Magnetic phase diagram of a spatially anisotropic, frustrated spin-$\frac{1}{2}$ Heisenberg antiferromagnet on a stacked square lattice

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Received 15 November 2010
Published 7 January 2011
Online at stacks.iop.org/JPhysCM/23/046001

Abstract

The magnetic phase diagram of a spatially anisotropic, frustrated spin-$\frac{1}{2}$ Heisenberg antiferromagnet on a stacked square lattice is investigated using a second-order spin-wave expansion. The effects of interlayer coupling and the spatial anisotropy on the magnetic ordering of two ordered ground states are explicitly studied. It is shown that with increase in next nearest neighbor frustration the second-order corrections play a significant role in stabilizing the magnetization. We obtain two ordered magnetic phases (Néel and stripe) separated by a paramagnetic disordered phase. Within the second-order spin-wave expansion we find that the width of the disordered phase diminishes with increase in the interlayer coupling or with decrease in spatial anisotropy but it does not disappear. Our obtained phase diagram differs significantly from the phase diagram obtained using linear spin-wave theory.

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

1. Introduction

The availability of new magnetic materials and the recent discovery of superconductivity at relatively high temperatures in the iron pnictide family of materials have spurred a flurry of interest in understanding the properties of frustrated magnets [1–14]. For the last two decades the properties of quantum spin-$\frac{1}{2}$ Heisenberg antiferromagnets (HAFMs) with nearest neighbor (NN) $J_1$ and next nearest neighbor (NNN) exchange interactions $J_2$ on a square lattice have been studied extensively by various analytical and numerical techniques [15–41]. It is now well established that at low temperatures these systems exhibit new types of magnetic order and novel quantum phases [42, 43]. For $J_2 = 0$ the ground state is antiferromagnetically ordered at low temperatures. Addition of NNN interactions induces a strong frustration and breaks the antiferromagnetic (AF) order. The competition between NN and NNN interactions for the square lattice is characterized by the frustration parameter $\eta = J_2/J_1$. A disordered paramagnetic phase, probably a columnar dimer, exists between $\eta_{1c} \approx 0.38$ and $\eta_{2c} \approx 0.60$. For $\eta < \eta_{1c}$ the square lattice is AF ordered whereas for $\eta > \eta_{2c}$ a degenerate collinear antiferromagnetic (CAF) stripe phase emerges. Experimentally, by applying high pressure, the ground state phase diagram of these frustrated spin systems can be explored from low $\eta = J_2/J_1$ to high $\eta$. For example, in Li$_2$VOSiO$_4$ x-ray diffraction measurements show that the value of $\eta$ decreases by about 40% with increase in pressure from zero to 7.6 GPa [44]. Moreover, nuclear magnetic resonance, magnetization, specific heat, and muon spin rotation measurements on the compounds Li$_2$VOSiO$_4$, Li$_2$VOGeO$_4$, VOMoO$_4$, and BaCdVO(PO$_4$)$_2$ show significant coupling between NN and NNN neighbors [6–8]. In addition, these experiments on Li$_2$VOSiO$_4$ have shown that it undergoes a phase transition at a low temperature (2.8 K) to collinear AF order with magnetic moments lying in the $a$–$b$ plane with $J_2 + J_1 \sim 8.2(1)$ K and $J_2/J_1 \sim 1.1(1)$ [8, 9].

A generalization of the frustrated $J_1$–$J_2$ model is the $J_1$–$J'_{1}$$-J_2$ model where $\xi = J'/J_1$ is the directional anisotropy parameter [23, 26, 27]. A possible candidate for this model may be the compound (NO)Cu(NO$_3$)$_3$ [45]. Extensive band structure calculations [36] for the vanadium phosphate...
with a staggered magnetic moment of waves [67]. These studies have also revealed that the parent neutron scattering spectra for the pnictides show sharp spin-

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However, spin-wave calculations for the 3D $J_1-J_2$ model on the body centered cubic (BCC) lattice and for the simple cubic (SC) lattice show no signs of an intermediate disordered paramagnetic phase [46, 47, 61]. Instead, for the BCC and the SC lattice, a direct first-order phase transition occurs from a two sublattice Néel ordered AF phase for small $J_2$ to a collinear antiferromagnetic ordered state for large $J_2$ [46, 47]. For Li$_3$VOSiO$_4$, a layered material that can be described by a square lattice $J_1-J_2$ model with large $J_2$, the interlayer coupling $J_{\perp}/J_1 \approx 0.07$ is not negligible [10]. Due to a finite interlayer magnetic coupling $J_{\perp}$ these experimental systems are quasi-2D.

Another example is the recently discovered iron pnictide superconductors [11]. The parent phases of these materials have been found to be metallic, but with AF order and magnetic excitations, and have been shown to play an important role in the superconducting state [11–14, 62–66]. Although magnetism in these materials is still debated, neutron scattering spectra for the pnictides show sharp spin-waves [67]. These studies have also revealed that the parent compounds exhibit a columnar antiferromagnetic ordering with a staggered magnetic moment of $(0.3–0.4) \mu_B$ in LaOFeAs and $(0.8–1.01) \mu_B$ in Sr(Ba,Ca)Fe$_2$As$_2$ [14, 67]. Moreover, at low temperatures there is orthorhombic distortion and the exchange constants have been found to be substantially anisotropic [68, 69]. Motivated by the observation of spatially anisotropic exchange constants in these materials the spin-wave spectra and the low-temperature phases of the model can be studied by the spatially anisotropic $J_1-J_1'-J_2$ Heisenberg antiferromagnet on a stacked square lattice. We use a spin-wave expansion based on Holstein–Primakoff transformation up to second-order in $1/S$ to numerically calculate the sublattice magnetization for each of the two ordered magnetic phases. The paper is organized as follows. Section 2 provides an introduction to the Hamiltonian for the Heisenberg spin-$\frac{1}{2}$ AF on a spatially anisotropic stacked square lattice. The classical ground state configurations of the model and the two phases are then briefly discussed. In sections 2.1 and 2.2 the spin Hamiltonian is mapped to the Hamiltonian of interacting spin-wave excitations, and the spin-wave expansion for sublattice magnetizations is presented for the two phases. Magnetizations for the two phases are numerically calculated with different values of interlayer coupling and spatial anisotropy and the results are plotted and discussed in section 3. Finally we summarize our results in section 4.

