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Kinetic modeling of N incorporation in GaInNAs growth by plasma-assisted molecular-beam epitaxy
Kinetics versus thermodynamics of the metal incorporation in molecular beam epitaxy of (In$_x$Ga$_{1-x}$)$_2$O$_3$

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We present a detailed study of the reaction kinetics and thermodynamics of the plasma-assisted oxide molecular beam epitaxy of the ternary compound (In$_x$Ga$_{1-x}$)$_2$O$_3$ for $0 \leq x \leq 1$. We measured the growth rate of the alloy in situ by laser reflectometry as a function of growth temperature $T_G$ for different metal-to-oxygen flux ratios $r_{Me}$ and nominal In concentrations $x_{nom}$ in the metal flux. We determined ex situ the In and Ga concentrations in the grown film by energy dispersive X-ray spectroscopy. The measured In concentration $x$ shows a strong dependence on the growth parameters $T_G$, $r_{Me}$, and $x_{nom}$ whereas growth on different co-loaded substrates shows that in the macroscopic regime of $\sim \mu m^3$ $x$ does neither depend on the detailed layer crystallinity nor on crystal orientation. The data unveil that, in the presence of In, Ga incorporation is kinetically limited by Ga$_2$O desorption the same way as during Ga$_2$O$_3$ growth. In contrast, In incorporation during ternary growth is thermodynamically suppressed by the presence of Ga due to stronger Ga–O bonds. Our experiments revealed that Ga adatoms decompose/etch the In–O bonds whereas In adatoms do not decompose/etch the Ga–O bonds. This result is supported by our thermochemical calculations. In addition we found that a low $T_G$ and/or excessively low $r_{Me}$ kinetically enables In incorporation into (In$_x$Ga$_{1-x}$)$_2$O$_3$. This study may help growing high-quality ternary compounds (In$_x$Ga$_{1-x}$)$_2$O$_3$ allowing band gap engineering over the range of 2.7–4.7 eV. © 2016 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/). [http://dx.doi.org/10.1063/1.4961513]

The transparent (semi)conducting oxide, indium sesquioxide In$_2$O$_3$, has been for decades a well studied material and reached industrial significance due to its good transparency in the visible regime of light and quite large band gap of $E_g = (2.7 \pm 0.1)$ eV.$^1$ In contrast, gallium sesquioxide Ga$_2$O$_3$ is a transparent semiconducting oxide and a rather new material. Due to its direct large band gap of $E_g = (4.7 \pm 0.2)$ eV,$^2$ it is transparent up to the deep ultra-violet (DUV) regime of light and a suitable material for DUV detectors and power devices with higher breakdown voltage and efficiency than SiC and GaN,$^2$ for instance. Alloying both materials into (In$_x$Ga$_{1-x}$)$_2$O$_3$ should enable to widen the wavelength range for DUV detectors and the growth of heterostructures.$^3$

The applications mentioned above and the control of the compound concentration require a high degree of purity and crystallinity, that can be achieved by plasma-assisted oxide molecular beam epitaxy (MBE).$^4$–$^9$

So far, theoretical reports have predicted the miscibility of In$_2$O$_3$ and Ga$_2$O$_3$.$^{10}$–$^{12}$ Experimental studies about the crystal structure and optical properties of (In$_x$Ga$_{1-x}$)$_2$O$_3$ grown by MBE,$^{13}$ pulsed laser deposition,$^{14,15}$ and metal-organic vapor phase epitaxy (MOVPE)$^{16,17}$ were carried out. For the latter method, it has been shown that no In was incorporated at too low O pressures in the growth chamber,$^{16}$ and that In likely has a tendency to float on the growth surface.$^{17}$ However, systematic

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investigations of the reaction kinetics and studies of the In and Ga incorporation for the (In\(_{x}\)Ga\(_{1-x}\))\(_2\)O\(_3\) growth are lacking.

