Observation of a van Hove Singularity in $Bi_2Sr_2Ca_1Cu_2O_{8+x}$ with Angle Resolved Photoemission

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(October 13, 2018)

Abstract

We have performed high energy resolution angle-resolved photoemission studies of the normal state band structure of oxygen overdoped $Bi_2Sr_2Ca_1Cu_2O_{8+x}$. We find that there is an extended saddle point singularity in the density of states along $\Gamma - \bar{M} - Z$ direction. The data also indicate that there is an asymmetry in the Fermi surface for both the $\Gamma - \bar{M} - Z$ and perpendicular directions.

PACS numbers: 73.20.At, 79.60.-i, 74.25.Jb, 74.72.Hs
I. INTRODUCTION:

The possibility of a van Hove singularity has been of considerable interest since it was proposed as a means of enhancing the superconducting transition temperature, $T_c$. \[1–4\] The idea is that the tendency toward superconductivity in a two-dimensional system can be enhanced when the Fermi level lies at or close to the energy of a logarithmic van Hove singularity (VHS) in the density of states. \[2,4\] Several investigators have studied such a mechanism for raising the superconducting transition temperature $T_c$ \[3,9\] and there has been indirect experimental evidence indicating the possibility of a van Hove singularity. \[10,11\] More recently, the first direct spectroscopic observations of a van Hove singularity, in the $YBa_2Cu_3O_{7-x}$ and $YBa_2Cu_4O_{7-x}$ systems have been reported. \[12,13\] Gofron et.al. show that there is an extended saddle point singularity along the $\Gamma - Y$ direction in reciprocal space, centered at the Y-point. \[12\] This report motivated us to study the $Bi_2Sr_2Ca_1Cu_2O_{8+x}$ system.

Our main result is that there is indeed an extended saddle point type of van Hove singularity in the oxygen overdoped $Bi_2Sr_2Ca_1Cu_2O_{8+x}$ system. We have also obtained two additional results: the electronic band structure is asymmetric along both the $\Gamma - \bar{M} - Z$ and perpendicular directions, and there is no evidence for an electronic state just above the Fermi energy along the $\Gamma - \bar{M} - Z$ direction.

II. EXPERIMENTAL:

The experiments were performed using the four meter normal incidence monochromator at the Wisconsin Synchrotron Radiation Center in Stoughton, WI. The beamline provides highly (> 95%) linearly polarized light with the electric vector in the horizontal plane and with photon energy resolution better than 10 meV. The angle-resolved photoemission chamber includes a reverse-view low energy electron diffraction (LEED) optics used to orient the sample \textit{in situ} after cleaving. The electron energy analyzer is a 50 mm VSW hemispherical
analyzer mounted on a two axis goniometer, with an acceptance full angle of 2°. The base pressure is $6 \times 10^{-11}$ torr. The incidence angle between the photon Poynting vector and surface normal was 45° unless otherwise noted.

The single crystal samples were annealed in 1 atmosphere of oxygen at 530 °C for 20 hours. Figure 1 illustrates the results of AC susceptibility measurements. Note particularly that $T_c$ is 83K and the 10-90% temperature width is 1.3K. The crystals were characterized using four-point resistivity, X-ray diffraction, and transmission electron microscopy. The samples were transferred from a load lock chamber with a base pressure of $5 \times 10^{-9}$ torr to the main chamber, and cleaved at 30K in a vacuum of $6 - 8 \times 10^{-11}$ torr. The sample holder includes the capability to rotate the sample in situ about the surface normal, at low temperatures, for precision alignment with respect to the photon electric field. To measure the normal state electronic band structure, the temperature was raised to 95K, above $T_c$. The stability of the temperature was ±1K. We obtained a Fermi edge reference by using the spectra of freshly deposited gold films. We have obtained an energy resolution of 15 meV using 1 eV pass energy. For the present study, the overall energy resolution employed was 35 meV unless otherwise stated.

