Order-disorder quantum phase transition in the quasi-one-dimensional spin-1/2 collinear antiferromagnetic Heisenberg model

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The ground-state properties of the quasi-one-dimensional spin-1/2 antiferromagnetic Heisenberg model is investigated by using a variational method. Spins on chains along the x direction are antiferromagnetically coupled with exchange $J > 0$, while spins between chains in the y direction are coupled either ferromagnetically ($J' < 0$) or antiferromagnetically ($J' > 0$). The staggered and the colinear antiferromagnetic magnetizations are computed and their dependence on the anisotropy parameter $\lambda = |J'|/J$ is analyzed. It is found that an infinitesimal interchain coupling parameter is sufficient to stabilize a long-range order with either a staggered magnetization m_s ($J' > 0$) or a colinear antiferromagnetic magnetization m_{caf} ($J' < 0$), both behaving as $\simeq\lambda^{1/2}$ for $\lambda \to 0$.

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The study of low-dimensional spin systems is an interesting theoretical and experimental problem in condensed matter physics [1]. Experimentally, the interest in these systems is related to the unusual magnetic properties of layered perovskites (quasi-two-dimensional compounds) such as Rb_2MnF_4, K_2NiF_4 [2,3], K_2MnF_4 [4] (easy-axis anisotropy), K_2CuF_4, $NiCl_2$, $BaNi(PO_4)_2$ [5] (easy-plane anisotropy), organic compounds [6], ferromagnetic films, multi layers, and surfaces [7]. Another interesting category of low-dimensional magnetic systems are the quasi-one-dimensional compounds containing chains of magnetic atoms with a weak interchain magnetic exchange, as for example the Sr_2CuO_3, Ca_2CuO_3 [8], $Sr_2V_3O_9$ [9], $BaCu_2Si_2O_7$ [10], $Sr_2Cu(PO_4)_2$ and $Ba_2\ Cu(PO_4)_2$ [11], and Cs_2CuCl_4 [12], where the magnetic properties are well theoretically described by a quasi-one-dimensional spin-1/2 Heisenberg antiferromagnet (HAF).

Most of the above real compounds exhibit a finite Néel critical temperature T_N, which is caused by a weak interchain (or interplane) exchange and/or anisotropy. For example, the Cs_2CuCl_4 quasi-one-dimensional compound has an orthorhombic crystal structure [12]. The spin-1/2 Cu^{2+} spins are coupled into chains running parallel to the b axis, with four such chains passing through each unit cell. The magnetic properties, such as the susceptibility, are consistent with the theoretical results [13] of a quasi-one-dimensional spin-1/2 HAF with an interaction $J = 0.34 \pm 0.02$ meV and chains coupled in the c direction by a small exchange $J' = 0.175$ meV. Below $T_N = 0.62$ K the spins order into a cycloid along the chain direction with an incommensurate wave vector $\vec{q} = (0,0.472,0)$; the incommensurate ordering is due to the frustration caused by the staggering of chains with respect to their neighbors. A small anisotropy confines the spins to rotate within a plane containing the b direction and making a small angle with the (b,c) plane. This is a typical realization of a system having weak interchain interactions. More details in this particular material can be found in Refs. [14,15] and references therein.

Bearing in mind the experimental results of such magnetic materials as a motivation for analytical models, a series of previous theoretical studies, done by several authors [16–20], have considered the ground-state magnetic properties of the quasi-one-dimensional spin-1/2 HAF, as well as isotropic and frustrated HAF models [21–25]. In particular, one basic problem is the influence of spatial anisotropy on the ground-state behavior of the staggered magnetization, m_s, of the two-dimensional HAF. Although in some materials (as described above for the Cs_2CuCl_4) the underlying lattice of interactions is an anisotropic triangular lattice (or more complex situations as treated in Refs. [26,27]), the model we are concerned with is described by the following Hamiltonian defined on an anisotropic simple square lattice as

$$\mathcal{H} = J \sum_{\langle i,j\rangle_x} \vec{\sigma}_i \cdot \vec{\sigma}_j + J' \sum_{\langle i,j\rangle_y} \vec{\sigma}_i \cdot \vec{\sigma}_j, \tag{1}$$

where $\langle i,j\rangle_{x(y)}$ denote nearest neighbors (NN) along the $x(y)$ direction on a square lattice, $\vec{\sigma}_i = (\sigma_i^x,\sigma_i^y,\sigma_i^z)$ is the Pauli vector spin operator at site i, $J > 0$ ($J' > 0$ or $J' < 0$) is the NN exchange interaction along the $x(y)$ direction. Here we define the anisotropy parameter $\lambda = |J'|/J \in [0,1]$. The model is symmetric for $\lambda > 1$.

