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Magnetization plateaus
in the ferromagnetic–ferromagnetic–antiferromagnetic Ising chain

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Abstract

The Ising chain consisting of the antiferromagneticaly coupled ferromagnetic trimer is considered in the external magnetic field. In the framework of the transfer-matrix formalism the thermodynamics of the system is described. The magnetization per site (m) is obtained as the explicit function of the external magnetic field (H). The corresponding plots of $m(H)$ are drawn. Two qualitatively different regions of the values of coupling constants are established: weak antiferromagnetic coupling ($J_A < 3J_F$) and the strong antiferromagnetic coupling ($J_A \geqslant 3J_F$). For the latter case the magnetization curve with plateau at $m/m_{sat} = 1/3$ is obtained. It is proven that the plateau is caused by the stability of spatially modulated spin structure $\langle 3111 \rangle$. The values of magnetic field determining the width of the plateau are obtained in the limit of zero temperature.
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1. Introduction

Quite recently, the family of known non-trivial quantum effects in the condensed matter physics is enriched with the novel phenomenon—intermediate plateaus in the magnetization processes. Namely, the system ceases to respond on changes of the external magnetic field within some range of the latter. As a consequence, for some intermediate values of H ($H_1 < H < H_2$) a horizontal region forms in the magnetization (m) versus the external magnetic field (H) curve.

These plateau structures were experimentally observed in series of compounds: the triangular antiferromagnets (C_6Eu [1], $CsCuCl_3$ [2], $RbFe(MoO_4)_2$ [3]); quasi-one-dimensional Ni-compounds [4]; quasi-two-dimensional compound with structure of Shastry–Sutherland lattice ($SrCu_2(BO_3)_2$ [5]). At present there are numerous theoretical papers where the magnetization plateaus were obtained for various non-homogeneous Heisenberg chains [6–10], for spin-ladders [11], and as well as for some two-dimensional models [12–18].

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In the majority of plateau mechanisms which have been proposed up to now the purely quantum phenomena play a crucial role. The concepts of magnetic quasiparticles and the strong quantum fluctuations are regarded to be of first importance for understanding of these processes. Particularly, for a number of systems it was shown that the plateaus at $m \neq 0$ are caused by the presence of the spin gap in the spectrum of magnetic excitations in the external magnetic field. Another mechanism lies in so-called crystallization of the magnetic particles [7,17].

Nevertheless, we suppose that the quasiclassical picture in which the main dynamics of spins can be presented in terms of spin "up" and spin "down" is also relevant to the problem of magnetization plateaus. The problem formulated in the quasiclassical language allows to use the Ising type variable instead of the quantum spin operators. The advantages of this approximation are obvious: in the case of one-dimensional systems one can develop purely analytic and quite simple technique for the magnetization plateau problem.

One of the pioneering theoretical works where the appearance of the magnetization plateau was predicted is the paper of Hida [6]. In this paper the ferromagnetic–ferromagnetic–antiferromagnetic Heisenberg chain was considered as a model of $3CuCl_2 \cdot 2$ dioxane compound which consists of the antiferromagneticaly coupled ferromagnetic trimers. The Hamiltonian, considered by Hida has the following form:

$$\mathcal{H} = \sum_i \left(\mathcal{H}_i^{\mathrm{trim}} + \mathcal{H}_i^{\mathrm{int}} \right) + \mathcal{H}_Z, \tag{1}$$

where

$$\mathcal{H}_i^{\mathrm{trim}} = -2J_F(\boldsymbol{s}_i \cdot \boldsymbol{\tau}_i + \boldsymbol{\tau}_i \cdot \boldsymbol{\sigma}_i),$$
$$\mathcal{H}_i^{\mathrm{int}} = 2J_A(\boldsymbol{\sigma}_i \cdot \boldsymbol{s}_{i+1}),$$
$$\mathcal{H}_Z = -g\mu_B H \sum_i \left(s_i^z + \sigma_i^z + \tau_i^z \right), \tag{2}$$

here J_A and J_F are the antiferomagnetic and ferromagnetic coupling constants, respectively, $\boldsymbol{s}_i$, $\boldsymbol{\tau}_i$ and $\boldsymbol{\sigma}_i$ are the $S = 1/2$ spin operators and $\mathcal{H}_Z$ is the standard Zeeman Hamiltonian. Using the method of the numerical diagonalization of finite size system Hida obtained a magnetization curve with the plateau at $m/m_{\mathrm{sat}} = 1/3$, where m_{sat} is the saturation magnetization.

