

Coherence in semiconductor nanostructures

Part III: Spontaneous collective coherence & superradiance in ensembles of excitons & polaritons

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UNIWERSYTET
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ZINTEGROWANY
PROGRAM ROZWOJU

Equipe mixte CEA-CNRS "Nanophysique et semiconducteurs"
Institut Néel - CNRS
Grenoble France

Warsaw University, October-December 2020

Outline

1 Condensation Phenomenon

2 Why Polariton Condensation ?

3 Demonstration of the Polariton Condensation

- Massive Occupation (N, T)
- Thermalization
- Long Range Spatial Coherence

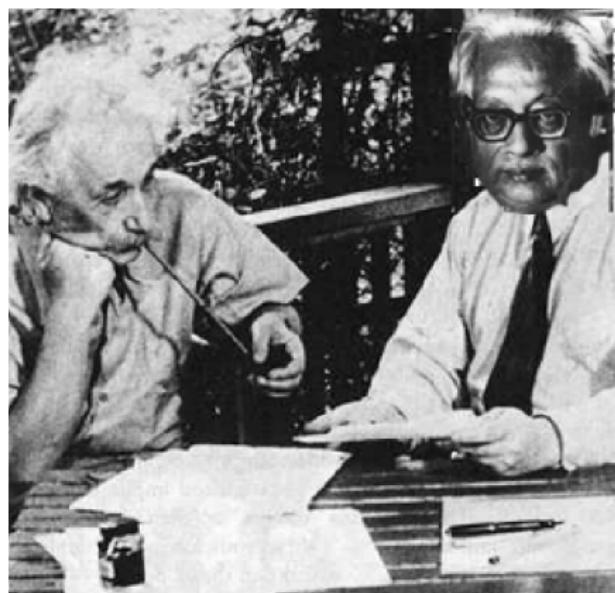
4 Discussion

- Single State
- Quantum State

Condensation Phenomenon

Idea, 1924: The theory is pretty, but is there also some truth to it?

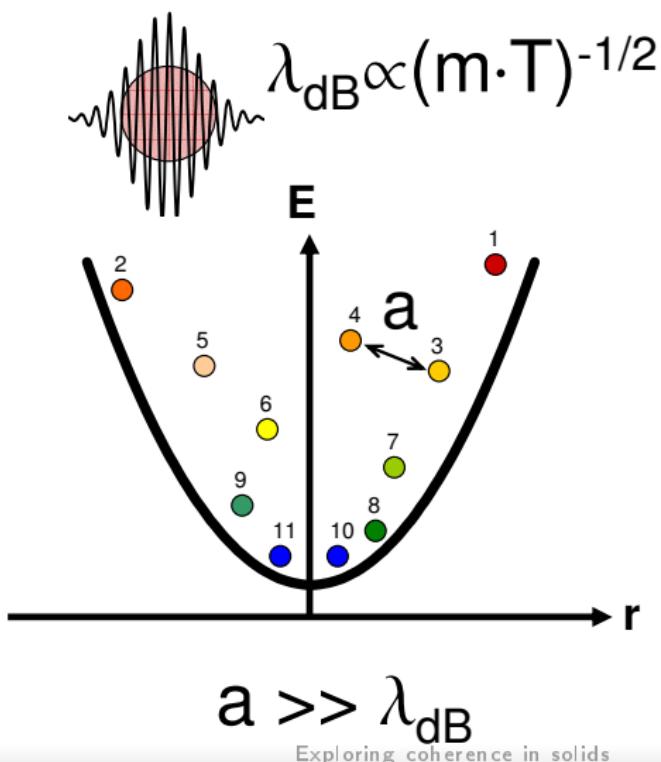
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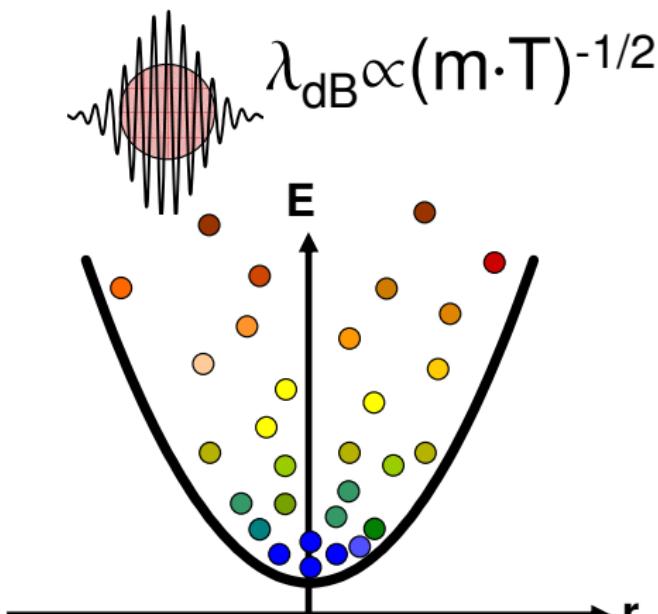
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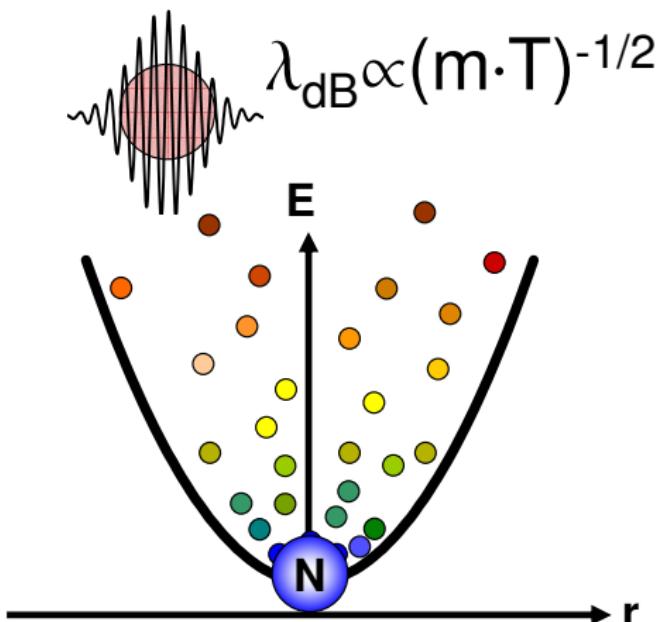
$$a \sim \lambda_{dB}$$

Exploring coherence in solids

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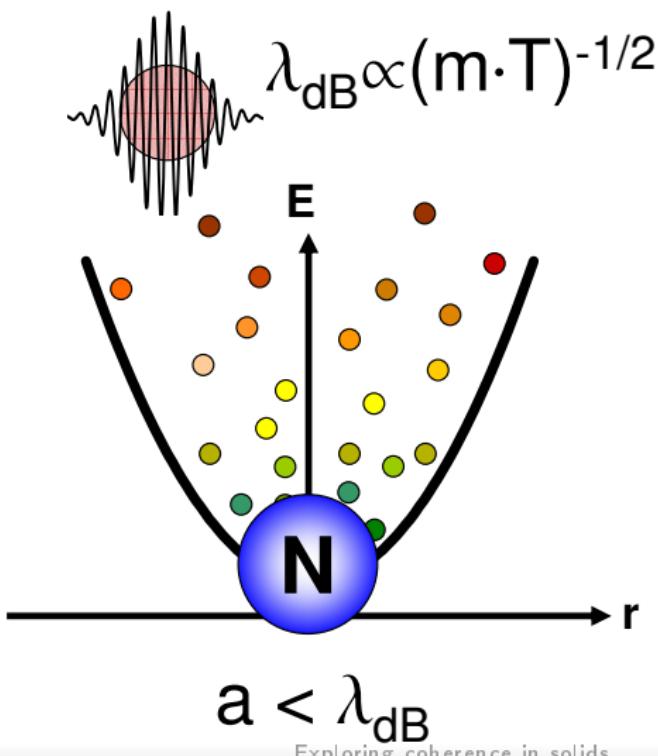
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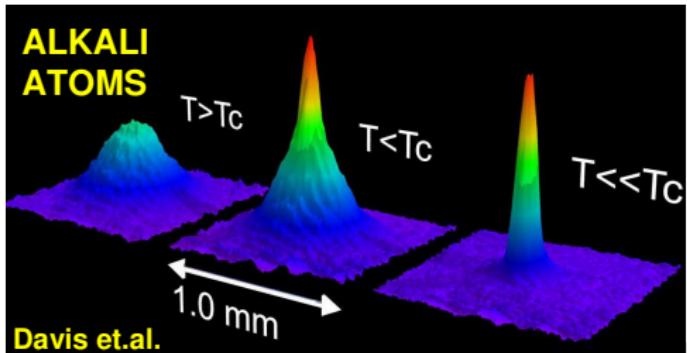
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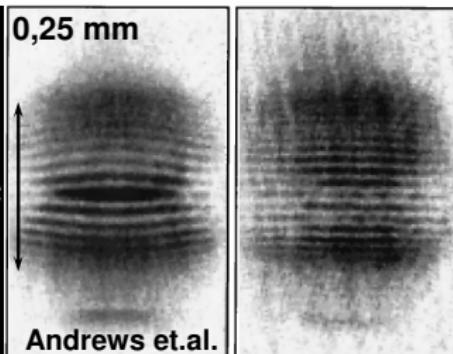
Bose-Einstein Condensation

First Achievement: Boulder, MIT - 1995

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- Long Range Coherence Interference

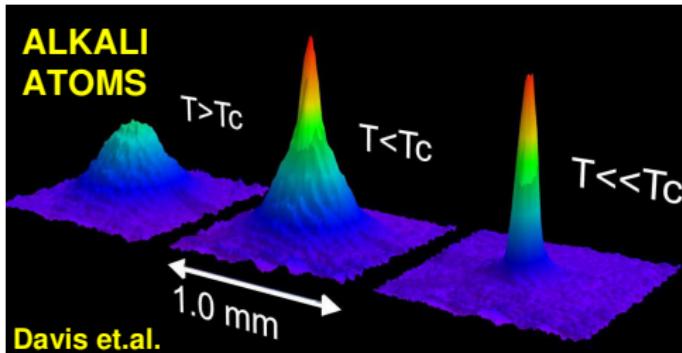


- Atoms: $(T_c, N_c) \sim (10^{-7} K, 10^8 cm^{-2})$
- Polaritons: $\sim (20 K, 5 \times 10^8 cm^{-2})$
- $m_{\text{Polariton}} \sim 10^{-8} m_{\text{Atom}}$**

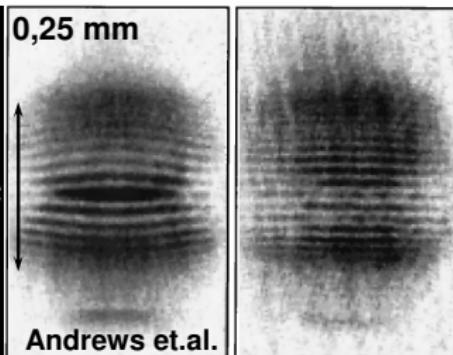
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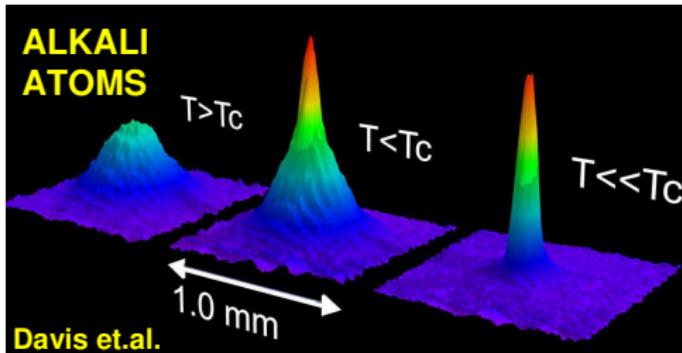
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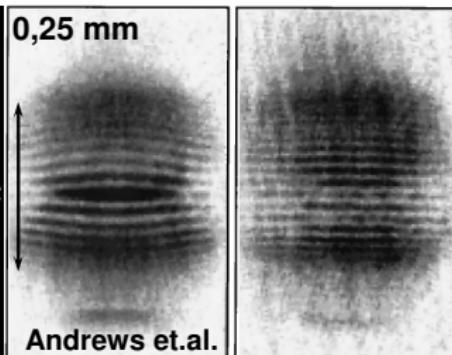
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Davis et.al.

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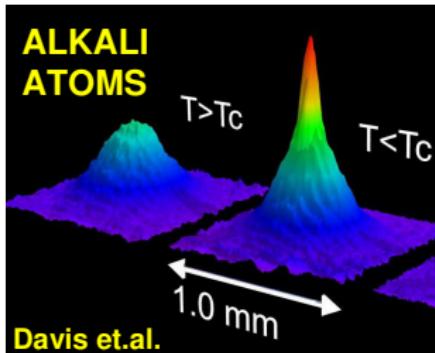
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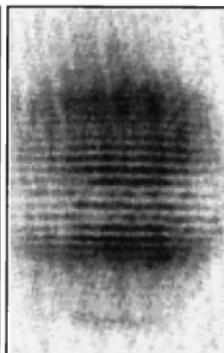
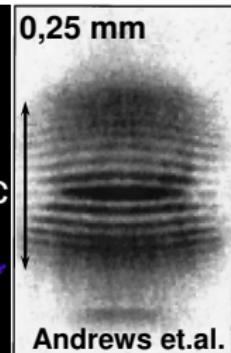
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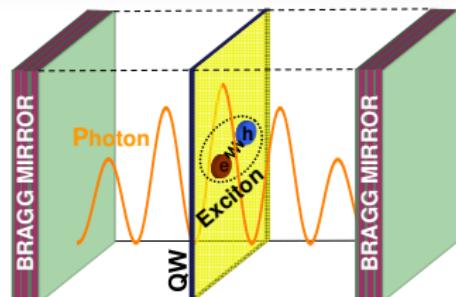
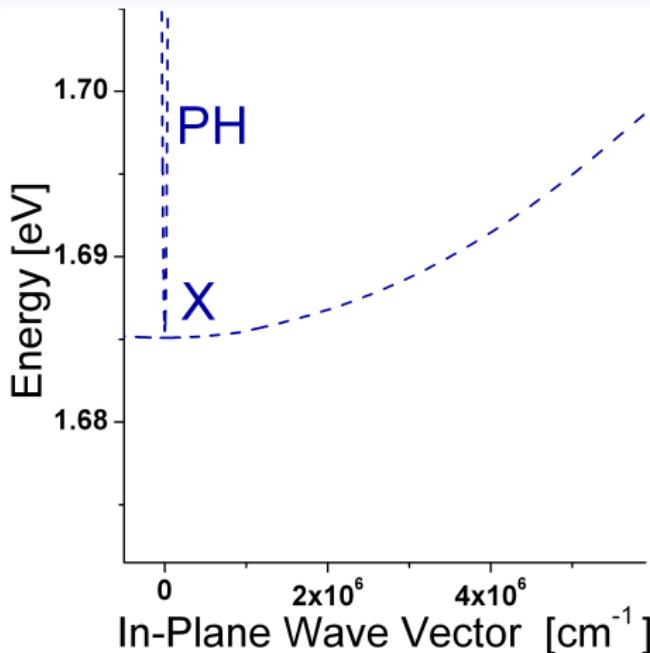
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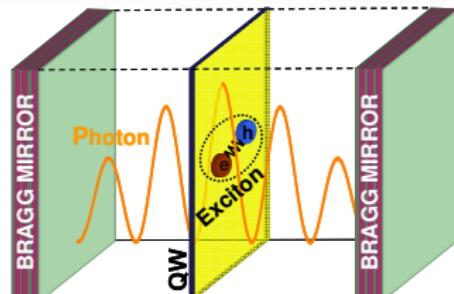
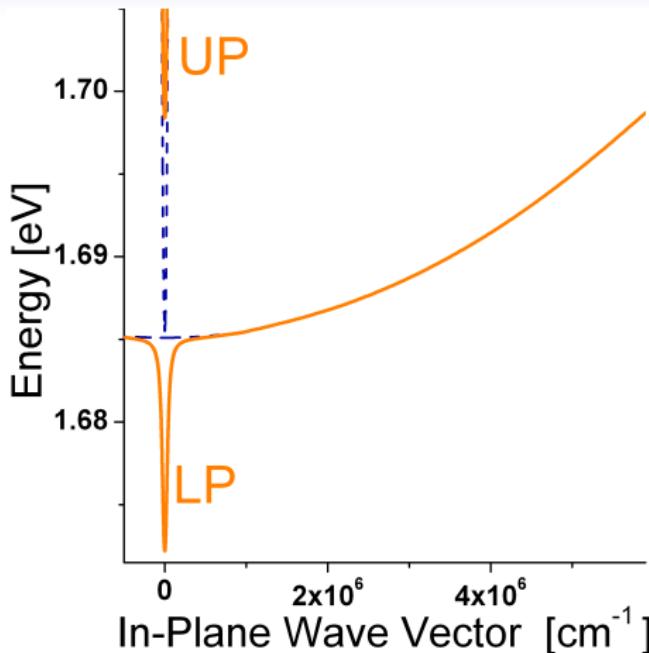


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Momentum Conservation

$$k_{||} = \frac{E}{\hbar c} \sin \theta_{ext}$$

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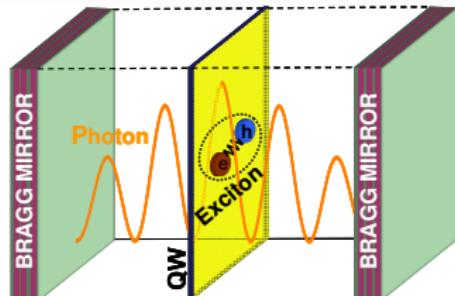
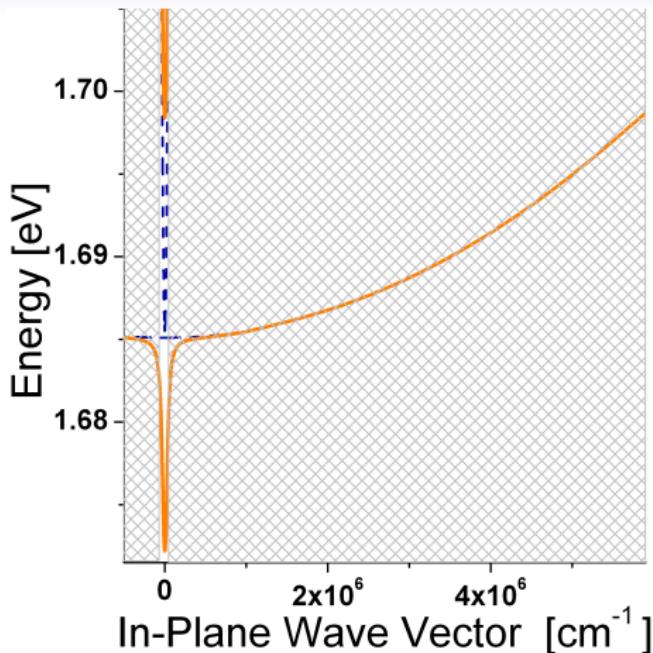
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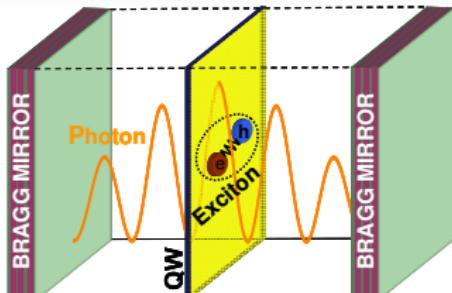
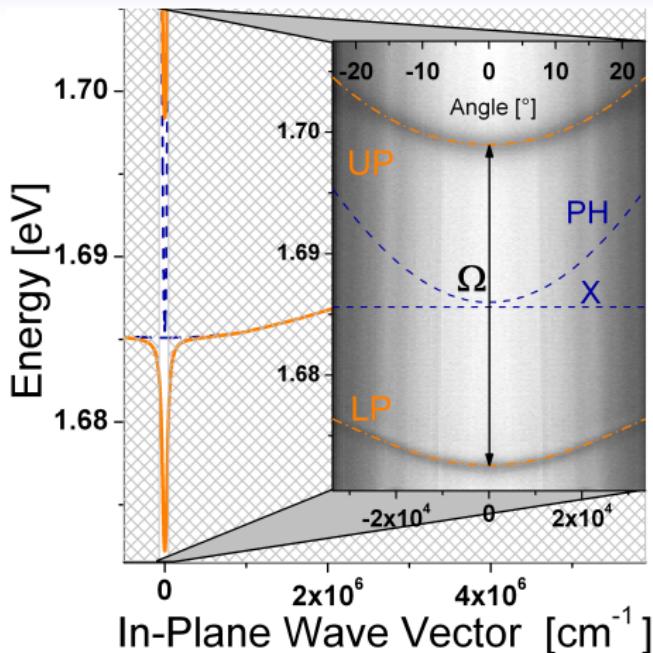
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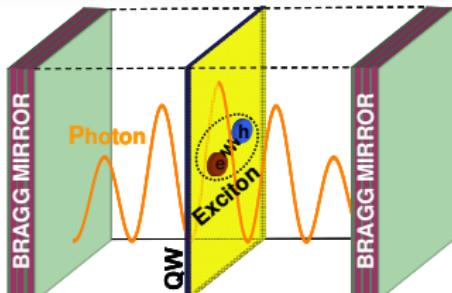
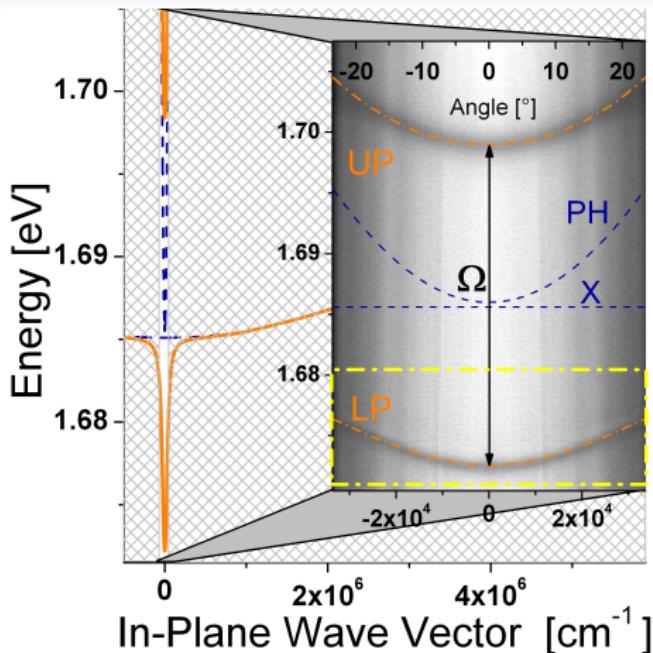
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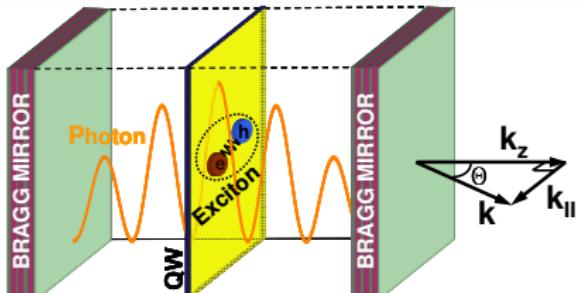
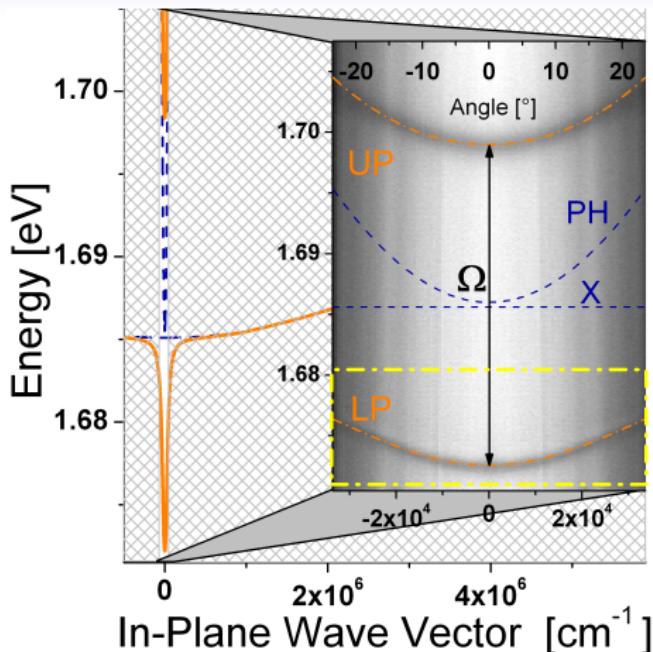
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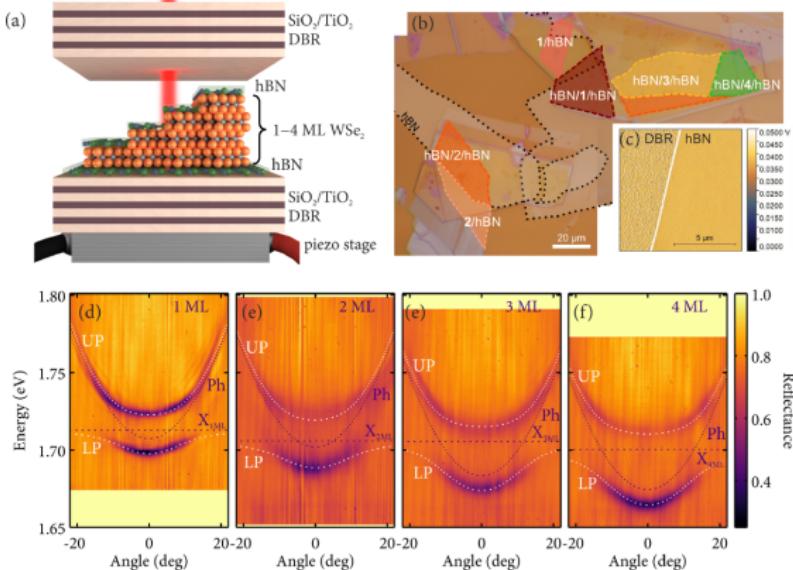
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Digression: Polaritons in a tunable cavity with TMDs



M. Król et al. 2D Materials 7, 015006 (2020)

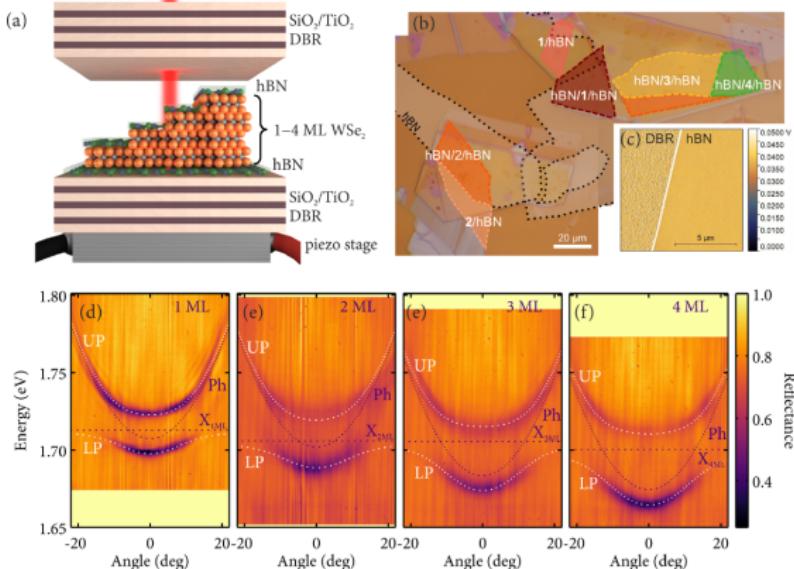


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Polariton versus Atom

Polariton \approx Boson

$$N \ll N_{sat} \sim 10^{11} \text{ cm}^{-2}$$

Parameter

- $m [m_e]$
- $T [\text{K}]$
- $\lambda_{dB} \sim a [\mu\text{m}]$
- $N [\text{cm}^{-2}]$
- Life Time

Atom

- 10^4
- 10^{-6}
- 0.7
- 10^8
- "long" s

Polariton

- 10^{-4}
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- 3
- $5 \cdot 10^8$ (10^{10})
- "very short" 10^{-12} s

Thermalization

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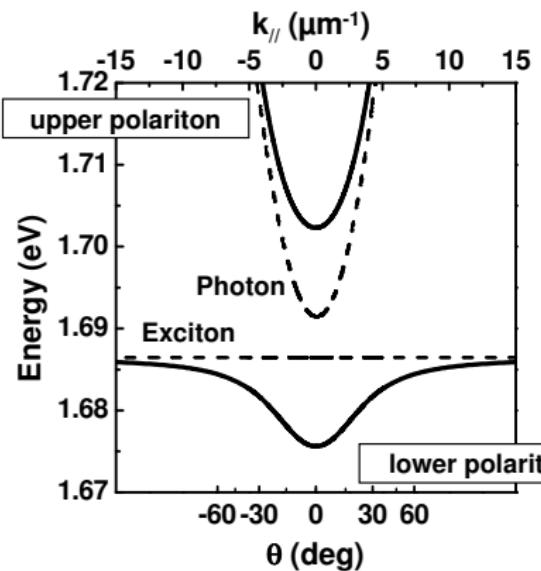
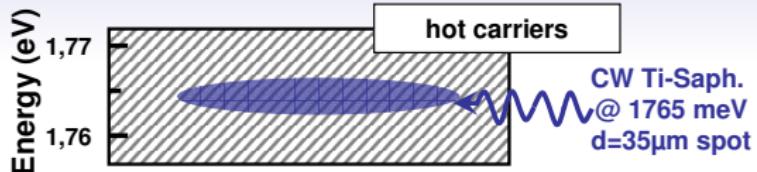
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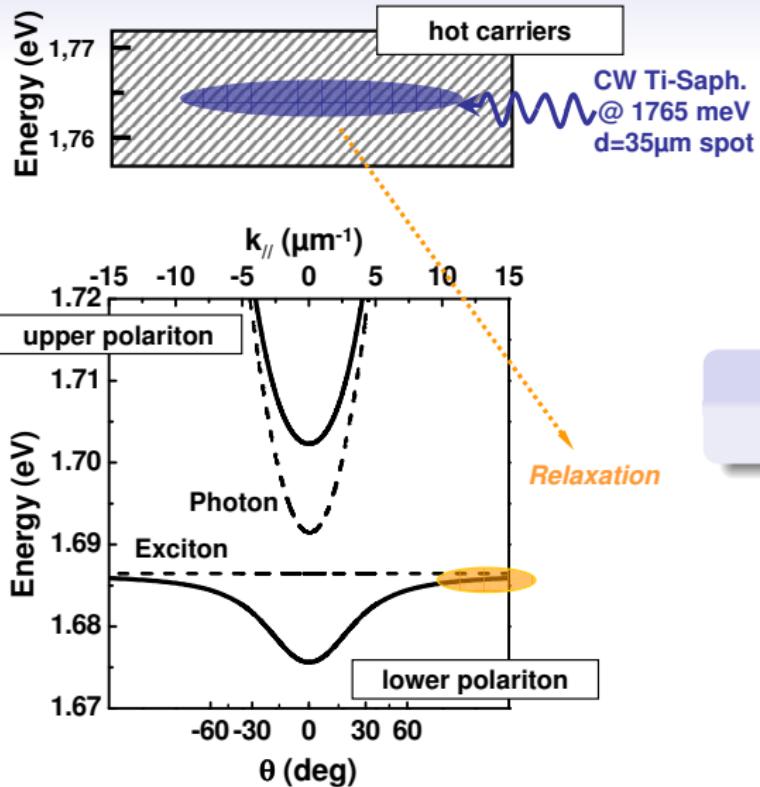
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How to demonstrate polariton condensation ??



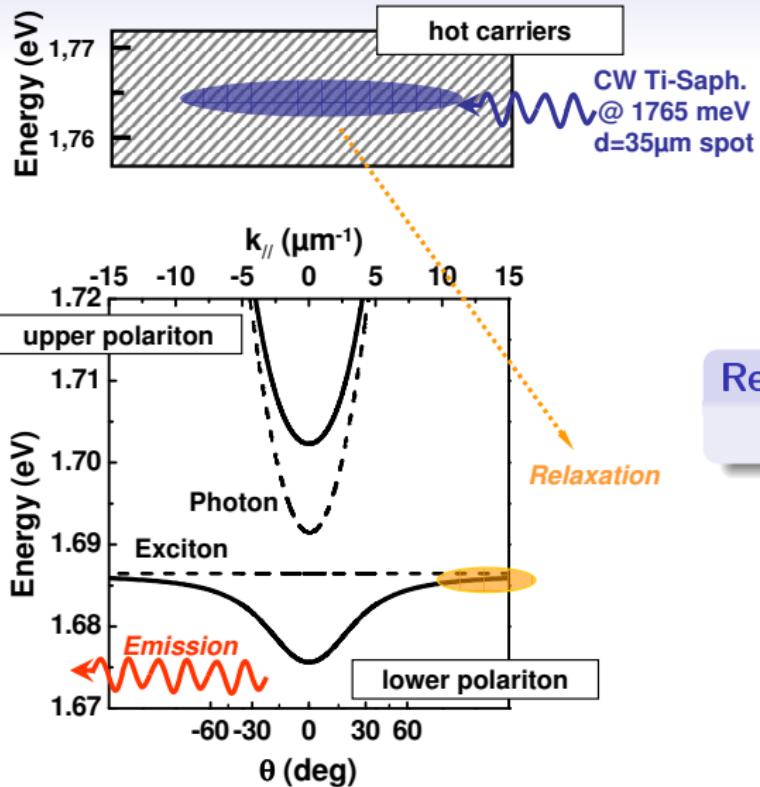
Excitation
Strictly Non-resonant

How to demonstrate polariton condensation ??



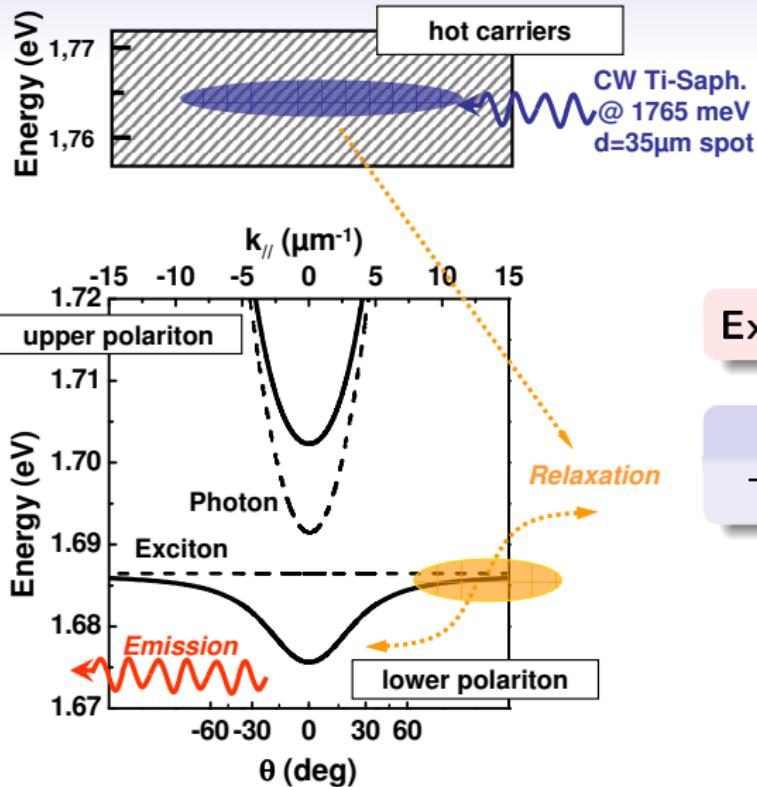
Initial State
Incoherent Excitons

How to demonstrate polariton condensation ??



Relaxation < Emission
Bottleneck

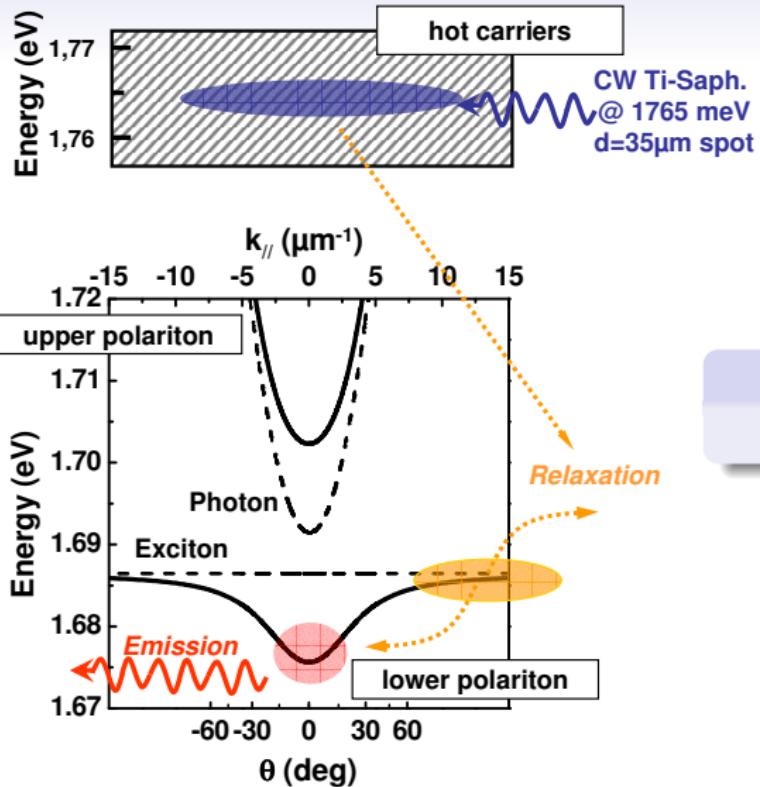
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Exciton-Polariton Scattering

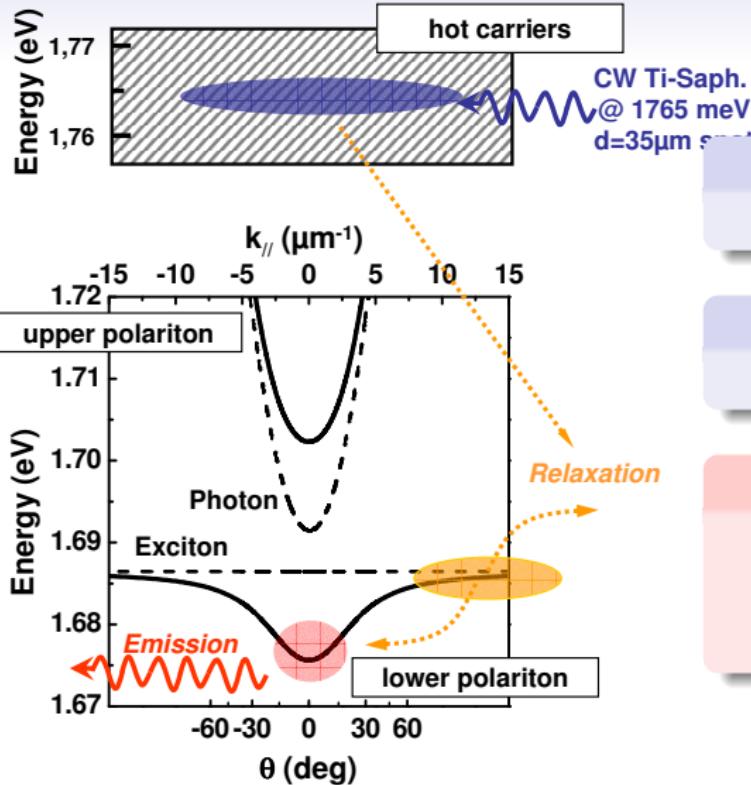
Relaxation \sim Emission
Thermalization, Stimulation

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Final State
Coherent Condensate

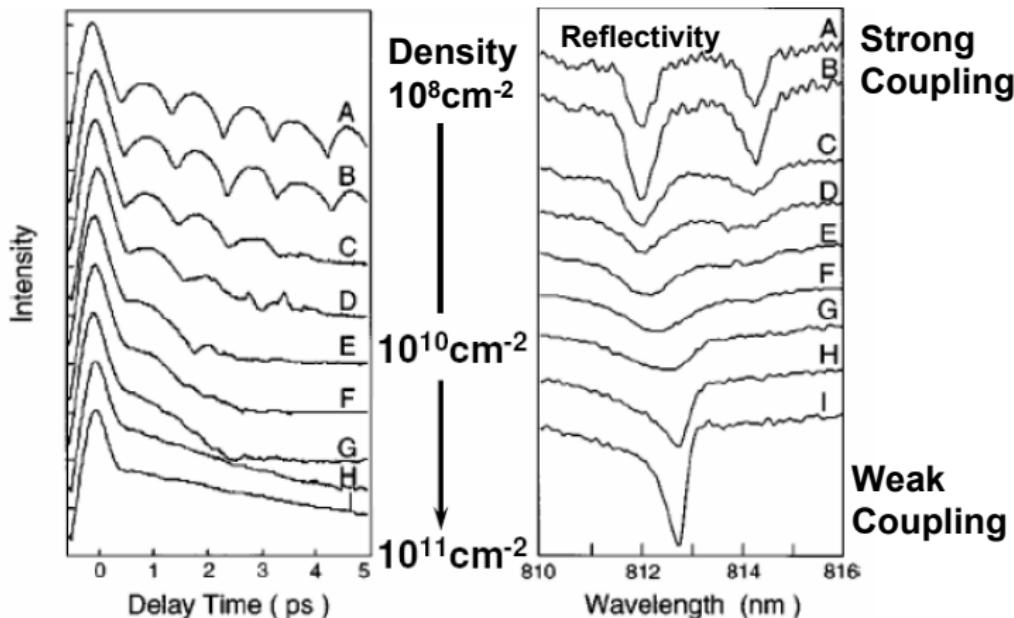
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GaAs Microcavity with a single QW



