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Simulation of magnetic properties of the two-dimensional magnetic with anisotropic antiferromagnetic interactions and cluster ordering by quantum Monte Carlo

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ABSTRACT

Magnetic with anisotropic antiferromagnetic exchange interactions and special topology of coupling in the square lattice with spins pairs ordering is studied by quantum Monte Carlo method. The antiferromagnetic order is found to be more stable as compared to spin liquid state. Exchange interactions and wave vector of structure modulation for $Cu_3B_2O_6$ is estimated. Neel temperature versus strength of exchange in spin pair is calculated. Plateau and modulation of magnetic structure in field magnetization dependence is revealed.

1. Introduction

In recent years, much attention has been devoted to investigation into the properties of low-dimensional quantum spin system. These are given by the close proximity of competing phases with long range order and short range correlated states, for example Neel order and a spin liquid. The dynamic of such system is governed even at finite but low temperatures by quantum fluctuations instead of thermal fluctuations. Excitation spectrum of spin liquid may be gapless or contains gap in the triplet spin excitation spectrum. Existence of gap is determined by a 2D exchange topology [1,2], additional frustrating exchange terms or geometry of lattice [3,4]. The ladder system $SrCu_2O_3$ formed by two coupled chains has a large singlet–triplet gap $\Delta \sim 0.5J$ [5]. The plaquette system CaV_4O_9 [6] consists of two-dimensional layers of VO_5 pyramids with Ca ions embedded between them. The ground state of the system is singlet with an energy of gap of 107 K. The simplest realization of a spin gap is a spin dimer. The dimers in the system $SrCu_2B_2O_6$ [7] arises from strong frustration between the diagonal coupling and square plane exchange. This compound has a spin gap of $\delta = 34$ K and very localized triplet excitations. The magnetic susceptibility of gapped spin liquid vanishes as $T \to 0$. The temperature dependance of the heat capacity is described by the exponential function.

Similar properties reveal the $Cu_3B_2O_6$ [8] crystal. The temperature dependence of the magnetic susceptibility passes through a broad maximum near 40 K, followed by a sharp decrease at temperatures below 10 K and a arise at 5 K. Incidently, the magnetization is only $\sim 0.05\mu_B$/Cu in a magnetic field of 30 T at 1.5 K [8]. Heat capacity has an anomaly at $T_c = 9.8$ K, that depends on magnetic field approximately as H^2 [9]. The analyze of the experimental data suggest that spin subsystem consists of single spins, clusters of pairs and fours of spins interacting with one another. In authors [9] opinion a antiferromagnetic order with the spin direction parallel to the *bc* plane appears at temperatures below 10 K. The spin entropy is much smaller than the value of the free-spin entropy at $T > 10$ K. Another point of view [8] that is the possibility of the existence of the spin-singlet ground state in $Cu_3B_2O_6$. NMR measurements suggest [10] that part of the magnetic system

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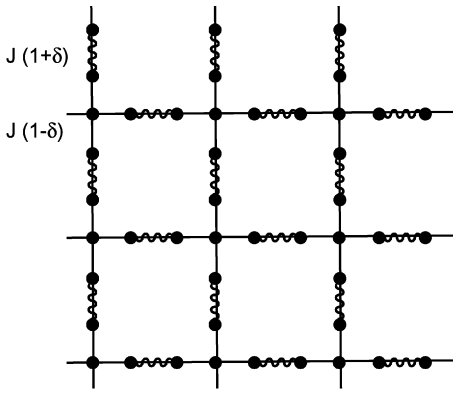

Fig. 1. Exchange bonds topology in lattice. $J(1+\delta)$ is denoted by wavy line, $J(1-\delta)$ is thin line.

forms a modulated spin density wave and μSR spectroscopic data supports coexistence of antiferromagnetically ordered pairs with singlet spins. Relation $T_N/\Theta \simeq 0.02$ is fairly small for large exchange anisotropy $\Theta(\|bc)/\Theta(\perp bc) \sim J^{xx}/J^{zz} = 0.8$ [9], where Θ is paramagnetic Curie temperature.

