Essay

Orientational Phase Transitions

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Orientational Phase Transitions

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Abstract

A brief review of orientational phase transitions and thermodynamic properties of various magnets. These methodological guidelines are intended for students studying the section "Theory of Phase Transitions" in the course "Theory of Solids", as well as the section "Magnetic Phase Transitions" in the course "Theory of Magnetism". They can be used in preparation for laboratory and seminar classes in these courses, and for independent research work by students of the Faculty of Physics.

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1. Phase Transitions

1.1. Introduction

The definition of the phase (phase state) concept is based on the symmetry of the crystal, both spatial and magnetic. Phase transitions (PT), as a rule, occur with a change in the crystal's symmetry. Thus, during structural phase transitions, the crystal before and after the transition has a different crystal structure. For small atomic displacements, some symmetry elements are lost, and symmetry is lowered (distortional PT); however, there is a subgroup relationship between the low-symmetry (usually low-temperature) and high-symmetry (usually high-temperature) phases. For large atomic displacements during PT, a subgroup relationship between the symmetry groups of the two phases may not exist (reconstructive PT). In both cases, we are dealing with PTs of the displacement type. A structural PT can also occur as an "order-disorder" transition (atomic ordering in alloys like CuZn...). Classical objects for the physics of PT are magnetic materials, which is primarily due to the diversity of observed magnetic structures. Among magnetic PTs, the best-known are "order-disorder" type transitions at Curie (T_C – ferromagnetic ordering temperature) and Néel (T_N – antiferromagnetic ordering temperature) points. A common characteristic feature of such transitions is the appearance, for $T < T_C$ (T_N), of a non-zero average magnetic moment per atom. In other words, these PTs occur with a change in the magnitude of the average atomic magnetic moment. Among magnetic "order-disorder" type PTs, the so-called magnetic orientational PTs or spin-reorientation (SR) transitions are particularly distinguished. A characteristic feature of such transitions is a smooth or sharp change in the orientation (direction) of the atomic magnetic moments (spins).

Like other phase transitions, SR transitions can be spontaneous, i.e., occur solely under the influence of temperature change, or induced by an external magnetic field, electric field, or mechanical stress. A number of authors include in the concept of an SR transition only transitions with a change in magnetic state (orientation of magnetic moments) but without a change in the magnetic structure (ferro-, ferri-, or antiferromagnetic). In many cases, the concept of an SR transition is extended to all transitions accompanied by a change in the orientation of the magnetic moments of atoms or magnetic sublattices in multi-sublattice magnets. In this case, SR transitions include such well-known magnetic ones as spin-flop (reorientation of sublattices in antiferromagnets in an external magnetic field), the Morin transition (ferro – antiferromagnet transition), the metamagnetic transition (antiferromagnet – ferromagnet transition in highly anisotropic materials in an external field).

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A feature of orientational PTs is the wide applicability range of the simple Landau theory, which, unlike PTs at the Curie point, is valid up to temperatures differing from the transition temperature by an extremely small amount $10^{-6} \div 10^{-8}$ K.

1.2. Elements of Landau Theory

In the thermodynamic description of PT, the order parameter η introduced by L.D. Landau is of fundamental importance. This parameter always characterizes some new property that appears in the system as a result of a PT from the original phase where it was absent. In other words, the order parameter is zero in the original (high-symmetry) phase and non-zero in the new (low-symmetry) phase. Thus, during a 2nd order phase transition, a symmetry breaking of the system occurs.

In the general case, the parameter η can be multi-component (e.g., two angles of magnetic moment orientation). Below we consider the simplest theory of spontaneous 2nd order PTs.

The thermodynamic potential is written as a polynomial including invariant combinations of the order parameter to various powers. For example, for a single-component parameter η:

$$\Phi(\eta) = \Phi_0 + A\eta^2 + B\eta^4 + \cdots \tag{1}$$

A linear term in the expression for $\Phi(\eta)$ is absent because the condition for the energy minimum for the equilibrium of the new phase requires that

$$\frac{\partial \Phi}{\partial \eta} = 0, \tag{2}$$

including at the transition point itself ($\eta = 0$). Along with the minimum condition (1.2), the condition for phase stability must be fulfilled

$$\left.\frac{\partial^2 \Phi}{\partial \eta^2}\right|_{\eta=\eta_0} > 0, \tag{3}$$

Bellow we will only use terms with η^2, η^4. Depending on A and B parameters there are 4 cases for $\Phi(\eta)$ dependency (Figure 1).

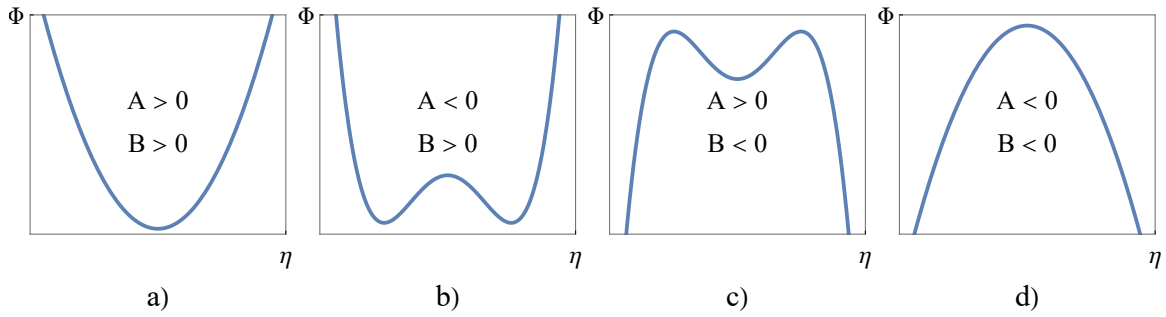

Figure 1

Case (d) is uninteresting because it does not correspond to any stable states. Case (c) corresponds to the existence of a metastable state. The most interesting are cases (a) and (b). In case (a), the energy minimum corresponds to $\eta = 0$ (original phase); in case (b) — to $\eta \neq 0$ (new phase). During the transition from (a) to (b), the coefficient A changes sign. Since we are considering a spontaneous PT, the only reason for the change in A can be a change in temperature T. Expanding $A(T)$ near the transition point ($T = T_c$) in a Taylor series and keeping only the first (linear) term, we have

$$A = A_0(T - T_c). \tag{4}$$

We will consider the coefficient B to be constant hereafter: $B = \text{const}$.

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From the condition for the minimum of the thermodynamic potential, we find

$$\frac{\partial \Phi}{\partial \eta} = (2A + 4B\eta^2)\eta = 0, \tag{5}$$

which gives two solutions:

1. $\eta = 0$ for original phase;
2. $\eta^2 = -\frac{A}{2B}$ for new phase ($B > 0$).

The stability condition gives

$$\frac{\partial^2 \Phi}{\partial \eta^2} = 2A + 12B\eta^2 > 0, \tag{6}$$

i.e., the original phase ($\eta = 0$) is stable for $A > 0$, and the new phase ($\eta \neq 0$) for $A < 0$. In the new phase

$$\eta^2 = -\frac{A}{2B} = -\frac{A_0}{2B}(T - T_c), \tag{7}$$

or

$$\eta = \pm\left[\frac{A_0}{2B}(T_c - T)\right]^{\frac{1}{2}}. \tag{8}$$

The obtained temperature dependence of the order parameter near T_c is characteristic of Landau theory.

Note also that in the low-symmetry phase, the parameter η can be either positive or negative. This fact reflects the so-called Curie principle, according to which a dissymmetry appearing in a system must be present in the causes that give rise to it.

According to this principle, the symmetry of a crystal should not seem to change with temperature, since the latter is a scalar. This contradiction between the Curie principle and the aforementioned emergence of symmetry breaking during second-order phase transitions is resolved by the crystal breaking into domains with different signs of η for $T < T_c$. The symmetry within each domain is lower than in the high-temperature phase; however, their arrangement in the crystal is determined by those symmetry elements that were lost during the phase transition, as a result of which the overall symmetry of the crystal remains unchanged.

1.3. Elements of Thermodynamics of Solids

In the thermodynamic description of solids, it is customary to introduce the so-called generalized thermodynamic forces (temperature T, electric E and magnetic H field, mechanical stress σ) and generalized thermodynamic coordinates (entropy S, electric displacement field D, magnetic flux density B, strain ε).

The thermodynamic potential (TDP) Φ, or Gibbs free energy, is a function of the form

$$\Phi = \mathcal{U} - TS - \sigma_{ij}\varepsilon_{ij} - \cdots = \mathcal{U} - X_i x_i, \tag{9}$$

where $\mathcal{U}$ is an internal energy, X_i is a generalized force, x_i is a generalized coordinate. The total differential of Φ is

$$d\Phi = -SdT - \varepsilon_{ij}d\sigma_{ij} - \cdots = -x_i dX_i, \tag{10}$$

since for the internal energy

$$d\mathcal{U} = TdS + \sigma_{ij}d\varepsilon_{ij} + \cdots = X_i dx_i. \tag{11}$$

If Φ is considered as a function of the generalized forces, then for the generalized coordinates we have

$$x_i = -\frac{\partial \Phi}{\partial X_i}. \tag{12}$$

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Let us consider as the initial state one in which external forces are absent ($\mathbf{E} = 0$, $\mathbf{H} = 0$, $\sigma = 0$, $T = T_0$). Expanding the generalized coordinates x_i in a Taylor series, we obtain

$$x_i = -\frac{\partial \Phi}{\partial X_i}\bigg|_0 - \frac{\partial^2 \Phi}{\partial X_i \partial X_j}\bigg|_0 X_j - \cdots = -\frac{\partial \Phi}{\partial X_i}\bigg|_0 + M_{ij} X_j - \cdots . \tag{13}$$

Then the TDP can be represented as

$$\Phi = \Phi_0 + \frac{\partial \Phi}{\partial X_i}\bigg|_0 X_i + \frac{1}{2}\frac{\partial^2 \Phi}{\partial X_i \partial X_j}\bigg|_0 X_i X_j + \cdots = \Phi_0 + \frac{\partial \Phi}{\partial X_i}\bigg|_0 X_i - \frac{1}{2} M_{ij} X_i X_j + \cdots . \tag{14}$$

Most often, the second term in the expansion of Φ is absent (exceptions are crystals without spontaneous electric or magnetic polarization, nor spontaneous strain, and ferroelectrics, ferromagnets, and ferroelastics — crystals with spontaneous deformation). The tensors $M_{ij} = -\partial^2 \Phi / \partial X_i \partial X_j \big|_0$ have the meaning of generalized susceptibilities, for example:

$$-\frac{\partial^2 \Phi}{\partial T^2}\bigg|_0 = \frac{C_p}{T},$$

where C_p is heat capacity at constant pressure;

$$-\frac{\partial^2 \Phi}{\partial E_i \partial E_j}\bigg|_0 = \varepsilon_{ij}$$

