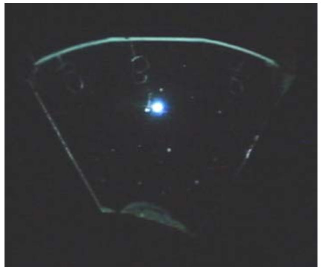
GaN Polariton Lasing

μphotonics

Jeremy J. Baumberg

UNIVERSITY OF CAMBRIDGE



Stavros Christopoulos
Alexei Kavokin
Guillaume Malpuech

Giorgio Baldassarri
Höger von Högersthal
Alastair Grundy
Pavlos Lagoudakis

University of Southampton

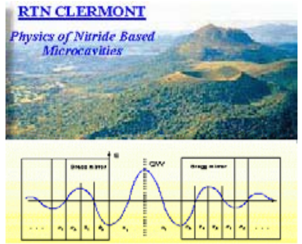
Gabriel Christmann
Raphael Butté
E. Feltin
J.-F. Carlin,
Nicolas Grandjean

EPFL
ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

EPSRC NanoPhotonics Centre

np.phy.cam.ac.uk

STIMSCAT



Funders: EPSRC, EU, Royal Society

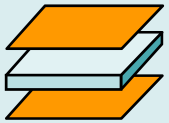
μphotonics NanoPhotonics Centre

nanostructure materials ⟷ modify light-matter interaction

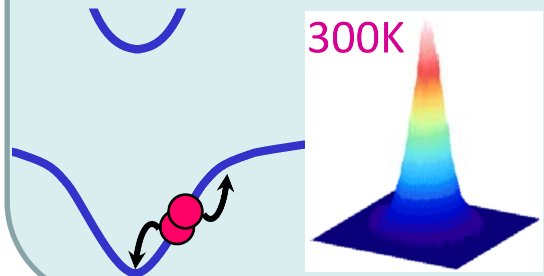
np.phy.cam.ac.uk

metamaterials

semiconductor microcavities

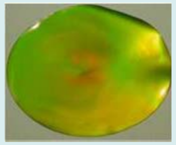


new dispersions
strong interactions
polariton lasers
Bose condensation

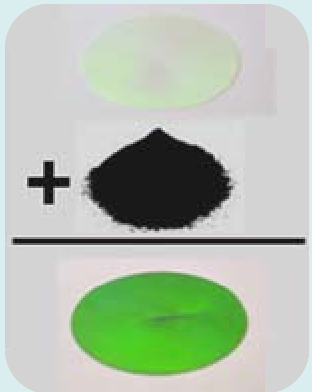


nano-assembly

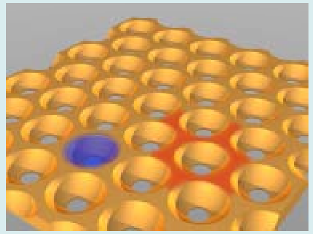
elastomeric polymer opals



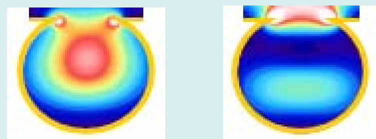
structural colour
long range order
enhanced scattering



plasmonics



localised plasmons
strong optical fields
300K strong coupling
molecular sensing



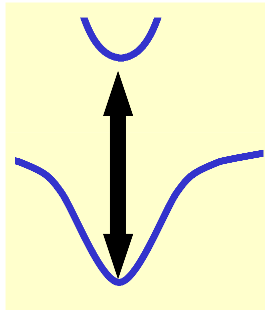
NanoPhotonics Centre move

- lab building and commissioning: opening April'08
- microcavity GaN and III-V experiments running

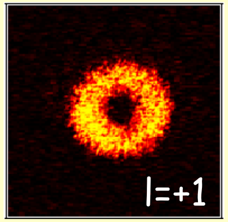
Strong coupling Microcavities

New directions:

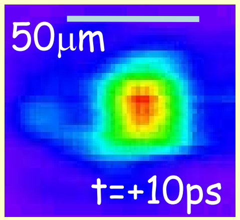
- understanding non-equilibrium coherent polariton states
- properties of coherent polariton states
- strong coupling at room temperature (devices)



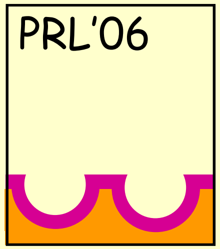
vortex dynamics



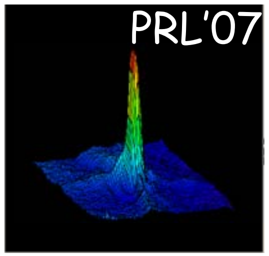
spatio-temporal dynamics



300K organic exciton-plasmons



300K polariton -laser



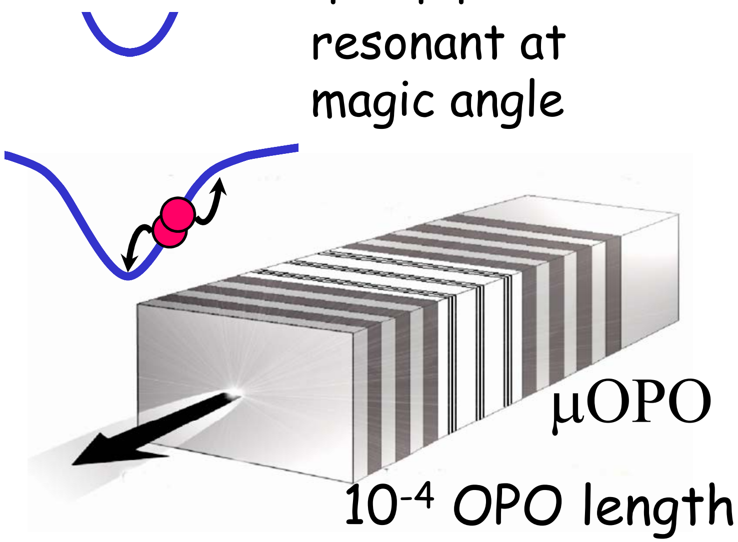
large Rabi splitting:

- plasmonics
- organics
- **GaN**, ZnSe

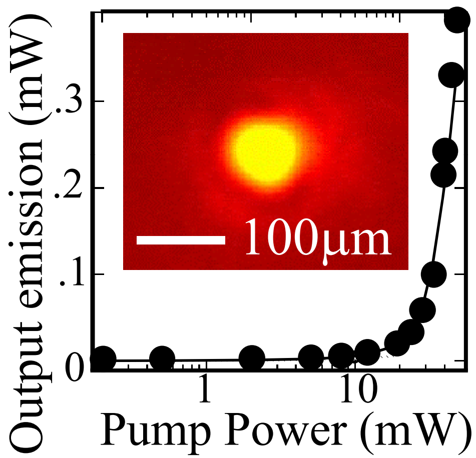
CW μOPO : Coherent Polariton State

- **strong polariton interactions**
  - dipole-dipole exciton mechanism: polaritons ~ ½ excitons
  - *E,k* matching: dispersion engineered by microcavity
- **stimulated scattering**
  - polaritons are bosons: scattering in $\propto 1 + N$ (ultrafast gain)
- **polariton OPO, OPA, lasing**: *plasers*

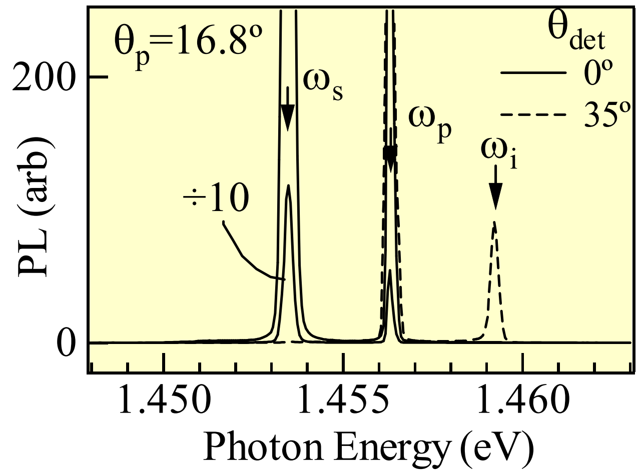
CW pump polaritons



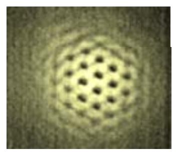
ultralow threshold



PRL **85**, 3680 (2000)
PRB **62**, R16247 (2000)

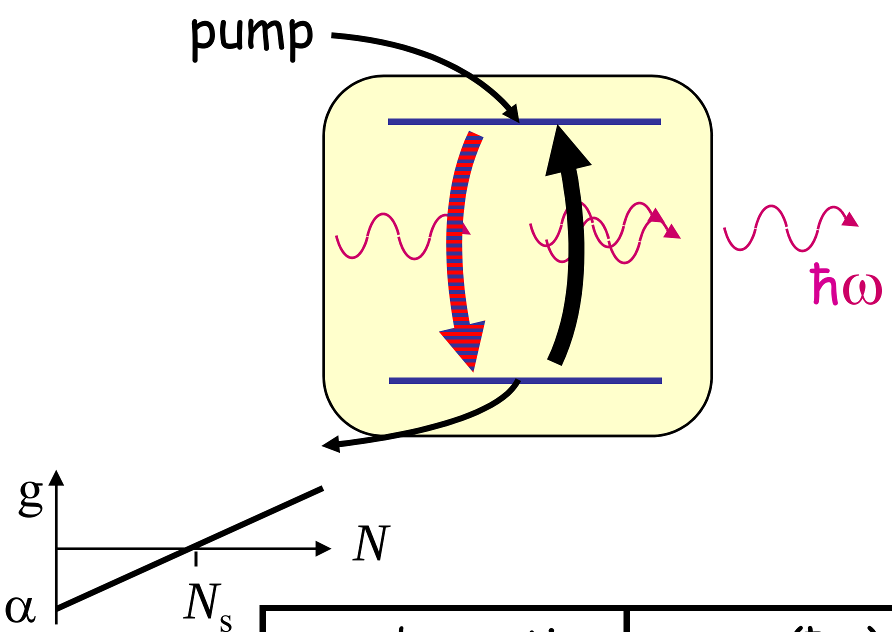


- **polariton condensation**
  - analogue of GP equation: atomic BECs
  - superfluid transport & interferometers



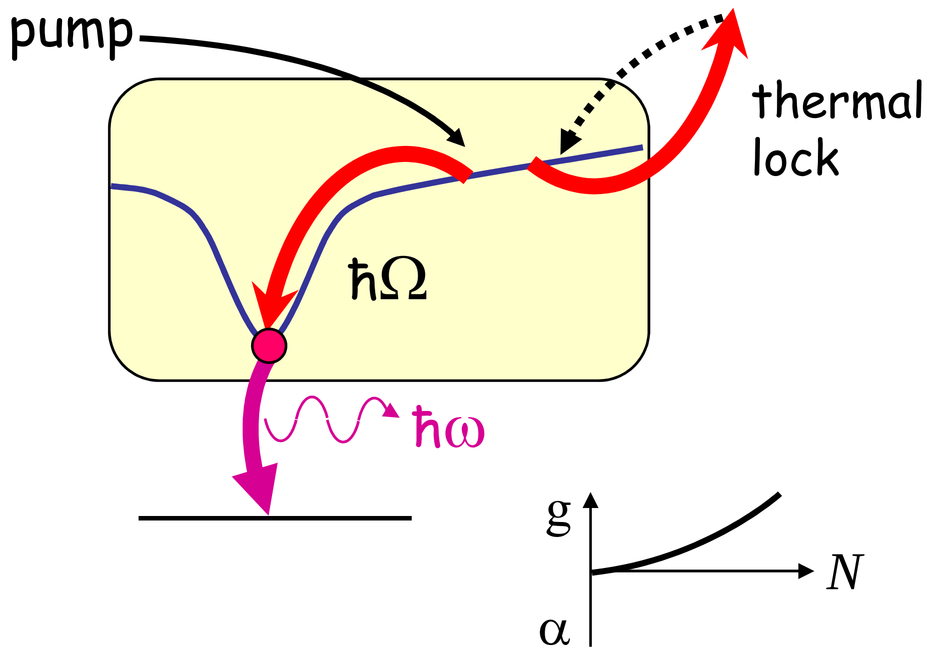
Polariton lasers (plasers)

- one particle stimulated emission



re-absorption	yes (ħω)
inversion	essential
matter	*e-h* plasma
low threshold	small volume