For the binary oxides In\(_2\)O\(_3\) and Ga\(_2\)O\(_3\), we have already shown that the growth rate \(I'\) decreases in the Me-rich regime due to the oxygen-deficiency-induced formation of the volatile suboxide Me\(_2\)O (Me = In, Ga).\(^{4-6}\) At high growth temperatures \(T_G\) \(I'\) also decreases because of Me\(_2\)O desorption in the Me-rich and O-rich regimes. This decrease of \(I'\) is stronger with increasing \(T_G\) for a larger Me-to-O flux ratio \(r_{\text{Me},O} = \Phi_{\text{In, Ga}}/\Phi_O\) (see Fig. 1 and Refs. 5 and 6). The In, Ga, and active O fluxes are denoted as \(\Phi_{\text{In}}, \Phi_{\text{Ga}},\) and \(\Phi_O\), respectively.

In this letter, we present a systematic study of the In and Ga incorporation into the ternary oxide (In\(_x\)Ga\(_{1-x}\))\(_2\)O\(_3\) as a function of \(T_G\) for different Me-to-O flux ratios \(r_{\text{Me}} = (\Phi_{\text{In}} + \Phi_{\text{Ga}})/\Phi_O\) ranging from 0.1 to 2.0, and In-to-Ga flux ratios \(x_{\text{nom}} = \Phi_{\text{In}}/(\Phi_{\text{In}} + \Phi_{\text{Ga}})\) ranging from 0 to 1. The results are discussed in terms of kinetic and thermodynamic limitations. Growth conditions to force metal incorporation are demonstrated as well. However, the miscibility of In\(_2\)O\(_3\) and Ga\(_2\)O\(_3\) as well as the crystallinity of (In\(_x\)Ga\(_{1-x}\))\(_2\)O\(_3\) are beyond the scope of this study.

The samples were grown in a custom made MBE system equipped with a laser-reflectometry set-up (LR) that allows the in-situ measurement of \(I'\) and a line-of-sight quadrupole mass spectrometer (QMS) that also allows to monitor in-situ the actual desorbing flux from the substrate.\(^{4}\) Standard shuttered hot-lip effusion cells were used to evaporate liquid In and Ga (7N purity). The beam equivalent pressure (BEP) which is proportional to the particle flux was measured by a nude filament ion gauge positioned at the substrate location.

The Me fluxes were calibrated by measuring \(I'\) of the In\(_2\)O\(_3\) and Ga\(_2\)O\(_3\) layers in the O-rich regime at sufficiently low \(T_G\) where neither Me\(_2\)O desorption nor Me accumulation occurs.\(^{4-6}\) With this calibration we converted the measured BEPs into equivalent Me\(_2\)O growth rates that are used throughout this study.

A radio frequency plasma source with a mass flow controller supplied \(\Phi_O\) from the research-grade O\(_2\) gas (6N purity). The radio frequency power of the plasma source was maintained at 300 W. The O\(_2\) mass flows \(\Phi_{O_2}\) were set to 0.5, 1.0, and 2.0 standard cubic centimeter per minute (SCCM). Due to the different oxidation efficiencies of In\(^4\) and Ga\(^4\), \(\Phi_O\) was calculated for all values of \(x_{\text{nom}}\) as

\[
\Phi_O = \Phi_O^4(x_{\text{nom}} = 1) = 5.7 \text{ Å}/(s \times \text{SCCM}) \quad \text{and} \quad \Phi_O^{Ga}(x_{\text{nom}} = 0) = 1.7 \text{ Å}/(s \times \text{SCCM})
\]

with \(\Phi_O^{In} = \Phi_O(x_{\text{nom}} = 1) = 5.7 \text{ Å}/(s \times \text{SCCM})\) and \(\Phi_O^{Ga} = \Phi_O(x_{\text{nom}} = 0) = 1.7 \text{ Å}/(s \times \text{SCCM})\) determined from the growth at stoichiometric condition of the binary oxide In\(_2\)O\(_3\) and Ga\(_2\)O\(_3\), respectively.\(^{4}\) Table I summarizes the growth parameters employed throughout this study.
Growth parameters and corresponding figure symbols in Figs. 1–4 for the \( (\text{In}_x\text{Ga}_{1-x})_2\text{O}_3 \) growth.

