Biquadratic magnetic interaction in parent ferropnictides

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Abstract. We discuss an effective spin Hamiltonian with biquadratic interaction for ferropnictide superconductors from the point of view of band structure theory and available experimental data. This model is consistent with electronic structure calculations and captures many observed magnetic properties, including the anisotropy of the exchange coupling, thin domain walls, and the crossover from first to second-order phase transition under doping. The parameters of the model are analyzed as a function of the local spin moment using first-principles calculations. Calculations show the biquadratic coupling is negative in stoichiometric KFe$_2$Se$_2$, and the phase diagram is extended into this region. We also consider magnetic short-range order and discuss the limitations of this model in comparison with experiment.

1. Introduction

Strong evidence points toward spin fluctuations as the dominant pairing mechanisms in ferropnictide superconductors [1]. Therefore, intense attention has been focused on the magnetic properties of these materials, and in particular on understanding the character of spin fluctuations in both parent and doped compounds. An intermediate magnitude of the local moments ($\mu \sim 1\mu_B$) observed in neutron scattering experiments, as well as the fact that parent compounds are metallic, suggest that strong itinerant effects may be expected, but, on the other hand, that an effective model based on localized spin variables may provide a reasonable starting point. It has also been suggested that spin dynamics should be described based on significantly larger local moments, which are strongly reduced as a result of quantum spin fluctuations.

Application of the Heisenberg model to ferropnictides meets with problems. Although the collinear antiferromagnetic (AFM) ground state (“stripe phase”) may be obtained in the widely studied $J_1$-$J_2$ Heisenberg model with $J_2 > J_1/2$, the magnetic properties are poorly described by this model. First, it is well established both experimentally [2–5] and theoretically [6, 7] that the nearest-neighbor exchange constant (obtained from spin wave analysis or from linear response calculations) is strongly anisotropic in the stripe phase. This anisotropy strongly depends on the local moment and can even change sign at large values of $\mu$ [7, 8]. Second, the Heisenberg model of any range fails to describe the energies of noncollinear structures connecting the degenerate AFM domains. While in this model the $(\pi,0)$ and $(0,\pi)$ AFM domains are connected by a continuously degenerate set of noncollinear states, band structure calculations find a rather high
energy barrier between them [7, 9]. This is consistent with the experimental observation of microscopically thin twin domain walls between such stripe states [10]. Within the Heisenberg model such domain walls would be very thick due to the continuous degeneracy (in fact, infinitely thick in the classical description at zero temperature).

These essential features missing in the Heisenberg model can be reintroduced in different ways. It has been argued that anisotropic spin fluctuations are linked to orbital ordering [11–14]. Orbital degrees of freedom can be included in the model either as electronic degrees of freedom coupled to local spins [15, 16], or as Ising variables in a Kugel-Khomski-type model [13, 14]. However, a simpler description of magnetic properties may be achieved by adding non-Heisenberg terms in the effective spin-only Hamiltonian. Biquadratic coupling naturally appears when the total energy of an itinerant electron system is mapped to an effective spin Hamiltonian. This coupling comes from the complicated dependence of the electronic energy on the spin configuration, and a specific mechanism for ferropnictides was proposed [17]. The importance of biquadratic interaction in ferropnictides and related materials was discussed in Refs. [7, 9, 18], and the thermodynamics of this model was studied in Ref. [19]. The viewpoint based on the fully itinerant picture may be found in Refs. [17, 20].

In this paper we discuss the magnetic properties of ferropnictides within the effective spin model with biquadratic interaction [19]. After briefly reviewing the model in Section 2, we discuss the numerical values of the parameters based on first-principles calculations in Section 3. In this section we analyze the dependence of the interaction parameters on the local moment in CaFe$_2$As$_2$, as well as the frequency dependence of the biquadratic coupling. Using the stoichiometric KFe$_2$Se$_2$ as an example, we also show that the biquadratic coupling can change sign depending on the band filling, while remaining quite sizeable. In Section 4 we describe the phase diagram of the model, extending prior results [19] to the case of negative biquadratic coupling. Magnetic short-range order is discussed in Section 5, and the overall properties of the effective spin model are summarized in Section 6.

