# Opening of the superconducting gap in the hole pockets of $Ba(Fe_{1-x}Co_x)_2As_2$ as seen via angle-resolved photoelectron spectroscopy

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We present an angle-resolved photoelectron spectroscopy study of the changes in the electronic structure of electron-doped $Ba(Fe_{1-x}Co_x)_2As_2$ across the superconducting phase transition. By changing the polarization of the incoming light, we were able to observe the opening of the gap for the inner hole pocket $\alpha$ and to compare its behavior with the outer holelike band $\beta$. Measurements along high-symmetry directions show that the behavior of $\beta$ is consistent with an isotropic gap opening, while slight anisotropies are detected for the inner band $\alpha$. The implications of these results for the $s\pm$ symmetry of the superconducting order parameter are discussed, in relation to the nature of the different iron orbitals contributing to the electronic structure of this multiband system.

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## I. INTRODUCTION

After the recent discovery of iron-based superconducting compounds,[1,2] several experimental and theoretical studies tried to explain the mechanisms underlying their high critical temperatures ($T_c$). To this end, the symmetry of the order parameter is one critical piece of information.[3] For instance, considerable interest was raised by early theoretical predictions,[4,5] describing the possibility of a $s\pm$ symmetry with a gap following the law $\Delta = \Delta_0[\cos(k_x) + \cos(k_y)]$, implying a dephasing of $\pi$ between hole and electron pockets of the Fermi surface.

Experimentally, angle-resolved photoelectron spectroscopy (ARPES) has been widely used in this search, thanks to its capability of resolving in $k$ space the electronic structure of materials.[6] This technique has been employed to measure the amplitude of the superconducting (SC) gap in all the existing iron-based compound families: the 1111 ($RE(O_{1-x}F_x)FePn$, RE being a rare earth and Pn a pnictogen),[7,8] 11 ($FeSe_{1-x}$, $FeSe_{1-x}Te_x$, and $FeTe_{1-x}S_x$),[9] 111 (LiFeAs and CaFeAs),[10,11] in the iron selenides $A_xFe_2Se_2$ ($A$ = Cs, K),[12] and in the 122 compounds (doped $AEFe_2As_2$, AE being an alcaline-earth metal).

In particular, this latter family has been extensively studied due also to the availability of high-quality samples. The 122 compounds present a Fermi surface composed of three hole pockets (two almost degenerate inner $\alpha$ bands and an outer $\beta$ band) located around the ΓZ direction, and two electron pockets ($\gamma$ and $\delta$) located around XA. The Fermi surface and the folded first Brillouin zone in two dimensions are shown in Fig. 1(d). In a recent study, we showed that selective measurements of these bands are possible with polarization-dependent ARPES, which also gives insight into the orbital character for each one of them.[13] The highest $T_c$ in this family is obtained in optimally hole-doped $Ba_{0.6}K_{0.4}Fe_2As_2$, for which several ARPES experiments showed that the SC gap amplitude was band dependent[14–17] and presented a three-dimensional character for one of the hole pockets.[18,19] However, another recent study showed an orbital independent SC gap amplitude in both hole-doped $Ba_{1-x}K_xFe_2As_2$ and isovalent $BaFe_2(As_{1-x}P_x)_2$,[20] raising the question of a possible orbital-fluctuations mediated superconductivity mechanism.

Only one ARPES measurement of the SC gap has been reported so far in the electron-doped system $Ba(Fe_{1-x}Co_x)_2As_2$.[21] The SC gap was measured to be larger in $\beta$ than in $\gamma$; no information was obtained on the $\alpha$ gap, since this band could not be seen to cross the Fermi level. Interestingly, the SC gap appeared isotropic for both $\gamma$ and $\beta$.

