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MAGNETISM

Magnetoreflection and Magnetooptical Kerr Effect in $La_{2/3}Ba_{1/3}MnO_3$ Films at Room Temperature

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Abstract—Results of studies of magnetooptical Kerr effect and magnetoreflection of natural light in $La_{2/3}Ba_{1/3}MnO_3/SrTiO_3$ films of different thickness are presented. The Kerr effect was shown to be the most prominent in visible and near IR range; magnetoreflection was found to achieve its maximum of about 10% in the mid-IR range near the room temperature. Physical mechanisms defining the value and sign of the effects and the influence of the thin-film state on the magnetooptical properties are discussed. Magnetoreflection is estimated in the framework of the magnetorefractive effect theory.

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1. INTRODUCTION

Optical properties of magnetized media are investigated in polarized (Faraday, Cotton–Mouton, Kerr effects; see [1–3]) and in natural light (magnetoreflection and magnetoabsorption effects [4, 5]). A special consideration in studies of magnetooptical effects is given to concentrated magnetic semiconductors, whose optical properties can be controlled by magnetic and electric fields, temperature, and pressure. This class of materials includes manganites, which possess colossal magnetoresistance (CMR) and high-frequency response to the CMR—magnetotransmission and magnetoreflection of light. Physical phenomena underlying these effects in manganites in visible and infrared (IR) ranges were revealed; conditions for the maximal values of the effects were found (see review [6] and references therein). It was shown that the value and spectral dependence of the magnetoreflection can be influenced by surface and bulk defect levels in single crystals, films, and thin-film structures, as well as interfacial and resonance effects [6].

In most doped manganites with CMR the magnetoreflection effects were observed at temperatures below room temperature, which limits their possible practical applications. Authors are aware of some research [7–9] in which magnetoreflection of IR radiation in manganites near 300 K and in weak magnetic fields was studied. Magnetoresistance in $La_{2/3}Ba_{1/3}MnO_3$ is known to attain its maximum at $T_R \sim 310$–338 K. Authors of [10] revealed magnetotransmission of ~10% in natural light in a $La_{0.85}Ba_{0.15}MnO_3$ single crystal in the field of $H \sim$ 8 kOe and $T \sim 200$ K. Thus, we can suggest the presence of a noticeable magnetoreflection effect in the IR range in the optimally doped manganite $La_{2/3}Ba_{1/3}MnO_3$ at the room temperature.

In this paper we present original results of studying of magnetoreflection in unpolarized light compared to the data on equatorial Kerr effect in $La_{2/3}Ba_{1/3}MnO_3$ epitaxial films with the Curie point above room temperature.

2. EXPERIMENTAL

Epitaxial films of $La_{2/3}Ba_{1/3}MnO_3$ with the thickness of (1) $d = 80$ nm and (2) 110 nm were grown on single-crystal $SrTiO_3$ (001) substrates ($a_0 = 3.905$ Å) with the substrate temperature of $T = 730$ K and the oxygen pressure of $P = 0.4$ mbar by means of laser ablation (excimer pulsed ArF laser with $\lambda = 247$ nm was used). Film thickness was determined from the exposition time. Pressured tablet of polycrystalline $La_{2/3}Ba_{1/3}MnO_3$ ($a_0 = 3.909$ Å) with the diameter of 12 mm was used as a target. The films were annealed in the oxygen flow at the atmospheric pressure and $T =$ 730 K to achieve a stoichiometric fraction of oxygen. Analysis of the topology and magnetic structure of the films was conducted with the aid of a SolverNext automated scanning probe microscope (made by NT-MDT) (Fig. 1a). Mean roughness of the film surface was ~25 nm, which is larger than that for thin epitaxial films grown by magnetron sputtering [11]. The islet formation was also observed, which is typical of this method and small thicknesses. Structural quality

