



October 2004



Temporal and spatial-resolved spectroscopy of InGaN/GaN

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collaborated with

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¹Caltech, USA, ²Kyoto Univ., Japan



Motivation

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Development of optical devices



Light emitting diode (LED), Laser diode (LD),
Optical Memory, Optical communication
Optical computing, 3D-hologram display

(1) Elucidation of the optical properties and function of several materials.

Compound Semiconductor (**GaN**, ZnSe, ZnO-based quantum well, dot, etc.)

Organic thin films (Polysilane Films, Alq₃-based EL, etc.)

Nano-particles of Metal, Semiconductor (Pt, Au, CdS, etc.)

Molecular organized system (solvation, micell, LB-layer, liquid crystal, etc.)

Biological living cell (Onion, Drosera capensis, Nerve cell of mouse, dopamine), **etc.**

(2) Development of the unique laser spectroscopy with time and spatial resolution

Time resolved micro photoluminescence spectroscopy (TR- μ -PL)

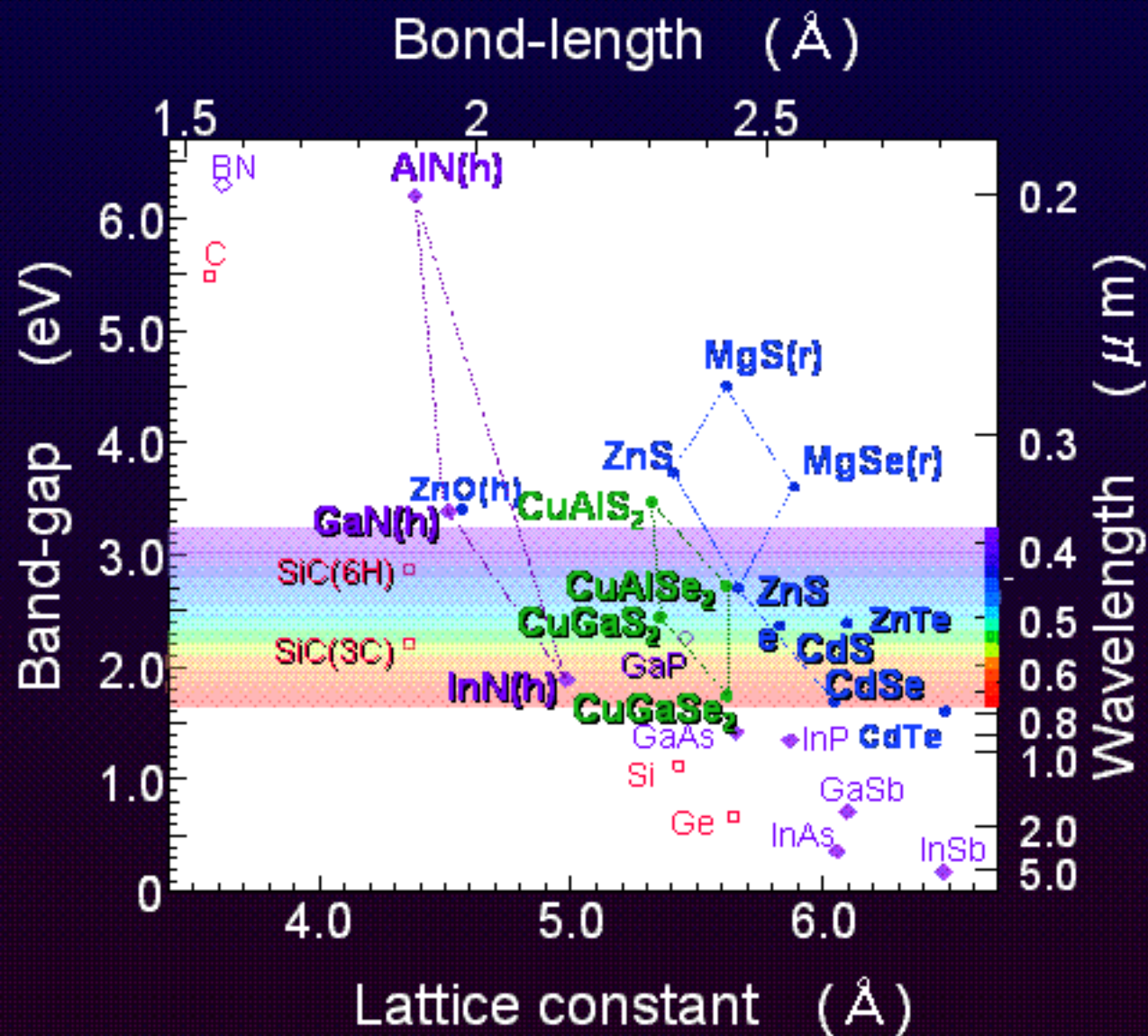
Scanning near-field optical microscopy (SNOM(=NSOM)) with i-c mode.

Third order nonlinear optical spectroscopy - Transient grating (TG) method, etc.



Bandgap of semiconductors

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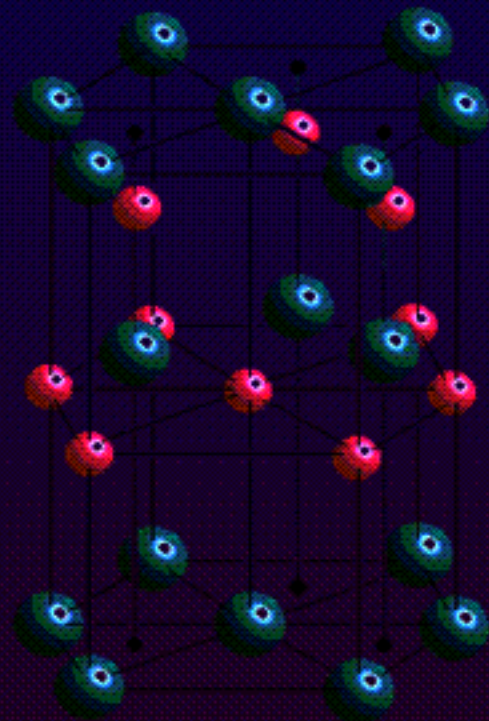


II	III	IV	V	VI
	5	6	7	8
	B	C	N	O
	13	14	15	16
	Al	Si	P	S
30	31	32	33	34
Zn	Ga	Ge	As	Se
48	49	50	51	52
Cd	In	Sn	Sb	Te
80	81	82	83	84
Hg	Tl	Pb	Bi	Po



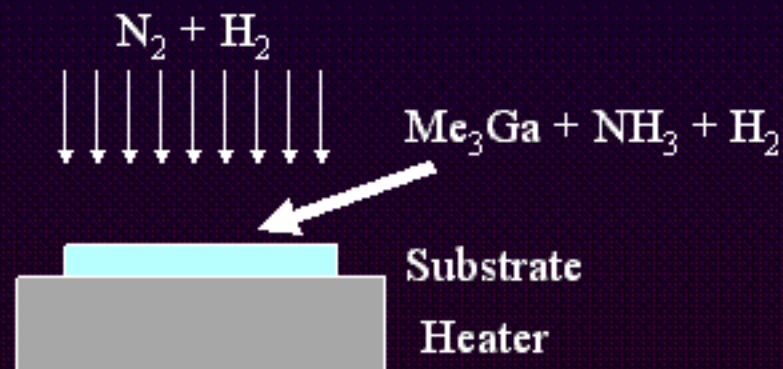
III-V Semiconductors

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hexagonal wurtzite

	GaN	AlN	InN	
Molecular weight:	83.72	40.98	128.82	
Density:	3.04	1.63	3.04	(gcm ⁻³)
Lattice Constant (a=)	3.19	3.11	3.54	(Å)
Lattice Constant (b=)	5.19	4.98	5.71	(Å)
Boiling point	1773, 2273	2673	1473	(°C)
Band-gap	3.4	6.2	1.9	(eV)
	366	200	653	(nm)

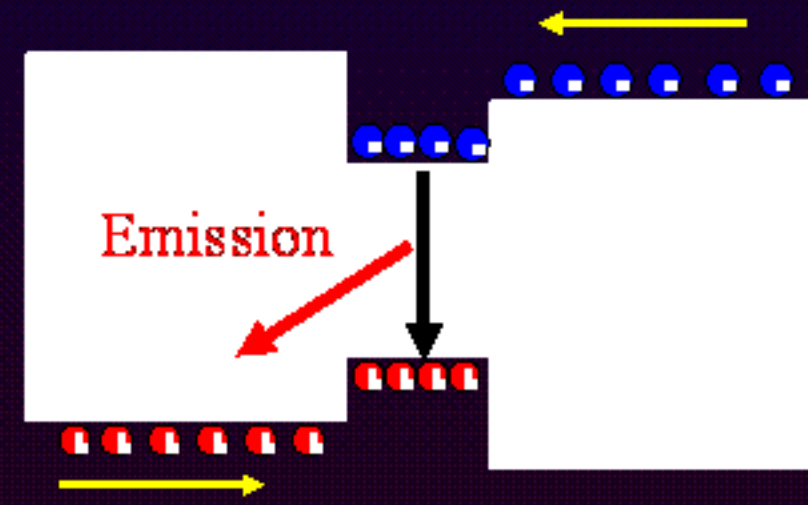
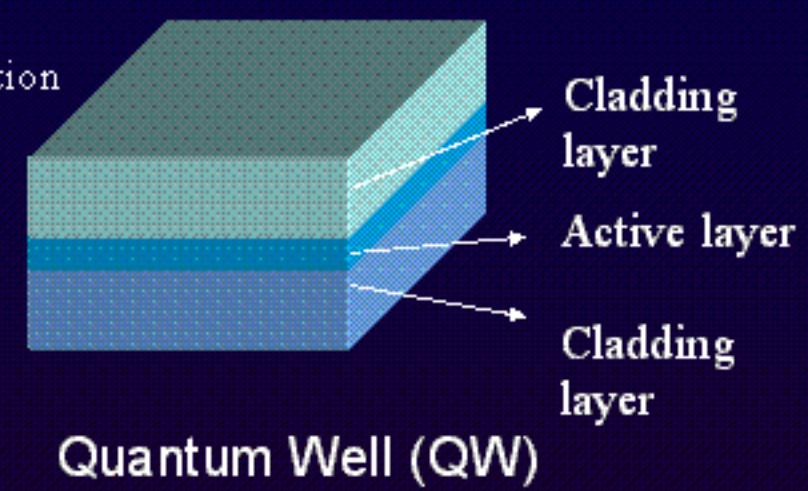
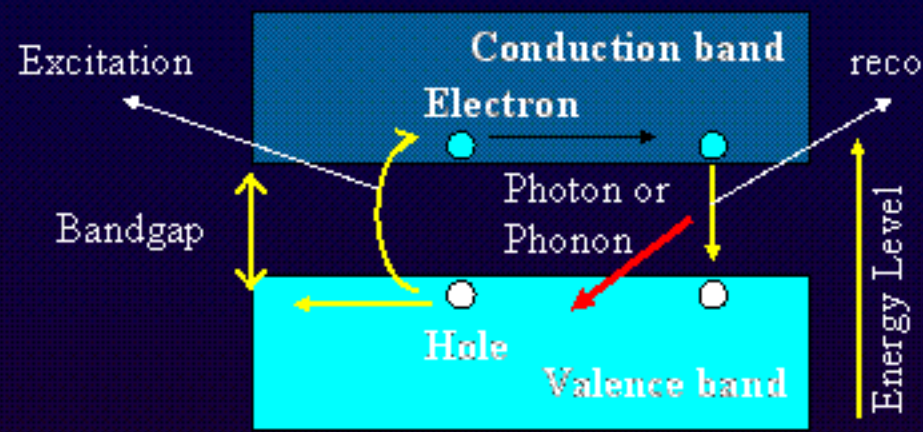


metalorganic chemical vapor deposition (MOCVD)



QW and LED

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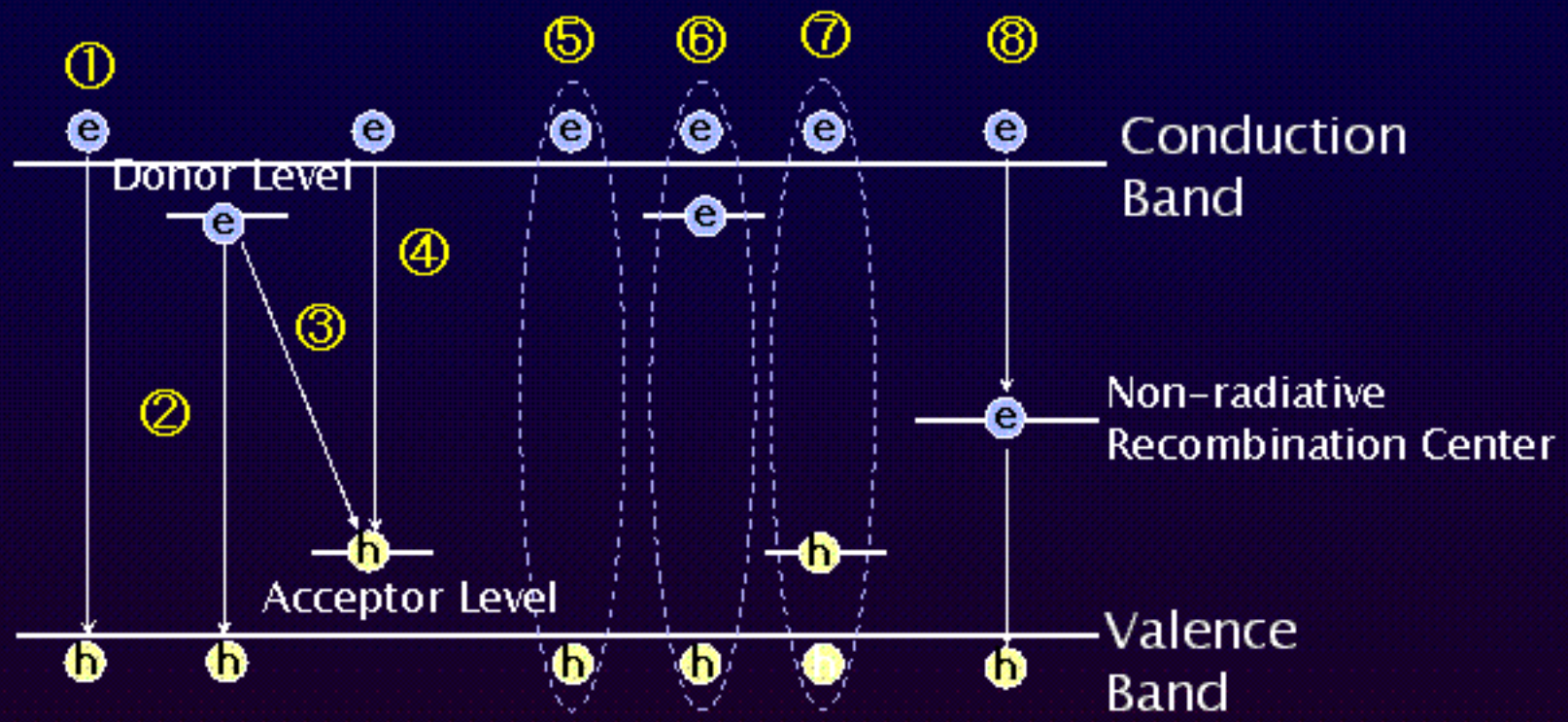


Light-emitting diode (LED)



Electron-Hole recombinations

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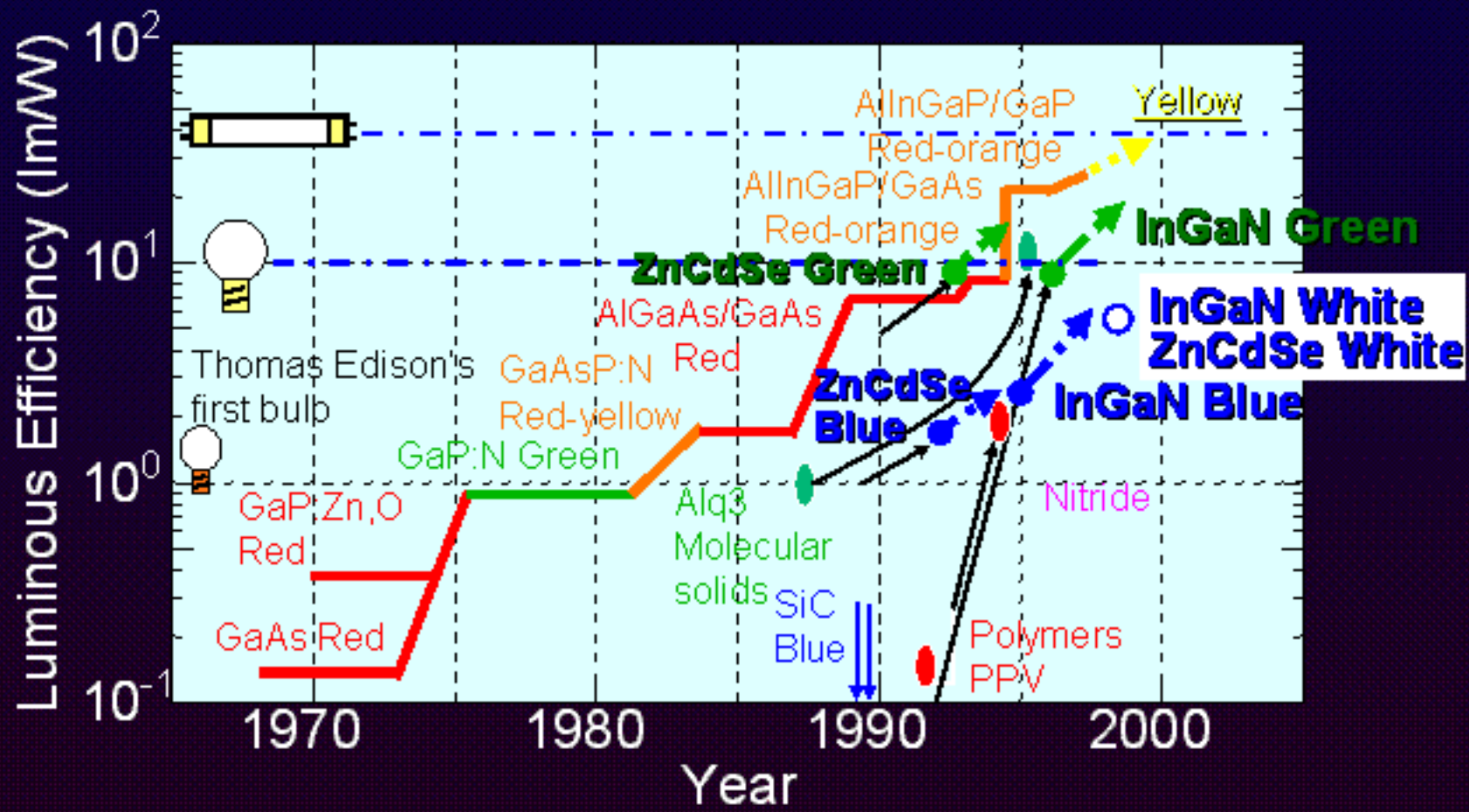
- | | |
|-------------------------------|-----------------------------------|
| ① Inter-band Emission | ⑤ Free Exciton Emission |
| ② BF (bound to free) Emission | ⑥ Donor bound Exciton Emission |
| ③ Donor-Acceptor Emission | ⑦ Acceptor bound Exciton Emission |
| ④ FB (free to bound) Emission | ⑧ Non-Radiative Recombination |

①~⑦ : Radiative (Emission) Processes ⑧ : Non-Radiative



Historical Development of LEDs

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InGaN/GaN-based LEDs

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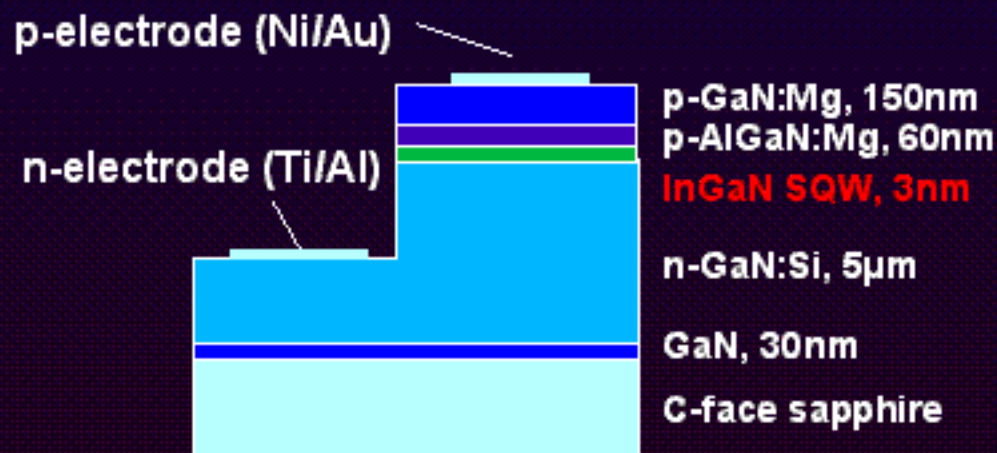
InGaN/GaN semiconductors

Very advantageous materials for light emitting diode (LED) and laser diode (LD).

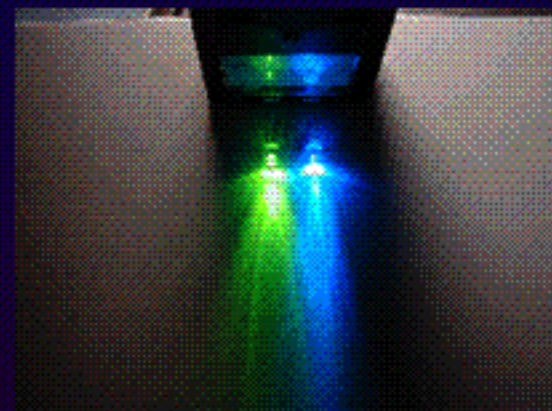
1993 GaN/InGaN based Blue-LED was developed by Shuji Nakamura in Nichia Co. (present; UCSB)

1995 GaN/InGaN based Blue-LD was also developed by Shuji Nakamura in Nichia Co.

Now: Cree, HP, Toyota gose, Omron, etc.



GaN/InGaN single quantum well structure of LEDs grown by two-flow metalorganic chemical vapor deposition (**MOCVD**)



Super bright LEDs and LD of InGaN/GaN/AlGaN by Nichia



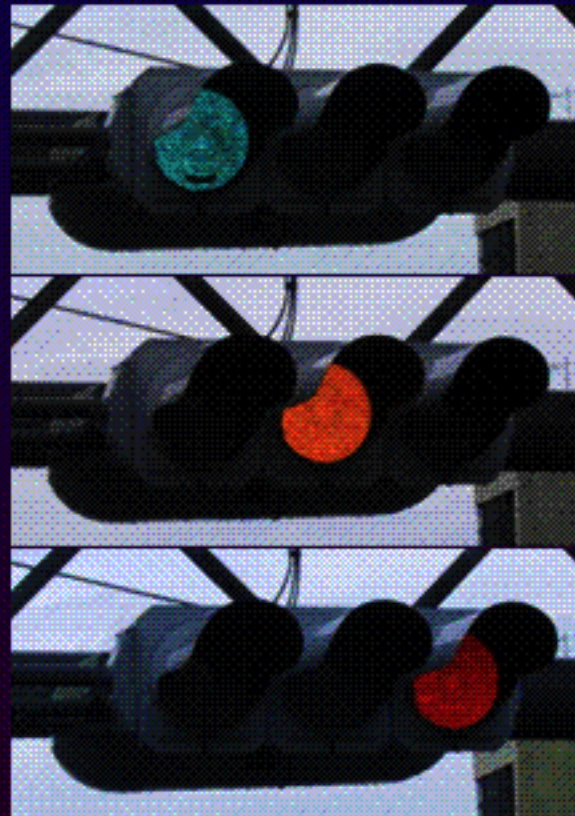
New Application of InGaN/GaN LED

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Primary color of light (RGB) were completed by LEDs



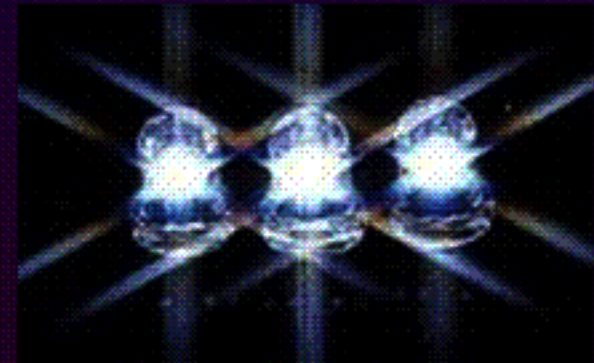
Full color LED display



LED Signal



Agriculture (Red and



White light LED
(blue LED and yellow fluorescent)



Application for Surgical Operation

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September 11, 2000 (Monday) in Kyoto, Japan



Operator Surgeon

Dr. J. Shimada (Kyoto Yosanoumi Hosp.)

Associate Surgeon

Dr. H. Amachi (Kyoto Yosanoumi Hosp.)

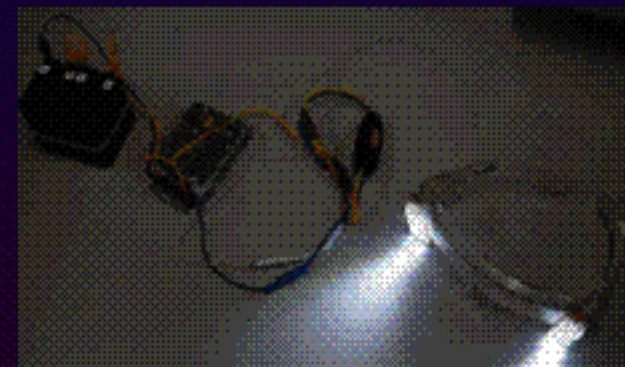
Nurse

Miss. Y. Shakamoto

Technical stuff

Dr. Y. Kawakami (Kyoto Univ.)

Dr. K. Okamoto (Kyoto Univ.)



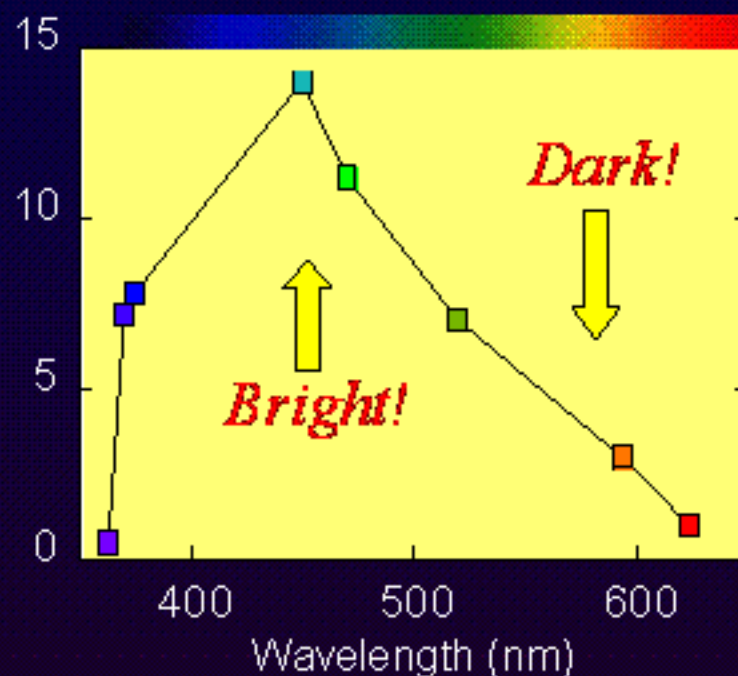
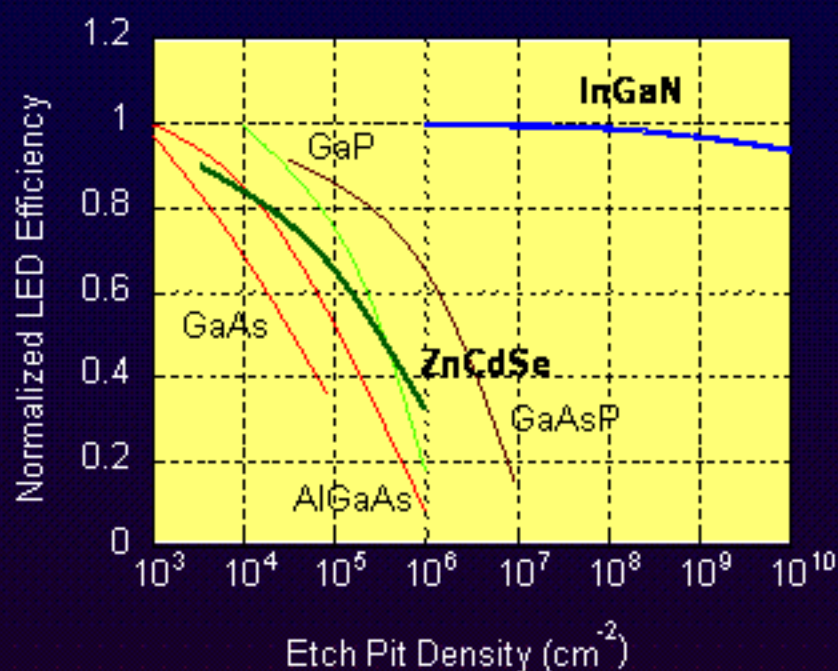
This head-mounted light source uses **112** white LEDs to illuminated surgical operation.

Surgeons and scientists in Kyoto are collaborating on a head-mounted prototype white LED lighting system designed to provide directed illumination during surgical operations. The LEDs were mounted on simple lightweight goggles and made headlines in Japan recently following their use in a successful 2-hour forearm operation.



Optical Properties of InGaN/GaN

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In spite of high threading dislocation density (10^8 - 10^{10} cm^{-2}), the external quantum efficiency (η_{ext}) of emission are very high (10-15%) (20 % is now achieved in Lab.)

However, η_{ext} values are still lower for LEDs out of this blue spectral range.

