

**QUASIPARTICLE SPECTROSCOPY AND HIGH-FIELD
PHASE DIAGRAMS OF CUPRATE SUPERCONDUCTORS
– AN INVESTIGATION OF COMPETING ORDERS
AND QUANTUM CRITICALITY**

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We present scanning tunneling spectroscopic and high-field thermodynamic studies of hole- and electron-doped (p- and n-type) cuprate superconductors. Our experimental results are consistent with the notion that the ground state of cuprates is in proximity to a quantum critical point (QCP) that separates a pure superconducting (SC) phase from a phase comprised of coexisting SC and a competing order, and the competing order is likely a spin-density wave (SDW). The effect of applied magnetic field, tunneling current, and disorder on the revelation of competing orders and on the low-energy excitations of the cuprates is discussed.

Keywords: Competing orders; quantum critical point; spin density waves; pseudogap.

1. Introduction

There has been emerging consensus that the existence of competing orders^{1,2,3} in the ground state of cuprate superconductors is likely responsible for a variety of non-universal phenomena such as the pairing symmetry, pseudogap, commensuration of the low-energy spin excitations, and the spectral homogeneity of quasiparticle spectra.^{4,5,6,7} Among probable competing orders such as the spin-density waves (SDW),^{8,9} charge-density waves (CDW),¹⁰ stripes,³ and the staggered-flux phase,¹¹ the dominant competing order and its interplay with superconductivity (SC) remain not well understood. In this work, we report experimental investigation of these issues via quasiparticle spectroscopic and high-field thermodynamic studies. We compare our results with a conjecture that cuprate superconductivity occurs near a quantum critical point (QCP)^{2,12} that separates a pure SC phase from a phase with coexisting SC and SDW.^{8,9} The scenario of SDW as the relevant competing order can be rationalized by the proximity of cuprate SC to the Mott antiferromagnetism, and also by experimental evidence for spin fluctuations in the SC state of various

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cuprates.^{13,14,15,16} Possible relevance of SDW in the cuprates to the occurrence of strong quantum fluctuations and the pseudogap phenomenon will be discussed.

2. Competing Orders and Quantum Criticality

We consider a conjecture of competing SDW and SC near a non-universal QCP at $\alpha = \alpha_c$,⁸ where α is a material-dependent parameter that may represent the doping level, the electronic anisotropy, spin correlation, orbital ordering, or the degree of disorder for a given family of cuprates. As schematically illustrated in Fig. 1(a), in the absence of magnetic field H , the ground state consists of a pure SC phase if $\alpha_c < \alpha < \alpha_2$, a pure SDW phase if $\alpha < \alpha_1$, and a SDW/SC coexisting state if $\alpha_1 < \alpha < \alpha_c$. Upon applying magnetic field, delocalized spin fluctuations can be induced due to magnetic scattering from excitons around vortex cores, eventually leading to the occurrence of SDW coexisting with SC for fields satisfying $H^*(\alpha) < H < H_{c2}(\alpha)$ if the cuprate is sufficiently close to the QCP,⁸ and H_{c2} is the upper critical field. In general we expect stronger quantum fluctuations of the SC order parameter in the SDW/SC phase than in the pure SC phase because of excess low-energy excitations associated with the competing SDW. In principle such a difference between coexisting SDW/SC and pure SC phases can be manifested in the field dependence of the thermodynamic properties of the cuprates at $T \rightarrow 0$. That is, the proximity of a cuprate to the QCP at α_c can be estimated by determining a characteristic field H^* using thermodynamic measurements at $T \rightarrow 0$, and a smaller magnitude of (H^*/H_{c2}^0) would indicate a closer proximity to α_c if $\alpha > \alpha_c$, where H_{c2}^0 denotes the upper critical field of a given sample at $T = 0$. In contrast, for a cuprate superconductor with $\alpha_1 < \alpha < \alpha_c$, we find $H^* = 0$ so that SDW coexists with SC even in the absence of external fields, implying gapless SDW excitations (i.e. $\Delta_{SDW} = 0$) and strong excess fluctuations in the SC state. Thus, the thermodynamic quantity $h^* \equiv (H^*/H_{c2}^0)$ for a given cuprate is expected to reflect its susceptibility to low-energy excitations and its SC stiffness, which can be confirmed via studies of the quasiparticle spectra taken with a low-temperature scanning tunneling microscope (STM).

