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The frustrated J_1 – J_2 model in high magnetic fields

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Abstract

We investigate thermodynamic properties of the frustrated J_1 – J_2 spin $S = 1/2$ model by using the finite-temperature Lanczos method for small clusters on a square lattice. We focus on the global properties of specific heat, susceptibility and spin structure factor as a function of the frustration angle $\phi = \tan^{-1}(J_2/J_1)$. Specifically, we discuss the high-field properties and the saturation field and its dependence on ϕ . We show that the asymmetry between collinear and Néel antiferromagnetic states gives an additional criterion to distinguish between these phases. Furthermore, we investigate the magnetocaloric effect and show that large peaks occur close to the saturation field. The J_1 – J_2 spin model is realized in some layered quasi-2D vanadium perovskites. They have characteristic values for J_1 and J_2 of about 10 K. The associated saturation fields are of the order of 40 T and therefore easily accessible in pulsed-field experiments.

1. Introduction

Low-dimensional spin systems offer the possibility of studying genuine quantum effects, which are usually hidden in higher dimension due to the onset of long-range magnetic order at finite temperature, where quantum fluctuations are still masked by thermal fluctuations. For a 2D square lattice the moment reduction factor of 0.63 in the Néel antiferromagnetic (AF) ground state is a prominent example of the effect of quantum fluctuations. Earlier hopes that they might be strong enough to lead to a disordered RVB-like spin-liquid ground state have been disproven. To stabilize such a state one has two options. Firstly a different lattice structure with geometrical frustration such as triangular and Kagomé lattices leads to a continuous degeneracy of classical broken symmetry states and an ensuing disordered ground state. Secondly, on geometrically nonfrustrated lattices a more complex pattern of exchange bonds e.g. including nnn diagonal exchange J_2 in addition to nn exchange J_1 on a square lattice leads to a frustration of interactions. The corresponding J_1 – J_2 model is given by the Hamiltonian

$$\mathcal{H} = J_1 \sum_{\langle ij \rangle_1} \mathbf{S}_i \mathbf{S}_j + J_2 \sum_{\langle ij \rangle_2} \mathbf{S}_i \mathbf{S}_j. \quad (1)$$

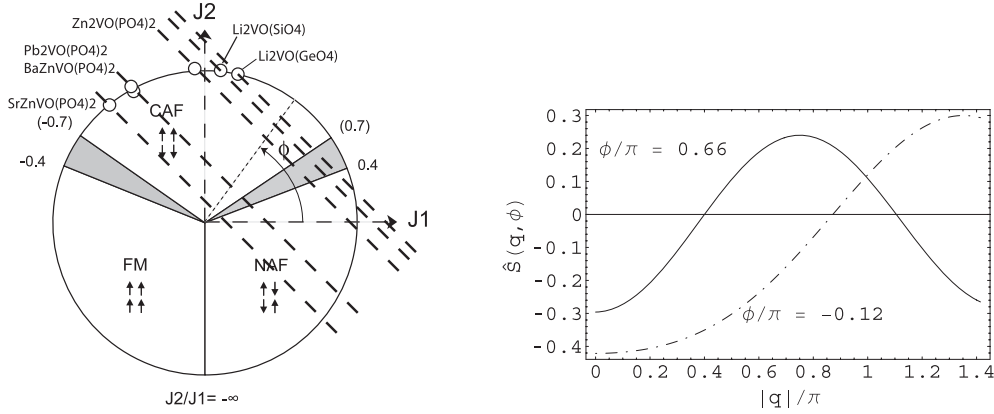


Figure 1. Left: phases of the J_1 – J_2 model. The AF order of CAF and NAF is indicated; the associated wavevectors are $\mathbf{Q} = (0, 1)$ or $(1, 0)$ and $(1, 1)$ (in units of π/a) respectively. The boundary between FM and NAF phases is the line $J_1 = 0, J_2 < 0$. Values of J_2/J_1 in parentheses show where zero-point fluctuations destroy the CAF order parameter [2]. Dashed lines correspond to the experimental $\Theta_{CW} = (J_1 + J_2)/k_B$ and refer to the known J_1 – J_2 compounds [7, 8]. Right: angular averaged prefactor of $(1/T)$ term in the high-temperature expansion of the structure factor (2). Frustration angles ϕ correspond to CAF (full line) and NAF (broken line) candidates for $Pb_2VO(PO_4)_2$.