- two particle stimulated scattering

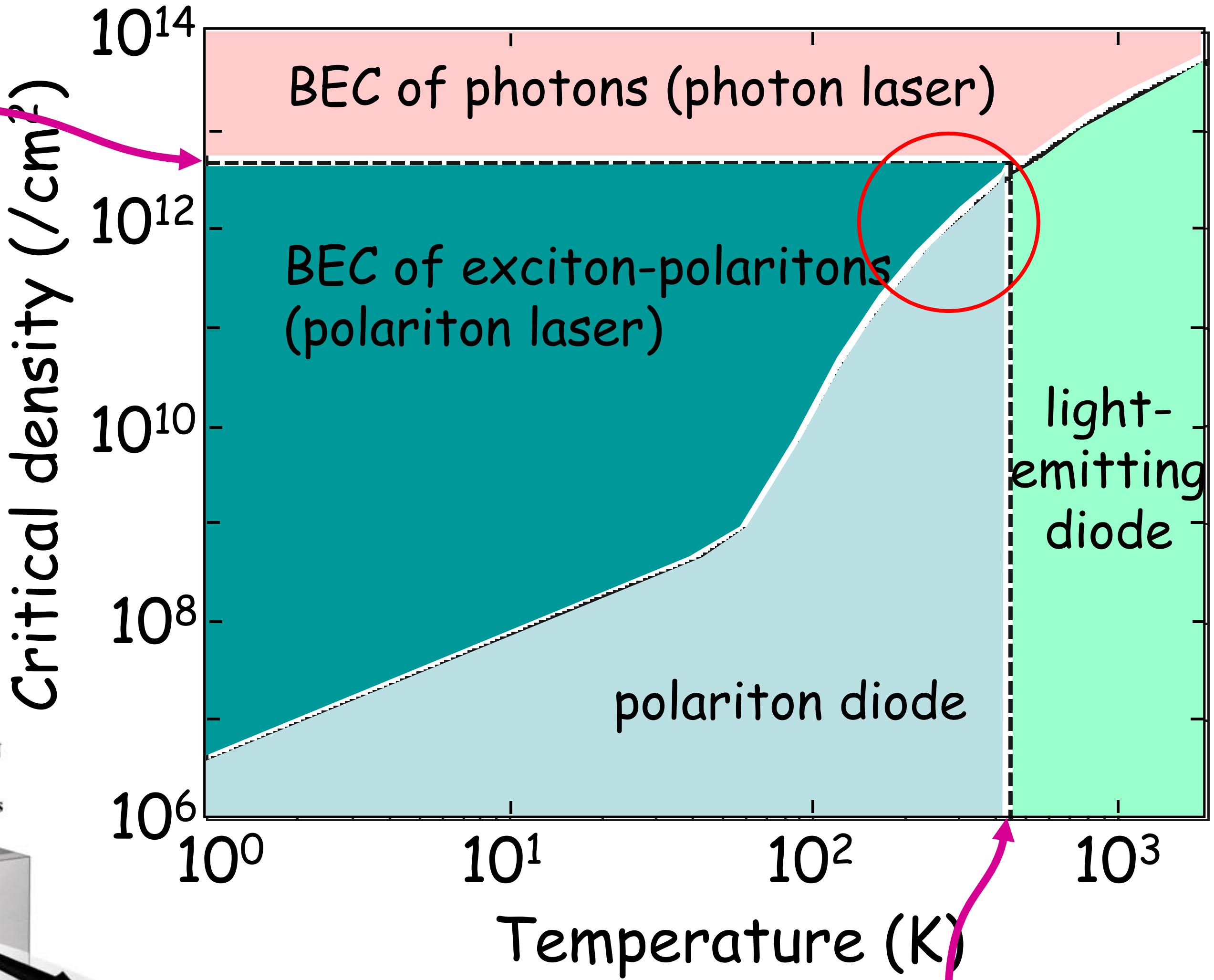


separates stimulation and emission ✓

issues:
- initiate pair scattering (eg. phonons, electron doping)
- need $kT \sim \Omega$: deep enough trap for 300K operation
- wide bandgap strong coupling: GaN, J-aggregate, ...
- thermalisation, localisation

Phase diagram: GaN

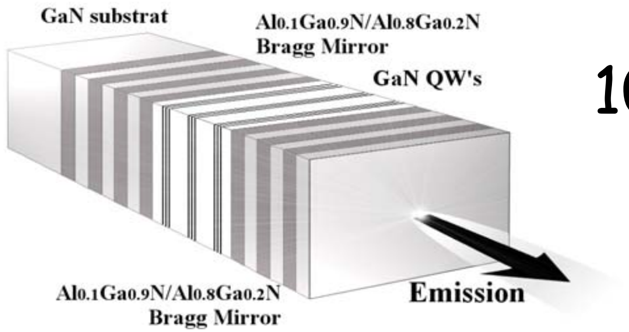
dissociation of excitons



ionization of excitons

GaN

f_{osc}	10x GaAs
E_b	10x GaAs
Ω_{Rabi}	10x GaAs



Malpuech,..., Baumberg *et al.*, *Appl. Phys. Lett.* **81**, 412 (2002)

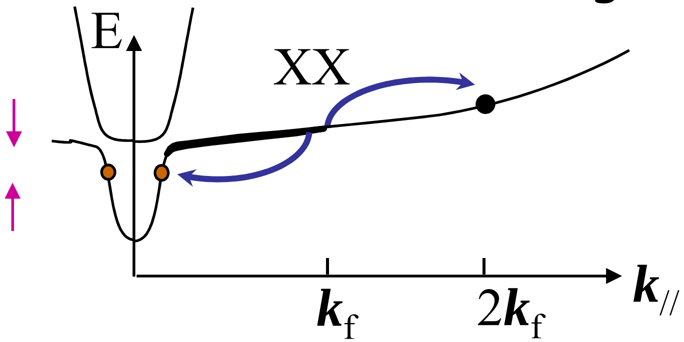
- models quasiclassical kinetics: fast formation

Encouraging Cooling

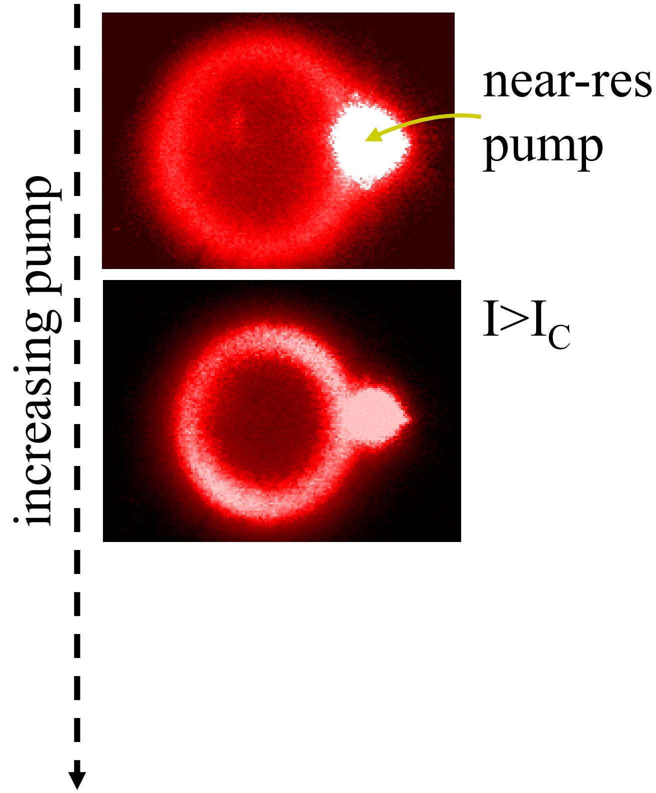
- scattering with electron reservoir $k_B T_e > (m_e/m_x)\ \Omega/2$

PRL **90**,206401 (2003)

- cooling from an exciton reservoir
  at low *T*, cooling **bottleneck:**



$k_B T_X > \Omega/2$



no optic phonons
acoustic phonons have too little energy
only hot excitons can pair scatter

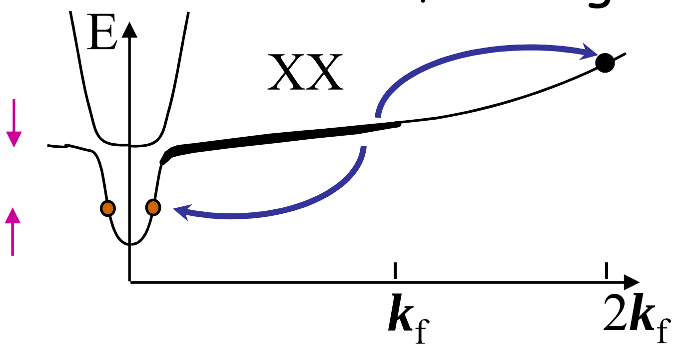
PRB **65**,73309 (2001)

Encouraging Cooling

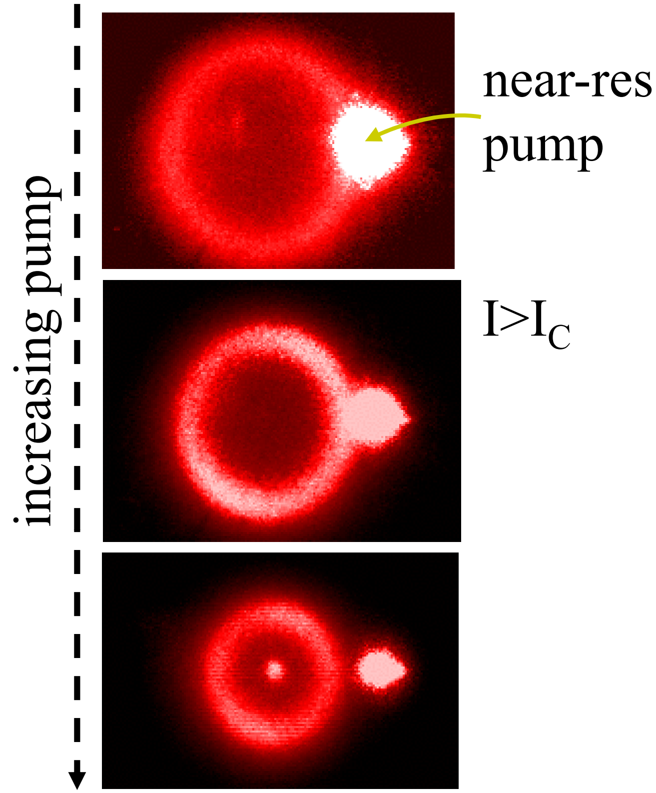
- scattering with electron reservoir $k_B T_e > (m_e/m_x)\,\Omega/2$

PRL **90**,206401 (2003)

- cooling from an exciton reservoir
  at low *T*, cooling **bottleneck:**



$k_B T_X > \Omega/2$



PRB **65**,73309 (2001)

no optic phonons
acoustic phonons have too little energy
only hot excitons can pair scatter

- .....but.... cooling in GaN is ultrafast,....

GaN Microcavities

- bulk GaN, for now since QW broadening, interfacial E-fields, ...
- different approaches for mirrors

G. Christmann
R. Butté
E. Feltin
J.-F. Carlin
N. Grandjean

ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

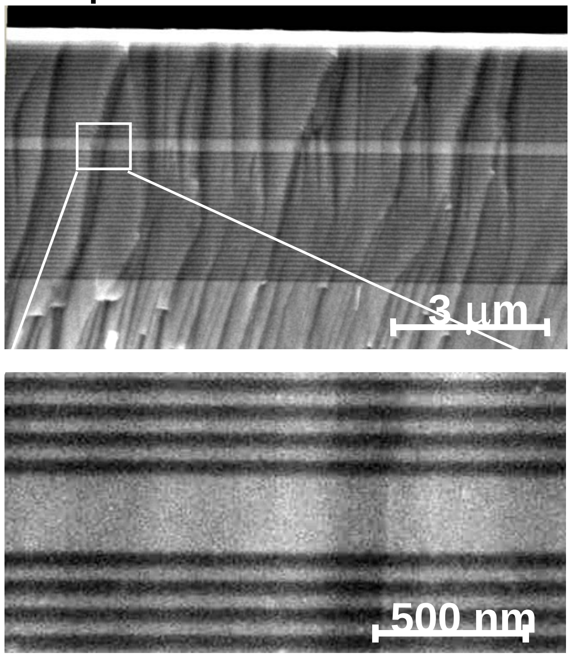
quantum photonics

e.g. full GaN microcavity

hybrid GaN μcav

key:
strain-relaxing template
lattice-matched
AlInN/AlGaN

28 pair AlInN/AlGaN DBR
3λ/2 GaN cavity
23 pair AlInN/AlGaN DBR



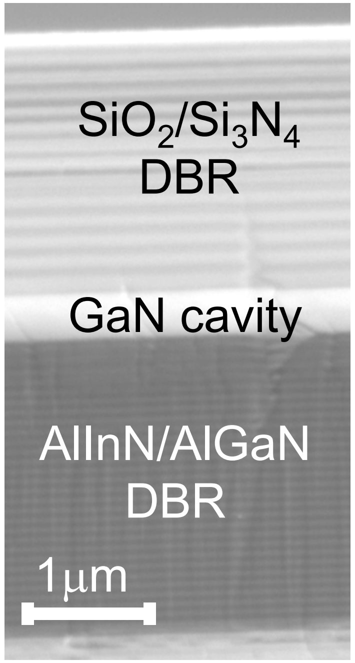
Carlin *et al.*, APL **86**, 031107 (2005)

Strong coupling at room temperature

previous work: 10K-300K CRHEA/LASMEA: PRB **68**, 153313 ('03)

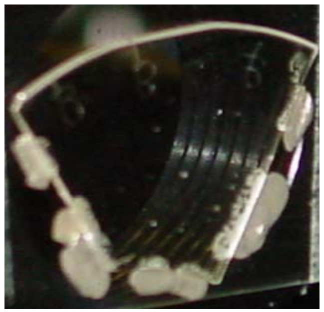
observed $\hbar\Omega$ = 31meV at 5K for $\lambda/2$ cavity

here: **hybrid bulk GaN µcav**



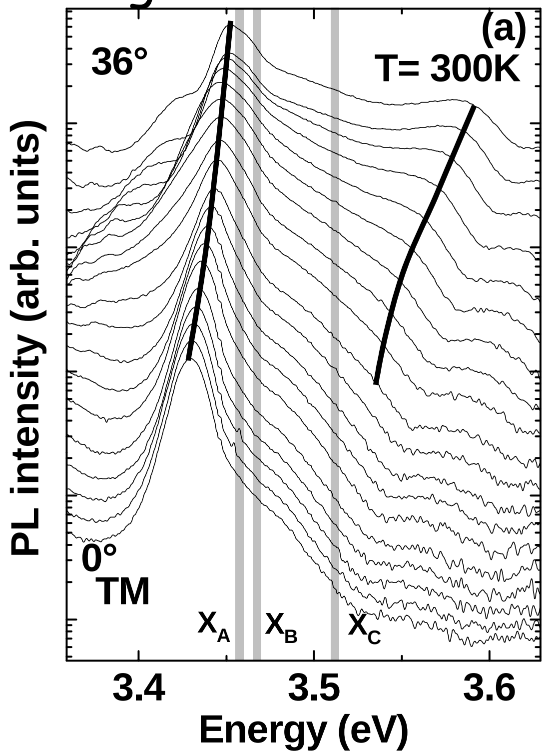
35 pair DBR, crack free
Q ~ 350, can be > 2000

high quality GaN
300K linewidths ~ 30meV
$3\lambda/2$ cavity: 210nm thick



- 244nm pump
- 50W/cm^2
- LP and UP visible
- separation ~ 60meV

angle-resolved PL



Butté, PRB **73**, 33315 ('06)

