Theory of reduced singlet pairing without the underlying state of charge stripes in the high-temperature superconductor YBa$_2$Cu$_3$O$_{6.45}$

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Recently, a strongly enhanced xy anisotropy of magnetic excitations was observed in YBa$_2$Cu$_3$O$_y$ (YBCO$_y$), with $y=6.45$ and $T_c=35$ K [V. Hinkov *et al.*, Science **319**, 597 (2008)]. Unlike the observation in YBCO$_{6.6}$ and YBCO$_{6.85}$, the anisotropy grows to be pronounced at lower temperature and at lower energy and is not suppressed by the onset of superconductivity. We propose that the effect of singlet pairing is substantially reduced in YBCO$_{6.45}$. This reduction concomitantly enhances an order competing with singlet pairing, a strong tendency of the so-called d-wave Pomeranchuk instability, leading to the magnetic excitations observed experimentally.

The spin-charge stripe order was widely discussed in high-T_c cuprates.[1] In this scenario, the two-dimensional CuO$_2$ plane is assumed to have the instability of one-dimensional charge order, the so-called stripe pattern, in the antiferromagnetic (AF) background with a π phase shift across the charge stripe. However, the charge order signal was observed only for limited materials with specific hole doping rates and had a broad spectrum with very weak intensity,[2–5] implying that the charge order is usually not well developed. Kivelson *et al.*[6] introduced a new concept of electronic nematic order, which was envisaged as the melting of charge stripe order,[7] where the translational symmetry is recovered but the orientational symmetry is still broken.

On the other hand, an alternative root to yield electronic nematic order was found in minimal models of high-T_c cuprates such as the t-J (Refs. 8–10) and Hubbard[11–15] models. These models show a tendency toward d-wave-type Fermi-surface deformations (dFSDs), the so-called d-wave Pomeranchuk instability. The Fermi surface (FS) expands along the k_x direction and shrinks along the k_y direction or vice versa. The dFSD state has the same symmetry as the electronic nematic state. But the physical origin is different from the stripe physics[6] since the dFSD is generated by forward-scattering processes of quasiparticles, not by fluctuating charge stripes.

The dFSD competes with superconductivity.[16] In the slave-boson mean-field[8] and variational Monte Carlo[10] analyses of the t-J model, superconductivity becomes dominant. Yet the system still has sizable dFSD correlations,[17] producing a giant response to a small external anisotropy, e.g., due to a lattice structure and anisotropic strain.[8,10] This theoretical insight yields a promising scenario[18] to understand the pronounced anisotropy of magnetic excitations observed in untwinned YBa$_2$Cu$_3$O$_y$ (YBCO$_y$), with $y=6.5$,[19] 6.6,[20,21] and 6.85,[20] where the lattice yields a small xy anisotropy.

Quite recently, a very strong anisotropy was observed in magnetic excitations in untwinned YBCO$_{6.45}$.[22] The observed spectra were qualitatively different from typical observations in Y-based cuprates. (i) The anisotropy starts to increase below $\sim$150 K and saturates below $\sim$50 K, in contrast to the case of YBCO$_{6.6}$ (Refs. 20 and 21) where the anisotropy is

most pronounced at relatively high temperature and is reduced at low temperature. (ii) The magnetic excitation spectra are hardly affected by the onset of superconductivity and in fact a gap feature is not found in the magnetic excitations. This is sharply different from the well-known observation in Y-based cuprates, where a broad spectrum around **q** $=(\pi,\pi)$ becomes very sharp and is strongly enhanced below the onset temperature of superconductivity, leading to a resonance at relatively high energy, followed by suppression of low-energy spectral weight.[23,24] These peculiar magnetic excitations in YBCO$_{6.45}$ are not captured even qualitatively by simple application of the theory in Ref. 18, implying failure of the standard resonating-valence-bond (RVB) mean-field theory in a strongly underdoped region.

