

SUPPLEMENTARY INFORMATION

Realization of 4-Inch and Thick β -Ga₂O₃ Single Crystals Using the Vertical Bridgman Method

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Simulation Model

The simulation considered factors including: heat transfer, phase change, convection in the melt and gas. The model used the finite volume method to numerically solve the governing equations describing heat and mass transfer. Natural convection in the melt was approximated using the Boussinesq assumption. Air was set as an incompressible ideal gas, and the Reynolds-Average Navier-Stokes (RANS) model was employed. A solidification-melting model was used to simulate the development of the solid-liquid interface during growth.

The governing equations solved are as follows:

General transport equation::

$$\frac{\partial(\rho\phi)}{\partial t} + \text{div}(\rho u\phi) = \text{div}(\Gamma \text{grad}\phi) + S \quad (1)$$

Where: ϕ is the general variable (can represent velocity, temperature, etc.); Γ is the generalized diffusion coefficient; S is the generalized source term; ρ is fluid density; t is time; u is velocity vector.

Mass conservation:

$$\nabla \cdot (\rho u) = 0 \quad (2)$$

Momentum equations in three dimensions:

$$\frac{\partial(\rho u)}{\partial t} + \text{div}(\rho u \vec{u}) = \text{div}(\mu \text{grad} u) - \frac{\partial p}{\partial x} + S_u \quad (4)$$

$$\frac{\partial(\rho v)}{\partial t} + \text{div}(\rho v \vec{u}) = \text{div}(\mu \text{grad} v) - \frac{\partial p}{\partial y} + S_v \quad (5)$$

$$\frac{\partial(\rho w)}{\partial t} + \text{div}(\rho w \vec{u}) = \text{div}(\mu \text{grad} w) - \frac{\partial p}{\partial z} + S_w \quad (6)$$

Where: $\text{grad}() = \partial()/\partial x + \partial()/\partial y + \partial()/\partial z$ is the fluid velocity vector;
 S_u , S_v , S_w are components of the generalized source term.

Energy equation:

$$\frac{\partial(\rho T)}{\partial t} + \text{div}(\rho \vec{u} T) = \text{div}\left(\frac{k}{c_p} \text{grad} T\right) + S_T \quad (7)$$

Where: T is temperature; k is thermal conductivity of the fluid; S_T is viscous dissipation term.

Radiation transfer equation:

$$\frac{dI(\vec{r}, \vec{s})}{ds} + (\alpha + \sigma_s)I(\vec{r}, \vec{s}) = \alpha n^2 \frac{\sigma T^4}{\pi} + \frac{\sigma_s}{4\pi} \int_0^{4\pi} I(\vec{r}, \vec{s}') \Phi(\vec{s}, \vec{s}') d\Omega' \quad (8)$$

Where:

\vec{r} - position vector;

\vec{s} - direction vector;

s - ray path length;

α - absorption coefficient;

σ_s - scattering coefficient;

I - radiation intensity, W/(m²·sr);

n -refractive index;

σ - Stefan-Boltzmann constant (5.67×10⁻⁸ W/m²·K⁴);

T - local temperature of object, K;

Φ - phase function;

\vec{s}' - scattering direction vector;

Ω' - solid angle.

In this study, the DO (Discrete Ordinates) radiation model was employed.

Solidification-Melting Model

The enthalpy method was used to model the solidification/melting process. The enthalpy of the material is calculated as follows:

$$H = h + \Delta H \quad (9)$$

$$h = h_{ref} + \int_{T_{ref}}^T C_p dT \quad (10)$$

Where h is sensible enthalpy, ΔH is latent heat, h_{ref} is reference enthalpy, T_{ref} is reference temperature, and C_p is specific heat at constant pressure.

The liquid fraction is defined as:

$$\beta = \begin{cases} \beta = 0 & \text{if } T < T_{solidus} \\ \frac{T - T_{solidus}}{T_{liquidus} - T_{solidus}} & \text{if } T_{solidus} < T < T_{liquidus} \\ \beta = 1 & \text{if } T > T_{liquidus} \end{cases} \quad (11)$$

The latent heat content can now be expressed in terms of latent heat to represent the heat of the material:

$$\Delta H = \beta L \quad (12)$$

The value of latent heat varies from 0 (for solids) to L (for liquids). For the problems of solidification and melting, the energy equation is:

$$\frac{\partial}{\partial t}(\rho H) + \nabla \cdot (\rho \vec{v} H) = \nabla \cdot (k \nabla T) + S \quad (13)$$

Where: H represents enthalpy, ρ represents density, v is the fluid velocity, and S is the source term.

The thermal insulation system in our model comprises two distinct materials (A and B), with their key properties listed shown in table S1.

