Crystal perfection and Thermostructural (300-1300K) Properties of β-Ga_2O_3: High-resolution 2D Mapping and Powder Diffraction Studies

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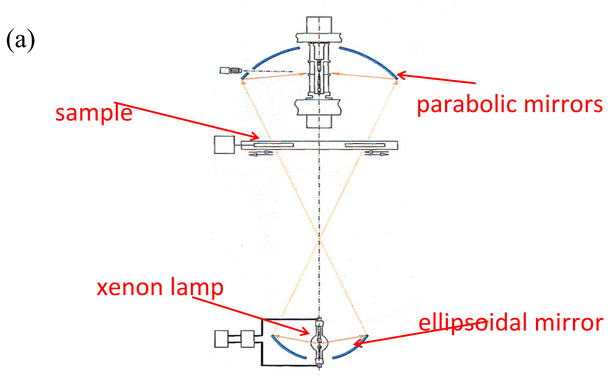
Introduction

Gallium oxide, Ga_2O_3 can crystallize in five polymorphs: *α, β, γ, δ,* and *ε*, with β-Ga_2O_3 (the monoclinic phase) being the most thermodynamically stable phase at ambient conditions and possessing great application potential especially for power electronic devices and high-frequency electronics [1].

In the present work we show the crystal perfection of β-Ga_2O_3 using HRXRD 2-D mapping and also show the high temperature behavior of structure of β-Ga_2O_3 as function of temperature just to understand the physical properties of such material .

Methods



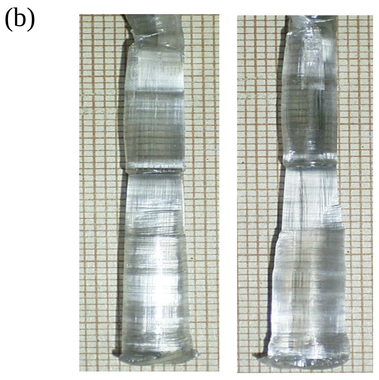


Fig. 1. a) Set-up for the OFZ growth.
b) Ga_2O_3 crystals grown in 010 direction.

The β-Ga_2O_3 crystals were grown using the Optical Floating Zone (OFZ) method (in ambient air) in optical heated furnace capable of operating up to 2000°C. This method is very useful for research applications due to the fact that it does not require a crucible, and doping is relatively easy. In the most of our crystals, the growth direction was [010]. For measurements the crystals were cleaved in (100) and (001) planes and cut along the (010) plane.

The volume crystal that we obtained is split into (100) natural cleavage plane to get a good crystal plate A. Selected reflection 12 0 0 ($2\theta \approx 102.20^{\circ}$) were measured in triple axis geometry (detector coupled with analyzer). We investigate step by step the sample surface doing both ω and 2θ/ω scans at every point.

X-Ray HRD Method 'digital topography'

The sample (half cylindrical column with two natural planes and two planes after cutting which are perpendicular to growth (see figure 5)) surface spot illuminated by the beam is 0.2x0.5 mm^2 (effect of the slit used, slit size 0.1 mm and mask 0.2 mm) where we have: Δx=0.2 mm (n=26 steps) and Δy=0.5 mm (n=8 points).

The segment drawn through the maxima of ω (OmTA) curves provide information about bending of (100) plane for the adjacent measurement points (marked bending radius, concave "+" and convex"-"). In general, on the entire surface of volume crystal, the OmTA curves are narrow i.e. FWHM<30". And near the edges, where grains appear, FWHM reaches 64".And this can also see in "Intensity OmTA (Max) vs XY".

Fig. 2. ωTA scans of (100) plane along the Y=-0.7 showing the curvature of plane and the numbers are showing radius of curvature .

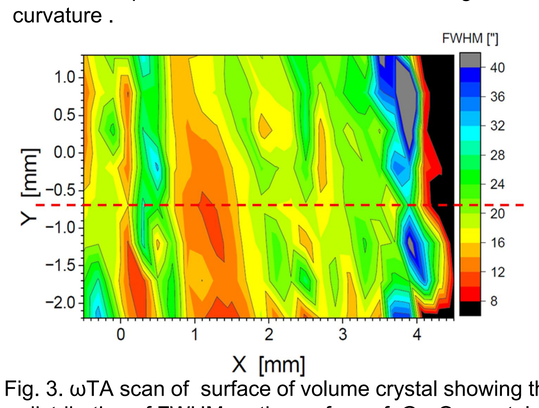


Fig. 3. ωTA scan of surface of volume crystal showing the distribution of FWHM on the surface of Ga_2O_3 crystal.

