RESOLVING THE FERMI SURFACE OF YBa\textsubscript{2}Cu\textsubscript{3}O\textsubscript{7-\delta} FROMPOSITRON ANNIHILATION DATA

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Abstract. A short survey is made of a recent research project devoted to reliable identification of the sheets of Fermi surface originating in the CuO chains (the ridge) in YBa\textsubscript{2}Cu\textsubscript{3}O\textsubscript{7-\delta} from 3D positron annihilation spectra. It is concluded that the ridge signature at third Umklapp components is the first one entirely attributable to the conduction electrons.

Key words: Fermi surface, high-\(T\textsubscript{c}\) superconductivity, YBa\textsubscript{2}Cu\textsubscript{3}O\textsubscript{7-\delta}, positron annihilation.

1. INTRODUCTION

The electronic properties of the normal and superconductor states of conventional superconductors are described in the frame of a Fermi liquid model. Fermi-liquidness behaviour of electrons within a perfect crystal is characterized by a number of characteristic features [1]: the occurrence of a sharp Fermi surface; low energy excitations consisting of quasiparticles whose properties (sharply peaked spectral density, unit charge, spin one-half) are well understood; characteristic properties in the limit \(T \rightarrow 0\), \(\omega \rightarrow 0\), such as a constant spin susceptibility and a linear-in-\(T\) heat capacity.

Whether the high temperature superconductors (HTS) are consistent with Fermi liquid behaviour or not is a question of highest interest. There is no answer in general. The experience accumulated at a given moment on a compound of interest can converge towards a possibly positive answer, or a certain property can be identified which is inconsistent with Fermi liquid behaviour. Among the HTS, most investigations were devoted to the archetypal compound YBa\textsubscript{2}Cu\textsubscript{3}O\textsubscript{7-\delta}, with increasing evidence which accommodate a Fermi liquid behaviour [1].

The present paper brings further confirmation to this point of view. In what follows, results of a recent research project are discussed. The project was devoted to the achievement of better resolution of the Fermi
surface signature in YBa$_2$Cu$_3$O$_{7-8}$ by means of the technique of two-dimensional angular correlation of the positron-electron annihilation radiation (2D-ACAR).

Previous efforts and difficulties of 2D-ACAR in HTS are summarized in section 2. Characterization of a window least squares method for off-line processing of 2D-ACAR statistics is given in section 3. New evidence of Fermi surface in YBa$_2$Cu$_3$O$_{7-8}$ is summarized in section 4. Both the method [2, 3] and the results [2, 4, 5] have been described in detail elsewhere. Here we add a few ideas stressing their physical meaning.

2. DIFFICULTIES AND PITFALLS OF 2D-ACAR IN HTS

Before their annihilation on electrons, the projectiles in a positron beam directed on a crystal loose most of their kinetic energies via interaction with the lattice phonons and electrons. The gamma rays representing the annihilation radiation emerging from a positron-electron pair carry away the energy and momentum of the $e^+ - e^-$ pair. Two planar detectors, parallel to a crystal plane of interest, can infer from 2D angular correlations of a pair of gamma rays the transverse (in-plane) component of the $e^+ - e^-$ momentum, which is stored on a memory device. Accumulation of annihilation radiation events yields a 2D-ACAR statistics which eventually resolves a 2D projection of the $e^+ - e^-$ momentum distribution onto the crystal plane. The classical theory and interpretation of the 2D-ACAR, [6], assumes that the positron momentum is negligible and hence the accumulated statistics represents a 2D projection of the electron momentum distribution inside the crystal. The Fermi surface of the specimen can then be inferred from characteristic discontinuities in the momentum distribution.

The 2D-ACAR shows unique advantages over other methods of resolving the Fermi surface: low sensitivity to sample surface effects, independence of resolution on the electron mean free path, accumulation of data on the whole topology of the Fermi surface in a single spectrum measurement.

The attempts at straightforward extension of the previously accumulated experience on the 2D-ACAR to HTS did, however, encounter many difficulties and pitfalls which were only gradually realized and solved.