- dielectric susceptibility (permittivity);

$$-\frac{\partial^2 \Phi}{\partial E_i \partial T}\bigg|_0 = p_i^E$$

- pyroelectric coefficients;

$$-\frac{\partial^2 \Phi}{\partial H_i \partial H_j}\bigg|_0 = \mu_{ij}$$

- magnetic susceptibility (permeability);

$$-\frac{\partial^2 \Phi}{\partial H_i \partial T}\bigg|_0 = p_i^M$$

- pyromagnetic coefficients;

$$-\frac{\partial^2 \Phi}{\partial \sigma_{ij} \partial \sigma_{kl}}\bigg|_0 = S_{ijkl}$$

- tensor of elastic compliances;

$$-\frac{\partial^2 \Phi}{\partial \sigma_{ij} \partial T}\bigg|_0 = \alpha_{ij}^{\sigma}$$

- coefficients of thermal expansion (at constant stress);

$$-\frac{\partial^2 \Phi}{\partial E_i \partial \sigma_{jk}}\bigg|_0 = d_{ijk}^E$$

- piezoelectric coefficients;

$$-\frac{\partial^2 \Phi}{\partial H_i \partial \sigma_{jk}}\bigg|_0 = d_{ijk}^M$$

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- piezomagnetic coefficients. The tensor M of generalized susceptibilities is symmetric: $M_{ij} = M_{ji}$, i.e., M_{ij} determines both the change in the coordinate x_i under the action of the force X_j, and vice versa, the change in x_j under the action of X_i. Thus, for example, the pyromagnetic coefficient p_i^M determines the change in magnetic induction with temperature and the change in entropy under the action of a magnetic field (the magnetocaloric effect). Incidentally, the magnetocaloric effect can be realized in two variants:

1. Isothermal process ($\Delta T = 0$)

$$\Delta S = p_k^M H_k = (\mathbf{p}^M \cdot \mathbf{H}), \tag{15}$$

i.e., the crystal absorbs heat from the surroundings:

$$\Delta Q = T\Delta S = T(\mathbf{p}^M \cdot \mathbf{H}). \tag{16}$$

(It releases heat if $\mathbf{p}^M \cdot \mathbf{H} < 0$).

2. Adiabatic process (heat exchange with the surroundings is excluded, and $\Delta S = 0$). In this case:

$$\Delta S = -\frac{C_p}{T}\Delta T + \mathbf{p}^M \cdot \mathbf{H} = 0, \tag{17}$$

and

$$\Delta T = \frac{T}{C_p}(\mathbf{p}^M \cdot \Delta\mathbf{H}). \tag{18}$$

Considering that, as a rule, for paramagnets

$$\mathbf{p}^M = -\frac{\partial^2\Phi}{\partial T \partial \mathbf{H}}\bigg|_0 = \frac{\partial \mathbf{I}}{\partial T}\bigg|_{\mathbf{H}} < 0,$$

we arrive at the conclusion that the temperature decreases when the field is reduced (magnetic cooling).

1.4. Features of Generalized Susceptibilities Near a 2nd Order PT

In the general case, the TDP of a crystal is a function of the order parameter and thermodynamic forces $T, \mathbf{E}, \mathbf{H}, \sigma, \ldots$: $\Phi = \Phi(\eta, T, \mathbf{E}, \mathbf{H}, \sigma)$.

Here, the order parameter is itself a function of the forces $(T, \mathbf{E}, \mathbf{H}, \sigma, \ldots)$ and is determined from the condition of the minimum of the TDP; in other words, the only independent variables in the TDP are the forces. It is completely obvious that the nature of phase transitions from the high-temperature phase should not change if the crystal is preliminarily subjected to any symmetry transformation that maps it onto itself. Consequently, the TDP used to describe a phase transition must be invariant under the transformations of the symmetry group of the high-temperature phase.

The expansion of the TDP near the PT point includes three types of terms of minimal degree in η and X:

1. Terms of the type $\eta^2, \eta^4, \ldots$;
2. Terms of the type $X_i X_j, \ldots$;
3. Terms of the type $\eta X_i, \eta^2 X_i, \ldots, \eta X_i, \eta^2 X_i X_j$.

The order parameter is found from the condition of the minimum of the TDP:

$$\frac{\partial\Phi(T, X, \eta)}{\partial\eta}\bigg|_{X=X_0} = 0, \quad \frac{\partial^2\Phi(T, X, \eta)}{\partial\eta^2}\bigg|_{X=X_0} > 0, \tag{19}$$

where

$$\Phi = \Phi_0 + A\eta^2 + B\eta^4 - \frac{1}{2}MX^2 + N\eta X + K\eta^2 X + \cdots \tag{20}$$

(for simplicity, we restrict ourselves to only one force X). Below we analyze the PT, keeping in the TDP only the terms highlighted in (20).

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1.4.1. Accounting for Terms of the Type ηX ($N \neq 0, K = 0$)

The first of the TDP minimum conditions (19) in this case reduces to

$$\frac{\partial \Phi}{\partial \eta} = 2A\eta + 4B\eta^3 + NX = 0. \tag{21}$$

Differentiating this equality with respect to X, we get

$$2A\frac{\partial \eta}{\partial X} + 12B\eta^2\frac{\partial \eta}{\partial X} + N = 0, \tag{22}$$

whence

$$\frac{\partial \eta}{\partial X} = -\frac{N}{2A + 12B\eta^2} = \begin{cases} -\dfrac{N}{2A} & \text{(high-symmetry phase)}, \\ -\dfrac{N}{2A + 12B\eta^2} & \text{(low-symmetry phase)}. \end{cases}$$

Let us find the generalized coordinate x:

$$x = -\frac{d\Phi}{dX} = -\frac{\partial \Phi}{\partial X} - \frac{\partial \Phi}{\partial \eta}\Big|_X \frac{\partial \eta}{\partial X} = -\frac{\partial \Phi}{\partial X}\Big|_\eta, \tag{I.22}$$

since $\partial\Phi/\partial\eta\big|_X = 0$. Thus, differentiating x in (20) with respect to X and using (21), we obtain

$$x = MX - N\eta. \tag{23}$$

Let us define the effective (renormalized) susceptibility M_{eff} as

$$M_{\text{eff}} = \frac{\partial x}{\partial X} = -\frac{\partial^2 \Phi}{\partial X^2} = M + N\frac{\partial \eta}{\partial X} =$$

$$= M + \begin{cases} -\dfrac{N^2}{2A} & \text{(high-symmetry phase)}, \\ -\dfrac{N^2}{2A + 12B\eta^2} & \text{(low-symmetry phase)}. \end{cases}$$

Substituting $\eta^2 = -A/2B$, we get

$$M_{\text{eff}} = M + \begin{cases} -\dfrac{N^2}{2A} & \text{(high-symmetry phase)}, \\ +\dfrac{N^2}{4A} & \text{(low-symmetry phase)}. \end{cases} \tag{24}$$

Considering that near $T \sim T_c$, $A = A_0(T - T_c)$ and $A_0 > 0$, we obtain

$$M_{\text{eff}} = M_0 + \begin{cases} -\dfrac{N^2}{2A_0}\tau^{-1}, & T > T_c, \\ +\dfrac{N^2}{4A_0}|\tau|^{-1}, & T < T_c, \end{cases} \tag{25}$$

where $\tau = T - T_c$.

Thus, when terms of the type ηX, linear in the order parameter and the force, are present in the TDP, the generalized susceptibility corresponding to the force X — generalized susceptibility has at T_c a characteristic hyperbolic singularity.

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1.4.2. Accounting for Terms of the Type $\eta^2 X$ ($N = 0, K \neq 0$)

In this case, the first of the minimum conditions (19) for the TDP reduces to

$$\frac{\partial \Phi}{\partial \eta} = 2\eta(A + KX) + 4B\eta^3 = 0.$$

Differentiating with respect to X gives

$$2\eta'(A + KX) + 2K\eta + 12B\eta^2\eta' = 0,$$

or

$$\eta' = \frac{\partial \eta}{\partial X} = -\frac{K\eta}{A + KX + 6B\eta^2}.$$

For the coordinate, we have

$$x = MX - K\eta^2,$$

and for the effective generalized susceptibility (at $X = 0$)

$$M_{\text{eff}} = M - 2K\eta\frac{\partial \eta}{\partial X} = M + \frac{2K^2\eta^2}{A + 6B\eta^2},$$

$$M_{\text{eff}} = \begin{cases} M, & T > T_c, \\ M + \dfrac{K^2}{|A|}, & T < T_c, \end{cases} \tag{26}$$

taking into account that $\eta_0^2 = -\dfrac{A}{2B}$.

Thus, if the TDP contains only terms of the type $\eta^2 X$, which are quadratic in the order parameter but linear in the force X, then the corresponding generalized susceptibility experiences a jump at the same 2nd order PT point.

2. Spontaneous Orientational Phase Transitions

2.1. Orientational Phase Transitions in Uniaxial Magnets

Without an external magnetic field a thermodynamic potential (TDP) of an uniaxial magnet has the following form:

$$\Phi = K_1 \sin^2\theta + K_2 \sin^4\theta, \tag{27}$$

where K_1 and K_2 are the first and second anisotropy constants, θ is the angle between the z-axis and a magnetic moment $\overrightarrow{M}$. The TDP minimum condition

$$\frac{\partial \Phi}{\partial \theta} = \sin 2\theta\left(K_1 + 2K_2 \sin^2\theta\right) = 0 \tag{28}$$

gives us

$$\begin{aligned} &\text{I. } \sin 2\theta = 0, \text{ i.e. the phases } M_{||} \ (\theta = 0) \text{ and } M_{\perp} \ (\theta = \pi/2) \\ &\text{II. } \sin^2\theta = -\frac{K_1}{2K_2}, \text{ i.e. the angular phase } M_{>}. \end{aligned} \tag{29}$$

The thermodynamic stability condition of the phases

$$\frac{\partial^2 \Phi}{\partial \theta^2} = 2\cos 2\theta\left(K_1 + K_2 \sin^2\theta\right) + 2K_2 \sin^2 2\theta \geq 0 \tag{30}$$

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leads us to the following relations:

$$\begin{aligned} &a)\ K_1 \geq 0 \text{ for the } M_{||}\text{-phase,} \\ &b)\ K_1 + 2K_2 \leq 0 \text{ for the } M_{\perp}\text{-phase,} \\ &c)\ K_2 \geq 0 \text{ at } K_1 \leq 0 \text{ and } K_1 + 2K_2 \geq 0 \text{ for the } M_{>}\text{-phase,} \end{aligned} \tag{31}$$

which define a regions of the phase existence (Figure 2).

