

Magnetic phase diagram of a quasi-one-dimensional quantum spin system

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We propose an analytical ansatz, using which the ordering temperature of a quasi-one-dimensional (quasi-1D) antiferromagnetic (AF) system (weakly coupled quantum spin-1/2 chains) in the presence of the external magnetic field is calculated. The field dependence of the critical exponents for correlation functions of 1D subsystems plays a very important role. It determines the region of possible re-entrant phase transition, governed by the field. It is shown how the quantum-critical point between two phases of the 1D subsystem, caused by spin-frustrating next-nearest neighbor and multispin ringlike exchanges, affects the field dependence of the ordering temperature. Our results qualitatively agree with the features, observed in experiments on quasi-1D AF systems.

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I. INTRODUCTION

The progress in preparation of quantum spin substances with well-defined one-dimensional (1D) subsystems has motivated the interest in studies of them during last years. Another reason for the investigation of properties of quasi-1D spin systems is the relatively rare possibility of comparison experimental data with results of exact theories for many-body models. According to the Mermin-Wagner theorem,¹ totally 1D spin systems with isotropic spin-spin interactions cannot have a magnetic ordering at nonzero temperatures. However, for quasi-1D spin systems, which 1D subsystems have gapless spectrum of low-lying excitations, the magnetic susceptibility, specific heat, and muon spin relaxation often manifest peculiarities, characteristic for phase transitions to magnetically ordered states. The critical temperature of the ordering of a quasi-1D AF Heisenberg spin system was first calculated in Ref. 2 in the absence of the external magnetic field. However, for quasi-1D spin systems, in which (the largest) exchange constants along the distinguished direction are relatively small ($\sim 1-20$ K), the transition to the ordered state can be governed by the external magnetic field. Nowadays in low-temperature experiments high values of the magnetic field (about 20 T for stationary fields and about 60 T for pulse fields) can be used. Therefore, for many quasi-1D spin systems it is possible to investigate experimentally how the external magnetic field affects the Néel ordering, i.e., to determine the H - T phase diagram.

This is why, the objection of the present study is to construct an analytical theory (convenient for comparison with experiments), which has to show how the external magnetic field affects the magnetic ordering in a system of weakly coupled AF Heisenberg spin chains. To calculate the ordering temperature of the quasi-1D system we use the mean-field approximation for a weak interchain interactions. In this approach 1D subsystems are considered as clusters (of infinite size), for the description of which we can use nonperturbative results.³ As a result, we propose a relatively simple analytical ansatz for the magnetic field dependence for the Néel temperature of a quasi-1D Heisenberg AF system.

II. MEAN-FIELD APPROXIMATION

Consider a three-dimensional spin-1/2 system with AF interactions between spins, which form a hyper-cubic lattice. In the quasi-1D situation the Hamiltonians of the 1D subsystems are Heisenberg Hamiltonians of AF spin chains

$$\mathcal{H}_{1D} = J \sum_n (\mathbf{S}_n \cdot \mathbf{S}_{n+1}) - H \sum_n S_n^z, \quad (1)$$

where $J > 0$ is the AF exchange coupling between nearest neighbor spins in the chain, $H = g\mu_B B$, B is the magnetic field, g is the g factor of magnetic ions, and μ_B is Bohr's magneton. Denote by $J' \ll J$ the weak interchain coupling between spins belonging to different 1D subsystems of the quasi-1D system. If the system is AF ordered, we can write the magnetization of the n th site of the system as

$$\mathbf{M}_n = M \mathbf{e}_z + (-1)^n m_N \mathbf{e}_x, \quad (2)$$

where $\mathbf{e}_{x,z}$ are the unit vectors in the x or z directions, M is the average magnetization, and m_N is the staggered magnetization in the direction, perpendicular to the external field (the order parameter in the considered case). The interchain interaction can be taken into account in the mean-field approximation. In that approximation in the AF phase we write the Hamiltonian of the total system as

$$\mathcal{H}_{mf} = \mathcal{H}_{1D} + zJ'M \sum_n S_n^z - h_N \sum_n (-1)^n S_n^x + \text{const}, \quad (3)$$