For a quasi-two-dimensional system such as $Bi_2Sr_2Ca_1Cu_2O_{8+x}$ the initial state of the electron can be determined by measuring the component of the electron momentum parallel to the sample surface ($k_{//}$). By measuring the energy distribution curves (EDC’s) for different directions ($\theta$, $\phi$) of the emitted photoelectron relative to the surface normal, the $k_{//}$ of the initial state is derived from the relation: $k_{//} = 0.512\text{Å}^{-1}\sqrt{E_{\text{kin}}(\sin \theta \cos \phi \hat{k}_x + \sin \phi \hat{k}_y)}$, where $E_{\text{kin}}$ is the kinetic energy of measured photoelectrons in the unit of eV, $\hat{k}_x$ and $\hat{k}_y$ denote unit vectors along two Cu-O bond axis ($\Gamma - \bar{M}$ directions). The binding energy of the state is determined by using the Fermi edge of the gold film as a reference.
III. RESULTS AND DISCUSSION:

We first oriented the sample so that the $\Gamma - \bar{M} - Z$ direction was in the horizontal plane. We measured the dispersion of the normal state quasiparticle band along the $\Gamma - \bar{M} - Z$ horizontal axis. Figure 2(a) and (b) illustrate the spectra obtained using an energy resolution of (a) 55 meV and (b) 35 meV, and a photon energy of (a) 21 eV and (b) 25 eV. The emission angle of the photoelectrons for each spectrum is noted in the figure.

A band develops and is visible for $8^\circ$ off normal at a binding energy of $335 \pm 5$ meV. The band disperses toward the Fermi energy and, at $\theta = 20^\circ$, is at a binding energy of 0-15 meV. The $\bar{M}$ point at the photon energy of 25 eV is at $21^\circ$. For this polarization (even symmetry) there is no indication of a Fermi level crossing, consistent with reports of Dessau et.al. Between $\theta = 20^\circ - 32^\circ$ the quasiparticle band remains at a binding energy of 0-15 meV. Starting at $34^\circ$ the quasiparticle band disperses to higher binding energy, reaching a binding energy of 120 meV at an angle of $40^\circ$. The dispersion curve is illustrated in Fig. 2(c). The data in Fig. 2 illustrate what is, to the best of our knowledge, the first observation of the quasiparticle dispersion near the Z-point which, for this photon energy, is $43^\circ$. All of the above results on the quasiparticle dispersion along the $\Gamma - \bar{M} - Z$ direction were confirmed using a photon energy of 21 eV. This behavior, with the quasiparticle band near the Fermi energy for an extended portion of the $\Gamma - \bar{M} - Z$ line, is consistent with an extended saddle point singularity. Note also that the dispersion is not symmetric with respect to the $\bar{M}$ point; we return later to this point.

The other crucial issue of the band structure topology is the behavior of the quasiparticle band along cuts perpendicular to the $\Gamma - \bar{M} - Z$ direction. To obtain definitive information, we made four cuts, at angles $14^\circ$, $16^\circ$, $18^\circ$ and $20^\circ$ along the $\Gamma - \bar{M} - Z$ direction. In Figs. 3-6, we illustrate the results obtained with 25 eV photon energy. We also confirmed the results of Figs. 3-6 by using a photon energy of 21 eV (data not shown). Fig. 3(a) and (b) illustrate the results for the cut at $\theta = 20^\circ$, very close to the $\bar{M}$ point along the $\Gamma - \bar{M} - Z$ line. Fig.
Figures 3(a) illustrate the spectra along the $Y - \bar{M} - X$ line. Along the $\bar{M} - X$ line, the intensity of the quasiparticle band decreases monotonically with increasing angle. At $\phi = -5^\circ$ only the background remains. The data indicate that the band disperses up through the Fermi energy almost immediately as one moves from $\bar{M}$ along the $\bar{M} - X$ line. By contrast, along the $\bar{M} - Y$ line, the band first disperses slightly away from the Fermi surface, then exhibits a clear Fermi surface crossing at $\phi = +7^\circ$. Figure 3(b) illustrates the dispersion along the $X - \bar{M} - Y$ direction, with Fermi surface crossings as one moves away from the $\bar{M}$ point in either direction, but the Fermi surface crossings in the $\bar{M} - X$ and $\bar{M} - Y$ directions are not symmetric with respect to the $\Gamma - \bar{M} - Z$ line.