The approach we are offering in the present Letter is based on very simple idea to use the Ising spins instead of the Heisenberg operators in Eq. (1) and to develop the transfer-matrix formalism (see, for example, Ref. [19]). It is worth to emphasize that in contrast to the majority of existing approaches to the problem of magnetization plateau, our technique is entirely based on analytical calculations and allows to obtain the magnetization profiles for arbitrary finite temperatures and arbitrary values of parameters J_A and J_F. As a result we obtained the diagrams of magnetization processes for emerged ferromagnetic–ferromagnetic–antiferromagnetic Ising chain with the same magnetization plateau as in Ref. [6].

Presently, there are the numerical results indicating the existence of magnetization plateau in two-dimensional Ising models [16]. Moreover, in the Ref. [20] the multisite interaction Ising model on the infinite-dimensional recurrent lattice was considered. There the magnetization curves with plateaus at $m = 0$ and $m/m_{\mathrm{sat}} = 1/2$ were also obtained there. However, till now, there are no any evidence of the appearance of the magnetization plateaus in the one-dimensional Ising chains.

2. The ferromagnetic–ferromagnetic–antiferromagnetic Ising chain

Replacing all Heisenberg operators in Eq. (1) by the Ising variables s, t, u one can obtain the model, describing by the follow Hamiltonian:

$$-\beta\mathcal{H} = \sum_i \left(J_F(s_i t_i + t_i u_i) - J_A u_i s_{i+1} + h(s_i + t_i + u_i) \right). \tag{3}$$

Here the $J_F = \beta \tilde{J}_F$ and $J_A = \beta \tilde{J}_A$ are dimensionless ferromagnetic and antiferromagnetic constants, respectively, $h = \beta H$ is dimensionless magnetic field and all the variables s, t, u take values equal to ± 1. The partition function for one-dimensional systems can be written in the following way:

$$Z = \sum_{(s,t,u)} e^{-\beta \mathcal{H}} = \sum_{(s,t,u)} \prod_i \exp\big(J_F(s_i t_i + t_i u_i) - J_A u_i s_{i+1} + h(s_i + t_i + u_i)\big). \tag{4}$$

Here the product is going over all sites of the chain. If the periodic boundary conditions are assumed, the partition function can be represented as a trace of product of the transfer-matrices:

$$Z = \mathbf{Sp}\big(\mathbf{T}^N\big). \tag{5}$$

In our case we can write down the elements of the transfer-matrix as

$$T_{s\acute{s}} = \sum_{t,u} \exp\big(J_F(st + tu) - J_{AF}u\acute{s} + h(s + t + u)\big) = \sum_{t,u} \exp\big(\alpha_1(t)s + \alpha_2(u)\acute{s} + \alpha_3(t,u)\big), \tag{6}$$

where the following notations are introduced:

$$\alpha_1 = J_F t + h, \qquad \alpha_2 = -J_A u, \qquad \alpha_3 = J_F ut + h(t + u). \tag{7}$$

Implementing the summation on t and u in Eq. (6) one can easily obtain the explicit form of the transfer-matrix:

$$\mathbf{T} = \begin{pmatrix} 2e^{J_A}\cosh h + 2e^{-J_A+2h}\cosh(2J_F + h) & 2e^{-J_A}\cosh h + 2e^{J_A+2h}\cosh(2J_F + h) \\ 2e^{-J_A}\cosh h + 2e^{J_A-2h}\cosh(2J_F - h) & 2e^{J_A}\cosh h + 2e^{-J_A-2h}\cosh(2J_F - h) \end{pmatrix}. \tag{8}$$

Following the general procedure, using the properties of the matrix trace, $\mathbf{Sp}(\mathbf{A_1 A_2 \ldots A_k B}) = \mathbf{Sp}(\mathbf{BA_1 A_2 \ldots A_k})$ it is easy to show that the partition function from Eq. (4) for one-dimensional model is the sum of the Nth power of the eigenvalues of transfer-matrix.

