Magneto-optical properties of terbium iron borate

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The Faraday effect induced by an external magnetic field in $TbFe_3(BO_3)_4$ and $TbAl_3(BO_3)_4$ borates at a wavelength 633 nm has been investigated. It was found that the terbium subsystem brings the dominant magnetic contribution to the Faraday rotation at low temperatures in borate $TbFe_3(BO_3)_4$. For both $TbFe_3(BO_3)_4$ and $TbAl_3(BO_3)_4$ the magneto-optical coefficients of the terbium subsystem were determined.

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1. Introduction

Rapidly developing scientific and technical fields such as spintronics and photonics require new magnetic and magneto-optical materials [1,2]. Currently, garnet films ordinarily are used for magneto-optical applications [3]. It is possible that the borates can compete with garnet in this area.

Recently the family of borate crystals with general formula $RM_3(BO_3)_4$ (M = Al, Ga, Sc, Cr, Fe) doped with rare-earth ions R for use as magneto-optic materials has received much attention among researchers due to the optimal optical properties of rare-earth ions in terms of losses and magnetic susceptibility [4]. The aluminum borates $RAl_3(BO_3)_4$, for example, can be used in lasers, as well as elements of nonlinear optics [5,6]. The noncentrosymmetric structure of $RFe_3(BO_3)_4$ borates makes these materials also an interesting candidate for optical applications based on their good luminescent and nonlinear optical properties [7,8].

All crystals of this family belong to a trigonal system with the space group $R32$. The replacement of Al by Fe changes significantly the magnetic behavior of the rare-earth borates, which may have an effect on the magneto-optical properties of such crystals. In some iron borates, such as $GdFe_3(BO_3)_4$ and $TbFe_3(BO_3)_4$ [9,10], the first-order structural phase transition from high-symmetry $R32$ to low-symmetry space group $P3_121$ was observed at temperatures of 156 and 192 K, respectively. Above Neel temperature ($T_N \approx 20$–40 K) the $RFe_3(BO_3)_4$ borates are paramagnetic. At Neel temperature, the iron subsystem orders antiferromagnetically in these borates [11]. At the same time the rare-earth subsystem remains paramagnetic below T_N. The magnetic moments of the rare-earth ions are polarized by the exchange coupling between the rare-earth and iron subsystems. Two types of magnetic anisotropy of the antiferromagnetic ordered state can be observed in the iron borates: the "easy plane" anisotropy for the compounds with Y, Nd, Er, and Tm and the "easy axis" one–with Tb, Pr, and Dy. The rare- earth iron borates with the "easy axis" anisotropy show spin-flop transitions in magnetic field $H \| c$ below T_N [10,12,13].

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It is well known that the Faraday rotation induced by an external magnetic field depends upon the magnetization and magneto-optical properties of the materials. The Faraday effect in $TbFe_3(BO_3)_4$ and $TbAl_3(BO_3)_4$ borates are investigated in this work. In an external magnetic field, the magnetization $M(H)$ of the $TbFe_3(BO_3)_4$ crystal ($T_N = 40$ K) can be represented as a sum of two contributions: the magnetization of the terbium subsystem $M_{Tb}(H)$ and the iron subsystem $M_{Fe}(H)$. Thus the rotation angle $\Phi(H)$ in magnetic field $H \| c$ can be described by the following expression:

$$\Phi(H) = (A \cdot M_{Tb}(H) + B \cdot M_{Fe}(H)) \cdot d, \qquad (1)$$

where A and B are the magneto-optical coefficients for the terbium and iron subsystems, respectively. d is the thickness of the sample.

In contrast to the terbium iron borate, the $TbAl_3(BO_3)_4$ crystal contains only one type of magnetic ions–the terbium subsystem. In external magnetic fields the magnetization and the Faraday rotation of this crystal are determined by the magnetization of terbium subsystem $M'_{Tb}(H)$. Therefore, the Faraday rotation $\Phi'(H)$ for $TbAl_3(BO_3)_4$ can be described as follows:

$$\Phi'(H) = A' \cdot M'_{Tb}(H) \cdot d, \qquad (2)$$

where A' is the magneto-optical coefficient for the terbium subsystem and d is the thickness of the sample.

The study of field dependences of Faraday rotation and magnetization of $TbAl_3(BO_3)_4$ and $TbFe_3(BO_3)_4$ borates can clarify the contributions of both magnetic subsystems to the Faraday rotation in the crystal $TbFe_3(BO_3)_4$ separately, due to the presence of the terbium subsystem as in $TbAl_3(BO_3)_4$ and $TbFe_3(BO_3)_4$ borates.

The purpose of this work is to study the Faraday effect and investigate how temperature variations influence the magneto-optic properties of both $TbAl_3(BO_3)_4$ and $TbFe_3(BO_3)_4$.

