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Fermi surface and effective masses in photoemission response of the $(\text{Ba}_{1-x}\text{K}_x)\text{Fe}_2\text{As}_2$ superconductor

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The angle-resolved photoemission spectra of the superconductor $(\text{Ba}_{1-x}\text{K}_x)\text{Fe}_2\text{As}_2$ have been investigated accounting coherently for spin-orbit coupling, disorder and electron correlation effects in the valence bands combined with final state, matrix element and surface effects. Our results explain the previously obscured origins of all salient features of the ARPES response of this paradigm pnictide compound and reveal the origin of the Lifshitz transition. Comparison of calculated ARPES spectra with the underlying DMFT band structure shows an important impact of final state effects, which result for three-dimensional states in a deviation of the ARPES spectra from the true spectral function. In particular, the apparent effective mass enhancement seen in the ARPES response is not an entirely intrinsic property of the quasiparticle valence bands but may have a significant extrinsic contribution from the photoemission process and thus differ from its true value. Because this effect is more pronounced for low photoexcitation energies, soft-X-ray ARPES delivers more accurate values of the mass enhancement due to a sharp definition of the 3D electron momentum. To demonstrate this effect in addition to the theoretical study, we show here new state of the art soft-X-ray and polarisation dependent ARPES measurements.

The iron pnictides are nowadays one of the most studied examples for unconventional superconductivity. Due to their complex properties standard theoretical methods based on a local density approximation (LDA) within density functional theory (DFT) often fail^{1–5}. This is especially true if one tries to explain angle-resolved photoemission (ARPES) spectra of the iron pnictides^{6–13}. In this context a significant discrepancy between the effective masses derived from experimental ARPES spectra m_{exp}^* and from LDA band structure calculations m_{LDA}^* was reported^{11, 13, 14}. Correct trends in the effective masses can be observed using dynamical a mean-field theory (DMFT) approach which quantifies the importance of correlation effects for the iron pnictides^{15, 16}.

Various advanced approaches have been applied in the field, accounting for different phenomena. This covers the treatment of disorder in an appropriate way^{17–19}, the inclusion of spin-orbit coupling (SOC)²⁰ and in order to calculate ARPES spectra correctly the influence of matrix element effects and surface effects was recently stressed^{21, 22}. Finally, electron-electron correlation effects are one of the most important issues discussed^{10, 15, 23–27}. All these aspects were shown to play a crucial role for the iron pnictides, yet most approaches so far can deal with only one of these issues at the same time.

In this work we will present a theoretical approach which accounts for all of the above mentioned issues leading in this way to a very satisfactory agreement with experimental ARPES data of the iron pnictides. Here we investigate one of the most prominent prototype systems in the family of iron pnictides, namely the K substituted $(\text{Ba}_{1-x}\text{K}_x)\text{Fe}_2\text{As}_2$ compound^{28, 29}, which was extensively studied by ARPES^{6–9, 12, 30, 31}. There is common agreement, that the Fermi surface (FS) of this compound is quite complex and cannot be obtained from plain DFT calculations. In fact, an exceptional propeller-like FS topology at the \bar{X} point is found^{16–8} which is discussed in terms of a Lifshitz transition,

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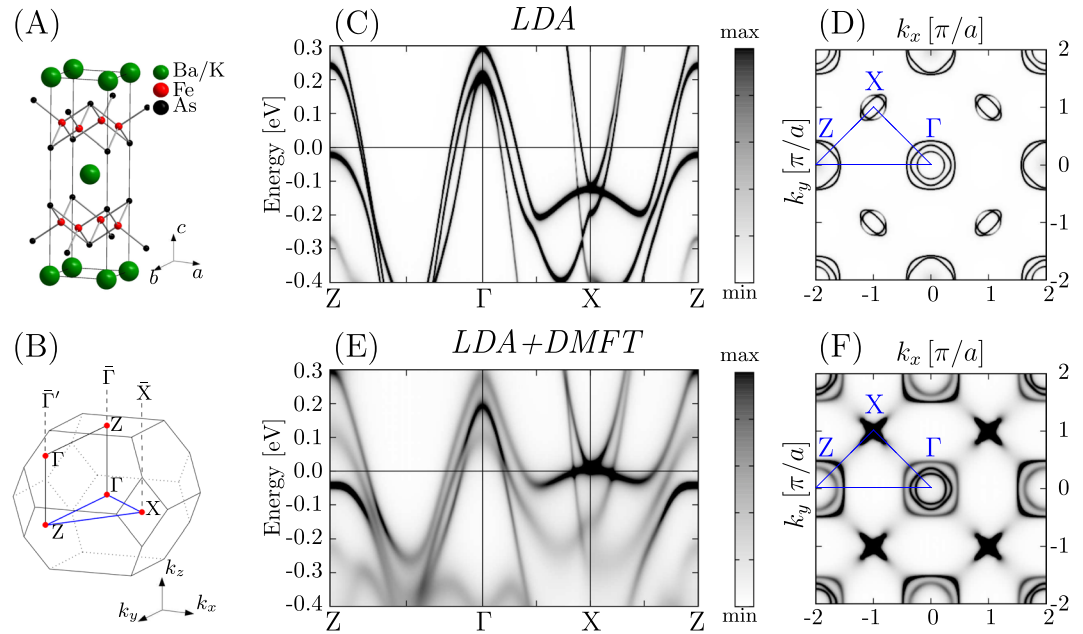


