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2D-ACAR SPECTRA OF INSULATING AND SUPERCONDUCTING Y-123

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ABSTRACT- An overview of the two-dimensional angular correlation (2D-ACAR) positron annihilation results for the three fundamental phases of $\text{YBa}_2\text{Cu}_3\text{O}_x$, namely, the normal metal, the superconductor, and the insulator, is presented. In addition to the c-axis projected momentum density, the recent results for the a-axis projection as well as the insulating Y123 are discussed. The experimental results are compared and contrasted with the corresponding band theory predictions as far as possible in order to gain insight into the electronic structure and Fermiology of this archetypal high- T_c superconductor.

Keywords: positron annihilation spectroscopy, superconductivity, Fermi surface.

I. INTRODUCTION

Positron annihilation (2D-ACAR) studies of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ started immediately after the discovery of this archetypal high temperature superconductor [1-3]. The 2D-ACAR experiment is ideally suited to answer one of the fundamental questions that arose early, namely, whether the electron momentum density in the new materials possesses Fermi surface breaks like the conventional metals. Results in this regard would provide guidance in searching for a suitable theoretical description of the electronic structure, and thus help in developing an understanding of the mechanism responsible for the occurrence of high superconducting transition temperatures. More generally, the accuracy of any specific electronic structure model can in principle be assessed by comparing the predicted 2D-ACAR spectra with the corresponding observations, although such comparisons to date have been possible only for the local density (LDA) based band theory model, which is the only model where detailed theoretical predictions are currently available.

Despite some unresolved issues, the recent Argonne Fermiology workshop [4-6] marked a turning point in that, by combining the results from 2D-ACAR [7-11], ARPES [12-15], and dHvA [16,17] experiments, a general consensus was achieved regarding the existence of a Fermi surface in at least $\text{YBa}_2\text{Cu}_3\text{O}_7$ in overall agreement with the band theory predictions. Concerning the 2D-ACAR spectroscopy in particular, extensive comparisons between the computed spectral intensities [18-22] and the measurements clearly establish the presence of the Cu-O chain related ridge Fermi surface in $\text{YBa}_2\text{Cu}_3\text{O}_7$; some evidence for other pieces of the Fermi surface is also indicated [11]. Further insight into the electronic structure of Y-123 will obviously be obtained by augmenting the studies of the metallic phase with those of the insulating phase $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ of this material [22-24].

Although the present work focuses on the Y123 system, we note that other high- T_c 's have been investigated via the 2D-ACAR spectroscopy [25-30]. Also, a large body of literature is devoted to Doppler broadening and lifetime studies of the high- T_c 's, for recent work in this area see, e.g., Ref. 31-33, and citations therein.

An outline of this article is as follows. The introductory remarks are followed in Section II by an overview of the band structure theory based methodology for computing the electron-positron momentum density involved in the analysis of the 2D-ACAR spectra. Section III gives a brief overall discussion of the temperature dependent 2D-ACAR spectra in various phases of Y123. The metallic

phase is considered in Section IV, including the c-axis projected momentum density (Section IV.1), the a-axis projection [34] (Section IV.2), and some remarks on the normal to superconducting transition. The insulating phase [35] and the metal/insulator transition are discussed in Section V, followed by a few concluding remarks.

II THEORETICAL OVERVIEW

The spectral quantity of relevance in connection with the 2D-ACAR experiment is the electron-positron momentum density $\rho_{2\gamma}(\mathbf{p})$, which can be expressed in the form

$$\rho_{2\gamma}(\mathbf{p}) = \frac{1}{\pi^2} \int d\mathbf{r} \int d\mathbf{r}' \exp[-i\mathbf{p} \cdot (\mathbf{r} - \mathbf{r}')] \int dE f(E) \int dE_+ f_+(E_+) \times \text{Im } G(\mathbf{r}, \mathbf{r}'; E) \text{Im } G_+(\mathbf{r}, \mathbf{r}'; E_+) \quad (1)$$

in terms of the electron and positron one-particle Green's functions $G(E)$ and $G_+(E)$, and the associated Fermi-Dirac distribution functions $f(E)$ and $f_+(E)$ respectively. Note that Eq. (1) assumes independent particles and implicitly neglects the effects of electron-positron correlation (see e.g. [21]). The 2D-ACAR experiment measures the 2D-projection of $\rho_{2\gamma}(\mathbf{p})$ along a specific direction in momentum space.

