ISSN 0021-3640, JETP Letters, 2015, Vol. 102, No. 2, pp. 100–104. © Pleiades Publishing, Inc., 2015.
Original Russian Text © S.N. Martynov, 2015, published in Pis'ma v Zhurnal Eksperimental'noi i Teoreticheckoi Fiziki, 2015, Vol. 102, No. 2, pp. 111–116.

Symmetrical Conical Incommensurate Structures of a Frustrated Isotropic Heisenberg Ferrimagnet

S. N. Martynov

Kirensky Institute of Physics, Siberian Branch, Russian Academy of Sciences, Krasnoyarsk, 660036 Russia

e-mail: unonav@iph.krasn.ru

Received May 22, 2015; in final form, June 11, 2015

The conditions of the appearance of conical incommensurate structures have been studied analytically for a ferrimagnet with the geometrically frustrated exchange between spins in different magnetic positions (subsystems) and the competition between exchanges in one of the subsystems. The phase transition temperatures to the conical states have been determined. The types of phase transitions have been determined by the numerical minimization of the free energy and (temperature–exchange parameters) phase diagrams have been obtained.

DOI: 10.1134/S0021364015140076

Interest in studying incommensurate magnetic structures in the last decade is mainly due to magnetoelectric effects accompanying this type of magnetic ordering [1–3]. The possibility of controlling the direction of the electric polarization by a low magnetic field at room temperature is of a great applied interest. Ferrimagnets with the conical type of incommensurate ordering are multiferroics with the corresponding magnetoelectric characteristics [2–6]. The necessary condition of controlling the direction of the ferrimagnetic moment by low magnetic fields is the small magnitude of the magnetic anisotropy. The formation of the conical ordering in such isotropic (or nearly isotropic) magnets is a particular case of non-collinearity caused by the frustration (competition) of isotropic exchanges. A specific type of the non-collinear magnetic order is determined first of all by the spatial distribution of exchange bonds in a crystal and the symmetry of the location of magnetic ions in the considered magnetic structure. Cubic spinel AB_2O_4 is the only example of the conical incommensurate ordering studied in detail theoretically [7, 8]. The three-cone incommensurate structure in it is determined by three positions of magnetic ions A, B_1, and B_2 nonequivalent with respect to the direction of the modulation vector of the magnetic structure **k** [1, 1, 0] [3, 8, 9]. In this work, we theoretically consider conical incommensurate structures arising in the isotropic Heisenberg model, where magnetic ions are in two nonequivalent positions both crystallographically and with respect to the arising magnetic structures. The number of independent variables of the problem is determined by the spin dimensionality and the number of nonequivalent positions (magnetic subsystems) and allows a relatively simple analytical solution on magnetic structure parameters in the considered two-subsystem case. In addition, there is a series of solutions with the coplanar orientation of spins within such a model [10–12]. Correspondingly, the conditions of the existence of conical solutions, i.e., exchange interactions and temperature at which their appearance is possible, as well as the types of phase transitions between states, can be determined by comparing free energies of different states.

Two types of frustrated exchanges are taken into account in the model: the geometrically frustrated exchange between spins in different subsystems and competing exchanges between nearest and next nearest magnetic neighbors in one subsystem. The directions of frustrated exchange bonds determining the direction of the modulation vector of the magnetic structure are the same in the considered model. This provides the conservation of the given direction under the variation of the type of incommensurate structure (Fig. 1). Such a scheme of exchange interactions is implemented in tetragonal ferrimagnet CuB_2O_4 [10, 11]. As will be shown below, it gives both flat and conical solutions. The Hamiltonian of the model has the form

$$H = J_a\sum_{ii'}\mathbf{S}_i\mathbf{S}_{i'} + J_{b1}\sum_{jj'}\mathbf{S}_j\mathbf{S}_{j'} + J_{b2}\sum_{jj''}\mathbf{S}_j\mathbf{S}_{j''} + J_{ab}\sum_{ij}\mathbf{S}_i\mathbf{S}_j, \quad i \in \mathrm{A},\ j \in \mathrm{B}, \quad (1)$$

$$z_aJ_a > z_{b1}|J_{b1}|, \quad z_{b2}J_{b2}, \quad z_{ab}J_{ab}, \quad z_{ab}J_{ab} > 0,$$

where i and j are the indices of spins of A and B subsystems, respectively; z_{ab} (z_{ba}) is the number of mag-

