

Full Length Article

In-situ spectral reflectance investigation of hetero-epitaxially grown β -Ga₂O₃ thin films on c-plane Al₂O₃ via MOVPE process

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ABSTRACT

Metalorganic vapor-phase epitaxy of β -Ga₂O₃/c-plane Al₂O₃ heterostructures was monitored *in-situ* by spectral reflectance in different wavelengths. The reflectance spectrum was analysed as a function of the growth time and the incident wavelength to estimate the growth rate and the refractive index at the growth temperatures. The obtained values are validated by *ex-situ* methods such as secondary ion mass spectrum measurement and spectroscopic ellipsometry. A theoretical simulation of the reflectance spectrum was carried out by combining a transfer matrix method with a multilayer model, and a good agreement with the experimental results is presented.

1. Introduction

In recent years, there has been an increasing focus on β -Ga₂O₃ for different heterostructure electronic device applications, such as high breakdown voltage devices [1–3], Schottky diodes [4], photodetectors [5–8], and gas sensing devices [9,10]. The increasing interest in β -Ga₂O₃ is mainly due to its excellent properties with an ultrawide bandgap of about 4.8 eV [11] and its consequent high electric breakdown field of up to 8 MV/cm [12]. Many techniques have been investigated for the heteroepitaxy process of β -Ga₂O₃ films to fulfill the requirements of device fabrication on different substrates, especially on c-plane Al₂O₃, including pulsed laser deposition (PLD) [8,13], molecular beam epitaxy (MBE) [14,15], halide vapor phase epitaxy (HVPE) [16,17], and metalorganic vapor phase epitaxy (MOVPE) [18–24].

Among the techniques mentioned above, the MOVPE technique offers many advantages for device fabrication, including homogeneous deposition over device topography and the capability for large-scale production. The growth evolution of heteroepitaxial β -Ga₂O₃ thin films using MOVPE is believed to be linked to structural imperfections and the functional properties of the final layers. Traditional characterization methods such as transmission electron microscopy (TEM), scanning electron microscopy (SEM), and secondary ion mass

spectroscopy (SIMS) have been used to evaluate the structural qualities, interfacial properties, and thickness of β -Ga₂O₃ films grown by MOVPE. However, these methods are time-consuming, destructive, and *ex-situ*, making them impractical for production purposes.

Therefore, there is a high demand for a non-destructive, *in-situ* monitoring tool. Reflectance-based methods, such as reflection high-energy electron diffraction (RHEED) [25,26], have been widely used in various deposition techniques for *in-situ* monitoring. However, RHEED is typically limited to specific growth environments, such as low temperatures or vacuum conditions [27], making it suitable only for certain tools like molecular beam epitaxy (MBE) and pulsed laser deposition (PLD) but not for MOVPE. To our present knowledge, there have been no *in-situ* observations of the heteroepitaxial β -Ga₂O₃/c-plane Al₂O₃ growth process in the MOVPE system due to the lack of available refractive index at high temperatures. To fill this gap, we demonstrated using reflectance spectroscopy (RS) to *in-situ* monitor the thin film growth rate and the refractive index at high temperatures with validation by high-temperature ellipsometry measurement. Simulation methods like effective medium approximation (EMA) and transfer matrix method (TMM) are used to interpret observed reflected signals and correlate them with film properties and spectroscopic ellipsometry (SE) measurements [28,29]. This addresses limitations in commercial

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software, which may lack specific material parameters and flexibility for simultaneous estimation of growth rate and surface roughness based on user-defined layer structures. The results obtained in this work are an important step towards the high-quality β -Ga₂O₃-based optoelectronic devices and the growth process control of β -Ga₂O₃-related alloys, β -(Al_xGa_{1-x})₂O₃/ β -Ga₂O₃ [30–32].

