

<sup>10</sup>A. K. Niessen and F. A. Staas, *Phys. Letters* **15**, 26 (1965).

<sup>11</sup>J. Bardeen and M. J. Stephen, *Phys. Rev.* **140**, A1197 (1965).

<sup>12</sup>W. S. Chow, *Phys. Rev. B* **1**, 2130 (1970).

<sup>13</sup>C. Caroli and K. Maki, *Phys. Rev.* **164**, 591 (1967).

<sup>14</sup>We point out that I. M. Tang [*Phys. Rev. B* **2**, 1299 (1970)] has misquoted our Eqs. (4) and (5). For details, see Ref. 3.

<sup>15</sup>D. Markowitz and L. P. Kadanoff, *Phys. Rev.* **131**, 563 (1963).

<sup>16</sup>P. Nozières and W. F. Vinen, *Phil. Mag.* **14**, 667 (1966).

<sup>17</sup>W. S. Chow, *Phys. Rev.* **188**, 783 (1969).

<sup>18</sup>W. A. Reed, E. Fawcett, and Y. B. Kim, *Phys. Rev. Letters* **14**, 790 (1965).

<sup>19</sup>A. T. Fiory and B. Serin, *Phys. Rev. Letters* **19**, 227 (1967).

PHYSICAL REVIEW B

VOLUME 5, NUMBER 11

1 JUNE 1972

## Electron-Phonon Interaction and Superconductivity in In-Tl Alloys

D. W. Taylor\*

*Physics Department, McMaster University, Hamilton-16, Canada*

and

P. Vashishta†

*Argonne National Laboratory, Argonne, Illinois 60439*

(Received 15 December 1971)

The superconducting tunneling results obtained by Dynes for the electron-phonon part  $\alpha^2(\omega)F(\omega)$  for indium-thallium alloys are examined in terms of both exact sum rules for alloy lattice dynamics and the self-consistent mass-defect theory. Detailed calculations are made of  $\alpha^2(\omega)F(\omega)$  for four alloy concentrations. Good over-all agreement is found between calculated and tunneling-derived results. Our analysis seems to support Dynes's conclusion that the phase transition in the alloy structure near 30-at.% thallium could be due to phonon-mode softening. Zero-temperature Eliashberg equations are solved using the calculated and tunneling-derived values of  $\alpha^2(\omega)F(\omega)$  to determine the gap and renormalization functions. Reduction in the dc Josephson current and condensation energies from their BCS value are calculated. Transition temperatures for the alloys are also calculated using the two sets of  $\alpha^2(\omega)F(\omega)$ . The agreement with the experiment is satisfactory.

### I. INTRODUCTION

Within the very sophisticated and accurate formulation of the pairing theory, the strong-coupling theory of superconductivity due to Eliashberg,<sup>1</sup> Nambu,<sup>2</sup> Gorkov,<sup>3</sup> Scalapino, Schrieffer, and Wilkins,<sup>4</sup> a superconductor in the "dirty" isotropic limit is completely characterized by a complex energy-dependent generalization  $\Delta(\omega)$  of the BCS energy gap and a renormalization function  $Z(\omega)$ . The central result of the strong-coupling theory is a set of nonlinear integral equations, the Eliashberg gap equations, which relate  $\Delta(\omega)$  and  $Z(\omega)$  to certain properties of the metal in its normal state. The equations need to be solved numerically because of their complexity. The problem of calculating the properties of a superconductor therefore reduces to (i) calculation of the kernels of the gap equations, and (ii) solution of the gap equations to determine the gap and renormalization functions which in turn determine the superconducting properties of the material. The kernels of the gap equations refer separately to the phonon-mediated part of the interaction between two electrons, denoted by  $\alpha^2(\omega)F(\omega)$ , and the basic Coulomb repulsion between them, denoted by  $\mu^*$ .  $\alpha(\omega)$  is a

measure of the electron-phonon coupling and  $F(\omega)$  is the phonon density of states.

It is widely accepted that the reason for unavailability of accurate calculations, for example, of the superconducting transition temperature  $T_c$  lies not in any basic failing of the superconductivity theory but rather in the fact that  $T_c$  depends sensitively on the normal-state properties of the material, which at present cannot be calculated to a very high degree of accuracy starting from fundamentals.

Using a model for the phonon density of states in Pb, Swihart, Scalapino, and Wada<sup>5</sup> performed calculations of the superconducting properties of Pb using the Eliashberg formulation. Their results agreed reasonably well with experiment.

A source of information on the normal-state parameters entering Eliashberg kernels is provided by quasiparticle-tunneling experiments. An image of the phonon density of states exists in the current-voltage characteristics of the tunnel junction. McMillan and Rowell<sup>6</sup> have devised a computer program which inverts the zero-temperature Eliashberg equations using the experimental tunneling density of states and determines, quite accurately, the functions  $\alpha^2(\omega)F(\omega)$  and  $\mu^*$  which

enter the Eliashberg kernels. Theoretically calculated Eliashberg kernels should, of course, agree with the kernels derived from tunneling data. Vashishta and Carbotte<sup>7</sup> have performed extensive calculations of the superconducting properties of several strong-coupling materials, including amorphous Bi and Ga at zero and finite temperatures using the tunneling-derived normal-state data. Their results are in good agreement with experiments,<sup>8,9</sup> confirming the accuracy of the Eliashberg equations at finite temperatures if the tunneling-derived kernels are used.

In recent years much has been added to our understanding of the normal-state properties of simple metals. A reliable calculation of the relevant normal-state properties to determine the superconducting properties can now be attempted. For the success of such a calculation it will, of course, be helpful to have results for the normal-state properties derived from quasiparticle tunneling. These can serve as a check on the accuracy of the calculations. Recent calculations of superconducting properties, mainly  $T_c$ , due to Seiden,<sup>10</sup> Carbotte and Dynes,<sup>11</sup> and Allen and Cohen<sup>12</sup> have been reasonably successful.

Recently Dynes<sup>13</sup> has made an experimental study of In-Tl alloys by the quasiparticle-tunneling technique. From the tunneling data he has extracted the functions  $\alpha^2(\omega)F(\omega)$  and  $\mu^*$  for the alloys. He has used  $\alpha^2(\omega)F(\omega)$  to discuss the possibility of phonon-mode softening near the phase transition in the alloy structure from face-centered tetragonal (fct) to face-centered cubic (fcc) at around 30-at. % thallium concentration. In particular he has used the expressions for the frequency moments of  $\alpha^2(\omega)F(\omega)$  in the alloys suggested by results for pure materials. Furthermore, he has made a detailed comparison between the  $T_c$  calculated from the expressions due to McMillan<sup>14</sup> and Garland and Allen<sup>15</sup> and the experimentally measured values.

Besides the superconductivity theory outlined above, there should enter into a discussion of these results the available theory of the dynamics of disordered crystals. On examining the frequency moments of  $\alpha^2(\omega)F(\omega)$  employed by Dynes, but using an expression for  $\alpha^2(\omega)F(\omega)$  applicable to alloys, it becomes clear that only the frequency moments of the lattice Green's functions are involved. In Sec. II we derive exact expressions for the latter moments (i. e., sum rules) which suggest a slightly different analysis than that given by Dynes. One of the sum rules can be applied exactly as far as the lattice dynamics is concerned and we use it to extract information on the alloy Fermi-surface density of states.

With regard to the detailed lattice dynamics of an alloy we have used the approximate self-consistent theory of Taylor<sup>16</sup> that is only applicable

for mass defects. This is essentially a procedure that describes the configuration-averaged alloy by an effective Green's function so defined that there is zero scattering produced by the departure from translational invariance. In the simplest approximation, which is the only one so far evaluated in detail,<sup>16</sup> this amounts to setting the average single-site scattering to zero. So, besides the omission of force-constant changes associated with the impurities, these calculations omit effects arising from clusters of impurities whether they occur in a random manner or are due to short-range order. The theory is applied in Sec. III to the In-Tl system in the nominally fcc region. (Beyond about 55-at. % thallium the structure is initially a mixture of phases and finally hexagonal close packed.<sup>17</sup>) While the omission of force-constant changes rules out the possibility of describing in any detail the fcc/fct phase transition, the fact that indium and thallium have the same valence suggests that this omission should not prevent our calculations from giving a reasonable over-all description of the behavior of  $\alpha^2(\omega)F(\omega)$  as a function of concentration.

