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The two-dimensional ($2D$) Heisenberg model with anisotropic exchange ($\Delta = 1 - J_x/J_z$) and with negative next nearest neighbor exchange (J_2) with $S = 1/2$ is investigated by using the quantum-mechanical Monte-Carlo method. The energy, magnetic moment at a site, heat capacity, and spin-spin correlation functions are calculated. The stability regions for Néel ordering of spins as well as the strip-phase and gapless quantum spin liquid are determined in the interval $\Delta/2 \leqslant J_2/J_1 \leqslant 1/(2\Delta)$. © *1998 American Institute of Physics.* [S1063-777X(98)00808-1]

Over the recent years, a large number of publications have been devoted to the theoretical and experimental studies of $2D$ Heisenberg antiferromagnets (AF) in a square lattice with spin $S = 1/2$. Following the discovery of high-temperature superconductivity in metal oxides, considerable attention has been paid to the investigations of frustrated antiferromagnets with a negative next nearest neighbor interaction. Several elegant theories have been proposed for new types of magnetic states. These are the resonant valence bond (RVB) states proposed by Anderson,[1] which are formed as a result of superposition over all realizations of singlet pairs. Calculations for a 4×4 lattice[2] confirm the closeness of the energies E_{RVB} to the exact value. The N-Fermi approach, in which N different flavors are introduced instead of the two projections of the electron spin S,[3,4] leads to a 1/2-flux state with gapless spin excitation and a power attenuation of the spin–spin correlation function. In the strong frustration region $J_2/J_1 \sim 0.6$, where J_α is the energy of antiferromagnetic exchange interaction between nearest or next nearest neighbors, the existence of gap magnetic states is assumed with an energy gap $\sim (0.1-0.2)J$. These states include that of a quantum spin liquid (SL) with scalar and vector chiral ordering,[4,5] and an ordered dimeric (spin-Peierls) state.[6,7] in the $1/N$ approximation (N is the number of flavors),[6] the energy of the dimeric state is slightly lower than the energy of the chiral spin state: $E_{\mathrm{chiral}}/E_{\mathrm{dimer}} = 0.994$. The exact diagonalization in small lattices[8] gives opposite results: the vector chiral order parameter is double the dimeric parameter. The latter states are described by fractional statistics and have a specific spectrum of spinon, holon, and anyon excitations described in detail by Izyumov *et al.*[9]

Most works devoted to investigation of the Heisenberg model with frustrations assume a long-range AF order at $T = 0$ and $J_2 = 0$. The critical value of J_2/J_1 corresponding to the violation of long-range magnetic order depends on the technique and approximations used in analytic computations and varies between 0.1 and 0.4.[7,10–12] The more precise the uncoupling of spin correlation functions, the smaller the critical value for the frustrated bond. For values of J_2/J_1 in the interval 0.6–0.8, a four-sublattice AF is formed.[7,11] All these investigations are based on Heisenberg's isotropic model although quasi-two-dimensional compounds have an exchange anisotropy $\sim (10^{-4}-10^{-2})J$. For example, CaV_3O_7 with an exchange $J_2/J_1 \approx 1.3$ has a temperature-independent static structural factor $S(q)$ above the Néel temperature.[13] The anisotropy lowers quantum fluctuations and minimizes computational error associated with the finite size of the lattice.

In an earlier work,[14] we studied the dimeric state in an antiferromagnetic Heisenberg chain with four-spin interaction and with a spin $S = 1/2$. The four-spin exchange may be a result of interaction of spins with lattice vibrations (phonons). Expanding the exchange integral in powers of atomic displacements, we obtain the spin–phonon interaction in first-order perturbation theory. Transforming the phonon operators through a displacement by a certain constant[15] determined by the condition of vanishing of terms linear in phonon operators, we obtain the four-spin exchange. Thus the bilinear and four-spin exchange correspond effectively to a spin-Peierls system with a spin-Peierls phase transition in the one-dimensional Heisenberg model.[16] Such a transition corresponds to the formation of spin pairs that are in singlet states with ordered centers of mass relative to one another. In this work, we study the properties of the dimeric state and calculate the dimeric state–paramagnet phase transition temperature as a function of the four-spin interaction. The transition vanishes in the absence of a four-spin interaction, and the calculated properties for the antiferromagnetic chain are in good agreement with the results of computations, e.g., the correlation radius varies according to a power law $\xi = A/T$ in the entire range of temperature T.

