Two-sublattice model of magnetic linear birefringences in rare-earth iron garnets

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A phenomenological analysis of magnetic linear birefringence is presented within the framework of the two-sublattice model. The magnetic linear birefringence of yttrium and terbium and terbium iron garnets is investigated experimentally in the temperature interval 80–450 K in fields up to 30 kOe at a wavelength 1.15 μm. It is shown that in the yttrium iron garnet at all temperatures, and in the terbium garnet in a sufficiently wide range of temperatures, the magnetic linear birefringence and its anisotropy can be described with good accuracy by the two-sublattice model. Deviations from the two-sublattice model in the terbium iron garget at low temperatures are attributed to changes of the noncubic distortions in the local environment of the magnetic ions.

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1. INTRODUCTION

Magnetic linear birefringence (MLB) of light, observed a few years ago in cubic rare-earth iron garnets, has a large value $\Delta n \sim 10^{-3}$–10^{-5} and a strong anisotropy, i.e., a dependence of the effect on the orientation of the magnetization in the crystal. By now, the temperature, field, angular, and spectral dependences of this phenomenon have been investigated for a number of rare-earth iron garnets,[1-5] but the theoretical description was carried out only within the framework of the phenomenological single-sublattice model.[2,6,7] This model describes well the angular dependences of the MLB, but is not suitable for the description of the temperature and field dependences. In particular, within the framework of the single-sublattice model it is impossible to explain the strong temperature dependence of the magneto-optical anisotropy parameter, a dependence typical of most rare-earth iron garnets.[2,3] It is possible that the single-sublattice model can describe sufficiently well the MLB in ferromagnets or antiferromagnets with equivalent magnetic sublattices, but for a correct description of MLB in crystals with nonequivalent sublattices it is apparently necessary to take into account the interaction of the light with each sublattice.

In this paper we propose a two-sublattice model and check it experimentally by investigating the temperature and field dependences, as well as the anisotropy of the MLB, in yttrium iron garnet $Y_3Fe_5O_{12}$ and in terbium iron garnet $Tb_3Fe_5O_{12}$.

2. PHENOMENOLOGICAL TWO-SUBLATTICE MODEL

We consider a cubic ferrimagnet with two collinear sublattices characterized by magnetizations $\mathbf{m}_1$ and $\mathbf{m}_2$. We write down the energy $\mathscr{E}$ of the interaction of the electromagnetic wave with the crystal in the transparency region, taking into account terms that are quadratic in the magnetization, in the form

$$8\pi\mathscr{E}=\varepsilon_0\mathbf{E}^2+\lambda_1{}^{ij}m_im_j\mathbf{E}^2+\lambda_2{}^{ij}m_im_j(\mathbf{eE})^2+\lambda_3{}^{ij}m_im_je_\alpha{}^2E_\alpha{}^2, \quad (1)$$

where $\mathbf{E}$ is the electric vector of the light wave; ε_0 is that part of the permittivity which does not depend on the magnetizations of the sublattices; $\mathbf{e}$ is a unit vector in the direction of the spontaneous magnetization $\mathbf{m}=\mathbf{m}_1+\mathbf{m}_2$; λ_k^{ij} are phenomenological constants, and ij take on the values 11, 12, 22; $\alpha=x, y, z$.

From (1) we can obtain an expression for the dielectric tensor and for the values of the MLB. If the wave vector $\mathbf{k}$ of the light is directed along the $[1\bar{1}0]$ axis, then we have for the measured value of the magnetic birefringence

$$\Delta n=a^{ij}m_im_j, \quad (2)$$

where $a^{ij}=2(\lambda_2^{ij}+\lambda_3^{ij})/n_0$ for $\mathbf{m}\|[001]$ and $a^{ij}=2\lambda_2^{ij}/n_0$ for $\mathbf{m}\|[111]$, and n_0 is the refractive index. It is assumed in the considered model that the coefficients a^{ij} do not depend on the temperature and magnetic field. For the field dependence of the MLB we can therefore write

$$d\Delta n/dH=a^{ij}(m_i\chi_j+m_j\chi_i), \quad (3)$$

where χ_i and χ_j are the susceptibilities of the sublattices.

The feasibility in principle of distinguishing between the two-sublattice and single-sublattice models, i.e., the possibility of determining the coefficients a^{ij}, is based on the difference between the temperature and field dependences of m_1 and m_2. If there is no such difference, i.e., if m_1/m_2 is constant, then expression (2) for the two-sublattice model goes over into the corresponding expression for the single-sublattice model. For yttrium iron garnets, in accord with results of an investigation of the nuclear magnetic resonance, there are exact data on the temperature dependences of the magnetizations of the tetrahedral and octahedral sub-

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lattices,[8] and this gives grounds for hoping to be able to distinguish the two-sublattice model from the single-sublattice model in this garnet.

