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Double-peak specific heat feature in frustrated antiferromagnetic clusters

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We study the nature of a double-peak specific heat structure in kagome clusters. A cluster containing 12 spins is considered thoroughly by means of numerical diagonalization. On base of analysis of exact spectra simple models are proposed to explain the nature of the low-T peak at $T_l < \Delta$ (Δ is the spin gap) in this case and in those of larger clusters studied so far. Using these models we show that the rapid increase in density of states just above the spin gap gives rise to the peak, and clarify the weak magnetic field sensitivity of the peak. Spin susceptibility and entropy are considered as well. Our approach could be appropriate for other frustrated antiferromagnetic systems.

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Introduction. A lot of both experimental and theoretical attention has been attracted to kagome and pyrochlore frustrated magnets during last decade (see [1 – 14]. The specific heat C measurements in spin-$\frac{3}{2}$ kagome compound SrCrGaO revealed a peak at $T \approx 5\,$K which was almost independent on the magnetic field H up to 12 T, and $C \propto T^2$ behaviour at lower T [1]. Furthermore, the uniform susceptibility χ was found to deviate from Curie-Weis law at much smaller temperatures than in non-frustrated materials.

In order to understand qualitatively peculiarities of these systems a number of finite clusters diagonalization studies have been carried out [2, 4 – 12]. The main finding of Refs. [4 – 8] on kagome Heisenberg spin-$\frac{1}{2}$ clusters (the number of sites $N \leq 36$) with periodic boundary conditions is the observation of a spin gap Δ separating the singlet ground state from the upper triplet levels, a band of non-magnetic singlet excitations being inside the gap. The number of states in the singlet band increases exponentially with the number of sites. Similar picture with many singlets inside the spin gap has been found for pyrochlore clusters [2].

Specific heat calculations of kagome clusters with $N = 12$, 18, 24 and 36 revealed two peaks with the low-T one at temperature $T_l \lesssim \Delta$ [8 – 11]. At $N = 18$, 36 the low-T peak was found to be weakly H-dependent, and $C \propto T^2$ in a narrow interval below T_l [11]. It is widely accepted now that the wealth of low-lying singlet excitations is responsible for the low-T peak [10, 11] and for its weak field dependence [1, 11]. At the same time it has been pointed out that upper triplet levels contribute to the peak as well [11, 9].

The double-peak specific heat structure is a general feature of many frustrated antiferromagnetic systems. It was observed numerically in Heisenberg spin-$\frac{1}{2}$ pyrochlore slab [12], in Δ-chain [15] and in clusters of triangular lattice within the model with multi-spins exchange [16, 17].

In the present paper we elaborate on nature of the double-peak specific heat structure in Heisenberg spin-$\frac{1}{2}$ kagome antiferromagnetic clusters.

Firstly, we perform numerical diagonalization of the kagome Heisenberg spin-$\frac{1}{2}$ star-like cluster (see inset of Fig.1). The star has doubly degenerate singlet ground

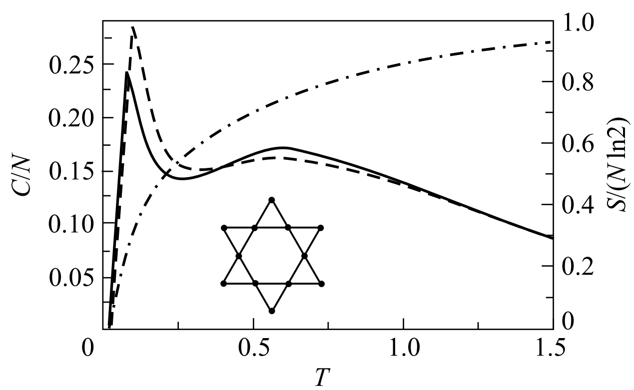

Fig.1. Specific heat $C(T)$ per spin (solid line) and entropy $\mathcal{S}(T)$ normalized by $\mathcal{S}(\infty) = N \ln 2$ (dashed dotted line) of the antiferromagnetic cluster with $N = 12$ shown below the curves (star). The specific heat calculated with eq. (3) is shown by dashed line

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state separated from the lower triplet level by the gap Δ [14]. As is shown in [14] the lower singlet band of kagome antiferromagnet appears naturally from the stars degenerate ground states as a result of inter-star interaction (cf. [4–8]).

Unlike clusters with periodic boundary conditions [4–11] no singlet states appear inside the spin gap in the star cluster. Nevertheless, we observe double-peak structure of C with the low-T peak at $T_l \ll \Delta$ which possesses weak field sensitivity.