Thus, we can single out two problems. The first one concerns the possibility of existence of a quantum spin liquid in an anisotropic frustrated AF and the effect of exchange anisotropy on the stability region of long-range antiferromagnetic order and the quantum spin liquid. The second problem concerns the existence of an energy gap in the excitation spectrum of a quantum spin liquid. The value of the

gap in the SL can be determined from the temperature dependence of heat capacity, susceptibility, and from the dependence of magnetization on the applied magnetic field. We shall use the trajectory algorithm[17] in the Monte-Carlo (MC) method. The basic idea underlying this algorithm is the transformation of a quantum D-dimensional problem into a classical $(D+1)$-dimensional problem by introducing ''time sections'' in the space of imaginary time $0<\tau<1/T$, and the realization of the MC procedure in the ''imaginary time-coordinate'' space.

1. MODEL AND COMPUTATIONAL TECHNIQUE

Let us consider an anisotropic Heisenberg AF with the next nearest neighbor antiferromagnetic interaction in a square lattice with spins $S=1/2$ localized at its sites and directed along the Z-axis coinciding with the direction of the applied field. The Hamiltonian can be presented in the form

$$H=-\sum_{\alpha=1}^{2}\sum_{h_a=1}^{4}\sum_{i=1}^{N}\{J_{\alpha}^{zz}(h_\alpha)S_i^zS_{i+h_\alpha}^z + J_{\alpha}^{x,y}(h_\alpha)(S_i^xS_{i+h_\alpha}^x+S_i^yS_{i+h_\alpha}^y)\} - H^z\sum_{i=1}^{N/2}(S_i^z+S_{i+h_\alpha}^z), \quad (1)$$

where $J\alpha<0$, the summation over h_α is carried out over all neighbors in the αth coordination sphere, $\Delta=1-J^{x,y}/J^z$ is the ''easy axis'' type exchange anisotropy, H^z is external magnetic field, and N the total number of spins.

The algorithm and the MC method were described in detail in an earlier publication.[18] The Hamiltonian is divided into clusters of four spins on a plaquette, and commutation between them is taken into consideration using Trotter's formula. In the MC procedure adopted in our work, we use periodic boundary conditions along Trotter's direction and over the lattice. The linear dimensions of the lattice are $L=40$, 48, 64 and 80, and $m=16$, 32, 48 (where m is a positive integer called Trotter's number). The number of MC steps per spin varies from 3000 to 10000. One MC step involves the rotation of all spins in a lattice of size $L\times L\times 4m$. The energy E and the heat capacity C are defined by formulas

$$E=\left\langle (1/2)\sum_{i,j,r}F_{i,j}^r\right\rangle,\quad F_i^r=-\partial/\partial\beta(\ln\rho_i^r),\quad C=dE/dT. \quad (2)$$

Here $\rho_{i,j}^r$ are matrix elements of the local density matrix $(i,j=1,\ldots,L,\ r=1,\ldots,m)$, and $\beta=1/(k_BT)$. Summation is carried out over eight spin clusters $L\times L\times m$, and the angle brackets indicate thermodynamic averaging. Magnetization M and longitudinal susceptibility χ are defined as

$$M=\left\langle\sum_{i,j,r}M_{i,j}^r\right\rangle,$$

$$M_{i,j}^r=\frac{1}{4m}\sum_{h_x h_y=0}^{1}(S_{i+h_x,j+h_y}^r+S_{i+h_x,j+h_y}^{r+1}),$$

$$(S_i=\pm 1/2),\quad \chi=M/H. \quad (3)$$

We calculated the longitudinal $R(r)=\langle S_0^zS_r^z\rangle$ spin-spin and four-spin $\langle S_0^zS_1^zS_r^zS_{r+1}^z\rangle$ correlation functions and their Fourier transform along the sides and diagonal of a square lattice. The thermodynamic mean of the spin at a site is defined as $\sigma=\lim_{r\to\infty}[\mathrm{abs}(\langle S_0^zS_r^z\rangle)]^{1/2}$. The correlation radius ξ of spin interaction and the pre-exponential index η are defined through the relation

