Magnetization Plateaux in the Antiferromagnetic Ising Chain with Single-Ion Anisotropy

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Two one-dimensional spin-1 antiferromagnetic Ising models with a single-ion anisotropy under external magnetic field at low temperatures are exactly investigated by the transfer-matrix technique. The magnetization per spin ($m$) is obtained for the two types of models (denoted by model 1 and 2) as an explicit function of the magnetic field ($H$) and of the anisotropy parameter ($D$). Model 1 is an extension of the recently one treated by Ohanyan and Ananikian [Phys. Lett. A \textbf{307} (2003) 76]: we have generalized their model to the spin-1 case and a single-ion anisotropy term have been included. In the limit of positive (or null) anisotropy ($D \geq 0$) and strong antiferromagnetic coupling ($\alpha = J_A/J_F \geq 3$) the $m \times H$ curves are qualitatively the same as for the spin $S = 1/2$ case, with the presence of only one plateau at $m/m_{\text{sat}} = 1/3$. On the other hand, for negative anisotropy ($D < 0$) we observe more plateaux ($m = 1/6$ and $2/3$), which depend on the values of $D$ and $\alpha$. The second model (model 2) is the same as the one recently studied by Chen et al. [J. Mag. Mag. Mat. \textbf{262} (2003) 258] using Monte Carlo simulation; here, the model is treated within an exact transfer-matrix framework.

PACS NUMBER: 75.10.Hk, 75.10.FP\textsuperscript{g}, 75.40.Cx, 75.50.Ee.
Keywords: A. Antiferromagnetic models; A. One-dimensional systems; D. Magnetic-field effects; D. Magnetization plateaux.

I. INTRODUCTION

For some one-dimensional (1D) antiferromagnets at low temperatures, it has been observed a spin gap, which is induced by a finite magnetic field, and a plateaux structure appears in the magnetization process. Experimentally, the magnetization plateaux were observed in high-field measurements of several magnetic materials such as the quasi one-dimensional compounds SrCu$_2$O$_2$, Y$_2$BaNiO$_5$, Ni(C$_2$H$_8$N$_2$)$_2$NO$_2$ClO$_4$ (abbreviated NENP), and Cu(NO$_3$)$_2$2.5H$_2$O, the triangular antiferromagnets C$_6$Eu, CsCuCl$_3$, and RbFe(MoO$_4$)$_2$, and the quasi two-dimensional compound,
with a Shastry-Sutherland lattice structure, SrCu$_2$(BO$_3$)$_2$. The mechanism for the appearance of these magnetization plateaux in quasi one-dimensional spin chains are dimerization, frustration, single-ion anisotropy, periodic field and so on.

From a general view point, Oshikawa et al.\(^\text{10}\) concluded that the necessary condition for the magnetization plateaux in spin-$S$ chains is $Q(S - m) =$integer, where $Q$ is the spatial periodicity of the magnetic ground state and $m$ is the magnetization per site. For some range of the magnetic field $H$ (i.e., $H_1 < H < H_2$), the system ceases responding to its increase and a plateau is formed in the magnetization versus the magnetic field curve. The values of $m$ at which the plateaux appear are sensitive to small changes in the parameters of the model and are not only restricted to integer spin (Haldane conjecture)\(^\text{11}\).

In the $S = 1/2$ antiferromagnetic Heisenberg model on a triangular lattice, a magnetization plateau was found at $m/m_{\text{sat}} = 1/3^6.7.8.12$. In an $S = 1/2$ trimerized Heisenberg model\(^\text{13}\), the plateau appears at $m/m_{\text{sat}} = 1/6$. Recently, plateaux at $m/m_{\text{sat}} = 1/8$ and $1/4$ have been observed in the SrCu$_2$(BO$_3$)$_2$, which has a Shastry-Sutherland lattice structure. However, irrational values have not been found, at least so far. Theoretically, various other models with spin $S = 1/2$ have been proposed to describe the magnetization plateaux. One of the first models was introduced by Hida\(^\text{14}\), where a Heisenberg chain was considered, with antiferromagnetically coupled ferromagnetic trimers ($p = 3$). The three-dimerized Hamiltonian proposed by Hida to describe the 3CuCl$_2$.2 dioxane compound is given by

$$H = H^{\text{trim}} + H^{\text{int}} + H^{\text{Zeeman}}, \quad (1)$$

with

$$H^{\text{trim}} = -J_F \sum_i (S_i \cdot \tau_i + \tau_i \cdot \sigma_i), \quad (2)$$

$$H^{\text{int}} = J_A \sum_i \sigma_i \cdot S_{i+1}, \quad (3)$$

and

$$H^{\text{Zeeman}} = -\mu_B H \sum_i (S_i^z + \sigma_i^z + \tau_i^z), \quad (4)$$

