Dimer state in the two-dimensional anisotropic alternated-exchange Heisenberg model

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An analysis is made of the two-dimensional Heisenberg model with $S=1/2$, anisotropic exchange interaction between nearest neighbors, and alternating exchange in two directions, [100] and [010] (corresponding to condensation of the (π,π) mode) and in one direction [100] (corresponding to condensation of the $(\pi,0)$ mode). The quantum Monte Carlo method is used to calculate the thermodynamic characteristics and the spin correlation functions which are used as the basis to determine the boundary of stability of an anisotropic antiferromagnetic with respect to alternation of exchange $\delta=(1-J^{x,y}/J^z)^{0.4}$ in the (π,π) model and $\delta=(1-J^{x,y}/J^z)^{0.31}$ in the $(\pi,0)$ model. In the $(\pi,0)$ model a disordered quantum state exists in the range $(1-J^{x,y}/J^z)^{0.31}<\delta<(0.3–0.35)$. The energy $(E-0.68)=0.36\delta^{1.80(6)}$ and $0.21\delta^{2.0(5)}$, the energy gap between the ground and excited states $H_c(\delta)=1.96\delta^{2.(1)}$, $1.8(1)(\delta-0.35(3))^{0.67(2)}$ were determined as a function of the alternation of exchange in the (π,π)- and $(\pi,0)$ models, respectively. © *1998 American Institute of Physics.* [S1063-7834(98)02506-4]

Dimerization of the lattice caused by electron–phonon interaction and leading to a spin–Peierls transition in one-dimensional systems has been analyzed in detail in the literature.[1,2] Following the discovery of high-temperature superconductors, the Peierls instability was investigated using the two-dimensional half-filled Hubbard model.[3,4] In the strong-attraction limit $(U/t)\gg 1$ of the adiabatic approximation, an exact diagonalization method[5] was used to analyze the alternating-exchange model which corresponds to the (π,π)- and $(\pi,0)$ phonon modes. According to these calculations, dimerization takes place in the [100] direction.

The region of stability of antiferromagnetic ordering with respect to exchange alternation was calculated by numerically solving a system of equations for the spin operators in the Schwinger representation using a 40×40 lattice[6] in the inhomogeneous Hartree–Fock approximation.[7,8] In all cases, the long-range antiferromagnetic order disappears for a critical dimerization of the lattice, which corresponds to a 50% change in volume, when the alternation is $\sim 0.5J$. For quasi-two-dimensional magnets $CuGeO_3$ (Ref. 9) and $Cs_3Cr_2Br_9$ (Ref. 10), in which a transition takes place to the dimer state, these estimates are not realistic. Possibly because of these high estimates of exchange alternation, interest in studies of two-dimensional alternating exchange models has declined.

Three problems are solved here. The first involves determining which phonon mode, (π,π) or $(\pi,0)$, gives the largest magnetic energy per alternated bond. The second involves studying the stability of the antiferromagnetic ordering relative to exchange alternation as a function of the volume anisotropy. The third involves identifying whether a disordered quantum state exists or whether the antiferromagnet is converted directly to the dimer state as the exchange alternation parameter increases. A quantum Monte Carlo method based on a trajectory algorithm is used to solve these problems.[11] The basic idea of the algorithm is to transform the quantum D-dimensional problem to a classical $D+1$-dimensional one by introducing ''time'' cutoffs in the imaginary time space $0<\tau<1/T$ and implementing a Monte Carlo procedure in the ''imaginary time–coordinate'' space.

MODEL AND GROUND STATE OF THE TWO-DIMENSIONAL HEISENBERG MODEL WITH ALTERNATING EXCHANGE

We consider a two-dimensional lattice with the spins $S=1/2$ localized at lattice sites. Exchange alternation will be considered using two models. In the first case, alternation takes place in one of the directions of the lattice (for example, [100]) and according to the notation used in Ref. 5, is caused by condensation of the $(\pi,0)$ phonon mode. In the second case, alternation takes place in two directions and is caused by condensation of the (π,π) mode, i.e., $J_{l,l+1}=J_0+\delta$, $J_{l+1,l+2}=J_0-\delta$ (Fig. 1). This exchange inhomogeneity may be caused by distortion of the lattice $J^c_{l,l+1}-J^c_{l+1,l+2}=\lambda'(u_l-u_{l+1})$, where u is the displacement of an atom from the equilibrium position, or by anharmonicity of the vibrations. The Hamiltonian in the $(\pi,0)$ model has the form