$$R(r)=A/r^{\eta}\exp(-r/\xi), \quad (4)$$

where $R(r)$ is the normalized correlation function $R(r)=\langle S^z(0)S^z(r)\rangle-\langle S^z\rangle^2$. In the model with competing interactions in the absence of a magnetization-type order parameter, a possible characteristic of the system may be the correlation function of locally calibrated operator over an elementary plaquette. These plaquettes may turn out to be ordered, and the type of this order may be determined from the eight-spin correlation function

$$\langle S^z(0)S^z(h_x)S^z(h_y)S^z(h_x+h_y)S^z(r)S^z(r+h_x)\times S^z(r+h_y)S^z(r+h_x+h_y)\rangle. \quad (5)$$

Chiral order may exist in the region of strong frustrations $(J_2\sim 0.5J_1)$. Let us determine the vector parameter of chirality over the smallest triangle

$$\mathbf{F}_{ijk}=(\mathbf{S}_i\times\mathbf{S}_j+\mathbf{S}_j\times\mathbf{S}_k+\mathbf{S}_k\times\mathbf{S}_i) \quad (6)$$

and calculate its z-projection F^z

$$F_{ijk}^z=i[(S_i^+S_j^--S_i^-S_j^+)+(S_j^+S_k^--S_j^-S_k^+)+(S_k^+S_i^--S_k^-S_i^+)]. \quad (7)$$

We calculate the correlation functions of normal and anomalous types of spin operators $\langle S^+(0)S^-(r)\rangle$, $\langle S^+(0)S^+(r)+S^-(0)S^-(r)\rangle$. by using the Hirsch technique.[19] The idea underlying this technique is that the world lines are ruptured at a distance $r=m$ in the Trotter direction, and the wave functions in the S^z representation become equal at this distance. The computation of these correlations requires a new MC procedure with free boundary conditions in the Trotter direction and a doubling of the computation time.

The statistical error in MC computation was determined by using the standard technique. The mean value was computed, the instantaneous value memorized, and the mean square deviation determined after completion of the MC procedure. This error lies in the interval (0.1–2)%. The systematic error arises due to a finite value of the number m and is proportional to $\sim A/(mT)^2$.

In a frustrated AF, the matrix elements in a 16×16 matrix corresponding to a four-spin cluster on a plaquette may assume negative values. However, the probability of local and closed rotations is an even function of negative matrix elements. The probability of global rotations is equal to the product of matrix elements from 1 to m, which may be negative. The statistical weight of these configurations is small and is determined as follows. The number of configurations in the space of negative (positive) weights $Z_-(Z_+)$ is determined by the MC procedure, and the quantity $Z_-/(Z_+$

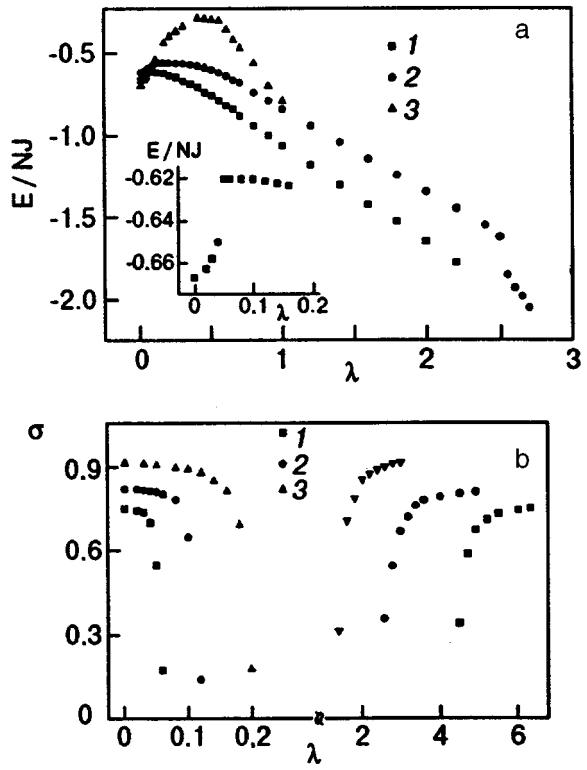

FIG. 1. Dependences of the energy E/NJ of an anisotropic AF with $\Delta=0.05$ (curve *1*), 0.2 (curve *2*), and an isotropic AF[11] (curve *3*) ($\Delta=0.1$ in the inset) (a) and of the magnetization σ at the site for an AF with $\Delta=0.1$ (curve *1*), 0.2 (curve *2*), and 0.4 (curve *3*) (b) on the normalized next nearest neighbor exchange $\lambda=J_2/J_1$.

