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SOLIDS

Quantum Monte Carlo Investigation of the Magnetic Properties of Weakly Interacting Antiferromagnetic Chains with an Alternating Exchange Interaction with Spin $S = 1/2$

S. S. Aplesnin

Kirenskiĭ Institute of Physics,
Siberian Division, Russian Academy of Sciences,
Krasnoyarsk, 660036 Russia;
e-mail: apl@iph.krasnoyarsk.su

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Abstract—An approximation dependence of the spontaneous magnetic moment at a site, $\sigma/\sigma(0) - 1 = 0.71(6)\delta^{2.5(2)}$, and the antiferromagnet-singlet state phase boundary, $J_2/J_1 = 0.52(3)\delta$, are determined by the quantum Monte Carlo method in the self-consistent sublattice molecular field approximation for weakly interacting (J_2) antiferromagnetic chains with spin $S = 1/2$ and alternating exchange interaction ($J_1 \pm \delta$). The Néél temperature and a number of critical temperatures which could be related with the filling energy of two singlets ($\Delta S^z = 0$) and one triplet ($\Delta S^z = 1$) spin bands, each of which is split by the sublattice field ($h^{x,y} \neq h^z$) into two subbands, are determined on the basis of the computed correlation radii of the two- and four-spin correlation function, the squared total spin $\langle (S^z)^2 \rangle$ with respect to the longitudinal components, the dimerization parameter, and the correlation functions between the nearest neighbors with respect to longitudinal and transverse spin components. On the basis of the Monte Carlo calculations, the critical temperatures and possible energy gaps at the band center are determined for the antiferromagnets $CuWO_4$ and Bi_2CuO_4 and for the singlet compounds $(VO)_2P_2O_7$ and $CuGeO_3$, agreeing satisfactorily with existing results, and new effects are also predicted. © *2000 MAIK "Nauka/Interperiodica".*

1. INTRODUCTION

There exists a wide class of magnetic compounds with spatially anisotropic distribution of exchange interactions and with a strong interaction between the magnetic and elastic subsystems that in certain cases results in a spin-Peierls transition. Ordinarily, a transition from a singlet state into the paraphrase is studied in the Hubbard or Heisenberg models with alternating exchange parameter J and hopping integral t using the mean-field theory or Green's functions together with perturbation theory. As a rule, spinon excitations are neglected in the analysis of these systems, resulting in overestimation of the temperature of the spin-Peierls transition when interchain exchange is taken into account, specifically, logarithmic behavior [1].

Alternating exchange can also be achieved by means of the geometry of the crystal lattice, such as in $CuWO_4$ [2], Bi_2CuO_4 [3, 4], $(VO)_2P_2O_7$ [5], and $(CH_3)CHNH_3CuBr_3$ [6]. These compounds are all three-dimensional magnetic systems with alternating exchange. For most of them, the exchange interactions in three directions of the corresponding crystal axes have been determined. The magnetic properties of the antiferromagnets $CuWO_4$ and Bi_2CuO_4 are interpreted in a two-sublattice Heisenberg model, and the existence of several branches of spin excitations, whose intensity becomes zero at different temperatures, and the presence of an energy gap at band center for $\omega = 1.4$ meV in $CuWO_4$ [2] and at $\omega_i = 0.7$, 1.7, 2.3, 3.4, and 4 meV in Bi_2CuO_4 [4, 7, 8] remain incomprehensible. Nonmonotonic temperature behavior of the susceptibility [9], the antiferromagnetic resonance field, and the linewidth in Bi_2CuO_4, whose temperature derivatives have several maxima [10], and an additional maximum of the specific heat at $T \simeq 17$ K ($T_N = 45$ K) in Bi_2CuO_4 [11], are observed in these antiferromagnets.

Several energy gaps in the spin excitation spectrum, which do not fit either into the conventional theory of the spin-Peierls transition with one triplet gap [12] or into the theory of the two-magnon excitation spectrum [13], have also been found in the singlet magnets $CuGeO_3$, $Na_2V_2O_5$, and $(VO)_2P_2O_7$. Of these compounds, $CuGeO_3$ has been studied in greatest detail. In this compound three temperature ranges have been found, $T_{c1} \sim$ (4–7) K, $T_{c2} \simeq 14$ K, and $T_{c3} \sim$ (20–25) K, where the EPR linewidth and intensity exhibit anomalous behavior [14, 15], and the magnetic thermal conductivity [16] and magnetostriction [17] possess maxima below and above the spin-Peierls transition temperature $T_N = 14$ K.

The present paper is devoted to an investigation of the region of stability of long-range antiferromagnetic order in an isotropic $3D$ antiferromagnet with a quite strong anisotropic distribution of exchange interactions

in the lattice relative to the magnitude of the alternating exchange, and the determination of the site magnetic moment, the Néél temperature, and the critical temperatures at which the correlation radii assume their maximum values. According to the dynamic scaling hypothesis, the relaxation time τ is proportional to the correlation radius, $\tau \propto \xi^z$, and the temperatures indicated above can be found from the temperature dependence of the EPR linewidth, antiferromagnetic resonance, and diffuse neutron scattering. Additional spin excitations, spinons [18], will be proposed on the basis of the four-spin correlation function. These excitations have several excitation bands, which make it possible to explain previously reported experimental results and, using the computed values of the critical temperatures, predict the existence of additional spin modes and a number of new effects.

