Research articles

Magneto-optical response to tunnel magnetoresistance in manganite films with a variant structure

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ABSTRACT

The optical, magneto-optical, electrical, and magnetic properties of $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3$ films grown on the SrTiO_3 and $\text{ZrO}_2(\text{Y}_2\text{O}_3)$ substrates have been studied. The temperature and magnetic field dependences of magnetotransmission of light in the $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3/\text{ZrO}_2(\text{Y}_2\text{O}_3)$ film with a variant structure exhibit features which can be attributed to an optical response to tunnel magnetoresistance – tunnel magnetotransmission. Unlike the optical response to colossal magnetoresistance, existing near the Curie temperature only, the tunnel magnetotransmission increases at $T \rightarrow 0$ K, has a complex hysteresis behavior with the change of sign of the effect, and saturates in magnetic fields above 1 kOe. The observed tunnel magnetotransmission has been interpreted within the magnetorefractive effect theory developed for granular films.

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1. Introduction

Numerous works have been devoted to identification of the physical mechanisms of optical response to colossal magnetoresistance (CMR) in doped manganites (see, for example, [1,2] and references therein). Such response can be considered as magnetoreflexion and magnetotransmission effects which take place in the CMR manganites in the infrared spectral range near the Curie temperature (T_C). Apart from CMR in granular manganite-based systems there exists the specific low-temperature magnetoresistance (MR) which is due to scattering and tunneling of spin-polarized charge carriers thorough grain boundaries – the so-called tunnel magnetoresistance (TMR) [3]. Noticeable TMR values were observed in manganite films with an equivalent (variant) structure [4]. Such structure can be formed

during growth of manganite films on the $\text{ZrO}_2(\text{Y}_2\text{O}_3)$ single crystalline substrates and can be considered as consisting of a set of identical crystallites – structural domains of equal size with fixed number and concentration of coherent high-angular boundaries within the film volume. This is the main structural difference from polycrystalline films. The variant structure leads to formation of highly conductive ferromagnetic nanosized grains (crystallites) separated by lowly conductive grain boundaries. As a result, an additional contribution to MR associated with TMR appears [4–7]. Taking into account different temperature dependences of $\text{CMR}(T)$ and $\text{TMR}(T)$ it is easy to separate their contributions to the total $\text{MR}(T)$ dependence by choosing the manganite composition with high T_C , for example, the $\text{La}_{2/3}\text{Ba}_{1/3}\text{MnO}_3$, and wide temperature range of interest. Though the TMR in manganites is well-known, however, there is clear lack of evidences for optical response to tunnel magnetoresistance. Earlier the optical response to TMR was considered theoretically for granular allows [8] and metallic superlattices [9,10]. As far as we know the few experimental attempts [11,12] to reveal an optical response to TMR in manganites have not been successful. The main problem of these attempts is in the absence of low-temperature optical measurements (close to zero Kelvin) and high absorbance of manganites

Abbreviations: CMR, colossal magnetoresistance; MR, magnetoresistance; TMR, tunnel magnetoresistance; IR, infrared; LMO, $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3$; SrTiO_3 , STO; YSZ, $\text{ZrO}_2(\text{Y}_2\text{O}_3)$.

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in a ferromagnetic state. In this work, we have made a comparative analysis of experimental data on optical properties and electrical resistance for the thin $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3$ films in the presence and absence of the variant structure in the wide temperature range. As a result, the contribution of optical response to TMR in magnetotransmission of unpolarized light was revealed.