$$H=-\frac{1}{2}\sum_{i,j=1}^{L}\{J^{z(010)}_{i,j}S^z_iS^z_j+J^{x,y(010)}_{i,j}(S^+_iS^-_j+S^-_iS^+_j)/2\}$$
$$\times\frac{1}{2}\sum_{i,j=1}^{L}\{(J^{z(100)}_{i,j}+(-1)^j\delta^z)S^z_iS^z_j+(J^{x,y(100)}_{i,j}$$
$$+(-1)^j\delta^{x,y})(S^+_iS^-_j+S^-_iS^+_j)/2\}-\sum_{i=1}^{N}h^zS^z_i,$$

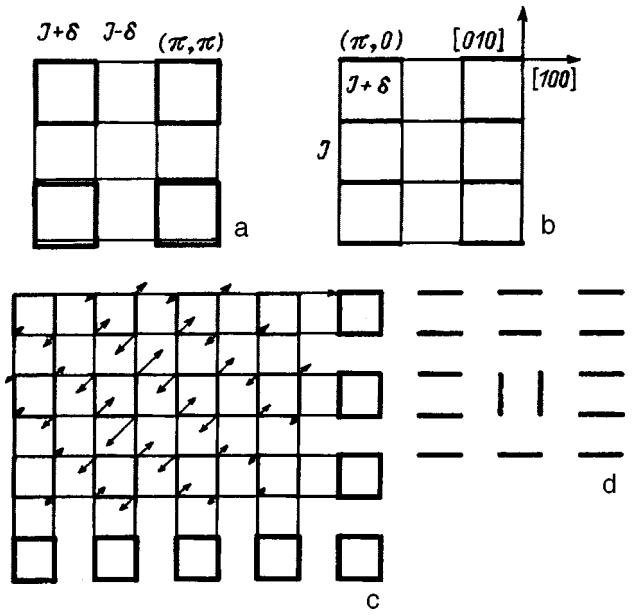

FIG. 1. Distribution of bonds on the lattice in two models: (π,π) (a) and $(\pi,0)$ (b), the arrows indicate a soliton (c), and the line segments corresponds to spin pairs in the singlet state — dimers (d).

and in the (π,π) model

$$H=-\frac{1}{2}\sum_{i,j=1}^{L}\{(J_{i,j}^{z}+(-1)^{j}\delta^{z})S_i^zS_j^z+(J_{i,j}^{x,y}+(-1)^{j}\delta^{x,y})\times(S_i^+S_j^-+S_i^-S_j^+)/2\}-\sum_{i=1}^{N}h^zS_i^z,$$

where $J^{z,x,y}<0$ is the anisotropic interaction, $J^z>J^{x,y}$, $\Delta=1-J^{x,y}/J^z$, $\delta^{z(x)}$ is the exchange alternation parameter, $H=h^z/J$ is the external magnetic field, and L is the linear dimension of the lattice $(N=L\times L)$ (Fig. 1a).

The algorithm and Monte Carlo method were described in detail in Ref. 12. The Hamiltonian is divided into clusters of four spins per square whose commutation is taken into account using the Trotter equation. Here periodic boundary conditions in the Trotter direction and along the lattice are used in the Monte Carlo procedure. The linear dimension of the lattice is $L=40$, 48, 64 and $m=16$, 24, 32. The number of Monte Carlo steps per spin varied between 3000 and 10 000. One Monte Carlo step is determined by the flip of all spins on a $L\times L\times 4m$ lattice.

We shall determine the order parameter of the dimers from the four-spin correlation function $\langle S_0^zS_1^zS_r^zS_{r+1}^z\rangle$ whose dependence on distance is oscillatory and has a difference between the minimum and the maximum of $\langle S_0^zS_1^zS_r^zS_{r+1}^z\rangle-\langle S_0^zS_1^zS_{r+1}^zS_{r+2}^z\rangle$. We calculate the pairwise spin–spin correlation functions for the longitudinal and transverse components of the spins, between which a relation must be satisfied at distance $r=1$ to establish a singlet state.

