

Majumdar-Ghosh-like spin models in low dimensions

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Abstract : The Majumdar-Ghosh model occupies a special position amongst the models of interacting spin systems. The model has motivated the search for other low-dimensional spin systems with quantum paramagnetic ground states and a gap, the so-called spin gap, in the excitation spectrum. In this brief review, some generic features of MG-like spin models will be described in terms of theoretical results and experimentally observed phenomena.

Keywords : Majumdar-Ghosh model, spin gap antiferromagnets, frustration, spin ladder, quantum spin liquid

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1. Introduction

The celebrated Majumdar-Ghosh (MG) model illustrates some characteristic features of low-dimensional antiferromagnetic (AFM) quantum spin systems. This has given rise to a flurry of research activity, both theoretical and experimental, to gain insight on the unique properties of such systems. In this brief overview, a number of spin models will be discussed which are descendants of the MG model and which describe a variety of novel phenomena in interacting spin systems. We assume the magnitude of the spins to be $S = 1/2$ unless mentioned otherwise.

2. Frustrated spin systems

The MG chain is the first example of a frustrated quantum spin model for which the ground state can be determined exactly. The origin of frustration in the model lies in the presence of both nearest-neighbour (*n.n.*) as well as next-nearest-neighbour (*n.n.n.*) spin-spin interactions. The spin-spin correlations in the ground state are short-ranged but one can define a four-spin correlation function $\langle S_i^x S_j^x S_i^y S_m^y \rangle$ which has off-diagonal long range order (LRO) in the two ground states. These states, because of a coherent structure, describe a quantum paramagnetic phase. A general MG-type model in one dimension (1d) has been proposed which includes exchange couplings to $2n$ - nearest neighbours with strengths $J_1 = 2n, J_2 = 2n - 1, \dots, J_{2n} = 1$ [1, 2]. The 2d models include the Shastry-Sutherland (SS) model [3], the $J_1 - J_2, J_1 - J_2 - J_3$ and the $J_1 - J_2 - J_3 - J_4 - J_5$ models

[4-6]. The SS model is defined on a square lattice and includes *n.n.* interactions (of strength J_1) as well as alternate diagonal exchange interactions (of strength J_2) in every second square. For J_1/J_2 below a critical value ~ 0.7 , the exact ground state consists of singlets along the diagonals. At the critical point, the ground state changes from the gapped disordered state to an antiferromagnetically ordered gapless state. The AFM compound $\text{SrCu}_2(\text{BO}_3)_2$ is an experimental realization of the SS model [7]. In the frustrated $J_1 - J_2$ and $J_1 - J_2 - J_3$ models, J_1, J_2 and J_3 are the strengths of the *n.n.*, diagonal and *n.n.n.* exchange interactions respectively. The $J_1 - J_2 - J_3 - J_4 - J_5$ model includes two more exchange interactions, knight's-move-distance-away and further-neighbour diagonal of strengths J_4 and J_5 respectively. In this case, the four columnar dimer (CD) states are the exact ground states for the ratio of interaction strengths

$$J_1 : J_2 : J_3 : J_4 : J_5 = 1 : 1 : \frac{1}{2} : \frac{1}{2} : \frac{1}{4}. \quad (1)$$

In a CD state, the spin pairs in the alternate columns of the 2d square lattice are in singlet (alternatively termed as dimer or valence bond (VB)) spin configurations. There is no rigorous proof as yet that the CD states are the exact ground states though approximate theories tend to support the conjecture [6]. One can, however, prove that any one of the CD states is the exact ground state when the dimer bonds are of strength $7J$ and the rest of the exchange interactions are of strengths as specified in eq. (1).

Two well-known examples of spin-disordered but quantum coherent states are the quantum spin liquid (QSL) and the dimer or VB states. A QSL state is a spin singlet with total spin $S = 0$ and has both spin rotational and translational symmetry. A well-known example of a QSL state is the resonating-valence-bond (RVB) state which is a coherent linear superposition of VB states. The linear superposition of the two ground states of the MG model is a simple example of a RVB state in 1d. In this particular case, however, there is no energy gain due to the superposition of the VB states. In the VB states, *e.g.*, the CD states, the spin rotational symmetry is present but the translational symmetry is broken. Some quantum paramagnetic states like the chiral, dimer, twisted and strip or collinear states are characterised by the non-zero expectation values of appropriately defined order parameters [8]. A recent work classifies and analyses MG-like spin models with dimer (VB) ground states [9].