We report here an observation of momentum and band-dependent SC gaps using polarization-dependent ARPES in $Ba(Fe_{1-x}Co_x)_2As_2$, which allowed us to detect the gap opening across $T_c$ for both $\alpha$ and $\beta$. These results confirm that the order parameter is consistent with a $s\pm$ symmetry. A slight anisotropy in the behavior of $\alpha$ along ΓX and ΓM could be detected, and we discuss how this may require the introduction of multiple harmonic terms for the description of the order parameter.

## II. EXPERIMENTAL

We studied single-crystal samples of $Ba(Fe_{1-x}Co_x)_2As_2$, $x = 0.07$ with $T_c = 24.5$ K, grown by the self-flux method.[22] They were fully characterized and oriented prior to our measurements. ARPES experiments were performed on the BaDElPh beamline at the Elettra synchrotron light source,[23] which is optimized for photon energies $h\nu$ below 10 eV, allowing bulk-sensitive measurements; all the results presented in this article have been obtained at a photon energy of 9 eV. The photoelectron analyser was a SPECS Phoibos 150, with an energy resolution of 5 meV, and an angular resolution of 0.1°. Measurements were performed on high-quality surfaces cleaved *in situ* under ultra-high vacuum with

 

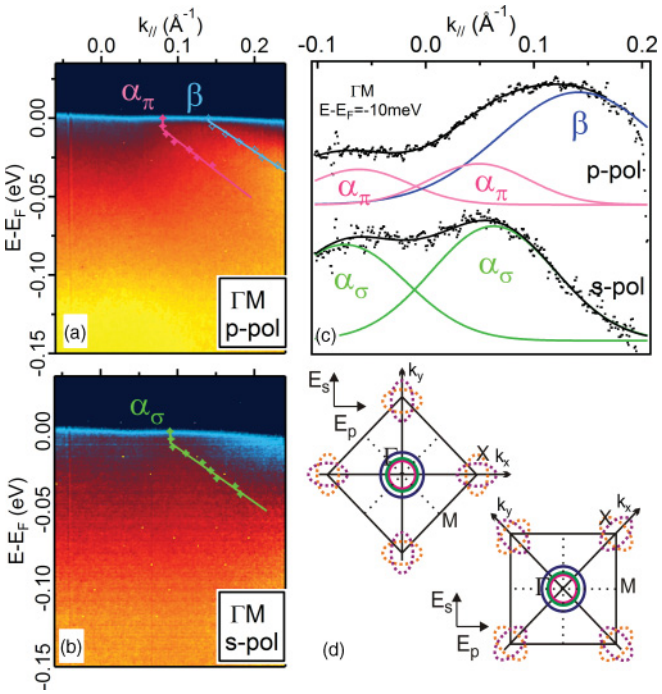


FIG. 1. (Color online) ARPES images acquired at $T = 14$ K along the ΓM sample orientation, (a) in $p$ and (b) in $s$ polarizations. (c) Momentum dispersion curves extracted from the images at $E - E_F = -10$ meV. (d) Experimental geometries for measurements along ΓX and ΓM.

pressure better than $5 \times 10^{-11}$ mbar. The reproducibility of all the experimental results presented here was verified via multiple thermal cycles across $T_c$, yielding consistent results.

## III. RESULTS

ARPES images in the ΓM direction are presented in Figs. 1(a) and 1(b). Three holelike pockets are visible, two almost degenerate inner $\alpha$ bands (that in our previous work[13] were named $\alpha_\sigma$ and $\alpha_\pi$) and an outer $\beta$ band, as shown in the momentum dispersion curves (MDCs) in Fig. 1(c). As shown in our previous study,[13] photon polarization and $k_z$ dispersion effects make it possible to obtain information on their orbital character. Namely, even orbitals with respect to the photoemission plane are measureable with $p$-polarized photons, while odd ones are visible only with $s$-polarized photons. Our experimental geometry is depicted in Fig. 1(d), and the notations for sample orientation and photon polarization are the same as in Ref. 13; in particular, ΓX corresponds to the nearest neighbours Fe-Fe bond direction. In the $k_z \cong 0.8\,[4\pi/c]$ plane, probed with $h\nu = 9$ eV, and along ΓM, $\beta$ has a marked $d_{z^2}$ character, which in our experimental configuration is even and thus measurable only in $p$ polarization. On the other hand, the $\alpha$ bands are formed by combinations of $d_{xz}$ and $d_{yz}$ orbitals and are detectable for every sample orientation and polarization. Consequently, in $p$ polarization, we simultaneously detect both the even $\alpha$ band and $\beta$, with $\beta$ having an overwhelming spectral weight. Conversely, in $s$ polarization, $\beta$'s photoemission yield is suppressed by matrix element effects, and the odd $\alpha$ band can

