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Effects of interlayer interactions on the critical temperature of high-T_c compounds

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We show that the dependence of T_c on the number of layers in the unit cell of high-T_c thallium compounds is quantitatively explained within the BCS framework, irrespective of the precise mechanism responsible for pairing. The intraplanar pairing is augmented by a weak interplanar scattering of the pair, thereby enhancing the T_c of layered compounds without increasing either the density of states or the number of carriers per CuO_2 plane. The origin of the interplanar coupling is investigated and implications of the present model to other high-T_c materials (bismuth and yttrium compounds) are discussed.

The recent discovery[1] of high-T_c superconductivity in bismuth and thallium compounds ($Bi_2Ca_{l-1}Sr_2Cu_lO_x$ and $Tl_2Ca_{l-1}Ba_2Cu_lO_x$, $l=1$, 2, and 3) not only sets a new record in the high-T_c race, but provides useful information in understanding the high-T_c mechanism, namely, a systematic increase in T_c as the number of CuO_2 layers in the unit cell (l as defined above) increases. One of the most controversial issues in high-T_c superconductivity has been whether the general framework of the BCS theory is valid in describing the new high-T_c oxides. Wheatley, Hsu, and Anderson[2] attempted to explain the multilayer effects of Bi and Tl compounds in terms of their resonating-valence-bond theory, whose formalism is totally different from the BCS theory. While the validity of their model will ultimately be determined by future experiments, some questions immediately arise regarding their results. An example is how the coupling between outer CuO_2 layers of more than 11 Å apart can be comparable ($\sim$60%) to that between inner layers of $\sim$3 Å apart in Tl compounds (i.e., $\Lambda_0/\Lambda_m \sim 0.6$ in their notation[2]). Another question is whether the conventional BCS theory can account for the behavior of Bi and Tl compounds as well (or better). There are also other theories[3] providing different explanations for the high-T_c superconductivity of these materials.

In the present paper, we show that the dependence of T_c on the number of layers of Tl compounds is quantitatively explained in terms of the BCS theory. Our model assumes that the (still unidentified) pairing interaction within a CuO_2 layer is strong enough to exhibit superconductivity by itself and a weak interlayer interaction enhances T_c further. The predicted limit of T_c attainable by stacking more CuO_2 layers ($l \rightarrow \infty$), however, turns out no more than 150 K (providing each layer has the same density of states as discussed below), not a significant improvement over 125 K for $l=3$. We will concentrate on the study of Tl compounds and a brief discussion on other high-T_c materials (Bi and Y compounds) will be given at the end, with details reserved for a longer paper. From the unusually high T_c of the Tl compounds (near 80 K even for the lowest case of the 2:0:2:1 compound), it seems clear that the phonon is not the major mediator of the pairing. The smallness of the isotope effect[4] in $YBa_2Cu_3O_7$ ($\alpha \leq 0.05$), when extrapolated to Tl compounds (we are unaware of isotope effect measurements for Tl and Bi compounds yet), is another indication that phonons do not play a significant role. We use the BCS formalism below imagining that some unidentified quasiparticles originated from the Fermi sea of charge carriers may give rise to Cooper pairing.

There are basically three parameters determining T_c in the BCS theory [$kT_c = 1.14\hbar\omega \exp(-1/NV)$], and the increase of T_c as a function of l should also be explained by them. Since the fundamental pairing mechanism should be independent of l, the prefactor $\hbar\omega$ which is essentially the range of interaction of the pairing mechanism would be unaffected by l. Remaining parameters are the density of states at the Fermi level N and the interaction (coupling) energy V. An important feature in our model is that increasing l above 1 adds the interlayer interaction energy to V, but keeps N per CuO_2 layer (contributed from CuO_2 units) essentially constant. The assumption of constant N per CuO_2 layer is justified in a number of ways. We first note that the observed structure of $Tl_2Ca_{l-1}Ba_2Cu_lO_x$ is surprisingly intact despite the intercalation of $CaCuO_2$ units as l increases. No driving force has been identified experimentally that can change the density of states per CuO_2 layer. Moreover, theoretical calculations[5] for the density of states of $Tl_2Ca_1Ba_2Cu_2O_8$ and $Tl_2Ca_2Ba_2Cu_3O_{10}$ indicate N per CuO_2 layer remains the same. The assumption of course requires more direct experimental verification in the future.

