Growth and characterization of indium containing monolayers in GaN

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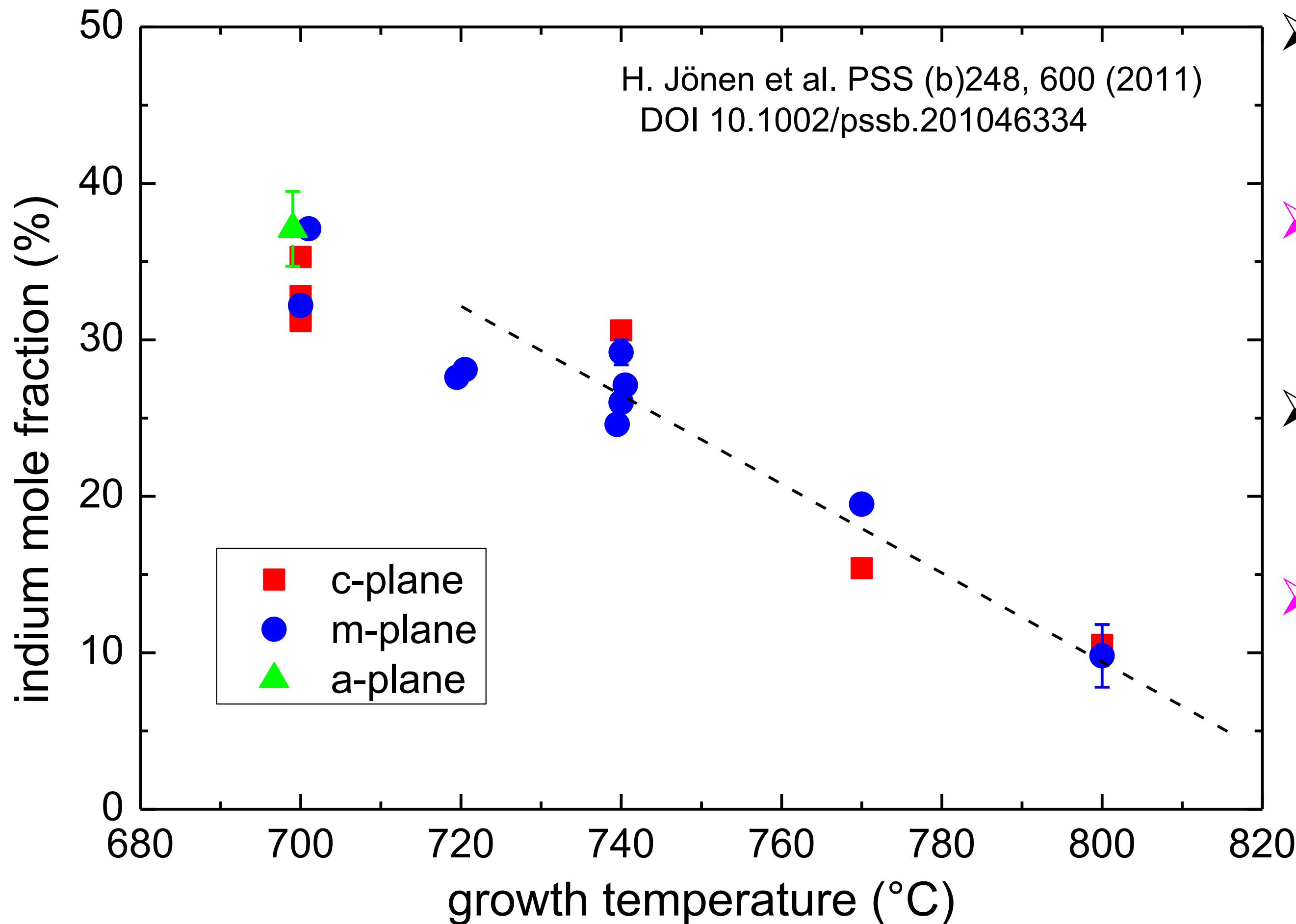
Introduction

- InGaN/GaN MQW optoelectronic devices visible/near UV.
- Indium incorporation not well understood, conflicting results (see discussion in R. Bhat, G. M. Guryanov JCG433, 7 (2016)): surface orientation, offcut angle, limited ammonia dissociation, role of hydrogen...
- Long wavelengths emitter, solar cell: need high indium concentrations.
- Aim: better understanding and control of the incorporation process.

Introduction: In-concentration QWs



- Similar for c- and m-plane of wurtzite structure
- Saturates around 30-33%, caused by strain?
- Stable phase 33% - S. Lee et al. PRB **90**, 245301 (2014).
- Indium desorption dominates at high T_G

What happens at T_G <740°C, saturation?

