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Features of RBaMn$_2$O$_6$ (R = Pr$_{0.5}$Nd$_{0.5}$, Nd$_{0.5}$Sm$_{0.5}$, Sm) double manganites with partial ordering

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Single crystals of double manganites RBaMn$_2$O$_6$ (R = Pr$_{0.5}$Nd$_{0.5}$, Nd$_{0.5}$Sm$_{0.5}$, Sm) were grown and the magnetic and optical properties in the near-IR range were studied. From the analysis of structural data, partial ordering of the R and Ba ions (A-site ordering) $\sim$ 50% was confirmed. According to magnetic data, the ferromagnetic contribution decreases and the coercive force increases as the average radius of the rare-earth ion decreases in the series Pr$_{0.5}$Nd$_{0.5}$ $\rightarrow$ Nd$_{0.5}$Sm$_{0.5}$ $\rightarrow$ Sm. From measurements of the temperature dependences of light transmission a change in the conductivity character from „semiconductor“ to „metallic“ at cooling was discovered: for manganite with Pr$_{0.5}$Nd$_{0.5}$ the conductivity change temperature T_{MI} is close to the magnetic phase transition temperature T_c, and for manganites with Nd$_{0.5}$Sm$_{0.5}$ or Sm the T_{MI} value is higher than T_c. Near T_{MI} in the manganites under consideration, the influence of the magnetic field on the transmission of light and the magnetotransmission effect were found. The highest magnetotransmission of 5% is observed in manganite with R = Pr$_{0.5}$Nd$_{0.5}$.

Keywords: double manganites, ordering, phase transitions, magnetotransmission effect.

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1. Introduction

Double manganites of the type RBaMn$_2$O$_6$ (O$_6$), where R are rare earth ions have been actively studied since early 2000 years. [1,2]. The study of a family of such manganites with R from La to Dy found that a significant change of physical properties occurs in these materials which are chemically related to the manganites $R_{0.5}$Ba$_{0.5}$MnO$_3$ (O$_3$) as a result of the layered ordering of ions R and Ba along the axis c (A-site ordering). In particular, a change occurs during the formation of a double manganite from the state of spin glass with a low temperature of the magnetic phase transition in disordered $R_{0.5}$Ba$_{0.5}$MnO$_3$ to a state with ferromagnetic (FM) or antiferromagnetic (AFM) ordering (depending on the type of rare earth ion) and high Curie or Neel temperatures, or with a set of magnetic transitions that coexist with structural phase transitions and with transitions to the state of orbital ordering. In addition, such manganites began to attract attention in connection with the discovery of the effect of colossal magnetoresistance in them for some compositions and the degree of ordering of [1,3–5], as well as the effect of magnetotransmission [6,7]. At the same time, the study of manganites PrBaMn$_2$O$_6$ and NdBaMn$_2$O$_6$, showed that not only temperatures depend on the degree of ordering by A-position magnetic and structural phase transitions, but also the magnetoresistance values [4,5]. It should be noted that the evolution of properties depending on the degree of ordering is mainly studied on polycrystalline or powder samples, the ordering in which either varies by annealing [4], or the synthesis temperature [5]. Recently, we showed that similar changes occur when growing a single crystal PrBaMn$_2$O$_6$ from a workpiece of the initial ordered manganite [6]. There are a few publications in the literature on the study of the properties of single-crystal samples of ordered SmBaMn$_2$O$_6$ [8] and NdBaMn$_2$O$_6$ [9], that contain very limited volume of experimental data due to the complexity of growing single crystals RBaMn$_2$O$_6$ of large sizes.

Single crystals of RBaMn$_2$O$_6$ (R = Pr$_{0.5}$Nd$_{0.5}$, Nd$_{0.5}$Sm$_{0.5}$, Sm) were grown and studied in this paper, the data obtained are compared on the one hand with the properties of polycrystals of ordered manganites of the same compositions. On the other hand, the obtained data are compared with the properties of a single crystal of PrBaMn$_2$O$_6$ from [6]. Understanding the process of formation of a partially ordered state in A position is important in the case of attempts to obtain manganites for applied tasks. In particular, since colossal magnetoresistance is observed in partially ordered manganites [1,3,4,5] and, as we find, magnetotransmission [6,7], these materials can be considered for applications in spintronics or magneto-optics.

2. Specimens and methods of study

Polycrystalline workpieces of ordered manganites RBaMn$_2$O$_6$ (R = Pr$_{0.5}$Nd$_{0.5}$, Nd$_{0.5}$Sm$_{0.5}$, Sm) were prepared for obtaining of a few crystals by solid-phase synthesis in an argon atmosphere (workpieces with R = Pr$_{0.5}$Nd$_{0.5}$, Nd$_{0.5}$Sm$_{0.5}$) and the topotactic reaction [10] (blank with R = Sm). Single crystals of RBaMn$_2$O$_{5+\delta}$ were grown using the method of crucibleless zone melting in

the URN-2-3P furnace in an argon atmosphere (5 atm.); the growth rate was ~ 5–7 mm/h. The single crystals were then annealed in an oxygen stream at 500°C for 25 h to achieve a maximum oxygen content.

