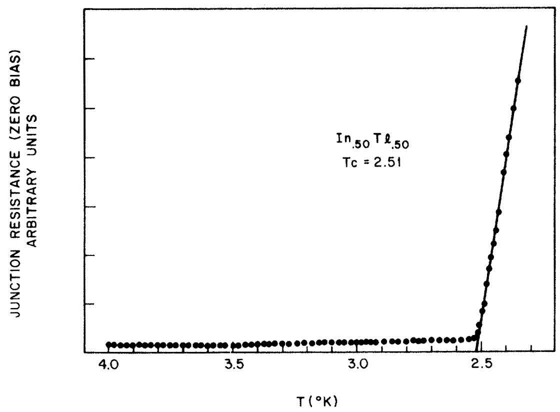

FIG. 2. Zero-bias junction resistance as a function of T for the alloy $\mathrm{In}_{0.50}\mathrm{Tl}_{0.50}$.

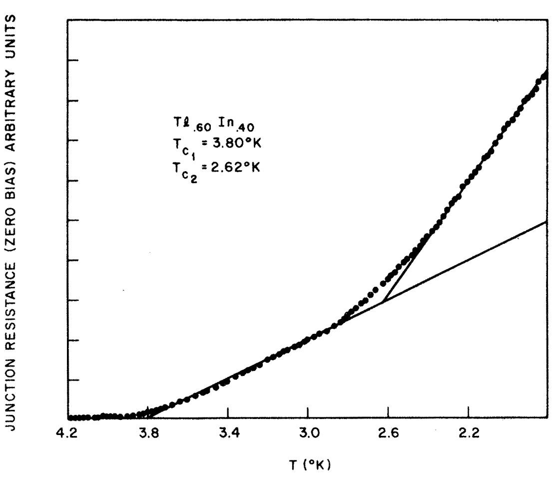

FIG. 3. Zero-bias junction resistance as a function of T for the alloy $\mathrm{Tl}_{0.60}\mathrm{In}_{0.40}$.

As has been pointed out previously,[22] the tunneling technique is quite sensitive to the detection of multiphases in an alloy series. This has been previously emphasized in the detection, in the I–V characteristics, of multiple energy-gap structure corresponding to tunneling into the different phases. This present technique is also sensitive to the measurement of various phases in that, at the critical temperature of each phase of the material, there is a change in slope in this R-versus-T plot. This point is better illustrated in Fig. 3, where zero-bias resistance versus T of a $\mathrm{Tl}_{0.6}\mathrm{In}_{0.4}$ alloy (a mixture of bcc and fcc) is plotted. Here we can fit two straight lines to the curve corresponding to the two transition temperatures

$$T_c(\mathrm{bcc}) = 3.80\ ^\circ\mathrm{K} \text{ and } T_c(\mathrm{fcc}) = 2.62\ ^\circ\mathrm{K}.$$

From the I–V characteristic at $T = 1.0\ ^\circ\mathrm{K}$, the energy gaps of the two phases are measured to be

$$\Delta_0(\mathrm{bcc}) = 0.630\,\mathrm{meV},\quad \Delta_0(\mathrm{fcc}) = 0.420\,\mathrm{meV},$$

yielding the BCS ratios of

$$(2\Delta_0/k_BT_c)_{\mathrm{bcc}} = 3.86,\ (2\Delta_0/k_BT_c)_{\mathrm{fcc}} = 3.73\ ,$$

the higher T_c phase displaying a stronger coupling ratio than the lower fcc phase.

The criteria outlined previously[22] for an acceptable tunnel-junction were, again, rigidly adhered to. In order to obtain reliable results for the product function $\alpha^2(\omega)F(\omega)$ and the normal-state parameters it is felt that the junctions must display a minimal amount of nontunneling "leakage" current and that the Coulomb pseudopotential term be fairly constant throughout the entire alloy series, 0.10–0.14.

The junctions were measured using the harmonic detection of a small modulating signal and a resistance bridge.[24] The ac signal (500 c/sec) was never greater than 100 μV rms and usually much less. As fine details of the phonon spectra of alloys are smeared due to the inherent nature of an alloy, the 100 μV did not contribute any additional smearing.

The Tl–In alloy series is of interest for several reasons. It has various crystallographic phase transformations such that we can investigate the phonon distributions of several different crystal structures by simply varying alloy concentration. There is no valence change throughout the system, so only the average atomic mass and interatomic forces are changed. When the crystal structure of a particular alloy film was in doubt in the course of this investigation, the phase was verified by x-ray diffraction. In no case did we find any conflict between our determinations of crystal structure and those of other investigators[25,26]; so, it is felt that the films were good representatives of the bulk alloy as investigated by others.

