Antiferromagnetically coupled alternating spin chains

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I. INTRODUCTION

The recent synthesisesis of quasi-onedimensional bimetallic magnets \([1,2]\), with each unit cell containing two kind of different spins, has spurred a growing interest in the low-temperature properties of quantum antiferromagnetic chains. After intensive studies \([3,4]\), it was possible to describe properly the thermodynamic properties observed in experiments by including the ferro and antiferromagnetic (AF) features that make this systems specially attractive. These results together with the possible experimental realization of new mixed spin compounds motivated the study of a variety of AF systems with different spin composition, all kind of interactions, and topologies \([10–13]\). From the experimental point of view, the ferrimagnetic compounds found are all composed of weakly coupled alternating spin chains \([1]\) (Fig.1(b)) represents the ladder case of such a topology). In other family of compounds like \(\text{MnCu}(\text{pba})(\text{H}_2\text{O})_3\cdot 2\text{H}_2\text{O}\), with pba=1,3-propylenebis(oxamato), the ground state is a non-magnetic one, although it is composed of weakly coupled alternating spin \((\text{Mn}^{II}\text{Cu}^{II})\) chains \([\text{Fig.1(a)}\) and (c)]. In fact, using the Heisenberg Hamiltonian for these interacting spin systems and applying Lieb-Mattis theorem \([4]\), it is easy to prove that in case (a) and (c) the ground state is AF with total spin \(S_{\text{tot}} = 0\) whereas in case (b) it is ferrimagnetic with \(S_{\text{tot}} \neq 0\). Nonetheless, in the 2D case we will see that the rotational spin symmetry can be broken in the thermodynamic limit. The aforementioned considerations motivated us to focus on the ground state properties of an alternating spin ladder of type (a) and a 2D array of type (c) with the following Hamiltonian,

\[
H = J \sum_{<i,j>_\parallel} S_i S_j + J_{\perp} \sum_{<i,j>_\perp} (S_i S_j + S_i S_j),
\]

where \(S_i(S_j)\) represents a spin-1 (spin-1/2)and \(<i,j>_\parallel\) \(<i,j>_\perp\) denotes nearest neighbors along horizontal (vertical) direction. To our knowledge, this is the first systematic study of antiferromagnetically coupled alternating spin chains. Only recently, this issue has been ad-

![FIG. 1. Three kind of alternating spin systems: (a) and (c) have a singlet ground state, (b) has a ferrimagnetic one.](image.png)

...dressed but for ferrimagnetic ladders (Fig.1(b)) \([5,6]\). Instead, in the alternating spin ladder we are interested in, (in what follows ASL), the global properties of the system change completely once the chains are coupled. Unlike uniform spin ladders, little is known about the effect of AF interchain coupling in ASL. For instance, in a uniform spin-1/2 ladder (USL), it is well known that an infinitesimal \(J_{\perp}\) is required to send the chains off-criticality –with an excitation gap growing linearly with \(J_{\perp}\) independently of its sign \([7,8]\). Besides, it has been pointed out that the absence of broken symmetry in the chains is indeed the crucial feature for such a behaviour \([7]\). On the other hand, in the spin-1 case, it has been argued that an interchain coupling \(J_{\perp}\) makes the gap (already existing in the chains) to decrease with \(J_{\perp}\), and again no matter the sign of \(J_{\perp}\) \([9]\). For the mixed case, we are concerned, the situation is rather different as the ferrimagnetic chains are neither gapped nor critical,
but they are gapless as a result of a broken symmetry ground state. Hence, one could intuitively say that the effect of interchain coupling should be different to the both uniform cases mentioned above and a finite amount of \( J_\perp \) could be needed to melt order. We shall see, however, that the low dimensionality of the problem (1D) is crucial and that any \( J_\perp > 0 \) drives the system to a gapped ground state. The study of the spin correlation functions confirms this picture, showing a strong resemblance to USL. Contrary to the ASL, in the 2D case we shall see that the dimensional crossover from 1D is completely smooth and keeps the ground state ordered for the whole range of \( J_\perp \).

The work is organized as follows: in section II we study the dimensional crossover from 1D to 2D within spin wave theory. In section III we give some details of the DMRG method implemented and discuss the results for the ladder case. In section IV we summarize the results and give the concluding remarks.