Klein [10] proposed a method of construction of MG-like spin models whose exact ground states are the n.n. VB states. In the Klein formalism, one defines a neighbourhood $N(i)$ of each site i which consists of the site itself and its $Z - 1$ n.n.s. The Klein Hamiltonian, H_{klein} , is written as a sum over projection operators $P_{\frac{Z}{2}}(\mathbf{S}_{N(i)})$ onto the maximally allowed spin state $S = \frac{Z}{2}$ of a neighbourhood.

$$H_{klein} = \sum_i P_{\frac{Z}{2}}(\mathbf{S}_{N(i)}), \quad (2)$$

where

$$\mathbf{S}_{N(i)} = \sum_{k \in N(i)} \mathbf{S}_k. \quad (3)$$

The projection operator P_j has the form

$$P_j(\mathbf{S}_{N(i)}) = \prod_{l \neq j} \frac{[\mathbf{S}_{N(i)}^2 - l(l+1)]}{[j(j+1) - l(l+1)]}, \quad (4)$$

where the product runs over the total spin values of the Z spins, other than the value j . Any n.n. VB state is an exact ground state of H_{klein} . In a VB configuration, each site of the lattice forms a VB with one of its n.n.s. The Z spins in the neighbourhood $N(i)$ can thus at most sum up to the total spin $S = \frac{Z}{2} - 1$. The projection operator $P_{\frac{Z}{2}}(\mathbf{S}_{N(i)})$ acting on the VB configuration gives zero. Since H_{klein} is a sum over projection operators, it is semi-positive definite with zero as the lowest eigenvalue. Thus the VB states are the exact ground states. In the case of the honeycomb lattice, one can show that the n.n. VB states are the only ground states [11] whereas other ground states are possible for lattices like the square and the triangular lattices. From eqs. (2)-(4), it is clear that the Klein Hamiltonian includes two-spin as

well as multi-spin interactions. The MG Hamiltonian is a special case of the Klein Hamiltonian with $Z = 3$. The Hamiltonian, apart from a numerical prefactor and a constant term, can be written as

$$H_{MG} = \sum_i P_{\frac{3}{2}}(\mathbf{S}_i + \mathbf{S}_{i+2} + \mathbf{S}_{i+1}). \quad (5)$$

3. Haldane-gap antiferromagnets

These are integer-spin 1d antiferromagnets which share common features with the spin $-1/2$ MG chain. Both have spin-disordered ground states and a gap in the excitation spectrum. From the Lieb-Schultz-Mattis theorem, one can show that the half-odd integer spin Heisenberg AFM (HAFM) chain with only n.n. spin-spin interactions, has a gapless excitation spectrum in the infinite chain length limit [12]. The theorem is not applicable to integer spin chains. Haldane, based on his analysis of the nonlinear σ model mapping of the large S HAFM Hamiltonian in 1d, conjectured that the HAFM spin chains with integer spins have a gap in the excitation spectrum. Haldane's conjecture has now been verified both theoretically and experimentally [13]. Affleck, Kennedy, Lieb and Tasaki (AKLT) constructed a spin-1 model in 1d the exact ground state of which is the valence bond solid (VBS) state [13]. In the case of the MG model, the ground state has broken translational symmetry. In the VBS state, the symmetry is unbroken and the ground state is non-degenerate. The state can be described as a true QSL. Each spin 1 can be considered as a symmetric combination of the two spin $-1/2$'s. In the VBS state, each spin $-1/2$ component of a spin 1 forms a singlet (VB) with a spin $-1/2$ at a neighbouring site. The principle behind the construction of the AKLT Hamiltonian is similar to that in the case of the Klein Hamiltonian.

$$H_{AKLT} = \sum_i P_2(\mathbf{S}_i + \mathbf{S}_{i+1}). \quad (6)$$

From (4), H_{AKLT} can be written as

$$H_{AKLT} = \sum_i \left[\frac{1}{2} (\mathbf{S}_i \cdot \mathbf{S}_{i+1}) + \frac{1}{6} (\mathbf{S}_i \cdot \mathbf{S}_{i+1})^2 + \frac{1}{3} \right]. \quad (7)$$

The presence of a VB between each neighbouring pair of sites implies that the total spin of each pair of spins cannot be 2. Thus, H_{AKLT} acting on the VBS state gives zero. The VBS state can be described as a quantum paramagnet.

