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Three-orbital study on the orbital distillation effect in the high T_c cuprates

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Abstract

Our recent study has revealed that the mixture of the d_{z^2} orbital component into the Fermi surface suppresses T_c in the cuprates such as La_2CuO_4 . We have also shown that applying hydrostatic pressure enhances T_c due to smaller mixing of the $\text{Cu}4s$ component. We call these the "orbital distillation" effect. In our previous study, the $4s$ orbital was taken into account through the hoppings in the $d_{x^2-y^2}$ sector, but here we consider a model in which of the $d_{x^2-y^2}$, d_{z^2} and $4s$ orbitals are all considered explicitly. The present study reinforces our conclusion that smaller $4s$ hybridization further enhances T_c .

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1. Introduction

One of the important, and still not fully understood, problems associated with the high- T_c cuprates is how to optimize their superconducting transition temperature, T_c [1,2]. It is well known that T_c varies strongly with the number of CuO_2 layers. However, even within the single layered cuprates, it is also known that there is significant material dependence of T_c , e.g., La_2CuO_4 ($\text{La}214$; $T_c \sim 40\text{K}$) and $\text{HgBa}_2\text{CuO}_4$ ($\text{Hg}1201$; $T_c \sim 100\text{K}$) [3]. In $\text{La}214$, the shape of the Fermi surface is observed to be square compared to that of $\text{Hg}1201$, so a warped Fermi surface apparently favours superconductivity [4,5]. This, however, conflict with theoretical many-body studies of Hubbard-type models that indicate warped Fermi surfaces are unfavourable for superconductivity. This has remained a long-standing puzzle in the field of the study of the cuprates [6].

As for the shape of the Fermi surface, some studies have pointed out that the contribution of the d_{z^2} [7-9] orbital and hence the apical oxygen height (h_O) are important [10-18]. On the other hand, some theoretical models which include the effect of the d_{z^2} orbital have explained the material dependence of T_c [19-22]. However, there seems to have been no persuasive solution for this problem between T_c and the shape of Fermi surface, at least, within many-body approaches for the Hubbard-type models with realistic values of the on-site U [23].

To solve this puzzle, we have introduced a $d_{x^2-y^2}$ - d_{z^2} two-orbital model in which not only the conventionally-considered $d_{x^2-y^2}$ Wannier orbital but also the d_{z^2} orbital is explicitly considered. By applying many-body analysis to the two-orbital model, we have shown that the admixture of the d_{z^2} orbital in the Fermi surface Bloch states is crucial in understanding the material dependence of T_c in the cuprates, i.e., in relatively low T_c materials such as $\text{La}214$, the d_{z^2} orbital suppresses the warping of the Fermi surface which would enhance T_c . However the increase of the d_{z^2}

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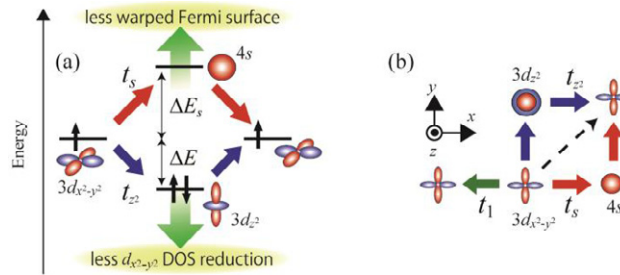


Fig.1 (a) Definitions of ΔE_s , t_s , ΔE and t_{z^2} . The thickest arrows represent the orbital distillation (see text). Two paths via Cu4s or Cu3d_{z²} (solid arrows t_s and t_{z^2}) which give the effective diagonal hopping (dashed arrow).

component of the density of states to the Fermi surface overcomes this enhancement and suppresses T_c . As a result, La214 with a relatively squared Fermi surface nevertheless exhibits a low T_c [17,18].

More recently, we have found in a study of the hydrostatic pressure enhancement of T_c [23,24] that the Cu4s orbital significantly affects T_c in cuprates in which the d_{z^2} orbital mixture is small[25]. In that study, the contribution from the 4s orbital was implicitly included in the hoppings of the $3d_{x^2-y^2}$ and the $3d_{z^2}$ orbitals, namely, a smaller 4s contribution reduces the second and third nearest neighbour hoppings between $3d_{x^2-y^2}$ Wannier orbitals (as will be explained later in detail), thereby reducing the warping of the Fermi surface and enhancing T_c . Combining the effect of $3d_{z^2}$ and 4s, we have concluded that the “orbital distillation” effect enhances T_c in the cuprates.

In the present work, we consider the 4s orbital explicitly in the model Hamiltonian, which now contains all of the $3d_{x^2-y^2}$, $3d_{z^2}$ and 4s orbitals, to study the effect of orbital distillation on superconductivity. Our ultimate goal along this line of study is to consider the possibility of new materials which can have T_c even higher than the cuprates.

