April 1991

ANL/MSD/PP--73474

**OBSERVATION OF THE ELECTRON RIDGE FERMI SURFACE
IN $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ BY POSITRON ANNIHILATION***

L.C. Smedskjaer,¹ A. Bansil,² U. Welp,³ Y. Fang,^{1,+} and K.G. Bailey¹

¹Materials Science Division

³Science and Technology Center for Superconductivity
Argonne National Laboratory, Argonne, IL 60439

²Physics Department, Northeastern University, Boston, MA 02115

The submitted manuscript has been authored by a contractor of the U.S. Government under contract No. W-31-109-ENG-38. Accordingly, the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U.S. Government purposes.

RECEIVED
OCT 14 1994
OSTI

jmc

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

*Work supported by U.S. Department of Energy, BES-Materials Sciences under contract #W-31-109-ENG-38 (LS, YF, KB), DOE grant #DE-FG02-85ER45223 (AB), and the National Science Foundation Office of Science and Technology Centers under contract #STC-8809854 (UW).

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

12

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

Observation of the Electron Ridge Fermi surface in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$
by Positron Annihilation

L.C. Smedskjaer¹, A. Bansil³, U. Welp², Y. Fang¹⁺, and K.G. Bailey¹.

¹ Materials Science Division and ² Science and Technology Center for Superconductivity. Argonne National Laboratory, Argonne, IL 60439.

³ Physics Department, Northeastern University, Boston, Massachusetts 02115

Abstract

Positron annihilation (two-dimensional-angular-correlation) experiments on an untwinned single crystal of metallic $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ sample are reported in the c-projection. The measurements were carried out at room temperature and involved 94 Mcounts. An analysis of the spectra reveals clearly for the first time the presence of the electron ridge Fermi surface associated with the one-dimensional chain bands, and orthorhombic anisotropies in momentum density in good agreement with the band theory predictions.

PACS: 71.25H, 74.70, 78.70B

⁺ Present address: University of Illinois at Chicago, Chicago IL 60680.

Two-dimensional-angular-correlation of annihilation radiation (2D-ACAR) constitutes a powerful spectroscopy of the electronic structure and Fermi surface of materials. With this motivation, a considerable effort has been devoted in the last few years to studying high- T_c superconductors via this technique, with the hope of determining the extent to which electronic states in these compounds can be described within the framework of the conventional band theory picture. The answer to this question is important in order to establish the degree to which correlations are essential in describing electronic states, and bears thus on the eventual nature and form of the theory of superconducting mechanism of these materials. Much of the 2D-ACAR work, experimental as well as theoretical, has been focused to date on the classic high- T_c superconductor $\text{YBa}_2\text{Cu}_3\text{O}_x$ ¹⁻⁷⁾, with a few very recent studies of the Bi-2212 compound^{8,9)}.

All the existing measurements on $\text{YBa}_2\text{Cu}_3\text{O}_x$ have so far employed *twinned* single crystals¹⁻³⁾. In this article we report the first 2D-ACAR experiments on a nearly *untwinned* single crystal of metallic $\text{YBa}_2\text{Cu}_3\text{O}_x$. The advantages of using untwinned single crystals are clear. The untwinned sample is closer to the ideal $\text{YBa}_2\text{Cu}_3\text{O}_7$ compound which is the basis of much of the theoretical work on this system. The measured spectrum from a twinned specimen will possess the tetragonal C_{4v} symmetry in the c-projection, whereas the ideal orthorhombic system possesses the C_{2v} symmetry. Therefore,

important aspects of the momentum density, e. g. the ΓY - ΓX asymmetry, cannot be determined for the twinned system.

We focus in this article on identifying the Fermi surface associated with the highly anisotropic one-dimensional chain band. This is the only electron sheet predicted by band theory in $\text{YBa}_2\text{Cu}_3\text{O}_7$. While some Fermi surface sheets have been observed via very recent dHvA¹⁰⁾ and angle-resolved photoemission experiments¹¹⁾, so far the electron ridge Fermi surface has, to our knowledge, not been observed. Here the positron should have an advantage in that it is predicted to sample these electron states preferentially, providing thus an especially discriminating test of the band theory predictions.

