

Antiferromagnetic Spin- S Chains with Exactly Dimerized Ground States

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We show that spin S Heisenberg spin chains with an additional three-body interaction of the form $(\mathbf{S}_{i-1} \cdot \mathbf{S}_i)(\mathbf{S}_i \cdot \mathbf{S}_{i+1}) + \text{H.c.}$ possess fully dimerized ground states if the ratio of the three-body interaction to the bilinear one is equal to $1/[4S(S+1) - 2]$. This result generalizes the Majumdar-Ghosh point of the $J_1 - J_2$ chain, to which the present model reduces for $S = 1/2$. For $S = 1$, we use the density matrix renormalization group method to show that the transition between the Haldane and the dimerized phases is continuous with a central charge $c = 3/2$. Finally, we show that such a three-body interaction appears naturally in a strong-coupling expansion of the Hubbard model, and we discuss the consequences for the dimerization of actual antiferromagnetic chains.

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Introduction.—Over the years, exact results have proved to be extremely useful in quantum and statistical physics [1,2]. In quantum magnetism, the Bethe ansatz solution of the spin-1/2 Heisenberg chain [3] has led to the first proof that the spectrum is gapless [4], and its extensions, e.g., to the $S = 1$ chain with bilinear and biquadratic interactions (BLBQ) with equal [5–7] or opposite [8,9] amplitudes, have helped a lot to clarify the physics of that model. In quantum frustrated magnetism [10], cases where an exact expression for the ground state wave function can be obtained have also played a very important role. For instance, for the spin-1 Heisenberg chain, the exact ground state of the Affleck-Kennedy-Lieb-Tasaki point [11] has been a milestone in the confirmation of Haldane’s prediction that the spectrum of integer- S spin chains is gapped [12]. For spin-1/2 magnets, the first example of a gapped spectrum goes back to the Majumdar-Ghosh [13] (MG) point $J_2/J_1 = 1/2$ of the J_1 - J_2 model defined by the Hamiltonian:

$$\mathcal{H}_{J_1-J_2} = \sum_i (J_1 \mathbf{S}_i \cdot \mathbf{S}_{i+1} + J_2 \mathbf{S}_i \cdot \mathbf{S}_{i+2}). \quad (1)$$

At that point, the two fully dimerized states obtained as products of singlets on consecutive dimers and defined by

$$|\psi_{\text{even,odd}}\rangle = \prod_{i \text{ even,odd}} |S(i, i+1)\rangle, \quad (2)$$

where $|S(i, i+1)\rangle$ denotes the singlet formed by the spins at sites i and $i+1$, have been shown by Majumdar and Ghosh to be exact ground states. Building on this result, it has been shown that the spectrum is gapped, and that this point is representative of an extended phase that covers the parameter range $0.2411 < J_2/J_1 < +\infty$ [14–16]. This seminal result has been at the origin of a long series of experimental investigations of frustrated spin-1/2 chains

which started about 20 years ago with CuGeO_3 and which remains a very active field of research [17].

Attempts at generalizing the MG point to come up with a realistic model with fully dimerized states as exact ground states for larger spins have failed so far. The simplest idea is to consider the model of Eq. (1) for spins $S \geq 1$ [18]. It is easy to convince oneself that the dimerized states of Eq. (2) remain exact eigenstates for any spin when $J_2/J_1 = 1/2$, but for $S \geq 1$ they are no longer ground states. The problem can be traced back to the properties of a single triangle into which the Hamiltonian of Eq. (1) can be decomposed for $J_2/J_1 = 1/2$: For $S = 1/2$, the product of a singlet built out of two spins times any state of the third spin is a ground state. For $S \geq 1$, the same state has a total spin S , and it is not the ground state, which has total spin 0 or $1/2$ for integer and half-integer S , respectively.

Following Klein [19], an interesting alternative consists in building Hamiltonians as sums of local projectors on three spins to ensure that the product of a singlet with a single spin state is a local ground state. The simplest Hamiltonian of that kind takes the form [20]

$$\mathcal{H}_{\text{Klein}} = - \sum_i P_{S_{\text{tot}}=S}^{i,i+1,i+2}, \quad (3)$$

where $P_{S_{\text{tot}}=S}^{i,i+1,i+2}$ is the projector on the subspace of total spin S [21]. This projector can be written as

$$P_{S_{\text{tot}}=S}^{i,i+1,i+2} = \prod_{\sigma \neq S} \frac{(\mathbf{S}_i + \mathbf{S}_{i+1} + \mathbf{S}_{i+2})^2 - \sigma(\sigma+1)}{S(S+1) - \sigma(\sigma+1)}, \quad (4)$$

