

# Optical spectroscopy



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« Nanophysique et semiconducteurs » Group



# Goal

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## Optical spectroscopy techniques

- ⇒ Some notions
  - ⇒ QWs
- ⇒ Examples from literature (III-N mainly)

## GaN peculiarities

- ⇒ Quantum Confined Stark Effect
  - ⇒ Localization...

Books on the topic:

Semiconductor Optics, Springer  
(*C. F. Klingshirn*)

Building Electro-Optical Systems : Making it all work, Wiley  
*P. C. D. Hobbs*

$$\lambda = \frac{c}{\nu} = \frac{hc}{E} = \frac{1}{\tilde{\nu}}$$

$$\lambda = 1 \mu m$$

$$\Leftrightarrow$$

$$\nu = 3.10^{14} Hz$$

$$\Leftrightarrow$$

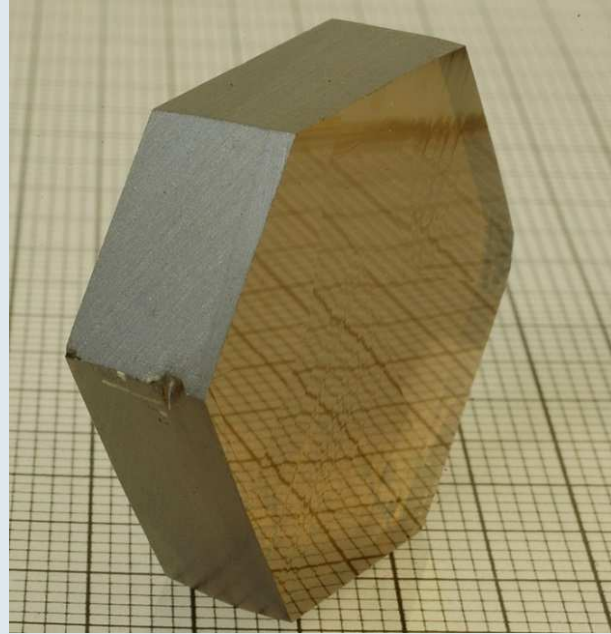
$$E = 1.24 eV$$

$$\Leftrightarrow$$

$$\tilde{\nu} = 10^4 cm^{-1}$$

# Spectroscopy of GaN

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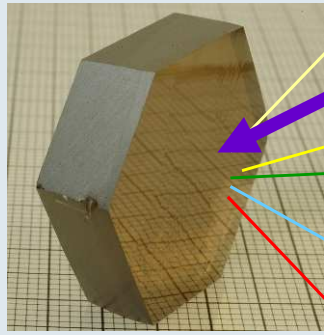
E&M, eanmint.co.jp

I have a piece of GaN and want to optically probe its optical properties...



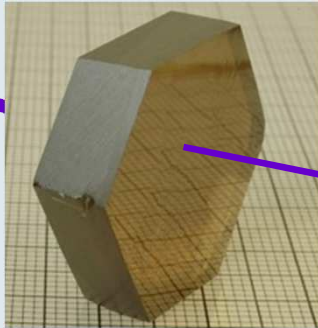
Another wide band gap semiconductor

# Spectroscopy of GaN



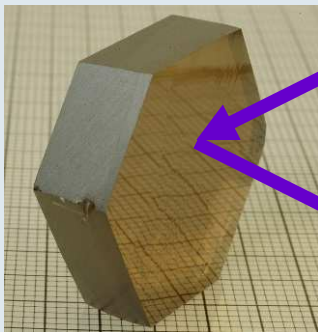
*Spectrometer  
+  
Detector*

Photoluminescence



*Spectrometer  
+  
Detector*

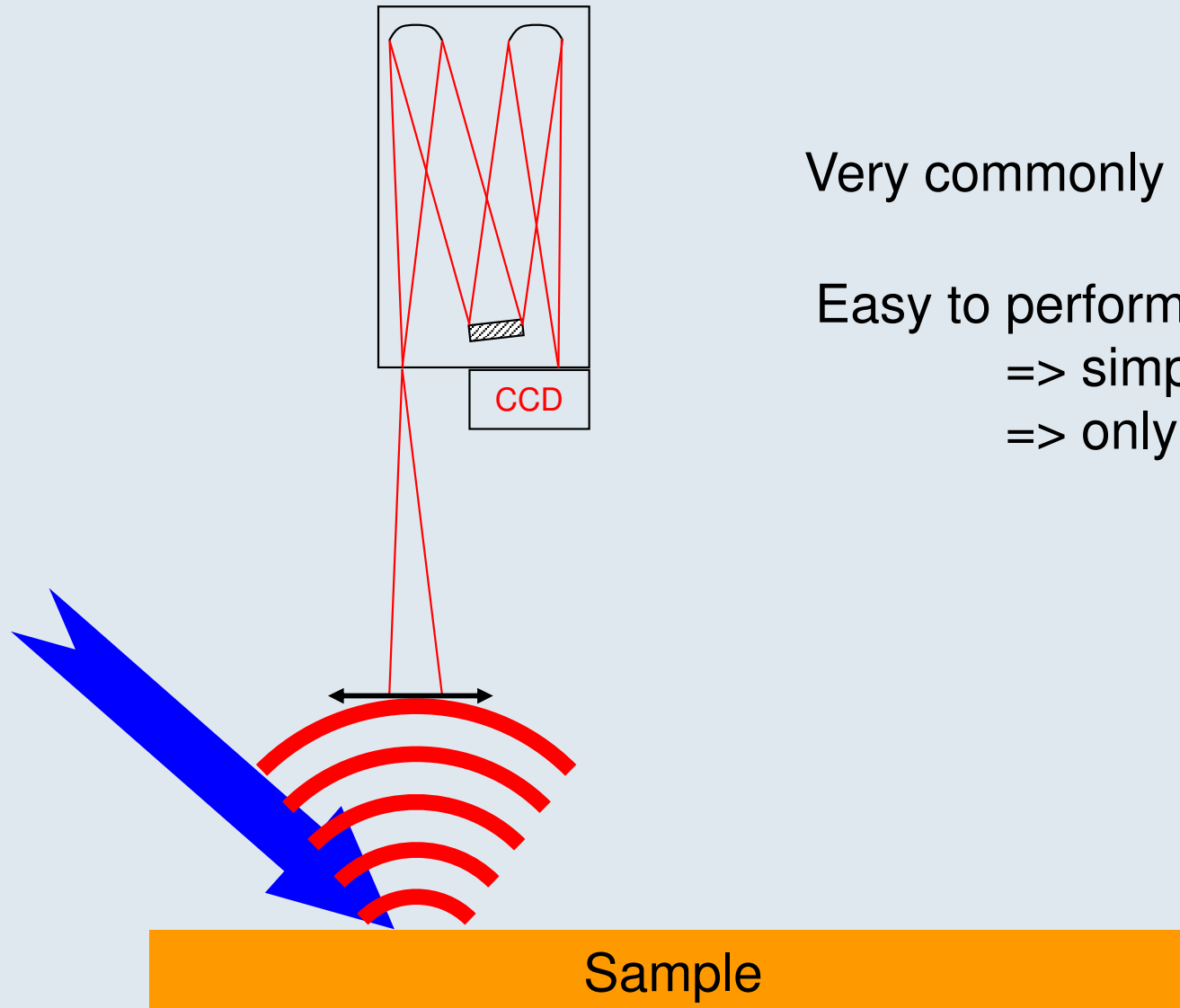
Absorption



*Spectrometer  
+  
Detector*

Reflectivity

# Spectroscopy of GaN : photoluminescence



Very commonly used

Easy to perform

=> simple set-up

=> only PL signal

# The grating spectrometer

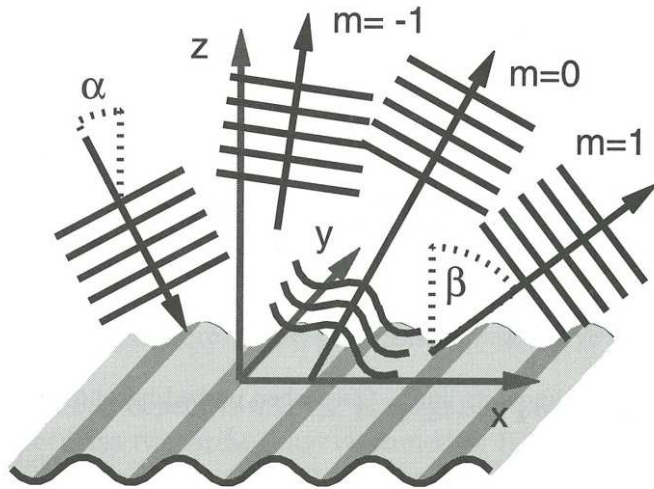


Figure 7.1 Plane diffraction grating.

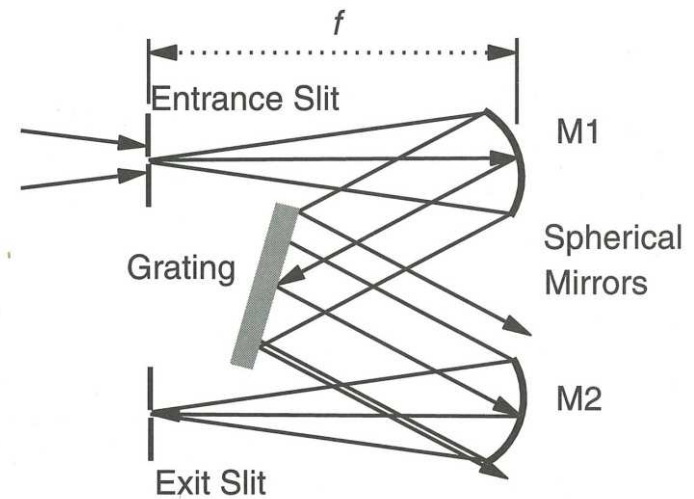
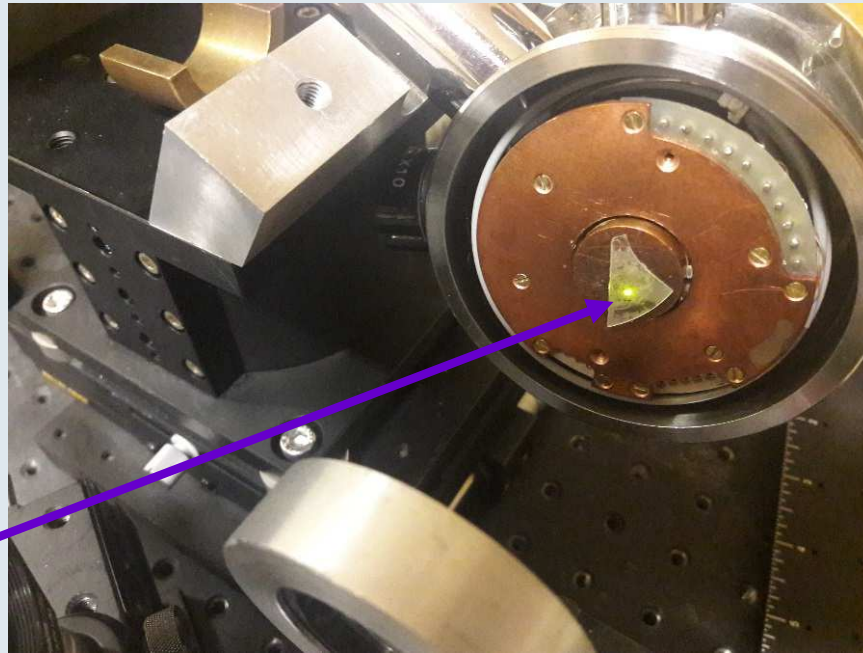


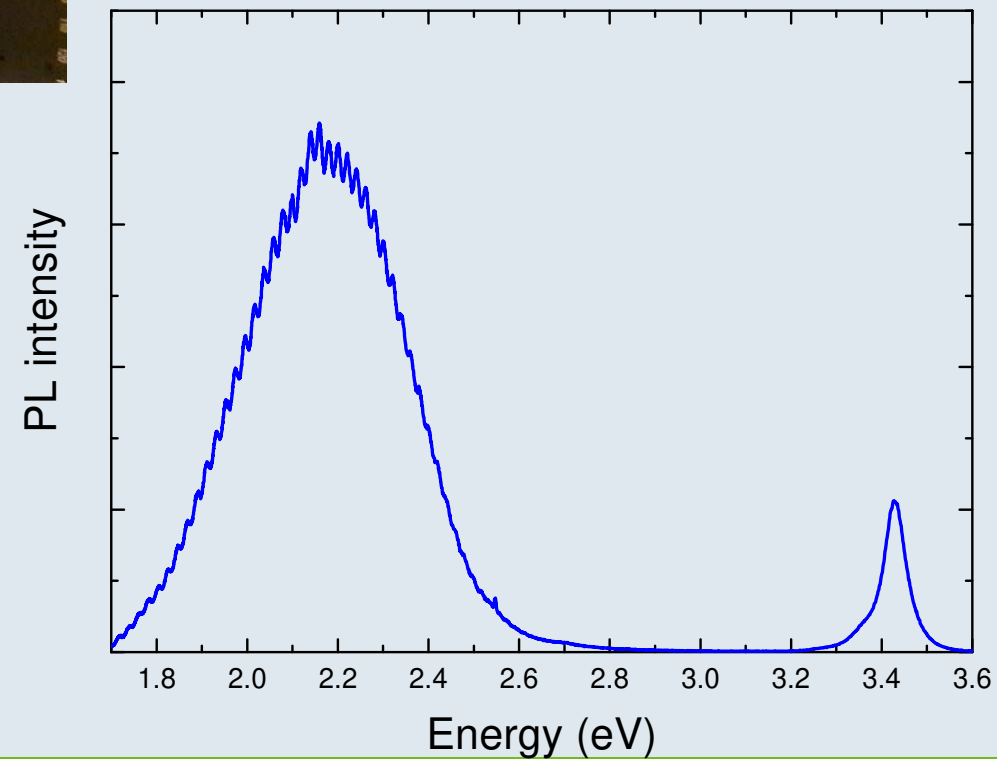
Figure 7.2 Czerny-Turner monochromator.

# Spectroscopy of GaN : photoluminescence

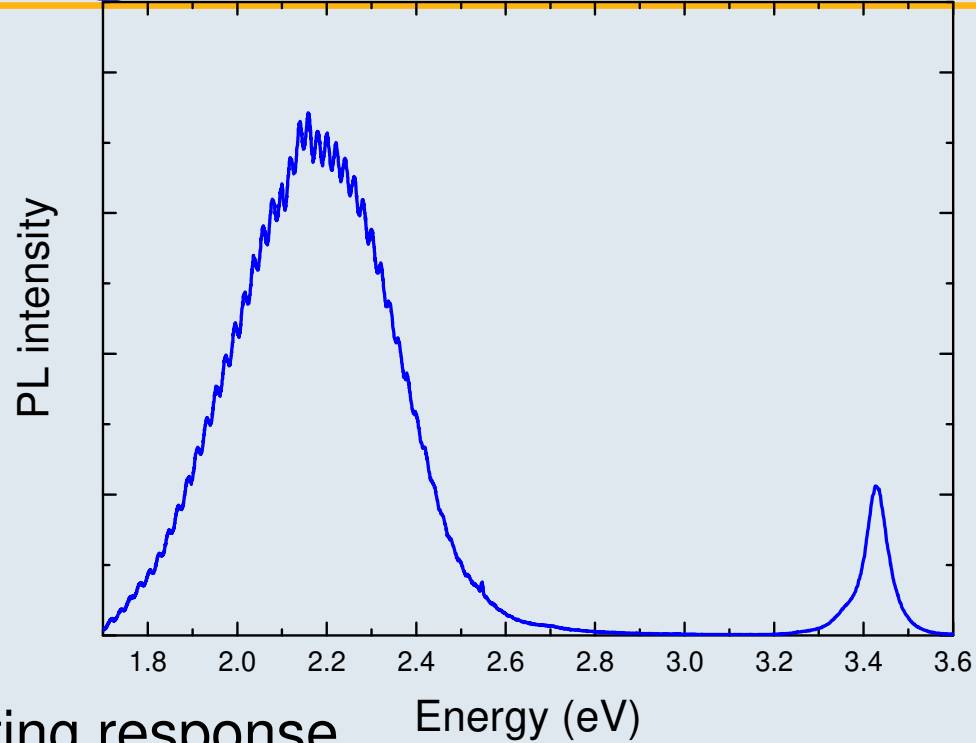


244 nm

300 K

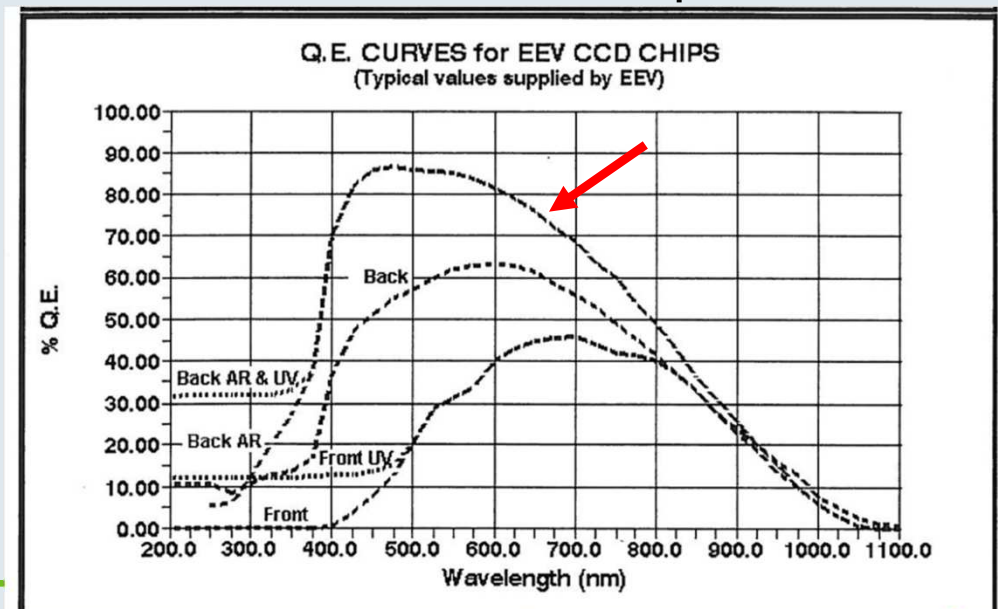
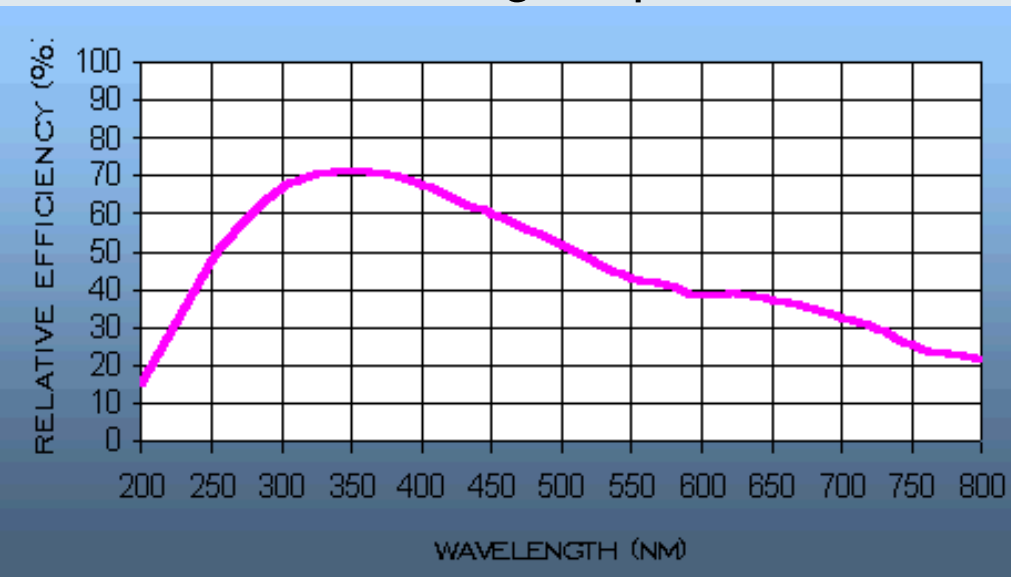


# Comparing intensities...



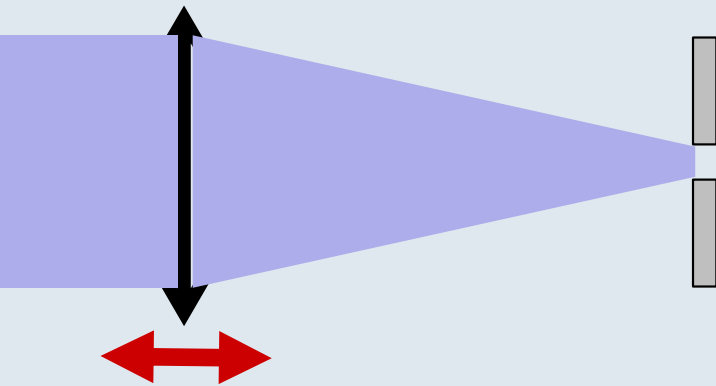
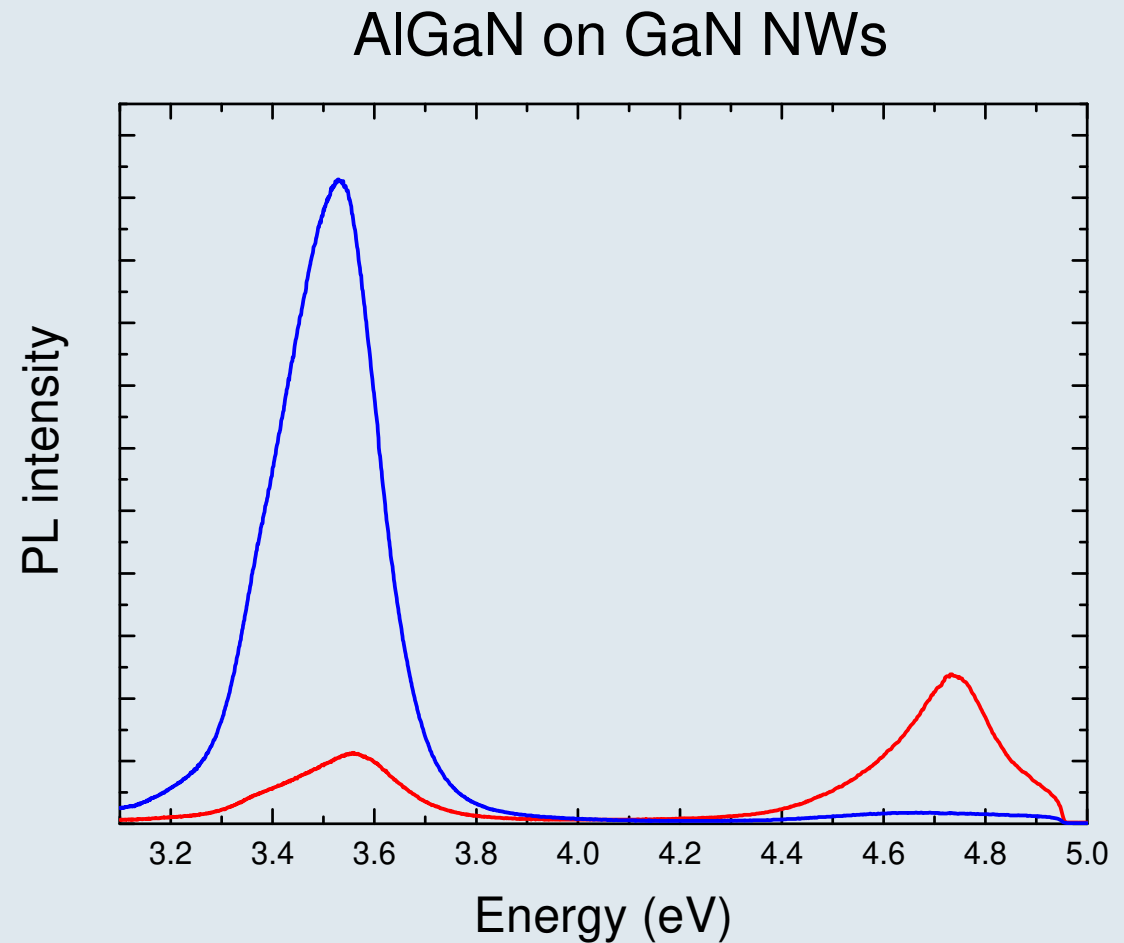
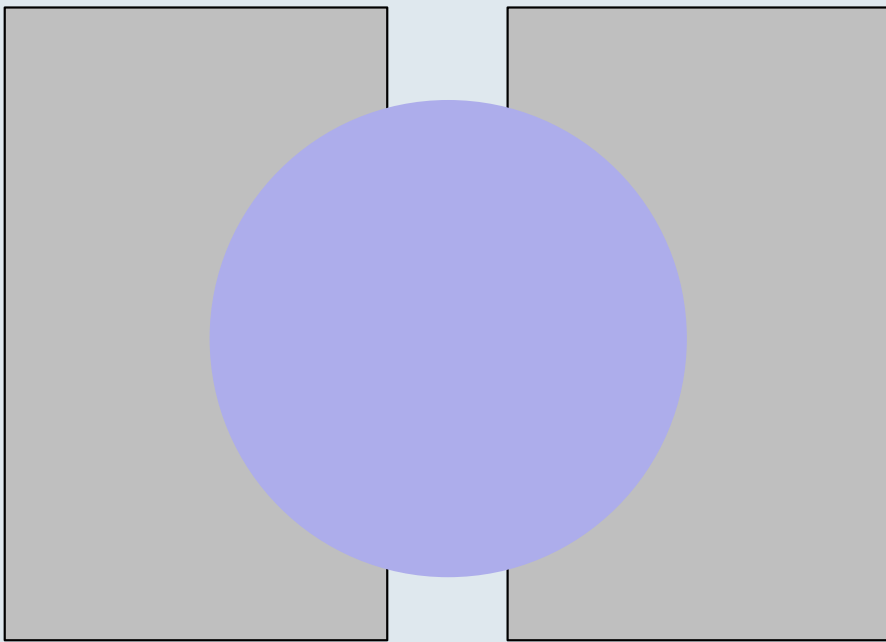
Grating response

Detector response



# Comparing intensities...

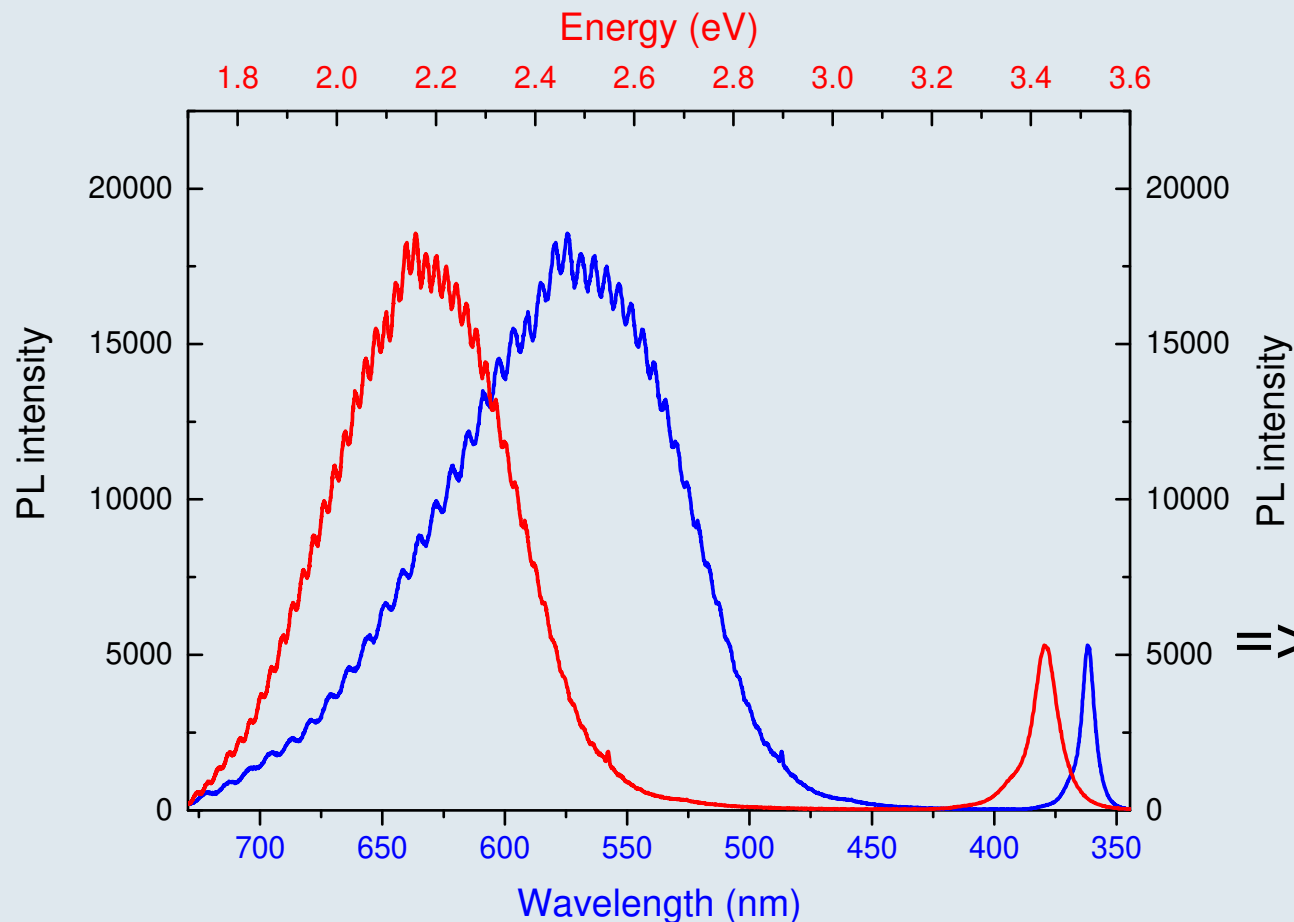
100  $\mu\text{m}$



⇒ For wide spectral ranges (especially in UV)  
off-axis parabolic mirrors are recommended !

# Comparing intensities...

$$\lambda(nm) = \frac{hc}{E} = \frac{1240}{E(eV)}$$



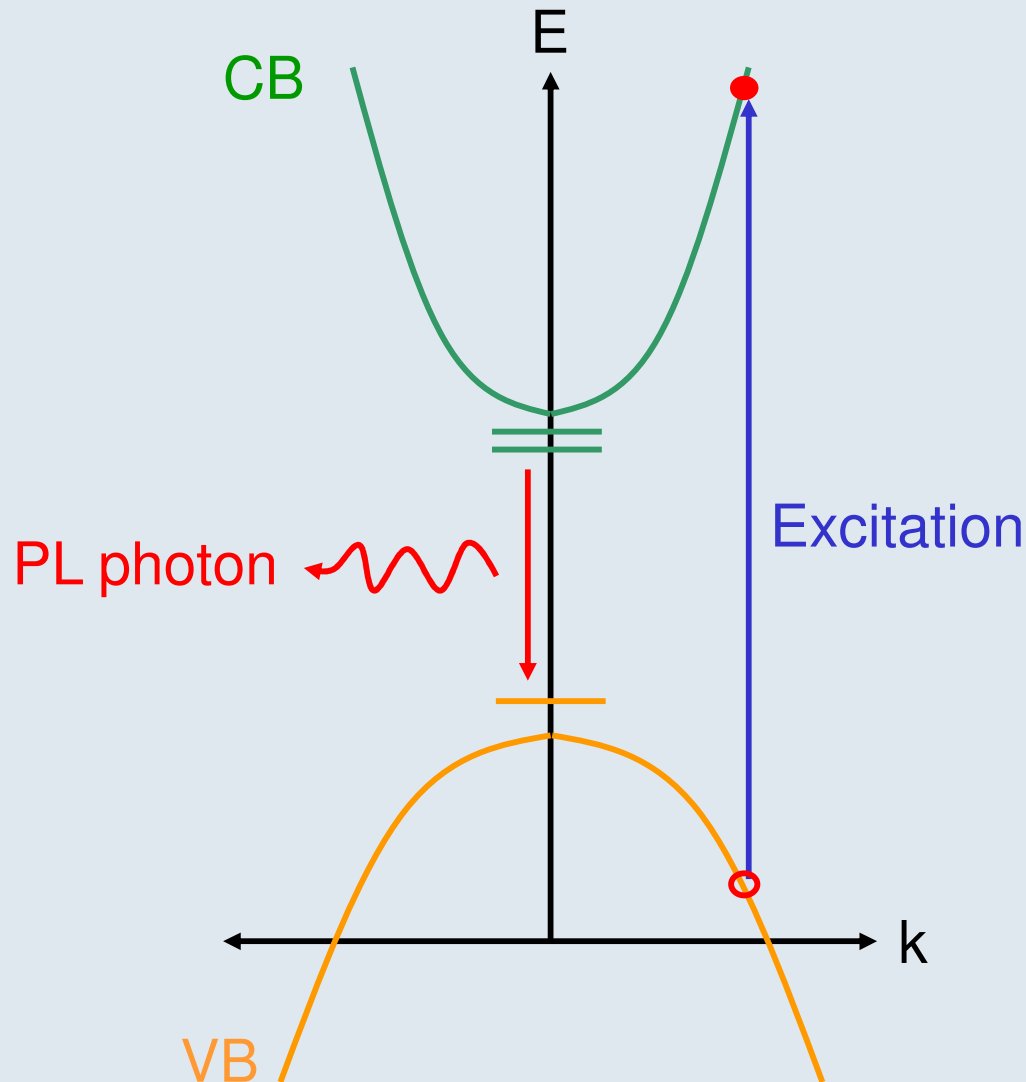
$$d\lambda \propto \frac{dE}{E^2}$$

=> Jacobian transformation

J. Mooney *et al.*, *J. Phys. Chem. Lett.* **4** 3316 (2013)

# Spectroscopy of GaN : PL

What is probed in PL??



