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Bogoliubov angle, particle-hole mixture, and angle-resolved photoemission spectroscopy in superconductors

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Superconducting state is characterized by excitations—Bogoliubov quasiparticles that represent a coherent mixture of electron ($-e$) and hole ($+e$) components. We propose an observable, which we call *Bogoliubov angle*, which can be extracted using angular resolved photoemission spectroscopy (ARPES) and reveal this particle-hole entanglement by measuring relative weight of particle and hole amplitude of the Bogoliubov quasiparticle. This angle can be measured using the ratio of ARPES spectral intensities at positive and negative energies. Bogoliubov angle could be used to identify variety of possible pairing states and for discussion on possible nature of the pseudogap states that could contain pairing correlations.

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(i) *Bogoliubov angle (BA) and particle-hole mixture in paired states*. Bogoliubov quasiparticles are the analog of the conduction electrons in the superconductors. Unlike electrons, the Bogoliubov quasiparticles do not carry definite charge. The same quantum-mechanical dualism that allows electron to be present at the same time in two states is at play when one considers the Bogoliubov quasiparticles in superconducting state: the quasiparticle is a coherent combination of an electron and its absence ("hole"). This particle-hole dualism of quasiparticles is responsible for a variety of profound phenomena in superconducting state. For example, Andreev reflection, the particle-hole conversion process, is only possible in superconductor due to the particle-hole dualism.

In this Rapid Communication we introduce a quantity that parametrizes the particle and hole mixture in terms of an angle we call BA. We discuss how angular resolved photoemission spectroscopy (ARPES) measurements allow one to extract the Bogoliubov angle from ARPES intensities and thus how one can reveal particle-hole dualism. Here we introduce BA for ARPES in a similar way as it has been introduced for scanning tunneling microscopy.[1] We present an explicit example of how BA can be extracted from the data using ARPES spectra.

To illustrate the point about particle and hole mixture and BA, we can look at the textbook BCS case first. We will show that convenient definition of BA is

$$\Theta_k = \arctan\left[\left(\frac{|u(\mathbf{k})|^2}{|v(\mathbf{k})|^2}\right)^{1/2}\right], \qquad (1)$$

with the conventional coherence factors that are

$$|u(\mathbf{k})|^2 = \frac{1}{2}\left(1 + \frac{\xi_\mathbf{k}}{E_\mathbf{k}}\right), \quad |v(\mathbf{k})|^2 = \frac{1}{2}\left(1 - \frac{\xi_\mathbf{k}}{E_\mathbf{k}}\right), \qquad (2)$$

where $\xi_\mathbf{k}$ is a normal quasiparticle energy counted from chemical potential and $E_{bk}^2 = \xi_\mathbf{k}^2 + |\Delta_k|^2$ is a superconducting state single-particle energy with the gap $\Delta_\mathbf{k}$.

Bogoliubov, Anderson, Nambu, and others have shown[2,3] that natural excitations in the superconducting state are a linear combination of particle and hole excitations with the coherence factors $u_\mathbf{k}$ and $v_\mathbf{k}$. They describe the unitary transformation from particle and hole operators to quasiparticles that have the form

$$\gamma_{\mathbf{k},\uparrow} = u_\mathbf{k} c_{\mathbf{k},\uparrow} + v_\mathbf{k} c^\dagger_{-\mathbf{k},\downarrow}, \qquad (3)$$

with the constraint $|u_\mathbf{k}|^2 + |v_\mathbf{k}|^2 = 1$ for any $\mathbf{k}$ (normalization). This constraint is satisfied with the choice

$$|u_k|^2 = \sin^2 \Theta_k,$$

$$|v_k|^2 = \cos^2 \Theta_k. \qquad (4)$$

To make a connection to ARPES we use the identity[2]

$$A(k,\omega > 0) = |u(\mathbf{k})|^2 \delta(\omega - \mathbf{E}_\mathbf{k}),$$

$$A(k,\omega < 0) = |v(\mathbf{k})|^2 \delta(\omega + \mathbf{E}_\mathbf{k}). \qquad (5)$$

Thus we have finally the main result to be used in ARPES analysis,

$$\Theta_k = \arctan\left(\frac{A(\mathbf{k},\omega > 0)}{A(\mathbf{k},\omega < 0)}\right)^{1/2}. \qquad (6)$$

Experimentally $A(\mathbf{k},\omega>0)$, $A(\mathbf{k},\omega<0)$ can be determined using ARPES intensity at positive (above E_F) and negative (below E_F) energies. We will focus on the peak of the spectral function and assume a smooth energy independent background in the energy range near Fermi surface, where peaks in spectral functions are seen.