2. Experiment

Single crystal samples of $TbAl_3(BO_3)_4$ and $TbFe_3(BO_3)_4$ were grown by the method described in [10,14]. For the optical measurements, the samples were cut in the form of plane-parallel plates perpendicular to the c axis. The thicknesses of the samples were $d = 150$ and 110 μm for $TbAl_3(BO_3)_4$ and $TbFe_3(BO_3)_4$, respectively. To reduce the elastic stresses after mechanically polishing, the samples were annealed at 800°C for 10 h. Both samples have good transparent properties for the visible range of light.

The Faraday rotation as a function of the applied magnetic field was measured by using an experimental system by incorporating modulation and the synchronous detection technique. The sample was maintained in a cold finger of an optical helium cryostat in vacuum. The temperature was controlled by a resistance thermometer to within 0.1 K. A superconducting solenoid with $H_{max} = 55$ kOe was used as a source of magnetic field coinciding with the direction of the light propagation. A filament lamp with an interference filter ($\lambda_{max} = 633$ nm, FWHM = 11 nm) was used as a light source.

The field dependences of magnetization of both samples were measured with a Quantum Design SQUID magnetometer MPMS-XL5 in the temperature range 8–35 K and magnetic fields 0–50 kOe.

The magnetic field was directed along the c axis of crystals in both experiments.

3. Experimental Results

A. $TbFe_3(BO_3)_4$

The magneto-optical properties of $TbFe_3(BO_3)_4$ single crystal were measured in an external magnetic field parallel to the c axis of the crystal. Figure 1 shows the field dependences of Faraday rotation $\Phi(H)$ in $TbFe_3(BO_3)_4$ at fixed temperatures of 8, 10, 15, 20, and 35 K. As can be seen in Fig. 1(a), at the lowest temperature $T = 8$ K the curve $\Phi(H)$ has a more pronounced nonlinear character and this nonlinearity gradually diminishes with increasing temperature. At temperature close to $T_N = 40$ K, the curve $\Phi(H)$ becomes almost linear.

In addition to the optical measurements, the magnetization M as a function of the applied magnetic field H was measured for $TbFe_3(BO_3)_4$ by using the standard SQUID technique. The magnetic

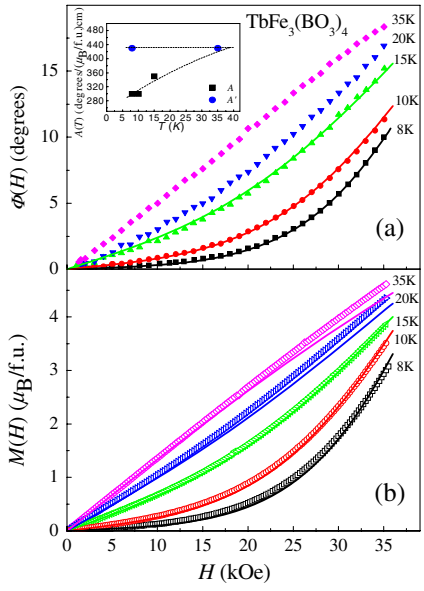

Fig. 1. (a) Faraday rotation as a function of the applied magnetic field $\Phi(H)$ and (b) the magnetization as a function of the applied magnetic field $M(H)$ in $TbFe_3(BO_3)_4$ single crystal at various temperatures for $H \| c$. The symbols are the experimental data; the lines are the curves calculated by using Eq. (1) for (a) and Eq. (5) for (b). The thickness of the investigated sample is $d = 110$ μm. Inset: the magneto-optical coefficients A for $TbFe_3(BO_3)_4$ and A' for $TbAl_3(BO_3)_4$ as a function of temperature.

experiments were performed at the same temperatures. Figure 1(b) shows the field dependences of magnetization $M(H)$ of the $TbFe_3(BO_3)_4$ borate for magnetic field $H \| c$. Evidently, the magnetization curves obtained are very similar to the results for the Faraday rotation experiments. At the low temperature the curves $M(H)$ have nonlinear character. At temperatures close to T_N the curves $M(H)$ become almost linear.

B. $TbAl_3(BO_3)_4$

We also consider the magneto-optical properties of $TbAl_3(BO_3)_4$ borate, which contains a single Tb magnetic subsystem. The optical and magnetic measurements of the $TbAl_3(BO_3)_4$ borate were performed under the same experimental conditions as for $TbFe_3(BO_3)_4$. Figures 2(a) and 2(b) show the field dependences of Faraday rotation and magnetization of $TbAl_3(BO_3)_4$ at $T = 8$ and 35 K and magnetic fields up to 35 kOe for orientation $H \| c$. As one would expect, the results obtained for magnetization are very similar to the results for the Faraday rotation experiments for $TbAl_3(BO_3)_4$. These dependences at $T = 8$ K demonstrate that the rotation angle of the polarization plane and magnetization of the $TbAl_3(BO_3)_4$ borate tend to a saturation value with increasing magnetic field. However, at 35 K the field dependences of Faraday rotation and magnetization do not tend to a saturation and demonstrate almost linear behavior.

