Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

SUPPLEMENTAL INFORMATION

1. *Flux calculations*

The Ga and In beam equivalent pressures are converted into particle fluxes Φ_{Me} ($cm^{-2}s^{-1}$) in Fig. 4b-d according to kinetic gas theory:[58]

$$\Phi_{Me}^{BEP} = C\frac{\rho_{Me}}{\sqrt{2\pi m_{Me}k_B T}}, \quad (9)$$

$$\Phi_{Me} = I_{Me}\Phi_{Me}^{BEP}, \quad (10)$$

Where m_{Me} is the atomic mass, T is the effusion cell temperature, $C = 0.013332$ $cm^{-2}s^{-1}$ is a prefactor to convert Φ_{Me}^{BEP} from torr to particle flux, and I_{Me} is an effective sensitivity factor determined from metal-limited calibration growth. The Ga sensitivity factor I_{Ga} was determined from homoepitaxial growth of Ga_2O_3 with a typical growth rate of 3.16 nm/min, and the In sensitivity factor I_{In} was estimated as $I_{In} = I_{Ga}(5.2/9.1)$ in accordance with the experimentally obtained factors reported in Ref. 60. The oxygen beam equivalent pressure $\Phi_{O}^{BEP} = 3.2\times10^{15}$ $cm^{-2}s^{-1}$ at 3.00 SCCM and 250 W was calculated from the growth rate of slightly metal-rich homoepitaxial Ga_2O_3 growth at 650 °C. The effective oxygen flux $\Phi_{O}^{*,Me}$ available for Ga_2O_3 and In_2O_3 growth varies for each metal species due to the differing oxidation efficiencies[34,54,58] and is given by:

$$\Phi_{O}^{*,Me} = J_{O}^{Me}\Phi_{O}^{BEP}. \quad (11)$$

In the presence of both Ga and In flux, the total effective oxygen flux Φ_{O}^{*} is modified by the metal flux ratio $R = \Phi_{In}/(\Phi_{Ga} + \Phi_{In})$, with[58]

$$\Phi_{O}^{*} = \Phi_{O}^{BEP}\left(RJ_{O}^{In} + (1-R)J_{O}^{Ga}\right). \quad (12)$$

The total metal/oxygen flux ratio is thus $(\Phi_{Ga} + \Phi_{In})/\Phi_{O}^{*}$ and the stoichiometric flux ratio shown in Fig. 4d is $\frac{3}{2}(\Phi_{Ga} + \Phi_{In})/\Phi_{O}^{*}$. The flux calibration parameters are summarized in Table I.

TABLE I. Flux calibration parameters for Ga and In.

Metal	Ga	In
Atomic mass, m_{Me} (amu)	69.723	114.818
Sensitivity factor, I_{Me}	10.60	6.06
Oxidation efficiency, J_{O}^{Me}	0.096	0.263

2. *X-ray diffraction of phase separated growth*

Fig. 7 shows a coupled ω-2θ x-ray diffraction scan for a targeted growth performed at 740 °C with relatively high In and Ga fluxes of 3×10^{-7} torr each. The dashed lines indicate the locations of the β-Ga_2O_3 (020) reflection at ω = 30.481° and the bixbyite In_2O_3 (444) reflection at ω = 31.824°. The presence of the measured bixbyite In_2O_3 (444) peak reflects the terminal RHEED pattern which developed into a spotty/faceted pattern typical of Fig. 3b, confirming that this pattern is due to the formation of In_2O_3 on the surface. No compressive features are observed at diffraction angles below the β-Ga_2O_3 (020) peak, confirming that In does not incorporate at these high metal flux growth conditions. RBS measurements for this sample (not shown) confirm the presence of a thin In-rich layer at the surface. Reaction equations (4) through (8) suggest that In_2O_3 formation is suppressed at these metal rich conditions due to a combination of In_2O formation and Ga cation exchange followed by Ga_2O decomposition. The presence of a thin In_2O_3 layer suggests that complete decomposition of In_2O_3 does not occur. The accumulation of In_2O_3 is very small, however, on the order of several nanometers over the course of a 1 hour growth.

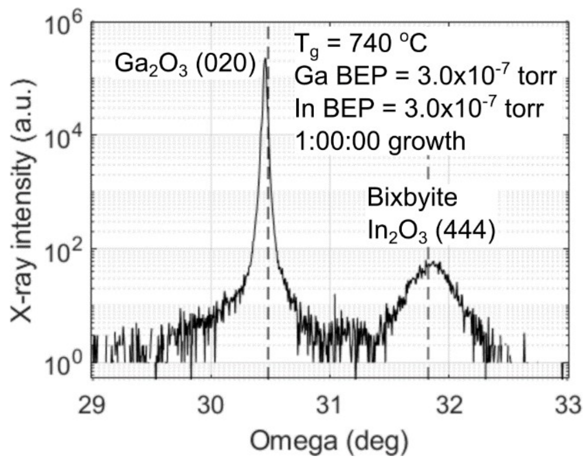

FIG. 7. Coupled ω-2θ x-ray diffraction (XRD) scan of targeted growth with spotty/faceted surface reconstruction. Growth conditions labeled directly on figure. The peak at ω = 31.824° is attributed to the (444) reflection from bixbyite In_2O_3, responsible for the spotty/faceted RHEED pattern.

3. *Orientation of bixbyite In_2O_3 on (010) β-Ga_2O_3*

Fig. 8 shows the possible orientation of bixbyite In_2O_3 on (010) oriented β-Ga_2O_3. The In_2O_3 [111] direction is parallel to the Ga_2O_3 [010] direction and is perpendicular to the growth plane of the substrate. The In_2O_3 [011] direction is perpendicular to the Ga_2O_3 [001] direction. This orientation is responsible for the bixbyite In_2O_3 (444) peak observed in the coupled ω-2θ XRD scan shown in Fig. 7 for the phase separated growth, and for the ~7.1 Å spot spacing observed in the RHEED pattern shown in Fig. 3b corresponding to diffraction from the In_2O_3 (110) plane.

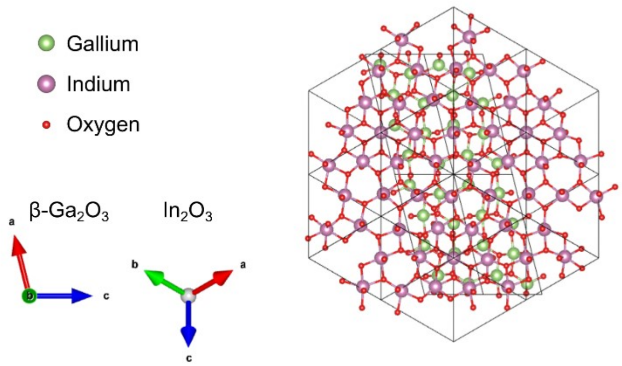

FIG. 8. Possible orientation of bixbyite In_2O_3 on (010) oriented β-Ga_2O_3.