Phase diagram of ferrimagnetic ladders with bond alternation

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(Received 30 June 1999; revised manuscript received 28 September 1999)

We study the phase diagram of a two-leg bond-alternation spin-(1/2, 1) ladder for two different configurations using a quantum renormalization-group approach. Although d-dimensional ferrimagnets show gapless behavior, we will explicitly show that the effect of the spin mixing and the bond alternation can open the possibility for observing an energy gap. We show that the gapless phases of such systems can be equivalent to the one-dimensional half-integer antiferromagnets, besides the gapless ferrimagnetic phases. We therefore propose a phase transition between these two gapless phases that can be seen in the parameter space.

I. INTRODUCTION

Since the seminal work of Haldane[1] quantum spin chains have been extensively studied as one of the simplest but most typical quantum many-body systems. According to Haldane, the one-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 Ref. 2 and references therein). The origin of the difference between half-integer and integer spin chains can be traced back to the topological ''θ'' term in the effective nonlinear σ-model description of antiferromagnetic spin chains and is believed to be due to nonperturbative 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 claim.[2–4] 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.*,[5] 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.[6,7] It has been also shown that the phase diagram of the $S=1$ chain decomposes into different phases by adding bond alternation.[8]

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.[9] These systems contain two transition-metal ions per unit cell and have the general formula $A\mathrm{Cu(pbaOH)(H_2O)_3.2H_2O}$ with pbaOH $=2$-hydroxo-propylenebis (oxamato) and $A=$Mn, Fe, Co, Ni. They have been characterized as the alternating (mixed) spin chains or ferrimagnets.[10] 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 behavior with linear dispersion relation.[11] More precisely, the effective Hamiltonian for the lowest spectrum of a one-dimensional Heisenberg ferrimagnet is a one-dimensional Heisenberg ferromagnet with $S=|S_1-S_2|$.[12] A spin-wave study also shows the similar behavior is seen in two and three dimensions.[12]

Another surprising investigation deals with spin ladders that have recently attracted a considerable amount of attention.[13] 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.[13] These predictions have been confirmed experimentally by compounds like $\mathrm{SrCu_2O_3}$ and $\mathrm{Sr_2Cu_3O_5}$. However, again bond alternation changes this universality. It has been shown that a gapless line that depends on the staggered bond-alternation (SBA) parameter γ, divides the gapful phase of a two-leg antiferromagnetic spin-1/2 ladder into two different phases.[14,15] Moreover, there are some other configurations, like the columnar bond alternation (CBA) [see Fig. 1(a)] that introduces new phases for the antiferromagnetic ladders.[16,27] The appearance of the magnetization pla-

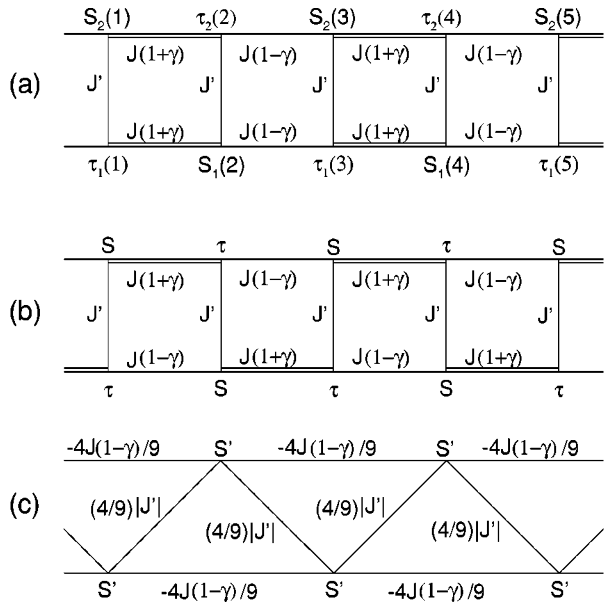

FIG. 1. The ladder realization $\tau=1/2$ and $S=1$. (a) Columnar bond alternation (CBA), (b) staggered bond alternation (SBA), (c) schematic illustration of effective Hamiltonian in SBA configuration at $|J'|\ll J$, where $S'=1/2$.

teaus for both chains[17] and ladders[18] and the appearance of the new phases for spin ladders,[19] 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 (QRG) group,[20] 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 CBA configuration consists of a gapful and two gapless phases, while the SBA 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 limits of BAFL. We have also shown the structure continued in the intermediate region.

