Phase diagram of ferrimagnetic ladders with bond-alternation

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1. INTRODUCTION

Since the seminal work of Haldane quantum spin chains have been extensively studied as one of the simplest but most typical quantum many-body systems. According to Haldane, the 1-dimensional integer-spin Heisenberg antiferromagnets have a unique disordered ground state with a finite excitation gap, while half-integer antiferromagnets are gapless and critical (see references therein). The origin of the difference between half-integer and integer spin chains can be traced back to the topological $\theta$ term in the effective non-linear $\sigma$-model description of antiferromagnetic spin chains and is believed to be due to non-perturbative effects. Haldane’s original predictions were based on large-spin arguments and although a general rigorous proof is still lacking, several theoretical developments have helped to clarify the situation and there is now strong experimental and numerical evidence in support of Haldane’s claims. The massive phase of the integer spin chains, called Haldane phase, has been understood as valence-bond-solid (VBS) states proposed by Affleck et al., wherein each spin-$S$ is viewed as a symmetrized product of $2S$ spinors. However bond-alternation may drastically change the low-energy behavior of these systems and produce phase transitions. For instance a bond-alternation spin-$1/2$ antiferromagnet chain has a finite energy gap as opposed to its uniform counterpart. It has been also shown that the phase diagram of the $S=1$ chain decomposes into different phases by adding bond-alternation.

Yet another challenge in this area has led to synthesizing quasi-one-dimensional bimetallic molecular magnets. The search for molecular ferromagnet has led to the discovery of many interesting molecular magnetic systems. In recent years, quasi-one-dimensional bimetallic molecular magnets, with each unit cell containing two spins of different spin value have been synthesized. These systems contain two transition metal ions per unit cell and have the general formula $ACu(PbaOH)(H_2O)_3\cdot2H_2O$ with $PbaOH=2$-hydroxo-propylenebis (oxamato) and $A=Mn, Fe, Co, Ni$. They have been characterized as the alternating (mixed) spin chains or ferrimagnets. It has been shown that one-dimensional ferrimagnets have two types of excitations in their low-lying spectrum. The lowest one has gapless excitations, i.e., they behave like ferromagnets with quadratic dispersion relation. The other one which is separated by an excitation gap from the primer one, has antiferromagnetic behaviour with linear dispersion relation. More precisely, the effective Hamiltonian for the lowest spectrum of a 1-dimensional Heisenberg ferrimagnets is a 1-dimensional Heisenberg ferromagnet with $S=|S_1-S_2|\gamma$. A spin wave study also show the similar behavior is seen in two and three dimensions.

Another surprising investigation deals with spin ladders which have recently attracted a considerable amount of attention. They consist of coupled one-dimensional chains and may be regarded as interpolating truly one and two-dimensional systems. These models are useful to study the properties of the high-$T_c$ superconductor materials. Theoretical studies have suggested that there are two different universality classes for the uniform-spin ladders, i.e., the antiferromagnetic spin-$1/2$ ladders are gapful or gapless, depending on whether $n_l$ (the number of legs) is even or odd. These predictions has been confirmed experimentally by compounds like $SrCu_2O_3$ and $Sr_2Cu_3O_5$. However, again bond-alternation changes this universality. It has been shown that a gapless line which depends on the staggered bond-alternation (SBA) parameter $\gamma$, divides the gapful phase of a 2-leg antiferromagnetic spin-$1/2$ ladder into two different phases. Moreover, there are some other configurations, like the columnar bond-alternation (CBA) that introduces new phases for the antiferromagnetic ladder.
chains and ladders, and the appearance of the new phases for spin ladders, are also some of the consequences of the bond-alternations.