We determine the region of stability of the antiferromagnetic and dimer states from the spin–spin correlation functions, the dimerization parameter, and the correlation radius calculated for three temperatures $T/J=0.1$, 0.15, and 0.2 as a function of the exchange alternation parameter for various values of the exchange anisotropy $\Delta=0$, 0.01, 0.05, 0.1, 0.15, 0.2, 0.25, 0.3, 0.4, and 0.5. The critical exchange alternation δ_c for which the long-range antiferromagnetic order disappears, is determined from the spin–spin correlation functions $\langle S_0^zS_r^z\rangle\to 0$ at the distance $r=L/2$ calculated for different lattice dimensions.

Figure 2 gives the spin correlation functions for three values of the exchange anisotropy parameter $\Delta=0$, 0.05, 0.25, calculated using the two models of exchange alternation. The exchange alternation corresponding to the point of inflection of the dimerization parameter $q(\delta)$ and the formation of a dimer state coincides with the critical value δ_c for which antiferromagnetic order is impaired in both the (π,π) and $(\pi,0)$ models for the exchange anisotropy $\Delta>0.02$. For the isotropic Heisenberg model, the dependence $q(\delta)$ is linear and passes through the origin for (π,π) dimerization (Fig. 2b) and intercepts the δ axis at $\delta_c\approx 0.3$ for $(\pi,0)$ (Fig. 2a). The spin correlation functions between the nearest neighbors along the longitudinal and transverse components of the spin do not vary significantly as the exchange alternation increases for (π,π) and $(\pi,0)$ dimerization in the [100] direction since an isotropic two-dimensional antiferromagnet is in the singlet state.[13,14] In an anisotropic antiferromagnet in the direction of exchange alternation the correlation function along the transverse components increases, which also indicates dimer formation, whereas in the [010] direction in the $(\pi,0)$ direction, $\langle S_0^+S_1^-\rangle$ decreases with increasing δ.

The correlation radius in the dimer state diverges following a power law as the critical value δ_c is approached (Figs. 2c and 2d). For (π,π) dimerization the relation $\xi=1/(\delta-\delta_c)^{\beta}$ is satisfied, where the exponent decreases with increasing exchange anisotropy. In the isotropic case, the correlation radius is well approximated by $\xi=1/\delta^{2.(15)}$ in the (π,π) model and by $\xi=4.(5)/(\delta-0.33(3))^{0.70(4)}$ in the $(\pi,0)$ model with the critical value $\delta_c=0.33(3)$. The corresponding interpolated dependences are given by the dashed lines in Figs. 2c and 2d, and to within the calculation error of $\sim 10\%$ do not depend on the lattice dimensions as shown in the figure for $L=48$ and 64. The calculated dependences $q(\delta)$, $\xi(\delta)$, and $\langle S_0^{z,+}S_1^{z,-}\rangle(\delta)$ indicate that alternation of exchange in two directions in the two-dimensional Heisenberg model is accompanied by the formation of a dimer state and an anisotropic antiferromagnetic is converted to the dimer state at a certain critical value of the exchange alternation parameter. A correlation in terms of longitudinal spin components exists between the dimers in a region of dimensions $\sim\xi^2$, shown in Fig. 1c. In the $(\pi,0)$ model an ordered dimer state is formed at the critical value $\delta_c=0.3$–0.35. The energy of an isolated dimer is $E/J=3/2(1+\delta)$. When two dimers commute in the [010] direction, as shown in Fig. 1d, the energy is reduced by $\Delta E/J=3\delta$. If this energy is lower than the triplet excitation energy $\Delta E/J=1$, then for $\delta<\delta_c=1/3$ no ordered dimer state exists. In the $(\pi,0)$ model the correlation radius is anisotropic and has a maximum in the [100] direction.