On the other hand, the charge carrier density n, not appearing explicitly in the BCS expression of the T_c, has been shown[6] to correlate with T_c very closely in $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{7-y}$ (n is not a nominal density from a valence count but the real density of carriers which are actively participating in conduction). This does not contradict the BCS formalism at all because N at the Fermi level is explicitly dependent on n and V may also implicitly depend on n. In particular, if the pairing is mediated by charge carriers as believed by many people, the prefactor $\hbar\omega$ ($\sim$Fermi energy) is a monotonically increasing function of n. This correlation between n and T_c, though undoubtedly true, is not relevant to the l dependence of T_c since the number of carriers n per CuO_2 layer located on the CuO_2 plane is independent of l for the same reason as

N per CuO_2 layer is unchanged.

Now the procedure of the calculation goes as follows. There is a strong intralayer interaction V_a and a weak interlayer interaction V_{er} between the CuO_2 layers of ~ 3.1 Å apart as schematically shown in Fig. 1. We can safely neglect the interaction between outer layers of ~ 11.5 Å apart. The formalism in the present model is remarkably similar to the two-band pair scattering version of the BCS theory.[7,8] The difference from previous works is that each layer is now considered to produce a two-dimensional energy band. A mixed representation of the carrier in terms of the momentum-space index k for the ab plane and the real-space index l for the lth layer in the c-axis direction is a convenient choice in this layered material. The reduced Hamiltonian used here is

$$H=\sum_{lk\sigma}\varepsilon_k c^*_{lk\sigma}c_{lk\sigma}-\sum_{lkk'}V_a c^*_{lk\uparrow}c^*_{l-k\downarrow}c_{l-k'\downarrow}c_{lk'\uparrow} \quad (1)$$

$$-\sum_{ll'kk'}V_{er}c^*_{lk\uparrow}c^*_{l'-k\downarrow}c_{l'-k'\downarrow}c_{l'k'\uparrow}\,,$$

where ε_k's are the normal-state band energy and c is the annihilation operator of the charge carrier. For simplicity, V_a and V_{er} are assumed independent of l, l', k, and k'. It is understood that $l\neq l'$. Note that the k summation should extend only over the two-dimensional reciprocal space for each layer. The k summation for the pair-interaction part (the second and third term on the right-hand side) is further restricted to within the energy range $\pm\hbar\omega$ (measured from the Fermi level) of whatever interaction responsible for the pairing. Applying the standard Bogoliubov transformation, we obtain a set of gap (Δ) equations to be solved simultaneously as in Ref. 7:

$$\Delta_l[1-V_aNF(\Delta_l)]=\sum_{l'}\Delta_{l'}V_{er}NF(\Delta_{l'})\,, \quad (2)$$

$$F(\Delta_l)=\int_0^{\hbar\omega}d\varepsilon\tanh[(\varepsilon^2+\Delta_l^2)^{1/2}/2kT]/(\varepsilon^2+\Delta_l^2)^{1/2}. \quad (3)$$

We note here that the interlayer scattering in general gives rise to two additional terms not included in the Hamiltonian in Eq. (1). These omitted terms are

$$H'=-\sum_{ll'kk'}V'_{er}c^*_{lk\uparrow}c^*_{l'-k\downarrow}c_{l-k'\downarrow}c_{l'k'\uparrow}$$

$$-\sum_{ll'kk'}V''_{er}c^*_{lk\uparrow}c^*_{l'-k\downarrow}c_{l-k'\downarrow}c_{l'k'\uparrow}\,. \quad (1')$$

