THE QUADRUPOLE PHASE IN A MAGNET WITH ANISOTROPIC BIQUADRATIC EXCHANGE

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UDC 537.61

The quadrupole phase in a uniaxial magnet with single-ion anisotropy of the easy-plane type and the anisotropic biquadratic exchange interaction is investigated. The case where the spin at a site equals unity is considered. A new method for the calculation of the spectrum of spin excitation modes at finite temperatures is proposed. It is shown that, in the limit case where $T = 0$, the results obtained are in full compliance with those reported by other researchers. The expressions for two branches of the spin excitation spectrum at finite temperatures are obtained, and the stability conditions for the spectrum modes are determined. The temperature, at which the stability of the spectrum modes breaks, is determined analytically as a function of the Hamiltonian parameters. It is proved that, under certain conditions as the temperature is lowered, the stability of the modes of the spin excitation spectrum first breaks and then is restored. The existence of a metastable state is predicted.

1. Introduction

Magnetic compounds, in which the constants of both SIA and BEI are of the same order as the usual exchange interaction, were discovered in the 1970s [1–6]. However, the theoretical study of such systems met difficulties, because the inclusion of both SIA and BEI in the Hamiltonian results in the fact that the algebra of the operators describing a system state goes beyond the limits of the SU(2) algebra. The attempts to overcome these difficulties continue to date with a greater or less success [7–29]. Unfortunately in almost all cases where the researchers accounted for BEI, the calculations were restricted to the consideration of isotropic BEI.

In works [7–9], the authors studied the magnets with an isotropic BEI in the mean field approximation and came to a conclusion that, for a certain range of the Hamiltonian parameters, the ferromagnetic and antiferromagnetic phases are unstable against the appearance of the quadrupole phase (QP) with $\langle S^Z \rangle = 0$. As was shown in work [10], a new type of ordering occurs in QP at $T = 0$, for which $S^Z = 0$ at each site to within zero-point oscillations. The spin-wave approximation was built for the $S = 1$ case, and it was shown that, for the case where the SIA and BEI constants, J_0 and K_0, respectively, satisfy the condition $K_0 > J_0 > 0$, QP becomes stable.

In work [22], the finite-temperature studies of the magnets with SIA and isotropic BEI were carried out for the case of $S = 1$. The possible kinds of ordered phases at $T = 0$ were determined, among which two phases were shown to display a quadrupole ordering with $\langle S^Z \rangle = 0$. At the same time, for the first phase, QP_1, the equality $\langle (S^Z)^2 \rangle = 1$ is valid, which means that the ordering occurs in both directions along the z axis. On the other hand, it was shown that $\langle (S^Z)^2 \rangle = 0$ for the second phase, QP_2, which corresponds to the ordering in the xy plane. At the zero temperature, the ordering in QP_1 doesn't differ from the antiferromagnetic one, i.e. it is inhomogeneous. It is also proved that, apart from the usual ferromagnetic phase FM_Z with the ordering along the z axis, a phase QFM_X can exist and is characterized by the ferromagnetic ordering along the x axis and the quadrupole one within the plane that forms a certain angle with the xy plane, i.e. the phase with a spontaneously broken symmetry.

In work [25], the results of the low-temperature studies were reported for the magnets with an anisotropic BEI. The possible types of homogeneous ordering were determined for the cases of the finite and zero external magnetic field h. It is demonstrated that, for the case of $h_Z = 0$, a quadrupole-angular phase (QAP) can exist, for which all average S^α values are equal to zero, but the plane of quadrupole ordering forms a certain angle with the xy plane. This phase, just as the QFM_X one, is a phase with spontaneously broken symmetry. However, contrary to the latter, QAP cannot be formed when BEI is isotropic. In the above work, a spin excitation spectrum was found for each of the ordered phases. It was shown that a quadratic dispersion law, $\omega(\mathbf{k}) = \Delta + \alpha k^2$, was observed in the long-wave limit for QP_2. The phase diagrams, which illustrated the field-induced phase transitions, were built. However, since the investigations were limited to the low-temperature region, it was impossible to observe the temperature evolution of the phase transitions.

The authors of works [28,29] considered the angular phase which appears as a result of the competition between the magnetic and strong crystalline fields in easy-plane magnets. It was shown in [28] that, in such systems, the Landau phenomenological theory is suitable for the description of phase transitions. The results of work [29] demonstrated that the phase transition from the singlet state to the antiferromagnetic one belongs to magnetic phase transitions of the substitution kind.

