Multi-orbital quantum antiferromagnetism in iron pnictides—effective spin couplings and quantum corrections to sublattice magnetization

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Abstract

Effective spin couplings and spin fluctuation induced quantum corrections to sublattice magnetization are obtained in the $(\pi,0)$ AF state of a realistic three-orbital interacting electron model involving $xz$, $yz$ and $xy$ Fe $3d$ orbitals, providing insight into the multi-orbital quantum antiferromagnetism in iron pnictides. The $xy$ orbital is found to be mainly responsible for the generation of strong ferromagnetic spin coupling in the $b$ direction, which is critically important to fully account for the spin wave dispersion as measured in inelastic neutron scattering experiments. The ferromagnetic spin coupling is strongly suppressed as the $xy$ band approaches half filling, and is ascribed to particle-hole exchange in the partially filled $xy$ band. The strongest AF spin coupling in the $a$ direction is found to be in the orbital off-diagonal sector involving the $xz$ and $xy$ orbitals. First order quantum corrections to sublattice magnetization are evaluated for the three orbitals, and yield a significant 37% average reduction from the Hartree–Fock value.

Keywords: spin couplings, multi-orbital models, iron pnictides, quantum magnetism

(Some figures may appear in colour only in the online journal)

1. Introduction

The rich phase diagram exhibited by iron pnictides [1–3] including magnetic, structural and superconducting phase transitions [4] have stimulated intensive investigations aimed at detailed understanding of their macroscopic physical behavior in terms of their complex multi-orbital electronic structure as revealed by first-principle calculations [5–12] and angle resolved photoemission spectroscopy (ARPES) experiments [13–17]. The magnetic state exhibits $(\pi,0)$ ordering of Fe moments in the $a$–$b$ plane, with a concomitant structural distortion $a > b$, possibly correlated with the ferro orbital order $n_{xz} > n_{yz}$ as seen in ARPES studies [15].

Inelastic neutron scattering studies of magnetic excitations in iron pnictides have been carried out extensively [18–24], and clearly reveal well defined spin wave excitations with energy scale $\sim$200 meV, persisting even above the Néel temperature [24], indicating that short-range antiferromagnetic (AF) and ferromagnetic (F) order remain in the $a$ and $b$ directions, respectively, even above the disordering temperature for long-range magnetic order. This persistence of short-range anisotropic magnetic order may account for the narrow nematic phase [25, 26] above the Néel temperature where the ferro orbital order [15] and structural distortion survive, as well as the temperature dependence of the measured anisotropies in $a$ and $b$ directions of magnetic excitations and resistivity [15, 24, 27].

Spin wave excitations in iron pnictides have been theoretically studied for multi-band models [28–32] in the random phase approximation (RPA). The magnitude of the intra-orbital Coulomb interaction term considered in these investigations typically lie in the intermediate coupling range
(\(U \approx 1-2\) eV), resulting in moderately well developed local moments in the \((\pi, 0)\) magnetic state. In this paper we will study the effective spin couplings generated by particle-hole exchange, their orbital contributions, and quantum corrections to sublattice magnetization in the \((\pi, 0)\) AF state of a realistic three-orbital model which yields Fermi surface structure, spin wave excitations, and ferro orbital ordering in quantitative agreement with experiments.

One important feature of the measured spin wave dispersion is that the spin wave energy is maximum at the ferromagnetic zone boundary (FZB), slightly higher than at the antiferromagnetic zone boundary (AFZB). This feature is quite significant as the FZB spin wave energy provides a sensitive measure of the effective ferromagnetic (F) spin coupling. Understanding the microscopic mechanism behind the origin of this strong F spin coupling should provide significant insight toward understanding the multi-orbital quantum antiferromagnetism in the iron pnictides.

Following a brief account in section 2 of the realistic three-orbital model in terms of the \(xz, yz\) and \(xy\) Fe 3d orbitals, effective spin couplings are introduced in section 3 as mediated by exchange of the particle-hole propagator \([\chi^0]\) in the \((\pi, 0)\) magnetic state, and results for the calculated effective spin couplings and their variation with the \(xy\) orbital energy offset are discussed. Evaluation of quantum corrections to sublattice magnetization for the three orbitals is discussed in section 4 in terms of transverse spin correlations \((S^+_i S^+_j)\) and \((S^+_i S^-_j)\), followed by conclusions in section 5.