2. Model spin Hamiltonian

Consider the following effective classical spin Hamiltonian:

$$H = \sum_{i<j} \left[ J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - K_{ij} (\mathbf{S}_i \cdot \mathbf{S}_j)^2 \right]$$

(1)

The first term is the conventional (isotropic) exchange interaction, and the second term is the pairwise biquadratic interaction. Ferropnictides with the stripe ground state can be described by the $J_1$-$K$-$J_2$-$J_c$ model, including two in-plane exchange parameters $J_1$ and $J_2$, an interplane coupling $J_c$, and a nearest-neighbor biquadratic coupling $K = \tilde{K} S^2$. Based on the electronic structure calculations, it is known that in ferropnictides the biquadratic coupling of electronic origin is much larger than the one generated by the “order-from-disorder” mechanism [21–23] or by magnetostructural coupling [24].

The linear-response exchange parameters $J_{ij}^{LR}$, which determine the spin-wave spectra, are defined as second derivatives of the total energy with respect to spin rotations. From the Hamiltonian (1) we find

$$J_{ij}^{LR} = J_{ij} - 2\tilde{K}_{ij} S^2 \mathbf{e}_i \cdot \mathbf{e}_j,$$

(2)

where $\mathbf{e}_i = \mathbf{S}_i / S$. For the stripe ground state, we find $J_{1a} = J_1 + 2\tilde{K} S^2$ and $J_{1b} = J_1 - 2\tilde{K} S^2$. Thus, the anisotropy of the nearest-neighbor linear-response exchange parameter in the stripe phase is captured by the biquadratic term in the Hamiltonian (1). The biquadratic term also eliminates the continuous degeneracy for noncollinear states connecting the $(\pi,0)$ and $(0,\pi)$ stripe states [7, 9]. This barrier also explains why twin domain walls in ferropnictides are microscopically thin [10].
For example, the spin wave spectrum of CaFe$_2$As$_2$ was measured in Refs. [3, 4] and fitted to the anisotropic $J_1$-$J_2$-$J_c$ Heisenberg model. Mapping these anisotropic linear-response exchange parameters to model (1), we find $S J_1 = 22$, $S K = 14$, $S J_2 = 19$, and $S J_c = 5.3$ meV, where $K = \tilde{K} S^2$.

3. **First-principles analysis of interaction parameters**

Band structure theory can provide unique information about the spin Hamiltonian of the system. Apart from identifying the ground magnetic state, it is able to describe the adiabatic energy surface for a wide range of spin configurations with different magnitudes of the spin moments and angles between them. The energies of such configurations can be evaluated using constrained density functional theory [25] and then mapped onto a model spin Hamiltonian.

Band structure calculations based on density functional theory (DFT) correctly produce the stripe antiferromagnetic configuration as the ground state for ferropnictides [26]. As explained above, contrary to the Heisenberg model, in DFT the ground state does not have an internal degeneracy [7, 9]. Let us consider the total energy as a function of the rotation angle $\theta$ between the two interpenetrating Néel sublattices for stoichiometric CaFe$_2$As$_2$ and KFe$_2$Se$_2$ systems (Fig. 1). Here we use local density approximation (LDA) and the full-potential linear muffin-tin orbital (LMTO) method, and the distance $R_{Fe-As}$ between Fe and As (Se) atoms is varied to obtain different magnitudes of the spin moments. In CaFe$_2$As$_2$ the ground state is the collinear stripe AFM configuration (Fig. 2a). The total energy depends strongly on the angle $\theta$ and reaches a maximum of 20-40 meV at $\theta = 90^\circ$ (depending on the local spin moment). The simplest way to map this dependence to an effective spin Hamiltonian is to use Eq. 1 with a nearest-neighbor biquadratic coupling. For CaFe$_2$As$_2$, as well as for all compounds with the collinear stripe ground state, $K$ is positive.

