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D. KASPROWICZ, M. DROZDOWSKI, A. PAJACZKOWSKA*, P. ZIOBROWSKI, A. KLOS*

Institute of Physics, Poznan University of Technology, Poland
*Institute of Electronic Materials Technology, Warsaw, Poland

Anisotropy of the Elastooptic Properties of SLA and SLG Crystals Studied by Brillouin Scattering

The influence of annealing of $SrLaAlO_4$ (SLA) and $SrLaGaO_4$ (SLG) crystals on their elastooptic properties has been reported. The results obtained are discussed in terms of the nature of the oxygen point defects which might be created in the crystals during the growth process.

Keywords: oxides, defects, Brillouin scattering.

1. Introduction

$SrLaAlO_4$ (SLA) and $SrLaGaO_4$ (SLG) crystals are promissing substrates for high temperature superconducting thin films. They crystallize in the perovskite-like, tetragonal $KNiF_4$-type structure of I4/mmm space group. The structure of the crystal is built up of layers of AlO_6 (GaO_6) octahedra formed in a-b plane. Between them are situated two terminal oxygens in the C_{4v} (O2 position) sites and the remaining four in the D_{2h} (O1 position) sites. Between the layers the Sr^{2+} and La^{3+} ions are randomly distributed in nine coordinated sites of C_{4v} symmetry with equal probability (RÜTER; BRITTEN et al.) (Fig.1).

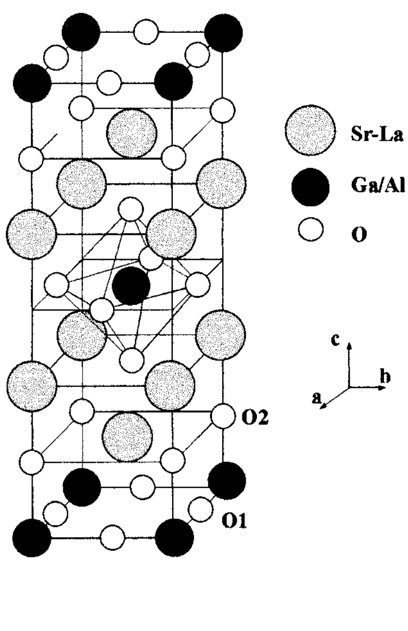

Fig.1: Schematic illustration of the unit cell of $ABCO_4$ crystals.

During last few years crystal structure and physical properties of SLA and SLG have been studied intensively using various experimental techniques (RYBA-ROMANOWSKI et al.; BYSZEWSKI et al.). Their elastic and elastooptic properties were investigated using Brillouin scattering method (DROZDOWSKI et al. 1996, 1997). Elastooptic constants of as - grown SLA and SLG crystals were reported earlier (DROZDOWSKI et al. 1997). In this work we investigated the influence of annealing on the elastooptic properties of SLA and SLG crystals. We have compared the values of the elastooptic constants for phonons propagating in the directions, which can be affected by the existence of the oxygen point defect (Table1).

2. Experimental

The Brillouin polarized spectra were taken in the standard 90^0 scattering geometry at room temperature. As a source of exciting light we used 488 nm line of an argon laser operating on a single mode. The scattered light was analysed through a piezoelectrically driven Fabry - Perot interferometer (BLASZCZAK et al.). The overall *finesse* (free spectral range devided by the instrumental full width at half maximum of the incident line) achieved was not less than 50. The calculations of the parameters of Brillouin lines were carried out using curve - fitting method. To ensure greater reproducibility we made a minimum 5 separate recordings for each scattering geometry.

SLA and SLG crystals used in our experiment were grown by Czochralski method (GLOUBOKOV et al. 1995). Samples of very good optical quality were cut in appropriate directions according to the imposed selection rules. For our measurements we used samples of sizes 5mm x 4mm x 3mm, with faces perpendicular to the [100], [010], [001], [110] directions.

The scattering geometry was determined by the directions of the incident and scattered light wave vectors and the difference between them, the acoustic wave vector **q**. The proper polarization of the incident and scattered light allowed us to distinguish longitudinal or transverse mode of the acoustic wave propagating in the **q** direction. In order to obtain the values of the elastooptic constants we used the substitution technique (CUMMINS, SCHOEN; NELSON, LAX) in which the spectrum of the crystal under study is compared with that of a standard scatterer. For this purpose we used the longitudinal mode of the acoustic wave propagating in the [100] direction of quartz for which a value of the elastooptic constant $p_{31} = 0.258$ is available from the literature (CUMMINS, SCHOEN). The accuracy in the estimation of the elastooptic constants p_{44} and p_{66} was 2 %.

