c-AXIS PROJECTED ELECTRON-POSITRON MOMENTUM DENSITY
IN $\text{YBa}_2\text{Cu}_3\text{O}_7^*$

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ABSTRACT

We present the theoretical c-axis projected electron-positron momentum density $N_{2\gamma}(p_x, p_y)$ in $YBa_2Cu_3O_7$ based on the local density approximation (LDA) framework along various lines in momentum space. The calculations use the Korringa-Kohn-Rostoker (KKR) band structure formalism. The anisotropic distribution defined by taking cuts through the calculated spectra along different lines in the (p_x, p_y) plane possesses complex structures which arise from both Fermi surface effects and the anisotropy of the smoothly varying underlying background from filled bands; the maximum size of the anisotropy is about 10% of $N_{2\gamma}(0, 0)$. The theoretically predicted $N_{2\gamma}(p_x, p_y)$ distribution is compared with the measured 2D-ACAR spectrum. Our considerations suggest that in interpreting the 2D-ACAR data on $YBa_2Cu_3O_7$ in terms of a band theory LDA picture, a substantial, largely isotropic, background should be subtracted from both the 2D-ACAR's and the associated LCW-folded spectra.

I. Introduction

During the last two years considerable attention has been devoted to obtaining accurate theoretical predictions for the electron-positron momentum density in $YBa_2Cu_3O_7$. This effort is motivated by the fact that any serious interpretation of the positron annihilation angular correlation (2D-ACAR) measurements and identification of possible Fermi surface (FS) signatures in the data cannot be carried out without concurrent theoretical model predictions for the spectra. In this connection, Bansil et al.¹ reported the first c-projected momentum densities in the 123 using the KKR band theory approach; Ref. 2 extended these computations to obtain the corresponding LCW-folded³ distributions which are better suited for studying FS features. Singh et al.⁴ have computed LCW distributions within the LAPW band structure scheme. Massidda⁵ has reported 2D-ACAR as well as LCW distributions based on the LAPW method. Jarlborg et al.⁶ have carried out the computations using the LMTO approach. The main features of the band-theory-based electron-positron momentum density in $YBa_2Cu_3O_7$ as deduced by different workers using a variety of methods are in reasonable accord, the relatively small differences between the various band theory computations arise from differences in the details of the underlying band structures and Fermi surfaces. Finally, we note that while the preceding theoretical work is based on the assumption of extended Bloch states for describing the positron as well as the electron states in the solid, Turchi et al.⁷ have pursued an alternate LCAO-MO model to mimic localized states in describing the system. On the experimental side, 2D-ACAR data in $YBa_2Cu_3O_{7-\delta}$ have been published by Smedskjaer et al.⁸, the Geneva group⁹, and Haghighi et al.¹⁰. The overall shapes of the data from various laboratories using different samples are quite similar, and at least in this respect the experimental results can be considered well established. The question of whether or not the data show FS signatures, which are expected to appear as a relatively small modulation on the spectra, is not yet clearly settled and requires further experimental work.^{11,12}

In this article, we present detailed cuts through the theoretical c-projected electron-positron momentum distribution along various lines in the first as well as higher Brillouin zones in (p_x, p_y) space. The theory is confronted with the corresponding measurements on an absolute scale and points of agreement/disagreement are delineated. The anisotropies in the momentum distributions are discussed. We emphasize that we are not directly concerned in this article with the FS signatures in the data which as noted above are still a matter of controversy. The details of the KKR formalism and the specific muffin-tin potentials used in the present work are identical to those used in our earlier work, and are therefore not presented in the interest of brevity; see Refs. 1,2, and 13. In particular, the computations implicitly assume an independent particle model; electron-positron correlation effects are neglected.

