Low-Lying Excited States and Low-Temperature Properties of an Alternating Spin-1 / Spin-$\frac{1}{2}$ Chain : A DMRG study

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Abstract

We report spin wave and DMRG studies of the ground and low-lying excited states of uniform and dimerized alternating spin chains. The DMRG procedure is also employed to obtain low-temperature thermodynamic properties of the system. The ground state of a 2N spin system with spin-1 and spin-$\frac{1}{2}$ alternating from site to site and interacting via an antiferromagnetic exchange is found to be ferrimagnetic with total spin $s_G = N/2$ from both DMRG and spin wave analysis. Both the studies also show that there is a gapless excitation to a state with spin $s_G - 1$ and a gapped excitation to a state with spin $s_G + 1$. Surprisingly, the correlation length in the ground state is found to be very small from both the studies for this gapless system. For this very reason, we show that the ground state can be described by a variational *ansatz* of the product type. DMRG analysis shows that the chain is susceptible to a conditional spin-Peierls' instability. The DMRG studies of magnetization, magnetic susceptibility (χ) and specific heat show strong magnetic-field dependence. The product χT shows a minimum as a function of temperature(T) at low-magnetic fields and the minimum vanishes at high-magnetic fields. This low-field behaviour is in agreement with earlier experimental observations. The specific heat shows a maximum as a function of temperature and the height of the maximum increases sharply at high-magnetic fields. It is hoped that these studies will motivate experimental studies at high-magnetic fields.

1 Introduction

The low-dimensional magnetic systems exhibit a wide variety of exotic physical phenomena. Many of these interesting and novel phenomena were first predicted from theoretical studies on spin systems in one-dimension [1, 2, 3]. Fascinating amongst these have been

the spin-Peierls instability and the Haldane conjecture. These predictions motivated a number of experimental efforts towards synthesis of low-dimensional magnetic systems. The experimental measurements[4, 5, 7] on some of these systems have provided support for the Haldane conjecture and also for the existence of spin-Peierls instability in quasi-one-dimensional systems. While experimental studies[4, 5] of the spin-1 antiferromagnet $Ni(C_2H_8N_2)_2NO_2(ClO_4)$ clearly show the existence of Haldane gap, the measurement of the magnetic properties in a series of quasi-one-dimensional compounds confirm the presence of spin-Peirels instability at low temperatures[6]. In recent years, purely inorganic systems that show low-dimensional behaviour have also been synthesized. The compound $CuGeO_3$ has been shown to be a quasi-one-dimensional system exhibiting spin-Peierls instability[7]. Another class of compounds that have become important in recent years are the systems with spin ladders. The compound that closely approximates a spin ladder is vanadyl pyrophosphate with molecular formula $(VO)_2P_2O_7$. A spin ladder with hole doping is predicted to support hole binding based on exchange energy considerations as two holes occupying the same rung of the ladder would be energetically favoured, if the exchange constant for the rung is larger than that along the leg. These systems are hence expected to exhibit non-BCS type of superconductivity[8].

Yet another challenge in the area of molecular magnetism has been to synthesize a molecular ferromagnet. While there have been many models for ferromagnetic exchange in molecular systems[9], the actual synthesis of molecular ferromagnets has been relatively recent. 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[10]. These systems contain two transition metal ions per unit cell and have the general formula $ACu(pbaOH)(H_2O)_3.2H_2O$ with $pbaOH$=2-hydroxo-1,3-propylenebis(oxamato) and $A = Mn$, Fe, Co, Ni and belong to the alternating or mixed spin chain family[11]. These alternating spin compounds are seen to exhibit ferrimagnetic behaviour. The ferromagnetic or ferrimagnetic alignment of spins in molecular systems is usually difficult to achieve due to the diffused nature of the molecular orbitals (MOs) which the unpaired electrons occupy. The direct exchange integrals involving MOs are much smaller than that found in transition metal ions. However, even with the smaller exchange integral, we could have a ferro or ferri magnetic spin alignment if the molecules are so arranged as to yield a small intermolecular transfer integral. In such a situation, the

kinetic stabilization of the electrons is rather small and parallel alignment of the effective spin of the electrons on the molecules is favoured leading to a magnetic ground state[12]

Thermodynamic properties of these alternating spin compounds show very interesting behaviour[11, 13]. In very low magnetic fields, these systems show one-dimensional *ferrimagnetic* behaviour. The χT vs T (χ is the magnetic susceptibility and T is the temperature) plots show a rounded minimum. As the temperature is increased, χT decreases sharply, goes through a minimum before increasing gradually. The temperature at which the minimum occurs differs from compound to compound. The behaviour of field induced magnetization with temperature is also quite interesting as the ground state of the system is a magnetic state. At moderate magnetic fields, with increase in temperature, the magnetization slowly increases, shows a broad peak and then decreases. Such behaviour is conjectured to be due to irregular variation in the spin multiplicities of the energy levels of the system. Theoretically, there has been an earlier study of the alternating spin sytem in three dimensions that focuses on the dependence of Curie temperature on the anisotropy of the spins[14].

Motivation for the present study comes from the above experimental observations on the quasi-one-dimensional alternating spin systems. Theoretical studies of spin chains so far, have however been concerned mainly with the antiferromagnetic spin systems with unique site spin, exceptions being dilute spin impurity problems. In this study, we first analyse the ground and low-lying excited states of the mixed or alternating antiferromagnetic spin chain by employing a spin wave theory. The spin wave theory is, however, not as accurate as the recently developed density matrix renormalization group (DMRG) method which has proved to be the best numerical tool for one-dimensional spin systems[15]. Ground state energy per site, the spin excitation gap and the two-spin correlation functions obtained from this method have been found to be accurate to several decimal places, in the case of the spin-$\frac{1}{2}$ Heisenberg antiferromagnet which is amenable to exact study. In the DMRG method, spin parity symmetry can be used to characterize the spin states along with the $s^z_{tot.}$ as the good quantum numbers [16]. There have been many interesting studies of spin-$\frac{1}{2}$ and spin-1 chains in recent years, employing the DMRG technique[17]. The spin-$\frac{1}{2}$ and spin-1 dimerized Heisenberg chains with nearest neighbour and next nearest neighbour antiferromagnetic interactions have been recently studied by us by this new and powerful technique[18]. There have also been some recent studies on a system with one or more spin-$\frac{1}{2}$ impurities embedded randomly in a spin-1 chain[19], and

solution of spin models dynamically coupled to dispersionless phonons[20] by the DMRG method.