Table S1 Characteristics and properties of insulation materials

Property	Symbol	Thermal insulation material a	Thermal insulation material b
Density	ρ (kg/m ³)	700	4400
Specific heat	C_p (J/kg*K)	1000	120
Thermal conductivity	λ (W/m*K)	0.39	0.9

Scanning Electron Microscope

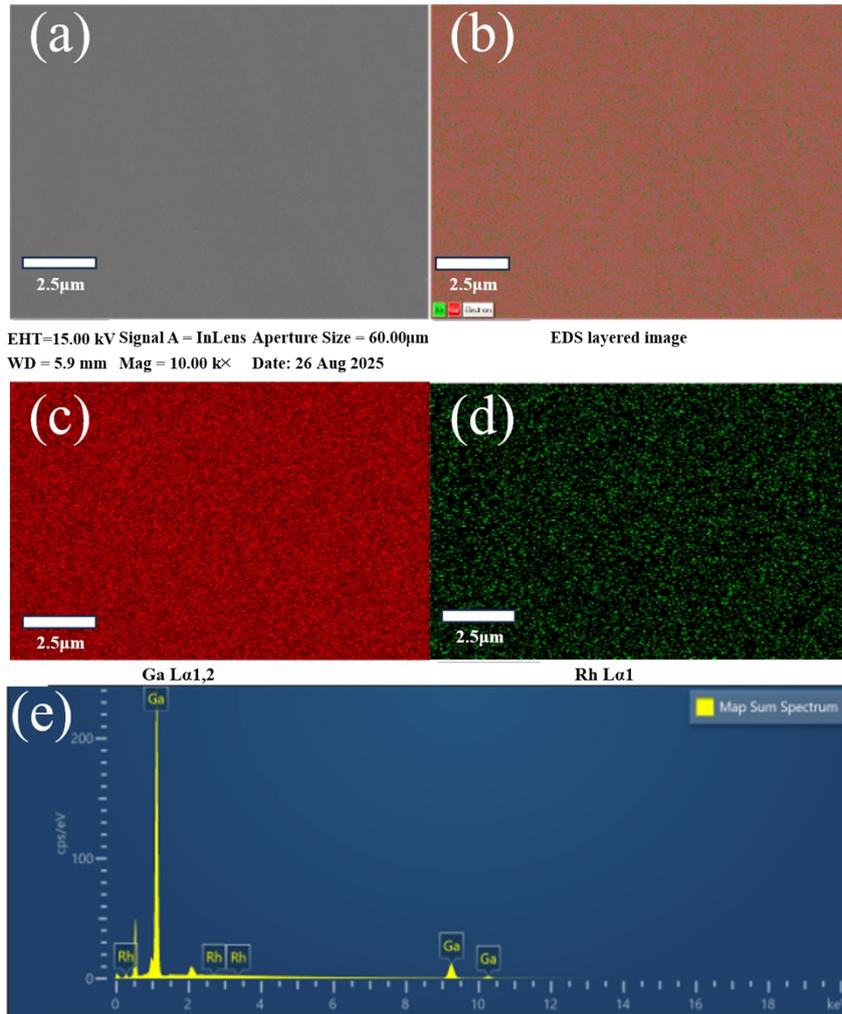


Figure S1. SEM characterization and elemental composition of β -Ga₂O₃ crystal (a) SEM micrograph of the polished β -Ga₂O₃ crystal surface. (b) Composite EDS elemental map (Ga, Rh) of the region shown in (a). (c) Ga elemental distribution map. (d) Rh elemental distribution map. (e) Map sum spectrum acquired over the entire scanned region.

Atomic Force Microscopy (AFM) Analysis

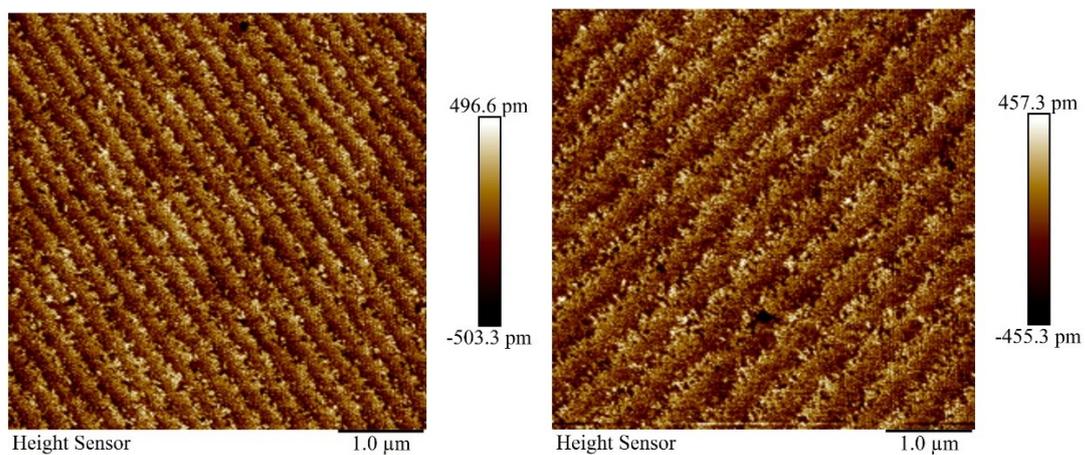


Figure S2. AFM height image of (100) β -Ga₂O₃ wafer.

Analysis of the (100)-oriented β -Ga₂O₃ wafer surface was performed in tapping mode under ambient conditions using a Bruker Dimension Icon system equipped with an RTESP-300 probe. As shown in **Fig. S2**, the $5 \times 5 \mu\text{m}^2$ height image reveals a well-defined step-terrace structure. No pits, cracks, or Rh-rich precipitates are observed, corroborating the high crystalline integrity inferred from HRXRD.