Investigating the Physical Origin of Unconventional Low-Energy Excitations and Pseudogap Phenomena in Cuprate Superconductors

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October 28, 2021

We investigate the physical origin of unconventional low-energy excitations in cuprate superconductors by considering the effect of coexisting competing orders (CO) and superconductivity (SC) and of quantum fluctuations and other bosonic modes on the low-energy charge excitation spectra. By incorporating both SC and CO in the bare Green’s function and quantum phase fluctuations in the self-energy, we can consistently account for various empirical findings in both the hole- and electron-type cuprates, including the excess subgap quasiparticle density of states, “dichotomy” in the fluctuation-renormalized quasiparticle spectral density in momentum space, and the occurrence and magnitude of a low-energy pseudogap being dependent on the relative gap strength of CO and SC. Comparing these calculated results with experiments of ours and others, we suggest that there are two energy scales associated with the pseudogap phenomena, with the high-energy pseudogap probably of magnetic origin and the low-energy pseudogap associated with competing orders.

PACS. 74.72.-h - Cuprate superconductors.
PACS. 74.50.+r - Pseudogap.
PACS. 74.25.Dw - Competing orders.

I. Introduction

High-temperature superconducting cuprates are doped Mott insulators with strongly correlated electronic ground states manifested in forms of different competing orders (CO) besides superconductivity (SC) [1, 2, 3, 4, 5]. Phenomenologically, the presence of coexisting SC and CO in the ground state has a number of important implications: 1) the occurrence of quantum criticality becomes a natural consequence of competing phases in the ground state [6]; 2) significant quantum fluctuations are expected as the result of proximity to quantum criticality [7, 8, 9]; 3) the low-energy excitations of coexisting SC and CO differ from conventional Bogoliubov quasiparticles of pure SC because of redistribution of the spectral weight between SC and CO [10, 11]; 4) the presence of competing orders and strong quantum fluctuations gives rise to weakened superconducting stiffness and extreme type-II nature in the cuprates [7, 8]; 5) the relevant competing orders can vary in different cuprates, giving rise to non-universal physical properties [6, 12, 13]; 6) the existence of competing orders and accompanied unconventional low-energy excitations are likely relevant to the occurrence of pseudogap phenomena [10]. We further note that empirically the pseudogap phenomena generally refer to two different energy scales;
the lower energy scale is correlated with the Nernst effect \[5, 14, 15\] and the suppressed quasiparticle density of states observed above \(T_c\), and is only found in hole-type cuprates; whereas the higher energy scale is present in both electron- and hole-type cuprates according to optical \[16, 17\] and neutron scattering \[18, 19\] experiments. As discussed in our recent work \[10\] and further elaborated below in this paper, the lower-energy pseudogap may be associated with the competing order, whereas the higher-energy pseudogap seems to be of magnetic origin.

In this work, we consider the effect of competing orders and quantum phase fluctuations on the quasiparticle spectral function and density of states (DOS) by incorporating both SC and CO in the bare Green’s function and quantum phase fluctuations in the proper self-energy. Using realistic bandstructures and physical parameters in our calculations and comparing the resulting spectra with experimental data on both hole- and electron-type cuprates, we find favorable comparison of the concept of coexisting CO and SC with a wide range of experimental phenomena \[10\], including the occurrence of excess subgap quasiparticle density of states (DOS) \[13\], spatial modulations in the low-temperature quasiparticle spectra that are unaccounted for by Bogoliubov quasiparticles alone \[11, 20, 21, 22\], “dichotomy” in the momentum-dependent quasiparticle spectral function from ARPES \[23\], and the presence (absence) of the low-energy pseudogap \[13, 24, 25\] in the hole (electron)-type cuprates above the SC transition. We also conjecture that the high-energy pseudogap observed in optical \[16, 17\] and neutron scattering \[18, 19\] experiments on the cuprates may be associated with dynamic spin fluctuations, which can be stabilized by an external field. In this context, we present experimental evidence for current-induced pseudogap in electron-type cuprates \[7, 8\]. Finally, we propose a unified phase diagram for all cuprates based on the findings of competing orders and two pseudogap energy scales.

