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Spin waves in the (0, \( \pi \)) and (0, \( \pi, \pi \)) ordered SDW states of the \( t-t' \) Hubbard model: application to doped iron pnictides

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Abstract
Spin waves in the (0, \( \pi \)) and (0, \( \pi, \pi \)) ordered spin-density-wave (SDW) states of the \( t-t' \) Hubbard model are investigated at finite doping. In the presence of small \( t' \), these composite ferro–antiferromagnetic (F–AF) states are found to be strongly stabilized at finite hole doping due to enhanced carrier-induced ferromagnetic spin couplings as in metallic ferromagnets. Anisotropic spin-wave velocities, a spin-wave energy scale of around 200 meV, reduced magnetic moment and rapid suppression of magnetic order with electron doping \( x \) (corresponding to F substitution of O atoms in LaO\(_{1-x}\)F\(_x\)FeAs or Ni substitution of Fe atoms in BaFe\(_{2-x}\)Ni\(_x\)As\(_2\)) obtained in this model are in agreement with observed magnetic properties of doped iron pnictides.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Following the recent discovery of superconductivity [1–4] in doped iron pnictides such as RO\(_{1-x}\)F\(_x\)FeAs (R = La, Ce, Nd, Sm, Gd), there has been great interest in their magnetic properties as well. Single-crystal neutron scattering studies of their parent compounds have indicated a commensurate magnetic ordering with iron moments ordered ferromagnetically in the \( b \) direction and antiferromagnetically in the \( a \) and \( c \) directions [5]. Inelastic neutron scattering measurements in AFe\(_2\)As\(_2\) (A = Ca, Ba, Sr) yield sharp spin-wave excitations on an energy scale \( \sim 200 \) meV [6–8].

All known compounds in these classes contain FeAs layers and exhibit a common phase diagram, with parent compounds exhibiting a magnetically ordered SDW state below \( T_N \approx 200 \) K and onset of superconductivity following the suppression of long-range magnetic order by electron doping or pressure. Contrast with cuprates has naturally followed in order to gain insight from the significant progress made in understanding superconductivity and magnetism in correlated electron systems [9]. While the onset of superconductivity at finite doping is a common feature, in contrast with the Mott insulating nature of cuprates, the pnictides appear to be commensurate SDW systems in the intermediate coupling regime. Appreciable hybridization between Fe 3d orbitals and As 3p orbitals possibly reduces the effective correlation term \( U \) as compared to cuprates [10, 11]. Comparison with x-ray photoemission spectra of the calculated density of states within the LDA + DMFT approach supports the physical picture of a multiband metal with intermediate correlations [12, 13].

In cuprates, the intense interest in the nature of magnetic excitations in the quantum antiferromagnet, their coupling with hole motion and scrambling of local AF order, strongly incoherent hole spectral function and the possibility of pairing interaction mediated by exchange of local magnetic excitations have contributed significantly to obtaining an insight and understanding of their magnetic and electronic properties.

Therefore, an investigation of magnetic excitations in the (0, \( \pi \)) and (0, \( \pi, \pi \)) ordered SDW states within a minimal itinerant electron model should be of interest, particularly within an approach which is valid in the full
range of interaction strengths including the relevant weak and intermediate coupling regimes. In this paper, we will therefore consider the $t-t'$ Hubbard model and obtain the magnon energies in the F–AF states in two and three dimensions, focusing especially on the role of finite $t'$ and doping in stabilizing the F–AF order.

Spin waves in the $(0, \pi)$ and $(0, \pi, \pi)$ ordered states were investigated earlier in the undoped $t-t'$ Hubbard model [14] and have been recently investigated within the $J_{1x-J_2y-J_2z}$ Heisenberg models on a square lattice [15, 16]. Reduced magnetic moments and suppression of magnetic ordering have been investigated in terms of the associated magnetic frustration effect in such models [17, 18]. An itinerant description of spin-wave excitations in iron-based superconductors and comparison with available experiments has been investigated using multiband models involving excitonic instability of nested electron-like and hole-like Fermi pockets [19–23]. However, doping dependence of magnetic properties, especially the rapid suppression of magnetic ordering with electron doping, was not investigated. Recently, doping dependence of spin fluctuations and electron correlations have been theoretically investigated within an effective five-band Hubbard model for iron pnictides using the FLEX approximation [24].