2. Methods

2.1. Experimental setup

The MOVPE system was utilized to grow Si-doped β -Ga₂O₃ thin films consisting of a vertical shower-head low-pressure reactor (Structured Materials Industries, Inc. -USA). Triethylgallium (TEGa) was used as the metalorganic precursor for Ga, and Tetraethylorthosilicate (TEOS) was used as the metalorganic precursor for the n-type doping by Si, while O₂ (5 N) was used as the oxidant, and high purity Ar (5 N) acts as the carrier gas. Commercial c-plane Al₂O₃ of dimensions size $10 \times 10 \times 0.5 \text{ mm}^3$ was prepared by CrysTec GmbH. β -Ga₂O₃ thin film growth was performed at a constant growth temperature of 800 °C to ensure the formation of β -phase, and the chamber pressure was kept at 25 mbar. The Ar flow rate through the chamber was constant at $235 \times 10^{-3} \text{ mol/min}$. Thin films were grown using TEGa molar flow rates of range between $1.9 \times 10^{-5} \text{ mol/min}$ and $5.5 \times 10^{-5} \text{ mol/min}$, and the oxygen molar flow rates varied between $7 \times 10^{-3} \text{ mol/min}$ and $2.2 \times 10^{-2} \text{ mol/min}$. The TEOS molar flow rates range from $4 \times 10^{-11} \text{ mol/min}$ to $3 \times 10^{-8} \text{ mol/min}$. During the thin film deposition, *in-situ* reflectance monitoring was performed by using a Laytec EpiNet system (EpiNet 2017, Laytec. -Germany), acquiring reflectance signals at 405 nm, 633 nm, and 950 nm with the LED light sources through a window normal to the substrate embedded on the top of the MOVPE chamber [33]. The light source module is switched on and off several times per revolution (200 Hz modulation). One revolution is sufficient to acquire all measurement data of the samples that sit within the same radius of the susceptor. The thin-film thickness and the Si concentration were determined by the secondary ion mass spectrometry (SIMS) performed by RTG Mikroanalyse GmbH Berlin. The films were structurally characterized by high-resolution x-ray diffraction (HR-XRD) using a Bruker D8 instrument behind a pre-collimating parabolic Göbel mirror, and an asymmetric Ge 220 double crystal monochromator was utilized to select the Cu K α 1 line at $\lambda = 1.54056 \text{ \AA}$ and to collimate the incident x-ray beam to about 36 arcsec. The surface morphology of the grown β -Ga₂O₃ films was investigated by atomic force microscopy (AFM) (Bruker Dimension Icon, USA). The room-temperature and temperature-dependent optical properties of the β -Ga₂O₃ thin film were determined by spectroscopic ellipsometry (SE) on a Woollam M-2000DI rotating compensator ellipsometer in the spectral range of 0.73 to 6.5 eV at an angle of incidence equal to 60°. Temperature-dependent SE measurements were carried out from room temperature (RT) and finally ramped up to 800 °C, and it should be noted that the stability of the sample was checked by comparing the spectra measured before and after the SE measurement, showing that the sample did not deteriorate during the procedure.

2.2. A multilayer stack simulation

The Effective Medium Approximation (EMA) is a widely utilized method for modeling the optical characteristics and ellipsometry parameters of thin films. It employs the effective dielectric function to estimate the surface roughness and employs two parameters to describe the roughness: the volume fraction and the thickness of the rough surface layer. In the EMA approach, the surface roughness layer is typically assumed to consist of a combination of bulk material and air. The void volume is assumed to remain constant at 50 %, while the thickness of the surface roughness layer is adjusted as an unknown parameter through the fitting.

Optical constants like the dielectric function ϵ and the complex

refractive index N are needed to perform the EMA simulation. The dielectric function can be expressed as a function of the complex refractive index as $\epsilon = N^2$ following the Maxwell equations, the complex refractive index N is defined as $N \equiv n + ik$ with n as the refractive index and k as the extinction coefficient. The surface roughness layer is generally assumed to be homogeneous with an effective dielectric function, ϵ_{eff} . The effective dielectric function of the surface roughness layer can be calculated based on a simplified microscopic model, with the polarization of components represented by point dipoles [34] in the following form (Eq. (1)):

$$f \frac{\epsilon_n - \epsilon_{eff}}{\epsilon_n + \epsilon_{eff}} + (1-f) \frac{\epsilon_v - \epsilon_{eff}}{\epsilon_v + \epsilon_{eff}} = 0 \quad (1)$$

where f and ϵ_n represent the volume fraction and dielectric function of the measured sample, respectively; ϵ_v is the dielectric function of void spaces.

