Quantum Monte Carlo investigation of the $2D$ Heisenberg model with $S=1/2$

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The two-dimensional ($2D$) Heisenberg model with anisotropic exchange ($\Delta=1-J_x/J_z$) and $S=1/2$ is investigated by the quantum Monte Carlo method. The energy, susceptibility, specific heat, spin-spin correlation functions, and correlation radius are calculated. The sublattice magnetization (σ) and the Néel temperature of the anisotropic antiferromagnet are logarithmic functions of the exchange anisotropy: $1/\sigma+1+0.13(1)\ln(1/\Delta)$. Crossover of the static magnetic structural factor as a function of temperature from power-law to exponential occurs for $T_c/J\approx 0.4$. The correlation radius can be approximated by $1/\xi=2.05T^{1.0(6)}/\exp(1.0(4)/T)$. For La_2CuO_4 the sublattice magnetization is calculated as $\sigma=0.45$, the exchange is $J=(1125-1305)$ K; for Er_2CuO_4 $J\sim 625$ K and the exchange anisotropy $\Delta\sim 0.003$. The temperature dependence of the static structural magnetic factor and the correlation radius above the Néel temperature in these compounds can be explained by the formation of topological excitations (spinons). © *1999 American Institute of Physics.* [S1063-7834(99)02401-6]

Many theoretical works on the two-dimensional ($2D$) Heisenberg antiferromagnet (AF) with a square lattice with spin $S=1/2$ have appeared in the last few years. Substantial theoretical efforts have been made to confirm or reject Anderson's idea of a quantum disordered ground state[1] in the $2D$ Heisenberg model with antiferromagnetic interactions. In a number of works, the critical spin below which there is no long-range AF order at $T=0$ was found to be $S_c=1-3/2$.[2,3] Chakravarty, Halperin, and Nelson,[4] using the renormalization-group method and the nonlinear σ model in 2 + 1 space, obtained long-range AF order (LRO) in the ground state. They neglected the Hopf term $h\theta H_{hopf}(\theta=2\pi S)$,[5] which can change the magnetic state substantially. The theories with topological invariants[6] do not assume long-range order in the ground state. A similar result is obtained in the monograph Ref. 7 where, on the basis of the Lieb, Schultz, and Mattis theorem,[8] it is asserted that the two-dimensional spin-1/2 Heisenberg antiferromagnet possesses a ground state which may turn out to be non-Néel and can have either broken translational symmetry or gapless excitations.

Most exact calculations for small lattices 4×4 and 4×6 give AF ordering[9–11] with energy $E/NJ=0.67-0.7$. Advances in computational technology make it possible to use large lattices with $N=26$ and 32,[12,13] which give asymptotic vanishing of the long-range antiferromagnetic order as $\sim 1/N$. Monte Carlo (MC) modeling makes it possible to use lattices ranging in size from 20×20 to 128×128.[14–18] These calculations, which use a variational algorithm[17] and the renormalization group,[19,20] likewise give the existence of $2D$ AF and $T=0$. The type of magnetic state calculated on the basis of variational methods depends on the choice of the initial (trial) wave function and, in some cases, it gives no long-range order in magnets. Trajectory MC methods[15,16,21] employ finite temperatures, and it is difficult to draw any conclusions about the ground state based on an investigation of an isotropic model only.

