Magnetism of the $N=42$ kagomé lattice antiferromagnet

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I. INTRODUCTION

The spin-1/2 kagomé Heisenberg antiferromagnet (KHAF) is one of the most prominent and at the same time challenging spin models in the field of frustrated quantum magnetism. The first challenge concerns the nature of the ground state (GS), on which a plethora of studies exist, see, e.g., Refs. 1–27. Although consensus on the absence of magnetic long-range order (LRO) is achieved, the precise nature of the spin-liquid GS, with quantum spin liquids and Dirac spin liquids as possible candidates,23,28 is not yet understood. Large-scale density matrix renormalization group (DMRG) and exact diagonalization (ED) studies13–15,22,23 suggest a tiny singlet-singlet gap $\Delta_s \sim (0.01 \ldots 0.05)J$, where $J$ denotes the exchange coupling in the Heisenberg model, and a sizeable singlet-triplet gap $\Delta_t \sim (0.13 \ldots 0.17)J$. However, a very recent DMRG study using adiabatic flux insertion provides indications for a much smaller spin-gap in agreement with variational and other numerical techniques.12,17,24,26 The very existence of a gap is deterministic for thermodynamics at low temperatures $T$. In addition, a triplet gap leads to an exponentially activated low-temperature behavior of the susceptibility. On the other hand, indications were found that a huge number of singlet states below the first triplet state may exist,2,3,7,13,14,16,22,28 being relevant for the specific heat $C$ at low temperatures.

Besides the theoretical work there is also large activity on the experimental side, see, e.g., Refs. 30–46 and in particular the review 28. Among the spin-1/2 kagomé compounds, Herbertsmithite ZnCu$_2$(OH)$_6$Cl$_2$ seems to be a promising candidate for a spin liquid.28,35–39,41,46,47

The second challenge concerns the thermodynamic properties of the quantum KHAF on which far less studies exist.4,5,26,29,48–59 While systematic high-temperature approaches48,53,54,56 provide reliable insight into the temperature dependence of physical quantities down to temperatures $T$ of about 40% of the exchange coupling $J$, a reliable picture of the temperature dependences at $0 \leq T \lesssim 0.4J$ is still missing. Various methods48–51,53,56,57 provide indications for an additional low-temperature peak of the specific heat signaling an extra low-energy scale set by low-lying singlets. However, instead of a true maximum a shoulder-like hump may characterize the low-$T$ profile of $C(T)$.26,51 Thus, the low-$T$ behavior of the specific heat is another issue (in some relation to the gaps) that is not yet settled.

The third challenge is given by the magnetization process of the spin-1/2 KHAF,26,27,60–68 A series of magnetization plateaus at $3/9 (= 1/3)$, 5/9 and 7/9 of the saturation is found,26,65,66 among which the 1/3-plateau, already found by Hida,60 is the widest. In addition to the above mentioned plateaus, there might be a tiny plateau at 1/9.26,65 The magnetic ordering within the plateau is well-described by valence-bond states, i.e., the plateau states are of quantum nature,63,65,66 Moreover, there is a macroscopic jump to saturation related to the existence of a huge manifold of localized multi-magnon states.61,69–71 At low enough temperatures and for specific values of the magnetic field the plateaus as well as the magnetization jump are well expressed features of the magnetization curve. From the experimental point of view the detection of these features at low temperatures provides smoking gun evidence of the proximity of the investigated magnetic kagomé compound to the ideal KHAF model.

In the present paper we discuss the thermodynamic properties of the spin-1/2 KHAF on a finite lattice of $N = 42$ sites. These results were obtained by large-scale numerical calculations (5 Mio. core hours) using the finite-temperature Lanczos method (FTLM).72–76 The
The ground state and the lowest energy levels, $R = 4$ for $M = 1$, $R = 2$ for $M = 2$, ..., $8$ and then again $R = 10$ for $8 < M < 16$. The number of Lanczos iterations for each random vector was determined by reaching convergence for the two lowest energy levels. In subspaces with $M \geq 16$ the Hamiltonian was diagonalized completely. The total computation time for the kagomé system of 42 sites was $\sim 5 \cdot 10^6$ core hours at the Leibniz Supercomputing Center’s supermuc.

Figure 1. (Color online) (a) Specific heat of the KHAF as function of temperature at $B = 0$ for various systems sizes (logarithmic temperature scale). (b) Specific heat of the KHAF and the SHAF as function of temperature at $B = 0$ (linear temperature scale).