**II. SPIN WAVE THEORY**

Though approximate, it has been shown that the spin wave (SW) series converges correctly to the DMRG results in ferrimagnetic chains \([2]\). Hence SW appears to be, in principle, a good starting point technique to study the crossover from 1D to 2D where SW is even more reliable. It is also known, however, that for a ladder system SW breaks down \([2]\) (see below). The problem requires a unit cell composed of two spins \( S = 1 \) and two spins \( S = 1/2 \) (see Fig 1(c)). At the classical level, it is assumed a Néel order and quantum fluctuations are incorporated by four kind of bosons leading to the following Holstein-Primakov transformation,

\[
\begin{align*}
S_A^z &= -s_1 + a^\dagger a, \\
S_B^z &= s_2 - b^\dagger b, \\
S_C^z &= s_1 - c^\dagger c, \\
S_D^z &= -s_2 + d^\dagger d.
\end{align*}
\]

Introducing this representation in the Hamiltonian \([3]\), and Fourier transforming to \( k \)-space, the SW Hamiltonian can be written as,

\[
H_{sw} = \text{const} + \sum_k \pi_k \left( \begin{array}{cc} d & \Delta \\ \Delta & d \end{array} \right) \pi_k
\]

where \( \pi_k = (a_k^\dagger, b_k^\dagger, c_k^\dagger, d_k^\dagger, a_k, b_k, c_k, d_k) \), \( d = \frac{1}{2} \text{diag}(A_1, A_2, A_1, A_2) \),

\[
\Delta = \frac{1}{2} \begin{pmatrix}
0 & C_k & D^\dagger_k & 0 \\
C_k & 0 & 0 & D^\dagger_k \\
D_k & 0 & 0 & C_k \\
0 & D^\dagger_k & C_k & 0
\end{pmatrix},
\]

\[
\text{const} = -4NJzs_1s_2 - N^2J_\perp(s_1^2 + s_2^2)
\]

\[
A_1 = (2Js_2 + 2J_\perp s_1), \quad A_2 = (2Js_1 + 2J_\perp s_2)
\]

\[
D^\dagger_k = 2s_1J_\perp \cos ky/2, \quad D^\dagger_k = 2s_2J_\perp \cos ky/2,
\]

\[
C_k = 2J\sqrt{s_1s_2} \cos kx/2.
\]

We have para-unitary diagonalized \([2]\) \( H_{sw} \) which allowed us to calculate the spectrum excitation (with an acoustic and an optic band each one twofold degenerate), the ground state energy and the magnetizations. For the particular case \( J_\perp = 0 \) we recover the SW predictions \([4,5,6]\) of a ferrimagnetic chain, that is, a quadratic gapless dispersion near \( k = 0 \) (Fig 3) (which manifests the ferromagnetic character of the system at low-temperature), an antiferromagnetic gapped mode, and the magnetization values \( m_1 = 0.6951 \) and \( m_2 = 0.1951 \) for the spin-1 and spin-1/2, respectively. As soon as \( J_\perp \) is switched on the gapless mode becomes linear reflecting the antiferromagnetic character \([21]\) of the coupled system, whereas the optical modes moves upward (Fig 4). For the isotropic case \( J_\perp = 1 \) we obtained a ground state energy \( e_0 = -1.3631 \), and \( m_1 = 0.7326 \), \( m_2 = 0.3694 \) for the magnetizations. Notice that \( m_2 \) is bigger than the SW value of the uniform spin-1 case \( m = 0.303 \). Using mean-field arguments, such a robust spin-1/2 magnetization is due to the more stronger field caused by the spin-1 neighbors (a similar reasoning explains why \( m_1 \) is smaller than the SW value of the uniform spin-1 case \( m = 0.803 \)). It is also interesting to note that the relative reduction of the magnetizations, \( m_1/S \) and \( m_2/S \), are both nearly 0.73 in the isotropic case, while \( m_1/S \sim 0.70 \) and \( m_2/S \sim 0.40 \) for the ferrimagnetic chain. In Fig 5 we show \( m_1 \) and \( m_2 \) versus \( J_\perp \). It can be seen that the effect of 2D coupling is to enhance the values of the magnetizations, being the crossover completely smooth. On the other hand, our SW calculation can be adapted to

![FIG. 2. Spin wave spectrum for the 2D array. Dashed, dotted, and solid lines are for \( J_\perp = 0, 0.2, \) and 0.5, respectively. For the ladder case the spectrum is qualitatively the same.](image-url)
The procedure follows the usual DMRG steps [22], thinking the ladder like a chain of rungs. We found out that to avoid great edge effects, because of the OBC, the definition of two different single sites is needed, each one composed of a spin-1/2 rung and spin-1 rung, respectively (see Fig.1(a)). The 4 and 9 states needed to span a minimal 2 rungs (otherwise we would need just 6 states) appear at the beginning as a complication to the number of dominant density matrix states kept, $m$, but they shown to be just of minor annoyance. Most of the calculations was carried out using $m$ between 200 and 400, getting a truncation error $O(10^{-8})$ at worst and $O(10^{-12})$ in the best case, reassuring us the reliability of the calculation. We have calculated the triplet spin gap which is defined as $\Delta(J) = E(L, S_\perp = 1) - E(L, S_\perp = 0)$, with $E(L, S_\perp)$ the ground state energy for a chain with $L$ rungs ($2 \times L$ is the number of sites), and $z$ component of total spin $S_z$ [25]. For each $J_\perp$, we have extrapolated the gap with $L$ ranging from 10 to 100 rungs using a polynomial fit of the form [24]:

$$\Delta(L) = \Delta + a_1/L + a_2/L^2 + a_3/L^3 + \ldots$$

In Fig.4 it is plotted the gap $\Delta$ vs. $J_\perp$. In the particular case, $J_\perp = 1$, we obtained a value for the gap of $\Delta = 0.334$. For small values of $J_\perp$ (below 0.3) the gap of the ASL behaves quadratically as $\Delta \sim 0.6J_\perp^2$ and beyond 0.3 it turns out quite linear corresponding to the strong coupling regime. Consistently with these features, when extrapolating the gap for different values of $J_\perp$, we detected a crossover between two different scaling regimes around $J_\perp \sim 0.3$. This is shown in the inset of Fig.4 where we have plotted the scaling for $J_\perp = 0.1, 0.3, 0.8$ and 1. It is interesting to compare our result for the weakly coupled regime with that of the USL, which is known to behave linearly as $\Delta \sim 0.34L^2$ and beyond. We think that this discrepancy reflects the quite distinct underlying physics behind the decoupled regime, that is, a critical state for uniform spin chains and a broken symmetry state for the ferrimagnetic chains. In order to get an insight of the ground state configuration of the ASL we computed the local bond strengths and compared them with the USL. The comparison was made after normalizing each value to its free bond case. Hence, for the very strong coupling regime the values of the bonds along the leg (rung) must go to zero (one). Actually, we calculated the $z$ component of these quantities and because of rotational invariance we multiply it by a factor 3. In Fig.5(a), we present the normalized values of $\langle S_i s_{i+1} \rangle$ along the leg. A similar monotonic behavior with $J_\perp$ is shown.

III. DENSITY MATRIX RENORMALIZATION GROUP

The difficulty of SW theory to describe ASL leads us to implement the DMRG method, which has shown a great versatility in different quasi-onedimensional systems. We have performed calculations using both, the finite and infinite algorithm, with open boundary conditions (OBC). The procedure follows the usual DMRG steps [22], thinking the ladder like a chain of rungs. We found out that to avoid great edge effects, because of the OBC, the definition of two different single sites is needed, each one composed of a spin-1/2 rung and spin-1 rung, respectively (see Fig.1(a)). The 4 and 9 states needed to span each rung (otherwise we would need just 6 states) appear at the beginning as a complication to the number of dominant density matrix states kept, $m$, but they shown to be just of minor annoyance. Most of the calculations was carried out using $m$ between 200 and 400, getting a truncation error $O(10^{-8})$ at worst and $O(10^{-12})$ in the
is observed for ASL and USL. On the other hand, in Fig. 5(b) we show the normalized values of \( \langle S_i S_{i+1} \rangle \) and \( \langle s_i s_{i+1} \rangle \), corresponding to the vertical spin-1 rungs and spin-1/2 rungs respectively. It can be noticed that the spin-1 rungs are always larger than the spin-1/2 ones, however, what is really remarkable is the crossing between the spin-1/2 rungs values for ASL and USL which occurs just at \( J_\perp \sim 0.3 \). Even if we have not a microscopic explanation for that we believe that our results for the gap and the local bond strengths are connected and indicate a change of regime for the above value of \( J_\perp \).

For the sake of completeness, we studied the correlation functions along the legs. In Fig. 5(a), we compare the spin correlations of the USL with the three possible correlations of the mixed spin case, corresponding to \( J_\perp = 0.1 \) and \( J_\perp = 1.0 \). In the inset it is shown in a semilog plot the exponential behaviour of the correlations with an estimated correlation length of \( \xi_{\perp,\text{USL}} \sim 25 \), and \( \xi_{\perp,\text{ASL}} \sim 30 \) lattice spacing. It can be noticed, also, that the three possible correlation lengths are the same. We have checked that the correlation functions along the leg decay exponentially for the whole range of interchain coupling and the correlation length \( \xi_{\perp,\text{ASL}} \rightarrow \infty \) in the limit \( J_\perp \rightarrow 0 \) like in USL \([22]\). When \( J_\perp \) is strictly zero there is a transition to two ferrimagnetically ordered chains in the ASL. In Fig. 5(b) we show the correlations for \( J_\perp = 1 \). Again, the semilog plot (inset) shows the exponential decaying of the correlations with an estimated correlation length of \( \xi_{\perp,\text{ASL}} \sim 5 \). This value is bigger than the isotropic one \( \xi_{\perp,\text{USL}} \sim 3 \) found for the uniform case \([24, 25]\).