Figure 1. (A) Crystallographic unit cell of tetragonal $(\text{Ba}_{1-x}\text{K}_x)\text{Fe}_2\text{As}_2$ with (B) corresponding Brillouin zones indicating the important high symmetric points. $\bar{\Gamma}$, $\bar{\Gamma}'$ and \bar{X} indicate the two-dimensional Brillouin zone for a (001) orientated surface. (C,D) BSF and FS of $(\text{Ba}_{0.6}\text{K}_{0.4})\text{Fe}_2\text{As}_2$ calculated on the basis of LDA. (E,F) Corresponding BSF and FS of $(\text{Ba}_{0.6}\text{K}_{0.4})\text{Fe}_2\text{As}_2$ calculated on the basis of LDA + DMFT. The blue lines always indicate the path chosen for the presented band structure.

meaning topological changes in the FS which mark the onset of superconductivity^{19,32}. Also a rather puzzling change in the intensity distribution at neighboring $\bar{\Gamma}$ points is known⁶. Until now there is no theoretical work which would explain all the salient features of the ARPES spectra of $(\text{Ba}_{1-x}\text{K}_x)\text{Fe}_2\text{As}_2$. Our theoretical work is supported by the new bulk sensitive ARPES measurements done in the soft X-ray regime using various linear polarisation of the light.

Results

Impact of correlations on the electronic structure. The crystal structure of $(\text{Ba}_{0.6}\text{K}_{0.4})\text{Fe}_2\text{As}_2$ is shown in Fig. 1(A), with the corresponding Brillouin zone (BZ) and its two-dimensional counterpart for a (001) orientated surface given in Fig. 1(B). The electronic structure is represented by means of the Bloch spectral function (BSF), which has the significant advantage that in the presented approach all disorder effects induced through substitution are fully accounted for (for more details see review³³). The LDA based band structure is shown in Fig. 1(C) with the corresponding Fermi surface (FS) cut shown in Fig. 1(D). The topology of this FS cut fails to explain the Fermi surface seen by ARPES⁶. It can neither reproduce the well-known propeller-like features at the \bar{X} point, nor it can explain the flower-like topology at $\bar{\Gamma}$ ⁶. Thus, the applied LDA approach is insufficient to deal accurately with these prominent features. To account for the necessary correlation effects fully self-consistently, we have applied subsequent LDA + DMFT calculations. We used for Fe an averaged on-site Coulomb interaction $U = 3.0$ eV and an exchange interaction $J = 0.9$ eV, which are commonly used for the iron pnictides^{10,34,35}. The impact of correlation effects represented by the DMFT on the band structure in Fig. 1(E) and on the Fermi surface cut in Fig. 1(F) is tremendous. We see strong renormalization of the d_{xy} and $d_{xz/yz}$ bands, in agreement with other literature¹⁶. However, most prominent are the changes around the X point where a significant upwards shift of the bands towards the Fermi level (E_F) leads to the hole and electron pockets responsible for the appearance of the propeller like topology at \bar{X} in agreement with experimental ARPES data⁶⁻⁸. Note, that a similar upwards shift at X of around 0.1 eV was also observed by Werner *et al.*¹⁰, although the qualitative agreement of our results with experiment seems slightly better. The X point in our notation corresponds to the M point in the notation used by Werner *et al.*¹⁰. In comparison, Werner *et al.* used a frequency dependent screening which leads to strong incoherence. However, based on suggestions by Tomczak *et al.*¹⁵ and because we look only at energies close to the Fermi level it seems like an acceptable approximation to use a static coulomb interaction U . In particular, we are able to fully account for the chemical disorder of the K-doped compound in terms of the coherent potential approximation (CPA) which seems to be more relevant for the problem at hand. Considering the LDA band structure from Fig. 1(C) one can already see strong band broadening for the hole band of interest at X. Thus, incoherence due to disorder effects is strongest for explicitly this band and it would be invalid to neglect this. Consequently, strong incoherence at X for the LDA + DMFT band structure makes it difficult to resolve the exact band shape. Based on the ARPES data of Zabolotnyy *et al.*⁶ the electron pocket and the hole pocket at X should hybridize even more than in the presented calculations. This effect is covered in our results due to the strong incoherence and it might be also a shortcoming of the applied FLEX DMFT solver.

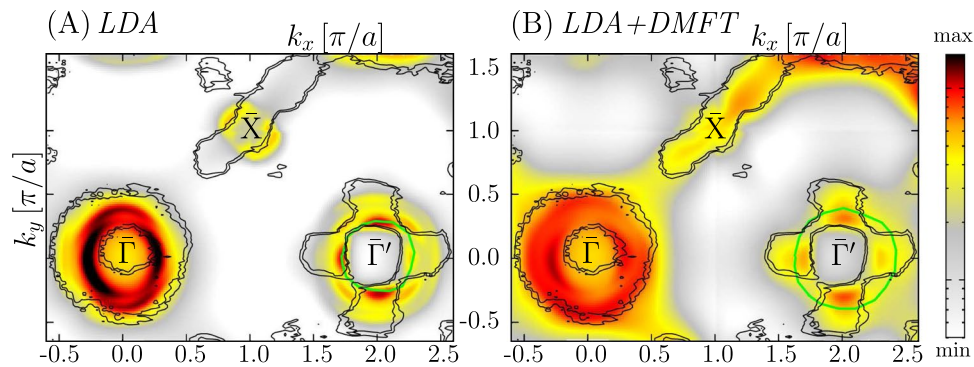


Figure 2. Fermi surfaces cuts of $(\text{Ba}_{0.6}\text{K}_{0.4})\text{Fe}_2\text{As}_2$ for $h\nu = 75$ eV as seen by one step model ARPES calculations for (A) LDA and (B) LDA + DMFT. The overlay of black isolines always corresponds to experimental ARPES data taken with permission from Zabolotnyy *et al.*⁶. The green solid lines are guides for the eyes to indicate surface state related features.

