Spin-wave excitations in the spin-density wave state of doped iron pnictides

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Abstract

We investigate spin-wave excitations in the spin-density wave state of doped iron pnictides within a five-orbital model. We find that the excitations along \((\pi, 0) \to (\pi, \pi)\) are very sensitive to dopings whereas they do not exhibit a similar sensitivity along \((0, 0) \to (\pi, 0)\). Secondly, the ellipticity of the elliptical ring-like excitations around \((\pi, 0)\) is also very much dependent on doping. Thirdly, the spin-wave spectral weight shifts towards the low-energy region as it moves away from zero doping. We find several features to be in qualitative agreement with the inelastic neutron-scattering measurements for the doped pnictides.

Keywords: five-orbital model, spin-wave excitations, doped iron pnictides

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

1. Introduction

Iron pnictides exhibit a very rich temperature-doping phase diagram, where the doping of either electrons or holes suppresses the long-range collinear magnetic order, giving way to sign-changing s-wave superconductivity (SC). The magnetic order in some of these materials is stabilized over a range of dopings \(x \lesssim 0.06\) and \(x \lesssim 0.3\) in the electron- and hole-doped regions, respectively [1, 2]. Despite the competing SC and magnetic long-range order with further doping, the superconducting state retains the spin fluctuations responsible for the pairing in a manner similar to that in high-\(T_c\) cuprates [3, 4]. Experimentally, the role of spin fluctuations in mediating SC can be probed with the help of inelastic neutron scattering (INS)—a powerful experimental tool [5].

The origin of the magnetic order in these materials lies in the Fermi surface (FS) instability because of the good nesting present between the Fermi pockets. According to angle-resolved photoemission spectroscopy (ARPES) as well as band structure calculations, FSs consist of concentric hole pockets around \((0, 0)\) and an elliptical electron pocket at \((\pi, 0)\) [6–13]. Nesting between these two sets of pockets leads to the \((\pi, 0)\) spin-density wave (SDW) state or collinear magnetic order. When electrons or holes are doped, FSs can be modified in a significant manner, thereby altering the nature of the nesting, and that can have a significant impact on the nature of the SDW state, as well as on the spin fluctuations responsible for the SC [14].

A remarkable high-energy excitation scale has been observed using INS in the SDW state of the parent compound, which is highly dispersive as well as sharp [15–19]. It can extend up to \(\sim 200\) meV, corresponding to the zone-boundary modes \(q = (\pi, \pi)\). There exists an in-plane anisotropy [17], which persists even in the nematic phase [20]. Excitations have also been studied extensively for various dopings, though in the superconducting state. For instance, the spin excitation is manifested as a resonance in the superconducting state [21, 22] because of its dependence on the BCS coherence factors for the wavevector equal to the nesting vector and the opposite signs of the superconducting gap on the electron and hole pockets. Spin-wave dispersion similar to that of the parent compound \(\text{BaFe}_2\text{As}_2\) has been observed along the high-symmetry directions for the superconducting \(\text{Ba}_{0.67}\text{K}_{0.33}\text{Fe}_2\text{As}_2\) with hole doping \(x \approx 0.33\), though with a significant zone-boundary softening [14, 23]. At the same time, magnetic-exchange coupling is reduced by 10%. High-energy spin excitations are suppressed and magnetic spectral weight is shifted to low energies. The excitations are gapped below \(\sim 50\) meV for the electron-doped \(\text{BaFe}_{1.7}\text{Ni}_{0.3}\text{As}_2\), while those with high-energy are largely unaffected. It has been further suggested that both the low- and high-energy excitations may be associated with SC [14].
For the SDW state, various theoretical and experimental studies have focused largely on spin excitations in the parent compound. The spin-wave dispersion has been described within the conventional Heisenberg model with anisotropic exchange couplings [24]. However, such a description cannot explain the spin-wave damping resulting from particle–hole excitations in the metallic SDW state, and the limitation can be overcome by considering additional terms which account for the bandstructure and coupling between the spin of the band electrons and the localized spin through Hund’s coupling [25, 26]. Several studies give an estimate of the intraorbital Coulomb interaction \((U)\) to be \(U/W \approx 0.25\), where \(W\) is the bandwidth [27, 28]. Therefore, a completely itinerant approach is best suited to describe the spin excitations in these materials.