Our Purpose

To elucidate the emission dynamics and optical properties

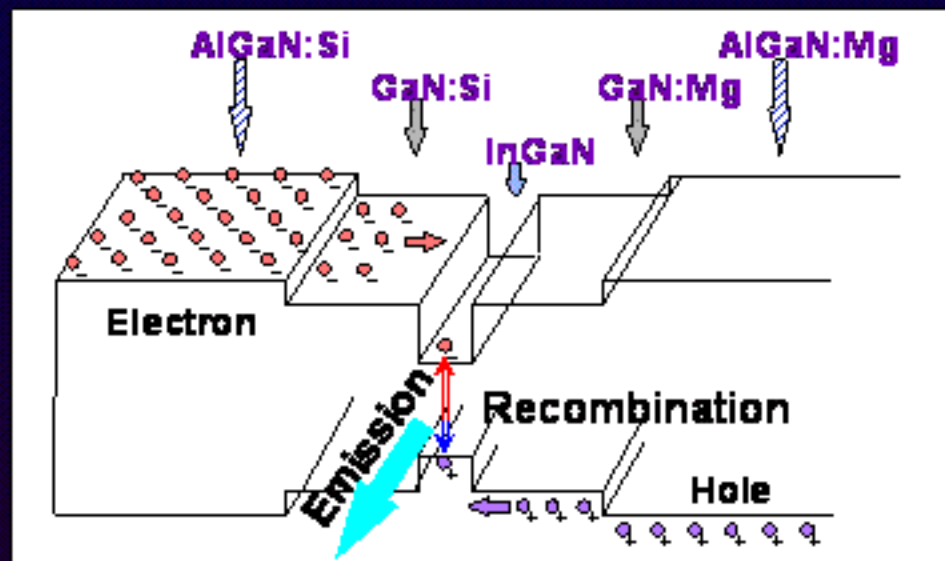
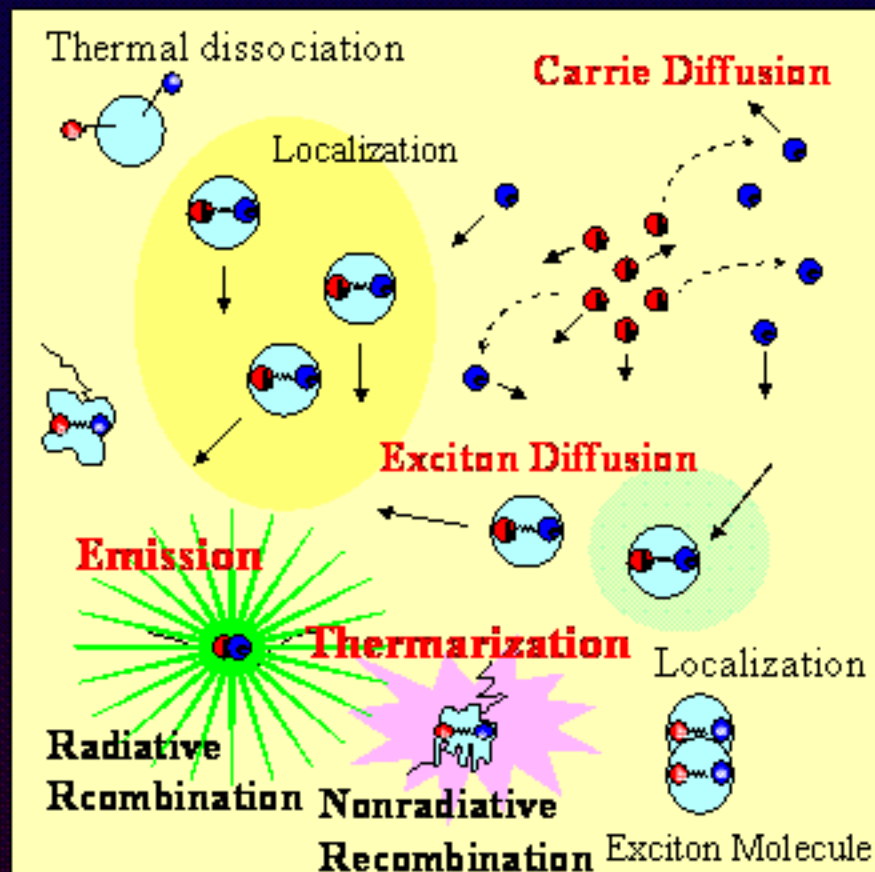
To obtain the way how to develop the emission efficiency



Carriers and Excitons Dynamics

© Koichi Okamoto

Optical properties are controlled by the dynamics of carriers and/or excitons



We try to detect

(1) Time and spatial resolved PL

by time-resolved macro-PL spectroscopy

(2) Nano-scale inhomogeneously of PL

by SNOM(NSOM) with I-C mode

(3) Carrier diffusion and thermarization

by third order nonlinear spectroscopy

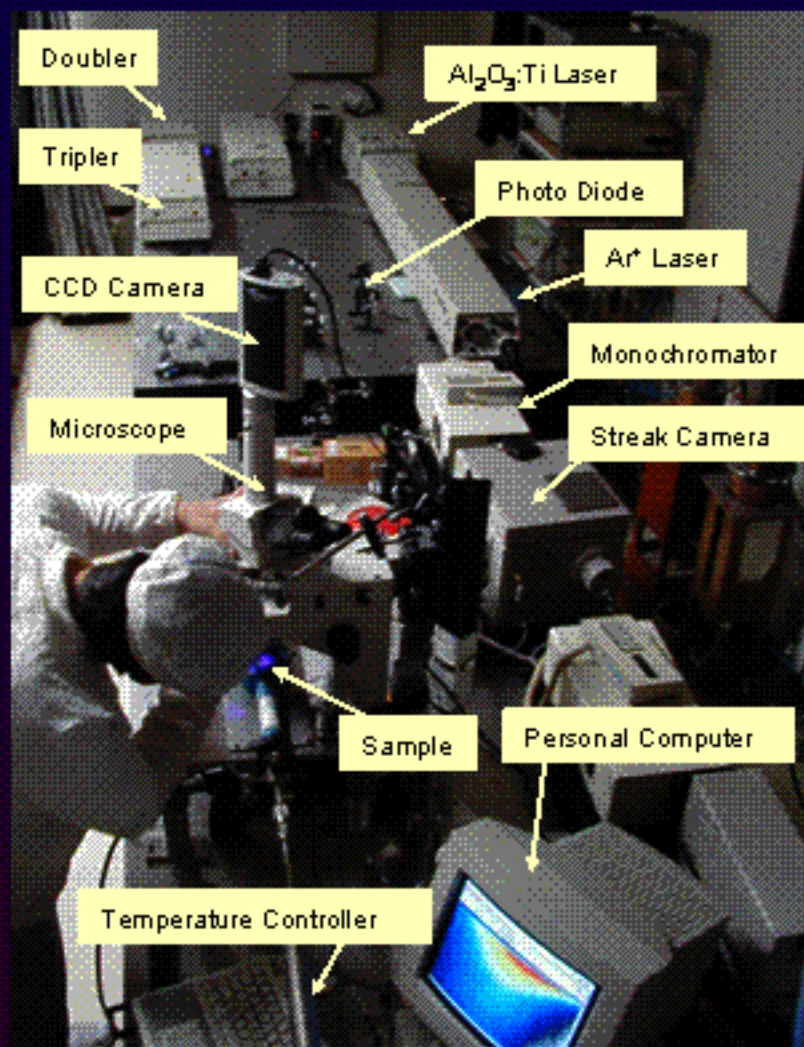
Until now, Photoluminescence (PL) spectroscopy, time resolved PL, micro-PL, Electroluminescence (EL), cathodoluminescence (CL), etc.



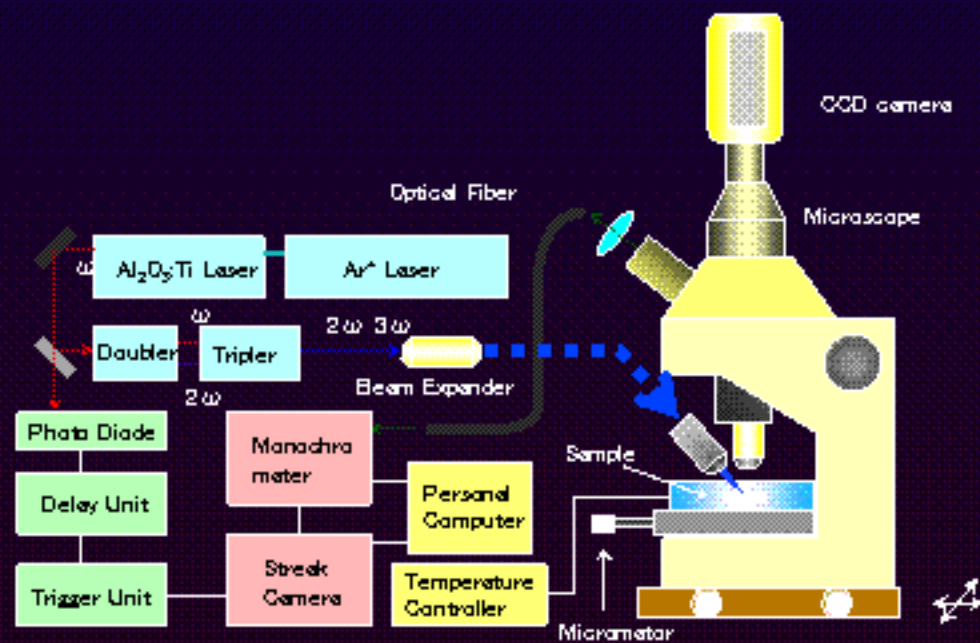
Time Resolved Micro PL

© Koichi Okamoto

K. Okamoto, et al, J. Crystal Growth, 214/215, 639 (2000).



Mode locked Al₂O₃:Ti laser
 (pumped by Ar⁺ laser)
 pulse width: 1.5 ps
 wavelength: 400 nm
 repetition rate: 80MHz
 power density: 20μJ/cm²

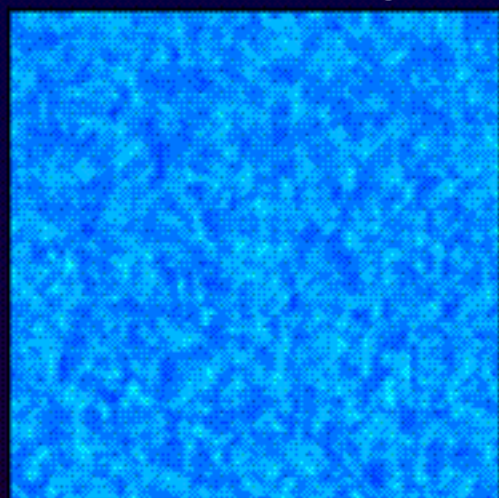




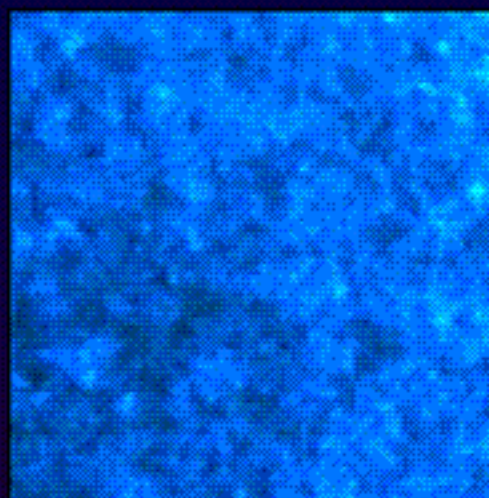
Microscopic Images

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@R.T.

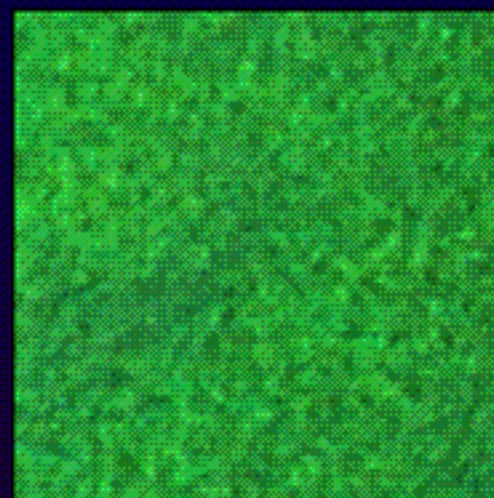
20 μm



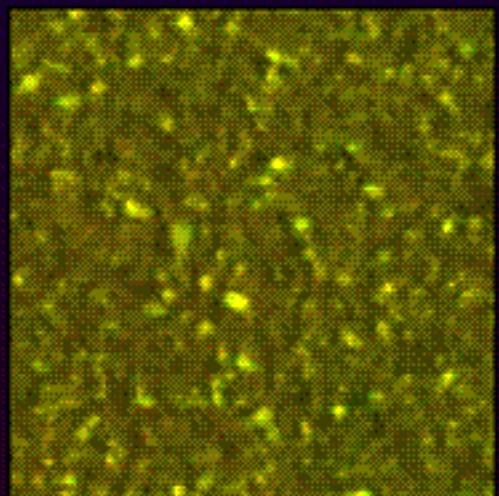
LED 460



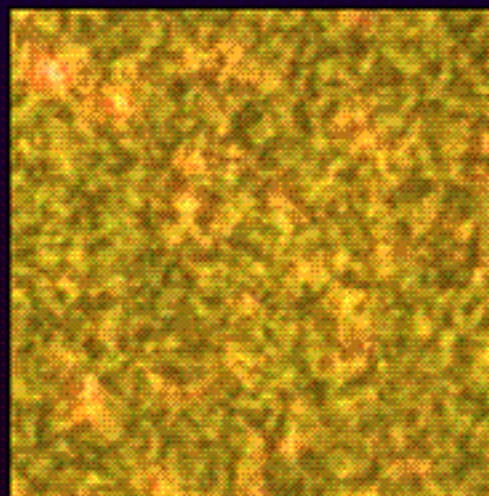
LED 470



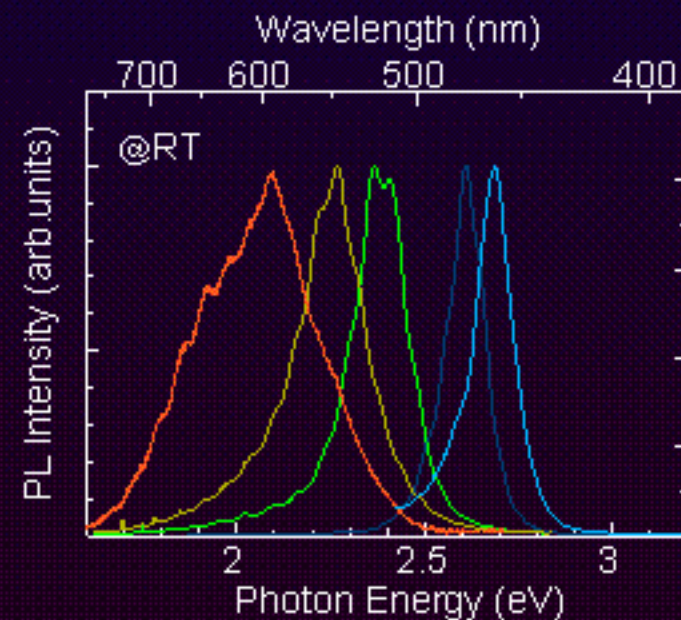
LED 510



LED 540



LED 600

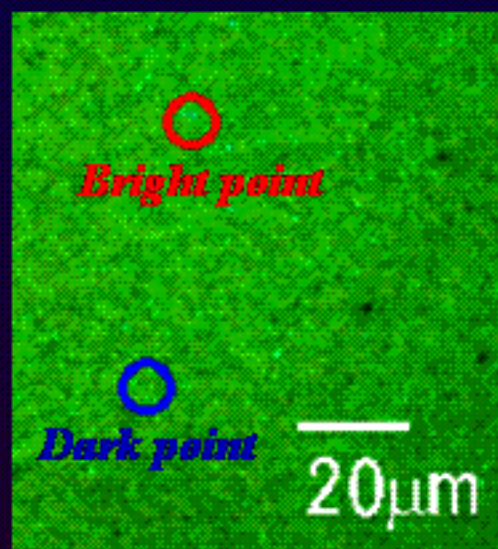




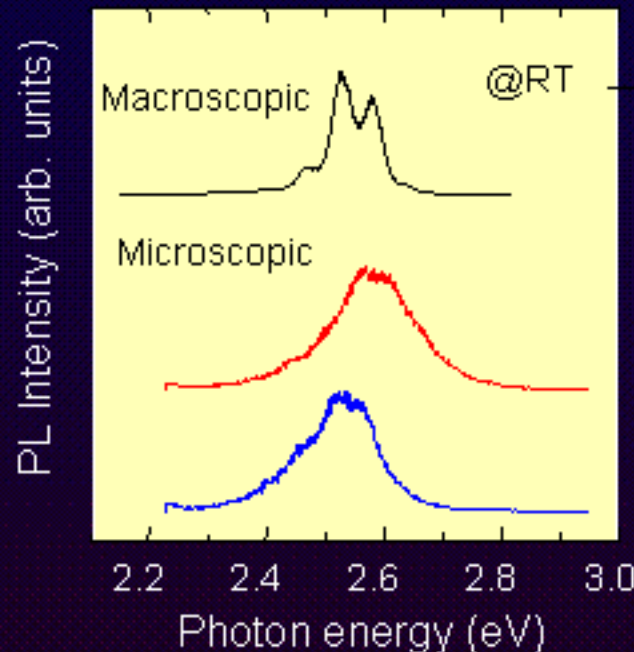
Results of the TRMPL

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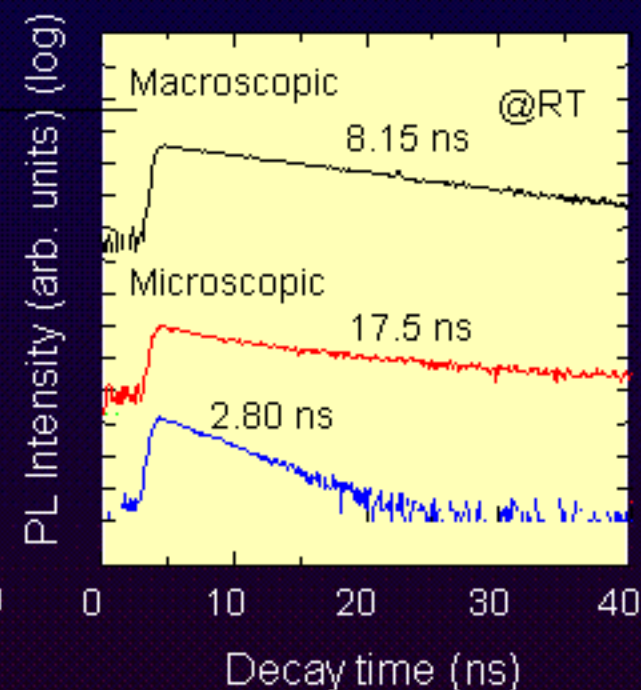
Spot Size ($2\mu\text{m}\times 2\mu\text{m}$)



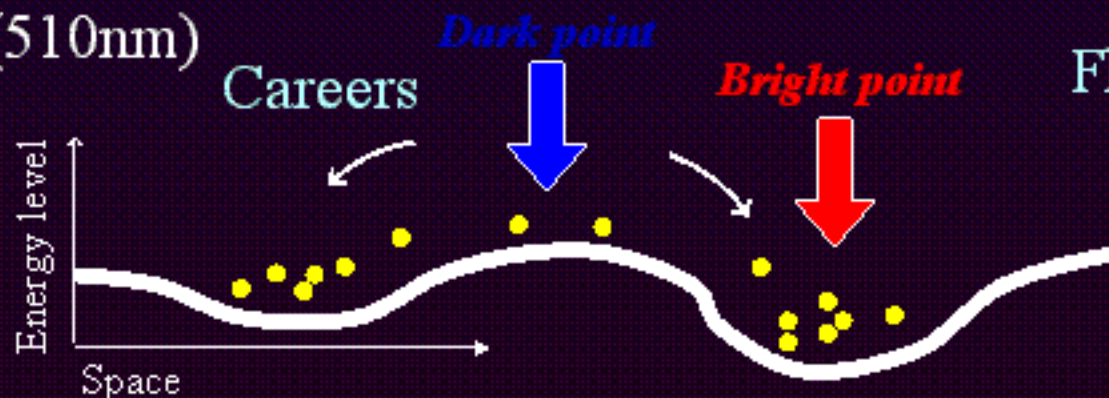
Spectrum



Time-profile



Green LED
(510nm)

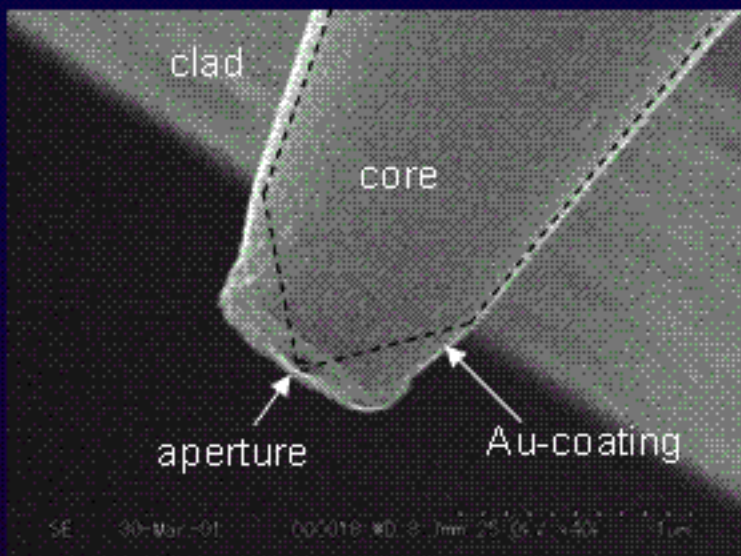


Fluctuation of Energy Level

In Composition
quantum well width
Piezoelectric field



Near-field Scanning Optical Microscopy (NSOM) © Koichi Okamoto



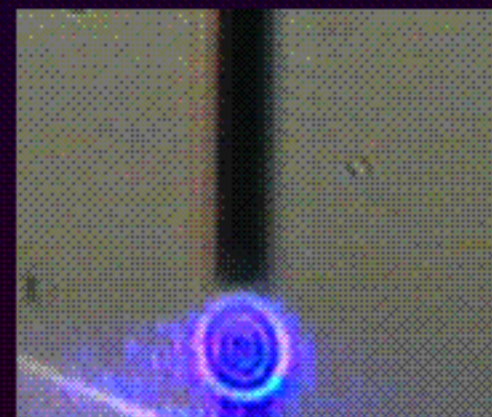
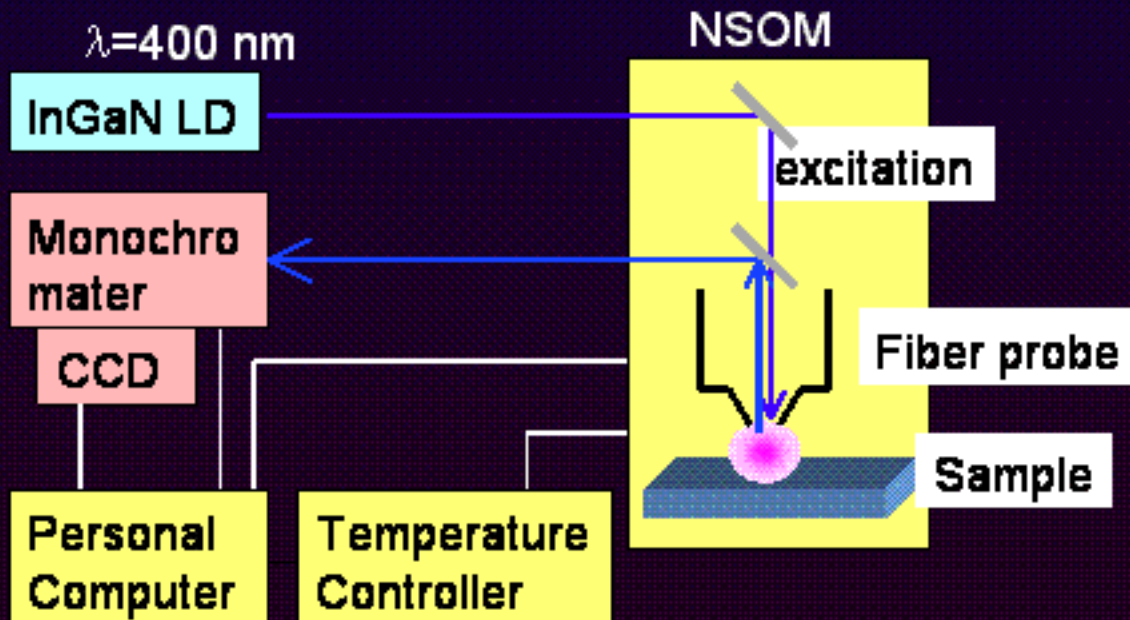
(NSOM)

Optical fiber tip

Evanescent light

Metal coat

Aperture size: ~ 150 nm



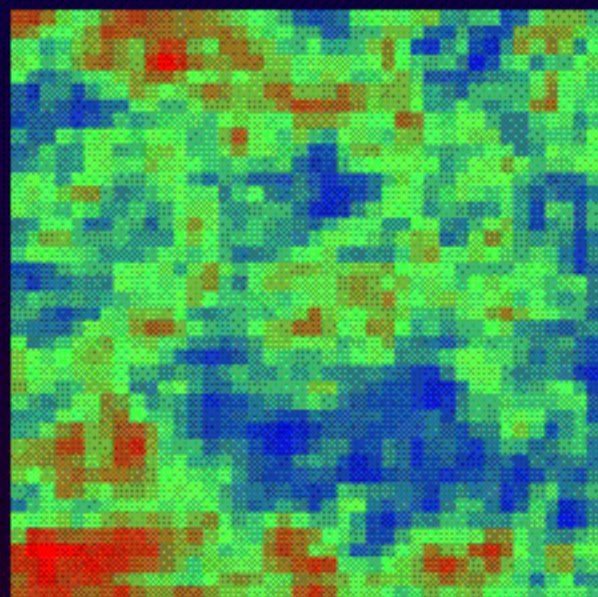


SNOM Images of LEDs

© Koichi Okamoto

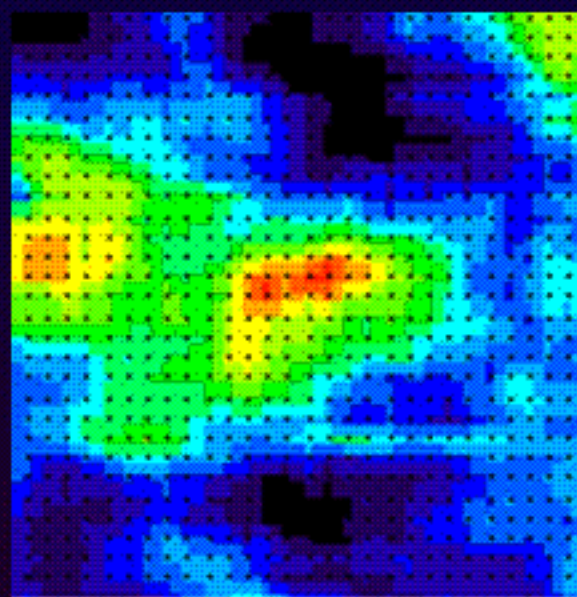
Bright!

Blue LED (420nm)

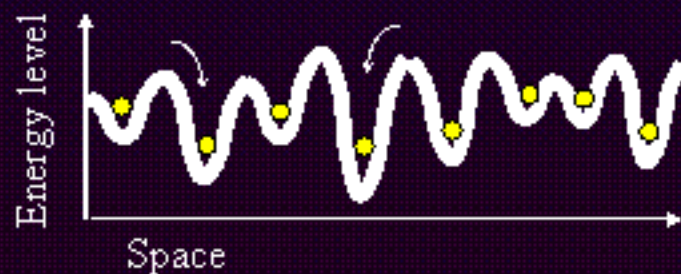


Dark!