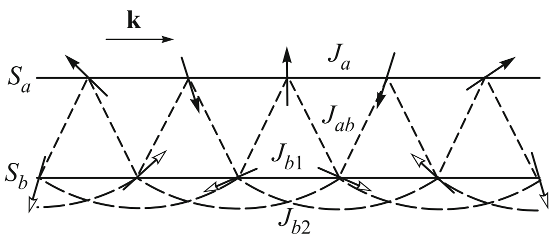

Fig. 1. Scheme of exchange interactions and orientations of spins in the antiferromagnetic flat helix at $J_{b1} > 0$.

netic neighbors in the position B (A) for the spin in the position A (B); and J_a, J_{b1}, and J_{b2} are exchanges inside the subsystem. The numbers of magnetic neighbors z_{ab} and z_{ba} for the intersubsystem exchange J_{ab} are related to the numbers of magnetic ions in subsystems N_a and N_b by the relation

$$\frac{z_{ab}}{z_{ba}} = \frac{N_b}{N_a} = n.$$

The antiferromagnetic exchange in the unfrustrated A subsystem is considered dominant. This makes it possible to use the mean field approximation (MFA) when considering the states arising at temperatures below the antiferromagnetic ordering temperature T_N in the A subsystem (in the antiferromagnetic phase). We consider the cases of antiferromagnetic ($J_{b1} > 0$) and ferromagnetic ($J_{b1} < 0$) exchanges between the nearest magnetic neighbors in the B subsystem giving different conical solutions. In the mean field approximation, the Hamiltonian is additive over spins:

$$\begin{gathered} H_{\mathrm{MFA}} = \sum_i \mathbf{h}_i \mathbf{S}_i + \sum_j \mathbf{h}_j \mathbf{S}_j, \\ \mathbf{h}_i = \frac{1}{2} J_a \sum_{i'} \mathbf{S}_{a,i'} + \frac{1}{2} J_{ab} \sum_j \mathbf{S}_{b,j}, \\ \mathbf{h}_j = \frac{1}{2} J_{b1} \sum_{j'} \mathbf{S}_{b,j'} + \frac{1}{2} J_{b2} \sum_{j''} \mathbf{S}_{b,j''} + \frac{1}{2} J_{ab} \sum_i \mathbf{S}_{a,i}, \end{gathered} \tag{2}$$

where $\mathbf{S}_{a,\,i}$ and $\mathbf{S}_{b,\,j}$ are the vectors of average spins in the A and B subsystems, respectively. The necessary condition for the existence of stationary states in the mean field approximation is the collinearity of average spins to the corresponding total fields [13]. This requirement is equivalent to the constraint imposed on the effective fields acting on spins: the transverse components of the fields should be zero. In our case of two nonequivalent magnetic positions, the fields on spins $\mathbf{h}_{i,\,j}$ are functions of four angles of the orientation of spins $\theta_{a,\,b}$ and $\varphi_{a,\,b}$ (polar and azimuthal angles in the local spherical coordinates of the corresponding spins) and two average values S_a and S_b. Four collinearity conditions and two self-consistency equations for the average values of spins in the mean field approximation [14] form a complete system of nonlinear equations for the variables of the problem:

$$h_{a,b}^{\theta,\varphi}(\theta_{a,b}, \varphi_{a,b}, S_{a,b}) = 0, \tag{3}$$

$$S_{a,b} = -S_{a,b}^0 B_{S_{a,b}^0}\left(\frac{h^{a,b} S_{a,b}^0}{T}\right), \tag{4}$$