II. Results and Discussion

Fig. 1 gives cuts through the c-projected theoretical $N_{2\gamma}(p_x, p_y)$ distribution along various lines in the first as well as the "Umklapp" higher Brillouin zones (BZ). Along the three principal symmetry directions, ΓX , ΓY , and ΓS , the distribution decreases by about 10% of its value at $p=0$ by 3 mrad, by about 50% by 6.5 mrad, and by 90% by momenta p between 9.8 and 11 mrad, as seen from curves a1, c1, and e1. The overall distribution is thus quite isotropic; it is roughly Gaussian, being flat in the first BZ, varying rapidly in the 2nd, 3rd, and 4th BZ's, and is nearly zero for momenta in the 5th and higher zones. This picture is consistent with the results for "Umklapp" lines presented in Fig. 1. For example, the curves a1 and a2 (or c1 and c2) possess a similar shape, with a2 extending to about 8.1 mrad which is somewhat less than the extent of 9.8 mrad for the curve a1 (a similar situation holds for c1 and c2 with slightly higher momentum values). The curves e1 and e2 on the other hand are quite different. These differences are easily understood if we envision drawing a circle of 11 mrad radius in Fig. 1(f) of momentum space; the distribution will then be seen to possess only a somewhat smaller extent along the line a2 compared to a1, but

will decrease more rapidly along e_2 compared to the line e_1 . Similar remarks are applicable in comparing curves b_1 and b_2 or d_1 and d_2 . The line a_3 (or c_3) essentially lies outside the distribution and the maximum value of momentum density along this line is an order of magnitude less than the peak value in the curves a_1 and a_2 (or c_1 and c_2).

The effects of the orthorhombic distortion are apparent in Fig. 1, as can be seen by comparing the curves a_1 and c_1 , or a_2 and c_2 , or b_1 and d_1 etc. In a tetragonal crystal there would be no difference in the momentum density along the ΓX and ΓY directions; in the present case differences arise not only due to changes in the smooth part of the distribution but also because there are no Fermi surface (FS) breaks along ΓX while there are many FS breaks along ΓY . Interestingly, although the curves e_2 and e_3 are similar in shape and magnitude, they are seen to differ in detail, once again as a result of the orthorhombic distortion; note that both curves involve the same set of reduced $\vec{k}$ -points (along the line ΓS) but one represents the $2\pi/a$ Umklapp while the other corresponds to the $2\pi/b$ Umklapp momentum density.

The theoretical results of Fig. 1 are in good overall accord with the corresponding experimental results in Fig. 2. We emphasize once again that although we show the data of Ref. 8 in Fig. 2, all the published 2D-ACAR measurements⁸⁻¹⁰ in this regard are in good agreement; the interpretation of the data in terms of the FS is not our direct concern in this article. In comparing Figs. 1 and 2, note that the experimental data cannot distinguish between the ΓX and ΓY directions because all measurements so far have been on twinned crystals; also, the experimental data in Fig. 2 has been corrected by subtracting a smooth isotropic 'background' curve for reasons discussed below. While a quantitative comparison between theory and experiment is taken up shortly, Figs. 1 and 2 already show agreement in many details, e.g. similarity of shapes of a_1 and a_2 , a_3 being an order of magnitude less than a_1 or a_2 , e_2 decaying much more rapidly than e_1 , etc.

Fig. 3 shows the anisotropy of the theoretical distribution. A sense of scale of the theoretically predicted anisotropy may be given by noting that in the top-most ΓS - ΓX curve the positive peak at 8.5 mrad is 7% and the negative peak at 5 mrad is 11% of the value of momentum density at $p=0$. The presence of quite sharp FS jumps is now more evident, although the FS structure is sitting on a smoothly varying anisotropic contribution of an amplitude generally comparable to the FS jumps. The anisotropies are vanishingly small beyond 16 mrad. The smoothly varying part of the anisotropy in Fig. 3 is characteristic of the details of the electron-positron matrix elements associated with the filled states in $YBa_2Cu_3O_7$. For example, even though there are no unfilled bands along the ΓX direction, the momentum density along ΓS (with a smaller number of filled bands) is larger for $p > 7$ mrad and produces the peak around 8.5 mrad (top-most curve). Similarly, the ΓY - ΓX curve displays a positive peak around 10.5 mrad. Massidda⁵ has recently computed the ΓS - ΓX anisotropy using the LAPW scheme; the results of Ref. 5 are similar to ours, both calculations showing in particular a dip followed by a peak with the curve crossing zero around 7 mrad.