3. Experimental Approach and Results

To investigate the conjecture outlined above, we employ in this work measurements of the penetration depth $\lambda(T, H)$, magnetization $M(T, H)$, and third-harmonic susceptibility $\chi_3(T, H)$ on different cuprates to determine the irreversibility field $H_{irr}(T)$ and the upper critical field $H_{c2}(T)$. The degree of quantum fluctuations in each sample is then estimated by the ratio $h^* \equiv (H^*/H_{c2}^0)$, where the characteristic field H^* is defined as $H^* \equiv H_{irr}(T \rightarrow 0)$. The magnitude of h^* for different cuprates is determined and compared with the corresponding quasiparticle spectra.

Specifically, the experiments results reported in this work consist of studies on the n-type optimally doped infinite-layer cuprate $\text{La}_{0.1}\text{Sr}_{0.9}\text{CuO}_2$ (La-112, $T_c = 43$ K) and one-layer $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-\delta}$ (NCCO, $T_c = 21$ K); and the p-type

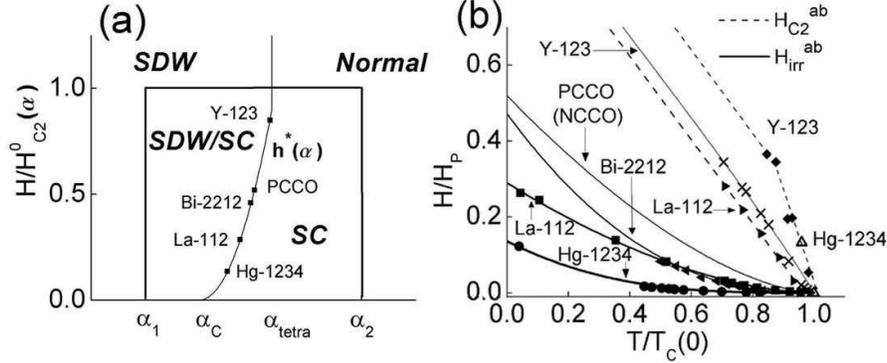


Fig. 1. Phase diagrams of cuprate superconductors based on the conjecture of competing SDW and SC:⁸ (a) Reduced field (H/H_{c2}^0) vs. material parameter (α) phase diagram at $T = 0$. Here $h^*(\alpha) \equiv H^*(\alpha)/H_{c2}^0$ denotes the phase boundary that separates a pure SC phase from a coexisting SDW/SC phase. The points along h^* represent the reduced irreversibility fields, $H_{irr}(T \rightarrow 0)/H_{c2}^0$ obtained from data taken on Y-123^{17,18}, PCCO (NCCO)^{19,20}, Bi-2212²¹, La-112²², and Hg-1234. (b) Comparison of the reduced irreversibility fields (solid lines) and upper critical fields (dashed lines) vs. reduced temperature (T/T_c) phase diagram for Hg-1234, La-112, Bi-2212, NCCO, and Y-123 with $H \parallel ab$ -plane, so that the corresponding H_{c2}^0 is limited by the paramagnetic field H_p .

optimally doped $\text{HgBa}_2\text{Ca}_3\text{Cu}_4\text{O}_x$ (Hg-1234, $T_c = 125$ K) and $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (Y-123, $T_c = 93$ K). Results obtained by other groups on p-type underdoped Y-123 ($T_c = 87$ K),¹⁸ over- and optimally doped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$ (Bi-2212, $T_c = 60$ K and 93 K)^{21,23}; and n-type optimally doped $\text{Pr}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-\delta}$ (PCCO, $T_c = 21$ K),²⁰ are also included for comparison. The optimally doped samples of La-112 and Hg-1234 were prepared under high pressures, with details of the synthesis and characterizations described elsewhere.^{24,25,26} Detailed physical properties of the optimally doped Y-123 single crystal¹⁷ and NCCO epitaxial thin-film¹⁹ have also been given elsewhere.

