Nonmonotonic $d_{x^2-y^2}$-Wave Superconductivity in Electron-Doped Cuprates Viewing from the Strong-Coupling Side

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Applying a variational Monte Carlo method to a two-dimensional $t$-$J$ model, we study the nonmonotonic $d_{x^2-y^2}$-wave superconductivity, observed by Raman scattering and ARPES experiments in the electron-doped cuprates. As a gap function in the trial state, we extend the $d$-wave form (ext.$d$) so as to have its maxima located near the hot spots of the system. It is found that, in contrast to the hole-doped case, the ext.$d$ wave is always more stable than the simple $d$ wave in the electron-doped case, and the magnetic correlation of the wave vector $(\pi, \pi)$ as well as the pair correlation is enhanced. These results corroborate spin-correlation-mediated superconductivity in cuprates, recently argued from a FLEX calculation. In addition, we confirm that $s$- and $p$-wave symmetries are never stabilized even in the over-doped regime.

KEYWORDS: superconductivity, electron-doped High-$T_c$ cuprate, nonmonotonic $d_{x^2-y^2}$ wave, antiferromagnetic correlation, hot spot, $t$-$J$ model, variational Monte Carlo method

Introduction: Antiferromagnetic (AF) correlation is probably the primary origin to form Cooper pairs in the high-$T_c$ cuprates. A recent neutron scattering experiment$^3$ in the electron-doped ($n$-type) cuprates discovered peaks at magnetic Bragg spots in both normal and superconducting (SC) phases, which fact indicates that the spin correlation of the AF wave vector $Q = (\pi, \pi)$ plays an important role in $n$-type cuprates.

As in the hole-doped ($p$-type) cuprates, the pairing symmetry of the $n$-type ones was ascertained to be a $d_{x^2-y^2}$-wave type, as far as the doping rate $\delta$ is smaller than 0.15, by a scanning SQUID microscope$^4$ and angle-resolved photoelectron spectroscopy (ARPES)$^5,6$ However, recent experiments by Raman scattering$^7$ and ARPES$^8$ have concluded that the pairing symmetry in the $n$-type cuprates is not the typical $d_{x^2-y^2}$ wave characterized by $\Delta_k \propto \cos k_x - \cos k_y$, but exhibits a non-monotonic behavior. Namely, the maximum of $\Delta_k$ is located midway between the Brillouin-zone boundary $(\pi, 0)$ and the zone diagonal $(\pi/2, \pi/2)$. Furthermore, for $T > T_c$, pseudogap-like behavior$^9$ or an AF gap$^{10}$ arises at this midway locus, in contrast to the $p$-type cuprates, in which the gap maximum and pseudogap behavior take place around $(\pi, 0)$.

These results can be explained by the fact that the loci of the gap maximum in the $p$ and $n$ types roughly coincide with their respective hot spots—the intersection of the Fermi surface and the magnetic Brillouin zone boundary [See Fig. 1(a)]. This observation affords confirmatory evidence that the superconductivity in cuprates is induced by the spin correlation (or fluctuation) of the wave vector $Q$, which connects the hot spots.

Theoretically, Yoshimura and Hirashima$^{11}$ recently treated this issue, applying a fluctuation exchange approximation (FLEX) to the Hubbard model. They obtained a non-monotonic behavior of $\Delta_k$ and consistent results of Raman spectral functions and spin susceptibility etc. with the experiments. Since the electron correlation is not weak even in the $n$-type cuprates, the results of FLEX, which is basically a weak-coupling theory, should be checked by complementary studies from the strong-coupling and low-carrier-density sides. In this paper, we study this issue, applying a variational Monte Carlo (VMC) method$^{12}$ to a $t$-$J$-type model. So far, taking this approach, various aspects of the cuprates have been elucidated on a strong-coupling footing.$^{13}$

A secondary interest of this paper is possible variation of the pairing symmetry from $d$ to $s$ wave in the over-doped regime of $n$-type cuprates, observed by tunneling spectroscopy$^{14}$ and measurement of magnetic penetration depth.$^{15}$ Although a BCS-level calculation$^{16}$ supports these experiments, one has to confirm this issue using a less biased approach.