A comparison between the theoretical and experimental anisotropies is made in Fig. 4. For this purpose, the ΓX and ΓY curves of Fig. 1 have been averaged and resolution broadened. The units chosen in Fig. 4 allow an absolute comparison between theory and experiment; also in these units the curves will be independent of the total number of counts in the spectrum as well as the pixel size in the data. Note that many of the published anisotropic curves in the literature are defined by subtracting a smooth background from the measured curves and should not be compared directly with Fig. 4 which involves a *difference* in the momentum density along two directions and is thus independent of the presence of an isotropic background in the data. Also, the isotropic part of positron-electron correlation effects cancels in these difference curves.

While both curves in Fig. 4 show a peak of essentially the same height at 8.5 mrad, and a dip at 11 mrad, they also possess significant differences. For example, the theoretical peak at 8.5 mrad is narrower, and the pronounced dip

in the theory at 6 mrad is absent in the measurements; this dip will be reduced in size if the 'ridge' electron FS sheet is absent in the real material as proposed in Ref. 2. The discrepancy between theory and experiment may have many contributing factors. These include: the lack of a proper treatment of electron-electron and electron-positron correlations (in particular the anisotropic part of these), differences between the theoretical and experimental band structures, the presence of defects, and as suggested by Massidda⁵ an admixture of O6 in the samples. Further computations of the momentum density associated with defect structures, and more accurate measurements on well-characterized samples are necessary to discriminate between the various possibilities.

Figs. 5-7 consider an absolute comparison between the theoretical and experimental 2D-ACAR curves along the ΓS and Γ -(XY) directions. Figs. 5 and 6 show clearly that the experimental curve (triangles) is significantly higher than the theoretical curve in the first BZ; the two are in reasonable accord for higher momenta as noted previously in Ref. 1 in considering the momentum density along the XS and YS directions. In order to shed some light on this discrepancy, Figs. 5 and 6 also show an isotropic 'background' curve (circles). The precise choice of this background function is not important for the main point we want to make here; the particular curve shown is in fact the measured 2D-ACAR from the glue used to bind the 123 single crystal to the wire in the sample holder. We have made a simple least squares fit between the experimental distribution and a linear function of the form $\alpha N_{2\gamma}(\text{theory}) + \beta N_{2\gamma}(\text{background})$ where α and β are constants. Using both directions ΓS and Γ -(XY) in the fitting procedure, the coefficient β was found to be 0.36. The experimental curves (squares) are obtained by subtracting $\beta N_{2\gamma}(\text{background})$ from the measured 2D-ACAR spectrum; the major discrepancy in the first zone is now seen to virtually disappear in Figs. 5 and 6.¹⁴

The residual differences between theory and the background corrected experimental spectra are given in Fig. 7; the subtraction of the background is seen to reduce the discrepancies generally by an order of magnitude. The residuals in

Fig. 7 could be attributed to various mechanisms indicated in connection with Fig. 4 above. The fit between theory and experiment can probably be improved significantly by adjusting the shape of the 'background' function. We did not however make such an attempt. Our main purpose is to emphasize that, should one attempt to interpret experimental spectra via LDA computations in the 123, it is essential to subtract at least a substantial isotropic background before a meaningful comparison can be made. Furthermore, since this background function is quite peaked in the first BZ, the measured 2D-ACAR (without such a subtraction) will yield a much larger variation upon LCW folding of the data than predicted by the LDA, and is likely to make it harder to identify possible FS signatures in the data. We stress that the preceding remarks are not necessarily meant to favor an LDA interpretation of the data, but merely to point out that it is inconsistent to compare measured and computed band-theory-based LCW amplitudes if the measured and calculated 2D-ACAR's possess a large discrepancy.

It is reasonable to ask: what is the physical origin of a possible background contribution in the measured 2D-ACAR's? Obvious mechanisms that come to mind are the annihilations in the sample support, presence of defects and inhomogeneities in oxygen concentration, and an admixture of the insulating phase in the sample. The fact that 2D-ACAR measurements from various laboratories with different metallic samples are in essential overall accord with respect to the data shown in Figs. 5 and 6, would favor the view that the origin of the background is intrinsic (i.e. defects etc.) rather than extrinsic (i.e. glue, etc.) in nature. Differences between theory and experiment (before subtracting a background) of the magnitude seen in Figs. 5 and 6 cannot likely be explained by variations in band structures and other uncertainties in the theory concerning the treatment of electron-electron and electron-positron interactions.

