Orbital weight redistribution triggered by spin order in the pnictides

M. Daghofer,1 Q.-L. Luo,2 R. Yu,3 D. X. Yao,2,4 A. Moreo,2 and E. Dagotto2

1IFW Dresden, P.O. Box 27 01 16, D-01171 Dresden, Germany
2Department of Physics and Astronomy, University of Tennessee, Knoxville, TN 37996-1200 and Materials Science and Technology Division, ORNL, Oak Ridge, TN 37831-6032, USA
3Department of Physics & Astronomy, Rice University, Houston, Texas 77005, USA
4School of Physics and Engineering, Sun Yat-Sen University, Guangzhou 510275, China

(Dated: May 28, 2010)

The one-particle spectral function and its orbital composition are investigated in a three-orbital model for the undoped parent compounds of the iron-based superconductors. In the realistic parameter regime, where results fit experimental data best, it is observed that the magnetization in the xz and yz orbitals are markedly different and the Fermi surface presents mostly xz character, as recently observed in photoemission experiments [T. Shimojima et al., Phys. Rev. Lett. 104, 057002 (2010)]. Since the ferro-orbital order in this regime is at most a few percent, these results are mainly driven by the magnetic order. An analogous analysis for a five-orbital model leads to similar conclusions.

PACS numbers: 71.10.-w, 71.10.Fd, 74.20.Rp

Introduction. In contrast to the cuprate superconductors, more than one band cross the chemical potential in the pnictide superconductors. According to density functional theory (DFT) calculations, most of the spectral weight at the Fermi surface (FS) arises from two of the iron d-orbitals, namely the xz and yz orbitals (inset of Fig. 1), which are degenerate in the high-temperature tetragonal phase. Lowering the temperature, the undoped parent compounds undergo a structural as well as a magnetic phase transition to an orthorhombic phase with antiferromagnetic (AF) order with wavevector $(\pi, 0)\xi$ (inset of Fig. 1). This spin order breaks the rotational symmetry of the original tetragonal lattice. In this regime, scanning tunneling microscopy and resistivity measurements have indicated that the electronic system presents symmetry-breaking properties that far exceed the relatively modest difference of the lattice constants before and after the transition.

To rationalize these results, orbital ordering has been suggested to occur together with the magnetic ordering, lifting the degeneracy between the $xz$ and $yz$ orbitals before and after the transition. Finally, ARPES results can be fitted by DFT if the magnetic moment in the calculation is artificially suppressed towards experimentally observed values, and these calculations then yield a far smaller rearrangement of the two hole pockets, indicating that FO order and the ordered magnetic moment may be linked.

In this paper, the spectral function of a three-orbital model is investigated with an emphasis on features related to orbital polarization effects. This model allows for the stabilization of a regime with small ordered magnetic moments by selecting intermediate values for the Hubbard repulsion. One-particle spectral functions calculated in this regime have already been shown to be qualitatively similar to experimental ones including the presence of small hole- or electron-like extra pockets near the electron and hole pockets of the uncorrelated bands. We focus here on the orbital composition of the Fermi surface, which was not analyzed in those previous investigations. It will be shown that the FS has predominantly xz character, similar to recent experimental results obtained with Laser-ARPES. In this regime

![FIG. 1: (Color online) Density of states of the xz and yz orbitals in the (π, 0)-AF phase of the three-orbital model at U = 0.7 and J = U/4. For these values of U and J, the orbital densities are $n_{xz} = n_{xz,\uparrow} + n_{xz,\downarrow} = 1.590 \approx n_{yz} = 1.586$, and the magnetizations $m_{xz} = n_{xz,\uparrow} - n_{xz,\downarrow} = 0.04 \ll m_{yz} = 0.15$. For U = 0, N(ω) is identical for both orbitals (solid line). A Gaussian broadening with $\sigma = 0.005$ was used. The inset illustrates the (π, 0)-AF order considered here and the xz, yz, and xy orbitals (left to right).](image-url)
with the polarized FS, the orbital magnetizations show a substantial difference between the \(yz\) and \(xz\) orbitals, but there is hardly any static orbital order. Our analysis of the three-orbital model is complemented by a discussion of a five-orbital model where similar results are found. This model admits a regime with moderate FO order of \(\approx 30\%\); the spectral density and FS, however, more closely resemble ARPES results if the FO is at most a few percent and the ordered magnetic moment is small or intermediate.\(^6\)

