Series expansion analysis of a tetrahedral cluster spin chain

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Using series expansion by continuous unitary transformations we study the magnetic properties of a frustrated tetrahedral spin–\(\frac{1}{2}\) chain. Starting from the limit of isolated tetrahedra we analyze the evolution of the ground state energy and the elementary triplet dispersion as a function of the inter-tetrahedral coupling. The quantum phase diagram is evaluated and is shown to incorporate a singlet product, a dimer, and a Haldane phase. Comparison of our results with those from several other techniques, such as density matrix renormalization group, exact diagonalization and bond-operator theory are provided and convincing agreement is found.

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INTRODUCTION

Geometric frustration is a central issue in present days research on quantum magnets. In particular, materials with spin-tetrahedra as building blocks are under scrutiny, since they bear the potential of exotic magnetic properties \([1]\). Many investigations have focused on dense tetrahedral magnets, e.g. on pyrochlore lattices \([1]\). Quantum magnets with spatially more separated spin-tetrahedra have been considered far less exhaustive. The isostructural tellurate compounds Cu\(_2\)Te\(_2\)O\(_5\)X\(_2\) (X=Br or Cl) \([2,3]\) are a prominent example of such ‘diluted’ systems. They contain disjoint tetrahedral clusters of Cu\(^{2+}\) with \(S=1/2\) \([2]\). The tetrahedra display two longer and four shorter edges. They form chains along the \(c\) direction and are separated by lone-pair cations in the \(a, b\) plane \([2]\). Analysis of the susceptibility \([2,4,5]\) is consistent with a strongly frustrated antiferromagnetic intra-tetrahedral exchange of \(J_1=47.5(40.7)\text{K}\) along the shorter edges and a ratio of \(J_2/J_1=0.7(1)\) between the long and the short edges for X=Br(Cl). Both systems show incommensurate ordering with strongly reduced moments \([6]\) below a transition temperature of \(T_c=11.4(18.2)\text{K}\) for X=Br(Cl). This \(T_c\) is consistent with bulk thermodynamics \([2,3,4,5]\). Observation of longitudinal magnons suggests that the Br-system could be close to quantum criticality \([4,6]\).

Despite the low-temperature ordering of the tellurates, the effects of inter-tetrahedral coupling at various dimensionality and on an energy scale of the intra-tetrahedral exchange are open issues in such magnets. In the tellurates dispersive features have been observed in inelastic neutron scattering on polycrystals \([3]\). Two-magnon Raman scattering provides evidence for inter-tetrahedral dispersion along the \(c\)-axis only \([5]\). Considerations of the electronic structure however, suggested that \(a, b\)-planar exchange via O-Te-O bonds is important also \([10,11]\).

Theoretically, inter-tetrahedral coupling in diluted tetrahedral magnets has been analyzed in terms of one-dimensional (1D) tetrahedral spin chains using bond-operator methods, exact diagonalization \([12]\) and effective Hamiltonians \([12]\). The chains have been found to allow for a singlet, a dimer, and a Haldane phase with low-energy singlet excitations close to the dimer-to-singlet transition which is of first order. In 3D molecular field theories \([4,5]\) have been used to model the magnetic transition in the tellurates. Moreover series expansion (SE) up to 4th order in various 3D inter-tetrahedral couplings has been carried out \([12]\) resulting in a rich quantum phase diagram with several competing states. Motivated by this situation, the aim of the present work is to extend and improve the analysis of 1D tetrahedral chains carried out in ref. \([12]\) by employing the SE method of ref. \([13]\), however in 1D only, which will enable us obtain results to higher orders.

