Frustrated ferromagnetic spin-$\frac{1}{2}$ chain in a magnetic field

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We study the ground state properties of the Heisenberg spin-$\frac{1}{2}$ chain with ferromagnetic nearest-neighbor and antiferromagnetic next-nearest-neighbor interactions using two approximate methods. One of them is the Jordan-Wigner mean-field theory and another approach is based on the transformation of spin operators to Bose ones and on the variational treatment of bosonic Hamiltonian. Both approaches give close results for the ground state energy and the magnetization curve at $T=0$. It is proved that quantum fluctuations change the classical critical exponents in the vicinity of the transition point from the ferromagnetic to the singlet ground state. The magnetization processes display a different behavior in the regions near and far from the transition point. The relation of the obtained results to the experimental magnetization curve in $Rb_2Cu_2Mo_3O_{12}$ is discussed.

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I. INTRODUCTION

Lately, there has been considerable interest in low-dimensional quantum spin systems that exhibit frustration. The spin-$\frac{1}{2}$ chain with next-nearest-neighbor interactions (which is equivalent to a zigzag ladder) is a typical model with the frustration. The Hamiltonian of the model is given by

$$H=\sum_{n=1}^{N}(J_1\mathbf{S}_n\cdot\mathbf{S}_{n+1}+J_2\mathbf{S}_n\cdot\mathbf{S}_{n+2}), \tag{1}$$

where S is the spin-$\frac{1}{2}$ operator, J_1 and J_2 denote the nearest- (NN) and the next-nearest-neighbor (NNN) interactions.

This model with both NN and NNN antiferromagnetic interactions (J_1, $J_2>0$) is well studied.[1–6] There is a critical value $J_{2c}=0.2411J_1$, which separates doubly degenerated dimer phase (at $J_2>J_{2c}$) characterized by the excitation gap and the gapless spin-fluid phase (at $J_2<J_{2c}$). Relatively less is known about model (1) with the ferromagnetic NN and the antiferromagnetic NNN interactions ($J_1<0$, $J_2>0$). Though the latter model has been a subject of many studies[4,7–10] the complete picture of the phases of this model remains unclear up to now. It is known that the ground state is ferromagnetic for $J_2<|J_1|/4$. At $J_2=|J_1|/4$ the ferromagnetic state is degenerated with the singlet state. The wave function of the singlet state at $J_2=|J_1|/4$ is known exactly.[11,12] For $J_2>|J_1|/4$ the ground state is an incommensurate singlet. Controversial conclusions exist about the presence of a gap at $J_2>|J_1|/4$. It has been long believed that the model is gapless[6,13] but the one-loop renormalization group analysis indicates[9,14] that the gap is open due to a Lorentz symmetry-breaking perturbation. However, existence of the gap has not been verified numerically.[9] On the basis of a field theory consideration it has been proposed[15] that a very tiny but finite gap exists which cannot be observed numerically.

Besides a general interest in frustrated quantum spin models there is an additional motivation for the study of model (1) with $J_1<0$. Really, the ferromagnetic NN interaction is expected to exist in compounds containing CuO chains with edge-sharing CuO_4 units. The Cu-O-Cu angle in these compounds is close to 90°, and usual antiferromagnetic NN exchange between Cu ion spins is suppressed. This means that the sign of J_1 can be negative, while the NNN exchange is antiferromagnetic. Several compounds with edge-sharing chains are known,[16] such as Li_2CuO_2, $La_6Ca_8Cu_{21}O_{41}$, and $Ca_2Y_2Cu_5O_{10}$. However, in these compounds the antiferromagnetic long range order appears at low temperatures due to small interchain interactions. Recently, the crystal $Rb_2Cu_2Mo_3O_{12}$ with edge-sharing chains has been synthesized and studied experimentally.[17,18] Remarkably, no magnetic phase transition appears down to 2 K, which testifies that interchain interaction is very small in this compound. Therefore, $Rb_2Cu_2Mo_3O_{12}$ can be considered as an ideal model compound with the ferromagnetic (NN) interaction. According to Ref. 17 it is described by the Hamiltonian (1) with $J_1\simeq-140$ K and $J_2/|J_1|\simeq0.4$.

One of the interesting peculiarities of model (1) is the dependence of the magnetization on the applied magnetic field at $T=0$. The magnetization curve for $J_1<0$ is quite different from that for the case $J_1>0$. It is characterized by a rapid increase (or even discontinuity) in the magnetization if the external field exceeds a critical value. It is expected that the magnetization displays a true jump (the metamagnetic transition) when the NNN interaction J_2 is slightly larger than $|J_1|/4$.[19] This conclusion has been made on the basis of the exact diagonalization calculations for finite chains. However, the extrapolation of these results to the thermodynamic limit is rather difficult due to strong nonmonotonic finite-size effects.

In this paper the ground state energy and the magnetization processes in model (1) with $J_1<0$ are studied using two variational approaches. One of these approaches is based on the Jordan-Wigner transformation of spin-$\frac{1}{2}$ operators to the Fermi ones with the subsequent mean-field treatment of the Fermi Hamiltonian. Another variational approach is applied to the Bose Hamiltonian arising from a special transformation of spin operators to Bose ones.

The paper is organized as follows. In Sec. II the details of the Jordan-Wigner mean-field approximation are presented. The zero-temperature magnetization process is studied and the form of the magnetization curve in different regions of the parameter $J_2/|J_1|$ is found. For the case $h=0$ we have focused on the behavior of the model in the vicinity of the transition point from the ferromagnetic to the singlet ground state. The critical exponents characterizing this behavior are determined. In Sec. III we describe the variational method for the treatment of the considered Hamiltonian rewritten in bosonic form. Scaling estimates of the critical exponent near the transition point are presented in Sec. IV. In Sec. V we summarize our results.

II. JORDAN-WIGNER MEAN-FIELD APPROACH

It is convenient to represent the Hamiltonian of the spin-$\frac{1}{2}$ chain with the ferromagnetic NN and the antiferromagnetic NNN interactions in the form

$$H=-\sum_{n=1}^{N}\left(\mathbf{S}_n\cdot\mathbf{S}_{n+1}-\frac{1}{4}\right)+\alpha\sum_{n=1}^{N}\left(\mathbf{S}_n\cdot\mathbf{S}_{n+2}-\frac{1}{4}\right)-h\sum_{n=1}^{N}S_n^y, \tag{2}$$

where $\alpha=J_2/|J_1|$ and $h=g\mu_B B/|J_1|$ is the effective dimensionless magnetic field. The constant shifts in Eq. (2) secure the energy of the fully polarized state to be zero.

