Inhomogeneuos state of ferrimagnetic film near compensation point

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Abstract. Recently, an investigation of the Faraday rotation in thin iron garnet film (LuBi)₃(FeAlGa)₅O₁₂ revealed an unusual “precursor” of transition to a noncollinear phase. It was observed in the vicinity of the compensation temperature. However, this feature cannot be ascribed to the uniform phase transition associated with uniaxial anisotropy. A model for the “precursor” based on inhomogeneous magnetic structure at the substrate-film interface is presented. To interpret the Faraday rotation, two diamagnetic optical transitions attributed to tetrahedral and octahedral iron sublattices are considered.

1. Introduction
Iron garnets are widely used in magnetooptical and novel magnonic devices due to their unique properties [1, 2]. Despite the fact that Bi³⁺ ion itself is nonmagnetic, Bi-substitution in dodecahedral position of iron garnets leads to a drastic enhancement of magneto-optical effects [3-5]. The mechanism of this phenomenon was intensively investigated [6] and presently it is related to an influence of Bi³⁺ on optical transitions of ions Fe³⁺ in the tetrahedral and octahedral positions. Iron garnets allow fabrication of solid solutions and adjustment of magnetic and optical characteristics in a wide range.

Recently, a magnetic phase diagram of thin iron garnet film (LuBi)₃(FeAlGa)₅O₁₂ about 8 μm in thickness was investigated by means of magnetooptical Faraday effect [7]. The film was epitaxially grown on a (111) Gd₃Ga₅O₁₂ substrate. Since ions in the dodecahedral environment (Bi³⁺, Lu³⁺) were nonmagnetic, the magnetic structure was formed by two iron sublattices: tetrahedral and octahedral. The ions Fe³⁺ were in the same state S=5/2 in both crystallographic positions. A ratio of the iron ion numbers in the tetrahedral sublattice to that in octahedral one is 3:2 in undiluted iron garnet. Due to a strong AFM interaction between sublattices, this substance turned out to be ferrimagnetic. However, a selective substitution of iron ions by nonmagnetic Ga³⁺ and Al³⁺ ions made it possible to get almost equal magnetic moments of the two sublattices. That is why, (LuBi)₃(FeAlGa)₅O₁₂ compound had a compensation point at temperature about 65 K [7]. The film had a uniaxial anisotropy perpendicular to the surface.

The investigation of the (LuBi)₃(FeAlGa)₅O₁₂ film grown on the gadolinium gallium garnet revealed an unusual “precursor” of the transition to noncollinear phase in the vicinity of the compensation temperature [7] (figure 1). It looked like a small step and a plateau just before the phase transition. The “precursor” was observed in increasing and decreasing magnetic field without a noticeable hysteresis. The two branches are shown in figure 1 for T = 80 K by dash and solid lines. It was impossible to explain
the shape of the curve by the traditional theory for uniform almost compensated ferrimagnets [8] especially that was observed at 80 K because the AFM angle and, therefore, the Faraday rotation angle should strongly depend on the external magnetic field in the noncollinear phase. This fact contradicted to the plateau observed in figure 1.

In the present work, we propose a model for description of the “precursor” assuming induced inhomogeneous magnetic structure at the substrate-film interface.

2. Model
The specific Faraday rotation $\Theta_F$ in compensated or almost compensated iron garnets originates from different contribution of the tetrahedral (d) and octahedral (a) iron sites [1]. Here we assume the ions in dodecahedral positions to be nonmagnetic. Then magnetooptical Faraday effect (figure 1) can be described in terms of the Neel model as a result of rotation of sublattice magnetizations $M_d$ and $M_a$ [1, 4, 5], i.e.

$$\Theta_F = C_d M_d \cos(\vartheta_d) + C_a M_a \cos(\vartheta_a),$$

(1)

where $C_d, C_a > 0$ are magneto-optical coefficients and $\vartheta_d, \vartheta_a$ are angles between applied magnetic field and tetrahedral and octahedral magnetization vectors, correspondingly (magnetic field is applied perpendicular to the film surface).

