Low-energy excitations of the kagomé antiferromagnet and the spin-gap issue

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Abstract – In this paper we report the latest results of exact diagonalizations of SU(2) invariant models on various lattices (square, triangular, hexagonal, checkerboard and kagomé lattices) and revisit critically in this light some of our previous analysis. We focus on the low-lying levels in each $S$ sector. The differences in behavior between gapless systems and gapped ones are exhibited. The plausibility of a gapless spin liquid in the Heisenberg model on the kagomé lattice is discussed.

A rough estimate of the spin susceptibility in such an hypothesis is given. The evolution of the intra-$S$-channel spectra under the effect of a small perturbation is consistent with the proximity of a quantum critical point. We reemphasize that the very small intra-$S$-channel energy scale observed in exact spectra is a very interesting information to understand the low-$T$ dynamics of this model.

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The RVB concept of a quantum spin liquid due to Anderson in the beginning of the seventies has motivated a very large number of works both theoretical and experimental [1,2]. Very recently a specifically interesting candidate for a spin liquid has emerged: the Herbersmithite characterized by a fluctuating behavior down to temperatures of the order of $10^{-4}$ times the coupling constant [3–7]. In this compound the magnetic Cu ions are located at the vertices of a kagomé lattice and the interactions between these spin-1/2 ions are supposed to be fairly isotropic. The possibility that a simple Heisenberg model might capture the physics of this compound, has therefore attracted interest, although a significant amount of defects and other interactions, e.g. Dzyaloshinskii-Moriya interactions, are likely to be present. However, in spite of many studies, the nature of the ground-state and first excitations of the spin-1/2 kagomé Heisenberg antiferromagnet (KHA) is still disputed [8–16].

Exact diagonalizations [8–10,17,18] have very early shown that something special happens in the KHA. n-points correlations functions (up to $n=8$) decrease at short distances [8,16–21]. The spectral properties observed on these small samples (up to $N=36$ sites) are quite unusual: these spectra exhibit two low-energy scales: besides a finite-size spin-gap scale, there is a second much lower-energy scale inside each sector of total spin $S$. A crude $1/N$ extrapolation of the finite-size spin-gaps to the thermodynamic limit lead to an “estimate” of a putative thermodynamic spin-gap of about $1/20$ of the coupling constant [10]: an analysis and a result that we want to revisit critically at the light of recent more extensive studies of the spectra of samples up to $N=36$ spins.

Many other numerical approaches have been used to attack this problem: CORE renormalization approaches (Budnik et al. [11] and Capponi et al. [22]), sophisticated series expansion (Singh et al. [12,15]), DMRG calculations (Jiang et al. [16]), and more recently MERA renormalization procedure (Evenbly et al. [23]), give evaluations of the ground-state energy and for most of them of the spin-gap.

Since 1998 and our publication [10] the performances of computers have increased noticeably (not enough to compute the spectrum of samples much larger than 36) but sufficiently for computing a large number of levels in any $S$ sector of various clusters up to $N=36$ spins and thus allowing a full analysis of the exact $T=0$ magnetism of this “large molecule”. The object of this paper is to display these results and compare them to the
behaviour of the Heisenberg antiferromagnet on various other lattices: square (SHA), hexagonal (HHA), triangular (THA), checkerboard (CHA) of similar sizes. We show that up to this size, the KHA has a magnetic behavior which compares extremely well to other antiferromagnets known to be gapless in the thermodynamic limit and that, the extrapolation of such finite-size data although it does not rule out a finite spin-gap, cannot yield a significant measurement of an eventual very small spin-gap: our paper written in 1998 was on this point misleading [10]. From the methodological point of view the analysis of the data that we perform in this paper can readily be applied to any results obtained by other numerical finite-size calculations (DMRG etc.). The third message of this paper is that the much lower-energy scale that appears inside each $S$ sector play a key role in the true dynamics of this magnet and suggests a critical or quasi-critical behavior.

**Gapless and gapped magnets through exact diagonalizations: ground-state energy vs. total spin.** – In fig. 1, we display the lowest eigen-levels of the SHA, THA, HHA, KHA and CHA samples of 36 spins (with periodic boundary conditions) and study how they evolve with the total spin value $S$ of the sample. The Hamiltonian is

$$\mathcal{H} = \sum_{\langle i,j \rangle} \vec{s}_i \cdot \vec{s}_j,$$

(1)

where $\vec{s}_i$ is the individual spin $1/2$ at site $i$ and the sum $\langle i,j \rangle$ runs over pairs of nearest neighbors ($\vec{S} = \sum \vec{s}_i$).

