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AN UNEXPECTED VARIATION OF THE COULOMB PSEUPOTENTIAL*

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ABSTRACT: The Coulomb pseudopotential, as determined from tunneling measurements, is found to vary over a wider range in superconducting *fcc* PbTl and PbBi than accounted for by currently accepted theory. This variation is shown to reflect structure in the density of states, as estimated from the rigid band model.

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INTRODUCTION

In their numerical solutions of the Eliashberg gap equations[1] for strong-coupling superconductors, Scalapino, Schrieffer and Wilkins accounted[2,3] for the screened Coulomb repulsion between electrons in an approximate way by making use of an effective Coulomb pseudopotential. This concept had previously been introduced by Bogoliubov, Tolmachev and Shirkov[4], and was subsequently clarified by Morel and Anderson in their treatment[5] of phonon retardation effects in a superconductor having an Einstein frequency spectrum.

Morel and Anderson found it convenient to represent the Coulomb interaction by the dimensionless pseudopotential

$$N(0)U_c = N(0)V_c/[1 + N(0)V_c \ln (E_F/\omega_E)] \qquad (1)$$

where $N(0)$ is the band-structure density of states of the metal at the Fermi energy, E_F, ω_E the frequency of the Einstein peak, and V_c is the angular average over the Fermi surface of the screened Coulomb interaction which, in the Thomas-Fermi approximation, gives

$$N(0)V_c = a^2 \ln \{1 + a^{-2}\} \qquad (2)$$

Here $a^2 = k_s^2/4k_F^2$, where k_s and k_F are the screening and Fermi wavevectors, respectively. Thus the effective Coulomb repulsion, competing with the phonon mediated attraction, was shown to be reduced by a factor of $\simeq 5$ (i.e. the denominator in Eq. 1) because of the instantaneous correlation it produces.

On the basis of their model and available normal state parameters, Morel and Anderson found $N(0)U_c$ to fall within a narrow band between 0.10 and 0.13 in all metallic elements, being 0.10 in Pb. These calculations have guided the thinking of many subsequent workers in the field.

In the strong-coupling theory[2,3] the same expression for $N(0)U_c$ arises (Eq. 1), with the exception that ω_E is replaced by a Coulomb cutoff usually taken to be five times the upper limit of the phonon spectrum.

McMillan and Rowell[6,7] showed how the analysis of the differential conductance of superconducting tunnel junctions can be made by inverting the Eliashberg equations. This provides a powerful technique for obtaining the phonon spectrum of the superconductor, multiplied by the electron-phonon coupling function, from which the electron-phonon renormalization factor can

be calculated. The constant $N(0)U_c$ is obtained as a byproduct of this analysis, by adjusting each iteration of the complex gap function to equal the measured energy gap at $\omega = \Delta_0$. In their tunneling studies, McMillan and Rowell determined[7] $N(0)U_c$ to be 0.12 for Pb.

The present author applied this technique to a series of *fcc* PbBi and PbTl alloys, with the principal findings reported elsewhere[8,9]. This series is well suited for such studies for several reasons. For one, substitutional *fcc* alloys exist over a wide range, from about 88% Tl in Pb to 18% Bi in Pb.[10] Because of the near equality of the constituent masses the electron density could reasonably be expected to be the dominant parameter