![Figure 1](image-url)  

**Figure 1.** Calculations of the total energy in CaFe$_2$As$_2$ (bottom panel) and KFe$_2$Se$_2$ (top panel) as a function of the angle between the two interpenetrating Néel sublattices for different values of the Fe-As(Se) bond length. The curves are labeled by the value of the magnetic moment $\mu$ on Fe atoms in the stripe structure. The end points (0 and 180°) represent two degenerate stripe configurations.
Figure 2. Ground state spin orderings for the $J_1$-$K$-$J_2$-$J_c$ model. (a) Stripe ordering for $K > 0$. (b) Noncollinear ordering with a $90^\circ$ angle between two Néel sublattices for $K < 0$.

The situation in the case of KFe$_2$Se$_2$ is opposite: the total energy reaches its minimum at $\theta = 90^\circ$ (Fig. 2b), corresponding to negative $K$ in Eq. (1). This result shows that depending on the electronic structure (band filling) the biquadratic coupling can change sign, stabilizing either collinear or noncollinear magnetic orderings. Thus, in materials like stoichiometric KFe$_2$Se$_2$ the intrinsic non-Heisenberg interaction can lift the degeneracy in an opposite way compared to the order-from-disorder mechanism.

To further study the character of magnetic interactions in ferropnictides, we evaluate the effective exchange coupling for CaFe$_2$As$_2$ using the linear response technique. Table 1 and Fig. 3 show the effective exchange parameters for several nearest neighbors as a function of the local spin moment, which is manipulated by varying $R_{Fe-As}$. Table 1 also includes the values of $SK^*$ obtained from the energy barrier determined as shown in Fig. 1.

Large values of $SK$ at large spin moments $\mu$ show that in this regime the AFM stripe order is very stable, while the isotropic $J_1$-$J_2$ Heisenberg model is decidedly inappropriate for the description of the low-lying magnetic states. Comparing the values of $SK$ (obtained from the linear-response calculation for the nearest neighbors) with $SK^*$ (obtained from the energy barrier), we find that at small $\mu$ they differ by about a factor of 3. This means that non-Heisenberg coupling at small $\mu$ extends over at least a few coordination spheres. A significant anisotropy of the $J_3$ parameter (Table 1) confirms this interpretation. Note that the $J_3$ interaction is between the sites on the same Néel sublattice. Therefore, in order to include its anisotropy in the effective spin model, it would be necessary to add non-pairwise quartic terms in (1).

For larger $\mu$ the values of $SK$ and $SK^*$ come fairly close to each other, demonstrating a gradual transition from itinerant (long-ranged) to localized magnetic interaction. Note that the dependence of $K$ on $\mu$ (Fig. 3) is much stronger than that of $J_{ij}$, consistent with the fact that it may be interpreted as a higher-order term in the expansion of the total energy with respect to the non-magnetic state.

The anisotropy of $J_1$ was obtained by linear response technique [8] even before it was found experimentally. Once mapped to the $J_1$-$K$-$J_2$-$J_c$ model, the experimental anisotropy [4] appears to be larger compared to calculations by nearly a factor of 3 ($J_{1b}$ even changes sign in this model). Anisotropy of $J_1$ of this magnitude can only be obtained in band theory at very large values of $\mu$ [8]. This discrepancy may, at least partially, be due to the anisotropic contribution from non-nearest neighbors (as mentioned above for $J_3$). Another possibility is that it is necessary to consider dynamical magnetic interaction, which cannot be directly incorporated in a localized
Table 1. The exchange coupling parameters (in meV) and the Neel temperature (in K) in the stripe AFM phase of CaFe$_2$As$_2$ as a function of the magnetic moment $\mu$ (in $\mu_B$) of Fe. Indices $a$ and $b$ correspond to antiparallel and parallel spin pairs, respectively. $SK^*$ is obtained from the height of the energy barrier (Fig. 1). $T_N$ is estimated using Monte Carlo results from Section 4.

| $\mu$ | $SJ_1$ | $SK$ | $SJ_2$ | $SJ_c$ | $SJ_{3a}$ | $SJ_{3b}$ | $SK^*$ | $T_N$ |
|-------|--------|------|--------|--------|-----------|-----------|--------|-------|
| 0.76  | 37     | 2.1  | 24     | 10     | 0.64      | -8.6      | 6.6    | 99    |
| 1.07  | 50     | 4.6  | 40     | 9.4    | 9.7       | 1.1       | 11     | 182   |
| 1.39  | 49     | 8.8  | 44     | 8.6    | 14        | 5.3       | 13     | 245   |
| 1.57  | 43     | 11.2 | 42     | 7.9    | 16        | 6.6       | 14     | 253   |

Figure 3. Exchange and biquadratic coupling parameters as a function of the local moment.

