Specific heat of the $S=\frac{1}{2}$ Heisenberg model on the kagome lattice: High-temperature series expansion analysis

G. Misguich*

Service de Physique Théorique, URA 2306 of CNRS CEA Saclay, 91191 Gif-sur-Yvette Cédex, France

B. Bernu†

Laboratoire de Physique Théorique des Liquides, Université P. et M. Curie, URA 765 of CNRS Case 121, 4 Place Jussieu, 75252 Paris Cédex 05, France

We compute specific heat of the antiferromagnetic spin-$\frac{1}{2}$ Heisenberg model on the kagome lattice. We use a recently introduced technique to analyze high-temperature series expansion based on the knowledge of high-temperature series expansions, the total entropy of the system and the low-temperature expected behavior of the specific heat, as well as the ground-state energy. In the case of the kagome-lattice antiferromagnet, this method predicts a low-temperature peak at $T/J \lesssim 0.1$.

I. INTRODUCTION

We consider the nearest-neighbor Heisenberg model on the kagome lattice:

$$\mathcal{H} = 2\sum_{\langle i,j \rangle} \vec{S}_i \cdot \vec{S}_j. \tag{1}$$

Because of its unconventional properties, the spin-$\frac{1}{2}$ kagome antiferromagnet (KAF) has been subject to an intense activity these last years. All studies agree that this frustrated two-dimensional magnet has no long-ranged magnetic order at zero temperature.[1–9] Exact diagonalization studies have established that the low-energy spectrum of the kagome-lattice Heisenberg antiferromagnet has a large number of spin-singlet states before the first spin 1 excited state.[10] Among the different theories developed to explain this unconventional spectrum, short-range resonating valence-bond (RVB) pictures have been proposed.[11–15]

The high-temperature (HT) expansion of the specific heat as been computed up to order $1/T^{16}$ by Elstner and Young.[16] We have checked and extended this series to order $1/T$.[17] The additional term for the specific heat per site is given by $c_v(T) = \frac{3}{2}\beta^2 + \cdots + 1\,845\,286\,680\,253/366\,912\,000\beta^{17}$. Elstner and Young analyzed the series through conventional Padé approximants with the additional constraint that the specific heat must vanish at $T=0$. At the highest orders, they found a specific heat curve with a single maximum around $T=1.3$ but with a large entropy deficit of about 40%: $\int_0^\infty c_v(T)/T dT \simeq 0.6 \ln(2)$. They concluded the existence of a low-temperature structure corresponding to an entropy of about 40% of ln 2 and claimed that this low-energy structure could not be accessed from the high-temperature expansion of the specific heat. They argued that even though the *classical* kagome antiferromagnet has a nonvanishing ground-state entropy, quantum fluctuations in the spin-$\frac{1}{2}$ model are expected to lift this degeneracy.

In this paper, we revisit the question of the specific heat with the help of a new method to analyze high-temperature series data. Compared to the usual Padé approximant approach, this method[17] takes advantage of additional information on the system: the two sum rules on the energy and on the entropy are exactly satisfied. In many simple systems (one- and two-dimensional ferro- or antiferromagnets), this technique allows one to compute accurately the specific heat *down to zero temperature*,[17] which is not the case if one does a direct Padé analysis of the series. For the present kagome model we show that this method provides rich semiquantitative information on the specific heat curve, although a full convergence down to zero temperature cannot be achieved.

II. DIRECT HIGH-TEMPERATURE EXPANSION OF THE SPECIFIC HEAT

We reproduce here the first attempt by Elstner and Young[16] to compute the specific heat from its high-temperature expansion alone. We use Padé approximants to extrapolate the series. We impose the specific heat to vanish at low temperature as T, T^2, or T^3. At orders 9 to 17, only six such approximants do not develop poles or zeros in the interval $T \in \,]0,\infty]$ (Fig. 1). One should notice at this point that the remaining Padé approximants agree reasonably well down to zero temperature. This is usually not the case in a two-dimensional antiferromagnet where even the position of the peak ($T \simeq 1$) can hardly be obtained by the use of direct Padé approximants to the series for the specific heat.[17] From this point of view, the HT series expansion of the kagome model seems to have a faster convergence than models such as the triangular-lattice antiferromagnet.