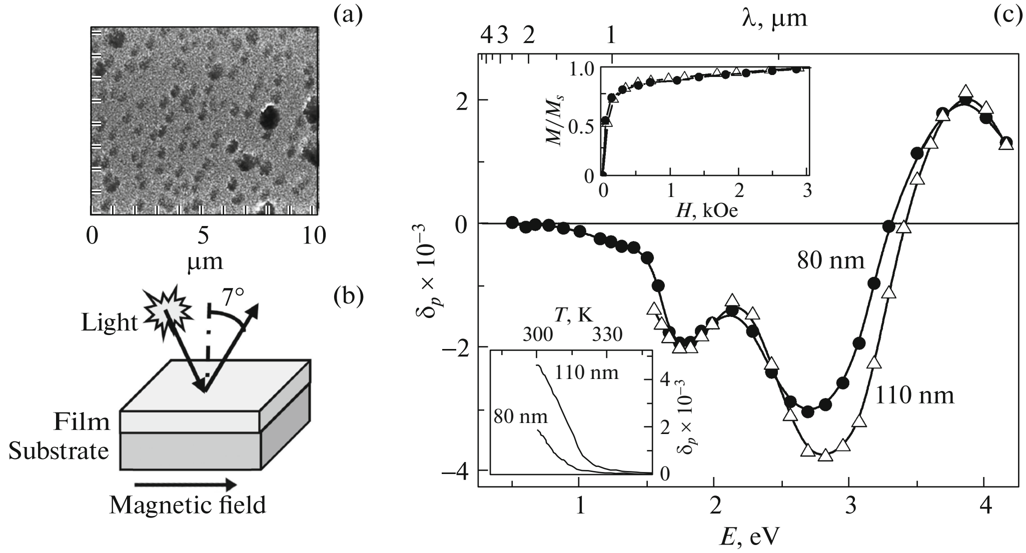

Fig. 1. (a) Magnetic-force microscopy (the size of the scanned region is 10 μm), (b) scheme of the experiment for measuring the magnetoreflection, and (c) spectra of the equatorial Kerr effect δ_p for the $La_{2/3}Ba_{1/3}MnO_3$ films at $T = 295$ K and $H = 2.5$ kOe. Upper inset: field dependences of the relative magnetization of the films M/M_s at $T = 295$ K and $E = 2.7$ eV. Bottom inset: temperature dependences $\delta_p(T)$ for the films at $E = 2.7$ eV.

of the films was attested by a Dron-4M X-ray diffractometer. The epitaxial character and the presence of a single phase were proved by clear reflexes of manganite and the substrate and by the absence of additional reflexes after the annealing procedure. The lattice parameter for the films was $a_0 = 3.97$ Å; it was independent of the film thickness. This value is close to that of the target.

An equatorial Kerr effect was measured at the angle of incidence of 68° in the energy range from 1.5 to 4.2 eV, temperature range from 30 to 350 K, and in static magnetic fields of up to 3.5 kOe. The relative change in the intensity of *p*-polarized light reflected from the sample was measured: $\delta_p = (I_H - I_0)/I_0$, where I_H and I_0 are the intensities of reflected light with and without the magnetic field [12]. The specular reflection coefficient was defined as $R = I_S/I_{Al}$, where I_S and I_{Al} are the intensities of light reflected from the sample and from an aluminum mirror, respectively. Magnetoreflection $\Delta R/R_0 = (R_H - R_0)/R_0$ was measured in magnetic fields up to 4 kOe directed along the sample surface and at the incidence angles of light of ~7°, in the IR range of wavelength from 0.8 to 30 μm, in the temperature range 200 K $\leq T \leq$ 360 K (Fig. 1b). Field and spectral dependences of $\Delta R/R_0$ were measured at the temperatures corresponding to the maximal values of the effects. The relative error in finding the reflection and magnetoreflection coefficients was ~0.2%. Electro- and magnetoresistance $\Delta\rho/\rho_0 = (\rho_H - \rho_0)/\rho_0$ of the films (where ρ_H and $\rho_0$are the specific resistances with and without the magnetic field) was measured by the two-contact method using direct current in the fields up to 8 kOe.

3. RESULTS AND DISCUSSION

3.1. Equatorial Kerr Effect

Compared to bulk samples, investigation of magnetic properties of thin films is complicated by a significant contribution from the diamagnetic substrate. The Kerr effect is the most convenient method of study in this case. The thickness of the skin layer becomes an important parameter, since the behavior of magnetization of the sample is defined on the scale of this layer. At the wavelength of λ ~ 0.6 μm, the skin layer thickness $s = (2\rho/\mu\mu_0\omega)^{1/2}$ in the $La_{2/3}Ba_{1/3}MnO_3$ films is 160 nm in the paramagnetic state ($T > T_C$) and 60 nm in the ferromagnetic state ($T < T_C$), which is comparable to or less than the thickness of the samples. Thus, behavior of magnetization inside the film can be analyzed based on magnetooptical data and the substrate can be considered not to contribute to the Kerr effect.