**Model.** The Hamiltonian studied here consists of the kinetic energy (tight binding) previously used for three\(^8\) or five\(^5\) \(d\) orbitals, as well as the standard onsite Coulomb interaction terms comprised of the intraorbital repulsion \(U\), interorbital repulsion \(U'\), and the \(z\)-component of the Hund’s rule interaction regulated by a coupling \(J\), with \(U = U' + 2J\). The reader is referred to Refs.\(^{8,16}\) for more details. The overall electronic density per site \(n\) is 4 (6) for the three (five) orbital model. The spin-flip and pair-hopping terms, which are by symmetry also part of the onsite interaction, drop out in our previous and current mean-field studies.\(^6\) The interacting Hamiltonian is then treated with a mean-field approximation\(^5\) where we can compare a variety of phases with different magnetic and orbital orders.\(^5\) For a two-orbital model, our method was compared to the Variational Cluster Approximation\(^6\) and found to give similar results.\(^8,12\) As previously reported, small to intermediate Coulomb repulsions \(U \lesssim 1\text{eV}\) stabilize an AF metal in agreement with experiments.\(^5,8\) Approximations beyond mean-field will likely increase the actual values of \(U\) and \(J\) in the realistic regime.

**Results for three orbitals.** Figure 1 shows the \(xz\) and \(yz\) contributions to the density of states, both for the AF metal found at \(U = 0.7\) and \(J = U/4\), and for the uncorrelated system. In agreement with the interpretation given to Laser-ARPES results in Ref.\(^5\), we find that most of the weight at the FS arises from the \(xz\) orbital in the AF state, while both orbitals contribute equally in the tetragonal nonmagnetic state. The total densities in the \(xz\) and \(yz\) orbitals are, however, almost the same \(n_{xz} = 1.590 \approx n_{yz} = 1.586\), i.e., there is (almost) no FO order. Only the states near the chemical potential \(\mu\) are \(xz\)-polarized, and a strong \(yz\) peak at energies \(\approx -50\text{meV}\) approximately compensates for the missing \(yz\)-weight around \(\mu\). This peak comes from the opening of a gap that stabilizes the AF order and that affects mainly the \(yz\) portions of the bands. The system remains metallic, because the \(xz\) orbital does not have a gap around \(\mu\). The stronger impact of the magnetic \((\pi,0)\)-order on the \(yz\) orbital also leads to a larger magnetization for this orbital, with \(m_{yz} = 0.15 \gg m_{xz} = 0.04 \gg m_{xy} = 0.014\). Such a larger value of \(m_{yz}\) has been explained by a larger \(yz\) hopping along the \(x\)-direction\(^5\) but in the present three-orbital model the \(xz\) orbital has the largest hopping amplitude along \(x\).\(^{20}\) While we have observed before a dominant \(m_{xz}\) in the large-\(U\) limit\(^5\) we attribute the relatively large value of \(m_{yz} > m_{xz}\) observed at smaller \(U\) to the orbital character of the electron pockets. The pocket found around \((\pi,0)\) in the uncorrelated model, with mostly \(yz\) character, gets folded into the central hole pockets and forms the gap mentioned above, while the \(xz\) pocket at \((0,\pi)\) is far less affected.\(^{21}\) Since the \(yz\) orbital develops a pseudogap at \(\mu\) and the ungapped \(xz\) orbital consequently determines the states at the Fermi level, the slightly higher resistivity in the ferromagnetic (FM) \(y\)-direction\(^5\) might be due to the fact that the \(xz\) orbital has larger hopping amplitudes in the AF \(x\)-direction.