In the checkerboard lattice, also known as the pyrochlore slab lattice, tetraedra share corners and are disposed on a checkerboard: all couplings along tetraedra edges are identical.

Figure 1 (top) shows that in most of the cases (except CHA), the evolution of the first level in each $S$ sector is extremely well described by a $S(S+1)$ behavior which extend to $S = S_{\text{max}} = N/2$ (not shown in the figure for the sake of clarity).

Such a behavior can be described by the equation:

$$e_N(S) = e_N(0) + \alpha_N \frac{S(S+1)}{N^2},$$

(2)

where $e_N(S)$ is the lowest energy per spin in the $S$ channel for a sample of size $N$. The limit of eq. (2), when the size of the sample goes to infinity is the well-known formula of the energy vs. magnetization of classical antiferromagnets:

$$e(m) = e(0) + \frac{1}{2\chi} m^2,$$

(3)

where $m$ is the magnetization ($0 \leq m \leq 1$) of the sample and $\chi$ the dimensionless susceptibility. (With this definition, the magnetic moment per spin in a magnetic field $H$ reads $\mu = (g\mu_B/2)^2 H$ and $\alpha_N = 2/\chi N$).

In fact there is a tiny cusp at $S = 6$ (1/3 of the maximum magnetization) both for the triangular system and the kagomé system which signals a magnetization plateau, and a change of behavior between $S_{\text{max}} - 1$ and $S_{\text{max}}$ which is the manifestation of localized states in the nearly magnetized KHA [24].

The low-energy effective behavior described by eq. (2) is expected from first principles for a uniaxial ordered antiferromagnet [25], as is the case of the SHA and HHA on the square and hexagonal lattices [26]. In such a case, eq. (2) describes the free precession of the order parameter of the magnet (which has the symmetry of a rigid rotator) in an SU(2) invariant environment. A slightly more complicated behavior is expected for a biaxial magnet like the THA (on the triangular lattice): the order parameter
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has now the symmetry of a quantum top and two different susceptibilities are needed to describe its free motion [27]. It can indeed be observed in fig. 1 that the fit of the low-lying levels of the THA with eq. (2) is not as good as in the case of collinear Néel order (SHA et HHA). In fact the Pearson product-moment correlation coefficient of the fit of the 6 first levels (S_{tot} = 0 \cdots 5) of the exact spectra to eq. (2) is 1 for HHA, respectively 0.99997 for SHA, 0.99996 for KHA and only 0.99849 for THA.

In addition to the above discussed data, fig. 1 (top) exhibits the low lying levels of the spectrum of the CHA (a two dimensional pyrochlore lattice). This system is a good example of a Valence Bond Crystal with a large gap of the order of 0.7 [28,29]. The S-dependence of the CHA energy is clearly different from that of the previous models. For high values of the total spin CHA energy displays the standard semi-classical quadratic behavior described in eq. (2). But for S \leq 4 the energy of the magnet deviates clearly downwards from this quadratic behavior, and appears dominated by a linear in S behavior (fig. 1 (bottom))^2. The following heuristic law is then a good first order approximation of the small S behavior:3

\[ E_N(S) = E_N(0) + \Delta_N S, \]

where \( E_N(S) \) is the total energy of the ground state of the N-sites sample in the S channel. In a magnetic field this equation becomes

\[ E_N(S) = E_N(0) + \Delta_N S^2 - g_\mu_B H S^2 + \cdots, \]

where \( \Delta_N \), the spin-gap (\( \sim 0.7 \) in the case of the CHA) is a measure of the critical field \( H_c \) needed to observe a magnetic answer of the system (in units where \( g_\mu_B = 1 \)). At that point we can do the contact between the usual point of view in term of one-particle excitations and our many body approach. The so-called Bose-Einstein condensation of spin excitations at \( H_c \) translates in our approach in a collapse of a finite fraction of eigen-levels with different total S^2 when \( H = H_c = \Delta \). In our 36-site CHA sample, the linear-in-S behavior of eq. (4) implies that the eigen-levels with total spin components \( S^2 = 1, 2, 3 \) (which represent, respectively, states with 1, 2, 3 bosons) become degenerate when \( H \) reaches \( H_c \).

