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Orientational phase transitions in anisotropic rare-earth magnets at low temperatures within the Heisenberg model

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Abstract. Orientational phase transitions are investigated within the Heisenberg model with single-site anisotropy. The temperature dependence of the cone angle is calculated analytically within the spin-wave theory. The role of the quantum renormalizations of anisotropy constants is discussed. A comparison with the experimental data on the cone–plane orientational transition in holmium is performed.

1. Introduction

The old problem of magnetic structure of rare-earth metals and their compounds is still a subject of experimental and theoretical investigation. These substances have complicated phase diagrams and demonstrate a number of orientational phase transitions. In particular, such transitions take place in the orthoferrites and the practically important intermetallic compounds RCo_5 (R = Pr, Nd, Tb, Dy, Ho); see, e.g., reference [1]. A qualitative explanation of these transitions was obtained many years ago within the Heisenberg model with inclusion of magnetic anisotropy [2]. In a number of systems, lattice (magnetoelastic) effects are important. The standard description is usually performed within mean-field approaches (see [2, 3] and references therein). However, quantitative comparison with experimental data requires a more detailed treatment.

Provided that the orientational transition temperature is low (in comparison with the magnetic ordering point), spin-wave theory is applicable [2]. In the simplest case of the second-order anisotropy the magnetization lies either along the easy axis or in the easy plane. Inclusion of higher-order anisotropy constants can lead to cone phases where the magnetization makes the angle θ with the z-axis. The case $\theta = \pi/2$ was considered in references [3–6] where the temperature renormalization of the anisotropy constants and the spin-wave spectrum in Tb and Dy within the standard spin-wave theory were calculated.

In the present paper we consider the cone phase with arbitrary $0 \leqslant \theta \leqslant \pi/2$. The situation here is analogous to that for the field-induced orientational phase transitions, e.g., in the transverse-field Ising model (see reference [7] and references therein). Unlike for the latter model, one can expect at low enough temperatures and small values of anisotropy the spin-wave theory to be applicable for an arbitrary relation between anisotropy parameters, not only close to the orientational phase transition. Even for the second-order easy-plane anisotropy, the Holstein–Primakoff representation for spin operators used in references [4–6] leads to so-called kinematical inconsistencies because of incorrect treatment of on-site kinematical relations. To

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avoid this difficulty, we use the technique of spin coherent states. Our approach is to some extent similar to the operator approach used in reference [3], but gives the possibility of treating more simply higher-order anisotropy constants, as well as of calculating higher-order terms of the $1/S$ expansion.

The anisotropic Heisenberg model used is formulated in section 2. In section 3 we develop a special form of the $1/S$ expansion which gives the possibility of taking into account exactly on-site kinematical relations. In section 4 we treat the cone–plane transition owing to the temperature dependence of the cone angle θ and discuss experimental data on the rare-earth metals.

2. The model and mean-field approximation

We start from the Hamiltonian of the Heisenberg model with inclusion of single-site anisotropy

$$\mathcal{H} = -\frac{J}{2}\sum_{\langle ij\rangle} \boldsymbol{S}_i \boldsymbol{S}_j + B_2^0 \sum_i (O_2^0)_i + B_4^0 \sum_i (O_4^0)_i \tag{1}$$

where $J > 0$ is the exchange parameter,

$$\begin{aligned} O_2^0 &= 3(S^z)^2 - S(S+1) \\ O_4^0 &= 35(S^z)^4 - 30S(S+1)(S^z)^2 + 25(S^z)^2 + 3S^2(S+1)^2 - 6S(S+1) \end{aligned} \tag{2}$$

are the irreducible tensor operators of second and fourth orders, B_l^m are the corresponding anisotropy constants.