<table>
<thead>
<tr>
<th>Figure symbols</th>
<th>( r_{\text{Me}} )</th>
<th>( x_{\text{nom}} )</th>
<th>( \Phi_{\text{In}} (\text{Å/s}) )</th>
<th>( \Phi_{\text{Ga}} (\text{Å/s}) )</th>
<th>( \Phi_{\text{O}} (\text{Å/s}) )</th>
</tr>
</thead>
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<td>Centered (inset in Fig.)</td>
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<td>0.50</td>
<td>0.50</td>
<td>0.50</td>
<td>10.80</td>
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<tr>
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<td>1.44</td>
<td>1.81</td>
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<tr>
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<td>1.12</td>
<td>1.60</td>
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<tr>
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<td>0.25</td>
<td>0.50</td>
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<td>2.65</td>
</tr>
<tr>
<td>Crossed</td>
<td>2.00</td>
<td>0.50</td>
<td>1.80</td>
<td>1.80</td>
<td>1.80</td>
</tr>
</tbody>
</table>

Using QMS, \( T_G \) was calibrated by monitoring the total reflection of a known \( \Phi_{\text{Ga}} \) desorbing from an \( \text{Al}_2\text{O}_3 \) (0001) surface. In order to obtain \( T_G \), we slowly decreased the thermo-couple temperature of the substrate heater \( T_{TC} \) until \( \text{Me} \) atoms started adsorbing on the substrate. Besides, \( T_G \) was calculated by applying kinetic theory of gases as reported in Ref. 18 that enabled determining the offset between \( T_{TC} \) and \( T_G (\Delta T = T_{TC} - T_G) \). Depending on \( T_G, r_{\text{Me}}, \) and \( x_{\text{nom}} \) the layers were either amorphous, polycrystalline, or crystalline. The \( \text{In} \) layer concentration \( x \) was obtained by measuring a macroscopic volume of the epilayer of \( \sim 4 \mu m^3 \) in a scanning electron microscope by means of energy dispersive X-ray spectroscopy (EDX).

In order to investigate whether \( x \) depends on the substrate material and orientation, different cleaved substrates of \( \text{Al}_2\text{O}_3 \) ((0001),(1120),(1012)) and Y-stabilized \( \text{ZrO}_2 \) (YSZ)((111),(100)) were \( \text{In} \)-bonded on a Si wafer and the \( (\text{In}_x\text{Ga}_{1-x})_2\text{O}_3 \) layers were grown simultaneously on different substrates. It turned out that at a scale of a macroscopic volume \( x \) neither depends on the substrate material and orientation, nor on the crystallinity of the film. The crystallinity and structure of the alloys were identified \textit{in-situ} by reflection high energy electron diffraction during growth and \textit{ex-situ} by X-ray diffraction \( \omega-2\theta \) wide-range scans (not shown).

Figure 1 shows the measurement of \( \Gamma \) of the binary grown \( \text{In}_2\text{O}_3 \) and \( \text{Ga}_2\text{O}_3 \) as a function of \( T_G \). The dependencies of \( \Gamma \) on \( T_G \) for both materials are discussed in detail in Refs. 5 and 6. The stronger decrease of \( \Gamma_{\text{Ga}_2\text{O}_3} \) with \( T_G \) and \( r_{\text{Ga}} = \Phi_{\text{Ga}}/\Phi_{\text{O}} \) is kinetically explained by the higher vapor pressure of its suboxide \( \text{Ga}_3\text{O}_2 \) as compared to the suboxide \( \text{In}_3\text{O}_2 \). Similarly, for the ternary film \( (\text{In}_x\text{Ga}_{1-x})_2\text{O}_3 \) (Fig. 2) we do not observe any decrease of \( \Gamma \) up to \( T_G = 800 \) °C.