The spin wave analysis, in this paper, is followed by a report of our results from extensive DMRG studies on the alternating spin system. The DMRG calculations have been carried out on chains/rings with alternate spin-1 and spin-$\frac{1}{2}$ sites. Studies on the ground state and low-lying excited states are reported in detail. Furthermore, by resorting to full diagonalization of the DMRG Hamiltonian matrix in different total M_s sectors, we have also obtained the low-temperature thermodynamic properties of the alternating spin chain with periodic boundary condition. The thermodynamic properties we discuss will include low and high field magnetization, magnetic susceptibility and specific heat at low temperatures. These properties are compared with experimental studies on bimetallic chains.

The paper is organized as follows. In the next section, we present properties of the ground and low-lying excited states obtained from spin wave analysis and DMRG calculations on long alternating spin chain with and without dimerization. We also show that the short correlation length allows the ground state to be approximated by a variational *ansatz*. In the third section, we present the low-temperature thermodynamic properties of the alternating spin chain. We end the paper with a summary of all the results.

2 Properties of the Ground and Low-Lying Excited states

2.1 Spin-Wave analysis

We begin with the Hamiltonian for a chain with spins s_1 and s_2 on alternating sites.

$$H = J \sum_n [\hat{\mathbf{S}}_{1,n} \cdot \hat{\mathbf{S}}_{2,n} + \hat{\mathbf{S}}_{2,n} \cdot \hat{\mathbf{S}}_{1,n+1}] \tag{1}$$

where the total number of sites (or bonds) is $2N$, and we use periodic boundary conditions in this section with $\hat{\mathbf{S}}_{1,N+1} = \hat{\mathbf{S}}_{1,1}$.

The notation is illustrated in fig.1. We assume that $s_1 > s_2$, and will use spin wave theory[21] to compute the leading order corrections to the state shown in fig.1, where, the z-component of the spin is s_1 for the spin-s_1 sites and $-s_2$ for spin-s_2 sites. The

Holstein-Primakoff transformations take the form

$$\begin{aligned}
\hat{\mathbf{S}}^z_{1,n} &= s_1 - \hat{a}^\dagger_n \hat{a}_n \\
\hat{\mathbf{S}}^+_{1,n} &= \left(\sqrt{2s_1 - \hat{a}^\dagger_n \hat{a}_n}\right)\hat{a}_n \\
\hat{\mathbf{S}}^-_{1,n} &= \hat{a}^\dagger_n \left(\sqrt{2s_1 - \hat{a}^\dagger_n \hat{a}_n}\right)
\end{aligned} \tag{2}$$

for the spin-s_1 sites, and

$$\begin{aligned}
\hat{\mathbf{S}}^z_{2,n} &= -s_2 + \hat{b}^\dagger_n \hat{b}_n \\
\hat{\mathbf{S}}^+_{2,n} &= \hat{b}^\dagger_n \left(\sqrt{2s_2 - \hat{b}^\dagger_n \hat{b}_n}\right) \\
\hat{\mathbf{S}}^-_{2,n} &= \left(\sqrt{2s_2 - \hat{b}^\dagger_n \hat{b}_n}\right)\hat{b}_n
\end{aligned} \tag{3}$$

for the spin-s_2 sites. We then rewrite the Hamiltonian in terms of the bosonic operators $\hat{a}_n$, $\hat{b}_n$, $\hat{a}^\dagger_n$ and $\hat{b}^\dagger_n$, expand to quadratic order and Fourier transform to get

$$H = -2NJs_1s_2 + 2J\sum_k [s_1 \hat{b}^\dagger_{-k}\hat{b}_{-k} + s_2 \hat{a}^\dagger_k \hat{a}_k + \sqrt{s_1 s_2}\cos(k/2)(\hat{a}_k \hat{b}_{-k} + \hat{a}^\dagger_k \hat{b}^\dagger_{-k})] \tag{4}$$

This can be transformed using the Bogoliubov transformation, in the form

$$\begin{aligned}
\hat{c}_k &= \hat{a}_k \cosh\theta_k + \hat{b}^\dagger_{-k}\sinh\theta_k \\
\hat{d}_k &= \hat{b}_{-k}\cosh\theta_k + \hat{a}^\dagger_k \sinh\theta_k \\
\tanh(2\theta_k) &= \frac{2\sqrt{s_1 s_2}}{s_1 + s_2}\cos(k/2)
\end{aligned} \tag{5}$$

We then get

$$H = -2NJs_1s_2 + \sum_k [\omega_{1k}\hat{c}^\dagger_k \hat{c}_k + \omega_{2k}\hat{d}^\dagger_k \hat{d}_k + z_k] \tag{6}$$

where the mode energies ω_{ik} and zero-point energy z_k are given by

$$\begin{aligned}
\omega_{1k} &= J(-s_1 + s_2) + \omega_k \\
\omega_{2k} &= J(s_1 - s_2) + \omega_k \\
z_k &= -J(s_1 + s_2) + \omega_k \\
\omega_k &= J\sqrt{(s_1 - s_2)^2 + 4s_1 s_2 \sin^2(k/2)}
\end{aligned} \tag{7}$$

The ground state $|\psi_0>$ is the state annihilated by all the operators $\hat{c}_k$ and $\hat{d}_k$, because ω_{1k} and ω_{2k} are positive for all k. We see that the modes denoted by ω_{1k} are gapless at $k = 0$, where they have a ferromagnetic dispersion $\omega_{1k} \sim k^2$. The modes ω_{2k} are gapped for all k, with a minimum gap $\Delta = 2J(s_1 - s_2)$ at $k = 0$.

