# **OPTICS II** Nanophotonics and quantum optics

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«NanoPhysique et SemiConducteurs» CEA-Grenoble

Ganex School 2013, Montpellier

#### Light-matter coupling $\Rightarrow$ Some notions, peculiarities of GaN $\Rightarrow$ The "strong exciton" in GaN

#### **Microcavity physics**

 $\Rightarrow$  From the weak to the strong coupling  $\Rightarrow$  Why do microcavity physics in GaN?

Books on the topic:

Semiconductor Optics, Springer (C. F. Klingshirn)

Confined Electrons and Photons: New Physics and Applications, Springer *(E. Burstein and C. Weisbuch)* 

Units

$$\mathcal{L} = \frac{c}{\upsilon} = \frac{hc}{E} = \frac{1}{\tilde{\upsilon}}$$

$$\begin{aligned}
\lambda = 1 \,\mu m \\
\Leftrightarrow \\
\upsilon = 3.10^{14} \,Hz \\
\Leftrightarrow \\
E = 1.24 \,eV \\
\Leftrightarrow \\
\tilde{\upsilon} = 10^4 \,cm^{-1}
\end{aligned}$$

## **Excitons and polaritons in GaN**

The excitons in GaN

- $\Rightarrow$  See Optics I (M. Leroux)
- $\Rightarrow$  A, B and C excitons

What does an exciton look like?

- $\Rightarrow$  A well defined wavevector for the center of mass
- $\Rightarrow$  The electron and hole are closely bound (Bohr radius)



#### **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<sup>‡</sup> Physics Department, Cornell University, Ithaca, New York (Received July 16, 1958)

#### Polariton = mixed exciton-photon particles

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# **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



#### **Excitons and polaritons in GaN**



#### **Excitons and polaritons in GaN**



#### **Consequences of the polariton picture**





### **Consequences of the polariton picture**



#### **Consequences of the polariton picture**

#### Luminescence



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

#### **The polariton in GaAs**

Volume 27, Number 24

#### PHYSICAL REVIEW LETTERS

**13 December 1971** 

# **Observation of Polaritons in GaAs:** A New Interpretation of the Free-Exciton Reflectance and Luminescence

D. D. Sell, R. Dingle, S. E. Stokowski, and J. V. DiLorenzo Bell Laboratories, Murray Hill, New Jersey 07974 (Received 1 November 1971)



#### **Excitons and polaritons in GaN**



Fig. 1. 2 K reflectance spectrum (dashed line) and the corresponding 2 K photoluminescence (open circles) spectrum which shows the LPB and UPB contributions to the GaN fluorescence.



B. Gil et al., Solid State Commun. 104 264 (1997)

#### **Confined electronic structures**

The strong light-matter coupling in bulk GaN....

... creates strong polaritons, which are lousy emitters

What about QWs?

A confined exciton in a QW is coupled to a large number of photonic states  $\Rightarrow$  Luminescence allowed on the first order

V. M. Agranovitch and O. A. Dubovskii, JETP Lett. **3**, 223 (1966) C. Weisbuch *et al.*, Solid State Commun. **37**, 219 (1981)

 $\Rightarrow$  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)

# **Confined electronic structures : GaN**

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**



$$F_{\rm rad} \propto {1 \over {\rm nE}^2 {
m f}}$$



Strong confinement

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



Lateral weak confinement  $\psi_X(r_e, r_h) \propto f(z_e) g(z_h) e^{-r/a_{2D}} e^{-R_p^2/2\xi}$   $f = \frac{8E_p}{E} \left(\frac{\xi}{a_{2D}}\right)^2$ 

 $\Rightarrow$  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

#### **Excitonic effects in QWs**

2D limit = intrinsic lifetime of two-dimensional exciton

#### ~ 10 ps for a GaAs/AIAs 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**

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

In a non perfect structure, the fundamental emitting state 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**

Polar QWs



J. Renard *et al.*, Appl. Phys. Lett. **95** 131903 (2009)

P. Lefebvre et al., Phys. Rev. B 59 15363 (1999)

#### **Excitonic effects in GaN 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??

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

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

 $\Rightarrow$  Quite strong localization effects, no giant oscillator strength

 $\Rightarrow$  Strong excitonic effects in III-N, but ordinary recombination rates

More control on light-matter coupling

What are microcavities ?