The low fraction of the conduction electrons within the electron population inside an HTS (below 3 percent) makes the 2D-ACAR very demanding: accumulation of gigacount statistics is required to get the topology of the Fermi surface well resolved.

The interpretation of the quickly varying momentum distributions in HTS as Fermi surface jumps is a delicate point.

On one side, the electron momentum distribution in such complex materials is highly structured, the structure arising both from completely and partially filled bands. Due to the convolution with the finite resolution of the setup, the difference between an actual jump and a finite slope becomes hardly noticeable and needs a lot of expertise to reach the correct solution [7 – 10]. In fact, as shown elsewhere [11], early interpre-
tation of such quickly variable momentum distributions as Fermi surface breaks was misleading.

On the other side, preferential positron accumulation at particular regions inside an HTS results in the failure of the standard assumption of quasifree thermalized positrons before annihilation. Positron-electron correlation effects (occurrence of a polarization cloud surrounding the positron [7]) come into play; the identification of the \( e^+ - e^- \) pair momentum distribution with the electron momentum distribution becomes doubtful. A number of structures suggesting occurrence of Fermi surface breaks are shown to represent, in fact, positron effects (see, e.g., [12]).

A further specific complication of the 2D-ACAR spectra comes from the occurrence of the so-called "high-momentum components" [6]. Let us consider an electron of momentum \( \mathbf{k} \) which contributes to the 2D-ACAR spectrum. Due to the fact that the electron is embedded into the crystal, its contribution will be noticed not only at the momentum \( \mathbf{p} = \mathbf{k} \), but at all momenta \( \mathbf{p} = \mathbf{k} + \mathbf{G} \), where \( \mathbf{G} \) is a translation vector of the reciprocal lattice of the crystal. If \( g_1, g_2, \) and \( g_3 \) are the primitive translation vectors, every vector \( \mathbf{G} \) can be written, \( \mathbf{G} = m_1 g_1 + m_2 g_2 + m_3 g_3 \). In what follows, these "Umklapp annihilations" occurring at \( m_1 = m_2 = m_3 = m \), where \( m \) is a positive integer, will be called the \( m \)-th Umklapp components of the 2D-ACAR spectrum. Since the amplitudes of the \( m \)-th Umklapp components decrease with \( m \) as the coefficients of a Fourier series, only a few, statistics dependent, values of \( m \) will bring noticeable contributions to the spectrum.

To increase the height of the useful statistical signal, Lekh Crisp and West proposed [13] the folding of the 2D-ACAR spectrum from the extended to the reduced zone scheme. In view of the above-mentioned difficulties associated with the interpretation of the 2D-ACAR spectrum, a serious question mark rises on the reliability of the LCW folding. Thus, an extended zone analysis at higher order periodicity zones, based on explicit or implicit use of the periodicity cells mapped by the reciprocal lattice of the crystal onto the histogram plane, is to be foreseen.

Within this intricate process emerging towards correct identification of the Fermi surface breaks, the information brought by theoretical models of the electron energy bands in HTS and of the \( e^+ - e^- \) pair momentum distribution was very important. From the point of view of the electron energy bands in HTS, it resulted in the identification of Fermi surface sheets expected to occur in a given compound. On this basis, the 2D projections of the theoretically predicted Fermi surface breaks (as obtained after convolution with the resolution of the setup) provided important hints on the \( a \) priori expectation of achieved good or poor resolution of a particular sheet of Fermi surface by 2D-ACAR. Identification, within the smoothed 2D-ACAR spectra, of the Fermi surface breaks primarily attributable to electrons (and elimination of breaks carrying overwhelming positron-induced contribution) rested entirely with theory.

As a feedback of this process, the better agreement of the 2D-ACAR data with the outputs of the FLAPW theoretical approach [14] to the above-mentioned problems singled it out as the most realistic theoretical representation of the energy band structures of HTS in the local density approximation.
A good confirmation of the fact that the methods of off-line analysis of 2D-ACAR spectra [2] have ripened is provided by the identification of a new, characteristic, Fermi surface break of the CuO chain related electron ridge in YBa_{2}Cu_{3}O_{7-δ} at third Umklapp components. This was first found from the analysis of experimental data [2] and afterwards confirmed [4] by FLAPW theoretical calculations, as well as by an alternative method of analysis of 2D-ACAR spectra [6].