Another noteworthy point about Fig. 3(a) is that we obtained data all the way from the $\bar{M}$ point to the $Y$ point. Notice that there is only one Fermi surface crossing visible in the data. Since there is a part of the Fermi surface around the $Y$-point, our data indicate that there is no Bi-pocket around the $\bar{M}$ point along the $\bar{M} - Y$ line. [10]

Figure 4 illustrates the cut at $18^\circ$ parallel to the $X - \bar{M} - Y$ line. The spectra in Fig. 4(a) exhibit an abrupt reduction in the quasiparticle spectral feature intensity between $\theta/\phi = 18^\circ/4^\circ$ and $\theta/\phi = 18^\circ/5^\circ$ in going toward $X$, but the corresponding Fermi surface crossing in going toward $Y$ occurs between $\theta/\phi = 18^\circ/5^\circ$ and $\theta/\phi = 18^\circ/6^\circ$. Fig. 4(b) illustrates the details of the dispersion observed in Fig. 4(a). Note in particular that the dispersion is again asymmetric about the $\Gamma - \bar{M} - Z$ line, with a maximum binding energy of 60 meV at $\phi = +3^\circ$. Also, it is noteworthy that for this cut, again, the quasiparticle state disperses above the Fermi energy as one moves away from the $\Gamma - \bar{M} - Z$ line in either direction.

We also studied a cut at $\theta = 16^\circ$ parallel to the $X - \bar{M} - Y$ line. Figure 5(a) illustrates the spectra, and Fig. 5(b) the resulting dispersion relation. For this cut, the dispersion is almost symmetric, with a saddle point centered at $\phi = -1^\circ$. Note that the quasiparticle state again disperses above the Fermi energy as one moves away from the $\Gamma - \bar{M} - Z$ line in either direction. The results of the cuts at $16^\circ$, $18^\circ$ and $20^\circ$ all indicate that the Fermi surface topology is markedly different around the $X$ and the $Y$ points.
Fig. 6 illustrates a cut at $\theta = 14^\circ$ parallel to the $X - \bar{M} - Y$ line. Note that the binding energy, 130 meV, along the $\Gamma - \bar{M} - Z$ line for this cut places the quasiparticle state well below the Fermi energy. For this reason, we would expect the behavior of the quasiparticle state in this part of the Brillouin zone not to be strongly involved in the superconducting properties. Note, however, that we observe asymmetric Fermi surface crossings along this cut at $\theta/\phi = 14^\circ/ -5^\circ$ and at $\theta/\phi = 14^\circ/ +7^\circ$, another indication that the shape of the Fermi surface around the X and the Y points is different.

Figure 7 summarises (a) our partial Fermi surface mapping and (b) the shape of the Fermi surface near the $\Gamma - \bar{M} - Z$ direction. The mapping is a purely experimental derivation; we have made no a priori assumptions regarding the symmetry of the Fermi surface. As noted above, we observe an asymmetric Fermi surface shape with respect to the $\bar{M}$ point along the $\Gamma - \bar{M} - Z$ line. The cause of this asymmetry is unknown; however, our observations are consistent with our finding that the $\Gamma - Y$ and $\Gamma - X$ lines are inequivalent. Note also that the shape of the Fermi surface is as expected for an extended saddle point type of van Hove singularity.

Our results are different from those reported by Ref. [15] who, however, studied more lightly doped samples (ours are overdoped with oxygen), and who assumed tetragonal symmetry. Our data establish that for the overdoped samples, the Fermi surface exhibits orthorhombic, rather than tetragonal, symmetry. Such a result is consistent with X-ray and transmission electron microscopy studies of our crystals, which both indicate orthorhombic symmetry. Earlier reports indicate that the orthorhombic structure arises from a Bi-O buckling distortion that also affects the $CuO_2$ planes.