Up to an unimportant constant we can rewrite the Hamiltonian (1) in the form

$$\mathcal{H} = -\frac{J}{2}\sum_{\langle ij\rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j + D\sum_i (S_i^z)^2 + D'\sum_i (S_i^z)^4 \tag{3}$$

where

$$\begin{aligned} D &= 3B_2^0 - [30S(S+1) - 25]B_4^0 \\ D' &= 35B_4^0. \end{aligned} \tag{4}$$

For $D, D' > 0$, spins of magnetic ions lie in the easy plane xy, while for $D, D' < 0$, we have the easy axis z. For $D > 0$, $D' < 0$, a first-order transition takes place between the easy plane (which is favoured by second-order anisotropy) and easy axis (which is favoured by large $|D'|$). We consider only the case $D < 0$, $D' > 0$ where the cone phase occurs at intermediate values of $D/(2D'S^2)$, so that the spin orientation direction makes the angle θ with the z-axis and the orientational phase transitions are of second order. This is the case for Gd and also for Ho, Er in low-temperature phases.

In the phenomenological approach it is supposed (see, e.g., references [1, 2]) that

$$F = F_{\mathrm{is}} + D(T)(S\cos\theta)^2 + D'(T)(S\cos\theta)^4 \tag{5}$$

where F_{is} is the isotropic (θ-independent) part of the free energy. Then we obtain by minimization of F

$$\cos^2\theta(T) = -\frac{D(T)}{2D'(T)S^2} \tag{6}$$

so at the point where $D(T) = 0$ the spins become directed in the xy-plane while when $|D(T)| \geqslant 2D'(T)S^2$ they are aligned along the z-axis. The temperature dependence of $D(T)$ is supposed to have the form

$$D(T) = 2D'S^2(T_1 - T)/(T_2 - T_1) \tag{7}$$

with $D'(T) > 0$. Thus at $T = T_1$ the transition from the easy-plane to the cone structure takes place, while at $T = T_2$ the transition from the cone to the easy-axis structure occurs. At the same time, Zener's [8] result for the temperature dependence of the anisotropy constants in an axially symmetric state with $\theta = 0$ has the form

$$B_l^0(T) = B_l^0 M^{l(l+1)/2} \tag{8}$$

where $M = \langle \widetilde{S}^z \rangle / S$ is the relative magnetization, and $D(T)$, $D'(T)$ are determined by the same relations (4) with $B_l^0 \to B_l^0(T)$. As pointed out in references [3–6], the temperature dependences of the anisotropy constants have a more complicated form for the cone structures with $\theta > 0$ (in fact, only the case $\theta = \pi/2$ is discussed in references [3–6]).

A systematic way of calculating temperature dependences of anisotropy constants is using the $1/S$ expansion which is considered in the next section.

3. The $1/S$ expansion of the partition function

The $1/S$ expansion developed here is slightly different from the standard scheme of $1/S$ expansion [4–6] since it gives the possibility of taking into account exactly the kinematical relations between powers of spin operators on each site. We use the coherent state approach (see, e.g., reference [9]) to write down the partition function in the form

$$\mathcal{Z} = \int \mathrm{D}\boldsymbol{\pi}\ \exp\left\{ \mathrm{i}S \int_0^\beta \mathrm{d}\tau\ (1 - \cos\vartheta) \frac{\partial \varphi}{\partial \tau} - \langle \pi | \mathcal{H} | \pi \rangle \right\} \tag{9}$$

where $\boldsymbol{\pi}$ is the unit-length vector, ϑ and φ are its polar and azimuthal angles respectively, $|\pi\rangle = \exp(\mathrm{i}\vartheta S^y + \mathrm{i}\varphi S^z)|S\rangle$ are the coherent states ($S^z|S\rangle = S|S\rangle$). To construct the $1/S$ expansion we rotate the coordinate system around the y-axis through the angle θ. The Hamiltonian (1) takes the form

