Ground state properties of a generalized spin-1/2 distorted diamond chain

Ground state properties of a generalized spin-$\frac{1}{2}$ distorted diamond chain

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Abstract

With the help of the bond operator representation for three $S = \frac{1}{2}$ spins, we study the effective Hamiltonian and the phase diagram of a generalized spin-$\frac{1}{2}$ distorted diamond chain. In the weak-intertriangle-coupling limit, the magnetism of the effective Hamiltonian is studied with second-order perturbation theory and mean-field decoupling. Various phases such as the spin-fluid phase, the dimerized phase and the ferrimagnetic phase are shown to compete. For larger intertriangle interactions, the spin-fluid phase and the dimerized phase can be also described by the effective spin–orbital model and the region of the dimerized phase enlarges around the symmetric point of $J_1 = J_2 = J_3$ (J_1, J_2 and J_3 are the intratriangle interactions). The magnetization plateaus at $\frac{1}{3}M_s$ and $\frac{2}{3}M_s$ in the magnetic field are also studied.

1. Introduction

In recent years, frustrated magnetic systems have been attracting a lot of attention. Antiferromagnetic spin systems on geometrically frustrated lattices show many unusual behaviours of magnetic and thermal properties. For example, Heisenberg antiferromagnets on the triangular lattice [1, 2] are believed to have three-sublattice long-range Néel order, while Heisenberg antiferromagnets on the two-dimensional Kagóme lattice [3, 4] or on the three-dimensional pyrochlore lattice [5–7] have disordered ground states with high degeneracy. $SrCu_2(BO_3)_2$ [8, 9], which can be described by the Shastry–Sutherland model [10], has a spin-liquid ground state with a finite spin gap. The $m = \frac{1}{8}$, $\frac{1}{4}$ and $\frac{1}{3}$ magnetization plateaus observed in external magnetic fields [8, 11] have been studied extensively [12–16]. The planar model of weakly coupled spin-$\frac{1}{2}$ trimers describing the triangular spin-cluster compound $La_4Cu_3MoO_{12}$ [17] shows various long-range-ordered states to compete, depending on the ratio of the intratriangle coupling constants [18, 19]. Very recently, a quasi-one-dimensional material, $Cu_3Cl_6(H_2O)_2 \cdot 2H_8C_4SO_2$, consisting of $S = \frac{1}{2}$ trimer spin chains, has been studied experimentally [20] and theoretically [21–23]. From the measured
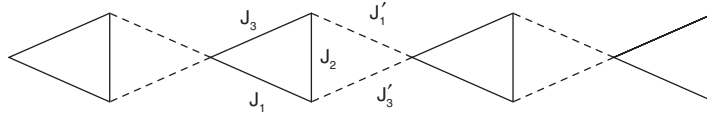
Figure 1. The DDC model with general intertriangle interactions. When $J_1 = J_1'$ and $J_3 = J_3'$, the model describes $Cu_3Cl_6(H_2O)_2 \cdot 2H_8C_4SO_2$.

susceptibility and magnetization curves [20], it is found that $Cu_3Cl_6(H_2O)_2 \cdot 2H_8C_4SO_2$ has a spin-singlet ground state with an excitation gap. Ishii *et al* then suggested a distorted diamond chain (DDC) model whose translational symmetry is spontaneously broken. In this paper, we study a generalized diamond chain model:

$$H = H_0 + H_1, \tag{1}$$

with

$$H_0 = \sum_i (J_1 \vec{S}_{i1} \cdot \vec{S}_{i2} + J_2 \vec{S}_{i2} \cdot \vec{S}_{i3} + J_3 \vec{S}_{i3} \cdot \vec{S}_{i1}),$$

$$H_1 = \sum_i (J_1' \vec{S}_{i3} \cdot \vec{S}_{i+1,1} + J_3' \vec{S}_{i2} \cdot \vec{S}_{i+1,1}), \tag{2}$$

where, as shown in figure 1, J_1, J_2 and J_3 denote intratriangle interactions and J_1' and J_3' the intertriangle interactions. $\vec{S}_{i,j}$ ($j = 1, 2, 3$) represents a spin-$\frac{1}{2}$ degree of freedom at site j of the triangle centred at position i. When $J_1 = J_1'$ and $J_3 = J_3'$, the Hamiltonian is used to describe $Cu_3Cl_6(H_2O)_2 \cdot 2H_8C_4SO_2$ and was called the DDC model. Furthermore, when $J_1 = J_3$, the system returns to the diamond chain model studied by Takano *et al* [24]. They showed that there exist three phases in the parameter space: the ferrimagnetic phase for $J_2/J_1 < 0.909$, the tetramer–dimer (TD) singlet phase for $0.909 < J_2/J_1 < 2$ and the dimer–monomer (DM) phase for $J_2/J_1 > 2$. When $J_1 \neq J_3$ (the DDC model), the symmetry of the tetramer cluster in the TD phase is broken and the dimerization is weighted on the J_1-bond (if $J_1 > J_3$). A complete phase diagram for the DDC model was given by Okamoto, Tonegawa *et al* [21, 22] and also by Sano and Takano [23]. In addition to the ferrimagnetic phase and the gapless spin-fluid phase, a gapped dimerized phase exists around the symmetric point of $J_1 = J_2 = J_3$. In section 2, with the help of the bond operator representation for three $S = \frac{1}{2}$ spins [19, 25], we map the model Hamiltonian (1) to an effective spin–orbit model and then study its phase diagram by second-order perturbation theory. In section 3, it is shown that the model is dimerized around the symmetric point of $J_1 = J_2 = J_3$ and the dimerized region increases with increasing J_1' and J_3'. The $\frac{1}{3}M_s$ and $\frac{2}{3}M_s$ magnetization plateaus predicted in the DDC model and in $Cu_3Cl_6(H_2O)_2 \cdot 2H_8C_4SO_2$ are studied in section 4.

