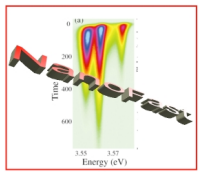
Spettroscopia ottica avanzata di nanostrutture per dispositivi optoelettronici nell'UV

Dipartimento di Fisica ed Astronomia



Partecipanti:

2 strutturati Fis (Vinattieri,Gurioli)

+1 strutturato Energ (Bogani)

1 assegnista (Cavigli)

1PhD (Gabrieli, XXV ciclo)

1PhD (Stokker-Cheregi, XXI ciclo)

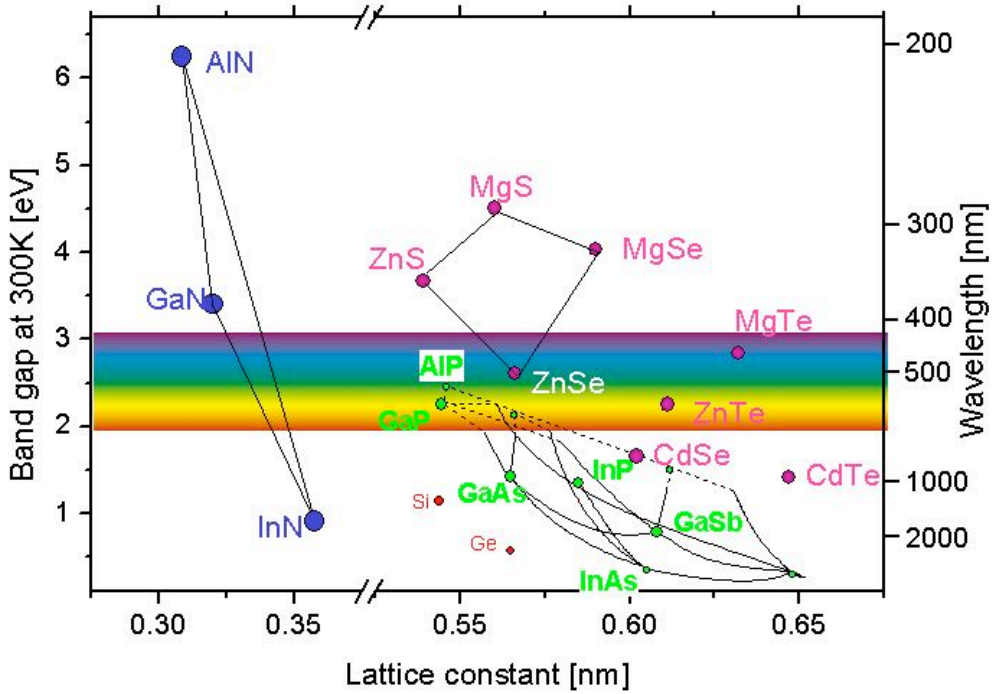
Linee ricerca

High spatial and temporal resolution optical spectroscopy

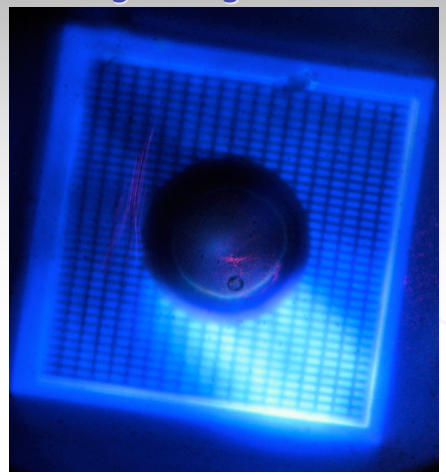
- Nitrides heterostructures (VI programma EU-Clermont2, collaborazioni CHREA-Valbonne, EPFL-Losanna)
- Semiconductor NPs (TiO_2) for innovative photovoltaic energy conversion (DE, PoliTO, CNR & DipFisPI: Richiesta PAR-FAS Regione Toscana)
- GaN optoelectronic devices for future applications (DE, DipIng-PD, CNR-Pr: PRIN)

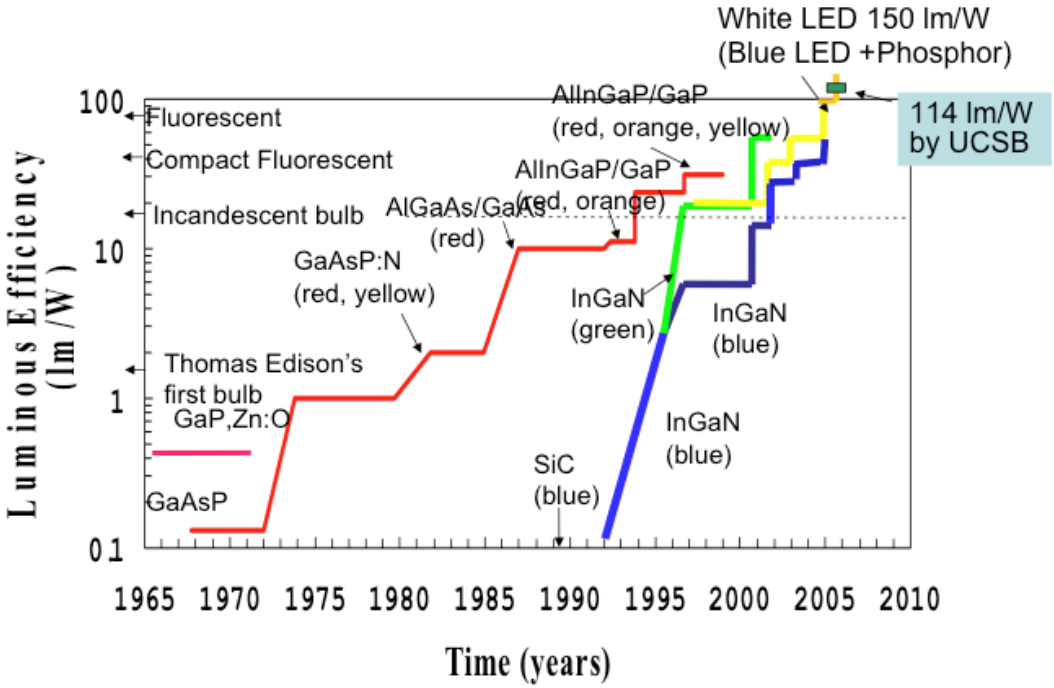
High resolution optical spectroscopy of Nitride Heterostructures

- GaN bulk μCavities for room T polariton lasers
- Exciton dynamics in high quality GaN/AlGaN QWs



Nitrides nicely fit the UV region: GaN and InN, GaN,AlN ternary-quaternary alloys for high efficiency solid state lightening devices





Dipartimento di Fisica ed Astronomia

Advantages:

High tunability NUV-NIR

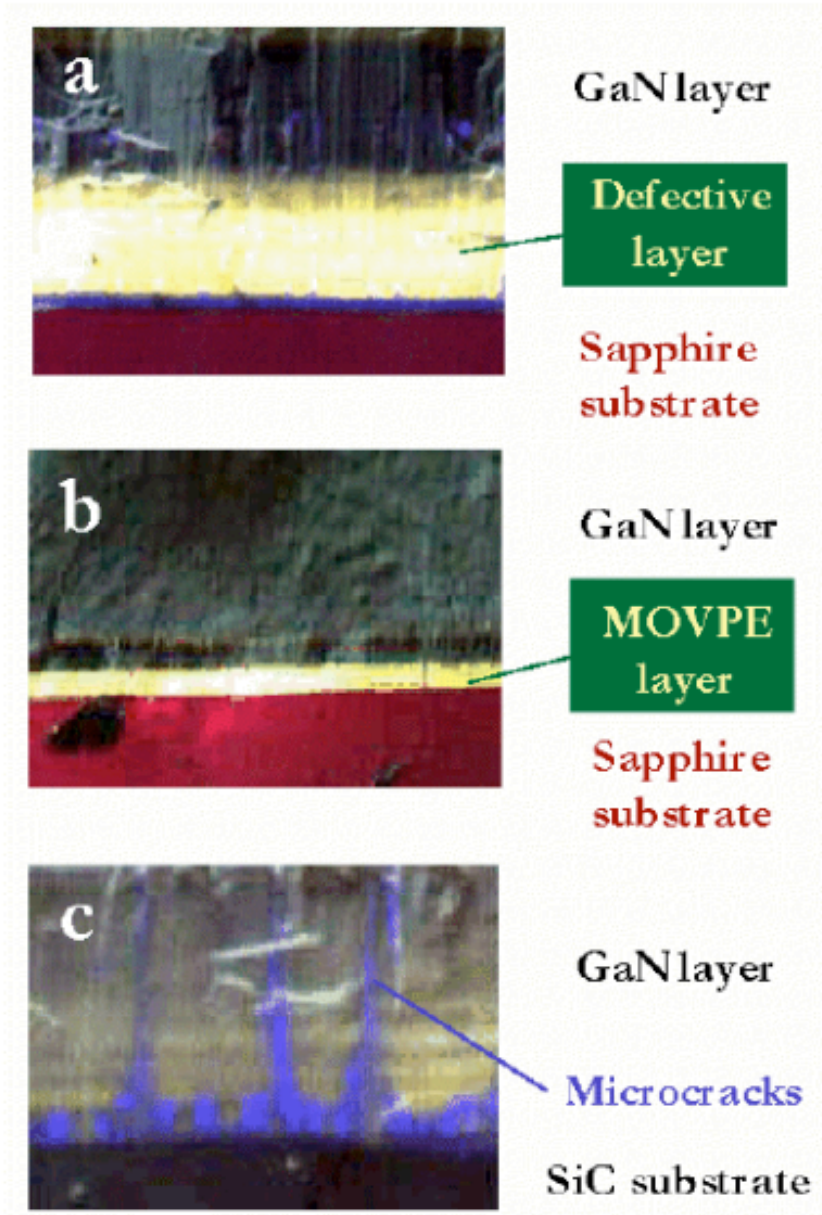
High radiative efficiency

Low maintenance cost

But: poor material quality
- Piezoelectricity
- High content of defects
- Poor control of doping