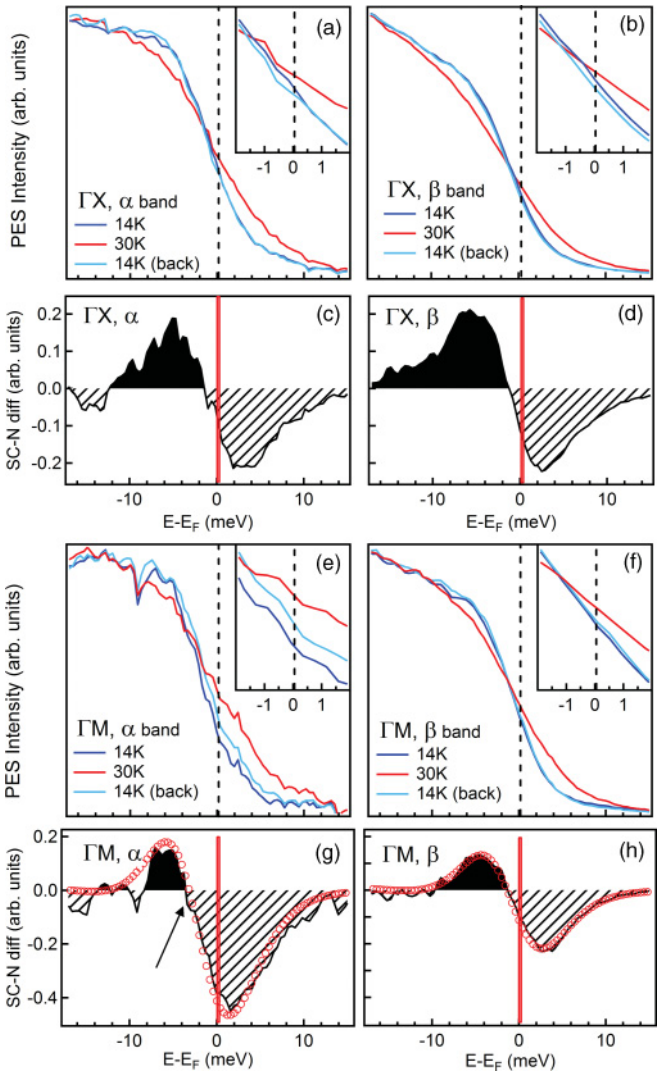


FIG. 2. (Color online) (a)–(c) Energy dispersion curves for the $\alpha$ and $\beta$ bands, along ΓX (a) and (b) and ΓM (e) and (f); the insets show in more detail the Fermi level region. (c) and (d) and (g) and (h) Corresponding differences between superconducting phase and normal state. Red dots in (g) and (h) correspond to simulations using a BCS-like function, giving for $\beta$ a coherent quasiparticle with width 3.5 meV and centered at 2 meV and for $\alpha$, the same width but at 3.5 meV away from the Fermi level.

be measured in a very accurate way, which was not possible in the previous gap study on this system.[21]