Why do the qualitative features of magnetic excitations change so drastic in YBCO$_{6.45}$? Since YBCO$_{6.45}$ lies closer to the AF instability, strong AF fluctuations might play an important role. However, even for the lower hole-doped system YBCO$_{6.353}$, and thus much closer to the AF instability, the experimental data[25] showed that magnetic correlation length is still short ranged. In this Brief Report, we show an integral role of dFSD correlations, which are concomitantly enhanced by the suppression of singlet pairing, leading to results very similar to the experimental observation.[22]

We analyze the bilayer t-J model on a square lattice, a minimal model for Y-based cuprates,[26]

$$H = -\sum_{\mathbf{r},\mathbf{r}',\sigma} t_\tau \widetilde{c}_{\mathbf{r}\sigma}^\dagger \widetilde{c}_{\mathbf{r}'\sigma} + \sum_{\langle \mathbf{r},\mathbf{r}' \rangle} J_\tau \mathbf{S}_\mathbf{r} \cdot \mathbf{S}_{\mathbf{r}'}, \tag{1}$$

defined in the Fock space with no doubly occupied sites. The operator $\widetilde{c}_{\mathbf{r}\sigma}^\dagger$ ($\widetilde{c}_{\mathbf{r}\sigma}$) creates (annihilates) an electron with spin σ on site $\mathbf{r}$, and $\mathbf{S}_\mathbf{r}$ is the spin operator. J_τ (>0) is a super-exchange coupling between the nearest-neighbor sites along each direction, $\tau=x,y,z$. We take into account hopping amplitudes t_τ between $\mathbf{r}$ and $\mathbf{r}'$ up to third-nearest neighbors, and the direction of $\mathbf{r}'-\mathbf{r}$ is represented by τ. Hamiltonian (1) is analyzed in the standard slave-boson method in the t-J model by introducing the so-called RVB mean fields. However, in contrast to the standard RVB theory, we assume the mean field of singlet pairing Δ to be zero in order to mimic possibly substantial suppression of singlet pairing in
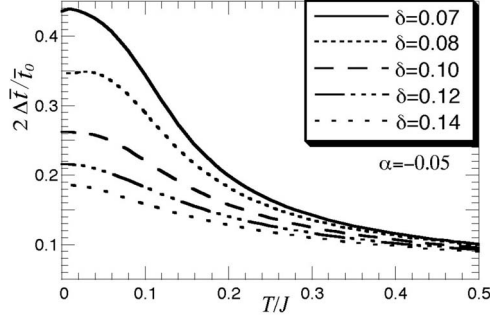
FIG. 1. T dependence of the anisotropy of renormalized band, $2\Delta\bar{t}/\Delta\bar{t}_0$, for several choices of δ for $\alpha=-0.05$.

YBCO$_{6.45}$, as we will discuss later in detail. The formalism and the band parameters are otherwise the same as those in Ref. 18, where we successfully discussed the pronounced anisotropy of magnetic excitations in YBCO$_{6.85}$ (Ref. 20) and YBCO$_{6.6}$.[20,21] We introduce 5% xy anisotropy into the hopping integrals t_τ and 10% into the superexchange coupling J_τ, twice as large, as imposed by the superexchange mechanism. The anisotropy then can be parametrized by a single parameter $\alpha=-0.05$. The minus sign indicates that the band parameters are more enhanced along the y direction, parallel to the direction of the CuO chains, which indeed reproduces[18,27,28] qualitatively the same anisotropic distribution of magnetic excitations as observed previously.[20,21] Interestingly the negative sign of α is also implied in a spin spiral state to understand the anisotropy of magnetic excitations.[29]

The most important quantity is the renormalized in-plane nearest-neighbor hopping $\bar{t}_\tau$, which is given by $\bar{t}_\tau=t_\tau\delta +\frac{3}{8}J_\tau\chi_\tau$, with $\tau=x,y$, δ is the hole density, and χ_τ is the RVB bond order along the τ direction. The order parameter of the dFSD is then defined as $\chi_d=(\chi_x-\chi_y)/2$. Figure 1 shows the temperature (T) dependence of the anisotropy of renormalized band $2\Delta\bar{t}/\Delta\bar{t}_0$, where $\Delta\bar{t}=|\bar{t}_x-\bar{t}_y|/2=|(t_x-t_y)\delta/2+\frac{3}{8}(J_x-J_y)\chi_d|$ and $\bar{t}_0=(\bar{t}_x+\bar{t}_y)/2$. The anisotropy is strongly enhanced at lower T, especially for lower δ. This enhancement comes from the development of χ_d, namely, the underlying dFSD instability in the t-J model.[8–10] The presence of the anisotropy in t_τ and J_τ, however, smears the singularity associated with the dFSD instability, which would appear below $T\approx 0.03J(0.05J)$ for $\delta=0.08(0.07)$ and disappear for $\delta \gtrsim 0.09$ if the input anisotropy is zero.

