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Excitation and emission spectra of $LaInO_3$-based solid solutions doped with Sm^{3+}, Sb^{3+}

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ABSTRACT

X-ray analysis showed that all the ceramic samples of $La_{1-x}Sm_xInO_3$ ($0.010 \le x \le 0.025$) solid solutions were single-phased but the samples of nominal composition of $LaIn_{0.98}Sb_{0.02}O_3$, $La_{0.98}Sm_{0.02}In_{0.98}Sb_{0.02}O_3$ contained a small amount of impurity phase of $LaSbO_3$-based solid solutions. It was established that $La_{0.98}Sm_{0.02}InO_3$ solid solution under the excitation of 275 nm and 320 nm exhibits the strongest photoluminescence among $La_{1-x}Sm_xInO_3$ solid solutions with $0.010 \le x \le 0.025$. Photoluminescence bands located in wavelength ranges of 550–580 nm, 585–625 nm and 630–680 nm exhibit 2–3 clear maxima each. According to the locations of these maxima we calculated the magnitudes of Stark splitting of $^6H_{5/2}$, $^6H_{7/2}$, $^6H_{9/2}$ multiplets of the main ^6H term of Sm^{3+} ion by crystal field of $La_{1-x}Sm_xInO_3$ solid solutions with orthorhombically distorted perovskite structure. It was established that the intensity of PL spectra obtained at $\lambda_{ex}=320$, 405 and 470 nm is significantly higher for sample of $La_{0.98}Sm_{0.02}In_{0.98}Sb_{0.02}O_3$ nominal composition than that of $La_{0.98}Sm_{0.02}InO_3$ solid solution. It could be explained by sensitizing effect of Sb^{3+} ions on Sm^{3+} ions photoluminescence or by higher PL intensity of Sm^{3+} ions of impurity phase than of $LaInO_3$ matrix.

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1. Introduction

In the past ten years much attention was drawn to optical properties of perovskite $LaInO_3$-based solid solutions doped with RE ions (RE=Pr^{3+}, Sm^{3+}, Eu^{3+}, Tb^{3+}) and/or Bi^{3+} ion because of their visible light emission [1–4]. Substitution of La^{3+} ions by Bi^{3+} ions in $LaInO_3$: Eu^{3+} shows sensitizing effect on Eu^{3+} luminescence [3]. Sb^{3+} ions have $5s^2$ electron configuration similar to $6s^2$ electron configuration of Bi^{3+} ions. So Sb^{3+} ions are expected to be efficient sensitizer of RE ions. Luminescent properties of Sb^{3+} and Bi^{3+} ions in $LnBO_3$ (*Ln*=Sc, Y, La, Gd, Lu) were investigated in [5–10]. In these compounds Sb^{3+} and Bi^{3+} ions are located in Ln^{3+} ions sublattice. Bi^{3+} ions are sensitizers of Eu^{3+} luminescence in (Y, Gd)BO_3 and energy transfer is $Bi^{3+} \rightarrow Gd^{3+} \ldots Gd^{3+} \rightarrow Eu^{3+}$ [6,7]. At the same time in the work [8] no energy transfer between Sb^{3+} ions and Eu^{3+} ions in YBO_3-based solid solution was observed and both Sb^{3+} and Eu^{3+} ions acted as co-activators. There is no research devoted to luminescent properties of Sb^{3+} ions located in In^{3+} ions sublattice of $LaInO_3$. Such a substitution is possible because Sb^{3+} ionic radius is only 0.02 Å less than that of In^{3+} ($r_{In^{3+}}=0.92$ Å [11]) and 0.14 Å less than that of La^{3+} ($r_{La^{3+}}=1.04$ Å [11]). In the present work we investigate excitation and emission spectra of $LaIn_{0.98}Sb_{0.02}O_3$, $La_{0.98}Sm_{0.02}In_{0.98}Sb_{0.02}O_3$ and $La_{1-x}Sm_xInO_3$ ($0.010 \le x \le 0.025$) solid solutions. All $LaInO_3$-based solid solutions were synthesized by solid-state reaction method and had the structure of orthorhombically distorted perovskite.