Θ_k is the central quantity we are interested in and we define it as a Bogoliubov angle (see related local scanned probes discussion in Ref. 1). It represents a mixture between particle and hole excitations for an eigenstate n (momentum k eigenstate in this context). For example, for $\Theta_k=0$ the Bogoliubov excitation will be a hole. In the opposite case of $\Theta_k=\pi/2$ quasiparticle is essentially an electron. The angle that corresponds to the strongest admixture between particle and holes is $\Theta_k=\pi/4=45°$.

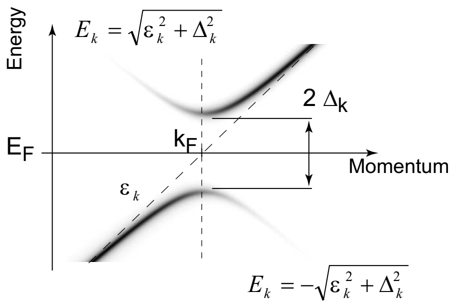

FIG. 1. Particle hole admixture in conventional BCS superconductor as a function of energy is shown. The experimental challenge is to find out if BA is a function of momentum on FS in real materials such as high-T_c superconductors.

(ii) *ARPES and BA analysis*. We will now demonstrate how one can extract BA from ARPES. Although ARPES probes mostly the occupied portion of the single-particle spectral function[4] (i.e., states below E_F, $\omega < 0$) due to the Fermi-Dirac function $f(\omega, T)$ cutoff near E_F, it is still possible to obtain some information about the states above E_F. Because of the high T_c of some superconducting cuprates, ARPES measurements could be performed in the superconducting state at a relatively high temperature, such that the upper branch of the Bogoliubov quasiparticle dispersion [i.e., $A(k, \omega > 0)$] near the nodal region, where the gap is smaller, could lay within the energy range where the value of $f(\omega, T)$ still appreciably differs from zero. In this situation, the Bogoliubov band dispersion above E_F could be seen in the raw spectra allowing a further analysis of its property.[5]

In a recent study of a high-T_c cuprate, Bi2212, the temperature dependence of the Bogoliubov dispersion was measured revealing a sudden onset of the superconducting gap at T_c near the nodal region.[6] In this section, we address the Bogoliubov angle analysis near the nodal region to demonstrate that angle can be extracted from ARPES data. Experimental details of the data presented here can be found in Ref. 6.

Figure 2(a) demonstrates a false-color plot of raw ARPES spectra along the cut position indicated in the inset of Fig. 2(d) at 87 K. In addition to the high intensity region below the Fermi energy (the occupied band dispersion), there is also a less-bright region above E_F, which is the thermally populated Bogoliubov band above E_F. The raw energy distribution curves (EDCs) in the region where the Bogoliubov quasiparticle dispersion is visible are displayed in Fig. 2(c). A small peak above E_F representing the upper branch of Bogoliubov dispersion can be clearly seen near the Fermi crossing momentum k_F, where the gap is minimal. This small peak above E_F becomes less pronounced when we move away from k_F. If the peak position of the Bogoliubov dispersion is moving away from E_F (see also Fig. 1), there are fewer thermally populated excitations at this temperature. Therefore intensity of ARPES signal at positive energy rapidly decreases.

To illuminate these small features above E_F, the ARPES spectrum is divided by an effective Fermi-Dirac (FD) function, which is generated by convolving $f(\omega, T=87\ K)$ with 3.2 meV instrument resolution via a Gaussian convolution.