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

II. COLUMNAR BOND ALTERNATION

Our model consists of mixed spins $\tau=1/2$ and $S=1$ on a two-leg ladder (see Fig. 1). The Hamiltonian for this system can be divided into three parts: $H=H_1^l+H_2^l+H_r$, where H_μ^l ($\mu=1,2$) and H_r are the exchange interaction of the spins inside the μth leg, and the interaction between different legs, as shown in Fig. 1. The explicit form of the Hamiltonian is

$$H_1^l=J\sum_{i=1}^{N/2}\,[(1+\gamma)\vec{\tau}_1(2i-1)\cdot\vec{S}_1(2i)+(1-\gamma)\vec{S}_1(2i)\cdot\vec{\tau}_1(2i+1)]$$

$$H_2^l=J\sum_{i=1}^{N/2}\,[(1+\gamma)\vec{S}_2(2i-1)\cdot\vec{\tau}_2(2i)+(1-\gamma)\vec{\tau}_2(2i)\cdot\vec{S}_2(2i+1)]$$

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

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. Throughout 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, $\gamma\rightarrow-\gamma$ amounts to sublattice exchange $n\rightarrow n+1$, and therefore we consider $0\leqslant\gamma\leqslant1$.

By QRG, we divide the Hamiltonian into intrablock (H^B) and interblock (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[21] (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.[22] In this paper four interesting configurations are in order: positive and negative value of J' for both CBA and SBA configurations.

A. Case $J'>0$

The CBA configuration [Fig. 1(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 spins 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 ($H_{\rm eff}=T^\dagger HT$).[23] Finally, the effective Hamiltonian (in which each rung is mapped to a single site) can be obtained

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

where $S'=1/2$ and i is the label of the sites on a new chain that represents the ith rung of the original ladder. Equation (2) is the Hamiltonian of a spin-1/2 (bond-alternation) ferromagnetic 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

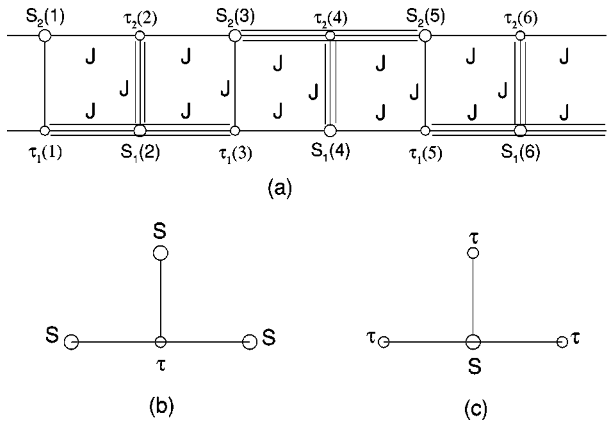

FIG. 2. (a) Decomposition of ladder into four-sites blocks in the intermediate region ($J \sim J'$), (b) three spin-1 and a spin-1/2; (c) three spin-1/2 and a spin-1.

magnetization ($m = \langle \tau \rangle + \langle S \rangle = 0.5$) and the staggered one ($sm = \langle S \rangle - \langle \tau \rangle = 5/6$) are nonzero with a gapless excitation.