where $h_N = zJ'm_N$ and z is the coordination number. The order parameter m_N (or h_N) has to be determined self-consistently. The self-consistency equation reads $m_N = M_N(H, h_N, T)$, where $M_N(H, h_N, T)$ is the magnetization per site of the 1D subsystem in the effective field $H - MzJ'$ at the temperature T . In other words, the susceptibility of the quasi-1D system can be written in the mean-field approximation as

$$\chi_{q1D} = \frac{\chi_N}{1 - zJ'\chi_N} \quad (4)$$

and the ordering takes place at the values of the temperature and the field, at which the denominator becomes zero. Then the transition temperature to the ordered state has to be determined from the equation

$$1 = zJ'\chi_N, \quad (5)$$

$$\chi_N = [\partial M_N(H, h_N, T) / \partial h_N]_{h_N \rightarrow 0}.$$

Notice that χ_N is exponentially small for the situation with gapped low-energy eigenstates of the spin chain. It takes place, e.g., for the Heisenberg spin chain for $H > H_s$ in the ground state ($H_s = 2J$ is the critical value of the magnetic field, at which the spin chain undergoes a quantum phase transition to the spin-saturated phase). In that case weak couplings J' cannot yield a magnetically ordered state of a quasi-1D system. Therefore, in the following we consider only the case with gapless low-energy eigenstates of the spin chain.

III. SUSCEPTIBILITY OF THE ONE-DIMENSIONAL SUBSYSTEM

The nonuniform static susceptibility of the 1D subsystem at low temperatures can be written as

$$\chi_\alpha(q, T) = -i \sum_n \int dt e^{-iqn} \Theta(t) \langle [S^\alpha(n, t), S^\alpha(0, 0)] \rangle_T, \quad (6)$$

where q is the wave vector, $\alpha = x, y, z$, and $\langle \dots \rangle_T$ denotes the thermal average at the temperature T . Asymptotic behavior of correlation functions for an integrable spin chain for the gapless case can be obtained in the conformal field theory limit³ and it is possible to write the staggered part of the correlation function for the transverse to the magnetic field components in the ground state (related to χ_N) as

$$\langle S_n^x(t) S_0^x(0) \rangle \approx (-1)^n \frac{C}{[n^2 - (vt)^2]^{\eta/2}} + \dots, \quad (7)$$

where v is the Fermi velocity of low-energy excitations, $\eta = 1/2Z^2$ is the correlation function exponent, Z is the dressed charge of low-lying excitations (low-energy eigenstates), and C is a nonuniversal constant.⁵ Asymptotic behavior can be extended for weak nonzero temperatures using the conformal mapping $(n \pm vt) \rightarrow (v/\pi T) \sinh[\pi T(n \pm vt)/v]$. Then, we can calculate susceptibilities for $q = \pi$ (we use the main approximation), Fourier transforming of the conformal mapping of Eq. (7) at low temperatures as

$$\chi_N = \frac{C}{v} \left(\frac{2\pi T}{v} \right)^{2-\eta} B^2 \left(\frac{\eta}{4}, \frac{2-\eta}{2} \right), \quad (8)$$

where $B(x, y) = \Gamma(x)\Gamma(y)/\Gamma(x+y)$ is the Euler's beta function. Finally, we obtain the expression for the Néel temperature below which the magnetic ordering takes place

$$T_N \approx \frac{v}{2\pi} \left[C \frac{zJ'}{v} \sin \left(\frac{\pi\eta}{2} \right) B^2 \left(\frac{\eta}{4}, \frac{2-\eta}{2} \right) \right]^{1/2-\eta}. \quad (9)$$