Dislocation density GaAs on Si:
10^4-10^5 cm^{-2}

Dislocation density GaN on Sapphire:
10^{10} cm^{-2} now 10^7 cm^{-2}



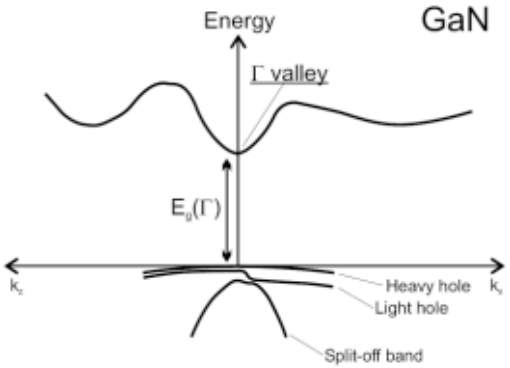
Reducing the defects density

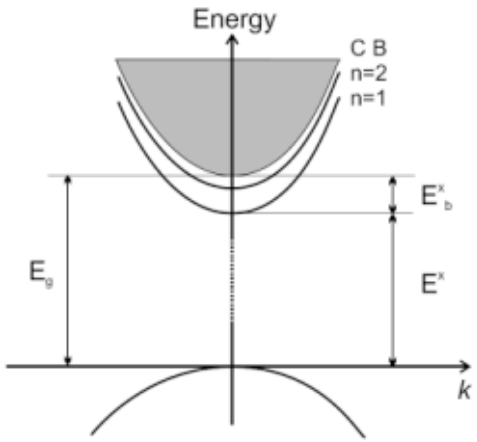
↓

Exciton dynamics

GaN	ZnO	GaAs	CdSe
3.5 eV 25 meV	3.4 eV 60meV	1.5 eV 4meV	3.5 eV 20 meV

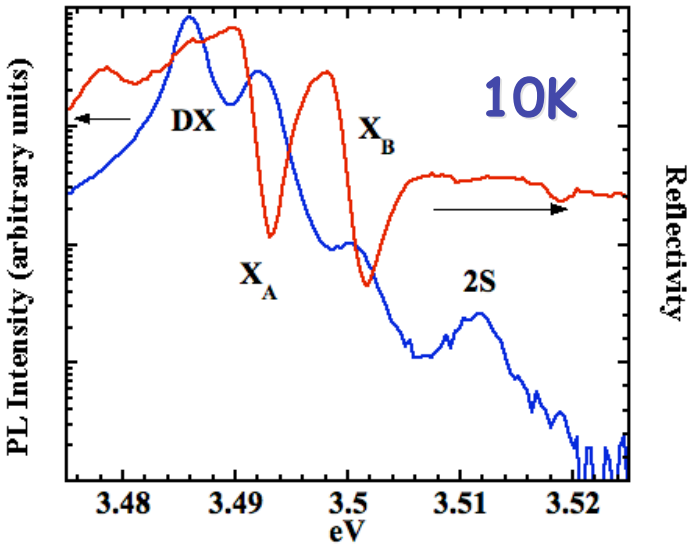
Excitons @ RT: new devices

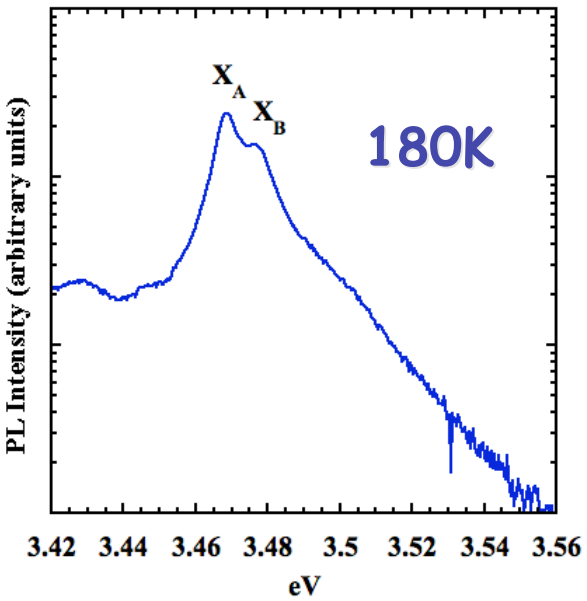




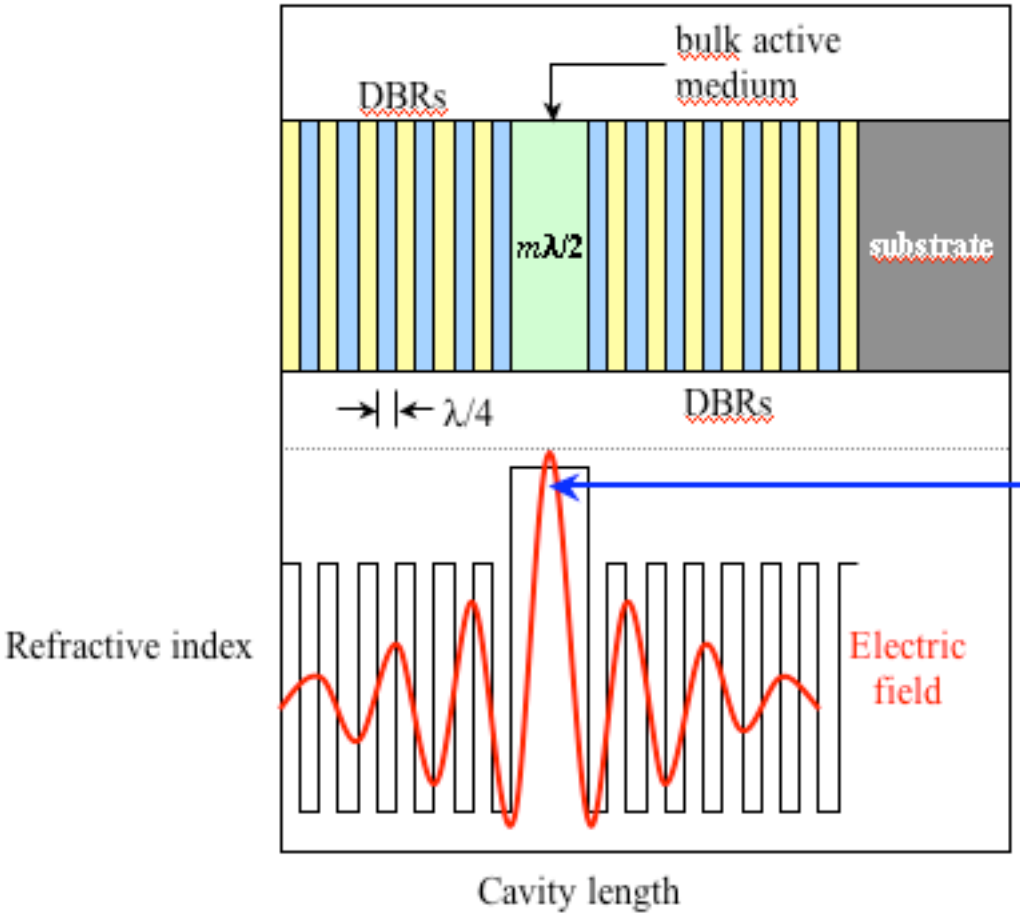
$$E^x_n = E_g - E^x_b \frac{1}{n^2} \qquad E^x_b = 13.6eV \frac{\mu}{\varepsilon^2}$$