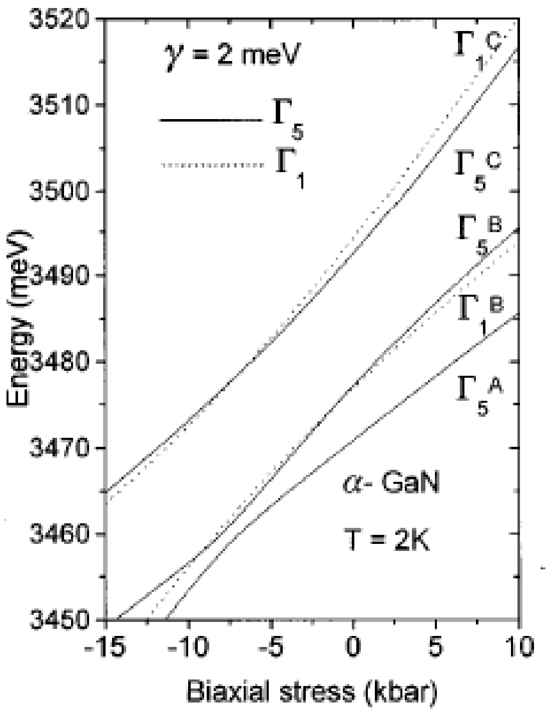
ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

Bulk GaN optical properties

3 optically active free excitons (X_A, X_B and X_C)

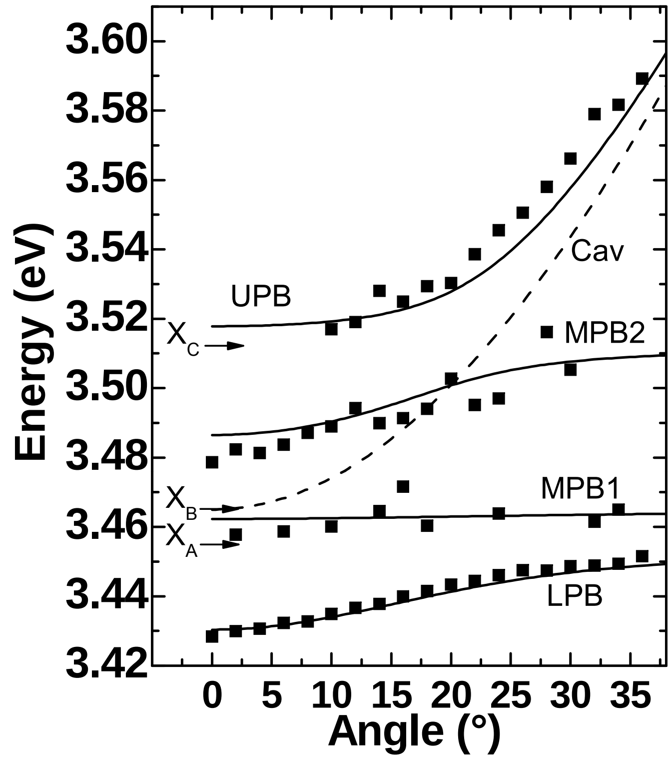
$E_x + f_{osc}$ depend on biaxial stress and polarization

$\Rightarrow$ half-MC ~ 30 kbar



B. Gil, O. Briot, PRB **55**, 2530 ('97)

microcavity dispersion

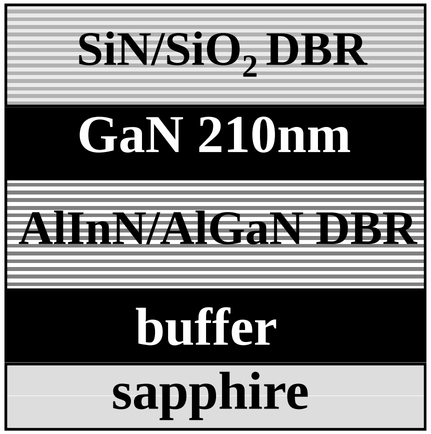


Interactions *A-C* = 25, 15, 15 meV

Cavity *Q* ~ 350

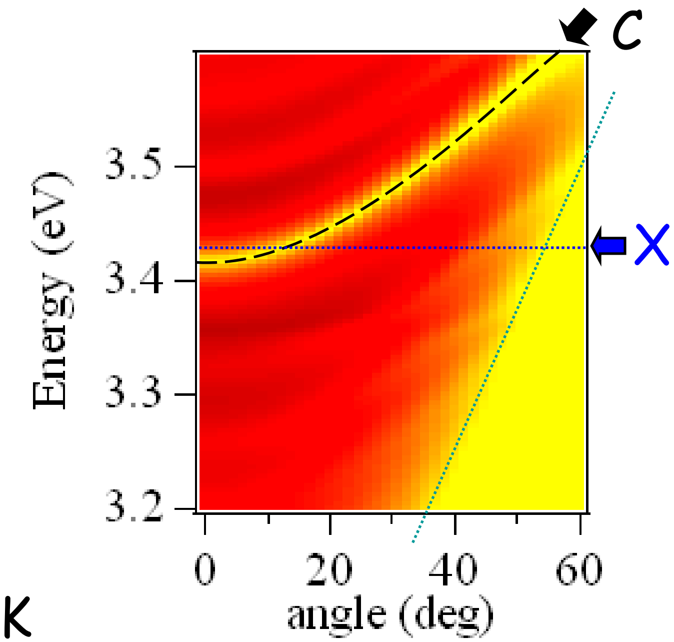
Angular dispersion

bulk GaN microcavity:



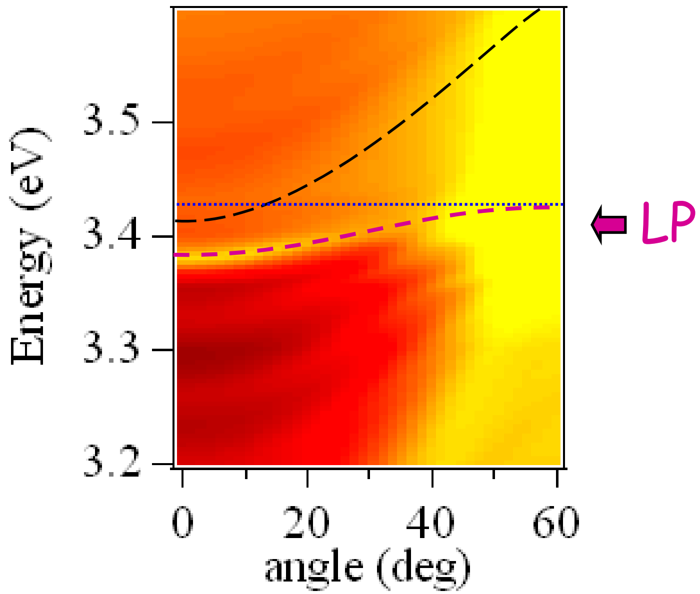
model

GaN without exciton



Weak coupling

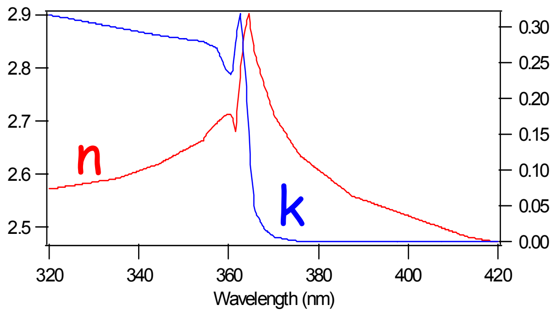
GaN with exciton



Strong coupling

- for modelling

ε of GaN at 300K



JAP **89**, 2779 (2001)

- UP broadened and attenuated
- extra lower DBR modes visible
- top DBR stopband tunes strongly

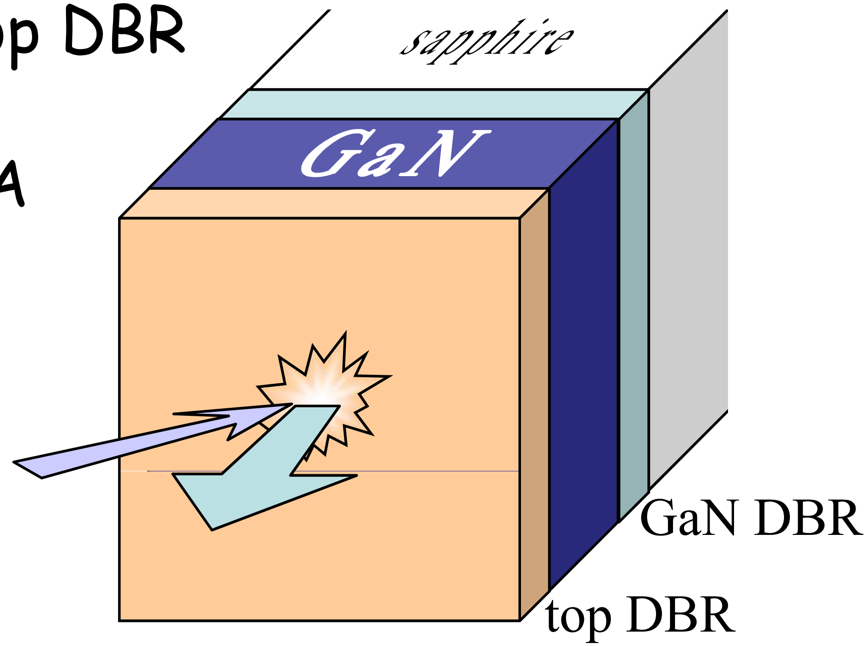
Slide 13

Σ6 The theoretical ang.disp. of the absorption of our GaN MC has been calculated using a transfer matrix formalism using the full dielectric constants for both with and without the resonant excitonic component.(emphasizing the effect of the strong coupling)
Σταύρος, 16/10/2006

Σ8 -lower is strongly flattened by the coupling
-upper is broadened and almost completely attenuated by the excitonic continuum at the same energy
-Bragg modes of the lower DBRs are visible and cross the exciton dispersion
-High angles the top DBR becomes essentially transparent allowing the direct interrogation of the exciton distribution
Σταύρος, 16/10/2006

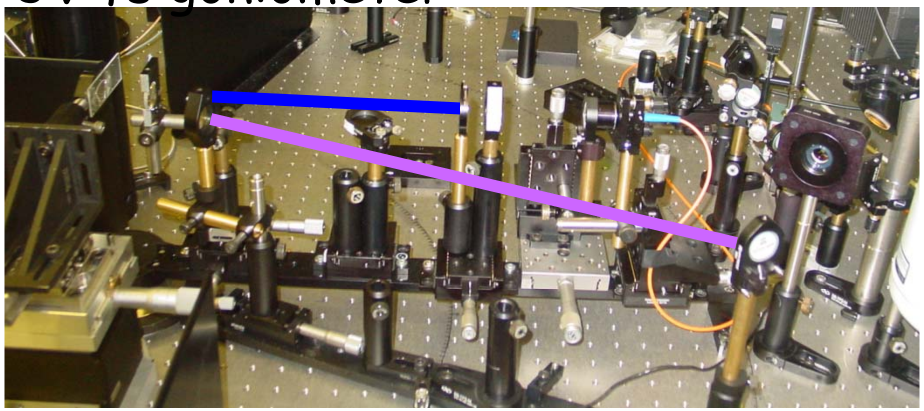
Emission experiments

- excite short pulse through top DBR
- match pump angle and energy
- UV pulses: doubled 120fs OPA
  250kHz, 270-360nm

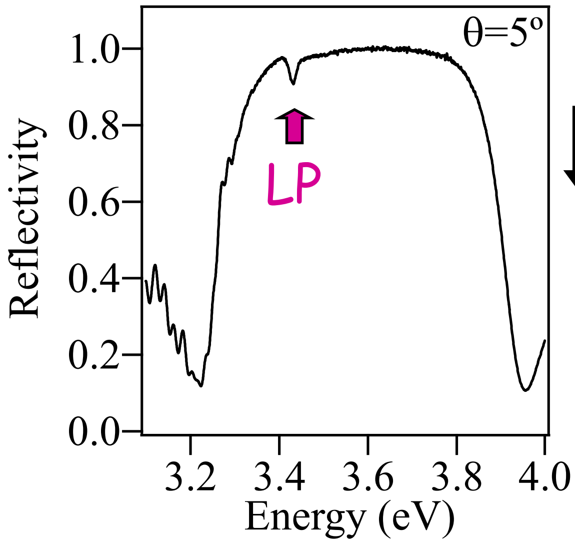


measure emission
- power
- spectra
- spatially
- angularly
- time-resolved
- polarization

