Superconductivity and Stoichiometry in the BSCCO-family Materials

Marshall Onellion

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We report on magnetization, c-axis and ab-plane resistivity, critical current, electronic band structure and superconducting gap properties. Bulk measurements and photoemission data were taken on similar samples.

KEY WORDS: Resistivity; band structure; superconducting gap; symmetry.

I have two purposes in writing this report. One is to provide experimental data that has been reproduced in different laboratories, and so can be viewed as reliable, to serve as the basis for theoretical models. The other is to argue that there is a close connection between changes observed in the superconducting properties (gap size and temperature dependence, critical current, magnetic field dependence) and normal state properties (symmetry of electronic states, topology of Fermi surface, c-axis resistivity).

In our studies, we have found c-axis resistivity data versus oxygen content that are consistent with and extend earlier reports.[1-4] Note that the c-axis resistivity is reduced by a factor of as much as \times 250 for overdoped compared to underdoped samples. However, neither we nor our colleagues [1-4] have yet incorporated sufficient oxygen to observe fully metallic (dR/dT > 0) behavior for all temperatures down to the superconducting transition temperature ($T_c$), as has been observed for YBCO-123.[5] The significant point to these measurements is that there is a marked change in the c-axis resistivity, and hence interlayer coupling, with oxygen stoichiometry.

For the same samples for which we measured c-axis resistivity, we also performed angle-resolved photoemission measurements of the normal state.[4] The issue was to determine whether samples that exhibited the change in c-axis resistivity also exhibited any differences in their Fermi surfaces. We have reported elsewhere [6] that there is a finite interlayer coupling between the adjacent CuO$_2$ planes of the bilayer. We chose Pb-doped BSCCO-2212 samples. By performing careful TEM measurements,[4] we determined that the overdoped and underdoped samples were isostructural in the CuO$_2$ (ab-)planes. The only structural difference is a change in the periodicity of the superlattice modulation, with the overdoped samples exhibiting a larger period. We found that there is a change of symmetry in the normal state quasiparticle band states as the amount of oxygen is changed. Specifically, the electronic states that comprise the Fermi surface in the $k_x = \pm k_y$ directions exhibited a change in symmetry with oxygen stoichiometry, while the symmetry of the states in the $k_x$ and $k_y$ directions was unaffected by the oxygen stoichiometry. Our results indicate one of two possibilities. One is that the c-axis (interlayer coupling) affects the symmetry of the electronic states. The other possibility is that the electronic states arise due to many-body effects.[7] If the electronic states are interpreted as many-body states,[7] preliminary calculations indicate that interactions beyond the nearest-neighbor must be included to explain the data.[7]

In addition to the symmetry of the normal state electronic states, our data indicate two other important points about the Fermi surface. We find
that there is an extended van Hove singularity, consistent with other reports.\cite{8} The extent of the singularity is reduced for overdoped samples.\cite{4} A pocket, absent for underdoped samples, develops around the $(\pi,0)$ point for overdoped samples,\cite{4,8} consistent with earlier work by C. Olson and colleagues.\cite{12,13} Further, it is noteworthy that P. Aebi and colleagues have used our samples to study the presence and strength of the shadow bands they reported earlier.\cite{14} In their previous work,\cite{14} they reported observing shadow bands only on samples that were particularly flat and well-ordered (to reduce scattering that averages over the Brillouin zone). Using our samples, they find such shadow bands, and find that the shadow bands are weaker, but present, for the overdoped samples ($T_c = 75$ K) compared to the underdoped samples ($T_c = 80$ K).\cite{15} These data indicate that both the symmetry of the Fermi surface electronic states, and the topology of the Fermi surface, change with oxygen doping.

Note that we have found results consistent with other investigators for those measurements where consistency is expected. For that reason, our results as to the symmetry and topology of the Fermi surface electronic states appear representative of samples in different laboratories. As an additional check of our samples, we have studied the current versus voltage measurements for supercurrent along the c-axis and applied magnetic field in the ab-plane.\cite{16} The issue is whether the change in c-axis resistivity, and Fermi surface electronic states, both normal state properties, are related to superconducting state properties.

We found that for our samples, the CuO$_2$ planes are stacked very close to parallel, with an ab-plane misalignment below 0.02 degrees. This allowed us to orient the applied magnetic field parallel to the ab-plane with high accuracy. The results at lower magnetic fields (below 1 Tesla) reproduce earlier work by Kleiner et al.\cite{17,18} For underdoped samples, the data indicate a S-I-S Josephson junction stacking of planes along the c-axis. For overdoped samples, the critical current density increases between ($\times 100$ − $\times 1000$) and the stacking is S-N(S')-S. Our results indicate a marked increase in critical current density for overdoped samples, the same samples that exhibit the changes in normal state properties.

