## Quasiparticle Liquid in the Highly Overdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$

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(Received 19 April 2001; published 8 April 2002)

Results from the study of a highly overdoped (OD) Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8+ $\delta$ </sub> with a  $T_c = 51$  K using angleresolved photoemission spectroscopy are presented. We observe a sharp peak in the spectra near ( $\pi$ , 0) that persists well above  $T_c$ , a nodal self-energy which approaches that seen for the Mo(110) surface state, and a more k-independent line shape at the Fermi surface than the lower-doped cuprates. This allows for a realistic comparison of the lifetime values to the experimental resistivity measurements. These observations point to the validity of the quasiparticle picture for the OD even in the normal state.

DOI: 10.1103/PhysRevLett.88.167006

PACS numbers: 74.25.Jb, 71.18.+y, 79.60.Bm

The question of whether the Fermi liquid (FL) [1] model is applicable for the high- $T_c$  cuprates has been one of the major issues in condensed matter. For the optimally doped (OP) and underdoped (UD) cuprates, there are many results which indicate that the FL picture may not be valid [2,3] especially in the normal state. It is speculated that, as doping increases beyond the optimal value, the cuprate may become a FL [4]. However, experimental evidence from the highly overdoped (OD) regime showing FL properties is scant. Moreover, the term "FL" has become imprecise in recent literature. In the strictest definition of a FL, transport properties and the self-energy,  $\Sigma$ , of the single-particle excitations or quasiparticles (QP) are proportional to the square of energy,  $\omega^2$ , and temperature,  $T^2$ , reflecting the electron-electron interactions. For regular metals, this is not the case for most T and  $\omega$  because the electron-phonon interaction dominates both the scattering rate and the resistivity. At the other extreme, the term FL has been used to describe any material that has singleparticle excitations regardless of the nature of the QP scattering or transport. Indeed, this has been used to argue for the presence of QP in the OP and UD cuprates from angleresolved photoemission spectroscopy (ARPES) below  $T_c$ [5]. However, the existence of a sharp peak is not in itself proof of a FL. In the normal state of the OP and UD, there may be a sharp peak, but only in the nodal direction. In the superconducting state, although the sharp peak appears around the entire Fermi surface, it is only within a short range of  $\omega$ , and the line shape is highly anisotropic. Analysis of OP cuprates in the nodal direction reveals that  $\Sigma$  is unlike that for a typical metal [2]. These observations indicate that the presence of a sharp peak in ARPES spectra does not automatically imply that the FL picture is valid.

Because of such uncertainty, we use the term "quasiparticle liquid" (QPL) as a way to describe a system which has single-particle excitations around the Fermi surface that govern the transport properties even if these properties do not have the  $\omega^2$  and  $T^2$  or electron-phonon-like behavior. These excitations can, in principle, be studied using ARPES, which is the best method to determine the nature of the electronic excitations in a 2D solid. In model 2D metals, the extracted  $\Sigma$  values are amenable to analyses in terms of traditional scattering mechanisms [6]. More exotic phenomenologies have been invoked to understand similar data for the OP [2]. This Letter focuses on ARPES results from highly OD Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8+ $\delta$ </sub> (Bi2212) with  $T_c \sim 51$  K. The OD has a clear sharp peak in the ARPES spectra over the whole Fermi surface and well into the nor*mal state*. Furthermore, the line shape of the spectra appears to be more *isotropic* than the lower-doped Bi2212. This enables us to relate ARPES results to transport measurement. Hence, the normal state of the OD exhibits properties of a QPL. Additionally, analysis in the nodal direction shows a linear dependence of Im $\Sigma$  on  $\omega$  and the insensitivity of the scattering rate to temperature changes across  $T_c$ , similar to that observed for OP Bi2212 [2]. Thus, we explicitly describe the challenges to theory in constructing the temperature and doping dependence of  $\Sigma$ .