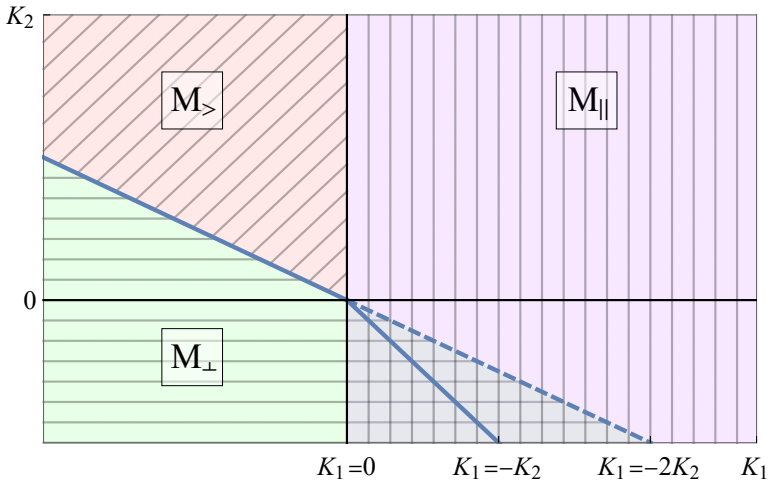

Figure 2

The equality to zero in the relations (31) define lines of the stability loss (lability borders). Values of the TDP in the different phases

$$\Phi(M_{||}) = 0;\ \Phi(M_{\perp}) = K_1 + K_2;\ \Phi(M_{>}) = \frac{K_1^2}{4K_2} \tag{32}$$

allow us to find the exact form of equations, which define the lines of the phase transitions:

$$\begin{aligned} &a)\ K_1 + K_2 = 0,\ K_1 \geq 0\ \left(M_{||} \leftrightarrow M_{\perp}\right); \\ &b)\ K_1 = 0,\ K_2 \geq 0\ \left(M_{||} \leftrightarrow M_{>}\right); \\ &c)\ K_1 + 2K_2 = 0,\ K_1 \leq 0\ (M_{\perp} \leftrightarrow M_{>}). \end{aligned} \tag{33}$$

A magnetic phase diagram of the uniaxial magnet in the (K_1, K_2) coordinates shown in the Figure 2 (the bold lines is the PT lines, the dashed lines represent the phase stability loss, or the lability borders).

Let us pay attention to the presence of the phase coexistence areas ($M_{||}$ and $M_{\perp}$) in the last case, which makes possible an appearance of a hysteresis and metastable states. A schematic dependence of the orientation angle of the vector $\overrightarrow{M}$ versus the anisotropy K_1 at $K_2 = \text{const} < 0$ shown in the Figure 3. The sectors $2 \to 3$ and $5 \to 0$ correspond to the metastable states, which can appear in a single-domain sample. The transition $M_{||} - M_{\perp}$ could occur without hysteresis loops in the way $1 \leftrightarrow 2 \leftrightarrow 5 \leftrightarrow 4$, because there are nucleus of the new phase in the domain borders of the old phase, which grows through the all crystal volume at K_1 changes with temperature.

A smooth $M_{||} \to M_{\perp}$ orientational phase transition (OPT) realized with two second-order phase transition ($M_{||} \to M_{>}$ and $M_{>} \to M_{\perp}$) through the intermediate angular phase $M_{>}$. Schematically the dependence of the orientation angle θ versus the K_1 at $K_2 = \text{const} > 0$ given in the Figure 4. Note that in the angular phase the derivative

$$\frac{\partial \theta}{\partial K_1} = -(2K_2 \sin 2\theta)^{-1} \tag{34}$$

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becomes infinity when $\theta = 0$ and $\theta = \pi/2$. In a conventional OPT model it is considered that in the transition region $K_2 = \text{const}$ and K_1 is the linear function of temperature:

$$K_1(T) = -2K_2 \frac{T - T_2}{T_1 - T_2}, \tag{35}$$

where T_1 and T_2 are the transition temperatures $M_\perp \leftrightarrow M_>$ and $M_{||} \leftrightarrow M_>$ respectively. Then in the angular phase we have the following equations (with Figure 5):

$$\sin^2\theta = \frac{T - T_2}{T_1 - T_2},$$
$$\frac{\partial\theta}{\partial T} = \frac{1}{(T_1 - T_2)\sin 2\theta} = \frac{1}{2\sqrt{(T - T_1)(T_2 - T)}}, \quad T_1 < T < T_2. \tag{36}$$

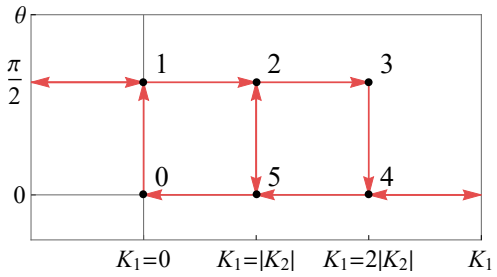

Figure 3

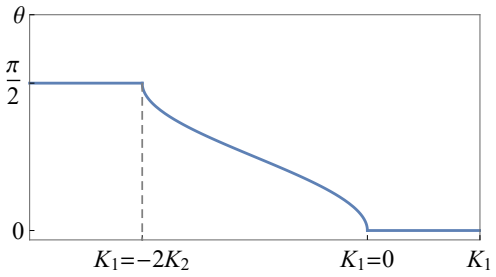

Figure 4

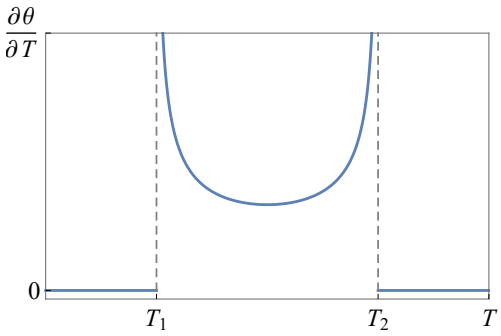

Figure 5

In the uniaxial magnet the $M_{||}$-phase could be named "easy-axis" phase, and the $M_\perp$-phase is "easy-plane". In the angular phase $M_>$ takes place a cone of the easy-axes.

The given analysis could be applicable to an OPT in a certain crystallographic plane of a rhombic magnet.

2.2. Magnetic Part of a Heat Capacity at an OPT in Uniaxial Magnets

We use the well-known relation for a heat capacity at constant pressure

$$C_p = -T\frac{\partial^2\Phi}{\partial T^2}, \tag{37}$$

and taking into account $\Phi = \Phi(\theta, T)$ for an uniaxial magnet we get

$$C_p(m) = -T\left[\frac{\partial^2\Phi}{\partial T^2}\bigg|_\theta + 2\frac{\partial^2\Phi}{\partial\theta\partial T}\cdot\frac{\partial\theta}{\partial T} + \frac{\partial^2\Phi}{\partial\theta^2}\cdot\left(\frac{\partial\theta}{\partial T}\right)^2\right], \tag{38}$$

where we used the TDP minimum condition $\partial\Phi/\partial\theta = 0$.

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From differentiating the minimum condition by temperature T we get

$$\frac{\partial^2\Phi}{\partial\theta\partial T}+\frac{\partial^2\Phi}{\partial\theta^2}\cdot\frac{\partial\theta}{\partial T}=0,$$

or

$$\frac{\partial\theta}{\partial T}=-\frac{\Phi''_{\theta T}}{\Phi''_{\theta\theta}}. \tag{39}$$

Thus, we can rewrite the magnet part of the heat capacity in the form

$$\begin{aligned} C_p(m)&=-T\left[\left.\frac{\partial^2\Phi}{\partial T^2}\right|_\theta-\left.\frac{\partial^2\Phi}{\partial\theta^2}\right|_T\left(\frac{\partial\theta}{\partial T}\right)^2\right]= \\ &=-T\left(\Phi''_{TT}-\frac{\left(\Phi''_{\theta T}\right)^2}{\Phi''_{\theta\theta}}\right)=C'_p(m)+C''_p(m), \end{aligned} \tag{40}$$

where the second term is considered as the pure "orientational" contribution. With the account of (30) for $\Phi''_{\theta\theta}$ and

$$\Phi''_{\theta T}=\sin 2\theta\left(\frac{\partial K_1}{\partial T}+2\frac{\partial K_2}{\partial T}\sin^2\theta\right) \tag{41}$$

we get the following equation for the orientational term in $C_p(m)$:

$$\begin{aligned} C''_p(m)&=T\frac{\sin^2 2\theta\left(\frac{\partial K_1}{\partial T}+2\frac{\partial K_2}{\partial T}\sin^2\theta\right)}{2\cos 2\theta\left(K_1+2K_2\sin^2\theta\right)+2K_2\sin^2 2\theta}= \\ &=\begin{cases} 0, & M_{||}\text{-phase} \\ \dfrac{T}{2K_2}\left(\dfrac{\partial K_1}{\partial T}-\dfrac{\partial K_2}{\partial T}\cdot\dfrac{K_1}{K_2}\right)^2, & M_{>}\text{-phase} \\ 0, & M_{\perp}\text{-phase} \end{cases} \end{aligned} \tag{42}$$

Assuming $K_2=\text{const}$ and a linear temperature dependence of $K_1(T)$ like in (35) we have $C'_p(m)=0$ and

$$C''_p(m)=\begin{cases} 0, & M_{||}\text{-phase} \\ \dfrac{2K_2T}{(T_1-T_2)^2}, & M_{>}\text{-phase} \\ 0, & M_{\perp}\text{-phase} \end{cases} \tag{43}$$

A schematic temperature dependence of $C''_p(m)$ at a smooth OPT $M_{||}\to M_{\perp}$ shown in the Figure 6.

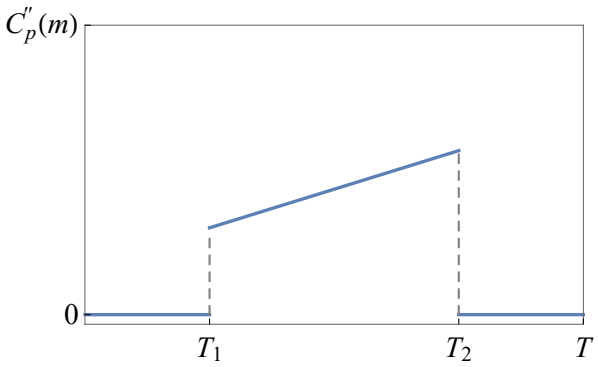

Figure 6

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2.3. Behavior Features of a Magnetic Susceptibility at an OPT

We write a TDP of an uniaxial or a rhombic ferromagnet ("plane" OPT) in an external magnetic field in the following form:

$$\Phi = K_1 \sin^2\theta + K_2 \sin^4\theta - \vec{m}\vec{H} - \frac{1}{2}\chi H^2. \tag{44}$$

At an analysis of the OPT $M_{||} \to M_{>}$ the $\sin\theta$, or the angle θ itself at the small rotations of $\vec{m}$, plays a role of an order parameter. Then we have

$$\Phi \simeq \Phi_0 + K_1\theta^2 + K_2\theta^4 - mH_{\perp}\theta + \frac{1}{2}m_0 H_{||}\theta^2 - \frac{1}{2}\chi H^2. \tag{45}$$

There is also a completely different behavior of the effective magnetic susceptibilities $\chi_{\perp}$ and $\chi_{||}$, corresponding to $H_{\perp}$ and $H_{||}$, i.e.