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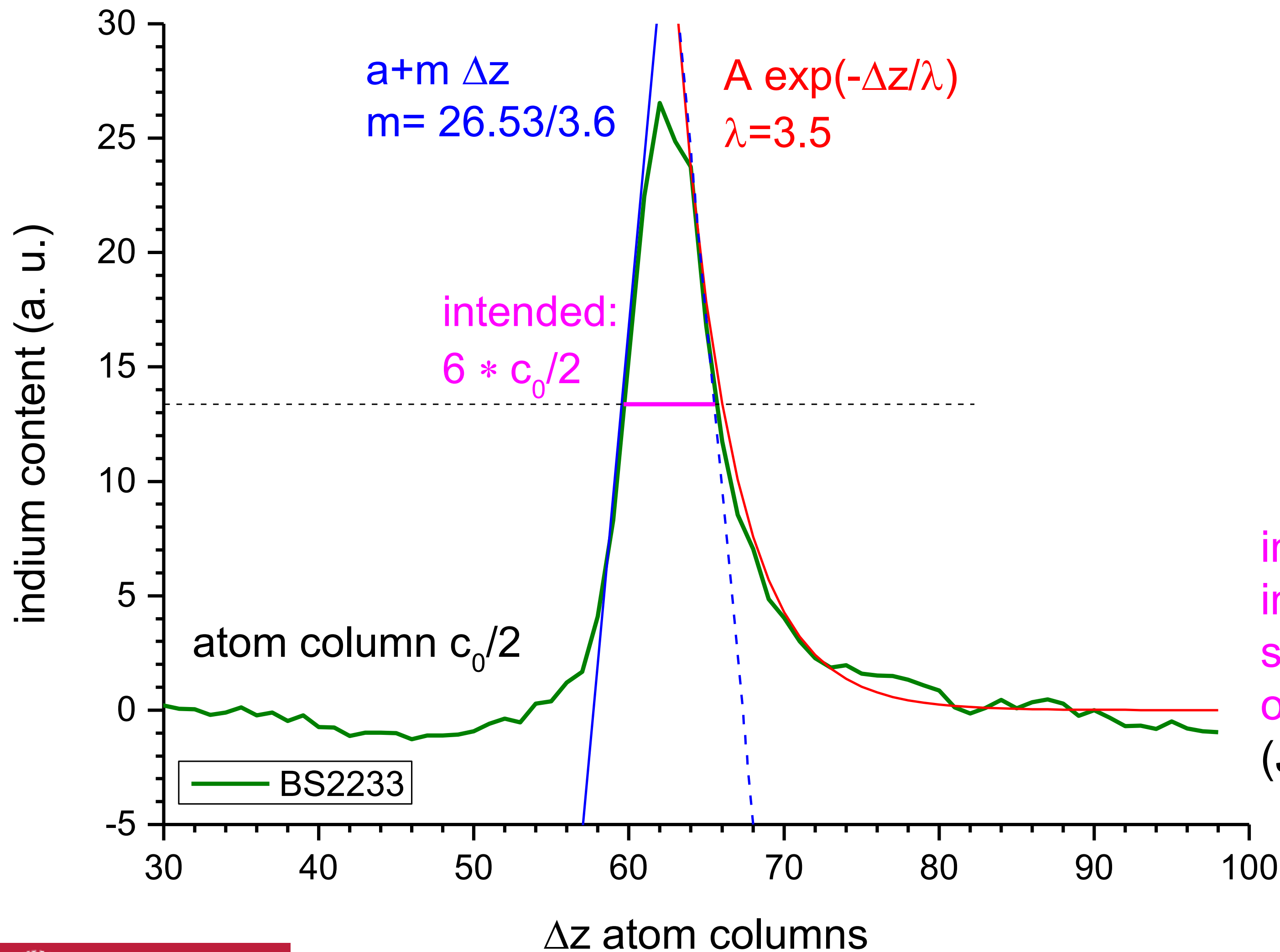
Introduction: STEM profile InGaN QW



indium incorporated into barriers ⇒ non sharp upper interface or step-like profile (JCG **414**, 49 (2015)).

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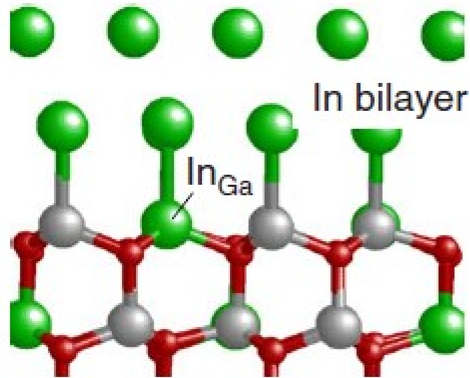
Introduction: Model by theory - adlayer



Structure of InGaN c-plane surface: InGa+In bilayer, In atoms green, Ga gray, N red, H white.
(J. E. Northrup: PRB 79, 041306 (R)).

Liquid-like layer on surface proposed by theory (C. G. Van de Walle, J. Neugebauer, J. Northrup):

- Ga or In incorporation from this adlayer.
- Effect surface strain lowered by In atom on top of incorporated In atom.
- C-plane and m-plane similar, (11-22) favored.

Introduction: Indium incorporation

- At high T_G adlayer is incomplete
- At low T_G or high TMI doses adlayer fully developed, excess indium may cluster
- Exchange indium adlayer - clusters
- Indium incorporated into barriers – clusters as indium source when TMI switched off

(see JCG **414**, 49 (2015), JCG **464**, 112 (2017))

Better control by pulsed growth, single layers?