Still, the consequence of this phenomenon is a topological change in the FS contour, indicating a so-called Lifshitz transition which is crucial for the emergence of superconductivity^{19,32}. This Lifshitz transition was already discussed for high K concentrations ($x \approx 0.9$) within a LDA framework^{19,31}. The present as well as previous experimental work^{6–8}, however, clearly shows the emergence of the petal topology around \bar{X} already for optimal doping ($x \approx 0.4$). Based on the latter discussion and by comparing Fig. 1(C,D) one can see that the origin of the Lifshitz transition at lower K concentrations is fully controlled by correlation effects accounted for by the applied LDA + DMFT approach. Consequently, the Lifshitz transition can also qualitatively explain the breakdown of magnetic order for $(\text{Ba}_{1-x}\text{K}_x)\text{Fe}_2\text{As}_2$, which takes place at low doping ratios $x < 0.4$ ²⁹, as it destroys the nesting condition³⁶. More details showing the clear dependence on the Coulomb interaction U are found in the Supplemental Material. Although, the applied self-consistent DMFT approach has brought important new insights on the topology around \bar{X} , it is not able to reproduce the flower-like intensity distribution observed at $\bar{\Gamma}'$ compared to $\bar{\Gamma}$.

Impact of ARPES response effects. To understand this flower-like feature, additional calculations based on the one-step model of ARPES have been done, accounting for the experimental geometry^{37,38} including surface effects as well as matrix element effects. These calculations were performed using the LDA potentials and within the LDA + DMFT framework. The corresponding spectroscopic Fermi surface cuts obtained from the one-step model ARPES calculations of $(\text{Ba}_{0.6}\text{K}_{0.4})\text{Fe}_2\text{As}_2$ for $h\nu = 75$ eV are shown in Fig. 2(A) for LDA and in Fig. 2(B) for the LDA + DMFT calculations. For comparison we show in both pictures the original experimental data from Zabolotnyy *et al.*⁶ as an overlay of black isolines, measured at $h\nu = 80$ eV. Please note that the theoretical photon energy of $h\nu = 75$ eV corresponds to the experimental value of $h\nu = 80$ eV and it has been found by the theoretical k_z scan (not shown here). This is typical shift in the energy of final time reversal LEED state and it is due to the limitations of the density functional theory.

The ARPES calculations based on the LDA + DMFT shown in Fig. 2(B) reveal very good agreement with the experimental data, concerning the Lifshitz transition induced propeller structure at \bar{X} (only one part of the propeller is clearly visible for the chosen light polarization as was also found in experiment⁶) indicating that the previously discussed electronic structure is correctly reproduced. Furthermore, we obtain good agreement concerning the flower intensity distribution at $\bar{\Gamma}'$. It should be stressed that there is no alternation of this circle and flower topology around $\bar{\Gamma}$ and $\bar{\Gamma}'$, respectively, with alternating Γ and Z points in the k_z direction by changing $h\nu$. Thus, the origin of this interesting topology is not connected to the alternation between Γ and Z points of the bulk Brillouin zone.

The general appearance of two different shapes between $\bar{\Gamma}$ and $\bar{\Gamma}'$ can be explained by the structure factor and the light polarization in terms of a 1-Fe or 2-Fe cell, as discussed by e.g. Moreschini *et al.*³⁹ or Lin *et al.*⁴⁰. Note, that a correct treatment of the phase difference between two atoms of a unit cell and the light polarization is by construction included in the one-step model of photoemission (see ref. 41), thus the theory can sufficiently account for this. However, we find that at the same time other effects can contribute, in order to obtain this flower topology in agreement with experiment. As can be seen from Fig. 2(A) the intensity distribution at $\bar{\Gamma}'$ has a four-fold rotational symmetry, although the flower-like topology is not adequately reproduced compared to the result in Fig. 2(B). This difference cannot be explained without additional contributions.

It is known that the intensity distributions in ARPES might change for neighboring Brillouin zones between $\bar{\Gamma}$ and $\bar{\Gamma}'$ due to matrix element effects, however, such a strong change in the intensity distribution as seen in experiment⁶ and reproduced in Fig. 2(B) is rather uncommon and unexpected. Yet, it is also known that the influence of matrix element effects can be enhanced in the vicinity of surface related states. Surface phenomena can be investigated by the applied method, as it is explained in more detail in ref. 42. In particular, we have recently shown that surface states have a significant influence on the ARPES spectra of Co-doped BaFe_2As_2 ²². Subsequently, one is able to identify in the spectra of $(\text{Ba}_{0.6}\text{K}_{0.4})\text{Fe}_2\text{As}_2$ surface resonance states which wave functions have bulk Bloch asymptotic behavior and exhibit a strong resonance at the vicinity of the surface. This means such surface resonances can show a k_z dispersion and they can be observed also for comparably high photon energies. The positions of these ring-shaped surface resonances is marked with solid green lines as an