In addition to revealing the extended saddle point van Hove singularity, our data allow us to comment on the possibility of an electronic state just above the Fermi energy in the $\Gamma - \bar{M} - Z$ direction. Recently, Ref. [15] argued that on optimally doped $Bi_2Sr_2Ca_1Cu_2O_x$ samples there are two closely spaced electronic states, one of even and the other of mixed symmetry. Along the $\Gamma - \bar{M} - Z$ direction and with even symmetry orientation, our data are, for the wavevector domain in common, similar to those reported by Ref. [15]. However,
as illustrated in Fig. 8, along the $\Gamma - \bar{M} - Z$ direction and with odd symmetry orientation, we find virtually no electronic state.

To insure that this is not due to a matrix element effect, we took data at several photon energies in the 17-30 eV range. The difference noted in Fig. 8 is independent of the photon energy.

If we define a polarization factor $P = (S_{\theta=22^\circ} - S_{\phi=22^\circ})/(S_{\theta=22^\circ} + S_{\phi=22^\circ})$, where $S_{\theta}$ and $S_{\phi}$ represent the photoemission spectral area at $\theta$ and $\phi$ after the subtraction of normal emission spectral area, respectively, the data indicate that for the overdoped samples, the electronic state along the $\Gamma - \bar{M} - Z$ direction is $P = 66\%$ of even symmetry, as indicated in Fig. 8(b). This result contrasts to that of Ref. [15] who, for lighter doped samples, report an electronic state in the odd symmetry orientation that disperses through the Fermi surface along the $\Gamma - \bar{M} - Z$ direction.

The two data sets indicate that the oxygen doping affects the symmetry of the quasiparticle state along the $\Gamma - \bar{M} - Z$ direction. Our results, illustrated in Fig. 8 and repeatedly reproduced for overdoped samples, establish that the quasiparticle state exhibits almost perfect even symmetry in the $\Gamma - \bar{M} - Z$ direction. We emphasize that our data for overdoped samples do not allow us to confirm or to exclude the possibility of a mixed-symmetry state, observable in odd symmetry orientation, that disperses through the Fermi surface. We simply do not observe strong state in the odd symmetry orientation.

**IV. CONCLUSION:**

We have mapped the quasiparticle dispersion along the $\Gamma - \bar{M} - Z$ direction up to nearly the Z point for the first time. We find an extended saddle point type of singularity in the Fermi surface, which can lead to a stronger than logarithmic enhancement of $T_c$. It is noteworthy, as Abrikosov has pointed out, that this enhancement is virtually independent of the choice of exchange boson. We have found that the overdoped $Bi_2Sr_2Ca_1Cu_2O_{8+x}$ system exhibits an orthorhombic, rather than a tetragonal, Fermi surface.
ACKNOWLEDGEMENT We benefitted from conversations with Andrey Chubukov and Robert Joynt. The staff at the Wisconsin Synchrotron Radiation Center, particularly Tom Baraniak, were most helpful. Financial support was provided by the U.S. NSF, both directly (DMR-9214701) and through support of the SRC, by Ecole Polytechnique Fédérale Lausanne and the Fonds National Suisse de la Recherche Scientifique, and by the Deutsche Forschungsgemeinschaft.
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FIGURES

FIG. 1. AC susceptibility data for an oxygen overdoped $Bi_2Sr_2Ca_1Cu_2O_{8+x}$ single crystal annealed in 1 atm $O_2$ at 530 °C for 20 hours. The onset $T_c$ is 83K, with 10-90% transition width $\Delta T = 1.3K$.

