Planar pyrochlore: a strong-coupling analysis

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Recent investigations of the two-dimensional spin-1/2 checkerboard lattice favor a valence bond crystal with long range quadrumer order [J.-B. Fouet *et al.*, preprint cond-mat/0108070]. Starting from the limit of isolated quadrumers, we perform a complementary analysis of the evolution of the spectrum as a function of the inter quadrumer coupling j using both, exact diagonalization (ED) and series expansion (SE) by continuous unitary transformation. We compute (i) the ground state energy, (ii) the elementary triplet excitations, (iii) singlet excitations on finite systems and find very good agreement between SE and ED. In the thermodynamic limit we find a ground state energy substantially lower than documented in the literature. The elementary triplet excitation is shown to be gapped and almost dispersionless, whereas the singlet sector contains strongly dispersive modes. Evidence is presented for the low energy singlet excitations in the spin gap in the vicinity of $j = 1$ to result from a large downward renormalization of local high-energy states.

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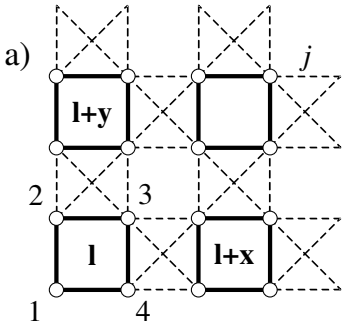
Quantum-magnetism in low dimensions has received considerable attention recently due to the discovery of numerous materials with spin-1/2 moments arranged in chain, ladder, and depleted planar structures. Many of these systems exhibit strongly gapped excitation spectra induced by various types of magnetic dimerization and discrete symmetry breakings. Yet, low energy collective spin dynamics may emerge from materials with strong geometrical frustration leading to ground states with near macroscopic degeneracy and possibly even zero temperature entropy[1,2]. In this respect quantum spin systems on the kagomé, and more recently on the pyrochlore lattice are of particular interest. While their classical counterparts have been studied in considerable detail the role of quantum fluctuations in such systems is an open issue. For the spin-1/2 kagomé lattice gapless singlet excitations and a high density of singlet states in the singlet-triplet gap have been established on finite systems by ED[2]. Similar analysis of the pyrochlore quantum-magnet is severely constrained by its three-dimensional structure. Therefore, and as a first step, several investigations[3–6] have focussed on the planar projection of the pyrochlore quantum-magnet, *i.e.* the spin-1/2 checkerboard lattice of Fig. 1a) for the case of $j = 1$.

In particular, ED at $j = 1$ has resulted in a sizeable spin gap and a large number of in-gap singlet states[3,6]. Moreover, recent ED[6] has given strong evidence in favor of a valence bond crystal (VBC) ground state with long range order in the $S = 0$ quadrumers shown in Fig. 1a). While this has been concluded from the quadrumer correlation at $j = 1$ it emphasizes the need for a perturbative investigation of the checkerboard magnet starting from the limit $j \to 0$ in Fig. 1a). Low-order perturbation theory has been performed previously for the checkerboard magnet[4] however with the unperturbed Hamiltonian chosen at variance with the VBC found in the ED[6]. Therefore, the purpose of this work is to explore the evolution of the spin spectrum of the checkerboard spin-1/2 magnet as a function of the inter quadrumer coupling j, contrasting exact diagonalization (ED) against high-order series expansion (SE).



b)

E	S	$q_{\mathbf{l}}$
1	2	3
0	$0 \oplus 1 \oplus 1$	2
−1	1	1
−2	0	0

FIG. 1: a) The checkerboard lattice. Spin-1/2 moments are located on the open circles. Full (dashed) lines label the quadrumer bonds (bonds corresponding to the expansion parameter j). b) Energy (E), spin (S), and local $q_{\mathbf{l}}$ quantum-number of a single quadrumer.