This paper is aimed at the finite-temperature study of QP_2 for a uniaxial magnet, which is characterized by SIA and anisotropic BEI. Here, we propose the method which makes it possible to find two branches of the spin excitation spectrum, with one of these branches becoming soft at the center of the Brillouin zone for certain values of the Hamiltonian parameters. At the same time, in the long-wave limit, one observes the quadratic dispersion law, $\omega(\mathbf{k}) \sim k^2$, when $h_Z \neq 0$, and the linear one, $\omega(\mathbf{k}) \sim k$, when $h_Z = 0$. It is demonstrated that, with lowering the temperature, the stability of the spectrum modes becomes broken at first under certain conditions, and then it is restored. The phase diagrams in the "phase – temperature" coordinates are constructed.

2. Operation Algebra and System Hamiltonian

In the case where the spin S at a site is equal to 1, the algebra of operators which describe all interactions in the system is the ASU(3) algebra. The generators of this algebra are the tensor operators O_l^m, where l is the tensor rank, and $m = 0, \pm 1, \ldots, \pm l$. A relation between these operators and the spin ones is described by the formulae

$$O_1^0 = S^Z,\ O_1^1 \equiv S^+ = \frac{1}{\sqrt{2}}\left(S^X - iS^Y\right),$$

$$O_1^{-1} \equiv S^- = \frac{-1}{\sqrt{2}}\left(S^X + iS^Y\right),\quad O_2^0 = \left(S^Z\right)^2 - \frac{2}{3},$$

$$O_2^{\pm 1} = -\left(S^Z S^\pm + S^\pm S^Z\right),\quad O_2^{\pm 2} = \left(S^\pm\right)^2. \tag{1}$$

The mean values of the O_l^m operators determine the spin order in the system, i.e. they form the multicomponent order parameter.

The studies will be carried out basing on the Hamiltonian which is invariant under the $\exp\left(i\varphi S^Z\right)$ transformation (here, φ is an arbitrary angle):

$$H = -h_Z \sum_i S_i^Z - \sum_{i,j(i\neq j)} J_{ij}\left[S_i^Z S_j^Z - 2\xi S_i^+ S_j^-\right] +$$

$$+D\sum_i O_{2i}^0 - \sum_{i,j(i\neq j)} K_{ij}\Big(3O_{2i}^0 O_{2j}^O -$$

$$-2\eta O_{2i}^1 O_{2j}^{-1} + 4\zeta O_{2i}^2 O_{2j}^{-2}\Big), \tag{2}$$

where h_Z is the z-component of the external magnetic field, J_{ij} are the exchange interaction constants, K_{ij} are the BEI constants, ξ is the anisotropy constant of the exchange interaction, and η, ζ are the BEI anisotropy constants.

In this work, the calculations are restricted to the case where all the constants of the Hamiltonian are positive. Under these conditions, only the homogeneous (one-sublattice) ordering occurs in the system. In addition, SIA is assumed to be of the easy-plane type.

In QP_2, only the diagonal components of the $\langle S^Z\rangle$ and $\langle O_2^0\rangle$ order parameters are nonzero. Thus, in the molecular field approximation, Hamiltonian (2) takes the form

$$H_0 = -\left(h_Z + 2J_0\langle S^Z\rangle\right)\sum_i S_i^Z + \left(D - 6K_0\langle O_2^0\rangle\right)\sum_i O_{2i}^0, \tag{3}$$

where J_0 and K_0 are:

$$J_0 \equiv \sum_i J_{ij}, K_0 \equiv \sum_i K_{ij}.$$

The atomic energy levels depend on the value of the spin projection on the z axis ($S^Z = 1$, 0, or -1). Thus, we have

$$E_1 = -h_Z - 2J_0\langle S^Z\rangle + \frac{1}{3}D - 2K_0\langle O_2^0\rangle,$$

$$E_0 = -\frac{2}{3}D + 4K_0\langle O_2^0\rangle,$$

$$E_{-1} = h_Z + 2J_0\langle S^Z\rangle + \frac{1}{3}D - 2K_0\langle O_2^0\rangle.. \quad (4)$$

For QP_2, it follows from the $h_Z = 0$ condition that $\langle S^Z\rangle = 0$ and, thus, $E_1 = E_{-1}$. This means that the doublet with $S^Z = \pm 1$ lies above the singlet ground state with $S^Z = 0$. If $h_Z \neq 0$, the doublet splits ($E_1 < E_{-1}$) by the value $2(h_Z + 2J_0\langle S^Z\rangle)$.

Thus, QP_2 can be formed provided that $E_0 < E_1$, which is equivalent to the inequality

$$D - 6K_0\langle O_2^0\rangle > h_Z + 2J_0\langle S^Z\rangle. \quad (5)$$

It should be noted that inequality (5) is just the condition of the QP_2 stability against the transitions to the FM_Z phase. When $h_Z = 0$, condition (5) is fulfilled automatically, which means that the FM_Z phase is not realized.