### 2. Three-orbital model and \((\pi, 0)\) magnetic state

We consider a minimal three-orbital model \([32]\) involving \(d_{xz}, d_{yz}\) and \(d_{xy}\) Fe 3d orbitals. The tight binding Hamiltonian is defined as:

\[
H_0 = -\sum_{ij} \sum_{\mu, \sigma} t_{ij}^{\mu\sigma} (a_{i\mu \sigma}^\dagger a_{j\mu \sigma} + \text{H.c.}) + \varepsilon_{xy} \sum_i n_{i, \mu = xy, \sigma} \tag{1}
\]

where \(a_{i\mu \sigma}^\dagger\) creates an electron at lattice site \(i\) with spin \(\sigma\) in orbital \(\mu\), \(t_{ij}^{\mu\sigma}\) are the in-plane hopping terms including nearest-neighbour (NN) and next-nearest-neighbour (NNN) hoppings, and \(\varepsilon_{xy}\) is the energy difference between the \(xy\) and the degenerate \(xz/yz\) orbitals. The hopping parameter values considered are given in table 1. Here \(t_1\) and \(t_2\) are the intra-orbital hoppings for \(xz, yz\) along \(x(y)\) and \(y(x)\) directions, respectively, \(t_3\) and \(t_4\) are the intra and inter-orbital hoppings along diagonal direction for \(xz, yz\), \(t_5\) and \(t_6\) are intra-orbital NN and NNN hoppings for \(xy\), while \(t_7\) and \(t_8\) the NN and NNN hybridization between \(xy\) and \(xz/yz\).

The Fermi surface for the three-orbital Hamiltonian (1) for hopping parameter values given in table 1 and near half filling shows two circular hole pockets around the center and elliptical electron pockets around \((\pm \pi, 0)\) and \((0, \pm \pi)\) in the unfolded BZ. \([32]\) The two hole pockets involve primarily the \(xz\) and \(yz\) orbitals, while the electron pockets centered at \((\pm \pi, 0)\) \([0, \pm \pi]\) arise mainly from the hybridization of the \(xy\) and \(yz/xz\) orbitals. All of these features are in good agreement with results from DFT calculations and ARPES experiments.

We now consider the \((\pi, 0)\) ordered SDW state of this model. The various electron–electron interaction terms included are:

\[
H_I = U \sum_{i, \mu} n_{i, \mu} n_{i, \mu} + (U' - J/2) \sum_{i, \mu} n_{i, \mu} n_{i, \mu} - 2J \sum_{i, \mu, \nu} S^\mu_i \cdot S^\nu_i + J' \sum_{i, \mu, \nu} (a_{i\mu \sigma}^\dagger a_{i\mu \sigma} a_{i\nu \sigma} a_{i\nu \sigma}^\dagger + \text{H.c.}), \tag{2}
\]

where \(S_i^\mu (n_i^\mu)\) refer to the local spin (charge) density operators for orbital \(\mu\). The first and second terms are the intra-orbital and inter-orbital Coulomb interactions respectively, the third term the pair-hopping and the fourth term the ‘pair-hopping’ term. In the following, we will consider \(U = 1.2\) eV and \(J \approx U/4\). This interaction strength corresponds to the intermediate coupling regime, which is in accord with recent DFT + DMFT study of magnetism in iron pnictides \([33]\).