FIG. 2. Normal state (T=95K) angle-resolved photoemission spectra for an oxygen overdoped $Bi_2Sr_2Ca_1Cu_2O_{8+x}$ single crystal of $T_c = 83K$ along the $\Gamma - M - Z$ direction in the Brillouin zone. The $\Gamma - M - Z$ direction lies in the photon polarization plane (even symmetry). Photon energy of (a) 21 eV, (b) 25 eV employed. (c) Dispersion curve obtained from (a) and (b). Note the dispersing electronic state that disperses toward the Fermi energy $E_f$ away from $\Gamma$, the extended flat region between 0.75Å$^{-1}$–1.25Å$^{-1}$, and the dispersion away from $E_f$ at wavevectors greater than 1.25Å$^{-1}$. The dotted line indicates the maximum binding energy for which a photoemission superconducting condensate peak is observed.[18]

FIG. 3. (a) Normal state (T=95K) angle-resolved photoemission spectra for an oxygen overdoped $Bi_2Sr_2Ca_1Cu_2O_{8+x}$ single crystal of $T_c = 83K$ along the $\Gamma - M - Z$ direction. The $\Gamma - M - Z$ direction lies in the photon polarization plane (even symmetry). A photon energy of 25 eV was employed. (b) The dispersion curve obtained from Fig. 3(a). Positive values of $k_y$ correspond to a wavevector pointed along the $M - Y$ direction.

FIG. 4. (a).Normal state (95K) angle-resolved photoemission spectra for an oxygen overdoped $Bi_2Sr_2Ca_1Cu_2O_{8+x}$ single crystal of $T_c$ along a direction parallel to the $X - M - Y$ direction at $\theta = 18^\circ$. The $\Gamma - M - Z$ direction lies in the photon polarization plane (even symmetry). The photon energy was 25 eV. (b). The dispersion curve obtained from Fig.4(a). Positive values of $k_y$ correspond to a wavevector parallel to the $M - Y$ direction.
FIG. 5. (a) Normal state (95K) angle-resolved photoemission spectra for an oxygen overdoped Bi$_2$Sr$_2$Ca$_1$Cu$_2$O$_{8+x}$ single crystal $T_c = 83$K along a direction parallel to the $X - \bar{M} - Y$ line at $\theta = 16^\circ$. The $\Gamma - \bar{M} - Z$ direction lies in the photon polarization plane (even symmetry). The photon energy was 25 eV. (b) The dispersion curve obtained from Fig. 5(a). Positive values of $k_y$ correspond to a wave vector parallel to the $\bar{M} - Y$ direction.

FIG. 6. (a) Normal state (95K) angle-resolved photoemission spectra for an oxygen overdoped Bi$_2$Sr$_2$Ca$_1$Cu$_2$O$_{8+x}$ single crystal of $T_c = 83$K along a direction parallel to the $X - \bar{M} - Y$ direction at $\theta = 14^\circ$. The $\Gamma - \bar{M} - Z$ direction lies in the photon polarization plane (even symmetry). The photon energy was 25 eV. (b) The dispersion curve obtained from Fig. 6(a). Positive values of $k_y$ correspond to a wavevector parallel to the $\bar{M} - Y$ direction.

FIG. 7. (a) The experimental positions of the Fermi surface crossings near the $\Gamma - \bar{M} - Z$ direction. The sample exhibits orthorhombic, not tetragonal, symmetry, in agreement with structural characterization. The plane containing the c-axis and the $\Gamma - \bar{M} - Z$ direction is not a plane of reflection symmetry of the Fermi surface. (b) The perspective drawing of the binding energy of the spectral features near the $\Gamma - \bar{M} - Z$ direction. Note that the band disperses through the Fermi energy as one moves away from the $\Gamma - \bar{M} - Z$ line in either perpendicular direction, as expected for a extended saddle point van Hove singularity.

FIG. 8. (a) Normal state ($T = 95$K) angle-resolved photoemission spectra for an oxygen overdoped Bi$_2$Sr$_2$Ca$_1$Cu$_2$O$_{8+x}$ single crystal of $T_c = 83$K along the $\Gamma - \bar{M} - Z$ direction in the Brillouin zone. The $\Gamma - \bar{M} - Z$ direction lies perpendicular to the photon polarization plane (odd symmetry). The photon energy was 21 eV. Note the loss of spectral intensity compared to Fig. 2(a). (b) Direct comparison of the spectra taken for two orientations (odd and even). The dotted lines are the spectra taken at normal emission. All spectra have been normalized above the Fermi energy and at high binding energy.