By integration of these approximants, we evaluate the ground-state energy $e_0 = \int_0^\infty c_v(T)dT$ and the ground-state entropy $s_0 = \log(2) - \int_0^\infty c_v(T)dT$. These values are indicated in Fig. 1. The ground-state energy is about -0.845, only slightly higher (0.02) than estimations obtained from exact diagonalizations. The entropy deficit is very large: 0.3 [40% of $\log(2)$]. Elstner and Young[16] argued that a low-temperature peak should be present in the specific heat in order to compensate the deficit of 40% of $\log(2)$.[18] However, this low-
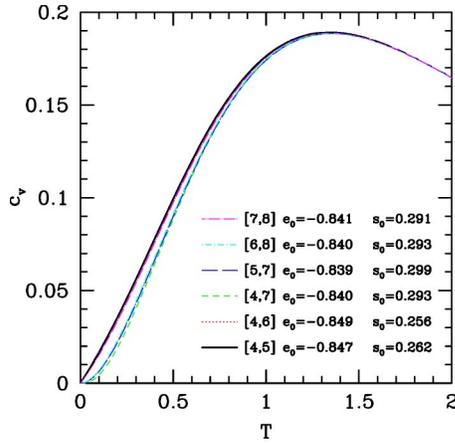
FIG. 1. (Color online) Specific heat obtained from Padé approximants to the high-temperature series of $c_v(T)$. Only approximations of degrees $[u, u+1]$, ($[u, u+2]$ and $[u, u+3]$) are considered. They vanish at zero temperature as T (T^2 and T^3). The only six such approximants from order 9 to 17 which are positive on the positive real axis are shown. The ground-state energy per site e_0 and entropy s_0 obtained by integrating these Padé approximations is indicated.

temperature peak should "contain" almost no energy (2%), which means that such peak would have to occur at very low temperatures. In order to estimate this temperature, one can add a δ-function peak to the curves of Fig. 1 in order to recover the correct energy and entropy variations. This constrains both the location (temperature T_1) and the weight of the δ peak. By averaging over the different curves of Fig. 1 one finds $T_1 \simeq 0.05$ ($T_1 \simeq 0.08$) for a ground-state energy $e_0 = -0.865$ ($e_0 = -0.875$). These estimates are in agreement with the conclusions of the more elaborate treatment described below.

III. ENTROPY METHOD

In this section we briefly summarize the method we use to compute the specific heat. More details can be found in Ref. 17. The specific heat c_v and the temperature T can be obtained from the entropy s as a function of the energy e using basic thermodynamic relations:

$$T(e) = 1/s'(e), \tag{2}$$

$$c_v(e) = -\frac{s'(e)^2}{s''(e)}. \tag{3}$$

From Eq. (3) one can convert[17] a high-temperature series for $c_v(T \to \infty)$ into a series for $s(e \to 0)$ [$e = 0$ at $T = \infty$ for the Hamiltonian of Eq. (1)]. The truncated series are plotted in Fig. 2. Using Eq. (2), the entropy can be plotted as a function of temperature (right of Fig. 2). A good convergence is observed down to relatively low energies ($e \sim -0.75$) but the corresponding entropy remains very large [more than 60% of $\ln(2)$], although the ground-state energy is not much lower
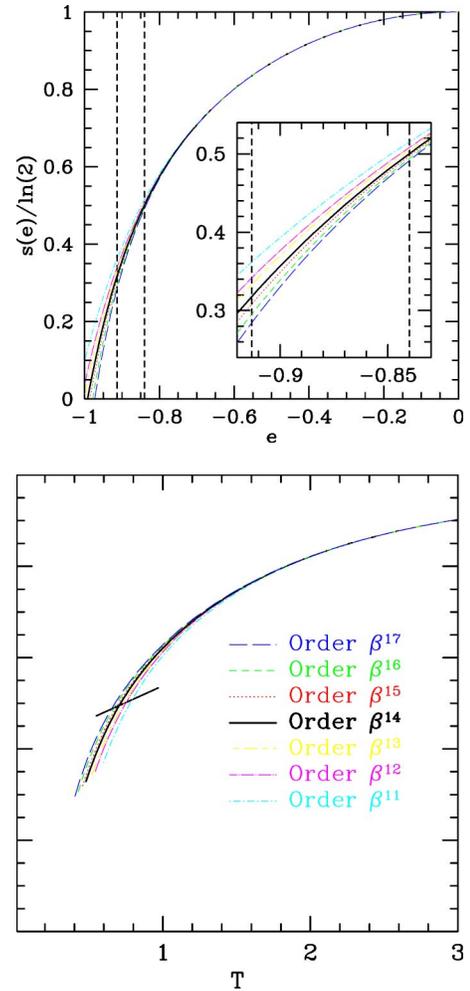
FIG. 2. (Color online) Left: Series expansion for $s(e)$ (entropy per site) as a function of the energy per site e. The result of the bare series are displayed for orders from β^{11} to β^{17}. The dashed vertical lines indicate upper ($e = -0.84267$) and lower ($e = -0.909952$) rigorous bounds on the ground-state energy in the thermodynamic limit (Ref. 19 Right: Same data as on the left panel but plotted as a function of temperature $T = 1/s'(e)$. For each curve the lowest temperature corresponds to $e = -0.909952$ (lower bound). The black segment corresponds to $e = -0.84267$ (upper bound).

(the ground-state energy lies between the dashed vertical lines in Fig. 2). These results are consistent with a direct analysis of the series for $c_v(T)$ (Fig. 1). In addition, it appears that the "true" $s(e)$ must be bent downward below the curves of the truncated series (shown in Fig. 2) between e_0 and ~ -0.75 in order to reach $s = 0$ at $e = e_0$. Due to Eq. (3), this almost certainly implies a low-energy (and therefore low-temperature) *peak* in $c_v(T)$. This paper makes this idea more precise by computing the specific heat obtained by forcing the entropy to vanish at $e = e_0$.

The advantage of working on $s(e)$ rather than $c_v(T)$ is that a two-point Padé interpolation can be used to set the ground-state energy and the total entropy of the system. However, $s(e)$ is singular at $e = e_0$ [since $T = 1/s'(e) \to 0$ when $e \to e_0$]. For this reason one cannot directly approximate $s(e)$ by a rational fraction (Padé approximant). If we assume that the specific-heat behaves as

$$c_v \simeq (T/c_0)^\alpha \qquad (4)$$

at low temperature (c_0 has the dimension of an energy) and $s(e_0)=0$, $s(e)$ behaves as

$$s(e \to e_0) \simeq \frac{(\alpha+1)^{\alpha/(\alpha+1)}}{\alpha} \left(\frac{e-e_0}{c_0} \right)^{\alpha/(\alpha+1)}. \qquad (5)$$

The quantity

$$G(e) = \frac{s(e)^{1+1/\alpha}}{e-e_0} \qquad (6)$$

is then nonsingular at $e=e_0$ and can be approximated by a Padé form.[20] The series for $s(e \to 0)$ must therefore be transformed into a series for $G(e \to 0)$ before Padé approximants can then be computed in the usual way. In what follows all the Padé approximants will be approximations to this function $G(e)$. If no finite-temperature phase transition is expected, all approximants where $G(e)$ has a pole or a zero, or where $s'(e)$ or $s''(e)$ vanishes somewhere in the interval $]e_0,0[$ must be discarded. The remaining ones are called "physical" for brevity.