In Sec. IV we turn to the solution of the zero-temperature Eliashberg gap equations, using both the experimental and calculated values for  $\alpha^2(\omega) \times F(\omega)$ . With the resulting gap and renormalization functions for the superconducting and normal states we calculate the reductions in the dc Josephson current and the condensation energy from their BCS values. The transition temperature  $T_c$  of the alloys is calculated for both sets of  $\alpha^2(\omega)F(\omega)$  using the simple equations due to Leavens and Carbotte.<sup>18</sup> A comparison is then made with the results for  $T_c$  obtained from the equations due to McMillan and to Garland and Allen. Our conclusions are drawn together in Sec. V.

## II. SUM RULES

The details of the lattice vibrations and the coupling of these vibrations to the electrons enter the strong-coupling superconductivity theory of Eliashberg via the function  $\alpha^2(\omega)F(\omega)$ .<sup>19</sup> For an alloy it can be written as<sup>6</sup>

$$\alpha^2(\omega)F(\omega) = \frac{N(0)}{8\pi^2 k_F^2} \int_{q \leq 2k_F} \frac{d^3 q}{q} \frac{1}{N^2} \sum_{i i'} \sum_{\alpha \beta} w_i^*(q) \times q_{\alpha} \text{Im} G_{\alpha \beta}(l, l'; \omega - i\delta) q_{\beta} w_{i'}(q) e^{-i\vec{q} \cdot (\vec{R}_l - \vec{R}_{l'})}, \quad (1)$$

where  $w_i(q)$  is the pseudopotential form factor appropriate to the kind of ion at the site  $\vec{R}_i$ . The Green's function in (1) is given by

$$G_{\alpha \beta}(l, l'; \omega) = \frac{1}{\hbar} \int_{-\infty}^{\infty} d\omega' e^{i\omega t} \langle\langle u_{\alpha}(l, t); u_{\beta}(l', 0) \rangle\rangle, \quad (2)$$

where  $\langle\langle \rangle\rangle$  indicates a double-time Green's function,

as defined by Zubarev.<sup>20</sup>  $u_\alpha(l, t)$  is the Heisenberg operator for a lattice displacement at  $\vec{R}_l$ , with  $\alpha$  the Cartesian coordinate.

Our expression for  $\alpha^2(\omega)F(\omega)$  refers to a given configuration of ions, i. e., no configuration averaging has been performed. It should also be noted that explicit effects on the electron states due to alloying have been ignored, although to some extent they could be considered to be included in the screening of the form factors and in the band-structure density of states at the Fermi surface,  $N(0)$ . It can be seen from (1) that to discuss the frequency moments of  $\alpha^2(\omega)F(\omega)$  we need only examine those for  $\text{Im}\hat{G}_{\alpha\beta}$ .

The first moment

$$\mu_{\alpha\beta}^{(1)}(l, l') = \int_0^\infty \omega \text{Im}G_{\alpha\beta}(l, l'; \omega - i\delta) d\omega \quad (3)$$

is just an  $f$  sum rule and an expression for it can be derived in the usual way.<sup>21</sup> The central identity involved is

$$[u_\alpha(l), [u_\beta(l'), H]] = \frac{-\hbar^2}{M(l)} \delta_{\alpha\beta} \delta_{ll'}, \quad (4)$$

where  $M(l)$  is the mass of the atom at  $\vec{R}_l$ . On taking a thermal average of both sides of (4), the left-hand side becomes related to  $\text{Im}G_{\alpha\beta}$ , and we obtain

$$\mu_{\alpha\beta}^{(1)}(l, l') = \frac{\pi}{2M(l)} \delta_{\alpha\beta} \delta_{ll'}. \quad (5)$$

This result is exact, as is the following sum rule [see (10)], irrespective of short-range order, anharmonicity, structure, and force-constant changes.

The first moment of  $\alpha^2(\omega)F(\omega)$  then becomes

$$\alpha^{(1)} = \int_0^\infty \omega \alpha^2(\omega)F(\omega) d\omega = \frac{N(0)}{4k_F^3 N^2} \sum_i \frac{\langle w_i^2 \rangle}{M(l)}, \quad (6)$$

with

$$\langle w_i^2 \rangle = \int_{q \leq 2q_F} q^3 |w_i(q)|^2 dq.$$

Even neglecting the difference in form factors between the two atoms, we see that it is the mean inverse mass  $\langle M^{-1} \rangle$ , rather than the inverse mean mass of Dynes,<sup>13</sup> that enters  $\alpha^{(1)}$ . Although, as Dynes indicates,  $\alpha^{(1)}$  does not contain information on the lattice vibrations, it does give information on the band-structure density of states and the form factors.

Taking available values for the bare pseudopotentials we can calculate the right-hand side of (6) using a free-electron density of states  $N(0)_{\text{FE}}$ . Then, from the experimental values of  $\alpha^{(1)}$ , values of  $N(0)$  can be obtained for the alloy system.<sup>11</sup> For the pure metals the pseudopotentials can be checked by comparing the calculated values of  $N(0)$  with those obtained from the specific heat  $\gamma$ .<sup>14</sup> For indium, using the Ashcroft potential with  $R_c$

= 0.575 Å,<sup>22</sup> and screening according to Singwi, Sjolander, Tosi, and Land,<sup>23</sup> we find  $N(0)/N(0)_{\text{FE}} = 0.701$ . This is in fair agreement with the value of 0.745 taken from the specific heat<sup>24</sup>  $\gamma$  suitably adjusted for the electron-phonon enhancement,  $1 + \lambda$ , using the results of Dynes<sup>13</sup> (see below).<sup>25</sup> Very close agreement is not to be expected as we have not taken into account departures from the  $N(0)_{\text{FE}}$  in calculating the screening. For thallium we have used the Heine-Abarenkov model<sup>26</sup> to obtain  $N(0)/N(0)_{\text{FE}} = 0.580$  compared with 0.640 obtained via<sup>27</sup>  $\gamma$  and  $\lambda$ .<sup>13</sup>

On using the experimental variation of volume with concentration<sup>28</sup> we find the results for  $N(0)/N(0)_{\text{FE}}$  given in Fig. 1. These results are not very different from those of Dynes for his quantity  $N(0)\langle v^2 \rangle$ , though we have taken into account in a more accurate way the mass and pseudopotential differences between indium and thallium. The small apparent jump in the region of the phase transition from fct to fcc is to be noted. It would be of interest to have experimental values for  $\gamma$  to examine this effect and to test the above manipulations.

The situation is different for the first inverse moment

$$\mu_{\alpha\beta}^{(-1)}(l, l') = \int_0^\infty \frac{d\omega}{\omega} \text{Im}G_{\alpha\beta}(l, l'; \omega - i\delta). \quad (7)$$

We can relate  $\mu_{\alpha\beta}^{(-1)}$  to  $\mu_{\alpha\beta}^{(1)}$  using the equation of motion for  $\underline{G}$ ,<sup>29</sup>

$$\omega^2 M(l) G_{\alpha\beta}(l, l'; \omega) = \delta_{\alpha\beta} \delta_{ll'} + \sum_{\gamma l_1} \Phi_{\alpha\gamma}(l, l_1) \times G_{\alpha\beta}(l_1, l'; \omega), \quad (8)$$

where  $\Phi$  is the force-constant matrix. On multiplying on the left-hand side by  $\Phi^{-1}$ , dividing by  $\omega$ , and then taking the discontinuity across the real axis of the resulting equation, we find

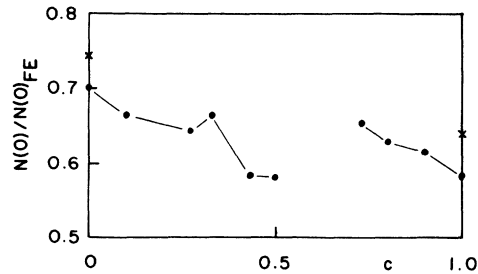


FIG. 1. Calculated ratio of band structure to free-electron Fermi-surface density of states,  $N(0)/N(0)_{\text{FE}}$ , as a function of thallium concentration  $c$ .  $\times$  indicates values obtained from specific heat  $\gamma$  for In (Ref. 24) and Tl (Ref. 27).

$$\sum_{l_1} \Phi_{\alpha\gamma}^{-1}(l, l_1) M_\gamma(l_1) \omega \operatorname{Im} G_{\gamma\beta}(l_1, l'; \omega) = -\pi \delta(\omega) \\ \times [\Phi_{\alpha\beta}^{-1}(l, l') + \operatorname{Re} G_{\alpha\beta}(l, l'; \omega)] + \frac{1}{\omega} \operatorname{Im} G_{\alpha\beta}(l, l'; \omega). \quad (9)$$

Setting  $\omega = 0$  in (8) shows that the first term on the right-hand side of (9) is zero. We obtain, finally,

$$\mu_{\alpha\beta}^{(-1)}(l, l') = \frac{1}{2} \pi \Phi_{\alpha\beta}^{-1}(l, l'). \quad (10)$$

This moment does contain information on the lattice dynamics but not in as clear a form as might be desired. Unfortunately, it is the trace of the inverse of the reduced-force-constant matrix  $[M(l)M(l')]^{-1/2} \Phi_{\alpha\beta}(l, l')$  that gives the second inverse-frequency moment of the phonon density of states. Only in a monatomic system does  $\operatorname{Tr} \mu^{(-1)}$  give this moment. However, it seems very reasonable to conclude that  $\mu^{(-1)}$  should tend to peak when there is phonon-mode softening.