$$Z = \sum_i \lambda_i^N. \tag{9}$$

It is easy to see that in the thermodynamical limit, $N \to \infty$, only the maximal eigenvalue contributes to the r.h.s. of Eq. (9). Thus, the problem of finding the partition function for the chain reduces to the problem of finding the maximal eigenvalue of the transfer-matrix. Solving the secular equation for the matrix (8), one can obtain the eigenvalues

$$\begin{aligned} \lambda_{\pm} = {}& 2\cosh h + e^{-J_A}\big(e^{2h}\cosh(2J_F + h) + e^{-2h}\cosh(2J_F - h) \\ & \pm \big((e^{2h}\cosh(2J_F + h) - e^{-2h}\cosh(2J_F - h))^2 \\ & + 4\big(\cosh h + e^{2(J_A+h)}\cosh(2J_F + h)\big)\big(\cosh h + e^{2(J_A-h)}\cosh(2J_F - h)\big)\big)^{1/2}\big). \end{aligned} \tag{10}$$

It is easy to see that for our purpose we have to take λ_+.

Thus, in the thermodynamical limit the free energy per site takes the form:

$$\begin{aligned} F = {}& \lim_{N\to\infty}\left(-\frac{\log \lambda_+^N}{3N\beta}\right) = -\frac{1}{3\beta}\log\big\{2\cosh h + e^{-J_A}\big(e^{2h}\cosh(2J_F + h) + e^{-2h}\cosh(2J_F - h) \\ & + \big((e^{2h}\cosh(2J_F + h) - e^{-2h}\cosh(2J_F - h))^2 \\ & + 4\big\{\cosh h + e^{2(J_A+h)}\cosh(2J_F + h)\big)\big(\cosh h + e^{2(J_A-h)}\cosh(2J_F - h)\big\}\big)^{1/2}\big)\big\}. \end{aligned} \tag{11}$$

The factor $1/3$ is introduced here because there are three spins in each site of the chain, and the total number of spins is $3N$. Now, using the general thermodynamical relations we can find the magnetization per site

$$m = -\left(\frac{\partial F}{\partial H}\right)_\beta = \frac{1}{3\beta\lambda_+}\left(\frac{\partial \lambda_+}{\partial H}\right)_\beta = \frac{1}{3\lambda_+}\left(\frac{\partial \lambda_+}{\partial h}\right)_\beta. \tag{12}$$

The explicit form of this expression is too cumbersome and we found no reason to write it down here. However, it is quite suitable to plot analytically the corresponding magnetization profiles (diagrams of m versus the external magnetic field H) with the aid of MATHEMATICA 4.0 package. The results are presented in the next section.

3. Magnetization processes

It is easy to see that the ground state of the system under consideration at $T = 0$ and $H = 0$ is the antiferromagnetic spatially modulated structure in which the trimers of spins pointed up are alternating with the trimers of spins pointed down ($\ldots \uparrow\uparrow\uparrow\downarrow\downarrow\downarrow\uparrow\uparrow\uparrow\downarrow\downarrow\downarrow \ldots$ and so on). Usually this modulated phase is denoted by $\langle 3 \rangle$ [21]. This is valid for all values of J_A and J_F in the absence of the external magnetic field.[1]

The profile of the magnetization as a function of the external magnetic field essentially depends on the ratio of coupling constants $\kappa = J_A/J_F$ and temperature. Actually one can distinguish two regions of values of κ: strong antiferromagnetic coupling and weak antiferromagnetic coupling. The limiting case of the latter, when the system reduces to the ordinary ferromagnetic Ising chain is $J_A = 0$. When the antiferromagnetic coupling J_A is included then the low temperature behaviour changes steeply. At $T = 0$ and $H = 0$ as is mentioned above the chain is in the ordered state with complex antiferromagnetic long-range order. At finite but sufficiently low temperatures the long-range order is breaking just by few solitons. The external magnetic field will have only small effects until it reaches a critical value H_c where it can flip a spin without cost in energy. Thus, the appearance of a magnetization plateau in the magnetization versus the external magnetic field curve is expected in this case. The width of the plateau is obviously equal to H_{c_1}. For the chain at $T = 0$ one, using the simple principles of minimal energy, can easily express H_{c_1} in terms of J_A and J_F.

Let us first consider the case when antifferromagnetic coupling between trimers is assumed to be so intensive that under the effect of the sufficiently low external magnetic field only the central spins of ferromagnetically coupled trimers flip (do respond); whereas the rest two spins are maintained in the same position as in the ground state. This regime we will call the strong antiferromagnetic coupling. So, in this case, when the magnitude of the external magnetic field reaches its critical value H_{c_1} the system will pass 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 of spins pointed along the field and another trimer with alternating orientation of spins (see Fig. 1). In order to obtain the value of the critical field H_{c_1} one must compare the energies of $\langle 3 \rangle$ and $\langle 3111 \rangle$ states and find the value of H at which the $\langle 3111 \rangle$ state becomes energetically more preferable than the ground state. Considering the single periodic spin sequences we have

$$-J_A - 2H \leqslant -4J_F - J_A, \quad \text{or} \quad H_{c_1} = 2J_F. \tag{13}$$

So, for the strong antiferromagnetic coupling regime the value of the critical field does not depend on J_A.