For a review see, e.g., [1]. More complex models involving additional ring exchange [2] have also been studied. Alternatively, one may set $J_{1,2} = J$ and leave out the diagonal exchange in every other square. This leads to the geometrically frustrated checkerboard lattice, which is a 2D projection of the 3D pyrochlore lattice. Thus the two different types of frustration are not mutually exclusive.

In this work we focus on the above J_1 – J_2 model on a square lattice. Depending on the exchange ratio, which may be parametrized by the ‘frustration angle’ $\phi = \tan^{-1}(J_2/J_1)$, this model exhibits both phases with long-range magnetic order and disordered ‘spin liquid’-like phases, that may, however, be characterized by hidden order parameters [2]. The possible phases in the J_1 – J_2 plane are shown in figure 1 comprising three ordered phases, NAF (Néel antiferromagnet), CAF (collinear antiferromagnet) and FM (ferromagnet), and two smaller sectors without long-range magnetic order. Classically, only the former exist and the lines separating them may be interpreted in a simple manner [3]. At the FM/AF boundary ($J_2 < 0$) one sublattice of the AF may be rotated by $\pi/2$ with simultaneous sign change $J_1 \rightarrow -J_1$. This leaves the classical energy unchanged. At the CAF/NAF boundary a breakup into stripes of the two phases is possible due to a vanishing domain wall energy. At the CAF/FM boundary arbitrary coplanar spiral states with wavevectors $(2\pi n/m, 0)$ with n, m being arbitrary integers become degenerate and destabilize the CAF phase. Quantum fluctuations are unimportant at the FM/NAF boundary because the orientation of sublattices is determined already on the mean field level for arbitrary small J_1 . At the other boundaries quantum fluctuations lead to narrow magnetically disordered sectors of finite widths due to a continuous degeneracy of classical phases. However, nonmagnetic hidden order has been proposed in these sectors. Actually, quantum fluctuations are also important within the CAF phase. At $J_1 = 0$ ($J_2 > 0$) there are two decoupled Néel sublattices, which may be rotated continuously. Because the energy gain due to quantum fluctuations is maximized when the two sublattices are parallel, the CAF order is selected. Thus at this (broken) line one has the ‘order-by-disorder’ selection of the true magnetic order.

Most theoretical work on this model was focused on the AF side with $J_2 > 0$ and especially on the spin liquid phase around $J_2/J_1 = 0.5$. However, most of the compounds discussed here rather lie on the ferromagnetic side of J_1 as was first proposed in [4].

2. J_1 – J_2 compounds

Physical realizations of J_1 – J_2 have been rather scarce; the first one was $\text{Li}_2\text{VO}(\text{SiO}_4)$ [5, 6] and more recently this was followed by $\text{Pb}_2\text{VO}(\text{PO}_4)_2$ [8]. The full list of compounds known is shown in the legend of figure 1. These quasi-2D layered perovskites can be grouped into two classes, namely Li_2VOXO_4 ($X = \text{Si}, \text{Ge}$) and $\text{AA}'\text{VO}(\text{PO}_4)_2$ ($A, A' = \text{Pb}, \text{Zn}, \text{Sr}, \text{Ba}$) [7, 8]. So far only polycrystalline samples are available. The V^{4+} ($S = 1/2$) ions are surrounded by oxygen polyhedra, which form a square lattice in each (generally buckled) layer. In all compounds the scale of exchange energies as given by $J_c = (J_1^2 + J_2^2)^{1/2}$ is of the order of 10 K, which is comparatively low for V oxides. This is a favourable circumstance since low-temperature specific heat is not strongly masked by phonon contributions; in addition it makes the compounds accessible for high-field experiments. All compounds order magnetically at Néel temperatures of a few kelvin due to the effect of inter-layer coupling. This is safely below the characteristic 2D fluctuation peak in the susceptibility $\chi(T)$, which is at a temperature of the order of J_c/k_B .