A number of problems also arise in investigations of the quasi-two-dimensional compounds La_2CuO_4,[22,23] Er_2CuO_4,[24–26] and $Sr_2CuO_2Cl_2$.[27] Experimental investigations of these crystals indicate ionic bonds,[28] while the ordered moment falls in the range $\sigma=0.4-0.44$, which is much less than the theoretical estimates for $2D$ Heisenberg AF $\sigma=0.6-0.64$.[4,29] Above the Néel temperature, up to $(1.5-2)T_N$, there exist strong two-dimensional spin correlations so that the correlation radius varies exponentially, for example, in La_2CuO_4 from 40 Å at 450 K up to 400 Å at 200 K,[23] while the static magnetic structure factor $S(Q)$ varies with temperature very slowly, which also does not agree with theoretical calculations predicting exponential behavior $S(Q)\sim T^2\exp(-A/T)$.[4] In the isostructural compounds La_2NiO_4[30] and La_2CoO_4[31] with spin $S=1/2$ and 3/2, a two-dimensional Ising phase transition is observed in the neutron scattering intensity near $1.02T_N$, in complete analogy with K_2NiF_4 which possesses magnetic anisotropy of the same order of magnitude as La_2CuO_4. The magnetic susceptibility $\chi(T)$ in these compounds[26] at temperatures $T>T_N$ demonstrates anomalous temperature behavior — a very smooth temperature dependence in the region $T_N<T<(1.5-2)T_N$, and an inflection in the temperature dependence $\chi(T)$ at the limit of this interval. Therefore the magnetic moment of the copper ion and the temperature behavior of the spin correlations above T_N are unique and cannot be explained on the basis of existing theoretical calculations in a $2D$ Heisenberg model with $S=1/2$.

Thus, two important problems can be identified from the review presented above. The first problem is the unknown nature of the ground state of a $2D$ AF. Quantum fluctuations that can completely destroy long-range order are very substantial because of the low dimension of the space and the

low spin. The second problem is to explain using the Heisenberg model with nearest-neighbor interaction and to estimate the magnetic moment, temperature dependence of the susceptibility, and static magnetic structure factor $S(Q)$ for $T > T_N$ in quasi-two-dimensional compounds based on copper ions with spin $S=1/2$ without using additional interactions.

In the present paper we use the quantum Monte Carlo method employing a trajectory algorithm.[32] The basic idea of the algorithm is to convert the quantum D-dimensional problem to a classical $(D+1)$-dimensional problem by introducing ''time'' cuts in imaginary time space $0<\tau<1/T$ and to implement an MC procedure in the ''imaginary time — coordinate'' space.

1. MODEL AND METHOD

Let us consider a Heisenberg anisotropic AF with antiferromagnetic nearest-neighbor interaction $(J<0)$ on a square lattice whose sites are occupied by spins $S=1/2$ directed along the OZ axis in the direction of the external field. The Hamiltonian has the form

$$H=-\frac{1}{2}\sum_{h=1}^{4}\sum_{i=1}^{N}\{J^{zz}(h)S_i^zS_{i+h}^z+J^{x,y}(h)\times(S_i^xS_{i+h}^x+S_i^yS_{i+h}^y)\}-\sum_{i=1}^{N}H_i^zS_i^z, \quad (1)$$

where $\Delta=1-J^{x,y}/J^z$ is the easy-axis type exchange anisotropy, H^z is the external magnetic field, and N is the total number of spins.

The MC algorithm and computational method have been described in detail in a previous work.[33] The Hamiltonian is divided into four-spin clusters on a placquette and the commutation between the clusters is taken into account by Trotter's relation. In the present paper, periodic boundary conditions in the Trotter direction and along the lattice are used in the MC procedure. The linear size of the lattice is $L=40,48,64$, and 80 and $m=16$, 32, and 48. The number of MC steps per spin ranges from 3000 to 10000. One MC step is determined by rotating all spins on a $L\times L\times 4m$ lattice. The following were calculated: the energy E, the specific $C=dE/dT$, the magnetization M, the susceptibility in an external field $\chi=M/H$, the pair and four-spin correlations in the longitudinal and transverse spin components, their Fourier spectrum, the sublattice magnetization $\sigma=\lim_{r\to\infty}\sqrt{\mathrm{abs}(\langle S_0^zS_r^z\rangle)}$, and the correlation radius ξ from the approximation of the spin correlation function as

$$R(r)=A/r^{\eta}\exp(-r/\xi), \quad (2)$$

where $R(r)$ is the normalized correlation function $R(r)=|\langle S^z(0)S^z(r)\rangle|-\langle S^z\rangle^2$.