Spin model. Consider the frequency-dependent exchange coupling defined as

$$J_{ij}(\omega) = \text{Re} \chi^{-1}_{ij}(\omega)$$

(3)

where $\chi^{-1}_{ij}(\omega)$ is the inverse Fourier transform of the dynamical transverse spin susceptibility. The dynamic biquadratic coupling can be obtained from the anisotropy of $J_1$ in the stripe AFM phase. To calculate the transverse dynamic spin susceptibility $\chi(q,\omega)$, we use an all-electron linear response technique developed in Refs. [27, 28]. In this method, the susceptibility $\tilde{\chi}(q,\omega)$ is first calculated in the full product basis set representation. Then it is projected onto the functions representing local spin densities on each magnetic site, which gives a matrix in basis site indices. This projection corresponds to the rigid spin approximation. Inversion of this matrix with a subsequent Fourier transform provides the real-space representation of the inverse susceptibility. Note that the local exchange enhancement does not affect the intersite exchange parameters.

The results for the anisotropy of the dynamic nearest-neighbor coupling in CaFe$_2$As$_2$ are shown in Fig. 4. (At $\omega = 0$ it corresponds to the static $SK$ in Table 1.) It is seen that $K(\omega)$ strongly depends on frequency, changing by a factor of 2 or 3 over the range of the spin-wave spectrum. This frequency dependence can not be incorporated in an effective spin Hamiltonian and complicates the interpretation of the measured spin excitation spectra. The relative importance of the longer-range interactions contributing to $SK^*$ and of the dynamical effects is currently unclear, but it is quite likely that the high-frequency part of the spectrum is more appropriately described in the itinerant picture. In the following we assume that the
thermodynamic properties are not sensitive to these high-frequency excitations and may be described by a spin model (1) with effective interaction parameters that may already include some reasonable averaging over the thermal frequency range.

Figure 4. Dynamical biquadratic exchange coupling in the spin wave region for CaFe$_2$As$_2$ in the antiferromagnetic (stripe) magnetic state, for two values of the local moment obtained by modifying the Fe-As bond length.

4. Phase diagram
The spin ground state of the $J_1$-$K$-$J_2$-$J_c$ model depends on the sign of $K$. At $K = 0$ the ground state has an internal O(3) degeneracy in addition to the global spin rotation. It is convenient to divide the original square lattice into two equivalent sublattices I and II, which are connected by next-nearest-neighbor bonds. The antiferromagnetic $J_2$ coupling enforces the Neél order within each sublattice, but the classical energy does not depend on the relative angle between them. The biquadratic interaction lifts this degeneracy. At $K > 0$ the collinear stripe ordering is stabilized (Fig. 2a), while at $K < 0$ the energy is minimized in a noncollinear (2Q) configuration with nearest-neighbor spins aligned perpendicular to each other (Fig. 2b). While the internal symmetry of the magnetic phase becomes discrete ($Z_2$) at $K > 0$, it is only lowered from O(3) to O(2) at $K < 0$. Here we consider the magnetic phase diagram of this model at both positive and negative $K$.

4.1. Mean-field approximation
At $K > 0$ the Hamiltonian takes the following form in the mean-field approximation (MFA):

$$H_{\text{MFA}}^{K>0} = -mJ_s \sum_i \eta_i S_{iz} - qK_0 \sum_i S_{iz}^2$$  \hspace{1cm} (4)

where $J_s = 4J_2 + 2J_c$, $K_0 = 4K$ and $\eta_i = \pm 1$ for $i \in I$ and $i \in II$. The MFA equations contain dipole and quadrupole effective fields [29], demanding self-consistency for both the staggered magnetization $m = \eta_i \langle S_i^z \rangle$ and the quadrupole order parameter $q = (3\langle S_i^2 \rangle - 1)/2$. The lowest free energy may be obtained in the paramagnetic phase ($m = q = 0$), the antiferromagnetic phase ($m \neq 0$, $q \neq 0$), or in the quadrupolar phase($m = 0$, $q \neq 0$).