How does an emitter in a microcavity behave ? Strong coupling Weak coupling

Two-dimensional microcavity polaritons Peculiarities of the III-N system

# **Optical cavities**

What is a mode of the E-M field?

 $\Rightarrow$  A solution of Maxwell equations in the absence of source

What is a confined mode?

« An optical resonator, the optical counterpart of an electronic resonant circuit, confines and stores light at certain resonant frequencies »

« Fundamentals of photonics » B.E.A. Saleh and M.C. Teich, Wiley ed.

« Optical resonators [...] are used primarily in order to build up large field intensities with moderate power inputs. They consist in most cases of two, or more, curved mirrors that serve to "trap," by repeated reflections and refocusing, an optical beam that thus becomes the mode of the resonator. »

> « Optical electronics in modern communications » A. Yariv, Oxford ed.

#### **Optical cavities**



 $\Rightarrow$  High intensity in a confined region of space for resonant frequencies

Quality factor measures the decay of stored E-M energy in mode :

$$E = E_0 e^{-\frac{\omega t}{Q}}$$

#### Various semiconductor microcavities



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#### **Spontaneous emission**

Spontaneous emission : irreversible emission of a photon





Influence of the E-M surrounding:



# Mirror

#### **Spontaneous emission**



Control of the *emitter-field* coupling

Control of the *density of states* for the EM field

# **Cavity Quantum ElectroDynamics**

**Free space emission** 



### **Strong coupling**



#### **Strong coupling**





#### Weak coupling, Purcell effect



### Weak coupling, Purcell effect

#### A famous paper :

B10. Spontaneous Emission Probabilities at Radio Frequencies. E. M. PURCELL, *Harvard University.*—For nuclear magnetic moment transitions at radio frequencies the probability of spontaneous emission, computed from

 $A_{\nu} = (8\pi r^2/c^3)h\nu(8\pi^3\mu^2/3h^2)$  sec.<sup>-1</sup>,

is so small that this process is not effective in bringing a spin system into thermal equilibrium with its surroundings. At 300°K, for  $\nu = 10^7$  sec.<sup>-1</sup>,  $\mu = 1$  nuclear magneton, the corresponding relaxation time would be  $5 \times 10^{21}$  seconds! However, for a system coupled to a resonant electrical circuit, the factor  $8\pi r^2/c^2$  no longer gives correctly the number of radiation oscillators per unit volume, in unit frequency range, there being now one oscillator in the frequency range  $\nu/Q$  associated with the circuit. The spontaneous emission probability is thereby increased, and the relaxation time reduced, by a factor  $\int = 3Q\lambda^3/4\pi^3 V$ , where V is the volume of the resonator. If a is a dimension characteristic of the circuit so that  $V \sim a^2$ , and if  $\delta$  is the skin-depth at frequency  $\nu$ ,  $\int \sim \lambda^3/a^3 \delta$ . For a non-resonant circuit  $f \sim \lambda^2/a^3$ , and for  $a < \delta$  it can be shown that  $f \sim \lambda^3/a\delta^2$ . If small metallic particles, of diameter 10<sup>-1</sup> cm are mixed with a nuclear-magnetic medium at room temperature, spontaneous emission should establish thermal equilibrium in a time of the order of minutes, for  $\nu = 10^{2}$  sec.<sup>-1</sup>.

E.M. Purcell Phys. Rev. **69**, 681 (1946)



# Weak coupling, emission diagram modification Atomes à l'Intérieur d'un Interférometre Perot-Fabry A. Kastler Example : 1 atom in a Fabry-Pérot cavity, emitting at $\omega$ θ $I(\omega, \theta) = \frac{1 + r^{2} + 2 * r \cos(2\phi_{1})}{|1 - r^{2}e^{-2i\phi}|^{2}}$ reflection coefficient r d n<sub>cav</sub> $\phi = n_{cav} L \cos(\theta) \frac{\omega}{c}$ $\phi_1 = n_{cav} d \cos(\theta) \frac{\omega}{c}$

A. Kastler, Appl. Optics 1,17 (1962)

# Weak coupling, emission diagram modification

A. Kastler, Appl. Optics 1,17 (1962)

Emission for a centered atom (varying  $\omega$ ) at normal incidence, r=0.9



# Weak coupling, emission diagram modification



#### Only **spectral and angular intensity redistribution** No spontaneous emission rate modification!!

 $\Rightarrow$  But can be used to extract more light out of a high index material !!