3. Thermodynamics, structure factor and the ambiguity of frustration angles

To determine in which sector of the phase diagram the V oxides are to be placed, the temperature dependences of specific heat $C_V(T)$ and magnetic susceptibility $\chi(T)$ have been measured. For example, from the peak in $C_V(T)$ one obtains the energy scale J_c and from the high-temperature tail of $\chi(T)$ one obtains the Curie–Weiss temperature $\Theta_{\text{CW}} = (J_1 + J_2)/k_B$. Their intersection in figure 1 should determine the frustration angle. It is obvious that there are always two solutions for ϕ lying generally in different sectors of the phase diagrams. For the V oxides under consideration NAF as well as CAF are possible candidates for the ground state as seen in figure 1. Choosing other criteria like the ratio $\Theta_{\text{CW}}/T_\chi$ (T_χ is the maximum position of $\chi(T)$) leads to the same result [3]. Even a comparison of the whole $C_V(T)$ or $\chi(T)$ curves cannot resolve this ambiguity [9]. Different methods of analysis are necessary to achieve this goal. The most direct approach is the investigation of the spin structure factor given by

$$S(\mathbf{q}, T) = \frac{1}{N} \sum_{i,j=1}^N e^{i\mathbf{q}(\mathbf{R}_i - \mathbf{R}_j)} \langle \mathbf{S}_i \mathbf{S}_j \rangle. \quad (2)$$

This correlation function has been calculated with the Lanczos method for the various sectors [3]. Since only polycrystalline samples are available, neutron diffraction experiments can only explore the \mathbf{q} -averaged structure factor.

From a high-temperature series expansion the angular averaged prefactor $\hat{S}(q, \phi)$ ($q = |\mathbf{q}|$) of the $(1/T)$ contribution in (2) may be obtained. It is shown in figure 1 (right) for the possible frustration angles ϕ corresponding to $\text{Pb}_2\text{VO}(\text{PO}_4)_2$. The maxima are expected at different moduli $|\mathbf{q}|$ of the momentum transfer for NAF and CAF phases. Recent neutron scattering experiments [10] for $\text{Pb}_2\text{VO}(\text{PO}_4)_2$ and $\text{Li}_2\text{VO}(\text{SiO}_4)$ have definitely shown that the $S(|\mathbf{q}|)$ behaviour for these compounds agrees with that of the CAF phase, therefore the positive value $\phi = 0.66\pi$ is the right choice for the Pb and $\phi \simeq 0.45\pi$ for the Li compound (there is a spread of acceptable values [3]). Although nothing certain is known yet for the other vanadates, given their similarities with the above compounds, one would rather speculate that they should

lie in the same CAF sector. This has been assumed in figure 1, where the ϕ values from thermodynamical data have been included for the remaining compounds. This would lead one to conclude that three of the compounds are rather close to the $J_1 = 0$ line where CAF is stabilized by quantum fluctuations while the other three have large negative J_1 , i.e. they have strongly frustrated nn FM bonds, which puts them closer to the disordered sector on the FM ($J_1 < 0$) side. It has also been proposed to resolve the ambiguity of ϕ by studying the nonlinear magnetic susceptibility [11], which should show characteristic asymmetry of peak heights among the CAF/NAF sectors.

4. High-field properties of the J_1 - J_2 -model

This asymmetry might also be suspected for the high-field behaviour of magnetization, specific heat and other quantities. As mentioned above, $J_c \sim 10$ K for the present compounds, which makes the saturation fields accessible. Therefore, we have investigated $C_V(T, H)$ and $M(T, H)$ as a function of the frustration angle using a 20-site cluster with periodic boundary conditions. The most convenient quantity to study is the saturation field $H_{\text{sat}}(\phi)$. The classical expression for this quantity is obtained from the one-magnon instability of the linear spin wave excitations with $\Delta S = 1$ of the fully polarized (FM) state. It is given by

$$g\mu_B H_{\text{sat}}^{(\text{cl})} = zSJ_c[\cos\phi(1 - \frac{1}{2}(\cos Q_x + \cos Q_y)) + \sin\phi(1 - \cos Q_x \cos Q_y)]. \quad (3)$$