Our theoretical treatment is based on the application of multiple-scattering theory techniques to a system of non-overlapping muffin-tin potentials (including 'empty' spheres) to represent the crystal potential. The form of $\rho_{2\gamma}(\mathbf{p})$ for a general lattice with basis may be described most simply in terms of the formula for the electron momentum density $\rho(\mathbf{p})$ [36]:

$$\rho(\mathbf{p}) = -\frac{(4\pi)^2}{\tau} \sum_j \frac{f(E_j)}{(E_j - p^2)^2} \frac{\left| \sum_{\mu} \exp(-i\mathbf{p} \cdot \mathbf{b}_{\mu}) \sum_L \bar{C}_{L\mu}(\mathbf{k}) s_{l\mu}(p, E_j) \cot \eta_{l\mu} Y_L(\mathbf{p}) \right|^2}{\sum_{\mu\nu} \sum_{LL'} \bar{C}_{L\mu}(\mathbf{k}) \dot{M}_{LL'}^{\mu\nu} \bar{C}_{L'\nu}(\mathbf{k})} \quad (2)$$

Here, all vectors and matrices are in the space of angular momentum and basis atom indices L and μ , and are to be evaluated at the energies $E = E_{\mathbf{k}j}$ of the Bloch levels at the reduced wave vector $\mathbf{k}$. $Y_L(\mathbf{p})$ is the L th real spherical harmonic, τ the unit cell volume, $\mathbf{b}_{\mu}$ denotes the position, and $\eta_{l\mu}$ the l th phase-shift of the μ th atom. $s_{l\mu}(p, E)$ are smoothly varying functions involving the solutions of the radial Schrödinger equation for the μ th muffin-tin potential. The remaining quantities are the KKR matrix M and its eigenvectors $C_{L\mu}(\mathbf{k})$. They satisfy the secular equation $MC = 0$. The momentum density of the positron in its ground-state ($\mathbf{k}=0$) is given by a formula similar to Eq. (2), except that in this case there is no summation over j . $\rho_{2\gamma}(\mathbf{p})$ is then obtained straightforwardly as a convolution of the e^- and e^+ momentum density functions, see Ref. 36 for details.

III. POSITRON SPECTRA IN Y123; AN OVERVIEW

A typical as observed 2D-ACAR spectrum is shown in Fig. 1. Fig. 2 shows sections through the 2D-ACAR spectrum for both the metal ($x=6.9$) and the insulator ($x=6.2$) at low and high temperatures [35]. The electronic structure effects which are our primary concern here manifest themselves as structure in the 2D-ACAR spectra. Following common practice (see, e.g., [2]), such structure may be exposed by considering the anisotropic spectrum $A(\mathbf{p})$ defined by

$$A(\mathbf{p}) = M(\mathbf{p}) - S(\mathbf{p}) \quad (3)$$

where $M(\mathbf{p})$ denotes the measured 2D-ACAR and $S(\mathbf{p})$ is a smooth radially symmetric function. In the following, we discuss the nature of the electronic states, including the Fermi surface, in the metallic

and insulating phases of Y123 in terms of the anisotropic spectra $A(p)$ which turn out to be relatively temperature independent.

Some comments concerning the large temperature effects seen in Fig. 2 are appropriate however. We see that as we go from the metallic to the insulating state, the 2D-ACAR becomes narrower or more peaked near $p=0$ at both temperatures (all spectra are area normalized); the change at the metal/insulator transition is however about two times larger at 300K than at 30K near $p=0$. The temperature effect, observed in going from 30K to 300K, is similar for for the metal and the insulator, but the direction is opposite, i.e., in the metal the 30K spectrum is narrower whereas in the insulator the 300K spectrum is narrower.

Fig.1 2D-ACAR data for $\text{YBa}_2\text{Cu}_3\text{O}_{6.9}$ in c-axis projection (300K).

