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# Magnetic ordering of weakly coupled frustrated quantum spin chains

A. A. Zvyagin[1,2] and S.-L. Drechsler[3]

[1]*Max-Planck-Institut für Physik Komplexer Systeme, Nöthnitzer Strasse, 38, D-01187 Dresden, Germany*

[2]*B. I. Verkin Institute for Low Temperature Physics and Engineering, National Academy of Sciences of Ukraine, Lenin Avenue, 47, Kharkov 61103, Ukraine*

[3]*Leibniz Institut für Festkörper- und Werkstoffforschung Dresden, PF 270116 D-01171 Dresden, Germany*

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The ordering temperature of a quasi-one-dimensional system, consisting of weakly interacting quantum spin-1/2 chains with antiferromagnetic spin-frustrating couplings (or zigzag ladder), is calculated. The results show that a quantum critical point between two phases of the one-dimensional subsystem plays a crucial role. If the one-dimensional subsystem is in the antiferromagneticlike phase in the ground state, similar to the phase of a spin chain without frustration, weak couplings yield magnetic ordering of the Néel type. For intrachain spin-frustrating interactions larger than the critical one (at which the quantum phase transition takes place), the quasi-one-dimensional spin system manifests a spiral magnetic incommensurate ordering. The obtained results of our quantum theory are compared with the quasiclassical approximations. The calculated features of magnetic ordering are expected to be generic for weakly-coupled quantum spin chains with gapless excitations and spin-frustrating nearest- and next-nearest-neighbor interactions.

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The interest in quasi-one-dimensional (quasi-1D) quantum spin systems has grown considerably during the last decades. The characteristic feature of quasi-1D magnets is the strong spin-spin interaction along one space direction, much stronger than the couplings along all other directions. The interest is motivated, on the one hand, by the progress in preparation of substances with well-defined 1D subsystems. Another reason for studying quasi-1D spin systems is the possibility to compare experimental data with results of nonperturbative theories for 1D models. Also, such systems often manifest quantum phase transitions that take place in the ground state and which are governed by other parameters than the temperature, such as external magnetic field, pressure, concentration of impurities (internal pressure), etc. From the experimental viewpoint, quasi-1D spin-1/2 systems differ from other magnets due to special features in the behavior of their characteristics. For example, the temperature dependence of the magnetic susceptibility and the specific heat of quasi-1D spin systems with dominant nearest-neighbor (NN) interactions reveal maxima (in a small external magnetic field at temperatures of the order of the exchange coupling constant along the distinguished direction).[1] For spin systems, which 1D subsystems have a gapless spectrum of low-lying excitations at temperatures, much lower than the temperature of the maximum of the $T$ dependence of the susceptibility, the latter and the specific heat often manifest peculiarities, characteristic for phase transitions to low-temperature magnetically ordered states at critical temperatures.

Also, a great attention has been recently given to spin systems with a spin frustration. For most of antiferromagnetic (AF) systems the ground state corresponds to Néel-type configurations. The standard quasiclassical description of AF systems uses quantization of small deviations of vectors of magnetizations (magnetic order parameters) of AF sublattices from their steady-state configuration. However, such a description of AF systems can be used for bipartite magnetic structures. For AF systems with a spin frustration the competing interactions produce a very high degeneracy of such steady-state configurations. Therefore, in most cases it is hopeless to use the approximation of magnetic sublattices. From the theoretical viewpoint the situation becomes even worse in quasi-1D spin systems with a spin frustration. For those systems quantum fluctuations are enhanced due to peculiarities in the 1D density of states. This is why, according to the famous Mermin-Wagner theorem,[2] 1D spin systems with short-range interactions cannot have any magnetic ordering even at $T=0$. Thus, approximate methods of theoretical physics often produce significant errors in the description of such systems. Hence, it is necessary to study them nonperturbatively, better exactly, which is, fortunately, possible for few cases for 1D quantum spin systems. Probably one of the simplest and most known examples of a quantum spin system with a spin frustration is the Heisenberg spin-1/2 chain with AF NN and AF next-nearest-neighbor (NNN) interactions. The Hamiltonian of such a model can be written as