S. Jiang et al. APL 73, 3031, 1998

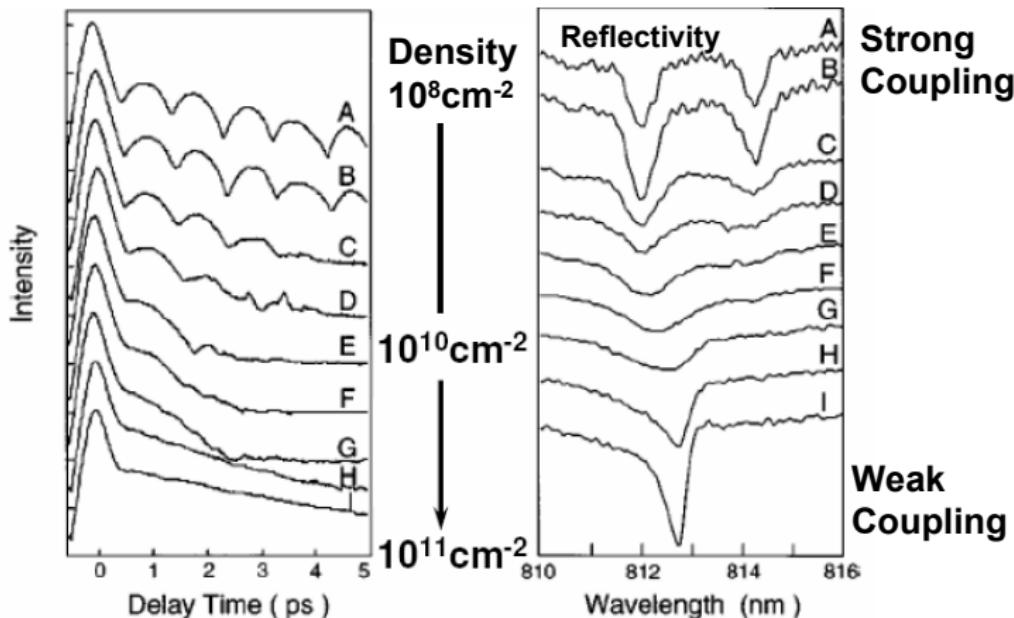


Exciton dissociation before condensation

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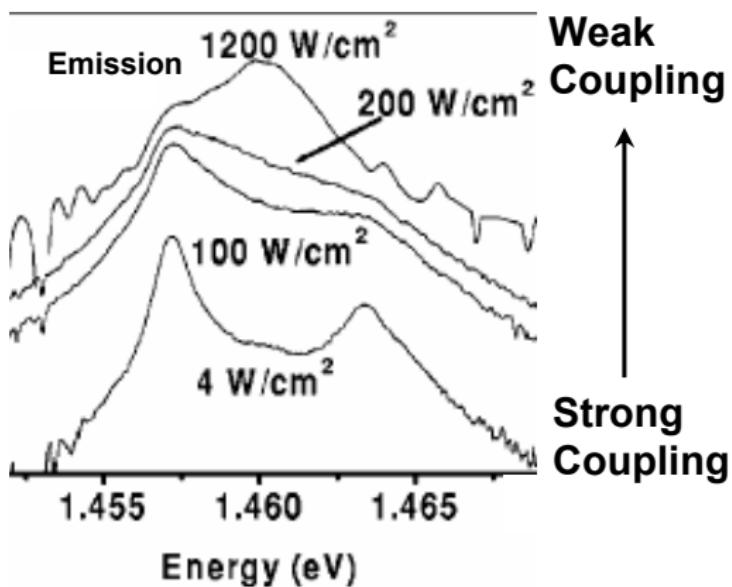
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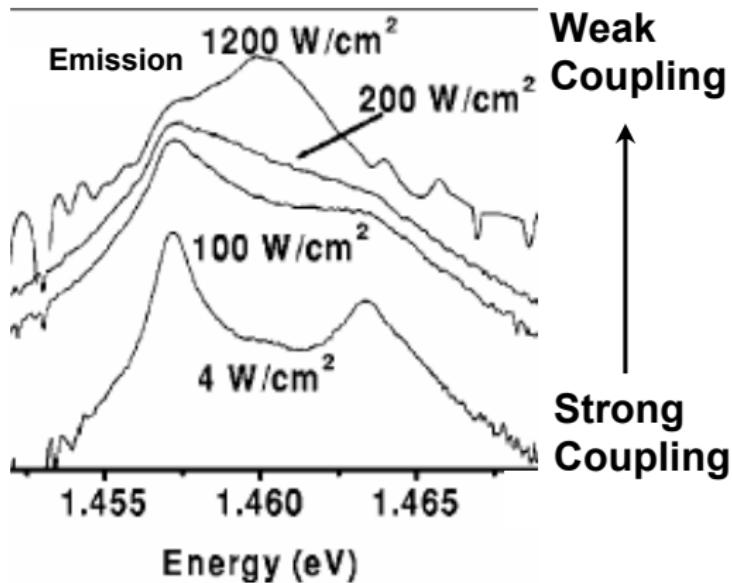
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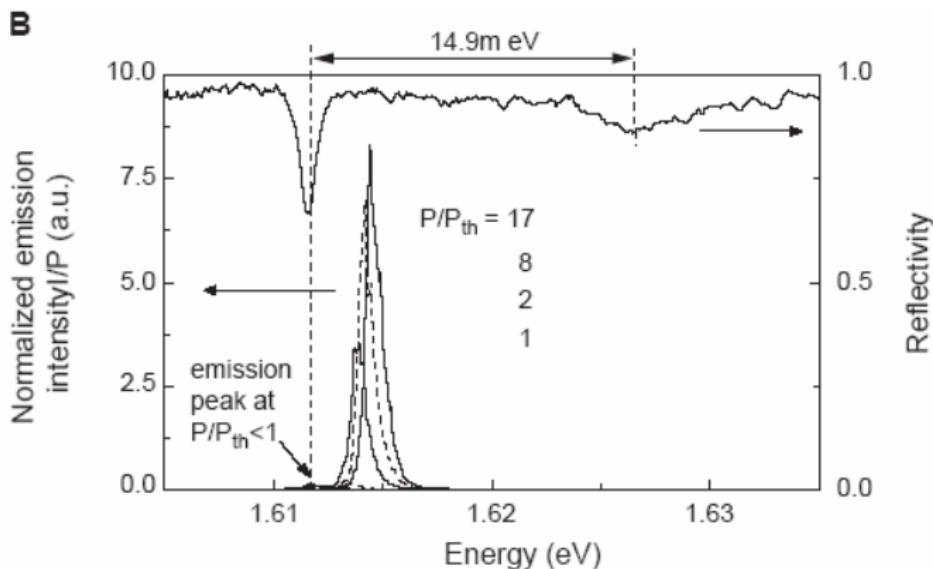
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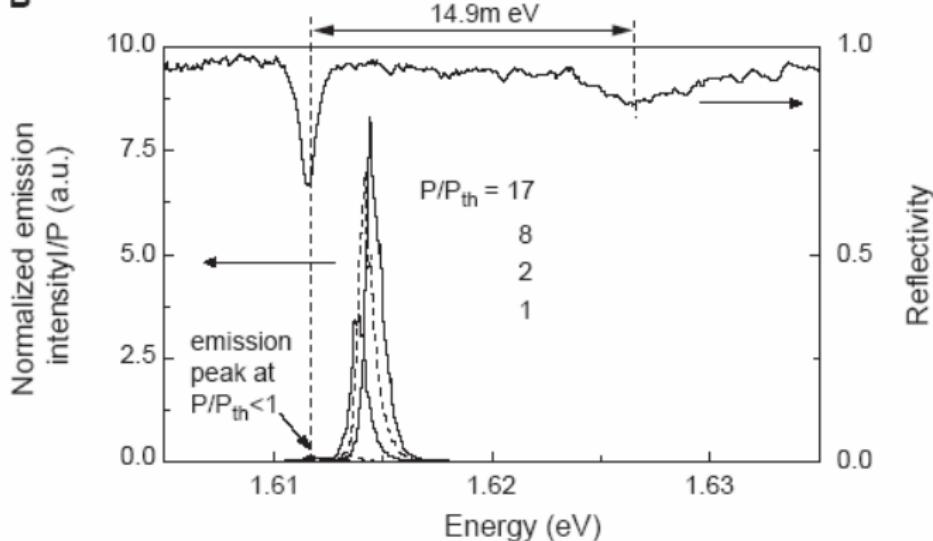


Stimulated scattering in strong coupling

Coherence ?? Non-resonant ??

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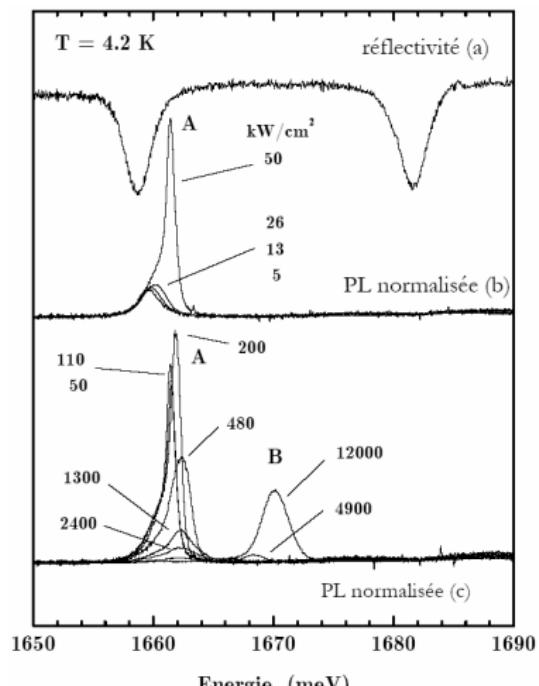
CdTe Microcavity with 16 QWs



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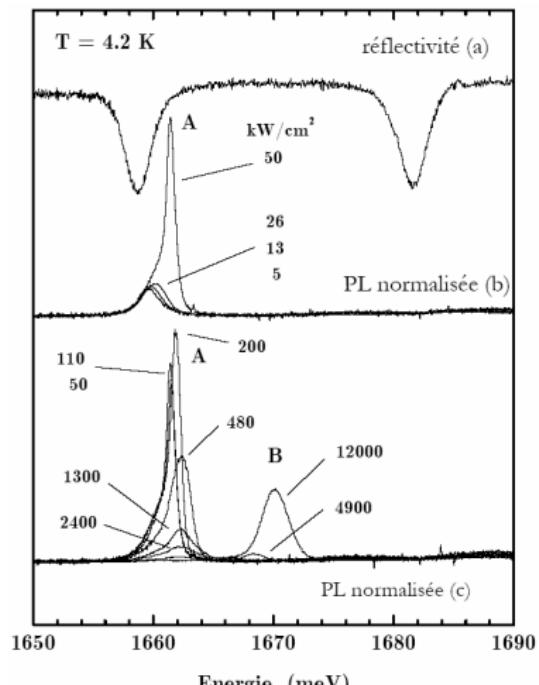
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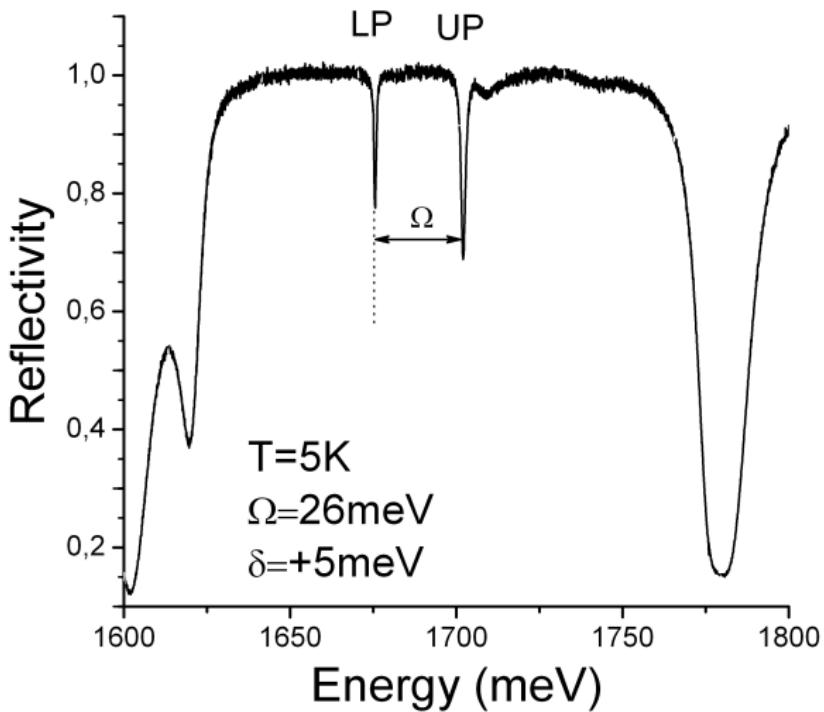
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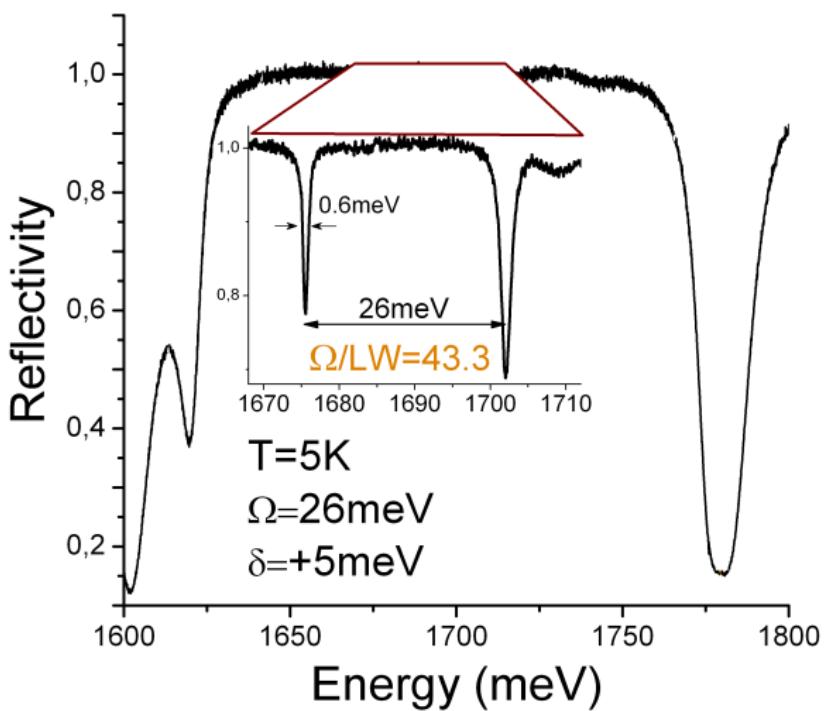
Optical Response of the CdTe Microcavity

$16PQs \Rightarrow \Omega = 26\text{meV}$



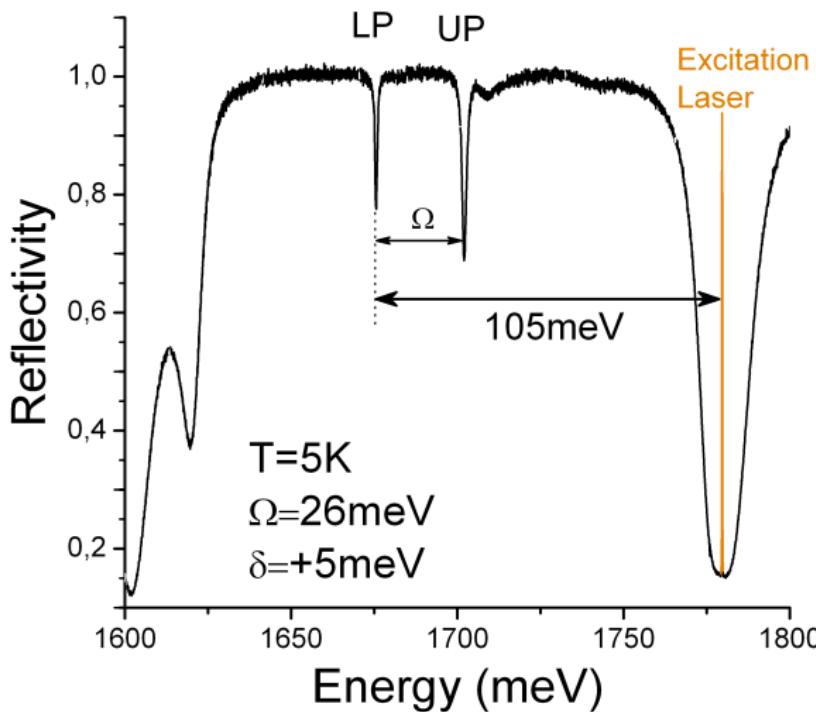
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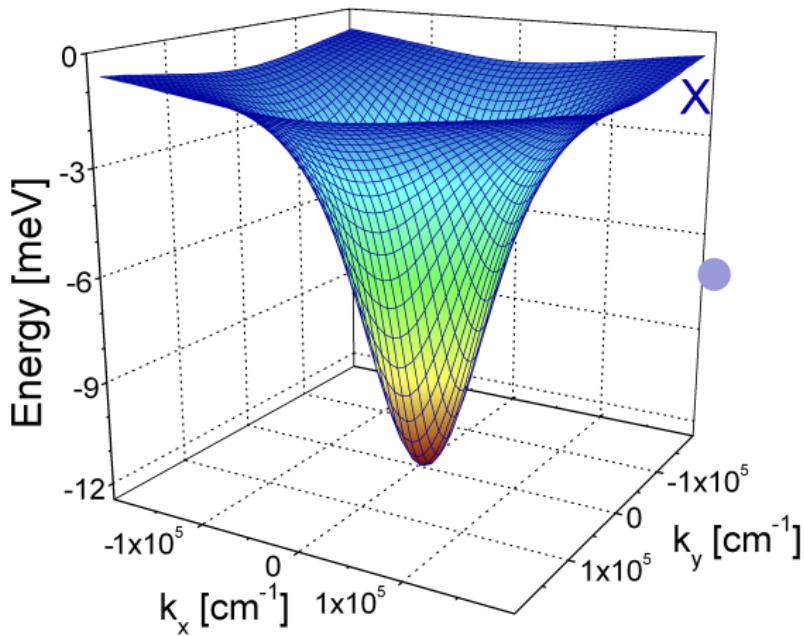
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Non-resonant Excitation



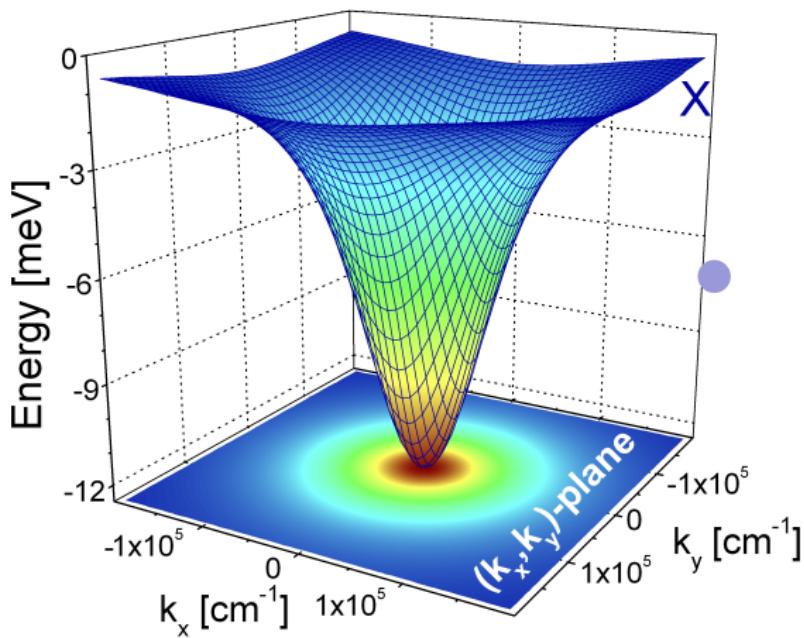
Polariton Condensation in Momentum space

Far Field Imaging



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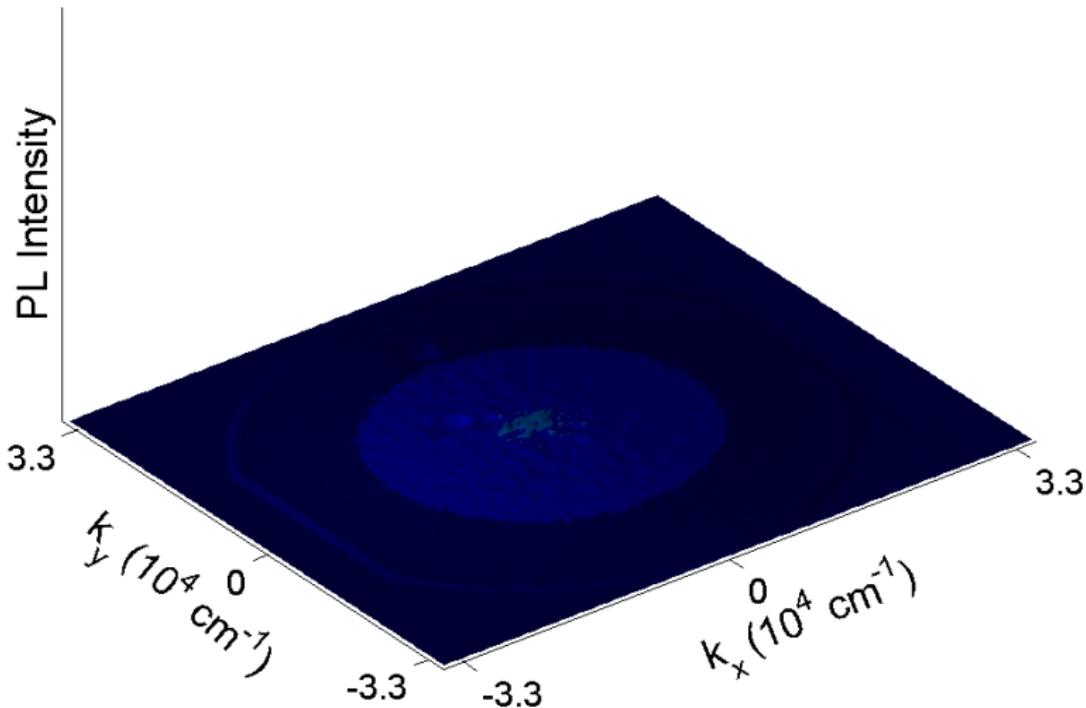
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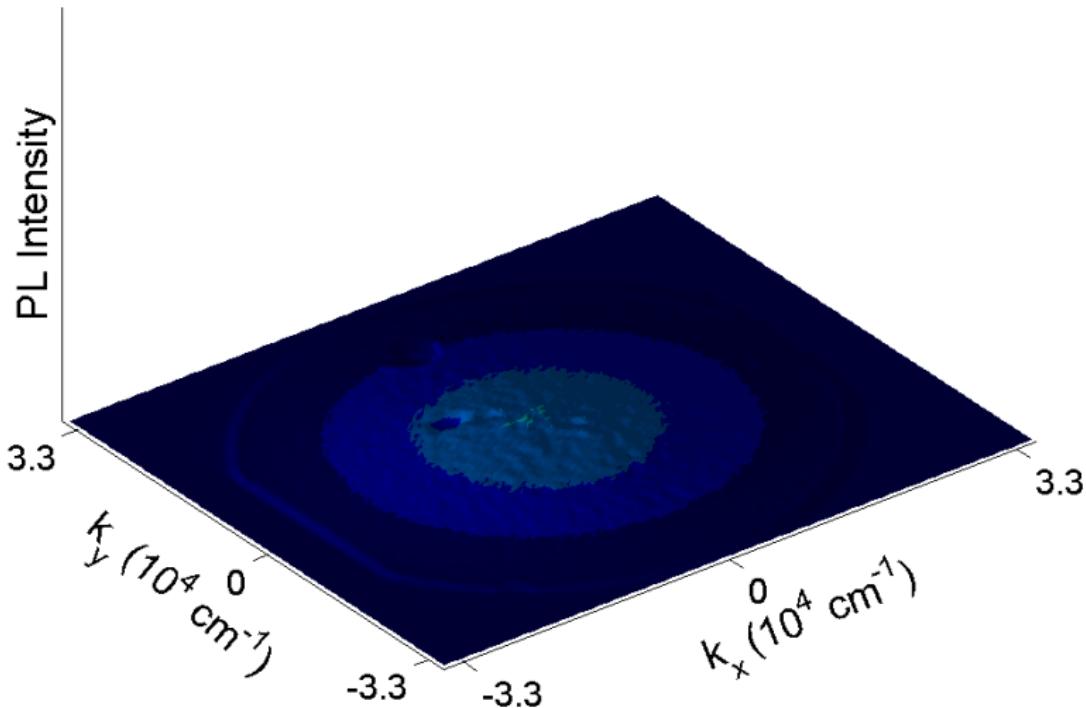
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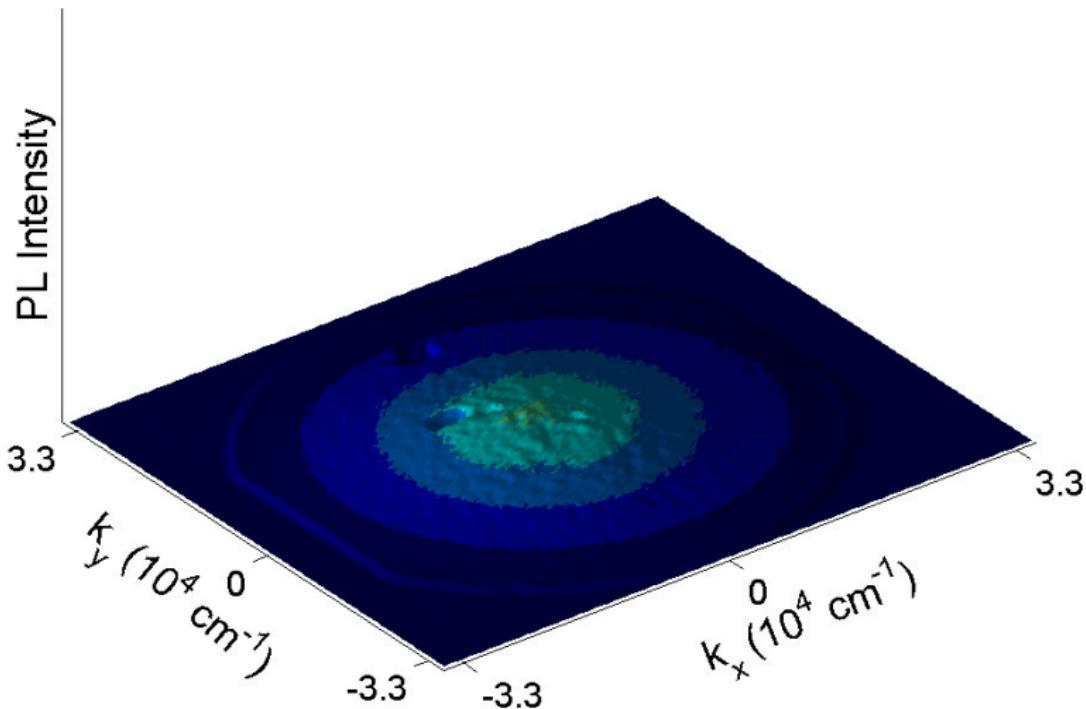
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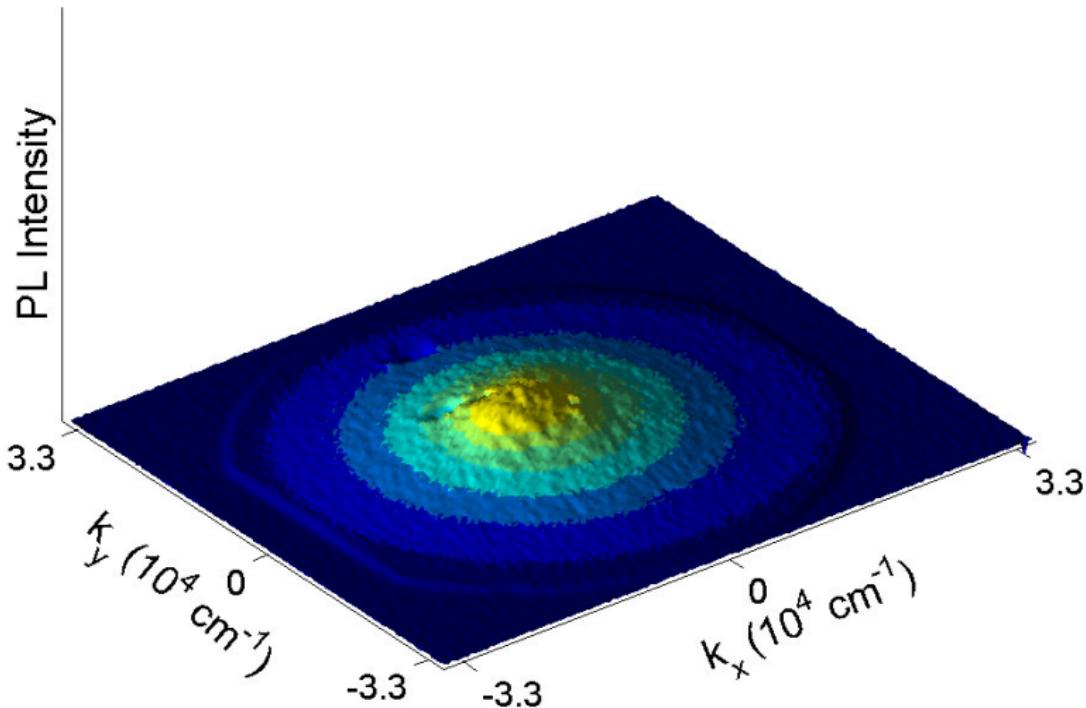
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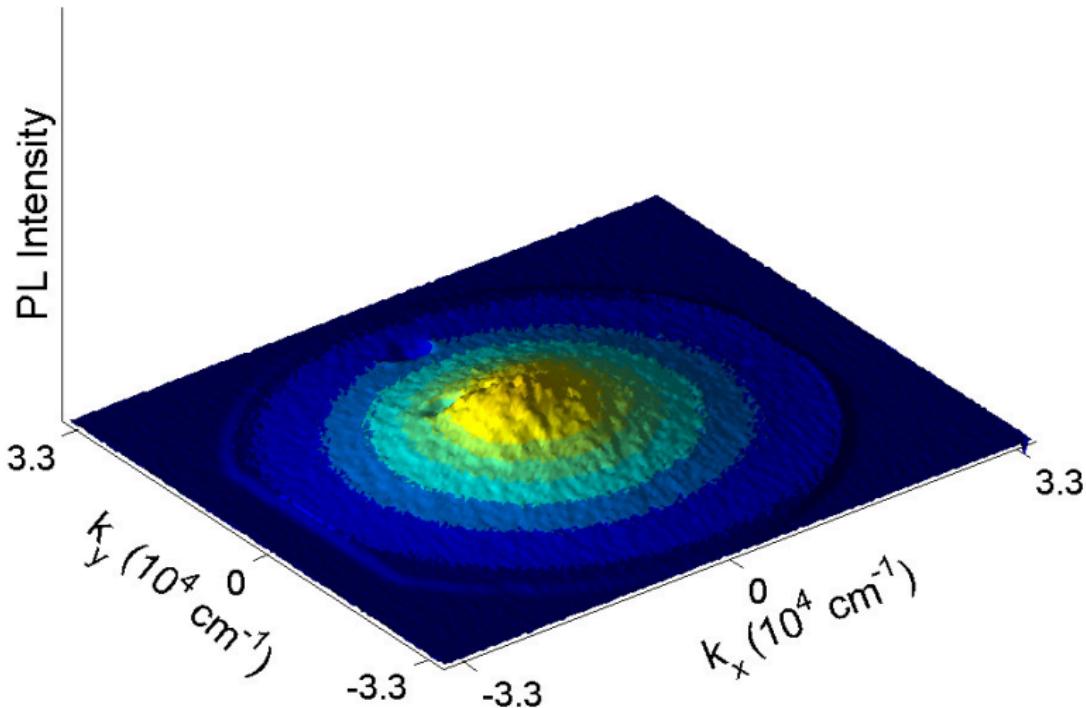
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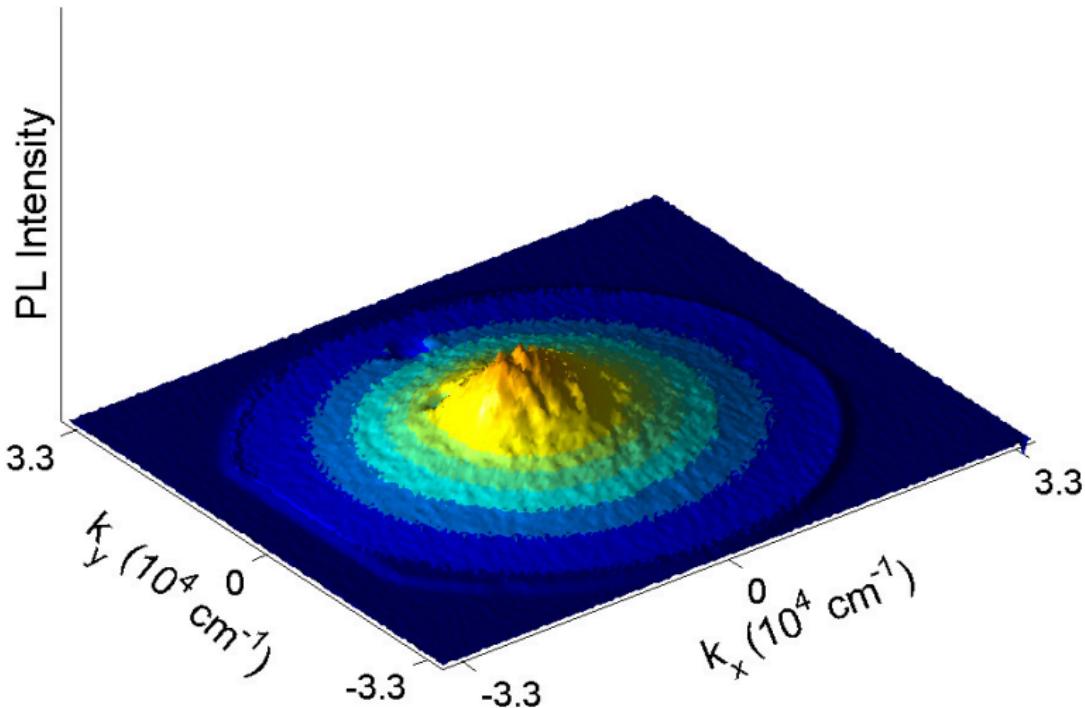
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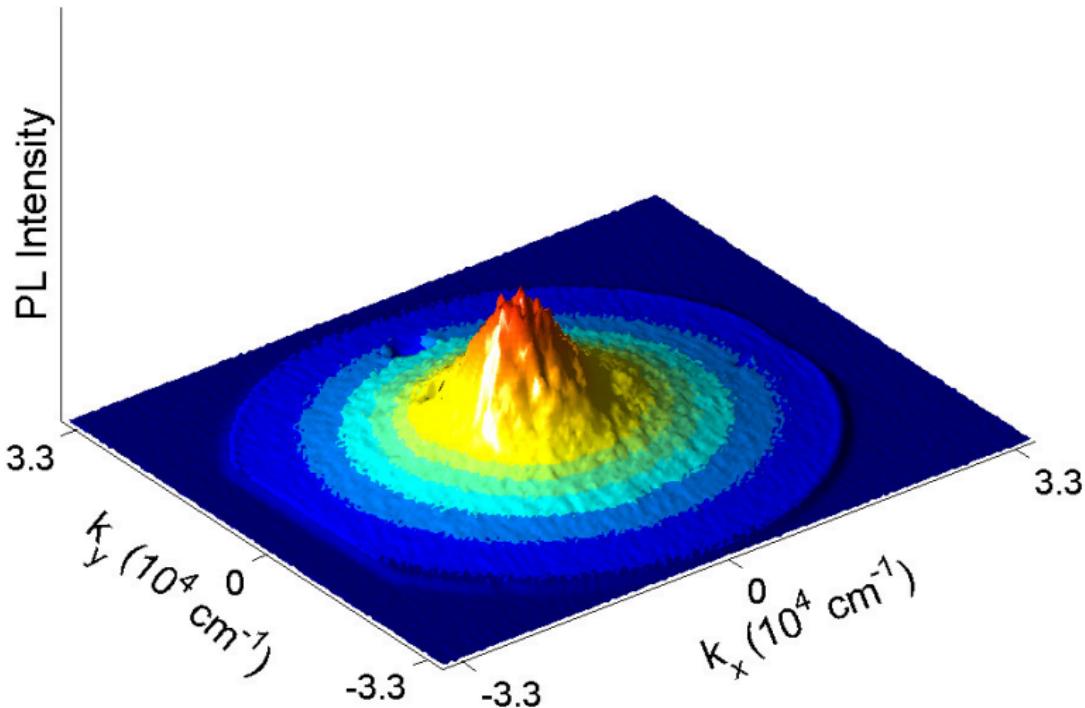
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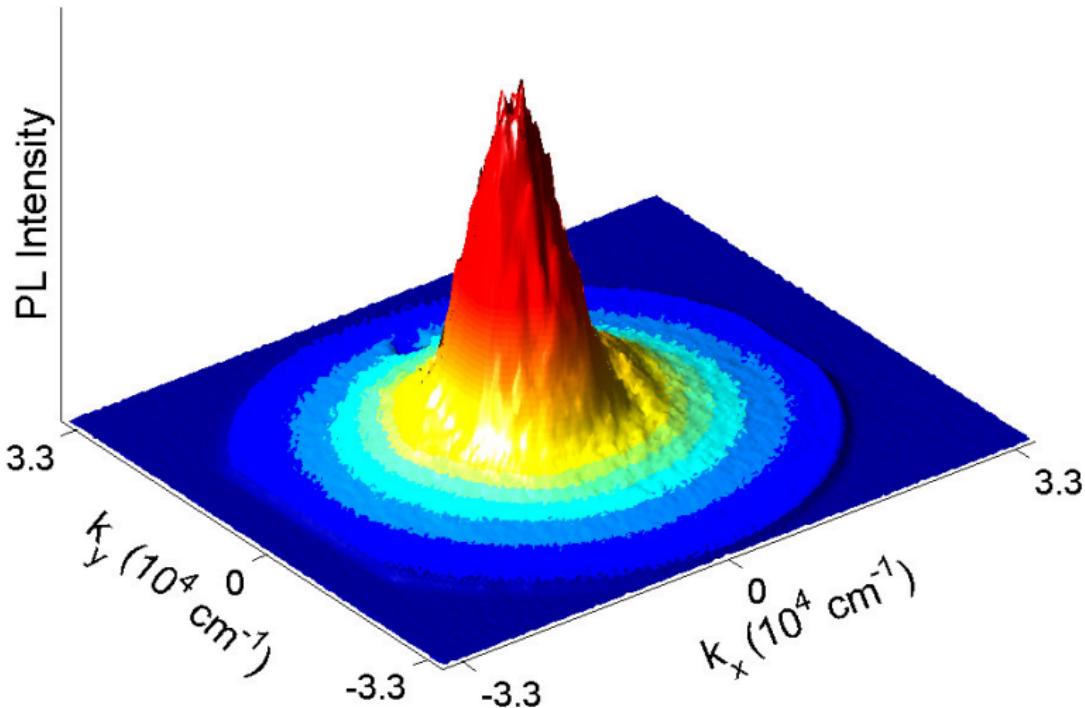
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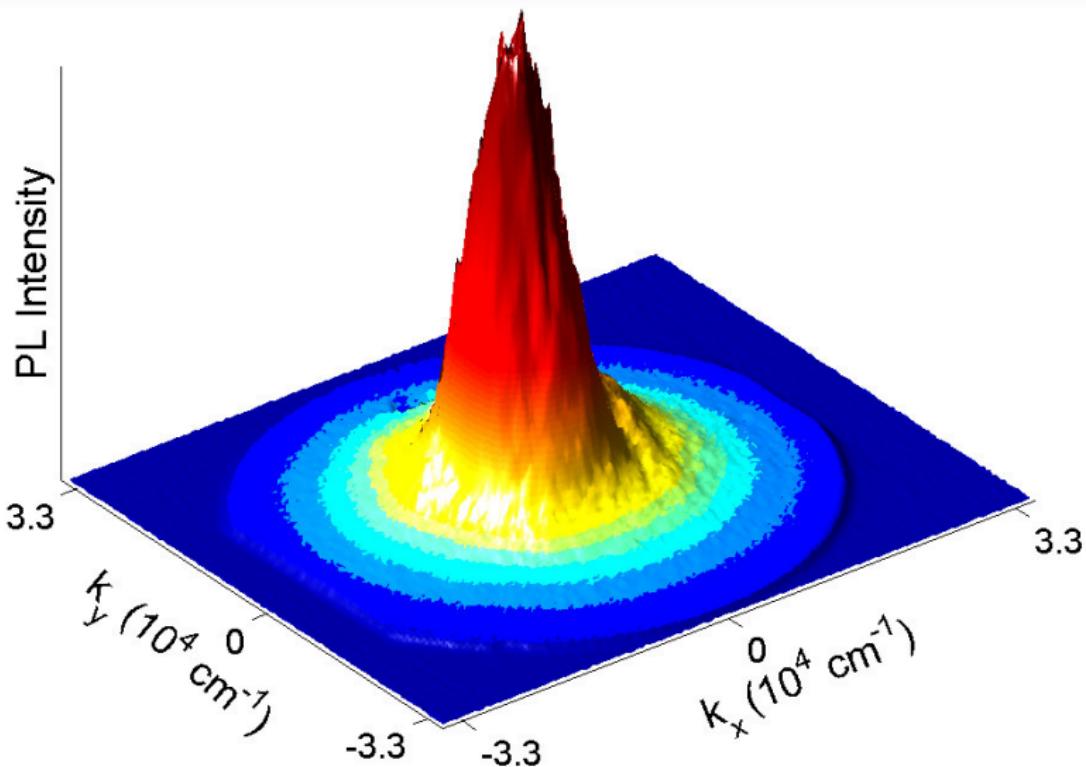
Polariton Condensation in Momentum Plane