2. MODEL AND METHODS

Let us consider a Heisenberg model with negative interactions between nearest neighbors with spin $S = 1/2$ in an external magnetic field oriented in the Z direction. The alternating interaction is taken in the strong-coupling direction $I = J_1 + \delta$ and $K = J_1 - \delta$. The Hamiltonian has the form

$$H = -J_1 \sum_{i,j} \mathbf{S}_{i,j}\mathbf{S}_{i+1,j} - \sum_{\substack{i,j,\gamma=1 \\ \alpha=x,y,z}} J_2^{\alpha}(\gamma) S_{i,j}^{\alpha} S_{i,j+\gamma}^{\alpha} - \sum_i H_i S_i^z, \quad (1)$$

where $J_1 < 0$ and $J_2 < 0$ are the intra- and interchain interactions, H is the external magnetic field, and γ signifies summation over the nearest neighbors between chains ($z = 4$). We transform the Hamiltonian of the 3D system to a one-dimensional chain of spins, which interact with the effective field, by means of the self-consistent molecular field approximation [19, 20]:

$$H = -\sum_{i=1}^{L/2} I_{2i,2i-1}\mathbf{S}_{2i}\mathbf{S}_{2i-1} - \sum_{i=1}^{L/2} K_{2i,2i+1}\mathbf{S}_{2i}\mathbf{S}_{2i+1} - \sum_{i=1}^{L} h_i^{\alpha} S_i^{\alpha} - \sum_{i=1}^{L} H_i S_i^z - 2NJ_2 m_0^2, \quad (2)$$

where m_0 and h are the sublattice magnetization and field $\mathbf{h}_i(h^z, h^+, h^-)$, determined in [19, 20] as $m_0 = (1/L)\sum_{i=1}^{L}(-1)^i\langle S_i^z\rangle$ and $h = -4J_2 m_0$. To take account of the quantum and temperature fluctuations, we shall determine these quantities from the spin–spin correlation function, which is a power-law function of on distance in a 1D antiferromagnet at $T = 0$. We shall assume that this dependence also holds also for the transverse spin components in the magnetically ordered region of a quasi-one-dimensional antiferromagnet, while in the paramagnetic and singlet states the instantaneous values of the sublattice field are proportional to the magnitude of the short-range order, i.e., the spin-spin correlation function of the nearest neighbors. The average value is $\langle h_i\rangle \approx 0$, and $\langle h_i^2\rangle \neq 0$ in the singlet state. Taking account of the fluctuations of the sublattice field in the singlet state leads to new effects, which will be described below. We shall study in the singlet and paramagnetic states two types of the sublattice field with respect to transverse spin components: isotropic $h^x = h^y = h^z$ and anisotropic, characteristic for $CuGeO_3$, $h^x = h^y = 1.4h^z$. In an antiferromagnet the sublattice fields have the form

$$m_0 = \frac{2}{L}\sum_{i=L}^{L/2}\sqrt{\mathrm{abs}(S_0^z S_i^z)},$$
$$h_i^z = 4J_2\,\mathrm{sign}(\langle S_0^z S_i^z\rangle)\sqrt{\mathrm{abs}(S_0^z S_i^z)}, \quad (3)$$
$$b - h_i^{+,-} = 4J_2(-1)^i\sqrt{\mathrm{abs}(S_0^+ S_1^-)/i}.$$

In this work, the quantum Monte Carlo method, which employs a trajectory algorithm of world lines, based on a transformation of a D-dimensional quantum system into a $(D + 1)$-dimensional classical system by discretization of the path integral in the space (imaginary time $0 < \tau < 1/T$, coordinate) [21, 22], is used. In the Monte Carlo calculations, Trotter's formula with the parameter $m = 32$, 64, 124, and 200 and periodic boundary conditions on a chain of length $L = 100$, 200, and 400 is used. One Monte Carlo step was determined by rotating all spins on a $L \times 2m$ lattice. From 4000 to 7000 Monte Carlo steps per spin were used to reach equilibrium, and 2000–5000 Monte Carlo steps per spin were used for averaging. The autocorrelation time τ required to establish thermodynamic equilibrium was estimated from the relation $\ln(\tau) = amT/J$ (T is the temperature) [23]. The systematic error due to quantum fluctuations is proportional to $\sim 1/(mT/J)^2$ and is of the order of 4% for the minimum temperature $T/J = 0.025$, used in the calculations. The rms errors of the computed quantities lie in the range (0.1–0.6)% for energy, (6–11)% for susceptibility, and ~10% for the correlation radius. The errors due to the finite dimensions of the lattice can be neglected, since $\xi < L/2$.