$+Z_-)=0.02-0.044$ decreases with increasing temperature. This leads to a systematic error of the same order of magnitude in the computed values:

$$\langle A\rangle \approx A_+\{1+Z_-/(Z_+-Z_-)\}. \tag{8}$$

Here A_+ is the sum of physical quantities in the space of positive weights. To improve convergence of thermodynamic means, the sign of the statistical weight of the configuration was not taken into consideration. This is valid if the ratio $(Z_+-Z_-)/(Z_++Z_-)$ tends to a constant value as $T\to 0$.[20] In the region of strong frustrations ($J_2/J_1=0.5$), the difference in the values of energy calculated by taking into account the sign of transition probability $W>0$ and without it abs(W) is $\sim 5\%$.

2. DISCUSSION OF RESULTS

While calculating the thermodynamic characteristics of an anisotropic AF with frustrated next nearest neighbor exchange in the ground state, we shall use the technique of asymptotic continuation of these quantities (calculated at finite temperatures) to $T=0$. The dependence of energy, magnetization at a lattice site, and spin correlation functions on the next-nearest neighbor exchange was calculated for several values of exchange anisotropy $\Delta=0.05$, 0.1, 0.2, 0.3, 0.4, 0.6, 0.75 and 0.8. The critical values of $\lambda_{c1,2}$ corresponding to the vanishing (emergence) of long-range order are determined from the kinks on the energy dependence $E(\lambda)$ and the vanishing of magnetization at a lattice site (Fig. 1).

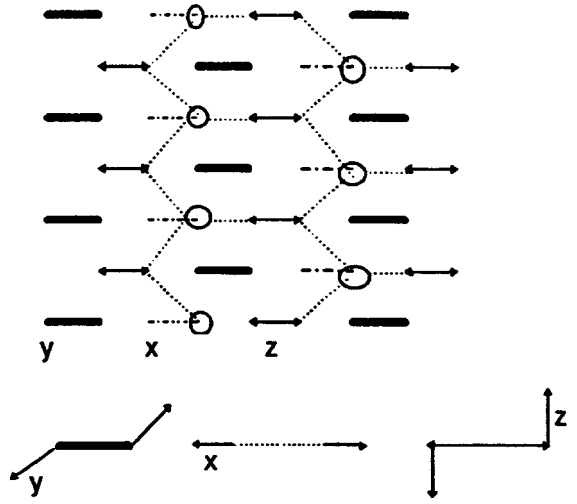

FIG. 2. Magnetic structure of a spin liquid: spin pairs directed along the X-axis (dashed line), Y-axis (solid line), and Z-axis (arrowed line). The dotted line indicates correlations along the longitudinal spin components.

Frustration decreases the absolute value of the energy of an AF by 5–8%. For $\lambda>\lambda_{c1}$, a dimeric state is formed in which dimers are arranged in a particular order. Such a structure can be presented schematically in the form of three types of dimers with a mutually orthogonal arrangement. The fine lines in Fig. 2 show pairs of spins directed along the X-axis, while the thick lines and lines with double arrows indicate respectively the spin pairs directed along the Y- and Z-axes. Spins directed along the Z-axis induce polarization along the longitudinal spin components on the nearest neighbors marked by circles in Fig. 2. The spin–spin correlations along longitudinal components are shown by dashed lines. In this region, the correlation functions along the diagonal of the square [110] at a distance $r=\sqrt{2}a$ are negative for longitudinal components and zero along transverse components as shown in Fig. 2.