The energy calculated by the Monte Carlo method for the two models, (π,π) and $(\pi,0)$, and different exchange anisotropies is accurately fitted by the power dependence $(E-E(0))=A\delta^{\alpha}$, where the exponent α increases with in-

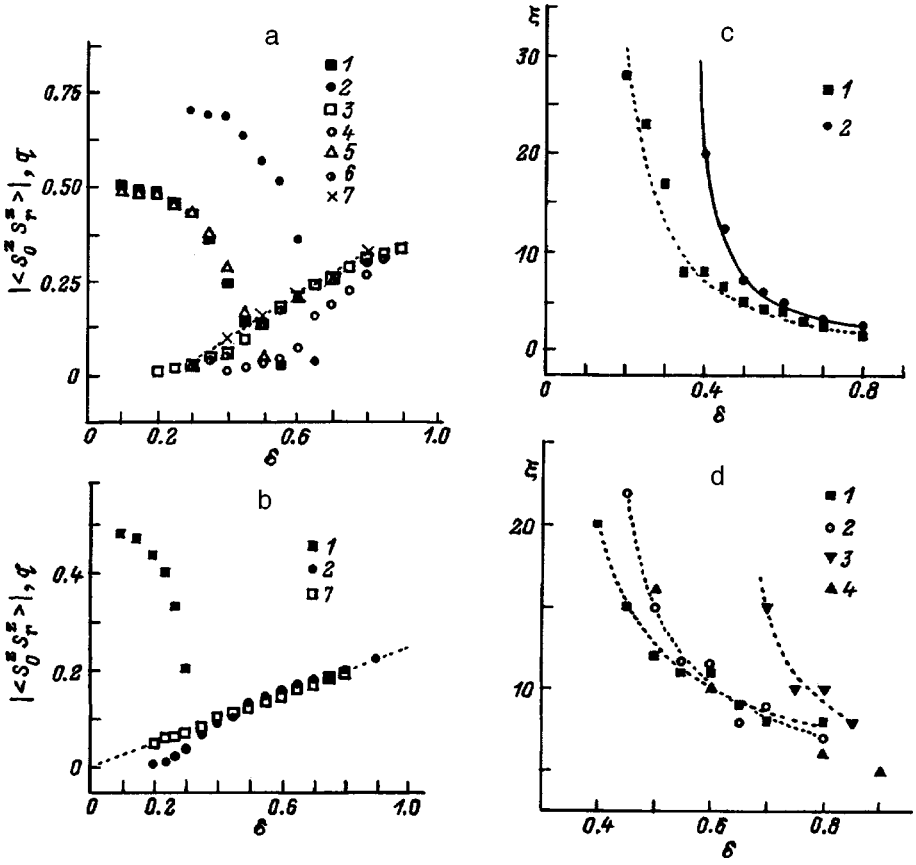

FIG. 2. Correlation function at the distance $r=22$ (*5, 6*), 30 (*1*), the dimerization parameter q (*2–4, 7*) for the exchange anisotropy $\Delta=0$ (*7*), 0.05 (*1–3, 5*), 0.25 (*4, 6*) on a lattice $L=64$ (*1, 2, 7*), 48 (*3, 5, 6*), and the correlation radius ξ for $\Delta=0$ (*1*), 0.05 (*2, 4*), 0.25 (*3*) on 64×64 (*1, 2*) and 48×48 (*3, 4*) lattices as a function of the exchange alternation in the $(\pi,0)$ (a, c) and (π,π) (b, d) models.

creasing exchange anisotropy. In the isotropic limit in the (π,π) and $(\pi,0)$ models, the interpolated dependences respectively have the form $(E-0.68)=0.36\delta^{1.80(6)}$ and $0.21\delta^{2.0(5)}$. For $\delta\approx0.50(4)$ the energies calculated using the two dimerization models and normalized to the number of alternated bonds are the same. For $\delta<0.50(4)$, the dimer state energy normalized to the number of alternated bonds has a higher absolute value in the (π,π) model compared to the $(\pi,0)$ model, and for $\delta>0.50(4)$ we find $E(\pi,\pi)<E(\pi,0)$. In a spin-Peierls transition, the increase in the magnetic energy achieved by dimerization should exceed the energy loss in the elastic system $0.36\delta^{1.80(6)}\geqslant Ku^2/2$ or $\lambda^{1.8}\geqslant1.4Ku^{0.2}$, where λ is the spin–phonon interaction constant, K is the modulus of elasticity, $u=|u_i-u_j|$ is the change in the distance between nearest neighbors, i.e., as a result of interaction between the elastic and magnetic subsystems for $\delta<0.5J$, dimerization of the magnetic structure takes place preferentially in two directions. For large spin–phonon interaction constants, dimers may be formed along one of the translation vectors of the lattice. Calculations made for small lattices[5] indicate that $(\pi,0)$ dimerization predominates. This may be caused by the finite dimensions of the 4×4 lattice. For example, the linear dimension of the lattice is equal to the correlation radius for $\delta=0.7$, and for this exchange alternation the Monte Carlo calculations give $(\pi,0)$ dimerization.

