Quantum phase transitions in alternating spin-($\frac{1}{2}, \frac{5}{2}$) Heisenberg chains

Antônio S F Tenório, R R Montenegro-Filho and M D Coutinho-Filho

Laboratório de Física Teórica e Computacional, Departamento de Física, Universidade Federal de Pernambuco, CEP 50670-901, Recife, Pernambuco, Brazil

Received 11 August 2011, in final form 5 November 2011
Published 1 December 2011
Online at stacks.iop.org/JPhysCM/23/506003

Abstract

The ground state spin-wave excitations and thermodynamic properties of two types of ferrimagnetic chains are investigated: the alternating spin-1/2 spin-5/2 chain and a similar chain with a spin-1/2 pendant attached to the spin-5/2 site. Results for magnetic susceptibility, magnetization and specific heat are obtained through the finite-temperature Lanczos method with the aim of describing the available experimental data, as well as comparison with theoretical results from the semiclassical approximation and the low-temperature susceptibility expansion derived from Takahashi’s modified spin-wave theory. In particular, we study in detail the temperature versus magnetic field phase diagram of the spin-1/2 spin-5/2 chain, in which several low-temperature quantum phases are identified: the Luttinger liquid phase, the ferrimagnetic plateau and the fully polarized phase, and the respective quantum critical points and crossover lines.

(Some figures may appear in colour only in the online journal)

1. Introduction

Quasi-one-dimensional magnetic materials form a class of compounds with magnetic properties that above a characteristic temperature can be described through one-dimensional models [1]. These include systems with a rotationally invariant singlet ground state (GS), modeled, for example, through spin-1 gapped and gapless critical spin-1/2 chains [1], as well as more complex structures, such as ladders [1] and spin tubes [2]. Typically, gapless one-dimensional systems exhibit power-law decay of the correlation functions and can be understood through the Bethe ansatz [3] or the Luttinger liquid theory [1]. In addition, in gapped one-dimensional (1D) systems the application of an external magnetic field $B$ can suppress the gap and induce a quantum phase transition [4] to a Luttinger liquid phase. In particular, an extensive study of the $B$–temperature ($T$) phase diagram of a spin-1/2 gapped ladder system was recently carried out [5, 6].

In contrast to the above mentioned systems, quasi-1D ferrimagnetic compounds display GS spontaneous magnetization and have ferromagnetic and antiferromagnetic (AF) spin-wave excitations. Usually the AF spin-wave mode is gapped and the magnetization curve exhibits a plateau, which can be explained by topological arguments [7]. Ferrimagnetism can arise from the topology of the unit cell [8], as in the phosphates with chemical formula $A_3Cu_3(PO_4)_4$, where $A = Ca, Sr$ or $Pb$. These materials have three $Cu^{2+}$ spin-1/2 ions [9] and can be modeled by a line of spin-1/2 trimer clusters [10, 11] with AF exchange couplings. Another class of ferrimagnets comprises the mixed-spin compounds of type $(A–X–B–X–)_n$, where $A$ and $B$ are two different magnetic components (single ions or more complex molecules) and $X$ is a bridging ligand. In particular, we are interested in compounds that can be modeled by spin-1/2 spin-5/2 chains ($sS$ chains); these include, for example, systems built from $Mn^{2+}$ and $Cu^{2+}$ ions linked through a dithioxalato ligand [12, 13]. Further, in the composition of some ferrimagnets, the magnetic elements can be organic radicals like the nitronyl nitroxide free radicals (NITR), where $R$ stands for an alkyl (methyl, ethyl) or aromatic group (phenyl). A family in this category consists of the $Mn$–NITR compounds [14], for which there is an AF exchange coupling between the spin-1/2 radicals and the spin-5/2 $Mn^{2+}$ ions.

In this work we present a numerical study of the thermodynamic properties of the ferrimagnetic chains illustrated in figure 1: a spin-1/2 spin-5/2 alternating chain ($sS$ chain) and...
In section 3, we estimate the model parameters suitable to describe the experimental data (susceptibility and magnetization) of the related compounds. In section 4, the one-magnon bands and the specific heats of the two systems are presented and the main features are discussed. In section 5, we exhibit the $T-R$ phase diagram of the $sS$ chain and discuss in detail its quantum critical points and crossover lines, the Luttinger liquid phase and the plateau regions. In section 6, we analyze the low-temperature behavior of the zero-field magnetic susceptibility and, finally, in section 7 we present a discussion of our relevant findings.