for bulk GaN: E^x_b ~ 25 meV

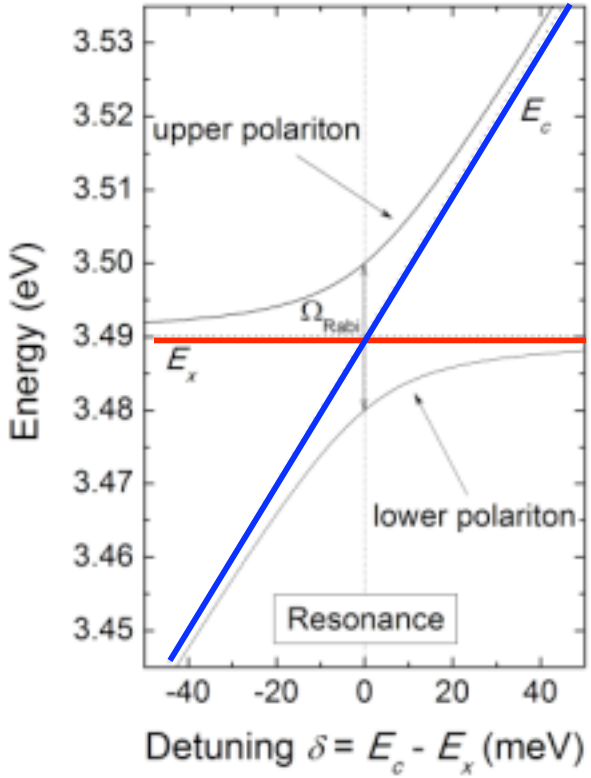


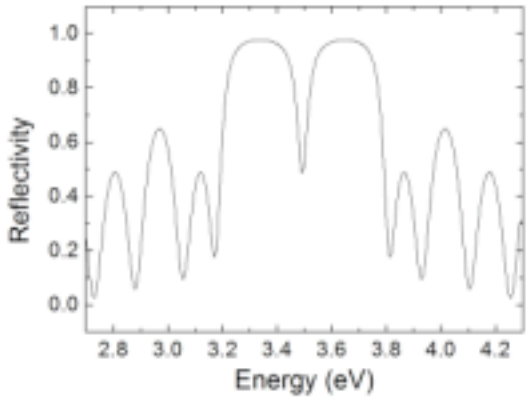


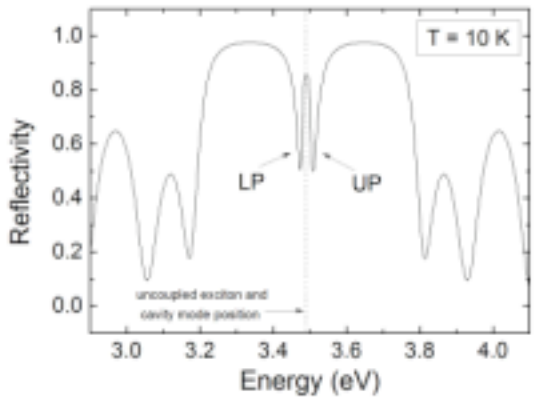
Microcavities: Strong coupling at RT



Strong amplification of the electric field in the active region





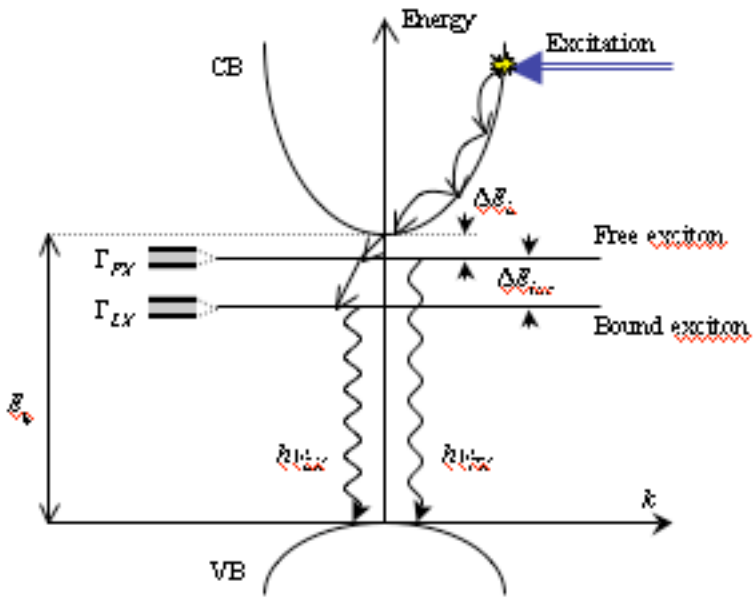


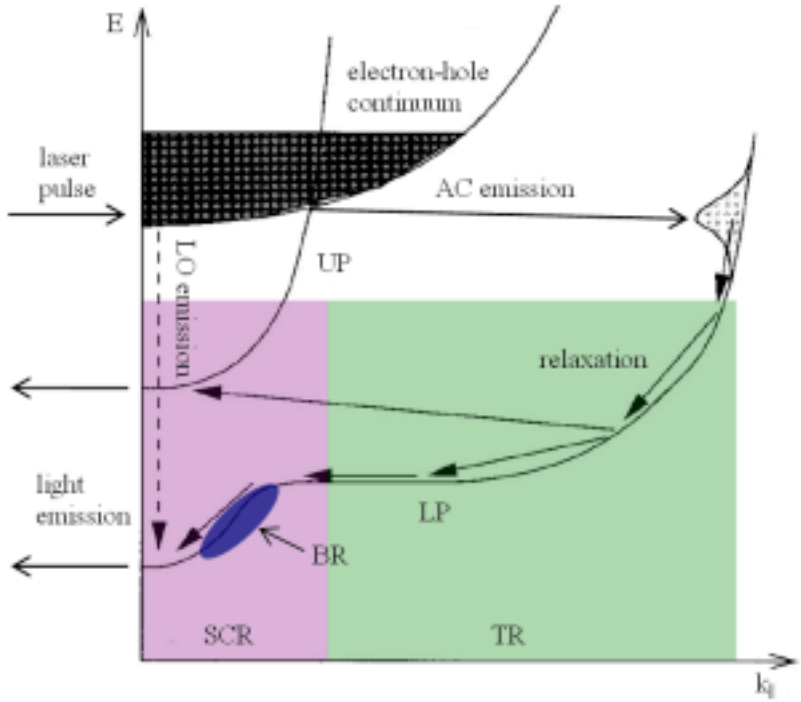
Rabi splitting in GaN-based μC 50-60 meV : strong coupling regime @ RT.

Polariton=Boson

Polariton effective mass $\approx 10^{-4}$ exciton mass $\approx 10^{-9}$ Rb mass

$\longrightarrow$ high T_c





bulk exciton photoluminescence

strong coupling polariton photoluminescence

Bose-Einstein Exciton Condensate & Polariton Lasing

(J.Kasprzak et al.: Nature 443, 409 (2006), S.Christopoulos et. al.:PRL 98, 126405 (2007), G.Christmann et al.: APL 93, 051102 (2008))

Our research focussed on:

- Investigation of SC regime
- Investigation of relaxation bottleneck and thermalization (Exciton-polariton dynamics)

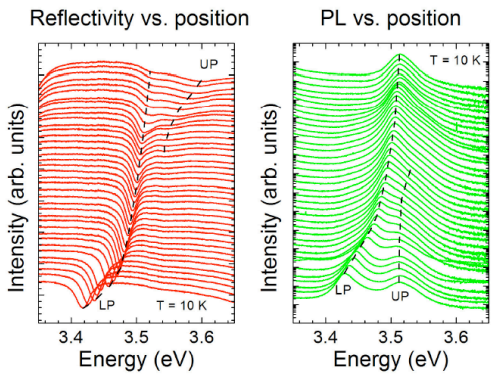
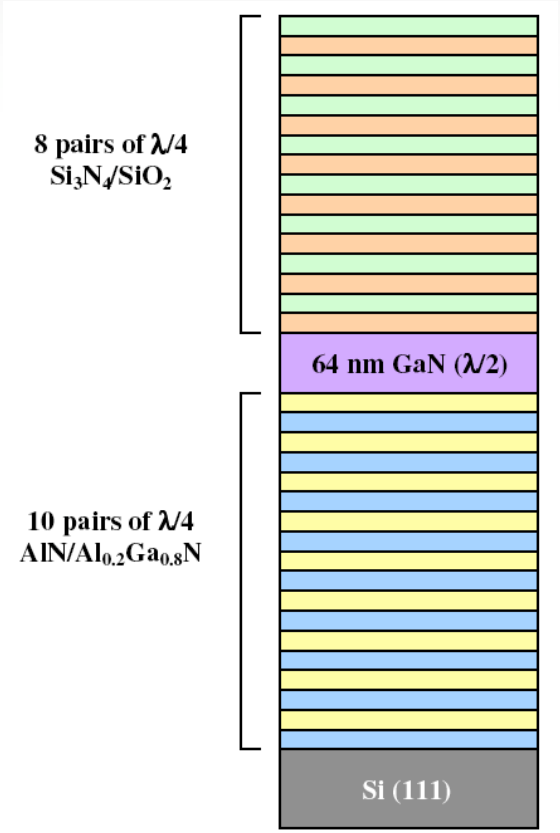


Figure 5.18. Simultaneous reflectivity (left) and TI PL (right) spectra at T = 10 K of the hybrid microcavity (sample 808) as a function of excitation position. Spectra are shifted in intensity for clarity. The dashed lines are guides to the eye.



