Study of the Chain Related Fermi Surface in $(R)Ba_2Cu_3O_{7-\delta}$

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We have observed, by 2D positron annihilation, the rigid-shaped Fermi surface related to the chains in the compound series $(R)Ba_2Cu_3O_{7-\delta}$ for R=Y, Dy, Ho, and Pr. In the case of Y we have sighted the clear signature of the ridge in the fourth Brillouin zone and have observed the effect of substituting Cu with Ni, Zn, and Al. The presence of the ridge is also seen in $PrBa_2Cu_3O_{7-\delta}$ which has an insulating character. This shows that chains have a Fermi surface but contribute little to conductivity. This observation is discussed in view of the above alloying experiments, and in view of the recent model of Fehrenbacher and Rice.

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The compounds $(R)Ba_2Cu_3O_{7-\delta}$ show high superconducting transition temperatures when R stands for most of the rare earth elements or for Y. In these compounds the CuO_2 planes are generally held responsible for superconductivity while the one-dimensional Cu-O chains may play an indirect role by fixing the number of charge carriers in the planes. A notable exception is the nonsuperconducting $PrBa_2Cu_3O_{7-\delta}$ which has an insulating character [1,2]. In $PrBa_2Cu_3O_{7-\delta}$ the problem is twofold, first to account for the nonsuperconducting behavior and second the nonmetallic behavior. Models have been proposed, notably the one by Fehrenbacher and Rice [3], where the absence of superconductivity is ascribed to the existence of a local $4f$ Pr^{IV} hybridized state which binds doped holes to Pr sites and may also act as a magnetic pair breaker. The insulating character is ascribed by these authors mainly to the extreme sensitivity of the Cu-O chains and the mixed valence state of the Pr ions to O vacancies.

We have performed a systematic study of $(R)Ba_2Cu_3O_{7-\delta}$ compounds by positron annihilation 2D-ACAR (two-dimensional angular correlation of the annihilation radiation). We find that not only in the Y-based compound but also in Dy, Ho, and Pr compounds there is a Fermi surface (FS) sheet due to the Cu-O chains. This is particularly noteworthy in the case of the insulating Pr compound. Like Fehrenbacher and Rice [3], we conclude that the dc conductivity, which is a macroscopic property, is very sensitive to vacancies and impurities: It can be easily blocked by a small amount of disorder along the chains. Moreover, we add a very important experimental fact: At a smaller scale, these chains remain conductive in their nondefective parts.

Corroborative evidence supporting this conclusion comes from at least three sources: (1) similarity of the 2D-ACAR results for both the insulating (R=Pr) and the related metallic phases with R=Y, Ho, and Dy; (2) investigation of the influence of local disorder along the chains upon the corresponding FS sheets. This was studied (a) by measuring $YBa_2(Cu_{1-x}M_x)_3O_{7-\delta}$ where M =Ni, Zn, or Al and (b) by changing δ between ~ 0 and ~ 1 when $x=0$; and (3) precise band structure calculations using the full-potential linearized augmented plane wave (FLAPW) method [4], to confirm the statements made about the FS in $YBa_2Cu_3O_{7-\delta}$ from 2D-ACAR measurements.

In this Letter, we report evidence for the intrinsic conductivity of the Cu-O chains in $PrBa_2Cu_3O_{7-\delta}$ despite the macroscopic insulating character of this compound. Some of our arguments are based on results presented elsewhere [5].

Experimental evidence supporting the existence of a FS in superconducting cuprates has been obtained by angular-resolved photoemission [6] and de Haas-van Alphen [7,8] and positron 2D-ACAR [9–11] measurements. Each of these techniques has resolved one or more of the FS sheets expected from band structure calculations [4] and resulted in a good agreement between the observed FS topology and calculations based on the local density approximation.

As for 2D-ACAR [12], due to the positron distribution in the lattice [13], only the ridgelike FS related to the Cu-O chain states can be observed in $YBa_2Cu_3O_{7-\delta}$. We provide in Fig. 1 slices of 2D-ACAR anisotropy which generalize this statement to compounds with Y substituted by Dy, Ho, and Pr. The ridge is a periodic feature occurring at $p_x=6.28(i-1)$ in mrad, where 1 (mrad) $=10^{-3}mc=0.137$ a.u., and $i=1,2,\ldots$ labels the Brillouin zone (BZ) centered at $p_y=0$. Because of the limited range of the wave-function effects, the ridge is clearly visible for $i=3$ [5,9]. In untwinned cases [Figs. 1(a)–1(c)] it appears as a bump at $(p_x;p_y)=(12.6;0)$