2. Models and methods

The $sS$ chain with a uniform exchange interaction $J (>0)$ and $N_c \{s, S\}$ cells is described by the following Hamiltonian:

$$H_{sS} = J \sum_{i} (S_i + S_{i+1}) \cdot s_i - g \mu_B B S^z,$$

where $s = 1/2$ and $S = 5/2$, the $g$-factor is assumed uniform, $\mu_B$ is the Bohr magneton, $B$ is an applied magnetic field in the $z$ direction and $S^z$ is the operator for the $z$ component of the total spin. This chain is bipartite, with $N_c$ sites with spin-5/2 in one sublattice and $N_c$ sites with spin-1/2 in the other; therefore, the Lieb and Mattis theorem [20] assures that the GS total spin, $S_{GS}$, is given by $N_c|S-s| = 2N_c$, i.e., spin 2 per unit cell. The GS magnetic ordering of this chain is sketched in figure 1(a).

The $ssS$ chain with $N_c \{s, S, s'\}$ cells is described by the following Hamiltonian:

$$H_{ssS} = J \sum_{i=1}^{N_c} s_i \cdot (S_i + S_{i+1}) + J \sum_{i=1}^{N_c} s_i \cdot s'_i - g \mu_B B S^z,$$

where $s = s' = 1/2$ and $S = 5/2$, while $J > 0$ and $J' > 0$. The Lieb and Mattis theorem assures that $S_{GS} = N_c|S-2s| = 3N_c/2$, i.e., spin $\frac{5}{2}$ per unit cell. The GS magnetic order of this chain is sketched in figure 1(b).

The FTLM [18] is based on the Lanczos diagonalization technique and random sampling. The fundamental relations used in the FTLM for the calculation of a static quantity associated to an operator $A$ are

$$\langle A \rangle \approx \frac{N_{st}}{Z R} \sum_{r=1}^{R} \sum_{j=0}^{M} e^{-\beta E_f(r)} \langle r|\psi_f(r)\rangle \langle \psi_f(r)|A|r \rangle,$$

$$Z \approx \frac{N_{st}}{R} \sum_{r=1}^{R} \sum_{j=0}^{M} e^{-\beta E_f(r)} |\langle r|\psi_f(r)\rangle|^2,$$

where the sampling is carried over $R$ random states $|r\rangle$, taken as initial states for an $M$-step Lanczos procedure which results in $M$ approximate eigenvalues $E_f(r)$ with respective eigenvectors $|\psi_f(r)\rangle$ in the $N_{st}$-dimensional Hilbert space. The method allow us to calculate the temperature dependence of the magnetization per unit cell $m_c$, magnetic susceptibility per unit cell $\chi$, and specific heat $C$ through $m_c = g\mu_B \frac{(S^z)^2}{N_c}$, $\chi = g^2 \mu_B^2 \frac{(S^z)^2}{N_c k_B T}$, and $C = \frac{(M^2) - (M)^2}{k_B T}$, where $k_B$ is the
Boltzmann constant. The total number of sites is \( N = 2N_c \) for the sS chain and \( N = 3N_c \) for the ssS chain. In the computation we have used periodic boundary conditions, \( M = 50 \) for both chains and \( R = 40,000 \ (50,000) \) for the sS (ssS) chain. A full diagonalization study of the specific heat and susceptibility for the sS chain with \( N_c = 3 \) can be found in [21].

3. Magnetic susceptibility and model parameters

Through a semiclassical approach, in which the S spins are treated as classical variables, Seiden [19] derived a closed formula for the magnetic susceptibility \( \chi \). In particular, the quantity \( T\chi(T) \) has a minimum at a temperature \( T_{\text{min}} \) which is generally situated in a region where \( \beta JS < 1 \), a feature which has been known to be typical of 1D ferrimagnets. Similarly, a closed expression for the susceptibility of the ssS chain can also be established [15].