2. The effective Hamiltonian and the phase diagram in the weak-coupling limit

A symmetric spin triangle with $J_1 = J_2 = J_3$ has a fourfold-degenerate ground state composed of two doublets corresponding to right and left chirality; the excited states are spin-$\frac{3}{2}$ quadruplets. Introducing eight bosons to denote the eight eigenstates, we obtain the

bond operator representation of the three spins S_p ($p = 1, 2, 3$) [25]:

$$
\begin{aligned}
S_p^+ = {}&-\tfrac{1}{3}(u_l^\dagger d_l + u_r^\dagger d_r) + \tfrac{1}{3}q_1^+ q_{-1} + \frac{1}{\sqrt{3}}(q_3^+ q_1 + q_{-1}^+ q_{-3}) \\
&+ \tfrac{1}{3}j^{2p}(2u_l^\dagger d_r - u_r^\dagger q_{-1} - q_1^\dagger d_l + \sqrt{3}d_r^\dagger q_{-3} + \sqrt{3}q_3^\dagger u_l) \\
&+ \tfrac{1}{3}j^p(2u_r^\dagger d_l - u_l^\dagger q_{-1} - q_1^\dagger d_r + \sqrt{3}d_l^\dagger q_{-3} + \sqrt{3}q_3^\dagger u_r), \\
S_p^z = {}&\tfrac{1}{6}(u_l^\dagger u_l + u_r^\dagger u_r + q_1^\dagger q_1 - d_l^\dagger d_l - d_r^\dagger d_r - q_{-1}^\dagger q_{-1}) + \tfrac{1}{2}(q_3^\dagger q_3 - q_{-3}^\dagger q_{-3}) \\
&+ \tfrac{1}{3}j^{2p}(d_l^\dagger d_r + d_r^\dagger q_{-1} + q_{-1}^+ d_l - u_l^\dagger u_r - u_r^\dagger q_1 - q_1^\dagger u_l) \\
&+ \tfrac{1}{3}j^p(d_r^\dagger d_l + q_{-1}^+ d_r + d_l^\dagger q_{-1} - u_r^+ u_l - q_1^\dagger u_r - u_l^\dagger q_1),
\end{aligned}
$$

(3)

where $j = e^{i\frac{2}{3}\pi}$, $u_{l(r)}^\dagger|0\rangle$, $d_{l(r)}^\dagger|0\rangle$ correspond to the doublets with left (right) chirality and $q_\alpha^\dagger|0\rangle$ with $\alpha = 3, 1, -1, -3$ to the quadruplets with $S_z = \frac{3}{2}, \frac{1}{2}, -\frac{1}{2}$ and $-\frac{3}{2}$ respectively. The restriction that the physical states are either the doublets or quadruplets leads to the constraint $u_l^\dagger u_l + u_r^\dagger u_r + d_l^\dagger d_l + d_r^\dagger d_r + q_\alpha^\dagger q_\alpha = 1$.

With the quadruplets projected out, the three $S = \frac{1}{2}$ spins S_p with $p = 1, 2, 3$ can be represented by the pseudospin operator and the chirality operator as [25–28]

$$
\begin{aligned}
S_p^+ &= \tfrac{1}{3}\mathcal{S}^+ - \tfrac{2}{3}j^{2p}\mathcal{S}^+\tau^+ - \tfrac{2}{3}j^p\mathcal{S}^+\tau^-, \\
S_p^z &= \tfrac{1}{3}\mathcal{S}^z - \tfrac{2}{3}j^{2p}\mathcal{S}^z\tau^+ - \tfrac{2}{3}j^p\mathcal{S}^z\tau^-,
\end{aligned}
$$

(4)

where $\mathcal{S} = S_1 + S_2 + S_3$ is the total spin of a spin triangle: $\mathcal{S}^+ = -(u_l^\dagger d_l + u_r^\dagger d_r)$ and $\mathcal{S}^z = \frac{1}{2}(u_l^\dagger u_l + u_r^\dagger u_r - d_l^\dagger d_l - d_r^\dagger d_r)$. The chirality operator can be expressed as: $\tau^+ = (u_l^\dagger u_r + d_l^\dagger d_r)$ and $\tau^z = \frac{1}{2}(u_l^\dagger u_l - u_r^\dagger u_r + d_l^\dagger d_l - d_r^\dagger d_r)$. It can be shown that $\mathcal{S}$ and τ are independent degrees of freedom.

Using the total spin operator $\mathcal{S}$ and the chirality operator τ, we obtain the effective Hamiltonian of model (1) at $J_1 = J_2 = J_3$:

$$
H_{eff} = -\tfrac{3}{4}NJ_1 + \sum_i \frac{J_1'}{9}A_{i,i+1}^\tau \mathcal{S}_i \cdot \mathcal{S}_{i+1},
$$

(5)

where N is the number of the spin triangles and $A_{i,i+1}^\tau$ the effective magnetic coupling coefficient containing operator τ: $A_{i,i+1}^\tau = [1 + \lambda - 2(1 + \lambda j)\tau_i^+ - 2(1 + \lambda j^2)\tau_i^-](1 - 2j^2\tau_{i+1}^+ - 2j\tau_{i+1}^-)$ with $\lambda = J_3'/J_1'$.