Formulation: We consider a two-dimensional $t$-$J$ model, $\mathcal{H} = \mathcal{H}_t + \mathcal{H}_J$ with $\mathcal{H}_t = -\sum_{(i,j)\sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma}$ + H.c.$\mathcal{P}_G$ and $\mathcal{H}_J = J \sum_{(i,j)} (\mathbf{S}_i \cdot \mathbf{S}_j - n_i n_j / 4)$, where $\mathcal{P}_G = \prod_j (1 - n_j n_j)$, the value of $J/t$ is fixed at 0.3, and $t_{ij} = t, t', t''$ or 0, according as the site $i$ is a first-, second-, third-nearest neighbor or farther site of the site $j$, respectively. An electron-doped (more-than-half-filled) system can be treated with a $t$-$J$ model as a less-than-half-filled case, by applying a particle-hole transformation $c_{j\sigma} \rightarrow \exp(i \mathbf{Q} \cdot \mathbf{r}_j) h_{j\sigma}$ with $t' t'' / t > 0$ for $n$- ($p$-) type cuprates. Actually, we adopt typical values$^{17}$ of $t'/t$ and $t'' / t$ given in the caption of Fig. 1.

To this model, we apply a VMC method, which accurately treats the local correlation $\mathcal{P}_G$. As a variational function for a SC state, a simple Gutzwiller-type wave function is used,

$$|\Psi_{SC}\rangle = \mathcal{P}_G |\Phi_{BCS}\rangle = \mathcal{P}_G \left( \sum_k \varphi_k c_{k\uparrow}^\dagger c_{-k\downarrow}^\dagger \right)^{\frac{\mathcal{P}_G}{2}} |0\rangle,$$

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with

$$\varphi_k = \frac{u_k}{v_k} = \frac{\Delta_k}{\varepsilon_k - \mu + \sqrt{(\varepsilon_k - \mu)^2 + \Delta_k^2}}. \quad (2)$$

Here, \(\Phi_{\text{BCS}}\) is the BCS wave function of a fixed particle number \(N_e\), \(\varepsilon_k = -2t(\cos k_x + \cos k_y) - 4t' \cos k_x \cos k_y - 2t''(\cos 2k_x + \cos 2k_y)\), and the parameter \(\mu\) is substituted by the chemical potential of the non-interacting case. It is already known that this type of wave function works well for \(t-J\)-type models.\(^{18,19}\) Anisotropy of the pairing potential is introduced into \(\Delta_k\), as \(\Delta_k^d = \Delta_0(\cos k_x - \cos k_y)\) for the simple \(d\) wave. We extend it so that it may have large amplitude near the hot spots,

$$\Delta_k^{\text{ext.d}} = \Delta_k^d + a \Delta_0 \left[ k_x^2 e^{-c(k_x-b)^2 - dk_x^2} + k_y^2 e^{-c(k_y-b)^2 - dk_y^2} - k_x^2 e^{-c(k_x+a)^2 - dk_x^2} - k_y^2 e^{-c(k_y+a)^2 - dk_y^2} \right], \quad (3)$$

and call it the extended-\(d\) (ext.\(d\)) wave. In \(\Delta_k^{\text{ext.d}}\), we add to \(\Delta_k^d\) eight Gaussian peaks, whose height, position and width in two directions are adjusted by parameter \(a,b,c\) and \(d\), respectively. Although \(\Delta_k\) should be determined variationally, for simplicity we fix the parameters for the Gaussian, except for trial calculations,\(^{20}\) at rough optimal values of the case \(\delta = 0.082\) (\(L = 14\)), namely, \(a = 3, b = 2, c = 1\) and \(d = 1\); \(\Delta_k\) thereof is shown in Fig.1(b). Note that the loci of the maximal \(\Delta_k\) are situated very closely to the hot spots, shown in Fig.1(c).

Thus, \(\Delta_0\) (amplitude of \(\Delta_k\)) becomes a sole parameter to be optimized. Unlike the BCS theory, \(\Delta_k\) here is not entirely equivalent to the SC gap, especially in \(\Delta_0\), but the symmetry of \(\Delta_k\) faithfully reflects the SC gap.

We have followed a conventional VMC scheme,\(^{12}\) and collected \(10^2-10^5\) samples, which suppress the statistical errors in energy at \(\sim 10^{-4}\). The system used has \(L \times L\) sites (\(L = 12,14\) and 16) with the periodic-antiperiodic boundary conditions. The particle density \(\delta\) is chosen so as to satisfy the closed-shell condition.