The ground state energy per bond is given by

$$\epsilon_0 = \frac{E_0}{2N} = -Js_1s_2 + \frac{1}{2}\int_0^\pi \frac{dk}{\pi}[-J(s_1 + s_2) + \omega_k] \tag{8}$$

For $s_1 = 1$ and $s_2 = \frac{1}{2}$, we get $\epsilon_0 = -0.718J$. The spin wave ground state $|\psi_0>$ can also be shown to be an eigenfunction of the $\hat{\mathbf{S}}^z_{tot.}$ operator with the eigenvalue $N(s_1 - s_2)$. The sublattice magnetizations are given by the expectation values of $\hat{\mathbf{S}}^z_{1,n}$ and $\hat{\mathbf{S}}^z_{2,n}$,

$$\begin{aligned} <\hat{\mathbf{S}}^z_{1,n}> &= (s_1 + \frac{1}{2}) - \frac{1}{2}\int_0^\pi \frac{dk}{\pi}\frac{(s_1 + s_2)J}{\omega_k} \\ <\hat{\mathbf{S}}^z_{2,n}> &= s_1 - s_2 - <\hat{\mathbf{S}}^z_{1,n}> \end{aligned} \tag{9}$$

For $s_1 = 1$, $s_2 = \frac{1}{2}$, we find $<\hat{\mathbf{S}}^z_{1,n}> = 0.695$ and $<\hat{\mathbf{S}}^z_{2,n}> = -0.195$. The operator

$$\hat{\mathbf{S}}^+_{tot.} = \sum_n [\hat{b}^+_n(\sqrt{2s_2}) + \hat{a}_n(\sqrt{2s_1})] = \hat{c}_0(\sqrt{2N(s_1 - s_2)}), \tag{10}$$

to linear order in the bosonic operators. Since this annihilates $|\psi_0>$, which is an eigenstate of $\hat{\mathbf{S}}^z_{tot.}$, we conclude that $|\psi_0>$ has $s_{tot.} = s^z_{tot.} = N(s_1 - s_2)$. Thus the ground state is a ferrimagnet.

We can show that the ω_{1k} modes are created by acting on $|\psi_0>$ with $\hat{\mathbf{S}}^-_k = \frac{1}{\sqrt{N}}\sum_n(\hat{\mathbf{S}}^-_{1,n} + \hat{\mathbf{S}}^-_{2,n})e^{ikn}$, where $k \neq 0$. The resultant states have $s^z_{tot.} = N(s_1 - s_2) - 1$ and are also annihilated by $\hat{\mathbf{S}}^+_{tot.}$. Therefore we conclude that these states have $s_{tot.} = s^z_{tot.} = N(s_1 - s_2) - 1$. Since ω_{1k} is a gapless branch, we further conclude that the system has gapless excitations to states with spin $N(s_1 - s_2) - 1$. Similarly, the ω_{2k} modes are created by acting on $|\psi_0>$ with $\hat{\mathbf{S}}^+_k = \frac{1}{\sqrt{N}}\sum_n(\hat{\mathbf{S}}^+_{1,n} + \hat{\mathbf{S}}^+_{2,n})e^{-ikn}$, where $k \neq 0$. By a similar argument, we can show that these states have $s_{tot.} = s^z_{tot.} = N(s_1 - s_2) + 1$. The branch ω_{2k} is separated from the ground state by a gap $\Delta = 2J(s_1 - s_2)$. Fig.2 shows these two excitation branches for $s_1 = 1$ and $s_2 = \frac{1}{2}$

Finally, we can calculate the two-spin correlation functions. There are three kinds of functions one can consider, namely $<\hat{\mathbf{S}}_{1,0}\cdot\hat{\mathbf{S}}_{1,n}>$, $<\hat{\mathbf{S}}_{2,0}\cdot\hat{\mathbf{S}}_{1,n}>$ and $<\hat{\mathbf{S}}_{2,0}\cdot\hat{\mathbf{S}}_{2,n}>$.

We will consider only the first case as an illustrative example. Since the ground state has long range order (with $s_{tot.} = s^z_{tot.} = N(s_1 - s_2)$), we consider the subtracted correlation function

$$\begin{aligned} < \hat{\mathbf{S}}_{1,0} \cdot \hat{\mathbf{S}}_{1,n} > - < \hat{\mathbf{S}}_{1,0} > \cdot < \hat{\mathbf{S}}_{1,n} > &= s_1 < \psi_0 | \hat{a}_0 \hat{a}^\dagger_n + \hat{a}^\dagger_0 \hat{a}_n | \psi_0 > \\ &= J s_1 \int_0^\pi \frac{dk}{\pi} (s_1 + s_2) \frac{\cos kn}{\omega_k} \end{aligned} \tag{11}$$

For $n \to \infty$, we can show that the leading behaviour of this is given by an exponentially decaying factor $e^{-n/\xi}$, where $\xi^{-1} = \ln(s_1/s_2)$. For $s_1 = 1$, $s_2 = \frac{1}{2}$, we get $\xi = 1.44$. This remarkably short correlation length agrees well with numerical results as will be seen later. It may also be compared to the much larger values for the pure spin-1 antiferromagnet in which $\xi \approx 6$[22, 24] and the pure spin-$\frac{1}{2}$ antiferromagnet in which $\xi = \infty$.

We can use spin wave theory to study a dimerized model described by the Hamiltonian

$$H = J \sum_n [(1+\delta) \hat{\mathbf{S}}_{1,n} \cdot \hat{\mathbf{S}}_{2,n} + (1-\delta) \hat{\mathbf{S}}_{2,n} \cdot \hat{\mathbf{S}}_{1,n+1}] \tag{12}$$

where the dimerization parameter δ lies in the range $[0, 1]$. We find that the ground state and low-energy excitations are qualitatively similar to the undimerized case, $\delta = 0$. Namely, the ground state has $s_{tot.} = N(s_1 - s_2)$. There is a gapless branch of excitations ω_{1k} with $s_{tot.} = N(s_1 - s_2) - 1$ and dispersion

$$\omega_{1k} = J(-s_1 + s_2) + J\sqrt{(s_1 - s_2)^2 + 4 s_1 s_2 (1 - \delta^2) \sin^2(k/2)}, \tag{13}$$

and a gapped branch of excitations ω_{2k} with $s_{tot.} = N(s_1 - s_2) + 1$ and whose dispersion is,

$$\omega_{2k} = \omega_{1k} + 2J(s_1 - s_2) \tag{14}$$

To this order in spin wave theory, the gap at $k = 0$ is given by $\Delta = 2J(s_1 - s_2)$ independent of δ. Numerically, however we will see below that the gap increases almost linearly with δ for the spin-1 / spin-$\frac{1}{2}$ alternating spin chain.