Extending the two-sublattice basis approach for the SDW state in a single-band model \([34]\) to a composite three-orbital, two-sublattice basis, the Hartree–Fock (HF) level Hamiltonian matrix in this composite basis \((A\pi z A\pi y A\pi x B\pi y B\pi x)\) is obtained as:

\[
H_{\text{HF}}(k) = \begin{bmatrix}
-\sigma \Delta_{xz} + \varepsilon_{2y}^k & 0 & 0 & \varepsilon_{4}^k + \varepsilon_{3}^k & \varepsilon_{2}^k + \varepsilon_{1}^k & \varepsilon_{2}^k + \varepsilon_{1}^k & \varepsilon_{2}^k + \varepsilon_{1}^k \\
0 & -\sigma \Delta_{yz} + \varepsilon_{2y}^k & \varepsilon_{2y}^k & \sigma \Delta_{xy} + \varepsilon_{2y}^k & \varepsilon_{2y}^k & \sigma \Delta_{xy} + \varepsilon_{2y}^k & 0 \\
-\sigma \Delta_{xy} + \varepsilon_{2y}^k & -\sigma \Delta_{xy} + \varepsilon_{2y}^k & 0 & 0 & 0 & 0 & 0 \\
\varepsilon_{2y}^k & -\varepsilon_{2y}^k & -\varepsilon_{2y}^k & 0 & 0 & 0 & 0 \\
\varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & 0 & 0 & 0 & 0 \\
\varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & 0 & \sigma \Delta_{yz} + \varepsilon_{2y}^k & \sigma \Delta_{yz} + \varepsilon_{2y}^k & \sigma \Delta_{yz} + \varepsilon_{2y}^k \\
-\varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & 0 & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k \\
-\varepsilon_{2y}^k & -\varepsilon_{2y}^k & -\varepsilon_{2y}^k & 0 & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k \\
\sigma \Delta_{xy} + \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k \\
\varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k \\
\sigma \Delta_{xy} + \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k \\
0 & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k \\
0 & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k \\
0 & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k \\
0 & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k & \varepsilon_{2y}^k
\end{bmatrix}
\tag{3}
\]

| Table 1. Hopping parameter values in the three-orbital model (in eV). |
|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| \(t_1\)         | \(t_2\)         | \(t_3\)         | \(t_4\)         | \(t_5\)         | \(t_6\)         | \(t_7\)         | \(t_8\)         |
| 0.1             | 0.32            | -0.29           | -0.06           | -0.3            | -0.16           | -0.15           | -0.02           |

and \(\varepsilon_{xy}\) is the energy difference between the \(xy\) and the degenerate \(xz/yz\) orbitals. The hopping parameter values considered are given in table 1.
in terms of the band energies (over the unfolded Brillouin zone $-\pi \leq k_x, k_y \leq \pi$) corresponding to hopping terms along different directions:
\[
\begin{align*}
\epsilon_{k}^{1x} &= -2t_{1} \cos k_x, \\
\epsilon_{k}^{1y} &= -2t_{1} \cos k_y, \\
\epsilon_{k}^{2x} &= -2t_{2} \cos k_x, \\
\epsilon_{k}^{2y} &= -2t_{2} \cos k_y, \\
\epsilon_{k}^{3x} &= -4t_{3} \cos k_x k_y, \\
\epsilon_{k}^{3y} &= -4t_{3} \cos k_k k_y, \\
\epsilon_{k}^{4x} &= -4t_{4} \sin k_x k_y, \\
\epsilon_{k}^{4y} &= -4t_{4} \sin k_x k_y,
\end{align*}
\] (4)

and the exchange fields $2\Delta_{\mu} = U m_{\mu} + J \sum_{\nu \neq \mu} m_{\nu}$ determined self-consistently from the sublattice magnetization $m_{\mu}$ for orbital $\mu$. The relative orbital energy shift arising in the HF approximation due to the density interaction term $(U - J/2)$ has been discussed in detail in [32]. For our choice of parameters, the relative energy shift between d$_{xz}$ and d$_{yz}$ orbitals is very small and is neglected in our calculations, while the shift between d$_{xz}$ and d$_{yz}$ orbitals has been absorbed in the renormalized energy offset $\xi_0$ for the d$_{xz}$ orbital. The pair hopping term $J'$ does not contribute to the HF Hamiltonian in the magnetic state.

The calculated spin wave spectral function [32] in the SDW state for the three-orbital model is shown in figure 1. Evidently, spin wave excitations are highly dispersive, and do not decay into the particle-hole continuum. The energy scale of spin excitations is $\sim 200$ meV and the spin wave energy at the FZB (0, $\pi$) is slightly higher than at the AFZB ($\pi/2$, 0). These features of spin wave excitations are in excellent agreement with results from inelastic neutron scattering measurements, confirming the realistic nature of the three-orbital model.