2. The $t-t'$ Hubbard model

We consider the $t-t'$ Hubbard model on square and simple cubic lattices, with hopping terms $t$ and $t'$ between nearest-neighbour (NN) and next-nearest-neighbour (NNN) pairs of sites, respectively:

$$H = -t \sum_{\langle i,j,\sigma \rangle} n_i^{\dagger}\sigma a_{i+\delta x,\sigma} - t' \sum_{\langle i,j,\sigma \rangle} n_i^{\dagger}\sigma a_{i+\delta x,\sigma} + U \sum_i n_i^{\dagger}n_i^{\dagger},$$

(1)

The observed asymmetry in the antiferromagnetism of cuprates with respect to hole and electron doping [25, 26], the existence of metallic antiferromagnetism at half-filling [27, 28] and correlated motion of electrons in metallic ferromagnets in fcc and bcc lattices [29–31] exemplify the few physical systems which have been investigated in terms of the above model.

The two-sublattice representation of the AF state (corresponding to ordering wavevector $(\pi, \pi)$ in two dimensions) conveniently allows for the investigation of spin waves, quantum corrections, Néel temperature, hole dynamics, etc. In analogy, we will consider F–AF SDW states with $Q = (0, \pi)$ and $(0, \pi, \pi)$ involving ferromagnetic spin ordering in one direction and antiferromagnetic spin ordering in the remaining direction(s). The self-consistent-field (Hartree–Fock) approximation provides a convenient basis in which many-body correlation effects can then be systematically incorporated. The two-sublattice structure for the F–AF SDW states in two and three dimensions is given below.

2.1. $Q = (0, \pi)$ ordered state

Spins are ordered ferromagnetically along the $x$ direction and antiferromagnetically along the $y$ direction. NN hopping terms in the $x$ direction connect sites of the same sublattice, while NN hopping terms in the $y$ direction and NNN hopping terms connect sites of opposite sublattices. The HF Hamiltonian matrix in the two-sublattice basis:

$$H^x_{\text{HF}}(k) = \begin{pmatrix}
-\sigma \Delta + e_k^x & e_k^x + \epsilon_k^x \\
-\epsilon_k^x + e_k^x & \sigma \Delta + e_k^x
\end{pmatrix} = \eta_k^x \mathbf{1} + \begin{pmatrix}
-\sigma \Delta & \eta_k \\
\eta_k & \sigma \Delta
\end{pmatrix}$$

(2)

for electron spin $\sigma$, where $\eta_k^x \equiv e_k^x = -2t \cos k_x$ and $\eta_k \equiv e_k^x + e_k^x = -2t \cos k_y - 2t' \cos k_y \cos k_z$, and the self-consistent exchange field is given by $2\Delta = mU$ in terms of the sublattice magnetization $m$, resulting in SDW state band energies $E_{\text{sdw}}^{x,\tau} = \eta_k^x \pm \sqrt{\Delta^2 + \eta_k^x}^2$.

2.2. $Q = (0, \pi, \pi)$ ordered state

Spins are ferromagnetically ordered along the $x$ direction and antiferromagnetically ordered along the $y$ and $z$ directions. NN hopping terms in the $y-z$ plane connect sites of opposite sublattices, while those in the $x$ direction connect sites of the same sublattice. Similarly, NNN hopping terms in the $y-z$ plane connect sites of the same sublattice while those in the $x-y$ plane connect sites of opposite sublattices. The HF Hamiltonian matrix:

$$H^y_{\text{HF}}(k) = \begin{pmatrix}
-\sigma \Delta + e_k^y + \epsilon_k^{yz} & \epsilon_k^y + e_k^{yz} + e_k^{xz} \\
-\epsilon_k^y - e_k^{yz} & \sigma \Delta + e_k^y + e_k^{yz} + e_k^{xy}
\end{pmatrix} = \eta_k^y \mathbf{1} + \begin{pmatrix}
-\sigma \Delta & \eta_k \\
\eta_k & \sigma \Delta
\end{pmatrix}$$