II. Unconventional low-energy excitations in cuprate superconductors

In order to consider the effect of coexisting CO and SC on the low-energy excitations consistently, we begin by incorporating both CO and SC in the mean-field Hamiltonian, and then include the quantum phase fluctuations associated with the CO and SC phases in the proper self-energy to solve the Dyson’s equation self-consistently for the full Green’s function at \(T = 0\) so as to obtain the quasiparticle spectral density function \(A(k, \omega)\) and the density of states \(N(\omega)\), as detailed in Ref. \[10\]. For electron-type cuprates, we consider the case of coexisting s-wave SC and charge-density waves (CDW) \[4\], whereas for hole-type cuprates, we assume coexisting d-wave SC and disorder-pinned spin-density waves (SDW) \[26, 27\]. We also examine the possibility of d-density waves (DDW) \[2\] being the relevant CO in d-wave SC, and in all cases, we employ the realistic bandstructures and Fermi levels for given cuprates under consideration. Specifically, we consider the bare Green’s function \(G_0(k, \omega)\) associated with the mean-field Hamiltonian

\[
H_{MF} = H_{SC} + H_{CO} \tag{1}
\]

where \(H_{SC}\) is the BCS-like superconducting Hamiltonian for a given pairing potential \(\Delta_{SC}(k)\), with \(\Delta_{SC}(k) = \Delta_{SC}\) being independent of \(k\) in the case of s-wave pairing and \(\Delta_{SC}(k) \approx \Delta_{SC}[\cos k_x a - \cos k_y a]/2\) for d\(_{x^2-y^2}\)-wave pairing. Here \(a\) is the lattice constant of the two-dimensional CuO\(_2\) layer, \(H_{CO}\) is the mean-field CO Hamiltonian with the CO energy scale given
by $V_{CO}$ and the wave-vector of the CO (among CDW, SDW and DDW) given by $Q$ [10]. In the zero-temperature zero-field limit, the quantum phase fluctuations are dominated by the longitudinal phase fluctuations, which can be approximated by the one-loop velocity-velocity correlation in the proper self-energy $\Sigma^* [10]$. Thus, the full Green’s function $G(k, \omega)$ is determined self-consistently through the Dyson’s equation:

$$G^{-1}(k, \omega) = G_0^{-1}(k, \omega) - \Sigma^*(q, \omega),$$

where $\omega$ denotes the energy renormalized by the phase fluctuations. Equation (2) is solved self-consistently [10] 28 by first choosing an energy $\omega$, going over the $k$-values in the Brillouin zone by summing over a finite phase space in $q$, and then finding the corresponding $\xi_k$, $\tilde{\omega}$ and $\tilde{\Delta}$ until the solution to the full Green’s function $G(k, \tilde{\omega})$ converges using iteration method [28]. The converged Green’s function yields the spectral density function $A(k, \omega) \equiv -\text{Im} \left[ \Gamma(k, \tilde{\omega}(k, \omega)) / \pi \right]$ and the DOS $N(\omega) \equiv \sum_k A(k, \omega)$. We also confirm the conservation of spectral weight throughout the self-consistent calculations.