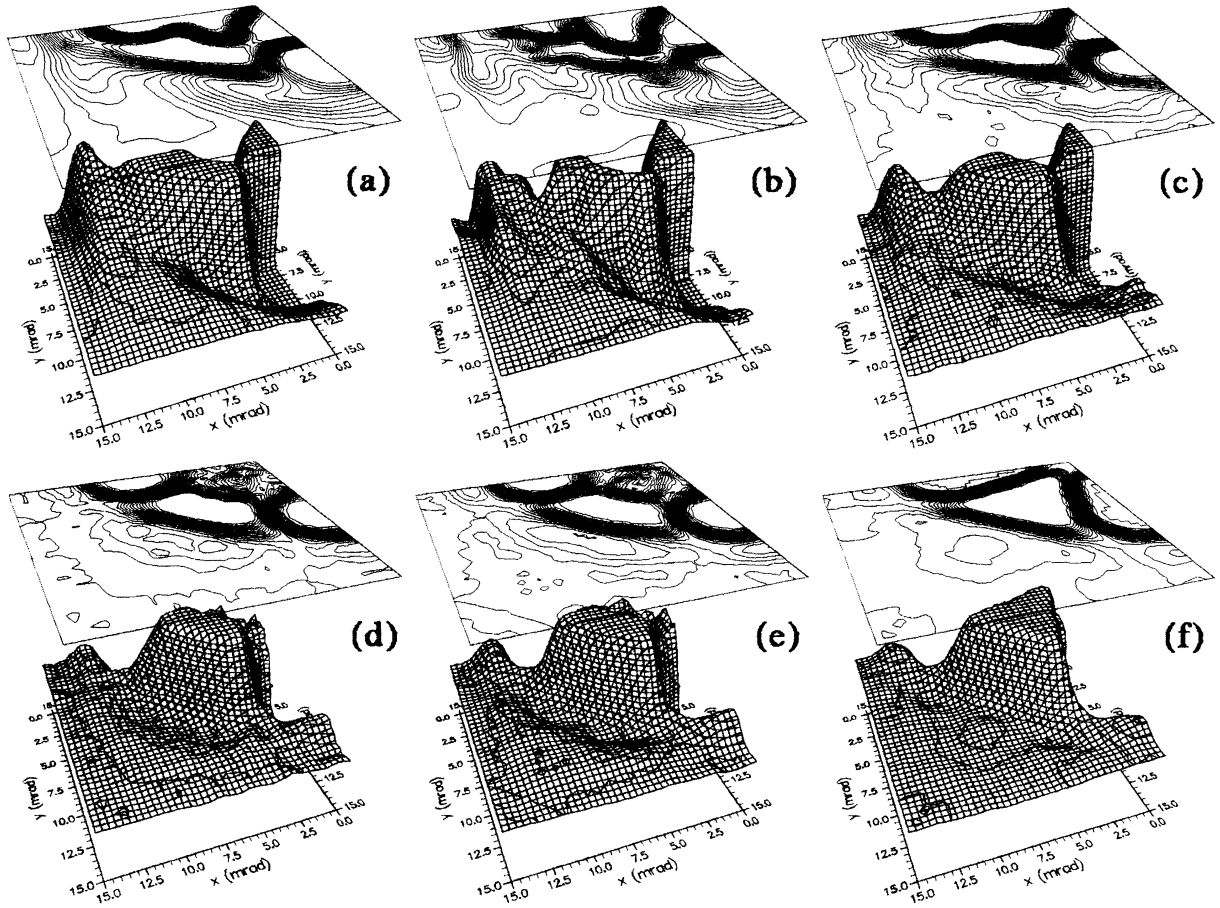

FIG. 1. 2D-ACAR anisotropic slices for $(R)Ba_2Cu_3O_{7-\delta}$, truncated to $\pm 10\%$. (a) $R=Y$, $\delta > 6.8$. (b) FLAPW calculation results of $YBa_2Cu_3O_7$. (c) $R=Dy$, $\delta > 6.8$. (d) $R=Pr$, $\delta > 6.8$. (e) $R=Ho$, $\delta > 6.8$. (f) O-deficient compound $YBa_2Cu_3O_{\sim 6.7}$. (a)–(c) Untwinned results; (d)–(f) twinned results. p_x is along [010] and p_y is along [100].

and extends along p_y. It is still visible at $p_y > 12$ (mrad). It can be seen in Fig. 1(b) that 2D-ACAR anisotropies evaluated from FLAPW calculations for $YBa_3Cu_3O_7$ are in excellent agreement with the measurement shown in Fig. 1(a). Moreover, we have observed that an arbitrary suppression of the FS from the calculations [5] completely modifies the structure which is thus indeed proved to originate in the ridge. For twinned samples ($R=$Pr and Ho) the ridge at $i=3$ appears systematically along both p_x and p_y as shown in Figs. 1(d) and 1(e). Our FLAPW calculations fully support this interpretation.