For a nonsymmetric spin triangle with general J_1, J_2 and J_3, the chirality symmetry no longer exists and the degeneracy of the two doublets will lift; we can define a transfer operator T to denote the change of the spin states from one doublet to another [19]: $T^+ = a_2^+ a_1 + b_2^+ b_1$, $T^z = \frac{1}{2}(a_2^+ a_2 + b_2^+ b_2 - a_1^+ a_1 - b_1^+ b_1)$, where $a_{1(2)}^\dagger|0\rangle$, $b_{1(2)}^\dagger|0\rangle$ denotes the lower (higher) doublet. The relations between the operators τ and T are

$$
\begin{aligned}
\tau^+ &= e^{i\delta}(T^z - iT^y), \\
\tau^- &= e^{-i\delta}(T^z + iT^y),
\end{aligned}
$$

(6)

where $\Delta e^{i\delta} = J_1 + jJ_2 + j^2 J_3$.

With the spin operator $\mathcal{S}$ and transfer operator T, we can map the original spin Hamiltonian (1) to an effective spin–orbital model:

$$
H_{eff}^{(1)} = H_a + H_b,
$$

(7)

with

$$
H_a = -\tfrac{1}{4}(J_1 + J_2 + J_3)N + \sum_i \Delta T_i^z,
$$

$$
H_b = \sum_i \frac{J_1'}{9}B_{ij}^T \mathcal{S}_i \mathcal{S}_{i+1},
$$

(8)

where

$$B_{ij}^T = \{1 + \lambda - 4[\cos\delta + \lambda\cos(\delta + \tfrac{2}{3}\pi)]T_i^z - 4[\sin\delta + \lambda\sin(\delta + \tfrac{2}{3}\pi)]T_i^y\}$$
$$\times [1 - 4\cos(\delta + \tfrac{4}{3}\pi)T_{i+1}^z - 4\sin(\delta + \tfrac{4}{3}\pi)T_{i+1}^y]. \tag{9}$$

In the following, we calculate the effective magnetic coupling coefficients in the weak-coupling limit of $J_1', J_3' \ll J_1, J_3, \Delta$. Since $[\vec{S}, \vec{T}] = 0$, we can regard the spin operators as c-numbers when we perform the perturbation with respect to the transfer operators. The ground state of H_a is $|\text{g.s.}\rangle = \prod_i |T_i^z = -\tfrac{1}{2}\rangle$. Up to first order, we have $T^z \sim -\tfrac{1}{2}$. Recalling that $e^{i\delta} = (J_1 + jJ_2 + j^2J_3)/\Delta$, we obtain the effective coefficient as

$$B = [1 + \lambda + (2 - \lambda)\cos\delta - \sqrt{3}\lambda\sin\delta](1 - \cos\delta + \sqrt{3}\sin\delta)$$
$$= \frac{1}{\Delta^2}[(1 + \lambda)\Delta + 2J_1 - (J_2 + J_3) - \lambda(J_1 + J_2 - 2J_3)](\Delta + 2J_2 - J_1 - J_3). \tag{10}$$

Along some special lines, the effective magnetic coupling coefficients to first order are zero and we have to calculate the higher-order perturbation. Considering lower-order excited states:

$$|1_i\rangle = |T_i^z = \tfrac{1}{2}\rangle \prod_{j\neq i} |T_j^z = -\tfrac{1}{2}\rangle,$$
$$|2_{ij}\rangle = |T_i^z = \tfrac{1}{2}\rangle|T_j^z = \tfrac{1}{2}\rangle \prod_{l\neq ij} |T_l^z = -\tfrac{1}{2}\rangle, \tag{11}$$

we get the effective magnetic coupling up to the second order as

$$H_{eff}^{(2)} = H_{eff}^{(2a)} + H_{eff}^{(2b)} \tag{12}$$

with

$$H_{eff}^{(2a)} = \frac{2J_1'^2}{81\Delta}(a^2b^2 + c^2d^2)\sum_i \mathcal{S}_i\mathcal{S}_{i+1} - \frac{2J_1'^2}{81\Delta}abcd\sum_i \mathcal{S}_i\mathcal{S}_{i+2},$$
$$H_{eff}^{(2b)} = \frac{8J_1'^2}{81\Delta}a^2d^2\sum_i \mathcal{S}_i\mathcal{S}_{i+1} \tag{13}$$

where $H_{eff}^{(2a)}$ and $H_{eff}^{(2b)}$ come from $|1_i\rangle$ and $|2_{ij}\rangle$ respectively and

$$a = \sin\delta + \lambda\sin(\delta + \tfrac{2}{3}\pi),$$
$$b = 1 + 2\cos(\delta + \tfrac{4}{3}\pi),$$
$$c = 1 + \lambda + 2[\cos\delta + \lambda\cos(\delta + \tfrac{2}{3}\pi)], \tag{14}$$
$$d = \sin(\delta + \tfrac{4}{3}\pi).$$

