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Low-energy singlet dynamics of spin-$\frac{1}{2}$ Kagomé Heisenberg antiferromagnets and low-temperature features in the specific heat of Kagomé clusters

A V Syromyatnikov[1] **and S V Maleyev**

Petersburg Nuclear Physics Institute, Gatchina, St Petersburg 188300, Russia

E-mail: syromyat@thd.pnpi.spb.ru

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Abstract
We suggest a new approach for describing the low-energy sector of spin-$\frac{1}{2}$ Kagomé Heisenberg antiferromagnets (KAFs). The Kagomé lattice is represented as a set of blocks (stars) arranged in a triangular lattice. Each of these stars has two degenerate singlet ground states. It is shown using symmetry considerations that the KAF lower singlet band is made by inter-star interaction from these degenerate states. The general form of the effective Hamiltonian describing this band is established. Low-T peculiarities in the specific heat of Kagomé clusters are also discussed.

1. Introduction

The unusual low-T properties of Kagomé antiferromagnets (KAFs) have attracted much attention from both theorists and experimenters in the last decade (see [1–6] and references therein). A qualitative understanding of the spin-$\frac{1}{2}$ KAF low-T physics was based mostly on the results of numerous finite cluster investigations with the number of sites $N \leqslant 36$ (see, e.g. [2–4]). They revealed a gap separating the ground state from the upper triplet levels and a band of nonmagnetic singlet excitations inside it. The number of states in the band increases exponentially with N. Meanwhile the origins of the singlet band as well as the nature of the ground state are still unclear.

Subsequently, specific heat C calculations of Kagomé clusters with even N revealed two peaks, with the low-T one at $T_l < \Delta$ (Δ is the spin gap) [3, 4]. It was obtained for $N = 18$, 36 in [3] that the low-T peak is weakly dependent on the magnetic field. It is now believed that the lower singlet band is responsible for the low-T peak in C and explains its weak field dependence [3, 4].

In section 2 of this paper we give a model describing the low-energy sector of spin-$\frac{1}{2}$ Kagomé Heisenberg antiferromagnets (KAFs). In our recent paper [5] we proposed to consider

[1] Author to whom any correspondence should be addressed.

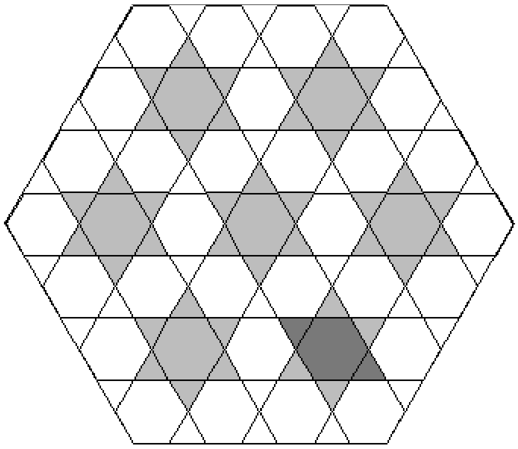

Figure 1. Kagomé lattice. This can be considered as a set of stars with 12 spins arranged in a triangular lattice. The dark region represents a unit cell. There are four unit cells per star.

a Kagomé lattice as a set of stars with 12 spins arranged in a triangular lattice (see figure 1). This representation is caused by the star properties. Numerical diagonalization gives that each star has two degenerate singlet ground states separated from the upper triplet levels by a gap. These states form a lower singlet energy band as a result of inter-star interaction. We show that it is described by the Hamiltonian of a magnet in the external magnetic field where degenerate states of stars are represented in terms of two projections of pseudospin-$\frac{1}{2}$. The general form of the effective Hamiltonian is established. The Hamiltonian parameters are calculated up to the third order of perturbation theory. The ground-state energy calculated in the considered model is lower than that evaluated numerically in the previous finite clusters studies. A way of experimental verification of this picture using neutron scattering is discussed.

In section 3 we reveal the nature of the specific heat features discussed above by the example of the star. Our conclusions contradict the point of view accepted now. There are no singlet states inside the spin gap in the star as there was in clusters with periodic boundary conditions studied so far. Nevertheless we have obtained in [6] the double-peak structure of C with a low-T peak at $T_l \approx \Delta/3$ which possesses weak field sensitivity similar to the results of previous works [3]. A simple model revealing the nature of these peculiarities is proposed. In particular, it is shown that the rapid increase in the density of states (DS) just above the gap is responsible for this peak. The nature of the double-peak structure in other frustrated antiferromagnetic systems can be the same (see [7]). We hope that this paper will stimulate corresponding studies.

