Growth and composition control of $Tb_3Ga_5O_{12}$ crystals

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[Introduction]

$Tb_3Ga_5O_{12}$ (TGG) single crystals have excellent characteristics for optical isolators, such as high Verdet constant, high thermal conductivity, and low optical loss. On the other hand, TGG single crystals grown by the Czochralski（Cz）method contain “radial stripes” besides defects such as cores and voids. The radial stripes are extended radially along the growth direction, and are a source of light scattering that degrades the quality of optical isolators. We have researched the radial stripes by polarizing microscope, etching, and X-ray topography, and identified them as giant helical dislocations [1]. Helical dislocations are formed by the interaction of dislocations and a lot of vacancies [2]. We considered that high-density vacancies were caused by deviation of the melt composition from the congruent composition. In this study, we determined the optimum melt composition by precise measurement of the change of TGG’s lattice constant, which were sensitive to the Tb/Ga composition ratio.

[Experiments and Results]

TGG single crystals were grown by the RF induction heating Cz method using an iridium crucible. The ratios of Tb/Ga in the feeds were the stoichiometric, Tb-excess, and Ga-excess compositions. The pulling orientation was <111>. Wafers of 1.0 mm in thickness with (111) plane were cut and polished from initial and final growth parts of grown ingots. Lattice spacing of 444 diffraction was measured by the Bond method using a high-resolution X-ray diffractometer (Rigaku ATX-E) with $CuK\alpha_1$ X-ray source taken through a Ge220 4-bounce monochromator.

The growth interface of the grown TGG was convex to the melt, and a core with three-fold symmetric {211} facets was observed at the center. The lattice constant of the {211} facets region was slightly larger than that of the off-facet region. This phenomenon is thought to be due to the different growth mechanisms between the two regions. For TGG crystals grown from stoichiometric melt, the lattice constant at the final growth part were smaller than that of the initial growth part. The temperature of liquidus and solidus are lower as the composition shifts from the congruent-melting composition. On the other hand, the lattice constant in TGG increases with increasing Tb/Ga ratio. Therefore, the congruent-melting composition is estimated to be a Tb-excess from the stoichiometric composition. TGG crystals were grown from various Tb/Ga ratio in the feeds. The optimum Tb/Ga ratio for the congruent-melting composition was determined by the minimum difference of the lattice parameter between initial and final growth part. TGG single crystals grown from the optimum congruent composition were not observed any giant helical dislocations successfully.

References

[1] Miki Watanabe et al., "Growth and characterization of $Tb_3Ga_5O_{12}$ single crystals", (Invitation to The 8th Asian Conference on Crystal Growth and Crystal Technology, C03-01-02).
[2] J.P. Hirth and J. Lothe, "Theory of Dislocations" McGraw-Hill, New York, (1968).