Temperature dependence $\delta_p(T)$ in the $La_{2/3}Ba_{1/3}MnO_3$ films comprises an abrupt increase in the intensity of reflected polarized light near the Curie point (inset in Fig. 1). The effective Curie point $*T_C$ of the films was defined from the position of the negative extremum of the first derivative of the $\delta_p(T)$ dependences. Analysis of experimental data gave the values of $*T_C = 302$ K for film 1 and $*T_C = 310$ K for film 2. This definition of $*T_C$ is due to the presence of a weak temperature hysteresis of $\Delta_p(T)$ in the vicinity of the magnetic phase transition. The $*T_C$ values of the studied films are close to the data for films of the same composition [11] and are 15–20 K lower that T_C for single crystals [10]; this can be attributed to the peculiarities of the film state, e.g., influence of epitaxial strains in the films [13, 14]. As the film thickness grows, the value of the Kerr effect and $*T_C$ increase (Fig. 1). This can be explained by relaxation of the epitaxial strain and an increase in the relative volume of the ferromagnetic phase in the film. A similar conclusion was made for manganite films of other compositions [14, 15].

Hysteresis and the absence of saturation for $\delta_p(T)$ at decreasing temperatures are most probably due to the presence of magnetic inhomogeneities in the films in paramagnetic and ferromagnetic states. Figure 1a shows the data of magnetic-force microscopy of the 80-nm-thick film at the room temperature; they may be interpreted as the presence of magnetic inhomogeneities comprising ferromagnetic (light islets) and paramagnetic (dark islets) regions. Similar results on magnetic phase separation were reported also for other manganites with CMR [15, 16].

The $\delta_p(E)$ spectrum for $La_{2/3}Ba_{1/3}MnO_3$ films (Fig. 1c) at T = 295 K is close to the spectra of $La_{1-x}Ba_xMnO_3$ single crystals [10] and doped manganites with CMR (see [6, 17] and references therein). At ~3.5 eV the spectra of $\delta_p(E)$ are defined by electric dipole transitions with charge transfer in octahedral complexes $[MnO_6]^{9-}$. At ~2 eV the main contribution is from spin-allowed transitions in Mn ions of different valence state. Threefold decrease in the intensity of the negative extremum at ~2.7 eV as compared to the data for δ_p in single crystals [10] and nonzero value of δ_p between the bands at 2.7 and 1.5 eV are also related to the magnetic inhomogeneity of the films. This conclusion is supported by the changes in $\delta_p(E)$ and $*T_C$ upon increasing the film thickness. Analogous behavior was observed for $\delta_p(E)$ and $*T_C$ in $La_{0.8}Ag_{0.1}MnO_3$ films of different thickness [18]. At the energies E < 1.5 eV the Kerr effect tends to zero. This behavior is common for manganites with CMR [17] independently of the doping cation type; it is also an important fact for comparison with the data on magnetoreflection.

Field dependences of the relative magnetization of the films $M/M_s = \delta_p(H)/\delta_{sp}(H)$ (where $\delta_{sp}(H)$ is the value of the Kerr effect at H = 3.5 kOe and E = 2.7 eV; see inset in Fig. 1c) show a typical of soft-magnetic materials sharp increase in the magnetization in weak fields and saturation at H > 500 Oe. The same behavior of magnetization was observed in $La_{0.7}Ba_{0.3}MnO_3$ single crystals [10], but at temperatures 10–15 K above than that for the thin-film samples studied in this work. In our opinion, this is also due to magnetic inhomogeneity of the films.

