

# Gutzwiller Theory of Band Magnetism in LaOFeAs

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Five years after the discovery of high- $T_c$  superconductivity in F-doped LaOFeAs, understanding the phase diagram of these systems is still a challenging problem. This wide class of compounds is characterized by two-dimensional layers, in which iron atoms form a square lattice, and are engaged in tetrahedra, formed by pnictogen or chalcogen atoms sitting above or below the planes. Typically, at ambient pressure and zero doping, the spins of the iron atoms order below a spin-density-wave (SDW) transition temperature  $T_{SDW}$ . For most pnictides, the ordering pattern is an antiferromagnetic (AFM) *stripe*, in which Fe spins align ferromagnetically along one Fe-Fe bond, and antiferromagnetically along the other. Usually, superconductivity occurs when the SDW order is destroyed by doping or pressure.

Understanding the nature of the magnetically ordered state in Fe-based superconductors, and its relation to superconductivity, has been the subject of an intense debate. Several points are puzzling in this respect: the values of the measured magnetic moments range from  $\sim 0.5 \mu_B$  in some pnictides to  $\sim 2.0 \mu_B$  in the chalcogenides, with no obvious relation to the critical temperature; the moments are much smaller than the saturation value of  $\sim 4.0 \mu_B$  for Fe in the  $d^6$  configuration. The stripe AFM pattern corresponds to a spin density wave with  $\mathbf{Q} = (0, \pi)$ , which is a nesting vector of the Fermi surface for most compounds. This hints towards a weak-coupling SDW. However, compounds with no static magnetism or different ordering patterns exhibit the same Fermi surface topology. Therefore, an entirely itinerant picture seems to be inadequate to describe magnetism in Fe pnictides. In fact, local-spin density functional (LSDFT) calculations reproduce the SDW order observed in experiments, however, the **magnitude** of the magnetic moment ( $\sim 2.0 \mu_B$ ) is in most cases much larger than its experimental value and almost independent of the compound. Moreover, the same moment values are found for a large range of dopings where experiment sees no trace of long-range magnetic order.

Adding local correlations, in the form of multiband Hubbard models, sensibly improves the agreement between theory and experiment. Studies which employed dynamical mean field theory (DMFT) have revealed that in Fe-based superconductors two different magnetic moments coexist: a large, *local* magnetic moment, ( $m \sim 2.0 \mu_B$ ), which is also present in the paramagnetic phase, and a much smaller ordered moment ( $m \sim 0.3 - 1.0 \mu_B$ ) [1]. Due to the large computational cost of DMFT calculations, however, most of these studies have focused on single points of the phase diagram and did not work with the full (i.e., rotationally invariant) local Hamiltonian of the corresponding multiband Hubbard models.

In our work [2], we have therefore combined local density functional and Gutzwiller theory (LDA+GT), to study the phase diagram and other properties of an eight-band Hubbard model for Fe-based superconductors. The Gutzwiller theory is a variational approach which allows for the calculation of ground-state properties as well as quasi-particle band structures. In our study, we considered a multiband Hubbard Hamiltonian  $\hat{H} = \hat{H}_0 + \hat{H}_C$  with a non-interacting eight-band Hamiltonian  $\hat{H}_0$  that was constructed by downfolding the electronic structure of the prototype compound LaOFeAs to Fe3d and As4p NMT0 Wannier orbitals [3]. The Hamiltonian  $\hat{H}_C$  contains the local Coulomb interaction of electrons in the Fe orbitals. In a spherical approximation, all terms in this Hamiltonian are determined by the intra-orbital Hubbard interaction  $U$  and an average Hund's-rule interaction  $J$ .

The main point of our work is summarized in Fig. 1, where we plot the magnetic moment  $m$  in the stripe AFM state, as a function of the interaction parameters ( $U, J$ ). Establishing the SDW order, with a small, finite magnetic

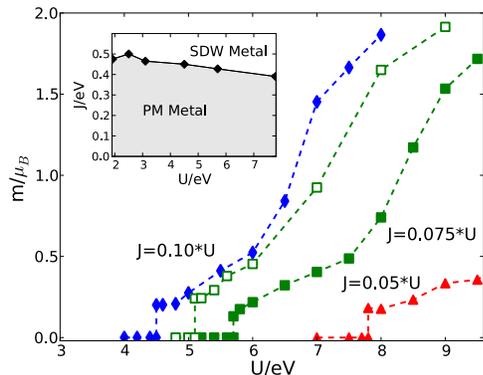


Figure 1: GT ordered magnetic moment as a function of  $U$  for various values of  $J/U$ . Full symbols: full atomic Hamiltonian; open symbols: restricted atomic Hamiltonian with density-density interactions only. Inset: Ground-state phase diagram as a function of  $U$  and  $J$ .

