Orbital order induced stabilization of the \((\pi, 0)\) ordered magnetic state in a minimal two-band model for iron pnictides

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Spin wave excitations and stability of the \((\pi, 0)\) ordered magnetic state are investigated in a minimal two-band itinerant-electron model for iron pnictides. Presence of hopping anisotropy generates a strong ferro-orbital order in the \(d_{xz}\) and \(d_{yz}\) Fe orbitals. The orbital order sign is as observed in experiments. By optimizing the strength of the emergent AF and F spin couplings through optimal band fillings in the two orbitals, the induced ferro-orbital order is shown to strongly enhance the spin wave energy scale and stabilize the magnetic state. The calculated spin-wave dispersion is in quantitative agreement with neutron scattering measurements. Finite inter-orbital Hund’s coupling is shown to strongly stabilize the SDW state by coupling the two magnetic sub-systems.

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The iron pnictides exhibit a typical phase diagram \(^1\text{–}^2\) in which the parent compound goes through a tetragonal-to-orthorhombic structural phase transition (at \(T_S\)) and spin ordering transition (at \(T_N\)). Upon doping, both transitions are suppressed and superconductivity emerges. Single crystal neutron scattering experiments show that Fe moments align antiferromagnetically (AF) along the \(a\) direction and ferromagnetically (F) along the \(b\) direction \(^3\), so that the magnetically ordered state can be viewed as a \((\pi, 0)\) ordered spin density wave (SDW) state. Inelastic neutron scattering experiments yield well-defined spin-wave excitations up to the zone boundary on an energy scale \(\sim 200\) meV \(^4\text{–}^9\).

Several weak coupling models with Fermi surface (FS) nesting have been proposed to account for the observed magnetic order \(^1\text{–}^2\). Although these models can explain low-energy magnetic excitations, they fail at higher energies and suggest that spin waves enter the particle-hole continuum at high energies and become higher energies and indicate that spin waves enter the particle-hole continuum at high energies and become.

In this paper, we will investigate the effect of orbital ordering on the SDW state stability within a minimal two-band itinerant-electron model. We will study how the variations in orbital disparity affects the induced AF and F spin couplings, which provides a microscopic understanding of the role of orbital ordering on the stability of the \((\pi, 0)\) ordered SDW state in the relevant intermediate coupling regime.

The iron pnictides have a quasi two-dimensional structure with layers of FeAs stacked along the \(c\) axis. Among the five Fe 3d orbitals, only \(d_{xy}\) and \(d_{yz}\) contribute to orbital ordering (due to \(C_4\) symmetry of others) and we retain only these two in our model. The hybridization of Fe 3d orbitals with themselves as well as through the As 3p orbitals lying above and below the square plaquettes formed by the Fe atoms leads to hopping parameters in our two-orbital model. The hopping amplitudes are shown in Fig. 1 in which the \(d_{xz}\) and \(d_{yz}\) orbitals are taken to be extended along \(x(a)\) and \(y(b)\) direction respectively since the cores of Fe \(d_{xz}\) and \(d_{yz}\) Wannier Functions extend towards the direction of magnetic ordering \(^1\text{–}^2\). Although hybridization between the orbitals can lead to finite \(t_s\) (i.e. \(\pi\) overlap), for simplicity we consider \(t_s\) (i.e. \(\sigma\) overlap) only in our model. However, orbital ordering in our model persists as long as there is hopping anisotropy i.e. \(t_s\) is larger than \(t_{\pi}\). A finite intra-orbital next-nearest-neighbor (NNN) hopping \(t'_s\), expected due to presence of Fe-As-Fe path along plaquette diagonals, is also included. Finite \(t'_s\) plays a very important role in stabilizing the SDW ordering.

We start with the two-orbital Hubbard model Hamiltonian:

\[ H = - \sum_{(ij)\mu\nu\sigma} t_{ij}^{\mu\nu} a_{i\mu\sigma}^\dagger a_{j\nu\sigma} + \sum_{i\mu} U_{\mu} n_{i\mu\uparrow} n_{i\mu\downarrow} \]

where \(i, j\) refer to lattice sites; \(\mu, \nu\) are the orbital indices, \(t_{ij}^{\mu\nu}\) are the hopping terms as shown in Fig. 1, and \(U_{\mu}\) are the intra-orbital Coulomb interactions. The role of inter-orbital density interaction and Hund’s coupling will be discussed later.