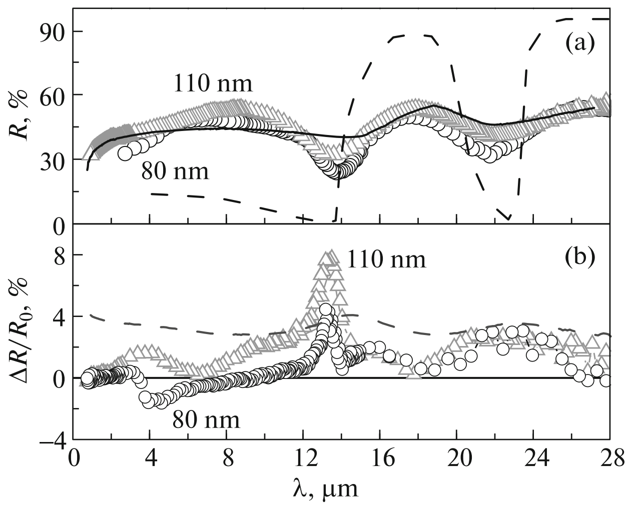

Fig. 2. Reflection R and magnetoreflection $\Delta R/R_0$ spectra for the $La_{2/3}Ba_{1/3}MnO_3$ films at T = 300 K and H = 3 kOe. (a) Dashed line shows the reflection spectrum of the $SrTiO_3$ substrate; solid line, reflection spectrum of a single crystal [10]. (b) Dashed line shows the calculated spectrum $\Delta R/R$ for the film with d = 110 nm.

Thus, the peculiarities in spectral, field, and temperature dependences of the Kerr effect in the studied films as compared to the data on single crystals of a similar composition are most probably due to magnetic and charge inhomogeneity caused by epitaxial strains and/or nonstoichiometric composition of sublattices.

3.2. Reflection and Magnetoreflection

Spectral dependence of the reflection coefficient R of the $La_{2/3}Ba_{1/3}MnO_3$ films at the room temperature (Fig. 2a) resembles that of the $La_{1-x}Ba_xMnO_3$ single crystal (x = 0.25) [10, 19]. At λ > 14 μm (0.09 eV) the shape of the dependence is defined by the interaction of light with lattice oscillations (phonons); at λ < 14 μm, by interaction of light with charge carriers. A decrease in R at wavelengths λ < 4 μm is most probably due to the plasma frequency ω_p (for example, ω_p = 1.5 eV for the $La_{1-x}Ba_xMnO_3$ single crystal (x = 0.25) at T = 95 K [10]).

The $R(\lambda)$ spectra of the films show prominent minima at λ ~ 14 and ~22 μm, and in the range $4 \le \lambda <$ 12 μm the reflection coefficient is larger than that of a single crystal (solid curve). The peculiarities in the $R(\lambda)$ spectra of the films arise due to an additional contribution to the reflection from the perovskite $SrTiO_3$ substrate (Fig. 2a) [14, 20]. A larger reflection coefficient for the films compared to that for the single crystal is also connected with reflection from the substrate and probably with a larger concentration of delocalized charge carriers in the film.

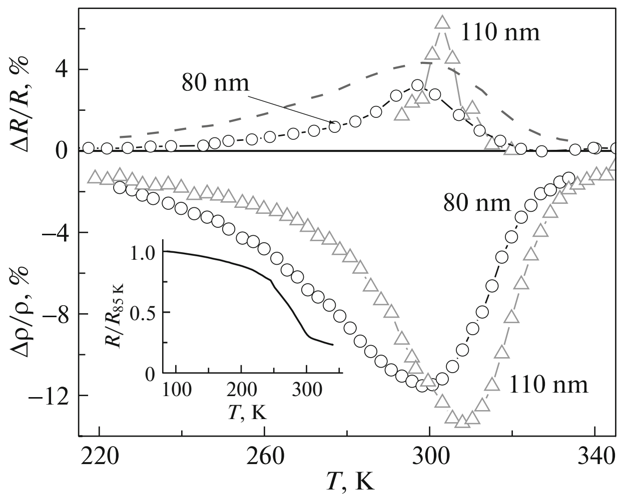

Fig. 3. Temperature dependences of the magnetoreflection $\Delta R/R_0$ at $H = 3$ kOe and $\lambda = 13.4$ μm and of the magnetoresistance $\Delta\rho/\rho_0$ at $H = 7.5$ kOe for the $La_{2/3}Ba_{1/3}MnO_3$ films. Dashed line represents the calculation following Eq. (1) for a film with $d = 80$ nm. The inset shows the temperature dependence of the reduced reflectance $R/R_{85\ K}$ for a film with $d = 80$ nm and $\lambda = 13.4$ μm.