In figure 2(a) we present data for the magnetic susceptibility of the compound CuMnDTO (from [13, 19]) together with FTLM, with \( J/k_B = 44.8 \) K and \( g = 1.90 \), and semiclassical approximation [19] results, with \( J/k_B = 59.7 \) K and \( g = 1.9 \), for the sS chain. For the FTLM results, the estimation of \( J \) is made by using the value of the minimum of the experimental curve: \( T_{\text{min}} = 130 \) K. We see that both the FTLM and the semiclassical approach agree with the experimental data in the mid- and high-temperature regimes. As the temperature is lowered below \( T_{\text{min}} \), \( \chi T \) increases and presents a maximum at \( T_{\text{max}} = 7.5 \) K, which marks the onset of the tridimensional ordering. For a strictly one-dimensional system it is expected, from the Mermin–Wagner theorem [22], that long-range order (LRO) may occur only at \( T = 0 \). We remark that for quantum ferromagnetic [23] chains the correlation length diverges as \( 1/T \) and the susceptibility as \( 1/T^2 \); further, the low-lying magnetic excitations of ferrimagnetic chains present a ferromagnetic character (see below) and the same critical behavior as in [24] is shown to hold, which explains the increase in the curve of \( \chi T \) just below \( T = T_{\text{min}} \).

In figure 2(b) we present FTLM and semiclassical results [15] for the ssS chain, and experimental data [15] for the MnNN compound. A profile similar to that of figure 2(a) is observed with \( T_{\text{min}} = 255 \) K. Taking \( g = 2.0 \), our estimates for the model parameters, \( J \) and \( J' \), are \( J = 1.7I \), with \( J/k_B = 150 \) K and \( J'/k_B = 255 \) K. We also note that the FTLM curve for this chain departs from the experimental one at a higher temperature than the curve for the sS chain. This behavior is in fact a finite-size effect since the number of unit cells used in the FTLM calculation for the ssS chain (6 unit cells, with 18 sites) is effectively less than the number used for the sS chain (8 unit cells, with 16 sites).

In figure 3 we compare our results at \( T = 4.2 \) K with experimental data from [13]. The temperature is lower than the one at which the maximum of the \( \chi T \) curve is observed, \( T_{\text{max}} \approx 7.5 \) K. However, due to the low value of the coupling between chains, \( J_{\text{inter-chain}}/k_B \approx 0.1 \) K, we expect that at \( T = 4.2 \) K and for fields higher than \( \sim 0.1 \) T, the ferrimagnetic correlations along the chain are the relevant ones to determine the behavior of the magnetization as a function of \( B \). Since the correlation length along the chains diverges [24, 25] as \( 1/T \), a finite number of unit cells are correlated at \( 4.2 \) K. Thus, we can treat the system as composed of independent linear clusters, each cluster carrying a total spin \( S \), and a superparamagnetic behavior is expected for the magnetization curve. Within this context, we try to estimate the number of correlated cells in the chain from the experimental data shown in figure 3(b) by comparing with the FTLM data and the molar magnetization of a Brillouin paramagnet (BP) with total spin \( S \), given by \( M_m(B, T) = N_A g_\mu_B(S - s)B_\gamma(x) \), where \( N_A \) is the Avogadro constant and \( B_\gamma(x) \) is the Brillouin function. As shown in figure 3(b), the experimental data are well described by the BP curve with \( S = 15 \), indicating that approximately 8 unit cells (size used in the FTLM calculation) are ferrimagnetically correlated at \( 4.2 \) K. This enforces the one-dimensional description of the experimental magnetization for these temperature and field values, as well as the superparamagnetic behavior. We remark that the authors of [13] estimate that approximately 10 cells of the compound CuMnDTO are ferrimagnetically correlated at \( T = 7.9 \) K (just above \( T_{\text{max}} \)) for \( B = 0 \).
one-magnon bands, in units of magnetic field, of the non-interacting spin-wave (SW) results [26]:

\[ \omega^-(q) = E_{\text{min}}(S_{\text{GS}} - 1, q) - E_{\text{GS}}, \]

\[ \omega^+(q) = E_{\text{min}}(S_{\text{GS}} + 1, q) - E_{\text{GS}}, \]

where \( E_{\text{min}}(S_i, q) \) indicates the lowest energy in the total spin sector \( S_i \) and lattice wavenumber \( q = 2\pi l/N_c \), with \( l = 0, 1, 2, \ldots, N_c - 1 \).

In figures 4(a) and (b) we display the lower energy one-magnon bands, in units of magnetic field, of the \( ss \) and \( ssS \) chains. For the \( ssS \) chain, we also plot in figure 4(a) non-interacting spin-wave (SW) results [26]:

\[ \omega_{\text{SW}}^-(q) = -J(S - s) + \omega_q + g\mu_B B, \]

\[ \omega_{\text{SW}}^+(q) = J(S - s) + \omega_q - g\mu_B B, \]

where \( \omega_q = J\sqrt{(S - s)^2 + 4s\sin^2(q/2)}, s = 1/2 \) and \( S = 5/2 \).