In general, a k -magnon instability associated with $\Delta S = k$ magnon excitations and an onset field $H_{\text{sat}}^{(k)}$ may be defined. It corresponds to the crossing point of total energies $E(S_z, H)$ with total spin components $S_z = N/2$ and $S_z = N/2 - k$ respectively. Since the totally polarized state is an exact eigenstate of the system, the classical result $H_{\text{sat}}^{(\text{cl})}$ for $H_{\text{sat}}^{(1)}$ is exact. It is shown in a polar plot in figure 2 (left) as a function of ϕ . The two-magnon instability field $H_{\text{sat}}^{(2)}$ has been computed for the 16-, 20- and 24-site clusters; it is shown as a dashed line for 20 sites. One observes a crossing of the line of one- and two-magnon instabilities at $\phi = 0.5\pi$ ($J_1 = 0$) independent of the cluster size. In the thermodynamic limit such a crossing is possibly expected when ϕ enters the disordered regime on the FM side, which is characterized by a hidden spin nematic order [2]. While the magnitudes of the saturation field are different, the qualitative magnetization behaviours as shown in figure 2 (right) are quite similar for both possible ϕ values of $\text{Pb}_2\text{VO}(\text{PO}_4)_2$. We mention that the plateaux in the $T = 0$ magnetization reported previously [12] cannot be seen in our case, where $T = 0.2J_c/k_B$ is apparently large enough to overcome the associated spin gap energy.

5. Specific heat and magnetocaloric effect

The situation is more promising for the high-field specific heat. The results of the 20-site cluster calculation are shown in figure 3 (left). On the FM side ($\phi = -0.12\pi$) the specific heat is smaller by a factor of two as compared to $\phi = 0.66\pi$ on the NAF side. This large asymmetry in $C_V(H, \phi)$ should be useful to determine the proper frustration angle in the other compounds. In addition, we have considered the magnetocaloric effect, which is the adiabatic temperature change caused by the application of a field. It is given by

$$\left(\frac{\partial T}{\partial H}\right)_S = -\left(\frac{\partial S}{\partial H}\right)_T \bigg/ \left(\frac{\partial S}{\partial T}\right)_H = -\frac{T(H)}{C_V(H, T)} \left(\frac{\partial M(H, T)}{\partial T}\right)_H, \quad (4)$$

where S and M are entropy and magnetization respectively. This may be expressed in terms of thermodynamic ensemble averages of total energy and magnetization, which can be evaluated

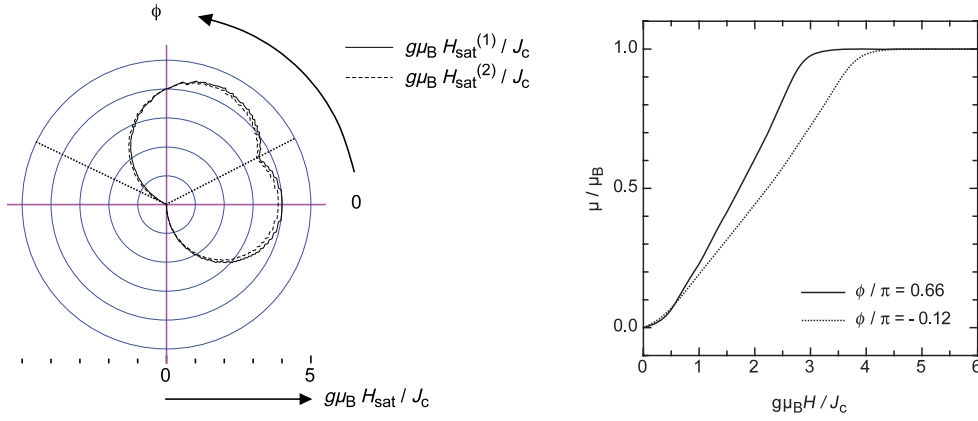


Figure 2. Left: polar plot of saturation field $H_{\text{sat}}^{(1)}$ (full line) and $H_{\text{sat}}^{(2)}$ (broken line) as a function of the frustration angle ϕ . Both lines are calculated for the 20-site cluster. $H_{\text{sat}}^{(1)}$ agrees with the classical expression (3). Right: magnetization for ϕ_{\pm} of $\text{Pb}_2\text{VO}(\text{PO}_4)_2$ at $T = 0.2J_c/k_B$. (This figure is in colour only in the electronic version)

