Giant Magnetorefractive Effect in La_{0.7}Ca_{0.3}MnO_3 Films


Abstract—Complex experimental investigations of the structural, optical, and magneto-optical properties (magnetotransmission, magnetoreflection, and transversal Kerr effect, as well as the magnetoresistance, of La_{0.7}Ca_{0.3}MnO_3 epitaxial films indicate that magnetoreflection and magnetotransmission in manganite films can reach giant values and depend strongly on the magnetic and charge homogeneity of the films, their thickness, and spectral range under investigation. It has been shown that the optical enhancement of the magnetorefractive effect is most pronounced in thin films as compared to manganite crystals. In the region of the minimum of the reflectance near the first phonon band, the resonance-like magnetorefractive effect has been observed, which is accompanied by change of the sign of the magnetoreflection. A model based on the theory of the magnetorefractive effect has been proposed to qualitatively explain this behavior.

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1. INTRODUCTION

The possibility of the high-speed control of the propagation and polarization of light is actively investigated in view of the development of the optical methods of the transmission and processing of information, holography, optical computers, contactless sensors, etc. The highest performance speed is inherent in magnetic materials in which the reorientation of the magnetic moment or magnetization can occur in times certainly shorter than nanoseconds. The efficiency of the high-speed control of the propagation direction and modulation of light by means of a magnetic field and magneto-optical effects, in particular, using magnetophotonic crystals was confirmed experimentally [1].

However, traditional Faraday and Kerr magneto-optical effects are small because of their relativistic origin [2]. For this reason, it is necessary to seek new magneto-optical effects and corresponding functional materials for solving this problem. One of such promising effects is the magnetorefractive effect, which is a magnetic field-induced change in the reflection and transmission (absorption) of natural light in magnetic materials having large magnetoresistance [1, 3, 4]. In contrast to traditional magneto-optical phenomena, the magnetorefractive effect is not attributed to the spin–orbit interaction; hence, the magnetoreflection and magnetotransmission of light due to the magnetorefractive effect can reach giant for magnetooptics values of 10–20% [3, 4]. The magnetorefractive effect is most pronounced in manganites with the colossal magnetoresistance [5–14], because the magnetoresistance in manganites is sufficiently high and their electrical properties are sensitive to an external magnetic field so that the conductivity type can even change. In view of this circumstance, it is difficult to describe the effects of magnetoreflection and magnetotransmission of light in manganites, particularly in films, because the developed theory of the magnetorefractive effect is applicable only for metallic [15] and tunneling conductivities [16] and ignores the effects associated with magnetic field-induced change in the relation of various magnetic phases, concentration of charge carriers, absorption edge, effective polaron mass [10, 17], etc. The experimental data for manganite films are contradictory, scarce, and exist either only for magnetoreflection (see, e.g., [7, 14]) or for magnetotransmission (see [11–13] and references therein) often in a limited range of wavelengths, fields, and temperatures without comparison with the data for crystals of the same composition and analysis for films of various thicknesses. Recently, we carried out complex investigations of magnetoreflection in La_{0.7}Ca_{0.3}MnO_3 crystals [10] and showed that magnetoreflection in this case near the Curie temperature is the optical response of colossal magnetoresistance. That work was a necessary stage of the study of the magnetoreflection and magnetotransmission of the manganite films of the same composition, which is the aim of this work. In this work, it is shown that the thickness of the films, as well as their inhomogeneity caused apparently by both nonstoichiometry in oxygen and strain stresses, strongly affects the spectra, magnitude, and even sign
of the studied magneto-optical phenomena concerning the reflection and transmission of light. The optical enhancement of the magneto-optical effects in films due to multiple passage of light through a film leads to giant values of the magnetoreflection and magnetotransmission reaching 10–20%. Moreover, the investigation demonstrates that significant magnetoreflection and magnetotransmission of light in manganites can be reached near optical resonance. This makes it possible, on the one hand, to explain the presence of the magnetorefractive effect in the visible spectral range for some compounds [5–7] and, on the other hand, to strongly expand the spectral range of the possible applications of the magnetorefractive effect, whereas it was previously [3, 4] thought that this range is reduced to the near infrared range.

2. SAMPLES AND EXPERIMENTAL CONDITIONS

We investigate the La$_{0.7}$Ca$_{0.3}$MnO$_3$ compound whose physical properties are known in detail; this circumstance facilitates comparative analysis of the nature of the magnetorefractive effect in films and crystals [10]. Thin epitaxial La$_{0.7}$Ca$_{0.3}$MnO$_3$ films were grown by the method of chemical vapor deposition from the vapors of metalloorganic compounds (MOCVD) on LaAlO$_3$ (001)$_{\text{cub}}$ single crystal substrates as was described in [18]. A series of 50, 180, and 320 nm thick films were prepared under the same experimental conditions. The films after the deposition were annealed in an oxygen flow for an hour at a temperature of 800°C. X-ray diffraction patterns (see Fig. 1) indicate that the films contain only one phase, highly oriented manganite La$_{0.7}$Ca$_{0.3}$MnO$_3$ in the (001)$_{\text{cub}}$ orientation. The lattice parameter determined from the X-ray diffraction pattern shown in Fig. 1 is $c = 0.3857(1)$ nm. The FWHM of the rocking curve of the (002)$_{\text{cub}}$ reflection of the film is 0.11°. The $\Psi$ scans of the (110)$_{\text{cub}}$ reflections of the LaAlO$_3$ substrate and La$_{0.7}$Ca$_{0.3}$MnO$_3$ film (see Fig. 1) demonstrate a high epitaxial quality of the film with the cube-on-cube growth.

The thickness of the La$_{0.7}$Ca$_{0.3}$MnO$_3$ films is comparable to the thickness of the skin layer; for this reason, we determined the effective Curie temperature $T^*_C$ using the transversal Kerr effect, which is the most convenient method for determining the magnetic-ordering temperature of thin films and surface structures. The Kerr effect was investigated at the angle of incidence of light of 67° in the energy range from 1.5 to 4.2 eV, the temperature range from 30 to 350 K, and in magnetic fields up to 3.5 kOe. The relative change in the intensity of the $p$-polarized light reflected from the sample, $\delta = (I_H - I_0)/I_0$, where $I_H$ and $I_0$ are the intensities of the reflected polarized light with and without magnetic field, respectively, was measured by the method described in [19].

The optical properties of the films were studied with a highly sensitive infrared spectrometer in the wavelength range of 0.8 $\mu$m $\leq \lambda \leq$ 27 $\mu$m at temperatures from 80 to 380 K. The design of the spectrometer allows the measurement of the light reflectance in magnetic fields up to $H = 3.5$ kOe for the field orientation along the surface of the sample and the light transmittance in magnetic fields up to $H = 10$ kOe for the field orientation perpendicular to the surface of the sample. The relative error of the determination of the transmittance and reflectance of light was 0.1%.

The light reflectances $R$ of the films were determined with respect to the reflectance from the Al mirror ($R = I/I_{Al}$, where $I$ and $I_{Al}$ are the intensities of light reflected from the sample and Al mirror, respectively) for the nearly normal incidence ($\sim 7^\circ$) of natural light. The sample and Al mirror were in one plane.

![Graph](image_url)