$$\begin{aligned} \chi_{\perp} =& \chi^0_{\perp} + \begin{cases} \frac{m_0^2}{2K_1}, & M_{||}\text{-phase} \\ -\frac{m_0^2}{4K_1}, & M_{>}\text{-phase} \end{cases} \\ \chi_{||} =& \chi^0_{||} + \begin{cases} 0, & M_{||}\text{-phase} \\ \frac{m_0^2}{8K_2}, & M_{>}\text{-phase} \end{cases} \end{aligned} \tag{46}$$

At an analysis of the OPT $M_{>} \to M_{\perp}$ the angle $\theta' = \pi/2 - \theta$ could play a role of an order parameter, so at $\theta \sim \pi/2$

$$\Phi \simeq \Phi_0 - (K_1 + 2K_2)\theta'^2 + K_2\theta'^4 - m_0 H_{||}\theta' + \frac{1}{2}m_0 H_{\perp}\theta'^2 - \frac{1}{2}\chi H^2. \tag{47}$$

Thus, at the $M_{>} \to M_{\perp}$ transition we have

$$\begin{aligned} \chi_{\perp} =& \chi^0_{\perp} + \begin{cases} \frac{m_0^2}{8K_2}, & M_{>}\text{-phase} \\ 0, & M_{\perp}\text{-phase} \end{cases} \\ \chi_{||} =& \chi^0_{||} + \begin{cases} \frac{m_0^2}{4(K_1+2K_2)}, & M_{>}\text{-phase} \\ -\frac{m_0^2}{2(K_1+2K_2)}, & M_{\perp}\text{-phase} \end{cases} \end{aligned} \tag{48}$$

Schematically the temperature behavior of the susceptibilities $\chi_{||}$ and $\chi_{\perp}$ at the smooth OPT $M_{||} \to M_{\perp}$ shown in the Figure 7. With the linear temperature dependence of $K_1(T)$ at the transition region and with $K_2 = \text{const}$ the susceptibility $\chi_{\perp}$ near the transition $M_{||} \to M_{>}$ and $\chi_{||}$ near the transition $M_{>} \to M_{\perp}$ have a characteristic for the Landau theory hyperbolic dependence:

$$\chi_{\perp} \sim |T - T_2|^{-1},\ \chi_{||} \sim |T - T_1|^{-1}. \tag{49}$$

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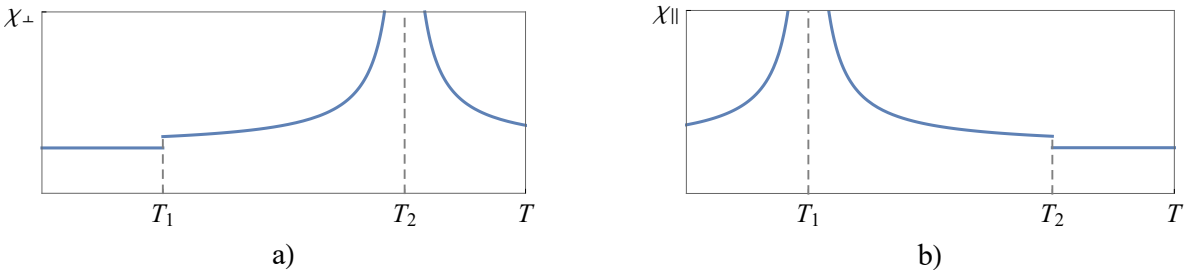

Figure 7

2.4. Spatial Spin-Reorientation in Rhombic Magnets

Without external magnetic fields a magnetic anisotropy energy of rhombic magnets with a consideration of second and fourth order anisotropy has the following form:

$$\Phi_{an} = K_1 \sin^2\theta\left(1 + \alpha \sin^2\varphi\right) + K_2 \sin^4\theta\left(1 + \beta \sin^2\varphi + \gamma \sin^4\varphi\right), \tag{50}$$

where θ, φ is the polar and azimuth orientation angles of a magnetization vector (or an antiferromagnetism vector). The constants K_1, K_2, α, β, γ can be connected with the first and the second anisotropy constants in the different crystallographic planes:

$$\begin{aligned} &K_1(ac) = K_1;\ K_1(bc) = K_1(1+\alpha);\ K_1(ab) = \alpha K_1 + \beta K_2 \\ &K_2(ac) = K_2;\ K_2(bc) = K_2(1+\beta+\gamma);\ K_2(ab) = \alpha K_2 + \gamma K_2. \end{aligned} \tag{51}$$

As it is accepted for perovskites $R\mathrm{FeO_3}$ and $R\mathrm{CrO_3}$, let us select the magnetic configurations Γ_1, Γ_2, Γ_4, Γ_{12}, Γ_{14}, Γ_{24}, Γ_{124}, which are different by the antiferromagnetism vector orientation: $\Gamma_1(\vec{l}||b)$, $\Gamma_2(\vec{l}||c)$, $\Gamma_4(\vec{l}||a)$ are the "clear" phases, $\Gamma_{12}(\vec{l} \perp a)$, $\Gamma_{14}(\vec{l} \perp c)$, $\Gamma_{24}(\vec{l} \perp b)$ are the angular phases and Γ_{124} is the phase with a spatial orientation of $\vec{l}$. An equilibrium orientation of the vector $\vec{l}$ can be found from the minimization of Φ_{an}. A phase diagram of the rhombic magnet in coordinates K_1 and $K_1' = \alpha K_1$ shown in the Figure 8 at a condition of the positive second anisotropy constants in all of the crystallographic planes. The stability strips width of the phases Γ_{24}, Γ_{12}, Γ_{14} are respectively $2K_2(ac)$, $\sqrt{2}K_2(bc)$, $2K_2(ab)$. The borderline between the phases Γ_4 and Γ_{14} defined by the equation $K_1' = -\beta K_2$ ($K_1(ab) = 0$), and between the Γ_2 and Γ_{12}-phases by the equation $K_1' = -K_1$ ($K_1(bc) = 0$).

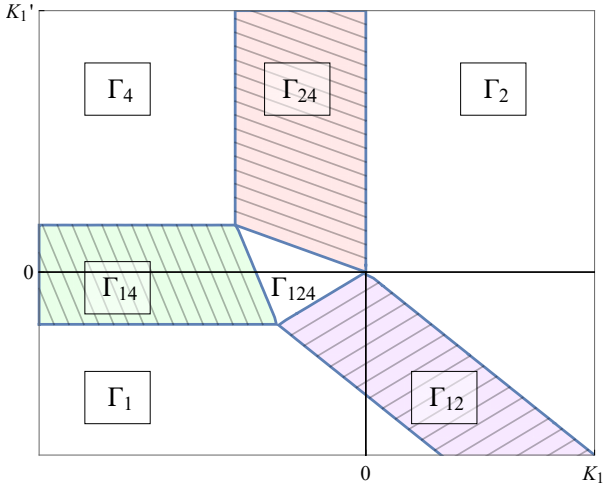

Figure 8

The triangle region in the center is the Γ_{124} configuration stability region and it realizes only if $4\gamma - \beta^2 > 0$. Actually, at $4\gamma = \beta^2$ the triangle region degenerate into a segment, which is occur to be a borderline between two angular phases on the one side and the third angular phase on the other side (e.g. Γ_{14}, Γ_{24}, Γ_{12}). From the form of the diagram follows that the most possible transitions

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with the spatial orientation of the antiferromagnetism vector are complex cascading transitions like $\Gamma_4 - \Gamma_{14} - \Gamma_{124} - \Gamma_{12} - \Gamma_1$, $\Gamma_2 - \Gamma_{24} - \Gamma_{124} - \Gamma_{14} - \Gamma_1$, and etc., i.e. those scenarios, in which the transition to the Γ_{124}-phase and out from it goes through a intermediate angular phase.

2.5. Spin-Reorientation Transitions in Cubic Magnets

Without an external magnetic field a TDP of a cubic magnet has the form

$$\Phi_{\text{an}} = K_1\left(\alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_1^2\alpha_3^2\right) + K_2\alpha_1^2\alpha_2^2\alpha_3^2 = \tag{52}$$

$$= \frac{1}{4}\left(K_1 \sin^2 2\theta + \left(K_1 + K_2\cos^2\theta\right)\sin^4\theta\cos^2 2\varphi\right),$$

where α_i are the direction cosines of the magnetic moment vector $\vec{M}$ (or the antiferromagnetism vector in the cubic antiferromagnet), θ, φ are the polar and azimuth angles of this vector orientation, K_1 and K_2 are the first and second anisotropy constants.

From minimizing (52) on θ and φ we find that minimum of the TDP realizes only when the vector $\vec{M}$ oriented along one of the three main crystallographic directions [100], [110] and [111]:

$$\begin{aligned} &\vec{M} \,||\, [100] \text{ at } K_1 \geq 0, \\ &\vec{M} \,||\, [110] \text{ at } 0 \geq K_1 \geq -\tfrac{1}{2}K_2, \\ &\vec{M} \,||\, [111] \text{ at } K_1 \leq -\tfrac{1}{3}K_2. \end{aligned} \tag{53}$$

The equalities in the equations (53) correspond to the lines of the phases stability loss and, as it seen from (53), there is regions of the K_1 and K_2 magnitudes at which there is coexistence of the different phases. Comparing the TDP values we could find lines of the phase transitions:

$$\begin{aligned} &\Phi_{[100]} = \Phi_{[110]} \text{ at } K_1 = 0,\ K_2 \geq 0, \\ &\Phi_{[110]} = \Phi_{[111]} \text{ at } 9K_1 + 4K_2 = 0,\ K_1 \leq 0, \\ &\Phi_{[111]} = \Phi_{[100]} \text{ at } 9K_1 + K_2 = 0,\ K_1 \geq 0. \end{aligned} \tag{54}$$

The phase transitions in the cubic magnet with considering only two anisotropy constants K_1 and K_2 occur sharply at the absence of external magnetic fields, i.e. the spin-reorientation is a first order phase transition.

2.6. SR Peculiarities in Magnets with a Fluctuating Magnetic Anisotropy

Compounds with a second order fluctuating magnetic anisotropy (i.e. many of disordered magnets, as well as compounds with substitution, containing ions with a different contribution to the magnetic anisotropy energy: $R_{1-x}R'_xFeO_3$, $RFe_{1-x}Co_xO_3$, ...) have an energy density

$$\varepsilon_{\text{an}} = K_1(\vec{r})\sin^2\theta(\vec{r}). \tag{55}$$

Characteristics of a random field of the local anisotropy constants $K_1(\vec{r})$ are the average value $\overline{K}_1$ and a correlation function of the fluctuations $\delta K_1(\vec{r}) = K_1(\vec{r}) - \overline{K}_1$:

$$K_{\text{an}}(\vec{r} - \vec{r}\,') = \frac{1}{4}\left\langle K_1(\vec{r})K_1(\vec{r}\,') - \vec{K}_1^2\right\rangle = \left\langle \delta K_1(\vec{r})\delta K_1(\vec{r}\,')\right\rangle. \tag{56}$$

A reasonable approximation for the $K_{\text{an}}(\vec{r} - \vec{r}\,')$ is

$$K_{\text{an}}(\vec{r} - \vec{r}\,') = D(K_1)\exp\left(-\frac{|\vec{r} - \vec{r}\,'|}{R_c}\right), \tag{57}$$

where $D(K_1)$ is the dispersion and R_c is the correlation radius of the anisotropy fluctuations.