In this paper, we study the bond-alternation ferrimagnetic ladders (BAFL). This model contains a rich phase diagram. By making use of a quantum renormalization group (QRG), we demonstrate that the combination of both spin-mixing and bond-alternation may open the possibility of observing the energy ‘gap’ in the ground state of these systems. We obtain the phase diagram of BAFL in both SBA and CBA configurations. The SBA configuration consists of a gapfull and two gapless phases, while the CBS one is composed of two gapless phases. The boundary of these phases where phase transition takes place is calculated by QRG method. By using QRG we have obtained the effective Hamiltonian in both strong and weak coupling limit of BAFL. We have also shown the structure continued in the intermediate region.

The paper is organized as follows. In Section II, we present the effective Hamiltonian of CBA configuration by QRG method. We show that the phase transition occurs in the $J' < 0$ region. In Sec. III, we then present the SBA aconfiguration of ferrimagnetic ladder and discuss the structure of phase diagram in both positive and negative $J'$ region. Sec. IV. is devoted to conclusions.

II. COLUMNAR BOND ALTERNATION

Our model consists of mixed spins $\tau = 1/2$ and $S = 1$ on a 2-leg ladder (see Fig. 1). The Hamiltonian for this system can be divided into three parts: $H = H'_1 + H'_2 + H_r$, where $H'_\mu$ ($\mu = 1, 2$), and $H_r$ are the exchange interaction of the spins inside the $\mu$th leg, and the interaction between different legs, as shown in Fig. 1. The explicit form of the Hamiltonian is

$$H'_1 = J \sum_{i=1}^{N/2} [(1 + \gamma) \vec{\tau}_1(2i - 1) \cdot \vec{S}_1(2i) + (1 - \gamma) \vec{S}_1(2i) \cdot \vec{\tau}_1(2i + 1)]$$

$$H'_2 = J \sum_{i=1}^{N/2} [(1 + \gamma) \vec{S}_2(2i - 1) \cdot \vec{\tau}_2(2i) + (1 - \gamma) \vec{\tau}_2(2i) \cdot \vec{S}_2(2i + 1)]$$

$$H_r = J' \sum_{i=1}^{N/2} \vec{\tau}_1(2i - 1) \cdot \vec{S}_2(2i - 1) + \vec{S}_1(2i) \cdot \vec{\tau}_2(2i)$$

(1)

The ladder contains $N$ rungs and by assuming the periodic boundary condition for each leg, we identify the first rung to the $N + 1$-th rung. Through out this paper we assume $J$ is positive but $J'$ can be positive or negative as a tunable parameter. Note that for the bond-alternation parameter, $\gamma$ becomes $-\gamma$ amounts to sublattice exchange $n \rightarrow n + 1$, and therefore we consider $0 \leq \gamma \leq 1$.

By QRG, we divide the Hamiltonian into intra-block ($H^B$) and inter-block ($H^{BB}$) parts. After diagonalizing the first part, a number of low-energy eigenstates are kept to project the full Hamiltonian onto the renormalized one. As opposed to other powerful techniques, such as the density matrix renormalization group (DMRG), the QRG approach is much less complicated yielding analytical results, although QRG does not give as accurate numerical results as DMRG, nevertheless its simplicity can give a good qualitative picture of the phase diagram. In this paper four interesting configurations are in order: positive and negative value of $J'$ for both CBA and SBA configurations.

A. The case $J' > 0$

The CBA configuration [Fig. (a)] and $J' > 0$. Let us first consider the strong coupling limit ($J' \gg J$). Since the interaction between two sites on each rung is strong, then each rung can be considered as the isolated block in the first step of the QRG, i.e., $H^B = H_r$. The Hilbert space of each block ($\tau - S$) consists of two multiplets whose total spin are $3/2$ and $1/2$. The corresponding energies for these two configurations are $J'/2$ and $-J'$. Therefore we keep the $S = 1/2$ multiplet as the basis for constructing the embedding operator $T$ to project the full Hamiltonian onto the truncated Hilbert space ($H_{\text{eff}} = T^\dagger H_T \overline{T}$). Finally the effective Hamiltonian (in which each rung is mapped to a single site) can be obtained

$$H_{\text{eff}} = -NJ' - \frac{8}{9} J \sum_{i=1}^{N/2} [(1 + \gamma) \vec{S}'(2i - 1) \cdot \vec{S}'(2i) + (1 - \gamma) \vec{S}'(2i) \cdot \vec{S}'(2i + 1)],$$