IV. BETHE-ANSATZ APPROACH

Fermi velocity and the critical exponent η can be calculated exactly using the Bethe ansatz.³ In the ground-state phase with gapless low-energy eigenstates at $H < H_s$, we can write $Z = \xi(A)$, $v = \varepsilon(A)/2\pi\sigma(A)$, where $\xi(x)$ and $\sigma(x)$ and $\varepsilon(A)$ are determined from the solution of the Fredholm integral equations of the second kind

$$\sigma(x) + \frac{1}{2\pi} \int_{-A}^A dy \frac{4\sigma(y)}{(x-y)^2 + 4} = \frac{1}{\pi(1+x^2)},$$

$$\rho(x) + \frac{1}{2\pi} \int_{-A}^A dy \frac{4\rho(y)}{(x-y)^2 + 4} = -\frac{4x}{2\pi[1+(x-A)^2]},$$

$$\xi(x) + \frac{1}{2\pi} \int_{-A}^A dy \frac{4\xi(y)}{(x-y)^2 + 4} = 1 \quad (10)$$

and

$$\varepsilon(A) = \frac{4A}{(1+A^2)^2} + \int_{-A}^A dx \rho(x) \left(H - \frac{2J}{x^2 + 1} \right). \quad (11)$$

The boundaries of integrations are related to the value of the magnetic field ($0 \leq H \leq H_s$ for the phase with gapless excitations while for $H > H_s$ the spin chain is in the spin-saturated phase with gapped excitations) via $H = 2\pi J\sigma(A)/\xi(A)$. Equations (10) can be solved analytically only in some limiting cases and numerically in other cases. In the absence of interactions between z components of neighboring spins (so-called XY model, the Hamiltonian of which can be exactly mapped to the one of the noninteracting fermion model using the Jordan-Wigner transformation), the dressed charge is equal to 1. It is also equal to unity for the isotropic Heisenberg chain at $H = H_s$, where $A = 0$. In the absence of the magnetic field we have $A = \infty$, and the solution of integral equations can be obtained by the Fourier transformation, which yields $v = \pi J/2$, and $Z = 1/\sqrt{2}$. The numerical solution for intermediate values of A shows that the dressed charge as a function of H grows from $1/\sqrt{2}$ to 1 for $0 \leq H \leq H_s$, see, e.g., Ref. 3, i.e., η decreases from 1 to 1/2 in this domain of field values. Similarly, the velocity of low-energy excitations decreases with the growth of the field from $\pi J/2$ to zero in the domain $0 \leq H \leq H_s$. Numerical solution for the Néel temperature was given, e.g., in Ref. 4.

V. SIMPLE ANALYTIC ANSATZ

It is not convenient, however, from the viewpoint of application of the results for comparisons with experimental data to use numerical solutions. This is why, we propose the simple ansatz for the magnetic field behavior of the velocity v and correlation function exponent η , valid in the interval $0 \leq H \leq H_s$

$$v = \frac{\pi J}{2} \sqrt{[1 - (H/H_s)][1 - (H/H_s) + (2H/\pi J)]},$$

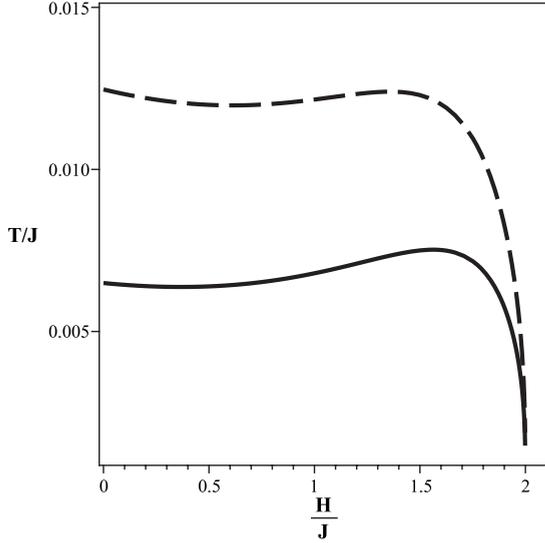


FIG. 1. The H - T phase diagram of the quasi-1D Heisenberg spin-1/2 system with AF interactions between nearest neighbors (the value of the spin saturation field $H_s=2J$). The dashed line represents the result without logarithmic corrections while the solid one is related to the case with taken into account logarithmic corrections.