Experiment

Substrates:

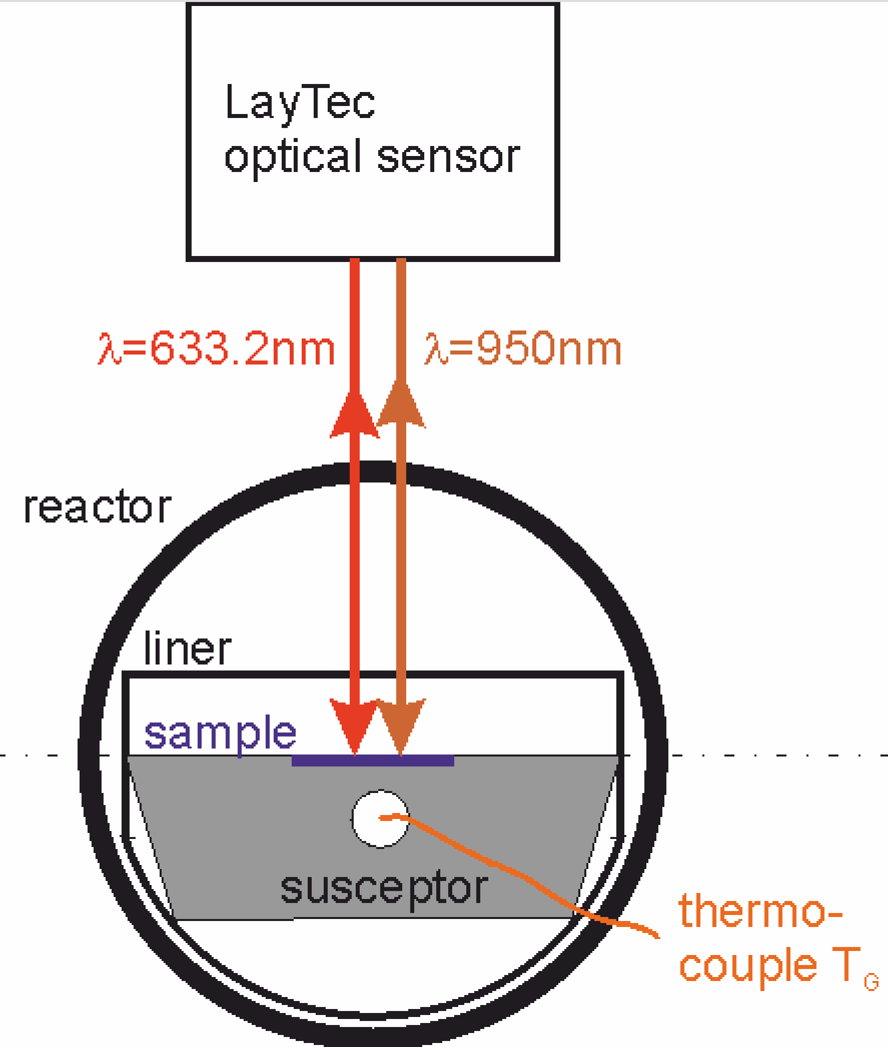
- Sapphire, (0001) on-axis, 330µm, single-side polished.

Growth:

- Aixtron AIX200RF
- InGaN: N_2 carrier gas, 200mbar, 6slpm, TEG 8sccm (TMG), TMI 40sccm (III/III 3.8) or 47sccm (III/III 4.5), NH_3 (124 mmol/min), V/III > 11000

Characterization:

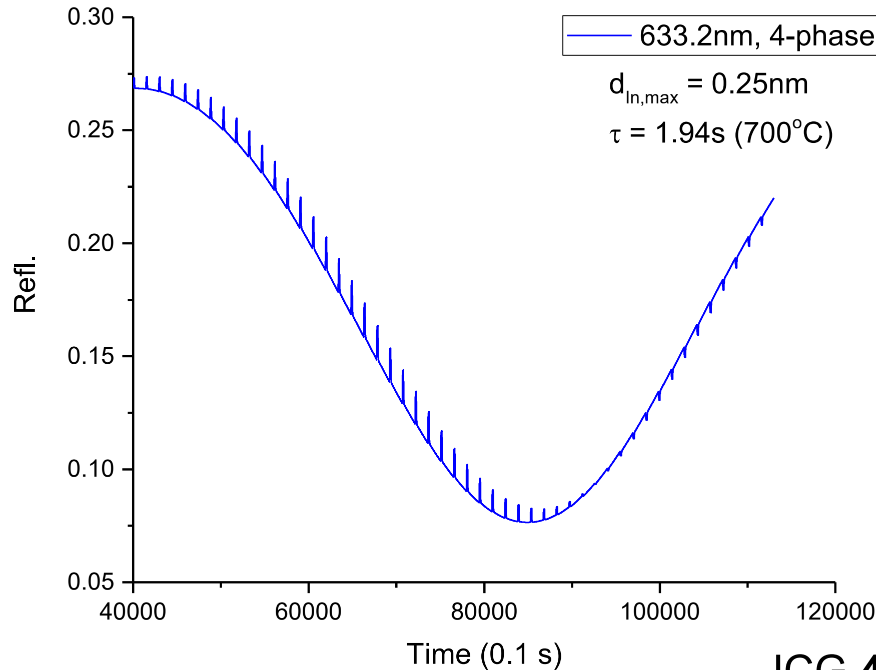
- PL 4K -- 300K: low exc. Ar 380nm, 365nm, 350nm, 334nm
- Raman (TU Chemnitz),
- HRXRD,
- REM, AFM, STEM (Univ. Bremen).

