Spatially Anisotropic Spin $J_1 - J_2$ Heisenberg Model for an Antiferromagnetic Square Lattice: Phase Diagrams

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The new magnetic materials such as the layered oxide high-temperature superconductor can be well described by the Heisenberg spin model with nearest-neighbor coupling J_1 and next-nearest-neighbor coupling J_2. A generalization of the $J_1 - J_2$ model is the $J_1^x - J_1^y - J_2$ model where the nearest-neighbor bonds have different strengths J_1^x and J_1^y in the x and y directions, respectively. The effect of the couplings J_2 and J_1^y on the antiferromagnetic Néel state is investigated within the quantum many-body Green function method.

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1. Introduction

The physics of two-dimensional Heisenberg antiferromagnet continues to attract considerable attention due to the discovery and availability of new magnetic materials such as a new class of iron pnictide superconductors ΛFe_2As_2 (Λ = Ba, Ca, Sr) with transition temperature T_c reaching 55 K [1]. These systems can be well described by the spatially anisotropic $J_1^x - J_1^y - J_2$ Heisenberg model with nearest-neighbour (NN) exchanges J_1^x along the x axis, J_1^y along the y axis, and with next-nearest-neighbour (NNN) exchange J_2 along the diagonals in the xy plane [2, 3]. The competition between NN and NNN interactions is characterized by the parameter $\alpha = J_2/J_1$, and the spatial anisotropy is characterized by the parameter $\eta = J^y/J^x$. It is now well known that the quantum spin-$\frac{1}{2}$ antiferromagnetic model on the square lattice exhibits new types of magnetic order and novel quantum phases [4, 5]. For $J_2 = 0$ the ground state is antiferromagnetically ordered at zero temperature. The addition of next-nearest-neighbor interaction induces a strong frustration and breaks the antiferromagnetic order at some critical value α_c. It has been found that a paramagnetic phase exists between α_{c1} and α_{c2}. For $\alpha < \alpha_{c1}$ the square lattice is antiferromagnetically ordered whereas for $\alpha > \alpha_{c2}$ a collinear antiferromagnetic stripe phase emerges. The effects of quantum fluctuations due to spatial anisotropy and frustration between the nearest neighbors and next-nearest neighbors of the quantum spin-$\frac{1}{2}$ Heisenberg antiferromagnet on a square lattice was investigated within the second-order spin-wave expansion in [6].

Inelastic magnetic neutron scattering experiments showed that the $J_1^x - J_1^y - J_2$ Heisenberg model quite well describes spin waves of $CaFe_2As_2$ [7]. The spin wave excitation spectrum in the anisotropic $J_1^x - J_1^y - J_2$ Heisenberg model with parameters close to those found in ΛFe_2As_2 compounds and Néel temperature was calculated within many-body Green function theory in [8].

We present in this work a study of magnetic phase diagrams and the effect of quantum fluctuations on the magnetization in the spatially antiferromagnetic Heisenberg model on the square lattice for various NN and NNN exchange interactions. We use the many-body double-time Green function method for arbitrary spin S [9–11].

2. Model Hamiltonian and method

According to the Mermin–Wagner theorem [12], the two-dimensional antiferromagnetic Heisenberg model with exchange interaction alone cannot show finite magnetization. In order to obtain finite magnetization, one can introduce anisotropies. We use the exchange anisotropy.

Consider the Hamiltonian

$$H = \tfrac{1}{2} \sum_{\langle ij \rangle} J_{ij} (S_i^- S_j^+ + S_i^z S_j^z) + \tfrac{1}{2} \sum_{\langle ij \rangle} D_{ij}^z S_i^z S_j^z, \qquad (1)$$

where the exchange interaction and exchange anisotropy strengths are positive ($J_{ij} > 0$ and $D_{ij}^z > 0$).