$$\eta = \frac{\sqrt{4f^2 - 3H^2}}{2f}, \quad f = \pi J \left(1 - \frac{H}{H_s}\right) + H. \quad (12)$$

The nonuniversal constant is equal to 0.18 at $H=0$ and near the saturation it behaves approximately as $C \sim 0.18\sqrt{1-2M}$, where M is the average spin moment per site, cf. Ref. 5, which leads to the field dependence $C \approx 0.18[2(H_s - H)/2\pi H_s]^{1/4}$ in the vicinity of the critical saturation point H_s . Our ansatz is exact at the points $H=0$ and $H=H_s$ and in the vicinity of $H=H_s$. The main deviations of our ansatz from exact results take place for intermediate field values, between zero and H_s . In the above expression for the Néel temperature (and in the ones for the velocity and the critical exponent) we did not take into account logarithmic corrections, which exist for the characteristics of the isotropic Heisenberg AF spin-1/2 chain near $H=0$, see, e.g., Ref. 3. Those corrections can be taken into account, which yield for the susceptibility⁶

$$\chi_N \rightarrow \chi_N \frac{\sqrt{\ln(24.27J/T)}}{(2\pi)^{3/2}}. \quad (13)$$

Then for the Néel temperature we can modify our Eq. (9) as (cf. Ref. 5)

$$C \rightarrow (2\pi)^{-7/4} \sqrt{\sqrt{2(H_s - H)/H_s} \ln([48.54\pi J/v])}. \quad (14)$$

Equations (9), (12), and (14) are the main result of our work.

VI. RESULTS FOR THE MAGNETIC PHASE DIAGRAM

In Fig. 1 we present the Néel temperature of a quasi-1D spin-1/2 AF system for $z=4$ and $J'=0.1J$ as a function of the

external magnetic field H , i.e., the H - T phase diagram of the system (our results qualitatively agree with the results of numerical calculations for Bethe-ansatz equations, see, e.g., Ref. 4). The quasi-1D system is in the magnetically ordered state in the interval of fields and temperatures, limited by the line of the second-order phase transition. Logarithmic corrections do not change the qualitative behavior of the Néel temperature as a function of the field. However, the values of the critical temperatures become smaller due to logarithmic corrections. The Néel temperature as a function of the external field first grows and then goes to zero at the critical field $H=H_s$. Hence, there exists a (narrow) interval of temperatures, at which one can observe a re-entrant phase transition. In this domain of temperatures, if we enlarge the value of the field, a quasi-1D spin system is first in the paramagnetic short-range phase. Then the system undergoes a phase transition to the magnetically ordered phase, and then, for larger values of the field, it returns to the paramagnetic phase. It would be interesting to observe such a re-entrant phase transition in real quasi-1D spin systems. Such a behavior is the consequence of the field dependence of the critical exponent. If the critical exponent does not depend on the magnetic field (e.g., in the XY chain), the Néel temperature as a function of the field only decreases (following the field dependence of the velocity). Exact calculation of the Néel temperature for quasi-1D spin system with the Dzyaloshinskii-Moriya (DM) interaction⁶ also revealed similar to Fig. 1 behavior, i.e., the maximum in the field dependence of the critical temperature. Our calculations for spin systems with DM interactions or with the “easy-plane” magnetic anisotropy show that such magnetically anisotropic interactions produce the reduction of the maximum in the field dependence of the critical temperature. Notice that the critical temperature for weakly coupled XY spin chains is higher than for Heisenberg chains due to field-independent exponent. Also, if the symmetry of the lattice of the total system is lower than hyper-cubic, the ordering can take place not at $q=\pi$, but for some values of q , which depend on the lattice structure and relativistic interactions (such a case can be analyzed in the random phase approximation). In that case the Néel temperature also becomes smaller than in the hyper-cubic situation, cf. Ref. 6.

