Control of Superconducting Correlations in High-Tc Cuprates

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(Received March 24, 2022)

A strategy to enhance d-wave superconducting correlations is proposed based on our numerical study for correlated electron models for high-Tc cuprates. We observe that the pairing is enhanced when the single-electron level around \((\pi, 0)\) is close to the Fermi level \(E_F\), while the d-wave pairing interaction itself contains elements to disfavor the pairing due to shift of the \((\pi, 0)\)-level. Angle-resolved photoemission results in the cuprates are consistently explained in the presence of the d-wave pairing interaction. Our proposal is the tuning of the \((\pi, 0)\)-level under the many-body effects to \(E_F\) by optimal design of band structure.

KEYWORDS: high-Tc superconductivity, pairing correlations, single-electron level, angle-resolved photoemission

We report a new aspect of control of superconducting correlations in high-Tc cuprates. Our new observation in the light of numerical studies is about the critical interplay between single-electron level at wave vector \((\pi, 0)\) and \((0, \pi)\) and electron-pair hopping processes, which are usually overlooked. One potential implication is that the superconducting transition temperature \(T_c\) may be controlled by tuning the single-electron level and the pair-hopping coupling constant in a self-consistent fashion. This could provide a guideline for the quest of new materials with higher \(T_c\).

Since the first report of high-Tc superconductivity in 1986, various materials with the common CuO\(_2\) layer structure have been found to show superconductivity with \(T_c\) up to 133 [K] in mercury compounds and 164 [K] under pressure. One noticeable character common among the high-Tc family is the presence of a flat band around the \((\pi, 0)\) and equivalent \((0, \pi)\) points in the Brillouin zone. The anomalous flat dispersion determined by the angular-resolved photoemission (ARPES) experiments may be fitted by a \(k^4\)-type form and numerical calculations on the two-dimensional (2D) Hubbard and t-J models indicate that it is due to strong electron-electron correlations rather than band effects. ARPES data in fact show the d-wave gap starts growing from the flat-dispersion region in underdoped samples suggesting a key role of this region. However, in the insulating or very lightly doped samples, the broad quasiparticle peak at \((\pi, 0)\) appears substantially lower than the level at \((\pi/2, \pi/2)\), which implies rapid change in the shape of the Fermi surface with increasing doping. Experimentally, the rigid band picture thus fails and it is yet clear how the electron correlation and pairing interaction effects modify the single-particle dispersion.

In this letter, we first show how the dynamical superconducting correlations are enhanced by controlling the single-electron level at the wave vector \(\mathbf{k} = (\pi, 0)\), and we will show later that self-consistent determination of the flat-dispersion level around \((\pi, 0)\) under strong correlation effects opens a way to control the enhancement. We employ the 2D t-J model for our numerical investigation, and introduce the second and third neighbor hoppings in order to control this energy level:

\[
H_{IJ} = -t \sum_i \sum_{\sigma} c^\dagger_{i \sigma} c_{i+\sigma} + J \sum_i S_i \cdot S_i,
\]

where the hopping integrals are \(t_{IJ} = t\) for the nearest-neighbor pairs, \(t_1\) and \(t_2\) for the second and third neighbor pairs, respectively, and otherwise 0. The values of \(t\) and \(J\) are determined from various experiments, and a good estimate is \(t=0.3\) eV and \(J=0.12\) eV in all high-Tc compounds. Hereafter, we will use these values unless mention explicitly, and measure energy in units of \(t\). The single-electron kinetic energy is then given by \(\epsilon_0(k) = -2t(k_x \cos k_x + \cos k_y) - 4t_1 \cos k_x \cos k_y - 2t_2 \cos 2k_x + \cos 2k_y\), and the tight-binding Fermi surface changes its shape at half filling from the \(\pi/4\)-rotated square due to the added \(t_1\)- and \(t_2\)-terms. Fermi surface in various high-Tc compounds has been determined by ARPES experiments and based on comparison with the band calculation: for example, for YBa\(_2\)CuO\(_7\), see Ref. 10. A couple of previous studies argue that these distant hoppings alone account for the change in Fermi surface. Another explanation is provided for the deep \((\pi, 0)\)-level, based on the spin-charge separation scenario. We take an alternative interpretation on this difference to account for superconducting correlations.

In order to investigate the enhancement of superconductivity, we have calculated its dynamic correlation function, \(P_\sigma(\omega)\), instead of the usual equal-time correlations. It is essential to separate the coherent part to measure the enhancement, since only the low-energy dynamics of Cooper pairs is relevant to the superconducting transition. Typical results are shown in Figure 1 for various \(t_1\) and \(t_2\) for creating a spin-singlet hole pair on neighboring sites, \(\Delta \equiv \sum_{j, \alpha} f_\sigma(a)c^\dagger_{j \gamma}c^\dagger_{j+\delta \gamma} + \text{H.c.}\) with the \(d_{x^2-y^2}\) form factor \(f_\sigma(a) = +1\) \((-1)\) when \(a = \pm x\) \((\pm y)\). This is calculated at zero temperature from the ground-state wave function obtained by exact diagonalization for the \(4\times4\)-site cluster. The cluster is chosen such that we...
can compare the electron level between the two characteristic k-points on the Fermi surface at half filling, (π, 0) and (π/2, π/2). Note that in the noninteracting system, the difference in kinetic energy, ε0(π, 0) − ε0(π/2, π/2), is given by u0 ≡ 4(t1 − 2t2).

