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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_{z2} 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 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 $d_{x^2-y^2}$ sector, but here we consider a model in which of the $d_{x^2-y^2}$, d_{z^2} and 4*s* orbitals are all considered explicitly. The present study reinforces our conclusion that smaller 4*s* hybridization further enhances T_c.

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Keywords: cuprates; superconductivity; two-orbital model; band calculation; flucutuation 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_2CuO_4 (La214; T_c ~40K) and $HgBa_2CuO_4$ (Hg1201; T_c ~100K)[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_{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 La214, 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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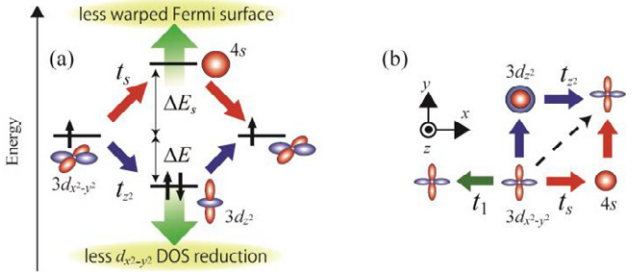

Fig.1 (a) Definitions of ΔE_s, t_s, ΔE and t_{z^2}. The thickest arrows represent the orbital distillation (see text). Two paths via Cu4*s* or Cu3d_{z^2} (solid arrows t_s and t_{z^2}) which give the effective diagonal hopping (dashed arrow).

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 Cu4*s* 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 4*s* orbital was implicitly included in the hoppings of the $3d_{x^2-y^2}$ and the $3d_{z^2}$ orbitals, namely, a smaller 4*s* 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 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 $3d_{x^2-y^2}$, $3d_{z^2}$ 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.

2. Calculation methods

First, we perform first-principles band calculation to obtain the structural parameter of $HgBa_2CuO_4$ [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 4*s* 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 4*s* 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 7-10t (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)[31-33] 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=0.015eV, 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 Cu4*s* orbital can mediate the electron hopping path $3d_{x^2-y^2}$ →4*s*→$3d_{x^2-y^2}$, so that the 4*s* 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 4*s* orbital effect is implicitly included in the $3d_{x^2-y^2}$ and the

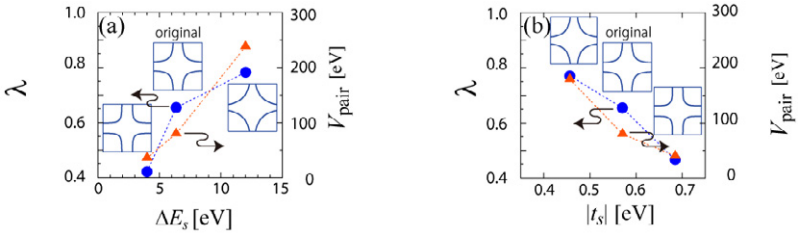

Fig.2 Eigenvalue λ(circle) and the pairing interaction V_{pair}(triangle) plotted against ΔE_s(with a fixed $|t_s|$=0.57 eV ; left panel) or $|t_s|$(with a fixed ΔE_s =6.4 eV; right) for $HgBa_2CuO_4$.

$3d_{z^2}$ Wannier orbitals, where the 4*s* 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 4*s* orbitals. In the following, we vary these two parameters hypothetically to see more directly the basic mechanism of T_c suppression by the 4*s* 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 4*s* 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 4*s* orbital) comes from the fact that ΔE (~1eV) is considerably smaller than U (~3eV), while ΔE_s(~7eV) is much larger [18].

Note that there are two effects that can enhance T_c when the 4*s* contribution is reduced : (i)the reduction of the warping due to less $3d_{x^2-y^2}$-4*s* 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 4*s* 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 ($3d_{x^2-y^2}$-$3d_{z^2}$-4*s*) and two($3d_{x^2-y^2}$-$3d_{z^2}$, 4*s* 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 4*s* orbital in the actual materials. Thus a strategy for having higher T_c is to reduce the effects of d_{z^2} and 4*s* 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 $3d_{x^2-y^2}$-$3d_{z^2}$-4*s* three-orbital model for $HgBa_2CuO_4$ 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 4*s* 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 4*s* 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, $U/8t$~1, square lattice), then there is a possibility that T_c may be optimized even further.

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