High-Pressure Chemical Vapor Deposition: An enabling technology for the fabrication of embedded indium-rich multiple heterostructures

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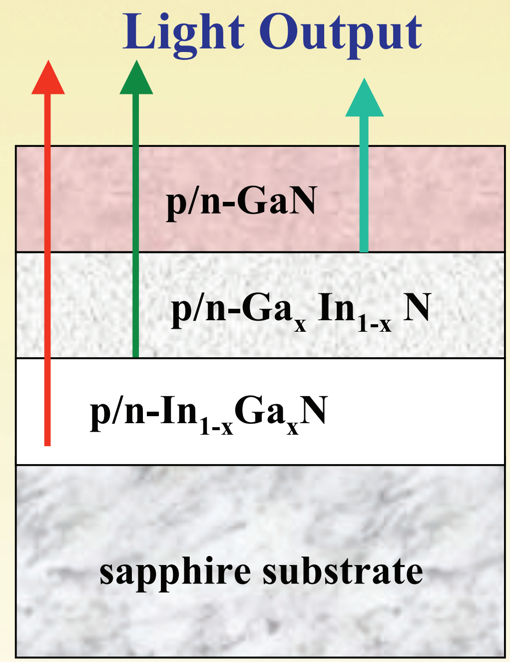
Need for embedded InN and indium rich group III-nitride alloys and heterostructures

- **High-efficient energy conversion systems**
  - Solid State Lightning (SSL)
  - frequency agile lasers in wide spectral range
  - monolithic integrated LED's / displays
  - Monolithic integrated dual UV/IR detectors



- High Speed Electronic Switching
- Monolithic integrated optoelectronics
- High Power Amplification
- High temperature device operation
- Room-temp. ferromagnetic group III-nitrides
- Terahertz device structures (emitters and detectors)

However, high quality InN and indium-rich alloys are difficult to fabricated at optimum processing conditions!

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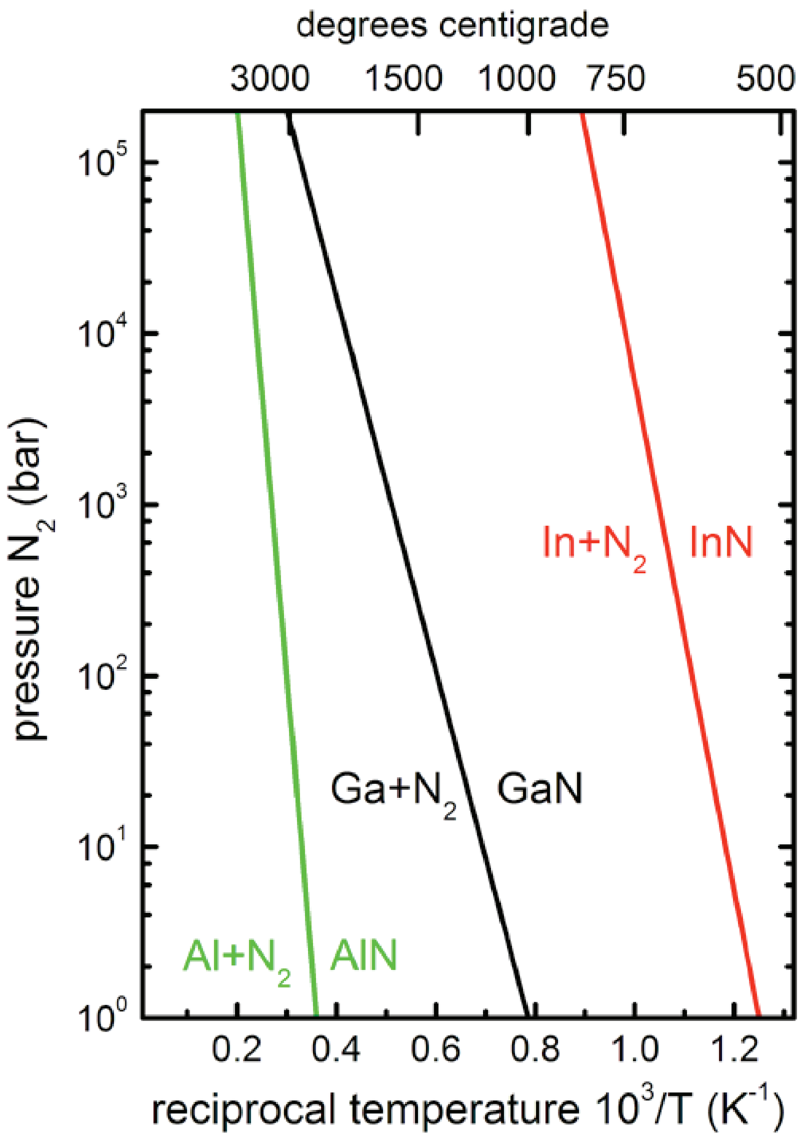
The need of high-pressure CVD for InN growth and the fabrication of embedded GaN/$In_{1-x}Ga_xN$/GaN structures



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Stabilization of InN under blanket of high pressure molecular nitrogen will enable processing at higher temperatures T than attainable at atmospheric and sub-atmospheric pressure.

p = 100 atm T = 900 - 1100 K

Potential gains of high T:

- *Improved nucleation kinetics*
- *Improved surface morphology*
- *Improved microstructure at growth temperature*
- ***Match of InN - GaN - AlN processing conditions***
  → control of decomposition kinetics

Drawback:

flow-kinetics can not be neglected!