Written in a form adapted to the VBC symmetry breaking and normalized to an overall unit of energy the Hamiltonian of the checkerboard magnet reads

$$H = \sum_{\mathbf{l}} \Big[\frac{1}{2}(\mathbf{P}^2_{1234\mathbf{l}} - \mathbf{P}^2_{13\mathbf{l}} - \mathbf{P}^2_{24\mathbf{l}}) + j\,(\mathbf{P}_{34\mathbf{l}}\mathbf{P}_{12\mathbf{l}+\mathbf{x}} + \mathbf{P}_{23\mathbf{l}}\mathbf{P}_{14\mathbf{l}+\mathbf{y}})\Big] \quad (1)$$

$$= H_0 + \; j \sum_{n=-N}^{N} T_n \quad (2)$$

where $\mathbf{P}_{i\ldots j\mathbf{l}} = \mathbf{S}_{i\mathbf{l}} + \ldots + \mathbf{S}_{j\mathbf{l}}$ and $\mathbf{S}_{i\mathbf{l}}$ refers to spin-1/2 operators residing on the vertices $i = 1 \ldots 4$ of the quadrumer at site $\mathbf{l}$, c.f. Fig. 1a). H_0 is the sum over local quadrumer Hamiltonians the spectrum of which, c.f. Fig. 1b), consists of four equidistant levels which can be labeled by spin S and the number of local energy quanta $q_{\mathbf{l}}$. H_0 displays an equidistant ladder spectrum labeled by $Q = \sum_{\mathbf{l}} q_{\mathbf{l}}$. The $Q = 0$ sector is the *unperturbed* ground

state $|0\rangle$ of H_0, which is the VBC of quadrumer-singlets. The $Q = 1$-sector contains local $S = 1$ single-particle excitations of the VBC with $q_{\mathbf{l}} = 1$, where $\mathbf{l}$ runs over the lattice. At $Q = 2$ the spectrum of H_0 has a total $S = 0, 1$, or 2 and is of multiparticle nature. For $S = 0$ at $Q = 2$ it consists of one-particle singlets with $q_{\mathbf{l}} = 2$ and two-particle singlets constructed from triplets with $q_{\mathbf{l}} = q_{\mathbf{m}} = 1$ and $\mathbf{l} \neq \mathbf{m}$. Consequently the perturbing terms $\propto j$ in (1) can be written as a sum of operators T_n which *nonlocally* create(destroy) $n \geq 0$ ($n < 0$) quanta within the ladder spectrum of H_0.

It has been shown recently[7–9] that problems of type (2) allow for high-order SE using a continuous unitary transformation generated by the flow equation method of Wegner[10]. The unitarily rotated effective Hamiltonian H_{eff} reads[7,9]

$$H_{\text{eff}} = H_0 + \sum_{n=1}^{\infty} j^n \sum_{\substack{|\mathbf{m}| = n \\ M(\mathbf{m})=0}} C(\mathbf{m})\, T_{m_1} T_{m_2} \dots T_{m_n} \quad (3)$$

where $\mathbf{m} = (m_1 \dots m_n)$ is an $n = |\mathbf{m}|$-tuple of integers, each in a range of $m_i \in \{0, \pm 1, \dots, \pm N\}$. In contrast to H of (1), H_{eff} conserves the total number of quanta Q. This is evident from the constraint $M(\mathbf{m}) = \sum_{i=1}^{n} m_i = 0$. The amplitudes $C(\mathbf{m})$ are rational numbers computed from the flow equation method[7,9].

Previous applications of this method to spin models have been confined to dimer systems[9] where $N \leq 2$. Here we report on its first application using a quadrumer basis where $N \leq 6$. Explicit tabulation[11] of the T_n shows that for the checkerboard magnet $N = 4$. Because the quadrumer basis consists of 16(4) states(spins) rather than only 4(2) states(spins) as for dimer systems, the maximum order achievable by a quadrumer expansion is lower in general than for a dimer expansion. Here we present results up to $O(7)$ for the ground state energy and the elementary triplet as well as up to $O(6)$ for the $Q = 2$ singlets. The $C(\mathbf{m})$-table is available on request[11].

Q-conservation leads to a ground state energy of

$$E_g = \langle 0 | H_{\text{eff}} | 0 \rangle \,. \quad (4)$$

Evaluating this matrix element on clusters with periodic boundary conditions, sufficiently large not to allow for wrap around at graph-length n one can obtain SEs valid to $O(n)$ in the thermodynamic limit, *i.e.* for systems of infinite size.