It is well-known that magnetization vector is of almost constant magnitude under a low magnetic field (in the vicinity to the phase transition to the noncollinear phase) [1]. Furthermore, due to strong AFM interaction the angle between sublattices approximates to 180 degree in weak magnetic fields. Therefore, in vicinity of the phase transition, specific Faraday rotation can be written as

$$\Theta_F \approx (C_d M_d - C_a M_a) \cos(\vartheta_{AFM}),$$

(2)

where $\vartheta_{AFM} \equiv \vartheta$ is the angle between the AFM vector and magnetic field direction.

In the vicinity of the compensation temperature there is a point on the magnetic phase diagram, which is sometimes called a “throat” [8], where the uniaxial anisotropy is compensated by an induced transverse anisotropy. While approaching this point, the magnetic structure becomes sensitive to small perturbations. As was shown in [9] a surface anisotropy can cause magnetic inhomogeneity in a surface layer or shift phase transitions in ultrathin films. In the present work we discuss another source of the inhomogeneity, namely, magnetic interaction of the film with paramagnetic gadolinium gallium garnet substrate.

The volume energy density of the almost compensated ferrimagnet [8] can be presented in dimensionless form as

![Figure 1. Faraday rotation in (LuBi)$_3$(FeAlGa)$_5$O$_{12}$ film at different temperatures [7]. The increasing and decreasing field branches for 80 K are plotted by dash and solid lines. The precursor of the transition to noncollinear phase is shown by the arrow. The dash-dot lines are guide for the eye.](image-url)
\( f_v = -\eta h \cos (\vartheta) - (k_u - h^2) \cos^2 (\vartheta), \)  \hspace{1cm} (3)

where \( \eta = m_d - m_a, m_d \) and \( m_a \) are dimensionless magnetic moments of the tetrahedral and octahedral sublattices, \( m_d + m_a = 2, k_u > 0 \) is the easy-axis anisotropy constant (the easy axis is perpendicular to the surface of the film), \( \vartheta \) is the angle between the easy axis and the AFM vector, and \( h \) is the external magnetic field that was directed perpendicular to the film surface (along the easy axis). The last term in the brackets corresponds to the transverse susceptibility [8]. The equilibrium value of the angle in the volume \( \vartheta \), determined by minimization of \( f_v \) with respect to \( \vartheta \).

The substrate contains the paramagnetic ions \( \text{Gd}^{3+} (S=7/2) \) which interact with the iron sublattices of the film within an area of about a lattice period in thickness. This leads to the interface energy

\[ f_s = h \chi \lambda \cos (\vartheta_s), \]  \hspace{1cm} (4)

where \( \chi \) is the susceptibility of gadolinium ion, \( \lambda \) is the interaction constant which can be estimated from molecular field coefficients for \( \text{Ga}_3\text{Fe}_5\text{O}_{12} \) [11], \( \vartheta_s = \vartheta (0) \) is the AFM angle at the interface. It should be mentioned that above the compensation temperature \( \lambda > 0 \), therefore the interface effective field is directed oppositely to the external field.

Finally, the total energy takes the form of a functional of \( \vartheta (z) \) [9]

\[ F = \int_0^d \left[ A \left( \frac{d \vartheta}{dz} \right)^2 + f_v (\vartheta) \right] dz + f_s, \]  \hspace{1cm} (5)

where \( d \) is the film thickness, \( A \) is a constant of the order of \( a^2 \), \( a \) is the distance between the nearest neighboring magnetic ions. As we see below the film used in [7] is thick films as compared to an inhomogeneity length. Then it is possible to replace \( d \to \infty \) [12].

Following [9] one can derive an equation for calculation of the AFM angle at the surface by minimization of the functional (5). The Lagrange-Euler equation takes the form

\[ 2A \frac{d^2 \vartheta}{dz^2} - \frac{\partial f_v}{\partial \vartheta} = 0. \]  \hspace{1cm} (6)

Combining it with the boundary conditions we obtain the equation for the AFM angle at the interface \( \vartheta_s \)

\[ \frac{df_s}{d\vartheta_s} = -A^{1/2} \sqrt{f_v (\vartheta_s) - f_v (\vartheta_s)}. \]  \hspace{1cm} (7)

The spatial distribution \( \vartheta (z) \) can be evaluated from the following expression

\[ A^{1/2} \frac{d \vartheta}{dz} = -\sqrt{f_v (\vartheta) - f_v (\vartheta)}. \]  \hspace{1cm} (8)

When \( f_v (\vartheta) - f_v (\vartheta) \) is small, the equation (5) can be linearized. Then the solution is given by

\[ \vartheta = \exp (-z/\delta) + \vartheta_s, \hspace{0.5cm} \delta = \sqrt{A^2 h \eta - 2k_u + 2h^2}. \]  \hspace{1cm} (9)

From here one can obtain an estimation for the characteristic inhomogeneity length.