Let us consider the possible spin excitations in this model. If the wave function of the ground state is schematically represented as a sum of the Néél configuration and a set of singlet states of spins with different weight ratios, then, besides ordinary excitations of the spin-wave type, there can exist excitations in singlet regions that can be divided into two groups: the longitudinal component of the spin vector does not change, i.e., $\Delta S^z = 0$ (we call such excitations singlet excitations), and the longitudinal component of the spin vector changes by one unit, i.e., $\Delta S^z = 1$, which correspond

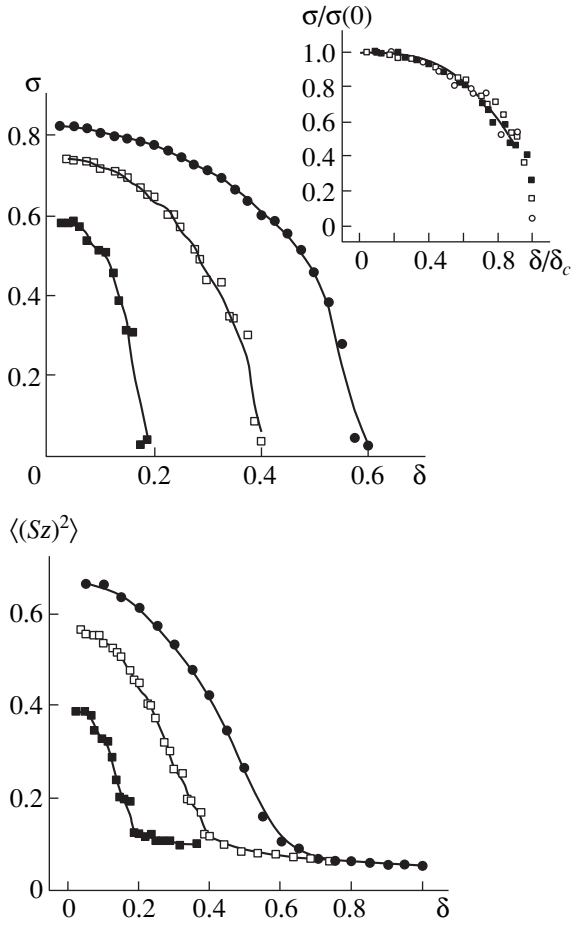

Fig. 1. Site magnetic moment (σ) and squared longitudinal component of the total spin ($\langle (S^z)^2 \rangle$) of an antiferromagnet with λ = 0.1 (■), 0.2 (□), and 0.3 (●) versus the exchange alternation. Inset: Normalized values of the magnetization for the same parameters.

to triplet excitations. According to Anderson's theory [24], the singlet state is described well by a generalized resonance valence bond (RVB) model whose wave function is represented in the form of singlet pairs of spins over all possible configurations. In the presence of exchange alternation the generalized model reduces to a simple RVB model in which pairing of nearest spins is taken into account. Since two types of exchange interactions, differing in magnitude, exist here, the energies of the singlet pairs and the corresponding excitations on *K* bonds differ from the energy on *I* interactions. For this reason, the characteristic features of the temperature behavior of the magnetic characteristics of an antiferromagnet with alternating exchange can be calculated and understood on the basis of the four-spin correlation function of spin pairs $\langle \mathbf{S}_0\mathbf{S}_1\mathbf{S}_r\mathbf{S}_{r+1} \rangle$ and the dimmer ordering parameter *q*:

$$q^\alpha = \frac{4}{L}\sum_{i=2}^{L/4}(\langle S_0^\alpha S_1^\alpha S_{2i-2}^\alpha S_{2i-1}^\alpha \rangle - \langle S_1^\alpha S_2^\alpha S_{2i-1}^\alpha S_{2i}^\alpha \rangle), \quad (4)$$

$$\alpha = x, y, z.$$

A classical excitation of the kink type, which in what follows we shall call a spinon excitation, corresponds to the excited state in the simple RVB model with one dangling bond. We shall determine the correlation radius ξ_4 of the spinons and the parameter η_4 from the four-spin correlation function

$$\left|\langle S_0^z S_1^z S_{r-1}^z S_r^z \rangle - \langle S_0^z S_1^z S_{L/2-1}^z S_{L/2}^z \rangle\right| = \frac{A}{r^{\eta_4}}\exp(-r/\xi_4), \quad (5)$$

where $r = 2i + 1$ and $i = 1, 2, 3 \ldots$.

The following quantities will be calculated below: the energy, the specific heat $C = dE/dT$, the magnetization, the susceptibility $\chi = M/H$ in an external field, the spin-spin correlation function between longitudinal $\langle S^z(0)S^z(r) \rangle$ and transverse $\langle S^+(0)S^-(1) \rangle$ spin components, the Fourier spectrum $S(q) = (2/L)\sum_{r=1}^{L/2}\exp(-iqr)(S_0^zS_r^z)$, and the magnetic structure factor. We shall determine the correlation radius ξ_2 and the parameter η_2 from the spin-spin correlation function

$$\left|\langle S^z(0)S^z(r) \rangle - \langle S^z(0)S^z(L/2) \rangle\right| = \frac{B}{r^{\eta_2}}\exp(-r/\xi_2). \quad (6)$$

The squared total spin will be calculated from the longitudinal component $\langle (S^z)^2 \rangle$. This parameter makes it possible to distinguish a singlet state from a paramagnetic state and is sensitive to a change in the spin excitation spectrum.