Results: Before going to the \(d\)-type pairings, we check the stability of the singlet \(s\)- (\(\Delta_k = \Delta_0\)), ext. \(s\)- (\(= \Delta_0(\cos k_x + \cos k_y)\)) and triplet \(p\)-wave (\(= \Delta_0 \sin k_x\)) symmetries. In Fig.2, the variational energies \(E_{\text{tot}}\) of the symmetries we treat are compared in the overdoped regime. Here, \(E_{\text{tot}}\)'s of the \(s\), ext.\(s\) and \(p\) waves monotonically increase, as \(\Delta_0\) increases, and are destabilized with respect to the normal state. We have also confirmed that the same behavior persists to a large doping rate, \(\delta = 0.245\), where SC states are no longer stabilized. Since our treatment is little biased in comparing pairing symmetries, we are confident that the \(p\) and \(s\)-type waves are not realized even in the overdoped regime; thus a pairing-symmetry transition is unlikely to arise.\(^{21}\)

Now, we turn to the \(d\)-type symmetries. By contrast, the variational energies of the \(d\)- and ext.\(d\)-wave SC states plotted in Fig.2 have minima at finite values of \(\Delta_0\), as indicated by arrows. The decrease in \(E_{\text{tot}}\) of the \(d_{x^2-y^2}\) wave has been well-known for the plain \(t-J\) model (\(t' = t'' = 0\) since the early stage.\(^{25}\) It should be noted here that the ext.\(d\) wave has an appreciably lower energy than the simple \(d\) wave even for such a large value of \(\delta\).

We have optimized \(E_{\text{tot}}\) similarly for various values of \(\delta\), and depict in Fig.3(a) the difference of \(E_{\text{tot}}\) between the \(d\) and ext.\(d\) waves, \(\Delta E = E_{\text{tot}}^d - E_{\text{tot}}^{\text{ext.d}}\).\(^{26}\) In electron-doped cases, the ext.\(d\) wave is always more stable than the \(d\) wave. The large value of \(\Delta E\) near half filling probably stems from the fact that the Gaussian peaks in \(\Delta_k^{\text{ext.d}}\) are close to the hot spots, as well as that the energy scale in the condensation energy \(E_{\text{cond}}^d = E_{\text{tot}}^{\text{cond}} - E_{\text{tot}}^d\) becomes large for \(\delta \to 0\). \(\Delta E\) vanishes at \(\delta = 0.222\), where the hot spots still survive, but \(E_{\text{cond}}^d\) vanishes for both waves. Conversely, in the hole-doped cases [open symbols in Fig.3(a)], the simple \(d\) wave is more stable than the ext.\(d\) wave except in the vicinity of half filling. In the optimal- and overdoped regime for \(\delta < 0\), the hot spots sit near \((\pi,0)\) and equivalent points, whereas near half filling the hot spots are still away from \((\pi,0)\) point and rather closer to the Gaussian positions we set (Fig.1(b)). Thus, the results in \(E_{\text{tot}}\) definitely indicate that the SC state becomes stable when the maximum of the gap \(\Delta_k\)
is located near the hot spots of the system.

To reinforce the above argument, we consider the $d$-wave SC correlation function of nearest-neighbor pairs,

$$P_d(r) = \frac{1}{N_s} \sum_j \sum_{\tau,\tau'=-\infty} (-1)^{1-\delta(\tau,\tau')} \langle \Delta_\tau(j) \Delta_{\tau'}(j+r) \rangle,$$

with $\Delta_\tau(j) = (c_{j\uparrow}^\dagger c_{j+\tau\downarrow} + c_{j+\tau\uparrow}^\dagger c_{j\downarrow})$. Since $P_d(r)$ rapidly decays with $|r|$ and is almost constant for $|r| \gtrsim 3$, the average for $|r| \gtrsim 3$, $P_d^{ave}$, gives an adequate estimate of the long-distance value. In Fig.3(b), $P_d^{ave}$ is plotted versus carrier density. The remarkable asymmetry between the $p$ and $n$ types can be attributed to the difference in DOS at the Fermi surface, particularly at the hot spots. Note that the ext. $d$ wave always exceeds the $d$ wave for the $n$ type, whereas the relation is inverse in the overdoped regime for the $p$ type. This tendency of $P_d(r)$ corresponds well with that of $\Delta E$ [Fig.3(a)]. Incidentally, in $n$-type systems, the SC correlation of long-distance pairs is probably enhanced, as will be discussed later.