Using the aforementioned approach, we find that many important features in the quasiparticle DOS of both the hole- and electron-type cuprates of varying doping levels can be well accounted for by a set of parameters ($\Delta_{SC}, V_{CO}, \eta$), provided that the CO specified couples well to the Bogoliubov quasiparticles of the cuprates near the Fermi level, and $\eta$ denotes the magnitude of the quantum phase fluctuations specified in Ref. [10]. As exemplified in Fig. 1, we compare the quasiparticle tunneling spectra of three different cuprates of varying doping levels with theoretically calculated quasiparticle DOS for (a) $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ ($\text{Bi}-2212$) [22], (b) $\text{YBa}_2\text{Cu}_3\text{O}_x$ (Y-123) [12] 22, and (c) optimally doped $\text{Sr}_{0.9}\text{La}_{0.1}\text{CuO}_2$ (La-112) [13]. The corresponding SC gap ($\Delta_{SC}$) and CO energy ($V_{CO}$) as a function of the doping level are determined from the fitting and shown in Fig. 1(d). In contrast to the common empirical practice that denotes the peak-to-peak or hump-to-hump energy differences as twice of the nominal SC gap values without considering CO, we find that our theoretical fitting to the quasiparticle DOS not only captures the primary features of the tunneling spectra but also yields doping dependent $\Delta_{SC}$ that better follows the doping dependence of $T_c$, whereas the doping dependence of $V_{CO}$ increases with decreasing doping level.

Another important finding of our calculations is to provide a natural explanation for the presence and the strength of low-energy pseudogap in hole-type and electron-type cuprate superconductors, respectively. That is, generally if $V_{CO} > \Delta_{SC}$, the quasiparticle spectra exhibit two sets of peaks at energies $\omega \approx \pm \Delta_{eff}$ and $\omega \approx \pm \Delta_{SC}$ where $\Delta_{eff} \equiv \sqrt{\Delta_{SC}^2 + V_{CO}^2}$. The peaks at $\pm \Delta_{SC}$ vanish at $T_c$ while those associated with $\pm \Delta_{eff}$ remain finite at $\omega \approx \pm V_{CO}$ above $T_c$ and become much broadened due to thermal fluctuations. Thus, these broadened features at $\omega \approx \pm V_{CO}$ above $T_c$ can be referred to as the lower-energy pseudogap. In contrast, for $V_{CO} < \Delta_{SC}$ and under finite quantum fluctuations, the quasiparticle spectra only exhibit one set of peaks at energies $\omega \approx \pm \Delta_{eff} \approx \pm \Delta_{SC}$ as exemplified in Fig. 1(c), and no discernible low-energy pseudogap can be observed above $T_c$. We therefore conclude that in under- and optimally doped hole-type cuprates, the condition $V_{CO} > \Delta_{SC}$ generally holds, whereas in electron-type cuprates, we generally find $V_{CO} < \Delta_{SC}$. The disparity in the strength of the competing energy scales may be related to the much better electronic coupling with the longitudinal optical phonons [30] for the hole-type electronic configuration along the Cu-O bonding direction in the CuO$_2$ plane, as...
FIG. 1. (a) Comparison of the c-axis (\{001\}) quasiparticle tunneling spectra on nominally underdoped and overdoped Bi-2212 cuprates (left panel, from Ref. [22]) with theoretical calculations (right panel) using the parameters $\Delta_{SC}$ and $V_{CO}$ indicated. (b) Comparison of the anti-nodal (\{100\}) quasiparticle tunneling spectra on underdoped ($T_c \approx 60$ K) and optimally doped ($T_c \approx 93$ K) Y-123 cuprates (symbols, from Ref. [12, 29]) with theoretical calculations (solid lines). We note that the optimally doped Y-123 was not perfectly aligned along the \{100\} so that some contributions from the zero-bias-conductance-peak in the nodal direction [29] were present in the spectra. (c) Comparison of the tunneling spectrum (open squares) on optimally doped La-112 cuprate [13] with theoretical calculations (solid line). (d) Hole-doping dependence of $\Delta_{SC}$, $V_{CO}$ and $T_c$ for Y-123 and Bi-2212, where $\Delta_{SC}$ and $V_{CO}$ are normalized to their corresponding SC gaps at the optimal doping, $\Delta_{SC}^0$, and $T_c$ is also normalized to the value at the optimal doping $T_c^0$. In addition, the onset temperatures for diamagnetism $T_{onset}$ in Bi-2212 with various doping levels from Ref. [15] are shown in crosses for comparison, and we note the good agreement between the doping dependence of ($T_{onset}/T_c^0$) and that of ($V_{CO}/\Delta_{SC}^0$).