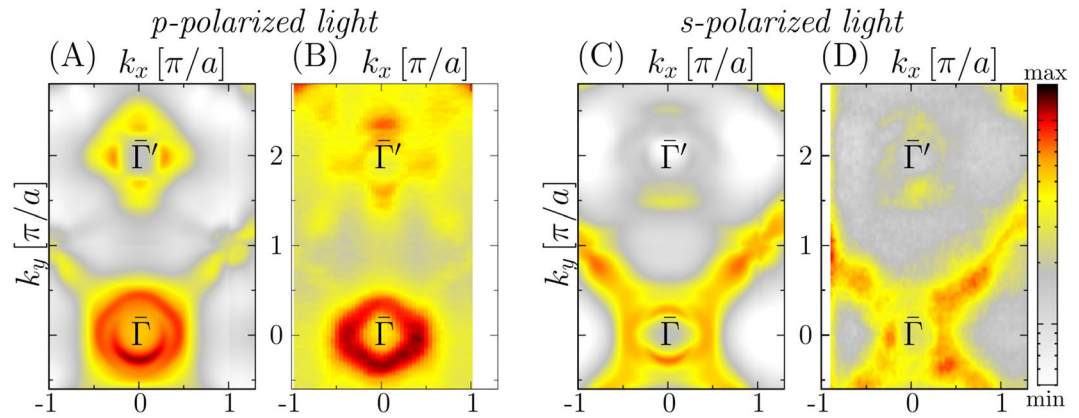


Figure 3. Fermi surface cuts of $(\text{Ba}_{0.6}\text{K}_{0.4})\text{Fe}_2\text{As}_2$ for (A,C) $h\nu = 425$ eV ARPES calculation using LDA + DMFT and (B,D) $h\nu = 430$ eV experimental data. The incoming light was either (A,B) p -polarized or (C,D) s -polarized.

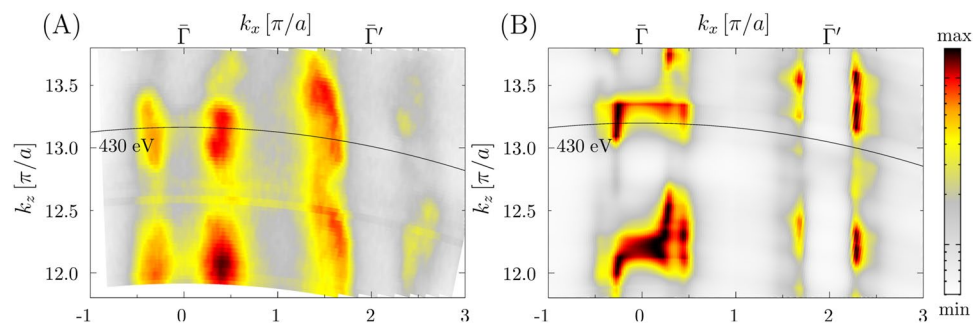


Figure 4. Experimental (A) and theoretical (B) k_z scans with p -polarized light for $(\text{Ba}_{0.6}\text{K}_{0.4})\text{Fe}_2\text{As}_2$. Clearly visible is the k_z dispersion at $\bar{\Gamma}$ and $\bar{\Gamma}'$. The photon energy of $h\nu = 430$ eV and $h\nu = 392$ eV are marked with a black line.

overlay in Fig. 2. For LDA in Fig. 2(A) one can see that this surface resonance is compressed and thus its influence on the intensity distribution at $\bar{\Gamma}'$ is less significant. In comparison, the surface resonance is shifted for the LDA + DMFT calculation in Fig. 2(B) where it cuts precisely through the clearly visible petals of the flower topology, affecting the intensity distribution at this position. We believe, that these contributions from the surface resonances can add up to the commonly discussed explanation based on the 1-Fe/2-Fe scheme, giving finally an overall good agreement with experimental data.

Bulk sensitive ARPES experiments. Additional bulk sensitive soft-X-ray photoemission measurements for $h\nu = 430$ eV were performed for samples of $(\text{Ba}_{0.6}\text{K}_{0.4})\text{Fe}_2\text{As}_2$. In the first surface Brillouin zone, this photon energy sets k_z to the Γ point of the bulk one. The resulting spectra are presented in Fig. 3(B,D) for p -polarized and s -polarized light, respectively. Notably, for p -polarized light the flower shaped topology at $\bar{\Gamma}'$ is enhanced in intensity while for s -polarized light the propeller topologies at \bar{X} are enhanced. Corresponding calculations for $h\nu = 425$ eV are presented in Fig. 3(A,C), which show very good agreement with the experimental data concerning the relevant topologies and the polarization dependence. Thus, these experiments are fully in line with the argumentation of this work so far and further validate our results. Additional extended Fermi surface cuts for higher Brillouin zones can be found in the Supplemental Material.

Furthermore, the experimental and theoretical k_z scan are shown in Fig. 4 with the photon energy of $h\nu = 430$ eV explicitly marked with the black line. This shows clearly the k_z dispersion and thus the strong 3D character of the iron pnictides.

Effective masses derived from ARPES spectra. Finally, the electronic structure derived from BSF calculations and ARPES calculations was used to analyze the effective masses which are of great actual interest for the iron pnictides^{11,13,15,16}. The results for the mass ratios of the inner and outer hole pockets around $\bar{\Gamma}$ in the Γ Z direction and additionally for the hole pocket at X in the Γ X direction are summarized in Table 1. More details are found in the Supplemental Material.