3. DISCUSSION

We shall use a number of criteria to determine the antiferromagnet–singlet state phase boundary in the interchain interactions–alternating exchange plane: the sublattice magnetization is zero, $\sigma \longrightarrow 0$, and the correlation radii ξ_2 and ξ_4 for δ_c have their maximum value. The singlet state is distinguished from the paramagnetic state or the spin-glass state according to the following indicators. In the singlet state, in a model with alternating exchange, the total spin is zero, $S = 0$, and the eigenvalue of the operator $\hat{S}_z^2$ is also zero on the basis of the equality $\langle (S^z)^2 \rangle = S(S + 1)/3$. The dimmer ordering parameter is different from zero, $q \neq 0$, and the relation $\langle S_0^+S_1^- \rangle \simeq 2\langle S_0^zS_1^z \rangle$ holds between the longitudinal and transverse components of the spins. We shall calculate the characteristics indicated above, some of which are displayed in Fig. 1, at low temperatures, $(0.1–0.2)T_N$, for a number of values of the interchain exchange parameters $\lambda = J_2/J_1 = 0.05, 0.075, 0.1, 0.125, 0.15, 0.2, 0.25$, and 0.3 as a function of the magnitude of alternation. The normalized values of the sublattice magnetization and the energy can be approximated well by power-law dependences $\sigma/\sigma(0) - 1 = 0.71(6)\delta^{2.5(2)}$ and

$E/E(0) - 1 = 0.02\delta^{3.6(3)}$, where $\sigma(0) = 1.9(1)\sqrt{\lambda}$, and are shown in the inset in Fig. 1. In the singlet state, the absolute value of the internal energy increases with the exchange alternation, $(E - 0.85) \approx 0.63\delta^{1.2(1)}$, which agrees well with the results for a one-dimensional chain, $\sim\delta^{4/3}$ [25]. The difference in the exponent could be due to the correlation effects of the interaction of the chains, which are taken into account in the form of the self-consistent sublattice fields **h** (3). The magnetic state for $\delta > \delta_c$ is a singlet state, and the finite quantity $\langle(S^z)^2\rangle$ is due to the singlet excitations with $\Delta S^z = 0$, since Monte Carlo calculations are performed at finite temperatures. The phase boundary of the transition can be approximated well by the linear function $\lambda = 0.52(3)\delta$.

For an antiferromagnet with alternating exchange in the form of two sublattices with strong I and weak K exchange interactions, three types of spinon (pair) excitations can be distinguished: I–I, K–K, and I–K. In the sublattice field ($h^{+,-} \neq h^z$), each of these spinon bands can split into subbands with transverse and longitudinal spin excitations. The wave functions of these excitations can be represented as

$$\psi^{s1} \propto c_1(|\ldots\uparrow\uparrow\downarrow\downarrow\ldots\rangle - |\ldots\downarrow\downarrow\uparrow\uparrow\ldots\rangle) + c_2(|\ldots\downarrow\uparrow\downarrow\uparrow\ldots\rangle - |\ldots\uparrow\downarrow\uparrow\downarrow\ldots\rangle)$$

on the K–K bonds and in the form

$$\psi_l^{s2} \propto (|\ldots\uparrow\uparrow\ldots\downarrow\downarrow\ldots\rangle + |\ldots\downarrow\downarrow\ldots\uparrow\uparrow\ldots\rangle,$$
$$\psi_t^{s2} \propto (|\ldots\uparrow\downarrow\ldots\uparrow\downarrow\ldots\rangle + |\ldots\downarrow\uparrow\ldots\uparrow\downarrow\ldots\rangle \quad (7)$$

on the I bonds. Excitations of this type do not lead to a change in the z component of the total spin ($\Delta S^z = 0$) and do not contribute to the longitudinal susceptibility, so that the minimum in the temperature dependence $\chi(T)$ for some temperature T_{si} corresponds to filling of the band of singlet spinon excitations. Excitations on the I–K bonds change the z component of the spin $\Delta S^z = 1$ and are spinons, or spin waves; this gives rise to a maximum in the temperature dependence of the susceptibility at T_{ti}. The filling of the singlet band of excitations in the I–I sublattice will lead to an increase in the dimmer ordering parameter q (4), and in the K–K sublattice it will lead to a sharp decrease of the parameter q. We shall determine the splitting into subbands according to the magnitude of the temperature variation of the dimerization parameter $q^{x,y,z}$ and the near-range correlation functions $\langle S_0^\alpha S_1^\beta\rangle$ with respect to the longitudinal and transverse spin components.

