HYSTERETIC PHENOMENA IN MAGNETS WITH TENSOR INTERACTIONS

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Magnetic properties of a uniaxial singlet magnet with tensor interactions have been studied. A case where the atomic spins equal 1, and the external magnetic field is directed along the axis of crystal symmetry, is considered. The diagram of stability and metastability in the temperature versus field coordinates is plotted for the quadrupole (QP) and ferromagnetic (FM) phases. It revealed two metastability regions. In one of them, the QP phase is stable, but the FM one can be metastable only. In the other region, the opposite situation is realized. The "hysteretic" dependence of the magnetization on the field is obtained. An analytic dependence of the magnetization reversal energy on the Hamiltonian parameters of the system at zero temperature is derived.

1. Introduction

If atomic spins $S = 1$, there exist two types of tensor interactions: single-ion anisotropy (SIA) and biquadratic exchange interaction (BQEI). In magnetic compounds, for which the SIA and BQEI constants are of the same order as constants of the exchange interaction (EI) bilinear in spin operators, a QP phase can arise [1–4]. At zero temperature, the QP phase is stable in the external magnetic field range $h < h_{c1}$, and the FM phase in the range $h > h_{c2}$, where h_{c1} and h_{c2} are the critical fields which depend on the Hamiltonian parameters. If $h_{c1} < h_{c2}$, both phases are unstable in the interval $h_{c1} < h < h_{c2}$ of the external magnetic field. In this case, a phase with a spontaneously broken symmetry—the so-called canted phase—is realized [5–8]. If $h_{c1} > h_{c2}$, the canted phase does not take place at any temperature. In this case, the temperature versus field phase diagram includes regions, where metastable states can be realized, which results in the emergence of hysteresis phenomena in the system.

In the 1980s, magnetic compounds with large constants of tensor interactions were discovered experimentally (see, e.g., work [9]). This gave an additional impetus to theoretical researches which have been continued till now [10–26]. However, in the majority of works, where the BQEI was taken into account, the authors confined the consideration to the approximation of isotropic BQEI. At the same time, in those works, where the BQEI anisotropy was considered, the influence of BQEI anisotropy constants on magnetic properties of the system was not studied enough.

This work aimed at studying the influence of SIA and anisotropic BQEI on the field dynamics of the order parameter (OP) in the QP and FM phases. Therefore, we confined the consideration to such values of Hamiltonian parameters, at which the condition $h_{c1} > h_{c2}$ is satisfied, so that the canted phase is not realized.

2. Model of the System

Provided the atomic spins $S = 1$, all interactions in a magnetic system can be described with the help of operators of the $ASU(3)$ algebra. Three operators of spin projection S^α $(\alpha = x, y, z)$ and five tensor operators of the second rank O_2^m $(m = 0, \pm1, \pm2)$ can be chosen as the generators of this algebra. The relations between O_2^m operators and S^α ones $(\alpha = x, y, z)$ are as follows:

$$O_2^0 = \left(S^Z\right)^2 - \frac{2}{3}I, \quad O_2^{\pm 1} = -\left(S^Z S^\pm + S^\pm S^Z\right),$$

$$O_2^{\pm 2} = \left(S^\pm\right)^2, \tag{1}$$

where I is the identity operator, whereas the operators S^+ and S^- are defined by the formulas

$$S^+ = \frac{-1}{\sqrt{2}}\left(S^X + iS^Y\right), \quad S^- = \frac{1}{\sqrt{2}}\left(S^X - iS^Y\right). \tag{2}$$

In the case of the single-sublattice ordering, the average values $\langle S^\alpha\rangle$ and $\langle O_2^m\rangle$ completely determine the magnetic and quadrupole orders in the system; therefore, they can be regarded as the OP components. Without loss of generality, the coordinate system can be chosen so that the condition

$$\langle S^y\rangle = \langle O_2^1 + O_2^{-1}\rangle = \langle O_2^2 - O_2^{-2}\rangle = 0 \tag{3}$$

would be satisfied. Therefore, the maximal number of OP components equals five [12].