At the zero temperature ($T = 0; h_Z \neq 0$), the equalities $\langle S^Z\rangle = 0$ and $\langle O_2^0\rangle = -2/3$ are valid for QP_2. On the other hand, when $T \neq 0$, the $\langle S^Z\rangle$ and $\langle O_2^0\rangle$ values are determined by the formulae

$$\langle S^Z\rangle = \frac{\sum_n S_n^Z \exp(E_n/\theta)}{\sum_n \exp(E_n/\theta)}, \quad \langle O_0^2\rangle = \frac{\sum_n O_0^2 \exp(E_n/\theta)}{\sum_n \exp(E_n/\theta)}, \quad (6)$$

where θ is the temperature in the energy units ($\theta = kT$).

With regard for expressions (4), formulae (6) can be rewritten as

$$\langle S^Z\rangle = \frac{2\mathrm{sh}\frac{h_z + 2J_0\langle S^Z\rangle}{\theta} \exp\frac{6K_0\langle O_2^0\rangle - D}{\theta}}{1 + 2\mathrm{ch}\frac{h_z + 2J_0\langle S^Z\rangle}{\theta} \exp\frac{6K_0\langle O_2^0\rangle - D}{\theta}},$$

$$\langle O_2^0\rangle = \frac{1}{3} - \frac{1}{1 + 2\mathrm{ch}\frac{h_z + 2J_0\langle S^Z\rangle}{\theta} \exp\frac{6K_0\langle O_2^0\rangle - D}{\theta}}. \quad (7)$$

The values of $\langle S^Z\rangle$ and $\langle O_2^0\rangle$ grow with increase in θ. At the same time, the value of $\langle O_2^0\rangle$ remains negative.

For the case where $h_Z = 0$, a relation between the temperature θ and the $\langle O_2^0\rangle$ value can be obtained from the second equation of system (7):

$$\theta = \frac{D - 6K_0\langle O_2^0\rangle}{\ln\left[(2 - 6\langle O_2^0\rangle) / (2 + 3\langle O_2^0\rangle)\right]}. \quad (8)$$

3. Spin Excitations

For QP_2 at zero temperature, all spins are aligned in the xy plane, but their alignment within the plane is random and, thus, $\langle S^X\rangle = \langle S^Y\rangle = 0$. At finite temperatures, the spin excitations with $S^Z = 1$ and $S^Z = -1$ appear. They are generated by the Hubbard operators X^{10} and X^{-10}, respectively. Since, in the presence of a magnetic field ($h^Z \neq 0, T \neq 0$), the excitation energy is smaller for $S^Z = 1$ than that for $S^Z = -1$, a finite magnetization ($\langle S^Z\rangle > 0$) appears in the system and grows with increase in the temperature, which completely agrees with system (7). For $h^Z = 0$, the energies of excitations of both kinds are equal, and, thus, $\langle S^Z\rangle = 0$.

To calculate the spin excitation spectrum, we propose a method, whose essence is as follows. We will enumerate the atomic levels f, which correspond to the different values of spin projection on the z axis, by natural numbers n. Let the level with $n = 1$ be the lowest one. Then the Hubbard operators $X_f^{12}, X_f^{13}, \ldots, X_f^{1n}, \ldots$ are the annihilation operators for the spin excitations at the site f, and $X_f^{21}, X_f^{31}, \ldots, X_f^{n1}, \ldots$ are the creation operators for the corresponding excitations. The set of the X_f^{1n} and X_f^{n1} ($n = 2, 3, \ldots$) operators forms an ensemble $\tilde{X}$. With the use of the approximation, which will be described below [see (13)], the commutators of operators from this ensemble with Hamiltonian (2) can be presented as a linear combination of these operators. In the case where the commutators of annihilation operators with the Hamiltonian give only the linear combinations of annihilation operators, we can write

$$\begin{aligned}
&\left[X_f^{12}, H\right] = \\
&= a_{11}X_f^{1(1+1)} + a_{12}X_f^{1(1+2)} + \ldots + a_{1n}X_f^{1(1+n)} + \ldots, \\
&\left[X_f^{13}, H\right] = \\
&= a_{21}X_f^{1(1+1)} + a_{22}X_f^{1(1+2)} + \ldots + a_{2n}X_f^{1(1+n)} + \ldots, \\
&\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \\
&\left[X_f^{1(n+1)}, H\right] = \\
&= a_{n1}X_f^{1(1+1)} + a_{n2}X_f^{1(1+2)} + \ldots + a_{nn}X_f^{1(1+n)} + \ldots, \\
&\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots .
\end{aligned} \quad (9)$$