2. DISCUSSION

We shall determine the energy and spin correlation functions of an isotropic AF in the ground state by two methods: asymptotic continuation of these quantities determined for an anisotropic AF with $\Delta=1-J^{x,y}/J^z\to 0$ and for an isotropic AF with $T\to 0$. The finite lattice dimensions employed in the MC calculations cut off the region of long-wavelength excitations at the wave number $k\sim\pi/L$ and limit the minimum exchange anisotropy, which satisfies $\sqrt{\Delta}>\pi/L$. Under this condition the contribution of long-wavelength excitations to the thermodynamics will be exponentially small.

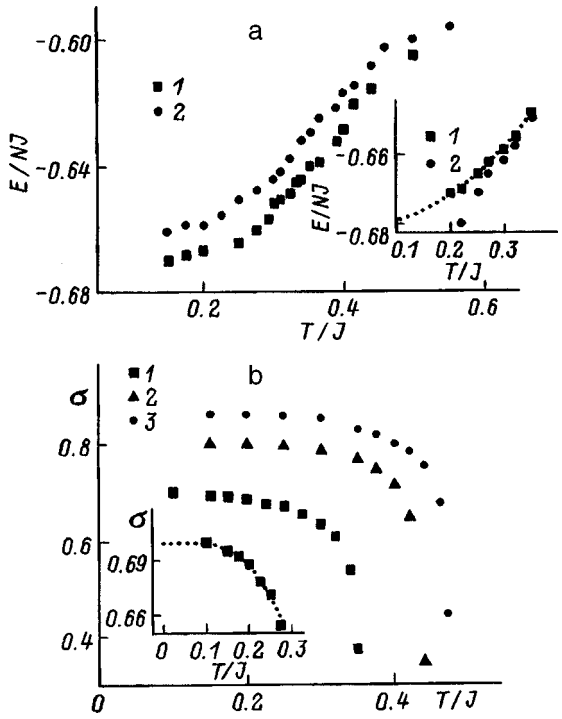

FIG. 1. Temperature dependences of the energy E/NJ of an anisotropic AF with $\Delta=0.02(1)$, $0.075(2)$ and an isotropic AF (inset: $\Delta=0.0, m=32(1)$, $16(2)$) (a) and the sublattice magnetization σ of an AF with $\Delta=0.05(1)$, $0.15(2)$, $0.25(3)$ (b). Inset: The dotted lines show the power-law functions $E=-0.682(4)+0.35T^{2.0(2)}$ (a) and $\sigma=0.70(3)-3.(4)T^{3.(2)}$ (b).

We shall calculate the temperature dependences of the energy, sublattice magnetization, and spin correlation functions for a number of exchange anisotropy constants $\Delta\geqslant 0.005$. The typical dependences are shown in Fig. 1. At low temperatures, less than the gap between the ground and excited state $T<4SJ\sqrt{\Delta(1+\Delta)}$, we extrapolate the computed quantity A using a power law $A=A(T=0)-\alpha T^{\beta}$ (shown by the dotted line in the inset in Fig. 1) and an exponential law $A=A(T=0)-\alpha\exp(-\beta/T)$ with three adjustable parameters α,β, and A at $T=0$. The extrapolated values of E and $\langle S^z(0)S^z(r=1)\rangle$ for an anisotropic AF in the ground state are shown in Fig. 2. The dependence of these quantities on the exchange anisotropy can be approximated by the function $A=A(\Delta=0)\pm 1/\exp(\alpha/\Delta^{\beta})$ with the adjustable parameters α,β, and $A(0)$. The corresponding parameters are: for the energy $\alpha=1.61(7)$, $\beta=0.26(5)$; for the correlation functions $\langle S^z(0)S^z(r=1)\rangle\alpha=2.(1)$, $\beta=0.165(7)$. The energy of the $2D$ isotropic Heisenberg model in the ground state $E=-0.684(6)$ agrees well with the energy obtained by exact diagonalization $E=-0.68445$.[12] The spin-spin correlation function $\langle S^z(0)S^z(r=1)\rangle=-0.120(4)$ agrees quite well with the result $\langle S^z(0)S^z(r=1)\rangle=-0.114(4)$.[11]

