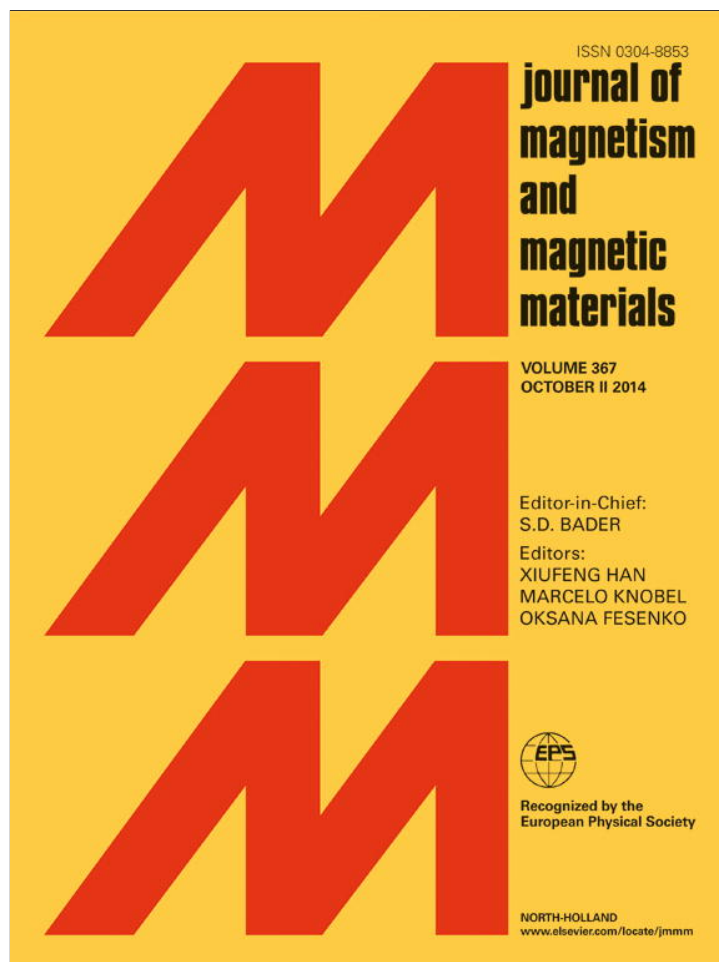


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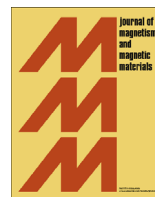
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journal homepage: www.elsevier.com/locate/jmmmMagnetorefractive effect in the $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ thin films grown by MOCVDYu.P. Sukhorukov^{a,*}, A.V. Telegin^a, V.D. Bessonov^{a,d}, E.A. Gan'shina^b, A.R. Kaul^b, I.E. Korsakov^b, N.S. Perov^b, L.Yu. Fetisov^b, A.N. Yurasov^c^a Institute of Metal Physics, Ural Division of RAS, 620990 Ekaterinburg, Russia^b Faculty of Physics, Moscow State University, Moscow 119991, Russia^c Moscow State Technical University of Radioengineering, Electronics and Automation, 119454 Moscow, Russia^d University of Bialystok, 15-424 Bialystok, Poland

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ABSTRACT

Thin epitaxial $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ films were grown using two-stage procedure. Influence of substitution of La^{3+} ions with K^+ ions on the optical and electrical properties of $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ films ($x=0.05, 0.10, 0.15$ и 0.18) has been studied in detail. A noticeable magnetorefractive effect in the films under study was detected in the infrared range. Magnetorefractive effect as well as transverse magneto-optical Kerr effect and magnetoresistance have the maximum in optimally doped sample with $x=0.18$ corresponding to the highest Curie temperature. The experimental data for compositions close to optimally doped films are in good agreement with the data calculated in the framework of a theory developed for manganites. The resonance-like contribution to magnetoreflexion spectra of manganite films has been observed in the vicinity of the phonon bands. It is shown that magnetic and charge inhomogeneities strongly influence on the magneto-optical effects in films. Thin films of $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ with the large values of Kerr and magnetorefractive effect are promising magneto-optical material in the infrared range.

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1. Introduction

After discovering the giant magnetoabsorption of light ($\sim 25\%$) in HgCr_2Se_4 spinels in 1988 [1], giant magnetotransmission in 1997 [2] and lately magnetoreflexion of light [3] in manganites with colossal magnetoresistance (MR) the investigations of mechanisms of interaction of unpolarized light with magnetic semiconductors in external magnetic fields started. It is important that absolute values of magnetoreflexion and magnetotransmission effects in manganites significantly exceed the linear magneto-optical Faraday and Kerr effects for a wide spectral range from middle infrared (IR) to ultraviolet one [3], which has been used for creation of modulators and contactless sensors, e.g. [4–7].