At the same time, a_{ij} are the expressions which depend on the Hamiltonian parameters. A set of all a_{ij} constitutes a dynamical matrix A_{ij}. It is clear that, if the dynamical matrix is a diagonal one ($a_{ij} = 0$ for $i \neq j$), the a_{ii} values are the energies of spin excitations from the ground state to the states with $n = 2, 3, ...$ at the site f. On the other hand, they are the eigenvalues of the matrix A_{ij}, since it is diagonal. In the **k**-space, the expressions for these eigenvalues coincide with those for the branches of the spin excitation spectrum. If A_{ij} is a non-diagonal matrix, the expressions for its eigenvalues in the **k**-space also coincide with those for the branches of the spin excitation spectrum: $\varepsilon_n = \omega_n$. To find these eigenvalues, a secular equation can be used:

$$\begin{vmatrix} a_{11}-\varepsilon_k & a_{12} & ... \\ a_{21} & a_{22}-\varepsilon_k & ... \\ \multicolumn{3}{c}{\dotfill} \end{vmatrix} = 0. \quad (10)$$

If the dynamical matrix consists of the creation operators for the spin excitations ($X_f^{k1}, X_f^{l1}, ...$), the expressions for its eigenvalues in the **k**-space differ in the sign from the expressions for the spectrum branches: $\varepsilon_n = -\omega_n$.

In the case where the construction of the dynamical matrix requires the use of both the annihilation and creation operators, the expressions for the eigenvalues, which correspond to the former, coincide with those for the corresponding spectrum branches. On the contrary, the expressions for the eigenvalues, which correspond to the latter, differ in the sign from the expressions for the corresponding spectrum branches.

To apply the proposed method to Hamiltonian (2), it is necessary to turn to the Hubbard operators. The relation of the quadrupole operators O_l^m to the Hubbard operators X^{ps} can be expressed as

$$S^Z = X^{11} - X^{-1-1}, \quad S^+ = -X^{10} - X^{0-1},$$

$$S^- = X^{01} + X^{-10}, \quad O_2^0 = X^{11} + X^{-1-1} - \frac{2}{3},$$

$$O_2^1 = X^{10} - X^{0-1}, \quad O_2^{-1} = -X^{01} + X^{-10},$$

$$O_2^2 = X^{1-1}, \quad O_2^{-2} = X^{-11}. \quad (11)$$

At the same time, the initial Hamiltonian (2) should be rewritten as

$$H = -h_Z \sum_i \left(X_i^{11} - X_i^{-1-1}\right) -$$

$$-\sum_{i,j} J_{ij} \Big[\left(X_i^{11} - X_i^{-1-1}\right)\left(X_j^{11} - X_j^{-1-1}\right) +$$

$$+2\xi\left(X_i^{10} + X_i^{0-1}\right)\left(X_j^{01} + X_j^{-10}\right)\Big] +$$

$$+D\sum_i \left(X_i^{11} + X_i^{-1-1} - \frac{2}{3}\right) -$$

$$-\sum_{i,j} K_{ij} \Big[3\left(X_i^{11} + X_i^{-1-1} - \frac{2}{3}\right)\left(X_j^{11} + X_j^{-1-1} - \frac{2}{3}\right) -$$

$$-2\eta\left(X_i^{10} - X_i^{0-1}\right)\left(-X_j^{01} + X_j^{-10}\right) + 4\zeta X_i^{1-1} X_j^{-11}\Big]. \quad (12)$$

Calculating the commutators $\left[X_p^{01}, H\right]$ and $\left[X_p^{-10}, H\right]$ and using the approximation

$$X_i X_j = X_i \langle X_j \rangle + \langle X_i \rangle X_j$$

$$\langle X^{nm} \rangle = 0 \ (n \neq m), \quad (13)$$

we obtain the equation of motion for the operators X_p^{01} and X_p^{-10}, which acquires, after the Fourier transformation, the form

$$\left[X_k^{01}, H\right] = \Big[D - h_Z - 2\langle S^Z \rangle J_0 - 6\langle O_2^0 \rangle K_0 +$$

$$+ \left(\langle S^Z \rangle + 3\langle O_2^0 \rangle\right)\left(\xi J_k + \eta K_k\right)\Big] X_k^{01} +$$

$$+ \left(\langle S^Z \rangle - 3\langle O_2^0 \rangle\right)\left(\xi J_k + \eta K_k\right)\Big] X_k^{-10},$$

$$\left[X_k^{-10}, H\right] = \left(\langle S^Z \rangle - 3\langle O_2^0 \rangle\right)\left(\xi J_k - \eta K_k\right) X_k^{01} +$$

$$+ \Big[-D - h_Z - 2\langle S^Z \rangle J_0 + 6\langle O_2^0 \rangle K_0 +$$

$$+ \left(\langle S^Z \rangle - 3\langle O_2^0 \rangle\right)\left(\xi J_k + \eta K_k\right)\Big] X_k^{-10}, \quad (14)$$

where X_k^{nm}, J_k, and K_k are the Fourier transforms of the quantities X_p^{nm}, J_{ij}, and K_{ij}, respectively.