Good quality yellow-coloured as - grown SLA and SLG single crystals were subjected to heat treatment at different temperatures and oxygen pressures. The oxygen pressure in the flowing nitrogen gas was constant with an accuracy of 1% during an annealing experiment. The Brillouin spectra were taken for the same scattering geometries for as - grown crystals and than for the same samples after annealing at 650°C and $p(O_2) = 8\cdot10^{-4}$ atm, 620°C and $p(O_2) = 10^{-2}$ atm, at 820°C and $p(O_2) = 10^{-2}$ atm and after the period of three month since last annealing. In every case samples were annealed for 16h and then slowly cooled down to 100 ^{0}C over a period of 6 h.

3. Results

The obtained values of the elastooptic constants p_{44} and p_{66} for as-grown crystals and after each annealing in mentioned above temperature and oxygen pressure conditions are summarized in the Table 1. The last column in Table 1 includes the values of the elastooptic constants p_{44} and p_{66} obtained for SLA and SLG crystals three month since last annealing.

For the as-grown yellow coloured SLG crystal we observed an anisotropy in the elastooptic interaction for the transverse modes of the acoustic waves propagating in q_1 = [100] and q_2 = [010] directions with polarization u = [001] and in q_3 = [101] and q_4 = [011] directions with polarization u = [010] and u = [001], respectively. The Brillouin spectra obtained for as-grown samples of SLA and SLG crystals are presented in Figs. 2 and 3, respectively.

Crys-tal	p_{ij}	q	u	as grown	650°C $8\cdot10^{-4}$atm	620°C 10^{-2}atm	820°C 10^{-2}atm	three month since last annealing
	p_{44}	q_1=[-100]	[001]	0.0452	0.0445	0.0450	0.0435	0.0433
SLA	p_{44}	q_2=[010]	[001]	0.0447	0.0450	0.0456	0.0440	0.0442
	p_{66}	q_3=[101]	[010]	0.0500	0.0488	0.0478	0.0479	0.0481
	p_{66}	q_4=[011]	[100]	0.0505	0.0493	0.0496	0.0489	0.0498
	p_{44}	q_1=[-100]	[001]	0.0497	0.0526	0.0598	0.0490	0.0459
SLG	p_{44}	q_2=[010]	[001]	0.0619	0.0599	0.0605	0.0462	0.0392
	p_{66}	q_3=[101]	[010]	0.0660	0.0760	0.0679	0.0644	0.0643
	p_{66}	q_4=[011]	[100]	0.0562	0.0569	0.0683	0.0510	0.0466

Table1: Elastooptic constants p_{44} and p_{66} of SLA and SLG crystals after annealing at different heat temperatures and oxygen pressures.

Table 1 reveals, that the difference in the value of the elastooptic constant p_{44}, which corresponds to phonons propagating in the q_1 and q_2 directions, decreases from the 20% for as-grown SLG crystal to 1% after annealing at 820 °C and $p(O_2) = 10^{-2}$ atm. Moreover, the difference in the value of the elastooptic constant p_{66}, which corresponds to phonons propagating in the q_3 and q_4 directions, decreases from the 15% for as-grown SLG crystal to 1% after annealing at 620 °C and $p(O_2) = 10^{-2}$ atm. A further increase in these differences after the next annealing at 820 °C and $p(O_2) = 10^{-2}$ atm is also caused by different absorption of the light and is connected with the colour change of the sample. It was observed that there is no change in the colour of SLG after the first annealing at 650°C and $p(O_2) = 8\cdot10^{-4}$ atm, and second annealing at 620 °C and $p(O_2) = 10^{-2}$ atm, while after annealing at 820 °C and $p(O_2) = 10^{-2}$ atm some parts of the volume of SLG crystal changed the colour from yellow to brown. In the case of the q_3 and q_4 directions the light was traversing through the brown region of the crystal.

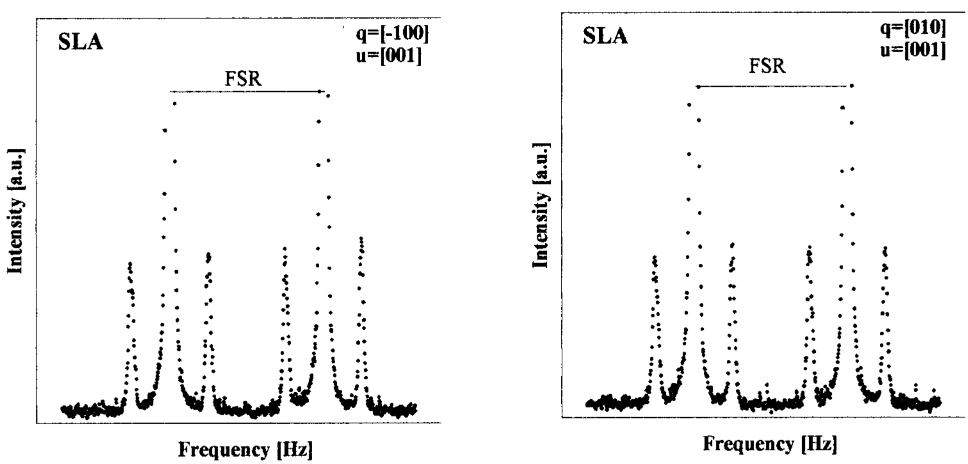

Fig.2: The Brillouin spectra obtained for phonons propagating in a-b plane for as-grown SLA (FSR = 92 GHz).

