Magnetic-structure instability resulting from the intersection of energy levels

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We investigated the instability of the magnetic structure of a ferrimagnetic crystal (of the iron-garnet type), which is produced in a magnetic field when the lower energy levels of one of the sublattices intersect. This phenonmenon is the magnetic analog of the Jahn-Teller effect. The phase diagram of this system is constructed.

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1. The Jahn—Teller effect in crystals is well known, and reduces to the fact that the crystalline environment of the ions with orbitally degenerate lower levels is unstable to a deformation that lowers the symmetry of the crystal and by the same token leads to lifting of the orbital degeneracy. An interesting modification of this phenomenon can be realized in a magnetic crystal. If in a magnetically ordered crystal (we shall call it a matrix) there are ions with degenerate ground states and having anisotropic exchange (or dipole) interaction with the matrix, then the degeneracy can be lifted by deformation of the magnetic structure with lowering of magnetic symmetry of the surrounding of the ion, and if the phenomenon has a cooperative character, the symmetry of the entire crystal can be lowered. The reason for this phenomenon can be explained in the following manner (in analogy with the usual Jahn—Teller effect): Assume that in the symmetrical phase the ground state of the ion is doubly degenerate. We shall characterize the magnetic-structure deformation that leads to a lowering of the symmetry by the angles of inclination of the magnetic moments of the matrix from the equilibrium position in the symmetrical phase. This deformation causes a level splitting that is linear in θ ($\Delta E = \pm a\theta$, where a is the constant of the interaction of the ion with the matrix). The energy of the ground state of the ion becomes lower. On the other hand, the deformation leads to an energy gain $b\theta^2/2 > 0$ (b is the "rigidity" of the matrix). The fact that the energy contains terms linear in θ leads to an instability of the system;

the quadratic terms hinder the development of the instability. The structure that is produced in the course of the instability (at $T = 0\,^\circ\text{K}$) is determined by the condition that the total energy be a minimum

$$E = \frac{b\theta^2}{2} - a|\theta| . \tag{1}$$

The minimum is reached at $\theta = \pm a/b$. At large concentrations of ions with degenerate levels, a cooperative magnetic phase transition takes place. Examples of substances for such phenomena can take place are rare-earth (RE) compounds with elements of the iron group. In these compounds, the role of the matrix is played by the iron ions, and a degenerate ground state can be realized in the RE ions.[1)]

2. The considered mechanism of the instability of the magnetic structure can be of particular interest in a situation in which the lower levels overlap in an external magnetic field H, a fact that is possible if their rates of change with changing magnetic field are different. Let us investigate, as a function of H and T, the character of the instability that is used thereby and the ensuing phase transitions. Let us describe the model in greater detail.[2)] Let the crystal have two subsystems, a magnetic "matrix" and a paramagnetic two-level system; the subsystems are coupled by an exchange interaction. The external and exchange fields are parallel and directed along a definite crystal axis. The energy of the "matrix," as before, is equal to $b\theta^2/2$, where the angle θ is reckoned from H in a definite plane. We confine ourselves below to the case of small angles. We assume that in the crystal there are two nonequivalent positions of the paramagnetic ions and this, as will be shown below, is important and conforms to the real situations. The energy levels of the ions depend on H and θ; at $\theta = 0$ they intersect in the field $H = H_0$; in the approximation linear in θ, they take the form

$$\text{position A: } E_{1,2} = \pm\Delta(H) \mp a\theta ; \qquad \text{position B: } E_{1,2} = \pm\Delta(H) \pm a\theta . \tag{2}$$

$\Delta(H)$ at the point of intersection (at $H = H_0$) reverses sign. The presence of two positions ensures stability of the state $\theta = 0$ far from the point of intersection of the levels, for in this case the linear terms in the expansion $E_{1,2}(\theta)$ cancel each other.

3. We consider the case $T = 0$. The total energy of the system is

$$E = \frac{b\theta^2}{2} - \frac{x}{2}\left(|\Delta - a\theta| + |\Delta + a\theta|\right), \tag{3}$$

where x is the concentration of the paramagnetic ions. We see that at the point of intersection $[\Delta(H) = 0]$ the energy (3) coincides with (1) with all the ensuing consequences. Near the intersection point, at $|\Delta| < xa^2/b$, the energy (3) has two minima at $\theta_1 = 0$ and $\theta_2 = \pm xa/b$. At $|\Delta| < xa^2/2b$, the states $\theta = \pm xa/b$ are absolute minima, and the state $\theta = 0$ is metastable; at $xa^2/2b < |\Delta| < xa^2/b$, the inverse situation is realized. This means that at the points $\Delta(H) = \pm xa^2/2b$ there occur first-order phase transitions with respect to the field. In the canted phase ($\theta \neq 0$) the levels of the paramagnetic ions will be repelled, and in different manners for positions A and B.