The sublattice magnetization can be interpolated by the logarithmic law $1/\sigma=1+0.13(1)\ln(1/\Delta)$ (Fig. 3). In the in-

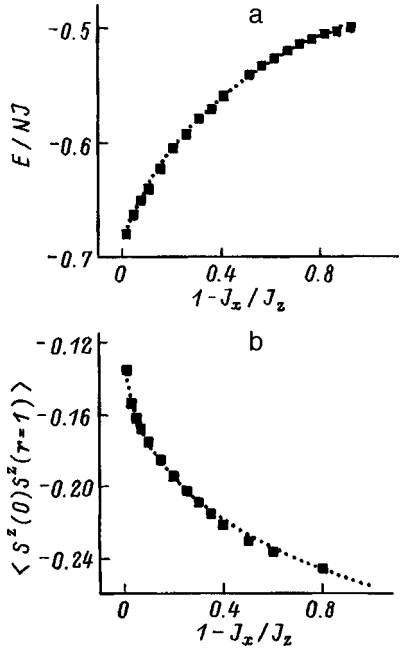

FIG. 2. The energy E/NJ (a) and nearest-neighbors correlation functions $\langle S_0^z S_1^z \rangle$ (b) of an AF in the ground state as a function of the exchange anisotropy $\Delta = 1 - J_x/J_z$. The lines show the interpolation functions $E = -0.684(6) + 1/\exp(1.61(7)/\Delta^{0.26(5)})$, $\langle S^z(0)S^z(r=1)\rangle = -0.120(4) + 1/\exp(2(1)/\Delta^{0.165(7)})$.

set in Fig. 3 the reciprocal of the magnetization is a function of $\ln(\Delta)$ is described well by a linear function. This means that there is no long-range antiferromagnetic order in the ground state in an isotropic ($\Delta = 0$, $\ln\Delta \to \infty$) $2D$ Heisenberg model. For the minimum anisotropy $\Delta = 0.005$ the sublattice magnetization is $\sigma = 0.29$, or (in Bohr magnetons with $g = 2$) $\sigma = 0.58\mu_B$. We shall determine the Néel temperature

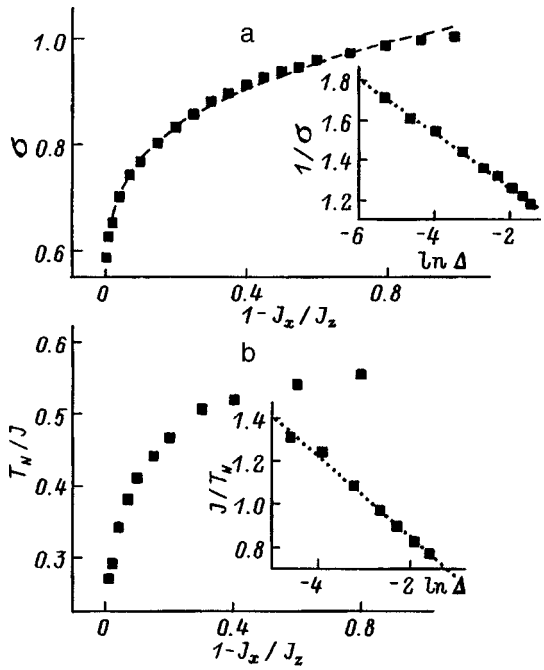

FIG. 3. Sublattice magnetization σ (a) and Néel temperature T_N/J (b) of an AF as a function of the exchange anisotropy. Inset: $1/\sigma$ (a) and J/T_N (b) versus $\ln\Delta$.