![Figure 2: Growth rate of \((\text{In}_x\text{Ga}_{1-x})_2\text{O}_3\) as a function of growth temperature \( T_G \) plotted for different \( r_{\text{Me}} = 0.47, 0.72, 0.75, 2.00 \) and \( x_{\text{nom}} = 0.45, 0.70, 0.25, 0.50 \) for filled, open, dotted, and crossed squares, respectively (see Table I). Inset: \( \Gamma_{(\text{In}_x\text{Ga}_{1-x})_2\text{O}_3} \) as a function of \( T_G \) for \( r_{\text{Me}} = 0.1 \) and \( x_{\text{nom}} = 0.50 \) (centered squares). The lines are guides to the eye.](image-url)
In order to unveil the relative loss of In and Ga contributing to the decrease of $I_i(\text{In}_x\text{Ga}_{1-x})\text{O}_3$ with $T_G$, we investigated $x$ as a function of $T_G$ for the growth conditions indicated in Table I. The results are plotted in Fig. 3. Under O-rich growth conditions of $r_{\text{Me}} < 1.0$ and $T_G < 600 \, ^\circ\text{C}$, $x = x_{\text{nom}}$. For $T_G \leq 600 \, ^\circ\text{C}$, $x$ decreases with increasing $T_G$ for $0.1 < r_{\text{Me}} < 1.0$. However, for $r_{\text{Me}} \leq 0.1$, $x$ does not depend on $T_G$ up to 800 $^\circ\text{C}$ and is equal to $x_{\text{nom}}$ (inset, Fig. 3). Under Me-rich growth conditions, for $r_{\text{Me}} = 2.0$ and $\Phi_\text{O} = \Phi_\text{In} = \Phi_\text{Ga}$, $x = 0$ for all measured $T_G$, i.e., only Ga$_2$O$_3$ is formed. Scanning electron microscopy and EDX measurements revealed that In accumulated as droplets on the growth surface for this sample at $T_G < 600 \, ^\circ\text{C}$ (not shown). Similar metal accumulation has been observed during MBE under metal-rich growth conditions at these low $T_G$ for In$_2$O$_3$ but not for Ga$_2$O$_3$.\(^6\) This finding suggests preferential incorporation of Ga over In.

Now, we focus on the growth experiments of (In$_x$Ga$_{1-x}$)$_2$O$_3$ for which $x_{\text{nom}}$ was set to 0.5 and 0.45 and discuss the individual incorporation rates of In and Ga into (In$_x$Ga$_{1-x}$)$_2$O$_3$. In order to access these quantities we introduce pseudo-binary growth rates $\gamma$ defined as

$$\gamma_{\text{In}_2\text{O}_3} = xI_i(\text{In}_x\text{Ga}_{1-x})\text{O}_3$$ \hspace{1cm} (2)

and

$$\gamma_{\text{Ga}_2\text{O}_3} = (1 - x)I_i(\text{In}_x\text{Ga}_{1-x})\text{O}_3.$$ \hspace{1cm} (3)

Figure 4 depicts the variation of $\gamma_{\text{In}_2\text{O}_3}$ and $\gamma_{\text{Ga}_2\text{O}_3}$ as a function of $T_G$. Note that the definitions of $\gamma_{\text{In}_2\text{O}_3}$ and $\gamma_{\text{Ga}_2\text{O}_3}$ do not take into account the difference in cation densities due to different possible crystal phases during (In$_x$Ga$_{1-x}$)$_2$O$_3$ growth. Therefore, we assume a maximum error of $\gamma_{\text{In}_2\text{O}_3}$ and $\gamma_{\text{Ga}_2\text{O}_3}$ of $\pm 10 \%$ based on the difference in cation densities of bixbyte In$_2$O$_3$ and monoclinic Ga$_2$O$_3$. This difference, however, does not change the qualitative evolution of $\gamma$ as a function of $T_G$. The evolution of $\gamma_{\text{Ga}_2\text{O}_3}$ follows the same trend as the one for $I_i(\text{Ga}_2\text{O}_3)$ plotted in Fig. 1. In contrast, $\gamma_{\text{In}_2\text{O}_3}$ is strongly reduced for $T_G > 600 \, ^\circ\text{C}$ and $r_{\text{Me}} > 0.1$ compared to the binary $I_i(\text{In}_2\text{O}_3)$ depicted in Fig. 1. However, the inset shows a constant $\gamma$ at $r_{\text{Me}} = 0.1$ for both materials up to $T_G = 800 \, ^\circ\text{C}$.