Now we discuss the phase diagram of the original spin Hamiltonian by checking the sign of the effective magnetic coupling. In figure 2, we show the effective magnetic coupling coefficients up to first order as functions of J_1 ($J_2 = 1$) for $J_3 = 0.4$ (solid curve), 0.8 (dashed curve), 1.2 (dotted curve) and 2.0 (dotted–dashed curve) given $\lambda = 1$ (figure 2 (a)) and $\lambda = J_3/J_1$ (figure 2 (b)). It is pointed out that the change of λ will not change the sign of the effective magnetic coupling coefficients. As shown in figure 3, on the lines of $J_1 = J_2 < J_3$ (line a), $J_2 = J_3 < J_1$ (line b) and $J_1 = J_3 < J_2$ (line c), the magnetic coupling coefficients up to first order are zero and the J_1–J_3 plane was separated into three regions. In the regions I and II, $B > 0$, the system is in the antiferromagnetic spin-fluid phase; while in the region III, $B < 0$, the system is in the ferrimagnetic phase. On lines a and b, the second-order perturbation gives $H_{eff}^{(2)} = \frac{J'^2}{18\Delta}(3\lambda^2 + 1)\sum_i \mathcal{S}_i\mathcal{S}_{i+1}$ and $H_{eff}^{(2)} = \frac{J'^2}{18\Delta}(3 + \lambda^2)\sum_i \mathcal{S}_i\mathcal{S}_{i+1}$ respectively, indicating that the system is still in the antiferromagnetic spin-fluid phase.
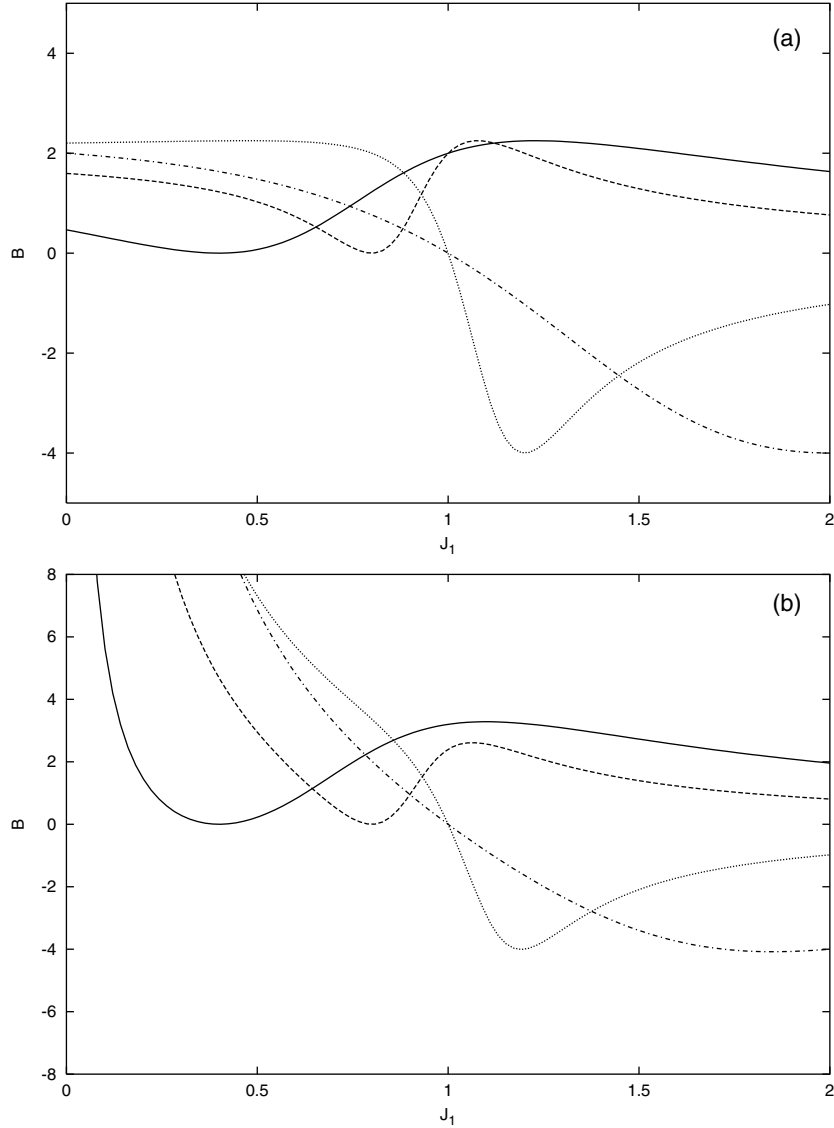
Figure 2. Effective magnetic coupling coefficients B as a function of J_1 with $J_3 = 0.4$ (solid curve), $J_3 = 0.8$ (dashed curve), $J_3 = 1.2$ (dotted curve) and $J_3 = 2.0$ (dashed–dotted curve) for (a) $\lambda = 1$ and (b) $\lambda = J_3/J_1$. J_2 is set to 1.