2.2 Variational Calculation

The short correlation length found above for the alternating spin-1 / spin-$\frac{1}{2}$ chain suggests that a *product* wave function[22] could be a good variational trial state for obtaining the

ground state properties. We work in the Fock space basis of the alternating spin chain. We are interested in a variational wave function in which the state $|\ldots \quad 1 \quad -\frac{1}{2} \quad 1 \quad -\frac{1}{2} \quad \ldots>$ of fig.1 is expected to have the largest amplitude, say, 1. We can then show that a state in which there are n spins on the spin-1 sublattice in the state $s^z = 0$ must have a real amplitude with the sign $(-1)^n$. This sign rule (which is analogous to the Marshall sign rule for the spin-$\frac{1}{2}$ chain[23]) can be proved by using the Perron-Frobenius theorem and the fact that the states with an odd number of spins in the $s^z = 0$ state are connected to states with an even number of spins in the $s^z = 0$ states by the operation of an odd number of exchange terms in the Hamiltonian (Eq.1).

Let us now introduce the following real and positive amplitudes for the six states possible for a nearest neighbour bond, namely, unit amplitude for $|1, -\frac{1}{2}>$ and $|-1, \frac{1}{2}>$, an amplitude η_1 for $|0, -\frac{1}{2}>$ and $|0, \frac{1}{2}>$ and amplitude η_2 for $|-1, -\frac{1}{2}>$ and $|1, \frac{1}{2}>$. Now, we consider a variational wave function of the form

$$|\Psi(\eta_1, \eta_2)>= \sum_a C_a |\psi_a> \tag{15}$$

where a runs over all the 6^N possible states of N spin-1 and spin-$\frac{1}{2}$ sites and $C_a = (-1)^n \prod_j \omega_{a,j}$, where the product is over all the $2N$ bonds, $\omega_{a,j}$ is the amplitude of the bond j and n denotes the number of spins in the $s^z = 0$ state in the chain state a.

The state $|\Psi(\eta_1, \eta_2)>$ is translationally invariant. However, it is not an eigenstate of the total spin $\hat{S}^2_{tot.}$ or even $\hat{S}^z_{tot.}$; further, since it includes states $\{s^z_n\}$ and $\{-s^z_n\}$ with equal amplitude, the expectation value of $\hat{\mathbf{S}}^z_{1,n}$ or $\hat{\mathbf{S}}^z_{2,n}$ is zero for any site n. In spite of these drawbacks, we will see below that the $|\Psi(\eta_1, \eta_2)>$ gives a good variational ground state energy for an appropriate choice of η_1 and η_2.

Calculations involving the state $|\Psi(\eta_1, \eta_2)>$ can be carried out using the transfer matrix method. For instance, the probabilities of the six possible bonds for $|\frac{1}{2}, n> \bigotimes < 1, n+1|$ are given by the 2×3 matrix.

$$A = \begin{pmatrix} \eta_2^2 & \eta_1^2 & 1 \\ 1 & \eta_1^2 & \eta_2^2 \end{pmatrix}$$

while the probabilities of the bonds $|1, n> \bigotimes < \frac{1}{2}, n|$ are given by the 3×2 matrix A^T. Then the norm $< \Psi(\eta_1, \eta_2)|\Psi(\eta_1, \eta_2)>= Tr(AA^T)^N$. The matrix AA^T has the eigenvalues $\lambda_+ = (1+\eta_2^2)^2 + 2\eta_1^4$ and $\lambda_- = (1-\eta_2^2)^2$. Hence, $< \Psi(\eta_1, \eta_2)|\Psi(\eta_1, \eta_2)>= \lambda_+^N + \lambda_-^N$, which is dominated by λ_+^N as $N \to \infty$.

We can now calculate the variational energy per bond $\epsilon(\eta_1, \eta_2)$ through the expression

$$\epsilon(\eta_1, \eta_2) < \Psi(\eta_1, \eta_2)|\Psi(\eta_1, \eta_2) >= \frac{1}{2N} < \Psi(\eta_1, \eta_2)|H|\Psi(\eta_1, \eta_2) > \tag{16}$$

This is equal to $< \Psi(\eta_1, \eta_2)|\hat{\mathbf{S}}_{\frac{1}{2},n} \cdot \hat{\mathbf{S}}_{1,n+1}|\Psi(\eta_1, \eta_2) >$ by translational invariance. We find that,
$\epsilon(\eta_1, \eta_2) =$

$$J\frac{[-\frac{1}{2}(1+\eta_2^2)^3(1-\eta_2^2) - 2\sqrt{2}(1+\eta_2^2)\eta_1^2\eta_2(1+\eta_2) - \eta_1^4(1-\eta_2^4) - 2\sqrt{2}\eta_1^6(1+\eta_2)]}{[(1+\eta_2^2)^2 + 2\eta_1^4]^2} \tag{17}$$

We now minimize this function of η_1 and η_2, and find that the minimum occurs at $\eta_1 = 0.842$, $\eta_2 = 0.445$, giving $\epsilon = -0.701J$. This compares favourably with the spin wave result of $-0.718J$.

We can compute the two-spin correlation function and determine how it decays asymptotically at large distances. The correlation length ξ for the asymptotic decay is given by, $\xi^{-1} = \ln(\lambda_+/\lambda_-)$. Hence ξ is 0.749 at the values of η_1, η_2 given above. This is even shorter than the value of 1.44 found from the spin wave theory.

We can improve the variational calculation by allowing the five independent (taking the amplitude for $|1, -\frac{1}{2} >= 1$) real amplitudes for the different possible states of the nearest neighbour bonds rather than only two amplitudes (η_1, η_2) as used above. However we will not do so here as such a calculation would be tedious and the two-amplitude *ansatz* has already given us a good understanding of the short correlation length and the ground state energy.