With the suggested optical structure from the EMA, the Transfer Matrix Method (TMM) is widely used as a mathematical approach to investigating wave reflection and transmission in one-dimensional structures, mostly on thin film. Considering the case of the hetero-epitaxy process, a simplified four-layer structure, as shown in Figure SI 1, is proposed to represent the grown film. n_1 , n_2 and n_3 are the real refractive index of the vacuum, the grown film (β -Ga₂O₃), and the substrate (c-plane Al₂O₃), respectively, while n_{eff} is the refractive index of the effective layer, d is the thickness of the β -Ga₂O₃ epilayer, and d_{eff} is the thickness of the effective layer. The light propagates in each layer according to the plane of incidence under an incident angle θ with respect to the reflection plane. Following Ref. [35], by projecting the electromagnetic wave vectors on the reflection plane with the boundary conditions at the interfaces, the Fresnel reflection coefficients r_s and r_p from layer 1 to layer 2 can be obtained as Eq. (2) and Eq. (3):

$$r_s = \frac{n_1 \cos \theta_1 - n_2 \cos \theta_2}{n_1 \cos \theta_1 + n_2 \cos \theta_2} \quad (2)$$

$$r_p = \frac{n_2 \cos \theta_1 - n_1 \cos \theta_2}{n_2 \cos \theta_1 + n_1 \cos \theta_2} \quad (3)$$

The Fresnel reflection coefficients involving more layers and interfaces can be determined with the same formulation. Together with the TMM, the total reflectivity of the film can be, therefore, expressed in the recurrence matrix form between the layers depicted in Figure SI 1, and a detailed deduction to calculate the total reflectivity can be found in Ref. [35,36].

3. Results and discussion

At optimal growth conditions, β -Ga₂O₃ grows with the (201) orientation parallel to (0001) Al₂O₃ plane [37,38]. Fig. 1 illustrates the 2 θ - ω HR-XRD scan of the β -Ga₂O₃ film on c-plane Al₂O₃. In the 2 θ range from 10° to 80°, only (201), (401) and (601) Bragg peaks of the monoclinic gallium oxide phase can be detected. The Inset of Fig. 1 shows the XRD rocking curve of the (201) diffraction peak of the β -Ga₂O₃ thin film. The full width at half maximum (FWHM) of the rocking curve is measured as 1.30°, indicating no significant dislocation densities. As a comparison, the reported FWHMs of β -Ga₂O₃ thin films grown on c-plane sapphire substrates are 1.5° (MOCVD, (201) diffraction peak) [37] and 1.49° (LPCVD, (201) diffraction peak) [39]. Fig. 2 shows *in-situ* collected reflectance spectra with wavelengths of 405 nm, 633 nm, and 950 nm from a $\sim 1000 \text{ nm}$ heteroepitaxial grown β -Ga₂O₃ film on c-plane Al₂O₃. Fabry-Perot (FP) oscillations appear in reflectance transients as the film growth starts due to differences in the refractive index of c-plane Al₂O₃ ($n_{\text{Al}_2\text{O}_3} = 1.77$ as the nominal value [40] at room temperature) and β -Ga₂O₃ ($n_{\text{Ga}_2\text{O}_3} = 1.95$ as the nominal value [41] at room temperature). Amplitudes of interference oscillation after a certain growth time are gradually getting smaller. Finally, no oscillation can be observed when

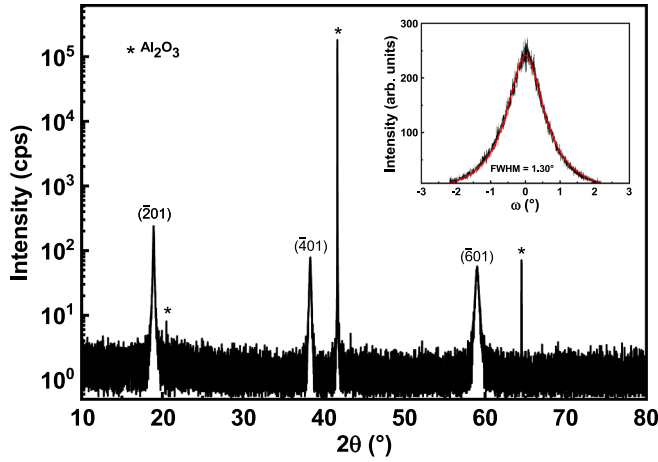


Fig. 1. 2θ-ω scans for the β-Ga₂O₃ thin film grown on the on-axis c-plane sapphire. Inset XRD rocking curve of (201) reflection of β-Ga₂O₃ thin film.