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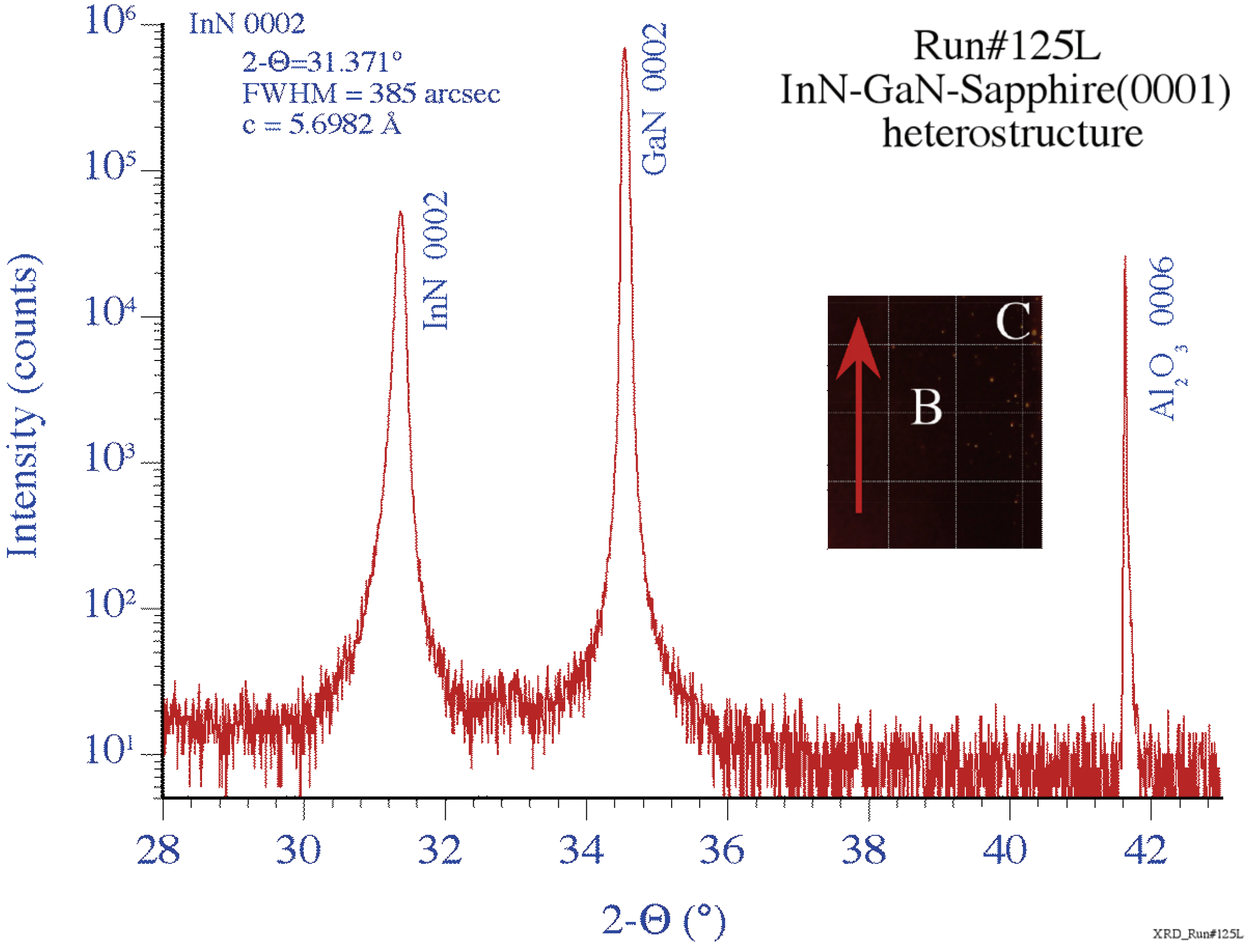
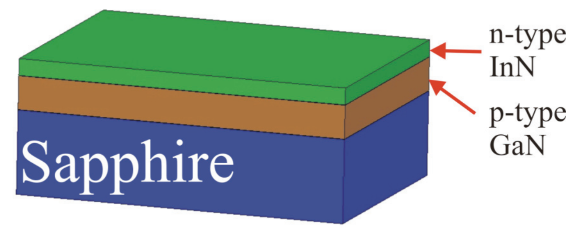
Does the HPCVD approach work? - Yes!