Fig.1 depicts the tetrahedral spin–\(\frac{1}{2}\) chain introduced in refs. \([12,13]\). The tetrahedra correspond to the sites 1–4 with couplings \(J_1\) and \(J_2=\alpha J_1\). Inter-tetrahedral exchange occurs through \(J_3=bJ_1\). The Hilbert space of a single tetrahedron consists of 16 states, i.e., two singlets \(S_1, 2\), three triplets \(T_1, 2, 3\) and one quintet \(Q\) whose energies are listed in Table II. This level scheme implies that a singlet resides within the singlet-triplet gap of the tetrahedron for \(1/2<\alpha<2\). The Hamiltonian of the chain can be written in terms of the total edge-spin operators \(P_{1,2,3}\) where \(P_{1,2}\) denotes a spin–\(1/2\) at vertex \(i\) on the tetrahedron at site \(l\). For the \(P_i\) quantum numbers refer to Table II.

\[
H = \frac{\alpha}{2} \sum_{i=1}^{L/2} \left[ P_{1,i} P_{2,i} + \frac{1}{2} ( P_{1,i}^2 + P_{2,i}^2 - 3 ) + bP_{2,i} P_{1, i+1} \right].
\]

Here \(L\) is the number of rungs (pairs of sites coupled by \(J_2=\alpha J_1\)). The Hamiltonian commutes with the edge spin, i.e. \([H, P_{1,2}^\mu = 0; \forall i, l; \mu = 1, 2]\). Hence, the Hilbert space decomposes into sectors of fixed distributions of locally conserved edge-spin eigenvalues \(P_{1,2}\), each corresponding to a sequences of spin–1 chain-segments \((P_{1,2} = 1)\) intermitted by chain-segments of localized singlets \((P_{1,2} = 0)\). It has been shown in ref. \([12]\) that the ground state of Eq. I occurs only within the homogeneous sector of purely \(S_2\) states or in the \(P_{1,2} = 1, \forall i, l\) sector.
sector. I.e., inhomogeneous phases consisting of both $P_{11} = 0$ and $P_{11} = 1$ sites will not be ground states. Keeping $b$ fixed and for $a \to \infty$, the ground state must result from local $S_2$ type of states. This is a trivial, decoupled singlet product with singlets on each rung. The ground state energy per tetrahedron in this phase is independent of $b$ and it is given by: $e_a S_2 = -3a/2$. In the opposite limit $a \to 0$ the ground state is in the $P_{11} = 1$, $\forall i, l$ sector. There is a line of a first order quantum phase transition in the $(a,b)$-plane that switches between both ground states, beginning at the non-interacting point $a = 1, b = 0$. In the next Sections we will determine this line by SE.

In the $P_{11} = 1$, $\forall i, l$ sector the tetrahedral chain is equivalent to that of a dimerized spin$-1/2$ chain with $L$ sites

$$
\frac{H}{J_1} = \sum_{i=1}^{L} [S_{2i-1} S_{2i} + b S_{2i} S_{2i+1}] + \frac{a}{4} L, \tag{2}
$$

where the edge-spin operators $P_{1,i} = S_{2i-1}$ and $P_{2,i} = S_{2i}$ have been relabelled in terms of spin$-1$ operators $S_i$. For $b = 0$, the chain decomposes in isolated dimers and for $b = 1$ it is the homogeneous spin$-1$ chain. In both cases the system has a spin gap. Trivially in the former one and according to Haldane [15, 16] in the latter. However, for a particular $b_c$ the gap closes, defining a second order quantum phase transition between the dimer phase ($b < b_c$) and the Haldane phase ($b > b_c$). This transition will be analyzed in the next Sections by calculating the evolution of the gap in terms of the perturbative parameter $b$ using SE around the non-interacting point ($b = 0$). Our results will be compared with those obtained by other methods.

| $P_1$ | $P_2$ | $E/J_1$ |
|-------|-------|---------|
| $S_1$ | 1     | $-2 + a/2$ |
| $S_2$ | 0     | $-3a/2$ |
| $T_1$ | 1     | $-1 + a/2$ |
| $T_{2,3}$ | 0.1 | $-a/2$ |
| $Q$   | 1     | $1 + a/2$ |

TABLE I: Eigenstates of a single tetrahedron: singlets ($S_1, S_2$); triplets ($T_1, T_{2,3}$) and Quintet ($Q$). Each state is labelled by the $P_{1,2}$ edge-spin quantum numbers and the energy $E/J_1$. Site index $l$ suppressed.