Structural studies were performed by X-ray powder diffraction method on fragments of ground single crystals using DRON-2.0 diffractometer. The degree of ordering of the system of RBaMn$_2$O$_6$/$R_{0.5}$Ba$_{0.5}$MnO$_3$ (number of phases of O_6 and phases O_3), according to X-ray data, was evaluated using the PowderCell 2.4 program [11]. A similar assessment of the degree of ordering from the analysis of X-ray data was done in [4].

The elemental analysis of some samples was carried out using a scanning electron microscope QUANTA 200 Pegasus at Collaborative Access Center (CAC) of the IMP UB RAS.

The magnetic properties were studied using a magnetometric unit (SQUID magnetometer) MPMS-XL-5 CAC IMP UB RAS. The temperature dependences of magnetization were measured in the field of 50 kOe and 100 Oe in standard cooling modes without a magnetic field followed by heating in a magnetic field (ZFC mode), as well as in the cooling mode in a magnetic field (FC mode). The magnetization curves $M(H)$ were measured at 5 K after cooling in a magnetic field of 50 kOe.

The temperature dependences of the light transmission of the studied manganites were measured to analyze changes in the charge subsystem in the near-infrared range. Optical composites based on CsI, used as a transparent matrix in the IR region, were prepared for optical studies since the studied single crystals are characterized by low resistance, have high absorption and are not transparent in the form of thin single crystal plates. For this purpose the single crystals were crushed in an agate mortar, mixed with CsI in an amount of 3 and 300 mg, respectively, thoroughly ground and then pressed under pressure of 0.5 MPa. As a result, semitransparent tablets with a diameter of ~ 10 and a thickness of ~ 1 mm were obtained. Measurements were carried out in the range of 0.1–0.54 eV in the temperature range of 90–350 K using an automated cryomagnetic unit based on a prism monochromator. The temperature dependences of light transmission were measured at fixed energy values in the pre-cooled heating mode without a magnetic field; the rate of temperature change was ~ 1 K/min. Light transmission was also measured when an external magnetic field of 8 kOe was applied. The optical transmission of manganite was calculated as

$$t(T) = I(T)/I_0(T),$$

where I and I_0 — the intensity of light transmitted through the manganite/CsI composite and a CsI tablet without manganite prepared using the same technology. The magnetotransmission was calculated using the following formula

$$MT(T) = \big(t_{\mathrm{H}}(T) - t_0(T)\big)/t_0(T) = \big(I_{\mathrm{H}}(T) - I(T)\big)/I(T),$$

where I_{H} is the intensity of light transmitted through the composite in a magnetic field.

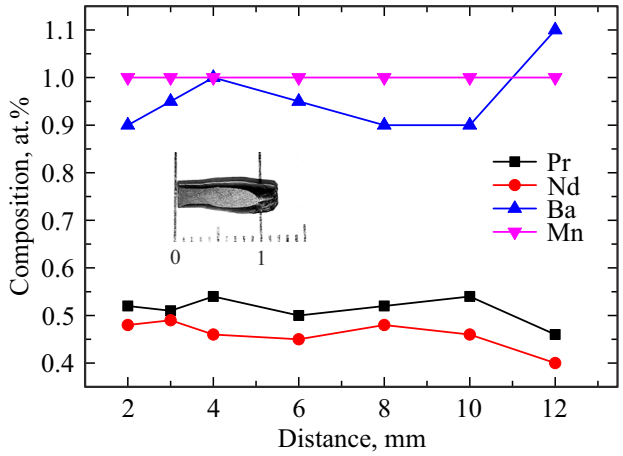

Figure 1. Distribution of chemical elements along the single crystal Pr$_{0.5}$Nd$_{0.5}$BaMn$_2$O$_6$. A photo of a single crystal with a surface prepared for SEM is shown in the insert.

3. Results and discussion

Figure 1 shows a photo of a single crystal of Pr$_{0.5}$Nd$_{0.5}$BaMn$_2$O$_6$ and the distribution of elements Pr, Nd, Ba, Mn along the growth axis determined by scanning electron microscopy (SEM). The content of the elements at each point is reduced to the index value Mn = 2. We believe that the distribution of elements is satisfactory for such a multicomponent system. There is no strong deviation from stoichiometry along the length of the single crystal, except for the region of the ingot edge.