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

$$H_{i\text{-eff}} = -\frac{9}{8} NJ + \frac{40}{63} J \sum_{i=1}^{N/4} [\vec{\tau}'(2i-1) \cdot \vec{\hat{S}}(2i) + \vec{\hat{S}}(2i) \cdot \vec{\tau}'(2i+1)]. \tag{3}$$

In the next step of QRG procedure the Hamiltonian in Eq. (3) is projected to a $\tilde{S} = 2$ ferromagnetic Heisenberg chain,[12]

$$H_{\text{eff}}(\tilde{S}=2) = -\frac{101}{72} NJ - \frac{10}{81} J \sum_{i=1}^{N/4} \vec{\tilde{S}}(i) \cdot \vec{\tilde{S}}(i+1). \tag{4}$$

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. 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

$$H_{2\text{ eff}} = \frac{NJ'}{2} + \frac{4}{9} J \sum_{i=1}^{N/2} [(1+\gamma) \vec{S}''(2i-1) \cdot \vec{S}''(2i) + (1-\gamma) \vec{S}''(2i) \cdot \vec{S}''(2i+1)], \tag{5}$$

where S'' is a spin 3/2. Hamiltonian (5) is a one-dimensional Heisenberg spin-3/2 antiferromagnet with alternating bonds. It is known[7] 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 yield the possibility for developing of an energy gap. This can be compared to the spin-1/2 CBA Heisenberg ladders[16] where the model is gapful in the whole range of parameter space (J, J', γ) 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[24] that XXZ$+h$ model has the critical line $h_c = \Delta + 1$, where Δ is the anisotropy in the $\hat{z}$ direction.[22] 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 $\gamma_c = 3/7 (\simeq 0.428)$ for our model that is close to the DMRG results ($\simeq 0.42$) for spin-3/2 dimerized chain.[7] Therefore in the ferromagnetic region ($J' < 0$) and strong coupling ($|J'| \gg J$) there is a critical value for γ 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 one-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

$$H_{3\text{ eff}} = \frac{8|J'|}{9} \sum_{i=1}^{N/2} \vec{S}'_1(i) \cdot \vec{S}'_2(i) - \frac{4J(1-\gamma)}{9} \sum_{i=1}^{N/2} \sum_{\mu=1,2} \vec{S}'_{\mu}(i) \cdot \vec{S}'_{\mu}(i+1). \tag{6}$$

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 intersite terms (for a moment), each couple of sites has a multiplet of states with total angular momentum $l = 0,1$,[25] where each pair of interlayer spins on the two layers behaves like a single-quantum rotor where $\vec{L} = \vec{S}_1 + \vec{S}_2$ and $\vec{n} = (\vec{S}_1 - \vec{S}_2)/2S$. Considering the intralayer terms, one may map the Hamiltonian (6) to that one-dimensional quantum rotor model,[26]

$$H_{3\ \mathrm{eff}}=\frac{g}{2}\sum_{i=1}^{N/2}\vec{L}_i^2-K\sum_{i=1}^{N/2}(\vec{n}_i\cdot\vec{n}_{i+1}+\vec{L}_i\cdot\vec{L}_{i+1}), \tag{7}$$

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 that 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 Eq. (6). Note that the dominant term in $H_{3\ \mathrm{eff}}$ is the ferromagnetic interaction along the legs. Then the ground state is composed of two ferromagnetic ordered chains that are aligned oppositely due to antiferromagnetic interaction $(8|J'|/9)$. But this classical antiferromagnetic alignment fluctuates along the $\hat{z}$ axis and the magnetization is reduced along this direction.

In the intermediate region (where $|J'|\simeq J$), the ladder is decomposed to four-sites plaquettes if $\gamma\rightarrow 1$. Note that $S_z=0$ is the unique ground state of any four-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 γ is negligible, the gapless phase behaves differently in the long-wavelength 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 antiferromagntic chain. The ground state is gapless with quasi-LRO, and the correlation functions falls off algebraically (the correlation length ξ 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 antiferromagnets (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 1st (2nd) 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).