H. Benisty, H. De Neve and C.Weisbuch, IEEE J. Quantum Electron., 34, pp. 1612–1643 (1998)

# Weak coupling, emission diagram modification



Grâce a l'interposition des faces semi-réfléchissantes, l'énergie lumineuse émise par la couche luminescente, au lieu d'être uniformément répartie dans toutes les directions, est concentrée dans des anneaux fins et extrêmement brillants.

A. Kastler, Appl. Optics 1,17 (1962)
#### Weak coupling, emission diagram modification



Were you there? Some of the participants in the Paris ICO meeting July 1961

Photo: Harand

A. Kastler, Appl. Optics 1,17 (1962)

Spontaneous emission depends on the E-M field mode structure

The SE radiation pattern is in general modified by the E-M field boundaries

When resonantly coupled to a cavity mode, the SE rate can be modified: - Weak coupling : faster SE in the « overdamped » regime => Purcell effect

Strong coupling : damped reversible SE
=> mixed emitter-photon modes

### Strong coupling in a planar cavity



Quantum Well



$$\Psi_{X}(r_{e}, r_{h}) \propto f(z_{e}) g(z_{h}) e^{-r/a_{2D}} e^{i\vec{K}_{x,y}.\vec{R}_{x,y}}$$
$$\vec{R}_{x,y} = \frac{m_{e}^{\dagger}\vec{r}_{e} + m_{h}^{\dagger}\vec{r}_{h}}{m_{e}^{\dagger} + m_{h}^{\dagger}}, \vec{K}_{x,y} = \vec{k}_{x,y}^{e} + \vec{k}_{x,y}^{h}, \vec{r} = \vec{r}_{e} - \vec{r}_{h}$$

In-plane translational invariance for confined field and QW excitons

 $\Rightarrow$  one to one coupling of confined modes and excitons of same  $\mathbf{k}_{x,v}$ 

#### Strong coupling in a planar cavity



#### $\Rightarrow$ Creation of mixed exciton-photon states : **2D polaritons**

C. Weisbuch et al., Phys. Rev. Lett. 69, 3314 (1992)

## **Bragg mirrors**

DBR = Distributed Bragg Reflector

 $\Rightarrow$  rely on periodicity to have Bragg reflection



# **Bragg mirrors**





#### **Comparison of III-N and III-As systems**

$$R_{max} = 1 - 4 \frac{n_{sup}}{n_{sub}} \left(\frac{n_{l}}{n_{h}}\right)^{2N}$$

III-As

n<sub>GaAs</sub>=3.5, n<sub>AIAs</sub>=3 Lattice-matched R=99%

N=16

III-N

n<sub>GaN</sub>=2.3, n<sub>AIN</sub>=2.1 Lattice mismatch=2.4%

R=99% N=20 to 60 for GaN/AlGaN DBR

Al<sub>0.2</sub>Ga<sub>0.8</sub>N/Al<sub>0.85</sub>In<sub>0.15</sub>N Lattice matched R=99% N=27

E. Feltin et al., Appl. Phys. Lett. 88 051108 (2006)

#### **DBR based Fabry-Pérot**



#### Bragg mirrors as 1D photonic band gap

R. P. Stanley et al., Phys. Rev. A 48, 2246 (1993)

Analogy between Maxwell and Schrödinger equations

 $\Rightarrow$  the Bragg reflector as a 1D photonic bandgap



### **DBR based Fabry-Pérot**

Longitudinal E-M field profile at resonance:



 $\Rightarrow$  Field peaked at cavity center, exponential decay in the DBRs Mode extent larger than for metal mirror cavity!

#### **Planar microcavities : the photon effective mass**



# Strong coupling in a planar cavity

New dispersion relation :





Relaxation bottleneckF. Tassone *et al.*, Phys. Rev. B 53, R7642 (1996)(high DOS vs low DOS at  $k_{x,y} \sim 0$ )

BUT excitons = bosons in the low density regime

- $\Rightarrow$  Stimulated emission of polaritons : R=R<sub>0</sub>(1+<N>)
- ⇒ Polariton laser or solid-state BEC

A. Imamoglu *et al.* Phys. Rev. A **53**, 4250 (1996)

### **Strong coupling in a planar cavity**

#### "Bose-Einstein condensation of exciton polaritons"

 $\Rightarrow$  reach stimulated relaxation of polaritons before exciton screening



