Phase diagram of the spin-one Heisenberg system with dimerization and frustration

EUROPHYSICS LETTERS 20 March 1996

Europhys. Lett., **33** (9), pp. 707-712 (1996)

Phase diagram of the spin-one Heisenberg system with dimerization and frustration

S. Pati[1], R. Chitra[2], D. Sen[3,4], H. R. Krishnamurthy[2,4] and S. Ramasesha[1,4]

[1] *Solid State and Structural Chemistry Unit, Indian Institute of Science Bangalore 560012, India*

[2] *Physics Department, Indian Institute of Science - Bangalore 560012, India*

[3] *Centre for Theoretical Studies, Indian Institute of Science - Bangalore 560012, India*

[4] *Jawaharlal Nehru Centre for Advanced Scientific Research Indian Institute of Science Campus - Bangalore 560012, India*

(received 15 September 1995; accepted 29 January 1996)

PACS. 75.10Jm – Quantized spin models.
PACS. 75.50Ee – Antiferromagnetics.

Abstract. – We use the density matrix renormalization group method to study the ground-state "phase" diagram and some low-energy properties of an antiferromagnetic spin-1 chain with a next-nearest-neighbour exchange J_2 and an alternation δ of the nearest-neighbour exchanges. We find a line running from a gapless point at $(J_2, \delta) = (0, 0.25 \pm 0.01)$ up to a "gapless" point at $(0.73 \pm 0.005, 0)$ such that the open-chain ground state is fourfold degenerate below the line and is unique above it. A disorder line $2J_2 + \delta = 1$ runs from $\delta = 1$ to about $\delta = 0.136$. To the left of this line, the peak in the structure factor $S(q)$ is at π, while to the right of the line, it is at less than π.

While the spin-(1/2) Heisenberg antiferromagnetic chain has been extensively studied using a variety of techniques [1], the corresponding spin-1 chain has been studied in much less detail [2]-[5]. Interest in spin-1 chains grew after Haldane's conjecture that integer spin chains should have a gap while half-integer spin chains should be gapless. This observation was based on a non-linear sigma-model (NLSM) field theory [6]. This approach can be generalized to include dimerization (an alternation δ of the nearest-neighbour (nn) exchanges) and a next-nearest-neighbour (nnn) exchange J_2 [7], and it leads to interesting predictions. For instance, the spin-1 model should be gapless at some critical value of δ. If the nnn exchange is large enough, the spin chain goes over from a Néel-like "phase" to a spiral "phase" ([1]) and a different kind of NLSM field theory is required [8]. This predicts a gap for *all* values of the spin.

([1]) We use the word "phase" only for convenience to distinguish between regions with different modulations of the two-spin correlation function. The model actually has no phase transition from Néel to spiral even at zero temperature.

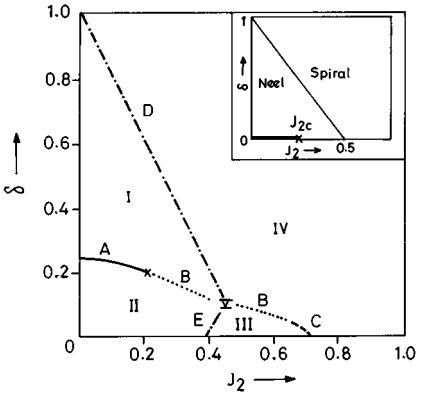

Fig. 1. – "Phase" diagram for the spin-1 chain in the (J_2, δ)-plane. The inset shows the "phase" diagram for the spin-(1/2) chain. The various features are explained in the text.

Real spin-(1/2) Heisenberg systems with both dimerization and frustration are now known [9]. However, the spin-1 analogues are yet to be synthesized. In what follows, we demonstrate that a spin-1 system exhibits a very rich "phase" diagram. It is hoped that this will provide motivation for experimental realizations of such systems.

In this letter, we study a spin-1 chain with both dimerization and frustration using the density matrix renormalization group (DMRG) method [2]. We compare our results with field-theoretic expectations as well as our recent study of the J_2-δ model for a spin-(1/2) chain [10]. The major surprise which we discover is a "gapless" (to numerical accuracy) point at $(J_2 = 0.73,\ \delta = 0)$ which is contrary to the field theory expectation. We suggest that this point may be close to a critical point described by a conformal field theory (CFT) [11], [12].