2. Materials and methods

Epitaxial films of the $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3$ composition with the thickness of $d = 80$ nm were grown on the single-crystal SrTiO_3 (001) (the cubic lattice constant $a_0 = 3.905$ Å), hereafter denoted as LBMO/STO, and the $\text{ZrO}_2(\text{Y}_2\text{O}_3)$ (100) (the cubic lattice constant $a_0 = 5.140$ Å), hereafter denoted as LBMO/YSZ, substrates. The square-shaped substrates with the surface area of 1 cm^2 were used. The growth process was carried out with the substrate temperature of $T_s = 1000$ K and the oxygen pressure of $P = 0.4$ mbar by pulsed laser deposition using the excimer KrF laser with $\lambda = 247$ nm. The films were postannealed in the deposition chamber under oxygen flow at atmospheric pressure and $T = 730$ K to improve the oxygen stoichiometry. The elemental composition of the films was determined by EDAX analysis using the Inspect F (FEI) scanning electron microscope. According to X-ray diffraction data, the films were single-phase, highly oriented, and had an orthorhombic perovskite structure. The effective Curie temperature (T_C^*) of the films was determined from magneto-optical and magnetization measurements using the Lake Shore 7400 VSM vibration sample magnetometer. The T_C^* value of ≈ 290 K was found for the LBMO/YSZ film while $T_C^* \approx 300$ K was determined for the LBMO/STO film. Optical properties of the films were studied using the home made IR prism spectrometer in IMP UB RAS and the Vortex 80v Fourier spectrometer in the Radboud University. Measurements were carried out for unpolarized light in the infrared spectral range $1 \leq \lambda \leq 22 \mu\text{m}$, temperature interval from 10 to 350 K, and magnetic fields of up to $H = 7.5$ kOe oriented along (the Voigt geometry, $H \perp q$) and perpendicular to (the Faraday geometry, $H \parallel q$) the surface of the film. The films were arbitrary oriented during these experiments. The coefficient of absorption of light was calculated as $K = (1/d) \ln [(1 - R)^2/t]$, where t is the transmission of light and R is the coefficient of reflection. The magnetotransmission was defined as $\Delta t/t = [t_H - t_0]/t_0$, where t_H and t_0 are the intensities of the transmitted light in the presence and absence of a magnetic field H . The relative error in determination of the magnetotransmission was about 0.1%. Electro- and magnetoresistance of the films $\Delta\rho/\rho_0 = [\rho_H - \rho_0]/\rho_0$ were measured by the two-point contact method with ultrasonically soldered silver contacts in magnetic fields of up to 8 kOe.

3. Results and discussion

3.1. Spectral dependences

The shape of spectral dependences of the absorption coefficient K for the films (inset in Fig. 1b) is similar to those observed for the single crystals [13,14] and manganite films with CMR (see, for example [15]). The strong absorption at short wavelengths is attributed to onset of the $d-d$ transitions (${}^5E_g - {}^5T_{2g}$) in Mn^{3+} ions forming the absorption edge in manganite films [16,17]. The spectral dependence of K follows the $K(h\nu) = (h\nu - \varepsilon_g)^{3/2}$ law with the optical gap $\varepsilon_g = 0.4$ eV. Consequently, the absorption edge is formed by the direct forbidden interband transitions [15]. Upon cooling below T_C the absorption, as well as reflection, of the film increase due to the strong contribution from free charge carriers in the mid-IR range (inset in Fig. 1b) [1,12]. Application of mag-

netic field results in increase of the absorption too, thus resulting in negative $\Delta t/t$.

The spectra of magnetotransmission of light for the LBMO/YSZ and LBMO/STO films recorded at $T = 295$ K are shown in Fig. 1a. The magnitude of $\Delta t/t$ for LBMO/YSZ is two times lower than that for LBMO/STO which can be due only to the presence of structural domains in the LBMO/YSZ film. Variation from in-plane $H \perp q$ geometry of the experiment to the out-of-plane $H \parallel q$ one does not principally affect the magnitude of $\Delta t/t$ (not shown) due to the maximal fluctuations of the magnetic moments near T_C . In general, the $\Delta t/t$ spectra are formed by competition of a positive contribution from the interband transitions at $\lambda < 1.5 \mu\text{m}$ and a negative contribution associated with the interaction of light with delocalized charge carriers and localized states at $\lambda > 1.5 \mu\text{m}$. According to the theory of the magnetorefractive effect developed for manganites with CMR [see, for example, 1 and references therein] $\Delta t/t$ should reach a maximum at $\lambda \sim 20 \mu\text{m}$ in agreement with the $\omega\tau \sim 1$ condition, where τ is the relaxation time of the electron momentum ($1/\tau = 2.3 \times 10^{14} \text{ s}^{-1}$ [14]). This explains the observed increase of $\Delta t/t$ at $\lambda > 10 \mu\text{m}$. This theory is applicable only for optimally doped manganites [12] in the spectral range of interaction of light with delocalized charge carriers (the near and middle IR spectral ranges). According to (1) the estimated values of $\Delta t/t$ for the films under study are in good agreement with the experimental data (see Fig. 1a).

$$\Delta t/t = 0.5 \cdot \Delta\rho/\rho(0) \cdot t(0) \cdot k^2 \cdot (2n^2 + n)/(n^2 + k^2) \quad (1)$$

where the indexes of refraction n and k were subtracted from the absorption and reflection data (not shown).