Non-resonant, CW Excitation



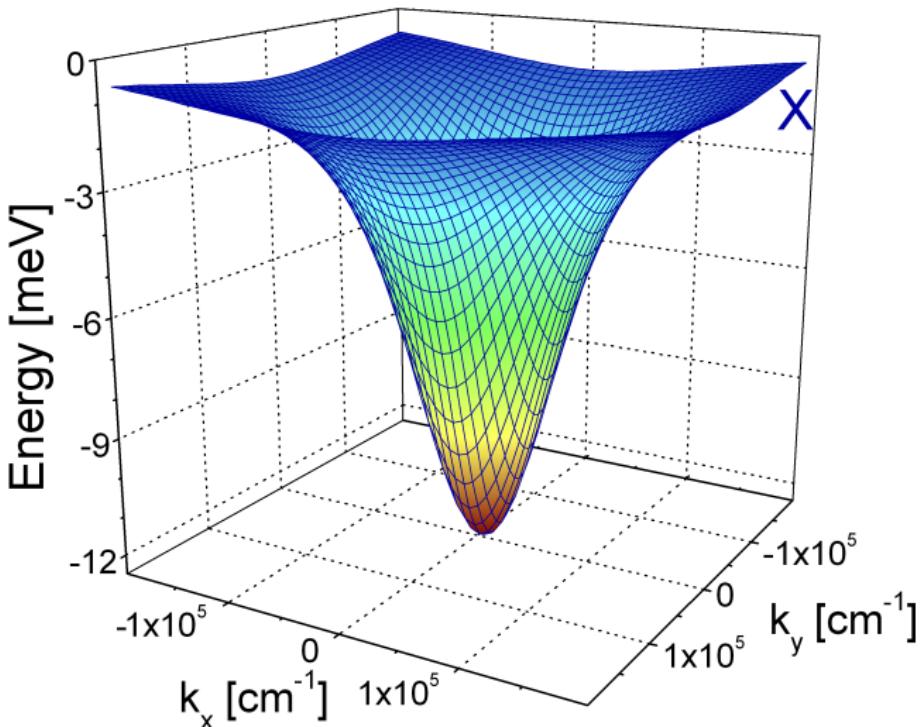
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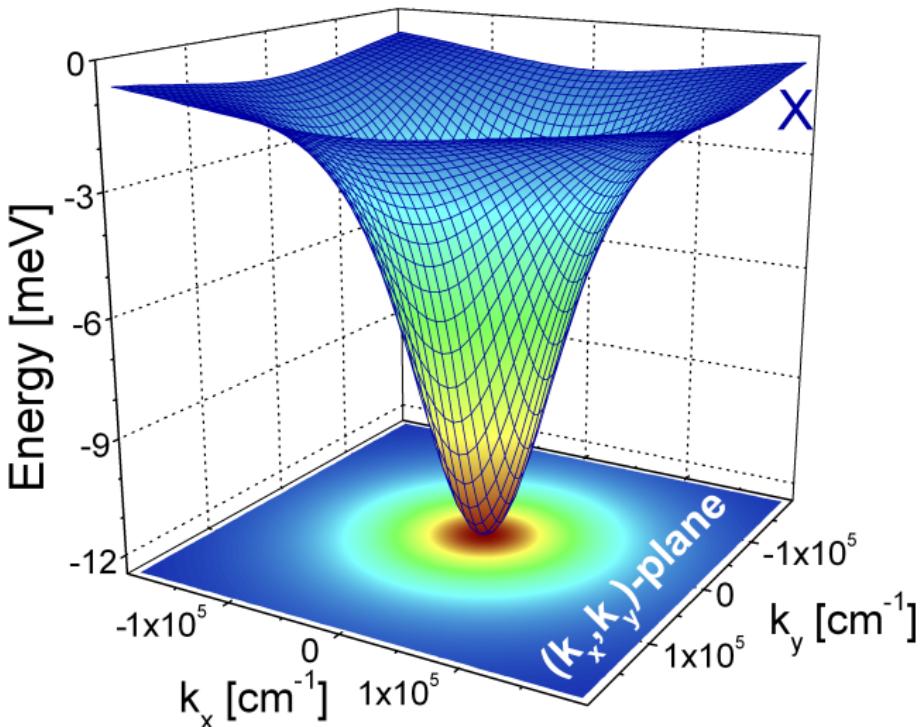
Polariton Condensation in Dispersion Plane

Spectrally Resolved Far Field Imaging



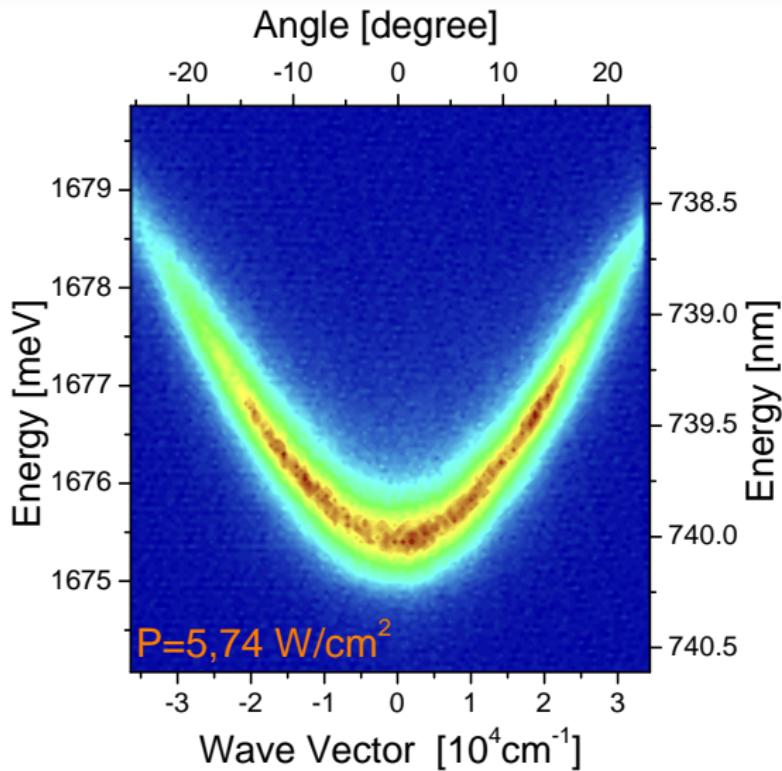
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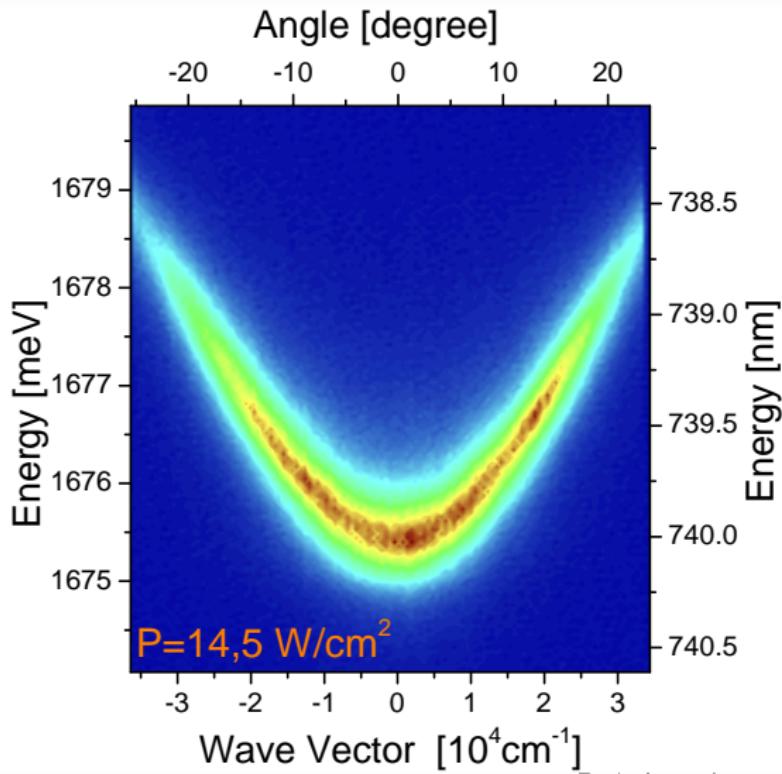
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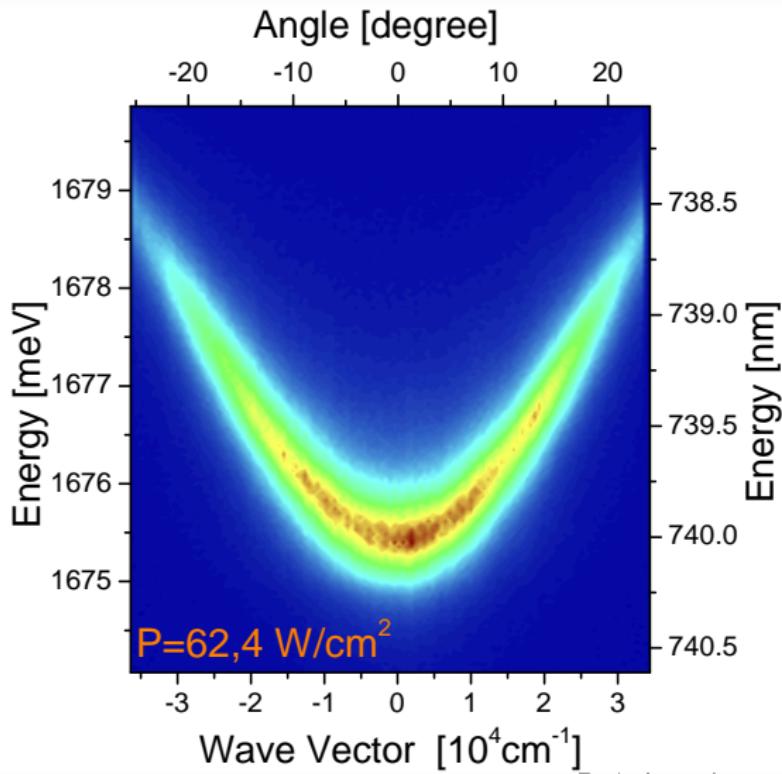
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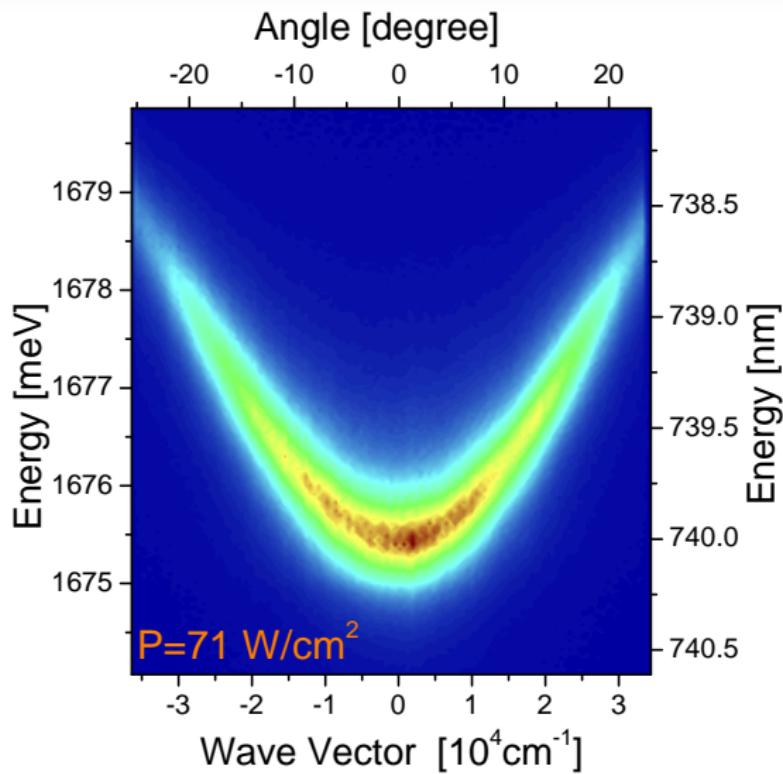
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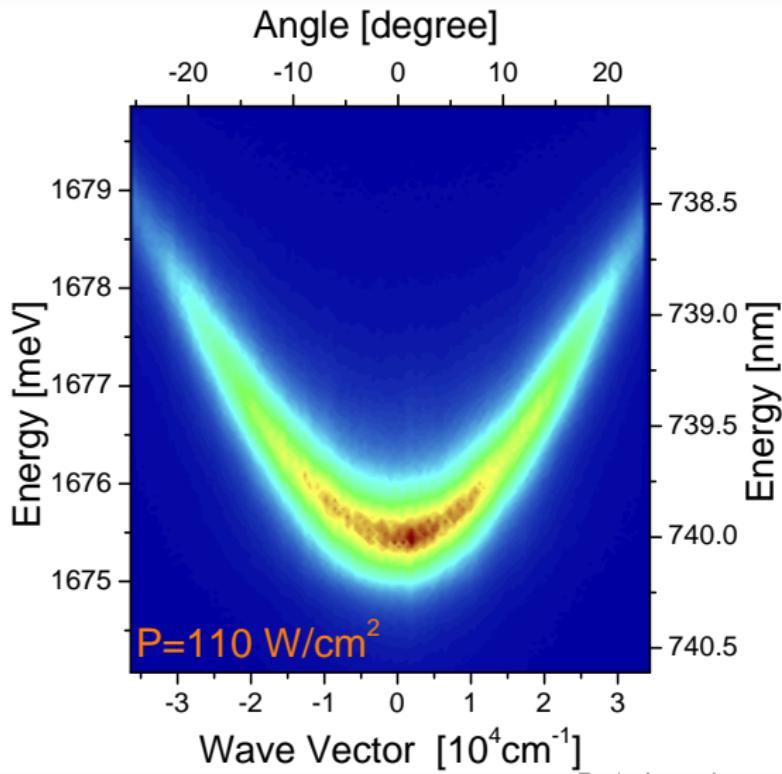
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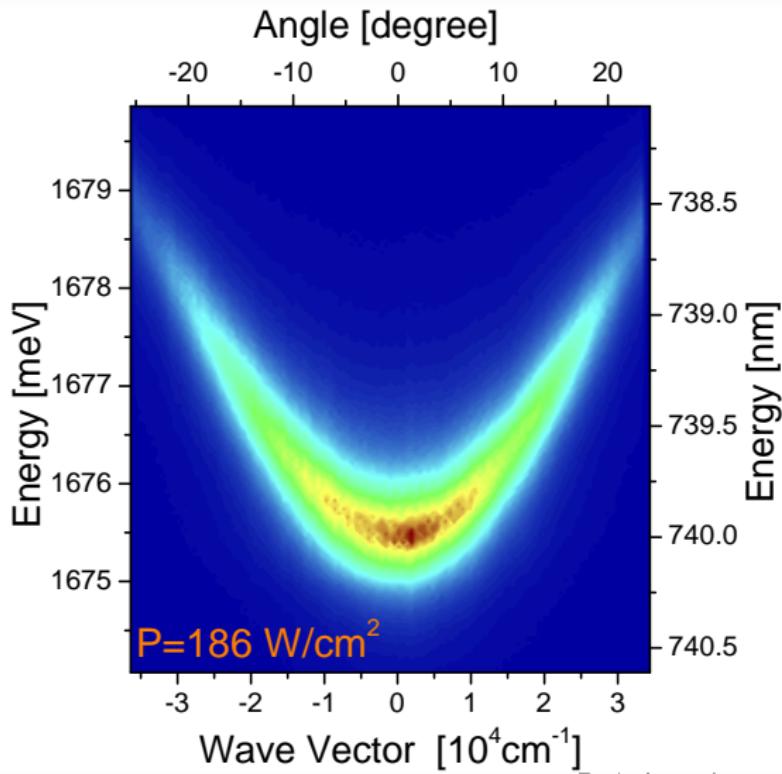
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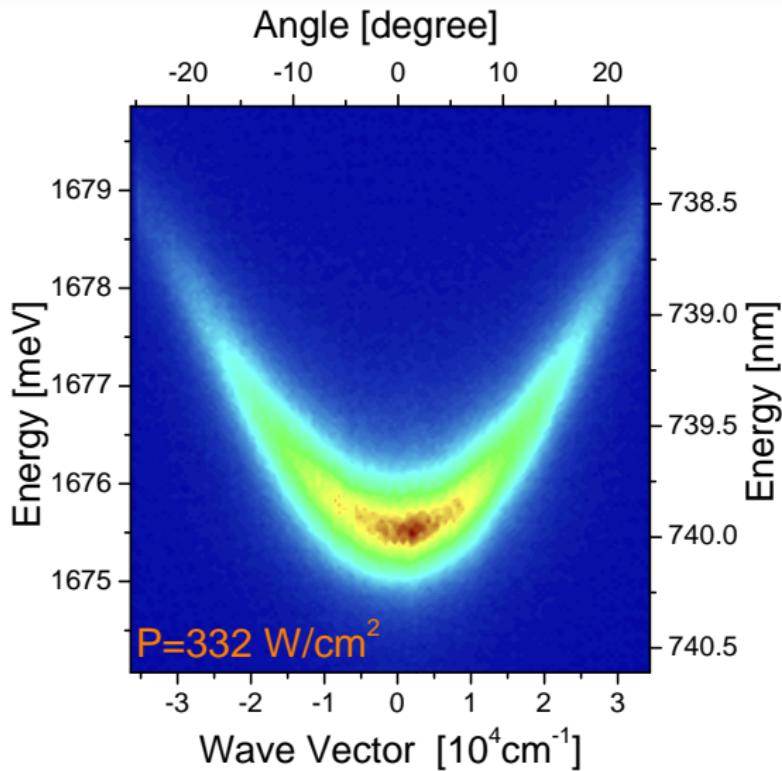
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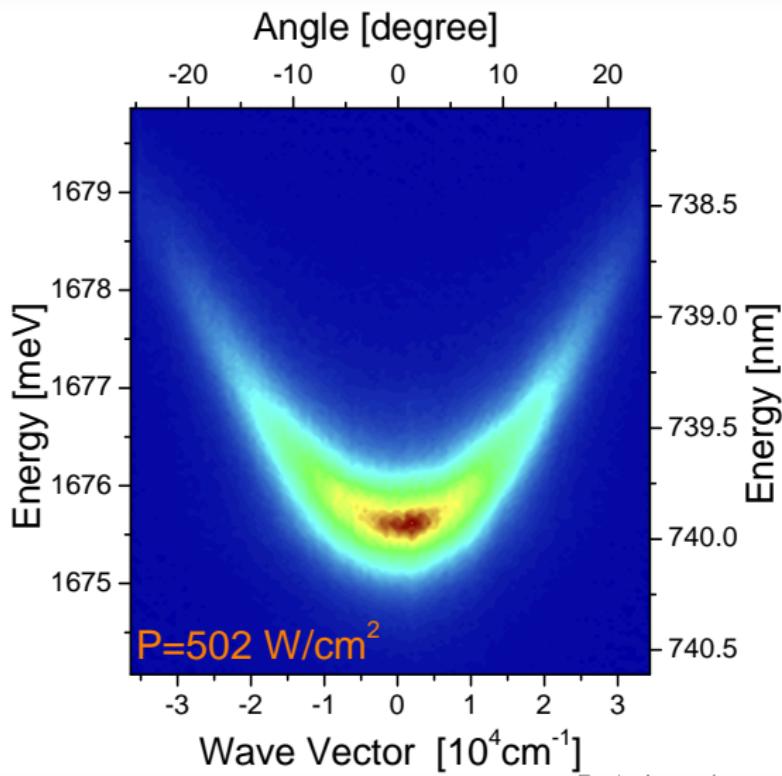
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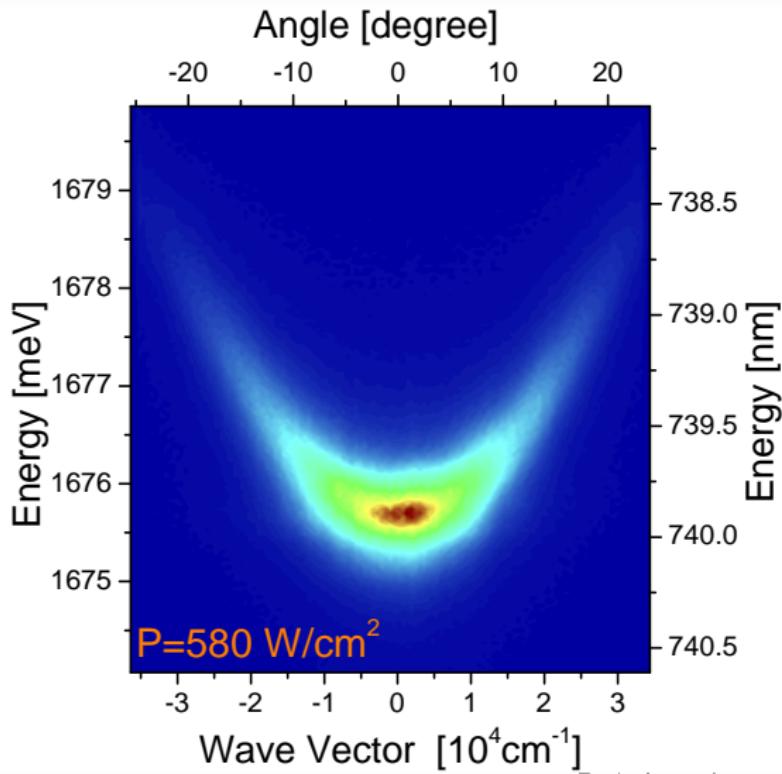
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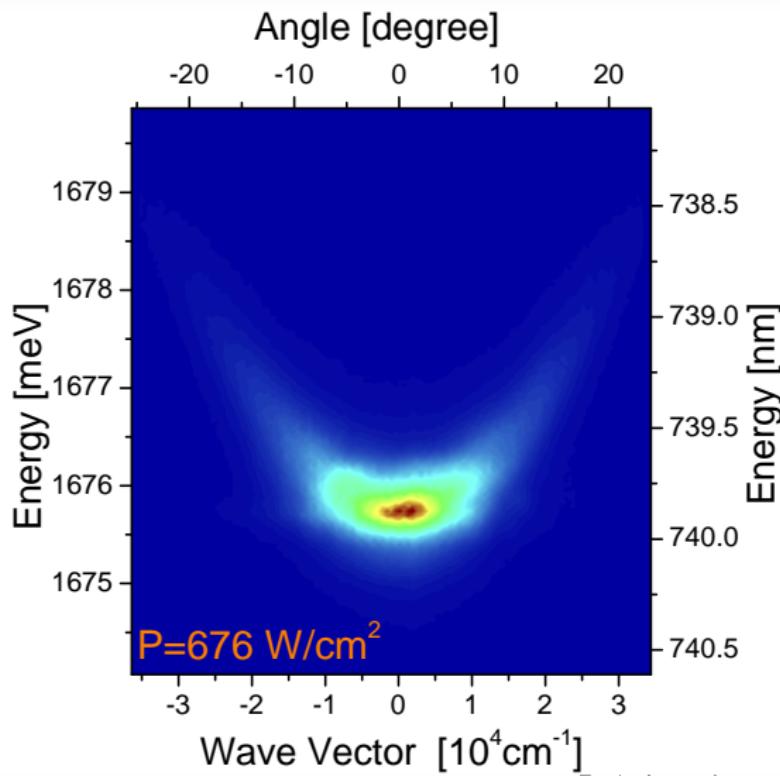
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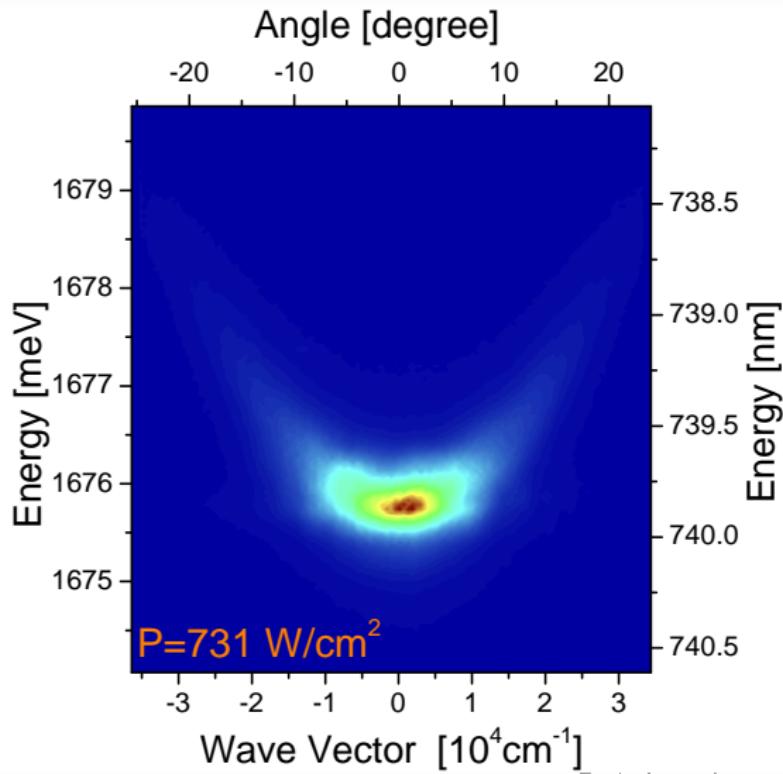
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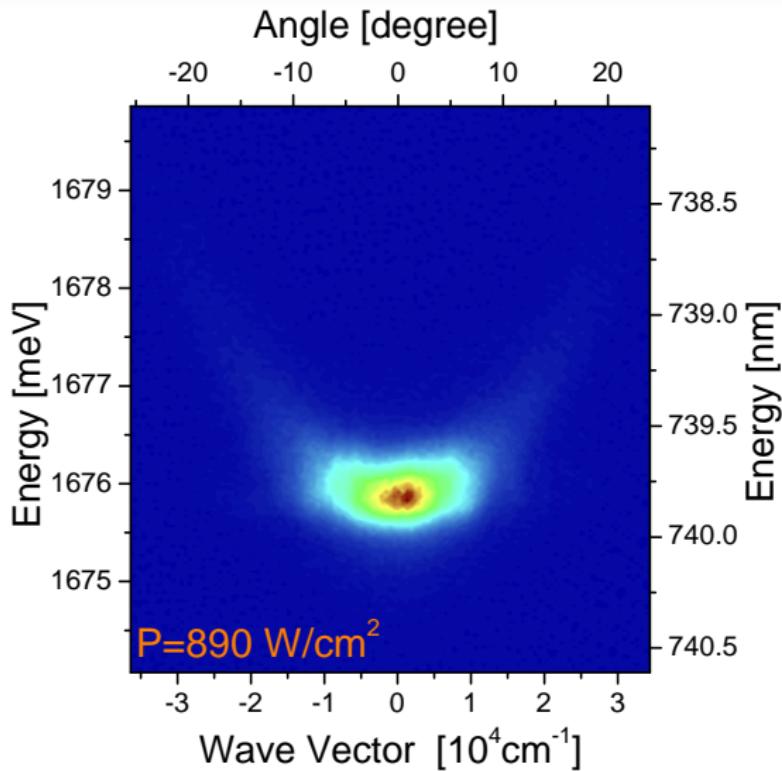
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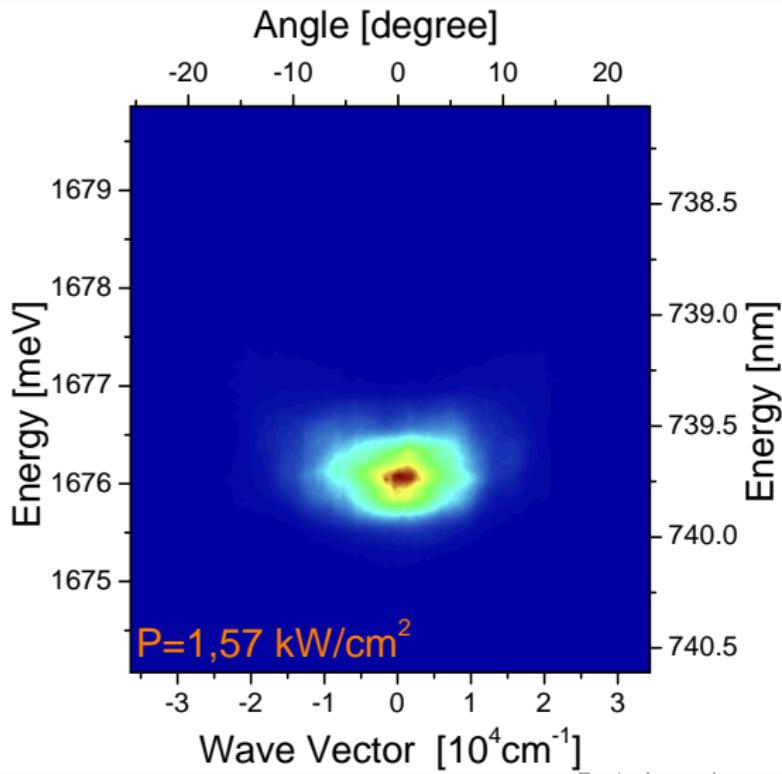
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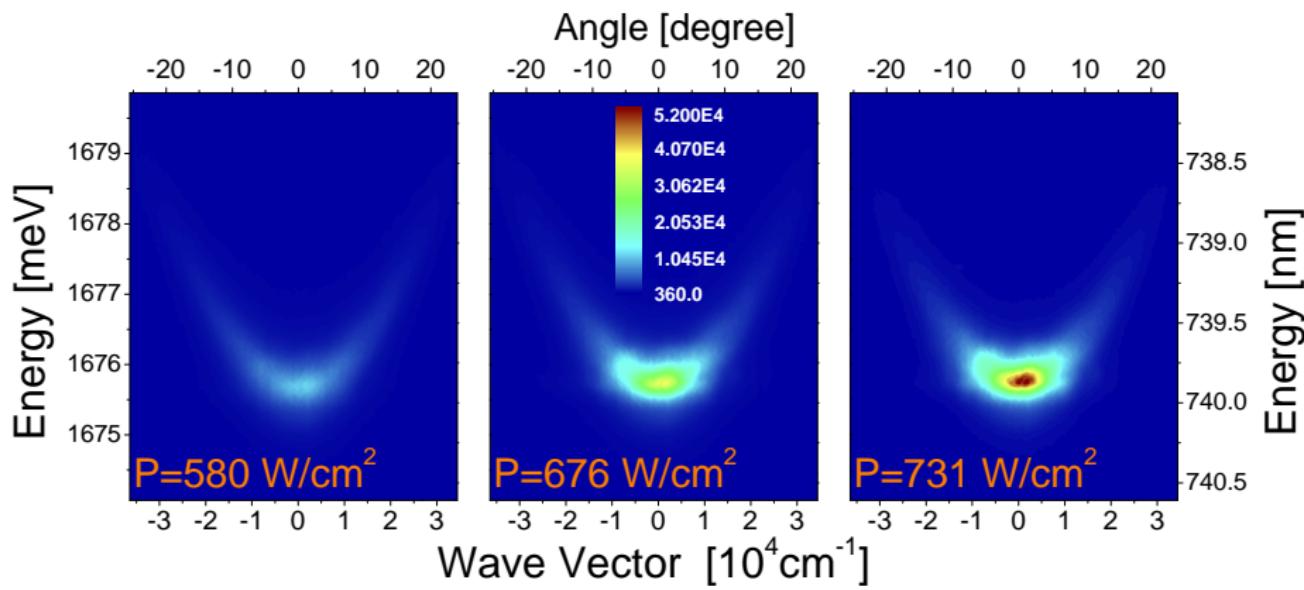


Polariton Condensation in Dispersion Plane

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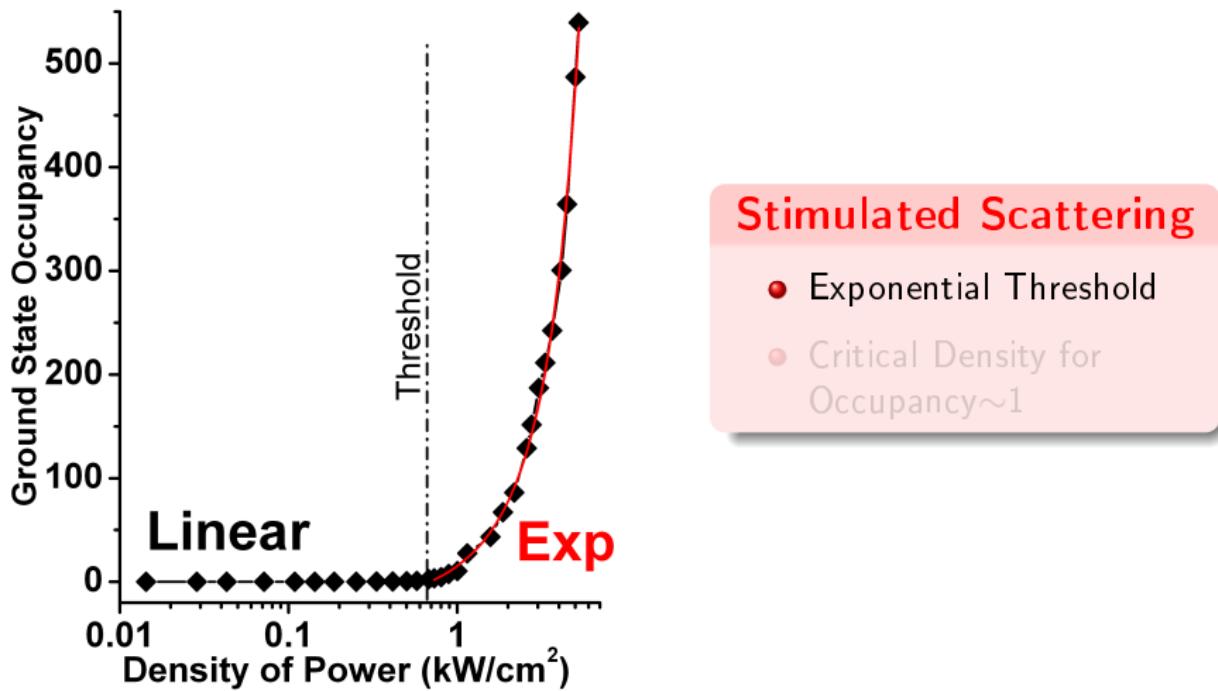


Saturation of the Excited States

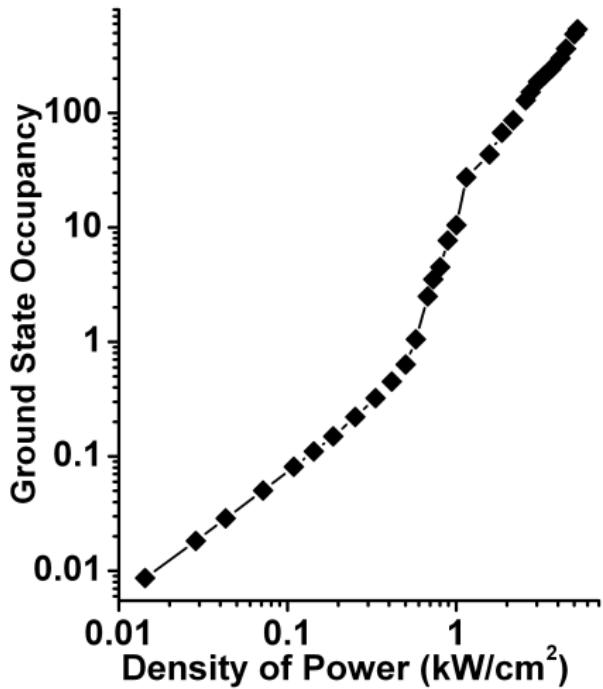


Log Scale

Massive Occupation of the Ground State



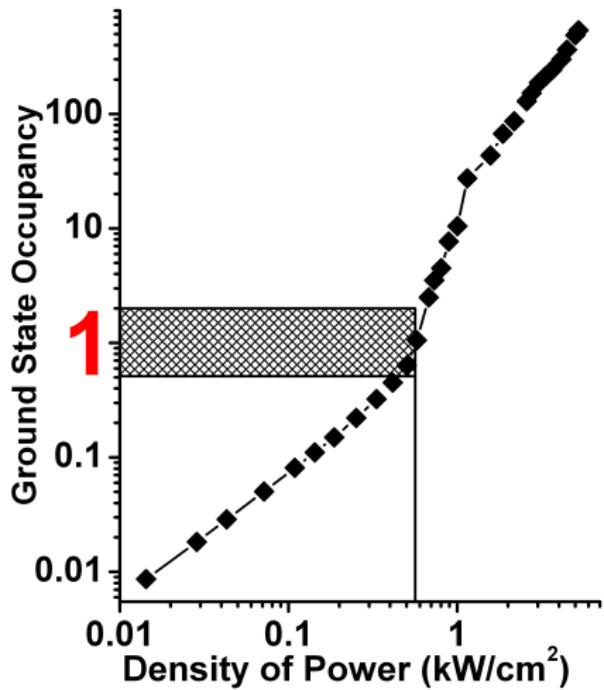
Massive Occupation of the Ground State



Stimulated Scattering

- Exponential Threshold
- Critical Density for Occupancy~1

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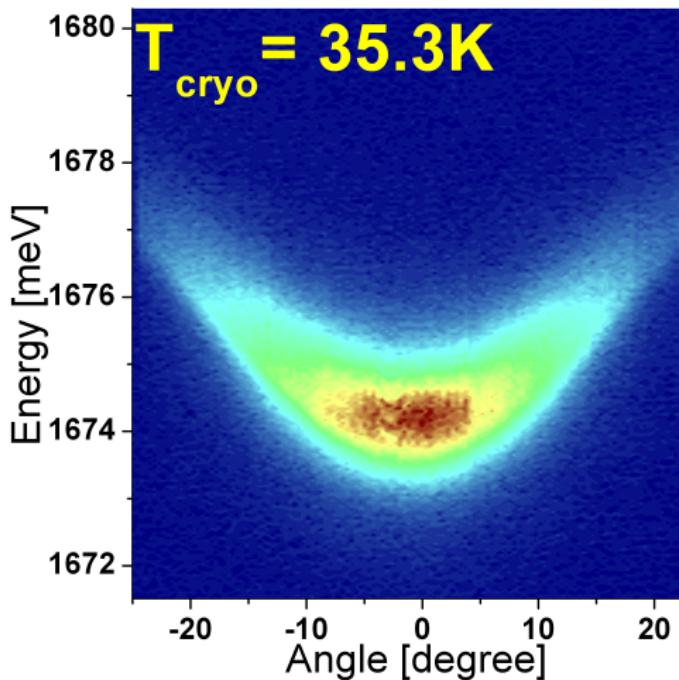
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Polariton Condensation in Dispersion Plane

Temperature as a Control Parameter

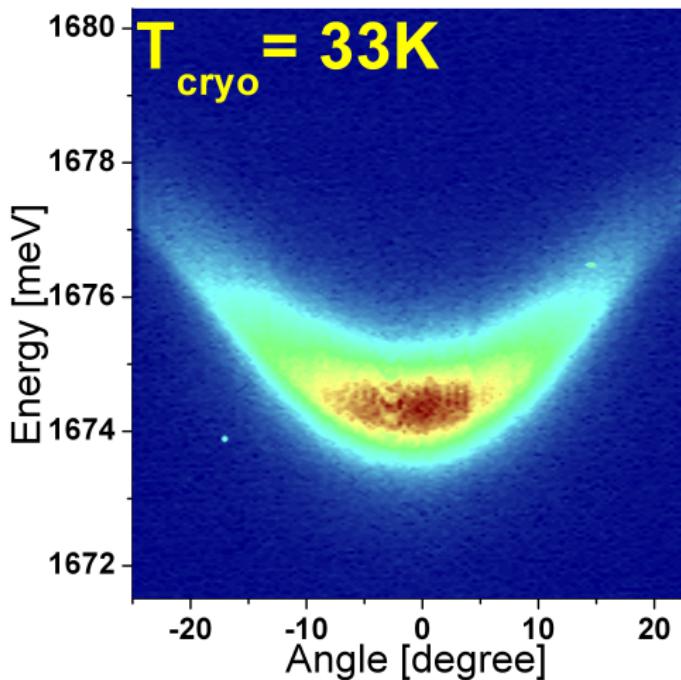
$P = \text{const}$, T_C Exists !