VII. EFFECT OF NEXT-NEAREST NEIGHBOR AND MULTISPIN EXCHANGE COUPLINGS

In real quasi-1D spin systems additional intrachain exchange interactions between next-nearest-neighbor (NNN) spins exist very often.⁷ To take into account such interactions we can consider the modified 1D Hamiltonian

$$\mathcal{H}_{\text{NNN}} = J_1 \sum_n (\mathbf{S}_n \cdot \mathbf{S}_{n+1}) + J_2 \sum_n (\mathbf{S}_n \cdot \mathbf{S}_{n+2}) - H \sum_n S_n^z, \quad (15)$$

where J_2 is the exchange integral for next-nearest-neighbor couplings. For $J_2 > 0$ such a spin chain reveals a spin frustration. Unfortunately, for this Hamiltonian an exact solution cannot be obtained analytically for any values of $J_{1,2}$. Nevertheless, approximate bosonization studies and numerical

calculations suggest that for $J_2 > 0.24 \dots J_1$ the spin gap is opened for low-energy excitations.⁸ For the above mentioned reasons a system of weakly coupled chains with gapped excitations cannot be ordered magnetically. However, as follows from Ref. 7, despite the fact that for most of studied compounds exchange constants satisfy the condition $J_2 > 0.24 \dots J_1$, the spin gap was not confirmed experimentally. To describe theoretically quasi-1D spin systems with spin frustration due to intrachain interactions without spin gap and with a weak interchain coupling, we consider another model, the Hamiltonian of which is \mathcal{H}_{NNN} with additional terms, describing multispin ringlike interactions. The advantage of that model is its exact integrability: the model permits an exact Bethe-ansatz solution. We do not state, naturally, that the model describes all features of the experiments.⁷ However, many properties of the model are similar to what was observed in Ref. 7, at least, for this model low-energy eigenstates are gapless. Hence, from this viewpoint, it qualitatively agrees with the data of experiments,⁷ unlike the model with the Hamiltonian \mathcal{H}_{NNN} . Multispin ring-exchange interactions are often present in oxides of transition metals, where a direct exchange between magnetic ions is complimented by a superexchange between magnetic ions via nonmagnetic ones.⁹ The modified Hamiltonian of such 1D subsystem has the form

$$\begin{aligned} \mathcal{H}_{\text{mod}} = & \mathcal{H}_{\text{NNN}} + J_4 \sum_n [(\mathbf{S}_{n-1} \mathbf{S}_{n+1})(\mathbf{S}_n \mathbf{S}_{n+2}) \\ & - (\mathbf{S}_{n-1} \mathbf{S}_{n+2})(\mathbf{S}_n \mathbf{S}_{n+1})]. \end{aligned} \quad (16)$$

Notice that multispin interactions are less relevant from the renormalization-group viewpoint than two-spin interactions. Quantum properties of the model can be seen from the exact solution,¹⁰ for the parametrization of coupling constants $J_1 = J(1-y)$, $J_2 = Jy/2$, and $J_4 = 2Jy$ for any J and y (in what follows we consider $J > 0$, $y \geq 0$). This exactly solvable model, while being formally less realistic than the model with the Hamiltonian \mathcal{H}_{NNN} , reveals features, more similar to the properties of experimentally studied quasi-1D systems with spin frustration.⁷ For $y=0$ the model describes the Heisenberg spin-1/2 chain. The ground state of the model depends on values of the parameter y and an external magnetic field.¹⁰ At $T=0$ for large values of the magnetic field the model is in the spin-saturated phase, divided from other phases by the line of the second-order quantum phase transition. For low values of y and H the model is in the phase, which properties are similar to the phase of the Heisenberg spin-1/2 chain in a weak magnetic field (Luttinger liquid).¹⁰ The model is in this phase for $y < y_{cr} = 4/\pi^2$ at $H=0$ and for $y < y_{cr}(H)$ for nonzero fields. The point y_{cr} is the quantum critical one. For $y > y_{cr}$ the model is in an incommensurate phase with nonzero spontaneous magnetization at $H=0$. Last two phases are divided from each other by the line of the second-order quantum phase transition. Quantum phase transitions can be observed in the temperature behavior of thermodynamic characteristics of the model, such as the magnetic susceptibility and the specific heat, that were also calculated exactly.¹⁰ This model permits us to know, how NNN interactions (together with the multispin ones), which