3. Effective spin couplings

In the ($\pi$, 0) AF state of the multi-orbital interacting electron model, the spin wave (transverse spin fluctuation) propagator in the random phase approximation (RPA) can be expressed in a symmetrised form:

$$
[\chi^{-1}(q, \omega)] = \frac{[\chi^{0}(q, \omega)]}{1 - [U][\chi^{0}(q, \omega)]} = \frac{1}{[U] - [U][\chi^{0}(q, \omega)][U]} - \frac{1}{[U]}
$$
(5)

where the matrix $[U][\chi^{0}(q, \omega)][U]$ is Hermitian. Here $[\chi^{0}]$ is the bare particle-hole propagator in the orbital-sublattice basis evaluated by integrating out the fermionic degrees of freedom, and $[U]$ is the local interaction matrix with intra-orbital terms $U$ and inter-orbital (Hund’s rule coupling) terms $J$. The bare particle-hole propagator is calculated from the Hartree–Fock (HF) level Green’s function:

$$
[\chi^{0}(q, \omega)]_{\alpha \beta} = i \int \frac{d\omega'}{2\pi} \sum_{k} G_{\alpha' \beta'}(k, \omega') \times G_{\alpha \beta}(k - q, \omega' - \omega)_{\alpha \beta},
$$
(6)

in the sublattice-orbital basis, and involves integrating out the fermions in the ($\pi$, 0) ordered spontaneously-broken-symmetry state. Here $E_{k}$ and $\phi_{k}$ are the eigenvalues and eigenvectors of the HF level Hamiltonian matrix and $l, m$ indicate the eigenvalue branches. The superscripts (+, −) refer to particle (hole) energies above (below) the Fermi energy, and both inter-band and intra-band particle-hole terms are included.

In our Hartree–Fock (HF) treatment of the Coulomb interaction terms (equation (2)), we have considered only expectation values of the spin operators $(S_{\mu}^{\alpha \beta} = \langle n_{\mu \alpha} - n_{\mu \beta} \rangle / 2)$ which yield the local magnetic moments for the three orbitals in the ($\pi$, 0) magnetic state. When condensates of the form $(d_{\mu \alpha}^{1} d_{\mu \beta}^{1})$ or $(d_{\nu \alpha}^{1} d_{\nu \beta}^{1})$ are included at the HF level, [30, 31, 35] the tensor structure of the transverse spin fluctuation propagator must be extended to $[\chi^{-1}(q, \omega)]_{\mu \nu}$, corresponding to the generalized spin operators $S_{\mu \alpha}^{\nu \beta} = d_{\mu \alpha}^{1} d_{\nu \beta}^{1}$ and $S_{\nu \alpha}^{\mu \beta} = d_{\nu \alpha}^{1} d_{\mu \beta}^{1}$. In this case, the corresponding interaction vertices must be incorporated at the RPA (ladder sum) level in order to preserve spin rotational symmetry.

Broadly, the important features of spin wave excitations in the spontaneously-broken-symmetry ($\pi$, 0) magnetic state of the three-band model are: (i) presence of zero-energy Goldstone mode related to continuous spin rotation symmetry of the interacting electron Hamiltonian and (ii) coupling between the three orbitals due to orbital hybridization in the tight-binding model. We therefore consider mapping to an effective spin model:

$$
H = \sum_{(\mu, \nu), \sigma} J_{g}^{\mu \nu} S_{\mu \sigma} S_{\nu \sigma}
$$
(7)

in terms of spin-1/2 operators $S_{\mu \sigma}$ corresponding to the three electronic 3d orbitals $\mu = xz, \gamma z, xy$, which allows for effective spin interactions (couplings) between different orbitals while preserving the spin rotation symmetry.