Along the line of $J_1 = J_3 < J_2$, we obtain

$$H_{eff}^{(2)} = \frac{J'^2}{6\Delta}(1 - \lambda)^2 \sum_i \mathcal{S}_i \mathcal{S}_{i+1}. \tag{15}$$

When $\lambda \neq 1$, $H_{eff}^{(2)} > 0$, the system is still in the spin-fluid phase. For $\lambda = 1$, we get $H_{eff}^{(2)} = 0$. Even up to higher-order perturbation, the effective magnetic coupling coefficient is still zero, which means that the system is in the DM state and every spin triangle is independent [24] with a free spin S_1 and a singlet formed by S_2 and S_3. The DM state is 2^N-fold degenerate. Tonegawa *et al* regarded it as a special case of a spin-fluid state with $S_{tot} = 0$ [22].
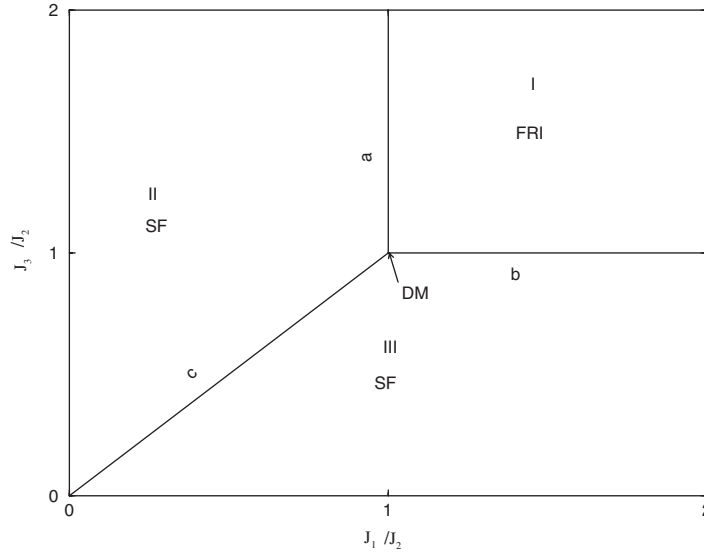
Figure 3. The phase diagram of the generalized DDC in the small-J_1' and small-J_3' limits.

3. Dimerized state in the generalized distorted diamond chain

At $J_1 = J_2 = J_3$, $\Delta = 0$, the perturbative theory in section 2 is not applicable. We can solve the effective spin–orbit model (5) by a mean-field decoupling that retains the quantum nature of the spin and chirality variables [3]. At the mean-field level, Hamiltonian (5) can be rewritten as

$$H_{eff} = -\tfrac{3}{4}NJ_1 + \sum_{\langle ij \rangle} \tfrac{1}{9}J_1'(a_{ij}^\tau \mathcal{S}_i \mathcal{S}_j + a_{ij}^s A_{ij}^\tau - a_{ij}^\tau a_{ij}^s), \tag{16}$$

where $a_{ij}^s = \langle \mathcal{S}_i \mathcal{S}_j \rangle$ and $a_{ij}^\tau = \langle A_{ij}^\tau \rangle$ should be determined self-consistently. For a two-triangle system (denoted by i and j), we use the basis $|\uparrow\uparrow\rangle$, $|\uparrow\downarrow\rangle$, $|\downarrow\uparrow\rangle$ and $|\downarrow\downarrow\rangle$ in the orbital space. The two-site orbital Hamiltonian A_{ij}^τ has eigenvalues $1+\lambda-4\sqrt{1-\lambda+\lambda^2}\pm 2(\lambda+1-\sqrt{\lambda^2-\lambda+1})$ and $1+\lambda+4\sqrt{1-\lambda+\lambda^2}\pm 2(\lambda+1+\sqrt{\lambda^2-\lambda+1})$. The corresponding eigenfunctions can be analytically obtained. We omit them here for simplicity. Considering the spin singlet, we get the lowest energy of $E_g = -\tfrac{3}{2}J_1 - \tfrac{1}{4}J_1'(1+\lambda+2\sqrt{1-\lambda+\lambda^2})$. The dimer has a wavefunction of $\phi_{ij}^\tau \otimes \phi_{ij}^\sigma$ with $|\phi_{ij}^\sigma\rangle$ a spin singlet and ϕ_{ij}^τ the eigenstate corresponding to the eigenenergy of $3(1+\lambda+2\sqrt{\lambda^2-\lambda+1})$. Following Mila [3], an N-triangle system is shown to have a wavefunction of $|\Phi_0(D)\rangle = \prod_{ij\in D} \phi_{ij}^\tau \otimes \phi_{ij}^\sigma$ since $\langle \phi_{ij}^\tau \phi_{kl}^\tau | A_{jk}^\tau | \phi_{ij}^\tau \phi_{kl}^\tau \rangle = 0$ and $\langle \phi_{ij}^\sigma \phi_{kl}^\sigma | \mathcal{S}_j \mathcal{S}_k | \phi_{ij}^\sigma \phi_{kl}^\sigma \rangle = 0$, which can be checked directly. The corresponding ground state energy is $E_g(N) = -\tfrac{3}{4}NJ_1 - \tfrac{1}{8}NJ_1'(1+\lambda+2\sqrt{1-\lambda+\lambda^2})$. For $\lambda = 1$, $E_g(N) = -\tfrac{3}{4}NJ_1 - \tfrac{1}{2}NJ_1'$ and $\phi_{ij}^\tau = \tfrac{1}{2}(-j, j^2, -j^2, 1)$.