In general, we have found that the resultant film-alloy concentrations were a few percent Tl-rich compared to the initially prepared alloy. This was determined simply by comparing our measured values of T_c with those of other workers[25,26] on bulk samples. In quoting concentrations we choose to assign those concentrations rather than our initial alloy concentrations. In all cases, this corresponds to only a slight change and has very little, if any, effect on the final analysis of the data. A comparison of the quoted and prepared concentrations is given in Table I. From this table it is clear that the flash evaporated film of these constituents displays T_c characteristics in fairly good agreement with the concentration desired. The alloy $\text{In}_{0.40}\,\text{Tl}_{0.60}$ was not extensively investigated owing to its strong two-phase nature. The two alloys $\text{In}_{0.07}\,\text{Tl}_{0.93}$ and $\text{In}_{0.17}\,\text{Tl}_{0.83}$ showed very slight traces of the bcc phase consistently but, as the transition was almost complete to hcp, these alloys were treated as single-phase materials. The only indication of the bcc phase in these alloys was a slight hump in the plots of dI/dV versus V at Δ_0 for this phase. This corresponds to a small current rise at Δ_0 (bcc) but the strength of this rise was < 1% of the current rise at Δ_0 (hcp) and was ignored.

TABLE I. Concentrations of alloys studied and their crystal structures.

Prepared alloy	Quoted alloy	X^{tal} structure
In	In	fct
$\text{In}_{0.90}\text{Tl}_{0.10}$	$\text{In}_{0.90}\text{Tl}_{0.10}$	fct
$\text{In}_{0.75}\text{Tl}_{0.25}$	$\text{In}_{0.73}\text{Tl}_{0.27}$	fct
$\text{In}_{0.70}\text{Tl}_{0.30}$	$\text{In}_{0.67}\text{Tl}_{0.33}$	fcc
$\text{In}_{0.65}\text{Tl}_{0.35}$	$\text{In}_{0.63}\text{Tl}_{0.37}$	fcc
$\text{In}_{0.60}\text{Tl}_{0.40}$	$\text{In}_{0.57}\text{Tl}_{0.43}$	fcc
$\text{In}_{0.50}\text{Tl}_{0.50}$	$\text{In}_{0.50}\text{Tl}_{0.50}$	fcc
$\text{In}_{0.30}\text{Tl}_{0.70}$	$\text{In}_{0.27}\text{Tl}_{0.73}$	bcc
$\text{In}_{0.20}\text{Tl}_{0.80}$	$\text{In}_{0.17}\text{Tl}_{0.83}$	hcp + weak bcc
$\text{In}_{0.10}\text{Tl}_{0.90}$	$\text{In}_{0.07}\text{Tl}_{0.93}$	hcp + very weak bcc
Tl	Tl	hcp

Annealing of the films in vacuum up to 72 h at room temperature had very little effect on the results except in this slightly mixed phase of bcc and hcp. The bcc contribution to these alloys decreased even more and was almost indiscernible in several samples.

IV. RESULTS AND DISCUSSION

In Fig. 4 the variation of T_c with alloy concentration and crystal structure is illustrated over the entire range, and the alloys studied in this work are illustrated as points on this curve. We have observed, in agreement with others,[25,26] that the bcc phase is the highest T_c phase. The abrupt changes in crystal structure from fct to fcc at ≈ 30% Tl and from fcc to bcc between 50 and 70% are clearly evident in the characteristics of this figure, while the transition bcc to hcp is barely discernible due to its gradual nature.

The calculated $\alpha^2(\omega)F(\omega)$ and μ^* determined from the conductance of the junction for several of the typical alloys studied is illustrated in Fig. 5. Immediately obvious are the differences in shapes of the various distributions depending upon their individual crystal structures. The indium-rich fct and fcc structures display the typical shape of a face-centered system: two clearly resolved peaks, one transverse peak and one longitudinal peak. The typical bcc phonon distribution has two separate transverse phonon peaks and one longitudinal mode, the familiar shape of the alkali metals, which is clearly evident in the bcc $\text{Tl}_{0.73}\,\text{In}_{0.27}$ system. The other distinct crystal structure studied, the hcp system typical of Tl, shows a distinctively strong low-energy mode characteristic of hexagonal systems and a much more complicated structure at higher energies. These distinctive fingerprints of each crystal structure serve as additional evidence that the structure of the films studied is representative of the bulk alloy.

In Fig. 5, it is also of interest to note that as the pure material (In or Tl) is alloyed, the fine structure in the phonon distributions becomes rapidly smeared out and only the gross features remain. This phenomenon has been reported previously[16,22] and is a general trait of alloy systems. This smearing is a result of the fact that because of the random nature of an alloy there will be microscopic fluctuations in the interatomic forces and the atomic masses. This results in a finite phonon lifetime causing a substantial uncertainty smearing in energy of the eigenfrequencies of the solid. This smearing results in the replacement of the δ functions in Eq. (6) with functions of a finite linewidth repre-