2.3 DMRG Studies

We have performed DMRG calculations on alternating spin-1 / spin-$\frac{1}{2}$ chain with open boundary condition for the Hamiltonian in Eq.(1). We compute the ground state properties by studying chains of upto 50 to 100 sites. The number of dominant density matrix eigenstates, m, that we have retained at each DMRG iteration is between 80 to 100. The DMRG procedure follows the usual steps for chains discussed in earlier papers[15, 17, 18] except that the chains do not have the symmetry between the left and right halves and the density matrices for these two halves have to be constructed at every iteration of the calculations. The DMRG results reported in this paper are all specialized to the case of

$s_1 = 1$ and $s_2 = \frac{1}{2}$. The ground state of the chain lies in the $M_s = N(s_1 - s_2)$ sector for a $2N$ sites system and we cannot use spin parity symmetry in this sector. In general, for $M_s \neq 0$, parity cannot be exploited, keeping M_s a good quantum number. The ground state is confirmed to be in $M_s = N(s_1 - s_2)$ sector for a chain of $2N$ sites from extensive checks carried out by obtaining the low-energy eigenstates in different M_s sectors of a 20-site chain. A state corresponding to the lowest energy in $M_s = N(s_1 - s_2)$ is found in all subspaces with $|M_s| \leq N(s_1 - s_2)$ and is absent in subspaces with $M_s > N(s_1 - s_2)$. This shows that the spin in the ground state, $s_G = N(s_1 - s_2)$. In fig.3, we show the extrapolation of the energy per site as function of inverse system size. The energy/site, ϵ_0, extrapolates to $-0.72704J$ for the ground state of the system. The spin wave analysis value of $-0.718J$ for ϵ_0 is higher than the DMRG value. It is worth mentioning at this point that the DMRG ground state energy/site agrees to better than 10^{-7} with the exact solution in the case of uniform spin-$\frac{1}{2}$ Heisenberg antiferromagnet[15, 16]. It is interesting to note that, in the alternating spin case, the energy/site lies in between the values for the pure spin-$\frac{1}{2}$ uniform chain ($-0.443147J$) [25] and the pure spin-1 uniform chain ($-1.401484J$) [16].

In fig.4, we show the spin-density at all the sites of a chain of 100 sites. The spin-density is uniform on each of the sublattices in the chain. The expectation value of $\hat{\mathbf{S}}_z$ at the spin-1 site reduces from the classical value of 1 to 0.79248, while, at the spin-$\frac{1}{2}$ site, it is -0.29248. This can be compared with spin wave values of 0.695 and -0.195 for spin-1 and spin-$\frac{1}{2}$ sites respectively. We note that the spin wave analysis overestimates the quantum fluctuations. It is very interesting to note that, the spin-density distribution in alternating spin chain behaves more like in a ferromagnetic chain than like in an antiferromagnet, with the net spin of each unit cell perfectly aligned (but with small fluctuations on the individual sublattices). In a ferromagnetic ground state the spin-density at each site has the classical value appropriate to the site spin, whereas for an antiferromagnet, this averages out to zero at each site as the ground state is nonmagnetic. From this viewpoint, the ferrimagnet is similar to a ferromagnet and is quite unlike an antiferromagnet. The spin wave analysis also yields the same physical picture.

Owing to the alternation of the spin-1 and spin-$\frac{1}{2}$ sites along the chain, one has to distinguish between $< \hat{\mathbf{S}}^z_{1,0}\hat{\mathbf{S}}^z_{1,n} >$, $< \hat{\mathbf{S}}^z_{2,0}\hat{\mathbf{S}}^z_{2,n} >$ and $< \hat{\mathbf{S}}^z_{1,0}\hat{\mathbf{S}}^z_{2,n} >$ pair correlations. We calculate all the three correlation functions with the mean values subtracted out as in Eq.(11), since the mean values are nonzero in this system unlike in a pure antiferromag-

netic spin chain. In the DMRG procedure, we have computed these correlation functions from the sites inserted at the last iteration, to minimize numerical errors. In fig.5, we plot the two-spin correlation functions in the ground state as a function of the distance between the spins for an open chain of 100 sites. All three correlation functions decay rapidly with distance. From the figure it is clear that, except for $< \hat{\mathbf{S}}^z_{1,0}\hat{\mathbf{S}}^z_{2,n} >$ correlation, other correlations are almost zero even for the shortest possible distances. The $< \hat{\mathbf{S}}^z_{1,0}\hat{\mathbf{S}}^z_{2,n} >$ correlation has a finite value (-0.094) only for the nearest neighbours. This rapid decay of the correlation functions do not easily allow one to find the exact correlation length ξ for a lattice model though it is clear that ξ is very small (*i.e.* less than one unit). As mentioned earlier, spin wave theory gives $\xi = 1.44$ while the variational calculation gives $\xi = 0.75$. This type of decay could be compared with the behaviour of the correlation function at the Majumdar-Ghosh point[26] for the pure spin-$\frac{1}{2}$ Heisenberg chain with nearest and next nearest neighbour antiferromagnetic exchange interactions ($J_1 - J_2$ model at $J_2 = 0.5J_1$). The AKLT model at the exactly solvable point[22] in the case of a pure spin-1 chain described by a bilinear-biquadratic Hamiltonian also has a very short correlation length ($\xi = 0.91$). Both the cases compared above, however, have one property in common, *i.e.*, the exactly solvable point in both models lie in the gapped phase.

The lowest spin excitation of the alternating spin chain is to a state with $s = s_G - 1$. To get this state, we target the 2nd state in $M_s = s_G - 1$ sector of the chain. To confirm that this state is the $s = s_G - 1$ state, we have computed the 2nd state in $M_s = 0$ sector and find that it also has the same energy. However, the corresponding state is absent in M_s sectors with $M_s > |s_G - 1|$. Besides, from exact diagonalization of all the states of an alternating spin chain with 8 sites, we find that the ordering of the state is such that the lowest excitation is to the state with spin $s = s_G - 1$. We have obtained the excitation gap of the alternating spin chain in the limit of infinite chain length by extrapolating from the plot of spin gap *vs* the inverse of the chain length (fig.6). We find that this excitation is gapless in the infinite chain limit. This is very unusual in that the correlation function in the ground state decays exponentially (in fact $\xi < 1$) but the system has a gapless excitation, in agreement with the spin wave analysis.

