Growth issues and optical properties of nonpolar (Al,In,Ga)N films and quantum wells

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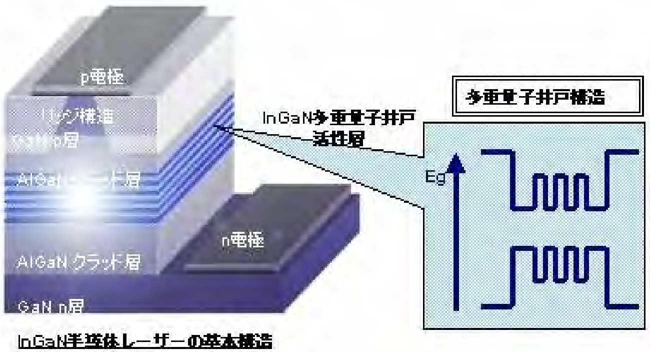
Optoelectronic devices

Wide bandgap semiconductor quantum nanostructures

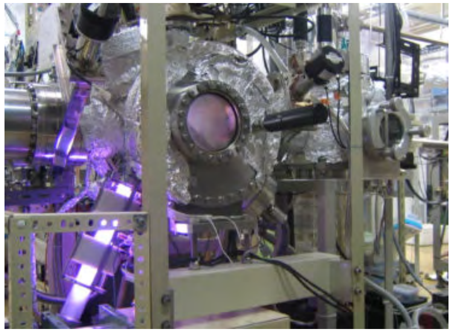
Epitaxial growth **GaN, ZnO** etc **Material Science**

New functional and planet conscious semiconductor optoelectronic devices, material growth, and material engineering

MOVPE

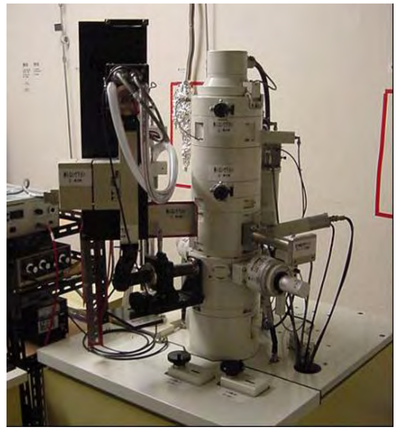


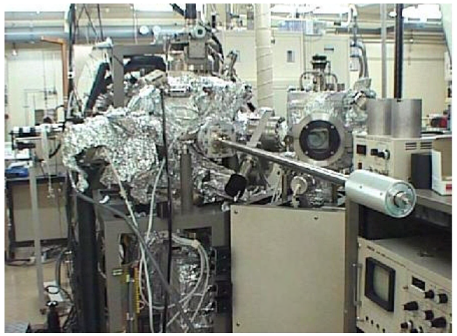
Quantum well laser structure



HWPSE for ZnO/MgZnO heterostructures

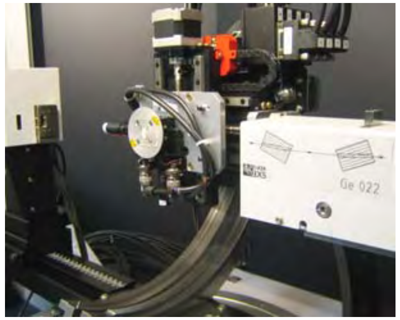
SEM / CL





NH_3-MBE

Femtosecond Ti:sapphire laser



HR XRD Bruker D8

東北大学 TOHOKU UNIVERSITY · IMRAM · 筑波大学 University of Tsukuba · Excellence in Electronics ROHM · 三菱化学 · UCSB · JST · ERATO · 窒化物光半導体のフロンティア −材料潜在能力の極限発現− · Optoelectronics Chichibu Lab 東北大学 秩父研究室

Contributors & Acknowledgments

Laboratory members

MOVPE Growth and Characterization:

T. Onuma, K. Hazu, T. Koyama, T. Koida, M. Kubota, L. Zhao, H. Yamaguchi

Samples

***m*-plane GaN substrate : K. Fujito, H. Namita, T. Nagao (Mitsubishi Chemical)**

Quantum wells and Devices :

S. Nakamura, S. P. DenBaars, J. S. Speck, U. K. Mishra, S. Keller, P. Fini, B. Haskell, A. Chakraborty, H. Masui (UCSB & ERATO-JST), H. Ohta, K. Okamoto, H. Takasu (RHOM)

Budgets

Nakamura Inhomogeneous Crystal Project-ERATO-JST, Grant-in-Aid for Scientific Research in Priority Areas No. 18069001 under MEXT, AOARD/AFOSR, ROHM, Mitsubishi Chemical, NGK etc.

Outline

1. Introduction

2. Issues on heteroepitaxial nonpolar (Al,In,Ga)N [UCSB samples]
   √ Planar growth and lateral epitaxial overgrowth of GaN
   √ Optical properties of InGaN/GaN and AlGaN/GaN quantum wells

3. Homoepitaxial nonpolar (In,Ga)N [Tohoku-films ROHM-devices]
   √ Low defect density freestanding (FS) *m*-plane GaN substrate
   √ GaN and InGaN growth by MOVPE
   √ Device performance Digest -- *m*-plane LEDs and LDs

4. Summary

papers available from http://www.tagen.tohoku.ac.jp/labo/chichibu/SHIGEFUSA/paper/GaN.html

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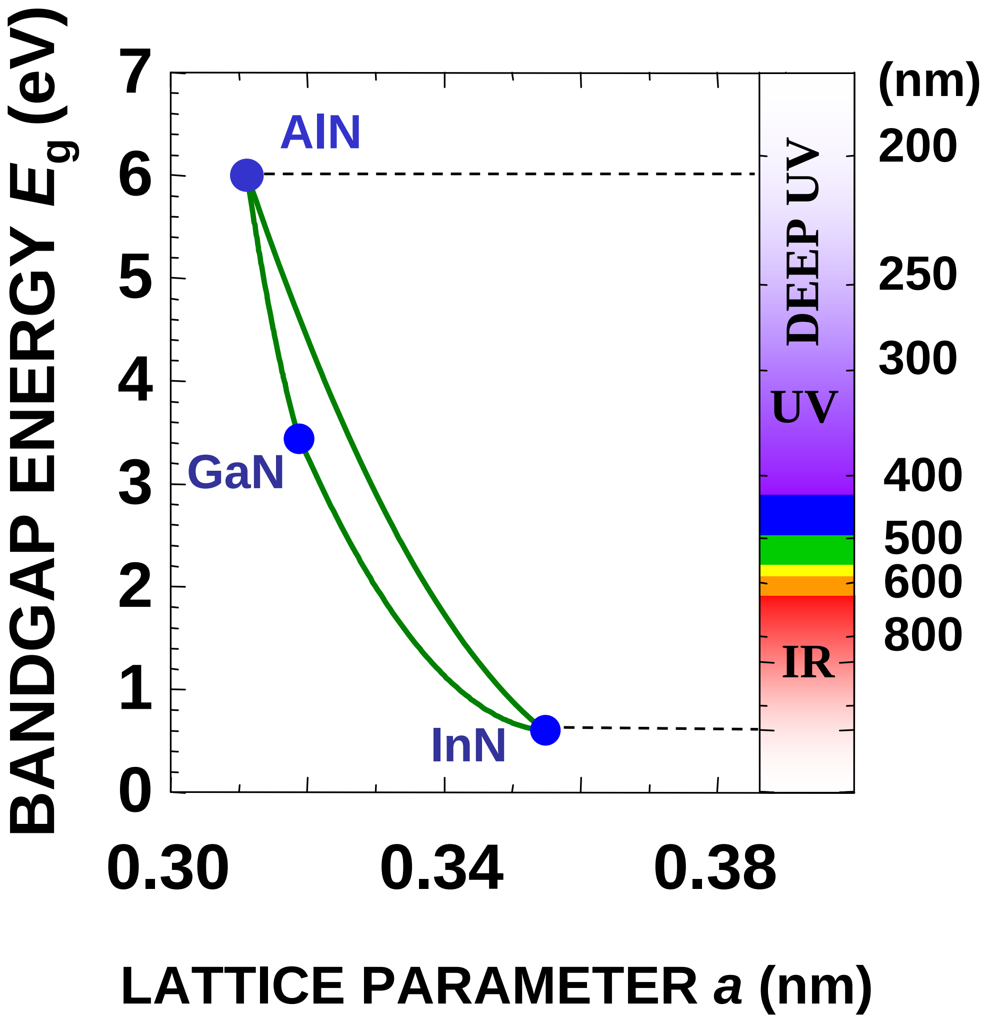
Group-III Nitride Semiconductors



Group-III Nitride Semiconductors
(Al, Ga, In)N

Wide Direct Bandgap range

- √ AlN 6.01 eV
- √ GaN 3.43 eV
- √ InN 0.67 eV

From deep UV to IR

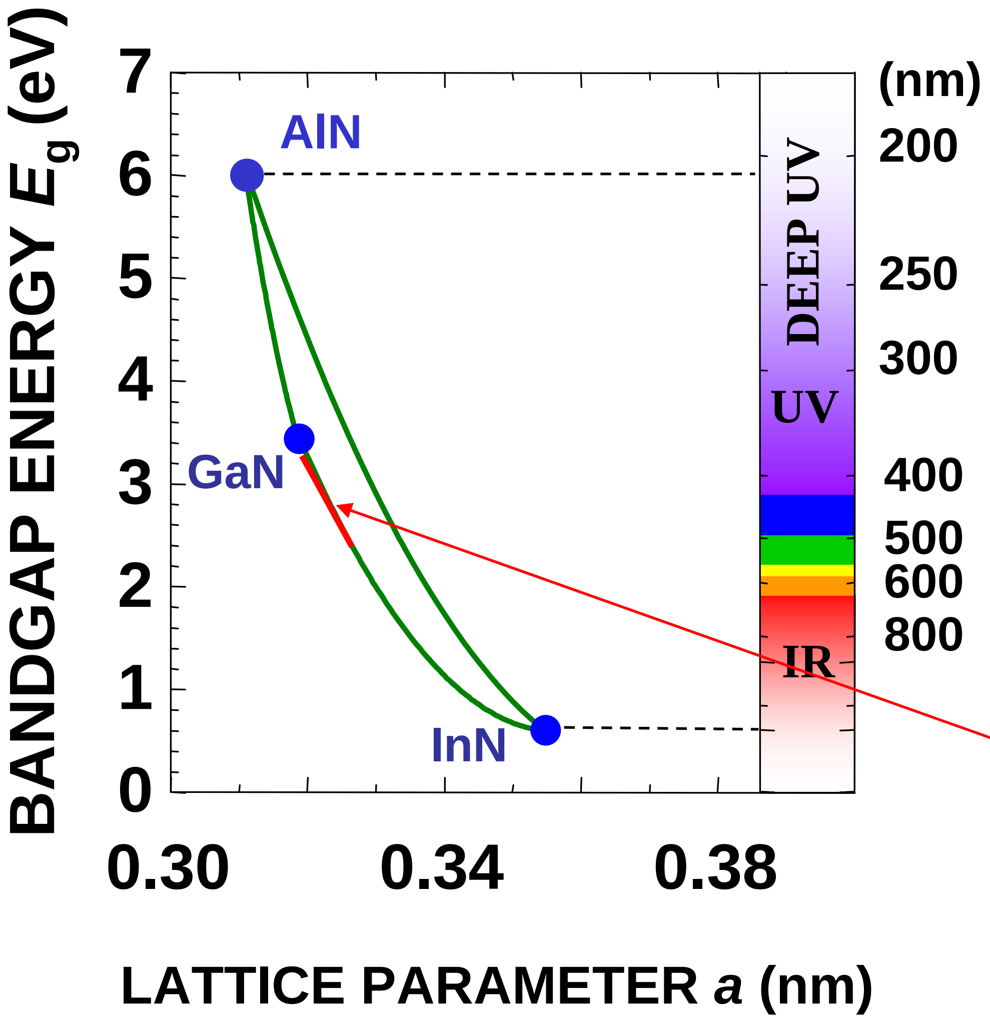
Hard material

High-power, high-frequency Electronic Devices

Blue, green, white LEDs and 400nm LDs

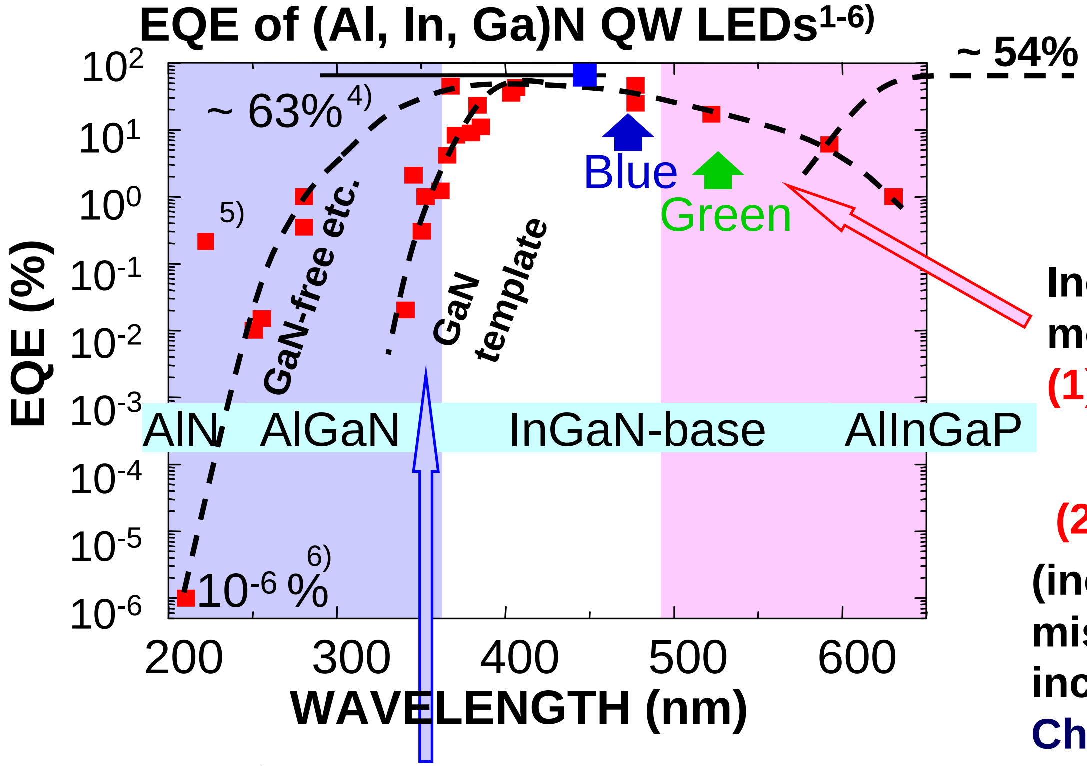
東北大学 TOHOKU UNIVERSITY
筑波大学 University of Tsukuba
Excellence in Electronics ROHM
三菱化学
UCSB
JST
ERATO
窒化物光半導体のフロンティア
−材料潜在能力の極限発現−
Optoelectronics Chichibu Lab
東北大学 秩父研究室