Figure 3a shows the spin correlation functions varying with the exchange J_2 in accordance with the magnetic structure presented in Fig. 2. For the critical value $\lambda_1=(1+\Delta)/4$, the sign of $\langle S^z(0)S^z(r=\sqrt{2})\rangle$ varies from positive to negative, and the signs of spin correlation functions of nearest and next nearest neighbors coincide with the signs of the exchange, and the frustration disappears. This leads to an increase in the absolute value of energy. The theoretical value of the energy of the disordered state exceeds all energy values obtained by other methods. Thus, for $\lambda=1/2$ the exact diagonalization gives $E/J=-0.53$,[21] the spherically-symmetric spin-wave theory gives $E/J=-0.26$,[11] while the $1/N$- fermion representation gives $E/J=-0.23$.[4]

Upon a further increase in the exchange J_2, the short-range order in transverse spin components along the diagonal decreases by $r=\sqrt{2}a$ and becomes equal to zero $\langle S^+(0)S^-(r=\sqrt{2})\rangle\approx 0$ (Fig. 3b) for $\lambda=\lambda_2$. Spin correlation functions have a power dependence on distance $\langle S^z(0)S^z(r)\rangle\sim 1/r^\eta$, and can be approximated quite well by a straight line on the logarithmic scale (Fig. 3c) where the exponent η varies in the interval 2.5–3.5 for $\lambda_1<\lambda<\lambda_2$. The chirality parameter calculated over the entire lattice ac-

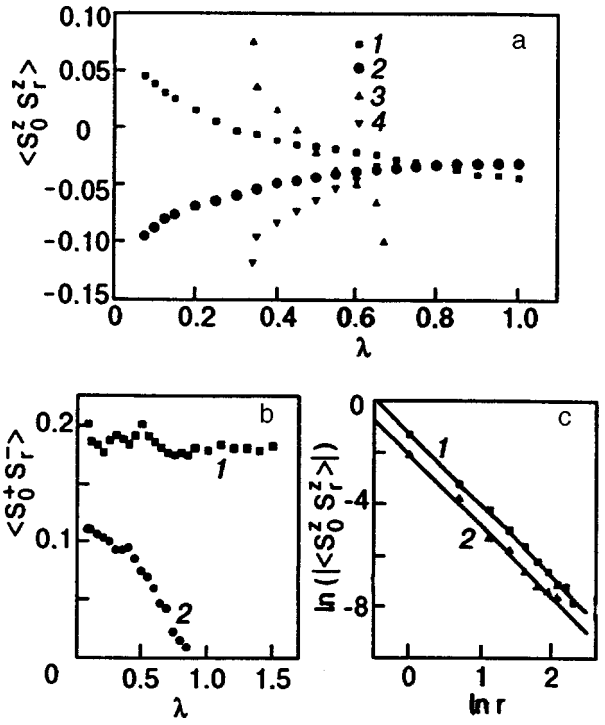

FIG. 3. Spin–spin correlation functions along the longitudinal spin components $\langle S_0^z S_r^z\rangle$ in an SL with $\Delta=0.1$ (*1,2*), 0.75 (*3,4*) at $r=1$ (*2,4*), $\sqrt{2}$ (*1,3*) (a) along transverse spin components $\langle S_0^+ S_r^-\rangle$ with $\Delta=0.1$, $r=1$ (*1*), $r=\sqrt{2}$ (*2*) (b), (c) shows the dependence of the correlation function of an SL with $\Delta=0.05$, $\lambda=0.25$ (*1*), 0.8 (*2*) on logarithmic scale.

cording to formula (8) is equal to zero ($F=0$), i.e., there is no chiral order. The proposed[7] ordering of dimers over sublattices embedded into one another in a staggered manner can be calculated by using the four-spin correlation function defined in these sublattices. The MC computation of four-spin correlation functions along the sides, diagonals and sublattices $\langle S_0 S_1 S_r S_{r+1}\rangle \approx 0$ for $r=L/2$, and does not lead to a long-range order of singlet pairs. There is no correlation between spin plaquettes described by formula (5). At distances $r>a$, the function (5) tends to zero. Hence the long-range order parameter for chiral and singlet ordering of pairs is equal to zero in this state.