(3)

where

$$\eta_k^y \equiv e_k^y + \epsilon_k^{yz} = -2t \cos k_x - 2t' \cos k_x \cos k_z$$

$$\eta_k \equiv e_k^y + e_k^{yz} + e_k^{xy}$$

$$\eta_k = [-2t - 2t' \cos k_x](\cos k_y + \cos k_z).$$

3. Magnon propagator

Magnon excitations in the F–AF state, corresponding to transverse spin fluctuations about the ordering direction (assumed $z$), are obtained from the time-ordered propagator of the transverse spin operators $S_i^x$ and $S_i^y$ at sites $x$ and $y$:

$$\chi^{-1}(q, \omega) = \int d\tau\sum_i \langle \Psi_G | T \{ S_i^x(\tau) S_i^y(\tau') \} | \Psi_G \rangle.$$  

(5)

In the random phase approximation (RPA), the magnon propagator:

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

(6)

where the bare particle–hole propagator $[\chi^{(0)}(q, \omega)]$ is evaluated by integrating out the fermions in the spontaneously broken-symmetry F–AF state. In the metallic state, $[\chi^{(0)}(q, \omega)]$ involves both inter-band and intra-band particle–hole processes [26].
4. Insulating \((0, \pi, \pi)\) state in the strong coupling limit

In the analytically simple strong coupling limit, the magnon propagator in the half-filled insulating \((0, \pi, \pi)\) state was evaluated earlier [14] and the RPA level magnon energies were obtained as

\[
\left( \frac{\omega_q}{2J} \right)^2 = \left[ \left( 1 + \frac{2J'}{J} \right) \right. \\
- \frac{1}{2} \left[ \left( 1 - \cos q_x \right) + \frac{2J'}{J} \left( 1 - \cos q_y \cos q_z \right) \right]^2 \\
- \left[ \left( 1 + \frac{2J'}{J} \cos q_z \right) \left( \cos q_x + \cos q_y \right) \right]^2 \\
\left. \right] \tag{7}
\]

which reduces to

\[
\left( \frac{\omega_q}{2J} \right)^2 \approx \frac{1}{2} \left( 1 + \frac{2J'}{J} \right) [\alpha q_x^2 + q_y^2 + q_z^2] \tag{8}
\]

in the long wavelength limit. The coefficient \(\alpha = \left( \frac{4J'}{J} - 1 \right)\) becomes negative for \(J'/J < 1/4\), signalling the instability of the F–AF phase at \(J'/J = 1/4\). Anisotropic spin-wave velocities naturally follow from the different \(q^2\) coefficients in the F and AF directions. Here \(J = 4t^2/\mathcal{U}\) and similarly for \(J'\).

As an illustration of quantum corrections beyond the HF approximation, the spin-fluctuation correction \(\delta m_{\text{SF}}\) to sublattice magnetization, evaluated from the magnon propagator in terms of transverse spin correlations [14], is shown in figure 1. The correction in the AF phase is also shown for comparison. Near the transition point \(J'/J = 1/4\), the correction in the F–AF phase is seen to be nearly half that in the AF phase, indicating greater robustness of the F–AF phase with respect to quantum spin fluctuations. The spin-fluctuation correction in both phases approaches one (the HF value of sublattice magnetization) only very close to the critical value \(J'/J = 1/4\). This implies that (up to first order) \(m\) vanishes only very close to \(J'/J = 1/4\), so that the extent of the spin-disordered phase is quite narrow. This is unlike the \(d = 2\) case, where AF order is lost at \(J'/J \approx 0.37\), well before the F–AF state appears at \(J'/J \gtrsim 0.5\).

5. Stabilization of the hole-doped F–AF state

As shown in section 4, AF NNN spin couplings \(J' = 4t'^2/\mathcal{U}\) stabilize the undoped F–AF state (for \(t'/t > 1/\sqrt{2}\) in two and \(t'/t > 1/2\) in three dimensions). AF NNN spin couplings are generated even in the absence of \(t'\), as in the \(t-U\) model at finite doping. Indeed, these effectively frustrating spin couplings are responsible for destabilizing the AF state at any finite doping, as observed in hole-doped cuprates [26]. However, as the F–AF state is actually stabilized rather than being frustrated by the AF NNN spin couplings, these doping-induced spin couplings should actually favour the F–AF state by supplementing the \(t'\)-induced spin couplings.