- **Single crystal InN succesfully deposited on GaN and sapphire epilayers**
- **FWHM of InN (0002) presently around 385 arc sec**
- **Carrier concentration in mid 10^{19} cm^{-3}**
- **Mobility around 600 $cm^2V^{-1}s^{-1}$**
- **Absorption edge above 1.5 eV**

Processing temperatures around 850 - 900 °C (@15 bar) enables the fabrication of thick GaAlN/$In_{1-x}Ga_xN$/GaAlN multiple heterostructures.

High-Pressure Chemical Vapor Deposition "HPCVD"

*Reactor Characteristics and Implementation**

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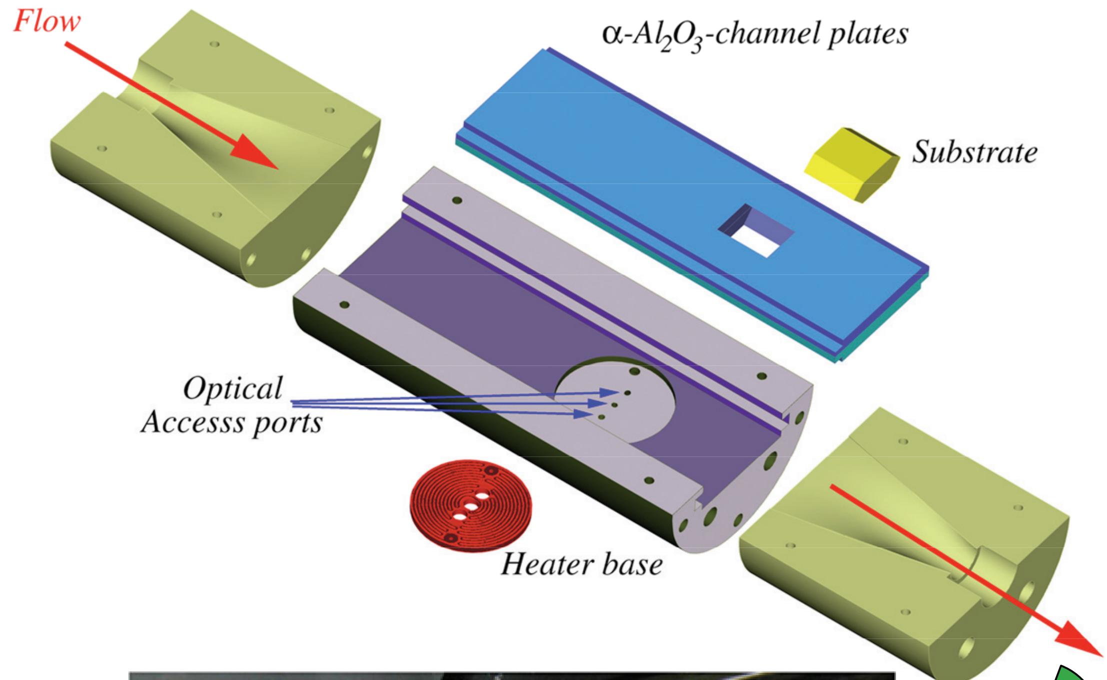
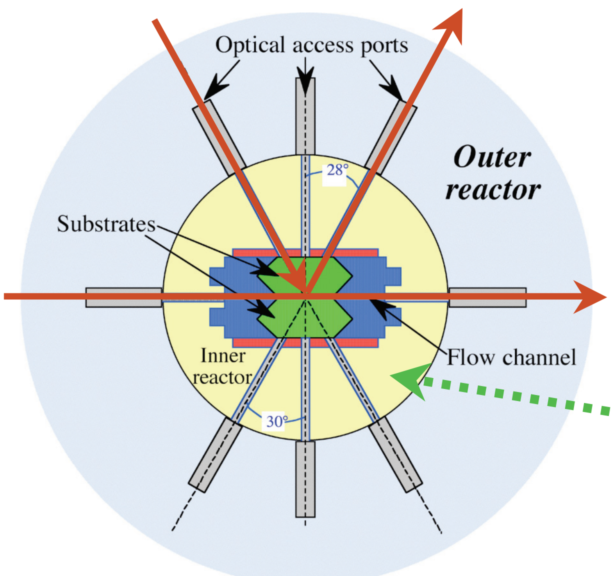
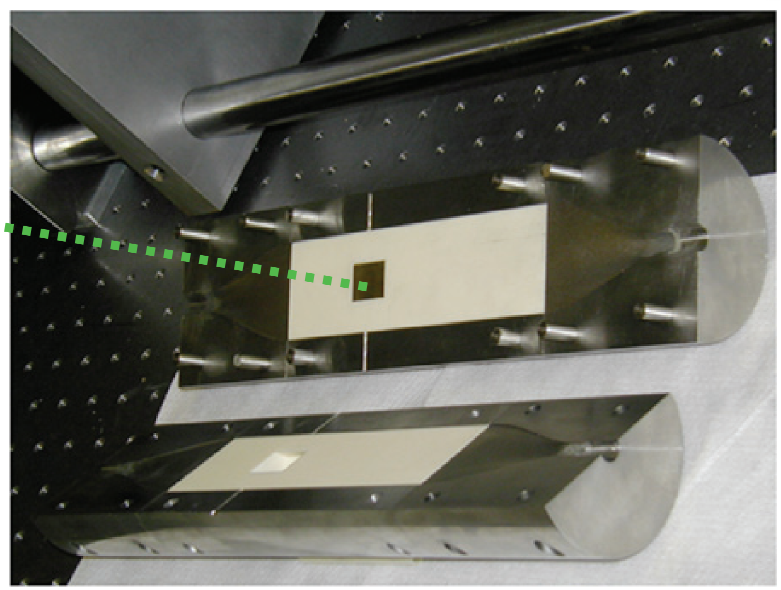
Design Criteria