Small differences between the experimental and calculated data are probably due to underestimation of the contribution of localized states and interband transitions in the theoretical model [1]. The most important result obtained is in revealing the noticeable magnetotransmission effect for the films with the variant structure far below T_C (at $T < 200$ K) where the TMR contribution exists even in the low field of $H = 2$ kOe. Upon $T \rightarrow 0$ K magnetotransmission increases and reaches a magnitude close to that observed near T_C . In other words, we can observe both a pronounced optical response to the CMR near T_C and optical response to TMR at low temperatures – the so-called tunnel magnetotransmission. As one can see, the $\Delta t/t$ spectra at low temperatures are identical to those at $T \sim T_C$ (Fig. 1b). The more detailed information can be derived from comparative analysis of the temperature and magnetic field dependencies of $\Delta t/t$ and MR effects.

In order to separate the contributions related to optical response to CMR and TMR in the manganite film we tried to model the $\Delta t/t(\lambda, T)$ dependence at different temperatures starting from 13 K (formulae 2). The same approach was used for calculation of magnetorefractive effect in granular systems with TMR [18].

$$\Delta t/t(T_2) = [\Delta t/t(T_1) \times T_2 \times \Delta\rho/\rho(T_1)]/T_1 \alpha \Delta\rho/\rho(T_2) \quad (2)$$

where $T_1 = 13$ K, $T_2 = 80$ K, 200 K and 300 K, $\Delta t/t$ and $\Delta\rho/\rho$ are magnetotransmission and MR of the film with the variant structure recorded at the appropriate temperatures.

One can see that this modeling gives a quite good qualitative result for $\Delta t/t$ at low temperatures and underestimated magnitude of the effect at temperatures close to T_C (300 K) (Fig. 1b). This can be attributed to two mechanisms of $\Delta t/t$. The first one is related to the existence of ferromagnetic domains with high conductivity, the $\Delta\rho/\rho(T)$ dependence having maximum near T_C , and comparable in magnitude with magnetotransmission for the film without variant structure. The second one is related to scattering at the grain boundaries of crystallographic structural grains with low-conductivity. At the temperature scale a borderline between these contributions is located near $T = 200$ K, where for the LBMO/STO

Fig. 1. (1.5-column fitting image) Spectral dependences of magnetotransmission (a) $\Delta t/t$ for LBM0/YSZ and LBM0/STO films at $H \perp q$ and $H = 7$ kOe and $T = 295$ K, dashed lines – calculations according to the expression (1); (b) magnetotransmission for LBM0/YSZ film at different temperatures and $H = 2$ kOe; dashed lines – calculated spectra according to the expression (2). In the inset: absorption spectra for the LBM0/YSZ film.

film $\Delta t/t$ is vanished. Possible size-dependent effect in the film with nanosized structural grains and magnetic domains should also be taken into account. Thus our modeling can be applied for estimation of the optical responses to TMR and CMR for such complex materials as manganites.

3.2. Temperature dependences

The temperature and magnetic field dependencies of $\Delta t/t$ as well as of $\Delta \rho/\rho_0$ differed significantly for the LBM0/YSZ and