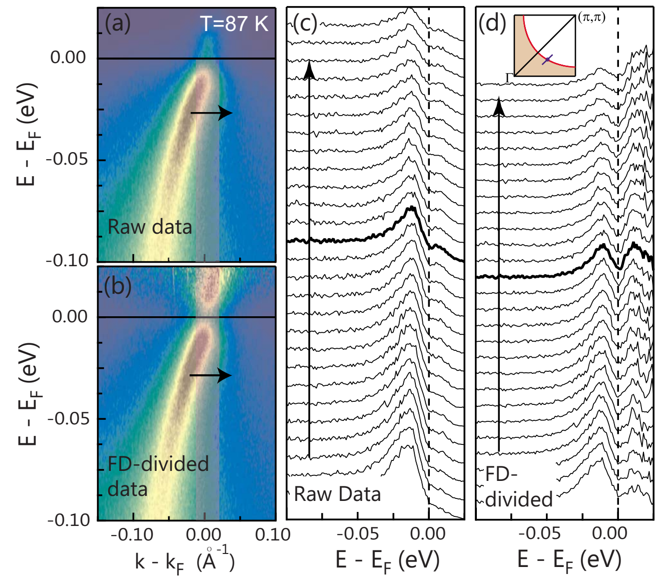

FIG. 2. (Color online) (a) The false-color plot of raw ARPES spectra along the cut indicated in the inset of (d) in unit of the detector angle. The data were taken on a slightly underdoped Bi2212 single crystal with T_c=92 K at a temperature of 87 K using 7 eV photons to excite the photoelectrons. The total-energy resolution is set to 3.2 meV. (b) Fermi-Dirac function divided spectrum of (a). (c) EDC stack plot of the raw spectra within the shaded area of (a). (d) EDC stack plot of the FD-divided spectra within the same shade area as indicated in (b). The thick solid curve is the EDC where $|u_k|^2 \sim |v_k|^2$ and could be defined as the Fermi crossing point, k_F.

The FD-divided ARPES spectrum image is shown in Fig. 2(b). The intensity break near the E_F vividly demonstrates the existence of a gap and two branches of dispersion centered at E_F, as expected for the Bogoliubov quasiparticle dispersion of a superconductor. The EDCs within the shaded area are plotted in Fig. 2(d). Two peaks in each EDC can now be clearly seen exhibiting an evolution of the relative peak height at different momentum positions due to the coherence factors. Before reaching k_F (EDCs below the thick solid curve), the peak below E_F has a higher intensity than that above E_F. After passing k_F (EDCs above the thick solid curves), the relation reverses; the peak above E_F now has a higher intensity than that below E_F. This crossover behavior near k_F is also known to be a characteristic of the Bogoliubov quasiparticle dispersion of a superconductor. Since the peak intensities are related to the coherence factors $|u_k|^2$ and $|v_k|^2$, they could be used for the Bogoliubov angle analysis. Here we note that we are going beyond the claim of existence of the particle-hole peaks, as was clearly demonstrated in Ref. 5. We introduce a specific spectroscopic quantity that captures this particle-hole mixture in a direct way and propose a specific analysis of the relative intensities above or below Fermi energy to extract BA.

To extract the peak intensity, we simply fit the FD-divided EDCs with two equal width Lorentzians in a narrow energy widow, ranging from −20 to 20 meV, in which the signal has not yet completely masked by the amplified noise due to the Fermi function division. This noise amplification at higher

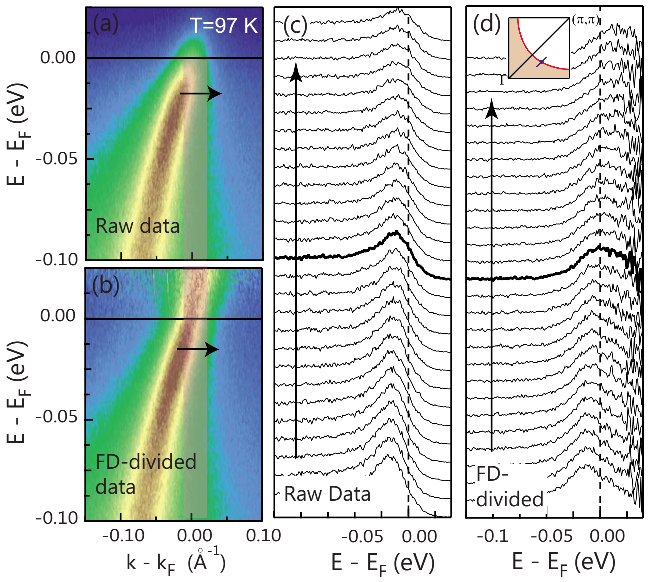

FIG. 3. (Color online) Spectra taken at a temperature of $T=97$ K (above T_c) at the same momentum position as that shown in Fig. 2.

