Investigation of Growth Striations in LGS, LGT and LGN Crystals with SWBXT
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Introduction: Langasite (La$_3$Ga$_5$SiO$_{14}$ or LGS) and its two isomorphs Langanite (La$_3$Ga$_{5.5}$Nb$_{0.5}$O$_{14}$ or LGN) and Langatate (La$_3$Ga$_{5.5}$Ta$_{0.5}$O$_{14}$ or LGT) with the Ga$_3$Ga$_2$Ge$_4$O$_{14}$-type structure exhibit piezoelectric properties intermediate between those of quartz and LiNbO$_3$ and LiTaO$_3$. The combination of the properties, with the availability of high quality crystals, makes these crystals currently the most promising piezoelectric materials. X-ray topography enable one to observe the true microstructure developed during the crystal growth process, and is imperative for understanding the nature and distribution of imperfections, mainly growth striations, and enable one to assess the influence of striations, especially on Q factor.

Methods and Materials: A systematic study of the growth striations configuration of LGX crystal boules, wafers and resonators was conducted by examining LGX crystals with SWBXT in both reflection and transmission geometry at various stages throughout the device fabrication procedure, i.e., from as-grown LGX boules to crystal wafers cut, both parallel and perpendicular to the growth axis, from adjacent locations in the boule to resonators made from the crystals. This enables some insight to be gained as to the evolution of the three-dimensional defect microstructure, including the interface shape, during growth.

Results: Figure 1 to 4 is a selection of SWBXT data recorded from LGX crystal as-grown boule, Y-cut and Z-cut wafers and resonator made from a LGX crystal.

Conclusions: Striations are the most significant defects in LGX crystal growth. They are not visible optically, and there is no report of striations revealing in LGX by chemical etching or other techniques. The presence of striations was only revealed by X-ray topography, so far. In LGX, the striations originate due compositional variations or non-stoichiometry in the melt. These can arise due to: (a) the small size or even absence of the field of congruent melting; (b) small but finite evaporation of Ga$_2$O$_3$ from the melt. This non-stoichiometry can also give rise to the effect of constitutional supercooling, which can drive interface breakdown or cellular growth, Figure 3 give out strong evidence. To prevent striations, some possible ways are: (a) carefully prepare starting melt composition; (b) improve temperature control, keep growing proceed at a most exactly stoichiometric melt composition; (c) reduce temperature fluctuation during crystal growth; and (d) reduce evaporation by improving the growth atmosphere. Since striations are associated with slight variations in crystal structure (variations in composition and lattice spacing), they will result in spatial variations of the elastic constants. However, they will not necessarily have an anelastic response to acoustic waves and, therefore, may not affect the Q factor. This depends on whether applied stress induces a slight restructuring of the atoms in the striations. So, a lack of effect on Q factor is possible. Furthermore, striations could affect the temperature independence of oscillators. Local variations of elastic constants are bound to produce changes in the bulk elastic constants (at wavelengths long compared to the periodicity of the striations) and their temperature derivatives. In addition, the effects can be expected to be different for different elastic constants, since striations have a directionality.