UV fs goniometer

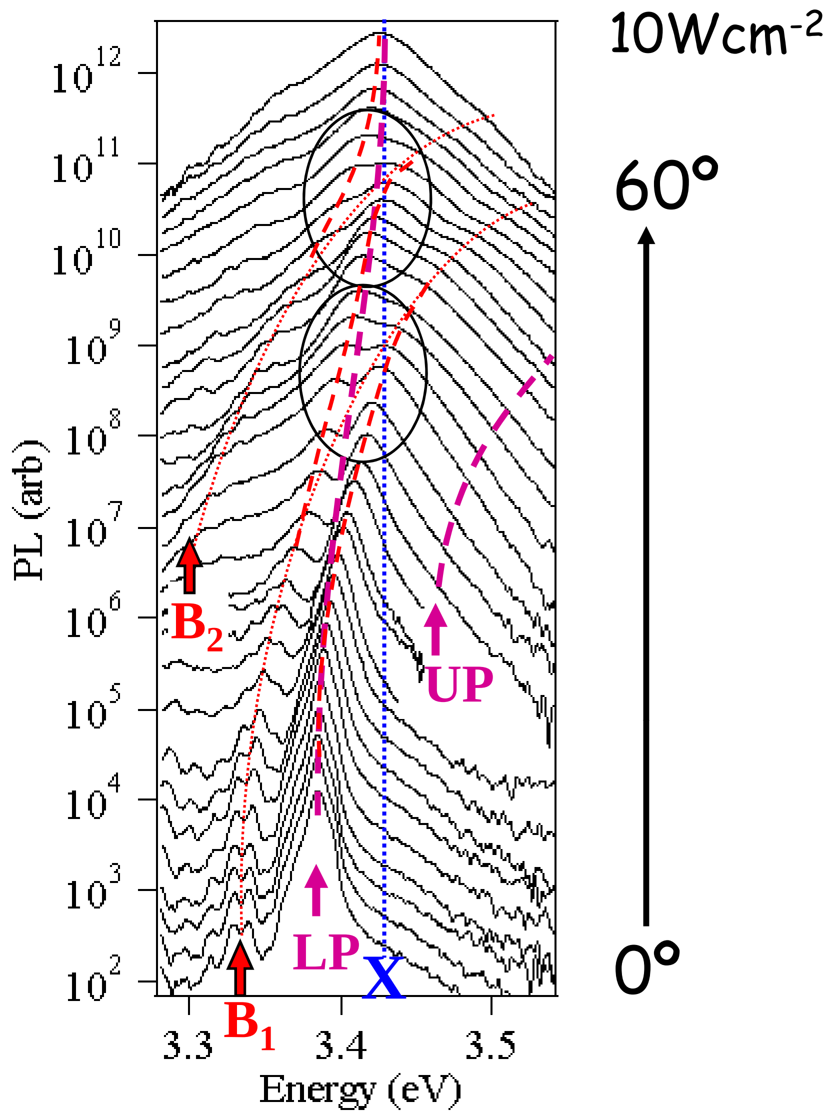


Angular dispersion II



pump at 20°

150fs tuneable

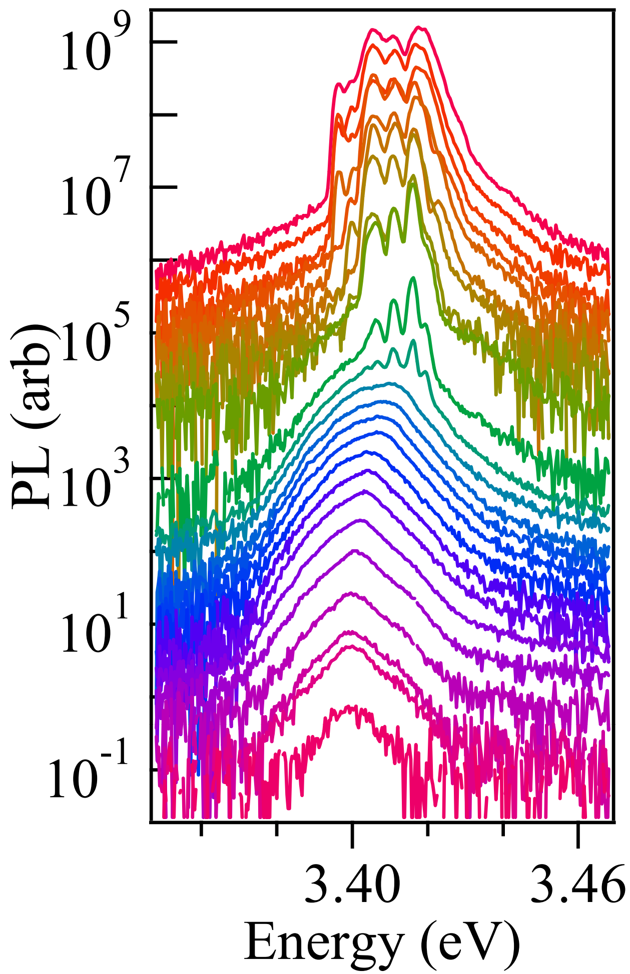


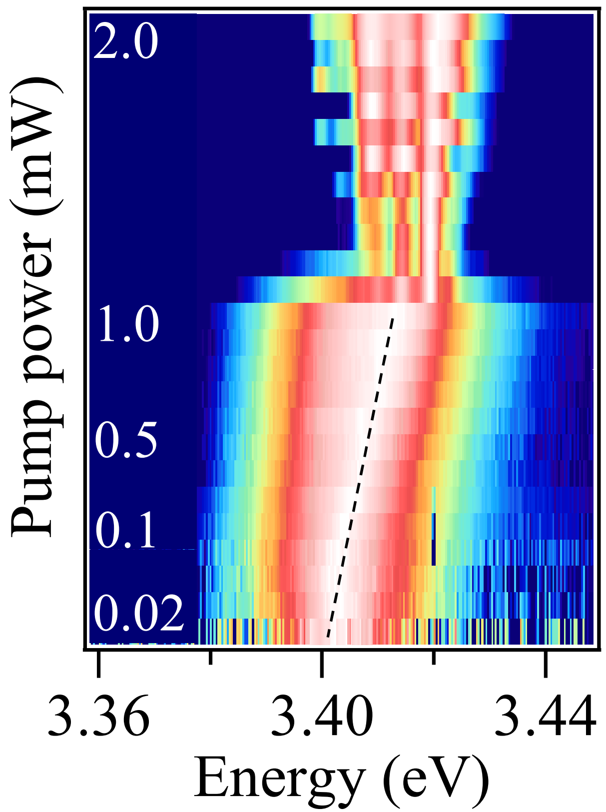
- optimise pump (ω,θ) for PL
- low power: angular dispersion
- observe:
  - lower polariton
  - Bragg modes
    - extra anti-crossings
  - exciton at high angles

sample not wedged

Power dependence I

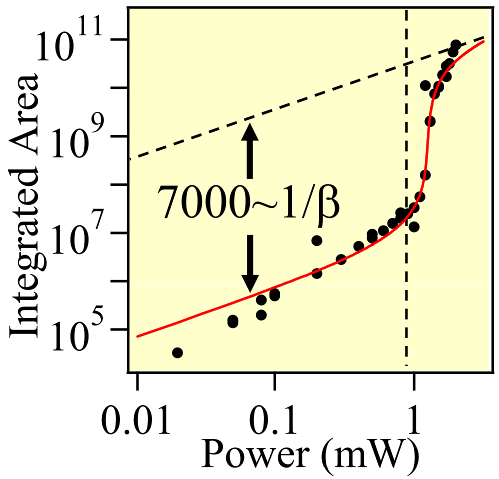
T=300K
150fs
4.14eV
5mW
250kHz

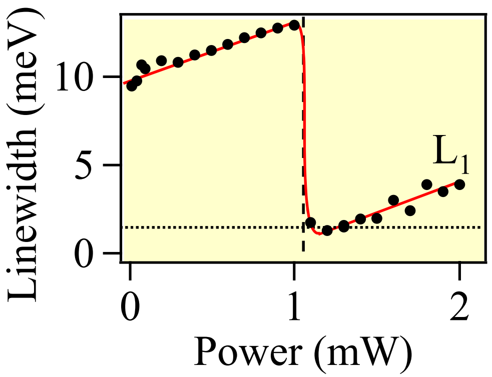




- many places on wafer, similar threshold ~ 1mW
- one/several lines: power increases by 10^4
- collect emission from entire spot (pump 60μm)

Power dependence II

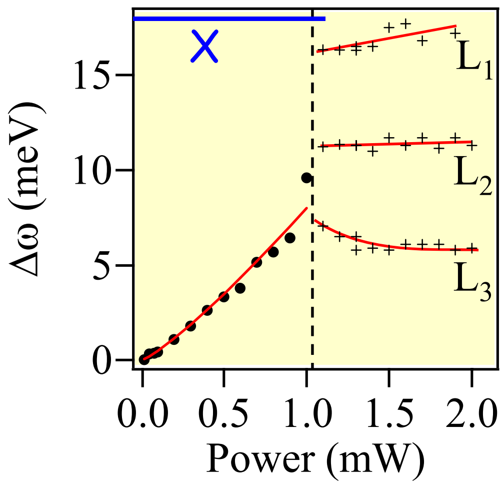




- from power dependence, extract β
- 1mW -> 3×10^{18} cm^{-3}

- below Mott density ~ 10^{19} cm^{-3} @ 300K
- vs VCSEL: Song, APL **76**, 1662 ('00)
  - best expt, InGaN QWs: ~$10^{19}cm^{-3}$
- below transparency condition
- linewidth narrows at threshold
- further increase due to X-X int$^{\underline{n}}$s

Power dependence III



- blue shift below threshold

$$\hbar\Delta\omega = 3.3\pi\, E_b\, a_B^3\, N_{3D}$$

28meV 3.5nm Haug & Koch pss(b) **82**, 531 ('77)

signature of exciton-exciton interactions

- no blue shift for empty microcavity
- previous work: always PL red shift
  - bulk GaN
  - thermal expansion
  - blue shift not seen for X

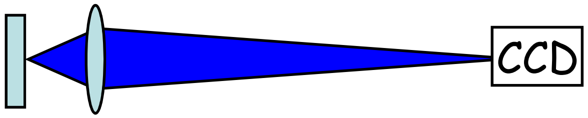
implies $N < 10^{18}$ cm^{-3}

multimode emission:
- not regularly spaced
- vary with position, power
- not $X_{A,B,C}$
- not polariton C of M quantization

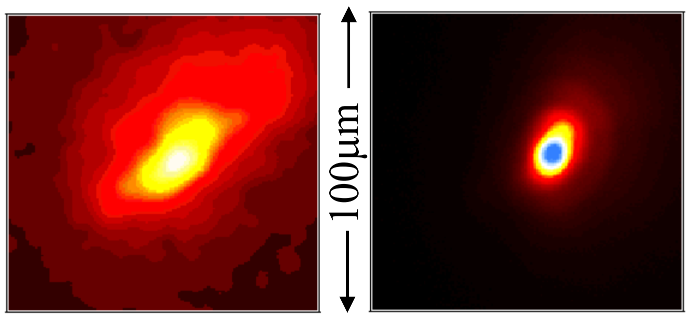
⇒ spatial localization

Spatial imaging

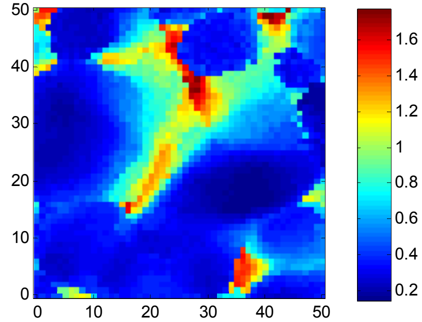
real space



below I_{th} above I_{th}



photonic disorder



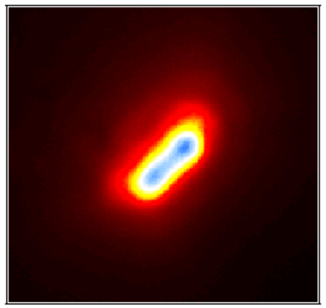
Butté et al.

- collapse at threshold
- 5µm spatial confinement gives transverse modes of 5meV

100μW from 5μm
~50% efficiency

multiple spots

real space

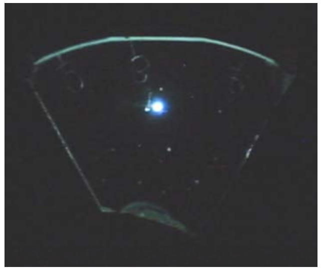


can be correlated with multiple spectra

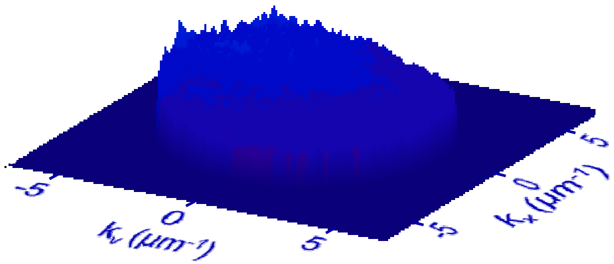
k-space imaging

- population drops into lowest state

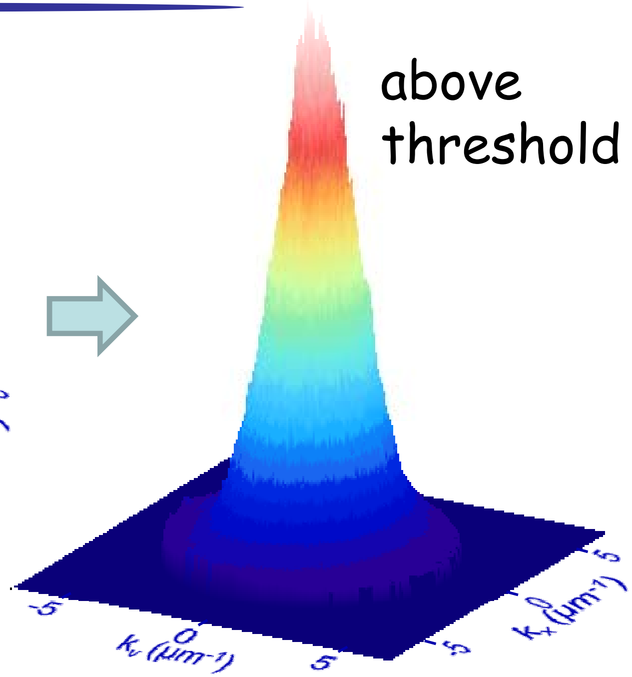
stimulated scattering



below threshold



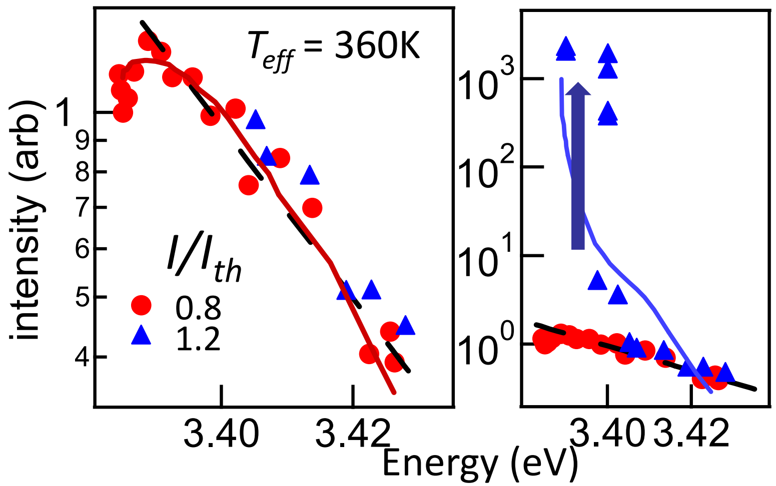
above threshold

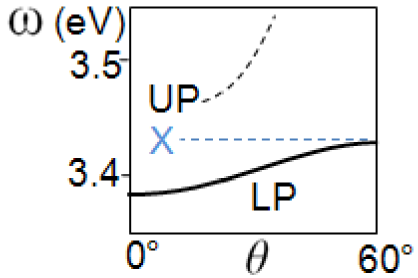


- thermalisation ?

