Preliminary studies on halide vapor phase epitaxy of AlGaN alloy on GaN substrates

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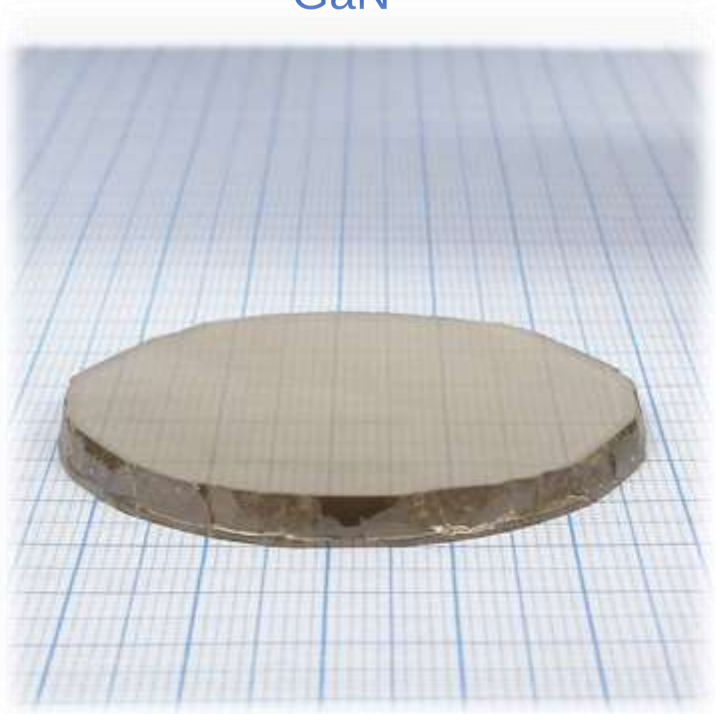
GaN – AlN – AlGaN

GaN



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Halide **V**apor **P**hase **E**pitaxy

Ammonothermal (Acidic/Basic)

Na-Flux

AlGaN

lack of
bulk

AlGaN
crystal

AlN

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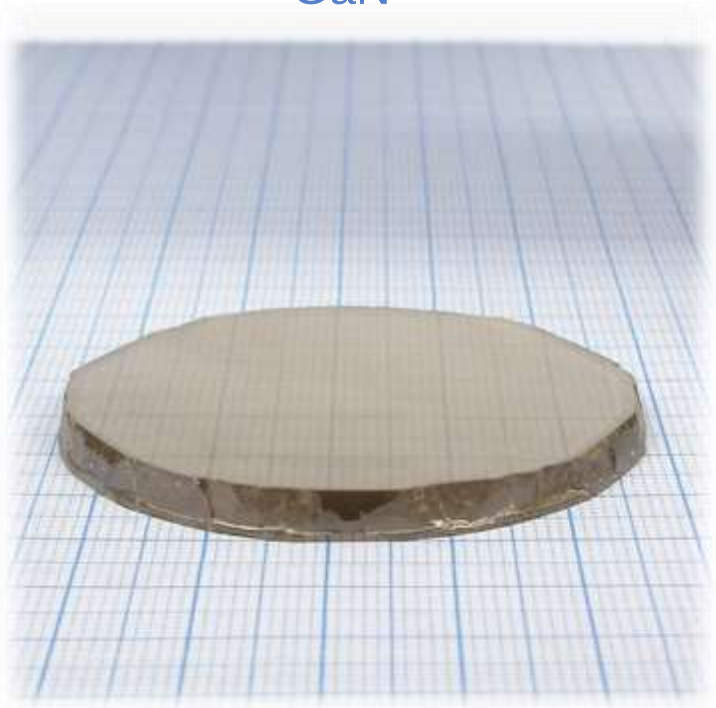
Halide **V**apor **P**hase **E**pitaxy

Physical **V**apor **T**ransport

GaN – AlN – AlGaN

GaN



http://unipress.waw.pl/

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crystal

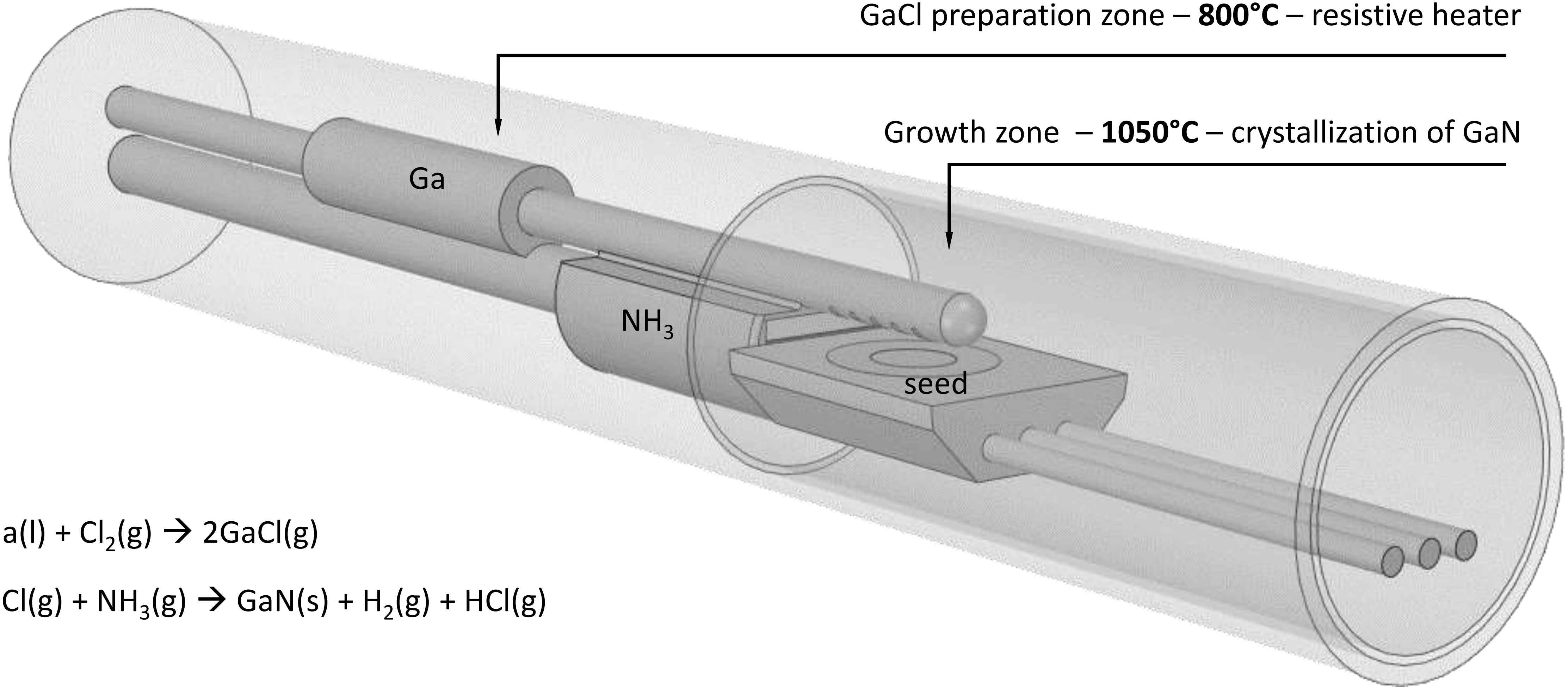
AlN

www.hexatechinc.com

Halide **V**apor **P**hase **E**pitaxy

Physical **V**apor **T**ransport

HVPE reactor for GaN growth



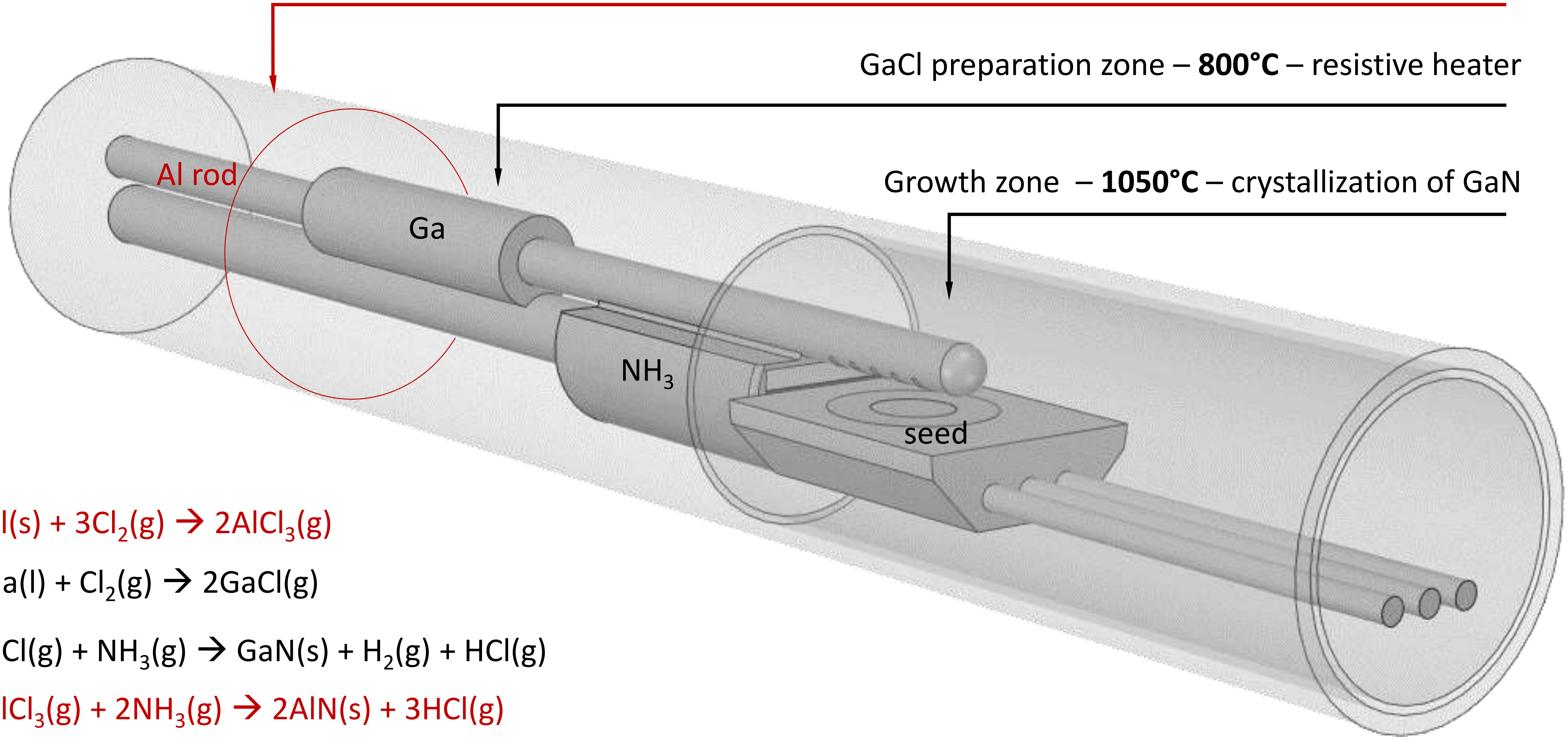
GaCl preparation zone – **800°C** – resistive heater