In Fig. 2, we present the energy dispersion curves (EDCs), extracted from ARPES images along ΓM and ΓX, at the Fermi wave vector corresponding to each band and for two temperatures across the superconducting (SC) phase transition. Upon cooling down, a shift of the Fermi leading edge (FLE) is observed, corresponding to the opening of the SC gap, in every band. This shift can be also seen in the differences between the SC and normal state EDCs, shown in Figs. 2(c), 2(d), 2(g), and 2(h). The simple thermal contribution to the Fermi level broadening—different at $T = 14$ and 30 K—would give a symmetric peak-valley feature in the differences. For our data, the zero-level crossing happens at energies lower than the Fermi energy [indicated by an arrow in Fig. 2(g)], which

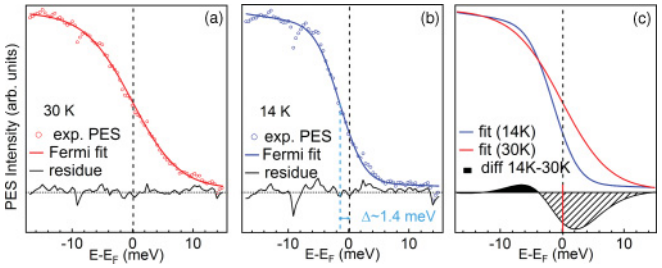


FIG. 3. (Color online) Energy dispersion curves fitting by a Fermi function for $\alpha$ band along ΓM at (a) 30 and (b) 14 K. (c) Fitting functions with their difference; the Fermi level crossing is shifted toward negative energies, indicating the opening of the superconducting gap.

is a very sensitive indication of the shift of the leading edge. Since no prominent quasiparticle peak could be observed in the SC state, we did not try to evaluate the gap value by modeling the spectral line shape.[24,25]

We preferred to carefully fit the EDCs with Fermi functions in order to precisely quantify the FLE shift, which is proportional to the gap, and measure this value for high-symmetry directions in $k$ space. The fitting functions together with the experimental spectra for the $\alpha$ band along ΓM are shown in Figs. 3(a) and 3(b), and the difference between the two fitting functions across $T_c$ in Fig. 3(c). The FLE shifts obtained by this procedure are the following: along ΓM, we obtain for $\beta = 0.5 \pm 0.4$ meV and for $\alpha = 1.3 \pm 0.4$ meV. On the other hand, along ΓX the FLE shifts are $0.55 \pm 0.4$ for $\beta$ and $0.6 \pm 0.4$ for $\alpha$, as summarized in Fig. 4(c).

This small but unambiguous difference between the FLE shifts of $\alpha$ and $\beta$ deserves further attention, but first the question that should be addressed is if other factors than the opening of the SC gap can affect such shifts. One important factor that should be kept in mind is that the FLE is directly affected by the scattering rate: the stronger the scattering, the broader the superconducting coherent peak, and thus the smaller the Fermi leading edge. The scattering can be, in general, band- and momentum-dependent and may produce an effective gap anisotropy along ΓM and ΓX.

We tried to evaluate these possible effects on our data by using a BCS-like function to fit the differences in the EDC's presented in Figs. 2(g) and 2(h), in order to model the transfer of spectral weight into the superconducting coherent peak for the two bands along ΓM. As already mentioned, the absence of a marked SC gap makes it hard to unambiguously fit the superconducting coherent peak. Once the hypothesis on the SC peak line shape is made (BCS in our case), fitting the SC-normal differences is instead a very sensitive way of obtaining information on the coherent peak position—the price to pay is of course the absence of any information on the overall density of states in the two phases. Our fits show that the differences for both $\alpha$ and $\beta$ can be correctly reproduced only by using the same values for the full width of the coherent peak (3.5 meV) in the two cases, but with different values for its position (3.5 and 2 meV for $\alpha$ and $\beta$, respectively). This suggests that the difference in scattering rate should not be regarded as the main factor in determining the inequivalent behavior for $\alpha$ and $\beta$ along ΓM; furthermore, it corroborates the hypothesis that