More importantly, carrier-induced F NN spin couplings responsible for metallic ferromagnetism become increasingly important at finite doping. This is especially so in the presence of small \(t'\), which can cause a strongly peaked electronic spectral distribution due to band dispersion saddle points \((\mathcal{V} \mathcal{V}_k = 0)\), which strongly enhance band ferromagnetism by increasing the delocalization contribution \(\sim (\mathcal{V} \mathcal{V}_k)^2\) to spin stiffness while strongly suppressing the correlation-induced exchange contribution \(\sim (\mathcal{V} \mathcal{V}_k)^2/\mathcal{U}\) due to correlated motion of electrons [31]. The F–AF state at finite doping is therefore expected to be stabilized at even smaller \(t'\) values.

Figure 2 shows the spin-wave energy along symmetry directions in the Brillouin zone for the doped F–AF state, with orderings \((0, \pi)\) and \((0, \pi, \pi)\), as considered earlier. The variation of wavevector \(q\) follows the sequence \((0, 0) \rightarrow (0, \pi) \rightarrow (\pi, \pi) \rightarrow (0, 0) \rightarrow (\pi, 0, 0) \rightarrow (\pi, \pi, \pi)\) (in (a)) and \((0, 0, 0) \rightarrow (0, \pi, \pi) \rightarrow (0, 0, 0) \rightarrow (\pi, 0, 0) \rightarrow (\pi, \pi, \pi)\) (in (b)). In order to focus on doping dependence, the SDW bands were kept unchanged with fixed \(\Delta/t = 4\). The rapid crossover from negative to positive energy magnon modes shows a strong stabilization of the F–AF state upon hole doping. This stabilization occurs for much smaller \(t'\) values compared to the critical values required in the undoped F–AF state. A finite \(t'\) is quite realistic in view of the hybridization between Fe and As orbitals.

The anisotropic spin-wave velocities, evident from the different slopes in the F and AF directions in figure 2(a), can be understood readily in terms of the independent origin (delocalization and exchange) of the effective F and AF spin couplings, resulting in different coefficients of \(q_x^2\) and \(q_y^2\), as in equation (8). Furthermore, for \(t \sim 200\) meV and \(\mathcal{U}\) in the intermediate coupling regime, the calculated spin-wave energy scale of around 200 meV is as observed in neutron scattering measurements of iron pnictides.

Are the doping-induced spin couplings sufficient to stabilize the F–AF state without any finite \(t'\)? We find that in the absence of \(t'\) the F–AF state is not stabilized for any doping. While AF and F orderings do get separately stabilized at low and high hole dopings, respectively, as indicated by the spin-wave dispersion along the AF and F directions, both are not simultaneously stabilized. Thus, small \(t'\) plays a crucial role in stabilizing the doped F–AF state with respect to transverse spin fluctuations in the entire Brillouin zone. A possible link between orthorhombic distortion and an effective NNN hopping \(t'\) would then explain why this distortion
The magnon energy scale also determines the finite-temperature magnetic properties. Magnon thermal excitation yields the fall off of magnetization with temperature and the magnetic ordering temperature. Thus, the rapid suppression of magnetic ordering with electron doping. The rapid magnon energy suppression and crossover to negative-energy modes indicating destabilization of the F–AF state provides an understanding of the observed reduced magnetic moment and rapid suppression of magnetic ordering in iron pnictides. Enhanced spin-fluctuation quantum correction in the vicinity of the magnetic instability point (as in figure 1) is also possibly important in reducing the magnetic moment.

6. Conclusions

Spin waves in the (0, π) and (0, π, π) ordered F–AF SDW states of the $t$–$t'$ Hubbard model were investigated at finite doping in the intermediate coupling regime. Including both inter-band and intra-band processes in the particle–hole propagator, spin-wave dispersion was obtained along different symmetry directions in the Brillouin zone. The F–AF state was found to be strongly stabilized at finite hole doping, as shown by the rapid crossover from negative to positive spin-wave energies, most noticeably in the F direction.

This stabilization was ascribed mainly to carrier-induced ferromagnetic spin couplings (along the F chains) as in metallic ferromagnets, and was found to be strongly enhanced in the presence of finite $t'$ due to the band structure saddle point effect. This calculated doping behaviour is in agreement with the observed rapid suppression of the magnetic order in doped pnictides such as LaO$_{1-x}$F$_x$FeAs on electron doping arising from F substitution of O atoms. The doping and $t'$ dependence of spin waves obtained within the minimal $t$–$t'$ Hubbard model accounts for many of the observed magnetic properties of iron pnictides, including anisotropic spin-wave velocities, spin-wave energy scale, reduced magnetic moment and rapid suppression of magnetic ordering with electron doping. The spin-wave velocity anisotropy is strongly enhanced on electron doping near the instability point.