Figures 2(a) and (b) show the \(xz\) and the \(yz\) contributions to the Fermi surface for the three parameters as in Fig. 1. As mentioned above, and as also reported previously\(^{15}\) most of the FS is given by \(xz\) states, but we also find small features coming from the \(yz\) orbital. These small \(yz\) electron-like pockets, see also Fig. 6(a) in Ref.\(^3\) are similar to \(V\)-shaped features reported in Laser-ARPES\(^5\) and their \(yz\) character does not contradict the experimental findings: the laser spot is expected to catch signals both from \((\pi,0)\) - and \((0,\pi)\)-ordered domains, and the two polarizations pick up either the \(xz\) or the \(yz\) orbital. Since the \(yz\) orbital takes the same role for \((0,\pi)\) that \(xz\) has for \((\pi,0)\), the polarization sensitive to \(xz\) symmetry is expected to find states with \(xz\) character from \((\pi,0)\) domains together with features having what corresponds effectively to ‘\(yz\)’ from the rotated \((0,\pi)\) domains. Similarly, changing the polarization leads to \(yz\) for \((\pi,0)\) plus ‘\(xz\)’ for \((0,\pi)\).\(^{15}\) This situation can be modeled by adding to the \(xz\)-weight at the FS the \(yz\) contributions rotated by 90 degrees, because they stem from domains with rotated AF order. Figures 2(c) and 2(d)
filled four- and two-orbital models. Effects could therefore occur more easily than in the half-model is away from half-filling, and orbital ordering effects grow continuously in this regime and magnetic moment, grows continuously in this regime and inflection point in the magnetization. For small\textit{c}lectronically unchanged orbital densities. As was reported for inflection point in the orbital densities, which coincides well with an inflection point in the magnetization.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig3}
\caption{(Color online) Orbital dependent (a) magnetizations (in \(\mu_B\)) and (b) electronic densities of the five-orbital model varying the strength of the Coulomb repulsion \(U\), at \(J = U/4\). In (a), the total magnetization is also given. The gray area denotes the regime with small ordered magnetic moment and almost no FO order. Its lower boundary is given by the onset of a finite ordered moment. Since orbital order develops more gradually, its upper boundary was defined as the inflection point of the orbital densities, which coincides well with an inflection point in the magnetization.}
\end{figure}

show the expected result for the two polarizations, and one clearly observes the rotation of all features by 90 degrees, as seen in experiments. The rotation would only break down for a hybridized FS that contains substantial contributions from both the \(xz\) and \(yz\) orbitals. Both hole pockets present such a mixture in the uncorrelated bands, but Figs. 2(a) and (b) clearly show that the AF order removes the hybridization and each feature of the FS in the AF phase has (almost) only \(xz\) or \(yz\) character.

\textbf{Results for five orbitals.} A similar analysis was carried out for a five-orbital model and Fig. 3 shows the mean-field results for the magnetizations and densities in the five \(d\)-orbitals, varying the strength of the on-site Coulomb repulsion \(U\). A realistic constant ratio \(J = U/4\) was chosen, and it was checked that a slightly larger or smaller \(J\) does not qualitatively alter our conclusions. For small \(U\), the system remains an uncorrelated metal without any magnetic ordering and practically unchanged orbital densities. As was reported for the other multi-orbital models, AF order starts to develop at a critical value of \(U\), see Fig. 3(a). The staggered magnetization per site, which corresponds to the ordered magnetic moment, grows continuously in this regime and remains smaller than 1.5\(\mu_B\).