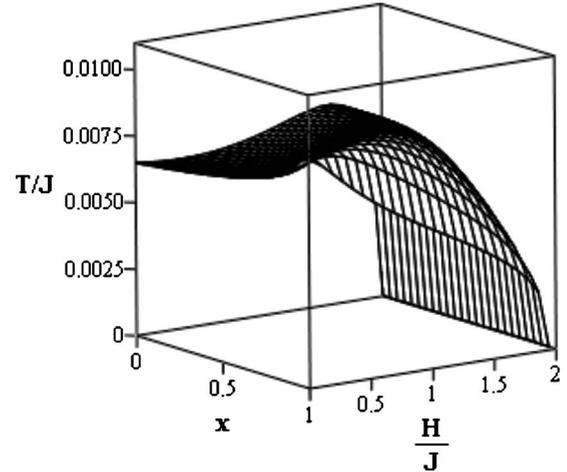


FIG. 2. The phase diagram for a quasi-1D spin-1/2 chain with spin frustration, caused by nearest, NNN interactions and the ring exchange. The Néel temperature is a function of the parameter x , which shows how close the quantum-critical point (caused by spin-frustrating interactions) $x=1$ is, and the external magnetic field.

cause the quantum phase transition, can modify the H - T phase diagram, obtained above for the case of only nearest-neighbor couplings in chains. In this situation the expressions for the velocity and the critical exponent can be modified using the substitution $\pi J \rightarrow \pi J(1-x)$ (cf. Ref. 10), where $x = y/y_{cr}$. We concentrate on the case $y < y_{cr}$ ($0 \leq x \leq 1$) for the 1D subsystem, which has Luttinger liquid properties. In this case a quasi-1D spin system undergoes the transition to the AF ordered state.¹¹ Ordering in the incommensurate phase was studied in Ref. 11. The results of our analysis are presented in Fig. 2. The ordered phase is inside the region, limited by the surface, at which the second-order phase transition takes place. The maximum in the field dependence of the critical temperature, cf. Fig. 1, is shifted toward low values of the field with the growth of x , i.e., spin-frustrating NNN and multispin couplings can reduce the domain of temperatures, at which re-entrant phase transition can take place. We believe, that while the considered model seems less realistic, the mentioned feature has the generic nature for quasi-1D spin systems.

Phase diagrams, similar to the ones, presented in Figs. 1 and 2, were obtained experimentally for real quasi-1D compounds.¹² Phase H - T diagrams in those compounds show maxima of the field dependencies of critical temperatures. Namely, the ordering temperature in studied quasi-1D copper oxides first increases with the growth of the value of the field, reaches its maximum, and then decreases to zero at the value of the field, where the spin chain has the spin saturation. Notice, that in one of those compounds measurements reveal different values of the ordering temperatures for different directions of the external field, which can be caused by the weak magnetic anisotropy of the intrachain exchange interactions.

VIII. CONCLUSIONS

In summary, we have used a simple analytical ansatz to calculate the ordering temperature of a quasi-1D system,

consisting of weakly interacting quantum spin-1/2 chains with AF couplings in the presence of the external magnetic field, when the weak interchain coupling is taken into account in the mean-field approximation, and the characteristics of spin chains are obtained nonperturbatively. Our results show that the field dependence of the critical exponents for correlation functions of 1D subsystems plays a very important role. In particular, that dependence determines the region of possible re-entrant phase transition, governed by the field. We have shown also how a quantum-critical point between two phases of the 1D subsystem, caused by spin-frustrating NNN and multispin ringlike exchanges, affects the field dependence of the ordering temperature. Our results qualita-

tively agree with the features, observed in experiments on quasi-1D AF systems. We expect that our results are generic for quasi-1D systems and that they can be helpful for experimentalists, who study magnetic properties of such systems, especially due to the recent progress in obtaining high values of the magnetic field in experiments.

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