Figure 2 shows X-ray images of the obtained manganites. The absence of impurity phases is visible for all samples (other than RBaMn$_2$O$_6$ or $R_{0.5}$Ba$_{0.5}$MnO$_3$). It is known from the literature (see, for example, [4,5]) that A-site disordered manganites of the type $R_{0.5}$Ba$_{0.5}$MnO$_3$ are characterized by a cubic structure, and ordered manganites are characterized by orthorhombic or tetragonal structure. The diffraction patterns provided in Figure 2 show peaks corresponding to both phases. The inserts to Figure 2 show the lines from the planes (210) for phase (O_3); (202), (201), (104) for phases (O_6) Pr$_{0.5}$Nd$_{0.5}$BaMn$_2$O$_6$ and Nd$_{0.5}$Sm$_{0.5}$BaMn$_2$O$_6$ and lines (601), (442), (281) for the phase SmBaMn$_2$O$_6$, which demonstrate the qualitative decomposition of the diffractogram into phases with structures O_3 and O_6. Therefore, the experimental spectra were adjusted under the assumption of the coexistence of phases O_6 and O_3. The best agreement of the theoretical curves with the experimental ones is achieved under the assumption of a quantitative ratio of the mass fraction of phases 47/53, 43/57 and 48/52 for manganites with R = Pr$_{0.5}$Nd$_{0.5}$, Nd$_{0.5}$Sm$_{0.5}$ and Sm, respectively (table). It should be noted that a value close to 50% of the degree of ordering at A-position was previously obtained in a single crystal of PrBaMn$_2$O$_6$ grown under the same conditions [6]. It can be assumed that the formation of a state with an ordering of about 50% in double manganites