Motivated by the spin-wave calculation, we also have computed the energy of the $s = s_G + 1$ state by targetting the lowest state in $M_s = s_G + 1$ sector. In fig.7, we plot the excitation gap to the $s = s_G + 1$ state from the ground state as a function of the

inverse of the chain length. The gap saturates to a finite value of $(1.2795 \pm 0.0001)J$. The S_z expectation values computed in this state are found to be uniform on each of the sublattices (independent of the site) and leading us to believe that this excitation is not a magnon like excitation (quantized in a box) as has been observed for a spin-1 chain in the Haldane phase[16].

It would be interesting to know the total spin of the states as a function of their energies. For a smaller alternating spin system, it is possible to characterize all the states by their energy and total spin value, by resorting to an exact diagonalization scheme. The total spin value of a state is naturally fixed if we exploit the total spin conservation property of the Hamiltonian while constructing the Hamiltonian matrix. This can, for example, achieved by using a valence bond basis[27] for setting up the Hamiltonian matrix. Alternately, we can also compute the expectation value of the total spin operator in each eigenstate in the $M_s = 0$ sector to provide a spin label for each of the states. We have followed the latter procedure. In fig.8, we present the energy levels as a function of the total spin of the states for an eight site ferrimagnetic ring and twelve site spin-$\frac{1}{2}$ ferromagnetic and antiferromagnetic rings. We find that the spin of the state appears to vary irregularly with energy unlike in the case of pure spin-$\frac{1}{2}$ ferro and antiferromagnets (fig.8). Careful comparison of the three plots in the figure shows that the low-lying excitations of the ferrimagnet to spin states $s_{tot} < s_G$ is ferromagnetic like and to states with $s_{tot} > s_G$ is antiferromagnetic like, for finite systems.

We have also studied the dimerized alternating spin chain defined by the Hamiltonian (Eq.12) with δ, the dimerization parameter, in the range $0 < \delta \leq 1$. Earlier works on spin chains[28] have revealed that with the alternation δ in the exchange parameter, the half integer spin chain will have an unconditional spin-Peierls transition whereas for integer spin chain the transition is conditional. This conclusion has been drawn from the fact that, with the inclusion of δ, the magnetic energy gain ΔE can be defined as,

$$\Delta E(2N, \delta) = \frac{1}{2N}[E(2N, \delta) - E(2N, 0)] \tag{18}$$

where $E(2N, \delta)$ is the ground state energy of the $2N$ sites system with alternation δ in the exchange integral and $E(2N, 0)$ is the ground state energy of the uniform chain of $2N$ sites. For the pure spin chain, if we assume that ΔE varies as δ^ν for small δ, we find that $\nu = 4/3$ for the spin-$\frac{1}{2}$ chain and $\nu = 2$ for the spin-1 chain[28]. Thus, for the spin-$\frac{1}{2}$ chain, the stabilization energy always overcomes the elastic energy, whereas for the spin-1

case, it depends on the stiffness of the lattice.

We have employed a DMRG calculations to obtain $\Delta E(2N, \delta)$, for small values of δ for the alternating spin chain. To determine the exact functional form of the magnetic energy gain, we varied the chain length from 50 sites to 100 sites and also m(the number of states retained in each DMRG iteration) from 80 to 100 to check the convergence of ΔE with the chain length. The dependence of $\Delta E(2N, \delta)$ on $1/2N$ is linear for the δ values we have studied. Fig.9 gives a sample variation of $\Delta E(2N, \delta)$ upon $1/2N$. This allows us to extrapolate $\Delta E(2N, \delta)$ to the infinite chain limit reliably. In fig.10, we show the plot of $\Delta E(2N, \delta)$ vs. δ for finite $2N$ values and also the extrapolated infinite chain. We see that there is a gain in magnetic energy upon dimerization even in the infinite chain limit. To obtain the exponent ν, we plot $\ln \Delta E(2N, \delta)$ vs. $\ln \delta$ for the infinite chain (fig.11). From this figure, we find that in the alternating spin case, for the infinite chain $\Delta E \approx \delta^{2.00 \pm 0.01}$. Thus, the spin-Peierls transition appears to be close to being conditional in this system. The magnetic energy gain/site for finite chains is larger than that of the infinite chain for any δ values (fig.10). It is possible that the distortion in finite chain is unconditional while that of the infinite chain is conditional.

We have also studied the spin excitations in the dimerized alternating spin-1 / spin-$\frac{1}{2}$ chain. We calculate the lowest spin excitation to the $s = s_G - 1$ state from the ground state. We find that the $s = s_G - 1$ state is gapless from the ground state for all δ values. This result agrees with the spin wave analysis. The system remains gapless even while dimerized unlike the pure antiferromagnetic dimerized spin chains. There is a smooth increase of the spin excitation gap to $s = s_G + 1$ state from ground state with increasing δ. We have plotted this gap with δ in fig.12. The gap shows almost a linear behaviour as a function of δ, with an exponent of 1.07 ± 0.01. The spin wave analysis however shows that this excitation gap is independent of δ.

3 Low-Temperature Properties

In this section, we present results of our DMRG calculations of the thermodynamic properties of the alternating spin system. The size of the system varies from 8 to 20 sites. We impose periodic boundary conditions to minimize finite size effects. We set-up the Hamiltonian matrices in the DMRG basis for all allowed M_s sectors for a ring of 2N sites.

We can diagonalize these matrices completely to obtain all the eigenvalues in each of the M_s sectors. As the number of DMRG basis states increases rapidly with increasing m, we retain a smaller number of dominant density matrix eigenvectors in the DMRG procedure, *i.e.*, $50 \leq m \leq 65$, depending on the M_s sector as well as the size of the system. We have checked to find the dependence of properties (with m in the range $50 \leq m \leq 65$) for the system sizes we have studied ($8 \leq 2N \leq 20$) and have confirmed that the properties do not vary significantly for the temperatures at which they are computed. The above extension of the DMRG procedure is found to be accurate by comparing with exact diagonalization results for small systems. It may appear surprising that the DMRG technique which essentially targets a single state, usually the lowest energy state in a chosen sector, should provide accurate thermodynamic properties since these properties are governed by energy level spacings and not the absolute energy of the ground state. However, there are two reasons why the DMRG procedure yields reasonable thermodynamic properties. Firstly, the projection of the low-lying excited state eigenfunctions on the DMRG space in which the ground state is obtained is substantial and hence these excited states are well described in the chosen DMRG space. The second reason is that the low-lying excitations of the full system are often lowest energy states in different sectors in DMRG procedure and thus their energies are quite accurate even on an absolute scale.

