# Two-Orbital Model Explains the Higher Transition Temperature of the Single-Layer Hg-Cuprate Superconductor Compared to That of the La-Cuprate Superconductor

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In order to explore the reason why the single-layered cuprates, $La_{2-x}(Sr/Ba)_xCuO_4$ ($T_c \simeq 40$ K) and $HgBa_2CuO_{4+\delta}$ ($T_c \simeq 90$ K) have such a significant difference in $T_c$, we study a two-orbital model that incorporates the $d_{z^2}$ orbital on top of the $d_{x^2-y^2}$ orbital. It is found, with the fluctuation exchange approximation, that the $d_{z^2}$ orbital contribution to the Fermi surface, which is stronger in the La system, works against $d$-wave superconductivity, thereby dominating over the effect of the Fermi surface shape. The result resolves the long-standing contradiction between the theoretical results on Hubbard-type models and the experimental material dependence of $T_c$ in the cuprates.

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The physics of high-$T_c$ superconductivity, despite its long history, harbors rich problems which are still open. Specifically, given the seminal discovery of the iron-based superconductors [1] and their striking material dependence of $T_c$ [2], it should be important as well as intriguing to have a fresh look at the cuprates, which still have the highest $T_c$ to date, to understand their material dependence of the $T_c$. One of the basic problems is the significant difference in $T_c$ within the single-layered materials, i.e., $La_{2-x}(Sr/Ba)_xCuO_4$ with a maximum $T_c$ of about 40 K versus $HgBa_2CuO_{4+\delta}$ with a $T_c \simeq 90$ K. Phenomenologically, it has been recognized that the materials with $T_c \sim 100$ K tend to have "round" Fermi surfaces, while the Fermi surface of the La system is closer to a square shape which implies a relatively better nesting [3,4].

Conventionally, the materials with a rounded Fermi surface have been modeled by a single-band model with large second [$t_2(>0)$] and third [$t_3(<0)$] neighbor hopping integrals, while the "low-$T_c$" La system has been considered to have smaller $t_2$, $t_3$. This, however, has brought about a contradiction between theories and experiments. Namely, while some phenomenological [5] and $t$-$J$ model [6,7] studies give a tendency consistent with the experiments, a number of many-body approaches for the Hubbard-type models with realistic values of on-site $U$ show suppression of superconductivity for large $t_2 > 0$ and/or $t_3 < 0$, as we shall indeed confirm below [8].

To resolve this discrepancy, here we consider a two-orbital model that explicitly incorporates the $d_{z^2}$ orbital on top of the $d_{x^2-y^2}$ orbital. The former component has in fact a significant contribution to the Fermi surface in the La system. We shall show that the key parameter that determines $T_c$ is the energy level difference between the $d_{x^2-y^2}$ and $d_{z^2}$ orbitals, i.e., the weaker the $d_{z^2}$ contribution to the Fermi surface, the better for $d$-wave superconductivity, where a weaker contribution of the $d_{z^2}$ results in a rounded Fermi surface (which in itself is not desirable for superconductivity), but it is the "single-orbital nature" that favors a higher $T_c$ dominating over the effect of the Fermi surface shape for the La system.

Let us start with a conventional calculation for the single-band Hubbard Hamiltonian, $H = \sum_{ij\sigma} t_{ij} c^\dagger_{i\sigma} c_{j\sigma} + U\sum_i n_{i\uparrow} n_{i\downarrow}$. Here we take the nearest-neighbor hopping $-t_1$ ($\simeq$0.4 eV, see Table I) to be the unit of energy, $U = 6$, the temperature $T = 0.03$, and the band filling $n = 0.85$ are fixed, while we vary $t_2 = -t_3$ with $t_2 > 0$. We then apply the fluctuation exchange approximation (FLEX) [9,10] to solve the linearized Eliashberg equation. $T_c$ is the temperature at which the eigenvalue $\lambda$ of the Eliashberg equation reaches unity, so $\lambda$ at a fixed temperature can be used as a measure for the strength of the superconducting instability. We show in Fig. 1 $\lambda$ as a function of $(|t_2| + |t_3|)/|t_1|$ ($=2|t_2|/|t_1|$ here), which just confirm that, within the single-band model, $\lambda$ (hence $T_c$) monotonically decreases with increasing $|t_2|$ and $|t_3|$. A calculation with the dynamical cluster approximation (DCA) shows that a negative $t_2$ works destructively against $d$-wave superconductivity [11], and a more realistic DCA calculation that