Since positive spin wave energies are associated with increase in spin interaction energy corresponding to specific spin twisting modes, the effective spin couplings are inherently present in the RPA level spin propagator of equation (5).
Based on earlier spin wave studies in interacting electron models [32, 36, 37], as briefly discussed below, we will consider the correspondence:

$$\frac{-J_{\mu\nu}^{\mu\nu}}{2} = \sum_{\mathbf{q}} \sum_{\mu \neq \nu} \{U^{\mu\mu} \chi_{\mu}^{(0)}(\mathbf{q})U^{\nu\nu} \chi_{\nu}^{(0)} - e^{i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)}\}$$

(8)

where indices $s, s'$ correspond to the sublattices (A/B) which sites $i$ and $j$ belong to. Here we have set $\omega = 0$ in the bare particle-hole propagator $\chi^{(0)}(\mathbf{q}, \omega)$. Neglect of dynamical effects should not affect the results qualitatively in the intermediate coupling regime. The above form clearly suggests exchange of the particle-hole propagator $\chi^{(0)}$ as the origin of the effective inter-site spin couplings in interacting electron models having only local interactions of the form $-U^{\mu\mu}S^{\mu\mu}S^{\mu\mu}$. In the following, we will focus on the first neighbor (1a and 1b) spin couplings in the $a$ (AF) and $b$ (F) directions and the second neighbor (2) coupling along the diagonal direction, evaluated by taking the corresponding lattice vectors $\delta = \mathbf{r}_i - \mathbf{r}_j$ in equation (8).

The above correspondence is known to be exact in the strong coupling limit for several models. It yields the AF first and second neighbor effective spin couplings $J_1 = 4t^2/U$ and $J_2 = 4t^2U/2$ for the $t-t'-U$ model at half filling in the $(\pi, \pi)$ and $(\pi, 0)$ AF states, [32, 36] and also effective F spin coupling of opposite sign in the $(\pi, 0)$ AF state at finite doping. Similarly, in the ferromagnetic Kondo lattice model for doped manganites, the calculated spin wave dispersion in the double exchange limit ($J_H/\pi \rightarrow \infty$) corresponds exactly to that for the quantum Heisenberg ferromagnet with effective spin coupling given by $J_0^2 \chi^{(0)}(\mathbf{q})$ [37]. Finally, the above correspondence reduces to the well known RKKY interaction in the weak coupling limit.

The effective spin couplings $J_{\mu\nu}^{\mu\nu}$ were calculated from equation (8) with the $\mathbf{q}$ summation over the two dimensional Brillouin zone performed for grids up to $60 \times 60$ to ensure no significant variation. The results are shown in table 2 for the first and second neighbors indicated by $\delta = 1a, 1b, 2$, and for the reference case $\epsilon_{x_y} = 0.8}$eV. Among the diagonal terms, the F (negative) spin coupling (1b) is maximum for the $xy$ orbital, whereas AF spin couplings are strong for $xy$ and $yz$ orbitals (1a) and for $xz$ and $yz$ orbitals (2). The dominant off-diagonal term is the AF spin coupling (1a) involving the $xz$ and $xy$ orbitals. Interestingly, this off-diagonal term is the maximum AF spin coupling, highlighting the multi-orbital character of quantum antiferromagnetism and the role of strong orbital hybridization in the three-orbital model.

As all three orbitals follow identical ($\pi, 0$) magnetic ordering, it is useful to consider the total (orbital summed) spin couplings

$$J_0 = \sum_{\mu, \nu} J_{\mu\nu}^{\mu\nu}$$

(9)

which yield the effective couplings as considered in phenomenological spin models to describe the spin wave dispersion in iron pnictides [20, 22, 23]. The most significant feature of the total spin couplings, given in table 3, is the ferromagnetic (negative) first neighbor spin coupling (1b), which is in agreement with [20, 22, 23] where it was shown that spin wave dispersion throughout the BZ and the maximum at $(0, \pi)$ can be explained by a suitably parameterized Heisenberg Hamiltonian with an effective ferromagnetic exchange interaction ($J_{ab} < 0$) in the $b$ direction. The present work provides the microscopic origin of this ferromagnetic interaction as due to the usual particle-hole exchange process in an itinerant-electron model, with the $xy$ orbital being mainly responsible for the strong F spin coupling generated.