Gradiente spessore: possibilita' di tuning muovendosi sul campione

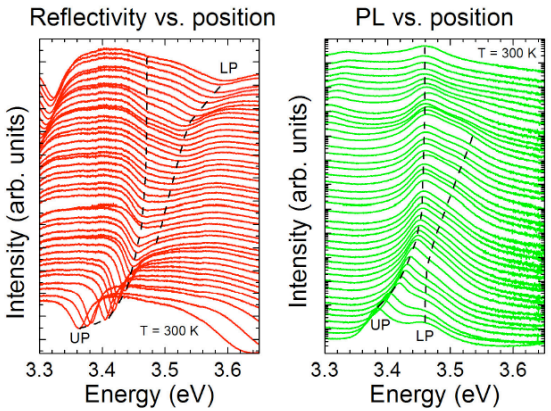


Figure 5.20. Simultaneous reflectivity (left) and TI PL (right) spectra at T = 300 K of the hybrid microcavity (sample 808) as a function of excitation position. Spectra are shifted in intensity for clarity. The dashed lines are guides to the eye.

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Superlattices and Microstructures 41 (2007) 376–380
Superlattices and Microstructures
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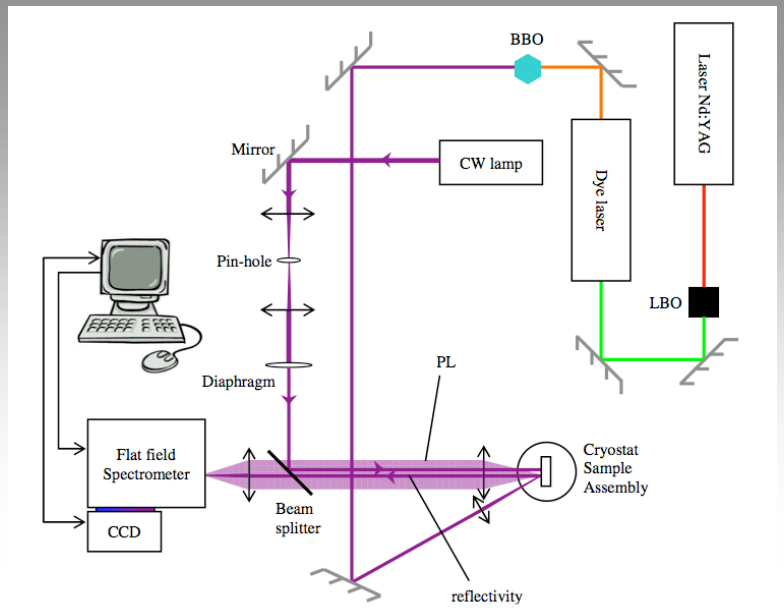
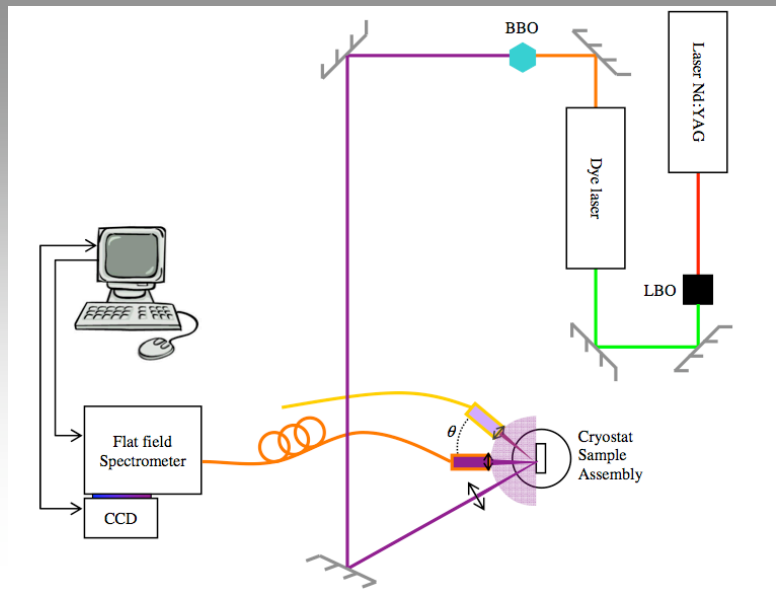
Polariton thermalization in GaN microcavities in the strong light–matter coupling regime

F. Stokker-Cheregi[a,*], M. Zamfirescu[a], A. Vinattieri[a], M. Gurioli[a], I. Sellers[b], F. Semond[b], M. Leroux[b], J. Massies[b]

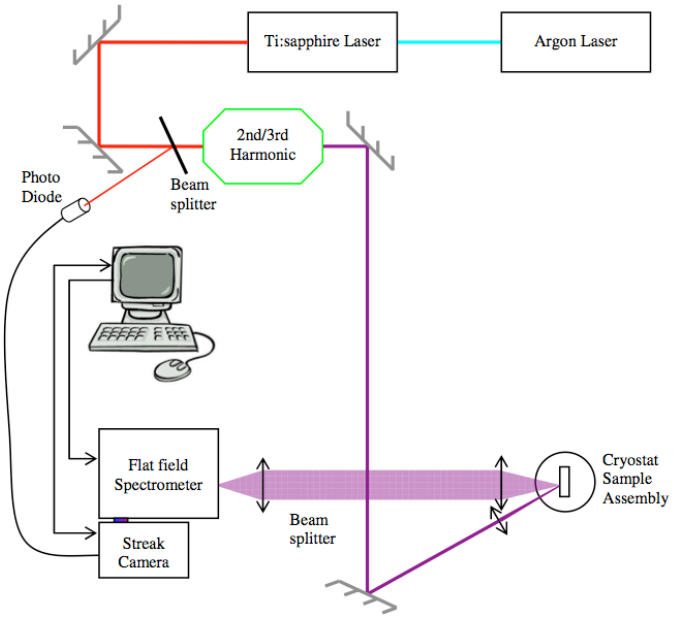
[a] LENS and Dipartimento di Fisica, Universita di Firenze, 50019 Sesto Fiorentino, Italy
[b] CRHEA-CNRS, Rue Bernard Gregory, Valbonne 06560, France



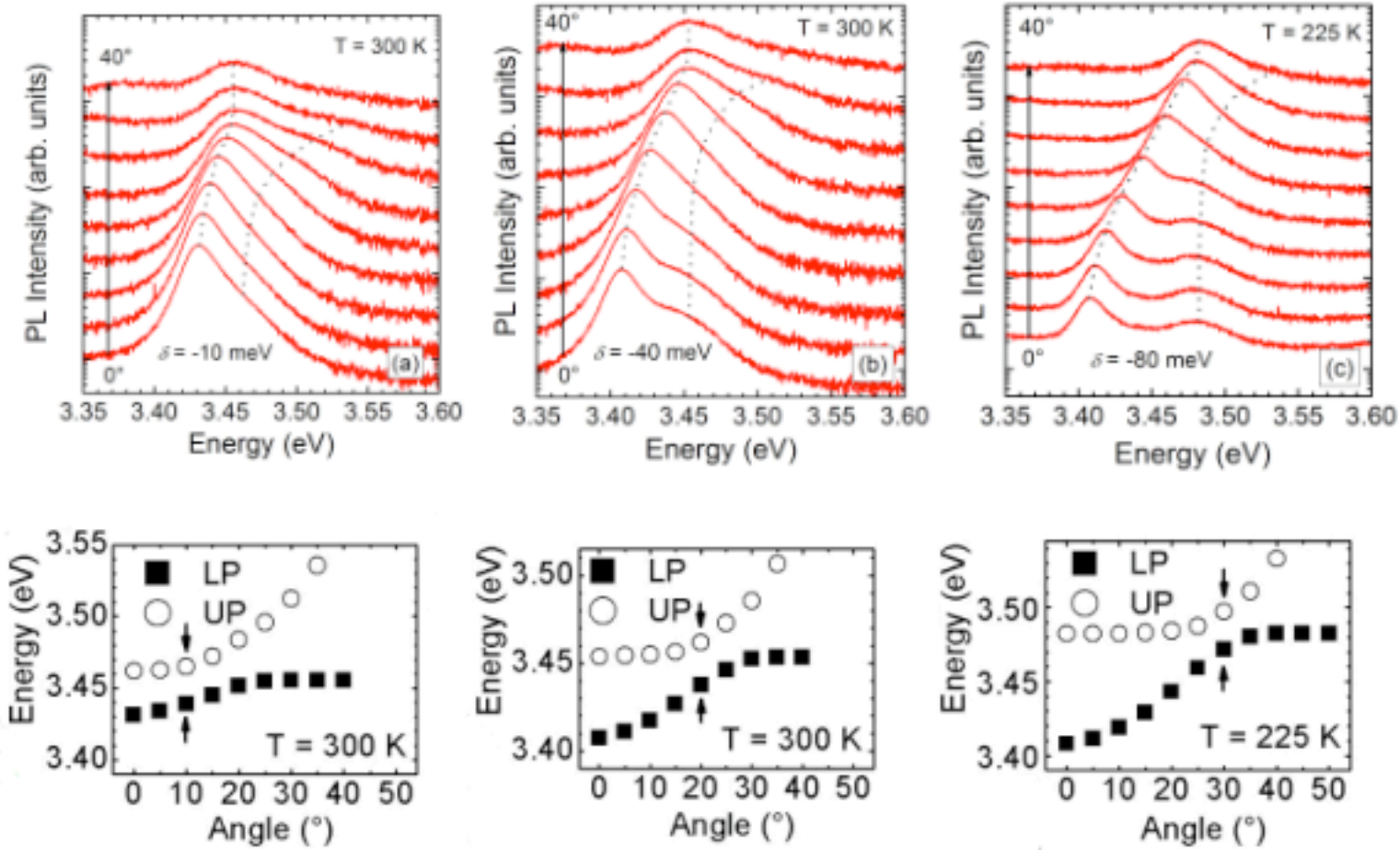
Position-angle resolved Reflectivity & PL



ps Time-resolved PL



Strong Coupling Regime @ RT



Exp Rabi splitting: 30 meV → SC @ RT

PHYSICAL REVIEW B 74, 193308 (2006)