2. Theory

We consider a spatially anisotropic, frustrated spin-$\frac{1}{2}$ HAFM on a $N_L \times N_L \times N_L$ cubic lattice with four types of exchange interactions between spins: $J_1$ along the $x$ (row) direction, $J_1'$ along the $y$ (column) direction, $J_2$ along the diagonals in the $xy$ plane, and $J_\perp$ is the interlayer coupling. We assume all interactions to be AF and positive i.e. $J_1, J_1', J_2, J_\perp > 0$. This $J_1-J_1'-J_2-J_\perp$ spin system is described by the Heisenberg Hamiltonian

\begin{equation}
H = \frac{1}{2} \sum_{i,\ell} \left[ J_1 |S_{i,\ell} \cdot S_{i+\delta_x,\ell}| + J_1' \sum_{\ell} |S_{i,\ell} \cdot S_{i+\delta_y,\ell}| \right] + J_2 |S_{i,\ell} \cdot S_{i+\delta_x+\delta_y,\ell}| + \frac{1}{2} J_\perp \sum_{i,\ell} |S_{i,\ell} \cdot S_{i,\ell+1}|.
\end{equation}
where $\ell$ labels the layers, $i$ runs over all lattice sites, and $i + \delta_x$ ($\delta_x = \pm 1$) and $i + \delta_y$ ($\delta_y = \pm 1$) are the NNs to the $i$th site along the row and the column direction. The third term represents the interaction between the NNs, which are along the diagonals in the $xy$ plane and the last term is for the NN coupling between the layers. This model is different from the fully frustrated simple cubic lattice which has the additional NNN interactions in the $xz$ and $yz$ planes [46].

At zero temperature this model exhibits three types of classical ground state (GS) configurations: the Néel or the $(\pi, \pi, \pi)$ state, the two stripe states which are the columnar stripe $(\pi, 0, \pi)$ and the row stripe $(0, \pi, \pi)$. The classical ground state energies of these states are

$$
E_{\text{AF}}^{\text{class}} / N = -\frac{1}{2} J_1 S^2 \zeta [1 + \xi - 2\eta - \delta],
$$
$$
E_{\text{col}}^{\text{class}} / N = -\frac{1}{2} J_1 S^2 \zeta [1 - \xi + 2\eta + \delta],
$$
$$
E_{\text{row}}^{\text{class}} / N = -\frac{1}{2} J_1 S^2 \zeta [-1 + \xi + 2\eta + \delta],
$$

where $\zeta = J_2 / J_1$ measures the directional anisotropy, $\eta = J_3 / J_1$ is the magnetic frustration between the NN (row direction) and NNN spins, and $\delta = J_4 / J_1$ is the interlayer coupling parameter. $\zeta = 2$ is the number of NN sites along the row (column) direction. For $\eta < \zeta / 2$ the classical GS is the AF Néel state and for $\eta > 1 / 2$ (with $\zeta = 1$) the GS is doubly degenerate. Otherwise for $\eta < 1$ the GS is the columnar AF ($\pi, 0, \pi$) state. The classical first-order phase transition between the AF and CAF state occurs at the critical value $\eta_{\text{class}}^c = \zeta / 2$, which is independent of $\delta$.

The motivation of this paper is to investigate the role of interlayer coupling $\delta$ and spatial anisotropy $\xi$ to the quantum phases of this model. How do the quantum fluctuations due to $\delta$ and $\xi$ affect the disordered paramagnetic phase? Is there a critical point for $\delta$ (or $\xi$) above (or below) which the intermediate quantum paramagnetic GS does not exist and we have a direct first-order phase transition from the AF to the CAF ordered phase? For our study we follow the standard procedure by first expressing the fluctuations around the ‘classical’ ground state in terms of the boson operators using the Holstein–Primakoff transformation. The quadratic term in the boson operators corresponds to the LSWT, whereas the higher-order terms represent spin-wave (magnon) interactions. We keep terms up to second-order in $1/S$. The staggered magnetizations per spin to the leading order in $1/S^2$ for the AF and CAF phases are then obtained from the renormalized magnon Green’s functions and self-energies. We will follow the theoretical framework described in detail in [23]. However, for completeness we provide the equations that are required for numerical computations and that are needed to follow the present work.