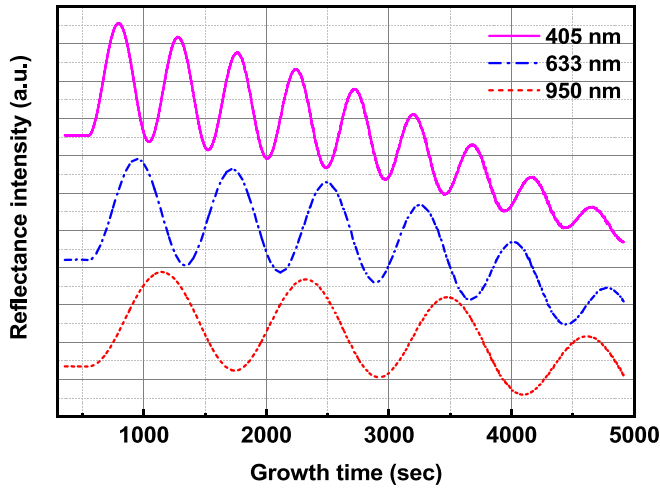


Fig. 2. In-situ reflectance transients recorded during the film growth at different wavelengths of 405 nm, 633 nm, and 950 nm.

the film is over a certain thickness, which is attributed to the increasing light scattering from the roughening surface (see Fig. 3) due to three-dimensional (3D) island growth caused by large lattice mismatch ($\sim 10.60\%$) between Al₂O₃ and β-Ga₂O₃ [42].

Besides, it can also be observed from Fig. 2 that the reflectance reduction is more pronounced at a smaller wavelength, which can be described qualitatively as [43] (Eq. (4)):

$$R(t) R_0 e^{-(4\pi\sigma/\lambda)^2} \quad (4)$$

where R_0 is the initial reflectance value, and σ is the root-mean-square (RMS) roughness of the sample surface. Following Eq. (4), when considering a decrease in surface roughness, a smaller incident wavelength exhibits a higher sensitivity by experiencing a relatively significant reduction compared to longer wavelengths. This phenomenon arises because the reflectance near the normal angle relies on both the surface roughness and the diffuse reflectance, with the latter being more prominent at smaller wavelengths and negligible at longer wavelengths [43].

From the peak-to-peak (or valley-to-valley) distance of the oscillation at the selected wavelength, $\tau(\lambda)$ (sec), the growth rate can be calculated by applying the FP equation for the interference oscillations (Eq. (5)):

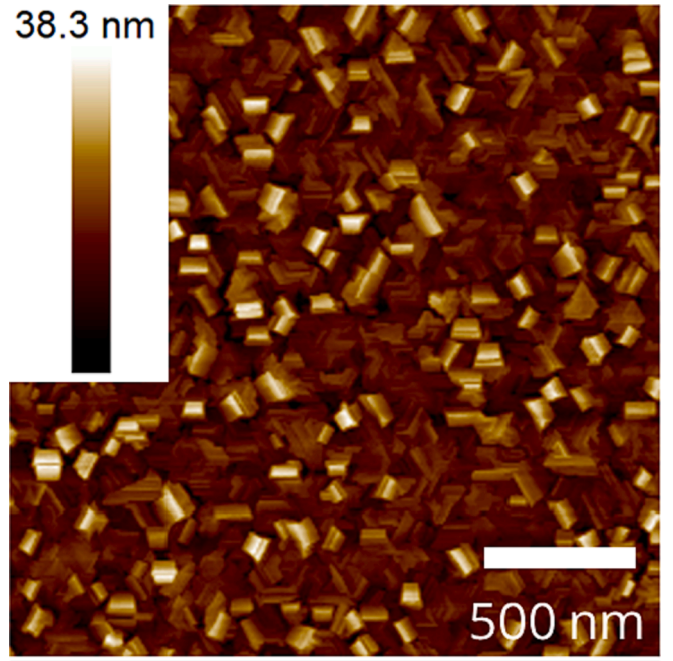


Fig. 3. AFM images of a β-Ga₂O₃ thin film grown on c-plane Al₂O₃ with a thickness of around 150 nm.

$$\text{Growth rate (nm/sec)} = \frac{\lambda}{2n(\lambda, T)\tau(\lambda)} \quad (5)$$

where λ is the wavelength of the incident light (nm), $n(\lambda, T)$ is the refractive index of the grown film at the incident wavelength, λ , and at growth temperature, T . To estimate the growth rate from FP oscillation, one should consider the thermo-optic effect while determining the refractive index since the reflectance transient is recorded at 800 °C. However, there was no measured value of the refractive index for (201) β-Ga₂O₃ at high temperatures up to 800 °C. Therefore, to determine the refractive index of the β-Ga₂O₃ film at high temperature (800 °C), temperature-dependent SE measurements were performed from room temperature up to 800 °C. To evaluate the refractive index and bandgap energy, we used a model to fit the SE parameters, Ψ and Δ , which consists of a four-component stack composed of substrate/film/ surface roughness/air, as shown in Figure SI 1. The optical properties of the surface roughness layer are analyzed by a Bruggeman effective medium approximation [44] consisting of a 50 % bulk film/50 % void mixture.