We consider chains with an even number of sites with the Hamiltonian

$$H = \sum_i [1 - (-1)^i \delta] \mathbf{S}_i \cdot \mathbf{S}_{i+1} + J_2 \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+2}\,, \tag{1}$$

where $J_2 \geq 0$ and $0 \leq \delta \leq 1$. We study various regions in the (J_2, δ)-plane using the DMRG. The DMRG involves building up the chain to a desired number of sites starting from a very short chain by adding two sites at a time. The initial chain of $2n$ sites (with n a small integer) is diagonalized exactly. The density matrix (DM) for the left n sites is computed from the ground state of the $2n$-site Hamiltonian by integrating over the states of the right n sites. This DM is diagonalized, and the n-site Hamiltonian is obtained in a truncated basis with m basis vectors which are the eigenvectors of the DM corresponding to its m largest eigenvalues. The Hamiltonian for the $2n+2$ chain is obtained in the $(2s+1)^2 m^2$-dimensional direct-product subspace constructed using the basis of the left and the right halves of the $2n$ chain and the full space of the two additional spins which are inserted in the middle. After obtaining the ground state of the $2n+2$ chain, the DM of half the chain, now with $n+1$ sites, is computed. The procedure is repeated up to the desired chain length N. The DMRG allows us to study a few low-lying states in a sector with a given value of the total spin S_z. The ground state is always the first (lowest-energy) state in the $S_z = 0$ sector. The accuracy of the DMRG depends crucially on the number of eigenvalues m of the DM which are retained. We have worked with $m = 100$ to 120 over the entire (J_2, δ)-plane after checking that the DMRG results obtained with these values of m agree well with exact numerical diagonalizations of chains with up to 16 sites [4]. The chain lengths we studied varied from 150 sites for $J_2 > 0$ to 200 sites for

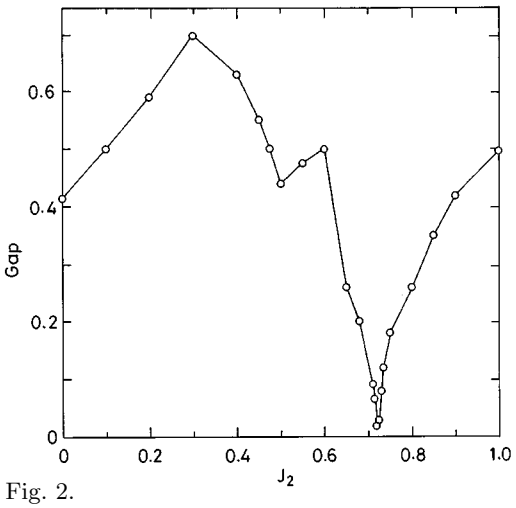

Fig. 2.

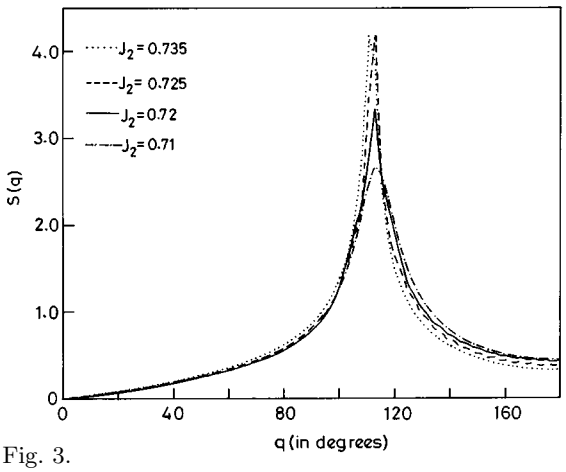

Fig. 3.

Fig. 2. – Dependence of the gap on J_2 for $\delta = 0$.

Fig. 3. – Structure factor $S(q)$ *vs.* q for $J_2 = 0.71$, 0.72, 0.725 and 0.735 at $\delta = 0$.

$J_2 = 0$. We tracked our results as a function of N to check that convergence had been reached well before 150 sites. We find that the numerical results are much better convergent for open chains than for periodic chains, a feature generic to DMRG studies [13], [14]. Hence all the data shown in fig. 1 to 3 are based on open-chain results.

