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 $d_{xz}$ orbital component into the Fermi surface suppresses $T_c$ in the cuprates such as La$_2$CuO$_4$. We have also shown that applying hydrostatic pressure enhances $T_c$ due to smaller mixing of the Cu4$s$ component. We call these the "orbital distillation" effect. In our previous study, the 4$s$ orbital was taken into account through the hoppings in the $d_{xz,zy}$ sector, but here we consider a model in which the $d_{xz,zy}$, $d_{xy}$ and 4$s$ orbitals are all considered explicitly. The present study reinforces our conclusion that smaller 4$s$ hybridization further enhances $T_c$.

Keywords: cuprates; superconductivity; two-orbital model; band calculation; fluctuation exchange approximation (FLEX); spin fluctuation

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$_2$CuO$_4$ (La214; $T_c \approx 40$K) and HgBa$_2$CuO$_4$ (Hg1201; $T_c \approx 100$K)[3]. In La214, the shape of the Fermi surface is observed to be square compared to that of Hg1201, 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 $d_{xz}$ [7,8,9] orbital and hence the apical oxygen height ($h_0$) are important[10,11,12,13,14,15,16,17,18]. On the other hand, some theoretical models which include the effect of the $d_{xz}$ orbital have explained the material dependence of $T_c$ [19,20,21,22]. 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 $d_{xz,yz}$-$d_{xy}$ two-orbital model in which not only the conventionally-considered $d_{xz}$ Wannier orbital but also the $d_{xy}$ orbital is explicitly considered. By applying many-body analysis to the two-orbital model, we have shown that the admixture of the $d_{xz}$ 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 La214, the $d_{xz}$ orbital suppresses the warping of the Fermi surface which would enhance $T_c$. However, the increase of the $d_{xy}$ 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 Cu4$s$ orbital significantly affects $T_c$ in cuprates in which the Cu4$s$ orbital mixture is small[25]. In that study, the contribution from the 4$s$ orbital was implicitly included in the hoppings of the $3d_{xz,zy}$ and the $3d_{xy}$ orbitals, namely, a smaller 4$s$ contribution reduces the second and third nearest neighbour hoppings between $3d_{xz,zy}$ 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 $3d_{xz}$ and 4$s$, we have concluded that the “orbital distillation” effect enhances $T_c$ in the cuprates.

In the present work, we consider the 4$s$ orbital explicitly in the model Hamiltonian, which now contains all of the $3d_{xz,zy}$, $3d_{xy}$ and 4$s$ 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.

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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 3dₓ²₋ᵧ² orbital is negligible because of the large level offset ΔE between the 3dₓ²₋ᵧ² and 3dₓz⁻rᵧz 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 3dₓ²₋ᵧ² orbital, the 3dₓz⁻rᵧz orbital and the 4s orbital explicitly as discussed above.

1.1. 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=0.3 eV. In the recent estimations by first-principles, U in cuprates is considered to be 7-10e (namely, about 3-4.5 eV) and J(J′)~0.1U, so the values chosen here are within the widely accepted range.

Then we apply the fluctuation exchange approximation (FLEX) [31,32,33] 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 Tc, so we can use λ at a fixed temperature as a measure of Tc. We set T=0.015eV, and the number of electrons per copper site to be n=2.85 (i.e., 15% doped in the main band). We take the 32×32×4 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 3dₓ²₋ᵧ²→4s→3dₓ²₋ᵧ², so that the 4s orbital effectively enhances the second and third neighbour hoppings between 3dₓ²₋ᵧ² orbitals (Fig.1) [11]. In our previous two-orbital model, the 4s orbital effect is implicitly included in the 3dₓ²₋ᵧ² and the 3dₓz⁻rᵧz Wannier orbitals, where the 4s effect is taken into account mainly via the second (t₂) and third (t₃) neighbour hoppings of the 3dₓ²₋ᵧ² 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, and the nearest neighbour inter-orbital hopping tₓ between the 3dₓ²₋ᵧ² and 4s orbitals. In the following, we vary these two parameters hypothetically to see more directly the basic mechanism of Tc suppression by the 4s orbital.
Applying the warped Fermi surface, the Fermi level offset is decreased (~3eV) is expected to enhance the Tc. We can see that λ increases with ΔEg. To explore the mechanism for the enhanced Tc, we have also plotted the maximum value of the pairing interaction Vpair along with the shape of Fermi surface at k∥=0 in Fig.2. Vpair is seen to increase as the warping of the Fermi surface is reduced, i.e., Vpair 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,, we obtain a similar result (panel b in Fig.2), namely the reduction of |t,| makes the Fermi surface less warped, hence higher Tc. From this viewpoint, searching for materials with reduced t, may be a promising way to obtain high Tc materials. Here let us make a comment on the pressure effect studied in our previous paper [25]. Both t, and ΔEg increase with applying hydrostatic pressure, but the effect of the increased t, is nearly cancelled out by the increase of the nearest neighbour hopping t (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 ΔEg increase remains, which enhances (suppresses) the Tc (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, orbital because ΔE and ΔEg 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, orbital [17,18]. However, for small ΔE, the 3d, orbital component partially replaces the 3d, orbital component, and this reduction of the density of 3d, states overcomes the Tc enhancement associated with the reduced warping of the Fermi surface. The reason why the 3d, orbital cannot be integrated out before the many-body analysis (in contrast to the case of 4s orbital) comes from the fact that ΔE (~1eV) is considerably smaller than U (~3eV), while ΔEg (~7eV) is much larger [18].

Note that there are two effects that can enhance Tc when the 4s contribution is reduced: (i) the reduction of the warping due to less 3d, orbital mixing, and (ii) the increase of the 3d, 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, in La214), so that effect (i) governs over effect (ii). This is confirmed by the fact that λ is very close between three (3d, orb., 3d, 4s) and two(3d, 4s) 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, 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 Tc is to reduce the effects of d, 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 Tc.

4. Conclusion

In summary, we have studied the 3d, 3d, 4s three-orbital model for HgBa2CuO4 derived from first principles. Applying FLEX approximation to this model, we have shown that the increase in the level offset ΔE enhances Tc, by suppressing the warping of the Fermi surface, and that the reduction of the inter-orbital hopping t, works in a similar manner.
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 $t_1$ and $t_2$ 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 $\Delta E$ to greater extent in actual materials, but we believe there exists a possibility of reducing the inter-orbital hopping $t_3$. If such an orbital distillation is realized in some materials other than the cuprates, thereby preserving the “favorable conditions” enjoyed by the cuprates (near half-filling, $U/8t \approx 1$, square lattice), then there is a possibility that $T_c$ may be optimized even further.

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