We notice that in zero field the ferromagnetic excitation is gapless (which is expected from the spontaneously broken symmetry of the GS) and displays a quadratic dispersion relation in the long wavelength limit, as predicted by conformal invariance [27], while a gap \( \Delta \) exists for the AF excitation. The ferromagnetic branch obtained through non-interacting SW theory for the \( ss \) chain is in good agreement with the ED data, while for the AF branch the value of the zero-field gap \( \Delta = 2J(S - s) = 4J \) departs from the ED value, as is often the case in other ferromagnetic systems [28], due to quantum fluctuation effects. In fact, we estimate that in the thermodynamic limit \( \Delta = 4.9046J (3.88J) \) for the \( ss \) (\( ssS \)) chain.

For the \( ssS \) system, we find no published experimental data for the magnetization. However, considering the FTLM results, figure 3(b), we estimate that at \( T = 15 \) \( K \) the number of ferrimagnetically correlated unit cells is \( \approx 6 \) (size used in the FTLM calculation), due to the good agreement between the FTLM results and the BP curve with \( \bar{S} = 8.75 \).

4. One-magnon bands and specific heat

Due to the ferrimagnetic order of the GS, there are two kinds of elementary excitation in the systems: ferromagnetic magnons, which lower the total spin by one unit and AF magnons, which increase the total spin by one unit. The dispersion relations of the lower energy magnons are calculated, respectively, through

\[ \omega^-(q) = E_{\text{min}}(S_{\text{GS}} - 1, q) - E_{\text{GS}}, \]

\[ \omega^+(q) = E_{\text{min}}(S_{\text{GS}} + 1, q) - E_{\text{GS}}, \]

where \( E_{\text{min}}(S_i, q) \) indicates the lowest energy in the total spin sector \( S_i \) and lattice wavenumber \( q = 2\pi l/N_c \), with \( l = 0, 1, 2, \ldots, N_c - 1 \).

In figures 4(a) and (b) we display the lower energy one-magnon bands, in units of magnetic field, of the \( ss \) and \( ssS \) chains. For the \( ssS \) chain, we also plot in figure 4(a) non-interacting spin-wave (SW) results [26]:

\[ \omega_{\text{SW}}^-(q) = -J(S - s) + \omega_q + g\mu_B B, \]

\[ \omega_{\text{SW}}^+(q) = J(S - s) + \omega_q - g\mu_B B, \]

where \( \omega_q = J\sqrt{(S - s)^2 + 4s\sin^2(q/2)}, s = 1/2 \) and \( S = 5/2 \).

We notice that in zero field the ferromagnetic excitation is gapless (which is expected from the spontaneously broken symmetry of the GS) and displays a quadratic dispersion relation in the long wavelength limit, as predicted by conformal invariance [27], while a gap \( \Delta \) exists for the AF excitation. The ferromagnetic branch obtained through non-interacting SW theory for the \( ss \) chain is in good agreement with the ED data, while for the AF branch the value of the zero-field gap \( \Delta = 2J(S - s) = 4J \) departs from the ED value, as is often the case in other ferromagnetic systems [28], due to quantum fluctuation effects. In fact, we estimate that in the thermodynamic limit \( \Delta = 4.9046J (3.88J) \) for the \( ss \) (\( ssS \)) chain.

In figure 5(a) we show the specific heat of the \( ss \) and \( ssS \) chains in zero field. Due to the LRO ferrimagnetic state at \( T = 0 \), with low-energy gapless ferromagnetic excitations, it is expected that \( C \sim \sqrt{T} \). Another feature is the occurrence of double peaks [29–31]; it turns out that the main peak is well described by the Schottky formula [29, 30], \[ C = \frac{\delta^2}{Nk_B} \left( \frac{A}{2k_B T} \right)^2 \left( \frac{\delta^2}{2k_B T} \right)^2, \] where \( \delta \) is the Schottky gap and \( A \) is the amplitude parameter. The Schottky gap for the \( ss \) chain \( \Delta_{ss} \approx 4.1J \) is in accord with the AF spin-wave gap \( \Delta_{ss} \approx 3.9J \). However, for the \( ss \) chain the value of \( \delta_{ss} \approx 3.4J \) significantly departs from the AF spin-wave gap value, \( \Delta_{ss} \approx 4.9J \), indicating strong influence of the lower
energy ferromagnetic excitations. Since these states have a total spin \(S_t = S_g - 1\) lower than the one of the AF branch \((S_t = S_g + 1)\), we expect that a field \(B\) can wash it out. In fact, the center of the AF branch (see figure 4(a)) is found at \(\Delta_{\text{AF}}(0) = 5.4J\) and is lowered in the presence of a magnetic field through \(\Delta_{\text{AF}}(B) = \Delta_{\text{AF}}(0) - g\mu_B B\). In figure 5(b) we present the specific heat for fields up to 103.8 T, and in figure 5(c) we compare \(\Delta_{\text{AF}}(B)\) with the Schottky gap \(\Delta_{\text{S}}(B)\). As we can see in the figure, the values of the two quantities are nearly equal for moderate values of \(B\).