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The fluctuations of the constant $K_1(\vec{r})$ lead to appearing of a peculiar stochastic magnetic structure, in which a local orientation of the magnetic moments fluctuate relative to its average value $\overline{\theta}$, which is defining the orientation of the average magnetic moment of the system. However, such magnetic structures, which correspond to the minimum of the energy $E = E_{\text{an}} + \frac{1}{2}\alpha \cdot (\vec{\nabla}\theta)^2$ (the second term is the exchange energy), are not energetically advantageous for every particular local value of $E(\vec{r})$. We can tell only about a tendency to the local orientation of $\theta(\vec{r})$, corresponding to the minimum of the local anisotropy energy, a consequence of which is the occurrence of the fluctuations $\delta\theta(\vec{r})$.

Neglecting exchange fluctuations and excluding $\delta\theta(\vec{r})$ from the thermodynamic potential, we can write the density of the TDP in the following form:

$$\Omega_{\text{eff}}(\overline{\theta}) = K_1 \sin^2\overline{\theta} + K_2 \sin^4\overline{\theta}, \tag{58}$$

where

$$K_1 = \overline{K}_1 - D(K_1)\frac{R_c^2}{2\alpha};\ K_2 = \overline{K}_2 + D(K_1)\frac{R_c^2}{2\alpha} \geq 0, \tag{59}$$

(α is the exchange parameter). Thus, taking into account orientation fluctuations of the parameter θ, defined by the fluctuation of the second order magnetic anisotropy constants, leads to the renormalization of the TDP and to the occurrence of the effective anisotropy constants of the higher orders.

Let us point out that at $\overline{K}_1 = 0$, $\overline{K}_2 = 0$ a minimum of the density Ω_{eff} realized at $\overline{\theta} = \pm\pi/4$. It can be shown that the value of $\overline{\theta} = \pm\pi/4$ also corresponds to a maximal dispersion of the local easy-axes directions or the values of $\theta(\vec{r})$. The appearance of a positive effective constant K_2 can lead to that the orientation transition between the phases $\theta = 0$ and $\theta = \pi/2$ at a partial system disordering will change it character from a sharp first-order transition to a smooth spin-reorientation transition, realized by two second-order phase transitions.

At $\overline{K}_2 > 0$ second-order anisotropy fluctuations lead to an increasing of the spin-reorientation interval.

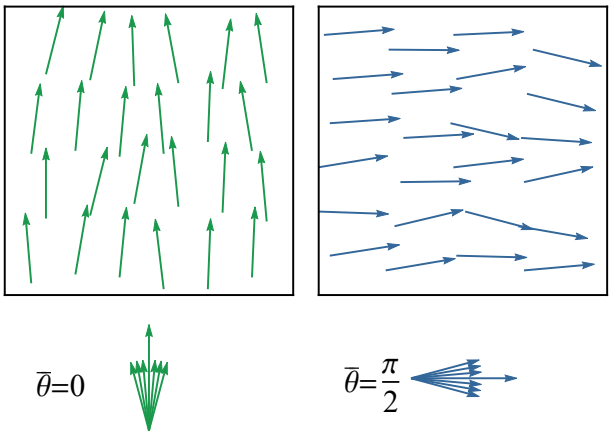

Figure 9. Schematic illustration of the orientational fluctuations of magnetic moments in the asperomagnetic phases.

The presence in the system of a competing magnetic anisotropy (in this case the local values of $K_1(\vec{r})$ can have different signs) affects on the SR transitions from the phases $\overline{\theta} = 0$, $\overline{\theta} = \pi/2$ to the angular phase $\overline{\theta} \neq 0, \pi/2$. The SR happens after the formation of asperomagnetic structures, in which the orientation of the average magnetic moment defines by the angles $\overline{\theta} = 0$ or $\overline{\theta} = \pi/2$, while in directions, perpendicular to the average magnetic moment, components of the local vectors $\vec{M}(\vec{r})$ orientate chaotic with a zero average value (Figure 9). In the angular phase complex heterophasic states can occur, in which the magnet breaks up to domains, i.e. the nuclei either of the phase $\overline{\theta} = 0$ or the phase $\overline{\theta} = \pi/2$.

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2.7. Dynamics of an Order Parameter at Spin-Reorientation Transitions. Soft Mode

For an analysis of homogeneous oscillations of a magnetic moment we use the Landau–Lifshitz equation

$$\frac{d\vec{M}}{dt} = -\gamma\left[\vec{M}\times\vec{H}\right] - \frac{\lambda}{M^2}\left[\vec{M}\times\left[\vec{M}\times\vec{H}\right]\right], \tag{60}$$

and in the spherical coordinates we have

$$\begin{aligned} -\frac{1}{\gamma}M\dot{\theta}\sin\theta &= \frac{\partial\Phi}{\partial\varphi} + \lambda\sin\theta\frac{\partial\Phi}{\partial\theta}, \\ \frac{1}{\gamma}M\dot{\varphi}\sin\theta &= \frac{\partial\Phi}{\partial\theta} - \frac{\lambda}{\sin\theta}\frac{\partial\Phi}{\partial\varphi}, \end{aligned} \tag{61}$$

where Φ is the TDP of the crystal, $\vec{M}$ is the magnetic moment, θ and φ are the polar and azimuth orientation angles of the vector $\vec{M}$, λ is the dimensionless damping parameter, γ is the gyromagnetic ratio. Let us represent the TDP in the form

$$\Phi = K_1\sin^2\theta + K_2\sin^4\theta + K_{ab}\sin^2\theta\sin^2\varphi, \tag{62}$$

including the anisotropy in the xy-plane ($K_{ab} > 0$, because in thermal equilibrium $\varphi = 0$). For small oscillations

$$\theta = \theta_0 + \delta\theta e^{-i\omega t},\ \varphi = \varphi_0 + \delta\varphi e^{-i\omega t}, \tag{63}$$

where $\varphi_0 = 0$, and θ_0 corresponds to the condition $\left.\frac{\partial\Phi}{\partial\theta}\right|_{\varphi=0} = 0$.

A linearization of the Landau–Lifshitz equations gives us in this case

$$\begin{aligned} \frac{i\omega}{\gamma}M\sin\theta_0\delta\theta &= \Phi_{\varphi\varphi}\delta\varphi + \lambda\sin\theta_0\Phi_{\theta\theta}\delta\theta, \\ -\frac{i\omega}{\gamma}M\sin\theta_0\delta\varphi &= \Phi_{\theta\theta}\delta\varphi - \frac{\lambda}{\sin\theta_0}\Phi_{\varphi\varphi}\delta\varphi, \end{aligned} \tag{64}$$

where $\Phi_{\varphi\varphi}$, $\Phi_{\theta\theta}$ are the second derivatives of Φ on φ and θ. A non-trivial solution of this system exists only for the complex frequency ω:

$$\omega = \omega_p + i\Delta\omega, \tag{65}$$

where in the low damping approximation

$$\omega_p = \frac{\gamma}{M}\left(\frac{\Phi_{\theta\theta}\Phi_{\varphi\varphi}}{\sin^2\theta_0}\right)^{1/2},\ \Delta\omega = \frac{\lambda\gamma}{M}\left(\Phi_{\theta\theta} + \frac{1}{\sin^2\theta_0}\Phi_{\varphi\varphi}\right), \tag{66}$$

where $\sin^{-2}\theta_0\Phi_{\varphi\varphi} = 2K_{ab}$ has no singularities at the transition points T_1 and T_2, so the critical behavior of the oscillation frequency determined by only $\Phi_{\theta\theta}$, which is turning to zero at $T = T_1$ and $T = T_2$.

Near the temperatures T_1 and T_2 the frequency ω_p could be rewritten as

$$\omega_p^2 = \omega_1\omega_2\frac{|(T-T_1)(T-T_2)|}{(T_2-T_1)^2};\ \omega_1 = \gamma\frac{2K_{ab}}{M};\ \omega_2 = \gamma\frac{2K_2}{M}. \tag{67}$$

Schematically the frequency $\omega_p(T)$ shown in the Figure 10. That dependence is the frequency of a homogeneous precession of the magnetic moment (FMR frequency for ferromagnets or AFMR frequency for antiferromagnets). It is the limit at $k \to 0$ for the corresponding spin wave branch, which in this case called the soft mode; it means that the frequency ω_p turn zero at T_1 and T_2 (a "gap" disappearance in the spectrum of the corresponding oscillations).

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At a reorientation in an uniaxial magnet ($K_{ab} = 0$) the eigenfrequency of the order parameter, when approaching the transition temperature from the high-symmetry phase, goes to zero and keeps equal to zero at the low-symmetry phase (Figure 11). This soft mode called a "Goldstone mode".

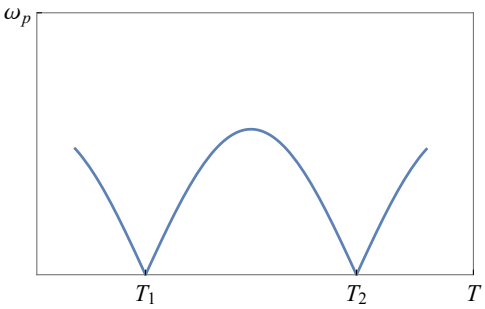

Figure 10

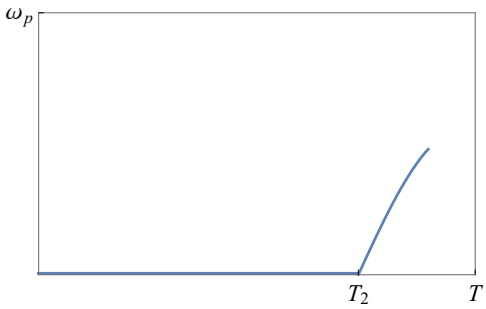

Figure 11

3. Orientational Phase Transitions Induced by External Magnetic Fields

3.1. Uniaxial Ferromagnets

Consider an uniaxial ferromagnet with an easy-axis in parallel to the z-axis in an external magnetic field in a basal plane. A thermodynamic potential in this case has the following form

$$\Phi = K_1 \sin^2\theta + K_2 \sin^4\theta - m_0 H \sin\theta, \ (K_1 > 0), \tag{68}$$

where m_0 is the saturation magnetic moment. Equilibrium directions $\vec{m}$ can be determined from the condition

$$\frac{\partial \Phi}{\partial \theta} = \sin 2\theta \left(K_1 + 2K_2 \sin^2\theta\right) - m_0 H \cos\theta = 0, \tag{69}$$

which gives us two phases:

a) the "high-field" phase: $\theta = \pi/2$, $\cos\theta = 0$;
b) the angular phase: $\theta \neq \pi/2$, $\sin\theta\left(K_1 + 2K_2 \sin^2\theta\right) - m_0 H = 0$.