where $h_{a,b}^{\theta,\varphi}$ are the transverse fields directed along vectors $\mathbf{e}_{i,j}^{\theta}$ and $\mathbf{e}_{i,j}^{\varphi}$ of local coordinate systems, $h^{a,\,b}$ are transverse fields, and $B_{S_{a,b}^0}(x)$ is the Brillouin function for the spins $S_{a,b}^0$. In local coordinate systems, $S_{a,\,b} > 0$ and $h_{a,\,b} < 0$. We define the dimensionless exchange parameters of the model and the longitudinal effective fields normalized to the complete exchange interactions between A spins, as well as the frustration parameter of exchanges between B spins R and the temperature t normalized to the Néel temperature of the A subsystem:

$$\begin{gathered} j_b = \frac{z_{b1} J_{b1}}{z_a J_a}, \quad j_{ab} = \frac{z_{ab} J_{ab}}{z_a J_a}, \quad j_{ba} = \frac{z_{ba} J_{ab}}{z_a J_a}, \\ h_{a,b} = \frac{h^{a,b}}{z_a J_a}, \quad R = \frac{z_{b2} J_{b2}}{z_{b1} |J_{b1}|}, \\ t = \frac{T}{T_N} = \frac{6T}{S_a^0 (S_a^0 + 1) z_a J_a}, \\ j_{ab}, \quad j_{ba}, \quad R, \quad t \in \{0,1\}, \quad -1 < j_b < 1. \end{gathered} \tag{5}$$

In this notation, self-consistency equations (4) have the form

$$S_{a,b} = -S_{a,b}^0 B_{S_{a,b}^0}\left[\frac{6 h_{a,b} S_{a,b}^0}{S_a^0 (S_a^0 + 1) t}\right]. \tag{6}$$

The system of Eqs. (3) and (4) determines all solutions with two nonequivalent magnetic positions. To find the solution with the minimum free energy,

$$\begin{gathered} F = -T \ln Z, \\ Z_{\mathrm{MFA}} = \mathrm{Sp}\exp\left(\frac{H_{\mathrm{MFA}}}{T}\right) = Z_a^{N_a} Z_b^{N_b}, \end{gathered} \tag{7}$$

where $Z_{a,\,b}$ are the single-particle partition functions, the free energy is varied over the variables of the problem

$$\delta F = N_a S_a \delta h_a + N_b S_b \delta h_b = 0. \tag{8}$$

Structures with two cones in the subsystem with the main unfrustrated exchange that are located symmetrically with respect to the plane of spins of the second subsystem are conical structures where magnetic moments have a three-dimensional orientation and the number of nonequivalent magnetic positions is two (Fig. 2). When the symmetrical distribution of the projections of spins of the subsystem A on the plane of B spins is preserved (Fig. 2b), the conditions of vanishing of two transverse components on the spins $\mathbf{S}_{b,\,j}$ and the h_a^{φ} component on the spins $\mathbf{S}_{a,\,i}$ are fulfilled

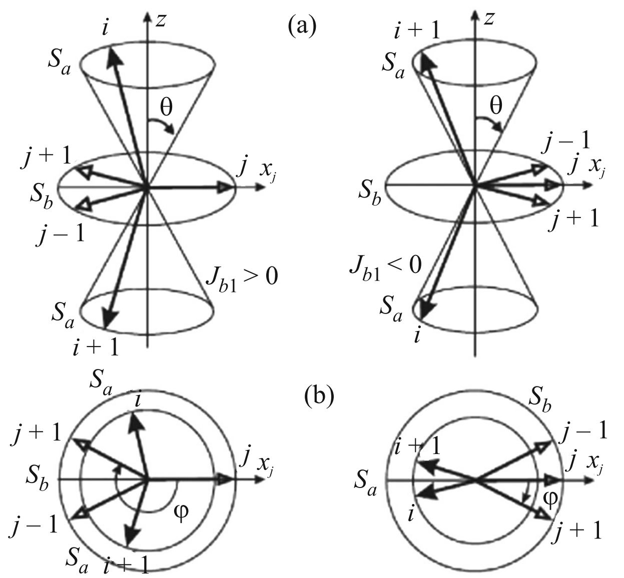

Fig. 2. (a) Orientations of spins in symmetrical conical helices at $J_{b1} > 0$ and $J_{b1} < 0$ and (b) the projections of spins of the helix plane.