One can define a non-local string order parameter which has a non-zero expectation value in the VBS state [13]. Several Haldane gap antiferromagnets have been discovered so far. Some examples are $\text{Ni}(\text{C}_2\text{H}_8\text{N}_2)_2\text{NO}_2(\text{ClO}_4)$ (NENP), $\text{Ni}(\text{C}_3\text{D}_{14}\text{N}_2)_2\text{N}_3(\text{PF}_6)$ (NDMAP), $\text{Ni}(\text{C}_5\text{H}_{14}\text{N}_2)_2\text{N}_3(\text{ClO}_4)$ (ND-MAZ) *etc* [14]. Experiments carried out on some of these compounds show that the VBS state provides the correct physical picture for understanding the observed properties.

The Haldane gap antiferromagnets exhibit rich physics in the presence of strong magnetic fields. In the absence of the magnetic field, the lowest excitations are a $S = 1$ triplet separated from the ground state by an energy gap. In the presence of the magnetic field, there is a Zeeman splitting of the excited levels. As the field increases, the energy gap of the lowest mode decreases and vanishes at a critical value of the field. This is accompanied by a 1d Bose condensation of magnons [14]. Two possible scenarios have been suggested at higher fields: a quantum critical state with power-law correlations and a state with AFM LRO. The nickel-based organometallic compound, with abbreviated name NTENP, represents a $S = 1$ chain with exchange interactions of alternating strength. The alternation parameter is $\delta = (J_1 - J_2)/(J_1 + J_2)$, where J_1 and J_2 are the strengths of the alternate n.n. exchange interactions. As δ increases from zero, the energy gap decreases and vanishes at a critical value δ_c . The spin gap is reopened beyond this quantum critical point. The ground state is no longer the VBS state but a dimerized one in which two VBs form on each strong link.

4. Spin ladders

The simplest ladder model consists of two chains coupled by rungs. More generally, a ladder may consist of n chains. Ladders provide a bridge between 1d and 2d many body systems and are ideally suited to study how physical properties change as one goes from a single chain to the square lattice limit [15, 16]. In the spin ladder model, each site of the ladder is occupied by a spin and the spins interact *via* the HAFM exchange interaction Hamiltonian. The intra-chain n.n. and the rung exchange interactions are of strength J and J_R respectively. When $J_R = 0$, there is a decoupling of the chains for which the excitation spectrum is known to be gapless. When $J_R \neq 0$, an energy gap opens up provided the number of chains in the ladder is even. For an odd number of chains, the excitation spectrum is gapless. Thus, ladders with an even number of chains belong to the general class of spin gap (SG) antiferromagnets of which the MG chain is a prime example. In such systems, the energy gap does not occur due to some anisotropy but is quantum mechanical in origin. A large number of compounds with ladder-like structure are now known. The family of compounds $\text{Sr}_{n-1}\text{Cu}_{n+1}\text{O}_{2n}$ consists of planes of weakly coupled ladders of $(n+1)/2$ chains. For $n = 3$ and 5 , one gets the two-chain and three-chain ladder compounds SrCu_2O_3 and $\text{Sr}_2\text{Cu}_3\text{O}_5$ respectively. Experimental observations on these compounds are consistent with the theoretical predictions that for an n -chain ladder, the excitation spectrum is gapped (gapless) when n is even (odd). The origin of the SG can be understood by considering a two-chain ladder in the strong coupling limit, *i.e.*, $J_R \gg J$. In the limit $J = 0$, the ground state of the ladder consists of singlets (VBs) along the rungs. The ground state energy is

$-3NJ_R/4$, where N is the number of rungs in the ladder. When J is not equal to zero but small, the corrections to the energy and the wavefunction are in powers of J so that the ground state more or less retains its form in which the VBs occupy the rungs. An $S = 1$ excitation may be created in the ladder by promoting one of the rung singlets to the $S = 1$ triplet state. The weak coupling along the chains gives rise to a propagating $S = 1$ magnon. In first order perturbation theory, the correction to the ground state energy is zero. The dispersion relation, with respect to the ground state energy, is

$$\omega(k) = J_R + J_{\text{cos}k} \quad (8)$$

where k is the momentum wave vector of the propagating excitation. The SG is given by

$$\Delta = \omega(\pi) \approx J_R - J. \quad (9)$$

Though the arguments given above strictly hold true in the strong-rung coupling limit, a SG exists for all values of $J_R > 0$.