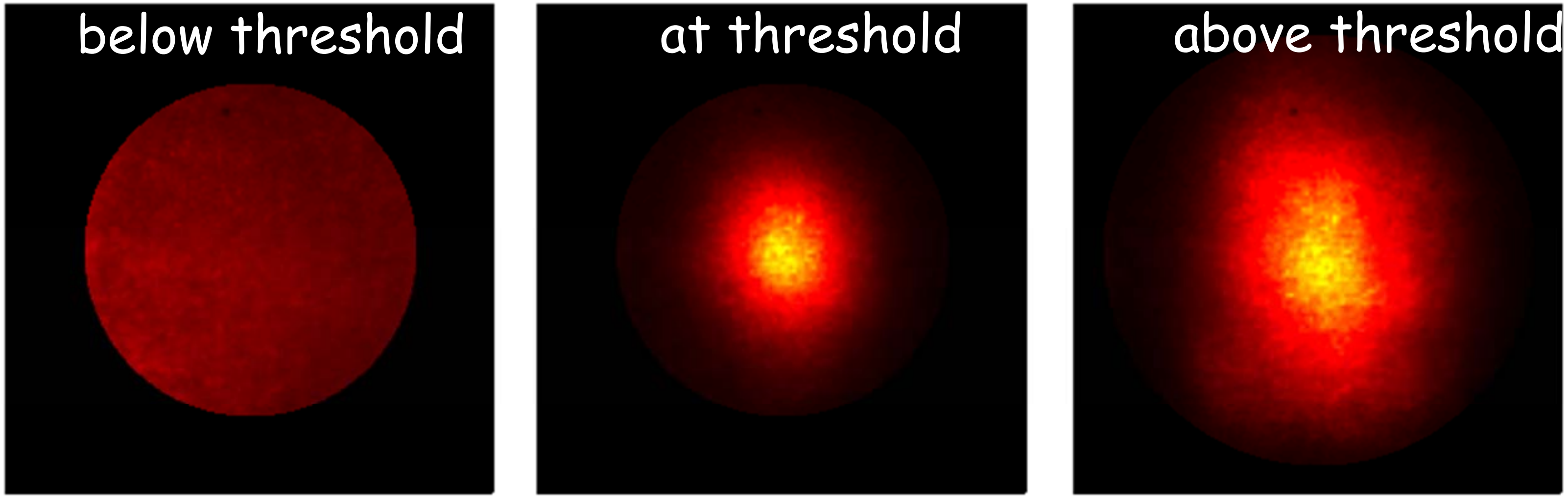
effective T ~ 360K

full kinetic model in agreement (Malpuech et al.)





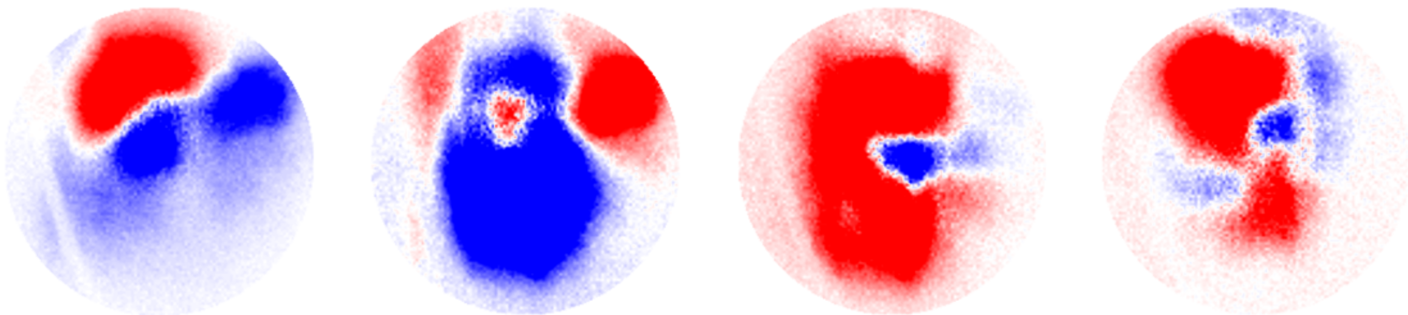
Momentum Distribution



elliptical pump spot:  

• fluctuations: 200ms apart

above threshold



• angular patches: 5°

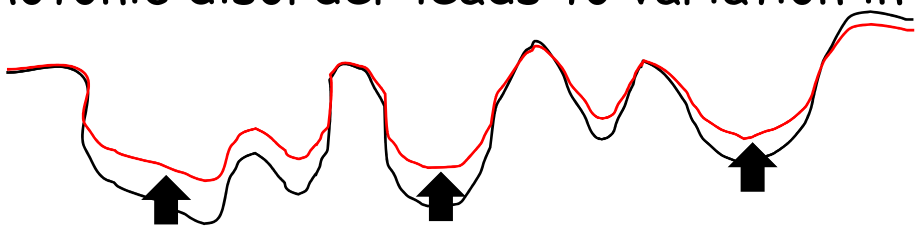
driven by what fluctuations?: incident power? spot position? cooling?

Effects of disorder potential

GaAs vs CdTe vs GaN

localisation starts to dominate

photonic disorder leads to variation in lower polariton energy



$$\hbar\Delta\omega = 3.3\pi\, E_b\, a_B^3\, N_{3D}$$

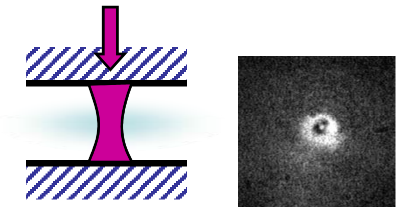
~1 at N_{Mott}

local model of polariton energy shifts

- disorder annealing by occupation
- as E_b increases, and Q decreases, less important
- is there spatially-indirect polariton scattering?

or **spatial solitons**

diffraction balanced by effective self-focussing



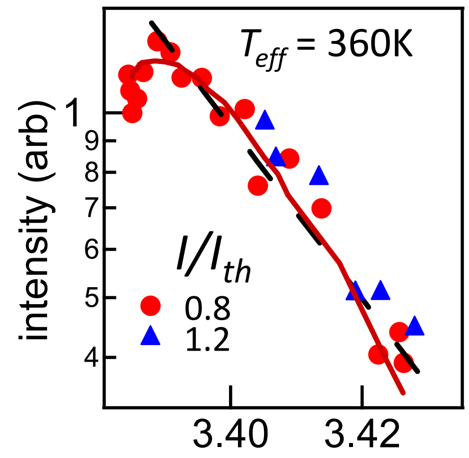
PRA **66**, 55801 (2002)

or **quasi-condensate:**

$$l_c = (4\pi a N)^{-1/2} \propto (\Delta\omega)^{-1/2}$$

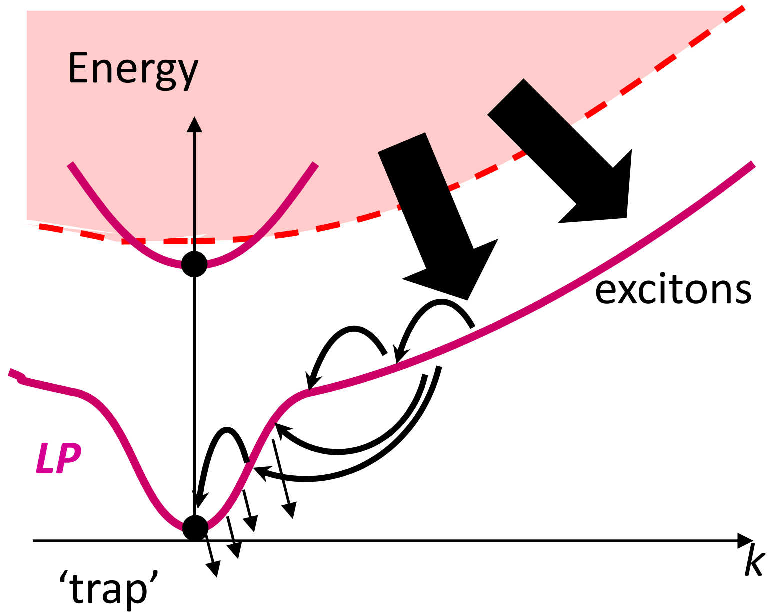
characteristic size $\propto 1/\sqrt{E_b}$

Thermalization



- many experiments seem to show Boltzman-like energy distribution

- does this prove thermalisation?



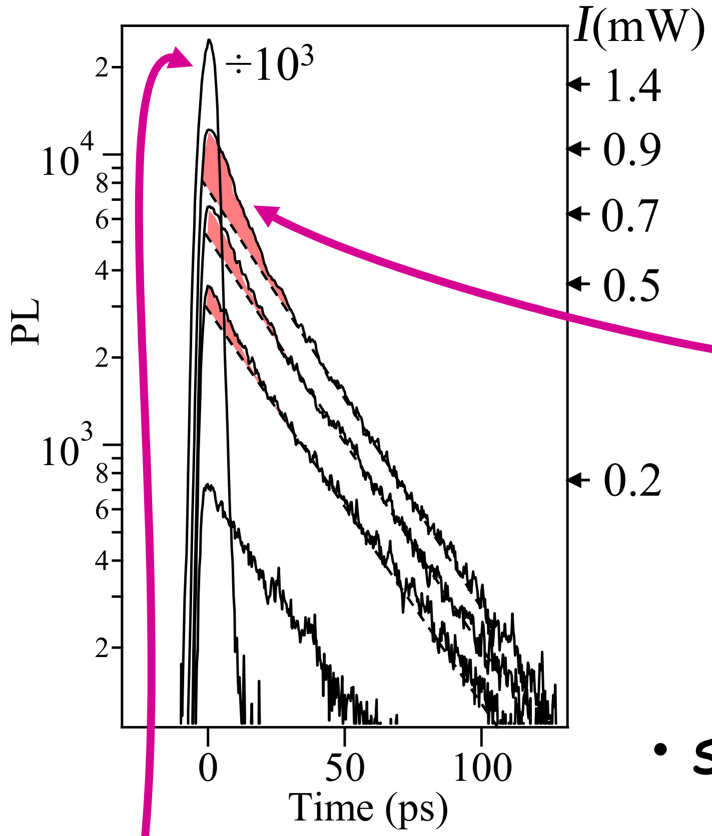
- scattering 'mimic'

- is there multiple scattering within the trap?

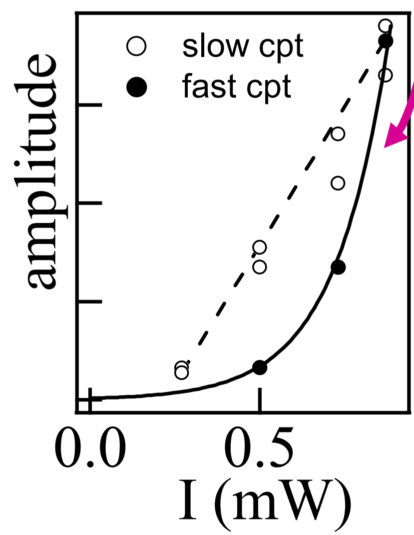
scattering events/polariton before escape? laser vs. BEC

Dynamics

- time-resolved emission on Streak camera: ~ 3ps resolution

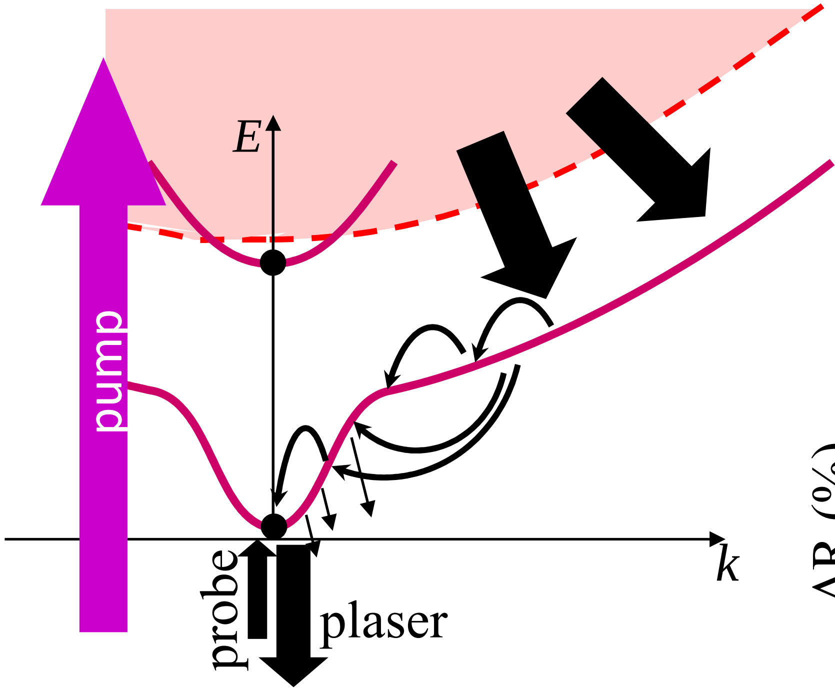


- 30ps decay,
  trapping by non-radiative defects
- near threshold:
  extra, fast component: 8ps
  exponentially rises in amplitude



