The two-dimensional frustrated Heisenberg model on the orthorhombic lattice

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Abstract. We discuss new high-field magnetization data recently obtained by Tsirlin et al. for layered vanadium phosphates in the framework of the square-lattice model. Our predictions for the saturation fields compare exceptionally well to the experimental findings, and the strong bending of the curves below saturation agrees very well with the experimental field dependence. Furthermore we discuss the remarkably good agreement of the frustrated Heisenberg model on the square lattice in spite of the fact that the compounds described with this model actually have a lower crystallographic symmetry. We present results from our calculations on the thermodynamics of the model on the orthorhombic (i.e., rectangular) lattice, in particular the temperature dependence of the magnetic susceptibility. This analysis also sheds light on the discussion of magnetic frustration and anisotropy of a class of iron pnictide parent compounds, where several alternative suggestions for the magnetic exchange models were proposed.

1. Introduction

The frustrated \( S = 1/2 \) Heisenberg model on the square lattice, the \( J_1-J_2 \) model, appears to describe well the thermodynamic and magnetic properties of two classes of vanadium compounds of type \( \text{Li}_2\text{VOXO}_4 \) (\( X = \text{Si, Ge} \)) [1] and \( \text{AA}’\text{V}(\text{PO}_4)_2 \) (\( A, A’ = \text{Pb, Zn, Sr, Ba} \)) [2, 3]. They consist of V-oxide pyramid layers containing \( \text{V}^{4+} \) ions with \( S = 1/2 \). From the analysis of the temperature dependence of the heat capacity and the magnetic susceptibility in zero (or small) fields, the frustration ratio \( J_2/J_1 \) can be obtained [4]. However, an ambiguity remains with respect to the relative sign of the two exchange constants, which can be resolved by analyzing the behavior of these materials in finite fields [5]. The average exchange constants \( J_c = \sqrt{J_1^2 + J_2^2} \) of these materials are low enough such that their saturation fields are experimentally accessible. In this article we discuss the high-field magnetization of the \( J_1-J_2 \) model and compare our findings to recent measurements [6].

Although we describe the physics of the \( J_1-J_2 \) compounds using a square-lattice model, their true crystal structure corresponds to a two-dimensional lattice with lower symmetry. Therefore, we introduce an additional spatial anisotropy in the ab plane assuming orthorhombic symmetry, i.e., a rectangular lattice.

2. Model Hamiltonian

The effective Hamiltonian on the rectangular lattice has the form

\[
\mathcal{H} = J_{1a} \sum_{\langle ij \rangle_{1a}} \vec{S}_i \cdot \vec{S}_j + J_{1b} \sum_{\langle ij \rangle_{1b}} \vec{S}_i \cdot \vec{S}_j + J_2 \sum_{\langle ik \rangle_{2}} \vec{S}_i \cdot \vec{S}_k
\]
Fig. 1. Left: Comparison of saturation fields from ED and spin-wave theory with \( \theta = \frac{\pi}{4} \), and from high-field experiments [6]. The compounds are (1) PbZnVO(PO₄)₂, (2) Na₃₋₅VOPO₄F₀₋₅, (3) Pb₂VO(PO₄)₂, (4) SrZnVO(PO₄)₂, and (5) BaCdVO(PO₄)₂. The agreement with the saturation fields for the columnar phase gives a direct proof that all compounds have CAF order. Right: Magnetization for BaCdVO(PO₄)₂ [3]. Solid line denotes experimental data and filled symbols the data from \( T = 0 \) Lanczos calculations for different cluster sizes.

where \( J_{1a} \) and \( J_{1b} \) denote the nearest-neighbor exchange along the \( a \) and \( b \) directions, and \( J_2 \) labels the diagonal next-nearest neighbor exchange. A more convenient parametrization is

\[
J_{1a} = \sqrt{2} J_c \cos \phi \cos \theta, \quad J_{1b} = J_c \sin \phi, \\
J_2 = J_c = \sqrt{\frac{1}{2} (J_{1a}^2 + J_{1b}^2 + J_2^2)},
\]

(2)

introducing a frustration angle \( \phi \), an anisotropy parameter \( \theta \), and an overall energy scale \( J_c \). For \( \theta = \pi/4 \), the above Hamiltonian reduces to the square-lattice case.

The results presented in the following paragraphs are obtained by applying linear spin-wave theory (LSW) and exact diagonalization (ED) for finite clusters at finite temperatures (FTLM) [4, 5, 7].

3. Saturation field

From LSW the saturation field for the square lattice is given by the instability of the fully polarized state against a single-magnon excitation. This result is in exact agreement with ED for antiferromagnetic \( J_2 \). However, exact diagonalization for our finite clusters reveals that for ferromagnetic \( J_1 \), a \( \Delta S = 2 \) instability determines the saturation field [5].

The left-hand side of Fig. 1 shows a comparison of the saturation fields determined by LSW and ED for the columnar (CAF) and Néel (NAF) antiferromagnetic phases with the experimental values determined from high-field measurements [6]. The predicted theoretical values are based on fits of our FTLM data and of a high-temperature series expansion [2, 3, 6, 8] to the temperature dependences of the low-field susceptibilities. The experiments agree surprisingly well with the predicted CAF values, demonstrating that all compounds order in a columnar magnetic structure at low temperatures.