Q-number conservation guarantees the $Q = 1$-triplets to remain genuine one-particle states. *A priori* single-particle states from sectors with $Q > 1$ will not only disperse via H_{eff}, but can decay into multi-particle states. *A posteriori* however such decay may happen only at high order in j leaving the excitations almost true one-particle states. The dispersion of the single-particle excitations is

$$E_\mu(\mathbf{k}) = \sum_{lm} t_{\mu,lm}\, e^{i(k_x l + k_y m)} \quad (5)$$

where $t_{\mu,lm} = \langle \mu, lm | H_{\text{eff}} | \mu, 00 \rangle - \delta_{lm,00} E_g^{obc}$ are hopping matrix elements from site $(0,0)$ to site (l,m) for a quadrumer excitation μ inserted into the unperturbed ground state. For the thermodynamic limit $t_{\mu,lm}$ has to be evaluated on clusters with open boundary conditions large enough to embed all linked paths of length n connecting sites $(0,0)$ to (l,m) at $O(n)$ of the perturbation. $E_g^{obc} = \langle 0 | H_{\text{eff}} | 0 \rangle$ on the $t_{\mu,00}$-cluster.

To assess the convergence of the SEs, as well as to identify the remaining excitation spectrum, we will contrast the SEs with ED of finite systems. For this purpose eqns. (4,5) apply equally well, with however the cluster topologies set by the finite systems. We emphasize, that for some of the SEs generated by (3) on finite systems we were also able to compute standard SEs using a completely different code[12]. In all cases we found both SEs to agree which serves as an independent check.

Fig. 2 summarizes our results for the ground state energy *per spin* $e_g(j)$, the spin gap $E_T(j)$, and the spectrum of the low lying singlets. The figure compares the SEs with our ED data on $2(3) \times 2 \cdot 4$ systems, *i.e.* 16(24) spins, as a function of j, as well as available ED data[6] on $3 \times 3 \cdot 4$ sites, *i.e.* 36 spins, at $j = 1$. SEs are shown both for the *identical* system sizes and also for the thermodynamic limit to $O(j^7)$ for e_g and E_T as well as to $O(j^6)$ for the $Q = 2$-singlets. All SE data refers to the actual series, no Padé continuations have been applied.

First we focus on the ground state energy in Fig. 2a). In the thermodynamic limit we find

$$\begin{aligned} e_g^{\infty}(j) = \; & -\frac{1}{2} - \frac{j^2}{96} - \frac{j^3}{768} - \frac{451\, j^4}{387072} - \frac{127\, j^5}{338688} \\ & - \frac{865153\, j^6}{3072577536} - \frac{24293225710331\, j^7}{178396309802188800} \end{aligned} \quad (6)$$

which is lower than the decoupled quadrumer-value of $e_g(0) = -1/2$ for all $j \leq 1$ and yields $e_g^{\infty}(1) \approx -0.513677$. This is also lower than the value of $e_g^{\infty}(1) \approx -0.49622$ which is *above* the bare quadrumer limit and has been reported from a different third-order SE starting from decoupled *tetrahedra*[4]. While the SEs are not related to a variational principle the preceding is consistent with the decoupled quadrumers to incorporate the proper ground state correlations for $j \leq 1$ rather than the tetrahedral limit. This is confirmed on the finite systems in Fig. 2a), where practically quantitative agreement is found for $j \lesssim 0.7$ between ED and SE for $2(3) \times 2 \cdot 4$ sites. At $j \sim 1$ very small deviations occur, which however decrease with system size. They are $\sim 2\%$ on the 16(24)-spin lattices and only 0.7% on the $3 \times 3 \cdot 4$ site system[6]. Regarding the SEs this agreement is remarkable also, since the high-order decrease of SE-coefficients[11] tends to be slowed down on finite geometries. On the $2(3) \times 2 \cdot 4$ lattice we observe a kink in the $e_g(j)$ as obtained from the ED at $j \approx 1$ which suggests a change of the nature of the ground state on these systems in the vicinity of $j = 1$. This is probably a special property of the 16(24)-spin lattices[6].