Polariton Condensation in Dispersion Plane

Temperature as a Control Parameter

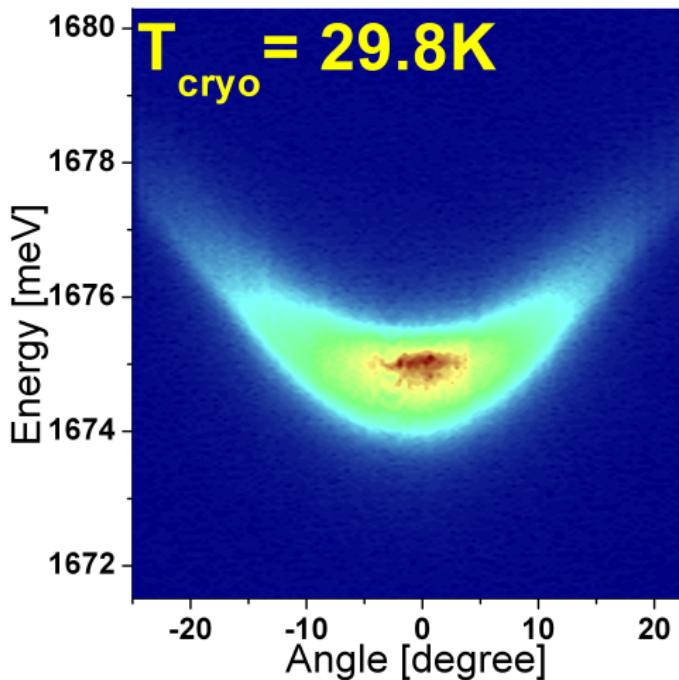
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Polariton Condensation in Dispersion Plane

Temperature as a Control Parameter

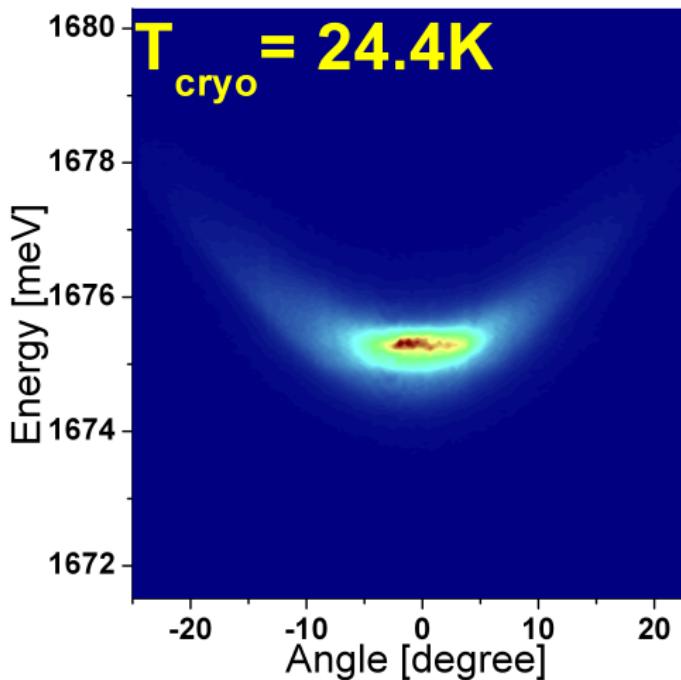
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Polariton Condensation in Dispersion Plane

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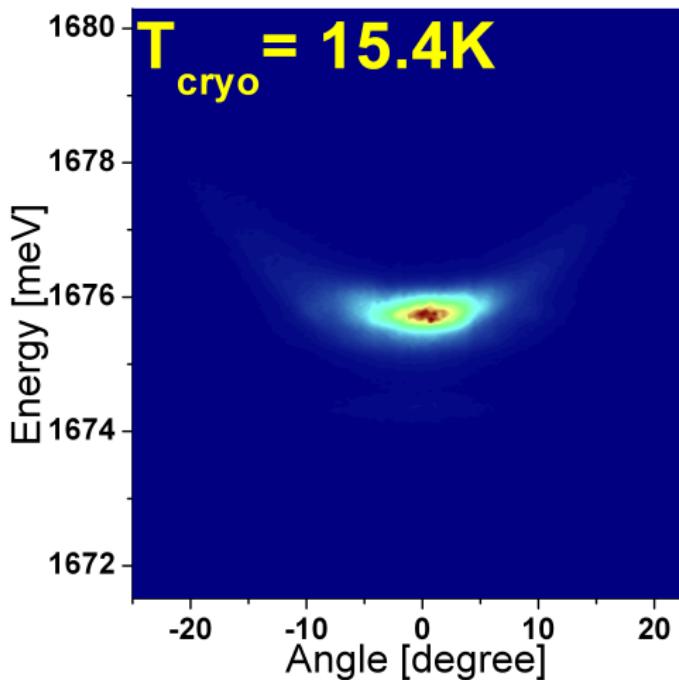
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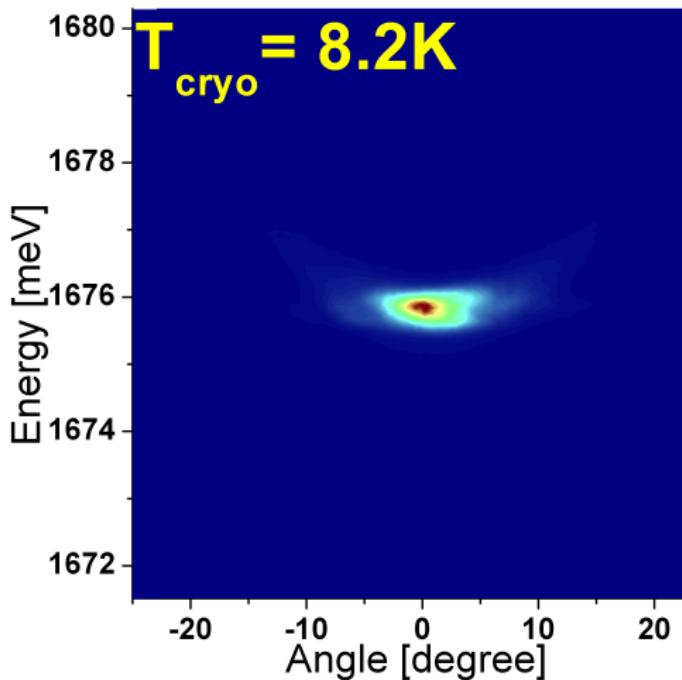
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Polariton Condensation in Dispersion Plane

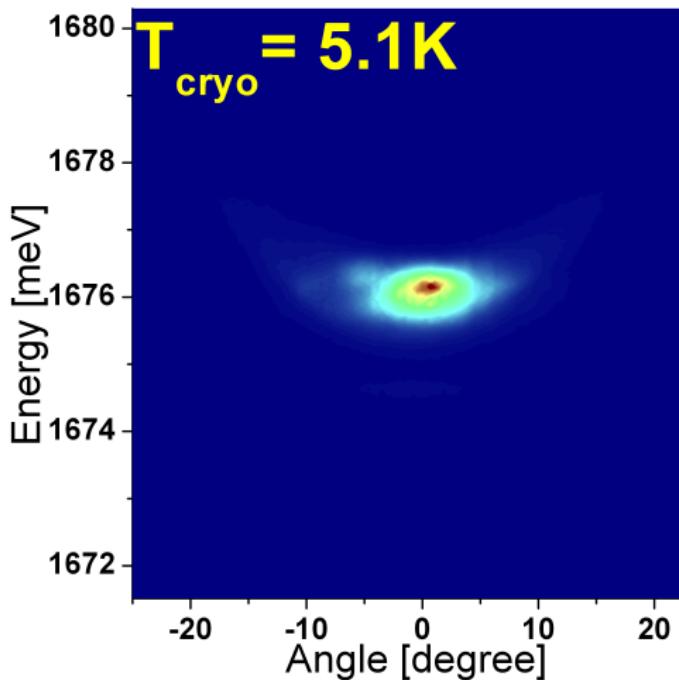
Temperature as a Control Parameter
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Polariton Condensation in Dispersion Plane

Temperature as a Control Parameter

$P = \text{const}$, T_C Exists !



Plan

1 Condensation Phenomenon

2 Why Polariton Condensation ?

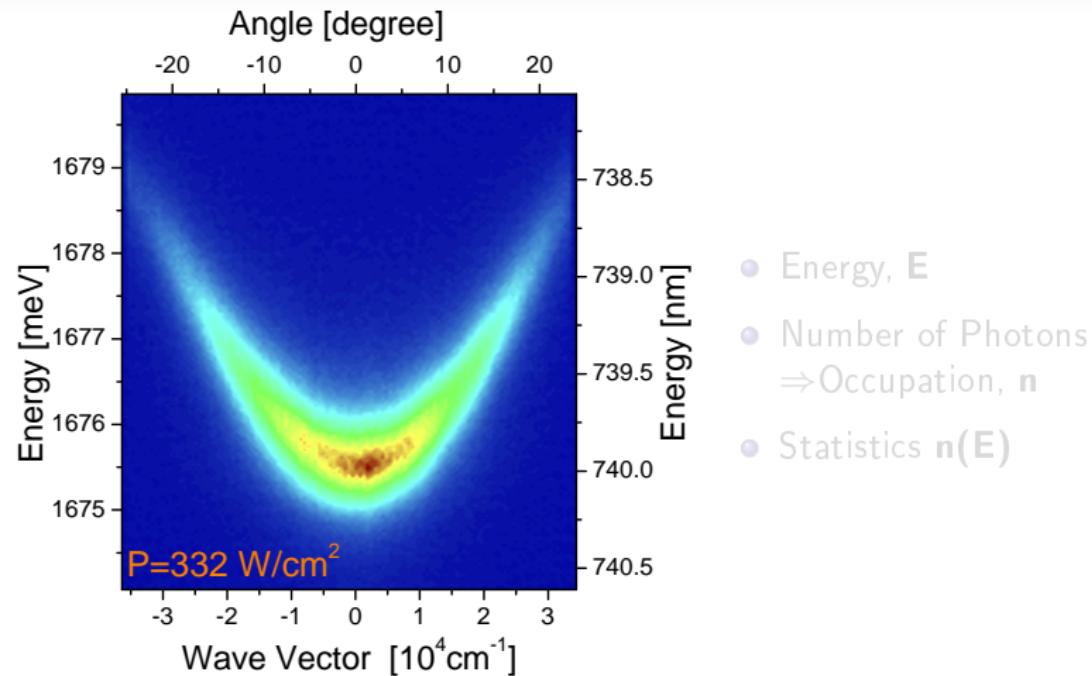
3 Demonstration of the Polariton Condensation

- Massive Occupation (N, T)
- Thermalization
- Long Range Spatial Coherence

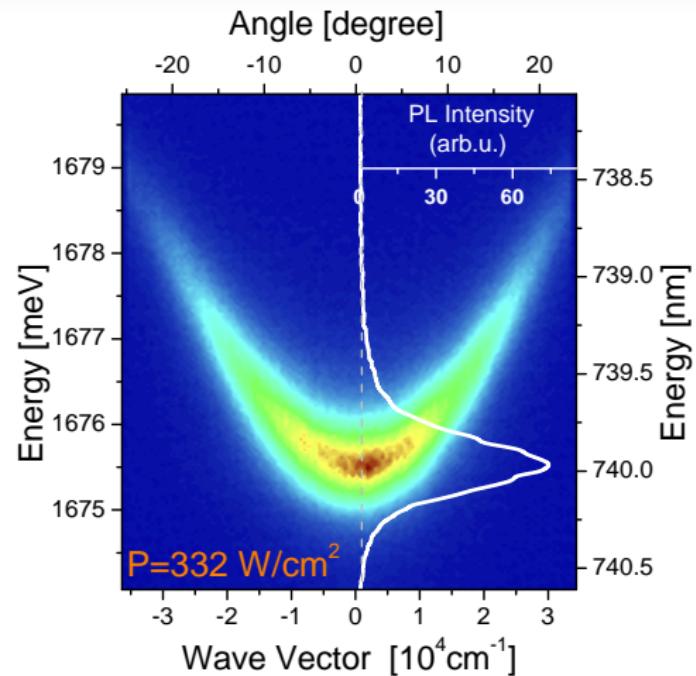
4 Discussion

- Single State
- Quantum State

Thermalized Polariton Gas ??

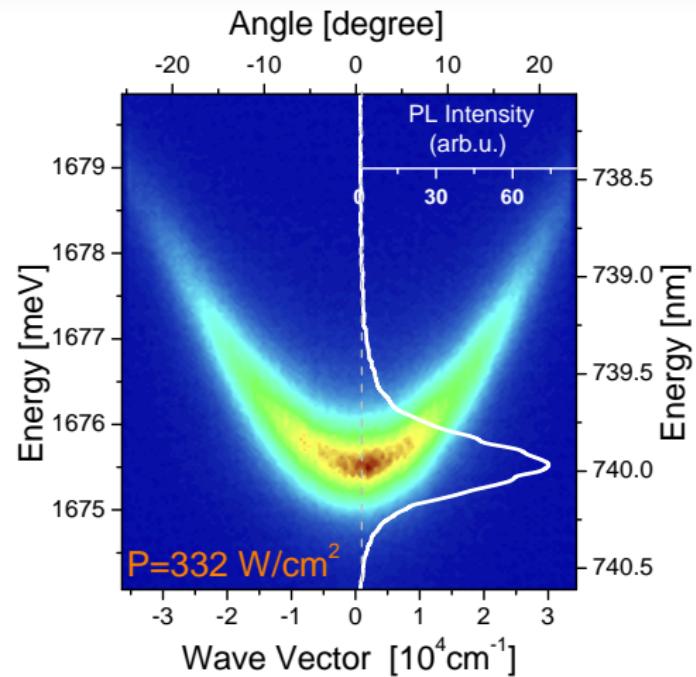


Thermalized Polariton Gas ??



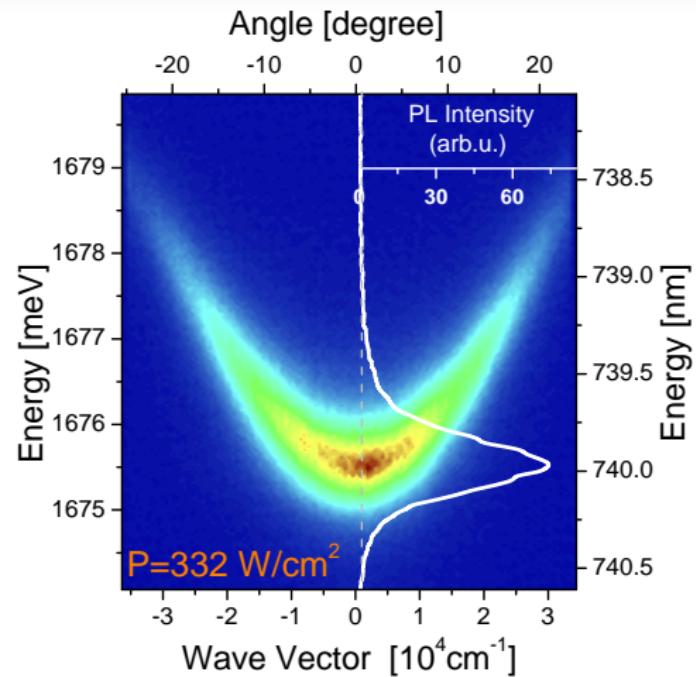
- Energy, E
- Number of Photons
→ Occupation, n
- Statistics $n(E)$

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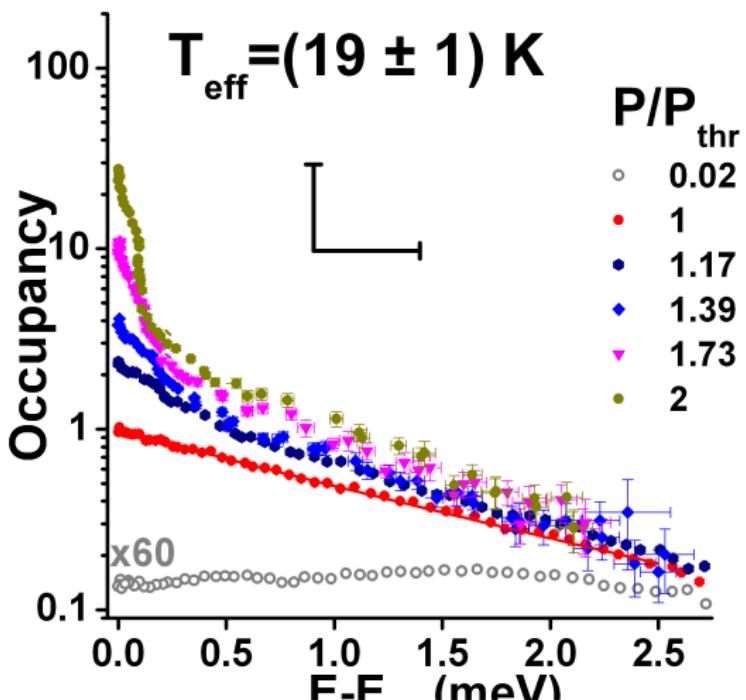


- Energy, \mathbf{E}
- Number of Photons
⇒ Occupation, \mathbf{n}
- Statistics $\mathbf{n}(\mathbf{E})$

Internal Temperature of the Polariton Gas

... In from the cold: $T_{C\text{Polariton}} \simeq 100\,000\,000 \times T_{C\text{Atom}}$

Critical Density: $N_{C\text{Polariton}} \simeq 5 \times 10^8 \text{ cm}^{-2}$



Plan

1 Condensation Phenomenon

2 Why Polariton Condensation ?

3 Demonstration of the Polariton Condensation

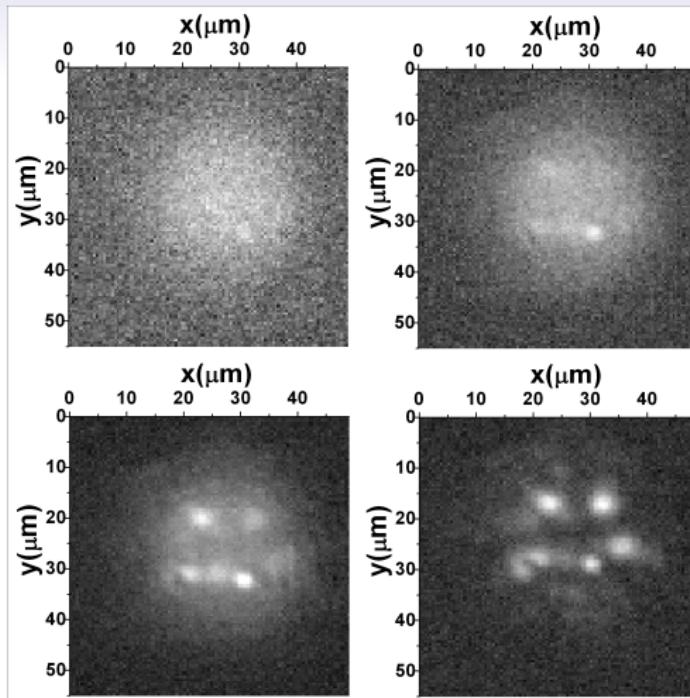
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Condensation in Real Space

Condensation in Real Space

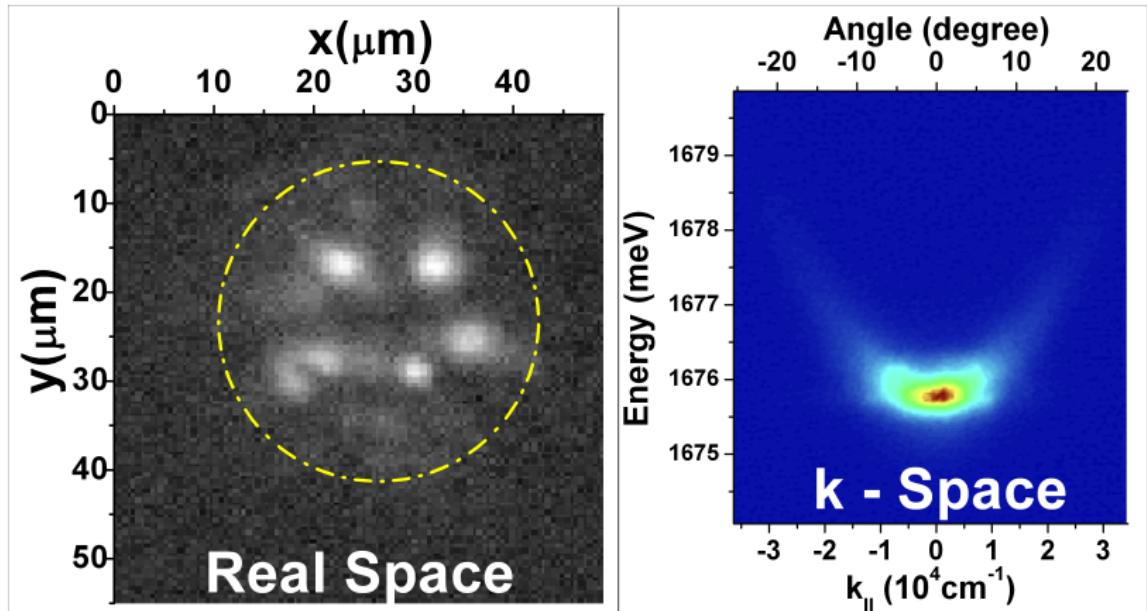


Spatial Localization by the Photonic Disorder

Long Range Spatial Coherence



M. Richard et al. PRB 72, 201301(R), 2005



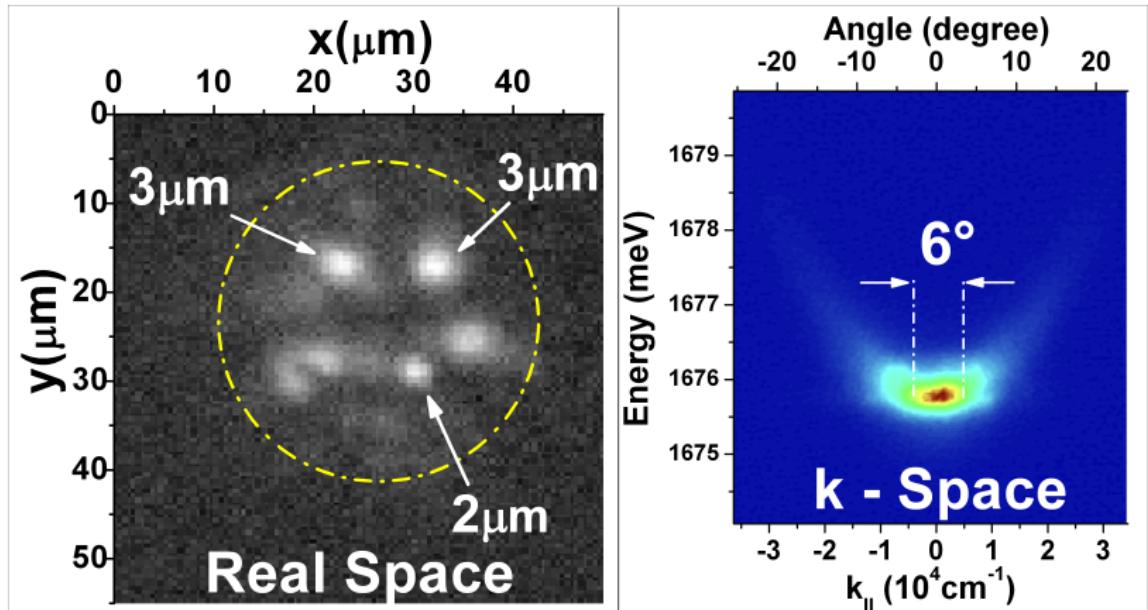
Mutual Coherence Between Localized Spots

Exploring coherence in solids

Long Range Spatial Coherence



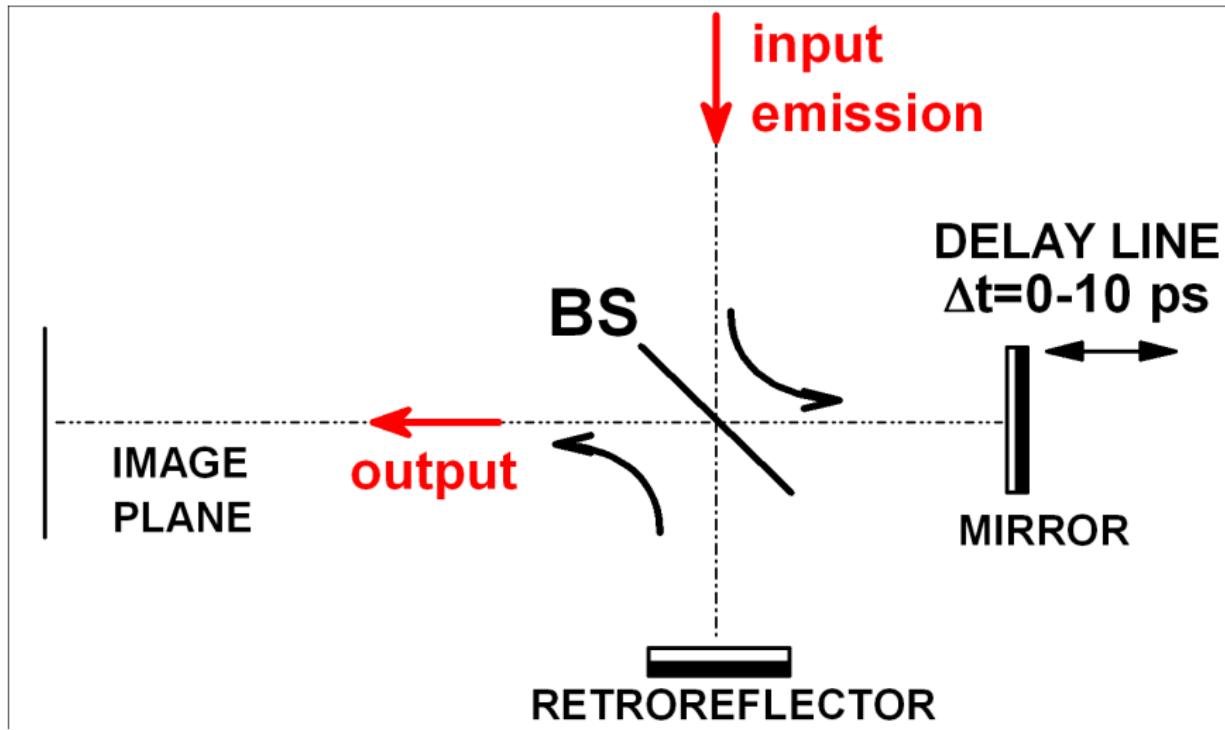
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Mutual Coherence Between Localized Spots

Long Range Spatial Coherence

Michelson Interferometer

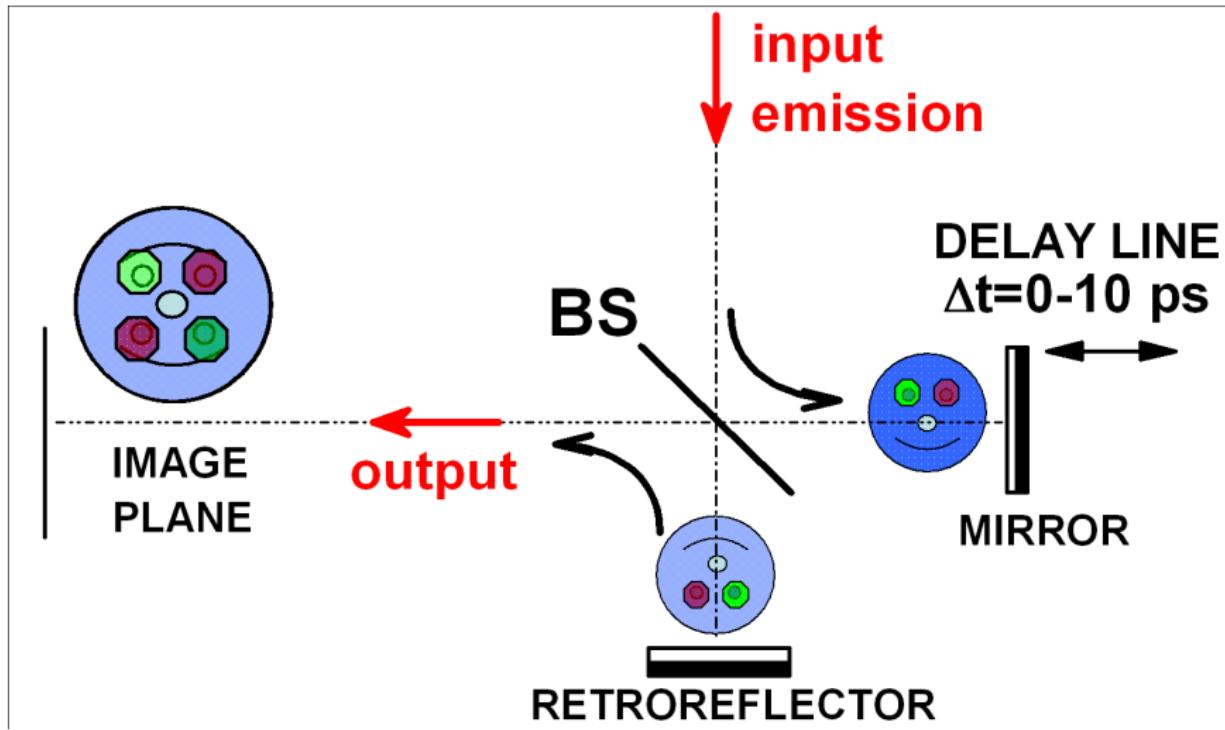


RETROREFLECTOR

Exploring coherence in solids

Long Range Spatial Coherence

Michelson Interferometer



RETROREFLECTOR

Exploring coherence in solids

Long Range Spatial Coherence

Interference over the Entire Excitation Spot



+

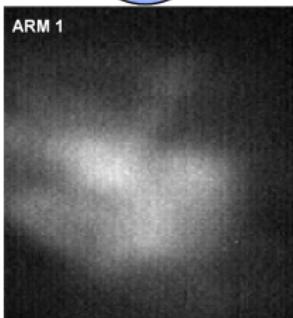


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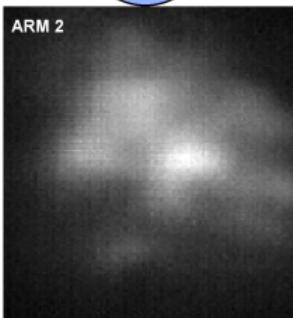


$1.9P_{thr}$

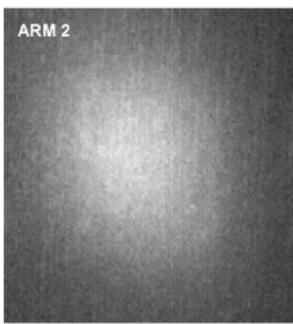
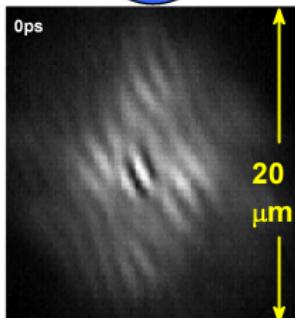
$0.3P_{thr}$



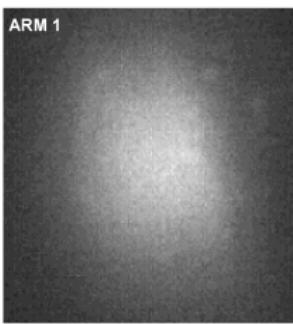
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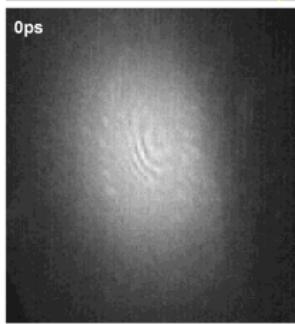
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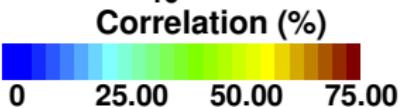
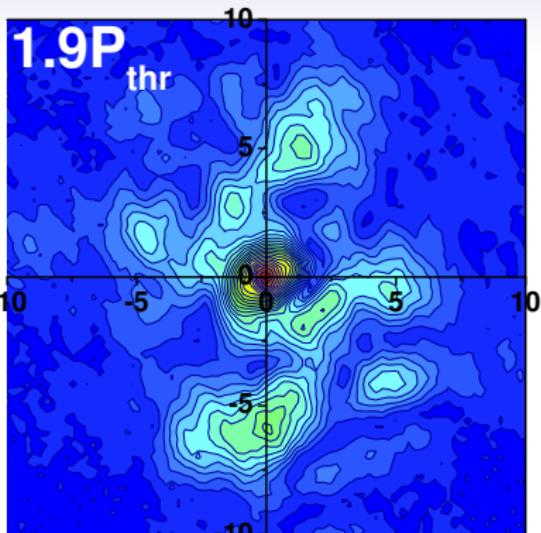
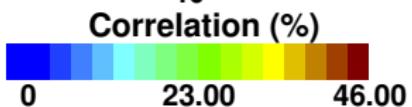
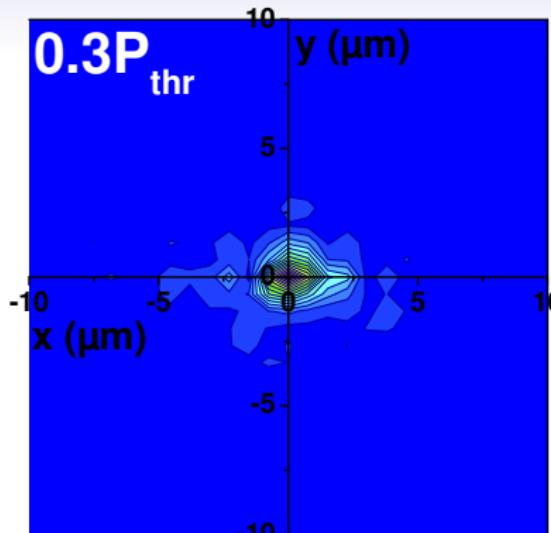
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Long Range Spatial Coherence

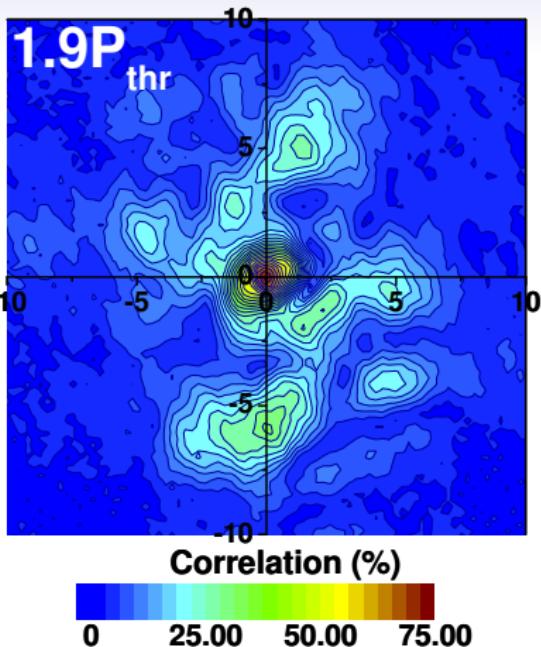
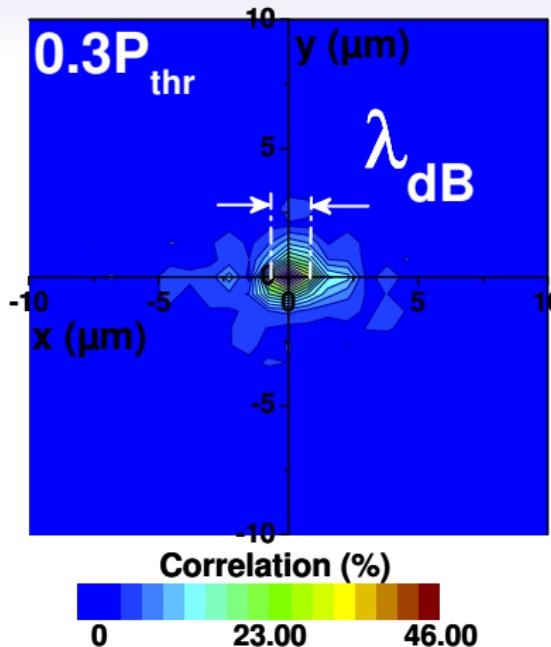
Probing of the Phase...

Mapping of the First Order Correlation



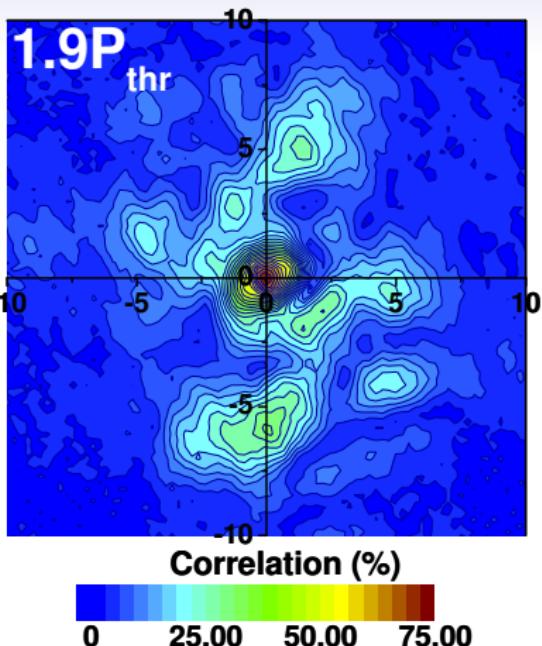
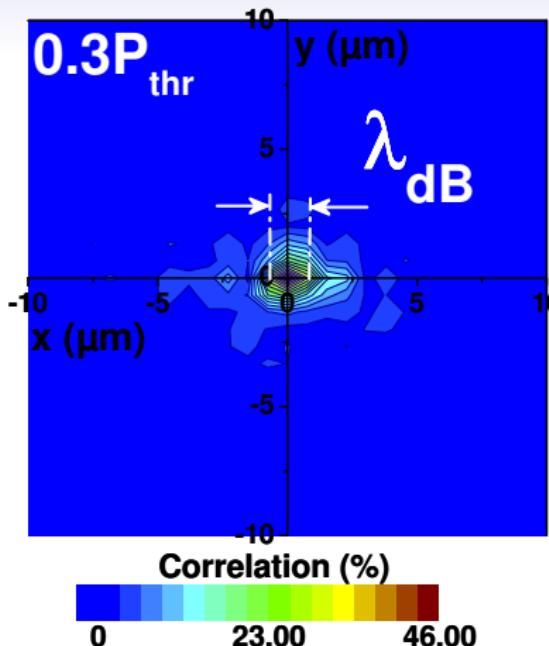
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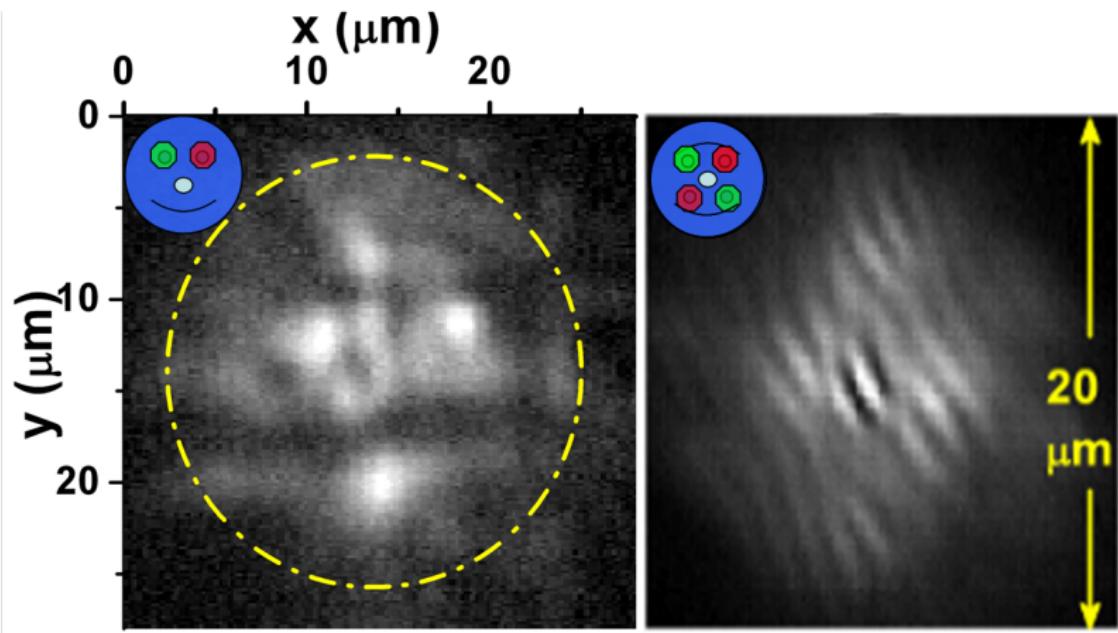
Outline

- 1 Condensation Phenomenon
- 2 Why Polariton Condensation ?
- 3 Demonstration of the Polariton Condensation
 - Massive Occupation (N, T)
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- 4 Discussion
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Condensation into the Single State ??



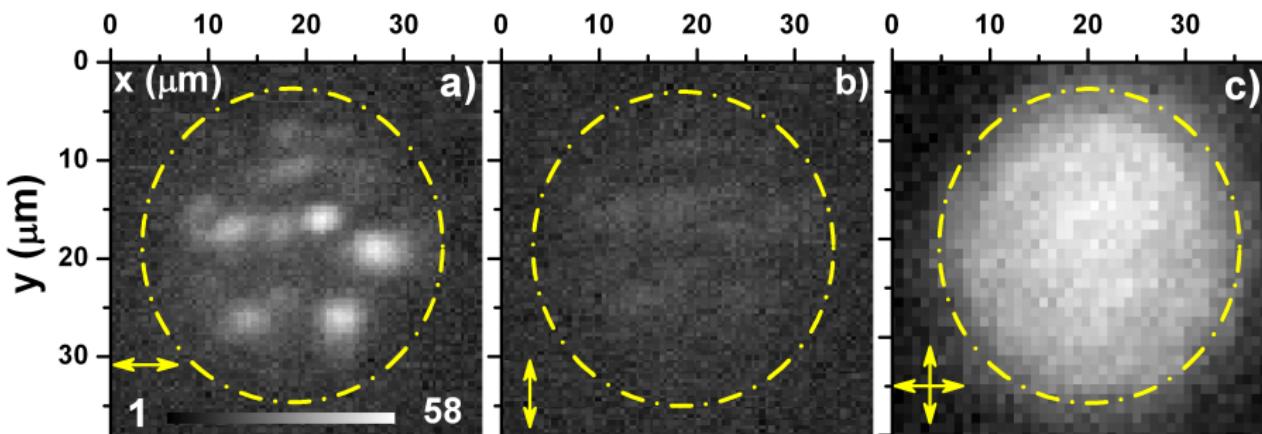
Stationary Interference Pattern !!

Exploring coherence in solids

Condensation into the Single State ??

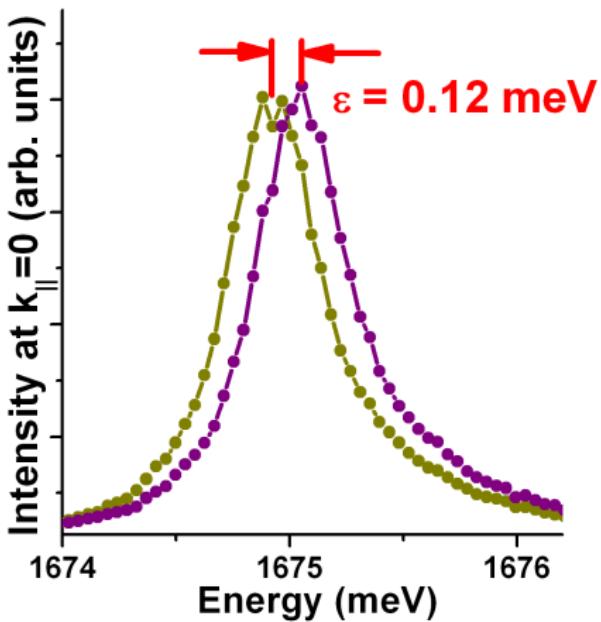
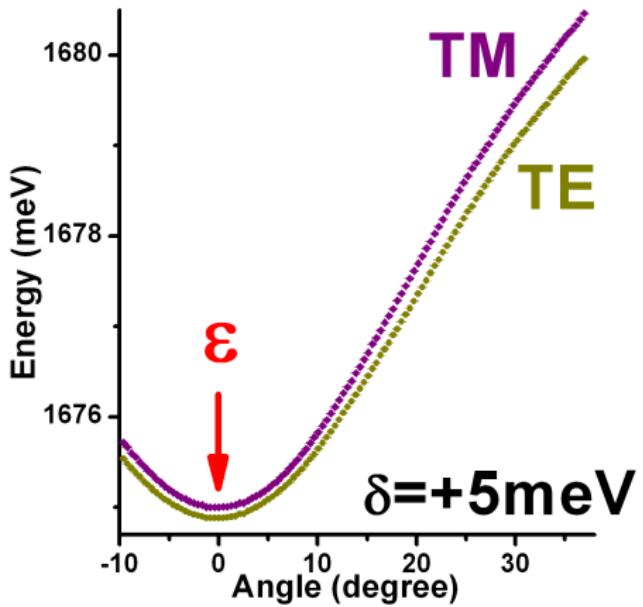
Stationary Interference Pattern !!