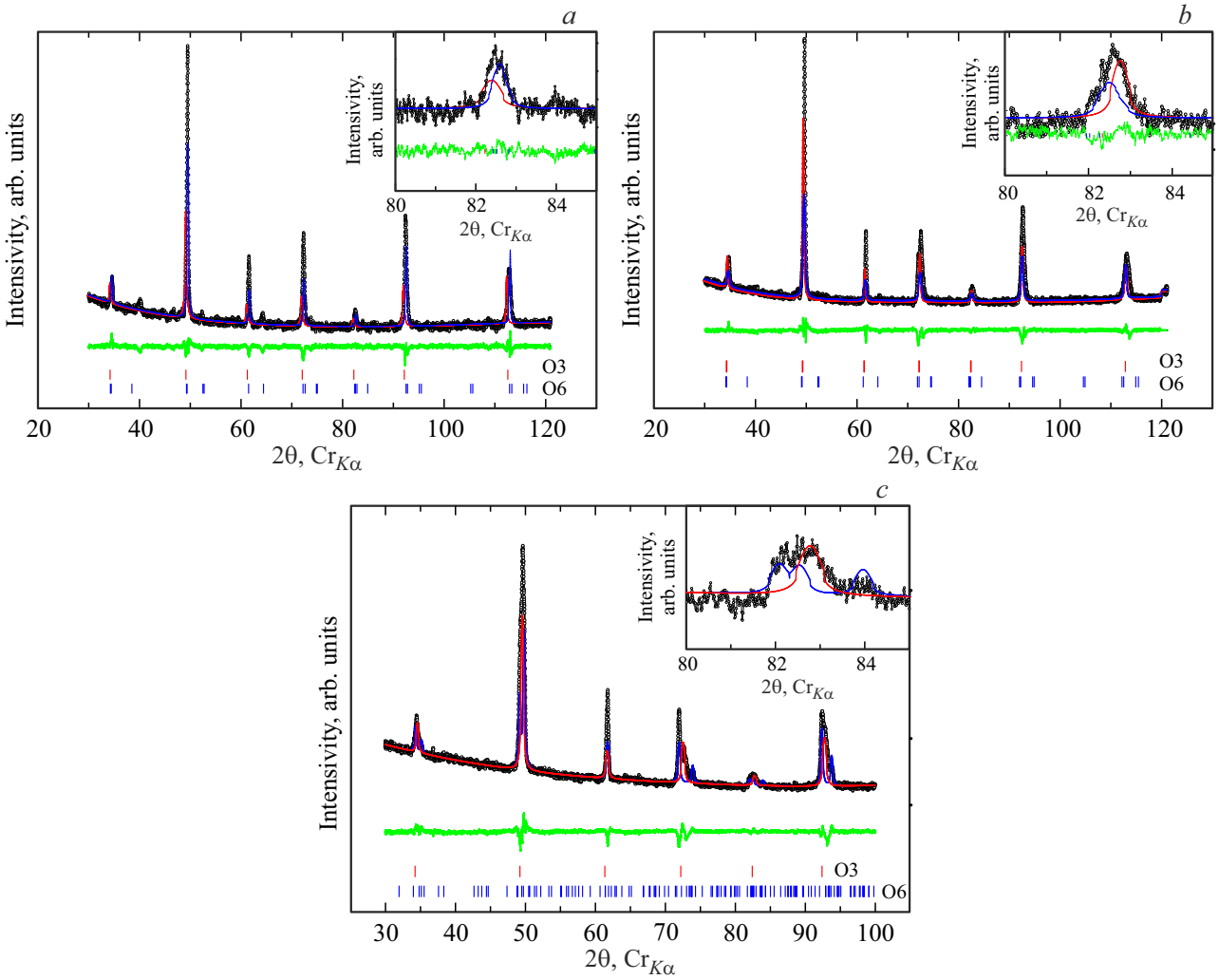

Figure 2. Diffractograms of ground single crystals $Pr_{0.5}Nd_{0.5}BaMn_2O_6$ (a), $Nd_{0.5}Sm_{0.5}BaMn_2O_6$ (b), $SmBaMn_2O_6$ (c). Symbols — experimental values, solid lines — calculation for $RBaMn_2O_6$ — blue line, calculation for $R_{0.5}Ba_{0.5}MnO_3$ — red line. Calculation/experiment difference — green line. The vertical labels at the bottom of the figure refer to the Bragg corners of the structures $R_{0.5}Ba_{0.5}MnO_3$ (O_3), $RBaMn_2O_6$ (O_6). The inserts contain fragments of diffraction patterns in the area of angles $2\theta \sim 82°$.

under the synthesis conditions indicated in this paper is a general trend for double manganites, regardless of the type of rare earth ion.

The lattice parameters of the studied manganites were also determined from the adjustment of diffraction patterns. The data obtained are shown in table. It can be seen that a decrease of the average ionic radius of a rare earth element results in a slight decrease of the lattice parameter for the disordered phase O_3, and the same trends are observed for samples with $R = Pr_{0.5}Nd_{0.5}$ and $Nd_{0.5}Sm_{0.5}$ that have the same tetragonal symmetry group in an ordered state $P4/mmm$. The data obtained for our manganites on the parameters of the crystal lattice are consistent with the literature data.

Figure 3 shows the temperature dependences of the magnetization of the studied single crystals of manganites $RBaMn_2O_6$. It can be seen that manganite $Pr_{0.5}Nd_{0.5}BaMn_2O_6$ is characterized by a transition to a ferromagnetic state near 150 K. The divergence of the curves $M(T)$ measured in the ZFC and FC modes at $T < T_C$ and the decrease of the magnetization at low temperatures indicates the presence of an inhomogeneous magnetic state. At the same time, the predominantly ferromagnetic nature of the ground state is confirmed by the magnetization curve measured at 5 K (Figure 4): a very narrow hysteresis loop ($H_c \approx 110$ Oe) is observed for this manganite, a sharp increase of magnetization in the region of weak magnetic fields and an the occurrence of the linear dependence of magnetization on the magnetic field at $H > 6$ kOe.

A series of manganites $Nd_{1-x}Pr_xBaMn_2O_6$ with $x = 0.25$, 0.5, 0.75 is considered in Ref. [10]. The dependence $M(T)$ has a different form for a sample with $Nd_{0.5}Pr_{0.5}$: an increase of magnetization is observed near room temperature in case of cooling 350 K, then the magnetization reaches a maximum at ~ 270 K, and the magnetization decreases to almost zero with a further

Number of phases O_6 and O_3 in single crystals, spatial group and lattice cell parameters

Composition	Number of phase, mass%		Spatial group	Parameters of the lattice cells, Å
$Pr_{0.5}Nd_{0.5}BaMn_2O_6/(Pr_{0.5}Nd_{0.5})_{0.5}Ba_{0.5}MnO_3$	O_6	43	#123, $P4/mmm$	$a = 3.895(8)$ $c = 7.770(3)$
	O_3	57	#221, $Pm3m$	$a = 3.900(3)$
$Nd_{0.5}Sm_{0.5}BaMn_2O_6/(Nd_{0.5}Sm_{0.5})_{0.5}Ba_{0.5}MnO_3$	O_6	48	#123, $P4/mmm$	$a = 3.904(6)$ $c = 7.778(3)$
	O_3	52	#221, $Pm3m$	$a = 3.889(6)$
$SmBaMn_2O_6/Sm_{0.5}Ba_{0.5}MnO_3$	O_6	47	#62, $Pnma$	$a = 11.066(1)$ $b = 15.291(9)$ $c = 5.533(6)$
	O_3	53	#221, $Pm3m$	$a = 3.885(4)$