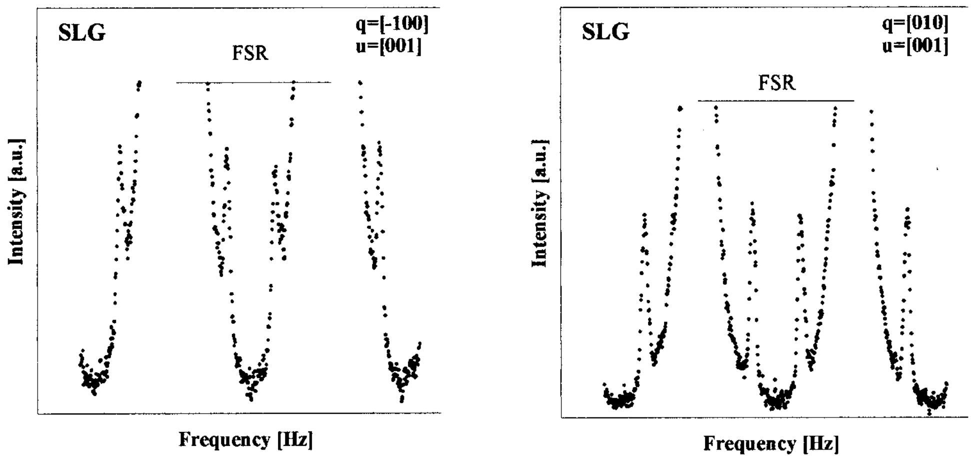

Fig.3: The Brillouin spectra obtained for phonons propagating in a-b plane for as-grown SLG (FSR = 63 GHz) crystals.

It was found that the annealing procedure does not change the colour of SLA. Moreover, it should be also noted that we did not observe an anisotropy in the q_1 and q_2 directions for the as-grown SLA crystal. The difference between the values of the elastooptic constants p_{44} and p_{66} are in the range of experimental error.

4. Discussion

The results obtained are consistent with our previous study of the elastic properties of SLA and SLG crystals (DROZDOWSKI et al. 1996, 1997). We found earlier an anisotropy in the phase velocity of the acoustic wave propagating in a and b directions for the SLG crystal. Moreover, the temperature dependences of hypersound velocities in SLA and SLG crystals obtained for more sensitive directions exhibit discontinuities at the characteristic temperature 460 K (DROZDOWSKI et al. 1997).

Thermal analysis of SLA crystals indicated oxygen deficiency in as-grown crystals but a different behaviour was observed for SLG crystals where a loss in mass during the heating of SLG crystals was detected (BYSZEWSKI et al.). These effects are associated with oxygen point defects which can appear in the lattices of as-grown SLG and SLA crystals (RYBA-ROMANOWSKI et al.; BYSZEWSKI et al.).

The observed anisotropy in the elastooptic properties for as-grown SLG monocrystal during Brillouin scattering study is probably associated with the displacement of the oxygen ions from their original O1 and O2 positions. This phenomenon can lead to the distortion of the oxygen octahedra which consequently leads to a lower symmetry of the crystal structure from the ideal tetragonal I4/mmm.

Moreover, the positions of oxygen ions are sensitive to annealing conditions. A strong influence of annealing on SLG crystals is evident in the disappearance of the anisotropy of the elastooptic properties in the a and b directions after a double annealing procedure. The anisotropy in the elastooptic properties is not observed for SLA crystals. Also, the influence of the annealing process is not observed for SLA. Thus, we can conclude, that SLA crystals are not sensitive to the heat treatment and oxygen atmosphere under the annealing conditions used in this study.

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Authors' addresses:

mgr D. KASPROWICZ, Dr hab. M. DROZDOWSKI,
Dr P. ZIOBROWSKI
Institute of Physics, Poznan University of Technology,
Piotrowo 3, 60-965 Poznan, Poland
E - mail : dobkas@phys.put.poznan.pl
Fax : (061) 8782-324

Prof. dr hab. A. PAJACZKOWSKA, mgr A. KLOS
Instytute of Electronic Materials Technology
Wólczynska 133, 01-919 Warszawa, Poland