$$\mathcal{H}_{\mathrm{NNN}} = J_1 \sum_n \mathbf{S}_n \mathbf{S}_{n+1} + J_2 \sum_n \mathbf{S}_n \mathbf{S}_{n+2}, \tag{1}$$

where $J_1$ and $J_2$ are the couplings between NN and NNN, respectively (here we consider only the case with an even number of spins $N$). Such a system is equivalent to a zigzag spin ladder with obvious renotation of indices. The system with Hamiltonian (1) is, obviously, spin frustrated. Several limiting cases are known exactly. Namely, for $J_2=0$ (or for $J_1=0$) Hamiltonian (1) is reduced to the Hamiltonian of one (or two decoupled) Heisenberg AF spin-1/2 chain(s). The ground state for those cases is a nondegenerate singlet, without long-range orderings, and the low-energy excitations are gapless spinons.[1] The other limiting case, for which the ground state is known exactly, is the so-called Majumdar-Ghosh point, $J_1=2J_2$.[3] In that case the

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Hamiltonian of the zigzag spin ladder can be rewritten as $\mathcal{H}_{\rm NNN}=(J_1/4)\Sigma_n(\mathbf{S}_n+\mathbf{S}_{n+1}+\mathbf{S}_{n+2})^2-9N/4$. The ground state is given by two degenerate singlets of the resonance valence bond type, without long-range ordering, and the low-lying excitations are gapped. For other values of the coupling constants it is, unfortunately, impossible to obtain exact answers. Nevertheless, an approximate bosonization description and numerical calculations suggest that there is no long-range magnetic ordering in the system and that for $J_2>0.2411\cdots J_1$ a spin gap is opened for the low-lying excitations.[4] A quasiclassical approximation of the model yields the following. If one replaces the spin operators by classical vectors, two steady-state configurations are possible. The first one is the period 2 commensurate and collinear AF Néel configuration, stable for $J_1>4J_2$. The second one, stable for $J_1<4J_2$, is a noncollinear incommensurate spiral magnetic structure with the pitch angle $\cos\phi=-J_1/4J_2$. Such a description implies a long-range magnetic order. This means that it might be approximately valid for, e.g., a quasi-1D spin system, consisting of weakly-coupled 1D spin chains with NN and NNN AF interactions at temperatures lower than the ordering temperature. However, for quasi-1D spin systems with a spin gap $\Delta_{\rm sp}$ for low-energy excitations of their 1D subsystems, a weak interchain coupling, as a rule, does not produce a magnetic ordering.[5] (This is plausible at least for spin systems with isotropic exchange interactions: the exponential decay of the long-range spin-spin-correlation function $\propto\exp(-n/\xi)$ with a finite coherence length $\xi=\hbar v/\pi\Delta_{\rm sp}$, where $v$ is the Fermi velocity of the low-lying spin excitations, conflicts with the magnetic order requiring asymptotically nondecaying spin-spin-correlation functions.)

On the other hand, it is clear that spin frustration in a 1D subsystem has to yield features in transitions to possible magnetically ordered state for a quasi-1D system. Moreover, as follows from Ref. 6 (see also Ref. 7) despite the fact that for most of studied compounds exchange constants satisfy the condition $J_2>0.2411\cdots J_1$, the spin gap was not confirmed experimentally. To describe such experimental situations (i.e., quasi-1D AF spin systems with spin frustration of intrachain interactions without a spin gap and with a weak interchain coupling), we consider another model, the Hamiltonian of which consists of $\mathcal{H}_{\rm NNN}$ with multispin interaction. Such a model is known to have gapless low-energy excitations. Those multispin interactions do not change the spin frustration property from the classical viewpoint.[8] The advantage of the proposed model is the exact integrability: The model permits an exact solution by means of the Bethe's ansatz. We do not state here, naturally, that the model describes all features of the materials of current experimental interest.[6] However, many properties of the model are similar to what was observed in Ref. 6. At least, for this model the low-lying excitations are gapless. Hence, from this viewpoint, they qualitatively agree with the data of experiments, unlike the model with the Hamiltonian $\mathcal{H}_{\rm NNN}$. Multiple spin-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] Models with multispin interactions are believed to be closer to real quasi-1D magnets compared to standard ones with only NN spin couplings.[9] Multispin-exchange models were introduced by Thouless[10] already in 1965. Later similar models were used to study some cuprates[11] and spin ladders.[12] For the consistent explanation of several experiments[13] by means of inelastic neutron scattering, optical conductivity, and nuclear-magnetic resonance, one needs to account for relatively large values of NNN spin-spin interactions and multispin interactions between four neighboring sites of the spin ladder (the so-called ring exchange). Similar four-spin interactions were used recently in the theory of two-dimensional (2D) quantum spin systems, where they regulate the quantum phase transition between the Néel-type ground state and the resonance valence bond solid one.[14] The Hamiltonian of the 1D subsystem of the quasi-1D model, studied in our work, has the form