The effect of orbital order $n_{x_y} - n_{x_z}$ on spin wave energies and effective spin couplings was investigated by varying the $d_{x_y}$ orbital energy offset $\epsilon_{x_y}$. Figure 2 shows the variation of (a) the electronic densities ($n_{\nu}^\rho$) for the three orbitals and (b) the spin wave energies at the F (0, $\pi$) and AF ($\pi/2, 0$) zone boundaries. While $n_{x_y}$ remains fixed at 1 (half filling), there is a significant transfer of electronic density from $xy$ to $xz$ orbital with increasing $xy$ orbital energy offset. The strong enhancement in the FZB spin wave energy with the depletion in $xy$ orbital electronic density from 1 is due to the corresponding enhancement in the F spin coupling contribution of the $xy$ orbital, as discussed below.

Figure 3(a) shows the variation (again with $\epsilon_{x_y}$) of the total (orbital summed) spin couplings for the first neighbors (1a, 1b) in the $a$ and $b$ directions and the second neighbor (2) along the diagonal direction. For higher values of the $xy$ orbital energy offset, the effective spin coupling $J_{1b}$ is strongly negative, clearly highlighting the strong F spin generated by the particle-hole process mainly in the partially filled $xy$ band. With decreasing $xy$ orbital offset energy, as the $xy$ band approaches half filling, the F component of the spin coupling generated by particle-hole exchange decreases substantially, resulting in the net spin coupling changing sign from negative to positive.

| $\delta$ | 1a | 1b | 2 |
|---------|----|----|---|
| $J_0$ (meV) | 226.48 | -64.52 | 68.86 |

Table 3. The total (orbital summed) effective spin couplings for first and second neighbors.
Due to orbital hybridization in the three-orbital tight binding model (1), the effective spin couplings considered earlier include contributions from the orbital off-diagonal parts $\chi_{\mu \nu} \neq 0$ of the particle-hole propagator as well. In order to identify the individual orbital contributions to the effective spin couplings, we therefore consider the orbital diagonal terms:

$$-\frac{J_\delta}{2} = U^2 \sum_q [\chi_0^q(\mathbf{r})]_{\mu \mu} e^{i\mathbf{q} \cdot (\mathbf{r} - \mathbf{r}')}$$  \hspace{1cm} (10)

which are shown in figure 3(b) for the three orbitals $\mu = xz, yz, xy$. The $xy$ orbital yields strong F spin coupling (1b) for higher values of $xy$ orbital offset energy, which sharply decreases in magnitude, even changing sign to AF spin coupling, with decreasing $xy$ orbital offset energy. This reduction is evidently related to the $xy$ orbital approaching half filling, which strongly suppresses the F spin coupling generated by the particle-hole exchange.

We now address the origin of the strong F spin coupling generated in the $xy$ orbital sector as seen in figure 3(b). We note that the $xy$ orbital sector is partially filled in the $(\pi, 0)$ AF state (figure 2(a)) due to the strong hybridization with $xz/yz$ orbitals. Furthermore, the (negative) F spin coupling magnitude ($xy$) is maximum for highest $\varepsilon_{xy}$ (figure 3(b)) where the $xy$ electronic density is maximally depleted ($n_{xy} \approx 0.75$), and with decreasing $\varepsilon_{xy}$, as the $xy$ orbital approaches half filling ($n_{xy} \approx 1$), the F spin coupling component decreases substantially, resulting in the net spin coupling changing sign from negative to positive. We therefore attribute the generation of strong F spin coupling in the $b$ direction to particle-hole processes in the partially filled $xy$ orbital sector, as in band ferromagnets with partially filled bands.

As also seen from figure 3(b), the AF spin coupling (1a) for the half-filled $yz$ orbital shows no variation. For the $xz$ orbital, the strongly AF spin coupling (1b) near half filling decreases substantially with $\varepsilon_{xy}$ as the $xz$ band becomes over-filled (figure 2(a)), indicating F contribution from the additional states transferred from the $xy$ band. This reduced AF spin coupling in the ferromagnetic (b) direction also contributes to the stabilization of the $(\pi, 0)$ state by suppressing magnetic frustration.