Single crystals of Bi2212 were synthesized using the floating-zone technique [7] that yielded optimal  $T_c$  onset of 95 K. Overdoping is achieved by annealing the samples in oxygen at 400 °C for 5 days, yielding  $T_c \sim 51$  K [8]. The samples were kept in the cells under oxygen pressure until they were mounted in the vacuum chamber. The crystals were cleaved in situ below 150 K under vacuum with a base pressure of  $8 \times 10^{-11}$  Torr. Crystal orientation was determined using low-energy electron diffraction (LEED). All data shown here were obtained within 12 h after cleaving to minimize variation in doping of the exposed surface with time.  $T_c$  of the samples were determined to be  $\sim$ 51 K  $\pm$  5 K using a SQUID magnetometer. All ARPES measurements were performed at beam line U13UB of the NSLS with 22 eV photon energy and a Scienta SES200 analyzer that simultaneously collects a large energy (0.5 to 1 eV) and angular (12°) window. The resulting energy resolution is  $\sim$ 10 meV with an angular resolution of better than 0.2°.

Figure 1a shows the T dependence of the energy distribution curves (EDC) near the M point ( $\Gamma M$  direction). The spectra show a well-defined sharp peak along with a broader hump that persists well into the normal state, a behavior that was also seen in a similar, less overdoped Bi2212 [9]. This is noticeably different than the OP and UD cuprates where the sharp peak disappears around  $T_c$ [10]. A recent report [11] shows the sharp peak disappearing at slightly above  $T_c$  on lightly OD crystals and claims the peak intensity is a measure of the superfluid density. This cannot be the case for our more OD samples since the sharp peak persists to temperatures well above  $T_c$  where there is no superfluid. There is no change in the line shape as T goes above  $T_c$  other than a shift in the leading edge to indicate the closing of the gap, as shown in Fig. 1b. The gap size estimated from this shift is 10 meV, which is consistent with that observed in tunneling measurements on similar crystals [12]. Furthermore, the spectra show no pseudogap above  $T_c$  at this doping level.

From the spectra in the  $\Gamma Y$  direction, we obtain  $k_F$ for the OD to be  $0.39A^{-1}$  or  $0.34(\pi, \pi)$ , compared to  $0.446A^{-1}$  or  $0.391(\pi, \pi)$  for OP Bi2212 [2], indicating a shift of the Fermi surface towards the  $\Gamma$  point. The width  $\Delta k$  of the momentum distribution curve (MDC) is used to obtain Im $\Sigma$  using the relation  $\hbar v_k \Delta k \sim 2 \text{ Im} \Sigma$  [2], where  $v_k$  is the noninteracting band velocity. Within the energy range studied here,  $v_k$  is assumed to be a constant [13]. Figure 2 shows Im $\Sigma$  at T = 22 K along with similar data for the OP [2] and the Mo [6]. None of the data sets in the figure show Im $\Sigma$  varying as  $\omega^2$  for small energies. As noted earlier, the Mo data can be broken into contributions from three scattering mechanisms: electron-electron, electron-phonon, and defect scattering [6]. The phonon mechanism dominates in the vicinity of  $E_F$ . Unlike Mo,  $2 \text{ Im}\Sigma$  for the OD and the OP Bi2212 [2] have a predominantly linear dependence on  $\omega$ . Within the error bars [14], Im  $\Sigma$  versus  $\omega$  for the OD sample shows no change between data taken at 22 K and that taken above  $T_c$ . It has been shown that a change in the dispersion at different temperatures can result in a different energy dependence of Im $\Sigma$  [15]. We have shown in a previous paper that this OD cuprate shows no significant change in the dispersion across  $T_c$  [13]. This is consistent with the current observation that Im $\Sigma$  versus  $\omega$  looks similar above and below  $T_c$ . Of the three data in Fig. 3, Im $\Sigma$  of the OP sample increases the fastest with  $\omega$  while, for the OD sample, the slope of Im $\Sigma$  versus  $\omega$  is intermediate between the OP and the Mo. It has been previously noted that  $Im\Sigma$  versus  $\omega$  is characteristic of a marginal Fermi liquid (MFL) where Im $\Sigma \sim \max(\omega, T)$  [16]. Such an identification is less clear in the OD sample, but appears possible. What is not possible is an interpretation of  $Im\Sigma$  of the OD in terms of the scattering processes in typical metals.