Let m_1 and m_2 denote the relative magnetizations of the tetrahedral and octahedral sublattices; we then have in the investigated temperature range $m_1, m_2 \gg \Delta m \gg \Delta m^2 (\Delta m = m_1 - m_2)$ (Ref. 1). Under this condition, expression (2) can be transformed into

$$\Delta n = (a^{11}+a^{12}+a^{22}) m_1^2 - (a^{12}+2a^{22}) m_1 \Delta m,$$
$$\Delta n = (a^{11}+a^{12}+a^{22}) m_2^2 + (a^{12}+2a^{11}) m_2 \Delta m. \tag{4}$$

If the two-sublattice model is valid, then the quantity $\Delta n/m_i^2$ should be linear function of the argument $\Delta m/m_i$.

The MLB in a terbium iron garnet should obviously be described by a three-sublattice model, but the difference in the temperature and field behavior of the magnetization of the rare-earth sublattices, compared with the iron sublattice, is so strong that the small difference between the tetrahedral and octahedral sublattices can be neglected. We denote, for the terbium iron garnet, the relative magnetizations of the combined iron and terbium sublattices by m_1 and m_2, respectively. In this case the condition $m_1, m_2 \gg \Delta m$ is no longer satisfied, and it is more convenient to analyze the temperature behavior of the field dependences. Since the susceptibility of the rare-earth sublattice greatly exceeds the susceptibility of the summary iron sublattice, expression (3) can be represented in the form

$$d\Delta n/dH = \chi_2 (a^{12} m_1 + 2a^{22} m_2). \tag{5}$$

If the MLB in terbium iron garnet is described by the two-sublattice model, then the dependence of $d\Delta n/dH\chi_2 m_1$ on m_2/m_1 should be linear.

3. EXPERIMENTAL PROCEDURE

We have investigated the field and temperature dependences on the MLB in yttrium and terbium iron garnets at a wavelength $\lambda = 1.15$ µm in fields up to 30 kOe and in the temperature interval 80–450 K. In the comparison of the theory with experiment in the range 20–100 K, we used also the data of Vien *et al.*[4] The temperature was stabilized accurate to ± 0.1 K. The crystals were cut in the (110) and (111) planes, polished, and annealed. The yttrium iron garnet samples were 5–15 mm thick, and the terbium samples were 1–2 mm thick.

To measure the MLB we used an optical system consisting of a light source, polarizer, sample, quarter-wave plate, polarization modulator, analyzer, and photoreceiver. The alternating signal was registered by an ordinary synchronous-detection circuit. The sensitivity of the measurements of the birefringence was $\Delta n \sim 10^{-8}$ for the terbium iron garnet and ~3×10^{-9} for the yttrium garnet. In the experiment we measured the phase difference β of orthogonally polarized light waves; this difference is connected with the birefringence by the relation $\beta = 2\pi\Delta n/\lambda$.

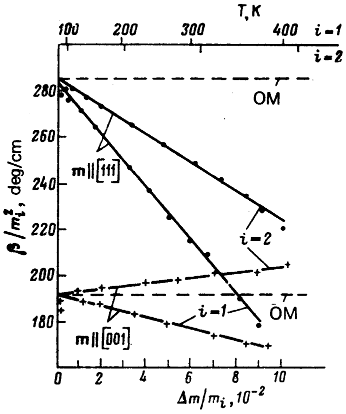

FIG. 1. Plot of β/m_i^2 against $\Delta m/m_i$ in $Y_3Fe_5O_{12}$ for m‖ [111] and m‖ [001]; dashed—single-sublattice model.

4. EXPERIMENTAL RESULTS

Figure 1 shows the experimental results on the temperature dependence of the birefringence in yttrium iron garnet in the form of a plot of β/m_i^2 against $\Delta m/m_i$ for magnetization orientations m‖[001] and m‖[111]. We see that when plotted in these coordinates the experimental data are described, accurate to 2%, by a linear law. At the same time the parameters of the linear dependences satisfy the two-sublattice-model requirements that follow from (4), namely: 1) the free terms of both dependences should be equal, 2) the difference between the slopes should be equal to double the free term (see the table). It must be pointed out that in the single-sublattice model the quantity β/m_i^2 must depend on the temperature. It is seen from Fig. 1 that for this model the deviations of the theory from experiment reach 30% (dashed lines).

In the yttrium iron garnets we have observed for the MLB a field dependence that was linear above the saturation field. Figure 2 shows the variation of the slope of the field dependence $d\beta/dH$ with changing temperature in fields above saturation at k‖[111]. The field dependences have an anisotropic character. On the basis of (3), using the values of the magnetic susceptibility of the yttrium iron garnet,[9] and assuming $\chi_1/\chi_2 = \frac{3}{2}$, we obtain at room temperature the slopes 0.04 and 0.2 deg/cm-kOe at m‖[001] and m‖[110], respectively.