In general, relaxation is very fast  
=> much faster than radiative rates

=> PL probes the  
lowest available energy states

# Spectroscopy of GaN : PL

**PL PROBES THE LOWEST  
AVAILABLE ENERGY STATES**



# Spectroscopy of GaN : photoluminescence

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## **Advantages of photoluminescence:**

Very versatile and easy to set-up technique

Usually large (positive) signal

No issue with substrate

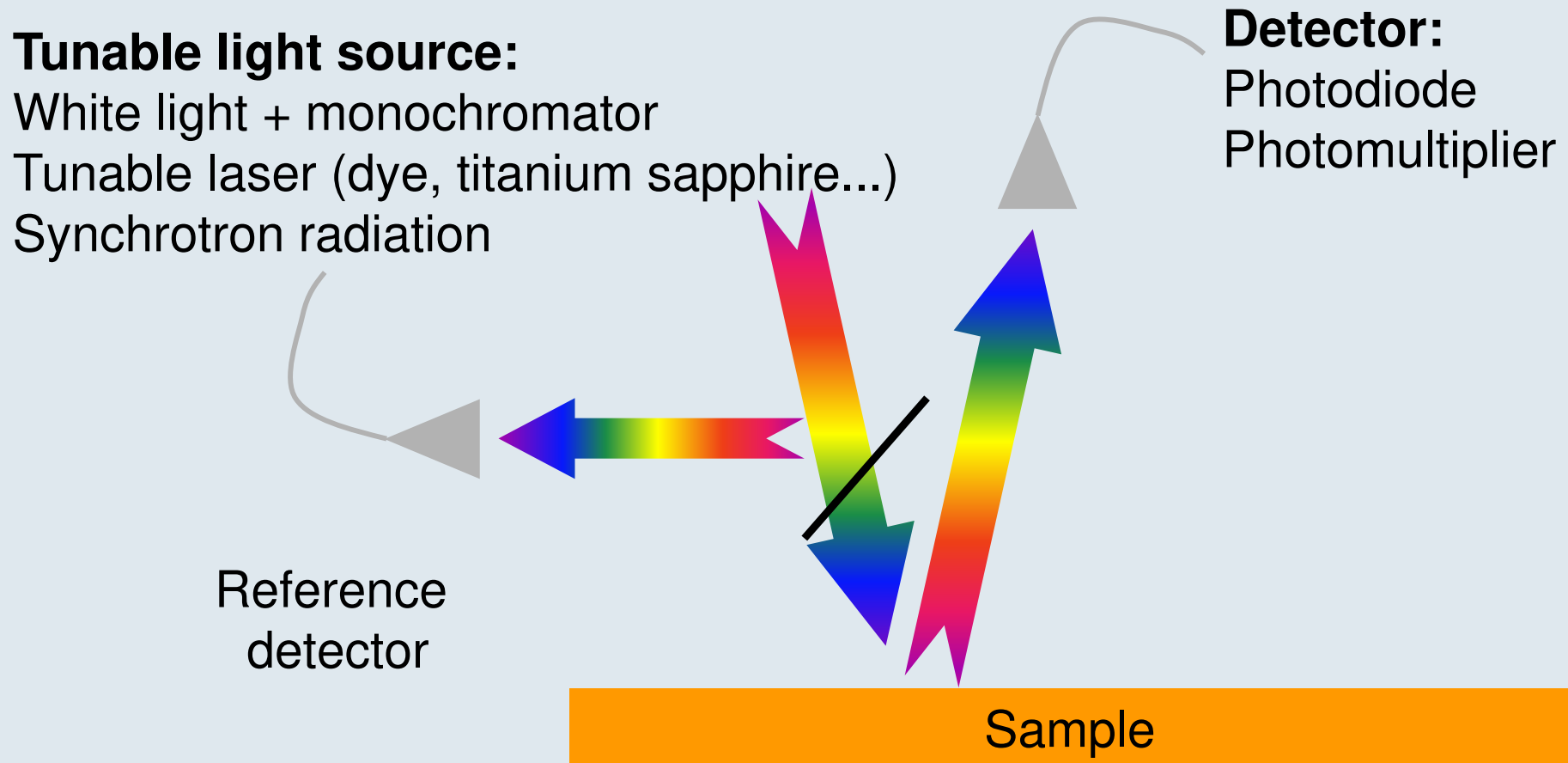
## **Limitations of photoluminescence:**

Only lowest energy state probed

Pump laser might affect the electronic properties

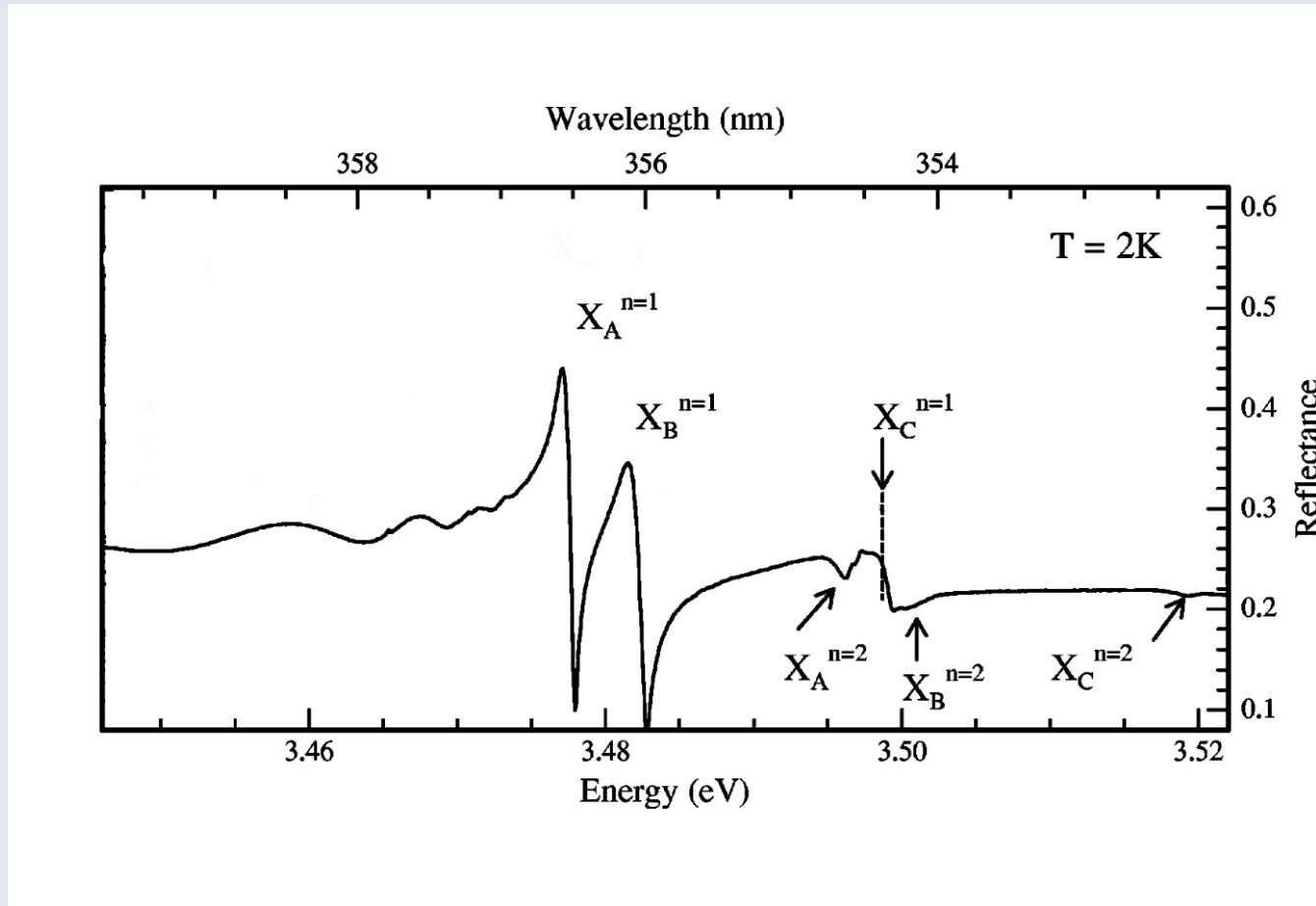
Data interpretation ?

# Spectroscopy of GaN : reflectivity



Often used in conjunction with modulation techniques  
=> what is really measured?

# Spectroscopy of GaN : reflectivity



1.5  $\mu\text{m}$  GaN on bulk GaN substrate

Kornitzer *et al.* PRB **60** 1471 (1999)

# Spectroscopy of GaN : reflectivity

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## **Advantages of reflectivity:**

Probing of the electronic states (coupled to the light!)

Measurement does not perturb the system at low power

No issue with substrate

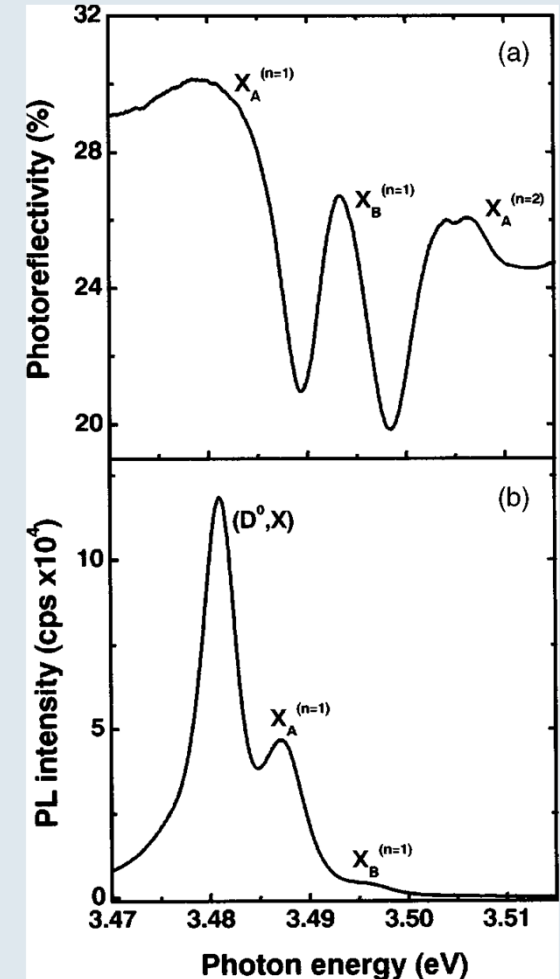
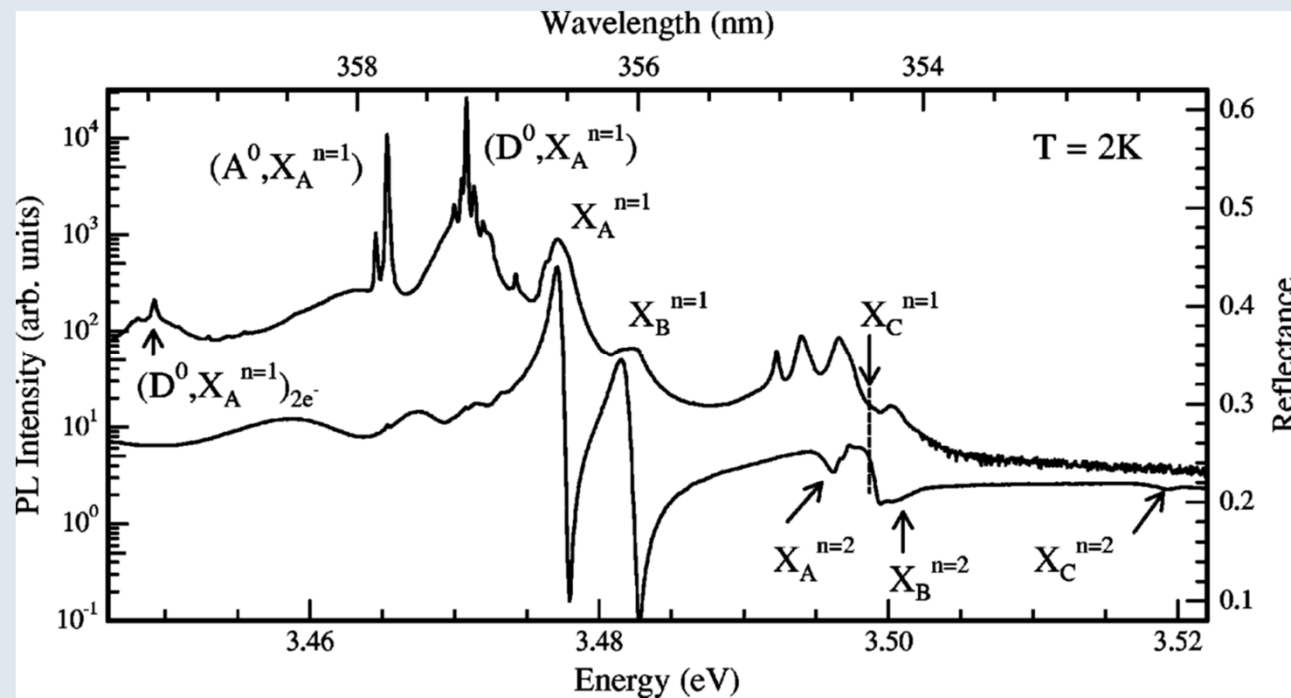
## **Limitations of reflectivity:**

Weak signal (especially if large broadening)

Complex data interpretation

# Spectroscopy of GaN : reflectivity vs PL

10 K  
1.3  $\mu\text{m}$  GaN on sapphire

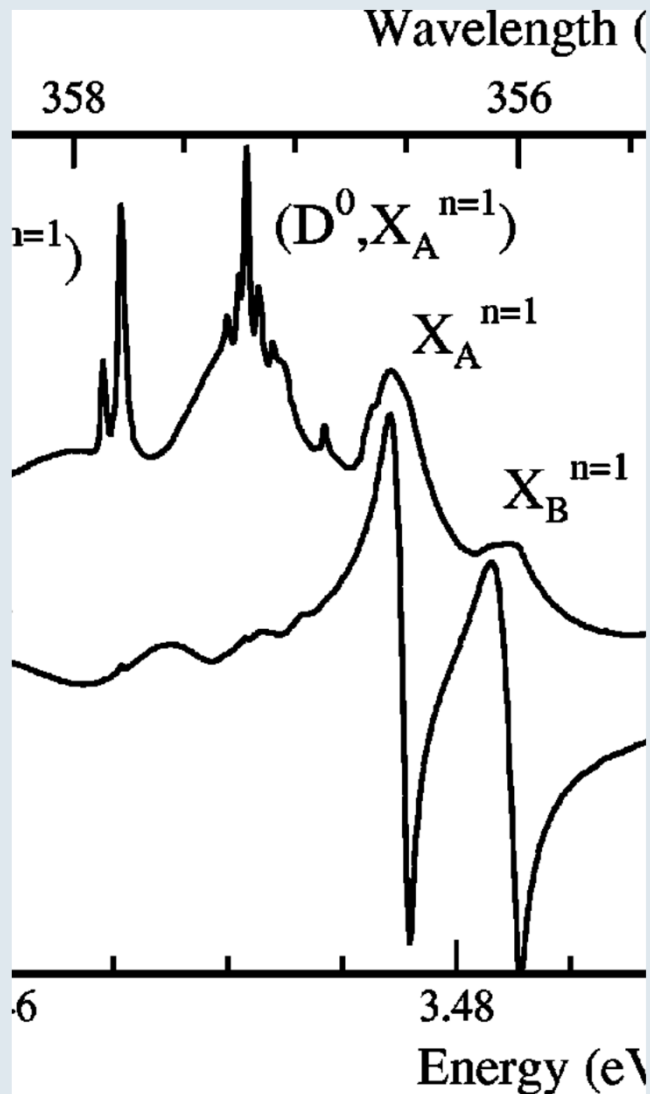


Ursaki *et al.* **JAP** 94 4813 (2003)

1.5  $\mu\text{m}$  GaN on bulk GaN substrate

Kornitzer *et al.* PRB **60** 1471 (1999)

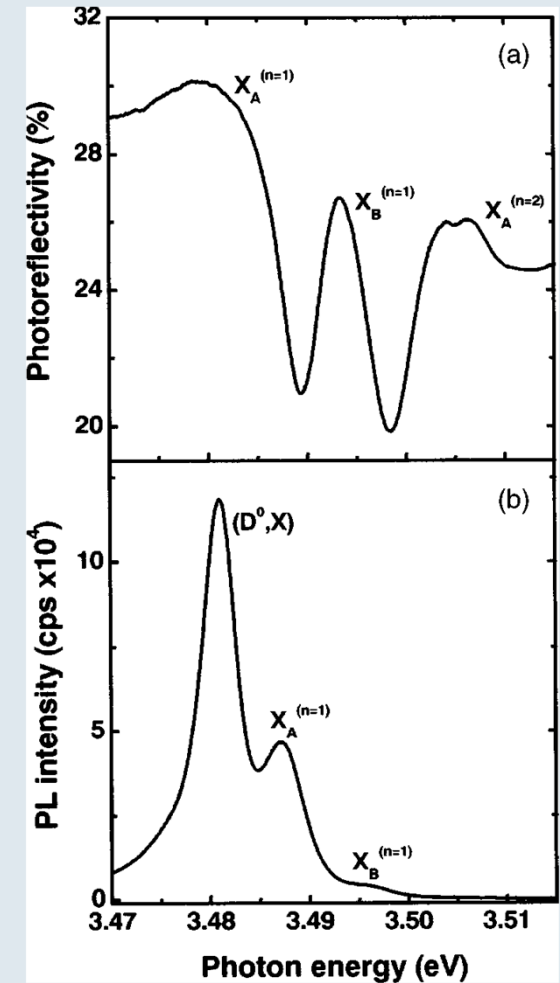
# Spectroscopy of GaN : peak position



1.5  $\mu\text{m}$  GaN on bulk GaN substrate

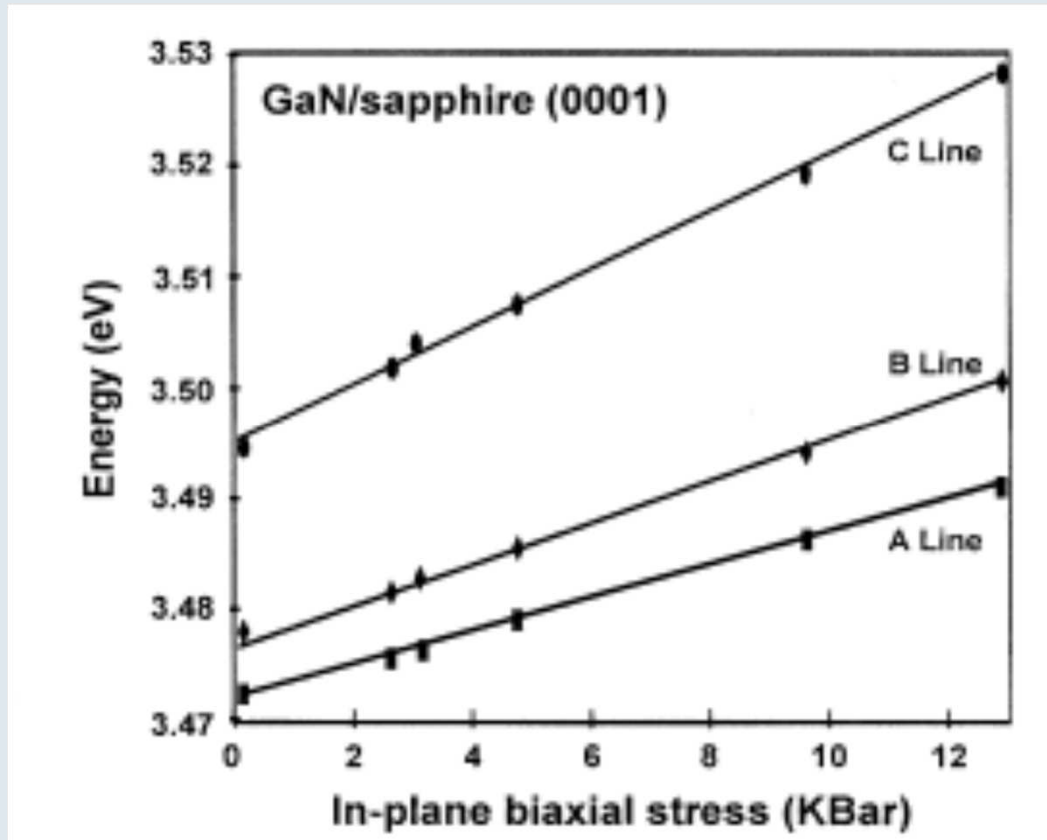
Kornitzer *et al.* PRB **60** 1471 (1999)

10 K  
1.3  $\mu\text{m}$  GaN on sapphire



Ursaki *et al.* JAP **94** 4813 (2003)

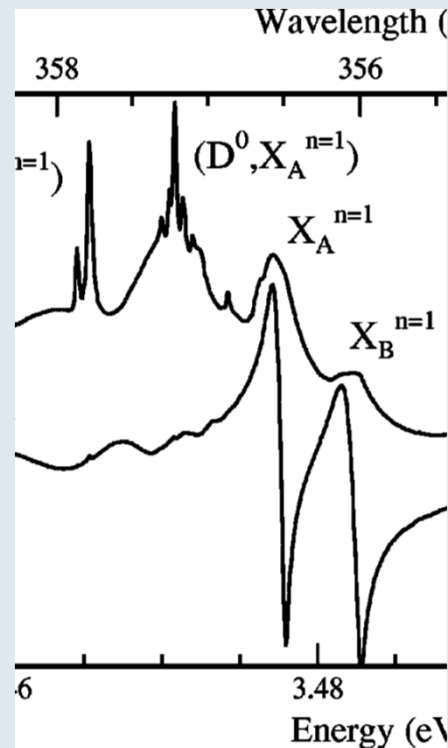
# Spectroscopy of GaN : strain dependence



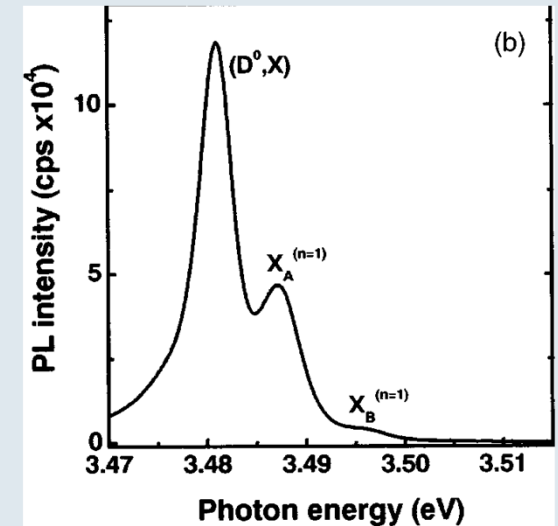
The « Bernard Gil » plot

Gil *et al.*, PRB **52** R17028 (1995)

# Spectroscopy of GaN : broadening



100  $\mu\text{eV}$



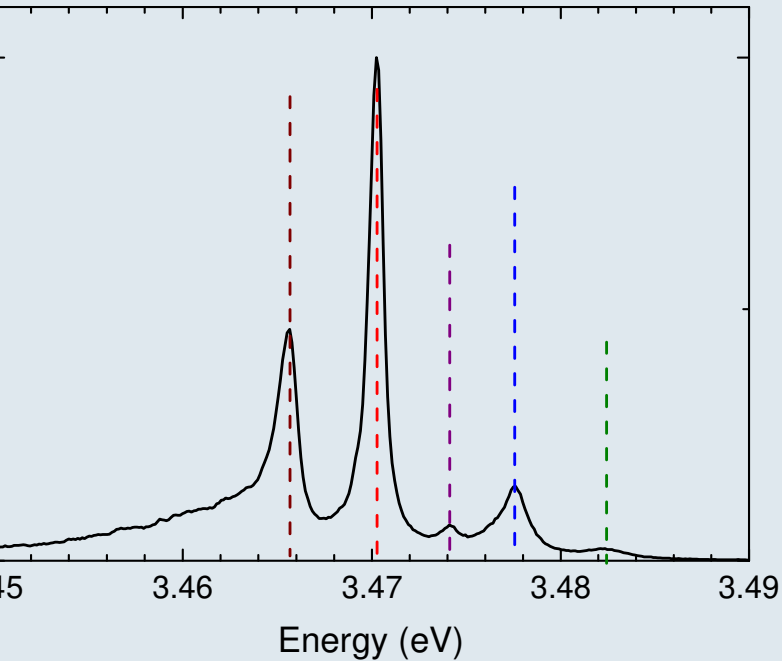
5 meV

Temperature ( $kT@ 2K = 0.15 \text{ meV}$ )

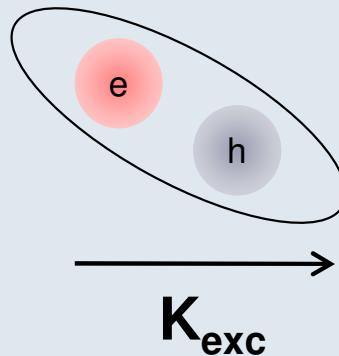
Strain inhomogeneity (due to extended defects, point defects....)

Lifetime ( $\Delta E \Delta t > \hbar/2$ )

# Excitons, what are they?



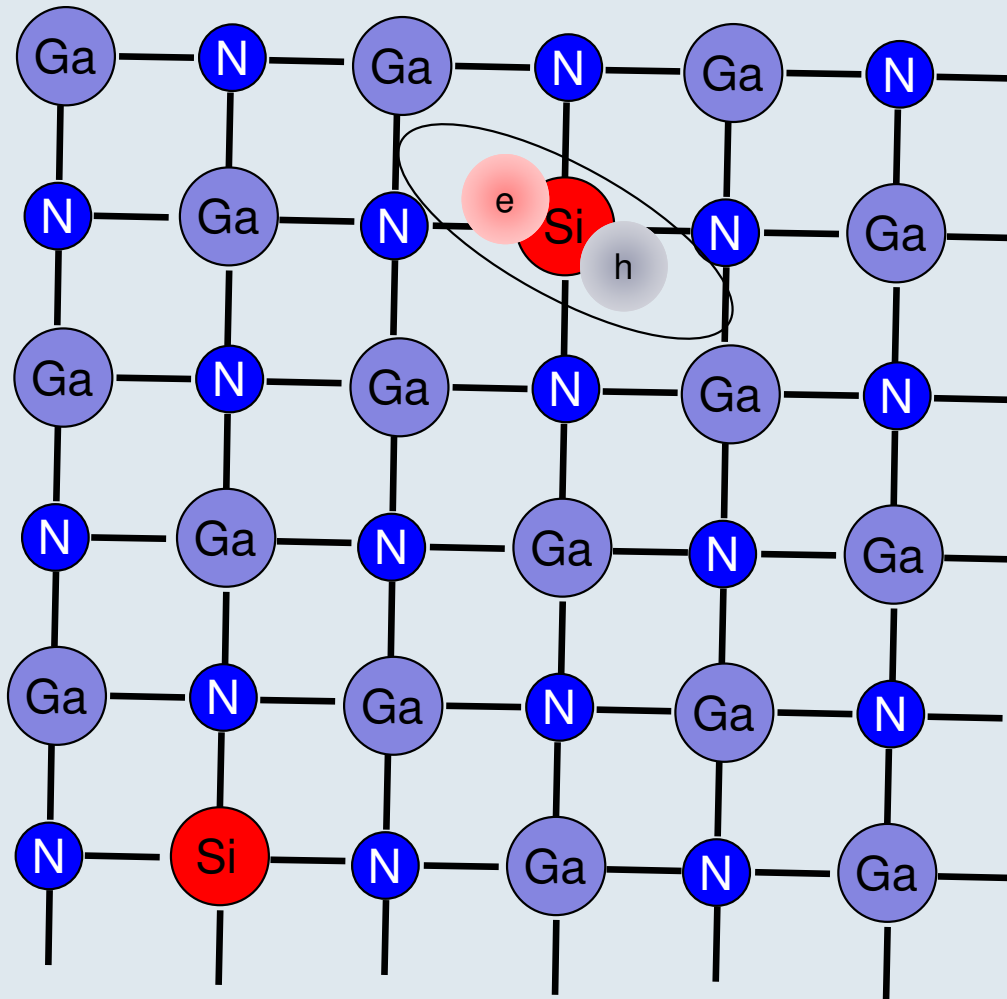
One electron + one hole  
bound by Coulomb interaction  
=> Hydrogen-like particle



|      | $a$ (nm) | $E_x$ (meV) |
|------|----------|-------------|
| GaN  | 2.8      | 25          |
| GaAs | 12       | 4.8         |

$kT$  @ 300K  
25 meV

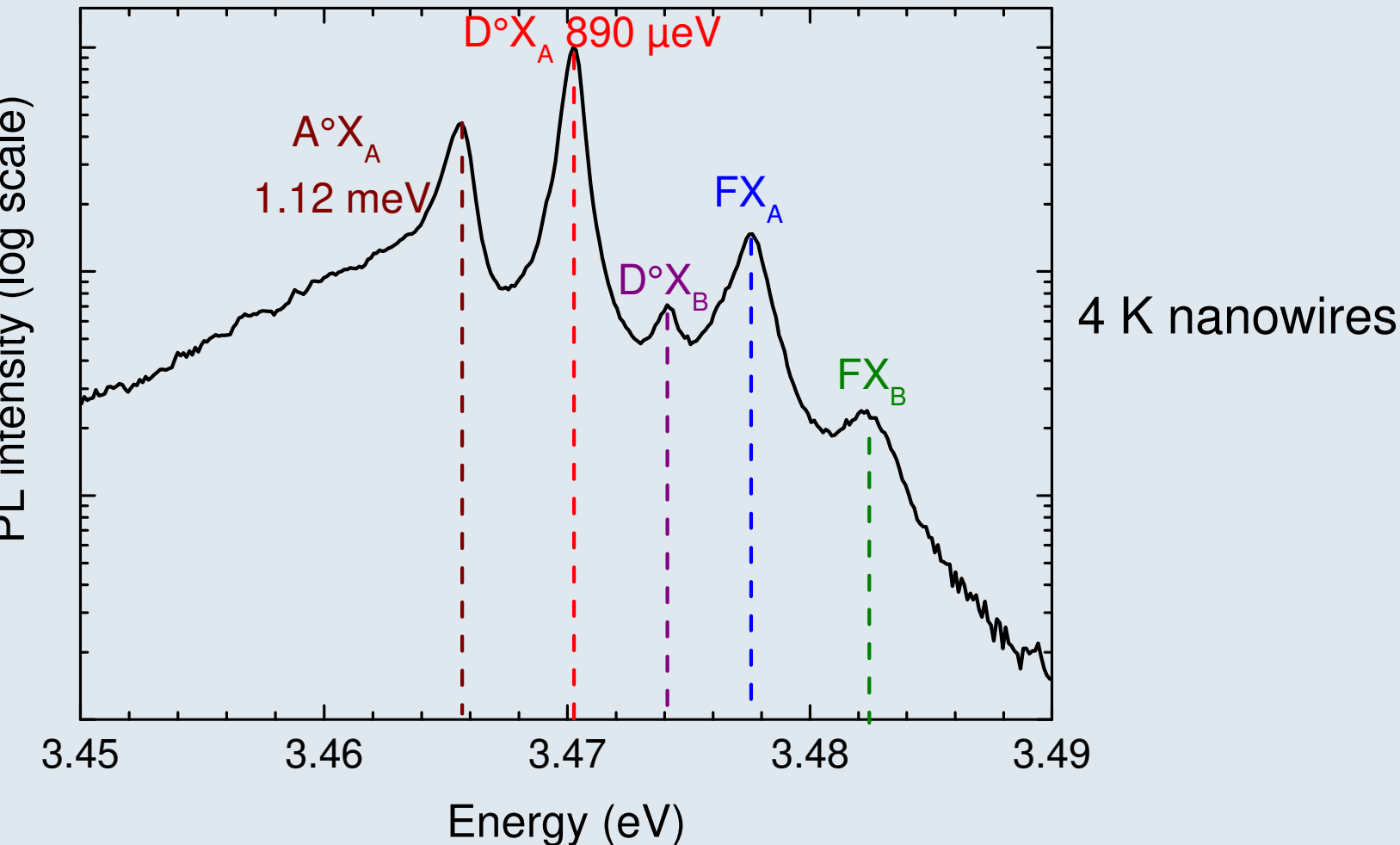
# Excitons, what are they?



|                                      |                                        |                                       |                                        |                                        |                                       |                                     |
|--------------------------------------|----------------------------------------|---------------------------------------|----------------------------------------|----------------------------------------|---------------------------------------|-------------------------------------|
|                                      |                                        |                                       |                                        |                                        |                                       | helium<br>2<br><b>He</b><br>4.0026  |
|                                      | boron<br>5<br><b>B</b><br>10.811       | carbon<br>6<br><b>C</b><br>12.011     | nitrogen<br>7<br><b>N</b><br>14.007    | oxygen<br>8<br><b>O</b><br>15.999      | fluorine<br>9<br><b>F</b><br>18.998   | neon<br>10<br><b>Ne</b><br>20.180   |
|                                      | aluminium<br>13<br><b>Al</b><br>26.982 | silicon<br>14<br><b>Si</b><br>28.086  | phosphorus<br>15<br><b>P</b><br>30.974 | sulfur<br>16<br><b>S</b><br>32.065     | chlorine<br>17<br><b>Cl</b><br>35.453 | argon<br>18<br><b>Ar</b><br>39.948  |
| zinc<br>30<br><b>Zn</b><br>65.39     | gallium<br>31<br><b>Ga</b><br>69.723   | germanium<br>32<br><b>Ge</b><br>72.61 | arsenic<br>33<br><b>As</b><br>74.922   | selenium<br>34<br><b>Se</b><br>78.96   | bromine<br>35<br><b>Br</b><br>79.904  | krypton<br>36<br><b>Kr</b><br>83.80 |
| cadmium<br>48<br><b>Cd</b><br>112.41 | indium<br>49<br><b>In</b><br>114.82    | tin<br>50<br><b>Sn</b><br>118.71      | antimony<br>51<br><b>Sb</b><br>121.76  | tellurium<br>52<br><b>Te</b><br>127.60 | iodine<br>53<br><b>I</b><br>126.90    | xenon<br>54<br><b>Xe</b><br>131.29  |
| mercury<br>80<br><b>Hg</b><br>200.59 | thallium<br>81<br><b>Tl</b><br>204.38  | lead<br>82<br><b>Pb</b><br>207.2      | bismuth<br>83<br><b>Bi</b><br>208.98   | polonium<br>84<br><b>Po</b><br>[209]   | astatine<br>85<br><b>At</b><br>[210]  | radon<br>86<br><b>Rn</b><br>[222]   |

Neutral donor bound exciton  $D_0X$

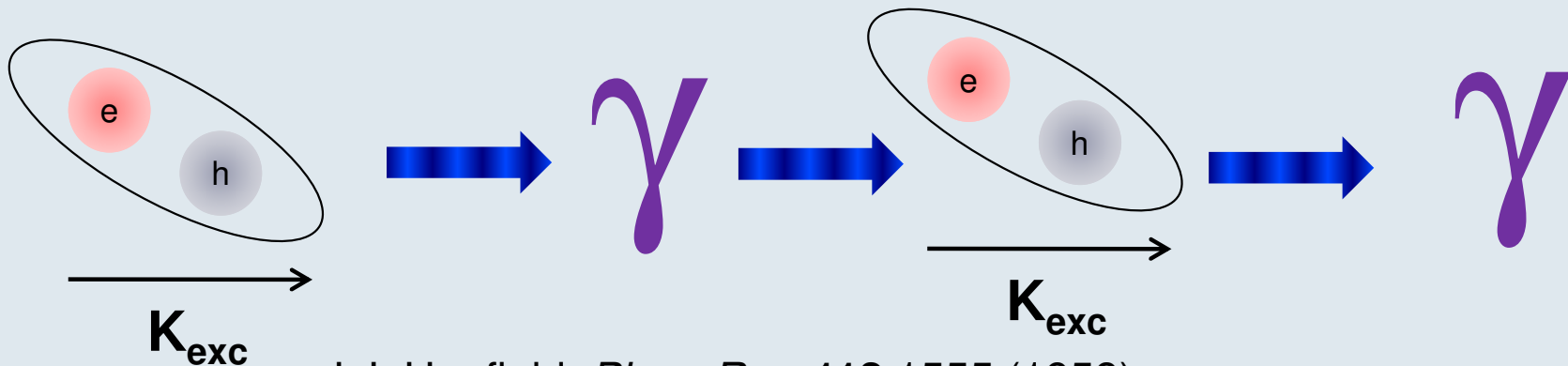
# Photoluminescence of bulk GaN



1 Si atom for  $10^7$  Ga atom ( $\sim 10$  per  $(100 \text{ nm})^3$ ) is enough to have a low temperature PL dominated by neutral donor bound exciton

# Excitons and polaritons in GaN

*Does the exciton really exist?*



J.J. Hopfield, *Phys. Rev.* **112** 1555 (1958)  
+ Pekar, Agranovich...