2. Calculation methods

First, we perform first-principles band calculation to obtain the structural parameter of HgBa₂CuO₄ [26]. Namely, we calculate the total energy varying the lattice constants, and fit the result with the standard Burch-Marnaghan formula [27] to obtain the structure at the most stable point. From this we obtain the crystal structure, which turns out to be within 1% discrepancy from the experimentally determined lattice constants [28]. In the Hg compound, the effect of the $3d_{z^2}$ orbital is negligible because of the large level offset ΔE between the $3d_{x^2-y^2}$ and the $3d_{z^2}$ orbitals, so that we can focus on the effect of the 4s orbital. From this we construct maximally localized Wannier orbitals [29,30] to obtain the hopping integrals of the present three-orbital model, in which we consider the $3d_{x^2-y^2}$ orbital, the $3d_{z^2}$ orbital and the 4s orbital explicitly as discussed above.

2.1. Applying the fluctuation exchange approximation

The electron-electron interactions considered in the present study are the following: the on-site intra-orbital Coulomb repulsion U , the inter-orbital repulsion U' , the Hund's coupling J and pair-hopping J' . Here we observe the orbital SU(2) requirement, $U-U'=2J$. Here we fix the values at $U=3.0$ eV, $U'=2.4$ eV and $J=J'=0.3$ eV. In the recent estimations by first-principles, U in cuprates is considered to be 7-10 t (namely, about 3-4.5 eV) and $J(J')\sim 0.1U$, so the values chosen here are within the widely accepted range.

Then we apply the fluctuation exchange approximation (FLEX)[31-33] to the present model to obtain the Green's function renormalized by the many-body self-energy correction. In FLEX, the contributions from bubble and ladder diagrams are included in the self-energy, for which we solve the Dyson's equation in a self-consistent manner. Then we substitute the Green's function to linearized Eliashberg equation to evaluate the strength of the superconducting instability. The eigenvalue λ of Eliashberg equation reaches unity at T_c , so we can use λ at a fixed temperature as a measure of T_c . We set $T=0.015$ eV, and the number of electrons per copper site to be $n=2.85$ (i.e., 15% doped in the main band). We take the $32\times 32\times 4$ k -point meshes and 1024 Matsubara frequencies.

3. Results and discussion

3.1. Effects of the Cu4s orbital

Due to the symmetry of the wavefunctions, the Cu4s orbital can mediate the electron hopping path $3d_{x^2-y^2} \rightarrow 4s \rightarrow 3d_{x^2-y^2}$, so that the 4s orbital effectively enhances the second and third neighbour hoppings between $3d_{x^2-y^2}$ orbitals (Fig.1) [11]. In our previous two-orbital model, the 4s orbital effect is implicitly included in the $3d_{x^2-y^2}$ and the

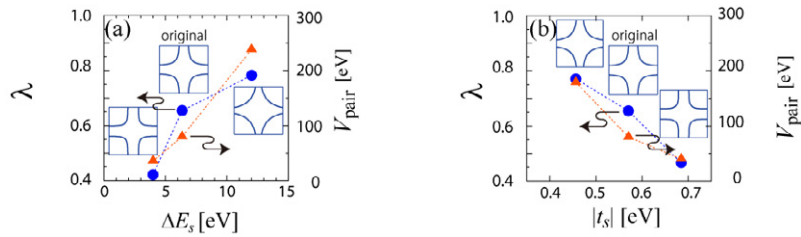


Fig.2 Eigenvalue λ (circle) and the pairing interaction V_{pair} (triangle) plotted against ΔE_s (with a fixed $|t_s|=0.57$ eV ; left panel) or $|t_s|$ (with a fixed $\Delta E_s=6.4$ eV; right) for $\text{HgBa}_2\text{CuO}_4$.

$3d_{z^2}$ Wannier orbitals, where the $4s$ effect is taken into account mainly via the second (t_2) and third (t_3) neighbour hoppings of the $3d_{x^2-y^2}$ Wannier orbital sector, and hence the warping of the Fermi surface [17].

In the present three orbital model, the warping of the Fermi surface is controlled by two parameters; the level offset ΔE_s and the nearest neighbour inter-orbital hopping t_s between the $3d_{x^2-y^2}$ and $4s$ orbitals. In the following, we vary these two parameters hypothetically to see more directly the basic mechanism of T_c suppression by the $4s$ orbital.