(2)

where $S' = 1/2$ and $i$ is the label of the sites on new chain which represents the $i$th rung of the original ladder. Eq. (3) is the Hamiltonian of a spin-1/2 (bond-alternation) ferrimagnetic chain. It exhibits gapless excitations as well as the ferromagnetic ground state. Equivalently, the original ladder exhibits the ferrimagnetic ground state, in the sense that both magnetization ($m = -\tau > +S > 0.5$) and the staggered one ($sm = -S > -\tau > 5/6$) are non-zero with a gapless excitations.

Now, let us consider the weak coupling limit ($J' \ll J$). In this case the stronger bonds, e.g. $J(1 + \gamma)$, appear on the legs. They are considered as the isolated blocks. The other bonds (the weaker ones) are considered as $H^{BB}$. The QRG procedure leads to the Heisenberg (spin-1/2) ferromagnet ladder. In this regime the spectrum of the system is similar to the strong coupling limit.

For the intermediate region where $J' \approx J$, the two types of blocking which has been considered in the strong and weak coupling limits seems to be not suitable. In this regard we have considered a 4-sites block which consists of both rung and leg interactions [Fig. (a)]. Decomposition of ladder to 4-sites blocks leads to two types of
building blocks, which is shown in [Fig. 2(b)] and [Fig. 2(c)]. The lowest energy multiplet of the Hamiltonian in [Fig. 2(b)] has total spin $\tilde{S} = 5/2$ and the corresponding one in [Fig. 2(c)] has $\tilde{r}' = 1/2$. The embedding operator is constructed from these two multiplets and finally the effective Hamiltonian ($H_{\text{eff}} = T^HT$) is a (1/2, 5/2) ferrimagnetic chain.

$$H_{\text{eff}} = -\frac{g}{8} NJ + \frac{40}{63} \sum_{i=1}^{N/2} [\tilde{\tau}'(2i - 1) \cdot \tilde{S}(2i) + \tilde{S}(2i) \cdot \tilde{\tau}'(2i + 1)]$$

In the next step of QRG procedure the Hamiltonian in Eq. (2) is projected to a $\tilde{S} = 2$ ferrimagnetic Heisenberg chain:

$$H_{\text{eff}}(\tilde{S} = 2) = -\frac{9}{8} J + \frac{40}{63} \sum_{i=1}^{N/4} \tilde{S}(i) + \tilde{S}(i + 1)$$

Thus the magnetization ($m$) and staggered magnetization ($sm$) of the original ladder in the intermediate regime are: $m \simeq 0.2292$; $sm \simeq 0.4514$, which shows ferrimagnetic order. We therefore find the different regimes have similar structure (the ferrimagnetic ground state with the gapless excitations) as long as $J'$ is positive.

### B. The case $J' < 0$

This configuration has more interesting features. Let us first consider the strong coupling limit where $H^B$ is constructed by $H_r$. Since $J'$ is negative the low-energy multiplet has total spin 3/2. Thus this subspace is considered as the effective Hilbert space for the first step of the QRG. The effective Hamiltonian ($H_{2\text{eff}}$) can be obtained by projecting each operator onto the effective Hilbert space

$$H_{2\text{eff}} = \frac{N J'}{2} + \frac{4}{9} \sum_{i=1}^{N/2} [(1 + \gamma)\tilde{S}'(2i - 1) \cdot \tilde{S}''(2i) + (1 - \gamma)\tilde{S}''(2i) \cdot \tilde{S}'(2i + 1)]$$

