Pnictides as frustrated quantum antiferromagnets close to a quantum phase transition

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(Received 23 January 2009; published 23 March 2009)

We present a detailed description of the dynamics of the magnetic modes in the recently discovered superconducting pnictides using reliable self-consistent spin-wave theory and series expansion. Contrary to linear spin-wave theory, no gapless mode occurs at the Néel wave vector. We discuss the scenario that the static magnetic moment is strongly reduced by magnetic fluctuations arising from the vicinity to a quantum phase transition. Smoking gun experiments to verify this scenario are proposed and possible results are predicted. Intriguingly in this scenario, the structural transition at finite temperature would be driven by an Ising transition in directional degrees of freedom.

DOI: 10.1103/PhysRevB.79.092416 PACS number(s): 74.70.−b, 75.10.Jm, 75.40.Gb, 75.30.Ds

The seminal discovery of superconductivity in the pnictide family of materials at relatively high temperatures[1] has led to tremendous excitement and research activity. This discovery raises many fundamental questions. Foremost among them is whether high-temperature superconductivity in these materials is in some fundamental sense closely related to those in the cuprate family of materials. Indeed the quasi-two-dimensional layered structure for the two families and antiferromagnetism in the parent compounds suggests potential similarities. However, many doubts have also been raised about any correspondence, such as: is the origin and nature of spin fluctuations in the two families related given that the parent compounds are metallic in the pnictides whereas they are insulating in the cuprates? Are the pnictide materials even strongly correlated or are local density approximation based approaches adequate? Are spin models appropriate for describing spin fluctuations in these materials? In order to address these very basic questions, it is necessary to have detailed quantitative comparisons between theory and experiments.

On the experimental front, magnetic long-range order was established in $LaFeAsO_{1-x}F_x$ by neutron scattering (NS) (Ref. 2) and by muon spin resonance (μSR).[3] The NS provides evidence for a columnar antiferromagnetic ordering with a staggered magnetic moment of $0.36(5)\mu_B$. For simplicity we consider here only the square lattice which is formed by the Fe ions ignoring a small orthorhombic and even monoclinic structural distortion. Along the a axis the spin directions alternate whereas they are the same along the b axis; see Fig. 1(a). The μSR also provides evidence that the spin order is commensurate but with a small staggered moment of $0.25\mu_B$. First results on the dispersion of the magnetic excitations have just become available.[4,5] A tiny anisotropy gap is found to be ≈ 6 meV, the spin-wave velocity $v_\perp$ perpendicular to the stripes to be 205 ± 20 meV in units of $1/g$, where g is the inverse Fe-Fe distance, and a small interplane coupling J_z is found to be ≈ 5 meV. Results for the parallel spin-wave velocity $v_\parallel$ are not available so far, but they are expected soon.

Theoretically, the columnar antiferromagnetic ordering was also found to be the most stable in band-structure calculation.[6,7] So there is agreement on the static structure. But the smallness of the staggered magnetic moment is a matter of controversy. On the one hand, band-structure results indicate a local moment of up to $2.3\mu_B$.[7–9] This has led to the suggestion that the magnetic fluctuations themselves strongly reduce the static local moment.[10] We will show that this scenario implies that the pnictides are in the direct vicinity of a quantum phase transition. On the other hand, there are studies suggesting that the strong reduction in the local magnetic moment can be explained by electronic effects such as hybridization, spin-orbit coupling, and a particular low symmetry.[11,12] Hence, there are two different scenarios: (i) the local static moment is reduced by the magnetic fluctuations. In this case the ratio of couplings must be fixed to an appropriate value. (ii) The local electronic orbitals account for the sizable reduction so that the magnetic couplings are not determined by the value of the magnetic moment.

Together with upcoming experimental results, our work will help decide on the degree of strong correlation and on the closeness to a quantum phase transition.