Even though the magnetic susceptibility of the yttrium iron garnet decreases rapidly with decreasing temperature and becomes negligibly small below 190 K,[9] the MLB changes noticeably with increasing field even at 100 K (Fig. 2). One might assume that the small slope is due to the presence of rare-earth impurities in the yttrium iron garnet. An estimate shows, however, that to account for the observed value of $d\beta/dH$ at low tem-

TABLE I.

Orientation	Parameters			Orientation	Parameters		
	$a^{11}+a^{12}+a^{22}$, deg/cm	$a^{12}+2a^{22}$, deg/cm	$a^{12}-2a^{11}$, deg/cm		$a^{11}+a^{12}+a^{22}$, deg/cm	$a^{12}-2a^{22}$, deg/cm	$a^{12}+2a^{11}$, deg/cm
m‖[111]	285±5	1180±30	−615±20	m‖[001]	191±4	256±8	122±4

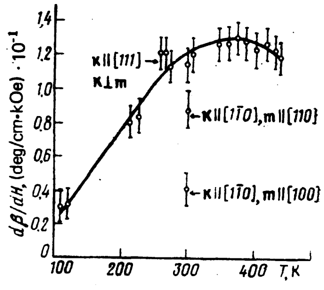

FIG. 2. Slopes of the field dependences of the magnetic birefringence in $Y_3Fe_5O_{12}$ as functions of the temperature.

peratures the impurity concentration must be ~0.1%, which is much higher than the possible value. It must therefore be admitted that, just as in the case of the Faraday effect,[10,11] the phenomenological coefficients of the MLB are noticeably altered when the field is increased.

Investigations of the terbium iron garnet have shown that above the saturation field the MLB changes linearly with the field, and the values of $d\beta/dH$ have opposite signs above and below the magnetic-compensation point.[2] Figure 3 shows the experimental results in the form of plots of $d\beta/dH\chi_2 m_1$ against m_2/m_1 at m∥[001] and m∥[111]. We used in calculations the data of Refs. 9 and 12 on the temperature dependences of the magnetization and of the susceptibility.

In a sufficiently wide temperature interval, from 100 to 380 K at m∥[111] and from 200 to 450 K at m∥[001], the plots shown in Fig. 3 are linear. Using the value of the MLB at one value of the temperature within the limits of the linear section, we can obtain the missing coefficient a^{11}. As a result, the expressions for the MLB take the following form:

$$\beta=-460m_1^2+2250m_1m_2-4930m_2^2 \quad \text{for } \mathbf{m}\parallel[111],$$
$$\beta=-210m_1^2-420m_1m_2+4080m_2^2 \quad \text{for } \mathbf{m}\parallel[001]. \tag{6}$$

(The relative error in the determination of the values of the coefficients is 10%.) These coefficients make it possible to calculate the contributions from the indiv-

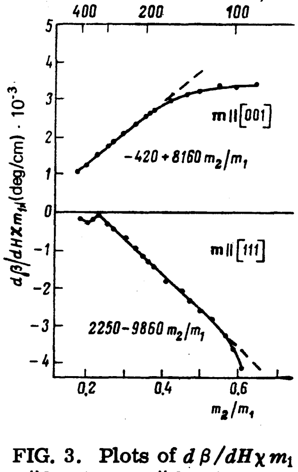

FIG. 3. Plots of $d\beta/dH\chi m_1$ against m_2/m_1 in $Tb_3Fe_5O_{12}$ for m∥[001] and m∥[111].

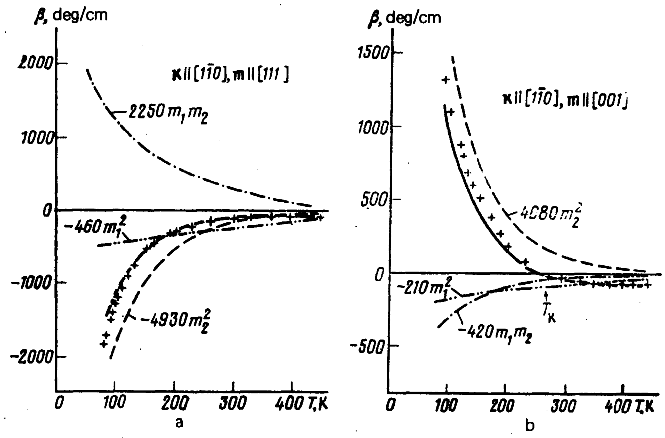

FIG. 4. Calculation of the temperature dependences of the magnetic birefringence in Tb_3FeO_{12} for m∥[111] (a), and m∥[001] (b) at λ=1.15 m. Solid curve—calculation, points (+)—experiment.

idual terms to the temperature dependence of the MLB, as illustrated in Fig. 4 for m∥[111] and m∥[001]. At high temperatures, the competing terms $a^{11}m_1^2+a^{12}m_1m_2$ predominate, and at low temperatures the predominant contribution is that of the rare-earth sublattice, $a^{22}m_2^2$. The contributions $a^{12}m_1m_2$ and $a^{22}m_2^2$, which are connected with the Tb sublattice, are anisotropic, and have opposite signs for m∥[111] and m∥[001].

