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RADIO PHENOMENA IN SOLIDS AND PLASMA

Magneto-Optic Properties of Ultrathin Bismuth-Containing Ferrite-Garnet Films Obtained Using Radio-Frequency Magnetron Sputtering

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Abstract—Magneto-optic properties of ultrathin ferrite-garnet ($Bi_2Dy_1Fe_4Ga_1O_{12}$) films with layer thicknesses ranging from 0.6 to 100 nm are studied. The films are produced with the aid of the radio-frequency magnetron sputtering and subsequent high-temperature crystallization annealing. The magnetic circular dichroism is measured in the wavelength interval 300–650 nm. Relatively weak magneto-optic activity is revealed in the films with nominal thicknesses of 0.6 and 3.7 nm at room temperature but the measured spectrum does not correspond to the spectral dependence of the magnetic circular dichroism of bismuth-containing ferrite-garnets. The spectral dependence of the magnetic circular dichroism that is typical of bismuth-containing ferrite-garnets is observed in the film with a thickness of 10.3 nm and thicker films. The spectra of the magnetic circular dichroism that are typical of nanocrystallites of bismuth-containing ferrite-garnets are obtained in the measurements in the temperature interval 8–200 K.

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INTRODUCTION

Bismuth-containing ferrite-garnets are the most promising magneto-optic (MO) materials for applications in various MO devices, magnetic photonic crystals, and photonics. With respect to practical applications of ferrite-garnets, the most promising process involves the radio-frequency magnetron sputtering of amorphous films on a gadolinium–gallium-garnet substrate at room temperature with the subsequent annealing of such films using a high-temperature procedure in the temperature interval 550–650°C.

The properties of the substrate–film transition layer play a decisive role in such crystallization annealing of the ferrite-garnet films, since the crystallization processes in the structure under study are initiated in the transition layer [1].

To study the initial crystallization stage of the ferrite-garnet films, we employ a series of films with nominal composition $Bi_2Dy_1Fe_4Ga_1O_{12}$ and thicknesses of 0.6, 3.7, 10.3, 50, and 100 nm on the $Gd_3Ga_5O_{12}$ substrates with the substrate orientation in the (111) plane. The spectra of magnetic circular dichroism (MCD) are measured in the wavelength interval 250–650 nm. In the measurements in the temperature interval 200-8 K, we observe the MCD spectra that are typical of the nanocrystallites of bismuth-containing ferrite-garnets.

1. FABRICATION OF EXPERIMENTAL SAMPLES

A two-stage procedure is employed in the fabrication of the ultrathin ferrite-garnet films. At the first stage, the films with nominal thicknesses of 0.6, 3.7, 10.3, 50, and 100 nm are obtained using the radio-frequency magnetron sputtering of a hot-pressed $Bi_2Dy_1Fe_4Ga_1O_{12}$ target on the gadolinium–gallium substrate. The film thicknesses are calculated using the deposition times on the assumption that the deposition rate does not depend on the deposition time. The gadolinium–gallium garnet ($Gd_3Ga_5O_{12}$) serves as the substrate. A KVS-T4065 magnetron system is used for deposition of the ferrite-garnet films. Standard substrates with the substrate surface orientation in the (111) plane are employed.

In the course of the magnetron sputtering, the substrates are heated to a temperature of 250°C. The X-ray measurements of the films with a thickness of 50 nm (and thicker films with thicknesses ranging

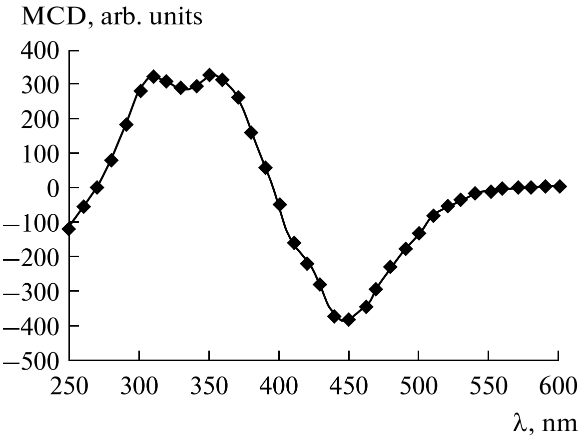

Fig. 1. MCD spectrum of the ferrite-garnet film with nominal composition $Bi_2Dy_1Fe_4Ga_1O_{12}$ and a thickness of 10.3 nm on the $Gd_3Ga_5O_{12}$ substrate (the sensitivity scale of the dichrograph is 5×10^{-6}).

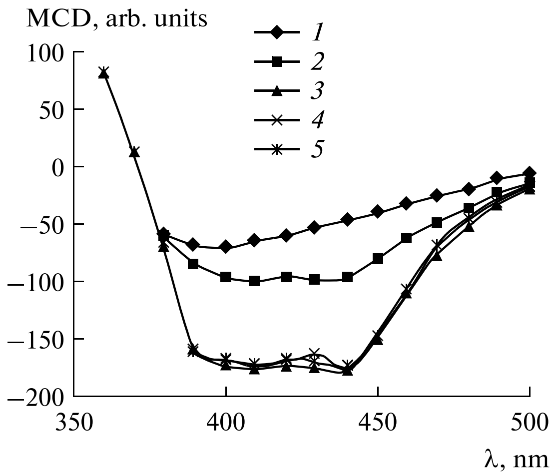

Fig. 2. MCD spectra of the ferrite-garnet film with nominal composition $Bi_2Dy_1Fe_4Ga_1O_{12}$ and a thickness of 3.7 nm on the $Gd_3Ga_5O_{12}$ substrate measured in the wavelength interval 360–500 nm at temperatures of (*1*) 200, (*2*) 150, (*3*) 100, (*4*) 50, and (*5*) 8 K (the sensitivity scale of the dichrograph is 5×10^{-6}).

from 50 to 100 nm) yield the amorphous state of the freshly deposited films. The films are deposited in argon atmosphere at a pressure of 1 mTorr, the radio-frequency power on the target is 4 W/cm^2, the deposition rate is 4 nm/min, the distance between the target and substrate is 18 cm, and the rotation rate of the substrate is 30 rpm.