III. Conclusions

We present and discuss the theoretical c-axis projected electron-positron mo-

momentum density $N_{2\gamma}(p_x, p_y)$ in $YBa_2Cu_3O_7$ based on the LDA band theory framework along various lines in momentum space. The theoretical distribution is of a roughly Gaussian shape extending to about 11 mrad. The theoretical results are confronted with the corresponding 2D-ACAR measurements on an absolute scale, and the latter are found to contain a pronounced narrow peak near $p=0$ in the first BZ. This discrepancy between theory and experiment essentially exists in all available 2D-ACAR measurements¹⁵ on 123 metallic samples, the controversies surrounding the FS signatures in the data notwithstanding. We show that a good overall agreement between the calculated and measured 2D-ACAR's is possible provided an isotropic 'background' contribution is subtracted from the experimental spectra; the remaining differences could be rationalized on the basis of detailed effects not included in the computations and the sample quality. Our considerations imply that in order to interpret FS signatures in the 2D-ACAR measurements on the 123 via LCW-folding of the data, a significant isotropic contribution should be subtracted. The anisotropic theoretical distributions defined by taking differences in the spectra along different lines possess complex structure with dips and peaks which arise partly from Fermi surfaces and partly from the smoothly varying underlying background from filled bands; the maximum size of the anisotropy is on the order of 10% of the peak value $N_{2\gamma}(0, 0)$. In comparing the theoretical and experimental anisotropies some similarities are found, but there also are significant differences.

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14. It is seen from Figs. 2, 5 and, 6 that the corrected experimental spectra possess a small dip at $p=0$. Although this may be a physical effect, we should keep in mind the fact that this dip is not present in the uncorrected data in Figs. 5 and 6, and may thus result from an overcorrection in estimating the background contribution.
15. See Refs. 9 and 5 for the Geneva data. Also, the contour level diagram in Fig. 2 of Ref. 10 seems to be consistent with a distribution rather like that of Refs. 8 and 9.

Figure Captions

Fig. 1. Theoretical c-projected electron positron momentum density $N_{2\gamma}(p_x, p_y)$ in $YBa_2Cu_3O_7$ along various lines in (p_x, p_y) -space in arbitrary units. The curves are not broadened with experimental resolution in order to show the Fermi surface breaks clearly. The curve a1 corresponds to the line a1 in Fig. 1(f), other curves are similarly keyed. Filled circles in (f) designate the origins of the various curves in (a)-(d); note that (f) is not drawn to scale. The horizontal scale is in milliradians (1 mrad = 10^{-3} mc).

Fig. 2. Experimental 2D-ACAR's (c-projection) along various lines in momentum space in units of counts/annihilation/mrad²; this unit is independent of the total number of counts in the spectrum or the pixel size in the measurements. The data have been corrected by subtracting an isotropic background (see text). Various curves are keyed to the box on the lower right hand side.

Fig. 3. Theoretical anisotropic momentum distribution. Results for the differences $\Gamma S - \Gamma X$, $\Gamma S - \Gamma Y$, and $\Gamma Y - \Gamma X$ are shown.

Fig. 4. Theoretical and experimental anisotropy curves $\Gamma S - \Gamma(XY)$. The vertical scale is as in Fig. 2 and allows an absolute comparison between the theory and experiment.

Fig. 5. The 'background' isotropic spectrum (circles), the experimental spectrum (triangles), the experimental spectrum after subtracting an appropriately weighted 'background' spectrum (squares, see text), and the theory curve broadened to reflect experimental resolution along the ΓS direction (solid). All spectra are given in units of counts/annihilation/mrad² to allow an absolute comparison.

Fig. 6. Same as Fig. 5, except that this figure refers to the $\Gamma - (XY)$ direction. The theoretical curve is the resolution broadened average of ΓX and ΓY curves.

Fig. 7. Residual difference between the theory and experiment after background correction, i.e. differences between the squares and solid lines in Figs. 5 and 6.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7