$$\mathcal{H} = -\frac{J}{2} \sum_{\langle ij \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j + \sum_i \sum_{l,m} \sum_{m'=-l}^{l} B_l^m \sqrt{\frac{(l+m)!(l-|m'|)!}{(l-m)!(l+|m'|)!}} \frac{A_l^m}{A_l^{|m'|}} d_{mm'}^l(\theta) (\widetilde{O}_l^{|m'|})_i \tag{10}$$

where $d_{mm'}^l(\theta)$ are the Wigner matrices of the rotation group irreducible representation,

$$A_l^m = \frac{(l-m)!}{(l+[m]-1)!!} \frac{1}{K_l^m} \tag{11}$$

($[m] = m$ for m even and $[m] = m+1$ for m odd), for $l \leqslant 4$ we have $K_l^m = 1$, and the tilde sign here and hereafter refers to the rotated coordinate system. Since the partition function (9) is invariant under rotation of the states $|\pi\rangle$, it is convenient to use the coherent states defined in the same coordinate system, i.e., $|\widetilde{\pi}\rangle = \exp(\mathrm{i}\vartheta \widetilde{S}^y + \mathrm{i}\varphi \widetilde{S}^z)|\widetilde{S}\rangle$ with $\widetilde{S}^z|\widetilde{S}\rangle = S|\widetilde{S}\rangle$. The advantage of using the coherent states is the simple form of the averages of the tensor operators (2) over $|\widetilde{\pi}\rangle$. By direct calculation we obtain

$$\langle \widetilde{\pi} | \widetilde{O}_l^m | \widetilde{\pi} \rangle = S_l A_l^m P_l^m(\cos\vartheta) \cos m\varphi \tag{12}$$

where $P_l^m(x)$ are the associated Legendre polynomials, and the factors $S_l = S(S - 1/2) \cdots [S - (l-1)/2]$ take into account properly the kinematical relations on each site. In particular, the second-order anisotropy term vanishes for $S = 1/2$, and the fourth-order term for $S = 1/2, 1, 3/2$, as it should (unlike the results for boson representations in references [4–6]). Using (12) we obtain for the case $B_l^m = B_l^0 \delta_{m0}$ under consideration the result

$$\langle \widetilde{\pi} | \mathcal{H} | \widetilde{\pi} \rangle = -\frac{JS^2}{2} \sum_{\langle ij \rangle} \widetilde{\boldsymbol{\pi}}_i \cdot \widetilde{\boldsymbol{\pi}}_j + \sum_i \sum_{l=2,4} \sum_{m=-l}^{l} S_l B_l^0 A_l^0 \frac{(l-|m|)!}{(l+|m|)!} P_l^{|m|}(\cos\theta) P_l^{|m|}(\cos\vartheta_i) \cos m\varphi_i . \tag{13}$$

Further calculations are performed along the same lines as in reference [7]. Using the representation

$$\cos\vartheta_i = \sqrt{1-\sin^2\vartheta_i}$$

and expanding in $\sin\vartheta_i$ we obtain the $1/S$ expansion of the partition function. It should be stressed that we retain the factors S_l, as well as the S-dependences in (4), non-expanded. By performing decouplings, terms of third order are reduced to linear ones, and terms of fourth order to quadratic ones. The requirement of the absence of $\sin\vartheta$-linear terms leads to the result for the cone angle θ

$$\cos^2\theta = \frac{3}{7}\left[1 - X + Y - \frac{1}{10}\frac{B_2^0 S_2}{B_4^0 S_4}\left(1 - \frac{7}{2S} + 6X + Y\right)\right] \tag{14}$$

where

$$\begin{aligned} X &= \langle \pi_{xi}^2\rangle \equiv \langle \sin^2\vartheta\cos^2\varphi\rangle = \sum_q \frac{J_0 - J_q}{2E_q}\coth\frac{E_q}{2T} \\ Y &= \langle \pi_{yi}^2\rangle \equiv \langle \sin^2\vartheta\sin^2\varphi\rangle = \sum_q \frac{J_0 - J_q + \Delta_0/S}{2E_q}\coth\frac{E_q}{2T} \end{aligned} \tag{15}$$