discussed in Refs. [7, 10].
FIG. 2. (a) Schematic illustration of the LDOS in a slightly underdoped Bi-2212 system with coexisting SC and CO. Our theoretical analysis of experimental data suggests that $\Delta_{SC}$ generally varies slowly within a given sample, while the apparent spatial variations in the LDOS is primarily due to stronger variations in $V_{CO}$ and $\eta$. Here we denote the A-type region as areas of larger $V_{CO}$ and $\eta$, whereas the B-type region as areas of smaller and nearly homogeneous $V_{CO}$ and $\eta$. (b) An example of varying spectral characteristics due to varying quantum phase fluctuation (parameterized by $\eta$) alone while keeping $\Delta_{SC}$ and $V_{CO}$ constant [10].

In addition to accounting for the primary features of doping dependent quasiparticle tunneling spectra, the notion of coexisting CO and SC can also explain the spatially varying local density of states (LDOS) and the corresponding Fourier transformation of the LDOS (FT-LDOS) in Bi-2212 [20, 21, 22]. As schematically illustrated in Fig. 2(a), the spatially varying LDOS can be simulated by designating spatially varying parameters ($\Delta_{SC}, V_{CO}, \eta$) so that the corresponding LDOS $\mathcal{N}(\mathbf{r}, \omega)$ for a given bulk doping level can reproduce the empirical finding. Specifically, we note that overall $\Delta_{SC}$ does not vary much within each sample, as manifested in Fig. 1(d), while the magnitude of $V_{CO}$ changes more significantly with the doping level and in space, particularly for the underdoped cuprates. We may approximately divide the spectral characteristics of Bi-2212 into two types of regions: the “A”-type region with larger $V_{CO}$ accompanied by stronger fluctuations (large $\eta$), and the “B”-type region with smaller $V_{CO}$ accompanied by small $\eta$. On the other hand, $\Delta_{SC}$ remains relatively homogeneous throughout A and B regions for a sample of a nominal doping level. The total area of the B-type region appears to increases with increasing doping level at the cost of the A-type region. Therefore, the appearance of spectral inhomogeneity may be largely attribute to variations in $V_{CO}$ and $\eta$, as exemplified in Fig. 2(b) for a special case showing spectral variations due to increasing $\eta$ while keeping $\Delta_{SC}$ and $V_{CO}$ fixed. We further note that the variations in $V_{CO}$ and $\eta$ can effectively result in scattering of quasiparticles. Consequently, we expect the FT-LDOS of such quasiparticle spectra in the first Brillouin zone to reveal not only excess contributions associated with the CO at the wave-vector $\mathbf{Q}$ [11], but also quasiparticle interference effects [11] as the result of spatial inhomogeneity in $V_{CO}$ and $\eta$. We further emphasize that the spectral inhomogeneity is NOT the result of large variations in $\Delta_{SC}$. Moreover, $V_{CO}$ in Bi-2212 system apparently exhibit much stronger spatial variations than that in other cuprates such as Y-123 and La-112, probably due to the stronger two dimensionality in Bi-2212.