As the first inverse moment of  $\alpha^2(\omega)F(\omega)$  is one-half the electron-phonon-coupling constant<sup>14</sup>  $\lambda$ , we quote the expression for the latter parameter arising from (10):

$$\lambda = \frac{N(0)}{8\pi k_F^2} \int_{q \leq 2k_F} \frac{d^3q}{q} \frac{1}{N^2} \sum_{l, l'} \sum_{\alpha\beta} w_l^*(q) q_\alpha \Phi_{\alpha\beta}^{-1}(l, l') \\ \times q_\beta w_{l'}(q) e^{-i\vec{q} \cdot (\vec{R}_l - \vec{R}_{l'})}. \quad (11)$$

Unfortunately it appears that this expression cannot be simplified unless we neglect both the differences in the form factors of the different atoms and the variations in force constants. Then we regain the familiar result<sup>14</sup>

$$\lambda = \frac{N(0)}{8\pi MNk_F^2} \sum_j \int \frac{d^3q}{q} \frac{|w(q)\vec{q} \cdot \vec{\sigma}^j(\vec{q})|^2}{w_j^2(q)}, \quad (12)$$

where  $\omega_j(\vec{q})$  and  $\vec{\sigma}^j(\vec{q})$  are the eigenfrequency and eigenvector, respectively, for the phonon mode  $(j, \vec{q})$  in the host material. This result indicates that if only the mass differences are taken into account,  $\lambda$  remains constant through the alloy system since  $M$  in (12) is the host atomic mass. It appears that we must rely on (11) to suggest that in an alloy  $\lambda$ , as given by (11), should tend to peak when a phonon mode softens.

Unlike (6), (11) is not calculable at present as it requires both a model for the lattice dynamics and a knowledge of the atomic configuration of the alloy.

If we neglect the difference in form factors for the different atoms we note that, aside from the exponential factor in (11), it is the same  $q$  moment of the form factors that enters both (6) and (11). This suggests that we use the experimental value for  $\alpha^{(1)}$  to approximately remove from  $\lambda$  the changes in the electronic factors. This can be

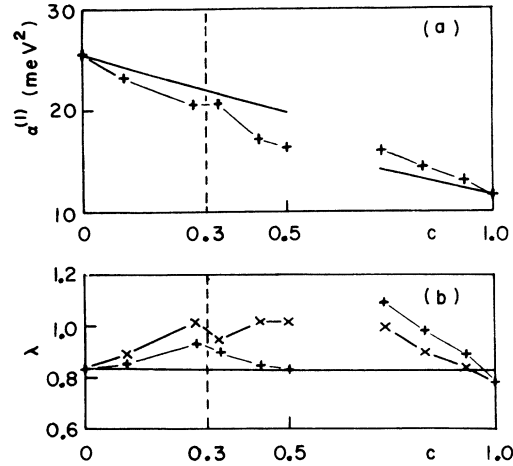


FIG. 2. Frequency moments of  $\alpha^2(\omega)F(\omega)$  as a function of thallium concentration  $c$ . Solid line is calculated assuming only mass difference; + experiment;  $\times$  adjusted experiment [see text after Eq. (12)]. (a) First moment  $\alpha^{(1)}$ ; (b) inverse moment or electron-phonon-coupling parameter  $\lambda$ .

done by dividing  $\lambda$  by the ratio of the experimental  $\alpha^{(1)}$  [ + in Fig. 2(a)] to that obtained by taking into account just the mass change [solid line in Fig. 2(a)]. The result is shown in Fig. 2(b), where the experimental values of  $\lambda$  (+) are compared to those adjusted in the above manner ( $\times$ ). We think this is preferable to examining  $\lambda$  normalized by the area under  $\alpha^2(\omega)F(\omega)$  ( $\langle\omega\rangle^{-1}$  of Dynes) because  $\alpha^{(1)}$  does not contain any information about mode softening due to force-constant changes.

The behavior of the adjusted  $\lambda$  as a function of thallium concentration contains a sharp increase on crossing the critical thallium concentration ( $c \approx 0.3$ ) from the fcc phase ( $c > 0.3$ ) to the fct phase ( $c < 0.3$ ). This indicates a decrease in phonon frequencies and may be attributable to some of the phonon modes becoming very soft, heralding the instability responsible for the transition. However, this evidence for mode softening obtained from our analysis of the experimental data is not significantly different, in numerical sense, from that presented by Dynes using  $\langle\omega\rangle$  (Fig. 9, Ref. 13).

The general trend of  $\lambda$  adjusted as  $c$  increases,  $c < 0.5$ , does indicate a general reduction in phonon frequencies that can be attributed to a general reduction in force constants according to (11). A similar reduction is indicated in Fig. 2(b) as indium is added to thallium in the bcc, hcp region ( $0.7 \lesssim c \leq 1.0$ ). This is quite different from the behavior of  $\langle\omega\rangle$  which, except at the fct/fcc phase transition, decreases monotonically with thallium concentration. We suspect that this is partly a consequence of dividing two quantities both con-

taining information about the phonon-frequency distribution.

### III. DETAILED CALCULATION OF ALLOY

$$\alpha^2(\omega)F(\omega)$$

An examination of the experimental  $\alpha^2(\omega)F(\omega)$  near the fct/fcc phase transition reveals no spectacular evidence of mode softening. It is of interest to have a reference with which to compare these results in order to obtain further information on possible mode softening. Such a reference can be obtained by calculating the alloy lattice dynamics taking into account just the mass difference, as this is not expected to induce a phase transition.

We have used the self-consistent theory for random alloys<sup>16</sup> to do this, working with indium as the host material and so treating thallium as the impurity. As mentioned in Sec. I this involves setting to zero the average single-site scattering in the effective crystal. The consequent neglect of effects due to clusters of impurities or, equivalently, the variations in the local environment of an impurity atom should not be too important as we are concerned only with the region in which the heavier atom (thallium) has fractional concentration  $c < 0.5$ . Comparison with machine calculations has shown that the theory is quite accurate in these circumstances.<sup>16</sup> It is the neglect of force-constant changes and the use of a theory developed for a cubic system that are the more serious approximations involved in our calculations.

As only  $G_{\alpha\alpha}(l, l; \omega)$  is involved in calculating the scattering described above we only need the indium phonon density of states  $F(\omega)$ . We have extracted this from the experimental  $\alpha^2(\omega)F(\omega)$  in the following manner. Although theoretical pseudopotential attempts to calculate  $F(\omega)$  have not been too successful,<sup>30</sup> we have used them to estimate  $\alpha^2(\omega)_{\text{cal}}$ , defined by

$$\alpha^2(\omega)_{\text{cal}} = \alpha^2(\omega)F(\omega)_{\text{cal}}/F(\omega)_{\text{cal}}, \quad (13)$$

and then obtained

$$F(\omega) = \alpha^2(\omega)F(\omega)_{\text{expt}}/\alpha^2(\omega)_{\text{cal}}. \quad (14)$$

As the theoretical discrepancy is mainly an overestimate of the transverse frequencies, we have increased  $\alpha^2(\omega)_{\text{cal}}$  in this region by about 20% and then only an over-all scaling of 5% is required to obtain a correctly normalized  $F(\omega)$ .