Group-III Nitride Semiconductors



Practical devices exclusively use c-plane (0001) InGaN quantum well active region

Issues on EQE vs wavelength (*c*-plane)



maximum value for AlInGaP LEDs

√ Substrate absorption
√ Increase in TDD and point defects

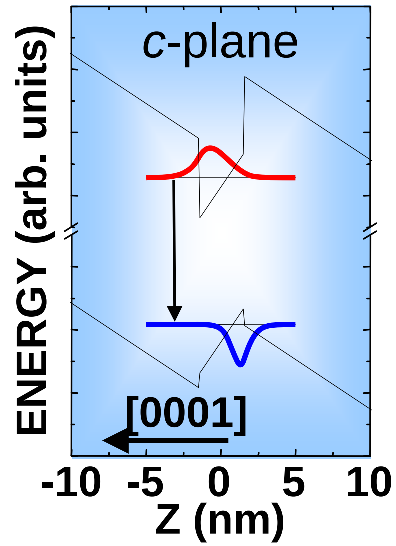
Increase in InN molar fraction

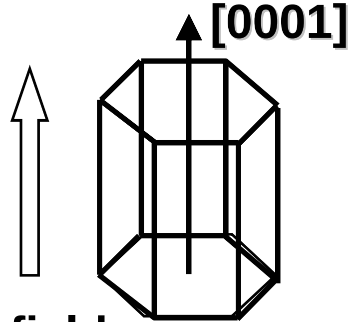
(1) point defects
(low *Tg* of InGaN)

(2) polarization effects

(increased lattice mismatch → increased QCSEs)

Chichibu *et al.* APL 69, 4188 (1996).
Takeuchi et al. JJAP 36, L382 (1997).





polarization fields

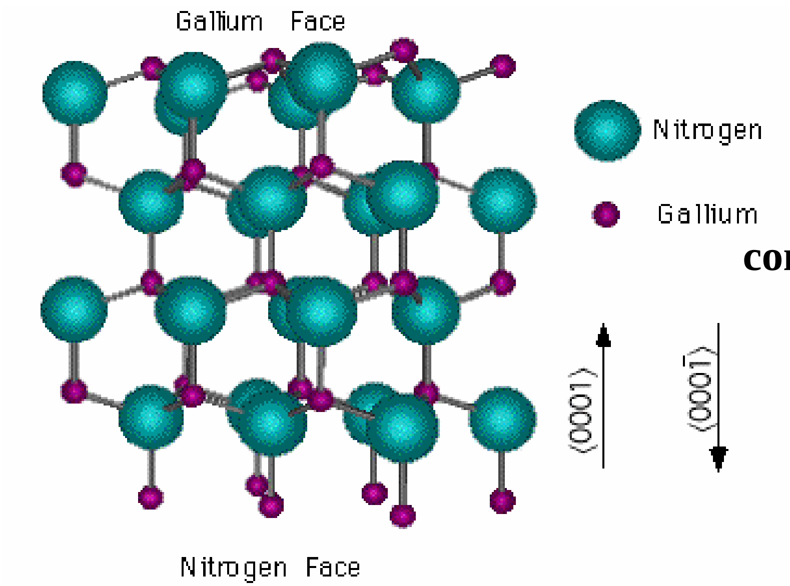
1) Khan *et al*., Nat. Photon. **2**, 77 (2008).
2) Shur *et al*., Proc. SPIE **6894**, 689419 (2008).
3) Yasan *et al*., APL **83**, 4701 (2003).
4) Narukawa *et al*., JJAP **45**, L1084 (2006).
5) Hirayama *et al*., APEX **1**, 051101 (2008).
6) Taniyasu *et al*., Nature **441**, 325 (2006).

Polarization discontinuity at heterointerfaces

Wurtzite lattice

$C_{6v}{}^4$: uniaxial anisotropy

no inversion symmetry along the *c*-axis

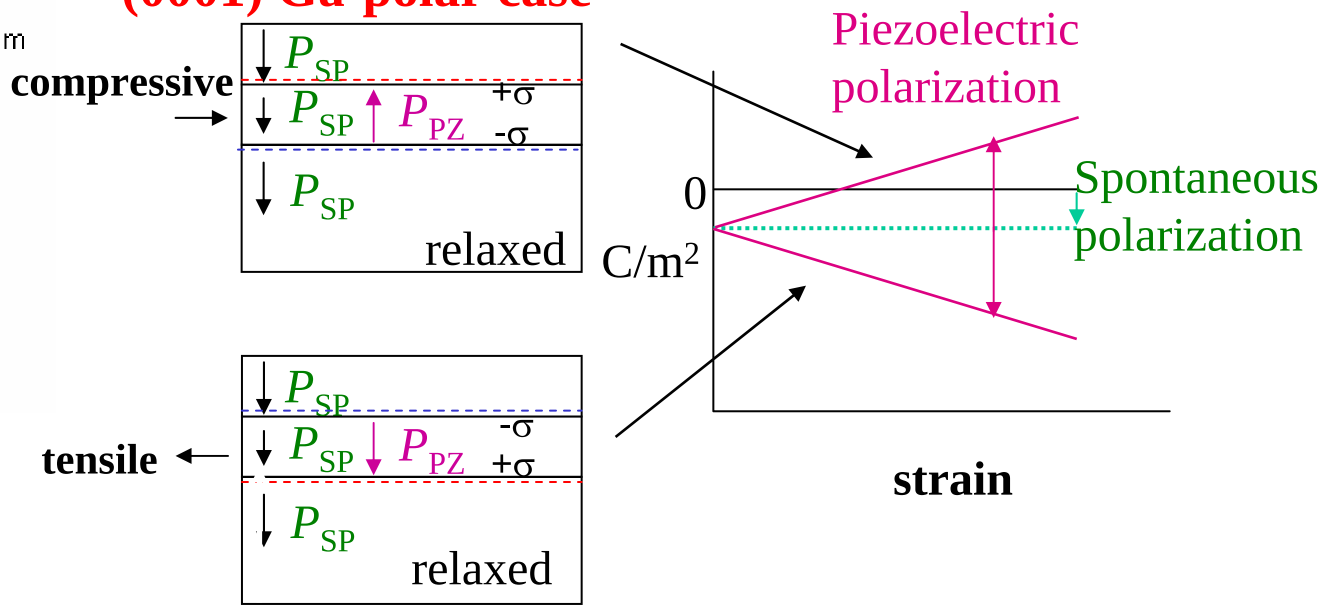


E. Hellman, MRS Internet J. Nitride Semicond. Res. **3**,11 (1998).

Low crystal symmetry :
No inversion symmetry along the *c*-axis
→spontaneous polarization (P_{SP})
Lattice mismatched STRAINED heterostructures
→piezoelectric polarization (P_{PZ})

Polarization discontinuity produces immobile charges (±σ)at the interfaces

(0001) Ga-polar case



F. Bernardini, V. Fiorentini, and D. Vanderbilt, Phys. Rev. B 56, R10024 (1997).

Avoid polarization fields - off *c*-axis semipolar -

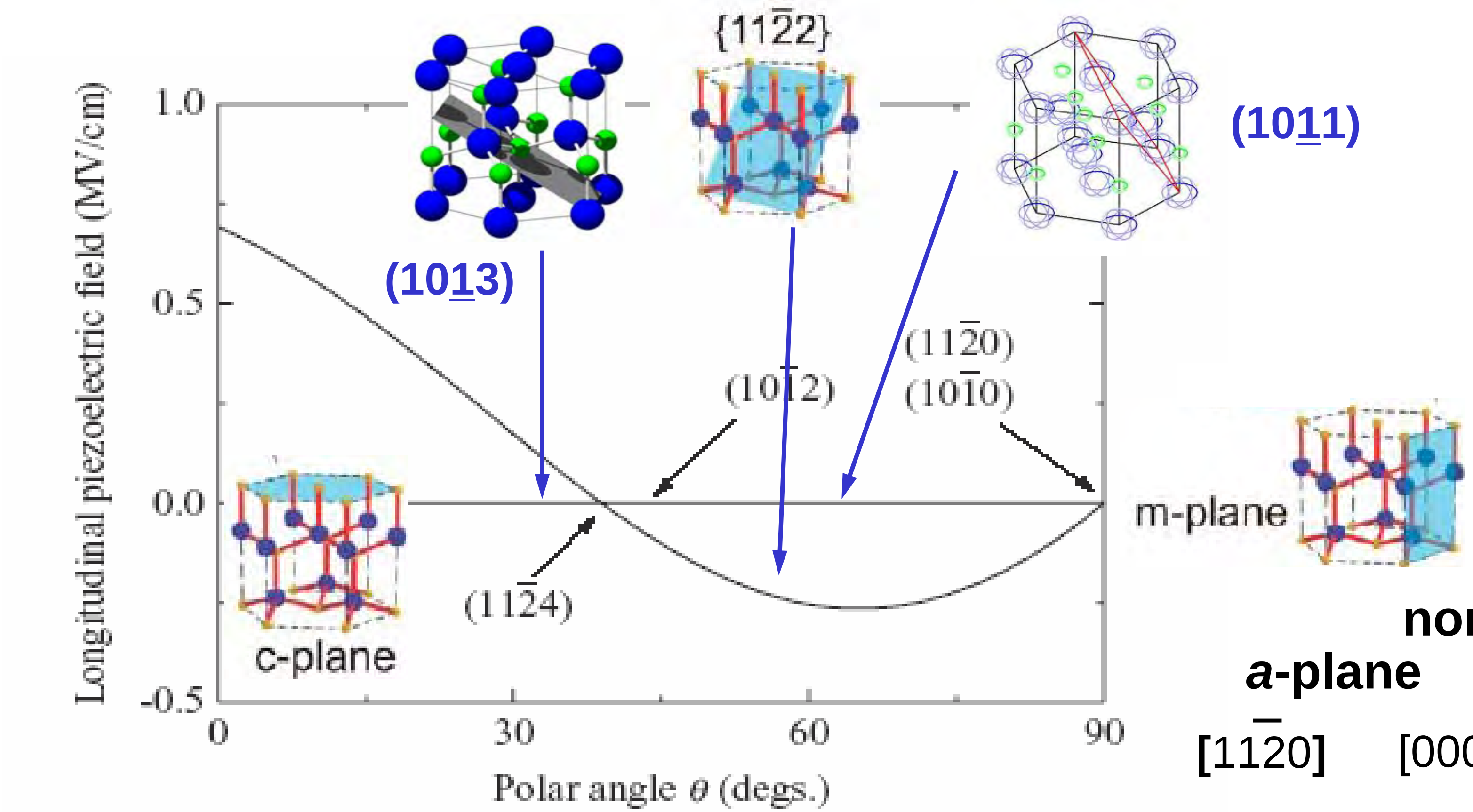
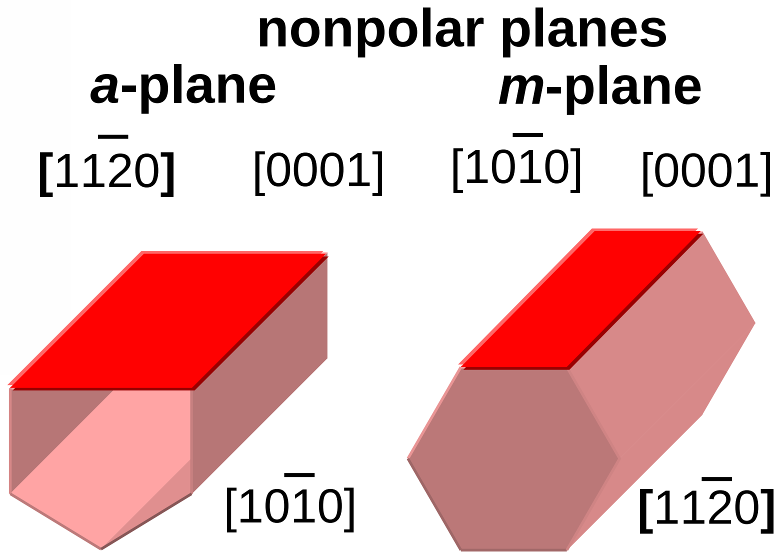


Fig. 2. Calculated longitudinal piezoelectric field in strained $Ga_{0.9}In_{0.1}N$ on GaN as a function of the polar angle from (0001).*