and the ‘bare’ magnon spectrum reads

$$\begin{aligned} E_q &= S\sqrt{(J_0 - J_q)(J_0 - J_q + \Delta_0/S)} \\ \Delta_0 &= 2\left[3B_2^0 S_2\cos 2\theta - 10B_4^0 S_4(28\cos^4\theta - 27\cos^2\theta + 3)\right] \end{aligned} \tag{16}$$

where Δ_0 is the energy gap. To obtain the correct description of the thermodynamics at not too low temperatures the corrections in (14) can be collected into powers in the same way as in references [10, 11],

$$1 - \alpha X \to 1/X^\alpha \qquad 1 - \alpha Y \to 1/Y^\alpha. \tag{17}$$

(In the presence of higher-order anisotropy this is essential since the coefficients at X, Y increase $\sim l^2/2$ with the anisotropy order). Then we have

$$\cos^2\theta = \frac{3}{7}\frac{Z_X}{Z_Y}\left[1 - \frac{1}{10}\frac{B_2^0(T)S_2}{B_4^0(T)S_4}\right] \tag{18}$$

where

$$B_2^0(T) = Z_X^2 Z_Y B_2^0 \qquad B_4^0(T) = Z_X^9 Z_Y B_4^0 \tag{19}$$

are the temperature-renormalized anisotropy constants, and

$$Z_X = 1 + \frac{1}{2S} - X \qquad Z_Y = 1 + \frac{1}{2S} - Y. \tag{20}$$

The relations (19) extend the results of references [3–6] to the case where spins make a non-zero angle with the z-axis. The renormalized gap has the form

$$\begin{aligned} \Delta = {} & 6\cos 2\theta B_2^0 S_2 - 20B_4^0 S_4[3(1 - 7\widetilde{X}) - 3\cos^2\theta(9 - 56\widetilde{X} - 7\widetilde{Y}) \\ & + 28\cos^4\theta(1 - 6\widetilde{X} - \widetilde{Y})] - 196\sin^2\theta\cos^2\theta \\ & \times \sum_{k,\omega_n}\left[\frac{3B_2^0 S_2(J_0 - J_k) - 10B_4^0(S_4/S)\Delta_0\cos^2\theta}{\omega_n^2 + S^2(J_0 - J_k)(J_0 - J_k + \Delta_0/S)}\right]^2 \end{aligned} \tag{21}$$

where

$$\widetilde{X} = X - 1/(2S) \qquad \widetilde{Y} = Y - 1/(2S) \qquad \omega_n = 2\pi nT.$$

After introducing the temperature-renormalized second- and fourth-order anisotropy parameters $D(T)$ and $D'(T)$,

$$\begin{aligned} D(T)S^2 &= 3B_2^0(T)S_2 - 30B_4^0(T)S_4 \\ D'(T)S^4 &= 35B_4^0(T)S_4(Z_Y/Z_X) \end{aligned} \tag{22}$$

the expression for $\cos\theta$ coincides with that of the phenomenological theory (6). Collecting again corrections in (21) into powers, we obtain for the renormalized gap in the notation (22) the expression

$$\begin{aligned} \Delta = {} & 2D(T)S^2\cos 2\theta + 2D'(T)S^4\cos^4\theta + 6D'(T)S^4\sin^2\theta\cos^2\theta \\ & - 196\sin^2\theta\cos^2\theta \sum_{k,\omega_n}\left[\frac{3B_2^0S_2(J_0-J_{\boldsymbol{k}}) - 10B_4^0(S_4/S)\Delta_0\cos^2\theta}{\omega_n^2 + S^2(J_0-J_{\boldsymbol{k}})(J_0-J_{\boldsymbol{k}}+\Delta_0/S)}\right]^2 \end{aligned} \tag{23}$$

which also coincides with that obtained in the phenomenological theory except for the last term. Note that at $\theta > 0$ the renormalizations (22) are present even at $T = 0$, which should be taken into account when treating experimental data.