The "phase" diagram obtained is shown in fig. 1. A solid line marked A runs from $(0, 0.25)$ to about $(0.22 \pm 0.02, 0.20 \pm 0.02)$ shown by a cross. Within our numerical accuracy, the gap is zero on this line and the correlation length ξ is as large as the system size N. The rest of the phase diagram is gapped. However, the gapped portion can be divided into different regions characterized by other interesting features. On the dotted lines marked B, the gap is finite and ξ is much smaller than N. On the dashed line C extending from $(0.65, 0.05)$ to about $(0.73, 0)$, the gap appears to be zero (to numerical accuracy), and ξ is very large but not as large as N. In regions II and III, the ground state for an *open* chain has a fourfold degeneracy, whereas it is non-degenerate in regions I and IV. The dashed line marked D is defined by $2J_2 + \delta = 1$, has an exactly dimerized ground state, and extends from $(0, 1)$ to about $(0.432, 0.136)$. The line E separating regions II and III begins at about $(0.39, 0)$ and extends up to region V. In regions I and II, the peak in the structure factor is at π (Néel), while in regions III and IV, the structure factor peak is at less than π (spiral).

The phase diagram of the spin-1 chain is much more complicated than that of a spin-(1/2) chain shown in the inset of fig. 1. For spin-(1/2), there is a gapless line from $(0, 0)$ to $(0.24, 0)$. The rest of the parameter space is gapped. The line $2J_2 + \delta = 1$ extends throughout the (J_2, δ)-plane and separates the Néel "phase" from the spiral "phase".

For reasons explained below, the "gapless" point at $(0.73, 0)$ is quite unexpected. So we examine that point more in detail. Figure 2 shows a plot of the gap *vs.* J_2 for $\delta = 0$. It is non-monotonic and is "gapless" at about $J_2 = 0.73$. In regions II and III, *i.e.* for $J_2 \leq 0.735$, the open-chain ground state is found to be fourfold degenerate. By comparing the energies of the low-lying states in sectors with $S_z = 0, 1$ and 2, we find that the four ground states have $S = 0$ and 1. We therefore define the gap as the energy difference between the first state in the $S_z = 0$ sector and the *second* state with $S_z = 1$, since the gap to the first state with $S_z = 1$

is zero. This is the correct definition of the gap since the finite ground-state degeneracy does not contribute to thermodynamic properties. In region IV, *i.e.* for $J_2 > 0.735$, the ground state is found to be unique with $S = 0$. So the gap is defined as the energy difference between the first states in the $S_z = 0$ and $S_z = 1$ sectors. In all cases, we extrapolate the gap Δ to infinite system size by fitting it to N through the formula $\Delta = A + B/N^{\alpha}$, and finding the best possible values of A, B and α for each J_2. For $J_2 \leq 0.735$, we also studied the separation between the second and the third states with $S_z = 1$. This separation is appreciably larger than the extent of non-monotonicity in the region around $J_2 = 0.5$. This confirms that the non-monotonic behaviour in that region is not an artifact arising from convergence of the DMRG to higher excited states.

Figure 3 is a plot of the static structure factor $S(q)$ *vs.* q at four values of J_2 near 0.73 obtained from open-chain studies with 150 sites [14]. For J_2 between 0.725 and 0.735, we see a pronounced peak at about $q_{\max} = 112°$. The peak decreases in height and becomes broader as one moves away from this interval. We estimate the maximum value of ξ to be about 60 sites. (Interestingly, Tonegawa *et al.* [4] did find a pronounced peak in $S(q)$ at $J_2 = 0.7$, but they did not investigate it further.) It is natural to speculate that $(0.73, 0)$ lies close to some critical point which exists in a bigger parameter space. We believe that the appropriate critical point may be the one discussed in ref. [11], [12]. Reference [11] solves a spin-1 chain with nn interactions of the form

$$H = \sum_i [\mathbf{S}_i \cdot \mathbf{S}_{i+1} + (\mathbf{S}_i \cdot \mathbf{S}_{i+1})^2] , \tag{2}$$

and finds gapless modes at $q = 0$ and $\pm 120°$. This implies a peak in the structure factor at $q = 120°$ which is not very far from the value we observe numerically. Reference [12] argues that the long-distance physics of this model is described by a CFT with $SU(3)$ symmetry ([2]).