III. Relevant Competing Orders to Cuprate Superconductivity

While we have demonstrated that CDW and SDW are relevant CO in accounting for the unconventional low-energy excitations and the lower-energy pseudogap phenomena, the im-
portance of considering the realistic bandstructures and Fermi level in determining whether a CO is relevant to cuprate superconductivity cannot be over-emphasized. For instance, in our consideration of either CDW or SDW as the coexisting CO with SC, although the wave-vector $Q$ of the density waves need not be commensurate with $(\pi/a)$, maximum effect of CO on the low-energy excitations only occurs when $|k|$ and $|k+Q|$ are comparable to the Fermi momentum $k_F$ for $Q$ along either $(0,\pi)$ or $(\pi,0)$ direction. Specifically, maximum effect of CO occurs when $|Q| \sim 2k_{Fx}$ for $Q$ along $(0,\pi)$ and $|Q| \sim 2k_{Fy}$ for $Q$ along $(\pi,0)$. If we relax the condition for $Q$ such that $|k+Q|$ deviates substantially from $k_F$, the effect of CO becomes much weakened. As exemplified in Fig. 3, we compare the effective order parameter $\Delta_{\text{eff}}$ in the first quadrant of the Brillouin zone of $s$-wave SC with coexisting CDW (first row) and $d_{x^2-y^2}$-wave SC with coexisting SDW (second row). For the CO wave-vector $Q$ varying from $|Q| < 2k_F$ (left panels), $|Q| = 2k_F$ (middle panels), to $|Q| > 2k_F$ (right panels), we find the strongest CO-induced dichotomy (i.e., anisotropic momentum dependence of $\Delta_{\text{eff}}$) for $|Q| = 2k_F$, implying the maximum effect of the CO on the ground state and the low-energy excitations of the cuprates if the CO wave-vector is correlated with the Fermi momentum.

In this context, we examine the effect of DDW on cuprate superconductivity and find that the DDW order parameter does not couple well with the doped cuprates. Specifically, its effect is only significant for the nearly nested Fermi surface (see Fig. 4(a)), which corresponds to a nearly half-filling (and thus insulating) condition with bandstructures not representative of the cuprates. On the other hand, the contribution of DDW to quasiparticle tunneling spectra becomes essentially uncorrelated with experimental observation if we consider the Fermi surface of a realistic cuprate with a doping level deviating from half-filling, as shown in Fig. 4(b).

IV. Two pseudogap energy scales and the generic phase diagram

Thus far with the notion of coexisting CO and SC and quantum fluctuations, we have successfully accounted for the experimental findings of apparent pseudogap phenomena in ARPES and STM of hole-type cuprate superconductors by the condition $V_{\text{CO}} > \Delta_{\text{SC}}$ and for the absence of such phenomena in electron-type by the condition $V_{\text{CO}} \leq \Delta_{\text{SC}}$. This pseudogap at a lower energy scale (i.e., on the order of $\Delta_{\text{SC}}$) is well correlated with the anomalous Nernst effect [14] and has only been found in hole-type cuprates. In contrast, the higher-energy pseudogap (on the order of the Neel temperature) observed in optical [16, 17] and neutron scattering [18, 19] experiments on both electron- and hole-type cuprates may have its physical origin primarily in magnetism. In the case of Bi-2212 system, the higher-energy pseudogap seems correlated with the so-called “dip-hump” features in the quasiparticle tunneling spectra, whose characteristic energies appear to be related to the energies of spin fluctuations found in Raman spectroscopy and neutron scattering [31]. If the higher-energy pseudogap is indeed of magnetic origin and is dynamic in nature, one would expect that the presence of an external field can stabilize such a dynamic phase, leading to interesting experimental consequences.

A piece of suggestive evidence associated with this conjecture has been recently observed in our tunneling spectroscopic studies of the electron-type La-112 system. As shown in the upper panel of Fig. 5(a), for sufficiently small tunneling currents the quasiparticle spectrum of optimally doped La-112 exhibits no pseudogap above $T_c$ [13], which is as expected because
our fitting to the data reveals $V_{CO} < \Delta_{SC}$ [10]. Interestingly, however, if we increase the tunneling currents beyond a critical value ($I_{cr} \sim 45$ nA) that corresponds to a local magnetic field on the order of a few tens of Tesla, we find that the original sharp peaks associated with $\omega = \pm \Delta_{eff} \sim \pm \Delta_{SC}$ become suppressed, and a second set of peaks at higher energies $\omega = \pm V_{PG}$ emerge [7, 8], as shown in the lower panel of Figure 5(a). The magnitude of $V_{PG}$ is significantly larger than $\Delta_{SC}$, and is therefore of a different physical origin from the competing order energy scale $V_{CO}$ and appears to have been stabilized by the large tunneling currents.