Growth zone – **1050°C** – crystallization of GaN

$2Ga(l) + Cl_2(g) \rightarrow 2GaCl(g)$

$GaCl(g) + NH_3(g) \rightarrow GaN(s) + H_2(g) + HCl(g)$

HVPE reactor for AlGaN growth



$AlCl_3$ preparation zone – **500°C** – passive/resistive heater

GaCl preparation zone – **800°C** – resistive heater

Growth zone – **1050°C** – crystallization of GaN

$2Al(s) + 3Cl_2(g) \rightarrow 2AlCl_3(g)$

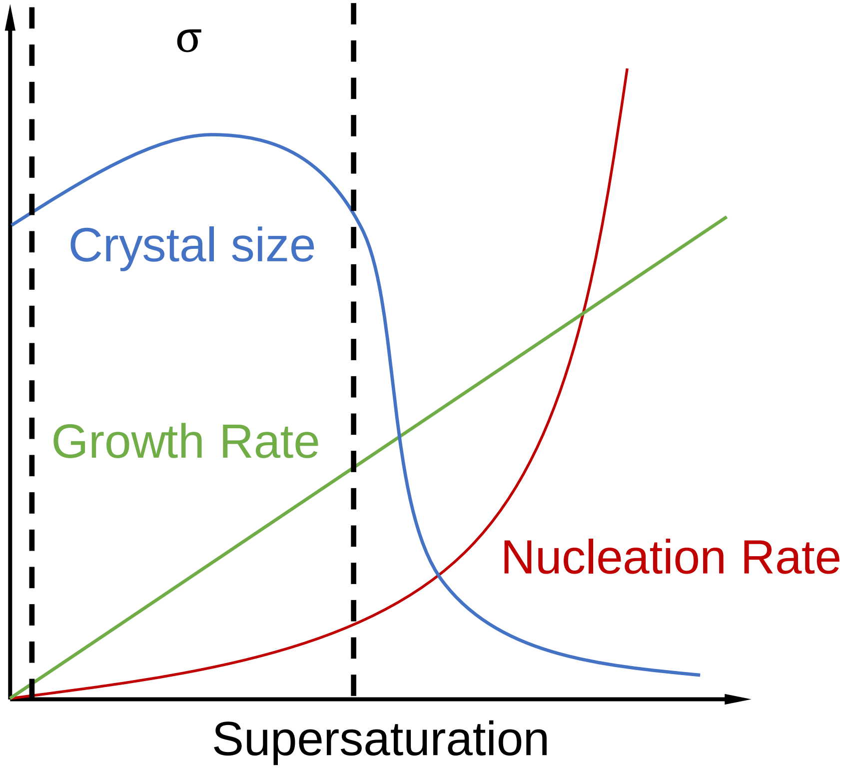
$2Ga(l) + Cl_2(g) \rightarrow 2GaCl(g)$

$GaCl(g) + NH_3(g) \rightarrow GaN(s) + H_2(g) + HCl(g)$

$2AlCl_3(g) + 2NH_3(g) \rightarrow 2AlN(s) + 3HCl(g)$

Supersaturation

Optimal conditions for HVPE
GaN and AlN growth



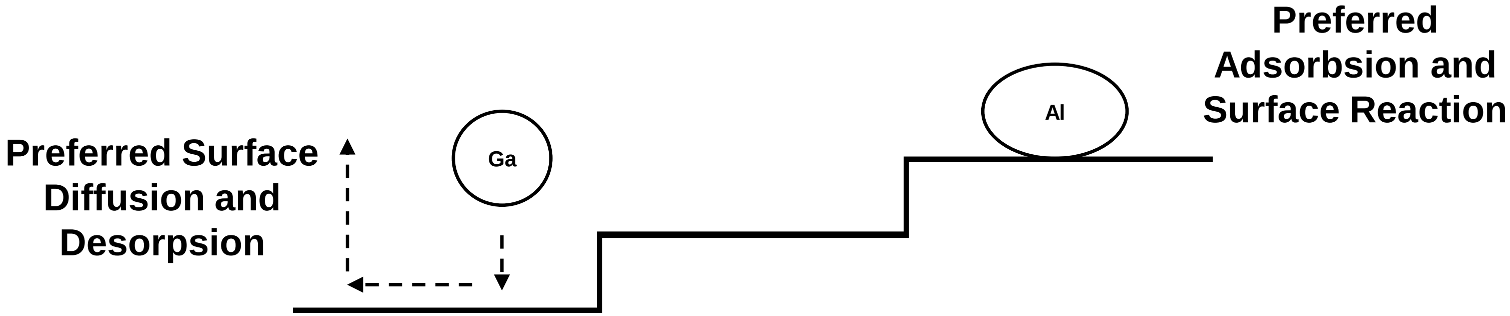
$$\sigma_{Ga} = \frac{P^{\circ}_{GaCl} - P_{GaCl}}{P_{GaCl}}$$

$$\sigma_{Al} = \frac{P^{\circ}_{AlCl_3} - P_{AlCl_3}}{P_{AlCl_3}}$$

σ - supersaturation
P_i°- input partial pressure
P_i- equilibrium partial pressure

GaN and AlN bulk growth by HVPE method

	GaN	AlN
Growth T [°C]	1050	1450
Source T [°C]	800 – 900	450 – 500
P [atm]	0.8	0.8
NH_3 [sccm/min]	1000	-
V/III	20	-
Growth rate [μm/h]	240 – 480	150 – 170*

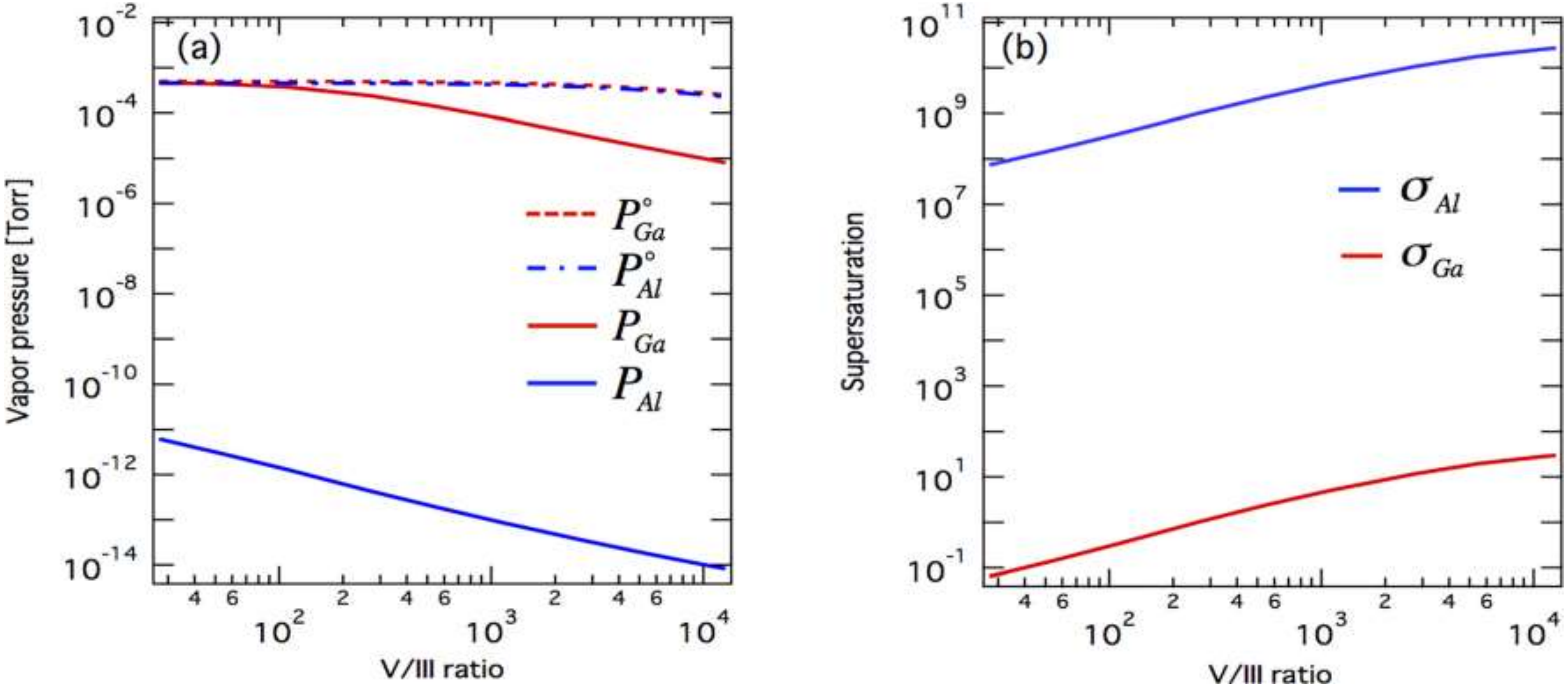


Preferred Surface Diffusion and Desorpsion

Preferred Adsorbsion and Surface Reaction

** Y. Kumagai et al. Appl. Phys. Express 15 115501 (2022)*

Difference of Supersaturation



P_i° - input partial pressure
P_i - equilibrium partial pressure

$$\sigma = \frac{P_i^\circ - P_i}{P_i}$$

Shun Washiyama et al. Journal of Applied Physics 124, 115304 (2018); doi: 10.1063/1.5045058

GaN and AlN bulk growth by HVPE method

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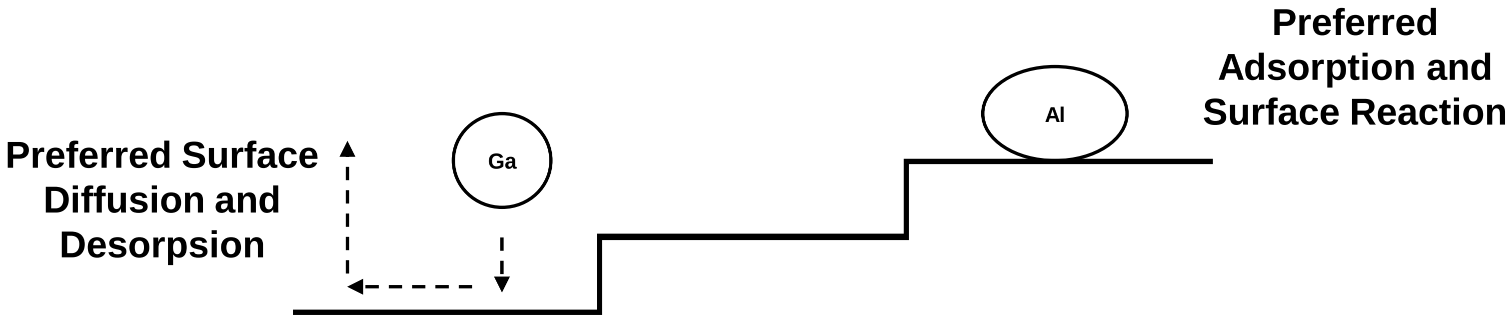


* *Y. Kumagai et al. Appl. Phys. Express 15 115501 (2022)*

Parameters that affect the saturation

Influence of various parameters on σ

	~~T ↑~~	P°_{III} ↑	*V/III ratio* ↑	~~$P^{\circ}_{H_2}$ ↑~~	α ↑
σ	~~↓~~	↑	↑	~~↓~~	↓

Constant

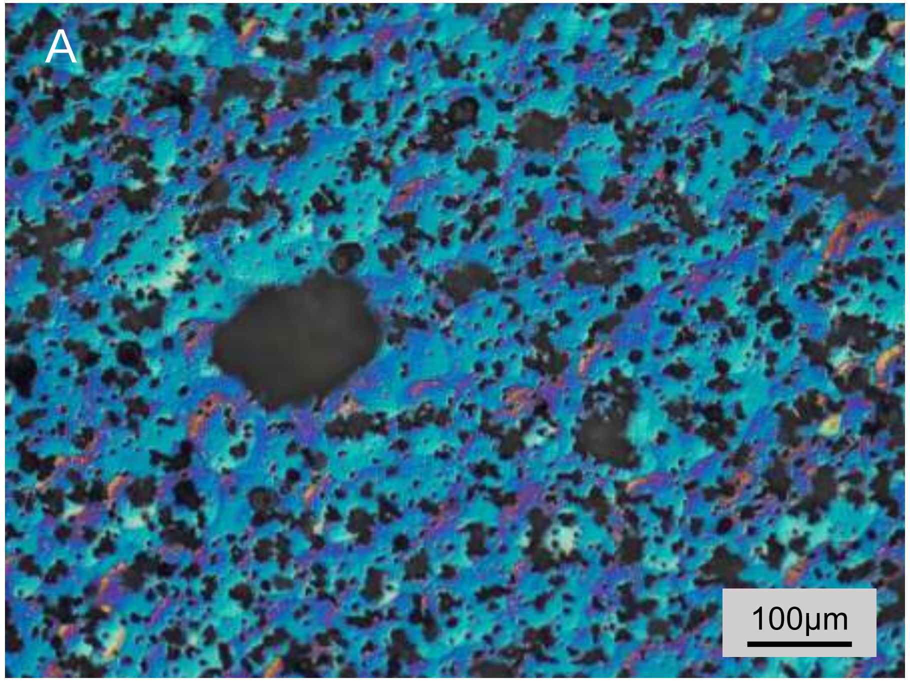
Not too low due to decomposition of GaN

Not too high due to decomposition of GaN

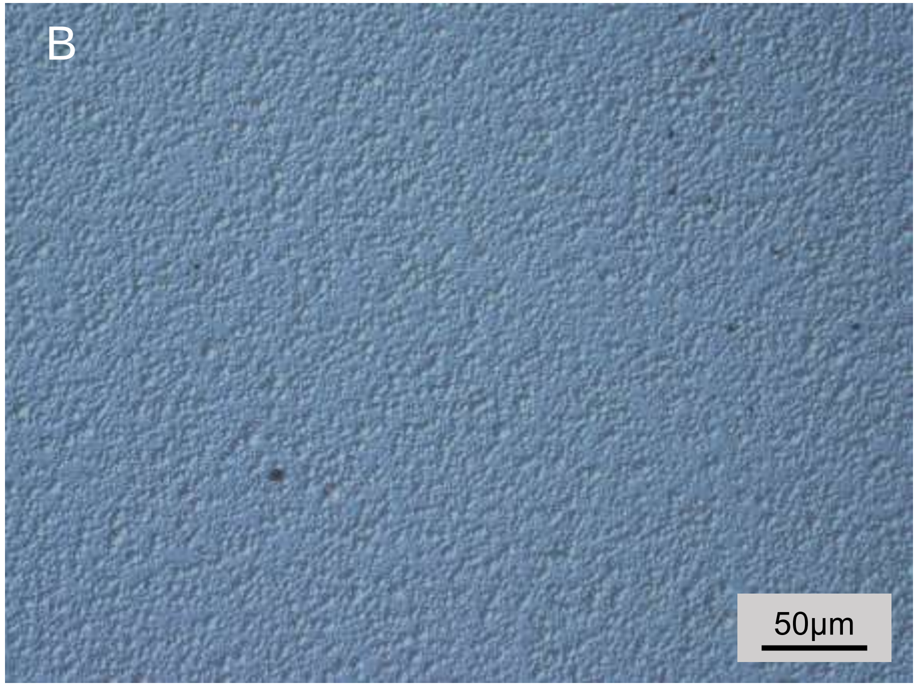
Shun Washiyama et al. Journal of Applied Physics 124, 115304 (2018)