We can transform the Green's function in (1) using the host-crystal eigenvectors to obtain

$$G^j(\vec{q}, \omega) = \frac{M}{N} \sum_{\alpha, \beta} [\sigma_{\alpha}^j(\vec{q})]^* G_{\alpha\beta}(l, l'; \omega) \sigma_{\beta}^j(\vec{q}) e^{-i\vec{q} \cdot (\vec{R}_l - \vec{R}_{l'})}. \quad (15)$$

In our approximation for the alloy lattice dynamics the Green's function given in (15) depends on  $(j, \vec{q})$  only through  $\omega_j(\vec{q})$ , and so we can write

$$G^j(\vec{q}, \omega) = G(\omega_j(\vec{q}), \omega). \quad (16)$$

Then, on neglecting the variation in form factors,  $\alpha^2(\omega)F(\omega)$  from (1) becomes

$$\begin{aligned} \alpha^2(\omega)F(\omega)_{\text{allor}} &= \frac{N(0)}{8\pi^2 k_F^2 N} \int_{q \leq 2k_F} \frac{d^3 q}{q} |w(q)|^2 \\ &\quad \times |\vec{q} \cdot \vec{\sigma}^j(\vec{q})|^2 \text{Im}G(\omega_j(\vec{q}), \omega - i\delta) \\ &= \frac{2}{\pi} \int d\omega' \alpha^2(\omega')_{\text{host}} \omega' \text{Im}G(\omega', \omega). \end{aligned} \quad (17)$$

The properties of the spectral function  $\text{Im}G(\omega', \omega)$  have been discussed in detail by Taylor<sup>16</sup> for the case of a very heavy impurity with a mass ratio of 3.1:1. In the present case the thallium/indium mass ratio is 1.7:1 and is insufficient to produce any strong resonance effects. Consequently the spectral function does not have any of the strong features found by Taylor, such as the occurrence of two peaks in the region of a resonance. Rather, it always has a single peak although it is sometimes rather asymmetric.

We have compared our calculations of (17) with experiment for various thallium concentrations  $c$ . In Fig. 3 we give the results for  $c = 0.27, 0.33$ , and  $0.50$ , comparing experiment with (17) and with (17) adjusted for changes in  $N(0)$ , as suggested by Fig. 1. The results for indium are given as a reference.

Although in Sec. II we adjusted  $\lambda$  to take into account variations in both the form factors and the electron density of states we have not included the former in Fig. 3 as we are now examining the frequency dependence  $\alpha^2(\omega)F(\omega)$ . The indium and thallium form factors are not that different, so it is the deviations of  $N(0)$  from  $N(0)_{\text{FE}}$  that dominate the over-all scaling. Because the low-frequency region will tend to be dominated by the thallium motion, the main consequence of the difference in form factors will be a frequency-dependent scaling. Detailed calculations, equivalent to the inclusion of different scattering lengths in neutron-scattering cross sections,<sup>16,29</sup> would be required to test this effect.

When compared to the indium  $\alpha^2(\omega)F(\omega)$  it can be seen in Fig. 3 that the calculated  $\alpha^2(\omega)F(\omega)$  for alloys is in reasonable accord with the experimental results, particularly when adjusted for variations in  $N(0)$ . In the longitudinal region the only discrepancy is the location of the peak and this can be taken as evidence for reductions in the force constants. Such a reduction would enhance the tendency to form resonant modes in the transverse region. Detailed calculations for low concentrations of gold in copper definitely indicate this tendency.<sup>31</sup> Thus the omission of these changes in our calculations could be the major reason for the large discrepancy

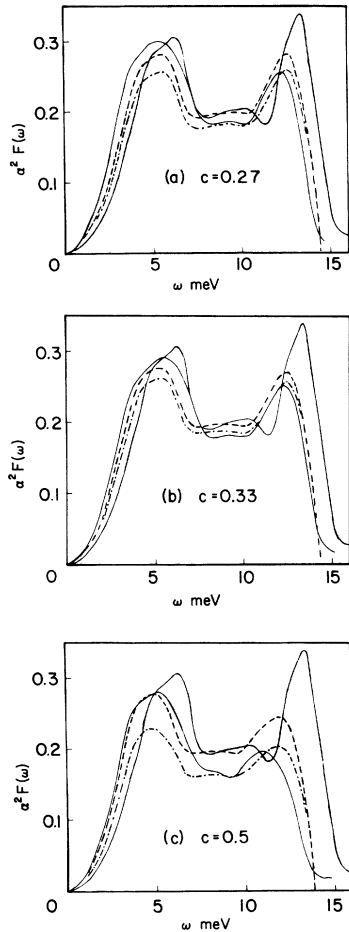


FIG. 3. Comparison of  $\alpha^2(\omega)F(\omega)$  from experiment (solid line), mass-defect theory (dashed line), and adjusted mass-defect theory (dash-dot line) for thallium concentrations  $c$ ; (a)  $c = 0.27$ , (b)  $c = 0.33$ , (c)  $c = 0.50$ . Indium  $\alpha^2(\omega)F(\omega)$  is also shown by continuous curve; it is the one with highest peak around 13 meV.

in the magnitude of  $\alpha^2(\omega)F(\omega)$  in the transverse region.

In order to see whether the calculations contain any information on mode softening at the phase transition the discrepancies between calculation and experiment should be compared for  $c = 0.27$ ,  $0.33$ . Besides the large discrepancy for the strength of the transverse region for  $c = 0.27$  there is also a greater discrepancy in the position of the peaks for this concentration. Both these effects produce an enhancement in  $\lambda$ . The first effect is due to a redistribution of modes towards lower frequencies, while the second largely involves a uniform shift.

However, we see that even using the calculated  $\alpha^2(\omega)F(\omega)$  as a reference there is no clear-cut evidence of specific mode softening, although this may be contained in the mode redistribution men-

tioned above. It should be noted that although we find that phonons described by  $(j, \vec{q})$  have widths  $\Delta\omega/\omega$  of about 6–10% for  $c = 0.3$ , we can still talk about normal modes and their softening for a harmonic Hamiltonian. It is just that these modes are not described by  $(j, \vec{q})$ .

#### IV. CALCULATION OF SUPERCONDUCTING PROPERTIES

We have seen in Sec. III that the phonon part calculated from (17),  $[\alpha^2(\omega)F(\omega)]_{\text{cal}}$ , compares satisfactorily with  $[\alpha^2(\omega)F(\omega)]_{\text{tun}}$ , the tunneling-derived phonon part due to Dynes.<sup>13</sup> The Eliashberg formulation of the pairing theory provides us with the necessary framework for the calculation of the superconducting properties starting from the relevant normal-state data. For a derivation and discussion of the gap equations we refer the reader to the review article by Scalapino.<sup>32</sup> We have solved the zero-temperature Eliashberg equations using  $(\alpha^2 F)_{\text{cal}}$  and  $(\alpha^2 F)_{\text{tun}}$  as the normal-state data. It should be noted that throughout this section we use the unadjusted  $(\alpha^2 F)_{\text{cal}}$  obtained directly from (17). One could in fact proceed further to solve the Eliashberg equations at various temperatures and determine the temperature-dependent gap and renormalization functions  $\Delta(\omega, T)$  and  $Z(\omega, T)$ , which could in turn be used to study the thermodynamic properties of the alloys. The transition temperature can simply be determined by plotting the square of the calculated gap against temperature, which gives a straight line near the transition temperature, and by finding out the intersection by extrapolating. It is possible to carry out such a calculation, but in terms of computer time it is very expensive. Furthermore, unlike the strong-coupling materials, e.g., Pb, Hg, Pb<sub>0.9</sub>, Bi<sub>0.1</sub>, amorphous Bi, Ga, etc., for which the deviations from BCS predictions are large, for the intermediate-coupling materials like In, Tl, and their alloys the deviations from BCS predictions are not very pronounced. For the above reasons we feel that a full program of solving the finite-temperature Eliashberg equations for In-Tl alloys is not worthwhile. We shall solve only the zero-temperature Eliashberg equations.

Our computer program for solving the Eliashberg equations at  $T = 0$  accepts numerical data for  $\alpha^2(\omega)F(\omega)$ . We have used a 45-point mesh in the interval  $(0, \omega_D)$ , where  $\omega_D$  is the maximum phonon frequency in  $\alpha^2(\omega)F(\omega)$ . For a particular value of the upper cutoff  $\omega_c = 70$  meV ( $\omega_c \approx 5\omega_D$ ) in the Eliashberg equations, the value of the Coulomb part was adjusted to  $\mu_E^*$  such that the calculated gap  $\Delta_0$  was equal to the experimental value  $\Delta_0^{\text{exp}} = \Delta_0 = \text{Re}\Delta(\Delta_0)$ . Relevant experimental data about the alloys are given in Table I. The values of  $\mu_E^*$  are different for  $(\alpha^2 F)_{\text{cal}}$  ( $\mu_E^* = 0.140, 0.112, 0.122, 0.169$  for  $c = 0.10, 0.27, 0.33, 0.50$ , respectively) and  $(\alpha^2 F)_{\text{tun}}$  ( $\mu_E^* = 0.137, 0.133, 0.137, 0.138, 0.142, 0.127$  for

TABLE I. Values of the energy gap and transition temperature from experiment. Transition temperatures calculated from  $(\alpha^2 F)_{\text{tun}}$  using the equations due to McMillan and to Garland and Allen, and calculated from  $(\alpha^2 F)_{\text{tun}}$  and  $(\alpha^2 F)_{\text{cal}}$  using Leavens-Carbotte equation.