A variety of mechanisms are probably involved in the effects seen in Fig. 2. Since the positron annihilation experiment measures the electron-positron momentum density, the spectra reflect changes in the nature of *both* the positron and the electron states of the system (intrinsic effect), as well as possible changes in positron trapping (extrinsic effect) in the sample. While these questions are of interest in their own right, the key point to keep in mind for our present purpose is the aforementioned insensitivity of the anisotropic spectra $A(p)$ to temperature, suggesting that the effects of Fig. 2 are likely not intrinsic; see Ref. 11, 35 for a further discussion.

Fig. 2: Sections along the ΓX direction through the c-projected 2D-ACAR spectrum of $\text{YBa}_2\text{Cu}_3\text{O}_{6.2}$ at 35K and 300K (upper pair), and $\text{YBa}_2\text{Cu}_3\text{O}_{6.9}$ at 30K and 300K (lower pair). All spectra are area normalized.

IV THE METALLIC PHASE

IV.1 THE C-PROJECTION

The anisotropic distributions, shown in Fig. 3 at 30 and 300K, are rather similar in that these consist of a central ridge-like feature surrounded by four mountains. In addition, a less prominent sideridge along the ΓX direction is visible in both spectra at around 12 mrad; results in agreement with those of Fig. 3 (300K) have also been reported by Refs. 9 and 10.

The ridge feature and the associated umklapps (sideridges) are signatures of the one-dimensional Fermi surface, which is an electron sheet parallel to the a-c plane in the first zone, and originates from electron states in the chain region [37-39]. The four mountains, which exhibit C_{4v} symmetry, are most likely in part caused by electron states in the Cu-O planes, and in part by states in the chain area. These spectra do not show a clear imprint of the Cu-O

Fig. 3 A three-dimensional rendition of the anisotropic spectra $A(p)$ from untwinned $\text{YBa}_2\text{Cu}_3\text{O}_{6.9}$ in c-axis projection at 30 (left) and 300K (right).

plane Fermi surfaces; this is because the positron wave function overlap with the Cu-O plane electrons is small compared to the overlap with electrons in the chain region [18-22,33].

In comparing band theory predictions with experimental results (for twinned samples), it was noted early that the shapes of the computed and measured 2D-ACAR's differ substantially, the theoretical results being flatter near $p=0$ in comparison to the measurements; this discrepancy is seen from Fig. 4 to be present for the untwinned specimen as well. In analyzing twinned data, Ref. 19 found that if an isotropic 'background' is subtracted from the data, then the theory and experiment agree in many details, leading to the suggestion that the experimental spectra should be corrected for background before making such a comparison. The physical origin of the background is not clear, but it arises presumably from defects, oxygen inhomogeneities and other effects not included in the computations that assume a perfect crystal. In Ref. 11 the temperature effect is reexamined and it is shown how a more satisfactory background correction can be made in terms of the temperature dependent spectra.

Fig. 5 compares the background corrected spectrum with the normalized theoretical spectrum along the high symmetry directions in the first as well as higher Brillouin zones. A reference to Figs. 4 and 5 shows a dramatic improvement regarding the overall agreement between the theory and experiment. To enhance FS signatures, anisotropies -- in the form of difference sections for several pairs of directions -- are considered in Fig. 6 for the background corrected data. We comment on the theoretical results of Fig. 6(a) first. The complexity of these spectra arises not only from the presence of various FS sheets in $\text{YBa}_2\text{Cu}_3\text{O}_7$, but also from the detailed character of the electronic wave functions of various filled bands. Several of the 'breaks' associated with the Fermi surface are indicated (e.g., r_1 , r_2 , s_1 , ..., etc.). The sharpness of such breaks reflects the nearly dispersionless nature of the band structure of $\text{YBa}_2\text{Cu}_3\text{O}_7$ along the c-axis.

The signatures of various FS sheets in Fig. 6(a) are as follows. The ridge FS gives the rapid variation (r_0) near $p=0$; since the ridge lies along the ΓX direction, in the ΓS - ΓX spectrum or other difference spectra involving the Γ -point, the

Fig.4 Comparison between theory and the as observed sections through typical 2D-ACAR spectra for untwinned $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$. The results are shown for the 300K measurements.