TABLE I. Hopping integrals within the $d_{x^2-y^2}$ orbital for the single and two-orbital models, and $\Delta E \equiv E_{x^2-y^2} - E_{z^2}$.

| | One-orbital | | Two-orbital | |
|---|---|---|---|---|
| | La | Hg | La | Hg |
| $t_1$[eV] | −0.444 | −0.453 | −0.471 | −0.456 |
| $t_2$[eV] | 0.0284 | 0.0874 | 0.0932 | 0.0993 |
| $t_3$[eV] | −0.0357 | −0.0825 | −0.0734 | −0.0897 |
| $(\|t_2\| + \|t_3\|)/\|t_1\|$ | 0.14 | 0.37 | 0.35 | 0.41 |
| $\Delta E$[eV] | ··· | ··· | 0.91 | 2.19 |

 

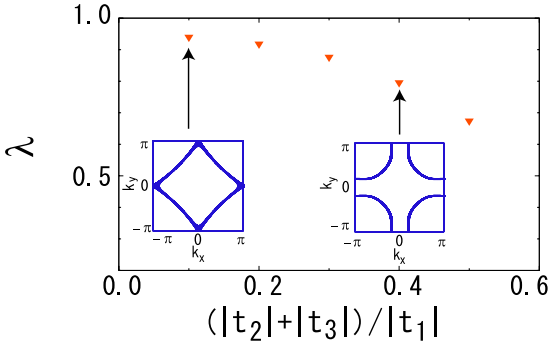


FIG. 1 (color online). FLEX result for the eigenvalue $\lambda$ of the Eliashberg equation for the single-band Hubbard model plotted as a function of $(|t_2| + |t_3|)/|t_1|$, where we take $t_2 = -t_3 > 0$ for $U = 6|t_1|$, $T = 0.03|t_1|$, and the band filling $n = 0.85$. Fermi surfaces are displayed for two cases (indicated by arrows).

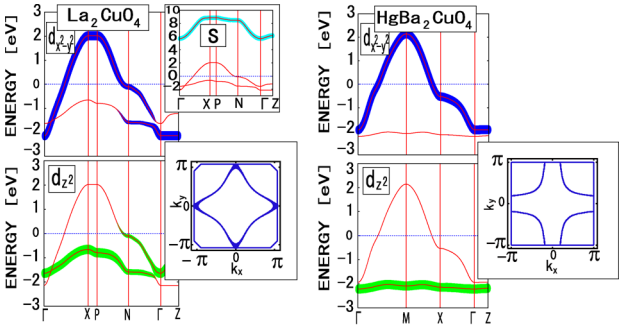


FIG. 2 (color online). The band structure in the two $(d_{x^2-y^2} - d_{z^2})$ orbital model for $La_2CuO_4$ (left) and $HgBa_2CuO_4$ (right). The top (bottom) panels depict the strength of the $d_{x^2-y^2}$ ($d_{z^2}$) characters with thickened lines, while the lower insets depict the Fermi surfaces (for a total band filling $n = 2.85$). The upper inset shows the band structure of the three-orbital model (see text) for La system, where the $4s$ character is indicated.

considers the oxygen $p_\sigma$ orbitals for the La and Hg cuprates also indicates a similar tendency [12]. As mentioned above, this seems to contradict with the experimental results that the materials with larger $t_2$ and $t_3$ have actually higher $T_c$'s [3].

To resolve this, we now introduce the $d_{x^2-y^2} - d_{z^2}$ two-orbital model. For the La system, it has long been known that a band with a strong $d_{z^2}$ character lies rather close to the Fermi energy [13–15]. More recently, it has been discussed in Refs. [3,16] that the shape of the Fermi surface is determined by the energy level of the "axial state" consisting of a mixture of Cu $d_{z^2}$-O $p_z$ and Cu $4s$ orbitals, and that the strength of the $d_{z^2}$ contribution causes the difference in the Fermi surface shape between the La and Hg systems. Namely, the $d_{z^2}$ contribution is large in the La system making the Fermi surface closer to a square, while the contribution is small in the Hg system making the Fermi surface more rounded. In Fig. 2, we show the present, first-principles [17] result for band structures in the two-orbital model for the La and Hg systems, obtained by constructing maximally localized Wannier orbitals [18]. The lattice parameters adopted here are experimentally determined ones for the doped materials [19,20]. We can here confirm that in the La system the main band (usually considered to be the "$d_{x^2-y^2}$ band") has in fact a strong $d_{z^2}$ character on the Fermi surface near the N point, which corresponds to the wave vectors $(\pi, 0)$, $(0, \pi)$ in the Brillouin zone of the square lattice. The $d_{z^2}$ contribution is seen to "push up" the van Hove singularity (vHS) of the main band, resulting in a seemingly well nested (square shaped) Fermi surface. In the Hg system, on the other hand, the $d_{z^2}$ band stays well away from $E_F$, and consequently the vHS is lowered, resulting in a rounded Fermi surface.