The field-theoretic analysis of spin chains with the inclusion of J_2 and δ proceeds as follows. In the $S \to \infty$ limit, a classical treatment shows that the ground state is in the Néel phase for $4J_2 + \delta^2 < 1$, and in a spiral phase for $4J_2 + \delta^2 > 1$. To next order in $1/S$, one derives a field theory to describe the long-wavelength low-energy excitations. The field theory in the Néel phase is the O(3) NLSM with a topological term [6], [7]. The Lagrangian is

$$\mathcal{L} = \frac{1}{2cg^2}\dot{\boldsymbol{\phi}}^2 - \frac{c}{2g^2}\boldsymbol{\phi}'^2 + \frac{\theta}{4\pi}\boldsymbol{\phi} \cdot \boldsymbol{\phi}' \times \dot{\boldsymbol{\phi}} , \tag{3}$$

where $\boldsymbol{\phi}^2 = 1$, $c = 2S(1 - 4J_2 - \delta^2)^{1/2}$ is the spin-wave velocity, $g^2 = 2/[S(1 - 4J_2 - \delta^2)^{1/2}]$ is the coupling constant, and $\theta = 2\pi S(1 - \delta)$ is the coefficient of the topological term. (Dots and primes denote time and space derivatives, respectively.) Note that θ is independent of J_2 in the NLSM. For $\theta = \pi \bmod 2\pi$ and g^2 less than a critical value, the system is gapless and is described by a CFT with an $SU(2)$ symmetry [7], [12]. For any other value of θ, the system is gapped. For $J_2 = \delta = 0$, one therefore expects integer (half-integer) spin chains to be gapped (gapless). This is known to be true even for small values of S like 1/2 (analytically) and 1 (numerically) although the field theory is only derived for large S. In the presence of dimerization, one expects a gapless system at certain special values of δ. The special value is predicted to be $\delta_c = 0.5$ for $S = 1$. We see that the *existence* of a gapless point is correctly predicted by the NLSM. However, according to the DMRG results, δ_c is at 0.25 for $J_2 = 0$ [3] and decreases with J_2 (see fig. 1). These deviations from field theory are probably due to higher-order corrections in $1/S$ which have not been studied analytically so far.

([2]) At the $SU(3)$ symmetric critical point, the two-spin correlation should asymptotically decay as the 4/3 power of the distance [12]. We tried to verify this but the finite correlation length prevented us from obtaining an accurate estimate of the power.

In the spiral phase, it is necessary to use a different NLSM which is known for $\delta = 0$ [8]. The field variable is an $SO(3)$ matrix $\underline{R}$ and the Lagrangian is

$$\mathcal{L} = \frac{1}{2cg^2}\mathrm{tr}\Big(\dot{\underline{R}}^T\,\dot{\underline{R}}P_0\Big) - \frac{c}{2g^2}\mathrm{tr}\Big(\underline{R}'^T\underline{R}'P_1\Big), \tag{4}$$

where $c = S(1+y)\sqrt{1-y^2}/y$, $g^2 = 2\sqrt{(1+y)/(1-y)}/S$ with $1/y = 4J_2$; P_0 and P_1 are diagonal matrices with elements $(1, 1, 2y(1-y)/(2y^2-2y+1))$ and $(1, 1, 0)$, respectively. Since there is no topological term, there is no apparent difference between integer and half-integer spin chains. A one-loop renormalization group and large-N analysis [8] indicate that the system should have a gap for all values of J_2 and S, and there is no reason for a particularly small gap at any special value of J_2. The "gapless" point at $J_2 = 0.73$ for spin-1 is therefore surprising.

For $\delta < 0.25$ and $J_2 = 0$, the spin-1 chain is known to exhibit a "hidden" $Z_2 \times Z_2$ symmetry breaking described by a non-local order parameter [3], [15]. This leads to a fourfold degeneracy of the ground state for the open chain. The degeneracy may be understood in terms of spin-(1/2) states living at the ends of an open chain whose mutual interaction decreases exponentially with the chain length [15]. We have oberved this ground-state degeneracy at all points in regions II and III in fig. 1, where the gap between the singlet and triplet states vanishes exponentially with increasing chain length. In regions I and IV, the ground state is unique. The situation is reminiscent of the $Z_2 \times Z_2$ symmetry breaking mentioned above. However, we have not directly studied the non-local order parameter using the DMRG.