*** T. Takeuchi *et al*., Jpn. J. Appl. Phys. 39, L413 (2000).**
and U. Schwarz and M. Kneissl, PSS (PRL) 1, A44 (2007).

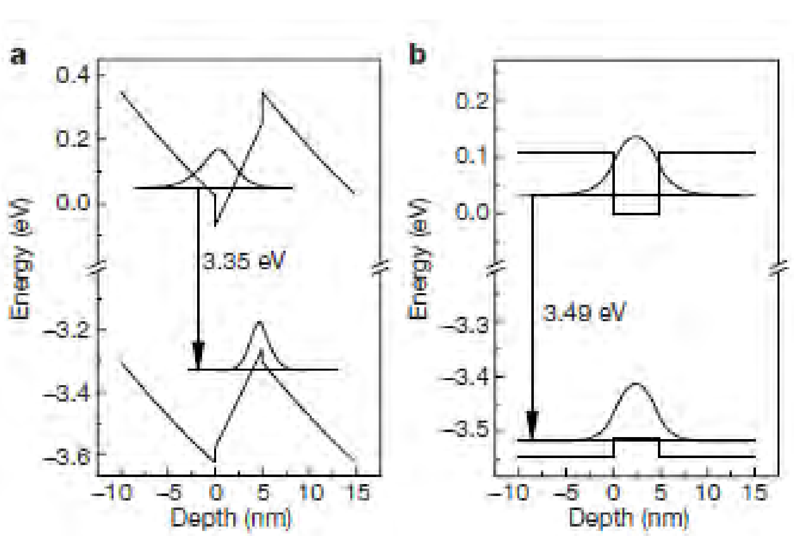
m-plane GaN and AlGaN/GaN / γ-$LiAlO_2$

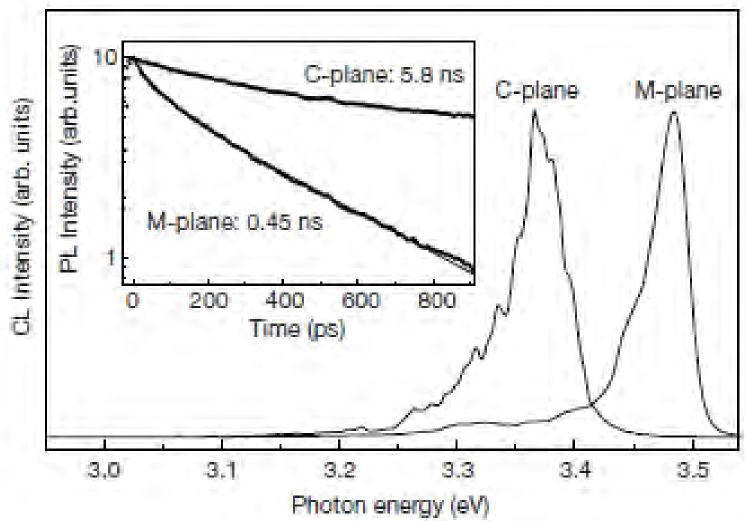
Nitride semiconductors free of electrostatic fields for efficient white light-emitting diodes

P. Waltereit, O. Brandt, A. Trampert, H. T. Grahn, J. Menniger, M. Ramsteiner, M. Reiche & K. H. Ploog

Paul-Drude-Institut für Festkörperelektronik, Hausvogteiplatz 5-7, D-10117 Berlin, Germany

Compact solid-state lamps based on light-emitting diodes (LEDs)[1,2] are of current technological interest as an alternative to conventional light bulbs. The brightest LEDs available so far emit red light and exhibit higher luminous efficiency than fluorescent lamps. If this luminous efficiency could be transferred to white LEDs, power consumption would be dramatically reduced, with great economic and ecological consequences. But the luminous efficiency of existing white LEDs is still very low, owing to the presence of electrostatic fields within the active layers[3]. These fields are generated by the spontaneous and piezoelectric polarization along the [0001] axis of hexagonal group-III nitrides—the commonly used materials for light generation[4–6]. Unfortunately, as this crystallographic orientation corresponds to the natural growth direction of these materials deposited on currently available substrates[7]. Here we demonstrate that the epitaxial growth of GaN/(Al,Ga)N on tetragonal $LiAlO_2$ in a non-polar direction allows the fabrication of structures free of electrostatic fields, resulting in an improved quantum efficiency. We expect that this approach will pave the way towards highly efficient white LEDs.



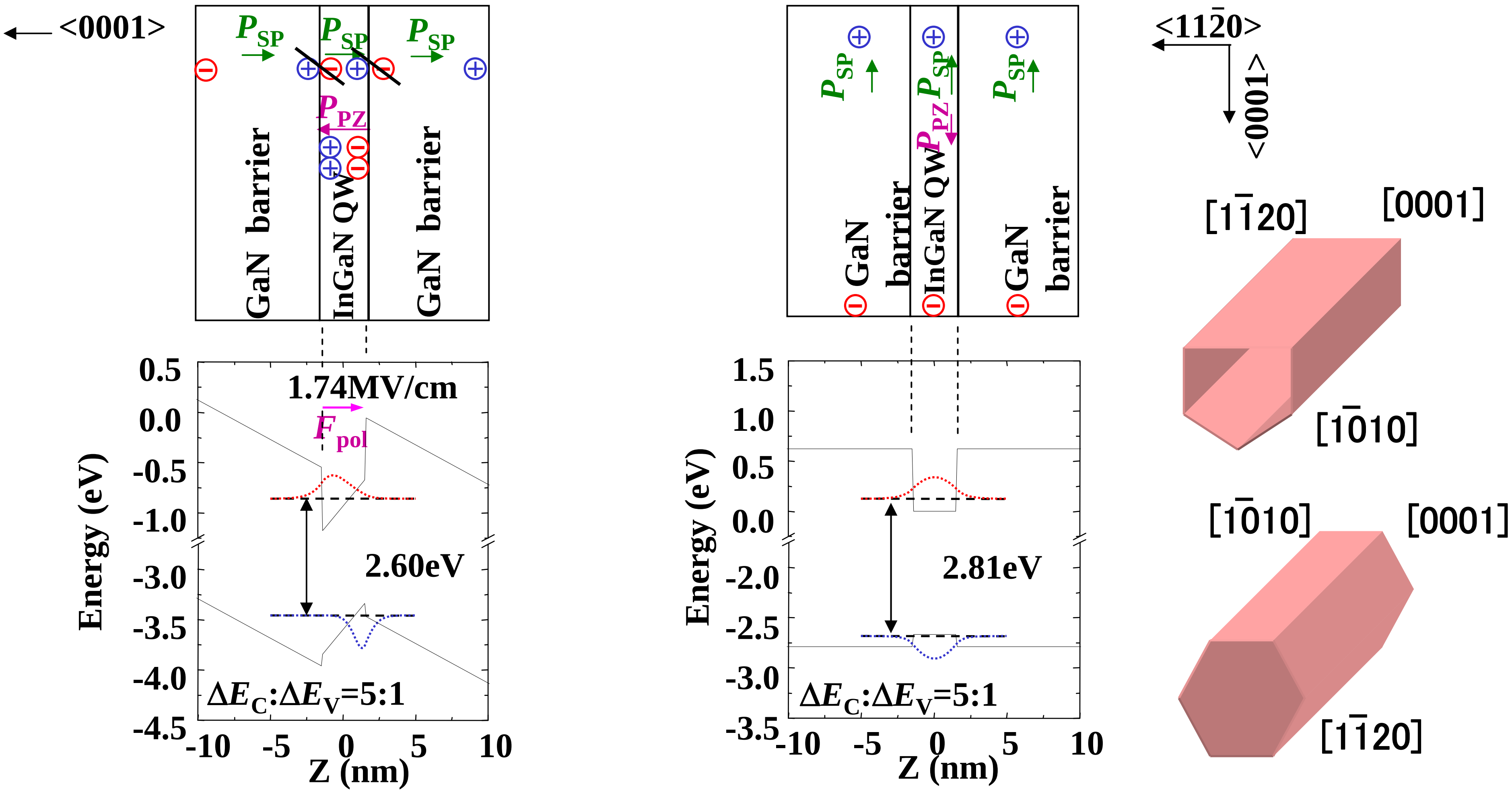


P. Waltereit *et al*., Nature 406, 865 (2000).

Nonpolar *m*- and *a*-plane InGaN/GaN

polar (0001)
$In_{0.15}Ga_{0.85}N$ (3nm)/GaN(15nm)

Nonpolar (1120), (1$\bar{1}$00), (001)
$In_{0.15}Ga_{0.85}N$ (3nm)/GaN(15nm)



SFC *et al*., Nat. Mater. 5, 810 (2006)

Nonpolar light-emitting diodes (LEDs)

***a*-plane**

C. Q. Chen, V. Adivarahan, J. W. Yang, M. Shatalov, E. Kuokstis and M. A. Khan: Jpn. J. Appl. Phys. 42, L1039 (2003).
MOCVD, GaN / $Al_{0.12}Ga_{0.88}N$ (3x), on *r*-plane Al_2O_3 University of South Carolina

A. Chitnis, C. Chen, V. Adivarahan, M. Shatalov, E. Kuokstis, V. Mandavilli, J. Yang and M. A. Khan: Appl. Phys. Lett. 84, 3663 (2004).
MOCVD, $In_{0.15}Ga_{0.85}N$ / GaN (3x), on *r*-plane Al_2O_3 University of South Carolina

A. Chakraborty, B. Haskell, S. Keller, J. S. Speck, S. P. DenBaars, S. Nakamura and U. K. Mishra: Appl. Phys. Lett. 85, 5143 (2004).
MOCVD, $In_{0.17}Ga_{0.83}N$ / GaN (5x), on HVPE LEO *a*-plane GaN template UCSB

***m*-plane**

A. Chakraborty, B. Haskell, S. Keller, J. S. Speck, S. P. DenBaars, S. Nakamura and U. K. Mishra: Jpn. J. Appl. Phys. 44, L173 (2004).
MOCVD, $In_{0.17}Ga_{0.83}N$ / GaN (5x), on free-standing *m*-plane GaN template UCSB

N. F. Gardner, J. C. Kim, J. J. Wierer, Y. C. Shen, and M. R. Krames: Appl. Phys. Lett. 86, 111101 (2005).
MOCVD, InGaN / GaN, on *m*-plane 4H-SiC Lumileds Lighting

A. Chakraborty, B. Haskell, H. Masui, S. Keller, J. S. Speck, S. P. DenBaars, S. Nakamura and U. K. Mishra: Jpn. J. Appl. Phys. 45, 739 (2006).
MOCVD, $In_{0.16}Ga_{0.84}N$ / GaN (5x), on free-standing *m*-plane GaN template UCSB

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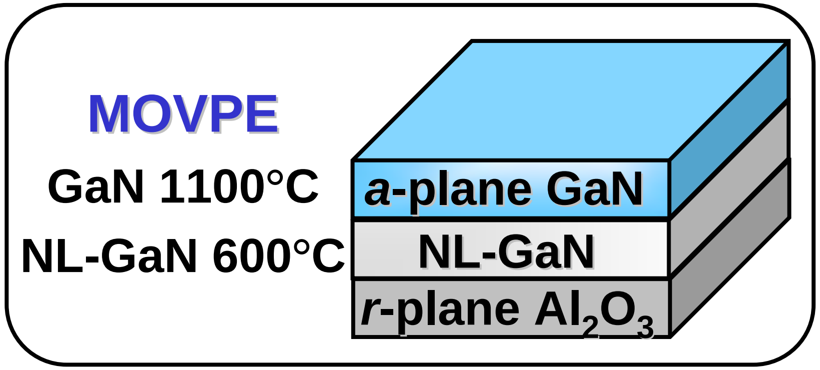
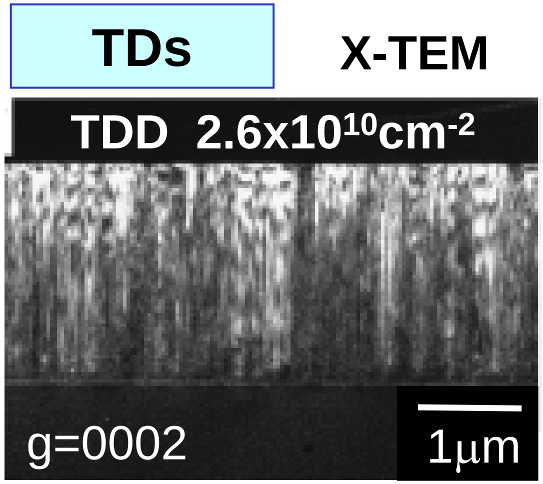
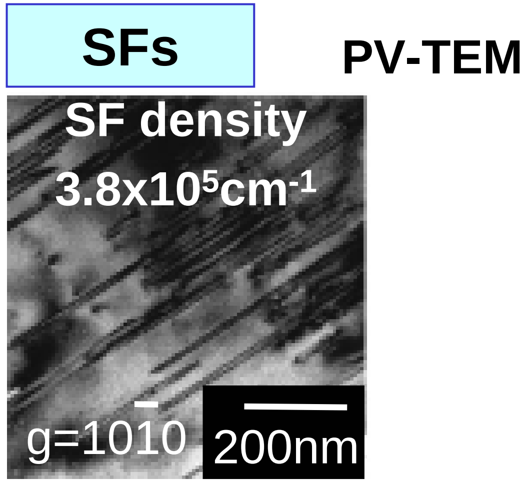
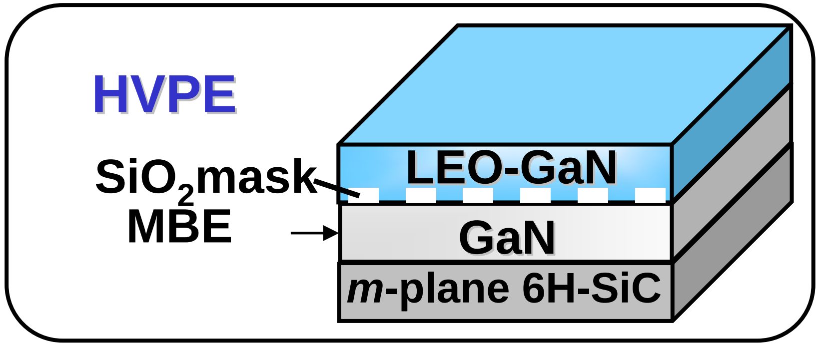
Issues in nonpolar GaN heteroepitaxy