Alloy	$\Delta_0^{\text{Expt}^b}$ (meV)	$T_c$ calculated using $(\alpha^2 F)_{\text{tun}}$			$T_c$ from Leavens and Carbotte, Eq. (25) $\mu_{LC}^*$ (alloy) = 0.10492 <sup>a</sup>	
		$T_c^{\text{Expt}^b}$ (K)	McMillan <sup>b</sup> (K)	Garland-Allen <sup>b</sup> (K)	Calculated using $(\alpha^2 F)_{\text{tun}}$ (K)	Calculated using $(\alpha^2 F)_{\text{cal}}$ from (17) (K)
In	0.5400	3.40	3.44	2.57	3.581	...
In <sub>0.90</sub> Tl <sub>0.10</sub>	0.5300	3.28	3.42	2.40	3.454	3.494
In <sub>0.73</sub> Tl <sub>0.27</sub>	0.5700	3.36	3.60	2.47	3.584	3.317
In <sub>0.67</sub> Tl <sub>0.33</sub>	0.5360	3.26	3.42	2.21	3.464	3.267
In <sub>0.50</sub> Tl <sub>0.50</sub>	0.4110	2.52	2.58	1.57	2.857	3.119
Tl	0.3690	2.33	2.10	1.41	2.363	...

$$^a \mu_{LC}^* (\text{alloy}) = \frac{1}{2} [\mu_{LC}^* (\text{In}) + \mu_{LC}^* (\text{Tl})].$$

<sup>b</sup>From Ref. 13.

$c = 0.00, 0.10, 0.27, 0.33, 0.50, 1.00$ , respectively) because for the two sets of  $\alpha^2 F$  the value of the calculated gap is equal to the experimental gap. The differences in  $(\alpha^2 F)_{\text{cal}}$  and  $(\alpha^2 F)_{\text{tun}}$  are reflected in their respective  $\mu_E^*$  values. Our values of  $\mu_E^*$  pertaining to  $(\alpha^2 F)_{\text{tun}}$  compare quite well with those of Dynes ( $\mu_E^* = 0.125, 0.122, 0.126, 0.127, 0.133, 0.127$  for  $c = 0.00, 0.10, 0.27, 0.33, 0.50, 1.00$ , respectively). Slight differences between Dynes's and our  $\mu_E^*$  values from  $(\alpha^2 F)_{\text{tun}}$  are due to different mesh sizes and cutoff values  $\omega_c$ .

The results for the real and imaginary parts of the gap function  $\Delta(\omega)$  and the renormalization functions  $Z_S(\omega)$  and  $Z_N(\omega)$  for the superconducting and normal states are shown in Figs. 4(a)–4(c), respectively, for In<sub>0.73</sub>Tl<sub>0.27</sub>. The continuous and broken curves refer to the solutions obtained by using  $(\alpha^2 F)_{\text{tun}}$  and  $(\alpha^2 F)_{\text{cal}}$ , respectively. In Fig. 4(a) the two sets of curves for  $\Delta_1$  and  $\Delta_2$  compare quite well. The continuous and broken curves for  $\Delta_1$  join together at small  $\omega$ . This is because the calculated gap  $\Delta_0$  is equal to  $\Delta_0^{\text{Expt}}$  for both sets of  $\alpha^2 F$ . In Figs. 4(b) and 4(c) the values of  $Z_{1S}(0)$  and  $Z_{1N}(0)$  are different for the continuous and broken curves. This can be understood by noting that  $Z_{1N}(0)$  is related to the strength of electron-phonon coupling  $\lambda$  according to

$$Z_{1N}(0) = Z_N(0) = 1 + \lambda = 1 + 2 \int \frac{\alpha^2(\omega)F(\omega)}{\omega} d\omega, \quad (18)$$

and as we mentioned at the beginning of this section we are using the unadjusted values for  $(\alpha^2 F)_{\text{cal}}$  taken directly from (17). Keeping in mind the approximations made in the calculation of  $\alpha^2(\omega)F(\omega)$  from (17), the agreement between two sets of curves for  $Z_{1N}(\omega)$ ,  $Z_{2N}(\omega)$ ,  $Z_{1S}(\omega)$ , and  $Z_{2S}(\omega)$  in Figs. 3(b) and 3(c) is quite reasonable.

#### A. dc Josephson Current

From our  $T = 0$  solutions of the Eliashberg equa-

tions we shall now calculate the reduction in the dc Josephson current from its BCS value. Fulton and McCumber<sup>33</sup> have discussed the problem of the (maximum) dc Josephson current that can flow at zero voltage across a tunnel junction made up of two identical superconductors. Strong-coupling effects in Pb reduce this current to 0.788 of the BCS value.<sup>6</sup> This result has been confirmed experimentally by Schwidtal and Finnegan.<sup>34</sup>

For a symmetric tunnel junction Fulton and McCumber have given an expression for the dc Josephson current  $J_s$  at temperature  $T$ . For  $T = 0$  it becomes

$$J_s = \frac{\pi \Delta_0}{2eR_N} [1 - \Delta_1'(\Delta_0)]^{-1} - \frac{1}{2R_N} \int_{\Delta_0^*}^{\infty} d\omega \text{Im} \left( \frac{\Delta^2(\omega)}{\omega^2 - \Delta^2(\omega)} \right), \quad (19)$$

where  $R_N$  is the normal-state resistance,  $e$  is the electron charge, and  $\Delta_1'(\omega)$  stands for the first derivative with respect to  $\omega$  of the real part of the gap function.  $\Delta_0^*$  means that the contribution from the pole  $\omega = \Delta_0$  is to be excluded from the integration. Equation (19) is to be compared with the result of Ambegaokar and Baratoff<sup>35</sup> for a weak-coupling superconductor at temperature  $T$ :

$$J_s^{\text{BCS}} = \frac{\pi \Delta_0}{2eR_N} \tanh \frac{\Delta_0}{2k_B T}. \quad (20)$$

Only the gap function enters in these expressions; the renormalization function does not occur explicitly. From our two sets of solutions for  $\Delta(\omega)$  we can easily calculate (19). The results are given in Table II. The results for  $J_s/J_s^{\text{BCS}}$  for the four alloy concentrations calculated using  $(\alpha^2 F)_{\text{cal}}$  are in good agreement with those calculated using  $(\alpha^2 F)_{\text{tun}}$ . Finally we would like to remark that in Table II the ratio  $J_s/J_s^{\text{BCS}}$  calculated from  $(\alpha^2 F)_{\text{tun}}$

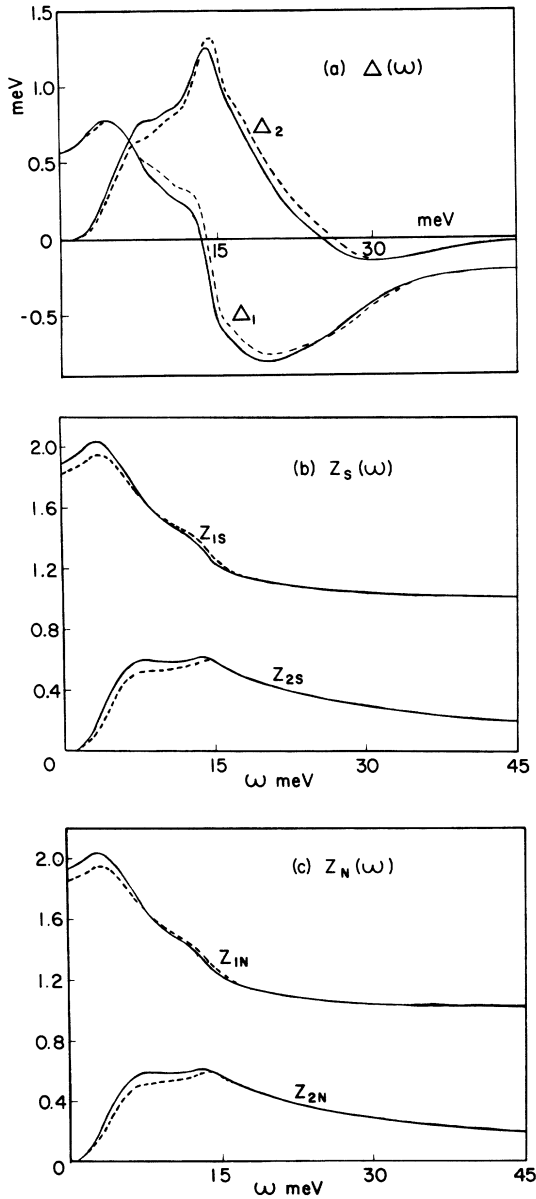


FIG. 4. Gap and renormalization functions for  $T = 0$  K for  $\text{In}_{0.73}\text{Tl}_{0.27}$ . Solid line is using tunneling-derived  $\alpha^2(\omega)F(\omega)$ ; dashed line is using calculated  $\alpha^2(\omega)F(\omega)$  from (17). (a) Real,  $\Delta_1$ , and imaginary,  $\Delta_2$ , parts of the gap function; (b) real,  $Z_{1s}$ , and imaginary,  $Z_{2s}$ , parts of the superconducting-state renormalization function; (c) real,  $Z_{1N}$ , and imaginary,  $Z_{2N}$ , parts of the normal-state renormalization function.

follows the same trend as the experimental value of the ratio  $(2\Delta_0/k_B T_c)$  for In, Tl, and four alloys.