Consequently, the dynamical matrix has the dimension 2×2 and the solution of the secular equation has two eigenvalues:

$$\varepsilon_k^{1,2} = h_Z + \langle S^Z \rangle (2J_0 - \xi J_k - \eta K_k) \pm$$

$$\pm \Big\{ \left(\langle S^Z \rangle\right)^2 (\xi J_k - \eta K_k)^2 + \left[D - 6\langle O_2^0 \rangle (K_0 - \xi J_k)\right] \times$$

$$\times \left[D - 6\langle O_2^0 \rangle (K_0 - \eta K_k)\right] \Big\}^{1/2}. \quad (15)$$

The operator X_k^{01}, which is the annihilation operator, corresponds to the eigenvalue ε_k^1. On the other hand, the operator X_k^{-10}, which is the creation operator, corresponds to the eigenvalue ε_k^2. For this reason, the $\omega_k^{1,2}$ spectrum branches are determined by the expressions

$$\omega_1 (\mathbf{k}) = \varepsilon_k^1; \;\; \omega_2 (\mathbf{k}) = -\varepsilon_k^2, \quad (16)$$

or

$$\omega_{1,2} (\mathbf{k}) = \left[D - 6\langle O_2^0 \rangle (K_0 - \xi J_k)\right] \times$$

$$\times \left[D - 6\langle O_2^0 \rangle (K_0 - \eta K_k)\right] +$$

$$+ \left(\langle S^Z \rangle\right)^2 (\xi J_k - \eta K_k)^{21/2} \pm$$

$$\pm \left[h_Z + \langle S^Z \rangle (2J_0 - \xi J_k - \eta K_k)\right]. \quad (17)$$

In the low-temperature limit ($T = 0$), we obtain $\langle S^Z \rangle = 0$ and $\langle O_2^0 \rangle = -2/3$. In this case, expressions (17) take the form

$$\omega_{1,2} (\mathbf{k}) = \Big\{ [D + 4 (K_0 - \xi J_k)] \times$$

$$\times [D - 4 (K_0 - \eta K_k)] \Big\}^{1/2} \pm h_Z. \quad (18)$$

Expressions (18) coincide with those obtained by the method of transformation of spin operators to the secondary quantization operators in work [25]. However, contrary to the results of work [25], the method proposed here makes it also possible to easily study both the cases, $T = 0$ and $T \neq 0$.

Since $\omega_1(\mathbf{k}) \geq \omega_2(\mathbf{k})$, the modes of spectrum (17) are stable provided that $\omega_2(\mathbf{k}) > 0$ for all values $\mathbf{k}$.

In the system, the one-sublattice ordering occurs. For this reason for the corresponding values of the Hamiltonian parameters, the $\omega_2(\mathbf{k})$ branch becomes soft at the center of the Brillouin zone, i.e. when $\mathbf{k} = 0$. When $\mathbf{k} \neq 0$, the inequality $\omega_2(\mathbf{k}) > \omega_2(0)$ is valid [30]. Thus, the stability condition for the modes of spectrum (17) for all $\mathbf{k}$ values is $\omega_2(0) > 0$ or

$$\Big\{ [D - 6\langle O_2^0 \rangle (K_0 - \xi J_0)][D - 6\langle O_2^0 \rangle K_0 (1 - \eta)] +$$

$$+ \left(\langle S^Z \rangle\right)^2 (\xi J_0 - \eta K_0)^2 \Big\}^{1/2} - h_Z -$$

$$- \langle S^Z \rangle (2J_0 - \xi J_0 - \eta K_0) > 0. \quad (19)$$

The stability threshold can be calculated as

$$\Big\{ \left[D - 6\langle O_2^0 \rangle (K_0 - \xi\, J_0)\right] \left[D - 6\langle O_2^0 \rangle K_0 (1 - \eta)\right] +$$

$$+ \left(\langle S^Z \rangle\right)^2 (\xi\, J_0 - \eta\, K_0)^2 \Big\}^{1/2} =$$

$$= h_Z + \langle S^Z \rangle (2J_0 - \xi\, J_0 - \eta\, K_0). \quad (20)$$

For the case where condition (20) is valid and $h^Z \neq 0$, the spin alignments in the easy plane and along the z axis are energetically equivalent, i.e. the corresponding excitations have zero energy. The energy of the transitions to the state with the spin alignment opposite to the z axis remains positive.

Since J_k and K_k are the even functions of the wave vector $\mathbf{k}$, the soft mode displays the quadratic dispersion law, $\omega (\mathbf{k}) \sim k^2$, for $h^Z \neq 0$.