4. At a finite temperature T, the nonequilibrium thermodynamic potential (TP) of the system can be represented in the form

$$\Phi = -\frac{xT}{2}\sum_{i=1}^{2}\ln\left[\exp\left(-\frac{E_1^i}{T}\right) + \exp\left(-\frac{E_2^i}{T}\right)\right] + \frac{b\,\theta^2}{2}, \tag{4}$$

where the index i numbers the positions A and B in terms of the dimensionless variables $\Theta = \theta/\theta_0$, $W = \Delta/a\theta_0$, $\tau = T/2a\theta_0$, and $F = \Phi/b\theta_0^2$, where $\theta_0 = ax/b$, Eq. (4) takes the form

$$F = \frac{\Theta^2}{2} - \tau\ln\left[\mathrm{ch}\left(\frac{W+\Theta}{2\tau}\right)\mathrm{ch}\left(\frac{W-\Theta}{2\tau}\right)\right]. \tag{5}$$

Thus, the dependence on the physical parameters (x, a, b) enters into these expressions only in terms of the scales of the quantities θ, Δ, and T. The formal problem of minimizing TP reduces to the following picture (see Fig. 1). There

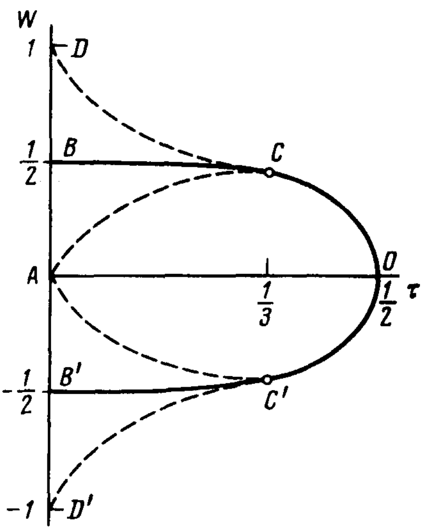

FIG. 1. Phase diagram (numerical calculation): BC, B′, C′—first-order phase-transition line; CC′—second-order phase-transition line; AC, AC′—line of collinear-phase stability loss; C′D′, CD—line of canted-phase loss line.

are two solutions (two phases): a) the collinear phase $\Theta = 0$; b) the canted phase $\Theta_1 = \Theta(W, \tau)$, $\Theta_2 = -\Theta_1$. The collinear phase on the $W\tau$ plane is stable everywhere with the exception of the region bounded by the curve ACOC′A, and this curve, which is the line where the phase $\Theta = 0$ loses stability, is given by the following equations in parametric form

$$W = \eta(1 + \mathrm{ch}\,\eta)^{-1}; \qquad \tau = (1 + \mathrm{ch}\,\eta)^{-1}. \tag{6}$$

The canted phase [the function $\Theta(W, \tau)$, calculated numerically, is shown in Fig. 2] is stable in the region bounded by the curve DCOC′D′, which is given in parametric form as follows:

$$W = \frac{\mathrm{cth}\,\xi - \xi^{-1}}{\xi}\,\mathrm{Arch}\left(\frac{\xi - \mathrm{ch}\,\xi\,\mathrm{sh}\,\xi}{\mathrm{sh}\,\xi - \xi\,\mathrm{ch}\,\xi}\right); \qquad \tau = \frac{1}{\xi}\mathrm{cth}\,\xi - \frac{1}{\xi^2}. \tag{7}$$

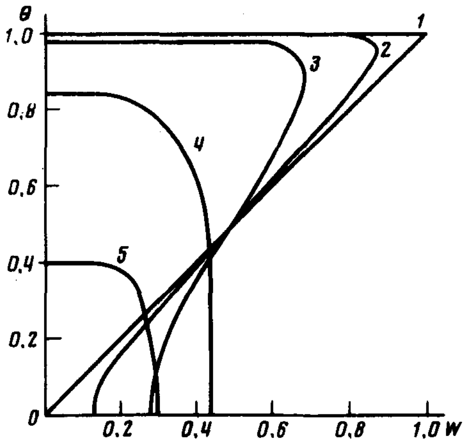

FIG. 2. Dependence of the angle Θ on $W(H)$ at different τ: 1) $\tau=0$, 2) 0.025, 3) 0.1, 4) 0.325, 5) 0.45.

At the points C and C′, the stability-loss curves (6) and (7) coalesce smoothly (with equal tangents) into a second-order phase-transition line. In the regions DCA and D′C′A the phases coexist. We note the possibility of the appearance of a domain structure in the region bounded by the curve DCOC′D′. The domain structure can be of two types: a) "thin" type because of the double degeneracy of the canted phase in the region ACOC′A; b) intermediate-state type near the lines of the coexistence of the phases (the lines DC and D′C′). It is quite possible that the similarities observed in[1] in the magnetization and magnetostriction of yttrium-terbium garnets is a manifestation of the considered instability of the magnetic structure.

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1)From this point of view we can consider, for example, the orientational transition $HoFeO_3$. In this substance at $T > 50$ °K the antiferromagnetism vector **l** is parallel to the **a** axis of the crystal, and the anisotropy of the g-factor is such that the exchange splitting of the lower levels is equal to zero. The deviation of 1 from the axis in the ac plane causes their splitting, which reaches a maximum at **l** ∥ **c** (~3—4 cm^{-1}). The transition of **l** from **a** to **c** in accordance with the foregoing is favored and is actually observed at $T = 50$ °K.

2)The nearest prototype of this model is YIG with RE ion impurities (Tb, Ho, ...).

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