A survey of the investigation of YBa_{2}Cu_{3}O_{7-δ} by 2D-ACAR is highly illustrative for the progressive articulation of the relationship between theory and experiment until the reliable identification of a sheet of Fermi surface in the data.

Band structure calculations of YBa_{2}Cu_{3}O_{7} [15, 16] predicted the occurrence of a Fermi surface consisting of several sheets. The electron ridge along the ΓX direction and the small hole pillbox around the S point were found to originate in the CuO chains, while the large hole barrel centred at S and the hole arm around Y, in the CuO planes.

Simulation of the convolution of the predicted Fermi surface jumps with the finite resolution of the setup suggested [17] a good resolution by 2D-ACAR of the jumps associated to the one-dimensional ridge and mediocre to poor resolutions of the jumps associated to the other, two-dimensional, Fermi surface sheets. Thus, the efforts of the 2D-ACAR practitioners concentrated towards the resolution of the ridge from their data.

At 0 < δ < 0.2, YBa_{2}Cu_{3}O_{7-δ} is superconducting, with a Tc above 77 K and a width of the transition directly related to the quality of the sample. The crystalline structure is orthorhombic, with a very small difference between the magnitudes of the a and b primitive vectors of the elementary cell (a = 3.3163 Å, b = 3.8860 Å, c = 11.6830 Å). At δ = 1, one gets an insulator, YBa_{2}Cu_{3}O_{4}, characterised by absence of the CuO chains and tetragonal symmetry of the single crystal structure.

Due to the small difference between the lattice constants along the a and b directions, the YBa_{2}Cu_{3}O_{7-δ} crystals grown using a flux technique are twinned. That is, their structure along the c-axis (the growth axis) consists of successive layered regions showing the typical a and b structural details interchanged with each other (i.e., rotated by 90° from layer to layer). (Here, layer is not to be understood in the sense of atomic layer, its thickness extends over a great many elementary atomic layers). As a consequence, a 2D-ACAR spectrum projected onto the crystal a − b plane would show 4mm symmetry instead of the 2mm symmetry characteristic to the orthorhombic single crystal. The similarity of early resolved jumps in the 2D-ACAR spectra on twinned metallic YBa_{2}Cu_{3}O_{7-δ} and insulating YBa_{2}Cu_{3}O_{4+δ}, (δ < 0.2), compounds, respectively, raised doubts on their interpretation as Fermi surface jumps [11].

As a consequence, the main efforts of the 2D-ACAR teams engaged in the study of YBa_{2}Cu_{3}O_{7-δ} were directed towards the solution of the difficult task of producing untwinned single crystals and then towards the accumulation of high enough statistics. As a result of such effort, by the beginning of 1991, the Texas-Livermore group [18, 19] reported clear resolution of Fermi surface breaks, attributable to the electron ridge, at second Umklapp components. The result was immediately confirmed by the Argonne-Northeastern group [20, 21] and then by the Geneva group.
The last group also reliably identified signatures of the second Umklapp component of the Fermi surface ridge in 2D-ACAR spectra collected on twinned YBa$_2$Cu$_3$O$_{7-\delta}$ crystals [23, 12].

3. PIECEWISE CONTINUOUS WINDOW LEAST SQUARES SMOOTHING

In a recent effort towards improvement of the quality of the off-line analysis of the 2D-ACAR spectra, a method was derived [2], with a twofold purpose: (a) to perform statistical noise smoothing by a simple and efficient algorithm and (b) to secure independent evaluation of two basic parameters: occurrence or absence of twinning in the investigated crystal(s) and the accuracy of the crystal-detector tuning (consisting of two operations: parallel alignment of the wished crystal plane with the histogram (detector defined) plane and orientation of the symmetry lines of the 2D-ACAR spectrum along the detector coordinate system with respect to which the information is sampled into pixels).