5. \(T-B\) phase diagram

The GS magnetization per unit cell, \(m_c\), of one-dimensional systems under an applied magnetic field can exhibit plateaus at values such that \(S_c - m_c = \text{integer}\), where \(S_c\) is the maximum total spin of a unit cell [7]. This condition implies that a plateau can be observed at values of \(m_c\) differing from its saturated value by an integer number of spin flips. In particular, a magnetization plateau at 1/3 of the saturation magnetization was observed in the magnetization curve of the mineral azurite [32], which is generally modeled through the distorted diamond chain [33]. Other compounds exhibiting the 1/3 magnetization plateau are the trimer chain systems \(\text{Cu}_2(\text{PO}_3)_3\) [34] and the phosphates [9] \(A_3\text{Cu}_3(\text{PO}_4)_4\), where \(A = \text{Ca}, \text{Sr} \text{or Pb}\). Further, the thermal properties of a variety of models [35–37] presenting plateaus in their magnetization curves have been analyzed in recent years and it was evidenced that the 1/3 magnetization plateau is also a characteristic feature of frustrated spin-S chains [38].

For the \(sS\) chain studied here, possible plateaus should be observed at \(m_c = m_{1M} = g\mu_B(S - s)\) (Lieb–Mattis (LM) magnetization) and \(m_c = m_{1P} = g\mu_B(S + s)\) (fully polarized magnetization), as confirmed by the numerical results shown in figure 6(a). In zero field the GS is ferrimagnetic with gapless ferromagnetic excitations (equation (6)), while for \(B \neq 0\) this mode acquires the gap \(\Delta_{\text{AF}}(B) = g\mu_B B\). Also, as \(B\) increases, the gap for the AF mode (equation (7)) decreases linearly with \(B\), \(\Delta_{\text{AF}}(B) = \Delta_{\text{AF}}(0) - g\mu_B B\), and for \(g\mu_B B \geq \Delta_{\text{AF}}(0)\) its gap is equal to the ferromagnetic one. At \(g\mu_B B = \Delta_{\text{AF}}(0) = g\mu_B B_{c,AF}\) the AF gap vanishes and the system undergoes a quantum phase transition (condensation of AF magnons, each carrying a spin \(+1\)) to a gapless Luttinger liquid (LL) phase [40], with power-law decay of the transverse correlation functions. In fact, the quantum critical point \(B_{c,AF}\) separates an incompressible phase (plateau) from a compressible one (LL phase). For \(B > B_{c,AF}\), a low density of magnons is found in the system and the asymptotic singular form of the magnetization can be obtained [41] by considering the system as a free Fermi gas or hard-core bosons. In this limit, the magnons will occupy single particle states with \(q \rightarrow 0\) and the dispersion relation, equation (7), can be used by replacing the linear spin-wave gap, \(\Delta_{\text{SW}} = 2J(S - s)\), by the computed gap in figure 4(a),
The density of magnons is given by
\[ \omega_{AF}^+ = -\mu + \frac{v^2}{2\Delta_S}q^2, \quad q \to 0, \]  
where \( v = J\sqrt{2sS} = J\sqrt{5/2} \) and \( \mu = g\mu_B B - \Delta_S = g\mu_B (B - B_{c,AF}) \). The energy density can thus be written (fermionic map) as
\[ \epsilon = \int_{-k_F}^{k_F} \frac{dk}{2\pi} (\epsilon_k - \mu), \]
where \( \epsilon_k = v^2 k^2 / 2\Delta_S \), \( k_F = \pi n \), and \( n \) is the density of particles. The value of \( n \) for a prescribed \( \mu \) can be obtained from the condition \( \partial_n \epsilon = 0 \):
\[ n = \sqrt{g\mu_B} \sqrt{\frac{2B_{c,AF}}{\pi^2v^2} \sqrt{\mu}}, \]  
which implies
\[ \frac{m_c}{g\mu_B} = 2 + \frac{g\mu_B}{J} \sqrt{\frac{4B_{c,AF}}{5\pi^2} (B - B_{c,AF})}, \]  
In figure 6(a) we show the very good agreement between the numerical data and \( m_c \) given by equation (11) in the LL phase.