automatically. The requirement of vanishing of the component h_a^θ imposes the additional constraint on the angles and average spins

$$h_a^\theta = \cos\frac{\varphi}{2}\cos\theta\left(S_a\sin\theta\cos\frac{\varphi}{2} \pm S_b\frac{j_{ab}}{2}\right) = 0, \quad (9)$$

giving three solutions:

(i) $\cos(\varphi/2) = 0$. The solution arises for the antiferromagnetic exchange ($J_{b1} > 0$) and corresponds to the antiferromagnetic ordering in both subsystems of the "cross" type [15] with the ground state degenerate with respect to the mutual orientation of the antiferromagnetism vectors of the subsystems. This state is globally unstable and transfers into a flat incommensurate structure at any infinitesimally small values of the intersubsystem exchange [11].

(ii) $\cos\theta = 0$, corresponding to an antiferromagnetic flat helix (AFH) at $J_{b1} > 0$ and a flat triangular Yafet–Kittel (YK) structure [15] at $J_{b1} < 0$.

(iii) Symmetrical conical helices (SCHs) with the opening angles of the cone determined by the relation

$$\sin\theta = \mp\frac{j_{ab}S_b}{2S_a\cos(\varphi/2)}. \quad (10)$$

The upper and lower signs in Eqs. (9) and (10) and below refer to the cases of antiferromagnetic and ferromagnetic exchanges J_{b1}, respectively. For SCH solutions, the transverse fields on spins have the form

$$\begin{aligned} h_a^{\mathrm{SCH}} &= -\frac{S_a}{2}\cos^2\theta + \frac{S_a}{2}\sin^2\theta\cos\varphi \\ &\pm \frac{S_b}{2}j_{ab}\sin\theta\cos\frac{\varphi}{2}, \\ h_b^{\mathrm{SCH}} &= \frac{S_b}{2}j_b(\cos\varphi \pm R\cos2\varphi) \\ &\pm \frac{S_a}{2}j_{ba}\sin\theta\cos\frac{\varphi}{2}. \end{aligned} \quad (11)$$

The substitution of Eq. (10) into Eqs. (11) makes it possible to exclude the angle θ from the independent variables of the problem:

$$\begin{aligned} h_a^{\mathrm{SCH}} &= -\frac{S_a}{2}, \\ h_b^{\mathrm{SCH}} &= \frac{S_b}{2}\left(j_b(\cos\varphi \pm R\cos2\varphi) - \frac{j_{ab}j_{ba}}{2}\right). \end{aligned} \quad (12)$$

In the conical phase, the decrease in the exchange field on A spins owing to their non-collinearity is exactly compensated by the field from B spins. As a result, this field remains equal to the field at the antiparallel orientation of antiferromagnetic sublattices of A spins and depends neither on average S_b values nor on the helix step φ. For the B subsystem, the interaction with A spins is reduced to the additional effective exchange between B spins. Thus, the minimization of the free energy given by Eq. (8) is reduced to the variation of the longitudinal field h_b^{SCH} (12) over the helix step that gives the standard expressions for $R > 1/4$:

$$\cos\varphi = \mp(4R)^{-1}. \quad (13)$$

At $R < 1/4$, $\varphi = \pi$ and solutions specified by Eq. (10) at finite S_b do not exist. Substituting Eq. (13) into Eqs. (10) and (12), we obtain

$$\sin\theta = \frac{j_{ab}S_b}{S_a}\left(2 \mp \frac{1}{2R}\right)^{-1/2}, \quad (14)$$

$$h_b^{\mathrm{SCH}} = \mp\frac{S_b}{2}\left[j_b\left(R + \frac{1}{8R}\right) \pm \frac{j_{ab}j_{ba}}{2}\right]. \quad (15)$$

Thus, conical phases arise from the antiferromagnetic phase with the appearance of the magnetization of B sites when the threshold condition is fulfilled. The opening angle of the cone increases continuously with the further decrease in the temperature: A spins tend to the B plane.