V_{er} in Eq. (1) corresponds to the scattering of the pair with both mates in layer 1 into the state with both mates in layer 2, whereas V'_{er} and V''_{er} in Eq. (1') correspond to the scattering of the pair consisting of a mate in layer 1 and the other in layer 2. It turns out that the usual Bogoliubov transformation of H', unlike the case of V_{er}, does not give an additional contribution to the energy [i.e., to the gap equations (2) and (3)] in the first order. A more appropriate way to handle the situation would be to introduce a modified Bogoliubov transformation which includes a small contribution (i.e., mixing) from carriers in the neighboring layers. Corresponding changes in the gap equations, however, can be shown to be second order in the degree of mixing, and hence neglected.

Among l distinct solutions of Eqs. (2) and (3), the physically realized solution should be the one with the lowest free energy. The smallest positive F produces the lowest free energy, as well as the highest T_c through the relation, $kT_c\cong 1.14\hbar\omega\exp(-F)$. The desired solution of Eqs. (2) and (3) turns out

$$F=1/[\lambda_a+p(l)\lambda_{er}]\,, \quad (4)$$

where $\lambda_a=NV_a$, $\lambda_{er}=NV_{er}$, and the geometrical factor $p(l)=0$, 1, $\sqrt{2}$, $(\sqrt{5}+1)/2$, $\sqrt{3}$, and 2 for $l=1$, 2, 3, 4, 5, and ∞, respectively. In Tl compounds, the reported[1] T_c's were <80, 108, and 125 K for $l=1$, 2, and 3, respectively. We vary $\hbar\omega$ in a wide range and obtain λ_a and λ_{er} which fit the measured T_c's. If $\hbar\omega$ is taken comparable to phonon energy, unphysical values for λ_a and λ_{er} are obtained whether the BCS form or strong-coupling forms are used for T_c. This reassures us that the phonons are ruled out from the major mediator of pairing. Now it is reasonable to take $\hbar\omega$ comparable to the Fermi energy of carriers, in the spirit that some kind of charge carrier-mediated pairing mechanism is operating.[9] Results are presented in Table I for $\hbar\omega=0.1$ and 0.2 eV. The fitting is successfully done with reasonable values of λ_a and λ_{er}. (The measured T_c for $l=1$ is not as accurately known as those for $l=2$ and 3.) The magnitude of the calculated coupling parameter $\lambda_a(\sim 0.3)$ is not unusual for a weak-coupling superconductor. The relatively small size of λ_{er} with respect to λ_a is also acceptable. These calculated λ_a

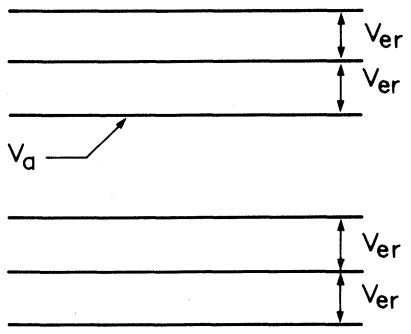

FIG. 1. Schematic diagram of the coupling interactions in our model for $l=3$. Each horizontal line represents a CuO_2 layer. V_a exists in each layer (only one of them is indicated for visual clarity).

TABLE I. The calculated T_c^l for $Tl_2Ca_{l-1}Ba_2Cu_lO_x$ and the parameters used in Eq. (4). In each row, $\hbar\omega$ is first chosen and λ_a and λ_{er} are determined to fit starred (*) values.