The equation of motion for the Green function $G_{ij}^{(l)}(\omega) = \langle\langle S_i^+ | (S_j^z)^l S_j^- \rangle\rangle_\omega$ in energy space is written as

$$\omega G_{ij}^{(l)}(\omega) = [S_i^+, (S_j^z)^l S_j^-] \delta_{ij} + \langle\langle [S_i^+, H] | (S_j^z)^l S_j^- \rangle\rangle_\omega, \qquad (2)$$

where $l \leq 2S - 1$ is integer, necessary for dealing with higher spin values S.

Using the spin commutator relations, one obtains

$$[S_i^+, H] = \sum_i J_{ik} (S_i^z S_k^+ - S_k^z S_i^+) - \sum_i D_{ik}^z S_k^z S_i^+. \quad (3)$$

The equation of motion is then

$$\omega G_{ij}^{(l)}(\omega) = [S_i^+, (S_j^z)^l S_j^-] \delta_{ij}$$

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$$-\sum_k D^z_{ik}\langle\langle S^z_k S^+_i|(S^z_j)^l S^-_j\rangle\rangle_\omega$$
$$+\sum_k J_{ik}(\langle\langle S^z_i S^+_k|(S^z_j)^l S^-_j\rangle\rangle_\omega$$
$$-\langle\langle S^z_k S^+_i|(S^z_j)^l S^-_j\rangle\rangle_\omega). \quad (4)$$

We adopt the random phase approximation (RPA) based on the Tyablikov self-consistent approach for the higher order of the Green functions occurring on the right-hand side

$$\langle\langle S^z_i S^+_k|(S^z_j)^l S^-_j\rangle\rangle_\omega \cong \langle S^z_i\rangle\langle\langle S^+_k|(S^z_j)^l S^-_j\rangle\rangle_\omega,$$
$$\langle\langle S^z_k S^+_i|(S^z_j)^l S^-_j\rangle\rangle_\omega \cong \langle S^z_k\rangle\langle\langle S^+_i|(S^z_j)^l S^-_j\rangle\rangle_\omega. \quad (5)$$

This leads to the equation

$$\left(\omega + \sum_k (J_{ik} + D^z_{ik})\langle S^z_k\rangle\right) G^{(l)}_{ij}(\omega)$$
$$-\langle S^z_i\rangle \sum_k J_{ik} G^{(j)}_{kj}(\omega) = Z^{(l)}_{ij}\delta_{ij}, \quad (6)$$

where $Z^{(l)}_i$ is the inhomogeneity term

$$Z^{(l)}_i \equiv [S^+_i, (S^z_i)^l S^-_i] = 2\left\langle (S^z_i - 1)^l S^z_i\right\rangle$$
$$+\left\langle \{(S^z_i - 1)^l - (S^z_i)^l\}\{S(S+1) - S^z_i - (S^z_i)^2\}\right\rangle. \quad (7)$$

To describe the antiferromagnetic (AFM) long-range order (LRO) in the square lattice, we consider the $J^x_1 - J^y_1 - J_2$ Heisenberg model. We write the Hamiltonian for the model in the form

$$H = \tfrac{1}{2}\sum_{i,j} J_{1,ij} S_i \cdot S_j + \tfrac{1}{2}\sum_{i,j} J_{2,ij} S_i \cdot S_j$$
$$+\tfrac{1}{2}\sum_{i,j} D^z_{ij} S^z_i S^z_j, \quad (8)$$

where the lattice sites are denoted by $i \equiv r_i$ and $j \equiv r_j$. The exchange interactions are

$$J_{1,ij} = J^x_1 \delta_{r_j, r_i \pm A_x} - J^y_1 \delta_{r_j, r_i \pm A_y},$$
$$J_{2,ij} = J_2 \delta_{r_j, r_i \pm d_{xy}}, \quad (9)$$

where J^x_1 and J_{1y} are the respective nearest-neighbour in-plane exchange interactions along the x and y axes, J_2 is the next-nearest-neighbour interaction along the diagonal $d_{xy} = A_x \pm A_y$ in the plane. The last term represents the exchange anisotropy. Taking inelastic magnetic neutron scattering experiments into account [13, 14], we assume *antiferromagnetic* coupling $J^x_1 > 0$ in the x axis and $J_2 > 0$ in the diagonal, where $J_2 < J^x$. Along the y axis there is *ferromagnetic* coupling: $J^y_1 > 0$. For these interaction values, there is no frustration.

