Evaluation of optical and acoustical properties of $Ba_{1-x}Sr_xTiO_3$ material library by a multi-technique approach including picosecond laser ultrasonics

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Background – Materials with graded chemical composition have diverse applications, such as laterally graded aperiodic crystals based on $Si_{1-x}Ge_x$ used for radiation path control in high-resolution X-ray monochromators and the out-of-plane graded bandgap semiconductors to improve solar energy harvesting. When films with continuously varying chemical composition are prepared for particular applications via epitaxial growth, their local physical parameters are generally different from bulk materials of the same chemical composition. So, the continuous spatial variation of the materials composition requires developing a high throughput measurement technique to locally and non-destructively characterize the library with high spatial resolution. The typical experimental methods used to examine such material systems are the energy or wavelength dispersive X-ray spectroscopy, high-resolution X-ray diffraction, and Raman spectroscopy/microscopy. However, optical, acoustical, and acousto-optical parameters have never been evaluated in vertical or lateral compositionally graded films. In this work, we report on the first application of the picosecond laser ultrasonics (PLU) technique for the evaluation of acoustical and optical parameters of lateral compositionally graded film, the $Ba_{1-x}Sr_xTiO_3$ ($0 \le x \le 1$) material library [1], denominated BST-x for short in the following.

Methods – An epitaxial 250 nm-thick BST-x ($0 \le x \le 1$) continuous composition spread film library was prepared by combinatorial pulsed laser deposition onto a (001) $SrTiO_3$ 1 cm^2 substrate buffered by a 120 nm-thick $La_{0.9}Sr_{1.1}NiO_4$ electrode. The lateral composition of the BST-x film was characterized by EDS and WDS. Opaque $La_{0.9}Sr_{1.1}NiO_4$ film served as an optoacoustic transducer for launching CAPs in BST-x coating, which was transparent at optical wavelengths of 354, 517, and 535 nm of our laser pulses. We implemented PLU with a fast data acquisition technique based on asynchronous optical sampling.

Results and Conclusions – We demonstrated an application of PLU in combination with XRD, EDS, EPMA, SEM and AFM for the measurements of the dependencies on x of the optical refractive index n and of the sound velocity v in an epitaxially grown thin film BST-x library (Fig. 1). We applied new

protocol for modeling and fitting the PLU signals, which accounts for the significant roughness of the film surface revealed by AFM measurements. The film was not dedicatedly prepared for its opto-acousto-optic evaluation by PLU, exhibiting significant lateral variations in thickness and surface roughness. Therefore, the achieved measurements of the sound velocity and of the optical refractive index, and characterization of the surface roughness confirm the robustness of the PLU technique for thin film evaluation. Our results on the acoustical and optical properties of epitaxial grown BST-*x* library layer by PLU technique accomplished here could provide the parameters required for more extended predictive design of the phononic, photonic and phoxonic mirrors and cavities with superior properties/functionalities for novel multifunctional nanodevices.

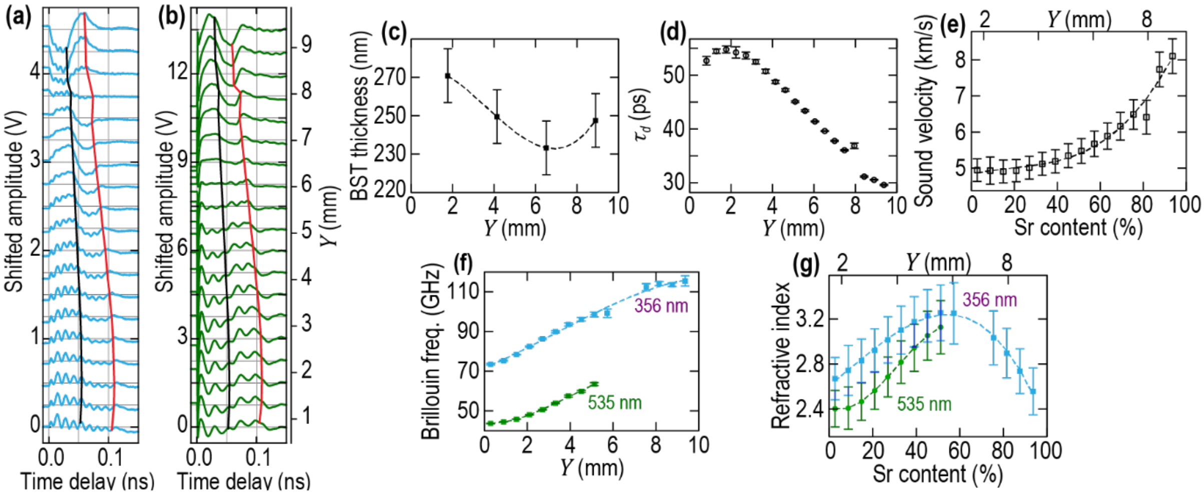

Fig. 1. **(a,b)** Raw transient reflectivity signals detected at different lateral positions of the BST-*x* library using 517 nm pump and either (a) 354 nm or (b) 535 nm probe. The black (red) solid lines connecting the raw signals vertically stand for the CAP arrival times at the free surface of the BST-*x* library layer (at the surface between the BST-*x* library layer and the OAT). **(c)** BST-*x* library thickness as a function of the position along the graded direction. **(d)** Time-of-flight of the acoustic wave propagating from the generator surface through the BST-*x* layer to its free surface estimated using the fitting procedure. **(e)** Sound velocity in the BST-*x* library as a function of the Sr content (bottom horizontal axis) and the position along the graded direction (top horizontal axis) estimated from the data in (c, d). **(f)** Brillouin frequencies as a function of the position along the graded direction. (g) Refractive index in the BST-*x* library as a function of the Sr content (bottom horizontal axis) and the position along the graded direction (top horizontal axis) estimated from the data in (e, f)

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References

[1] S. Sandeep, S. Raetz, J. Wolfman, B. Negulescu, G. Liu, J.-L. Longuet, T. Thréard, V.E. Gusev, Evaluation of Optical and Acoustical Properties of $Ba_{1-x}Sr_xTiO_3$ Thin Film Material Library via Conjugation of Picosecond Laser Ultrasonics with X-ray Diffraction, Energy Dispersive Spectroscopy, Electron Probe Micro Analysis, Scanning Electron and Atomic Force Microscopies, Nanomaterials, 11 (2021) 3131. https://doi.org/10.3390/nano11113131.