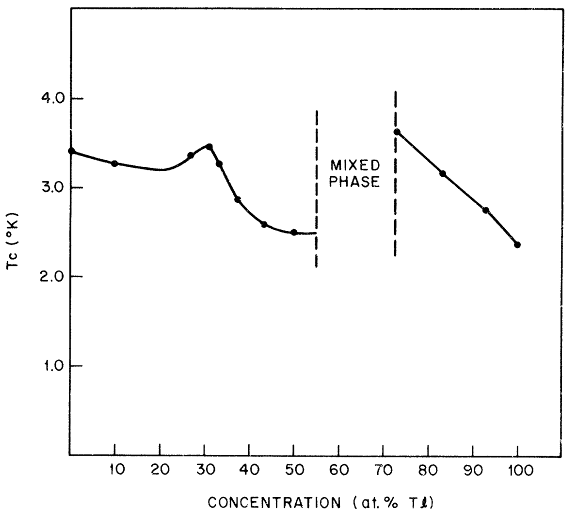

FIG. 4. T_c versus concentration for the Tl-In system. The points on the curve represent alloys studied in this work.

sentative of the smearing, causing a much smoother $F(\omega)$ distribution than in the pure material. In a perturbative calculation,[22] it has been shown that this smearing is a complicated function of energy and momentum of the frequency modes of the crystal, but in general, the longitudinal peak is substantially more smeared than the transverse modes. As a first approximation one can assume that the energy smearing $\Delta\omega$ varies linearly with energy throughout the distribution so that the ratio $\Delta\omega/\omega$ is a constant.

In the spirit of this overly simplified assumption, we have started with the pure In and pure Tl $\alpha^2(\omega) \times F(\omega)$ distributions and determined a best fit to the alloy data by convoluting these distributions with Lorentzian functions according to the prescription $\Delta\omega/\omega = \text{const}$, where $\Delta\omega$ equals the Lorentzian linewidth. The details and justification for this procedure have been reported previously.[22] The calculation was performed using the pure In distribution throughout the fct and fcc regions (up to 50% Tl) and repeated from the Tl end up to the end of the strong hcp phase. In this fashion, it is possible to obtain an estimate of the amount of energy smearing in the very high-concentration alloys. The results of this simple-minded calculation are illustrated in Fig. 6, where $\Delta\omega/\omega$ is plotted as a function of concentration. Although the results should be treated simply as an indication of the degree of smearing, in view of the simplicity of the calculation, it is interesting to note that it appears that the amount of energy smearing does not depend drastically upon crystal structure.

The point is, that from a tunneling investigation of this type, where a pure material is subsequent-

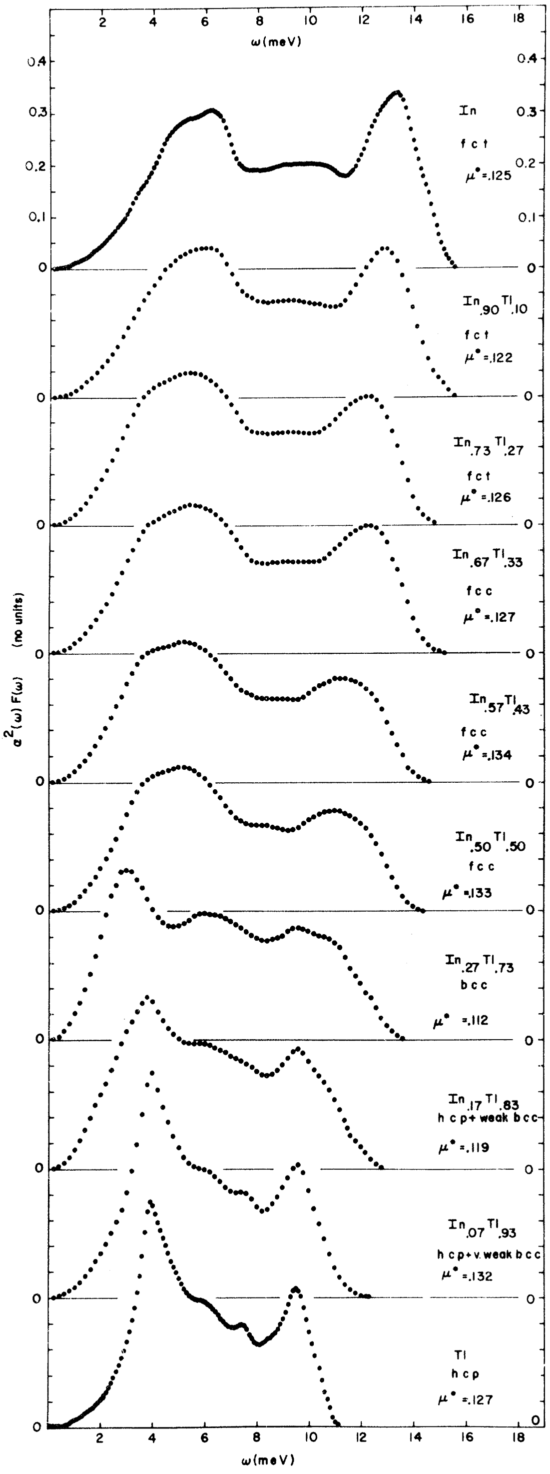

FIG. 5. $\alpha^2(\omega)F(\omega)$ versus ω for most of the alloys studied.