Secondly, on base of analysis of exact spectra obtained numerically we propose simple models revealing the low-T peak for both star and other larger kagome antiferromagnetic clusters. All these models share the common feature of rapid increase in density of states (DS) just above the gap. We argue that this feature is responsible for appearance of the low-T peak. The weak magnetic field sensitivity of the peak can be understood within the framework of these models as well.

It should be stressed that our conclusions concerning the origin of the low-T peak contradict to those proposed in previous works [10, 11].

We also investigate how such a behaviour of DS affects entropy and spin susceptibility of the system.

We believe that the double-peak structure in other frustrated antiferromagnetic systems can have the same origin as the one proposed in this paper. We hope our analysis will stimulate corresponding studies.

Analysis of the star. We begin with consideration of the Heisenberg spin-$\frac{1}{2}$ antiferromagnetic cluster shown in the inset of Fig.1. The Hamiltonian has the form

$$\mathcal{H} = \sum_{\langle i,j \rangle} \mathbf{S}_i \mathbf{S}_j - H \sum_i S_i^z, \tag{1}$$

where $\langle i,j \rangle$ denote nearest neighbours and the value of exchange constant is chosen to be unity so the temperature T and the magnetic field H are measured in coupling constant units. We start with $H = 0$ in eq. (1). As the Hamiltonian commutes with all projections of the total spin operator, the star levels are classified by the values of S, irreducible representations (IRs) of its symmetry group C_{6v}, and are degenerated with respect to S^z. In the basis of IRs the matrix of the Hamiltonian has a block structure. Each block has been diagonalized numerically. Low-lying levels are presented in the Table.

As is seen from the Table the star has doubly degenerate singlet ground state separated from the lower triplet level by the spin gap $\Delta \approx 0.26$ [14]. Energies of the ground state and the upper level are -4.5 and 4.5, respectively. Importantly, within the range $-4.5 + \Delta < E < 4.5$ the levels are very close to each other: distances between them are of order of $0.1 \div 0.01 \ll \Delta$. A part of

Low-lying levels of the star. They are classified by S

Energies	Number of levels		
	$S = 0$	$S = 1$	$S = 2$
−4.500000	2	0	0
−4.240331	0	1	0
−4.236220	0	2	0
−4.232400	0	2	0
−4.202448	0	1	0
−4.183814	1	0	0
−4.182320	2	0	0
−4.141850	2	0	0
−4.077928	0	1	0
−4.068850	0	2	0
−4.056472	1	0	0
−4.010310	0	2	0
−3.913465	0	1	0
−3.865010	2	0	0
−3.832691	0	1	0
−3.829460	0	0	1

the spectrum shown in the Table reflects this feature. As is demonstrated below this peculiarity plays the crucial role for the low-T properties of star cluster.

Specific heat C calculated numerically with this spectrum is shown in Fig.1. It has a double-peak structure with the low-T peak at $T_l \approx 0.085$ and high-T one at $T_h \approx 0.6$. The most intriguing feature of C is the fact that $T_l \ll \Delta$. Indeed, according to the common point of view specific heat of a fully gaped system should decrease exponentially with T at $T < \Delta$. Meanwhile the exponential decay for the star-cluster occurs at $T < T_l \approx \Delta/3$ only. As is demonstrated below, the low-T peak is not associated with any peak in DS. Instead, the extremely high DS just above the gap is the origin of this behaviour.

A plot of integrated density of states (IDS) $\mathcal{N}(E)$ (the number of states with energies lower than E) is presented in Fig.2. It is well fitted by the function $\mathcal{N}(E) = 2200 \tanh(0.39E) + 2060$. Since distances between levels at $-4.5 + \Delta < E < 4.5$ are of the order of $0.1 \div 0.01$ one can use this function as IDS above the gap to model the specific heat at $T \gg 0.01$. Then the DS given by $\rho(E) = d\mathcal{N}(E)/dE$ can be approximated within the interval $-4.5 + \Delta < E < 4.5$ as follows:

$$\rho(E) = \frac{w}{\cosh^2(gE)}, \tag{2}$$

with $w = 858$ and $g = 0.39$. In this model C has the form

$$C = \frac{d\overline{E}}{dT} = \frac{d}{dT}\left(\frac{\int_\Delta^9 dE e^{-E/T} E \rho(E-4.5)}{d + \int_\Delta^9 dE e^{-E/T} \rho(E-4.5)} \right), \tag{3}$$