The last equation determine the magnetization curve in the angular phase if we consider $\sin\theta = m_\perp / m_0 = \mu$:

$$H = \frac{1}{m_0}\left(2K_1\mu + 4K_2\mu^3\right). \tag{70}$$

From the condition $\frac{\partial^2 \Phi}{\partial \theta^2} > 0$ at $\cos\theta = 0$ we can find the high-field phase stability region:

$$H \geq \frac{2K_1 + 4K_2}{m_0} = H_1 \tag{71}$$

(the equality corresponds to the magnetic field at which the phase loses its stability). The stability region of the angular phase can be easily determined if we consider Φ in the form

$$\Phi = K_1\mu^2 + K_2\mu^4 - m_0 H\mu. \tag{72}$$

In this case

$$\frac{\partial \Phi}{\partial \mu} = 2K_1\mu + 4K_2\mu^3 - m_0 H = 0 \tag{73}$$

and

$$\frac{\partial^2 \Phi}{\partial \mu^2} = 2K_1 + 12K_2\mu^2 > 0. \tag{74}$$

At $K_2 \geq 0$ the angular phase stable for any μ: $0 \leq \mu \leq 1$, so the orientational phase transition from the angular phase into the high-field phase will always be a second-order phase transition

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with the critical field H_1 (Figure 12), at which the phase stability loss occurs and the values of the thermodynamic potentials equalize. At $K_2 > 0$ the angular phase loses stability at

$$\mu^2 \geq -\frac{K_1}{6K_2}, \tag{75}$$

which corresponds to the value of the magnetic field

$$H \geq \frac{4}{3}\frac{K_1}{m_0}\sqrt{-\frac{K_1}{6K_2}} = H_2 \geq H_1. \tag{76}$$

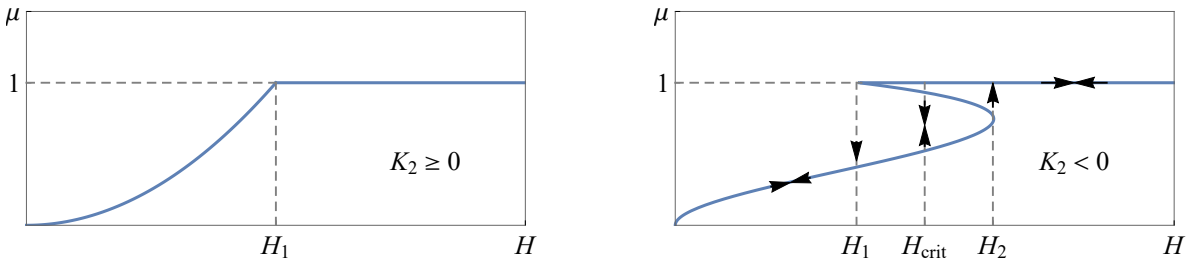

Figure 12

The field H_2 of the stability loss of the angular phase corresponds to a point on the magnetization curve, in which

$$\partial H/\partial \mu = 0.$$

The TDP of the angular and high-field phases match up at $H = H_{\text{crit}}$, which can be determined by the equation

$$K_1\mu^2 + K_2\mu^4 - m_0\mu H_{\text{crit}} = K_1 + K_2 - m_0 H_{\text{crit}},$$

at

$$H_{\text{crit}} = \frac{1}{m_0}\left(2K_1\mu + 4K_2\mu^3\right). \tag{77}$$

The solution of these equations gives us

$$H_{\text{crit}} = \frac{2K_2}{27m_0}\left(9\frac{K_1}{K_2} + 10 + \left(2 + 3\frac{K_1}{K_2}\right)\sqrt{-2 - 3\frac{K_1}{K_2}}\right), \quad H_1 \leq H_{\text{crit}} \leq H_2. \tag{78}$$

Thus, at $K_2 > 0$ the transition from the angular phase into the high-field phase is a first-order phase transition.

Two different options of magnetization curves in a hard direction of the uniaxial ferromagnet shown in the Figure 12.

There is regions at $K_2 > 0$ in the magnetization curve corresponding to metastable states, which leads to hysteresis in single-domain samples. In multi-domain samples a hysteresis-free option of the first-order transition is possible.

All of the results above can be used for the analysis of a transition from the easy-plane phase $\vec{m} \perp z$ into the easy-axis phase $\vec{m}||z$ at $\vec{H}||z$. For this analysis the replacement $K_1 \to -K_1 - 2K_2$ is needed in all of the expressions for the critical fields.

3.2. Anisotropic Antiferromagnets. "Spin-Flop" Transitions

An interaction energy of an antiferromagnet with an external magnetic field can be determined in the following form:

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$$\Phi_H = -\frac{1}{2}\chi_\perp H^2 + \frac{1}{2}\left(\chi_\perp - \chi_{||}\right)\left(\vec{l}\cdot\vec{H}\right)^2, \tag{79}$$

where $\vec{l}$ is the normalized antiferromagnetism vector ($2M_0\vec{l} = \vec{M}_1 - \vec{M}_2$ where $\vec{M}_{1,2}$ are the magnetization of the sublattices, $M_0 = |\vec{M}_1| = |\vec{M}_2|$); $\chi_\perp$ is the perpendicular susceptibility (susceptibility at $\vec{H} \perp \vec{l}$), $\chi_{||}$ is the parallel susceptibility (susceptibility at $\vec{H}||\vec{l}$).

A characteristic temperature dependence of $\chi_\perp$ and $\chi_{||}$ for weakly anisotropic antiferromagnets like MnF_2 shown in the Figure 13.

In the transverse phase ($\vec{l} \perp \vec{H}$) of the antiferromagnet the Zeeman energy is less by $\frac{1}{2}\left(\chi_\perp \cdot \chi_{||}\right)H^2$ than in the longitudinal phase ($\vec{l}||\vec{H}$).

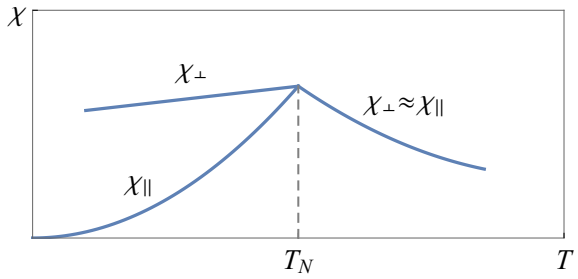

Figure 13

Let us consider a rhombic anisotropic antiferromagnet in external magnetic fields oriented along one of the crystallographic axes, e.g. z-axis. A TDP, which is describing rotation processes of $\vec{l}$ in one of the planes, will has the following form:

$$\begin{aligned}\Phi &= K_1\sin^2\theta + K_2\sin^4\theta - \frac{1}{2}\chi_\perp H^2 + \frac{1}{2}\left(\chi_\perp - \chi_{||}\right)H^2\cos^2\theta \\ &= \left[K_1 - \frac{1}{2}\left(\chi_\perp - \chi_{||}\right)H^2\right]\sin^2\theta + K_2\sin^4\theta - \frac{1}{2}\chi_{||}H^2,\end{aligned} \tag{80}$$

where θ is the angle between the z-axis and the vector $\vec{l}$.

Obviously, that taking into account the external magnetic field in this case leads to a renormalization of the first anisotropy constant. Thus, the analysis of the antiferromagnet behavior in the field can be based on the Section 2.1, in particular the phase diagram on the Figure 2.

At $K_2 > 0$ and the initial phase $\vec{l}||z$ the inclusion of the external magnetic field $\vec{H}||z$ leads to a "spin-flop" transition from the longitudinal phase $\vec{l}||\vec{H}$ into the transverse phase $\vec{l} \perp \vec{H}$, which is starting at

$$H_{||} = \sqrt{\frac{2K_1}{\chi_\perp - \chi_{||}}}, \tag{81}$$

as a second order phase transition (the transition into the angular phase), and ending at

$$H_\perp = \sqrt{\frac{2K_1 + 4K_2}{\chi_\perp - \chi_{||}}} \tag{82}$$

also, as a second order phase transition (the transition from the angular phase into transverse phase).

At $K_2 \le 0$ and $K_1 > 2|K_2|$ the initial longitudinal phase $\vec{l}||\vec{H}$ loses the stability at $H_{||}$, and the transverse phase became stable at $H = H_\perp$ ($H_\perp < H_{||}$). Thus, in the region of fields $H_\perp < H < H_{||}$ the longitudinal and transverse phases can coexist. Equality of the thermodynamic potentials of both phases takes place at $H = H_{\text{crit}} = \sqrt{\frac{2K_1+2K_2}{\chi_\perp - \chi_{||}}}$, and $H_\perp < H_{\text{crit}} < H_{||}$.

In an external magnetic field, which is exceeded the maximum of the critical fields $H_\perp$, $H_{||}$, H_{crit}, gradual "contraction" of the magnetic moments of the sublattices 1 and 2 to the direction of the

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magnetic field H occurs. So, in the sufficiently large magnetic field ($H \approx H_E$ for the weakly anisotropic antiferromagnets, $H_E = \frac{2m_0}{\chi_\perp}$ is the exchange field) a ferromagnetic structure $\vec{M}_1 || \vec{M}_2 || H$ is realized.

Schematically a magnetization curve of the antiferromagnet in our case shown in the Figure 14. The magnetic susceptibility on the region $0 \leq H \leq H_{||}$ (Figure 14a) or $0 \leq H \leq H_\perp$ (Figure 14b) matches with $\chi_{||}$, and on the region $H_{||} \leq H \leq H_E$ it matches with $\chi_\perp$. On the region $H_{||} \leq H \leq H_\perp$ (Figure 14a) the susceptibility has a "rotational" character:

$$\chi_{\text{rot}} = \frac{\partial m}{\partial H} = \chi_\perp - \left(\chi_\perp - \chi_{||}\right)\left(\cos^2\theta - \frac{2H^2}{H_\perp^2 - H_{||}^2}\right), \tag{83}$$

where we used the following relations:

$$m = \chi_\perp H - \left(\chi_\perp - \chi_{||}\right)\cos^2\theta \cdot H, \tag{84}$$

$$\frac{\partial\theta}{\partial H} = \frac{2H}{\left(H_\perp^2 - H_{||}^2\right)\sin 2\theta}. \tag{85}$$

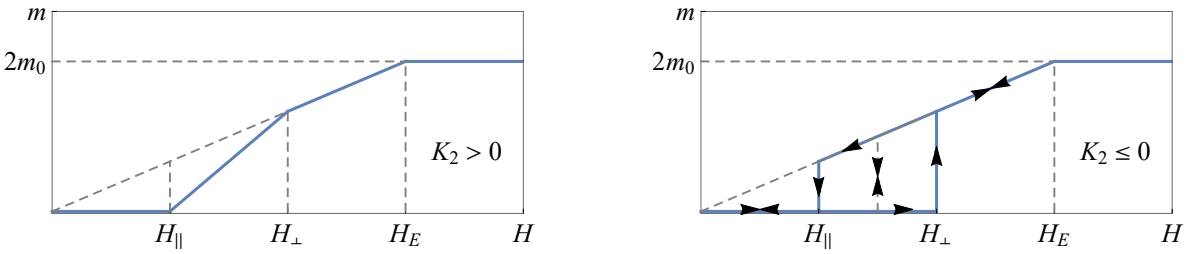

Figure 14

Let us pay attention to that $\partial\theta/\partial H \to \infty$ at $H \to H_{||}$ from the right and at $H \to H_\perp$ from the left. At $\chi_\perp \gg \chi_{||}$ (low temperature) and $\left(H_\perp - H_{||}\right) \ll H_{||}$ ($K_2 \ll K_1$) the rotational susceptibility

$$\chi_{\text{rot}} \approx \frac{H_{||}}{H_\perp - H_{||}}\chi_\perp \tag{86}$$

can significantly exceed $\chi_\perp$.