Condensation into the Single State ??



Homogenous Linear Polarization

Condensation into the Single State ??



Ground State Splitting

$$\epsilon \simeq 0.12 \text{ meV} \ll kT \simeq 1.6 \text{ meV}$$

Exploring coherence in solids

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Condensation into the Quantum State ??

Photon Statistics \Rightarrow Intensity Correlation

Second Order Coherence

$$g_2(\tau) \propto \langle I(t) \cdot I(t + \tau) \rangle_t$$

Source

- Coherent
- Thermal

Statistics

- Poisson
- B-E

$g_2(\tau = 0)$

- 1
- 2

$g_2(\tau = \infty)$

- 1
- 2

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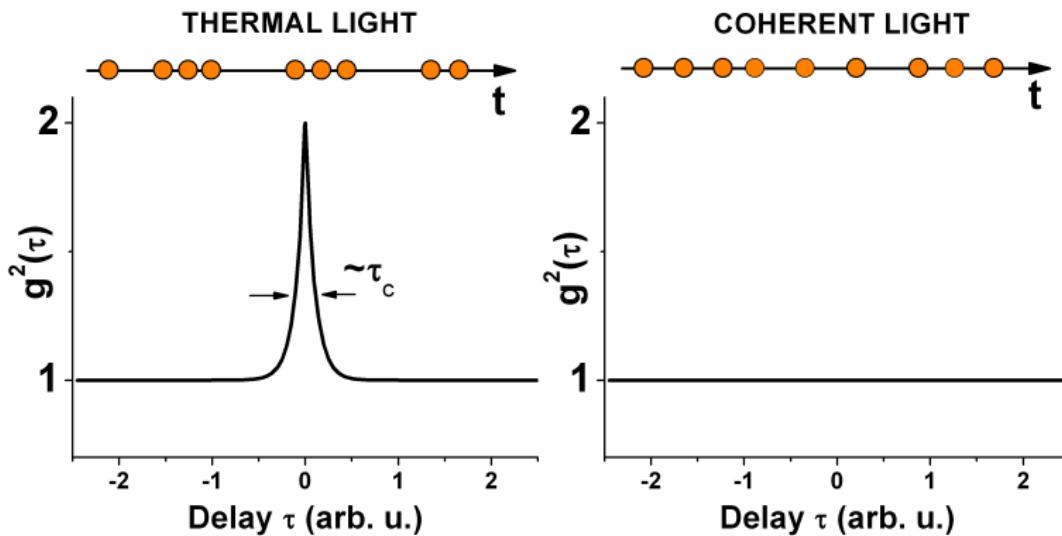
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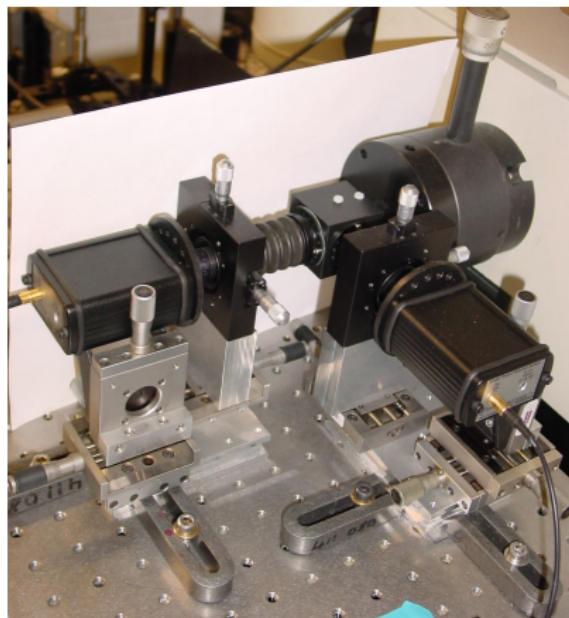
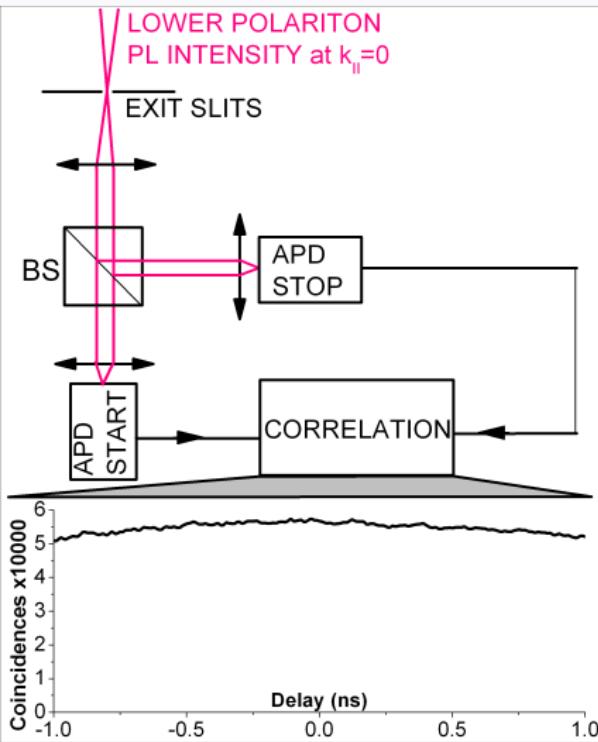
Condensation into the Quantum State ??

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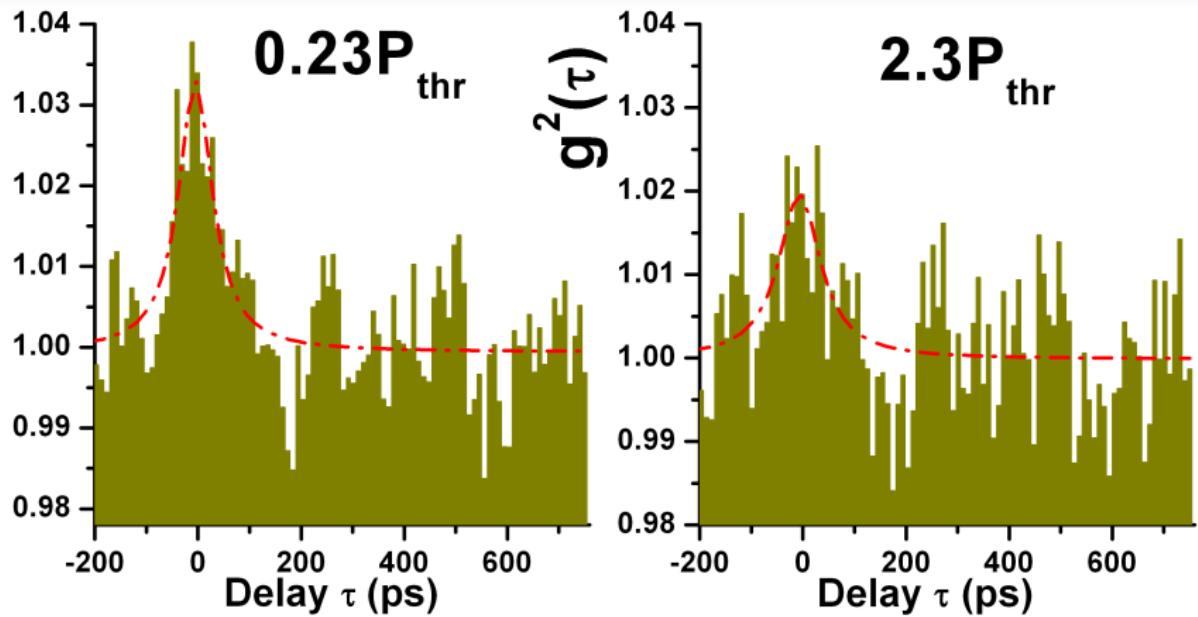


Intensity Correlation

Experimental Setup

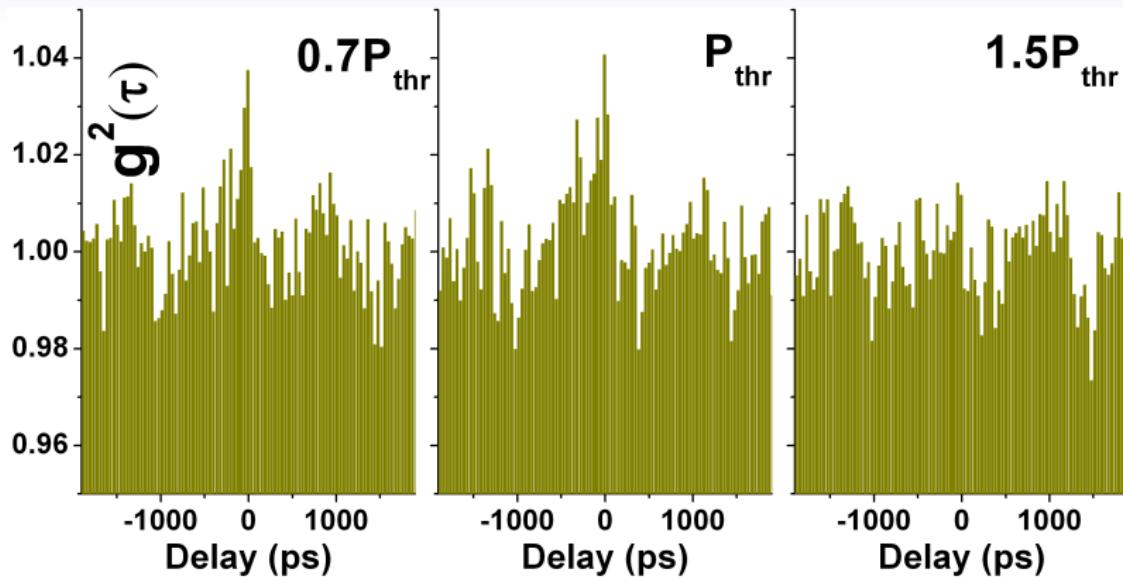


Intensity Correlation Reduction of the Bunching



Build up of the Second Order Coherence

Intensity Correlation



Transition from Thermal towards Coherent State

Conclusions

Realization of the Polariton Condensation

- Thermalization $T_{\text{eff}} = (16 - 20)K$
- Massive Occupation of the Ground State
- Saturation of the Excited States
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- Homogenous, Linear Polarization of the Condensate
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Alternatives for spontaneous coherence in solids

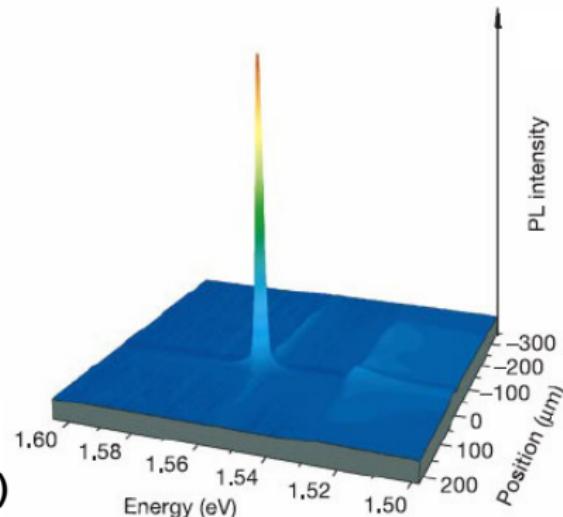
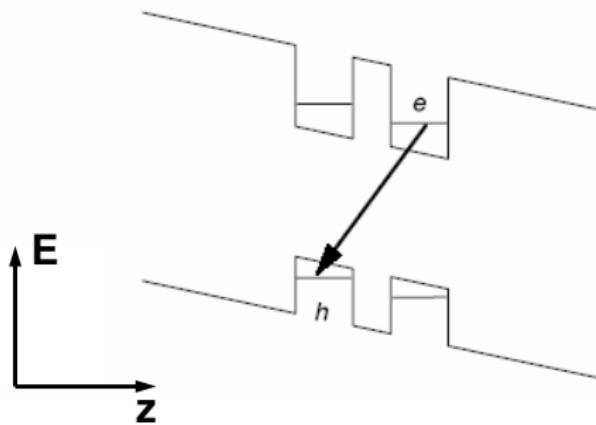
Indirect excitons in coupled quantum wells

Spatial Separation

Long Lifetime: $10^{-7} s$

Coherence, Statistics

?????



Butov et. al., Nature, 214, 47 (2002)

Alternatives for spontaneous coherence in solids

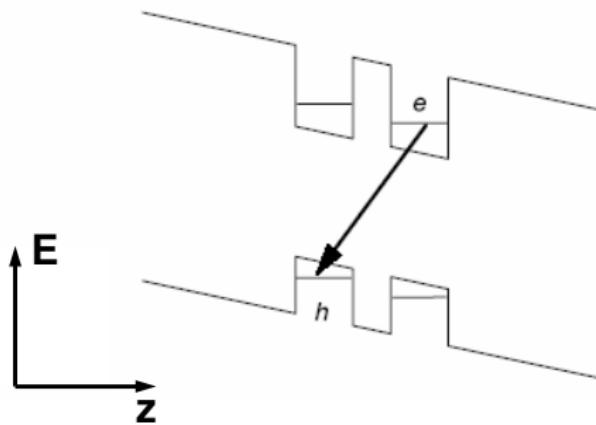
Indirect excitons in coupled quantum wells

Spatial Separation

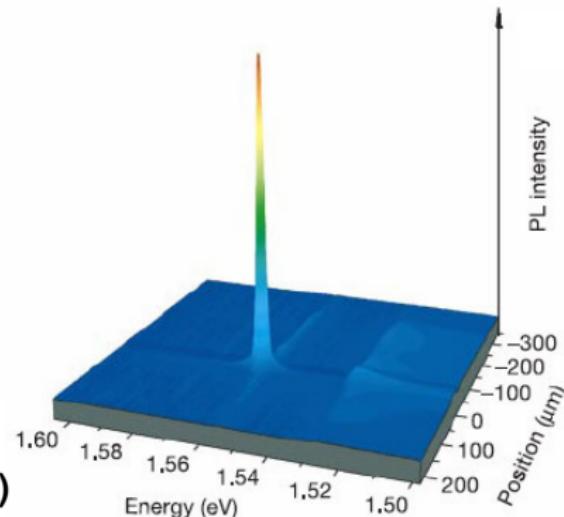
Long Lifetime: $10^{-7} s$

Coherence, Statistics

?????

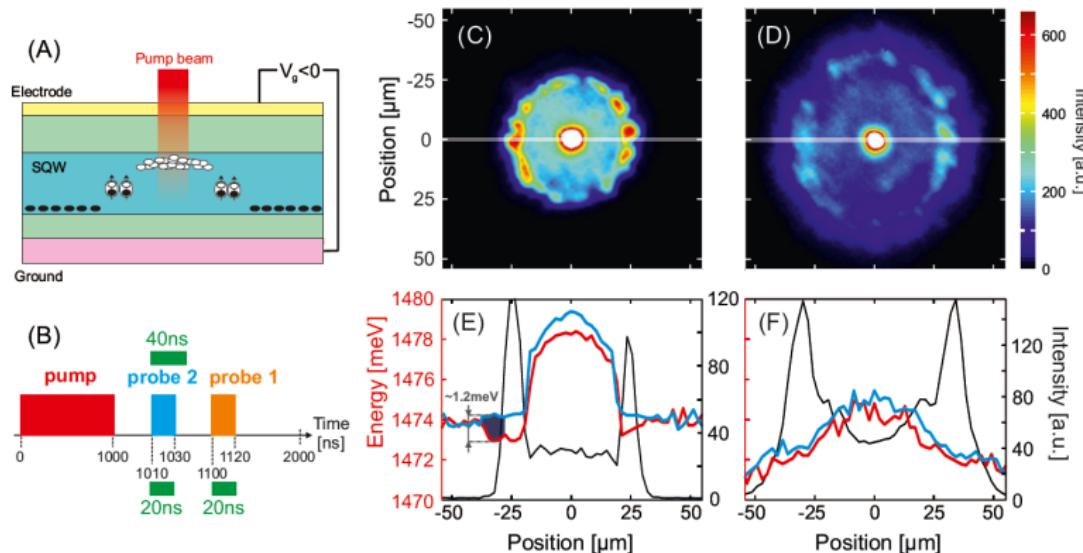


Butov et. al., Nature, 214, 47 (2002)



Alternatives for spontaneous coherence in solids

Excitons in a 25 nm GaAs QW in a field-effect device:
electrons and holes are spatially separated



Evidence for a BEC of excitons at 500 mK, EPL (2014)

Exciton gas in spatial traps, linear polarization, spatial coherence

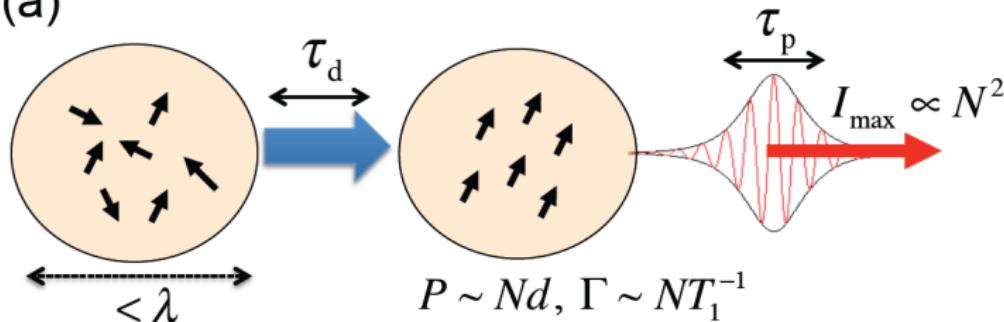
Alternatives for spontaneous coherence in solids

Superradiance \Rightarrow spontaneous and self-organized build up of coherent radiation within an ensemble of quasi-degenerate emitters [M. Gross and S. Haroche, Physics Reports 93, 301 (1982)]. It was first noted by R. Dicke back in 1954 [Phys. Rev. 93, 99 (1954)] that, with increasing their density, the collection of N emitters starts to radiate much faster and stronger comparing to spontaneous emission of individuals or their diluted ensemble. More precisely, when packing up N identical emitters into a volume of size much smaller than the radiation wavelength, instead of observing isotropic and exponentially decaying emission, one produces a fierce, directional radiation blast, having a lagged peak intensity scaling like N^2 and N-times reduced duration with respect to the spontaneous emission.

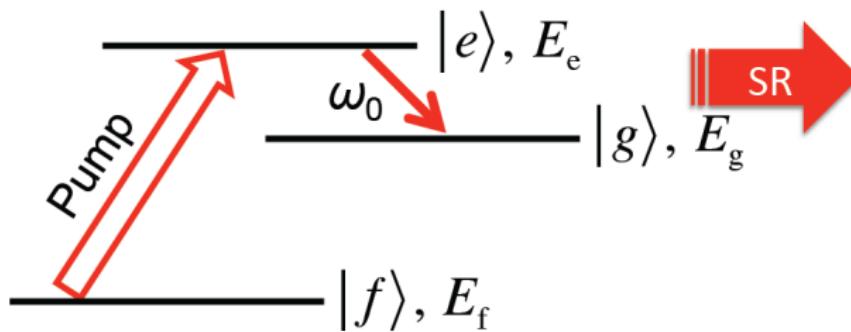
Alternatives for spontaneous coherence in solids

Superradiance in Solids, J. Kono JOSAB (2016)

(a)

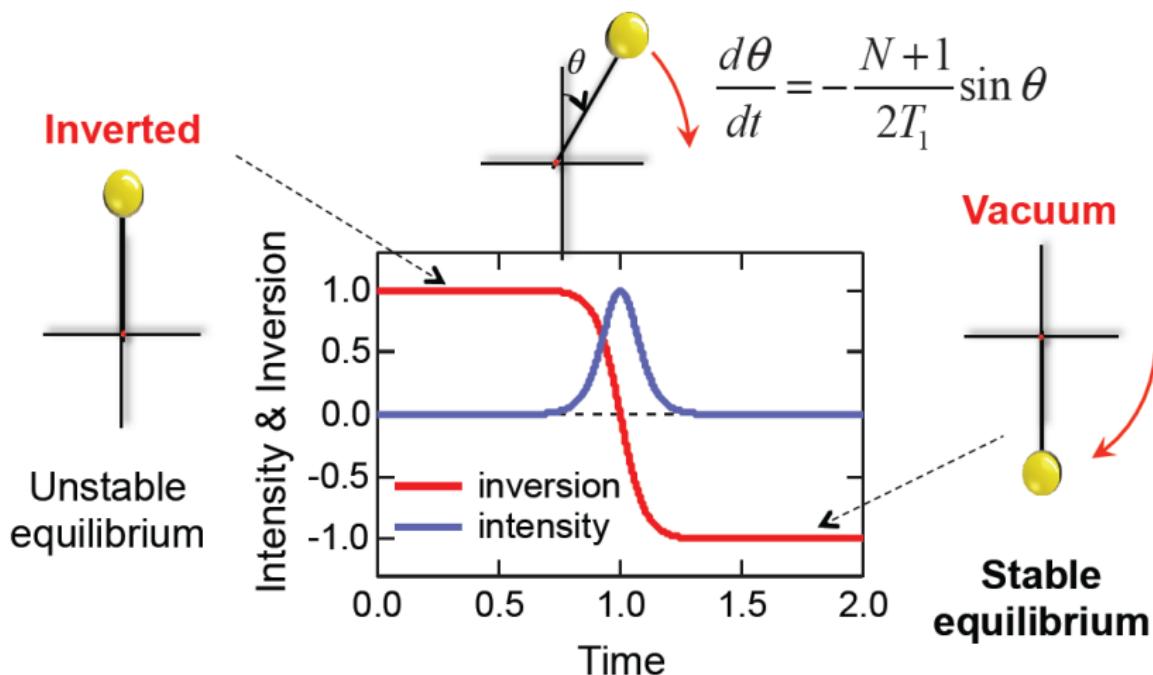


(b)



Alternatives for spontaneous coherence in solids

Superradiance in Solids, J. Kono JOSAB (2016)



Alternatives for spontaneous coherence in solids

Superradiance in Solids: most convincing examples

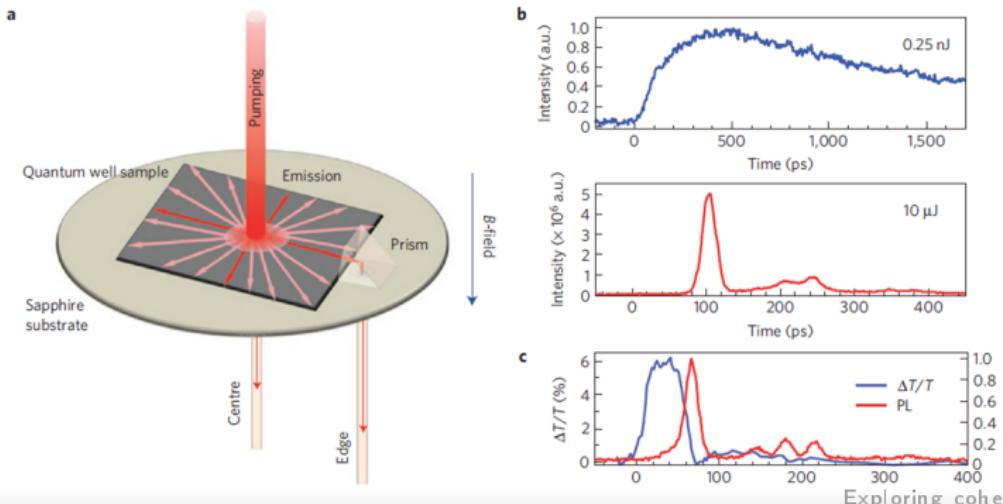
nature
physics

ARTICLES

PUBLISHED ONLINE: 29 JANUARY 2012 | DOI: 10.1038/NPHYS2207

Giant superfluorescent bursts from a semiconductor magneto-plasma

G. Timothy Noe II¹, Ji-Hee Kim¹, Jinho Lee², Yongrui Wang³, Aleksander K. Wójcik³, Stephen A. McGill⁴, David H. Reitze², Alexey A. Belyanin³ and Junichiro Kono^{1*}



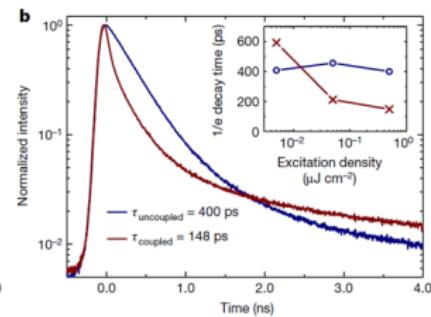
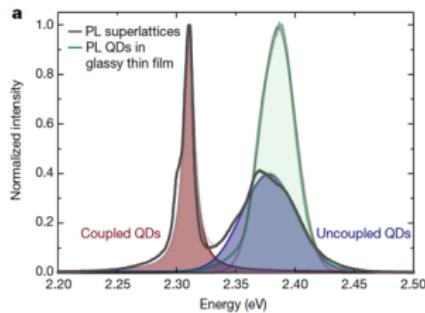
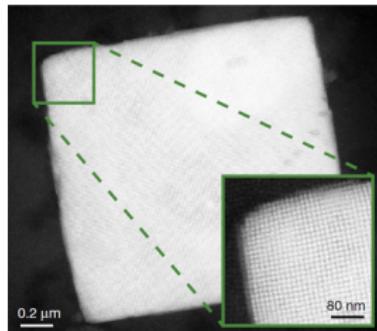
Alternatives for spontaneous coherence in solids

Superradiance in Solids: most convincing examples

LETTER

Superfluorescence from lead halide perovskite quantum dot superlattices

Gabriele Rainò^{1,2,3,5*}, Michael A. Becker^{3,4,5}, Maryna I. Bodnarchuk², Rainer F. Mahrt³, Maksym V. Kovalenko^{1,2*} & Thilo Stöferle^{3*}



Outline

5 Appendix

- Experimental Setup
- Transition Characteristics
- Non-Equilibrium Polariton Condensation
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- Depletion of the Condensate ?
- Dispersion Flattening
- Saturation of the excited states
- Real Space More
- Linear Polarization Build-up
- Coherence More
- g^2
- High Temperature Limit
- Polaritons at room temperature
- Polariton Lasing vs. Photon Lasing (VCSEL)
- Stimulation on the Ring of States

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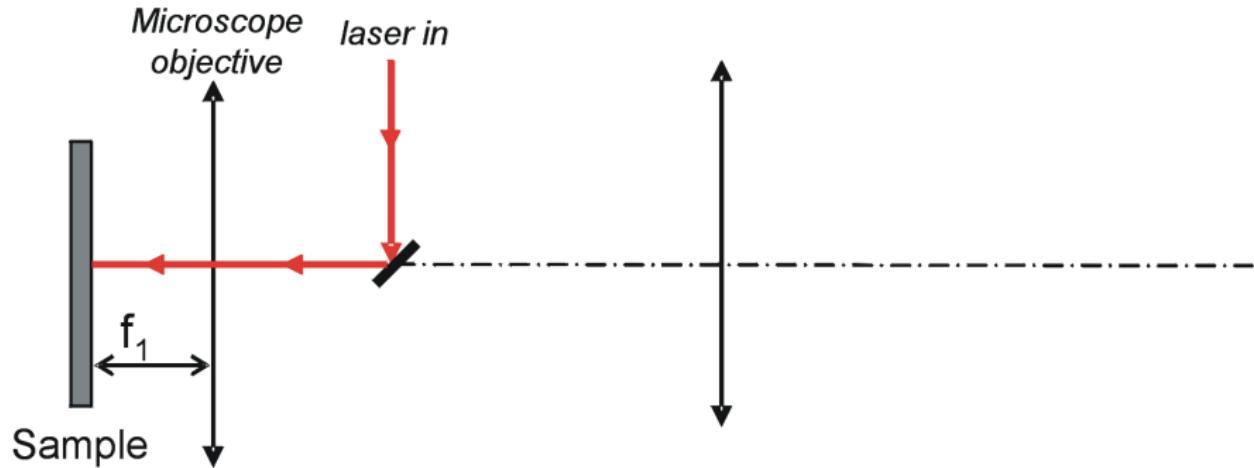
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Imaging

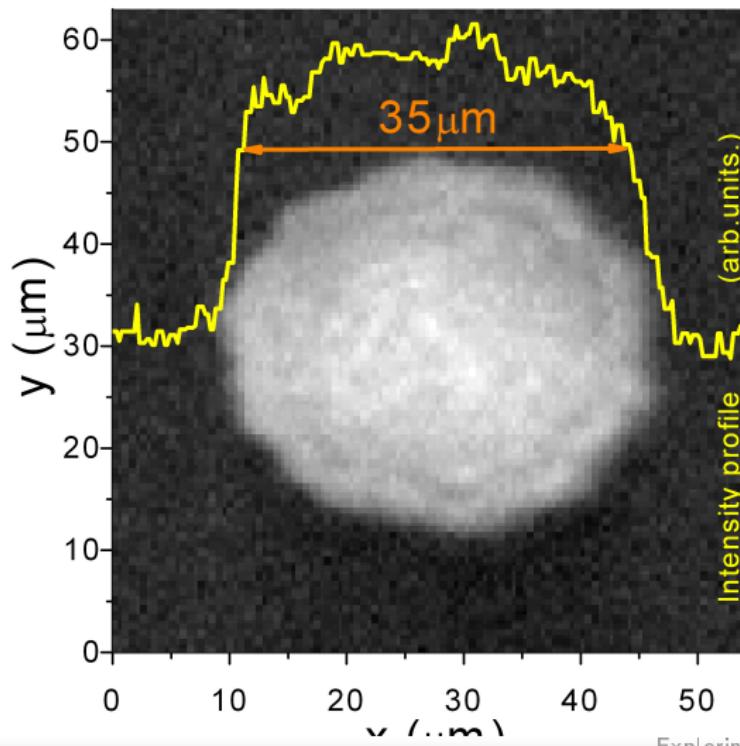
Excitation - Ti:Sapphire, CW



◀ Far Field

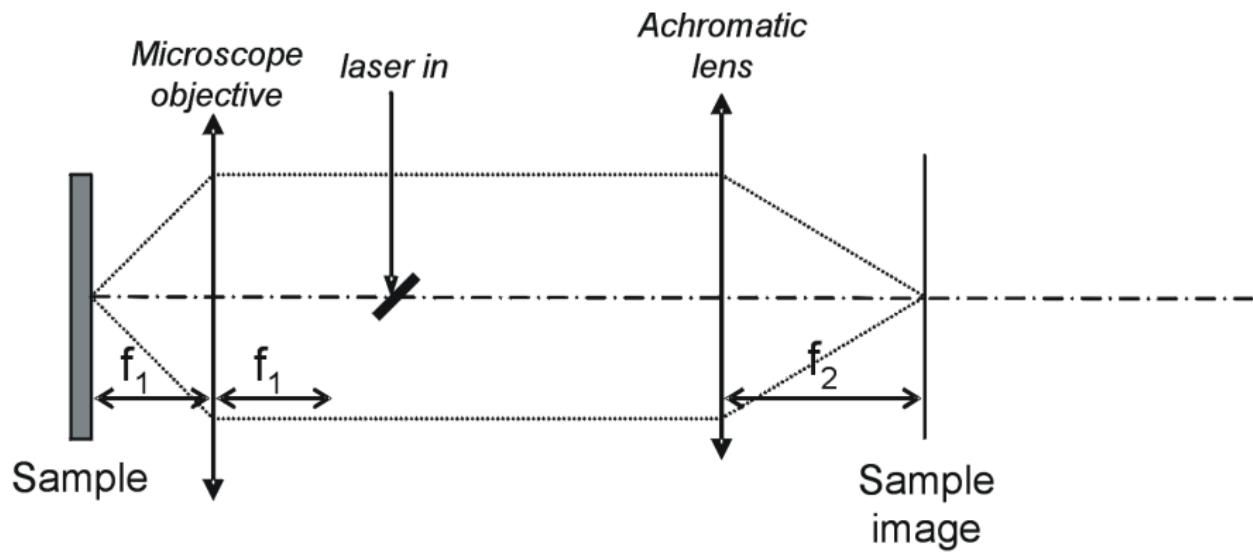
Excitation

"Top Hat" Spot $\sim 30\mu m$



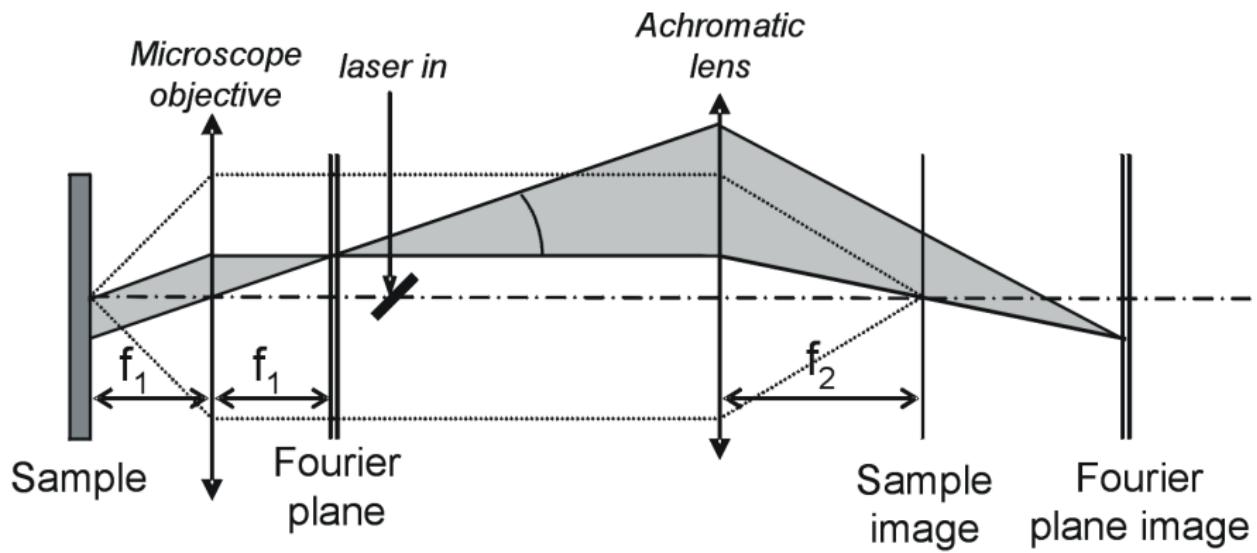
Imaging

Near Field: Sample Image - $f_1 = 9\text{mm}$, N.A. = 0.4



Imaging

Far Field: Fourier Plane Image

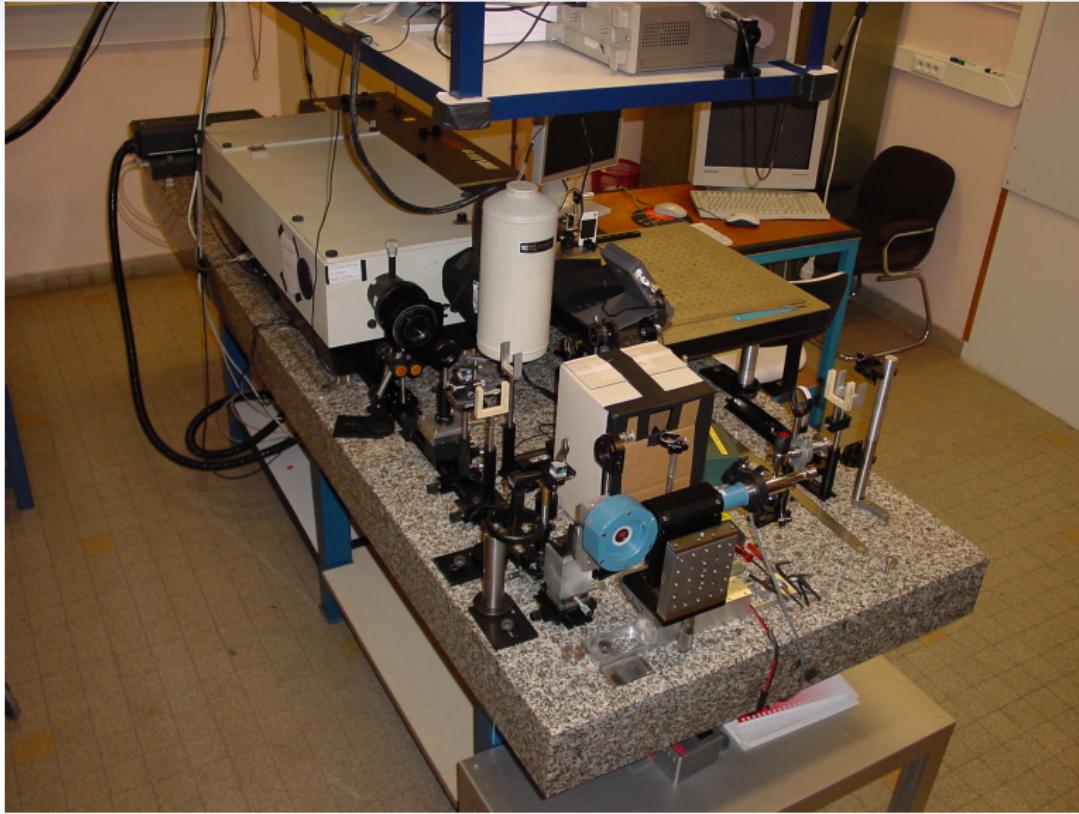


◀ Far Field

Experimental Setup - Photos



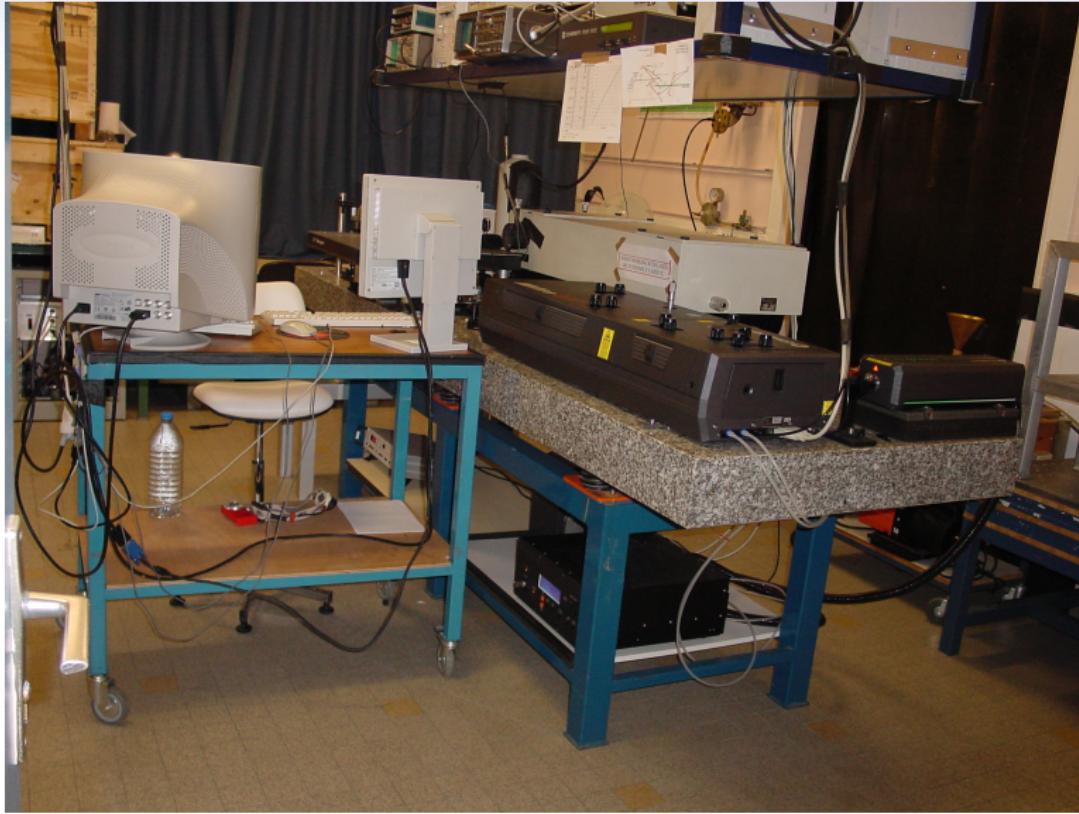
Experimental Setup - Photos



Experimental Setup - Photos



Experimental Setup - Photos



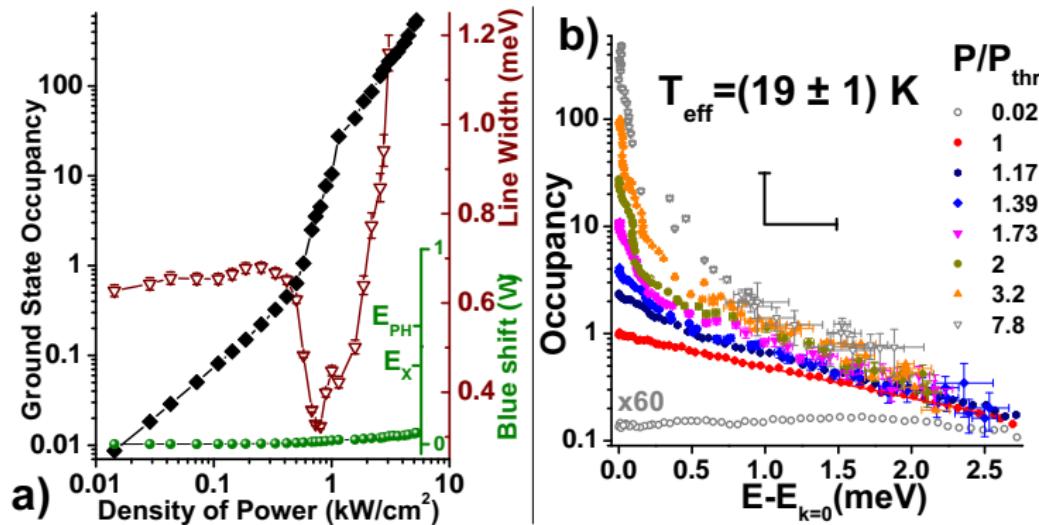
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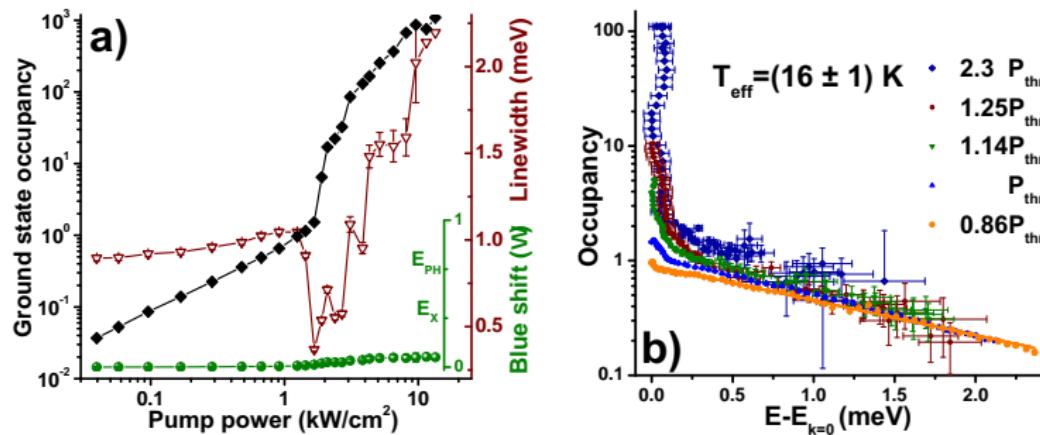
Polariton Condensation in Momentum Space

Threshold, Linewidth, Blueshift, Statistics



Polariton Condensation in Momentum Space

Threshold, Linewidth, Blueshift, Statistics



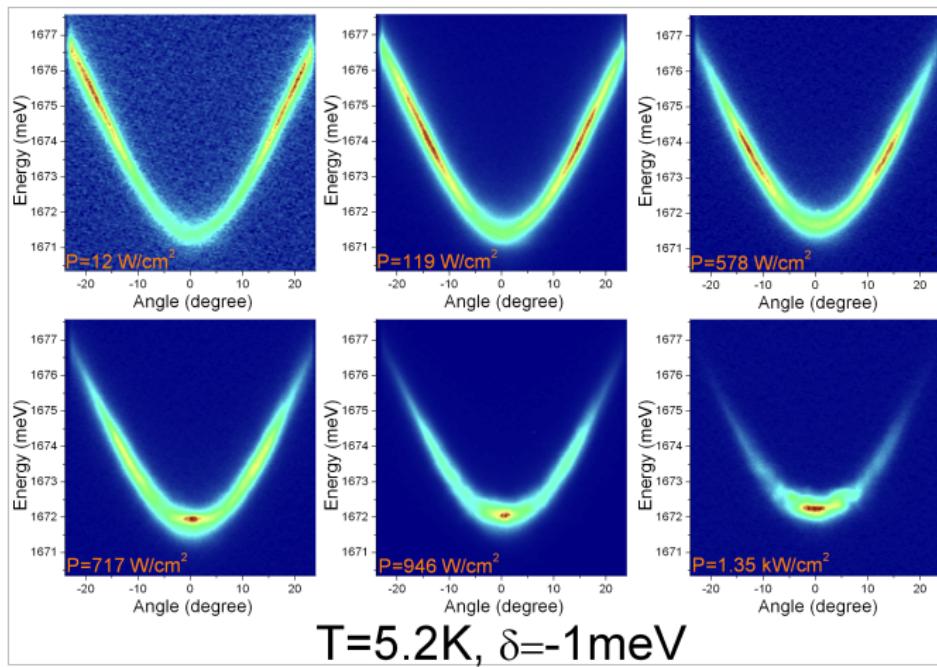
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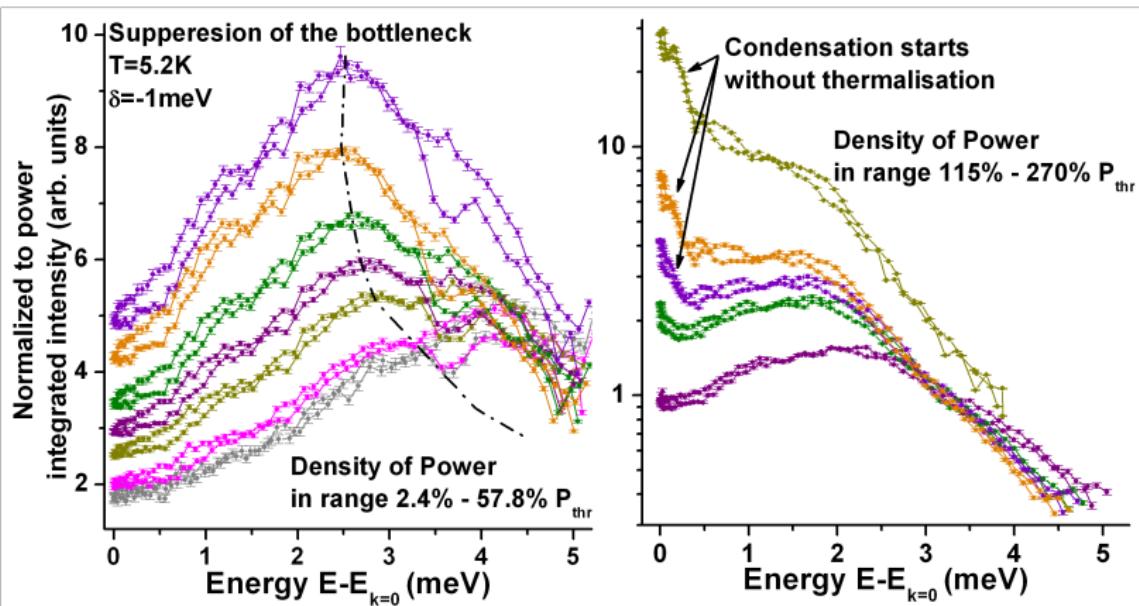
How to get Thermalization ??