J. Kasprzak et al., Nature 443, 409 (2006)

B. Deveaud-Plédran "On the condensation of polaritons" J. Opt. Soc. Am. B **29** A138 (2012)

It is central at this stage to correct a misuse of words: conventional lasing in semiconductors does not correspond to population inversion (which would mean having more electrons in the conduction band than electrons in the valence band). The lasing condition in semiconductors is the well-known Bernard– Durraffourg condition [63], i.e., that the distance between the quasi-Fermi levels for electron and holes is larger than the energy of the gap. It is therefore misleading to talk, in semiconductors, about lasing without inversion. D. Bajoni "Polariton lasers. Hybrid lightmatter lasers without inversion" J. Appl. Phys. D: Appl. Phys. **45** 409501 (2012)

The phenomenon of polariton condensation is exactly equivalent to the polariton lasing we have described in the previous section, and further investigation showed that thermal equilibrium is not achieved by the polariton gas, so that **the term Bose-Einstein condensate is not exact** and a weaker definition of condensation, which does not involve thermal equilibrium, has been applied to the justify the term condensation for polaritons

#### **Reference system : GaAs/AIAs cavities, InGaAs QWs**

 $\Rightarrow$  Very large Q factors (10000-50000)

 $\Rightarrow$  long polariton lifetime ~15 ps and little disorder induced localization

But : restricted to low temperatures restricted to low pump powers

III-N system:

- $\Rightarrow$  Large intrinsic light matter coupling
- $\Rightarrow$  QW exciton stable at 300 K, more stable at large pump power

But : high quality mirrors difficult to fabricate, much photonic disorder

The "Mott density" for 2D excitons

$$n \approx \frac{1}{25 a_B^2}$$

Smooth transition to e-h plasma, due to phase-space filling

	a (nm)	N <sub>mott</sub> (cm-2)
GaN	2.8	5 10 <sup>11</sup>
GaAs	12	2.5 10 <sup>10</sup>

N. F. Mott, Philos. Mag. **6** 287 (1961) S. Schmitt-Rink *et al.*, Phys. Rev. B **32** 6601 (1985)



- (1) GaN excitons are stable at room temperature;
- (2) the light-matter coupling is enormously strong, as we will show below; and
- (3) GaN technology is now increasingly well developed, and the observation of the strong coupling regime in such structures seems likely to be achieved.







#### Bulk GaN, Q~100, 5 K, but strong coupling $\Omega$ =30 meV

A. Antoine-Vincent et al., Phys. Rev. B 68 153313 (2003)



F. Semond et al., Appl. Phys. Lett. 87 021102 (2005)



Room temperature strong coupling !



R. Butté et al., Phys. Rev. B 73 033315 (2006)







#### Room temperature polariton lasing

G. Christmann et al., Appl. Phys. Lett. 93 051102 (2008)

# **The Purcell effect**



$$\Gamma = \Gamma_{cav} + \Gamma_{leak} \sim \Gamma_0(F_p + 1)$$

 $\beta$ =(photons in confined mode)/(emitted photons)= $\Gamma_{cav}/\Gamma$ 

$$\beta = F_p / (F_p + 1)$$

 $\Rightarrow$  For high Fp, **monomode emission** without inhibition!!

## **The Purcell effect**

#### **Application prospects**



# **The Purcell effect**



High  $\beta$  regime : all photons feed into mode *below threshold!!* 

 $P_{th} = \gamma_{cav} (1+\beta)/2\beta => P_{th} \text{ scales as } 1/\beta$ For  $\beta \sim 1$  and Q $\sim 10000$ ,  $P_{th} \sim 20$  nA!

Y. Yamamoto et al., Phys. Rev. A 44, 657 (1991)

Whispering gallery modes confined by total internal reflection



G. Mie, Ann. Phys. (NY) **25**, 377 (1908) Lord Rayleigh, « The problem of the whispering gallery », Scientic Papers **5**, 617 (1912)





#### InGaN QWs in a GaN slab

#### Lasing at room temperature in a microlaser

A. C. Tamboli et al., Nature Photon. 1 61 (2007)



GaN quantum dots in AIN slab on Si



M. Mexis et al., Opt. Lett. 36 2203 (2011)





M. Arita et al., Appl. Phys. Exp. 5 126502 (2012)



#### GaN quantum dots in AIN on Si



D. Sam-Giao et al., Appl. Phys. Lett. 100 191104 (2012)

#### **Prospects**

- UV microlasers based on the Purcell effect

- Strong coupling in other geometries than planar cavity  $\Rightarrow$  Strong coupling in photonic crystal slabs

### **Microdisk cavities**



FIG. 1. Scanning electron microscope images of DWELL-containing microdisk cavities after the (a)  $Si_xN_y$  etch, and (b) and (c) AlGaAs etch and undercut.