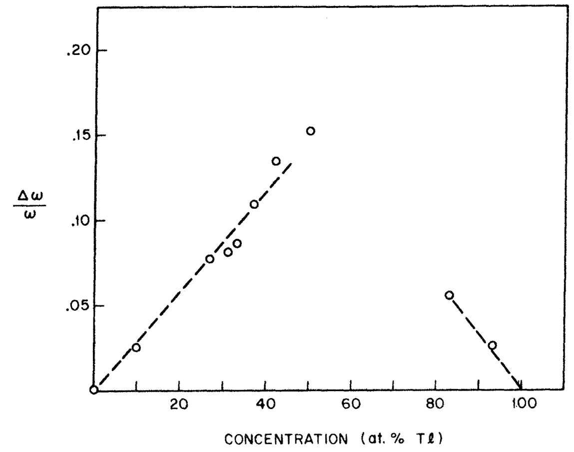

FIG. 6. $\Delta\omega/\omega$ as a function of concentration obtained as a best fit to the data by smearing the pure metal distributions of Fig. 5.

ly alloyed, we are now in a position to determine what effect these impurities have on the scattering time of the phonons of the system. Inelastic neutron scattering methods as yet do not have the resolution to make quantitative statements of this type. As we shall see in Sec. V, this phenomenon has the effect of washing out fine structure due to individual phonon modes, making some types of analyses more difficult.

From the data of Fig. 5, we are now in a position to calculate the parameters $\langle\omega\rangle$, $\langle\omega^2\rangle$, λ, and T_c as defined in Sec. II. As we can readily calculate $\langle\omega\rangle$ from our data and as it is felt that this is a more significant number than Θ_D to describe the phonon distribution, we choose to calculate T_c using the McMillan expression of Eq. (5). The parameters, as well as the measured T_c and those values of T_c calculated from the work of McMillan and of Garland and Allen, are given in Table II, and a comparison of the calculated and measured values is shown in Fig. 7. From this comparison it becomes immediately clear that the calculated T_c's are not in the anticipated good agreement with the measured values. One expression consistently predicts higher T_c's and the other lower. Both expressions qualitatively predict the correct concentration dependence, with that of Garland performing somewhat better in this respect but at the expense of an additional parameter.

From this comparison of the measured and calculated values of T_c, it is clear that neither expression has great success for this simple metal system, even when all the required data are carefully measured. It is interesting to note that the McMillan expression is most accurate for Tl, but large deviations occur as In is added. It is also interesting to note that with increasing alloy concentration there is a significant redistribution of the phonon energies by smearing and the $\alpha^2(\omega)F(\omega)$ for very low ω is enhanced. This shifting of modes to low energies has the over-all effect of increasing significantly the value of λ, as the lower energies in $\alpha^2(\omega)F(\omega)$ are more heavily weighted by virtue of the $1/\omega$ term in Eq. (2). In contrast, these lower modes will not have nearly as significant an effect on the actual T_c of the material. In the calculations of McMillan, the model $\alpha^2(\omega)F(\omega)$ distribution was cut off at lower energies thus effectively decreasing the λ for a particular measured T_c. It is felt that applying similar analysis and a phonon distribution with a more realistic low-energy spectrum, the value of 1.04 in Eq. (5) (slope of $\ln\langle\omega\rangle/T_c$ versus $(1+\lambda)/\lambda$ for $\mu^*=0$) would be increased, thus tending to decrease the predicted T_c's for this system and other systems known to have very low-energy phonon modes.

In view of these results and previous tunneling results on amorphous systems[12,13] we can see qualitatively that as the phonon spectrum becomes more and more distorted from that of niobium, the increasing difference between the predicted T_c using McMillan's expression and that obtained experimentally indicates that a more realistic shape for $\alpha^2(\omega)F(\omega)$ must be considered.

On the other hand, it is not clear why the expression of Garland and Allen, which explicitly attempts

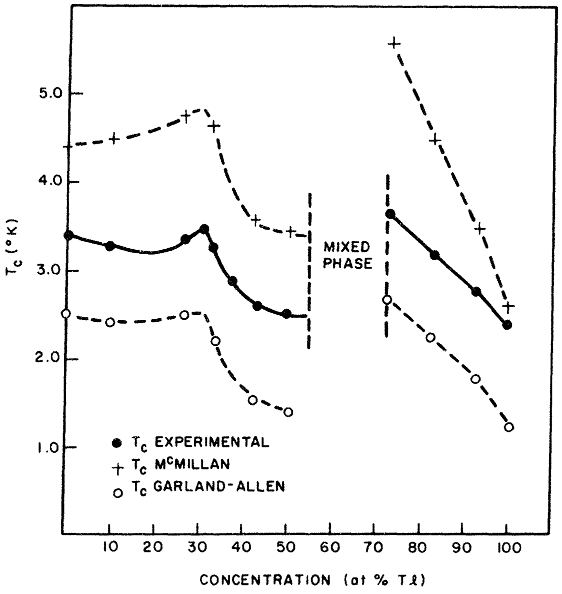

FIG. 7. Comparison of the experimentally measured T_c's with those obtained from the theoretical expressions of McMillan and Garland and Allen.