- stimulated scattering near threshold
- above threshold
  < 3ps emission
- instant rise time
- dynamics not polarisation dependent

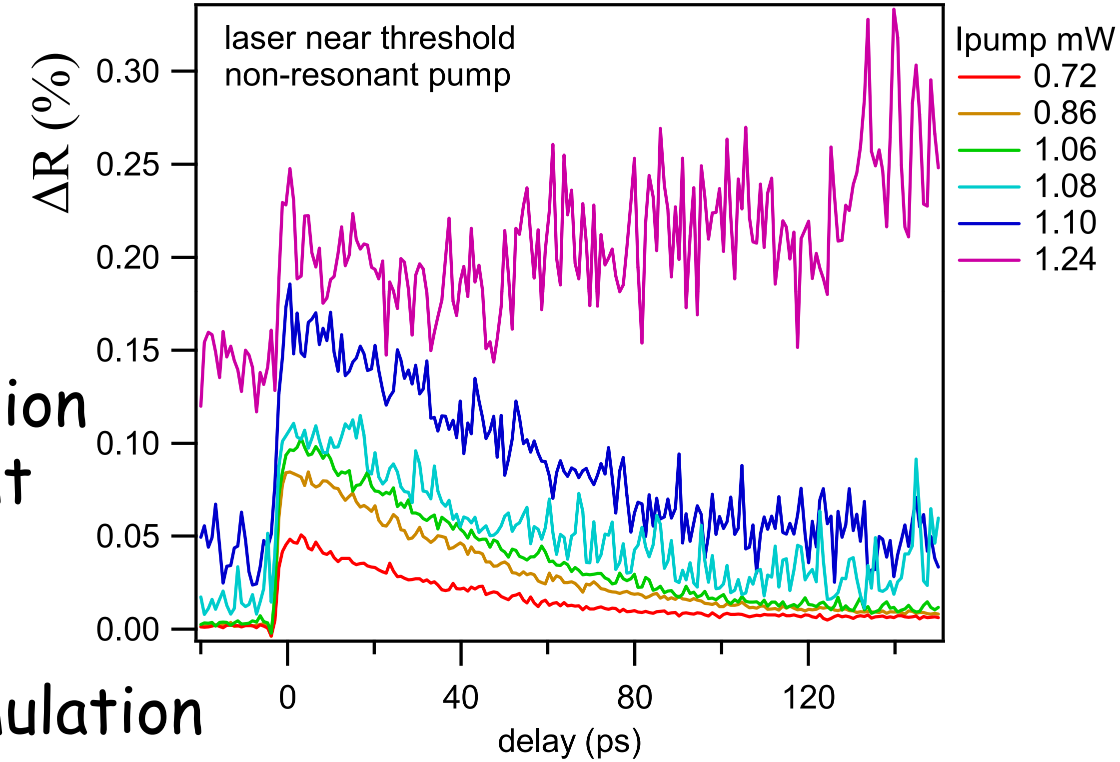
Time-resolved measurements



(I) non-resonant pump

- probe coherent emission above/below threshold

300K, 300nm pump, 363nm probe

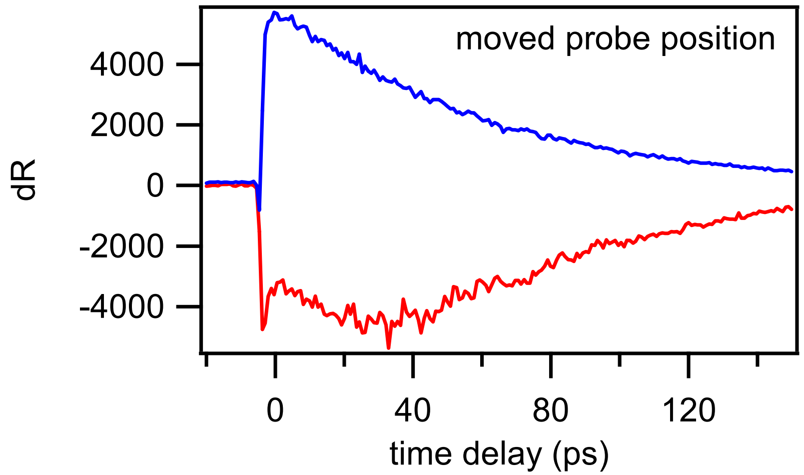


- probe increases emission
- saturation of transient above threshold
- decay close to τ_{ex}
- weak evidence of stimulation

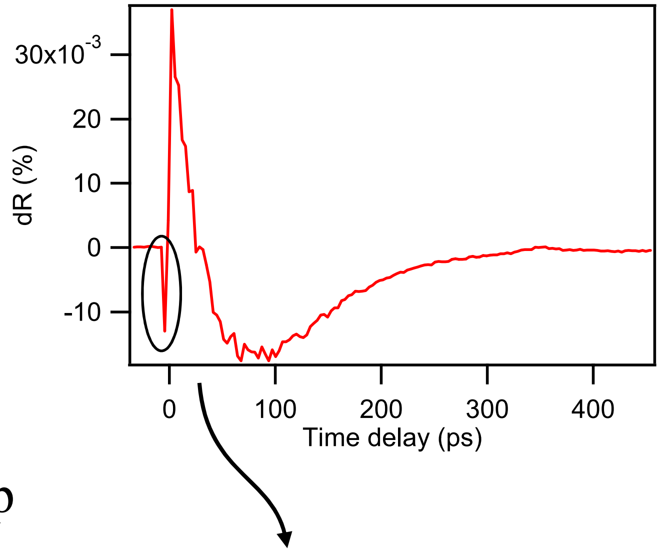
Time-resolved measurements

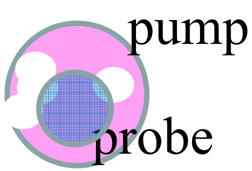
signal characteristics dependent on position/degradation

signal sign dependent on overlap

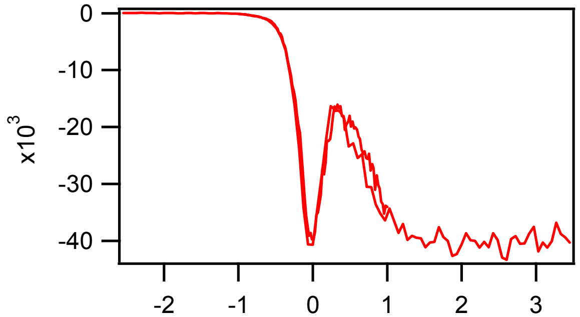


oscillatory behaviours





time resolution ~ 200fs

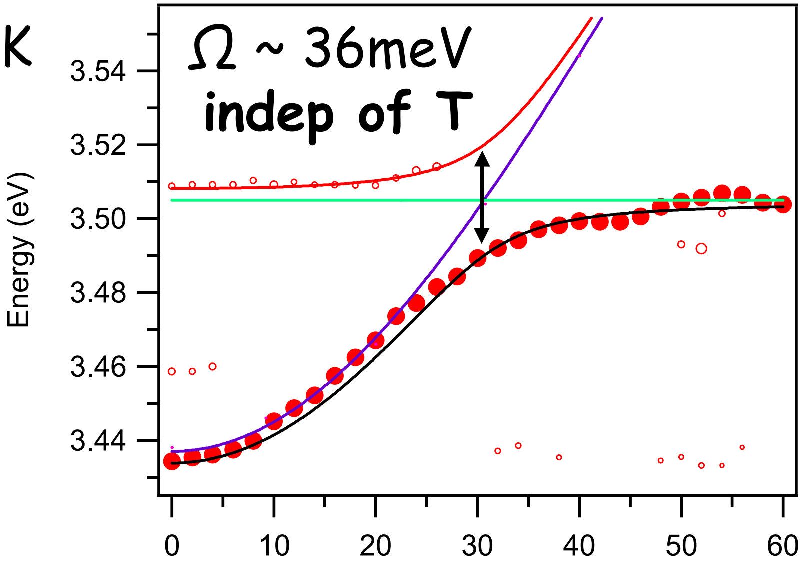


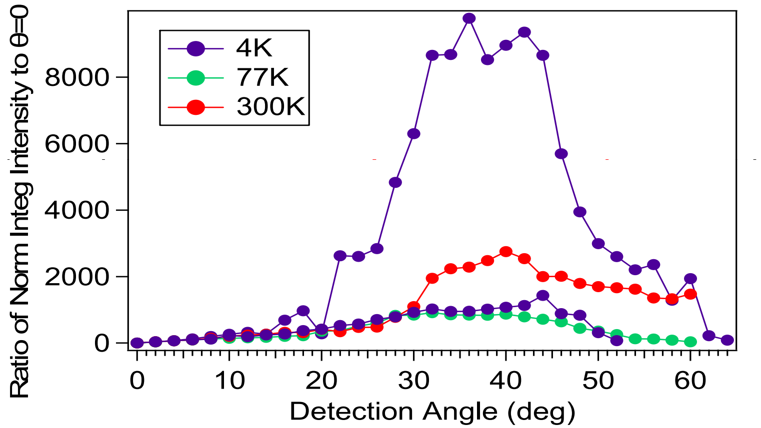
- decay times vary from 60ps-100ps
- exploring spatial behaviour
- correlated with multiline emission
- resonant stimulated scattering expts

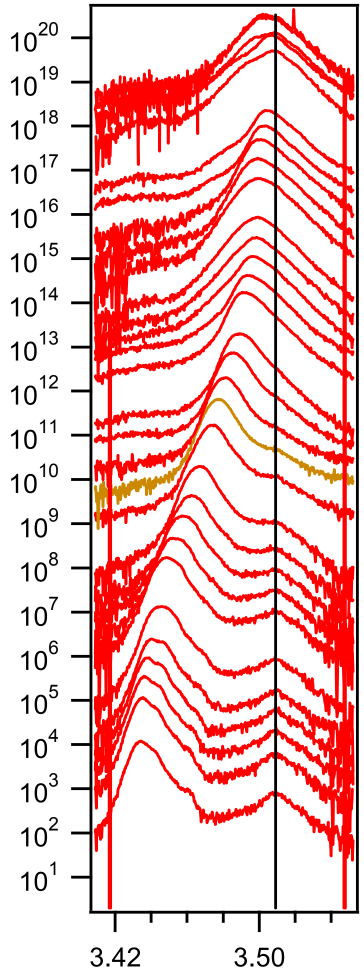
Temperature dependence

4-300K

T=77K





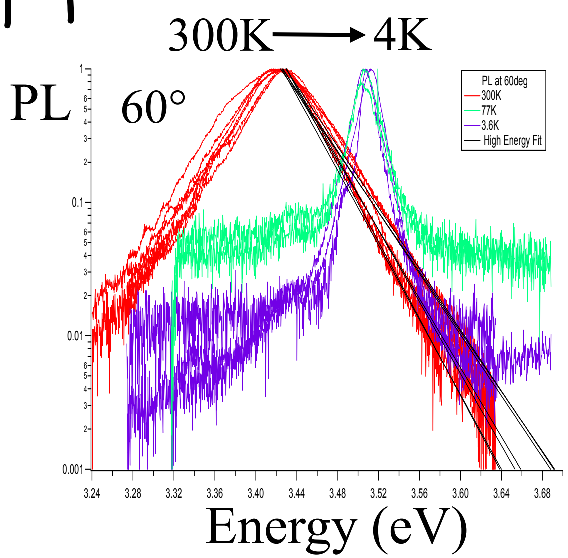


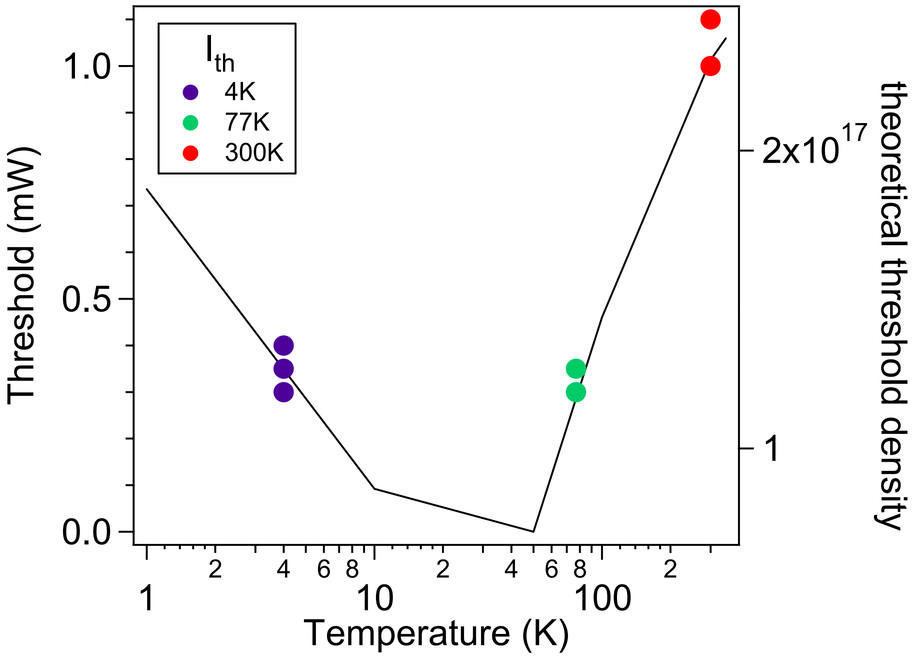
PL

- contribution of Bragg modes much weaker as T decreases
- very sensitive to sample position (disorder)
- now single shot dispersion measurements
- bottleneck sensitive to sample position and temperature

Temperature dependent plasing

- Rabi splitting ~36meV, independent of T
- PL bottleneck stronger at low T
- linewidths drop to 20meV at 4K
- but no A,B,C exciton seen in bulk





- polariton lasing 4K-300K
- threshold down to 0.1mW
- good match to theory (Malpuech/Kavokin)
- same polarisation emission

Polariton laser ?