4. Discussion

The magnetic properties of the terbium iron borate are governed by both magnetic subsystems interacting with each other. As mentioned above, the

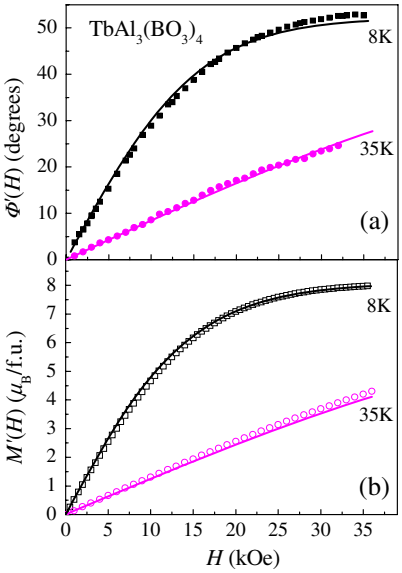

Fig. 2. (a) Faraday rotation as a function of the applied magnetic field $\Phi'(H)$ and (b) the magnetization as a function of the applied magnetic field $M'(H)$ in $TbAl_3(BO_3)_4$ single crystal at $T = 8$ and 35 K for $H \| c$. The symbols are the experimental data; the lines are the curves calculated by using Eq. (2) for (a) and Eq. (5) for (b). The thickness of the investigated sample is $d = 150$ μm.

Fe subsystem orders antiferromagnetically with magnetic moments along the c axis below T_N. The magnetic moments of the Tb subsystem are polarized in the same directions by the field of exchange interaction with the iron subsystem $H_{\text{Tb-Fe}}$. It is known that the ground multiplet of Tb^{3+} ions is $^7F_6$ ($S = 3, L = 3, J = 6$). In $TbFe_3(BO_3)_4$ this multiplet is split into $(2J + 1) = 13$ Stark levels by the crystal field [15,16]. The ground state of the Tb^{3+} ion is a quasi-doublet, and the energy interval between the ground quasi-doublet and the nearest excited states is of the order of 190 cm^{-1} [15,16]. Below T_N the ground quasi-doublet of the Tb^{3+} ions is split by the crystal field and exchange field $H_{\text{Tb-Fe}}$. Without an external magnetic field the energy gap between the levels of this quasi-doublet Δ can be determined by the following expression:

$$\Delta = \sqrt{\delta^2 + (g_{\text{eff}}\mu_B H_{\text{Tb-Fe}})^2}, \tag{3}$$

where δ is the splitting of the quasi-doublet of Tb^{3+} ions by the crystal field, the value of this splitting is ~0.2 cm^{-1} [16], μ_B is the Bohr magneton, and $g_{\text{eff}} \approx 17.5 \pm 0.3$ is the effective g-factor of the ground quasi-doublet [15,16].

The fact that the ground quasi-doublet of the Tb^{3+} ion is well separated from the excited one allows us to use the "quasi-doublet" approximation for estimation of terbium subsystem magnetization at $T < T_N$. Within this approximation the Tb^{3+} system should be divided into two subsystems, each characterized by the magnetic moment (per formula unit):

$$m_{i\text{Tb}}(H) = \frac{g_{\text{eff}}\mu_B}{4} \cdot \tanh\left(\frac{g_{\text{eff}}\mu_B(H \pm H_{\text{Tb-Fe}})}{2kT}\right), \tag{4}$$

where the sign ± before exchange field $H_{\text{Tb-Fe}}$ corresponds to two antiferromagnetic sublattices of Fe, and the subscript $i = 1, 2$ specifies the terbium subsystems. This expression does not take into account the splitting of the quasi-doublet by the crystal field δ. The full magnetization (per formula unit) of the terbium subsystem $M_{\text{Tb}}(H) = m_{1\text{Tb}} + m_{2\text{Tb}}$ in a magnetic field $H \| c$ will be determined by the following expression:

$$M_{\text{Tb}}(H) = \frac{g_{\text{eff}}\mu_B}{4} \cdot \left[\tanh\left(\frac{g_{\text{eff}}\mu_B(H + H_{\text{Tb-Fe}})}{2kT}\right) + \tanh\left(\frac{g_{\text{eff}}\mu_B(H - H_{\text{Tb-Fe}})}{2kT}\right)\right]. \tag{5}$$

In order to calculate $M_{\text{Tb}}(H)$ at temperatures 8, 10, 15, 20, and 35 K it is necessary to determine the value of $H_{\text{Tb-Fe}}$ at those temperatures. If δ is neglected in Eq. (3), then exchange field $H_{\text{Tb-Fe}} \approx \Delta/g_{\text{eff}} \cdot \mu_B$. Using the temperature dependence of Δ [15], $H_{\text{Tb-Fe}}$ can be estimated for different temperatures. The results of estimation are listed in Table 1.

It is not difficult to calculate the field dependence of $M_{\text{Tb}}(H)$ by Eq. (5), and compare it with the

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