For $K < 0$ we assume that the magnetization on the sublattice I (II) lies along the $x$ ($y$) axis. The MFA Hamiltonian can be then written as

$$H_{\text{MFA}}^{K<0} = -mJ_s \sum_i \eta_i S_{i\parallel} - K_0 \sum_i \left( q_{\parallel} S_{i\parallel}^2 + q_{\perp} S_{i\perp}^2 \right)$$  \hspace{1cm} (5)
where \( S_{\parallel} (S_{\perp}) \) is the component of \( S \) parallel (perpendicular) to the magnetization of site \( i \), while \( q_{\parallel} = \langle S_{iy}^2 - S_{iz}^2 \rangle = \langle S_{ix}^2 - S_{iz}^2 \rangle \) and \( q_{\perp}^I = \langle S_{ix}^2 - S_{iy}^2 \rangle = \langle S_{iy}^2 - S_{iz}^2 \rangle \) are the two parameters characterizing the quadrupole ordering tensor. Minimization of the free energy with respect to \( m, q_{\perp} \) while \( q_{\parallel} = 0 \) leads to a set of three self-consistent equations. Similarly to the case of positive \( K \), there are three possible types of solutions: paramagnetic \( (m = q_{\parallel} = q_{\perp} = 0) \), antiferromagnetic \( (m \neq 0, q_{\parallel} \neq 0, q_{\perp} \neq 0) \), and quadrupolar \( (m = 0, q_{\parallel} \neq 0, q_{\perp} \neq 0) \).

![Figure 5](image_url)

**Figure 5.** MFA phase diagram for the \( J_1-K-J_2-J_c \) model. Temperature is measured in units of \( T_N^{MFA} = J_s / 3 \), which is the second-order \( T_N \) in MFA. AFM\(_I\) and AFM\(_{II}\) denote stripe and noncollinear (\( \theta = 90^\circ \)) orderings, respectively, while Q\(_I\) and Q\(_{II}\) denote the corresponding quadrupolar phases. Solid and dashed lines denote second and first-order transitions, respectively. Points A, A', C and C' are tricritical points, point B is a triple point, and B' is a critical endpoint.

The MFA phase diagram is shown in Fig. 5. For small positive \( K \) there is a second order transition from the AFM stripe phase to the paramagnetic state with \( T_N^{MFA} = J_s / 3 \) being independent of \( K \). At the tricritical point A \( (K_A = 5J_s/24) \) the transition changes to first order, and at \( K > K_A \) the transition temperature grows approximately linearly in \( K \). Note that first-order transitions are common in systems with biquadratic coupling \([29, 30]\). The tricritical point A is within the range of realistic parameters for ferropnictides.

The quadrupolar phase appears at a much larger value \( K \approx J_s \) (point B). This phase is separated from the paramagnetic phase by a first-order transition, but the transition from the AFM to quadrupolar phase is first order at \( K < K_C \) and second-order at \( K > K_C \).

For \( K < 0 \) the phase diagram is quite similar. The noncollinear AFM phase (labeled AFM\(_{II}\) in Fig. 5) disorders through a second-order transition up to the tricritical point A' at \( K_A' \approx -0.27J_s \). At \( |K| > |K_A'| \) the transition temperature is constant and equal to \( J_s / 3 \), while the first-order transition temperature at larger \( |K| \) grows approximately linearly in \( K \). The quadrupolar phase appears at a critical endpoint at \( K \approx -0.9J_s \). The transition from the noncollinear phase to the quadrupolar phase changes from first to second order at a slightly higher \( |K| \) (point C'). The quadrupolar phase disorders through a second-order transition, contrary to the quadrupolar phase appearing at large \( K > 0 \). Both quadrupolar phases appear at very large values of \( |K| \) and are unlikely to be realized in ferropnictides and related materials.
4.2. Monte Carlo simulations