To solve point (a), a moving window least squares (WLS) method was developed. A necessary condition for its success was the consistent incorporation of a number of underlying features of the 2D-ACAR spectra: occurrence of crystal generated symmetry elements (symmetry inversion centre, symmetry lines) of the 2D projection of the momentum distribution; splitting of the momentum distribution into Umklapp components, distributed over the histogram area following the translation periodicity of the reciprocal lattice of the crystal, with amplitudes which decay away from the projection of the first Brillouin zone; discretization into pixels, in a chessboard pattern, of the accumulated information, which blurs the expected symmetry properties; occurrence of measurements of different precisions (obeying a Poisson distribution law) at different pixels within the histogram plane. The statistical noise smoothing had therefore to be able to restore accurately the noise-free information from an arbitrary discretization into pixels.

To cope both with the discretization and symmetry in a simple way, the WLS method was conceived to be piecewise continuous: each pixel of interest was approached independently, but smoothed information was computed at crystal symmetry defined coordinates inside it.

Independent pixel approach was achieved by defining, around each pixel of interest, a local window of quasi-circular shape, consisting of entire pixels and centred at that pixel. The smoothing window moves therefore with the pixel in the histogram plane. Within such a window, the assumption of measurements of equal precisions (resulting in the assignment of constant weights to the data at the pixels) proved to be a good working hypothesis which resulted in a highly simplified, constant weight smoothing WLS formula (OW-WLSP).

To get information recovery at arbitrary coordinates inside a pixel, the approximating space of smoothed data embedded into the noise contaminated input 2D-ACAR data was chosen to be spanned by continuous polynomial basis functions of two variables up to 3rd degree, orthogonal to each other over the area of the (information dependent) smoothing window.
A validation study of CW-WLSF [3], done on suitably chosen case studies, investigated: output reliability limitations originating in signal distortion caused by CW-WLSF itself; the capacity of CW-WLSF to remove inner signal symmetry distortions induced by inappropriate information sampling (discretization) into pixels; the efficiency of statistical noise smoothing; the consistency of CW-WLSF with the underlying features of the 2D-ACAR data. The obtained results allowed definition of precise reliability bounds of the CW-WLSF smoothing of 2D-ACAR spectra, able to avoid artifacts coming from analysis.

Since the collected 2D-ACAR spectrum on untwinned YBa$_2$Cu$_3$O$_{7-δ}$ single crystals was projected onto the $a - b$ crystal plane, to get the small anisotropic part showing 2 mm symmetry, elimination of the overwhelming background of 4 mm symmetry was necessary. This was achieved by the simplest possible method, which consisted in the subtraction of the 90° rotated smoothed spectrum from the as-got, properly centred, smoothed one.

4. OUTPUTS OF OFF-LINE PROCESSING OF 2D-ACAR SPECTRA

The off-line analysis of 2D-ACAR spectra performed in [2] involved eight steps:

(i) removal of detector induced systematic errors stemming from finite angular aperture and local variations of the efficiency of gamma-ray recording;

(ii) rough location (to half-pixel accuracy) of the symmetry centre (the $Γ$ point) of the histogram (which corresponds to the zero-momentum projection of the $e^+ + e^-$ pair momentum distribution onto the histogram plane);

(iii) location of the $Γ$ point to working accuracy (fine location);

(iv) assessment of crystal quality;

(v) CW-WLSF data smoothing at fractionary pixel coordinates, determined by the fine location of the $Γ$ point inside the central pixel, and pixel relabeling so as to get the centre of the smoothed spectrum at $(0, 0)$ pixel coordinates;

(vi) assessment of the correctness of the identification of the histogram plane with the $a - b$ crystal plane and of the detector related coordinate axes with the symmetry lines of the 2D-ACAR spectrum;

(vii) subtraction of the 4 mm symmetry background;

(viii) cut of convenient slices in the obtained 2 mm anisotropy spectrum such as to put into evidence one-dimensional structures at various Umklapp components.

While complete description of these points is given in reference [2], the following additions are helpful.