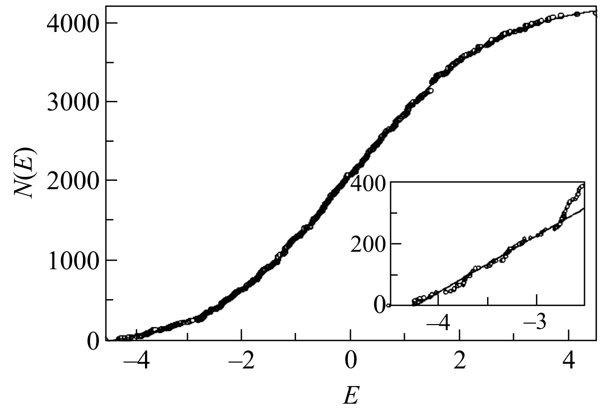

Fig.2. Integrated density of states of the star $\mathcal{N}(E)$ which is the number of states with energies lower than E. The fit is by $2200\tanh(0.39E) + 2060$. The inset shows the low-energy sector where $\mathcal{N}(E)$ is fitted by $180(E + 4.24)$

where $\rho(E)$ is given by eq. (2) and d is the degeneracy of the ground state ($d = 2$ for the star cluster). For the sake of convenience in eq. (3) and below we shifted the energy scale so the ground state energy is equal to 0. Specific heat calculated from eqs. (2) and (3) by numerical integration is also presented in Fig.1. It has a double-peak structure and coincides well with that obtained with the real spectrum. Let us mention an excellent agreement between real- and model-spectrum calculations at high temperatures and the good one around the low-T peak. The latter appearing at $T_l \approx 0.1$ is 15% higher for the model spectrum than for the real one. This pretty good coincidence encourages us to use our model in further analysis.

Low-T peak. Let us consider in detail the specific heat at $T \lesssim \Delta$. We derive now an analytic expression for C on the assumption that $1/\Delta \gg g$ which is held for the star ($1/\Delta \approx 4$ and $g \approx 0.4$). It is easy to show using eq. (2) that under these conditions one can replace $\rho(E - 4.5)$ in eq. (3) by constant $W \approx \rho(\Delta - 4.5) \approx 117$. Unfortunately, at low energies eq. (2) is not very good. As is shown in Fig.2, the precise value of W is 180 because the real $\mathcal{N}(E)$ is well fitted by $180(E + 4.24)$ in the range $\Delta - 4.5 < E < -2.7$. As a result, at $T \lesssim \Delta$ we have from eq. (3):

$$C = \frac{1 + Zxe^x(x^2 + 2x + 2)}{(1 + Zxe^x)^2}, \qquad (4)$$

where $x = \Delta/T$ and $Z = d/(W\Delta)$. Position of the peak T_l is determined by the condition $dC/dx = 0$ which has the form

$$Ze^x = \frac{x + 4}{x^2 + 2}. \qquad (5)$$

As follows from eq. (5) the peak appears if $Z < Z_c \approx 2$. For the star $Z \approx 0.04$ and numerical solution of eq. (5) gives $T_l \approx 0.093$ which is very close to its real value 0.085. Analysis of eqs. (4) and (5) shows that the peak becomes smaller and T_l tends to zero as Δ decreases. At the same time T_l becomes larger as W decreases. It can even exceed the value of Δ when W is small enough. Anyway, the peak disappears as soon as Z amounts to Z_c. Therefore, we conclude that Δ and W play the role of driving parameters responsible for the peculiar low-T behaviour of the star.

Field dependence. We turn now to the discussion how the external magnetic field H affects the low-T behaviour of the specific heat C. In isotropic unfrustrated antiferromagnet a finite-T peak in C implies the transition to the long-range ordered state. This peak could be shifted to zero and suppressed by applying external magnetic field of order of the peak temperature. In this respect the field dependence of the star cluster at low T is also unusual. The results of our calculations of C using the exact spectrum of the Hamiltonian eq. (1) obtained numerically are presented in the left panel of Fig.3. At $H = T_l \approx 0.085$ the peak height is reduced by 11% only and T_l is diminished negligibly. The field