We conclude (1) that Ho, Dy, and Pr compounds have an electronic structure very similar to $YBa_2Cu_3O_7$, as suggested by other band structure calculations [14] and (2) that $PrBa_2Cu_3O_{7-\delta}$ has a ridge-shaped FS due to the Cu-O chains, like metallic Ho, Y, and Dy compounds, and despite its insulating character. The presence of the ridge is also compatible with optical reflectivity measurements [15], suggesting that the chains of the Pr compound behave like those of $YBa_2Cu_3O_y$ while the planes behave like the planes of $YBa_2Cu_3O_6$.

We have found that the temperature dependence of the 2D-ACAR from the Pr compound is very weak and the ridge is clearly resolved from 20 to 300 K. This is in contrast with $YBa_2Cu_3O_{7-\delta}$ where shallow trapping of positrons [10] occurs at low T, hence weakening the FS signal. Another interesting feature follows from Fig. 1(f): An $YBa_2Cu_3O_{6.7}$ compound also exhibits a ridge-related FS, despite the partial disorder on the chains. But when the oxygen deficiency is further increased the sample becomes insulating and the FS signal disappears [5]. This tells us that the ridge FS can survive a partial O deficiency on the chains.

Confidence in the presence of the FS is further reinforced by looking at $i=4$ (fourth BZ), but this requires measurements with statistics larger (10^9 counts) than usual. We show in Fig. 2(a) the region of interest for an untwinned $YBa_2Cu_3O_{6.95}$ sample [16], where we have plotted [9] the difference between the 2D-ACAR and its transposition so as to remove most of the wave-function effects. The ridge-shaped FS is most clearly visible in this figure [17]. Again it is in perfect agreement with our

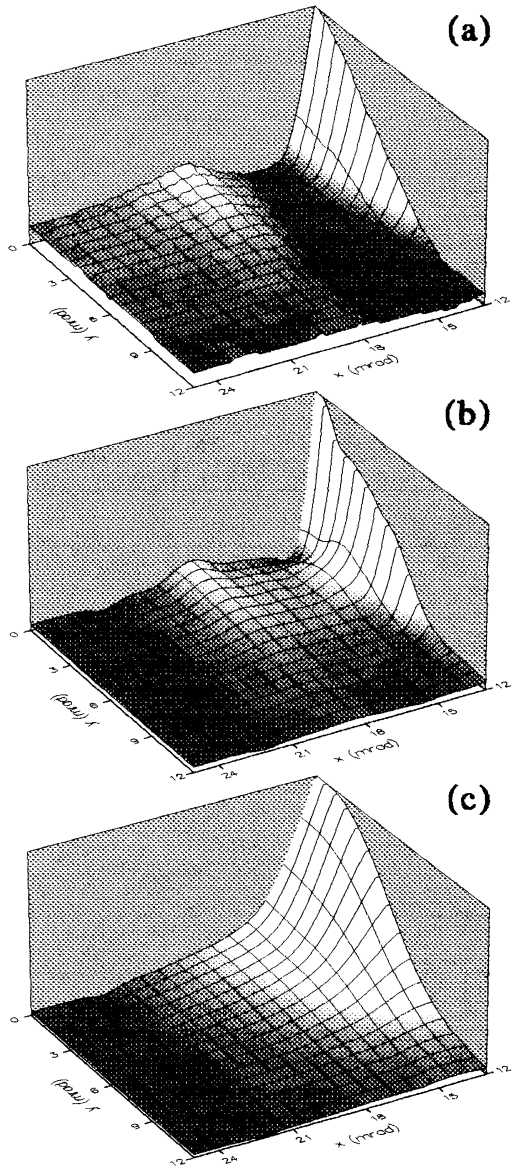

FIG. 2. Signature of the ridge-shaped Fermi surface of $YBa_2Cu_3O_{7-\delta}$ seen in the fourth Brillouin zone along the ΓY direction, in the anisotropic part of the measured 2D-ACAR. (a) Measurement, (b) FLAPW calculations, and (c) FLAPW calculations with artificially removed Fermi surface. p_x is along [010] and p_y is along [100].