where the product runs from 0 or $1/2$ for integer or half-integer spins to $3S$. For $S = 1/2$, this Hamiltonian reduces to the MG point of the J_1 - J_2 chain, but for $S \geq 1$, it is a polynomial in scalar products of pairs of spins of degree $3S$ or $3S - 1/2$ for integer or half-integer spins, hence a very complicated Hamiltonian that seems difficult to realize in actual systems. The same remark applies to a spin-3/2

model recently investigated by Rachel [22], whose ground states are partially dimerized valence bond solid states, or to the generalizations proposed by Rachel and Greiter [23] that lead to exactly trimerized and tetramerized ground states for $S = 1$ and $S = 3/2$ models, respectively.

In this Letter, we propose another generalization to arbitrary S of the spin-1/2 J_1 - J_2 model defined by the Hamiltonian,

$$\mathcal{H} = \sum_i (J_1 \mathbf{S}_i \cdot \mathbf{S}_{i+1} + J_3 [(\mathbf{S}_{i-1} \cdot \mathbf{S}_i)(\mathbf{S}_i \cdot \mathbf{S}_{i+1}) + \text{H.c.}]), \quad (5)$$

with $J_1 > 0$. The number of sites N is assumed to be even, and we concentrate on periodic boundary conditions [24]. As we shall see, this Hamiltonian possesses, for any value of S , the equivalent of a MG point when $J_3/J_1 = 1/[4S(S+1) - 2]$, at which the states of Eq. (2) are exact ground states, and it is realistic in the sense that it appears to the next-to-leading order in the $1/U$ expansion of the two-band Hubbard model that leads to the $S = 1$ Heisenberg model.

For $S = 1/2$, it is easy to check that the Hamiltonian of Eq. (5) reduces to that of Eq. (1) with $J_2 = J_3/2$. For $S \geq 1$, the three-spin interaction does not reduce to a next-nearest neighbor two-spin interaction, and the proof that the states of Eq. (2) are exact eigenstates is not a trivial extension of the MG proof.

As in the $S = 1/2$ case, let us first determine under which condition the states of Eq. (2) might be exact eigenstates of Eq. (5). To be specific, let us consider $|\psi_{\text{odd}}\rangle$. For i odd, $\mathbf{S}_i \cdot \mathbf{S}_{i+1} |\psi_{\text{odd}}\rangle = -S(S+1) |\psi_{\text{odd}}\rangle$. By contrast, for i even, the singlets on bonds $(i-1, i)$ and $(i+1, i+2)$ are affected by $\mathbf{S}_i \cdot \mathbf{S}_{i+1}$. However, the resulting wave function does not contain states with arbitrary spin for the pairs $(i-1, i)$ and $(i+1, i+2)$, but only triplets. Indeed, for two spins \mathbf{S}_1 and \mathbf{S}_2 , $S_1^\alpha |S(1, 2)\rangle$ is a triplet for all spin components $\alpha = x, y, z$. This is clear for the z component since the $\text{SU}(2)$ commutation relations imply that

$$\begin{aligned} (S_1^- + S_2^-)^2 S_1^z |S(1, 2)\rangle &= 0, \\ (S_1^+ + S_2^+)(S_1^- + S_2^-) S_1^z |S(1, 2)\rangle &= 2S_1^z |S(1, 2)\rangle, \end{aligned}$$

and by rotational symmetry, this has to be true of the other components as well. So, for i even, one can write

$$\begin{aligned} \mathbf{S}_i \cdot \mathbf{S}_{i+1} |\psi_{\text{odd}}\rangle &= \sum_{\sigma, \sigma'} C_{\sigma, \sigma'} |T_\sigma(i-1, i)\rangle |T_{\sigma'}(i+1, i+2)\rangle \\ &\times \prod_{j \text{ odd}} |S(j, j+1)\rangle, \end{aligned}$$

where the product over j is limited to $j \neq i-1, i+1$, and where the indices $\sigma, \sigma' = 0, \pm 1$ keep track of the three possible triplets of a pair of spins. Since the total wave function is a singlet, all coefficients must be equal to zero except $C_{1,-1}$, $C_{-1,1}$, and $C_{0,0}$, which must be related by $C_{1,-1} = C_{-1,1} = -C_{0,0}$. Their common absolute value can

be derived with the help of Clebsch-Gordan coefficients, but this is unimportant for our present purpose. The only relevant fact is that, since only triplets are involved, acting with $\mathbf{S}_{i-1} \cdot \mathbf{S}_i$ or $\mathbf{S}_{i+1} \cdot \mathbf{S}_{i+2}$ on $\mathbf{S}_i \cdot \mathbf{S}_{i+1} |\psi_{\text{odd}}\rangle$ will just multiply it by $1 - S(S+1)$. This leads to

$$\begin{aligned} \mathcal{H} |\psi_{\text{odd}}\rangle &= -\frac{J_1 N}{2} S(S+1) |\psi_{\text{odd}}\rangle + \{J_1 - [4S(S+1) \\ &\quad - 2]J_3\} \sum_{i \text{ even}} \mathbf{S}_i \cdot \mathbf{S}_{i+1} |\psi_{\text{odd}}\rangle. \end{aligned} \quad (6)$$