We start with the discussion of the specific heat $C(T)$, the entropy $S(T)$ and the uniform susceptibility $\chi_0(T)$ using a logarithmic scale for $T$ in order to make the low-temperature features transparent, see Figs. 1(a), 3(a), and 4(a). The main maximum in the specific heat, set by the exchange $J$, is at $T = 0.67$, its height is...
Below $T = 0.25$ the curvature of $C(T)$ changes and a shoulder-like profile is present for $0.1 < T < 0.25$. This feature seems to be size-independent, i.e., finite-size effects appear likely only at $T < 0.1$. In this low-temperature region also a difference between odd and even lattice sizes $N$ occurs, that is related to the GS value of the total spin (doublet vs singlet), where even $N$ with a singlet GS seem to better fit to the spin-liquid GS present for thermodynamically large systems.

The behavior at very low temperatures $T < 0.1$ deserves a specific discussion, where we focus on even $N = 30, 36$ and $42$. First we notice that for $N = 36$ and $42$ just below the shoulder there is a rather flat maximum at about $T = 0.05$. At very low temperatures we observe a well pronounced extra peak in the specific heat. This peak marks the appearance of low-lying singlets above the ground state and is thus related to the singlet-singlet gap. Common expectations are that such gaps shrink with increasing size $N$. But in accordance with recent exact diagonalization studies, this peak moves towards higher temperatures with increasing $N$, as highlighted by the arrow in Fig. 1(a). One reason is that the singlet-singlet gap as well as the singlet-triplet gap do not shrink (considerably) when going from $N = 36$ to $N = 42$ and even $N = 48$, instead the singlet-singlet gap grows and the singlet-triplet gap shrinks only slightly. This behavior can be further rationalized by looking at the total density of states $n(E^*)$ as a function of the respective excitation energy $E^*$ and the contributions of the different sectors of total magnetization $M$ to $n(E^*)$ as displayed in Fig. 2. The density of states is evaluated by histograming the Krylov space energy eigenvalues together with their respective weights. The bin size is chosen as $J/100$. From Fig. 2 it becomes obvious which sector of $M$ contributes to $C(T)/N$ at various low-temperature regimes.

Having in mind the sum rule
\[
\int_0^\infty \frac{C(T)}{NT} dT = \int_{T=0}^{T=\infty} ds = s_\infty - s_0 = k_B \log(2),
\]
we may speculate that the weight of the extra peak at very low temperatures moves towards the shoulder with increasing $N \to \infty$, thus making the shoulder more pronounced. To conclude, we argue that our results are in favor of a low-temperature shoulder rather than an additional low-temperature maximum, compare Fig. 1(b). This is in accordance with recent tensor network calculations.

![Figure 2](image.png)

Figure 2. (Color online) Binned density of states for $N = 36$ (dashed curves) and $N = 42$ (solid curves) as a function of the respective excitation energy $E^*$: total density of states – black, total density of states for $|M| = 1$ – red, for $|M| = 2$ – green, and for $|M| = 3$ – blue.

![Figure 3](image.png)

Figure 3. (Color online) (a) Entropy of the KHAF as function of temperature at $B = 0$ for various systems sizes (logarithmic temperature scale); the arrow marks the movement of the low-lying density of states. (b) Entropy of the KHAF and the SHAF as function of temperature at $B = 0$ (linear temperature scale).

The behavior of the low-temperature peak also means that concerning the density of singlet states, weight is not simply shifted towards lower and lower energies with increasing $N$. It may be that the singlet-singlet gap closes with increasing $N$, but the density profile seems to behave differently, as can be noted by comparing the dashed ($N = 36$) and solid black ($N = 42$) curves in Fig. 2. This observation is further supported by the behavior of the entropy $S(T)$ at $B = 0$, which is shown in Fig. 3. As highlighted by the arrow, the temperature above which the entropy rises moves towards higher temperatures with increasing $N$ in accordance with the motion of the low-temperature maximum of $C(T)$. 
Figure 4. (Color online) (a) Susceptibility of the KHAF as function of temperature at $B = 0$ for various systems sizes (logarithmic temperature scale). (b) Susceptibility of the KHAF and the SHAF as function of temperature at $B = 0$ (linear temperature scale).