PHYSICAL REVIEW

VOLUME 112, NUMBER 5

DECEMBER 1, 1958

## Theory of the Contribution of Excitons to the Complex Dielectric Constant of Crystals\*†

J. J. HOPFIELD†

*Physics Department, Cornell University, Ithaca, New York*

(Received July 16, 1958)

Polariton = mixed exciton-photon particles

J.J. Hopfield, *Annu. Rev. Condens. Matter Phys.* Vol. 5, 1 (2014)

# JJ Hopfield

*Very numerous papers on the theory of semiconductor optics...*

*... but his most famous paper is :*

*Proc. Natl. Acad. Sci. USA*  
Vol. 79, pp. 2554–2558, April 1982  
Biophysics

## **Neural networks and physical systems with emergent collective computational abilities**

(associative memory/parallel processing/categorization/content-addressable memory/fail-soft devices)

J. J. HOPFIELD

Division of Chemistry and Biology, California Institute of Technology, Pasadena, California 91125; and Bell Laboratories, Murray Hill, New Jersey 07974

### John J. Hopfield Facts



© Nobel Prize Outreach.  
Photo: Nanaka Adachi

John J. Hopfield  
Nobel Prize in Physics 2024

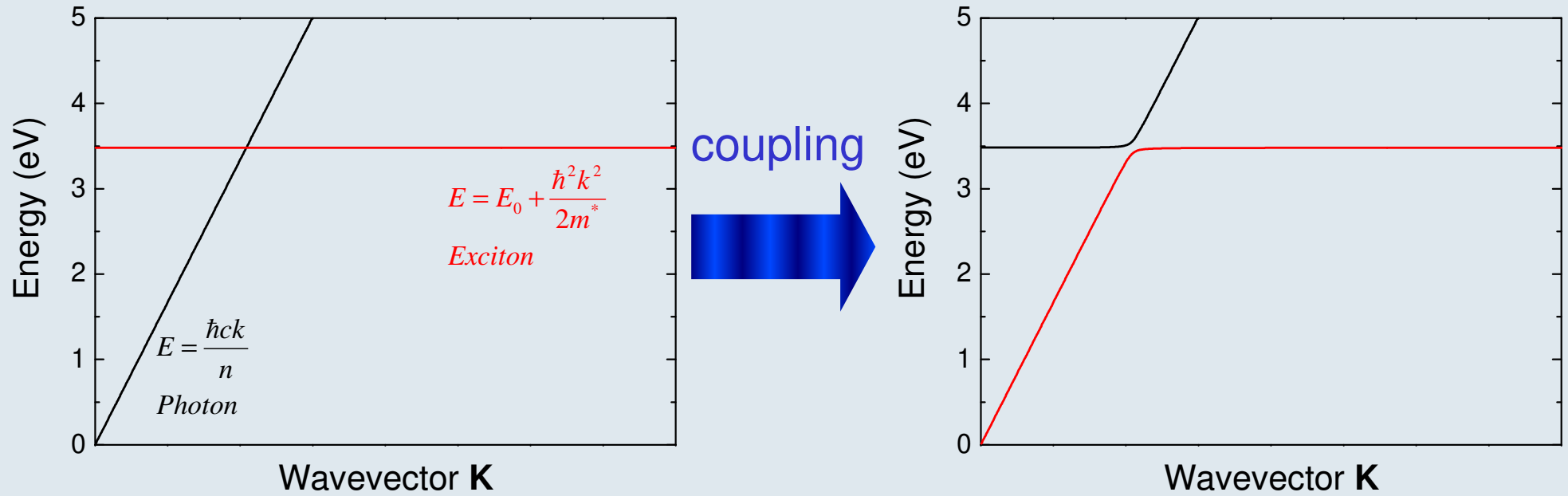
Born: 15 July 1933, Chicago, IL, USA

Affiliation at the time of the award: Princeton University,  
Princeton, NJ, USA

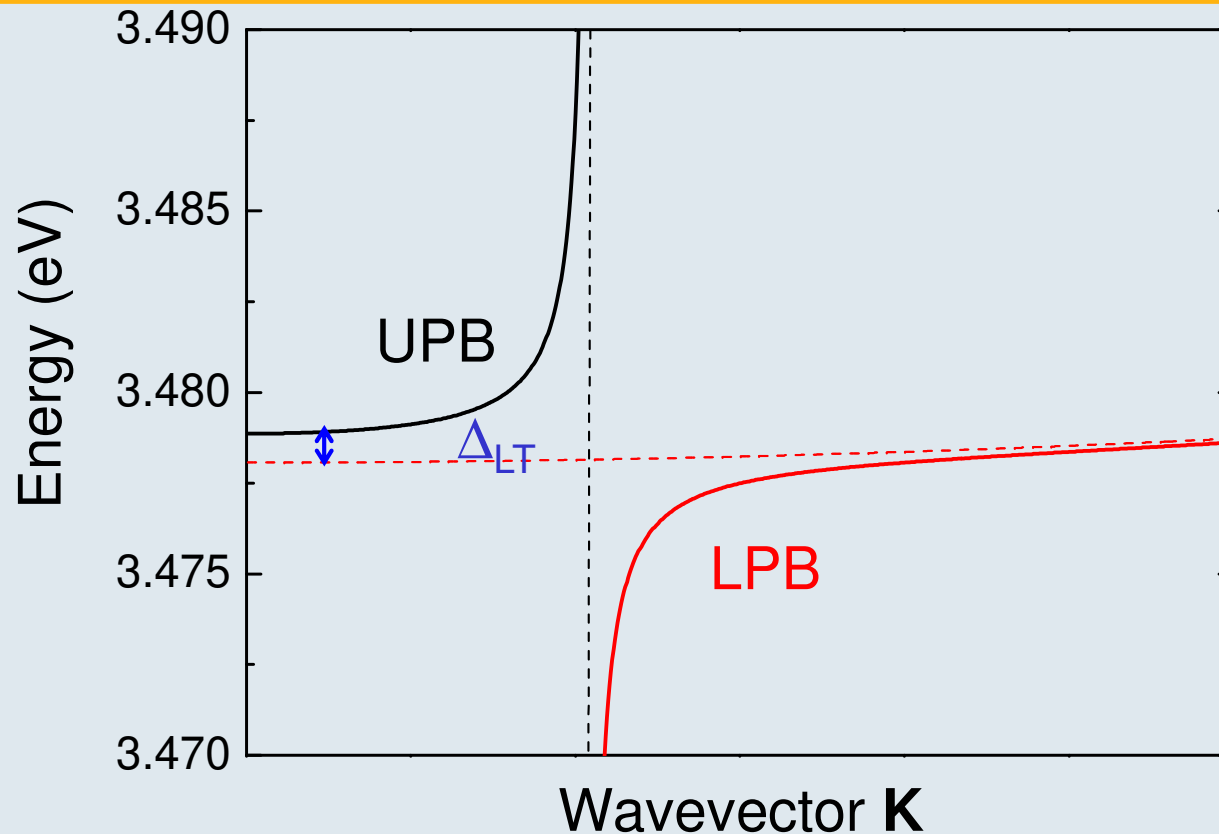
Prize motivation: “for foundational discoveries and  
inventions that enable machine learning with artificial  
neural networks”

Prize share: 1/2

# Excitons and polaritons in GaN



# Excitons and polaritons in GaN



$\Delta_{LT}$  measures the intrinsic light-exciton coupling in the material

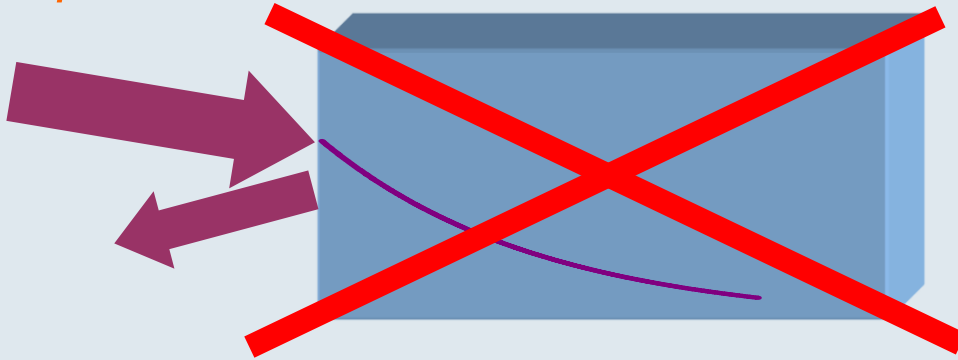
Note : in WZ GaN : A, B and C excitons + anisotropy

GaN :  $\Delta_{LT}(A)=0.8$  meV

GaAs :  $\Delta_{LT}=0.08$  meV

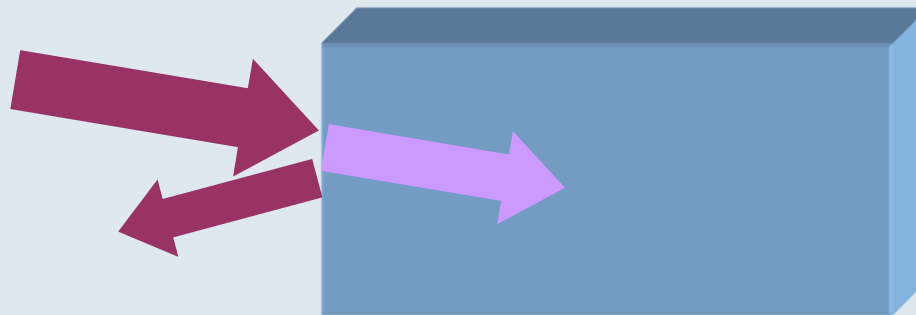
# Consequences of the polariton picture

Absorption



Beer-Lambert law

$$I = I_0 e^{-\alpha L}$$

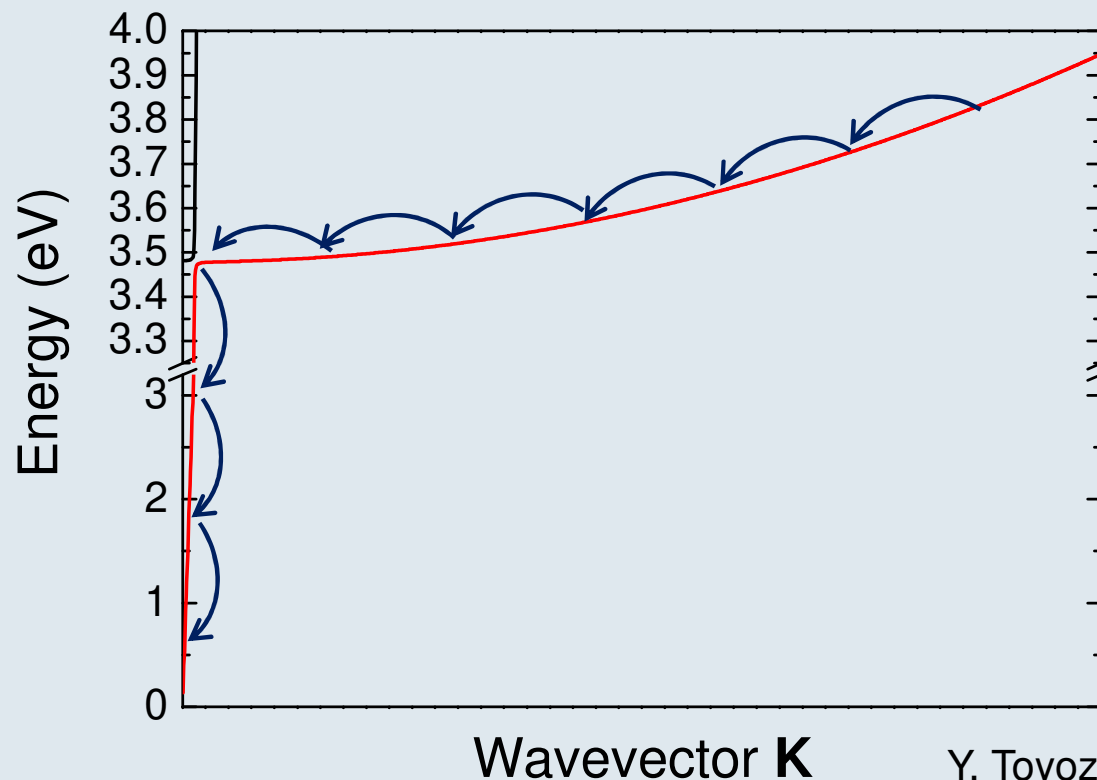
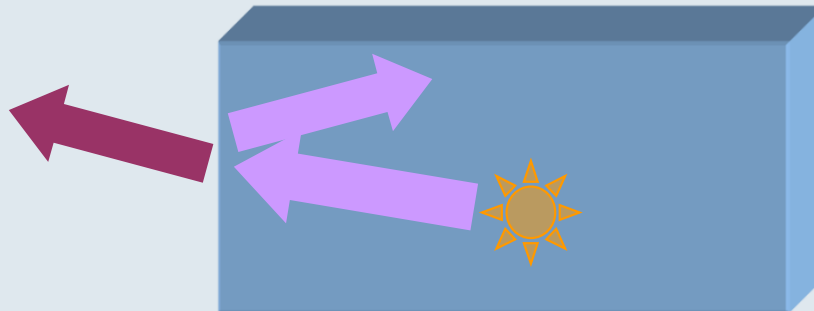


Propagating polariton  
(attenuation = scattering)

# Consequences of the polariton picture

## Luminescence

Polariton propagating to the surface !



Infrared catastrophe?

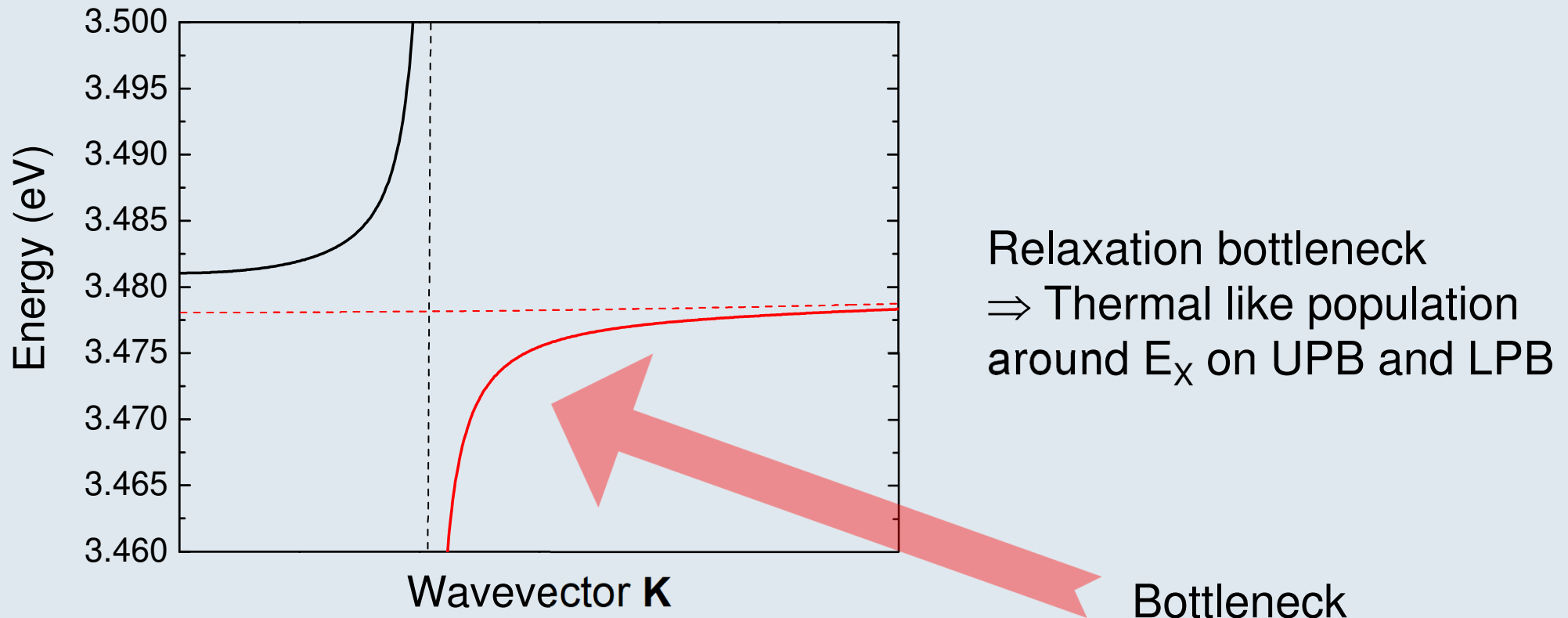
J. J. Hopfield, J. Phys. Soc. Jap. **21** 77 (1966)

Relaxation bottleneck  
⇒ Thermal like population

Y. Toyozawa, Prog. Theor. Phys. Suppl. **12** 111 (1959)

# Consequences of the polariton picture

## Luminescence



Luminescence properties are very complex to model !!  
(sample and experiments dependent...)

# Consequences of the polariton picture

## Luminescence

PHYSICAL REVIEW B

VOLUME 34, NUMBER 2

15 JULY 1986

### Effect of neutral donor scattering on the time-dependent exciton-polariton photoluminescence line shape in GaAs

T. Steiner\* and M. L. W. Thewalt

*Department of Physics, Simon Fraser University, Burnaby British Columbia, Canada V5A 1S6*

E. S. Koteles and J. P. Salerno

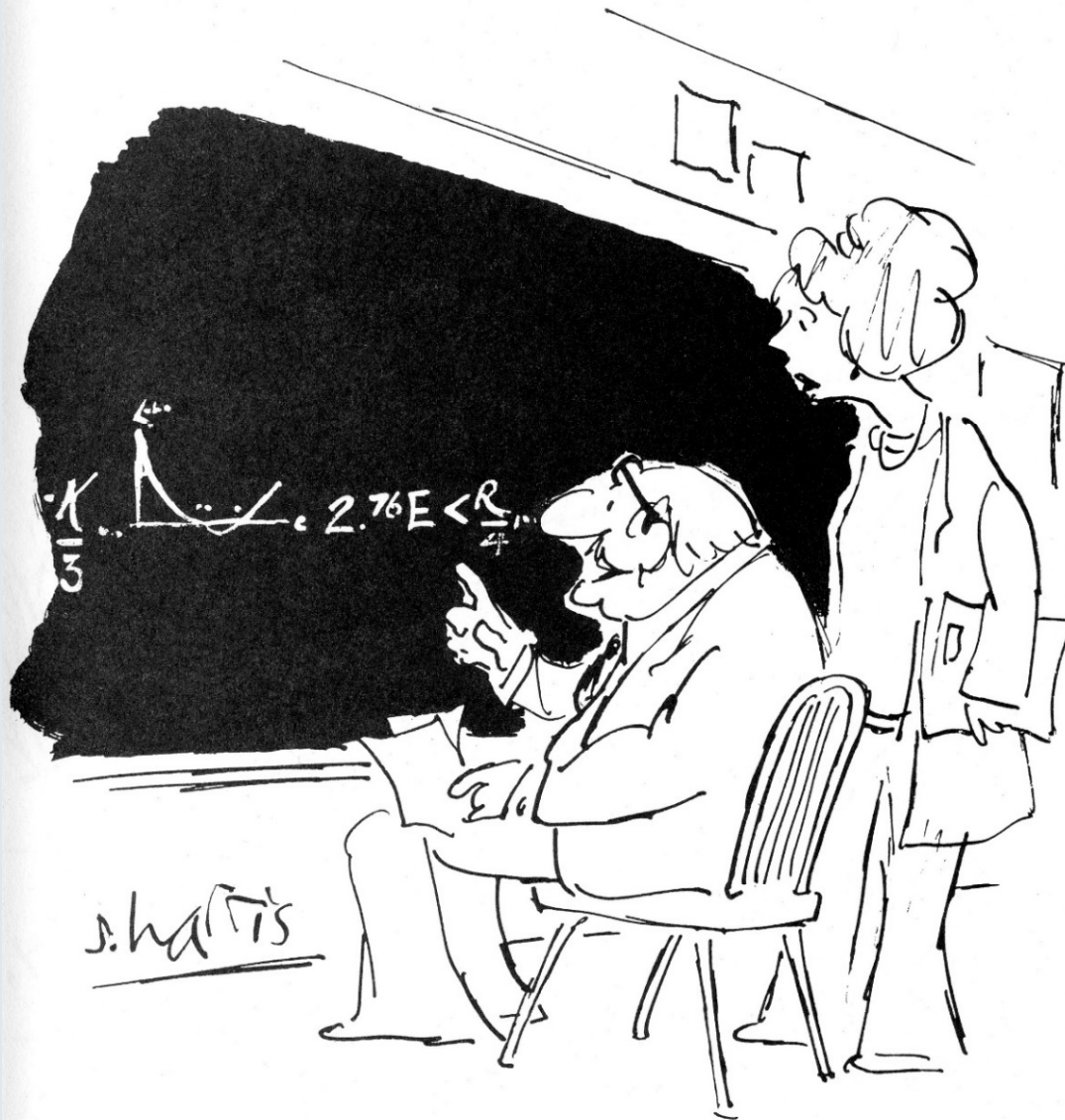
*GTE Laboratories, Inc., Waltham, Massachusetts 02254*

(Received 10 February 1986)

However, it is important to understand the complexity of the problem and hence the limitations of a simplified model. Any mechanism which affects the transport of polaritons to the sample surface, the probability of transmission at the surface, the distribution of polaritons in the crystal, or the polariton energy distribution can influence the observed polariton luminescence. Consequently, one can expect great variations in the observed emission with different samples, surface preparation, or experimental conditions.

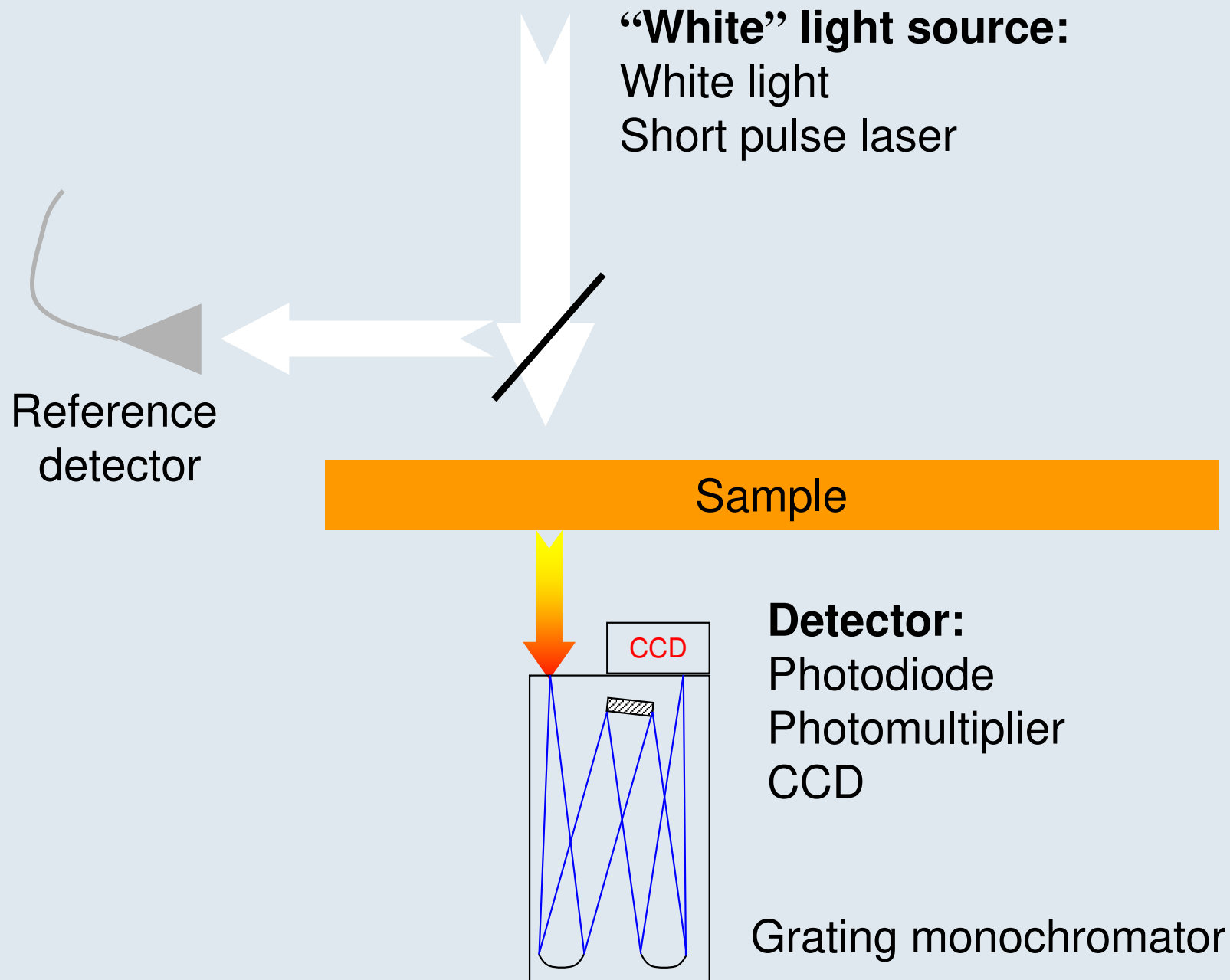
*"No single mechanism can explain the huge variety of observed polariton spectra in various crystals"*

# Excitons and polaritons in GaN

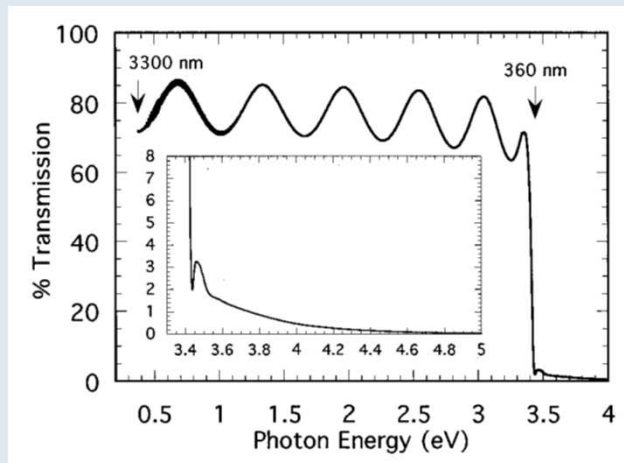


"THE BEAUTY OF THIS IS THAT IT IS ONLY OF THEORETICAL IMPORTANCE, AND THERE IS NO WAY IT CAN BE OF ANY PRACTICAL USE WHATSOEVER."

# Spectroscopy of GaN : Absorption

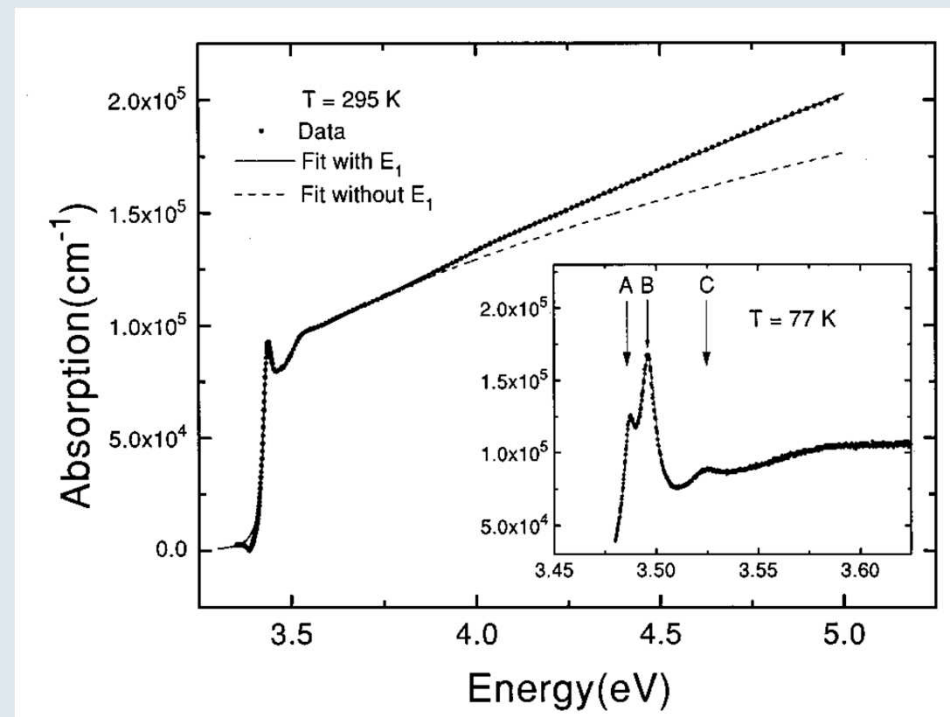


# Spectroscopy of GaN : Absorption



GaN, 0.4  $\mu\text{m}$

Sapphire



Muth *et al.* APL **71** 2572 (1997)

# Spectroscopy of GaN : Reflectivity

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## **Advantages of absorption:**

Direct probing of the electronic states (coupled to the light!)

Measurement does not perturb the system at low power

## **Limitations of absorption:**

Absorbing substrate (if substrate removal impossible)

Weak signal (especially if large broadening)

Complex structures

# Quantum wells, quantum dots in III-N

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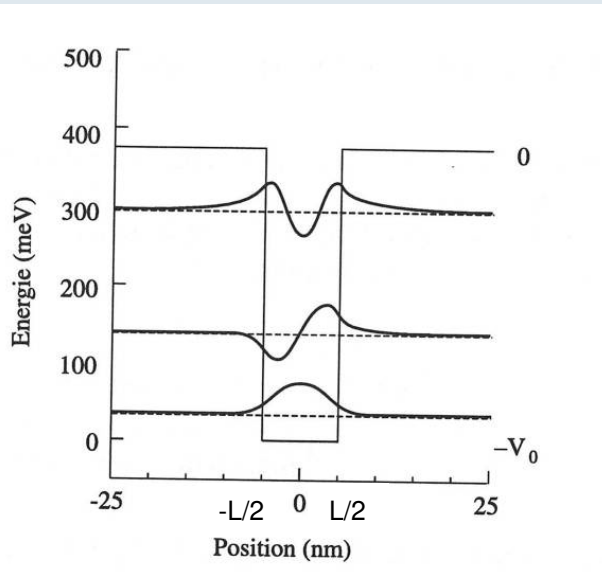
Localization, quantum confined Stark effect, excitons and all that...

# Quantum wells, the perfect case

$$E_{\vec{0},n} = \frac{\pi^2 \hbar^2 n^2}{2m^* L^2} + \frac{\hbar^2 k_{x,y}^2}{2m^*}$$

E

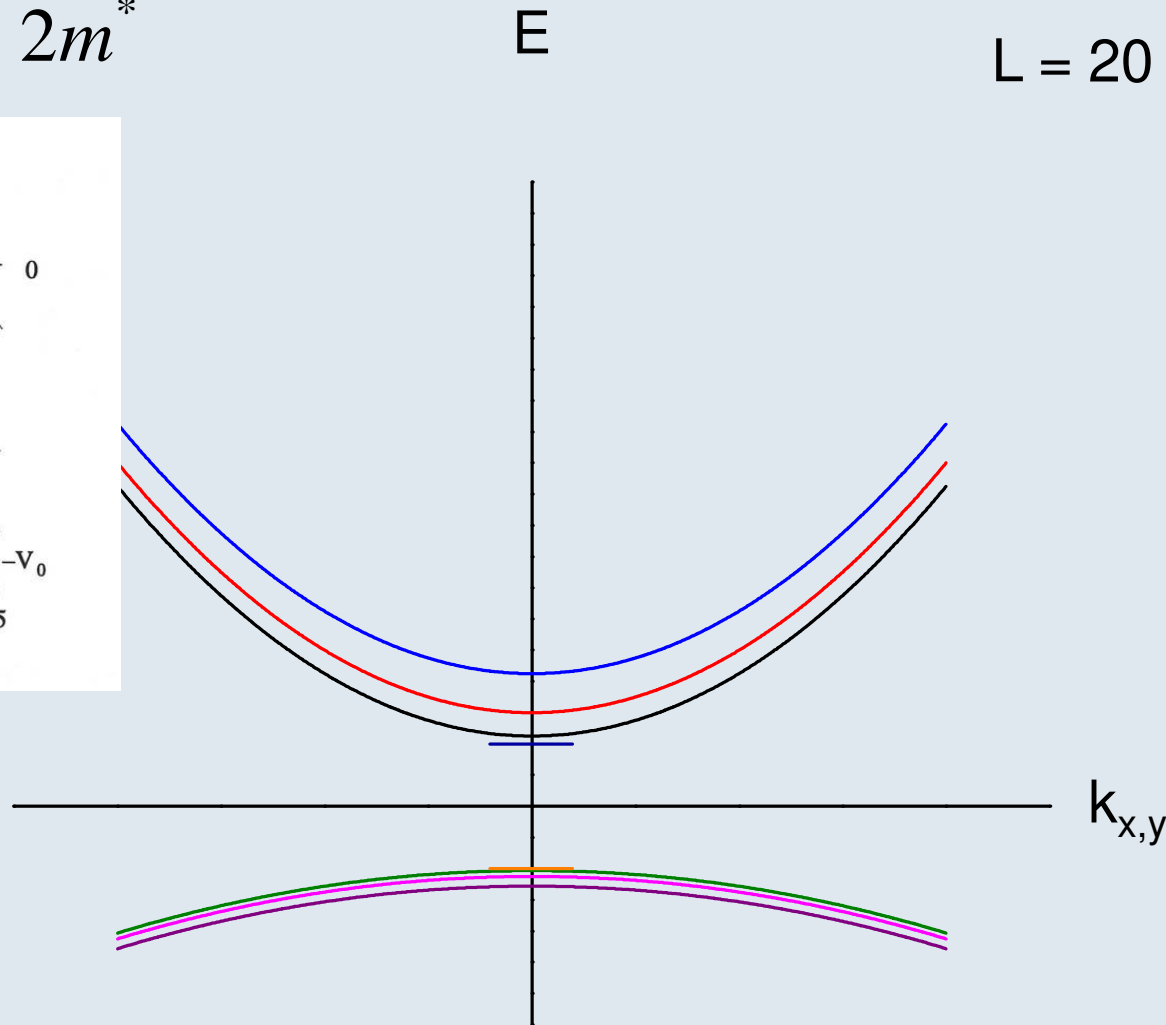
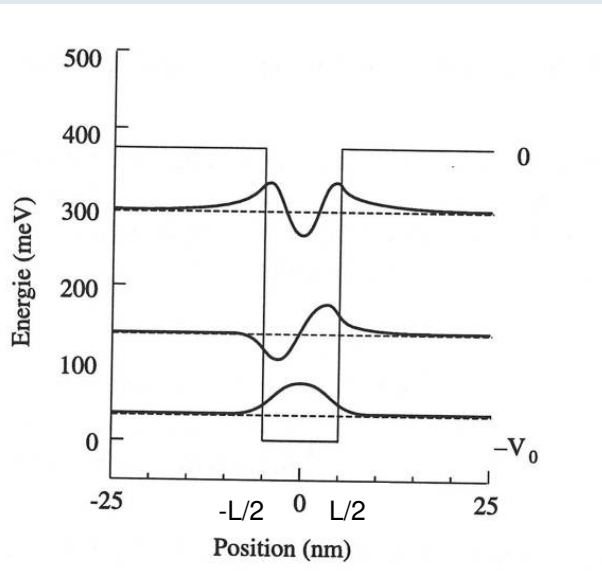
L = 100 nm



# Quantum wells, the perfect case

$$E_{\vec{0},n} = \frac{\pi^2 \hbar^2 n^2}{2m^* L^2} + \frac{\hbar^2 k_{x,y}^2}{2m^*}$$

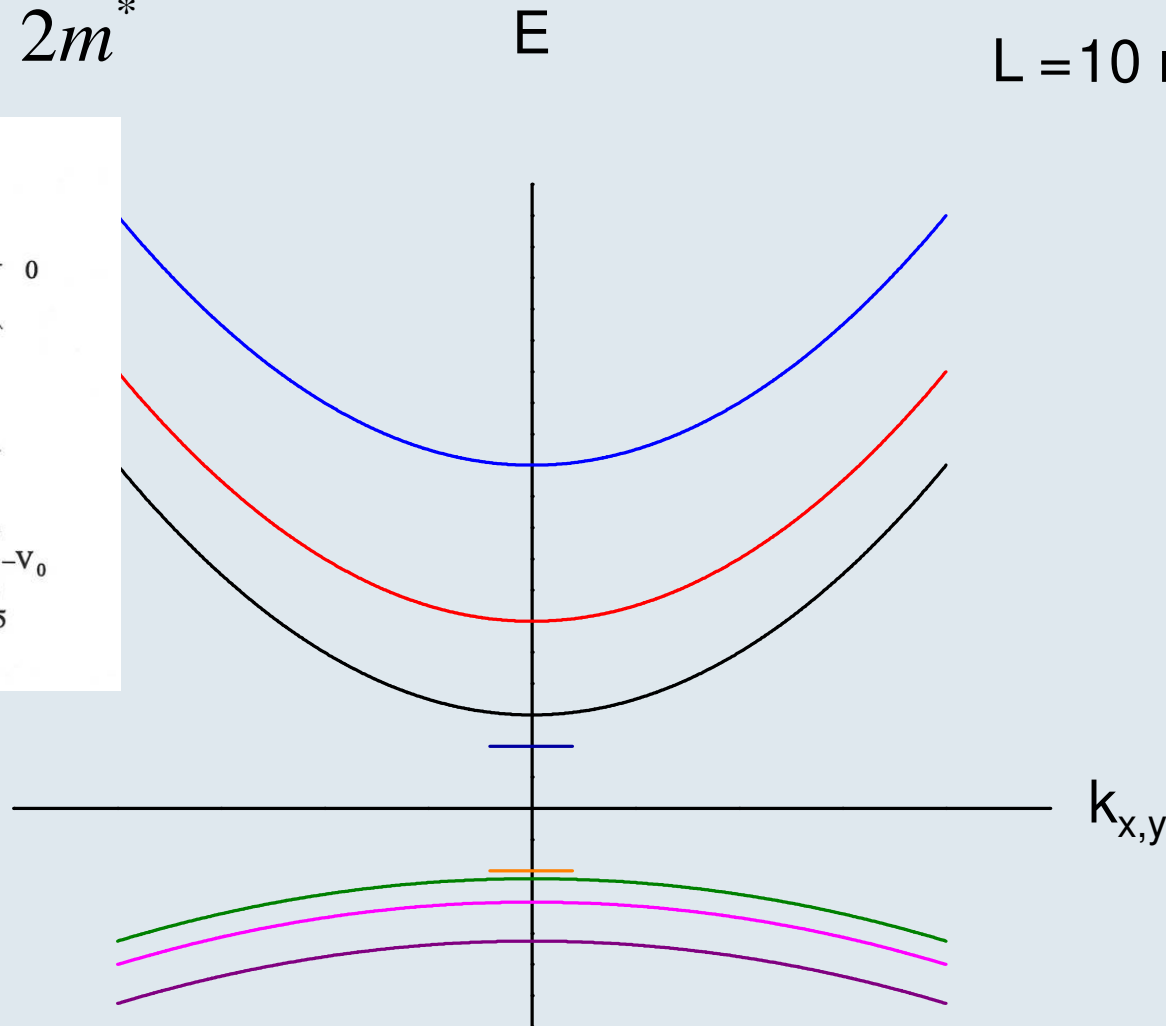
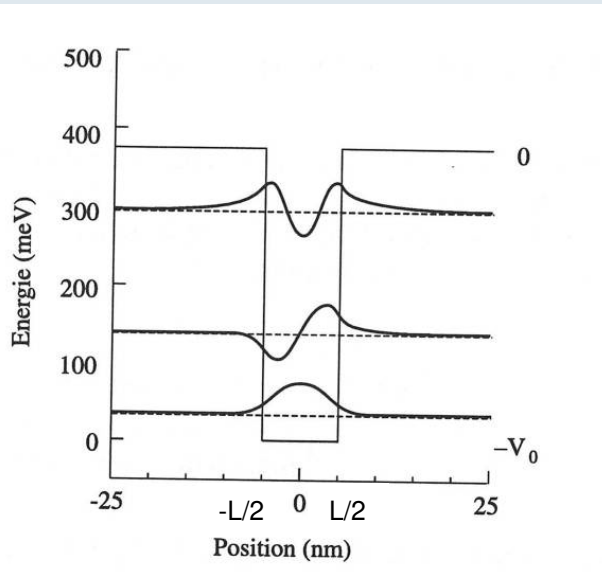
$L = 20 \text{ nm}$



# Quantum wells, the perfect case

$$E_{\vec{0},n} = \frac{\pi^2 \hbar^2 n^2}{2m^* L^2} + \frac{\hbar^2 k_{x,y}^2}{2m^*}$$