Now we can give a quantitative criterion for this domain of parameters. The antiferromagnetic interaction between edge spins of trimers is strong in the sense described above when the flip of the central spin does not lead to the flip of its adjacent one at the critical value of the external field calculated above. In the other words, the values of coupling J_A and J_F are such that the $\langle 3111 \rangle$ structure is energetically more preferable than the spin configuration where at least one of the spins adjacent to the central one is pointed up at $H = H_{c_1}$. Comparing corresponding energies for the single periodic sequence one can obtain the following condition of strong ferromagnetic coupling (Fig. 1):

$$-2J_A - 2H_{c_1} \leqslant -2J_F - 4H_{c_1}, \quad \text{or} \quad J_A \geqslant 3J_F. \tag{14}$$

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[1] Hereafter for convenience we will identify $\tilde{J}_A$ by J_A and $\tilde{J}_f$ by J_F.

(a)

$$E_0 = -4J_F - 2J_A$$

(b)

$$E' = -2J_A - 2H$$

H
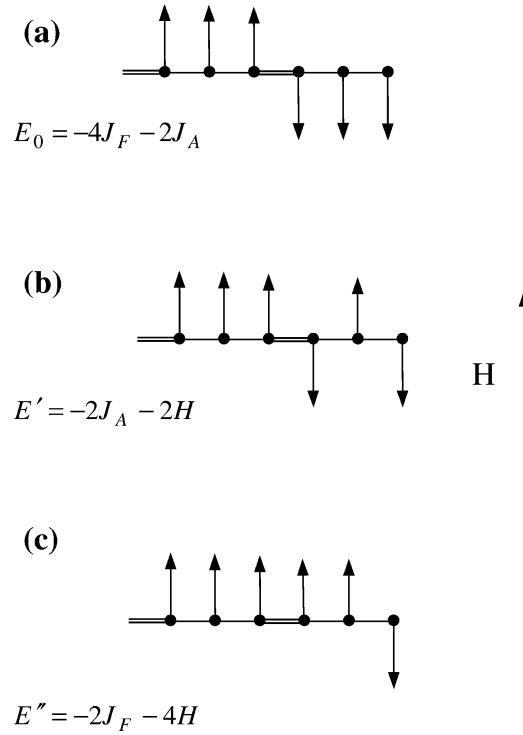
(c)

$$E'' = -2J_F - 4H$$

Fig. 1. Periodic spin sequences and corresponding energies for (a): ground state configuration at $T = 0$ and $H = 0$; (b): the plateau state, so-called $\langle 3111 \rangle$ configuration; (c): one of the possible intermediate configurations which forms after the plateau state. Double line between the sites denote the antiferromagnetic interaction J_A.

The weak antiferromagnetic coupling, $J_A < 3J_F$, supposes that the ferromagnetic interaction inside the trimers is strong enough, so that the three spins the trimer consists of behave like single Ising spin, which can take values ± 3 with effective antiferromagnetic coupling $J_{\mathrm{eff}} = J_A/3$. This case corresponds to the $S = 3/2$ singlets in the quantum model considered by Hida [6]. Thus, for $J_A < 3J_F$ the system under consideration can be regarded as the simple antiferromagetic Ising chain with the restrictions given above. The value of the critical field magnitude can be easily obtained in this case. Obviously, $H_{c_1} = 2J_{\mathrm{eff}} = \frac{2}{3}J_A$, thus,

$$H_{c_1} = \begin{cases} 2J_F, & \text{if } J_A \geqslant 3J_F, \\ \frac{2}{3}J_A, & \text{if } J_A < 3J_F. \end{cases} \tag{15}$$

The plots, presented in Fig. 2 illustrate these results. Note, that the high temperature magnetic behaviour for all regions of parameters is rather uniform and the typical magnetization profile is given by the curve of Langevin type. So, in that follows we will consider only low temperature region. At finite temperatures the values of H_{c_1} are obviously different from Eq. (15) due to existence of thermal fluctuations of spins. This difference becomes smaller with the decrease of temperature. So, for sufficiently low temperatures

$$H_{c_1}(T) = H_{c_1} - \theta(T), \quad \lim_{T \to 0} \theta(T) = 0. \tag{16}$$

Fig. 2(a)–(c) show that already at $T = 0.1J_F$ value of critical field is quite close to $\frac{2}{3}J_A$.