Let us start from the classical picture of the ground state of Eq. (2). In the classical approximation the spins are vectors which form the spiral structure in the xz plane with a pitch angle φ between neighboring spins and all spin vectors are inclined towards the y axis by an angle θ. The classical energy per site,

$$\epsilon(\varphi,\theta)=\frac{1}{4}[1-\cos\varphi-\alpha(1-\cos(2\varphi))]\cos^2\theta-\frac{h}{2}\sin\theta, \tag{3}$$

is minimized by the angles

$$\varphi_{cl}=\cos^{-1}\frac{1}{4\alpha},\quad \theta_{cl}=\sin^{-1}\frac{\alpha h}{2\gamma^2}, \tag{4}$$

where $\gamma=\alpha-\frac{1}{4}$.

The classical ground state energy is

$$\frac{E_{cl}}{N}=-\frac{1}{2\alpha}\gamma^2-\frac{\alpha h^2}{8\gamma^2}. \tag{5}$$

Following this picture we transform local axes on nth site by a rotation about the y axis by φn and then by a rotation about the x axis by θ. The transformation to new spin-$\frac{1}{2}$ operators η_n has a form

$$\mathbf{S}_n=R_y(\varphi n)R_x(\theta)\eta_n, \tag{6}$$

where $R_y(\varphi n)$ and $R_x(\theta)$ are the operators of the corresponding rotations.

Substituting (6) into (2) we obtain the transformed Hamiltonian in terms of the η operators,

$$H=H_1+H_2+H_3,$$

$$\begin{aligned}H_1=&N\epsilon(\varphi,\theta)\\&+\sum_{n=1}^{N}\left[J_{1x}\eta_n^x\eta_{n+1}^x+J_{1y}\eta_n^y\eta_{n+1}^y+J_{1z}\left(\eta_n^z\eta_{n+1}^z-\frac{1}{4}\right)\right]\\&+\sum_{n=1}^{N}\left[J_{2x}\eta_n^x\eta_{n+2}^x+J_{2y}\eta_n^y\eta_{n+2}^y+J_{2z}\left(\eta_n^z\eta_{n+2}^z-\frac{1}{4}\right)\right]\\&-h\sin\theta\sum_{n=1}^{N}\left(\eta_n^z-\frac{1}{2}\right),\end{aligned}$$

$$\begin{aligned}H_2=\sin\theta\sum_{n=1}^{N}&[\sin\varphi(\eta_n^x\eta_{n+1}^y-\eta_n^y\eta_{n+1}^x)\\&-\alpha\sin2\varphi(\eta_n^x\eta_{n+2}^y-\eta_n^y\eta_{n+2}^x)],\end{aligned}$$

$$\begin{aligned}H_3=-\sum_{n=1}^{N}&\left[\frac{\sin2\theta}{2}(1-\cos\varphi)(\eta_n^z\eta_{n+1}^y+\eta_n^y\eta_{n+1}^z)\right.\\&-\alpha\frac{\sin2\theta}{2}(1-\cos2\varphi)(\eta_n^z\eta_{n+2}^y+\eta_n^y\eta_{n+2}^z)\\&+\sin\varphi\cos\theta(\eta_n^x\eta_{n+1}^z-\eta_n^z\eta_{n+1}^x)\\&\left.-\alpha\sin2\varphi\cos\theta(\eta_n^x\eta_{n+2}^z-\eta_n^z\eta_{n+2}^x)\right]-h\cos\theta\sum_{n=1}^{N}\eta_n^y,\end{aligned} \tag{7}$$

where

$$J_{1x}=-\cos\varphi,\ J_{1y}=-\cos\varphi\sin^2\theta-\cos^2\theta,\quad J_{1z}=-\cos\varphi\cos^2\theta-\sin^2\theta$$

$$J_{2x}=\alpha\cos2\varphi,\quad J_{2y}=\alpha(\cos2\varphi\sin^2\theta+\cos^2\theta),\quad J_{2z}=\alpha(\cos\varphi\cos^2\theta+\sin^2\theta). \tag{8}$$

The Hamiltonian (7) for $h=0$ ($\theta=0$) has been considered before in Refs. 4 and 10. In Ref. 10 the ground state in the vicinity of the point $\alpha=\frac{1}{4}$ has been studied on the basis of the spin-wave theory taking φ by its classical value. It was shown that the transition at $\alpha=\frac{1}{4}$ from the ferromagnetic state to the singlet one is of the second order and the ground state energy is $-4\gamma^2$ for $0<\gamma\ll1$. In Ref. 4 the dependence of the ground state energy and the pitch angle φ on α has been found using the coupled cluster method. In particular, the ground state energy is proportional to γ^2 for $0<\gamma\ll1$ too. However, both cited approaches are not variational ones. Besides, the magnetization curve has not been studied.

Our primary interest is how quantum effects alter the classical ground state structure. In this section we use the Jordan-Wigner transformation which converts the spin Hamiltonian (7) into a model of interacting spinless fermions

$$\eta_n^+=c_n\exp\left(i\pi\sum_{j=1}^{n-1}c_j^+c_j\right),\quad \eta_n^z=\frac{1}{2}-c_n^+c_n. \tag{9}$$

The Hamiltonians H_1 and H_2 in Eq. (7) are transformed to Fermi Hamiltonians having the following forms:

$$H_{1f}=N\epsilon(\varphi,\theta)+(h\sin\theta-J_{1z}-J_{2z})\sum_{n=1}^{N}c_n^+c_n+J_{1z}\sum_{n=1}^{N}c_n^+c_nc_{n+1}^+c_{n+1}+J_{2z}\sum_{n=1}^{N}c_n^+c_nc_{n+2}^+c_{n+2}+\frac{1}{4}(J_{1x}+J_{1y})\sum_{n=1}^{N}(c_n^+c_{n+1}+c_{n+1}^+c_n)+\frac{1}{4}(J_{2x}+J_{2y})\times\sum_{n=1}^{N}(c_n^+c_{n+2}+c_{n+2}^+c_n)(1-2c_{n+1}^+c_{n+1})+\frac{1}{4}(J_{1x}-J_{1y})\times\sum_{n=1}^{N}(c_n^+c_{n+1}^++c_{n+1}c_n)+\frac{1}{4}(J_{2x}-J_{2y})\times\sum_{n=1}^{N}(c_n^+c_{n+2}^++c_{n+2}c_n)(1-2c_{n+1}^+c_{n+1}),$$

$$H_{2f}=\frac{i}{2}\sin\theta\sum_{n=1}^{N}[\sin\varphi(c_n^+c_{n+1}-c_{n+1}^+c_n)-\alpha\sin2\varphi(c_n^+c_{n+2}-c_{n+2}^+c_n)(1-2c_{n+1}^+c_{n+1})]. \quad (10)$$

We do not present here the Hamiltonian H_{3f} because it has a very complicated form containing nonlocal interaction and, as will be shown below, does not contribute to the energy in the mean-field approach.