SERIES EXPANSION BY CONTINUOUS UNITARY TRANSFORMATION

The Hamiltonian of the tetrahedral chain (Eq. (1)) can be written as

$$
\frac{H}{J_1} = H_0 + a T_{a0} + b \sum_{n=-N}^{N} T_{bn}. \tag{3}
$$

$H_0$ is the sum over local tetrahedral Hamiltonians at $a = 0$. Their spectra consist of four equidistant energy levels $E/J_1 = -2, -1, 0, 1$ (see Table I). With these levels we associate a number $q_l$ of local energy quanta $q_l = 0, \ldots, 3$. Together with $P_{1,2}$, this characterizes the basis. $H_0$ has an equidistant ladder spectrum labelled by $Q = \sum_l q_l$. $Q = 0$ refers to the unperturbed ground state of $H_0$: $|0\rangle \equiv |S_{1,1} \ldots S_{1,L/2}\rangle$, i.e. an $S_1$ singlet product. The $Q = 1$ sector of $H_0$ consists of linear combinations of local $T_{1,j}$ triplet excitations $|t_j\rangle \equiv |S_{1,1} \ldots T_{1,j} \ldots S_{1,L/2}\rangle$, with a $T_{1,j}$ triplet on the tetrahedron at site $j$. $T_{a0}$ refers to the sum over the local terms proportional to $a$ in Eq. (1). By construction this term is diagonal in the basis of $H_0$. The third term in Eq. (3) refers to the inter-tetrahedral coupling via $b$. The operators $T_{bn}$ non-locally create (destroy) $n \geq 0$ quanta within the ladder spectrum of $H_0$. For our model $N \leq N_{\text{max}} \equiv 6$, in principle. Explicit calculation of the $T_{bn}$ however shows that $N \leq 4$ [17].

Note that by a shift of one half of the unit cell, i.e. $P_{2i(1+1)} \rightarrow P_{1i (2l)}$ Eq. (1) is symmetric under the operation $(J_1, a, b) \rightarrow (J_1 b, a/b, 1/b)$. Therefore, in order to cover the complete parameter space for $a, b > 0$ it is sufficient to consider the phase diagram in the range of $a \in [0, \infty]$ and $b \in [0, 1]$.

It has been shown [18, 19, 20] that models of type Eq. (1) allow for high-order SE using a continuous unitary transformation generated by the flow equation method of Wegner [21]. Adapted to our case, the mapping of Eq. (3) onto the unitarily rotated effective Hamiltonian $H_{\text{eff}}$ reads [18, 20]

$$
H_{\text{eff}} = H_0 + a T_{a0}
$$
where $\mathbf{m} = (m_1, \ldots, m_n)$ is an $n = |\mathbf{m}|$-tuple of integers, each in a range of $m_i \in \{0, \pm 1, \ldots, \pm N\}$ and $M(\mathbf{m}) \equiv \sum_{i=1}^{n} m_i$. A main advantage of this method is that in contrast to $H$ of Eq. (3), $H_{\text{eff}}$ is constructed to conserve the total number of quanta $Q$ at each order $n$. This is clear from the constraint $M(\mathbf{m}) = 0$. $Q$ conservation allows the SE of several quantities using the bare eigenstates of $H_0$ like the ground state energy and the elementary triplet dispersion. This will be done in the next Sections. The amplitudes $C(\mathbf{m})$ are rational numbers computed from the flow equation method \[18, 20\]. The $C(\mathbf{m})$ table is available on \[17\].