### 2.1. $(\pi, \pi, \pi)$ AF Néel phase

For the AF ordered phase, NN interactions are between A and B sublattices and NNN interactions are between A–A and B–B sublattices. The Hamiltonian in equation (1) takes the form

$$
H = J_1 \sum_{i, \ell} S_{i, \ell}^A \cdot S_{i, \ell + \delta_x, \ell}^B + J_1' \sum_{i, \ell} S_{i, \ell}^A \cdot S_{i, \ell + \delta_y, \ell}^B + \frac{1}{2} J_2 \sum_{i, \ell} S_{i, \ell}^A \cdot S_{i, \ell + \delta_x, \ell + \delta_y, \ell}^B + S_{i, \ell}^B \cdot S_{i, \ell + \delta_y, \ell + \delta_x, \ell}^B + J_\perp \sum_{i, \ell} S_{i, \ell}^A \cdot S_{i, \ell + \delta_y, \ell + \delta_x}. \tag{3}
$$

This Hamiltonian is mapped into an equivalent Hamiltonian of interacting bosons by transforming the spin operators to bosonic creation and annihilation operators $a_i^\dagger$, $a_i$ for ‘up’ and $b_i^\dagger$, $b_i$ for ‘down’ sublattices using the Holstein–Primakoff transformations, keeping only terms up to the order of $1/S^2$.

$$
S_{i, \ell}^A \approx \sqrt{2 S} \left[ \frac{1}{2} a_i^\dagger a_i \frac{1}{2} a_i^\dagger a_i \right] a_i^\dagger, \tag{4}
$$
$$
S_{i, \ell}^B \approx \sqrt{2 S} b_i^\dagger b_i \left[ \frac{1}{2} b_i^\dagger b_i \frac{1}{2} b_i^\dagger b_i \right] \tag{5}
$$

In powers of $1/S$ the Hamiltonian is now written as

$$
H = -\frac{1}{2} NJ_1 S^2 \zeta (1 + \xi) \left[ 1 - \frac{2\eta}{1 + \xi} \right] + H_0 + H_1 + H_2 + \cdots \tag{6}
$$

The first term corresponds to the classical energy of the AF ground state (equation (2)). Then the real space Hamiltonian is transformed to the $k$-space Hamiltonian. Momentum $k$ is defined in the first Brillouin zone (BZ): $-\pi < k_x \leq \pi$, $-\pi < k_y \leq \pi$, $-\pi < k_z \leq \pi$ (with unit lattice spacings). Next we diagonalize the quadratic part $H_0$ by transforming the operators $a_k$ and $b_k$ to magnon operators $\alpha_k$ and $\beta_k$ using the Bogoliubov (BG) transformations

$$
\alpha_k^\dagger = l_k \alpha_{k_0}^\dagger + m_k \beta_{k_0}, \quad \beta_{-k} = m_k \alpha_{k_0}^\dagger + l_k \beta_{k_0}, \tag{7}
$$

where the coefficients $l_k$ and $m_k$ are defined as

$$
l_k = \frac{\left[ 1 + \epsilon_k \right]^{1/2}}{2 \epsilon_k}, \tag{8}
$$
$$
m_k = -\text{sgn}(\gamma_k) \frac{\left[ 1 - \epsilon_k \right]^{1/2}}{2 \epsilon_k} = -\chi_k \frac{1}{l_k},\tag{9}
$$

with

$$
\epsilon_k = \left( 1 - \gamma_k^2 \right)^{1/2}, \quad \gamma_k = \gamma_k / \kappa_k, \tag{10}
$$
$$
\gamma_{k_0} = [\cos(k_x) + \zeta \cos(k_y) + \delta \cos(k_z)] / (1 + \xi), \tag{11}
$$
$$
\gamma_{2k} = \cos(k_x) \cos(k_y), \tag{12}
$$
$$
\kappa_k = 1 - \frac{2\eta}{1 + \xi} (1 - \gamma_k) + \frac{\delta}{1 + \xi}. \tag{13}
$$

This completes the description of the quantum fluctuations for the AF Néel phase.
$\gamma_k$ is negative in certain parts of the first BZ, so it is essential to keep track of the sign of $\gamma_k$ through the function $\text{sgn}(\gamma_k)$. After these transformations, the quadratic part of the Hamiltonian becomes

$$H_0 = J_1 S_z (1 + \zeta) \sum_k \kappa_k (e_k - 1) + J_1 S_z (1 + \zeta) \sum_k \kappa_k e_k (a_k^\dagger a_k + \beta_k^\dagger \beta_k).$$

(9)

The first term is the zero-point energy and the second term represents the excitation energy of the magnons within LSWT.

The part $H_1$ corresponds to $1/S$ correction to the Hamiltonian. We follow the same procedure as described above. The resulting expression, after transforming the bosonic operators to the magnon operators, is

$$H_1 = \frac{J_1 S_z (1 + \zeta)}{2S} \sum_k [a_k^\dagger a_k + \beta_k^\dagger \beta_k]$$
$$+ B_k (a_k^\dagger a_{k+1} + \beta_k^\dagger \beta_{k+1}) - \frac{J_1 S_z (1 + \zeta)}{2SN} \times \sum \delta_k (1 + 2 - 3 - 4) f_{1234} [a_1^\dagger a_2^\dagger a_3^\dagger a_4^\dagger V_{1234}^{(1)}$$
$$+ \beta_1^\dagger \beta_2^\dagger \beta_3^\dagger \beta_4^\dagger V_{1234}^{(2)}$$
$$+ 4 a_1^\dagger a_2^\dagger a_3^\dagger a_4^\dagger V_{1234}^{(3)} + (2 a_1^\dagger \beta_2^\dagger a_3^\dagger a_4^\dagger V_{1234}^{(4)}$$
$$+ 2 \beta_1^\dagger \beta_2^\dagger \beta_3^\dagger \beta_4^\dagger V_{1234}^{(5)} + a_1^\dagger a_2^\dagger a_3^\dagger a_4^\dagger V_{1234}^{(6)}$$
$$+ \text{h.c.}].]$$