The irreducible dynamical magnetic susceptibility $\chi_0(\mathbf{q},\omega)$ reads

$$\chi_0(\mathbf{q},\omega) = \frac{1}{4N}\sum_{\mathbf{k}} \frac{\tanh\dfrac{\xi_{\mathbf{k}}}{2T} - \tanh\dfrac{\xi_{\mathbf{k+q}}}{2T}}{\xi_{\mathbf{k}} - \xi_{\mathbf{k+q}} + \omega + i\Gamma}, \quad (2)$$

where N is the total number of (bilayer) lattice sites and Γ is a positive infinitesimal. We calculate magnetic excitations in the renormalized random-phase approximation (RPA),[30,31]

$$\chi(\mathbf{q},\omega) = \frac{\chi_0(\mathbf{q},\omega)}{1 + J(\mathbf{q})\chi_0(\mathbf{q},\omega)}. \quad (3)$$
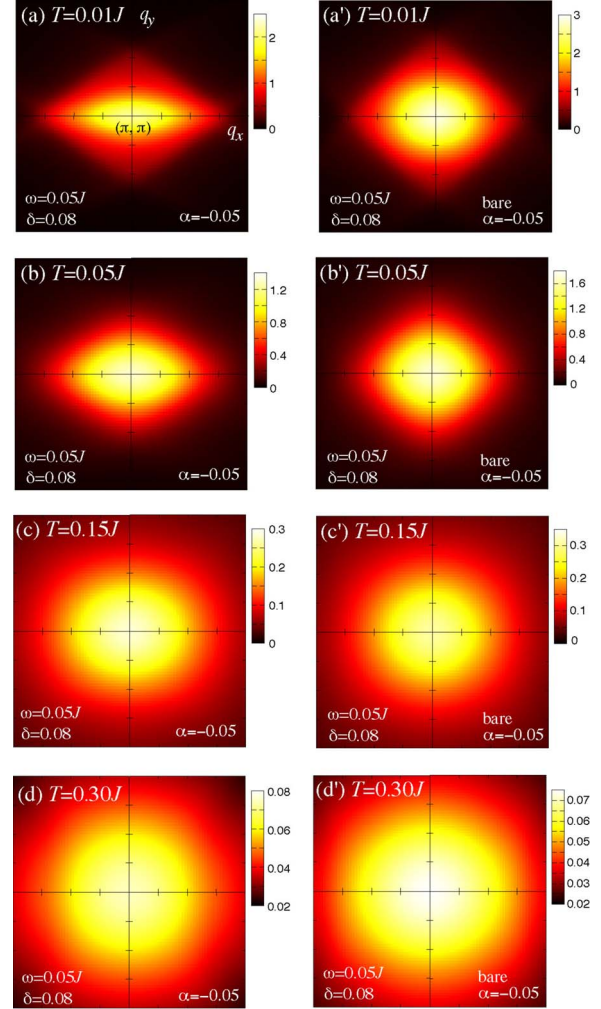
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FIG. 2. (Color online) Left-hand panels: **q** maps of Im $\chi(\mathbf{q},\omega)$ for a sequence of T in $0.6\pi \leq q_x,q_y \leq 1.4\pi$ for $\delta=0.08$, $\omega=0.05J$, and $\alpha=-0.05$. Right-hand panels: corresponding results for the bare anisotropy, namely, without dFSD correlations.

$$J(\mathbf{q}) = 2r(J_x \cos q_x + J_y \cos q_y) + J_z \cos q_z \quad (4)$$

with a renormalization factor r. While $r=1$ in the plain RPA, the value of r is reduced in the renormalized RPA such that the AF instability appears in a doping region comparable with actual materials. We set $r=0.4$, which confines the AF instability to $\delta \lesssim 0.068$.

We calculate $\chi(\mathbf{q},\omega)$ numerically by choosing $\Gamma=0.05J$. This choice of Γ is mainly due to numerical convenience but simulates damping of electrons by static defects in real materials and broadening due to limited energy resolution in neutron-scattering experiments. Since neutron-scattering measurements for untwinned YBCO are performed for the odd channel, we focus on this channel ($q_z=\pi$). Considering that the hole density in YBCO$_{6.45}$ is estimated as 8.5%,[22] we fix $\delta=0.08$.