$$\mathcal{H}_{\rm 1D}=\mathcal{H}_{\rm NNN}+J_4\sum_n[(\mathbf{S}_{n-1}\mathbf{S}_{n+1})(\mathbf{S}_n\mathbf{S}_{n+2})-(\mathbf{S}_{n-1}\mathbf{S}_{n+2})(\mathbf{S}_n\mathbf{S}_{n+1})]. \quad (2)$$

The model is also spin frustrated. The classical counterpart of the model (if one replaces the spin operators by classical vectors) reveals a long-range magnetic ordering with a Néel steady-state configuration or with a spiral magnetic structure, where the four-spin ring exchange renormalizes the spiral pitch angle as $\cos\phi=-2J_1/(8J_2-J_4)$. However, quantum properties of the model with the Hamiltonian $\mathcal{H}_{\rm 1D}$ differ from the one with $\mathcal{H}_{\rm NNN}$ in a much more drastic way than of their classical counterparts. This can be seen from the exact solution (the exactly solvable model was introduced in Ref. 15), which is known for the parametrization of coupling constants $J_1=J(1-x)$, $J_2=Jx/2$, and $J_4=2Jx$ for any $J$ and $x$ (in what follows we shall consider $J>0$ and $x>0$). For $x=0$ the model describes the Heisenberg spin-1/2 AF chain. As one can see from exact results, the high degeneracy of low-energy states, caused by the spin frustration of NN and NNN interactions, is removed by adding the ring exchange, which is also spin frustrated. According to the exactly known properties, the ground state of the model [Eq. (2)] depends on the values of the coupling constant $x$ and an external magnetic field $H$.[16] For large values of $H$ the model is in the spin-saturated (ferromagnetic) phase. This phase has a trivial long-range magnetic order and gapped low-lying excitations. It is divided from other phases by the line of the second-order quantum phase transition. For low values of $x$ and $H$ the model is in the phase, which behavior is similar in properties to the phase of the Heisenberg spin-1/2 AF chain in a weak magnetic field.[1] Note that the model is in this phase for $x<x_{\rm cr}=4/\pi^2$ even at $H=0$. At $x_{\rm cr}$ a second-order quantum phase transition takes place. For $x>x_{\rm cr}$ and $H=0$ the model reveals an incommensurate ordering with nonzero spontaneous magnetization. For nonzero values of $H$ and large enough values of $x$ the model is in the incommensurate magnetic phase with nonzero weak magnetization.[16] The degeneracy in the direction of the spontaneous magnetization can be removed if one first puts the system into an external magnetic field and, then, removes the field. These two phases are divided from each other by the line of the second-order quantum phase transition (quantum critical point).