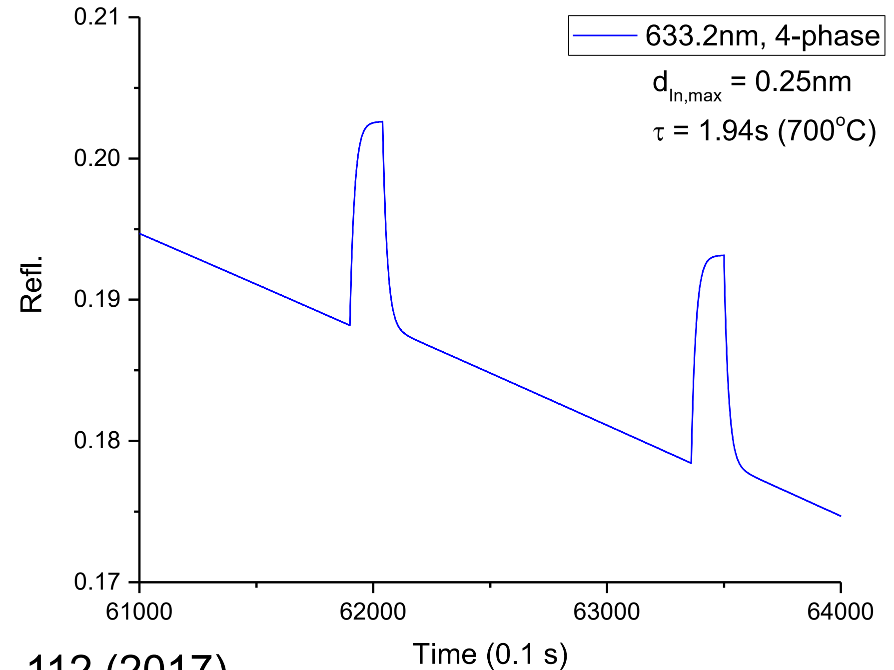


Expected transient

4-phase model:

ambient (0) – layer (1) – layer (2) – substrate (3)
ambient – indium – GaN /InGaN – sapphire

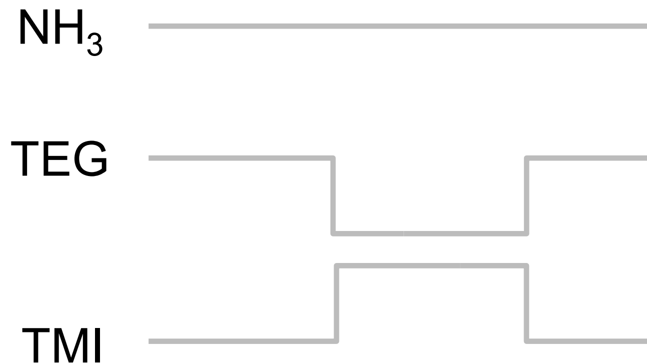




JCG **464**, 112 (2017)

- GaN – indium on surface by TMI puls – GaN
- Even a thin layer of indium should be measurable.