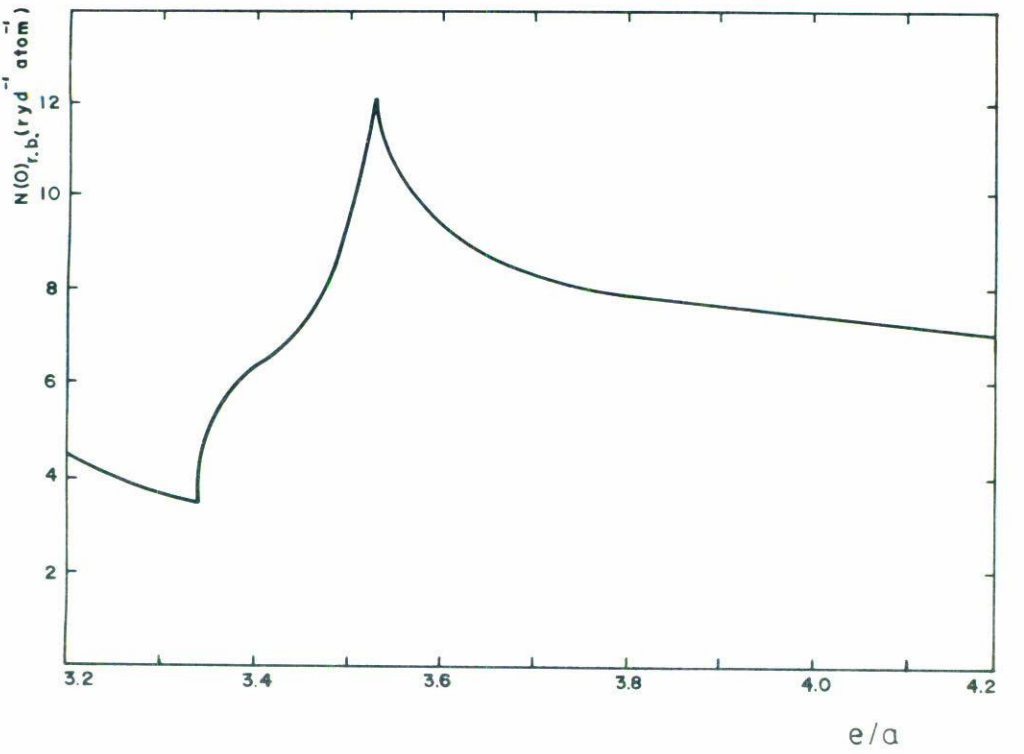

Fig. 1. Band structure density of states of PbTl and PbBi, as estimated from the rigid band model applied to Pb (Ref. 11). In the neighborhood of Pb the filling of the second zone produces a monotonically decreasing density of states. According to the rigid band calculation, the third zone begins to fill at $e/a \simeq 3.34$, its contribution reaches a peak at $e/a \simeq 3.53$, then falls quickly to a constant value. The fourth zone is expected to start filling at $e/a \simeq 4.08$, with a very slowly rising density of states. This contribution was neglected in the calculation because of its extreme smallness and because Anderson and Gold did not compute the larger contribution of the third zone above this point. The contribution of the third zone is expected to remain constant to well beyond the *fcc* phase limit. The ordinates have been doubled to include the spin degeneracy.

characterizing the system. In addition, a growing body of data, particularly based on phonon and superconductivity studies, is available for these alloys.

The values of $N(0)U_c$ computed in this work for many compositions fall outside the narrow limits suggested by Morel and Anderson. Furthermore, they do not vary monotonically with electron density. On the other hand, large variations would be expected in $N(0)$ over the compositional range on the basis of the rigid band model applied to the density of states curve computed for Pb,[11] and these fluctuations are such that $N(0)U_c$ varies smoothly with $N(0)_{r.b.}$. Here $N(0)_{r.b.}$ is the density of states curve estimated from the rigid band model and shown in Fig. 1. Thus the Coulomb pseudopotential reflects structure in $N(0)_{r.b.}$. In Fig. 2 the variation of "U_c" $= N(0)U_c/N(0)_{r.b.}$ with $N(0)_{r.b.}$ is plotted. It is interesting that a smooth curve through the data presented in Fig. 2 suggests $N(0)U_c = 0.13$ for Pb, a value considered appropriate by several workers[12-15].