K. Srinivasan et al., Appl. Phys. Lett. 86, 151106 (2005)

Remark : Result for empty cavities... probably less when QDs are inserted due to the increase of residual background absorption

#### **2D photonic band gap cavities**



Y. Akahane et al., Nature 425, 944 (2003)

# Fundamental papers (my very subjective selection)

#### 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<sup>‡</sup> Physics Department, Cornell University, Ithaca, New York (Received July 16, 1958)

VOLUME 27, NUMBER 24 PHYSICAL REVIEW LETTERS 13 DECEMBER 1971

#### **Observation of Polaritons in GaAs:** A New Interpretation of the Free-Exciton Reflectance and Luminescence

D. D. Sell, R. Dingle, S. E. Stokowski, and J. V. DiLorenzo Bell Laboratories, Murray Hill, New Jersey 07974 (Received 1 November 1971)

VOLUME 33, NUMBER 14

PHYSICAL REVIEW LETTERS

30 September 1974

#### Quantum States of Confined Carriers in Very Thin Al<sub>x</sub>Ga<sub>1-x</sub>As-GaAs-Al<sub>x</sub>Ga<sub>1-x</sub>As Heterostructures

R. Dingle, W. Wiegmann, and C. H. Henry Bell Laboratories, Murray Hill, New Jersey 07974 (Received 24 June 1974)

RAPID COMMUNICATIONS

PHYSICAL REVIEW B

VOLUME 30, NUMBER 2

15 JULY 1984

Reflectance of two-dimensional excitons in GaAs-AlGaAs quantum wells

L. Schultheis<sup>\*</sup> and K. Ploog Max-Planck-Institut für Festkörperforschung, D-7000 Stuttgart 80, Federal Republic of Germany (Received 27 March 1984)

# Fundamental papers (my very subjective selection)

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VOLUME 58, NUMBER 20 PHYSICAL REVIEW LETTERS	18 1	MAY 1987			
Inhibited Spontaneous Emission in Solid-State Physics	s and Electronics				
Eli Yablonovitch Bell Communications Research, Navesink Research Center, Red Ban (Received 23 December 1986)	k, New Jersey 07701				
		Volume 67,	NUMBER 17	PHYSICAL REVIEW LETTERS	21 October 1991
			Enhanced Radi	ative Recombination of Free Excitons in GaAs Quant	ım Wells
			Centre	B. Deveaud and F. Clérot National d'Etudes de Télécommunications, 22300 Lannion, France	
				N. Roy, K. Satzke, and B. Sermage	
			Centre	National à Etudes de l'elecommunications, 92120 Bagneux, France D. S. Katzer	
			(Rec	Naval Research Laboratory, Washington, D.C. 20375 eived 15 April 1991; revised manuscript received 15 August 1991)	
VOLUME 40 NUMBER 22 DILVELOAL DEVIEW LETTERS					
VOLUME 09, NUMBER 23 FRISICAL REVIEW LETTERS	7 DECEMBER 199	_			
Observation of the Coupled Exciton-Photon Mode Splitting in a Semiconductor Quantum Microcavity					
C. Weisbuch, <sup>(a)</sup> M. Nishioka, <sup>(b)</sup> A. Ishikawa, and Y. Arakawa Research Center for Advanced Science and Technology, University of Tokyo, 4-6-1 Meguro-ku, Tokyo 153, Japan (Received 12 May 1992)					
	phys. stat. sol. (b) <b>424</b> , No. 111, 234		9–2356 (2005) /	<b>DOI</b> 10.1002/pssb.200560972	
	Feature Article				
	Microcavities in Ecole Polytechnique Fédérale de Lausanne, Ecole Polytechnique (France) and elsewhere: past, present and future			ne, it	
	Claude Weisbuch <sup>*, 1, 2, 3</sup> and Henri Benisty <sup>**, 4</sup>				

# Fundamental papers (my very subjective selection)