The temperature dependences of the above-indicated characteristics are calculated for three interchain exchange parameters, $\lambda = 0.05$, 0.1, and 0.25, and the corresponding values of the exchange alternation, $\delta = 0.05$, 0.075, 0.12, 0.14, and 0.2; $\delta = 0.1$, 0.15, 0.2, 0.3, 0.45, 0.6; and, $\delta = 0.15$, 0.3, 0.45, 0.6, 0.75. The critical

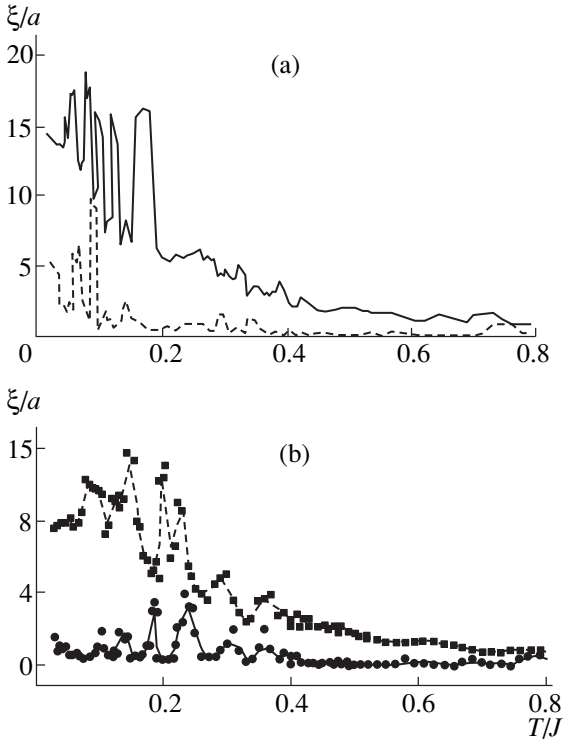

Fig. 2. Temperature dependence of the correlation radii of two-spin (solid line in Fig. a and ■ in Fig. b) and four-spin (broken line in Fig. a and ● in Fig. b) correlation functions for the antiferromagnetic state with $\lambda = 0.1$ and $\delta = 0.15$ (a) and for the singlet state with $\lambda = 0.05$, $\delta = 0.14$, and $h^{+,-} = 1.4h^z$ (b).

temperatures are determined according to the maxima of the correlation radii $\xi_2(T)$ and $\xi_4(T)$, which are shown in Fig. 2 for antiferromagnetic and singlet states, according to the maximum change in the longitudinal component of the squared total spin $\langle(S^z)^2\rangle$, i.e., according to the maxima of $d\langle(S^z)^2\rangle/dT$ and the extremal points of the temperature dependence $q(t)$, presented in Fig. 3. On the basis of an analysis of the temperature behavior of the susceptibility $\chi(T)$ (Fig. 4), the critical temperatures were associated to the filling energy of triplet (χ = max) and singlet (χ = min) spin excitation bands. The filling of these bands forms three maxima in the temperature dependence of the specific heat (Fig. 4).

A qualitative estimate of the relations between these temperatures, $T_{ti} - T_{si} \propto \sqrt{\lambda^2 \pm 2\delta\lambda + \delta^2}$, where the minus sign corresponds to T_{s1} and the plus sign to T_{s2}, apparently, will also be valid for the energy gaps between these excitation bands. An even weaker effect is the splitting of the proposed spin bands by the sublattice field, which occurs for $\delta > 0.1$. The temperature at which the change in the correlation function between nearest neighbors with respect to the longitudinal spin components is much greater than this change with respect to transverse components (this appears most

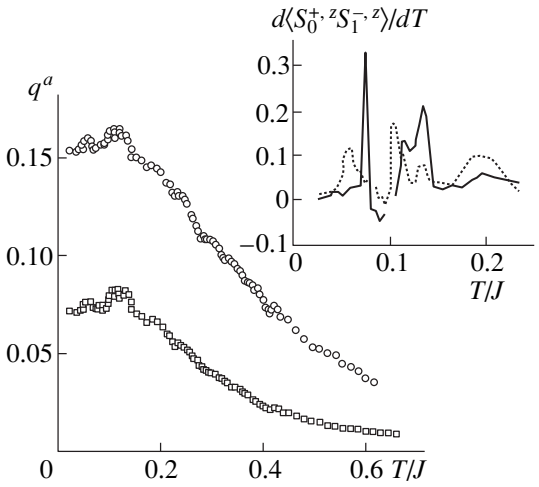

Fig. 3. Temperature dependence of the dimmer ordering parameter q^α ($\alpha = z$ (□), x, y = (○)) in an antiferromagnet with $\lambda = 0.1$ and $\delta = 0.15$. Inset: Temperature dependence of the derivative $d\langle S_0^\alpha S_1^\beta\rangle/dT$ with $\alpha, \beta = z$ (dotted line) and with $\alpha = +, \beta = -$ (solid line).