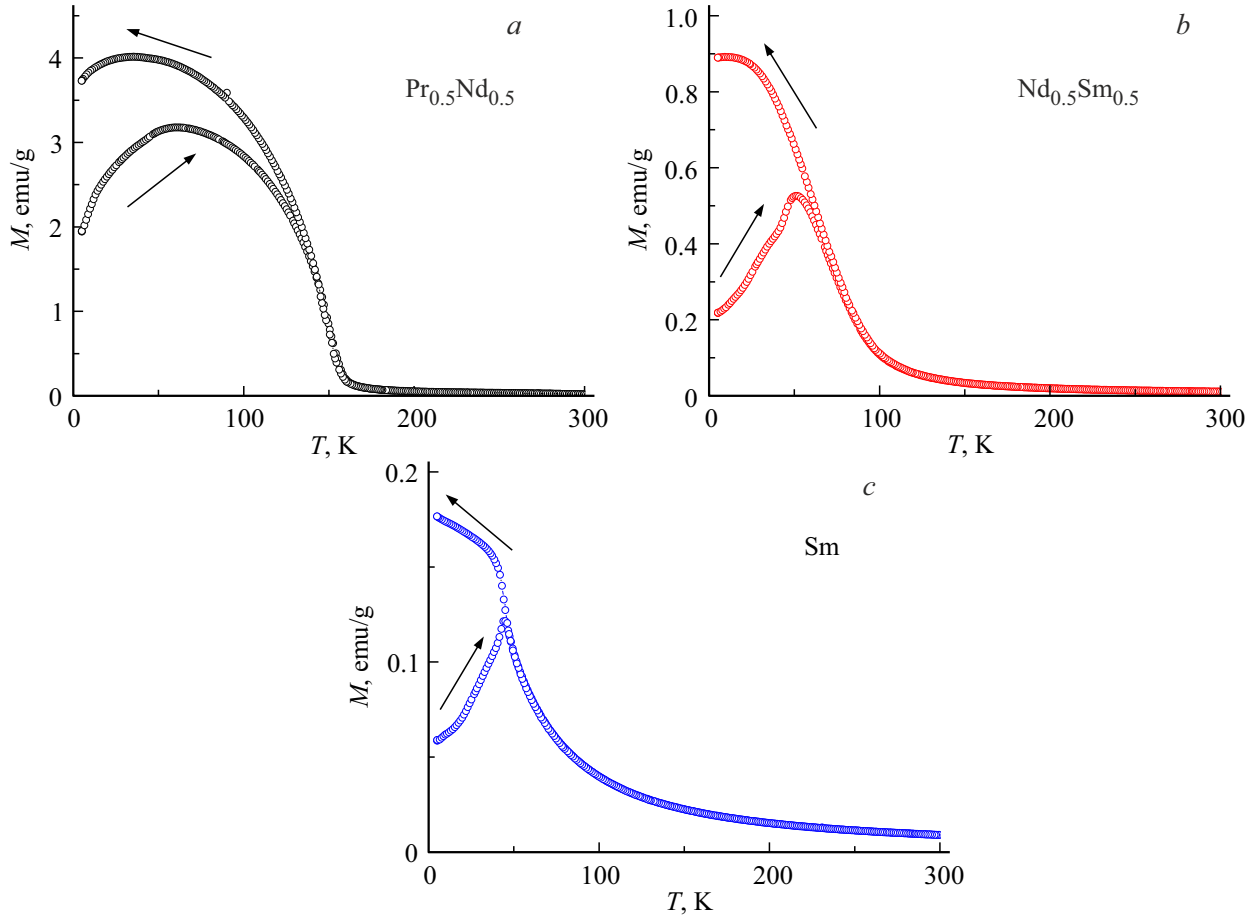

Figure 3. Temperature dependences of the magnetization of single crystals of $Pr_{0.5}Nd_{0.5}BaMn_2O_6$ (a), $Nd_{0.5}Sm_{0.5}BaMn_2O_6$ (b) and $SmBaMn_2O_6$ (c) measured in field of 100 Oe in ZFC and FC mode.

decrease in temperature. This behavior was explained by the coexistence of AFM and FM correlations in Ref. [10]. At the same time, manganite $Nd_{0.5}Pr_{0.5}BaMn_2O_6$ is in the AFM A-type state in the low temperature region, according to $M(H)$, with ferromagnetic ordering in planes ab and antiferromagnetic ordering between planes [10]. Thus, the magnetic data for our single crystal $Pr_{0.5}Nd_{0.5}BaMn_2O_6$ and a sample of the same composition from [10] significantly differ, and this difference in the nature of the curves $M(T)$ and $M(H)$ is explained by the difference in the degree of

ordering at A-position: as can be seen from the table, in our case the ordering is approximately 50%. It should be noted that a similar difference in the nature of magnetic properties was observed in $PrBaMn_2O_6$ with varying degrees of ordering at A-position [4,6]. In particular, these studies show that a decrease of the degree of ordering from 96 to 57% in $PrBaMn_2O_6$ results in a decrease of the Curie temperature from $\sim$ 300 to 240 K and the disappearance of the transition to the antiferromagnetic state.