- Maintenance of laminar gas flow
  (constant cross section)
- Symmetric substrates arrangements
- Access to Real Time monitoring







Real-Time Monitoring

- Characterization of gas phase reactions: **UVAS**
- Surface chemistry & kinetics **PARS**
- Gas flow kinetics, Surface roughness **LLS**

*In collaboration with Prof. K.J. Bachmann and T.H. Banks (NCSU)
Supported by: NASA (real-time diagnostics); AFOSR (reactor development)

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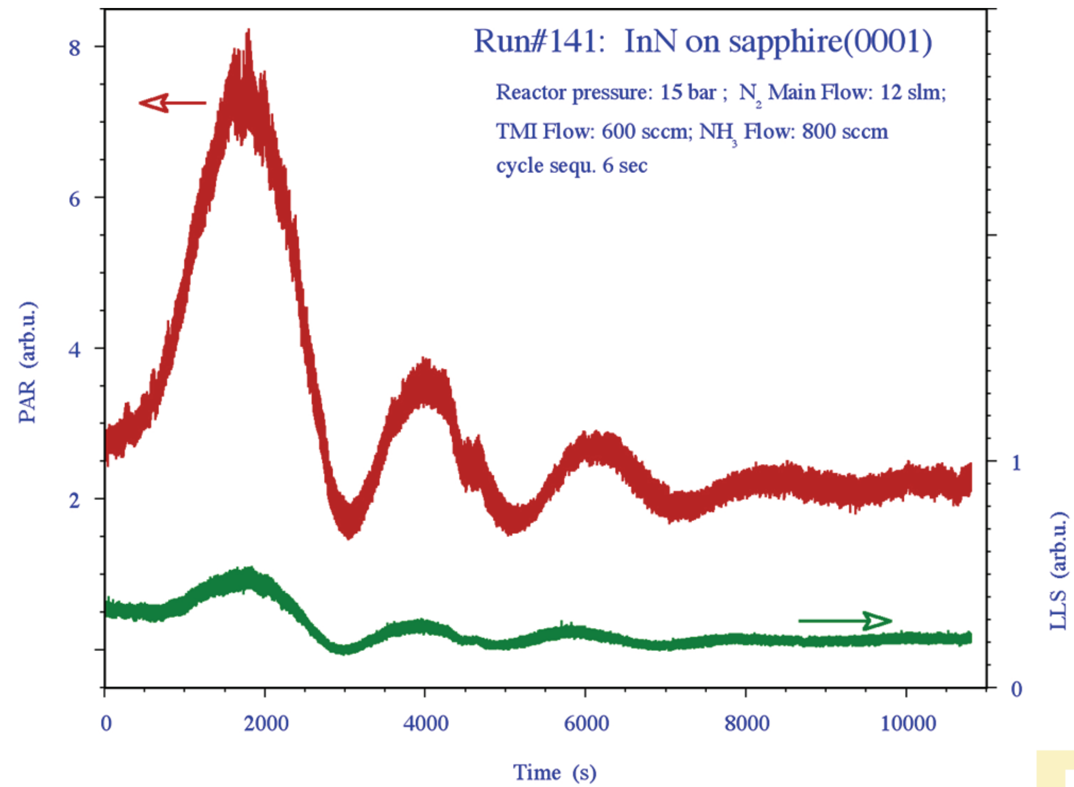
Real-time growth monitoring at elevated pressures



Principal Angle Reflectance Spectroscopy (PARS):