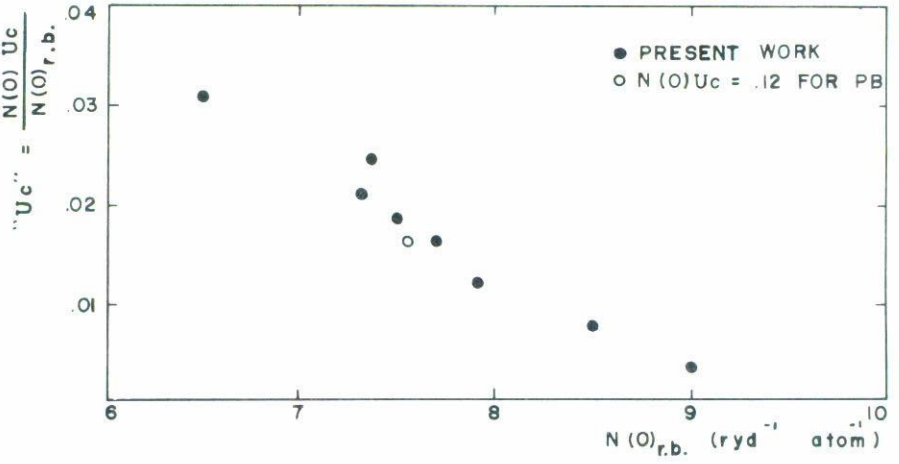

Fig. 2. Effective Coulomb potential "U_c" vs. $N(0)_{r.b.}$ in the series of PbTl and PbBi alloys. The point for Pb has been taken from Ref. 7.

Although it is not possible to state the uncertainty of a given determination of $N(0)U_c$*, a simple test suggests indirectly that most of the

* Owing to the non-linearity of the Eliashberg equations and the intricacy of the inversion scheme, it is not possible to estimate the uncertainty of a given determination of $N(0)U_c$. In this work the energy gap was measured to ± 0.005 mV and the differential conductance determined with a precision of a few parts in 10^5. The gap function was then computed until it and the resulting effective phonon spectrum converged in successive iterations to 1%. The range of integration extended to 40 mV.

data presented here are consistent with the measured electron-phonon interaction and the transition temperature, T_c. Leavens and Carbotte have recently pointed out[16] that at least for superconductors whose electron-phonon renormalization parameter, $Z_N(0) = 1 + \lambda$, lies in an "optimum" range ($1.15 \leqslant \lambda \leqslant 2.46$), a plot of T_c vs. A is linear with an empirical slope of 0.1477, where

$$A = \int d\omega\ \alpha^2(\omega)\ F(\omega) / \int d\omega F(\omega) \tag{3}$$

and

$$\lambda = 2 \int d\omega\ \alpha^2(\omega)\ F(\omega)/\omega \quad . \tag{4}$$

Here $F(\omega)$ is the phonon spectrum and $\alpha^2(\omega)$ the electron-phonon coupling function at frequency ω. (The data used by Leavens and Carbotte encompassed the interval $0.105 \leqslant N(0)U_c \leqslant 0.14$). The present data for all samples with $N(0)U_c > 0.10$ lie within a few percent of the Leavens-Carbotte line. The three other samples, having $N(0)U_c < 0.10$ show deviations of as much as 25% from this line, in a direction suggesting that *if the $N(0)U_c$ obtained were above 0.10*, A should be larger to conform with T_c. In any case, the interpretation of these data in the light of Leavens and Carbotte is not entirely appropriate, as several values of the Coulomb pseudopotential fall well outside the interval considered by them.

Two considerations encouraged the use of the rigid band model. In his study of the phonon dispersion curves of these alloys, Ng concluded[17,18] that the Fermi surface varies in accord with this model. In particular, he found that for $Pb_{40}Tl_{60}$ only a small part of the Fermi surface remains in the third zone. Furthermore, Clune and Green discovered[19] that the rigid band model satisfactorily accounts for electronic specific heats in normal state PbTl and PbBi, provided electron-phonon renormalization is properly included.

In an attempt to understand the cause of this variation, $N(0)U_c$ was computed for each alloy studied, from Eqs. 1 and 2. For this purpose the square of the screening wave-vector was assumed to be[20]

$$k_S^2 = 6\pi n_0 e^2 / E_F \tag{5}$$

where n_0 is the electronic density of the alloy. Since Anderson and Gold

showed[11] that Pb is, surprisingly, a nearly free electron metal, the dispersion relation was taken to be parabolic, i.e.

$$k_F^2 = 2m^* E_F/\hbar^2 \tag{6}$$

Here m^* is assumed to be proportional to the band-structure density of states, $N(0)$. Thus the quantity (see Eq. 2)

$$a^2 = X(e/a)/\left(N(0)\, E_F^2\right) \tag{7}$$

where X is a constant determined from the value $a^2 = 0.38$ found by Morel and Anderson[5] for Pb, and $N(0)$ and E_F are estimated from the rigid band model.