TMI pulses - GaN

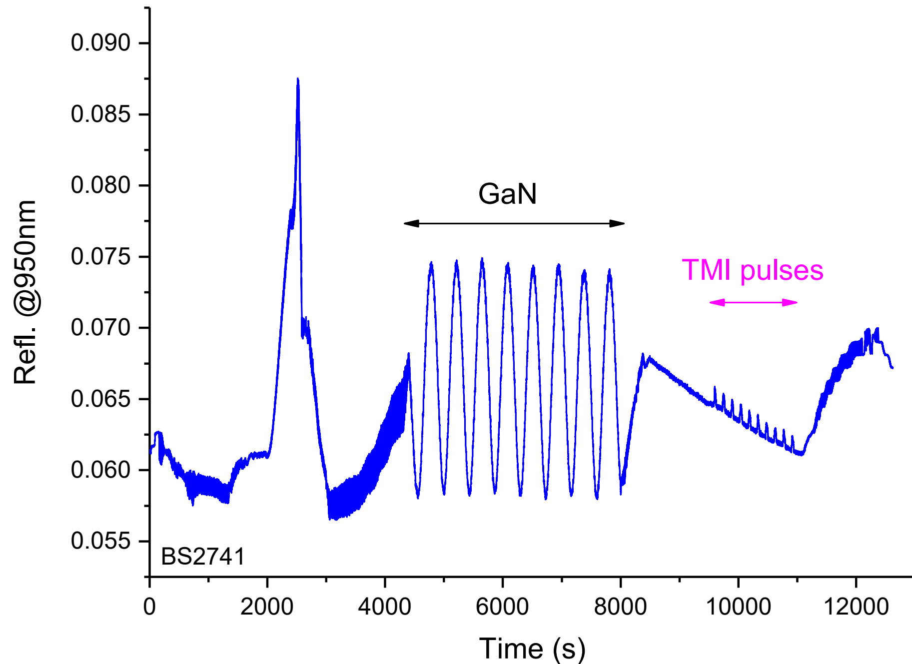


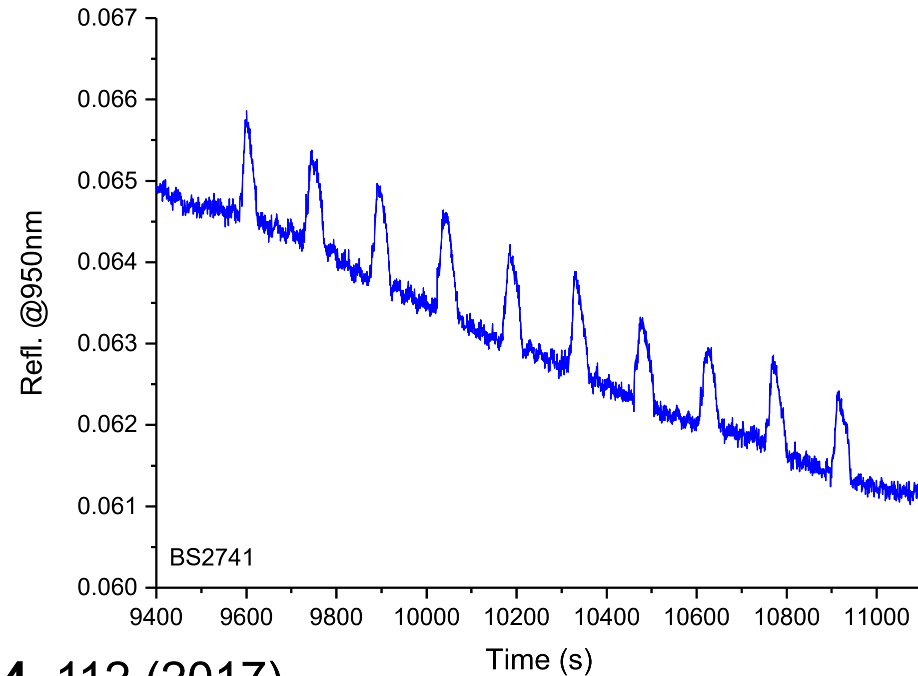
- TMI and TEG pulsed, ammonia always on.
- Intended (from QW growth): GaN 1.83nm (132s GaN), 28% In in $c_{0/2} \rightarrow x_{In,eff} = 3.86\%$
- pulsed mode: $x_{In,eff}$ smaller than expected, nonlinear for long pulses.

Here: 670°C - 770°C, and 6, 10 x (9-18s TMI, 0s/10s delay, 132s/330s TEG)

Measured reflection transients TMI pulses

670°C, 10 x (14s TMI, 132s TEG) ,TMI/TEG = 40sccm/8sccm





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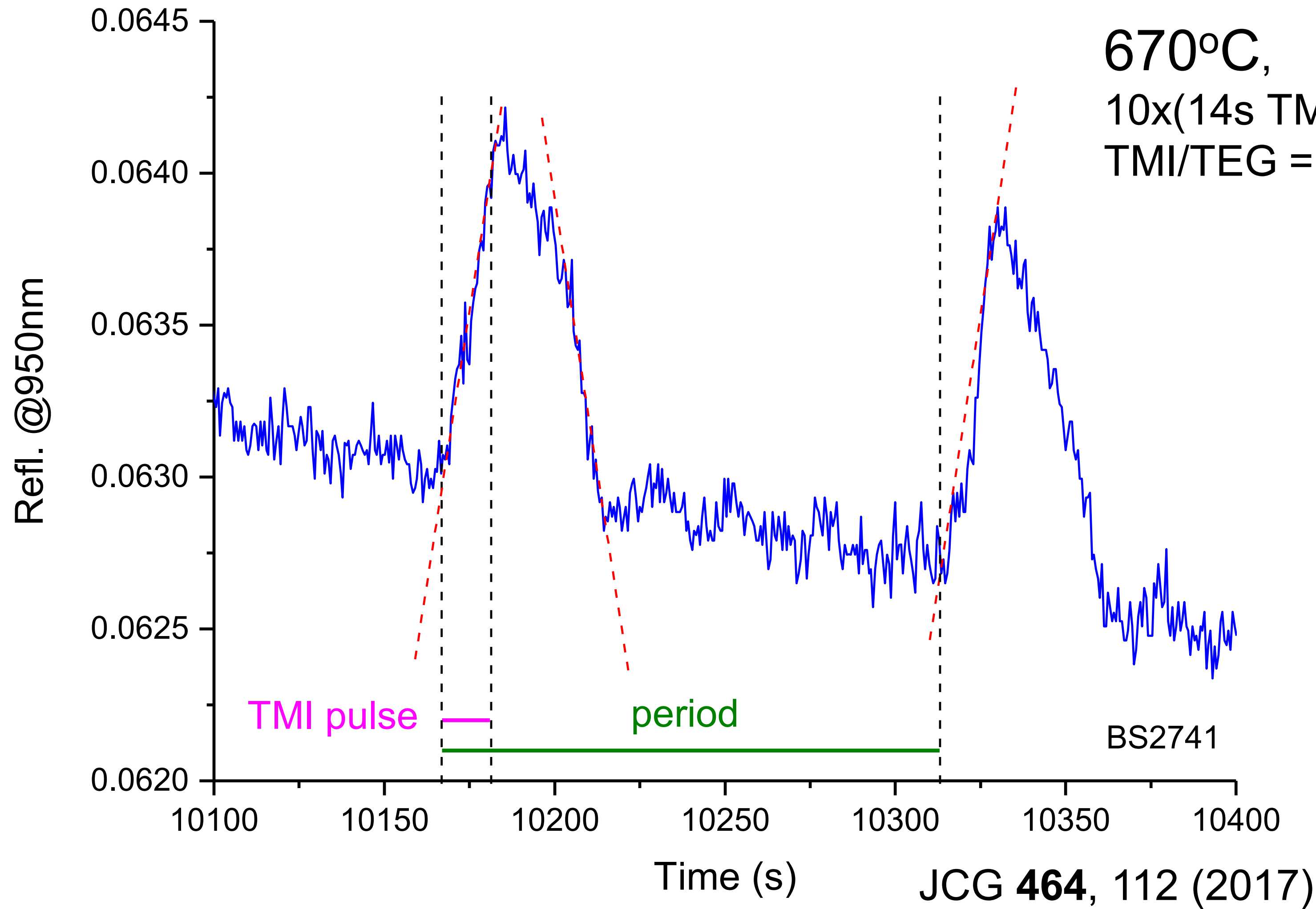
- Pulses clearly visible