AlGaN growth – reduction of NH_3

A – high V/III ratio



B – low V/III ratio



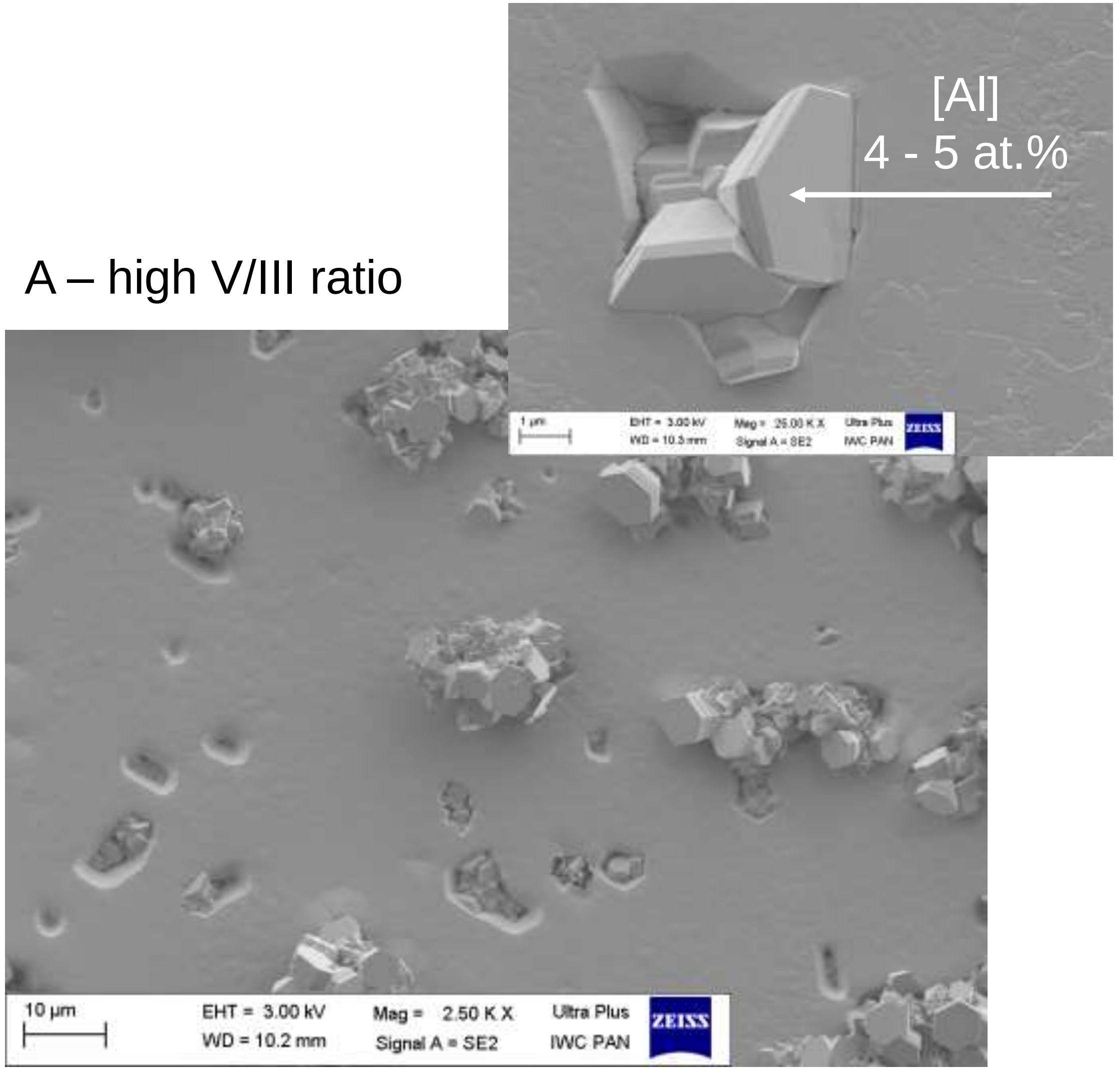
$$V/III = \frac{P^{\circ}_{NH_3}}{P^{\circ}_{AlCl_3} + P^{\circ}_{GaCl}}$$

	A	**B**
T [°C]	1050	1050
P [atm]	0.8	0.8
NH_3 [sccm/min]	**1100**	**400**
V/III	**79**	**21**
R	0.31	0.31

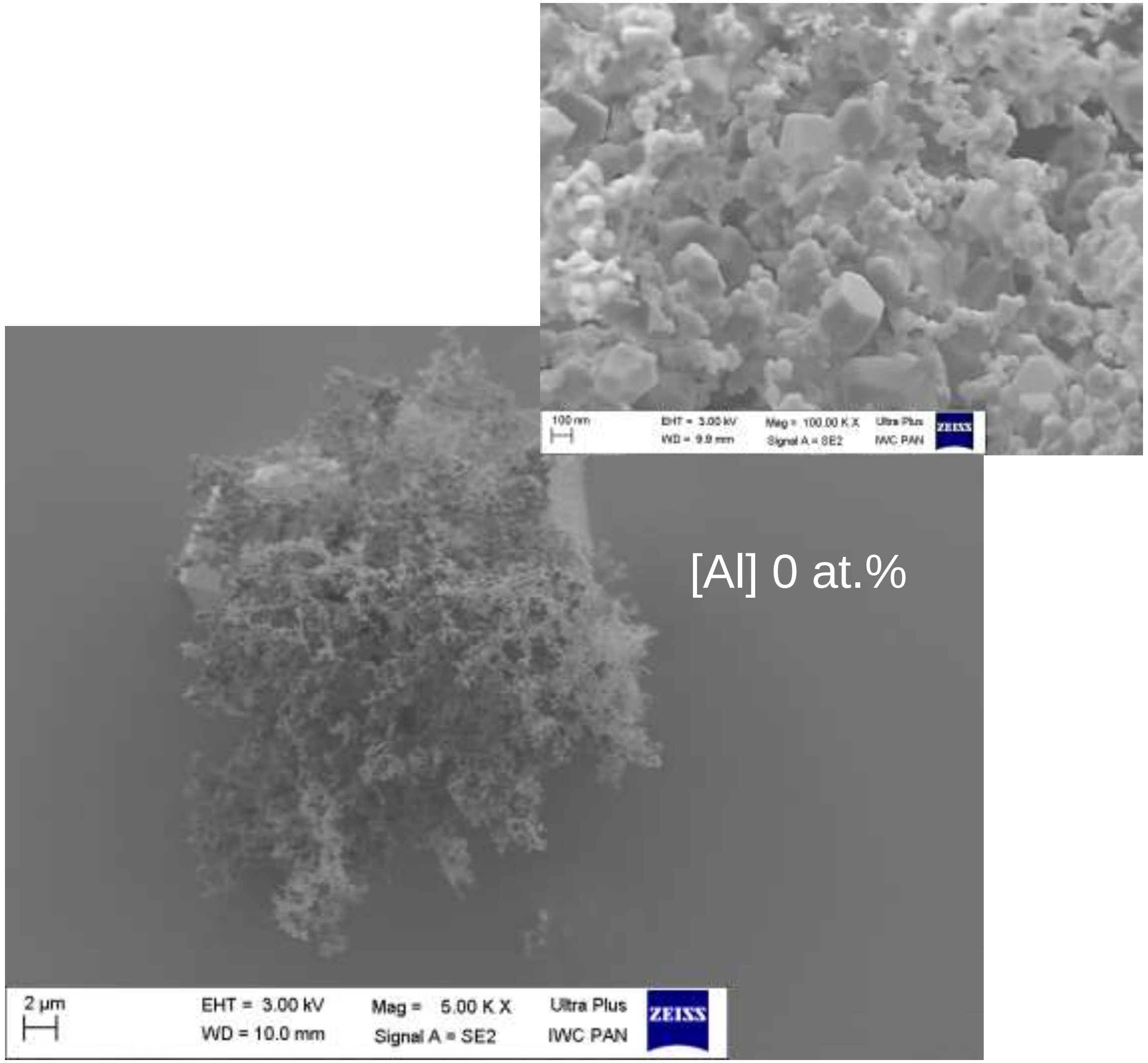
$$R = \frac{P^{\circ}_{AlCl_3}}{P^{\circ}_{AlCl_3} + P^{\circ}_{GaCl}}$$

