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The Czochralski growth and characterization of $SrLaGa_3O_7$: Ho^{3+} single crystals

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ABSTRACT

Rare – earth (Nd, Pr, Dy) doped $SrLaGa_3O_7$ single crystals are promising laser materials. Also crystals doped with Ho^{3+} ions could be used as efficient laser in visible and near infrared regions. Laser materials, generating in visible and infrared regions, can be used for optical data recording in microphotolitography and in medicine.

$SrLaGa_3O_7$ single crystals doped with 0.3, 1.5 and 2 at % of Ho^{3+}, respectively were grown by the Czochralski method with use of iridium crucible and afterheater. According to EPMA measurements distribution coefficient of Ho^{3+} in $SrLaGa_3O_7$ was estimated to be $k \cong 0.22$. Optical absorption spectra in visible and infrared regions were measured at 300 K and 12 K, respectively.

Optical quality of single crystals was checked by the use of computerized imaging spectropolarimeter and polariscopic measurements. Temperature dependence of capacitance and conductivity for different dopant concentrations were also measured.

Keywords: crystal growth, oxide compounds, laser materials, doping.

1. INTRODUCTION

Recently, solid-state laser materials, which generate in blue and UV regions are of great interest due to their potential applications for optical data storage, in microphotolitography and medicine. Moreover, lasers with up-conversion mechanism pumped with gallium arsenide infra-red diode lasers can transfer energy to shorter wavelengths (visible).

According to the energy level scheme of Ho^{3+} ion it can be concluded that there are metastable energy levels which correspond to emission transitions in the region from near infrared to ultraviolet.

It is known that Ho^{3+} ion is responsible for emission near 2 and 2.9μm regions[1,2] which correspond to transitions between $^5I_2$ and $^5I_6$ Stark excited levels and the ground level $^5I_8$.

There are many works on Ho^{3+} ion behavior in different host materials, and especially in fluorides and in glasses. In the case of oxide materials only for YAG[3] and YAP and YSGG[4] energy level positions of the Ho^{3+} ion were determined and up-conversion excitations were investigated for different dopant concentrations.

$SrLaGa_3O_7$ single crystals exhibit advantageous spectroscopic properties, and first of all low probability of non-radiative transitions, and therefore, long fluorescence lifetime of the excited states.

In this work the grow conditions in the Czochralski technique of $SrLaGa_3O_7$ single crystals doped with Ho^{3+} ions have been elaborated and optical and spectroscopic properties of the obtained crystals have been determined.

2. EXPERIMENTAL

The charge material was prepared starting from $SrCO_3$, La_2O_3, Ga_2O_3, and Ho_2O_3 (at least 4N purity) according to the formula $Sr_{1.04}La_{0.92}Ho_{0.03}Ga_{3.02}O_7$ (for 0.3at. % Ho^{3+} concentration in the charge). Ho was substituted for La in the charge. The following dopant concentrations were introduced into the charge 0.3, 1.5 and 2 at. %.

Single crystals were grown by the Czochralski method with the use of MSR-2 equipment (Metals Research Limited, England). Thermal system with iridium crucible of 50mm in diameter and height, respectively, and the passive iridium afterheater of 50mm in diameter was used.

The following growth conditions have been applied:

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- pulling rate 1 – 2mm/h,
- rotation rate 30 – 40 rpm,
- growth direction [001],
- atmosphere: N_2.

Single crystals 20mm in diameter, with length up to 70mm were obtained.

The optical quality of crystals was determined by the polariscopic and polarimetric methods. Computer controlled imaging spectropolarimeter was used for studying of birefringence. The absorption spectra at temperatures 12 and 300K were measured using the Bruker IFS 113V FT-IR spectrometer.

Electrical conductivity and capacitance were measured as a function of temperature for undoped and holmium doped specimens (with 0.3, 1.5 and 2 at. %). Hewlett Packard LCR Meter type HP 4263B was used. The measurements were performed in resistivity furnace in air atmosphere at f=1kHz.

3. OPTICAL INVESTIGATIONS

For optical measurements samples were cut perpendicularly to the growth direction [001] and were both sides optically polished. Absorption spectra at temperatures of 12K and 300K were measured with a Bruker IFS 113V FT-IR spectrometer. They are shown in Figs.1 and 2.

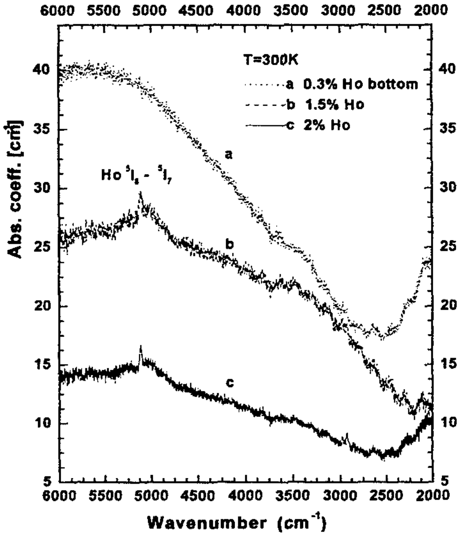

Fig.1.