For finite and positive values of the interchain exchange, $J' > 0$, the classical ground-state of the HAF is characterized by the system possessing an antiferromagnetic (AF) long-range order. Due to the quantum fluctuations the staggered magnetization is smaller than the saturated value, i.e., $m_s(\lambda) = 1/2 - \Gamma(\lambda)$, which continuously increases when λ increases. In the case of negative interchain exchange, $J' < 0$, we have a collinear state, which is characterized by a parallel spin orientation of NN in the vertical direction and an antiparallel spin orientation of NN in the horizontal direction, therefore exhibiting a Néel order of vertical chains along the x direction. Figure 1 shows schematically the AF and the CAF phases. In this case we denote this the Heisenberg collinear AF (HCAF) model. *A priori*, the inversion in sign of the

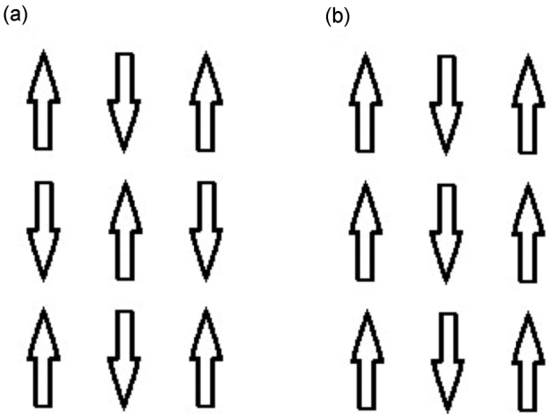

FIG. 1. The semiclassical ground-state of the spin-1/2 HAF model on an anisotropic square lattice. We have two ordered states: (a) antiferromagnetic $J' > 0$ (AF) and (b) colinear antiferromagnetic $J' < 0$ (CAF).

interchain exchange ($J' \to -J'$) could modify the value of the staggered magnetization, i.e., $m_s(-J') \neq m_s(J')$, as observed in quantum Monte Carlo results [28]. However, within the well-developed chain mean-field theory (see, for instance, Ref. [29]), the results are not sensitive to the sign of J'. In the isotropic square lattice limit ($\lambda = 1$) with $J' > 0$, the system exhibits an AF long-range order (LRO) at zero temperature with a finite value for $m_s(\lambda = 1) \simeq 0.3075$, while the one-dimensional isotropic limit ($\lambda = 0$) indicates an absence of LRO [$m_s(\lambda = 0) = 0$] [30]. The exact solution of the spin-1/2 HAF chain shows that the low-lying excitations are spin-1/2 objects (now called spinons) [31], quite different from standard spin waves. At finite temperature and dimension $d \leqslant 2$, the thermal fluctuations destroy the LRO of the isotropic HAF or HCAF [32].

There is still a quite basic question that has been intensively investigated by several authors regarding the presence of an order-disorder transition in model (1), i.e., the existence of a critical value $\lambda_c > 0$ where there occurs a quantum phase transition from a LRO ($\lambda > \lambda_c$) to a spin liquid state with pronounced AF short-range order (SRO) for $\lambda < \lambda_c$. For example, spin wave linear (SWL) theory [16] gives $\lambda_c = 0.034$ and the one-loop renormalization group analysis [33] found $\lambda_c = 0.047$. The series approach [20] with Padé approximants to Ising expansions for $m_s(\lambda)$ suggested that $\lambda_c \lesssim 0.02$, and field theoretical studies indicate $\alpha < 0.1$ [34]. On the other hand, some results [16,18,35–39] have found a null value $\lambda_c = 0$, which seems to be more plausible, because the ground state of the one-dimensional spin-1/2 HAF model is not gapped. Model (1) has also been solved using the mean-field approximation (MFA). The MFA is based on the Jordan-Wigner transformation of spin-1/2 operators to the Fermi ones with the subsequent mean-field treatment of the four-fermion term [40]. This method gives a coherent description of the ordered AF state and produces nontrivial quantitative predictions for static and dynamic quantities that have been successfully compared to experiments. In particular, the mean-field treatment [29,41] of the interchain ratio leads to $m_s(\lambda) \simeq \lambda^{1/2}$, up to logarithmic corrections. These relations are consistent with general scaling arguments [42]. On the other hand, the renormalized SW theory [43] predicts a behavior $m_s(\lambda) \simeq 1/\ln(1/\lambda)$, in clear contradiction with scaling. It can be seen that even the problem of the weakly coupled Heisenberg spin chains is indeed an unsolved fundamental task in condensed matter physics. It is not surprising that it has received a great deal of attention over the last five decades. It encapsulates the physics of the crossover from truly one-dimensional systems, dominated by quantum fluctuation effects, to two-dimensional, renormalized classical behavior.