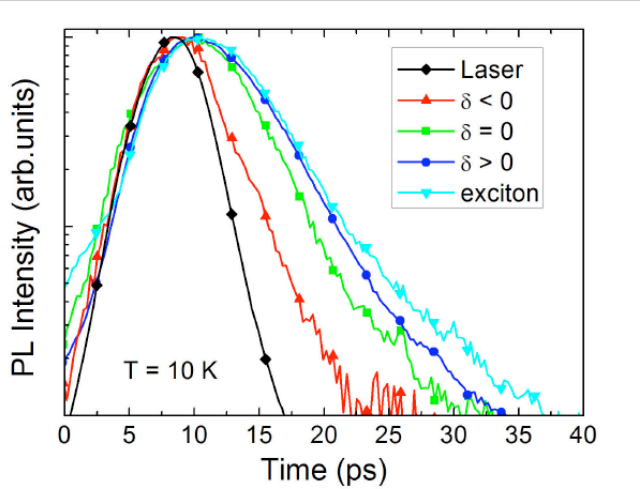
Polariton emission and reflectivity in GaN microcavities as a function of angle and temperature

I. R. Sellers,* F Semond, M. Leroux, and J. Massies
CRHEA-CNRS, Rue Bernard Gregory, Valbonne 06560, France

M. Zamfirescu, F. Stokker-Cheregi, M. Gurioli, A. Vinattieri, and M. Colocci
LENS, Dipartimento di Fisica, Universitia di Firenze, 50019 Sesto Florentino, Italy

A. Tahraoui and A. A. Khalifa
Department of Physics & Astronomy, Hicks Building, Hounsfield Road, University of Sheffield, S3 7RH, United Kingdom

Modification of the Radiative Recombination Rate in presence of SC



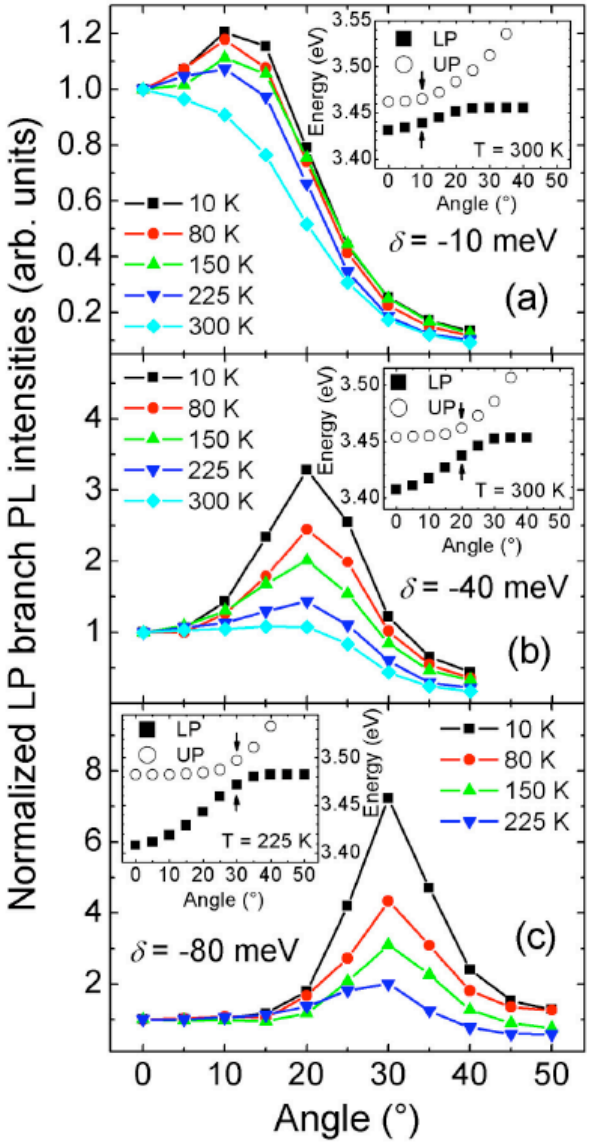
Moreover, the LP and UP populations are thermalized at T = 300 K,despite the very short lifetimes. This denotes very efficient energy relaxation processes in the LP, as well as an efficient population of the UP states through acoustic phonon interactions in our bulk GaN microcavity.

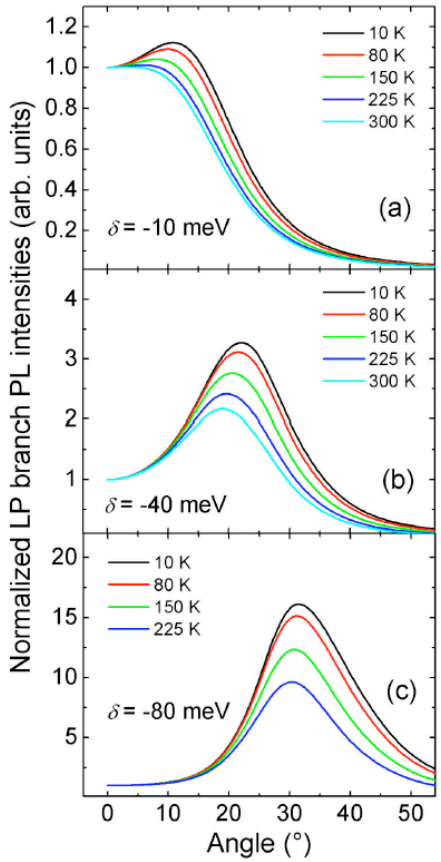
APPLIED PHYSICS LETTERS **92**, 042119 (2008)

Polariton relaxation bottleneck and its thermal suppression in bulk GaN microcavities

F. Stokker-Cheregi,[1,a)] A. Vinattieri,[1] F. Semond,[2] M. Leroux,[2] I. R. Sellers,[2] J. Massies,[2] D. Solnyshkov,[3] G. Malpuech,[3] M. Colocci,[1] and M. Gurioli[1]
[1]*LENS and Department of Physics, University of Florence, I-50019 Sesto Fiorentino, Italy*
[2]*CRHEA-CNRS, Rue Bernard Gregory, Valbonne 06560, France*
[3]*LASMEA, CNRS, Université Blaise Pascal, 24, av des Landais, 63177 Aubière, France*