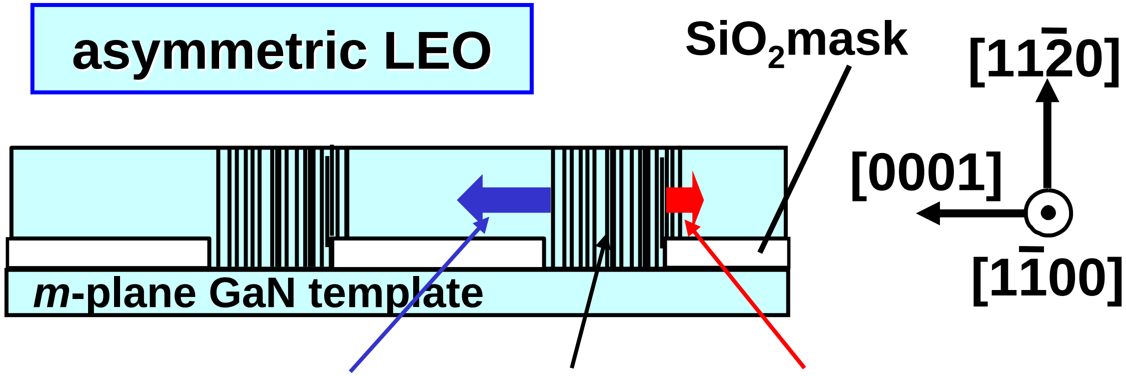
1) Craven *et al.*, APL 81, 469 (2002).





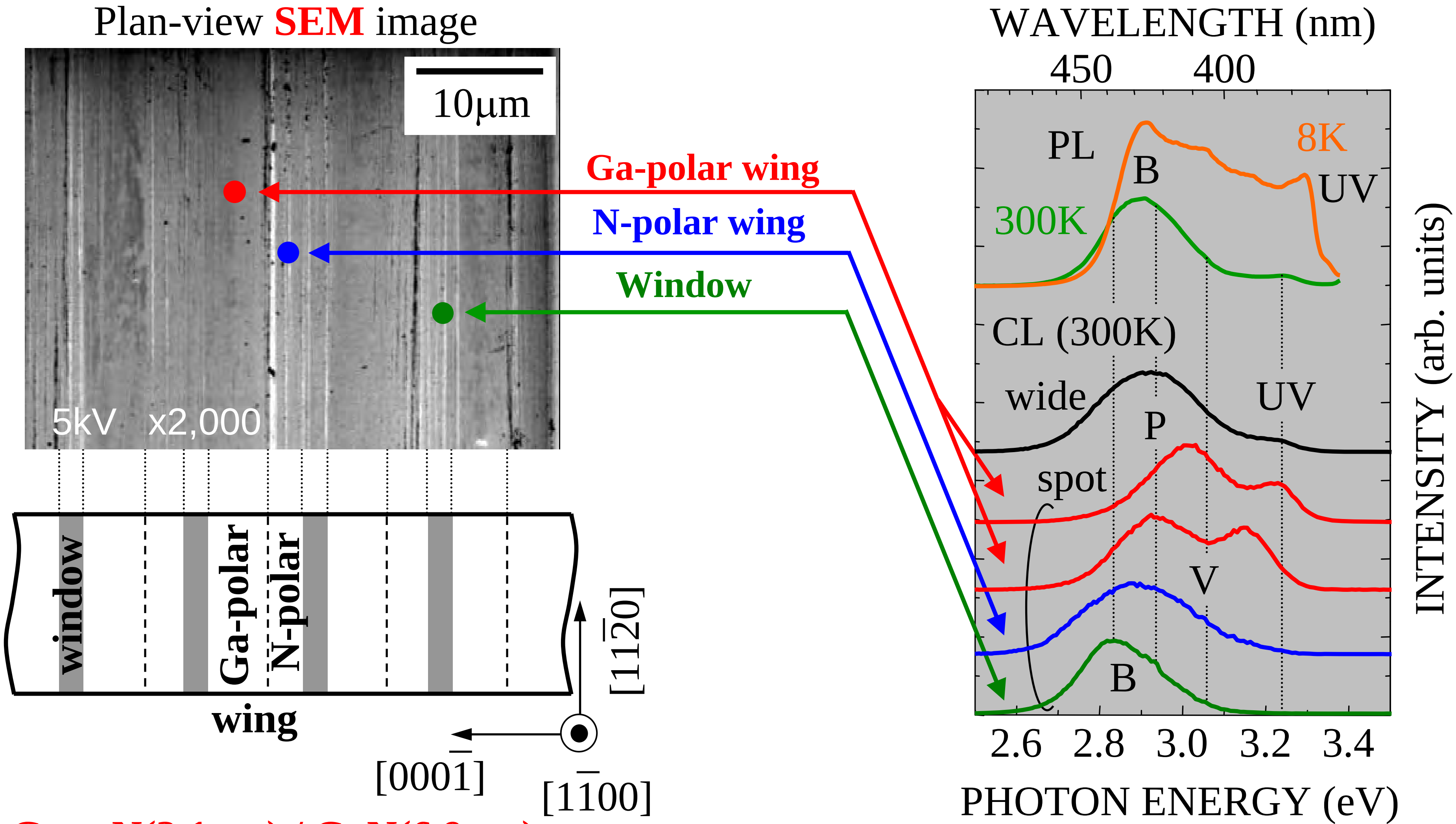


2) Haskell *et al.*, APL 86, 111917 (2005).



	m-plane GaN template		Ga-polar Wing		Window		N-polar Wing
TDD (cm^{-2})	2×10^9	>>	5×10^6	<<	4×10^9	>>	5×10^6
SFD (cm^{-1})	1×10^5	>>	3×10^3	<<	1×10^5	=	1×10^5

m-plane InGaN QWs grown on LEO GaN base



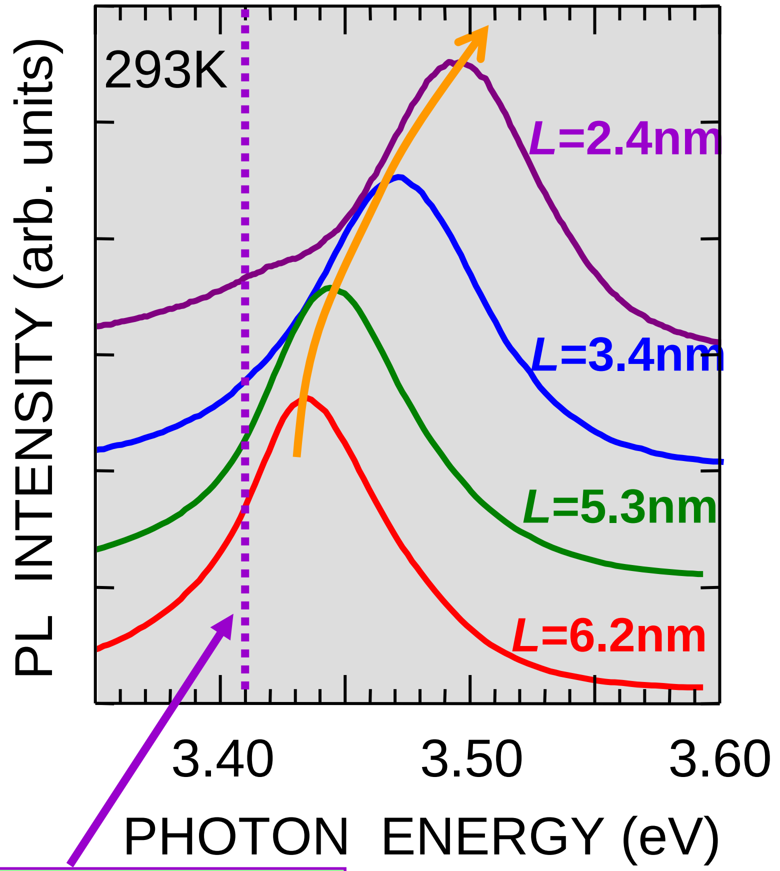
$In_{0.08}Ga_{0.92}N$(3.1nm) / GaN(6.9nm)

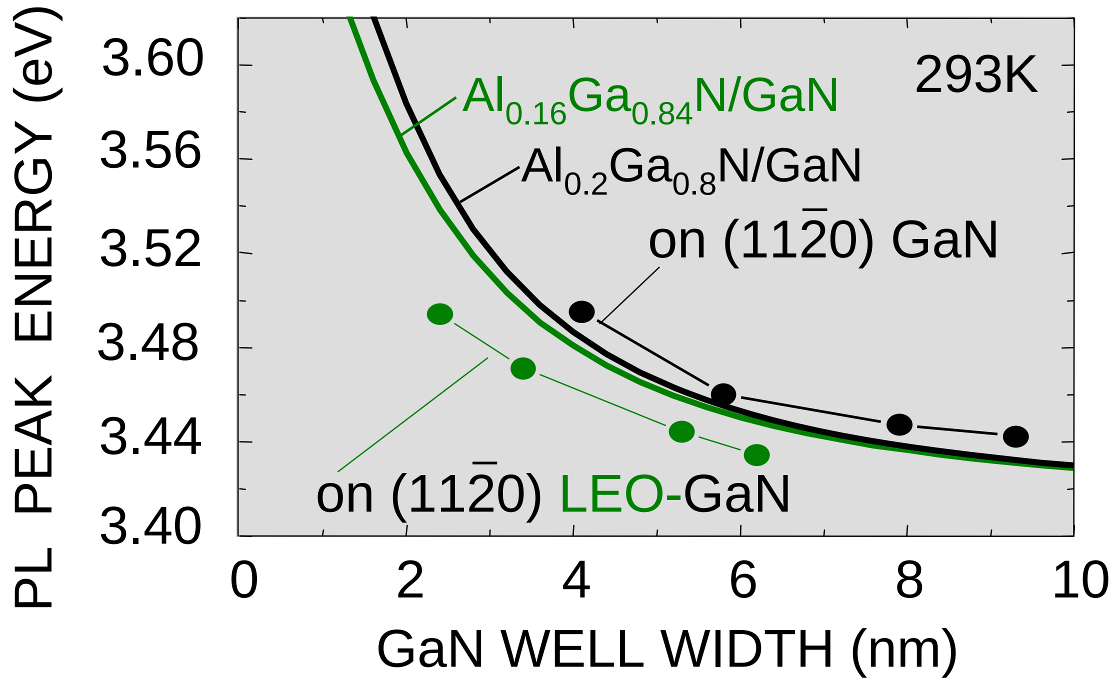
Onuma *et al*., JVST B <u>25</u>, 1524 (2007).

InN molar fraction: depends on the base structure

Nonpolar *a*-plane AlGaN/GaN QWs

on (11$\bar{2}$0) LEO-GaN

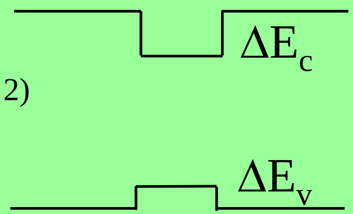




strain-free
(0001)LEO-GaN

no QCSE !

Schrödinger eq.