Based on the data plotted in Figs. 2–4, we can define a range of $r_{\text{Me}}$ where $x$ decreases with $T_G$,

$$0.1 < r_{\text{Me}} < 2.0.$$ \hspace{1cm} (4)

For $r_{\text{Me}} \leq 0.1$ (inset in Figs. 2–4 for $r_{\text{Me}} = 0.1$) all In is incorporated, whereas, for $r_{\text{Me}} \geq 2.0$ (crossed symbols in Figs. 2–4 for $r_{\text{Me}} = 2.0$) no In incorporation could be measured.

Our results strongly suggest that the presence of Ga inhibits the incorporation of In whereas the presence of In has no qualitative effect on Ga incorporation. However, considering kinetic factors, one would expect incorporation of In to be higher than that of Ga since the oxidation efficiency of In into In$_2$O$_3$ is higher than that of Ga into Ga$_2$O$_3$ and the vapor pressure of In$_2$O is lower than the
one of Ga2O3. To explain the contradicting results between $\Gamma$ of the binary grown In2O3 and Ga2O3, and $\gamma$ derived from the ternary growth, we suggest the suppressed In incorporation could be of thermodynamic origin since the Ga–O bond energy is higher than the In–O bond energy, as calculated by first-principles density functional calculations.\textsuperscript{19} We verify this hypothesis in two ways: (i) We did complementary etch-experiments to try to decompose the In–O and Ga–O bonds with Ga and In, respectively. (ii) We performed thermochemical calculations (see the supplementary material).

In a former publication we have already shown that In2O3 and Ga2O3 layers can be etched (i.e., decomposed into Me2O which then may desorb off or diffuse on the growth surface) by In and Ga, respectively.\textsuperscript{4} In the present etch-experiment, we exposed the In2O3 layer to $\Phi_{\text{Ga}}$, and reciprocally, the Ga2O3 layer to $\Phi_{\text{In}}$, to see if one of the metals decomposes the oxide layer. Potential decomposition reactions read as

\[
\begin{align*}
6\text{Ga}(a) + \text{In}_2\text{O}_3(s) & \rightarrow 3\text{Ga}_2\text{O}(g) + 2\text{In}(a \text{ or } g), \quad (5) \\
4\text{Ga}(a) + \text{In}_2\text{O}_3(s) & \rightarrow 2\text{Ga}_2\text{O}(g) + \text{In}_2\text{O}(a \text{ or } g), \quad (6) \\
6\text{In}(a) + \text{Ga}_2\text{O}_3(s) & \rightarrow 3\text{In}_2\text{O}(g) + 2\text{Ga}(a \text{ or } g), \quad (7) \\
4\text{In}(a) + \text{Ga}_2\text{O}_3(s) & \rightarrow 2\text{In}_2\text{O}(g) + \text{Ga}_2\text{O}(a \text{ or } g), \quad (8)
\end{align*}
\]

where $a$, $g$, and $s$ denote the adsorbate, gaseous, and solid phase, respectively. During the exposure of an In2O3 layer to $\Phi_{\text{Ga}}$, the desorption of Ga2O was detected by QMS and the intensity of the reflected laser signal changed. These results indicate that reactions (5) and/or (6) occurred. In contrast, In did not etch the Ga2O3 layer, as only desorbing In atoms were detected by QMS and the intensity of the detected laser signal remained constant. These observations indicate that reactions (7) and (8) did not take place.

Furthermore, our thermochemical calculations yield a negative Gibbs free enthalpy $\Delta G$ for reactions (5) and (6) and a positive one for reactions (7) and (8) (see the supplementary material for more details). Therefore reactions (5) and (6) should be feasible and reactions (7) and (8) should be hindered.