(10)

In the above equation 3D momenta $k_1, k_2, k_3, k_4$ are abbreviated as 1, 2, 3, and 4. The first term in equation (10) is obtained by setting the products of four boson operators into normal ordered forms with respect to the magnon operators, where $A_k$ and $B_k$ are

$$A_k = A_1 \frac{1}{k_k e_k} [y_k^2 - 1] + A_2 \frac{1}{\kappa_k} [1 - y_k],$$

(12)

$$B_k = B_1 \frac{1}{k_k e_k} y_k [1 - y_k],$$

with

$$A_1 = \frac{2}{N} \sum_p \frac{1}{\kappa_p} \left[ \gamma_p^2 + \epsilon_p - 1 \right],$$

(13)

$$A_2 = \frac{2}{N} \sum_p \frac{1}{\kappa_p} \left[ -1 - \gamma_p \right],$$

(14)

$$B_1 = \frac{2}{N} \sum_p \frac{1}{\kappa_p} \left[ \gamma_p \right].$$

(15)

The second term in equation (10) represents scattering between spin-waves where the 3D delta function $\delta_k (1 + 2 - 3 - 4)$ ensures that momentum is conserved within a reciprocal lattice vector $\mathbf{G}$. Explicit forms of the vertex factors $V_{1234}^{(1)} \sim 4$ are given in [23]. Here we provide two of the vertex factors that are needed to calculate the magnetization. They are

$$V_{1234}^{(4)} = -\gamma_1 (2 - 4) x_4 - \gamma_1 (1 - 4) x_1 x_2 x_4$$
$$- \gamma_1 (2 - 3) x_3 - \gamma_1 (1 - 3) x_1 x_2 x_3$$
$$+ \frac{1}{2} \gamma_1 (2) + \gamma_1 (1) x_1 x_2 + \gamma_1 (3) x_3 x_4 + \gamma_1 (4) x_2 x_4$$
$$+ \gamma_1 (2 - 3 - 4) x_3 x_4 + \gamma_1 (1 - 3 - 4) x_1 x_2 x_3 x_4$$
$$+ \gamma_1 (3 - 2 - 4) x_1 x_2 x_3 + \gamma_1 (4 - 2 - 4) x_1 x_2 x_3 x_4$$
$$+ \frac{2\eta}{1 + \zeta} f_{1234} [x_2 + \text{sgn}(\gamma_k) x_1 x_4].$$

(16)

$$V_{1234}^{(6)} = \gamma_1 (2 - 4) x_2 x_3 + \gamma_1 (2 - 3) x_2 x_4$$
$$+ \gamma_1 (1 - 3) x_1 x_4 + \gamma_1 (1 - 4) x_1 x_3$$
$$- \frac{1}{2} \gamma_1 (2) x_2 x_3 x_4 + \gamma_1 (3) x_4 + \gamma_1 (2 - 3) x_2$$
$$+ \gamma_1 (3 - 2 - 1) x_1 x_2 x_4$$
$$+ \gamma_1 (1) x_1 x_3 x_4 + \gamma_1 (4) x_4 + \gamma_1 (1 - 3) x_1$$
$$+ \gamma_1 (4 - 2 - 1) x_1 x_2 x_3$$
$$- \frac{2\eta}{1 + \zeta} f_{1234} [x_3 x_4 + \text{sgn}(\gamma_k) x_1 x_2].$$

(17)

with

$$f_{1234} = \frac{1}{2} \left[ \gamma_2 (1 - 3) + \gamma_2 (1 - 4) + \gamma_2 (2 - 3)$$
$$+ \gamma_2 (2 - 4) - \gamma_2 (1) - \gamma_2 (2) - \gamma_2 (3) - \gamma_2 (4) \right].$$

(18)

The second-order term, $H_2$ is composed of six boson operators. After transformation to magnon operators $\alpha_k, \beta_k$ the Hamiltonian in normal ordered form reduces to

$$H_2 = \frac{J_1 S_z (1 + \zeta)}{(2S)^2} \sum_k [C_{1k} (a_k^\dagger a_k + \beta_k^\dagger \beta_k)$$
$$+ C_{2k} (a_k^\dagger \beta_k^\dagger + \beta_k^\dagger a_k) + \cdots].$$

(19)

The dotted terms contribute to higher than second-order corrections and are thus omitted in our calculations. The coefficients $C_{1k}$ and $C_{2k}$ are

$$C_{1k} = \frac{1}{2} l_k^2 \left[ \frac{2}{N} \sum \frac{l_{12}^2}{l_{12}^2 - 6} \gamma_1 (2 - 1 - k) x_1 x_2$$
$$+ \gamma_1 (2) x_2 x_2 + \gamma_1 (2) x_1 x_2$$
$$+ 2 \gamma_1 (k) x_2 x_2 + 2 \gamma_1 (k x_2 x_2 + 2 \gamma_1 (1) x_1 x_1 + \gamma_1 (1) x_2 x_2)$$
$$- \frac{1}{4} \frac{(2\eta)}{(1 + \zeta)} f_k^2 (1 + x_k^2) \tilde{C}_k,$$