4. Orientational phase transitions

Now we can pass to description of possible orientational phase transitions. Consider first the case of a small enough constant B_4^0 (or, equivalently, D'), so that $\cos^2\theta(0)$ is close to unity. Then $\cos^2\theta(T)$ increases with temperature and there occurs a transition to the easy-axis phase at the point determined by

$$\frac{3}{7}\frac{Z_X}{Z_Y}\left[1 - \frac{1}{10}\frac{B_2^0(T)S_2}{B_4^0(T)S_4}\right] = 1. \tag{24}$$

In the opposite case of a large enough B_4^0, $\cos^2\theta(0)$ is small, and $\cos^2\theta(T)$ decreases with temperature, so at the point where

$$B_2^0(T)S_2 = 10B_4^0(T)S_4 \tag{25}$$

a phase transition to the easy-plane phase occurs. Thus one can expect that there exists a critical value θ_c: for $\theta_0 = \theta(0) < \theta_c$ we have a decrease of $\theta(T)$ with T and the phase transition from cone to easy-axis phase, while for $\pi/2 > \theta_0 > \theta_c$ we have an increase of $\theta(T)$ with T and the phase transition from cone to easy-plane phase. The numerical computations for the simple cubic lattice (see figure 1) yield $\theta_c \simeq 50^\circ$. Figure 2 shows the corresponding temperature dependences of the anisotropy constants $D(T)$, $D'(T)$. For simplicity, $J_{\boldsymbol{q}}$ is taken for the simple cubic lattice.

The phase transitions described by equations (24) and (25) are analogous to those in the phenomenological theory of reference [1] that occur at $D(T) = 0$ and $D(T) = -2D'(T)S^2$, respectively. However, unlike the phenomenological approach, microscopical consideration leads to either cone–easy-axis or cone–easy-plane transition with increasing temperature, depending on the zero-temperature value of θ. At the same time, the transition from the easy-plane to the easy-axis structure (through the intermediate cone phase) cannot be explained by purely magnetic renormalizations of anisotropy constants.

The result (14) gives the mean-field values of the critical exponents for both the ground-state and temperature orientational phase transitions (e.g., for the order parameter $\langle S^z\rangle \propto \cos\theta$ we have $\beta = 1/2$). Unlike the systems discussed in reference [7], the system under consideration has the dynamical critical exponent $z = 2$ (i.e., a single excitation mode with nearly quadratic dispersion is present). Thus the upper critical dimensionality for the ground-state quantum phase transition (QPT) is $d_c^+ = 4 - z = 2$. In this respect, the system is

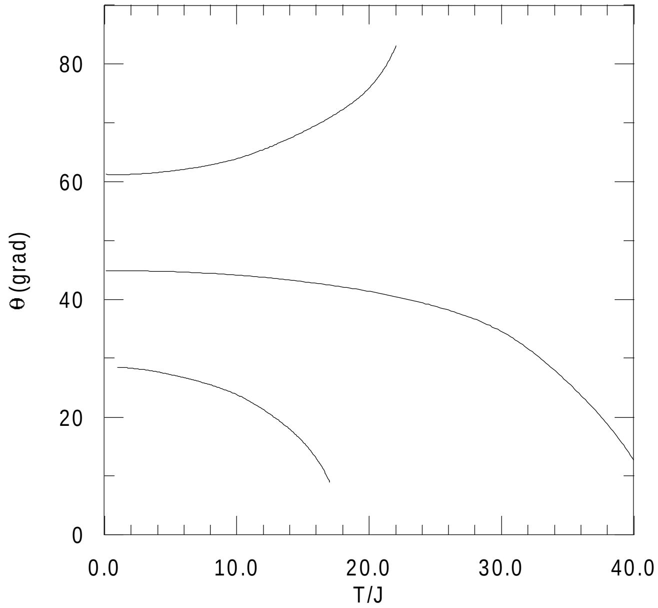

Figure 1. The theoretical temperature dependences of the cone angle $\theta(T)$ for $S = 7/2$ and different values of second-order anisotropy: $D/J = 0.004$; 0.005; 0.006 from upper to lower curve. The value of D'/J is 3.7×10^{-4}.