LBM0/STO films. The $\Delta t/t(T)$ and $\Delta \rho/\rho(T)$ temperature dependences for the LBM0/STO film have a clear maxima near T_C (Fig. 2a) associated with the suppression fluctuations of the magnetic moments by magnetic field near T_C (Fig. 2b). In the case of the LBM0/YSZ film the $\Delta \rho/\rho(T)$ dependence shows only a shoulder near T_C but $\Delta t/t(T)$ dependence has a maximum of $\sim -5.5\%$ ($H = 7.5$ kOe) (Fig. 2a). This maximum can be reasonably related to the optical response to CMR. The values of $\Delta t/t$ for the LBM0/STO and LBM0/YSZ films are almost the same near T_C . Unlike the LBM0/STO film the $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3/\text{ZrO}_2(\text{Y}_2\text{O}_3)$ one with the variant structure demonstrates the significant low-temperature magnetoresistance (up to 30% at $T \rightarrow 0$ K) associated with the tunneling of spin-polarized electrons through the structural grain boundaries. Similar $\Delta \rho/\rho(T)$ behavior was observed for other manganite films with variant structure [5,9]. Moreover, we revealed substantial magnetotransmission in the LBM0/YSZ film at $T < 150$ K (Fig. 2a). The value of $\Delta t/t$ reaches about -1% at $T = 13$ K which is comparable with the value of $\Delta t/t$ near T_C .

In nanomaterials TMR is usually described by the expression $\Delta \rho/\rho \approx -P^2 m^2 / (1 + \sqrt{C/T})$, where C is a constant [3,19]. In our case the experimental curve $\Delta \rho/\rho(T)$ is better described by the $f = a + b/(T + c)$ dependence proposed for bulk polycrystalline manganites (Fig. 2b) [20]. The authors of [21,22] experimentally showed that such temperature dependence of TMR in manganite films on YSZ substrates is associated with destruction of the coherence of the structural domain boundaries and appearance of polycrystallinity. Estimation of the degree of spin-polarization at $T \rightarrow 0$ K using the expression $\Delta \rho/\rho = 2P^2/(1 - P^2)$ from [3] gives the value of $P \approx 0.36$ which is close to $P \approx 0.41$ obtained for the $\text{La}_{0.8}\text{Ag}_{0.1}\text{MnO}_{3+\delta}/\text{YSZ}$ film [11]. Using the expression (1) and the experimental temperature dependence of TMR we calculated the $\Delta t/t(T)$ dependence. Good correlation between calculated and experimental data was obtained only at $T = 13$ K (taking into account the value of H). Above 150 K one should consider an extra significant contribution related to the optical response to the metal-insulator transition (inset in Fig. 2a) within structural domain that has a maximum near T_C for the film material.

Fig. 2. (1.5-column fitting image) Temperature dependences of (a) magnetotransmission $\Delta t/t$ and (b) magnetoresistance $\Delta \rho/\rho$ for LBM0 films on different substrates at $H = 7.5$ kOe. Solid line – $\Delta t/t(T)$ dependence obtained for the LBM0/YSZ film at $H = 2$ kOe; dashed lines – the results of calculations of $\Delta t/t$ and $\Delta \rho/\rho$ according to the expression (1) and $f = a + b/(T + c)$ [3], respectively. In the inset: (a) the temperature dependence of the transparency $t(T)$ at $\lambda = 6$ μm and (b) the field dependence of $\Delta \rho/\rho(H)$ at $T = 4.2$ K for the LBM0/YSZ film.

Fig. 3. (1.5-column fitting image) Magnetic field dependences of (a) magnetotransmission $\Delta t/t$ for the LBM0/YSZ and LBM0/STO films at $\lambda = 6 \mu\text{m}$, $T = 295 \text{ K}$ and $H \perp q$; (b) magnetoresistance $\Delta \rho/\rho$ for the LBM0/ZYO and LBM0/STO films at different temperatures and magnetic field direction; (c) magnetotransmission $\Delta t/t$ for the LBM0/YSZ film at $\lambda = 6 \mu\text{m}$ and $T = 80 \text{ K}$.

3.3. Magnetic field dependences

The differences between the CMR and TMR, as well as optical responses to CMR and TMR, are most clearly manifest themselves in the magnetic field dependences of these effects (Fig. 3b). First of all, upon variation of the geometry of experiment and application of magnetic field either along ($H \parallel q$) or perpendicular ($H \perp q$) to the film's surface, the influence of the form factor was 3 times less than the expected $N = 4\pi$ for thin films. The reduced N value for the film with variant structure may be explained by the influence of structural domain boundaries on the dynamics of magnetic domains in applied magnetic field [21].