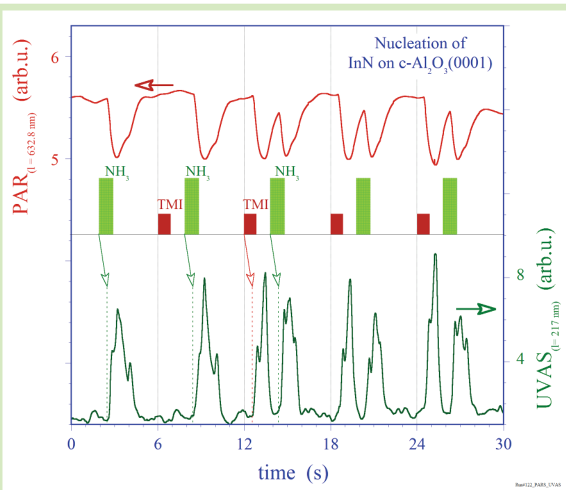
- *Extract and track average growth rate and dielectric properties of growing layer*
- *Characterize and monitor surface chemistry processes with sub-monolayer resolution*

Laser Light Scattering (LLS):

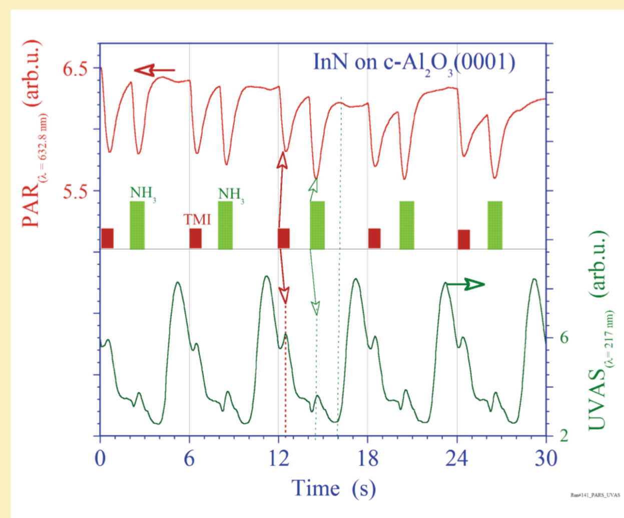
- *Monitor surface morphology / roughness*
- *Analyze reactor flow kinetics*

Ultra-violet Absorption Spectroscopy (UVAS):

- *Characterize precursor decomposition (e.g. NH_3 / TMI / TMG).*
- *Analyze/track precursor concentration/profile.*