$m_e=0.18m_0$, $m_h=1.52m_0$ [2)]

$\Delta E_c:\Delta E_v=3:1$

$E_g(Al_xGa_{1-x}N)$
$=6.138x+3.412(1-x)-0.82x(1-x)$ [3)]

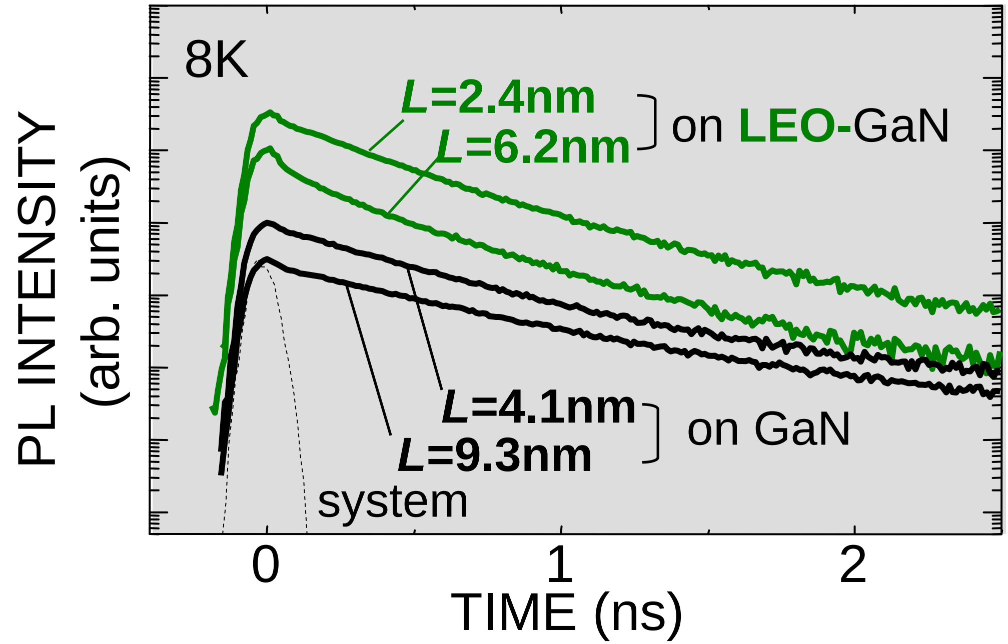
[2)] **M. Suzuki *et al.*, PRB 52, 8132 (1995) .**

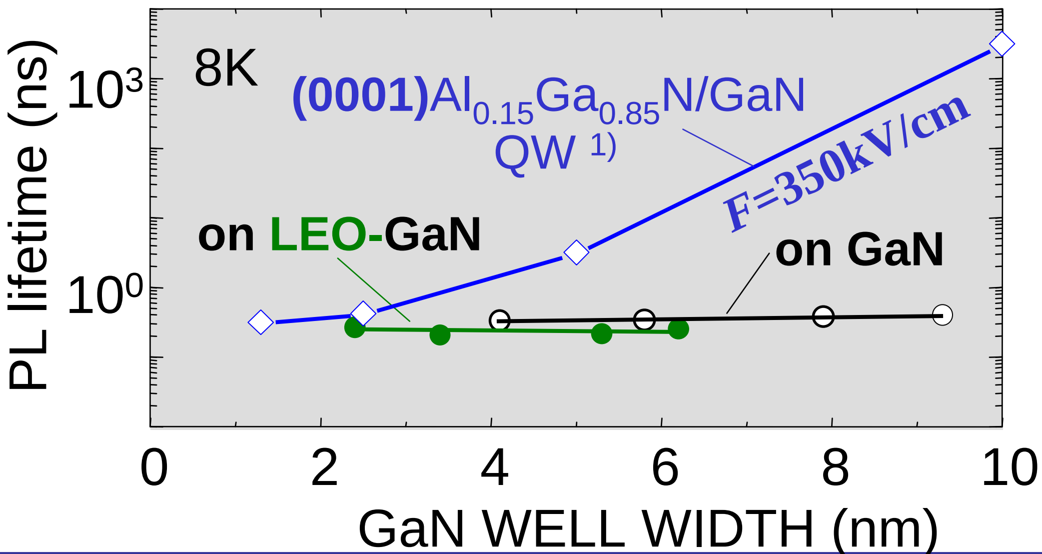
[3)] **T. Onuma *et al.*, JAP 95, 2495 (2004).**

Koida *et al.*, APL 84, 3768 (2004).

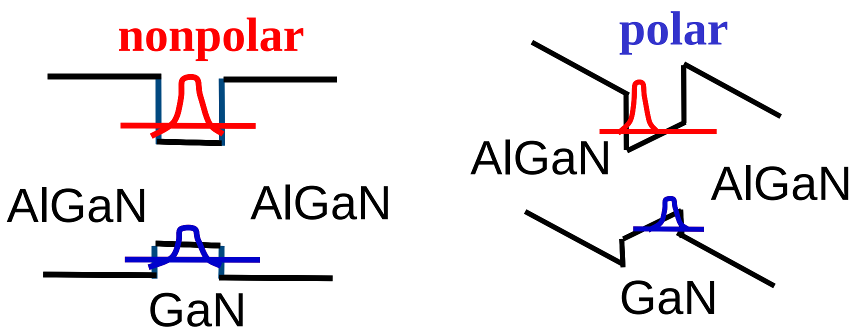
Nonpolar *a*-plane AlGaN/GaN QWs

Ti:sapphire 242nm 2mW **APL 84, 3768 (2004).**





τ_{PL} independent of L ⇨ no polarization fields



short τ_R
comparable to (0001)AlGaN/GaN QW
(L=1.3, 2.5nm)

$\tau_{PL} \sim \tau_R$ (L.T.)

[1] Im *et al.*, PRB 57, R9435 (1998).

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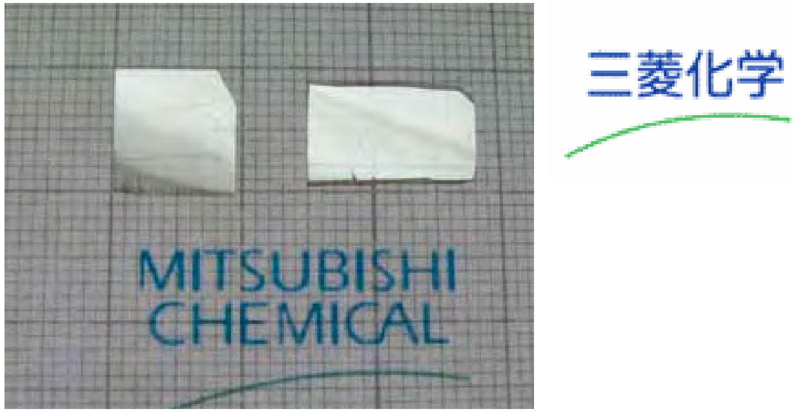
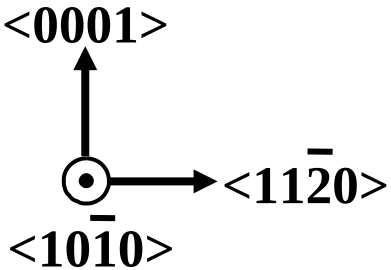
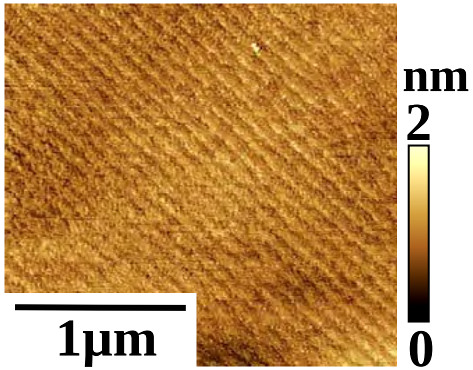
m-plane free-standing (FS)-GaN substrates

m-plane FS-GaN wafers



TDD$<5\times10^{6}$ cm^{-2},
SFD$<1\times10^{3}$ cm^{-1}



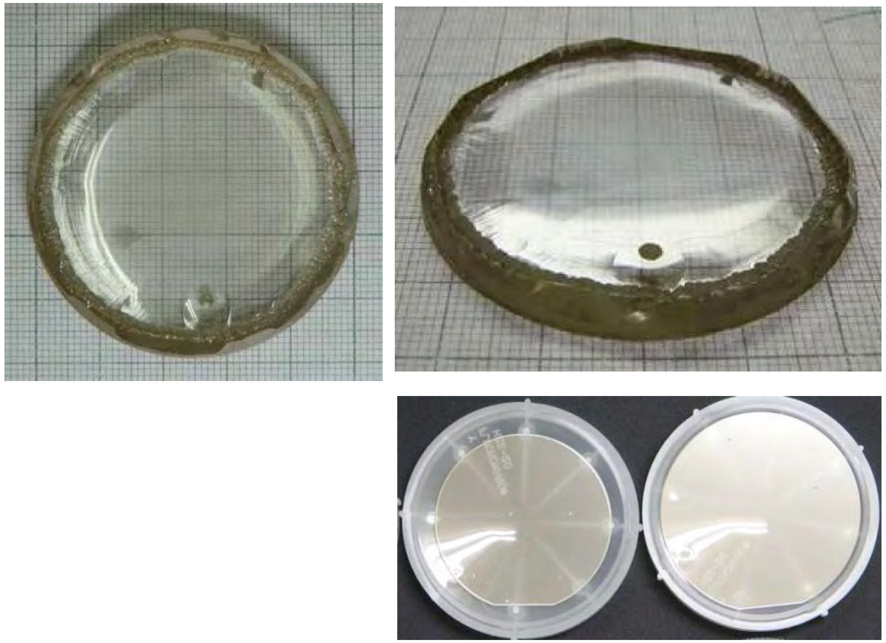


ML step 0.26nm
RMS ≅0.072nm

Fujito *et al*., pss(a) 205, 1056 (2008).

cut from 10-mm-thick *c*-plane GaN grown by HVPE (Mitsubishi Chemical Co.)

c-plane FS-GaN boule



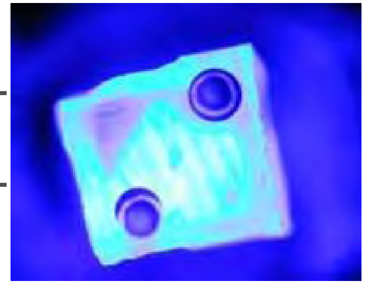
SFC *et al*., APL 92, 091912 (2008).

Progress of *m*-plane InGaN devices using low defect density FS-GaN substrates

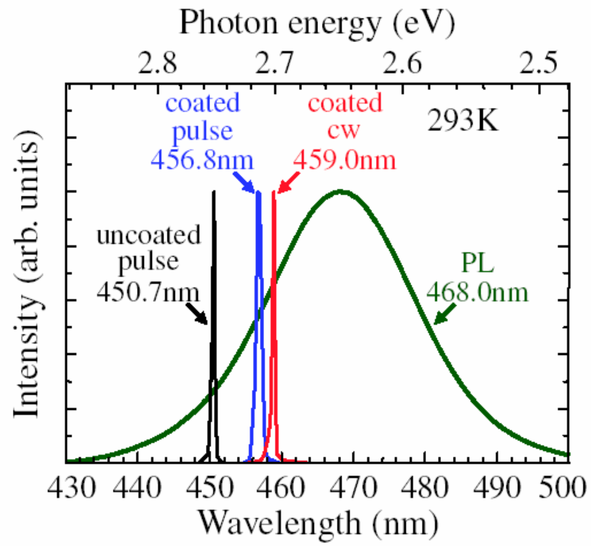
UCSB — Excellence in Electronics ROHM

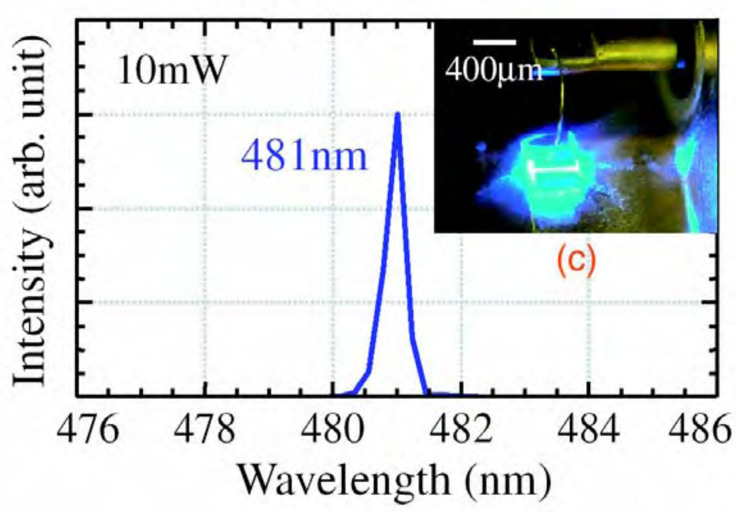
m-plane InGaN **LEDs**

		Driving current 20mA	
Wavelength (nm)	407.4		435
Output power (mW)	23.7		1.79
EQE (%)	38.9		3.1



Schmidt *et al*., JJAP 46, L126 (2007). **Okamoto *et al*., JJAP 45, L1197 (2006).**





m-plane InGaN **LDs**

	Pulsed	CW	Pulsed	CW	CW
Lasing wavelength (nm)	405.5	404	451.8	459 ⇨	481
J_{th} (kA/cm^2)	7.5	4.0	22.3	5.0	6.1

Schmidt *et al*., JJAP 46, L190 (2007). JJAP 46, L187; L820 (2007). APEX 1, 011102 (2008). APEX 1, 072201 (2008).

Issues in (In,Ga)N growth on *m*-plane substrate

√ **Is the *m*-plane substrate ready for homoepitaxy ?**
√ **Are the growth conditions similar to *c*-plane growth ?**
√ **Are InGaN alloys grown coherently ?**
√ **Are the optical properties promising ?**
√ **.... etc.**

Labor the issues in (In,Ga)N growth by MOVPE on the low DD *m*-plane FS-GaN substrates

Outline

1. Introduction

2. Issues on heteroepitaxial nonpolar (Al,In,Ga)N [UCSB samples]
   √ Planar growth and lateral epitaxial overgrowth of GaN
   √ Optical properties of InGaN/GaN and AlGaN/GaN quantum wells

3. Homoepitaxial nonpolar (In,Ga)N [Tohoku-films ROHM-devices]
   √ Low defect density freestanding (FS) *m*-plane GaN substrate
   √ **GaN and InGaN growth by MOVPE**
   √ Device performance Digest -- *m*-plane LEDs and LDs

4. Summary

papers available from http://www.tagen.tohoku.ac.jp/labo/chichibu/SHIGEFUSA/paper/GaN.html

Growth conditions

Horizontal MOVPE (Tohoku Univ.)

InGaN 200-250nm
GaN 1.5μm
m-plane GaN substrate
325μm

InGaN

P_{growth} : 6.6×10^4 Pa
N_2 carrier
TMGa+TMIn+NH_3
R_g=0.1 μm/h
T_g=750-820 °C
V/III =40000-1000000
APL 93, 151908 (2008).