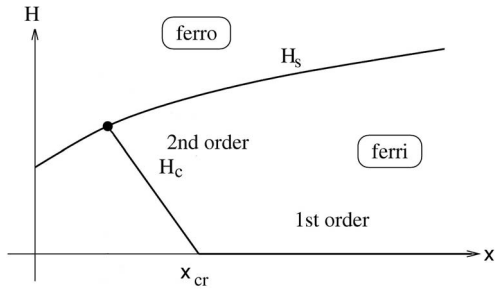


FIG. 1. The phase diagram $H-x$ of the one-dimensional integrable spin model. At the lines $H_s$ and $H_c$ second-order quantum phase transitions to the ferromagnetic (spin-saturated) phase and the ferrimagnetic spiral one, respectively, take place. In the point $H=0$ and $x=x_{cr}$ the second-order quantum phase transition takes place. At the line $H=0$ for $x>x_{cr}$ the first-order phase transition takes place.

Figure 1 shows the ground-state phase diagram of the one-dimensional integrable model. The mentioned 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, which were also calculated exactly.[17]

The goal of our present study is to find how the weak coupling between frustrated spin chains can produce magnetic orderings and what are specific features of such orderings. According to the above, one can suppose two different types of low-temperature magnetic ordering in the quasi-1D system under consideration. For the first one, the Néel ordering, one can write the magnetization of the $n$th site, e.g., as $\mathbf{M}_n=M\mathbf{e}_z+(-1)^n m_N\mathbf{e}_x$, 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. Another possibility is the three-dimensional (3D) generalization of the ground-state incommensurate phase of the 1D subsystem, the spiral incommensurate state with the magnetization of the $n$th site, written as $\mathbf{M}_n=M\mathbf{e}_z+\cos(Qn)m_{sp}\mathbf{e}_z$, where $m_{sp}$ is the modulated component of the $z$ projection of the magnetization around the average magnetization $M$ and $Q=\pi(1-2M)$ is the wave vector of the 1D modulated structure.

As usually, we study the weak interchain coupling $J'$ in the mean-field approximation. In that approximation in the Néel phase we can write the mean-field Hamiltonian of the 1D subsystem as

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

where $h_N=zJ'm_N$ and $z$ is the coordination number. For the spiral phase the mean-field Hamiltonian is

$$\mathcal{H}_{sp}^{mf}=\mathcal{H}_{1D}-(H-zJ'M)\sum_n S_n^z-h_{sp}\sum_n\cos(Qn)S_n^z+\text{const}, \tag{4}$$

where $h_{sp}=zJ'm_{sp}$. Renormalization grouplike approach[1] implies that both $h_N$ and $h_{sp}$ are relevant perturbations. They generate spin gaps $\Delta E_N\sim h_N^{2/(4-\eta)}$ and $\Delta E_{sp}\sim h_{sp}^{2\eta/(4\eta-1)}$, respectively, for low-energy excitations. Here $\eta$ is the correlation function exponent (see below), which determines the asymptotical behavior of the spin-spin-correlation functions of the 1D subsystem in the conformal limit.[17]

The order parameters $m_N$ and $m_{sp}$ (or $h_N$ and $h_{sp}$) have to be determined self-consistently. In the mean-field approximation the corresponding self-consistency equations can be written as

$$m_{N,sp}=M_{N,sp}(H,h_{N,sp},T), \tag{5}$$

where $M_{N,sp}(H,h_{N,sp},T)$ is the magnetization per site of the 1D subsystem in the effective field $H-MzJ'$ and $h_{N,sp}$ at the temperature $T$. Then the transition temperature to the ordered state has to be determined from the equation

$$1=zJ'\chi_{N,st},$$

$$\chi_{N,st}=\left(\frac{\partial M_{N,st}(H,H_{N,sp},T)}{\partial H_{N,sp}}\right)_{H_{N,sp}\to 0}. \tag{6}$$

