Third-order magnetic susceptibility of the frustrated square-lattice antiferromagnet

Burkhard Schmidt * Peter Thalmeier

Max-Planck-Institut für Chemische Physik fester Stoffe, Dresden, Germany

Abstract

We present results from our analysis of the finite-temperature properties of the spin 1/2 \( J_1 \)-\( J_2 \) Heisenberg model on a square lattice. The analysis is based on the exact diagonalization of small clusters with 16 and 20 sites utilizing the finite-temperature Lanczos method. In particular, we focus on the temperature dependence of the third-order magnetic susceptibility as a method to resolve the ambiguity of exchange constants. We discuss the entire range of the frustration angle \( \phi = \tan^{-1}(J_2/J_1) \) parameterizing the different possible phases of the model, including the large region in the phase diagram with at least one ferromagnetic exchange constant.

Key words: frustrated Heisenberg model, third-order magnetic susceptibility, finite-temperature Lanczos method

The spin-1/2 Heisenberg model on a two-dimensional square lattice with next-nearest neighbour interaction belongs to the most intensely studied models for frustrated spin systems. Its Hamiltonian is of the form

\[
H = J_1 \sum_{(ij)_1} \vec{S}_i \cdot \vec{S}_j + J_2 \sum_{(ik)_2} \vec{S}_i \cdot \vec{S}_k, \tag{1}
\]

where the sum on \((ij)_1\) runs over nearest neighbour and the sum \((ik)_2\) over diagonal next-nearest neighbour bonds. We allow the exchange constants \(J_1\) and \(J_2\) to be negative (FM) as well as positive (AF). Only the relative size of the exchange couplings determines the physics of the model. Therefore it is convenient to introduce an overall energy scale \(J_c = \sqrt{J_1^2 + J_2^2}\) and a frustration angle \(\phi = \tan^{-1}(J_2/J_1)\) to characterize the model. The schematic phase diagram of the model, which is shown in the left part of Fig. 1, can roughly be subdivided into three ordered phases, plus two spin-gapped phases or families of phases. The former are characterised by anomalies in the susceptibility \(\chi(q)\) at the ordering vectors \(q = (0, 0)\) (FM), \(q = (\pi, 0)\), or \((0, \pi)\) (collinear antiferromagnet, CAF), and \(q = (\pi, \pi)\) (Néel-type antiferromagnet, NAF).

Experimental realisations of the frustrated square-lattice antiferromagnet include the quasi-two-dimensional compounds \(\text{Li}_2\text{VO(Si,Ge)O}_4\) and \(\text{Pb}_2\text{VO(PO}_4)_2\) [1]. The latter is believed to have one ferromagnetic exchange constant. In the following, we shall refer to this compound when discussing the experimental situation.

In principle, the average interaction constant \(J_c\) can be determined from the asymptotic behaviour of the heat capacity and the magnetic susceptibility at high temperatures. However, the determination of the correct frustration angle, and therefore the correct values of the exchange constants of the compounds mentioned above has proved difficult. Diffuse neutron scattering would provide a means to determine \(J_1\) and \(J_2\) separately, therefore we have calculated the static spin structure factor \(S(q, T)\) previously [2]. Combining the parameter dependence of the third-order susceptibility presented here with results obtained earlier for the heat capacity and the linear susceptibility, our method provides a way to unambiguously determine the experimental values of \(J_1\) and \(J_2\) from the thermodynamic properties of the compounds.
χ high-temperature asymptotic behaviour of kN tem size. As usual, Table 1 the thermodynamic traces in Eq. 3. To give an exam-
ple, the right part of Fig. 1 shows the temperature dependence of χ'''(T) of a 20-site cluster for two different values ϕ± of the frustration angle. These values are chosen such that they correspond to those determined for Pb2VO(PO4)2 [1, 2]. The frustration angle ϕ = −0.11π (solid line) corresponds to the Néel antiferromagnet, while ϕ = 0.64π (dashed line) describes the collinear phase.

In all non-ferromagnetic phases of the model, the temperature dependence of χ'''(T) has a pronounced maximum at a temperature $T_{\text{max}}$, vanishes at a temperature $T_{\text{min}}$, passes through a tiny minimum at a temperature $T_{\text{max}}$, and eventually approaches the high-temperature $T^{-3}$ dependence. We have followed these characteristic temperatures as a function of the frustration angle. The initial maximum temperature, together with the value of χ''' at that point, are shown for the 16-site (solid dots) and 20-site cluster (open circles) in Fig. 2. The maxima occur at temperatures $T_{\text{max}} \ll J_c$, therefore finite-size effects are large.

The characteristic temperatures, together with the values of χ''' at maximum and minimum, are compiled in Table 1 for ϕ = −0.11π and 0.64π. Also shown is the ratio $\Theta_{\text{CW}}/T_{\text{max}}$ of the Curie-Weiss temperature to the position of the characteristic maximum of the linear susceptibility χ(T) taken from Ref. 2, which is equal for both values $\phi = \phi_\pm$. Together with the results from Ref. 2, our findings should be useful in determining the precise value of $\phi$ for a given $J_1$-$J_2$ compound, as exemplified here for Pb2VO(PO4)2.

| ϕ  | $T_{\text{max}}$ | $T_{\text{min}}$ | $\chi'''_{\text{max}}$ | $\chi'''_{\text{min}}$ | $\Theta_{\text{CW}}$ |
|----|------------------|------------------|------------------------|------------------------|-----------------|
| −0.11 | 0.49             | 1.11             | 1.69                  | 2.37                  | 1.41            |
| 0.64  | 0.49             | 0.21             | 1.45                  | 2.02                  | 0.64            |

References
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