GaN

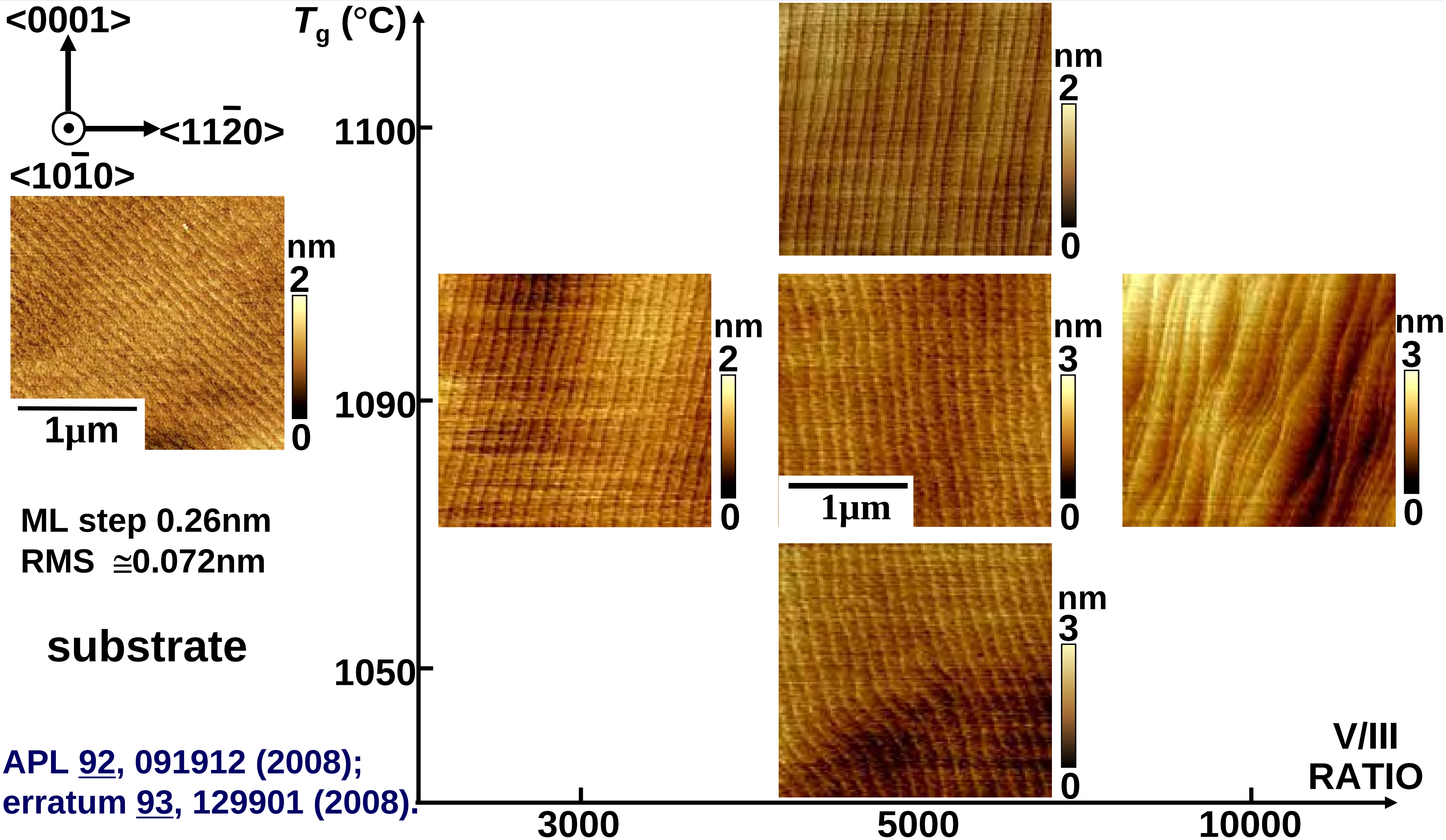
P_{growth} : 5.3×10^4 Pa
H_2 carrier
TMGa+NH_3
R_g=1.5 μm/h
T_g=1090 °C
V/III =5000
APL 92, 091912 (2008); erratum 93, 129901 (2008).

m-plane GaN cut from 10mm-thick *c*-plane GaN substrate (Mitsubishi)

TDD < 5×10^6 cm^{-2},
SFD < 10^3 cm^{-1},
$\Delta\omega$(10-10)≅90 arcsec

c-plane FS-GaN (TDD < 10^7 cm^{-2})
and c-plane GaN/(0001)Al_2O_3 for comparison

MOVPE *m*-plane GaN homoepitaxy (1.5μm)

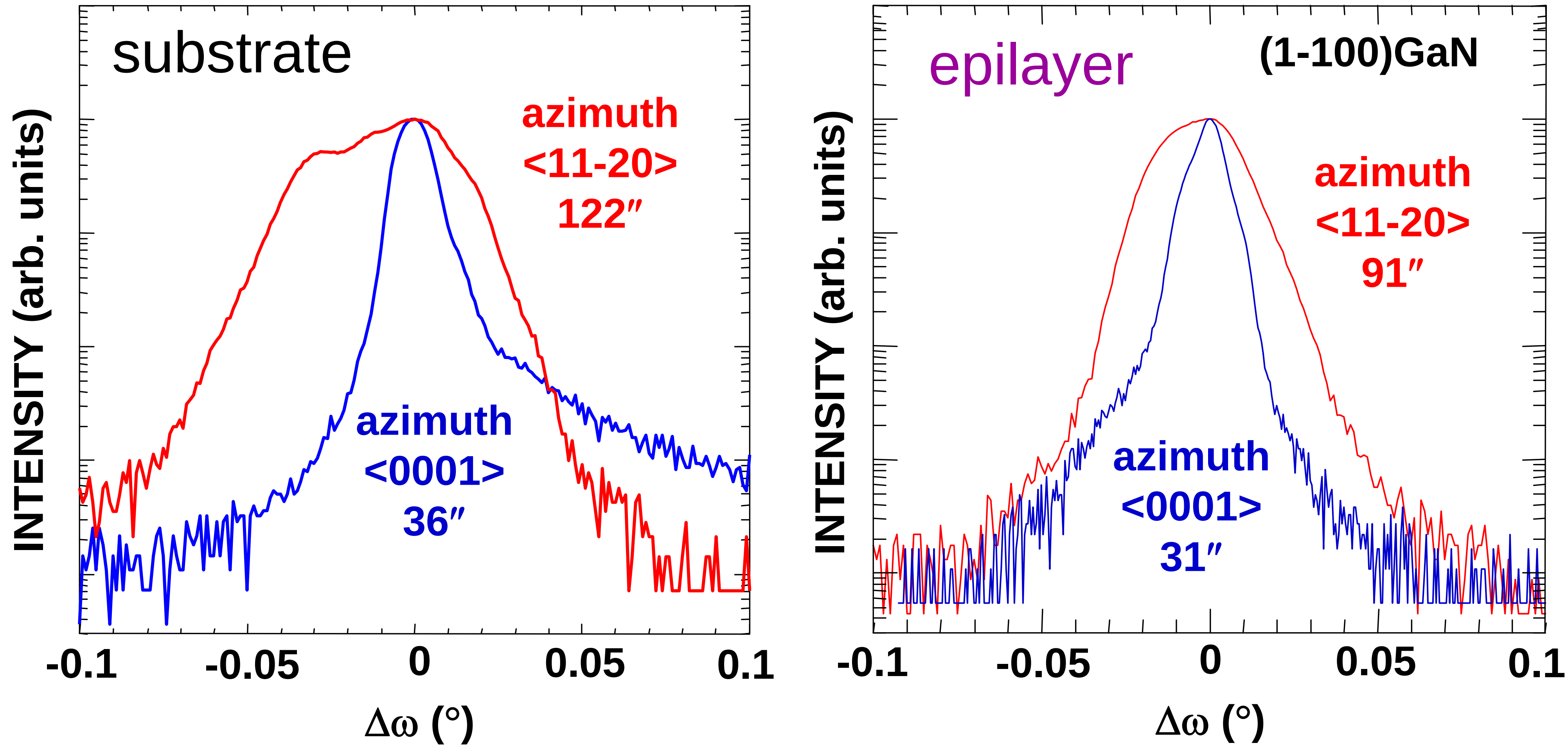


APL 92, 091912 (2008);
erratum 93, 129901 (2008).

XRCs of GaN substrate and epilayer

T_g=1050°C
V/III=5000

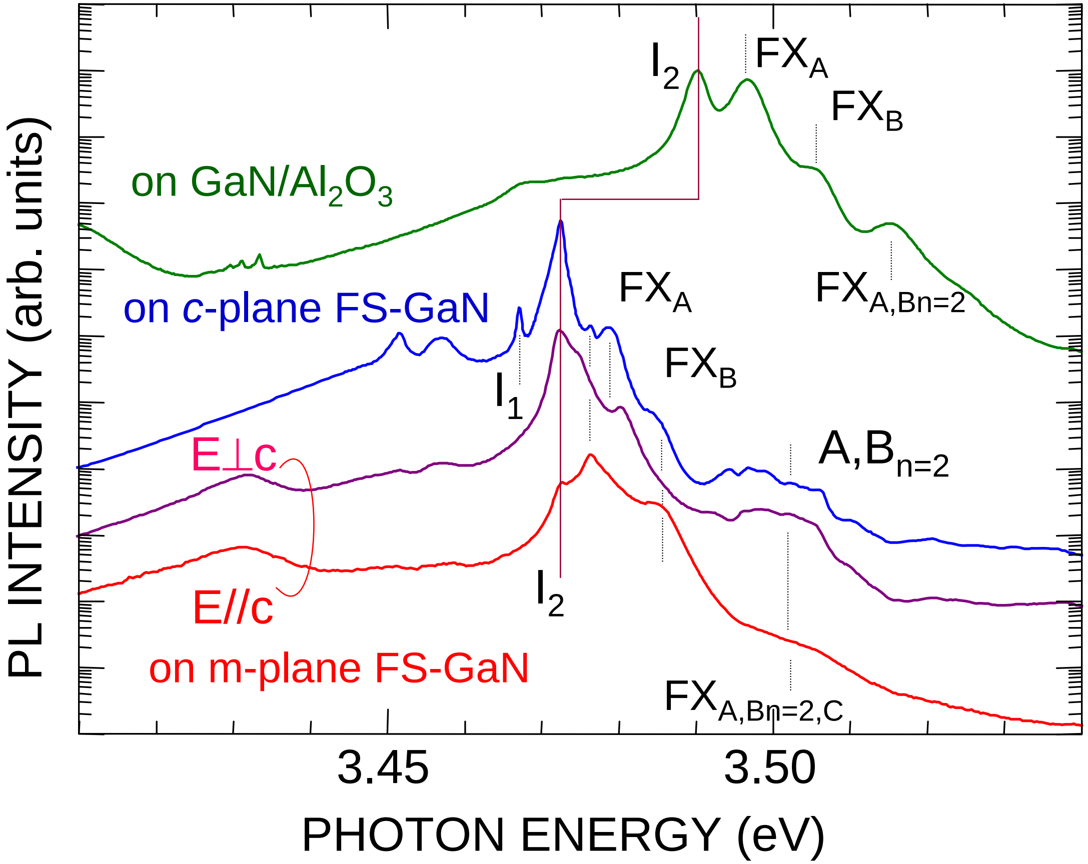
<(10$\bar{1}$0)XRC>

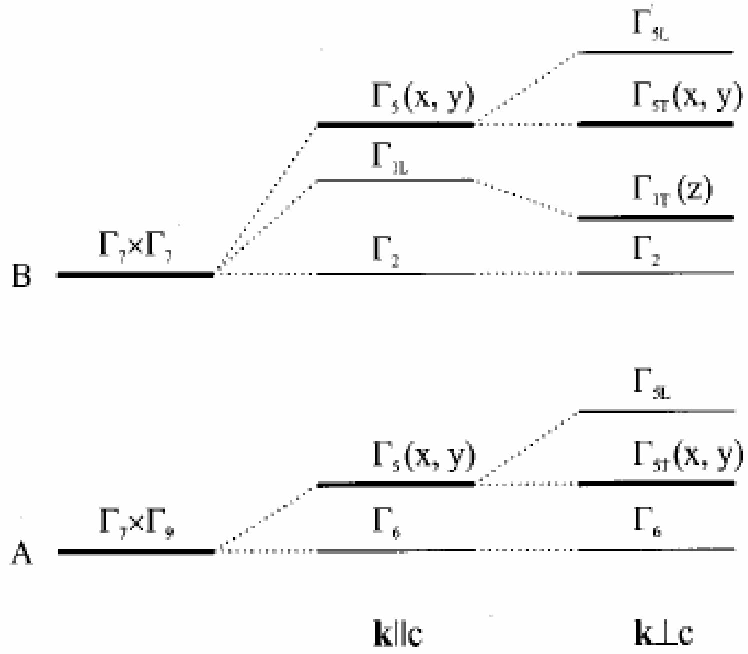


SFC *et al.*, APL 92, 091912 (2008);erratum 93, 129901 (2008).