The regime of strong antiferromagnetic coupling is characterized by additional feature which is absent in case of $\kappa < 3$. It is the magnetization plateau at $m/m_{\mathrm{sat}} = 1/3$. Corresponding plots are presented in Fig. 3. One can easily see that the width of plateau depends on the value of κ, namely it becomes broader with the increase of κ. In
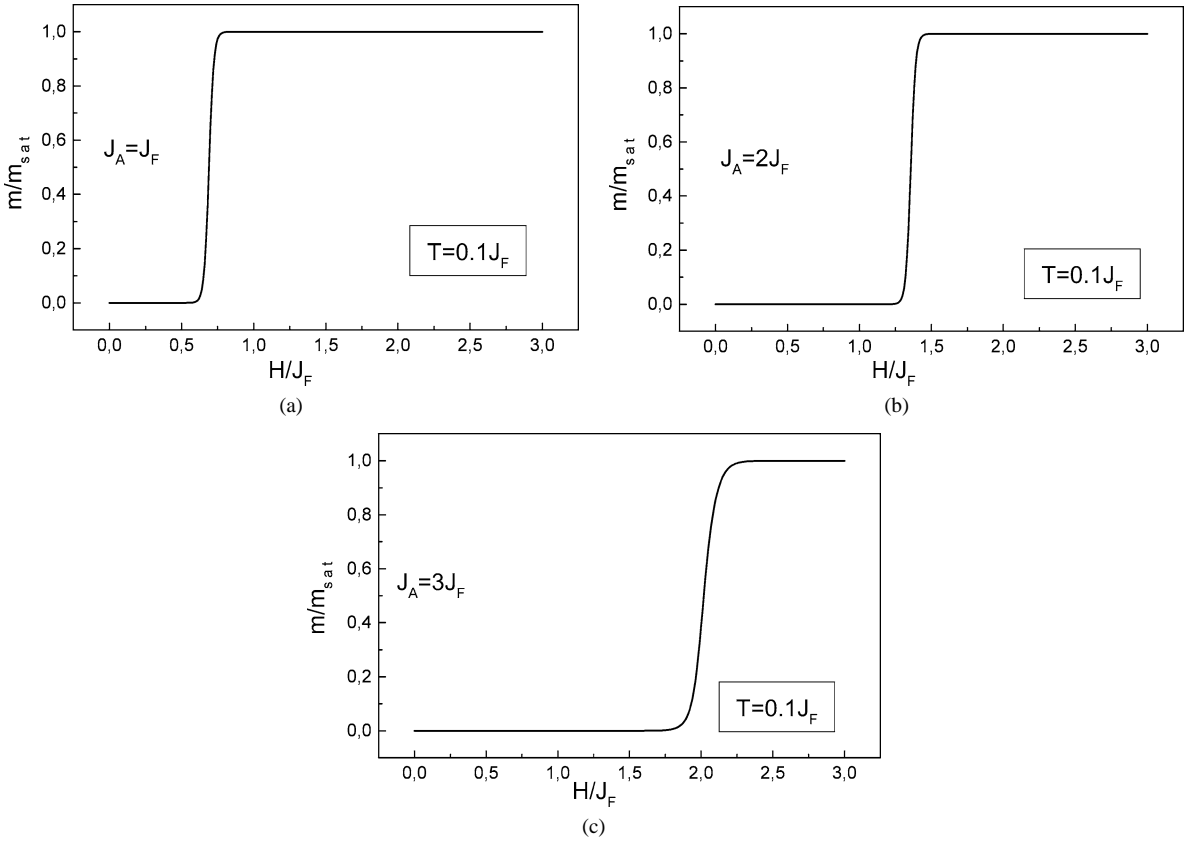
Fig. 2. Magnetization curves in the regime of weak antiferromagnetic coupling, $J_A \leqslant 3J_F$ at $T = 0.1J_F$ for different values of κ: (a) $\kappa = 1$; (b) $\kappa = 2$; (c) $\kappa = 3$. The value of the critical field magnitude in these cases is $3/2J_A$.