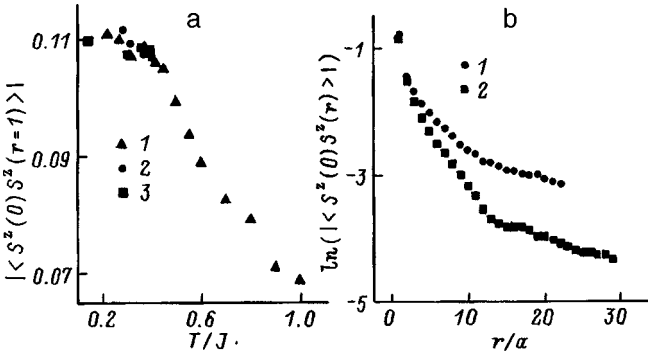

FIG. 4. Correlation functions with respect to the longitudinal components for AFs on 40×40 (*1*), 64×64 (*2*), 80×80 (*3*) lattices at distance $r/a = 1$ (a) and distance dependence of the logarithm of the spin-spin correlation function for $T/J = 0.28$ (*1*) 0.37 (*2*) (b).

of anisotropic AF from the maximum of the heat capacity, the point of inflection of the susceptibility, and $\sigma \to 0$ (Fig. 1). The dependence of $T_N(\Delta)$ on the exchange anisotropy (Fig. 3) likewise can be interpolated well by a logarithmic law for $\Delta \ll 1$ $T_N = 2/\ln(11/\Delta)$.

The temperature dependence of the energy of an isotropic AF in the interval $0.15 \leqslant T/J \leqslant 0.35$ can be approximated by a cubic polynomial with zero coefficients for the odd-numbered powers $E(T) = -0.682(4) + 0.35(3)T^{2.0(2)}$ (Fig. 1). The asymptotic continuation of the spin correlation functions in this interval for the longitudinal and transverse components (Fig. 4) gives $\langle S^z(0)S^z(r=1)\rangle = -0.113(3)$ and $\langle S^+(0)S^-(r=1)\rangle = -0.228(5)$, which agrees well with the results $\langle S^+(0)S^-(r=1)\rangle = -0.22823(2)$.[12] This confirms Anderson's idea of the existence of a singlet ground state, where $2\langle S^z(0)S^z(r=1)\rangle \approx \langle S^+(0)S^-(r=1)\rangle$ and the magnetic state can be represented by a superposition over all realizations of the singlet pairs. In other words, each pair of neighboring spins in the lattice plane is in a singlet state, but the combination of neighboring spins in a pair is always changing. If the singlets are ordered, then the four-spin correlation function at odd distances is greater than at even distances. The MC calculation of $\langle S_0^z S_1^z S_r^z S_{r+1}^z \rangle$ does not lead to this conclusion.

The correlation radius calculated from the distance dependence of the spin-spin correlation function (Fig. 4b) can be approximated well at temperatures $T/J \geqslant 0.26$ by the function $1/\xi = 2.05T^{1.0(6)}/\exp(1.0(4)/T)$, shown in Fig. 5a. In the singlet state the spin correlation function is a power-law function of distance. The excitations in this model are interacting spinons or solitons. The probability of excitation of a spinon (soliton) is $W \sim \exp(-E_s/T)$, where $E_s = J$ in the $1D$ Heisenberg model, and in the $2D$ model the excitation energy of a spinon (soliton) is two times higher since two singlet pairs must be broken in order to preserve cubic symmetry. The average distance between the temperature-excited spinons is $l(T) \sim 1\sqrt{W} \sim \exp(J/T)$. This can be represented in the form of a quasilattice embedded in a gas of singlet pairs (Fig. 6), where the spinons are represented by arrows. The spinons are correlated with one another. The correlation could vary as power law $\xi_s/l \sim A/T^{\alpha}$, where l is the quasilattice constant of the spins and decreases with increasing