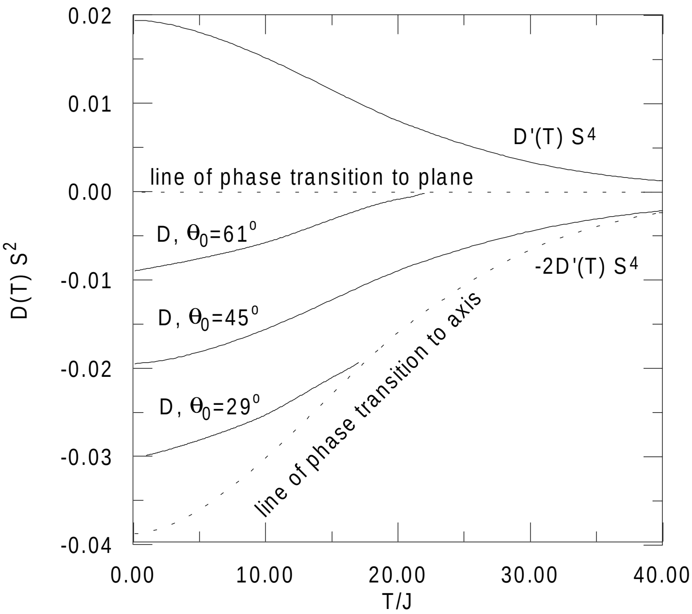

Figure 2. The temperature dependences of the anisotropy constants $D(T)$, $D'(T)$ corresponding to figure 1. Temperature transition points correspond to the intersection of the curve $D(T)$ with one of the dashed lines.

analogous to the XY model in the transverse magnetic field [12]. A characteristic feature of such systems is the mean-field behaviour of critical exponents both above and below the critical dimensionality. For (hypothetical) systems with $d = 2$, logarithmic corrections to ground-state properties near QPT are present (see, e.g., reference [13]). At the same time, the upper critical dimensionality for the temperature phase transition is $d_{cT}^{+} = 4$, and at $d < d_{cT}^{+}$ the temperature transition critical exponents differ from their mean-field values.

Now we discuss the experimental situation. In Gd (see, e.g., references [2, 14]) the orientational phase transition from cone phase to easy-axis phase is observed at $T_c = 240$ K. The temperature dependence of the cone angle at $T < T_c$ (and also of the magnetic anisotropy constants) is non-monotonic, unlike the dependence discussed in section 2. This complicated situation is connected with the absence of orbital momentum and the smallness of the anisotropy in gadolinium.

In holmium the low-temperature phase is a conical spiral one, the angle of the cone changing from $\approx 80^\circ$ to 90° in the temperature interval 0–20 K. The spiral angle makes up about 30°. At the same time, the temperature of the magnetic phase transition with vanishing magnetization is $T_N = 132$ K, so we have $T \ll T_N$ in the temperature interval discussed. Since the sixth-order and dipolar anisotropy are important, we use the Hamiltonian [15]

$$\mathcal{H}_{\mathrm{Ho}} = \mathcal{H} - \frac{1}{2}\sum_{ij}\mathcal{J}_{ij}^{D} S_i^z S_j^z + B_6^0 \sum_i (O_6^0)_i + B_6^6 \sum_i (O_6^6)_i \tag{26}$$

with

$$\mathcal{J}_D(\boldsymbol{q}) = -\mathcal{J}_{dd}\{0.919 + 0.0816\cos(q_z/2) - 0.0006\cos q_z\} \tag{27}$$