In this Letter, we consider the influence of the exchange interactions topology in the unit cell consisted single and pairs of spins relative to the stability of antiferromagnetic order or the formation of spin liquid state in two-dimensional lattice into account strong quantum fluctuation.

2. Model and discussion

We study the Heisenberg model with alternating exchange and spin $S = 1/2$. Hamiltonian is given by

$$H = -\sum_{i,j,\alpha}\big((1-\delta)J^{\alpha\alpha}S^{\alpha}_{i,j}S^{\alpha}_{i+1,j} + (1+\delta)J^{\alpha\alpha}S^{\alpha}_{i+1,j}S^{\alpha}_{i+2,j} + (1-\delta)J^{\alpha\alpha}S^{\alpha}_{i+2,j}S^{\alpha}_{i+3,j} + (1-\delta)J^{\alpha\alpha}S^{\alpha}_{i,j}S^{\alpha}_{i,j+1} + (1+\delta)J^{\alpha\alpha}S^{\alpha}_{i,j+1}S^{\alpha}_{i,j+2} + (1-\delta)J^{\alpha\alpha}S^{\alpha}_{i,j+2}S^{\alpha}_{i,j+3}\big) - \sum_i H_i S^z_i, \quad (1)$$

where $J(1+\delta)$, $J(1-\delta)$, $J<0$ are the exchange interactions between the nearest neighbors, H is the external magnetic field, $\eta = (J^{zz} - J^{xx(yy)})/J^{zz}$ is the exchange anisotropy. As a calculation method, we take the quantum Monte Carlo method unifying two algorithms, world-lines and continuous time for the spin $S = 1/2$. The continuous time world-line Monte Carlo approach [11] based on the expansion of the statistical evolution operator $e^{-H/T}$ in powers of J is applied. The world-line configuration of spins are updated through the space–time motions of the creation and annihilation operators [12]. The periodic boundary conditions are applied on $L = 48, 60$ square lattices. From 40 000 to 60 000 Monte Carlo steps (MCS) per site are spent to reach equilibrium and another 80 000–100 000 MCS are used for the averaging. The root mean square errors of the computed quantities lie in the range 0.1–2%.

We consider specific topology of exchange bonds plotted in Fig. 1. There are two local exchange field $E_1 = 4(1-\delta)JS$ and $E_2 = 2JS$. In limit case $\delta \to 1$ magnetic structure consists from part of spins located in small exchange field and the pairs of spins in the singlet state. Energy related to one bond is $E/J = 0.44$ for chain and $E/J = 0.75$ for dimer and $E/J = 0.34$ for square lattice. The effective antiferromagnetic interaction derived from integrating out the singlets created by pair of spins is $J_{\text{eff}} = 0.5J(1-\delta)^2/(1+\delta)$ and energy of antiferromagnetic with long range magnetic order (LRMO) is equal to $E/J = 0.68J_{\text{eff}}$, that is closely to energy of