The canonical partition function Z for the 2N site ring can be written as

$$Z = \sum_j e^{-\beta(E_j - B(M_s)_j)} \tag{19}$$

where, the sum is over all the DMRG energy levels of the 2N site system in all the M_s sectors. E_j and $(M_s)_j$ are energy and z-component of the total spin of the state j, B is the strength of the magnetic field in units of $J/g\mu_B$ (g is the gyromagnetic ratio and μ_B is the Bohr magneton) along **z** direction and $\beta = J/k_BT$ with k_B and T being the Boltzmann constant and temperature respectively. The field induced magnetization, $< M >$, can be defined as

$$< M >= \frac{\sum_j (M_s)_j e^{-\beta(E_j - B(M_s)_j)}}{Z} \tag{20}$$

the magnetic susceptibility, χ, by relating it to the fluctuation in magnetization,

$$\chi = \beta[< M^2 > - < M >^2] \tag{21}$$

and similarly the specific heat, C, by relating it to the fluctuation in energy, can be written

as,

$$C = \frac{\beta}{T}[< E^2 > - < E >^2] \tag{22}$$

The dimensionalities of the DMRG Hamiltonian matrices that we completely diagonalize vary from 2500 to 3000, depending upon the DMRG parameter m and the M_s value of the targetted sector, for rings of sizes greater than 12. These matrices are not very sparse, owing to the cyclic boundary condition imposed on the system. The DMRG properties compare very well with exact results for small system sizes amenable to exact diagonalization studies. In the discussion to follow, we present results on the 20-site ring although all calculations have been carried out for system sizes from 8 to 20 sites. This is because the qualitative behaviour of the properties we have studied are similar for all the ring sizes in this range.

We present the dependence of magnetization on temperature for different magnetic field strengths in fig.13. At low magnetic fields, the magnetization shows a sharp decrease at low-temperatures and shows paramagnetic behaviour at high temperatures. As the field strength is increased, the magnetization shows a slower decrease with temperature and for field strength comparable to the exchange constant, the magnetization shows a broad maximum. This behaviour could be understood from the type of spin excitations present in the system. The lowest energy excitation at low magnetic fields is to a state with spin s less than s_G. Therefore, the magnetization initially decreases at low temperatures. As the field strength is increased, the gap to spin states with $s > s_G$ decreases as the Zeeman coupling to these states is stronger than to the states with $s \leq s_G$. The behaviour of the system at even stronger fields turns out to be remarkable. The magnetization in the ground state ($T = 0$) shows an abrupt increase signalling that the ground state at this field strength has $M_s > s_G$. The temperature dependence of the magnetization shows a broad maximum indicating the presence of states with even higher spin values lying above the ground state in the presence of this strong field. Only at very intense fields do we find the magnetization decreasing with increasing temperature. This happens because at such large field strengths, the ground state is the highest spin state possible for the system.

The dependence of magnetization on the magnetic field is shown at different temperatures in fig.14. At low temperature the magnetization shows a plateau. The width of the plateau decreases as the temperature is raised and eventually the plateau disappears. The existence of the plateau shows that the higher spin states are not accessible at the

chosen temperature. At higher fields, the larger Zeeman splittings of higher spin states become accessible leading to an increase in the magnetization. All these curves intersect at $B = J/g\mu_B$ and $B = 2.5J/g\mu_B$ and these fields are close to the field strengths at which the ground state switches from one M_s value to another higher value.

The dependence of $\chi T/2N$ on temperature for different field strengths are shown in fig.15. For zero field, the zero temperature value of χT is infinite in the thermodynamic limit and for finite rings is finite and equal to the fluctuation in magnetization. For the ferrimagnetic ground state $\chi T/2N$, as $T \to 0$, is given by $s_G(s_G + 1)/6N$. As the temperature increases, this product decreases and shows a minimum around $k_B T = 0.5J$ before increasing again. The minimum manifests due to the states with $M_s < s_G$ getting populated at low-temperatures. These states in the infinite chain limit turn out to be the gapless excitations in the spectrum. The subsequent increase in the χT product is due to the higher energy and higher spin states accessed with further increase in temperature. Experimentally, it has been found in the bimetallic chain compounds that the temperature at which the minimum occurs in the χT product depends upon the magnitude of the spins s_1 and s_2[13]. The $Ni^{II} - Cu^{II}$ bimetallic chain shows a minimum in $\chi T/2N$ at a temperature corresponding to 55 cm^{-1} $(80K)$ and independent estimate of the exchange constant in this system is 100 cm^{-1}[29]. This is in very good agreement with the minimum theoritically found at temperature $(0.5 \pm 0.1)J$. The minimum in $\chi T/2N$ vanishes at $B = 0.1J/g\mu_B$ corresponding to $\sim 10T$ and it would be interesting to study the magnetic susceptibility of other systems experimentally under such high fields. The low-temperature zero-field behaviour of our system can be compared with the one-dimensional ferromagnet. In the latter, the spin wave analysis shows that the χT product increases as $\frac{1}{T}$ at low temperatures[30].

In finite but weak field, the behaviour of χT is different. The magnetic field opens up a gap and χT goes exponentially to zero for temperatures less than the gap in the applied field. Even in this case a minimum is found at the same temperature as in the zero-field case, for the same reason discussed in the zero field case.

In stronger magnetic fields, the behaviour of χT from zero temperature upto $k_B T = 0.5J$ is qualitatively different. The minimum in this case vanishes. In these field strengths, the states with higher M_s values are accessed even below $k_B T = 0.5J$. The dependence of χT above $k_B T = 0.5J$ is the same in all cases. In even stronger magnetic fields, the

initial sharp increase is suppressed. At very low temperature, the χT product is nearly zero and increases linearly with T over the temperature range we have studied. This can be attributed to a switch in the ground state at this field strength. The very high temperature behaviour of χT should be independent of field strength and should saturate to the Curie law value corresponding to the mean of spin-1 and spin-$\frac{1}{2}$ values which is 11/24.