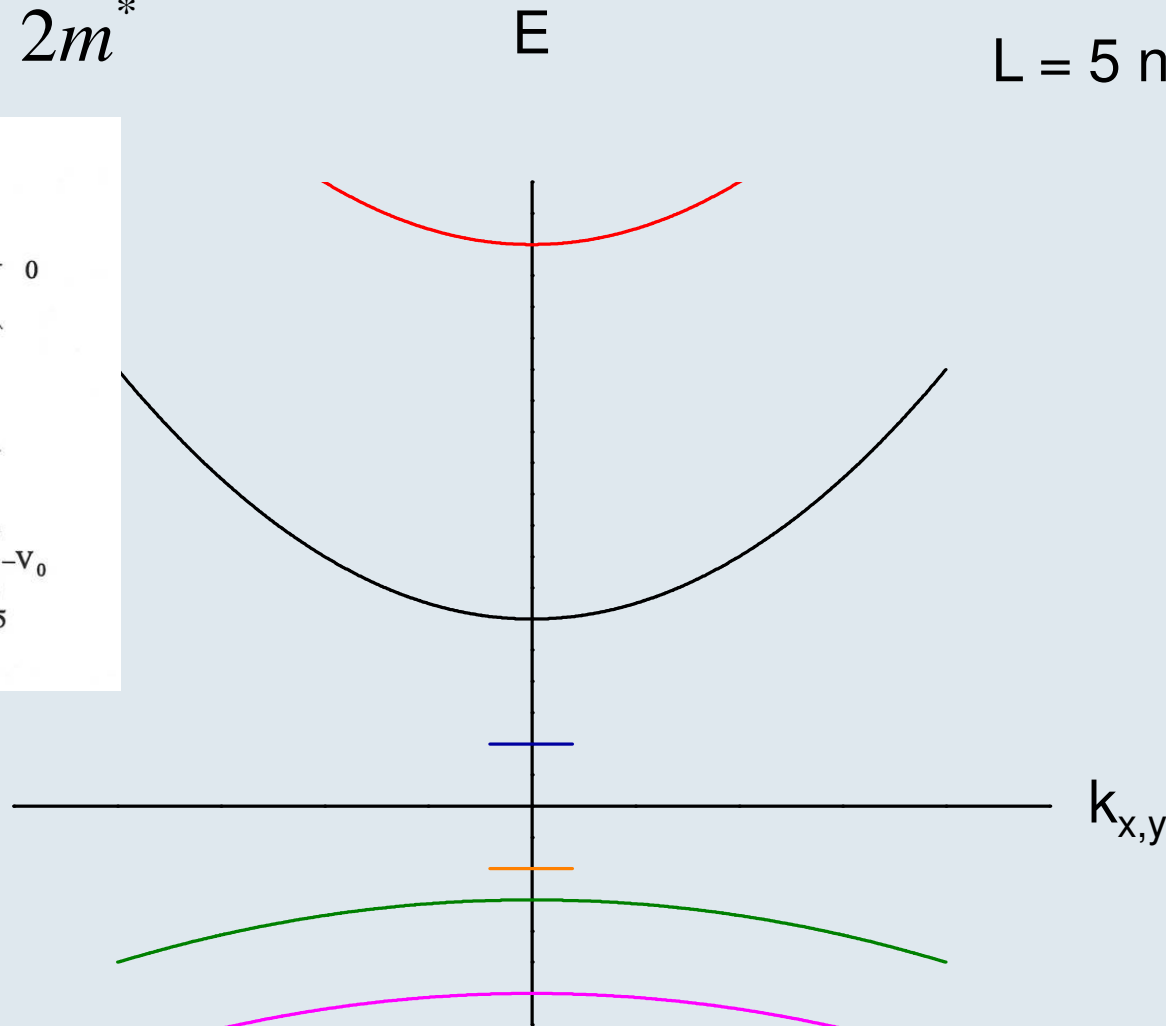
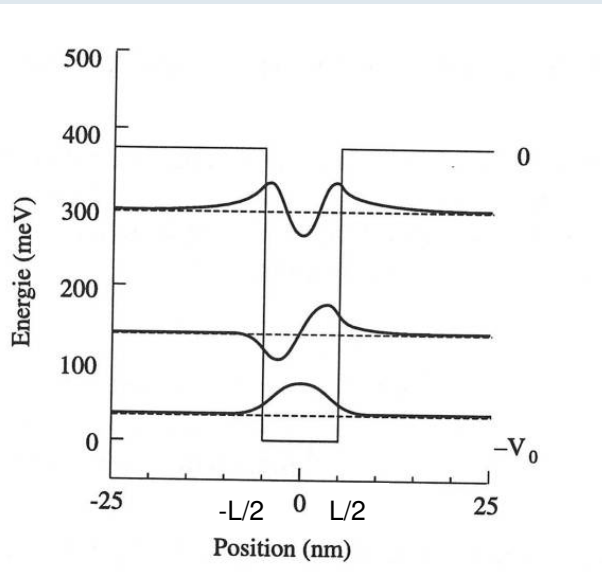
$L = 10 \text{ nm}$



# Quantum wells, the perfect case

$$E_{\vec{0},n} = \frac{\pi^2 \hbar^2 n^2}{2m^* L^2} + \frac{\hbar^2 k_{x,y}^2}{2m^*}$$

$L = 5 \text{ nm}$



# Quantum wells, the perfect case

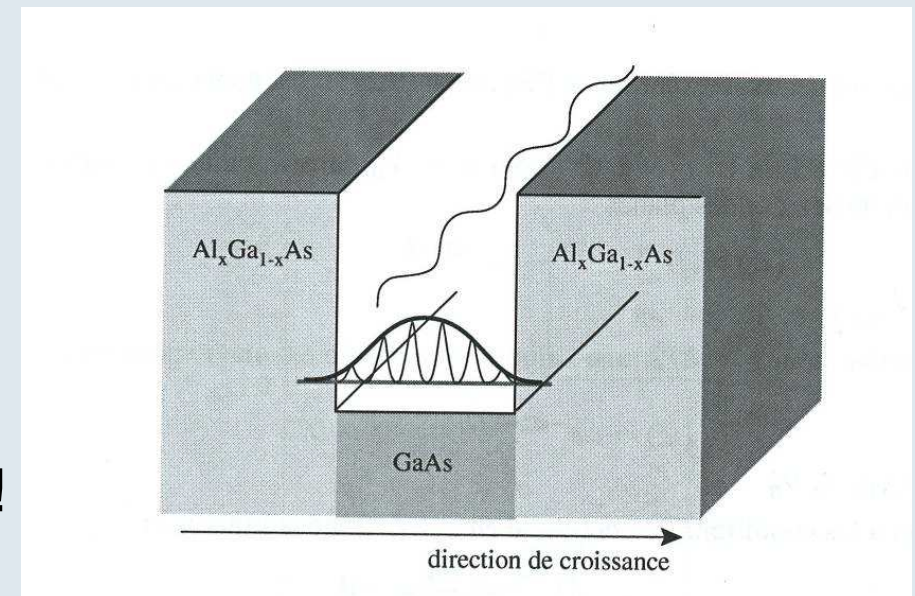
Ideally, the quantum well exciton is laterally fully delocalized

⇒ A pure 2-D state

⇒ Note that the exciton binding energy is up to 4 times larger in 2D structures

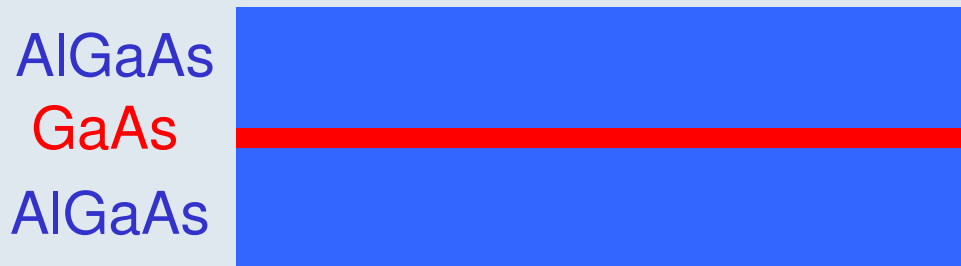
What about the Bohr radius ???

2 times smaller in 2D structures !!



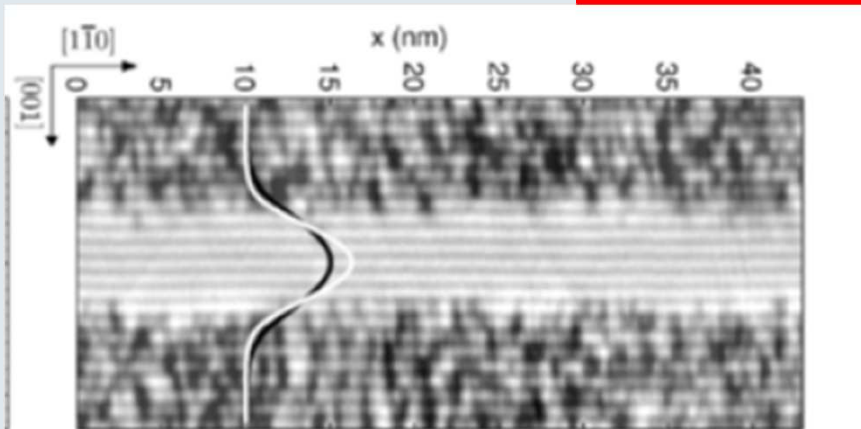
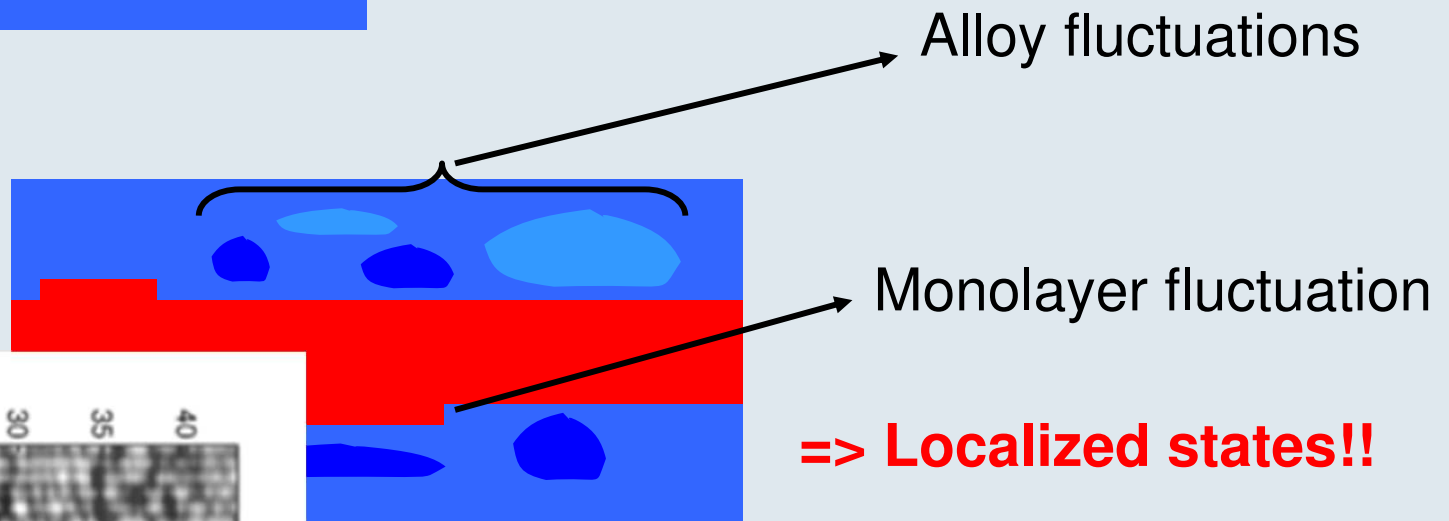
# Quantum wells, reality

Stokes-shift for quantum wells:



Perfect QW:  
electronic states  
with a well defined  
in-plane momentum

Reality:



GaAs/AlGaAs quantum well

See Nicolas Grandjean's lecture

# Cultural pause : Sir George Gabriel Stokes, 1st Baronet

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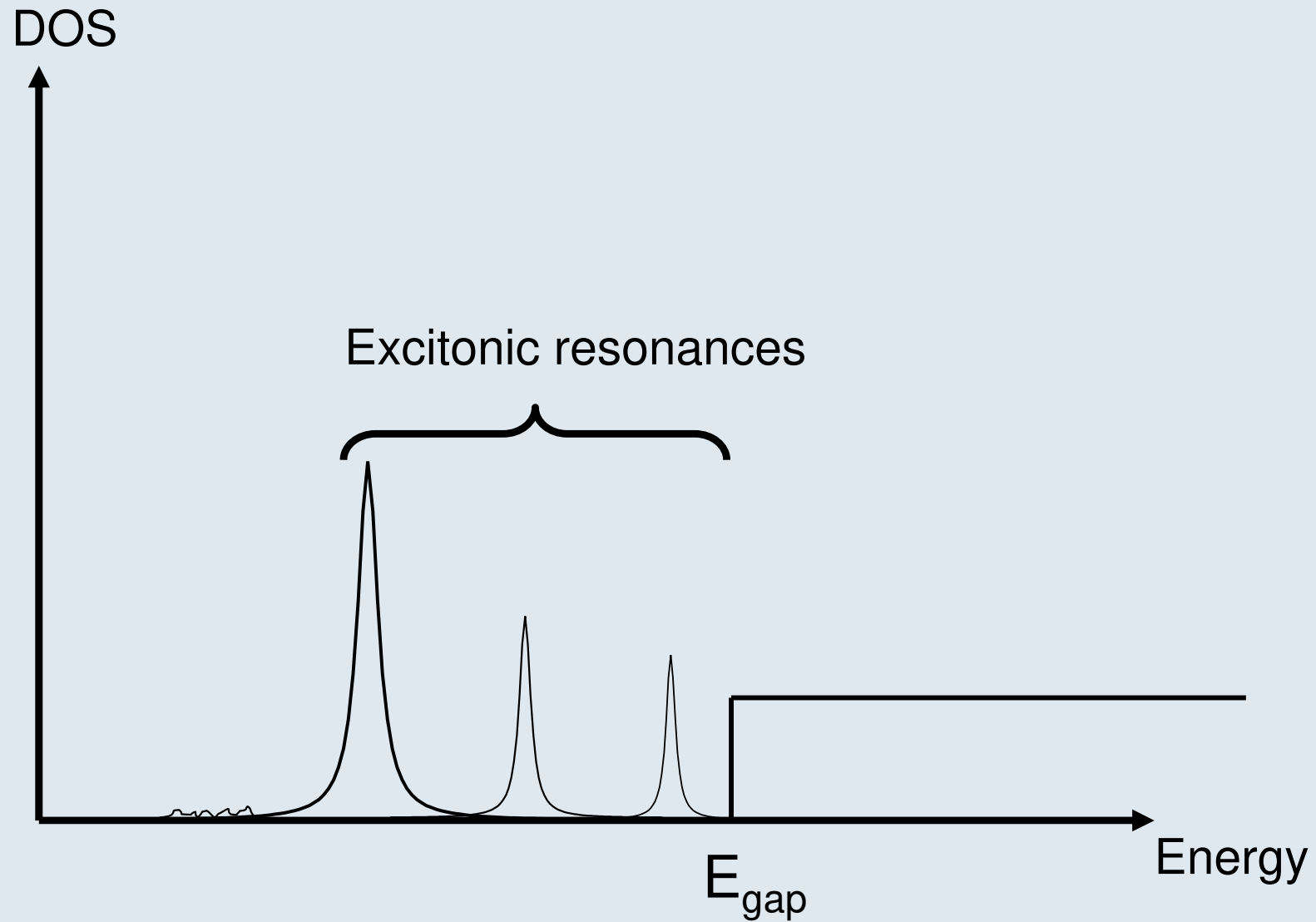
1819-1903

Major contributions in Mathematics (Stokes theorem), fluid mechanics (Navier-Stokes equation), optics (fluorescence, diffraction), biology (role of hemoglobin)...

*We all admit that the book of Nature and the book of Revelation come alike from God, and that consequently there can be no real discrepancy between the two if rightly interpreted.*

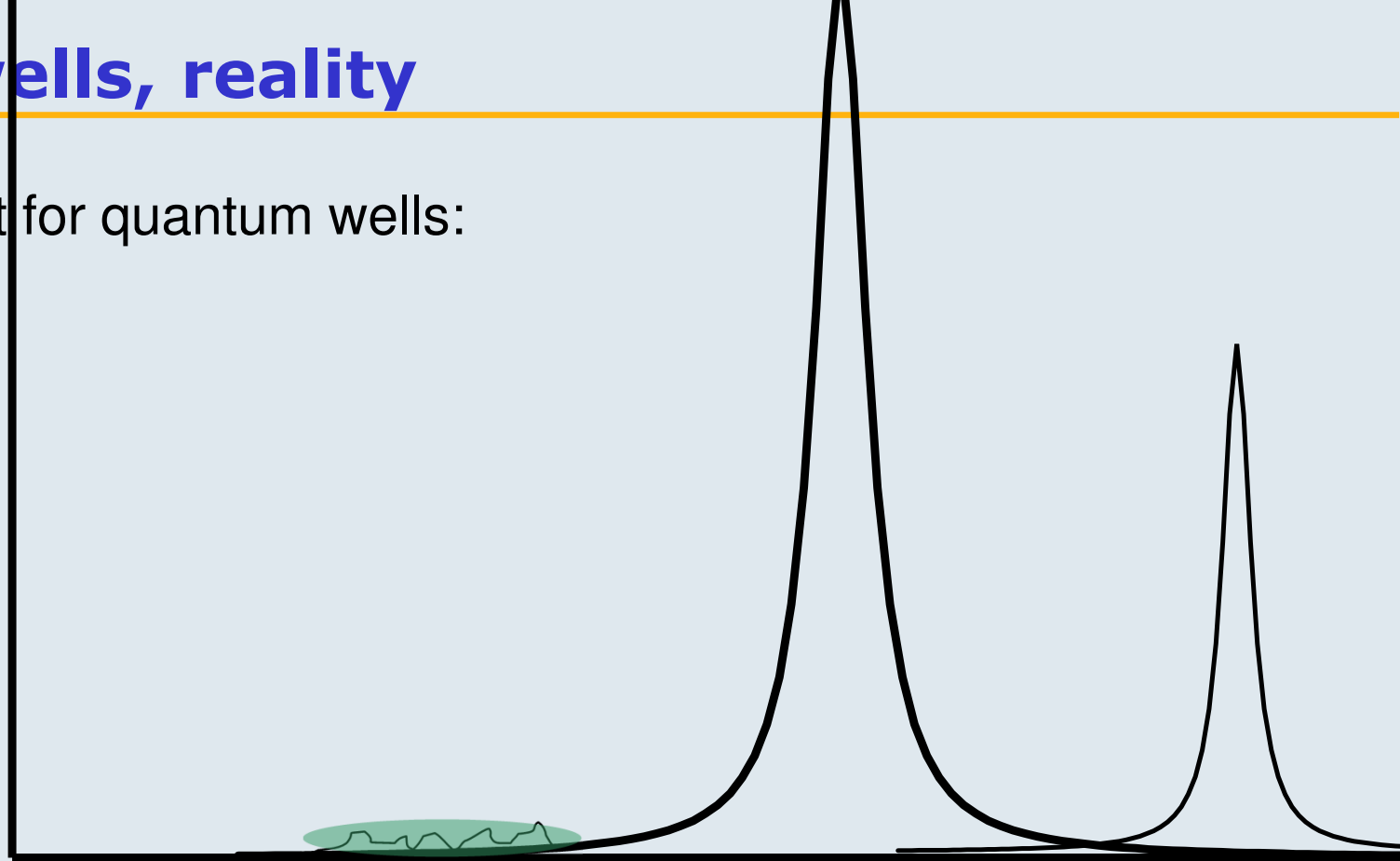
# Quantum wells, reality

Stokes-shift for quantum wells:



# Quantum wells, reality

Stokes-shift for quantum wells:

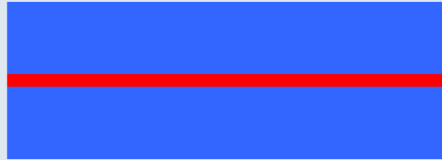


Localized states  
monolayer fluctuations  
alloy fluctuations  
impurity bound excitons

=> PL is not where the majority of states are

# Quantum wells, reality

Ideal QW

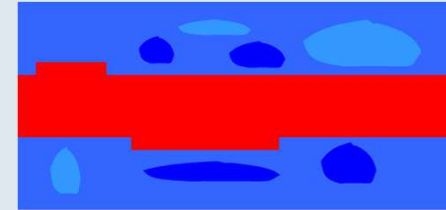


Fully in-plane delocalized exciton

Very short intrinsic lifetime

Radiative lifetime /  $kT$  limited broadening

Real QW



More or less localized « exciton »

Lifetime depends on lateral localization

Broadening reflects potential inhomogeneities

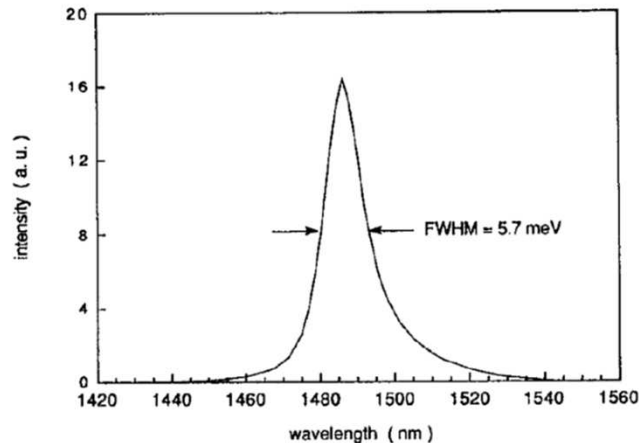
# Quantum wells, InGaN/GaN QWs

## Optical properties of high-quality InGaAs/InAlAs multiple quantum wells

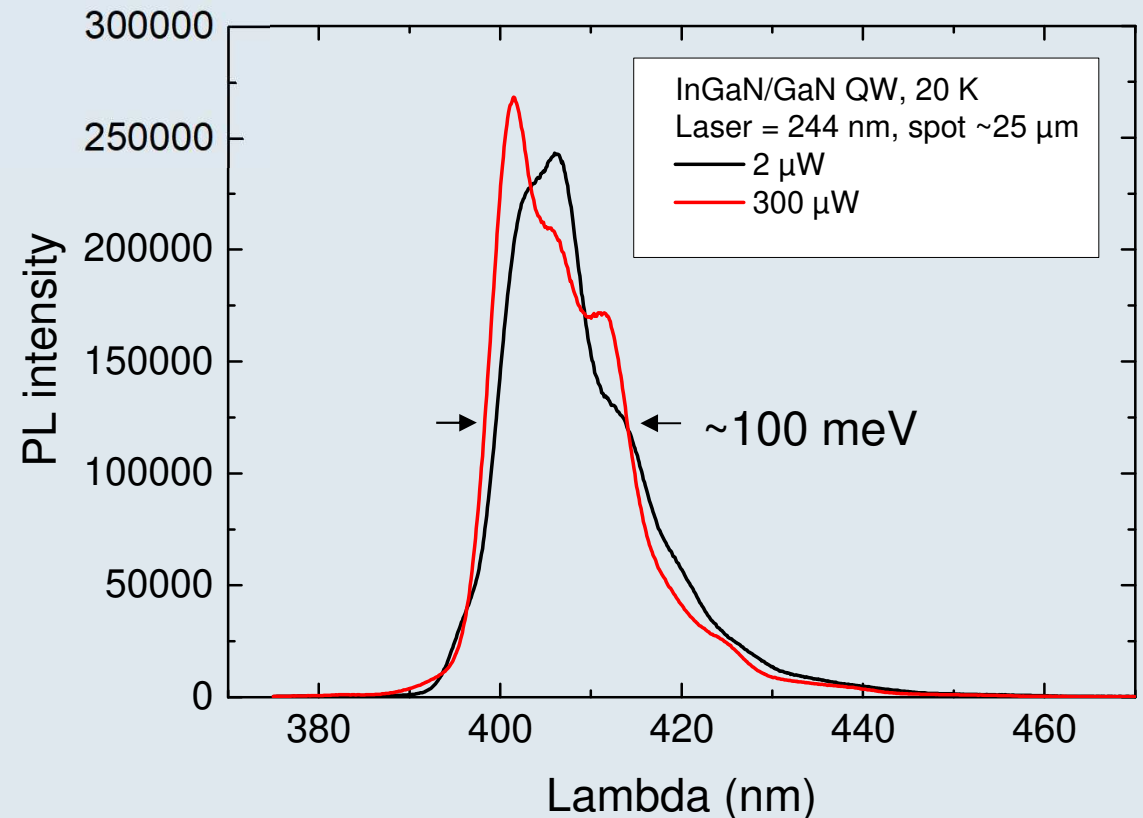
S. Gupta, P. K. Bhattacharya, J. Pamulapati, and G. Mourou

*Ultrafast Science Laboratory and Solid State Electronics Laboratory, Department of Electrical Engineering and Computer Science, University of Michigan, Ann Arbor, Michigan 48109*

(Received 12 September 1990; accepted for publication 6 December 1990)



Composition inhomogeneities  
at various scales



# Quantum wells, InGaN/GaN QWs

## Localization landscape theory of disorder in semiconductors III: Application to carrier transport and recombination in light emitting diodes

Chi-Kang Li,<sup>1</sup> Marco Piccardo,<sup>2</sup> Li-Shuo Lu,<sup>1</sup> Svitlana Mayboroda,<sup>3</sup> Lucio Martinelli,<sup>2</sup> Jacques Peretti,<sup>2</sup> James S. Speck,<sup>4</sup> Claude Weisbuch,<sup>2,4</sup> Marcel Filoche,<sup>2</sup> and Yuh-Renn Wu<sup>1,\*</sup>

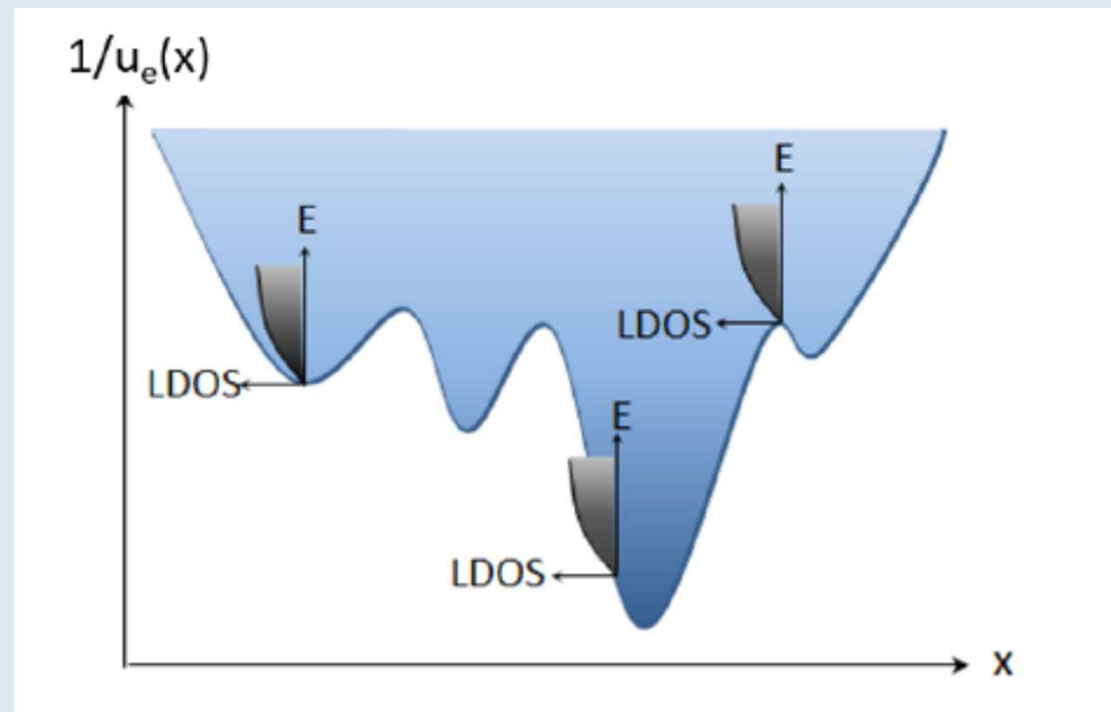
<sup>1</sup>*Graduate Institute of Photonics and Optoelectronics and Department of Electrical Engineering, National Taiwan University, Taipei 10617, Taiwan*

<sup>2</sup>*Laboratoire de Physique de la Matière Condensée, Ecole polytechnique, CNRS, Université Paris Saclay, 91128 Palaiseau Cedex, France*

<sup>3</sup>*School of Mathematics, University of Minnesota, Minneapolis, Minnesota 55455, USA*

<sup>4</sup>*Materials Department, University of California, Santa Barbara, California 93106, USA*

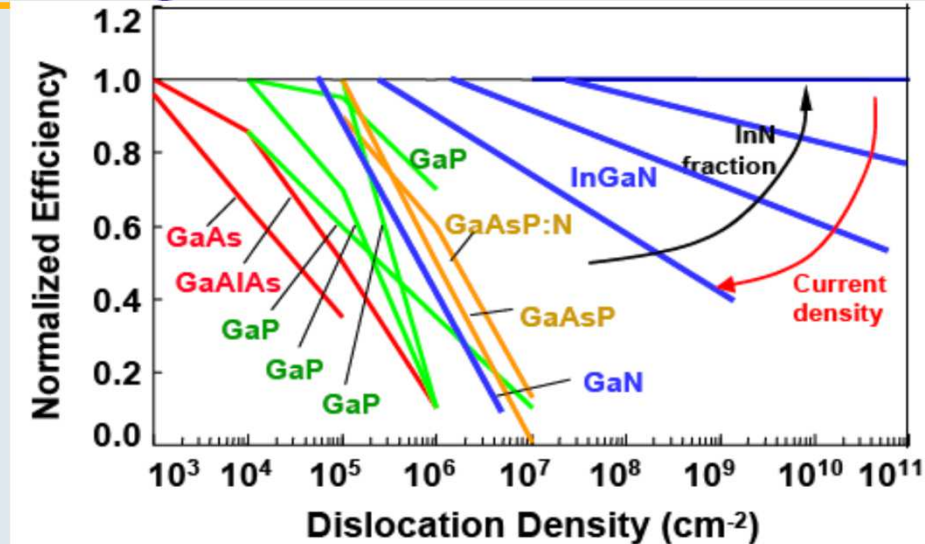
Strong localization effects in  
InGaN QWs ??



# Quantum wells, InGaN/GaN QWs

InGaN/GaN QWs are happily pathological

A vigorous debate in the last 20 years  
...not settled yet (getting closer)



~~Dislocations are not NR centers~~

~~Indium clustering localization~~

Dislocations « protected » by V-pits

~~Localization intrinsic to random InGaN alloy~~

# Quantum wells, InGaN/GaN QWs

---

InGaN quantum wells luminescence :

- Role of dislocations ( $\Rightarrow$  V-pits)
- Role of point defects ( $\Rightarrow$  Underlayer)
- Dependency on excitation density ("carrier temperature")

*Corollary 1* : the most efficient semiconductor emitting structure, the most sold optoelectronic layer is still not yet well understood (...getting there !)

*Corollary 2* : industrials have successfully spent big money to optimize InGaN/GaN QWs, with very little physical understanding of what's going on

# III-N quantum wells and the QCSE

Spontaneous polarization : it's complicated ...

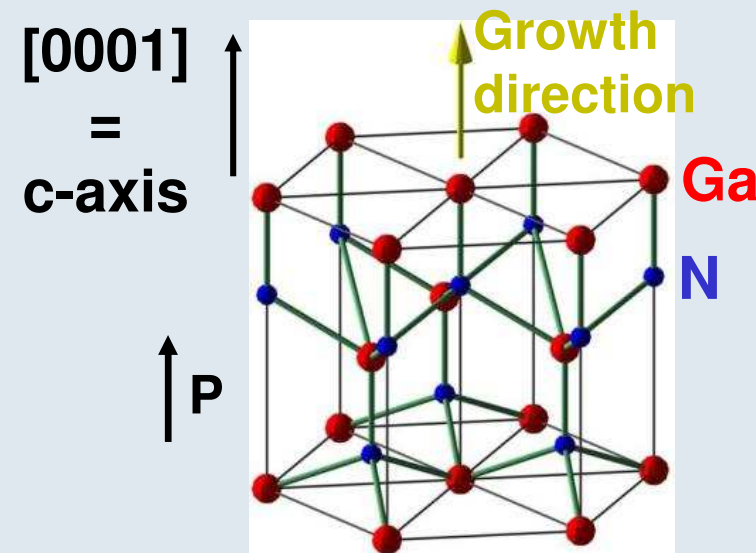
Macroscopic electric polarization is a fundamental concept in the physics of matter, upon which the phenomenological description of dielectrics is based (Landau and Lifshitz, 1984). Notwithstanding, this concept has long evaded even a precise microscopic definition. A typical incorrect statement—often found in textbooks—is that the macroscopic polarization of a solid is the dipole of a unit cell. It is easy to realize that such a quantity is neither measurable nor model-independent: the dipole of a periodic charge distribution is in fact ill defined

R. Resta, Rev. Mod. Phys. **66**, 899 (1994)

Only polarization differences can be measured !