The next step of the approach is to treat the Hamiltonian $H_f=H_{1f}+H_{2f}+H_{3f}$ in the mean-field approximation (MFA). We use the approximation scheme proposed in Ref. 20. The ground state wave function in this approximation has a BCS-like form,

$$|\Psi\rangle=\prod_{k>0}(\cos\phi_k+\sin\phi_kc_k^+c_{-k}^+)|0\rangle,$$

where c_k^+ is Fourier transform of the Fermi operators c_n^+.

The expectation value of H_{2f} over the function Ψ is $\langle H_{2f}\rangle=0$ due to the fact that $\Sigma_k\sin k\langle c_k^+c_k\rangle=0$ and $\Sigma_k\cos k\langle c_k^+c_{-k}^+\rangle=0$. The Hamiltonian H_{3f} includes an odd number of the Fermi operators and, therefore, it is zero in the MFA $\langle H_{3f}\rangle=0$ as well. Thus, both Hamiltonians H_{2f} and H_{3f} do not contribute to the energy in this approximation.

In the MFA, the four fermions terms in Eqs. (10) are decoupled in all possible ways. After Fourier transformation, the mean-field Hamiltonian takes the form

$$H_{\mathrm{MFA}}=NC+\sum_{k>0}a(k)(c_k^+c_k+c_{-k}^+c_{-k})+\sum_{k>0}b(k)(c_k^+c_{-k}^++c_{-k}c_k), \quad (11)$$

where

$$C=\epsilon(\varphi,\theta)-(J_{1z}+J_{2z})n^2+J_{1z}(t_1^2-g_1^2)+J_{2z}(t_2^2-g_2^2)+(J_{2x}+J_{2y})(nt_2-t_1^2-g_1^2)+(J_{2x}-J_{2y})(ng_2-2t_1g_1), \quad (12)$$

$$a(k)=u+\nu_1\cos k+\nu_2\cos2k,$$

$$b(k)=w_1\sin k+w_2\sin2k, \quad (13)$$

where

$$u=-(J_{1z}+J_{2z})(1-2n)-(J_{2x}+J_{2y})t_2-(J_{2x}-J_{2y})g_2+h\sin\theta,$$

$$\nu_1=\frac{1}{2}(J_{1x}+J_{1y})-2J_{1z}t_1+2(J_{2x}+J_{2y})t_1+2(J_{2x}-J_{2y})g_1,$$

$$\nu_2=(J_{2x}+J_{2y})\left(\frac{1}{2}-n\right)-2J_{2z}t_2,$$

$$w_1=\frac{1}{2}(J_{1x}-J_{1y})+2(J_{1z}+J_{2x}+J_{2y})g_1+2(J_{2x}-J_{2y})t_1,$$

$$w_2=(J_{2x}-J_{2y})\left(\frac{1}{2}-n\right)+2J_{2z}g_2. \quad (14)$$

The ground state energy, the one-particle excitation spectrum $\varepsilon(k)$, and the magnetization $M=\langle S_n^y\rangle$ are

$$\frac{E}{N}=\epsilon(\varphi,\theta)+F(\varphi,\theta),$$

$$\varepsilon(k)=\sqrt{a^2(k)+b^2(k)},$$

$$M=\left(\frac{1}{2}-n\right)\sin\theta, \quad (15)$$

where

$$F(\varphi,\theta)=(J_{1z}+J_{2z})n(1-n)+J_{1z}(g_1^2-t_1^2)+J_{2z}(g_2^2+t_2^2-2t_2)+\frac{1}{2}(J_{1x}+J_{1y})t_1+\frac{1}{2}(J_{1x}-J_{1y})g_1+(J_{2x}+J_{2y})\times(nt_2-t_1^2-g_1^2)+(J_{2x}-J_{2y})(ng_2-2t_1g_1). \quad (16)$$

Quantities n, t_1, t_2, g_1, and g_2 are the ground state expectation values, which are determined by the following self-consistency equations:

$$n=\langle c_n^+c_n\rangle=\int_0^{\pi}\frac{dk}{2\pi}\left(1-\frac{a(k)}{\varepsilon(k)}\right),$$

$$t_1=\langle c_n^+c_{n+1}\rangle=-\int_0^{\pi}\frac{dk}{2\pi}\frac{a(k)\cos k}{\varepsilon(k)},$$

$$t_2=\langle c_n^+c_{n+2}\rangle=-\int_0^{\pi}\frac{dk}{2\pi}\frac{a(k)\cos2k}{\varepsilon(k)},$$

$$g_1=\langle c_n^+c_{n+1}^+\rangle=-\int_0^{\pi}\frac{dk}{2\pi}\frac{b(k)\sin k}{\varepsilon(k)},$$

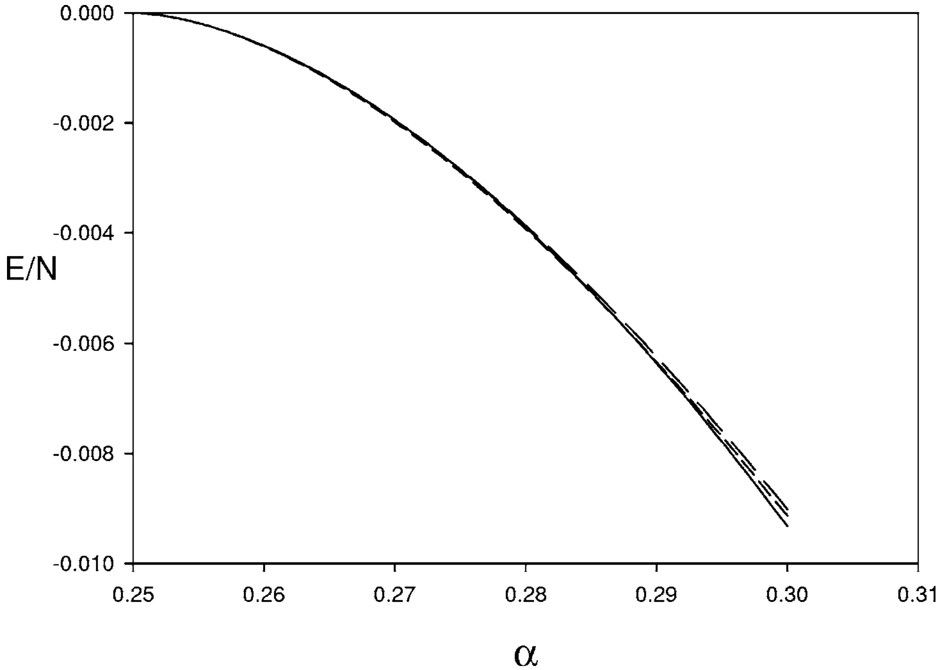

FIG. 1. The dependence of the ground state energy of model (2) on α in the MFA (short-dashed line) and the boson variational approach (long-dashed line). Solid line corresponds to the energy given by Eq. (20).