A. Low-temperature behavior of $c_v(T)$

Unlike some simpler magnets where the nature of the ground-state and elementary excitations is known,[17] the qualitative behavior of the specific heat when $T \to 0$ is unknown, although a $\sim T^2$ scenario has been proposed.[9,21] However, one of the striking facts about the model is the unusually high density of states *immediately above* the ground-state.[10] From this it is natural to expect gapless elementary excitations. If we assume quasiparticles with a dispersion relation $\epsilon_k \sim k^\gamma$ we get a specific heat $c_v \sim T^\alpha$ with $\alpha=D/\gamma$ in space dimension D. The (many body) density of states is $\rho(E_0+W) \sim \exp[N(W/N)^{\alpha/(\alpha+1)}]$ where N is the system size [consequence of Eq. (5) with $e-e_0=W/N$]. For an energy W of order one above the ground state, a density of states $\rho \sim 1.15^N$ was observed in exact spectra up to $N=36$ sites.[10] If this indeed holds up to the thermodynamic limit, it would imply $\alpha=0$ ($\gamma=\infty$) and an extensive entropy at zero temperature. This is unlikely in the present model[22] but this result points to a rather flat dispersion relation of the excitations, probably with $\gamma>1$. In the following we will consider the two cases $\gamma=1$ ($\alpha=2$) and $\gamma=2$ ($\alpha=1$).[23]