The solution of point (ii) is based on the definition of a chi-square sum which measures, in $L_2$-norm, the goodness of acceptance of the centre
of a given pixel for the Γ point of the physical histogram. It is to be stressed that the definition used in [2] is optimized in the sense that it involves all the symmetry elements expected to occur in the theoretical projection of the electron momentum distribution onto the histogram plane. (For the $a - b$ projection of interest of the momentum distribution inside $\text{YBa}_2\text{Cu}_3\text{O}_7$, it includes, besides a term originating in the symmetry inversion centre, contributions following from the two orthogonal to each other symmetry lines as well.)

The correctness of the solution devised in [2] to point (iii) is obviously conditioned by affirmative answers to the identifications specified at point (vi). In the meantime, a solution overcoming this limitation was devised, with the consequence that the systematic errors related to misfits in the tuning mentioned at point (vi) can be eliminated from the 2D-ACAR spectra during their off-line analysis. Details will be reported elsewhere.

Fortunately, for the 2D-ACAR spectrum considered in references [2] and [4], such systematic errors were small.

The argument is as follows. To solve problem (vi), recourse is made to chi-square sums similar to those mentioned at point (ii), with smoothed unsymmetrized data (v) for input. Nine chi-square sums are computed, at the (0, 0)-th pixel (the Γ point) and its eight nearest neighboring pixels, respectively, resulting in a $3 \times 3$ "reference" matrix, corresponding to this $3 \times 3$ pixel set centered at Λ. The pixel manifolds which bring contributions to such chi-square sums are chosen to consist of regions $D(m)$ of the histogram, over areas equivalent to that covered by the $e^+ - e^-$ annihilation processes up to the $m$-th Umklapp component (the $m$-th region is thus an area similar to the projection of the first Brillouin zone onto the histogram plane, with a scale factor of $2m + 1$). For the $401 \times 401$ pixel available statistics of pixel side $\omega_p = 0.15$ mrad, regions $D(m)$ corresponding to $m \in \{0, 1, 2, 3\}$ can be defined. Under consistent detector-crystal tuning, the property (vi) is equivalent to the statement that the symmetry properties of each of the $m$-dependent sets of $3 \times 3$ matrices have to coincide with that of the 2D projection of the reciprocal crystal lattice. This requirement was found to hold indeed, within three significant decimal figures at least, the differences at the next digits coming probably from the statistical noise in the input data.

As an illustration, the $D(3)$ set is reproduced below:

<table>
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<tr>
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<th>50.02</th>
<th>26.80</th>
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<tr>
<td></td>
<td>26.15</td>
<td>2.949</td>
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<tr>
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<td>50.03</td>
<td>26.80</td>
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The results obtained at point (viii) are fundamental for the valuation of the whole analysis. Comprehensive information obtained in graphic form under use of various finite sections in the histogram plane was reported in references [2, 4], and [5], respectively.

Following [2], we provide, always for the set of data considered in [2] and [4], a global view of the 2 mm anisotropic part of the $D(3)$ area of the processed 2D-ACAR histogram (fig. 1), as well as areas equivalent to the extension of the first Brillouin zone, comprising the central $(4\pi/b, 0)$ second Umklapp component (fig. 2) and the central $(6\pi/b, 0)$
Fig. 1. — Global view of the 2 mm anisotropic part of CW-WLSF-smoothed experimental 2D-ACAR data on $\text{YBa}_2\text{Ca}_2\text{O}_{7-\delta}$ single crystals (symmetrized output), ($p_x$ and $p_y$ absolute ranges: ± 22.2 mrad; distance between neighbouring isolines: 0.60 mrad).

Note: In Figs 1, 2, and 3, the values along z-axis follow from the number of counts in the accumulated statistics.

Fig. 2. — 2D window from figure 1 at central second Umklapp component (symmetrized output); $p_x \in (9.45$ mrad, 15.75 mrad), $p_y \in (-3.15$ mrad, 3.15 mrad); distance between neighbouring isolines: 0.15 mrad.
third Umklapp component, respectively (fig. 3), under use of smoothing windows of constant radial parameter (Eq. (3.1) of reference [2]), \( r = 11 \).