An important result is that upon doping a single hole pair into the half-filled insulator the coherent peak Pu holds. For u0 > 0, this is a clear evidence of the evolution of coherence of hole-pair motion. On the other hand, when u0 is negative and large, the coherent peak loses its weight, which means the motion of the hole pair is substantially damped. We can easily understand this behavior by considering the single-electron level. For larger u0, ε0(k) is higher around (π, 0) and the energy cost of creating a hole pair is smaller there. The d2−y2-wave Cooper pairs under consideration consist mainly of those holes, and consequently the superconducting correlations are enhanced. The u0-dependence is no longer monotonic when a hole pair is added to the systems already doped, as shown in Fig. 1 for the doping δ=0.125, but a similar interpretation holds. For u0 > 0, electrons have been already removed from the (π, 0) and (0, π) points at finite dopings, and an additional hole pair can hardly reside around these k-points. On the other hand, when u0 < 0, electrons are removed first from the (±π/2, ±π/2) and then from (π, 0) and (0, π) upon hole doping. Therefore, when u0 is negative, the electron occupancy at a finite doping may be such that the (π, 0)-level is still below and close to the Fermi energy. When this is the case, the superconducting correlations will be enhanced.

Since the above discussion on the single-electron level is within the free electron picture, we have to check it directly through the single-electron Green’s function. For the same set of parameters as in Fig. 1, the spectral function of the Green’s function, A_k(ω), is calculated and the results are plotted in Fig. 2. Fermi energy is here set to be zero, EF=0. Sharp peaks near EF are of quasiparticle or quasihole, and a broad incoherent background is spread at higher |ω|. Here, we define the difference in the quasiparticle level u ≡ ε(π, 0) − ε(π/2, π/2), from the peak positions in A_k(ω). The observed u-dependence qualitatively support the picture explained before. The quasiparticle level at (π, 0) shifts downwards with decreasing u0, despite the reduction of the size of the shift. The general trend holds that the (π, 0)-level is close to EF when the pairing correlations are enhanced. Similar behavior is also observed at other J/t’s. The reduction of the shift is due to electron-electron correlations, since single-electron hopping processes lose dispersion and coherence near half filling. We note that the t-J model with t1=t2=0 gives u ∼ 0, and this degeneracy also remains in much larger systems of the Hubbard model.

As is mentioned above, the ARPES results suggest that the (π, 0)-level is substantially lower than the (π/2, π/2)-level in the insulating or very lightly doped samples. From the above argument, if we employ the t-J model, the deep (π, 0)-level requires that the distant hopping satisfies u0 < 0 as in Refs. 36, 37. However, if this is the case, the pairing correlations are not optimally enhanced at realistic dopings around δ ≈ 0.15. Experimentally, the (π, 0)-level quickly rises up to the (π/2, π/2)-level with further doping. This behavior is difficult to explain in the rigid band picture, evidence of correlation effects presumably including strong pairing interaction.

To incorporate such pairing effects, we now examine microscopic processes which account for generic feature of d-wave pairing by employing the square of local kinetic energy. A part of similar interaction has been studied by several authors, 38 but many features remain unexplored. The Hamiltonian, say the t-J-W_d model, reads

\[ H_{I,tW_d} = H_{I,t} - W_d \sum_j \left( \sum_{\alpha, \sigma} f_d(a) c_{j \sigma}^\dagger c_{j+\alpha \sigma} + \text{H.c.} \right)^2, \]  

where a labels four nearest neighbors of each site, and the form factor f_d(a) is d_{x^2-y^2}-wave like as defined before.
control of superconducting correlations in high-Tc cuprates

The $W_d$-term consists of two-site terms and three-site terms. The former renormalizes the superexchange coupling constant as $J_{dr}=J+8W_d$, and its effects could be contained in those of the $J$-term. On the contrary, the three-site terms have new effects. Since it may be rewritten into the form of electron-pair hoppings, it is natural to expect further enhancement of superconductivity due to these terms. This $W_d$-term may be derived as the low-energy effective Hamiltonian starting from the $d$-$p$ model as Zhang-Rice construction of the $J$-term, but has customarily been omitted without examining its relevance. One may also obtain the three-site term starting from the single-band Hubbard model in the limit of strong on-site repulsion but then with a constant form factor, $f(\delta)$, and its effects have been studied by several authors. Generally, starting from the $d$-$p$ model, the $W$-term with the $d_{x^2-y^2}$-wave form factor, $W_d$, and that with the constant form factor, $W_s$, are both obtained. Figure 3 sketches two of typical fundamental processes contributing to the three-site terms. The difference of the two contributes to the $W_d$-term, while the sum leads to the $W_s$-term. Although usually neglected, these processes are in fact of lower order than those for superexchange, meaning a considerable size of the coupling constants, $W_d$ and $W_s$.