3.2. Effects of ΔE_s and t_s

First, we focus on the effect of ΔE_s . In the panel (a) of Fig.2, we plot the relationship between λ and ΔE_s . We can see that λ increases with ΔE_s . To explore the mechanism for the enhanced T_c , we have also plotted the maximum value of the pairing interaction V_{pair} along with the shape of Fermi surface at $k_z=0$ in Fig.2. V_{pair} is seen to increase as the warping of the Fermi surface is reduced, i.e., V_{pair} is enhanced by the nesting of the Fermi surface. In other words, the reduction of the $4s$ orbital mixing improves the Fermi surface nesting and this enhances the spin-fluctuation mediated superconductivity [6].

As for the effect of the hopping t_s , we obtain a similar result (panel (b) in Fig.2), namely the reduction of $|t_s|$ makes the Fermi surface less warped, hence higher T_c . From this viewpoint, *searching for materials with reduced t_s* may be a promising way to obtain high T_c materials. Here let us make a comment on the pressure effect studied in our previous paper [25]. Both t_s and ΔE_s increase with applying hydrostatic pressure, but the effect of the increased t_s is nearly cancelled out by the increase of the nearest neighbour hopping t_1 (note that the warping of the Fermi surface is determined by the *ratio* of the nearest and distant hoppings), so that only the effect of the ΔE_s increase remains, which enhances (suppresses) the T_c (warping of Fermi surface).

3.3. Orbital distillation

In some cuprates such as La214, the bending of the Fermi surface induced by the $4s$ orbital is cancelled by the $3d_{z^2}$ orbital because ΔE and ΔE_s have opposite signs (see Fig.1 (a)). Namely, the $4s$ orbital mediates a diagonal hopping whose sign is opposite to the one mediated by the $3d_{z^2}$ orbital [17,18]. However, for small ΔE , the $3d_{z^2}$ orbital component partially replaces the $3d_{x^2-y^2}$ orbital component, and this reduction of the density of $3d_{x^2-y^2}$ states overcomes the T_c enhancement associated with the reduced warping of the Fermi surface. The reason why the $3d_{z^2}$ orbital cannot be integrated out before the many-body analysis (in contrast to the case of $4s$ orbital) comes from the fact that ΔE ($\sim 1\text{eV}$) is considerably smaller than U ($\sim 3\text{eV}$), while ΔE_s ($\sim 7\text{eV}$) is much larger [18].

Note that there are two effects that can enhance T_c when the $4s$ contribution is reduced : (i)the reduction of the warping due to less $3d_{x^2-y^2}$ - $4s$ hybridization, and (ii)the increase of the $d_{x^2-y^2}$ density of states (the same effect as in the increase of ΔE). Actually, the $4s$ density of states in the Fermi surface is small (compared to that of d_{z^2} in La214), so that effect (i) governs over effect (ii). This is confirmed by the fact that λ is very close between three ($3d_{x^2-y^2}$ - $3d_{z^2}$ - $4s$) and two($3d_{x^2-y^2}$ - $3d_{z^2}$, $4s$ effectively included) orbital models having the same Fermi surface shape[17,18].

Now, we can unify the present results into a simple picture of ‘‘orbital distillation’’. While the $3d_{x^2-y^2}$ orbital is supposed to compose a nearly square shape Fermi surface in itself, the Fermi surface is bent by the $4s$ orbital in the actual materials. Thus a strategy for having higher T_c is to reduce the effects of d_{z^2} and $4s$ orbitals *simultaneously* (as symbolized by thick arrows on the Fig.1 (a)), and we propose this ‘‘orbital distillation’’ as a key to optimize T_c .

4. Conclusion

In summary, we have studied the $3d_{x^2-y^2}$ - $3d_{z^2}$ - $4s$ three-orbital model for $\text{HgBa}_2\text{CuO}_4$ derived from first principles. Applying FLEX approximation to this model, we have shown that the increase in the level offset ΔE_s enhances T_c by

suppressing the warping of the Fermi surface, and that the reduction of the inter-orbital hopping t_s works in a similar way. Both results can be summarized in a single picture in which the $4s$ orbital makes the Fermi surface more warped and hence suppresses T_c . This result indicates the same tendency as those calculations adopting the single-orbital Hubbard Hamiltonian tuning t_2 and t_3 in the $3d_{x^2-y^2}$ orbital sector because the $4s$ orbital can be integrated out before the many body analysis. Still, we stress that the present study has given access to T_c -controlling parameters that are *more directly connected to the lattice structure, orbital symmetry of the constituent element*. It may be difficult to increase ΔE_s to greater extent in actual materials, but we believe there remains a possibility of reducing the inter-orbital hopping t_s . If such an orbital distillation is realized in some materials other than the cuprates, thereby preserving the “favourable conditions” enjoyed by the cuprates (near half-filling, $U/8t \sim 1$, square lattice), then there is a possibility that T_c may be optimized even further.

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