Orbitally resolved lifetimes in \( \text{Ba(Fe}_{0.92}\text{Co}_{0.08})_{2}\text{As}_{2} \) measured by ARPES

V. Brout, M. Fuglsang Jensen, A. Nicolaou, A. Taleb-Ibrahimi, P. Le Févre, F. Bertran, A. Forget, and D. Colson

1Laboratoire de Physique des Solides, Université Paris-Sud, UMR 8502, Bât. 510, 91405 Orsay, France
2Synchrotron SOLEIL, L’Orme des Merisiers, Saint-Aubin-BP 48, 91192 Gif sur Yvette, France
3Service de Physique de l’Etat Condensé, Orme des Merisiers, CEA Saclay, CNRS-URA 2464, 91191 Gif sur Yvette Cedex, France

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There is a consensus that the multiband nature of iron pnictides is essential for defining their electronic properties. Many models for the superconducting and antiferromagnetic orders heavily rely on the interaction between different electron and hole Fermi Surface (FS) sheets. On the experimental side, transport, Raman, and quantum oscillations experiments seem to detect predominantly electrons, suggesting they have longer lifetimes than holes. However, ARPES experiments that can image independently hole and electron bands has not evidenced so far a clear difference between them that could explain this behavior. In fact, it has been more difficult to clearly image the electron pockets than the hole pockets with ARPES. Recently, an ARPES study resolved them in more details, but proposed a structure so different from theory, that it clearly calls for more investigation.

In this paper, we report ARPES measurements in \( \text{Ba(Fe}_{0.92}\text{Co}_{0.08})_{2}\text{As}_{2} \), which corresponds to optimal Co doping for superconductivity \( (T_{c}=23\text{K}) \). Similar features are found for other members of the \( \text{BaFe}_{2}\text{As}_{2} \) family. We first clarify that, in appropriate experimental conditions, the parity measured by ARPES are in good agreement with a recent study by Kemper et al. As the \( d_{xy} \) parts of the electron pockets are also associated with higher Fermi velocities on the electron pockets, this probably explains why electrons dominate in many experimental techniques. This also suggests that the two different orbitals play different roles in the electronic properties.

Single crystals were grown using a FeAs self-flux method. ARPES experiments were carried out at the CASSIOPEE beamline at the SOLEIL synchrotron, with a Scienta R4000 analyser, an angular resolution of 0.2° and an energy resolution better than 10 meV. Band structure calculations were performed within the local density approximation, using the Wien2K package and the experimental structure of \( \text{BaFe}_{2}\text{As}_{2} \).

Before presenting our ARPES data, we recall two particularities of the BZ structure that will be important for our discussion. First, as in all iron pnictides, the BZ (thick red lines in Fig. 1) corresponds to a unit cell with 2 Fe, because of the inequivalent positions of As above and below the Fe plane. Nevertheless, it is sometimes useful to think in an “unfolded” BZ containing just 1 Fe (dotted blue line), because this divides the number of bands by two. Moreover, folded bands typically have a very small intensity in photo emission (it is proportional to the strength of the potential at the origin of the folding) and they may not be detected. Second, this is specific to the stacking of the FeAs plane in \( \text{BaFe}_{2}\text{As}_{2} \). The folding takes place between different \( k_{z} \), namely along the vector \( q=(1,1,1) \) shown in the inset of Fig. 1. Following ref. [14], we sketch with solid lines the electron pockets expected in the unfolded BZ and with dotted lines the folded ones. The electron pockets are ellipses made out of \( d_{xy} \) and \( d_{xz}/d_{yz} \) orbitals with major axis oriented towards \( Z \), which rotates as a function of \( k_{z} \). This structure is completely at variance with...
We now examine in more details how many bands form these FS and how they connect together. Fig. 2 compares the measured and calculated dispersions in the 3 directions (a,b,c) indicated at bottom of Fig. 1. As odd measurement, we use those of panel α rotated by 90°, which are equivalent by symmetry. We indicate by colors their main orbital character, although bands are frequently hybridized here in a more complex way. For pure atomic orbitals, \(d_{xy}\) and \(d_{yz}\) would be odd with respect to the \(yz\) plane and \(d_{yz}\) even. Note that \(d_{xz}\) and \(d_{yz}\) switch roles along \(k_x\) and \(k_y\), but their parity with respect to \(xz\) and \(yz\) also switches, so that the selection rules remain identical. In the odd geometry (d-f), only one band is clearly observed. It corresponds to \(d_{xz}\) in the calculation (red line), which complies with the selection rule. It forms a shallow electron band of 50 meV along the minor axis of the ellipse (Fig. 2d) then becomes more squarish (Fig. 2e) and finally transforms into a band of opposite curvature 50 meV below \(E_F\) (Fig. 2f). At \(k_z=0\), this band is expected to stretch to form the ellipse long axis (see sketches in Fig. 3). In Fig. 3(b) and supplementary information, we show that this is actually well observed experimentally.