Two different types of itinerant models—excitonic [29, 30] and orbital [31–34]—have often been employed to understand spin-wave excitations as well their damping. The description of various characteristics of excitations has been challenging in view of the fact that the observed magnetic moments are small while the excitations are sharp and disperse up to 200 meV. Nonetheless, several characteristics have been captured within the five-orbital models [31, 34]. Recently, an orbital model has been used to study collective magnetic excitations of \(C_4\) symmetric magnetic states [35]. It has also been shown that a large Hund’s coupling plays an essential role in describing different features, such as sharpness, anisotropy around \((\pi, 0)\), and the spin-wave spectral function [36]. Another important factor that has a significant impact on the aforementioned features is the doping-induced modification in the bandstructure, which has not attracted much attention.

In this paper, we examine various aspects of spin-wave excitations in the \((\pi, 0)\) SDW state of doped iron pnictides within the doping range \(-0.4 \lesssim x \lesssim 0.05\). For this, we consider the five-orbital tight-binding model of Ikeda et al [37]. The model with a rigid bandshift is known to exhibit the \((\pi, 0)\) SDW state within the doping range of \(0 \lesssim x \lesssim 0.18\) for intraorbital Coulomb interaction \(U \sim 0.9\) eV [38], and the range can be expected to increase for relatively larger interaction parameters. The reconstructed FSs agree qualitatively with the ARPES measurements [9]. The quasiparticle interference obtained with the reconstructed bands within the model has reproduced several features of the local density of states modulation in the doped state [39, 40].

We use the above model to highlight three important consequences of doping on the spin-wave excitations. (i) The excitations along \((\pi, 0) \rightarrow (\pi, \pi)\) are very sensitive to doping and a similar sensitivity is absent along \((0, 0) \rightarrow (\pi, 0)\). (ii) Anisotropy around \((\pi, 0)\) in the form of an elliptical structure decreases towards the hole-doped region for low-energy excitations. It becomes minimal near \(n \approx 5.9\) and increases again on moving further into the hole-doped region. On the other hand, the elliptical shape of the anisotropic excitations becomes deformed for high-energy excitations, especially in the hole-doped region. (iii) The spin-wave spectral weight shifts towards the low-energy region on moving away from zero doping.

### 2. Transverse-spin fluctuations

In order to investigate the doping dependence of spin-wave excitations, we consider the following mean-field Hamiltonian in the \((\pi, 0)\) SDW state

\[
\hat{H}_\sigma = \sum_k \Psi_{k\sigma}^\dagger \left[ \hat{\epsilon}_k + N \frac{\text{sgn} \Delta}{\text{sgn} \Delta} \hat{\epsilon}_{k+Q+\tilde{N}} \right] \Psi_{k\sigma}. \tag{1}
\]

Here, \(\Psi_{k\sigma}^\dagger = (d_{k\uparrow}^\dagger, \ldots, d_{k\sigma}^\dagger, d_{k\downarrow}^\dagger, \ldots, d_{k\bar{\sigma}}^\dagger)\) is the electron creation (destruction) operator for the momentum \(k\) in the orbital \(i\) with spin \(\sigma\). \(i\) belongs to the set of five \(d\) orbitals of the iron atom. \(\hat{\epsilon}_k\) is the hopping matrix corresponding to the five-orbital model. \(Q = (\pi, 0)\) is the ordering wavevector. The elements of the matrices \(\Delta\) and \(\tilde{N}\) as described in the appendix are dependent on the onsite interaction parameters, orbital magnetizations, and charge densities. The Hamiltonian matrix is diagonalized and various order parameters are obtained in a self-consistent manner using eigenvalues and eigenvectors. Then, the eigenvalues and eigenvectors corresponding to the self-consistent SDW state are used to calculate the spin-wave excitations.