The $M(T, H)$ measurements were conducted in lower DC fields using a Quantum Design SQUID magnetometer at Caltech, and in high magnetic fields (up to 50 Tesla in a ^3He refrigerator) using a compensated coil in the pulsed-field facilities at the National High Magnetic Field Laboratory (NHMFL) in Los Alamos. The irreversibility field $H_{irr}(T)$ was identified from the onset of reversibility in the $M(T, H)$ loops, as exemplified in the inset of Fig. 2(a) for La-112 and in the main panel of Fig. 2(b) for Hg-1234. The penetration depths of La-112 and Hg-1234 were determined in pulsed fields up to 65 Tesla by measuring the frequency shift Δf of a tunnel diode oscillator (TDO) resonant tank circuit with the sample contained in one of the component inductors.²⁷ Small changes in the resonant frequency can be related to changes in the penetration depth $\Delta\lambda$ by $\Delta\lambda = -\frac{R^2}{r_s} \frac{\Delta f}{f_0}$, where R is the radius of the coil and r_s is the radius of the sample.²⁷ In our case, $R \sim r_s = 0.7$ mm and the reference frequency $f_0 \sim 60$ MHz such that $\Delta f \sim (0.16 \text{ MHz}/\mu\text{m})\Delta\lambda$. Further details

for the pulsed-field measurements of La-112 can be found in Ref.²². Third-harmonic magnetic susceptibility $\chi_3(T, H)$ measurements were also performed on Hg-1234 sample using a 9-Tesla DC magnet and Hall probe techniques.²⁸ The $\chi_3(T, H)$ data measured the non-linear response of the sample and were therefore sensitive to the occurrence of phase transformation.²⁸

Selected data of these thermodynamic measurements of La-112 and Hg-1234 are shown in Figs. 2(a)-(b), and a collection of measured $H_{irr}^{ab}(T)$ and $H_{c2}^{ab}(T)$ curves for various cuprates are summarized in Fig. 1(b), with the reduced characteristic fields (h^*) of several representative cuprates explicitly given in Fig.1(a). We note that the Hg-1234 sample, while having the highest T_c and upper critical field (estimated at $H_p \sim H_{c2}^{ab} \sim 500$ Tesla) among all cuprates shown here, has the lowest reduced irreversibility line ($H_{irr}^{ab}(T)/H_p$), where $H_p \equiv \Delta_{SC}^0/(\sqrt{2}\mu_B)$ is the paramagnetic field, and Δ_{SC}^0 denotes the superconducting gap at $T = 0$. The low irreversibility field is not only due to the extreme two-dimensionality (2D) of Hg-1234²⁶ that leads to strong thermal fluctuations at high temperatures, but also due to its close proximity to the QCP, yielding strong field-induced quantum fluctuations at low temperatures.