Fig. 5 A comparison of various sections through the background corrected 2D-ACAR spectrum for the untwinned $\text{YBa}_2\text{Cu}_3\text{O}_{6.9}$ sample with the corresponding theoretical predictions of Ref.[19]. The results for the high symmetry lines ΓY , ΓX , ΓS , YS , and XS and parallel lines through higher zones are shown as marked.

anisotropy is nearly zero for momenta (around $p=0$) of the order of the width of the ridge. The first umklapp of the ridge (r_1, r_2) is seen clearly around 6.3 mrad as a peak in ΓY - ΓX , and a dip in ΓS - ΓY ; only weak features are seen at this momentum in the ΓS - ΓX or XS - YS curves. The second image of the ridge (r_3, r_4) occurs around 13 mrad. The high momentum end r_4 is generally more clearly visible than the low momentum feature r_3 ; r_4 in particular coincides with the zero crossing in ΓS - ΓY . The FS S-centered hole (the "pillbox") yields relatively prominent features only in the XS - YS spectrum, and the breaks s_1, s_2, s_3, s_4, s_5 , and s_6 are all associated with the pillbox. Finally, as expected, the Cu-O plane sheets give essentially no discernible signatures in the 2D-ACAR's; this is due to the weak coupling of the positron Bloch state with the Cu-O planes [18-22].

Fig. 6(b) shows that experimental anisotropies are in good accord with theory regarding the detailed spectral features; also the agreement concerning the absolute magnitude of the overall undulations is good. In contrast, uncorrected data -- although having much the same features as shown in Fig. 6(b) -- have undulations that are about a factor of two too small. The level of discrepancies seen in Fig. 6(b) is common in even simpler materials in first principles comparisons between theory and experiment.

As noted above, the ridge FS gives three distinct features: (1) rapid variation in anisotropy near $p=0$, (2) an image around 6.3 mrad, and (3) a second image around 13 mrad. The experimental spectra display all three of these features. Although we do not expect any 'breaks' in the experimental data due to

Fig. 6 (a): Theoretical anisotropic momentum distribution, here defined by taking differences between spectra along four different pairs of directions. Thin vertical lines mark the position of various FS features r_0 , s_1 , etc. discussed in the text. (b) A comparison between the resolution broadened theory curves in (a) with the corresponding experimental difference sections obtained from the corrected data. The spectra have been offset relative to one another as indicated by the horizontal lines.

resolution effects, near $p=0$, both sets of curves in Fig. 6(b) possess similarly rapid variations and slopes; the width of the experimental ridge would however appear to be somewhat larger than that implicit in our band structure. The 6.3 mrad image of the ridge (r_1 , r_2) is particularly important in the $\Gamma Y-\Gamma X$ curve because this feature lies on a relatively gently varying background and remains quite prominent in the theory even after including resolution broadening, and it is crucial that this feature is indeed observed clearly in the experimental data. By contrast, in the $\Gamma S-\Gamma Y$ curve, the resolution broadening effects would make the 6.3 mrad ridge image difficult to distinguish from the background.

IV.2 THE A-AXIS PROJECTION

The above considerations all pertained to the c-axis projection. We now briefly consider results for the a-axis projection, which provides another view of the momentum density. Some current theoretical views hold that there may be fundamental differences in the description of the electron states (e.g., localized vs. itinerant) in the a- or b- directions compared to the c-direction [40]. In this regard, because an a-projection does not involve a momentum density integration in the c-direction, it is more sensitive to the electronic wave function character along the c-direction than is the c-projection. Also, since the c-projected momentum densities in $YBa_2Cu_3O_7$ are in good agreement with band theory predictions, it would be interesting to establish whether the band theory continues to provide a reasonable description of the a-projected spectra. In Fig. 7 anisotropic data for a c-axis and an a-axis projection both measured

Fig. 7 A three-dimensional rendition of the *anisotropic* spectra $A(p)$ for untwinned $\text{YBa}_2\text{Cu}_3\text{O}_{6.9}$. (a): in a-projection, and (b): in c-projection. The pixel size is 0.8 by 0.8 mrad.

at 300K are compared [34]. We note that the present results for the a-axis projection appear similar to more recent results of Ref. 41.