The transverse-spin susceptibility within the random-phase approximation can be obtained as

\[
\chi_{\text{RPA}}(q, i\omega_n) = \hat{\chi}(q, i\omega_n)(1 - U\hat{\chi}(q, i\omega_n))^{-1}, \tag{2}
\]

where \(1\) is a \(2n^2 \times 2n^2\) identity matrix with \(n\) as the number of orbitals. The elements of the block-diagonal matrix \(U_{\mu\nu, \mu'\nu'} = U, U - 2J, J\) and \(J\) for \(\mu = \mu' = \nu = \nu'\), \(\mu_1 = \mu_2 \neq \nu_3 = \nu_4\), \(\mu_1 = \mu_2 \neq \nu_1 = \nu_4\) and \(\mu_1 = \mu_2 \neq \nu_3 = \nu_4\), respectively; they vanish otherwise. \(U\) and \(J\) are the intraorbital and interorbital Coulomb interactions. The bare-level susceptibility matrix is

\[
\hat{\chi}(q, i\omega_n) = \left[ \hat{\chi}(q + Q, i\omega_n) \hat{\chi}(q + Q, i\omega_n) \right], \tag{3}
\]

where the elements in the ordered state are given by

\[
\chi_{\alpha\beta, \mu
u}(q, q, i\omega_n) = \chi_{\alpha\beta, \mu
u}(q, q, i\omega_n) + \chi_{\alpha\beta, \mu
u}(q, q, i\omega_n)
+ \chi_{\alpha\beta, \mu
u}(q, q, i\omega_n) + \chi_{\alpha\beta, \mu
u}(q, q, i\omega_n)
\]

including the Umklapp processes. The bar notation over the orbital indices corresponds to the shifting of momentum by \(Q\), as mentioned earlier with regard to equation (1). The physical transverse-spin susceptibility corresponding to the spin operators to be defined below is

\[
\chi^\mu(q, i\omega_n) = \sum_{\alpha\mu} \chi_{\alpha\mu}(q, q, i\omega_n). \tag{5}
\]

The transverse-spin susceptibility is defined as

\[
\chi_{\alpha\beta, \mu
u}(q, q', i\omega_n) = \int^3 d\tau e^{i\omega_n \tau} \langle T_\tau \hat{S}_{\alpha\beta}^\mu(q, \tau)\hat{S}_{\mu\nu}^\nu(-q', \tau) \rangle, \tag{6}
\]
Here, $\langle ... \rangle$ denotes thermal average, $T_\tau$ imaginary time ordering, and $\omega_n$ are the Bosonic Matsubara frequencies. The components of the spin operator (corresponding to the physical spin susceptibility) are

$$S_q^i = \sum_k \sum_{\mu\sigma} d_{\mu\sigma}^i(k + q) E_{\mu\sigma}^i \sigma_{\mu\sigma} d_{\mu\sigma}^i(k). \tag{7}$$

$E$ is a $5 \times 5$ identity matrix belonging to the orbital bases. $\sigma$'s are the Pauli matrices corresponding to the spin degree of freedom. Thus, the susceptibility when $q' = q$ is given in terms of Green’s functions as

$$\chi_{\alpha\beta,\mu\nu}(q, q, i\omega_n) = -\frac{T}{N} \sum_{k, \omega_n} G_{\alpha\beta}^{\mu\nu}(k + q, i\omega_n + i\omega_n) G_{\nu\beta}^{\mu\nu}(k, i\omega_n)$$

$$= \sum_k \sum_{\mu\nu} a_{k\mu}^\dagger a_{k\nu}^\dagger a_{k+q\mu} a_{k+q\nu}$$

$$\times \frac{n_f(\epsilon_k + i\eta)}{\omega_n + \epsilon_k + i\eta}$$

where $a_{k\sigma}$'s are the unitary coefficients of the $\nu$-orbital obtained from diagonalizing the Hamiltonian matrix in equation (1). $n_f(\epsilon)$ is the Fermi function with $\epsilon$ as eigen energy.

In the following, the analytic continuation $i\omega_n \rightarrow \omega + i\eta$ with $\eta = 2$ meV is used throughout. Interaction parameters $U$ and $J$ are set to be 1.1 eV and 0.25$U$ so as to obtain magnetic moment $m \sim 1$ for zero doping, which is motivated by the observed magnetic moments in the 122 series of pnictides [41]. The unit of energy is set to be eV unless stated otherwise.