The boundary of stability of long-range antiferromagnetic order is accurately approximated by the power dependence $\delta=(1-J^{x,y}/J^z)^{0.4}$ in the (π,π) model and $\delta=(1-J^{x,y}/J^z)^{0.31}$ in the $(\pi,0)$ model. In the $(\pi,0)$ model a disordered quantum state exists in the range of parameters $\delta<\delta_c=0.27-0.33$ and $\Delta\leqslant0.02$.

2. DETERMINATION OF THE SEQUENCE OF DIMER STATE–QUANTUM DISORDERED STATE–PARAMAGNETIC TRANSITIONS

Calculations of the specific heat and susceptibility as a function of temperature reveal two critical regions and two characteristic transition temperatures: T_{c1} and T_{c2}. Below T_{c1} the temperature dependence of the specific heat and the susceptibility is accurately approximated by an exponential dependence which indicates that there is an energy gap in the excitation spectrum. In the range $T_{c1}<T<T_{c2}$ the behavior of $C(T)$ obeys a power law. At low temperatures $T<T_{c1}$ an ordered dimer state is conserved in both models. The dimerization parameter q, the correlation radius, and the correlation functions along the longitudinal spin components at distance $r=1$ depend fairly weakly on temperature for $T<T_{c1}$ (Fig. 3). This is because the excitations are spinons (a concept introduced by Anderson[13]), i.e., the dimer breaks down into two spins separated by a certain distance. This type of excitation may be represented as a soliton, as shown in Fig. 1c. At T_{c1} soliton percolation occurs and in the range $T_{c1}<T<T_{c2}$ a soliton gas forms. With increasing temperature, the soliton density increases, the average distance between them decreases, and the correlation radius is therefore reduced. Near T_{c2} the temperature dependence of the correlation radius $\xi(T)$ may change from exponential to a power

