Thermodynamics of Ferro-Antiferro $J_1 – J_2$ Spin-Half Chain Materials

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We investigate finite temperature properties of a one-dimensional S=1/2 spin model expected to describe a family of recently synthesised copper based materials. We obtain results for the specific heat and magnetic susceptibility which may be used to fit experimental data. Additionally, we analyse the static magnetic structure factor and confirm the existence of incommensurate correlations in the strongly frustrated regime.

1. Introduction

There is considerable current interest in a class of materials containing edge-shared CuO_2 units, such as $LiCu_2O_2$, $LiCuVO_4$ and many others [1]. The nature of the exchange pathways is such that, magnetically, these materials can be approximated as weakly coupled chains with ferromagnetic nearest-neighbour exchange J_1 ($J_1 > 0$) and a (usually) stronger frustrating second-neighbour antiferromagnetic exchange J_2 ($J_2 < 0$). This is illustrated in Fig. 1.

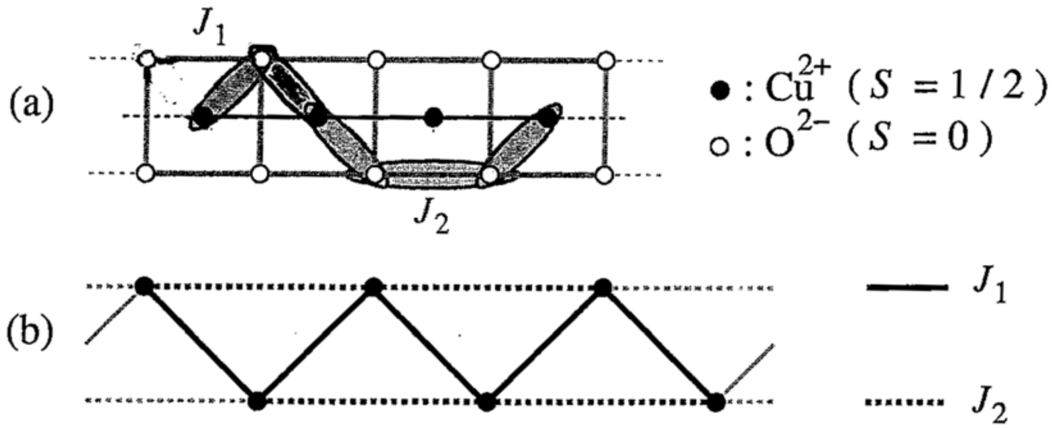

Fig. 1(a) Edge-joined CuO_4 plaquettes, showing nearest-neighbour and next-nearest neighbour
exchange paths. (b) The zig-zag J_1-J_2 chain.

This model, also termed the zig-zag chain, has the spin Hamiltonian

$$H = J_1 \sum_i \mathbf{S}_i . \mathbf{S}_{i+1} + J_2 \sum_i \mathbf{S}_i . \mathbf{S}_{i+2} \qquad (1)$$

It is rigorously known that for $J_2/J_1 > -0.25$ the ground state is fully aligned ferromagnetic. For $J_2/J_1 < -0.25$ the total magnetization is zero but the precise nature of the ground state remains enigmatic. Classically, the ground-state is an incommensurate spiral with successive spins rotated by an angle θ, with $\cos\theta = |J_1/4J_2|$. However, strong quantum fluctuations may invalidate this simple picture. Various predictions include *spin nematic* phases [2] and ferroelectric behaviour [3].

In the present work we use high-temperature series expansions [4] to investigate two aspects of this model. First, we calculate the susceptibility and specific heat of the model as a function of temperature for any choice of the parameters (J_1,J_2). Secondly, we obtain the static magnetic structure factor S(q) and look for the development of a characteristic peak at some wavevector q_0 as the temperature is lowered. This provides information on the nature of the correlations which develop with decreasing temperature and, indirectly, an inference about the nature of the ground state, over the range of the parameter ratio $r = J_1/J_2$.

2. High Temperature Thermodynamics

By standard methods [4] we obtain expressions for the specific heat and the susceptibility in the form (with $K = J_1/k_BT$

$$C/k_B = \sum_{n=0}^{\infty} a_n(r)\, K^n \qquad (2)$$

$$\bar{\chi} = k_B T\chi/(g\mu_B)^2 = \sum_{n=0}^{\infty} c_n\,(r)\, K^n$$

We have computed the coefficients through order K^{12} for various r. Earlier shorter series [5] were used to study the purely antiferromagnetic case, and provide a partial check on our results. Pade approximants are used in evaluating the series. Fig 2 shows typical results for the susceptibility obtained in this way. Specific heat results will be shown in a further more detailed publication.

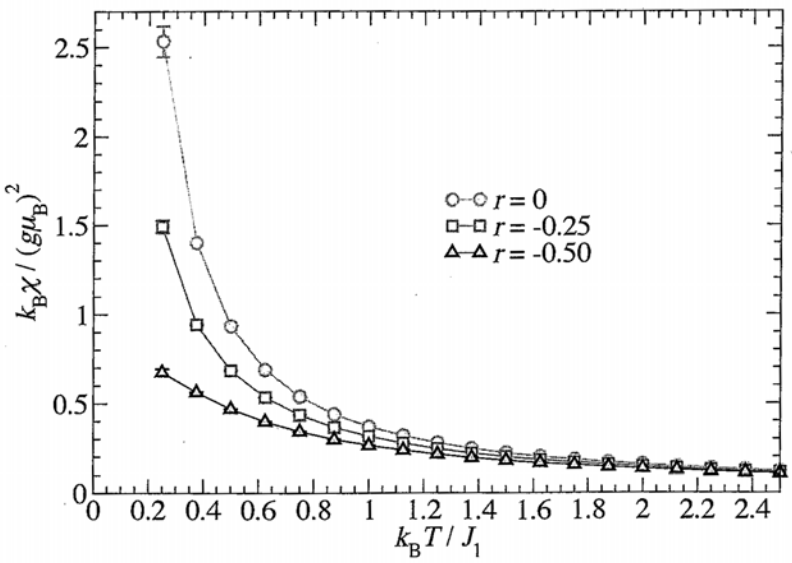

Fig. 2. Susceptibility as a function of temperature for r = 0, -0.25, and –0.50.