3. Results

FSs obtained in the unordered state for various dopings (a) $n = 5.6$, (b) $n = 5.8$, (c) $n = 6.0$, and (d) $n = 6.05$ are shown in figure 1. As can be seen, the nesting deteriorates fast on moving away from zero doping because the electron and hole pockets shrink and expand on moving from the electron-doped to the hole-doped region, respectively. Finally the electron pockets disappear, hinting at a Lifshitz transition [42, 43]. In fact, we also find a similar disappearance of the electron pockets in the SDW state along $k_x = 0$ near $n = 5.96$. Figures (e)-(h) show the reconstructed FSs in the SDW state for the respective dopings. It is evident that the reconstructed structure and topology of the FSs are very sensitive to doping when compared to the unordered state.

Figure 2 shows Im$\chi_{\mu\nu,\rho\sigma}^{RPA}$ obtained in the SDW state for various dopings (a) 5.6, (b) 5.7, (c) 5.8, (d) 5.9, (e) 6.0, and (f) 6.05. Note that the red color also represents those values of Im$\chi_{\mu\nu,\rho\sigma}^{RPA}$ that exceed 200. The excitations are heavily damped throughout, particularly along $1$–$X$–$M$ ((0, 0) $\rightarrow$ ($\pi$, 0) $\rightarrow$ ($\pi$, $\pi$)) for the electron doping $n = 6.05$, which is not surprising because there is a fast reduction in the net magnetization (figure 3), and hence of the magnetic-exchange gap on electron doping. That is also reflected in the particle–hole continuum extending down to low energies in the bare susceptibility (figures 2 (g)-(l)). The nesting between the electron pocket and the hole pocket in the unordered state is optimal in the vicinity of $n \approx 6.0$. Therefore, moving away from this band filling is expected to lead to a reduced magnetic-exchange gap according to the nesting-based scenario, though the reduction may not necessarily be symmetrical with respect to zero doping.

Spin-wave damping reduces quickly near the hole-doped region and the excitations become optimally sharp and well-defined for $q \approx 5.9$. In an earlier work, the maximum $T_c$ for the SDW state has also been shown to occur within the model for small hole doping [38]. By doping the holes further, the spin-wave excitations get softened rapidly along $X$–$M$, especially in the region close to M. Meanwhile, damping also
Figure 2. Imaginary part of the RPA-level physical transverse-spin susceptibility $\text{Im} \bar{\chi}_{\text{RPA}}^{\text{ps}}(q, \omega)$ calculated for $U = 1.1, J = 0.25U$ and several values of $n = (a) 5.6, (b) 5.7, (c) 5.8, (d) 5.9, (e) 6.0, and (f) 6.05$. The imaginary part of the bare spin susceptibility $\text{Im} \bar{\chi}_{\text{ps}}(q, \omega)$ calculated for the same set of parameters $n = (g) 5.6, (h) 5.7, (i) 5.8, (j) 5.9, (k) 6.0, and (l) 6.05$.

Figure 3. Orbital-resolved (a) magnetizations and (b) charge densities as a function of electron and hole dopings.
increases. Finally, the excitations disappear for \( n \approx 5.75 \) in a large part of \( X-M \). However, they remain largely unaffected along \( \Gamma-X \).

The magnetic moment grows on moving from the electron-doped region to the hole-doped region (figure 3) and so do the SDW gaps. This is not surprising because one approaches the half-filling or high-spin state \( S = 5/2 \) due to the strong Hund’s coupling. Thus, the largest intra-orbital SDW gap increases from \( \Delta_{xy} \sim 0.25 \) in the undoped case up to \( \Delta_{xy} \sim 0.5 \) when \( n = 5.6 \). SDW gaps of a similar magnitude have been obtained in the undoped cases within various orbital models, including the two- and three-orbital models. It is noteworthy that a significant reduction, as large as 40% in the sublattice magnetization, can occur due to quantum corrections, as illustrated in a three-orbital model [44].