3. Results and discussion

The results of calculation of inhomogeneous magnetic structure are shown in figure 2 (\( \eta = 10^{-4}, \chi \lambda = 2 \times 10^{-10}, k_u = 10^{-4}, A = 10^{-20} \text{ m}^2 \)). The function \( \vartheta (z) \) corresponds to magnetic fields below the transition (the lines 1 and 2) and above it (the line 3). The line 3 tends to the equilibrium volume value at \( z \to \infty \) (the angle is nonzero in the noncollinear phase). The critical magnetic field corresponding to...
the bulk transition to non collinear phase is \( h_C = 0.010025 \). The “precursor” appears at magnetic field of about \( h_P = 0.0072 \) and persists up to \( h_C \). In this range the nonuniform magnetic structure exists. The characteristic length of the area with inhomogeneous magnetic structure was typically much less than the film thickness (figure 2), which was about 8 \( \mu \text{m} \) [7]. That is why, the assumption of thick film occurred to be valid. Despite the small inhomogeneous area it was sufficient for experimental observation of the “precursor”. It also should be noted that the length of the inhomogeneous magnetic structure (from 0.1 to 1 \( \mu \text{m} \) according to figure 2) agrees with that was discussed for GdCo thin films [9, 13].

![Figure 2](image)

**Figure 2.** The distribution of the angle between the easy axis and the AFM vector near the film-substrate interface: \( h = 0.097, h = 0.01, \) and \( h = 0.0102 \) (the lines 1, 2, and 3) according to equation (8).

The total Faraday rotation at weak magnetic fields taking into account the interface contribution can be presented in the following form

\[
\Theta = C \int_0^d \cos \left[ \vartheta(z) \right] \, dz,
\]

where \( \vartheta(z) \) is the solution of the equation (8), \( C = C_e M_d - C_s M_u \) and \( d \) is the film thickness.

The calculation results of the Faraday rotation according to equation (10) are shown in figure 3. The dashed (1) and solid (2) lines were obtained for \( \eta = 10^{-4} \) and \( 2 \times 10^{-3} \). It should be mentioned that an increase of \( \eta \) in this model corresponds to the increase of the temperature. In first case the critical magnetic field \( h_C \) was about 0.010025 and “precursor” began in \( h_P = 0.0072 \). In second case: \( h_C = 0.0105125 \) and \( h_P = 0.0074 \).

![Figure 3](image)

**Figure 3.** The normalized Faraday rotation as a function of magnetic field for \( \eta = 10^{-4} \) (dashed line) and \( \eta = 2 \times 10^{-3} \) (solid line). The solid and dashed lines were calculated using \( \chi \lambda = 2 \times 10^{-10} \) and \( k_u = 10^{-4} \).
In conclusion, the “precursor” of the phase transition observed in [7] stems from the spatial inhomogeneity of the magnetic structure at film surface. A detailed investigation of the nonuniform magnetization distribution can be performed be means of magneto-optical Kerr effect measurements which are sensitive to surface magnetic structure [13]. The model proposed in the present article is based on the assumption that the magnetic inhomogeneity is induced by the film-substrate interface. This approach is similar to the surface anisotropy model proposed earlier [9]. The main difference between them lies in the form of nucleation disturbance. The surface energy in the present work has the angle dependence \( f_s \propto \cos(\phi_s) \) in contrast to \( f_s \propto \cos^2(\phi_s) \) in [9]. That is why, in our model the “precursor” should disappear below the compensation temperature (the surface energy changes the sign). Thus, an investigation of the transition to noncollinear phase on the both sides from the compensation point allows clarifying the origin of the “precursor”.

Acknowledgment
The work was partially supported by the Russian Foundation for Basic Research. (Project No.18-48-520006).

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