In the present work we use a variational approach where the fluctuations around the classical ground states (AF and CAF phases) are treated by considering a trial vector state of the form $|\Psi_0\rangle = \prod_{l=1}^{N/4} |\phi_{0l}\rangle$, where N is the total number of spins on a two-dimensional lattice, $|\phi_{0l}\rangle$ is the state of a plaquette of four spins as depicted in Fig. 2, and the product is taken over the $N/4$ no overlapping plaquettes. The plaquette state is itself defined as a linear combination of the vector basis of the σ^z operator in such a way that $\sum_{i=1}^{4} \langle \sigma_i^z \rangle = 0$, namely $\{|1\rangle = |{}^{+}_{-}{}^{-}_{+}\rangle; |2\rangle = |{}^{-}_{+}{}^{+}_{-}\rangle; |3\rangle = |{}^{+}_{-}{}^{+}_{-}\rangle; |4\rangle = |{}^{-}_{+}{}^{+}_{-}\rangle; |5\rangle = |{}^{-}_{+}{}^{-}_{+}\rangle; |6\rangle = |{}^{+}_{+}{}^{-}_{-}\rangle\}$. Thus one has

$$|\phi_{0l}\rangle = \sum_{n=1}^{6} a_n |n\rangle_l, \tag{2}$$

where the coefficients $\{a_n\}$ are real variational parameters obeying the normalization condition $\sum_{n=1}^{6} a_n^2 = 1$. With this choice of vector states, the mean value of the spin operator in each site of the plaquette is given by $\langle \vec{\sigma}_i \rangle = m_i \hat{z}$, where $i = 1,2,3,4$ as in Fig. 2, $m_i = \langle \sigma_i^z \rangle = \langle \phi_{0l} | \sigma_i^z | \phi_{0l} \rangle$, and we are left with null components in the x and y directions.

Using the trial vector state defined in the Eq. (2), we obtain the following magnetizations at each site

$$\begin{aligned} m_1 &= 2(xu + yv - zw), \quad m_2 = 2(-xu + yv + zw), \\ m_3 &= 2(xu - yv + zw), \quad m_4 = 2(-xu - yv - zw), \end{aligned} \tag{3}$$

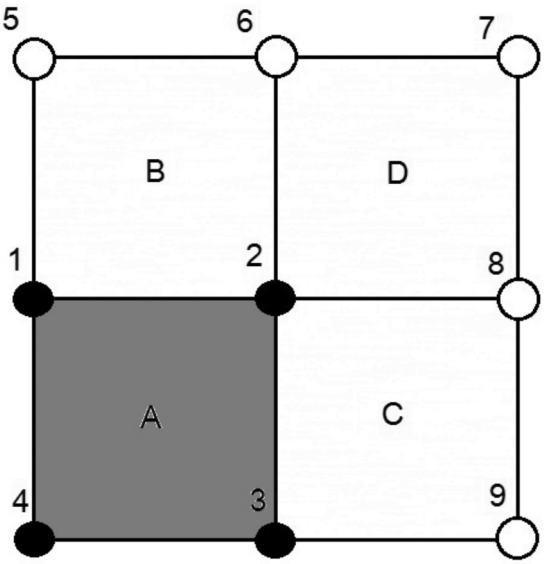

FIG. 2. Two-dimensional square lattice with a plaquette structure. The shadowed one is composed with the four $\vec{\sigma}_1$, $\vec{\sigma}_2$,$\vec{\sigma}_3$, and $\vec{\sigma}_4$ spin operators that are considered in Eq. (2).

where we have used the same set of parameters (canonical transformation) of Ref. [44], i.e., $x = (a_1 + a_2)/\sqrt{2}, y = (a_3 + a_5)/\sqrt{2}, z = (a_4 + a_6)/\sqrt{2}, u = (a_1 - a_2)/\sqrt{2}, v = (a_3 - a_5)/\sqrt{2}$, and $w = (a_4 - a_6)/\sqrt{2}$, which obey the normalization condition $x^2 + y^2 + z^2 + u^2 + v^2 + w^2 = 1$.

