

Integration of Single BaTiO₃ and LiNbO₃ Thin Films on Si: Synthesis, Microstructure and Characterization

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Ferroelectric thin film integration with Si and other integrated device substrates has the potential to enable new modes of photonics integration as well as a new class of high work/volume piezoelectric devices for MEMS integration. To date, most ferroelectrics integration efforts have focused either on growth of polycrystalline films with poor control of microstructure or on perovskite film epitaxial growth using the Si substrate or a bulk ceramic substrate as a template. However, realistic integration schemes with Si-based electronic and photonic devices probably will not allow growth on a single crystal silicon substrate, but rather will require perovskite oxide integration on top of amorphous dielectrics that encapsulates integrated circuit interconnects. Our group has thus focused on two approaches for ferroelectric oxide integration on Si using: 1) biaxially-textured MgO templates and 2) direct wafer bonding and layer transfer by ion implantation-induced layer splitting. This paper discussed the synthesis and properties of single crystal LiNbO₃ and BaTiO₃ thin films synthesized by the latter method.

Single crystal thin films of LiNbO₃ and BaTiO₃ on SiO₂/Si(100) have been synthesized via wafer bonding and implantation-induced layer. The layer transfer technique allows for the study of the ferroelectric domain structure and optical properties of thin films of LiNbO₃ and BaTiO₃ with crystal quality approaching that of the parent bulk crystal source material. In this study z-cut LiNbO₃ and (001) BaTiO₃ are used. LiNbO₃ crystals were co-implanted with H⁺ and He⁺ at 80keV with a dose of 1x10¹⁷cm⁻² and 115keV with a dose of 5x10¹⁶ cm⁻², respectively. BaTiO₃ crystals were co-implanted with H⁺ at 80keV with a dose of 1x10¹⁷cm⁻² and He⁺ with a dose of 7x10¹⁶ cm⁻². This creates a H-rich damage layer at a depth of approximately 700 nm based as indicated by electron microscopy and surface step profilometry. Thermal annealing at 320°C at a pressure of > 1 MPa for 5 hours induces layer splitting at the damaged interface. Co-implantation of H⁺ and He⁺ was found to be necessary for layer splitting as H⁺ alone was not observed to blister upon annealing with doses up to 2x10¹⁷cm⁻² at 80keV and temperatures up to 500°C..

The cavity formation that is a precursor to layer transfer occurs in a manner that is qualitatively similar to that observed for layer transfer of semiconductors, such as silicon and germanium. Thermodynamic and kinetic models of the nucleation and growth of cavity with ion implanted single crystal BaTiO₃ layer have been developed for the layer transfer of the ferroelectric thin film. The critical radius (r_{crit}^*) of the cavity is theoretically and experimentally obtained with respect to classical fracture and thermodynamic theories. From thermodynamic considerations, we obtain the cavity inner composition and dominant species present during cavity nucleation and growth, which include H₂, H, H₂O, Ba and BaOH. At the thermodynamic criteria (r_{crit}^*), H₂ molecules accumulate and the cavity begins to grow

toward the saturation radius (r_{sat}^*). Thermodynamic calculations also suggest that the addition of hydrogen to the Ba-Ti-O system can effectively suppress the volatile barium oxide formation during the annealing process.

Transmission electron microscopy of the exfoliated layers indicates that the crystalline quality of LiNbO₃ and BaTiO₃ films are retained during processing. Piezoresponse force microscopy measurements performed in transferred 700 nm BaTiO₃ thin films indicated ferroelectric film response with micron sized domains and domain structure that is quite different from the stripe domain structure in bulk BaTiO₃. Raman spectroscopy measurements indicate that while Raman active BaTiO₃ local vibrational modes are significantly reduced in as-implanted layers as compared with unimplanted substrates, the LO and TO mode intensities recover almost to the intensity seen in unimplanted substrates. Elastic recoil spectrometry ion scattering measurements have been used to profile residual hydrogen in BaTiO₃ following layer transfer, and a significant fraction of the implanted hydrogen remains following layer transfer, but can be reduced by subsequent post-transfer annealing. Transferred 700 nm thick LiNbO₃ thin films are found to be stoichiometric and optically transparent, and exhibit a film surface roughness of approximately 10 nm. Measurements of polarization-field characteristic and planar waveguide optical transmission loss will be presented for LiNbO₃ and BaTiO₃ films, in addition to more comprehensive piezoresponse force microscopy characterization of transferred thin film microstructure. Finally, the potential for ion implantation-induced layer transfer to fabricate high quality thin films suitable for piezoelectric and electro-optic devices will be assessed.