$\hbar\omega$ (eV)	λ_a	λ_{er}	T_c^1 (K)	T_c^2 (K)	T_c^3 (K)	T_c^4 (K)	T_c^5 (K)	T_c^∞ (K)
0.1	0.356	0.049	80*	111	125*	132	136	145
0.1	0.339	0.060	70	108*	125*	133	138	150
0.2	0.277	0.036	71	108*	125*	134	139	151

and λ_{er} of course correspond to effective, renormalized coupling parameters. An interesting question is how far T_c can go up as we increase l, let aside the problem of material fabrication. As shown in the same table, we get $T_c \sim 133$ K for $l=4$, ~ 138 K for $l=5$, and ~ 150 K for a large l limit. Therefore, our model makes a pessimistic prediction that fabricating higher l compounds, even if successful, does not help enhance T_c much further (unless λ_a or λ_{er} is strengthened simultaneously). In any case, as the prefactor $\hbar\omega$ increases from the phonon energy to near the Fermi energy of the carriers, relatively weak coupling parameters λ_a and λ_{er} are capable of accounting for T_c as high as 125 K. It is surprising that a tiny interlayer coupling λ_{er} can increase T_c by nearly 50 K for $l=3$. The reason, of course, is that λ_{er} enters the argument of a fast increasing exponential function. Although increasing l alone beyond three does not enhance T_c much further as shown above, there remains possibility of increasing λ_a and λ_{er} and achieving higher T_c since these coupling parameters do not seem to have saturated yet. For example, λ_a and λ_{er} would be strengthened if n could be increased by some means.

Another notable consequence of the model is that, if l equals $2m$ or $2m-1$, m distinct gaps exist. If normalized to T_c, the $l=2$ case is identical to the conventional BCS result for $l=1$ ($2\Delta(0)/kT_c=3.53$). There are two gaps for $l=3$ and 4. $2\Delta(0)/kT_c=3.92$ and 3.26, respectively for two gaps for $l=3$, and they are 3.80 and 3.10 for $l=4$ if $\hbar\omega=0.1$ eV is used. It would be interesting to verify experimentally if there should exist two gaps in $Tl_2Ca_2Ba_2Cu_3O_x$, one associated with the middle layer and the other with the outer layers. However, if we follow the above formalism in more detail, the situation turns out rather complicated. The energy dependence of λ_a or λ_{er} makes Δ energy dependent, and the layered structure produces gap anisotropy [$\Delta=\Delta(\mathbf{k})$]. The analysis of two sets of continuously distributed gaps is nontrivial and information from experiment is still insufficient. Furthermore, since the real sample is far from perfect, scattering will significantly average out the observed gap structure.

Now we are going to consider the origin of the proposed interlayer interaction. It is impossible at present to pinpoint the particular mechanism responsible for it, but we can speculate on a few possibilities. The soft plasmon (or demon) modes originated from screening of c-axis conduction carriers (of heavy mass) by ab plane conduction carriers (of light mass) can give rise to a weak interlayer coupling according to the two-band model.[8] Polarization effects (e.g., charge transfer excitations[10]) across nearest-neighbor layers are perhaps possible. Also, the van der Waals (fluctuating dipole) interaction probably exists between layers as partly manifested, for instance, in the cleavage energy of the layered Bi compounds. (We note, however, the first-order effect of fluctuating dipoles on conducting monopoles, unlike the dipole-dipole interaction, will be averaged out in time.) Another possibility we are going to consider in some detail is the most conventional phonon exchange. Although phonons have been ruled out as the major mediator of coupling, they might still be responsible for the very weak V_{er} only.

Let us suppose that V_{er} is primarily due to phonon scattering and V_a is primarily due to some other (charge carrier-mediated) mechanism. Now the interaction range of V_{er} changes to $\pm\hbar\omega_0$, where $\omega_0(<\omega)$ is the phonon frequency of the material. Then the gap equations [Eqs. (2) and (3)] are modified accordingly. The resulting equations are not much more complicated to solve: The only two changes to make in order to get T_c are (i) to replace λ_a by

$$\lambda_a^* = \lambda_a \Big/ \left(1-\lambda_a\int_{\hbar\omega_0}^{\hbar\omega} d\varepsilon \tanh(\varepsilon/2kT_c)/\varepsilon\right)$$