The quantitative goal of the present work is threefold. First, we provide a theory for the magnetic excitations based

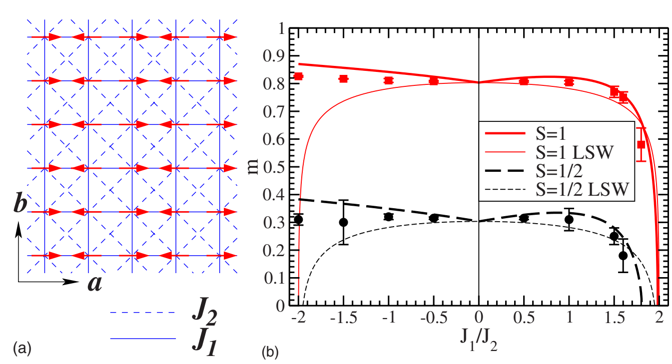

FIG. 1. (Color online) Panel (a) considered spin pattern; panel (b) staggered magnetization for $S=1/2$ and $S=1$ as function of the ratio of the couplings. For comparison the results of LSW theory are included. The self-consistent spin-wave theory is nicely supported by series expansion about the Ising limit (symbols).

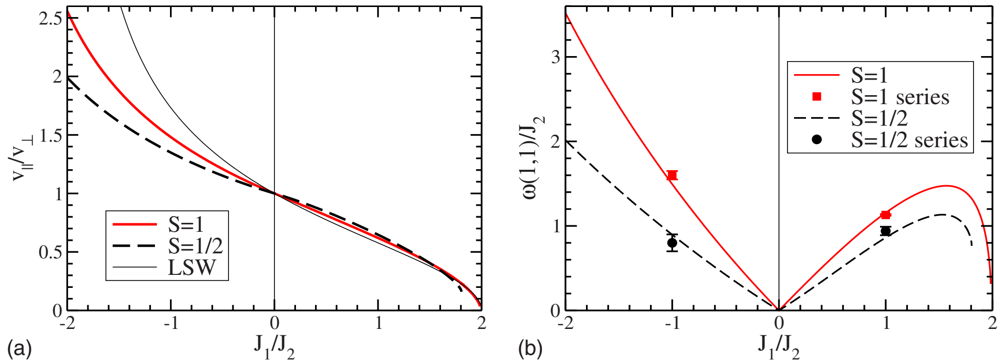

FIG. 2. (Color online) Panel (a) ratio of the spin-wave velocities parallel and perpendicular to the spin stripes, i.e., parallel is in b direction and perpendicular is in a direction; see Fig. 1(a). We stress that $v_\parallel$, though small, does not vanish where the magnetization vanishes. For comparison, LSW data are also included. Panel (b) depicts the spin-wave energy $\omega(\mathbf{q})$ at $\mathbf{q}=(1,1)$ and $\mathbf{q}=(0,1)$, respectively, in units of π/g (g lattice constant of the assumed square lattice). Note that $\omega(\mathbf{q})$, though small, does not vanish where the magnetization vanishes. The results are nicely corroborated by exemplary series-expansion data (symbols).

on a minimal spin model, namely, the J_1-J_2 Heisenberg model with spin $S=1$ ($S=1/2$ results are shown for comparison and to justify our approximations). Measurements of the spin-wave dispersion up to its maxima will further support the spin-Hamiltonian approach whose validity in turn shows that strong correlations dominate the pnictides. We show that a sizable finite energy of the spin waves is to be expected at $\mathbf{q}=(1,1)$ where we denote all wave vectors in units of π/g. This is in stark contrast to the results of linear spin-wave (LSW) theory where a vanishing spin-wave energy is predicted.[13,14] Second, we discuss the possibility that the magnetic fluctuations reduce the static local moment. Third, we make quantitative predictions of the dispersion and of the anisotropy in spin-wave velocities along ($v_\parallel$) and across ($v_\perp$) the magnetic stripes. Measurements of $v_\parallel/v_\perp$ can be used to determine the magnetic frustration J_2/J_1. The spin-wave spectra over the full Brillouin zone (BZ) show clear differences between a system deep in the columnar phase and one close to a quantum phase transition, where magnetic fluctuations dramatically reduce the static moment. This provides a robust experimental way to distinguish the two scenarios (i) and (ii).