As it is commonly done, all values of m^* are normalized to the LDA value m_{LDA}^* deduced from the ground state BSF. First, we consider the mass enhancement at $\bar{\Gamma}$ only, where the influence of DMFT on the band dispersion can be seen for the BSF, with an average mass enhancement of 2.15 (meaning an average over inner and outer hole pocket $\bar{\Gamma}$), being in good agreement with literature (e.g. 2.04 for KFe_2As_2 ¹⁶). Of more interest is the apparent mass enhancement deduced from the ARPES calculations compared to the BSF band dispersion. The difference is

$\frac{m_{\text{DMFT}}^*}{m_{\text{LDA}}^*}$	BSF	ARPES 75 eV	ARPES 425 eV
Inner pocket Γ	2.59	3.66	2.94
Outer pocket Γ	1.70	2.28	1.98
Hole pocket X	1.43	1.56	1.58

Table 1. Ratio of effective masses of m_{DMFT}^* to m_{LDA}^* for the Bloch spectral function (BSF) ground state calculations as well as for the ARPES calculations with $h\nu = 75$ eV and 425 eV, respectively. The values correspond to the inner and outer hole pockets around Γ showing strong k_z dispersion and to the hole pocket at X showing weak k_z dispersion.

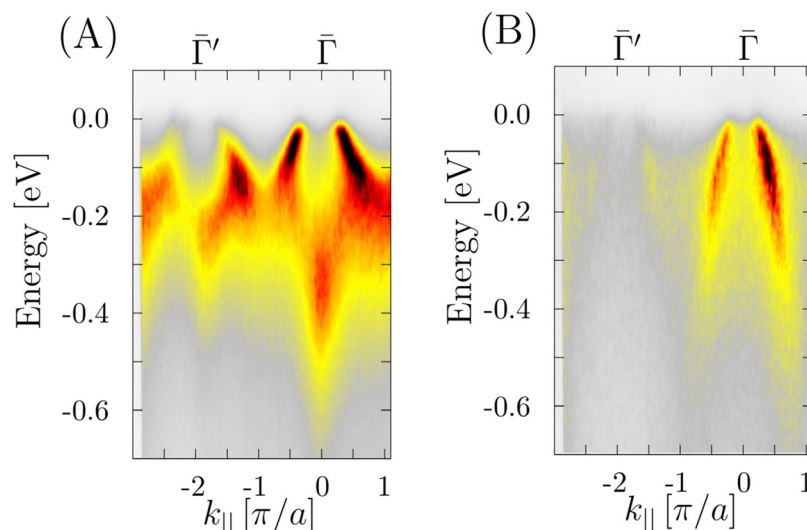


Figure 5. Experimental band dispersions of $(\text{Ba}_{0.6}\text{K}_{0.4})\text{Fe}_2\text{As}_2$ along the ΓZ line of the Brillouin zone measured at $h\nu = 425$ eV with (A) p-polarization, and (B) s-polarization selecting, respectively, the symmetric and antisymmetric d-states.

attributed to the fact that the calculated ARPES spectra include not only the correlation effects of DMFT but also final state effects which, as explained below, modify the ARPES spectral shape. On the experimental side, such an apparent mass enhancement has already been observed in ARPES for e.g. the BaFe_2As_2 parent compound and connected with the k_z dispersion of the valence states¹¹. The apparent mass enhancement is given by the fact that the ARPES response of the 3D valence states is formed by averaging of their matrix-element weighted k_z dispersion over an interval of the intrinsic final state k_z broadening (Δk_z) determined by the photoelectron mean free path λ ^{43,44}.

As illustrated in Fig. S5 in the Supplemental Material, near the extremes of the valence band k_z dispersions this averaging effectively shifts the ARPES peaks from true k_z dispersions into the band interior (for detailed physical picture see ref. 43). In k_{\parallel} dependent ARPES intensities this shift is seen as an apparent bandwidth reduction and corresponding mass enhancement. One can expect a stronger influence of these final state effects at lower photon energies where Δk_z is larger due to a smaller λ . Indeed, our calculations find significant differences in the mass enhancement at Γ depending on $h\nu$. For the low $h\nu$ of 75 eV ($\Delta k_z = 0.2779 \text{ \AA}^{-1}$, which makes about 30% of the perpendicular BZ dimension) we find an average mass enhancement of 2.97 at Γ which is higher than the value of 2.15 obtained from the BSF. The significantly higher $h\nu$ of 425 eV ($\Delta k_z = 0.1228 \text{ \AA}^{-1}$) increases λ and concomitantly improves the k_z definition. The final state effects have therefore a less pronounced contribution, reducing the average mass enhancement at Γ to 2.46. This is true for almost all bands in the iron pnictides as they are 3D materials with most bands showing a clear k_z dispersion. One of the rare exceptions for the $(\text{Ba}_{1-x}\text{K}_x)\text{Fe}_2\text{As}_2$ compound is the hole pocket at X which has almost 2D character and shows hardly a k_z dispersion as can be seen in Fig. 1(C,E) for the path ΓXZ . In such a case one would expect significantly less influence of the final-state effects and indeed, Table 1 shows that the apparent mass enhancement for high and low $h\nu$ at X is almost the same and very similar to the BSF mass enhancement. This finally explains discrepancies in the observed mass enhancement for the iron pnictides. To reduce these $h\nu$ dependent deviations of the ARPES response from the true 3D valence bands, we justify the use of higher $h\nu$ in the soft-X-ray regime to improve the k_z definition.