Role of Detuning



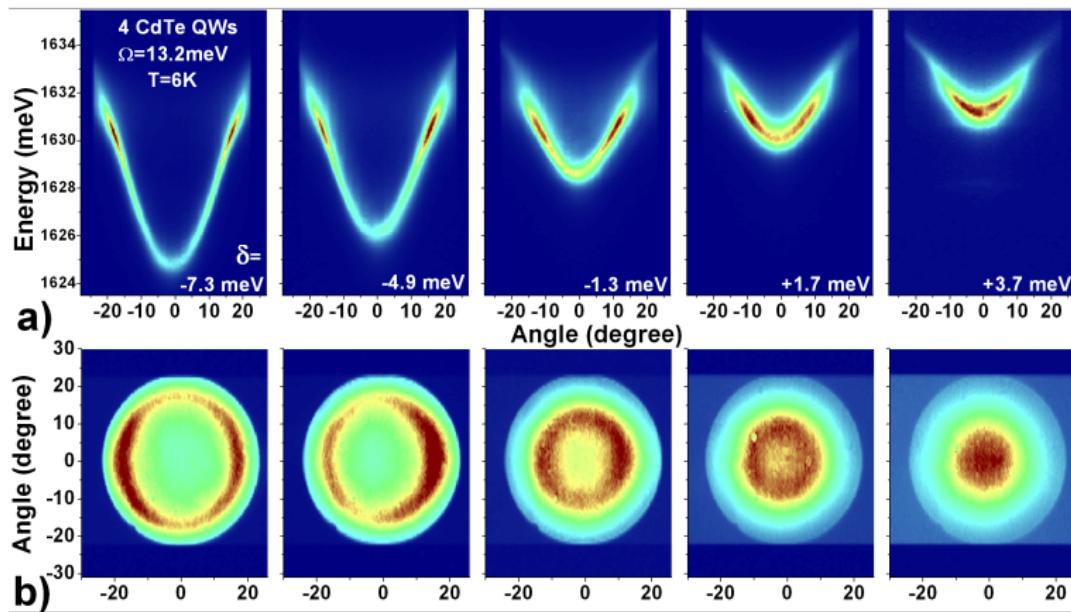
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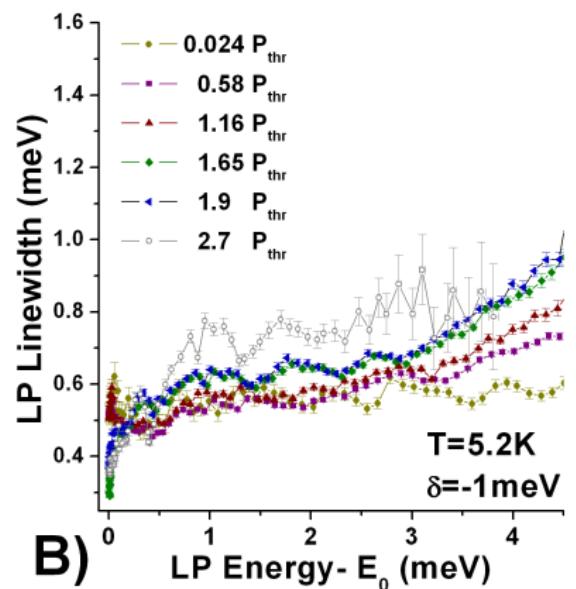
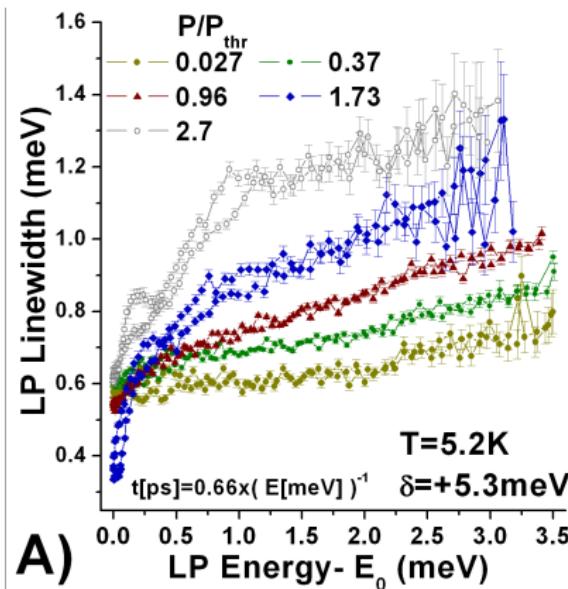
Positive Detuning

Shallower Trap, High X Fraction \Rightarrow Enhanced Relaxation



Positive Detuning

Shallower Trap, High X Fraction \Rightarrow Enhanced Relaxation

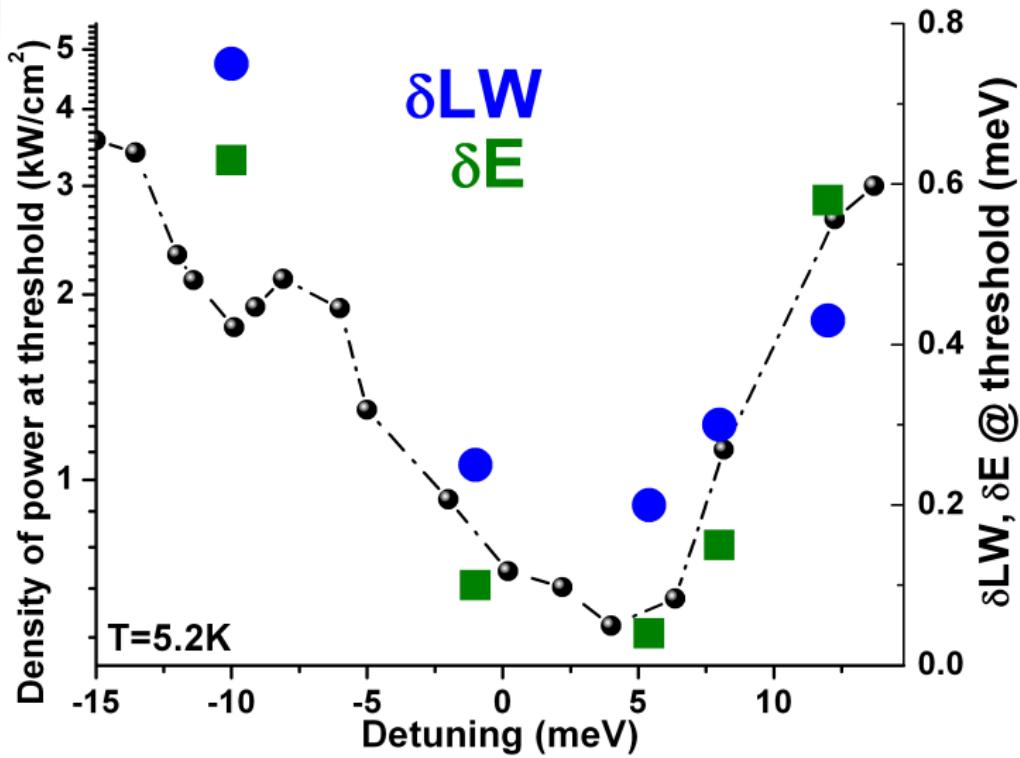


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Influence of the reservoir on the ground state

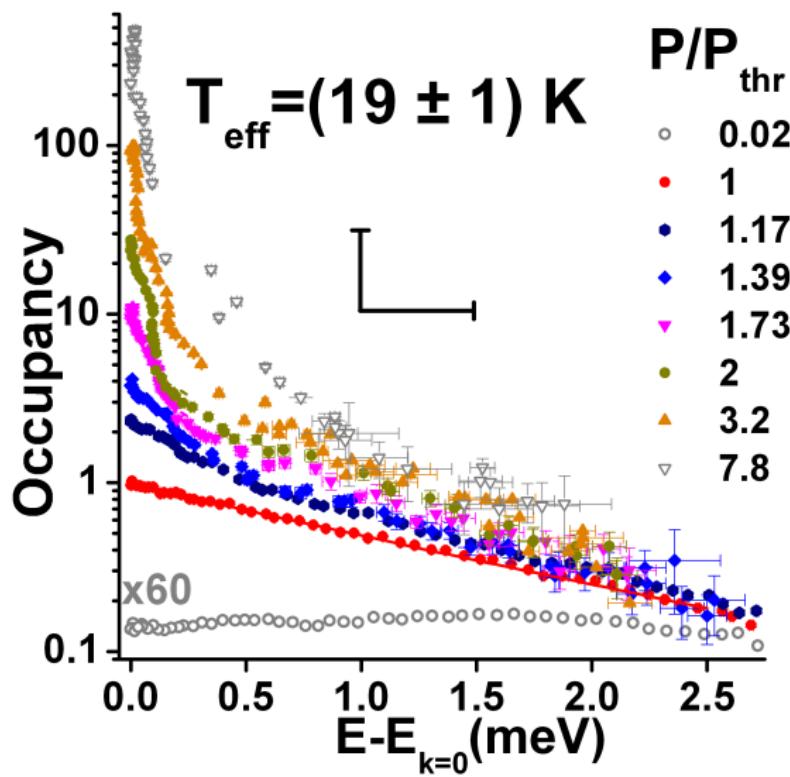


Plan

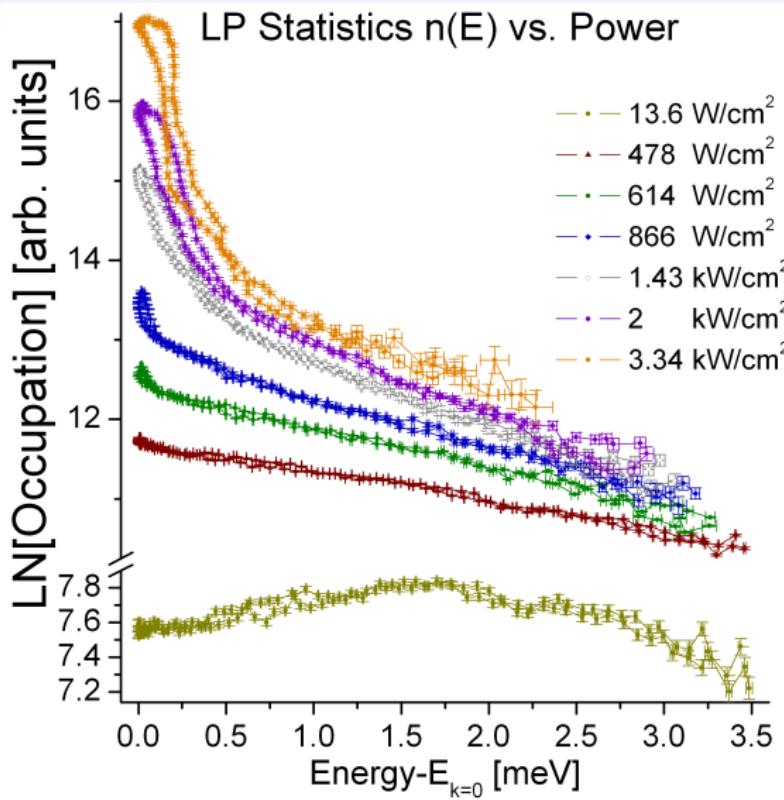
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Depletion of the Condensate ??



Depletion of the Condensate ??



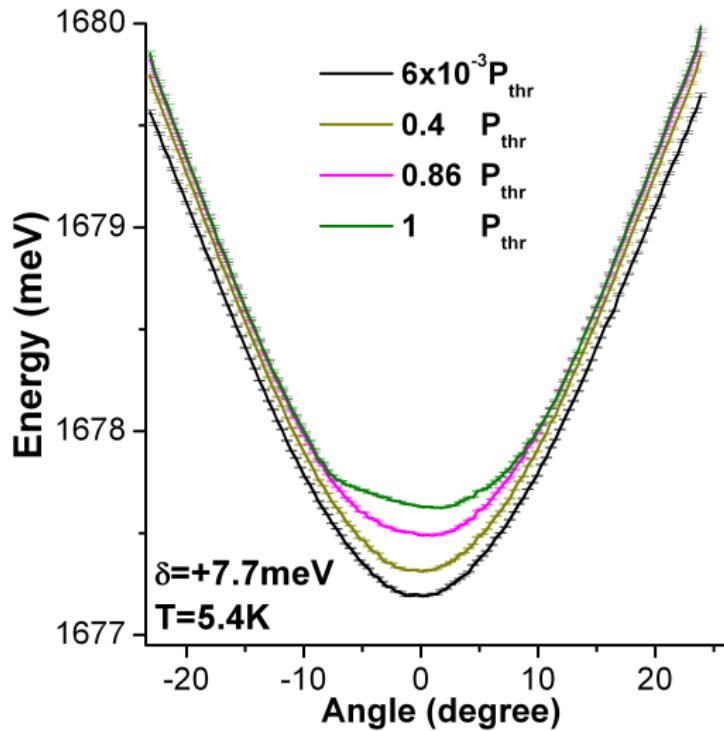
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Dispersion Flattening at k=0

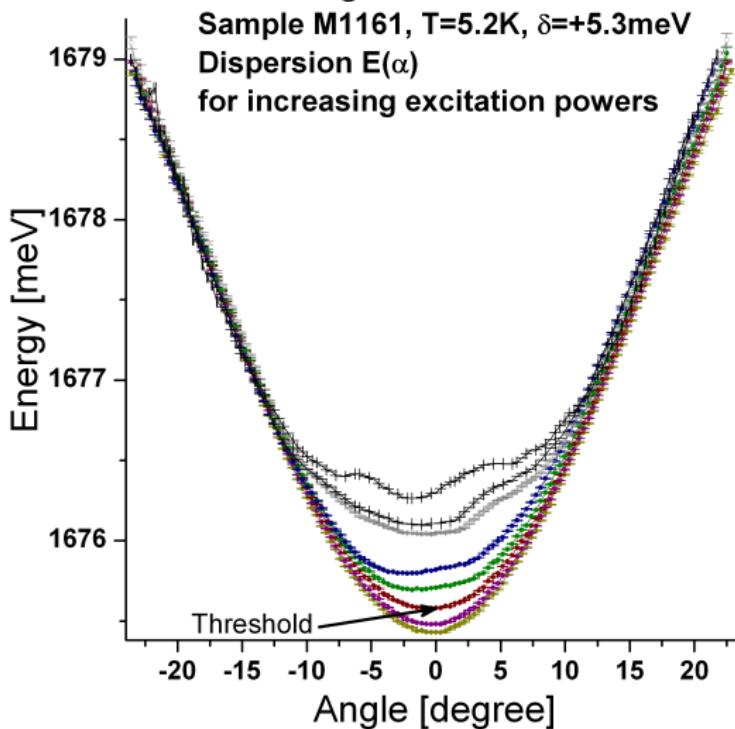
Localization, Non-equilibrium effects ??
Not a rigid shift !



Dispersion Flattening at $k=0$

Localization, Non-equilibrium effects ??

Not a rigid shift !



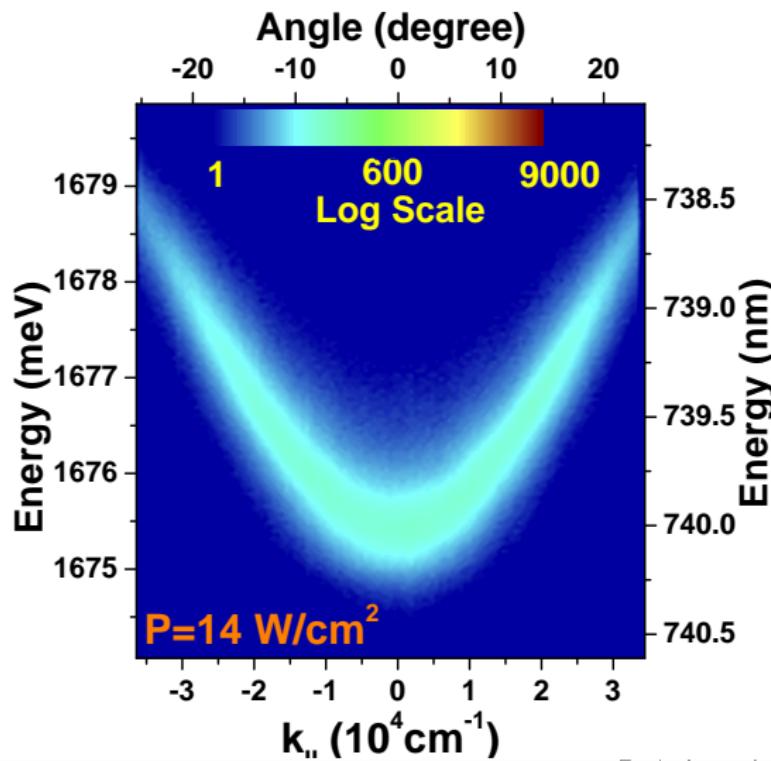
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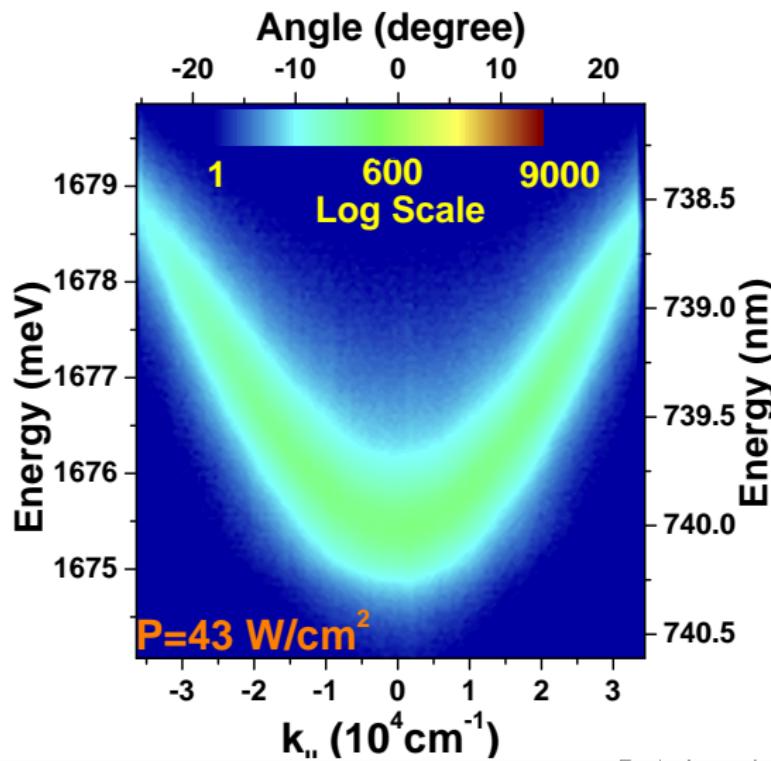
Polariton Condensation in Momentum Space

Saturation of Excited States [◀ Saturation](#)



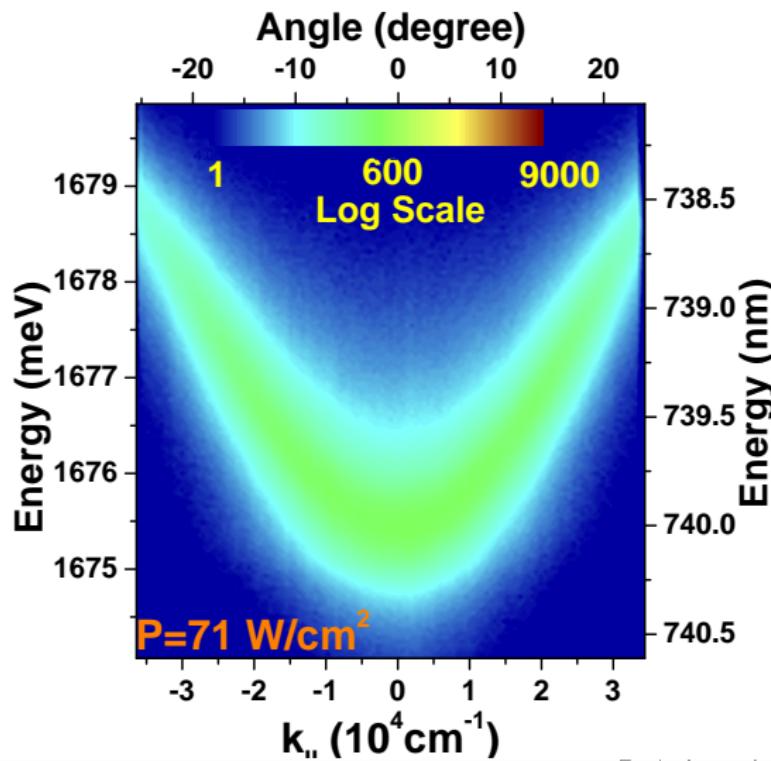
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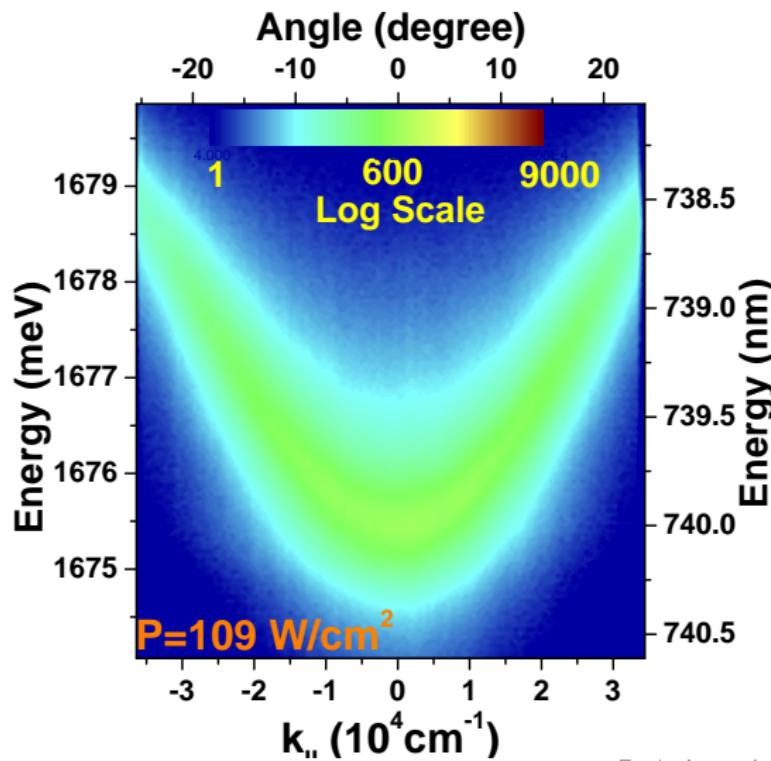
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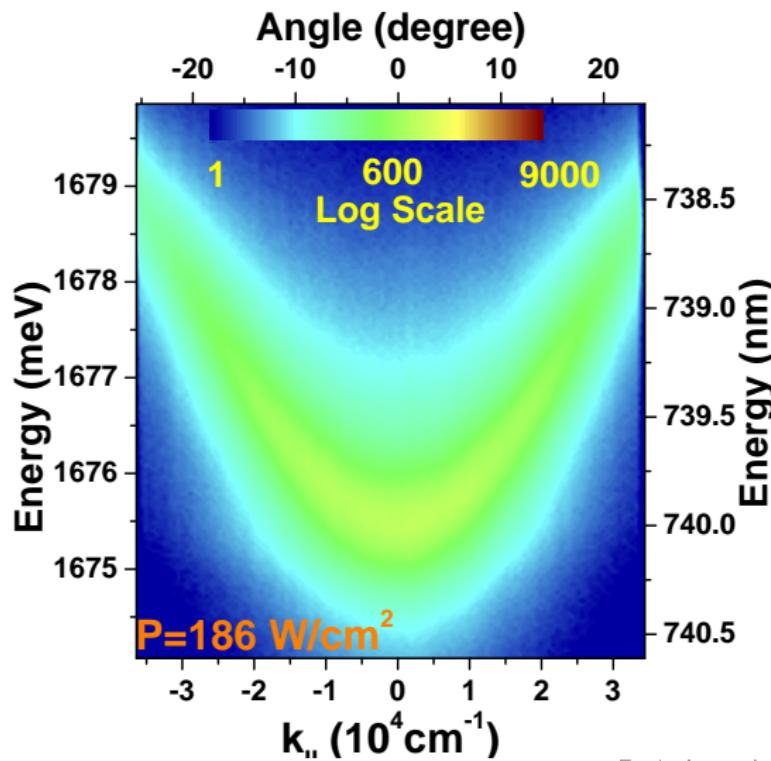
Polariton Condensation in Momentum Space

Saturation of Excited States Saturation



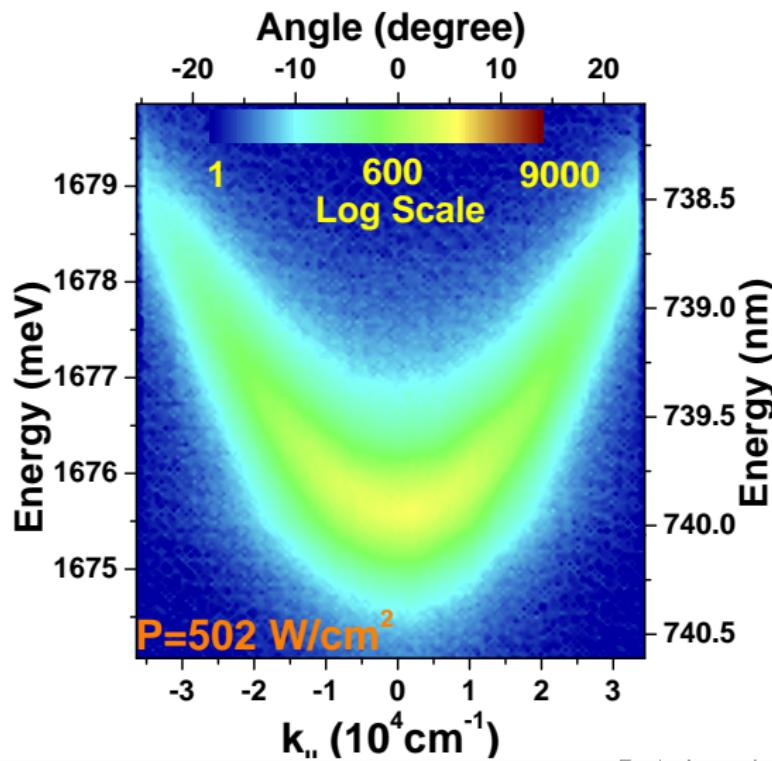
Polariton Condensation in Momentum Space

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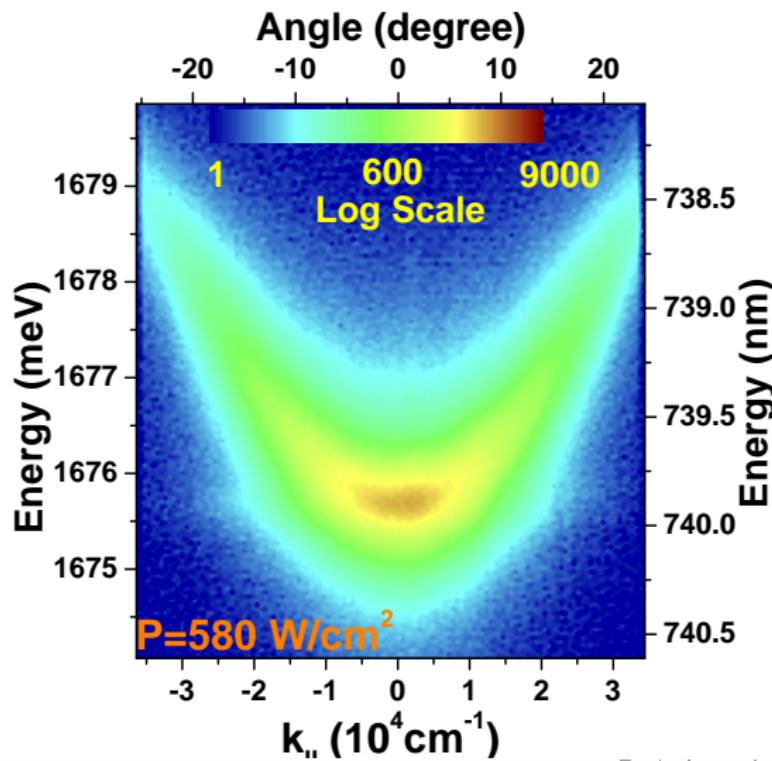
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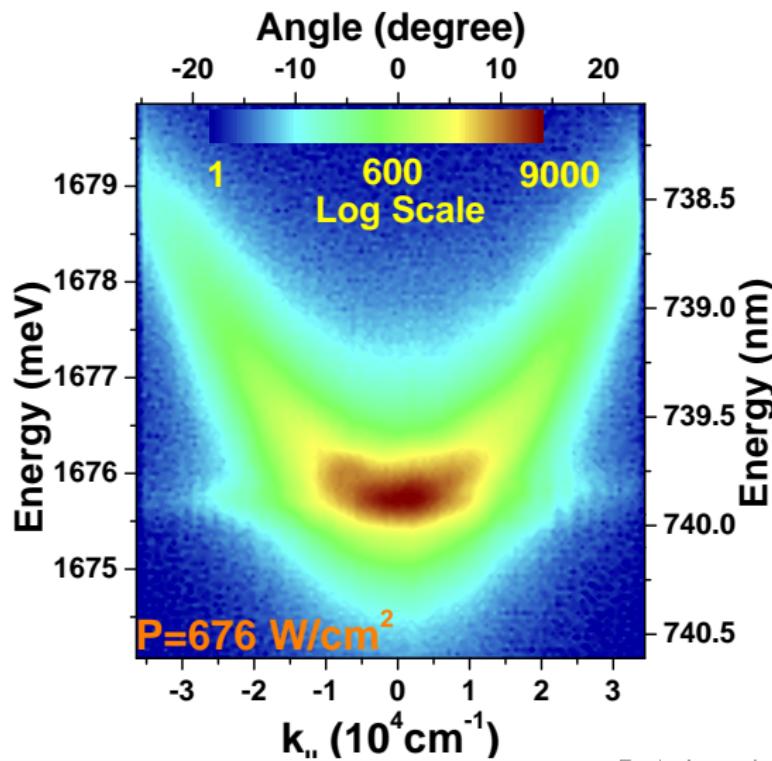
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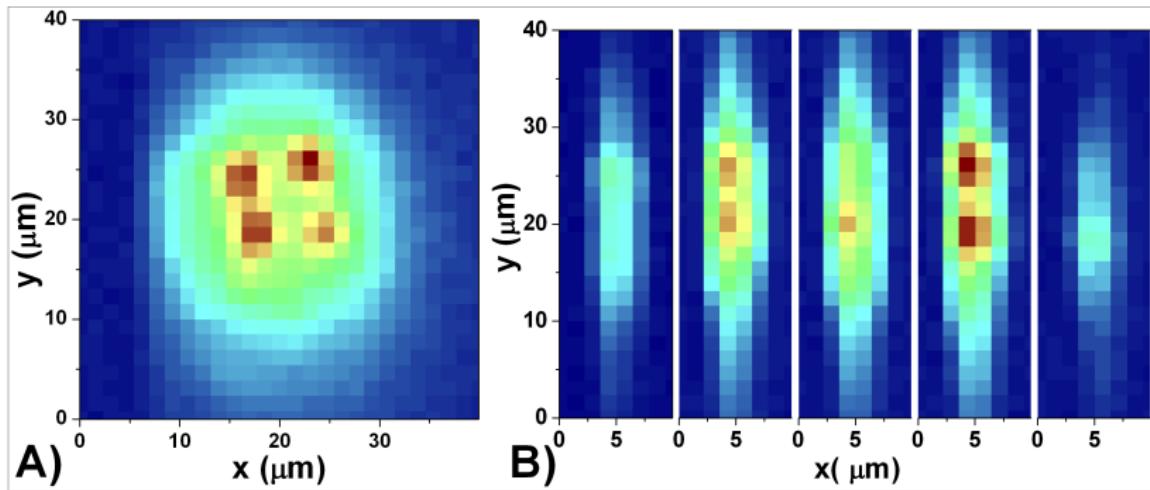
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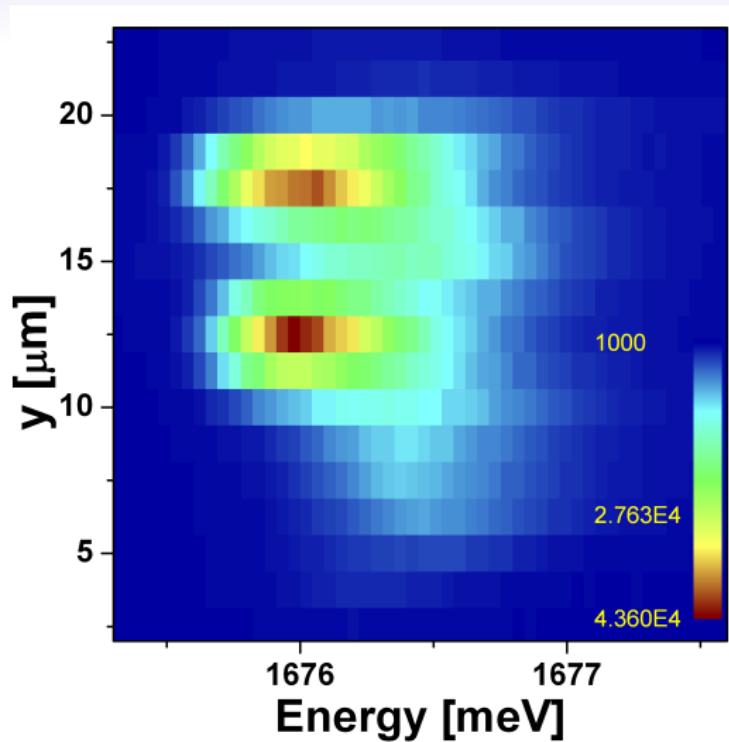
Plan

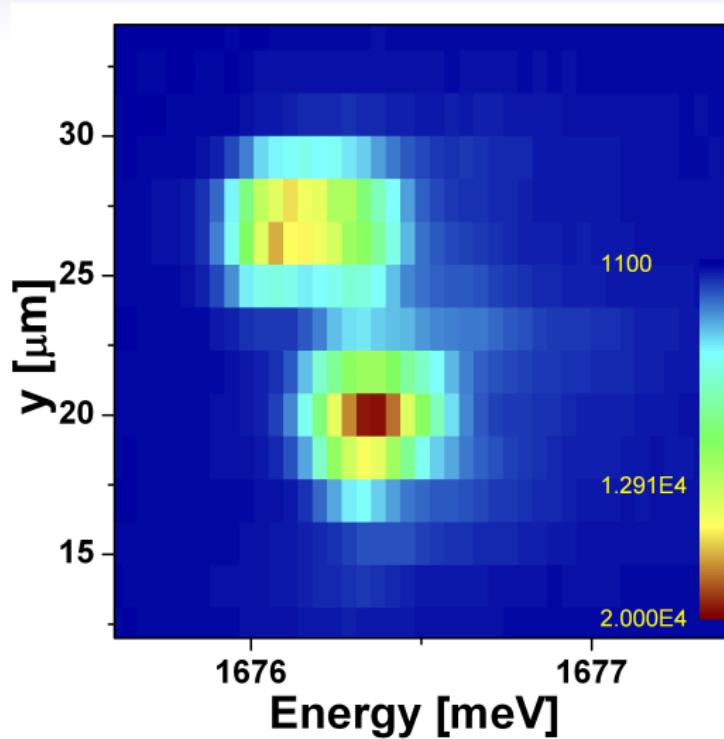
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(x, λ) -measurements