Bose and Gayen [17] have studied a two-chain ladder model with frustrating diagonal couplings. The intra-chain and diagonal couplings are of equal strength J . For $J_R \geq 2J$, the exact ground state consists of singlets along the rungs with the energy $E_g = -3NJ_R/4$. The triplet excitation is localized and separated by an energy gap from the ground state. Xian [18] later pointed out that as long as $(J_R/J) > (J_R/J)_C \approx 1.401$ the rung dimer state is the exact ground state. At the critical point, there is a first order transition from the rung singlet state to the Haldane phase of the $S = 1$ chain. Kolezhuk and Mikeska [19] have constructed a class of generalised $S = 1/2$ two-chain ladder models for which the ground state can be determined exactly. The Hamiltonian H is a sum over plaquette (square) Hamiltonians and each such Hamiltonian contains various two-spin as well as four-spin interaction terms. Several known models with exact ground states are special cases of the generalised ladder model. These include the MG model, the AKLT model and the ladder model with diagonal couplings. The $S = 1$ spins of the AKLT chain are composed from the $S = 1/2$ spins belonging to the ladder rungs. The two degenerate ground states of the MG chain correspond to singlets along the diagonals and on the rungs respectively. Kolezhuk and Mikeska [19] introduced a toy model which is a special case of the generalised ladder model. This model has a rich phase diagram with all the phase boundaries being exact. The ladder model with diagonal couplings is obtained from the toy model for special values of the parameters. The standard spin ladder models with bilinear exchange are not integrable. For integrability, multispin interaction terms have to be included in the Hamiltonian. Some integrable ladder models have already been constructed which exhibit rich phase diagrams [20]. A recent example of a spin ladder belonging to the organic family of materials is the

compound $(\text{C}_5\text{H}_{12}\text{N})_2\text{CuBr}_4$, a ladder system with strong rung coupling $[(J_R/J \approx 3.5)]$ [21]. The phase diagram of the AFM spin ladder in the presence of an external magnetic field is particularly interesting. In the absence of the magnetic field and at $T = 0$, the ground state is a QSL with a gap in the excitation spectrum. At a field H_{c1} , there is a transition to a gapless Luttinger liquid phase ($g\mu_B H_{c1} = \Delta_{SG}$, the spin gap, μ_B is the Bohr magneton and g the Landé splitting factor). There is another transition at an upper critical field H_{c2} to a fully polarized ferromagnetic state. Both H_{c1} and H_{c2} are quantum critical points. The quantum phase transition (QPT) from one ground state to another is brought about by changing the magnetic field. Quantum effects are persistent at small finite temperatures and can be probed experimentally. In the case of the ladder system $(\text{C}_5\text{H}_{12}\text{N})_2\text{CuBr}_4$, the magnetization data obtained experimentally exhibit universal scaling behaviour in the vicinity of the critical fields H_{c1} and H_{c2} . In the gapless regime $H_{c1} < H < H_{c2}$, the ladder model can be mapped on an XXZ HAFM chain, the thermodynamic properties of which can be calculated exactly by the Bethe Ansatz. The theoretically computed magnetization M versus magnetic field H curve is in excellent agreement with the experimental data. Organic spin ladders provide ideal testing grounds for the theories of quantum phase transitions. Other organic ladder compound exhibiting QPTs are $(5\text{IAP})_2\text{CuBr}_4 \cdot 2\text{H}_2\text{O}$ and $\text{Cu}_2(\text{C}_5\text{H}_{12}\text{N}_2)_2\text{Cl}_4$ [22].

5. Conclusion and outlook

The MG model has initiated a large body of work on spin models with dimer states as ground states. For short-ranged dimers (mainly between n.n.s), there is a gap in the excitation spectrum. In experiments, the presence of the gap Δ is confirmed through the measurement of properties like susceptibility, χ , which goes to zero exponentially at low T as $\chi \sim \exp(-\Delta/k_B T)$. Some other examples of SG antiferromagnets are [23]: spin-Peierls (SP) systems and AFM compounds consisting of weakly coupled spin dimers. The SP transition generally occurs in quasi-1d AFM spin systems with half-odd integer spins and is brought about by spin-phonon coupling. Below the SP transition temperature T_{SP} , a periodic deformation of the lattice sets in such that the distances between the neighbouring spins are no longer uniform but alternate in magnitude. This results in an alternation, $J(1+\delta)$ and $J(1-\delta)$, in the strengths of the n.n. exchange interaction strengths. The ground state is dimerized in which singlet spin pairs occupy the links with enhanced exchange couplings. MG-like spin models have been utilised to explain the properties of CuGeO_3 , the first example of an inorganic compound in which the SP transition has been observed [24]. Well-known examples of AFM compounds, which can be described as crystalline networks of spin dimers, include ACuCl_3 ($A = K, \text{Ti}$) in which a spin dimer arises from two anti-