Next we discuss the elementary triplet excitations.

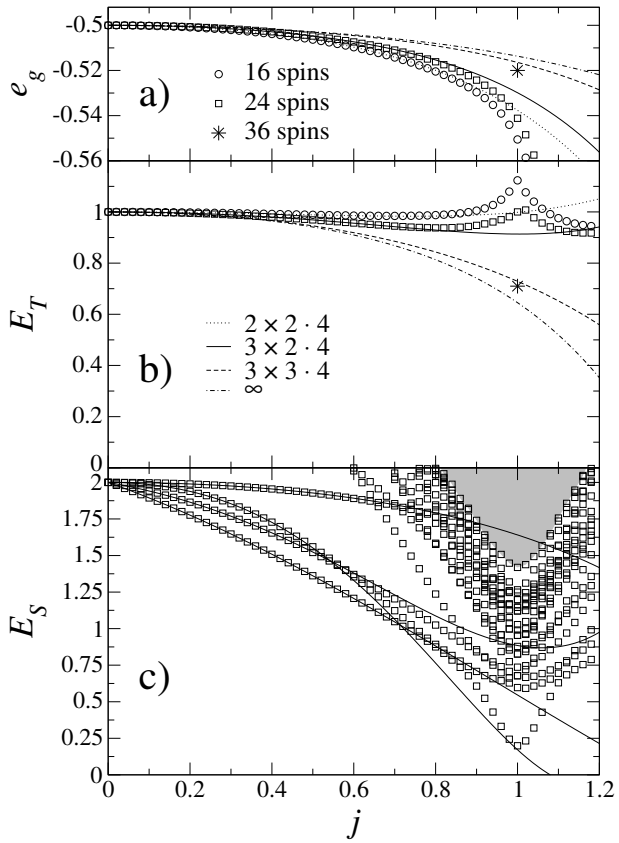


FIG. 2: a) Ground state energy per spin, b) spin gap and c) singlet excitations with $\mathbf{k} = \mathbf{0}$ as a function of j. Lines correspond to the series, symbols to exact diagonalization. The numerical data at $j = 1$ for 36 spins is taken from Ref. 6. The grey shaded area in panel c) indicates that our numerical data is not complete in this region.

The SE to $O(j^7)$ in the thermodynamic limit for the triplet dispersion reads

$$E_T^{\infty}(\mathbf{k},j) = 1 - \frac{7\,j^2}{36} - \frac{41\,j^3}{864} - \frac{329887\,j^4}{6531840} - \frac{580309487\,j^5}{21946982400} - \frac{16957803829\,j^6}{790091366400} - \frac{7822020675129119\,j^7}{557488468131840000} - \left(\frac{53\,j^5}{1036800} + \frac{59527\,j^6}{1306368000} + \frac{74504581093\,j^7}{948109639680000}\right)(\cos(k_x)+\cos(k_y)) + \left(\frac{1679\,j^6}{32659200} + \frac{2039741\,j^7}{438939648000}\right)\cos(k_x)\,\cos(k_y)\,. \quad (7)$$

Remarkably, the hopping amplitude is exceedingly small and sets in only at $O(j^5)$ hinting at a fairly extended polarization cloud necessary in order to allow for triplet motion[13]. From (7) the spin gap occurs at $\mathbf{k} = \mathbf{0}$.

The spin gap is shown in Fig. 2b)[14]. Again, on finite systems of $2(3)\times2\cdot4$ sites and for $j \lesssim 0.7$ the agreement between ED and $O(j^7)$-SE is very good. At $j = 1$ and for the SEs shown the absolute value of the relative difference to the numerically exact values for the spin gap improves from 11%, to 8.6%, to 2.7% on passing from 16, to 24, to 36 spins. A clear cusp is observable in $E_T(j)$ close to $j = 1$ on the $2(3)\times2\cdot4$ systems. This is closely related to the previously anticipated change in the nature of the ground state on the 16(24)-spin systems at $j \approx 1$ and is not captured by the SE. While no j-scan of E_T is available on $3\times3\cdot4$ sites it is remarkable that at $j = 1$ the SE agrees better with ED for 36 spins than for 16(24) spins and that the SE behaves qualitatively different, both on the $3\times3\cdot4$ lattice *and* in the thermodynamic limit as compared to the $2(3)\times2\cdot4$ lattices. *I.e.*, on the larger systems the gap decreases monotonously, while it increases beyond $j \gtrsim 1$ on the smaller two. This suggests an absence – or strong reduction – of the discontinuities close to $j = 1$ on larger systems improving the convergence of the SE.