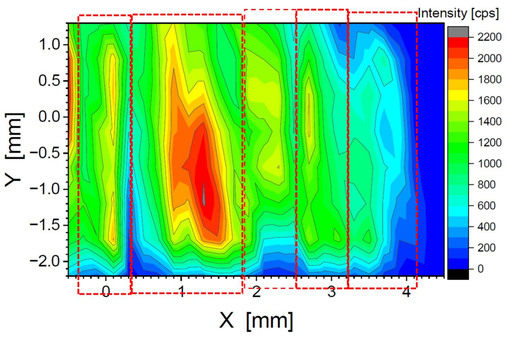


Fig. 4. ωTA scan of surface of volume crystal showing the distribution of Intensity on the surface of Ga_2O_3 crystal.

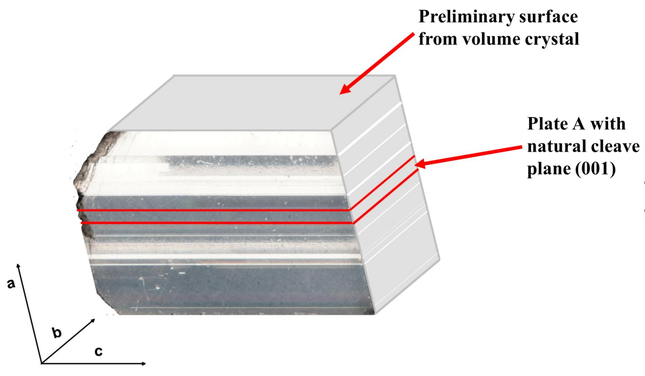


Fig. 5. Ga_2O_3 crystal half cylindrical column and the red lines showing the plate A which is obtained after cutting along c-axis.

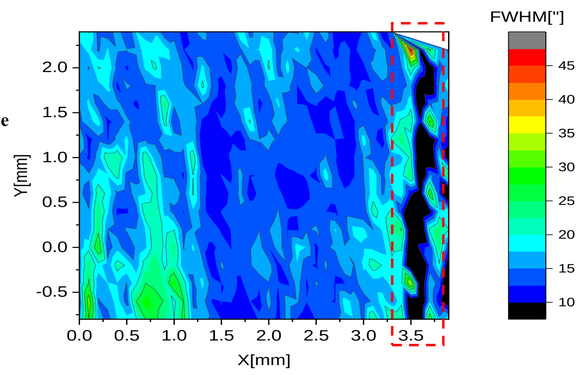


Fig. 6. ωTA scan Plate A of Ga_2O_3 crystal showing FWHM distribution .

The Plate A and A' (back side of plate A) were analysed using the same geometry. The sample surface illuminated by beam is 0.1×0.2 mm^2 (effect of the slit used, slit size 0.1 mm and mask 0.2 mm). We investigate step by step the sample surface where we have: Δx=0.1 mm (n=36 steps for Plate A' and n=40 for plate A) and Δy=0.2 mm (n=19 steps for Plate A' and n=18 steps for Plate A).

The quality of plate A and A' is same with thickness of 1.2 mm. In general, the ωTA curves are narrow over the entire surface i.e. 10<FWHM<25 arc seconds. The region close to 3.5 mm has some defects so it's not of our interest. This is also obvious in the figure of "intensity of ωTA(Max) vs XY". The Intensity is also maximum in the region between 0.0-3.25 mm so we can use this region as substrate for growth of crystal. The quality and homogeneity is really good and the FWHM is comparable with literature [2]. The lattice parameter distribution is uniform over entire surface. The value of *a* is derived from Bragg equation for 12 0 0 plane while considering the monoclinic crystal structure of Ga_2O_3 :

***a = (d*h)/sinβ* (here k, l=0 for 12 0 0)**

the median <*a*>= 12.2334(3)Å.