this case another critical value of the external magnetic field magnitude defining the width of the plateau appears. So, in the magnetization curve one can see a horizontal region for the values of H in interval $[H_{c_1}, H_{c_2}]$. With the aid of calculations analogous to those we made to obtain H_{c_1} one can easily get the second critical value H_{c_2} for the chain at $T = 0$ in the strong antiferromagnetic coupling region. Appearance of the plateau at $m/m_{\mathrm{sat}} = 1/3$ and $H = H_{c_1}$ is indication of the stability of $\langle 3111 \rangle$ configuration when $H \in [H_{c_1}, H_{c_2}]$. So, when H reaches its second critical value H_{c_2} the above mentioned structure begins to destroy due to further flip of the spins which previously were pointed down. The comparison of the corresponding energies of single periodic spin sequence yields (Fig. 1):

$$-2J_F - 4H \leqslant -2J_A - 2H, \quad \text{or} \quad H_{c_2} = J_A - J_F, \tag{17}$$

this is in full agreement with Fig. 3. The effects of finiteness of temperature play the same role as in the case of H_{c_1} i.e., they can only bring a slight decrease of the H_{c_2} at low values of T. It is worth to note once more that high temperatures always lead to disappearance of plateau due to strong intensity of thermal fluctuations of spins which in their turn destroy the ordered configuration. From all stated above we can conclude that the ferromagnetic–ferromagnetic–antiferromagnetic trimerized Ising chain in the region of strong antiferromagnetic coupling have a magnetization plateau, its width in the limit of zero temperature is equal to $H_{c_2} - H_{c_1} = J_A - 3J_F$.

In Fig. 4 the plots of magnetization for given values of coupling ($\kappa = 6$) and different values of temperature are shown. At $T = 2.5J_F$ the curve has no deviations from the usual high temperature magnetic behaviour of spin systems (Fig. 4(a)). But with decrease of temperature some peculiarities appear in the course of magnetization
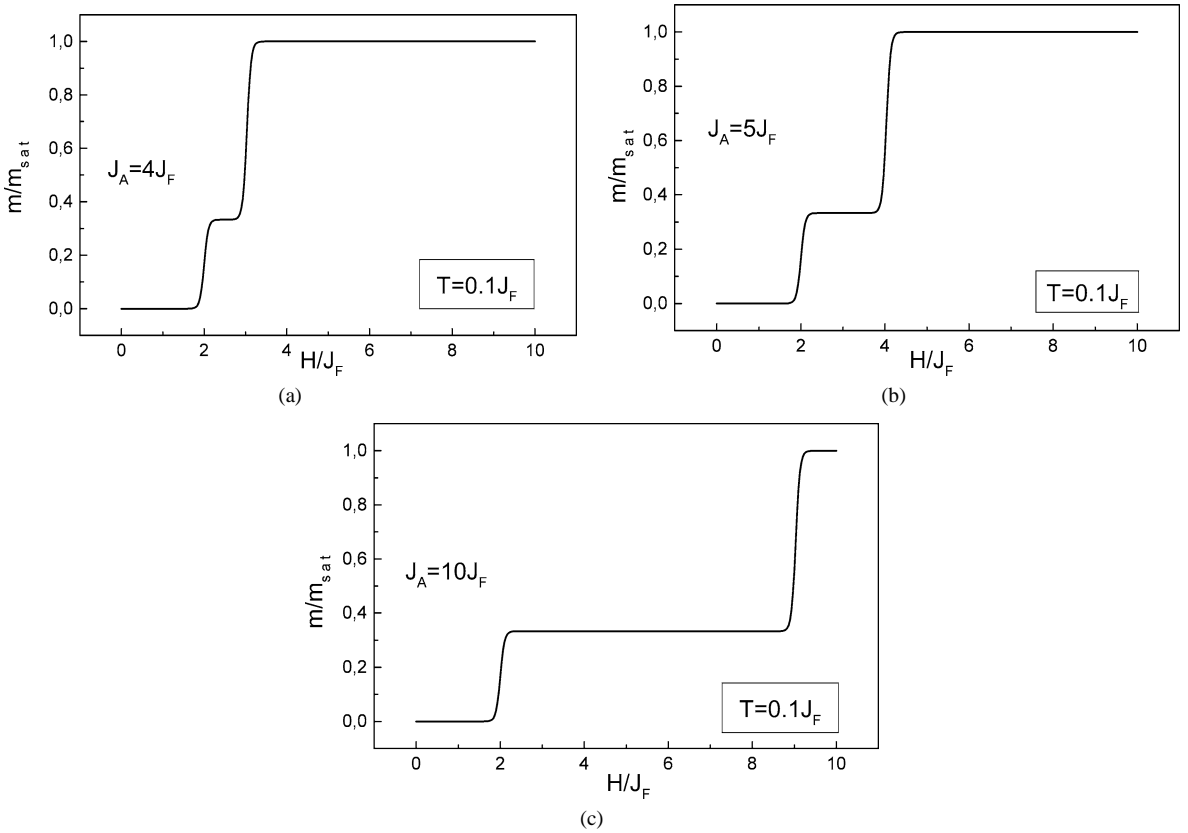
Fig. 3. Magnetization curves in the regime of strong antiferromagnetic coupling exhibiting the magnetization plateau at $m/m_{\mathrm{sat}} = 1/3$ for $\kappa = 4$ (a), $\kappa = 5$ (b) and $\kappa = 10$ at $T = 0.1 J_F$. The values of critical field magnitude, determining the width of plateau are $H_{c_1} = 2J_F$ and $H_{c_2} = J_A - J_F$.