If $J_3/J_1 = 1/[4S(S+1) - 2]$, the second term drops, and $|\psi_{\text{odd}}\rangle$ is an eigenstate of \mathcal{H} with an energy per site $-J_1 S(S+1)/2$. Since the Hamiltonian is translationally invariant, this is also true for $|\psi_{\text{even}}\rangle$.

To prove that these states are the ground states, let us decompose the Hamiltonian as $\mathcal{H} = J_1 \sum_i \mathcal{H}_i$ with

$$\begin{aligned} \mathcal{H}_i &= \frac{1}{2} (\mathbf{S}_{i-1} \cdot \mathbf{S}_i + \mathbf{S}_i \cdot \mathbf{S}_{i+1}) \\ &\quad + \frac{1}{4S(S+1) - 2} [(\mathbf{S}_{i-1} \cdot \mathbf{S}_i)(\mathbf{S}_i \cdot \mathbf{S}_{i+1}) + \text{H.c.}]. \end{aligned} \quad (7)$$

The spectrum of this three-spin Hamiltonian can be worked out analytically for $S = 1$ and numerically for larger spin, with the result that the ground state energy $E_{\text{GS}}(\mathcal{H}_i)$ is equal to $-S(S+1)/2$ (see Fig. 1). By the variational principle, $\langle \mathcal{H} \rangle \geq J_1 \sum_i E_{\text{GS}}(\mathcal{H}_i) = -NJ_1 S(S+1)/2$, a lower bound saturated by $|\psi_{\text{odd}}\rangle$ and $|\psi_{\text{even}}\rangle$. This completes the proof that they are ground states of the Hamiltonian of Eq. (5) when $J_3/J_1 = 1/[4S(S+1) - 2]$.

Finally, it is plausible that these are the only ground states since the only ground states of \mathcal{H}_i are the wave functions with a singlet $|S(i-1, i)\rangle$ or $|S(i, i+1)\rangle$, and the only common eigenstates are given by $|\psi_{\text{odd}}\rangle$ and $|\psi_{\text{even}}\rangle$. However, a mathematically rigorous proof that these are the only ground states for infinite systems would require an analysis similar to that of Ref. [11] for the MG point of the spin-1/2 J_1 - J_2 model.

Vicinity of the MG point for $S = 1$.—We now concentrate on the $S = 1$ model. At the Heisenberg point $J_3 = 0$, the system is in the Haldane phase, which is gapped but not

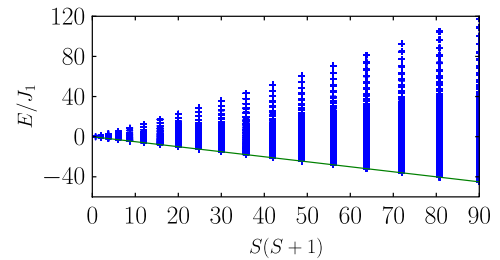


FIG. 1 (color online). Spectrum of the Hamiltonian \mathcal{H}_i [Eq. (7)] on three adjacent sites of the chain as a function of $S(S+1)$. The solid (green) line indicates the energy of the dimerized eigenstate on this three-site system, $E = -S(S+1)/2$.

dimerized. Therefore, a phase transition has to appear between the MG point $J_3/J_1 = 1/6$ and the Heisenberg point. Let us investigate the nature of this transition numerically using the density matrix renormalization group method (DMRG) [25,26].

The natural order parameter of this transition is the dimerization operator defined by $d = |\langle \mathbf{S}_i \cdot \mathbf{S}_{i+1} - \mathbf{S}_i \cdot \mathbf{S}_{i-1} \rangle|$ where $(i, i+1)$ is the central bond. Results for sizes up to 250 sites [27] are shown in Fig. 2. At the MG point, d is exactly equal to 2 for all sizes. The dimerization develops around $J_3/J_1 = 0.11$ in a way typical of a continuous transition. Assuming this to be the case, we have performed a finite-size scaling in the vicinity of the critical point, and we have identified the point where the dimerization decays to zero algebraically. This occurs at $J_3/J_1 = 0.111(1)$ (middle panel of the inset of Fig. 2).