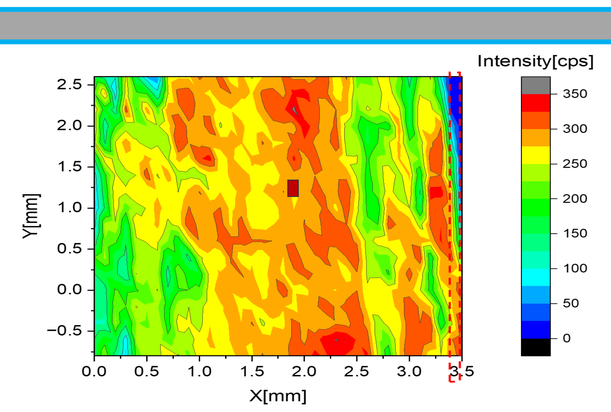


Fig. 7. ωTA scans of Plate A' of Ga_2O_3 crystal showing the distribution of Intensity .

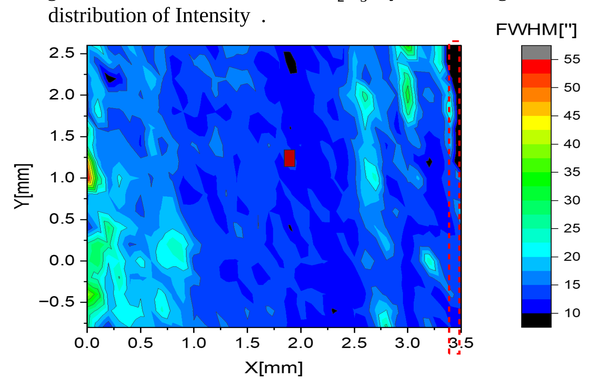


Fig. 8. ωTA scans of Plate A' of Ga_2O_3 crystal showing the distribution of FWHM .

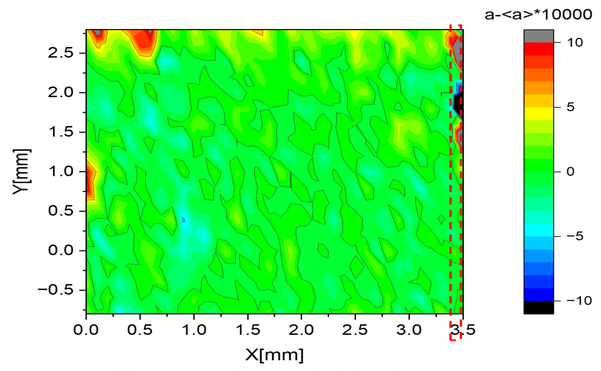


Fig. 9. 2θ/ ω scans of Plate A' of Ga_2O_3 crystal showing the distribution of lattice parameter *a* in terms of standard deviation (*a*-<*a*>*10000) .

Powder X-ray Diffraction and Rietveld Refinement Analysis

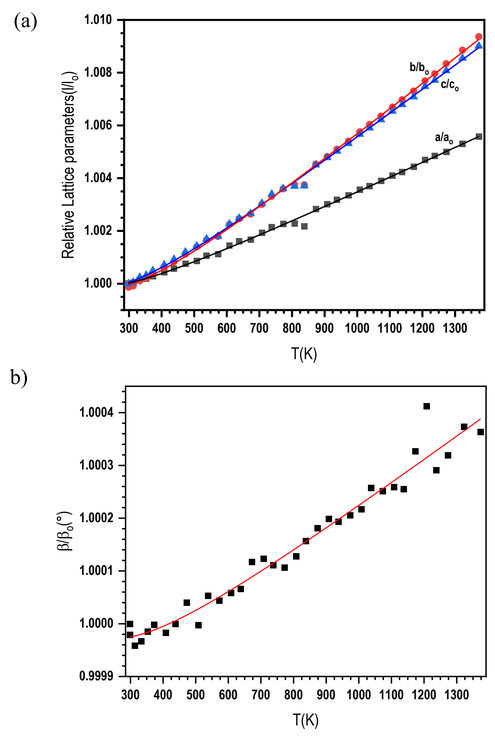
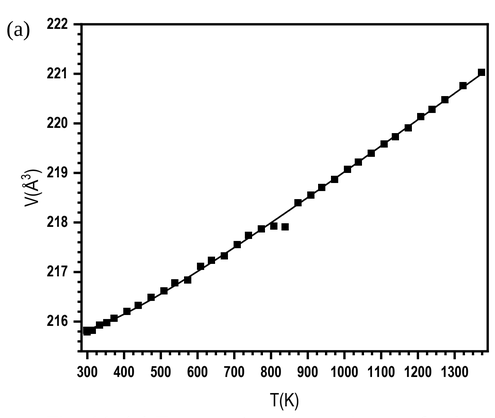


Fig. 10. (a) Temperature dependence of relative lattice parameters (l/l_o) of Ga_2O_3 sample.
(b) Temperature dependence of relative angle *β* of Ga_2O_3 sample.