In this model, the two orbitals are decoupled, with non-magnetic state dispersions \(\varepsilon_\alpha(k) = -2t \cos k_x - \)
4't cos k_x cos k_y for the d_{xz} and \( \varepsilon_\beta(k) = -2t \cos k_x - 4't \cos k_x \cos k_y \) for the d_{yz} orbitals. Although the dispersions are different, the energy bands are degenerate in the non-magnetic state wherein x and y directions are equivalent. This degeneracy is important to satisfy point-group symmetry conditions. It is noteworthy that the d_{zz} band along \text{Γ–Y} and d_{xy} along Γ–X are degenerate in energy, as indeed observed in ARPES studies on BaFe_{2}As_{2} in the non-magnetic state above the ordering temperature.21

In the (π, 0) ordered SDW state, the Hartree-Fock (HF) level Hamiltonian matrix is expressed in a composite two-orbital (α, β), two-sublattice (A B) basis as:

\[
H_{\text{HF}}(k) = \begin{bmatrix}
-\sigma \Delta_\alpha & \varepsilon_k^x + \varepsilon_k^y \\
\varepsilon_k^x + \varepsilon_k^y & 0 & 0 & 0 \\
0 & 0 & -\sigma \Delta_\beta & \varepsilon_k^y \\
0 & 0 & \varepsilon_k^y & \sigma \Delta_\beta + \varepsilon_k^y
\end{bmatrix}
\]

where \( \varepsilon_k^{x(y)} = -2t \cos k_x \cos k_y \), \( \varepsilon_k^x = -4't \cos k_x \cos k_y \), \( \sigma = \pm \) for the two spins. The self-consistent exchange fields are given by \( 2\Delta_\mu = U_\mu m_\mu \) in terms of the sublattice magnetizations \( m_\mu \) for the two orbitals \( \mu = \alpha, \beta \). As the intermediate-coupling regime will be considered throughout, the term “SDW state” is used here without any implicit weak-coupling connotation.

The density terms arising in the HF approximation, \( U_\mu n_\mu / 2 = \Delta_\mu + U_\mu n_\mu^\dagger \) for the two orbitals, have not been shown explicitly in (1). In the following, we will take identical exchange fields \( \Delta_\alpha = \Delta_\beta = \Delta \), for which we find the relative band shift \( U_\alpha n_\alpha^\dagger - U_\beta n_\beta^\dagger \) to be quite small, and this will be absorbed in an energy offset to be introduced later.

In iron pnictides, each iron ion has six electrons distributed among five 3d orbitals. Two \( e_g \) orbitals \( d_{xz} - d_{yz} \) and \( d_{xy} - d_{xy} \) are completely occupied by four electrons due to large crystal-field splitting between \( e_g \) and \( t_{2g} \) states.23 and the three \( t_{2g} \) orbitals are partially filled by the remaining two electrons. ARPES experiments show that the \( d_{xy} \) orbital has a finite contribution to the Fermi Surface. Therefore, the expected electron filling in our two-band model will be less than half filling, and will correspond to a “hole doped” condition.

The partial densities of states for the two orbitals are shown in Fig. 2 for the non-magnetic (Δ=0) and magnetic (Δ/|t|=2) cases. While the two orbitals are degenerate in the non-magnetic state, the degeneracy is lifted in the magnetic state since x and y directions are no longer equivalent. The hopping anisotropy together with magnetic ordering anisotropy naturally results in self orbital order in the magnetic state. For hole doping (\( E_F < 0 \)), the lower \( \beta \) band becomes partially unoccupied whereas the lower \( \alpha \) band remains nearly half-filled. For total electron occupation \( n \approx 0.75 \) per orbital (i.e. 25% hole doping), we have \( n_\alpha \approx 1.0 \) and \( n_\beta \approx 0.5 \). This sign of ferro-orbital order (\( n_\alpha - n_\beta > 0 \)) is in agreement with experiments.21, 22 Non-equivalence between \( \alpha \) and \( \beta \) DOS naturally leads to in-plane anisotropic behavior in FS and electronic properties in the (π, 0) SDW state.

Spin-wave excitations in this spontaneously-broken-symmetry SDW state are obtained from the transverse spin fluctuation propagator:

\[
[\chi_{\text{RPA}}(q, \omega)]_\alpha = \frac{[\chi_0^0(q, \omega)]_\alpha}{1 - [U][\chi_0^0(q, \omega)]}
\]

at the RPA level. The interaction matrix \([U]\) includes \( U_\alpha \) and \( U_\beta \) as diagonal matrix elements. The bare particle-hole propagator \([\chi_0^0(q, \omega)]\) is evaluated in the composite orbital-sublattice basis by integrating out the fermions in the (π, 0) ordered SDW state as22

\[
[\chi_0^0(q, \omega)]_{ab} = i \int \frac{d\omega'}{2\pi} \sum_{k'} [G_{HF}^\dagger(k', \omega')]_{ab}
\]

\[
[G_{HF}^\dagger(k - q, \omega' - \omega)]_{ba}
\]

where \([G_{HF}^\dagger(k, \omega)] = [\omega 1 - H_{HF}^\dagger(k)]^{-1}\) are the HF level
Green’s function in the SDW state. The spin-wave energies are obtained from the poles of Eq. (2).