It is convenient to describe the AFM-ordered Néel state as the two (A, B) sublattice model. The Green function method for the AFM Heisenberg model for spin $S = 1/2$ was considered in detail in [15].

We introduce the sublattice indices (i_m, j_n), where $(m, n) = (A, A)$, (B, A), (A, B) and (B, B). In the case of the approximation of the two sublattices we have to determine four Green functions that correspond to the four pairs of indices (m, n): $G^{(l)}_{i_A J_A}(\omega) = \langle\langle S^+_{i_A}|(S^z_{J_A})^l S^-_{J_A}\rangle\rangle_\omega$, $G^{(l)}_{i_B J_A}(\omega) = \langle\langle S^+_{i_B}|(S^z_{J_A})^l S^-_{J_A}\rangle\rangle_\omega$, $G^{(l)}_{i_B J_A}(\omega) = \langle\langle S^+_{i_B}|(S^z_{J_A})^l S^-_{J_A}\rangle\rangle_\omega$, $G^{(l)}_{i_A J_B}(\omega) = \langle\langle S^+_{i_A}|(S^z_{J_B})^l S^-_{J_B}\rangle\rangle_\omega$, and $G^{(l)}_{i_B J_A}(\omega) = \langle\langle S^+_{i_B}|(S^z_{J_A})^l S^-_{J_A}\rangle\rangle_\omega$. As $\langle S^z_{i_B}\rangle = -\langle S^z_{i_A}\rangle$ for an antiferromagnet, the four equations of motion decouple to two identical pairs of equations, which determine $\langle S^z_{i_A}\rangle$ or $\langle S^z_{i_B}\rangle$, respectively. Before replacing $\langle S^z_{i_B}\rangle$ by $-\langle S^z_{i_A}\rangle$, the equations for $G^{(l)}_{i_A j_A}$ and $G^{(l)}_{i_B j_A}$ are

$$\omega G^{(l)}_{i_A, j_A} + \sum_{k_A \neq i_A} \langle S^z_{k_A}\rangle[(J_2)_{i_A k_A} + D^z_{i_A k_A}] G^{(l)}_{i_A j_A}$$
$$-\sum_{k_A \neq j_A} \langle S^z_{i_A}\rangle (J_2)_{k_A j_A} G^{(l)}_{k_A j_A}$$
$$+\sum_{k_B} \langle S^z_{k_B}\rangle[(J^x_1)_{i_A k_B} - (J^y_1)_{i_A k_B} + 2D^z_{i_A k_B}] G^{(l)}_{i_A j_A}$$
$$-\sum_{k_B} \langle S^z_{i_A}\rangle[(J^x_1)_{i_A k_B} - (J^y_1)_{i_A k_B}] G^{(l)}_{k_B j_A} = Z^{(l)}_{i_A}, \quad (10)$$

$$\omega G^{(l)}_{i_B, j_A} + \sum_{k_B \neq i_B} \langle S^z_{k_B}\rangle[(J_2)_{i_B k_B} + D^z_{i_B k_B}] G^{(l)}_{i_B j_A}$$
$$-\sum_{k_B \neq j_A} \langle S^z_{i_B}\rangle (J_2)_{i_B j_B} G^{(l)}_{k_B j_A}$$
$$+\sum_{k_A} \langle S^z_{k_A}\rangle[(J^x_1)_{i_B k_A} - (J^y_1)_{i_B k_A} + 2D^z_{i_B k_A}] G^{(l)}_{i_B j_A}$$
$$-\sum_{k_A} \langle S^z_{i_B}\rangle[(J^x_1)_{i_B k_A} - (J^y_1)_{i_B k_A}] G^{(l)}_{i_A j_A} = 0. \quad (11)$$