Fig. 7 allows us to delineate similarities and differences between the signature of the Cu-O chain as revealed by the central ridge and its umklapp images in the two projections. The central ridge in both projections contains a minimum at $p=0$, and two peaks around ± 5 mrad, but in the a-projection, the anisotropic distribution extends to higher momenta and possesses additional peaks at ± 10 mrad. The 6 mrad first umklapp image of the ridge in the ΓY direction is clearer in the a-projection (ridge parallel to ΓZ); although hidden in the perspective view, this image is present also in the c-projection (ridge parallel to ΓX), but the mountains overlap with this feature, reducing the momentum range over which it can be observed. Finally, the second umklapp image of the ridge around 13 mrad is seen in both projections.

That the central ridge in both projections displays similar undulations in the ΓZ and ΓX directions may at first sight appear surprising, because in the a-projection the ridge extends along the c-direction, while in the c-projection it extends along the a-direction, and the BZ dimensions in the c- and a-directions are widely different. This insensitivity of the momentum density to the tripling of the unit cell in the c-direction indicates that the electron wave functions approximately behave as if the lattice dimension in the c-direction were 1/3 of the real lattice distance. The results of Fig. 7 are consistent with a picture where no dramatic change (e.g., itinerant vs. localized) takes place in the nature of the Cu-O chain wave functions between the a- and c-directions. In particular, our results suggest that the electronic states are not strongly localized along the c-direction, although a detailed comparison with theoretical calculations is needed to reach a firm conclusion.

IV.3 THE SUPERCONDUCTING TRANSITION

In order to investigate the transition from the normal to the superconducting state around 91K, we have also measured c-projected 2D-ACAR spectra for the untwinned single crystal of $\text{YBa}_2\text{Cu}_3\text{O}_{6.9}$ at 70K, 100K, and 185K. The spectra vary more or less uniformly with temperature and lie between the 30K and 300K results in a plot such as that of Fig. 2. A detailed comparison of the 70K and 100K data which bracket the superconducting transition does not reveal any substantial changes in either the 2D-ACAR or the anisotropic spectra $A(p)$, the latter appear similar to the data of Fig. 3. In this sense, the positron spectroscopy does not reveal significant changes in the electronic spectrum induced by pairing; we should keep in mind however that in Y123 the positron, as already noted, is more sensitive to the chain rather than the plane electrons.

Fig. 8 The anisotropic 2D-ACAR spectra, $A(\mathbf{p})$, for the insulator at 35K (left) and 300K (right).

V THE INSULATING PHASE; THE METAL-INSULATOR TRANSITION

Further insight into the electronic structure of Y-123 is obtained by augmenting the studies of the metallic phase with those of the insulating phase $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ [22-24, 35]. The close combination of band theory and experiment that has led to the understanding the 2D-ACAR spectra in the metallic Y-123 is less likely to succeed for the insulator, since the traditional band theory encounters well-known difficulties in describing the ground state of the insulating oxides. In the following we compare results from a c-axis projection of $\text{YBa}_2\text{Cu}_3\text{O}_{6.2}$ insulating sample measured at 35K and 300K to those of the metallic phase [35]. We note that our results appear similar to those reported at 40K in Ref. 22.

The anisotropic spectra of Fig. 8 and Fig. 3 allow us to delineate similarities and differences between the character of momentum density in the insulating and metallic phases of Y-123. A substantial overall change in the structure of the anisotropic 2D-ACAR spectra is seen to be induced by the metal/insulator transition at both the low and the high temperature. The most striking difference between the insulator and the metal is that of symmetry, the metal being orthorhombic and therefore resulting in a projection with C_{2v} symmetry, while the tetragonal insulator is C_{4v} symmetric. In the insulator the four mountain like features are present, but the central ridge and the side-ridges (see Fig. 3) are missing (see also [24]); a new feature connecting the four mountains in the shape of a 'cross' centered on $\mathbf{p}=0$ makes its appearance.