TABLE II. Relevant parameters calculated as described in Sec. II to determine T_c theoretically. $T_{c\,\mathrm{expt}}$ = experimental T_c. $T_{c\,\mathrm{McMill}} = T_c$ calculated using McMillan expression of Eq. (5). $T_{c\,\mathrm{G.A.}} = T_c$ calculated using Garland-Allen expression of Eq. (14).

Alloy	$\langle\omega\rangle$ (meV)	$\langle\omega^2\rangle$ (meV^2)	λ	$T_{c\,\mathrm{expt}}$ (°K)	$T_{c\,\mathrm{McMill}}$ (°K)	$T_{c\,\mathrm{G.A.}}$ (°K)	$\frac{\langle\omega^2\rangle}{\omega_0^2}$
In	8.86	61.17	0.834	3.40	4.41	2.57	0.2613
$In_{0.90}Tl_{0.10}$	8.41	54.30	0.850	3.28	4.46	2.40	0.2413
$In_{0.73}Tl_{0.27}$	7.67	44.19	0.933	3.36	4.79	2.47	0.2131
$In_{0.67}Tl_{0.33}$	7.81	46.00	0.899	3.26	4.51	2.21	0.2218
$In_{0.57}Tl_{0.43}$	7.33	40.50	0.847	2.60	3.56	1.68	0.2066
$In_{0.50}Tl_{0.50}$	7.20	39.32	0.835	2.52	3.41	1.57	0.2035
$In_{0.27}Tl_{0.73}$	6.46	29.32	1.092	3.64	5.59	2.76	0.1609
$In_{0.17}Tl_{0.83}$	6.30	29.45	0.980	3.19	4.46	2.26	0.1916
$In_{0.07}Tl_{0.93}$	6.09	29.61	0.889	2.77	3.47	1.79	0.2319
Tl	6.04	30.13	0.780	2.33	2.55	1.41	0.2584

to take into account this redistribution of the phonon modes, is not more accurate. The parameter $\langle\omega^2\rangle/\omega_0^2$ is certainly a measure of this shifting as is observed from Table II. A comparison of these values and the distributions of Fig. 5 yields the expected result, that as alloying increases there is a general trend for this parameter to decrease. This result is in some sense similar to the result in Fig. 6, where it was seen that the distributions could be characterized by a $\Delta\omega/\omega$ smearing factor which also tended to increase with increasing alloy concentration. It should be emphasized that from Fig. 7 it is clear that Garland and Allen correctly and quite accurately predict the shape of the T_c-versus-concentration plot. The absolute prediction, on the other hand, consistently underestimates by an almost constant amount.

For a given class of material, it was noted[8] that the product $N(0)\langle v^2\rangle$ was approximately a constant. From (8) we know that

$$N(0)\langle v^2\rangle = \lambda \bar{M}\langle\omega^2\rangle \ ,$$

where $\bar{M}$ is the average atomic mass for the particular concentration under consideration. The right-hand side of this equation consists of measurables from these results. Consequently, it is possible to obtain the product of the band-structure density of states at the Fermi surface times an average electron-ion potential for scattering around the Fermi surface.

This product is plotted as a function of concentration in Fig. 8, where it is seen that over the various crystal structures of the series there is approximately a 25% deviation from constancy. In fact, comparison of Fig. 8 with Fig. 4 reveals a remarkable similarity. Although the variation of $N(0)\ \langle v^2\rangle$ is not as great as the variation of T_c, the correlation is unmistakable and it is difficult to say whether this variation throughout the alloy series is mainly due to the band-structure density of states $N(0)$ or a result of strong fluctuations in $\langle v^2\rangle$ over various crystal structures. In fact, considering only In and Tl, we see from Fig. 8 that the value of $N(0)\ \langle v^2\rangle$ for In is 1.25 that of Tl. Employing the existing electronic specific-heat data for these two materials and Eq. (10), we find that the ratio of the band-structure density of states of the two materials $N(0)\,\mathrm{In}/N(0)\,\mathrm{Tl} = 1.12$, leaving approximately an equal amount of variation required in the $\langle v^2\rangle$ term to explain the results.

In order to separate the variation of these two parameters throughout the alloy system, it will be necessary to know either one of them. A measurement of the electronic contribution to the specific heat at low temperatures for this series would directly yield $N(0)$, and the variation of the two effects could be determined.