Based on our theoretical calculations and existing experimental observation of ours and others, we suggest that the generic temperature ($T$) vs. doping level ($\delta$) phase diagram of the cuprates is determined by the interplay of three primary energy scales: $V_{PG}$, $V_{CO}$ and $\Delta_{SC}$,
which correspond to temperature scales of $T_{PG}(\delta)$, $T^*(\delta)$ and $T_c(\delta)$. In the case of hole-type cuprates, generally $V_{CO} > \Delta_{SC}$ for a wide range of doping levels, probably due to enhanced charge transfer along the Cu-O bonding as the result of significant coupling of the electronic configuration to the longitudinal optical (LO) phonons 30, so that the CO phase occurs at $T^*(\delta) > T_c(\delta)$. In contrast, there is no discernible charge transfer due to the LO phonons in electron-type cuprates, and the corresponding competing order energy scale becomes much weaker so that $V_{CO} < \Delta_{SC}$. As a result, there is no apparent pseudogap associated with electron-type cuprates in the absence of external magnetic fields 13, 25. On the other hand, the higher-energy pseudogap exists in both electron- and hole-type cuprates, which may be related to spin fluctuations and thus $V_{PG} \gg \Delta_{SC}$. In the language of the slave-boson theory 5, $V_{PG}$ may be thought of as the spinon pseudogap in the underdoped limit. Hence, our proposed phase diagram in Fig. 5(b) has effectively unified the seemingly puzzling asymmetric phenomena between hole- and electron-type cuprates. In particular, we may consider the spinon pseudogap phase determined by the energy scale $V_{PG}$ as the highly degenerate “parent phase” of all cuprates, with antiferromagnetism (AFM), SC and CO being the broken-symmetry phases derived from the parent phase.

V. Summary

In summary, we have investigated the physical origin of unconventional low-energy excitations in cuprate superconductors by considering the effect of coexisting competing orders (CO) and superconductivity (SC) and that of quantum fluctuations and other bosonic modes on the low-energy charge excitation spectra. By incorporating both SC and CO in the bare Green’s function and quantum phase fluctuations in the proper self-energy, we can consistently account for various empirical findings in both hole- and electron-type cuprates. Moreover, based on our tunneling spectroscopic studies and experiments of others, we suggest that there are two energy scales associated with the pseudogap phenomena, with the high-energy pseudogap $V_{PG}$ probably of magnetic origin and the low-energy pseudogap $V_{CO}$ associated with the competing orders. A generic phase diagram of the cuprates that unifies various asymmetric phenomena in electron-
FIG. 5. (a) Evidence for two pseudogap energy scales in the electron-type La-112 system: The absence of the lower-energy pseudogap above $T_c$ \[13\] shown in the upper panel can be attributed to the condition $V_{CO} < \Delta_{SC}$, whereas the occurrence of a higher-energy pseudogap $V_{PG}$ induced by large tunneling currents at $T \ll T_c$ \[7, 8\] may be attributed a form of bosonic excitations stabilized by an effective large magnetic fields. (b) A proposed temperature ($T$) vs. doping level ($\delta$) generic phase diagram electron- and hole-type cuprates.

and hole-type cuprates is proposed, in which the interplay of three primary energy scales $V_{PG}$, $V_{CO}$ and $\Delta_{SC}$ determines the ground state phases and the unconventional low-energy excitations of the cuprates.

Acknowledgment

The work at Caltech is supported by the National Science Foundation through Grants #DMR-0405088, and at the Pohang University by the Ministry of Science and Technology of Korea. We thank Professor Xiao-Gang Wen for stimulating discussion.

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