where $J_A$ and $J_F$ are the antiferromagnetic and ferromagnetic interactions, respectively, $S_i, \tau_i$ and $\sigma_i$ are the $S = 1/2$ spin operators at site $i$, $\mu_B$ is the Bohr magneton and $H$ is the magnetic field. Using exact diagonalization of finite systems, Hida obtained, for $J_F$ comparable to or smaller than
$J_A$, a plateau at $m/m_{sat} = 1/3$. The plateau mechanism was considered to be a purely quantum phenomenon, where the concepts of magnetic quasiparticles and strong quantum fluctuations are regarded to be of major importance for understanding the process. On the other hand, Ohanyan and Ananikian have recently studied the Hida model by using the transfer-matrix technique, replacing the spin operators ($S_i, \tau_i$ and $\sigma_i$) by Ising variables ($S^x_i, \sigma^x_i, \tau^x_i$). It was shown that, for this classical model and for $T = 0$ (ground state) and $J_A \geq 3J_F$ (strong antiferromagnetic coupling), a magnetization curve with plateau at $m/m_{sat} = 1/3$ is observed, indicating that the appearance of plateaux is not a quantum manifestation, but may be caused by the stability of spatially modulated spin structures.

Another model which presents magnetization plateaux is the one-dimensional spin-1 antiferromagnetic Heisenberg with single-ion anisotropy. This model is described by the following Hamiltonian

$$H = J \sum_i S_i \cdot S_{i+1} - \mu_{B} H \sum_i S^z_i + D \sum_i (S^z_i)^2,$$

(5)

where $D$ is the single-ion anisotropy. For $D = 0$, the ground state is a singlet and the lowest excitation is a triplet (Haldane conjecture); increasing $D$, the triplet splits into a higher-energy singlet and a lower-lying doublet, with the Haldane gap for $D = 0$, $\Delta(0)$, splitting into two gaps, as observed in neutron scattering of NENP. The Haldane gap for general $D$, $\Delta(D)$, presents two different behaviors: for $D > D_c = J$, it increases with $D$, while for $D < D_c$ $\Delta(D)$ decreases as $D$ increases.

Recently, spin $S \geq 1$ Ising antiferromagnetic chains with single-ion anisotropy have been studied by using classical Monte Carlo simulation and it was observed the presence of $2S + 1$ plateaux for $D > 0$. Essentially, these classical models are obtained replacing the spin operators ($S_i$) by Ising variables ($S^z_i$) in Hamiltonian (5). From a theoretical point view, the model studied by Chen, et. al. represent the 1D antiferromagnetic Blume-Capel model, and it was observed also two different critical behaviors, which depend on the anisotropy parameter $D$ ($D < D_c$ and $D > D_c$, where $D_c = J$).

The purpose of this work is to obtain exact results for two classical models with spin $S = 1$ and in the presence of a single-ion anisotropy. In Section 2 the 1D models are presented and exactly solved by the transfer-matrix technique. The magnetization plateaux and ground-state phase diagrams are discussed in Section 3. Finally, the last section is devoted to conclusions.
II. MODELS AND FORMALISM

The transfer-matrix technique was proposed years ago by Kramers and Wannier\textsuperscript{20,21}, and it formed the basis for Onsager’s solution\textsuperscript{22} of the two-dimensional Ising model. In this section, we use this technique to obtain exact results for two one-dimensional models, in order to analyze the magnetization plateau mechanism.

A. Model 1: Three-dimerized chain

The first model we study is described by the following Hamiltonian:

\[
\mathcal{H}_1 = -J_F \sum_i (S_i^z \cdot \tau_i^z + \sigma_i^z \cdot \tau_i^z - \alpha \sigma_i^z \cdot S_{i+1}^z) - \mu_B H \sum_i (S_i^z + \tau_i^z + \sigma_i^z) - D \sum_i \left[ (S_i^z)^2 + (\tau_i^z)^2 + (\sigma_i^z)^2 \right],
\]

(6)

where \( \alpha = J_A/J_F \) and the spin variables \( S_i^z, \tau_i^z \) and \( \sigma_i^z \) can assume the values \(-1, 0, 1\). The above Hamiltonian represents a nonuniform spin system in which ferromagnetic trimers composed of \( S = 1 \) spins (\( S_i^z, \tau_i^z \) and \( \sigma_i^z \)) are coupled antiferromagnetically in one dimension, in the presence of a magnetic field (\( H \)) and single-ion anisotropy (\( D \)). In the limit \( \alpha \to 0 \) (strong intratrimer ferromagnetic interaction), the variables \( S_i^z, \tau_i^z \) and \( \sigma_i^z \) form a single spin \( \xi_i \) with magnitude 3. Thus, the system can be approximated by a spin \( S = 3 \) antiferromagnetic Blume-Capel chain.