Each spin coupling term (9) involves altogether nine terms corresponding to $\mu' \nu' = xz, yz, xy$, out of which eight terms involve the Hund’s rule coupling term $J$ in the effective spin couplings. Figure 4 shows the total (orbital summed) spin couplings $J_\delta = \sum_{\mu \nu} J_\delta^{\mu \nu}$ evaluated with and without $J$ for the three neighbors. The significant reduction (to nearly half in all three cases) when $J$ is set to zero highlights the importance of the Hund’s rule coupling term in

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**Figure 2.** Variation of (a) electronic density $n_{\mu}$ for the three orbitals and (b) spin wave energy at the F and AF zone boundaries with the $xy$ orbital energy offset $\varepsilon_{xy}$.

**Figure 3.** Variation of (a) the total (orbital summed) spin couplings $J_\delta$ and (b) the individual orbital contributions $J_\delta^{\mu \nu}$ to the effective spin couplings for the first and second neighbors with the $xy$ orbital energy offset $\varepsilon_{xy}$.
4. Quantum corrections to sublattice magnetization

In the AF ground state of the half-filled Hubbard model in two dimensions, quantum spin fluctuations substantially reduce the sublattice magnetization to nearly 60% of the classical (HF) value in the strong-coupling limit \((U/t \rightarrow \infty)\). We will therefore examine quantum corrections to sublattice magnetization for the individual orbitals in the \((\pi, 0)\) AF state, the relative magnitudes for the over-filled \((xz)\), half-filled \((yz)\) and under-filled \((xy)\) 3d orbitals being of particular interest. We will follow the approach in terms of transverse spin correlations which were applied over a wide range of interaction strengths in the Hubbard model and shown to interpolate properly to the strong coupling limit [38].

Expanding the spin identity \(S = \frac{1}{2} (S^+ S^- + S^z S^z)\) for the spin \(S\) system in terms of \(1/S\) and retaining the first-order terms, we obtain

\[
\langle S^z \rangle = \frac{1}{S} \left[ 1 - \frac{1}{2S} \left( \langle S^+ S^- + S^- S^+ \rangle \right) - 1 \right].
\]

Extending this approach to a multi-orbital antiferromagnet having spin operators \(S_{\mu}\) for orbital \(\mu\), with \(S\) replaced by \(\langle S_{\mu}^0 \rangle\) as effective spin quantum numbers, the corrected sublattice magnetization for orbital \(\mu\) is obtained as:

\[
\langle 2S_{\mu}^0 \rangle = m_{\mu} = m_{\mu}^{HF} - \delta m_{\mu}^{SF},
\]

where the first-order, quantum spin-fluctuation corrections:

\[
\delta m_{\mu}^{SF} = \left\{ \left( \frac{S_{\mu}^0 S_{\nu}^0 + S_{\nu}^0 S_{\mu}^0}{S_{\mu}^0 S_{\nu}^0} - 1 \right) - \left( \frac{S_{\mu}^0 S_{\nu}^0 + S_{\nu}^0 S_{\mu}^0}{2S_{\mu}^0} \right) - 1 \right\}
\]

The transverse spin correlations above (equal-time, same-site) are evaluated from the retarded part of the RPA level transverse spin fluctuation propagator:

\[
\langle S_{\mu}^0(t) S_{\nu}^0(t' \rightarrow t^-) \rangle = \sum_{q} \int_{0}^{\infty} - \frac{d\omega}{\pi} \text{Im} \chi^{-1}(q, \omega) \delta_{\mu\nu}^{A}
\]

\[
\langle S_{\mu}^0(t) S_{\nu}^0(t' \rightarrow t^-) \rangle = \sum_{q} \int_{0}^{\infty} - \frac{d\omega}{\pi} \text{Im} \chi^{-1}(q, \omega) \delta_{\mu\nu}^{B}
\]

for lattice site \(i\) on \(A\) sublattice, the last equation following from spin-sublattice symmetry in the AF state which relates correlations on \(A\) and \(B\) sublattices via \(\langle S_{\mu}^0 S_{\nu}^0 \rangle_A = \langle S_{\mu}^0 S_{\nu}^0 \rangle_B\).