Finally, Fig. 5 shows the experimental band dispersions measured at $h\nu = 425$ eV which correspond to ΓZ direction in the bulk Brillouin zone. In the soft-X-ray ARPES experimental geometry^{37,38} s-polarized light excites the dxz , dx^2-y^2 and dz^2 states symmetric relative to the ΓZM plane of the Brillouin zone, and p-polarized light the dyz and dxy states antisymmetric relative to ΓZM ⁴⁵. On top of these dipole selection rules, relative intensities of

	$\frac{m_{\text{EXP}}^*}{m_{\text{LDA}}^*}$	$\frac{m_{\text{EXP}}^*}{m_{\text{DMFT}}^*}$	$\frac{m_{\text{EXP}}^*}{m_{\text{ARPES DMFT}}^*}$
Inner pocket Γ	3.50	1.35	1.19
Outer pocket Γ	2.26	1.56	1.41

Table 2. Ratio of effective masses of the experimental data with respect to the various theoretical values. Experimental data are taken from Fig. 5. Values for m_{DMFT}^* and m_{LDA}^* are taken from the ground state BSF and $m_{\text{ARPES DMFT}}^*$ corresponds to the calculated one step model of photoemission spectra at $h\nu = 425$ eV as presented in Table 1.

these bands alters between the Γ and Z points because of the matrix element effects discussed above. From Fig. 5 we have extracted the corresponding experimental values of the effective masses for the inner and outer pockets at the Γ point. In the Table 2 we summarize the results for band renormalization between the experimental data and the corresponding ground state LDA and DMFT bands. Here we obtained an average value of $\frac{m_{\text{EXP}}^*}{m_{\text{LDA}}^*} = 3.05$ which, as we discussed before, due to reduced value of k_z broadening in the soft X-ray regime can be taken as a reference value for future studies. As a last point we show in the third column of Table 2 the ratio of the experimental value m_{EXP}^* to the theoretical value $m_{\text{ARPES DMFT}}^*$ obtained from the one-step model ARPES calculations. In the ideal case one would expect to reach a value of one. Due to the approximations we used for the DMFT calculations, we obtain an average value of 1.3. This discrepancy is mainly due to the use of perturbative nature of the DMFT solver (FLEX), and as it was discussed recently by Werner *et al.*¹⁰ additional dynamical screening effects are missing in our approach. However, by comparing the spectra calculated within one step model of photoemission with the state of the art experimental data, we are able to give a quantitative measure of the theoretical approximations used here. This would not be possible if we had used only ground state calculations as demonstrated in the second column of Table 2.

Conclusions

In conclusion, the presented LDA + DMFT + ARPES study is the first that quantitatively matches the theoretical description with the experimental ARPES data on the paradigm high-temperature superconductor ($\text{Ba}_{0.6}\text{K}_{0.4}$) Fe_2As_2 . These results enables a better physical understanding of the unconventional superconductivity in pnictides and will be of great importance for future studies on similar systems. In particular, the origin of the Lifshitz transition in $(\text{Ba}_{1-x}\text{K}_x)\text{Fe}_2\text{As}_2$, crucial for its superconductivity, is identified as fully controlled by electron correlation effects. Furthermore, we have shown that due to the inherently 3D nature of the iron pnictides their ARPES response is significantly influenced by final state effects, shifting the spectral peaks from the true quasiparticle valence bands. Their mass enhancement apparent in the ARPES spectra is then different from the true value and, moreover, will depend on the photon energy. Thus, the mass renormalization observed in previous ARPES works on iron pnictides is not an entirely intrinsic property of the quasiparticle valence band structure or spectral function, but has a significant contribution due to a peculiarity of the photoemission process extrinsic to the true valence band properties.

Methods

Computational method. Within the present work, the multiple scattering Korringa-Kohn-Rostoker-Green function (KKR-GF) method was applied which allows to deal simultaneously with all mentioned spectroscopic and many-body aspects. All calculations have been performed within the fully relativistic four component Dirac formalism^{33,46}, accounting this way for all effects induced by spin-orbit coupling. Disorder effects are dealt with by means of the coherent potential approximation (CPA)^{19,22,47}. ARPES calculations are based on the one-step model of photoemission in its spin density matrix formulation using the experimental geometry^{48,49}. Thus, the theory accounts for effects induced by the light polarization, matrix-element effects, final state effects and surface effects. To account for correlation effects fully self-consistently (concerning charge as well as self energy) the LDA + DMFT method using a FLEX solver was applied⁵⁰. For Fe an averaged on-site Coulomb interaction $U = 3.0$ eV and an exchange interaction $J = 0.9$ eV were applied. In the Supplemental Material calculations for different values of U are shown. The lattice constants of the tetragonal cell of $(\text{Ba}_{0.6}\text{K}_{0.4})\text{Fe}_2\text{As}_2$ were taken from experimental data²⁹.

ARPES experiments. New ARPES experiments in the soft-X-ray photon energy ($h\nu$) range above 400 eV were performed at the ADDRESS beamline of the Swiss Light Source synchrotron facility^{37,38}. By using higher $h\nu$ compared to the conventional ultraviolet ARPES, higher bulk sensitivity is achieved due to an increase of the photoelectron mean free path λ as expected from the well-known “universal curve”. Crucial for 3D materials like the iron pnictides is that the increase of λ results, by the Heisenberg uncertainty principle, in a sharp intrinsic definition of the momentum k_z perpendicular to the surface⁴³. As explained in the paper, the latter becomes important for the correct evaluation of the true valence band dispersions and effective masses.