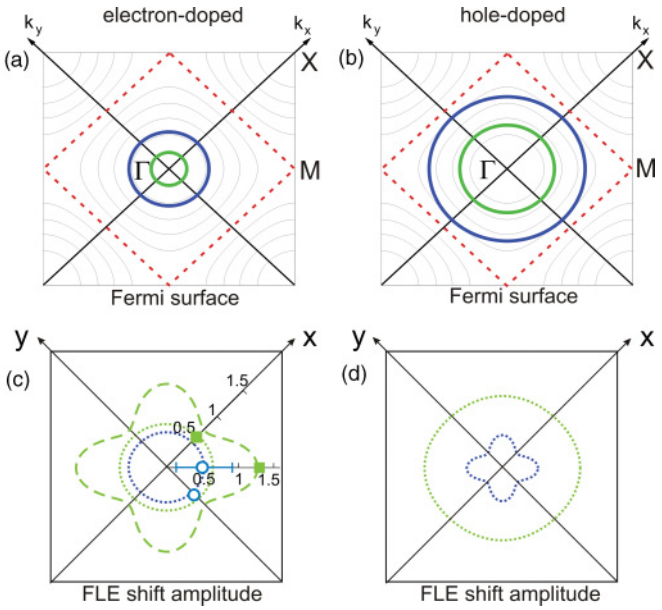


FIG. 4. (Color online) (a) Inner ($\alpha$, green) and outer ($\beta$, blue) holelike pockets in the folded Brillouin zone of $Ba(Fe_{1-x}Co_x)_2As_2$; the black lines represent the order parameter contour lines corresponding to the $s\pm$ symmetry, and the nodal lines (zero value) are the red dashed lines. (b) Example of a similar situation with larger Fermi surfaces (hole-doped compounds). (c) Expected gap amplitude dispersion (in dotted lines) for the situation described in (a) and experimental points; green squares correspond to the $\alpha$ band and blue circles to the $\beta$ one. The green dashed line is a guide to the eye illustrating a behavior for $\alpha$ compatible with the experimental data (d) expected gap amplitude dispersions for the situation described in (b).

the FLE shift is proportional to the SC gap, and indicates a slight anisotropy in the gap opening for the $\alpha$ band.

## IV. DISCUSSION

Before discussing the possible implications of this small anisotropy on the superconducting order parameter, we present in Fig. 4(a) a schematic view of the holelike pockets, together with the contour lines expected for an $s\pm$ order parameter with a $\cos(k_x) + \cos(k_y)$ behavior, and we show the corresponding $k$ dependence of the gap amplitude in Fig. 4(c), in dotted lines. In Figs. 4(b) and 4(d), we present a similar situation with larger Fermi surfaces, which would be representative of a hole-doped compound.

One should observe that the contour lines of the order parameter magnitude evolve from a circular shape around the Γ point to a diamond shape (where its value is zero) for larger momenta. Therefore its magnitude on a small circular Fermi surface around Γ [green curve in Fig. 4(a) and corresponding gap in Fig. 4(c)] will be nearly constant, while a flower-shaped modulation is expected for larger circular Fermi surfaces [blue curve in Fig. 4(b) and gap in Fig. 4(d)]. Also the overall magnitude is expected to be smaller in the latter case, because the Fermi surface approaches the node line (red dashed contour lines). This scenario is indeed found for the hole-doped pnictides of the 122 family[15] [see Figs. 4(b) and 4(d)].

Conversely, our results do not fit into this simple picture: the experimental FLE shift values [blue circles in Fig. 4(c)] for the larger Fermi surface ($\beta$, dark blue) are quite isotropic, while the experimental FLE shift values [green squares in Fig. 4(c)] for the smaller Fermi surface [$\alpha$, in green in Fig. 4(a)] are compatible with a flowerlike modulation.

Our results are consistent with the symmetry expected within the $s\pm$ scenario.[4,5] However, they also indicate that the simplest version of this model in which the superconducting order parameter follows a pure $\cos(k_x) + \cos(k_y)$ law, has to be extended;[5] in order to account for our findings, in particular, for the modulation of the $\alpha$ band, a band-dependent order parameter is needed.