AlGaN growth – NH_3 flow reduction

A – high V/III ratio



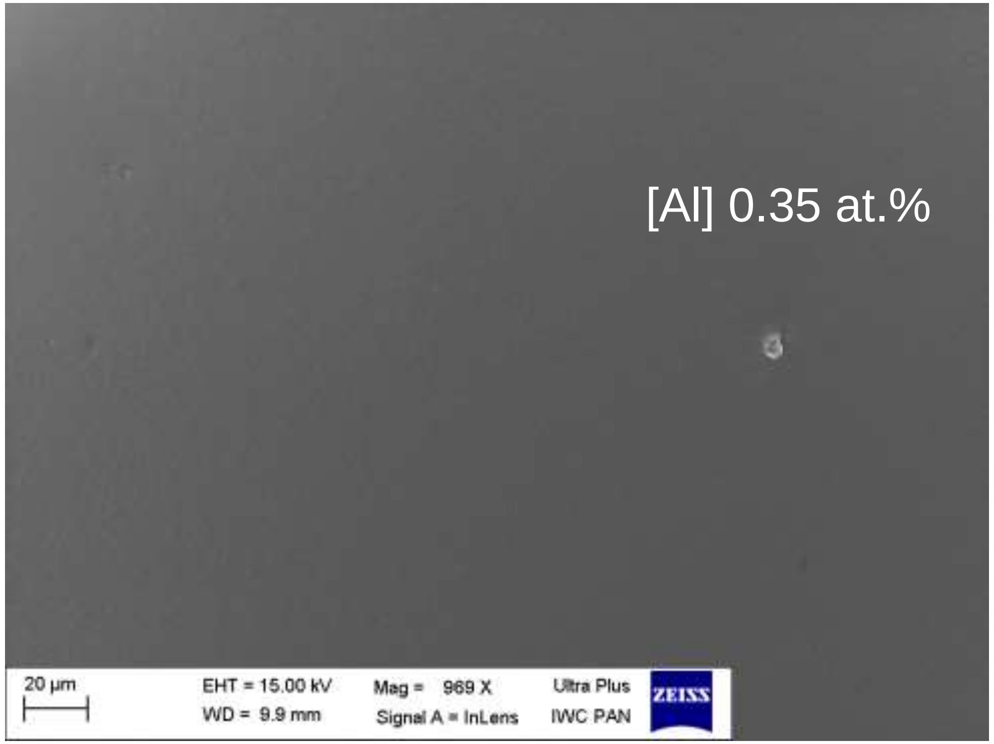
[Al]
4 - 5 at.%

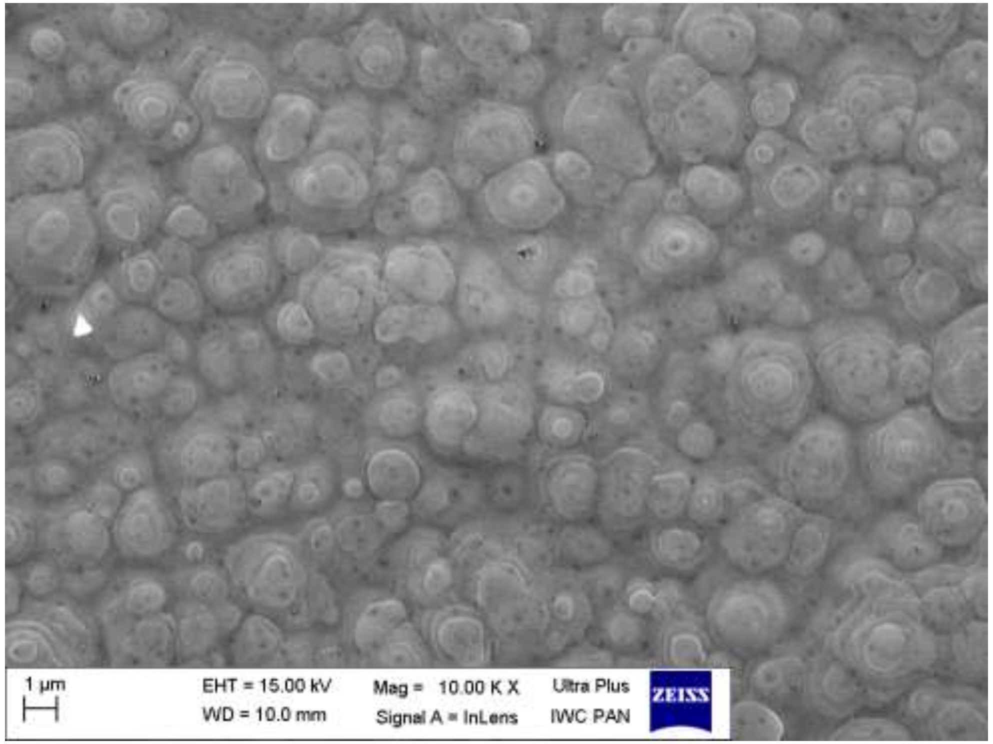


[Al] 0 at.%

AlGaN growth – NH_3 flow reduction

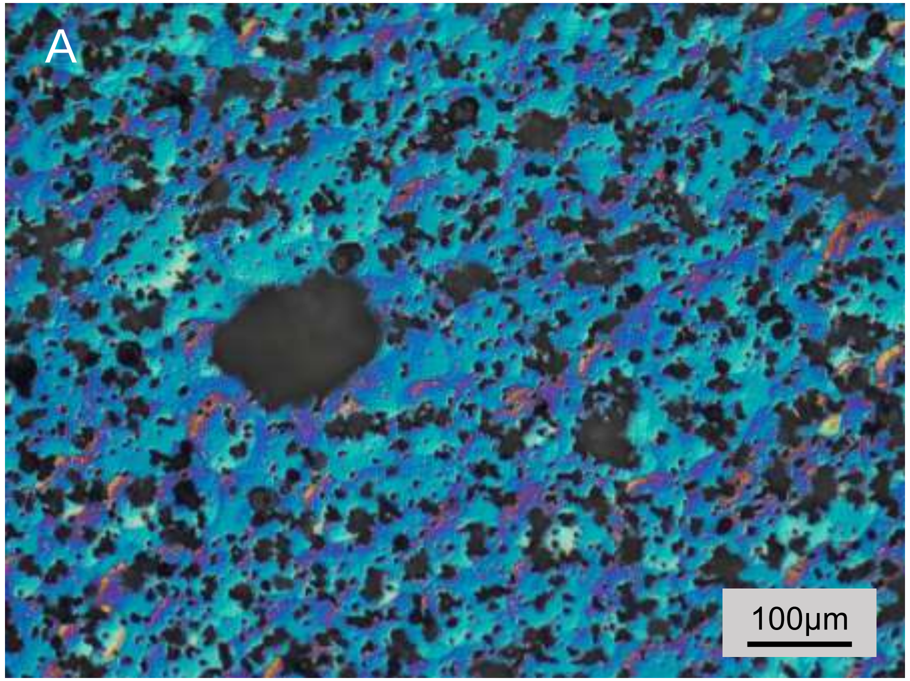
B – low V/III ratio


[Al] 0.35 at.%

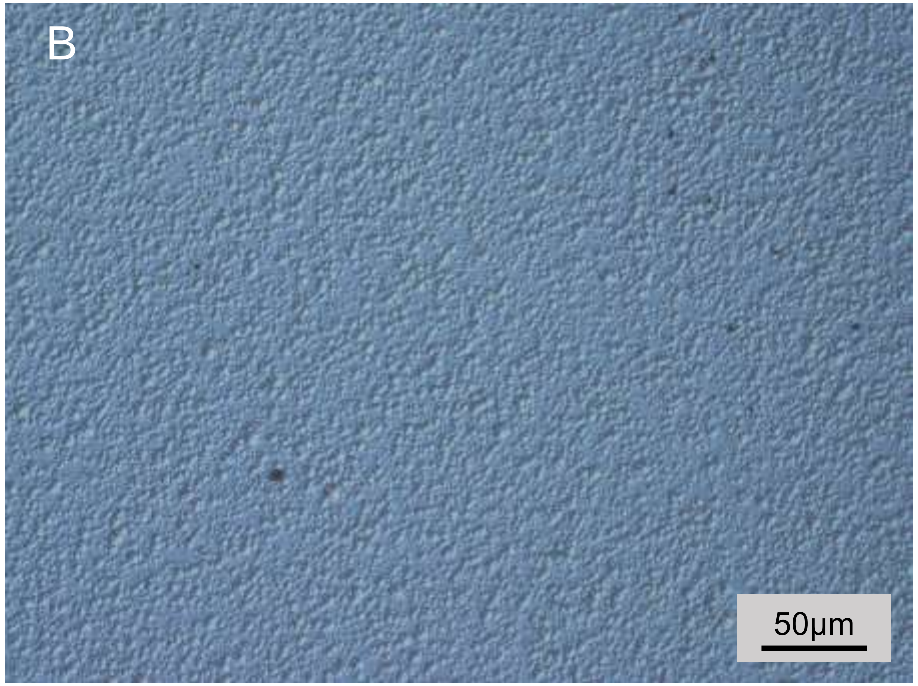


AlGaN growth – reduction of NH_3

A – high V/III ratio



B – low V/III ratio

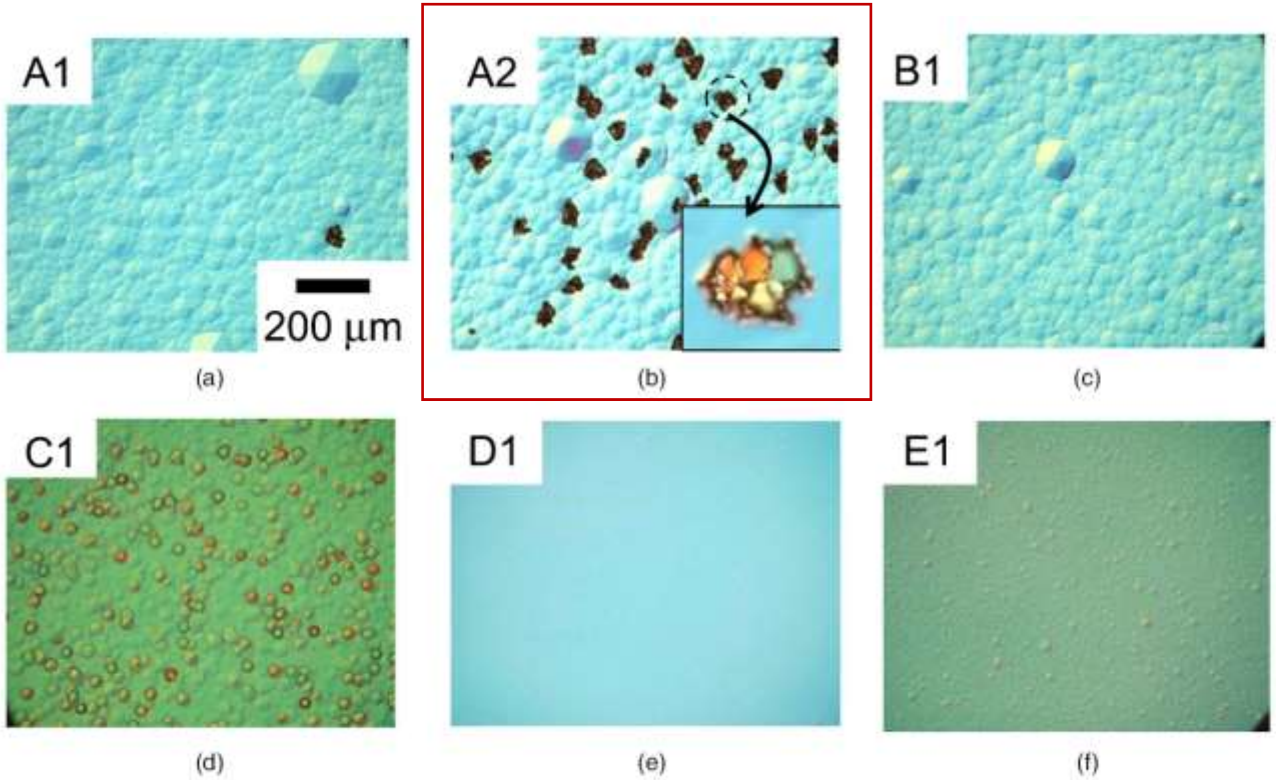


$$V/III = \frac{P^{\circ}_{NH_3}}{P^{\circ}_{AlCl_3} + P^{\circ}_{GaCl}}$$

	A	B
T [°C]	1050	1050
P [atm]	0.8	0.8
NH_3 [sccm/min]	**1100**	**400**
V/III	**79**	**21**
R	0.31	0.31

$$R = \frac{P^{\circ}_{AlCl_3}}{P^{\circ}_{AlCl_3} + P^{\circ}_{GaCl}}$$

AlGaN growth



(a) (b) (c)

(d) (e) (f)

Fig. 2. (Color online) Optical microscope images of the AlGaN surfaces of samples denoted in Fig. 1.

Applied Physics Express **15**, 085504 (2022)

https://doi.org/**10.35848/1882-0786/ac8412**

Hydride vapor phase epitaxial growth of AlGaN

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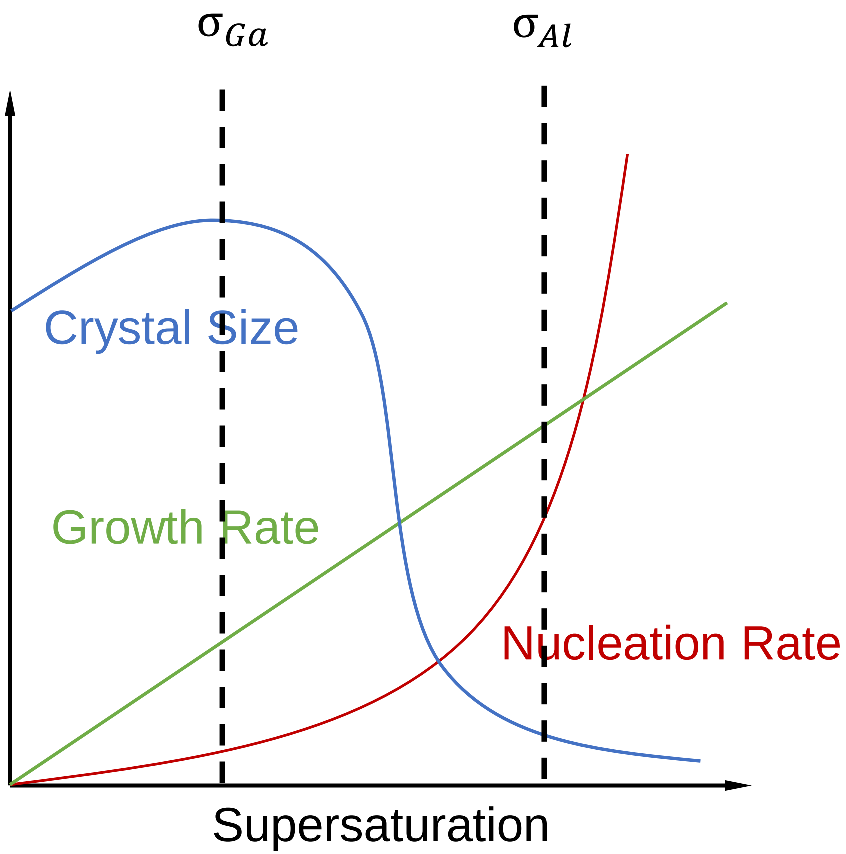
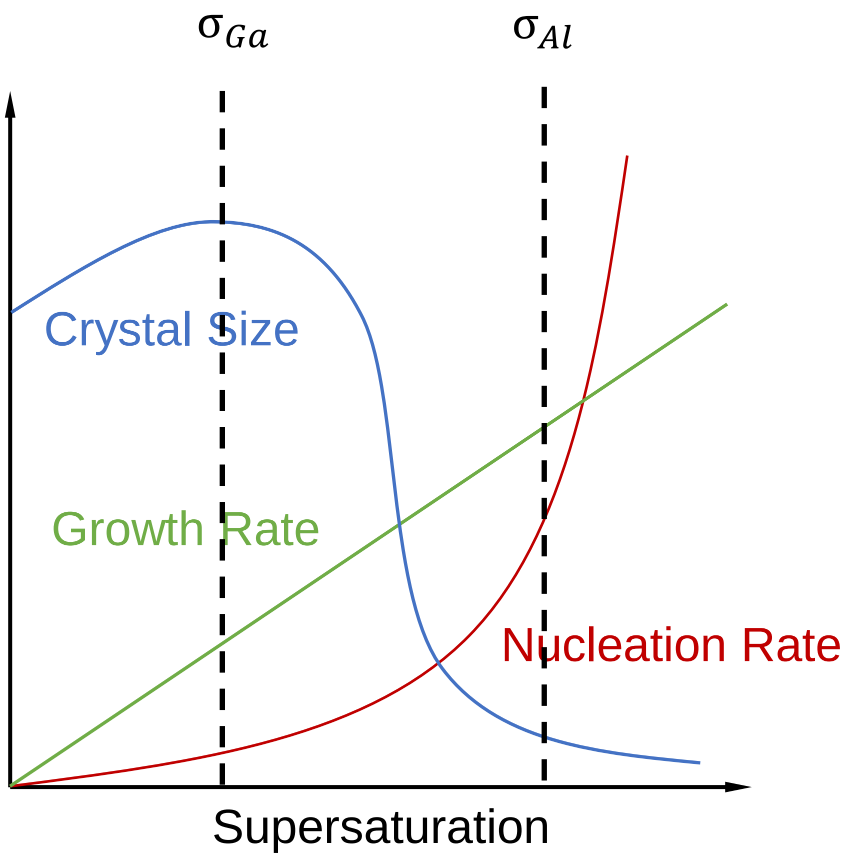
Received May 9, 2022; revised July 6, 2022; accepted July 25, 2022; published online August 3, 2022