The ground-state energy per spin in units of J_1, $E_0 = \langle\Psi_0|\mathcal{H}|\Psi_0\rangle/4NJ_1$, is given by

$$\begin{aligned} E_0 = {} & \langle\vec{\sigma}_1.\vec{\sigma}_2\rangle_A + \lambda\langle\vec{\sigma}_2.\vec{\sigma}_3\rangle_A + \langle\vec{\sigma}_3.\vec{\sigma}_4\rangle_A + \lambda\langle\vec{\sigma}_4.\vec{\sigma}_1\rangle_A \\ & + \lambda\langle\vec{\sigma}_1\rangle_A \cdot \langle\vec{\sigma}_5\rangle_B + \lambda\langle\vec{\sigma}_2\rangle_A \cdot \langle\vec{\sigma}_6\rangle_B + \langle\vec{\sigma}_2\rangle_A \cdot \langle\vec{\sigma}_8\rangle_C \\ & + \langle\vec{\sigma}_3\rangle_A \cdot \langle\vec{\sigma}_9\rangle_C, \end{aligned} \tag{4}$$

where $\langle\mathcal{O}\rangle_\mu = \langle\phi_{0\mu}|\mathcal{O}|\phi_{0\mu}\rangle$ is the mean value of a given observable O calculated in the vector state of the $\mu(= A,B,C,D)$ plaquette as illustrated in Fig. 2.

The variational energy can be evaluated using the properties of the spin-1/2 Pauli operator components $\sigma^z|\pm\rangle = \pm|\pm\rangle$, $\sigma^x|\pm\rangle = |\mp\rangle$ and $\sigma^y|\pm\rangle = \pm i|\mp\rangle$, which can be expressed as

$$\begin{aligned} E_0 = {} & -\left(\frac{\lambda+1}{2}\right)(x^2+u^2) - 2(\lambda+1)x^2u^2 \\ & + \frac{(1-\lambda)}{2}[y^2+v^2-z^2-w^2] \\ & + 2(1-\lambda)(y^2v^2 - z^2w^2) + 2x(y\lambda + z). \end{aligned} \tag{5}$$

To obtain the minimum energy with the boundary condition given by normalization $x^2 + y^2 + z^2 + u^2 + v^2 + w^2 = 1$ we use the Lagrange multiplier method, which corresponds to the minimization of the functional

$$\begin{aligned} & \mathcal{F}(x,y,z,u,v,w,\eta) \\ & \quad = E_0 - \eta(x^2 + y^2 + z^2 + u^2 + v^2 + w^2 - 1), \end{aligned} \tag{6}$$

in such a way that $\delta\mathcal{F} = 0$. We then get

$$\begin{aligned} -(\lambda+1)x(1+4u^2) + 2(y\lambda + z) &= 2\eta x, \\ (1-\lambda)y(1+4v^2) + 2x\lambda &= 2\eta y, \\ -(1-\lambda)z(1+4w^2) + 2x &= 2\eta z, \\ -(\lambda+1)(1+4x^2)u &= 2\eta u, \\ (1-\lambda)(1+4y^2)v &= 2\eta v, \\ -(1-\lambda)(1+4z^2)w &= 2\eta w, \end{aligned} \tag{7}$$

where η is the Lagrange multiplier.

The equations above can be solved numerically and when several solutions are found the stable one will be that which minimizes the energy. In this way, we always find a disordered solution $m_1 = m_2 = m_3 = m_4 = 0$ with an energy higher than those for the ordered phases.

In the AF phase, $J' > 0$, we have the staggered magnetization defined by $m_s = (m_1 - m_2 + m_2 - m_4)/4$, where, in the absence of any external field, gives $m_1 = -m_2 = m_3 = -m_4 = 2xu$ [see Fig. 1(a)]. The solution in this case is given by $v = w = 0$ and $u,x,y,z \neq 0$. The solution for u,x,y, and z depends on λ and the staggered magnetization so obtained is shown in Fig. 3. Note that in Fig. 3 we have defined $m_A = m_s/2 = \langle S_i^z\rangle$, where $\vec{S}_i = \frac{\vec{\sigma}_i}{2}(\hbar \equiv 1)$. In the isotropic limit we have $m_A = 0.44$, which should be compared to a more rigorous result $m_A = 0.3075$ from Ref. [30]. As $\lambda \to 0$ the magnetization goes to zero as $m_s \sim \lambda^{1/2}$.