In particular, bulk sensitive soft-X-ray photoemission measurements for $h\nu = 425$ eV were performed for *in-situ* cleaved samples of $(\text{Ba}_{0.6}\text{K}_{0.4})\text{Fe}_2\text{As}_2$ at a temperature of around 12 K and with an overall energy resolution of around 70 meV.

References

- Mazin, I. I. & Johannes, M. D. A key role for unusual spin dynamics in ferropnictides. *Nature Physics* **5**, 141–145 (2009).
- Mazin, I. I., Singh, D. J., Johannes, M. D. & Du, M. H. Unconventional superconductivity with a sign reversal in the order parameter of lafeaso1-xfx. *Phys. Rev. Lett.* **101**, 057003 (2008).
- Mazin, I. I., Johannes, M. D., Boeri, L., Koepfner, K. & Singh, D. J. Problems with reconciling density functional theory calculations with experiment in ferropnictides. *Phys. Rev. B* **78**, 085104 (2008).
- Kuroki, K., Usui, H., Onari, S., Arita, R. & Aoki, H. Pnictogen height as a possible switch between high- T_c nodeless and low- T_c nodal pairings in the iron-based superconductors. *Phys. Rev. B* **79**, 224511 (2009).
- Graser, S. *et al.* Spin fluctuations and superconductivity in a three-dimensional tight-binding model for bafe₂as₂. *Phys. Rev. B* **81**, 214503 (2010).
- Zabolotnyy, V. B. *et al.* π - π electronic order in iron arsenide superconductors. *Nature* **457**, 569–572 (2009).
- Evtushinsky, D. V. *et al.* Momentum-resolved superconducting gap in the bulk of ba1-xkxfe2as2 from combined arpes and sr measurements. *New Journal of Physics* **11**, 055069 (2009).
- Evtushinsky, D. V. *et al.* Propeller-like low temperature fermi surface of ba1-xkxfe2as2 from magnetotransport and photoemission measurements. *J. Phys. Soc. Japan* **80**, 023710 (2011).
- Evtushinsky, D. V. *et al.* Momentum dependence of the superconducting gap in ba1-xkxfe2as2. *Phys. Rev. B* **79**, 054517 (2009).
- Werner, P. *et al.* Satellites and large doping and temperature dependence of electronic properties in hole-doped bafe2as2. *Nature Physics* **8**, 331–337 (2012).
- Brouet, V. *et al.* Large temperature dependence of the number of carriers in co-doped bafe2as2. *Phys. Rev. Lett.* **110**, 167002 (2013).
- Evtushinsky, D. V. *et al.* Strong electron pairing at the iron 3d_{xz} orbitals in hole-doped bafe2as2 superconductors revealed by angle-resolved photoemission spectroscopy. *Phys. Rev. B* **89**, 064514 (2014).
- Brouet, V. *et al.* Arpes view of orbitally resolved quasiparticle lifetimes in iron pnictides. *Phys. Rev. B* **93**, 085137 (2016).
- Thirupathiah, S. *et al.* Effect of impurity substitution on band structure and mass renormalization of the correlated fete_{0.5}se_{0.5} superconductor. *Phys. Rev. B* **93**, 205143 (2016).
- Tomczak, J. M., van Schilfgaarde, M. & Kotliar, G. Many-body effects in iron pnictides and chalcogenides: Nonlocal versus dynamic origin of effective masses. *Phys. Rev. Lett.* **109**, 237010 (2012).
- Backes, S., Guterding, D., Jeschke, H. O. & Valent, R. Electronic structure and de haas–van alphen frequencies in kfe2as2 within lda + dmft. *New Journal of Physics* **16**, 083025 (2014).
- Berlijn, T., Lin, C.-H., Garber, W. & Ku, W. Do transition-metal substitutions dope carriers in iron-based superconductors? *Phys. Rev. Lett.* **108**, 207003 (2012).
- Wang, L. *et al.* Effects of disordered ru substitution in bafe2as2: Possible realization of superdiffusion in real materials. *Phys. Rev. Lett.* **110**, 037001 (2013).
- Khan, S. N. & Johnson, D. D. Lifshitz transition and chemical instabilities in bakfeas superconductors. *Phys. Rev. Lett.* **112**, 156401 (2014).
- Borisenko, S. V. *et al.* Direct observation of spin-orbit coupling in iron-based superconductors. *Nature Physics* **12**, 311–317, Letter (2016).
- Wang, X.-P. *et al.* Orbital characters determined from fermi surface intensity patterns using angle-resolved photoemission spectroscopy. *Phys. Rev. B* **85**, 214518 (2012).
- Derondeau, G., Braun, J., Ebert, H. & Minár, J. Theoretical study on the anisotropic electronic structure of antiferromagnetic bafe2as2 and co-doped ba(fe1-xcox)2as2 as seen by angle-resolved photoemission. *Phys. Rev. B* **93**, 144513 (2016).
- Yin, Z. P., Haule, K. & Kotliar, G. Magnetism and charge dynamics in iron pnictides. *Nature Physics* **7**, 294–297 (2011).
- Zhang, Y.-Z., Lee, H., Opahle, I., Jeschke, H. O. & Valent, R. Importance of itinerancy and quantum fluctuations for the magnetism in ironpnictides. *J. Phys. Chem. Sol.* **72**, 324–328 (2011).
- Ferber, J., Jeschke, H. O. & Valent, R. Fermi surface topology of lafepo and lifep. *Phys. Rev. Lett.* **109**, 236403 (2012).
- Xu, N. *et al.* Electronic band structure of baco2as2: A fully doped ferropnictide analog with reduced electronic correlations. *Phys. Rev. X* **3**, 011006 (2013).
- Yin, Z. P., Haule, K. & Kotliar, G. Spin dynamics and orbital-antiphase pairing symmetry in iron-based superconductors. *Nature Physics* **10**, 845–850 (2014).
- Rotter, M. *et al.* Spin-density-wave anomaly at 140 k in the ternary iron arsenide bafe2as2. *Phys. Rev. B* **78**, 020503 (2008).
- Rotter, M., Tegel, M. & Johrendt, D. Superconductivity at 38 k in the iron arsenide ba1-xkxfe2as2. *Phys. Rev. Lett.* **101**, 107006 (2008).
- Nakayama, K. *et al.* Universality of superconducting gaps in overdoped ba0.3k0.7fe2as2 observed by angle-resolved photoemission spectroscopy. *Phys. Rev. B* **83**, 020501 (2011).
- Xu, N. *et al.* Possible nodal superconducting gap and lifshitz transition in heavily hole-doped ba0.1k0.9fe2as2. *Phys. Rev. B* **88**, 220508 (2013).
- Liu, C. *et al.* Evidence for a lifshitz transition in electron-doped iron arsenic superconductors at the onset of superconductivity. *Nature Physics* **6**, 419–423 (2010).
- Ebert, H., Ködderitzsch, D. & Minár, J. Calculating condensed matter properties using the kkr–green’s function method – recent developments and applications. *Rep. Prog. Phys.* **74**, 096501 (2011).
- Ferber, J., Foyevtsova, K., Valent, R. & Jeschke, H. O. Lda + dmft study of the effects of correlation in lifeas. *Phys. Rev. B* **85**, 094505 (2012).
- Aichhorn, M. *et al.* Dynamical mean-field theory within an augmented plane-wave framework: Assessing electronic correlations in the iron pnictide lafeaso. *Phys. Rev. B* **80**, 085101 (2009).
- Kim, M. G. *et al.* Effects of transition metal substitutions on the incommensurability and spin fluctuations in bafe2as2 by elastic and inelastic neutron scattering. *Phys. Rev. Lett.* **109**, 167003 (2012).
- Strocov, V. N. *et al.* Soft-x-ray arpes facility at the adress beamline of the sls: concepts, technical realisation and scientific applications. *Synchrotron Radiat. News* **21**, 32–44 (2014).
- Strocov, V. N. *et al.* Soft-x-ray arpes at the swiss light source: From 3d materials to buried interfaces and impurities. *Synchrotron Radiat. News* **27**, 31–40 (2014).
- Moreschini, L. *et al.* Consequences of broken translational symmetry in fesexte1-x. *Phys. Rev. Lett.* **112**, 087602 (2014).
- Lin, C.-H. *et al.* One-fe versus two-fe brillouin zone of fe-based superconductors: Creation of the electron pockets by translational symmetry breaking. *Phys. Rev. Lett.* **107**, 257001 (2011).
- Lev, L. L. *et al.* Fermi surface of three-dimensional la1-xsrxmno3 explored by soft-x-ray arpes: Rhombohedral lattice distortion and its effect on magnetoresistance. *Phys. Rev. Lett.* **114**, 237601 (2015).
- Braun, J. & Donath, M. Contest between surface resonances and surface states at 3d ferromagnets. *Europhys. Lett.* **59**, 592 (2002).
- Strocov, V. N. Intrinsic accuracy in 3-dimensional photoemission band mapping. *J. Electron. Spectrosc. Relat. Phenom.* **130**, 65–78 (2003).
- Kurtz, R. L., Browne, D. A. & Mankey, G. J. Final state effects in photoemission studies of fermi surfaces. *J. Phys.: Cond. Mat.* **19**, 355001 (2007).
- Hajiri, T. *et al.* Three-dimensional electronic structure and interband nesting in the stoichiometric superconductor lifeas. *Phys. Rev. B* **85**, 094509 (2012).