**GROUND STATE ENERGY AND ELEMENTARY TRIPLET DISPERSION**

Now we discuss results for the ground state energy $E_g$ and the triplet dispersion $\omega(k)$ in the dimer phase as obtained from SE with respect to the inter-tetrahedral coupling $b$. $Q$-conservation leads to

$$ E_g = \langle 0 | H_{\text{eff}} | 0 \rangle, $$

where $|0\rangle = |S_{1,1} \ldots S_{1,L/2}\rangle$ and $H_{\text{eff}}$ is the effective Hamiltonian given by Eq. (4). Wrap-around of graphs up to length $n$ will not occur if this matrix element is evaluated on chains with $n+1$ tetrahedral clusters and periodic boundary conditions (PBC). This is required by linked-cluster theorem \[22\] and leads to SE’s valid to $O(n)$ in the thermodynamic limit, i.e. for infinite-sized systems.

We have evaluated $E_g$ up to $O(7)$. The ground state energy per tetrahedron $e_g$ in the dimer phase reads

$$ e_g(a, b) = -2 + \frac{a}{2} - \frac{2 b^2}{3} - \frac{3 b^3}{6} + \frac{b^4}{108} - \frac{67 b^5}{1620} - \frac{53273 b^6}{1749600} + \frac{27311519 b^7}{503848000}. $$

(6)

The first two terms in $\omega(k)$ correspond to the non-interacting energy of the $S_1$ state as in Table \[I\]. For $a = 0$ Eq. (6) is also a SE of the ground state energy per dimer of the dimerized spin–1 chain. To the best of our knowledge an analytic expression of this has not been published previously. Fig. 2 depicts the ground state energy which is a monotonously decreasing function of the inter-tetrahedral coupling $b$. We find, that plots of $e_g(a, b)$ to $O(6)$ and $O(7)$ are indistinguishable on the scale of Fig. 3 which provides an estimate of convergence. Using $e_g(a, b)$ we will discuss the first order critical line $b_c(a_c)$ for the dimer-to-singlet transition in the next Section. Trivially, the non-interacting critical point is $b_c(1) = 0$ (Table \[I\]).

![Fig. 2: Ground state energy $e_g(a, b) - e_g(a, b = 0)$ per tetrahedron in the dimer phase.](image)

To calculate the triplet dispersion we have to consider the subspace of exactly one $T_{1, j}$-type of excitation: $|t_j\rangle = |S_{1,1} \ldots T_{1, j} \ldots S_{1, L/2}\rangle$, i.e. one-triplet $T_{1, j}$ on a tetrahedron $j$. By $Q$ conservation the number of triplets is conserved, implying that $H_{\text{eff}}$ can only translate the triplets $|t_j\rangle$

$$ H_{\text{eff}}(j) = \sum_{i} c_i |j + i\rangle. $$

(7)

Due to translational invariance of our model, spatial Fourier transformation $|k\rangle = \sqrt{4/L} \sum_j \exp(ikj) |j\rangle$, diagonalizes Eq. (7) leading to the triplet dispersion

$$ \omega(k) \equiv \langle k | H_{\text{eff}} | k \rangle - E_g $$

$$ = c_0 - E_g + \sum_{j=1}^{\infty} 2 c_j \cos(kj). $$

(8)

We have evaluated the coefficients $c_i$ up to $O(7)$. From them, Eq. (9) in the thermodynamic limit reads

$$ \omega(k, b) = \left( 1 + \frac{8 b^2}{27} + \frac{19 b^3}{27} - \frac{571 b^4}{972} + \frac{18343 b^5}{233280} - \frac{391390595851 b^6}{380936908800} + \frac{913820919969227 b^7}{511979205427200} \right) \cos(k) $$

$$ + \left( - \frac{4 b}{3} - \frac{2 b^2}{3} + \frac{26 b^3}{27} - \frac{29 b^4}{54} + \frac{145237 b^5}{98415} - \frac{4087919 b^6}{1959552} + \frac{10916063988776383 b^7}{2879883030528000} \right) \cos(2k) $$

$$ + \left( - \frac{4 b^2}{9} + \frac{2 b^3}{9} - \frac{7285 b^4}{34992} + \frac{912407 b^5}{699840} - \frac{18113617135 b^6}{10158317568} + \frac{485683037077901 b^7}{142216445952000} \right) \cos(3k) $$

$$ + \left( - \frac{71 b^5}{21864} + \frac{10065 b^6}{205952} - \frac{325517 b^7}{5259744} \right) \cos(4k). $$

(9)
As for $e_g$ and since $\omega(k, b)$ is independent of $a$, this result may also be interpreted as a SE for the triplet dispersion of a dimerized spin–1 chain.