As it was shown earlier [8], regardless of the specific MR mechanism in manganites the simplest expressions in Eqs. (1) and (2) from [9] can be used for calculation of magnetoreflexion and magnetotransmission for $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ films for the normal incidence of light.

$$\begin{aligned} \Delta R/R_0 &= (R_H - R_0)/R_0 \\ &= -(1-R)\Delta\rho/\rho_0 k^2 \left[(3n^2 - k^2 - 1) / \left\{ (n^2 + k^2) \left[(1-n)^2 + k^2 \right] \right\} \right], \end{aligned} \quad (1)$$

$$\Delta T/T_0 = (T_H - T_0)/T_0 = 1/2 \Delta\rho/\rho_0 \cdot T k^2 (2n^2 + n)/(n^2 + k^2), \quad (2)$$

where $\Delta\rho/\rho_0 = [\rho_H - \rho_0]/\rho_0$ is magnetoresistance, R_H and R_0 , T_H and T_0 , ρ_H and ρ_0 are coefficients of reflection (R) and transmission (T) of unpolarized light and resistance (ρ) with and without magnetic field, respectively. This high-frequency response to magnetoresistance and its manifestation as magnetic field-induced changes in reflection and transmission coefficients of infrared light was called as a magnetorefractive effect (MRE) in magnetoreflexion and magnetotransmission modes, respectively.

The influence of the magnetic field (H) on the diagonal components of the dielectric permeability tensor ϵ_d and therefore on the refractive indices is described by the equation

$$\epsilon_d(\omega, H, T) = \epsilon' - i\epsilon'' = (n - ik)^2 = \epsilon_r(\omega, H, T) - i \frac{4\pi\sigma(\omega, H, T)}{\omega}, \quad (3)$$

where σ is a conductivity at frequency ω , temperature T and magnetic field (H); n and k are real and an imaginary parts of the complex refraction index. If the first term Eq. (3) characterizes the contributions of the displacement currents, electron-phonons interactions and so on to dielectric permeability, the second one is responsible for the magnetorefractive effect in manganites in the IR range.

The foregoing results of study of the magneto-optical and transport phenomena in optimally doped manganite crystals and films (see [3,10] and references therein) have proved that

* Corresponding author. Tel.: +7 343 3745154.

E-mail address: sukhorukov@imp.uran.ru (Yu.P. Sukhorukov).

magnetotransmission and magnetoreflexion are the consequences of colossal MR in the IR range and can be described in the framework of the magnetorefractive effect theory [8]. However, a large resonance-like contribution to magnetoreflexion ($\pm 20\%$ [10]) observed in $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ cannot be explained by the developed theory. This contribution is related with the displacement of the reflection minimum before the first phonon band in the reflection spectra of manganite in an external magnetic field. It should be also noticed that in the case of thin films it is necessary to take into account the effect of multiple reflection of light from the substrate that can increase the magnetoreflexion in films, unlike bulk crystals. In order to study the origin of the resonance-like contribution to magnetorefractive effect in manganites the complex investigations of single crystal and thin films with different thicknesses [10], levels and types of doping need to be completed to collect enough experimental data. One of the candidates are K^+ doped manganite films. Substitution of La^{3+} ions with K^+ one release twice as much charge carriers as in the case of Ca^{2+} doping ([11–15]). As a result $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ manganites possess both a high Curie temperature (T_C) above the room one and colossal MR for low K^+ concentration. Moreover, the influence of substitutions of La^{3+} ions with K^+ ions on MRE in manganite films has not been studied yet. Thus, the object of this paper is to study the $\Delta T/T_0$ and $\Delta R/R_0$ in $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ films in the IR range.

2. Sample preparation and experimental details

The $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ thin films with $x_K=0.05, 0.10, 0.15,$ and 0.18 and a thickness of $d=100$ nm were grown using the two-stage procedure described in detail in [11,16]. At the first stage the $\text{La}_x\text{MnO}_{3+\delta}$ epitaxial thin films were grown on single-crystalline (001) SrTiO_3 (STO) substrates ($a=3.905$ Å) by the metal-organic chemical vapor deposition (MOCVD) method. After deposition the films were annealed for 8 h in the flow of oxygen at $T=650^\circ\text{C}$. At second stage $\text{La}_x\text{MnO}_{3+\delta}$ films were enriched with K (isopiestic annealing). The process of K incorporation into the perovskite structure of $\text{La}_x\text{MnO}_{3+\delta}$ was controlled by using an energy-dispersive X-ray spectroscopy and by mass comparison with the ceramic target sample used as a standard. According to X-ray diffraction the $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ films were single-phase, highly oriented and had a rhombohedral perovskite structure (Fig. 1). The X-ray diffraction patterns indicate a small lattice mismatch (the parameters of the pseudo-cubic cell of the films and powders are differed in values less than 0.015 Å) and epitaxial growth of the films on the SrTiO_3 substrate. The inset in Fig. 1 shows the dependence of the unit cell parameters of $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ films on the potassium concentration. Incorporation of K^+ ions into the perovskite structure reduces the distance between planes of d (003) (reflex (003)) for the quasicubic $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ film from 1.2906 Å for $x_K=0.05$ to 1.2878 Å for $x_K=0.18$. Meanwhile, a large radius of K-cation must increase the unit cell volume and the d (003) distance. This feature can be connected with decreasing oxygen fraction and an increasing Mn^{4+} fraction or with the epitaxial strain at the film-substrate interface. Since accessible for oxygen ions interstices are not present in a perovskite structure, restricts the formation of additional vacancies of La and Mn ions that are being restricted by the oxidation process. The energy of La vacancies formation is lower and the mobility of La is higher than those for Mn vacancies [17]. To compensate the material and charge sites balance some defects have to be formed [18]. Thus, the oxidation of the films triggers the formation of cation vacancies and structural defects. As a result of isopiestic annealing the La vacancies fill with potassium and the structure of the film becomes non-equilibrium due to a process of slow relaxation.