If there is no external field ($h_Z = 0$), two branches coincide:

$$\omega_{1,2} \left(\vec{k}\right) =$$

$$= \sqrt{[D - 6\langle O_2^0 \rangle (K_0 - \xi\, J_k)][D - 6\langle O_2^0 \rangle (K_0 - \eta K_k)]}. \quad (21)$$

In this case, the expressions for the stability condition (19) and the stability threshold (20) take the form

$$\left[D - 6\langle O_2^0 \rangle (K_0 - \xi\, J_0)\right] \left[D - 6\langle O_2^0 \rangle K_0 (1 - \eta)\right] > 0, \quad (22)$$

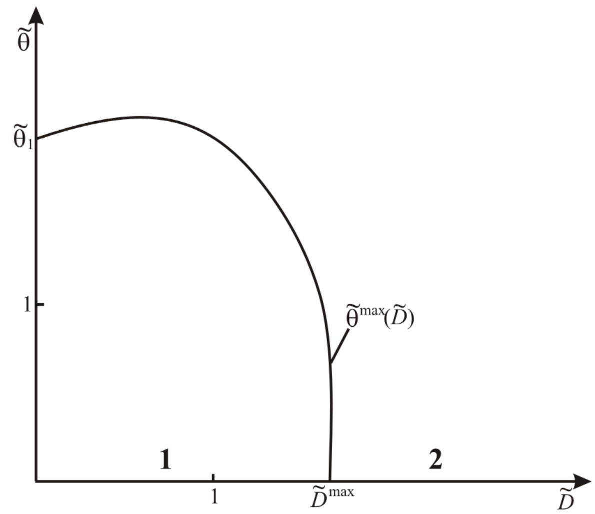

Fig. 1. The stability diagram for an easy-plane magnet with anisotropic BEI for $\beta_{\min} < 1 < \beta_{\max}$: *1* – a phase with the spontaneously broken symmetry; *2* – QP_2. The boundary between the phases is calculated for $\beta_{\max} = 1.4$. In the diagram, $\tilde{D}^{\max} = 1.6$ and $\tilde{\theta}_1 = 1.87$

and

$$\left[D - 6\langle O_2^0\rangle\,(K_0 - \xi\,J_0)\right]\left[D - 6\langle O_2^0\rangle K_0\,(1-\eta)\right] = 0, \quad (23)$$

respectively. If condition (23) is fulfilled, all three possible values of S^Z are energetically equivalent, and, thus, the energies of the transitions to the states with $S^Z = \pm 1$ equal zero.

For the case $h_Z = 0$, the soft mode displays the linear dispersion law, $\omega\,(\mathbf{k}) \sim k$.

4. Phase Boundaries

To analyze the phase boundaries for QP_2, it is pertinent to rewrite condition (22) as

$$\left[D - 6\langle O_2^0\rangle K_0\,(1-\beta_{\max})\right] \times$$

$$\times \left[D - 6\langle O_2^0\rangle K_0\,(1-\beta_{\min})\right] > 0, \quad (24)$$

where the designations

$$\beta_{\max} = \max\{\eta\,;\,\xi J_0/K_0\}, \quad \beta_{\min} = \min\{\eta\,;\,\xi J_0/K_0\} \quad (25)$$

are introduced.

For QP_2, the inequality $\langle O_2^0\rangle < 0$ is valid. Thus, under the conditions that

$$\beta_{\min} \le 1 \text{ and } \beta_{\max} \le 1, \quad (26)$$

the modes of spectrum (21) are stable for the arbitrary positive values of D.

In the case where only the former of inequalities (26) is fulfilled, the stability condition can be written as

$$\langle O_2^0\rangle \;>\; D/6K_0\,(1-\beta_{\max})\,.. \quad (27)$$

Since $\langle O_2^0\rangle$ is a function increasing with the temperature, the stability breaking occurs at

$$\langle O_2^0\rangle \;=\; D/6K_0\,(1-\beta_{\max}) \quad (28)$$

with decrease in the temperature.

With regard for (28), formula (8) gives the relation between the Hamiltonian parameters and the temperature of the stability breaking for the modes of spectrum (21):

$$\theta^{\max} = \frac{\beta_{\max}\,D}{(\beta_{\max}-1)\ln\dfrac{4K_0\,(\beta_{\max}-1)+2D}{4K_0\,(\beta_{\max}-1)-D}}. \quad (29)$$

It is seen from (29) that, as $D \to 4K_0\,(\beta_{\max}-1)$, $\theta^{\max}$ goes to zero, which means that there is a limit point in the case of the zero temperature:

$$D^{\max} = 4K_0\,(\beta_{\max}-1)\,. \quad (30)$$

After the introduction of the dimensionless parameters $\tilde{\theta} = \theta/K_0$ and $\tilde{D} = D/K_0$, we can build the phase diagram in the $\tilde{\theta} - \tilde{D}$ coordinates, which illustrates the case $\beta_{\min} < 1 < \beta_{\max}$ (Fig. 1).