The phase diagram at $K > 0$ was previously evaluated in Ref. [19]; here we extend it to the region of $K < 0$. This extension allows one to better understand the effects of biquadratic coupling, and, as shown in Section 3, negative $K$ may be realized in appropriately doped stoichiometric iron chalcogenides. We used cubic lattices $D \times D \times D$ with periodic boundary conditions and $D = 14, 16, 18$, and 20. Monte Carlo simulations were performed using the Metropolis algorithm, each step consisting in a trial random change of the spin direction for one site. The lengths of the equilibration and averaging runs were adjusted to achieve sufficient accuracy. We restrict ourselves to the realistically small range of $|K|/J_s < 0.4$, where the quadrupolar phase does not appear.

For the noncollinear ordering we used two alternative definitions of the order parameter: $\sqrt{(L_1^2 + L_2^2)/2}$ and $|L_1 \times L_2|$, where $L_J$ is the Néel order parameter for sublattice $J$. The results for both choices of the order parameters agree within the computational error bar.

The order of the phase transition was determined by analyzing the behavior of the fourth-order energy cumulant [31]. If this cumulant converges to 2/3 for all temperatures with increasing $D$, the transition is second-order. If the energy cumulant develops a minimum at some temperature, which sharpens with increasing $D$, the transition is of first order [31]. Second-order transition temperatures were determined using finite-size scaling of the Binder cumulant [32]. First-order transition temperatures were found from the peaks of the appropriate susceptibilities.

The magnetic phase diagram is shown in Fig. 6. To illustrate the dependence of the transition temperature on the model parameters, we include the dependence of the Néel temperature on $K$ for a few values of the $J_2/J_1$ and $J_c/J_1$ ratios. At $K > 0$ and $T = 0$ the stripe phase is stable for $J_2/J_1 > 0.5$. At $K < 0$ the noncollinear phase is stable with respect to the simple Néel order at $J_2/J_1 > 0.5(1 - |K|)$. Close to these points, as well as at smaller $J_c/J_1$, the transition temperature is significantly reduced compared to MFA due to strong fluctuations.

Using the parameters taken from the fitting of experimental spin wave dispersions for CaFe$_2$As$_2$ and BaFe$_2$As$_2$, the Néel temperatures are estimated at 90 K and 65 K, respectively (in both cases the transition is first-order in agreement with experiment [33]). These values are notably smaller compared to the experimental $T_N$ of 170 K and 140 K. We have also estimated $T_N$ using the calculated exchange parameters for CaFe$_2$As$_2$ (see the last column of Table 1). In this estimate, we first calculated $J_s$ taking $J_2$, symmetric $J_3$, and $J_c$ into account, found the mean-field estimate $T_{N\text{MFA}}$, and then appropriately reduced it using Monte Carlo results (Fig. 6) for the corresponding values of the parameter ratios. (Specifically, the mean-field $T_N$ were multiplied by 0.51, 0.56, 0.59, and 0.6 for the four lines in Table 1.) We see that the experimental $T_N$ for CaFe$_2$As$_2$ is reproduced for $\mu \sim 1$, in reasonable agreement with the measured values of the local spin moment. We also see that the contribution of $J_3$ is not negligible: for example, at $\mu = 1.39\mu_B$ it reduces the $T_N$ estimate by 20%. More distant intrasublattice couplings (not listed) are at least a few times smaller than $J_3$.

At $K > 0$ the phase transition changes from second to first order at increasing $K$. This is similar to MFA results, but the tricritical value of $K$ is significantly reduced by fluctuations. On the other hand, for $K < 0$ the transition remains second-order even beyond the MFA tricritical point, although the incipient tendency toward first-order transition increases with the magnitude of $K$. It is possible that the tricritical point at $K > 0$ is driven down by interaction with the nematic order parameter [34].