Difference of Supersaturation

A – high V/III ratio

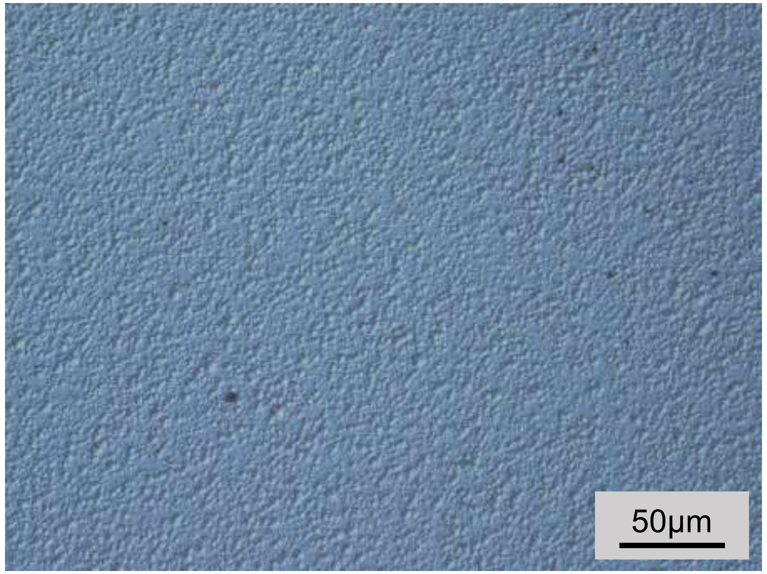


B – low V/III ratio

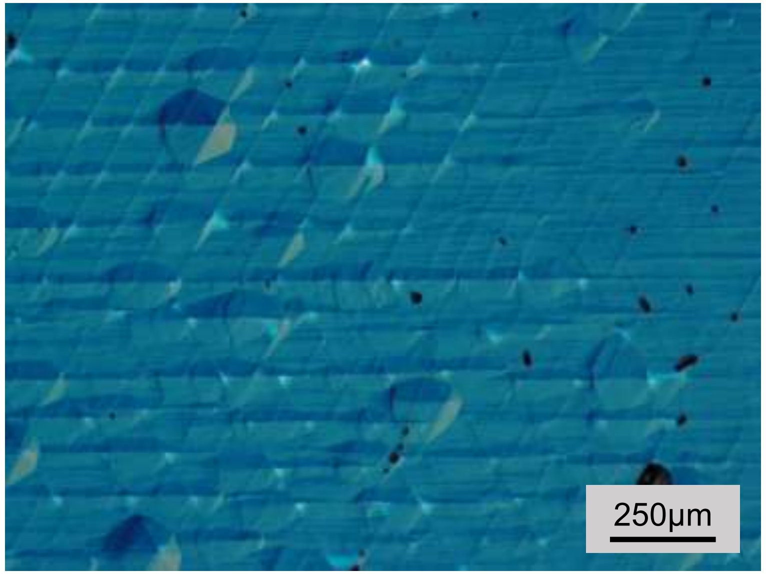


AlGaN growth – reduction of growth pressure

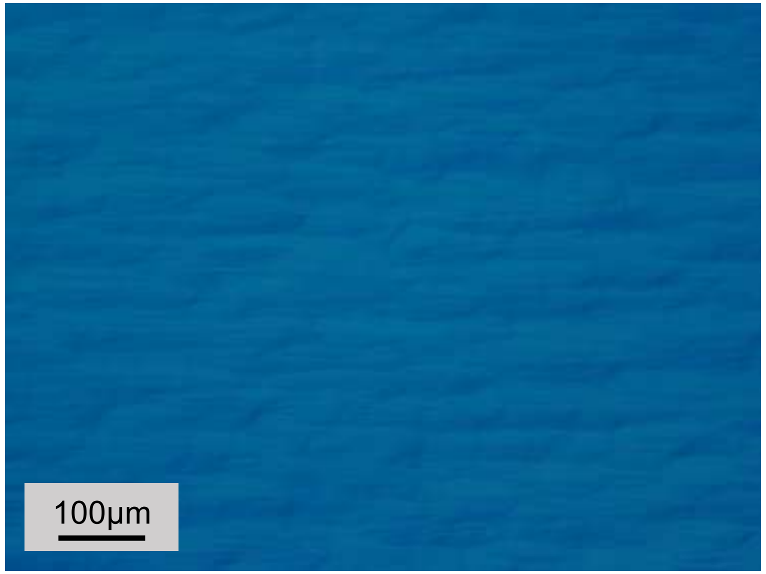
B. p = 0.8 [atm]



C. p = 0.4 [atm]



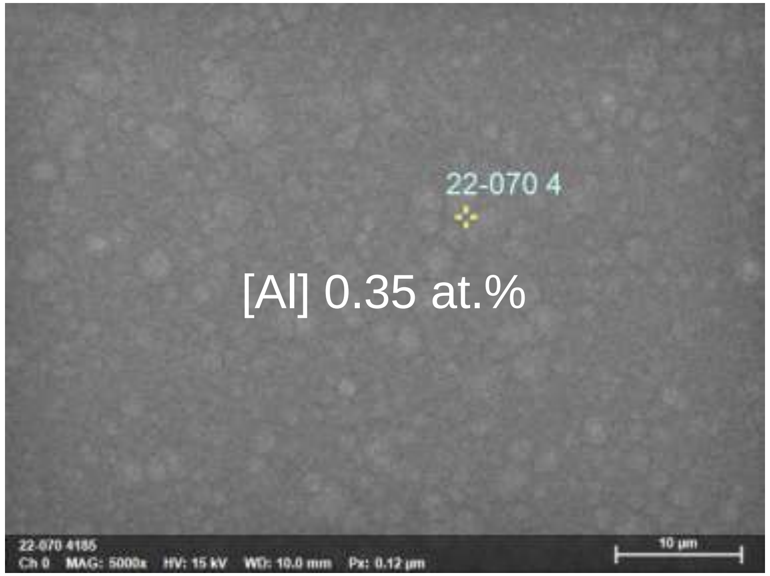
D. p = 0.2 [atm]



	B	C	D
T [°C]	1050	1050	1050
P [atm]	**0.8**	**0.4**	**0.2**
NH_3 [sccm/min]	400	400	400
V/III	21	21	21
R	0.31	0.31	0.31

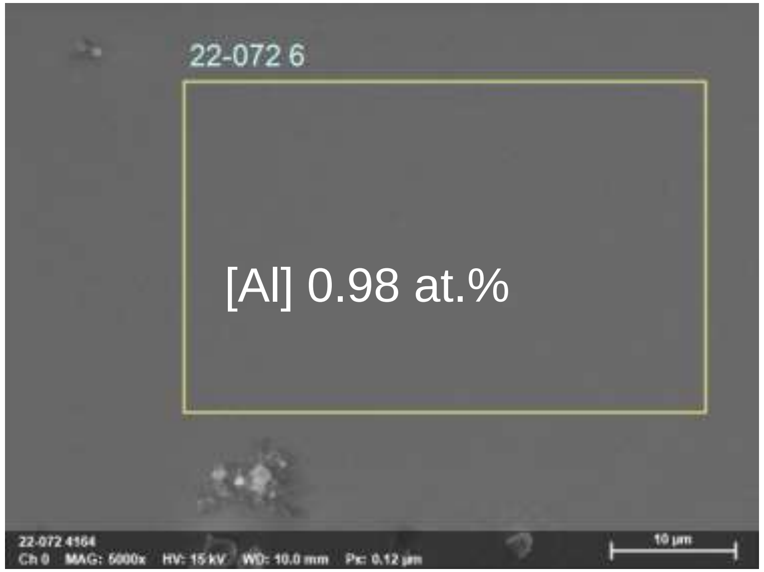
AlGaN growth – reduction of growth pressure

B. p = 0.8 [atm]



[Al] 0.35 at.%

C. p = 0.4 [atm]



[Al] 0.98 at.%

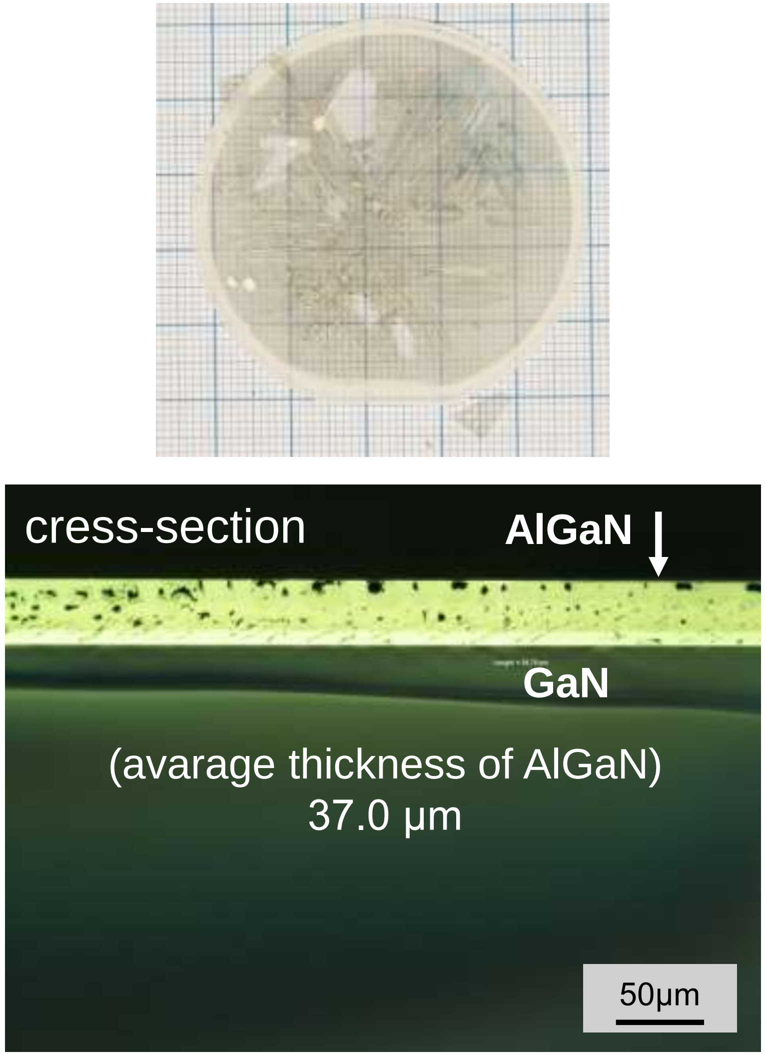
D. p = 0.2 [atm]

[Al] 2.15 at.%

	B	C	D
T [°C]	1050	1050	1050
P [atm]	**0.8**	**0.4**	**0.2**
NH_3 [sccm/min]	400	400	400
V/III	21	21	21
R	0.31	0.31	0.31

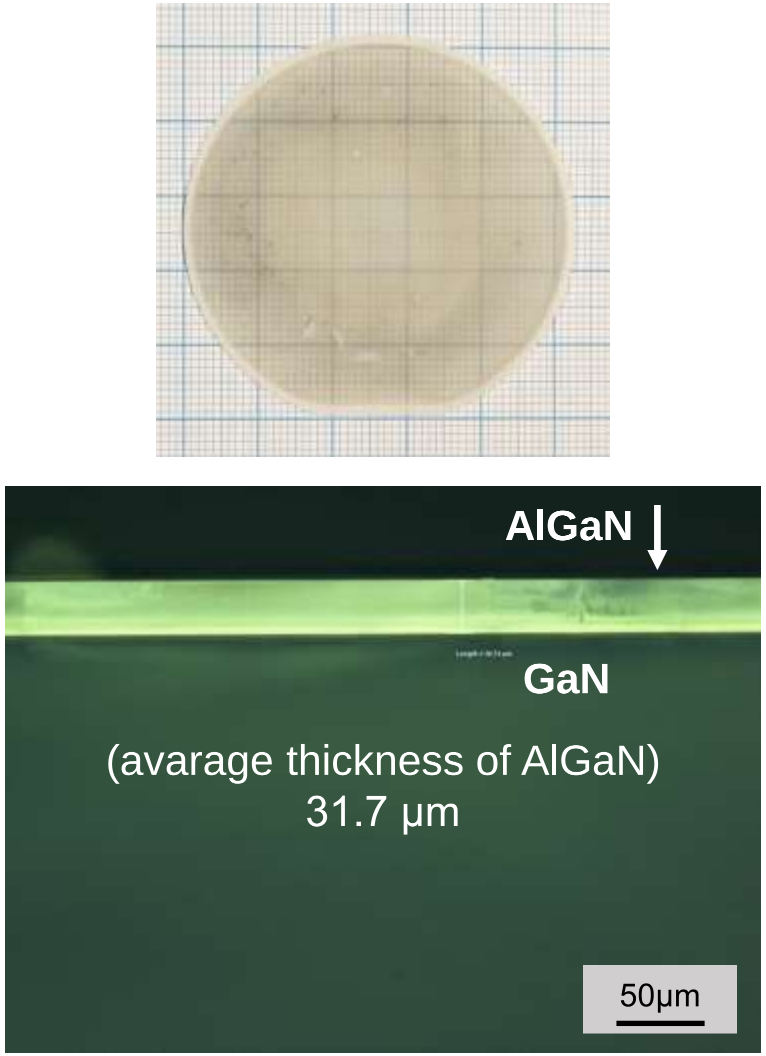
AlGaN growth – reduction of growth pressure