The \(d_{yz}\) band (green line) is not seen in this odd measurement, as expected from symmetry. On the other hand, the \(d_{xy}\) band should be observed, but is not. Surprisingly, it is the band detected in Fig. 2(i) in even geometry that perfectly matches the dispersion of \(d_{xy}\) [see also Fig. 4(b)]. It is a much deeper band with higher Fermi velocities. It clearly does not fit with \(d_{xz}\), which should cross \(E_F\) at similar points, but is much shallower, as clearly observed for its symmetric in odd geometry, both at \(k_z=0\) and \(k_z=1\). To convince oneself that this band is really \(d_{xy}\), it is interesting to follow how it transforms into the shallow band. Fig. 2(b) shows that...
as soon as one goes away from the diagonal, a gap opens between the $d_{xy}$ and $d_{xz}$ bands. The upper part of the deep band progressively transforms into the shallow band near $E_F$, while the $d_{xy}$ character is concentrated at the bottom of the band. This behavior is clearly observed in the data of Fig. 2(h), with a “kink” (white arrow) appearing in the dispersion of the deep band, where it crosses $d_{xz}$ that is not observed in this geometry. In Fig. 2(g), the intensity is quite weak and unclear, but probably contains residual contribution from $d_{xz}$, which is not strictly forbidden outside the $yz$ plane [22]. As the dispersion is intimately linked to the shape of one orbital, we conclude that the deep electron band is $d_{xy}$. The problem of parity is solved when one takes into account the 2 Fe of the unit cell [15]. Fig. 3 shows that it also follows qualitatively the expected behavior as a function of $k_z$ (its diameter shrinks away from $k_z=7\pi/c=1[\pi/c]$), although it strongly loses intensity near $k_z=6\pi/c=0[\pi/c]$.

The structure of the electron pocket is then quite simple, with only two interacting bands, whose dispersions are in remarkable agreement with theoretical predictions. A significant advantage of working with the electron bands is that the dispersion of the deep (blue, extracted from (a)) and shallow (red) electron bands at different $k_z$. For the shallow electron band, two samples were measured for $20<h\omega<45eV$ (circles) and $40<h\omega<90eV$ (stars). Thin lines are guides for the eyes. Ellipses on the right sketch how electron pockets evolve with $k_z$.

This clarified, we now show in Fig. 4 the dispersions and widths of the different bands at $k_z=1$ (for the shallow band, we use the equivalent of Fig. 2(a) at $k_z=0$ [22]. Some representative examples of the fits used to extract these quantities are shown in Fig. 4(c). Fermi velocities and linewidth are quoted in Table 1. Note that the deep electron band is quite narrow near $E_F$ ($\Delta k=0.04\pi/a$, i.e. an inverse lifetime $h/\tau=20meV$ after multiplication by $v_F$). This is just 1.5 larger than the outer hole band in LiFeAs that was claimed to be a particularly favorable case [24]. This invalidates the idea that lines would be extremely broadened by Co doping in BaFe$_2$As$_2$ [25] and too broad to perform meaningful self-energy analysis.

![Figure 3](image-url)  
Figure 3. (a) intensity integrated at $E_F$ for the deep electron band, for $25<h\omega<45eV$ (see [22] for translation into $k_z$). (b) $k_F$ for the deep (blue, extracted from (a)) and shallow (red) electron bands at different $k_z$. For the shallow electron band, two samples were measured for $20<h\omega<45eV$ (circles) and $40<h\omega<90eV$ (stars). Thin lines are guides for the eyes. Ellipses on the right sketch how electron pockets evolve with $k_z$.