In the following we will demonstrate based on numerical calculations an important role of the $W_d$-term for the enhancement of superconducting correlations. We also show that such pairing interaction simultaneously induces renormalization of single-particle dispersion. In contrast to the $W_d$-term, the $W_s$-term does not affect the single-electron level at $k$’s along the Fermi surface at half filling. Therefore, the deep $(\pi,0)$-level observed in experiments again require $u_0<0$ if $W_d=0$ and $W_s \neq 0$, and this implies that the enhancement of superconductivity remains not prominent. On the other hand, the $W_d$-term drives pairing instability, and simultaneously shifts the single-particle $(\pi,0)$-level downwards relative to $(\pi/2,\pi/2)$ as we show below. It turns out that these two phenomena are compatible only when the $W_d$-term is playing the driving mechanism of pairing. Based on these observations, we examine how to optimize superconductivity with the $W_d$-term.

Figure 4 shows the dynamic superconducting correlations of the $t$-$J$-$W_d$ model, and here $t_1 = t_2 = 0$. The growth of the coherent peak $P_d(\omega)$ upon switching $W_d$ is noticeable, and at the same time the weight of the incoherent background at higher energies is substantially suppressed. Thus the effects of the $W_d$-term on the enhancement of superconducting correlations are clear.

The $W_d$-term modifies single-electron dynamics as well. Electrons hop using this term to second and third neighbor sites with and without spin flip accompanied, where the single-electron part of the $W_d$-term renormalizes these hoppings as $t_1+2W_d$ and $t_2-W_d$, respectively. Quasiparticle energies $\varepsilon(k)$ are affected accordingly. Of course, electrons are strongly correlated near half filling, and we need numerical calculations to determine the real quasiparticle energy. We have calculated the Green’s function for the $t$-$J$-$W_d$ model now including $t_1$ and $t_2$, and determined the hole occupation and the quasiparticle/quasihole energy at each $k$. Some results are shown in Fig. 5. As was shown before, the $\delta$-dependence of the single-electron level is basically consistent with the rigid band picture, when $W_d = 0$. On the other hand, for finite $W_d$, the single-electron level is strongly renormalized to gain a large energy from the $W_d$-term rather than single-electron hopping term. Its important consequence is the pinning of the $(\pi,0)$-level near $E_F$ during hole doping, and the spectral function is found to have a sharp low-energy peak in both the particle and hole parts. This double peak structure agrees with what is expected in superconducting states, and it is more prominent than that in Fig. 2.

The single-electron part of the $W_d$-term, to some extent, suppresses superconductivity near half filling, since it pushes down the $(\pi,0)$-level away from $E_F$. On more general grounds, the $d$-wave pairing interaction necessarily induces a downward shift of $(\pi,0)$-level. The experimentally observed $(\pi,0)$ level lower than $(\pi/2,\pi/2)$ in the highly underdoped samples may at least partially result from such mechanism. We have calculated pairing correlations for the $t$-$J$-$W_d$ model including $t_1$ and $t_2$ hoppings and examined the relation with the $(\pi,0)$-level. Figure 6 shows its coherent part $P_d(\omega=0)$ at $\delta = 0$ as a function of the level difference between $(\pi,0)$ and $(\pi/2,\pi/2)$. As a realistic choice, we here set $J = 0$ and $W_d = 0.05t$, equivalent to $J_{dr} = 0.4t$, corresponding to Fig. 1. The level difference $u < 0$ at half filling and the enhancement of the pairing correlations with further doping now becomes more compatible. In addition, as in the $t$-$J$ model, superconductivity is more enhanced when the $(\pi,0)$-level is tuned close to the Fermi energy, and this indeed offers potential optimization beyond present
Fig. 5. Single-electron spectral function in the 2D t-W_d model including \( t_1 \) and \( t_2 \). \( J/t = 0 \) and \( W_d/t = 0.05 \). Results for \( k = (\pi, 0) \) and \( (\pi/2, \pi/2) \) are shown here for typical values of \( t_1 \) and \( t_2 \).

Fig. 6. Enhancement of superconductivity by level tuning in the 2D t-W_d model. The coherent part of electron-pair annihilation at \( \delta = 0 \) is plotted as a function of the level difference \( u \) between \( (\pi, 0) \) and \( (\pi/2, \pi/2) \). Level difference for the free electron case \( \omega_0 \) is also shown for comparison.

The authors thank Fakher Assaad for valuable discussions. This work is supported by a Grant-in-Aid for "Research for the Future" Program from Japan Society for the Promotion of Science under the project JSPS-RFTF97P01103.

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