curve. In Fig. 4(b) one can see the first signs of forthcoming plateaus at $m = 0$ and $m/m_{\mathrm{sat}} = 1/3$ and the complete formation of these plateaus at $T = 0.1 J_F$ (Fig. 4(c)). Fig. 4(c) also clearly indicates that the values of critical fields are just slightly different from H_{c_1} and H_{c_2} calculated above. And finally the curve, presented in Fig. 4(d), obtained for extremely low temperature $T = 0.001 J_F$ shows step-like behaviour which is typical for $T = 0$. In this case, one can see that the jumps in the course of the magnetization exactly coincide with $H = 2J_F$ and $H = J_A - J_F$ that is in full agreement with the pattern of transition from $\langle 3 \rangle$ state to $\langle 3111 \rangle$ state with further transition to the saturated state. Nevertheless, it is necessary to emphasize that there are no real jumps at any finite temperatures at all, because the system under consideration is one-dimensional and it is know that there are no phase transition in classical systems in $d = 1$. So, the results presented in Fig. 4(d) should be regarded not as jumps of magnetization but as extremely steep rises of the $m(H)$ function in the vicinity of critical values of H.

4. Conclusion

The main aim being pursued in this Letter is to establish the formation of magnetization plateaus in the classical one-dimensional spin systems with Ising interaction. As an example we chose the classical counterpart of the ferromagnetic–ferromagnetic–antiferromagnetic trimerized Heisenberg chain, considered by Hida [6]. The
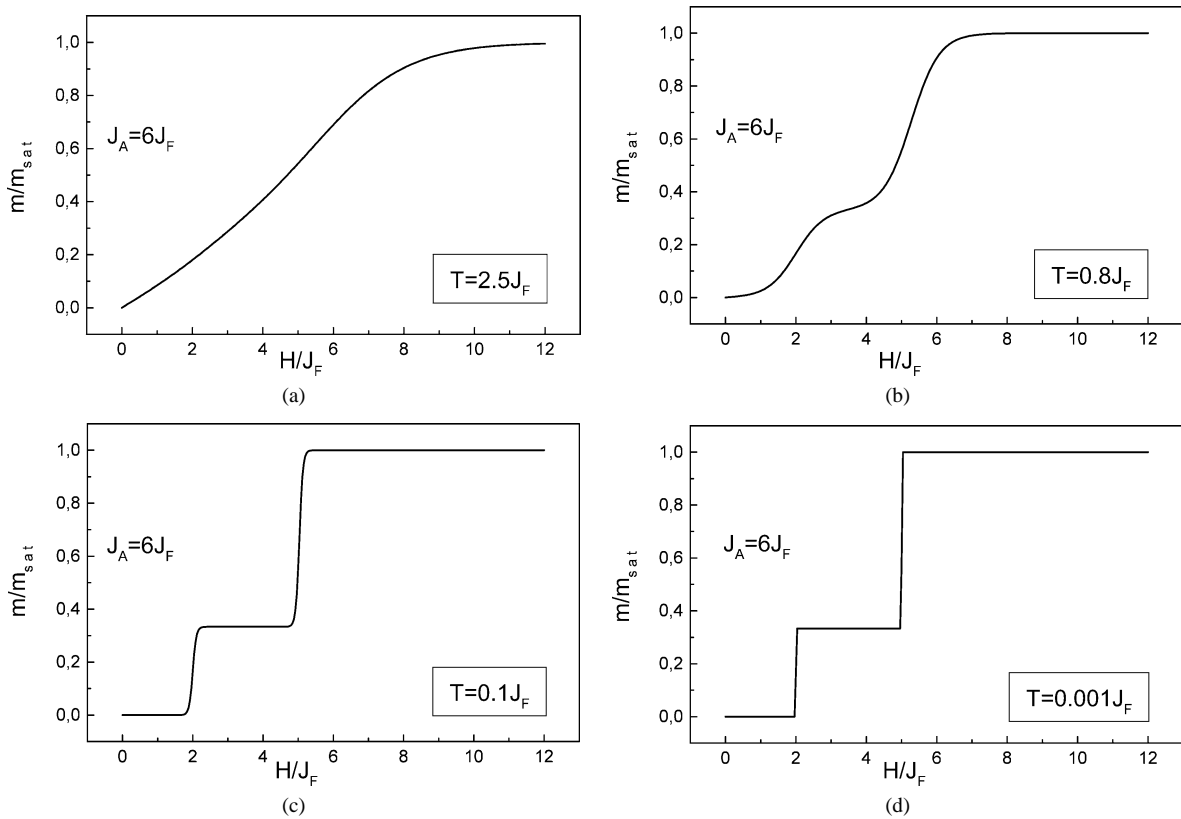
Fig. 4. The magnetization curves for $\kappa = 6$ at different temperatures: (a) $T = 2.5J_F$; (b) $T = 0.8J_F$; (c) $T = 0.1J_F$; (d) $T = 0.001J_F$.