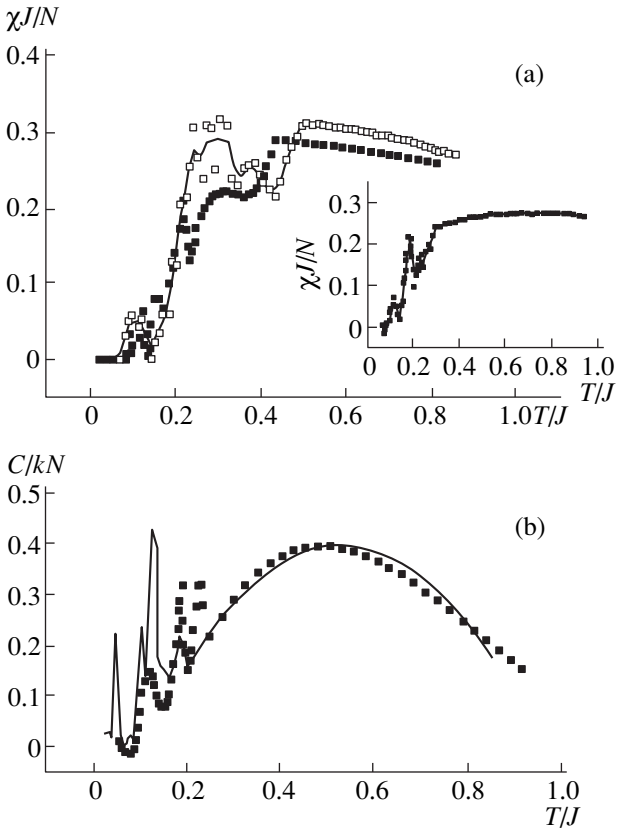

Fig. 4. (a) Temperature dependence of the susceptibility, calculated in the sublattice field $h^{+,-} = 0$ (□) and $h^{+,-} \neq 0$ (■) according to (3) in an antiferromagnet with $\lambda = 0.1$ and $\delta = 0.15$. Inset: $\chi(T)$ in the singlet state with $\lambda = 0.05$, $\delta = 0.14$, and $h^{+,-} = 1.4h^z$. (b) Temperature dependence of the specific heat in an antiferromagnet with alternating exchange and the parameters $\lambda = 0.1$ and $\delta = 0.15$ (solid line) and $\lambda = 0.05$ and $\delta = 0.14$ (■).

strikingly in the calculation of $d\langle S_0^{s,+} S_1^{z,-}\rangle/dT$ (inset in Fig. 3)) refers to the excitation energy of the longitudinal spin mode. In the singlet state, the sublattice field at a distance of the order of the correlation radius influences the spin excitations with wavelength $\sim\pi/\xi$, and for $\delta \gg \delta_c$ the interaction of chains has no effect on the redistribution of the spin excitation density (as compared with the one-dimensional chain) for $\lambda \leq 0.01$. It is possible that each spin subband is characterized by a definite wave vector of the structure $Q_i < \pi/a$, which can found from the Fourier spectrum of the spin correlation function $S(q)$, determined in the singlet state at a distance $k \sim 1/\xi_2$. Thus, $S(q)$ in the singlet state contains weak additional maxima at Q_i, whose number increases with temperature.

The Néél temperature was determined from the sublattice magnetization $\sigma \longrightarrow 0$. In the range of bond alternations close to the critical value, two sharp dropoffs appear clearly in the temperature behavior of the magnetization, for example, for $\delta = 0.15$ and $\lambda = 0.1$ for $T_{s1}/J_1 = 0.06$ and $T_t/J_1 = 0.11$; they are associated with the filling of the triplet spin excitation band in the temperature range $T_{s1} < T < T_t$ and in the spin-wave band at temperatures $T > T_t$. The computed critical temperatures for $\lambda = 0.1$ are presented in Fig. 5. The vanishing of the long-range antiferromagnetic order can be understood from this diagram. As exchange alternation increases, the spin-wave excitation density decreases and vanishes at $T_N \sim T_t$. When the sublattice fields are the same, $h^{+,-} = h^z$, the splitting into subbands vanishes, and for the singlet and paramagnetic states only two critical temperatures exist, above and below the spin-Peierls transition temperature, which are shown in Fig. 5 by the dashed and dotted lines. From the standpoint of symmetry, three phases can be distinguished in the temperature–exchange alternation plane: a region with long-range antiferromagnetic order, a region where the thermodynamic value of the spin is zero, i.e., a singlet state, and a region where $\langle S^z\rangle \sim H/k_BT$, i.e., the paramagnetic state. The phase diagrams (see Fig. 5b), calculated for the three parameters λ in normalized units, are the same to within the computational error.

A variety of incomprehensible experimental data for the antiferromagnet $CuWO_4$ with alternating exchange [2] can be explained on the basis of the results obtained: the existence of an energy gap at the center of the band of the spin excitation spectrum at $\omega = 1.4$ meV and the different temperature dependences of the intensity of spin modes, one of which vanishes at $T = 24$ K and the other (gapless) remains even at $T = 36$ K [2]. The temperature dependence of the susceptibility in the range $40 < T < 70$ K has a concave form [9]. Our computed intrachain exchange parameters $J = 11.6$ meV and $K = 8.9$ meV agree well with neutron diffraction measurements $J = 11.56$ and $K = 9.25$ meV [26]. The interchain exchange $J_2 \sim 1$ meV agrees fairly well with the aver-