#### B. Condensation Energy

For a strong-coupling superconductor the condensation energy is less than the BCS value. In Pb the reduction is 22% of the BCS value as calculated

by McMillan and Rowell,<sup>6</sup> which is in good agreement with experiment. The expression for the condensation energy as derived by Bardeen and Stephen<sup>36</sup> is

$$U = \text{Re}N(0) \left[ \int_0^\infty d\omega [Z_S(\omega) + Z_N(\omega)] \times \left( \omega - [\omega^2 - \Delta^2(\omega)]^{1/2} - \frac{\Delta^2(\omega)}{2[\omega^2 - \Delta^2(\omega)]^{1/2}} \right) - \int_0^\infty d\omega [Z_N(\omega) - Z_S(\omega)] \frac{\Delta^2(\omega)}{2[\omega^2 - \Delta^2(\omega)]^{1/2}} \right]. \quad (21)$$

To evaluate (21) we need  $Z_N(\omega)$  along with  $\Delta(\omega)$  and  $Z_S(\omega)$ .  $Z_N(\omega)$  is obtained by solving the Eliashberg equations with  $\Delta(\omega)$  equal to zero. The Bardeen-Stephen expression is to be compared with the BCS result<sup>32</sup>

$$U^{\text{BCS}} = \frac{1}{2} N(0) Z_N(0) \Delta_0^2, \quad (22)$$

where  $Z_N(0)$  is defined in (18) and  $N(0)$  is the band-structure density of states. In Table III we present our results for the condensation energy calculated from (21) using the two sets of solutions for  $\Delta(\omega)$ ,  $Z_S(\omega)$ , and  $Z_N(\omega)$ , obtained by using  $(\alpha^2 F)_{\text{tun}}$  and  $(\alpha^2 F)_{\text{cal}}$ . Results calculated from (22) are also given. The quantity entered in Table III is not  $U$  but  $U/N(0)$  and  $U^{\text{BCS}}/N(0)$  and the ratio  $R$  defined as

$$R = U/U^{\text{BCS}}.$$

From Table III it is clear that the values of the condensation energy obtained from (21) are always smaller than the BCS value  $U^{\text{BCS}}$ . The ratio  $R$  tends toward unity as the strength of the electron-phonon coupling decreases.<sup>37</sup> The values of  $U/N(0)$  and  $U^{\text{BCS}}/N(0)$  calculated using  $(\alpha^2 F)_{\text{cal}}$  are in good agreement with those calculated using  $(\alpha^2 F)_{\text{tun}}$ . Slight discrepancies in the values of the condensation energies are due to different values of  $Z_N(0) = 1 + \lambda$  from  $(\alpha^2 F)_{\text{cal}}$  and  $(\alpha^2 F)_{\text{tun}}$ . The critical magnetic field at zero temperature is related to the condensation energy by

TABLE II. Ratio of the strong-coupling to BCS value of the dc Josephson current for various In-Tl alloy compositions.

Alloy	$(2\Delta_0/k_B T_c)^{\text{Exp}^a}$	$J_s/J_s^{\text{BCS}}$ Using $(\alpha^2 F)_{\text{tun}}$	$J_s/J_s^{\text{BCS}}$ Using $(\alpha^2 F)_{\text{cal}}$ from (17)
In	3.683	0.898	...
$\text{In}_{0.90}\text{Tl}_{0.10}$	3.750	0.894	0.893
$\text{In}_{0.73}\text{Tl}_{0.27}$	3.937	0.879	0.887
$\text{In}_{0.67}\text{Tl}_{0.33}$	3.816	0.886	0.889
$\text{In}_{0.50}\text{Tl}_{0.50}$	3.786	0.899	0.901
Tl	3.676	0.905	...

<sup>a</sup>From Ref. 13.



TABLE III. Ratio of the strong-coupling to BCS value of the condensation energy for various In-Tl alloy compositions. Electron-phonon renormalization  $Z_N(0) = 1 + \lambda$  and condensation energy divided by band-structure density of states  $U/N(0)$  are also given in strong-coupling and BCS theories.

Alloy	$Z_N(0)$	Using $(\alpha^2 F)_{\text{tun}}$			$Z_N(0)$	Using $(\alpha^2 F)_{\text{cal}}$ from (17)		
		$U^{\text{BCS}}/N(0)$ (meV <sup>2</sup> )	$U/N(0)$ (meV <sup>2</sup> )	Ratio $R$		$U^{\text{BCS}}/N(0)$ (meV <sup>2</sup> )	$U/N(0)$ (meV <sup>2</sup> )	Ratio $R$
In	1.832	0.2670	0.248	0.931	...	...	...	...
In <sub>0.90</sub> Tl <sub>0.10</sub>	1.847	0.259	0.241	0.928	1.867	0.262	0.241	0.919
In <sub>0.73</sub> Tl <sub>0.27</sub>	1.929	0.313	0.283	0.904	1.857	0.302	0.276	0.915
In <sub>0.67</sub> Tl <sub>0.33</sub>	1.896	0.272	0.249	0.916	1.858	0.267	0.245	0.917
In <sub>0.50</sub> Tl <sub>0.50</sub>	1.831	0.155	0.144	0.934	1.856	0.157	0.145	0.927
Tl	1.777	0.121	0.115	0.949	...	...	...	...

$$H_c^2/8\pi = U = RU^{\text{BCS}}. \quad (23)$$

### C. Gap and Transition Temperature

Leavens and Carbotte(LC)<sup>18</sup> have derived simplified expressions for the gap and transition temperature of a weak- and intermediate-coupling superconductor by approximately solving the Eliashberg equations. Their equations are

$$\Delta_0 = 2\omega_D \exp[-(1 + \lambda(0) + \bar{\lambda})/(\lambda(0) - \mu^*)], \quad (24)$$

$$T_c = 1.134\omega_D \exp[-(1 + \lambda(T_c) + \bar{\lambda})/(\lambda(0) - \mu^*)]. \quad (25)$$

$\bar{\lambda}$  and  $\lambda(T)$  are defined as

$$\bar{\lambda} = 2 \int \frac{d\omega}{\omega} \alpha^2(\omega) F(\omega) \ln \left( 1 + \frac{\omega_D}{\omega} \right), \quad (26)$$

$$\lambda(T) = 2 \int dE \left( -\frac{\partial f(E)}{\partial E} \right) \int d\omega \frac{\alpha^2(\omega) F(\omega)}{\omega + E}, \quad (27)$$

where  $f(E) = (1 + e^{E/k_B T})^{-1}$  is the Fermi distribution,  $\lambda(0) = \lambda$ ,  $\omega_D$  is the maximum phonon frequency in  $\alpha^2 F$ , and  $\mu^*$  is the Coulomb part.<sup>38</sup> The main approximations involved in deriving (24) and (25) are that damping effects are neglected, that is to say  $\Delta_2 = Z_2 = 0$ , a model form is taken for the real part of the gap function  $\Delta(\omega)$ ,  $Z_S(\Delta_0)$  is taken to be equal to  $Z_N(0)$  at  $T = 0$  [this approximation is good for weak- and intermediate-coupling materials where the difference between  $Z_S(\Delta_0)$  and  $Z_N(0)$  is less than 1%<sup>18</sup>], and  $Z_S[\Delta(T), T]$  is taken to be equal to  $Z_N(0, T)$  near the transition temperature (this approximation is good for  $T$  near  $T_c$  and becomes exact at  $T_c$ ).