The transfer-matrix technique is based in the calculations of the eigenvalues \( \{ \lambda_i \} \), determined from the solution of the secular equation

\[
\det(W_1 - \lambda I) = 0,
\]

(7)

where \( I \) is the identity matrix \( 3 \times 3 \) and \( W_1 \) the Wannier matrix, with the elements defined by

\[
W_1(S, S') = \sum_{\sigma, \tau = 0, \pm 1} \exp[a(\tau)S + dS^2 + b(\sigma)S' + c(\tau, \sigma)],
\]

(8)

with

\[
a(\tau) = \beta J_F \tau + \beta \mu_B H,
\]

(9)

\[
b(\sigma) = -\alpha \beta J_F \sigma,
\]

(10)

\[
c(\tau, \sigma) = \beta J_F \sigma \tau + \beta \mu_B H (\tau + \sigma) + \beta D (\tau^2 + \sigma^2),
\]

(11)
and
\[ d = \beta D, \]  
(12)

where \( S, S' = 0, \pm 1 \).

Using properties of the matrix trace, the partition function \( Z = Tr(W^N) \) can be written as a sum of the \( N \)th power of the eigenvalues \( \{\lambda_i\} \) obtained from Eq.(7), i.e.,
\[ Z = \sum_{i=1}^{3} \lambda_i^N. \]  
(13)

In the thermodynamic limit \( (N \to \infty) \), the free energy, magnetization, magnetic susceptibility and specific heat (per atom) are expressed in terms of maximum eigenvalue \( \lambda_{\text{max}} \), respectively, as
\[ f = -\frac{T}{3} \ln \lambda_{\text{max}}, \]  
(14)
\[ m = \frac{T}{3 \lambda_{\text{max}}} \frac{\partial \lambda_{\text{max}}}{\partial H}, \]  
(15)
\[ \chi = \frac{\partial m}{\partial H} = \frac{T}{3} \frac{\partial}{\partial H} \left( \frac{1}{\lambda_{\text{max}}} \frac{\partial \lambda_{\text{max}}}{\partial H} \right), \]  
(16)

and
\[ c = \frac{2T}{3 \lambda_{\text{max}}} \frac{\partial \lambda_{\text{max}}}{\partial T} + T^2 \frac{\partial}{\partial T} \left( \frac{1}{\lambda_{\text{max}}} \frac{\partial \lambda_{\text{max}}}{\partial T} \right), \]  
(17)

where the factor \( 1/3 \) was introduced because there are three spins in each site of the chain, and the maximum eigenvalue \( \lambda_{\text{max}} \) is given by
\[ \lambda_{\text{max}} = -\frac{A}{3} + 2\sqrt{Q \cos \left( \frac{\theta}{3} \right)}, \]  
(18)
with
\[ A = W_{1}(1,1) + W_{1}(0,0) + W_{1}(-1,-1) = Tr(W_{1}), \]  
(19)
\[ Q = \frac{A + 3B}{9}, \]  
(20)
\[ B = W_{1}(1,0)W_{1}(0,1) + W_{1}(1,-1)W_{1}(-1,1) + W_{1}(-1,0)W_{1}(0,-1) - W_{1}(1,1)W_{1}(0,0) - W_{1}(1,-1)W_{1}(-1,1) - W_{1}(0,0)W_{1}(-1,-1), \]  
(21)
\[ R = \frac{9AB - 27C - 2A^3}{54}, \]  
(22)
where \( C = -\det(W_1) \), \( \lambda_{\text{max}} \) is given by Eq. (18), and \( \alpha = J_A/J_F \), \( \delta = D/J_F \), and on the reduced anisotropy parameter, \( \delta = D/J_F \).

For \( \delta \geq 0 \), the qualitative results are the same as those obtained by Ohanyan and Ananikian, namely: for \( \alpha > \alpha_c(\delta) \) (strong antiferromagnetic coupling) and \( T = 0 \) a magnetization plateau appears at \( m = 1/3 \), for magnetic fields in the interval \( H \in [H_{c1}, H_{c2}] \), where \( h_{c1} \equiv H_{c1}/J_F = 2.0 \) and \( h_{c2} \equiv H_{c2}/J_F = \alpha - 1.0 \). We have obtained, in this strong antiferromagnetic regime and positive anisotropy, that the value of the critical ratio \( \alpha_c \) does not depend on \( \delta \), i.e.,
\( \alpha_c(\delta) = 3.0 \). For infinity anisotropy \( (\delta \to \infty) \) our results reduce to the case of the three-dimerized Ising chain with spin 1/2. So, in this case, we obtain the same magnetization plateaux as in Ref.14.