The parent compound LaFeAsO is not a Mott-Hubbard insulator. It is rather a bad metal or semimetal without a Drude peak in the conductivity.[6,15] Even in the magnetically ordered phase the entire Fermi surface is not gapped.[6,9] Still, in the undoped system the magnetic excitations are long lived as they appear as sharp peaks[4] even at high energies.[5] So it is justified, though not undisputed,[16] to start with a model of well-localized spins coupled by Heisenberg interactions,

$$H = J_1 \sum_{\langle i,j\rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle\langle i,j\rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j, \tag{1}$$

where $\langle i,j\rangle$ stands for nearest neighbors (NN) and $\langle\langle i,j\rangle\rangle$ for next-nearest neighbors (NNN). The spin operators read $\mathbf{S}_i$ and represent $S=1$. This is not completely obvious in view of the complicated local electronic situations. But the success of two-band models strongly indicates that $S=1$ is the appropriate choice.[17,18] Furthermore, the band-structure results[7–9] with a local moment of up to $2.3\mu_B$ also indicate that there can be up to two electrons aligned. While our choice [Eq. (1)] neglects lifetime effects due to the decay into particle-hole pairs (Landau damping) we expect that the collective magnetic excitations and their dispersion are captured.

The choice of a J_1-J_2 Heisenberg model is justified because the superexchange is realized mostly via the As ion which sits in the middle of each Fe plaquette. Hence a NN contribution and a NNN contribution is to be expected.[10] Indeed, band-structure calculations show that the NN and the NNN couplings are sizable.[7,12,19] Both J_1 and J_2 turn out to be antiferromagnetic, i.e., positive and very similar in value. For this reason, we will choose for scenario (ii) the ratio $J_1/J_2=1$.

The technique employed is self-consistent spin-wave theory. It has been shown previously that this approximation works extremely well in the columnar, stripelike phase[20,21] for $S=1/2$; see also Figs. 1(b) and 2(b). Because spin-wave theory can be derived as a $1/S$ expansion, the results should only improve for $S=1$.

We have used the Dyson-Maleev as well as the Schwinger boson representation.[22] Both yield the same result on the level of self-consistent mean-field theory. In the symmetry broken phase the dispersion reads

$$\omega(\mathbf{q}) = CJ_2\sqrt{A^2 - B^2}, \tag{2a}$$

$$A = \lambda + x\beta\cos(\pi q_b), \tag{2b}$$

$$B = 2\cos(\pi q_a)\cos(\pi q_b) + x\alpha\cos(\pi q_a), \tag{2c}$$

with $x=J_1/J_2$. The expectation values C, α, and β are determined from the self-consistency conditions,