2. Singlet dynamics

In this section we study the nature of the lower singlet band in Heisenberg spin-$\frac{1}{2}$ Kagomé antiferromagnets for which the Hamiltonian has the form

$$\mathcal{H}_0 = J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \mathbf{S}_j + J_2 \sum_{(i,j)} \mathbf{S}_i \mathbf{S}_j - H \sum_i S_i^z, \qquad (1)$$

where $\langle i, j \rangle$ and (i, j) denote nearest and next-nearest neighbours on the Kagomé lattice, respectively. The case of $|J_2| \ll J_1$ is considered in this paper. As is shown below, in spite of its smallness the second term in equation (1) can be of importance for the low-energy properties. We start with $J_2 = 0$ and $H = 0$ in equation (1).

Table 1. Low-lying levels of the star. They are classified by S and degenerated with S^z. Here the energies are measured in units of J_1.

	Number of levels				Number of levels		
Energies	$S=0$	$S=1$	$S=2$	Energies	$S=0$	$S=1$	$S=2$
−4.500 000	2	0	0	−4.077 928	0	1	0
−4.240 331	0	1	0	−4.068 850	0	2	0
−4.236 220	0	2	0	−4.056 472	1	0	0
−4.232 400	0	2	0	−4.010 310	0	2	0
−4.202 448	0	1	0	−3.913 465	0	1	0
−4.183 814	1	0	0	−3.865 010	2	0	0
−4.182 320	2	0	0	−3.832 691	0	1	0
−4.141 850	2	0	0	−3.829 460	0	0	1

Table 2. Contributions to the parameters of the effective Hamiltonian equation (2) from terms V^1, V^2 and V^3 of the perturbation expansion. Interaction J_2 has been taken into account in V^1 and V^2 terms only. Here $\mathcal{N}$ is the number of stars in the lattice.

	V^1	V^2	V^3	Totals
$\mathcal{J}_z$	0	$-0.007J_1+0.002J_2$	$-0.003J_1$	$-0.010J_1+0.002J_2$
$\mathcal{J}_y$	0	$-0.001J_1+0.007J_2$	0.0	$-0.001J_1+0.007J_2$
$\mathcal{J}_x$	0	0	$0.067J_1$	$0.067J_1$
h	$-0.563J_2$	$-0.092J_1-0.218J_2$	$-0.081J_1$	$-0.173J_1-0.781J_2$
$\Delta\mathcal{C}$[a]	$-0.009J_2\mathcal{N}$	$-0.768J_1\mathcal{N}+1.530J_2\mathcal{N}$	$-0.057J_1\mathcal{N}$	$-0.825J_1\mathcal{N}+1.521J_2\mathcal{N}$

[a] Correction to the value $\mathcal{C}_0=-4.5J_1\mathcal{N}$ for non-interactive stars.

It is proposed to consider a Kagomé lattice as a set of stars with 12 spins arranged in a triangular lattice (see figure 1). The star Hamiltonian was diagonalized numerically. Some low-lying states are presented in table 1. As is seen, the star has two degenerate singlet ground states separated from the upper triplet levels by a gap $\Delta\approx 0.26J_1$ [5]. Evidently these states give rise to a singlet energy band as a result of inter-star interaction. As was discussed in [5], the two ground-state wavefunctions Ψ_+ and Ψ_- have different symmetry. It can be proved using this fact that this singlet band determines the KAF lower singlet sector [5]. Formal conditions for perturbation theory applicability are fulfilled for inter-star interaction when one studies the lower singlet band [5]. So despite its not being small compared to intra-star interaction, the inter-star one is treated as a perturbation in the following consideration.