The perfect adequation of eq. (2) for the description of the low-lying levels of the KHA (for all sizes up to \( N = 36 \)) opens a question: is it legitimate to ascribe the finite-size gap of the spectrum of this sample to a true spin-gap as it can be perfectly described in an alternate, ultimately gapless picture? Either a saturation of the gap with finite size^4 or a magnetization curve, as described in eq. (4), should be observed to ascertain a true thermodynamic spin-gap. With the knowledge of the exact spectra of the KHA up to \( N = 36 \), it seems impossible to decide if KHA is a gapless system or a gapped one with an extremely small gap.

In the hypothesis of a gapless system with linear in \( k \) low-lying excitations, \( e(0) \) (respectively \( \chi \)) would scale as \( N^{-3/2} \) (respectively \( N^{-1/2} \)). The fits in fig. 2 are not contradictory with this hypothesis. If this picture is valid up to the thermodynamic limit, the extrapolated value of the spin susceptibility is \( \lim_{N \to \infty} (\chi_N) \sim 0.99 \pm 0.015 \). Within the error bars (which are rather large), the extrapolated value of the susceptibility compares favorably to the bulk susceptibility measurement of the Herbertsmithite [6] and is consistent with the high-temperature series data [30].

The large finite-size spin-gap appears in this point of view as a simple manifestation of the finite value of the uniform spin susceptibility and of the spin quantization. In this aspect the behavior of KHA is not very different from that of SHA, HHA or THA. But, as we will show below, the situation is quite different for the intra-S-channel excitations, which do not suffer of the same limitation due to quantization and allow the exploration of a much lower-energy scale.

Néel-ordered magnets, energy scale of the excitations. – In the two first situations SHA and THA (figs. 3(a) and (b), respectively), all the low-lying levels in each S sector above the ground state describe the spin-waves. The analysis of the \( S = 1 \) sector is specially simple^5: one easily recognizes the soft mode(s) of the SHA (point X of the first Brillouin Zone (BZ) fig. 3(a)) and of the THA (points \( \Gamma \) and \( K \) of the BZ of the THA fig. 3(b)). For the THA the dispersion curve can directly be compared to the most sophisticated series calculation [31]. The agreement is qualitatively total (flat zone, local minima) and quantitatively rather good. In spite of the small size of the samples, the order of magnitude of the spin-wave bandwidth (2 for the SHA and only 1 for the THA) as well as the general behavior of the dispersion curve are in correct agreement with thermodynamic calculations. Note that in the typical bandwidth of one-magnon excitations, the exact spectra in the \( S = 1 \) sector exactly display one eigen-level per wave number in the SHA case and approximately the same number in the triangular case (crosses in figs. 3(a) and (b)). In that latter case the bi-axial nature of the magnet brings some minor complications.)

\(^2\)The downwards deviation is the manifestation that resonances which stabilize the quantum magnets are larger and larger when S decreases, as expected.

\(^3\)Indeed, specifically in one dimension, this mean-field approach and eq. (4) is certainly a bit naive. Non-analytic terms dominate the behavior of the spin susceptibility just above the spin-gap, but the overall thermodynamic behavior is correct.

\(^4\)In fact neither the Contractor Renormalization approach [11] (CORE), nor the Density Matrix Renormalisation Group [16] (DMRG) do observe a real saturation in the gap with increasing sizes up to 81 (respectively 108). In that last approach this saturation is only obtained for quasi–uni-dimensional samples with 3 or 4 legs. But the same saturation has not been exhibited for larger bands. The MERA renormalization [23] approach may be promising but has not given up to now an estimate of the gaps.