UVAS and PAR traces during the heteroepitaxial InN nucleation stage



UVAS and PAR traces during steady-state growth conditions

InN film characterization: XRD

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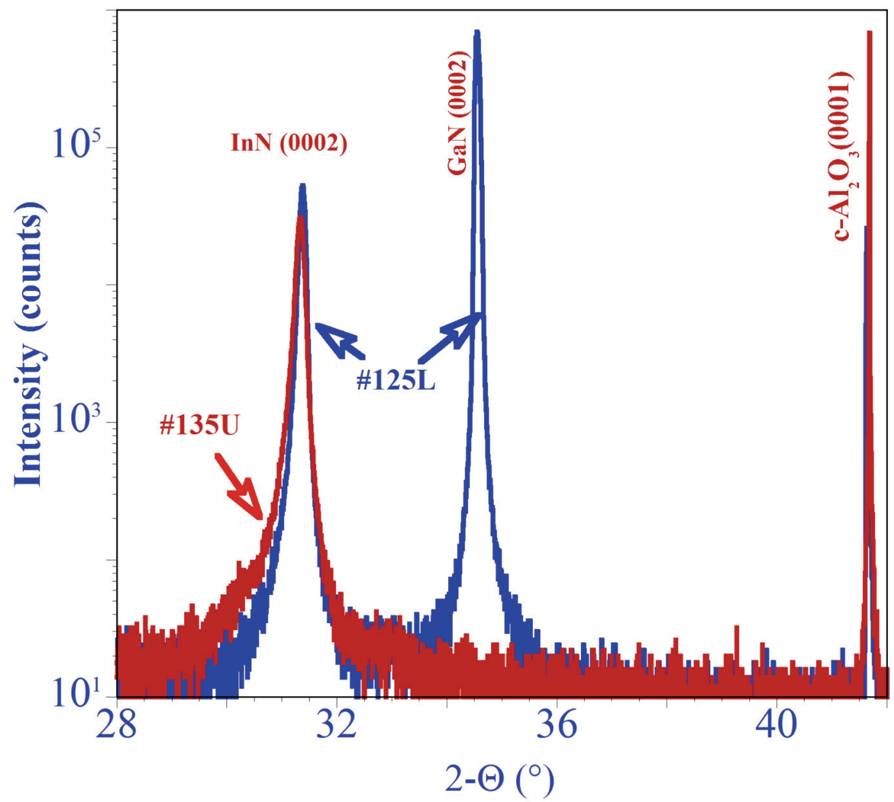
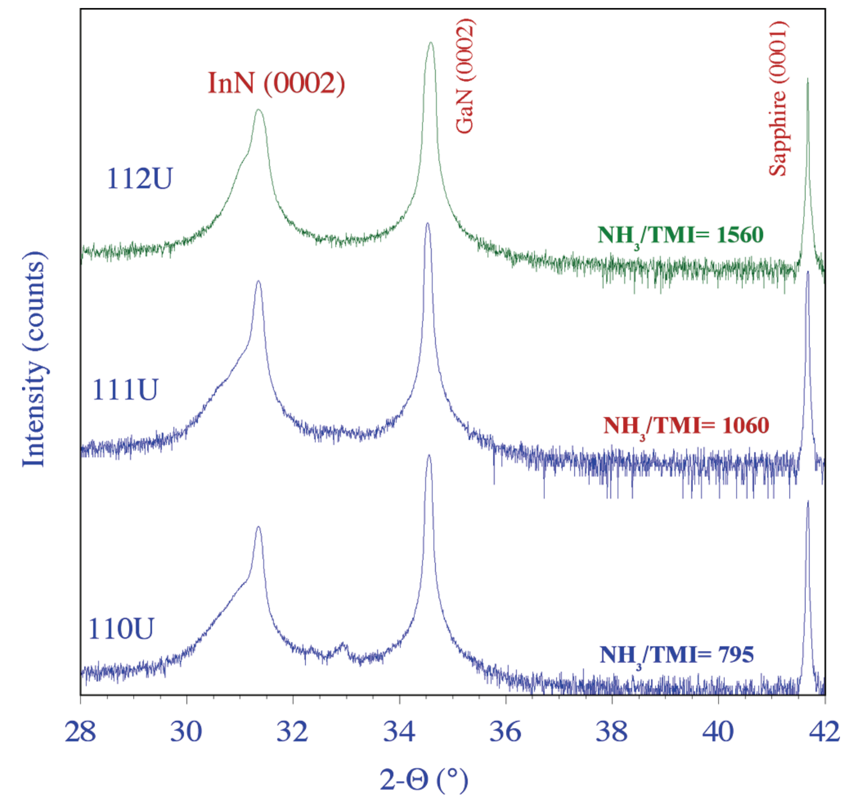


- Growth explored on sapphire and GaN/Sapphire
- For both templates, the growth of single phase InN (0002) epilayers achieved
- GaN/Sapphire FWHM ~ **385 arcsec**
- Sapphire FWHM ~ **492 arc sec**
- FWHM increases with lattice mismatch

Influence of Stoichiometry variations



- All samples were grown on GaN (0002)/ Sapphire(0001) templates
- The hexagonal phase InN (0002) peaks are centered around 2-Θ=31.3°
- FWHM values of InN(0002) increase with increasing NH_3/TMI ratio
- Lattice constants; c= 5.70 Å, a= 3.57 Å
- As the thickness of the InN film increases, InN (101) appear at ~ 33° - indicating a shift in processing window!