In fig. 3, dominant are the positron-induced structures (wave function effects) which consist, within the present procedure of subtracting the 4 mm symmetric part of the spectrum, of two maxima and two minima, all of the same amplitude.

\[
\text{\texttt{H5_x11s.W03}} \quad \text{\texttt{H5_x11s.W03}}
\]

![3D graph](image)

**Fig. 3.** 2D window from figure 1 at central third Umklapp component (symmetrized output): \( p_x \in (15.75 \text{ mrad}, 22.05 \text{ mrad}) \), \( p_y \in (-3.15 \text{ mrad}, 3.15 \text{ mrad}) \); distance between neighbouring isolines: 0.15 mrad.

Finite slopes at \( p_x = \pm 13.2 \text{ mrad} \), extending over \( \sim 12 \text{ mrad} \) along the \( p_y \) axis (from \( \sim -6 \text{ mrad} \) to \( \sim +6 \text{ mrad} \)), occur at the second Umklapp component, in agreement with earlier evidence [18–22, 7, 12]. The central region of the right hand slope (got at \( p_x \in (-3.15 \text{ mrad}, 3.15 \text{ mrad}) \)) is represented on a magnified scale in fig. 2.

At \( p_x \) coordinates corresponding to the third Umklapp component, the scale of fig. 1 is not informative about the occurrence of further structures of the momentum distribution. However, local magnification of the scale of the representation unveiled [2] the occurrence of one-dimensional ridges, centred at \( p_x = \pm 18.0 \text{ mrad} \), \( \sim 2.2 \text{ mrad} \) wide, well resolved over \( \sim 9.5 \text{ mrad} \) along the \( p_y \) axis (from \( -4.75 \text{ mrad} \) to \( +4.75 \text{ mrad} \)). Once again, the central region with respect to \( p_x \) of the right hand ridge is shown in fig. 3.

There are two contrasting aspects of figures 2 and 3, the clarification of which need further discussion.

First, the height of the structure resolved in fig. 2 is about one-third of that of the positron-induced structures. By comparison, the height of
the ridge in fig. 3 is about 1/20 of that of the structure resolved in fig. 2. As it concerns the amplitude of the ruled out statistical noise, this was estimated to lie, at the second Umklapp component (fig. 2), below fifty percent of the height of the resolved structure. By contrast, at the third Umklapp component (fig. 3), the amplitude of the ruled out statistical noise was an order of magnitude larger than the height of the resolved structure. It is therefore no wonder that, at the second Umklapp component, the use of local windows of radial parameter \( r = 6 \) secured good resolution of the output (compare fig. 7b of [2] with the present fig. 2), while a radial parameter \( r = 11 \) still left an important fraction of residual noise at the third Umklapp component. Therefore, to get uniformly smoothed spectra, smoothing windows of variable area (i.e., data adapted) have to be used. The implementation of a CW-WLSF showing this feature is in train to be completed and results will be reported elsewhere.

A second difference between figures 2 and 3 follows from the information brought by the contour lines in the basal plane. At the second Umklapp component, the closure of the contour lines starting at the middle of the finite slope proceeds inside the periodicity cell covering the first Umklapp component. Thus, the analysis in the extended zone scheme at the second Umklapp component does not put actually into evidence the expected one-dimensional ridge. In contradistinction to this, at the third Umklapp component, the expected ridge structure is resolved indeed.

To find out whether this is a fundamental difference or not, recourse was once again made to the FLAPW theoretical representation of the \( \epsilon^* - \epsilon^- \) momentum distribution. Under the assumption of a Fermi level artificially raised up to the forbidden energy band, the structure of the second Umklapp component was still there (with modified parameters, particularly the slope, however), while that of the third Umklapp component completely disappeared. This result, while serving at reliable identification of the occurrence of a second Umklapp component of the Fermi surface break, suggests that the third Umklapp component of the Fermi surface break can be entirely attributed to the conduction electrons.

REFERENCES