(20)

$$C_{2k} = \frac{1}{2} l_k^2 \left[ \frac{2}{N} \sum \frac{l_{12}^2}{l_{12}^2 - 13} \gamma_1 (2 - 1 - k) x_1 x_2$$
$$+ 3 \gamma_1 (2 - 1 - k) x_2 x_2 x_2 - 2 \gamma_1 (1) x_1 x_2 x_2$$
$$- 2 \gamma_1 (2) x_2 x_2 - 2 \gamma_1 (k x_2 x_2 + 2 \gamma_1 (1) x_1 x_2 x_2)$$
$$- \frac{1}{2} \frac{(2\eta)}{(1 + \zeta)} l_k m_k \tilde{C}_k,$$

(21)

The first-and second-order self-energies are written as

$$\Sigma_{\alpha\alpha}^{(1)} (k, \omega) = \Sigma_{\beta\beta}^{(1)} (k, \omega) = A_k,$$

(23)

$$\Sigma_{\alpha\beta}^{(1)} (k, \omega) = \Sigma_{\beta\alpha}^{(1)} (k, \omega) = B_k,$$

(24)
The Hamiltonian describing the CAF phase is

\[ H = J_1 \sum_{i,\ell} S_{i,\ell}^{A} S_{i+\delta,\ell}^{A} + J_2 \sum_{i,\ell} S_{i,\ell}^{A} S_{i+\delta,\ell}^{B} + J_3 \sum_{i,\ell} S_{i,\ell}^{B} S_{i+\delta,\ell}^{A} + J_4 \sum_{i,\ell} S_{i,\ell}^{B} S_{i+\delta,\ell}^{B} + J_5 \sum_{i,\ell} S_{i,\ell}^{A} S_{i+1,\ell}^{B} + J_6 \sum_{i,\ell} S_{i,\ell}^{B} S_{i+1,\ell}^{A}. \]

The procedure is the same as the AF phase. For this phase the structure factors \( y_{1k}', y_{2k}' \) along with other quantities required for the calculations are defined as

\[ y_{1k}' = [\cos(k_x)(1 + 2\eta \cos(k_y)) + \delta \cos(k_x)]/(1 + 2\eta), \]
\[ y_{2k}' = \cos(k_y), \quad y_{3k}' = y_{4k}'/y_{2k}', \]
\[ \kappa_k' = 1 - \frac{\zeta}{1 + 2\eta} (1 - y_{2k}') + \frac{\delta}{1 + 2\eta}, \]
\[ \epsilon_k' = [1 - y_{2k}']^{1/2}. \]

The coefficients that appear in the Hamiltonian \( H_1 \) are

\[ A_k' = A_1' \frac{1}{\kappa_k' \epsilon_k'}[\kappa_k' - y_{1k}'] + A_2' \frac{1}{\epsilon_k'}[1 - y_{2k}'], \]
\[ B_k' = B_1' \frac{1}{\kappa_k' \epsilon_k'} y_{1k}'[1 - y_{2k}']. \]

with

\[ A_1' = \frac{2}{N} \sum_{p} \frac{1}{\epsilon_p} \left[ \frac{\gamma_{ip}}{\epsilon_p} + \epsilon_p - 1 \right], \]
\[ A_2' = \left( \frac{\zeta}{1 + 2\eta} \right) \frac{2}{N} \sum_{p} \frac{1}{\epsilon_p} \left[ 1 - \epsilon_p - y_{2p} \right], \]
\[ B_1' = \left( \frac{\zeta}{1 + 2\eta} \right) \frac{2}{N} \sum_{p} \frac{1}{\epsilon_p} \left[ \gamma_{ip}^{2}/\epsilon_p \right]. \]

3. Magnetization and the phase diagram

We obtain the sublattice magnetization \( M \) for the two ordered phases with different values of \( \zeta, \eta, \) and \( \delta \) from equation (27) by numerically evaluating equations (28)–(30) (using similar expressions for the CAF phase). To obtain the second-order correction term \( M_Z \), we sum up the values of the \( N_L/8 \) points of \( k \) in the 1/8th part of the first BZ and \( N_L^3 \) points of the \( p \) and \( N_L^3 \) points of \( q \) in the full BZ. To check the convergence of our results we do the calculations for the AF phase with \( N_L = 8, 10, \) and 12 sites for \( \zeta = 1 \) and \( \delta = 0.1 \). The convergence is very good, as shown in figure 1. \( M_{AF} \) becomes zero at the critical point \( \eta_c \approx 0.460 \). Hereafter, we use \( N_L = 12 \) lattice sites for all of our numerical computations. Evaluation of the magnetization requires summing contributions from over 645 million points in the first BZ for each \( \zeta, \eta, \) and \( \delta \).

Figure 2 shows the sublattice magnetization \( M_0^{AF} \) for the AF phase with \( \zeta = 1 \) and \( \eta = 0 \) for different values of interlayer coupling \( \delta \). In the inset we plot the spin-deviation \( \Delta = 0.5M_0^{AF} \) with \( \delta \). \( \Delta \) is a measure of quantum fluctuations from the classical value of 0.5. We find that with increase in \( \delta \) the fluctuations decrease, thus \( M_0^{AF} \) (with second-order corrections) increases from 0.308 for \( \delta = 0 \) to 0.423 for
in the frustration parameter $\eta$

unfrustrated case (dotted line) is the result from LSWT and the solid line is with 1 decrease, thus with line), and 12 (open circles) lattice sites. The convergence is excellent with $N_L = 12$ sites.