5. Magnetic short-range order

Spin excitations in the paramagnetic phase AFe$_2$As$_2$ materials (A=Ca,Sr,Ba) were investigated using inelastic neutron scattering [35–37]. In general, these studies show the existence of strong short-ranged correlations peaked at the wave vector $Q_{AFM}$ of the low-temperature AFM structure. These correlations exhibit in-plane anisotropy [36, 37], whose magnitude is similar to
Figure 6. Magnetic phase diagram of the $J_1$-$K$-$J_2$-$J_c$ model obtained by MC simulations. The plot shows the transition temperatures for stripe-to-paramagnetic (positive $K$) and noncollinear-to-paramagnetic (negative $K$) phase transitions as well as the order of these transition as a function of biquadratic interaction $K$. Temperature is measured in units of $T_N^{\text{MFA}} = J_s/3$ which is the second-order $T_N$ in MFA. Each line type is labeled by a set of two parameters ($J_2/J_1$; $J_c/J_1$). The region of first-order transitions (empty symbols) is schematically highlighted by shading and thicker lines. The dashed line shows the point of inversion of $J_1 b$.

the anisotropy of spin waves at low temperatures. The interpretation of this anisotropy is an unresolved question.

The excitation spectrum of the two-dimensional $J_1$-$J_2$-$K$ model deep below the nematic phase transition has properties consistent with experimental measurements for the paramagnetic phase [38]. Although evidence of electronic nematicity in a wide range of temperatures above the Néel point has been reported in ferropnictides [39], it is currently unclear whether the underlying nature of the corresponding phase is captured by a strongly two-dimensional spin model with biquadratic interaction.

Here we show that in the paramagnetic phase of model (1) with a moderate interlayer coupling (which does not have a broad nematic phase) the anisotropy of the excitation spectrum is insensitive to biquadratic coupling $K$; this implies that in this model the anisotropy of the excitation spectrum increases as the temperature is raised into the paramagnetic phase.

Consider the static structure factor $C(q) = \langle |S(q)|^2 \rangle$, where $S(q)$ is the Fourier transform of $S_i$. This quantity is equal to the dynamical structure factor integrated over frequency, and its anisotropy is therefore representative of the anisotropy of the dynamical excitation spectrum. We have calculated $C(q ||)$ (at $q_z = 0$) in Monte Carlo simulations by collecting a number of spin configurations (each taken after an equilibration run), evaluating $|S(q_x, q_y, q_z)|^2$ using the fast Fourier transform for each atomic plane $z_k$, and averaging over the atomic planes and the spin configurations. For comparison, we also evaluated $C(q)$ using the Onsager reaction field approximation, in which the fluctuation-dissipation theorem is enforced by construction [40]. Similar to the random phase approximation (RPA), this method gives the transition temperature

$$
\frac{T_N^{\text{RPA}}}{T_N^{\text{MFA}}} = \left( \sum_q \frac{1}{1 - J_q} \right)^{-1}
$$
where \( J_\mathbf{q} \) is the Fourier transform of the normalized exchange parameters \( J_{ij}/J_s \). Note that the biquadratic term has no effect on Eq. (6), because it does not contribute to the paramagnetic susceptibility; the phase transition is always second-order in this approximation. The structure factor above \( T_N^{\text{RPA}} \) is given by

\[
C_{\text{RPA}}(\mathbf{q}) = \frac{S^2}{1 - (J_\mathbf{q} - \lambda)T_N^{\text{MFA}}/T}
\]

where the parameter \( \lambda \) satisfies the equation

\[
\lambda = \sum_\mathbf{q} \frac{J_\mathbf{q}}{1 - (J_\mathbf{q} - \lambda)T_N^{\text{MFA}}/T}
\]

The results for \( C(\mathbf{q}) \) at \( J_2/J_1 = 0.8, J_c/J_1 = 0.2 \), and \( T = 1.1T_N \) are shown in Fig. 7. First, we see that Monte Carlo results at \( K = 0 \) (panel (b)) agree very well with the RPA formula (panel (a)). As expected, the structure factor has elliptical peaks centered around the AFM wavevectors \( \{1,0\} \), with the major axis directed toward the corners of the Brillouin zone. This feature agrees with experimental excitation spectra. The peaks have elliptical shape reminiscent of the anisotropic spin wave excitations at low temperatures, even though the system is in the paramagnetic state, and the structure factor retains the full four-fold symmetry of the square lattice. (In Ref. [38] this symmetry is obtained in the nematic phase only after symmetrization, assuming the existence of macroscopic AFM domains.) The ellipticity of the peaks is a generic property of the Heisenberg model [35]. Further, Fig. 7c shows the structure factor for \( K/J_1 = 0.4 \)

\[\text{(a)}\quad \text{(b)}\quad \text{(c)}\]