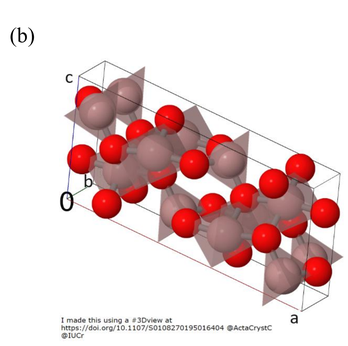


Fig. 11. (a) Temperature dependence of volume of Ga_2O_3 sample.
(b) β-Ga_2O_3 crystals have monoclinic structure with a space group *C2/m*. There is a right angle between the *a* and *b*, as well as *b* and *c* axes. The angle between the *a* and *c* axes is 103.8°.

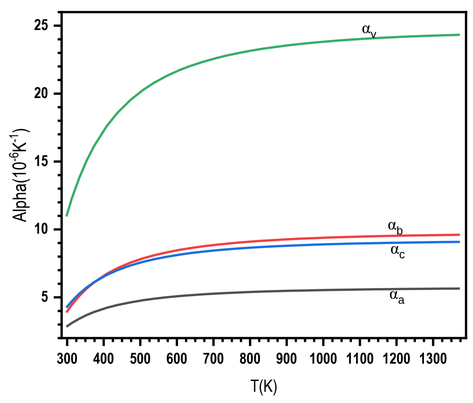


Fig. 12. Showing the variation of $\alpha_{(a, b, c, V)}$ of Ga_2O_3 with temperature

The PXRD and Rietveld refinement analysis shows that β-Ga_2O_3 belongs to monoclinic structure (space group *C2/m*) having unit-cell parameters *a*=12.2256(3) Å, *b*=3.0393(1) Å, *c*=5.8082(2) Å, *β*=103.8058(24) and volume, *V*=209.590(9) $Å^3$ at room temperature. No phase transition or decomposition is observed in whole temperature range. Due to heating the variation in lattice parameters is about 0.60% in *a*, 0.93% in *b*, 0.9% in *c*, 0.04% in *β* and 2.40% in *V* (see figure 9). We observe expansion anisotropy in *b*-direction [3,4].

The Variation in lattice parameters and volume are fitted using Laurent polynomial

$$y = A + B \cdot T + C/T$$

and then thermal expansion coefficient for lattice parameters and volume are calculated for whole temperature range. The TEC values of Ga_2O_3 at room temperature are $\alpha_a = 2.84\times10^{-6}\ K^{1}$, $\alpha_b = 3.88\times10^{-6}\ K^{-1}$, $\alpha_c = 4.24\times10^{-6}\ K^{-1}$ and $\alpha_v = 10.96\times10^{-6}\ K^{-1}$. The change in expansion coefficients of *b* and *c* are practically identical and roughly double that of *a* (See Figure 10).

Conclusions

- The *β*-Ga_2O_3 bulk crystals allowed us to access different crystallographic planes.
- The results of HRXRD " digital topography" of volume crystal shows the bending of plane (100), ωTA curves are narrow with FWHM<30' and the intensity is high.
- A wafer prepared from bulk crystal exhibit superior homogeneity in crystallographic properties (10<FWHM<25, Standard deviation of *a*= 0.0003Å) confirming the high quality of crystal plate A.
- Although the penetration depth of X-ray beam into plate is approximately 20μm, the fact that sides A and A' exhibit a mirror reflection of crystallographic properties indicates that interior of the wafer is also of high quality.
- The *β*-Ga_2O_3 crystalize into *C2/m* space group having monoclinic structure. The XRD results shows no phase transition and the increase in lattice parameters : *a, b, c, β* and *V* are 0.60%, 0.93%, 0.90%, 0.04% and 2.04%.
- The TEC values of Ga_2O_3 at room temperature are $\alpha_a = 2.84\times10^{-6}\ K^{1}$, $\alpha_b = 3.88\times10^{-6}\ K^{-1}$, $\alpha_c = 4.24\times10^{-6}\ K^{-1}$ and $\alpha_v = 10.96\times10^{-6}\ K^{-1}$ having expansion anisotropy that follow order $\alpha_b > \alpha_c > \alpha_a$.

References

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Acknowledgment

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