B. p = 0.8 [atm]



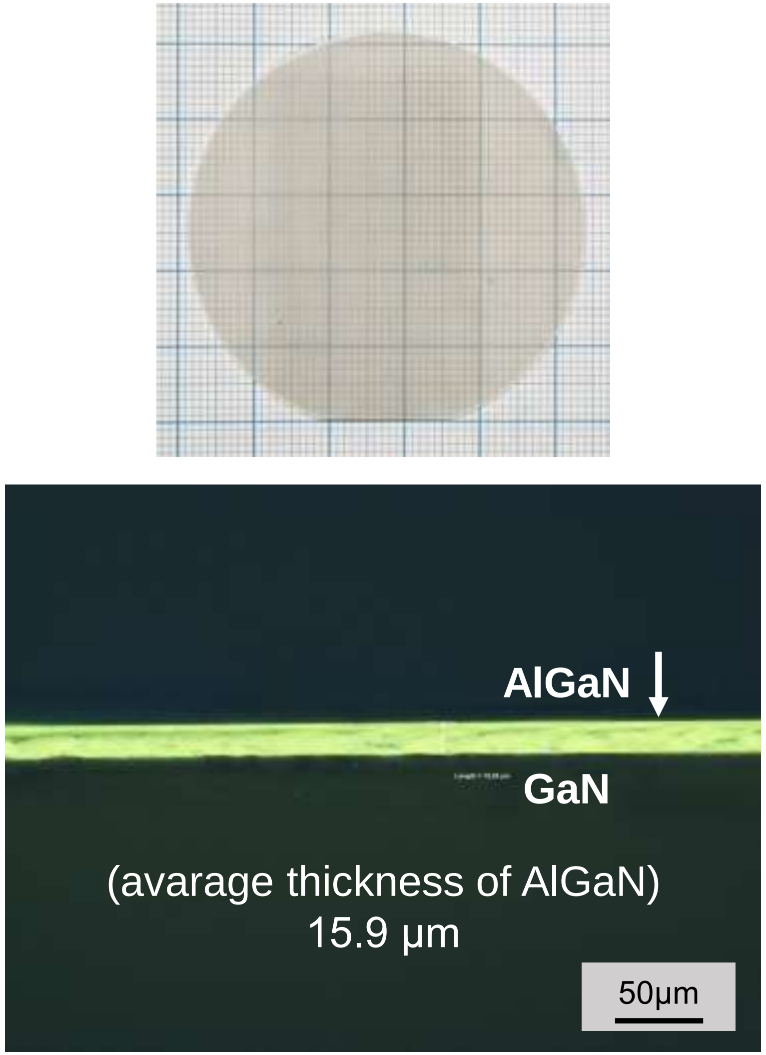
(avarage thickness of AlGaN)
37.0 µm

C. p = 0.4 [atm]



(avarage thickness of AlGaN)
31.7 µm

D. p = 0.2 [atm]

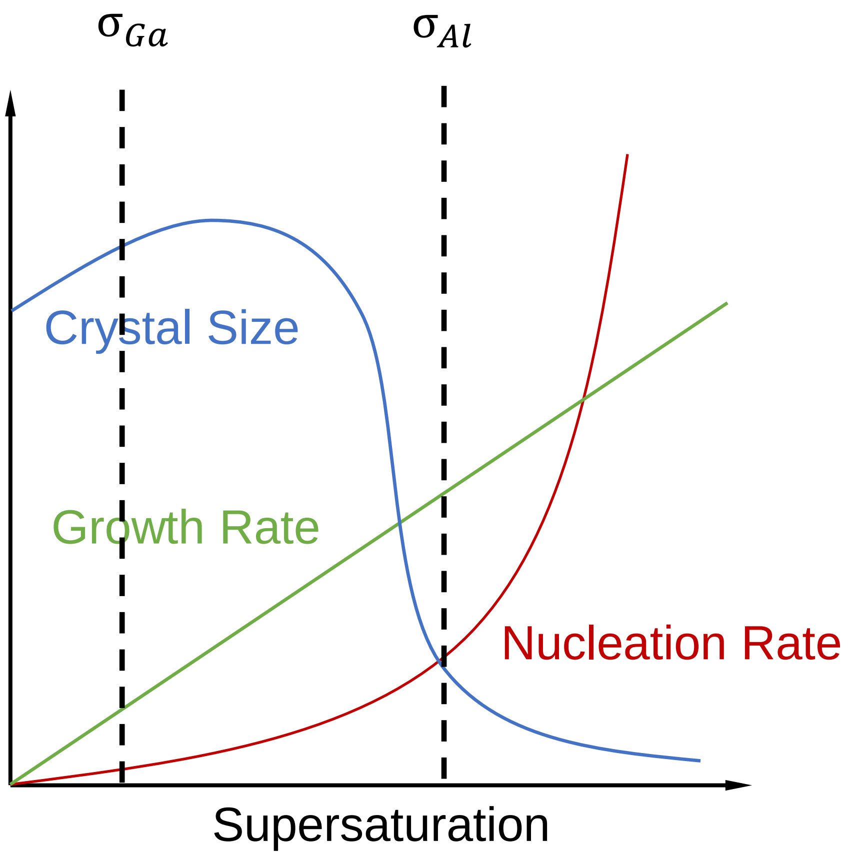


(avarage thickness of AlGaN)
15.9 µm

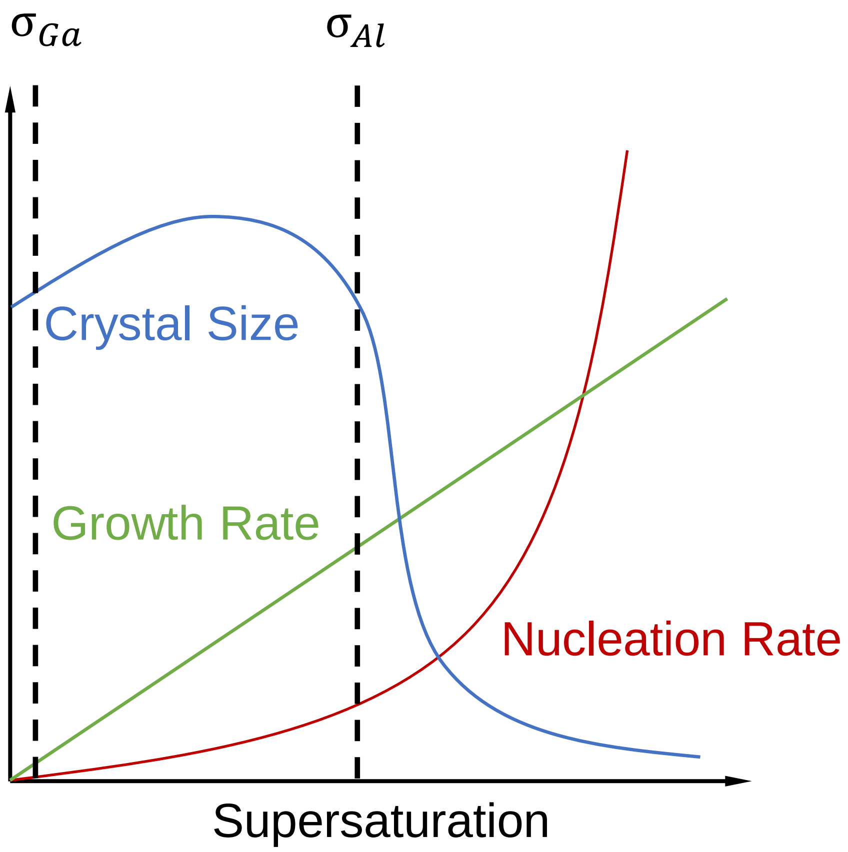
Constant growth time

Difference of Supersaturation

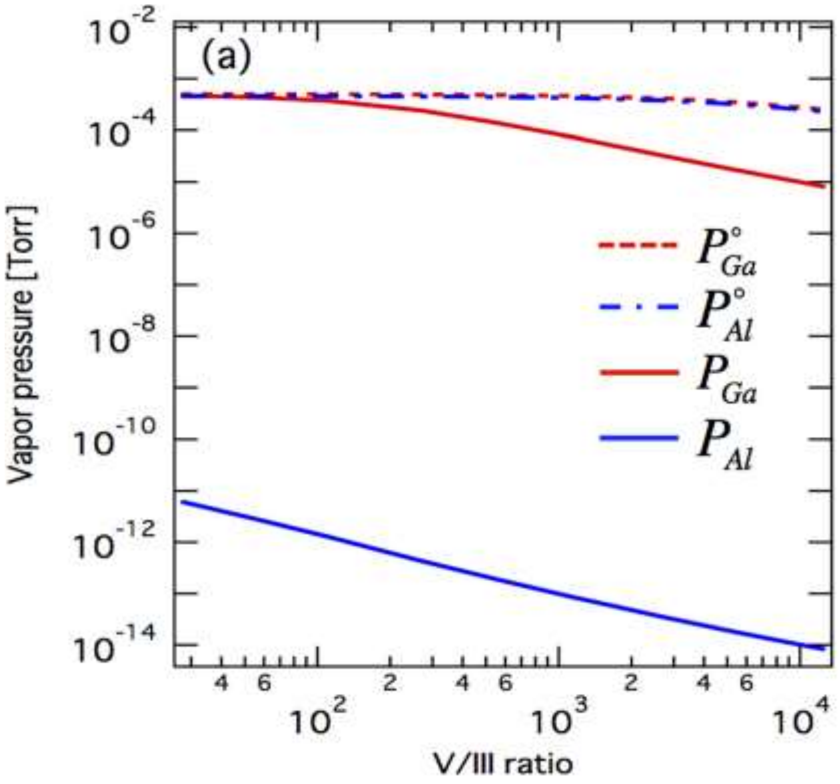
B – p = 0.8 atm



D – p = 0.2 atm

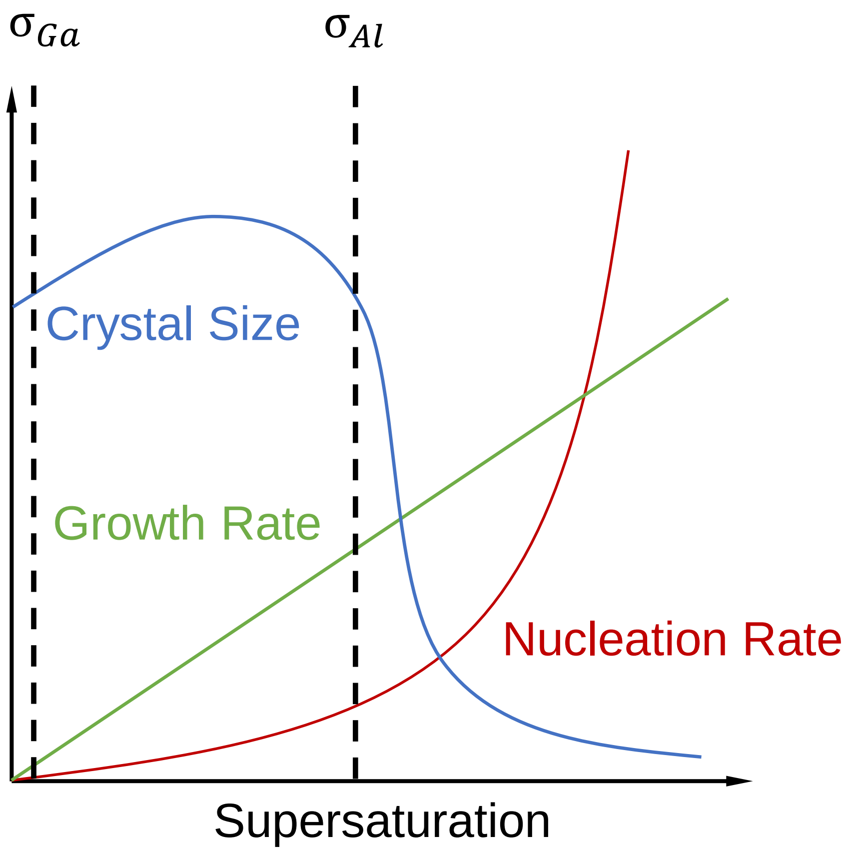


Difference of Supersaturation

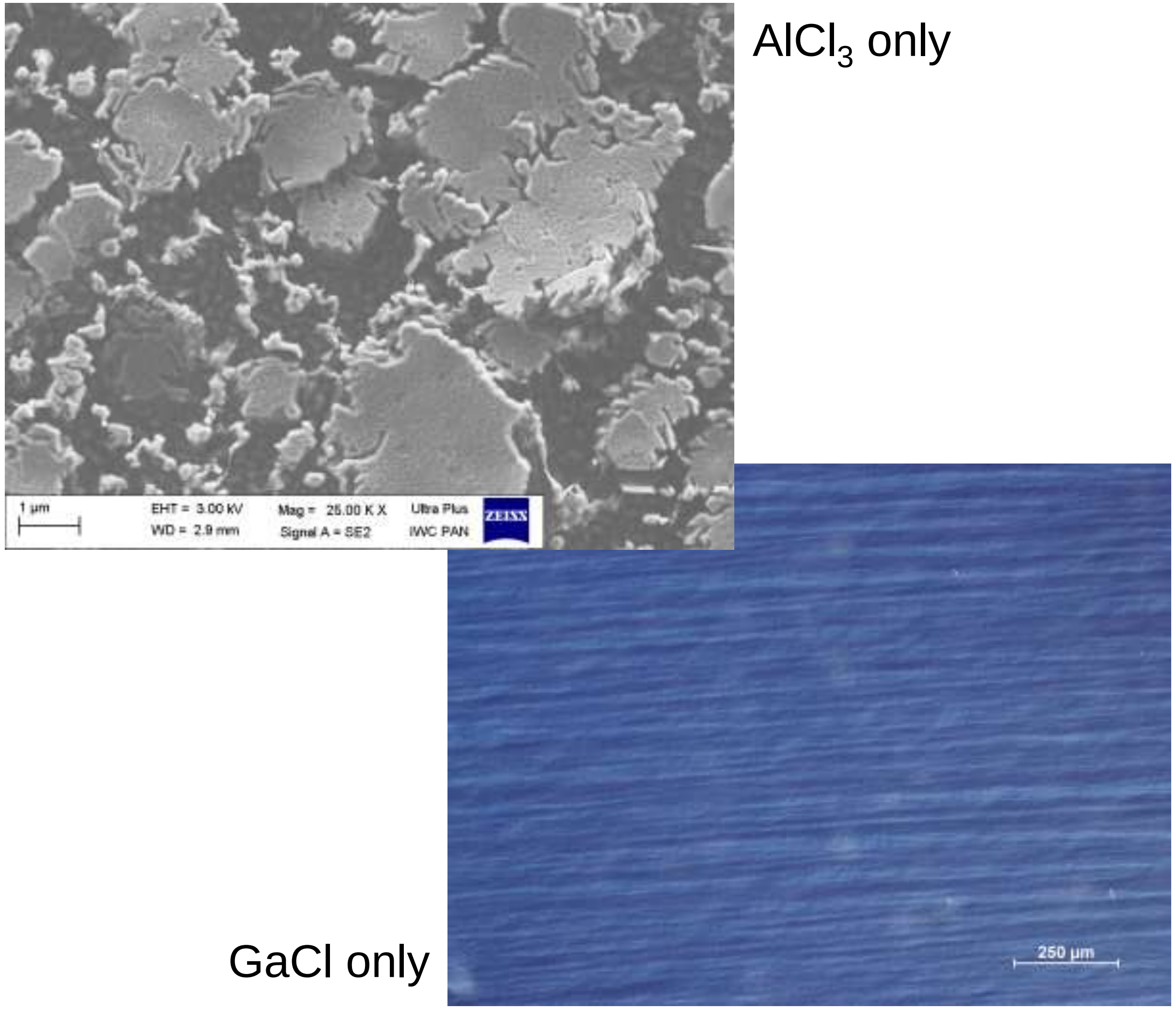


$$\sigma = \frac{P_i^\circ - P_i}{P_i}$$

D – p = 0.2 atm



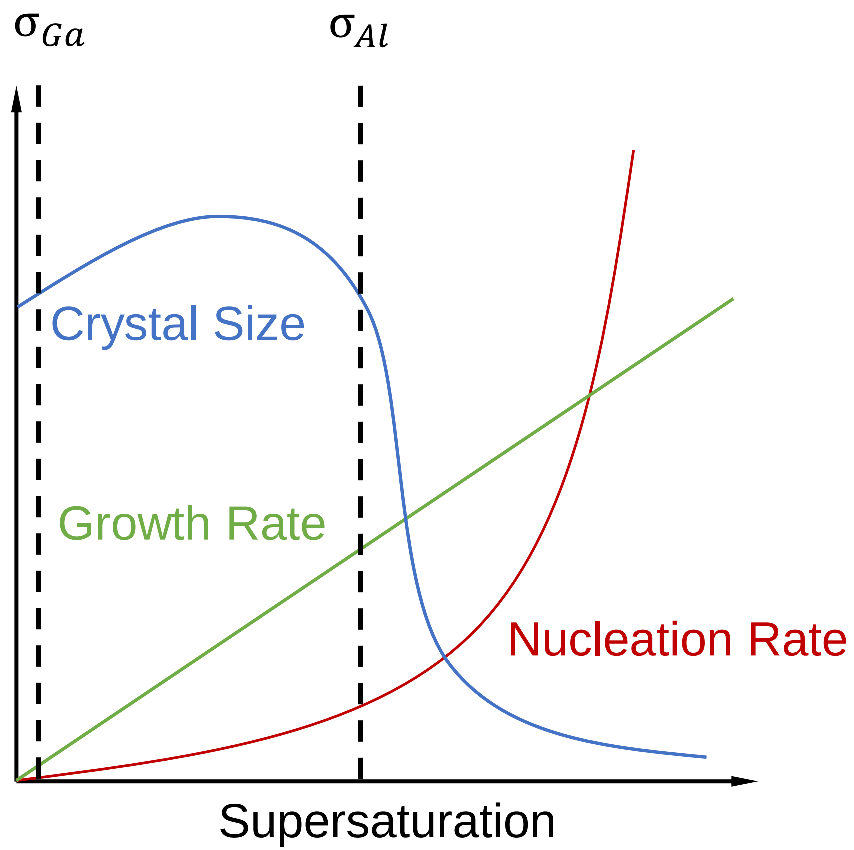
Difference of Supersaturation



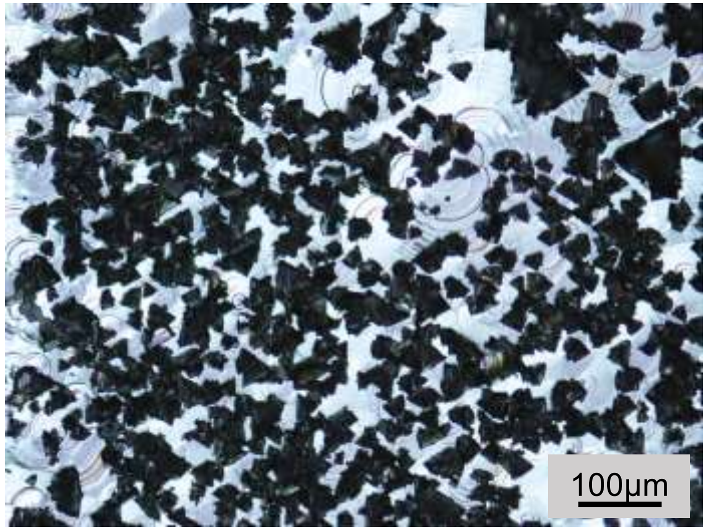
$AlCl_3$ only

GaCl only

D – p = 0.2 atm

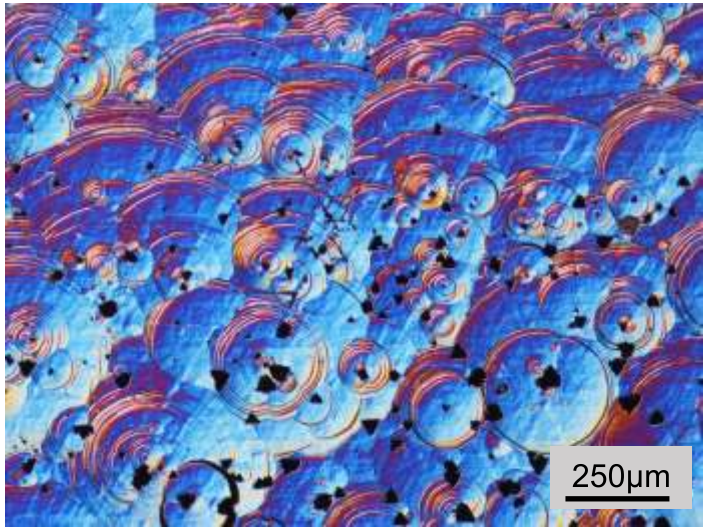


AlGaN growth – influence of the off-cut

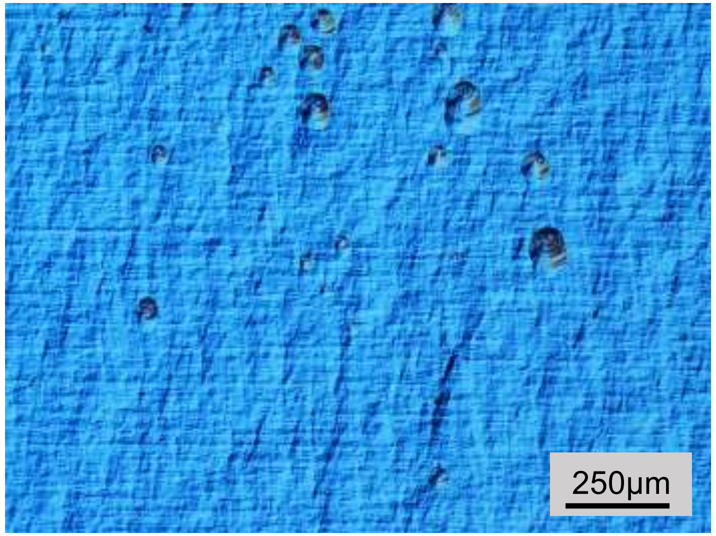


Off-cut 0.5 deg