We note that $N(0)V_c$ (see Table 1) calculated in this manner is generally increasing with increasing e/a, the exception being in $Pb_{40}Tl_{60}$ which has the smallest $N(0)_{r.b.}$ of the samples studied. Although $N(0)$ is a factor in the calculation of $N(0)V_c$, this quantity clearly varies systematically with electron density.

The remaining factor in Eq. 1, as modified in the strong coupling theory, is the logarithm of the ratio of E_F to an arbitrary cutoff of several times the maximum phonon frequency. The experimental results show the phonon limit to fall more or less linearly with increasing e/a, from 11.0 mV in $Pb_{40}Tl_{60}$ to 9.0 mV in $Pb_{85}Bi_{15}$. Assuming the cutoff to be five times this phonon limit, the factor $\ln(E_F/5\omega_{max})$ is calculated to increase slowly with e/a, from 5.07 in $Pb_{40}Tl_{60}$ to 5.36 in $Pb_{85}Bi_{15}$. A calculation then shows the predicted values of $N(0)U_c$ for these alloys all to lie in the narrow range 0.144 – 0.135, the smaller ones corresponding to larger e/a. These results as well as $N(0)U_c$ determined experimentally and "U_c" are summarized in Table 1, along with other pertinent data. We thus conclude that the expression for the Coulomb pseudopotential as commonly used in the strong coupling theory cannot be reconciled with the experimentally obtained pseudopotentials for this series of alloys.

Only one other tunneling study provides directly comparable data. Dynes[15] reported the following results:

	$N(0)U_c$	λ
$Pb_{40}Tl_{60}$	0.113	1.15
$Pb_{60}Tl_{40}$	.126	1.38
Pb	.131	1.55
$Pb_{90}Bi_{10}$	.10	1.66

TABLE I

ALLOY	e/a	$N(0)_{r.b.}$ [a]	E_F [a]	λ [b]	A [b]	$N(0)V_c$ [c]	$\omega_{max.}$ [b]	$N(0)U_c$ [d]	$N(0)U_c$ [b]	"U_c" [e]
$Pb_{40}Tl_{60}$	3.40	6.50	0.647	1.00	2.65	0.530	11.0	0.144	0.199	0.0306
$Pb_{63}Tl_{37}$	3.63	9.00	.671	0.97	2.50	.458	10.8	.138	.029	.0032
$Pb_{70}Tl_{30}$	3.70	8.50	.680	1.17	3.02	.469	10.5	.137	.065	.0076
$Pb_{84}Tl_{16}$	3.84	7.90	.697	1.32	3.46	.481	10.0	.136	095	.0120
$Pb_{93}Tl_{07}$	3.93	7.70	.709	1.49	3.90	.484	9.8	.136	.124	.0161
$Pb_{95}Bi_{05}$	4.05	7.50	.727	1.63	4.28	.486	9.4	.136	.138	.0184
$Pb_{89}Bi_{11}$	4.11	7.35	.738	1.77	4.42	.488	9.2	.135	.181	.0246
$Pb_{85}Bi_{15}$	4.15	7.30	.744	1.81	4.35	.489	9.0	.135	.152	.0208

Units: $N(0)_{r.b.}$ in $\{\text{ryd}^{-1}\,\text{atom}^{-1}\}$, E_F in $\{\text{ryd}\}$, A and ω_{max} in $\{\text{mV}\}$ and "U_c" in $\{\text{ryd atom}\}$

The remaining quantities are dimensionless.

a) Estimated from the rigid band model.

b) From tunneling experiment (Ref. 8).

c) Calculated from preceding data using Eqs. 2 and 7.

d) Calculated from preceding data using Eq. 1 with ω_E replaced by $5\omega_{max}$.