Yellow LED (540nm)



PL integrated Intensity (arb. units)

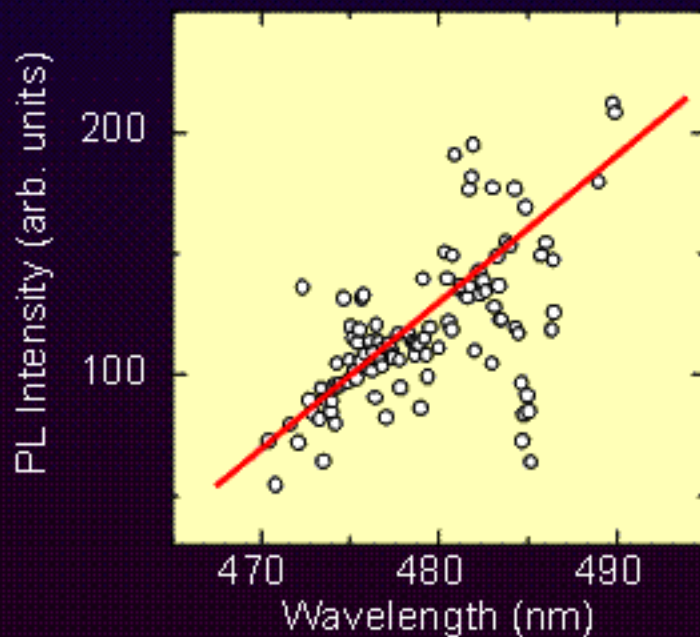
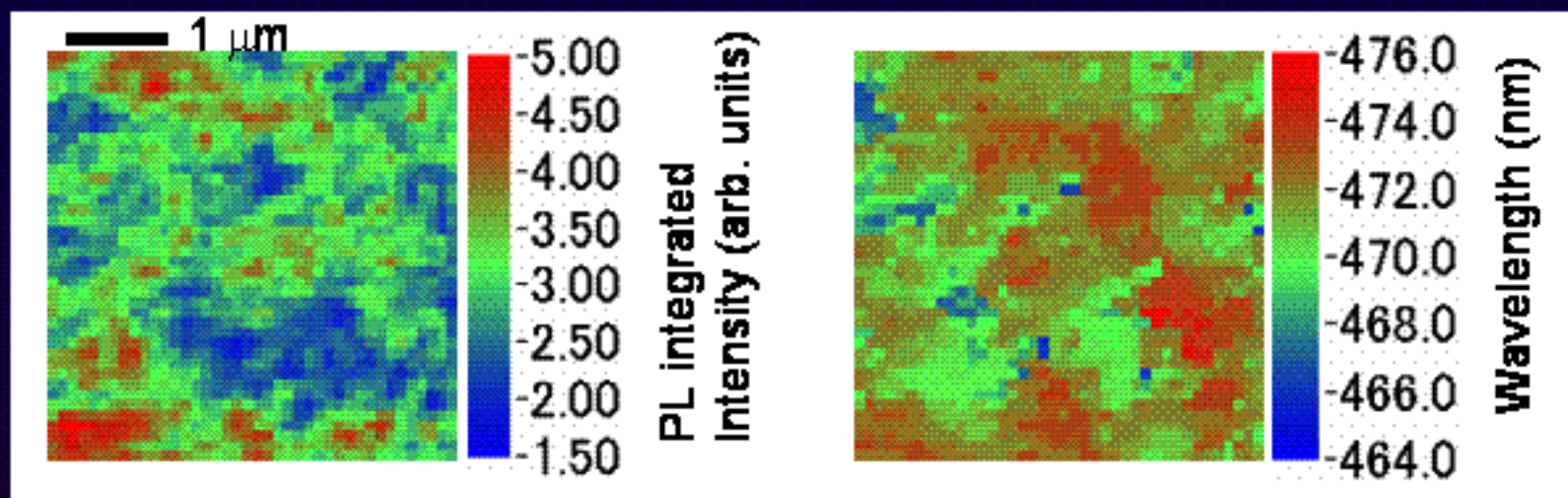




Correlation between

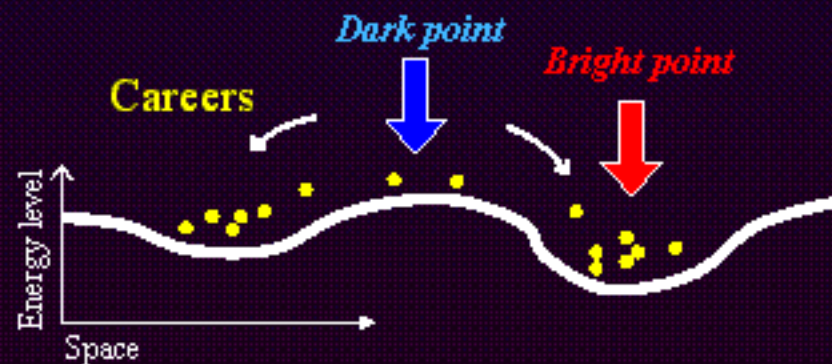
PL intensity and peak wavelength

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Strong PL region =

PL peak wavelength is longer

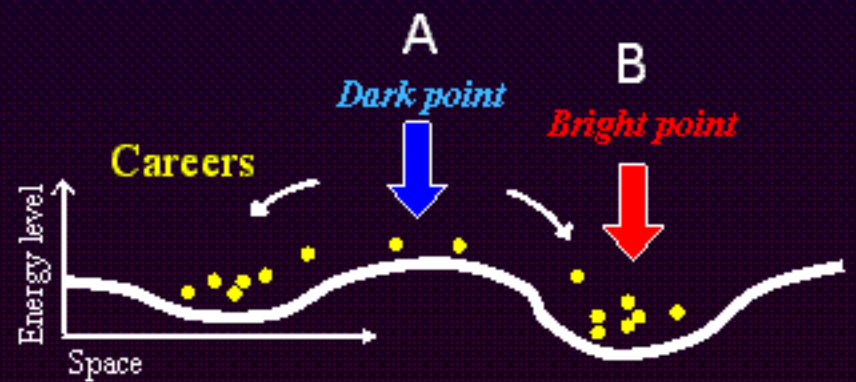
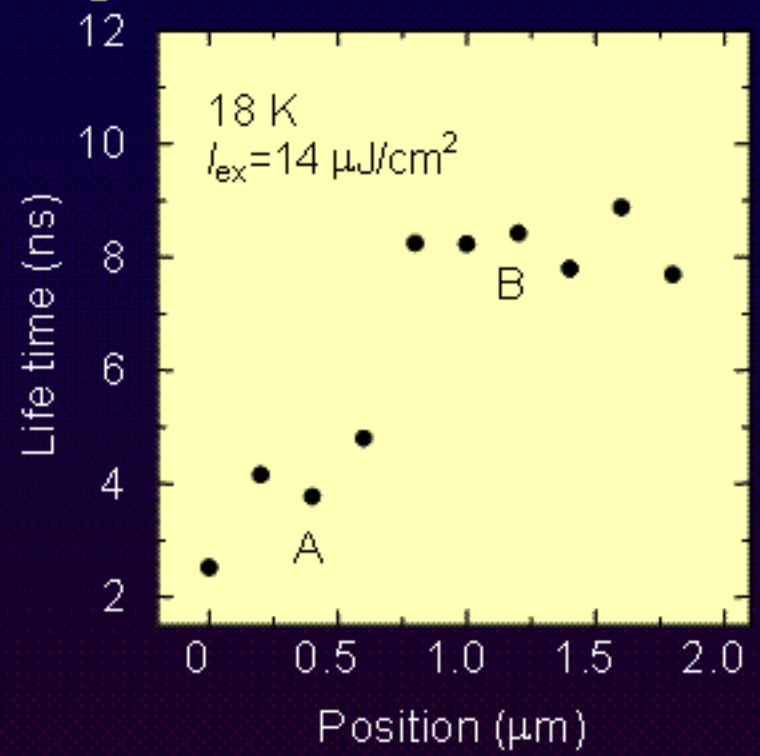
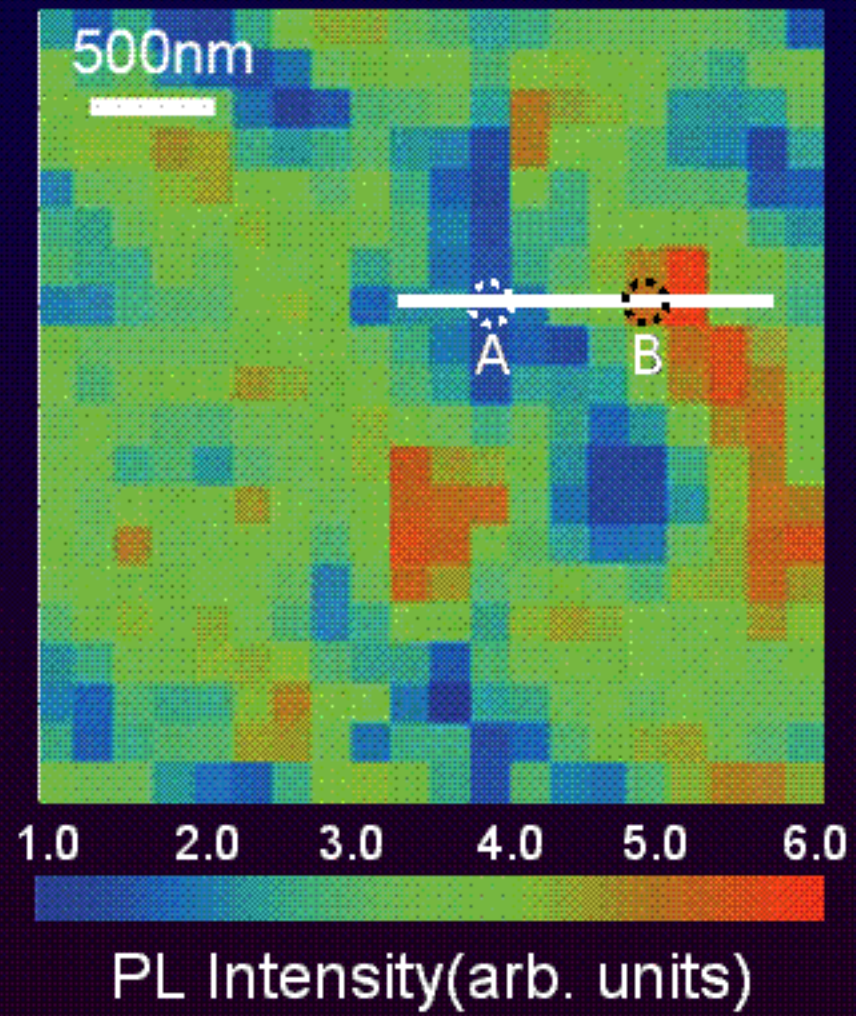




NSOM PL image and PL lifetimes along the white bar

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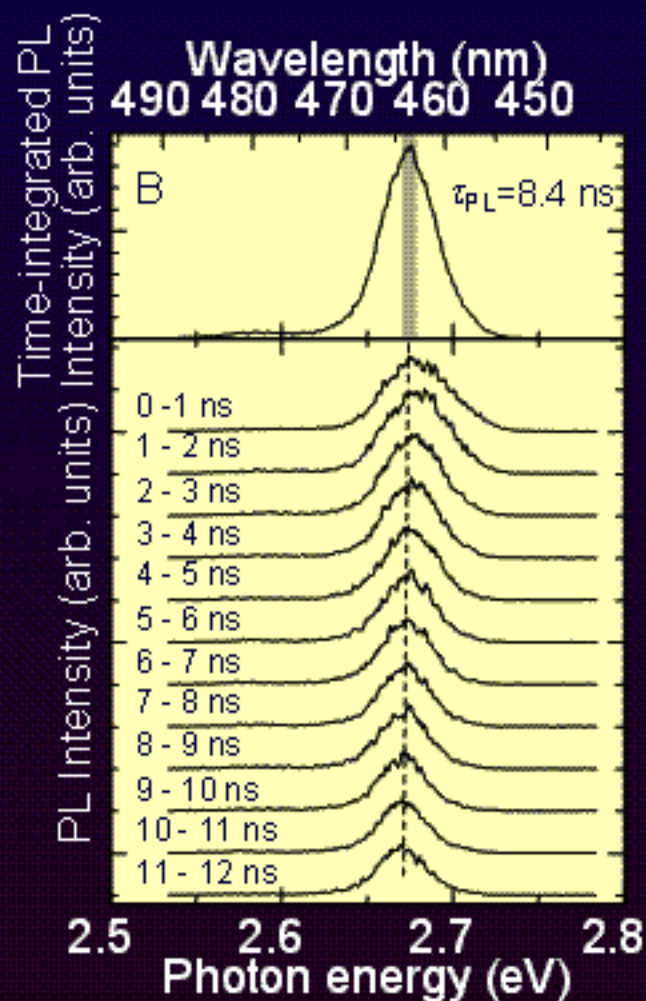
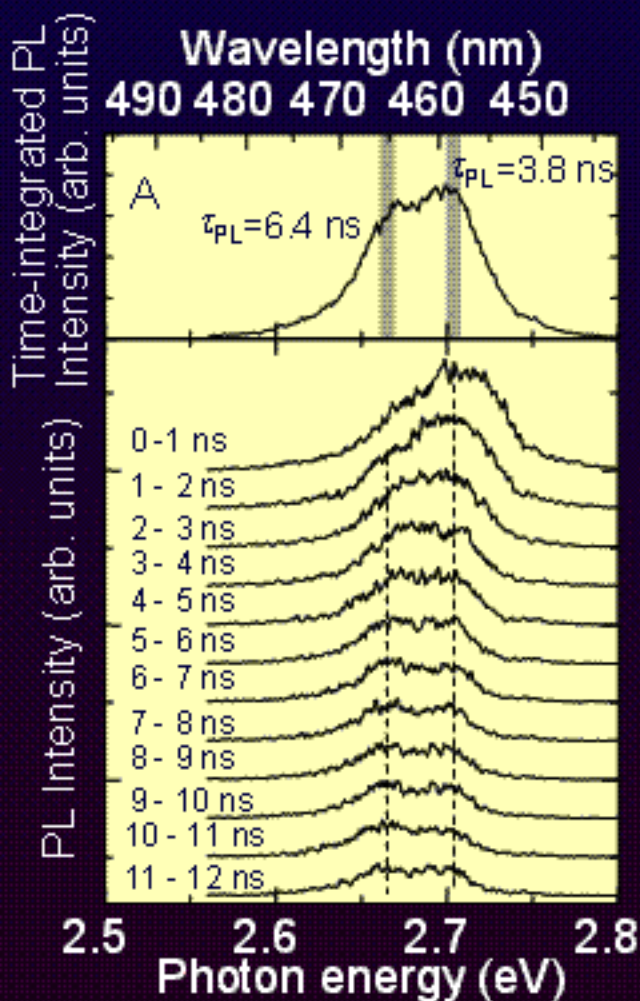
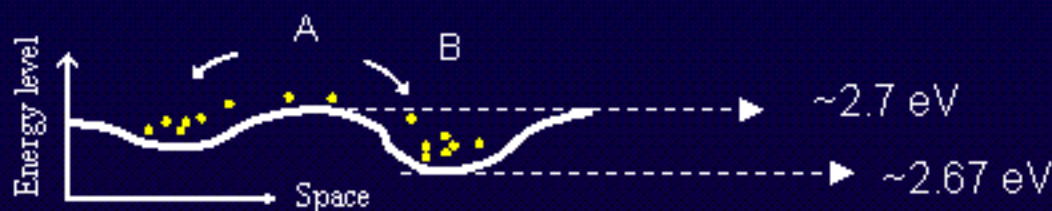
along the white bar





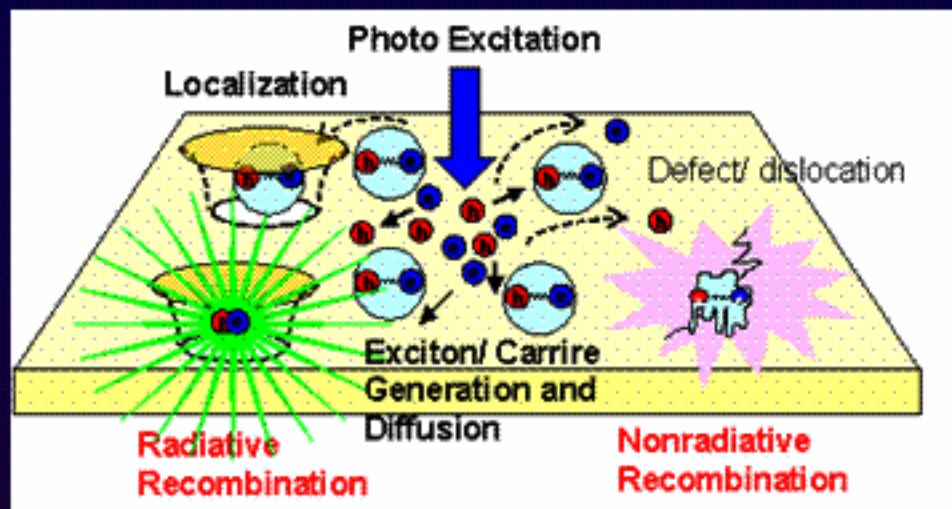
TR-PL spectra at A and B

© Koichi Okamoto





Observation of nonradiative processes © Koichi Okamoto



Radiative Processes

Well studied by
Photoluminescence (PL), Electricluminescence (EL), or Cathordoluminescence (CL)

Non-radiative Processes

Carrie dynamics, Therlnal dynamics
Also Important but very few studied

3rd order nonlinear optics

$$P / \epsilon_0 = \sum \chi_{ij}^{(1)} E_j + \sum \sum \chi_{ijk}^{(2)} E_j E_k + \sum \sum \sum \chi_{ijkl}^{(3)} E_j E_k E_l + \dots$$



Electronic polarization	Thermal dynamics	Clustering, Aggregation
Electron transfer	Volume, structure change	Molecular harmonic effect
Energy transfer	Density change	Nana particle growth
Carrier Dynamics	Ultrasonic, Acoustic wave	Crystal growth
Excitation Dynamics	Chemical reaction	Phase Transfer
Molecular vibration	Molecular translation	Metal diffusion

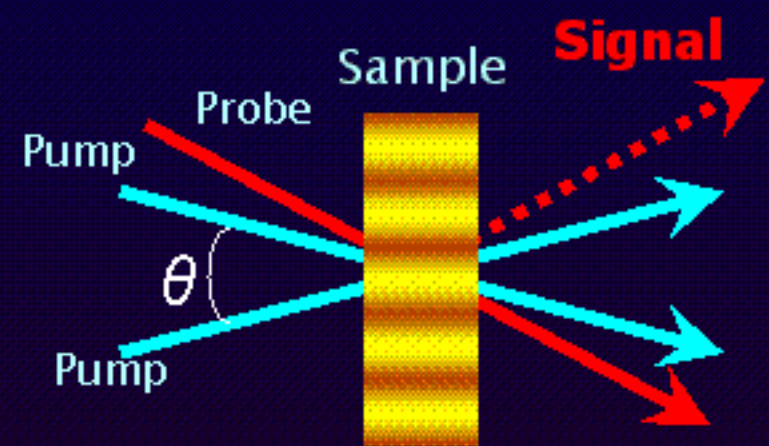


3rd order nonlinear spectroscopy

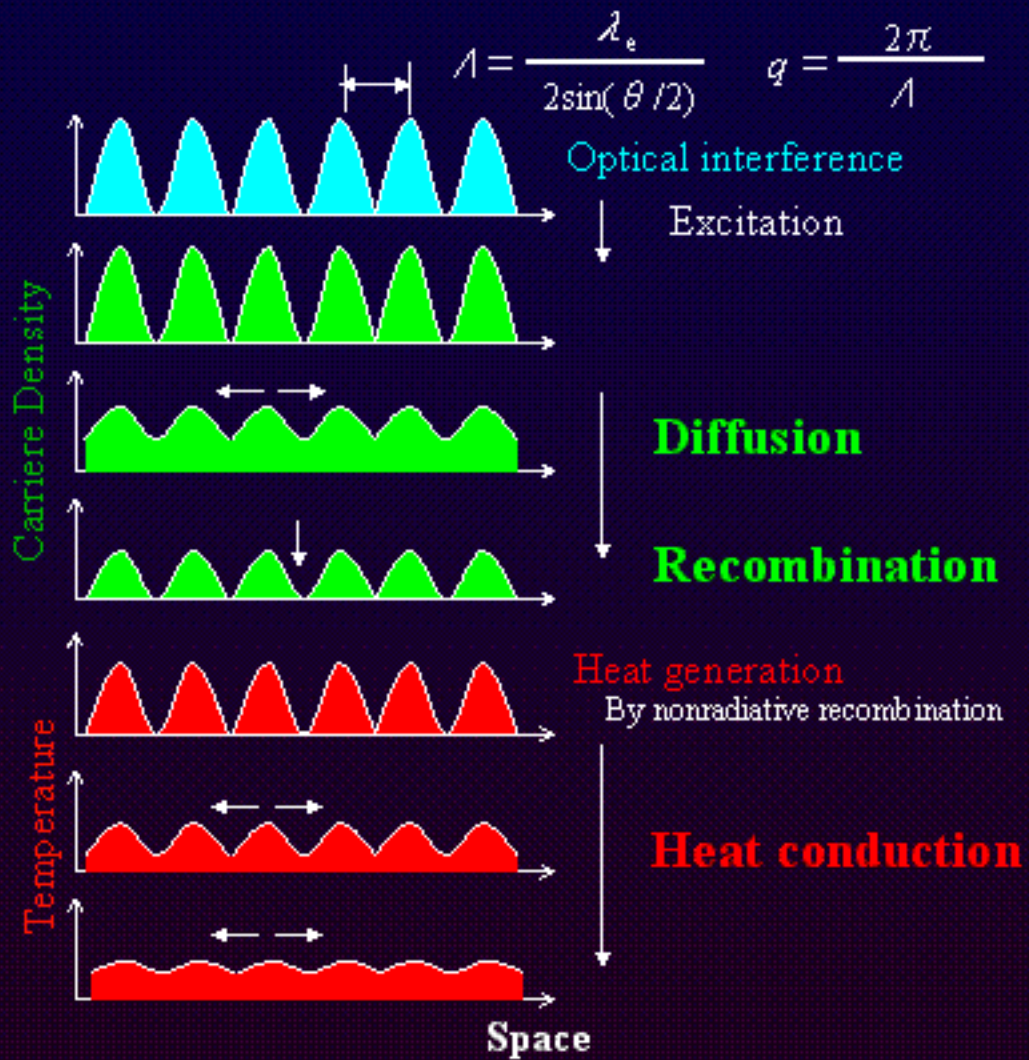
© Koichi Okamoto

K. Okamoto, et al, Analytical Science, 17, s312 (2001).

Transient Grating (TG)



Pump by the interference pattern created by crossing two beams
 Create the grating (moderation of the carrier and/or exciton densities or temperature)
Probe by the diffracted beam



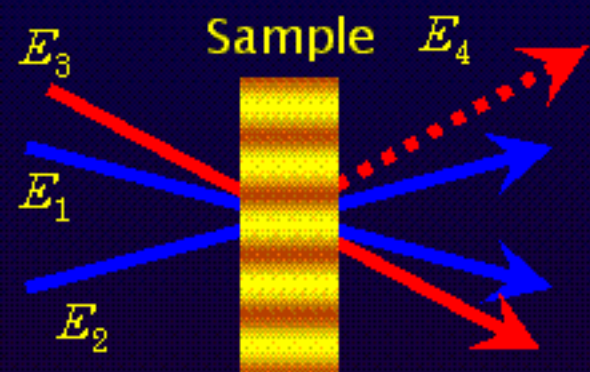
High sensitivities, high accuracy, high time resolution, and high spatial resolution



Application of TG method

© Koichi Okamoto

$$P | \varepsilon_0 = \sum \chi_{ij}^{(1)} E_j + \sum \sum \chi_{ijk}^{(2)} E_j E_k + \sum \sum \sum \chi_{ijkl}^{(3)} E_j E_k E_l + \dots$$



							Time
femto	pico	micro	mili	second	mint	hour	→
Electronic polarization		Thermal dynamics			Clustering, Aggregation		
Electron transfer		Volume, structure change			Molecular harmonic effect		
Energy transfer		Density change			Nana particle growth		
Carrier Dynamics		Ultrasonic, Acoustic wave			Crystal growth		
Excitation Dynamics		Chemical reaction			Phase Transfer		
Molecular vibration		Molecular translation			Metal diffusion		

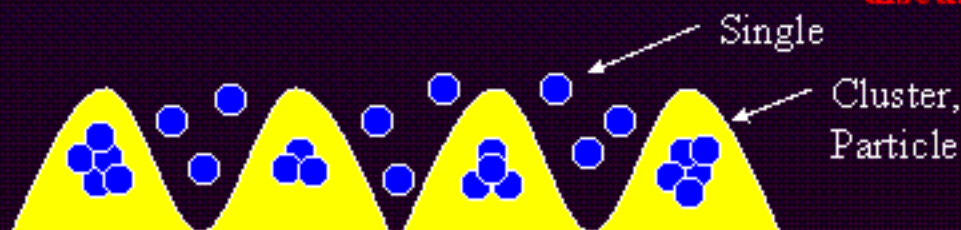
(For example) observation of molecular dynamics in solution



First time observation of diffusion of excited molecules

Both diffusions are quite different!!!

Caution!! Photochemical reaction should not be discusses based on only the hydrodynamic theory!!!



Real-time observation of molecular aggregation, ion clustering, metal particle growth

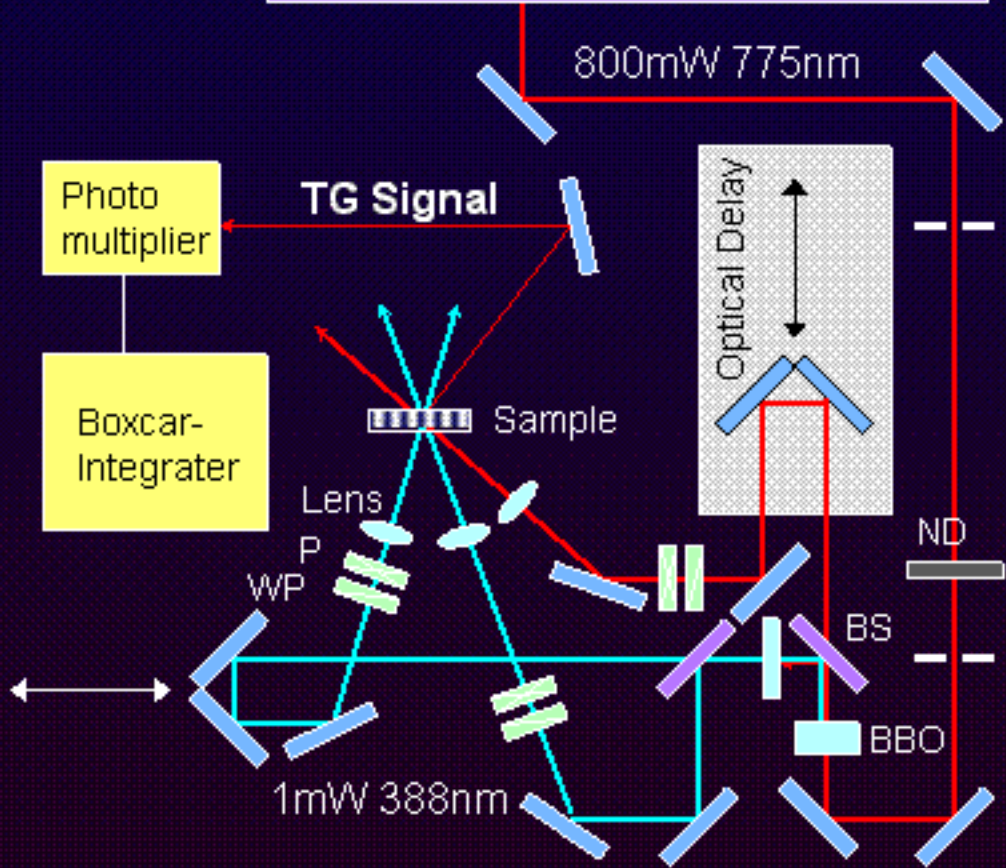


Experimental Setup of the TG method

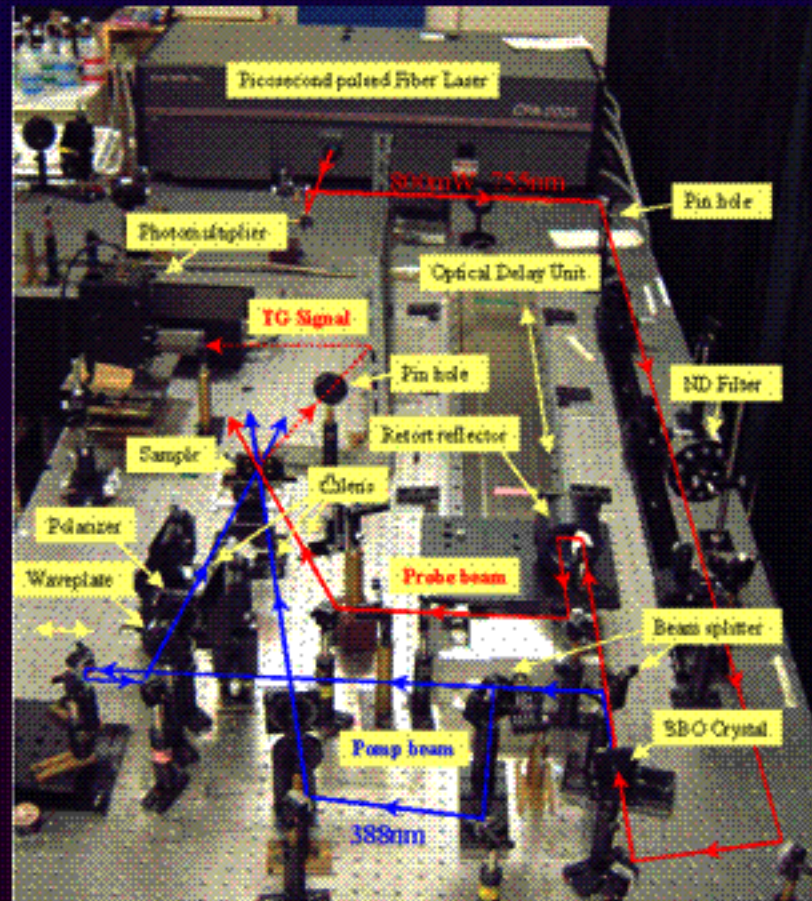
© Koichi Okamoto

**Mode-Locked Fiber Laser
Regenerate Amplifier**

Mode locked Fiber laser
frequency doubled beam
pulse width: 500 fs
wavelength: 388 nm
repetition rate: 1 kHz
power density: $0.5 \mu\text{J}/\text{cm}^2$



Picosecond time-scale

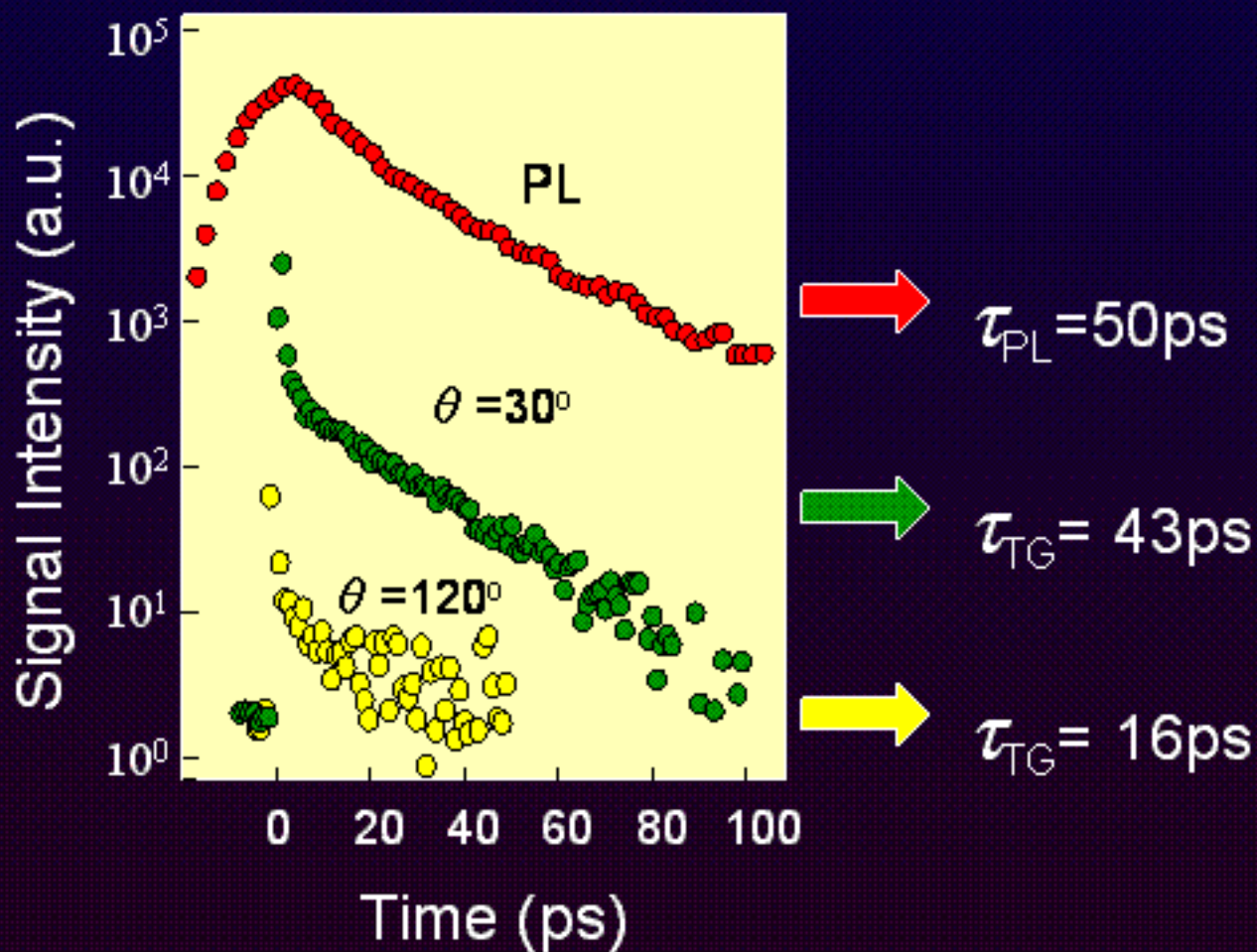




Time-profile of the TG signals of GaN

© Koichi Okamoto

GaN@R.T.