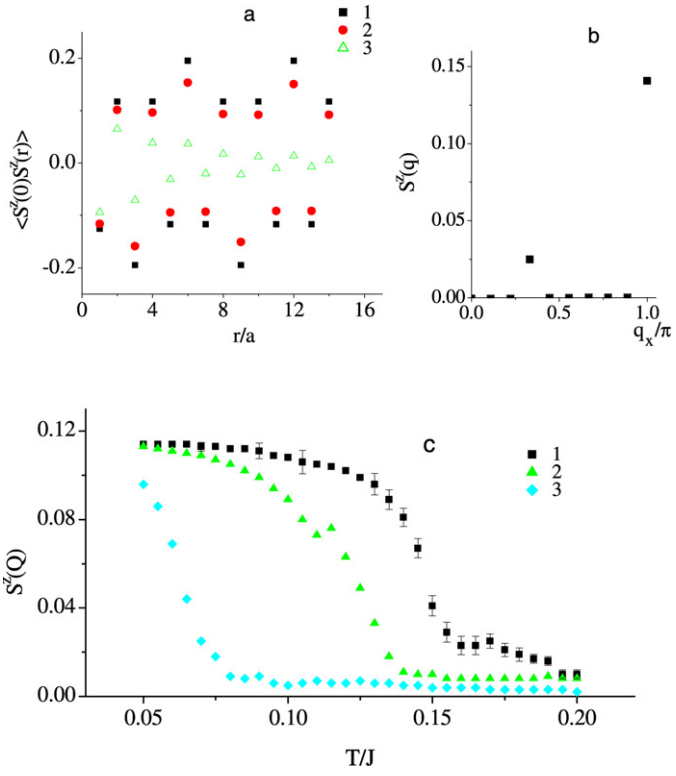

Fig. 2. Spin–spin correlation function $\langle S^z(0)S^z(r)\rangle$ of magnets calculated by MC at $\eta = 0.75$, $\delta = 0.3$, $T/T_N = 0.4$ (1), 0.7 (2), 1.1 (3) along [10] versus distance (a). Magnetic structure factor $S^z(q)$ along [10] at $\eta = 0.75$, $\delta = 0.3$ (b) and $S^z(Q)$ at $Q = (\pi,\pi)$, $\eta = 0.75$, $\delta = 0$ (1), 0.2 (2), 0.4 (3) versus temperature (c).

disordered ground state described by a resonating valence bond function. The exchange anisotropy contributes to rise of the stability of antiferromagnetic order.

More exact results may be obtained by Monte Carlo simulation. Correlation radius depends on temperature as a $\xi \sim \exp(-J/T)$ in two-dimensional isotropic quantum AFM. To eliminate the influence of finite lattice size $\xi \gg L$ using by MC simulation we restrict to investigation of AFM with exchange anisotropy, that is very strong in the single crystals of $Cu_3B_2O_6$. Magnetic structure in the ground state and magnetic properties are analyzed on the basis of spin–spin correlation function $\langle S^z(0)S^z(r)\rangle$, sublattice magnetization m_s, energy, susceptibility and magnetic structure factor $S^z(q) = 1/N\sum_r\langle S^z(0)S^z(r)\rangle\exp(-iqr)$.

The exchange alternating enhances the quantum fluctuations on the spin pairs denoted by the wavy line (Fig. 1) that causes a decreasing spin–spin correlation at the distance $r/a = 1, 2$ as compared to $r/a = 3$ as shown in Fig. 2(a). The distance between spins located in the square corner is $r/a = 3$ and strong spin–spin correlation between them indicates to existing of antiferromagnetic order between them. As a result of calculation of magnetic structure factor the additional satellites in the $S^z(q)$ at $q = \pi/3$ are discovered, the magnitude of which is $S^z(q = \pi/3)/S^z(q = \pi) = 0.1$–$0.2$ (Fig. 2(b)). Neel temperature is determined from the magnetic structure factor of $S^z(Q)$ at $Q = (\pi,\pi)$ (Fig. 2(c)) calculated at the various parameters of exchange alternating. The tails in the $S^z(Q)$ dependence are due to the finite lattice size using in the MC simulation. Normalized Neel temperature is plotted in Fig. 3 at various values of the exchange anisotropy. Dependence of T_N versus alternation is well fitted by $T_N(\delta)/T_N(0) = 1,7(1-\delta)^2/(1+\delta) \simeq zS(S+1)J_{\text{eff}}$ at $\delta \geqslant \delta_c = 0.3$.

Susceptibility of anisotropic antiferromagnetic sharp increases with temperature rise and maximum is observed in vicinity of Neel temperature (see Fig. 4). Such behavior is similar to anisotropic