The singlet-triplet gap is even larger than the singlet-singlet gap, therefore the zero-field susceptibility exhibits a gapped behavior, as displayed in Fig. 4. For odd $N$ the ground state possesses non-zero spin, therefore the susceptibility diverges Curie-like in these cases. Since the singlet-triplet gap does not move much with increasing size $N$, it is not possible to draw definite conclusions about the functional form of $\chi$ for $T \to 0$. Nevertheless, DMRG calculations suggest that the singlet-triplet gap does not vanish in the thermodynamic limit, but approaches $0.13(1)$.\(^{15}\)

Finally, we compare $C(T)$, $S(T)$ and $\chi_0(T)$ for the (highly frustrated) KHAF with the corresponding FTLM data for the (unfrustrated) spin-1/2 square-lattice Heisenberg antiferromagnet (SHAF) of $N = 32$ sites, see Figs. 1 (b), 3 (b), and 4 (b), where we use a linear temperature scale. The temperature profile of all three quantities exhibits significant differences between the KHAF and the SHAF illustrating the tremendous role of frustration in a wide temperature range and, in particular, at low temperatures.\(^{81}\) Note that at high temperatures the quantities $C(T)$, $S(T)$ and $\chi_0(T)$ for both models approach each other, since square and kagomé lattices have identical coordination number $z = 4$. Thus the high-temperature series for $C$ and $\chi_0$ are identical up to order $1/T^2$ and $1/T^3$, respectively, see, e.g., Refs. 54 and 82.

Figure 5. (Color online) Magnetization vs applied magnetic field for various temperatures: both magnetization and field are normalized by their saturation values.

Figure 6. (Color online) Derivative $dM/dB$ vs applied magnetic field $B$ for various temperatures for $N = 42$. Both, magnetization and field are normalized by their saturation values.

The KHAF exhibits a number of interesting properties in an applied magnetic field.\(^{27,60–67,69,70}\) Magnetization plateaus exist at $3/9 (= 1/3)$, $5/9$ and $7/9$ of the saturation magnetization $M_{\text{sat}}$ for the infinite system at $T = 0$, where the $1/3$ plateau is the widest. An additional tiny plateau at $1/9$ appears possible.\(^{65}\) Moreover, the magnetization curve at $T = 0$ shows a macroscopic jump to saturation due to the existence of independent localized magnons.\(^{61,69–71,83}\)

In a calculation of a small lattice the magnetization curve is unavoidably a sequence of steps, that happen at ground state level crossings at certain field values, compare Fig. 5. Thus, due to this discretization the existence of smaller plateaus cannot be unambiguously deduced from such a single magnetization curve. Moreover, a specific plateau value $M_{\text{plateau}}/M_{\text{sat}}$ can be missed in the $M(B)$ curve, if it does not fit to the lattice size $N$. For example, for our largest system of $N = 42$ the val-
uses at $M_{\text{plateau}}/M_{\text{sat}} = 5/9$ and $7/9$ are not present in Fig. 5 for $M(B)$ curves of other finite kagomé lattices, see, e.g., Refs. 27, 62, 66, and 67. Nevertheless, the major plateau at $1/3$ (marked by the blue horizontal arrow) is clearly visible in Fig. 5, since it is the widest of all plateaus and it is very robust as a function of $N$. We also mention that the pretty wide plateaus just above the $1/3$-plateau most likely disappear for $N \to \infty$. The influence of the temperature on the $M(B)$ curve is relevant for experimental studies. From Fig. 5 it is obvious that for slightly elevated temperatures the detection of plateaus by measuring $M(B)$ is difficult. Therefore, the first derivative $dM/dB$ as a function of $T$ as presented in Fig. 6 is often used in experiments to find plateaus, cf., e.g., Ref. 84. The $1/3$-plateau can be detected by the pronounced minimum in $dM/dB$. Note that the oscillations of the red $dM/dB$ curve are also due to finite-size effects. It is worth mentioning that the position of the minimum in $dM/dB$ stemming from the $1/3$-plateau is shifted to higher values of $B$ with increasing temperature. Thus, for $T = 0.05$ ($T = 0.02$) it is at $B/B_{\text{sat}} = 0.398$ ($B/B_{\text{sat}} = 0.381$) whereas the midpoint of the plateau is at $B/B_{\text{sat}} = 0.364$. For $T = 0.1$ the minimum in $dM/dB$ is hardly detectable, see Fig. 6. This shift is related to the ‘asymmetric melting’ of the plateau due to the larger density of low-lying excited states below the plateau than that of low-lying excitations above the plateau. Thus, for the KHAF the very existence of the plateau can be found by measuring $dM/dB$ at $T \lesssim 0.1$, but to determine the precise position of it requires very low temperatures. Last but not least, we notice that the jump of the magnetization to saturation at $T = 0$ is washed out at $T > 0$, but its existence leads to a high peak in $dM/dB$ at the saturation field.