The temperature dependence of specific heat also shows marked dependence on the magnetic field at strong-fields. This dependence is shown in fig.16 for various field strengths. In zero and weak magnetic fields, the specific heat shows a broad maximum around $k_BT = 0.6J$. At strong-magnetic field ($B = J$), there is a dramamtic increase in the peak height at about the same temperature, although the qualitative dependence is still the same as at low-magnetic fields. This indicates that the higher energy high-spin states are brought to within k_BT of the ground state at this magnetic field strength.

Studies on dimerized alternating spin chains show qualitatively similar trends as the uniform chains. This is not surprising as the low-energy spectrum of the system does not change qualitatively upon dimerization.

4 Summary

We have studied the alternating spin-1 / spin-$\frac{1}{2}$ model in detail. The ground and low-lying excited states have been analyzed by using a spin wave theory as well as DMRG calculations. Both the methods predict a ground state with spin $s_G = N(s_1 - s_2)$ for a $2N$ site system. They also predict a gapless excitation to a state with $s = s_G - 1$ in the infinite chain limit. The lowest gapped excitations are to states with spin $s = s_G + 1$. The very short correlation length in the ground state of the system motivated its description by a variational trial function of the product type. Interestingly, the spectrum is qualitatively unchanged upon dimerization. The dimerization is itself conditional in the infinite chain limit.

The DMRG technique is also employed to obtain the low-temperature thermodynamic properties. The magnetic susceptibility shows very interesting magnetic field dependence. The χT vs. T plot shows a minimum at low-magnetic fields. This minimum vanishes at high-magnetic fields. The specific heat shows a maximum as a function of temperature

at all fields. The height of the maximum shows a dramatic increase at high-magnetic field. Experimental systems describable by this model exist and have been studied quite extensively. It is hoped that our studies will motivate experimental studies of these systems in high magnetic fields.

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Figure Captions

Fig.1

Schematic picture of the arrangement of spins s_1 and s_2 on a chain with interactions discussed by the Hamiltonian in Eq.(1).

Fig.2

The two branches of the spin wave dispersion curves, ω_{1k} and ω_{2k} (Eq.(7)), in units of J, for the alternating spin system with $s_1 = 1$ and $s_2 = \frac{1}{2}$.

Fig.3

Extrapolation of the ground state energy/site (ϵ_0), in units of J, as a function of inverse system size, for the uniform alternating spin-1 /spin-$\frac{1}{2}$ chain.

Fig.4

Expectation value of the z-component of the site spin vs. site index, n. The upper and lower points are for the spin-1 ($i = 1$) and the spin-$\frac{1}{2}$ ($i = 2$) sites respectively.

Fig.5

Subtracted two-spin correlation functions (defined in the text) as a function of distance between the two spins. (a) spin-1 spin-1 correlations, (b) spin-$\frac{1}{2}$ spin-$\frac{1}{2}$ correlations and (c) spin-1 spin-$\frac{1}{2}$ correlations.

Fig.6

Energy gap (units of J) from the ground state to the lowest state with spin $s = s_G - 1$ as a function of inverse system size. s_G is the total spin of the ground state.

Fig.7

Excitation gap (units of J) from the ground state (spin $s = s_G$) to the state with spin $s = s_G + 1$ as a function of the inverse system size. The convergence to the infinite system is much faster for this gapped excitation, as compared to the gapless excitation in fig.6.

Fig.8

Plot of energy (units of J) vs. the total spin quantum number of the complete spectrum of (a) an 8 site ring with an alternating spin-1 / spin-$\frac{1}{2}$ arrangement (b) a ring of 12 site spin-$\frac{1}{2}$ antiferromagnet and (c) a ring of 12 site spin-$\frac{1}{2}$ ferromagnet. The states with very high energies are not shown, for (b) and (c).

Fig.9

Gain in magnetic energy (units of J) associated with dimerization vs. the inverse system

size for three different values of dimerization δ. (a) $\delta = 0.025$, (b) $\delta = 0.05$ and (c) $\delta = 0.075$.

Fig.10

Magnetic energy gain $\Delta E(2N, \delta)$ (units of J) as a function of dimerization parameter δ for different system sizes. In the figure $2N = 50$ (squares), $2N = 100$ (circles) and extrapolated values with $N \to \infty$ (triangles) are shown.

Fig.11

Log-log plot of extrapolated magnetic energy gain (units of J) for infinite system size and dimerization parameter δ. The slope is calculated to be 2.00 ± 0.01.

Fig.12

Excitation gap (units of J) to the state with spin $s = s_G + 1$ from the ground state ($s = s_G$) as a function of δ for the dimerized alternating spin-1 / spin-$\frac{1}{2}$ chain.

Fig.13

Plot of magnetization/site as a function of temperature T for five different values of magnetic fields, B. Squares are for $B = 0.1J/g\mu_B$, circles for $B = 0.5J/g\mu_B$, triangles for $B = J/g\mu_B$, diamonds for $B = 2J/g\mu_B$ and inverse triangles for $B = 3J/g\mu_B$.

Fig.14

Magnetization/site vs. the magnetic field strength B, in units of $J/g\mu_B$, for four different temperatures T. $T = 0.3J/k_B$ results are given by squares, $T = 0.5J/k_B$ by circles, $T = 0.7J/k_B$ by triangles and $T = J/k_B$ by diamonds.

Fig.15

χT (defined in the text) per site as a function of temperature T for various magnetic field strengths, B. Zero field results are shown by squares, $B = 0.01J/g\mu_B$ by circles, $B = 0.1J/g\mu_B$ by triangles and $B = J/g\mu_B$ by diamonds.

Fig.16

Specific heat/site as a function of temperature T for four different values of magnetic fields, B. Zero field data are shown by squares, $B = 0.01J/g\mu_B$ by circles, $B = 0.1J/g\mu_B$ by triangles and $B = J/g\mu_B$ by diamonds.