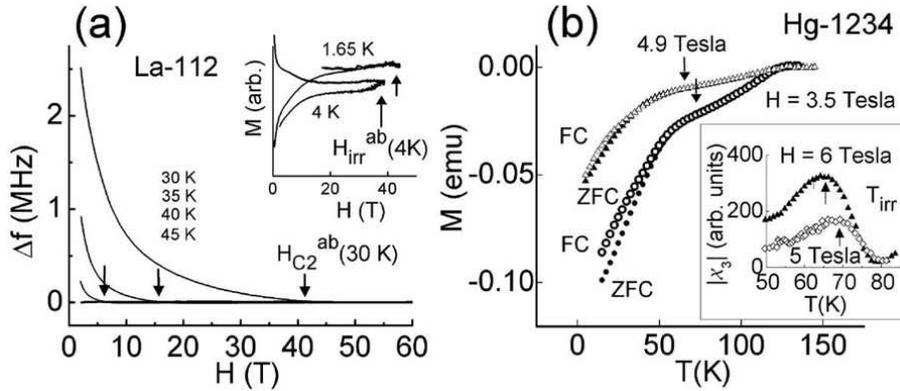


Fig. 2. (a) Main panel: selected data for changes in the resonant frequency Δf of the TDO tank circuit relative to the normal state of La-112 as a function of H at various T . The estimated H_{c2} values for $H \parallel ab$ -plane, $H_{c2}^{ab}(T)$, are indicated by arrows. Similar measurements on a grain-aligned La-112 sample have also been performed (not shown), which yield $H_{c2}^{ab}(T)$. Inset: $M(T, H)$ -vs.- H data on La-112 for $T = 1.65$ and 4.0 K, where the irreversibility field $H_{irr}^{ab}(T)$ are indicated by arrows.²² (b) Main panel: representative zero-field-cooled (ZFC) and field-cooled (FC) $M(T, H)$ -vs.- T data of Hg-1234 taken at $H = 3.5$ and 4.9 Tesla, with the corresponding irreversibility temperature $T_{irr}^{ab}(H)$ indicated by arrows, using the criterion $|M_{ZFC}(T_{irr}, H) - M_{FC}(T_{irr}, H)|/|M(T \rightarrow 0, H \rightarrow 0)| \sim 1.5\%$. Inset: Representative third-harmonic susceptibility²⁸ $|\chi_3(T, H)|$ data for Hg-1234 sample taken at 5 and 6 Tesla, with the corresponding $T_{irr}^{ab}(H)$ indicated by arrows.

To further evaluate our conjecture that cuprates with smaller h^* are in closer proximity to a QCP at α_c and are therefore associated with a smaller SDW gap

Δ_{SDW} and stronger SC fluctuations, we examine the SC energy gap $\Delta_{SC}(T)$ and the quasiparticle low-energy excitations of different cuprates. In Fig. 3(a), we compare the $\Delta_{SC}(T)$ data of La-112, taken with our low-temperature scanning tunneling microscope (STM), with those of Bi-2212 and PCCO obtained from intrinsic tunnel junctions²³ and grain-boundary junctions,²⁰ respectively. Noting that the h^* values are ~ 0.53 for PCCO (NCCO), ~ 0.45 for Bi-2212, and ~ 0.24 for La-112, we find that the rate of decrease in Δ_{SC} with T also follows the same trend. These differences in $\Delta_{SC}(T)$ cannot be attributed to different pairing symmetries, because La-112 is consistent with s -wave pairing symmetry,⁷ NCCO (PCCO) can exhibit s -wave²⁹ or d -wave pairing,³⁰ depending on both the cation and oxygen doping levels,^{31,32} and Bi-2212 is d -wave pairing.^{33,34,35} Therefore, the sharp contrast in the $\Delta_{SC}(T)$ data between La-112 and NCCO (PCCO) suggests that the proximity to the QCP at α_c plays an important role in determining the low-energy excitations of the cuprates. These experimental findings have been further corroborated by recent t - t' - U - V model calculations³⁶ for optimally doped p-type cuprates, which demonstrate that the competing order SDW can appear with increasing temperature even though $\alpha > \alpha_c$ at $T = 0$.

Another experimental confirmation for our conjecture can be found in the quasiparticle tunneling spectra exemplified in Fig. 3(b). We find that excess sub-gap spectral weight exists in La-112, although the quasiparticle spectra of La-112 are momentum-independent and its response to quantum impurities is consistent with s -wave pairing.^{6,7,37} The excess sub-gap spectral weight relative to the BCS prediction implies excess low-energy excitations that cannot be reconciled with simple quasiparticle excitations from a pure SC state, and is therefore strongly suggestive of the presence of a competing order with an additional channel of low-energy excitations. In contrast, substantial details of the quasiparticle spectrum on optimally doped Y-123 (for quasiparticle energies $|E|$ up to $> \sim \Delta_{SC}$) can be explained by the generalized BTK theory,^{5,38} implying that the spectral contribution due to the competing order is insignificant up to $|E| \sim \Delta_{SC}$. This finding is in agreement with a much larger h^* value for Y-123 than for La-112, and therefore much weaker quantum fluctuations in the former.

A further verification for the closer proximity of La-112 to the QCP than Y-123 is manifested in Figs. 4(a)-(b), where the quasiparticle tunneling spectra of La-112 are found to be dependent on the tunneling current I . For lower tunneling currents, we find that the coherence peaks at $E = \pm \Delta_{SC}$ are systematically suppressed by increasing I without changing the Δ_{SC} value. The coherence peaks eventually vanish while additional features at higher energies begin to emerge. Finally, in the large current limit, pseudogap-like features appear at $|E| \equiv \Delta_{PG} > \Delta_{SC}$, as illustrated in Fig. 4(a). More detailed evolution of the superconducting and pseudogap features with I is summarized in Fig. 4(b). We also notice that the magnitude of Δ_{SC} determined under smaller tunneling currents is highly spatially homogeneous, whereas the Δ_{PG} value appears to vary significantly from one location to another.