At the second stage, the crystallization annealing of the films is performed in air over 1 h at a temperature of 620°C. The X-ray measurements of the films with thicknesses of 300 and 1000 nm that are crystallized under similar conditions show that the mean size of nanocrystallites is 40 nm [2].

2. MEASUREMENTS OF THE MCD SPECTRA AT ROOM TEMPERATURE

A Jobin-Yvon Mark-IV dichrograph is used in the measurements of the MCD spectra. The measurements are performed at room temperature in the wavelength interval 300–650 nm. The films with thicknesses of 10 and 50 nm exhibit MCD spectra that are typical of bismuth-containing ferrite-garnets with the above composition [1, 2]. Figure 1 shows the MCD spectral curve for the sample with a thickness of 10.3 nm. The observed signs of the effect indicate that the magnetic compensation point of the material is above room temperature [1].

The MCD spectra of the samples with thicknesses of 0.6 and 3.7 nm on the gadolinium–gallium-garnet substrates that are measured at room temperature yield a low-intensity peak at about 360 nm the spectral dependence of which disagrees with the known MCD spectra of the bismuth-containing ferrite-garnets [1].

The observed MCD signal can be related to the presence of the magnetite nanocrystallites that are formed due to the loss of bismuth ions in the course of film deposition and subsequent crystallization annealing of ultrathin films. Note that the crystallization of the magnetite nanocrystallites is started at a temperature of about ~250°C [3].

3. MEASUREMENTS OF THE MCD SPECTRA AT LOW TEMPERATURES

To obtain additional data on the composition of the film-substrate transition layer and estimate variations in the composition of the film with respect to thickness, we measure the MCD spectra of the sample with a thickness of 3.7 nm at low temperatures ranging from 8 to 300 K (Fig. 2).

The experimental results show that the crystallization annealing at a temperature of 650°C does not lead to the total loss of bismuth even in the film with such a small thickness. The spectral dependences of the MCD are measured at temperatures of 200, 150, 100, 50, and 8 K. The negative MCD effect in the spectral interval 370-550 nm and the experimental shapes of spectral dependences at temperatures from 150 to 8 K (MCD peaks at 445 and 390 nm) are typical of the bismuth-containing ferrite-garnets with a relatively high content of the Ga^{3+} ions in the tetrahedral lattice of iron.

The negativeness of the MCD effect in the spectral interval 550–375 nm indicates that the magnetic moment of the octahedral sublattice of ferrite-garnet is oriented along the direction of the external magnetic field.

However, an alternative interpretation is needed for the spectral dependence of the MCD at a temperature of 200 K. A broad peak at about 400 nm can be related to the crystallization of the magnetite (Fe_3O_4) nanocrystallites [4].

The MCD signal level and shapes of spectral dependences at temperatures of no greater than 150°C point to the fact that the content of the Bi^{3+} ions in the ultrathin ferrite-garnet film under study is about one formula unit.

An unexpected result follows from an almost constant MCD signal level at temperatures ranging from 100 to 8°C. Based on the data of Fig. 2, we may conclude that the Neel temperature of the ferrite-garnet nanocrystallites is about 170 K.

The coincidence of the MCD curves and signal levels at temperatures of 100, 50, and 8 K shows that the part of the substrate–film transition layer contributing to the MCD signals at temperatures of no greater than 100 K has constant composition and the structural formula is $Bi_1Dy_2Fe_{2.5}Ga_{2.5}O_{12}$ with allowance for the above temperature of magnetic ordering.

This result is unusual, since a gradual variation in the composition of film in the transition layer is intuitively expected.

A possible interpretation is as follows: at the initial stage of the deposition of the ferrite-garnet film, the bombardment of substrate with the Ar^+ ions leads to the amorphization of substrate to a depth of 3 nm. In the course of the crystallization annealing in this region, the accelerated diffusion leads to the homogenization of the film in the amorphization region and the content of the Ga^{3+} ions in ferrite-garnet amounts to 2.5 formula units.

CONCLUSIONS

The magneto-optic properties of ultrathin films of bismuth-containing ferrite-garnets that are obtained with the aid of the radio-frequency magnetron sputtering on the gadolinium–gallium-garnet substrates and the subsequent crystallization annealing have been studied in the temperature interval from room temperature to 8 K. At room and liquid-helium temperatures, the spectral dependences of MCD are measured in the spectral intervals 650–250 and 650–360 nm, respectively. At room temperature, the MCD spectra typical of the bismuth-containing ferrite-garnets are obtained for the sample with a thickness of 10.3 nm.

For the films with thicknesses of 0.6 and 3.7 nm, the magneto-optic effects related to the bismuth-containing ferrite-garnet are not observed. At a temperature of 200 K, the magneto-optic activity in the film with a thickness of 3.7 nm is related to the magnetite nanocrystallites. At a temperature of no greater than 150 K, the MCD spectrum can be related to the $Bi_1Dy_2Fe_{2.5}Ga_{2.5}O_{12}$ nanocrystallites. This composition is determined using the measurements of the Neel temperature of the sample under study [5]. To estimate the bismuth content in the films we use the specific MCD signal. The calculated lattice parameter for the above composition ($a_f = 12.390$ nm) is close to the lattice parameter of the gadolinium–gallium-garnet substrate ($a_s = 12.383$ nm).

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