where $\tilde{S}''$ is a spin 3/2. Hamiltonian (3) is a 1-dimensional Heisenberg spin-3/2 antiferromagnet with alternating bonds. It is known that this model is gapful when $\gamma > \gamma_c$ and gapless otherwise. Although the one-, two- and three-dimensional ferrimagnets show gapless behavior, the combinations of the spin-mixing and bond-alternations yields the possibility for developing of an energy gap. This can be compared to the spin-1/2 CBA Heisenberg ladders, where the model is gapful in the whole range of parameter space ($J, J', \gamma$) except on a critical line, in the region where $J'$ is negative. In the next step of QRG, $H_{2\text{eff}}$ will be projected to a spin-1/2 XXZ model in the presence of an external magnetic field ($h$). It is known that XXZ+$h$ model has the critical line $h_c = \Delta + 1$, where $\Delta$ is the anisotropy in the $\tilde{z}$-direction. If $h > h_c$ (h < $h_c$) the model is gapful (gapless) and the value of gap is proportional to the strength of magnetic field. We find that $\gamma_c = 3/7 (\approx 0.428)$ for our model which is close to the DMRG results ($\approx 0.42$) for spin-3/2 dimerized chain. Therefore in the ferromagnetic region ($J' < 0$) and strong coupling ($|J'| \gg J$) there is a critical value for $\gamma$ below which ($\gamma < \gamma_c$) the BAFL is gapless and its ground state has quasi-long range order (quasi-LRO) where both $m$ and $sm$ are zero and correlations decay algebraically. This is equivalent to the uniform 1-dimensional spin-3/2 antiferromagnets. For $\gamma > \gamma_c$ the model is gapful.

In the weak coupling limit ($|J'| \ll J$), the strongest bonds, e.g. $J(1 + \gamma)$, of the ladder are considered as building blocks of QRG, and the remaining bonds are treated as $H^{BB}$. The effective Hamiltonian in this case is

$$H_{3\text{eff}} = \frac{8g |J'|}{9} \sum_{i=1}^{N/2} S''_i(i) \cdot S_2(i) - \frac{4J(1 - \gamma)}{9} \sum_{i=1}^{N/2} \sum_{\mu=1,2} \tilde{S}''_{\mu}(i) \cdot \tilde{S}''_{\mu}(i + 1)$$

Here $S' = 1/2$ and we have neglected a constant term. The effective Hamiltonian ($H_{3\text{eff}}$) is a ‘double-layer’ model of spin-1/2. Neglecting the inter-site terms (for a moment), each couple of sites has a multiplet of states with total angular momentum $\ell = 0, 1$, where each pair of inter-layer spins on the two layers, behaves like a single quantum rotor where $\tilde{L} = \tilde{S}_1 + \tilde{S}_2$ and $\tilde{n} = (\tilde{S}_1 - \tilde{S}_2)/2S$. Considering the intra-layer terms, one may map the Hamiltonian (4) to that 1-dimensional quantum rotor model (5)

$$H_{3\text{eff}} = \gamma g g \frac{g}{2} \sum_{i=1}^{N/2} \tilde{L}_i^2 - K \sum_{i=1}^{N/2} (\tilde{n}_i \cdot \tilde{n}_{i+1} + \tilde{L}_i \cdot \tilde{L}_{i+1})$$

where $g \equiv 16|J'|/9$ and $K = 4J(1 - \gamma)/9$. The mean field phase diagram of this model is governed by the gapped quantum paramagnet when $\gamma \rightarrow 1$ and the partially polarized ferromagnet when $\gamma \rightarrow 0$. The dominant term of the first limit is the antiferromagnetic exchange term along the rungs which makes singlets as the base structure for ground state of the renormalized ladder. Thus the ground state is unique and has a finite energy gap to the first excited state. This gapful phase occurs in the region where $|J'|/J \gg 1 - \gamma$, which is the continuation of the gapful phase in the strong coupling limit. Thus we have no-LRO in the ground state of the original ladder and finite gap in this region. For the latter limit, there is a competition between the ferromagnetic term and the antiferromagnetic term in (6). Note that the dominant term in $H_{3\text{eff}}$ is the ferromagnetic interaction along the legs. Then the ground state is composed of
two ferromagnetic ordered chains which are aligned oppositely due to antiferromagnetic interaction \((8|J'|/9)\). But this classical antiferromagnetic alignment fluctuates along the \(z\)-axis and the magnetization is reduced along this direction.