Suppression of Relaxation Bottleneck at RT

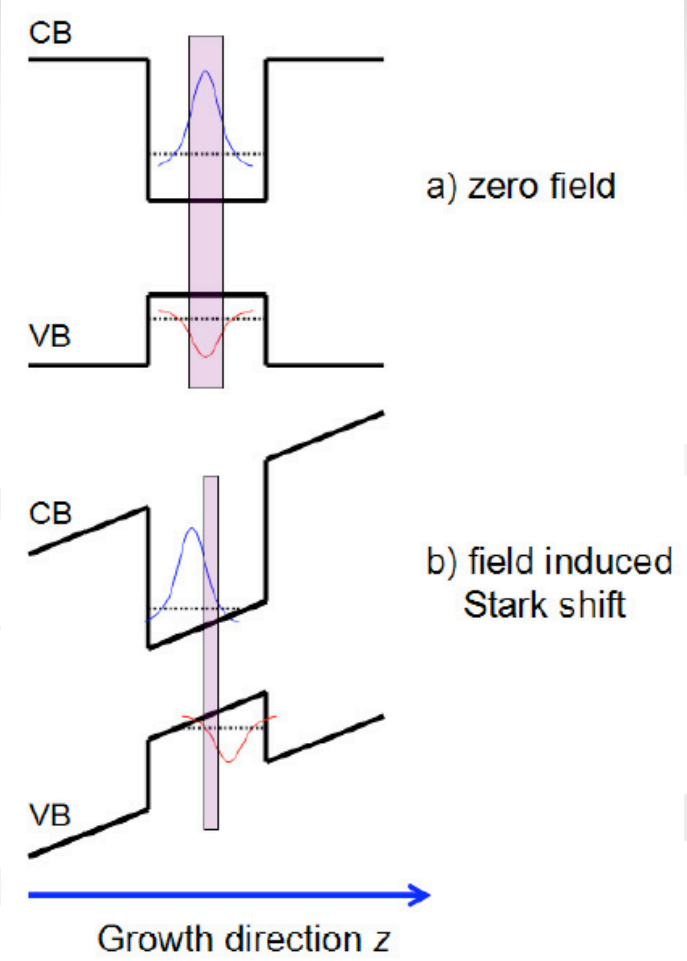




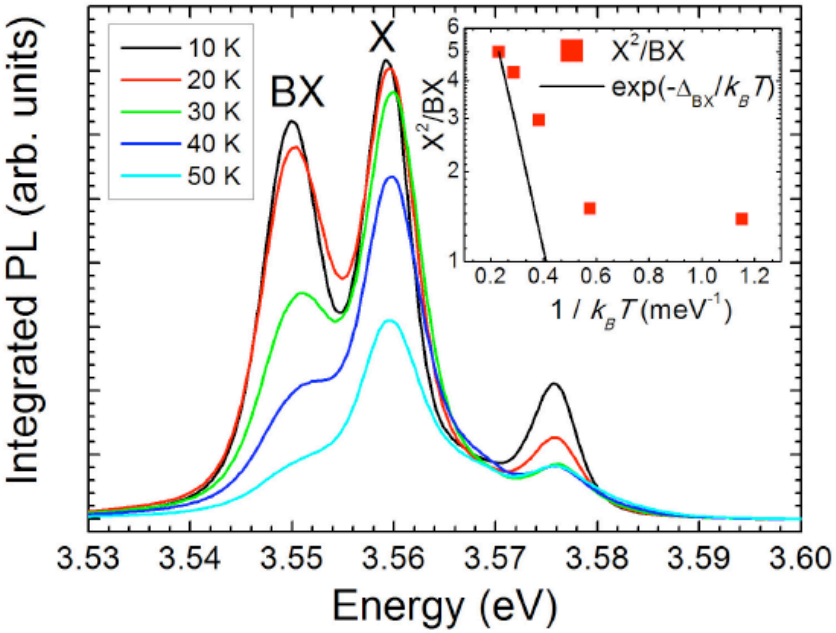
Exciton Dynamics in State-of-the-art GaN/AlGaN SQWs

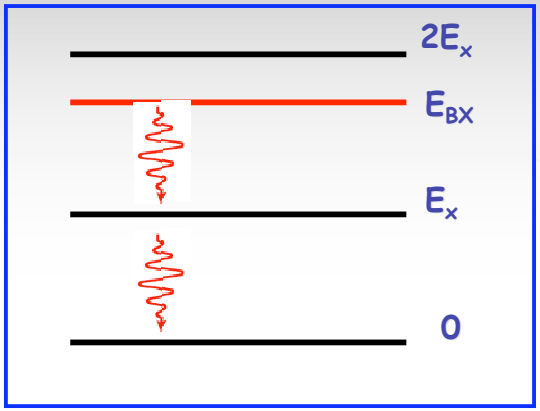
Strain & Internal Electric Field (piezo+ spontaneous polarization up to 1MV/cm) strongly affect the optical properties:

- Reduction of the exciton rad rate (Quantum Confined Stark Effect)
- Inhomogenous broadening
- Exciton localization