The dependence of magnetization on the external magnetic field is linear (Fig. 4c). the critical field $H_c=0$, and hence there is no energy gap between the ground and triplet excited states. The susceptibility is independent of temperature and applied magnetic field in SL. The temperature dependence of heat capacity can be approximated quite well by a power law $C(T)\sim AT^n$, where the exponent n varies between 2 and 3 depending on the value of the exchange J_2. For an exchange anisotropy $\Delta=0.05$, $J_2/J_1=3$ in an SL, the MC results are approximated correctly by a power dependence $C(T)\sim AT^{2.7(2)}$ (Fig. 4a). It was mentioned above that the negative sign of the matrix elements of transition probability leads to a small error of $\sim 3\%$ in the energy values for $\lambda=0.3$. Figure 4b shows the energy values calculated by taking into account the sign of the transition probability (curve *1*) and without taking the sign into consideration (curve *2*). The computational error decreases with increasing temperature.

Let us emphasize the basic characteristics of this state. The thermodynamic mean of the spin is equal to zero, a short-range order exists with a sharp attenuation of spin correlation functions according to a power law, and the energy gap between the ground state and the excited state is equal to zero, which corresponds to the definition of a gapless quantum spin liquid.[22] The magnetic properties of a quantum spin liquid are analogous to those of a ''tomographic'' Luttinger liquid for the two-dimensional case.[23] The existence of a gapless SL in the $2D$-Heisenberg model with frustrations is in accord with the general theorem of Lieb, Schultz and Mattis[24] applied to the two-dimensional case.[25] According to this theorem, the disordered phase of an antiferromagnet containing an arbitrary half-integral spin in a unit cell must have a broken symmetry or gapless excitations.

The energy of an SL in the isotropic case can be presented in the form of the energy of singlets in J_1 and the energy along longitudinal spin components with J_2 (the Ising component $\sim 2S^2J_2$): $E=-0.687-0.5\lambda$, which is in good agreement with the MC results for $\lambda>\lambda_2$ where the correlation functions along the longitudinal components become equal at distances $r=1$ and $r=\sqrt{2}$ (Fig. 3a). The exponent η begins to decrease with increasing exchange J_2 to $\eta\approx 1$ for

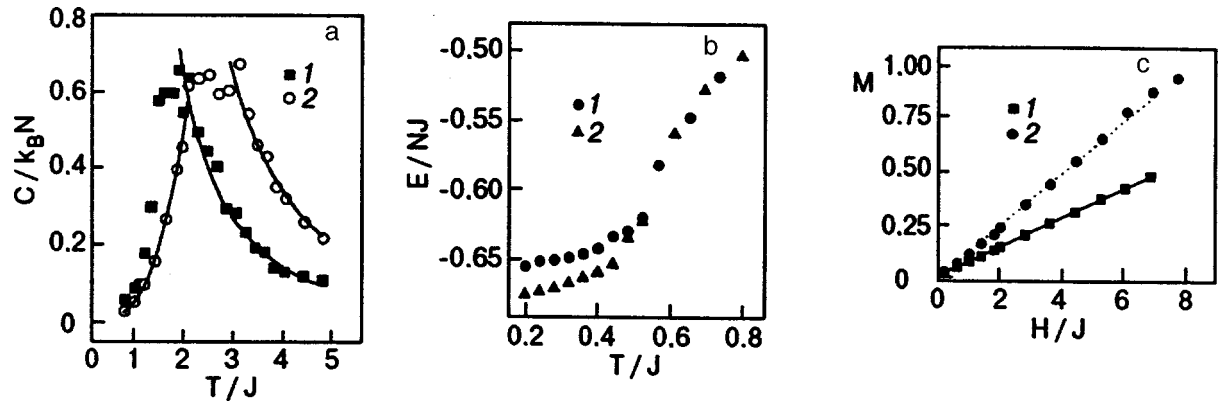

FIG. 4. (a) Temperature dependence of the heat capacity C/k_BN of a spin liquid for $\Delta=0.05$, $\lambda=2$ (curve *1*) and 3 (curve *2*); (b) temperature dependence of energy E/NJ calculated without taking (curve *2*) and taking (curve *1*) into consideration the sign of transition probability for $\Delta=0.05$, $\lambda=0.3$; (c) dependence of the magnetization M on the applied field in a spin liquid with $\Delta=0.05$, $\lambda=1$ (curve *1*) and 2 (curve *2*).