Results and Analysis of the TG signal

© Koichi Okamoto

The TG signal intensity is given by the refractive index change

$$I_{TG} / I_0 = \alpha \Delta n^2 + \beta \Delta k^2 \quad \text{In this time, } I_{TG}^{1st} \propto \Delta n$$

$\delta n(x, t)$ depend on the dynamics of carrier and/or exciton

$$\delta n(x, t) = \left(\frac{\partial n}{\partial N} \right) \delta N(x, t) + \left(\frac{\partial n}{\partial T} \right) \delta T(x, t) \quad \left(\frac{\partial n}{\partial N} \right) < 0, \quad \left(\frac{\partial n}{\partial T} \right) > 0$$

The carrier dynamics can be written by

$$\frac{d\delta N(x, t)}{dt} = D \frac{\partial^2 \delta N(x, t)}{\partial x^2} - \left(\frac{1}{\tau_{rad}} + \frac{1}{\tau_{non-rad}} \right) \delta N(x, t)$$

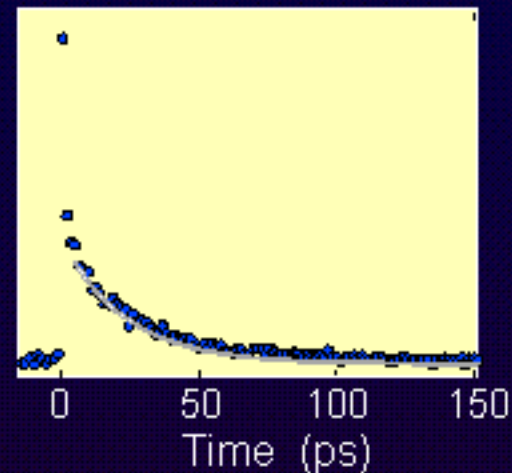
$$\frac{d\delta T(x, t)}{dt} = \frac{1}{\tau_{on-rad}} \frac{Q \delta N(x, t)}{\rho C_p} + D_{th} \frac{\partial^2 \delta T(x, t)}{\partial x^2}$$

By solving, the time profile of the TG signal was given by

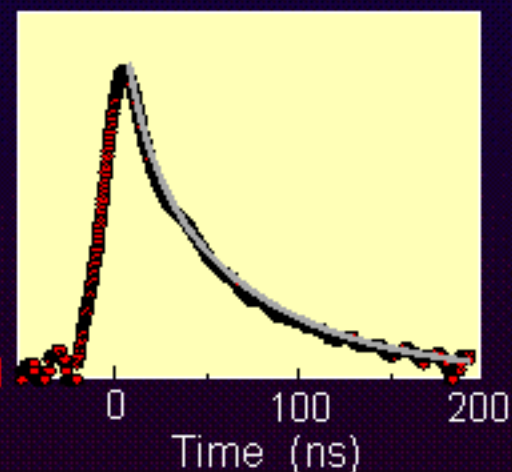
$$\delta \hat{n}(q, t) = \left(\frac{\partial n}{\partial N} \right) \delta \hat{N}(q, 0) \exp \left[- \left(Dq^2 + \frac{1}{\tau_{rad}} + \frac{1}{\tau_{non-rad}} \right) t \right]$$

$$+ \left(\frac{\partial n}{\partial T} \right) \frac{\tau_{rad}}{\tau_{rad} + \tau_{non-rad} + \tau_{rad} \tau_{non-rad} Dq^2} \frac{Q \delta \hat{N}(q, 0)}{\rho C_p} \left\{ - \exp \left[- \left(Dq^2 + \frac{1}{\tau_{rad}} + \frac{1}{\tau_{non-rad}} \right) t \right] + \exp \left(- D_{th} q^2 t \right) \right\}$$

Intensity (a.u.)



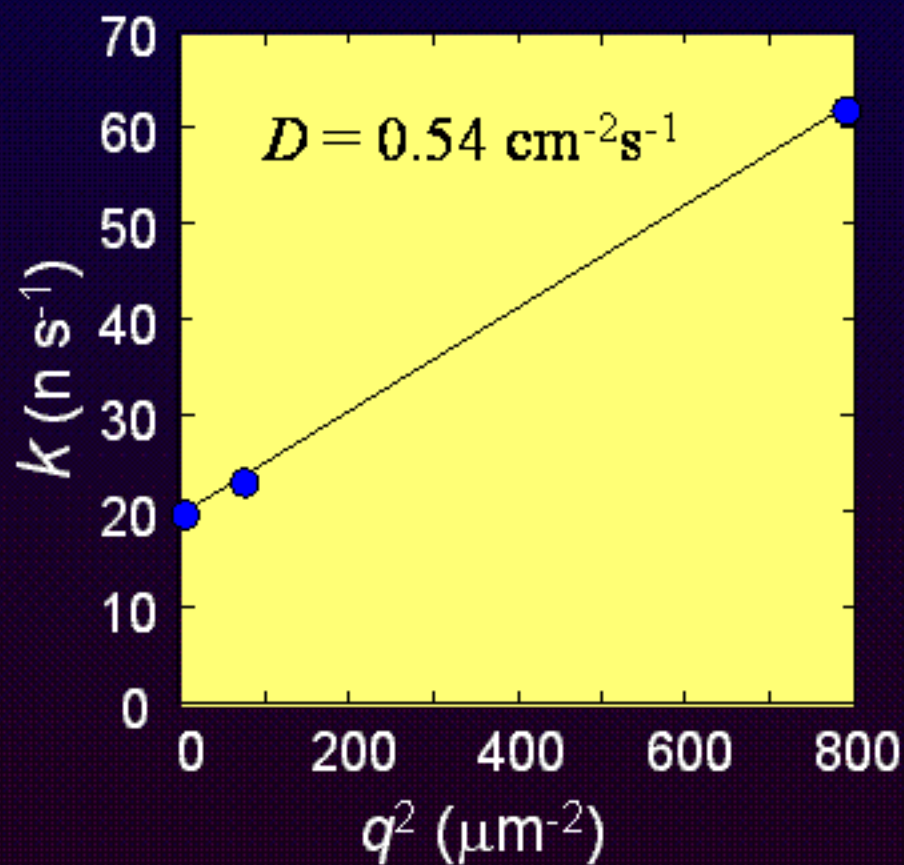
Intensity (a.u.)



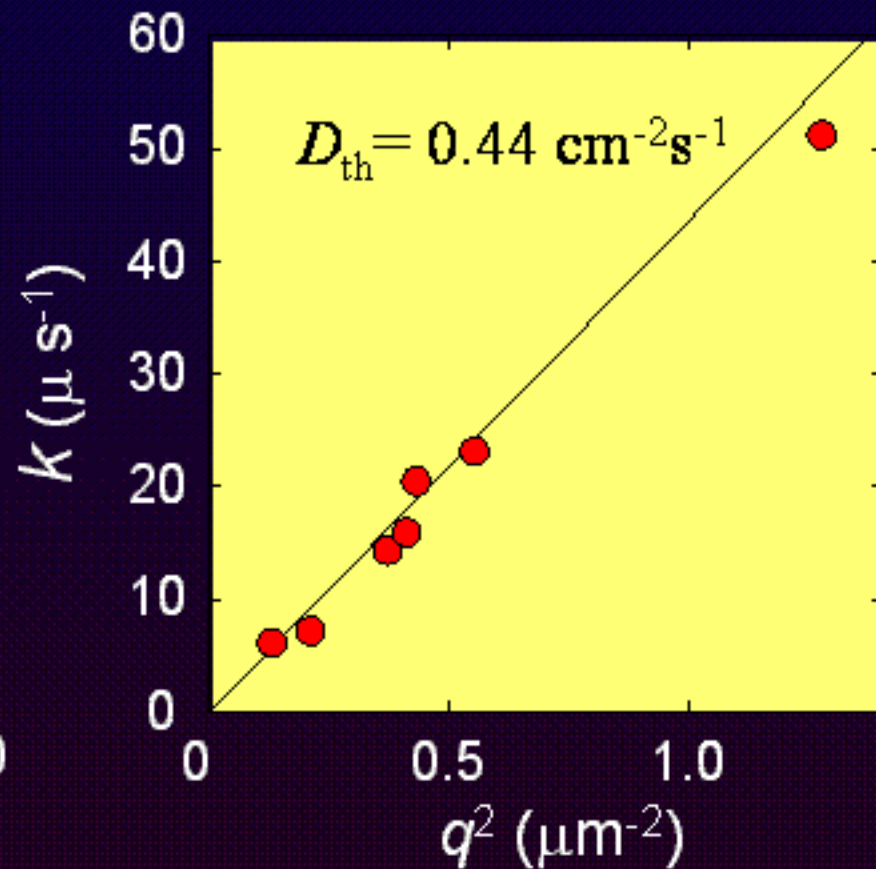


GaN@R.T.

Carrier density grating



Thermal grating

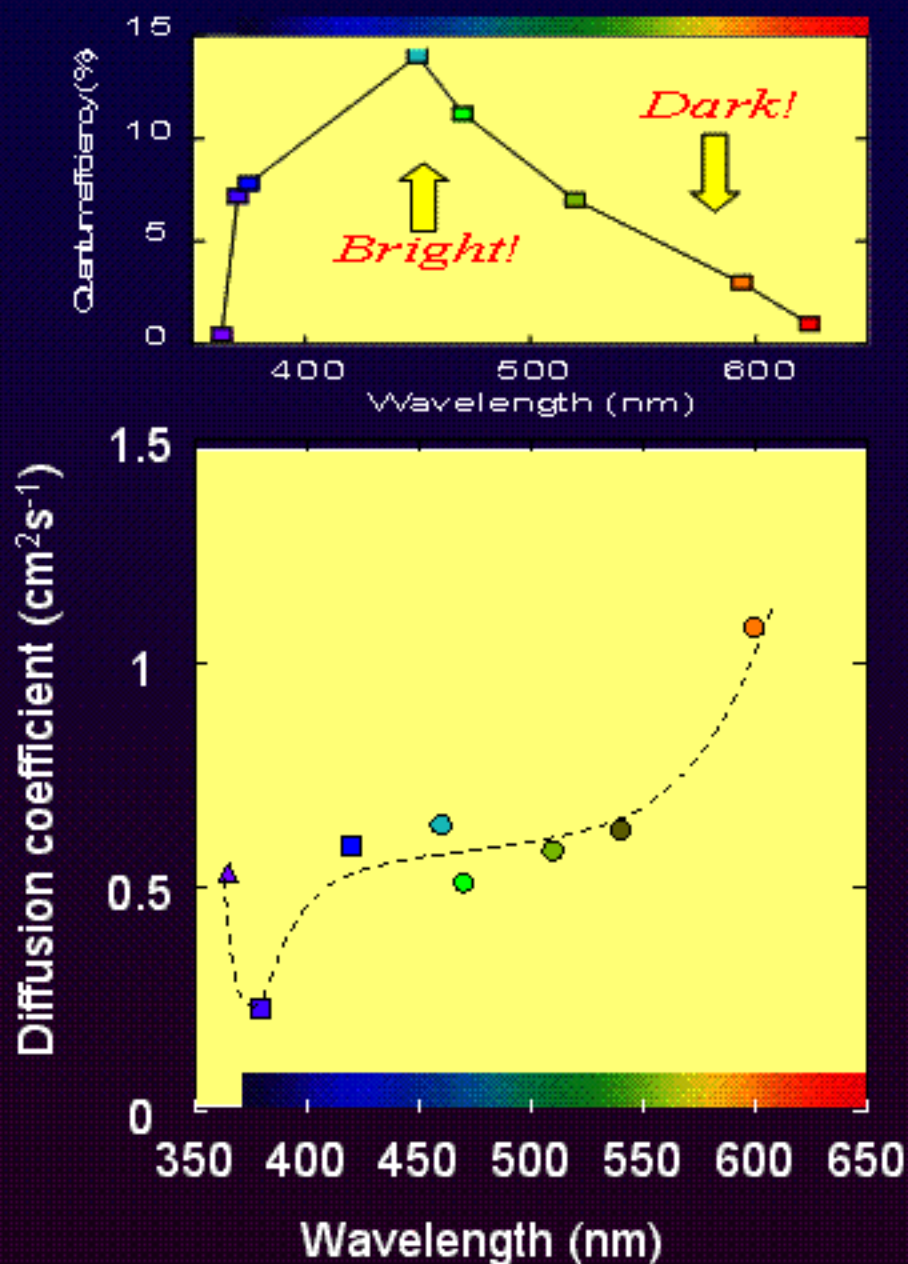
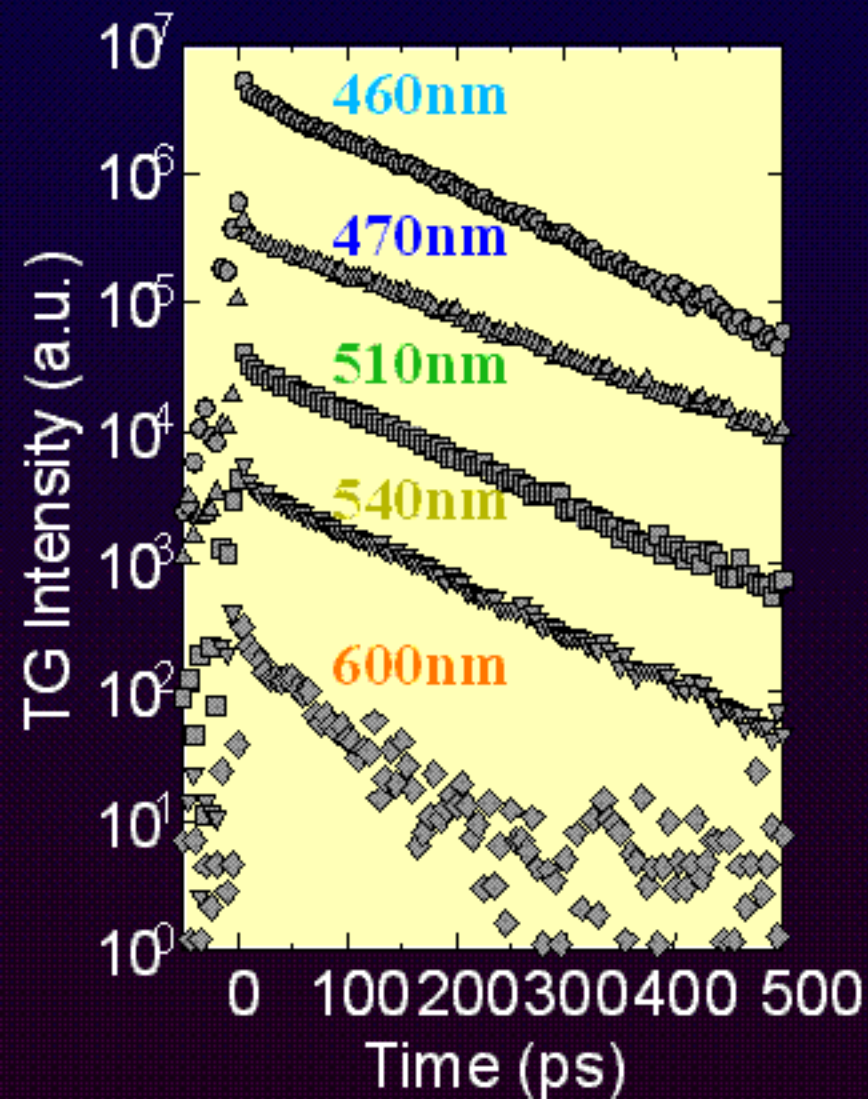




TG measurement for InGaN LEDs

© Koichi Okamoto

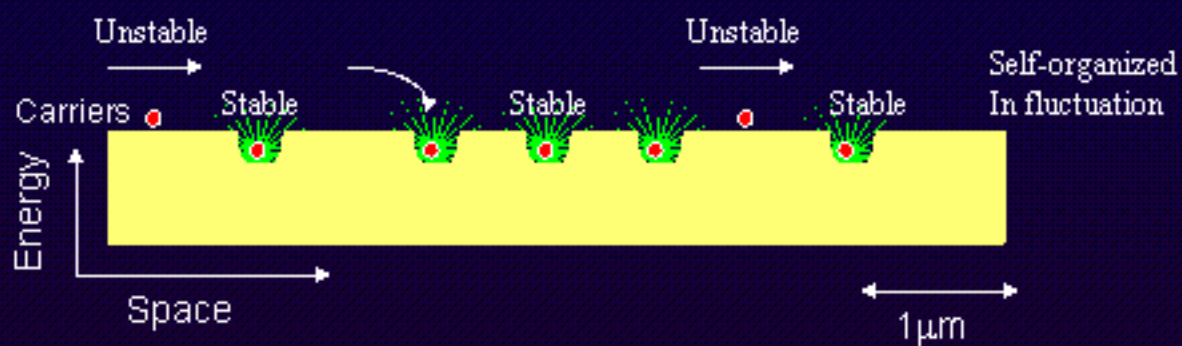
K. Okamoto, et al
Physica Status Solidi B, 228, 81 (2001)





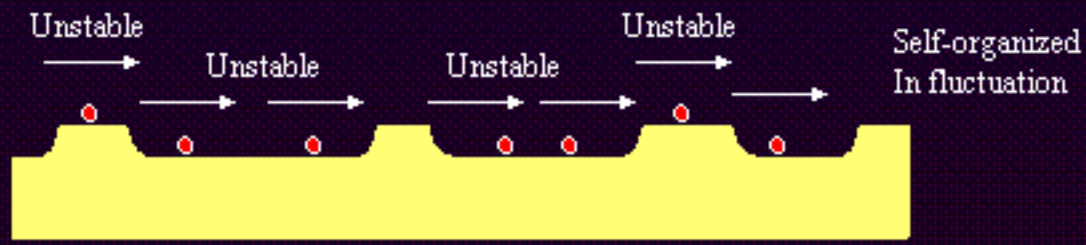
Carrier Diffusion and Localization

© Koichi Okamoto



Bright!

LED with blue-green
(low In doping)



Dark!

LED with yellow-red
(high In doping)

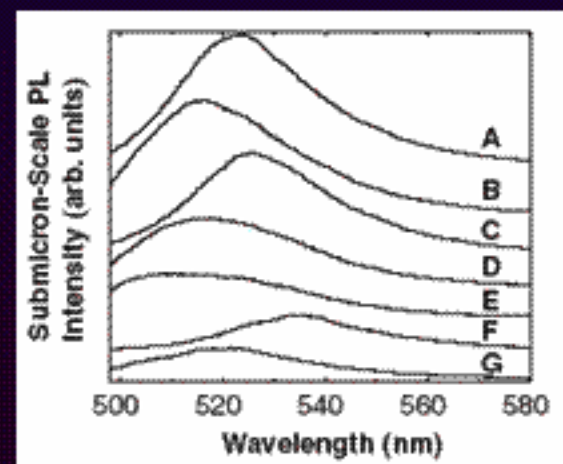
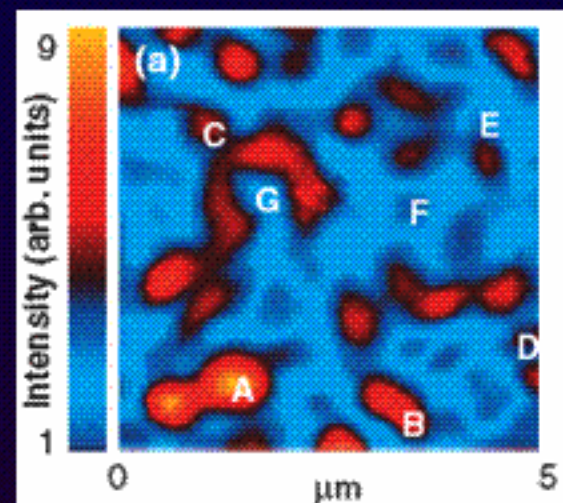
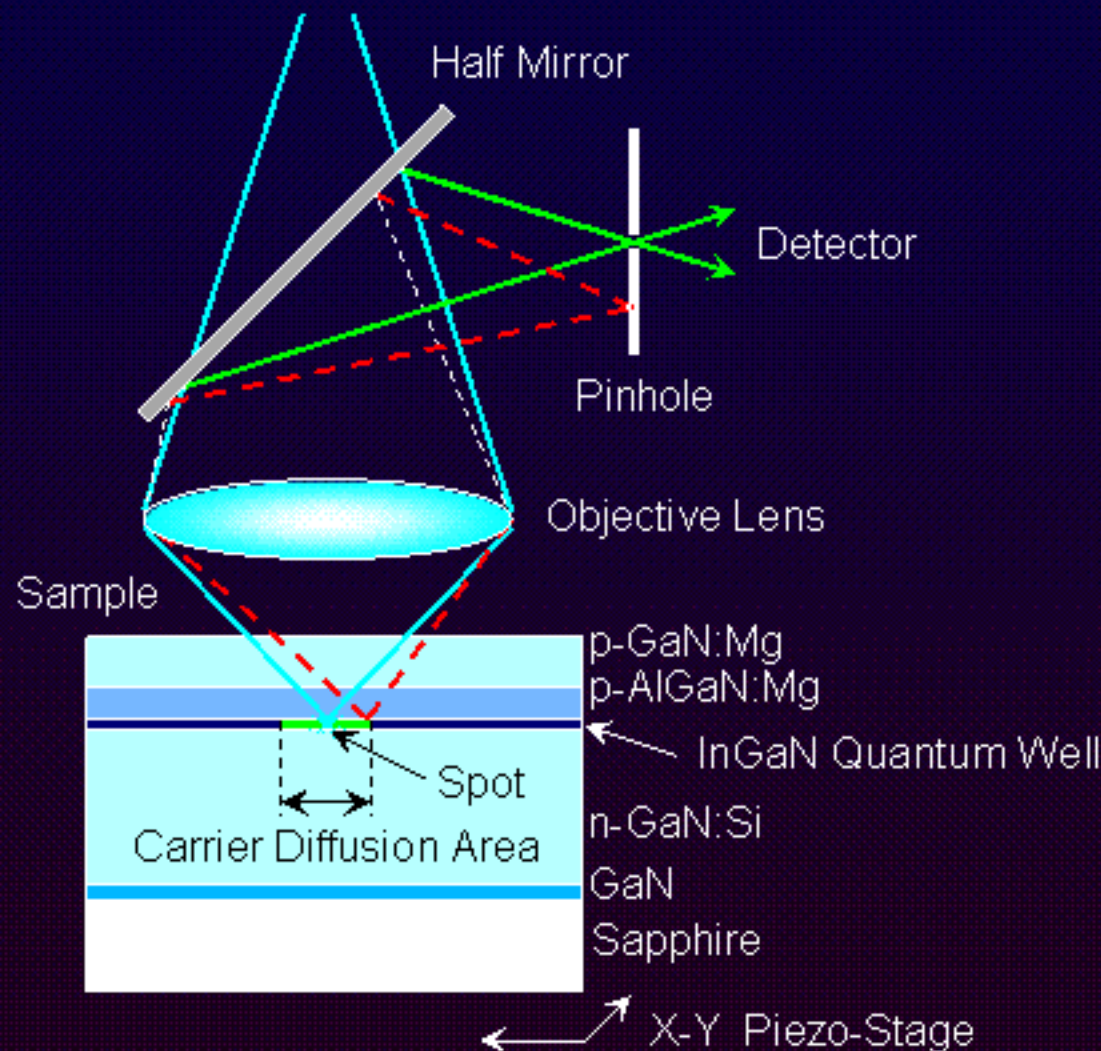


Confocal Microscopy

© Koichi Okamoto

K. Okamoto, et al., *Jpn. J. Appl. Phys.* 43, 839 (2004)

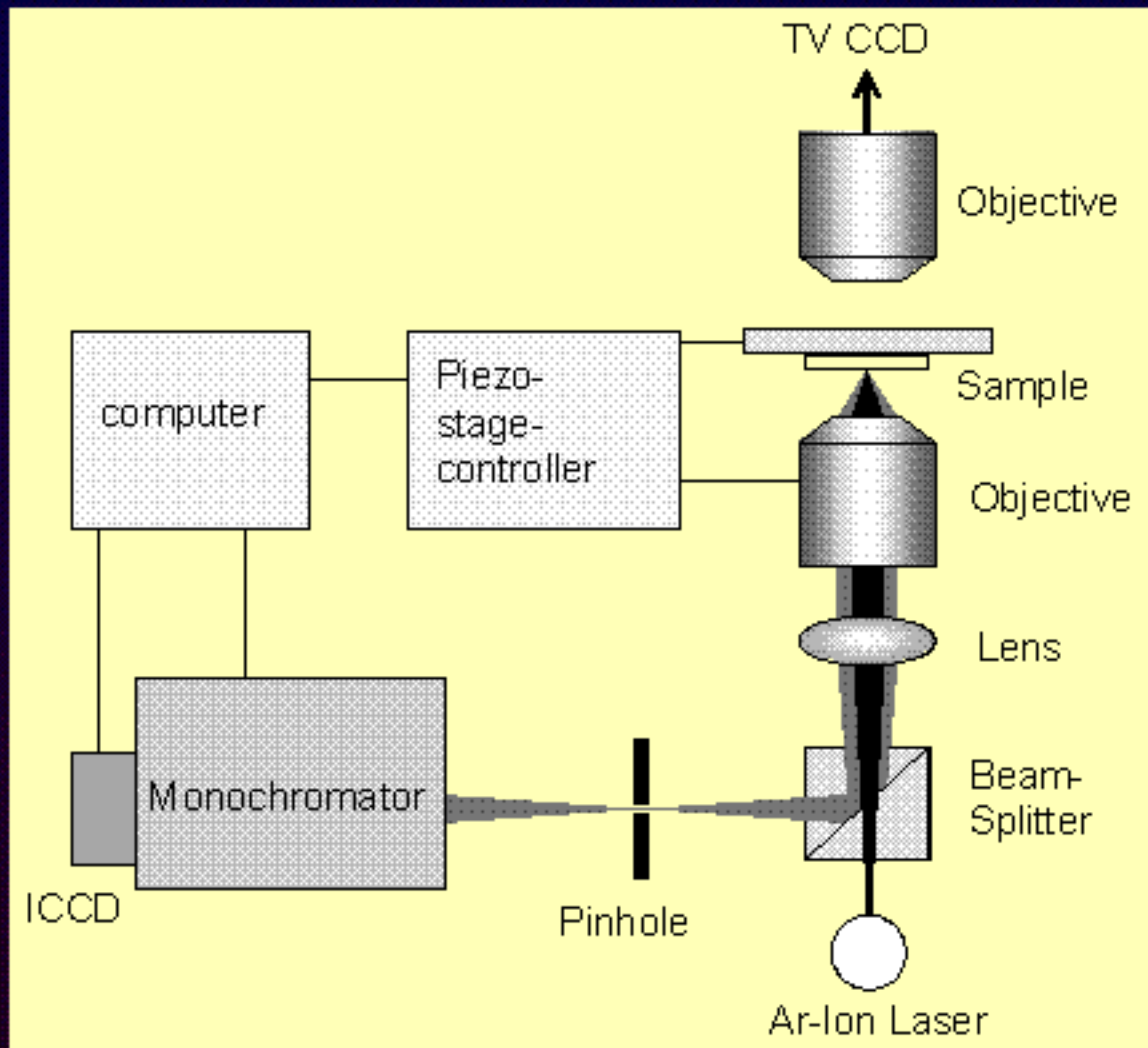
Excitation Laser





Experimental setup

© Koichi Okamoto

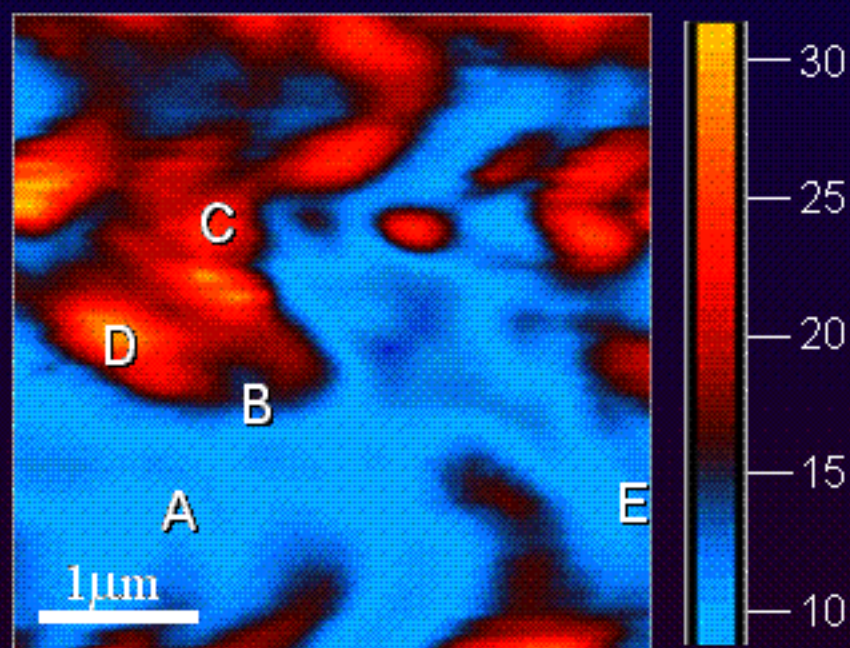




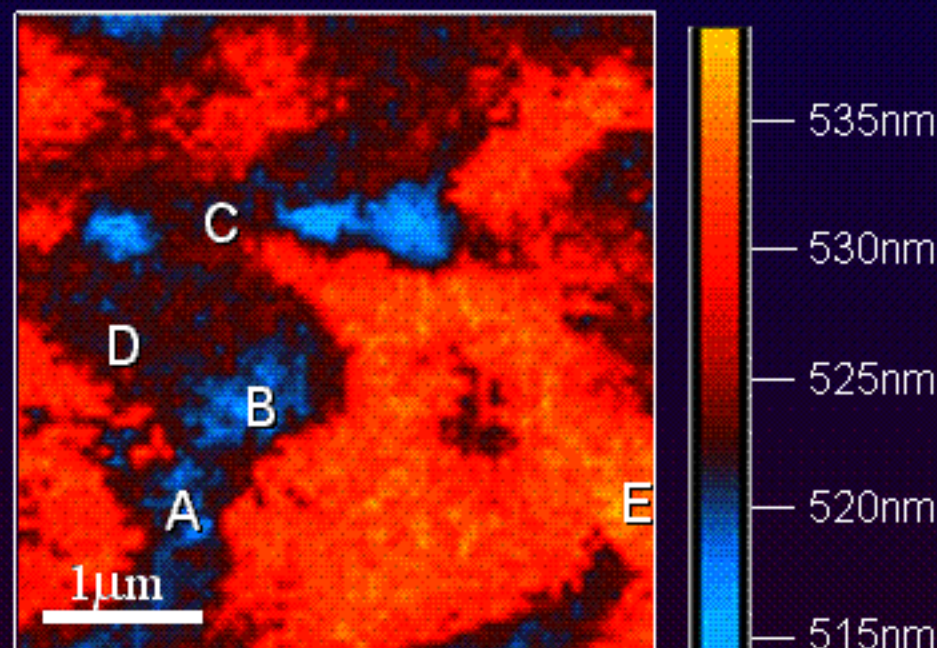
Confocal microscopic image

© Koichi Okamoto

InGaN/GaN LED 540nm @R.T. Excitation: Ar laser (488nm) $4 \times 4 \mu\text{m}$



Peak Intensity Mapping (a.u.)