We see that in the case of the terbium iron garnet the two-sublattice model is valid in a definite temperature interval, but it ceases to hold when the temperature is lowered to 200 K for m∥[001] and 100 K for m∥[111]. The deviation from the linear dependence (5) at an orientation of the magnetization along the difficult magnetization axis [001](~200 K) coincides with the appearance of noncubic lattice distortions in terbium yttrium garnet.[13] A similar deviation for m∥[111] in the case when the isotropic part of the birefringence is measured, appears at temperatures below 100 K. At these temperatures, according to the data of Vien,[14] the anisotropy of the MLB becomes essentially noncubic, i.e., this model cannot be used. The deviations from linearity at m∥[111] and $T\sim380$ K (Fig. 3) are apparently due to neglect of the magnetic susceptibility of the iron sublattice compared with the terbium sublattice. Indeed, in this temperature range the quantity $d\beta/dH$ in terbium iron garnet at m∥[111] is close to the analogous quantity in the yttrium iron garnet (~0.1 deg/cm-kOe). In the case m∥[001] the value of $d\beta/dH$ is much larger and linearity is preserved up to 450 K.

5. DISCUSSION OF RESULTS

Thus, the experimental results on the temperature dependence of MLB in the yttrium iron garnet in the entire temperature interval, and in the terbium garnet in a sufficiently wide interval, can be explained by the two-sublattice model. Let us dwell briefly on a qualitative microscopic explanation of the MLB in ferrimagnets. The sublattice contributions to the MLB depend on the characteristics of the electrodipole optical transitions (resonant frequencies, oscillator strengths) and on the splitting of the ground and excited states of the transitions in the effective magnetic field $H_{\rm eff}$. It is known that the MLB is proportional to $H^2_{\rm eff}$ (Ref. 15), and the connection between the MLB and the sublattice magnetization points to a connection of $\mathbf{H}_{\rm eff}$ with $\mathbf{m}_1$ and $\mathbf{m}_2$. If it is assumed that the effective field $\mathbf{H}^{1(2)}_{\rm eff}$ acting on the ions of the first (second) sublattice is given by $\mathbf{H}^{1(2)}_{\rm eff} = J\mathbf{m}_{2(1)}$, then the MLB should be proportional to a sum of the form $a^{11}m_1^2 + a^{22}m_2^2$. Experiment, however, yields a dependence of the type $a^{11}m_1^2 + a^{12}m_1m_2 + a^{22}m_2^2$, which can be explained only by assuming the existence of an effective field of the form

$$\mathbf{H}^{1(2)}_{\rm eff} = J\mathbf{m}_{2(1)} + \xi^{1(2)}\mathbf{m}_{1(2)}.$$

Only such an effective field, which includes also a dependence on the magnetization of the intrinsic sublattice, can explain the existence of the term $a_{12}m_1m_2$.

The constant $\xi^{i(j)}$ can include both the spin–orbit interaction and the exchange interaction between the ions in one sublattice. It appears that in the case of garnets the spin–orbit interaction is more important and is either comparable in magnitude or can exceed the sublattice exchange J, while by themselves these interactions do exceed the exchange between the sublattices.

The experimental results point to the existence of a considerable anisotropy of the MLB and to its variation with temperature. The anisotropy of the MLB is microscopically connected with the anisotropy of $\mathbf{H}_{\rm eff}$. In turn, the anisotropy of $\mathbf{H}_{\rm eff}$ is connected with violations of the cubic symmetry at the location of the magnetic ions, when the exchange and spin–orbit interactions are described by tensors of second rank. The spectroscopic investigations[16] indeed point to the existence of an appreciable anisotropy of $\mathbf{H}_{\rm eff}$, due to the action on the iron sublattice on the rare-earth sublattice. In the case of the terbium iron garnet, as noted above, appreciable lattice distortions appear with decreasing temperature.[13] This should be reflected in the values of the coefficients λ_3^{ij} and cause deviations from a two-sublattice model in which the phenomenological coefficients are constant (Fig. 3). It appears that in the yttrium iron garnet the local environment of the magnetic ions changes little with temperature and the two-sublattice model with constant coefficients holds true in the entire temperature interval (Fig. 1).

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