In materials with an inversion symmetry, the spontaneous polarization is null (by convention)

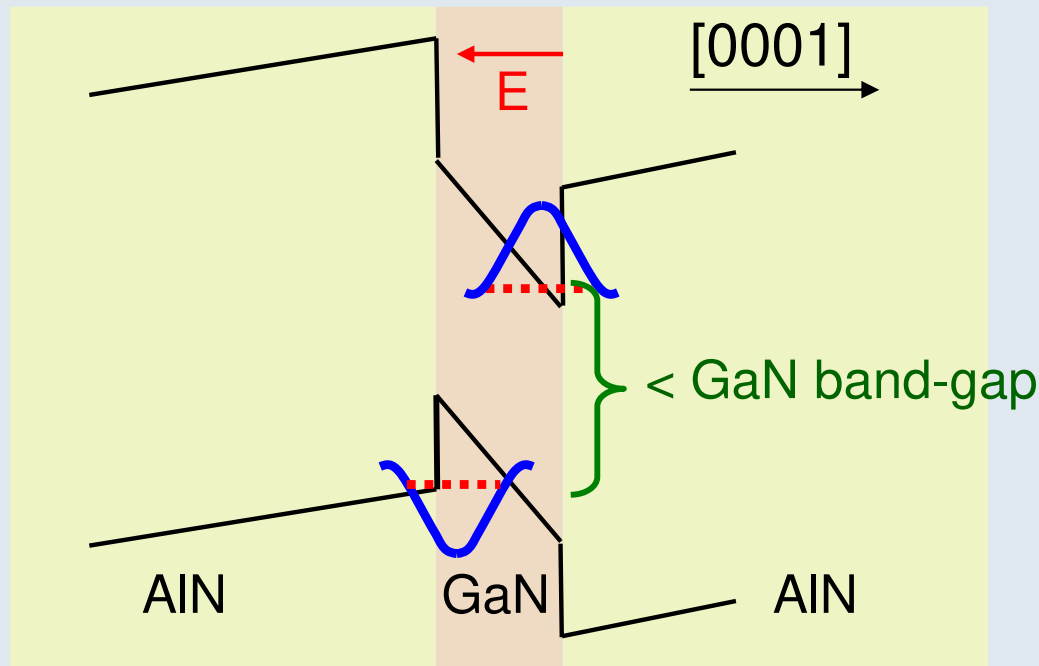
$$\vec{P} = \frac{1}{V} \left( -e \sum_l Z_l \vec{R}_l + \int d\vec{r} \rho(\vec{r}) \right)$$



$$\vec{F}_{QW} = \frac{\Delta \vec{P}}{\epsilon_0 \epsilon_r}$$

Review : Effects of Polarization in Optoelectronic Quantum Structures, R. Butté and N. Grandjean  
In "Polarization Effects in Semiconductors: From Ab Initio Theory to Device Applications", Springer, 2008

# III-N quantum wells and the QCSE



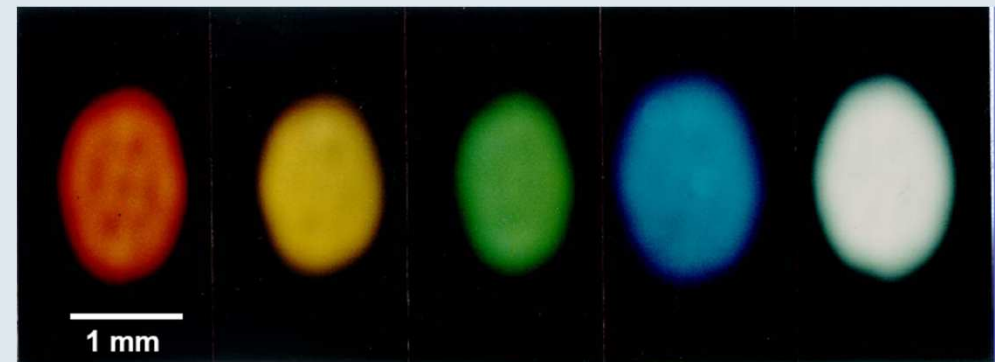
F. Bernardini *et al.*, Phys Rev B **56**, R10024 (1997)

Below well bandgap emission

**Experimental result:**

$$P_{\text{sp,GaN}} + P_{\text{pz,GaN}} - P_{\text{sp,AlN}} - P_{\text{pz,AlN}}$$

Reduced oscillator strength

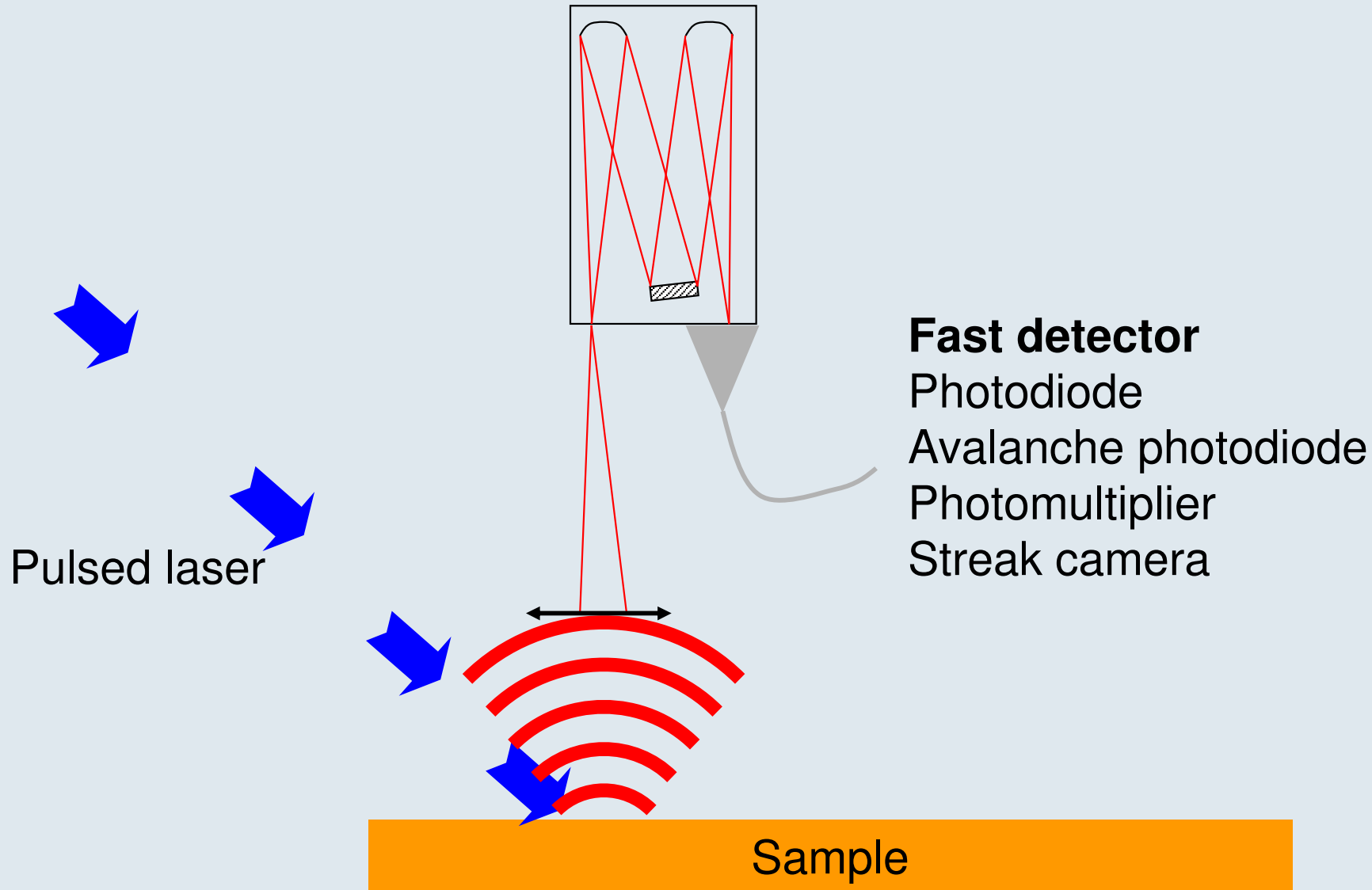


← 50 Å GaN QD height 25 Å QD stacking →

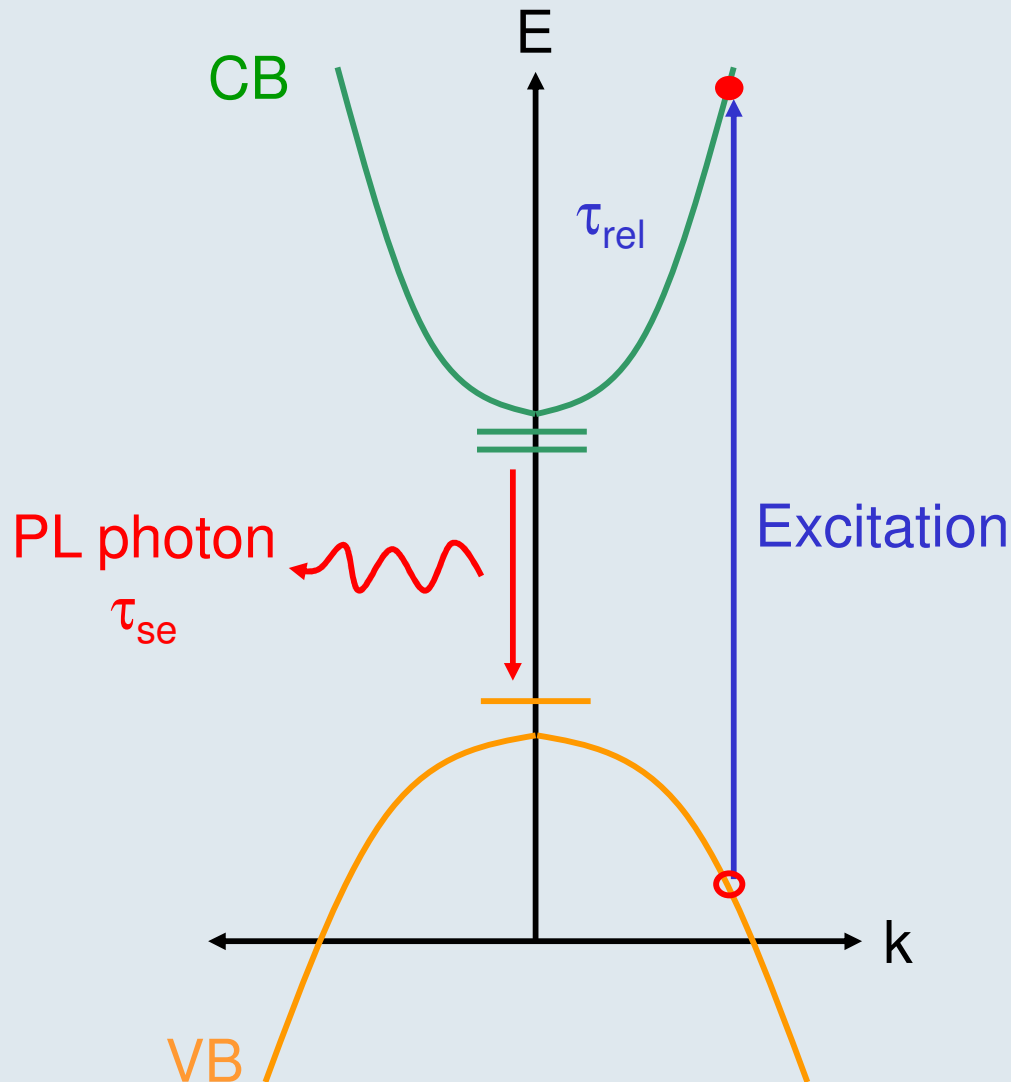
Damilano *et al.*, Appl. Phys. Lett. **75** 962 (1999)

Most important effect = radiative lifetime  
=> time-resolved PL

# Experimental technique : time-resolved PL



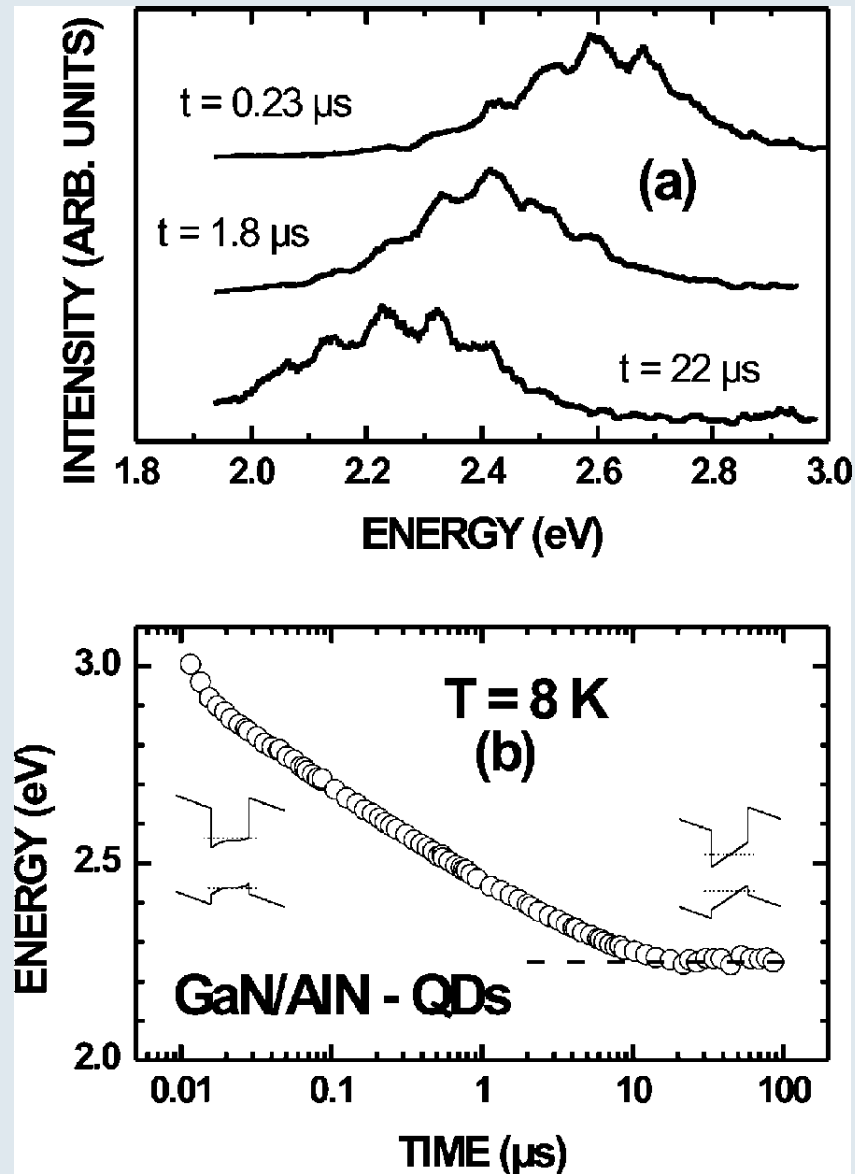
# Experimental technique : time-resolved PL



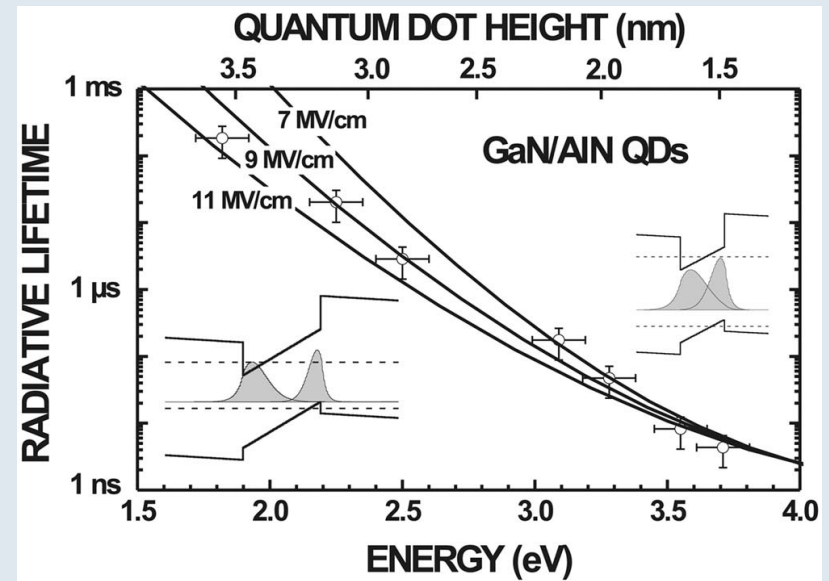
$$I(t, \lambda) \propto \frac{\tau_{se}}{\tau_{se} - \tau_{rel}} \left( e^{-\frac{t}{\tau_{se}}} - e^{-\frac{t}{\tau_{rel}}} \right)$$

⇒ Measurement of relaxation time and spontaneous emission time

# III-N quantum wells and the QCSE



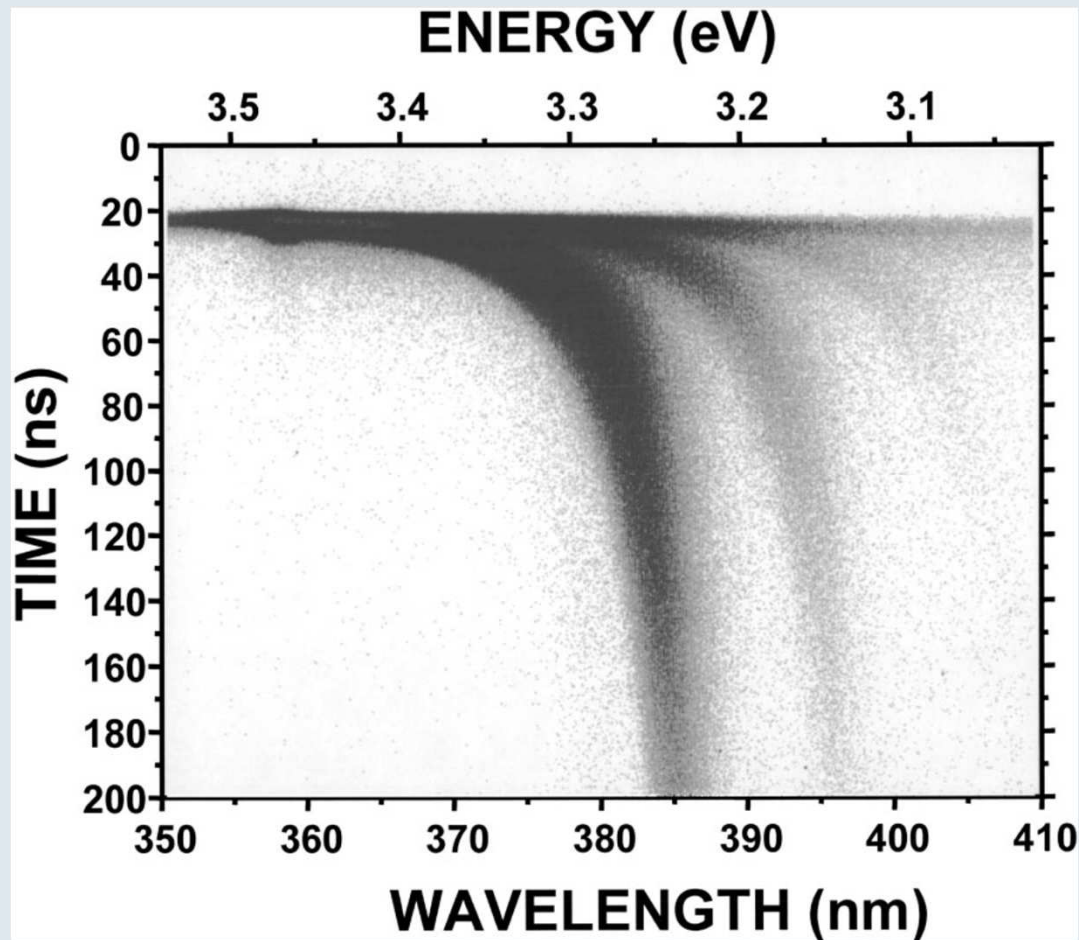
At « large » excitation density :  
screening of the QCSE



=> Extremely long lifetimes

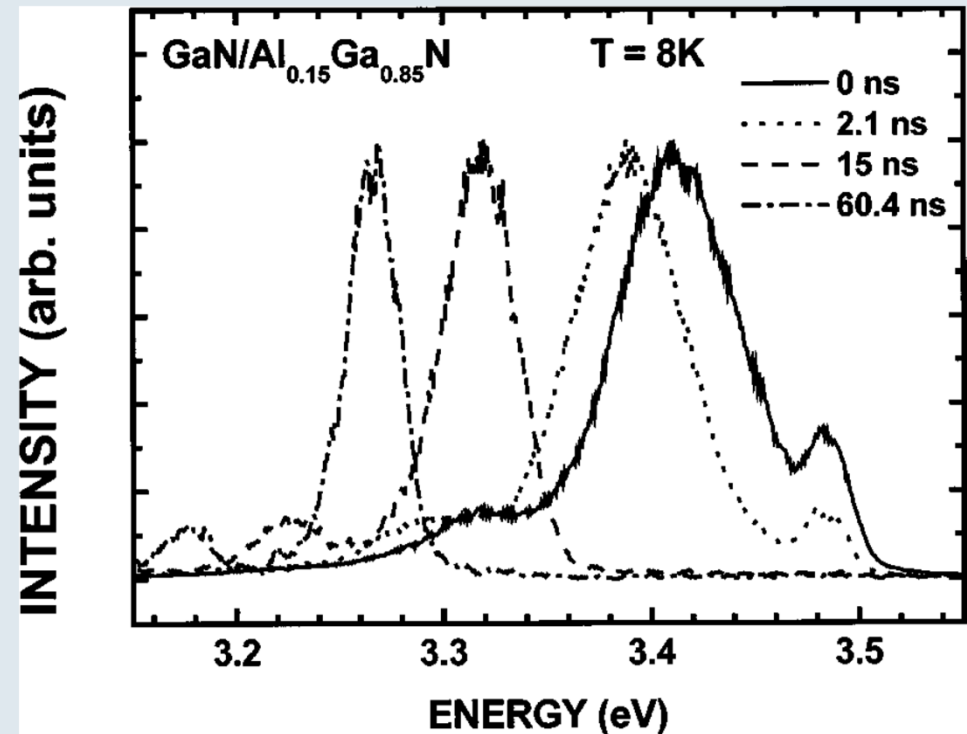
Bretagnon *et al.*, PRB **73**, 113304 (2006)

# III-N quantum wells and the QCSE



Importance of the streak camera

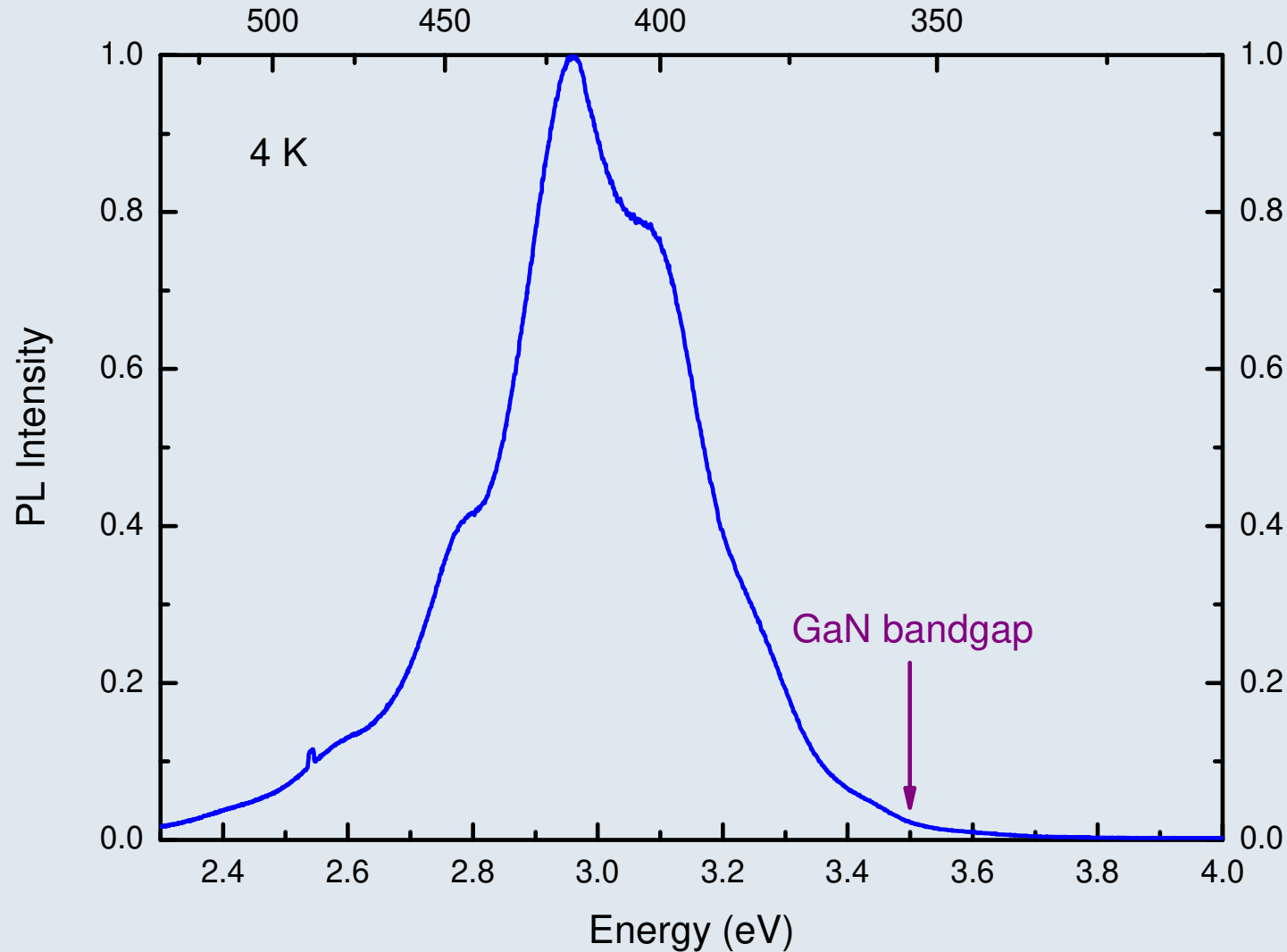
Beware of data interpretation!



Lefebvre *et al.* PRB **69** 035307 (2004)

# III-N quantum wells and the QCSE

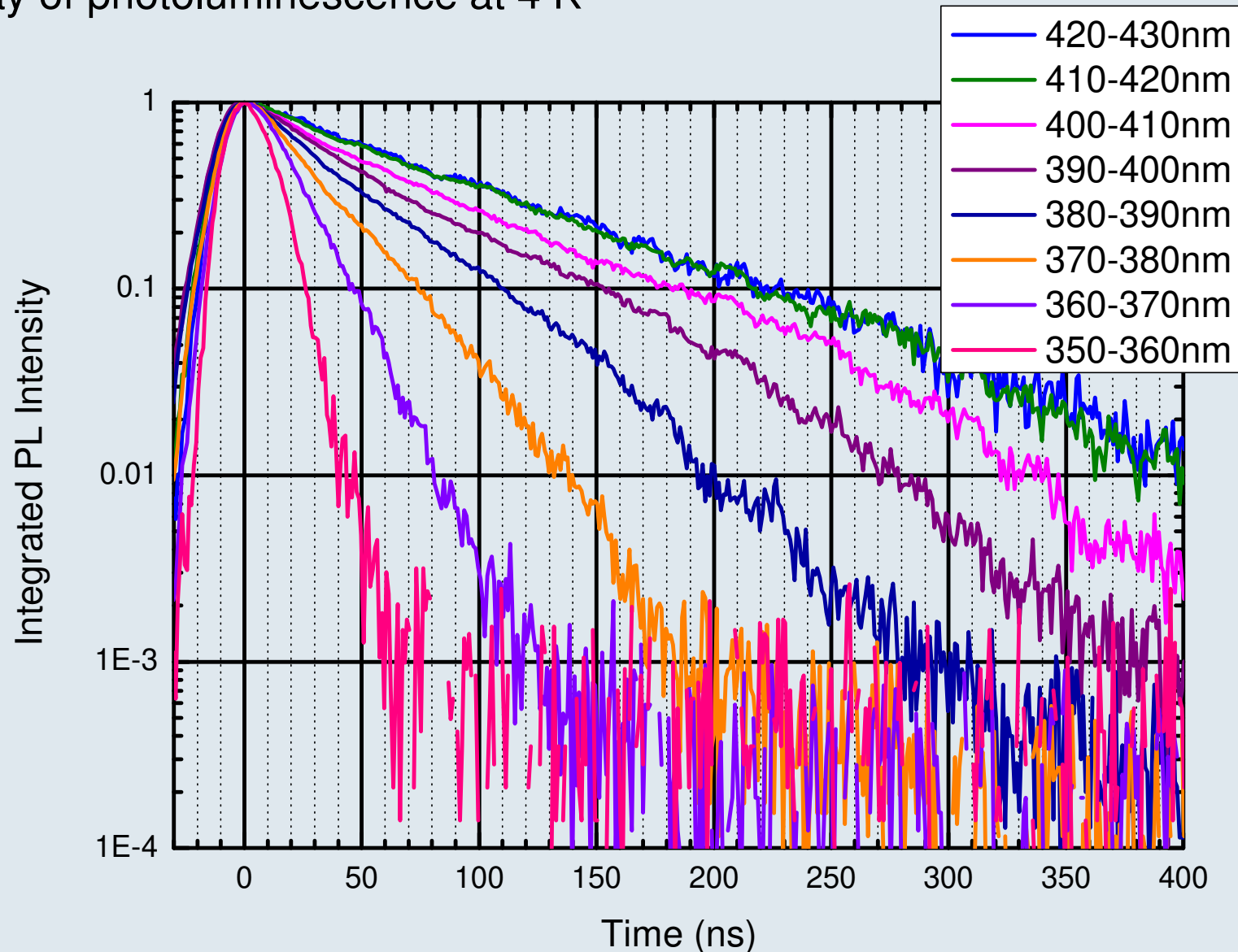
Inhomogeneously broadened GaN/AlN QDs



# III-N quantum wells and the QCSE

Inhomogeneously broadened GaN/AlN QDs

Decay of photoluminescence at 4 K

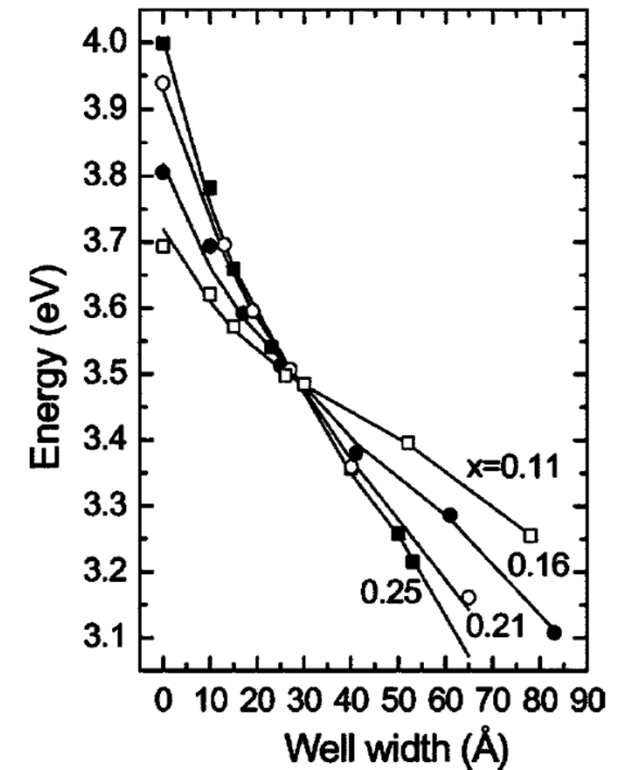
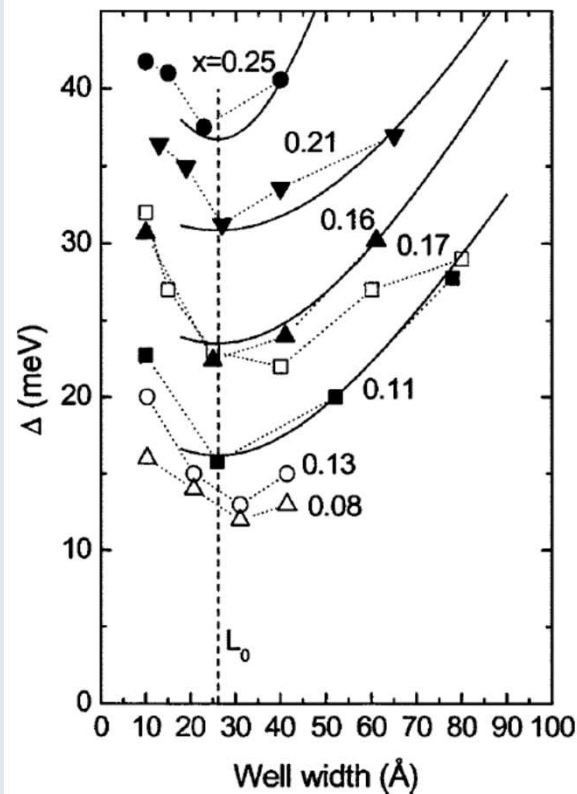
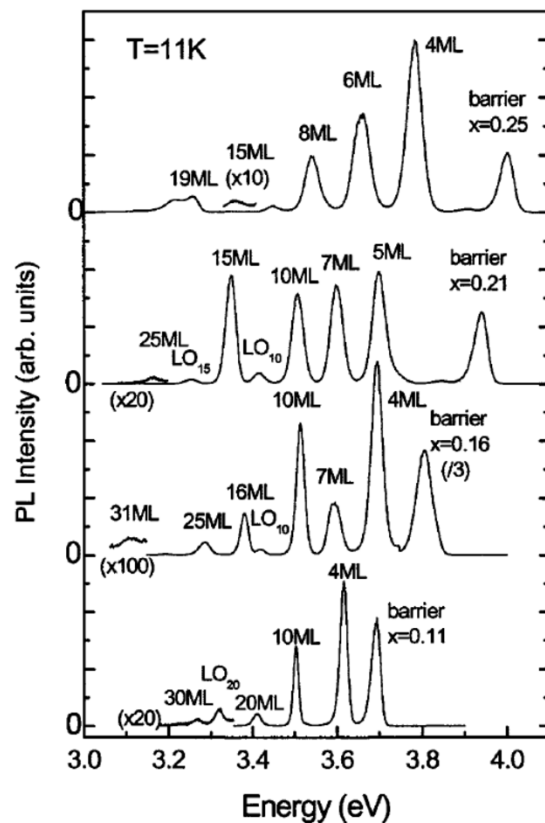


# Quantum wells, GaN/AlGaN QWs

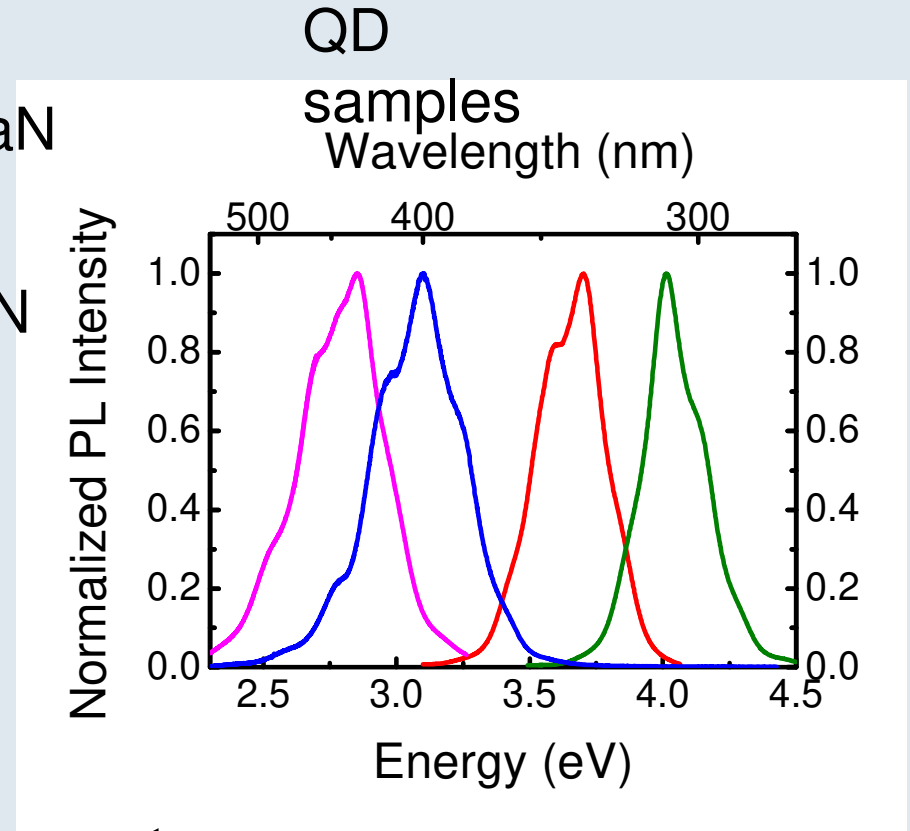
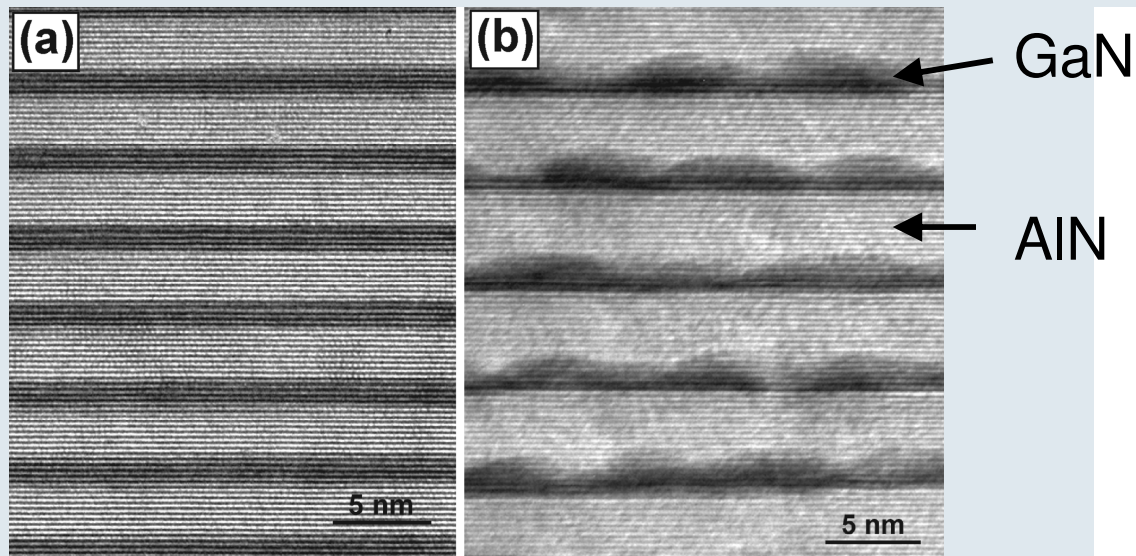
PHYSICAL REVIEW B **71**, 075311 (2005)

## Inhomogeneous broadening of $\text{Al}_x\text{Ga}_{1-x}\text{N}/\text{GaN}$ quantum wells

F. Natali, D. Byrne, M. Leroux, B. Damilano, F. Semond, A. Le Louarn, S. Vezian, N. Grandjean, and J. Massies  
*Centre de Recherche sur l'Hétéro-Epitaxie et ses Applications-CNRS, Rue Bernard Grégory, 06560 Valbonne, France*  
 (Received 17 November 2003; revised manuscript received 10 September 2004; published 15 February 2005)



# QDs vs QWs

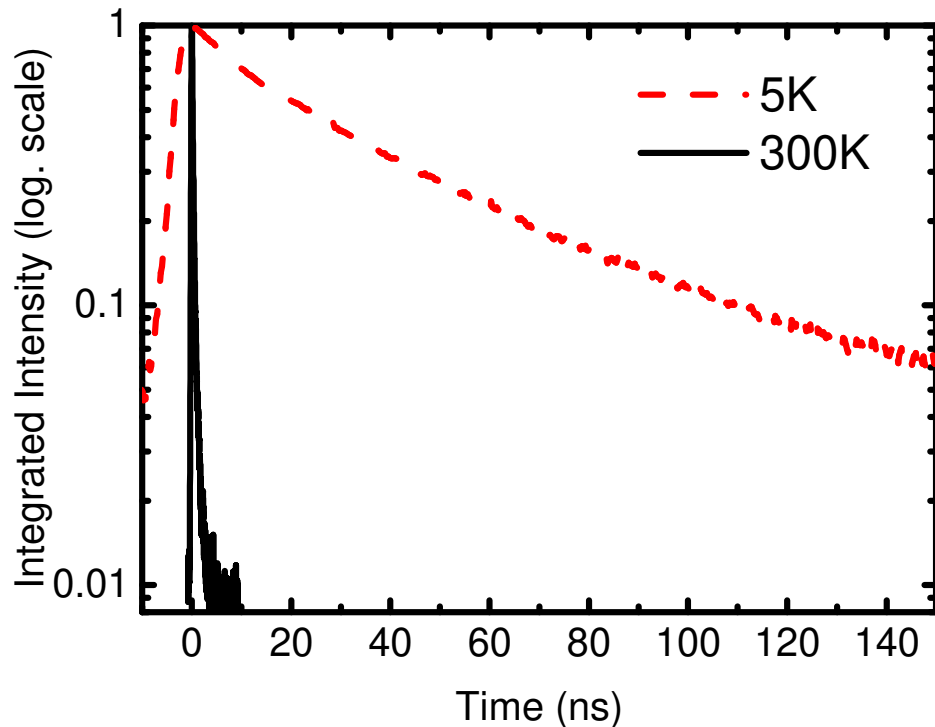


← Increasing thickness (1-3 nm)