Recent x-ray experiments support the scenario where the local moments grow with hole doping [45]. This, in turn, is
expected to suppress the density of states at the Fermi level and the electron movement in the lattice leading to the departure from metallicity [46]. It can be seen that the FSs exist for all dopings considered here because of a small ratio \( U/W \approx 0.25 \) with \( W \) as the electron bandwidth, and as a result the reconstructed bandstructure plays a crucial role in the spin-wave excitations. However, the system being away from half filling \( n = 5.0 \) is also partly responsible for the existence of the Fermi surfaces. For that very reason, a gap in the single particle spectrum may not appear for even larger \( U \); however, orbital-selective Mott transition is not ruled out [46].

Consequently, despite the large magnetic moments or SDW gaps, and unlike the excitonic scenario, the particle–hole continuum is not completely gapped due to the existence of the Fermi surfaces in the SDW state for the various dopings considered. In particular, the continuum can be seen to extend down to \( \sim 100 \) meV in several regions, especially at higher doping near \( n = 5.6 \). This leads to finite damping of the spin waves. In particular, we find that the particle–hole continuum in the bare-spin susceptibility for \( U/W \approx 0.25 \) shifts towards a low energy near \( M \), which should play a very important role in the damping of spin-wave excitations along \( X-M \) in the hole-doped region.

Figures 4 and 5 show the constant energy cuts for the band-filling \( n = 5.7, 5.9, 6.0 \) and 6.05, respectively. The anisotropy in the spin-wave excitations around \( X \) with an elliptical shape is sensitive to both energy as well as doping. The elliptical shape of the anisotropy is modified quickly as the energy is increased, particularly in the hole-doped region. On the other hand, the ellipticity decreases for the low-energy excitations \( \sim 50 \) meV when the holes are doped. It is minimal at \( \approx 5.9 \) (also figures 2(a)–(f)), and grows again on moving further into the hole-doped region. The anisotropic behavior around \( X \) at low energies, as well as the square-like shape of the excitations around \( M \) at higher energies, particularly when \( n \approx 5.9 \), is qualitatively similar to what is observed in the INS experiments [5, 14].

Figure 6 shows the doping dependence of the spin-wave spectral function \( \chi^{\prime\prime}(\omega) = \sum_{q} \text{Im} \chi_{\eta,0,0}^{\eta,0,0}(q, \omega) \). There are two separate peaks for \( n = 6.05 \)—one positioned near very small energy, and the other one around 200 meV. At zero doping, the peak near 200 meV becomes prominent, whereas the position of the peak shifts rapidly towards the low-energy region with hole doping. A similar observation has been made in the paramagnetic and superconducting phases of doped pnictides [14]. Moreover, the location of the peak in the spin-wave spectral function for optimal doping remains near 200 meV, which is also in accordance with the experiments.

4. Conclusions and discussions

In conclusion, we have investigated spin-wave excitations in the SDW state of doped iron pnictides. We focused on the role of change in the electronic bandstructure when either electrons or holes are doped, and doping-induced disorder is ignored for simplicity. We used a five-orbital model with a realistic electronic structure and a fixed set of interaction parameters corresponding to the magnetic moment \( m \approx 1 \) in the undoped case. We find that the excitations along \( (\pi, 0) \rightarrow (\pi, \pi) \) are very sharp and dispersive in a small doping range centered around \( n \approx 5.9 \) lying in the hole-doped region, and they become heavily damped on moving away from that doping on either side. Unlike \( (\pi, 0) \rightarrow (\pi, \pi) \), the excitations along \( (0, 0) \rightarrow (\pi, 0) \) do not show much variation with doping except in the electron-doped region, where they are damped heavily.

The spin-wave excitations around \( (\pi, 0) \) for low energy \( \sim 50 \) meV have an elliptical ring-like structure. With doping holes, the ellipticity first decreases and becomes minimum near \( n \approx 5.9 \), then increases again on moving further into the hole-doped region. On the other hand, the elliptical structure of the anisotropic excitations becomes deformed quickly with increasing energy, especially in the hole-doped region. We note that the anisotropy arises due to the imperfect nesting between the circular hole pocket and the elliptical electron pocket. Thus, when the nesting is optimal near \( n \approx 5.9 \), the anisotropy at low energy becomes minimum. In other words, the spin-wave anisotropy can provide valuable information regarding the relative structures of the electron and hole pockets.