Next, to identify the origin of the energy gain in $E_{tot}$, we compare the energy components, namely hopping energy $E_t = \langle H_t \rangle$ and exchange energy $E_J = \langle H_J \rangle$, among the $d$-wave, ext. $d$-wave and normal states. In Table I, we list the raw values of the $d$-wave, $E_t^d$ and $E_J^d$, and the differences between them and those of the other two states, namely, $\Delta E_t^N (= E_t^d - E_t^{normal})$, $\Delta E_J^N (= E_J^d - E_J^{ext.d})$, etc. for four kinds of doping. As compared with the normal state, the $d$- (also ext. $d$-) wave SC state is stabilized by the noticeable decrease in $E_J$; conversely, $E_t$ is more or less increases. Such magnetic origin of superconductivity is characteristic of $t$-$J$-type models, and closely related to the kinetic-energy-driven SC mechanism in the strong-correlation regime of the Hubbard model. In comparison of the $d$ and ext. $d$ waves, $E_{ext.d}^d$ is lower whenever the ext. $d$ wave has a lower total energy ($\delta \simeq -0.06$), whereas the ext. $d$ wave always possesses somewhat higher $E_t$. Thus, the gap structure of the ext. $d$ wave has further advantages to gain magnetic energy for $n$-type cases over the simple $d$ wave.

Now, we consider the momentum distribution function, $n(k) = 1/2 \sum_\epsilon |c_{k\epsilon}^\dagger c_{k\epsilon}|$, to actually observe the gap behavior in the momentum space—a milder slope at quasi-$k_F$. In Figs. 4(a) and (b), we depict $n(k)$ for an electron-doped case along two paths, namely OP in (a), which goes away from the hot spots, and QR in (b), which penetrates the hot-spot area. In (a), $n(k)$’s for the $d$ and ext. $d$ waves exhibit similar behavior, and the $d$ wave seems slightly mild. On the other hand, in (b) the ext. $d$ wave makes an obviously milder curve around the hot spot. Thus, the gap behavior around the hot spot is enhanced in the ext. $d$-wave state. Incidentally, in the node-of-gap direction, the difference between the $d$ and ext. $d$ waves is very small, and a clear Fermi surface [discontinuity in $n(k)$] can be seen (not shown).

Finally, we consider the spin correlation function. In Fig.5(a), the spin structure factor, $S(q) = 1/N_s \sum_{ij} c_{i\sigma}^\dagger c_{j\sigma} S(i,j)$ with $S(i,j) = \langle S_i^z S_j^z \rangle$, of the $d$ and ext. $d$ waves is plotted for several densities of electron doping. Both waves have the maximal amplitude at $(\pi, \pi)$ for all the electron densities, which is consistent
with the neutron experiment. As shown in the inset of Fig.5(a), the difference between the $d$ and $d$ waves is almost restricted to the vicinity of $(\pi, \pi)$, where $S(q)$ of the $d$ wave is sizably enhanced. This enhanced AF correlation naturally leads to the energy gain in $E_J$, mentioned above. Shown in Fig.5(b) is the ratio of the real-space spin correlation function for the $d$ wave to that for the simple $d$ wave. Although the $d$ and $d$ waves exhibit almost the same values for the nearest-neighbor sites ($|i-j|=1$), $S(i,j)$ of the $d$ wave for farther distances considerably increases, especially in the $x$ (or $y$) direction. This is because the harmonics of the $d$ wave, $\cos nk_x - \cos nk_y (n \geq 2)$, give substantial contribution to $\Delta^{ext,d}_{k}$. Unlike the pure $d$ wave ($n=1$), they elongate the coherence length (and magnetic correlation length); thereby the correlation strength in the electron-doped systems becomes effectively weaker. Such tendency has been actually observed by various experiments.

**Summary:** Using a variational Monte Carlo method for a $t$-$J$ model, we have studied a nonmonotonic (ext.) $d$-wave superconducting state, in which the amplitude of the gap parameter $\Delta$ is intentionally enhanced around the hot spots, so as to agree with recent experiments of Raman scattering and ARPES. This $d$-wave state has an appreciably lower energy than the simple $d$-wave state for all the densities of electron doping (also very low hole doping). This stabilization of the $d$ wave is caused by the gain in magnetic exchange energy, accompanied by a marked increase in the spin correlation of the wave vector $Q = (\pi, \pi)$. In addition, we have shown that the $s$-type and $p$ waves are unlikely to take place in the high-$T_c$ regime of $\delta$. Our results using a strong-coupling approach basically agree with the recent FLEX study; thereby, it is ensured that the AF spin correlation plays a crucial role for the high-$T_c$ superconductivity.

We believe that the essence of the nonmonotonic $d$-wave gap is grasped in this work, although we have simplified $\Delta^{ext,d}$ by both a naive assumption of a Gaussian form and fixing the parameters controlling the Gaussian. We should address quantitative refinement as well as related issues. For instance, (1) simultaneous optimization of all the variational parameters, including the renormalization of the quasi-Fermi surface, enables us to follow the continuous evolution of $\Delta_k$ versus $\delta$. (2) The relation to the AF ordered state is very important.

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