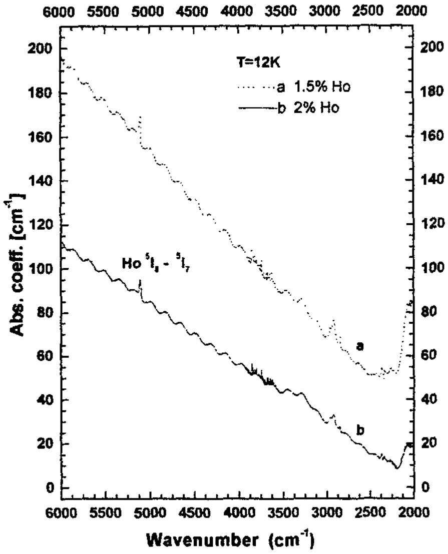

Fig.2.

Fig.1 and Fig.2. Absorption coefficient vs. wavenumber for $SrLaGa_3O_7$: Ho at temperatures of T=300K (Fig.1) and T=12K (Fig.2).

The crystal and wafers have been checked for their optical properties in the macroscale in the plane and circular polariscopes, an automated polarimeter capable of measuring the three maps on wafer area (birefringence, the principal azimuth (one of the principal residual stresses in the case of Z-cut wafers), and transmission)[5,6], and on automated spectropolarimeter used for mapping of parameters associated with birefringence dispersion on wafer area[7].

In this work due to the shortage of space we show only some characteristic results. In Fig.3 one can see the circular polariscope pictures of the wafer cut out from the top (Fig.3a), and from the bottom (Fig.3b) parts of the crystal doped with Ho 0.3 at.%. Besides the crack one can not observe any particular defects in Fig.3a. On the other hand, however, a core region is clearly visible in Fig.3b at the center of the wafer originating from the bottom part of the crystal. Moreover, one can also see the characteristic growth ridges on this picture at the perimeter of the wafer. The core region itself, as well as, the growth ridges and their neighborhood are the areas of mechanical stress, whereas the remaining part of the wafer is almost free from layer residual stress. Such is also the wafer cut from the top part of the crystal (Fig.3a) where only a few traces of the growth ridges can be observed.

In Fig.4 one can see the horizontal cross-section (along the horizontal diameter) of the BCD map (not shown in this work) measured for 770 nm wavelength in the automated spectropolarimeter. In the case of Z-cut wafer, like here, the BDC (Birefringence Dispersion Coefficient) equals to the ratio of the stress-optic coefficients[8], i.e.

$$BDC = \frac{Ci(\lambda_i)}{C(\lambda_{i+1})}$$

where λ_i is 770 nm, and λ_{i+1} is incremented by 10 nm. A good, quasi-radial BDC distribution seen on this figure, as well as on BDC maps, and cross-sections of another wafers well coresponding with the plane and circular polariscope pictures, also showing a quasi-radial residual stress distributions. However in the remaining wafers one could always observe the core region of their centers.

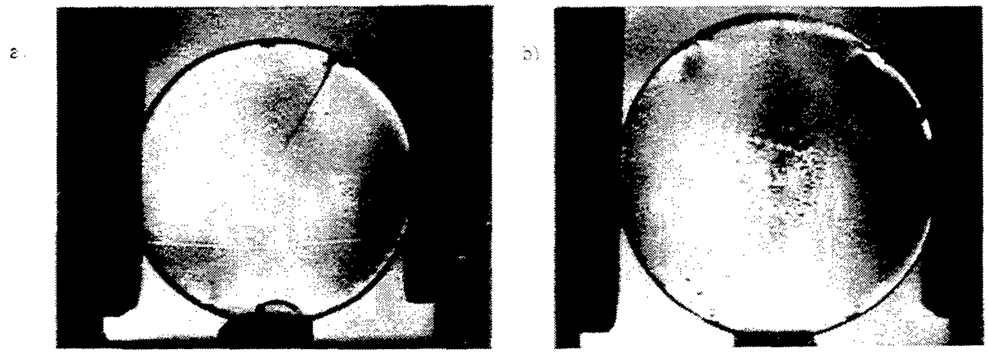

Fig.3. Circular polariscope pictures (quarterwave plates crossed) for 656nm wavelength of the wafer cut out from the top (Fig.3a), and the bottom (Fig.3b) parts of $SrLaGa_3O_7$ no 1 crystal doped with Ho 0.3 at.%

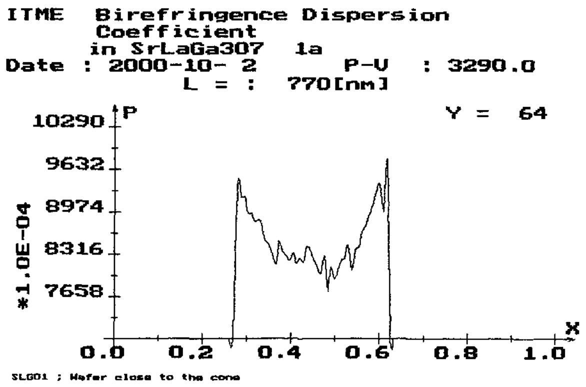

Fig.4. Horizontal cross-section of the BDC map (unshown in this work), corresponding to Fig.3a.