Similarly as for the three-orbital model, the five-orbital model is away from half-filling, and orbital ordering effects could therefore occur more easily than in the half-filled four- and two-orbital models. However, again similarly as for the three-orbital model, the first critical \(U\) turns out not to affect the orbital densities as strongly as the magnetization, see Fig. 3(b). The densities only slowly begin to vary after a robust magnetization has set in and for a finite window in \(U\) the difference remains in the low percent range, far smaller than the magnetization. Moreover, the difference in orbital densities is also smaller than the difference in orbital magnetizations with \(m_{yz} > m_{xz}\), due to the \(yz\) band being more strongly gapped around the chemical potential (see the discussion for the three-orbital model above). Only for larger values of \(U\), where the ordered magnetic moment is already quite large, a moderate FO order sets in with \(\approx 30\%\) more electrons in the \(xz\) orbital. Even in this phase, we find that all orbitals are affected to a similar degree, even though the \(xz\) and \(yz\) orbitals, which are degenerate in the uncorrelated case and make up most of the weight at the FS, might \textit{a priori} be expected to be particularly susceptible to symmetry-lowering orbital order.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig4}
\caption{(Color online) One-particle spectral function \(A(k, \omega)\) for (a) the AF metal without orbital order and (b) the AF metal with moderate FO order at \(U = 1.6\). In (a), for \(U = 1.35\) and \(J = U/4\), the total staggered magnetization is \(m_{\text{tot}} = 0.55\mu_B\), and the orbital contributions of the \(xz\) and \(yz\) orbitals are \(m_{xz} = 0.08\mu_B\) and \(m_{yz} = 0.18\mu_B\). The difference in the densities is \(n_{xz} - n_{yz} = 0.032\). In (b), for \(U = 1.6\) and \(J = 0.4\), \(m_{\text{tot}} = 2.52\mu_B\), \(m_{xz} = 0.37\mu_B\), and \(m_{yz} = 0.65\mu_B\); the difference in the densities is \(n_{xz} - n_{yz} = 0.26\). The thin black lines give the uncorrelated bands at \(U = 0\).}
\end{figure}
resulting “shadow” bands of magnetic origin form additional hole-pocket–like features next to the original electron pockets, in agreement with ARPES. Similar to the three-orbital model discussed above, \(xz\)-\(yz\)-hybridization of the hole pockets has given way to a large \(xz\)-polarized central pocket and small satellites with \(yz\) character. If a slightly larger \(U = 1.6\) is chosen, so that the system develops some FO order, see Fig. 3(b), the spectral function changes considerably, as it can be seen in Fig. 4(b), all remnants of the original electron pockets have completely disappeared, there are no longer bands just below the chemical potential around \((\pi, 0)/(0, \pi)\) as seen in ARPES and the features around \((0, 0)\) and \((\pi, 0)\) are far more symmetric to each other than in ARPES. The strong reconstruction into features that resemble neither the uncorrelated bands nor ARPES experiments arises because the interaction is now strong enough to involve the states at \((\pi, \pi)\), located just below the chemical potential at \(U = 0\), in the magnetic order.

Conclusions. In summary, both the three- and five-orbital models have instabilities towards orbitally ordered states, and the instability can be driven by \((\pi, 0)\)-AF order. However, significant orbital order requires a relatively strong onsite Hubbard repulsion \(U\) and goes together with a large ordered magnetic moment and a significant reconstruction of the one-particle bands and FS, see Ref. 8 and Figs. 3 and 4(b). At small to intermediate \(U\), a realistic AF metal with small ordered magnetic moments is found where the densities in the \(xz\) and \(yz\) orbitals differ by at most a few percent.

The orbital magnetization, on the other hand, is far stronger for the \(yz\) orbital [for AF order with ordering vector \((\pi, 0)\)] than it is for \(xz\), which suggests that the orbital degree of freedom strongly couples to the magnetic order. Such a more dynamic picture of the orbital degree of freedom in pnictides is also corroborated by the one-particle density of states, where the states near the Fermi surface have more \(xz\) character than \(yz\), leading to a FS with substantial orbital polarization, even in a regime where FO order is at most a few percent. Another effect of the magnetic order is the breakdown of the hybridization between the \(xz\) and \(yz\) orbitals: while the uncorrelated FS shows features with mixed \(xz\)-\(yz\) character, all features in the correlated FS are either purely \(xz\) or, for some smaller pockets, purely \(yz\), in agreement with Laser ARPES results.

Acknowledgments. This research was sponsored by the NSF grant DMR-0706020, the Division of Materials Science and Engineering, Office of Basic Energy Sciences, U.S. DOE (A.M. and E.D.), and by the Deutsche Forschungsgemeinschaft (DFG) under the Emmy-Noether program. We acknowledge valuable discussions with H. Rosner, P. M. R. Brydon, and K. Koepennik.