In the diagram, QP_2 is formed in domain *2*. To specify the kind of spin ordering in domain *1*, we consider the case of low temperatures in more details.

If

$$\eta > \xi\,J_0/K_0,, \quad (31)$$

formula (30) takes the form

$$D^{\max} = 4K_0\,(\eta-1)\,. \quad (32)$$

On the contrary, if

$$\eta < \xi\,J_0/K_0, \quad (33)$$

we obtain

$$D^{\max} = 4\,(\xi\,J_0 - K_0)\,. \quad (34)$$

Expressions (31)–(34) completely agree with the results of work [25] which describes what occurs in the system with decrease in D in the case where $T = 0$ and $h_Z = 0$. When condition (31) is fulfilled, QP_2 passes into QAP at the point which is given by expression (32). On the contrary, if condition (33) is valid, QP_2 transforms into the QFM_X phase at the point which is given by expression (34). Thus, in domain *1*, either the QAP or QFM_X phase is realized, depending on which of inequalities, (31) or (33), is valid.

It is noteworthy that, although Hamiltonian (2) is invariant under a rotation around the z axis, the QAP and the QFM_X phase do not possess such an invariance. This means that the QAP and the QFM_X phase are the phases with spontaneously broken symmetry [25].

If

$$\beta_{\min} > 1, \quad \text{and} \quad \beta_{\max} > 1, \tag{35}$$

inequality (24) is fulfilled, when both multipliers have the same sign. In the case where both of them are positive, the stability condition for the spectrum modes is condition (27), whereas the temperature of the stability breaking is given by formula (29). When both the multipliers are negative, the stability condition takes the form

$$\langle O_2^0 \rangle \; < \; D/6K_0\,(1-\beta_{\min})\,. \tag{36}$$

Correspondingly, the stability breaking occurs with increase in the temperature at the point

$$\langle O_2^0 \rangle \; = \; D/6K_0\,(1-\beta_{\min})\,. \tag{37}$$

The temperature of the stability breaking is determined by the expression

$$\theta^{\min} = \frac{\beta_{\min}\, D}{(\beta_{\min}-1)\ln\dfrac{4K_0\,(\beta_{\min}-1)+2D}{4K_0\,(\beta_{\min}-1)-D}}. \tag{38}$$

As a result, if the external magnetic field is zero, the modes of the spin excitation spectrum are stable either for $\theta < \theta^{\min}$ or for $\theta > \theta^{\max}$, provided that condition (35) is fulfilled.

If $\eta = \xi\, J_0/K_0$, the equality $\theta^{\min} = \theta^{\max}$ is valid. In this case, the modes of spectrum (21) are stable for all θ values, except for the point $\theta = \theta^{\min} = \theta^{\max}$.

Figure 2 shows the stability diagram in the $\tilde{\theta} - \tilde{D}$ coordinates. Its lines illustrate the boundaries of the stability breaking and the recovery for the modes of the QP_2 spectrum, i.e. the diagram reflects a reentrant behavior.

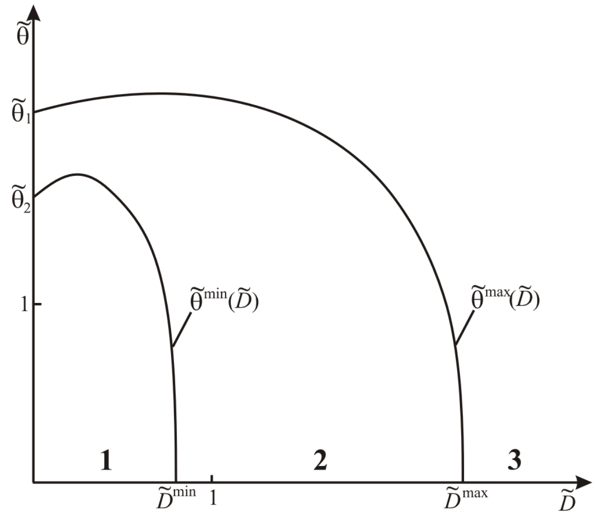

Fig. 2. The stability diagram for an easy-plane magnet with anisotropic BEI for $\beta_{\min} > 1$ and $\beta_{\max} > 1$: *1* – a metastability region for QP_2; *2* – a phase with spontaneously broken symmetry; *3* – QP_2. The boundary between the phases is calculated for $\beta_{\max} = 1.6$ and $\beta_{\min} = 1.2$. In the diagram, $\tilde{D}^{\max} = 2.4$, $\tilde{D}^{\min} = 0.8$, $\tilde{\theta}_1 = 2.13$, and $\tilde{\theta}_2 = 1.6$

A phase with spontaneously broken symmetry, either QAP provided that (31) is valid or the QFM_X phase if the condition (33) is fulfilled, is realized in domain *2*. The modes of the QP_2 spectrum are stable in domains *1* and *3*. However, in domain *1*, QP_2 can be realized as a metastable phase, since the ground state energy is greater for this phase than for the phase with spontaneously broken symmetry [25].