$$2S = \partial E_{\mathrm{MF}}/\partial(CJ_2\lambda) + 2m, \tag{3a}$$

$$2CJ_2 = -\partial E_{\mathrm{MF}}/\partial(CJ_2) + 4J_2 m, \tag{3b}$$

$$C\alpha J_1 = -\partial E_{\mathrm{MF}}/\partial(C\alpha) + 2J_1 m, \tag{3c}$$

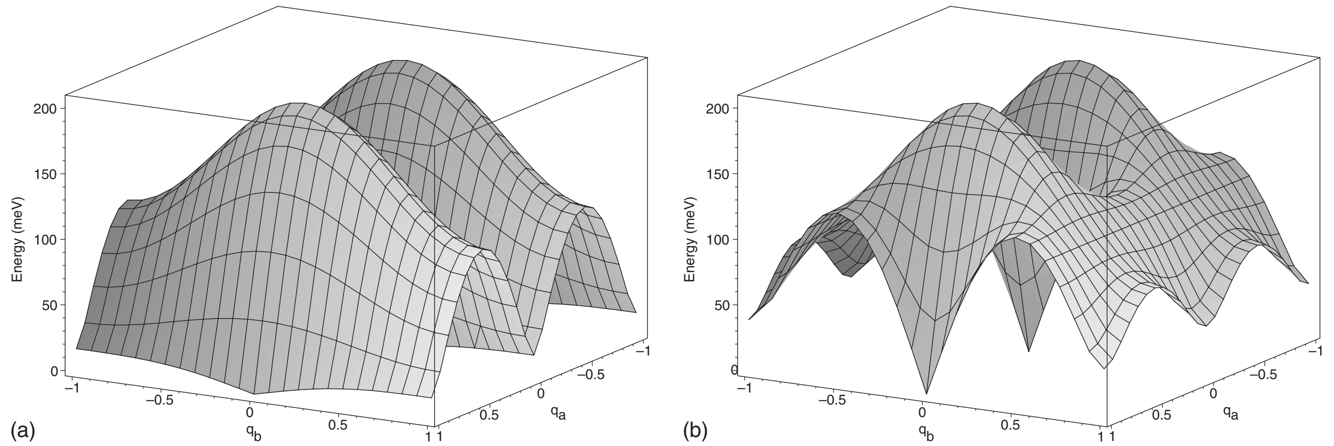

FIG. 3. (a) Dispersion of scenario (i) with $S=1$, $J_2=34$ meV, and $J_1/J_2=1.978$; (b) dispersion of scenario (ii) with $S=1$, $J_2=33$ meV and $J_1/J_2=1$. J_2 is chosen such that $v_\perp$ equals the experimental value (Ref. 4). Wave vectors in units of π/g.

$$C\beta J_1 = \partial E_{\mathrm{MF}}/\partial(C\beta) + 2J_1 m, \tag{3d}$$

where m is the staggered magnetization and the mean-field energy per spin

$$E_{\mathrm{MF}} = (2\pi)^{-2}\int_{\mathrm{BZ}}[\omega(\mathbf{q}) - ACJ_2]d^2q \tag{4}$$

is used. One integration can be done analytically, the other numerically. The self-consistency is solved by iteration. Gaplessness at zero wave vector implies $\lambda=2+x(\alpha-\beta)$ so that Eq. (3a) is used to determine m.

The resulting magnetizations are shown in Fig. 1(b). Note the extremely fast vanishing of the magnetization if x approaches 2. The vicinity of the classical first-order instability at $x=2$ (Ref. 23) makes $m(x)$ resemble a square root as $x \to x_c$ where m vanishes ($x_c|_{S=1/2}=1.8057$ and $x_c|_{S=1}=1.9836$) within the approximation. One may speculate that this is due to the Ising-type transition related to the breaking of directional symmetry,[13,24,25] but so far we cannot draw a definitive conclusion on this point. But in the light of the structural phase transition occurring before[2,3] or at[26,27] the magnetic phase transition this aspect is experimentally very interesting. The structural transition could easily be driven by the Ising transition in the directional degrees of freedom.

For completeness, we also include results for negative J_1. Around $x\approx-2$ another instability is expected.[28] But interestingly it does not lead to any precursors in the sublattice magnetization as found from self-consistent spin-wave theory and series expansion. We attribute this behavior to the fact that the classical instability to the ferromagnetic phase is completely first order in the sense that there are no precursive fluctuations because the ferromagnetic phase is free from quantum fluctuations.

The breaking of the directional symmetry implies that the spin-wave velocities depend on direction; see Fig. 2(a). This quantity is a much more robust probe for the value of the ratio $x=J_1/J_2$. The magnetization depends on matrix elements which in turn can depend on itinerancy, hybridization, and other effects. Energies in contrast only depend on the Hamiltonian and thus are much less ambiguous.

Furthermore, one notes that Eq. (2) implies that there is a *finite* excitation energy at $\mathbf{q}=(\pm1,\pm1)$ which is equivalent to $\mathbf{q}=(0,\pm1)$ if $x(\alpha-\beta)>0$. The results are plotted in Fig. 2(b). We stress that no dependence of the bare coupling J_1 on the bond direction is required.