4. CONDUCTIVITY AND CAPACITANCE MEASURMENTS

Temperature dependence of capacitance and conductivity of $SrLaGa_3O_7$ single crystals for different holmium concentrations was also measured. In Fig.5a conductivity dependence versus temperature is presented for undoped and $SrLaGa_3O_7$: Ho single crystals. For comparison, in Fig. 5b the similar dependence for other rare-earth dopants is presented. The band gap value calculated from the slopes of the presented curves is equal to about 2.4eV for undoped and rare-rarth doped specimens.

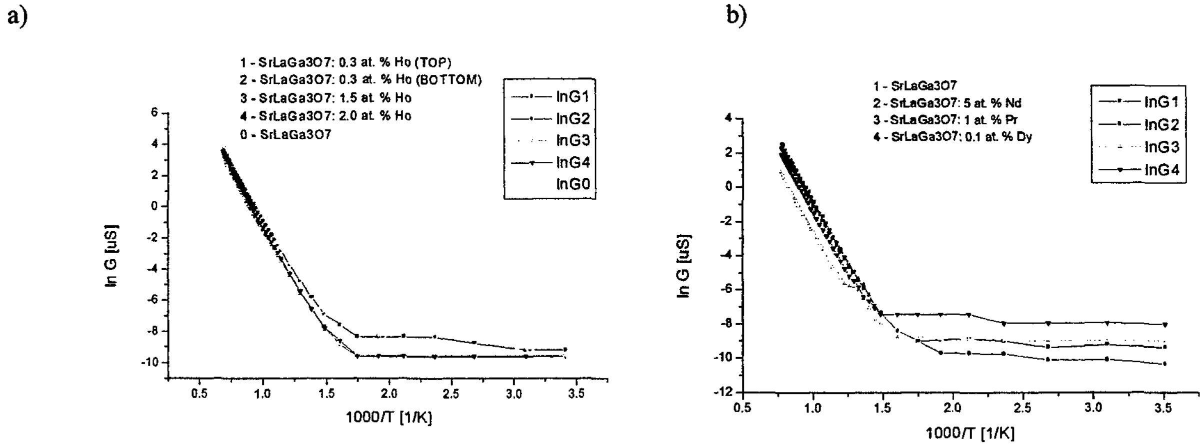

Fig. 5. Conductivity of $SrLaGa_3O_7$ in dependence on reciprocal temperature. (Fig. 5a – for holmium doped crystals, Fig. 5b – for other rare -earth dopands).

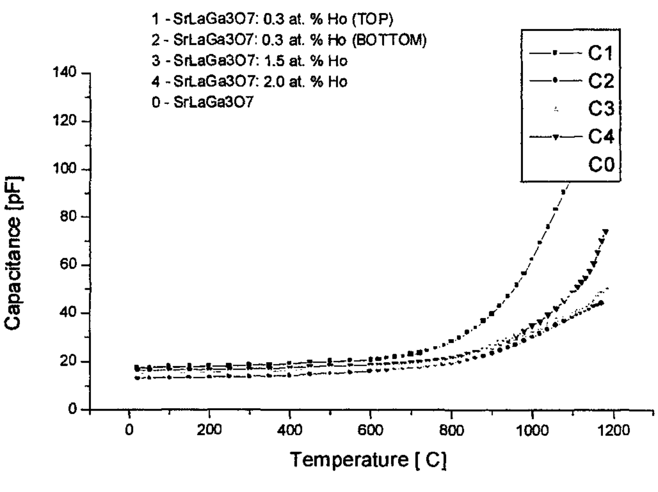

Fig. 6. Capacitance of $SrLaGa_3O_7$ in dependence on temperature.

5. CONCLUSIONS

Single crystals of $SrLaGa_3O_7$ doped with holmium were obtained by the Czochralski method. Optical homogenity of crystals was examined by computer controlled imaging polarimater.

The optical absorption at the temperature 300K and 12K was measured. It can be seen that more defective structure is observed for crystals with higher concentration of holmium. These results are well consistent with EPMA investigations which suggest that distribution coefficient of Ho in SrLaGa3O7 is relatively low and was estimated as close to $k\cong0.22$. Therefore, due to dopant rejection at interface its concentration increases during growth and crystal quality decreases, especially at the tail part.

According to conductivity measurements as function of temperature it can be concluded that doping with holmium and other rare-earths does not influence the conductance properties of material. Dielectric constant is stable and independent on temperature till 600^0 C.

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