The dimer wavefunction can be expressed in terms of the bond operators. At $\lambda = 1$,

$$\phi_{ij}^\tau \otimes \phi_{ij}^\sigma = \left(-\frac{j}{2\sqrt{2}}u_{li}^\dagger d_{lj}^\dagger + \frac{j^2}{2\sqrt{2}}u_{li}^\dagger d_{rj}^\dagger - \frac{j^2}{2\sqrt{2}}u_{ri}^\dagger d_{lj}^\dagger + \frac{1}{2\sqrt{2}}u_{ri}^\dagger d_{rj}^\dagger - d \longleftrightarrow u \right)|0\rangle \tag{17}$$

where $|0\rangle$ is the vacuum state and $d \longleftrightarrow u$ denotes similar expressions with d and u exchanged. Inserting the spin wavefunctions represented by $u(d)_{l(r)}^\dagger|0\rangle$ [19], we get the spin configuration

of the dimer as

$$\phi_{ij}^{\tau} \otimes \phi_{ij}^{\sigma} = \frac{j-1}{6\sqrt{2}}((|\uparrow_1\downarrow_3\rangle - |\downarrow_1\uparrow_3\rangle)_i(|\uparrow_{i2}\downarrow_{j1}\rangle - |\downarrow_{i2}\uparrow_{j1}\rangle)(|\uparrow_2\downarrow_3\rangle - |\downarrow_2\uparrow_3\rangle)_j$$
$$+ (|\uparrow_1\downarrow_2\rangle - |\downarrow_1\uparrow_2\rangle)_i(|\uparrow_{i3}\downarrow_{j1}\rangle - |\downarrow_{i3}\uparrow_{j1}\rangle)(|\uparrow_2\downarrow_3\rangle - |\downarrow_2\uparrow_3\rangle)_j), \qquad (18)$$

which is just the dimerized state found by Tonegawa *et al* (figures 1(c) and (d) in [22]). Thus we show that at the symmetric point of $J_1 = J_2 = J_3$, the DDC has a dimerized ground state and the ground state is just the one found by Tonegawa *et al*.

Very recently, Raghu *et al* [31] studied a spin-triangle chain with J_1 the intratriangle interaction and J_2 the intertriangle interaction. They derived the effective Hamiltonian to second order in the ratio of J_2/J_1 by means of degenerate perturbation theory and then compared the results obtained by exact diagonalizaiton of the effective Hamiltonian with those obtained for the full Hamiltonian using exact diagonalization and the density-matrix renormalization group method. For the model that they studied, the effective Hamiltonian gives an accurate ground state energy only when the ratio is <0.2. The case is different for the present model. The only approximation in our mapping to the effective spin–orbit Hamiltonian is to project out the $S = \frac{3}{2}$ spin states. If a spin singlet forms in every spin triangle in the ground state, the total spin of every spin triangle will be $\frac{1}{2}$, and then the effective spin–orbital Hamiltonian will describe well the low-temperature properties of the original spin Hamiltonian if the intertriangle interactions do not activate the $S = \frac{3}{2}$ spin states. Therefore, our effective Hamiltonian will be applicable to the DM and the TD states in the diamond model for $J_1 = J_3 = J_1' = J_3'$ [24], as well as the spin-fluid and the dimerized states in the DDC for $J_1' = J_1$ and $J_3' = J_3$ [22].

For those points not close to the symmetric point of $J_1 = J_2 = J_3$, Δ will be large and the perturbation theory in section 2 will be applicable. Therefore, when J_1' and J_3' increase, the outline of the phase diagram will not change, but the region of the dimerized state may enlarge. With the mean-field decoupling, the effective Hamiltonian (7) and (8) can be written as

$$H_{MF}^{(1)} = -\tfrac{1}{4}(J_1 + J_2 + J_3)N + \sum_i \Delta T_i^z + \sum_{\langle ij\rangle} \tfrac{1}{9}J_1'(a_{ij}^T S_i S_j + a_{ij}^s B_{ij}^T - a_{ij}^T a_{ij}^s), \qquad (19)$$

where a_{ij}^s and $a_{ij}^T = \langle B_{ij}^T\rangle$ should be determined self-consistently. In the basis $|\uparrow\uparrow\rangle$, $|\uparrow\downarrow\rangle$, $|\downarrow\uparrow\rangle$ and $|\downarrow\downarrow\rangle$, the two-site orbital Hamiltonian B_{ij}^T has eigenvalues $1 + \lambda - 4\sqrt{1-\lambda+\lambda^2} \pm 2(\lambda + 1 - \sqrt{\lambda^2 - \lambda + 1})$ and $1 + \lambda + 4\sqrt{1-\lambda+\lambda^2} \pm 2(\lambda + 1 + \sqrt{\lambda^2 - \lambda + 1})$. Taking into account the spin part, we find that the dimer is a spin singlet with a complicated orbital wavefunction corresponding to the eigenenergy $3(1 + \lambda + 2\sqrt{\lambda^2 - \lambda + 1})$. So in the dimerized state, the effective Hamiltonian (19) has a variational ground state energy $E_1 = -\tfrac{1}{4}N(J_1+J_2+J_3)-\tfrac{1}{8}NJ_1'(1+\lambda+2\sqrt{1-\lambda+\lambda^2})$; while in the spin-fluid state, the effective Hamiltonian has a variational energy $E_2 = -\tfrac{1}{4}N(J_1 + J_2 + J_3) - \tfrac{1}{2}\Delta N + \tfrac{J_1'}{9}B(-0.4431)N$, where -0.4431 is from the Bethe ansatz solution of the antiferromagnetic spin-$\frac{1}{2}$ chain. From $E_1 = E_2$, a first-order phase transition between the dimerized state and the spin-fluid state can be obtained. However, just as pointed out by Tonegawa *et al* [22], the transition between the spin-fluid phase and the dimerized phase is of second order except at some special points. In the following, we only discuss the transitions along the symmetric lines of $J_1 = J_3$, $J_1 = J_2$ and $J_2 = J_3$. On the line of $J_1 = J_3 < J_2$, we have $B = 0$, $\Delta = J_2 - J_1$ and $E_2 = -\tfrac{1}{4}N(J_1 + J_2 + J_3) - \tfrac{1}{2}\Delta N$. We can determine the transition from the dimerized state to the spin-fluid state occurring at $J_{1c} = J_2 - \tfrac{1}{4}J_1'(1 + \lambda + 2\sqrt{1-\lambda+\lambda^2})$. When $\lambda = 1$ and $J_1' = J_1$, we get $J_{1c} = \tfrac{1}{2}J_2$, which agrees exactly with the result of Tonegawa *et al* [22].