B. Ground-state energy and convergence of the different Padé approximants

In principle, the method above requires the knowledge of the ground-state energy e_0. If the value of e_0 is exact we expect the procedure to converge to the exact c_v if the number of known terms in the HT series increases to infinity.
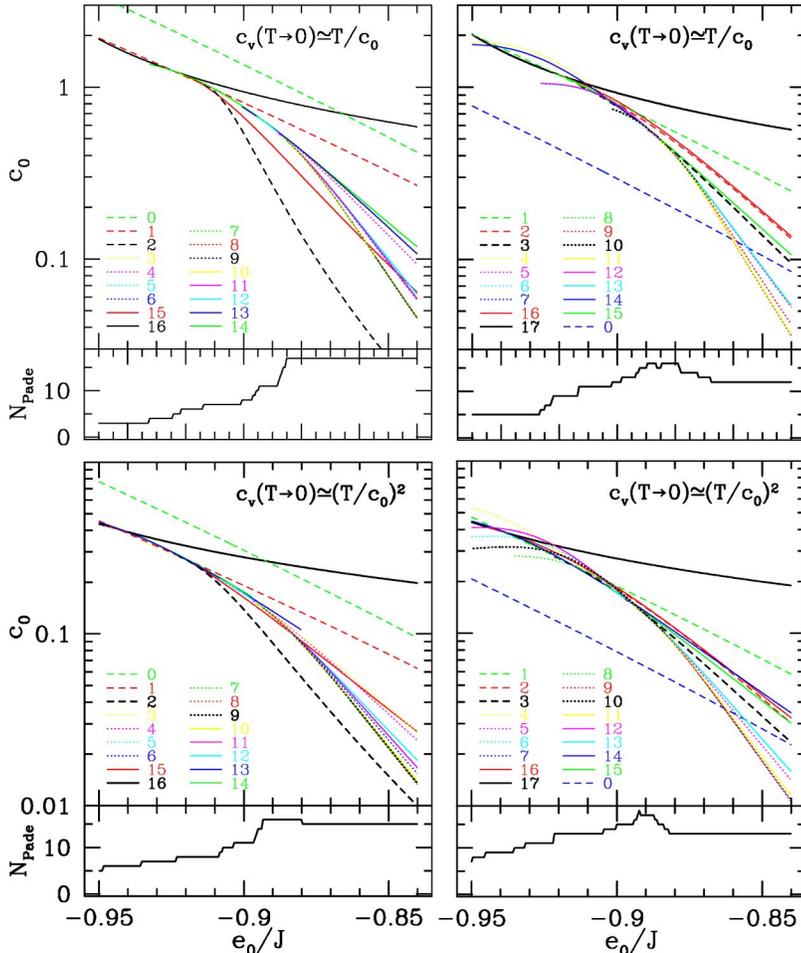
FIG. 3. (Color online) Zero temperature limit of c_0 [see Eq. (4)] for the different Padé approximants as a function of the ground-state energy. Top: a $c_v \sim (T/c_0)$ behavior is assumed at low temperature. Bottom: $c_v \sim (T/c_0)^2$. The degree u of the numerator of each approximant is given. The degree of the denominator is $d = n-u$ where n is the order of the series. Left: order $n=16$. Right: order $n=17$. The number of physical approximant (N_{Pade}) is plotted as a function of e_0 in each lower panel.
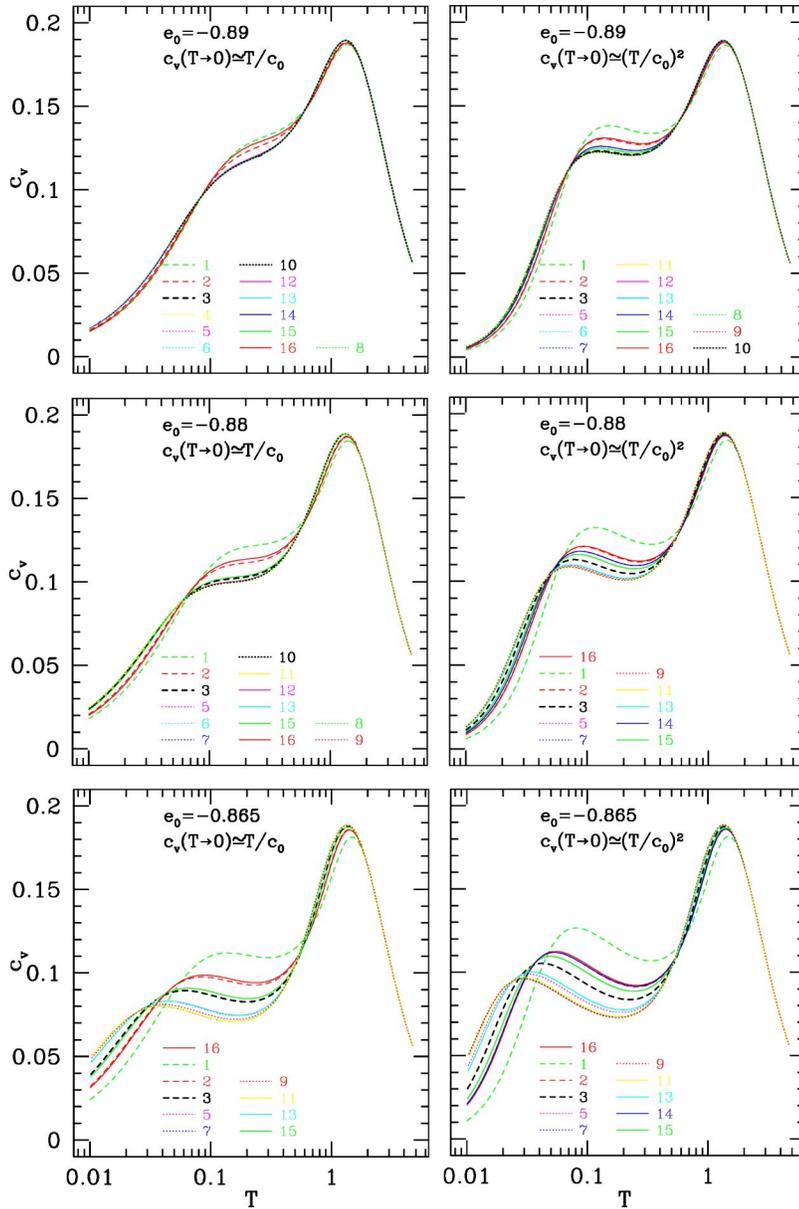
FIG. 4. (Color online) Specific heat computed at order β^{17} with the ground-state energies e_0 =−0.89 (top), e_0=−0.88 (center), and e_0=−0.865 (bottom). Left: c_v ∼T. Right: c_v∼T^2. The different curves correspond to all the physical Padés approximants. The degree u of the numerator is indicated (the denominator has degree $d=17-u$).