To calculate  $\Delta_0$  and  $T_c$  from (24) and (25), in addition to  $\alpha^2 F$  we need to know the value of  $\mu^*$  for the alloys. The experimentally observed energy gap  $\Delta_0^{\text{exp}}$  varies anything but smoothly from the value 0.540 meV for pure In to 0.369 for pure Tl, as the Tl concentration increases. The values of the Coulomb pseudopotential  $\mu_E^*$  (obtained from the  $T = 0$  Eliashberg equations by fixing  $\Delta_0$  equal to

$\Delta_0^{\text{exp}}$ ) do not vary smoothly in going from In to Tl and the variation from the average value is quite large. Furthermore there is a strong correlation between  $\mu_E^*$  and  $\Delta_0^{\text{exp}}$ ; when  $\Delta_0^{\text{exp}}$  is large,  $\mu_E^*$  is small and vice versa. The energy gap of an intermediate-coupling superconductor is fairly sensitive to the Coulomb part. To obtain good quantitative agreement with the experimental values one would need reliable values of  $\mu^*$  (which would certainly vary with  $c$  in the same manner as  $\mu_E^*$ ). Rather than become involved in the difficult task of trying to obtain reliable values of  $\mu^*$  to use in (24) and (25), we use a single value of  $\mu^*$  for all the alloy compositions. The value of  $\mu^*$  is taken to be

$$\mu_{\text{LC}}^*(\text{alloy}) = \frac{1}{2} [\mu_{\text{LC}}^{*\text{exp}}(\text{In}) + \mu_{\text{LC}}^{*\text{exp}}(\text{Tl})], \quad (28)$$

where  $\mu_{\text{LC}}^{*\text{exp}}(\text{In})$  and  $\mu_{\text{LC}}^{*\text{exp}}(\text{Tl})$  are obtained from (24) using  $(\alpha^2 F)_{\text{tun}}$  and requiring that the calculated energy gaps for pure indium and thallium be equal to the experimental gap values. This gives

$$\mu_{\text{LC}}^{*\text{exp}}(\text{In}) = 0.10808, \quad \mu_{\text{LC}}^{*\text{exp}}(\text{Tl}) = 0.10177,$$

and from (28) we get

$$\mu_{\text{LC}}^*(\text{alloy}) = 0.10492.$$

We shall use this value of the Coulomb part for all alloy compositions to study the concentration dependence of the gap and transition temperature when  $(\alpha^2 F)_{\text{tun}}$  and  $(\alpha^2 F)_{\text{cal}}$  are used to describe the phonon-mediated interaction. Of course, using the average value of  $\mu^* = \mu_{\text{LC}}^*(\text{alloy})$  we cannot hope to get good quantitative agreement with experiment.

In Fig. 5 we have plotted  $\Delta_0$  calculated from (24) by using  $\mu_{\text{LC}}^*(\text{alloy})$  for all values of  $c$ . The values given by crosses (x) and solid points were obtained by using  $(\alpha^2 F)_{\text{tun}}$  and  $(\alpha^2 F)_{\text{cal}}$ , respectively. The experimental gap values (+) are also shown. It is clear that the Leavens-Carbotte equation (24) with an average value of  $\mu_{\text{LC}}^*(\text{alloy})$  and  $(\alpha^2 F)_{\text{tun}}$

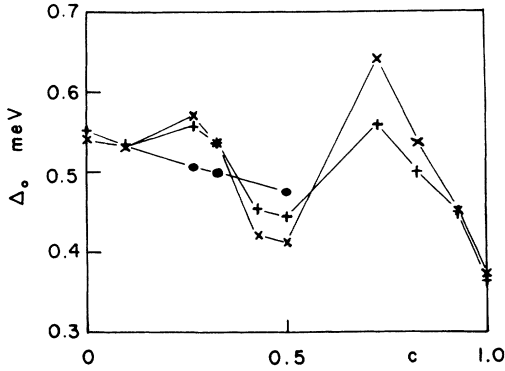


FIG. 5. Superconducting energy gap for various values of thallium concentration  $c$ ; + experiment,  $\times$  calculated from (24) using tunneling-derived  $\alpha^2(\omega)F(\omega)$  (Ref. 18), solid points calculated using  $\alpha^2(\omega)F(\omega)$  from (17).  $\mu_{LC}^*(\text{alloy}) = 0.10492$  for both sets of  $\alpha^2(\omega)F(\omega)$  for all values of  $c = 0-1$ .

gives results in good agreement with experiment.<sup>18</sup> The energy gap calculated from (24) with  $(\alpha^2 F)_{\text{cal}}$  and the same value of the Coulomb part are also in satisfactory agreement, considering the approximations involved in calculating  $\alpha^2 F$  for alloys from (17).

Using  $\mu_{LC}^*(\text{alloy}) = 0.10492$  in (25) and the two sets of  $\alpha^2 F$  we have self-consistently solved (25) and (27) for the transition temperature for various alloy compositions. In Fig. 6 we have plotted  $T_c^{\text{exp}}$  along with two sets of  $T_c$  obtained by using  $(\alpha^2 F)_{\text{tun}}$  and  $(\alpha^2 F)_{\text{cal}}$ . Again the agreement between  $T_c^{\text{exp}}$  and  $T_c$  calculated with  $(\alpha^2 F)_{\text{tun}}$  is quite satisfactory.  $T_c$  calculated from  $(\alpha^2 F)_{\text{cal}}$  is also in fair agreement with  $T_c^{\text{exp}}$  for the four alloy concentrations studied. With a simple-minded calculation of  $(\alpha^2 F)_{\text{cal}}$  from (17), with only one crystal structure for  $c = 0.10, 0.27, 0.33, 0.50$ , and neglecting such effects as changes in  $N(0)$ , we do not expect to reproduce such subtle features as the image of phase transition in the  $T_c$ -vs- $c$  curve for  $c \approx 0.30$ . In Table I the values of  $T_c$  calculated from the equations due to McMillan<sup>14</sup> and Garland and Allen<sup>15</sup> using  $(\alpha^2 F)_{\text{tun}}$  are given along with the values of  $T_c$  calculated from the equations due to Leavens and Carbotte<sup>18</sup> using  $(\alpha^2 F)_{\text{tun}}$  and  $(\alpha^2 F)_{\text{cal}}$ . It can be seen from Table I that  $T_c$  values from the Leavens-Carbotte equations, with a single value of  $\mu^*$  throughout, are in good agreement with experiment. The  $T_c$  values calculated from the McMillan equation, with values of  $\mu^*$  obtained from tunneling, are also in equally good agreement with experiment, whereas the Garland-Allen expression for  $T_c$  gives a much poorer estimate.

## V. CONCLUSION

We derive two exact sum rules for the alloy

phonon Green's functions, giving expressions for the first and first-inverse frequency moments ( $\mu^{(1)}$  and  $\mu^{(-1)}$ ) for the imaginary parts of these Green's functions. With their aid, expressions for the first frequency moment of  $\alpha^2(\omega)F(\omega)$  [ $\alpha^{(1)}$ , Eq. (6)] and the electron-phonon-coupling constant [ $\lambda$ , Eq. (11)] are obtained in which the only effect due to alloying that has been neglected is the effect of disorder on the electron states.  $\alpha^{(1)}$  is given in terms of the atomic masses, screened form factors, and the electron Fermi-surface density of states  $N(0)$ . Using this result the experimental values of  $\alpha^{(1)}$  can be used to estimate values of  $N(0)$  in an alloy. The expression for  $\lambda$  is more complicated, involving the atomic-force constants, and is not readily calculable. However, if just the difference in atomic masses is taken into account  $\lambda$  remains constant, independent of concentration.

Detailed calculations of alloy  $\alpha^2(\omega)F(\omega)$  are made in the framework of self-consistent defect theory. Confining ourselves to the nominally fcc region and taking into account only the mass difference between In and Tl, we obtain a fairly good over-all description of the concentration dependence of  $\alpha^2(\omega)F(\omega)$ . On comparing our theory with the experiment we reach the conclusion that, although there is a general weakening of the force constants near the phase transition fct-fcc, there is no clear-cut evidence for specific phonon-mode softening. In making a quantitatively correct calculation of the alloy  $\alpha^2(\omega)F(\omega)$  one will have to take into account the force-constant changes and such subtle features as the distorted cubic structure (In has a distorted cubic structure at low temperatures with  $c/a = 1.083$ ). Multiple-orthogonalized-plane-wave corrections may also be important.