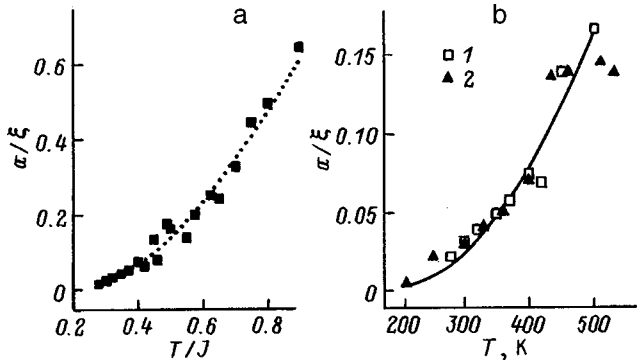

FIG. 5. Reciprocal of the correlation radius a/ξ, calculated by the MC method (a), (b, 1) and measured in a neutron scattering experiment in La_2CuO_4[23] (b, 2).[23]

temperature. The correlation radius at low temperatures should vary exponentially $\xi_s \sim A/T^{\alpha}\exp(J/T)$. There exists a minimum soliton size, determined by the ratio of the surface and volume magnetic energy for which the quasilattice becomes unstable at $T/J=0.37-0.4$.

The static magnetic factor, the Fourier component $S^z(Q)$ at $Q=\pi/a$, varies very little at temperatures $T/J<0.4$ as a function of temperature and the dimensions of the lattice (Fig. 7a). For $T/J>0.4$ the temperature dependence of $S^z(Q)$ is the same as in a paramagnet. At low temperatures the Fourier spectrum of the spin correlation function can be represented as a superposition of the Fourier correlation function of the singlets $S_{SN}(q)\sim A/q^{-(2-\eta)}$ and spinons $S_{SP}(k)\sim B/((k-\pi)^2+1/\xi^2)$. Singlet pairs make the main contribution to the neutron scattering intensity in the limit $q\to\pi$, since the spin density is low and the contribution of the spins is $\sim W_{SP}(q)$. The temperature derivatives of the specific heat and susceptibility in this temperature range (Fig. 8) have a maximum, and $d\chi/dT$ and dC/dT to the right and left of T_c differ substantially. For comparison, the values of $\chi(T)$ and $C(T)$ calculated with a super computer,[16] on a $L=128\times128$ lattice with two orders of magnitude more MC steps than the present work, are shown in Fig. 8. The authors of Ref. 16 also noted violation of the relation $\ln(S(Q)\xi^{-2})\sim\ln(T/J)$ for $T/J=0.35$. Just as in Ref. 16, for $T/J<0.4$ the exponent η in Eq. (2) tends to decrease with temperature from $\eta=0.55$ to 0.35 at $T/J=0.26$.

Weakly anisotropic AFs have two transition temperatures: the Néel temperature, associated with a breakdown of long-range order, and T_c associated with breakdown of topological magnetic formations (solitons). The size of a soliton is inversely proportional to the magnitude of the anisotropy. For this reason, a quasilattice of solitons (spinons) is formed when the exchange anisotropy reaches the critical value $\Delta\approx0.05$ at temperatures $T/J<0.4$. The magnetic structure of the factor $S(Q)$ (Fig. 7b), the spin correlation functions