m-plane GaN : Polarized low temp. NBE PL

He-Cd 325.0 nm 38W/cm² 10K





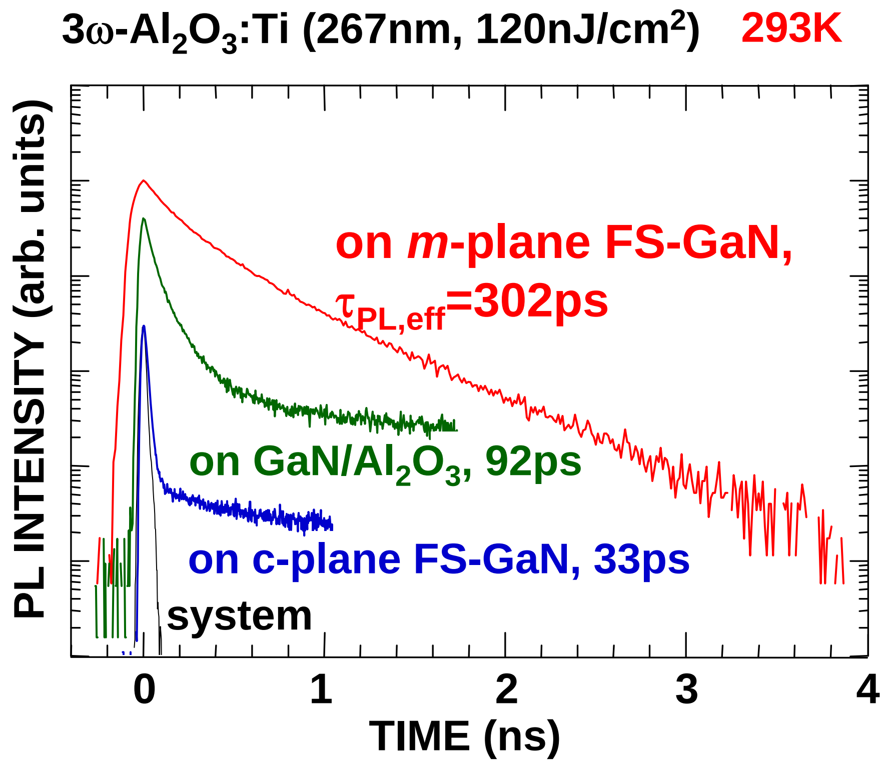
Paskov PR B 64, 115201(2001)

α-polarization (k//c)

σ-polarization (k⊥c, E⊥c)

π-polarization (k⊥c, E//c)

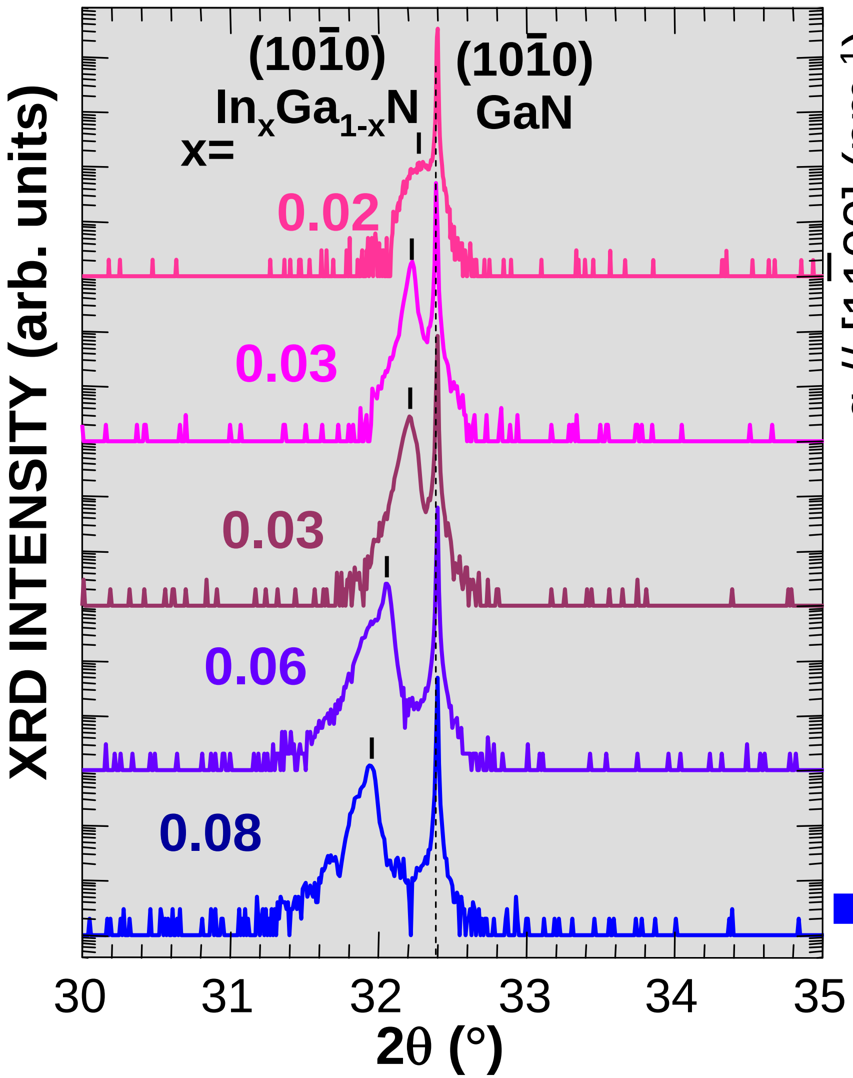
m-plane GaN homoepitaxy (1.5μm) - TRPL -



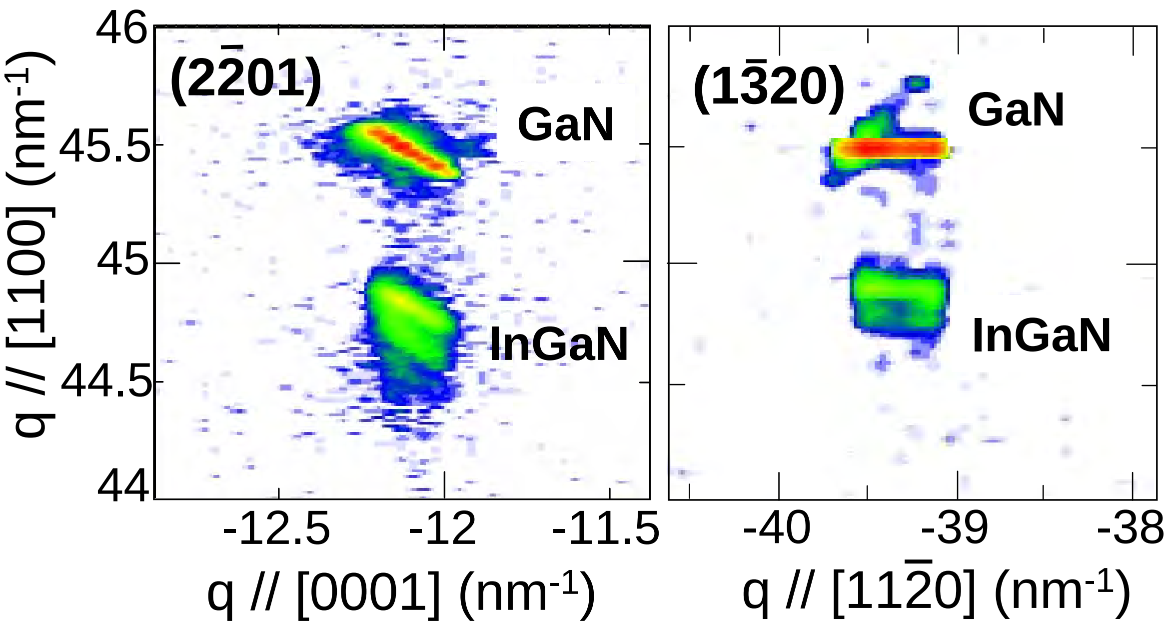
SFC *et al.*, APL <u>92</u>, 091912 (2008);erratum <u>93</u>, 129901 (2008).

m-plane InGaN (200nm) / GaN : x-ray analyses

<XRD>



<Reciprocal Space Mapping (x=0.08)>



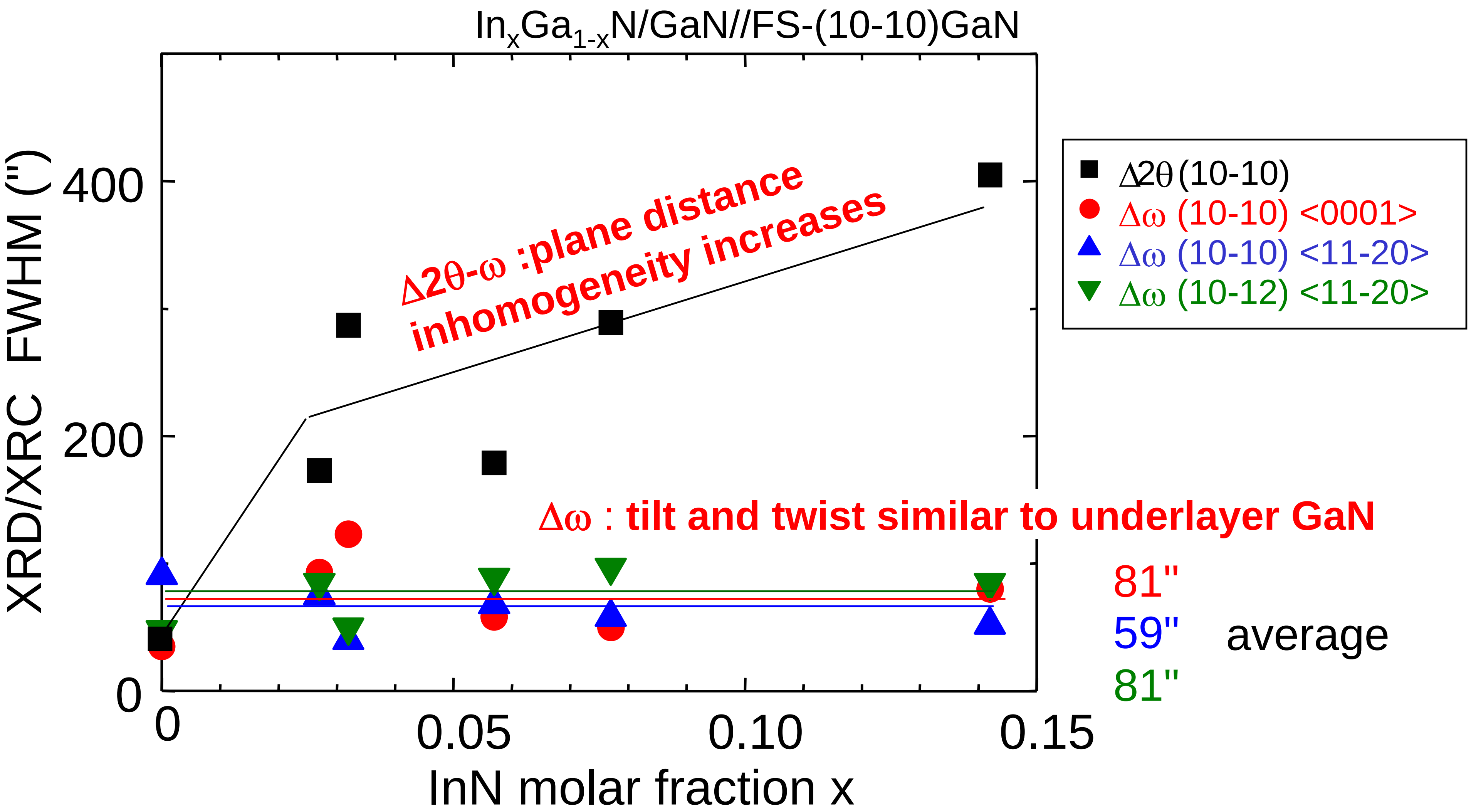
coherent growth
(anisotropic strain)

Tg : 750°C
V/III : 76000

$$\varepsilon_{yy} = \frac{-C_{12}\,\varepsilon_{xx} - C_{13}\,\varepsilon_{zz}}{C_{11}}$$

SFC *et al.*, APL 93, 151908 (2008).

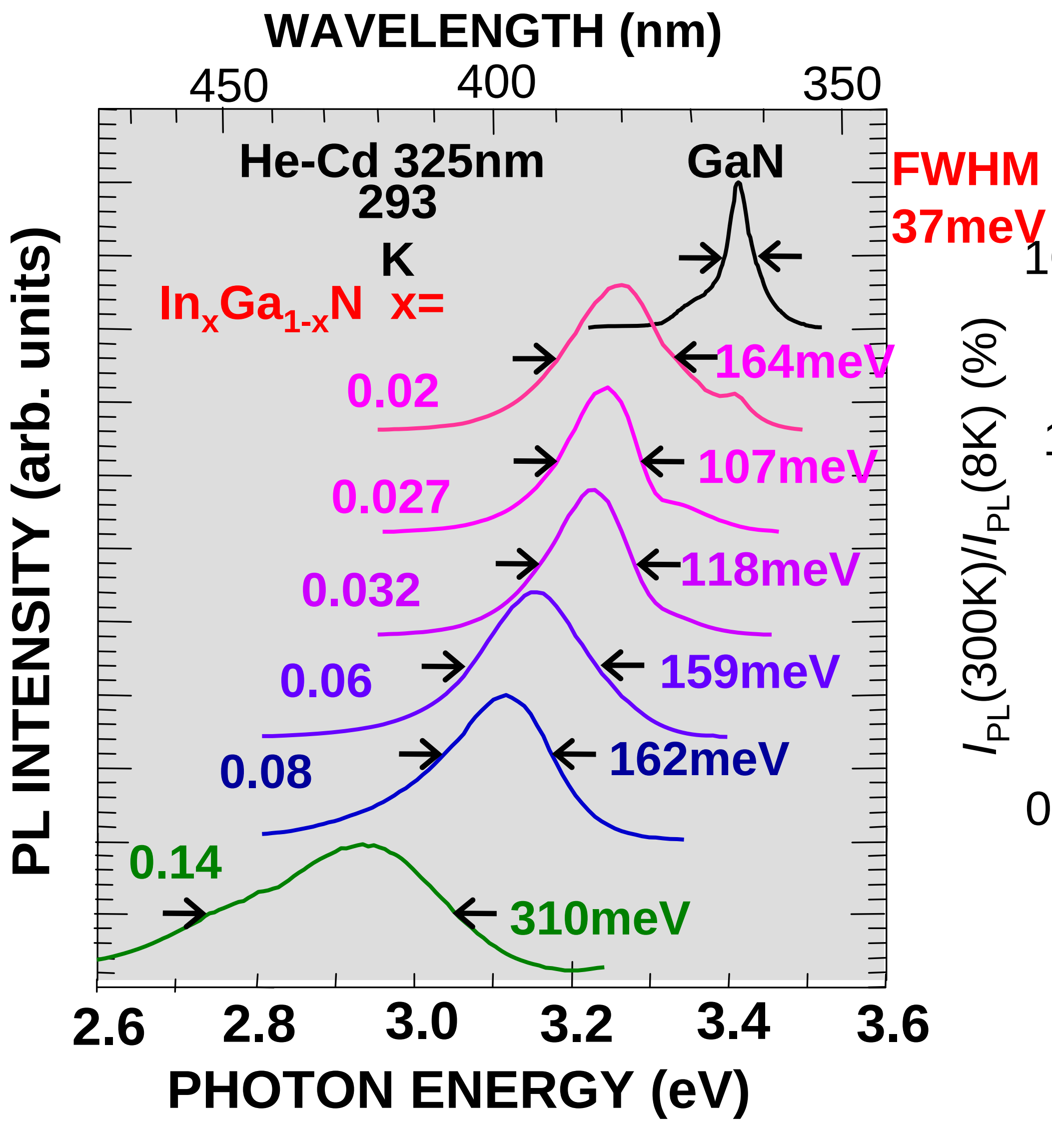
m-plane InGaN (200nm) / GaN : x-ray analyses