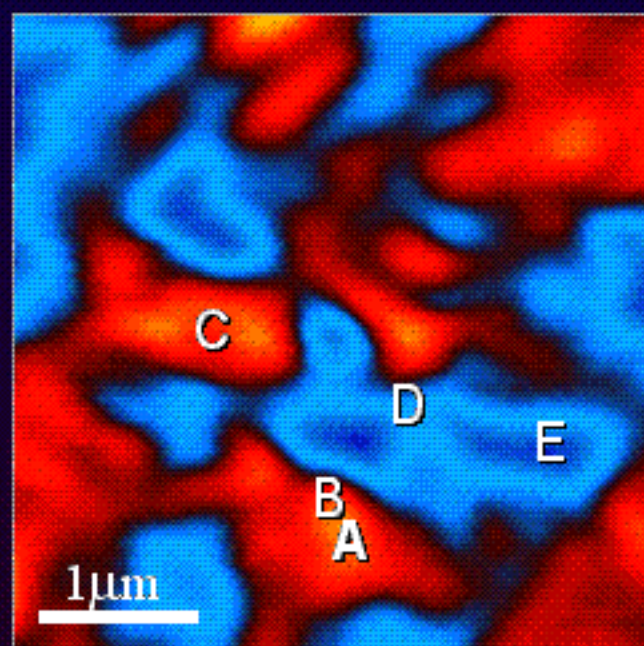
Peak Wavelength Mapping (nm)



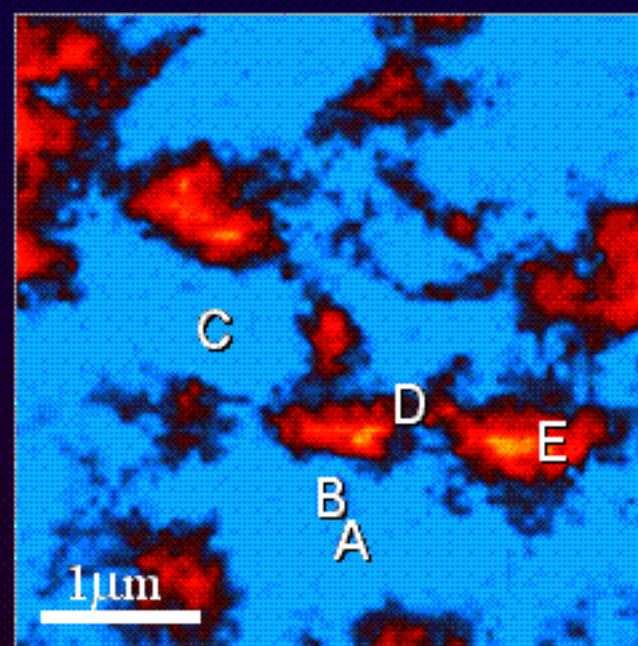
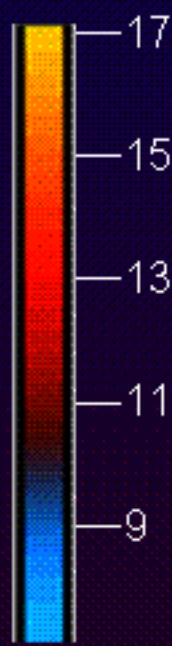
Confocal microscopic image

© Koichi Okamoto

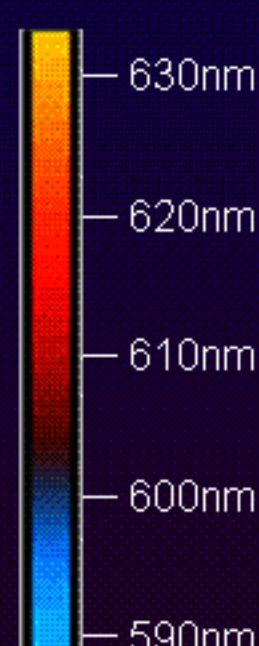
InGaN/GaN LED 600nm @R.T. Excitation: Ar laser (488nm) $4 \times 4 \mu\text{m}$



Peak Intensity Mapping (a.u.)



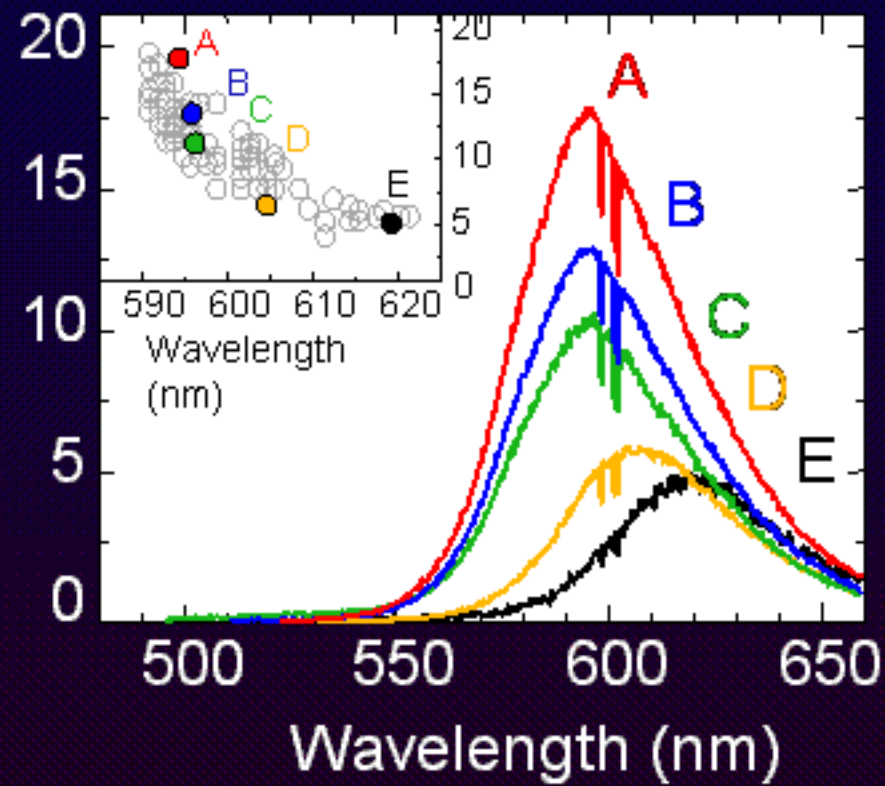
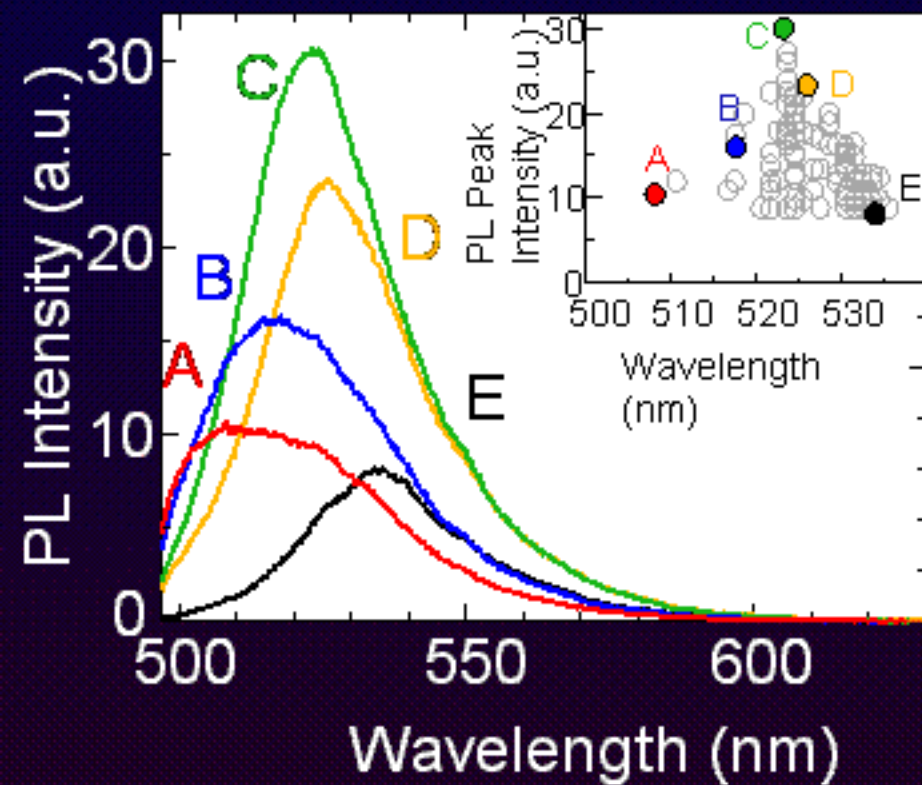
Peak Wavelength Mapping (nm)





Confocal microscopic PL spectra

© Koichi Okamoto

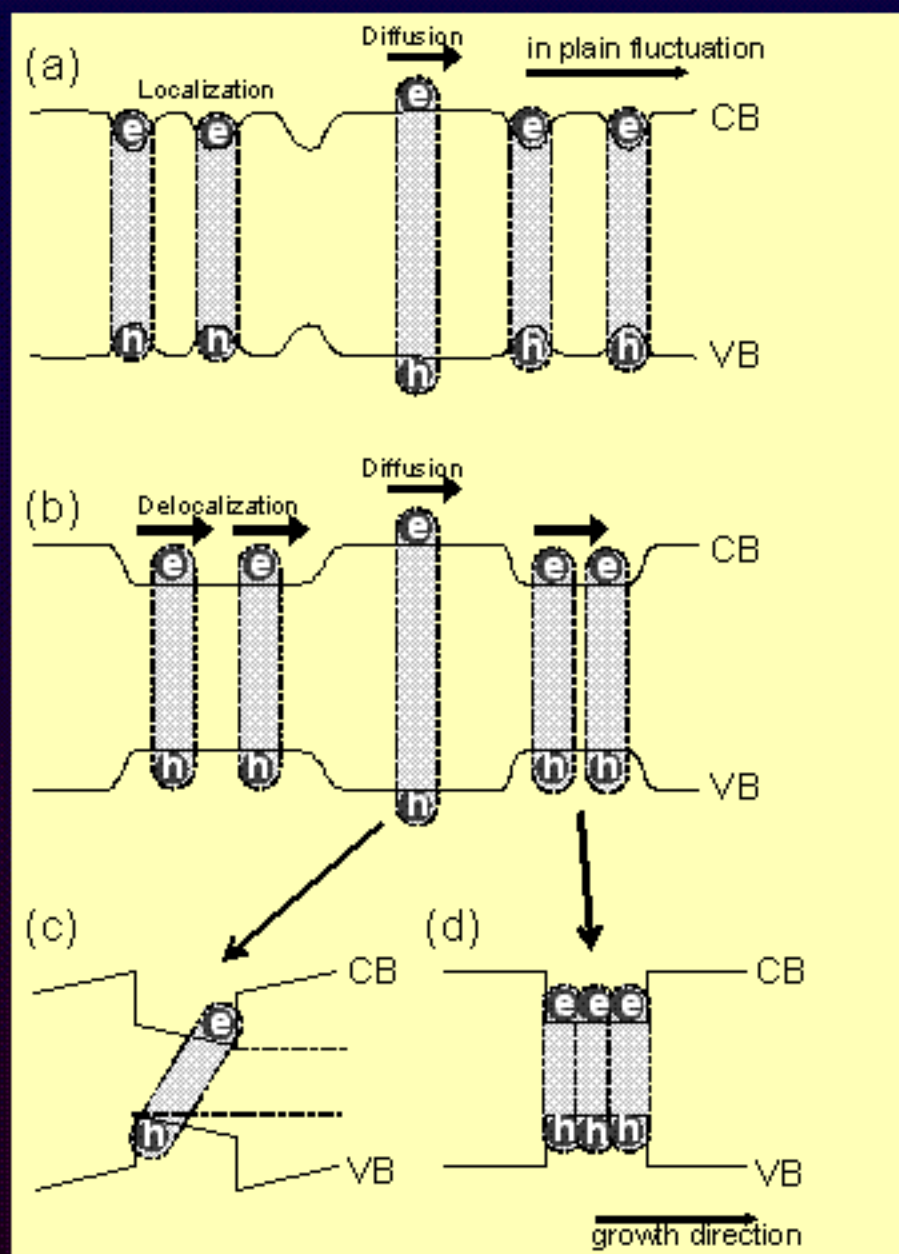




Band diagram fluctuation and

© Koichi Okamoto

exciton dynamics



Schematic band diagrams of in-plane fluctuation and diffusion, localization, and recombination dynamics of electron-hole pairs in InGaN active layers which have

(a) low indium composition (blue or green emission)

(b) high indium composition (amber emission).

(c) Schematic band diagram strained by strong piezoelectric field along the growth direction.

(d) Partial reduction of QCSE by band-filling at the high density regions of generated electron-hole pairs.

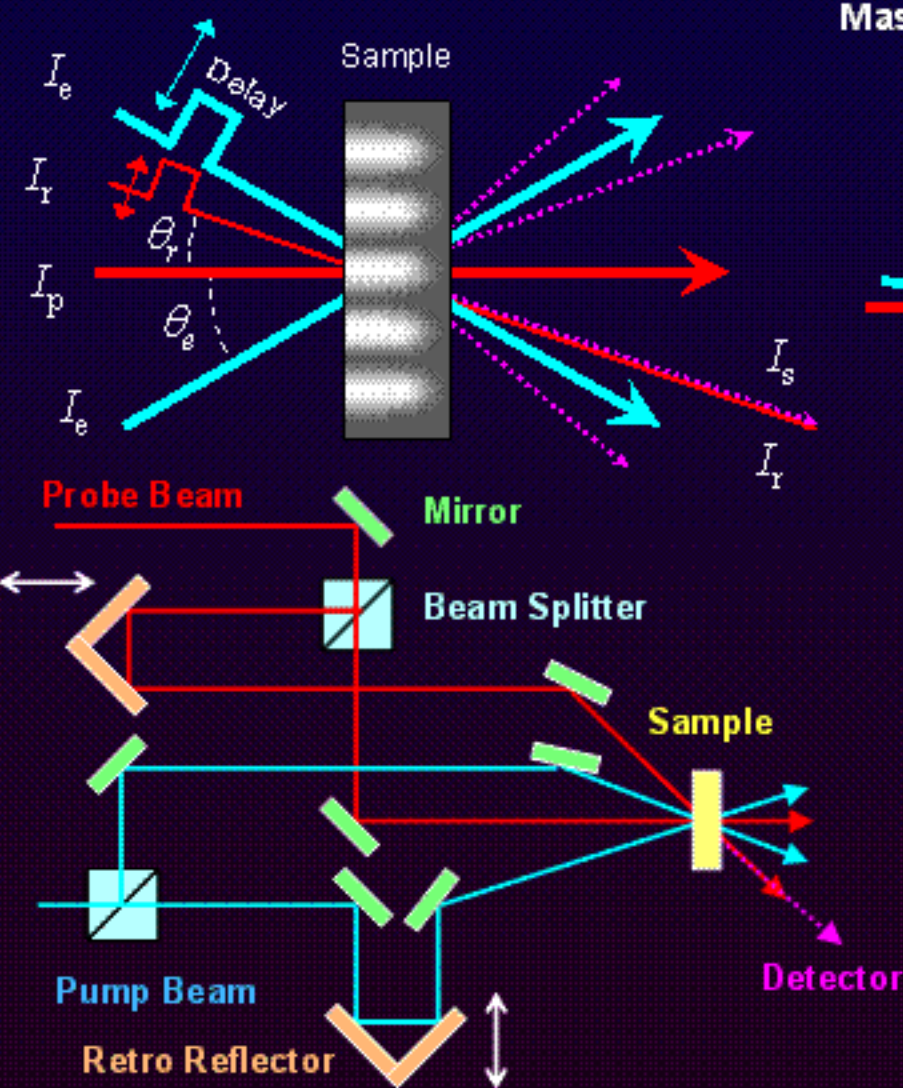


More advanced and simple technique

© Koichi Okamoto

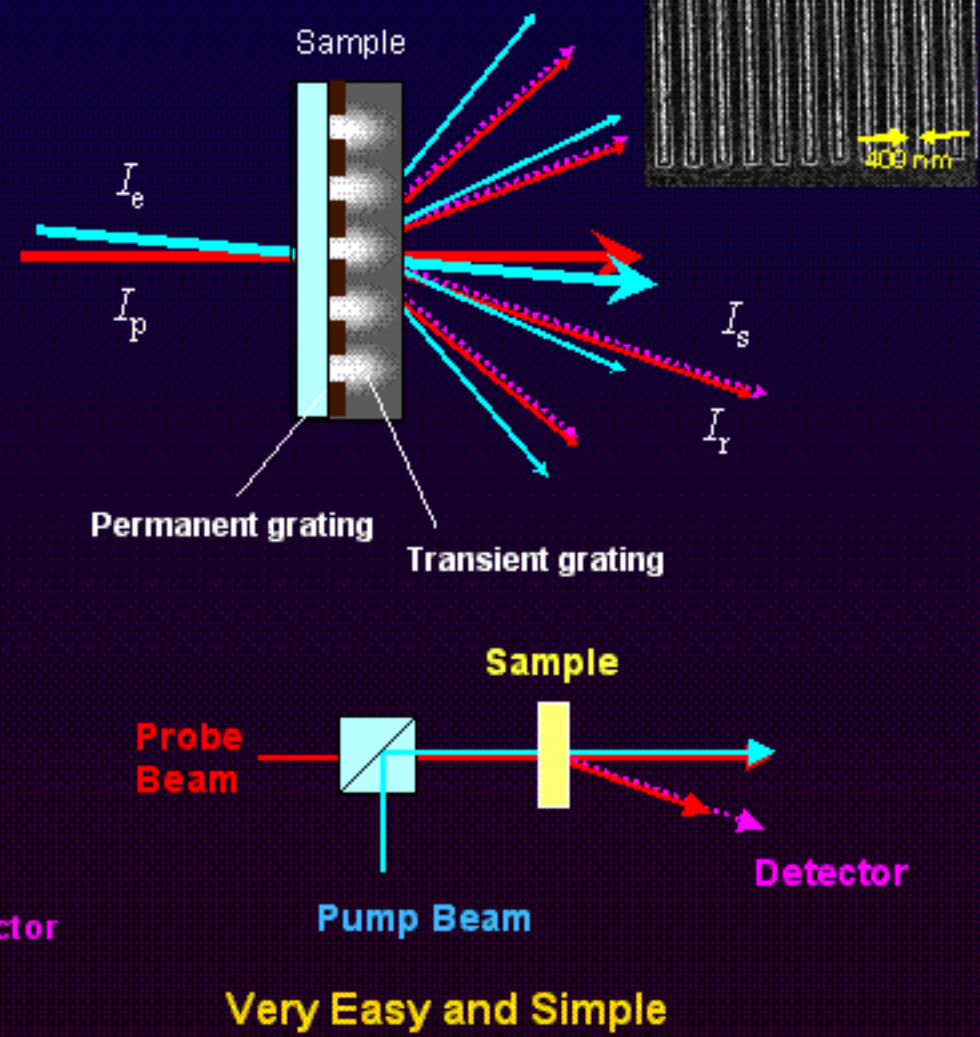
K. Okamoto, et al, Appl. Phys. Lett., 85, 4842 (2004)

Optical Heterodyne detected TG



Our invented new method

Mask pattern transfer-TG (MPT-TG)



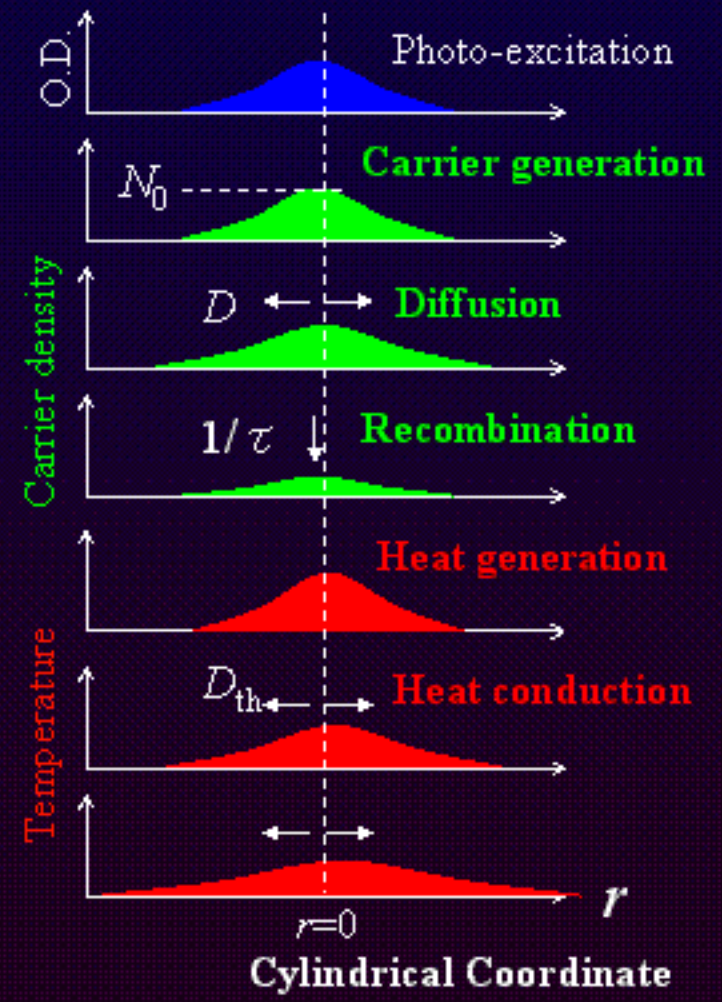
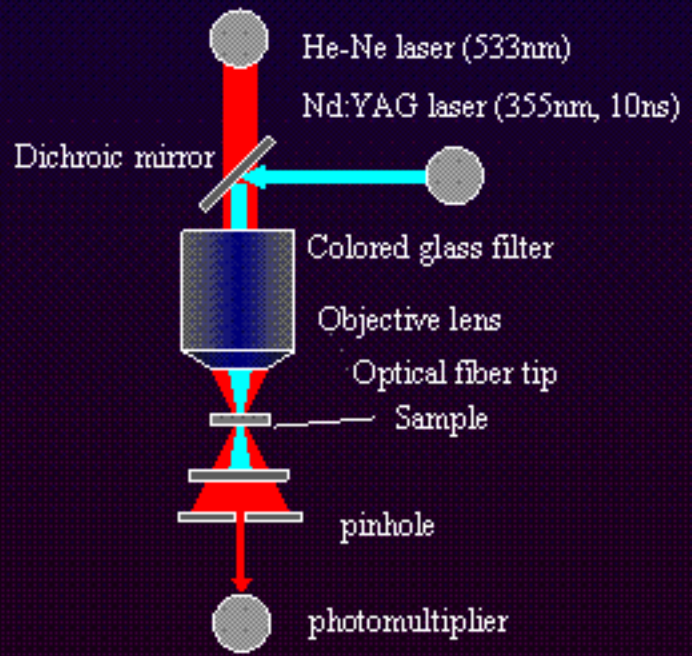
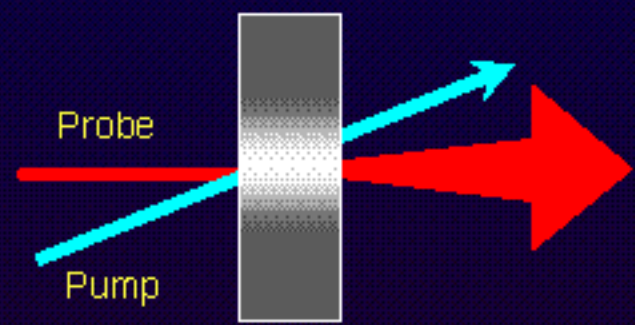


Temporal and spatial-resolved measurement

© Koichi Okamoto

3rd order Nonlinear Spectroscopy + Spatial Resolution

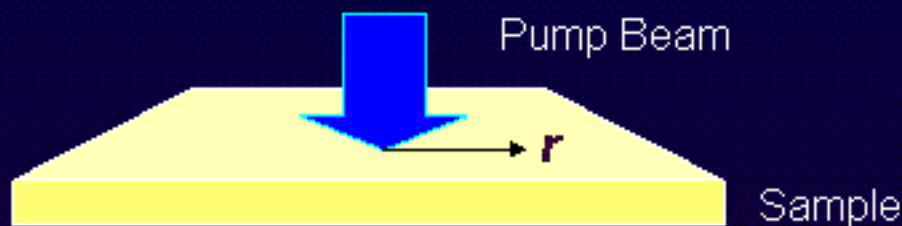
Transient Lens (TL) Spectroscopy



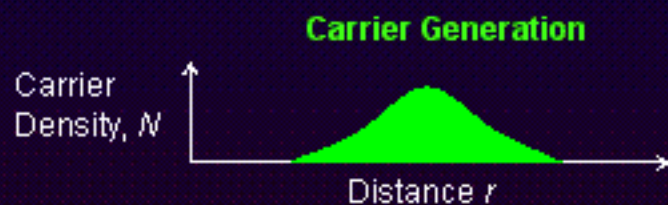
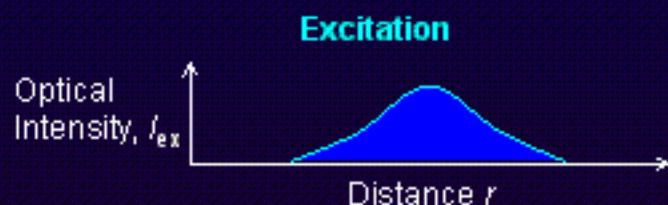