$$g_2 = \langle c_n^+ c_{n+2}^+ \rangle = -\int_0^\pi \frac{dk}{2\pi} \frac{b(k)\sin 2k}{\varepsilon(k)}. \tag{17}$$

The solution of the self-consistency equations gives the minimum of the ground state energy in a case of "one-particle" wave functions at given angles φ and θ. We treat the angles φ and θ as the variational parameters (not equal to their classical values). Thus, one should minimize the energy with respect to the angles φ and θ, solving the self-consistency equations for each value of φ and θ. This means that the proposed procedure remains a variational one.

To study the effect of the dimerization we added the staggered terms to the NN expectation values

$$\langle c_n^+ c_{n+1} \rangle = t_1 + t_1'(-1)^n,$$

$$\langle c_n^+ c_{n+1}^+ \rangle = g_1 + g_1'(-1)^n,$$

which leads to a more complicated form of the Hamiltonian H_{MFA} and two additional self-consistency equations for t_1' and g_1'. But the solution of the self-consistency equations with the minimization of the energy over φ and θ gives $t_1' = g_1' = 0$. This means that the MFA does not indicate the dimerization in the system.

A. Ground state energy near the transition point $\alpha = \frac{1}{4}$

At first we consider the Hamiltonian (11) at $h=0$ when $\theta=0$ and we are interested mainly in the behavior of the model in the vicinity of the point $\alpha=\frac{1}{4}$, where the transition from the ferromagnetic to the singlet ground state occurs. At $h=0$ the ground state energy per site is

$$\frac{E}{N} = \epsilon(\varphi,0) + F(\varphi,0), \tag{18}$$

where $F(\varphi,0)$ is a quantum correction to the classical part of the energy.

The analysis of the solution of Eqs. (17) shows that in the case $0<\gamma\ll 1$ and $\varphi \ll 1$ the functions $\epsilon(\varphi,0)$ and $F(\varphi,0)$ have forms

$$\epsilon(\varphi,0) = -\frac{\gamma\varphi^2}{2} + \frac{\varphi^4}{32},$$

$$F(\varphi,0) = -\frac{\varphi^4}{32} + A\varphi^{24/5}. \tag{19}$$

The coefficient $A \approx 0.0195$ is determined by the numerical solution of Eqs. (17).

Substituting (19) into (18) and minimizing E with respect to φ, we obtain the leading terms in γ for the angle $\varphi(\gamma)$ and the energy $E(\gamma)$,

$$\varphi = 2.331\gamma^{5/14},$$

$$\frac{E}{N} = -1.585\gamma^{12/7}. \tag{20}$$

The numerical solution of self-consistency Eqs. (17) confirms this dependence $E(\gamma)$ at $0<\gamma\ll 1$ (see Fig. 1).

Since the MFA is the variational approach, the found critical exponent for the ground state energy $\beta=\frac{12}{7}$ gives upper bound for the exact critical exponent $\beta \leq \frac{12}{7}$ and certainly it is less than $\beta=2$ obtained in classical approximation. We note that the use of more elaborate methods[4,10] does not change the exponent $\beta=2$ and change the numerical prefactor a in the ground state energy $E=-a\gamma^2$ only. This means that at present the MFA gives the best estimate of the ground state energy in the region $0<\gamma\ll 1$.

In Ref. 22 the model (2) (at $h=0$) has been studied using the Jordan-Wigner transformation and the mean-field theory. However, the pair correlations of type $\langle c_n^+ c_m^+ \rangle$ have been neglected. It was conjectured in Ref. 22 that the ground state at

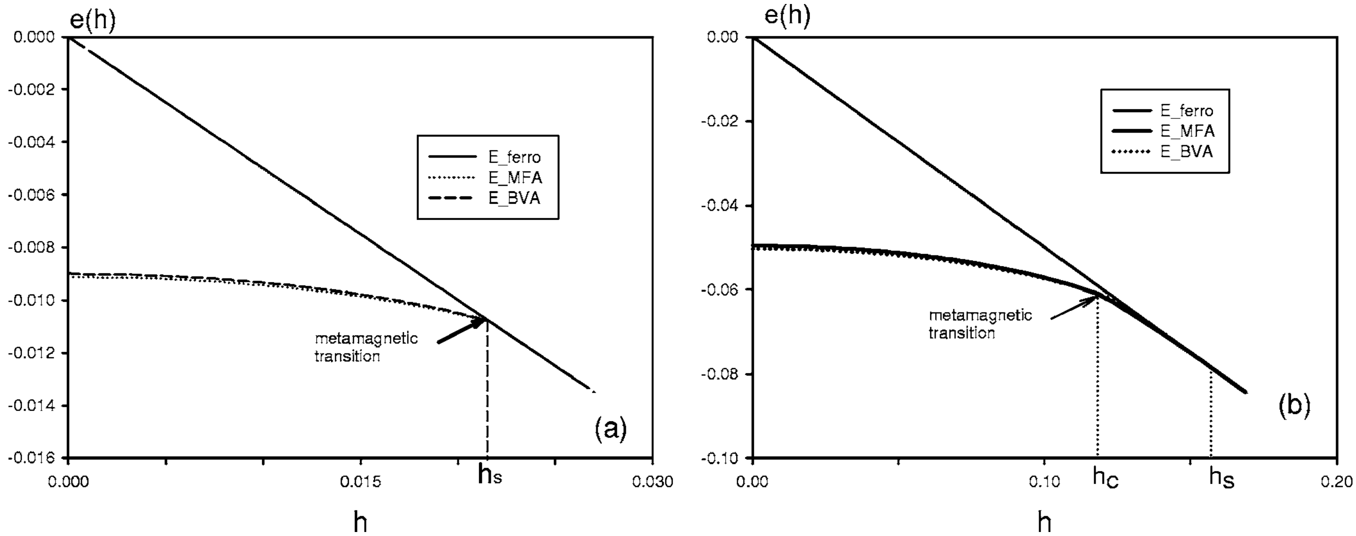

FIG. 2. The dependence of the ground state energy of model (2) on magnetic field $e(h)$ for (a) $\alpha=0.3$ and (b) $\alpha=0.4$.