This is in agreement with our experience on solvable models (such as the spin-$\frac{1}{2}$ XY chain, for instance[17]) where the full series as well as e_0 are known exactly. Inversely, wrong values of e_0 cannot lead to any convergence as the limiting c_v would have to satisfy the HT series at all orders but would have a different energy sum rule. As a consequence, when e_0 differs from the true ground-state energy, the physical approximants gets fewer (and/or more scattered) when the order of expansion gets larger. Of course, the smaller the error on e_0 the longer series is needed to observe this departure from convergence. From this we assume that the existence of a larger number of physical approximant is an *indication* that e_0 (and α) is closer to the exact value. However, because a limited number of terms of the series are known, this only provides qualitative information and does not allow one to determine the energy completely.

From exact diagonalizations on systems with up to 36 sites, e_0 was evaluated by Waldtmann *et al.*[10] to be e_0 =−0.865±0.015 (see also Refs. 26 and 27). Variational cal-

culations as well as rigorous bounds on e_0 will be discussed in a separate paper.[19,28]

The specific heat curve can be rather sensitive to the choice of e_0. Since e_0 is not exactly known, it is important to perform scans in order to see how the specific-heat curve depends on e_0. We observe that, for some choice of e_0 many Padé approximants at a given order give almost the same specific heat curve whereas some other choice of e_0 leads to some significant scattering in the specific heat curves. This can conveniently be seen, for instance, by looking at the value of the different Padé approximants at $e=e_0$. Since $G(e_0)$ and c_0 [defined by Eq. (4)] are simply related by

$$G(e_0) = \frac{\alpha+1}{c_0 \alpha^{1+1/\alpha}}, \tag{7}$$

we plot c_0 [which has a direct physical meaning in terms of $c_v(T{\to}0)$] in Fig. 3 as a function of e_0 for all physical Padé approximants at order β^{16} and β^{17} [both for $c_v(T)\sim T$ and
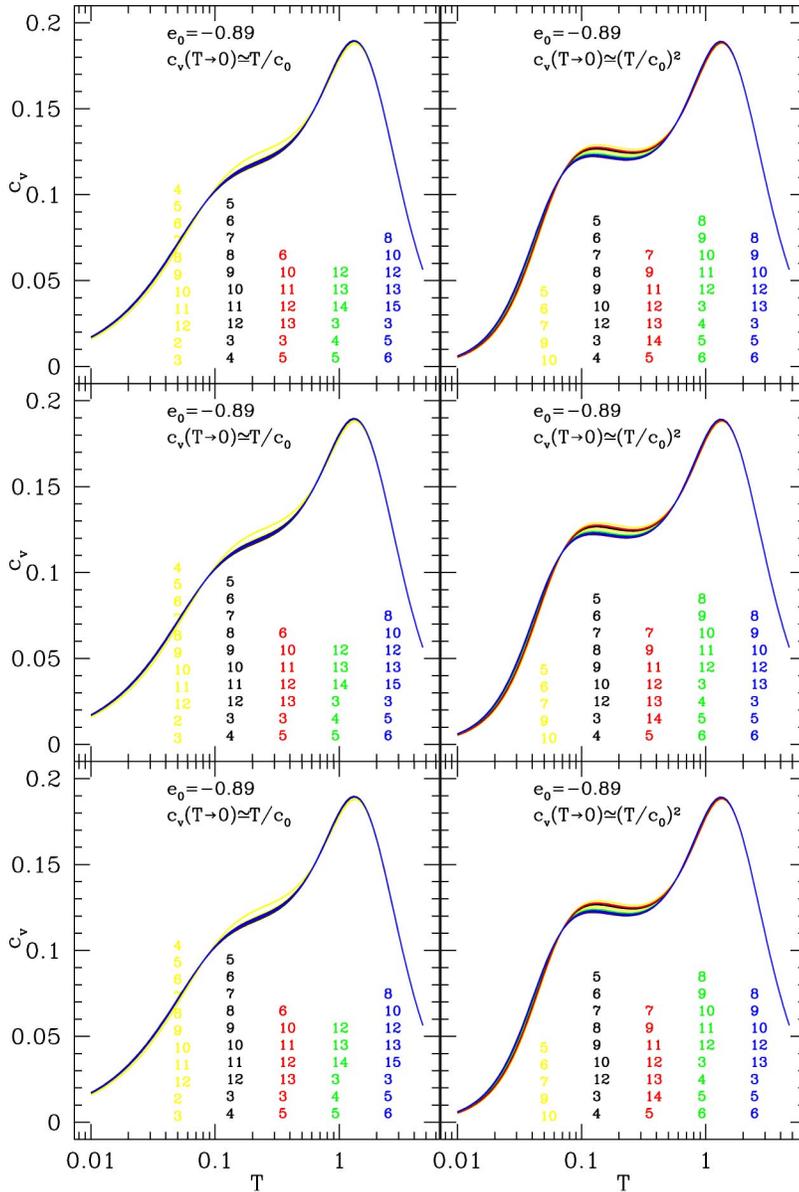
FIG. 5. (Color online) Specific heat curves from order β^{13} to β^{17} with $e_0=-0.89$ (top), $e_0=-0.88$ (center), and $e_0=-0.865$ (bottom). The degree of the numerator of each approximant is indicated and each column corresponds to a given order (β^n) of the series ($n=13$, 14, 15, 16, and 17 from left to right). Left panels: $c_v \sim T$. Right panels: $c_v \sim T^2$. In all cases the specific heat shows a maximum around $T \simeq 1.3$ [corresponding to $e(T) \simeq -0.7$, see Fig. 2] and a low-temperature peak (or shoulder).