The nonuniform susceptibilities of the 1D subsystem at low temperature can be found as

$$\chi_\alpha(q,T)=-i\sum_n\int dte^{-iqn}\Theta(t)\langle[S^\alpha(n,t),S^\alpha(0,0)]\rangle_T, \tag{7}$$

where $q$ is the wave vector, $\alpha=x,z$, and $\langle\cdots\rangle_T$ denotes the thermal average at the temperature $T$. Asymptotics of the correlation functions for an integrable spin chain can be obtained in the conformal field theory limit.[1] For the model with the Hamiltonian $\mathcal{H}_{1D}$ it was done in Ref. 17, and we can write for the ground-state correlation functions,

$$\langle S_n^z S_0^z\rangle\approx M^2+\frac{B^*\cos(Qn)}{[n^2-(vt)^2]^{\theta_z}}+\cdots,$$

$$\langle S_n^x S_0^x\rangle\approx(-1)^n\frac{C}{[n^2-(vt)^2]^{\theta_\perp}}+\cdots, \tag{8}$$

where $v$ is the Fermi velocity of low-energy excitations, $\theta_z=Z^2(\equiv 1/2\eta)$, $\theta_\perp=1/4Z^2\equiv(\eta/2)$, $Z$ is the dressed charge of low-lying excitations, and $B^*$ and $C$ are nonuniversal constants. In particular, we see that the symmetry of the ground state is lower than the symmetry of the Hamiltonian, caused by the ordering, i.e., for our model one deals with the manifestation of the Goldstone theorem. Equation (8) can be extended for weak nonzero temperatures using the conformal mapping $(n-vt)\to(2v/\pi T)\sinh[\pi T(n-vt)/v]$.[1] Then, calculating susceptibilities according to Eq. (7) (we use the main approximation) for $q=\pi$ for the Néel phase and for $q=Q$ for the spiral incommensurate phase, we obtain the expressions for the ordering temperatures,

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

and

$$T_{\rm sp} = \frac{v}{2\pi}\left[B^* \frac{zJ'}{v}\sin\left(\frac{\pi}{2\eta}\right)B^2\left(\frac{1}{4\eta},\frac{2\eta-1}{2\eta}\right)\right]^{\eta/(2\eta-1)}, \tag{10}$$

where $B(x,y)$ is the Euler's beta function. In those expressions the Fermi velocity and the critical exponent $\eta$ (or the dressed charge $Z$) can be calculated using the Bethe ansatz results.[17] Then, the question to be answered is which ordering temperature, $T_N$ or $T_{\rm sp}$, is higher for the quasi-1D spin chain with the spin frustration.

In what follows we limit ourselves with the case $H=0$ for simplicity. In this situation the effective Fermi velocity can be written as $v=(\pi/2)J|1-(x/x_{\rm cr})|$.[17] Consider first the ground-state phase $x<x_{\rm cr}$ for a 1D subsystem, which is similar to the ground state of the Heisenberg spin-1/2 chain with only NN AF interactions. In this case we have $M=0$ and $\eta=1$.[17] For this case we can use $B^*=C\sim 0.2$.[18] One can see that in this case (i.e., $\eta=1$) $T_N=T_{\rm sp}=(CzJ'/2\pi)B^2(1/4,1/2)$. We see that the critical temperature does not depend on $J$ and $x$ (obviously, any nonzero magnetic field $h\neq 0$ or an inclusion of a magnetic anisotropy will change this result). To get the $J$ and $x$ dependences even for $H=0$ and for the magnetically isotropic case, one has to include logarithmic corrections,[1] reproducing the known result for $x=0$,[19]

$$T_N = \frac{CzJ'}{2\pi}B^2(1/4,1/2)\sqrt{\ln\left(\frac{\pi^2 J|1-(x/x_{\rm cr})|}{CzJ'B^2(1/4,1/2)}\right)}, \tag{11}$$