The experimentally measured ellipsometry parameters, ψ and Δ , which are related to the optical and structural properties of the sample, are defined as (Eq. (6)):

$$\rho = \frac{R_p}{R_s} = \tan(\Psi)e^{i\Delta} \quad (6)$$

where R_p and R_s are the complex reflection coefficients of the light-polarized parallel (p) and perpendicular (s) to the plane of incidence, respectively. While β-Ga₂O₃ films heteroepitaxial grown on c-plane Al₂O₃ substrates contain threefold in-plane rotational domains[45], SE data were analyzed by assuming an isotropic material.

Since the films under investigation are optically transparent in the 0.73 – 4.3 eV spectral range, here the dispersion of the refractive index can be evaluated by the Sellmeier dispersion equation (first approximation, Eq. (7)):

$$n^2(\lambda, T) = 1 + \frac{B\lambda^2}{\lambda^2 - \lambda_0^2} \quad (7)$$

where B is a fitting parameter proportional to the density of effective

electron states, and λ_0 is the wavelength of the Sellmeier oscillator. The values of B and λ_0 at room temperature and 800 °C are listed in Table 1. The extinction coefficient k is considered to be zero since the absorption of the β -Ga₂O₃ film in the visible region can be neglected [46]. Based on the Sellmeier dispersion relation, we determined the refractive index of β -Ga₂O₃ film in the transparent region (Fig. 4). It is obvious that the refractive index continuously increases with decreasing the wavelength. Nearly the same dispersion behavior is observed for β -Ga₂O₃ films and bulk [45,46].

The evaluated n at 405 nm, 633 nm, and 905 nm of β -Ga₂O₃ film as a function of temperature is presented in Fig. 5. The refractive index at the three wavelengths reveals an increase with increasing temperature, and this behavior is attributed to electron-phonon interaction that is enhanced with increasing temperature [47].

Refractive indices for all three wavelengths at RT and 800 °C have been tabulated in Table 2. The value of n , especially at 633 nm (~ 1.93), is larger than the one reported by other authors [41,48]. As the higher n indicates more dense films without pores, this means that the MOVPE technique results in denser Ga₂O₃ layers. This can also explain the higher dielectric constant as compared to films deposited by other techniques.

It has been reported by Bhaumik [41] et al. that β -Ga₂O₃ (in (100) and (010) planes) has a positive thermo-optic coefficient around $10^{-5}/\text{K}$ in the temperature range of 30 °C to 175 °C. Fig. 5 shows the measured refractive indices as a function of temperature from room temperature to 800 °C at different wavelengths, and a slope of $\sim 10^{-5}/\text{K}$ can be observed, matching the reported values in the literature [41,48]. Table 3 summarizes the thermo-optic coefficients at different wavelengths from Fig. 5.

In Fig. 6a, the heteroepitaxial growth of a 300 nm Si-doped β -Ga₂O₃ film is monitored at different wavelengths. Using the refractive index at 800 °C and 405 nm from Table 2, the reflectivity response at 405 nm of the heteroepitaxial structure (Fig. 6a) has been fitted by a theoretical simulation model essentially based on the TMM simulation package in Python [36] and the four-layer stack structure (Figure SI 1).

In the following simulation, n_1 , n_2 and n_3 are the real refractive index of the air, the film (β -Ga₂O₃), and the substrate (c-plane Al₂O₃), respectively. We have used the obtained refractive indices of β -Ga₂O₃ (Table 2) and the refractive indices of c-plane Al₂O₃ (at 800 °C) [40,49]. The refractive index of the effective layer, n_{eff} , is calculated by fitting the reflectance spectrum at 405 nm. The simulation results given by the model are presented by the green line in Fig. 7. The two parameters which characterize the effective layer are the effective refractive index, n_{eff} , and the effective thickness, d_{eff} , respectively [50]. d_{eff} is obtained from the best fitting of the experimentally collected reflectance signals as a function of time from Fig. 7.