- threshold well below standard GaN VCSELs
- coherent, directed emission
- blue shifts suggest polariton interactions

- UV mirror degradation unhelpful

- hard to exactly know polariton density
- spatial mode very localised ⇨ spatial coherence not helpful nor imaging
- pulsed excitation only ⇨ conventional $g^{(1)}(\tau)$ not helpful

  picks random phase each time to develop coherent state
  no phase correlation between successive pulses

clearly aim towards CW operation ➡ next generation samples
positive detuning
new pump-probe expts

Polarization Signatures

- <u>integrated</u> coherent emission is **UNPOLARIZED** !

  unlike all previous lasers

- <u>all</u> VCSELs give linear polarization
  (but can flip with increasing I_p)

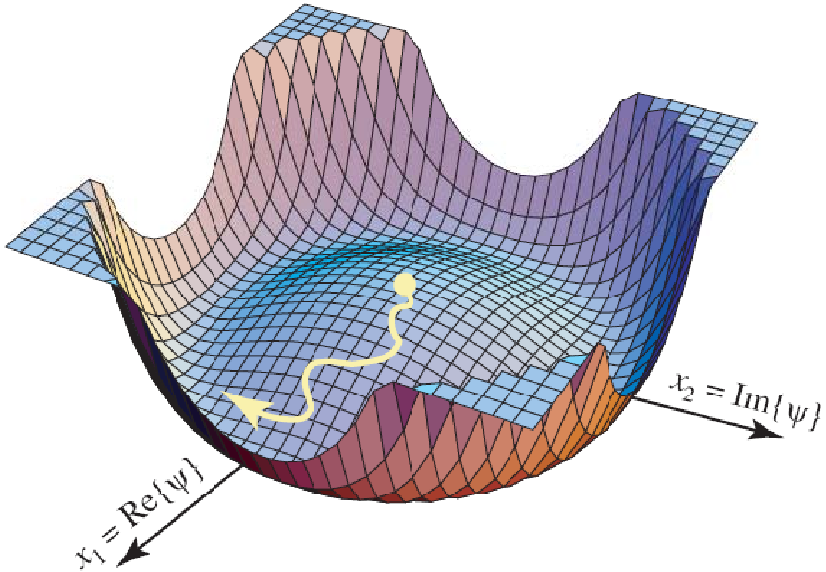
  including InGaN VCSELs made with same mirror DBRs

  and GaInAsP bulk VCSELs
  [Superl. & Micro. **32**, 103 (2002)]

Spontaneous polarisation

spinor order parameter

$$\psi(\underline{r}) = \begin{bmatrix} \psi_\uparrow(\underline{r}) \\ \psi_\downarrow(\underline{r}) \end{bmatrix}$$



polarisation degree $\rho = |\psi^2|/N_0$

along different axes, tracks spinor

effective potential

$$U = \{[W_{out}(t) - W_{in}(t)]\,|\psi|^2 + \alpha|\psi|^2\}/4,$$

pumping (↓ $W_{in}(t)$); radiative (↑ $W_{out}(t)$); interactions (↑ $\alpha|\psi|^2$)

below threshold: $|\psi| = 0$

above threshold: $W_{in} > W_{out}$ ⇨ finite $|\psi|$

Polarisation coherence

spin degeneracy lifted by fluctuations in spin-up/down density

classically, fluctuations wash out as density increases
so zero average spin

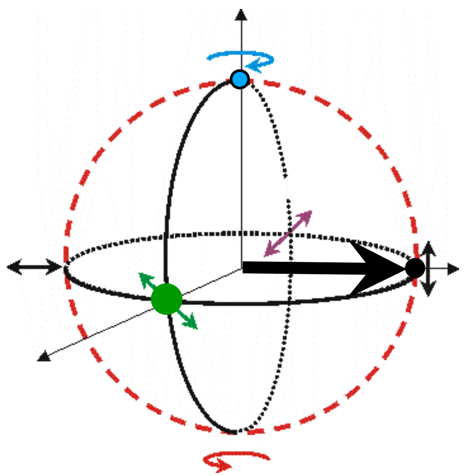
but stimulated scattering reinforces populations

$$p_{\uparrow\downarrow} = \frac{N_{0\uparrow\downarrow} + 1}{N_{0\uparrow} + N_{0\downarrow} + 1}$$

in-scattering

$$\Rightarrow \langle|\rho_c|\rangle = \frac{2 + N_0}{2 + 2N_0}$$

net <spin> $\langle|\rho_{tot}|\rangle \to \frac{1}{2}$

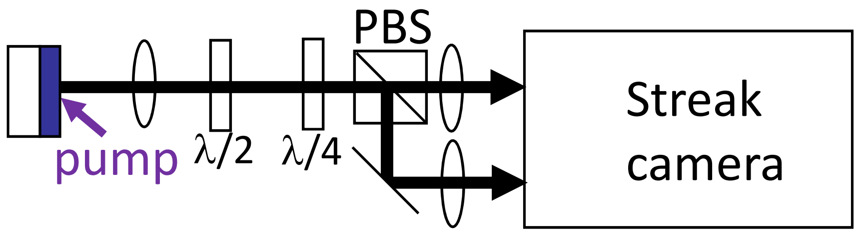


pseudo-spin dynamics also affected by
precession on Poincare sphere

Laussy et al., PRB **73**, 35315 ('06)

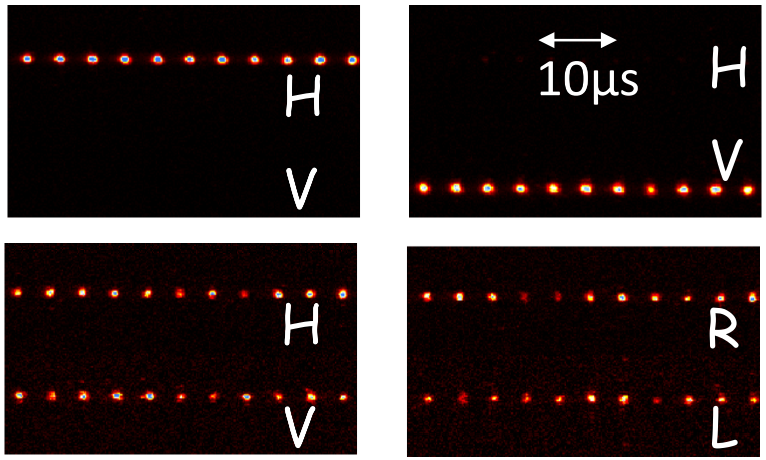
Polarization Dynamics

- single-shot Streak camera



time →

pump laser

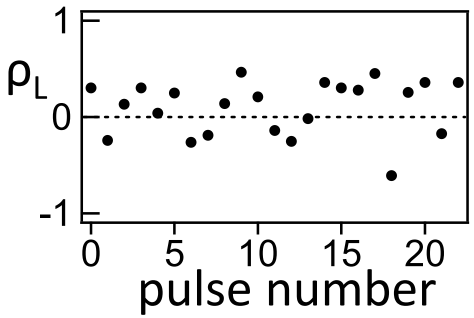


VCSEL
fixed polarised

pulsed emission

microcavity
random polarised

$$\rho_L = \frac{I_H - I_V}{I_H + I_V}$$

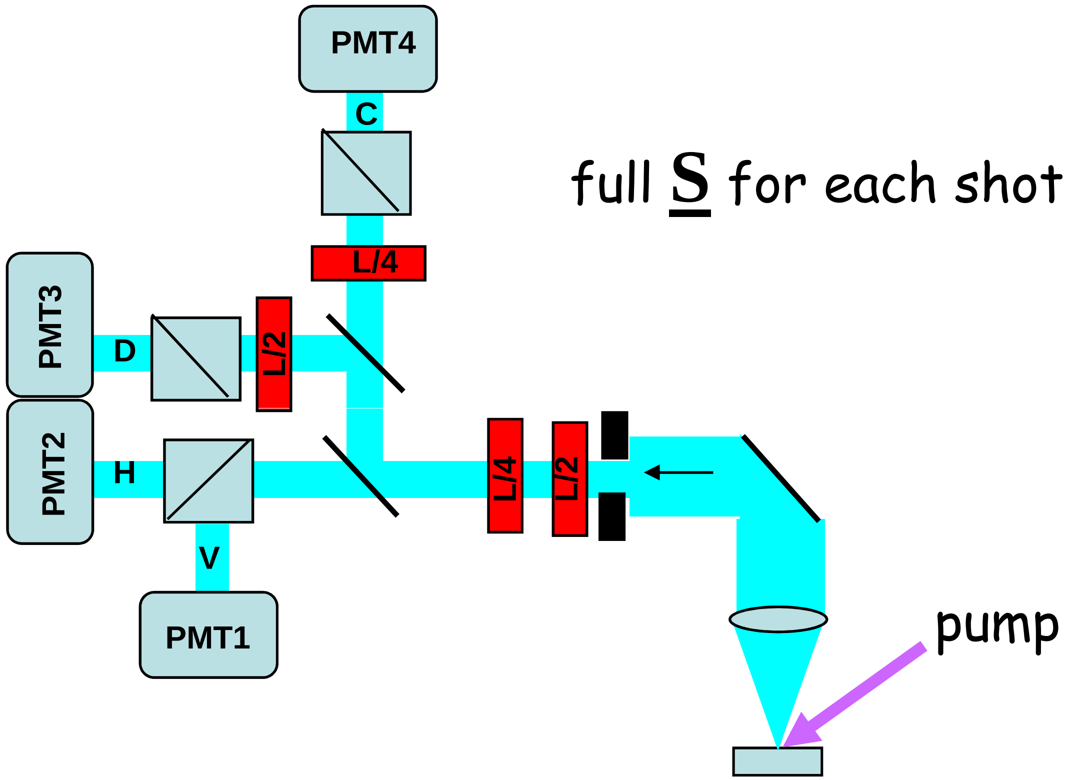


- individual pulses are partially polarised
- spin builds up from noise each time

Full Polarization Measure

- need to measure polarisation of each pulse

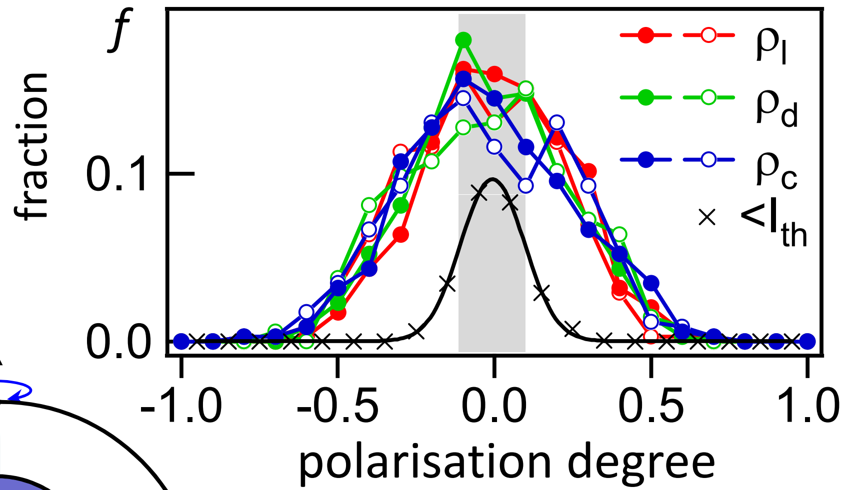
  Simultaneous full Stokes parameters



remove mis-balances etc. using full set of waveplate alignments
subtle to get measurement accurate

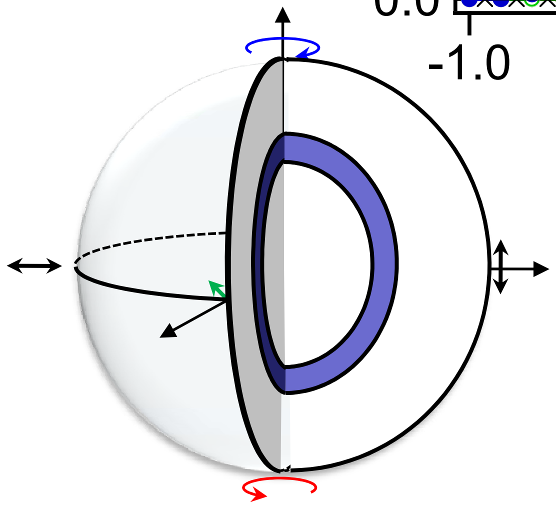
Spontaneous Polarisation

PRL ('08)