On the theoretical side, it may be noted that following the first momentum density computations in $\text{YBa}_2\text{Cu}_3\text{O}_7$ by Bansil *et al*⁴⁾, this system has been studied by a number of authors^{1,5,6)}. The main features of the LDA-band-theory-based theoretical momentum density in $\text{YBa}_2\text{Cu}_3\text{O}_7$ relevant for the 2D-ACAR experiment are well established, including the characteristic signatures of the various FS sheets in the unfolded as well as the LCW-folded c -projected momentum density; the relatively smaller differences in details of momentum density in various computations arise from differences in the underlying band structures and Fermi surfaces, and are quite common in first principles band structure work even in simpler materials.

The present experiments were carried out at room temperature on a partially untwinned single crystal of approximate dimensions 1 mm by

2mm by 0.1mm. The superconducting transition temperature of the sample was measured with a SQUID to be 91K, with a transition width of 1K. While the precise degree of detwinning of our sample was difficult to estimate, an examination of the sample in an optical microscope showed the absence of twin boundaries, except in one of the outermost corners of the specimen. The sample was mounted on a 25 μm diameter wire. The background annihilations associated with the sample support is estimated to be about 5 - 10% of the total counts.

The 2D-ACAR measurements were carried out using Anger Cameras of nominally 500 mm diameter positioned 12 m from the sample. The momentum resolution of the instrument is estimated to be 0.4 to 0.5 mrad, including the effects of the finite sample size; since the experiment was carried out at room temperature, the effective total momentum resolution (FWHM) is about 0.7 mrad, which includes positron thermal smearing effects.

The c-axis of the crystal was oriented so as to be parallel to the axis of the camera, and thus the present experiment measured the two-dimensional c-projection of the momentum density. The c-axis of the crystal was aligned *in situ*, and the alignment was checked during the progress of the experiment. The deviation between the camera axis and the c-axis was found to be 0.4 and 0.0 degree in the ΓY and ΓX directions respectively. The orientation of the a-b plane of crystal was determined from an analysis of the data.

All data were collected in listmode, from which a histogram on a 0.39 by 0.39 mrad/pixel (1/16th of the Brillouin zone dimension) grid was obtained. The pixels were placed such that the symmetry axis coincided with the pixel boundary, rather than the pixel center, which removed the problem of statistical singularities along high symmetry directions. Two types of fields were collected, namely the coincidence field and the floodfields. The floodfields were used to calculate the efficiency matrix, which in turn was used to correct the coincidence data.

The total experiment was divided into four groups, each with about 25 Mcounts. Each group of these data was checked for symmetry and possible instrumental errors, yielding a final spectrum with a total of 94 Mcounts. The data presented in this article have been C_{2v} symmetrized. However, before symmetrization, it was verified that the data exhibit C_{2v} symmetry. A more detailed discussion of our symmetrization and related data analysis procedures, is given elsewhere¹²⁾.

In order to expose structure in the observed 2D ACAR data, a smooth isotropic distribution was subtracted from the data, and the result is shown in fig. 1. Three main features are visible in the figure: (i) a central ridge-like feature running along the ΓX direction, (ii) 4 mountain-like features surrounding the ridge in the ΓS directions, and (iii) two smaller side-ridges running parallel to the main ridge at approximately 12.6 mrad from the center. The present results

strongly contrast those from a twinned sample, where the four mountains are seen, but where the ridge appears to take the form of a blurred cross with a lower relative intensity³⁾.