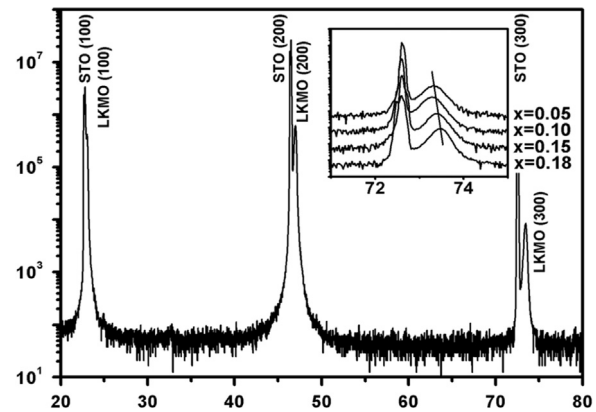


Fig. 1. X-ray diffraction patterns of the $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ films. The inset – the dependence of the position of (003) reflex on the potassium concentration.

There is a large difference in the vacancy relaxation rates at the surface (where vacancies are formed more easily) and in the bulk of the film. It leads to the difference in the physical properties of the surface and bulk of the $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ films. The comparative analysis of $\Delta\rho(T)/\rho_0$, $\Delta t(T)/t_0$ and $\Delta R(T)/R_0$ dependencies can reveal the degree of charge and magnetic inhomogeneity of manganite films.

The magnetic properties of the films were investigated using the vibration sample magnetometer and magneto-optical methods (Kerr effect and MRE). The transverse Kerr effect (TKE) was measured at an incidence angle 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 (Fig. 1). The relative change of the intensity of the p-polarized light reflected from the samples, $\text{TKE} = [I_H - I_0]/I_0$, where I_H and I_0 are the intensities of the reflected polarized light under and without magnetic field, respectively, that was measured by the method as described in [19]. The estimations of the skin layer thickness $\Delta = (2\rho/\mu\mu_0\omega)^{1/2}$ of the films have shown that in the paramagnetic (PM) state Δ changes from 180 to 450 nm, in the ferromagnetic (FM) state at wavelength $\lambda \sim 0.6$ μm the value of Δ varies from 60 to 110 nm depending on x_K . Thus, the thickness of the $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ films in the FM range (100 nm) is comparable or exceeds the skin-layer. It is a necessary condition for characterization of magnetic properties of the films by the TKE method. The skin-layer Δ is increased by more than 3 times when $\lambda \sim 6$ μm, thus making the investigation of the reflection and transmission spectra of unpolarized light in the manganite films possible. Due to the large change of absorption and reflection near T_C and a large absorption coefficient in the FM state an optical density (D) of the samples was measured.

$$D = \ln(Y_0/Y), \quad (4)$$

where Y_0 and Y are intensities of the incident and transmitted light, respectively. The reflectance (R) of the films was defined as $R = I_s/I_{Al}$, where I_s and I_{Al} are the intensities of light reflected from the sample and Al mirror, respectively.

Reflectance and transmittance of the light were measured under magnetic fields of up to $H=4$ kOe applied parallel to the sample surface (in-plane) and under the magnetic fields of up to $H=8$ kOe applied perpendicular to sample surface (out-of-plane), respectively. The optical density, reflectance and transmittance, magnetoreflexion and magnetotransmission were measured near the normal incidence of light in the wavelength range $0.8 \leq \lambda \leq 27$ μm at temperatures ranging from 80 to 380 K. The field and spectral dependences of $\Delta t/t_0$ and $\Delta R/R_0$ were measured at temperatures corresponding to maximum values of these effects. The relative errors of the determination of the reflectance and

transmittance, magnetoreflexion and magnetotransmission of light were less than 0.2%.

The electrical resistivity (ρ) was measured by the two-probe method under out-of-plane magnetic field of up to 8 kOe. Silver contacts were made on the films by In solder with using an ultrasonic iron.