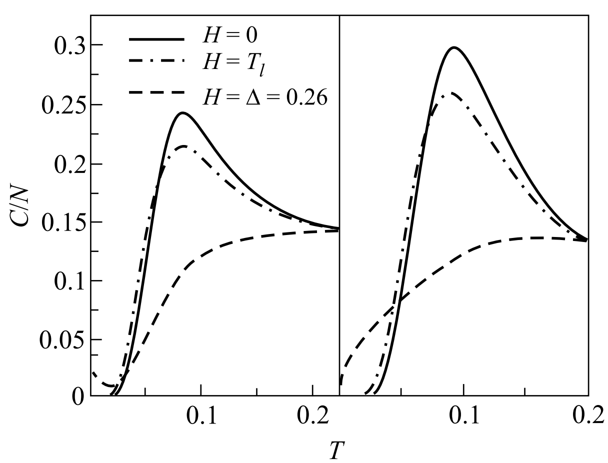

Fig.3. Low-T peak evolution for the star in the magnetic field H obtained by numerical diagonalization of the Hamiltonian eq. (1) (left) and by the model discussed in the text (right). In both cases $T_l \approx \Delta/3$

equal to the value of the spin gap is needed to smear out the peak.

The low-T peak evolution at $H = 0$ can be understood within the scheme described above. We apply the same scheme to explain the nature of the weak H sensitivity of the peak. The magnetic field splits levels with $S \neq 0$. It is clear from the Table that the low-energy sector above the gap is represented mostly by triplets.

So the DS at $H<\Delta$ can be modelled as follows: 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 is W at $E>\Delta+H$. The corresponding results of the peak evolution with H are presented in the right panel of Fig.3. Surprisingly, this model reproduces even quantitatively the main tendencies of the evolution (cf. left panel of Fig.3). The field broadens the area where the drop in DS takes place and in doing so it causes the peak reduction. *For the change in the peak to be significant $H\approx\Delta$ is needed. Therefore the reason for the weak field sensitivity of the low-T peak in the star cluster is $T_l\approx\Delta/3$.*

Larger kagome clusters. As was mentioned above the double-peak specific heat structure in kagome clusters with even N has been found in many previous numerical works [5, 8 – 11]. The height and the position of the high-T peak obtained by high-temperature expansion [10] and by finite cluster diagonalization [8 – 11] coincide with each other and with our results. The low-T peak at $T_l\lesssim\Delta$ was obtained in [8, 9, 10, 11] but its position and height were depended on N, cluster form and calculation technique. We show now that the reason for the low-T peak appearance in those cases is similar: it is the rapid increase in DS just above the spin gap. It will be argued that the weak field sensitivity of the low-T peak reported in [11] has the same origin as in our case.

We focus here on the largest cluster with $N=36$ studied before. Its low-energy spectrum has been obtained in [7, 8] and thermodynamic properties have been discussed in [11]. Similarly to the star case the spectrum above Δ is nearly continuous [7]. In contrast the spin gap there is filled with singlets separated from the non-degenerate singlet ground state by a very small gap $\Delta_s\ll\Delta$. According to [7] these singlets distribute quite uniformly and DS is of the order of 1000. The DS above the gap is much larger than that inside it.

The nature of the low-T peak can be understood qualitatively within the model similar to that proposed above for the star. Let us assume that the DS is W' at $E<\Delta'$, where $\Delta'\gtrsim\Delta$, and there is a jump in the DS at $E=\Delta'$ so it is equal to $W\gg W'$ above Δ'. Specific heat calculation based on analog of eq. (3) gives on the interval of interest $\Delta_s\ll T\lesssim\Delta'$ the following expression:

$$C=\frac{(1+re^x)^2+rx^2e^x+2rZxe^{2x}+Zxe^x(x^2+2x+2)}{(1+re^x+Zxe^x)^2}, \tag{6}$$

where now $x=\Delta'/T$, $Z=1/(\Delta'W)$, and $r=W'/W$. We have made an effort to reproduce the low-T peak in C/N obtained in [11] with the height of approximately 0.152 and $T_l\approx0.05<\Delta\approx0.074$. Specific heat calculated with the use of eq. (6) at $\Delta'=0.23$, $W=43000$, and $W'=1000$ reveals a peak with these parameters. However, the approach is too rough to get T^2 behaviour of C at $T<T_l$. Since $Z\Delta'/T_l\ll r$ the terms in eq. (6) containing Z do not affect the low-T peak. As a result, T_l is given by the equation

$$re^x=\frac{1}{x-1}. \tag{7}$$

It is seen from eqs. (6) and (7) that the parameter Δ' does not affect the peak height but it determines T_l. Evidently, T_l decreases as Δ' tends to zero. Simultaneously, the parameter r determines both the peak position and its height: the peak becomes smaller and T_l increases as r becomes larger. We conclude that the driving parameters for this model are Δ' and r. Furthermore the states lying both above and below Δ' contribute to the low-T peak. This finding is in accordance with Ref. [11].