The Fourier transforms to momentum space for the sublattices each consisting of $N/2$ lattice sites are

$$G^{(l)}_{mn}(\boldsymbol{q}) = \frac{2}{n}\sum_{i_m j_n} G^{(l)}_{i_m j_n} \mathrm{e}^{-\boldsymbol{q}\cdot(A_{i_m} - A_{j_n})},$$
$$G^{(l)}_{i_m j_n} = \frac{2}{n}\sum_{\boldsymbol{q}} G^{(l)}_{mn}(\boldsymbol{q}) \mathrm{e}^{\boldsymbol{q}\cdot(A_{i_m} - A_{j_n})}, \quad (12)$$

$$\frac{2}{n}\sum_{\boldsymbol{q}} \mathrm{e}^{-\boldsymbol{q}\cdot(A_{i_m} - A_{i_n})} = \delta_{i_m j_n},$$
$$\frac{2}{n}\sum_{\boldsymbol{q}} \mathrm{e}^{\mathrm{i}(\boldsymbol{q} - \boldsymbol{q}')\cdot A_{i_m}} = \delta_{\boldsymbol{q}\boldsymbol{q}'}, \quad (13)$$

where the subscripts (i_m, j_n) in Eqs. (12) and (13) now denote sublattice indices and not lattice sites. After replacing $\langle S^z\rangle_B$ by $-\langle S^z\rangle_A$, the equations for $G^{(l)}_{AA}(\boldsymbol{q})$ and $G^{(l)}_{BA}(\boldsymbol{q})$ are

$$\omega + \langle S^z_A\rangle\left(J_2(0) - J_2(\boldsymbol{q}) - [J^x_1(0) + J^y_1(0)]\right) G^{(l)}_{AA}(\boldsymbol{q})$$
$$-\langle S^z_A\rangle[J^x_1(q_x) - J^y_1(q_y)] G^{(l)}_{BA}(\boldsymbol{q}) = Z_A, \quad (14)$$
$$\omega - \langle S^z_A\rangle\left(J_2(0) - J_2(\boldsymbol{q}) - (J^x_1(0) + J^y_1(0))\right) G^{(l)}_{BA}(\boldsymbol{q})$$
$$+\langle S^z_A\rangle(J^x_1(q_x) - J^y_1(q_y)) G^{(l)}_{AA}(\boldsymbol{q}) = 0. \quad (15)$$

We have used the following denotations: $(J^x_1)_{AB} = (J^x_1)_{BA} \equiv J^x_1$, $(J^y_1)_{AB} = (J^y_1)_{BA} \equiv J^y_1$, $(J_2)_{AA} = (J_2)_{BB} \equiv J_2$, $D^z_{AA} = D^z_{BB} = D^z_{AB} = D^z_{BA} \equiv D^z$, and $J_2(0) = 2(J_2 + D^z)$, $J^x_1(0) = 2(J^x_1 + D^z)$, $J^y_1(0) = 2(-J^y_1 + D^z)$. In this case for a square lattice with lattice constant $a = 1$ we have

$$J_2(\boldsymbol{q}) = 4J_2\cos(q_x)\cos(q_y),\ \ J^x_1(q_x) = 2J^x_1\cos(q_x),$$
$$J^y_1(q_y) = 2J^y_1\cos(q_y).$$

We express the equations of motions (14), (15) in the matrix form

$$\begin{pmatrix} \omega + \langle S_A^z\rangle[J_2(0) - J_2(\boldsymbol{q}) - J_1(0)] & -\langle S_A^z\rangle[J_1(\boldsymbol{q})] \\ \langle S_A^z\rangle[J_1(\boldsymbol{q})] & \omega - \langle S_A^z\rangle[J_2(0) - J_2(\boldsymbol{q}) - J_1(0)] \end{pmatrix}$$
$$\times \begin{pmatrix} G_{AA}^{(l)}(\boldsymbol{q}) \\ G_{BA}^{(l)}(\boldsymbol{q}) \end{pmatrix} = \begin{pmatrix} Z_A^{(l)} \\ 0 \end{pmatrix}, \tag{16}$$

where $J_1(0) = J_1^x(0) + J_1^y(0)$, $J_1(\boldsymbol{q}) = J_1^x(q_x) - J_1^y(q_y)$.