In the vicinity of the Curie temperature the magnetic field dependences of $\Delta t/t(H)$ and $\Delta \rho/\rho(H)$ for both films are similar and typical for manganites with CMR [1,2]. The magnetic field dependences of colossal magnetoresistance and magnetotransmission are even on magnetization and demonstrate the absence of saturation in magnetic fields of up to 8 kOe regardless the direction of the in-plane field relative to the film surface. They are associated with the maximal fluctuations of the magnetic moments near T_C . At low temperatures ($T < 150 \text{ K}$) the CMR contribution in thin films becomes zero, while TMR appeared only in the LBM0/YSZ film (inset in Fig. 2b).

In contrast to CMR, the tunnel magnetoresistance has a complex hysteresis behavior with a positive value of up to $\sim +0.7\%$ in the area of technical magnetization associated with reorientation of magnetic domains in the fields below $H_1 = 300 \text{ Oe}$, and a negative value of up to $\sim -20\%$ associated with the tunneling effect at $H_2 = 1.9 \text{ kOe}$ ($H \perp q$) and $H_2 = 3.8 \text{ kOe}$ ($H \parallel q$) (Fig. 3b). Relatively high value of H_2 is due to the planar anisotropy of the film with TMR [7,22]. As can be seen from the Fig. 3c, magnetotransmission strictly follows the TMR field dependences and reaches about -1.1% at $H = 7.5 \text{ kOe}$. It differs significantly from the behavior of the magnetotransmission near T_C and is related to the optical response to TMR – the tunnel magnetotransmission. Weak hysteresis of tunnel magnetotransmission is preserved even in case of the Faraday geometry with $H \parallel q$. One can notice the different values of form-factor N obtained from the $\Delta t/t(H)$ and $M/M_s(H)$ dependences (not shown). Under these circumstances, additional study of the anisotropy of TMR and related to it tunnel magnetotransmission in the film with a variant structure for in-plane oriented magnetic field can be suggested.

4. Conclusion

Optical, magneto-optical, electrical, and magnetic properties of the $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3$ magnetoresistive epitaxial films grown on the SrTiO_3 and $\text{ZrO}_2(\text{Y}_2\text{O}_3)$ substrates have been studied. Apart from colossal magnetoresistance, the films on both substrates show significant magnetotransmission (5%) of unpolarized light in the wide infrared range with a maximum near the Curie temperature (290–300 K). The $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3/\text{ZrO}_2(\text{Y}_2\text{O}_3)$ film with the variant structure demonstrates the low-temperature magnetoresistance (up to 30% at $T = 4.2 \text{ K}$) associated with the tunneling of spin-polarized carriers through the boundaries of structural domains. Temperature dependence of this effect was well approximated by the function $f = a + b/(T + c)$, which points to polycrystalline structure of the film. The degree of spin-polarization of charge carriers was estimated as $P \approx 0.36$.

It is worth to notice, a direct optical response to the tunnel magnetoresistance was revealed in magnetotransmission of light for the $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3/\text{ZrO}_2(\text{Y}_2\text{O}_3)$ film for the first time – tunnel magnetotransmission. The so-called tunnel magnetotransmission follows the temperature and field dependences of tunnel magnetoresistance and reach the value of -1.1% at $T = 13 \text{ K}$ and $H = 2 \text{ kOe}$. The tunnel magnetotransmission depends on the direction of the magnetic field along or perpendicular to the film plane due to the contribution from the magnetic domain dynamics and the demagnetizing factor. Calculations of the spectral and temperature dependences of magnetotransmission in the framework of the magnetorefractive effect theory developed earlier for granular systems show good correlation with the experimental data.

The appearance of a substantial magnetoresistance and magnetotransmission in manganite films in relatively weak magnetic fields is of considerable interest for potential fabrication of optoelectronic sensors of magnetic field with high-sensitivity. For these purposes one can use both the appearance of sharp maxima of positive magnetoresistance and magnetotransmission in the region of technical magnetization (i.e., magnetization related to the reorientation of magnetic domains) in fields weaker than 300 Oe and that of large negative magnetoresistance (25%) and magnetotransmission (1%) related to the tunneling effect in magnetic fields of above 3 kOe.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at <https://doi.org/10.1016/j.jmmm.2017.10.006>.

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