In the presence of SIA and BQEI, the most general form of a uniaxial Hamiltonian with $S = 1$ looks like

$$
\begin{aligned}
&H = -g\mu_B h \sum_i S_i^Z - \sum_{i,j(i\neq j)} J_{ij}\Big[S_i^Z \cdot S_j^Z + \\
&+\xi\left(S_i^X \cdot S_j^X + S_i^Y \cdot S_j^Y\right)\Big] + D\sum_i O^0_{2i} - \sum_{i,j(i\neq j)} K_{ij}\times \\
&\times\Big\{3O^0_{2i}O^O_{2j} - \frac{1}{2}\eta\Big[\left(O^1_{2i}+O^{-1}_{2i}\right)\left(O^1_{2j}+O^{-1}_{2j}\right) - \\
&-\left(O^1_{2i}-O^{-1}_{2i}\right)\left(O^1_{2j}-O^{-1}_{2j}\right)\Big] + \varsigma\Big[\left(O^2_{2i}+O^{-2}_{2i}\right)\times \\
&\times\left(O^2_{2j}+O^{-2}_{2j}\right) - \left(O^2_{2i}-O^{-2}_{2i}\right)\left(O^2_{2j}-O^{-2}_{2j}\right)\Big]\Big\}, \qquad (4)
\end{aligned}
$$

where J_{ij} and K_{ij} are the EI and BQEI constants, respectively; D is the SIA constant; ξ the constant of EI anisotropy; and η and ζ are the constants of BQEI anisotropy which are adopted to be positive.

The first term in Hamiltonian (4) is the Zeeman energy. The second one is the EI energy which becomes isotropic at $\xi = 1$. The third term is the SIA energy. The fourth term is the BQEI energy. In the case where $\eta = \zeta = 1$, BQEI is isotropic:

$$H_{\mathrm{BQEI}} = -\sum_{i,j(i\neq j)} K'_{ij}\left(\mathbf{S}_i \cdot \mathbf{S}_j\right)^2, \qquad (5)$$

where K'_{ij} are the renormalized BQEI constants.

In this work, we confined the consideration to single-sublattice structures in easy-plane magnets, which is provided by the condition ($J_{ij} > 0$, $K_{ij} > 0$, and $D > 0$).

In the symmetric phases, only two OPs are different from zero, $\langle S^Z\rangle$ and $\langle O^0_2\rangle$. Therefore, in the molecular field approximation, the Hamiltonian looks like

$$
\begin{aligned}
&H_0 = -\left(g\mu_B h_Z + J_0\langle S^Z\rangle\right)\sum_i S_i^Z + \\
&+\left(D - 3\,K_0\langle O^0_2\rangle\right)\sum_i O^0_{2i}, \qquad (6)
\end{aligned}
$$

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

From Eq. (6), we obtain the expression for the average energy value:

$$E = N\Big(-g\mu_B h\langle S^Z\rangle - J_0\langle S^Z\rangle^2 + D\langle O^0_2\rangle - 3K_0\langle O^0_2\rangle^2\Big). \qquad (7)$$

Depending on the spin projection onto the axis z ($S^Z = 0, \pm 1$), the energy levels of an individual atom are determined by one of the formulas

$$E_1 = -\tilde{h} + \frac{1}{3}\tilde{d}, \quad E_0 = -\frac{2}{3}\tilde{d}, \quad E_{-1} = \tilde{h} + \frac{1}{3}\tilde{d}, \qquad (8)$$

where

$$\tilde{h} = g\mu_B h_Z + J_0\langle S^Z\rangle, \qquad \tilde{d} = D - 3\,K_0\langle O^0_2\rangle. \qquad (9)$$