3. Results and discussion

3.1. Kerr effect and magnetization

The temperature dependences of the Kerr effect and magnetization (M) (Fig. 2a,b) indicate the phase transition to the FM state in all $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ thin films below the room temperature. The effective Curie temperature ($*T_C$) was determined from the first-order derivative of the magnetization and TKE curves (see Table 1). The magnetic phase transition temperatures are lower than it could be predicted from the phase diagram because of poor quality of films and high defects value consequently. The increase of x_K leads to the growth of the FM phase volume and $*T_C$. One can notice the values of $*T_C$ for all films are by 40–70 K less than the data previously reported for polycrystals with the same doping level [20–23]. From the analysis of the $M(H)$ and $\text{TKE}(H)$ field dependences it was found that $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ films are soft ferromagnets with the coercive force $H_C \sim 60$ Oe and saturation fields $H > 200$ Oe (see the inset, Fig. 2c) and a weak magnetic anisotropy at $T=80$ K. The values of H_C are also increased by one order of magnitude compared to those for polycrystals [23].

The spectral behavior of Kerr effect for the films taken at $T=55$ K (Fig. 2c) is typical for doped manganites, e.g. by divalent ions like as for Sr^{2+} or Ca^{2+} or by univalent ions like as for Ag^+ (see [24–27] and references therein). However, small values of TKE indicate less volume of FM phase in the $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ thin films in comparison with $\text{La}_{1-x}\text{Ag}_x\text{MnO}_3$ or $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$ films. For $x_K=0.05$ the spectrum of TKE is similar to the $\text{La}_{0.95}\text{MnO}_{3+\delta}$ films, but the value of the one differs. It seems to be caused by a higher oxygen nonstoichiometry of $\text{La}_{0.95}\text{K}_{0.05}\text{MnO}_3$ film. For larger

x_K the TKE value is increased with a red-shift of a negative minimum that was earlier observed for manganites in [24,26]. It was shown that the TKE spectra for manganites are formed by the allowed electric-dipole transitions with charge transfer in the $[\text{MnO}_6]$ octahedral complexes and spin-allowed transitions in Mn ions of different valences [24]. The observed changes of TKE intensity, formation of the two bands and the red-shift of the minimum in spectra of the Kerr effect for the $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ thin films are probably related to their charge and magnetic inhomogeneity due to the two-stage method of deposition and high epitaxial strains at the film–substrate interface. Large influence of strains at the film–substrate interface on T_C and Kerr effect due to the stabilization of AFM phase and reduction of FM phase was found in La_xMnO_3 films as well [27].

3.2. Spectra of the reflectance and optical density

The spectra of the light reflection of $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ films, SrTiO_3 substrate and polycrystalline $\text{La}_{0.85}\text{K}_{0.15}\text{MnO}_3$ are shown in Fig. 3a. A large contribution of reflected light from the SrTiO_3 substrate changes the reflection spectra of films in contrast to the spectrum of the polycrystalline sample. Typically, in the FM state the reflectance is increased for manganites with colossal MR (for example, see [10,28]). It is explained by the contribution of free charge carriers and it is most visible in the minimum before the phonon spectrum of the film. An external magnetic field increases the intensity of the reflected light at $T \approx T_C$, thus giving rise to a positive magnetoreflexion effect.

The optical density spectra of the $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ thin films (Fig. 3b) are similar to those for manganite films in the IR range [10]. Spectra of $D(\lambda)$ are shaped by the absorption edge caused by the indirect interband transitions in the complexes $[\text{MnO}_6]^{9-}$ [29] at short wavelengths and by the MIR-bands. The MIR-band at $\lambda \sim 9 \mu\text{m}$ is formed by the intercenter transitions in hole complexes $[\text{MnO}_6]_{\text{JT}}^{8-}$ [24,30]. The absorption is small in the PM state and increases below T_C due to a large contribution from absorption of light by free charge carriers. The magnetic field increases the

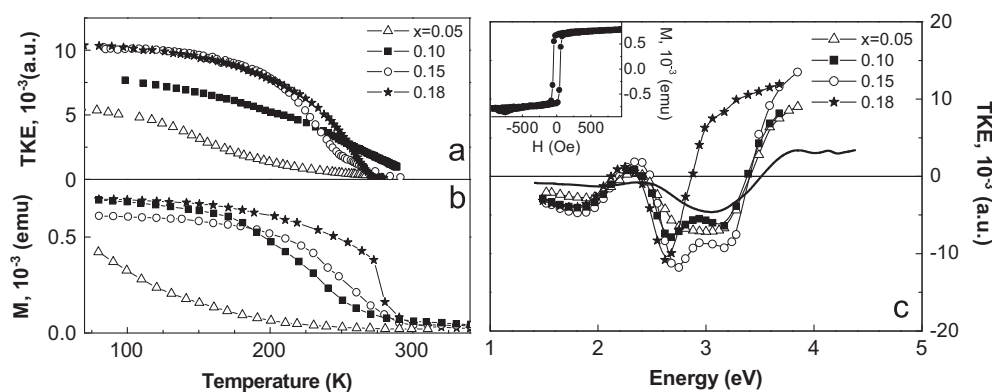


Fig. 2. Temperature dependences of (a) TKE at $E=2.65$ eV and $H=3.5$ kOe and (b) magnetization (M) in in-plane field $H=1$ kOe for $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ films, c) TKE spectra of $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ and $\text{La}_{0.95}\text{MnO}_{3+\delta}$ (solid line) films. On inset – the magnetic hysteresis for $x_K=0.15$.