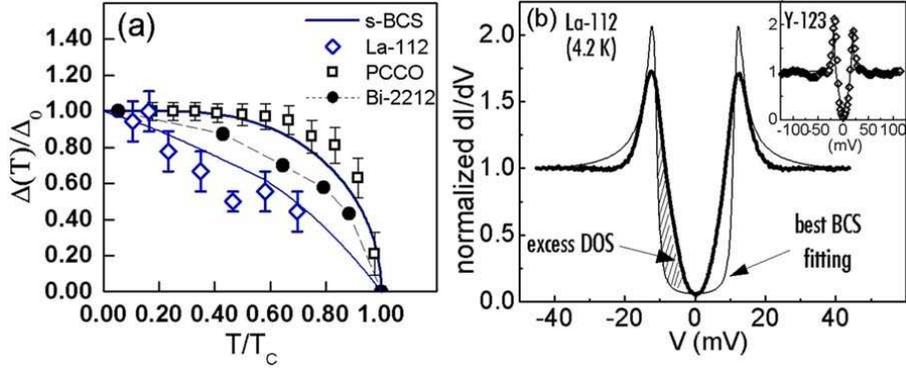


Fig. 3. (a) Comparison of the temperature (T) dependence of the normalized superconducting energy gap, $[\Delta_{SC}(T)/\Delta_{SC}^0]$, for NCCO (PCCO),²⁰ Bi-2212,²³ and La-112. (b) Main panel: comparison of the normalized quasiparticle spectrum of La-112 (points), obtained using a STM at $T = 4.2$ K, with the best BCS fitting (solid line), showing excess sub-gap and reduced post-gap spectral weight. Inset: comparison of the normalized c-axis quasiparticle tunneling spectrum (points) of Y-123, taken using a STM at 4.2 K, with the generalized BTK fitting^{5,38} for $d_{x^2-y^2}$ -wave superconductors (solid line). We note quality agreement between the generalized BTK fitting and data up to $|E| \sim \Delta_{SC}$.

We suggest two effects associated with increasing tunneling current I : First, the localized high current density under the STM tip can effectively suppress the SC order parameter and therefore reduce α . Second, the magnetic field induced by the localized high current density can also assist the evolution from an initial SC phase with $\alpha > \alpha_c$ into the coexisting SDW/SC state with increasing I . The current-induced SDW can be manifested in the DC tunneling spectrum through coupling of the SDW order parameter to disorder.³⁹ Hence, the resulting quasiparticle spectrum under large tunneling currents contains convoluted spectral information of SDW, SC, and the disorder potential, thus it can be spatially inhomogeneous. In contrast, for optimally doped Y-123, no noticeable spectral variation was found with tunneling currents in the same range as that used for studying La-112. This finding is again consistent with our conjecture that Y-123 is farther from the QCP than La-112, so that it is more difficult to induce SDW in Y-123.

4. Disorder Effect on Quasiparticle Spectra and Pseudogap

Next, we investigate the effect of disorder on the scenario depicted in Fig. 1(a). Generally speaking, disorder reduces the SC stiffness and tends to shift α closer to α_c if initially $\alpha > \alpha_c$.¹² Therefore one can envision spatially varying α values in a sample if the disorder potential is spatially inhomogeneous. In particular, for strongly 2D cuprates like Bi-2212 and Hg-1234, disorder can help pin the fluctuating SDW locally,⁴⁰ so that regions with the disorder-pinned SDW can coexist with SC, as schematically illustrated in Fig. 5(a). These randomly distributed regions of pinned SDW are scattering sites for quasiparticles at $T < T_c$ and for normal