If we estimate in the two-orbital model the ratio $(|t_2| + |t_3|)/|t_1|$ within the $d_{x^2-y^2}$ orbitals, we get 0.35 for the La system against 0.41 for Hg (Table I), which are rather close to each other. This sharply contrasts with the situation in which the model is constrained into a single band. There, the Wannier orbital has mainly $d_{x^2-y^2}$ character, but has "tails" with a $d_{z^2}$ character especially for the La system. Then the ratio $(|t_2| + |t_3|)/|t_1|$ in the single-orbital model reduces to 0.14 for La against 0.37 for Hg (Table I), which is just the conventional view mentioned in the introductory part. From this, we can confirm that it is the $d_{z^2}$ contribution that makes the Fermi surface in the La system square shaped, while the "intrinsic" Fermi surface of the high $T_c$ cuprate family is, as in the Hg system, rounded.

Now we come to the superconductivity in the two-orbital model. For the electron-electron interactions, it is widely accepted that the intraorbital $U$ is 7–10$t$ (with $t \sim 0.45$ eV) for the cuprates, so we take $U = 3.0$ eV. The Hund's coupling $J$ (=pair-hopping interaction $J'$) is typically $\sim 0.1U$, so here we take $J = J' = 0.3$ eV, which gives the interorbital $U' = U - 2J = 2.4$ eV [21]. The temperature is fixed at $k_BT = 0.01$ eV. As for the band filling (number of electrons/site), we concentrate on the total $n = 2.85$, for which the main band has 0.85. Here we apply the multiorbital FLEX, as described, e.g., in Ref. [22], for the three-dimensional lattice taking $32 \times 32 \times 4$ $k$-point meshes and 1024 Matsubara frequencies. We first focus on the La system, and investigate how the $d_{z^2}$ orbital affects superconductivity. Namely, while the on-site energy difference, $\Delta E \equiv E_{x^2-y^2} - E_{z^2}$, between the two orbitals is $\Delta E \simeq 0.9$ eV for $La_2CuO_4$ (Table I), we vary the value to probe how the Eliashberg eigenvalue $\lambda$ for $d$-wave superconductivity behaves. The result in Fig. 3 shows that $\lambda$ is small for the original value of $\Delta E$, but rapidly increases with $\Delta E$, until it saturates for sufficiently large $\Delta E$. Hence the superconductivity turns out to be enhanced as the $d_{z^2}$ band moves away from the main band. Note that this occurs despite the Fermi surface becoming more rounded with larger $\Delta E$, namely, the effect of the orbital character (smaller $d_{z^2}$ contribution) dominates over the Fermi surface shape effect. Conversely, the strong $d_{z^2}$ character in the Fermi surface around $(\pi, 0)$, $(0, \pi)$ works destructively against $d$-wave superconductivity. Physi-

cally, the reason for this may be explained as follows. First, although the La system has a better nested Fermi surface, we find that the strength of the antiferromagnetic spin fluctuations (the spin susceptibility obtained in FLEX) in La is only as large as that for Hg. This is intuitively understandable, since the two electrons on nearest-neighbor sites are less constrained to have antiparallel spins in order to gain kinetic energy when two orbitals are active as in La. Second, $d$-wave pairing has a rough tendency for higher $T_c$ in bands that are nearly half filled, whereas the $d_{z^2}$ orbital here is nearly full filled.