For evaluating quantum corrections, the \(q\) summation over the BZ in equation (14) was performed over a \(60 \times 60\) grid as earlier. The \(\omega\) integral was performed by evaluating the transverse spin fluctuation spectral function for 1000 \(\omega\) points extending up to 4 eV to ensure that particle-hole and optical mode excitations up to the highest energies were included. The momentum integrated spin wave spectral function evaluated from equation (5):
summed) quantum correction to sublattice magnetization \( \sum_{\mu} \delta m_{\mu} = 0.73 \), and the total HF level sublattice magnetization \( \sum_{\mu} m_{\mu}^{HF} = \sum_{\mu}(2S_{\mu}^{z}) = 1.95 \), the orbital-averaged reduction in sublattice magnetization due to spin fluctuation induced quantum corrections in the \((\pi, 0)\) AF state of the three-orbital model is quite significant at about 37%. The sublattice magnetization reduced to nearly half of the HF value is in agreement with experimentally measured values of \( \sim 1\mu_B \) for the Fe moment in the 122 family of iron pnictides [3].

5. Conclusions

Originating from particle-hole exchange, effective spin couplings \( J_{\mu\nu} \) for first and second neighbors were evaluated in the \((\pi, 0)\) magnetic state of a realistic three-orbital interacting electron model for iron pnictides involving \( xz, yz, xy \) Fe 3d orbitals. Variation of these spin couplings with the \( xy \) orbital energy offset provides valuable insight into the multi-orbital quantum antiferromagnetism in these compounds. The \( xy \) orbital was found to be mainly responsible for the generation of strong F spin coupling between first neighbors in the \( b \) direction, which is critically required to fully account for the spin wave dispersion measured from inelastic neutron scattering experiments. The F spin coupling is strongly suppressed with decreasing orbital order as the \( xy \) band approaches half filling, and is ascribed, as in band ferromagnets, to particle-hole exchange in the partially filled \( xy \) band. This study provides the microscopic basis for the negative (ferromagnetic) exchange interaction \( J_{1b} < 0 \) as considered in phenomenological spin models.

Significantly, the strongest AF spin coupling between first neighbors in the \( a \) direction lies in the orbital off-diagonal sector involving the \( xz \) and \( xy \) orbitals, highlighting the important role of strong orbital hybridization on effective spin couplings. While the AF spin coupling in \( a \) direction for the half-filled \( yz \) orbital was found to be constant with increasing \( xy \) orbital energy offset, the frustrating AF spin coupling in \( b \) direction for the \( xz \) orbital was found to decrease, thus reducing the magnetic frustration substantially. The Hund’s rule coupling term was found to contribute significantly to the effective spin couplings and therefore to the overall stabilization of the \((\pi, 0)\) magnetic state.

The first-order quantum corrections to sublattice magnetization due to quantum spin fluctuations were evaluated from transverse spin correlations. The maximum reduction was found for the over-filled \( xz \) orbital, which was more than double that of the minimum reduction for the half-filled \( yz \) orbital. The orbital-averaged reduction in sublattice magnetization due to quantum spin fluctuations in the \((\pi, 0)\) AF state of the three-orbital model was found to be quite significant at about 40%. The calculated sublattice magnetization including the quantum correction (nearly half of the HF value) is in agreement with the measured values of \( \sim 1\mu_B \) for the Fe moment in the 122 family of iron pnictides.

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Table 4. Transverse spin correlations and sublattice magnetization quantum corrections for the three orbitals.

| \( \mu \) | \( \langle S_{\mu}^{x}S_{\mu}^{y} \rangle \) | \( \langle S_{\mu}^{x}S_{\mu}^{z} \rangle \) | \( \langle (S_{\mu}^{x}S_{\mu}^{z})^{2} \rangle \) | \( m_{\mu}^{HF} \) | \( \delta m_{\mu} \) |
|---|---|---|---|---|
| \( xz \) | 0.64 | 0.888 | 0.55 | 0.57 | 0.32 |
| \( yz \) | 0.79 | 0.054 | 0.73 | 0.75 | 0.15 |
| \( xy \) | 0.70 | 0.081 | 0.62 | 0.63 | 0.26 |
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