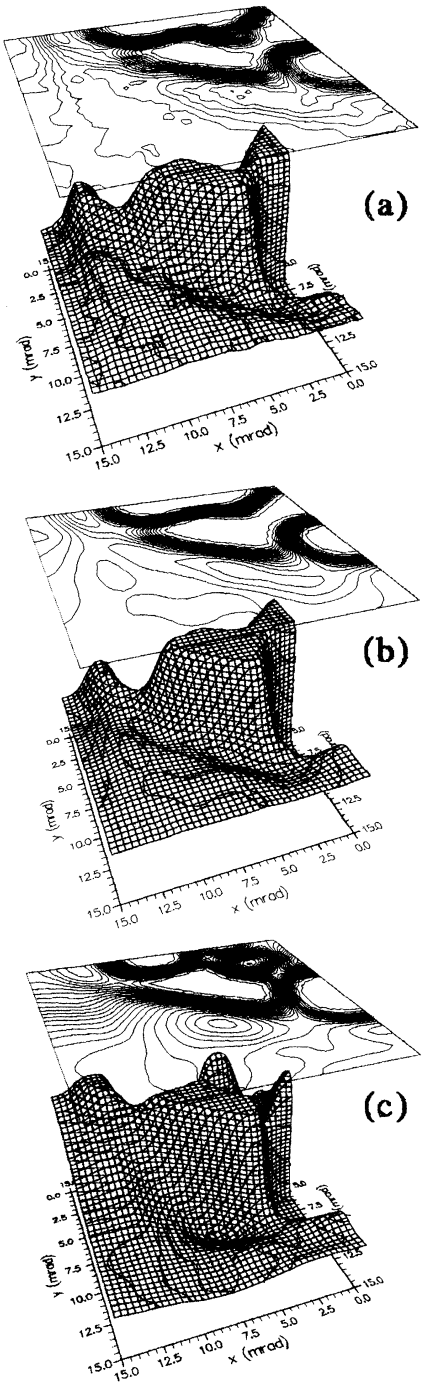

FIG. 3. 2D-ACAR anisotropic slices for $YBa_2(Cu_{1-x}M_x)O_{7-\delta}$, truncated to $\pm 10\%$. (a) M=Ni with $x=0.031$, (b) M=Zn with $x=0.035$, and (c) M=Al with $x=0.021$. p_x is along [010] and p_y is along [100].

FLAPW calculations [Fig. 2(b)], proving that the FS is the only origin of the feature observed: It totally disappears [Fig. 2(c)] when the FS is artificially removed from the calculations by filling the appropriate band.

To broaden our study of the FS in high-T_c Y-based cuprates we have investigated the effect of a partial substitution of Cu by 3.1 at.% Ni, 3.5 at.% Zn, and 2.1 at.% Al atoms. These substitutions lowered the superconducting transition temperature T_c to 73, 25, and 77 K, respectively. 2D-ACAR anisotropies are shown in Fig. 3. One observes that the distributions of the Ni- [Fig. 3(a)] and Zn-doped [Fig. 3(b)] compounds are similar to the pure $YBa_2Cu_3O_{7-\delta}$ [Fig. 1(a)]. The ridge width present in

both Ni- and Zn-doped samples is comparable to that of the undoped one. Ni substitutes for Cu atoms in CuO_2 planes, whereas the Cu-O chains are not affected by the substitution and the 1D band and its ridgelike FS are preserved. We remark that, despite a much lower T_c ($\sim$25 K), the Zn-doped sample does not exhibit a very different 2D-ACAR anisotropy and the ridge is still there; therefore we think that the positrons stay around the Cu-O chains for the Zn concentration we have, whereas Jean *et al.* [18] conclude from a positron lifetime study that the positron density transfers from the Cu-O chains to the CuO_2 planes. In the Al-doped sample [Fig. 3(c)], the anisotropy is quite different in the sense that it exhibits a nearly tetragonal symmetry and the ridgelike feature has almost completely vanished. The Al atoms substitute for the Cu atoms belonging to the Cu-O chains. If we assume that all Al atoms are located in the chains we have 9.3% of the Cu sites on the chains substituted by Al atoms. Our observations suggest that Cu-O chains are replaced by pieces of chains developing with increased Al concentration in both *a* and *b* directions. This suppresses the 1D band leading to the ridgelike FS.

To conclude, our 2D-ACAR investigation of the ridgelike FS related to the Cu-O chains in $(R)Ba_2Cu_3O_{7-\delta}$ with R=Y, Dy, Ho, and Pr has brought to light the existence of a FS in the insulating compound $PrBa_2Cu_3O_{7-\delta}$. Our interpretation is supported by the following observations: (1) Metallic Y-, Dy-, and Ho-based compounds provide data very similar to $PrBa_2Cu_3O_{7-\delta}$. (2) For $YBa_2Cu_3O_{7-\delta}$, FLAPW calculations, due to their very good agreement with the measurements, prove the FS origin of the observed structure in the third BZ. (3) A newly observed FS signal in the fourth BZ adds more confidence by its perfect agreement with calculations. (4) The FS signal disappears in $YBa_2Cu_3O_{7-\delta}$ when δ is increased enough to make the compound insulating.

Our data support the model proposed by Fehrenbacher and Rice [3] for the electronic properties of $PrBa_2Cu_3O_{7-\delta}$. We can ascribe the macroscopic insulating behavior of this compound to its extreme sensitivity to disorder on the chains which themselves remain metallic at the microscopic level until their 1D character is strongly damaged by defects and impurities, as is suggested by our experimental results for $YBa_2(Cu_{1-x}M_x)O_{7-\delta}$ with M=Zi, Zn, and Al.

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