2. Experimental

$LaInO_3$ indate and $La_{1-x}Sm_xInO_3$ (x=0.010, 0.015, 0.020, 0.025), $LaIn_{0.98}Sb_{0.02}O_3$, $La_{0.98}Sm_{0.02}In_{0.98}Sb_{0.02}O_3$ solid solutions ceramic samples were synthesized by solid-state reaction method using mixture of La_2O_3 (99.99%), Sm_2O_3 (99.99%), In_2O_3 (99.99%) and Sb_2O_3 (99.99%) oxides. La_2O_3 and Sm_2O_3 were preheated at 1273 K for 1 h. Stoichiometric amounts of the reactants were mixed with the aid of ethanol, ground in planetary mill (Pulverizette Fritch) in cups with zirconia balls and then pressed in pellets (D=25 mm, h=5–7 mm). The pellets were sintered at 1523 K for 6 h on the Al_2O_3 substrate. The pellets of different composition were not in contact with each other. In order to prevent the pellet-substrate

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interaction the pellets were separated from the substrate by thin powder layer of the same composition. Then the pellets were ground, milled and pressed in bars (5 × 5 × 30 mm). The bars were finally sintered at 1523 K for 6 h. The compounds were characterized by powder X-ray diffraction (XRD) analysis (Bruker D8 Advance) at room temperature using CuKα radiation. Crystal structure parameters of the investigated compounds have been calculated using X-ray structure tabular processor (RTP). SEM-images of indates were obtained with scanning electronic microscope JEOL JSM-5610LV with assistance of Energy Dispersive X-ray Spectrometer JED 22-01. Excitation and emission spectra of ceramic samples were recorded at 300 K using automatic spectrofluorometer SDL-2 composed of MDR-12 high-aperture excitation monochromator and MDR-23 recording monochromator at the Institute of Physics of the National Academy of Sciences of Belarus. Xe-lamp DKsSh-120 was used as excitation source.

3. Results and discussion

XRD patterns of $LaInO_3$ indate and $La_{1-x}Sm_xInO_3$ ($x=0.010$, 0.015, 0.020, 0.025) solid solutions (Fig. 1a) showed that all the samples were single-phased and had the structure of orthorhombically distorted perovskite ($GdFeO_3$-type, $a < c/\sqrt{2} < b$ [12]). Crystal structure parameters a, b, c and crystal cell volume V are presented in Table 1. Samples with nominal composition of $LaIn_{0.98}Sb_{0.02}O_3$ and $La_{0.98}Sm_{0.02}In_{0.98}Sb_{0.02}O_3$ had on their XRD patterns (Fig. 1a) not only the peaks corresponding to main perovskite phase but also one impurity peak of small intensity ($d=3.104$ Å, $2\Theta=28.76°$ for $La_{0.98}Sm_{0.02}In_{0.98}Sb_{0.02}O_3$ sample; $d=3.111$ Å, $2\Theta=28.70°$ for $LaIn_{0.98}Sb_{0.02}O_3$ sample). This reflex is probably referred to not-reacted Sb_2O_3 oxide or to intermediate

Table 1
Cell parameters (a, b, c), cell volume (V) and orthorhombical distortion degree (ε) for $LaInO_3$ and $LaInO_3$-based solid solutions doped with Sm^{3+}, Sb^{3+}.

Composition	Cell parameters					$c/\sqrt{2}$,Å
	a, Å	b, Å	c, Å	V, Å^3	$\varepsilon \cdot 10^2$	
$LaInO_3$	5.738	5.953	8.227	281.0	3.75	5.817
$La_{0.99}Sm_{0.01}InO_3$	5.732	5.943	8.228	280.3	3.68	5.818
$La_{0.985}Sm_{0.015}InO_3$	5.731	5.944	8.228	280.3	3.72	5.818
$La_{0.98}Sm_{0.02}InO_3$	5.732	5.944	8.223	280.2	3.70	5.814
$La_{0.975}Sm_{0.025}InO_3$	5.724	5.939	8.224	279.5	3.76	5.815
$La_{0.98}Sm_{0.02}In_{0.98}Sb_{0.02}O_3$	5.731	5.932	8.229	279.8	3.51	5.819
$LaIn_{0.98}Sb_{0.02}O_3$	5.735	5.937	8.234	280.3	3.52	5.822