We now focus on how the lattice structure affects $\Delta E$ and hence superconductivity. This is motivated by the fact that $\Delta E$ should be controlled by the ligand field, hence by the height, $h_O$, of the apical oxygen above the $CuO_2$ plane [14]. To single out this effect, let us examine the two-orbital model for which we increase $h_O$ from its original value 2.41 Å with other lattice parameters fixed. In Fig. 4(a), which plots the eigenvalue of the Eliashberg equation as a function of $h_O$, we can see that $\lambda$ monotonically increases with the height. As seen from the inset of Fig. 4(b), $\Delta E$ is positively correlated with $h_O$ as expected, and Fig. 4(b) confirms that the increase in $\lambda$ is due to the increase in $\Delta E$ [23]. In these figures, we have also plotted the values corresponding to the Hg system obtained with the actual lattice structure. We can see that, while $h_O \simeq 2.8$ Å for Hg is larger than $h_O \simeq 2.4$ Å for La, $\Delta E \simeq 2.2$ eV for Hg is even larger than $\Delta E \simeq 1.3$ eV, which is the value the La system would take for $h_O = 2.8$ Å. Consequently, $\lambda$ for Hg is somewhat larger than that for the La system with the same value of $h_O$. This implies that there are some effects other than the apical oxygen height that also enhance $\Delta E$ in the Hg system, thereby further favoring $d$-wave superconductivity. In this context, the present result reminds us of the so-called "Maekawa's plot," where a positive correlation between $T_c$ and the level of the apical oxygen $p_z$ hole was observed [24]. Since a higher $p_z$ hole level (i.e., a lower $p_z$ electron level) is likely to lower $E_{z^2}$, the positive correlation between $\Delta E$ and $T_c$ found here is indeed consistent with Maekawa's plot. It can be considered that in La cuprates, a considerable portion of the doped holes go into the apical oxygen $p_z$, and this effect is effectively taken into account in our model. A more detailed study on these issues is now under way, and will be discussed in a separate publication.

Finally, let us discuss the effect of Cu 4$s$ orbital, which is the main component of the "axial state" discussed in Refs. [3,16]. In the present two-orbital model the 4$s$ orbital is effectively incorporated in both of the $d_{x^2-y^2}$ and $d_{z^2}$ orbitals; i.e., the Wannier orbitals have tails that have the 4$s$ character. In order to make the examination more direct, we now consider a three-orbital model that explicitly considers the 4$s$ orbital. The band dispersion for the La system shown in the upper inset of Fig. 2 shows that the 4$s$ band lies well ($\simeq$7 eV) above the Fermi level. Nonetheless, the 4$s$ orbital gives an important contribution to the Fermi surface in that the ratio $(|t_2| + |t_3|)/|t_1|$ within the $d_{x^2-y^2}$ sector in the three-orbital model takes a much smaller value of 0.10, which should imply that it is the path $d_{x^2-y^2} \rightarrow 4s \rightarrow d_{x^2-y^2}$ that gives the effectively large $t_2$, $t_3$, and hence the round Fermi surface, as pointed out previously [3,16]. In this context, it is worth mentioning that the path $d_{x^2-y^2} \rightarrow d_{z^2} \rightarrow d_{x^2-y^2}$ also contributes to $t_2$, $t_3$, but has an opposite sign to the 4$s$ contribution because the $d_{z^2}$ level lies below $d_{x^2-y^2}$, while 4$s$ above $d_{x^2-y^2}$ [25]. So the two contributions to the main band cancel with each other, where the cancellation should be strong when the energy of the $d_{z^2}$ orbital is high as in La.

We now apply FLEX to the three-orbital model varying $\Delta E = E_{x^2-y^2} - E_{z^2}$ as in the two-orbital model, where we fix the on-site energy difference $E_{4s} - E_{z^2}$ at its original value. We have chosen this because a similar three-orbital model constructed for Hg (not shown) shows that the on-site energy difference between the 4$s$ and $d_{x^2-y^2}$ orbitals is smaller than in the La system by about 1 eV, so in the Hg system, both of $E_{x^2-y^2} - E_{z^2}$ and $E_{4s} - E_{x^2-y^2}$ are smaller by about 1 eV, which means that the $d_{z^2}$ and 4$s$ levels shift roughly in parallel relative to $d_{x^2-y^2}$. It can be seen in Fig. 3

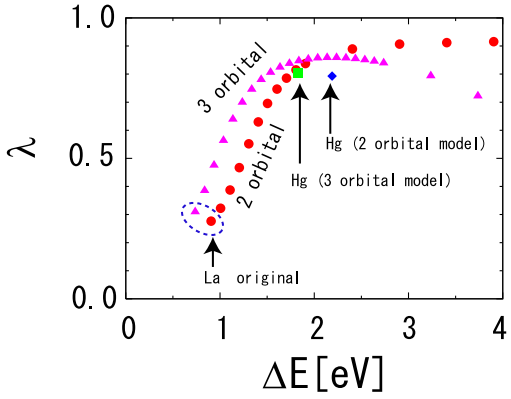


FIG. 3 (color online). The eigenvalue $\lambda$ of the Eliashberg equation for $d$-wave superconductivity is plotted against $\Delta E = E_{x^2-y^2} - E_{z^2}$ for the two-orbital (circles) or three-orbital (triangles) models for $La_2CuO_4$. Corresponding eigenvalues for $HgBa_2CuO_4$ are also indicated.