Large range of energy → large range of decay times (0.3 - 500 ns)

# QDs vs QWs

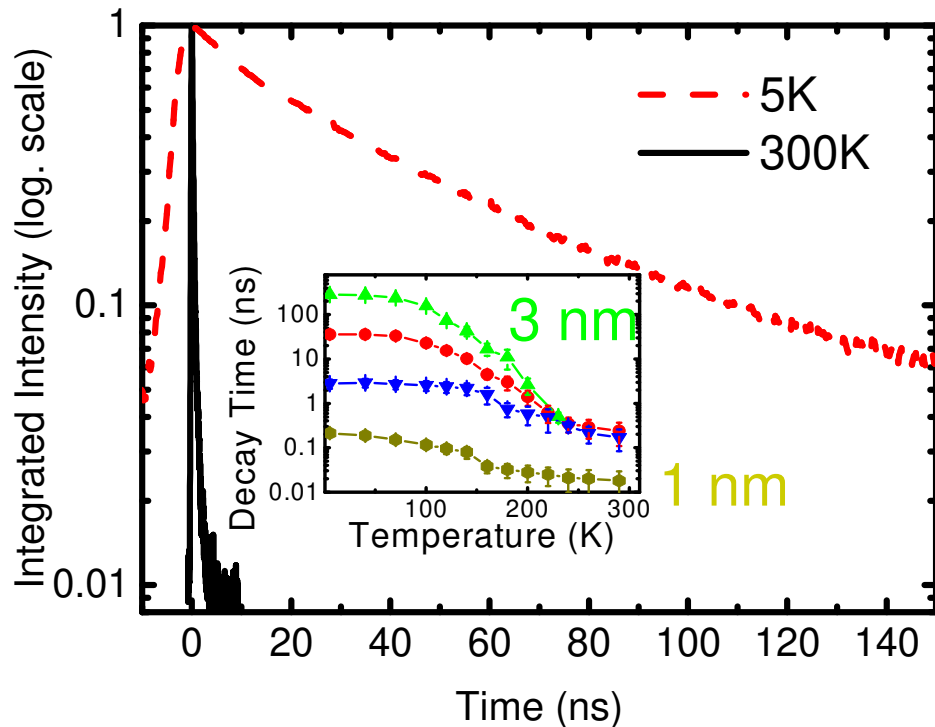
## Quantum wells



*QWs: strong decrease of the decay time → non radiative processes*

# QDs vs QWs

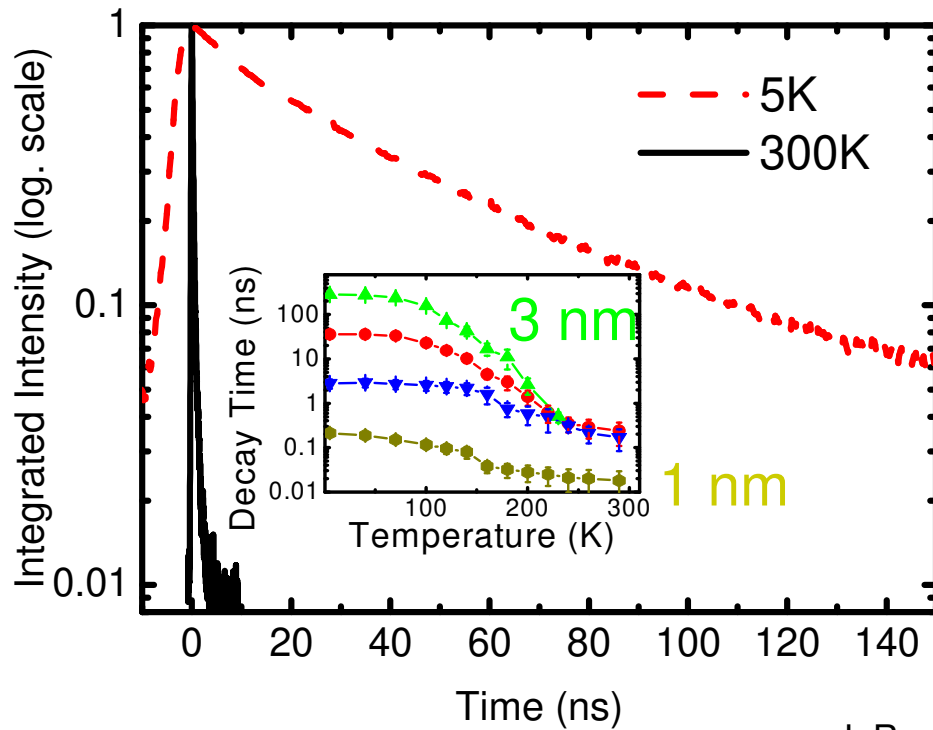
Quantum wells (1-3 nm)



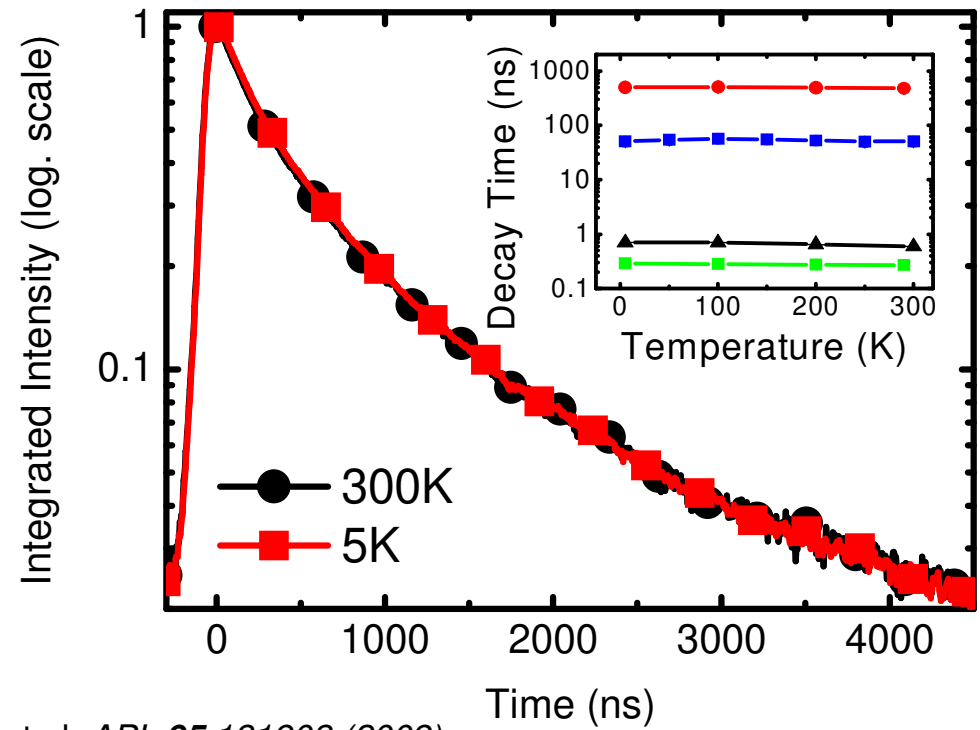
*QWs: strong decrease of the decay time → non radiative processes*

# TRPL STUDY

Quantum wells (1-3 nm)



Quantum dots (3-5 MLs)



J. Renard et al. *APL* **95** 131903 (2009)

*QWs: strong decrease of the decay time → non radiative processes*

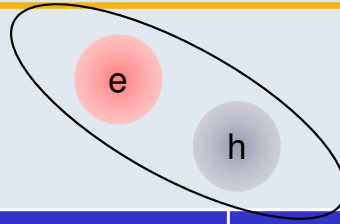
*QDs: constant decay time → negligible non radiative processes*

**Absence of thermal escape**

*Thermally activated !*

Low radiative emission rate does not imply low efficiency !!!

# The super exciton in GaN : it is useful?



|      | a (nm) | $E_x$ (meV) |
|------|--------|-------------|
| GaN  | 2.8    | 25          |
| GaAs | 12     | 4.8         |

$kT$  @ 300K  
25 meV



The excitonic force  
is strong in GaN

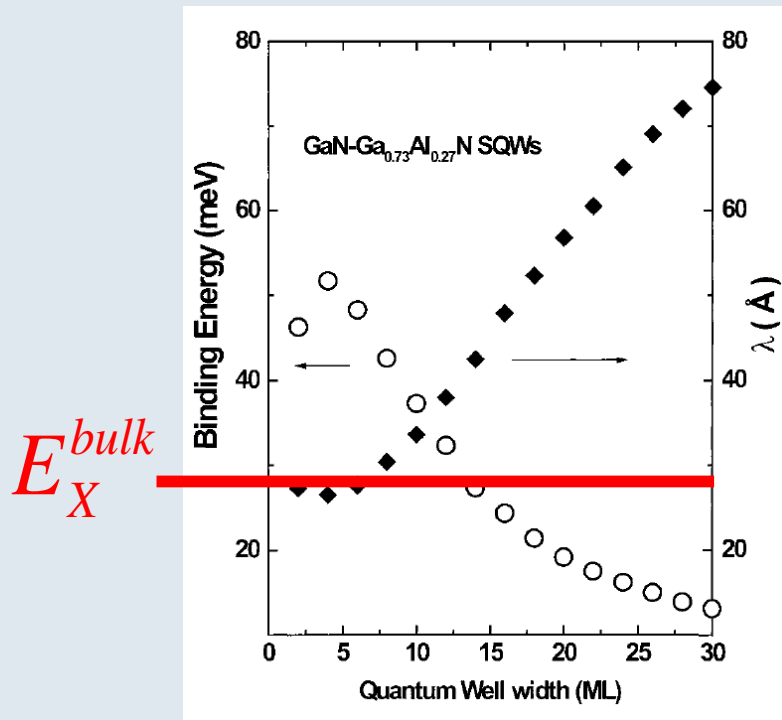
*Why is it important ?  
Is it useful ?*

⇒ The binding energy of excitons in QW is up to 4 times larger than in bulk

M. Shinada and S. Sugano, J. Phys. Soc. Jpn. **21** 1936 (1966)

# The super exciton in GaN : it is useful?

*The QCSE reduces the exciton binding energy...*



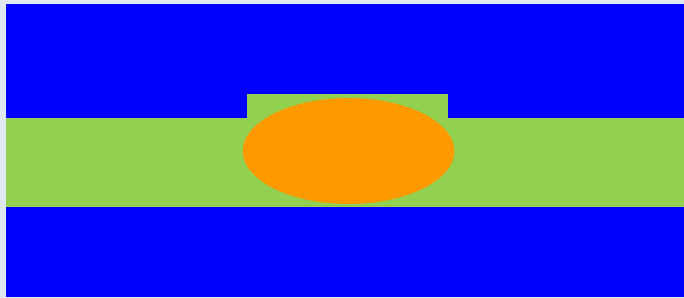
P. Bigenwald *et al.*, Phys. Stat. Sol. (b) **216** 371 (1999)

*... but in thin QWs or non-polar QWs, the exciton binding energy can be large!*

# Excitonic effects in QWs

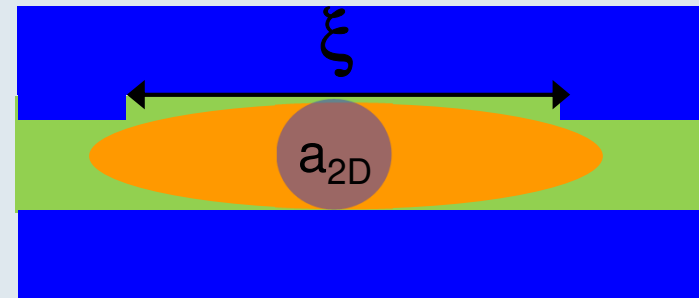
Do we get fast emission from a strong QW exciton?

$$\tau_{\text{rad}} \propto \frac{1}{nE^2 f}$$



Strong confinement

$$f = \frac{E_p}{E}$$



Lateral weak confinement

$$\psi_X(r_e, r_h) \propto f(z_e) g(z_h) e^{-r/a_{2D}} e^{-R_p^2/2\xi^2}$$

$$f = \frac{8E_p}{E} \left( \frac{\xi}{a_{2D}} \right)^2$$

⇒ Giant oscillator strength

A. V. Kavokin, Phys. Rev. B, **50**, 8000 (1994).

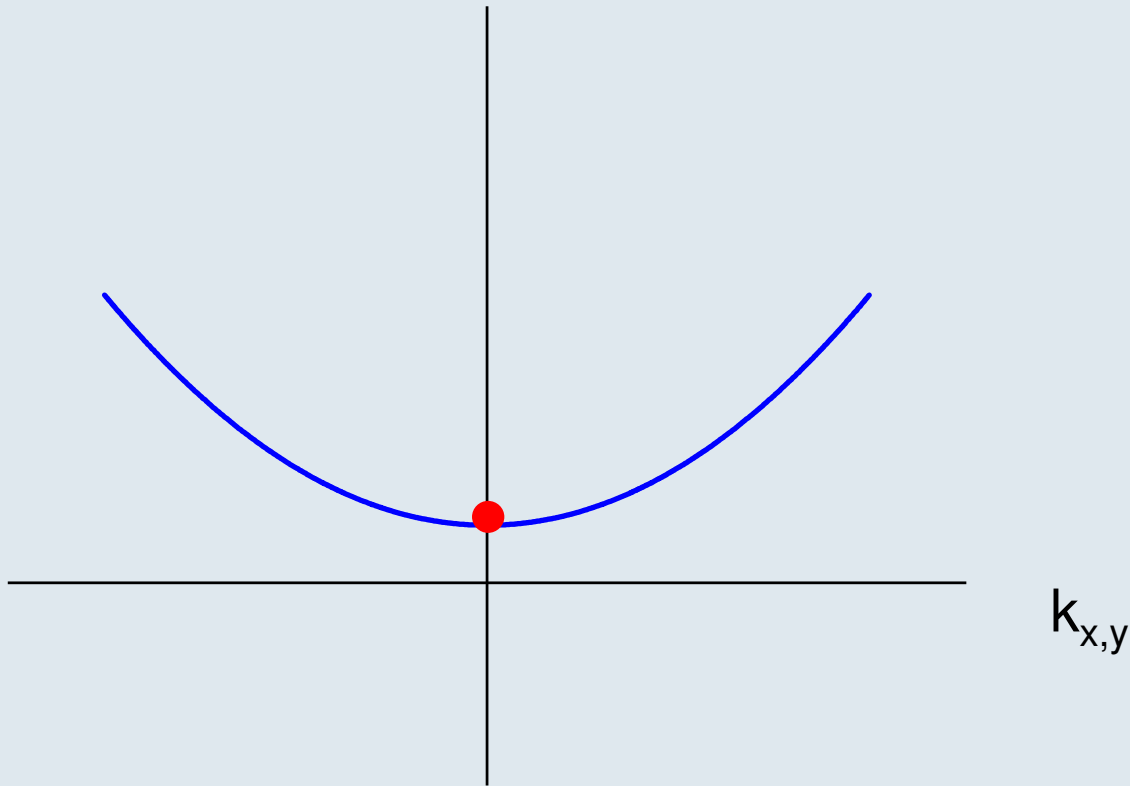
L. C. Andreani *et al.*, Phys. Rev. B **60** 13276 (1999).

Robert C. Hilborn, "Einstein coefficients, cross sections, f values, dipole moments, and all that"

<http://arxiv.org/ftp/physics/papers/0202/0202029.pdf>

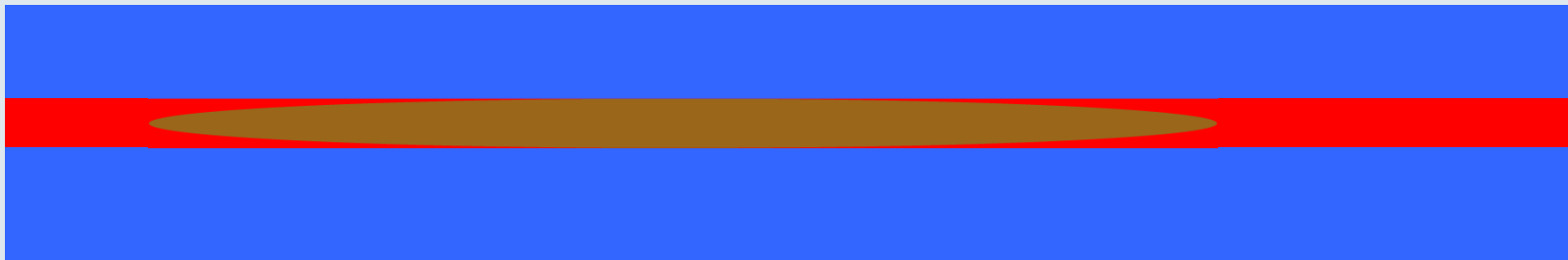
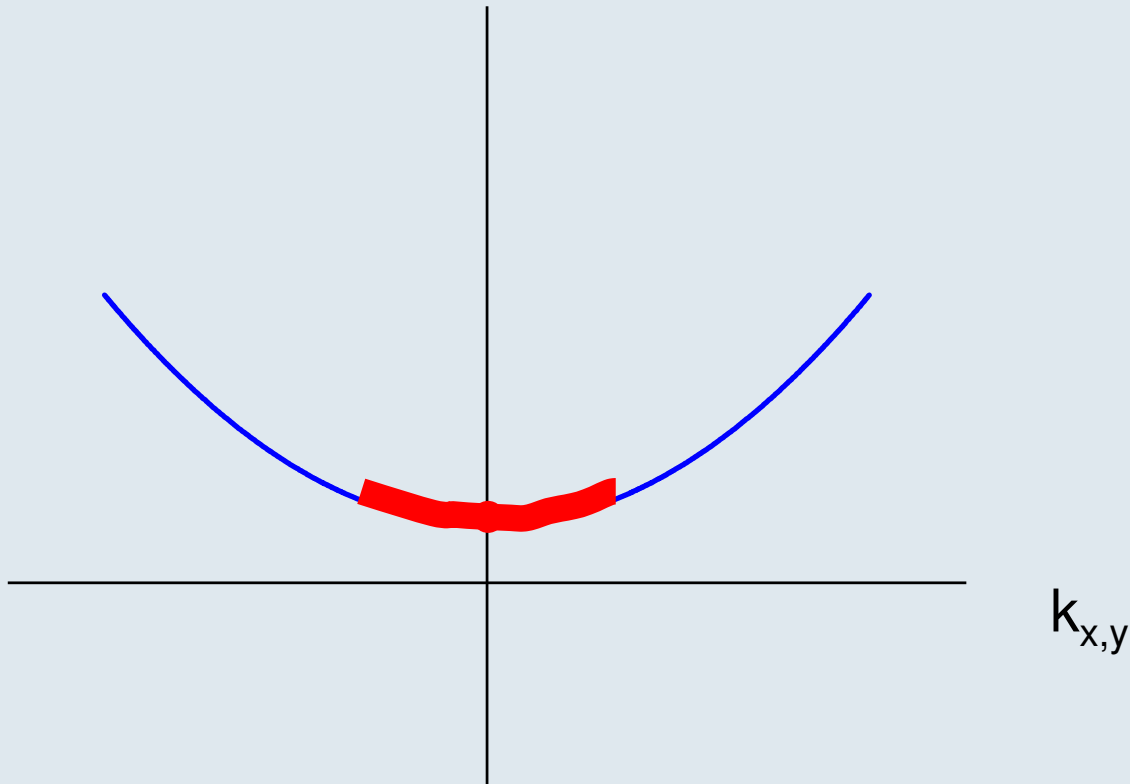
# Localization in k-space and real space

E

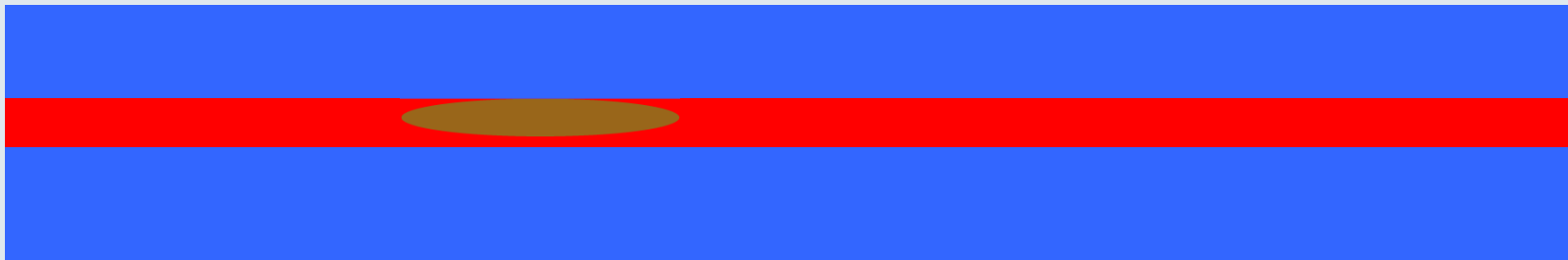
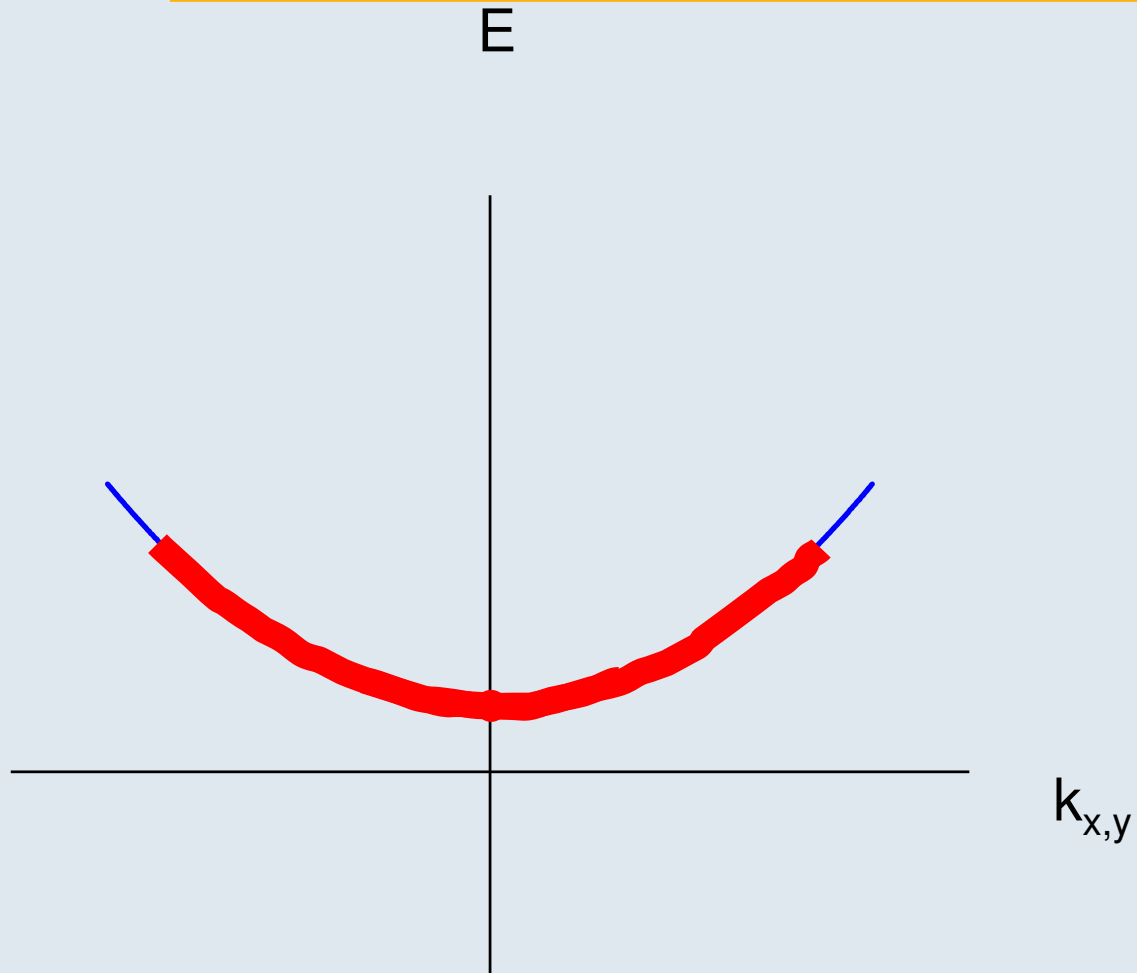


# Localization in k-space and real space

E



# Localization in k-space and real space



# Excitonic effects in QWs

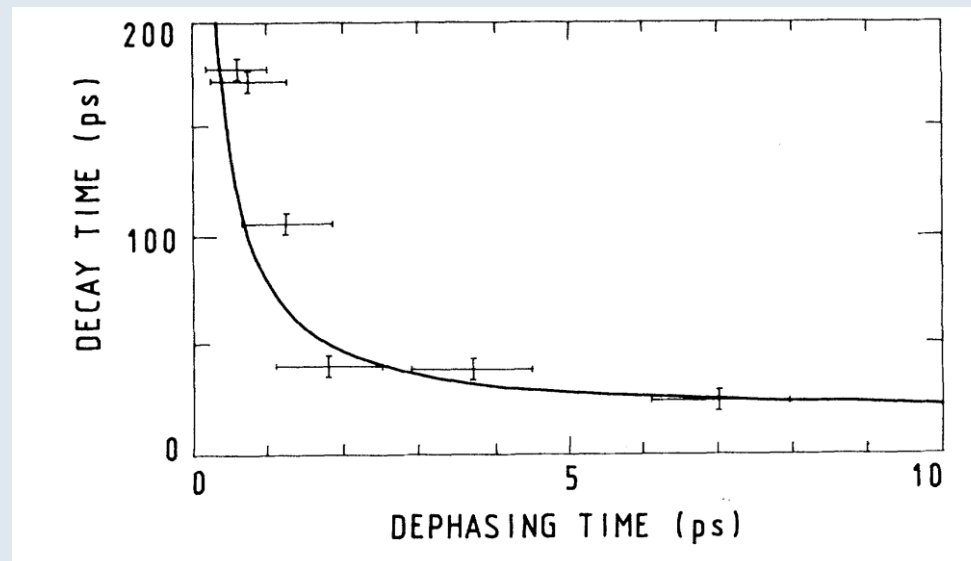
2D limit = intrinsic lifetime of two-dimensional exciton

**~ 10 ps for a GaAs/AlAs QW**

L. C. Andreani *et al.*, Solid State Commun. **77** 641 (1991)

B. Deveaud *et al.*, Phys. Rev. Lett. **67** 2355 (1991)

Dephasing (exciton-exciton or exciton-phonon scattering) limits the coherence surface and hence the radiative emission rate



# Excitonic effects in QWs

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The 2D quantum well excitons does couple efficiently to photons!

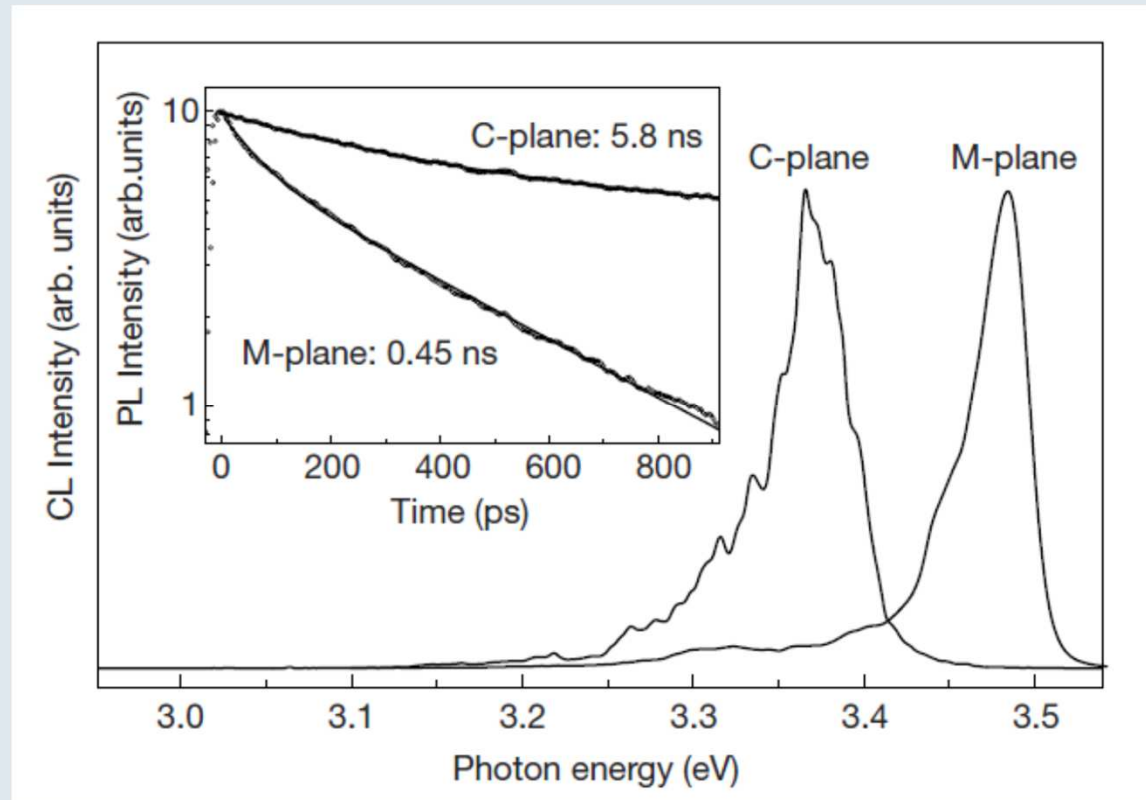
In a non perfect structure, the fundamental emitting states will be laterally localized states

Even in a very good (i.e. III-As) quantum well, it takes very peculiar experimental conditions to observe the intrinsic fast lifetimes of 2D excitons

- *Very low temperature (phonon scattering)*
- *Resonant and low excitation (carrier-carrier scattering)*

# Excitonic effects in GaN QWs

## Non-Polar QWs

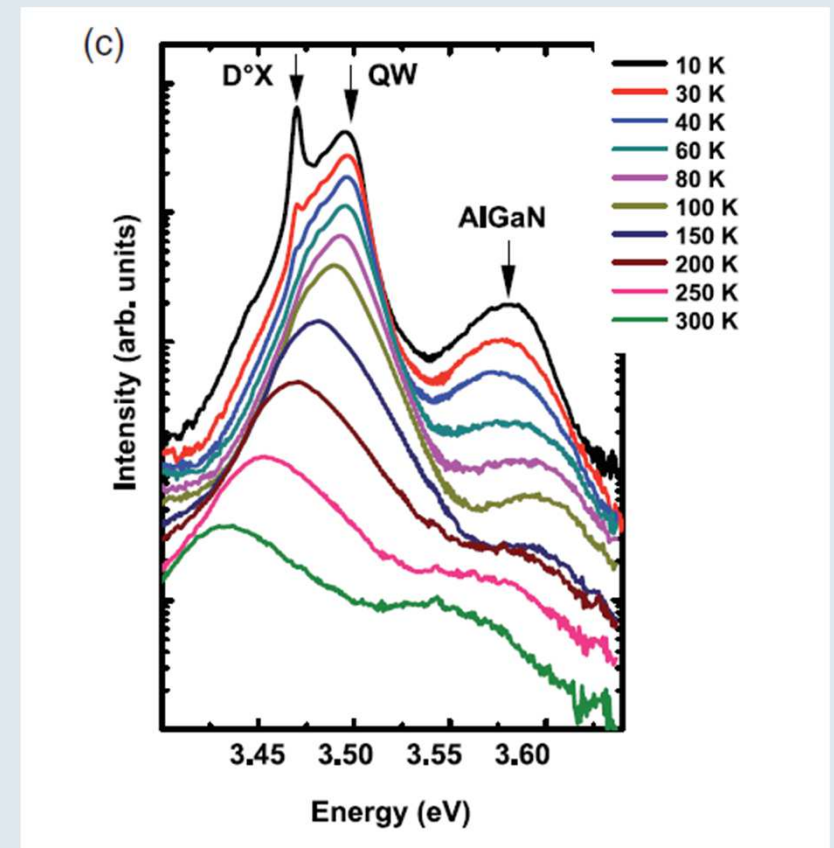
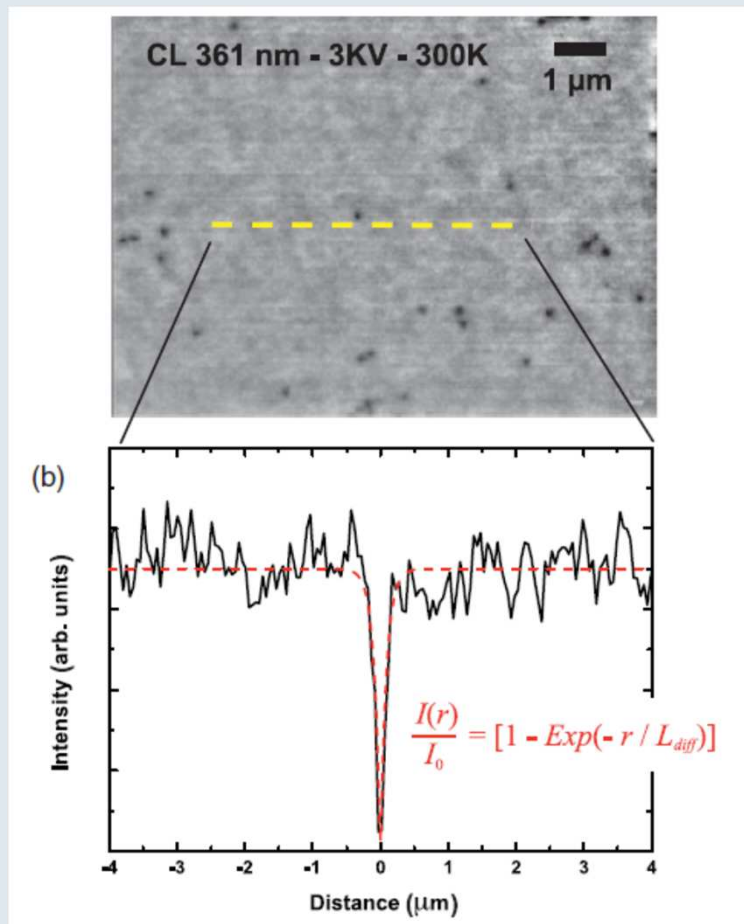