On the right-hand side of Fig. 1, the field dependence of the magnetization for BaCdVO(PO₄)₂ is displayed, together with zero-temperature data from our Lanczos calculations for different cluster sizes using a Bonner-Fisher construction [5, 9]. Given the small size of the clusters involved, the agreement is well, apart from low fields, where finite-size effects are most prominent.
Figure 2. FTLM results for the magnetic susceptibility $\chi(T)$. The left-hand figures show the position, the right-hand ones the temperature where $\chi(T)$ reaches its maximum. Top: Dependency on the frustration angle $\phi$. The vertical lines distinguish the different classical phases, FM, NAF and CAF. Bottom: Dependency on the anisotropy parameter $\theta$ at fixed frustration angle $\phi/\pi = 0.625$. The vertical line shows the isotropic case, $\theta = \pi/4$.

4. Extension to the orthorhombic (rectangular) case

Up to here, we discussed the layered vanadium phosphates in the context of the square-lattice Heisenberg model. However, their crystal structure corresponds to a lattice with lower symmetry [10]. We therefore investigate the impact of an additional anisotropy of the nearest-neighbor interactions in the ab plane, characterized by the angle $\theta$ defined in Eq. 2.

In Fig. 2, FTLM results for a tile of size 20 are shown. The position (left) and the value (right) of the broad maximum of the magnetic susceptibility $\chi(T)$ are plotted as a function of the frustration ($\phi$) and the anisotropy ($\theta$) angles. The top curves correspond to the isotropic case ($J_{1\alpha} = J_{1\beta}, \theta = \pi/4$), while the bottom ones show the effect of the anisotropy parameter $\theta$ for constant $\phi/\pi = 0.625$ (CAF regime). The change both in value and temperature is small. Therefore introducing an anisotropy within the columnar phase has comparatively little effect on the temperature dependence of $\chi(T)$. For further clarification, a contour plot of the ground state energy (right) of the Hamiltonian, Eq. 1 and the temperature of the maximum of $\chi(T)$ (left) as a function of $\phi$ and $\theta$ are shown in Fig. 3. The model has four classical phases, one FM, one Néel AF, and two columnar AF phases along the crystallographic $a$ and $b$ directions. Inside the AF regions, the parameter dependence is weak. This explains the validity of the square-lattice Heisenberg model in describing the experimental results on the thermodynamics of the compounds, which have lower than tetragonal symmetry.

It has been proposed that magnetic frustration is a key feature of the magnetic properties of ferropnictides. However, several different exchange models are discussed [11, 12, 13, 14], in particular models with a spatial anisotropy as described here. As an example, Table 1 shows the experimental and theoretical [15] values for moments and exchange constants in some iron pnictide parent compounds, which are all in the CAF regime. We conclude that the anisotropy of $J_{1\alpha,\beta}$ stabilizes the CAF phase. In particular the values with $J_{1\beta} \simeq 0$ and $J_{1\alpha}/2J_2 \simeq 1$ ($\theta \simeq 0, \phi \simeq 0.15\pi$) correspond to the stable CAF region. It is obvious from Fig. 3 (left) that
Figure 3. Left: Contour plot of ground state energy as function of anisotropy ($\theta$) and frustration ($\phi$). Open circle designates usual NAF ($\theta = \frac{\pi}{4}$, $\phi = 0$). Values in table 1 are represented by black (exp.) and white (theory) symbols. The white lines show the boundaries between the four classical phases, CAF$_{a,b}$, NAF and FM. Right: Contour plot for the position of the maximum susceptibility.

Table 1. Fe pnictide moment $\mu = 2S\mu_B$ and exchange interactions (in meV) from experiment (top) and theory (bottom).

| System       | Ref. | $S$ | $S_J_{1a}$ | $S_J_{1b}$ | $S_J_{2}$ | $S_J_{c}$ | $\phi/\pi$ | $\theta/\pi$ |
|--------------|------|-----|------------|------------|-----------|-----------|------------|------------|
| CaFe$_2$As$_2$ | 11   | -   | 41         | 10         | 21        | 36        | 0.19       | 0.08       |
| CaFe$_2$As$_2$ | 12   | 0.4 | 24-37      | 7-20       | 28-34     | 33-45     | 0.29       | 0.13       |
| CaFe$_2$As$_2$ | 14   | 0.22| 49.9       | -5.7       | 18.9      | 53.7      | 0.11       | -0.04      |
| BaFe$_2$As$_2$ | 13   | 0.28| 17.5       | 17.5       | 35        | 39.1      | 0.35       | 0.25       |
| BaFe$_2$As$_2$ | 13   | 0.54| 36         | -7         | 18        | 31.6      | 0.19       | -0.06      |
| CaFe$_2$As$_2$ | 15   | 0.75| 27.4       | -2.1       | 14.5      | 24.3      | 0.20       | -0.02      |
| BaFe$_2$As$_2$ | 15   | 0.84| 36.1       | -2.6       | 12.0      | 38.0      | 0.10       | -0.02      |
| SrFe$_2$As$_2$ | 15   | 0.84| 35.3       | 2.2        | 13.4      | 28.4      | 0.16       | 0.02       |

these values are quite distant to the strongly frustrated point ($\theta = 0.25\pi$, $\phi = 0.15\pi$) where the CAF$_{a,b}$ and NAF phases meet. The moment reduction by quantum fluctuations for the former values is comparable to that of the simple unfrustrated NAF (open circle).

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