In the intermediate region (where \(|J'| \approx J\)), the ladder is decomposed to 4-sites plaquettes if \(\gamma \to 1\). Note that \(S_z = 0\) is the unique ground state of any 4-site plaquette at \(2J = -J' = 1\) (one may see this after diagonalizing the Hamiltonian). But every plaquette is on the \(S_z = 0\) state, since the ladder is disconnected. This ground state is not degenerate and disorder (with no-LRO) and there is a finite energy gap to the first excited state. In other words the whole gapful phase of the ladder is in the VBS phase.

On the other extreme case, when \(\gamma\) is negligible, the gapless phase behaves differently in long wave-length limit. Let us first suppose \(|J'| > J\). In this case, each rung behaves as a spin-3/2 (after the first step of the QRG), since the spins on the same rung are coupled by the ferromagnetic interaction \((J' < 0)\). Hence the whole ladder is identified by a Heisenberg spin-3/2 antiferromagnetic chain. The ground state is gapless with quasi-LRO, and the correlation functions falls off algebraically (the correlation length \(\xi\), is infinite). But if \(|J'| < J\) the ladder can be considered as two quantum ferrimagnetic chains that interact via weak ferromagnetic coupling (through their rungs). The correlation length is small and the correlation functions falls off exponentially. One may naturally expect that at \(|J'| \sim J\) a phase transition is observed between two gapless phases. One phase is the half-integer quantum ferrimagnets (when \(|J'| \gg J\)) with \(\xi = \infty\) and quasi-LRO and another phase is the ferrimagnetic phase (when \(|J'| \ll J\)) with \(\xi \sim a\) (a is the lattice spacing). The order parameter to specify this phase transition is \(\tilde{m} = |m_1 - m_2|\), where \(m_{1(2)}\) is the magnetization per site of the 1-st (2-nd) leg of ladder. \(\tilde{m}\) is zero where \((J'/J) < -1\) and \(\tilde{m} = 0.5\) for \((J'/J) > -1\). The total magnetization \((m)\) and staggered one \((sm)\) are zero on both side of this critical line. This completes the phase diagram of the CBA configuration. It is depicted in Fig. 3(a).

\section*{III. STAGGERED BOND ALTERNATION}

\subsection*{A. The case \(J' > 0\)}

The SBA configuration is shown in [Fig. 3(b)]. In this region the effective Hamiltonian shows ferromagnetic behavior [similar to the CBA \((J' > 0)\) configuration ] and the ladder behaves like the gapless ferrimagnets. As an example, we may apply the “snake mechanism” of reference\(^4\), choosing \(\gamma = 1\) and \(J' = 2J\) the ladder degenerates into a uniform alternating spin-1/2 – 1 ferrimagnetic chain which has gapless excitations. More precisely the effective Hamiltonian \((H_{\text{eff}})\) in the strong coupling limit \((J' \gg J)\) is

\[
H_{4\text{eff}} = -NJ' - \frac{8}{9} J \sum_{i=1}^{N} \vec{S}_i^z \cdot \vec{S}_{i+1}^z
\]

where \(J' = 1/2\) and the block Hamiltonian for QRG procedure is considered to be \(H_r\). In the weak coupling limit \((J' \ll J)\) the strongest bonds are \(J(1+\gamma)\), the QRG procedure leads to a strip of triangular lattice as the effective Hamiltonian \((H_{5\text{eff}})\).

\[
H_{5\text{eff}} = -NJ(1+\gamma) - \frac{4}{9} J(1-\gamma) \sum_{\mu=1}^{N} \sum_{i=1}^{\mu} \vec{S}_i^\mu \cdot \vec{S}_{i+1}^\mu
\]

The effective Hamiltonians \((H_{4\text{eff}}, H_{5\text{eff}})\) are \(S = 1/2\) ferromagnetic Heisenberg model. Thus in both of these cases the model has gapless excitations. Since both of \(m\) and \(sm\) are not zero, we have ferrimagnetic order in the whole part of \((J' > 0)\) region.