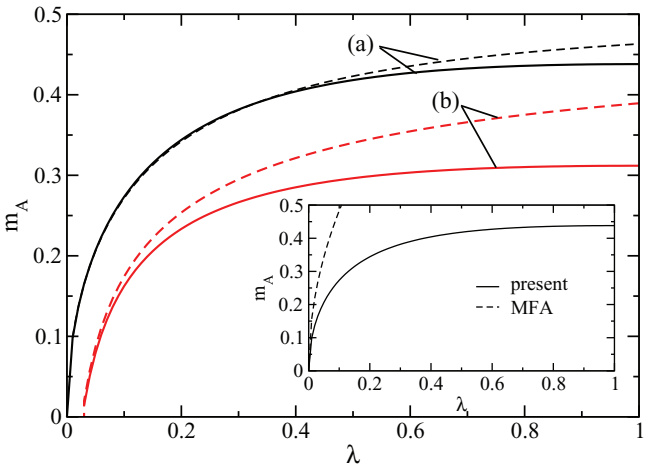

FIG. 3. (Color online) Staggered magnetization m_A as a function of interchain ratio λ of the quasi-one-dimensional spin-1/2 Heisenberg model obtained by using (a) variational method (present work), where the solid and dashed lines correspond to the AF and CAF states, respectively, and (b) spin wave linear theory [16]. The inset shows the comparison of our results (solid line) with those obtained by using a mean-field approximation (MFA) (dashed line) for the AF state [29].

On the other hand, in the CAF phase, $J' < 0$, we have $m_s = (m_1 - m_2 - m_3 + m_4)$, where $m_1 = -m_2 = -m_3 = m_4 = 2zw$ [see Fig. 1(b)]. The solution is now given by $u = w = 0$ and $v,x,y,z \neq 0$. The CAF staggered magnetization as a function of λ is also shown in Fig. 3 for the ferromagnetic interchain couplings. In the isotropic limit ($\lambda = 1$) we find $m_A = 0.46$ for the CAF states, which is higher than the value for the AF state, meaning that we have in fact $m_s(J' > 0) \neq m_s(J' < 0)$. In addition, a higher value of m_A in the CAF phase than in the AF phase has also been recently confirmed using a continuous-time Monte Carlo method [28]. In both cases, for any finite value of λ, the system is magnetically ordered, by a spontaneous breaking of the SU(2) spin symmetry. Our results for both signs of the transverse coupling, depicted in Fig. 3, have the same form at small λ given by $m_A \simeq \lambda^{1/2}$ as $\lambda \to 0$. This is the same critical behavior from the mean-field approach [29,41,42] in this limit, which is presented as a comparison in the inset of Fig. 3. This is not surprising because the variational approach used in this work corresponds to a mean-field-like approach. Our results for the AF and CAF staggered magnetizations deviate from the expected form at values below $\lambda \simeq 0.40$. The model (1) with AF interchain coupling was also studied by Sandvik [18] using quantum Monte Carlo simulation within a multichain mean-field theory, and reveals strong logarithmic corrections to the square-root dependence, $m_A \simeq \lambda^{1/2}\ln^{1/3}(\lambda)$. This logarithmic correction was not observed in the variational approach developed in this work. Also presented in Fig. 3 are the results obtained using SWL theory [16] that found a finite critical value $\lambda_c = 0.034$.

In summary, the present simple approach for the quasi-one-dimensional antiferromagnetic Heisenberg model is able to provide a zero critical value for the anisotropic exchange ratio $\lambda_c = 0$, as should be expected for this system [16,18,35–39], and a higher staggered magnetization in the CAF phase than in

the AF phase, in agreement to Monte Carlo simulations [28]. Although the asymptotic limit of the staggered magnetization m_A is the same as the MFA, its global behavior on λ is much better than the mean-field ones, as can be noted in the inset of Fig. 3. We can argue that the present results indeed reveal the true behavior of the AF and CAF spin-1/2 quasi-one-dimensional Heisenberg model.

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