Most relevant for interpreting our data are the calculations of Bansil *et al*⁴⁾ who have obtained the c-projected momentum density in $\text{YBa}_2\text{Cu}_3\text{O}_7$ within the band theory framework. A main theoretical prediction is the existence of a ridge-like Fermi surface (one-dimensional electron state) running along the ΓX direction, in accord with the results of fig. 1. A detailed comparison can be obtained from the directional anisotropies (here defined as differences in the projected momentum density along two different directions)¹³⁾. Figure 2 shows four such calculated anisotropic sections. The curves reflect the character of both the partially filled bands which give Fermi surface breaks, as well as the completely filled bands which yield more smooth variations in the momentum density, the spectrum being further modulated by the positron wave function effects. The signatures of the ridge Fermi surface are the break around r_0 in the first Brillouin zone (the break at the reflected point $-r_0$ not shown), and its first 'Umklapp' image (r_1, r_2) around 6.3 mrad, and the second image (r_3, r_4) around 12.6 mrad. [Recall that the Brillouin zone dimension is 6.3 mrad in $\text{YBa}_2\text{Cu}_3\text{O}_7$.] As noted, the ridge Fermi surface arises from a highly anisotropic one-dimensional band which is essentially full along ΓX but nearly empty along ΓY . The associated electronic states are predicted to couple strongly with the positron state in $\text{YBa}_2\text{Cu}_3\text{O}_7$ and yield clear features of the sort seen in fig. 2 in the momentum density.

Figure 2 shows other Fermi surface features, most notably the breaks denoted by (s_0, s_1) , and the umklapps (s'_0, s'_1) and (s_2, s_3) in the lowest panel, which are all related to a small hole Fermi surface around the symmetry point S. While the presence of this hole surface is predicted by most calculations, its detailed size and shape differs between various band computations. The larger hole sheets, due to the Cu-O planes, centered on the S-point do not couple well with the positron wave function and are not expected to give substantial signals in the 2D-momentum density.

Figure 3 compares theory and experiment. The theoretical results have been normalized using the same normalization factor for all four sections ¹⁴⁾. We see that the *relative* amplitudes of the theory and experiment in different sections are in good agreement; with the exception of the negative lobe in the $\Gamma S-\Gamma Y$ section both follow the same general behavior. These results are highly significant because, despite some differences in detail, the characteristic undulations in momentum density predicted by the LDA are more or less reproduced for the first time in measurements on a high- T_c superconductor. No such clear conclusion could obviously be reached from earlier positron studies which involved twinned samples, and therefore yielded the anisotropies $\Gamma Y-\Gamma X$ or $XS-YS$ to be strictly zero. Also, the $\Gamma S-\Gamma X$ and $\Gamma S-\Gamma Y$ anisotropies would be identical for a twinned sample.

It should be emphasized that the discrepancies of the level seen in Fig. 3 are not surprising in first principles comparisons and are commonly observed even in simpler materials. In the present case some of the

details of the band structure, especially around the symmetry points X, S, and Y, are quite sensitive and differ between various band calculations. There are also uncertainties inherent in the one-particle positron potential (e. g. the treatment of the electron-positron correlations). Bearing these considerations in mind, the overall agreement in Fig. 2 should be considered remarkably good.

We discuss now the detailed signature of the ridge Fermi surface. The rapid decrease or increase in the various sections of Fig. 3 around zero momentum is consistent with the break r_0 . This feature corresponds to the central ridge seen in fig. 1. The first Umklapp image (r_1, r_2) of the ridge around 6.3 mrad (hidden from view in fig. 1) predicted by the theory is seen at least as a shoulder in frame 3. The positive lobe seen in frame 3 corresponds to the sideridges seen in fig.1, and is mainly due to filled bands. The second image of the ridge coincides in part with the lobe and therefore theoretically yields a weak feature after resolution broadening around 13 mrad (r_4), but is responsible for the zero crossing seen in frame 2 at r_4 ¹⁵).

It is evident from Fig. 3 that there is much other structure in the data besides the ridge feature which has been the main focus of this article. A more detailed analysis of the data, including that of delicate differences between the band theory predictions of the Fermi surface around the symmetry points noted above and the measurements, is required for a convincing interpretation of the data in this regard. These details will be taken up elsewhere.