We obtain from the two equations in (16) the following Green function $G_{AA}^{(n)}(\omega, \boldsymbol{q})$:

$$G_{AA}^{(l)}(\boldsymbol{q}) = \left\{ Z_A^{(l)}[\omega - \langle S_A^z\rangle(J_2(0) - J_2(\boldsymbol{q}) - J_1(0)] \right\}$$
$$\Bigg/ \begin{vmatrix} \omega + \langle S_A^z\rangle[J_2(0) - J_2(\boldsymbol{q}) - J_1(0)] & -\langle S_A^z\rangle J_1(\boldsymbol{q}) \\ \langle S_A^z\rangle J_1(\boldsymbol{q}) & \omega - \langle S_A^z\rangle[J_2(0) - J_2(\boldsymbol{q}) - J_1(0)] \end{vmatrix} \tag{17}$$

with the poles

$$\omega_\pm = \pm\langle S_A^z\rangle[J_2(0) - J_1(0)]$$
$$\times\sqrt{1 + \frac{J_2^2(\boldsymbol{q}) - J_1^2(\boldsymbol{q}) - 2J_2(\boldsymbol{q})}{J_2(0) - J_1(0)}}. \tag{18}$$

From the spectral theorem, after integrating over the first Brillouin zone, the following equation for the correlation function $\langle (S_A^z)^l S_A^- S_A^+\rangle$ for the sublattice A results

$$\langle (S_A^z)^l S_A^- S_A^+\rangle = Z_A^{(l)}\Phi_A, \tag{19}$$

where

$$\langle (S_A^z)^l S_A^- S_A^+\rangle \equiv S(S+1)\langle (S_A^z)^l\rangle - \langle (S_A^z)^{l+1}\rangle$$
$$-\langle (S_A^z)^{l+2}\rangle \tag{20}$$

and

$$\Phi_A = -\frac{1}{2} + \frac{1}{2\pi^2}\int_0^\pi\int_0^\pi \frac{J_2(\boldsymbol{q}) - J_2(0) + J_1(0)}{\omega_+/\langle S_A^z\rangle}$$
$$\times\coth\frac{\omega_+}{2kT}\,\mathrm{d}q_x\,\mathrm{d}q_y. \tag{21}$$

The inhomogeneity term $Z_A^{(l)}$ is defined by relation (7).

For the spin $S = 1/2$, the average value of $\langle (S_A^z)^2\rangle = 1/4$ and Eq. (19) with $l = 0$ provides an equation for the magnetization $\langle S_A^z\rangle$:

$$\langle S_A^z\rangle = \frac{1}{2(1 + 2\Phi_A)}. \tag{22}$$

For the spin $S = 1$, the average value of $\langle (S_A^z)^3\rangle = \langle S_A^z\rangle$ and Eq. (19) with $l = 0, 1$ provides an equation for the magnetization

$$\langle S_A^z\rangle = \frac{1 + 2\Phi_A}{1 + 3\Phi_A + 3\Phi_A^3}. \tag{23}$$

For the spin $S = 3/2$, the average value of $\langle (S_A^z)^4\rangle = 5\langle (S_A^z)^2\rangle/2 - 9/16$ and Eq. (19) with $l = 0, 1, 2$ provides the equation for a magnetization

$$\langle S_A^z\rangle = \frac{1 + 10(\Phi_A + \Phi_A^2)}{2(1 + 2\Phi)[1 + 2(\Phi_A + \Phi_A^2)]}. \tag{24}$$