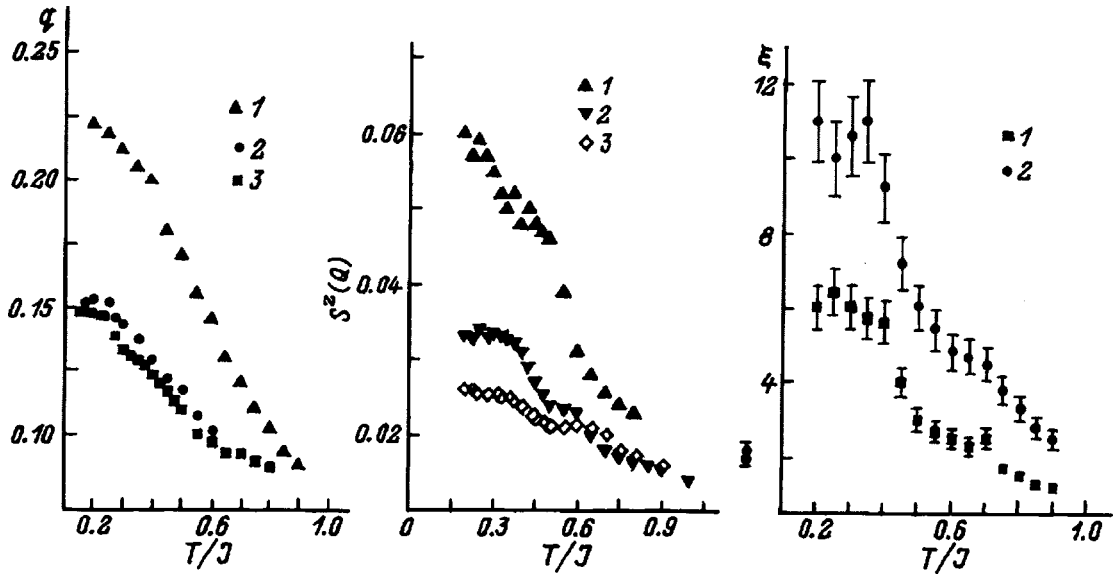

FIG. 3. Temperature dependences of the dimerization parameter q in the [100] direction, the correlation radius ξ in the [010] direction for $\Delta=0.05$, $\delta=0.65$ (*1*), $\Delta=0.0$, $\delta=0.5$ (*2, 3*) for $L=64$ (*1, 2*), 48 (*3*) and the static magnetic structure factor $S^z(Q)$ for $Q=\pi$ in the [010] (*1, 3*), and [100] (*1, 2*) direction for $\Delta=0.05$, $\delta=0.65$ (*2, 3*), and $\Delta=0.0$, $\delta=0.35$ (*1*) in the dimer state for the $(\pi,0)$ model.

dependence (Fig. 3). The correlation function $\langle S_0^{z,+}S_1^{z,-}\rangle$, and the static magnetic structure factor $S^z(Q)$ at $Q=\pi$ have two points of inflection caused by a transition from the dimer state to a disordered quantum state having short range dimer order and topological excitations (solitons) (QD) and by a quantum disorder–paramagnetic (QD–PM) transition. For $\delta=0.3$ in the $(\pi,0)$ model, $S^z(Q)$ has a single point of inflection at T_{c2} (Fig. 3).

The dependence of the DS–QD transition temperature is accurately described by the power law $T_{c1}=0.7(\delta$