which is valid, naturally, when the argument of the logarithm is larger or equal to 1. Next, let us consider the ground state of the 1D subsystem for $x>x_{\rm cr}$, which ground state has a spontaneous magnetic ordering. This spontaneous magnetization $M(x>x_{\rm cr})\neq 0$ is connected with holes in the ground-state distribution of quantum numbers, called rapidities, which form the Dirac sea of the 1D subsystem.[15,17] Those holes appear only for $x>x_{\rm cr}$.[15–17] Notice that in the previous case, $x<x_{\rm cr}$, there are no holes in the Dirac sea, and the ground-state rapidities can have any value in the range $-\infty,\ldots,\infty$.[15–17] It is impossible to find an analytic solution for $\eta$ in this case. We see that $\eta=1+a$, with $0\leq a\leq a_{\max}<1$ when $1\leq x/x_{\rm cr}<\infty$. Unfortunately, in this case we cannot obtain the values of the nonuniversal constants. We can only suppose that they are also of the order of 0.05–0.2.[18] It is easy to see that for most of the values of $J$, $J'$, and $x$ the temperature of the transition to the spiral incommensurate state for $x>x_{\rm cr}$ is higher than the Néel temperature. In Fig. 2 we plotted $T_N$ (lower surface) and $T_{\rm sp}$ (upper surface) for $J=1$, $J'=0.01$, $z=4$, and $B=C=0.2$ as functions of $a$ and $y=(x/x_{\rm cr})$. It turns out that $a$ is a function of $x/x_{\rm cr}$ also, but, unfortunately, one cannot find this dependence analytically. Note that for $a=0$ ($\eta=1$, $x=x_{\rm cr}$) both critical temperatures coincide. The Néel temperature can be larger than the temperature of the transition to the spiral incommensurate phase only in the vicinity of the quantum phase transition $x=x_{\rm cr}$ for $\eta$ being very large [close to 2, which seems to be an overestimation (cf. Ref. 15)]. For all other values of $x>x_{\rm cr}$ we get $T_{\rm sp}>T_N$. Hence, we can conclude that for a quasi-1D system, consisting of weakly-coupled spin-1/2 chains with AF spin-frustrating NN and NNN interactions and with the four-spin ring exchange, the low-temperature ordering depends on the behavior of 1D subsystems. For small values of the NNN interaction, the quasi-1D system undergoes a transition to the magnetically ordered AF Néel state. On the other hand, if the exchange constant of the NNN interactions exceeds the critical value, at which a quantum phase transition to the incommensurate state with the weak spontaneous magnetization takes place, a weak coupling between 1D subsystems produces the transition to the magnetically ordered incommensurate spiral state. The ordering temperature in the latter is

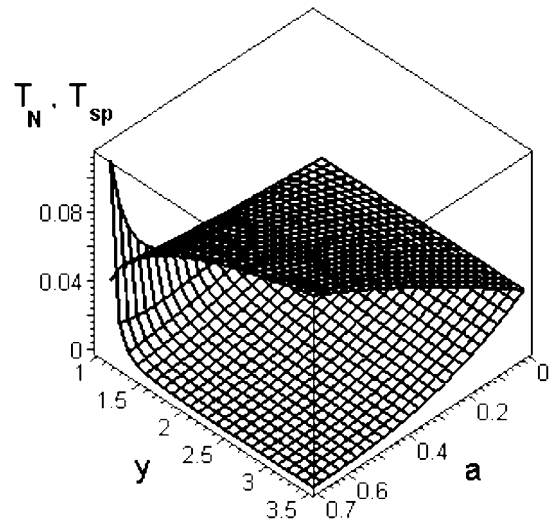


FIG. 2. The Néel temperature (the lower surface) and the ordering temperature of the transition to the spiral incommensurate phase for a quasi-1D spin-1/2 chain with the spin frustration, caused by AF NN, NNN interactions, and the ring exchange as functions of the deviation of the critical exponent $a=1-\eta$ and $y=x/x_{\rm cr}$, which shows how close the quantum critical point $y=1$ is.

$$T_{\rm sp} = \frac{J|1-y|}{4}\left[B^*\frac{2zJ'}{\pi J|1-y|}\sin\left(\frac{\pi}{2(1+a)}\right)\times B^2\left(\frac{1}{4(1+a)},\frac{1+2a}{2(1+a)}\right)\right]^{(1+a)/(1+2a)}. \tag{12}$$

Our quantum analysis qualitatively agrees with the quasiclassical description of the considered system. However, a difference between the quantum system and its quasiclassical counterpart exists: In the quantum system magnetic ordering takes place for $J'\neq 0$ only. We expect analogous expressions in Eqs. (11) and (12) to be valid also for the other critical point between a ferromagnetic and a spiral phase, i.e., if $J_1<0$. In this context, the determination of the magnetic order below their low-temperature phase transitions at few kelvins for $Li_2ZrCuO_4$ and $Pb[Cu(SO)_4(OH)_2]$,[20] both being close to that critical point, would be of interest.