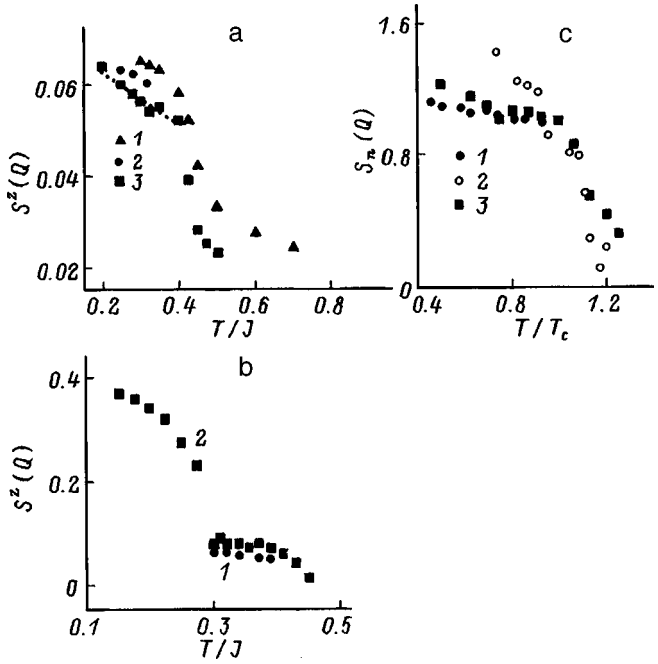

FIG. 7. Static magnetic structure factor $S^z(Q)$ for $Q=\pi/a$ and $L=40$ (*1*), 64 (*2*), 80 (*3*) in the isotropic (a) and anisotropic ($\Delta=0.02$,$L=40$ (*1*), 80 (*2*)) (b) cases as a function of temperature. c — Normalized static magnetic factor $S(Q,T)/S(Q,T=T_c)$, determined by the MC method for $T_c/J=0.4$ (*1*) and from neutron scattering in Er_2CuO_4[24] for $T_c=250$ K (*2*) and in La_2CuO_4[22] for $T_c=450$ K (*3*).

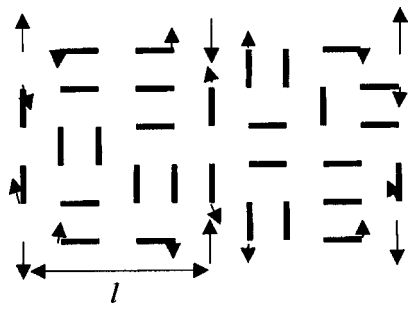

FIG. 6. Schematic representation of a quasilattice of singlet pairs of spins (segments) and spinons (arrows).

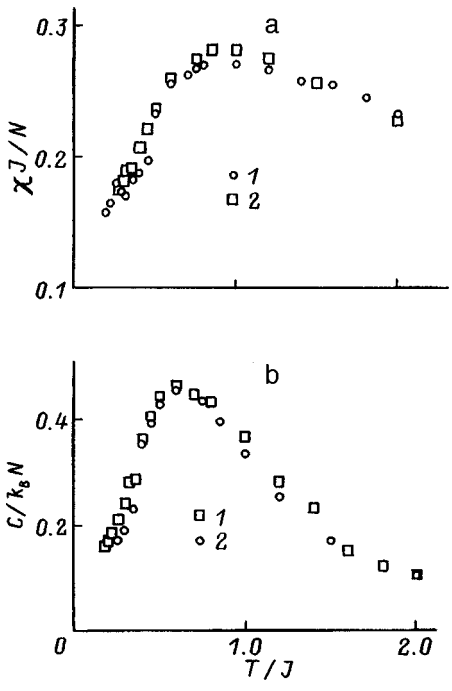

FIG. 8. Susceptibility $\chi J/N$ (a) and specific heat C/k_BN (b) calculated in the present work (*1*) and in Ref. 16 (*2*) as a function of temperature.

$\langle S^z(0)S^z(r=1)\rangle$, and the magnetic susceptibility are virtually temperature independent in the interval $T_N<T<T_c$.