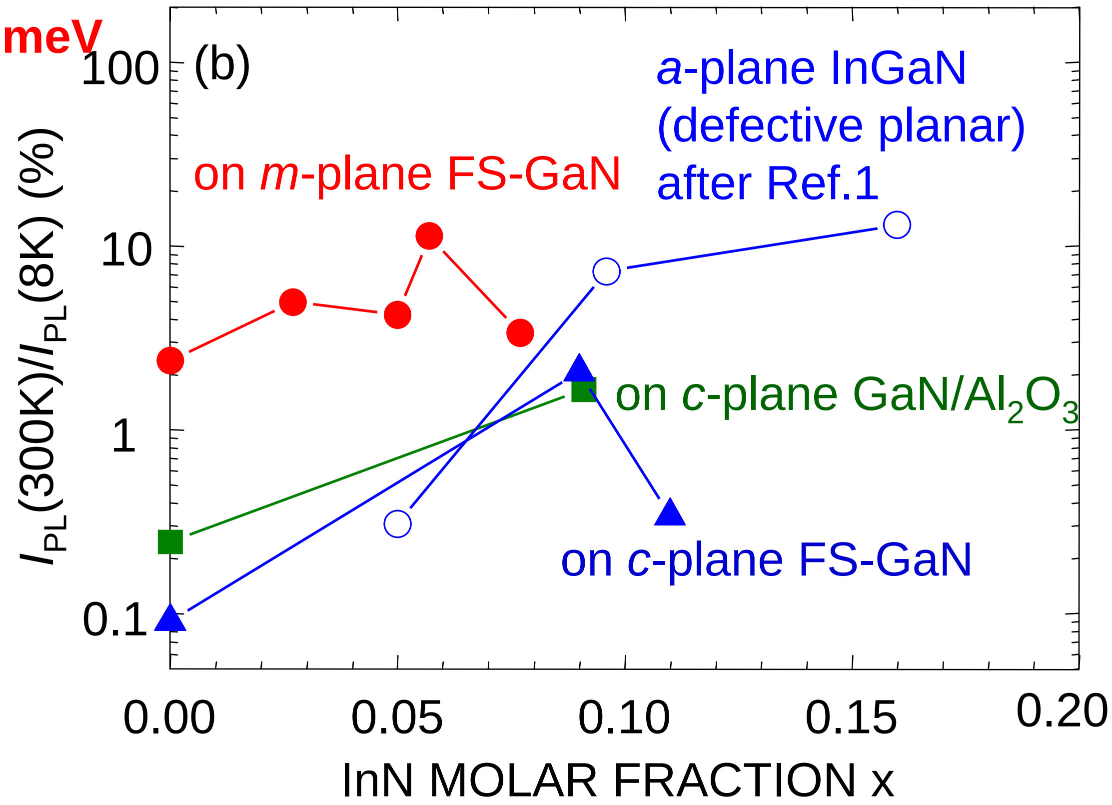


SFC *et al.*, APL <u>93</u>, 151908 (2008).

m-plane InGaN films -- room temperature PL

SFC *et al.*, APL 93, 151908 (2008).



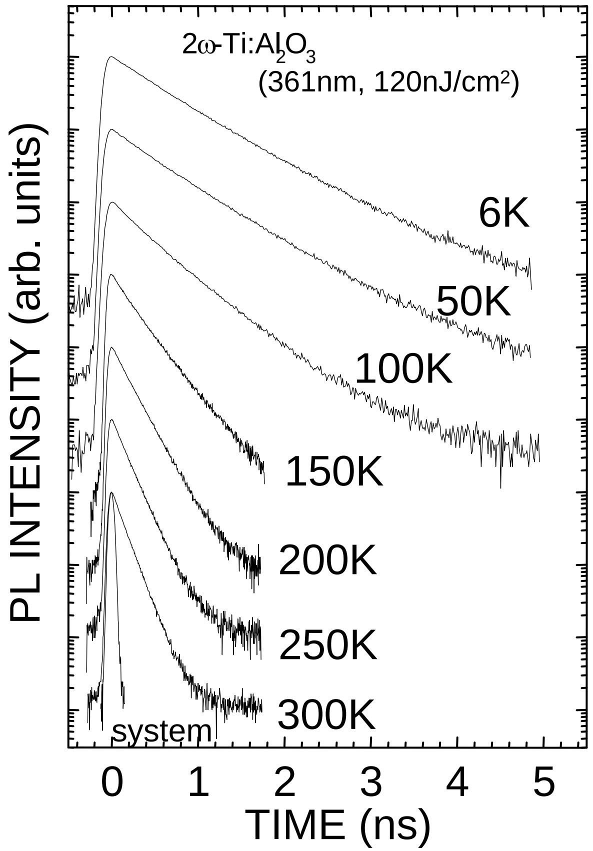


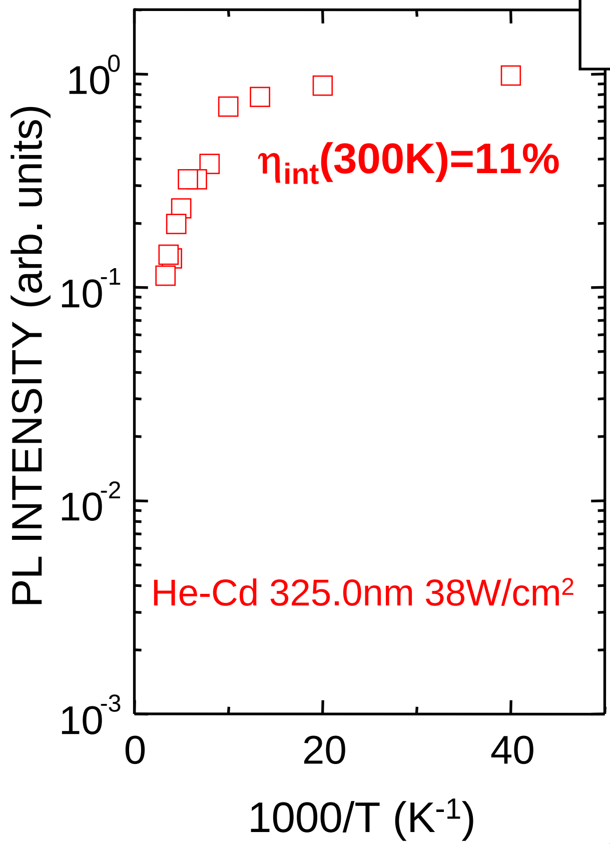
1) SFC *et al.*, Nat. Mater. 6, 810 (2006).

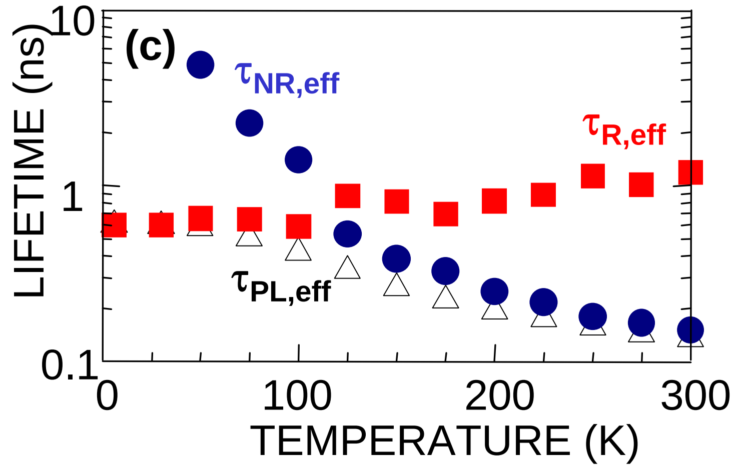
m-plane $In_{0.06}Ga_{0.94}N$ film -- TRPL results vs T

***m*-plane $In_{0.06}Ga_{0.94}N$ film**

Low temperature
Radiative lifetime = 600 ps
(nearly constant below 100 K)
Room temperature
Nonradiative lifetime 150 ps
Radiative lifetime 1.2 ns







SFC *et al.*, APL 93, 151908 (2008).

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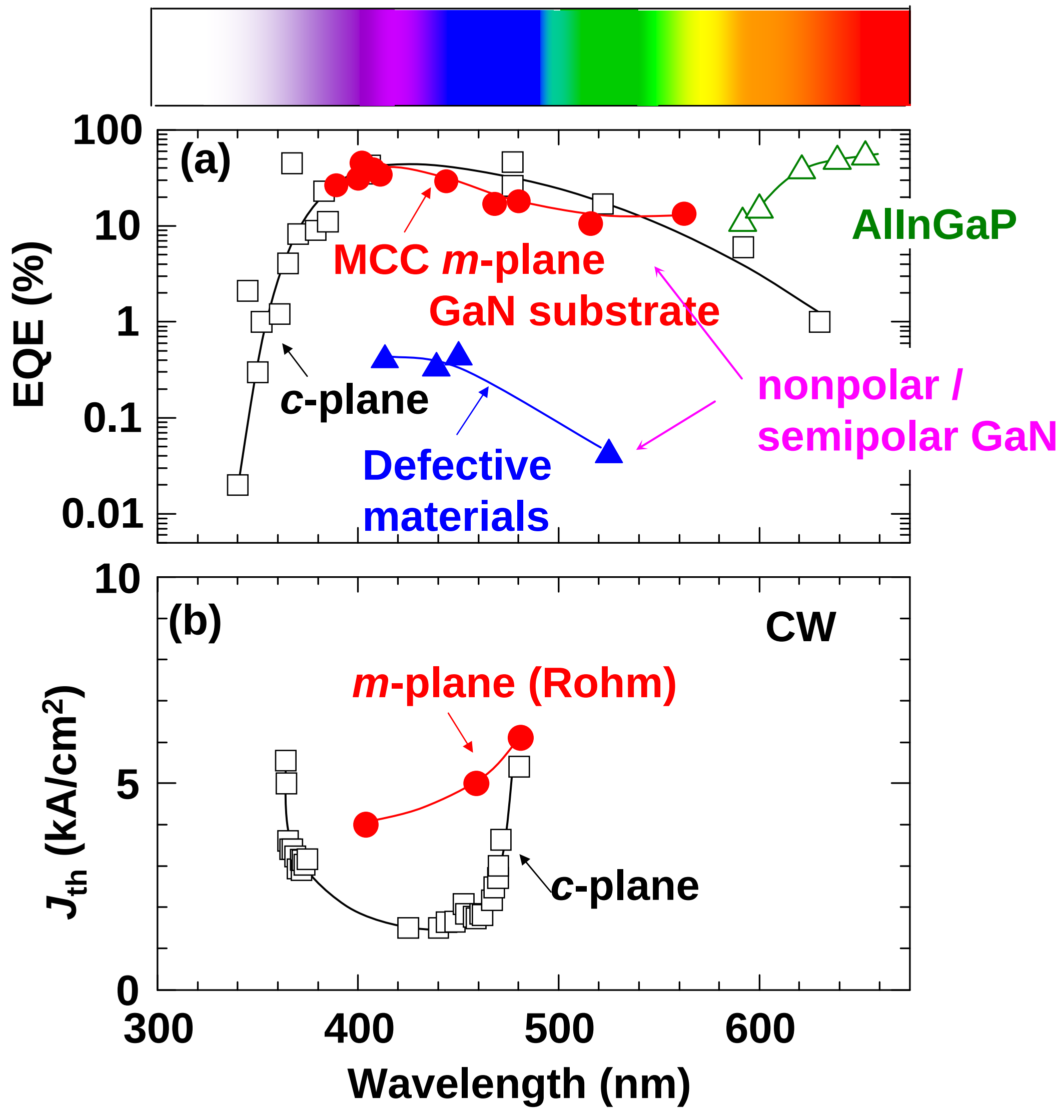
Summary of Device Characteristics

EQE of LEDs

Threshold current density of LDs



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Summary

√ **Planar heteroepitaxial nonpolar GaN / TD and SF problems unavoidable.**
√ **LEO does not work finely for nonpolar epitaxy**

√ **Advantages of nonpolar orientations confirmed (no QCSE, short radiative lifetime / polarized emission).**

√ ***m*-plane growths on free-standing GaN substrates**
TD and SF densities -- nearly equal to the underlayer value
Reasonable IQE as bulk InGaN films (11% for x=0.06) -- similar to *c*-plane
Improved radiative lifetime (at room temperature)

√ ***m*-plane LEDs and LDs on free-standing GaN substrates**
Coming up to overtake *c*-plane devices.