We expect that an interchain ferromagnetic interaction[21] for $x<x_{\rm cr}$ has to produce a AF low-temperature ordering of a special type. Namely, we expect low-temperature ordered phase, which consists of ferromagnetic planes with alternating magnetizations of planes. On the other hand, for $x>x_{\rm cr}$, a

ferromagnetic interchain interaction is expected to produce ferrimagnetic low-temperature ordering with a nonzero spontaneous total magnetization.

Finally, let us consider what happens if one studies the situation with only NN and NNN couplings, without the ring exchange. In that case, for $J_2 < 0.2411\cdots J_1$ the quasi-1D system undergoes a phase transition to the Néel state due to weak couplings between chains. For $J_2 > 0.2411\cdots J_1$, a spin gap is opened for low-lying excitations of the 1D subsystem, and the ordering temperature goes to zero. This case seems to contradict known experiments, in which magnetic ordering was observed even for $J_2 > 0.2411\cdots J_1$.[6] Therefore, we can conclude that for some real compounds with the properties of quasi-1D spin systems with spin-frustrating interactions in their 1D subsystems, some additional spin-spin interactions, such as the ring exchange, studied in this paper, probably exist, which close the spin gap and give rise to magnetic orderings at low temperature.

In summary, the ordering temperature of a quasi-one-dimensional system, consisting of weakly interacting quantum spin-1/2 chains with antiferromagnetic spin-frustrating couplings (or zigzag spin ladder), is calculated. Our results show that the quantum critical point between the two phases of the 1D subsystem plays an important role. If the one-dimensional subsystem is in the ground state in an antiferromagneticlike phase, similar to the phase of a spin chain without frustration, weak couplings yield a magnetic ordering of the Néel type. On the other hand, for intrachain spin-frustrating interactions larger than the critical one (at which the quantum phase transition takes place), an incommensurate spiral magnetic ordering of the quasi-one-dimensional spin system takes place. The obtained results of the quantum theory are compared with the quasiclassical approximations. We expect that the calculated features of the magnetic ordering are generic for weakly-coupled quantum spin chains with gapless excitations and with spin-frustrating nearest- and next-nearest-neighbor interactions. While up to now we do not know quasi-one-dimensional systems with NN AF spin interactions and large AF NNN ones in the spiral phase at low temperatures (cf. Ref. 6, see, though, Refs. 22 and 23), we believe that our results can be used for comparison with the observed temperatures of magnetic orderings in other spin-frustrated quasi-1D quantum spin systems

*Note added in proof.* Recently the predicted spiral ordering of $Li_2ZrCuO_4$ (Ref. 24) has been confirmed experimentally by $^7$Li-NMR measurements.[25] The derived pitch angle $\Phi$ amounts $33° \pm 2°$. This value corresponds to a ratio $-J_2/J_1 = 0.298$ within the framework of the classical relation for the pitch angle $\Phi$ given below Eq. (2) (provided $J_4 = 0$). It is surprisingly very close to the ratio $-J_2/J_1 = 0.3$ estimated from specific heat and susceptibility data analyzed within the 1D-spin-1/2 $J_1J_2$-model.[24] However, a recent quantum analysis within the coupled cluster method (CCM) in the SUB2-3 approximation for a realistic weak effective antiferromagnetic inter-chain coupling $J_\perp \lesssim 0.1|J_1|$ reveals a somewhat *larger* pitch angle: $\Phi \approx 42°$ for $-J_2/J_1 = 0.3$ (Ref. 26). Hence, other factors such as a sizable exchange anisotropy are expected to be relevant in this material. Such an anisotropy may lead to a modification of the classical spiral as well as to a reduction of quantum fluctuations compared with the isotropic spin model considered here. The corresponding effects are outside the scope of the present paper.

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