The left-hand panels of Fig. 2 show two-dimensional **q** maps of the imaginary part of $\chi(\mathbf{q},\omega)$ at $\omega=0.05J$ for a sequence of temperatures. At $T=0.01J$, significant spectral weight is centered around $\mathbf{q}=(\pi,\pi)$ and is strongly elon-
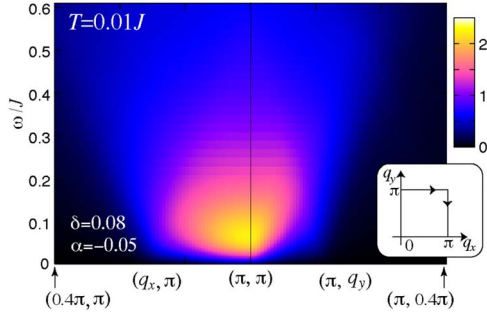
FIG. 3. (Color online) $(\mathbf{q}, \omega)$ map of Im $\chi(\mathbf{q}, \omega)$ at $T=0.01J$ for $\delta=0.08$ and $\alpha=-0.05$; the $\mathbf{q}$ directions are shown in the inset.

gated along the q_x direction. This pronounced anisotropy originates from the strong enhancement of the original band anisotropy due to dFSD correlations (Fig. 1). The anisotropy of magnetic excitations is reduced with increasing T. In particular, the distribution of the spectral weight becomes nearly symmetric around (π, π) at $T=0.30J$. To clarify the dFSD effect, we show the corresponding result for the *bare* anisotropy effect by switching off dFSD correlations in the right-hand panels of Fig. 2, where we impose the same anisotropy $\alpha=-0.05$ but assume $\chi_d \equiv 0$. Although the value of $\Delta \bar{t}$ does not depend on T, we see that the anisotropy of Im $\chi(\mathbf{q}, \omega)$ depends weakly on T and becomes visible with decreasing T. But the obtained anisotropy [Figs. 2(a′) and 2(b′)] is much weaker than the results due to dFSD correlations [Figs. 2(a) and 2(b)]. Hence the underlying dFSD correlations are particularly important to produce a strongly enhanced anisotropy at lower T.

To see the ω dependence of the anisotropy, the $(\mathbf{q}, \omega)$ map of Im $\chi(\mathbf{q}, \omega)$ is shown in Fig. 3 at low T. While the anisotropic distribution is still discernible for high energy where the spectral intensity is substantially reduced, a pronounced anisotropy appears especially in a low-energy region.

The evolution of the spectral intensity of Im $\chi(\mathbf{q}, \omega)$ is shown in Fig. 4 as a function of ω for several choices of T at $\mathbf{q}=\mathbf{Q}=(\pi, \pi)$. At high T, Im $\chi(\mathbf{Q}, \omega)$ shows a broad maximum at a moderate energy. With decreasing T, low-energy spectral weight is substantially enhanced yielding a peak structure at low energy.

When dFSD correlations strongly enhance a small xy anisotropy of the original band dispersion, a strong anisotropy of Im $\chi(\mathbf{q}, \omega)$ is induced with a characteristic T dependence.
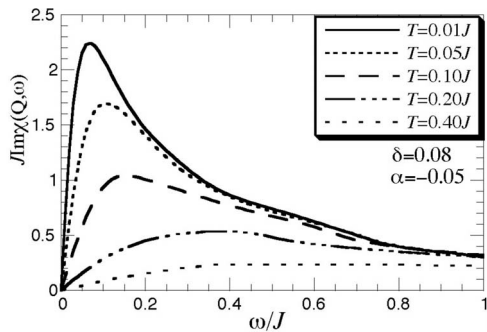

FIG. 4. ω dependence of Im $\chi(\mathbf{Q}, \omega)$ for several choices of T for $\delta=0.08$ and $\alpha=-0.05$.

In YBCO$_{6.45}$, the experimental data[22] suggest that fermions responsible for magnetic excitations feel a rather small mean field of singlet pairing in the sense that neither a gap feature nor a clear impact of the onset of superconductivity is observed in neutron scattering. This special experimental situation is mimicked by setting $\Delta \equiv 0$ in the slave-boson scheme of the t-J model. We have found that this phenomenological treatment well captures the most salient features observed in YBCO$_{6.45}$ (Ref. 22): (i) the strongly enhanced anisotropy of Im $\chi(\mathbf{q}, \omega)$ at lower T for low ω (Figs. 2 and 3) and (ii) the enhanced spectral weight of Im $\chi(\mathbf{Q}, \omega)$ at low ω for low T (Fig. 4).