InN film characterization: Transmission

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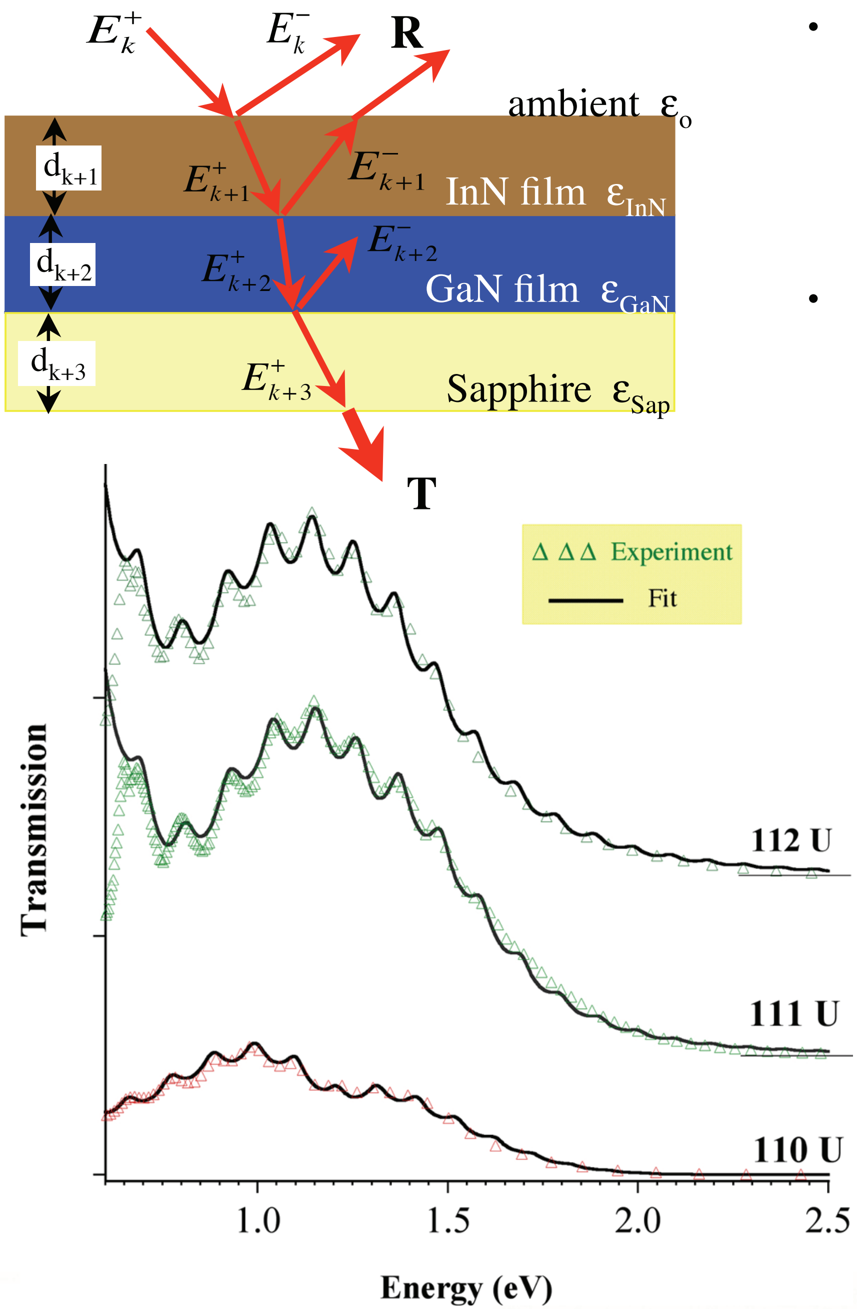

- Optical properties of InN (ε_{InN}, d_{InN}) are determined by fitting the transmittance of an "ambient/InN/GaN/Sapphire" stack using the dielectric functions of the group III-nitrides based on a "model dielectric function" (MDF) approach[1,2]

$$\varepsilon_{InN}(\hbar\omega) = \varepsilon_0(\hbar\omega) + \varepsilon_{0x}(\hbar\omega) + \varepsilon_1(\hbar\omega) + \varepsilon_{1x}(\hbar\omega) + \varepsilon_\infty$$

- A modified MDF model is used with two added oscillator at ≈0.8 and ≈ 1.2 eV, providing an excellent fit of the experimental transmission spectra:

$$\varepsilon_{InN'}(\hbar\omega) = \varepsilon_{InN}(\hbar\omega) + \sum_{n=1}^{2} \frac{S_{ab}}{(E_{ab}{}^2 - E^2) - iE\Gamma_{ab}}$$

S_{ab}: lorentzian oscillator strength,

E_{ab}: energetic position of absorption center,

Γ_{ab}: damping constant

- The optical constants, the refractive index and the extinction coefficient are determined from the transmission measurements for the films with different molar ratios

Sample	#110 U	#111 U	#112 U
d_{InN} (nm)	520	320	290
ε_∞	0.58	2.05	2.12
E_0 (eV)	1.92	1.58	1.48
S_{ab1}	0.46	0.37	0.37
E_{ab1}	0.64	0.8	0.8
Γ_{ab1}	0.57	0.34	0.35
S_{ab2}	0.075	0.01	-
E_{ab2}	1.2	1.3	-
Γ_{ab2}	0.22	0.1	-

1. A. B. Djusiric, and E. Herbert Li, J. Appl. Phys., **85**, 2848(1999).
2. T. Kawashima, H. Yoshikawa, S. Adachi, S. Fuke, and K. Ohtsuka, J. Appl. Phys., **82,** 3528(1997).