Figures 3a and 3b show the OD ARPES line shape along the  $\Gamma M$  and the  $\Gamma Y$  directions both below and above  $T_c$ , and for several  $\omega$ . There is a remarkable similarity in the line shape along these two symmetry directions. The main difference is that the broad hump seen for  $\omega_p \sim 12$  meV is present only in the  $\Gamma M$  spectra. This hump becomes less pronounced at higher  $\omega$ , where the spectra look more similar to each other. We compare this to the spectra of OP Bi2212 (Fig. 3c), where there is a dramatic difference between the spectra from the two directions. This is especially true above  $T_c$ , where there is no well-defined peak in the  $\Gamma M$  spectrum. We conclude that, for the OD, there



FIG. 1. EDCs for OD Bi2212 ( $T_c = 51$  K) near the *M* point (see inset) at various temperatures. The data in (a) have been shifted vertically for clarity. Six representative spectra are shown unshifted in (b).



FIG. 2.  $2 \text{ Im}\Sigma$  in the  $\Gamma Y$  direction for OD ( $T_C = 51$  K), OP ( $T_C = 91$  K) [2], and Mo(110) [6]. The legend shows the temperatures where the ARPES spectra were obtained.  $2 \text{ Im}\Sigma$  for the OD was obtained from MDC only and the values have been shifted vertically for clarity, whereas the OP values were obtained from MDC and EDC. The scattering due to impurities has been subtracted from the Mo data. The straight lines are guides to the eye.



FIG. 3. EDC line shapes for the OD [(a) and (b)] and OP [(c)] taken at the *M* point and the nodal direction. Data have been normalized to the intensity of the sharp peak, except for the normal state of OP where it has not been normalized.  $\omega_p$  is the binding energy of the peak intensity.

are QP over the entire Fermi surface even in the normal state, and that  $\Sigma$  is more *k* independent. This is profoundly different than the lower-doped compounds where the line shapes look very different in the two directions, and further there are no QP well above  $T_c$ .

There are several important implications for the OD from the result of Fig. 3. First, the presence of QP near the *M* point should enhance coupling along the *c* axis [17]. This should result in a lower *c*-axis resistivity,  $\rho_c$ , with increasing doping. This appears to be the case from resistivity measurements on Bi2212 [18], where  $\rho_c$  at a particular *T* monotonically decreases with increasing doping. In addition, such enhanced coupling should show the bilayer bands more clearly than for the lower-doped Bi2212 [19]. Since the splitting is negligible in the nodal direction, the analyses of spectra along this direction are not significantly affected.

Second, the isotropic line shape over the entire Fermi surface in the OD spectra would indicate that the ratio of  $\rho_c$  to  $\rho_{ab}$ , the in-plane resistivity, would be smaller with increasing doping. In fact, due to the absence of any pseudogap in the normal state, the *T* dependence of  $\rho_c$  should appear more similar to  $\rho_{ab}$  for the highly OD Bi2212. We expect the ratio  $\rho_c/\rho_{ab}$  over a wide *T* range to vary less dramatically for the highly OD Bi2212 than for the OP and UD. Again, resistivity measurements are consistent with this result [18]. Both  $\rho_c$  and  $\rho_{ab}$  show metallic behavior over the whole *T* range studied for the highly OD cuprate, unlike those for the lower-doped ones which show semiconducting behavior at low *T*.

Third, the more isotropic line shape of the spectra towards the overdoped regime is consistent with the argument that, with increasing hole doping, the influence of the antiferromagnetic "environment" in the system becomes weaker. This is reflected in the weaker coupling strength with increasing doping observed in the nodal direction [13].