$LaSbO_3$ compound which is formed during synthesis. The most intensive reflexes d parameters for that samples are [13,14] 3.151 and 3.196 Å, respectively. In order to identify the origin of the impurity phase in agate mortar there was made a mixture of $La_{0.98}Sm_{0.02}InO_3$ and Sb_2O_3 compounds with Sm^{3+}: Sb^{3+} ions ratio 1:1. On the XRD pattern of the mixture no peak of Sb_2O_3 phase was observed. The $LaIn_{0.98}Sb_{0.02}O_3$ sample was additionally sintered at 1523 K for 6 h. The impurity reflex intensity on the XRD pattern of additionally sintered sample did not decreased and remained the same (Fig. 1a). So the impurity reflex is probably referred to $LaSbO_3$ and $La_{1-y}Sm_ySbO_3$ solid solutions on its base. The ratio if intensities of the highest reflexes of the impurity phase ($2\Theta=28.70°$) and the main phase ($2\Theta=30.65°$) shows that in the $LaIn_{0.98}Sb_{0.02}O_3$ sample the amount of impurity phase is about 5% of the main phase. On SEM-images of $La_{0.98}Sm_{0.02}InO_3$, $La_{0.98}Sm_{0.02}In_{0.98}Sb_{0.02}O_3$, $LaIn_{0.98}Sb_{0.02}O_3$ ceramic samples (Fig. 1b) no sign of other phase was observed. Grain size was estimated to be about 0.5–3 μm.

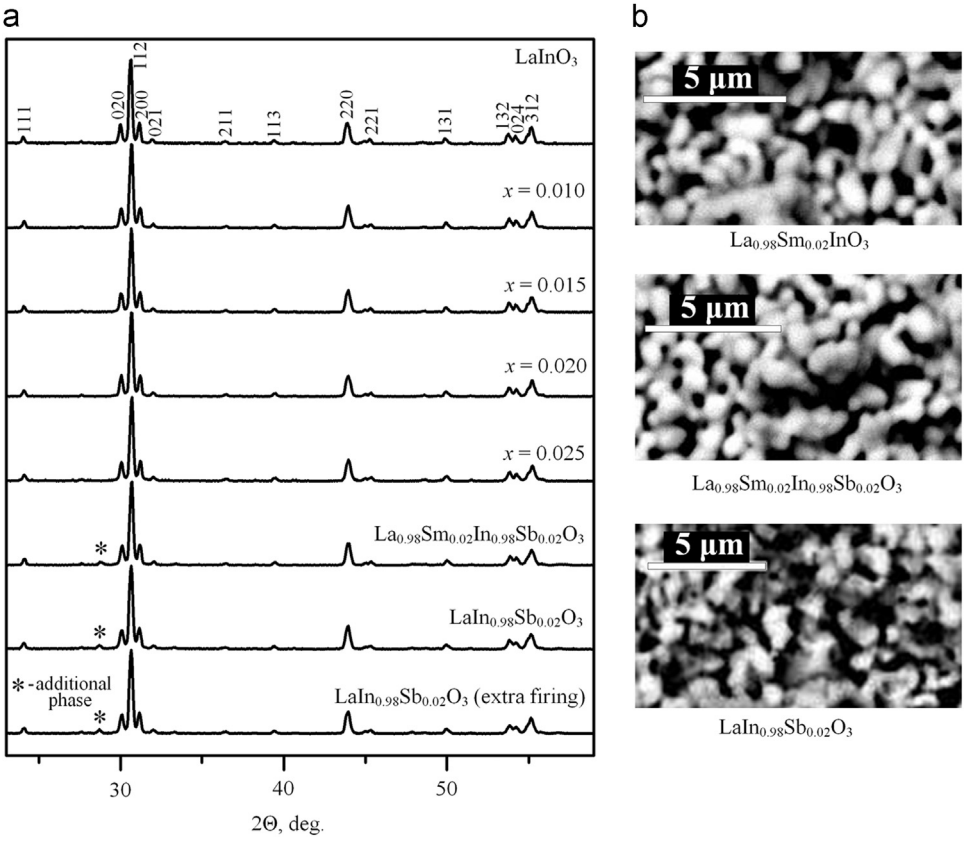

Fig. 1. XRD patterns of $LaInO_3$, $La_{1-x}Sm_xInO_3$ ($x=0.01$, 0.015, 0.02, 0.025), $La_{0.98}Sm_{0.02}In_{0.98}Sb_{0.02}O_3$, $LaIn_{0.98}Sb_{0.02}O_3$ (a); SEM-images of $La_{0.98}Sm_{0.02}InO_3$, $La_{0.98}Sm_{0.02}In_{0.98}Sb_{0.02}O_3$ and $LaIn_{0.98}Sb_{0.02}O_3$ ceramic samples (b).

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