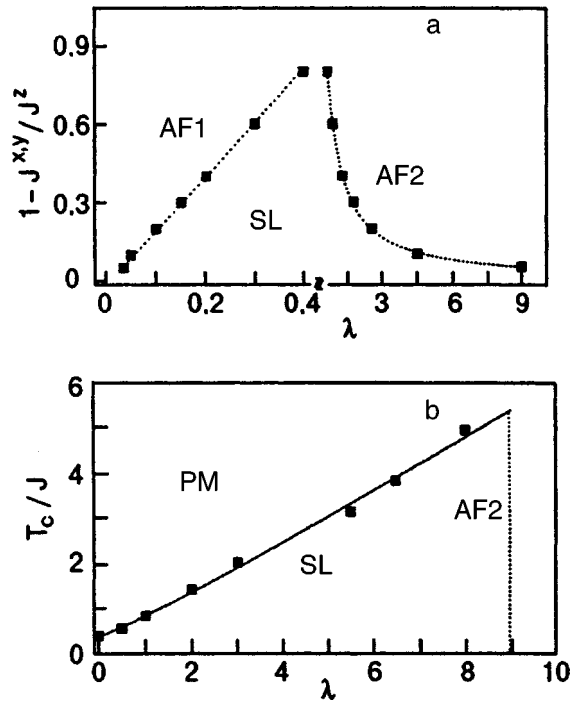

FIG. 5. Phase diagram of the ground state of a Néel antiferromagnet (AF1), strip-phase (AF2) and quantum spin liquid (SL) in the coordinates exchange anisotropy vs. normalized nearest neighbor interaction (a), and of a paramagnet (PM), AF2 and SL in the temperature vs. λ coordinates for $\Delta = 0.05$ (b).

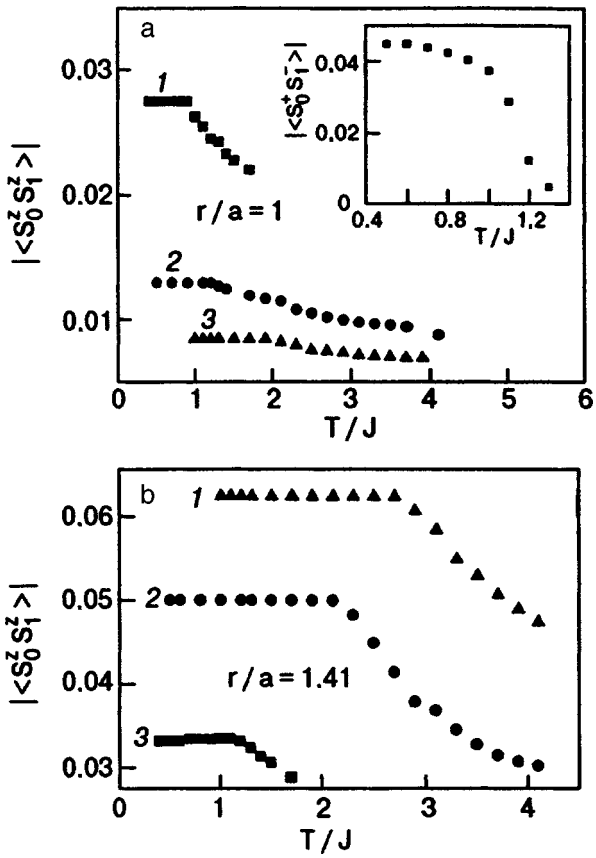

FIG. 6. Temperature dependence of spin correlation functions along longitudinal components in a spin liquid with $\Delta = 0.05$, $\lambda = 1$ (*1*), 2 (*2*), and 3 (*3*) at a distance $r/a = 1$ (a) and $\sqrt{2}$ (b). The inset shows the same dependence for $r/a = 1$ with $\lambda = 2$ along the transverse components.