In Fig. 3 we compare our SE results (solid lines) for $\omega(k, b)$ with other findings at an intermediate value of $b$ which will be shown later to reside still within the dimer phase. The first excited eigenvalues obtained from exact diagonalization (ED) on a finite dimerized spin–1 chain [14] are displayed with dots. Results from bond-boson mean field theory (MFT), bond-boson Linear Holstein Primakoff (LHF) methods [12, 23, 24] are shown with gray and dashed lines, respectively. As we can observe in this Fig. the agreement between the SE’s results and ED is very good, and the SE clearly improves on earlier uncontrolled approximations. Eq. (9) shows that there is a tendency to the closure of the gap: $\Delta = \omega(k = 0, b)$ as $b$ increases, which signals the transition to the Haldane phase. This will be analyzed in detail in the next Section. Finally we note, that in the 1D case our results extend to $O(7)$ an earlier SE which was obtained to $O(4)$ in ref. [14], however for a 3D tetrahedral spin model.

**QUANTUM PHASE TRANSITIONS**

In this Section we consider the quantum critical points of our model. Fig. 4 summarizes the phase diagram of the tetrahedral cluster spin chain as determined from the present SE and other methods [12, 13]. It displays the singlet product, dimer and Haldane phases. These regions are separated by a first order critical line between the dimerized spin–1 chain sector and the singlet product phase, as well as the second order critical line between the dimer and the Haldane phases.

We start by studying the dimer-to-singlet first order transition. The solid line represents our evaluation of the critical line $b_c(a_c)$ using SE. It has been obtained by numerical determination of $b(a)$ from $e_g(a, b) - e_g s_z = 0$, where $e_g s_z = -3a/2$ is the exact ground state energy of the singlet-product phase. Obviously, there is an excellent agreement with ED results [12, 25] (dotted line in Fig. 4). Surprisingly this is true even for $b$-values beyond the dimer-Haldane transition, where the SE is not expected to be valid anymore. From this we conclude that the ground state energy of the Haldane phase has little difference to that of an adiabatically continued dimer phase. While the accuracy of approximate approaches, as eg. bond-operator MFT [12], is hard to assess, it is interesting to see that the SE agrees well also with the latter approach at least up to the dimer-Haldane transition (dashed line in Fig. 4).

The dimer-Haldane transition of the dimerized spin–1 chain has been analyzed by several authors. From the analytical point of view, Haldane and Affleck [26, 27] have performed a mapping onto the $O(3)$ nonlinear $\sigma$–model (NLSM) in the large $S$ limit. They have shown that the topological angle $\theta$ is given by $\theta = 2\pi S(1 - \delta)$ and that the system is gapless and described by a conformal field theory with $SU(2)$ symmetry when $\theta/(2\pi)$ is half odd integer. $\delta$ is related to $b$ by $b = (1 - \delta)/(1 + \delta)$. Therefore, for $S = 1$ a gapless point was predicted to occur at $b_c = 1/3$, i.e. $\delta_c = 1/2$. While the prediction of a gapless point from the NLSM is expected to be correct, quantitative agreement for the critical point is not to be expected since $1/S$-correction may play a role and have not been analyzed to our knowledge.