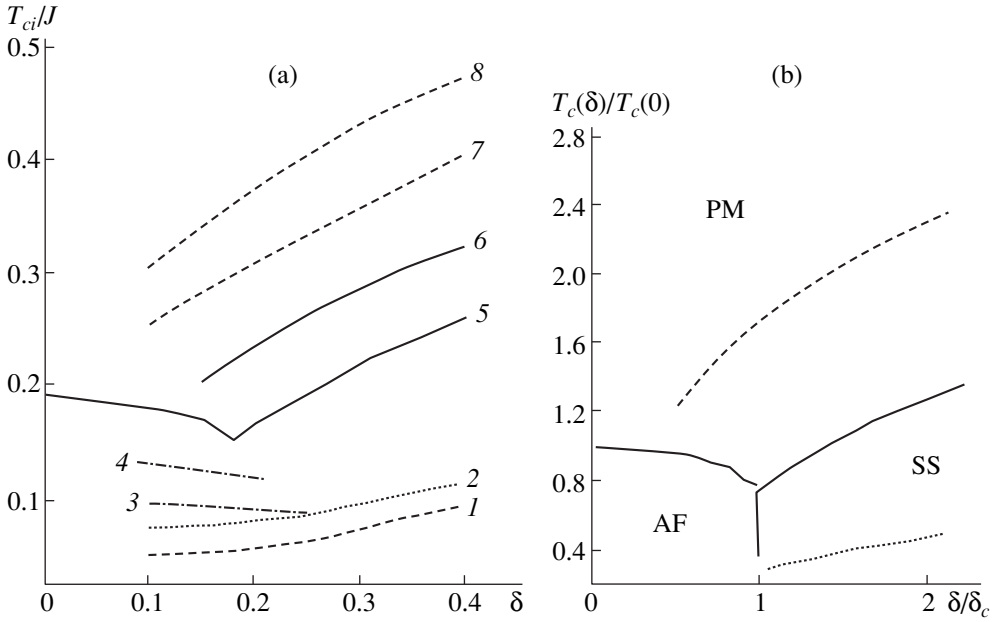

Fig. 5. Critical temperatures, associated with the filling energy of singlet (*1*, *2*, *7*, *8*) and triplet (*3*, *4*, *5*, *6*) spinon bands as a function of the exchange alternation for $h^{+,-} \neq h^z$ (a). Phase diagram of an antiferromagnet (AF), singlet state (SS), and paramagnet (PM) in the plane normalized temperature – normalized exchange alternation for $h^{+,-} = h^z$ (b).

age value $J_2 \approx 1.7$ meV [2]. According to the Monte Carlo calculations, the spin triplet band is filled in the temperature range $17 < T < 24$ K. At $T > 24$ K the gapless mode corresponds to spin-wave excitations, which should vanish at $T_c \approx 40$ K. The band of singlet excitations, not contributing to the magnetic susceptibility, is filled in the temperature range $52 < T < 86$ K, which gives rise to the inflection in the temperature behavior of $\chi(T)$. It is possible that another singlet mode with gap energy $\omega \sim 0.7$ meV exists at temperatures $T < 12$ K.

A series of energy gaps at the center of the band in Bi_2CuO_4 [4, 7] with tetragonal symmetry *P*4/*ncc* and alternating exchange in the [111] direction can be explained by the existence of singlet and triplet spin excitations. The temperature dependences of the resonance field H_0 and the anisotropy field H_a are nonmonotonic, and their derivatives dH_0/dT and dH_a/dT possess several maxima, with different magnitudes, at the corresponding temperatures, $T_{ci} = 8$, 12, 18, 26 K and $T_{ci} = 7$, 11, 17, 26, 38 K [10]. For the intra- and interchain exchange parameters chosen, $J_1 = 107$ K and $J_2 = 28$ K, the exchange alternation is $\delta = 0.2$, and our computed critical temperatures, related with the filling of the split singlet and triplet spinon bands at $T_{ci}^{MC} \simeq 7$, 11, 15, 25, 30, 35, agree well with the experimental results. It is possible that the energy gaps at $\omega_{s1} = 0.7$ meV and $\omega_{s2} = 3.4$ and 4 meV at the center of the Brillouin zone are due to singlet excitations of longitudinal and transverse spinon modes, and at $\omega_t = 1$, 7, and 2.1 meV by triplet excitations. Several spin-excitation modes also exist in singlet magnets with alternating exchange. For example, in $(VO)_2P_2O_7$ two gaps were found at $\omega = 3.12$ and 5.75 MeV [5]. It is possible that one other singlet mode with gap at band center at $\omega \simeq 0.6$ meV and with weak intensity exists in this compound. For $\lambda \simeq 0.02$ [5] our computed exchange alternation $\delta \approx 0.15(2)$ lies in the range $\delta_n = 0.12$ and $\delta_\chi = 0.18$, which were determined, respectively, in neutron resonance (*n*) experiments [5] and from the temperature dependence of the susceptibility [27].