$\sim T^2$ at low temperature]. It turns out that c_0 is representative of the full specific-heat curve in the sense that if two Padé approximants give "close" values of c_0 (say a relative difference less than 10^{-3}), their corresponding specific-heat curves are similar (typical relative difference of 10^{-2}) *for all temperatures*. This low-temperature coefficient c_0 is therefore a useful quantity to monitor how the $c_v(T)$ result depends on the choice of the degree of the Padé approximant.[30] In all cases the "optimal" energy region is around $e_0 \simeq -0.88 \pm 0.02$.[31] We also observe a gradual shift of the optimal region to higher energies as the order of the series is increased. We analyzed this effect and performed several extrapolations to the infinite-order limit (data not shown). It is not clear, however, that this indirect method to determine the ground-state energy is more accurate than the other available estimates.[19]

C. Low temperature peak in $c_v(T)$

The curves corresponding to all physical approximants at order β^{17} for $e_0=-0.865$, $e_0=-0.88$, and $e_0=-0.89$, and for $c_v \sim T$ and $c_v \sim T^2$ are shown in Fig. 4. Although some uncertainties remain concerning the ground-state energy of the model as well as the low-temperature behavior of the specific heat, the results are relatively well converged down to $T \simeq 0.7$ and the location of the high temperature peak is almost independent from the unknowns (e_0 and α) and is in agreement with previous studies.[6,9,16,21] In addition, all the scenarios we investigated gave rise to a low-temperature peak (or a shoulder) in the specific heat at $T \simeq 0.02$–0.1.

We also looked at the order dependence of the specific heat curves. For a given value of the ground-state energy some approximants give similar curves for $c_v(T)$ while some others are "isolated." The later ones can be recognized as isolated curves in Fig. 3. According to our experience[17] with this method, those isolated approximants do not reflect the convergence to the true function. We obtained the results of Fig. 5 by keeping only the approximants whose value of c_0 is at less than 3×10^{-3} from the c_0' of another approximant. This selection was repeated from orders β^{13} to β^{17} for the six combinations of ground-state energies and low-temperature

behaviors used before. As one can see, the low-temperature structure appears to be a robust feature, although a convergence of the full curve is not reached for $T \lesssim 0.6$. Still, a better convergence as a function of the order of the series (and a larger number of physical Padé approximants) is obtained when the ground-state energy is low ($e_0 = -0.89$ or $e_0 = -0.88$). This suggests that the actual value of e_0 may be lower than -0.865, although $e_0 = -0.89$ is probably too low (compared to the available estimates[1,10,26,27]).