The influence of the magnetic field $B$ on the specific heat $C$ is shown in Fig. 7 for $N = 42$. At very low temperatures and moderate fields the influence of $B$ is determined by the shift of the low-lying magnetic excitations with $M = 1$ and $M = 2$ towards and even beyond the zero-field singlet GS. As a result, the position and the height of the low-temperature (finite-size) peaks in $C(T)$ are substantially changed. At temperatures below the main maximum there is no obvious systematic behavior of $C(T)$ as a function of $B$, see Fig. 7(a). However, at magnetic fields slightly below and above the saturation field, the huge manifold of low-lying localized multimagnon states (already mentioned in the introduction) leads to an extra low-temperature maximum, see Fig. 7(b), persisting in the thermodynamic limit. It is worth mentioning, that for $B \lesssim B_{\text{sat}}$ in the thermodynamic limit this extra-maximum likely becomes a true singularity indicating a low-temperature order-disorder transition into a magnon-crystal phase.

Interestingly, the influence of $B$ on the main maximum of $C$ depicted in Fig. 8 shows some systematics (see also Ref. 26, Fig. 3): (i) The height of the maximum $C_{\text{max}}$ remains almost constant until $B \sim 0.8B_{\text{sat}}$ and increases smoothly for $B > B_{\text{sat}}$. (ii) The position of the maximum $T_{\text{max}}$ as a function of $B$ exhibits two maxima at $B = 0$ and $B \approx 1.1B_{\text{sat}}$ and two minima at $B \approx 0.5B_{\text{sat}}$ and $B \approx 1.4B_{\text{sat}}$ as well as two regions $0.65B_{\text{sat}} \lesssim B \lesssim 0.9B_{\text{sat}}$ and $B \gtrsim 1.5B_{\text{sat}}$ with an (almost) linear growth of $T_{\text{max}}$. To illustrate the role of frustration we contrast this behavior with that of the unfrustrated SHAF, also see Fig. 8. At low magnetic fields the value of $T_{\text{max}}$ is determined by $J$, and for both models $T_{\text{max}}$ behaves very similar. On the other hand, the difference in $C_{\text{max}}$ is significant and can be related to the
different low-energy physics which influences $C$ at higher $T$ according to the sum rule (3). Beyond $B \sim 0.5B_{\text{sat}}$ the different behavior is more evident. The almost straight increase in $T_{\text{max}}(B)$ for $0.65B_{\text{sat}} \lneq B \lneq 0.9B_{\text{sat}}$ in the case of the KHAF indicates a paramagnetic behavior. The maximum around $B = B_{\text{sat}}$ signals strong frustration because it is related to the manifold $W$ of localized multi-magnon states setting an extra low-energy scale in the vicinity of the saturation field in frustrated Heisenberg systems with a flat band. For the specific flat-band setting an extra low-energy scale in the magnetization curve and the temperature profile of the specific heat at magnetic fields near saturation.

In accordance with Ref. 22 we also found that in the low-field regime the convergence at $T \lneq 0.1J$ to the thermodynamic limit is slow. Both the singlet-singlet gap as well as the singlet-triplet gap change very little with increasing system size. Therefore, at $B = 0$ the behavior below $T \lneq 0.1J$ seen in our calculations is still dominated by finite-size effects.

Finally, we mention that the relation of our data to the experimental data of the spin-liquid candidate Herbertsmithite is limited for several reasons. First, the exchange interaction of this compound is estimated as $J \sim 190$ K. Low-temperature measurements of the magnetization as well as the specific heat, in particular for $2K \lneq T \lneq 10K$, are well below $0.1J$ and can thus, unfortunately, not be compared with our simulations due to the finite-size effects below $= 0.1J$. Second, it is up to now not settled how the interacting impurities as well as the lattice vacancies in Herbertsmithite can be dealt with in thermodynamic calculations. Moreover, there might be a noticeable spin anisotropy present in Herbertsmithite. Despite all the uncertainties, it is believed that the spin liquid ground state is a rather stable phenomenon.

In view of the numerical effort of our investigations we conjecture that exact diagonalization studies of the thermodynamic behavior of the KHAF might be feasible for $N = 45$ and $N = 48$, but larger systems must be dealt with by, e.g., DMRG and tensor network methods.

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