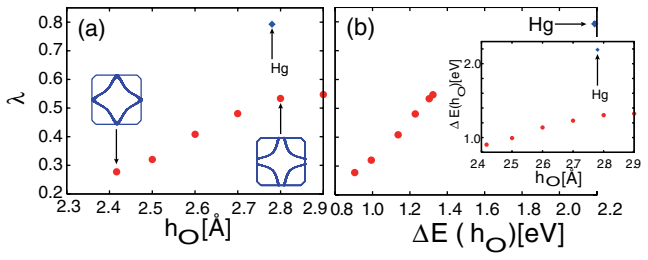


FIG. 4 (color online). The eigenvalue of the Eliashberg equation $\lambda$ (circles) when $h_O$ (a) or $\Delta E$ (b) is varied hypothetically in the lattice structure of $La_2CuO_4$. The diamond indicates the eigenvalue of $HgBa_2CuO_4$. The inset in (b) shows the relation between $h_O$ and $\Delta E$.

that the $\Delta E$ dependence of $\lambda$ in the three-orbital model resembles that of the two-orbital model in the realistic $\Delta E$ range. (When $\Delta E$ becomes unrealistically large, i.e., when $4s$ level is too close to the Fermi level, the Fermi surface becomes too deformed for superconductivity to be retained.) We have also calculated the eigenvalue for the Hg system in the three-orbital model, and obtained a value very similar to that obtained in the two-orbital model, as plotted in Fig. 3. If we summarize the three-orbital results, while the $4s$ orbital has an important effect on the shape of the Fermi surface, this can be effectively included in the $d_{x^2-y^2}$ and $d_{z^2}$ Wannier orbitals in the two-orbital model as far as the FLEX studies are concerned. This contrasts with the case of the $d_{z^2}$ orbital, which, if effectively included in the $d_{x^2-y^2}$ Wannier orbital to construct a single-orbital model, would result in a different result. This conclusion is natural, since the energy difference ($\simeq$1 eV) between $d_{x^2-y^2}$ and $d_{z^2}$ orbitals in the La system is smaller than the electron-electron interaction, which is why the $d_{z^2}$ orbital has to be explicitly considered in a many-body analysis, while the energy difference ($\simeq$7 eV) between $d_{x^2-y^2}$ and $4s$ orbitals is much larger than the electron-electron interaction, so that the $4s$ orbital can effectively be integrated out before the many-body analysis. So the message here is that the two-orbital ($d_{x^2-y^2} - d_{z^2}$) model suffices to discuss the material dependence of the $T_c$ in the cuprates. Whether the effect of the $d_{z^2}$ orbital can be further incorporated in the on-site $U$ or off-site $V$ values (i.e., material-dependent interaction values) in an effective, single-band model is a future problem.

To summarize, we have introduced a two-orbital model to understand the material dependence of $T_c$ in the cuprates. We have shown that the key parameter is the energy difference between the $d_{x^2-y^2}$ and $d_{z^2}$ orbitals, where the smaller the contribution of the $d_{z^2}$ orbital, the better for $d$-wave superconductivity, with the orbital-character effect superseding the effect of the Fermi surface shape. It is intriguing to note that the two high $T_c$ families, cuprates and iron pnictides, exhibit material dependence of $T_c$ that, according to the present study and Ref. [26], owes to the material dependent multiorbital band structures.

In the present view, the Hg cuprate is ''ideal'' in that the $d_{z^2}$ band lies far below the Fermi level. Nevertheless, there is still room for improvement: as mentioned in the outset, within single-orbital systems higher $T_c$ can be obtained for smaller $t_2$ and $t_3$. It may be difficult to make $t_2$ and $t_3$ smaller in the cuprates, since they are intrinsically large as far as the Cu $4s$ orbital is effective. Conversely, we can predict that materials with an isolated single band that has smaller $t_2$ and $t_3$ should accommodate even higher $T_c$ than the Hg cuprate, provided that the electron interaction is similar to those in the cuprates.

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