for $\boldsymbol{q}$ along the z-axis, $\mathcal{J}_{dd} = 4\pi(g\mu_B)^2 N/V \simeq 0.035$ meV. The hcp lattice is not of a Bravais type. However, if we neglect the optical mode (which is possible at $T \ll T_N$) one can put (see, e.g., reference [2])

$$J_{\boldsymbol{q}} = 2J\left[\cos q_x + 2\cos(q_x/2)\cos(\sqrt{3}q_y/2)\right] + 2J'\cos\frac{q_z}{2}\left|\exp(\mathrm{i}q_y/\sqrt{3}) + 2\cos\frac{q_x}{2}\exp(-\mathrm{i}q_y/2\sqrt{3})\right|. \tag{28}$$

The contribution of the dipolar anisotropy can be roughly taken into account by the renormalization of second-order anisotropy parameter

$$(B_2^0)_R = B_2^0 + 0.0816\mathcal{J}_{dd}. \tag{29}$$

Other parameters of the Hamiltonian were also taken from reference [15]: $J = 0.65$ K, $J' = 0.6J$, $B_2^0 = 0.3$ K, $B_4^0 = 0$, $B_6^0 = -1.1 \times 10^{-5}$ K, $B_6^6 = 1.07 \times 10^{-4}$ K. For simplicity, we restrict ourselves to a collinear magnetic structure (this is justified by the spiral angle in the rare earths being small, especially at low temperatures). Then the calculations with the Hamiltonian (26) are completely analogous to those in the previous section. The calculated dependence of the cone angle is compared with the result of the mean-field approximation and experimental data in figure 3. One can see that our results improve somewhat on those of the mean-field theory where the temperature dependence of the anisotropy constants is given by (8). Note that due to the inequality $T \ll T_N$ the decrease of the magnetization in the temperature interval 0–20 K under consideration makes up only about 1%. Thus one can expect our spin-wave results to be sufficiently accurate (unlike those from the mean-field approach, where errors are uncontrollable).

To conclude, we have formulated a consistent spin-wave approach to the description of thermodynamic properties of anisotropic magnets at low temperatures. The renormalizations of the anisotropy constants and the spin-wave spectrum for an arbitrary cone angle are calculated. This gives the possibility of describing the orientational phase transition between the cone and plane phases.

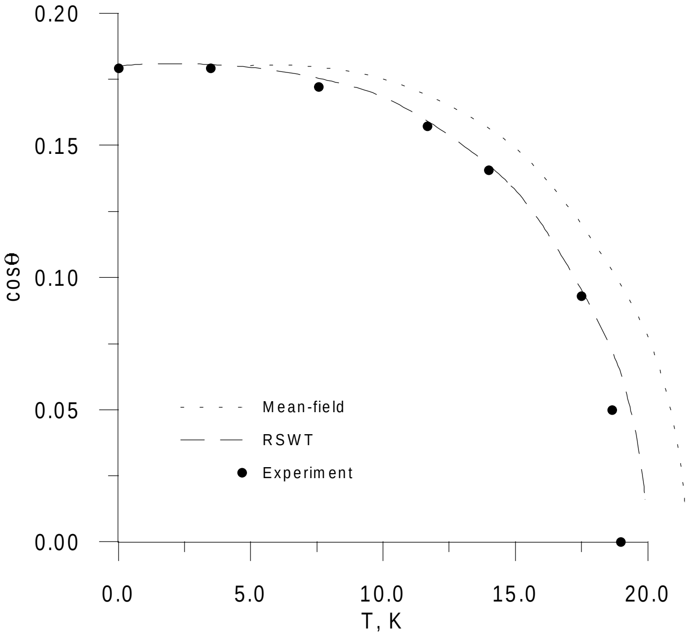

Figure 3. Calculated dependences of the cone angle in the mean-field approximation (short-dashed line) and renormalized spin-wave theory (RSWT, long-dashed line) as compared with experimental points for holmium (references [2, 16]).

Acknowledgment

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