The spin-wave spectral weight shifts towards lower energy when either the holes or electrons are doped, whereas it peaks at \( \approx 200 \) meV near zero doping. The origin of this behavior lies in the spectral-weight of the particle–hole continuum, which is suppressed at low energy on moving towards optimal doping.

In several doped pnictides, there is a phase transition from the high-temperature SDW state to the low-temperature superconducting state within a certain doping range. Some of the characteristics of the spin excitations, such as anisotropy around \( (\pi, 0) \), are expected to be retained across the phase transition in a manner similar to the phase transition from the paramagnetic to SDW state. We find several features, such as anisotropy around \( (\pi, 0) \), softening of zone-boundary modes, and shifting of the spectral weight on doping to the low-energy region, are qualitatively similar to those measured in the INS for the paramagnetic and superconducting phases.

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Appendix

The kinetic part of the model Hamiltonian that we consider is given by

\[
\mathcal{H}_0 = \sum_k \sum_{\mu, \nu} \sum_{\sigma} \varepsilon_{k, \mu, \nu} d_{k, \mu, \sigma}^\dagger d_{k, \nu, \sigma},
\]

(A.1)

where \( d_{k, \mu, \sigma}^\dagger \) (\( d_{k, \mu, \sigma} \)) are the electron creation (destruction) operators and \( \varepsilon_{k, \mu, \nu} \) are the hopping elements from orbital \( \mu \) to \( \nu \) for the momentum \( k \). \( \mu, \nu \) and \( \sigma \) belong to the set of five \( d \) orbitals.

The interaction term is given by

6
\[ \mathcal{H}_{\text{int}} = U \sum_{i<\nu} n_{i\mu\uparrow} n_{i\nu\downarrow} + \left( U' - \frac{J}{2} \right) \sum_{\nu<\mu} n_{i\mu\downarrow} n_{i\nu\uparrow} \]
\[ - 2J \sum_{i<\nu} \mathbf{S}_{i\mu}, \mathbf{S}_{i\nu} + J \sum_{\mu<\nu}(d_{i\mu\sigma}^\dagger d_{i\nu\sigma}^\dagger d_{i\nu\downarrow} + \text{h.c.}), \]
(A.2)

It has the intra- and inter-orbital Coulomb interaction terms as the first and second terms, respectively. The third and fourth terms represent Hund’s coupling and pair hopping. The Hamiltonian possesses only two independent interaction parameters due to the rotational invariance condition \( U = U' - 2J \).

Matrix elements of matrices \( \hat{\Delta} \) and \( \hat{N} \) in equation (1) are
\[ 2\Delta_{\mu\mu} = Um_{\mu\mu} + J \sum_{\nu \neq \mu} m_{\nu\mu} \]
\[ 2\Delta_{\mu\nu} = Jm_{\mu\nu} + (U - 2J)m_{\nu\mu} \]
(A.3)

and
\[ 2N_{\mu\mu} = Un_{\mu\mu} + (2U - 5J) \sum_{\nu \neq \mu} n_{\nu\mu} \]
\[ 2N_{\mu\nu} = Jn_{\mu\nu} + (4J - U)n_{\nu\mu}, \]
(A.4)

where charge densities and magnetizations are given by
\[ n_{\mu\sigma} = \sum_{k\sigma} \langle d_{k\mu\sigma}^\dagger d_{k\mu\sigma} \rangle, \quad m_{\mu\sigma} = \sum_{k\sigma} \langle d_{k\mu\sigma}^\dagger d_{k\nu\sigma} \rangle, \]
(A.5)

where \( d_{k\mu\sigma}^\dagger = d_{k+Q\mu\sigma}^\dagger \). The bar notation over the orbital indices corresponds to the shifting of momentum by \( Q \). k summation is in the first Brillouin zone.

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