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IV. SUMMARY AND CONCLUDING REMARKS

In summary, we have studied the effect of AF coupling in mixed spin chains for two particular topologies, the ladder and the two dimensional system. In the latter, SW theory leads to a smooth dimensional crossover from 1D to 2D with an enhancement of the magnetizations. Furthermore, as a consequence of the crossover, we detected an important difference in the relative magnetizations, being \( m_1/S \) and \( m_2/s \) around 0.73 for \( J_\perp = 1 \), whereas \( m_1/S \sim 0.70 \) and \( m_2/s \sim 0.40 \) for \( J_\perp = 0 \). For the ladder system, where SW breaks down, we used DMRG to study the gap and the correlation functions with \( J_\perp \). For \( J_\perp \neq 0 \) the ladder system is always gapped with a quadratic behaviour \( \Delta \sim 0.6J_\perp^2 \) for \( J_\perp \) lower than 0.3, and then it becomes quite linear for greater values of the interchain coupling. The study of the local bond strengths also indicate a change of regime at \( J_\perp \sim 0.3 \). In the particular case of \( J_\perp = 1 \) we found that the gap is \( \Delta = 0.334 \) and that the correlation functions decay exponentially with a correlation length about \( \xi_{\perp,\text{ASL}} \sim 5 \).
Except for the differences found in the weakly coupled regime, our results suggest a strong similarity between the ASL and the USL. This close resemblance encourage us to make some general statement regarding an arbitrary number of mixed spin chains, between the two limiting cases we have studied. First, we can rigorously say that any odd number of chains has always a ferromagnetic ground state, due to Lieb-Mattis theorem, so, it will be gapless—but ordered–. Then, if we complement our results with the conjecture that an even number of chains will be gapped and–similarly to uniform ladders–this gap will decrease to zero in the 2D limit, it is recovered an analogous spin gap behaviour to the uniform spin-1/2 case. In both cases, however, the nature of the gapless states is completely different. We hope our findings could be tested experimentally in a near future.

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[1] Y. Pei, M. Verdaguer, O. Kahm, J. Sletten and J. P. Renard, Inorg. Chem. 26, 138 (1987); O. Kahm, Y. Pei, M. Verdaguer, J. P. Renard and J. Sletten, J. Am. Chem. Soc. 110, 782 (1988).
[2] M. Hagiwara, K. Minami, Y. Narumi, K. Tatani, and K. Kindo, J. Phys. Soc. Jpn. 67, 2209 (1998).
[3] F. C. Alacraz and A. L. Malvezzi, J. Phys. Math. Gen. 30, 767 (1997).
[4] S. K. Pati, S. Ramasesha and D. Sen, Phys. Rev. B 55, 8894 (1997)
[5] G. S. Tian, Phys. Rev. B 56, 5355 (1997).
[6] N. B. Ivanov Phys. Rev. B 57, R14024 (1998); N. B. Ivanov, Phys. Rev. B 62, 3271 (2000).
[7] S. Yamamoto, S. Brehmer adn H. J. Mikeska, Phys. Rev. B 57, 13610 (1998); Phys. Rev. B 57, R14008 (1998); S. Yamamoto, T. Fukui, K. Maisinger and U. Schollwock, Phys. Condens. Matter 10, 11033 (1998); S. Yamamoto, T. Fukui and T. Sakai, Eur. Phys. J. B 15, 211 (2000) T. Fukui, Phys. Rev. B 57, R14008 (1998).
[8] S. Yamamoto, S. Brehmer adn H. J. Mikeska, Phys. Rev. B 57, 13610 (1998).
[9] C. Wu, B. C. Xi Dai, Y. Yu and Z. B. Su, Phys. Rev. B 60, 1057 (1999).
[10] A. Koga, S. Kumada, N. Kawakami and T. Fukui, J. Phys. Soc. Jpn. 67, 622 (1998); T. Fukui and N. Kawakami, Phys. Rev. B 56, 8799 (1997); A. Koga, S. and N. Kawakami, J. Phys. Soc. Jpn. 69, 1834 (2000).
[11] Y. Takushima, A. Koga and N. Kawakami, Phys. Rev. B 61, 6133 (2000).