The observed reduced magnetic moment in pnictides can be understood in terms of depletion of the predominantly magnetic states. Characteristic of the SDW state, electronic states at the top of the band are significantly more magnetic than states deeper in the band, especially in the weak coupling limit. Therefore hole doping of these states rapidly diminishes the local magnetic moment $m = 2\Delta / U$, which is reduced to $\approx 0.4$ at $x = 0$ in figure 3. While local moments will get enhanced on filling up these empty magnetic states by electron doping (F substitution of O), the rapidly diminished carrier-induced F NN spin couplings are then unable to sustain the F–AF ordering. This highlights the two distinctly different mechanisms behind the observed reduced magnetic moment.

Enhanced spin-fluctuation quantum correction in the vicinity of magnetic instability point (as in figure 1) is also possibly important in reducing the magnetic moment.

Figure 2. Spin–wave energy along symmetry directions in the Brillouin zone for F–AF states with (a) $(\pi, 0)$ and (b) $(0, \pi, \pi)$ ordering. Rapid crossover from negative to positive energy modes, especially in the F direction $\Gamma$–$M'$, shows a strong stabilization of F–AF states upon hole doping $y$. Here $t'/t = 0.5$ in (a) and 0.3 in (b).

Figure 3. Electron doping ($x$) dependence of the magnon energy at wavevector $q = (\pi/2, 0, 0)$, showing rapid suppression of magnon mode energy along the ferromagnetic ordering direction and destabilization of the F–AF state.

As the maximal sensitivity of magnon modes to doping is found along the ferromagnetic direction ($\Gamma$–$M'$), we have examined their doping dependence at an intermediate wavevector $q = (\pi/2, 0, 0)$. A pre-doped level with hole doping $y_0$ was taken to represent the partially filled band of the parent compound, with electron doping $x = y_0 - y$ defined as the reduction in hole doping from this level (representing F$^-$ substitution of O$^{2-}$ in the doped pnictides). Figure 3 shows the behaviour of the magnon mode energy with electron doping. The rapid magnon energy suppression and crossover to negative-energy modes indicating destabilization of the F–AF state provides an understanding of the observed rapid suppression of magnetic ordering temperature in iron pnictides with electron doping (due to F substitution of O atoms in LaO$_{1-x}$F$_x$FeAs or Ni substitution of Fe atoms in BaFe$_{2-x}$Ni$_x$As$_2$) [32]. Here, the interaction strength was fixed at $U = 8t$ in the intermediate coupling regime, and $y_0 = 0.35$.

The observed reduced magnetic moment in pnictides can be understood in terms of depletion of the predominantly magnetic states. Characteristic of the SDW state, electronic states at the top of the band are significantly more magnetic than states deeper in the band, especially in the weak coupling limit. Therefore hole doping of these states rapidly diminishes
of magnon energy with electron doping obtained in this paper accounts for the observed reduction of magnetic ordering temperature on F substitution of O atoms in LaO$_1$-$\delta$F,FeAs. Furthermore, spatially anisotropic magnetic couplings, with a small ratio $(J_\perp/J_\parallel = r < 1)$ of interlayer to planar magnetic couplings, reduces the ordering temperature to $\sim T_{c}^{iso}/\ln(1/r)$ compared to the isotropic case, which possibly accounts for the low ordering temperature observed in doped pnictides ($\sim 200$ K), as in the layered cuprate antiferromagnet La$_2$CuO$_4$, where $J \sim 1500$ K and magnon energy scale $\sim 250$ meV, but the Néel temperature is only about 400 K.

Correlation-induced quantum corrections to spin waves beyond RPA, investigated recently in multiband metallic ferromagnets [33] involving orbital degeneracy by incorporating self-energy and vertex corrections in a systematic Goldstone-mode-preserving approach, should be of interest, especially in view of recent magnetic form factor studies indicating that multiple d orbitals of iron atoms are occupied [34].

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