At zero temperature, the OP components $\langle S^Z\rangle$ and $\langle O^0_2\rangle$ equal 1 and 1/3, respectively, in the FM phase, and 0 and $-2/3$, respectively, in the QP one. At finite temperatures, the $\langle S^Z\rangle$- and $\langle O^0_2\rangle$-values are determined from the system of two equations,

$$\langle S^Z\rangle = \frac{\sum_n S_n^Z \exp\left(-E_n/\theta\right)}{\sum_n \exp\left(-E_n/\theta\right)}, \quad \langle O^0_2\rangle = \frac{\sum_n O^0_2 \exp\left(-E_n/\theta\right)}{\sum_n \exp\left(-E_n/\theta\right)}, \qquad (10)$$

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

Taking Eq. (8) into account, the system of equations (10) can be presented as

$$\langle S^Z\rangle = \frac{e^- - e^+}{1 + e^- + e^+}, \quad \langle O^0_2\rangle = 1/3 - \frac{1}{1 + e^- + e^+}\ , \qquad (11)$$

where the quantities e^- and e^+ are defined by the expressions

$$e^- = \exp\frac{-\tilde{h} + \tilde{d}}{\theta}, \quad e^+ = \exp\frac{\tilde{h} + \tilde{d}}{\theta}. \qquad (12)$$

3. Entropy and Free Energy of the System

In a system composed of N atoms, let N_1 atoms be in a state with $S^Z = 1$ and N_2 atoms in a state with $S^Z = -1$. Then, the OP components different from zero are

$$\langle S^Z\rangle = \frac{N_1}{N} - \frac{N_2}{N}, \quad \langle O^0_2\rangle = \frac{N_1}{N} + \frac{N_2}{N} - \frac{2}{3}. \qquad (13)$$

The entropy of the system can be written down as follows:

$$\sigma = k\ln\frac{N!}{N_1!N_2!\left(N - N_1 - N_2\right)!}. \qquad (14)$$

Using the Stirling formula, $\ln N! \approx N(\ln N - 1)$, expression (14) can be presented in the form

$$\sigma = -Nk\Bigg[\ln\left(1 - \frac{N_1}{N} - \frac{N_2}{N}\right) -$$

$$-\frac{N_1}{N}\ln\left(\frac{N}{N_1} - \frac{N_2}{N_1} - 1\right) - \frac{N_2}{N}\ln\left(\frac{N}{N_2} - \frac{N_1}{N_2} - 1\right)\Bigg]. \tag{15}$$

The free energy of the system is defined by the formula

$$F = E - T\,\sigma. \tag{16}$$

Taking Eqs. (7), (13), and (15) into account, the expression for the free energy per one atom reads

$$F = -g\mu_B h\langle S^Z\rangle - J_0\langle S^Z\rangle^2 + D\langle O_2^0\rangle - 3K_0\langle O_2^0\rangle^2 +$$

$$+\theta\Bigg\{\ln\left(\frac{1}{3} - \langle O_2^0\rangle\right) + \frac{\langle S^Z\rangle}{2}\ln\frac{2 + 3\langle O_2^0\rangle + 3\langle S^Z\rangle}{2 + 3\langle O_2^0\rangle - 3\langle S^Z\rangle} -$$

$$-\frac{2 + 3\langle O_2^0\rangle}{6}\ln\frac{4\left(1 - 3\langle O_2^0\rangle\right)^2}{\left(2 + 3\langle O_2^0\rangle\right)^2 - 9\langle S^Z\rangle^2}\Bigg\}, \tag{17}$$

where the quantities $\left\langle S^Z\right\rangle$ and $\left\langle O_2^0\right\rangle$ are determined from system (11).

4. Stability Boundaries for QP and FM Phases

In works [27,28], expressions were obtained in the framework of model (4) for stability boundaries of the QP and FM phases with respect to their mutual transitions. Both expressions have the same form:

$$g\mu_B h + \langle S^Z\rangle\left(2J_0 - \xi J_0 - \eta K_0\right) =$$

$$= \Bigg\{\left(\langle S^Z\rangle\right)^2\left(\xi J_0 - \eta K_0\right)^2 + \Bigg[D - 6\langle O_2^0\rangle\times$$

$$\times\left(K_0 - \xi J_0\right)\Bigg]\left[D - 6\langle O_2^0\rangle K_0\left(1 - \eta\right)\right]\Bigg\}^{1/2}, \tag{18}$$

but the stability boundaries of both phases in the temperature–field coordinates do not coincide. Such a situation is associated with the fact that formula (18) contains the OP components $\left\langle S^Z\right\rangle$ and $\left\langle O_2^0\right\rangle$ which acquire different values in different phases at the same θ and h.