![Figure 4](image-url)  
Figure 4. (a-b) Dispersion of the holes and electrons bands at $k_z=1$, compared with LDA band calculations at $k_z=0.5$ for BaFe$_2$As$_2$, renormalized by a factor 3.3. The open circles in (b) show maximum of EDCs. (c) Representative examples of MDC fits at $-0.03eV$ for holes (bottom), at $E_F$ for shallow (middle) and deep (top) electron bands. (d) Half width of the fitted lorentzians as function of the binding energy, averaged on the left and right bands.

| $d_{xz}$/$d_{yz}$ | $d_{xy}$ | $E_F$ (eV) | $m^*/m_0$ | $\delta k(E_F)$ (\pi/a) |
|------------------|--------|-----------|-----------|-----------------|
| hole             | 0.06   | 0.5       | 2.7       | 0.11            |
| hole             | 0.22   | 0.4       | 2.3       | 0.07            |
| electron         | 0.25   | 0.6       | 2.4       | 0.09            |
| electron         | 0.30   | 0.7/1.2   | 5/2.9     | 0.04            |

Table I. Properties of the different bands at $k_z=1$, extracted by the MDC fits of Fig. 4. For the $d_{xy}$ electron band, values at low ($v_{low}$) and high ($v_{tanh}$) binding energies are quoted.
In Fig. 4(a) and 4(b), the bands are compared with our LDA calculation at \( k_z = 0.5 \) renormalized by a factor 3.3. This order of magnitude is similar to those measured \[10\] or calculated \[28\] for related systems. The bands were not shifted, although the calculation was done for BaFe\(_2\)As\(_2\). We do observe shifts of the bands between BaFe\(_2\)As\(_2\) and this compound, consistent with electron doping \[21\], but some orbital and \( k_z \) dependent shifts would be needed to adjust the band structure to experimental data and this is not the scope of this paper. Calculations within DMFT give effective orbital masses of 2.4 for \( d_{xy} \) and 2.25 for \( d_{xz}/d_{yz} \), slightly different from our band masses, but the overall agreement of the calculated correlated band structure with experimental data is very good.

Fig. 4(d) shows that the deep electron band is a factor 2 narrower near \( E_F \) than the shallow one and that its linewidth increases much less steeply with binding energy. Interestingly, a similar anisotropy is observed for the hole bands having the same orbital characters. Although the hole bands are more difficult to separate [see Fig. 4(c)], we have found the same dependence for the 

Below 40meV, the dispersion of \( d_{xy} \) is more strongly renormalized and comes closer to that of the \( d_{xz}/d_{yz} \) band (see Table 1). Simultaneously, the linewidth nearly saturates. This behavior is of course reminiscent of a “kink”, which is often attributed to a coupling with a bosonic mode, phonons or magnons, and was sometimes proposed for iron pnictides \[25\]. Note, however, that the coupling would be very large here \( \lambda = \frac{\nu_{high}}{\nu_{low}} - 1 = 0.7 \), making this interpretation somewhat unlikely. In this multiband system, we rather believe that it is the signature of interactions between the different bands. Along the diagonal, a direct interaction, as in Fig. 2(h), is not expected. However, 40meV corresponds very well to the \( d_{xz}/d_{yz} \) bandwidth, suggesting that scattering between electrons in the two bands may take place below this energy scale and tend to homogenize their properties.

To conclude, we have detailed how \( d_{xy} \) and \( d_{xz}/d_{yz} \) combine to form electron pockets in Ba(Fe\(_{0.92}\)Co\(_{0.08}\))\(_2\)As\(_2\), reconciling ARPES and theory. We have then studied the self-energy for the different orbitals. It reveals a clear anisotropy between the behavior of the \( d_{xy} \) and \( d_{xz}/d_{yz} \), both for the hole and electron bands, with longer lifetimes on \( d_{xy} \). As \( d_{xy} \) form the extremities of the electron pocket, where \( \nu_F \) is also higher, these parts are likely a “cold spot” on the FS that could dominate some electronic properties, such as transport. We tentatively attribute the shorter lifetimes of \( d_{xz}/d_{yz} \) to stronger spin fluctuations. We also evidence inter-scattering effects between \( d_{xy} \) and \( d_{xz}/d_{yz} \) near the Fermi level. As they are probably very dependent on the relative nesting of these two bands, this could be a key to stabilize magnetism or superconductivity. To test this idea, following precisely the fate of \( d_{xy} \) under isovalent and aliovalent substitutions should be very instructive.