$\frac{1}{4}<\alpha<0.25854$ is not singlet but has a nonzero total spin. We have found no evidence of this fact and the numerical diagonalization of finite chains does not confirm this prediction as well.

As was written above, in our mean-field treatment the pitch angle φ is considered as the variational parameter. It should be noted that in the MFA a helical (spiral) long range order (LRO) exists as well as in the classical approximation. The helical LRO is characterized by the angle φ, though its value differs from the classical one. Of course, this LRO is destroyed by those quantum fluctuations which are ignored in the MFA. At the same time the obtained helical LRO is regarded to an incommensurate behavior of the correlation function and φ can be identified with the momentum $q_{\max}$ at which the static structure factor takes a maximal value.

B. Magnetization curve

In this section we consider the magnetization processes in model (2). The behavior of the magnetization in the region of the magnetic field close to saturation is of a particular interest. We note that the determination of the saturation field $h_s(\alpha)$ at which the transition to saturation takes place is generally not a simple problem. In model (1) with both antiferromagnetic NN and NNN interactions, this field is equal to the energy of a one-magnon state on the ferromagnetic background (all spins up), i.e., $h_s=-E_1$. It is not the case for model (2). The specific property of this model is the presence of bound magnon states (complexes of two or more flipped spins) arising as a result of magnon-magnon attraction. When the field h is reduced below the saturated value the number of spins flipping simultaneously is more than one. Then the saturated field is determined by the condition

$$h_s = \max\left\{\frac{|E_n|}{n}\right\}, \qquad (21)$$

where E_n is the energy of n-magnon state.

There are two possible scenarios of the behavior of the magnetization curve close to saturation. In the first case, Eq. (21) is satisfied at $n^*\sim o(N)$ and the magnetization is a continuous function at $h\to h_s$. In the second case $n^*\sim N$ and the magnetization jumps at $h=h_s$ from $M^*=\left(\frac{1}{2}-n^*/N\right)$ to $M=\frac{1}{2}$, i.e., the metamagnetic transition occurs. It means that the ground state energy as a function of magnetization $e(M)$ has a negative curvature and M^* is determined by the Maxwell construction. We note that the numerical estimate of h_s and n^* especially at $0<\gamma\ll 1$ is very difficult because the number of spins flipping simultaneously at $M\to\frac{1}{2}$ increases when $\alpha\to\frac{1}{4}$.[9,19]

We calculate the magnetization for model (2) using the MFA at $h>0$. In this case there are two variational parameters, φ and θ. We show the ground state energy per site e as a function of h for two values of α in Fig. 2 together with the energy of the ferromagnetic state $e_F=-h/2$. The saturation field h_s is determined by the crossing between the variational energy $e(h)$ and e_F. The dependence $e(h)$ shown in Fig. 2 demonstrates two different types of the behavior. For $\alpha\lesssim 0.38$ [Fig. 2(a)] $e(h)$ and $e_F(h)$ have different slopes at the crossing point. At $h=h_s$ the magnetization jumps from $M^*=\partial e/\partial h|_{h=h_s}$ to $\frac{1}{2}$ as it is shown in Fig. 3(a). It means that maximum in Eq. (21) is reached for macroscopic n and the metamagnetic transition takes place. The dependence of the saturated field on α is shown in Fig. 4. This dependence is consistent with those obtained in Ref. 19 by using numerical diagonalizations of finite chains. The magnetization curve at $0<\gamma\ll 1$ can be found analytically from the self-consistency Eqs. (17). The energy per site $e(M)$ at $M\ll\frac{1}{2}$ has a form

$$e(M) = -1.585\gamma^{12/7} + 3.69\gamma^{10/7}M^2 + O(\gamma^{12/7}). \qquad (22)$$

Performing the Maxwell construction we obtain

$$M^* = 0.429\gamma^{2/7},$$

$$h_s = 3.17\gamma^{12/7}. \qquad (23)$$

As follows from Eq. (21), h_s in Eq. (23) is the binding energy per magnon of a multimagnon complex. It is interesting to compare this value with the binding energy of a two-

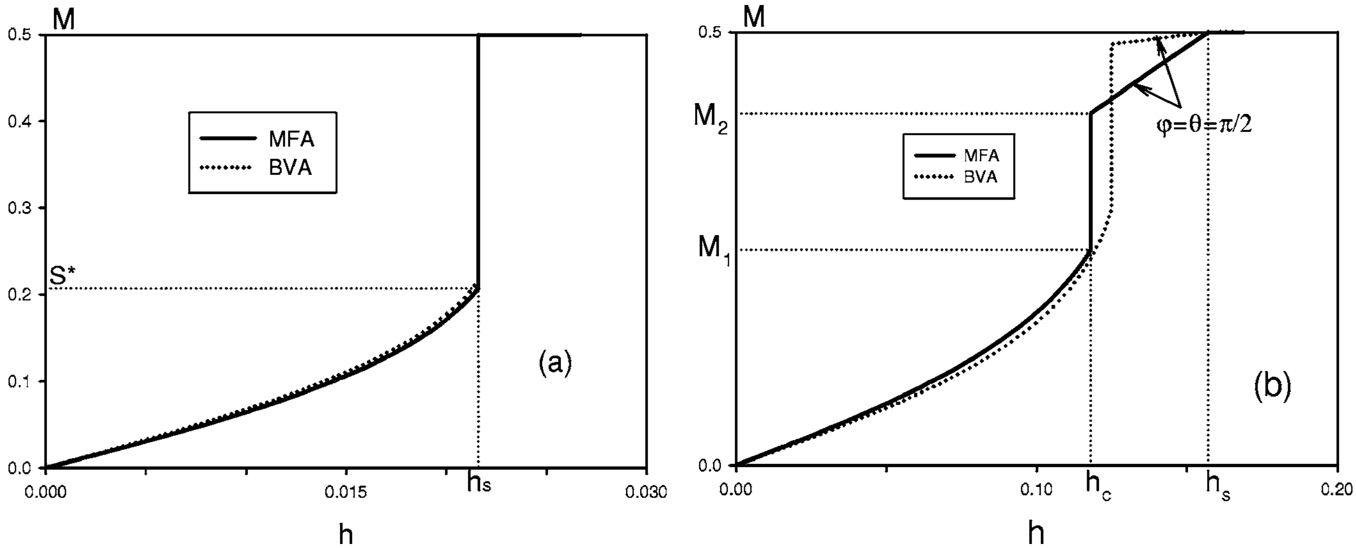

FIG. 3. The dependence of the magnetization of model (2) on magnetic field $M(h)$ for (a) $\alpha=0.3$ and (b) $\alpha=0.4$.

magnon complex which is $-72\gamma^3$ at $0<\gamma\ll 1$.[8,10] It is evident that interaction of a macroscopic number of magnons essentially lowers the binding energy.