Transient Lens Effect

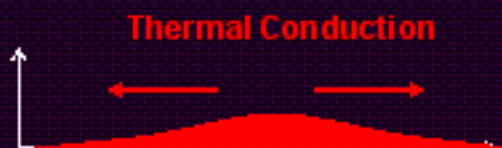
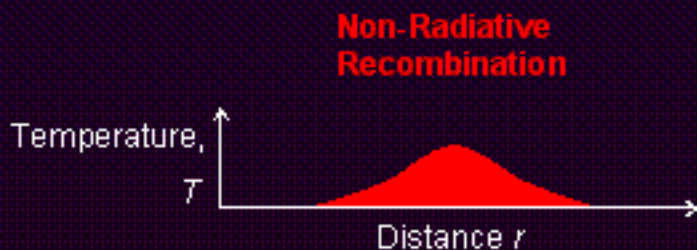
© Koichi Okamoto



Transient Lenses



$(\Delta n / \Delta N) < 0$
Concave Lens
Refractive index



$(\Delta n / \Delta T) > 0$
Convex Lens
Refractive index

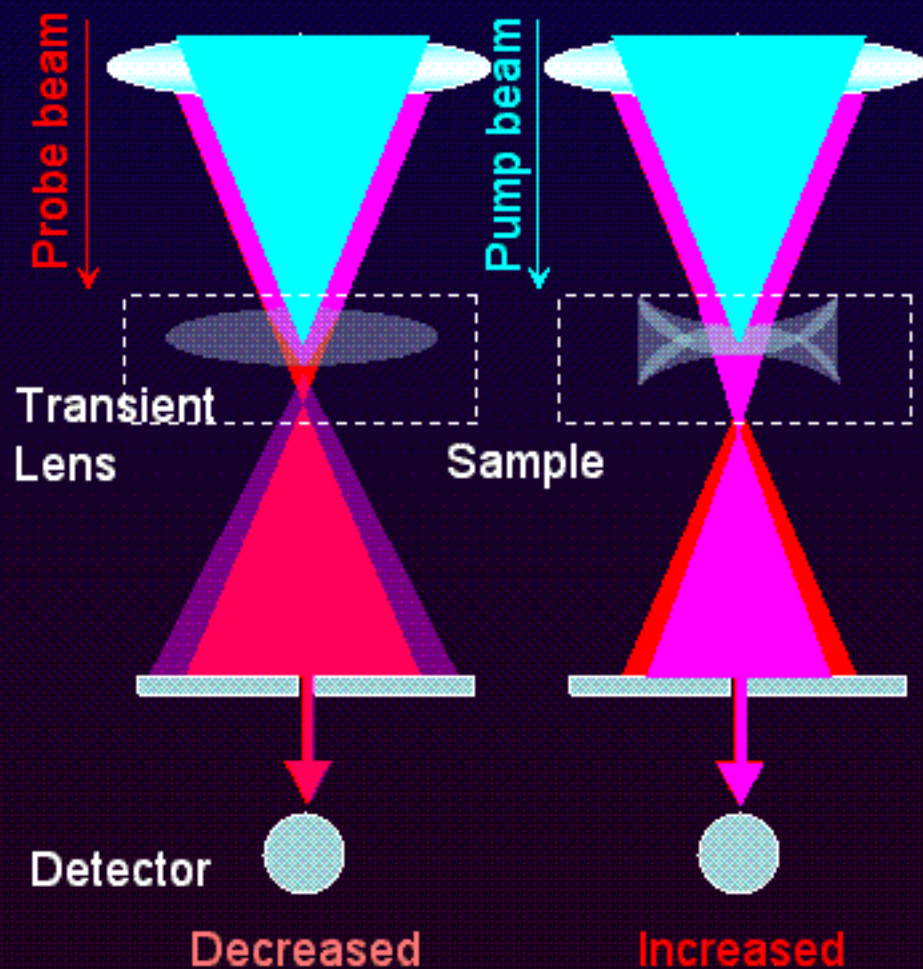




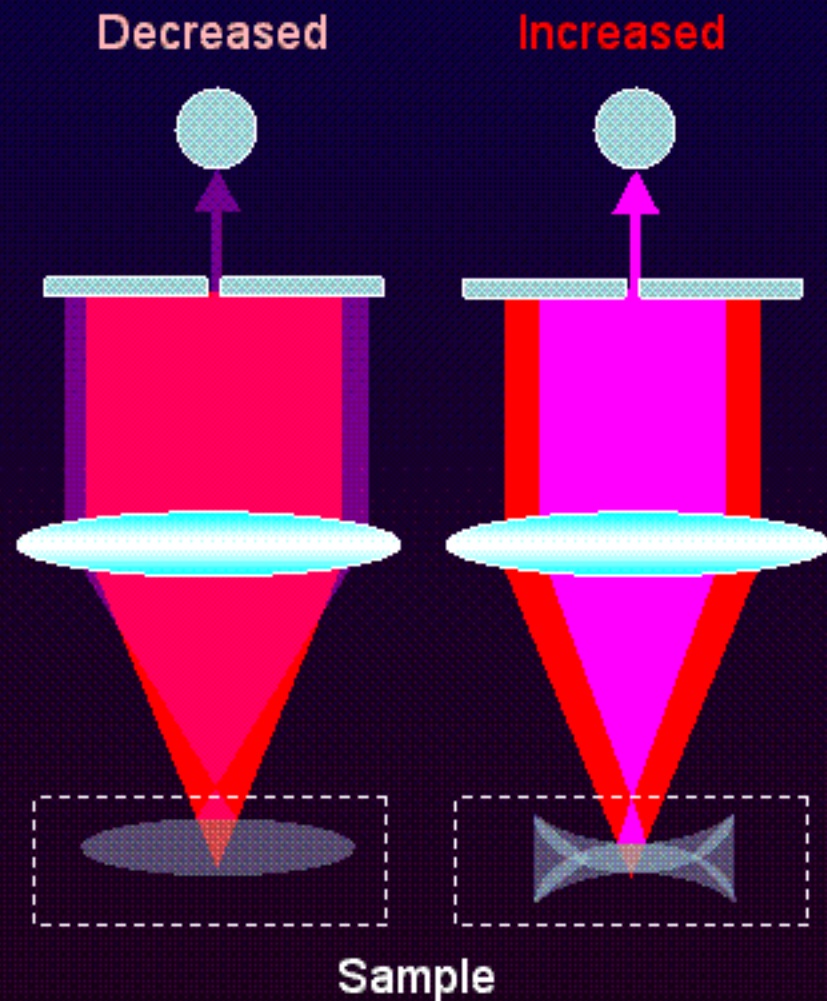
Detection of Transient Lens

© Koichi Okamoto

Transparent



Reflective



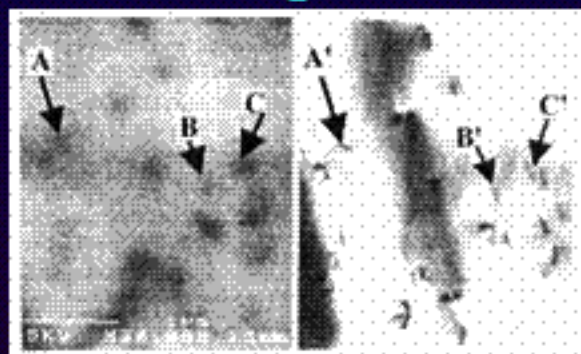


Threading dislocation (TD)

© Koichi Okamoto

At the room temperature, a large amount of carriers lost the energies by the nonradiative recombination

What is the origin of the nonradiative recombination center (NRC)??



T. Sugahara et al., Jpn. J. Appl. Phys. 37, L398 (1998)
Comparison between transmission electron microscopy (TEM) and cathodoluminescence (CL) mapping



Threading dislocation (TD) should act as the NRC

low dislocated GaN have been achieved by using the ELOG technique

T. Mukai et al., Jpn. J. Appl. Phys. 37 L839 (1998) *Epitaxial Lateral Over Growth*

Emission efficiency of the LEDs grown on ELOG-GaN were not so enhanced

S. Chichibu et al., Appl. Phys. Lett. 74 1460 (1999)

Optical properties of low dislocated region were not so difference with high dislocated region.

T. Izumi et al., Journal of Luminescence 87-89 1196 (2000)

Difference of emission spectra and lifetimes was not observed at

each region

TD should not act as NRC ??? Point defect should be more effective???



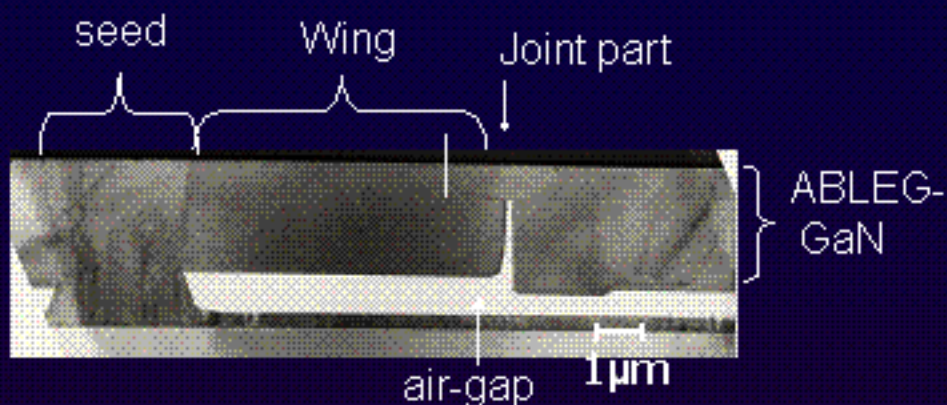
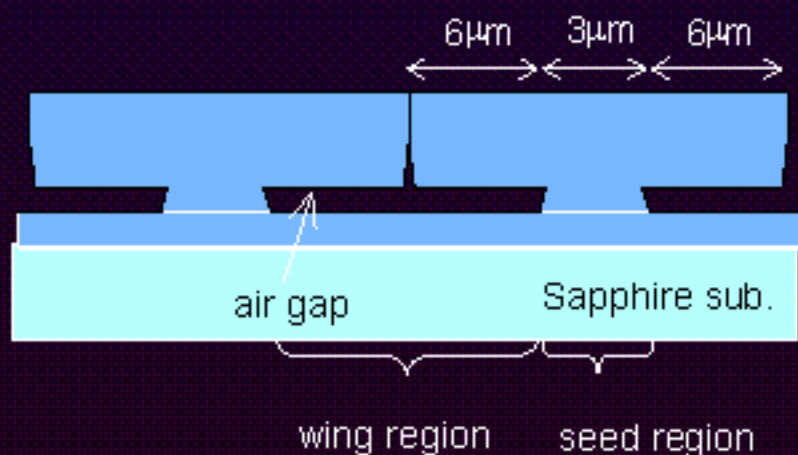
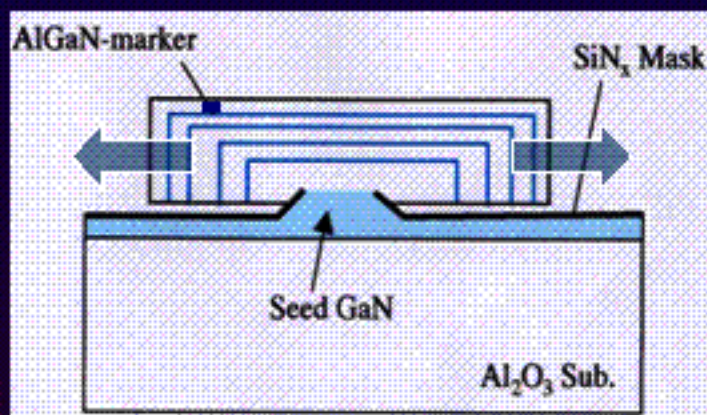
Air-Bridged Lateral Epitaxial Growth

© Koichi Okamoto

(ABLEG)

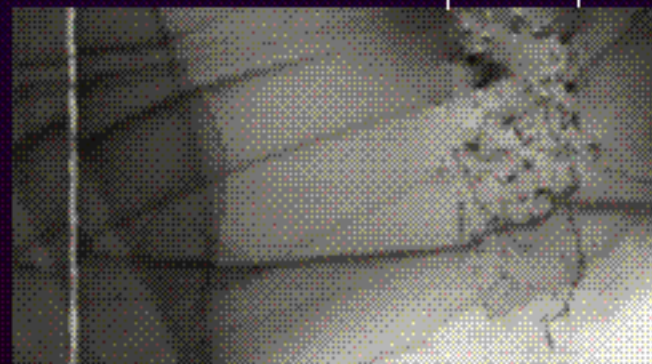
grown by A. Tsujimura, I. Kidoguchi,
Advanced Technology Research Labs., Matsushita Electric Industrial Co.

- Grew GaN (1 μm) on Sapphire(0001)
- Etched GaN for stripe structure
- Masked at bottom and side by SiN_x
- Grew GaN on the non-Mask seed region



TEM Image (cross-section)

$4 \times 10^7 \text{ cm}^{-2}$ (Seed GaN)
 10^6 cm^{-2} (Wing GaN)
 10^9 cm^{-2} (Seed GaN)



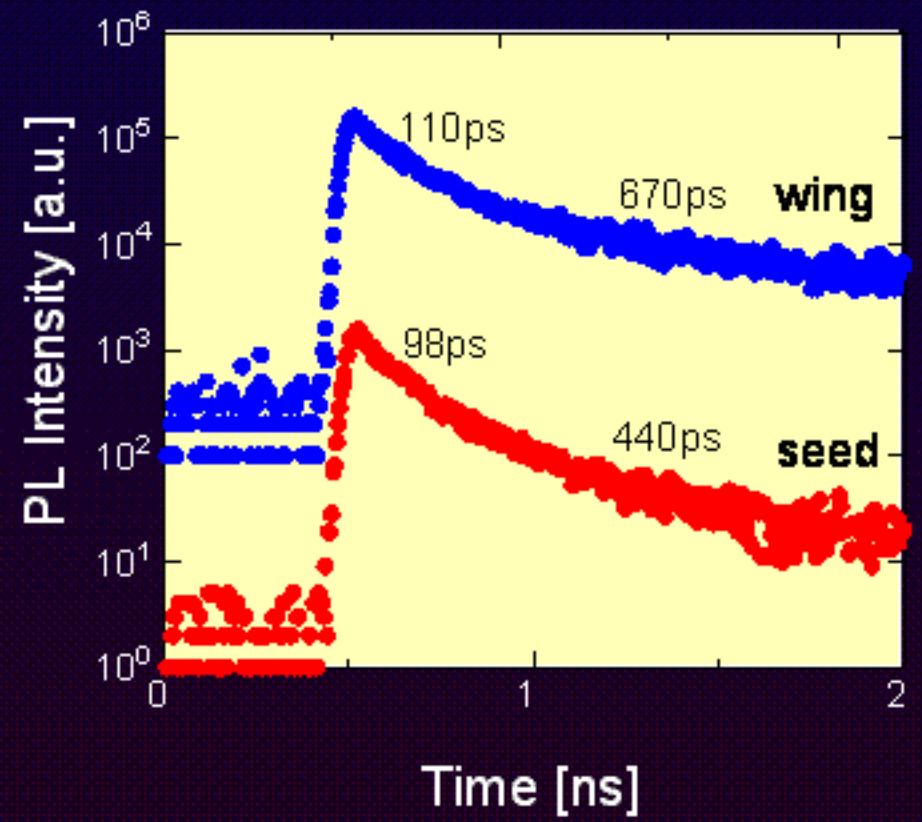
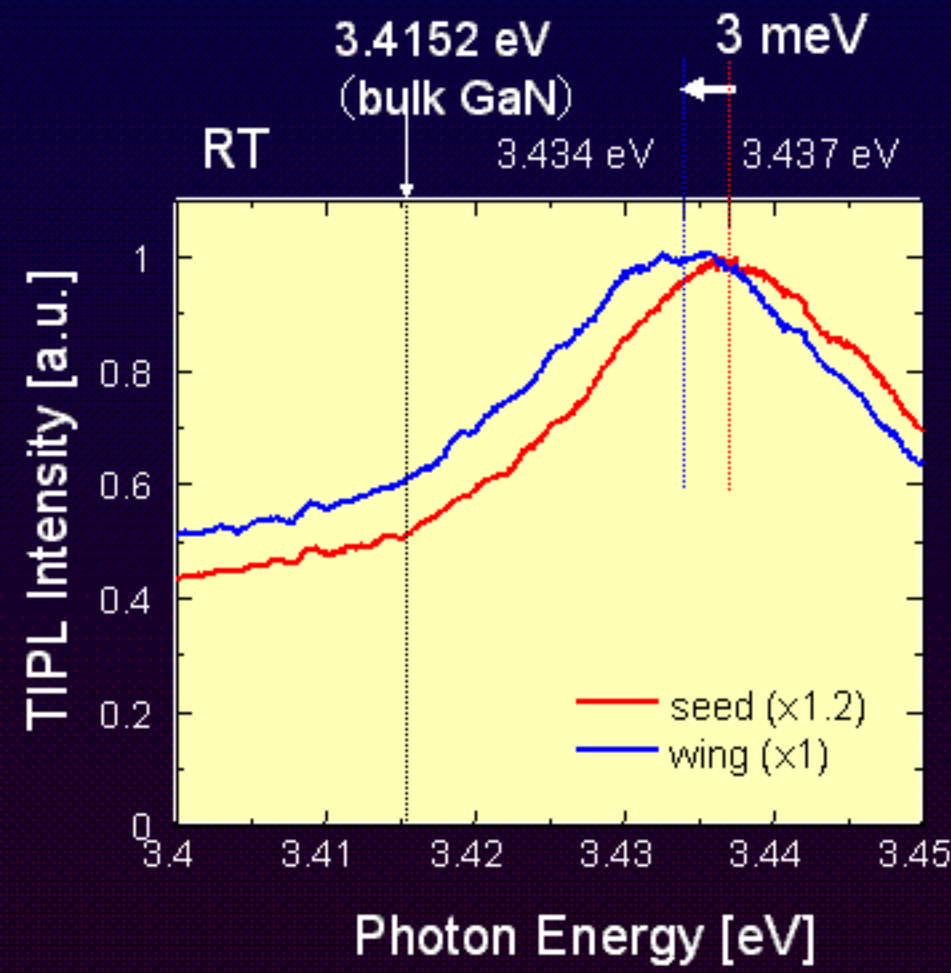
TEM Image

1 μm



Optical Properties of ABLEG-GaN

© Koichi Okamoto



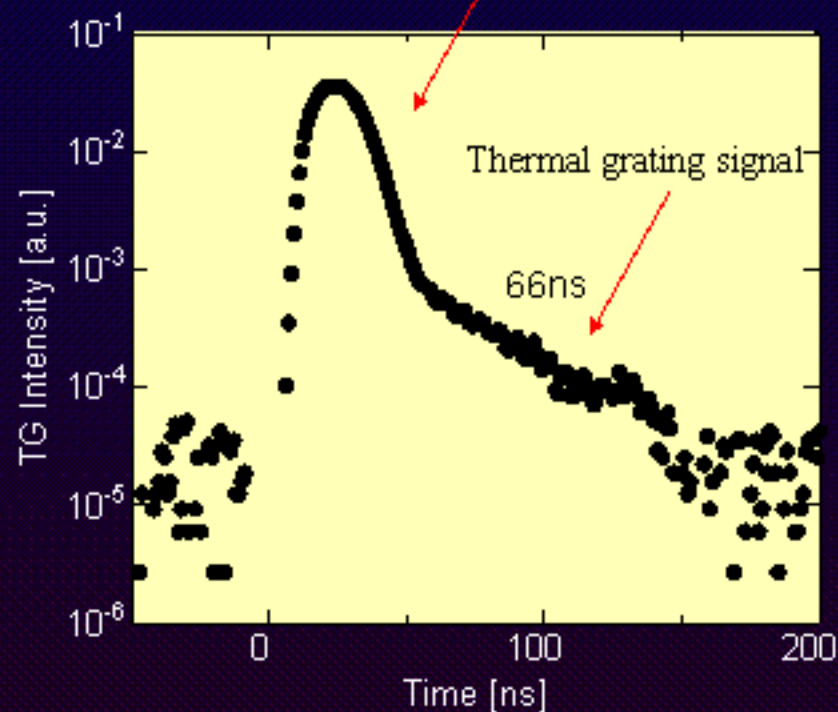


TG measurement of ABLEG-GaN

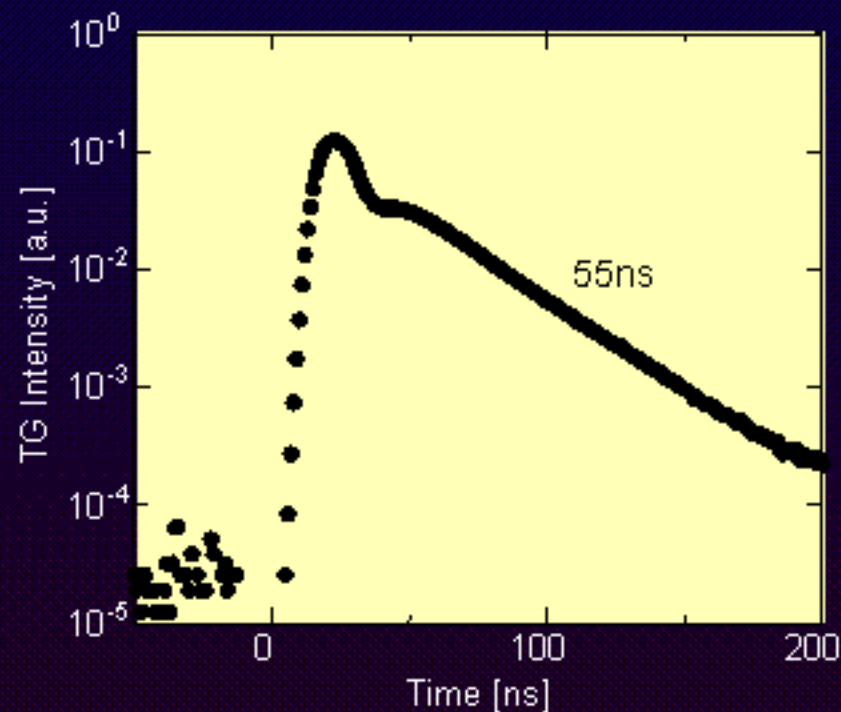
© Koichi Okamoto

ABLEG-GaN

Carrier grating signal



GaN



Thermal signal intensity, ABLEG-GaN < GaN

Heat generation of ABLEG-GaN should be reduced

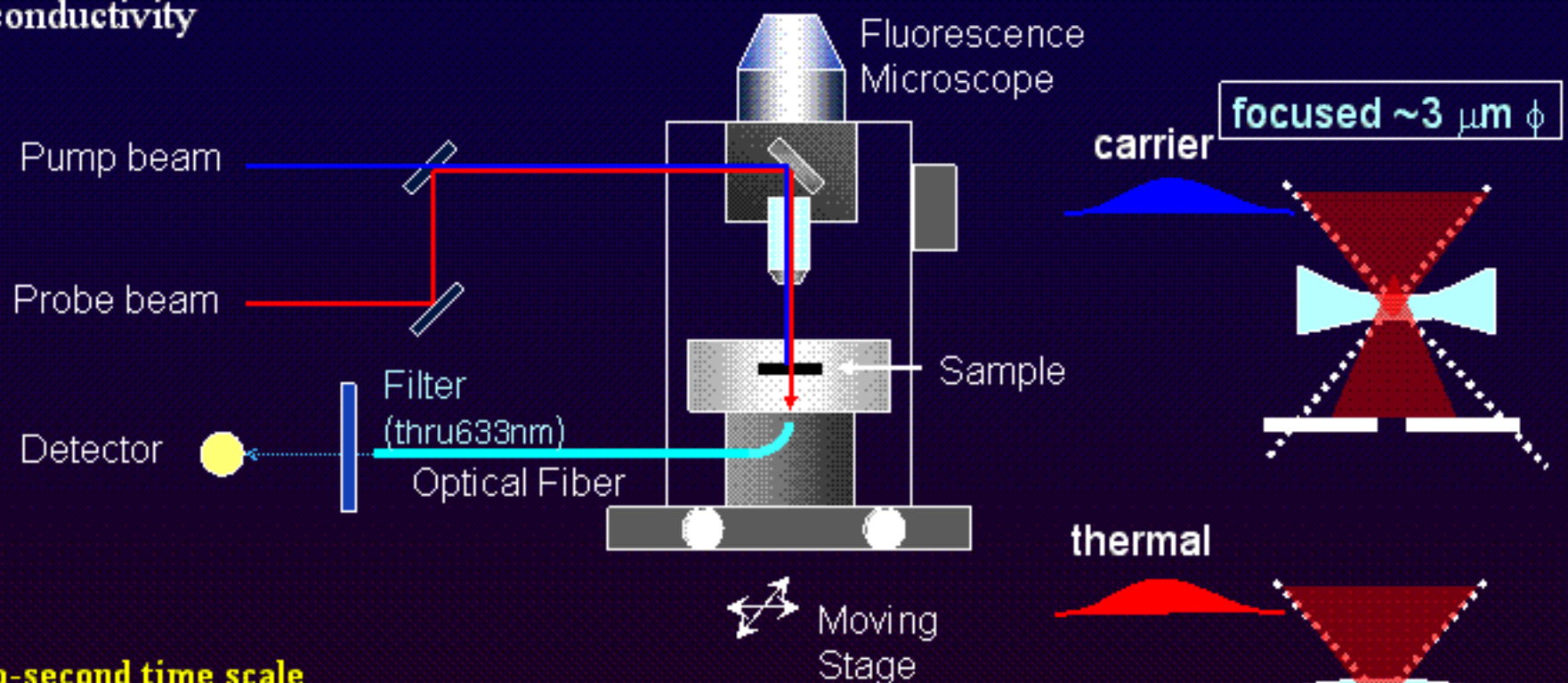
→ Is this difference due to at the Seed region and the Wing region??