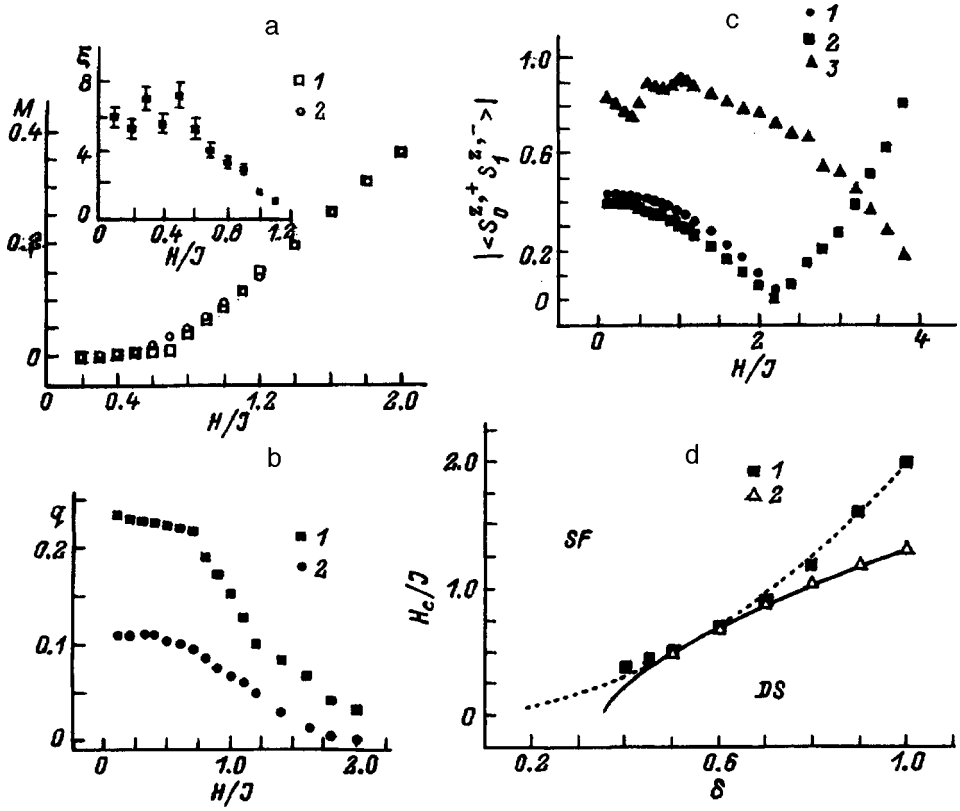

FIG. 4. Magnetization M (a), correlation radius ξ (inset), dimerization parameter q (b), and spin–spin correlation function at $r=1$ along the longitudinal (z) (*1, 2*) and transverse $(+,-)$ (*3*) components of the spin $\langle S_0^{z,+}S_1^{z,-}\rangle$ in the $(\pi,0)$ model, $\Delta=0.05$, $\delta=0.65$ (*1*) and in the (π,π) model, $\Delta=0.0$, $\delta=0.45$ (*2, 3*) (c) as a function of the external field. d — Phase diagram of the dimer state (DS), the spin flip phase (SF) on the field–exchange alternation plane in the (π,π) (*1*) and $(\pi,0)$ (*2*) models for isotropic exchange $\Delta=0$.

$-\delta_c)^{0.50(4)}$ in the $(\pi,0)$ model, where the parameter δ_c shows good agreement with the critical values of exchange alternation in the isotropic case $\delta_c=0.33(2)$. For (π,π) alternation of exchange $T_{c1}(\delta)=1.10(7)\delta^{2.0(7)}$.

The energy gap between the ground and excited states is determined from the dependence of the magnetization on the external magnetic field perpendicular to the lattice plane. For example, for the critical field H_c the magnetization is $M\neq 0$, and the correlation radius and dimerization parameter decrease abruptly with increasing field in both models (Fig. 4). The correlation functions for the transverse components vary negligibly. Here we can also identify a range of fields $H_c<H<H^*$, in which an inhomogeneous magnetic state exists which disappears when $\xi\to 0$. The dependence $M(H)$ is linear in this range of fields. In fields $H>H^*$ a classical spin flip state is formed. Figure 4 gives the critical fields as a function of the exchange alternation for the two models in the isotropic case. In the (π,π) and $(\pi,0)$ models these dependences are power laws $H_c=1.96\delta^{2.(1)}$ and $H_c=1.8(1)(\delta-0.35(3))^{0.67(2)}$, respectively. When exchange alternates in two directions, the ratio $H_c/T_{c1}\cong 1.78$ does not depend on the exchange alternation and in the $(\pi,0)$ model a dependence on δ is observed which can be approximately estimated as $H_c/T_{c1}\sim(\delta-0.34)^{0.17}$. Thus, this is related to the anisotropy of the correlation radius. As δ increases, the magnetic quasi-one-dimensionality increases and the density of states of the singlet and triplet excitations becomes redistributed, which is observed as a temperature shift of the maximum specific heat and susceptibility. In the two-dimensional Heisenberg model we find $T_{C\max}/T_{\chi\max}\approx 0.5$, and in the one-dimensional model $T_{C\max}/T_{\chi\max}\approx 0.76$. In the range $\delta\approx 0.5$, where the dimer state energies in the two models are the same, the energy gaps are also the same.

Thus, asymptotic dependences on the exchange alternation of the energies $(E-0.68)=0.36\delta^{1.80(6)}$ and $0.21\delta^{2.0(5)}$, and the energy gap between the ground and triplet states $H_c(\delta)=1.96\delta^{2.(1)}$ and $1.8(1)(\delta-0.35(3))^{0.67(2)}$ were obtained for exchange alternation along two translation vectors or along one of these. The boundaries of stability of an anisotropic antiferromagnet relative to exchange alternation $\delta=(1-J^{x,y}/J^z)^{0.4}$ in the (π,π) model and $\delta=(1-J^{x,y}/J^z)^{0.31}$ in the $(\pi,0)$ model were determined. The energy per alternated bond has a higher absolute value in the (π,π) model compared to the $(\pi,0)$ model for $\delta<0.5$. When exchange alternates along one of the translation vectors, an anisotropic antiferromagnet with the anisotropy $\Delta<0.02$ is transferred to the dimer state via a disordered quantum state. This state exists at temperatures between DS and PM.

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