Figure 2. Magnetization $M_{AF}^0$ for the AF phase plotted for the unfrustrated case ($\eta = 0$) with interlayer coupling $\delta$ for $\xi = 1.0$. The dotted line is the result from LSWT and the solid line is with $1/S^2$ corrections. $M_{AF}^0$ (with second-order corrections) increases from 0.308 for $\delta = 0$ to 0.423 for $\delta = 1.0$. The inset shows the spin-deviation $\Delta = 0.5M_{AF}^0$. With increase in $\delta$ the fluctuations decrease, thus $M_{AF}^0$ increases. This result is expected as with an increase in the interlayer coupling the system makes a transition from two to three dimensions.

$\delta = 1.0$. This result is expected as with the increase in interlayer coupling the system undergoes a dimensional transition from 2D to 3D. The dotted line is the result from LSWT. The result for $M_{AF}^0$ obtained from LSWT captures the essential physics both qualitatively and quantitatively as second-order corrections are small for the unfrustrated ($\eta = 0$) case.

Figures 3 and 4 show the magnetization with increase in the frustration parameter $\eta$ for two different values of the spatial anisotropy parameter $\xi = 0.9$ and 0.6. Similar to the 2D case (see [23] for details) our spin-wave expansion for the CAF phase becomes unreliable as $\xi$ gets close to 1. Thus we have not chosen $\xi = 1$ for our plots. For each $\xi$ and $\delta$, three different curves are plotted: the long-dashed lines represent LSWT results, the dotted lines include the first-order $1/S$ corrections, and the solid lines represent corrections up to second-order for the LSWT results. As $\eta$ approaches the classical transition point $\eta_{cl}^{class} = 0.45$ from both sides of the two ordered phases the dotted curves diverge. However, $1/S^2$ corrections ($M_2$) significantly increase to stabilize the divergence. We find that the magnetizations with $1/S^2$ corrections decrease steadily and then sharply drop to zero for both the phases as $\eta \to \eta_{cl}^{class}$. As an example, with $\xi = 0.9, \delta = 0.1$ magnetization for the AF phase begins from $\approx 0.377$ at $\eta = 0$, then decreases until $\eta \approx 0.41$, and sharply goes to zero at the critical point $\eta_{cl} \approx 0.427$. $M_2$ corrections start from a small positive number at $\eta = 0$ and then switch sign at $\eta \approx 0.32$. $M$ for the CAF phase shows the same feature, where we find $M \approx 0.393$ at $\eta = 1$ and $\eta_{2c} \approx 0.464$. However, in this case $M_2$ corrections are always negative. These results are qualitatively similar to the results for the 2D spatially anisotropic, frustrated HAFM on a square lattice ($\delta = 0$) [23].
both the phases at the critical points direct first-order phase transition from the AF to the CAF phase.

ζ shown for spatial anisotropy ζ decrease in spatial anisotropy (the results are not shown).

ζ decreases steadily and then sharply drops to zero for both the phases.

η large of the disordered PM phase diminishes but it survives even for

corrects) for both the phases at the critical points ζ = 0. The two phases are separated by a narrow disordered paramagnetic region. We repeat the calculations for ζ = 0.4 with different values of δ and obtain similar features (the results are not shown).

Another feature we observe from our data is that with decrease in spatial anisotropy ζ the width of the disordered region (η_2c − η_1c) diminishes but it never disappears. This is shown in figure 5 for δ = 0.1. The solid lines represent the critical points η_1c and η_2c for the AF and CAF phases. The dashed line shows the classical phase transition line η_{c\text{class}} = ζ/2 (independent of δ) between the two phases. For the AF phase η_{1c} ≈ 0.460 at ζ = 1. The spin-wave expansion for the CAF phase becomes unreliable for ζ near 1. We thus extrapolate our data to obtain η_{2c} ≈ 0.53 for ζ = 1. The width of the disordered region (η_{2c} − η_{1c}) increases with the anisotropy parameter ζ.

On the other hand, LSWT calculations for ζ = 0.9 and δ = 0.1 show that the magnetization goes to zero for the AF phase at η_{1c} ≈ 0.45 whereas it does not go to zero from the CAF phase. LSWT is not applicable at the classical transition point η = 0.45. Extrapolation of the CAF phase LSWT results show a direct transition from the AF to the CAF phase.

For ζ = 0.9 and δ = 1.0, M never goes to zero for either of the AF and CAF ordered phases (within LSWT), indicating that there is a direct first-order phase transition from the AF to the CAF phase. But with second-order corrections we find that M goes to zero for both the phases at the critical points η_{1c} ≈ 0.434 and η_{2c} ≈ 0.469. With increase in δ the width of the disordered PM phase diminishes but it survives even for large δ.

In figure 4 we show the results for ζ = 0.6. LSWT calculations show a first-order direct phase transition from AF to the CAF phase for both δ = 0.1 and 1.0. Similar to the ζ = 0.9 case we find that M vanishes (with second-order corrections) for both the phases at the critical points η_{1c} ≈ 0.288, η_{2c} ≈ 0.311 for δ = 0.1 and η_{1c} ≈ 0.290, η_{2c} ≈ 0.310 for δ = 1.0. The two phases are separated by a narrow disordered paramagnetic region. We repeat the calculations for ζ = 0.4 with different values of δ and obtain similar features (the results are not shown).