Finally we discuss the low energy singlet excitations. At $j \ll 1$ they result from the $Q = 2$ sector which involves solving a two-particle problem coupled to a one-particle problem. We have found the mixing between the one- and two-particle states in the $S = 0$, $Q = 2$ sector to be very weak. In fact, on the $2\times2\cdot4$ system the mixing *vanishes* identically in the ED as well as the SE up to $O(j^7)$. For all other systems analyzed by SE including the thermodynamic limit, the mixing sets in only at $O(j^5)$. Therefore the $Q = 2$ one-particle singlet remains an almost true one-particle state even at finite j. In addition its dispersion $E^{\infty}_{S,1pt}$ up to $O(j^4)$ is determined *exactly* by (5). In the thermodynamic limit we get

$$E^{\infty}_{S,1pt}\quad(\mathbf{k},j) = 2 - \frac{5\,j^2}{12} - \frac{11\,j^3}{96} - \frac{227\,j^4}{3024} - \left(\frac{j^2}{4} + \frac{j^3}{16} + \frac{23\,j^4}{2304}\right)(\cos(k_x)+\cos(k_y)) - \frac{3\,j^4}{512}(\cos(2\,k_x) + \cos(2\,k_y)) + \frac{53\,j^4}{1152}\cos(k_x)\,\cos(k_y)\,. \quad (8)$$

In sharp contrast to (7) this low energy singlet is strongly dispersive. Its gap resides at $\mathbf{k} = \mathbf{0}$.

Fig. 2c) shows the evolution of all eigenvalues of H from (1) in the singlet sector on the $3\times2\cdot4$ system at $\mathbf{k} = \mathbf{0}$ as well as the spectrum of $H_{\rm eff}$ at $O(j^6)$ in the $S = 0$, $Q = 2$ sector. For $j \lesssim 0.7$ the agreement between ED and SE is satisfying. Similar agreement is found at the remaining $\mathbf{k}$-vectors. The lowest $S = 0$ excitation can be identified with a singly degenerate $Q = 2$ two-triplet excitation. The binding energy for this two-particle state is approximately proportional to j with a coefficient of order unity. Starting at $j \gtrsim 0.7$ a large number of singlets from higher Q-sector enter the low energy spectrum. At $j = 1$ several of these singlets reside in the spin gap. This is consistent with ED at $j = 1$ in Refs. 3,6. The singlet gap occurs at $\mathbf{k} = \mathbf{0}$. Preliminary analysis indicates that the lowest singlets have a substantial overlap with the $Q = 4$ sector. The numerical spectrum in Fig. 2c) is very suggestive of a softening in the singlet sector in the vicinity of $j = 1$. If this is a feature particular only to the finite systems is an issue in clear need of further study. Close to $j = 0.7$ a near level crossing occurs, with the lowest state of the SE for $j \gtrsim 0.7$ corresponding to a

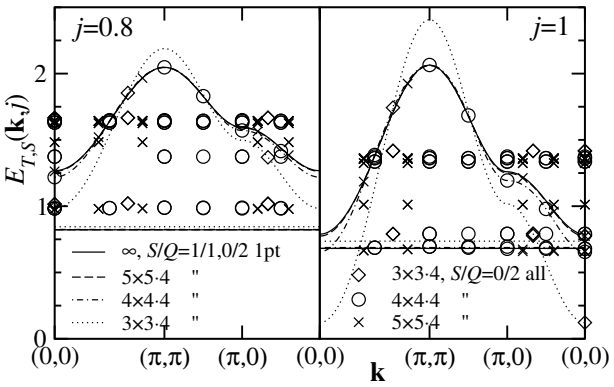


FIG. 3: System size dependence of the dispersion from SE for $Q = 1$, $S = 1$ and $Q = 2$, $S = 0$. Lines: $E_T(\mathbf{k}, j)$ (almost nondispersive) and $E_{S,1pt}(\mathbf{k}, j)$ (dispersive). Symbols: all eigenvalues of the $Q = 2$, $S = 0$ sector.

continuation of the $Q = 2$ one-particle singlet.