Similarly, along the lines of $J_1 = J_2 < J_3$ and $J_2 = J_3 < J_1$, we get the critical points of $J_{3c} = J_2 + \frac{1}{4}J_1'(1 + \lambda + 2\sqrt{1 - \lambda + \lambda^2})$ and $J_{1c} = J_2 + \frac{1}{4}J_1'(1 + \lambda + 2\sqrt{1 - \lambda + \lambda^2})$. It is obvious that the area of the dimerized state depends on the relation between J_1, J_2 and J_3 as well as J_1' and J_3'. More work is needed to study the second-order phase transition. It will be interesting to study the change of the energy gap in the dimerized phase.

4. Magnetization plateaus in the distorted diamond chain

The magnetization plateaus in the quantum spin systems have been actively studied in recent years [32–37]. One third of the saturated magnetization M_s was predicted in the spin-$\frac{1}{2}$ trimerized Heisenberg chains [32, 34, 35] or three-leg spin ladder [36]. For $Cu_3Cl_6(H_2O)_2 2H_8C_4SO_2$, a magnetization plateau of $\frac{1}{3}M_s$ was suggested to occur at the field of 58 T [20]. The $\frac{1}{3}M_s$ and $\frac{2}{3}M_s$ magnetization plateaus for the DDC model were also discussed [22]. In the following, we use the bond operator representations of the spin-$\frac{1}{2}$ triangles to study the magnetization of the generalized DDC model in the condition of weak intertriangle interactions [37]. To study the magnetization process, we have to include the $S = \frac{3}{2}$ quadruplets.

The Hamiltonian H_0 can be expressed in terms of the bond operators as

$$H_0 = \sum_i [-\frac{1}{4}(J_1 + J_2 + J_3)(a_{1i}^\dagger a_{1i} + b_{1i}^\dagger b_{1i} + a_{2i}^\dagger a_{2i} + b_{2i}^\dagger b_{2i})$$

$$+ \frac{1}{4}(J_1 + J_2 + J_3)(q_{1i}^\dagger q_{1i} + q_{-1i}^\dagger q_{-1i} + q_{3i}^\dagger q_{3i} + q_{-3i}^\dagger q_{-3i}) + \Delta T_i^z], \tag{20}$$

and the Zeeman term is

$$H_z = -h \sum_i [\frac{1}{2}(a_{1i}^\dagger a_{1i} + a_{2i}^\dagger a_{2i} + q_{1i}^\dagger q_{1i} - b_{1i}^\dagger b_{1i} - b_{2i}^\dagger b_{2i} - q_{-1i}^\dagger q_{-1i})$$

$$+ \frac{3}{2}(q_{3i}^\dagger q_{3i} - q_{-3i}^\dagger q_{-3i})]. \tag{21}$$

When the external magnetic field is small, the lowest two states of the spin triangle in the external magnetic field are $a_1^\dagger|0\rangle$ and $b_1^\dagger|0\rangle$. Projecting out the other six states, we get

$$S_p^+ \sim -\frac{1}{3}\left[1 + 2\cos\left(\frac{4\pi}{3}p + \delta\right)\right]\sigma^+,$$

$$S_p^z \sim \frac{1}{3}\left[1 + 2\cos\left(\frac{4\pi}{3}p + \delta\right)\right]\sigma^z, \tag{22}$$

with $\sigma^+ = a_1^\dagger b_1$ and $\sigma^z = \frac{1}{2}(a_1^\dagger a_1 - b_1^\dagger b_1)$. The total Hamiltonian $H_T = H_0 + H_1 + H_z$ can be represented by the pseudospin $\vec{\sigma}$ as

$$H_T^{(1)} = \sum_i [J_{xy}^{(1)}(\sigma_i^x \sigma_{i+1}^x + \sigma_i^y \sigma_{i+1}^y) + J_z^{(1)}\sigma_i^z \sigma_{i+1}^z] - h\sum_i \sigma_i^z \tag{23}$$

with $J_{xy}^{(1)} = J_z^{(1)} = \frac{1}{9}J_1'\{(1 + 2\cos\delta) + \lambda[1 + 2\cos(\delta + \frac{2}{3}\pi)]\}[1 + 2\cos(\delta + \frac{4}{3}\pi)]$.