One possibility for this is to have higher harmonics contributing to the order parameter, with orbital dependent weighting prefactors. Higher harmonics are needed for the gap on the inner Fermi surface to modulate, because they have more node lines, and they are closer to $\Gamma$; these harmonics have however to contribute much less to the gap of the outer Fermi surface. Such strong orbital dependence is in line with the analysis by Kuroki *et al.*[5,26]

A few considerations should be made on the differences observed for the electron- and hole-doped compounds of the 122 family. First, as just discussed, the order parameter can be described with a single harmonic term for the hole doped systems, while multiple terms might be necessary for the electron doped ones. Then another important difference concerns the relation between gap amplitude and Fermi surface nesting: in hole-doped $Ba_{0.6}K_{0.4}Fe_2As_2$, the largest SC gap opens in the hole pocket presenting the most efficient nesting with electron pockets,[17] which can be easily explained by interband scattering favoring a larger SC order parameter amplitude.

In the case of electron-doped $Ba(Fe_{1-x}Co_x)_2As_2$, nesting with the electron pockets was found for the $\beta$ band,[27] and this doesn't seem to be correlated to a larger gap; it may be related to a stronger scattering rate for $\beta$ than for $\alpha$, which would affect the effective FLE shift, but as discussed above this is unlikely to be the main factor emerging from our experimental data. Furthermore, it should be kept in mind that the gap values presented here were measured for a $k_z$ different from the values explored in Ref. 27. Since $Ba(Fe_{1-x}Co_x)_2As_2$ presents a marked three-dimensional character, an extensive $k_z$ dependent measurement of the gap (as performed in K-doped 122 compounds[18,19]), would be needed to get clear indications on its relation with nesting.

The differences between hole- and electron-doped compounds may come from the multiorbital character of their electronic structure. While the inner hole pockets of K-doped 122's are formed by $d_{xz}$ and $d_{yz}$, the outer band is constituted of $d_{xy}$.[18] On the other hand, the two inner bands of Co-doped 122's are constituted of $d_{xz}$ and $d_{yz}$, but the outermost one is hybridized in a more complex way, especially, with $d_{z^2}$ away from $k_z \cong 1\,[4\pi/c]$.[13] It should be noted that a recent ARPES study on both hole- and isovalent-doped 122 compounds by Shimojima *et al.* shows evidence of an orbital-independent order parameter amplitude for the different holelike pockets,[20] while for the electron-doped system presented here, we find instead indications of orbital dependence. The importance of these considerations should trigger more studies on the symmetry of the order parameter comparing electron- and hole-doped compounds of the 122 family. For ARPES experiments, it would be particularly important to compare more extensive experimental results with detailed calculations of the electronic structure of these compounds: this would allow to unambiguously extract quantitative values for the SC gap, going beyond the level of uncertainty coming from the use of FLE shifts and taking into account the density of states of these material when modeling the transfer of spectral weight from the normal to the superconducting state.

## V. CONCLUSION

In conclusion, we performed an angle-resolved photoemission study of the holelike Fermi surface above and below the superconducting phase transition in optimally doped $Ba(Fe_{1-x}Co_x)_2As_2$ along high-symmetry directions in $k$ space. The analysis of the Fermi leading edge shifts confirms that the SC gap is isotropic for the outer hole pocket $\beta$, while it shows slight anisotropies for the inner one, $\alpha$. These results are consistent with a $s\pm$ order parameter following a $\cos(k_x) + \cos(k_y)$ behavior, and suggest that multiple harmonic terms may be included in its description. More comparative studies between hole- and electron-doped materials of the 122 family appear necessary to completely understand the implications of these results, and the dependence of the superconducting order parameter on the nature of the various orbitals contributing to the physics of these multiband materials.

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