Experimental Setup of Micro-TL

© Koichi Okamoto

Time and Spatially resolved observation of carrier diffusion, thermalization, thermal conductivity



Pico-second time scale

Pump: OPA of Ti:Al₂O₃ laser (355nm), Probe: Ti:Al₂O₃ laser (800nm)

Nano-second time scale

Pump: Frequency-tripled of Nd:YAG laser (355nm) Probe: He-Ne laser (633nm)

Continuum wave

Pump: He-Cd laser (325nm), Probe: He-Ne laser (633nm)

$$\left(\frac{\partial n}{\partial N}\right) < 0, \quad \left(\frac{\partial n}{\partial T}\right) > 0$$



Theory of Transient lens -1-

© Koichi Okamoto

Gaussian spatially distribution of the refractive index change δn can be written by cylindrical coordinate

$$\delta n(r) = \delta n_0 \exp(-r^2/w_0^2)$$

w_0 : pump beam width

δn_0 : Refractive index change at the beam center

Modulation of the carrier density (δN) and the temperature (δT) by the nonradiative recombination are given by

$$\delta n(r,t) = \left(\frac{\partial n}{\partial N}\right) \delta N(r,t) + \left(\frac{\partial n}{\partial T}\right) \delta T(r,t) \quad \left(\frac{\partial n}{\partial N}\right) < 0, \quad \left(\frac{\partial n}{\partial T}\right) > 0$$

Time and Spatially dynamics of δn , δT are described by following rate-equation.

$$\frac{\partial \delta N(r,t)}{\partial t} = D \frac{\partial^2 \delta N(r,t)}{\partial r^2} - \left(\frac{1}{\tau_{rad}} + \frac{1}{\tau_{non-rad}}\right) \delta N(r,t)$$

Carrier density

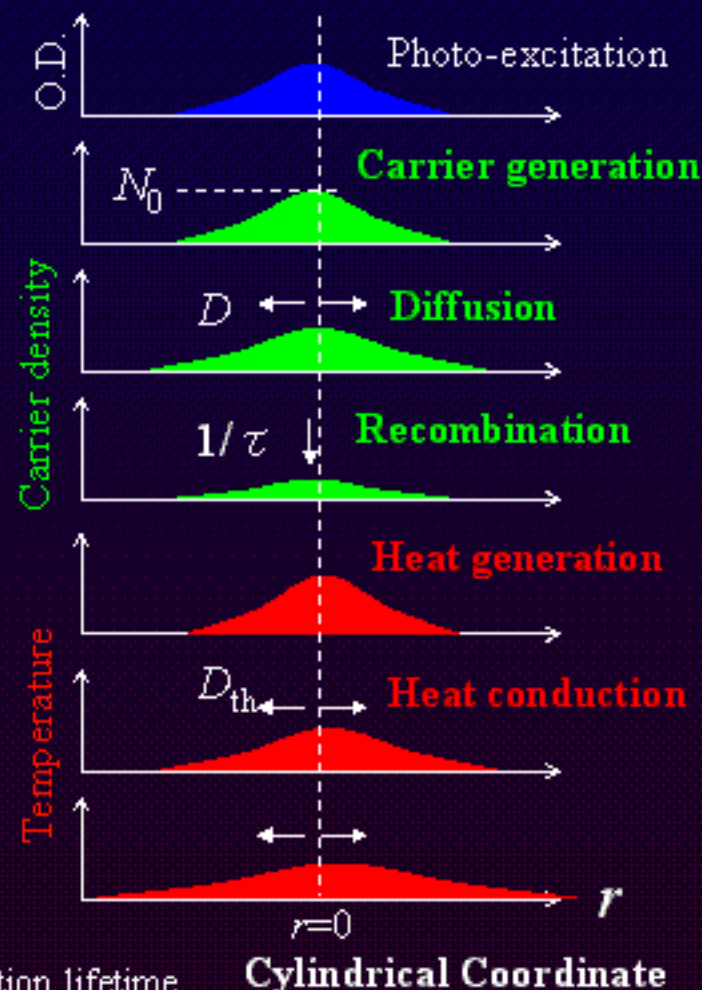
$$\frac{\partial \delta T(r,t)}{\partial t} = \frac{1}{\rho C_p} \frac{Q \delta N(r,t)}{\tau_{non-rad}} + D_{th} \frac{\partial^2 \delta T(r,t)}{\partial r^2}$$

Temperature

D : Diffusion coefficient of carriers, D_{th} : Thermal diffusion coefficient

τ_{rad} : Radiative recombination lifetime, $\tau_{non-rad}$: Non-radiative recombination lifetime

Q : Heat amount released from unit density of carriers



Cylindrical Coordinate



Theory of Transient lens -2-

© Koichi Okamoto

Solved the rate equation with the assumption of that the carrier recombination process is dominant because the pump beam width ($3\mu\text{m}$) is much larger than the diffusion length of carriers ($\sim 50\text{nm}$).

$$\delta N(r, t) = N_0 \exp\left[-\frac{r^2}{w^2} - \left(\frac{1}{\tau_{\text{rad}}} + \frac{1}{\tau_{\text{non-rad}}}\right)t\right]$$

Carrier Dynamics

Thermal Dynamics

$$\delta T(r, t) = \frac{\tau_{\text{rad}}}{\tau_{\text{rad}} + \tau_{\text{non-rad}}} \cdot \frac{QN_0}{\rho C_p} \times \left\{ \exp\left[-\frac{r^2}{w^2} - \left(\frac{1}{\tau_{\text{rad}}} + \frac{1}{\tau_{\text{non-rad}}}\right)t\right] + \frac{w_0^2}{4D_{\text{th}}t + w_0^2} \exp\left(-\frac{r^2}{4D_{\text{th}}t + w_0^2}\right) \right\}$$

The optical pass of the probe beam at the transient lens given by the ABCD law of Gaussian beam

$$S(f) = \frac{I(f) - I(0)}{I(0)} \propto \delta n(r=0, f)$$

Therefore, the time profiles of the carrier lens and thermal lens signal can be written by

$$S(f) = S_0 \exp\left[\left(\frac{1}{\tau_{\text{rad}}} + \frac{1}{\tau_{\text{non-rad}}}\right)ft\right] \quad S(t) = S'_0 \frac{w_0^2}{4D_{\text{th}}t + w_0^2}$$

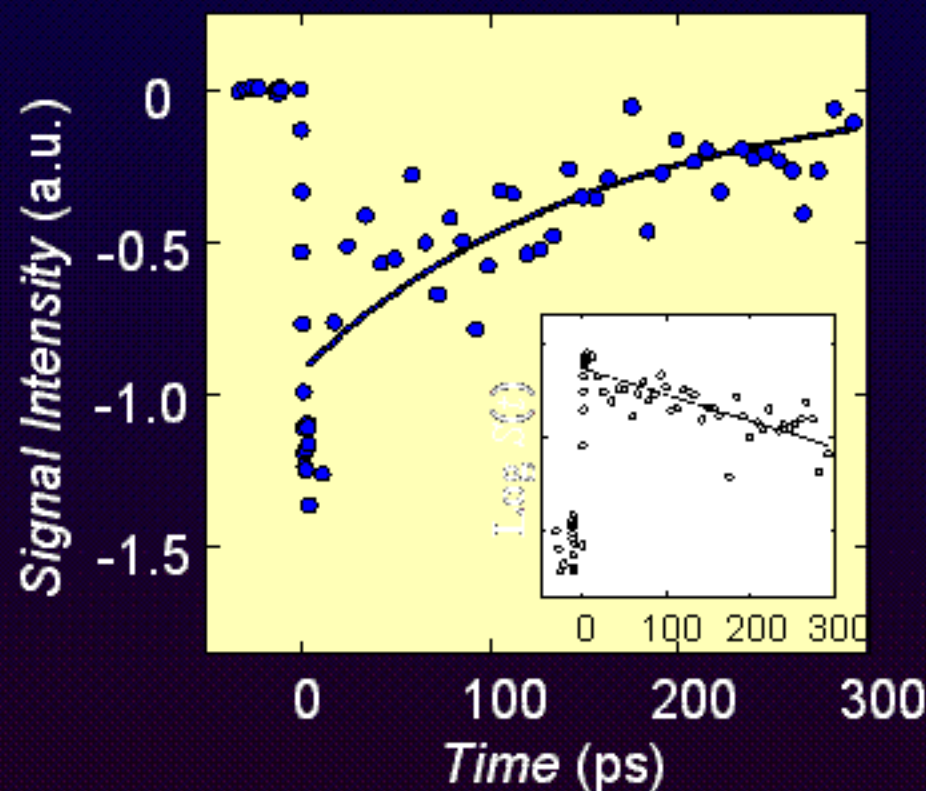
The information about carrier recombination lifetime, amount of the heat generation, thermal conductivity can be obtained by the analysis of the time profile of $S(t)$



Time profiles of MTL signal (GaN)

© Koichi Okamoto

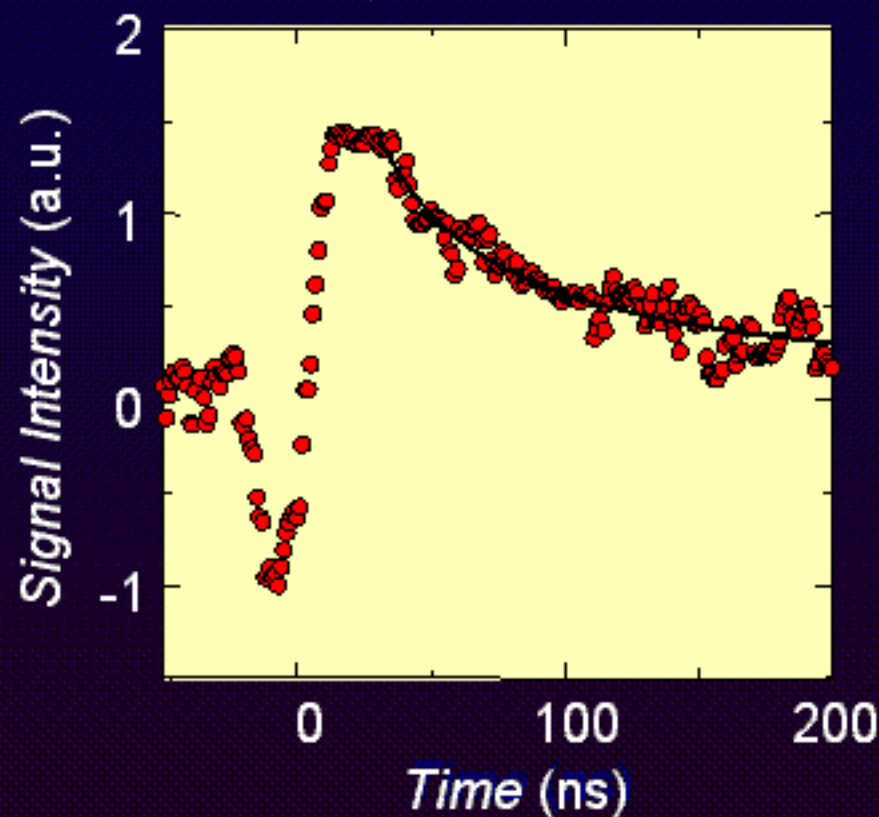
With Femto-second laser



$$S(t) = S_0 \exp \left[\left(\frac{1}{\tau_{rad}} + \frac{1}{\tau_{non-rad}} \right) t \right]$$

155ps

With Nano-second laser



$$S(t) = S'_0 \frac{w_0^2}{4D_{th}t + w_0^2}$$

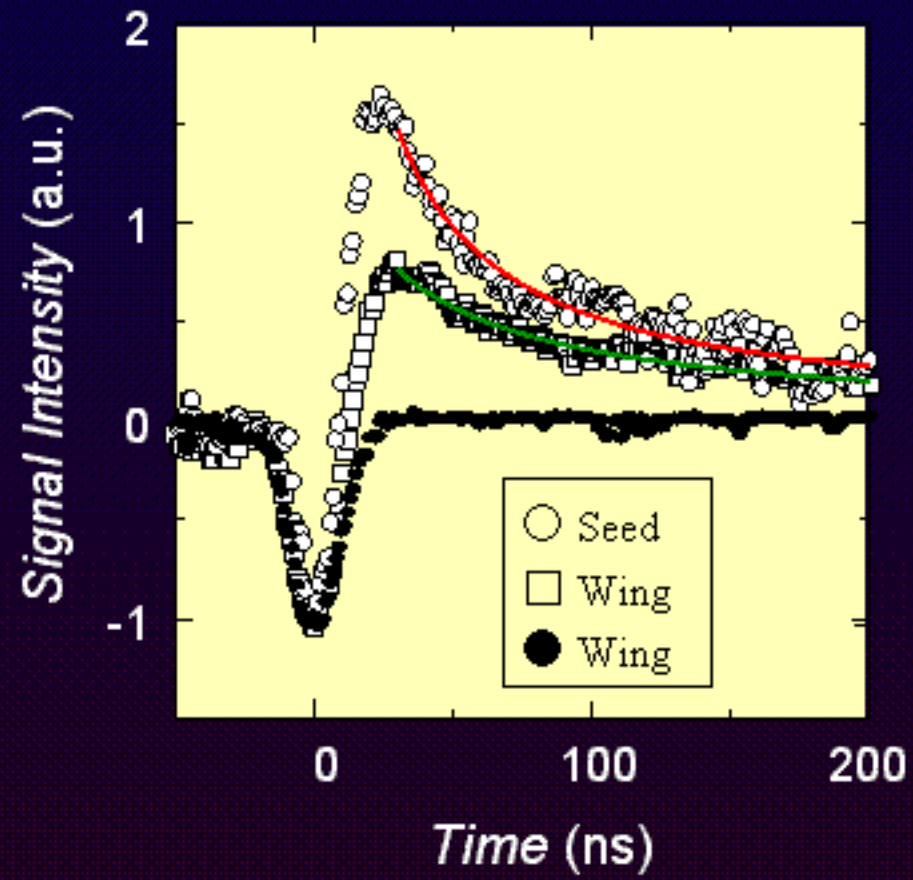
$D_{th} = 0.77 \text{ cm}^2 \text{ s}^{-1}$, $w_0 = 2.6 \mu\text{m}$



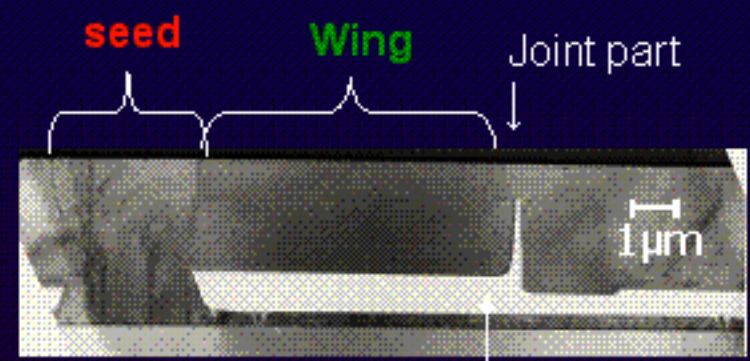
TL measurement for ABLEG-GaN © Koichi Okamoto

Spatial-resolved TL time-profiles

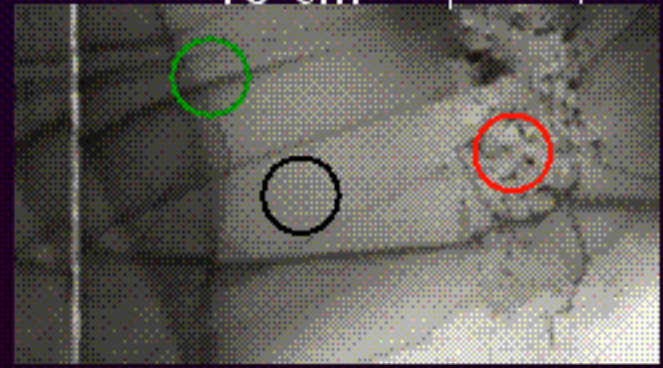
K. Okamoto, et al
Review of Scientific Instruments, 74, 575 (2003).



Thermal Signal Intensity
Seed region > Wing region



TDD = $4 \times 10^7 \text{cm}^{-2}$ 10^6cm^{-2} 10^9cm^{-2}
 Wing GaN Seed GaN





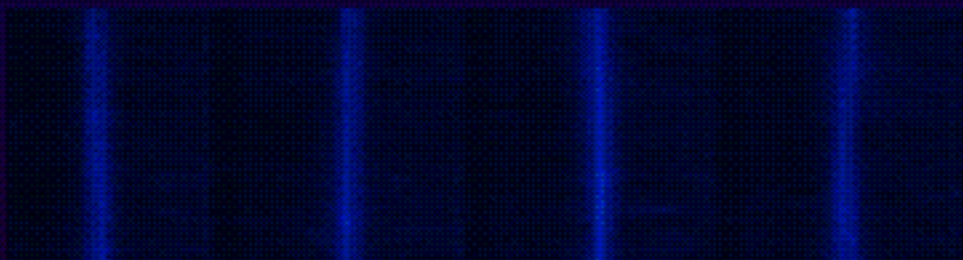
Spatial resolved measurement

© Koichi Okamoto

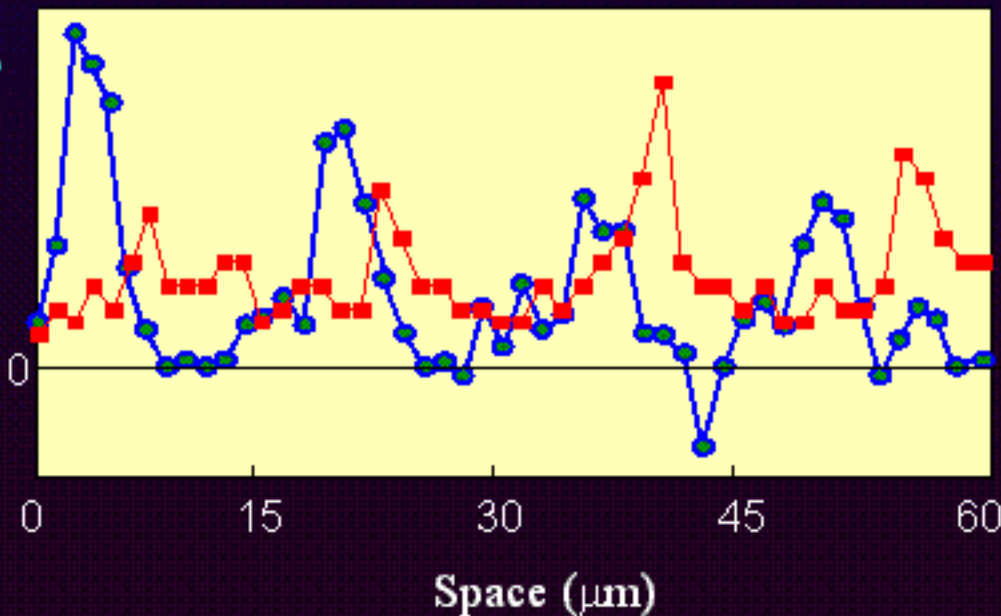
Transparent



Emission



TL Intensity



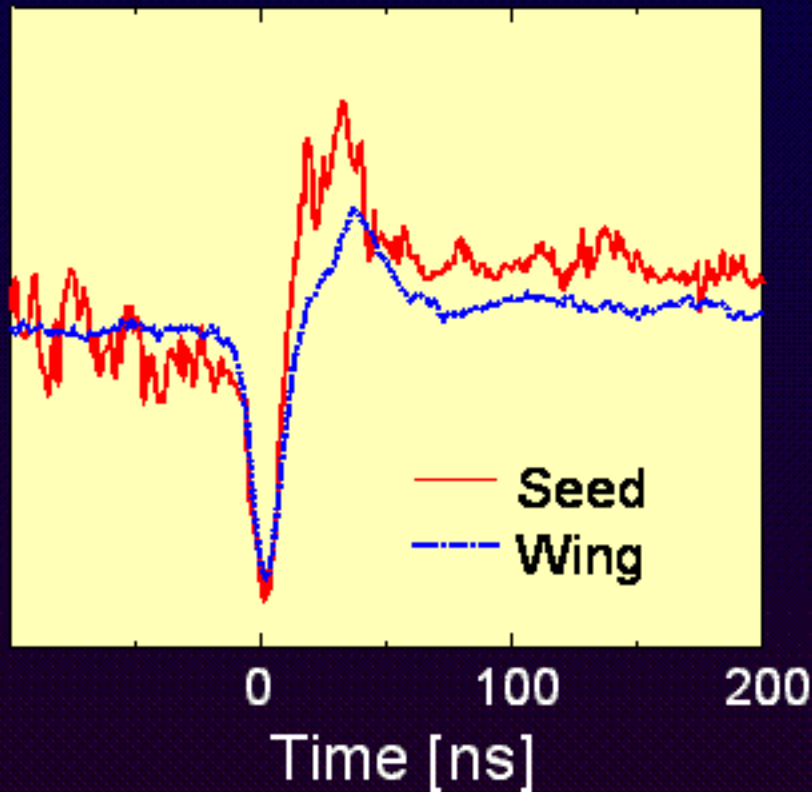
PL Intensity



MTL signal (3QW on ABLEG-GaN)

© Koichi Okamoto

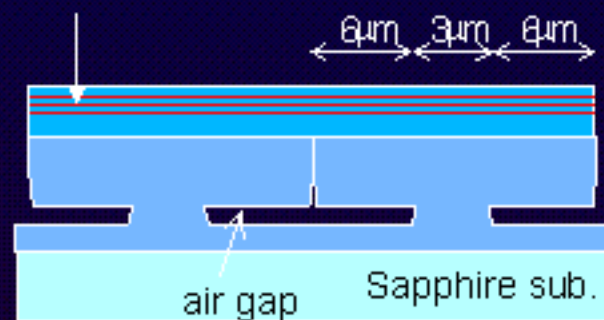
Relative Intensity [a.u.]



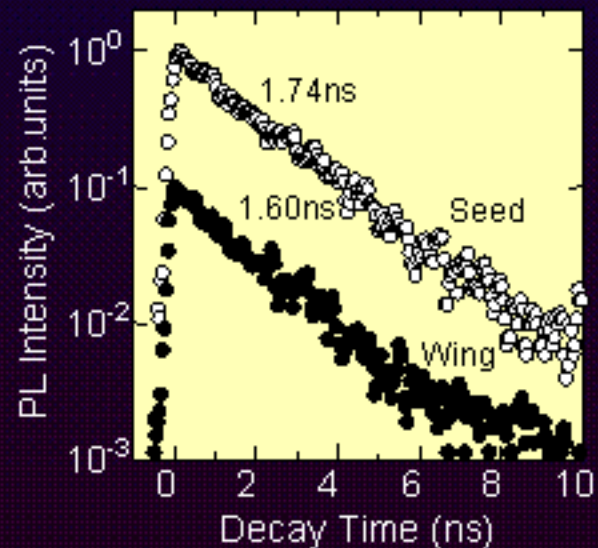
Thermal Signal Intensity

Seed region > Wing region

InGaN/GaN Quantum well layer



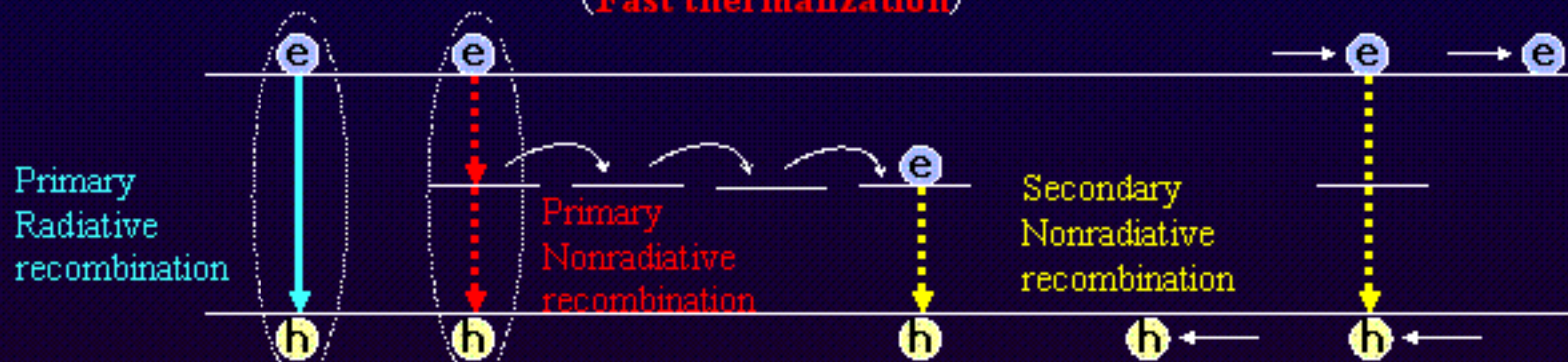
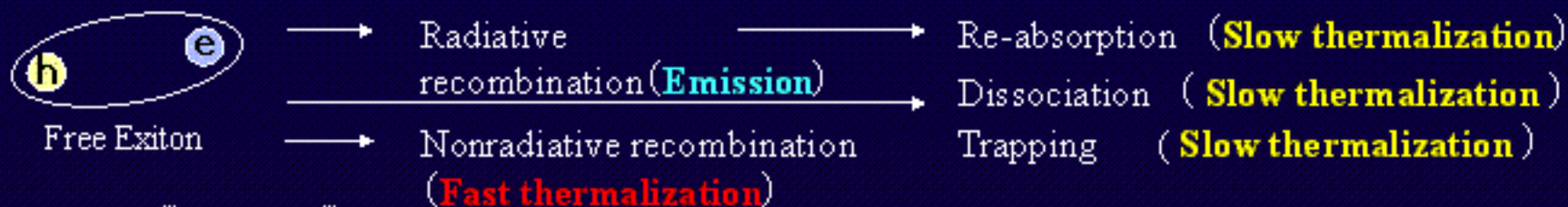
wing region seed region





Thermalization Processes

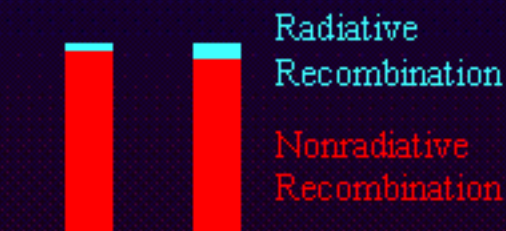
© Koichi Okamoto



$$\frac{1}{\tau_{PL}} = \frac{1}{\tau_{rad}} + \frac{1}{\tau_{non-rad}}$$

$$\eta_{int} = \frac{\frac{1}{\tau_{rad}}}{\frac{1}{\tau_{rad}} + \frac{1}{\tau_{non-rad}}}$$

$$\eta_{th} = \frac{\frac{1}{\tau_{non-rad}}}{\frac{1}{\tau_{rad}} + \frac{1}{\tau_{non-rad}}}$$

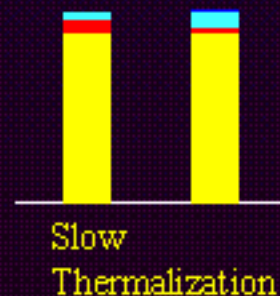


Take into consideration with re-absorption, thermal dissociation, or trapping in the deep level of exciton

$$\frac{1}{\tau_{PL}} = \frac{1}{\tau_{rad}} + \frac{1}{\tau_{nonrad}} + \frac{1}{\tau_{diss}} + \frac{1}{\tau_{trap}}$$

$$\eta_{int} = \frac{\frac{1}{\tau_{rad}}}{\frac{1}{\tau_{rad}} + \frac{1}{\tau_{non-rad}} + \frac{1}{\tau_{diss}} + \frac{1}{\tau_{trap}}}$$

$$\eta_{th} = \frac{\frac{1}{\tau_{non-rad}}}{\frac{1}{\tau_{rad}} + \frac{1}{\tau_{non-rad}} + \frac{1}{\tau_{diss}} + \frac{1}{\tau_{trap}}}$$

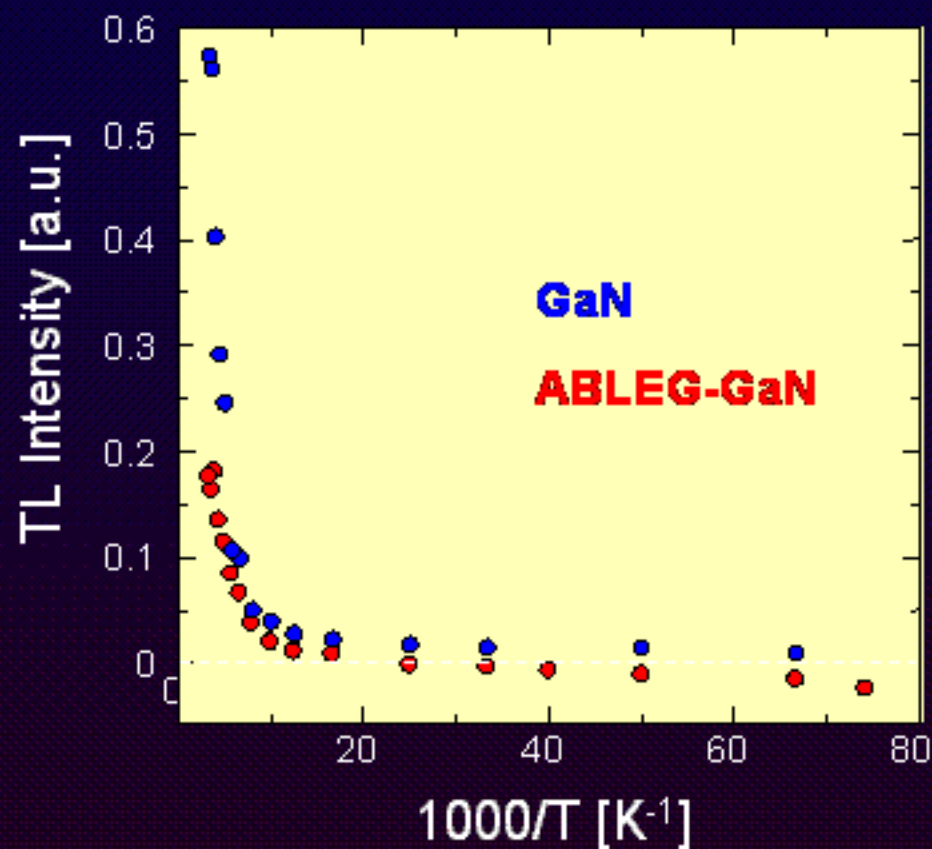
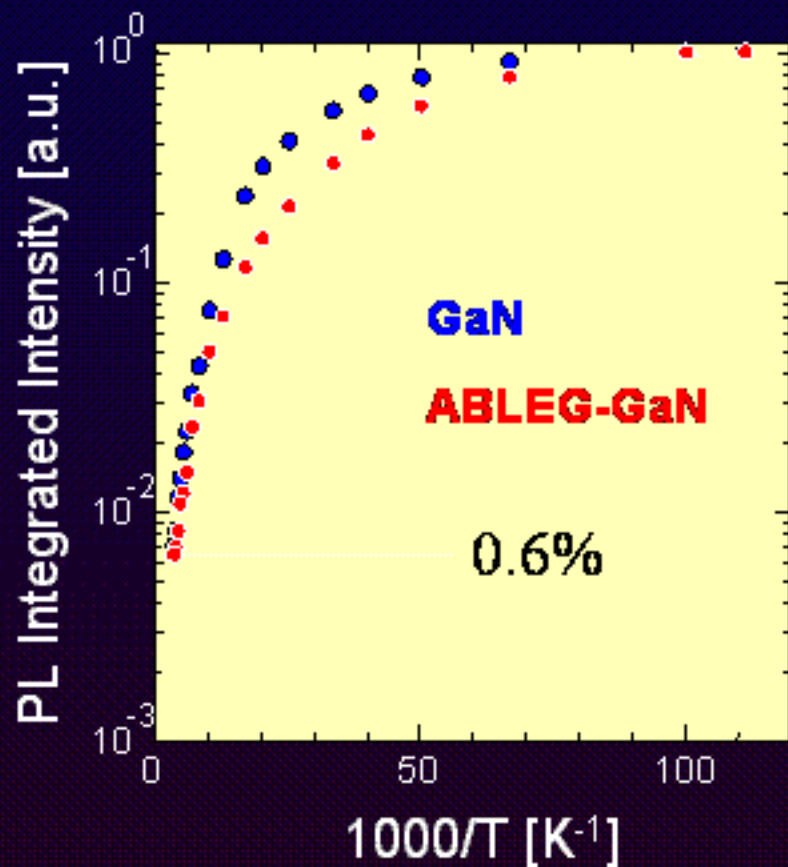


TD act as NRC, but it is not effective to the emission efficiency



Temperature dependence of CW-MTL

© Koichi Okamoto





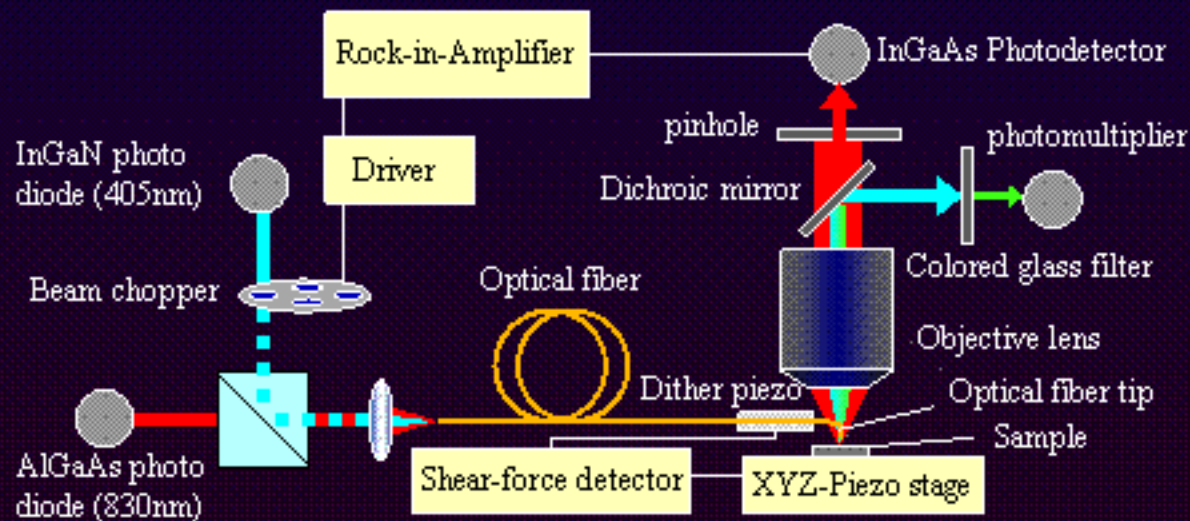
Near-field scanning optical microscope TL © Koichi Okamoto

(NSOM-TL)

3rd order Nonlinear Spectroscopy

+ Near-field scanning optical microscopy

K. Okamoto, et al, *Appl. Phys. Lett.*, 87, 161104 (2005).

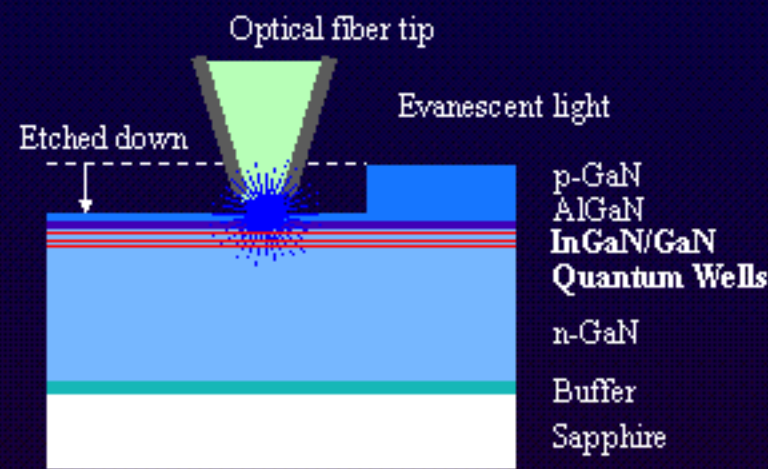


Experimental setup of Near-field scanning optical microscope TL (NSOM-TL)



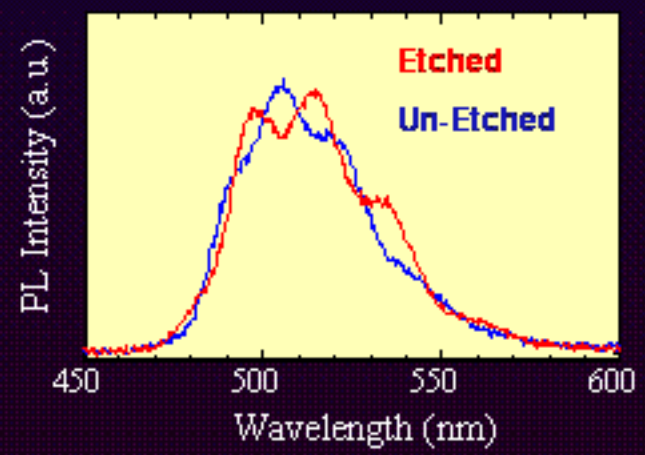
Sample Structure and Properties

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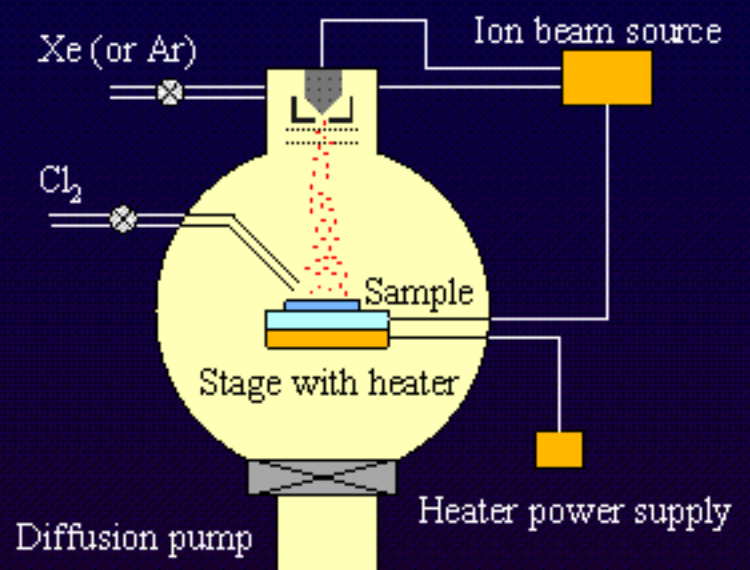


Purchased by AXT Inc.