Another feature we observe from our data is that with
Figure 6. Phase diagram for the $J_1-J_2-J_3-J_4$ model with $\zeta = 0.9$. The dashed line shows the classical phase transition line $\eta_{\text{class}} = 0.45$. The phase boundaries of the two ordered phases AF and CAF diminish but never disappear with increase in $\delta$. These two ordered phases are always separated by the magnetically disordered phase. For the AF phase $\eta_{\text{AF}}$ diminishes with decrease in the directional anisotropy. These points for the two phases differ from the LSWT predictions. Linear spin-wave theory calculations for this model show that there are two magnetic ordered phases (Néel and stripe) which are separated by a paramagnetic phase. For the AF phase $\eta_{\text{AF}}$ begins from $\approx 0.484$ and then slowly decreases to $\approx 0.461$ for $\delta = 0.3$. But from $\delta \approx 0.4$ the phase boundary for the CAF phase shows a slight upward rise. By excluding the $J_2$ interactions in the $xz$ and $yz$ planes our model becomes unrealistic in the strong interlayer coupling limit where $J_2$ is comparable to $J_1$ and $J'_2$ (especially in the CAF phase).

4. Conclusions

In this work using a second-order spin-wave expansion we have studied the effects of interlayer coupling and directional anisotropy on the magnetic phase diagram of a frustrated spin-$\frac{1}{2}$ Heisenberg antiferromagnet on a stacked square lattice. Linear spin-wave theory calculations for this model show that for small interlayer coupling there are two magnetic ordered phases, AF and CAF which are separated by a disordered paramagnetic state. However, when the interlayer coupling exceeds a critical value the disordered paramagnetic phase disappears and then there is a direct first-order phase transition from the AF to the CAF phase. Recent numerical calculations using coupled-cluster and rotation-invariant Green’s function methods support this picture [72, 73]. With our second-order spin-wave expansion we have found that with increase in next nearest neighbor frustration, $1/S^2$ corrections play a significant role in stabilizing the magnetization as the classical phase transition point is approached. As expected from linear spin-wave theory we have found that there are two ordered magnetic phases (Néel and stripe) which are separated by a paramagnetic disordered phase. But the values of the critical phase transition points for the two phases differ from the LSWT predictions. Our calculations show that the width of the disordered region diminishes with decrease in the directional anisotropy. These features are similar to the magnetic phase diagram of a 2D frustrated Heisenberg spin-$\frac{1}{2}$ antiferromagnet [23]. However, with increase in the interlayer coupling we have found that the parameter region of this disordered phase does not disappear. Our obtained phase diagram is significantly different from the phase diagram obtained using linear spin-wave theory which predicts a direct first-order phase transition from the AF to the CAF phase beyond a critical value of interlayer coupling.

In summary, with our present approach based on a second-order spin-wave expansion we do not find the existence of any critical interlayer coupling (or spatial anisotropy) beyond (or below) which there is a direct transition from one phase to the other ordered phase.

Acknowledgments

The author thanks H Johannesson for useful discussions and suggestions, and acknowledges the use of the Cornell Center for Advanced Computing’s ‘MATLAB on the TeraGrid’ experimental computing resource funded by NSF grant 0844032 in partnership with Purdue University, Dell, The MathWorks and Microsoft.