$$R = \frac{P^{\circ}_{AlCl_3}}{P^{\circ}_{AlCl_3} + P^{\circ}_{GaCl}}$$

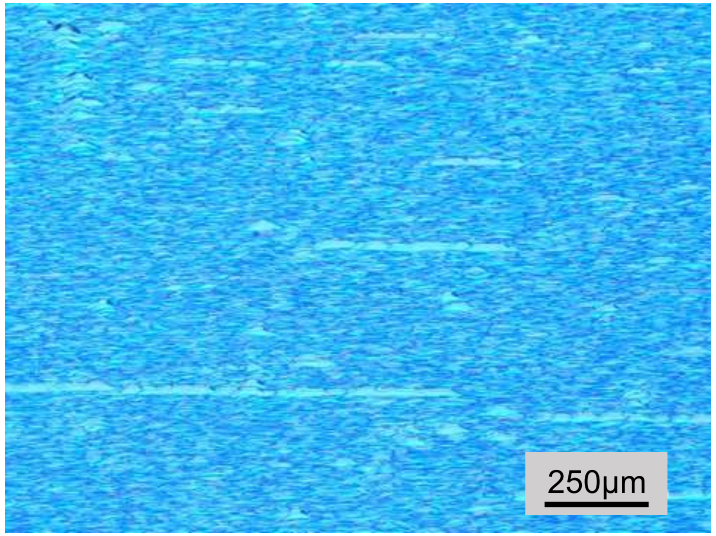


Off-cut 1.0 deg

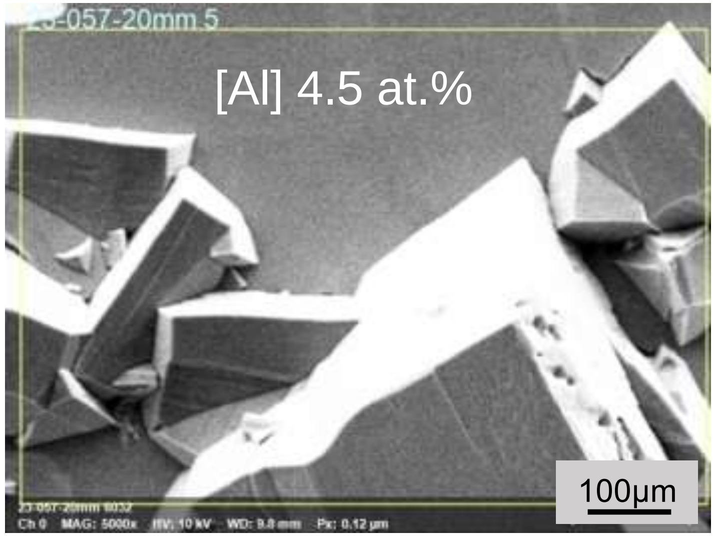


Off-cut 2.0 deg

Off-cut 4.0 deg

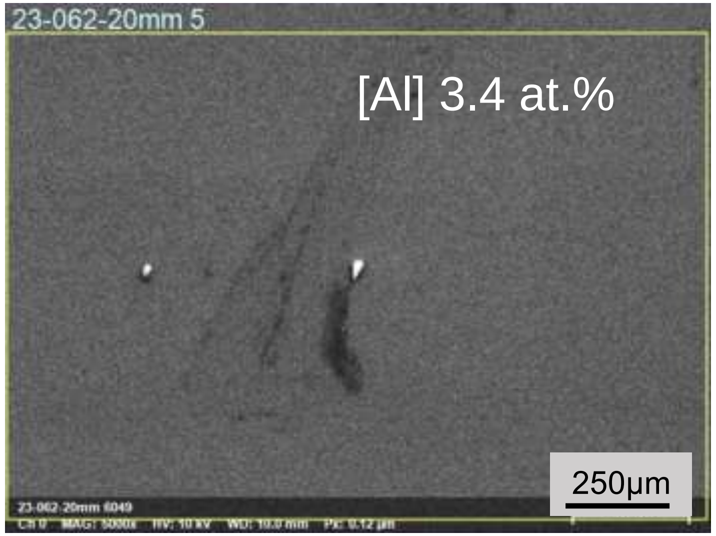


AlGaN growth – influence of the off-cut



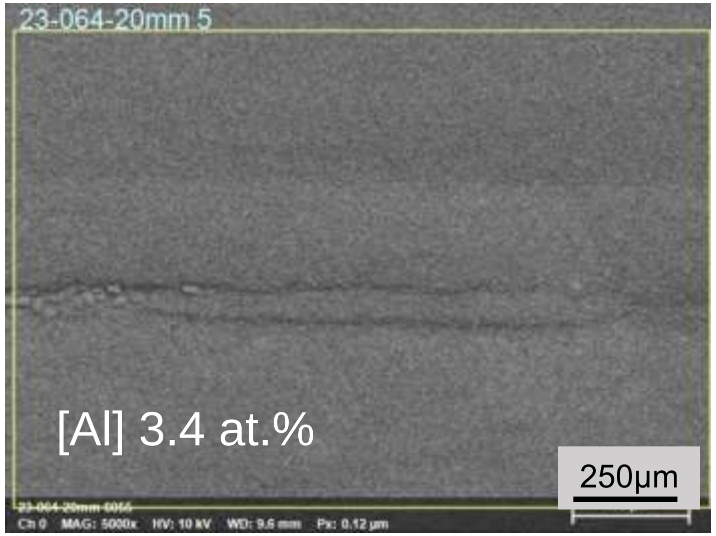
[Al] 4.5 at.%

Off-cut 0.5 deg



[Al] 3.4 at.%

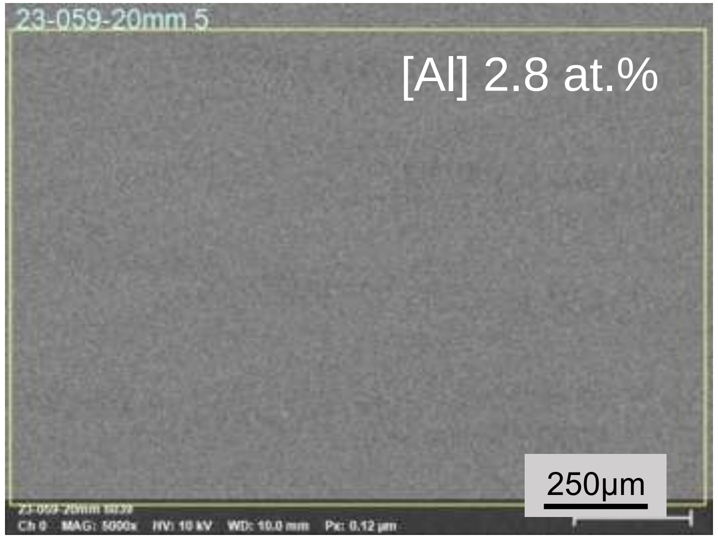
Off-cut 2.0 deg



[Al] 3.4 at.%

Off-cut 1.0 deg

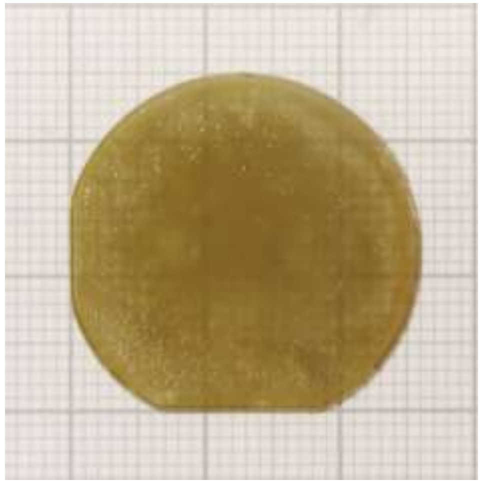
Off-cut 4.0 deg

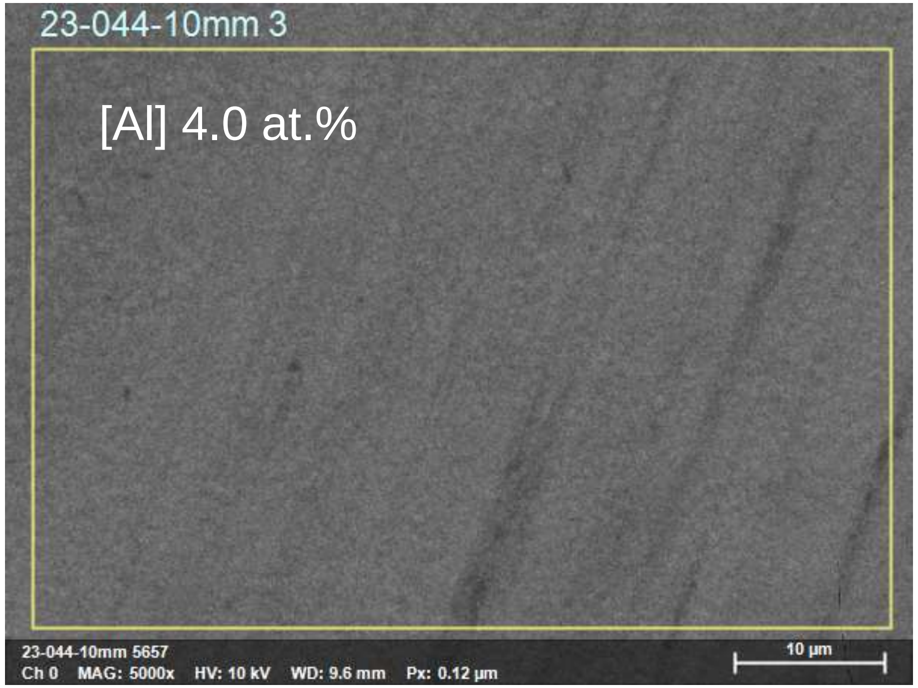


[Al] 2.8 at.%

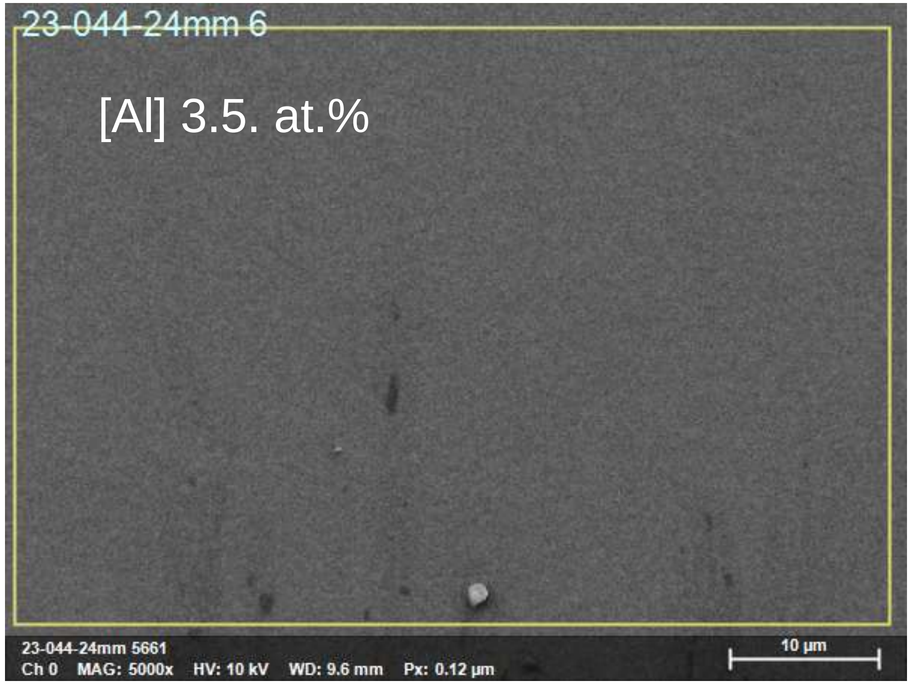
$$R = \frac{P^{\circ}_{AlCl_3}}{P^{\circ}_{AlCl_3} + P^{\circ}_{GaCl}}$$

Growth of AlGaN layer on native Am-GaN seed



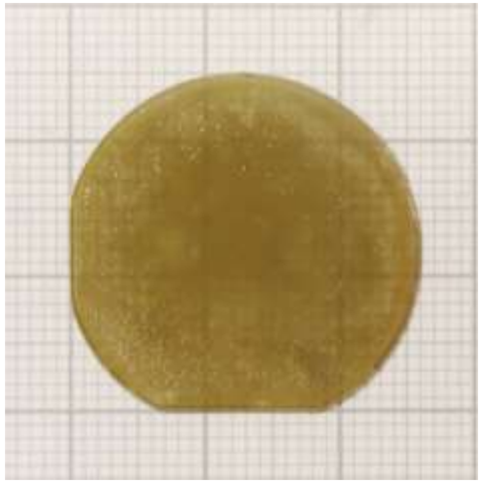


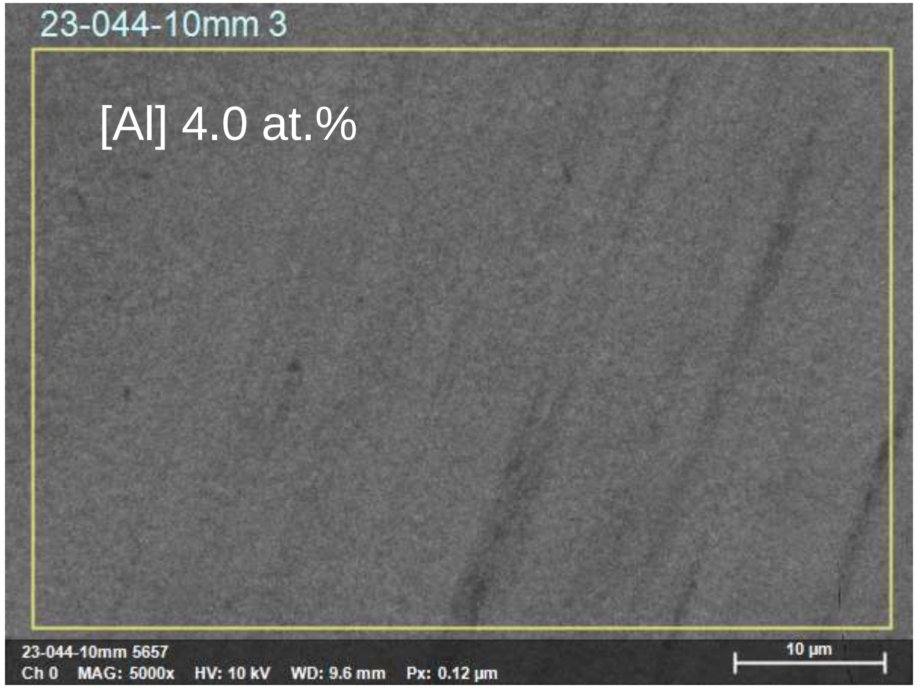
center



edge

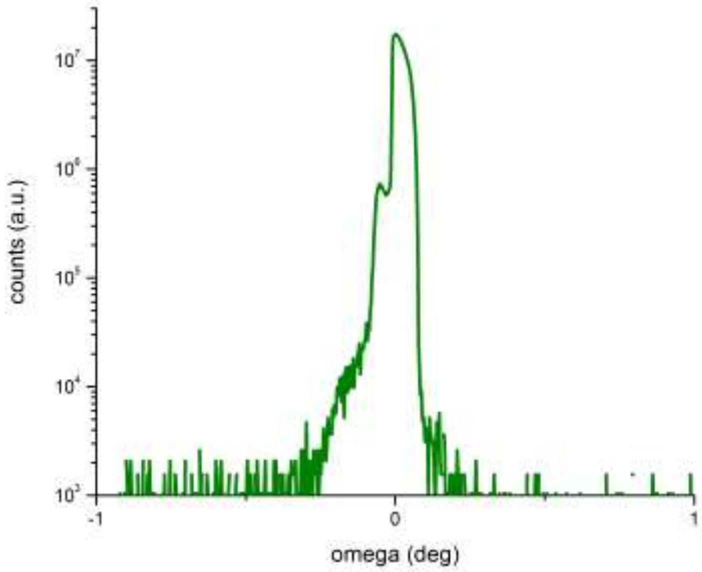
Growth of AlGaN layer on native Am-GaN seed





center

beam size: 1 mm x 10 mm



XRD for 002 reflection

Summary

- Crystallization of AlGaN with low concentration of Al is possible at the optimum temperature for GaN growth - 1050°C
- Under such conditions, high supersaturation for Al - favoring nucleation - can be effectively lowered by lowering the ammonia flow without GaN decomposition.
- Reducing the supersaturation for Al is also favored by reducing the pressure in the reactor.
- Reducing the pressure allows shifting the supersaturation for Al from the nucleation region to the crystal growth region.
- Increasing off-cut results in a radical improvement in growth morphology

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Acknowledgments

Sonata Project – National Science Center
2020/39/D/ST5/01611

Lider Project - National Centre for Research and Development
LIDER/23/0129/L-10/18/NCBR/2019

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Thank you for your attention

Appendix