The simulation result of the *in-situ* reflectance gives the β -Ga₂O₃ layer thickness of ~ 300 nm, and an effective layer thickness of about 25 nm can be determined, matching the cross-sectioned scanning electron microscopy (SEM) image and the SIMS profile of the film shown in Fig. 6b and Fig. 6c, respectively. On the other hand, ellipsometry is an optical method that accurately determines the optical constants of surfaces or the thickness and optical constants of superficial films on solid substrates [51,52]. Therefore, on the same sample, SE measurements have been performed, and the best-fitting adjustment gives a layer thickness of 299 nm and an effective thickness surface roughness of about 12 nm. The film thickness is in good agreement with the film thickness determined by SIMS and the *in-situ* reflectance. However, the noticeable

Table 1

A summary of the Sellmeier coefficients equation (Eq. (7)) at room temperature and 800 °C.

Coefficient	Room temp.	800 °C
B	2.59 ± 0.02	2.63 ± 0.02
λ_0 (nm)	136 ± 3	149 ± 3

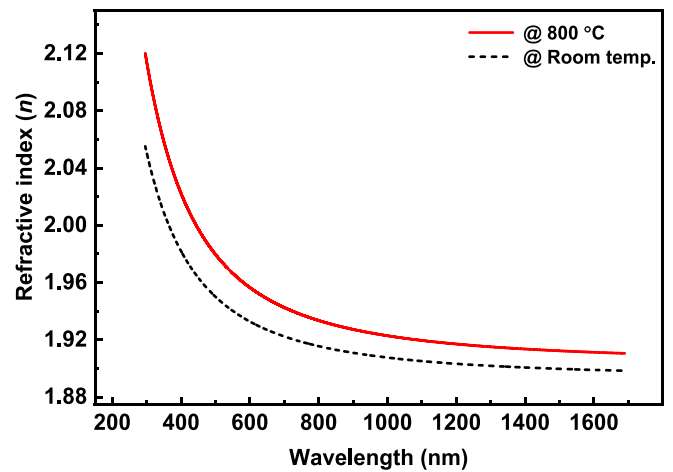


Fig. 4. The refractive index obtained by using a Sellmeier dispersion relation from experimentally measured Ψ and Δ spectra of β -Ga₂O₃ on c-plane Al₂O₃ at room temperature and 800 °C.

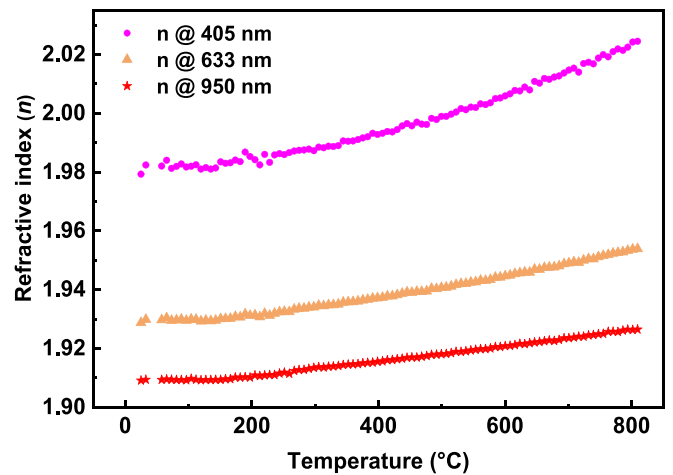


Fig. 5. The temperature-dependent refractive index of β -Ga₂O₃ on c-plane Al₂O₃ ranges from RT to 800 °C at the wavelengths of 405 nm, 633 nm, and 950 nm.

Table 2

The refractive index of β -Ga₂O₃ on c-plane Al₂O₃ at room temperature and 800 °C.

Wavelength (nm)	Refractive index	
	Room temp.	800 °C
405	1.98 ± 0.01	2.02 ± 0.01
633	1.93 ± 0.01	1.95 ± 0.01
950	1.90 ± 0.01	1.93 ± 0.01

Table 3

The thermo-optic coefficient of β -Ga₂O₃ on c-plane Al₂O₃ at different wavelengths.