The magnetization $M(h)$ at $0<\gamma\ll 1$ is

$$M(h) = 0.136\gamma^{-10/7}h, \quad h < h_s,$$

$$M(h) = \frac{1}{2}, \quad h > h_s. \tag{24}$$

According to Eq. (24) the susceptibility χ is

$$\chi = 0.136\gamma^{-10/7}. \tag{25}$$

As follows from Eq. (24) and as shown on Fig. 3, the magnetization is a linear function of h at $h\to 0$. This testifies to the absence of the gap in the spectrum. As was noted before, the subtle question about the gap in this model is still open. Numerical calculations show $1/N$ behavior for a gap,[9] while the one-loop renormalization group indicates so tiny an exponentially small gap[15] that it cannot be observed numerically. The proposed version of the MFA does not predict the dimerization and the gap for model (2). However, we are not sure that the mean-field approach is an adequate tool to answer so subtle a question.

For $\alpha \gtrsim 0.38$ the curve $e(h)$ [Fig. 2(b)] has a cusp at some critical magnetic field h_c as a result of the jump of variational parameters minimizing energy from $\varphi,\theta<\pi/2$ to $\varphi=\theta=\pi/2$. At $h=h_c$ the magnetization jumps from $M_1 = -\partial e(h)/\partial h|_{h=h_c-}$ to $M_2 = -\partial e(h)/\partial h|_{h=h_c+}$. According to Eq. (8), $e(h)$ at $h>h_c$ ($\varphi=\theta=\pi/2$) is the ground state energy of the Hamiltonian

$$H = -\sum_{n=1}^{N}\left(S_n^zS_{n+1}^z - \frac{1}{4}\right) + \alpha\sum_{n=1}^{N}\left(\mathbf{S}_n\mathbf{S}_{n+2} - \frac{1}{4}\right) - h\sum_{n=1}^{N}S_n^z. \tag{26}$$

The curve $e(h)$ in Fig. 2(b) is tangent to $e_F(h)$ at the crossing point. The saturation field h_s and the magnetization $M(h)$ near h_s are determined from the self-consistency Eqs. (17) with $\varphi=\theta=\pi/2$,

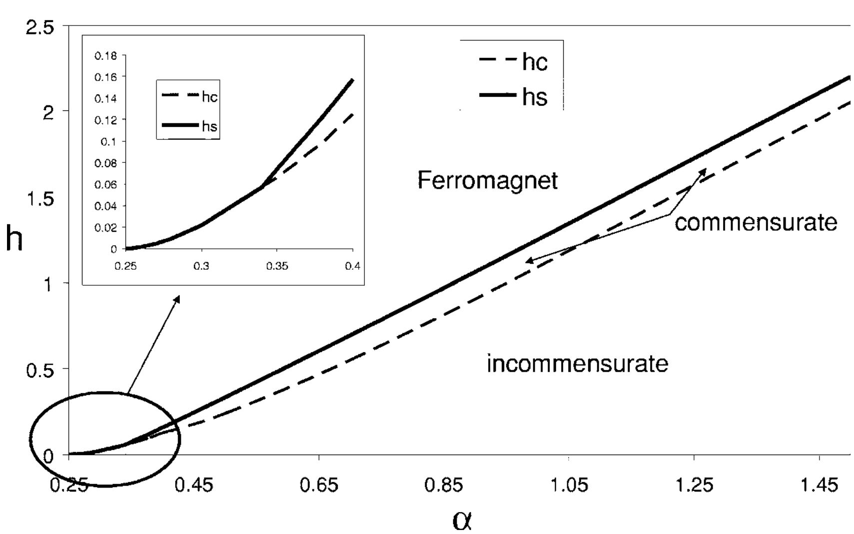

FIG. 4. The phase diagram of model (2) in the MFA in the (h,α) plane.

$$h_s = -\frac{E_2}{2},$$

$$M = \frac{1}{2} - \frac{2(1+\alpha)^4}{(1+2\alpha)(4\alpha^4+15\alpha^3+11\alpha^2-1)}(h_s - h), \tag{27}$$

where

$$E_2 = -\frac{4\alpha^2+2\alpha-1}{1+\alpha}. \tag{28}$$

It is interesting to note that E_2 given by this equation is the exact energy of the bound two-magnon state with a momentum $q=\pi$ of the Hamiltonian (26). The wave function of this state has a form

$$|\psi\rangle = \sqrt{\frac{(1-e^{-2/\xi})}{N}}\sum_{n,l}(-1)^{n+l}e^{-l/\xi}S_n^- S_{n+2l+1}^- |\uparrow\uparrow\uparrow \ldots \uparrow\rangle, \tag{29}$$

with a correlation length

$$\xi^{-1} = \ln\frac{1+\alpha}{\alpha}. \tag{30}$$

Remarkably, this function is the exact one of the two-magnon state of the Hamiltonian (2) as well because

$$\sum (S_n^x S_{n+1}^x + S_n^y S_{n+1}^y)|\psi\rangle = 0. \tag{31}$$

The state (29) has the lowest energy in the two-magnon sector of the Hamiltonian (2) at $\alpha > 0.38$.[8]

Thus, the maximum in Eq. (21) is reached at $n=2$ if $e(h)$ has the form similar to that shown in Fig. 2(b). This fact confirms the observation based on finite-size results[9] that in the region of α rather far from $\alpha=\frac{1}{4}$ only the flipping of pairs of spins participate in the magnetization process near the saturation.

The dependence $M(h)$ for two typical cases is shown in Fig. 3. At $\alpha > 0.38$ the MFA gives the saturation field exactly, though the linear behavior of $M(h)$ in Eq. (27) is in contrast with the expected square-root critical dependence.