Table 1

The effective Curie temperature ($*T_C$), the temperatures of maxima of magnetoreflexion ($^{max}T_{\Delta R/R}$), magnetotransmission ($^{max}T_{\Delta t/t}$), magnetoresistance effects ($^{max}T_{\Delta \rho/\rho}$) and $t(T)/t_{80}$, $R(T)/R_{min}$, $\rho(T)$ dependences for the $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ films on SrTiO_3 substrates. Magnetoreflexion ($\Delta R/R$) is taken for $\lambda=8.5 \mu\text{m}$ under the in-plane magnetic field 3 kOe, magnetotransmission ($\Delta t/t$) and magnetoresistance ($\Delta \rho/\rho$) for $\lambda=6 \mu\text{m}$ under the out-of-plane magnetic field 8 kOe.

$\text{La}_{1-x}\text{K}_x\text{MnO}_3/\text{SrTiO}_3$ (001)	$*T_C, \text{K}$	$^{max}T_{\Delta R/R}, \text{K}$	$^{max}T_{\Delta t/t}, \text{K}$	$^{max}T_{\Delta \rho/\rho}, \text{K}$	T^p, K	$\Delta R/R, \%$	$\Delta t/t, \%$	$\Delta \rho/\rho, \%$
$x=0.05$	150	190	180	200	210	1	-1	-14
$x=0.1$	240	245	230	230	240/250	2	-4.5	-8
$x=0.15$	230–250	255	255	255	260/270	5	-6	-19
$x=0.18$	260–280	275	275	275	280	6	-5.5	-19

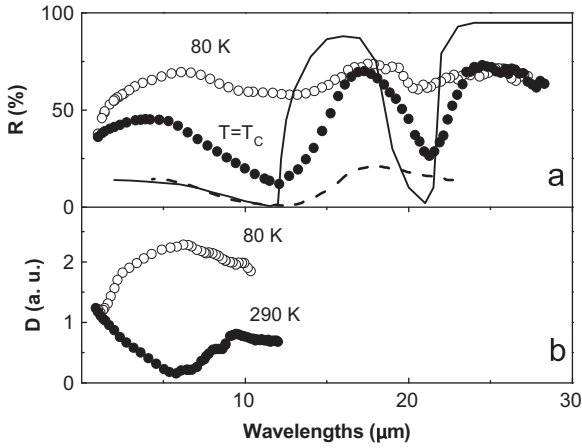


Fig. 3. The spectra of reflectance (R) of (a) $\text{La}_{0.85}\text{K}_{0.15}\text{MnO}_3$ film at $T=T_C$ and at $T=80\text{ K}$, dashed line – polycrystalline $\text{La}_{0.85}\text{K}_{0.15}\text{MnO}_3$, solid line – SrTiO_3 [31]; (b) optical density (D) for the $\text{La}_{0.85}\text{K}_{0.15}\text{MnO}_3$ film at two temperatures.

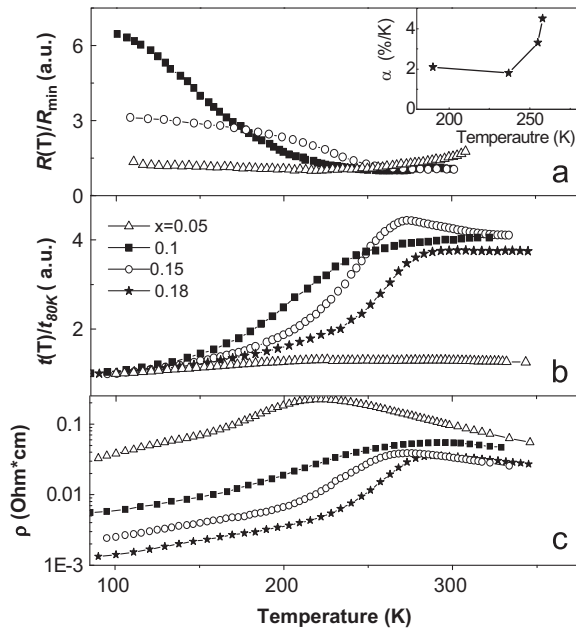


Fig. 4. The temperature dependences of (a) reflectance ($R(T)/R_{min}$) at $\lambda=8.5\ \mu\text{m}$, (b) transmittance ($t(T)/t_{80K}$) at $\lambda\sim 6\ \mu\text{m}$ and (c) the resistivity (ρ) of the $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ films. In inset the temperature dependence of thermal coefficient of resistance (α) for the film with $x_K=0$.

optical density at $T\approx T_C$ (magnetoabsorption) in the spectral range studied.