The gapless LL phase ends at the quantum critical point \( B = B_{c,FP} \); the system becomes fully polarized (FP) and presents gapped low-energy excitations. The two one-magnon excitations from the FP state, both carrying a spin \( -\frac{1}{2} \), can be exactly obtained [42] and the lower one has a dispersion relation given by
\[ \omega_{FP} = \sqrt{J} (s + S) - \sqrt{(s - S)^2 + 4s^2 \cos^2(\sqrt{q/2})} + g\mu_B B, \]  
which implies \( g\mu_B B_{c,FP} = 2\sqrt{(s + S)} = 6J \), in accord with the numerical results (figure 6(a)). For \( B \lesssim B_{c,FP} \), a low density of magnons is observed in the system and the same arguments used to obtain equation (10) can be used in this case. For \( q \to 0 \), equation (12) can be written as equation (8) with \( v = J\sqrt{2sS} = J\sqrt{5/2} \), \( \Delta_F = 2\sqrt{(s + S)} = 6J \) and \( \mu = g\mu_B (B_{c,FP} - B) \), which implies, from equation (10), that the density of magnons is given by
\[ n = \frac{g\mu_B}{J} \sqrt{\frac{4B_{c,FP}}{5\pi^2} \sqrt{B_{c,FP} - B}}, \]
and \( m_c \) now reads
\[ \frac{m_c}{g\mu_B} = 3 + \frac{g\mu_B}{J} \sqrt{\frac{4B_{c,FP}}{5\pi^2} \sqrt{B_{c,FP} - B}}, \]
which is plotted in figure 6(a) and is also in very good agreement with the numerical data.

In figure 6(b) we present FTLM data for \( m_c \) and \( \chi \) versus \( B \) for \( T \neq 0 \). We first notice that the magnetization in zero field is null and the system is in the thermal paramagnetic state, as expected from the Mermin–Wagner theorem [22]. Increasing \( B \) in the low-temperature regime, the LM (or ferrimagnetic) plateau is exponentially reached and the magnetization exhibits an inflection point at \( B = B_m \) (red diamond in figure 6(b)) that marks the changing of the gapped low-energy excitations from ferrimagnetic (\( B \leq B_m \)) to AF magnons (\( B \geq B_m \)); the ferromagnetic (antiferromagnetic) magnons are exponentially activated and \( m_c \) is lower (higher) than \( m_{LM} = g\mu_B (S - s) \). Also, by the same token, the FP plateau is exponentially reached from below for fields higher than \( B_{c,FP} \). Furthermore, the singular form of the magnetization near the quantum critical points (\( B = B_{c,AF} \) and \( B = B_{c,FP} \), at \( T = 0 \)), which implies \( \chi \to \infty \), is thermally smoothed out and the singularities in the susceptibility evolve into local maxima, thus providing the determination of the crossover lines. The LL phase, with linear dispersion relation \( \sim q \), is expected [40] between the two local maxima for a given \( T \) (see, e.g., the susceptibility curves for \( k_B T = 0.10J \) and \( 0.20J \) in figure 6(b)) with the two local maxima indicating a crossover to a region in which the excitations follow a non-relativistic dispersion relation \( \sim q^2 \), as previously discussed. On the other hand, as \( T \) increases, the LL phase ends and a single maximum is observed in the susceptibility curves (see, e.g., the susceptibility for \( k_B T = 0.40J \)). This single maximum defines a crossover from the regime in which the physics is determined by the excitations from the LM plateau to a regime in which the FP plateau is the relevant one. For sufficiently high temperatures, the system loses all information about the \( T = 0 \) LM magnetization plateau and the effect of \( B \) is to bring the system from the thermal paramagnetic state to the FP state at higher magnetic fields (see the case \( k_B T = 2.00J \)).