We have examined the structure factor $S(q)$. Since there is no long-range order anywhere in the (J_2, δ)-plane (except for algebraic order on the line A in fig. 1), $S(q)$ generally has a broad peak at some $q_{\max}$. In regions I and II in fig. 1, $q_{\max}$ is pinned at π, while in regions III and IV, $q_{\max} < \pi$. Above the curve ABC, the crossover from the Néel to the spiral "phase" occurs across the straight line D given by $2J_2 + \delta = 1$. Below ABC, the crossover has been determined numerically and occurs across the line E. The region of intersection between the crossovers from "Néel" to "spiral" and from fourfold degeneracy to a unique ground state is a small "hole" (region V) in the "phase" diagram centred about the point $(0.435, 0.12)$. Points in this "hole" turned out to be extremely difficult to study using the DMRG because of convergence difficulties with increasing chain lengths.

The segment D of the straight line $2J_2 + \delta = 1$ running from $\delta = 1$ to about $\delta = 0.136$ can be shown to have an exact ground state of the dimerized form $\psi = [1, 2][3, 4] \ldots [N-1, N]$, where $[i, j]$ denotes the singlet combination of the spins on sites i and j. (This will be shown elsewhere.) Since the segment D has an exact ground state with an extremely short correlation length (essentially, one site), and since there is a crossover from a Néel to a spiral "phase" across it, we may call D a disorder line just as in the spin-(1/2) case [10].

To summarize, we have studied a two-dimensional "phase" diagram for the ground state of an isotropic spin-1 chain. It has surprising features like a "gapless" point inside the spiral "phase". We have suggested that this point is close to a critical point of a particular kind. It would be interesting to establish this more definitively. In any case, our results show that frustrated spin chains with small values of S may exhibit features not anticipated from large-S field theories.

***

We thank B. Sriram Shastry for stimulating discussions, and Biswadeb Datta for assistance with the computer systems.

REFERENCES

[1] Tonegawa T. and Harada I., *J. Phys. Soc. Jpn.*, **56** (1987) 2153, and references therein.

[2] White S. R. and Huse D. A., *Phys. Rev. B*, **48** (1993) 3844.

[3] Totsuka K., Nishiyama Y., Hatano N. and Suzuki M., *J. Phys. Condens. Matter*, **7** (1995) 4895; Kato Y. and Tanaka A., *J. Phys. Soc. Jpn.*, **63** (1994) 1277.

[4] Tonegawa T., Kaburagi M., Ichikawa N. and Harada I., *J. Phys. Soc. Jpn.*, **61** (1992) 2890.

[5] Singh R. R. P. and Gelfand M. P., *Phys. Rev. Lett.*, **61** (1988) 2133; Guo D., Kennedy T. and Mazumdar S., *Phys. Rev. B*, **41** (1990) 9592.

[6] Haldane F. D. M., *Phys. Lett. A*, **93** (1983) 464; *Phys. Rev. Lett.*, **50** (1983) 1153.

[7] Affleck I., in *Fields, Strings and Critical Phenomena*, edited by E. Brezin and J. Zinn-Justin (North-Holland, Amsterdam) 1989; Affleck I. and Haldane F. D. M., *Phys. Rev. B*, **36** (1987) 5291.

[8] Rao S. and Sen D., *Nucl. Phys. B*, **424** (1994) 547; Allen D. and Senechal D., *Phys. Rev. B*, **51** (1995) 6394.

[9] Cava R. J. *et al.*, *J. Solid State Chem.*, **104** (1993) 437; Cava R. J. *et al.*, *J. Mater. Res.*, **9** (1994) 314; Sen D., Shastry B. S., Walstedt R. E. and Cava R., cond-mat/9508121, to appear in *Phys. Rev. B*.

[10] Chitra R., Pati S., Krishnamurthy H. R., Sen D. and Ramasesha S., *Phys. Rev. B*, **52** (1995) 6581.

[11] Sutherland B., *Phys. Rev. B*, **12** (1975) 3795.

[12] Affleck I., *Nucl. Phys. B*, **265** (1986) 409.

[13] White S. R., *Phys. Rev. Lett.*, **69** (1992) 2863; *Phys. Rev. B*, **48** (1993) 10345.

[14] Sorensen E. S. and Affleck I., *Phys. Rev. B*, **49** (1994) 15771.

[15] den Nijs M. and Rommelse K., *Phys. Rev. B*, **40** (1989) 4709; Tasaki H., *Phys. Rev. Lett.*, **66** (1991) 798; Kennedy T., *J. Phys. Condens. Matter*, **2** (1990) 5737; Affleck I., Kennedy T., Lieb E. H. and Tasaki H., *Commun. Math. Phys.*, **115** (1988) 477.