Representing degenerate states of stars in terms of projections of pseudospin-$\frac{1}{2}$ ($|\uparrow\rangle=\Psi_-$ and $|\downarrow\rangle=\Psi_+$), one can show that this band is described by an effective Hamiltonian, the general form of which established using symmetry considerations is [5]

$$\mathcal{H}=\sum_{\langle i,j\rangle}[\mathcal{J}_z s_i^z s_j^z+\mathcal{J}_x s_i^x s_j^x+\mathcal{J}_y s_i^y s_j^y]+h\sum_i s_i^z+\mathcal{C}, \tag{2}$$

where $\mathbf{s}$ is the operator of pseudospin-$\frac{1}{2}$, and $\langle i,j\rangle$ now labels nearest-neighbour pseudospins on the triangular lattice formed by stars. This Hamiltonian describes the low-energy singlet dynamics in KAF. The parameters of the Hamiltonian equation (2) calculated up to the third order of perturbation theory are summarized in table 2. As is seen, V^3 corrections to the parameters are still large and the analysis of the perturbation series cannot be finished at this point. Moreover, as is discussed in detail in [5], V^3 corrections to the initial lower singlet levels of stars are only two times smaller than V^2 ones. So the perturbation theory works quite badly and further studies are required to determine to what extent our approach is applicable to the Kagomé problem.

As is clear from table 2, $\mathcal{J}_x$ and h are the largest parameters of the Hamiltonian equation (2) in our approximation. So the KAF behaves like an Ising antiferromagnet in a perpendicular magnetic field. In this case the classical value of the field at which spin-flip occurs is $h_{\text{s-f}} = \mathcal{J}_x$, which is approximately 2.6 times smaller than h. So the ground state is ordered with all the stars in Ψ_- state.

The ground-state energy and that of the upper edge of the singlet band calculated using table 2 are $(-4.5J_1+\Delta\mathcal{C}+h/2+3\mathcal{J}_z/4)\mathcal{N} = -0.452J_1N$ and $(-4.5J_1+\Delta\mathcal{C}-h/2+3\mathcal{J}_z/4)\mathcal{N} = -0.437J_1N$, respectively [5]. At the same time the ground-state energy of the largest cluster (with $N = 36$) considered before numerically was $-0.438J_1N$ [2]. So we believe that the clusters used in previous studies were too small to reflect the low-energy sector at $J_2 = 0$ properly.

Corrections to the parameters of the effective Hamiltonian equation (2) from J_2-interaction are also presented in table 2. As is seen, the J_2-contribution to the value of h becomes significant if $|J_2| \sim 0.1J_1$. So despite its smallness, the next-nearest neighbour interaction can play an important role for KAF low-energy properties.

The physical picture described here can be verified experimentally using inelastic neutron scattering: the corresponding intensity for singlet–triplet transitions should have periodicity in the reciprocal space corresponding to the star lattice.

Finally we have shown numerically that the low-energy physics described above for spin-$\frac{1}{2}$ KAFs is not valid for KAFs with larger spin values [5].

3. Specific heat features

We turn now to the discussion of the double-peak C structure in Kagomé clusters using the example of the star. It is assumed in this section that $J_2 = 0$ and $J_1 = 1$ in equation (1). We start with $H = 0$ in equation (1). A distinguishing characteristic of the star spectrum in the range $\Delta - 4.5 < E < 4.5$ is that levels are very close to each other: the distances between them are of the order of 0.1–0.01 $\ll \Delta$. A part of the spectrum shown in table 1 reflects this feature. We show below that this peculiarity plays the crucial role for low-T star properties.

The specific heat C calculated with this spectrum is shown in figure 2. The most intriguing feature of C is the existence of the low-T peak at $T_l \approx \Delta/3$. The problem is that according to the common point of view the specific heat of a fully gapped system should decrease exponentially with T in the range $T < \Delta$. Meanwhile, exponential decay in the star occurs only at $T < T_l$. We show now that the extremely high DS just above the gap is the origin of this surprising behaviour.

It can be shown that the DS of the star above the gap can be modelled by a constant $w \approx 180$ in calculations of C at $T < \Delta$ [6]. As a result we have for C in this case

$$C = \frac{1 + Zx\mathrm{e}^x(x^2 + 2x + 2)}{(1 + Zx\mathrm{e}^x)^2}, \tag{3}$$

where $x = \Delta/T$ and $Z = 2/(w\Delta)$. This expression reproduces the low-T peak nicely (see figure 2). It can be shown that the peak disappears at $Z < 2$ ($Z \approx 0.04$ for the star). Hence the rapid increase in the DS just above the gap is responsible for the low-T peak.