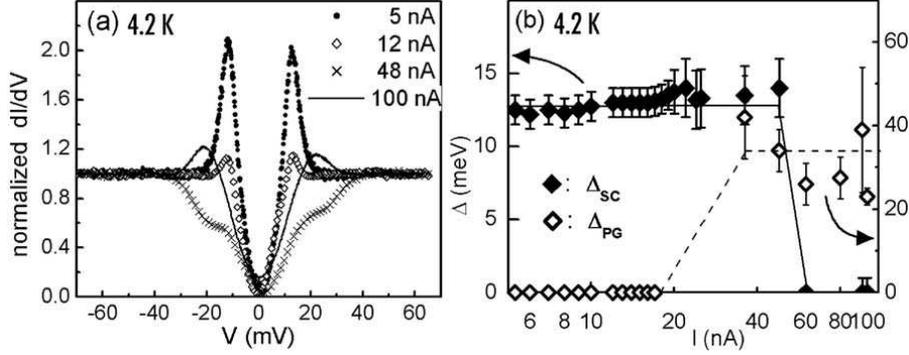


Fig. 4. (a) Evolution of the normalized quasiparticle tunneling spectrum of La-112 with increasing tunneling current I , taken at 4.2 K. (b) Evolution of Δ_{SC} and Δ_{PG} of La-112 with I , as determined from the quasiparticle tunneling spectra.

carriers at $T > T_c$, provided that the SDW persists above T_c . We have performed numerical calculations for the quasiparticle local density of states (LDOS) of a 2D d -wave superconductor to examine these related issues.⁴¹ We have considered two scenarios: one assumes that the ground state of the 2D d -wave cuprate is a pure SC with random point defects, and the other assumes that randomly pinned SDW regions coexist with SC at $T \ll T_c$. Using the Green's function techniques detailed in Ref.⁴¹, we find that the quasiparticle interference spectra for SC coexisting with a disorder-pinned SDW differ fundamentally from those due to pure SC with random point disorder. A representative real-space map of the quasiparticle LDOS, for a 2D $d_{x^2-y^2}$ -wave superconductor with 24 randomly distributed pinned SDW regions in an area of (400×400) unit cells, is shown in Fig. 5(b). The Fourier transformation (FT) of the LDOS is illustrated in Fig. 5(c) for a pure SC with random point disorder at $T = 0$, in Fig. 5(d) for the FT-LDOS of pinned SDW at $T = 0$, and in Fig. 5(e) for the FT-LDOS of pinned SDW at $T = T_c$. We note that the superposition of the FT-LDOS in Figs. 5(c) and 5(d) is consistent with experimental observation on a slightly underdoped Bi-2212 at $T \ll T_c$,^{42,43} whereas the FT-LDOS in Fig. 5(e) is consistent with experimental observation on a similar sample at $T > \sim T_c$.⁴⁴ These findings suggest that a disorder-pinned SDW coexists with SC in Bi-2212 at low temperatures, and that only the SDW persists above T_c .⁴¹ These studies therefore demonstrate the significant role of competing orders in determining the physical properties of cuprate superconductors. Moreover, disorder-pinned collective modes such as SDW can naturally account for the strong spatial inhomogeneity observed in the quasiparticle spectra of Bi-2212,^{45,46} and are also likely responsible for the pseudogap phenomenon⁴⁷ above T_c in under- and optimally doped p-type cuprates.