That these materials cannot be considered to be of the free-electron nature, which negates any accurate detailed first-principles calculation of the type previously performed in other cases,[6] can be illustrated by applying Eq. (11) in a straightforward fashion. Using an average Heine-Abarenkov[27] form for the pseudopotential and calculating r_s from existing data[25,26] on these materials, we can derive an almost-free-electron value of λ from (11). A comparison of this calculated value with that determined from tunneling data is illustrated in Table III. This table is essentially another way of presenting the data of Fig. 8. In this way, however, these deviations point out the fact that if truly accurate determinations of the superconducting properties of materials, whose Fermi surfaces are not almost spherical, are to be made, then band-structure effects must be carefully considered as an inherent part of the calculation. In addition, it can probably be safely assumed that all the deviation in Table III is not due to the deviations in band structure, and a more precise knowledge of the electron-ion pseudopotential is required. Again,

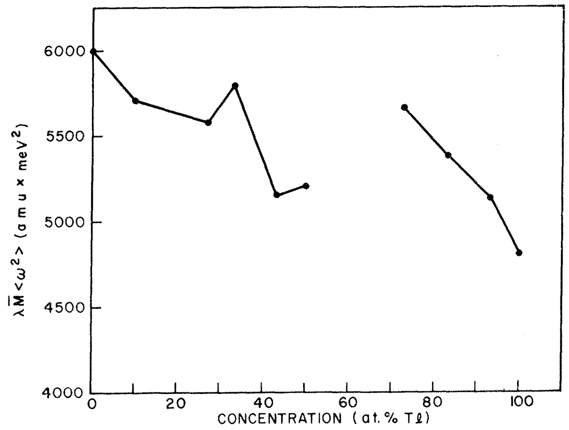

FIG. 8. $\lambda\bar{M}\langle\omega^2\rangle$ as a function of concentration. This is equal to the product $N(0)\langle v^2\rangle$, as explained in the text.

it is difficult without specific-heat measurements to separate the two, but the conclusion, that both details must be more reliably known than at the present time, remains.

V. fcc → fct TRANSITION

It has been suggested[28] that the transition fcc to fct is very reminiscent of a similar transition in V_3Si and several other βW structure superconductors. It has also been noted that this type of transformation (in the case of V_3Si, just above the superconducting transition temperature T_c) looks very much second order in that it goes to completion at T_t and the accompanying deformation is small and increases with $T_t - T$. In addition, the crystallography and the specific heat resemble second-order transitions. Anderson and Blount[21] have argued that, according to Landau's general theory of second-order phase transitions,[29] a transition from cubic to tetragonal in which the only change undergone is the macroscopic strain is necessarily first order. It is not clear that this transition is a strongly first-order transition and it was suggested by these authors, in order to account for this contradiction, that the symmetry of the phase heretofore considered tetragonal may be a less symmetric structure, which would be a result of the longitudinal or transverse phonon at the zone boundary point in the 100 direction going unstable. The peak in T_c of Fig. 4 at approximately the critical concentration for transformation was cited as a possible manifestation of this phonon instability. This unstable low-frequency mode would enhance the electron-phonon coupling and increase T_c in the region of the instability.

Referring to Fig. 5, it is quite clear that there are no obvious phonon instabilities in the set of $\alpha^2(\omega)F(\omega)$ distributions about the region where T_c is peaked ($\approx$ 30% Tl in In). Since we are working with an alloy system and we have shown that one can assign an estimate of the energy smearing ($\Delta\omega/\omega \approx$ 8%) for the alloys about this concentration, the distributions obtained are necessarily very smooth. Thus, individual critical points, corresponding to van Hove singularities in the phonon dispersion curves, are not discernable in the alloys. Hence, one has not much hope of seeing concentration dependence of a particular critical-point instability. One can, however, determine from these distributions an average phonon energy defined by Eq. (4). By studying this average phonon energy, it can be determined whether the increase in T_c at this point of instability is due to a shift in some particular phonon modes or due to some electronic parameter which would alter the $\alpha^2(\omega)$ function only and not appear in the determination of $\langle\omega\rangle$. Experimentally to vary concentration is easier than to vary temperature to study this $\langle\omega\rangle$; we have done it this way. In a sense it would be a more pleasing experiment to study $\langle\omega\rangle$ as one went through the transition on the temperature variable. The fact that we rely on the superconducting properties of such materials makes this a difficult task.

The results of the determination of $\langle\omega\rangle$ in the region of this transformation are illustrated in Fig. 9. It is quite clear that associated with this peak in T_c versus concentration there is a decrease in the average phonon energy $\langle\omega\rangle$. Although there are insufficient data points for a conclusive statement, the results suggest agreement with the surmise of Blount and Anderson that there is a phonon instability associated with the transformation. It is not clear from these data that there is a particular mode that is becoming unstable as we approach the transition. However, the results are certainly not in contradiction with this hypothesis. The results do indicate clearly that the fcc → fct transition is heralded by a gradual decrease in the mean phonon energy $\langle\omega\rangle$ from the general trend, certainly rein-

TABLE III. Comparison of λ determined from experiment and λ calculated from Eq. (11).