Last, in conventional metals, transport is governed by the scattering rate of the QP. The resistivity  $\rho$  in these materials is related to the lifetime or the mean-free path via Boltzmann transport theory that involves an integral over the Fermi surface [20]. The isotropic nature of the line shape and  $\Sigma$  for the OD makes the integral over the Fermi surface relatively trivial and calls for a comparison between the ARPES linewidths and  $\rho_{ab}$ . Although the lifetimes of ARPES QP are not identical to that in transport measurements, they should be roughly similar for most metals [21]. Boltzmann theory gives  $\rho_{ab}$  proportional to  $\Delta k$ , the width in k of the MDC peak at  $E_F$  that is also



FIG. 4. A comparison of the ARPES  $\Delta k$  (open circles) and experimental  $\rho_{ab}$  (solid line) from OD Bi2212 with  $T_c = 58$  K [8]. The error bars for  $\Delta k$  are roughly the size of the data points. The inset shows a fit of the ARPES data to the power law  $\Delta k = A + Bx^{C}$ .

the mean-free path of the ARPES QP. Figure 4 shows a comparison of the ARPES  $\Delta k$  with  $\rho_{ab}$  [8]. The resistivity data come from a different OD crystal that has roughly the same  $T_c$ , the best comparison available. Qualitatively, the curves clearly match.

To quantify the relationship between these quantities, we fit them to a power law of the form  $\rho/\Delta k = A + BT^{C}$ within the temperature range of 55 to 160 K. These fits are for the purpose of quantitatively comparing the temperature dependence over a limited T range and are not meant to imply underlying physics. The important parameter for comparing the functional form is C. This parameter, which determines the curve shape, is the same for the two data sets within the error bars (the fit to  $\Delta k$  is shown in the inset). The values are 2.59  $\pm$  0.5 for  $\Delta k$  and  $2.52 \pm 0.024$  for  $\rho_{ab}$ . The comparison is evidence that, in the normal state,  $\rho_{ab}$  is determined by the same interacting QP created in ARPES over the entire Fermi surface, an important element of a QPL. This is clearly not the case, for example, in the superconducting state, where dc transport proceeds via a two-particle channel. Furthermore, we note that, for OP Bi2212, the comparison between  $\Delta k$  and  $\rho_{ab}$ agrees if we consider only the nodal QP [2]. Since the OP  $\Sigma$  in the normal state are highly anisotropic, a summation over the entire Fermi surface would not produce the same T dependence between the ARPES  $\Delta k$  and  $\rho_{ab}$  [22]. This leads to the conclusion that, for the OP (and UD),  $\rho_{ab}$  is dominated by nodal QP, which is unlike that observed for the OD. We also point out that the smooth variation of  $\Delta k$ through  $T_c$  in the OD data is also seen along the nodal direction in OP Bi2212 [22]. It appears that, for the cuprates,  $\Delta k$  is insensitive to  $T_c$ .

Taken as a whole, the ARPES data indicate that the OD Bi2212 is a QPL in the normal state, though the primary interactions are different than those in a prototypical metal. The key elements arguing for the QPL nature are as follows: (i) the presence of well-defined peaks in the ARPES spectra over a wide range of T,  $\omega$ , and k, along with well-defined peaks for the entire Fermi surface; (ii)  $\Sigma$  that is largely k independent; and (iii)  $\rho_{ab}$  that is proportional to the mean-free path measured in ARPES as is expected from simple Boltzmann transport. These are characteristics of the OD that are different than the OP/UD. On the other hand, the OD also exhibits certain properties in common with the OP;  $Im\Sigma$  that appears to be linear in  $\omega$  and variation in  $\Delta k$  that is insensitive to  $T_c$ . The former seems to indicate that the fundamental interactions between the electrons and other excitations are similar for all of the cuprates, and not like those for a typical metal. One possible scenario involves some form of coupling to the magnetic modes observed in inelastic neutron scattering [23]. The overall changes in the ARPES  $\Sigma$ as a function of T and doping can be matched to the magnetic behavior [13,24]. However, regardless of the details of the interactions, there is a clear difference between the OD and OP cuprates in both the presence of QP and their relation to transport properties.

This work was supported in part by Department of Energy under Contracts No. DE-AC02-98CH10886, No. DE-FG02-00ER45801, and No. DOE-BES W-31-109-ENG-38. We acknowledge discussions with J. Zasadzinski, L. Coffey, P. Allen, D. Feng, and R. Gooding.

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