We find the temperature of the (antiferromagnet ⇒ symmetrical conical helix) second-order phase transition by linearizing the self-consistency equation (6) for S_b with allowance for Eq. (15):

$$t_{\mathrm{SCH}} = \frac{S_b^0(S_b^0+1)}{S_a^0(S_a^0+1)}\left[\pm j_b\left(R + \frac{1}{8R}\right) + \frac{j_{ab}j_{ba}}{2}\right]. \quad (16)$$

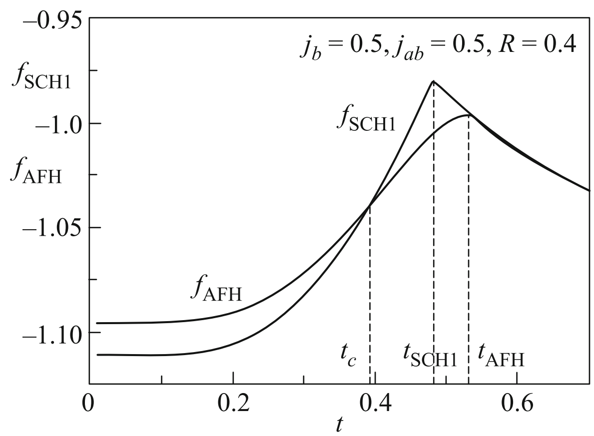

Fig. 3. Temperature dependences of the normalized free energy of incommensurate AFH and SCH1 states.

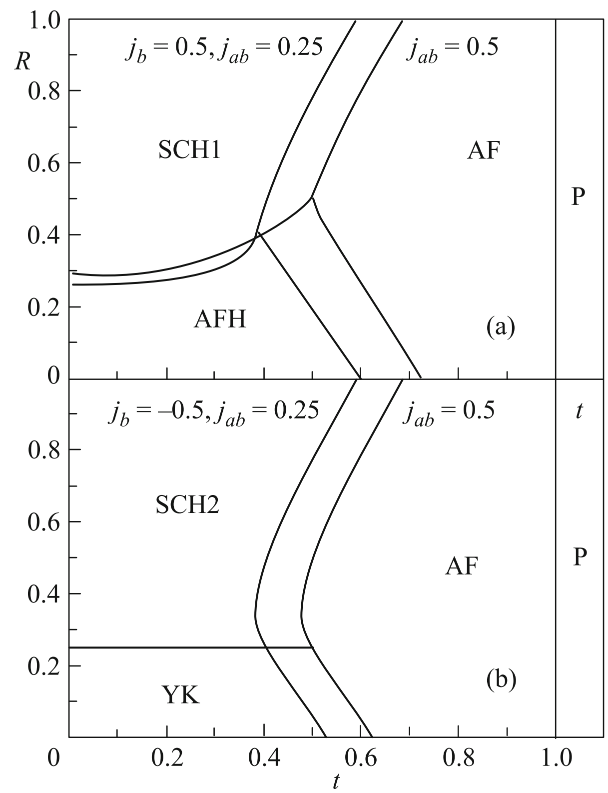

Fig. 4. (Temperature–ratio of competing exchanges in the B subsystem) phase diagrams at the fixed (a) antiferromagnetic and (b) ferromagnetic exchanges j_b and two values of the intersubsystem exchange: j_{ab} = 0.25 and 0.5; P is the paramagnetic phase, in which the spins of both subsystems are disordered.