The temperature dependences of magnetization for manganites $Nd_{0.5}Sm_{0.5}BaMn_2O_6$ and $SmBaMn_2O_6$ $M(T)$ have a qualitatively identical character: an increase of magnetization below $\sim$ 150 K with a maximum at $\sim$ 50 K in the ZFC mode and a strong divergence of curves measured in the ZFC and FC modes, especially in the magnetic field of 100 Oe. This behavior is associated with the transition to a spin glass type state in the literature [2–5]. At the same time, the magnetization value in the sample $Nd_{0.5}Sm_{0.5}$ is higher than in the sample with Sm. There is a small ferromagnetic contribution in crystals of $Nd_{0.5}Sm_{0.5}BaMn_2O_6$ and $SmBaMn_2O_6$ according to the data of magnetization curves $M(H)$, which manifests itself in a (more or less) sharp increase of magnetization in the region of weak magnetic fields. The ordered manganites $Nd_{1-x}Sm_xBaMn_2O_6$ were studied in [12], and it was shown that there are two maxima for samples with $Nd_{0.5}Sm_{0.5}$ and Sm depending on $M(T)$: at $\sim$ 250 K and less intensive at 350 and 370 K, respectively. At the same time, the magnetization of the sample with $Nd_{0.5}Sm_{0.5}$ is approximately three times higher than magnetization of the sample with Sm. The high-temperature feature on the curves $M(T)$ in [12] was explained by the transition to a state with charge/orbital ordering, and the low-temperature one was explained by the proximity of the temperature of the phase transition, at which the character of the orbital ordering changed. In addition, a conclusion was made from the analysis of the heat capacity data, in addition to the magnetic data, that these manganites have a spin glass state at $T < 60$ K, which coexists with the antiferromagnetic state [12]. Our single crystals have no features at 250 and 350 K, which, as well as for the sample with $R = Pr_{0.5}Nd_{0.5}$, is associated with a decrease of the degree of A-site ordering.

Comparing the magnetic data for the single crystals of double manganites studied by us, it can be noted that a decrease of the degree of ordering to $\sim$ 50% results in the absence of signs of transition to a ferromagnetic state on the curves $M(T)$, only a transition to a spin glass type state is observed unlike the sample with $R = Pr_{0.5}Nd_{0.5}$ in manganites with $Nd_{0.5}Sm_{0.5}$ and Sm. This difference may be due to the fact that in the ordered state, manganites with Pr are characterized by the formation of an antiferromagnetic A-type state with ferromagnetic exchange in ab-planes [2,4,13], manganites with Sm have a basic antiferromagnetic CE type state in which ferromagnetic ordering is formed in zigzag chains and antiferromagnetic ordering is formed between them [2,4,8], and manganites with Nd are located on the boundary between these two

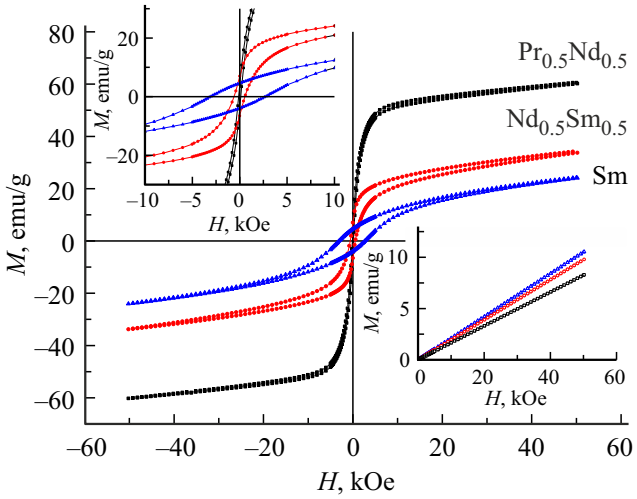

Figure 4. Field dependences of the magnetization of single crystals of $Pr_{0.5}Nd_{0.5}BaMn_2O_6$, $Nd_{0.5}Sm_{0.5}BaMn_2O_6$ and $SmBaMn_2O_6$, measured at $T = 5$ K. The upper insert shows the region of small magnetic fields, the lower insert shows the linear contribution to magnetization determined from the adjustment of experimental curves.

states in the phase diagram [2,4]. Therefore, it is possible to expect the existence of a ferromagnetic contribution in manganite with $R = Pr_{0.5}Nd_{0.5}$ with partial ordering because of the exchange in ab-planes [10] in contrast to manganite with $Nd_{0.5}Sm_{0.5}$.

The magnetization curves $M(H)$ for samples of manganites with $Nd_{0.5}Sm_{0.5}$ and Sm (Figure 4), as well as $Pr_{0.5}Nd_{0.5}$, demonstrate a sharp increase of magnetization in weak magnetic fields and close to linear magnetization growth with a weak slope in the high-field region. At the same time, the angle of inclination of the curves $M(H)$ in the area $H > 6$ kOe is almost the same. The observed character of the magnetization curves can be explained by the coexistence of two phases, one of which is antiferromagnetic, and the other has a ferromagnetic contribution. Subtracting the linear antiferromagnetic contribution from the dependencies $M(H)$ allows isolating the ferromagnetic contribution and determining the corresponding saturation magnetization value, which is $M_s = 52$ emu/g for $Pr_{0.5}Nd_{0.5}$, 24 emu/g for $Nd_{0.5}Sm_{0.5}$ and $\sim$ 14 emu/g for Sm. At the same time, the linear antiferromagnetic contribution for the considered manganites is close to each other (see bottom insert in Figure 4).