It was determined above that the presence of the cusp in the dependence $e(h)$ is the result of the jump of variational parameters. At $h < h_c$ the angle φ is incommensurate while at $h > h_c$ it corresponds to the commensurate phase. Therefore, the magnetic field h_c can be identified with the critical field at which the incommensurate-commensurate transition takes place. The phase diagram of model (2) in the MFA is shown in Fig. 4. The commensurate phase lies between the incommensurate and the ferromagnetic phases.

In the limit $\alpha \to \infty$ the Hamiltonian (2) describes two independent Heisenberg chains. The numerical solution of the MFA equations indicates that in this limit the width of the commensurate phase shrinks as $1/\alpha$. In the limit $\alpha \to \infty$ the pitch angle φ tends to $\pi/2$ for all values of h, while the canted angle θ changes from $\theta=0$ at $h=0$ to $\theta=\pi/2$ at $h=h_s$. This means that the MFA correctly describes the limit $\alpha \to \infty$, showing the incommensurate phase at $0 < h < h_s$.

The MFA shows the jump in magnetization at some critical field h_c. We are not sure whether a true magnetization curve has (or not) to jump. However, we believe that the jump obtained in our approximation testifies to the singular behavior of the magnetization at some critical field. A plausible reason for such singularity consists in the following physical picture. As was shown above at $\alpha > 0.38$ two magnons attract one another and form the bound two-magnon state (29) with the correlation length ξ (30). When the magnetic field is slightly lower than the saturated field h_s and the number of magnons is small, the ground state can be represented with high accuracy as a product of the bound magnon pairs, weakly interacted with each other. The bound pairs of magnons start to feel each other when the mean distance between bound pairs becomes of the order of the correlation length of the pair ξ. This happens at some critical magnon density $n_c \sim \xi^{-1}$. For larger density $n > n_c$ one cannot consider magnon pairs as independent quasiparticles and the above picture is destroyed. Certainly, these speculations can be well justified in the case of $\alpha \gg 1$ only, when the critical density is small, $n_c \sim 1/\alpha$.

Possible singularity in the magnetization is related to the magnetization curve in $Rb_2Cu_2Mo_3O_{12}$. The experimental magnetization at $T=2.6$ K demonstrates a sufficiently sharp increase up to $M \simeq 0.4$ at $B \simeq 14$ T followed by a gradual increase to the saturation. In light of the above, one can assume that the sharp change in the behavior of $M(h)$ at $B \simeq 14$ T is connected with the transition from the incommensurate regime to the commensurate one. Taking the values of J_1 and α estimated for $Rb_2Cu_2Mo_3O_{12}$ ($\alpha \simeq 0.4$, $J_1 \simeq -140$ K)[17] we obtain $B_c = 13.6$ T.

III. BOSON VARIATIONAL APPROACH

Another technique which can be used for an approximate analysis of model (2) is the boson variational approach (BVA) proposed in Ref. 23. This approach is based on the Agranovich-Toshich boson representation of spin $s=\frac{1}{2}$ operators[24]

$$S_n^z = -\frac{e^{i\pi b_n^+ b_n}}{2},$$

$$S_n^- = \frac{1-e^{i\pi b_n^+ b_n}}{2\sqrt{b_n^+ b_n}} b_n^+,$$

$$S_n^+ = b_n \frac{1-e^{i\pi b_n^+ b_n}}{2\sqrt{b_n^+ b_n}}, \tag{32}$$

where b_n^+ are the Bose operators. This transformation preserves all commutation relations for the spin operators and does not involve "nonphysical states," because all states with an odd number of bosons correspond to the same spin state $|\downarrow\rangle$ and the states with an even number of bosons correspond to the spin state $|\uparrow\rangle$. The latter implies that the transformation (32) is not unitary and each energy level of spin model corresponds to an infinite degenerated level of boson Hamiltonian. However, this transformation allows an estimate of the ground state energy of the spin model.

The Hamiltonian of the considered spin model rewritten in the bosonic form is treated by the variational function in the form[23]

$$|\Psi\rangle = \exp\left(\frac{1}{2}\sum_{i,n}\Lambda(n)b_i^+b_{i+n}^+\right)|0_b\rangle, \tag{33}$$

where $|0_b\rangle$ is the boson vacuum state corresponding to the ferromagnetic spin state with all spins pointing up and the function $\Lambda(n)$ is chosen by the condition of minimum of the total energy. Therefore, in contrast to spin-wave theory, this approach is variational. The procedure of calculation of the variational energy with the wave function (33) and energy minimization over function $\Lambda(n)$ was developed in Ref. 23, where the approach was successfully applied to construction of the ground state phase diagram of the frustrated two-dimensional (2D) Heisenberg model.

We have applied the above approach to the rotated spin Hamiltonian (7) and the rotation angles φ and θ were used as variational parameters, as in the MFA. The contribution to the energy of the parts H_2 and H_3 of the Hamiltonian (7) in this approach is also zero as in the MFA. We do not present here cumbersome details of calculations, because they are identical to those in Ref. 23, and give only the final results.

The approach shows very similar behavior of the ground state energy as in the Jordan-Wigner MFA. That is, for small $0<\gamma\ll 1$ both approaches give the same critical exponent for the ground state energy and for the parameter φ. Moreover, the numerical estimates of the ground state energy in both approaches are also very close (see Fig. 1). The behavior of the magnetization curve in the BVA is also very similar to that in the MFA (see Fig. 3). The perfect coincidence of physical properties of the model predicted by the MFA and the BVA looks somewhat surprising because of different nature of these approaches.

We note that the potential advantage of the boson variational approach consists in its applicability to any dimension lattices and possible modification to any spin value S.

IV. SCALING ESTIMATE OF THE CRITICAL EXPONENT NEAR THE TRANSITION POINT $\alpha=\frac{1}{4}$

As was shown in previous sections, the MFA and the BVA give the ground state energy $E(\gamma)\sim\gamma^{12/7}$ near the transition point $\alpha=\frac{1}{4}$. Since both approaches used are variational, the value $\beta=\frac{12}{7}$ is the upper bound for the critical exponent β. This implies an important and strict fact that quantum fluctuations definitely change the classical critical exponent $\beta=2$.