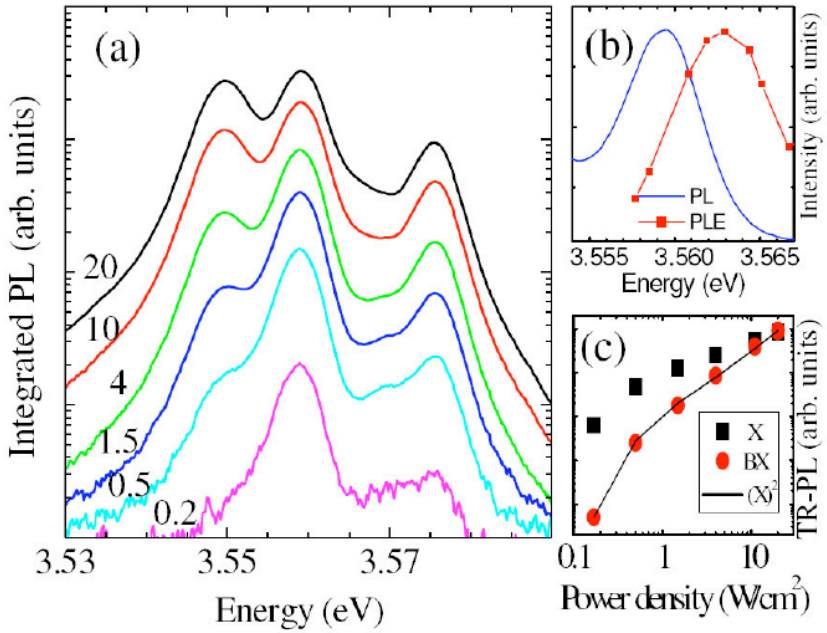


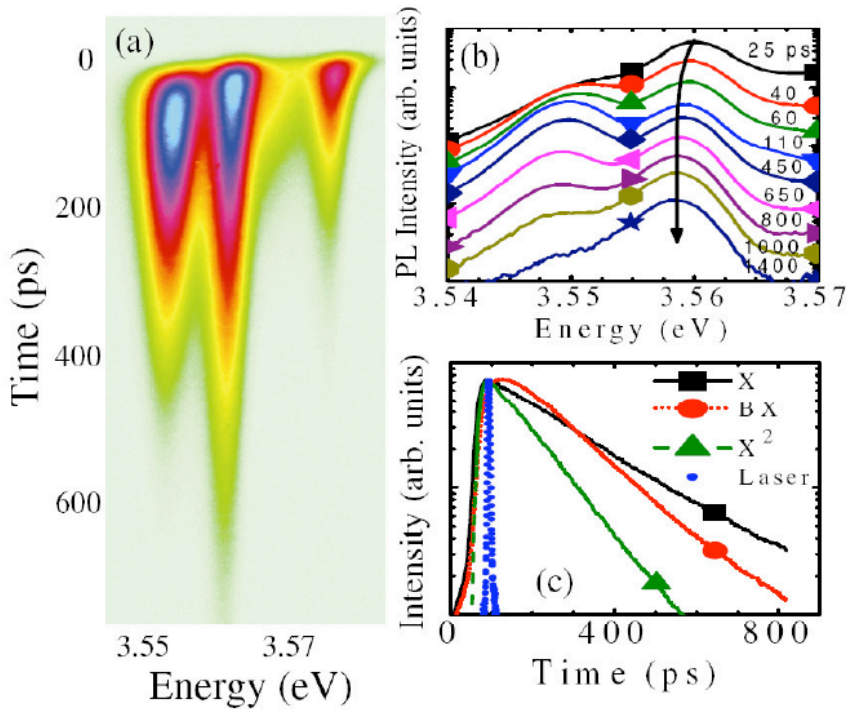
Biexciton Recombination Kinetics

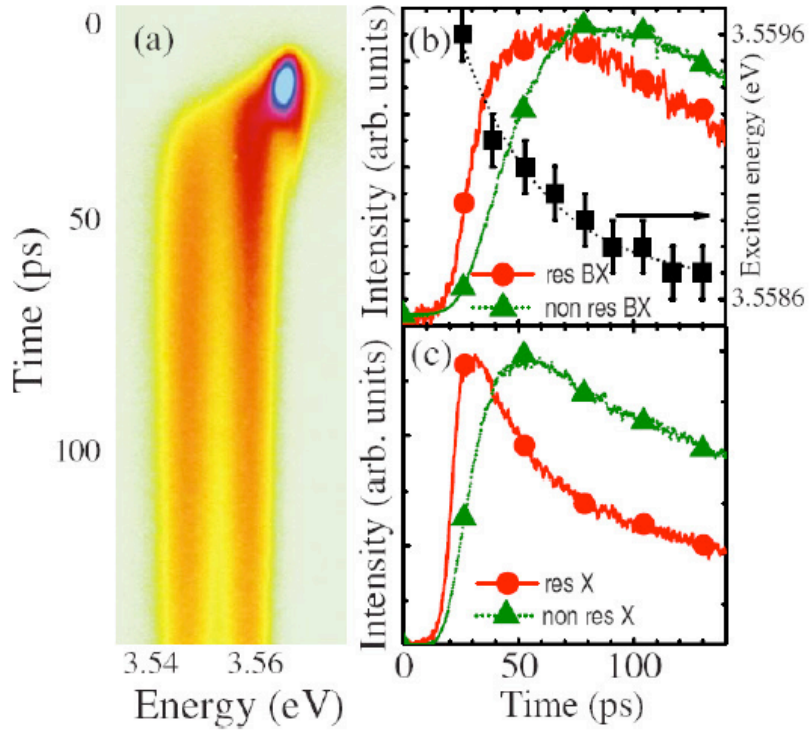




$$\frac{n_X^2}{n_{BX}} \propto \exp(-\Delta_{BX}/k_B T)$$







Non-resonant excitation

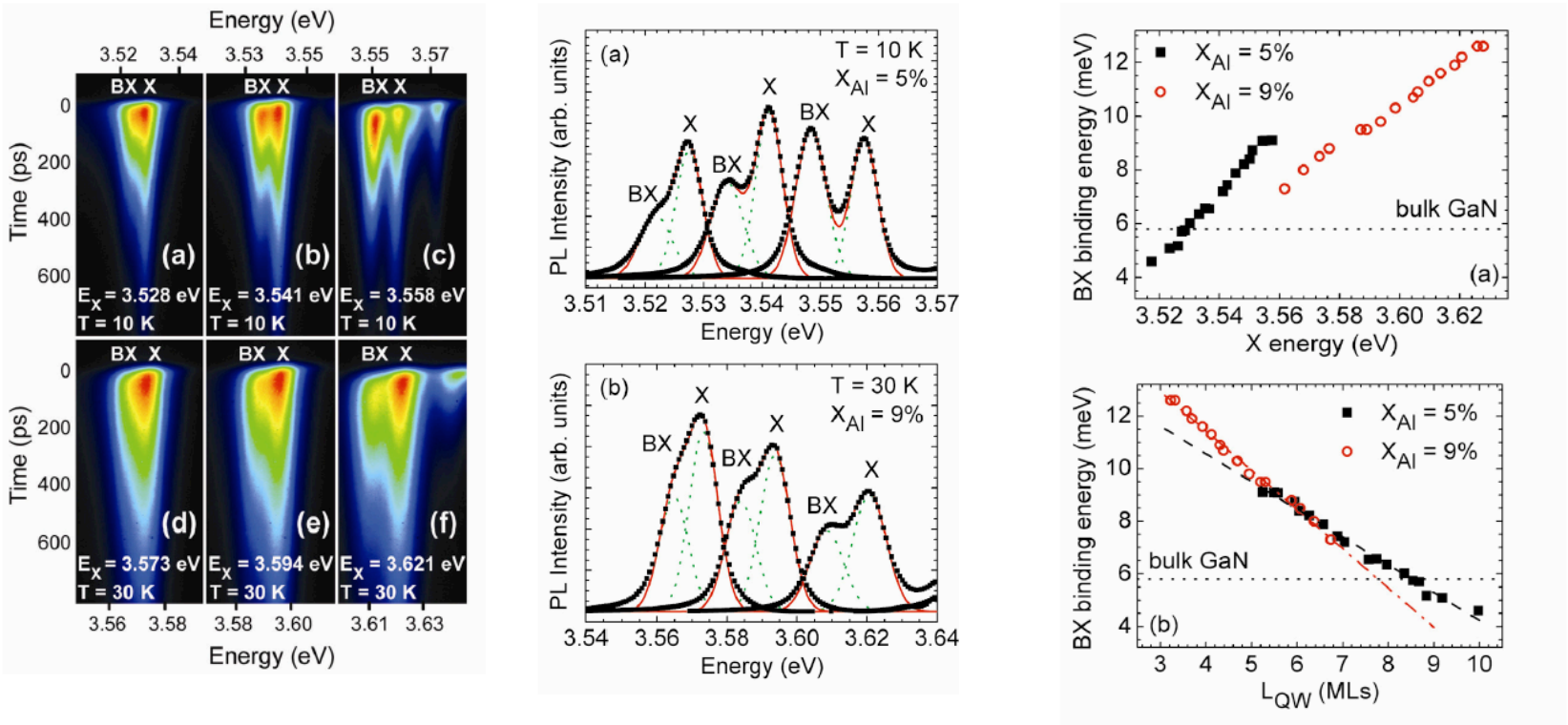
Resonant excitation

Only free excitons give rise to biexcitons: biexciton formation is quenched by exciton localization

PHYSICAL REVIEW B 77, 125342 (2008)

Biexciton kinetics in GaN quantum wells: Time-resolved and time-integrated photoluminescence measurements

Flavian Stokker Cheregi,[1] Anna Vinattieri,[1] Eric Feltin,[2] Dobri Simeonov,[2] Jean-François Carlin,[2] Raphaël Butté,[2] Nicolas Grandjean,[2] and Massimo Gurioli[1]
[1]*Department of Physics and LENS, Università di Firenze, 50019 Sesto Fiorentino, Italy*
[2]*Institute of Quantum Electronics and Photonics, Ecole Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland*

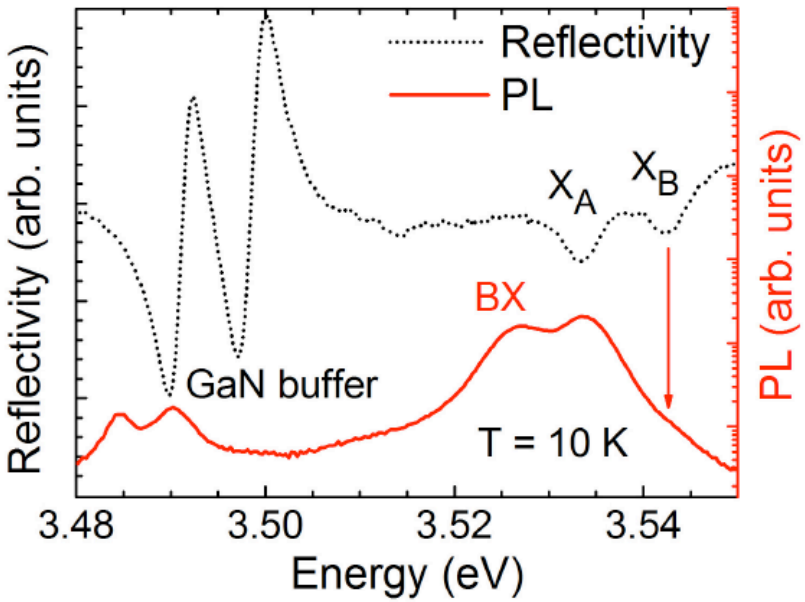


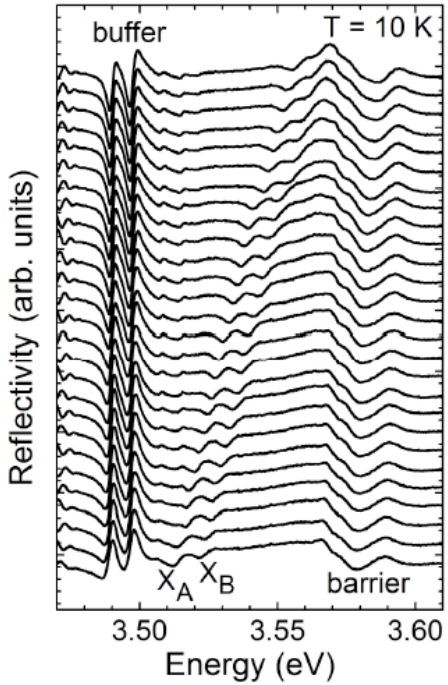
The biexciton binding energy increases with increasing quantum confinement. For low quantum confinement values, the BX binding energy is found to decrease in the $GaN/Al_{0.05}Ga_{0.95}N$ QW below that of bulk GaN. This result is a fingerprint of the strong QCSE present in these structures.

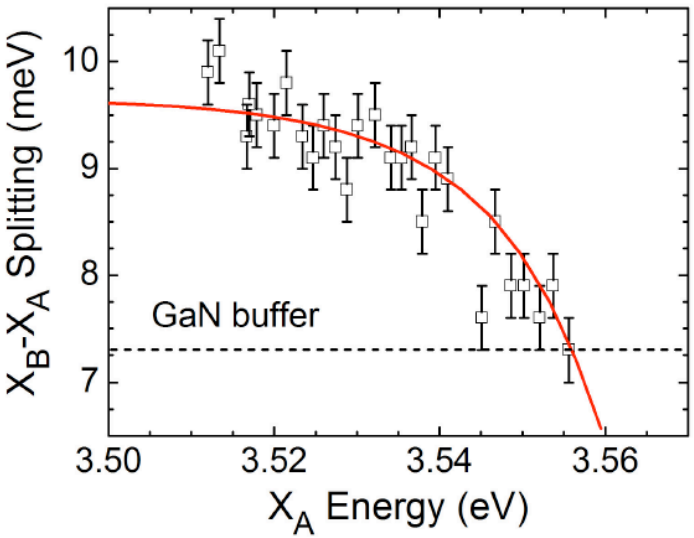
APPLIED PHYSICS LETTERS 93, 152105 (2008)

Impact of quantum confinement and quantum confined Stark effect on biexciton binding energy in GaN/AlGaN quantum wells

F. Stokker-Cheregi,[1,a)] A. Vinattieri,[1] E. Feltin,[2] D. Simeonov,[2] J. Levrat,[2] J.-F. Carlin,[2] R. Butté,[2] N. Grandjean,[2] and M. Gurioli[1]
[1]*LENS and Department of Physics, University of Florence, I-50019 Sesto Fiorentino, Italy*
[2]*Institute of Quantum Electronics and Photonics, Ecole Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland*







When decreasing the well thickness, while the energies of both X_A and X_B increase, X_B approaches the barrier limit before X_A does, so that the electron and hole wavefunctions for X_B are more sensitive to delocalization effects outside the well region making the overall energy enhancement due to the increase in quantum confinement smaller than for X_A.