3.3. Metamagnetic Transitions in High-Anisotropy Magnets

Considering the "spin-flop" transitions we implicitly assumed that a magnetic anisotropy energy is small in comparison with an exchange energy. There is, actually, a wide enough class of high-anisotropy magnets, which has the anisotropy energy comparable with the exchange energy, or even exceeds it. For a research of the peculiarities of orientational transitions induced by a magnetic field in such systems we restrict ourselves with $K_2 = 0$ case, and also with the region of sufficiently low temperatures, at which $\chi_{||} \approx 0$ in antiferromagnets.

A model thermodynamic potential for the analysis of the orientational phase transition in the certain plane can be determined in the following form:

$$\begin{aligned}\Phi = {} & K_1\left(\sin^2\theta_1 + \sin^2\theta_2\right) + IM_0^2\cos(\theta_1 - \theta_2) \\ & - M_0H(\cos(\theta_H - \theta_1) + \cos(\theta_H - \theta_2)),\end{aligned} \tag{87}$$

where I is the exchange constant, $\theta_{1,2}$ are the orientation angle of sublattices magnetic moments, θ_H is the orientation angle of the external magnetic field $\vec{H}$.

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A minimization of the TDP in the magnetic field H, which is directed along the anisotropy axis ($\theta_H = 0$), gives us three equilibrium phases:

1. $\theta_1 = 0;\ \theta_2 = \pi$ (antiferromagnet phase),
2. $\theta_1 = -\theta_2$ (phase with the flipped sublattices),
3. $\theta_1 = \theta_2 = 0$ (ferromagnet phase).

A research of the stability of this structures shows that the antiparallel ordering of the magnetic moments $\theta_1 = 0$, $\theta_2 = \pi$ exist in the region

$$0 < H < \sqrt{(2H_E + H_A)H_A} = H_c, \tag{88}$$

where $H_A = 2K_1/M_0$, $H_E = IM_0$ are the anisotropy and the exchange field respectively. The second state $\theta_1 = -\theta_2$ with the flipped lattices is stable in the region

$$H_c' = \sqrt{\frac{(2H_E - H_A)^2 H_A}{2H_E + H_A}} \le H \le H_c. \tag{89}$$

The magnetic fields H_c' and H_c are the analogues of $H_\perp$ and $H_{||}$ for the weakly anisotropic antiferromagnet (Figure 14b). The existing of overlapping phases witness about a possibility of hysteresis.

The state with the flipped sublattices at the increasing of the magnetic field smoothly transit into a "ferromagnet" state $\theta_1 = \theta_2 = 0$ in the magnetic field $H_{SF} = 2H_E - H_A$.

With the increasing of the anisotropy constant, or the anisotropy field H_A, the magnitude of the magnetic field H_{SF} decreases, but the magnitude of the magnetic field H_c increases. With this the stability regions of the antiferromagnet phase ($\theta_1 = 0$, $\theta_2 = \pi$) and ferromagnet phase ($\theta_1 = \theta_2 = 0$) expand, and simultaneously the stability region of the angular phase ($\theta_1 = -\theta_2$) narrows. Physically this fact corresponds to the minimum of the anisotropy energy in the ferro- and antiferromagnet phases, unlike the angular phase.

At $H_A = \frac{2}{3}H_E$ the upper border of the antiferromagnetic phase stability is compared with the lower border of the ferromagnetic phase stability H_{SF}. At H_{SF} energies of the three phases

$$\Phi_{AFM} = -M_0 H_E;\ \Phi_{FM} = -M_0(2H - H_E);$$
$$\Phi_{FL} = M_0\left(H_A - H_E - \frac{H_2}{2H_E - H_A}\right) \tag{90}$$

become equal and, with a further increase in H_A, the phase with flipped sublattices becomes energetically unfavorable for any H. Magnets for which the condition $H_A \ge H_E$ is valid are called metamagnets. Because of this condition, in metamagnets there is no phenomenon of flipping magnetic sublattices.

Figure 15 shows the phase diagram of the system described by the TDP (87) in coordinates $\alpha = H_A/H_E$, $\alpha_H = H/H_E$. It can be seen from the figure that at $\alpha \lesssim 1$, i.e. at $H_A \lesssim H_E$, there is always the phase with flipped sublattices FL (between the phase lines 1 and 3). The transition from the antiferromagnetic phases (AFM) to the FL-phase can be delayed, this phenomenon analogous to overheating during a first-order phase transition such as melting. The reverse transition FL–AFM can also be delayed (this is shown in Figure 15 by the lines 2 and 4). At $\alpha > 1$, the FL-phase is absent. With an increase in the external field at $\alpha \ge 1$, the AFM-phase transforms into the FM-phase, i.e. into a ferromagnet induced by the field. The region $\alpha > 1$ of the phase diagram is the region of the metamagnetic state existence.

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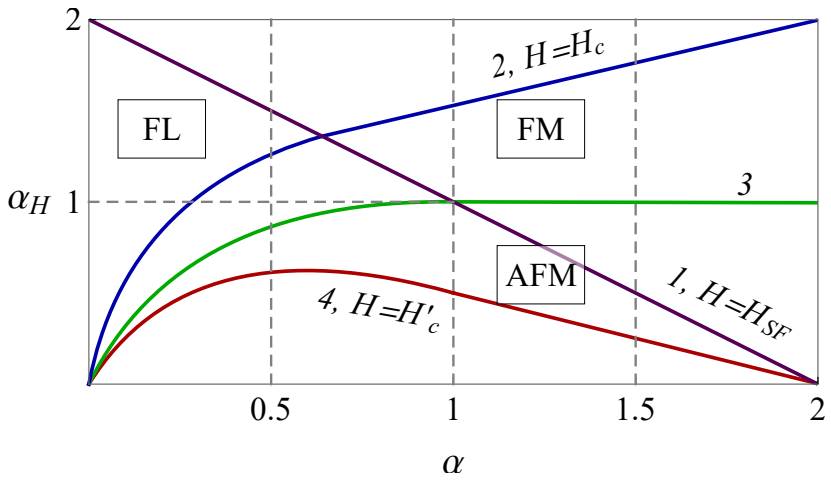

Figure 15

The transition AFM–FM in an external field is called the "spin-flip" transition. A typical magnetization curve for metamagnets is shown in Figure 16. We note that a decrease in the field to zero after the spin-flip transition can not be accompanied by a return of the system to the AFM state (this requires $H_A > 2H_E$). Thus, after turning off the field, the metamagnet can remain in the FM state, it will have remanent magnetization. If the value of $|H_A - 2H_E|$, which plays the role of the coercive force in this case, is sufficiently large, then such a metamagnet can be used as the material for permanent magnets.

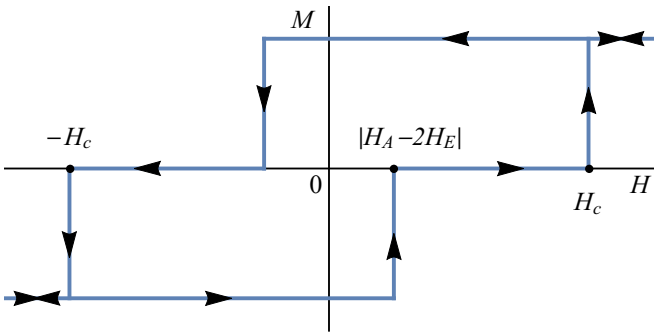

Figure 16

4. Elastic and Magnetoelastic Properties of Rhombic Magnets at SR Transitions

4.1. Magnetostriction at SR Transitions

Consider a rhombic magnet with a SR transition in one of the crystallographic planes (e.g. *ac*-plane). Along with Φ_{an} from (27), the TDP of the crystal will include the elastic and magnetoelastic energies:

$$\Phi_{\text{el}} = \tfrac{1}{2}c_{11}\varepsilon_{xx}^2 + \tfrac{1}{2}c_{22}\varepsilon_{yy}^2 + \tfrac{1}{2}c_{33}\varepsilon_{zz}^2 + c_{12}\varepsilon_{xx}\varepsilon_{yy} + c_{13}\varepsilon_{xx}\varepsilon_{zz} + c_{23}\varepsilon_{yy}\varepsilon_{zz} + 2c_{44}\varepsilon_{yz}^2 + 2c_{55}\varepsilon_{xz}^2 + 2c_{66}\varepsilon_{xy}^2\,, \tag{91}$$

$$\Phi_{\text{m-el}} = \left(L_a\varepsilon_{xx} + L_b\varepsilon_{yy} + L_c\varepsilon_{zz}\right)\sin^2\theta + \tfrac{1}{2}\mu_2\varepsilon_{xz}\sin 2\theta\,; \tag{92}$$

moreover, the form of $\Phi_{\text{m-el}}$ is determined by choosing the SR in the *ac*-plane.

Minimizing the TDP by deformations ε_{ij} in the absence of external mechanical stresses, we obtain:

$$\varepsilon_{ii} = -\frac{L_i}{c_{ii}}\sin^2\theta\,,\ \varepsilon_{xz} = -\frac{\mu_2}{8c_{55}}\sin 2\theta\,,\ \varepsilon_{yz} = \varepsilon_{xy} = 0\,.$$

In this way, at a smooth SR transition $M_z \to M_x$, along with the usual magnetostrictive deformations $\varepsilon_{xx,yy,zz}$ that do not change the rhombic symmetry of the crystal, shear (monoclinic) deformations

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appear in the plane of magnetic moments rotation, reaching a maximum at $\theta = \pm 45^\circ, \pm 135^\circ$. The ordinary deformations ε_{ii} are even, and the monoclinic deformation ε_{xz} is the odd function of the order parameter θ. Therefore, if the magnetostrictive deformations are insensitive to the domain structure, then the monoclinic deformation differs in sign in domains with different signs of the order parameter (Figure 17).