The exchange energy of frustrated interactions leading to the considered incommensurate structures has different dependences on the average values S_a and S_b:

$$E_{\rm AFH} \propto -AS_aS_b,$$

$$E_{\rm SCH} \propto -BS_b^2.$$

Different temperature dependences of the magnetization of subsystems lead to different temperature dependences of the free energy in the AFH and SCH states. At $t \ll 1$, spins S_a are close to saturation and, when AFH appears first, the faster decrease in the free energy of the SCH state with a further decrease in the temperature can lead to the change in the type of incommensurate ordering. To determine the type of such phase transition, we perform the numerical minimization of the free energy given by Eq. (4) for the particular case $S_a^0 = S_b^0 = 1/2$ and $n = 1$ with the fixed parameters $j_{ab} = j_{ba}, j_b$, and R. Figure 3 shows the temperature dependences of the free energy of the AFH and SCH1 states normalized to the number of spins N_a and the total exchange between neighboring A spins z_aJ_a. The type of incommensurate ordering changes at t_c and the angle θ varies stepwise to the value θ_c. The helical step and magnetizations of the subsystem also vary stepwise. The further decrease in the temperature leads to the increase in the angle θ, while the helical step remains constant (see Eq. (13)). Thus, the transition between the flat and conical incommensurate phases is a first-order phase transition.

(Temperature–exchange parameters) phase diagrams are shown in Figs. 4 and 5. At large values of the frustration parameter R of competing exchanges in the subsystem B, the conical SCH1 and SCH2 phases arise at the decrease in the temperature from the antiferromagnetic phase (AF), in which only spins of the A subsystem are ordered, with the appearance of the magnetization of the B subsystem through a second-order phase transition. At small R values, either the flat incommensurate phase (AFH at $j_b > 0$ in Fig. 4a) or the triangular Yafet–Kittel structure (YK at $j_b < 0$ in Fig. 4b) appears from the AF phase also through a second-order phase transition. In the former case, the further decrease in the temperature can lead to the first-order phase transition from the flat phase to the conical one. Since the mechanism of the formation of the flat incommensurate structure is the geometrical frustration of the intersubsystem exchange and the partial removal of degeneracy in such structures occurs according to this exchange, the interface between the flat and conical SCH1 phases depends on the exchange value. At the ferromagnetic exchange ($j_b < 0$), the interface between the triangular Yafet–Kittel phase and the conical SCH2 phase remains constant (R = 0.25) under the temperature variation for different values of the intersubsystem exchange (Fig. 4b). The conical phase arises starting from the zero helical step and the rotation begins around the axis lying in the plane of the triangular structure and normal to the direction of spins S_b (Fig. 2a). The SCH1 and SCH2 states are separated by the flat incommensurate phase, the width of which is determined by the intersubsystem exchange j_{ab} (Fig. 5). At

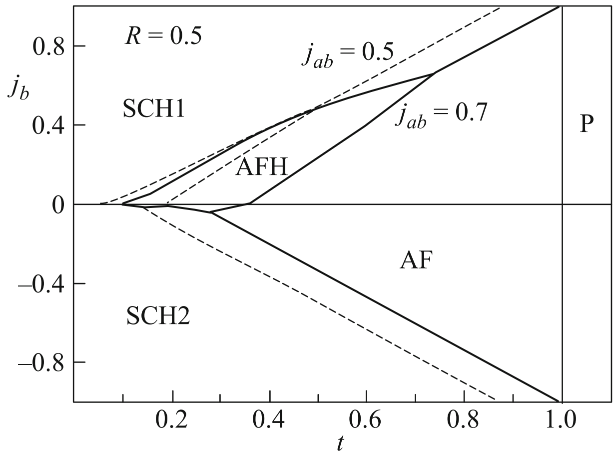

Fig. 5. (Temperature–exchange interaction in the B subsystem) phase diagram j_b at the fixed ratio of competing exchanges R and two values of the intersubsystem exchange: j_{ab} = (dashed lines) 0.5 and (solid lines) 0.7.

the same R values setting the helical step, the conical angles of these structures are different (see Eq. (14)) since they are determined by the angle $\phi/2$ between the projections of the spins of subsystems on the helical plane. In the long-wavelength limit ($R \to 0.25$), the expression determining the opening angle θ of the cone for the conical SCH1 and SCH2 structures should be used with $\phi/2 = \pi/2$ and 0, respectively.