Finally we speculate on the finite size dependence to be expected for systems presently inaccessible to ED focusing on the $Q = 1$ triplet- and the $Q = 2$, $S = 0$ singlet-sector by using SE instead. In Fig. 3 we show the spectrum of $H_{\rm eff}$ for these two sectors at $O(j^5)$ for $j = 0.8$ and 1.0 along a path in the Brillouin zone (BZ) on four systems ranging from $N = 3 \times 3 \cdot 4$ sites up to the thermodynamic limit. As we have pointed out SEs will converge more rapidly on larger systems. Therefore, at $j = 0.8$ the SE results of Fig. 3 will be rather well converged while at $j = 1$ they will be qualitatively correct. Fig. 3 includes the one-particle triplet and singlet dispersions, $E_T(\mathbf{k}, j)$ and $E_{S,1pt}(\mathbf{k}, j)$ with $\mathbf{k}$ varying continuously along the path in the BZ. For the latter curve $E_{S,1pt}(\mathbf{k}, j)$ has been evaluated at $O(j^5)$. Moreover in the thermodynamic limit and for $Q = 2$ only $E_{S,1pt}(\mathbf{k}, j)$ is depicted. First Fig. 3 illustrates the difference in strength of the dispersion between the elementary triplet and almost one-particle singlet from the $Q = 2$-sector. Second the figure shows that both, the elementary triplets and the corresponding, almost two-particle singlets with $Q = 2$ display only a small downward shift with increasing system size. However, the spectrum of the almost one-particle singlet with $Q = 2$ experiences a strong reduction of its band-width including an upward shift of its gap. In particular, at $j = 1$ the SE suggests that the $Q = 2$-singlets are above the spin gap for sufficiently large systems and that only singlets from sectors with $Q > 2$ might reside in the spin gap[15]. This underlines the need, both, for larger systems in ED and larger Q-values in SE studies of the singlet sector.

In conclusion, by comparing ED and SE we find the decoupled quadrumer-limit to be an excellent starting point for a perturbative treatment of the spin-1/2 checkerboard magnet even at $j \approx 1$. The remarkably fast convergence of the series for the ground state and the elementary triplet excitation up to at least $j = 1$ indicates that the points $j = 0$ and 1 belong to the same phase. This is consistent with the strong VBC correlations observed[6] at $j = 1$. We have further shown the elementary triplets of the checkerboard magnet to be almost nondispersive. Up to intermediate j the elementary singlet excitations are almost nondispersive two-particle states from the $Q = 2$ sector residing above the spin gap. On finite systems and in the vicinity of $j = 1$, we find a strong downward shift of singlets from higher Q sectors, entering the spin gap. In future work it would be interesting to study these higher Q-sectors by SE and to obtain j-dependent ED spectra on larger systems, extending in particular the 36-site results of Ref. 6 to $j \neq 1$.

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[14] We have performed a similar analysis for all distinct $\mathbf{k}$-sectors on the $2(3) \times 2 \cdot 4$ systems. We note that on the $2 \times 2 \cdot 4$ lattice $E_T^{2\times 2\cdot 4}(\mathbf{k}, j)$ is completely $\mathbf{k}$-independent, *both*, in ED and SE to $O(j^7)$.
[15] Since the VBC picture implies a two-fold ground state degeneracy[6] at $j = 1$, we expect at least one low-lying singlet excitation to come from these higher Q-sectors. Due to Q-conservation it is possible that the mixing of this state with the ground state as well as the convergence problems usually associated with this can be avoided in the present case.