\(^5\)The analysis in the \( S = 0 \) sector is more involved, selection rules on the coupling of angular momenta imply that the excited levels in the \( S = 0 \) sectors are uniquely two-magnons excitations when the order parameter is a rigid rotator, whereas for a top —as is the case of the THA— there are both one- and two-magnons excitations.
Specificity of the kagomé antiferromagnet. — By comparison the spectrum of excitations of the KHA fig. 4 is qualitatively and quantitatively totally different from the two previous systems. It exhibits a considerable density of low-lying excitations, seemingly continuous above the ground state in each $S$ sector. At same sizes, the finite-size gaps inside an $S$ sector are much smaller. A molecule of 36 spins with Heisenberg nearest-neighbor couplings and the coordination symmetry of a square would have internal Bohr frequencies for $\Delta S = 0$ transitions in the $S = 1$ sector of the order of 1.5. This number goes down to 0.5 for the triangular coordination and to $3 \cdot 10^{-3}$ for the kagomé network. This frequency scale at this size is only slightly larger than the one observed in Herbertsmithite — in which a fully developed dynamics down to temperatures of the order of $2.5 \cdot 10^{-4}$ [4,7] has been observed. The similar low energy of the $\Delta S = 0$ transitions in the $S = 0$ sector equally indicate fluctuation of singlets down to very low temperatures. A measurement of the energy scale inside
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The density of low-lying states for the kagomé sample is a few hundred times larger than in the two other magnets of the same size. The convergence of the Lanczos process is thus more tedious and is only achieved below the vertical red lines. In the range of non-converged levels, the displayed spectrum is less dense than in reality.

Each \(S\) sector on larger samples would be of the utmost interest as it might match the experimental scale observed in Herbertsmithite for \(N \sim 108\). Presently impossible with ED it should be reachable in DMRG [16] or MERA [23]. With some caveat, analysis in the dimer basis could also be interesting [32,33].

Proximity to a quantum critical point?. – The huge density of low-lying states in each \(S\) sector, and its evolution with the system size [10] which is the specificity of KHA, can be interpreted as the absence of an intrinsic energy scale (at least in these small samples). This opens the question of the criticality [13,14,34]. We show in fig. 5 the evolution of the low-lying levels in the \(S = 0\) (respectively \(S = 1/2\)) sector of the \(N = 36\) (respectively \(N = 27\)) sample under the action of an antiferromagnetic second neighbor coupling. Under the action of this perturbation, a gap opens linearly just above the ground state. The levels that would embody a VBC symmetry breaking [12,15,35] are immediately destabilized as well as all other excited states. The ground state (respectively in the \(S = 1/2\) and \(S = 0\) channels) has all the symmetries expected for the g.s. of the \(q = 0\) Néel order. At this scale and inside each \(S\) sector KAH looks like as it was critical\(^6\). In refs. [9,37], we concluded that a non-zero critical coupling (either a second neighbor interaction [9], or Dzyaloshinski-Moriya [37]) was needed to achieve full \(q = 0\) Néel order. These two determinations are implicitly dependent on the large energy scale set by the finite-size gap (\(\Delta_{N=36} = 0.2\)). At the light of the present study on can at best ascertain that the values obtained in these papers are certainly upper bounds. The present study cannot either ascertain that the system is critical. In some sense it may be instructive to compare the finite-size situation to a non-zero temperature regime: in the vicinity of a \(T = 0\) critical point, it is widely accepted that there exists a \(T \neq 0\) quantum critical regime which extends on a finite range of couplings around the quantum

\(^6\)The same situation was observed when destroying the three sublattice long-range order on a triangular lattice by a cyclic 4-spin exchange [36].
critical point. It seems quite reasonable to argue that the behavior observed in refs. [9,37] around the pure Heisenberg kagomé point looks like a quantum critical regime. Such an observation does not preclude at lower temperatures/larger sizes a subsequent crystallization of the pure KAH in a very large Valence Bond Crystal or a small gap RVB phase, but it gives some support to the idea that the system could be near a critical point, and that in a non-negligible range of energy, time and length scale it could behave as “quasi-critical”. Such a point of view is also supported by the conclusion of a finite-temperature DMFT calculation done some years ago [38].

In this paper we have explicitly shown that the KHA spin-gap measured in numerics for sizes up to 36, could be an extrinsic property: on such small samples it is impossible to distinguish between a gapless system and an system with a very small gap. We have shown that the intra-S-channel spectra could signal the proximity of a quantum critical point. We suggest that polarized inelastic neutron scattering on Herbertsmithite at temperature low enough but larger than the residual Dzyaloshinskii-Moriya perturbation would bring an interesting insight on the spin-1/2 compound, with a possible comparison to SrCrGaO$_3$ [39].

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