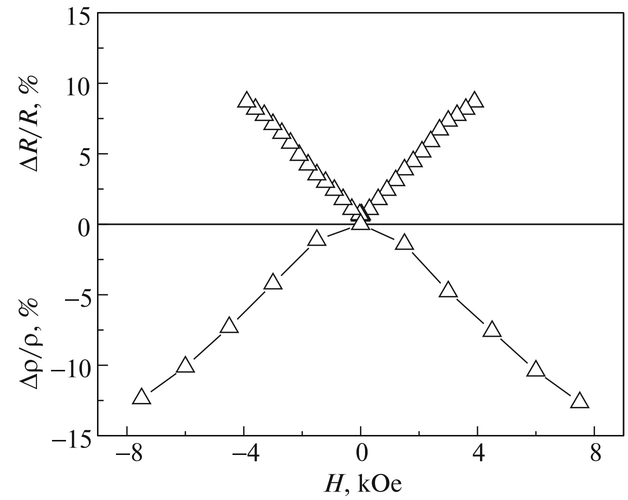

Fig. 4. Field dependences of the magnetoreflection $\Delta R/R_0$ and magnetoresistance $\Delta\rho/\rho_0$ for the $La_{2/3}Ba_{1/3}MnO_3$ film with the thickness of 110 nm at $T = 308$ K and $\lambda = 13.4$ μm.

Note that additional thermal treatment of the films in the oxygen atmosphere at the temperature of synthesis had negligible effect on the reflection spectra, which means that the nonstoichiometry in the anion sublattice is small.

As the temperature decreases ($T < T_C$), the reflection coefficient grows (inset in Fig. 3) which is caused by an increase in the contribution of free (delocalized) charge carriers. This effect is most prominent in the minimum before the phonon spectrum.

The external magnetic field leads to a change in the intensity of the reflected natural light and appearance of the magnetoreflection effect; the value of the latter, unlike the Kerr effect, is maximal in a relatively narrow temperature range near $^*T_C$. Magnetoreflection takes place in a wide IR spectral range from 1 to 28 μm (Fig. 2b) where the Kerr effect for manganites is negligible. Several ranges with typical features can be distinguished in $\Delta R/R_0$ spectra of the $La_{2/3}Ba_{1/3}MnO_3$ films at the room temperature: (i) the region of the plasma frequency and absorption edge at $\lambda \sim 1$ μm; (ii) near $\lambda \sim 3$ μm; (iii) near the reflection minimum before the first phonon band at $\lambda \sim 14$ μm; and (iv) near the minimum before the second phonon band at $\lambda \sim 22$ μm. Similar peculiarities in $\Delta R/R_0$ spectra were observed earlier in $La_{1-x}K_xMnO_3$ and $La_{0.7}Ca_{0.3}MnO_3$ films [9, 14]. We suggest that the feature (i) arises due to a shift of the plasma frequency in manganite induced by the magnetic field. The peculiarity (ii) is connected with a field-induced change in the density of electron states of the "narrow impurity band" at the energy of $E \approx 0.4$ eV (~3.1 μm). A similar band was observed in dielectric permittivity spectra of ε_{2xx} in a $La_{0.75}Ba_{0.25}MnO_3$ single crystal [10]. The features (iii) and (iv) are due to shift of the minima before phonon bands in the reflection spectra induced by the magnetic field [21, 22]. Physical phenomena underlying the features (iii) and (iv) were analyzed in [14]. It was shown that they are related to the influence of the magnetic field on the electron-phonon interaction and with the shift of phonon bands. Authors of [23] observed an increase in $\Delta R/R_0$ upon increasing the thickness of Fe_3O_4 films caused by the shift in the phonon band and appearance of a new phonon band under the influence of the magnetic field. In the case of our films, additional phonon bands were not observed in the magnetic field.

As the film thickness grows, the value of $\Delta R/R_0$ increases twice in the wavelength range $6 < \lambda < 14$ μm (Fig. 2b). A similar phenomenon was observed earlier in $La_{0.7}Ca_{0.3}MnO_3$ films [14], which was explained by a lesser charge and magnetic inhomogeneity of thick films due to relaxation of mechanical strains in the film/substrate system.