Nevertheless, there are some reasons to believe that the true value of the critical exponent is $\beta=\frac{5}{3}$. The arguments in favor of this conjecture are based on the following consideration with use of the results of numerical calculations of finite-size chains.[7,21,25]

The ground state of model (2) is the singlet at $\alpha>\frac{1}{4}$ for any even N. For a cyclic chain there are a number of level crossings of the two lowest singlets with momenta $q=0$ and $q=\pi$. For example, for $N=4k$ there are $k-1$ crossing points $\frac{1}{4}<\alpha_1(N)<\alpha_2(N)<\cdots<\alpha_{k-1}(N)$.[7,21] This fact means that in the thermodynamic limit the ground state is at least twofold degenerated at $\alpha>\frac{1}{4}$. Let us determine the dependence of α_1 (actually, $\gamma_1=\alpha_1-\frac{1}{4}$) on N. It is known this dependence defines the scaling parameter in one-dimensional models, where the crossings of two ground state levels occur. At $\alpha=\frac{1}{4}$ ($\gamma=0$) the ground state in the sector $S=0$ has momentum $q=\pi$, while the first excited state has $q=0$ and the excitation energy $\Delta E\sim N^{-4}$.[25] The first order correction to the energy of these states in γ is

$$\delta E_{1(2)}(\gamma) = \gamma\sum_n\langle\Psi_{1(2)}|\mathbf{S}_n\cdot\mathbf{S}_{n+2}|\Psi_{1(2)}\rangle, \tag{34}$$

where Ψ_1 and Ψ_2 are the wave functions of the ground state and the first excited singlet at $\alpha=\frac{1}{4}$. These two states have a spiral ordering at $N\to\infty$ with a period of spirals N and $N/2$ respectively.[7,25] According to this fact, the two-spin correlation functions at $N\to\infty$ are

$$\langle\Psi_1|\mathbf{S}_n\cdot\mathbf{S}_{n+l}|\Psi_1\rangle = \frac{1}{4}\cos\frac{2\pi l}{N},$$
$$\langle\Psi_2|\mathbf{S}_n\cdot\mathbf{S}_{n+l}|\Psi_2\rangle = \frac{1}{4}\cos\frac{4\pi l}{N}. \tag{35}$$

The accuracy of the above equations for $l=2$ is of the order of $O(N^{-3})$.[11,25]

The value γ_1 is determined from the condition

$$\delta E_1(\gamma_1)-\delta E_2(\gamma_1)=\Delta E, \tag{36}$$

which gives

$$\gamma_1\sim N^{-3}. \tag{37}$$

Therefore, the scaling parameter of model (2) in the vicinity of the transition point $\alpha=\frac{1}{4}$ is $x=\gamma N^3$. It means that the perturbation theory in γ contains infrared divergencies and the ground state energy has a form

$$E(\gamma)=\frac{\gamma}{N}f(x), \tag{38}$$

where the scaling function $f(x)$ at $x\to 0$ is given by the first order correction (34). In the thermodynamic limit ($x\to\infty$) the behavior of $f(x)$ is generally unknown, but the condition $E\sim N$ at $N\to\infty$ requires

$$E(\gamma)\sim -N\gamma^{\beta}. \tag{39}$$

According to Eqs. (38) and (39) $\beta=\frac{5}{3}$. This value is close to $\beta=\frac{12}{7}$ which indicates high quality of the variational approaches used. A possible reason for the discrepancy between the variational and scaling estimates of the exponent may be related to the fact that in the variational approaches the terms H_2 and H_3 in Eq. (7) are irrelevant. It can be also expected that the true dependences in Eqs. (20) and (23) at small $0<\gamma\ll 1$ are

$$\varphi\sim\gamma^{1/3},\quad M^*\sim\gamma^{1/3},\quad h_s\sim\gamma^{5/3},\quad \chi\sim\gamma^{-4/3}. \tag{40}$$

V. SUMMARY

We have studied the frustrated spin-$\frac{1}{2}$ Heisenberg chain with the NN ferromagnetic and the NNN antiferromagnetic exchange interactions using two different variational approximations: the MFA and the BVA. The first step of both approaches consists in rotation of the coordinate system on the pitch angle φ and the canted angle θ, which are not equal to their classical values and used as variational parameters of the approaches. Then, in the MFA the rotated spin Hamiltonian is mapped into the model of interacting spinless fermions by means of the Jordan-Wigner transformation. The latter is treated in the mean-field approximation with the inclusion of superconducting like correlations. Within the BVA we use the Agranovich-Toshich boson transformation of spin operators and the variational treatment of the bosonic Hamiltonian. It is remarkable that, despite the difference of these approaches, they give quantitatively close results.

The variational approaches used allowed us to estimate the critical exponent of the ground state energy β in the vicinity of the transition point from the ferromagnetic state to the singlet one. Both approaches give $\beta=\frac{12}{7}$ which differs from the classical value $\beta=2$. Since approaches used are variational, we have established an important and strict fact that quantum fluctuations definitely change the classical critical exponent. Using the results of finite-size calculations we presented also some scaling arguments in favor of the critical exponent $\beta=\frac{5}{3}$. This value is close to $\beta=\frac{12}{7}$ which indicates high quality of the variational approaches used.

The behavior of the magnetization curve is different in parameter regions $\alpha\lesssim 0.38$ and $\alpha\gtrsim 0.38$. In the region $\alpha\lesssim 0.38$ the metamagnetic transition to saturation takes place. At $\alpha\gtrsim 0.38$ the magnetization increases with h until the field reaches the critical value h_c, where the magnetization jumps from M_1 to $M_2<\frac{1}{2}$. This magnetization jump accompanies the jump of the pitch angle φ and canted angle θ. At $h<h_c$ both angles are incommensurate, while at $h>h_c$ they correspond to the commensurate phase $\varphi=\theta=\pi/2$. Therefore, we associate this jump with the incommensurate-commensurate transition induced by the magnetic field.

We believe that our approaches correctly predict the existence of the incommensurate-commensurate transition at some critical field h_c and $\alpha>0.38$. This transition must accompany some singular behavior of the magnetization curve, though we are not sure whether the true magnetization has the jump at $h=h_c$.

Both approximations used yield the exact value of the saturation field at $\alpha>0.38$, though the field dependence near the saturation does not correspond to the expected universal square-root behavior. We note that the magnetization behavior near the saturation in the region of sufficiently large value of α is described by model (26), which looks simpler than model (2). In this respect an accurate study of model (26) is of a particular interest.

We note that the magnetization curve in Fig. 3 resembles (apart from the magnetization jump) that observed in $Rb_2Cu_2Mo_3O_{12}$. This resemblance allows us to provide a qualitative explanation of the peculiarity of the magnetization process in this compound. The experimental curve is characterized by a sharp change of the susceptibility at the magnetic field $B\simeq 14$ T. We assume that this peculiarity in the magnetization behavior is related to the crossover between the incommensurate and the commensurate states.

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