P. Waltereit *et al.*, Nature, **406** 865 (2000)

# Excitonic effects in GaN QWs

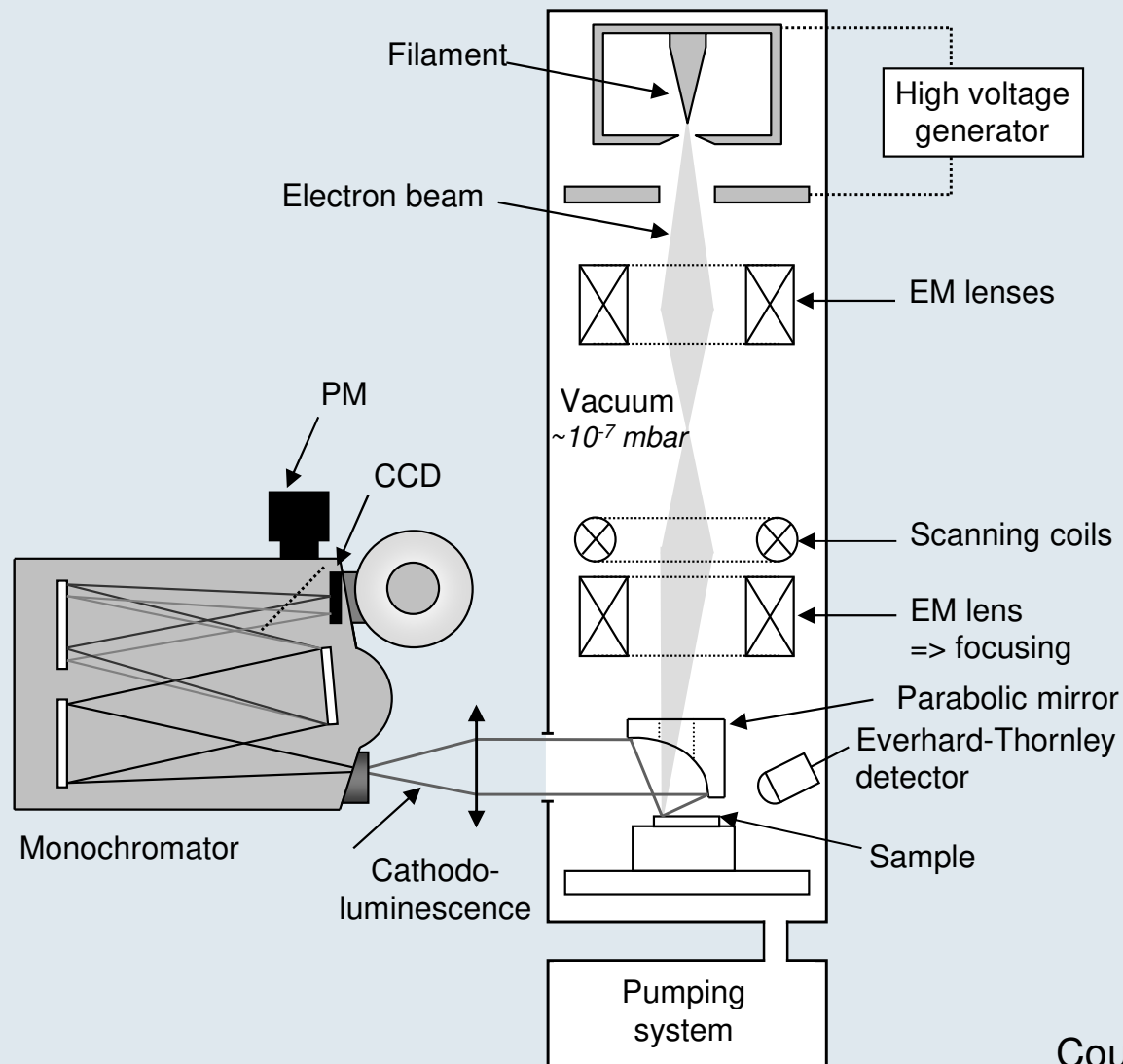
## Non-Polar QWs

GaN/Al<sub>0.06</sub>Ga<sub>0.94</sub>N QWs



P. Corfdir *et al.*, Phys. Rev. B **83** 245326 (2011)

# Experimental technique : cathodoluminescence



Courtesy F. Rol, PhD thesis 2007

# Experimental technique : cathodoluminescence

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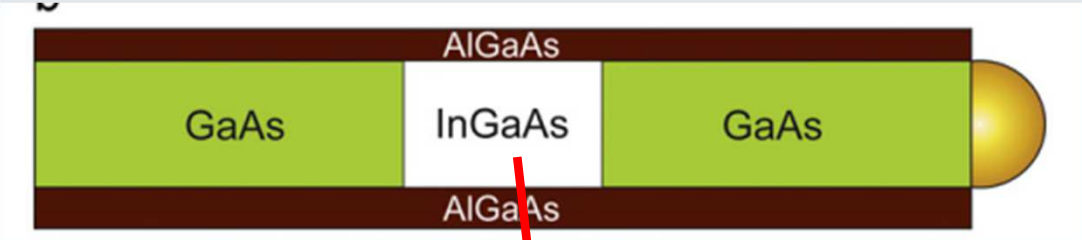
## Advantages of CL :

- Excites any semiconductor (including large gap)
- High spatial resolution
- Easy scanning of sample

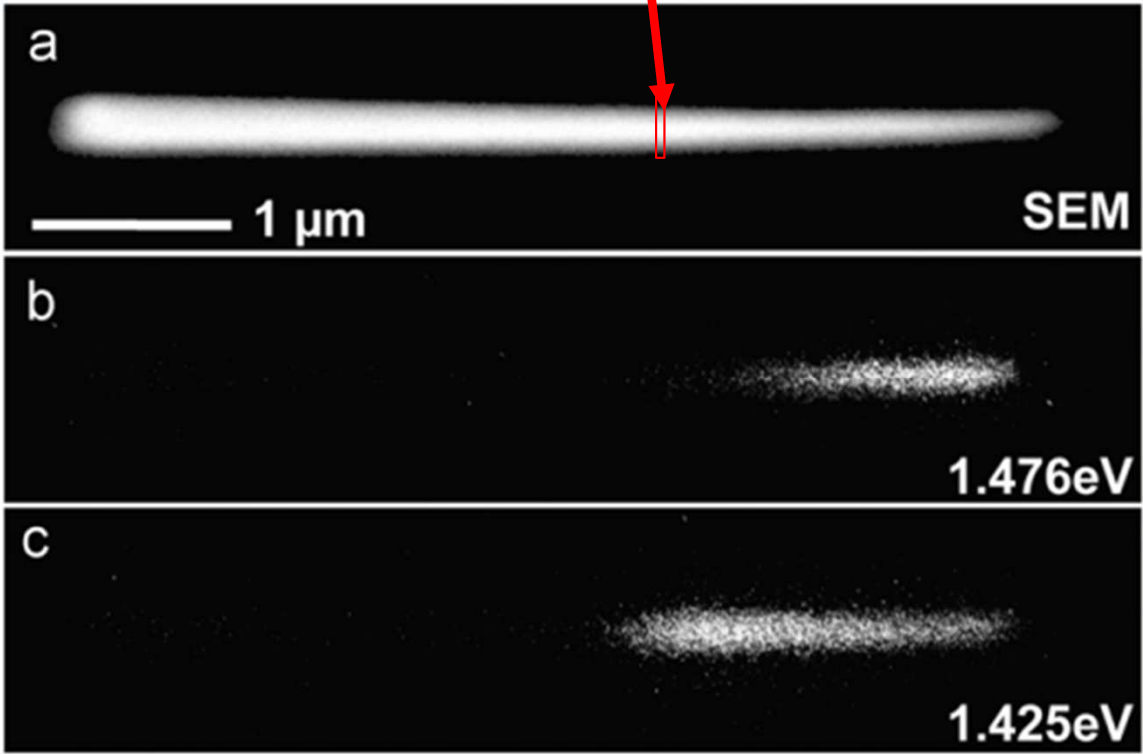
## Limitations of CL :

- Requires a conducting substrate
- Messy excitation (lots of hot carriers created)
- Low collection efficiency

# Cathodoluminescence $\neq$ imaging technique !



50 nm



GaAs luminescence

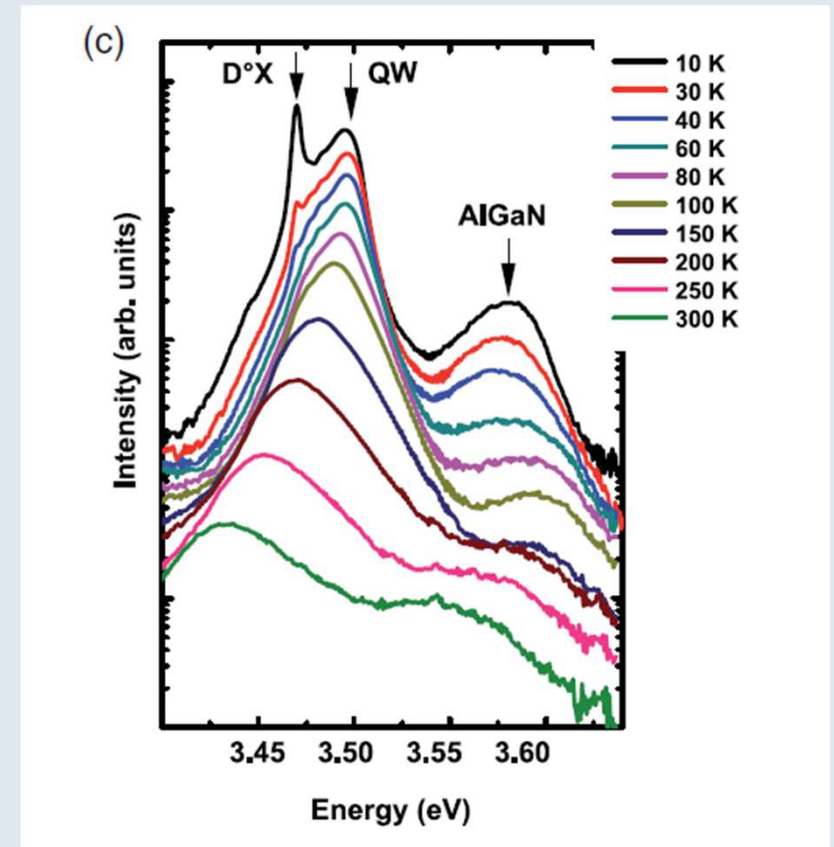
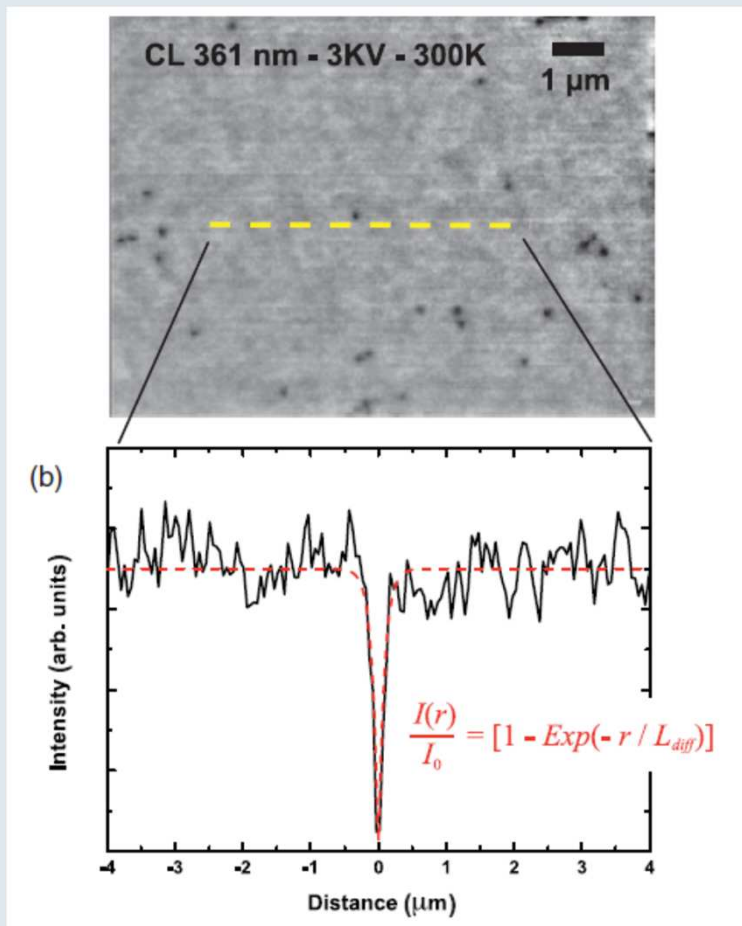
InGaAs luminescence

J. Bolinsson *et al.* *J. Cryst Growth* **315** 138 (2011)

# Excitonic effects in GaN QWs

## Non-Polar QWs

GaN/Al<sub>0.06</sub>Ga<sub>0.94</sub>N QWs  
grown on bulk GaN



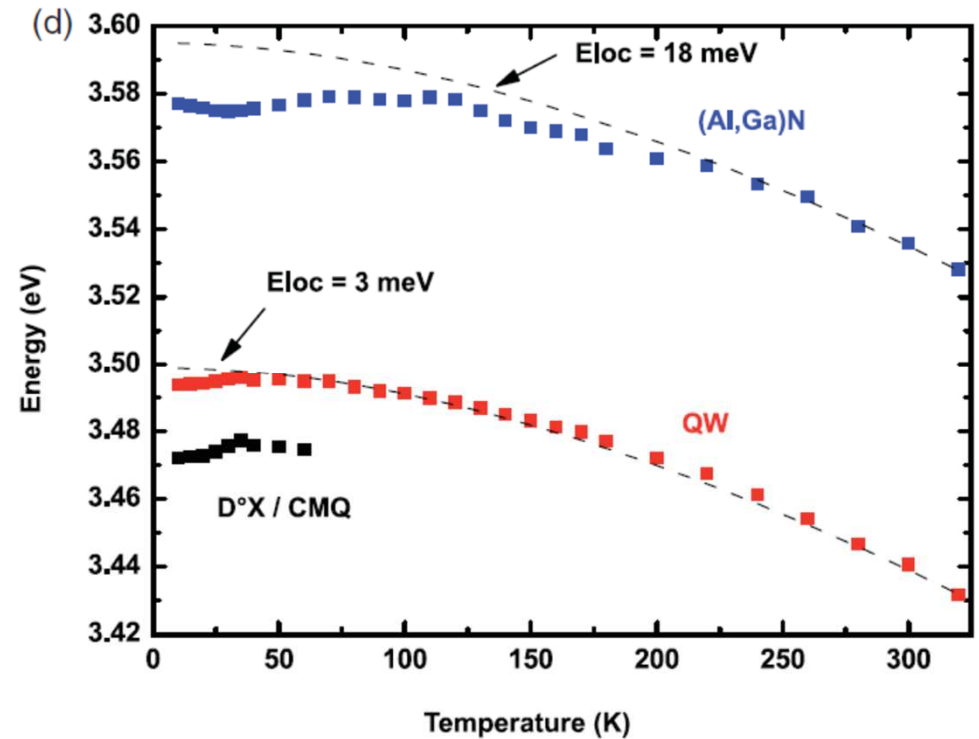
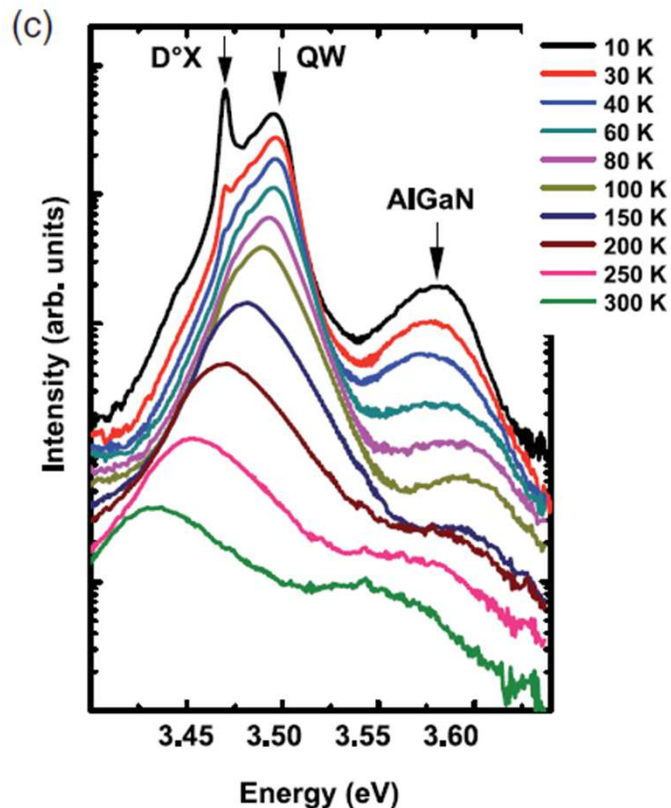
~100 nm in-plane diffusion length

P. Corfdir *et al.*, Phys. Rev. B **83** 245326 (2011)

# Excitonic effects in GaN QWs

## Non-Polar QWs

GaN/Al<sub>0.06</sub>Ga<sub>0.94</sub>N QWs



P. Corfdir *et al.*, Phys. Rev. B **83** 245326 (2011)

# Temperature dependence of GaN bandgap

.... beyond the Varshni empirical law

phys. stat. sol. (b) **200**, 155 (1997)

Subject classification: 71.20.Nr; S5.11, S5.12, S7.12, S7.14, S8.11, S8.12

## Basic Model Relations for Temperature Dependencies of Fundamental Energy Gaps in Semiconductors

R. PÄSSLER<sup>1)</sup>

*Institut für Physik, Technische Universität Chemnitz–Zwickau,  
D-09107 Chemnitz, Germany*

JOURNAL OF APPLIED PHYSICS

VOLUME 90, NUMBER 8

15 OCTOBER 2001

## Dispersion-related assessments of temperature dependences for the fundamental band gap of hexagonal GaN

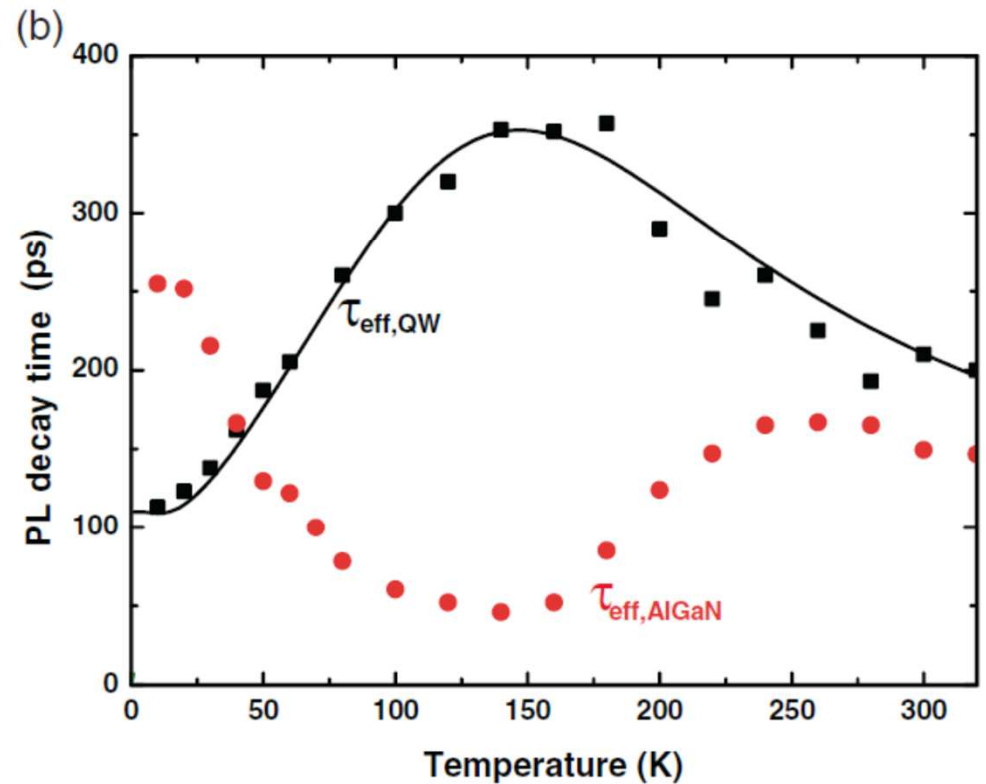
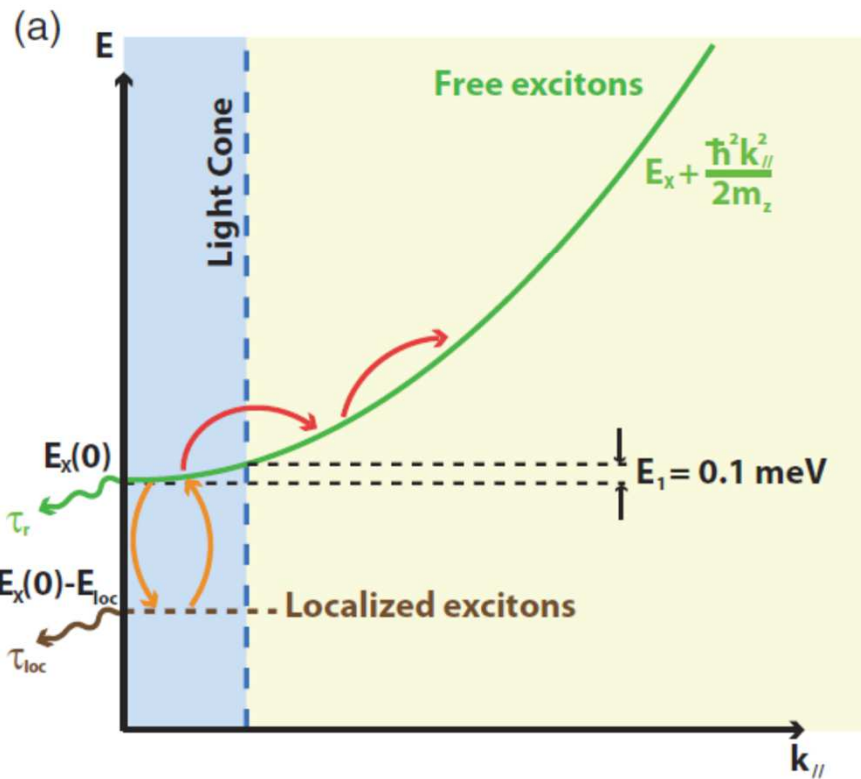
R. Pässler<sup>a)</sup>

*Technische Universität Chemnitz, Institut für Physik, D-09107 Chemnitz, Germany*

+ other references by R. Pässler

# Excitonic effects in GaN QWs

## Non-Polar QWs



P. Corfdir *et al.*, Phys. Rev. B **83** 245326 (2011)

# Excitonic effects in GaN QWs

*Is 100 ps @ 3.5 eV a fast recombination??*

$$\tau_{\text{rad}} \propto \frac{1}{nE^2f}$$

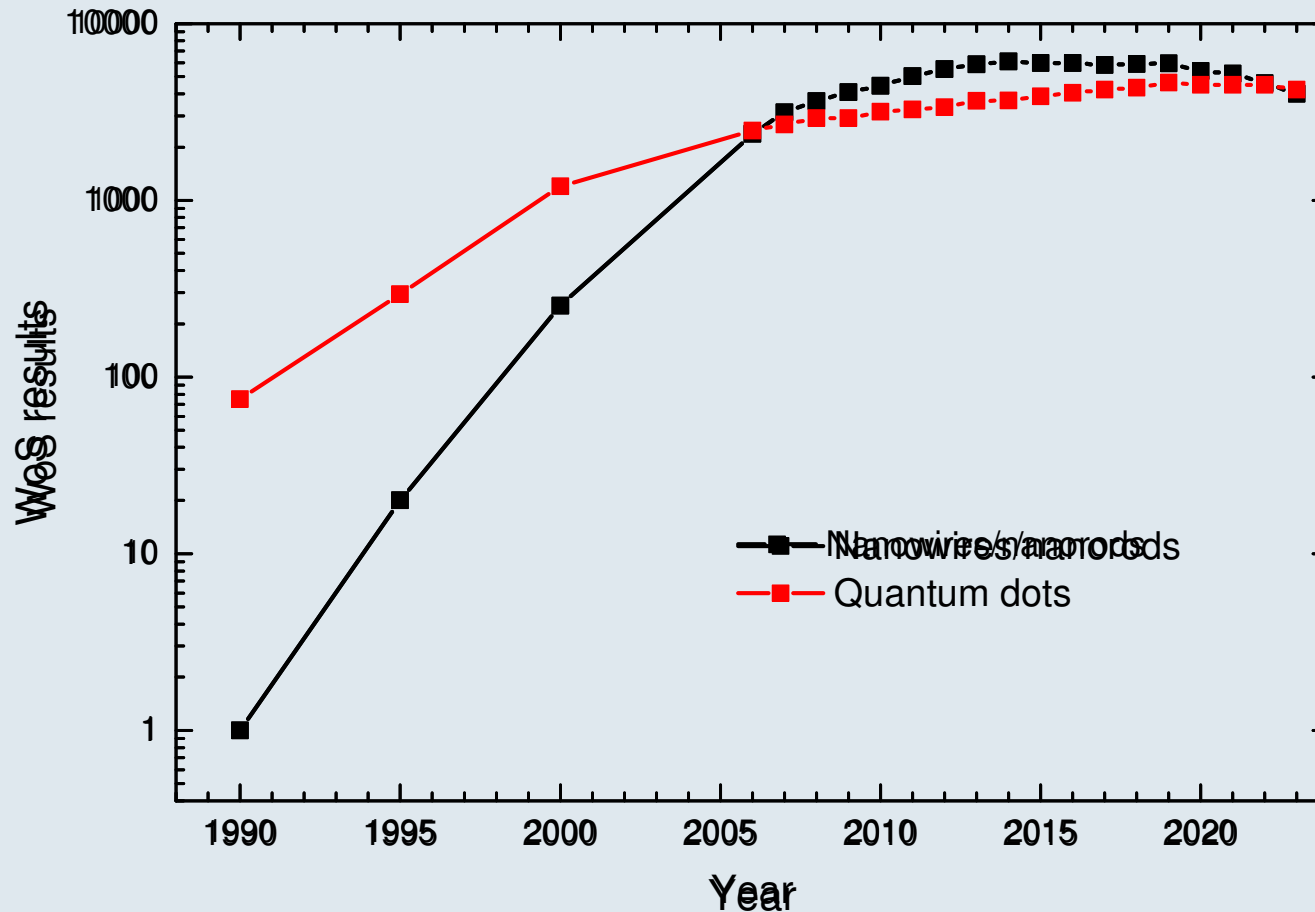
GaAs QW emitting at 1.7 eV with a lifetime of 25 ps : **f=320**

GaN QW emitting at 3.5 eV with a lifetime of 100 ps : **f=24**

⇒ Quite strong localization effects, no giant oscillator strength

⇒ Strong excitonic effects in III-N, but ordinary recombination rates

# Context : Nanowires



Physics Today, 50(12), Dec. 1997, p. 62

# Context : nitride nanowires

Jpn. J. Appl. Phys. Vol. 36 (1997) pp. L 459–L 462  
Part 2, No. 4B, 15 April 1997

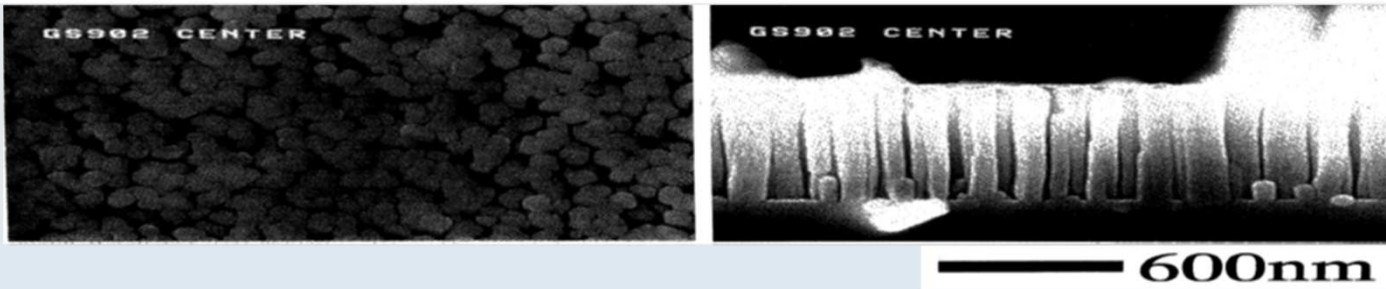
## Growth of Self-Organized GaN Nanostructures on $\text{Al}_2\text{O}_3(0001)$ by RF-Radical Source Molecular Beam Epitaxy

Masaki YOSHIKAWA, Akihiko KIKUCHI, Masashi MORI, Nobuhiko FUJITA and Katsumi KISHINO  
*Department of Electrical and Electronics Engineering, Sophia University, 7-1 Kioi-cho, Chiyoda-ku, Tokyo 102, Japan*

(Received July 18, 1996; accepted for publication February 28, 1997)

Columnar GaN nanostructures (GaN nanocolumns) were grown on  $\text{Al}_2\text{O}_3(0001)$  by RF-radical source molecular beam epitaxy (RF-MBE) through a self-organization process. The nanocolumns were grown at high density, with the *c*-axis maintained perpendicular to the substrate surface. The dependence of column diameter and density on growth conditions was systematically investigated. The average diameter was minimized to 53 nm and the density of the GaN nanocolumns was  $130 \times 10^{12}$  columns per square meter.

KEYWORDS: self-organization, GaN, nanostructure, nanocolumn,  $\text{Al}_2\text{O}_3(0001)$ , RF-MBE



# Context : nitride nanowires

PHYSICAL REVIEW B

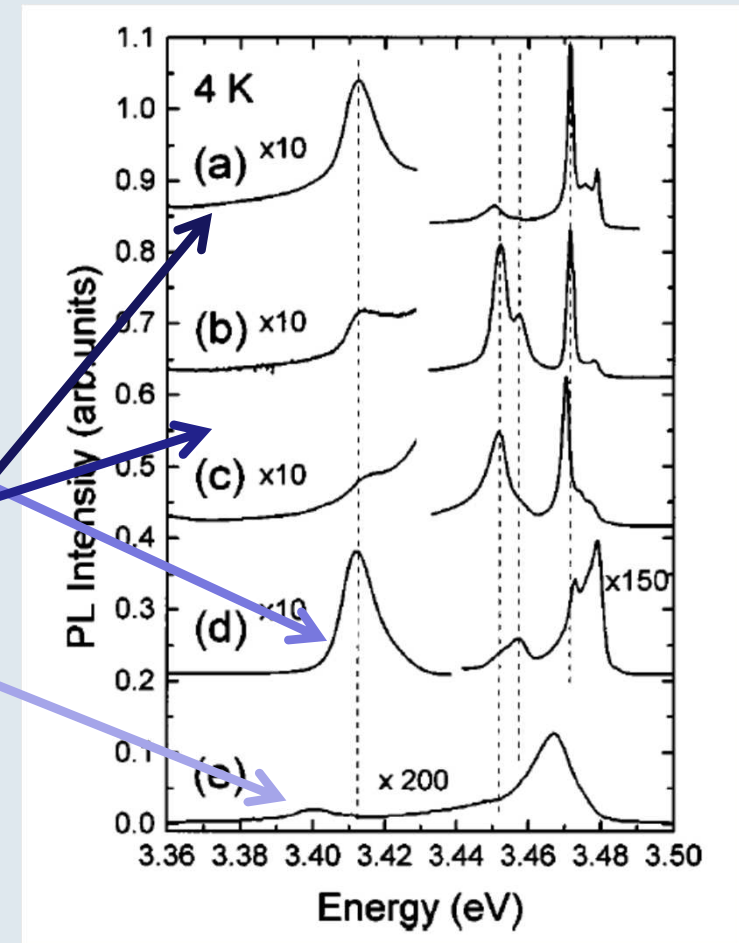
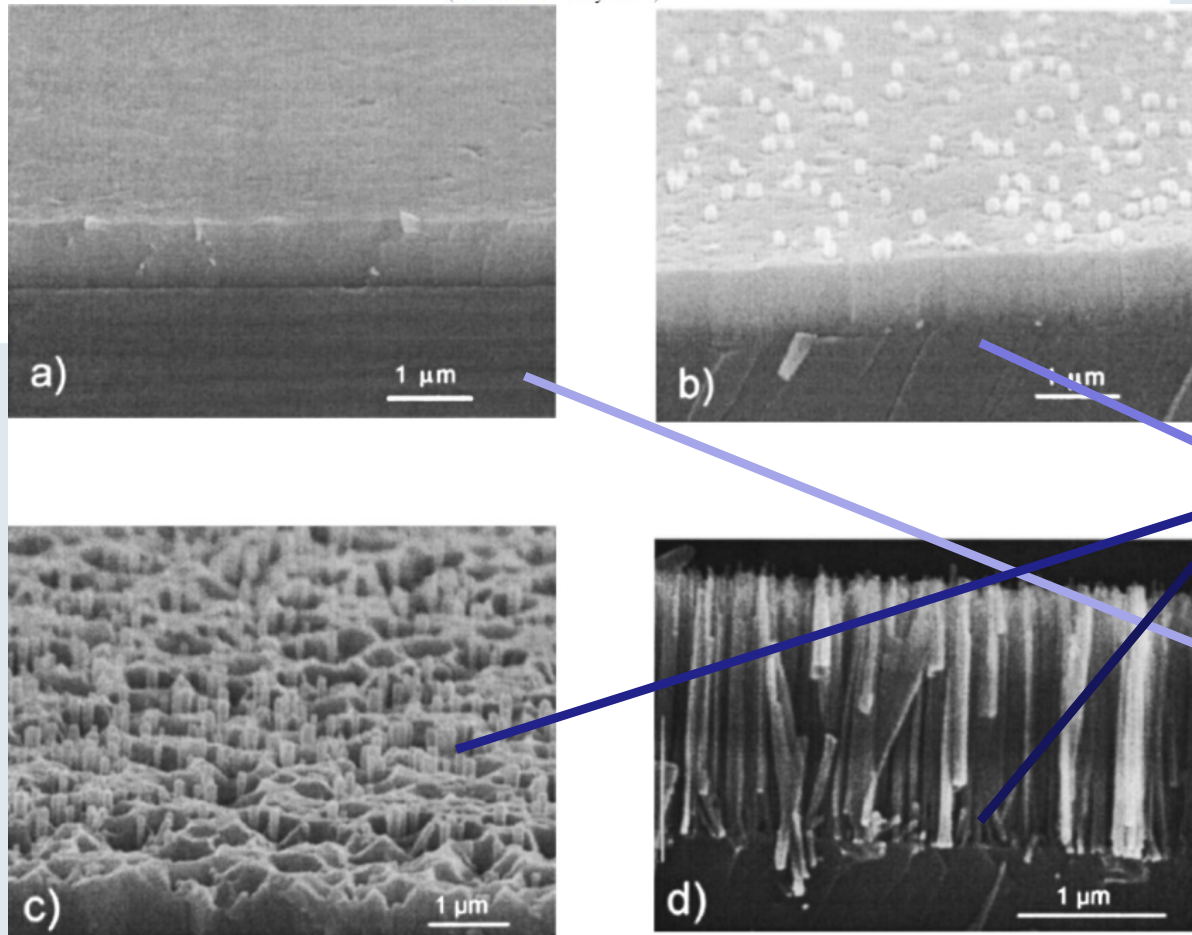
VOLUME 62, NUMBER 24

15 DECEMBER 2000-II

## Luminescence properties and defects in GaN nanocolumns grown by molecular beam epitaxy

E. Calleja, M. A. Sánchez-García, F. J. Sánchez, F. Calle, F. B. Naranjo, and E. Muñoz  
*Departamento Ingeniería Electrónica, ETSI Telecomunicación, Universidad Politécnica, 28040 Madrid, Spain*

U. Jahn and K. Ploog  
*Paul Drude Institut für Festkörperelektronik, Hausvogteiplatz 5-7, D-10117 Berlin, Germany*  
(Received 17 May 2000)



# GaN nanowires : fundamental studies

Can nanowires help answer fundamental questions about GaN?

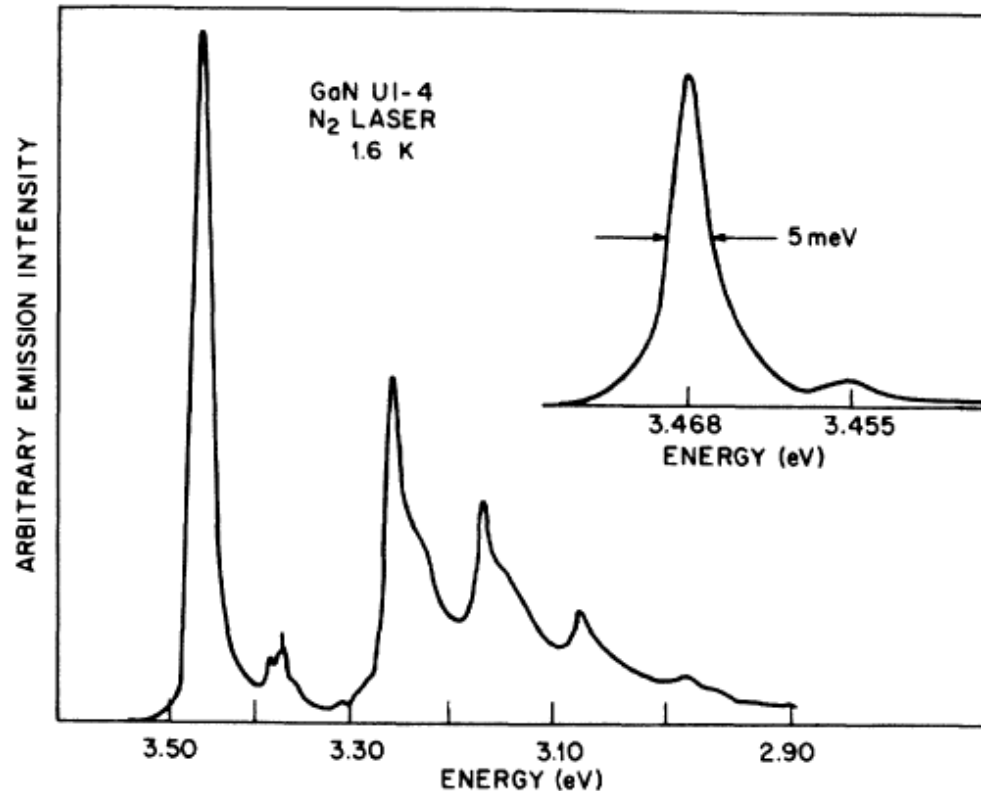
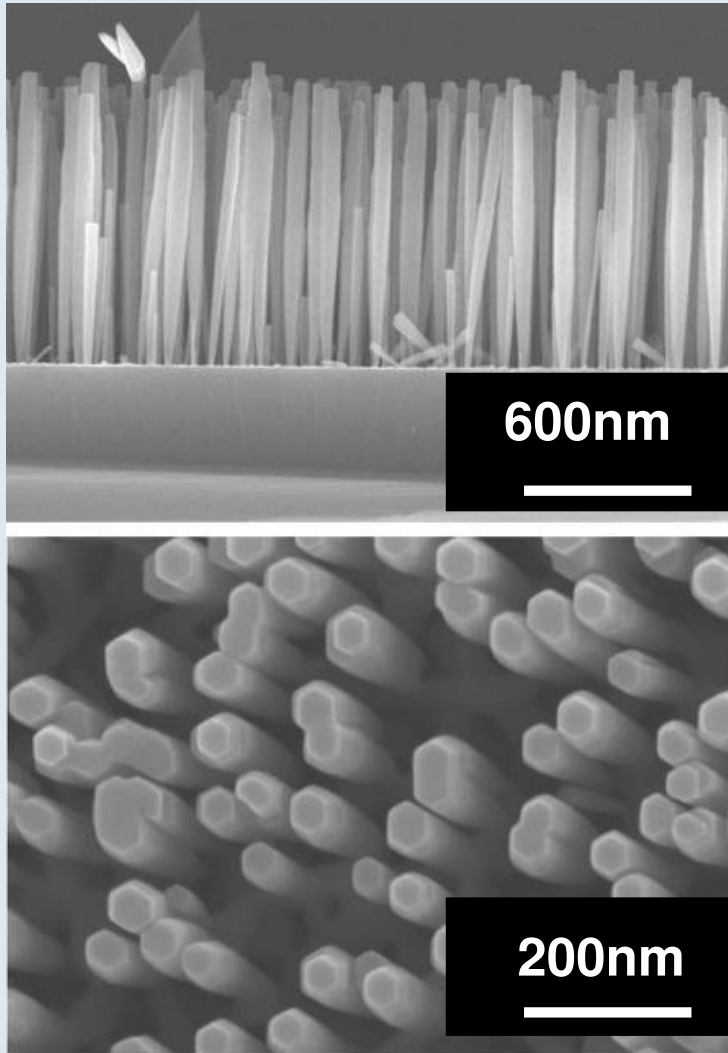


FIG. 5. Typical photoluminescence spectrum of type-I GaN at 1.6 K. The insert is a more detailed view of the strong high-energy peak near 3.468 eV, taken with low excitation intensity and high resolution.