Alloy	λ_{calc} from (11)	λ_{expt}
In	0.99	0.834
$In_{0.9}Tl_{0.1}$	1.06	0.850
$In_{0.73}Tl_{0.27}$	1.21	0.933
$In_{0.67}Tl_{0.33}$	1.12	0.899
$In_{0.57}Tl_{0.43}$	1.20	0.847
$In_{0.50}Tl_{0.50}$	1.22	0.835
$In_{0.27}Tl_{0.73}$	1.50	1.092
$In_{0.17}Tl_{0.83}$	1.36	0.980
$In_{0.07}Tl_{0.43}$	1.39	0.889
Tl	1.34	0.780

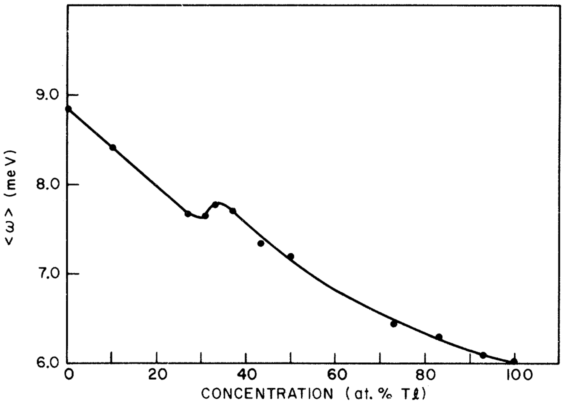

FIG. 9. Average phonon energy $\langle\omega\rangle$ as a function of alloy concentration.

forcing the suggestion that the transition is second order.

VI. CONCLUSIONS

A tunneling study of the Tl-In alloy system has been performed. An investigation of the T_c and Δ_0 of the various phases as well as μ^* and the product function $\alpha^2(\omega)F(\omega)$ have indicated that the various crystallographic phases have somewhat distinctive characteristics and that the bcc phase has the highest T_c of the system.

In a direct comparison with the existing expressions which allow calculation of the T_c from the electron-phonon coupling strength λ, the electron pseudopotential μ^*, and a characteristic phonon energy, the results are not very encouraging. In a simple metal alloy system such as Tl-In we find the results to be as much as 50% in error. These results leave one with the pessimistic attitude that aside from a first-principles calculation in which all the normal-state parameters are treated with extreme caution, a general expression for T_c to describe all superconductors may not be more accurate than this work indicates. It is possible that in view of these new data a modification of the existing expressions could be achieved. These data offer several variations of phonon spectra from almost crystalline to something approaching amorphous materials.

The spectra of the various phases display the characteristic fingerprints of their respective crystal structures. The fcc and fct systems have their typical single transverse and single longitudinal peaks, reminiscent of the well-known fcc spectra, while the bcc phase displays three distinct peaks in its spectrum. The hcp system has the most complicated distribution, containing a strong peak at low energies and various weaker critical points at higher energies.

In addition to this study of the various shapes of the spectra we have attempted to determine whether there exists an instability in one of the phonon modes in the region of the fcc → fct phase transition, thus supporting the idea of a second-order transition. Although nothing obvious as a function of concentration is observed, as there is no clear evidence of a single mode becoming unstable as the transition is approached it is felt that a study of the average phonon energy $\langle\omega\rangle$ supports the concept of an unstable mode. As the transition is approached, it apparently is signaled by a gradual tailing off of this $\langle\omega\rangle$ from its extrapolated value, reaching a maximum deviation at the transition into the indium phase.

ACKNOWLEDGMENTS

It is a pleasure to thank W. L. McMillan and J. M. Rowell for valuable discussions and suggestions throughout the course of this work. Also, the technical assistance of W. L. Feldmann is greatly appreciated.

---

[1] B. T. Matthias, in *Superconductivity*, edited by P. R. Wallace (Gordon and Breach, New York, 1969).

[2] J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. 108, 1175 (1957).

[3] J. R. Schrieffer, *Theory of Superconductivity* (Benjamin, New York, 1964); or D. J. Scalapino, in *Superconductivity*, edited by R. D. Parks (Marcel Dekker, New York, 1969).

[4] G. M. Eliashberg, Zh. Eksperim. i Teor. Fiz. 38, 996 (1960). [Soviet Phys. JETP 11, 696 (1960)].

[5] J. C. Swihart, D. J. Scalapino, and Y. Wada, Phys. Rev. Letters 14, 106 (1965).

[6] J. P. Carbotte and R. C. Dynes, Phys. Rev. 172, 476 (1968).

[7] P. B. Allen and M. L. Cohen, Phys. Rev. 187, 525 (1969).

[8] W. L. McMillan, Phys. Rev. 167, 331 (1968).

[9] P. Morel and P. W. Anderson, Phys. Rev. 125, 1263 (1962).

[10] R. J. Higgins, H. D. Kaehn, and J. H. Condon, Phys. Rev. 181, 1059 (1969).

[11] J. W. Garland and P. B. Allen, in International Conference on the Science of Superconductivity, Stanford, 1969 (unpublished).