The model discussed allows also to understand the nature of the weak H sensitivity of the low-T peak. As a result of C calculations using the spectrum of the Hamiltonian equation (1) obtained numerically, we found that the peak height is reduced only 11% and T_l is diminished slightly at $H = T_l$ (see figure 3). Using the fact that the low-energy sector above the gap is represented mostly by triplets and that the field splits levels with $S \neq 0$, the DS can be modelled at $H < \Delta$ in the following way: it is 0 at $0 < E < \Delta - H$, the DS is $W/3$ at $\Delta - H < E < \Delta$, it is $2W/3$ at $\Delta < E < \Delta + H$ and the DS remains W at $E > \Delta + H$.

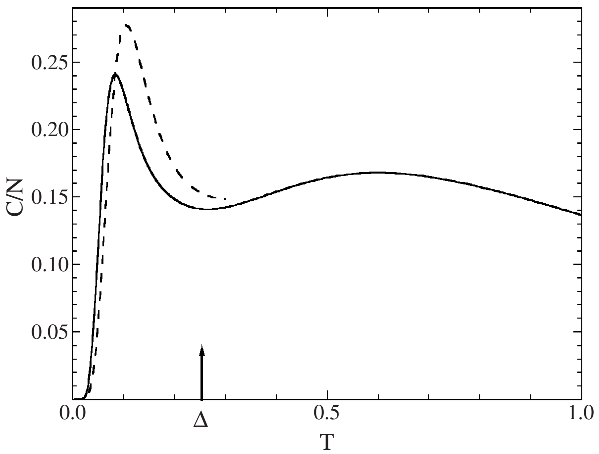

Figure 2. The specific heat $C(T)$ per spin of the star (solid curve). The point $T = \Delta$ is marked by the arrow. The dashed curve is the specific heat calculated with equation (3).

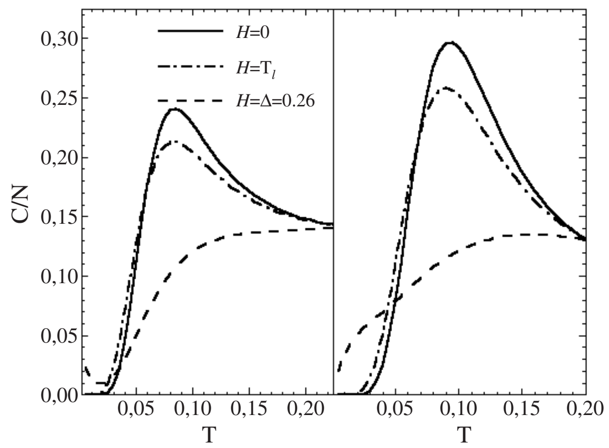

Figure 3. Low-T peak evolution for the star in the magnetic field H obtained by numerical diagonalization of the Hamiltonian equation (1) (left) and by the model discussed in the text (right). In both cases $T_l \approx \Delta/3$.

Results of the peak evolution with H in this approach even reproduce quantitatively the main tendencies in low-T peak evolution with H (see figure 3). The field smooths the area where the drop in DS takes place and in doing so it causes the peak reduction. *For changes in the peak to be significant, $H \approx \Delta$ is needed. So the reason for the weak field sensitivity of the low-Tpeak in the star is that $T_l \approx \Delta/3$.* As is shown in [6], the reason for the low-T peak appearance and the origin of its weak field sensitivity in larger clusters are the same as they are in the star.

4. Conclusion

In conclusion we present a model for the low-energy physics of spin-$\frac{1}{2}$ Kagomé Heisenberg antiferromagnets (KAFs). The spin–lattice is represented as a set of stars (see figure 1). Each star has two degenerate singlet ground states which are described in terms of pseudospin-

$\frac{1}{2}$. It is shown that interaction between stars leads to a band of singlet excitations which determines the KAF low-energy properties. The low-energy singlet dynamics is described by the effective Hamiltonian equation (2) whose parameters calculated using perturbation theory are summarized in table 2. The ground-state energy is lower than that calculated in previous finite cluster studies.

The nature of the double-peak specific heat structure in Kagomé clusters is also studied by the example of the star. A simple model is proposed revealing the low-T peak nature. We show, in particular, that the rapid increase in the density of states just above the spin gap gives rise to this peak. Our consideration could be appropriate for other frustrated antiferromagnetic systems too.

Acknowledgments

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