The compound $CuGeO_3$ with a spin-Peierls transition was investigated in greatest detail. According to the neutron diffraction data, the ratio of the exchange interactions is $J_c : J_b : J_a = 100 : 10 : 1$ [28] and, according to our estimates, for $\delta > \delta_c \simeq 0.1$ a magnet with such an exchange ratio can be in a singlet state. Inelastic neutron scattering data revealed three gaps at band center at $\omega_i = 0.8$ meV (1.9–2.1) meV [29] and 4(±1) meV [30] and a wide maximum in neutron scattering $\omega \simeq 6$ meV [29], two values for which are close to the results $\omega = 1.86$ and 4.74 meV obtained from EPR measurements [14]. Light scattering at the boundary of the Brillouin zone also leads to three energy gaps $\omega_i = 2.2$, 3.6, and 5.8 meV [31, 32]. The magnetic thermal conductivity in a magnetic field up to $H = 14$ T has two maxima at $T = 5.5$ and 22 K [16], and the width of the EPR line diverges at $T \simeq 4$ and 14 K [15]. In a magnetic field applied in the direction of the *c* axis, the temperature dependence of the magnetostriction constants has three maxima at $T \simeq 6$, 13, and 26 K and in a field $H \parallel b$ at $T \simeq 4$, 11, and 20 K [17].

These results can all be explained well by choosing the two intrachain exchange parameters $I = 145$ K and

K = 109 K, which agree fairly well with the data obtained based on the 1D Heisenberg model, in which the triplet gap $\omega \simeq 2$ meV has been calculated [33] and the spin-Peierls transition temperature is T_{sp} = 14 K. The observation of thermodynamic anomalies above and below T_{sp}, additional energy gaps, anisotropy of the critical magnetic fields, which are encountered in a ~10% range, where the field H_c = 13.9 T is applied in the direction of the exchange alternation axis and the field H_b = 12.6 T is applied perpendicular to this axis [34], remains unexplained. As indicated above, at a distance of the order of the correlation radius $\xi_2 \simeq 10c$, the interchain interaction leads to splitting of the low-energy band of singlet excitations with respect to its center into longitudinal and transverse excitation modes with gap energies $\omega_b \simeq 0.5$ meV and $\omega_c \simeq 0.8$ meV and corresponding critical temperatures $T_b \simeq 3.7$ K and $T_c \simeq$ 6 K, splitting of the triplet modes with gap energies $\omega_b \simeq 1.7$ meV and $\omega_c \simeq 2$ meV and corresponding critical temperatures $T_b \simeq 14$ K and $T_c \simeq 18$ K, and splitting of the high-energy longitudinal and transverse singlet modes with $\omega_b \simeq 4.5$ meV and $\omega_c \simeq 5.8$ meV and corresponding temperatures $T_b \simeq 30$ K and $T_c \simeq 39$ K. However, at temperatures $T > T_{sp}$ small structural distortions are observed in $CuGeO_3$, which lead to a change in the magnitude of the exchange and its alternation. For this reason, in the temperature range $20 < T <$ 26 K one can talk about a qualitative agreement between the temperature interval ΔT_{ex} = 6 K and the Monte Carlo computational results ΔT_{MC} = 9 K. It is possible that in $CuGeO_3$ in the singlet state the quantization axis is directed along the b axis, and then the anisotropy of the critical magnetic field is explained well. Thus, our calculations predict a polarization dependence of light scattering and inelastic neutron scattering along the dimerization axis of a chain. At low temperatures the system is nonlinear, so that the definition of the nonlinear susceptibility $M_\gamma^3 = \chi_{\gamma,\alpha,\beta,\delta} H_\beta H_\alpha H_\delta$ must be used to calculate the resonance absorption frequencies. It is possible that as a result of the nonlinear interaction of the field with the spin subsystem, a transition occurs from the singlet ground state into a singlet excited state, as is observed in EPR measurements at frequency ω = 294 GHz [14], and transitions occur between the subbands $\psi_l^s \longrightarrow \psi_t^s$ at the frequency f = 34 GHz in a field $H \approx 12$ kOe [15]. The intensity of both resonances has a maximum at T = 6 K and vanishes at $T < 2$ K; this agrees well with our estimates for the energy of the singlet gap and the critical temperature.

Thus, the long-range antiferromagnetic order in the quasi-low-dimensional antiferromagnet with alternating exchange remains for $\lambda \le 0.52(3)\delta$. Alternation gives rise to quantum spin reduction at a site $\sigma/\sigma(0) - 1 = 0.71(6)\delta^{2.5(2)}$, where $\sigma(0) = 1.9\sqrt{\lambda}$. Several temperatures, at which the correlation radii are maximum and the thermodynamic characteristics exhibit features, which can be interpreted on the basis of the model of additional spinon singlet and triplet excitations, were found in an alternating antiferromagnet and in the singlet state. The sublattice self-consistent field ($h^{+,-} \neq h^z$) splits the spinon bands into longitudinal and transverse excitation modes. The temperatures corresponding to the maxima of the derivatives of the resonance field and the linewidths as a function of temperature in Bi_2CuO_4 were calculated using dynamic scaling between the relaxation time and the correlation radius. Possible spinon excitation modes and energy gaps at the center of the band in antiferromagnetic states of the compounds $CuWO_4$ and Bi_2CuO_4 and in the singlet states of the compounds $(VO)_2P_2O_7$ and $CuGeO_3$ were predicted. The temperatures corresponding to the maxima of the magnetic thermal conductivity, the magnetostriction constants, and the divergence of the EPR linewidth in $CuGeO_3$ were calculated. A polarization dependence of light and neutron scattering along the dimerization axis of a chain was predicted.

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