[12] T. T. Chen, J. T. Chen, J. D. Leslie, and H. J. T. Smith, Phys. Rev. Letters 22, 526 (1969).

[13] H. Wühl, J. E. Jackson, and C. W. Briscoe, Phys. Rev. Letters 20, 1496 (1968).

[14] Myron Strongin, O. F. Kammerer, J. E. Crowe, R. D. Parks, D. H. Douglass, Jr., and M. A. Jensen, Phys. Rev. Letters 21, 1320 (1968).

[15] W. L. McMillan and J. M. Rowell, Phys. Rev.

Letters 19, 108 (1965).
[16]R. C. Dynes, J. P. Carbotte, D. W. Taylor, and C. K. Campbell, Phys. Rev. 178, 713 (1969).
[17]J. M. Rowell, W. L. McMillan, and W. L. Feldmann, Phys. Rev. 178, 896 (1969).
[18]W. L. McMillan and J. M. Rowell, in *Superconductivity*, Ref. 3.
[19]D. J. Scalapino, Y. Wada, and J. C. Swihart, Phys. Rev. Letters 14, 102 (1965).
[20]M. Hansen, *Constitution of Binary Alloys* (McGraw-Hill, New York, 1958).
[21]P. W. Anderson and E. J. Blount, Phys. Rev. Letters 14, 217 (1965).
[22]R. C. Dynes and J. M. Rowell, Phys. Rev. 187, 821 (1969).
[23]W. L. Feldmann and J. M. Rowell, J. Appl. Phys. 40, 312 (1969).
[24]J. M. Rowell, W. L. McMillan, and W. L. Feldmann, Phys. Rev. 180, 658 (1969).
[25]J. W. Stout and Lester Guttman, Phys. Rev. 88, 703 (1952).
[26]H. L. Luo and R. H. Willins, Phys. Rev. 154, 436 (1967).
[27]From W. A. Harrison, *Pseudopotentials in the Theory of Metals* (Benjamin, New York, 1966).
[28]B. W. Batterman and C. S. Barrett, Phys. Rev. Letters 13, 390 (1964).
[29]L. D. Landau and E. M. Lifshitz, *Statistical Physics* (Addison-Wesley, Reading, Mass., 1958).

PHYSICAL REVIEW B VOLUME 2, NUMBER 3 1 AUGUST 1970

Microwave-Photon-Assisted Tunneling in Sn-I-Sn Superconducting Tunnel Junctions

J. N. Sweet and G. I. Rochlin
Department of Physics, University of California,
and
Inorganic Materials Research Division, Lawrence Radiation Laboratory, Berkeley, California 94720

(Received 13 February 1970)

We have made an experimental study of comparatively low-frequency (3.93 GHz) microwave–photon-assisted quasiparticle tunneling in superconducting Sn-I-Sn tunnel junctions. The junctions were situated in a perpendicular rf electric field of frequency ω, with microwave voltages V_{rf} satisfying the condition $eV_{rf}/\hbar\omega \lesssim 18$. Excellent agreement with the rf power dependence predicted by the theory of Tien and Gordon has been obtained for junctions with normal resistances $\gtrsim 1\ \Omega$, although the calculated junction cavity fields remain an order of magnitude below field values needed to fit the data. As the junction resistance is decreased, agreement remains good at high rf power levels, but systematic discrepancies between theory and experiment occur at lower power levels. The interaction of microwave radiation with the zero-voltage Josephson current has also been studied on the same junctions, and the response compared to the theoretical predictions of Werthamer. In this case quantitative agreement with the theory is generally poor and does not appear to be correlated with sample resistance.

I. INTRODUCTION

The quasiparticle tunneling currents which flow through an insulating layer between two superconductors can be profoundly altered when time-varying electromagnetic fields are present in or near the barrier region. The exact form of the modified quasiparticle tunneling characteristic depends on the applied microwave frequency ω and the quantity $\alpha \equiv eV_{rf}/\hbar\omega$, where V_{rf} is the magnitude of the effective microwave voltage appearing across the oxide barrier. This inelastic process may be thought of as photon-assisted tunneling, in which the quasiparticles absorb or emit one or more photons while tunneling through the insulating layer. For a junction composed of two identical superconductors, the tunneling current will in general be increased for applied dc bias voltages $V < 2\Delta/e$ and decreased for $V > 2\Delta/e$, where 2Δ equals the superconducting energy gap. The exact form of the modified current for this system at a given temperature depends only on the parameters ω and α.

We have made a series of detailed photon-assisted-tunneling measurements utilizing comparatively low-frequency (3.93 GHz) microwaves and high-quality Sn-SnO-Sn junctions. Our results are in good agreement with the theoretical predictions of Tien and Gordon[1] when junction resistances are $\gtrsim 1\,\Omega$. Using a single adjustable parameter to scale the rf power, we are able to construct an excellent detailed fit to the theoretical power dependence of the tunneling current as a function of bias for experimentally determined values of α as large as 18. Systematic deviations from the theory,