The results obtained explain well the experimental data on the magnetization and the neutron scattering intensity in the quasi-two-dimensional compounds $Sr_2CuO_2Cl_2$,[27] La_2CuO_4,[23], and Er_2CuO_4.[24] The experimental values of the sublattice magnetization for $Sr_2CuO_2Cl_2$ and La_2CuO_4 are, respectively, $\sigma=0.42\mu_B$ and $\sigma=0.44\mu_B$. The exchange anisotropy in these compounds is of the order of $\Delta\sim10^{-4}$ and MC calculations give $\sigma=0.45\mu_B$. The very small decrease of the sublattice magnetization could be due to a covalence effect. The neutron scattering intensity $S(k=0)(k=\pi-q)$ in these compounds is essentially temperature independent above the Néel temperature up to some temperature $T_c/T_N=1.5-2.1$. For Er_2CuO_4, magnetic and resonance investigations[25,26] give a Néel temperature $T_N=165$ K, while neutron diffraction measurements give $T_N=(250-265)$ K.[24] The normalized magnetic structure factors — the experimental factor $S(Q,T)/S(Q,T=250\text{ K})$ and the theoretical factor $S(Q,T)/S(Q,T=0.4\text{ J})$ — qualitatively agree in the interval $(0.7-1)T/T_c$ (Fig. 7c). Taking account of the exchange anisotropy, as done in Fig. 7b, will improve the agreement with experiment. The magnitude of the exchange in Er_2CuO_4 can be estimated as $J\approx630$ K from the temperature T_c. For La_2CuO_4 the normalized intensity $S(Q,T)/S(Q,T=450\text{ K})$ agrees well with the MC results (Fig. 7c). This is probably due to the weaker exchange anisotropy, since the temperature interval between T_N and T_c decreases with increasing anisotropy. The magnitude of the exchange for La_2CuO_4, estimated from T_c, is $J\approx1125$ K, while the value estimated from an approximation of the correlation radius in the interval $T/J=0.26-0.46$ by the function $1/\xi=0.25T^{0.35}/\exp(1302/T)$ gives $J\approx1302$ K (Fig. 6c). An independent value of the exchange can also be obtained from the Néel temperature $T_N=2/\ln(11/\Delta)$ and $J=1160$ K.

These estimates of the exchange fall near the values 900 and 1500 K obtained on the basis of different theoretical and experimental methods. For example, the exchange values obtained from the function $1/\xi(T)$ determined using a nonlinear σ model and the normalization group or quantum MC method are $J=1175$[4] and 1450 K,[16] respectively. The exchange anisotropy $\Delta\sim0.003$ and sublattice magnetization $\sigma\approx0.56$ can be estimated from the Néel temperature $T_N=165$ K for Er_2CuO_4. It is desirable to perform for this compound more careful neutron diffraction measurements to determine the site moment. We note that the MC calculations are in good agreement with experiment in the temperature range of existence of singlets and spinons for $\Delta=0.003$: $(T_N/T_c)^{MC}=0.62$ and $(T_N/T_c)^{ex}=0.66$. The objective of this work was not only to determine a more accurate value for the exchange but also to investigate the temperature dependence of the magnetic structure factor and the correlation radius. In the compound, above the Néel temperature, the magnetic structure can be represented in the form of singlet pairs and spinons (solitons). Singlets make the main contribution to the magnetic structure factor while spinons, whose density decreases exponentially with increasing temperature, make the main contribution to the correlation radius.

Summarizing the results obtained in this paper, we can draw the following conclusions. There is no long-range antiferromagnetic order in the ground state in a two-dimensional isotropic Heisenberg model. The exchange anisotropy dependence of the sublattice magnetization of an anisotropic antiferromagnet is logarithmic: $1/\sigma=1+0.13(1)\ln(1/\Delta)$. At low temperatures, $T/J<0.4$, the magnetic static factor $S(Q)$ is essentially temperature-independent, while the correlation radius varies exponentially. This could be due to the existence of singlets, which make the main contribution to $S(Q)$, and the temperature-excited spinons, which give an exponential behavior of the correlation radius. The sublattice magnetization, correlation radius, and static magnetic structure factor above the Néel temperature in Er_2CuO_4, La_2CuO_4, and $Sr_2CuO_2Cl_2$ are described well by the $2D$ Heisenberg model with spin $S=1/2$.

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