Wavelength (nm)	Thermo-optic coefficient (1/K)
405	5.8×10^{-5}
633	3.2×10^{-5}
950	2.2×10^{-5}

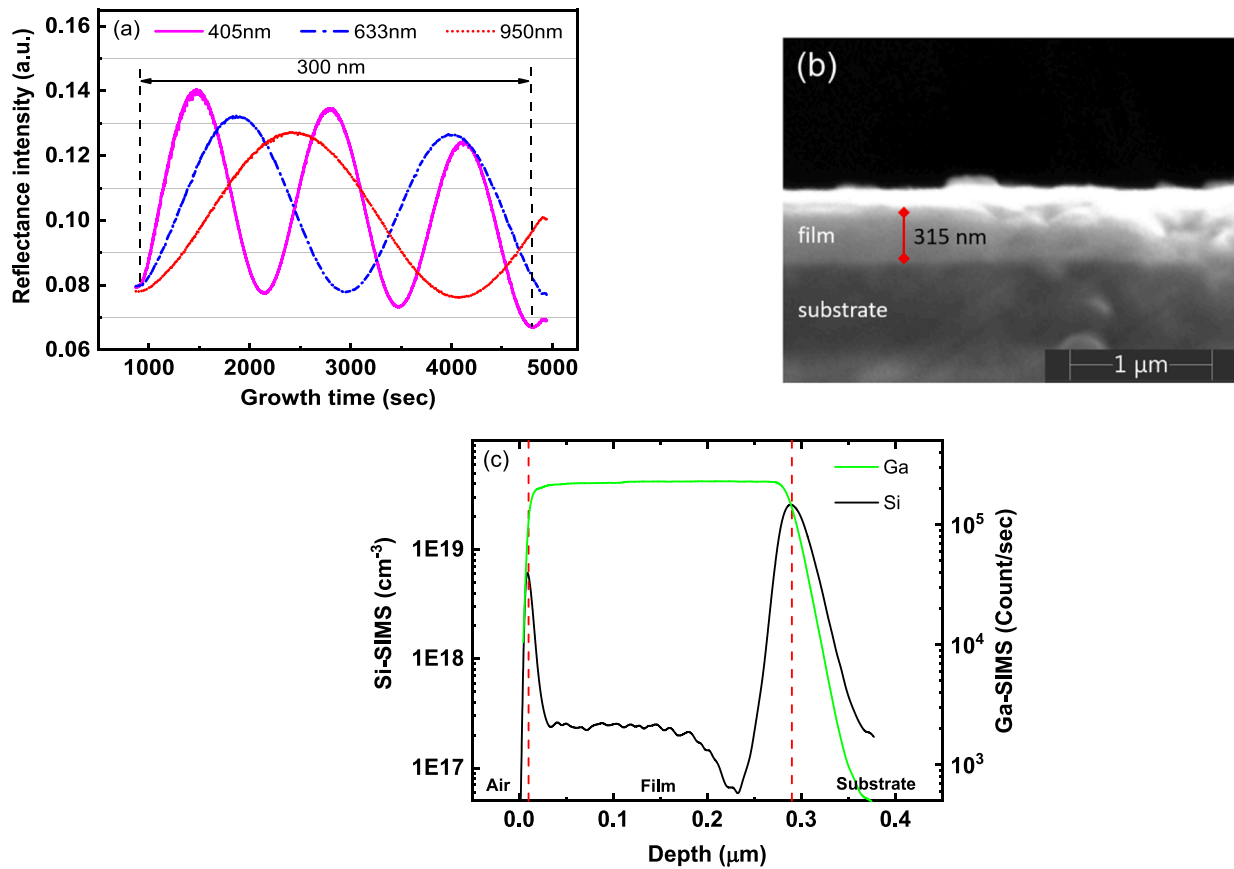


Fig. 6. (a) Reflectance transients (curves) recorded during sample growth at different wavelengths of 405 nm, 633 nm, and 950 nm, and the evolution of the spectra during the growth. (b) A SEM image (cross-section view) of the film from Fig. 6a. (c) The depth profile determined by the SIMS measurement via the intentionally doped Si concentration and the Ga intensity in the film from Fig. 6a.