**Figure 7.** Static structure factor \( C(q_x, q_y, 0) \) within the first Brillouin zone at \( T = 1.1T_N \). (a) \( C_{\text{RPA}}(q_x, q_y, 0) \) for \( J_2/J_1 = 0.8, J_c/J_1 = 0.2 \) and \( K = 0 \). (b) Monte Carlo results for the same parameters. (c) Monte Carlo results for \( J_2/J_1 = 0.8, J_c/J_1 = 0.2 \) and \( K/J_1 = 0.4 \). The X point is placed in the center of the frame; the \( \Gamma \) point is in the middle of the vertical edges. Normalization corresponds to \( S = 1 \). The functions in all panels have full four-fold symmetry.

(Other parameters are the same as in panel (b)). We see that the biquadratic coupling of this sizeable magnitude does not affect the eccentricity of the elliptical zone-boundary peaks.

Using the RPA structure factor (7), it is easy to see that in the Heisenberg model (at \( K = 0 \)) the anisotropy of the paramagnetic excitation spectrum is identical to the anisotropy of the spin wave velocity at \( T = 0 \). Positive \( K \) decreases the anisotropy of the spin-wave velocity at \( T = 0 \) [19]. Therefore, the fact that \( K \) has little effect on the anisotropy of the paramagnetic spectrum implies that in model (1) with large \( K \) the anisotropy of the excitation spectrum should notably increase as the temperature is raised into the paramagnetic phase.

Note that the anisotropy of the spin excitation spectrum in 122 compounds increases with Co doping [41, 42]. According to first-principles calculations (Table 1), the \( J_2/J_1 \) ratio decreases with decreasing \( \mu \). As long as the effect of doping is mainly to decrease \( \mu \), these two trends are consistent with each other.
6. Discussion and conclusions

The addition of biquadratic terms to the Heisenberg model allows one to reconcile it with numerous experimental observations, including the anisotropy of the spin wave spectrum, microscopic thickness of the twin domain walls between different variants of the stripe phase, as well as the existence of a tricritical point as a function of doping [33, 43–45]. (Doping is expected to reduce the spin moments, and thereby the importance of biquadratic coupling, see Section 3.) Biquadratic terms eliminate the continuous degeneracy of the ground state, in agreement with electronic structure calculations. With the inclusion of the third-neighbor exchange coupling $J_3$, this model was also used to describe the magnetic interaction in other families of iron-based superconductors [46].

The localized spin model should be viewed as an effective mapping of complicated itinerant interactions. The significant frequency dependence of the dynamic analog of the biquadratic term (Section 3) indicates a limitation of the model, which may also be related to the high-frequency behavior of the spin fluctuation spectrum. As we have seen in Section 5, in a moderately three-dimensional effective spin model with strong biquadratic interaction the anisotropy of the spin fluctuation spectrum should increase as the system is heated above the Néel temperature. On the other hand, experiments show that this anisotropy is essentially independent of temperature [35–37]. One possible interpretation is that the effective biquadratic interaction is not very strong in ferropnictides. It was argued [42] that the anisotropy of $J_1$ extracted from the spin-wave spectrum [4] reflects the high-energy part of the spectrum dominated by itinerant effects, while the low-energy part can be described by an isotropic $J_1$-$J_2$ model [42]. This interpretation is consistent with first-principles calculations of the frequency-dependent exchange parameters (Section 3). On the other hand, pure Heisenberg model has a continuous ground-state degeneracy, which is inconsistent with first-principles calculations and with the observation of microscopically thin domain walls. Another possible interpretation is the existence of a strong magnetic short-range order above the Néel temperature [47]. Although strong short-range order is unattainable in models with moderate interplane coupling (as assumed in Section 5), it can be reached in the symmetry-broken nematic phase in the strongly two-dimensional limit [38]. The situation remains unresolved, particularly in view of recent experiments suggesting broken symmetry in a broad temperature range above $T_N$ [39].

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