The critical fields h_{c1} and h_{c2} are the low-temperature limits of the stability regions for the QP and FM phases, respectively. Their values can be easy obtained from expression (18), putting $T = 0$:
for the QP phase,

$$g\mu_B h_{c1} = \sqrt{\left[D + 4K_0(1 - \eta)\right]\left[D + 4(K_0 - \xi J_0)\right]}, \tag{19}$$

for the FM phase,

$$g\mu_B h_{c2} = -2J_0 + \xi J_0 + \eta K_0 + \left|D - 2K_0 + \xi J_0 + \eta K_0\right|. \tag{20}$$

In the case where the condition

$$D - 2K_0 + \xi J_0 + \eta K_0 > 0, \tag{21}$$

is satisfied, formula ([20]) reads

$$g\mu_B h_{c2} = D - 2J_0(1 - \xi) - 2K_0(1 - \eta)\ . \tag{22}$$

Expressions ([19]) and (22) coincide with those for critical fields which were obtained in an alternative way in work [6].

5. Diagram of Stability and Metastability for the QP and FM Phases

For further researches, we select the values for Hamiltonian parameters so that the condition $h_{c1} > h_{c2}$ is satisfied. In Fig. 1, a diagram of stability and metastability for the QP and FM phases is exhibited in the dimensionless coordinates $\tilde{\theta}$ versus $\tilde{h}$ ($\tilde{\theta} = \theta/J_0, \tilde{h} = g\mu_B h/J_0$). Curves *1* and *2* are the stability boundaries for the FM and QP phases, respectively, which are described by expression (18). Therefore, in region *a* (*a1, a2*), both phases are stable simultaneously. Curve *3* is defined by the condition of equal free energies in both phases,

$$F\left(\langle S^Z\rangle_{\rm FM}, \langle O_2^0\rangle_{\rm FM}\right) = F\left(\langle S^Z\rangle_{\rm QP}, \langle O_2^0\rangle_{\rm QP}\right). \tag{23}$$

In region *a1*, the QP phase is stable, but the FM one can exist only as metastable. In region *a2*, the FM phase is stable, whereas the QP one is metastable. In region *b*, the FM phase is unstable, so that only the QP phase is realized, whereas the opposite situation is observed in region *c*. The difference between the values of OP components for the FM and QP phases along curve *3* diminishes as the temperature grows, by vanishing at point *A* characterized by the coordinates $(\tilde{\theta}^*, \tilde{h}^*)$, i.e., point *A* is critical. The critical point coordinate $\tilde{\theta}^*$ depends on the dimensionless parameters $\tilde{D} = D/J_0$ and

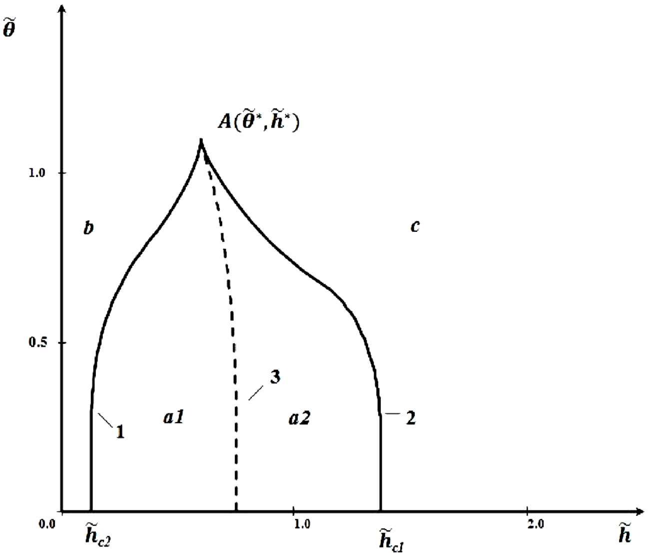

Fig. 1. Diagram of stability and metastability for the QP and FM phases at $\tilde{D} = 0.5$, $\tilde{K}_0 = 1.25$, $\eta = 0.85$, and $\xi = 1$