3.3. Temperature dependences of resistivity, reflection and transmission of light

The temperature dependencies of the relative values of reflected ($R(T)/R_{min}$) and transmitted ($t(T)/t_{80K}$) light clearly indicate the influence of magnetic order (PM–FM phase transition) on the process of interaction of light with the free charge carriers in the $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ films (Fig. 4). Note that the reflection was measured far away from the phonons using a wavelength of radiation $\lambda=8.5\ \mu\text{m}$, and the transmittance was obtained using a wavelength $\lambda=6\ \mu\text{m}$, where no influence of interband transitions on the absorption coefficient of the films can be detected. The optical and transport properties of the films correlate well. It appears in the similarity of the temperature dependence of

reflection and transmission of light, electrical resistivity and indicates the presence of the high-frequency (optical) response to the metal–insulator transition (for example, [3,24–26] and references therein) in the films as well as the proximity of T_C to the characteristic temperatures of the maxima of transmittance, electrical resistivity and minimum of the reflectance. These characteristic temperatures are shifted to the high-temperature region while x_K increases, which is caused by the increase in the FM metallic phase volume.

A thermal coefficient of the electrical resistance $\alpha=(1/\rho)(\delta\rho/\delta T)$ of the films can reach up to $\alpha\sim 4.5\%K^{-1}$ for $x_K=0.18$ at $T=T_C$. A large value of α for $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ is comparable with that for $\text{La}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$ [32] and can be used for creation of an infrared bolometer operating near the room temperature.

3.4. The magnetorefractive effect and colossal magnetoresistance

The spectra of magnetoreflexion (a) and magnetotransmission (b) for $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ films are shown in Fig. 5 for in-plane and out-of-plane magnetic field, respectively. When the volume of potassium rises up to $x_K=0.15$, the MRE increases both in the magnetoreflexion and magnetotransmission modes as it was for $\text{La}_x\text{MnO}_{3+\delta}$ [24]. The resonance-like contributions to magnetoreflexion effect were observed in the region of minimum of reflectance near the first phonon band for all the films. However, the highest magnetoreflexion (from +6% up to –10%) has been observed for the films with $x_K=0.15$, and 0.18. The data are shown in Table 1 without taking into account the resonance-like peaks. It is important that the intensive resonance-like peaks in the MRE ($\Delta R/R_0$) spectra were detected near the first and the second phonon bands from the reflection spectrum of the film with $x_K=0.18$ (Fig. 5a). As it was shown earlier [10], the resonance-like peak can be explained by shift of the position of the observed minimum before the first phonon band only in reflection spectrum under the magnetic field applied. In this experiment we showed this shift of minima in magnetic field is specific for other phonon bands. However, the physical origin of the resonance is still unknown. By analyzing the $\Delta R/R_0$ spectra of manganite films one can deduce two main reasons responsible for this peculiarity. The

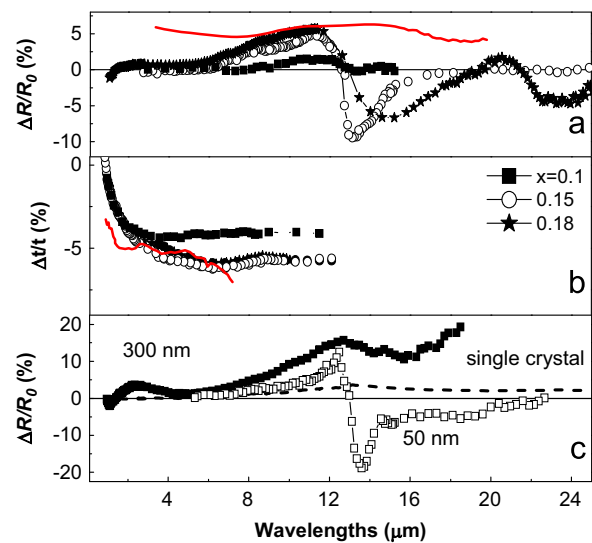


Fig. 5. The spectra of (a) magnetoreflexion ($\Delta R/R_0$) under the in-plane magnetic field $H=3\text{ kOe}$, (b) magnetotransmission ($\Delta t/t_0$) under the out-of-plane magnetic field $H=8\text{ kOe}$ for the $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ films (solid lines are the data calculated from Eqs. (1) and (2) for $x_K=0.15$), (c) magnetoreflexion ($\Delta R/R_0$) for single crystal and thin films of $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ with different thicknesses. All data were obtained at the temperature corresponding to the highest effect.