As is seen from the diagram, the situation that the stability firstly breaks at the point $\tilde{\theta} = \tilde{\theta}^{\max}$ and subsequently is restored at the point $\tilde{\theta} = \tilde{\theta}^{\min}$ occurs only under condition that $\tilde{D} < \tilde{D}^{\min}$. If $\tilde{D}^{\min} < \tilde{D} < \tilde{D}^{\max}$, the spectrum modes are stable only for $\tilde{\theta} > \tilde{\theta}^{\max}$. In the case where $\tilde{D} > \tilde{D}^{\max}$, the spectrum modes are stable at arbitrary temperatures.

Thus, the line $\tilde{\theta}^{\max}\left(\tilde{D}\right)$ coincides with the line of the phase transition from QP_2 to a phase with broken symmetry (either QAP or the QFM_X phase). Since the components of the order parameter (the mean values of the spin and tensor operators) are the continuous functions of the field and temperature, these phase transitions are of the second order. It is noteworthy that the line $\tilde{\theta}^{\min}\left(\tilde{D}\right)$ is only the line of the stability recovery for QP_2.

It is seen from (29) that if $\tilde{D} \to 4\,(\beta_{\max}-1)$, then $\tilde{\theta} \to 0$. On the other hand, when $\tilde{D} \to 0$, then $\tilde{\theta} \to$

$(4/3)\,\beta_{\max}$. This allows us to obtain the expressions for $\tilde{D}^{\max}$ and $\tilde{\theta}_1$:

$$\tilde{D}^{\max} = 4\,(\beta_{\max} - 1)\,, \quad \tilde{\theta}_1 = (4/3)\,\beta_{\max}. \tag{39}$$

$\tilde{D}^{\min}$ and $\tilde{\theta}_2$ can be found from Eq. (38):

$$\tilde{D}^{\min} = 4\,(\beta_{\min} - 1)\,, \quad \tilde{\theta}_2 = (4/3)\,\beta_{\min}. \tag{40}$$

5. Discussion of Results

As follows from the results of this paper, in the magnets with anisotropic BEI, the value of the anisotropy constant η affects the stability condition for the modes of the spin excitation spectrum for QP_2. At the same time, if condition (35) is valid, with decrease in the temperature, the stability first breaks at the point $\theta^{\max}$ and then is restored at $\theta^{\min}$. However, if $\tilde{D} < \tilde{D}^{\min}$, QP_2 can exist only as a metastable phase after the stability recovery.

It should be noted that the situation described above is only possible if BEI is anisotropic in the system. Otherwise, if BEI is isotropic, i.e. $\eta = 1$, the modes of spectrum (21) can be unstable only under condition that $\xi\, J_0 > K_0$. When also $\xi = 1$, the modes become unstable only for $J_0 > K_0$. These conclusions are in full compliance with the results of work [22], where only the isotropic exchange interaction and BEI were considered.

Thus, the anisotropic BEI gives rise to the appearance of the reentrant effect in the system (see Fig. 2) only for a certain range of the Hamiltonia parameters [see condition (35)]. If this condition is not fulfilled, the situation shown in Fig. 1 is realized.

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Received 27.08.07.
Translated from Ukrainian by A.I. Tovstolytkin

КВАДРУПОЛЬНА ФАЗА МАГНЕТИКА З АНІЗОТРОПНОЮ БІКВАДРАТНОЮ ОБМІННОЮ ВЗАЄМОДІЄЮ

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Р е з ю м е

Досліджено квадрупольну фазу одновісного магнетика з одноіонною анізотропією (ОА) типу “легка площина” та анізотропною біквадратною обмінною взаємодією (БОВ). Розглянуто випадок, коли значення вузельного спіну дорівнює одиниці. Запропоновано метод розрахунку мод спектра спінових збуджень при скінченних температурах, який у низькотемпературній границі добре узгоджується з відомими результатами для T=0. Одержано вирази для двох віток спектра спінових збуджень при скінченних температурах та визначено умови стійкості мод спектра. Визначена аналітична залежність температури порушення стійкості мод спектра від параметрів гамільтоніана. Доведено, що за певних умов в системі зі зниженням температури спочатку відбувається порушення стійкості мод спектра спінових збуджень, а потім, з подальшим зниженням температури, стійкість мод спектра відновлюється. Передбачено існування метастабільної фази.