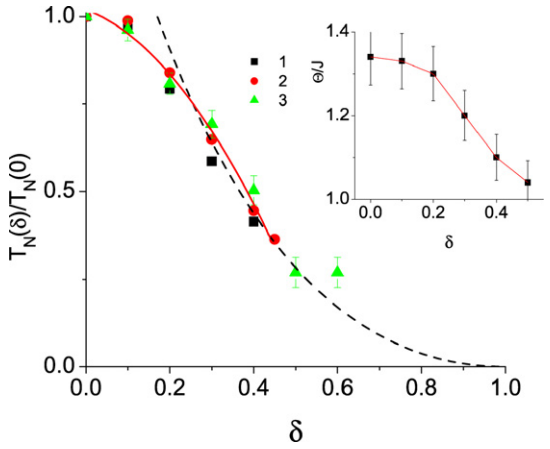

Fig. 3. Normalized Neel temperature at exchange anisotropy $\eta = 0.25$ (1), 0.75 (2), 0.95 (3) versus exchange alternation. Fitting function $T_N(\delta)/T_N(0) = 1.7(1-\delta)^2/(1+\delta)$ is plotted by dot line and fit polynomial $T_N(\delta)/T_N(0) = 1 - 0.5\delta - 2.3\delta^2$ is solid line. Paramagnetic Curie temperature at $\eta = 0.25$ versus exchange alternation are given in the insert.

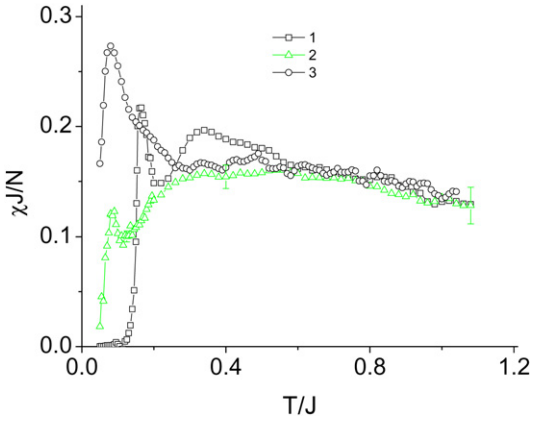

Fig. 4. Susceptibility of antiferromagnetic at $\eta = 0.75$, $\delta = 0$ (1), 0.4 (2), 0.8 (3) versus temperature.

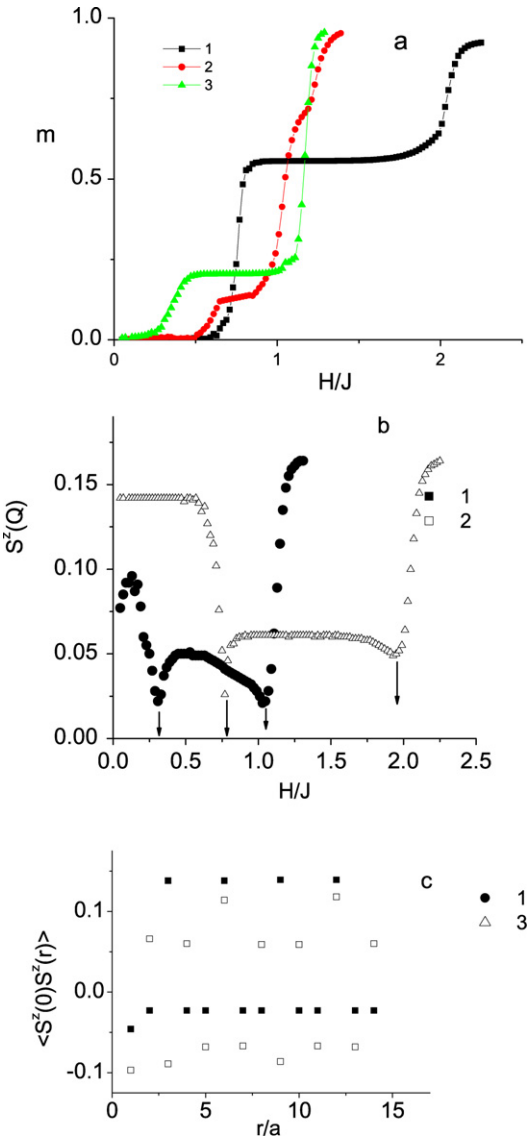

Fig. 5. Magnetization (m) (a) and magnetic structure factor (b) versus magnetic field at various parameters of exchange alternating $\eta = 0.75$; $\delta = 0$ (1), 0.4 (2), 0.8 (3). Spin-spin correlation function at $\eta = 0.75$; $\delta = 0$, $H/J = 0.1$ (1), 1 (2) versus distance.