Measured reflection transients TMI pulses



670°C,
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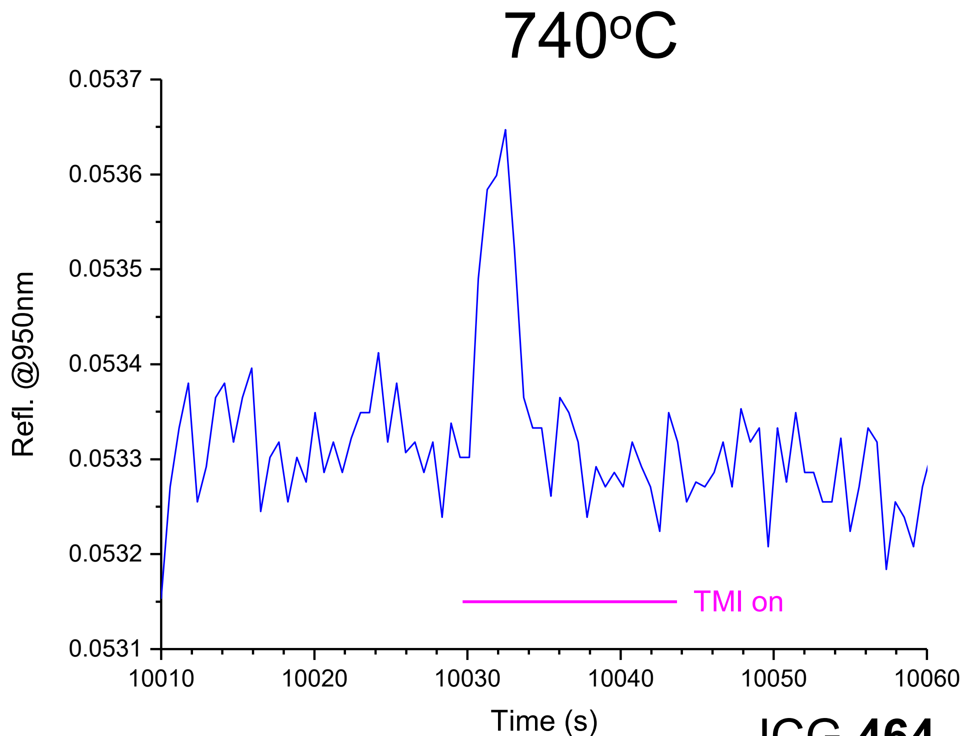
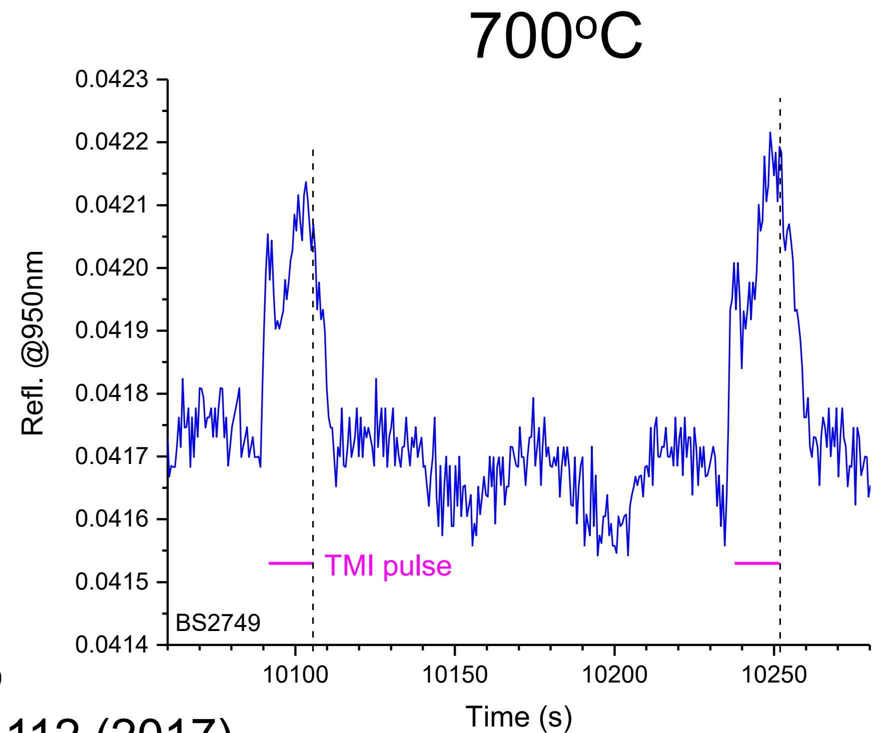
Measured reflection transients TMI pulses