ferromagnetically coupled Cu^{2+} ions [25]. The triplet excitation created on a particular dimer propagates through the dimer network due to the weak inter-dimer exchange couplings. The field-induced 3d magnetic ordering observed in these compounds can be described in terms of a Bose-Einstein condensation of low-lying magnons. The critical point $h = h_{c1}$ separates a gapped QSL state ($h < h_{c1}$) from a field-induced magnetically ordered state ($h > h_{c1}$). In the condensed state, the state of each dimer is found to be a coherent superposition of the singlet and the $S_z = +1$ triplet states. The phase in the superposition specifies the orientation of the staggered magnetization in the plane transverse to the magnetic field direction. The number of magnons in the condensed state is not, however, infinite as magnons cannot occupy the same sites in a spin system due to a hard-core repulsion between them. The interaction restricts the number of magnons to be large but finite.

Another novel phenomenon observed in SG AFMs is that of magnetization plateaus in which the magnetization versus the magnetic field curve exhibits plateaus at certain rational values of the magnetization per site m . For general spin systems, the quantization condition can be written as [26]

$$S_U - m_U = \text{integer}, \quad (10)$$

where $S_U = nS$, n being the number of spins of magnitude S in unit period of the ground state and $m_U = nm$ is the magnetization associated with the unit cell. The quantization condition is necessary but not sufficient as not all the plateaus predicted by the condition exist in general. High-field measurements reveal the existence of magnetization plateaus in several AFM compounds. The $S = 1/2$ SG antiferromagnet $\text{SrCu}_2(\text{BO}_3)_2$ (described by the SS model) is the first example of a 2d spin system for which magnetization plateaus have been observed experimentally [27]. The triplet excitations in the SS model are almost localized. In the presence of a magnetic field and at special values of the magnetization, the triplet excitations localize into a superlattice structure to minimize the energy so that the magnetization remains constant. Momoi and Totsuka [28] have suggested that the appearance of plateaus in $\text{SrCu}_2(\text{BO}_3)_2$ is due to the transition from a superfluid to a Mott insulating state of magnetic excitations. In the presence of a magnetic field in the z -direction, the $S_z = +1$ excitation is the lowest in energy. These excitations can be regarded as bosons with a hard-core repulsion. The repulsive interaction arises from the z -component of the exchange interaction and disallows the occupation of a single dimer by more than one boson. The xy -part of the exchange interaction is responsible for the hopping of the triplet excitation to neighbouring dimers. One thus has a system of interacting bosons in which itinerancy competes with localization. The transition from itinerancy to localization is analogous to the Mott metal-insulator transition in electronic

systems. If repulsive interactions dominate, the triplet excitations (bosons) localize to form a superlattice. A direct measurement of the superlattice in $\text{SrCu}_2(\text{BO}_3)_2$ has been made by Kodama *et al* using a high-field NMR facility [29]. The superlattice corresponds to $M = m/8 = 1/8$ which requires a high magnetic field of strength 27T for its observation. Superlattice structures for the other plateaus at $M = 1/3$ and $1/4$ have not been detected as yet because of the requirement of very high magnetic fields.

A few topics not covered in this overview include: the nature of spin excitations in MG-like spin models and doped VB systems. The elementary excitations in the MG model are a pair of spin $-1/2$ ‘defects’ separating the MG ground states. The pair gives rise to a continuum of scattering excitations. A bound state of the two spin $-1/2$ s can form in a restricted range of the momentum wave vectors. The elementary excitations of the Heisenberg AFM in 1d are also a pair of spin $-1/2$ objects, the ‘spinons’. There is now a fair amount of work on spin dimer models with spin $-1/2$ excitations. Low-dimensional candidate compounds with similar excitations are now known. The study of doped SG systems like the spin ladders and the Haldane gap antiferromagnets yields lots of useful information on charge transport in a background of antiferromagnetically interacting spins. There has been a considerable amount of experimental activity on doped spin ladder compounds. The variety of phenomena exhibited by such systems include superconductivity mediated by bound pairs of holes. The superconducting transition temperature is, however, low in contrast to the case of the cuprate compounds which exhibit high temperature superconductivity when doped with holes. SG systems have recently been suggested as candidates for the realization of quantum computation and communication protocols. The spin systems considered so far include some SG antiferromagnets like the MG model, the Haldane gap chain and the two-chain spin ladder. A number of reviews exist from which more information can be picked up on the MG-like models [23, 30-32]. In summary, the MG model has spawned wide-ranging research activities in theoretical and experimental magnetism. One anticipates that similar research efforts will continue unabated in the coming years.

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