References

[1] Kim Y J et al 1999 Phys. Rev. Lett. 83 852
[2] Coldea R, Hayden S M, Aepli G, Perring T G, Frost C D, Mason T E, Cheong S-W and Fisk Z 2001 Phys. Rev. Lett. 86 5377
[3] Ronnow H M, McMorrow D F, Coldea R, Harrison A, Youngson I D, Perring T G, Aepli G, Syljuasen O, Lefmann K and Rischel C 2001 Phys. Rev. Lett. 87 037202
[4] Christensen N B, McMorrow D F, Ronnow H M, Harrison A, Perring T G and Coldea R 2004 J. Magn. Magn. Mater. 272–276 896
[5] Christensen N B, Ronnow H M, McMorrow D F, Harrison A, Perring T G, Enderle M, Coldea R, Regnault L P and Aepli G 2007 Prog. Natl Acad. Sci. USA 104 15264
[6] Bombardi A, Rodriguez-Carvajal J, Matteo S D, de Bergevin F, Paolasini L, Carretta P, Millet P and Caciuffo R 2004 Phys. Rev. Lett. 93 027202
[7] Melzi R, Carretta P, Lascialfari A, Mambrini M, Troyer M, Millet P and Mila F 2000 Phys. Rev. Lett. 85 1318
[8] Melzi R, Aldrovandi S, Tedoldi F, Carretta P, Millet P and Mila F 2001 Phys. Rev. B 64 024409
[9] Carretta P, Papinutto N, Azzoni C B, Mozzati M C, Parvarini E, Gontier S and Millet P 2002 Phys. Rev. B 66 094420
[10] Rosner H, Singh R, Zheng W H, Oitmaa J, Drechsler S L and Picket W E 2002 Phys. Rev. Lett. 88 186405
[11] Kamihara Y, Watanabe T, Hiramatsu N and Hosono H 2008 J. Am. Chem. Soc. 130 3296
[12] de la Cruz C et al 2008 Nature 453 899
[13] Klaus H-H et al 2008 Phys. Rev. Lett. 101 077005
[14] Dong J et al 2008 Europhys. Lett. 83 27006
[15] Harris A B, Kumar D, Halperin B I and Hohenberg P C 1971 Phys. Rev. B 3 961
[16] Chandra P and Doucet B 1988 Phys. Rev. B 38 9335
[17] Chakravarty S, Halperin B I and Nelson D R 1989 Phys. Rev. B 39 2344
[18] Castilla G E and Chakravarty S 1991 Phys. Rev. B 43 13687
[19] Canali C M and Girvin S M 1992 Phys. Rev. B 45 7127
[20] Igarashi J I 1992 Phys. Rev. B 46 10763
[21] Igarashi J I 1993 J. Phys. Soc. Japan 62 4449
[22] Capriotti L 2003 Int. J. Mod. Phys. B 17 4819
[23] Majumdar K 2010 Phys. Rev. B 82 144407
[24] Dotsenko A V and Sushkov O P 1994 Phys. Rev. B 50 13821
[25] Kotov V N, Oitmaa J, Sushkov O P and Zheng W H 1999 Phys. Rev. B 60 14613
[26] Nersesyan A A and Tsvelik A M 2003 Phys. Rev. B 67 024422
[27] Starykh O and Balents L 2004 Phys. Rev. Lett. 93 127202
[28] Shannon N, Schmidt B, Penc K and Thalmeier P 2004 Eur. Phys. J. B 38 599
[29] Iaiev L, Ortiz G and Dukelsky J 2009 Phys. Rev. B 79 024409
[30] Weiwhong Z, Oitmaa J and Hamer C J 1991 Phys. Rev. B 44 11869
[31] Hamer C J, Weiwhong Z and Arndt P 1992 Phys. Rev. B 46 6276
[32] Oitmaa J and Weiwhong Z 1996 Phys. Rev. B 54 3022
[33] Zheng W, Oitmaa J and Hamer C J 2005 Phys. Rev. B 71 184440
[34] Sindzingre P 2004 Phys. Rev. B 69 094418
[35] White S R and Chernyshev A L 2007 Phys. Rev. Lett. 99 127004
[36] Tsirlin A A and Rosner H 2009 Phys. Rev. B 79 214417
[37] Sandvik A W and Singh R R P 2001 Phys. Rev. Lett. 86 528
[38] Capriotti L, Becca F, Parola A and Sorella S 2003 Phys. Rev. B 67 212402
[39] Yunoki S and Sorella S 2004 Phys. Rev. Lett. 92 157003
[40] Einarsson T and Johansson H 1991 Phys. Rev. B 43 5867
[41] Einarsson T, Fröjd P and Johansson H 1992 Phys. Rev. B 45 13121
[42] Diep H T 2004 Frustrated Spin Systems 1st edn (Singapore: World Scientific)
[43] Sachdev S 2001 Quantum Phase Transitions 1st edn (Cambridge: Cambridge University Press)
[44] Pavarini E, Tarantino S C, Ballaran T B, Zema M, Ghigna P and Carretta P 2008 Phys. Rev. B 77 014425
[45] Volkova O et al 2010 Phys. Rev. B 82 054413
[46] Majumdar K and Datta T 2010 J. Stat. Phys. 139 714
[47] Majumdar K and Datta T 2009 J. Phys.: Condens. Matter 21 406004
[48] Oitmaa J and Zheng W 2004 Phys. Rev. B 69 064416
[49] Lallemand P, Diep H T, Ghazali A and Toulouse G 1985 J. Physique Lett. 46 1087
[50] Azaria P 1986 J. Phys. C: Solid State Phys. 19 2773
[51] Derrida B, Pomeau Y, Toulouse G and Vannimenus J 1979 J. Physique 40 617
[52] Derrida B, Pomeau Y, Toulouse G and Vannimenus J 1980 J. Physique 41 213
[53] Oguchi T, Nishimori H and Taguchi Y 1985 J. Phys. Soc. Japan 54 4494
[54] Ignatenko A N, Katanin A A and Irkhin V Y 2008 JETP Lett. 87 1
[55] Banavar J R, Jasnow D and Landau D P 1979 Phys. Rev. B 20 3820
[56] Canals B and Lacroix C 1998 Phys. Rev. Lett. 80 2933
[57] Fak B, Coomer F C, Harrison A, Visser D and Zhitomirsky M E 2008 Eur. Phys. Lett. 81 17006
[58] Sachdev S 1992 Phys. Rev. B 45 12377
[59] Harris A B, Kallin C and Berlinsky A J 1992 Phys. Rev. B 45 2899
[60] Chubukov A 1992 Phys. Rev. Lett. 69 832
[61] Schmidt R, Schulenberg J and Richter J 2002 Phys. Rev. B 66 224406
[62] Applegate R, Oitmaa J and Singh R R P 2010 Phys. Rev. B 81 024505
[63] Singh R R P 2009 Supercond. Sci. Technol. 22 015005
[64] Uhrig G S, Holt M, Oitmaa J, Sushkov O P and Singh R R P 2009 Phys. Rev. B 79 092416
[65] Yao D X and Carlson E W 2008 Phys. Rev. B 78 052507
[66] Yao D X and Carlson E W 2010 Front. Phys. China 5 166
[67] Zhao J et al 2008 Phys. Rev. Lett. 101 167203
[68] Diallo S O et al 2009 Phys. Rev. Lett. 102 187206
[69] Zhao J, Adroja D T, Yao D-X, Bewley R, Li S, Wang X F, Wu G, Chen X H, Hu J and Dai P 2009 Nat. Phys. 5 555
[70] Ewings R, Perrin T, Bewley R, Guidi T, Pitcher M, Parker D R, Clarke S J and Boothroyd A T 2008 Phys. Rev. B 78 220501(R)
[71] Yuan H Q, Singleton J, Balakirev F F, Baily S A, Chen G F, Luo J L and Wang N L 2009 Nature 457 565
[72] Schmalfuß D, Darradi R, Richter J, Schulenburg J and Ihle D 2006 Phys. Rev. Lett. 97 157201
[73] Nunes W A, de Sousa J, Viana J R and Richter J 2010 J. Phys.: Condens. Matter 22 146004