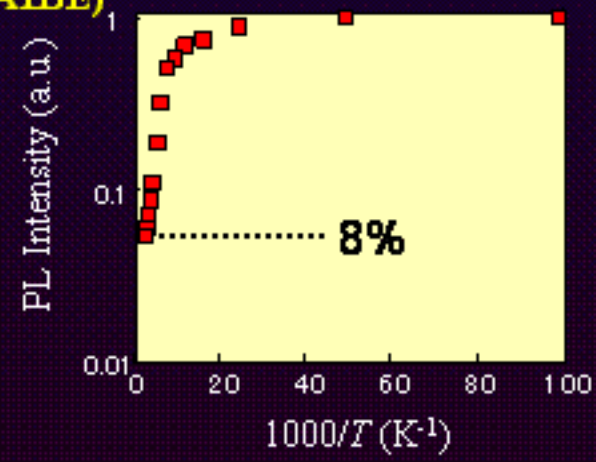
Sample structure of InGaN/GaN 3QWs



Photoluminescence (PL) spectra



Chemically assisted ion beam etching (CAIBE)



Temperature dependence of PL intensity



Numerical Calculation of TL time-profile

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$$\delta n(r) = \delta n_0 \exp(-r^2/w_0^2)$$

w_0 : pump beam width

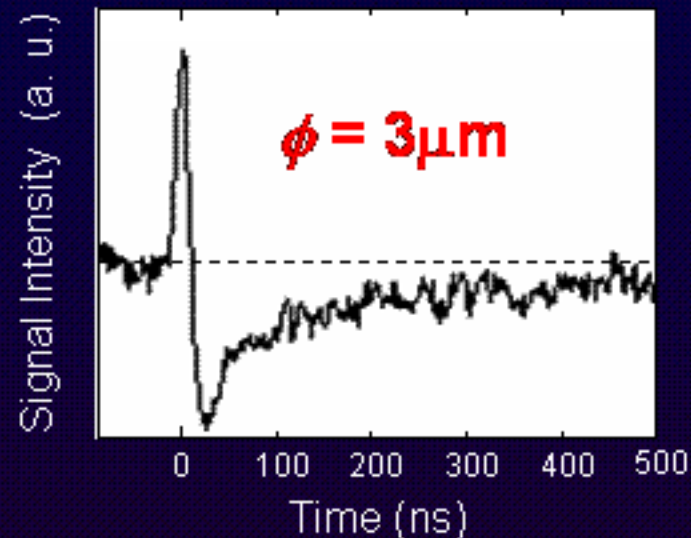
n_0 : Refractive index change at the beam center

$$\delta n(r,t) = \left(\frac{\partial n}{\partial N}\right) \delta N(r,t) + \left(\frac{\partial n}{\partial T}\right) \delta T(r,t) \quad \left(\frac{\partial n}{\partial N}\right) < 0, \quad \left(\frac{\partial n}{\partial T}\right) > 0$$

The carrier dynamics can be written by

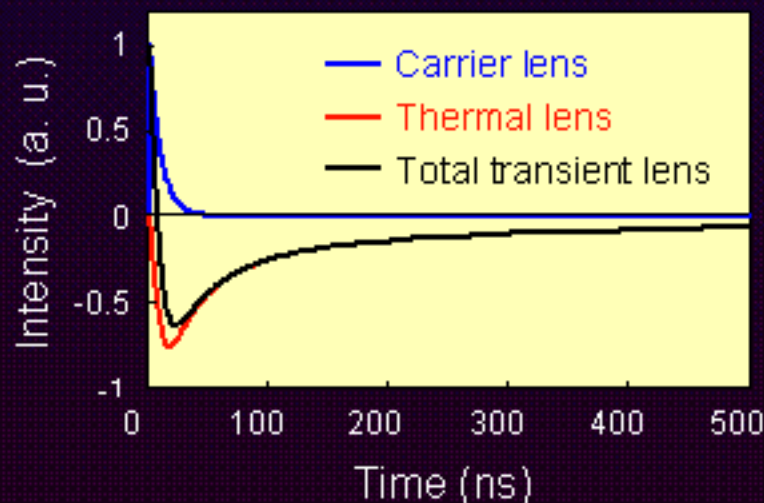
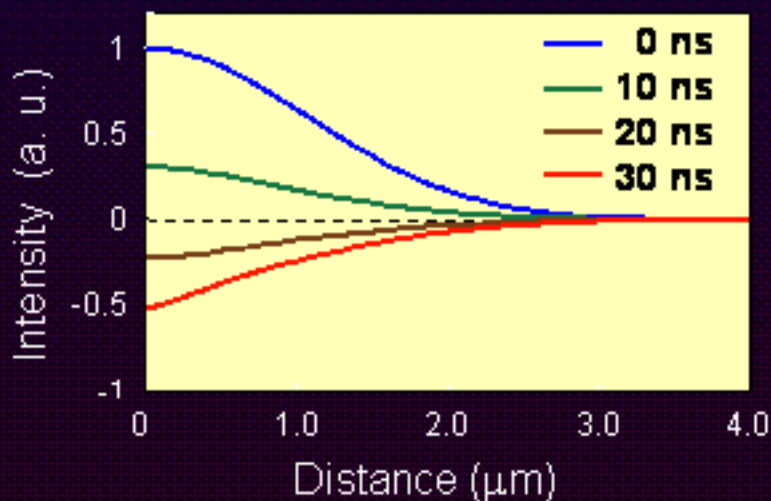
$$\frac{\partial \delta N(r,t)}{\partial t} = D \frac{\partial^2 \delta N(r,t)}{\partial r^2} - \left(\frac{1}{\tau_{\text{rad}}} + \frac{1}{\tau_{\text{non-rad}}}\right) \delta N(r,t) \quad \text{Carrier density}$$

$$\frac{\partial \delta T(r,t)}{\partial t} = \frac{1}{\tau_{\text{non-rad}}} \frac{Q \delta N(r,t)}{\rho C_p} + D_{\text{th}} \frac{\partial^2 \delta T(r,t)}{\partial r^2} \quad \text{Temperature}$$



Numerical Calculation

r : 100 pixel, t : 500 pixel, $D=0.5 \text{ cm}^2\text{s}^{-1}$, $D_{\text{th}}=1.0 \text{ cm}^2\text{s}^{-1}$
 $\tau_{\text{PL}} = 20 \text{ ns}$ ($1/\tau_{\text{PL}} = 1/\tau_{\text{rad}} + 1/\tau_{\text{non-rad}}$)

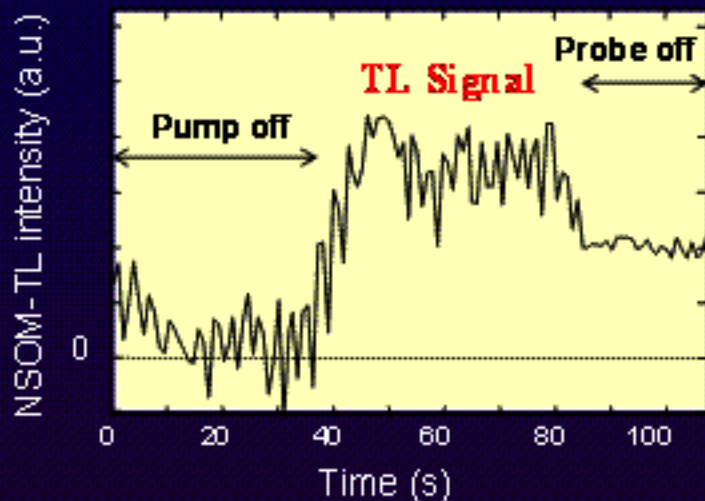




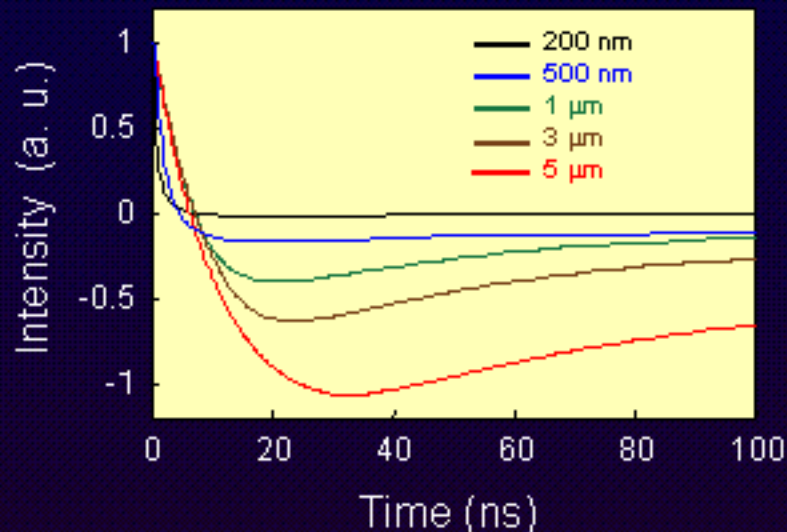
Results and Analysis of NSOM-TL

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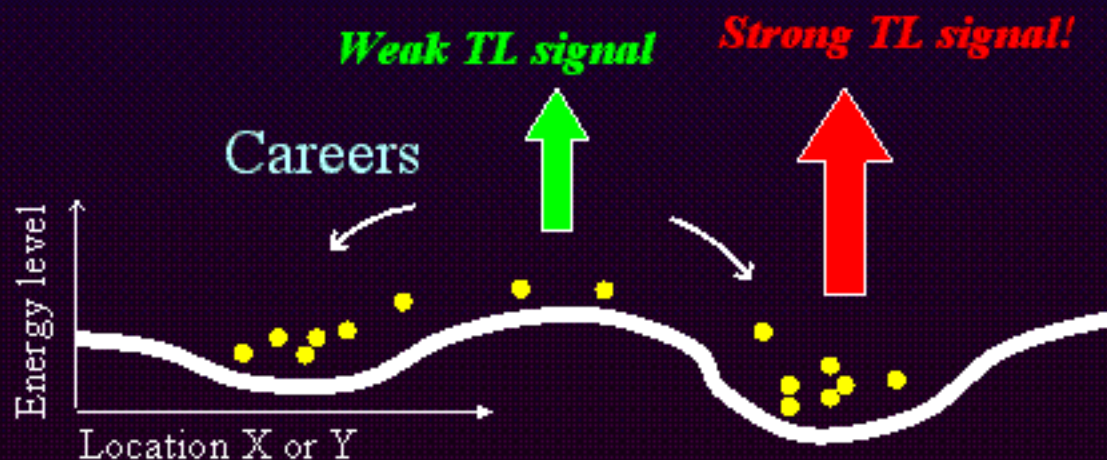
Obtained signal $\phi = 200$ nm



Numerical Calculation



Spot size = 200nm; main component of the TL signal is **Carrier density lens**

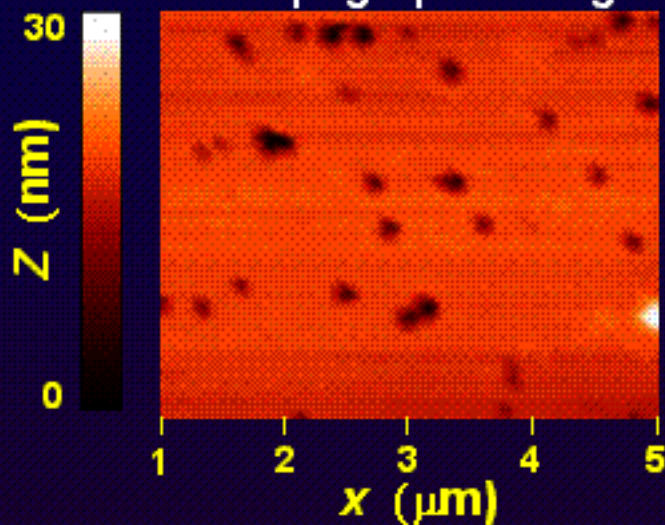




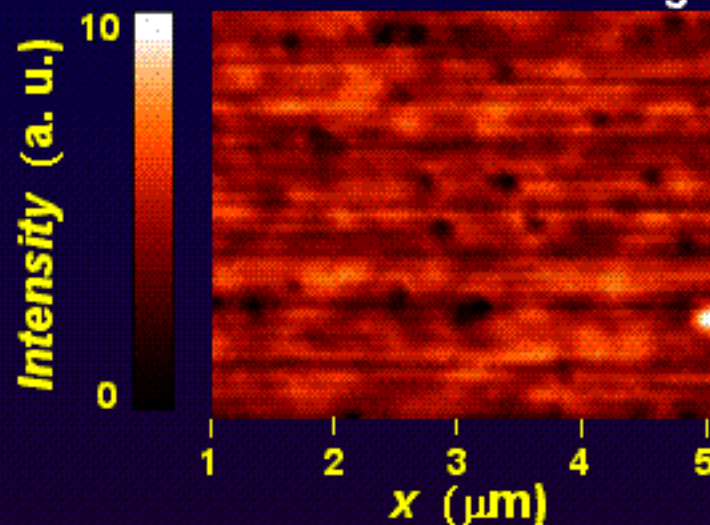
NSOM Images

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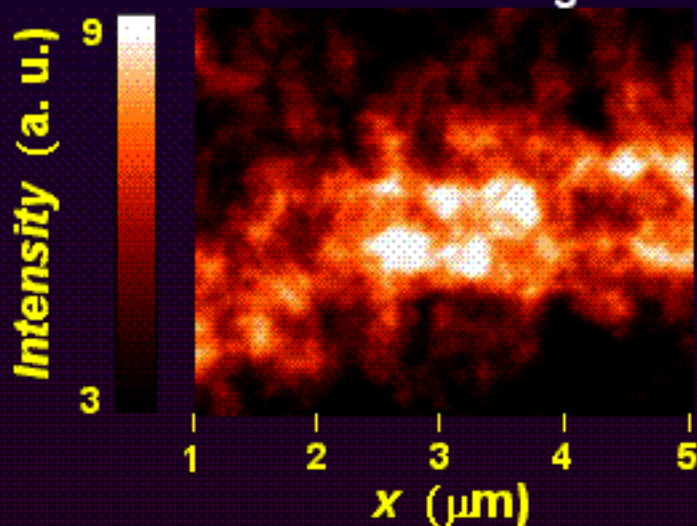
SFM Topographic Image



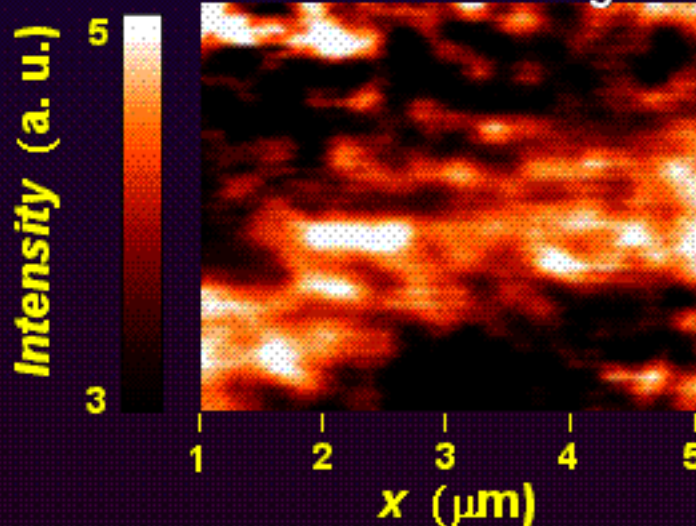
NSOM Reflection Image



NSOM-PL Image



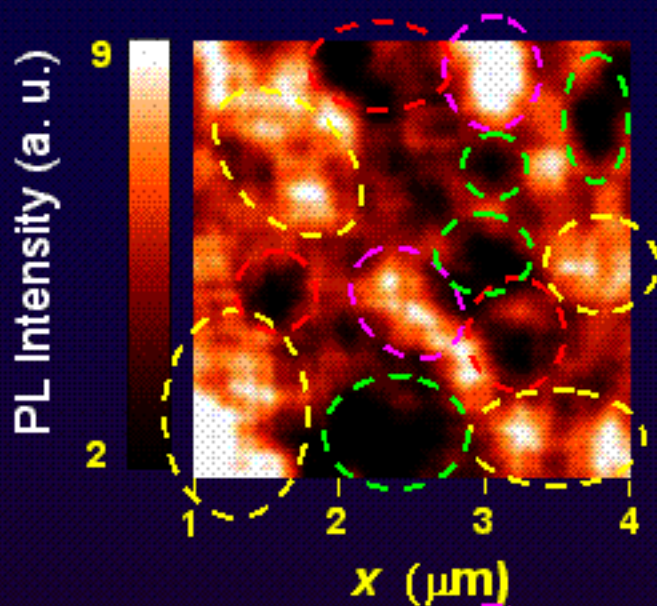
NSOM-TL image





NSOM-PL and -TL Images

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Area A

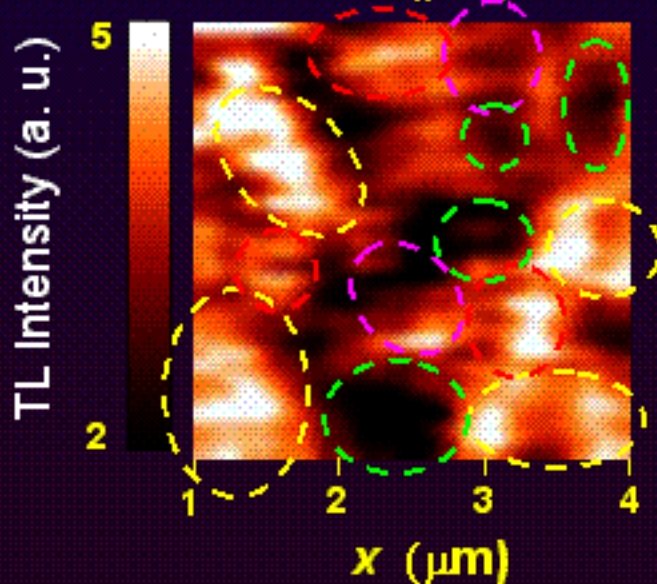
PL signal Strong

TL signal Strong

Area B

PL signal Weak

TL signal Weak



Area C

PL signal Weak

TL signal Strong

Area D

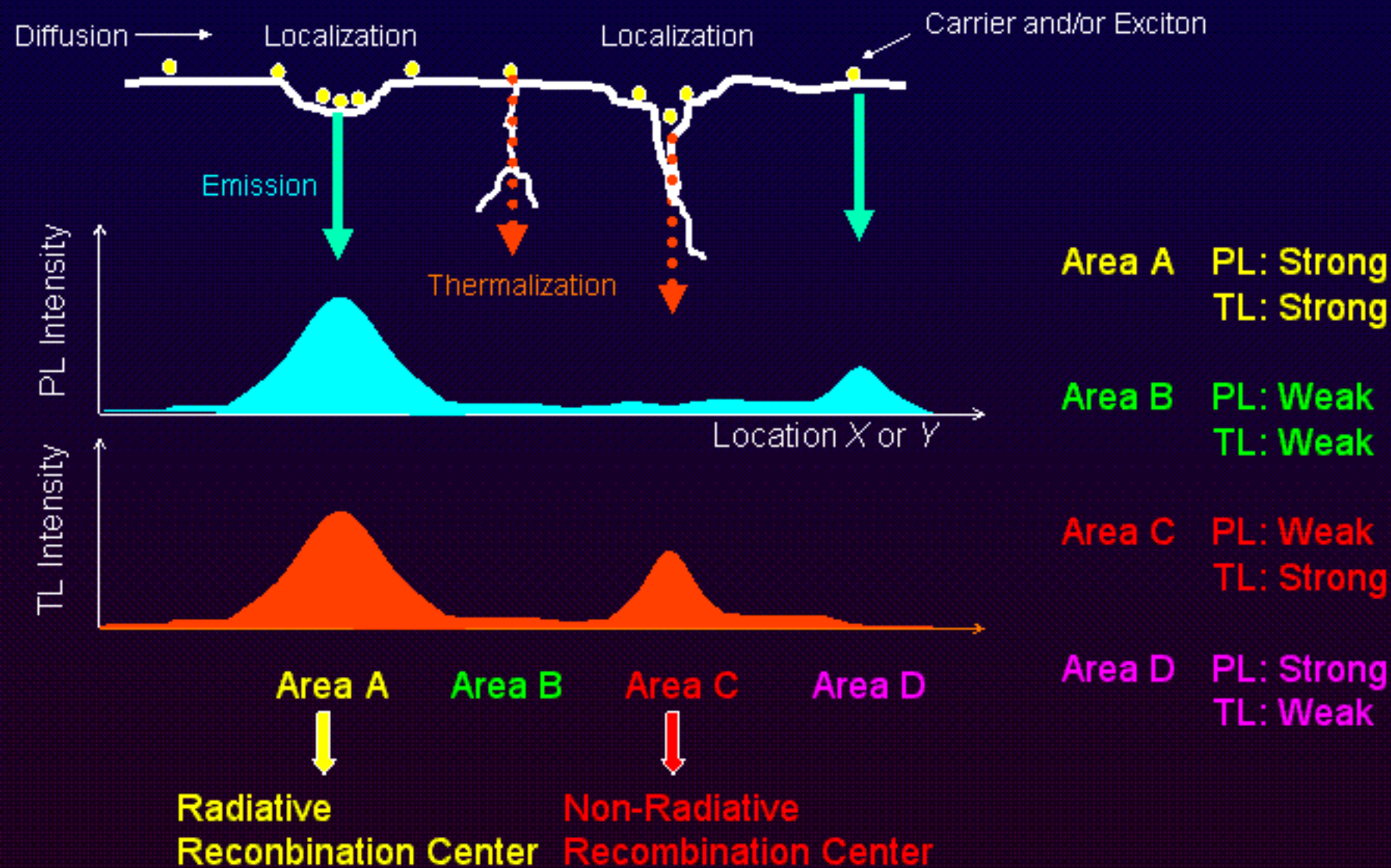
PL signal Strong

TL signal Weak



Carrier Dynamics

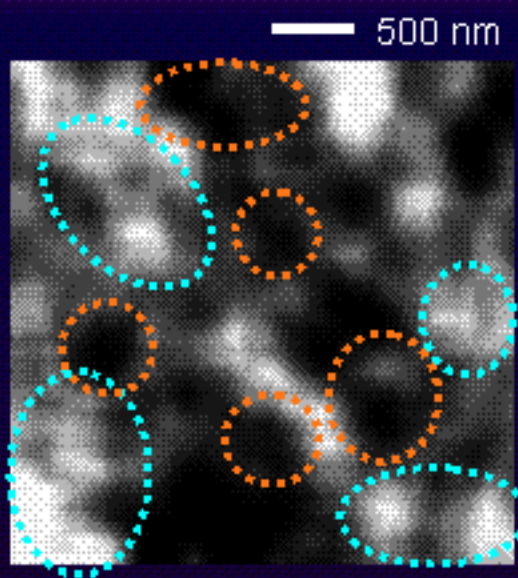
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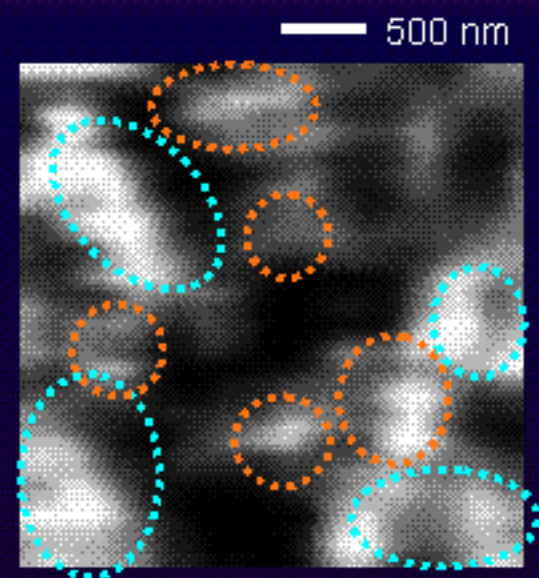


Diffusion and Recombination

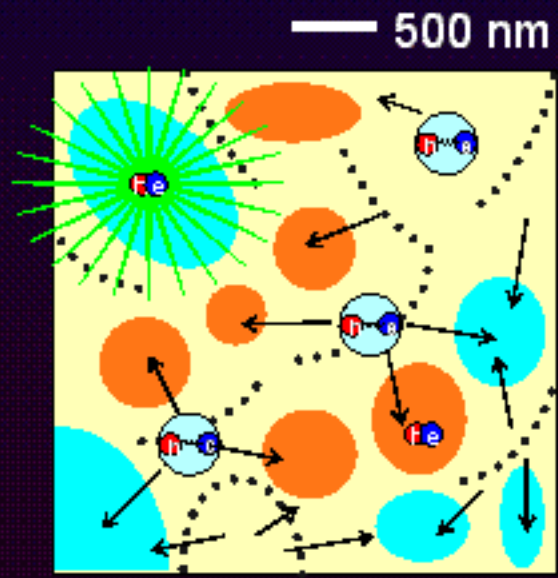
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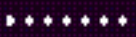


PL Intensity (arb. units)



TL Intensity (arb. units)



-  Radiative recombination centers
-  Non-radiative recombination centers
-  Carriers and/or excitons transfer
-  High potential energy region



Summary

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Temporal and spatial-resolved nonlinear spectroscopies are very useful and powerful tools to characterize nonradiative processes (carrier diffusion, localization, recombination, and thermalization, etc) in GaN and InGaN/GaN.

Such information are very important to understand the emission mechanism and dynamics in order to develop the device performances and light emission efficiencies of optical materials.



For example, based on such measurements, we have invented the novel technique to enhance the emission efficiencies of InGaN/GaN