In summary, we have reported the first 2D-ACAR positron experiments on an untwinned single crystal of metallic $\text{YBa}_2\text{Cu}_3\text{O}_7$, and identified the presence of the electron ridge Fermi surface in good accord with the corresponding band theory predictions.

Work supported by the U.S. Department of Energy, Basic Energy Sciences, Division of Materials Sciences under contract #W-31-109-ENG-38 (LS, YF ,KB), and 85ER45223 (AB), and the National Science Foundation's Office of Science and Technology Centers under contract #STC-8809854 (UW). This work benefited from the allocation of supercomputer time on the NERSC and San Diego Supercomputer centers.

Figure Captions

Figure 1. A three dimensional rendition of the anisotropic spectrum obtained by subtracting a smooth background from the observed c-axis projected 2D-ACAR spectrum. Dispersion is 1.57 mrad/channel.

Figure 2. Theoretical difference sections for various sections indicated on the figure. r_0 , s_0 , etc denotes breaks associated with various Fermi surface features discussed in the text.

Figure 3. Comparison between the difference sections of Fig. 2 after resolution broadening (solid) with the corresponding experimental spectra.

References:

- 1) B. Barbiellini *et al* Phys. Rev. B43 7810 (1991)
L. Hoffman *et al* Europhys. Lett. 6 61 (1988).
- 2) H. Haghighi *et al* J. Phys. Cond. Matter 2, 1911 (1990)
- 3) L.C. Smedskjaer *et al* Physica C156, 269 (1988)
- 4) A. Bansil *et al*, Phys. Rev. Lett. 61 2480 (1988).
A. Bansil, P.E. Mijnarends, and L.C. Smedskjaer Physica C172
175 (1990). Phys. Rev. B 43, 3667(1991).
- 5) D. Singh *et al* Phys. Rev. B42, 2696 (1990)
D. Singh *et al* Phys. Rev. B39, 9667 (1989)
- 6) S. Massida, Physica C 169, 137 (1990)
- 7) H. Haghighi *et al* Bull. Am. Phys. Soc. 36 no 3, 376 (1991)
R.N. West *et al* Workshop on Fermiology of high-T_c
superconductors, Argonne National Laboratory, March 25-27
(1991)
- 8) L.P. Chan *et al* Bull. Am. Phys. Soc. 35 483 (1990)
- 9) P.E. Mijnarends *et al*, Physica C (to appear) (1991)
- 10) F.M. Mueller *et al* Bull. Am. Phys. Soc. 35 550 (1990)
C. M. Fowler *et al* Phys. Rev. Lett. (submitted) (1991)
G. Kido *et al* Bull. Am. Phys. Soc. 36 515 (1991)
- 11) C.G. Olsen *et al* Phys. Rev. B42, 381 (1990)
J.C. Campuzano *et al* Phys. Rev. Lett. 64, 2308 (1990)
- 12) L.C. Smedskjaer and D.G. Legnini, NIM A292, 487 (1990)
- 13) As emphasized in Ref. 4, it is most sensible to compare
computed and measured anisotropies because the anisotropic
spectra are less sensitive to possible effects of the 'background',
i. e. contributions from sample support, effects of oxygen
inhomogeneities, annihilations from trapped positrons, etc.
which are largely expected to be isotropic.
- 14) An absolute comparison between theory and experiment is
difficult due to the presence of 'background' contribution in the
experiment which is not included in the theoretical spectra.
- 15) It is gratifying that the present data and its interpretation is in
good accord with the recent independent 2 D ACAR study of
Ref 7.

Figure 1
Smedskjaer et al

Figure 2
Smedskjaer et al

Figure 3
Smedskjaer et al

Figure 1
Smedskjaer et al

Figure 2
Smedskjaer et al

Figure 3
Smedskjaer et al