Some of the differences seen between Fig. 8 and Fig. 3 may be understood by recalling that the positron preferentially samples the chains in the O7 phase, and that the removal of O-atoms from the chains tends to localize the positron state in this area of the unit cell. We therefore expect that as the metallic, nearly O7, sample is deoxygenated and the Cu-O chains are disrupted, the signatures of the Cu-O chains in the 2D-ACAR will rapidly disappear due not only to the removal of the chains themselves but also due to a reduced overlap with the positron wave-function. These conclusions are consistent with the insulator spectra of Fig. 8, where we see the absence of the central ridge like structures extending along ΓX (or ΓY) directions, along with the absence of the side-ridges. The remaining major feature of the O7 2D-ACAR, namely the four mountains along ΓS , arise from various filled states including those associated with Cu-O planes and chains, and appear to be insensitive to the removal of chains. Finally, the new feature in the insulator, the aforementioned 'cross', appears to have no simple explanation in terms of the metallic 2D-ACAR.

The anisotropic spectra of Fig. 8 obviously contain what appear to be quite sharp structures. It turns out however that much of this structure lies close to Brillouin zone boundaries, and cannot therefore be taken to represent Fermi surfaces (see also [42]). Based on this and other arguments, Ref. [35] concludes that no Fermi surface signatures can be inferred in the 2D-ACAR spectra of insulating Y123.

Finally, we compare in Fig. 9 the anisotropy in the form of difference between the ΓS and ΓX sections for the insulator with the corresponding theoretical results of Ref. 20. While the use of a paramagnetic metallic band structure, implicit in the work of Ref. 20, to obtain electron-positron momentum density in O_6 , which is an antiferromagnetic insulator, is not satisfactory, the computation is nevertheless seen to capture some of the main features of the experimental data. The ΓS - ΓX anisotropy is seen to change dramatically at the metal/insulator transition; the metal contains (Fig. 6; ΓS - ΓX section) a broad negative lobe followed by a positive peak around 8 mrad, whereas the insulator gives two positive peaks centered around 4 and 8 mrad. Despite some discrepancies, the overall agreement between the theory and experiment in the insulator is surprisingly good, and as noted by Ref. 20, may reflect the lack of a significant overlap between the positron wave function and the Cu-O plane related bands which in the band theory incorrectly turn out to be metallic.

VI CONCLUSIONS

We have presented positron annihilation (2D-ACAR) results for the three fundamental phases of $YBa_2Cu_3O_{7-x}$, namely, the normal metal, superconductor, and the insulator. A clear signature of the Fermi surface associated with the one-dimensional Cu-O chain band is found in both the normal and the superconducting metallic state. The a-axis projected spectrum is consistent with a picture where no dramatic change (e.g., itinerant vs localized) takes place in the nature of at least the Cu-O chain wave functions between the a- and c-directions. No Fermi surfaces could be identified in the spectra from the insulator. Detailed comparisons are made between the measured 2D-ACAR's with the corresponding first-principles band theory predictions, and an impressive level of agreement is found in the metallic phase; surprisingly, the band theory appears to capture some major features of the observed spectra from the insulator, even though the band theory incorrectly predicts $YBa_2Cu_3O_6$ to be metallic.

Finally, we note that the 2D-ACAR spectra show substantial temperature effects in the metallic as well as the insulating samples, large changes are evident at the metal/insulator transition but not at the superconducting transition. However the important structure in the spectra, as revealed via the anisotropies in the 2D-ACAR which are the most relevant aspects of the spectra for delineating the electronic structure and Fermiology, are relatively insensitive to temperature. Our analysis suggests that the temperature effects are most likely extrinsic in nature (shallow positron traps, oxygen inhomogeneities, etc.), and do not reflect the character of the electronic states of the 'ideal' $Y123$.

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Fig. 9 The experimental difference section ΓS - ΓX in the insulator at 35K (triangles) and at 300K (circles). The corresponding theoretical results of Ref. 20 for $YBa_2Cu_3O_6$ (solid line) are scaled arbitrarily to obtain visual agreement with the experimental data.

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