This is further corroborated by our results for the correlation length, which we have obtained by fitting the exponential decay of the spin-spin correlation function with $x^{-1/2} \exp(-x/\xi)$. The results up to 250 sites shown in Fig. 3 are consistent with a divergence at $J_3/J_1 \approx 0.11$. At the MG point, the correlation vanishes rigorously for all sizes. Together with the results for the dimerization, we therefore conclude that the transition is located at $J_3/J_1 \approx 0.111$. In the Supplemental Material [28], we also report on a scaling analysis of the fidelity susceptibility [29] that agrees with this estimate.

Let us now try to further characterize the universality class of this phase transition. To this end, we have computed the central charge c from the block entropy of the system, $S_\ell = -\text{Tr} \rho_\ell \ln \rho_\ell$, with ρ_ℓ the reduced density matrix of a subsystem of size ℓ . For a gapless system,

$$S_\ell = \frac{c}{3} \ln \left[\frac{L}{\pi} \sin \left(\frac{\pi \ell}{L} \right) \right] + g_{\text{PBC}}, \quad (8)$$

in the presence of periodic boundary conditions, so that the central charge c is obtained by fitting the numerical results to Eq. (8) [30]. Results for 50, 80, and 100 sites [31] are shown in Fig. 4. They point rather convincingly to

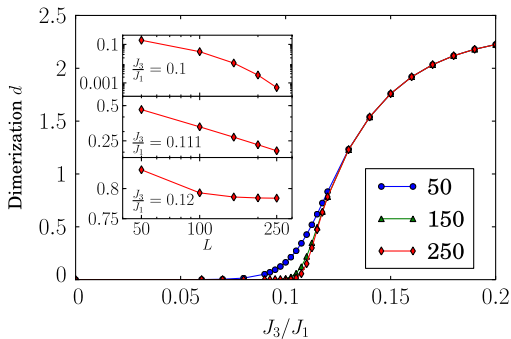


FIG. 2 (color online). Dimerization as a function of J_3/J_1 for different system sizes up to $L = 250$ sites in the vicinity of the phase transition $J_3/J_1 \approx 0.11$. The insets show the size dependence at $J_3/J_1 = 0.1$, 0.111 , and 0.12 , respectively.

$c = 3/2$. This suggests that the transition might be in the $SU(2)_{k=2}$ Wess-Zumino-Witten-Novikov (WZWN) universality class [32], as the Takhtajan-Babujian point of the $S = 1$ BLBQ chain, at which a transition from a gapped Haldane phase to a gapped dimerized phase takes place [8,9].

To further test this conclusion, we have attempted to determine the scaling dimensions at the critical point which determine the exponents of the algebraic decay of the spin and quadrupolar correlation functions [33,34]

$$\begin{aligned} C_S(i, j) &\equiv \langle S_i^z S_j^z \rangle \sim (-1)^{i-j} (i-j)^{-(1/4-\pi)/(2\alpha^2)}, \\ C_Q(i, j) &\equiv \langle \frac{1}{2} (S_i^+)^2 (S_j^-)^2 + \text{H.c.} \rangle \sim (i-j)^{-2\pi/\alpha^2}. \end{aligned} \quad (9)$$

For the $SU(2)_{k=2}$ WZWN transition, $\alpha = \sqrt{\pi}$, i.e., the correlation functions decay with exponents $3/4$ and 2 , respectively. A fit to the DMRG data at $J_3/J_1 = 0.111$ leads to exponents 0.72 and 1.83 for the corresponding correlation functions (see the inset of Fig. 3), in reasonable agreement with the field theory prediction. Furthermore, the finite-size scaling of the correlation length at the critical point is linear to a very good accuracy, which indicates that $\nu = 1$. Finally, at the critical point we find $d \propto L^{-0.47}$, implying $\beta/\nu \approx 0.47$, hence $\beta \approx 0.47$ since $\nu = 1$. In a related model, Nersisyan and Tselik [35] have predicted that the dimerization order parameter can be described as the product of four Ising fields. Three of them are ordered in the dimerized phase, one is disordered, and they are all critical at the transition point. Since the Ising exponent β is equal to $1/8$, we expect the product of four critical Ising fields to scale with exponent $\beta = 1/2$. Again, the numerical estimate is in reasonable agreement with this prediction [36].

We therefore safely conclude that the MG point is representative of an extended phase which is separated from the Haldane phase by a continuous phase transition at $J_3/J_1 \approx 0.111$, and which extends to large values of J_3 , as in the $S = 1/2$ case [16]. The results are summarized in the phase diagram of Fig. 5.