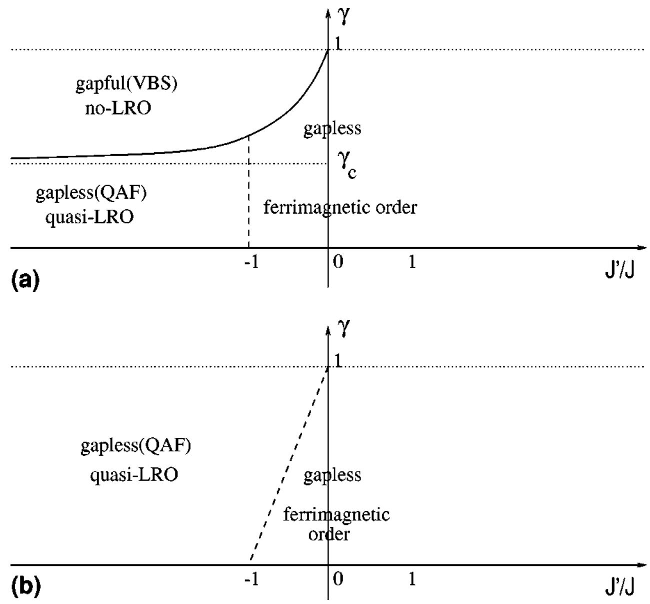

FIG. 3. Phase digram of a bond-alternation ferrimagnetic two-leg ladder. (a) Columnar bond alternation (CBA), (b) staggered bond alternation (SBA), solid line is the critical line between gapless and gapful phases and the dashed one shows the critical line between two gapless phases.

III. STAGGERED BOND ALTERNATION

A. Case $J'>0$

The SBA configuration is shown in Fig. 1(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 Ref. 14: choosing $\gamma=1$ and $J'=2J$ the ladder degenerates into a uniform alternating spin-1/2$-$1 ferrimagnetic chain that has gapless excitations. More precisely the effective Hamiltonian ($H_{4\ \mathrm{eff}}$) in the strong-coupling limit $(J'\gg J)$ is

$$H_{4\ \mathrm{eff}}=-NJ'-\frac{8}{9}J\sum_{i=1}^{N}\vec{S}'(i)\cdot\vec{S}'(i+1), \tag{8}$$

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

$$H_{5\ \mathrm{eff}}=-NJ(1+\gamma)-\frac{4}{9}J(1-\gamma)\sum_{\mu=1}^{2}\sum_{i=1}^{N}\vec{S}'_\mu(i)\cdot\vec{S}'_\mu(i+1)$$
$$-\frac{4}{9}J'\sum_{i=1}^{N}[\vec{S}'_1(i)\cdot\vec{S}'_2(i)+\vec{S}'_2(i)\cdot\vec{S}'_1(i+1)]. \tag{9}$$

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

B. 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 disappears 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 two one-dimensional spin-1/2 ferromagnetic Heisenberg chains. These two chains interact by an antiferromagnetic coupling on a triangular ladder as shown in Fig. 1(c). At $\gamma=1$ the ladder transforms to a one-dimensional spin-1/2 antiferromagnetic chain that is gapless with quasi-LRO. But at $\gamma=0$ (where the CBA is equivalent to SBA) the ladder is equivalent to two ferromagnetic chains that 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. 1(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) that separates these two phases represents the critical line.

IV. CONCLUSION

In summary, we have obtained the phase digram $(J'/J,\gamma)$ of a two-leg bond-alternating (1/2, 1) ferrimagnetic ladder. In the CBA configuration, Fig. 1(a), there exists a gapful (VBS) phase that 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 no longer true in ladders if one considers the bond-alternation effects, where both spin mixing and bond alternation may change the quantum phases. The dependence on different spins and the number of legs should be considered in future investigations.

ACKNOWLEDGMENTS

We would like to thank H. Hamidian, Hsiu-Hau Lin, and G. Sierra for helpful discussions and useful comments. M.A. acknowledges support from EPS-9720651 and a grant from the Oklahoma State Regents for Higher Education. M.A.M.-D. is supported by the DGES Spanish Grant No. PB97-1190.

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