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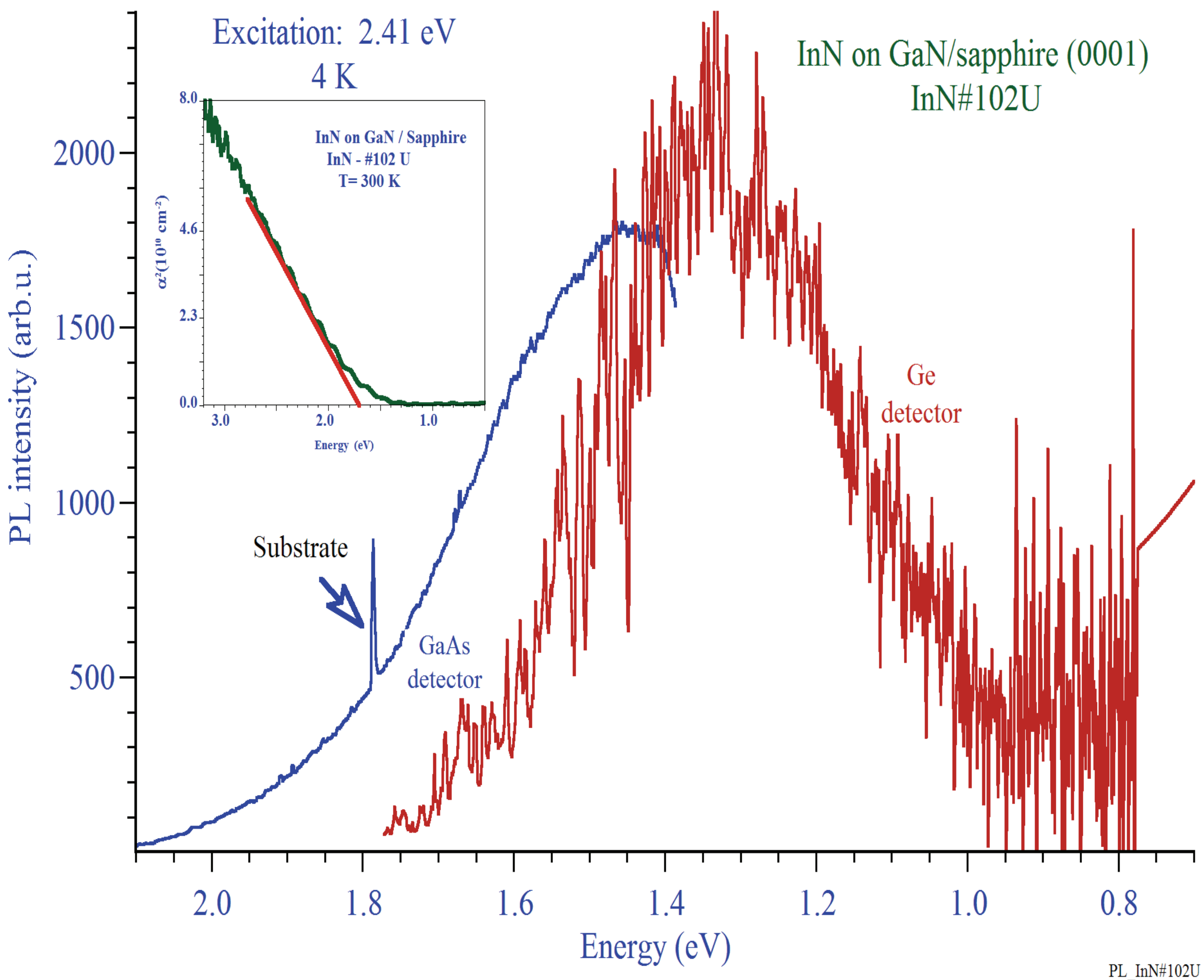
InN film characterization: ***Photoluminescence***



- PL spectra measured at 4 K
- broad PL features obtained due to the high defect concentration (> 10^{19} cm^{-3})
- The absorption coefficient (inset) was calculated by using the film thickness from the fitting of reflectance and transmission data
- The intercept of linear fits indicates absorption edge around 1.5 - 1.6 eV
- No PL detected below 1.0 eV

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The growth of InN by HPCVD

The need of high-pressure conditions

➢ ***Enabling the embedding of InN and indium-rich layers in wide-bandgap III-nitrides***

- growth temperatures at 1100K - 1200K for pressures 10-15 bar
- stabilization of volatile In-N-fragments under nitrogen blanket

➢ ***improved layer quality***

- higher mobility of growth surface fragments
- InGaN alloys by suppressing decomposition (phase segregation)

➢ ***engineered gas flow - and decomposition kinetics***

- onset of ammonia decomposition lowered at higher pressures
- MMIn and atomic In fragments dominant at pressures above 15 bar
- low ammonia : TMI ratio needed

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InN growth by HPCVD - *Open tasks*

- What is the origin of absorption centers appearing around 1.2-1.0 eV and 0.8-0.7 eV ?
  - Are the energy levels associated with conduction band states? If so, why would they vary with In:N ratio?
  - Could it be that the oxygen defect energy states lay in the conduction band of InN and couples with InN band states?

- Next steps
  - Reduction of external impurities
  - Understanding the influence of oxygen impurity
  - Optimize InN layer properties and assess turbulent flow regime at ***higher reactor pressures (20-100bar)***
  - Move to indium rich $In_{1-x}Ga_xN$ alloys & and embedded multiple $GaN/In_{1-x}Ga_xN/GaN$ device structures.