On the other hand, in the standard slave-boson mean-field theory of the t-J model,[18] the onset temperature of singlet pairing is higher than that of the dFSD, and the singlet formation suppresses the susceptibility of the dFSD, which then does not diverge.[8] The susceptibility, however, remains appreciable[17] so that a small external anisotropy introduced in the t-J model is strongly enhanced even in the singlet pairing state.[8] This effect is more pronounced for a lower doping rate. Such an enhanced anisotropy nonetheless turns out not to be sufficient to capture the anisotropy of Im $\chi(\mathbf{q}, \omega)$ observed in YBCO$_{6.45}$ [22] even at qualitative level for a realistic choice of parameters. In particular, the anisotropy is enhanced at relatively high T (Fig. 20 of Ref. 18). Moreover the experimental observation[22] that the onset of superconductivity hardly affects magnetic excitations is difficult to be captured in existing microscopic theories for cuprates.

Inclusion of singlet pairing is not so straightforward, and something beyond the existing theories happens in YBCO$_{6.45}$. Here focusing on the context of the present Brief Report we discuss several possibilities that dFSD correlations still remain strong enough to be compatible with the present result even in a full calculation. First, since the dFSD instability concerns discrete symmetry breaking, the spontaneous symmetry breaking is allowed at finite temperature even in the exact analysis of the two-dimensional model in contrast to superconductivity and antiferromagnetism. Hence suppression of the dFSD due to fluctuations is less crucial than the latter two, in favor of strong correlations of the dFSD. In the presence of a small external xy anisotropy, then the anisotropy can be strongly enhanced as we have seen in Fig. 1. The feedback of the enhanced anisotropy appears as further reduction in the magnitude of singlet pairing compared with the mean-field value in the slave-boson theory of the t-J model. Second, YBCO$_{6.45}$ has an orthorhombic crystal structure but is close to the continuous transition to a tetragonal structure. The dFSD order parameter couples to a phonon related to the orthorhombic-tetragonal structural transition, which contributes to an enhancement of dFSD correlations. In this case, the phonon spectrum may also show strong anisotropy in the orthorhombic phase. Last, there might be a phase segregation into hole-poor AF domains with nanoscale correlation lengths and hole-rich superconducting islands in YBCO$_{6.45}$, and the neutron-scattering signals originate mainly from the AF domains. Such finite-sized domains are often discussed in a strongly underdoped superconducting region in μSR measurements.[32,33]

While the present result well captures the neutron-scattering data in $YBCO_{6.45}$,[22] a detailed comparison reveals disagreement about several aspects, which may be mainly due to our simple calculation ignoring singlet pairing. (i) A weakly incommensurate structure was observed along the q_x direction, whereas we have obtained a single peak at (π, π) as read off from Fig. 2. In a fermiology scenario, the incommensurate structure in Y-based cuprates is explained by the suppression of the commensurate peak due to the development of d-wave singlet pairing.[18,34] Hence the observed weak incommensurate peak can be due to the development of relatively weak singlet pairing. (ii) The observed data at $\omega = 50$ meV was interpreted to be isotropic.[22] In the present calculation, the anisotropy becomes less pronounced for high ω but is still discernible (Fig. 3). On the other hand, if we include singlet pairing in the slave-boson mean-field theory, it is known[18] that the anisotropy of Im $\chi(\mathbf{q}, \omega)$ at high ω depends strongly on energy. More detailed experiments as well as more complete calculations are necessary to discuss the anisotropy at high ω.

The present phenomenological calculation suggests that d-wave singlet pairing is substantially reduced in $YBCO_{6.45}$, which then concomitantly enhances strong dFSD correlations, leading to a magnetic excitation spectrum very similar to the experimental data.[22] Since the magnitude of d-wave singlet pairing is often discussed to become larger with de-creasing doping rate, the present Brief Report implies an interesting direction to understand the evolution of d-wave singlet pairing as well as its connection with dFSD correlations in underdoped cuprates. Recent angle-resolved photoemission spectroscopy data[35] is in fact suggestive of relatively small d-wave singlet pairing. It demonstrated two distinct gaps, the amplitude of one of which increases for lower doping rates, while the other, which shows d-wave symmetry, decreases. In addition, strong dFSD fluctuations substantially reduce the lifetime of quasiparticles in the antinodal region of the FS while not in the nodal direction as shown in Ref. 36. Hence strong dFSD fluctuations may play an integral role to understand the pseudogap in the strongly underdoped regime of YBCO. Strong dFSD fluctuations are also expected in a scenario of a quantum phase transition into the dFSD state in a underdoped region.[37] Such a scenario is, however, based on the assumption of the existence of robust d-wave pairing, which is different from the present theory where we have invoked the substantial suppression of d-wave pairing due to the competition[16,17] with the dFSD in a strongly underdoped region.

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