\subsection*{B. The case \(J' < 0\)}

In this region and at the strong coupling limit \((|J'| \gg J)\) the ladder is mapped to a uniform Heisenberg spin-3/2 antiferromagnetic chain, where the alternation parameter is disappeared at the first step of the QRG. Thus the whole range of \(\gamma \in [0,1]\) is gapless and disordered. The system exhibits quasi-LRO with \(\xi = \infty\). But when \(|J'| \ll J\), the effective Hamiltonian is equivalent to a two 1-dimensional spin-1/2 ferromagnetic Heisenberg chains. These two chains interact by an antiferromagnetic coupling on a triangular ladder as shown in Fig. 3(c). At \(\gamma = 1\) the ladder transforms to a 1-dimensional spin-1/2 antiferromagnetic chain which is gapless with quasi-LRO. But at \(\gamma = 0\) (where the CBA is equivalent to SBA) the ladder is equivalent to two ferromagnetic chain which are coupled antiferromagnetically. It represents a ferrimagnetic phase where \(\xi \sim a\). As we have illustrated above the competition between the coupling constants in Fig. 3(c) leads to one of the above extreme cases. In other words, if \(|J'|/J > 1 - \gamma\) the system is equivalent to the Heisenberg spin-1/2 antiferromagnetic chains. For another opposite limit, the system is equivalent to the Heisenberg ferrimagnetic chains. The dashed line in Fig. 2(b) which separates these two phases, represents the critical line.

\section*{IV. CONCLUSION}

In summary, we have obtained the phase diagram \((J'/J, \gamma)\) of a 2-leg bond-alternating \((1/2,1)\) ferrimagnetic ladder. In CBA configuration, Fig. 1(a), there
exists a gapful (VBS) phase which is separated from two different gapless phases by the critical lines. The phase transition between gapless phases takes place when $J'/J \sim -1$. For the SBA configuration, Fig.1(b), the whole phase diagram is gapless. One phase contains the ferrimagnetic behavior and another one is equivalent to the Heisenberg spin-1/2 antiferromagnetic chains. In the latter region, the correlation function falls off algebraically. The transition between these two gapless phases takes place when $J'/J \sim (\gamma - 1)$.

Although the $(1/2, 1)$ ferrimagnetic system is considered as a generic model for all $(S_1, S_2)$ systems, we have found that this is not longer true in ladders if one considers the bond-alternation effects, where both spin-mixing and bond-alternation may change the quantum phase transitions. The dependence on different spins and the number of legs should be considered in future investigations.

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FIG. 1. The ladder realization $\tau = 1/2$ and $S = 1$, (a) Columnar bond-alternation (CBA), (b) staggered bond-alternation (SBA), (c) schematic illustration of effective Hamiltonian in SBA configuration at $|J'| \ll J$, (where $S' = 1/2$).

FIG. 2. (a) Decomposition of ladder into 4-sites blocks in the intermediate region ($J \sim J'$), (b) three spin-1 and a spin-1/2, (c) three spin-1/2 and a spin-1.

FIG. 3. Phase diagram of bond-alternation ferrimagnetic 2-leg ladder, (a) Columnar bond-alternation (CBA), (b) staggered bond-alternation (SBA), solid line is the critical line between gapless and gapful phases and the dashed one shows the critical line between two gapless phases.
Fig. 1
Fig. 2
Fig. 3

(a) gapful (VBS), no-LRO

(b) gapless (QAF), quasi-LRO

Gapless ferrimagnetic order

\( \gamma_{c} \)

\( \gamma \)

\( J'/J \)