3. Results

We obtain the saturation sub-lattice magnetization $M(0)$ for the AF ordered phase with several values of α and η from (22), (23) and (24) by numerical evaluation. Figure 1 shows the reduced magnetization $M(0)/S$ for spin $S = 1/2$ with increase in the parameter $\alpha = J_2/J_1^x$ for several values of the spatial anisotropy parameter $\eta = J_1^y/J_1^x = 0, 0.1, 0.3, 0.45$ and $d = D/J_1^x = 0.5$. We find that the saturation magnetization steadily decreases with increase in the parameter α and then suddenly drops to zero at a certain value of the parameter α_c. The parameter α_c decreases with increasing parameter η. For example (curve (a)), for the pairs ($\alpha < 0.241, \eta = 0.1$) the system is in the AFM LRO, as can be seen in Fig. 2 where the temperature dependence of magnetizations are plotted for different values of $\alpha = J_2/J_1^x$ when $\eta = J_1^y/J_1^x = 0.1$. For the pair ($\alpha > 0.241, \eta = 0.1$), the system is in the paramagnetic state. Saturation magnetizations jump to zero at the critical values $\alpha_c = J_{2c}/J_1^x$ for each value of $\eta = J_1^y/J_1^x$ from the interval: $\eta \in \langle 0, 0.5)$.

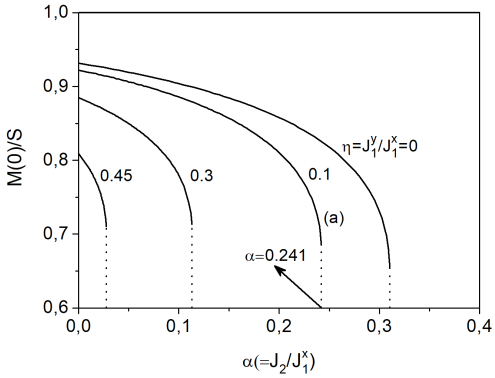

Fig. 1. The reduced sublattice magnetizations $M(0)/S$ for spin $S = 1/2$ are plotted with increase in the parameter $\alpha = J_2/J_1^x$ for several values of the ferromagnetic spatial anisotropy parameter $\eta = J_1^y/J_1^x = 0, 0.1, 0.3, 0.45$ and $d = 0.5$.

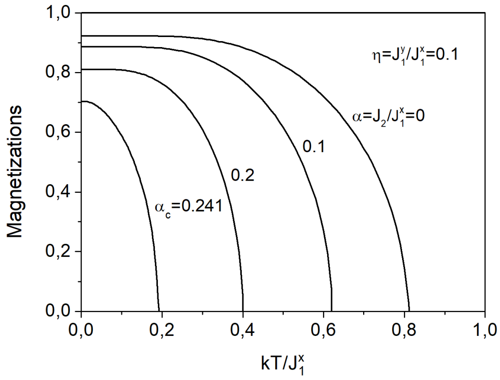

Fig. 2. The temperature dependence of sublattice magnetizations for spin $S = 1/2$ are plotted for different values of $\alpha = J_2/J_1^x = 0,\ 0.1,\ 0.2,\ 0.241$ when $\eta = J_1^y/J_1^x = 0.1$.

The ground-state magnetization $M(0)$ per spin is reduced from its classical value $M(0)_{class}$ by spin fluctua-

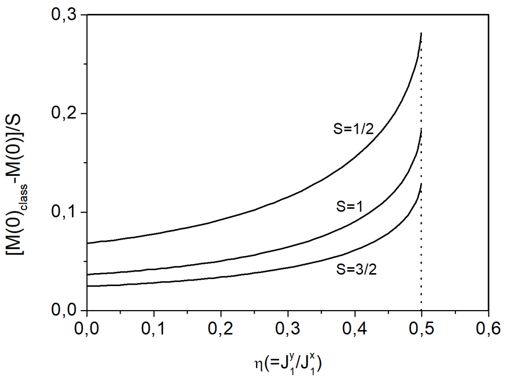

Fig. 3. Spin deviation $\Delta = (M(0) - M(0)_{class})/S$ from the classical value is plotted as a function of $\eta(= J_1^y/J_1^x)$ for the AF ordered phase along the y axis (with no NNN coupling, i.e. $\alpha = 0$).