R. Dingle *et al.*, Phys. Rev. B **4** 1211 (1971)

# Fundamental properties of MBE grown GaN nanowires



Plasma-assisted MBE

N-rich conditions

Si (111) substrate

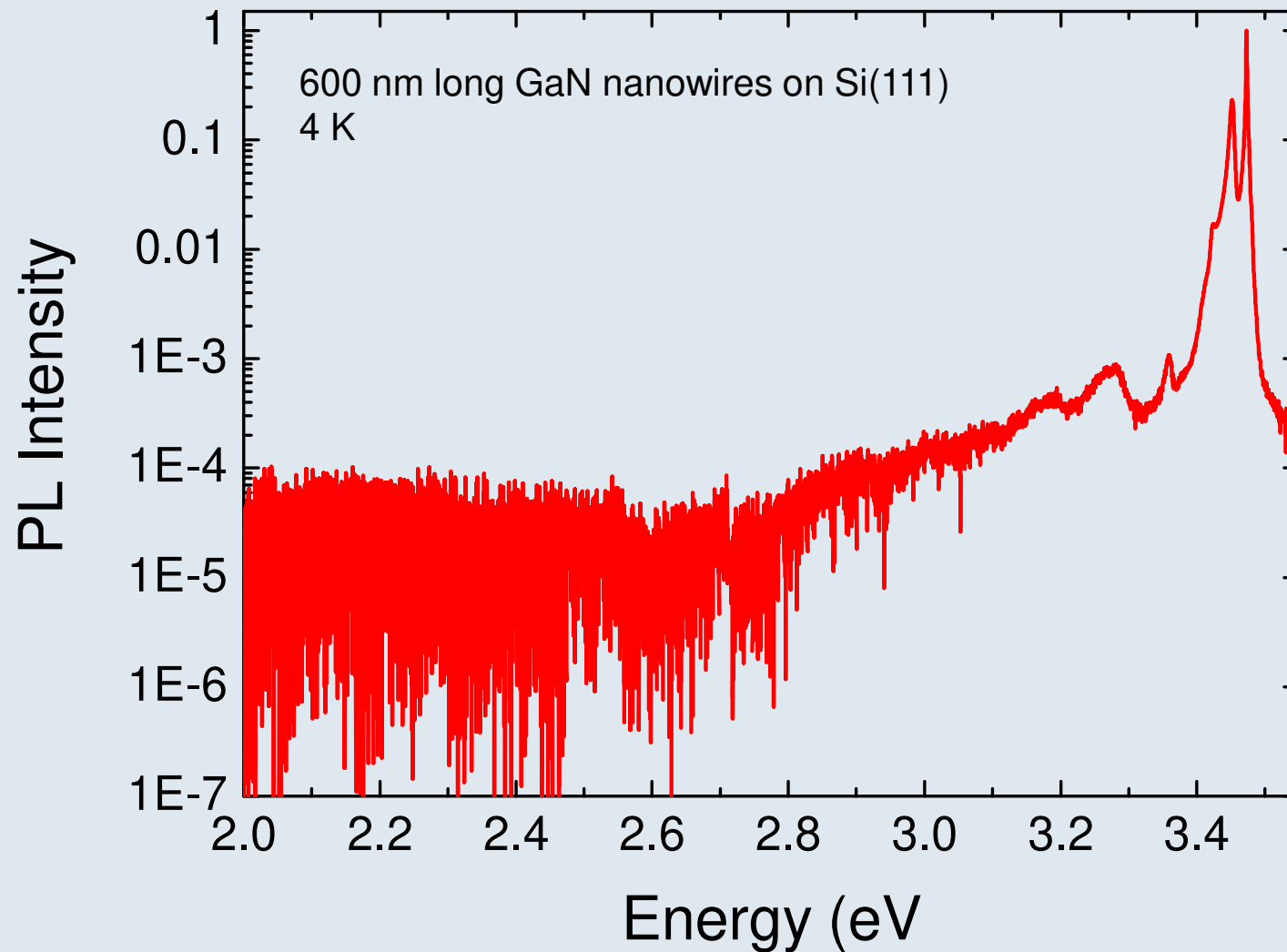
M.A. Sanchez-Garcia *et al.*, J. Crystal Growth **183** 23 (1998)

**Density :  $10^{10} \text{ cm}^{-2}$**

**Diameter :  $\sim 40 \text{ nm}$**

MBE growth: R. Songmuang *et al.* APL **91**, 251902 (2007)

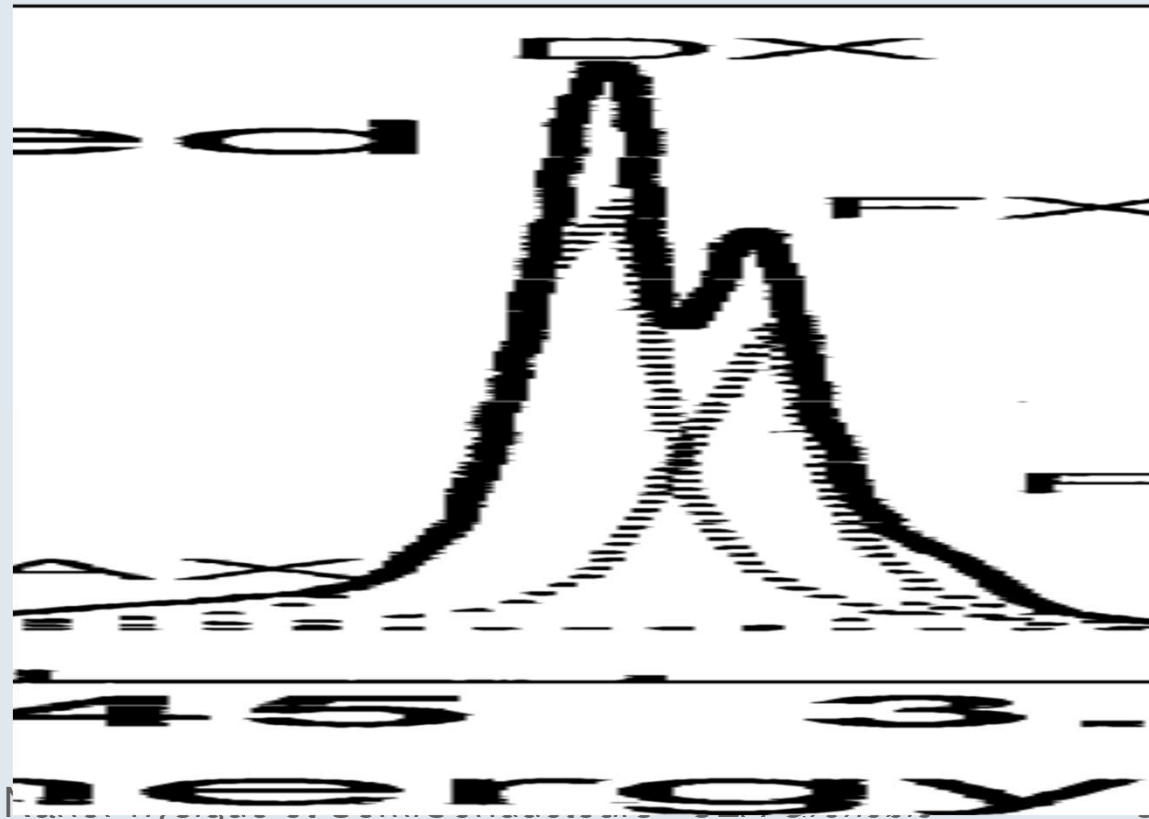
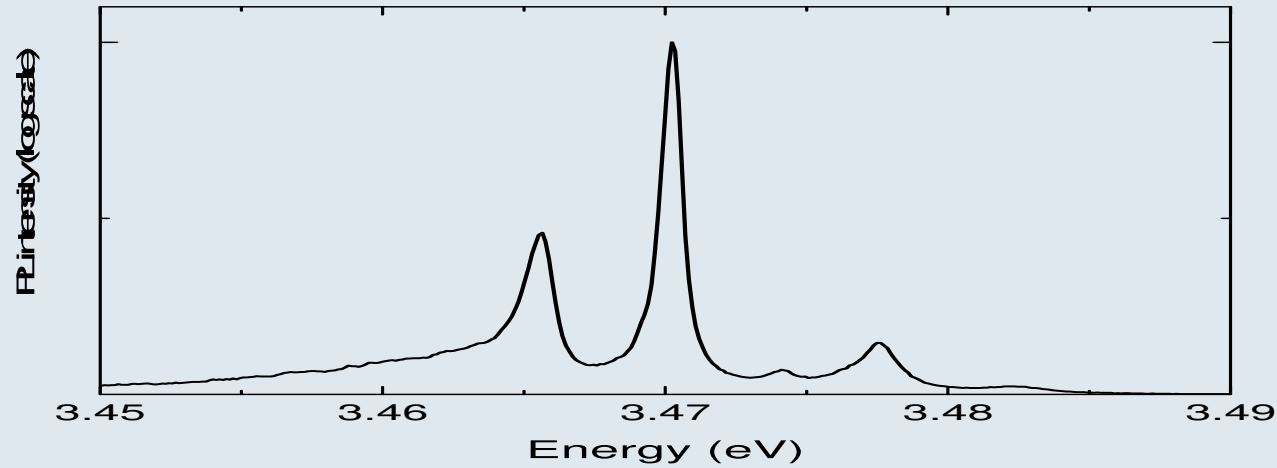
# GaN nanowires : bulk properties



No deep defect (yellow) band

# GaN nanowires : bulk properties

Near Band Edge PL of GaN:Mg nanowires, 4K

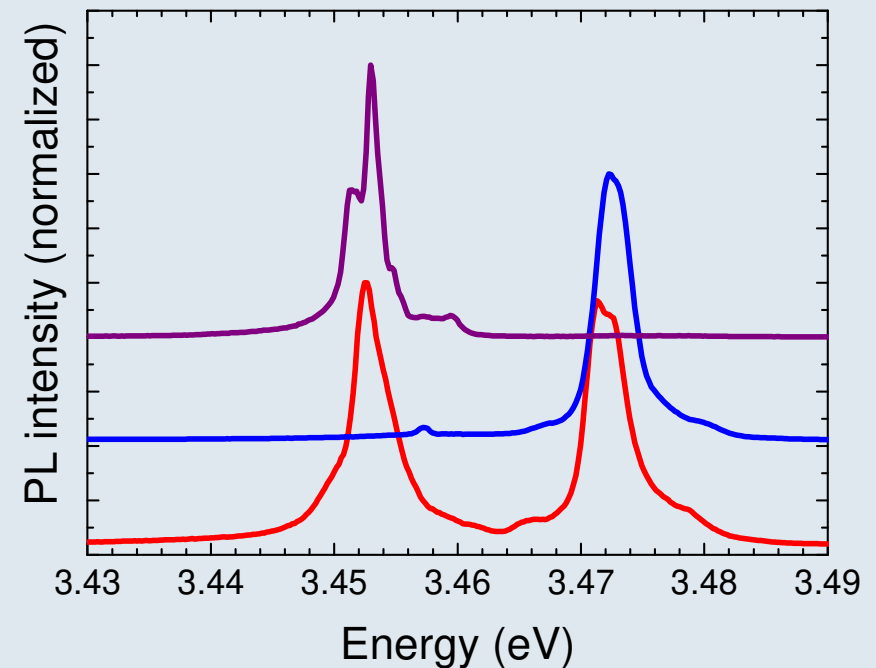
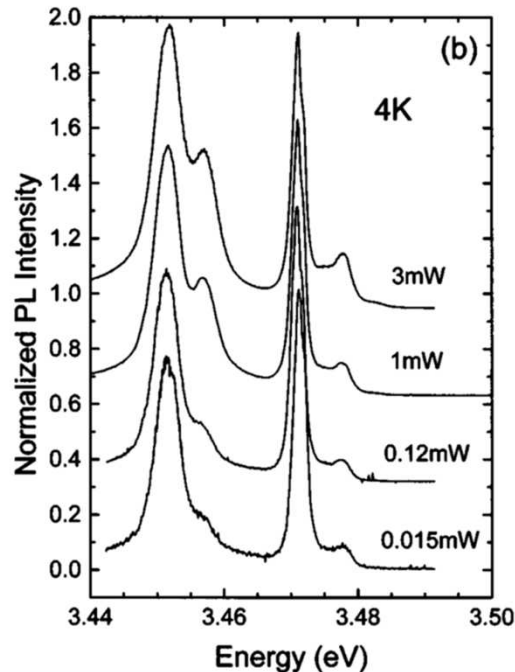


3  $\mu\text{m}$  GaN on sapphire

# A mysterious emission line

Many discussions on **3.45 eV** luminescence line in PA-MBE-grown GaN nanowires

$\mu$ PL on single NWs

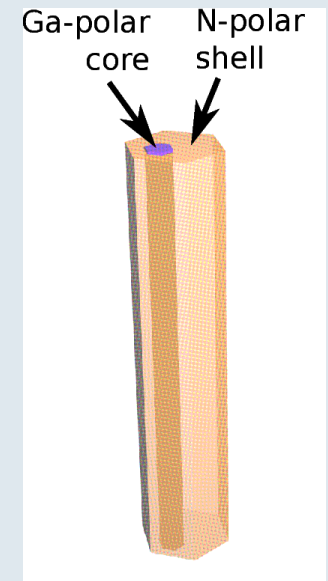
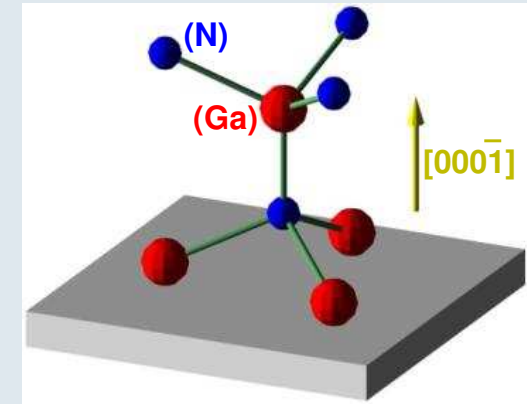
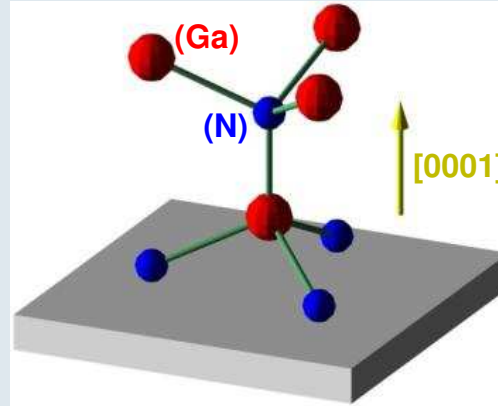
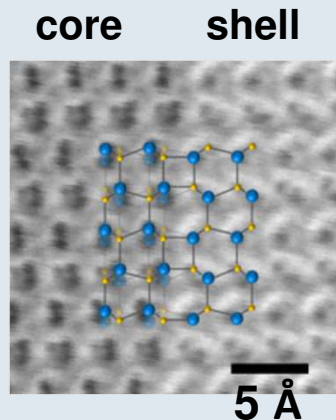
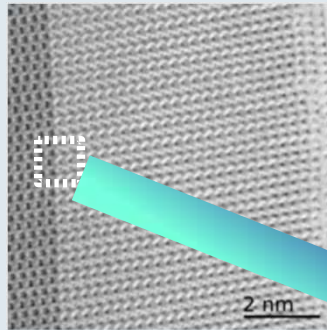
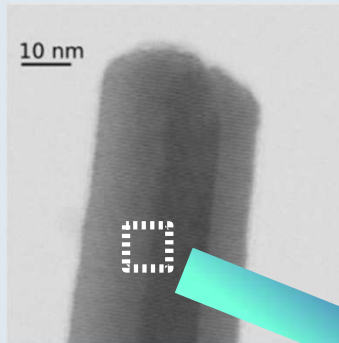


E. Calleja *et al.*, PRB **62** 16826 (2000)

D. Sam-Giao *et al.*, JAP **113** 043102 (2013)

# A mysterious emission line

Many discussions on **polarity** of  
PA-MBE-grown GaN nanowires  
-N or -Ga ??

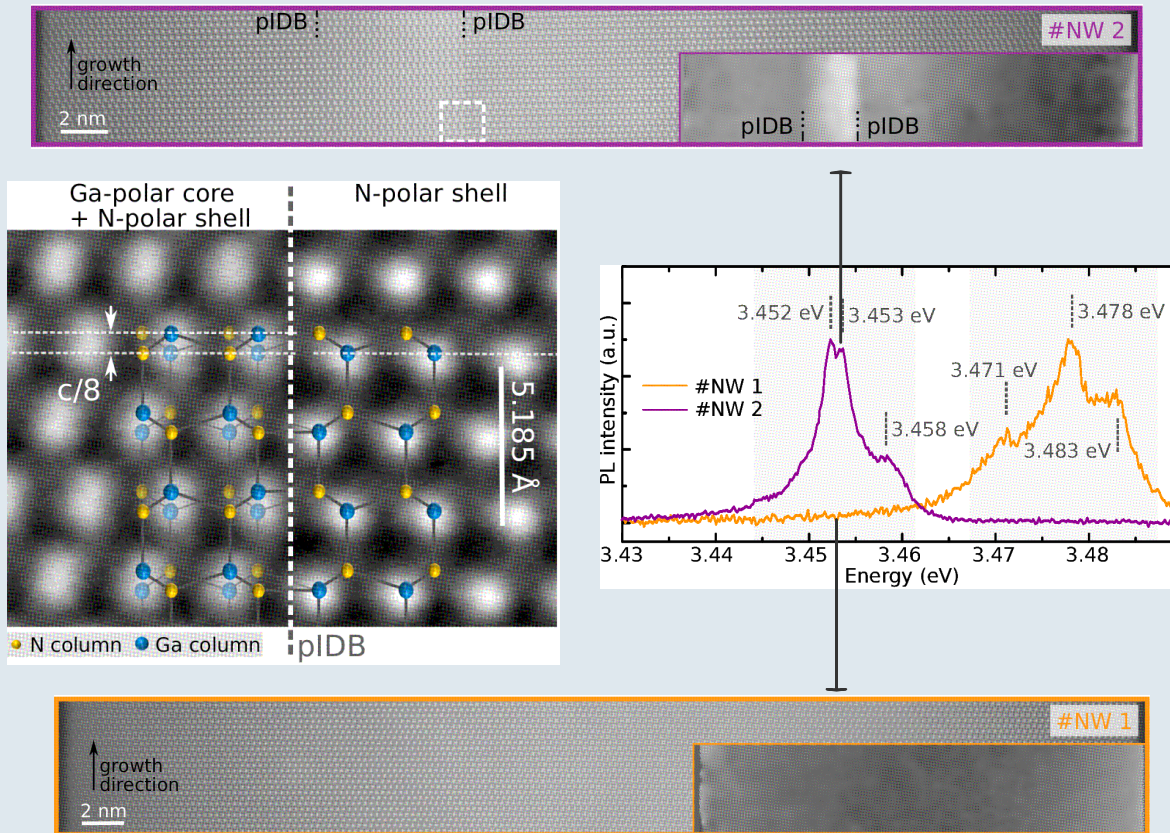


## Core-shell structures !!

T. Auzelle *et al.*, J. Appl. Phys. **117**, 245303 (2015)

# Inversion domain bound exciton

Correlated microPL and TEM studies on single nanowires



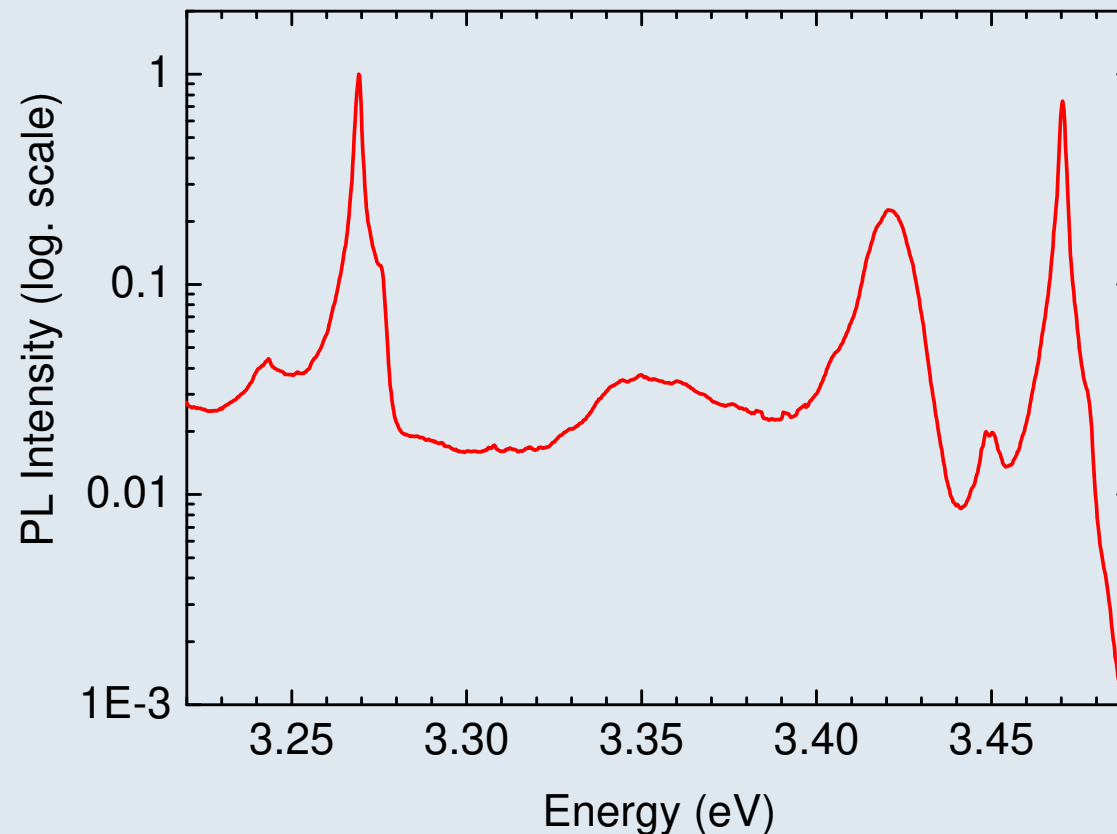
Systematic **correlated existence** of an inversion domain boundary (IDB) and a 3.45 eV luminescence.

T. Auzelle *et al.*, Appl. Phys. Lett. **107**, 051904 (2015)

# GaN nanowires : cubic phase

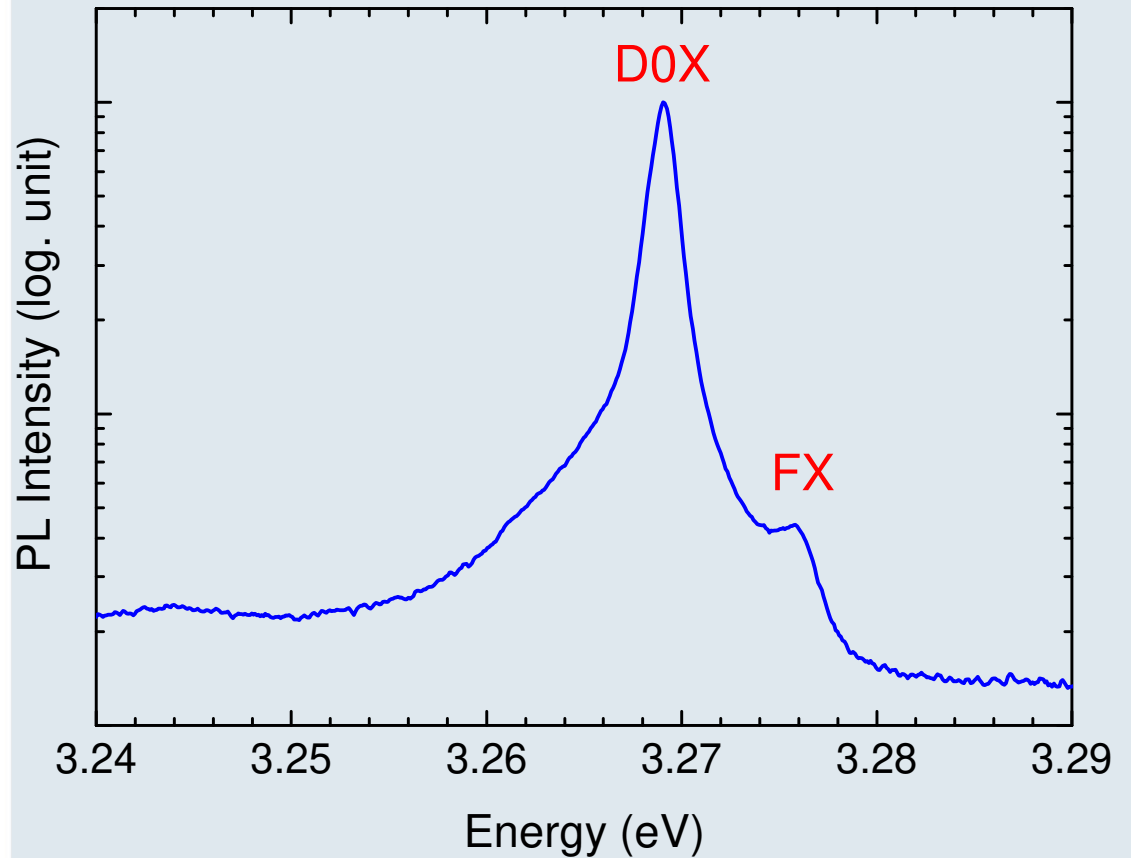
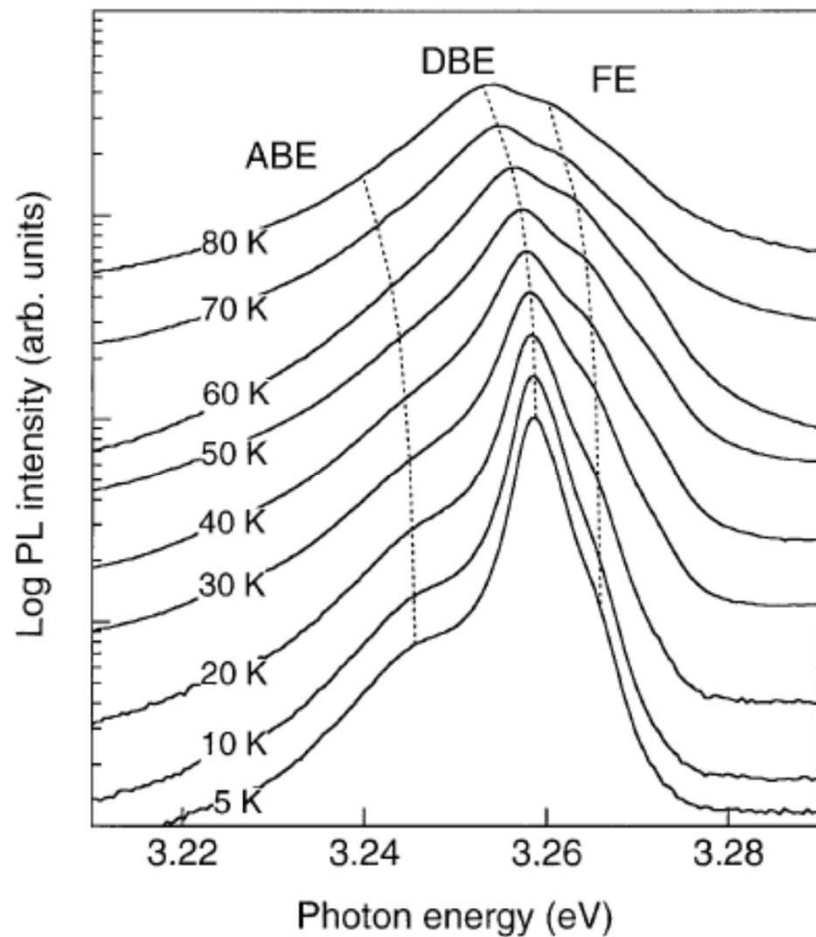


**Cubic phase** GaN by lowering the growth temperature



# GaN nanowires : cubic phase

**Cubic phase** GaN by lowering the growth temperature

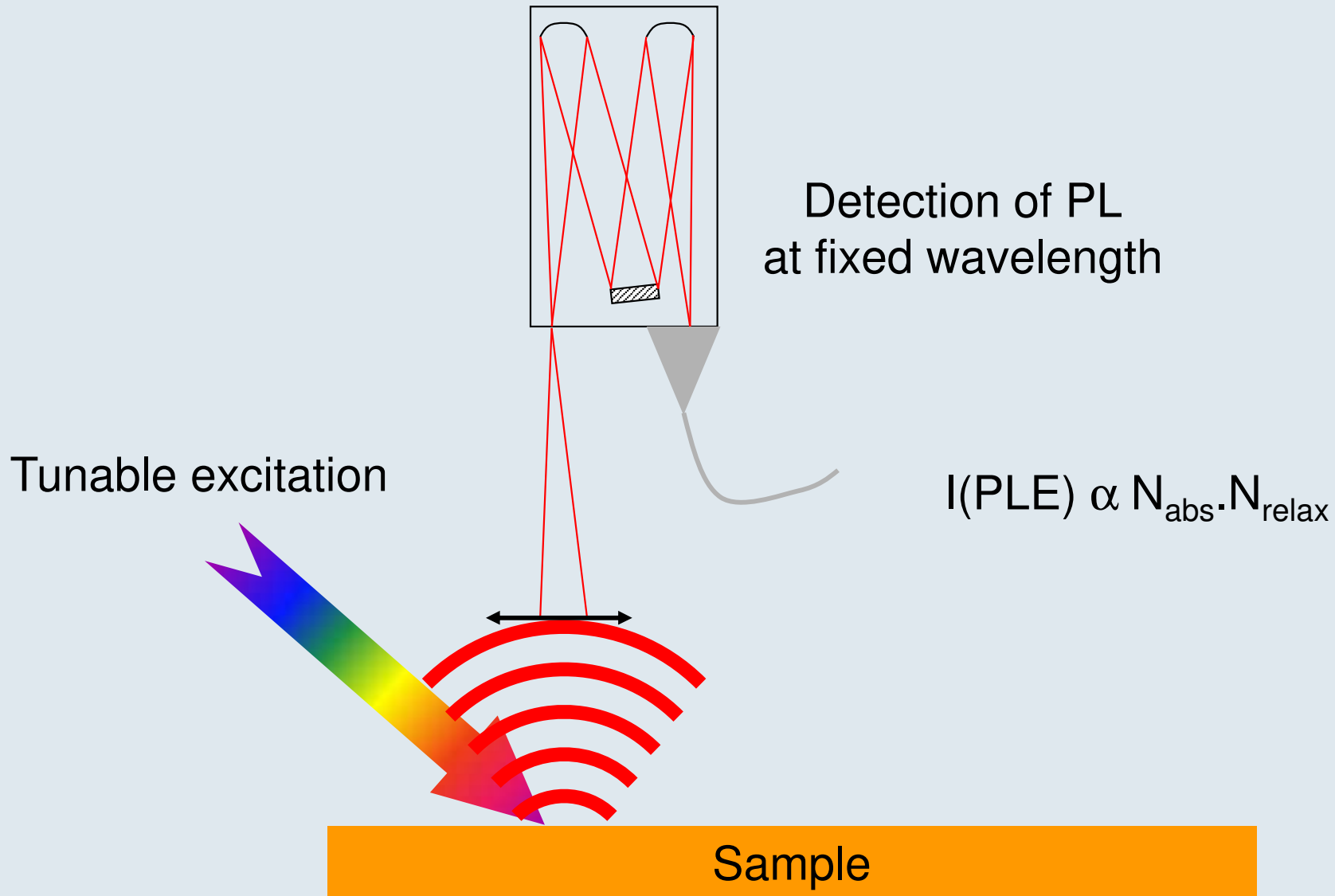


**D0X FWHM : 1.6 meV**  
**(previous state-of-the-art : 5 meV)**

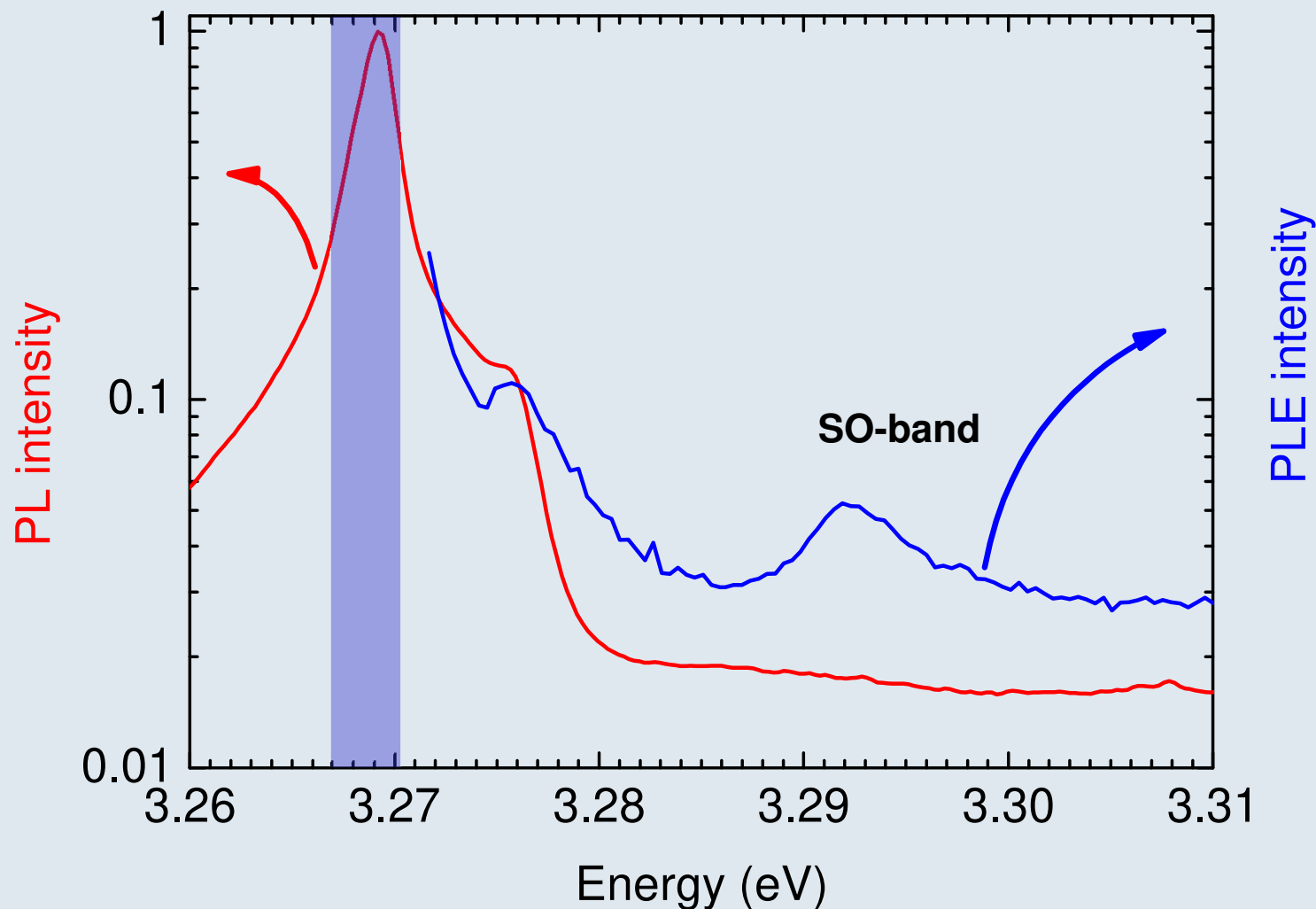
J. Renard *et al.*,  
Appl. Phys. Lett. **97** 081910 (2010)

# Experimental technique : PLE

=> Photoluminescence excitation



# GaN nanowires : cubic phase



Measurement of split-off exciton spectral position