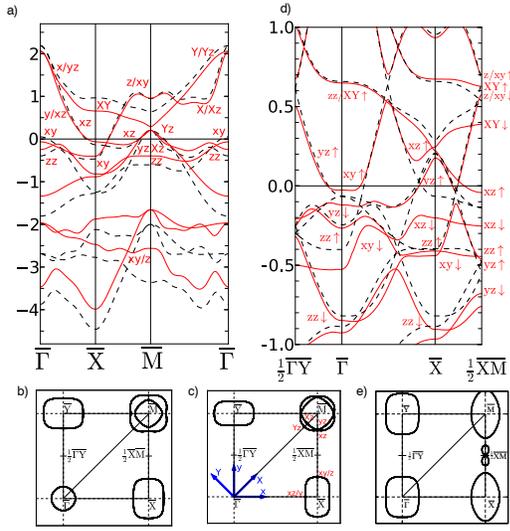


Figure 2: (Top: Fe3d–As4p bandstructure of LaOFeAs along the high-symmetry lines of the 2D Brillouin zone containing one formula unit per cell. *a*) DFT[3] (dashed) and GT (solid) in the PM phase for  $(U, J) = (8, 0.6)$  eV. The bands are lined up at the Fermi level ( $E = 0$ ), energies on the ordinate are in eV, the DFT bandwidth is  $W_{\text{DFT}} = 6.5$  eV. *b*) GT bands for  $(U, J) = (8, 0.6)$  eV in the PM (dashed) and SDW (solid) phases in the Brillouin zone folded along the line  $\frac{1}{2}\bar{\Gamma}\bar{Y}-\frac{1}{2}\bar{X}\bar{M}$ . High symmetry points and the directions  $x, y$  and  $X, Y$  are defined in panel *d*. Spin- $\uparrow$  are minority and spin- $\downarrow$  are majority spin states.  $z/xy$  means more  $z$  and less  $xy$  character. Bottom: Fermi surfaces for the PM state in DFT (*c*) and GT (*d*), and for the SDW state in GT (*e*). The magnetic moment of the SDW state in panels *b* and *e* is  $0.7 \mu_B$ .

moment  $m \lesssim 0.3\mu_B$ , requires a finite Hund's coupling  $J$ . The stripe moment then slowly rises up to  $m = 0.5\mu_B$ , where the  $m$  vs  $U$  curves bend up until  $m = 2.0\mu_B$ . We found that not only the onset points of the SDW, but also the full magnetization curves, collapse onto each other, if the data are plotted as a function of the energy  $I \equiv J + 0.017U$ , see Ref. [2]. The overall shape of the  $m$  vs  $I$  curves then strongly resembles the susceptibility curve for a stripe SDW in LaOFeAs, that we constructed in Ref. [3] using a simple Stoner model.

In Fig. 2, we focus on a representative point  $(U, J) = (8, 0.6)$  eV which, in the SDW state, exhibits a moment of  $m \sim 0.7 \mu_B$ . Panel (a) shows the effect of local correlations on the PM electronic structure, comparing the PM bands of the original DFT  $\hat{H}_0$  (black), with those of GT (red). The corresponding Fermi surfaces are shown in panels (b) and (c), respectively. Local correlations renormalize the quasi-particle weight of all  $d$  states and cause orbital-dependent energy shifts, leading to a partial rearrangement of the electronic bands. Around the Fermi level, the  $xy$  band around the  $\bar{\Gamma}$  point is pulled  $\sim 50$  meV down in energy, and the corresponding  $\bar{\Gamma}$ -centred hole pocket disappears. The electronic structure of the SDW phase in GT, shown in red in Fig. 2 (d), can be understood starting from the folded-in paramagnetic bands, shown in black.

In a simplified Stoner model, the SDW bands split approximately by  $\Delta$  (the staggered 'exchange field') times the overlap of their  $d$ -orbital characters, where the PM  $E(\mathbf{k})$ -bands cross the  $E(\mathbf{k} + \mathbf{q})$  bands ( $\mathbf{q} = [\bar{\Gamma}\bar{Y}] = [\bar{X}\bar{M}]$ ) acquiring majority ( $\downarrow$ ) and minority ( $\uparrow$ ) spin character [3]. This is also seen in the GT bands, which we decorated with partial spin and orbital characters. One clearly sees, for example, that the  $xz$  states give the biggest contribution to the magnetic moment, since the corresponding majority/minority characters sit mostly below/above  $E_F$ .  $xz$  states are easily polarized, also for small moments  $m$ , because they contribute to both hole ( $\bar{M}$ ) and electron ( $\bar{X}$ ) sheets at  $E_F$ , which fold on top of each other for an AFM stripe with  $\mathbf{Q} = (0, \pi)$ .

Besides the ordered magnetic moment ( $m = \langle \hat{S}_z \rangle$ ), we have used our approximate ground-state wavefunction to compute also the local magnetic moment, as the average of the local spin operator. For our representative point, we found  $\langle \hat{S}_i^2 \rangle = \sum_s p(s)s(s+1) \approx \langle \hat{S}_i^2 \rangle \approx 1.62$  with only a small difference between the PM and SDW state. This is in agreement with corresponding DMFT calculations.

In summary, we have investigated the magnetic phase diagram of Fe pnictides, combining LDA and Gutzwiller theory, for the solution of a realistic multiband Hubbard model of the electronic structure [2,3]. We found that a stripe SDW state is stable only if the exchange interaction  $J$  is finite. The value of the ordered moment in the SDW state is then in the range of the experimental data over a wide range of Coulomb-interaction parameters.

## References:

- [1] See, e.g., Aichhorn, M. et al., Phys. Rev. B **80** 085101 (2009); Yin, Z.P. et al., Nature physics **7**, 294 (2011).
- [2] Schickling, T., F. Gebhard, J. Bünemann, L. Boeri, O.K. Andersen, W. Weber. Phys. Rev. Lett. **108**, 036406 (2012).
- [3] Andersen, O.K. and L. Boeri. Annalen der Physik (Leipzig) **523**, 8 (2011).

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