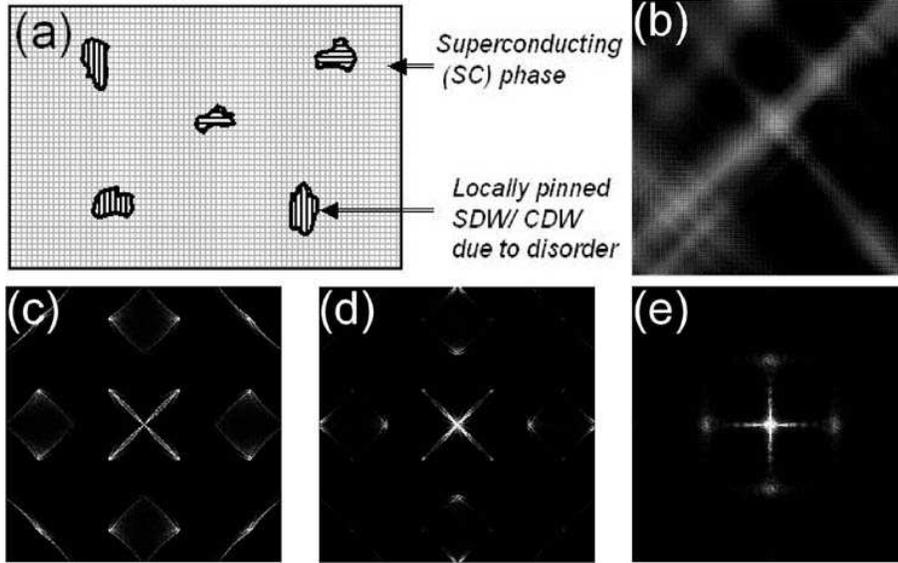


Fig. 5. (a) Schematic illustration of randomly distributed disorder-pinned SDW regions in a 2D superconductor. (b) Quasiparticle LDOS for the scenario depicted in (a), for a $d_{x^2-y^2}$ -wave superconductor at $T = 0$, with a SC gap $\Delta_{SC} = 40$ meV and the quasiparticle energy $E = 24$ meV. (c) Quasiparticle FT-LDOS in the first Brillouin zone of a 2D $d_{x^2-y^2}$ -wave superconductor with random point defects at $T = 0$. (d) The FT-LDOS of Part (b) in the first Brillouin zone at $T = 0$. (e) The FT-LDOS of Part (b) at $T = T_c$. More details are given in Ref. 41.

5. Discussion

The experimental results presented in the previous sections are generally consistent with the notion that significant quantum fluctuations can be induced by a magnetic field in all cuprate superconductors, and that the degree of excess low-energy excitations can be correlated with the proximity of the cuprates to a QCP. However, to further our understanding, systematic studies of more cuprates will be necessary. In particular, neutron scattering studies will be important for determining the SDW gaps of different cuprates. It is also imperative to examine the correlation between the low-temperature high-field phase diagram and the low-energy excitations of different cuprates through quasiparticle spectroscopic studies. For instance, quasiparticle tunneling spectroscopic studies of the highly 2D Hg-1234 can substantiate our conjecture if the following can be verified: (1) $\Delta_{SC}(T)$ decreases more rapidly with T than other p-type cuprates with larger values of h^* ; (2) the quasiparticle DOS exhibits strong spatial variation below T_c , similar to Bi-2212; (3) excess sub-gap DOS than the BTK prediction exists because of the relatively smaller Δ_{SDW} that provides an additional channel of low-energy excitations; and (4) strong quasiparticle spectral dependence on the tunneling current. Similarly, determination of the characteristic field h^* as a function of impurity concentration can provide further

verification for our conjecture that h^* varies with increasing disorder.

Despite consensus for the existence of competing orders in the ground state of the cuprates, whether SDW is the dominant competing order is yet to be further verified. For instance, we note that certain experimental consequences (such as the quasiparticle spectra) due to a disorder-pinned SDW cannot be trivially distinguished from a disorder-pinned CDW in tetragonal cuprates. Therefore neutron scattering studies will be necessary to distinguish between these two competing orders. As for the staggered flux phase, unit-cell doubling features for the quasiparticle LDOS along the CuO_2 bonds must be demonstrated around a vortex core if the staggered flux phase is a relevant competing order.¹¹ Finally, the most important issue remaining to be addressed in the studies of competing orders and quantum criticality is to investigate possible correlation between the existence of competing orders and the occurrence of cuprates superconductivity.

6. Summary

In summary, we have investigated the quasiparticle tunneling spectra and the thermodynamic high-field phase diagrams of various electron- and hole-doped cuprate superconductors. The experimental data reveal significant magnetic field-induced quantum fluctuations in all cuprate superconductors, and the degree of quantum fluctuations appears to correlate well with the magnitude of excess low-energy excitations as the result of competing orders in the ground state. Moreover, our experimental results support the notion that the ground state of cuprates is in proximity to a QCP separating pure SC from coexisting SC/SDW, and our theoretical analysis further suggests that disorder-pinned fluctuating SDW can have significant effect on the LDOS of highly 2D cuprates. Additionally, the persistence of disorder-pinned SDW above T_c may be accountable for the pseudogap phenomenon observed in the spectroscopic studies of highly 2D hole-doped cuprates.

Acknowledgments

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