e) "U_c" $= N(0)U_c\,(\text{exp})/N(0)_{\text{r.b.}}$

which vary less strongly with composition than those reported in the present paper. In particular, the values of $N(0)U_c$ he found lie within the Morel and Anderson range. Wu also reported[21] $N(0)U_c$ near 0.11 for Pb and three compositions of PbTl, based on preliminary data of the present author which he considers to be unreliable. Rowell *et al* made a detailed investigation[22] of Pb and $Pb_{40}Tl_{60}$ but they neglected to report the values of $N(0)U_c$ obtained. Adler and Chen[23] have reported $N(0)U_c = 0.15$ in $Pb_{70}Bi_{30}$ and alloy beyond the *fcc* phase.

A similar determination of the Coulomb pseudopotential was made by Dynes[24] for the series of InTl alloys, a system in which e/a is constant. Dynes found $N(0)U_c$ to be within 0.01 of 0.12 in both elements and all intermediate compositions.

There is a growing body of evidence[25, 26] that the Coulomb pseudopotential is enhanced in amorphous metals and alloys above its crystalline value, parallel to the increase in electron-phonon coupling strength. The value $N(0)U_c = 0.19$ has been reported[26] for amorphous Pb (+ 5% Bi). It is believed that this effect cannot be responsible for the "anomalous" values reported here, because in fact some of these are depressed below the Morel and Anderson estimates. Moreover, in this work the thin films were deposited on substrates at room temperature under conditions not conducive to amorphicity.

CONCLUSIONS

The purpose of this Note is to point out that in the superconducting *fcc* alloys PbTl and PbBi, the Coulomb pseudopotential varies more widely than expected on the basis of the expression customarily employed in the theory of strong coupling superconductivity[2, 3]. Furthermore, the variation of $N(0)U_c$ with e/a reflects structure expected in the density of states. Correspondingly, $N(0)U_c$ and "U_c" $= N(0)U_c/N(0)_{r.b.}$ vary monotonically with $N(0)_{r.b.}$

It is important to investigate in more detail the range on composition of PbTl for which $N(0)_{r.b.}$ varies rapidly in order to resolve the discrepancies between the data presented here and those of Dynes[15]. Such a study, when combined with normal state electronic specific heat coefficients, should serve as well to determine the density of states of the alloys, $N(0)$, without recourse to rigid band calculations, beyond the narrow range studied by Clune and Green.[19]

It should be emphasized that in the realm of soft superconductors, interest in $N(0)U_c$ has languished for several years. This neglect is equally a consequence of the intractability of the field theoretic expressions for the Coulomb interaction and the outstanding success of the strong-coupling theory in unraveling phonon effects in tunneling experiments once a "reasonable" value of $N(0)U_c$ has been fixed.

By contrast the Coulomb pseudopotential in transition metals has elicited somewhat more attention[27]. Estimates of this quantity have come from the isotope effect[28,29] and the enhancement of spin paramagnetism[30]. For example, in the *hcp* alloy series ScZr ($3 \leqslant e/a \leqslant 4$), $N(0)U_c$ is estimated[28] to vary widely, well outside the Morel and Anderson range. Gladstone *et al*[27] have pointed out that a properly chosen constant U_c and a constant phonon interaction term suffice to explain the wide variation of T_c in the central transition metals ($4 \leqslant e/a \leqslant 6$). They also point out that if $N(0)$ varies rapidly near the Fermi surface, in Eq. 1 its occurrence *in the denominator* should be replaced by an average over an energy interval of order E_F, or at least of order ω_{max}. (In any case this modification would not significantly affect the calculations presented in the present Note, as the dominant factor is the numerator in which $N(0)$ is unaltered).

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RESUMEN

Se encuentra que el pseudopotencial coulombiano, determinado a partir del efecto túnel, varía más ampliamente en aleaciones superconductivas de PbTl y PbBi (*ccc*) de lo que predicen las teorías de aceptación general. Se demuestra que esta variación refleja estructura en la densidad de estados, que es estimada empleando el modelo de bandas rígidas.