However, neither our results\cite{16} nor those of our colleagues,\cite{17,18} conclusively establish whether the c-axis coupling in overdoped samples is Josephson-junction or three dimensional. One new, and pertinent, result is that overdoped samples obey the Kim-Stevens relation,\cite{19} as do three-dimensional superconductors. We found $H_{c2}$ is already 14 Tesla only 4 K below $T_c$.\cite{16}

In magnetization studies of cuprate superconductors, much has been made of the “fishtail” behavior,\cite{20-22} a region of applied magnetic field for which the magnetization increases with increasing applied field. Using samples similar to those used in photoemission studies, X.Y. Cai et al.\cite{22} reported that the fishtail exists for both underdoped and overdoped samples, although such samples exhibit very different quantitative magnetization response. The data were interpreted as indicating that there are always stronger and weaker superconducting regions. These are the same sample types described above. Consequently, the fact that all samples exhibit the fishtail behavior indicates that all samples exhibit both stronger and weaker superconducting regions.

\begin{figure}
\centering
\includegraphics[width=\textwidth]{Fig.1.png}
\caption{Photoemission spectra in the normal state along the $k_x = k_y$ direction for He-annealed, Pb-doped sample (gap = 0.2 meV) and oxygen-annealed, overdoped sample (gap = 10-12 meV).\cite{27}}
\end{figure}
For the same sample types, we also conducted angle-resolved photoemission measurements of the size of the superconducting gap for two high-symmetry directions, $k_x$ (Cu-O-Cu bond axis in real space) and $k_x = k_y$ (Bi-O-Bi bond axis perpendicular to superlattice modulation).[23,24] Figure One illustrates the results. We found that for samples with less oxygen, particularly if underdoped, the gap along $k_x = k_y$ is indistinguishable from zero ($2 \pm 2$ meV), consistent with earlier reports by B.O. Wells and Z.-X. Shen.[25,26] However, for overdoped samples, the data indicate unambiguously that the superconducting gap in the $k_x = k_y$ direction is non-zero.[27] Observing a non-zero gap in the $k_x = k_y$ direction has also been reported earlier by several research groups.[28-31] One significant point is that this study is the first to directly connect the size of the superconducting gap and the bulk critical current and magnetization properties of a cuprate superconductor system.

These results can be regarded as solid, because they either reproduce earlier work or have been independently confirmed by other investigators, including other investigators using our samples.[15] The last experimental result that we wish to report is the temperature and momentum-resolved study of the superconducting gap for oxygen overdoped samples.[32] We studied the variation of the superconducting gap with temperature for two Brillouin zone directions, $k_x$ and $k_x = k_y$. Figure Two illustrates the results. We found that the temperature dependence of the superconducting gap along the two symmetry directions is qualitatively different. In particular, the gap along the $k_x = k_y$ direction becomes indistinguishable from zero at $0.82T_c$, while along $k_x$ the gap remains at 90-100% of its value at $0.35T_c$. Some parts of the data in Fig. 2 have been independently confirmed. The rapid, non-BCS, increase of the gap with decreasing temperature in the $k_x$ direction has been confirmed by J.C. Campuzano and colleagues.[33] The non-zero gap in the $k_x = k_y$ direction at lower temperatures has been reported earlier.[28-31]

What inferences can be drawn that are based strictly on data reproduced in different laboratories? I am confident of the following:

- Saying that the symmetry of the order parameter (gap) remains identical for all stoichiometry has been ruled out.[27-29,31-34]
- There is a marked increase in interlayer coupling for overdoped samples, as reflected in bulk measurements [1-4,16-18] and the symmetry of the normal state electronic states.[6]
- The shadow bands reported earlier by P. Aebi et.al. [14] are observed for samples from different laboratories, and exist well into the overdoped regime.[15]
- The gap increases more rapidly for temperatures below $T_c$ than a would a BCS superconductor.[32,33]

In addition, if all the results of of Ref. 32 are confirmed by independent work, these results indicate that, for overdoped samples, just below $T_c$ there may be only d-wave pairing, while at lower temperatures a more complicated pairing interaction exists. However, for underdoped samples, the small value of the gap along the $k_x = k_y$ direction (at $0.35T_c$) indicates that a predominant d-wave pairing extends to lower temperature.
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