approach developed by Hida was based on a numerical diagonalization of finite size system and allowed him to explain the low temperature magnetization data for the compound $3CuCl_2 \cdot 2dx$ as well as to predict the appearance of a magnetization plateau at $m/m_{sat} = 1/3$ when the ferromagnetic exchange energy is comparable to or smaller than the antiferromagnetic exchange energy. Using entirely analytic technique of the transfer matrix for corresponding system of Ising spins we have obtained the exact explicit expression for magnetization function. Despite the extremal complexity of the expression, nevertheless it permits to draw the plots of magnetization processes for arbitrary finite temperature and couplings. Obtained plots completely reproduce the result of Hida, moreover in our model the exact condition of existence of plateau $J_A \geqslant 3J_F$ is derived. It is also possible using our approach to calculate the width of the plateau at $m/m_{sat} = 1/3$. The latter is equal to $J_A - 3J_F$ in the limit $T \to 0$.

The phenomenon of magnetization plateaus are often regarded to have purely quantum origin, however it is obviously not always true since there are some two-dimensional classical spin models which exhibit a magnetization plateaus at zero temperature. For instance, in Ref. [12] Kubo and Momoi investigated the Multiple Spin Exchange (MSE) model on the triangular lattice in the classical limit and have found a large range of parameters where magnetization plateaus at $m/m_{sat} = 1/3$ and $m/m_{sat} = 1/2$ occur at zero temperature. There are also results on the two-dimensional Ising models [16] as well as on the calculations on recurrent lattice [20]. However, for one-dimensional systems these results are obtained for the first time.

Moreover, these allow to suppose that there exists a class of one-dimensional spin systems which being both considered in terms of Heisenberg and Ising spins leads to the qualitatively same structures of the magnetization profiles, particularly to the formation of magnetization plateaus. In contrast to the quantum systems the plateau in

our model has very simple nature associated with the stability of spatially modulated structure, so-called $\langle 3111 \rangle$ configuration of spins which we proved to exists at $T = 0$. Apparently, for finite but sufficiently low temperatures the plateau is still caused by the same $\langle 3111 \rangle$ structure, though the other configurations are also possible, for instance so-called *uud* phase which is responsible for plateau at $m/m_{\mathrm{sat}} = 1/3$ in triangular antiferromagnets and MSE model.

In conclusion note, that the approach based on Ising spins is remarkable for its simplicity and clarity. It does not require any numerical calculations in contrast to the majority of methods, which are used in this field of research, and can serve as a very useful supplement to the generally applicable techniques such as the numerical diagonalizations of finite clusters, quantum Monte Carlo calculations, series expansions and bosonization technique.

Acknowledgements

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