46. Ebert, H. *et al.* *The Munich SPR-KKR package*, version 6.3, <http://olymp.cup.uni-muenchen.de/ak/ebert/SPRKKR>. <http://olymp.cup.uni-muenchen.de/ak/ebert/SPRKKR> (2012).
47. Derondeau, G., Polesya, S., Mankovsky, S., Ebert, H. & Minár, J. Theoretical investigation of the electronic and magnetic properties of the orthorhombic phase of $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$. *Phys. Rev. B* **90**, 184509 (2014).
48. Minár, J., Braun, J. & Ebert, H. Correlation effects in magnetic materials: An ab initio investigation on electronic structure and spectroscopy. *J. Electron. Spectrosc. Relat. Phenom.* **189**, 129 (2013).
49. Braun, J. *et al.* Exceptional behavior of d-like surface resonances on $\text{W}(110)$: the one-step model in its density matrix formulation. *New Journal of Physics* **16**, 015005 (2014).
50. Minár, J. Correlation effects in transition metals and their alloys studied using the fully self-consistent kkr-based lsd + dmft scheme. *J. Phys.: Cond. Mat.* **23**, 253201 (2011).

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Author Contributions

G.D., V.N.S. and J.Mi. wrote the manuscript. F.B., V.A.R., M.K., T.S., J.Ma. and V.N.S. performed the experiments and analyzed the data. G.D. performed the calculations. G.D., F.B., J.B., M.S., H.D., H.E., V.N.S. and J.Mi. participated at the discussions. J.Mi. supervised the theoretical part, V.N.S. supervised the experimental part.

Additional Information

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