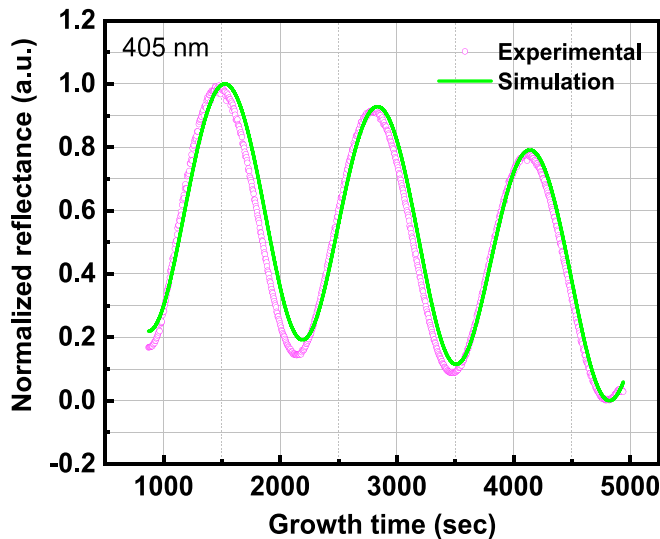


Fig. 7. The best fit of the normalized reflectance (from Fig. 6a) obtained from reflectance spectroscopy using the effective layer/ β -Ga₂O₃/c-plane Al₂O₃ substrate system at an incident wavelength of 405 nm.

difference in the effective layer thickness (25 nm from the reflectance spectrum and 12 nm from SE) can be attributed to variations in spot size between reflectance spectroscopy and spectroscopic ellipsometry and local roughness fluctuations on the film surface. Moreover, it is anticipated that the total film thickness may deviate by approximately 5 %

due to temperature-induced expansion (using a temperature expansion coefficient of approximately $\sim 1 \times 10^{-5} / \text{K}$ [42,53]). To check the validity and reproducibility of the model, several β -Ga₂O₃ films were grown on c-plane Al₂O₃ substrate at different deposition times. The film thickness values obtained from *in-situ* reflectance fitted by TMM and *ex-situ* SE measurements are comparable and presented in Fig. 8. A good match between the simulation results (triangles) and the experimental measurement (circles) is clearly evident.

4. Conclusions

This work investigated the heteroepitaxial growth of β -Ga₂O₃ thin film on c-plane Al₂O₃ using a multiwavelength (405 nm, 633 nm, and 950 nm) *in-situ* monitoring system as well as *ex-situ* spectroscopic ellipsometry. Room temperature and high-temperature (800 °C) refractive index of β -Ga₂O₃ on c-plane Al₂O₃ were determined. The thermo-optic coefficient from room temperature to 800 °C at different wavelengths was reported. The EMA model and the TMM simulation are combined to estimate the β -Ga₂O₃ thin film thickness and surface roughness using the measured high-temperature refractive index and were compared with the *ex-situ* spectroscopic ellipsometry values. A good agreement between the simulation approach and the experimental results is established.

CRediT authorship contribution statement

Ta-Shun Chou: Writing – original draft, Methodology, Investigation, Formal analysis, Data curation. **Saud Bin Anooz:** Writing – original draft, Methodology, Investigation, Formal analysis, Data curation. **Raimund Grüneberg:** Methodology, Data curation. **Jana Rehm:**

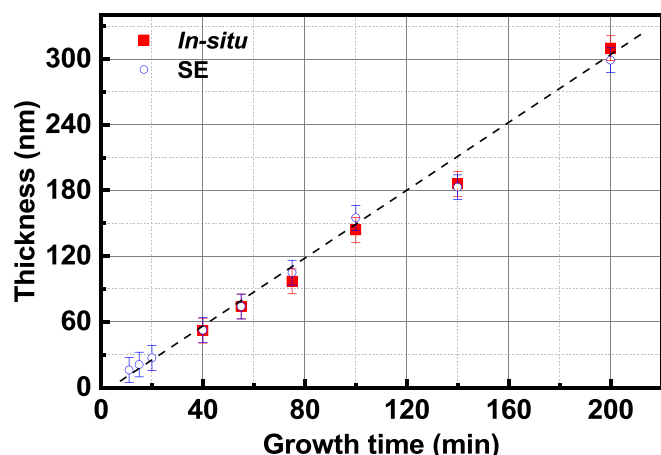


Fig. 8. A comparison between the thickness values obtained from measured values using SE (circles) and the fitted values derived from the in-situ reflectance spectrum (triangles).

Methodology, Data curation. **Arub Ahktar**: Data curation. **Deshabrato Mukherjee**: Methodology, Data curation. **Peter Petrik**: Data curation, Resources, Supervision, Writing – review & editing. **Andreas Popp**: Investigation, Project administration, Resources, Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Author contributions

Ta-Shun Chou and Saud Bin Anooz contributed equally to this work.

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