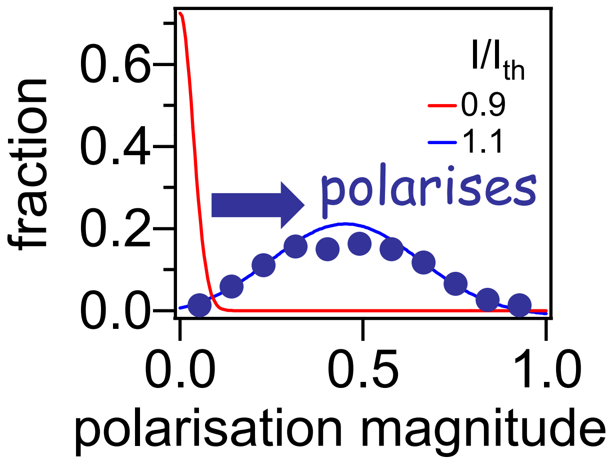


each axis 27% polarised
total polarisation 47%

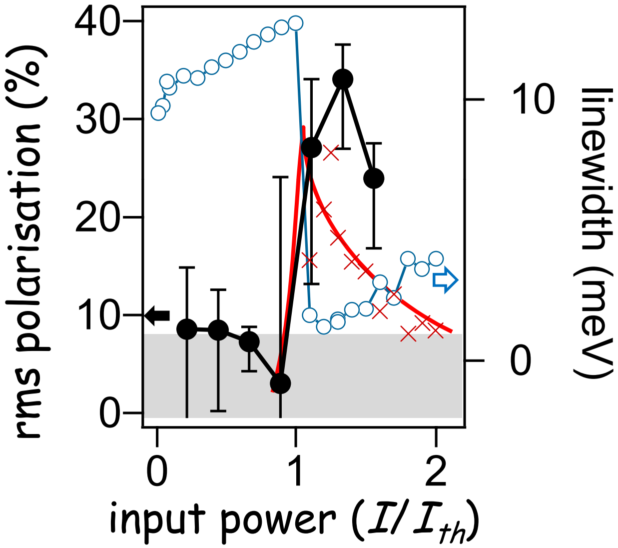
theory predicts 50%



shell of polarisation



spin coherence model

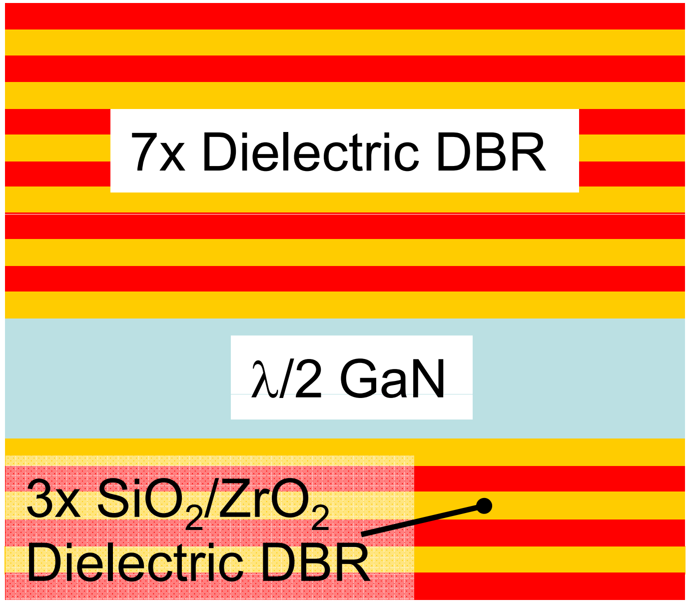


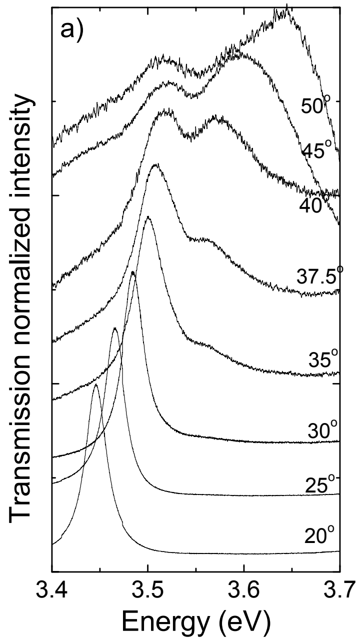
- elliptical polarisation each shot, uncorrelated successively
- extremely unusual coherent emitter

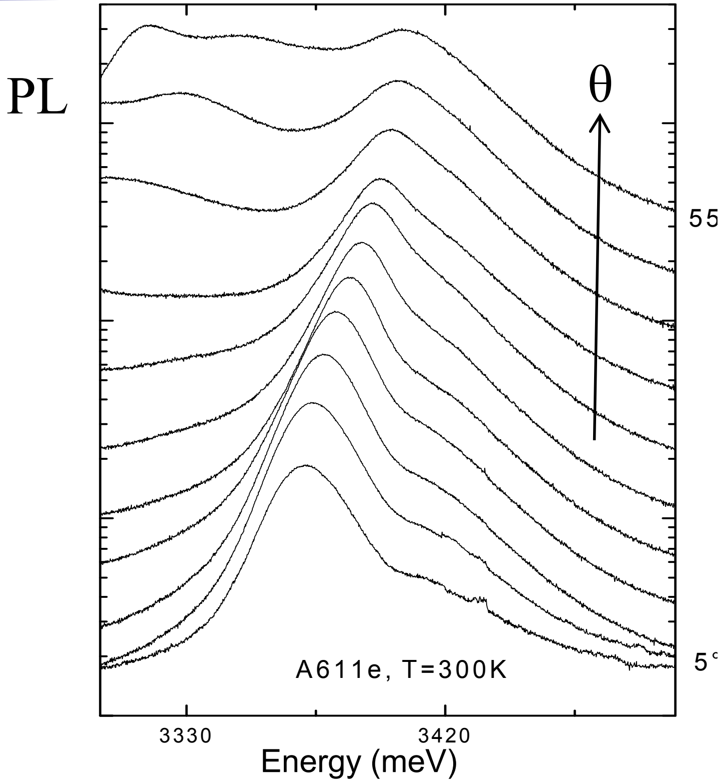
Rob Martin *et al.*
(Strathclyde/ CHREA)

other routes to 300K

GaN + double dielectric DBR

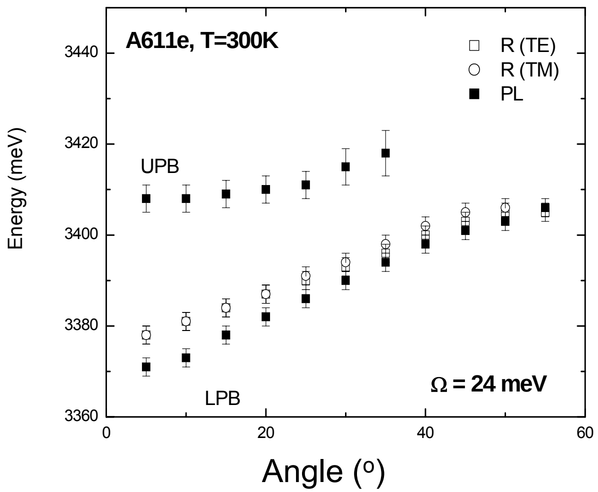




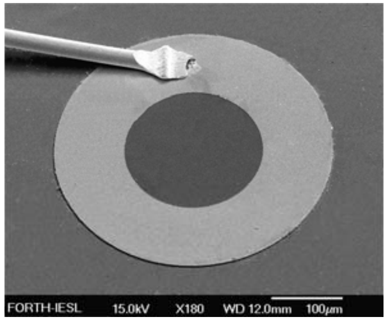


Si: wet etch & RIE thinning

- can observe strong coupling in R,T, PL
  good at 5K
  indicative at 300K
- but currently cavity Q too low



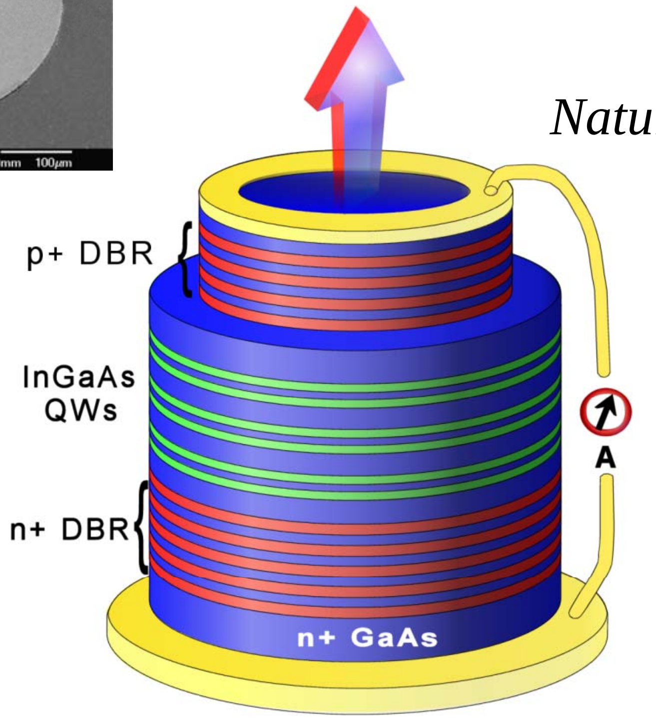
K. Bejtka *et al.*, *APL* **92** 241105 (2008)

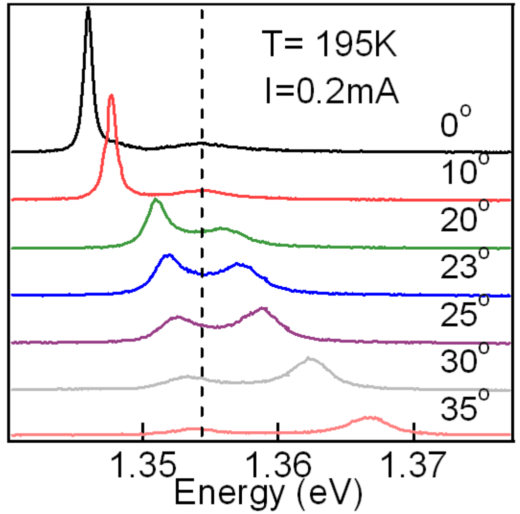


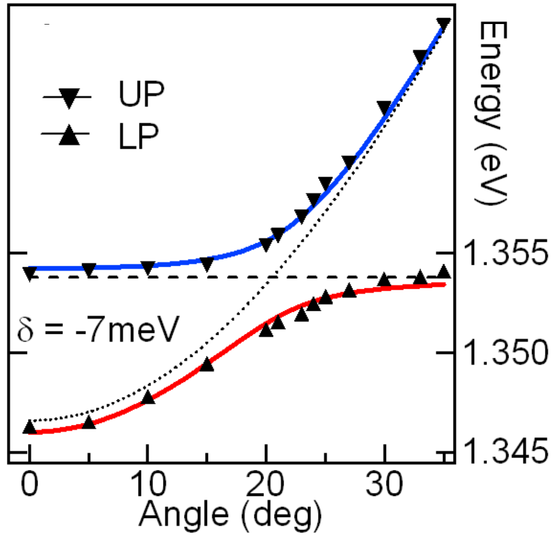
Polariton LED

Pavlos Savvidis *et al.*

Nature (‘08)







- Designed to operate at >200K
- MQWs enhance Rabi splitting
- **now** demonstrated stimulated scattering

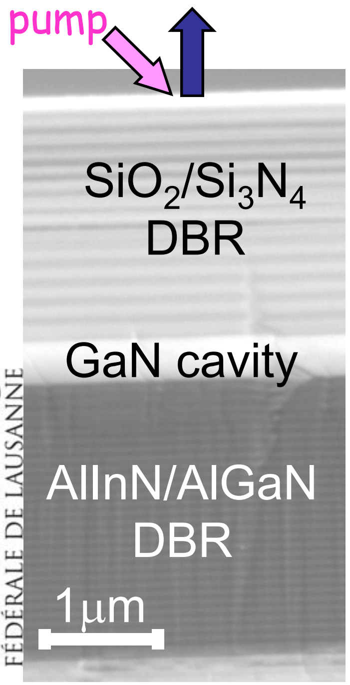
GaN polariton lasing

µphotonics

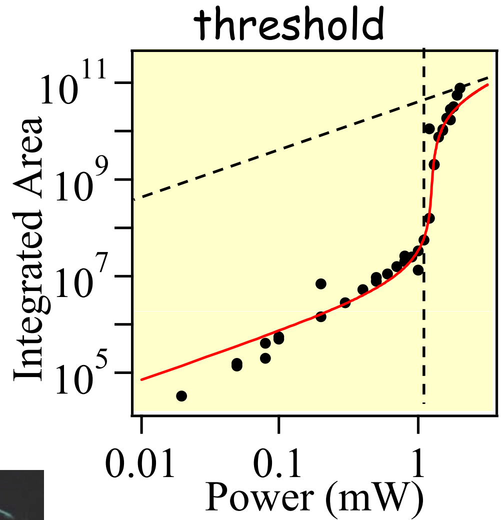
UNIVERSITY OF CAMBRIDGE

np.phy.cam.ac.uk

ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

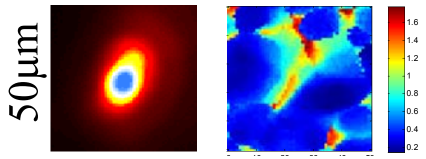


angle resolved emission
pulsed UV excitation



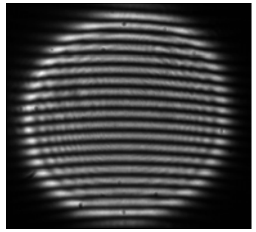
GaN > GaAs

300K 50K

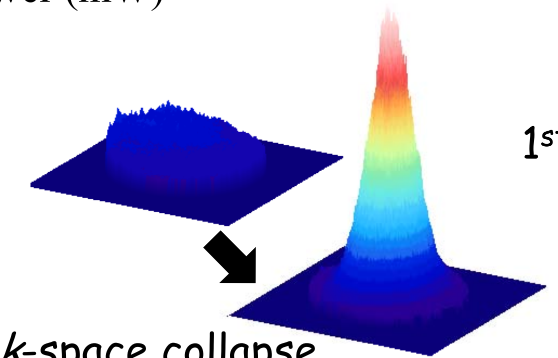


photonic disorder

spatially localised

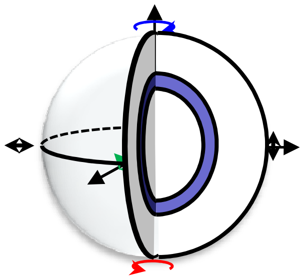


1st order coherence



k-space collapse

- ultralow threshold: 0.1mW
- linewidth narrowing
- coherent emission
- polariton lasing/BEC
- ultrafast dynamics



spontaneous polarization

PRL in press (2008)
PRL **98**, 126405 (2007)
Nature N&V, Science