energy above E_F is also the main reason why we use the peak intensity for the BA analysis instead of using peak area. We also note that this Lorentzian fitting is primarily used for obtaining the peak height not for achieving a good fit to the spectrum line shape. A small constant background fit from the raw spectrum well above E_F (+30–+50 meV) has been subtracted before the FD function division. This small constant background is due to the photon contamination of the higher harmonic light from the monochromator. No other background was used when fitting the peak height of the FD-divided spectrum. Figure 4(a) shows the extracted peak heights, which are normalized by the average sum of the two peaks in each FD-divided EDC within this momentum region. The normalized peak height of the peak below E_F is assigned to $|v_k|^2$, while that of the peak above E_F is assigned to $|u_k|^2$. The $|u_k|^2$ and $|v_k|^2$ extracted form the data are qualitatively consistent with what is expected in the conventional BCS superconductor. We also note that the sum of the extracted $|u_k|^2$ and $|v_k|^2$ is constant within the experimental error bars. This suggests that the normalization condition of $|u_k|^2+|v_k|^2=1$ is satisfied at this momentum position. This analysis is fully consistent with an earlier work on a different cuprate.[5]

The Bogoliubov angle Θ_k calculated using Eq. (6) is displayed in Fig. 4(b). The Θ_k increase monotonically across the Fermi crossing point k_F suggesting a continuous evolution of the particle and hole mixing within this momentum window. Furthermore, $\Theta_k=\pi/4$ at k_F within the error bar of our experimental data. This confirms our observation that particle and hole mix equally at k_F, as expected for a superconductor.

Situation is very different, however, at $T>T_c$. Neither Bogoliubov quasiparticle dispersion nor a spectral gap at E_F can be resolved at this momentum position, as can be seen in Fig. 3 (see also Ref. 6). There is only one peak that can be identified in the Fermi function divided EDCs, which disperses

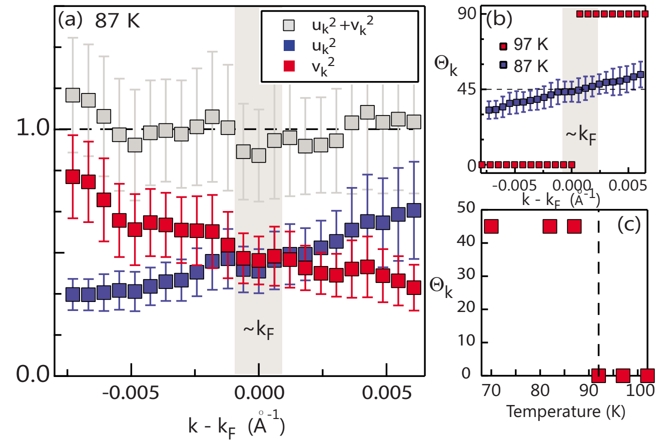

FIG. 4. (Color online) (a) The extracted $|u_k|^2$ and $|v_k|^2$ at 87 K. (b) The Bogoliubov angle Θ_k near the Fermi crossing point at 87 K. (c) The temperature dependence of Θ_k at k_F. The error bars are estimated by the 99.7% confidence interval of the fitting parameters.

across E_F [Figs. 3(b) and 3(d)] suggesting that there is no gap in the spectrum. To generalize Bogoliubov angle for this situation, we define $(u_k,v_k)=(1,0)$ when the quasiparticle peak in the FD-divided EDC is below or at E_F and $(u_k,v_k)=(0,1)$ when the peak of the FD-divided EDC is above E_F. Hence we used the notion that Bogoliubov angle is zero for filled states. This convention will induce a jump from 0° to 90° near the k_F as shown in Fig. 4(b). Thus, Θ_k equals to zero at k_F and a sudden jump across the K_F suggests an absence of the particle and hole mixing above T_c at this momentum position. The Θ_k at k_F at several different temperatures is shown in Fig. 4(c). Behavior of the electron at this momentum position (near nodal region) appears to be conventional despite the existence of a pseudogap (PG) near the antinodal region (see also Ref. 6 and the reference therein).