Based on the results presented above we conclude that the incorporation of Ga into $(\text{In}_x\text{Ga}_{1-x})_2\text{O}_3$ is thermodynamically favored compared to the one of In. Particularly, our etch-experiments indicate that during $(\text{In}_x\text{Ga}_{1-x})_2\text{O}_3$ growth, already oxidized In may be replaced by Ga, resulting in a segregation of In at the growth surface. A similar phenomenon has already been reported for $(\text{In}_x\text{Ga}_{1-x})_2\text{O}_3$ MOVPE growth where it is suggested that In floats on the growth surface.\textsuperscript{17} Similarly, In segregation at the growth surface has been observed during MBE growth of $(\text{In}_x\text{Ga}_{1-x})_2\text{In}_x$.\textsuperscript{20} In case of $(\text{In}_x\text{Ga}_{1-x})_2\text{O}_3$ MBE growth, In atoms segregate either as In droplets or In2O which desorbs at elevated $T_G$, explaining
the decrease of $x$ and $y_{\text{In}_2\text{O}_3}$ with increasing $T_G$ in Figs. 3 and 4. This explanation is in accordance with our observations that increasing $\Phi_O$ and/or $x_{\text{nom}}$ for $r_{\text{Me}} < 1$ leads to a higher $x$ at given $T_G$. We suggest that at higher $\Phi_O$ Ga adatoms may experience a higher diffusion barrier, and therefore, they may be already oxidized into Ga$_2$O$_3$ before etching the In–O bonds. This explanation holds also true for an increased $x_{\text{nom}}$ since (statistically) more In atoms may be oxidized into In$_2$O$_3$ and are not decomposed by Ga adatoms leading to an increase of $x$ at same $T_G$.

In summary, we have studied the metal incorporation during the plasma-assisted oxide molecular beam epitaxial growth of (In$_x$Ga$_{1-x}$)$_3$O$_4$. The metal fluxes, metal-to-oxygen ratios, and growth temperature were systematically changed in order to understand the competing In and Ga incorporation into (In$_x$Ga$_{1-x}$)$_3$O$_3$. During binary growth, the metal incorporation in In$_2$O$_3$ and Ga$_2$O$_3$ is kinetically limited by the suboxide desorption of In$_2$O and Ga$_2$O, respectively. During ternary growth the Ga incorporation is not affected by the presence of In adatoms and remains kinetically limited by Ga$_2$O desorption. In contrast, the In incorporation into (In$_x$Ga$_{1-x}$)$_3$O$_3$ is controlled by the presence of Ga.

Our experiments revealed that Ga atoms are more favorably incorporated in (In$_x$Ga$_{1-x}$)$_3$O$_3$ than In atoms. We conducted etch-experiments that revealed that the Ga–O bonds are stronger than the In–O bonds, and therefore, already oxidized In atoms may be reduced by Ga adatoms. These experiments unveil that the Ga incorporation is thermodynamically favored compared to In incorporation, as also supported by our thermochemical calculations. Furthermore, we found that In either accumulates as droplets at the growth surface for growth temperatures below 600 °C or desorbs eventually as In or In$_2$O for growth temperatures above 600 °C. However, excess of oxygen kinetically enables (full) In incorporation for all investigated growth temperatures in the range from 500 °C to 800 °C. The thermodynamically favored Ga incorporation and kinetically enabled In incorporation likely also explains the different In incorporation rates at low and high O-pressures observed during metal-organic vapor phase epitaxy and pulsed laser deposition of (In$_x$Ga$_{1-x}$)$_3$O$_3$. Our findings may help growing high-quality (In$_x$Ga$_{1-x}$)$_3$O$_3$ layers with desired In and Ga concentrations. They may also constitute a qualitative guidance for the growth of (In$_x$Ga$_{1-x}$)$_3$O$_3$ by ozone molecular beam epitaxy, pulsed laser deposition, and metal–organic vapor phase epitaxy.

See supplementary material for thermochemical calculations of the given reactions.

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