![Spin-wave dispersions for the two magnet modes α and β along symmetry directions of the BZ for 25% hole doping. Here \( t' = -200 \text{ meV}, t'/t = 0.5, \Delta/|t| = 2.0, \) and the Fermi energy \( E_F/|t| \approx -2.0 \).

FIG. 3: Spin-wave dispersions for the two magnet modes α and β along symmetry directions of the BZ for 25% hole doping. Here \( t' = -200 \text{ meV}, t'/t = 0.5, \Delta/|t| = 2.0, \) and the Fermi energy \( E_F/|t| \approx -2.0 \).

In the absence of any Hund’s coupling, the two orbitals are decoupled and the magnetic system reduces to two independent magnetic sub-systems α and β involving AF ordering in \( x \) direction (α magnet) and F ordering in \( y \) direction (β magnet). The spin-wave dispersions for the two magnet modes α and β are shown in Fig. 3. For \( t' = 0 \), the system further reduces to independent AF and F chains, which yield zero spin wave energy for wave vectors in the respectively perpendicular directions (due to absence of any spin coupling along those directions), implying instabilities of the magnetic state. Fig. 3 thus highlights the important role of finite \( t' \) and the corresponding finite inter-chain spin couplings in stabilizing the two α and β magnets with respect to spin twisting in any direction, as indicated by positive spin wave energies over the entire BZ.

To investigate the effect of orbital order on the SDW state stability, we plot the spin-wave dispersions for different orbital polarization \( \delta n = n_\alpha - n_\beta \) in Fig. 4. As magnetic state instability is indicated by the spin-wave energy crossing zero and going negative, we will focus only on the lowest (out of the two α and β mode) energies. Here, we have maintained a constant SDW order parameter \( \Delta \) and total electron filling \( n = 0.75 \) per orbital, and the occupations \( n_\alpha \) and \( n_\beta \) of the two orbitals are controlled by introducing an energy offset \( \Delta_{\alpha \beta} \) between the two orbitals. The orbital occupations are (a) \( n_\alpha = 0.85, n_\beta = 0.65 (\Delta_{\alpha \beta} = 0.38) \), (b) \( n_\alpha = 0.9, n_\beta = 0.6 (\Delta_{\alpha \beta} = 0.29) \), (c) \( n_\alpha = 0.95, n_\beta = 0.55 (\Delta_{\alpha \beta} = 0.20) \) and (d) \( n_\alpha = 1.0, n_\beta = 0.5 (\Delta_{\alpha \beta} = 0) \). Figure 4 shows a strong stabilization of the SDW state with orbital order, as seen by crossover from negative to positive energy modes.

The origin of this orbital-order-induced stabilization is as follows. Effectively, the AF and F spin couplings are optimized by the electron density redistribution associated with orbital order. The increased electron density in α band favors AF coupling (super exchange) in the \( x \) direction (NN) and in the diagonal direction (NNN), whereas the increased hole density in β band favors F coupling (carrier-mediated) in the \( y \) direction (NN) and AF coupling in the diagonal direction (NNN). Thus, all the spin couplings work together and the α and β magnets both reinforce \((\pi, 0)\) ordering without any frustration. It is important to note here that the magnetic and orbital orderings effectively stabilize each other and constitute a composite spin-orbital ordered state with \( m \neq 0, \delta n \neq 0 \). Our model thus provides a microscopic understanding of the close relation between in-plane anisotropy, orbital order, and the SDW magnetic order. If the inter-orbital Coulomb interaction term \( V n_{i\alpha} n_{i\beta} \) is included in our model, it will only enhance the orbital offset \( \Delta_{\alpha \beta} \) due to orbital disparity, and therefore enhance the effect discussed above.

Ferro orbital order was also reported in a recent study of magnetic excitations in iron pnictides within a degenerate double-exchange model including antiferromagnetic superexchange interactions. Orbital and spin polarizations were studied as function of band filling and the spin-fermion coupling strength. Breaking of in-plane lattice symmetry by ferro orbital order was linked to the orthorhombic lattice distortion, strong electronic anisotropy, Fermi surface reconstruction, and transport anomalies. However, the sign of the ferro orbital order reported in this work \( n_{2x} > n_{2z} \) does not agree with experiments. Furthermore, for a realistic NN hopping value of 200 meV, their calculated spin wave energy scale of around 30 meV is well below the nearly 200 meV energy scale measured in neutron scattering experiments. Also, inter-orbital Hund’s coupling was not included.