first reason may be related to the existence of tensile or compressive strains at the film-substrate interface which affects the electron-phonon interaction, magnetoelectric coupling [33] and so on, and depends on the external magnetic field. A small resonance contribution for single-crystals and thick films (for example, $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ on Fig. 5c) with small epitaxial strains [10] proves it. In our experiments the resonance-like peak is more typical for highly strained thin films (Fig. 5). The strains have a critical value when the film thickness becomes very small, which results in a change of (i) magnetoelastic [34] and (ii) magnetic properties [35–37], (iii) the temperature of the metal-insulator transition [38], (iv) the structure and transport properties [16,21,22,39] and, finally, (v) leads to the appearance of the resonance-like contribution (Fig. 5c). For examples, the authors of [40] showed an important role of a strong electron-phonon interaction in the formation of the phonon spectrum in manganites. Hence, the strains and magnetic field can influence the elastic constants and the phonon spectra of the films. For example, strains lead to the hardening of the phonon lines in thin films compared to a single crystal [41]. On the contrary, magnetic field causes softening of the phonon spectrum of both optical and acoustic phonons [42,43]. From the data of the BLS spectroscopy [42] it can be seen that the shift of the phonon band in the colossal MR manganites can reach up to 0.7% near the T_C at $H=3$ T. Probably, the same behavior of the phonon bands was observed in the reflection spectra of the $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ thin films.

The second reason that may be responsible for the large resonance-like contribution to magnetoreflexion is connected with the charge and magnetic inhomogeneities in the K^+ substitution films caused by the two-stage deposition procedure. The presence of such inhomogeneities should also lead to additional stresses in the films and can influence on the magnetoelastic constants and the phonon spectra of the films.

Note also, that the results of the calculations of magnetoreflexion in framework of the developed MRE theory (from Eqs. (1) and (2)) are agreed well with the experimental data for a single crystal, but not for the thin films of manganites [10], which can be explained by the additional contribution of the light reflected from the substrate. In this case, it is necessary to calculate MRE spectra in terms of the generalized Frenel's formulas. Nevertheless, there was no amplification of $\Delta R/R_0$ detected for the $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ films unlike Ca-substituted one.

If the magnetotransmission of the $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ films with the same thickness varies from 4.4% to 6.2%, the magnetoreflexion changes from 1.5% to 6% (Fig. 5). The difference is probably due to the high defectiveness of $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ thin film surface compared to a less defective volume of the film. The calculated (from Eqs. (1) and (2)) R/R_0 and $\Delta t/t_0$ data (solid lines on Figs. 6a and 6b) are close only to the experimental data for the films with $x_K=0.15$ and 0.18 (Fig. 5) and exceed experimental data for $x_K=0.10$ and 0.05. This fact indicates the presence of defects and magnetic/charge inhomogeneities in the volume and at the surface of the films too.

If magnetoreflexion and magnetotransmission effects are proportional to the volume of FM phase, the magnetoresistance reaches high value near the percolation threshold (for example, La_xMnO_3 films in [24]). Calculations of the temperature dependences of MRE according to Eqs. (1) and (2) (solid lines on Fig. 6) are in a good agreement with the experimental data for films with $x_K > 0.1$. However, for $x_K \leq 0.1$ there was no good correlation with the magnetoresistance. Taking into account the above results it was concluded that the theory of MRE developed for the manganites [8,44] may be applied only for chemical compositions that close to optimal doping. In our case it can be used for the films with $x_K > 0.1$, when the volume of a highly conductive FM phase tends to its maximum. To calculate the $\Delta R(T)/R_0$ and $\Delta t(T)/t_0$ for manganites with low doping level it is necessary to use the

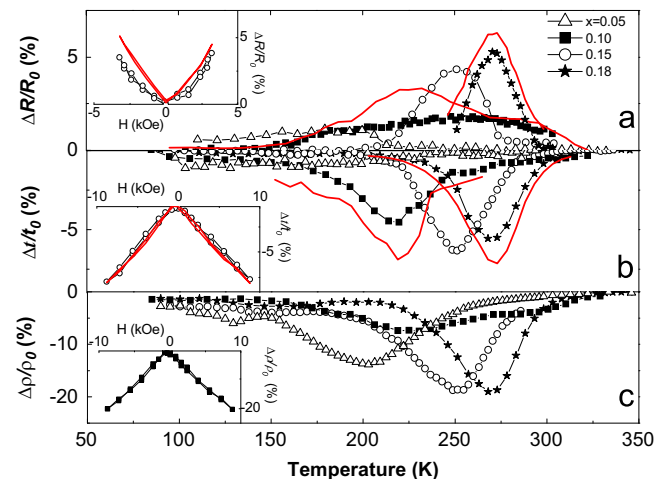


Fig. 6. The temperature dependences of (a) magnetoreflexion ($\Delta R/R_0$) for $\lambda=8.5$ μm under the in-plane magnetic field, (b) magnetotransmission ($\Delta t/t_0$) for $\lambda=6$ μm and (c) magnetoresistance ($\Delta\rho/\rho_0$) for $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ films under the out-of-plane magnetic field (solid lines were derived from the Eqs. (1) and (2) for $x_K=0.10$ and 0.18). The insets show the magnetic-field dependences of (a) $\Delta R/R_0$, (b) $\Delta t/t_0$ and (c) $\Delta\rho/\rho_0$ for $x_K=0.15$. The solid lines are the data calculated from Eqs. (1) and (2).

effective medium approach. For example, the spectra of $\Delta R(\lambda)/R_0$ and $\Delta t(\lambda)/t_0$ calculated in the framework of the effective medium approach (the form-factor $L=0.33$ for spherical particles or $L=0.2$ and 0.4 for ellipsoidal particles) had a good agreement with experimental data for manganites with a low doping level [45].