In the light of the recent experiments by Schwidtal and Finnegan, who have confirmed experimentally the theoretical prediction of Fulten and McCumber for Pb, it may be of some interest to experimentally measure the reduction in the (maximum) dc Josephson current, from the BCS value, in the intermediate-coupling superconductors like In, Tl,

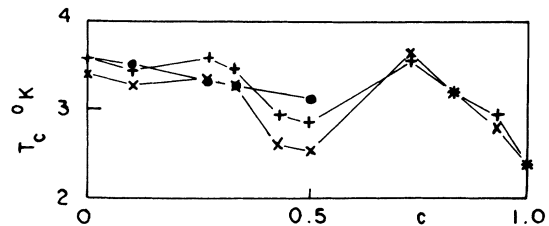


FIG. 6. Transition temperature  $T_c$  for various values of thallium concentration  $c$ ; + experiment,  $\times$  calculated using tunneling-derived  $\alpha^2(\omega)F(\omega)$ , solid points calculated using  $\alpha^2(\omega)F(\omega)$  from (17).  $\mu_{LC}^*(\text{alloy}) = 0.10492$  for both sets of  $\alpha^2(\omega)F(\omega)$  for all values of  $c = 0-1$ .

and In-Tl alloys. Experimental determinations of the critical magnetic field to check the predictions for the condensation energies of In-Tl alloys will also be of interest.

For In-Tl alloys (25), due to Leavens and Carbotte with a single value of the Coulomb part gives values of  $T_c$  as good as those obtained from the McMillan equation, which are in good agreement with the experiment. The Garland-Allen expression gives a much poorer estimate of  $T_c$ . It is rather gratifying to see that with a single average value of the Coulomb part  $\mu_{LC}^*(\text{alloy}) = \frac{1}{2}[\mu_{LC}^{*EP}(\text{In}) + \mu_{LC}^{*EP}(\text{Tl})]$  and the  $\alpha^2(\omega)F(\omega)$  from tunneling, one can obtain  $T_c$ 's from (25) which are in semiquantitative agreement with the experiment and reproduce reasonably

well the observed concentration dependence of  $T_c$ . The values of  $T_c$  from (25) with  $\alpha^2(\omega)F(\omega)$  calculated from (17) and the same  $\mu_{LC}^*(\text{alloy})$  are also in satisfactory agreement with the experiment. Keeping in mind the approximations made in calculating  $\alpha^2(\omega)F(\omega)$  from (17), one of course does not hope to reproduce such subtle features as the image of phase transition in the alloy structure in the  $T_c$  calculated with  $(\alpha^2F)_{\text{cal}}$  in (25).

#### ACKNOWLEDGMENTS

We wish to thank R. C. Dynes for tabulations of his experimental  $\alpha^2(\omega)F(\omega)$  prior to publication and R. C. Leavens and J. P. Carbotte for many discussions.

\*Research supported in part by the National Research Council of Canada.

<sup>†</sup>Present address: Physics Department, Western Michigan University, Kalamazoo, Mich. 49001.

<sup>1</sup>G. M. Eliashberg, Zh. Eksperim. i Teor. Fiz. **38**, 996 (1961) [Sov. Phys. JETP **11**, 696 (1960)].

<sup>2</sup>T. Nambu, Phys. Rev. **117**, 649 (1960).

<sup>3</sup>L. P. Gorkov, Zh. Eksperim. i Teor. Fiz. **34**, 735 (1958) [Sov. Phys. JETP **7**, 505 (1958)].

<sup>4</sup>D. J. Scalapino, J. R. Schrieffer, and J. W. Wilkins, Phys. Rev. **148**, 263 (1966).

<sup>5</sup>J. C. Swihart, D. J. Scalapino, and T. Wada, Phys. Rev. Letters **1**, 106 (1965).

<sup>6</sup>W. L. McMillan and J. M. Rowell, in *Superconductivity*, edited by R. D. Parks (Marcel Dekker, New York, 1968).

<sup>7</sup>P. Vashishta and J. P. Carbotte, Solid State Commun. **8**, 161 (1970); **8**, 1661 (1970); Can. J. Phys. **49**, 1413 (1971).

<sup>8</sup>C. S. Lim, J. D. Leslie, H. J. T. Smith, P. Vashishta, and J. P. Carbotte, Phys. Rev. B **2**, 1651 (1970).

<sup>9</sup>P. W. Wyatt and A. Yelon, Phys. Rev. B **2**, 4461 (1970).

<sup>10</sup>P. E. Seiden, Phys. Rev. **168**, 403 (1968).

<sup>11</sup>J. P. Carbotte and R. C. Dynes, Phys. Rev. **172**, 476 (1968).

<sup>12</sup>P. B. Allen and M. L. Cohen, Phys. Rev. **187**, 525 (1969).

<sup>13</sup>R. C. Dynes, Phys. Rev. B **4**, 3255 (1971); **2**, 644 (1970).

<sup>14</sup>W. L. McMillan, Phys. Rev. **167**, 331 (1968).

<sup>15</sup>J. W. Garland and P. B. Allen, in *International Conference on Science of Superconductivity*, Stanford, 1969 (published in Physica 1971).

<sup>16</sup>D. W. Taylor, Phys. Rev. **156**, 1017 (1967).

<sup>17</sup>M. Hansen, *Constitution of Binary Alloys* (McGraw-Hill, New York, 1958).

<sup>18</sup>R. C. Leavens and J. P. Carbotte, Can. J. Phys. **49**, 724 (1971).

<sup>19</sup>P. N. Trofimenkoff, thesis (McMaster University, 1969) (unpublished).

<sup>20</sup>D. N. Zubarev, Usp. Fiz. Nauk SSSR [Sov. Phys. Usp. **3**, 320 (1960)].

<sup>21</sup>D. Pines and P. Nozieres, *Theory of Quantum Fluids I* (Benjamin, New York, 1966).

<sup>22</sup>N. W. Ashcroft and W. E. Lawrence, Phys. Rev. **175**, 938 (1968).

<sup>23</sup>K. S. Singwi, A. Sjolander, M. P. Tosi, and R. H. Land, Phys. Rev. B **1**, 1044 (1970).

<sup>24</sup>H. W. White and D. C. McCollom, Phys. Rev. B **1**, 552 (1970).

<sup>25</sup> $R_c = 0.589 \text{ \AA}$  is required for agreement.

<sup>26</sup>A. O. E. Animalu and V. Heine, Phil. Mag. **12**, 1049 (1965).

<sup>27</sup>B. J. C. Van der Hoeven, Jr. and P. H. Keesom, Phys. Rev. **135**, A631 (1964).

<sup>28</sup>R. W. Meyerhoff and J. F. Smith, Acta Met. **11**, 529 (1963).

<sup>29</sup>R. J. Elliott and D. W. Taylor, Proc. Roy. Soc. (London) **A296**, 161 (1967).

<sup>30</sup>D. Buchanan and D. W. Taylor (unpublished); D. W. Taylor and R. C. Dynes, Bull. Am. Phys. Soc. **13**, 451 (1968).

<sup>31</sup>R. Bruno and D. W. Taylor, Can. J. Phys. **49**, 2496 (1971).

<sup>32</sup>D. J. Scalapino, in *Superconductivity*, edited by R. D. Parks (Marcel Dekker, New York, 1969).

<sup>33</sup>T. A. Fulton and D. E. McCumber, Phys. Rev. **175**, 585 (1968).

<sup>34</sup>K. Schwidtal and R. D. Finnegan, Phys. Rev. B **1**, 148 (1970).

<sup>35</sup>V. Ambegaokar and A. Baratoff, Phys. Rev. Letters **10**, 486 (1963).

<sup>36</sup>J. Bardeen and M. J. Stephen, Phys. Rev. **136**, A1485 (1964).

<sup>37</sup>J. P. Carbotte and P. Vashishta, Phys. Letters **33A**, 227 (1970).

<sup>38</sup>It is to be noted that the value of the Coulomb part  $\mu^*$  depends on the cutoff  $\omega_c$ . In the  $T=0$  Eliashberg equations we have used  $\omega_c = 70 \text{ meV}$  ( $\omega_c \approx 5\omega_D$ ), whereas in Leavens-Carbotte equation (22)  $\omega_c = \omega_D$ ; for this reason the respective Coulomb parts are

$$\mu_{LC}^* = \frac{N(0)V_c}{1 + N(0)V_c \ln(E_F/\omega_c)},$$

$$\mu_{LC}^* = \frac{N(0)V_c}{1 + N(0)V_c \ln(E_F/\omega_D)}.$$