Finally, we investigate the effect of finite inter-orbital Hund’s coupling \( J \), and consider the Hamiltonian:

\[
H = - \sum_{\langle ij \rangle \mu \nu} \mu \nu (a_{i \mu \sigma}^\dagger a_{j \nu \sigma} + a_{j \nu \sigma}^\dagger a_{i \mu \sigma}) - \sum_{i \mu \nu} U_{\mu \nu} S_{\mu} \cdot S_{\nu},
\]

(4)

where the interaction matrix elements \( U_{\mu \nu} = U_{\mu \nu} \) for \( \mu = \nu \) and \( U_{\mu \nu} = 2J \) for \( \mu \neq \nu \). The self-consistent exchange fields.
are now given by $2\Delta_\alpha = U_\alpha m_\alpha + J m_\beta$ and $2\Delta_\beta = U_\beta m_\beta + J m_\alpha$.

The transverse spin fluctuation propagator now includes both $U$ and $J$ ladders at RPA level. Figure 5 shows the spin-wave dispersion with and without Hund’s coupling, displaying a strong enhancement of spin-wave energies with Hund’s coupling, as expected since now the $\alpha$ and $\beta$ magnets are coupled.

For $J=0$, there are two independent Goldstone modes, as the $\alpha$ and $\beta$ magnets are independent. When finite Hund’s coupling is included, the two modes get coupled, leading to a single Goldstone mode corresponding to “in-phase” fluctuations (acoustic mode). The “out-of-phase” mode is now converted to an optical branch which rapidly becomes significantly gapped even at small values of $J$.

The spin-waves for the acoustic and optical modes (blue and red respectively) are shown in the inset [Fig. 5].

The calculated spin-wave energy scale ($\sim 200$ meV) agrees well with neutron scattering experiments for $|t| = 200$ meV and $U$ in the intermediate coupling range. Furthermore, the spin-wave dispersion shows a peak at the zone boundary which is consistent with experiments. For $\Delta/|t| = 2.0$, the SDW state effective gap $\sim 400$ meV is well above the maximum spin wave energy. Thus, contrary to other itinerant models, spin wave excitations in our model do not rapidly dissolve into the particle-hole continuum, as indeed not observed experimentally up to energies of 200 meV. Furthermore, the large separation between the moment-melting and moment-disordering temperatures in our model is consistent with the experimental observation of short-range spin wave excitations even above the magnetic transition temperature.

The spin wave energy scales in the $\Gamma$-$M$ (AF) and $M$-$X$ (F) directions are indicative of the AF and F spin coupling strengths. The hole doping dependence of spin wave dispersion (Fig. 6) effectively shows the evolution of emergent spin couplings. At high hole doping, the AF spin coupling is strongly suppressed due to electron density depletion in the AF band, whereas the F spin coupling is optimized due to enhanced hole doping in the F band. On the other hand, at low hole doping, the AF spin coupling gets saturated as the AF (lower) band is filled, whereas the F spin coupling is strongly weakened.

Maximum SDW state stability is seen for about 25% hole doping. If this corresponds to the parent compound, then electron doping (reduction in hole doping from this level) results in crossover to negative-energy modes, indicating destabilization of the SDW state. This is in agreement with the observed rapid decrease of magnetic ordering temperature in iron pnictides with electron doping.

Spin waves were studied earlier in a two-band model with isotropic hopping and therefore no orbital order. For somewhat larger interaction strength ($\Delta'/|t'| = 3$) and hole doping ($\sim 40\%$), the spin wave energies are qualitatively similar as in Fig. 6. However, for lower interaction strength and hole doping, increasing hopping anisotropy significantly enhances the spin wave energy scale and SDW state stability, as shown in Fig. 7. This confirms that the emergent AF and F spin couplings become stronger due to the electron density redistribution and orbital order in the two orbitals, corresponding to enhanced electron and hole densities in the $d_{xz}$ and $d_{yz}$ Fe orbitals, respectively.

In conclusion, we have shown that within a minimal two-orbital itinerant-electron model with hopping anisotropy, the magnetically anisotropic $(\pi,0)$ ordered SDW state can be self-consistently stabilized through the generation of orbital order which optimizes both AF and Hund’s coupling.
F spin couplings. We have also shown that with the inclusion of inter-orbital Hund’s coupling, the spin stiffness further increases substantially due to the coupling of the two magnetic sub-systems. The calculated spin-wave energy scale, nature of spin-wave dispersion and presence of ferro-orbital order are in agreement with experimental observations. Thus, the proclivity of iron arsenides towards $(\pi,0)$ magnetic ordering may actively involve the orbital degree of freedom. The presence of orbitally ordered state in our model allows the possibility of exploring orbital fluctuations which have been seen in recent experiments. Orbital fluctuations may play an important role on the electron-paring mechanism and can induce $s+\pi$ wave superconducting state and therefore a combination of orbital and spin fluctuations as a possible mechanism for superconductivity also needs to be explored in these materials.

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