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Supplementary information

Fermi Surface at $k_z=0$

In Fig. 5, we show the FS in similar conditions as those of Fig. 1, but at a photon energy of 27eV, corresponding approximately to $k_z=0$ on the electron pockets. The real trajectories in a $yz$ plane at 27 and 34eV are sketched in Fig. 5b, using $k_z = \sqrt{0.512^2(h\omega - W + V_0) - k_{yy}^2}$, where $k_{yy}$ is component of the momentum $k$ in the surface plane, $W$ is the work function (4.2eV) and $V_0$ an inner potential estimated to be 14eV [26].

![Figure 5](image)

Figure 5. (a) FS in same condition as Fig. 1, except the photon energy was 27eV. (b) Sketch of the BZ in the $yz$ plane. Black lines indicate trajectories in k-space for the two photon energies used in this study.

In odd geometry, the FS at $k_z=0$ and $k_z=1$ have similar appearances, except that the FS at $k_z=0$ is stretched in the $k_y$ direction. As detailed in Fig. 6, they are both created by the shallow electron band, which stretches, almost as expected theoretically (see also Fig. 3 and ref. [12]). The data in Fig. 4 for the shallow electron band correspond to Fig. 6d. The diameter of the band is somewhat smaller than in theory, for all $k_z$. As one can expect larger electron pockets for this Co doped compound than in pure BaFe$_2$As$_2$, for which the calculation was performed, this is a significant difference. However, the tendency of having smaller pockets than predicted theoretically was already noted before (see for example ref. 20) and probably corresponds to an upward shift of these bands. At 39eV, one starts to distinguish the two electron bands (see below).

In even geometry, the evolution is more complicated, as one would also expect the FS to keep a similar appearance (in this case, it should shrink near $k_z=0$), but the high intensity parts seem to rotate. We have observed a similar FS at the next $k_z=0$ point (48eV), ensuring that it is a periodic effect. A similar behavior was also observed in ref. [12] and it was proposed that the high intensity parts in panels $\beta$ and $\delta$ form a large square pocket of $d_{x^2-y^2}$ symmetry.

In Fig. 7, we present the dispersions along $k_x$ and $k_y$ to better understand the evolution. In Fig. 7a, we observe the deep electron band, corresponding well to $d_{xy}$ in the calculation. As shown in Fig. 3a, it loses intensity at lower $k_z$ and it is indeed not detected anymore at $k_z=0$ (Fig. 7a). Another band is observed that does not cross $E_F$. It could correspond to $d_{yz}$, but this is unlikely as it is not allowed by symmetry. It is rather the folded $d_{xy}$ that would be shifted up by about 50meV compared to the calculation. Hence, this is as if intensity had switched from the main bands at $k_z=1$ to the folded bands at $k_z=0$. At least, there are strong modulations of the intensity with $k_z$ that are not expected by orbital symmetry alone.

In Fig. 7c, the overall intensity is much weaker and more difficult to interpret. We probably observe the main $d_{xy}$ band at high binding energies and some traces of the $d_{yz}$ band near $E_F$. Although, this band is not allowed by

[29] M. Aichhorn, private communication. $U = 3.4$ eV, $J = 0.85$ eV, and doping $z = 0.1$ were used.
symmetry, this is strictly true only in the $yz$ plane (i.e. for the cut along $k_y$) and the intensity is quite weak anyway, so that even a small misalignment could produce this residual intensity. Having observed this, one realizes that the dispersion in Fig. 7 is in fact very similar to that in 7e, maybe with some more intensity of the folded $d_{xy}$ band. Therefore, the different appearance of the FS is not at all due to a rotation, but to a different contrast in the intensity of the bands along $k_x$ and $k_y$, resulting mainly from the loss of intensity of the deep electron near $k_z=0$. For this reason, we believe these two parts are unlikely to form a large square pocket of $d_{x^2-y^2}$ symmetry, as proposed in ref. 12, because they are not the same bands.

Figure 6. (a-b) Zoom on the electron pockets odd/yz of panels $\alpha$ and $\epsilon$ (Fig. 1 and S1), together with sketches of the electron pockets. (c-d) Corresponding dispersions along $k_y$, together with calculated bands rescaled by a factor 3. Blue lines correspond to $d_{xy}$, red lines to $d_{xz}$ and green lines to $d_{yz}$. Solid and dashed lines correspond to main and folded bands. (e) Same at 39eV.

Figure 7. (a-b) Zoom on the electron pockets even/yz of panels $\beta$ and $\delta$ (Fig. 1 and S1), together with sketches of the electron pockets. (c-f) Corresponding dispersions along $k_y$ (c-d) and $k_x$ (e-f), together with calculated bands rescaled by a factor 3.