$\lambda = \lambda_{c2}$. A four-sublattice AF structure obtained by the method of exact diagonalization[12,21] or analytically[11,26] can be singled out over several nearest neighbors. This structure is formed by two spin sublattices with a staggered ordering and a unit cell size $\sqrt{2}a$ embedded one into the other. On account of an inadequate consideration of spin correlations in analytic computations and the small size of lattice in an exact diagonalization, the variation in the short-range order for $\lambda_1 = (1+\Delta)/4$ and λ_2 is naturally associated with the violation of the Néel state and the emergence of a strip phase (AF2) with a ferromagnetic ordering of spins along one side of a square, and an antiferromagnetic ordering along the other side. The strip phase, determined from the Fourier spectrum $\langle S_{-q}S_q\rangle$ in MC computations, is degenerate for two vectors $\mathbf{Q}(0,\pi/a)$ and $\mathbf{Q}(\pi/a,0)$, and a spontaneous breaking of symmetry occurs along one of these vectors for $\lambda \geqslant \lambda_{c2}$. The interpolation of the critical values λ_{c1} and λ_{c2} calculated by the MC technique leads respectively to a linear $\lambda_{c1} = \Delta/2$ and inverse $\lambda_{c2} = 1/2\Delta$ dependence on exchange anisotropy. An asymptotic continuation for the isotropic case reveals the absence of a long-range order of the strip-phase type. Figure 5a shows the phase diagram of the ground state of an anisotropic Néel antiferromagnet (AF1), strip phase (AF2), and of a quantum spin liquid. Only one line corresponding to the isotropic case $\Delta = 0$ was investigated earlier. Our computations reveal the absence of a long-range order for all values of the exchange $J_2 < 0$, and two types of short-range order for $\lambda < 0.25$ and $\lambda > 0.75$ with a power dependence of the spin correlation functions on distance. In the interval $0.25 < \lambda < 0.75$, the spin–spin correlation function decreases sharply with distance: $\langle S^z(0)S^z(r)\rangle \sim 1/r^{3.5(3)}$.

For the order parameters in a quantum spin liquid, we can take correlation functions at distances $r = 1, \sqrt{2}$. The correlation functions along the longitudinal components are independent of temperature (in contrast to those along the transverse components). Since the excitations in SL are spinons, a singlet pair is transformed into a triplet with $S^z = 0$. The static structural factor $s(Q)$, viz., the Fourier harmonic of the pair correlation function at $Q = \pi/a$ for $\lambda < \lambda_2$ and $Q = \pi/(\sqrt{2}a)$ for $\lambda > \lambda_2$, has a temperature dependence analogous to the dependence $\langle S^z(0)S^z(r = 1,\sqrt{2}a)\rangle$ shown in Fig. 6. Two temperature transitions appear in the spin liquid for $\lambda > \lambda_2$. The first transition is associated with the violation of the dimeric order at $T = T_{1c}$, where the relation $\langle S^+(0)S^-(r=1)\rangle - \langle S^z(0)S^z(r=1)\rangle$, characteristic for an AF with Néel ordering of spins is satisfied. The developed long-range magnetic order in longitudinal components shown by a dashed line in Fig. 2 is preserved for $T > T_{1c}$, and is transformed into a paraphrase at $T = T_{2c}$. The spin correlation function $\langle S^z(0)S^z(r=\sqrt{2})\rangle$ has a singularity at this temperature (Fig. 6b), and the dependence of the spin–spin correlation function on distance changes from power to exponential. In the temperature interval $T_{1c} < T < T_{2c}$, the heat capacity (Fig. 4) and susceptibility have their maximum values. For $T > T_{2c}$, the temperature dependence of heat capacity is analogous to that for a paramagnet:

$C(T) \sim A/T^2$ (Fig. 4). Figure 5b shows the range of existence of the spin liquid on the temperature-exchange (normalized to next nearest neighbor) plane. The long-range magnetic order[13] is formed in CaV_3O_7 due to a weak interaction between planes. Above $T_N = 2$ K, the magnetic static structural factor $s(Q)$ is independent of temperature up to $T = 40$ K, and has a noncommensurate vector $\mathbf{Q} \sim 0.7\pi(1,1)$ in the basal plane. A quantum spin liquid is probably formed in this temperature interval.

Summarizing the results of this research, we can conclude that for the anisotropic frustrated $2D$-Heisenberg model with antiferromagnetic interactions, a gapless spin liquid exists in the interval $\Delta/2 < \lambda < 1/2\Delta$. For $\lambda > (1+\Delta)/4$, the spin-spin correlation functions for the nearest and next-nearest neighbors are negative and frustration vanishes. Long-range chiral and dimeric order do not exist in a quantum spin liquid. The spin–spin correlation function has a power dependence on distance in SL.

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