When the magnetic field increases, the energy of $q_3^\dagger|0\rangle$ will become lower than that of $b_1^\dagger|0\rangle$. So for large enough magnetic field, we preserve the two states of $a_1^\dagger|0\rangle$ and $q_3^\dagger|0\rangle$, and then the spins can be expressed with a pseudospin $\vec{\Omega}$:

$$\mathcal{S}_p^+ \sim (-1)^p \frac{2}{\sqrt{6}} e^{i\frac{\pi+\delta}{2}} \sin\left(\frac{\pi}{3}p - \frac{1}{2}\delta\right)\Omega^+,$$

$$\mathcal{S}_p^z \sim \frac{1}{3}\left[1 - \cos\left(\frac{4\pi}{3}p + \delta\right)\right]\Omega^z + \frac{1}{3} + \frac{1}{6}\cos\left(\frac{4\pi}{3}p + \delta\right), \tag{24}$$

with $\Omega^+ = q_3^\dagger a_1$ and $\Omega^z = \frac{1}{2}(q_3^\dagger q_3 - a_1^\dagger a_1)$. The total Hamiltonian then becomes

$$H_T^{(2)} = \sum_i [J_{xy}^{(2)}(\Omega_i^x \Omega_{i+1}^x + \Omega_i^y \Omega_{i+1}^y) + J_z^{(2)}\Omega_i^z \Omega_{i+1}^z] - h_{eff}\sum_i \Omega_i^z, \qquad (25)$$

where

$$J_{xy}^{(2)} = \frac{2}{3}J_1'\left[\sin\frac{\delta}{2}\sin\left(\frac{1}{3}\pi - \frac{\delta}{2}\right) - \lambda\sin\left(\frac{2}{3}\pi - \frac{\delta}{2}\right)\sin\left(\frac{\pi}{3} - \frac{\delta}{2}\right)\right],$$

$$J_z^{(2)} = \frac{1}{9}J_1'\left\{(1 - \cos\delta) + \lambda\left[1 - \cos\left(\frac{2\pi}{3} + \delta\right)\right]\right\}\left[1 - \cos\left(\frac{4\pi}{3} + \delta\right)\right], \qquad (26)$$

$$h_{eff} = h - \frac{1}{2}\Delta - \frac{1}{2}(J_1 + J_2 + J_3) - h',$$

with $h' = \frac{1}{9}J_1'\{2 + \frac{1}{2}\cos(\frac{2\pi}{3} + \delta) - \cos\delta\cos(\frac{4\pi}{3} + \delta) + \lambda[2 + \frac{1}{2}\cos\delta - \cos(\frac{2\pi}{3} + \delta)\cos(\frac{4\pi}{3} + \delta)]\}$. The saturated magnetic field of the effective Hamiltonian (23) is $h_s^{(1)} = 2J_{xy}^{(1)}$. At $h = \Delta$, the energies of the two states $b_1^\dagger|0\rangle$ and $a_2^\dagger|0\rangle$ will cross. So at $h \geqslant \min(\Delta, h_s^{(1)})$, the magnetization of the original spin Hamiltonian (1) will be $\frac{1}{3}M_s$. On the other hand, when the effective Hamiltonian (25) saturates in a negative effective magnetic field, the magnetization will be $\frac{1}{3}M_s$. So when $\min(\Delta, h_s^{(1)}) \leqslant h \leqslant h_s^{(2)} = \frac{1}{2}\Delta + \frac{1}{2}(J_1 + J_2 + J_3) + h' - |J_{xy}^{(2)}| - J_z^{(2)}$, we will have a $\frac{1}{3}M_s$ magnetization plateau with every spin triangle in the state $a_1^\dagger|0\rangle$. Furthermore, when $|J_z^{(2)}/J_{xy}^{(2)}|\rangle 1$ and below a critical magnetic field depending on $J_z^{(2)}/J_{xy}^{(2)}$, the effective Hamiltonian (22) will be antiferromagnetic and gapped with the two states $a_1^\dagger|0\rangle$ and $q_3^\dagger|0\rangle$ appearing alternatively, which means a $\frac{2}{3}M_s$ magnetization plateau since $\sum_{p=1}^{3}\cos(\frac{4\pi}{3}p + \delta) = 0$. The width of the plateau is [38]

$$\Delta h = 2|J_{xy}^{(2)}|\sinh\alpha\sum_{n=-\infty}^{\infty}\frac{(-1)^n}{2\cosh n\alpha} \qquad (27)$$

with $\cosh\alpha = J_z^{(2)}/J_{xy}^{(2)}$ and $\alpha > 0$.

5. Summary

In summary, we have studied the ground state properties of a generalized DDC with the help of the bond operator representations for three $S = \frac{1}{2}$ spins. We mapped the model Hamiltonian of the antiferromagnetic spin-$\frac{1}{2}$ trimer chain systems to an effective spin–orbit model and then studied the phase diagram of the effective model by means of second-order perturbation theory and the mean-field decoupling. Depending on the ratio of the intratriangle coupling constants, the ferrimagnetic phase, the spin-fluid phase and the dimerized phase are shown to compete at low temperatures. The region of the dimerized phase around the symmetric point of $J_1 = J_2 = J_3$ increases with increasing intertriangle interactions. The $\frac{1}{3}M_s$ and $\frac{2}{3}M_s$ magnetization plateaus in the external magnetic field were also studied.

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