In the considered model, the increase in the frustration of exchanges inside one subsystem is accompanied by the formation of the three-dimensional incommensurate structure because of the competition of optimal non-collinearities different for different partial subsystems. The large non-collinearity of B spins is energy favorable at large parameters R. Such cant of the antiferromagnetic sublattices of the unfrustrated A subsystem in the flat helix becomes unfavorable with respect to the energy of the dominant exchange J_a, and spins S_a come out of the helical plane. As a result, the cant between sublattices decreases and the initial exchange field on the spins of the subsystem is recovered. This leads to the considerable decrease in the energy with respect to the case of the flat helix with the locally triangular orientation of spins (Yafet–Kittel helix) considered earlier [12]. In this case, the helical step is intermediate between the optimal one determined by standard relation (13) and zero favorable in terms of the dominant exchange. As a result, the energy of exchange interactions in both subsystems for such incommensurate structure increases with respect to that of the conical helix. The additional magnetic anisotropy of the “easy-plane” type can make the flat incommensurate structure more energy favorable. At the same values of frustrated interactions, the three-dimensional orientation of isotropic Heisenberg spins is an additional opportunity to decrease the energy with respect to the anisotropic Ising and *XY* spin systems. The model disregards not only anisotropy but also the effect of magnetostriction playing a significant role first of all in the appearance of the electrical polarization in incommensurate magnets, e.g., via the inverse Dzyaloshinskii–Moriya effect [3, 16]. The appearance of such an additional mechanism of the incommensurability of the magnetic structure, as well as the striction change in isotropic exchanges, will not lead to qualitative variation of phase diagrams and conditions of the appearance of conical phases at the considered large values of frustrated exchanges of the model.

REFERENCES

1. S.-W. Cheong and M. Mostovoy, Nature Mater. **6**, 13 (2007).
2. T. Kimura, Ann. Rev. Mater. Res. **37**, 387 (2007).
3. Y. Tokura and S. Seki, Adv. Mater. **22**, 1554 (2010).
4. S. Ishiwata, Y. Taguchi, H. Murakawa, Y. Onose, and Y. Tokura, Science **319**, 1643 (2008).
5. Y. Kitagawa, Y. Hiraoka, T. Honda, T. Ishikura, H. Nakamura, and T. Kimura, Nature Mater. **9**, 797 (2010).
6. M. Soda, T. Ishikura, H. Nakamura, Y. Wakabayashi, and T. Kimura, Phys. Rev. Lett. **106**, 087201 (2011).
7. T. A. Kaplan, Phys. Rev. **119**, 1460 (1960).
8. D. H. Lyons, T. A. Kaplan, K. Dwight, and N. Menyuk, Phys. Rev. **126**, 540 (1962).
9. K. Tomiyasu, J. Fukunaga, and H. Suzuki, Phys. Rev. B **70**, 214434 (2004).
10. S. N. Martynov and A. D. Balaev, JETP Lett. **85**, 649 (2007).
11. S. N. Martynov, J. Exp. Theor. Phys. **108**, 72 (2009).
12. S. N. Martynov, JETP Lett. **95**, 188 (2012).
13. T. A. Kaplan and N. Menyuk, Philos. Mag. **87**, 3711 (2007).
14. J. S. Smart, *Effective Field Theories of Magnetism* (W. B. Saunders, Philadelphia, London, 1966)].
15. Y. Yafet and C. Kittel, Phys. Rev. **87**, 290 (1952).
16. I. A. Sergienko and E. Dagotto, Phys. Rev. B **73**, 094434 (2006).

Translated by L. Mosina