quasi-one-dimensional antiferromagnet. Sharp and broad maximum are associated with disappearing LRMO and short range magnetic order at T_{χ_m}. Quantum fluctuation suppress the antiferromagnetic order and lead to rise of the relation of $T_{\chi_m}/T_N = (2\text{–}5)$ at variation of exchange alternation from $\delta = 0$ to $\delta = 0.5$. The susceptibility is follow the Curie–Weiss law at high temperatures and expressed as $\chi = C/(T - \Theta)$, where Θ is paramagnetic Curie temperature, that monotonely decreasing at increasing exchange alternating as shown in Fig. 3.

Exchange anisotropy for $Cu_3B_2O_6$ may be estimated from the $\mu_{\text{eff}} = g\mu_B\sqrt{(S(1+S))}$ effective Bohr magneton, where g-factor is anisotropic value. From the data fitting in the temperature range between 150 and 300 K, the $\mu_{\text{eff}}(\| bc) = 1.91\mu_B$ and $\mu_{\text{eff}}(\perp bc) = 2.36\mu_B$ [9]. Exchanges in plane bc and perpendicular to bc differ in value $J(\perp bc)/J(\| bc) = (2.36/1.91)^2 = 1.53$. On the basis of comparison calculated data with experimental results $T_N/\Theta = 0.02$, $T_{\chi_m}/T_N = 4$ the alternating exchange is determined as $\delta \simeq 0.55$. Exchange in pairs exceed exchange in vertex of square approximately in three time for $Cu_3B_2O_6$. Estimated modulation of antiferromagnetic order is agreement with the interpretation of NMR measurements [10] for $Cu_3B_2O_6$ suggesting existence of the modulated spin density wave. The exchange interaction found from the fitting analysis using the function of $C = \gamma T + \beta T^3$ for quasi-one-dimensional system above T_N is $J = 277$ K and for quasi-two-dimensional system is $J = 24$ K [9]. Exchange $J = 136$ K is estimated from the temperature dependence of the susceptibility that obtained on basis of clustering [8]. Our computing gives $J^{\text{MC}} = 190$ K.

The $m(H)$ dependence reveals plateau, plotted in Fig. 5, that arises from two local anisotropic exchange fields E_1, E_2 in the unit cell. In this case the two spin-flop field $H_1 = J\sqrt{(1-(1-\eta)^2)}$, $H_2 = 2J(1-\delta)\sqrt{(1-(1-\eta)^2)}$ exist if interaction between spins S_1, S_2 located in these fields is ignore. Interaction between S_1, S_2 spins induces the noncollinear spin configuration in the field region of $H_1 < H < H_2$. Wave vector of structure determined from spin–spin correlation function (see Fig. 5(b)) is changed from $q = (\pi, \pi)$ to $q = (\pi/3, \pi/3)$ in the vicinity H_1 field. Maximum of magnetic structure factor $S(q)$ attains at $q = (0, 0)$ above H_2 field. Small magnetization value for $Cu_3B_2O_6$ is associated with a little magnetic field of 30 T as compared to spin-flop field $H_{\text{sf}} \sim 110$ T found in our computation. The spins located in small exchange fields are full polarized by external magnetic field in limit case $\delta \to 1$. Saturation magnetization is equal to weight of the spins $m = W_1 = 0.2\mu_B$, that agree with MC result given in Fig. 5.

3. Conclusion

In summary, we analyzed Heisenberg model with anisotropic antiferromagnetic exchange and specifical topology of the coupling constant in order to investigate the type of magnetic structure. As a result of calculations of magnetic structure factor we proved stability of the long range antiferromagnetic order as compared to spin liquid state. Dependence of Neel temperature and paramagnetic Curie temperature versus alternating exchange have been found. For $Cu_3B_2O_6$ the value of exchange in pair of spins exceeds exchange interaction in the vertex square in three time. Antiferromagnetic with cluster ordering reveals plateau in field dependence of magnetization that attributes to formation of modulated structure.

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