Moreover, there is another factor associated with charge and magnetic inhomogeneity of the $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ thin films. As it was shown earlier [24–26], the inhomogeneity of manganites leads to the weakening of MRE, smoothing of the $\Delta\rho(T)/\rho_0$ and $\Delta t(T)/t_0$ dependences or appearance of extra peaks. The same features are shown in Fig. 6a for $\Delta R(T)/R_0$. Moreover, the surface layer defects can lead to the observed difference in $\Delta R(T)/R_0$ and $\Delta t(T)/t_0$ dependences. The surface defects linked with inhomogeneity of $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ films most likely come from the process of saturation with potassium and their simultaneous oxidation [46]. The method of the comparative analysis of $\Delta\rho(T)/\rho_0$, $\Delta t(T)/t_0$ and $\Delta R(T)/R_0$ dependencies may be proposed to investigate the surface inhomogeneity of magnetic materials.

The applicability of the MRE theory [8] for the compositions of $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ films close to optimally-doped ($x_K=0.15$, and 0.18) is confirmed by the good agreement between the experimental and calculated data for $\Delta R(H)/R_0$ and $\Delta t(H)/t_0$ (for example, Fig. 6). According to the theory (Eqs. (1) and (2)) and experimental data, all formal criteria are valid: (i) the $\Delta R/R_0$ effect is positive, and (ii) $\Delta t/t_0$ as well as the magnetoresistance are negative, (iii) $\Delta R/R_0$ and $\Delta t/t_0$ as well as $\Delta\rho/\rho_0$ has similar dependence on magnetic field and are not saturated under the magnetic field of up to 8 kOe (insets in Fig. 6), and (iv) MRE reaches its maximum near T_C similarly to the colossal MR effect (Table 1). The surface layer defects do not affect the field dependence of $\Delta R/R_0$ of the films. The observed difference in the dependences of $\Delta R/R_0$, $\Delta t/t_0$ and $\Delta\rho/\rho_0$ on magnetic field is caused by the influence of the demagnetizing factor near the T_C in the case of the magnetic field applied in-plane and out-of-plane to the film surface.

The large value of MRE in magnetotransmission ($\sim 9\%$ close to $\Delta t/t_0$ for $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$, $\text{La}_{1-x}\text{Ag}_x\text{MnO}_3$ and $\text{La}_{0.82}\text{Na}_{0.18}\text{MnO}_3$ [5,24–26]) and magnetoreflexion ($\sim +5\%$ and -10% close to $\Delta R/R_0$ for $\text{La}_{0.9}\text{Ag}_{0.1}\text{MnO}_3$ [3]) modes in the $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ thin films under relatively small magnetic fields make it possible to propose these films as a new magneto-optical functional material working at temperatures close to the room one in a wide IR range.

4. Conclusion

Complex investigations of the structural, optical, magneto-optical (transversal Kerr effect, magnetoreflexion, magnetotransmission) and magnetotransport properties of the $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ films ($x_K=0.05, 0.10, 0.15,$ and 0.18) with thickness of 100 nm grown with using the two-stage procedure deposition on dielectric SrTiO_3 substrate have shown that these new films are promising magneto-optical materials with large values of the Kerr and magnetorefractive effects near the room temperature. The noticeable magnetorefractive effect in films under investigation can reach giant values ($-10\% < \Delta R/R_0 < 5\%$, $\Delta t/t_0 \sim -9\%$ in the IR range) and depends strongly on the level of doping by K^+ ions. If the Kerr and magnetorefractive effects reach their maximum in films close to optimally-doped compositions ($x=0.15, 0.18$) with high Curie temperature, the magnetoresistance does not change significantly. The obtained experimental data for films close to optimally-doped compositions were explained in the framework of the MRE theory [8]. The original method of the comparative analysis of $\Delta\rho(T)/\rho_0$, $\Delta t(T)/t_0$ and $\Delta R(T)/R_0$ dependencies for investigation of the surface inhomogeneity of films was used. The magnetic and charge inhomogeneities as well as doping level strongly influence on the value of magneto-optical effects. The multiply resonance-like contributions to magnetoreflexion observed in $\text{La}_{1-x}\text{K}_x\text{MnO}_3$ ($x_K=0.15,$ and 0.18) are explained by the epitaxial strains and charge and magnetic inhomogeneity of the films. The MRE spectra in magnetoreflexion mode shows local enhancement and appearance of resonance-like peak in the vicinity of the second phonon bands.

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