Temperature and field dependences $\Delta R/R_0$ for the $La_{2/3}Ba_{1/3}MnO_3$ films in the range of interaction between light and charge carriers correlate with the dependence of the static magnetoresistance (Figs. 3 and 4). This correlation manifests itself in the proximity of the maxima of the effects to the $^*T_C$ values for the films, in the likeness of the shapes and similarity of the band half-widths of the $\Delta R/R(T)$ and $\Delta\rho/\rho(T)$ dependences. The temperature behavior of $\Delta R(T)/R_0$ and $\Delta\rho/\rho(T)$ is defined by the suppression of magnetic moments fluctuations by the magnetic field, which are maximal near T_C (Fig. 3). Note that unlike magnetoreflection, the maximum of the Kerr effect is achieved at $T \ll {}^*T_C$ in the region of maximal magnetization of the sample (Fig. 1c).

Correlation of the temperature and field dependences of the magnetoreflection and static magnetoresistance can be explained in the theory of the magnetorefractive effect (MRE) that was introduced for manganites with CMR [6]. According to this theory, the ratio $\Delta R/R$ is determined to a first approximation by $\Delta\rho/\rho$ in the case of normal incidence of light as given by Eq. (1):

$$\frac{\Delta R}{R} = -\frac{1}{2}(1-R)\frac{\Delta\rho}{\rho} \qquad (1)$$

and should have a maximum provided that $\omega\tau \le 1$ [24].

Estimate of $\Delta R/R_0$ at the room temperature using Eq. (1), which did not allow for the peculiarities near the reflection minima before the phonon and plasma frequencies, yielded a satisfactory agreement with the experimental data. This means that the MRE theory is applicable to manganites of optimally doped composition (Figs. 2 and 3). Nevertheless, probably an additional mechanism influencing the value and sign of the magnetoreflection in manganites and related to the plasma frequency should be allowed for in the theory to describe $\Delta R/R_0$ in a more comprehensive way.

As was shown above, the Kerr effect undergoes saturation in weak fields since the film transforms to a state close to a single-domain one. Unlike the Kerr effect, $\Delta R/R_0$ is an even function of the magnetic field and does not show a hysteresis and saturation in the fields up to 8 kOe in different spectral regions (Fig. 4). This means that the peculiarities of the field dependence of $\Delta R/R_0$ in natural light are due to field-induced changes in only diagonal components of the complex dielectric permittivity; also this gives evidence that the odd components with respect to magnetization magnetooptical phenomena do not give a noticeable contribution.

CONCLUSIONS

Studies of magnetooptical effects in polarized (Kerr effect in the visible band) and natural (magnetoreflection in the IR band) light in $La_{2/3}Ba_{1/3}MnO_3$ films having thickness of 80 and 110 nm and the magnetic phase transition point close to the room temperature have proven that the Kerr effect tends to disappear when wavelength is larger than 1 μm and the magnetoreflection, on the contrary, increases. The Kerr effect achieves its maximum in the ferromagnetic region at temperatures significantly lower than the Curie point, which is due to the film magnetization process. At the same time, the magnetoreflection is the most prominent near the Curie point due to suppression of temperature fluctuations of magnetic moments by the external magnetic field. The Kerr effect is saturated in weak fields, since the films go to a state close to the single-domain one, whereas the magnetoreflection and magnetoresistance effects do not undergo saturation in fields up to 8 kOe.

The magnetoreflection effect in the $La_{2/3}Ba_{1/3}MnO_3$ films is defined by the influence of the external magnetic field on the position of the plasma frequency and the absorption edge, on the intensity of the localized states band, on the ratio of delocalized and localized charge carriers, and on the shift of the minima before the phonon bands in the light reflection spectra. An increase in the film thickness leads to an increase in the observed effects due to the relaxation of epitaxial (mechanical) strains in the film/substrate system and to an increase in the volume fraction of the ferromagnetic phase.

A satisfactory agreement of the experimental spectra and temperature dependences of the magnetoreflection with the data obtained within the magnetorefractive effect theory was achieved. However, the theory requires further development to allow for localized states and a contribution caused by a change in the plasma frequency.

The magnetoreflection effect in natural light in the $La_{2/3}Ba_{1/3}MnO_3$ films attains ~10% in the field of 4 kOe at the room temperature, which substantially exceeds by intensity the typical magnetooptical effects in the IR range. This makes the considered films a promising material for creating new functional devices and magnetooptoelectrical elements.

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