One can show that no other sets of parameters exist in this model which could describe the peak discussed in [11]. Indeed, since according to [7], DS below Δ' is equal to 1000 and $W'\ll W$ the values of Δ' and W are determined unambiguously by the low-T peak obtained in [11].

Using this model it is easy to understand qualitatively the weak sensitivity of C to the magnetic field discussed in [11]. It has been demonstrated that at $H=T_l\approx0.05$ the peak height is decreased by 10% only and T_l is also decreased negligibly. It resembles the star case considered above. As magnetic field splits levels with $S\neq0$ the DS in the field $H<\Delta'$ will be as follows: the DS at $0<E<\Delta'-H$ remains W', it is $W'+W/3$ at $\Delta'-H<E<\Delta'$, the DS is $2W/3$ at $\Delta'<E<\Delta'+H$ and the DS is W above $\Delta'+H$. We also assume here that the spectrum above Δ' at $H=0$ is formed mostly by triplets. As a result of calculations of the specific heat in our model we obtain that the field $H=T_l\approx0.05$ reduces the low-T peak by 8% and slightly diminishes T_l. These findings are even in *quantitative* agreement with Ref. [11]. Furthermore, the reduction of the peak at $H=\Delta'/2$ calculated in our model is about 28%. So we see that the field as large as $H\sim\Delta'\gg T_l$ is needed to change the low-T peak significantly.

We turn now to brief discussion of entropy $\mathcal{S}(T)$ and spin susceptibility (SS) $\chi(T)$ of kagome clusters and sketch what peculiarities in their behaviour the considered feature of the spectrum leads to.

Entropy. Entropy of the star cluster is presented in Fig.1. Its temperature dependence reproduces the most characteristic feature of those of larger clusters: in all cases there is 50% of the total entropy in the low-T peak of the specific heat at $T < 0.2$ [11, 10]. It is seen from the above consideration that this pecularity in kagome clusters stems from the rapid increase in DS and states above Δ' give the main contribution to $\mathcal{S}$ at $T < 0.2$. Such an entropy feature has been obtained in other models of frustrated antiferromagnets mentioned above which posessed the double-peak structure of C [16, 17].

Spin susceptibility. SS of clusters with $N = 12$ and 18 studied in [10] has one maximum at quite low temperature $T_\chi \approx 0.14 \approx \Delta/2$ and weakly depends on N. SS of the star cluster deviates slightly from those reported in Ref. [10]. As was obtained in [13], $\chi(T)$ of cluster with $N = 36$ differs significantly from $\chi(T)$ of smaller ones ($T_\chi \approx 0.045$ and the peak 50% higher). Authors of [13] attributed this effect to the increase in DS above the gap. One can lead to the same conclusion modelling $\chi(T)$ as we did above for the specific heat. Within the same approximations we get the following expressions for SS of the star χ_s and of the cluster with $N = 36$ χ_{36}:

$$\chi_s = \frac{2}{3}\frac{1}{\Delta}\frac{x}{Zxe^x + 1} \quad \chi_{36} = \frac{2}{3}\frac{1}{\Delta'}x\frac{re^{x(1-\alpha)} + 1}{re^x + 1}, \qquad (8)$$

where $\alpha = \Delta/\Delta'$. Our analysis shows that χ_s reproduces the peak nicely, whereas χ_{36} calculated with $\Delta = 0.074$ gives a peak of the right height but with $T_\chi \approx 0.065$. In both cases the models demonstrate stronger decrease of SS at $T \gtrsim T_\chi$ than in real-spectrum calculations. Meanwhile they correctly pick up the interrelation between the peak characteristics and the DS properties just above the gap mentioned in [13]: the peak becomes higher and T_χ becomes smaller as Z and r decreas.

Conclusion. In this paper we study the nature of the double-peak structure of the specific heat in kagome clusters. The star-like cluster containing 12 spins (see inset of Fig.1) is considered thoroughly by numerical diagonalization. Simple models are proposed to explain properties of the low-T peak at $T_l < \Delta$ (Δ is the spin gap) in this case and in those of larger clusters studied so far numerically [8 – 11]. We show that the rapid increase in density of states just above the gap gives rise to the peak and explain its weak sensitivity to the magnetic field. Our consideration could be appropriate for other frustrated antiferromagnetic systems.

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