In figure 7 we present the contour plot of \( m_c \) in the \( T-B \) plane and a schematic phase diagram. The \( T-B \) crossover lines enclosing the region of the LL phase, limited at \( T = 0 \) by \( B = B_{c,AF} \) and \( B = B_{c,FP} \), are obtained [43] from the local extrema of \( m_c(T) \) versus \( T \) for a given \( B \), as shown in figure 8(a). Further, as \( B \to B_c \) these crossover lines follow a universal function [43]: \( a(B - B_c) \) with \( a = 0.76238 \); as shown in figure 7, our numerical data confirm this asymptotic behavior for the two quantum critical points at \( B = B_{c,AF} \) and \( B_{c,FP} \). Moreover, as \( T \) increases beyond the crossover
the magnon densities [40], the excited magnons, as discussed above. On the other hand, the critical point with dynamical exponent in each region the system is thus governed by the quantum points [40, 44]. Last, we stress that the crossover lines and the lines of the two plateaus, gapless phases are reached [40, 44]. In addition, by increasing $B$ under a fixed $T$, local maxima are observed in the specific heat per cell $C$ as a function of $B$ for the indicated values of temperature. Scaling of the magnon density $n$ around the quantum critical points at $B = B_{c, AF}$ and $B = B_{c, FP}$.

as shown in figures 8(c) and (d). A better scaling behavior is observed for $B < B_{c, AF}$ ($B > B_{c, FP}$) in figure 8(c) (8(d)) since for $B > B_{c, AF}$ ($B < B_{c, FP}$) the zone of influence of the quantum critical point at $B = B_{c, FP}$ ($B = B_{c, AF}$) merges with the zone of influence of the point at $B = B_{c, AF}$ ($B = B_{c, FP}$). The guideline $k_B T = g \mu_B B$ in figure 7 is discussed below.

In figure 9 we present the contour plot of $C/T$ in the $B$ phase diagram [5], including the above-discussed crossover lines. At the plateaus, $C/T \to 0$ as $T \to 0$ due to the gaps, as evidenced in the plot. As we can see, the guideline $k_B T = g \mu_B B$ does not coincide with the local maxima of $C(B)$ in the low-$B$ region (see figure 8(b)) due to the LRO ferrimagnetic state at $T = 0 = B$; since $C \sim \sqrt{T}$, $C/T \to \infty$ as $T \to 0$ at $B = 0$ and an enhancement in the intensity of $C/T$ is observed near $T = 0 = B$. In spite of this fact, the plot shows a depression in the values of $C/T$ near the $T = 0$ LM plateau which, by increasing $T$, varies in a symmetrical fashion with respect to $B = B_m$ (dome-shaped) and is limited by the $k_B T = g \mu_B B$ and $k_B T = g \mu_B (B_{c, AF} - B)$ asymptotic crossover lines. Further, the LL dome is also clearly seen and the crossover lines of the FP and LM gapped phases can be visualized.

Next, we exhibit in figure 10 the magnetization of the $sS$ chain at $T = 0$. For this chain, the first plateau is found at $m_c = g \mu_B (S - 2 \alpha)$, i.e., the LM plateau, and the second is the FP plateau at $m_c = g \mu_B (S + 2 \alpha)$; the LL phase is expected to occur between these two plateaus. A third plateau could be found [7] at $m_c = g \mu_B S$; however our numerical analysis shows no evidence of this plateau. We remark that we did not perform a detailed analysis of the $T$–$B$ phase diagram of this chain, but we expect that it should display similar features to those already reported for the $sS$ chain.

The huge values of the quantum critical magnetic fields of the CuMnDTO ($sS$ chain) and MnNN ($ssS$ chain) compounds make the experimental investigation of the full $T$–$B$ phase diagram of these systems very difficult. However, magnetic phase transitions induced by very large magnetic fields (up to 400 T) in the low-temperature regime have been reported [46].
Further, materials physically described by similar models may have lower values for the exchange coupling and thus a more experimentally accessible phase diagram.

We also mention that ferrimagnetism can be destabilized by competing (or frustrating) interactions [47, 48], which can give rise to other critical points. Unconventional ferrimagnetism (non-bipartite lattices) was indeed found in one-dimensional frustrated structures [49, 50] and in the Kagomé lattice [51]. Further, the magnetocaloric effect in the kinetically frustrated diamond chain was recently investigated [52].