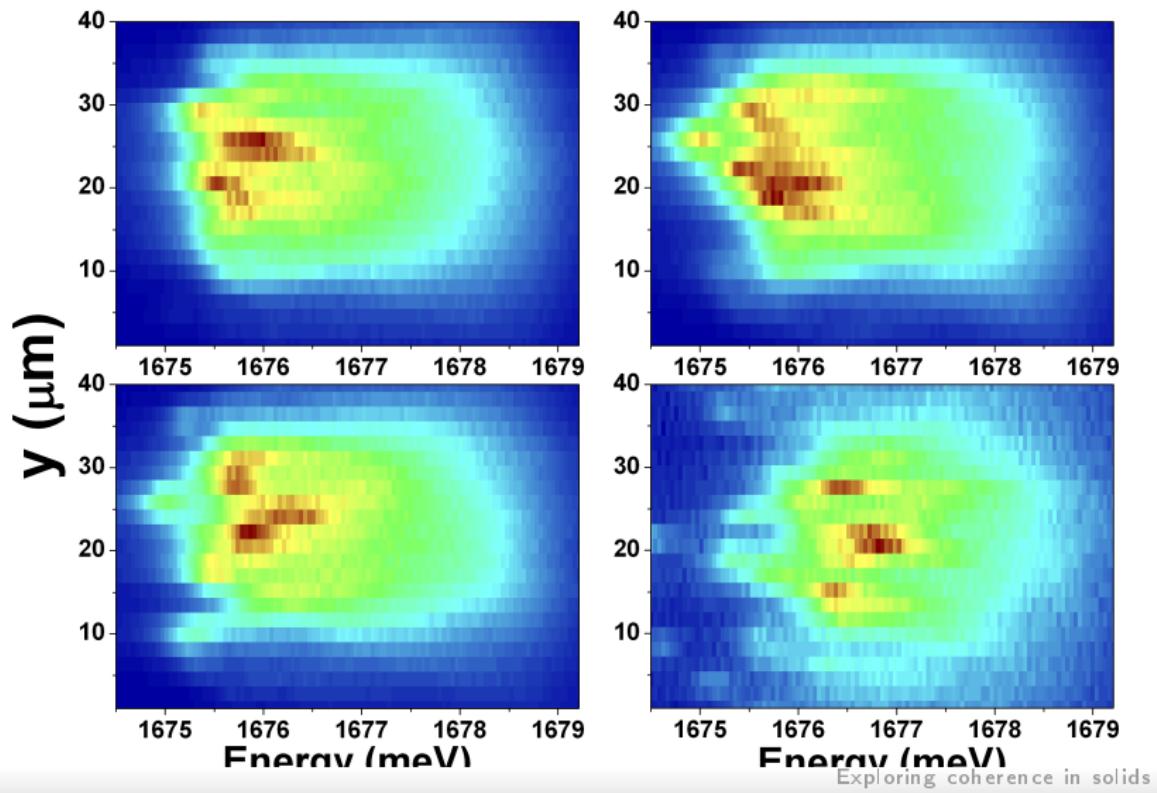
(x, λ) -measurements



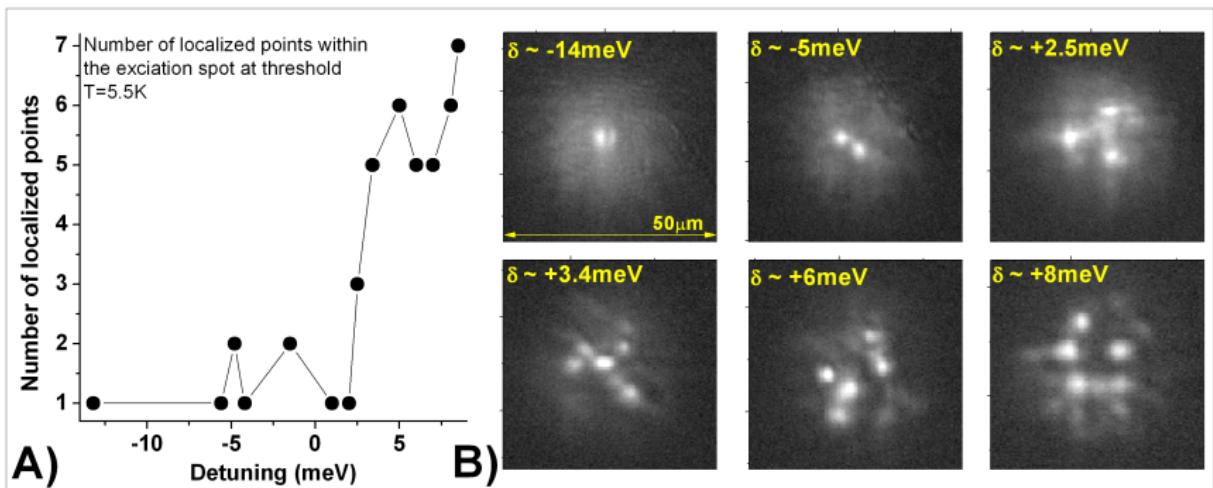
(x, λ) -measurements

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Desorder visible below threshold



Detuning dependence

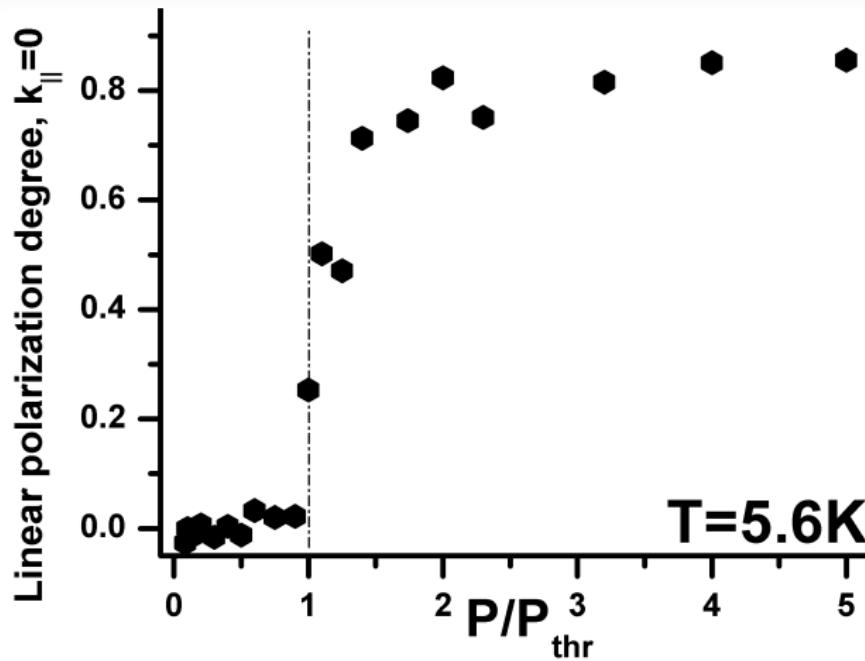


Plan

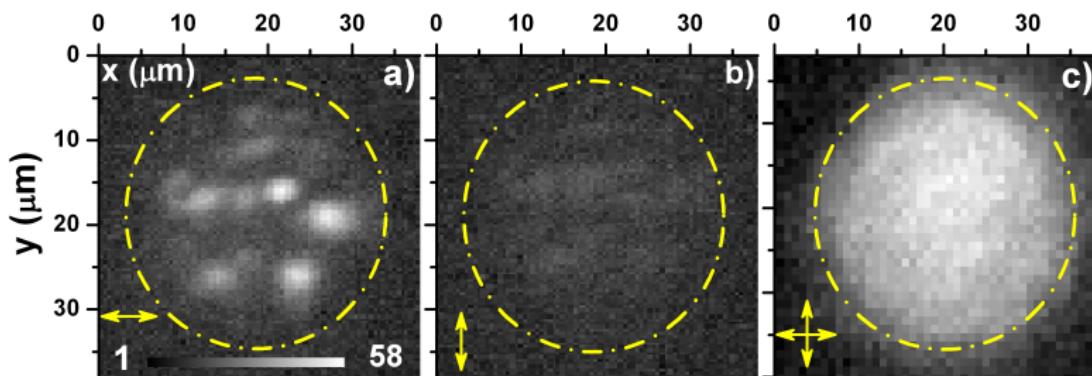
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Linear Polarization Build-up

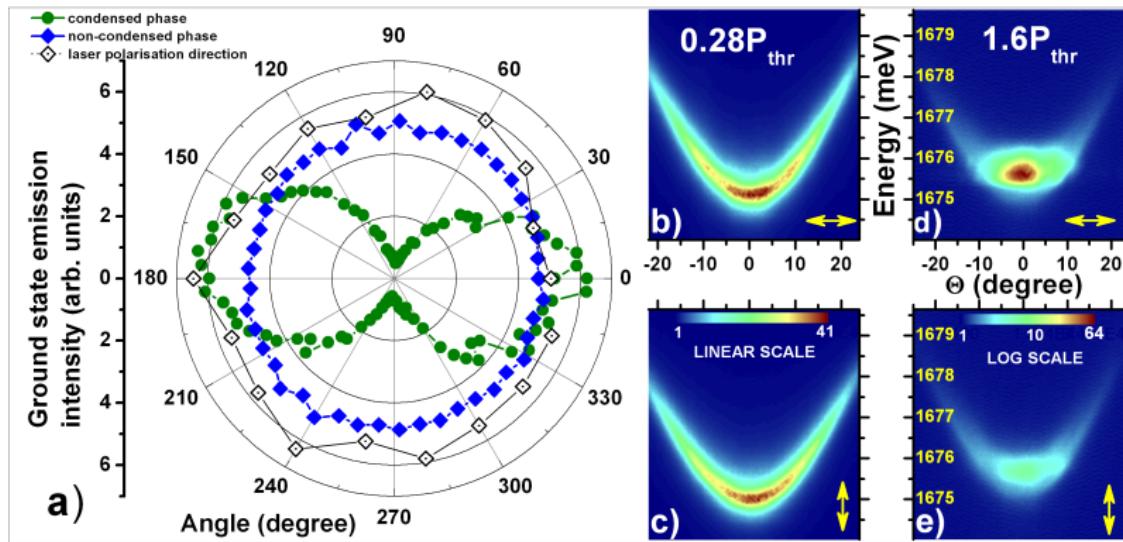


Emission Structure vs. Power vs. Polarization

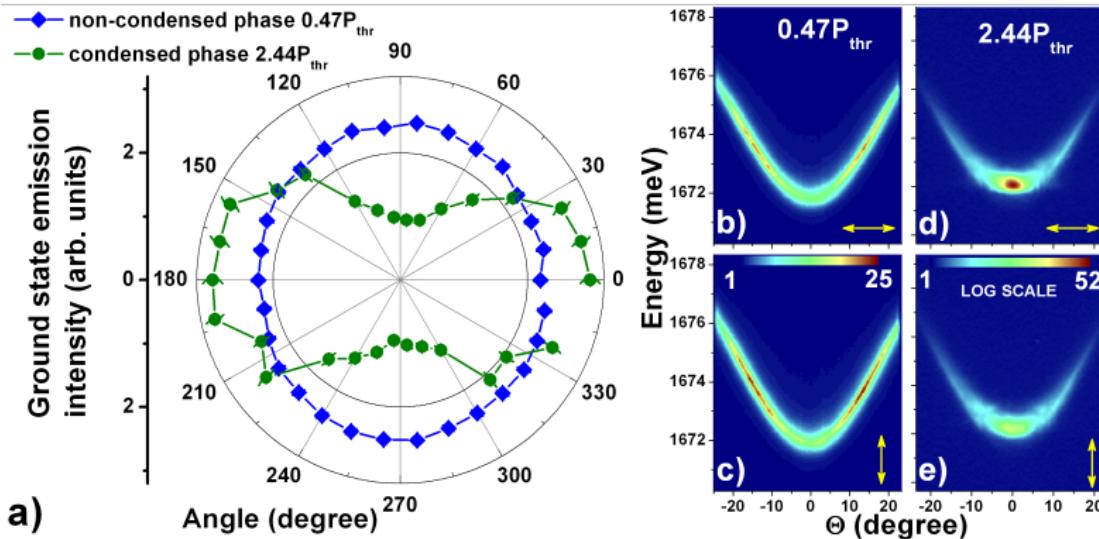


Linear Polarization Probing

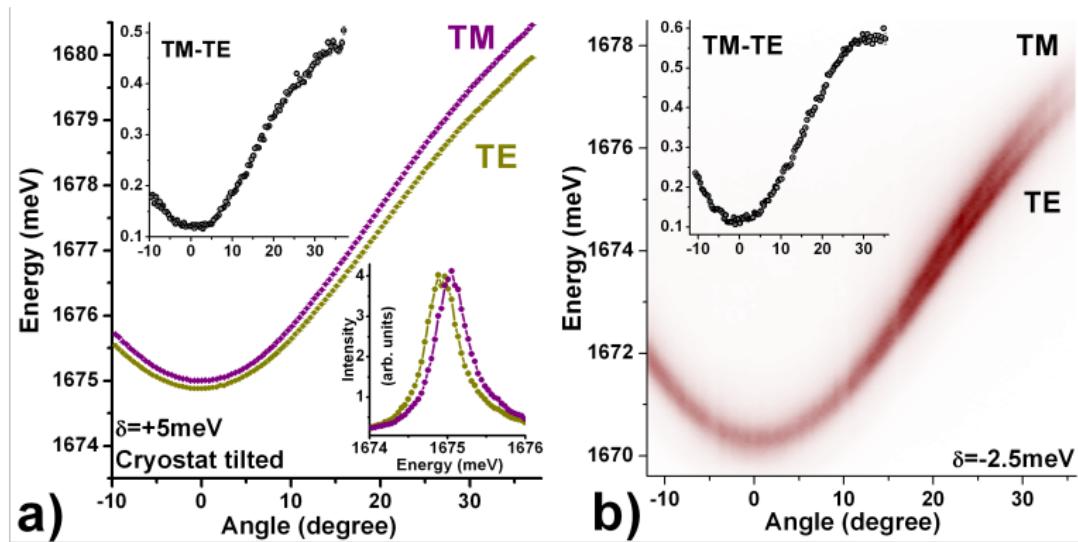
Spontaneous Build-up of Linear Polarization



Spontaneous Build-up of Linear Polarization



Origin of the Linear Polarization Ground State Splitting

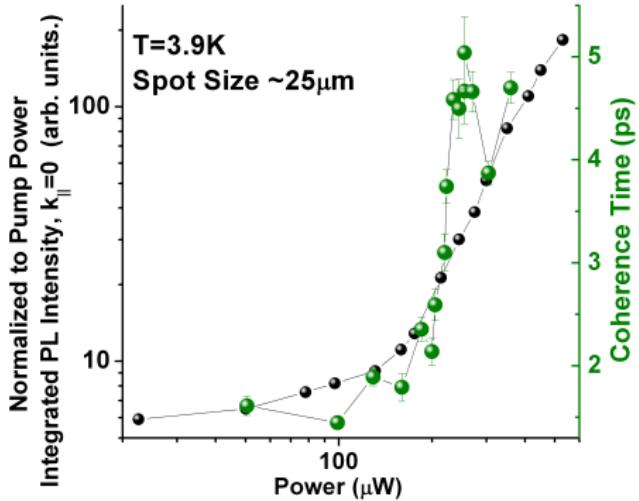
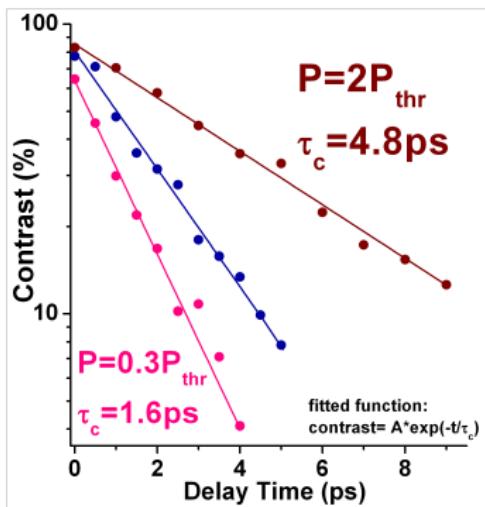


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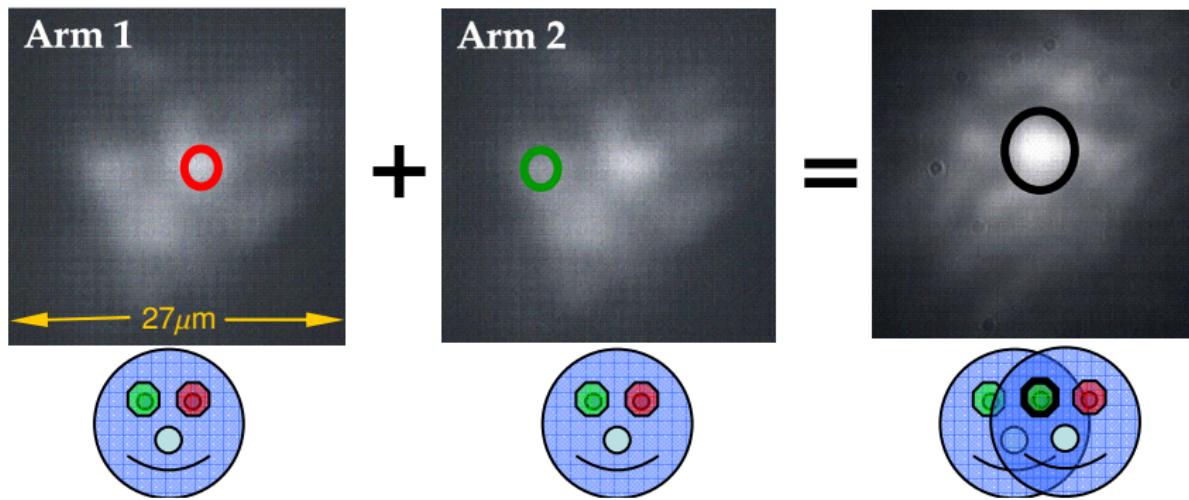
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Temporal Coherence



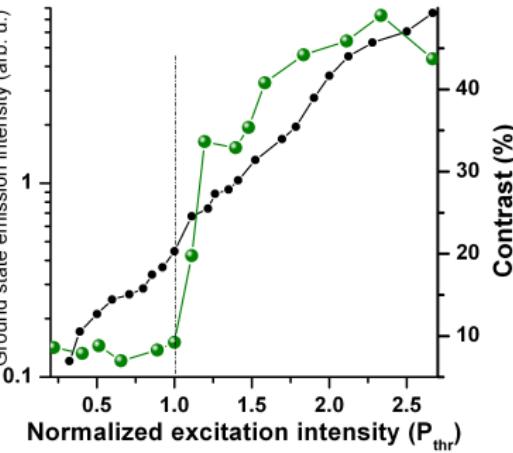
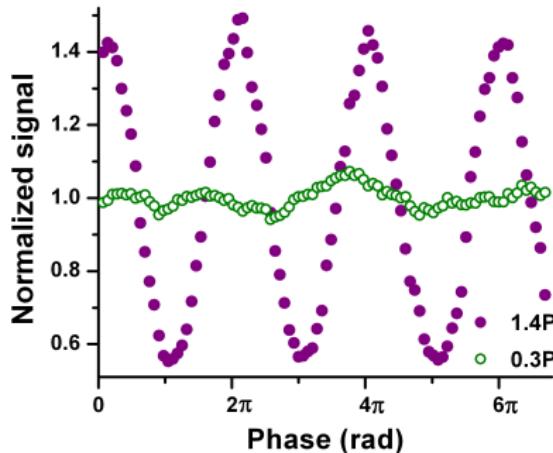
Spatial Coherence

Interference Between Different Spots



Spatial Coherence

Interference Between Different Spots



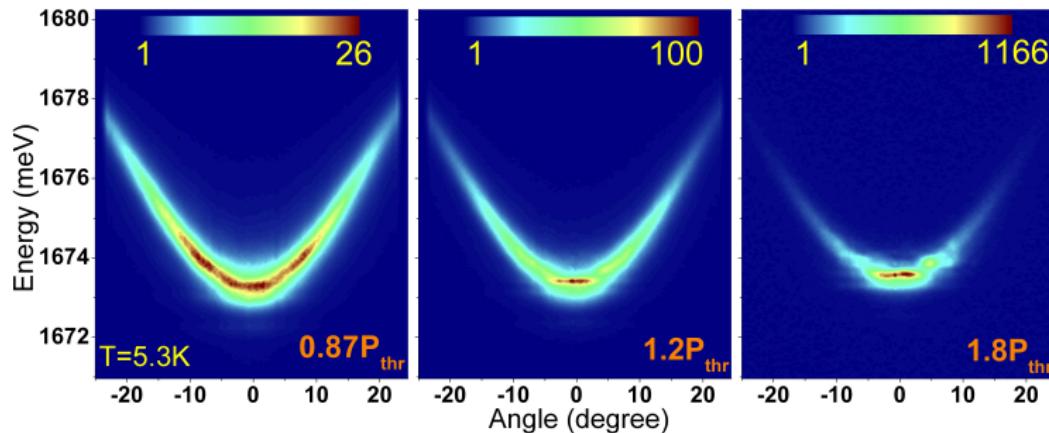
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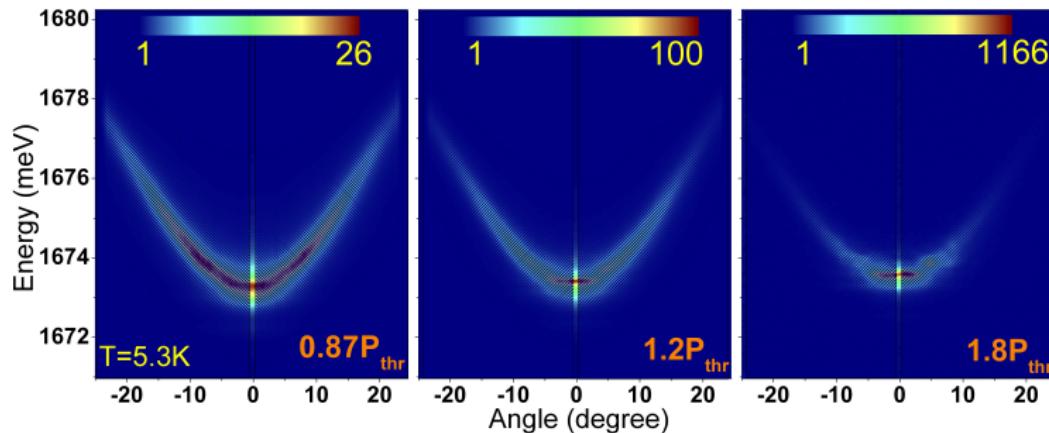
Nonequilibrium Polariton Condensation

Non-resonant, Pulsed Excitation



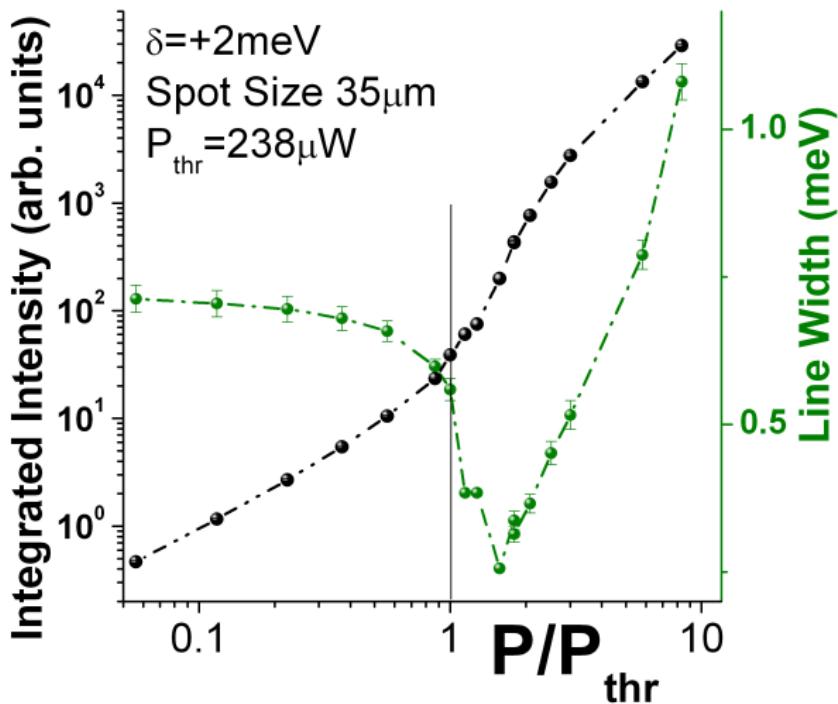
Nonequilibrium Polariton Condensation

Non-resonant, Pulsed Excitation



Transition Characteristics

Threshold, Line Width



What Can We Learn From the Photon Statistics

2nd Order Coherence Function

$$g_2(\tau) \propto \langle I(t) \cdot I(t + \tau) \rangle_t$$

Source

- Coherent
- Thermal

Statistics

- Poisson
- B-E

$g_2(\tau = 0)$

- 1
- 2

$g_2(\tau = \infty)$

- 1
- 2

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- Thermal

Statistics

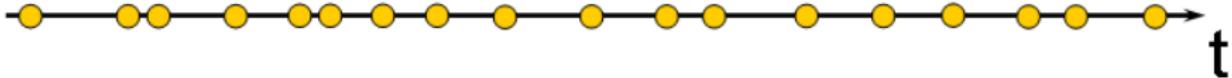
- Poisson
- B-E

$g_2(\tau = 0)$

- 1
- 2

$g_2(\tau = \infty)$

- 1
- 1



What Can We Learn From the Photon Statistics

2nd Order Coherence Function

$$g_2(\tau) \propto \langle I(t) \cdot I(t + \tau) \rangle_t$$

Source

- Coherent
- Thermal

Statistics

- Poisson
- B-E

$g_2(\tau = 0)$

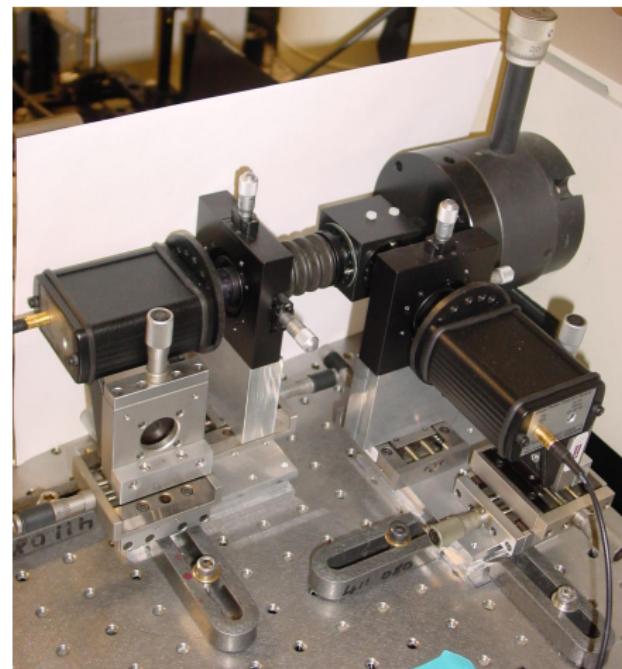
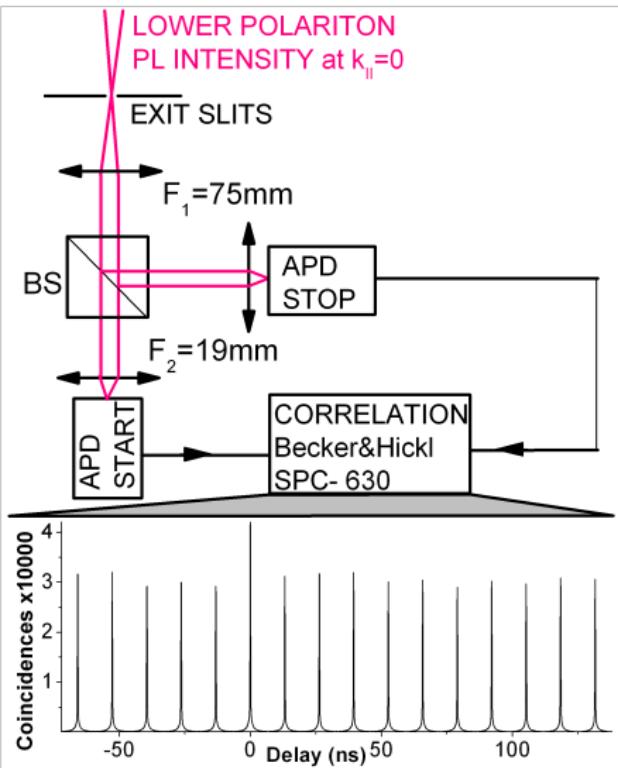
- 1
- 2

$g_2(\tau = \infty)$

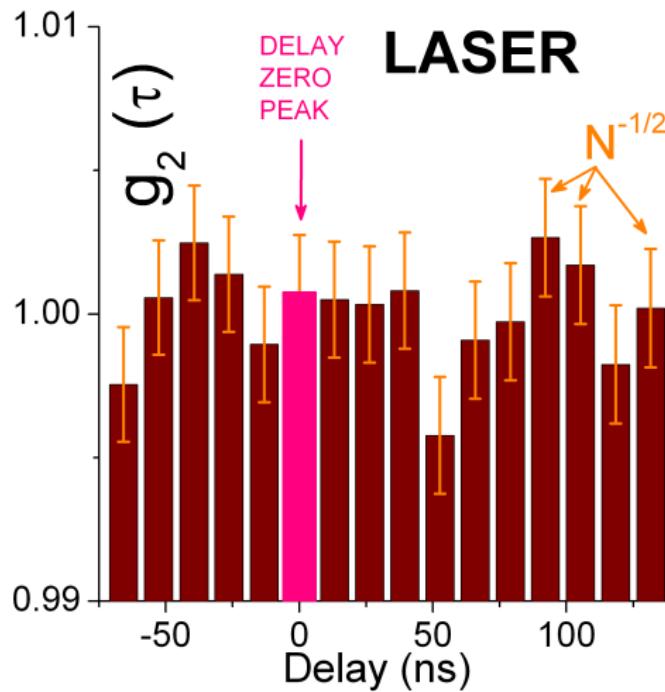
- 1
- 1



Intensity Correlation Experiment



Laser Check



What Do We Expect ?

Characteristic Timescales

P/P_{thr}	Decay Time [ps]	Coherence Time [ps]	Expected Bunching
• < 1	• 100-200	• 1-2	• 1%
• $(1, 2)$	• 7	• 5-7	• No Bunching
• > 2	• 3-7	• 1	• 10 – 30%

What Do We Expect ?

Characteristic Timescales

P/P_{thr}	Decay Time [ps]	Coherence Time [ps]	Expected Bunching
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What Do We Expect ?

Characteristic Timescales

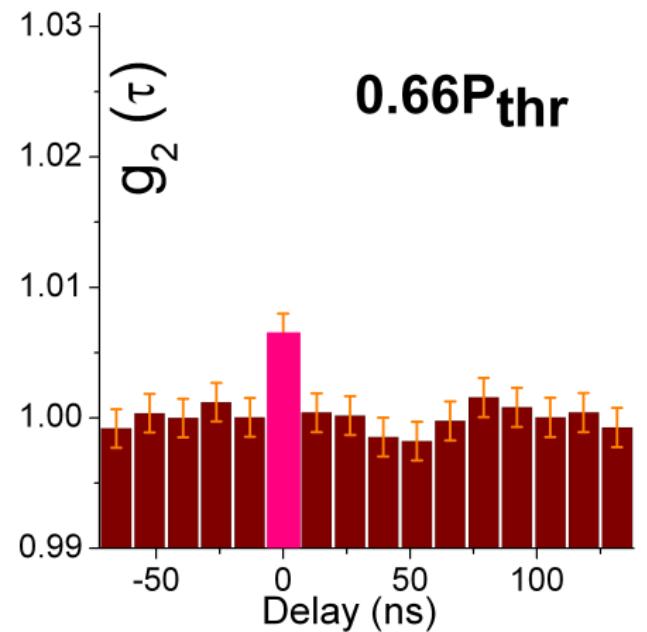
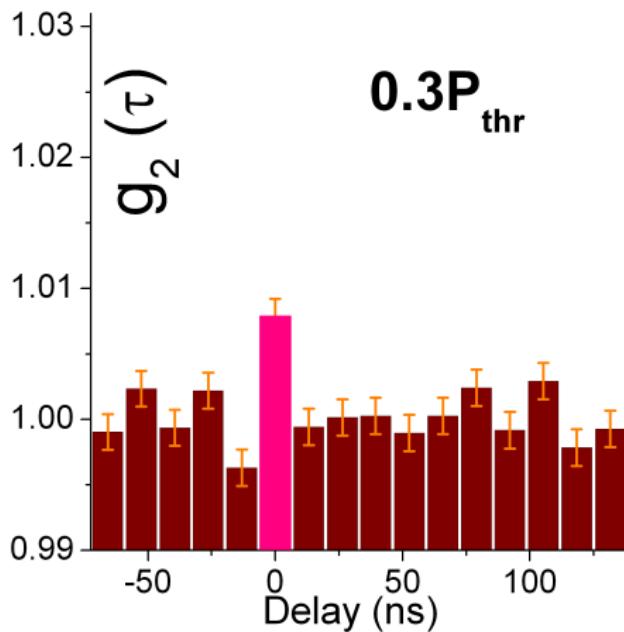
P/P_{thr}	Decay Time [ps]	Coherence Time [ps]	Expected Bunching
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What Do We Expect ?

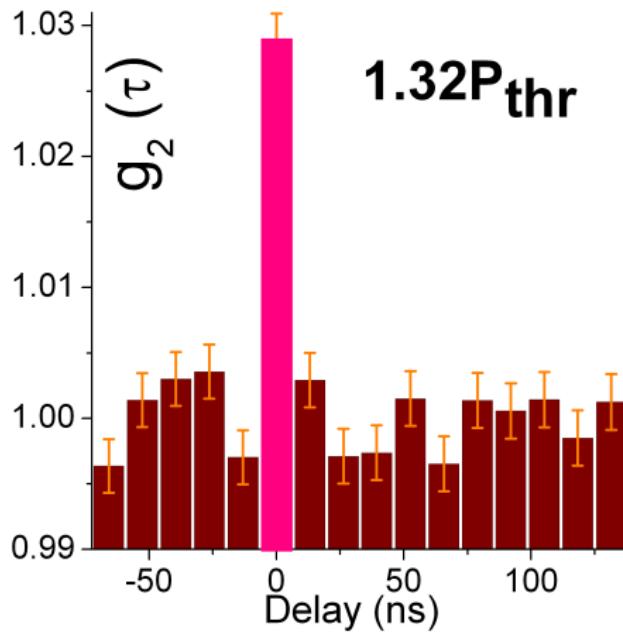
Characteristic Timescales

P/P_{thr}	Decay Time [ps]	Coherence Time [ps]	Expected Bunching
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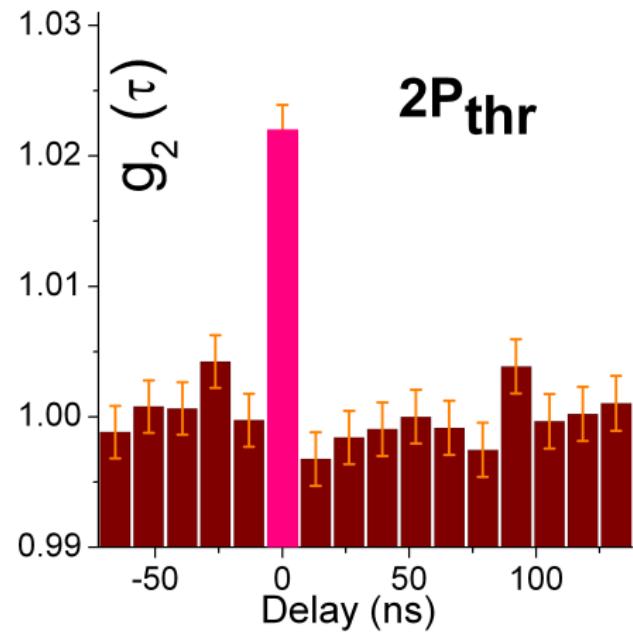
Result - $g_2(\tau = 0)$ Values Below Threshold



Result - $g_2(\tau = 0)$ Values At Threshold



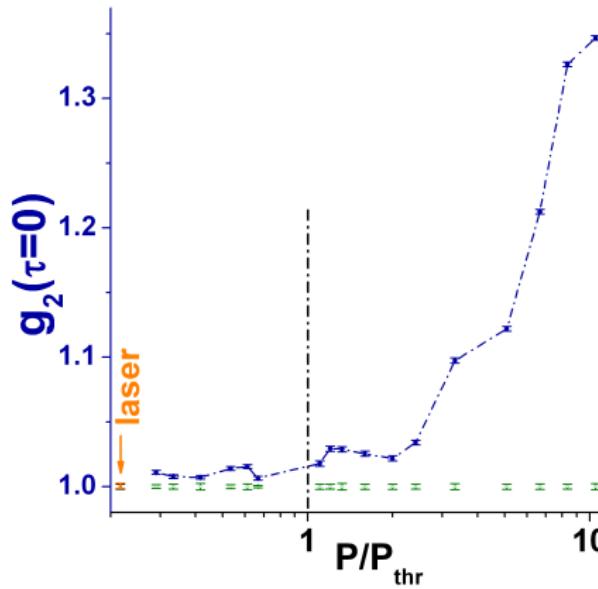
1.32P_{thr}



2P_{thr}

Summary

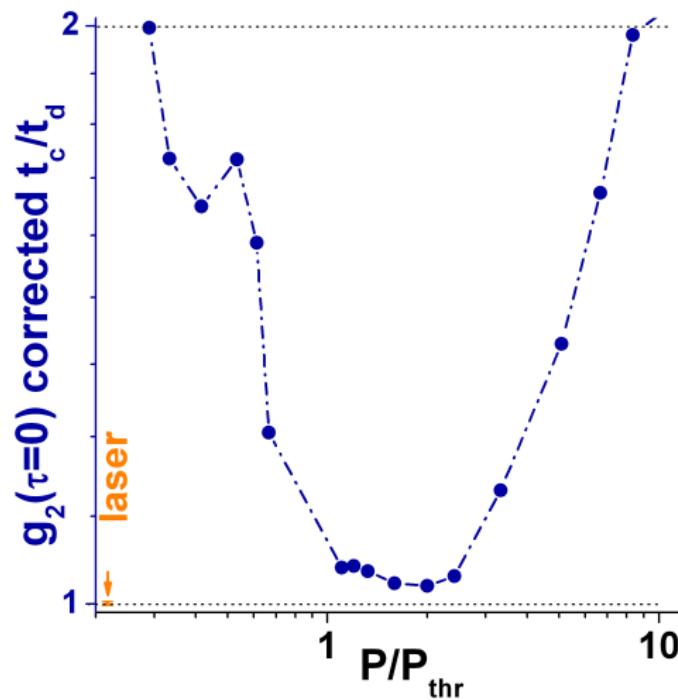
$\delta \approx +2\text{meV}$, $T=5.4\text{K}$, Counting Rate $\sim 3 \cdot 10^5 \frac{\text{c}}{\text{s}}$, Pulsed, Non-Resonant



Relaxation

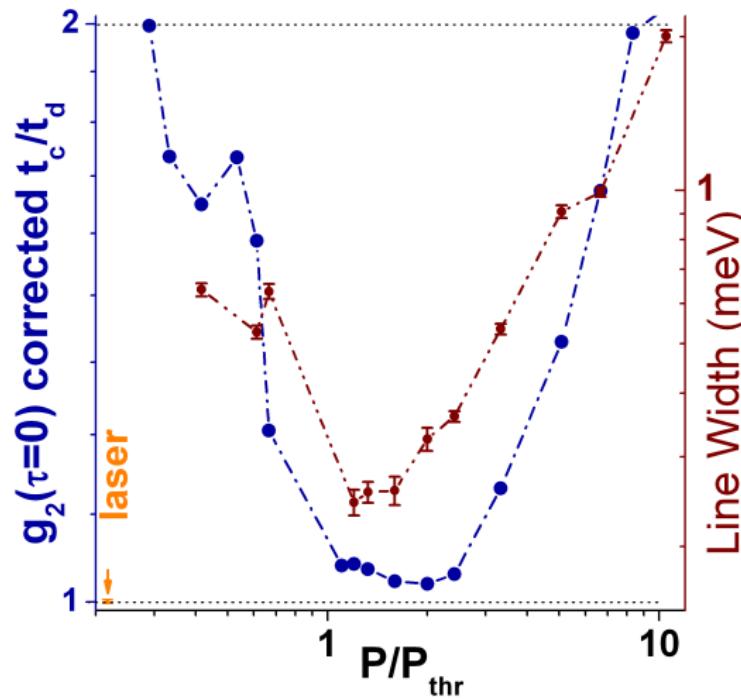
Summary

$g_2(\tau = 0)$ Corrected



Summary

$g_2(\tau = 0)$ Corrected



Interpretation

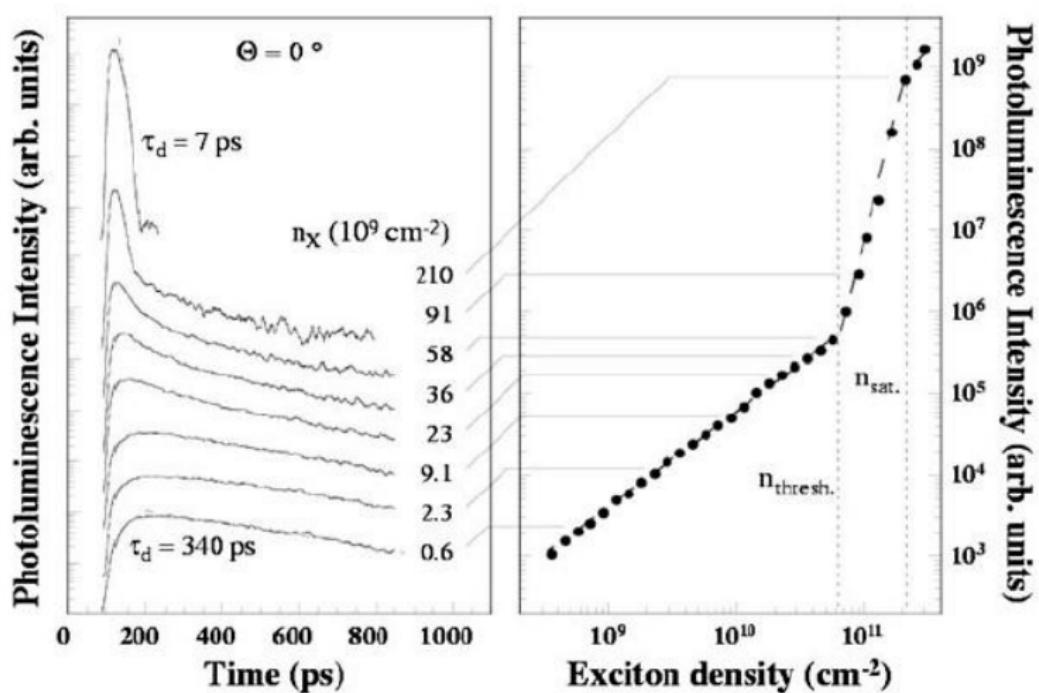
$P/P_{thr} < 1$ - Thermal Source

$1 < P/P_{thr} < 2$ - Coherent Source

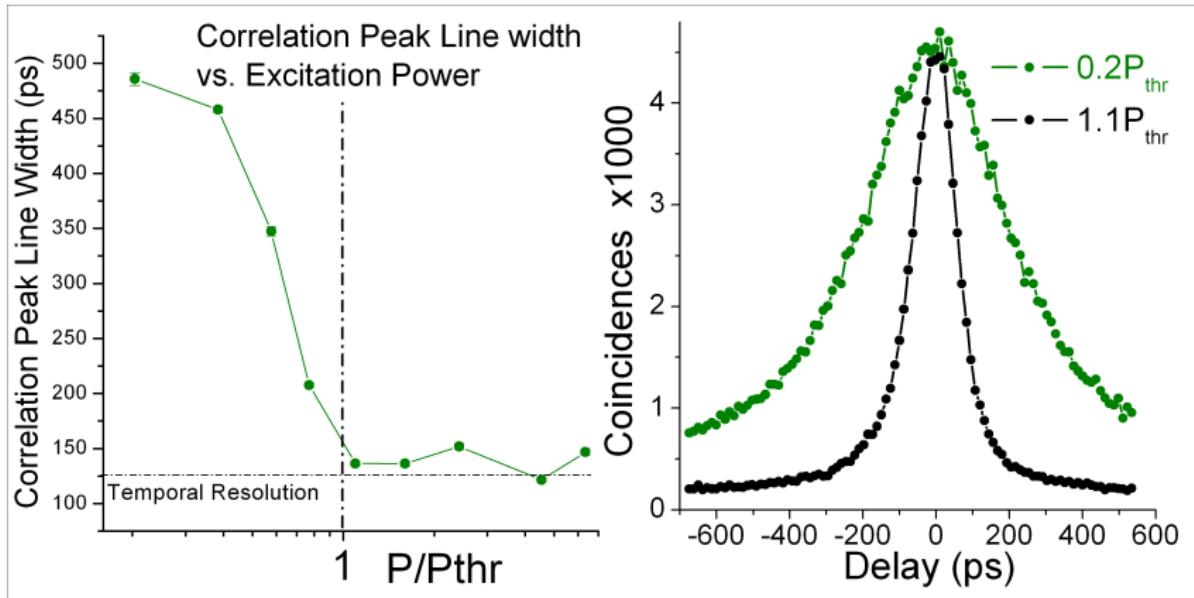
$P/P_{thr} > 2$ - Decoherence - Interaction Within the Condensate

Speed Up of the Relaxation

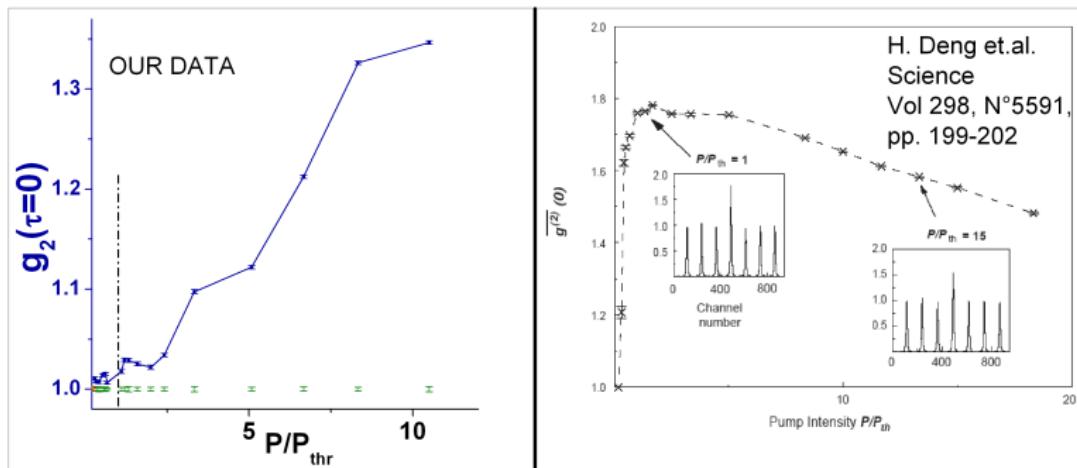
M. Müller Thesis - Grenoble 2000



Speed Up of the Relaxation

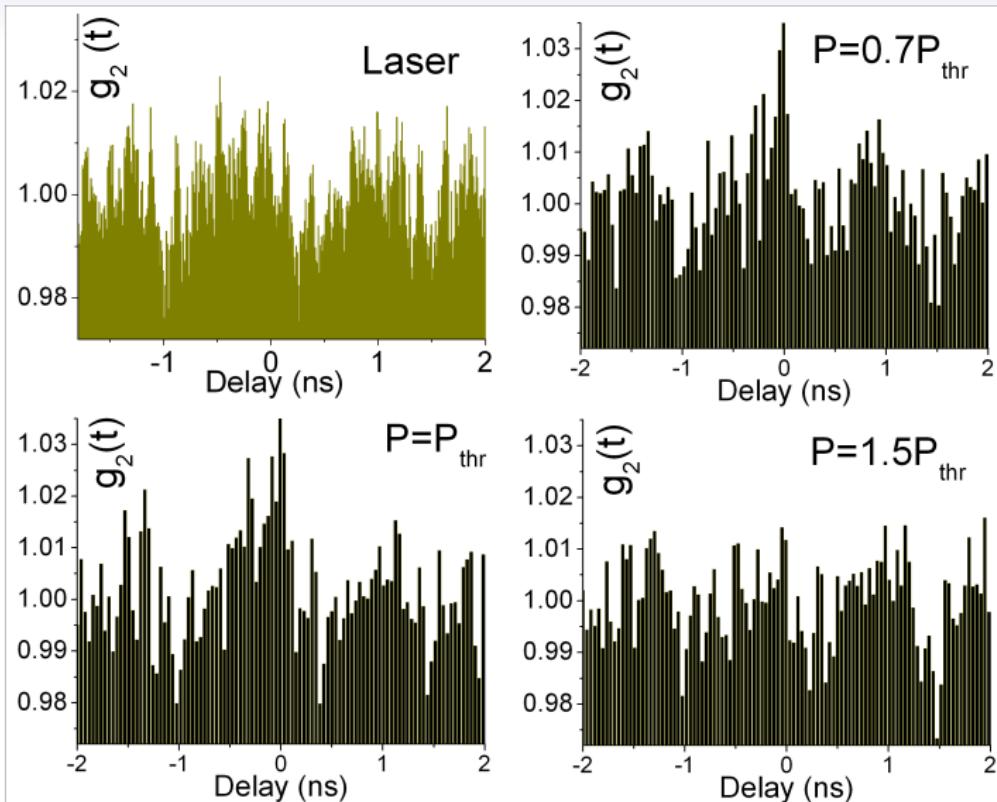


How About the Other Results ?