From the numerical point of view, the dimer-Haldane transition has been studied employing Density Ma-
FIG. 4: Quantum phase diagram of the tetrahedral cluster chain. Solid: first order dimer-to-singlet critical line \( b_c(a_c) \) obtained from SE. Dashed and dots: \( b_c(a_c) \) from bond-boson mean field theory (MFT) and exact diagonalization (ED) in the dimerized spin–1 sector for \( N = 16 \) sites and PBC (ref. \[12\]). Horizontal solid line at \( b_c \in [0.612, 0.614] \): second order dimer-to-Haldane transition obtained by Dlog-Padé analysis of the triplet-gap closure of Eq. (9). (See also Fig. 5.)

Fig. 4 shows the integrated (4,3)-Dlog-Padé approximant for the triplet gap from our SE at \( O(7) \). As from Eq. (4), the gap is located at \( k = 0, \pi \). The axes have been scaled to allow for a comparison with two other approaches, i.e. ED on \( N = 16 \) sites with PBC in the dimerized spin–1 chain sector \[12\] and DMRG results from ref. \[28\]. The latter two cover the complete range of \( b(\delta) \in [0,1][1,0] \). For \( b(\delta) \geq 0.4(0.4) \), there is a very good agreement among all three approaches displayed. For greater(smaller) values of \( b(\delta) \) finite size effects become evident in the ED data. The results from the SE however remains very close to the DMRG over the complete range of inter-tetrahedral couplings, up until the critical point. From the inset of Fig. 5 one can observe that the SE slightly over(under)estimate the critical value of \( b_c(\delta_c) \) as compared to the findings of DMRG. In fact, performing standard Dlog-Padé error analysis, by evaluating the scatter of the critical point

\[ b_c(\delta_c) \in [0.612, 0.614] \] (\( \in [0.239, 0.240] \)), whereas DMRG estimates \( b_c(\delta_c) \in [0.59, 0.61] \). The small difference may be due to non analytic corrections to a plain power-law behavior of \( \Delta \). Indeed, the gap has been claimed to close as \( \Delta \sim (\lambda)^{\lambda^2/\log \lambda} \) with \( \lambda = |b-b_c|/b_c \), in the vicinity of the critical point \[28\]. In case of such logarithmic corrections, deviations as those shown in the inset of Fig. 5 are likely to occur and even higher order SE would be required to improve the agreement very close the critical point. Further evidence for the relevance of logarithmic corrections stems from the critical exponent of \( \nu \in [0.98, 0.99] \) which we extract from the Dlog-Padé approximant which, as can be seen also from Fig. 4, varies almost linearly with \( b(\delta) \) close to the critical point. This is at variance with extrapolation of ED data, which predict a critical effective exponent \( \nu \in [0.7, 0.8] \) (\( \Delta \sim \lambda^{\nu} \)) \[28\], and with the onset of additional curvature which can be observed in the DMRG results close to \( b_c(\delta_c) \) in the inset of Fig. 5. This issue should be addressed in future studies.

FIG. 5: Solid line: integrated Dlog-Padé (4,3) approximant to the \( O(7) \) SE, Eq. (9), for the one-triplet gap \( \Delta \equiv \omega(k=0, b) \). Axes have been scaled to allow for a comparison with other methods. Dots: ED on \( N = 16 \) sites with PBC in the dimerized spin–1 sector \[12\]. Crosses: DMRG results from \[28\]. Inset: zoom of the transition region.

CONCLUSIONS

To summarize, we have studied zero temperature properties of a tetrahedral cluster spin chain using a series expansion technique based on Wegner’s flow equation method. Starting from the limit of isolated tetrahedra, we have obtained expansions up to \( O(7) \) in the inter-tetrahedral couplings for the ground state energy and
the dispersion of the elementary one-triplet excitations within the dimer phase. The ground state energy has been used to determine a first order quantum critical line which separates a dimerized spin−1 chain sector from a singlet product phase. Our findings are in excellent agreement with those of ED. A second order critical line for a dimer-to-Haldane phase transition was obtained by analyzing the closure of the triplet gap. Again, very good agreement was found with the previous results from ED, DMRG and QMC.

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