The ground state \( (T = 0) \), in the absence of a magnetic field \( (H = 0) \), is the antiferromagnetic spatially modulated structure, in which trimers of spins pointing up \( (S_i = 1) \) alternate with trimers of spins pointing down \( (S_i = -1) \) (i.e., .......... ↑↑↑↓↓↓↑↑↑ ...... and so on), for all values of \( \delta \geq 0 \) and \( \alpha \). We denote this modulated phase by \langle 3 \rangle. In the low field region \( (H < H_{c_1}) \), no magnetization is observed \( (m = 0) \). When the magnitude of the external magnetic field increases, at the critical value \( H_{c_1} \) the system passes from its ground state \langle 3 \rangle to the novel spatially modulated structure \langle 3111 \rangle, in which the periodic sequence of spins consists of one trimer pointing along the field, in the spin state \( S_i = 1 \) (↑↑↑), and another trimer with alternating orientation of spins (↓↑↓). In this spin state \langle 3111 \rangle, \( m = 1/3 \) and increases discontinuously to the saturation value \( m = 1 \) at the second critical field \( h_{c_2} = \alpha - 1.0 \). In the above one-dimensional model, the indispensable condition for the appearance of the plateau at \( m = 1/3 \) is the strong antiferromagnetic coupling, characterized by \( \alpha > 3.0 \). Therefore, no plateau is found for \( \alpha \leq 3.0 \).

On the other hand, when the anisotropy is negative the spin state \( S_i = 0 \) (represented by \( \bigcirc \)) is energetically favorable, when compared to the spin states \( S_i = 1 \) and \( -1 \). For certain values of \( \alpha \) and \( \delta < 0 \), we can observe various types of phase transition: first, a flip of the central spin to the state \( S_i = 0 \) with a modulated structure \langle 3101 \rangle (corresponding to the spin configuration .......... ↑↑↑↓↓↓↑↑↑ ......) and magnetization \( m = 1/6 \); second, a flip of the central spin to the state \( S_i = 1 \) with a modulated structure \langle 3111 \rangle (corresponding to the spin configuration .......... ↑↑↑↓↓↓↑↑↑ ......) and magnetization \( m = 1/3 \); third, a flip of the surface spins to the state \( S_i = 0 \) with a modulated structure \langle 3010 \rangle (corresponding to the spin configuration .......... ↑↑↑↓↓↓↑↑↑ ......) and magnetization \( m = 2/3 \), and, finally, the saturated state \langle 3^2 \rangle (corresponding to the spin configuration .......... ↑↑↑↓↓↓↑↑↑ ......), with a magnetization \( m = 1 \).

In order to obtain the values of the critical fields \( h_{c_1} \) (transition between the modulated structure \langle 3 \rangle and \langle 3101 \rangle), \( h_{c_2} \) (transition between the modulated structure \langle 3101 \rangle and \langle 3111 \rangle), \( h_{c_3} \) (transition between the modulated structure \langle 3111 \rangle and \langle 3010 \rangle) and \( h_{c_4} \) (transition between the modulated structure \langle 3010 \rangle and \langle 3^2 \rangle), we compare the energies for the respective periodic sequence, finding the following critical fields:
By solving Eq. (26), we obtain the critical frontiers which separate the various modulated phases, corresponding to magnetization plateaux at $m = 1/6$, $1/3$ and $2/3$ (see discussion in the previous paragraph). We find that, for $0 < h < h_{c1}$ and $h > h_{cs}$, the magnetizations are $m = 0$ (disordered state) and $m = 1$ (saturated state), respectively. Note that for $\delta < -2.0$ there is no disordered state for positive field $(H > 0)$. Depending on the values of the parameters $\alpha$ and $\delta < 0$, we can have various magnetization plateaux at $m = 1/6$, $1/3$ and $2/3$. In Figs 1, 2, and 3 the ground state phase diagrams for $\delta = -1.0, -2.0$, and $-3.0$ are depicted, respectively, where we indicate the various magnetization plateaux.

IV. CONCLUSIONS

We have treated two soluble models: model 1 (Eq. (6)) and model 2 (Eq. (25)), by using a transfer-matrix technique. In the thermodynamical limit ($N \to \infty$), the partition function was obtained and ground state phase diagrams were calculated, in order to analyze the plateaux structure in the magnetization. In contrast to the majority of existing approaches (numerical diagonalization) to treat the problem of magnetization plateaux in quantum models (see, for example, Ref.13), the present formalism (transfer-matrix technique) is entirely based on analytical (exact) calculations and allows for the calculation of magnetization profiles for arbitrary finite temperatures and values of the parameters $\alpha$ and $\delta$.

ACKNOWLEDGEMENT

This work was partially supported by CNPq, FAPEAM and CAPES (Brazilian Research Agencies).

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