Discussion.—Finally, let us discuss the implications of the present results for actual spin chains. For simplicity,

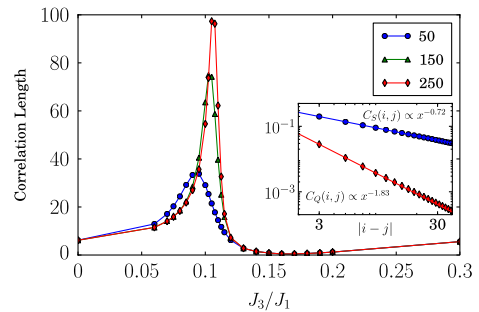


FIG. 3 (color online). Correlation length as a function of J_3/J_1 for different system sizes. Inset: Spin and quadrupolar correlation functions of Eq. (9) at the critical point $J_3/J_1 = 0.111$.

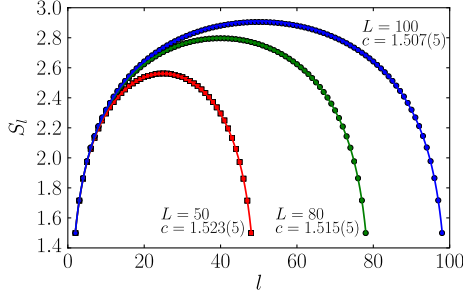


FIG. 4 (color online). Fit of the Calabrese and Cardy formula [Eq. (8), continuous line] to the DMRG results (dots) for the block entropy at $J_3/J_1 = 0.111$.

we concentrate on spin-1 chains [37]. Starting from a two-orbital Hubbard model with repulsion U and Hund's rule coupling, a strong-coupling expansion leads, to second order in the hopping integrals, to the $S = 1$ Heisenberg model with bilinear coupling J_1 . At fourth order, three extra terms appear: the three-body interaction J_3 of Eq. (5), a next-nearest neighbor bilinear coupling J_2 , as in Eq. (1), and a biquadratic interaction $J_{\text{biq}}(\mathbf{S}_i \cdot \mathbf{S}_{i+1})^2$ (see Supplemental Material [28]). The nature of the phase induced by these terms will depend on the microscopic parameters, but a reasonable case in favor of a spontaneous dimerization in a realistic parameter range can be articulated around four points: (1) The J_3 coupling generated by the fourth-order perturbation theory is essentially always positive. (2) The critical ratio for dimerization $J_3/J_1 = 0.111$ is quite small and can be reached for reasonable values of U . (3) The biquadratic interaction may be positive or negative. If it is negative, it favors dimerization. If it is positive, it is typically of the same order as J_3 , and preliminary results show that it should be significantly larger than J_3 to suppress dimerization. (4) To fourth order, the next-nearest neighbor interaction is essentially ferromagnetic, and this would compete with dimerization. However, in actual antiferromagnets, it is, in fact, more likely to be antiferromagnetic due to the residual direct superexchange, hence, to be compatible with dimerization. So, we believe that the dimerization mechanism described by the model of Eq. (5) is a realistic potential source of dimerization in actual antiferromagnetic spin chains. We also note that for systems of ultracold alkaline earth atoms on optical lattices, higher order perturbation theory leads to the three-body term of Eq. (5) as well [38,39]. In actual systems, this dimerization should be observable provided that the interchain coupling and the temperature are both smaller than an energy scale of the order of the gap, a reasonable condition since the gap at the Majumdar-Ghosh point is expected to be a significant fraction of J_1 (see Supplemental Material for a detailed discussion [28]).

Conclusions.—We have shown that it is possible to generalize the spin-1/2 J_1 - J_2 model to larger spins in

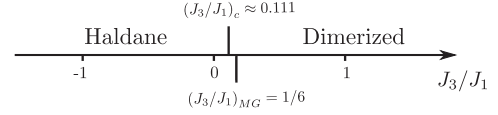


FIG. 5. Phase diagram of the J_1 - J_3 chain of Eq. (5) for $S = 1$.

such a way that a Majumdar-Ghosh point, where dimerized states are exact ground states, is still present without making the model unrealistically complicated. For spin 1, the additional interaction is a three-site term that appears naturally at fourth order in a $1/U$ expansion of a two-band Hubbard model, and we have also shown that the MG point is representative of an extended dimerized phase separated from the Haldane phase by a continuous transition in the $SU(2)_{k=2}$ WZN universality class. We hope that this new model will motivate the search for experimental realizations in quantum magnets and cold atoms.

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