In addition, there is a significant increase of coercive force from $H_c = 110$ Oe for $Pr_{0.5}Nd_{0.5}$ to 550 Oe for $Nd_{0.5}Sm_{0.5}$ and to 2660 Oe for Sm as can be seen from the curves $M(H)$ (upper inset in Figure 4). An increase of the coercive force and a decrease of the magnitude of magnetization in case of the transition from $Nd_{0.5}Sm_{0.5}$ to Sm was also observed in Ref. [12] which considered the manganites ordered at A position. The observed changes in magnetic properties are obviously the result of an increase of the local distortions of the crystal lattice with a decrease of

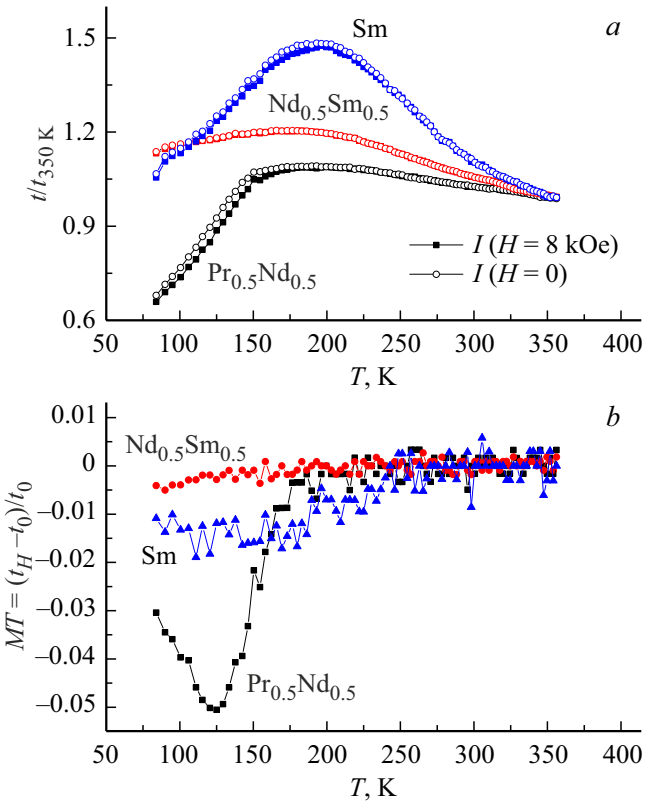

Figure 5. Temperature dependences of relative light transmission $t/t_{350\,K}(T)$ (a) without a magnetic field (light symbols) and in a magnetic field of 8 kOe (dark symbols) and temperature dependences of magnetotransmittance (b) at energy $E = 0.11$ eV ($\lambda = 10.25\,\mu$m) manganites $RBaMn_2O_6$.

the average ionic radius of the rare earth ion in the series Pr → $Pr_{0.5}Nd_{0.5}$ → $Nd_{0.5}Sm_{0.5}$ → Sm, which destroy collinear magnetic structures.

For studying the changes of the charge subsystem that occur in the studied manganites we measured the temperature dependences of light transmittance $t(T)$ in the energy range between the beginning of the phonon band region ($E \approx 0.09$ eV) and the edge of the lowest energy fundamental absorption band corresponding to $d-d$-transitions and according to the data from Ref. [8,9] is at an energy of 0.9 or 1.1 eV for $NdBaMn_2O_6$ or $SmBaMn_2O_6$, respectively. Assuming a homogeneous charge state in this spectral region, where light mainly interacts with charge carriers, the dependences $t(T)$ are similar to the temperature dependences of electrical resistance $\rho(T)$ and allow drawing qualitative conclusions about the nature of charge carriers and possible transitions „metal–insulator". The temperature dependences of the light transmittance of the studied manganites, reduced to the transmission value at 350 K are shown in Figure 5, *a*. We previously showed [14] that the contribution from charge carriers to the temperature dependences of light transmission depends on energy and is stronger at energies near the beginning of phonon bands. Therefore, curves $t(T)$ measured at 0.11 eV are presented for the analysis of data on the charge subsystem (Figure 5, *a*). It can be seen that the curves $t(T)$ differ for manganites with different rare earth ions.

A slight change of light transmittance with a temperature in the temperature range above 150 K and a rather sharp „metallic" dependence course ($dt/dT > 0$) at $T < 150$ K is observed for $Pr_{0.5}Nd_{0.5}$ (Figure 5, *a*). The temperature of the observed transition „metal–insulator" on the curve $t(T)$ for this sample is close to the temperature below which the ferromagnetic contribution appears on the curve $M(T)$ (Figure 3). A magnetic field influences an impact on the light transmittance below $T = 150$ K, i. e. a negative effect of magnetic transmittance is observed (Figure 5, *b*). The magnitude of the effect at $E = 0.11$ eV in a magnetic field of 8 kOe reaches the highest (in absolute magnitude) value at 125 K and is 5%. The effect of magnetotransmittance decreases with an increase of energy, but it does not disappear completely in the measured spectral range up to 0.5 eV but amounts to 2%. A similar wide spectral range of the existence of the magnetotransmittance effect with close MT values was observed in the manganites of $PrBaMn_2O_6$and $NdBaMn_2O_6$ in the case of a state with partial ordering at A-position [14].