---

* Electronic address: M.Daghofer@ifw-dresden.de

1. S. Lebègue, Phys. Rev. B 75, 035110 (2007); D. J. Singh and M.-H. Du, Phys. Rev. Lett. 100, 237003 (2008); G. Xu et al., EPL 82, 67002 (2008); C. Cao, P. J. Hirschfeld, and H.-P. Cheng, Phys. Rev. B 77, 220506 (2008); H.-J. Zhang, G. Xu, X. Dai, and Z. Fang, Chin. Phys. Lett. 26, 017401 (2009); L. Boeri, O. V. Dolgov, and A. A. Golubov, Phys. Rev. Lett. 101, 026403 (2008).

2. J. Dong et al., EPL 83, 27006 (2008); C. de la Cruz et al., Nature 453, 899 (2008); Y. Chen et al., Phys. Rev. B 78, 064515 (2008).

3. T. Chuang et al., Science 327, 181 (2010).

4. J.-H. Chu et al., arXiv:1002.3364 (unpublished).

5. F. Krüger, S. Kumar, J. Zaanen, and J. van den Brink, Phys. Rev. B 79, 054504 (2009).

6. W. Lv, F. Krüger, and P. Phillips, arXiv:1002.3163 (unpublished).

7. C.-C. Lee, W.-G. Yin, and W. Ku, Phys. Rev. Lett. 103, 267001 (2009).

8. M. Daghofer, A. Nicholson, A. Moreo, and E. Dagotto, Phys. Rev. B 81, 014511 (2010).

9. J. Zhao et al., Phys. Rev. Lett. 101, 167203 (2008).

10. D. H. Lu et al., Nature 455, 81 (2008).

11. M. Yi et al., Phys. Rev. B 80, 174510 (2009).

12. R. Yu et al., Phys. Rev. B 79, 104510 (2009).

13. M. Daghofer et al., Phys. Rev. Lett. 101, 237004 (2008).

14. V. B. Zabolotnyy et al., Nature 457, 569 (2009).

15. T. Shimojima et al., Phys. Rev. Lett. 104, 057002 (2010).

16. S. Graser, T. A. Maier, P. J. Hirschfeld, and D. J. Scalapino, New J. Phys. 11, 025016 (2009).

17. E. Kaneshita, T. Morinari, and T. Tohyama, Phys. Rev. Lett. 103, 247202 (2009).

18. A. M. Oleś, Phys. Rev. B 28, 327 (1983).

19. M. Puertas, M. Aichhorn, and C. Dahnken, Phys. Rev. Lett. 91, 206402 (2003).

20. Not only is the intraorbital hopping in the \(x\)-direction smaller for \(yz\) than for \(xz\), but the substantial interorbital hopping connecting the \(yz/yz\) orbitals to \(xy\) is zero along \(x\) (\(y\)) for symmetry reasons.

21. Since our mean-field approach breaks the orbital symmetry by neglecting the “pair-hopping” and spin-flip terms of the onsite interaction, we have to check that the \(yz\) electron pocket’s predominant interaction with the \(yz\) parts of the hole pockets is not an artifact of the approximation. We perform the mean-field calculation in a rotated basis with orbitals \(|a\rangle = \cos \alpha |xz\rangle + \sin \alpha |yz\rangle\) and \(|b\rangle = -\sin \alpha |xz\rangle + \cos \alpha |yz\rangle\) and find that (i) the energy is minimal for the original \(\alpha = 0\) and that (ii) \(m_{yz} > m_{xz}\) persists for \(\alpha \neq 0\).

22. For the intermediate-\(U\) regime with small ordered moment, the \((\pi, 0)\) phase has actually slightly higher energy than a \((\pi, \pi)\)-AF phase with large ordered moment, but since the five-band model is widely used by several authors, we are nevertheless going to discuss its properties under the assumption of \((\pi, 0)\) order.

23. A. Moreo, M. Daghofer, J. A. Riera, and E. Dagotto, Phys. Rev. B 79, 134502 (2009).

24. K. Kubo and P. Thalmeier, J. Phys. Soc. Jpn. 78, 083704 (2009).

25. S. Haas, A. Moreo, and E. Dagotto, Phys. Rev. Lett. 74, 4281 (1995).