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- A second loss mechanism: indium incorporation?
  → delayed incorporation

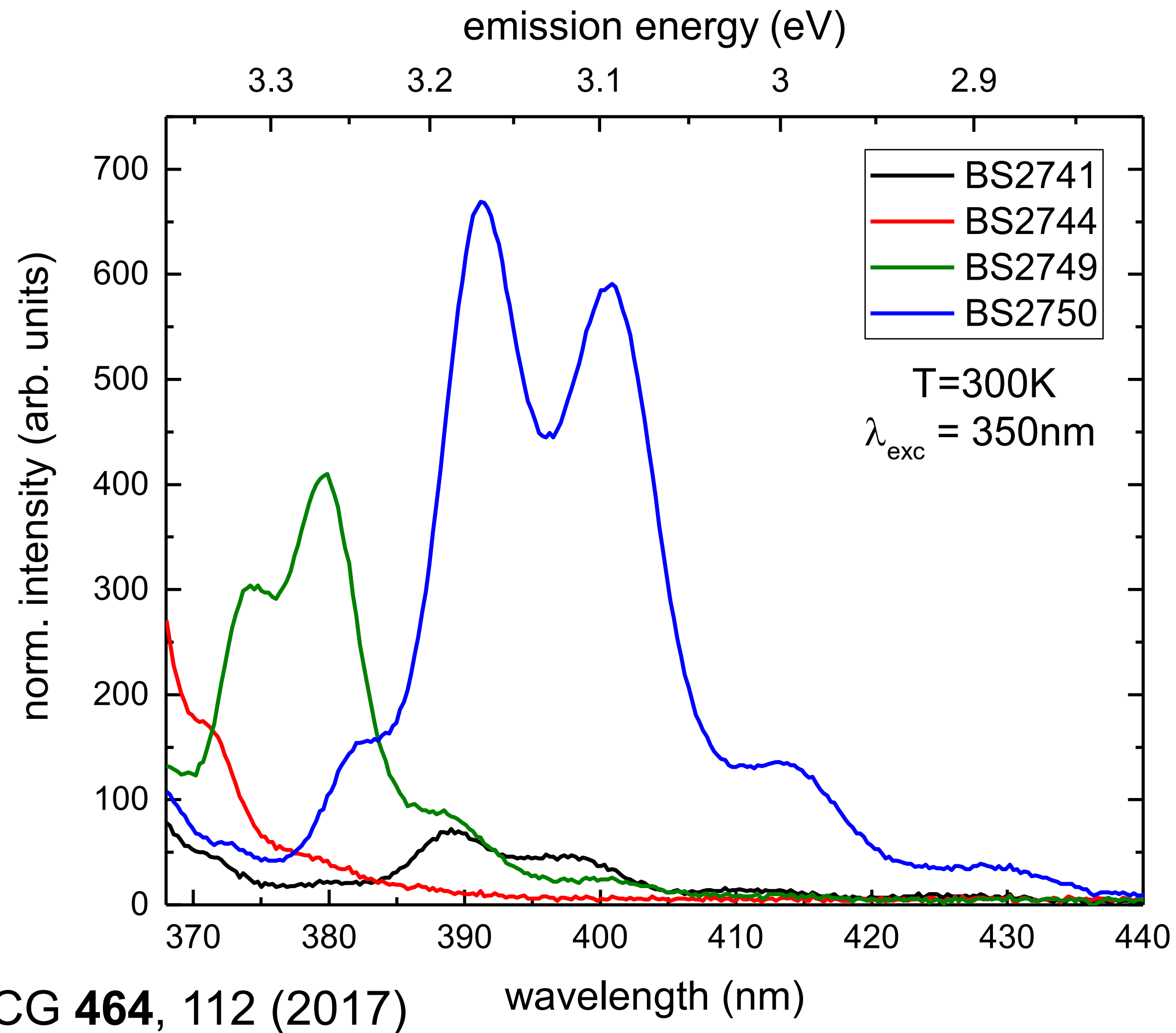
1. Initial phase
2. incorporation?
3. more indium supplied?
4. desorption