The dependence $t(T)$ is also nonmonotonic for manganite with $Nd_{0.5}Sm_{0.5}$, but the changes are significantly weaker than in $Pr_{0.5}Nd_{0.5}$, and the influence of the magnetic field is weak: the magnitude of magnetotransmission does not exceed 0.5%. It was shown earlier in [14,15] that in the case of a A-position ordered state, manganite with such a composition of $Nd_{0.5}Sm_{0.5}BaMn_2O_6$ has a sharp transition from a state of „bad metal" type to an insulating state below 340 K, associated with the appearance of an orbital ordering, and light transmission changed 10 times, and a transition to „metallic" state $t(T)$ is observed at temperatures below ~ 200 K, in case of the change of the type of orbital ordering. It can be seen that although the changes of the temperature dependence of the transmission of the sample we studied with $R =$ $Nd_{0.5}Sm_{0.5}$, which has approximately 50% A-site ordering, are weaker than the changes of the temperature dependence in the ordered manganite of the same composition from [14,15], in general, the trend of change $t(T)$ is similar. This turns out to be unexpected, since the studied single crystal and the sample from [14] significantly differ according to the magnetic data. The coexistence of phases with different degrees of ordering by A-position may be one of possible reasons for this situation. In the case of the existence of a phase with „metallic" conductivity, it can manifest itself in the transmission (absorption) of light, even if the volume of this phase is very small [16].

A rather pronounced transition „metal–insulator" at $T = 200$ K is observed for a single crystal of manganite with Sm in the temperature dependence of light transmission. A phase transition with a change of the type of orbital ordering is observed near this temperature in the $SmBaMn_2O_6$ manganite ordered at A position according to data of [14,15,17]. At the same time, there was only a weak „metallic" decrease of light transmission below

$T = 200$ K in the dependencies $t(T)$ in [14,15], and the semiconductor character was again $t(T)$ at lower temperatures. The fact of the existence of a magnetotransmission effect in a sample of a single crystal of manganite with Sm is of particular interest, which reaches $\sim 1.5\%$ at 150 K. Previously, we were unable to detect the effect of a magnetic field on the light transmission in the ordered or highly disordered manganites $SmBaMn_2O_6$. This suggests that the state of the $SmBaMn_2O_6$ manganite, which occurs as a result of growing a single crystal, differs from that formed in case of disordering during annealing of the initial ordered manganites.

4. Conclusion

Structural, magnetic and optical properties of single crystals of double manganites $Pr_{0.5}Nd_{0.5}BaMn_2O_6$, $Nd_{0.5}Sm_{0.5}BaMn_2O_6$ and $SmBaMn_2O_6$ grown using the method of crucibleless zonal melting. It was shown that a state with partial ordering of rare earth ions and barium ions is formed (A-site ordering) as a result of single crystal growing, the relative degree of ordering is estimated as 50%. In this case, partial ordering is realized through a two-phase state and the coexistence of A-position ordered position and disordered phase. This is confirmed by the fact that the X-ray images of the studied single crystals of manganites are satisfactorily described by a mixture of two crystallographic phases with lattice constants corresponding to ordered and disordered manganites. A similar conclusion that the process of disordering in double manganites goes through a two-phase state was previously made in [18], where the evolution of the properties of $PrBaMn_2O_6$ and $NdBaMn_2O_6$ depending on the degree of ordering by A-position as a result of high-temperature annealing. At the same time, it was noted in [18], that the partially disordered state obtained after annealing from the initial ordered state or formed directly as a result of synthesis may differ macroscopically. Therefore, the data obtained in this work are additional evidence that the scenario for the formation of a partially ordered state depends on the method of obtaining it.

The magnetic data indicate that the ferromagnetic contribution to magnetization decreases with a decrease of the average ionic radius of a rare-earth ion, and the coercive force increases, which is associated with an increase of local distortions in the series $Pr_{0.5}Nd_{0.5} \rightarrow Nd_{0.5}Sm_{0.5} \rightarrow Sm$ due to a mismatch of ionic radii of rare earth ions and barium. A change of the character of conductivity was found for all the considered manganites, which can be conditionally called the „metal–insulator“ transition, as well as the influence of the magnetic field on light transmission near the temperature of this transition. The magnitude of the negative effect of magnetic transmission reaches 5% for manganite $Pr_{0.5}Nd_{0.5}BaMn_2O_6$, 0.5% for $Nd_{0.5}Sm_{0.5}BaMn_2O_6$ and 1.5% for $SmBaMn_2O_6$. The fact that the effect of magnetic transmission was found in a single crystal of partially ordered $SmBaMn_2O_6$, whereas previously the effect of magnetotransmission was not observed in samples of the same composition with partial ordering obtained by high-temperature annealing, confirms the above assumption about the difference in the internal structure and distribution of regions with different degrees of ordering depending on the technology of transition to a partially disordered state.

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Conflict of interest

The authors declare that they have no conflict of interest.

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