Now we turn to the two scenarios presented. In scenario (i) we attribute the reduction of m to renormalization by the spin fluctuations. Equating the ratio 0.36/2.3 as from experiment[2] and band-structure theory[8,9] to m in Fig. 1(b) leads to $J_1=1.978J_2$. From Fig. 1(b) it is obvious that considerable fine tuning is needed. The resulting dispersion is shown in the upper panel in Fig. 3. The lower panel shows the dispersion for $J_1=J_2$ as suggested by band-structure calculations.[12,19] In both cases the overall scale of the coupling is adjusted to fit to the measured spin velocity $v_\perp$.[4] We stress that the value for J_2 of about 33 meV agree very well with the estimates from band-structure calculations.[12,19]

Comparing the two panels of Fig. 3, the difference in the dispersion of the spin waves in both scenarios is striking. Thus a measurement of the energetically higher lying modes will easily distinguish both scenarios. To facilitate the distinction we plot in Fig. 4 the dispersion along a generic path in the BZ. The perpendicular spin-wave velocity $v_\perp$ is fixed to its experimental value[4] by the appropriate choice of J_2. Obviously, the motion across the stripes (along $a^\star$) is the

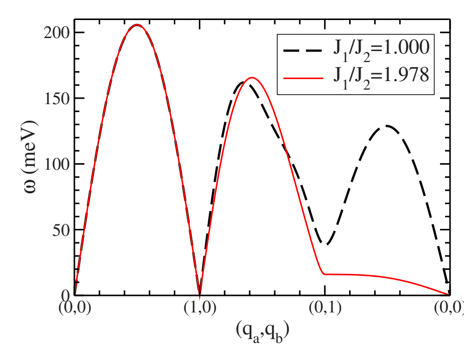

FIG. 4. (Color online) Dispersions of both scenarios for a generic path through the BZ. They coincide along $(0,0)\to(1,0)$, but differ strongly along $(0,1)\to(0,0)$. In LSW the dispersions at (0,1) would vanish spuriously. Wave vectors are given in units of π/g.

same for both sets of parameters. The important difference occurs in the motion along the stripes (along $b^\star$). If the spin fluctuations renormalize the magnetic moment the spin mode along $b^\star$ is extremely soft. Interestingly, this softness opens an additional channel for the magnons to decay at energies of about twice the energy at (0,1), i.e., above about 40–50 meV. So in this scenario significant line widths in inelastic neutron scattering are to be expected.

In conclusion, we presented a quantitative theory for the dispersion of the spin waves in the recently discovered superconducting pnictides. It is based on self-consistent spin-wave theory and series expansion for the $S=1$ J_1-J_2 Heisenberg model. Measurements at higher energies will further support a model of localized spins, for first evidence see Refs. 5 and 29. We predict a strong anisotropy of the spin-wave velocities and a finite excitation energy at the wave vectors (0,1) and (1,1).

Two scenarios for the strong reduction in the local magnetic moment are considered. The scenario (i) attributes the reduction to the magnetic fluctuations. We point out that the strongly varying static moments from $0.25\mu_B$ (Ref. 3) over $0.36\mu_B$ (Ref. 2) to $0.8\mu_B$ (Ref. 30) and $0.9\mu_B$ (Ref. 5) find a natural explanation if the pnictides are close to the quantum phase transition at $x\approx 2$ where the renormalized magnetization changes very rapidly on small parameter changes. This scenario implies the fascinating aspect that the pnictides realize a spin-isotropic system which displays an Ising transition in the orientation of its ferromagnetic stripes.[23–25]

The alternative scenario (ii) attributes the low magnetic moment to the local electronic configuration. Then $J_1/J_2 \approx 1$ is plausible which does not renormalize the magnetic values sizably (about 18%).

The measured anisotropies will allow one to decide how close the system is to a quantum phase transition with fascinating features such as directional Ising transitions. A quantitative understanding of spin fluctuations in the parent materials would help clarify one of the most intriguing issues in the field, namely, the similarities and differences between the pnictide and the cuprate family of materials at a fundamental level.

We thank Chris Hamer for very helpful discussions. The financial support (GSU) by the Heinrich-Hertz Stiftung NRW and the Gordon Godfrey Fund is gratefully acknowledged.

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