TMI pulses – PL at RT

HRXRD

sample	T_G	$x_{In,eff}$
BS2744	740°C	2.65%
BS2749	700°C	2.75%
BS2741	670°C	3.60%
BS2750 (delay)	700°C	4.23%

designed: $x_{In,eff}$ = 3.86%



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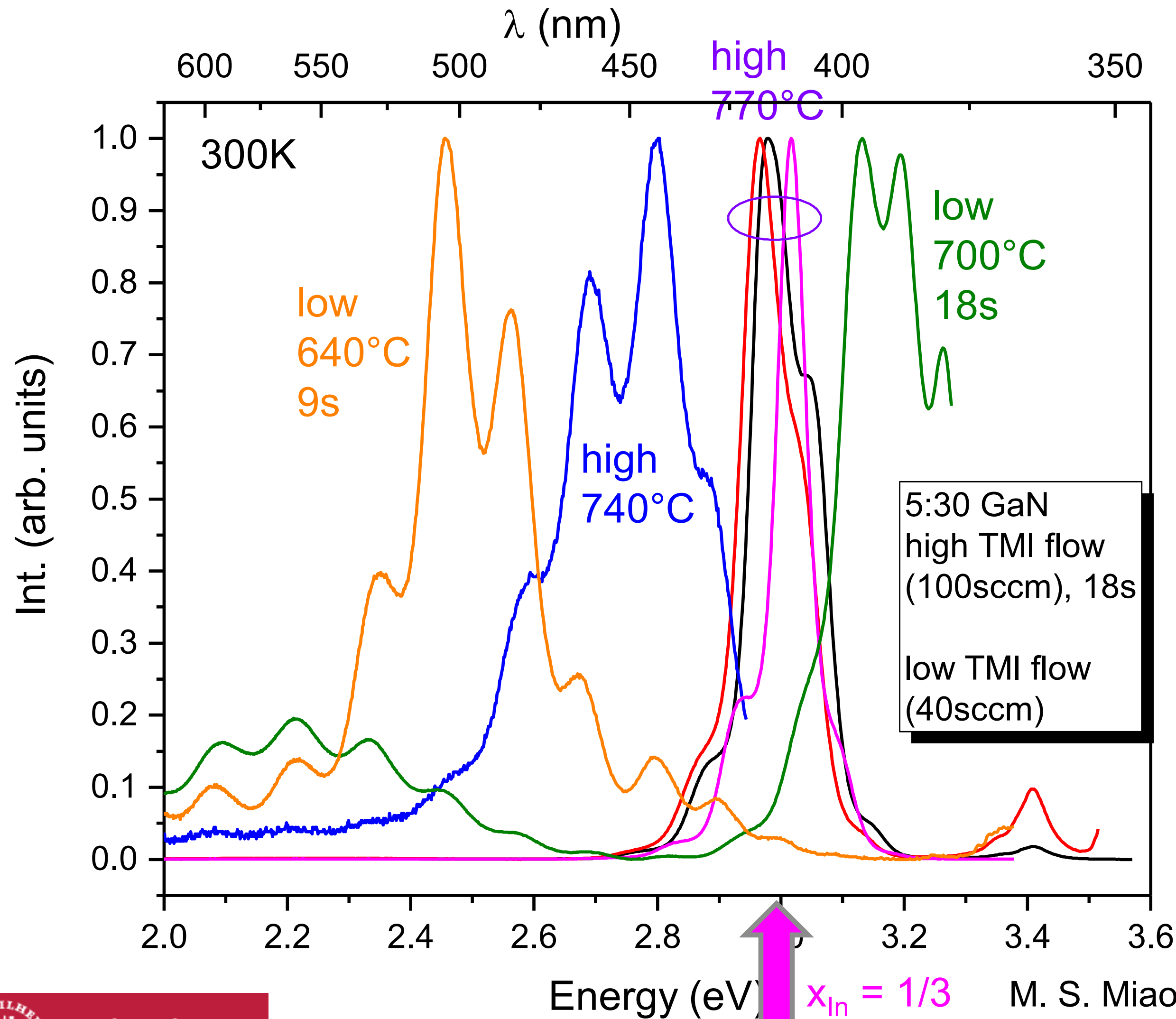
TMI pulses – PL at RT



$x_{In,eff}$ Exp.	$x_{In,eff}$ XRD
-	7.16%
-	7.58%
-	7.99%
-	6.22%
2.02%	1.51%
-	3.47%

M. S. Miao et al. APL **102**, 102103 (2013)

Raman spectra of InGaN

Literature data from S. Hernández et al.
JAP 98, 013511 (2005) show shift of A_1 and E_2 modes
with increasing indium content of $In_xGa_{(1-x)}N$

Raman spectra of InGaN

- Preliminary Raman data indicate that a third of the group-III layer is filled with indium (33% indium).
- PL and high $x_{In,eff}$ by HRXRD can only be understood if more than one group-III layer contains indium: we estimate approximately two layers.

Summary and Conclusions

- Results are in agreement with the adlayer model, adlayer stable.
- For low T_{QW} adlayer fills but competing process is metallic In cluster formation: Indium reservoir on surface, feeds adlayer → indium may incorporated into GaN barriers.
- Indium "loss": desorption + ?, delayed incorporation?
- "Saturation" of x_{In} observed in MOVPE (and MBE) → material property (not: gas phase, chemistry, V/III, NH_3 decomposition,…) → evidence for stable phase 1/3 coverage (in agreement with J. Neugebauer PRB**90**, 245301 (2014)).
- Monolayers are promising for active layers, pseudo alloys

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Thank you for the attention!