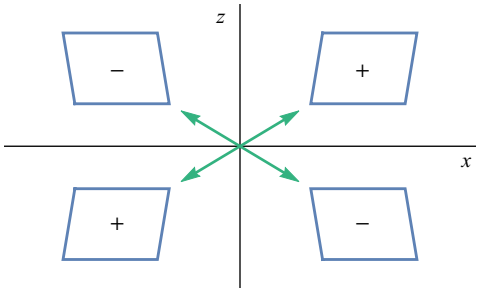

Figure 17

During the SR transition $M_z \to M_x$ induced by an external magnetic field $\vec{H}||x$, the nature of the magnetostrictive deformations in the rhombic ferromagnet will have the form shown schematically in Figure 18.

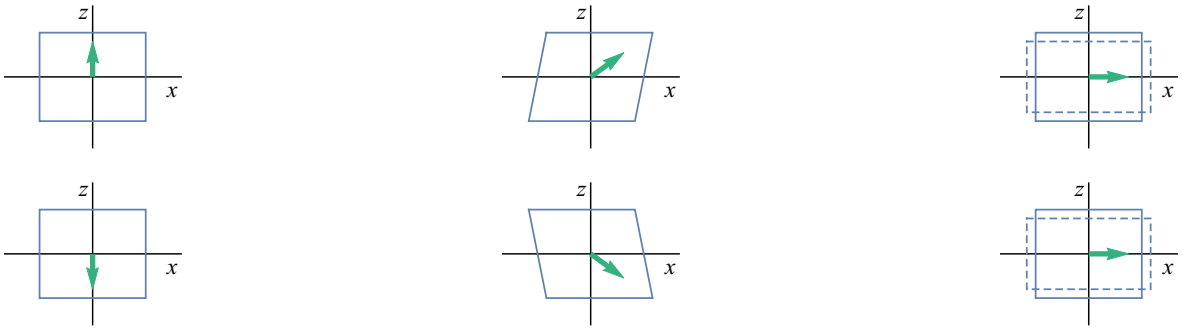

Figure 18

It is clear that the sample remagnetization by domain boundary offsets in the angular phase M_{zx}, for which small external magnetic fields are sufficient, will be accompanied by a change in the sign of the monoclinic deformation.

4.2. OPT Induced by External Mechanical Stresses

Below we consider a rhombic magnet with magnetic anisotropy of the form

$$\Phi_{\text{an}} = K_1 \sin^2\theta + K_2 \sin^4\theta\,, \tag{93}$$

where θ is the orientation angle of a magnetic moment (or antiferromagnetism) vector in one of the crystallographic planes (e.g. *ac*-plane).

The action of external mechanical stresses (pressure) leads to elastic deformations of the sample. Under uniaxial compression along the *c*-axis, the components of the elastic deformation tensor in the rhombic crystal is determined according to

$$\varepsilon_{ij} = -pS_{ijzz}\delta_{ij}\,, \tag{94}$$

where S_{ijkl} is the elastic compliance tensor. Under bulk compression of the rhombic crystal, we have

$$\varepsilon_{ii} = -p\sum_j S_{iijj}\,. \tag{95}$$

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If pressure is applied in the ac-plane at an angle of α to the a or c-axis, then it also causes the appearance of shear deformations. At $\alpha = 45^\circ$

$$\varepsilon_{ii} = -\tfrac{1}{2}p\left(S_{ii33} + S_{ii11}\right), \; \varepsilon_{ac} = \tfrac{1}{2}pS_{1313}\,. \tag{96}$$

When analyzing the OPT in the ac-plane, we represent the magnetoelastic energy in the form (92). It is clear that bulk compression, or compression along the crystallographic axes a, b, c of the rhombic crystal does not change the lattice symmetry and, due to the magnetoelastic coupling (92), it leads only to the change in the anisotropy constant K_1:

$$\Delta K_1 = L_a\varepsilon_{aa} + L_b\varepsilon_{bb} + L_c\varepsilon_{cc} = \begin{cases} -p\sum_j L_i S_{ii33} & (c\text{-axis compression}), \\ -p\sum_{ij} L_i S_{iijj} & (\text{bulk compression}). \end{cases} \tag{97}$$

If we assume that K_2, $L_{a,b,c}$ do not depend on temperature and K_1 depends linearly on T according to the relation (35), then the effect of pressure in this case will be reduced only to the shift in the beginning and end of the SR without changing the SR interval:

$$\frac{\partial T_1}{\partial p} = \frac{\partial T_2}{\partial p} = -\frac{(T_2 - T_1)}{2K_2}\frac{\partial K_1}{\partial p}\,, \quad \frac{\partial (T_2 - T_1)}{\partial p} = 0\,. \tag{98}$$

For the values of the parameters that are characteristic of a number of orthoferrites $R\mathrm{FeO_3}$: $(T_2 - T_1) \sim 10$ K, $K_2 \sim 5 \cdot 10^4$ erg $\cdot$ cm^{-3}, $\frac{\partial K_1}{\partial p} \sim 10^5$ erg $\cdot$ cm$^{-3}\cdot$ kbar^{-1}, we have:

$$\frac{\partial T_{1,2}}{\partial p} \sim 10\,\frac{\mathrm{K}}{\mathrm{kbar}}\,. \tag{99}$$

The presence of the shear component of mechanical stresses leads to a change in the rhombic symmetry of the crystal and an OPT induction in the ac-plane even at an arbitrarily small value of ε_{ac}. The equilibrium value of the angle θ is determined in this case by the equation

$$\tan 2\theta = -\frac{\mu_2\,\varepsilon_{ac}}{K_1 + \Delta K_1 + 2K_2\sin^2\theta} \tag{100}$$

at $\cos 2\theta \neq 0$. For the "magnetoelastic" susceptibility, we have

$$\left.\frac{\partial\theta}{\partial\varepsilon_{ac}}\right|_0 = \begin{cases} -\dfrac{\mu_2}{2K_1}\,, \; \theta = 0; \\ -\dfrac{\mu_2}{2(K_1 + 2K_2)}\,, \; \theta = \frac{\pi}{2}; \\ -\dfrac{\mu_2\cos 2\theta}{2K_2\sin^2 2\theta}\,, \; 0 < \theta < \frac{\pi}{2} \end{cases} \tag{101}$$

(where the equation $\partial\Phi/\partial\theta = 0$ is used). Thus, the magnetoelastic susceptibility $\left.\frac{\partial\theta}{\partial\varepsilon_{ac}}\right|_0$ has a singularity at the points T_1 and T_2, i.e. the beginning and end of the smooth SR transition $\theta = 0 \to \theta = \pi/2$ (Figure 19).

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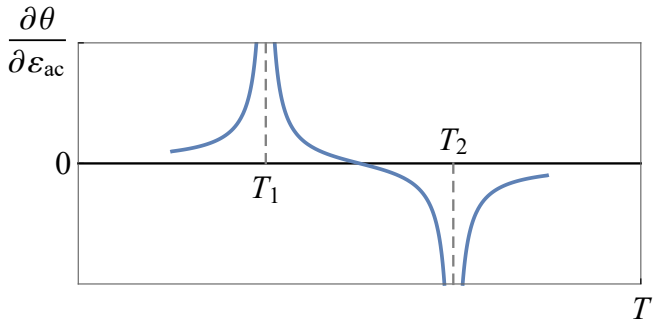

Figure 19

At a linear dependency of $K_1(T)$ this singularity is a characteristic of the Landau theory. Close to T_1 or T_2, we have $\frac{\partial\theta}{\partial\varepsilon_{ac}}\Big|_0 \sim (T - T_{1,2})^{-1}$.

Note that for large shear deformations ε_{ac} and small values of $K_{1.2}$, the equilibrium value of angle θ will approach $\pm\pi/2$. The sign of θ will then be determined by the signs of μ and ε_{ac}.

4.3. Behavior of Elastic Modules at SR Transitions

The components of an elastic compliance tensor $S_{\mu\nu}$ play a role of elastic susceptibilities and have singularities in SR transition regions.

For a rhombic crystal with the spontaneous SR transition in the *ac*-plane, the elastic modules $S_{11,22,23}$ and $S_{12,13,23}$ will change sharply at the PT points $M_z \to M_{zx}$ and $M_{zx} \to M_x$, since the associated with ε_{ii} and mechanical stresses σ_{ii} term in $\Phi_{\text{m-el}}$ is quadratic in the order parameter θ or $\theta' = \pi/2 - \theta$. The elastic modulus S_{55} will have a hyperbolic type divergence at these points, since the associated with ε_{ac} and σ_{ac} ($\varepsilon_{ac} = S_{55}\sigma_{ac}$) term in $\Phi_{\text{m-el}}$ is linear in the order parameter. The modules S_{44}, S_{66} do not have singularities in the SR transition region of the *ac*-plane.

Of practical interest is an analysis of the behavior of constants $C_{\mu\nu}$ (inverse elastic susceptibilities) during the SR transitions, which are easily determined experimentally in terms of sound propagation velocities. For example, for C_{55} at the SR transition $M_z \to M_x$ in the *ac*-plane, we have (Figure 19)

$$C_{55} = C_{55}^{(0)} + \begin{cases} -\dfrac{\mu_2^2}{4K_1}, & M_z\text{-phase;} \\ +\dfrac{\mu_2^2}{8K_1}, & M_{xz}\text{-phase, } \theta \geq 0; \\ -\dfrac{\mu_2^2}{8(K_1+2K_2)}, & M_{xz}\text{-phase, } \theta \leq \frac{\pi}{2}; \\ +\dfrac{\mu_2^2}{4(K_1+2K_2)}, & M_x\text{-phase.} \end{cases} \tag{102}$$

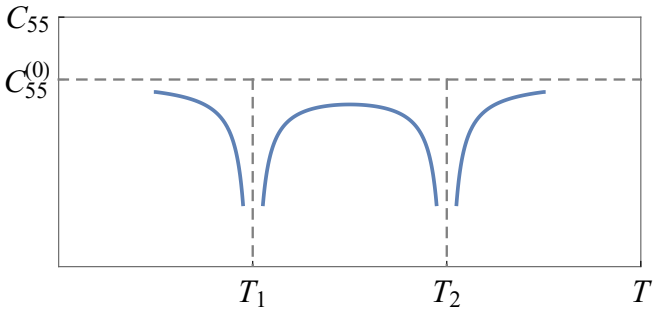

Figure 20

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A sharp decrease in the shear elastic constant C_{55} at the points T_1 and T_2 of the SR transitions $M_{zx} \to M_x$ and $M_z \to M_{zx}$ leads to a corresponding sharp drop in the transverse sound velocity

$$v_{xz}^T = \sqrt{C_{55}/p} \tag{103}$$

and an anomalous "softening" of the lattice with respect to shear mechanical stresses in the plane of a magnetic moments rotation.

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Conflicts of Interest: The authors declare no conflicts of interest.

Abbreviations

The following abbreviations are used in this manuscript:

PT	Phase transition
TDP	Thermodynamic potential
SR	Spin reorientation
FM	Ferromagnetic
AFM	Antiferromagnetic
FL	Flipped sublattices

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