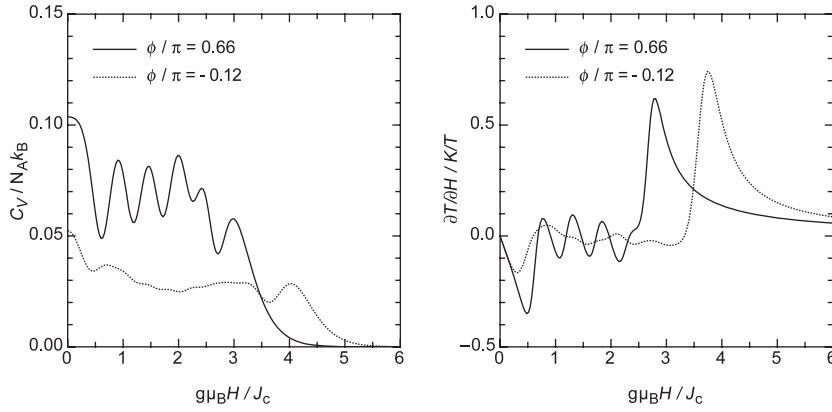


Figure 3. Left: $C_V(H)$ for ϕ -values of $\text{Pb}_2\text{VO}(\text{PO}_4)_2$ at $T = 0.2J_c/k_B$. Right: magnetocaloric coefficient for the same values. Giant peaks at the saturation fields $H_{\text{sat}}^{(1)}(\phi = 0.66\pi) = 2.54$ and $H_{\text{sat}}^{(1)}(\phi = -0.12\pi) = 3.70$ (in natural units) are due to the dispersionless J_1 - J_2 magnon spectrum along symmetry directions. In both cases a 20-site cluster and a temperature $T = 0.2J_c/k_B$ was used. Rapid oscillations are a finite size effect caused by the crossing of discrete levels with different S_z . They are more pronounced for $\phi/\pi = 0.66$ because there only levels with even S_z can cross with the ground state, corresponding to steps with $\Delta S_z = 2$, while for $\phi/\pi = -0.12$ all steps with $\Delta S_z = 1$ are possible, which leads to reduced finite size oscillations.

by the Lanczos method

$$\left(\frac{\partial T}{\partial H}\right)_S = -g\mu_B T \frac{\langle \mathcal{H} S_z \rangle - \langle \mathcal{H} \rangle \langle S_z \rangle}{\langle \mathcal{H}^2 \rangle - \langle \mathcal{H} \rangle^2}. \quad (5)$$

The magnetocaloric effect has become a useful tool to map out high-field phase boundaries and should be applicable for the J_1 - J_2 materials. From geometrically frustrated spin systems like pyrochlore or chequerboard it is known [13] that they have giant $(\partial T / \partial H)_S$ at the saturation field. This is due to the existence of flat magnon branches or equivalently, localized magnons which condense suddenly out of the fully polarized state below the saturation field. In the

J_1 - J_2 model, line zeros in the magnon spectrum should also lead to a (weaker) enhancement of the MCE. We have evaluated (5) for the 20-site cluster at $T = 0.2J_c/k_B$ for ϕ values of $\text{Pb}_2\text{VO}(\text{PO}_4)_2$ shown in figure 3 (right). Pronounced peaks are seen at the saturation fields for both frustration angles. Their peak heights differ slightly. The peak persists for all ϕ and is reduced in the disordered region around $\phi \sim 0.154\pi$. According to (4) the magnetocaloric effect is large when specific heat is small or the entropy changes rapidly with field. Comparison with the left panel of figure 3 shows that in the present case the rapid entropy change close to H_{sat} is the origin of the magnetocaloric peak. This rapid entropy change is associated with the flatness of magnon branches along symmetry directions. The maximum saturation field is obtained for $\phi_{\text{max}}/\pi = \tan^{-1}(2) \sim 0.35$ where $g\mu_B H_{\text{sat}}^{\text{max}} = 4.5J_c$. With $g = 2$ and assuming a typical $J_c \sim 10$ K for the compounds discussed one obtains saturation fields below 33.5 T, which are easily accessible for pulsed fields.

6. Summary

A new class of insulating layered vanadium compounds is an excellent realization of the frustrated J_1 - J_2 Heisenberg model. The latter exhibits various magnetically ordered and disordered phases as a function of the frustration angle ϕ . Thermodynamic measurements are not able to determine ϕ uniquely. We have proposed high-field experiments which show that the saturation field should provide further evidence for the proper ground state. Furthermore, large magnetocaloric effects are predicted just above the moderate and accessible saturation fields of these compounds. The asymmetry of adiabatic temperature change with respect to ϕ may also be helpful in resolving the ambiguity of the frustration ratio.

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