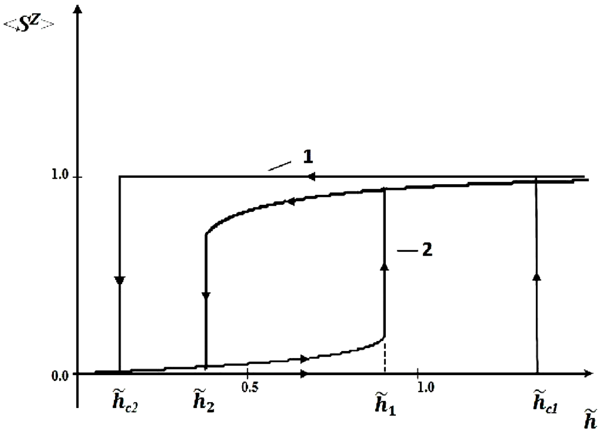

Fig. 2. Dependences of the relative magnetization on $\tilde{h}$ at $\tilde{D} = 0.5$ and $\tilde{K}_0 = 1.25$ for $\tilde{\theta} = 0$ (*1*) and 0.8 (*2*)

$\tilde{K}_0 = K_0/J_0$ of the Hamiltonian. In this case, if $\tilde{D}$ grows from 0.5 to 0.75 and $\tilde{K}_0 = 1.25$, the quantity $\tilde{\theta}^*$ changes almost linearly from 1.11 to 1.16. On the other hand, if $\tilde{K}_0$ grows from 1.25 to 1.7 and $\tilde{D} = 0.5$, the quantity $\tilde{\theta}^*$ also changes almost linearly from 1.11 to 1.41.

At $\tilde{\theta} > \tilde{\theta}^*$, there exists a region, in which the paramagnetic phase is realized. In what follows, we examine the processes that occur only under condition $\tilde{\theta} < \tilde{\theta}^*$. Therefore, we do not dwell on the issue of the boundaries between the paramagnetic phase, on the one hand, and the QP and FM phases, on the other hand, which has been studied well enough in the literature (see works [9, 19, 22, 29] and others).

6. Hysteresis and Magnetic Reversal Energy

Let us consider a case of low temperatures. If the fields in the system are low, the stable QP phase is realized. As the external magnetic field grows and the system crosses a curve, where the free energies in both phases are equal to each other (curve *3* in Fig. 1), the QP phase changes from the stable state into a metastable one, with the quantity $\langle S^Z \rangle$, which has the meaning of relative magnetization, remaining continuous. As the quantity h grows further, a phase transition of the first kind into the FM state takes place at the stability boundary of the QP phase (curve *2* in Fig. 1), which is accompanied by a jump-like increase of the $\langle S^Z \rangle$-value. If the external magnetic field h changes in the opposite direction, the FM phase, when crossing the curve of equal free energies, continuously changes into a metastable state, whereas the phase transition of the first kind, which is accompanied by a jump-like reduction of $\langle S^Z \rangle$-value, occurs at the stability boundary of the FM phase (curve *1* in Fig. 1). So, the dependence of the relative magnetization on h looks like a hysteresis loop (Fig. 2). The temperature growth is accompanied by a reduction of the loop area.