and (ii) to integrate F only up to $\hbar\omega_0$ rather than $\hbar\omega$. What interests us here most is the nonzero isotope effect from phonon contributions. The exact expression for the index of the isotope effect $\alpha(T_c \propto M^{-\alpha})$ in this model is, by differentiating Eq. (4) with above modification,

$$\alpha = 0.5 \Big/ \left(1+\frac{\tanh(\hbar\omega/2kT_c)}{\tanh(\hbar\omega_0/2kT_c)}\frac{\lambda_a^{*2}}{[\lambda_a^*+p(l)\lambda_{er}]^2-\lambda_a^{*2}}\right), \tag{5}$$

which is smaller than 0.5. With reasonable values for $\hbar\omega_0$ (a few tens of meV), we can fit T_c's for $l=1$, 2, and 3 and predict α. A typical set of values is $\hbar\omega=0.2$ eV, $\hbar\omega_0=0.04$ eV, $\lambda_a=0.271$, and $\lambda_{er}=0.151$ (compared with $\hbar\omega=0.2$ eV, $\lambda_a=0.277$, and $\lambda_{er}=0.036$ in Table I). We see that as the interaction range of λ_{er} is reduced, the magnitude of λ_{er} has to increase to produce the same increment of T_c for $l>1$. The size of λ_{er} ($=0.151$) is not unreasonable for phonon coupling. However, the calculated α in this case turns out 0.21, 0.25, and 0.27 for $l=2$, 3, and 4. Assuming that the isotope effect of Tl (or Bi) compounds is very small as in $YBa_2Cu_3O_7$ ($\alpha \lesssim 0.05$),[4] this calculation strongly indicates that phonons are *not* the major mediator of either the in-plane or the interlayer coupling.

Applications of the model to Bi compounds are withheld so far despite obvious kinship between Tl and Bi compounds. Not only is a single-phase Bi sample difficult to obtain but the structure is nonuniform as a function of l (especially for $l=1$, compared with $l=2$, and 3). In other words, we suspect that the number of carriers n or the density of states N per CuO_2 layer is not the same for different l and more information is necessary to solve the gap equations. (Approximate calculations are certainly possible assuming a constant n as in Tl compounds.)

One may wonder if our model is relevant to $YBa_2Cu_3O_{7-y}$. There are two CuO_2 planes and one CuO chain interacting each other in the unit cell of $YBa_2Cu_3O_{7-y}$. We have analyzed the system in the same way as Tl compounds. Since the number of CuO_2 layers is fixed here, the intra- or interlayer interaction is not an interesting parameter. Rather, we pay attention to the experimental results that the T_c is, crudely speaking, ~ 90 K for $0<y<0.2$ and $\lesssim 60$ K for $0.2<y<0.5$. It is believed that the Cu chain is preserved in the composition range of $0<y<0.2$ and significantly interrupted for $0.2<y<0.5$. We have transformed our formalism so that λ_a' now represents the total pairing interaction within the complex of *two* CuO_2 planes and λ_{er}' represents the chain-plane coupling. (The chain presumably does not superconduct by

itself.) When the chain structure is destroyed, the distance between outer CuO_2 planes is so large ($\sim$8.6 Å) that only λ_a' survives. Reasonable results obtained from this model by fitting to T_c=90 and 60 K ($\hbar\omega$=0.2 eV, λ_a'=0.264, and λ_{er}'=0.067) seem to suggest that the chain-plane interaction does play a supplementary role for high T_c. However, since n, N, and V_{er} will actually change as y changes, it is essential to take into account these changes for detailed understanding of $YBa_2Cu_3O_{7-y}$.

To summarize, we have shown that the dependence of the T_c on the number of CuO_2 layers l is well explained by the BCS theory if the intra-and interlayer coupling coexist. Simply increasing l does not enhance T_c much (150 K at best), but the weakness of the coupling constants λ_a and λ_{er} seems to indicate that there may be room for strengthening them (say, by increasing n) and raising T_c further.

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