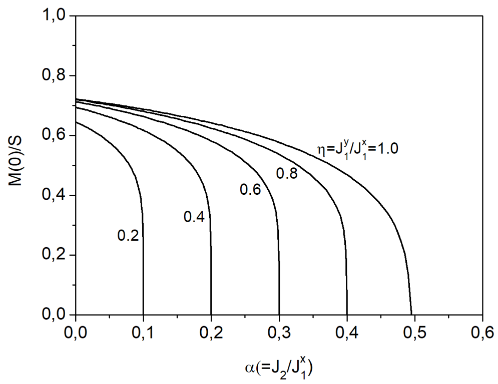

Fig. 4. The reduced sublattice magnetizations $M(0)/S$ are plotted for AF ordered phase with frustration for spin $S = 1/2$ with increase in the parameter $\alpha = J_2/J_1^x$ for different values of the antiferromagnetic spatial anisotropy parameter $\eta = J_1^y/J_1^x = 0.2$, 0.4, 0.6, 0.8, and 1.0.

tions. In Fig. 3 the "spin reduction" $(M(0) - M(0)_{class})/S$ is plotted for spins $S = 1/2$, $S = 1$ and $S = 3/2$. The spin fluctuations increase with values of η and decrease with increase of spin S. The long-range order disappears for anisotropy parameter $\eta > 0.5$.

The result in the case of frustration, i.e. when J_1^y is the antiferromagnetic coupling, is completely different from the case when there is no frustration. This can be seen in Fig. 4, where the reduced saturation sublattice magnetization $M(0)/S$ with spin $S = 1/2$ is presented as a function of the parameter α for AFM LRO for different values of spatial anisotropy η. Saturation magnetizations become continuous to zero at the critical values $\alpha_c = J_{2c}/J_1^x$ for each value of $\eta = J_1^y/J_1^x$ from the interval: $\eta \in \langle 0, 1\rangle$. We can see from Fig. 4 that the spin fluctuations decrease with increasing parameter anisotropy η. This result is opposite to the case when there is no frustration.

4. Conclusions

We have employed the Green function theory to calculate the phase diagrams in the spatially anisotropic antiferromagnetic $J_1^x - J_1^y - J_2$ Heisenberg model for the square lattice with arbitrary spins S. We assumed antiferromagnetic couplings: $J_1^x > 0$ in the x axis, $J_2 > 0$ in the diagonal and a ferromagnetic coupling along the y axis: $J_1^y > 0$. For these interaction values, there is no frustration. In this case, we observed the AFM LRO only for the parameters α and η from a certain interval. Saturation magnetizations jump to zero at the critical values $\alpha_c = J_{2c}/J_1^x$ for value of $\eta = J_1^y/J_1^x$ only from the interval: $\eta \in \langle 0, 0.5)$. The LRO disappears for the anisotropy parameter $\eta > 0.5$. In this case, the spin fluctuations increase with increase of parameter anisotropy η.

When the exchange coupling along the y axis is antiferromagnetic ($J_1^y < 0$), then there is frustration. Saturation magnetizations become continuous to zero at the critical values $\alpha_c = J_{2c}/J_1^x$ for each value of $\eta = J_1^y/J_1^x$ from the interval: $\eta \in \langle 0, 1\rangle$. In this case, the spin fluctuations decrease with increase of parameter anisotropy η. This result is opposite to the case when there is no frustration.

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