If the spin value $S = 1$, the magnetic field induction B is determined by the expression

$$B = h + 4\pi\, g\mu_B \langle S^Z \rangle\, n\,, \tag{24}$$

where n is the concentration of magnetic atoms. It follows from formula (24) that, along with the dependence $\langle S^Z \rangle(h)$, the dependence $B(h)$ is also hysteretic. In the $B - h$ coordinates, the loop area equals the energy of magnetic reversal in a unit volume of the system,

$$W_V = \int_{h2}^{h_1} (B_{\rm FM} - B_{\rm QP})dh, \tag{25}$$

where the fields h_1 and h_2 are determined as the solutions of the system of equations (18) and (11) at a fixed temperature; they are the limiting stability points for the FM and QP phases, respectively.

With regard for Eq. (24), the quantity W_V can be presented in the form

$$W_V = 4\pi g\mu_B n \int_{h_2}^{h_1} \big(\langle S^Z \rangle_{\rm FM} - \langle S^Z \rangle_{\rm QP}\big)dh. \tag{26}$$

Then, the dimensionless energy of magnetic reversal per one atom is determined by the formula

$$\tilde{W}_1 = 4\pi \int\limits_{\tilde{h}_2}^{\tilde{h}_1} \left(\langle S^Z \rangle_{\rm FM} - \langle S^Z \rangle_{\rm QP} \right) d\tilde{h}. \tag{27}$$

At $T = 0$, the equalities $h_1 = h_{c1}$, $h_2 = h_{c2}$, $\langle S^Z \rangle_{\rm FM} = 1$, and $\langle S^Z \rangle_{\rm QP} = 0$ are valid, and, accordingly, we obtain the following expression for $\tilde{W}_1$:

$$\tilde{W}_1 = 4\pi \left(\tilde{h}_{c1} - \tilde{h}_{c2} \right). \tag{28}$$

Taking Eqs. (19) and (22) into account, we obtain an analytic dependence of $\tilde{W}_1$ on the Hamiltonian parameters:

$$\tilde{W}_1 = 4\pi \Biggl(\sqrt{\left[\tilde{D} + 4\tilde{K}_0(1-\eta) \right] \left[\tilde{D} + 4(\tilde{K}_0 - \xi) \right]} - $$

$$ - \tilde{D} + 2(1-\xi) + 2\tilde{K}_0(1-\eta) \Biggr). \tag{29}$$

Hence, only the constant η of two constants of BQEI anisotropy (η and ζ) affects $\tilde{W}_1$. For example, at $\tilde{D} = 0.5$, $\tilde{K}_0 = 1.25$, $\xi = 1$, and an arbitrary value of ζ, the variation of the constant η from 0 to 0.8 results in the almost linear decrease of $\tilde{W}_1$ from 61.2 to 18.8.

7. Discussion of Results

Hence, the metastable quadrupole and ferromagnetic states can be realized in singlet magnets in a certain ranges of Hamiltonian parameters and at low enough temperatures. On the $T - h$ diagram, the boundary between regions, in which those states are realized, is a curve, where the free energies in the QP and FM phases are equal. The presence of metastable regions results in that the field dependence of the relative magnetization has a hysteretic form. The dimensionless energy of magnetic reversal per one atom, $\tilde{W}_1$, decreases, as the temperature increases. The magnitude of $\tilde{W}_1$ depends substantially on the BQEI anisotropy constant η.

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ГІСТЕРЕЗИСНІ ЯВИЩА В МАГНЕТИКАХ З ТЕНЗОРНИМИ ВЗАЄМОДІЯМИ

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Резюме

Досліджено магнітні властивості одноосьового синглетного магнетика з тензорними взаємодіями. Розглянуто випадок, коли значення атомного спіну дорівнює одиниці $S = 1$, а зовнішнє магнітне поле спрямоване уздовж осі симетрії кристала. У координатах температура – поле побудовано діаграму стабільності й метастабільності квадрупольної (КФ) і феромагнітної (ФМФ) фаз, на якій є дві області метастабільності. В одній із них КФ стабільна, а ФМФ – метастабільна, в іншій – реалізується протилежна ситуація. Побудовано польову залежність намагніченості, яка має "гістерезисний" вигляд. При нульовій температурі визначено аналітичну залежність енергії перемагнічування від параметрів гамільтоніана системи.