6. Low-temperature magnetic susceptibility

We now consider the temperature regime where ferromagnetic excitations tend to be a predominant feature. In order to test and illustrate the accuracy of the FTLM in describing the susceptibility behavior at very low temperatures, we have calculated the susceptibility of the spin-1/2 linear ferromagnetic chain; the results for \( \chi \) as a function of \( T \) are shown in figure 11 for systems with 8 and 24 sites. The crossover to zero of the FTLM results as \( T \to 0 \) is due to finite-size effects. We note that for \( (k_B T/J) \geq 0.3 \) the curves for the two chain sizes superimpose, thus suggesting that the thermodynamic-limit behavior is already attained, within numerical accuracy. Also, in the temperature range 0.06 \( \lesssim (k_B T/J) \approx 0.1 \), the results for the larger system are in good agreement with the expansion formula from Takahashi’s MSW theory [23], which up to second order in \( t \equiv k_B T/J \) reads

\[
\chi_J^{(2)} = \frac{A}{g \mu_B} = t^{-2} \left[ \frac{3}{2} s^2 - 25 s^2 A t^2 + sA^2 t + O(t^3) \right],
\]

where

\[
A = \frac{\chi(\frac{1}{2})}{\sqrt{2\pi}} \approx -0.582597 \quad \text{and} \quad g = 2.
\]

For \( s = 1/2 \) we obtain

\[
\chi_J^{\frac{1}{2}} = \frac{\chi_J}{g \mu_B} = t^{-2} \left[ \frac{3}{2} s + 0.145649 t + 0.16971 t^2 + O(t^3) \right].
\]

(15)

We stress that in the range \( 0 < (k_B T/J) < 0.1 \), equation (15) is in very good agreement with predictions from the Bethe ansatz approach [53], while the fitting of the FTLM results for \( N = 8 \) and \( 0.5 < (k_B T/J) < 0.9 \) yields \( a_0 = 0.140 \) and \( a_1 = 0.186 \), in good agreement with the MSW coefficients.

We now turn our attention to the low-temperature regime of the \( s \)-chain susceptibility displayed in figure 12. Firstly, we note that for \( (k_B T/J) \geq 0.5 \) the FTLM results for \( N = 8 \) and 14 (not shown) and \( N = 16 \) (8 cells) coincide, indicating that the thermodynamic limit has been attained in this temperature range. The experimental data normalized by \( J/k_B = 44.8 \quad \text{K} \) (\( g = 1.88 \)) and \( J/k_B = 59.7 \quad \text{K} \) (\( g = 1.9 \)) show the expected agreement with the FTLM results and the semiclassical formula, respectively, as already displayed in figure 2(a). The theoretical MSW results from the expansion formula derived by Yamamoto et al [29], which up to second order in \( t \) reads

\[
\chi_J^{\frac{1}{2}} = \frac{\chi_J}{g \mu_B} = t^{-2} \left[ \frac{3}{2} s + 0.145649 t + 0.16971 t^2 + O(t^3) \right].
\]
we recover the Takahashi expansion for the ferromagnetic agreement for the half-integer-power coefficient. For the integer-power coefficient and an order-of-magnitude of approaching the constant value $\chi \propto T^{\gamma}$, one has $\gamma < 0$, implying that $\chi T^2 \to 0$ as $T \to 0$.

7. Summary and discussion

We have presented a thorough numerical study of the GS and thermodynamic properties of two one-dimensional models related to quasi-one-dimensional ferrimagnetic compounds: CuMnDTO and MnNN. In fact, the models are associated with two types of ferrimagnetic chain: the alternating spin-1/2 spin-5/2 chain and the spin-1/2 spin-5/2 alternating chain with a spin-1/2 pendant attached to the spin-5/2 site. The finite-temperature Lanczos method proved quite reliable, except at very low temperatures where finite-size effects hinder its accuracy. A particular feature of these systems is the presence of gapless ferromagnetic and gapped AF spin-wave (magnon) branches in zero field. As the magnetic field is increased, the low-energy excitation changes from ferromagnetic to AF and the magnetic field versus temperature phase diagram displays characteristic crossover lines which distinguish these systems from spin-1 Haldane chains and two-leg ladder models. In particular, for the $s$ chain we have identified the quantum critical points and the crossover lines, the Luttinger liquid phase, the ferrimagnetic (LM) and the fully polarized plateaus.

The values of the exchange coupling parameters of the compounds discussed in the text are indeed very high. However, magnetic phase transitions induced by very large magnetic fields (up to 400 T) in the low-temperature regime have been experimentally investigated [46]. Also, other compounds described by similar models can have lower values of the exchange parameters and more experimentally accessible phase diagrams. We anticipate that this work will stimulate experimental and theoretical research with focus on the phase transitions induced by an applied magnetic field in the low-temperature regime of the large class of quasi-one-dimensional ferrimagnetic compounds.

Acknowledgments

This work was supported by CNPq, FACEPE, CAPES, and Finep (Brazilian agencies).

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