3. Static Structure Factor

The static structure factor is given by

$$S(q) = \sum_j \langle S_0^z S_j^z \rangle_T \, e^{iqRj} \qquad (3)$$

Standard methods are used to derive high-temperature expansions for the various correlators which are then combined to give a high-temperature series (to order K^{12}) for the structure factor, for arbitrary wavevector q. These are evaluated again with Pade approximants. Fig. 3 shows curves of S(q) versus q for two values of r. In each case a number of curves are shown, for decreasing temperatures.

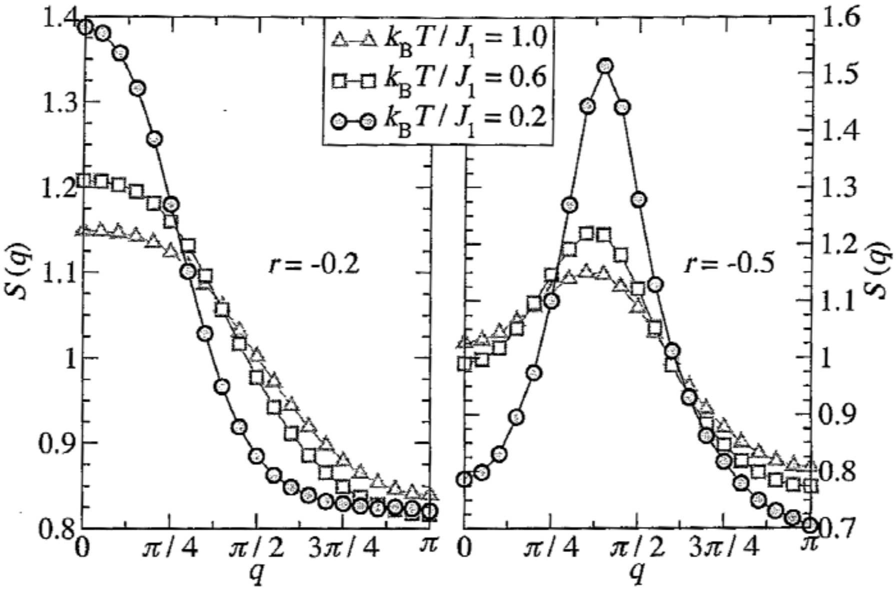

Fig. 3. Static structure factor S(q), defined in Eq.(3), as a function of q and for temperatures indicated in the inset, for two values of $r = J_1/J_2$, -0.2 (left) and –0.5 (right).

As we see in Fig. 3, for r = -0.20 the maximum in S(q) remains at $q_0 = 0$, as expected for the ferromagnetic phase, with the peak increasing in height as the temperature is lowered. At T=0, which of course we cannot reach, S(q) would become a delta function. On the other hand, for r = -0.5, a coupling ratio in the classical spiral phase, S(q) peaks at the incommensurate wavevector $q_0 = 0.42\pi$. We have made similar estimates of q_0 for various r, and in Fig.4 the results are compared to the classical results.

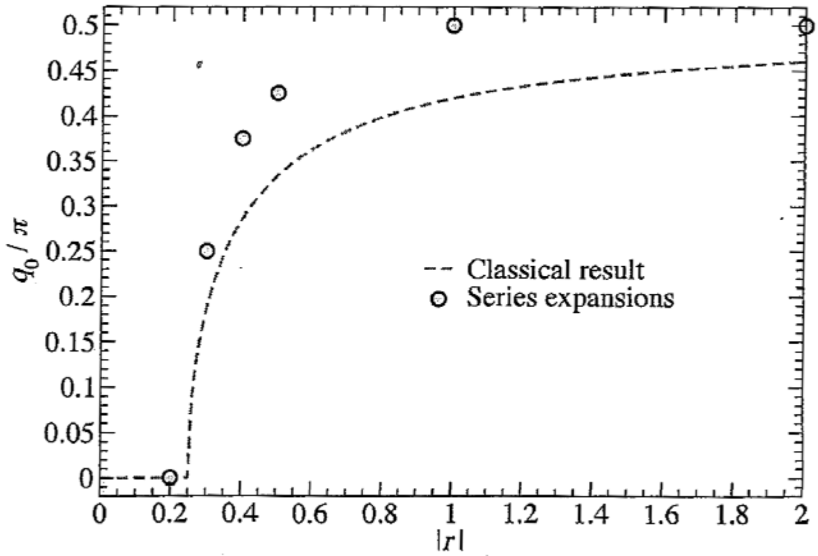

Fig. 4. Location of the incommensurate peak q_0 in S(q) as a function of $r = J_1/J_2$.

4. Conclusions and Future Work

Summarising, we have derived high-temperature expansions to investigate the thermodynamic properties of a one-dimensional magnet with competing ferro- and antiferromagnetic interactions, expected to describe the magnetic properties of a number of recently synthesised materials. Work in progress, which will be reported elsewhere, will fit our theoretical results for the specific heat and susceptibility to experimental data, to test the validity of the model and to estimate values for the exchange parameters. Additionally we have analysed the temperature dependence of the magnetic structure factor for various coupling ratios r, and find evidence for incommensurate ordering in the strongly frustrated regime.

Acknowledgments

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References

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