The BA at several different momentum positions within the near nodal region of the Fermi surface at a temperature of 82 K is shown in Fig. 5(a). The BA is found to be $\pi/4$ suggesting again that the particle and hole mix equally on the Fermi surface at least near the nodal region. For the region in the shaded area indicated in Fig. 5(a), the upper branches of the Bogoliubov band disperse far way from the E_F due to a larger gap magnitude and become less accessible by the excitations with thermal energy at this temperature. We note that although the Bogoliubov peak above E_F can still be identified in the raw EDCs near k_F up to $\phi=15°$, the Bogoliubov peak above E_F in the FD-divided spectrum is too noisy to be useful for the BA analysis [thinner curves in Fig. 5(b)]. Therefore, we cannot obtain any definitive information about the BA for the momentum positions within this shaded area of Fig. 5. We also remark that at a temperature above T_c [lower panel of Fig. 5(b)], the Bogoliubov quasiparticle peak *above* E_F has not yet been resolved in the pseudogap state in this momentum region even through it can be seen at 82 K. This may suggest a qualitative difference of the particle-hole mixing in the pseudogap state from that of the superconducting state. However, we could not rule out the possibility that the absence of a clear signature of a Bogoliubov quasiparti-

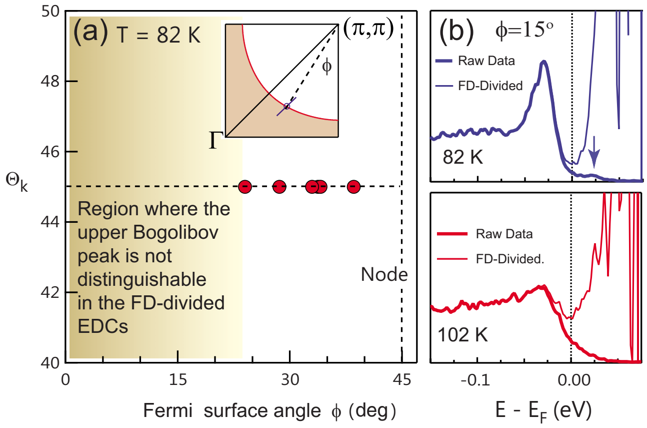

FIG. 5. (Color online) (a) The Bogoliubov angle at a temperature 82 K along the Fermi surface where the Bogoliubov peak can be unambiguously distinguished from the noise level in the FD-divided spectrum. (b) Raw EDCs (thicker curves) and the corresponding FD-divided EDCs (thinner curves) at $\phi=15°$ at temperatures above (102 K) and below T_c (82 K). At this momentum position, a gap (pseudogap) still exists in the spectrum of 102 K. We note that the FD-divided spectrum at this momentum position is too noisy to be useful for BA analysis.

cle dispersion feature above T_C at this momentum position is due to the broadening of the peak in the spectrum. Further study is needed to clarify this issue.

In conclusion, we have introduced a spectroscopic measure, the so-called Bogoliubov angle $\Theta(\mathbf{k})$ that can be extracted from existing ARPES data. This measure allows one to directly image particle-hole admixture in the superconducting state using observed ARPES intensities.

The ideas presented here are quite general and are applicable to a variety of superconductors, including conventional superconductors. One can investigate Bogoliubov angle in a variety of states, including vortex state and normal state with superconducting correlations, e.g., the so-called PG state. As a future application of these ideas we suggest using Bogoliubov angle to identify how robust the particle-hole mixture is in the normal state of cuprates. At present stage we do not have enough resolution to perform this analysis. Another interesting question is the BA behavior with temperature. Answers to these questions will shed light on the nature of PG state and would allow us to differentiate between different scenarios of PG state, e.g., flux phases[7] and *D*-wave density wave (DDW).[8]

Note added: After this Rapid Communication had been submitted, we became aware of the recent paper by Yang *et al.*,[9] where similar mixture of particle and hole component is discussed without using Bogoliubov angle analysis, as proposed here.

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