PHYSICAL REVIEW B **79**, 245316 (2009)

Quantum confinement dependence of the energy splitting and recombination dynamics of A and B excitons in a GaN/AlGaN quantum well

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A. Vinattieri
Department of Physics and LENS, University of Florence, Sesto, I-50019 Fiorentino, Italy

E. Feltin, D. Simeonov, J.-F. Carlin, R. Butté, and N. Grandjean
Institute of Quantum Electronics and Photonics, Ecole Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland

F. Sacconi, M. Povolotskyi, and A. Di Carlo
Department of Electronic Engineering, University of Rome "Tor Vergata," 00133 Rome, Italy

M. Gurioli
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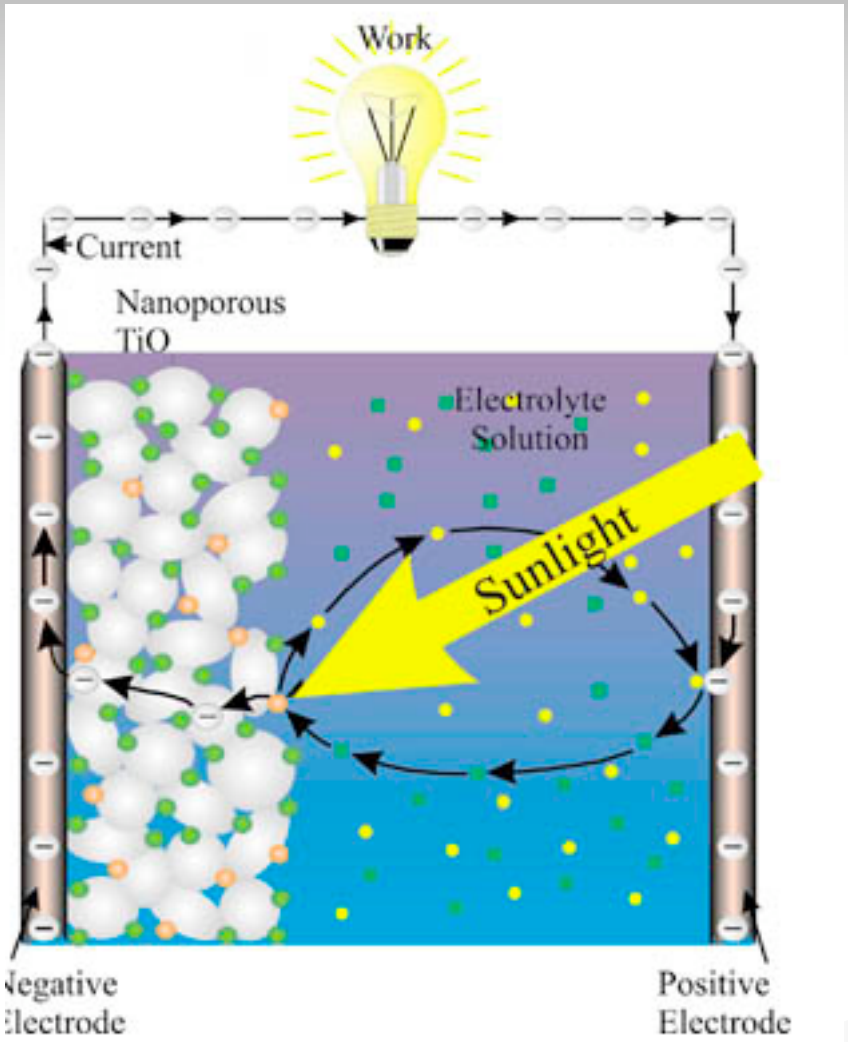
Nanoparticles for dye-sensitized solar cells



Graetzel, Nature 414,338 (2001)

$\eta \approx 11\ \%$

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Open questions

- **Charge collection & transport**
  - Solar spectrum coverage
  - Lifetime (dye & electrolyte)

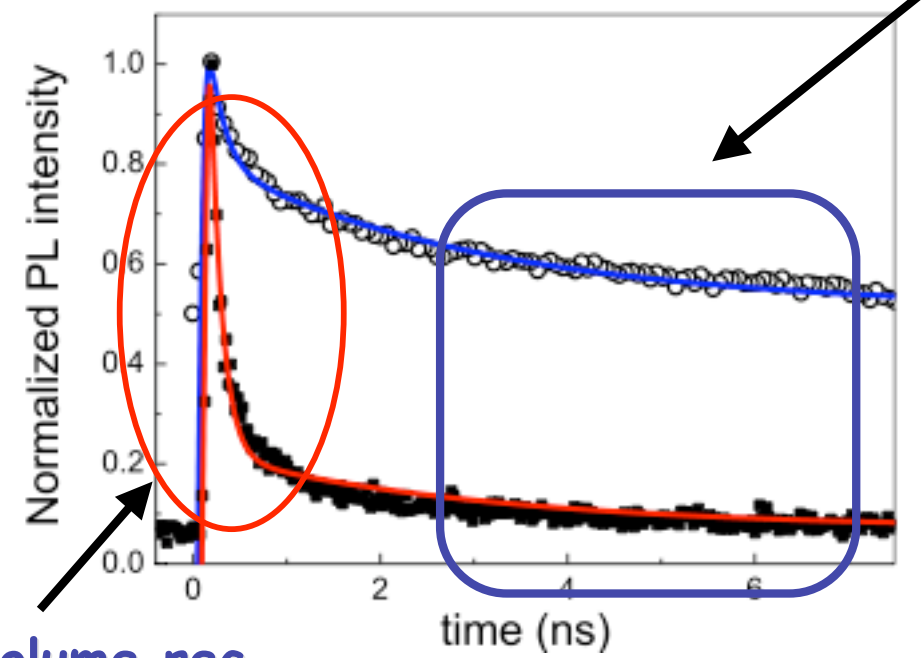
It's necessary to understand the physics of charge transfer and transport in the semiconductor mesoporous strate

Surface vs Volume contributions

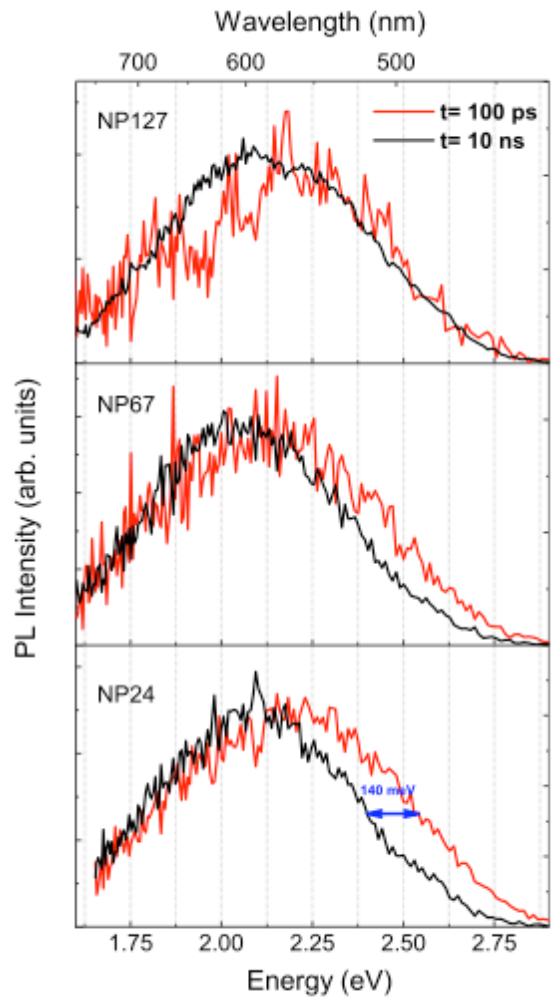
Controlling the surface state recombination

Modifying the NPs environment and size

Surface rec



Volume rec



JOURNAL OF APPLIED PHYSICS **106**, 053516 (2009)

Volume versus surface-mediated recombination in anatase TiO_2 nanoparticles

Lucia Cavigli,[1,a)] Franco Bogani,[1] Anna Vinattieri,[2] Valentina Faso,[3] and Giovanni Baldi[3]
[1]*Dipartimento di Energetica, Università di Firenze, I-50134 Firenze, Italy*
[2]*Dipartimento di Fisica, CNISM, and LENS, Università di Firenze, I-50019 Sesto Fiorentino (Firenze), Italy*
[3]*Centro Ricerche Colorobbia (Ce.Ri.Col.) Italia, I-50053 Sovigliana-Vinci (Firenze), Italy*

Work in progress

- Nitride heterostructures: Exciton-phonon interaction

L.Cavigli et al: Probing the exciton density of states through the phonon assisted emission in GaN epilayers: the A and B exciton contribution, PRB under submission

- NPs for DSSC: Nanostructuring of the Semiconductor layer - Nanorods, Nanotubes, CNTs scaffolds (PoliTO, CNR-Pi)

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