For $N = 18$ spins, exact diagonalizations[16] gave a low-temperature peak of the specific heat at $T \simeq 0.2$ and $c_v \simeq 0.17$. A hybrid method[21] based on exact diagonalizations and high-temperature series expansion gave a peak at $T \simeq 0.2$ and $c_v \simeq 0.17$ for $N = 36$ (see also Ref. 6). Quantum Monte Carlo simulations for $N = 72$ spins[6] indicated that a peak may exist below $T \simeq 0.3$ for this system. Those results obtained for small systems are qualitatively consistent with those of Fig. 5 but our peak is located at a lower temperature by at least a factor of 2. We think that this discrepancy is likely to be due to finite-size effects in previous studies.

IV. CONCLUSIONS

By means of a detailed high-temperature series analysis we provided quantitative estimates for the specific heat curve of the spin-$\frac{1}{2}$ Heisenberg antiferromagnet on the kagome lattice. Those results show a low-temperature peak in the specific heat of the model for $T \lesssim 0.1$, although its precise location cannot be determined due to uncertainties on the ground-state energy. The corresponding degrees of freedom are also responsible for the large density of singlet states observed in exact diagonalization studies but their nature, as well as the nature of the ground state itself, remains to be explained.

*Electronic address: misguich@spht.saclay.cea.fr

†Electronic address: bernu@lptl.jussieu.fr

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[20] Equation (6) is slightly different from the original choice made in Ref. 17.

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[22] See Ref. 13 for a related model where this property is explicitly demonstrated. This, however, requires some fine-tuning of the Hamiltonian and there is no reason to think that the kagome Heisenberg model may have such a very special property. From finite-size spectra it can, however, be difficult to resolve the difference between a model with an extensive entropy at $T = 0$ and a system with vanishing entropy at $T = 0$ but a large low-temperature peak in the specific heat for $T \ll J$. We stress that the $T = 0$ entropy that follows from $\rho \sim 1.15^N$ is consistent with that of the low-temperature peak predicted by the present method: about 20–25% of log(2).

[23] The possibility of having a gapped spectrum with a (very) small gap cannot be excluded. This would be the case if the system realizes a $\mathbb{Z}_2$ spin-liquid, as predicted in large-N approaches (Ref. 24) or if the ground-state has some valence-bond long-ranged order (Refs. 14, 15, and 25). A thermally activated behavior $c_v \sim \exp(-\Delta/T)$ can be treated within the entropy method (Ref. 17) and we applied it to the present model. The gaps we obtained are rather large (of order 1) but the specific-heat curves also exhibited a low-temperature peak consistent with the $c_v \sim T^\alpha$ results discussed below.

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[28] The expectation value of the Hamiltonian in any first-neighbor valence-bond covering is $e = -0.75$. By optimizing the wave function in the vicinity of each triangle without any dimer, Elser obtained a variational state with energy $e = -0.8333$. This rigorous upper bound was refined by Pierre (Ref. 29 who used an improved variational state to prove that $e_0 \leq -0.84267$; and exact lower bound can be obtained by remarking that the ground-state energy per triangle in the kagome AF is necessarily larger than the ground-state energy of an isolated triangle. This gives $e_0 \geq -1$. By extending this reasoning to larger clusters we can improve this lower bound. We can prove that $e_0 \geq -0.909952$ by

exactly diagonalizing a 24-site cluster with open boundary conditions. Farnell *et al.* used a coupled cluster method and predicted $e_0 \simeq -0.8504$ (Ref. 26). They recently improved their calculation (Ref. 27) and obtained $e_0 \simeq -0.86208$ by fitting their nth-order results by $1/n^2$ corrections. We observed that their bare data are better described by $1/n$ corrections. In that case a fit gives $e_0 \simeq -0.875$.

[29] L. Pierre (unpublished).

[30] The physical meaning of this observation is that if two specific heat curves have (i) the high-temperature expansion up to some relatively high order, (ii) the same ground-state energy, (iii) the same entropy, and (iv) the same $c_v \sim (T/c_0)^\alpha$ limit at low temperature they must be very "similar."

[31] For lower energies the curves gets closer but the number of physical Padé approximants actually gets smaller and smaller, as can be seen in the lower panels of Fig. 3.