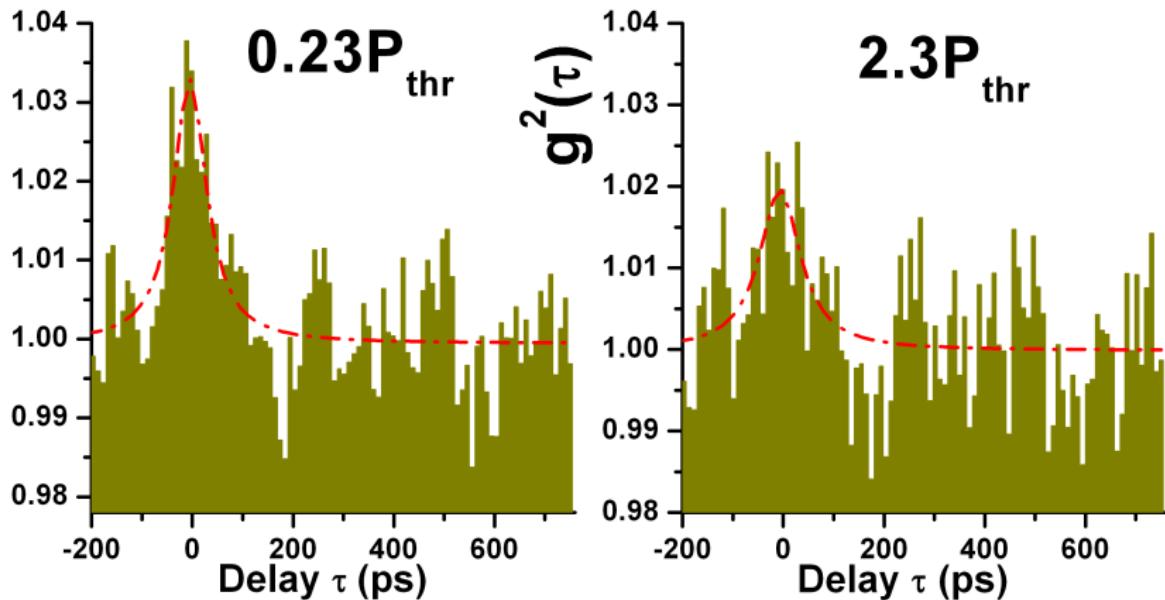
CW Measurements

...Much More Difficult !!!



CW Measurements

Reduction of bunching \Leftrightarrow Build up of the 2nd Order Coherence



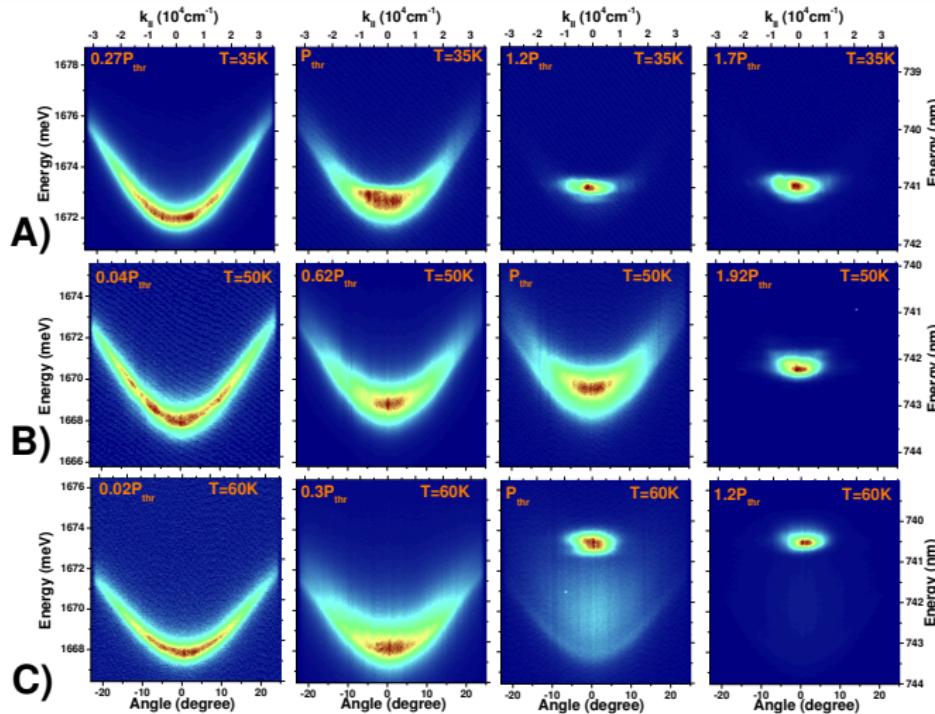
Plan

5 Appendix

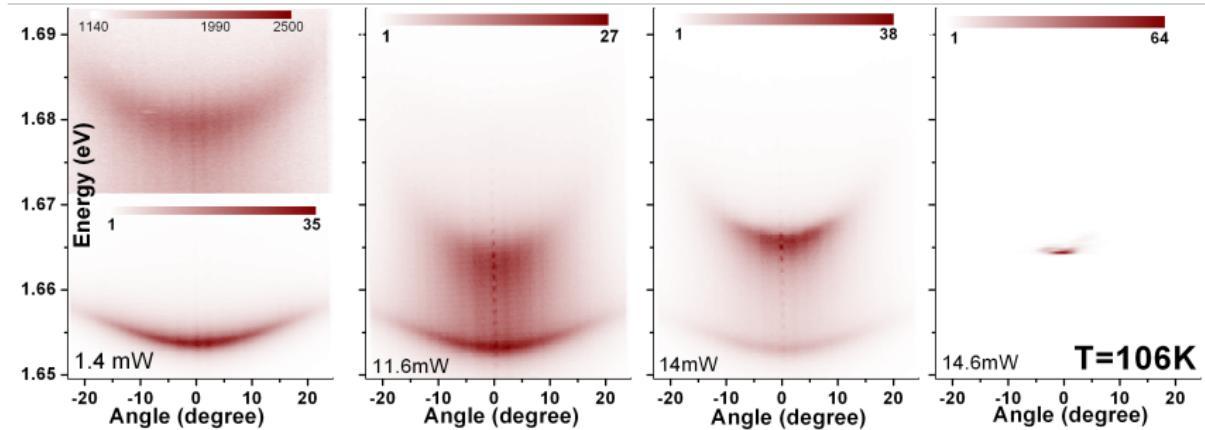
- Experimental Setup
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- Linear Polarization Build-up
- Coherence More
- g^2
- **High Temperature Limit**
- Polaritons at room temperature
- Polariton Lasing vs. Photon Lasing (VCSEL)
- Stimulation on the Ring of States

Polariton Condensation in CdTe Microcavities

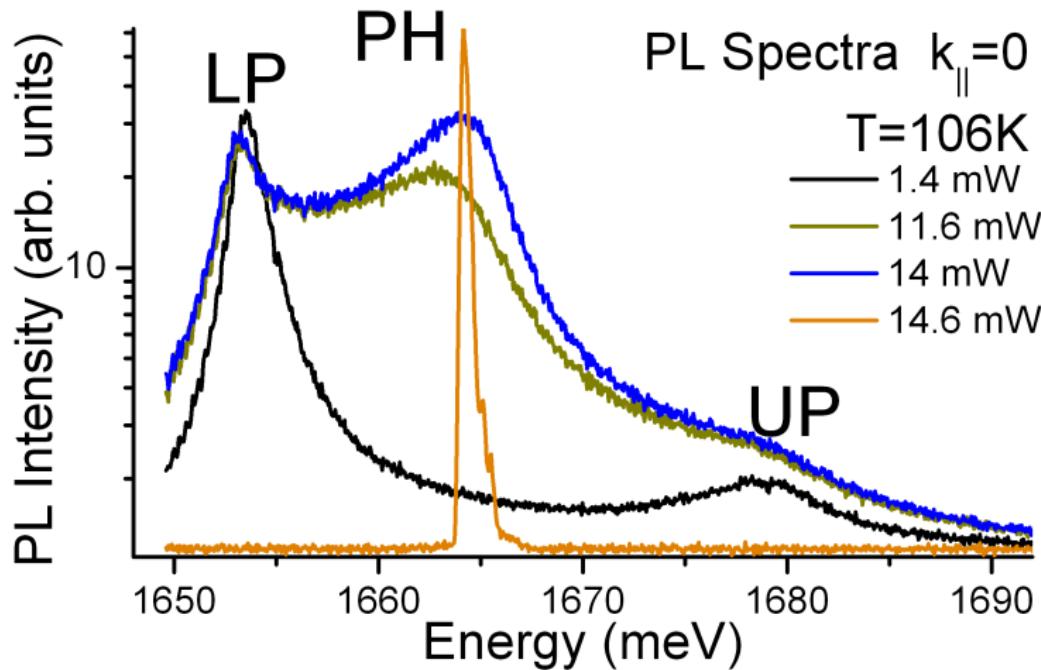
High Temperature Limit - $T_{\text{cryo}} = 50K$



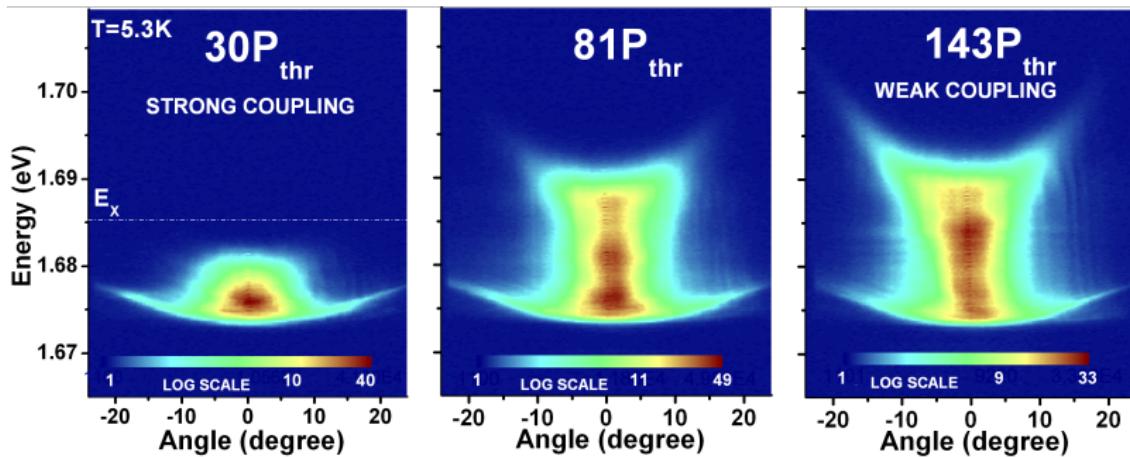
Transition Towards Weak Coupling Regime

$$T_{cryo} = 106K$$


Transition Towards Weak Coupling Regime

$$T_{cryo} = 106K$$


Transition Towards Weak Coupling Regime

$$T_{cryo} = 5K$$


Polariton Lasing versus Photon Lasing

High finesse microcavity + QW Exciton \Rightarrow Strong Coupling

High excitation

Large E_{Bx}

Small E_{Bx}

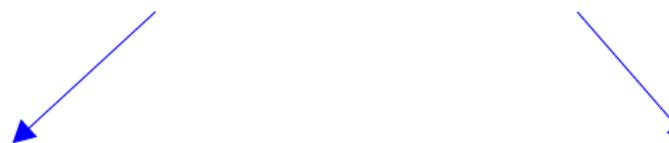
Polariton Lasing versus Photon Lasing

High finesse microcavity + QW Exciton \Rightarrow Strong Coupling

High excitation

Large E_{BX}

Small E_{BX}



X - Strong Coupling

BEC, Polariton Lasing

(e,h) - Weak Coupling

VCSEL

Polariton Lasing versus Photon Lasing

High finesse microcavity + QW Exciton \Rightarrow Strong Coupling

High excitation

Large E_{BX}

Small E_{BX}

X - Strong Coupling

BEC, Polariton Lasing

(e,h) - Weak Coupling

VCSEL

Higher Excitation and/or high T

Exciton Saturation

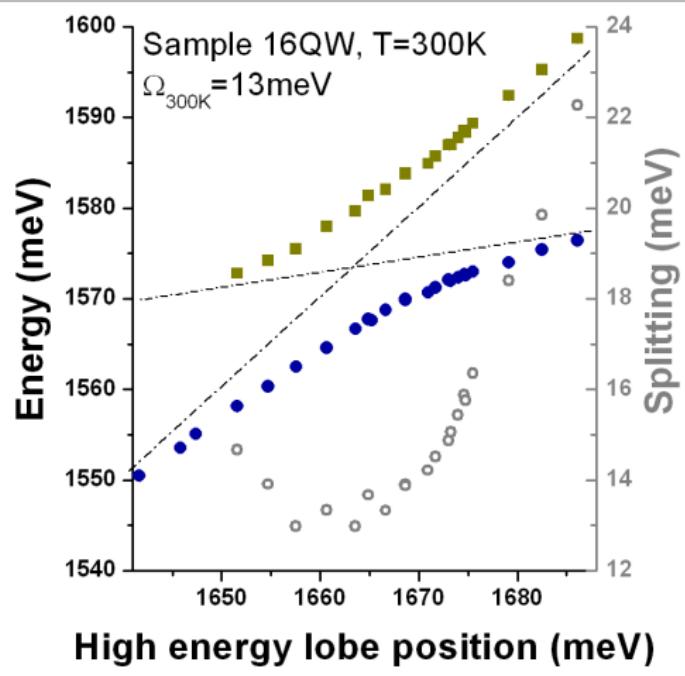
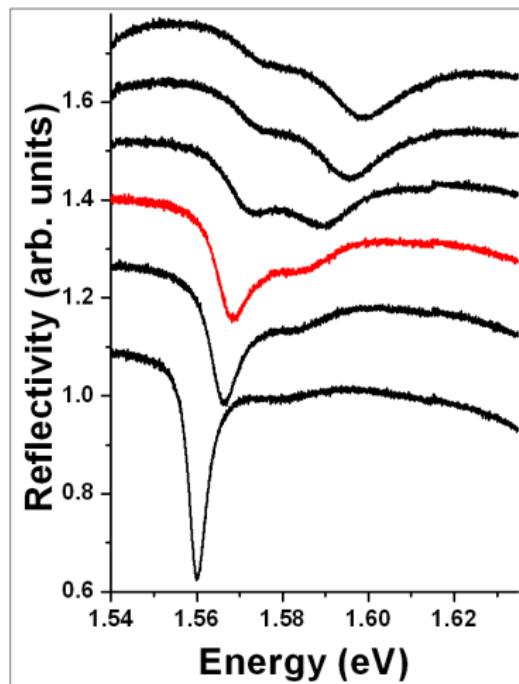
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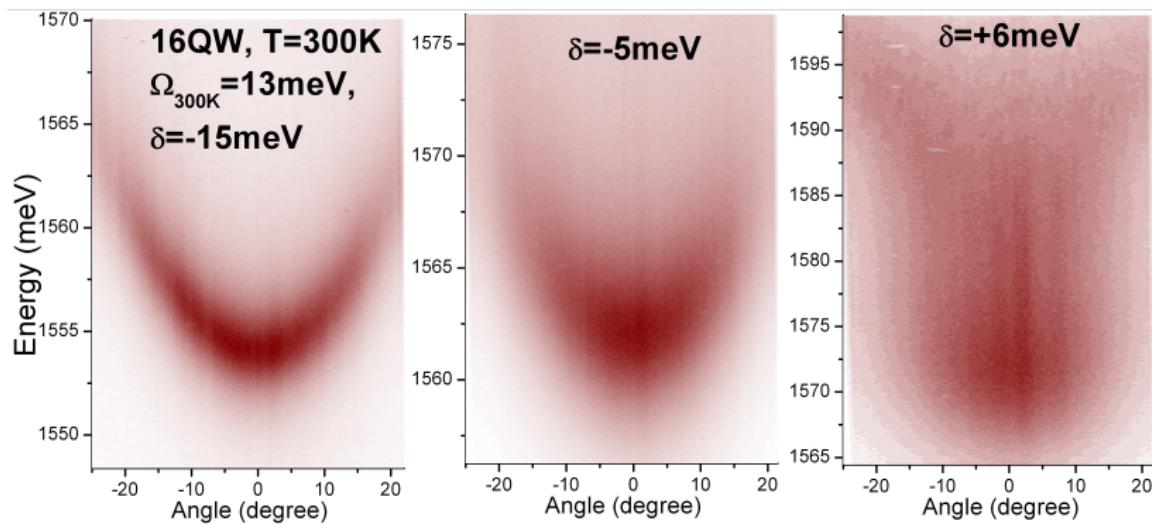
Strong coupling at room temperature

Nonlinearities not observed



Strong coupling at room temperature

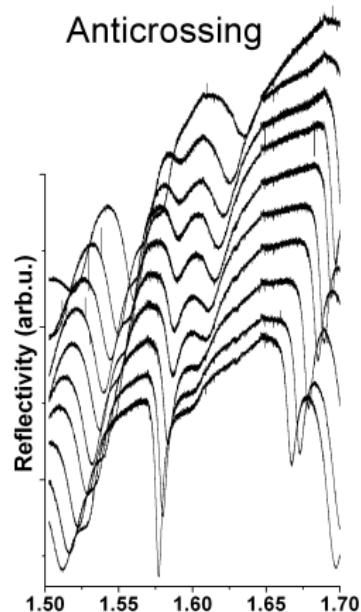
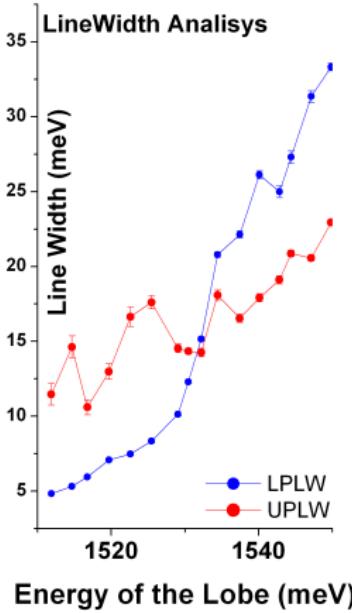
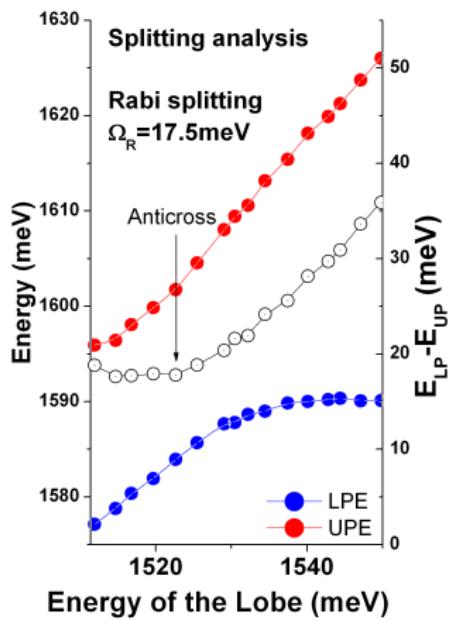
Nonlinearities not observed



Strong coupling at room temperature

Nonlinearities not observed

T=300K, sample 24QW

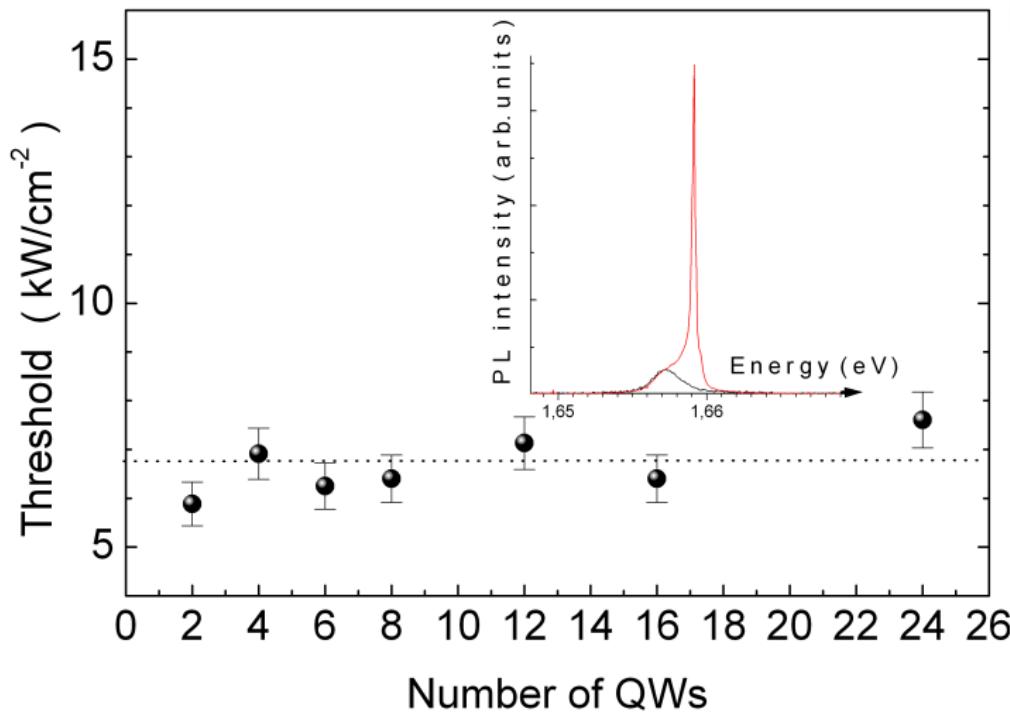


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Threshold vs. Number of QWs



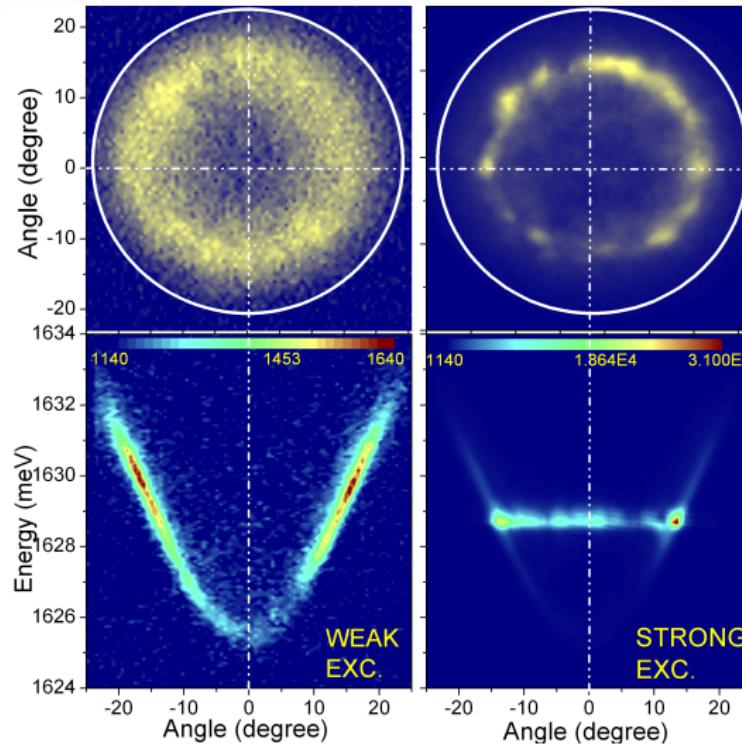
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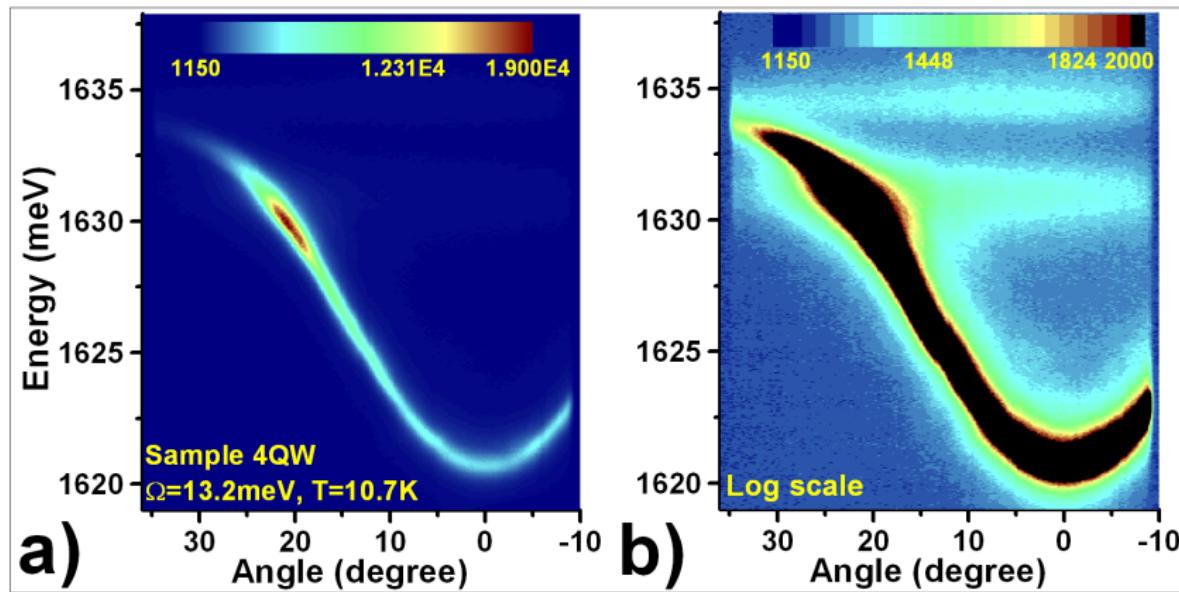
Stimulation on the Ring of Excited States

Spot Size $\sim 3\mu\text{m}$ - Strong Localization



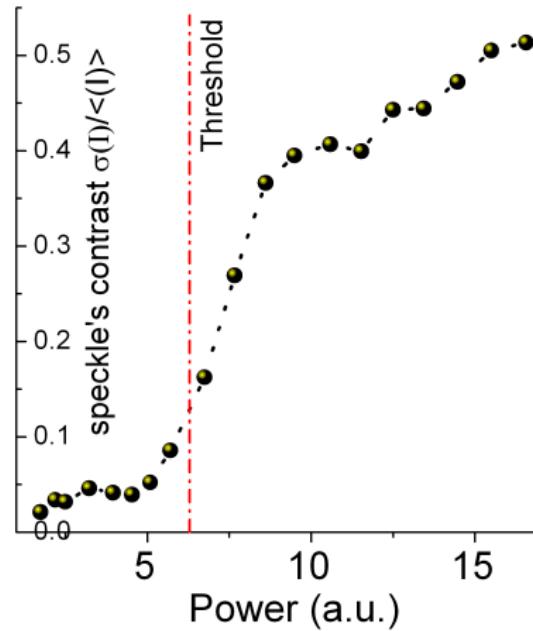
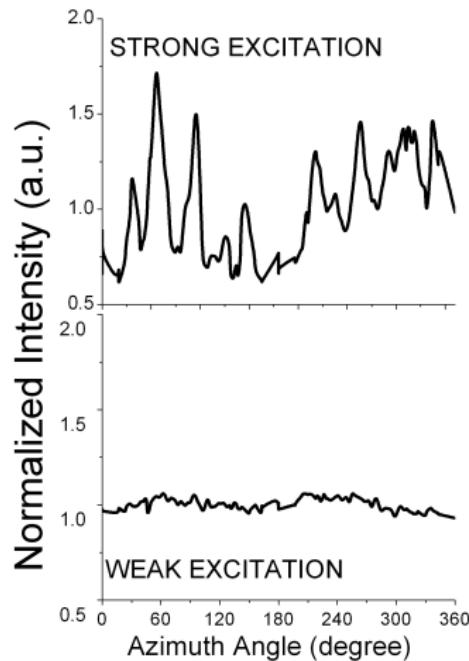
Stimulation on the Ring of Excited States

Non-dispersive Bar-Geometrical Artefact



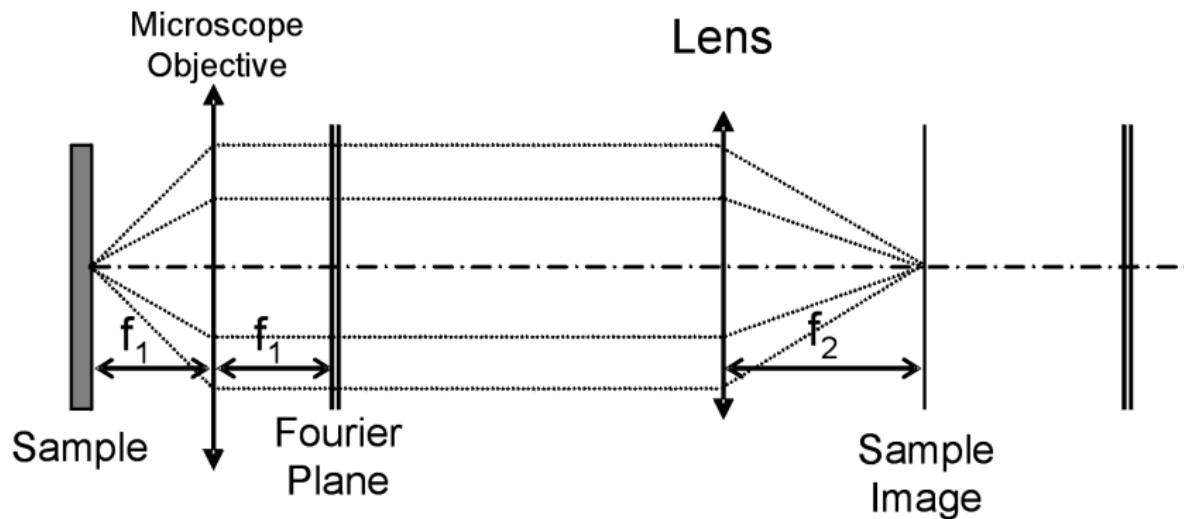
Stimulation on the Ring of Excited States

Speckled Emission Above Threshold



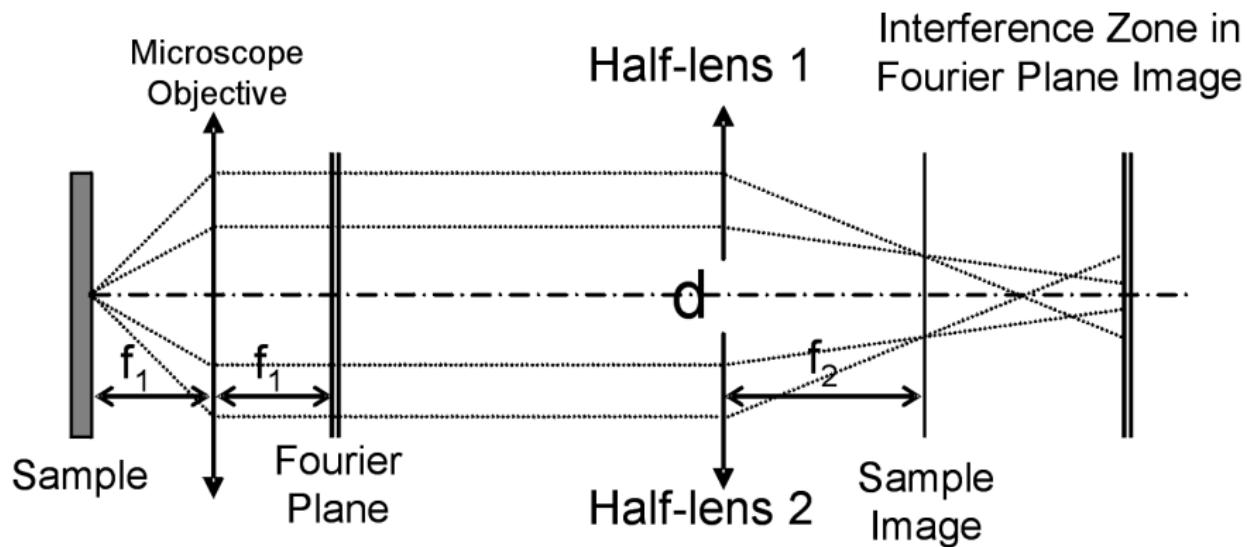
Stimulation on the Ring of Excited States

Transverse Field Interferometer - Principle



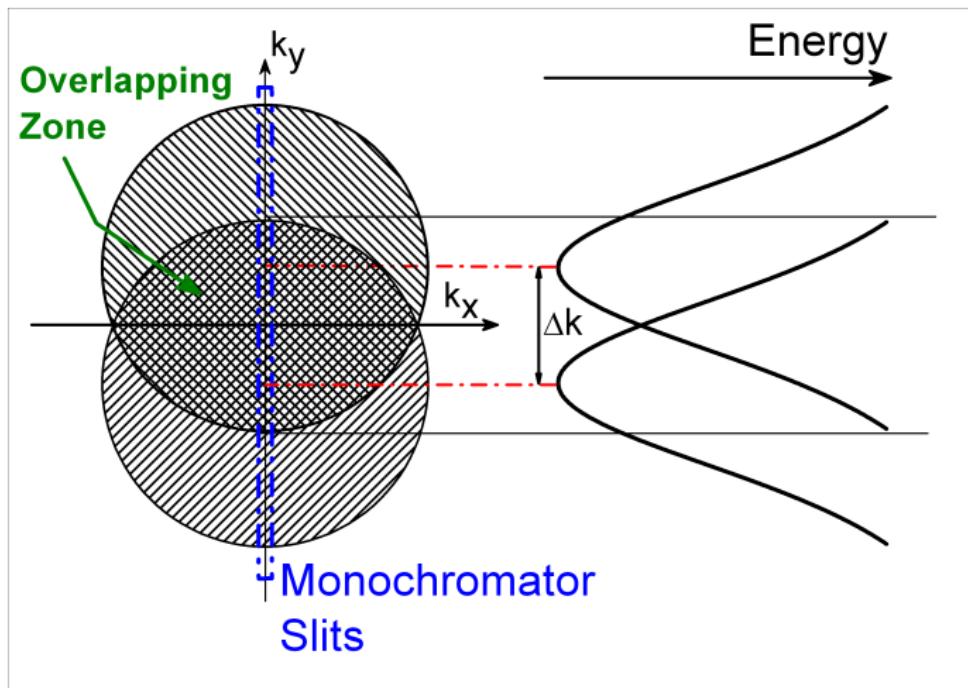
Stimulation on the Ring of Excited States

Transverse Field Interferometer - Principle



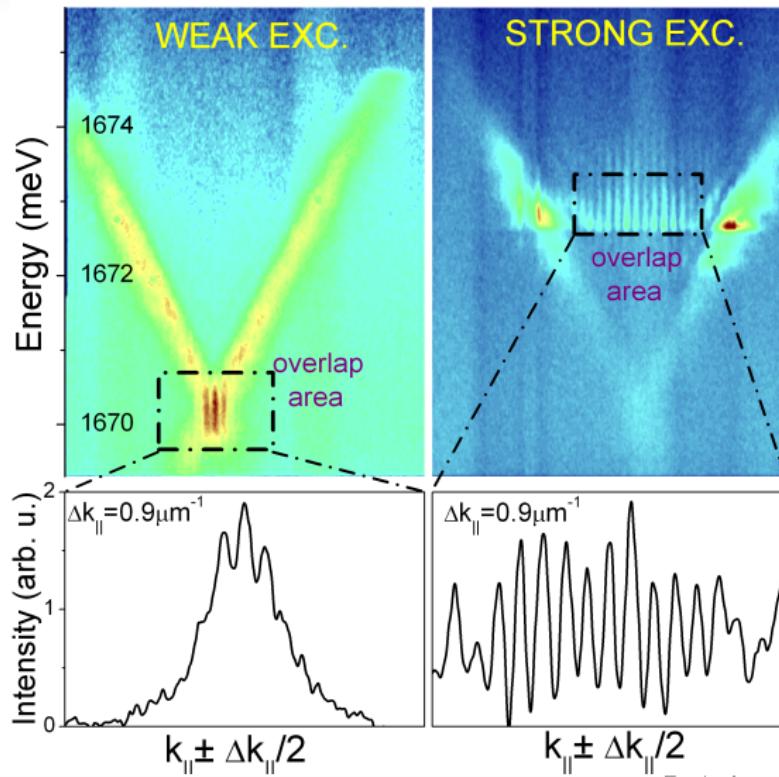
Stimulation on the Ring of Excited States

Billet Interferometer - Zone of the Overlap



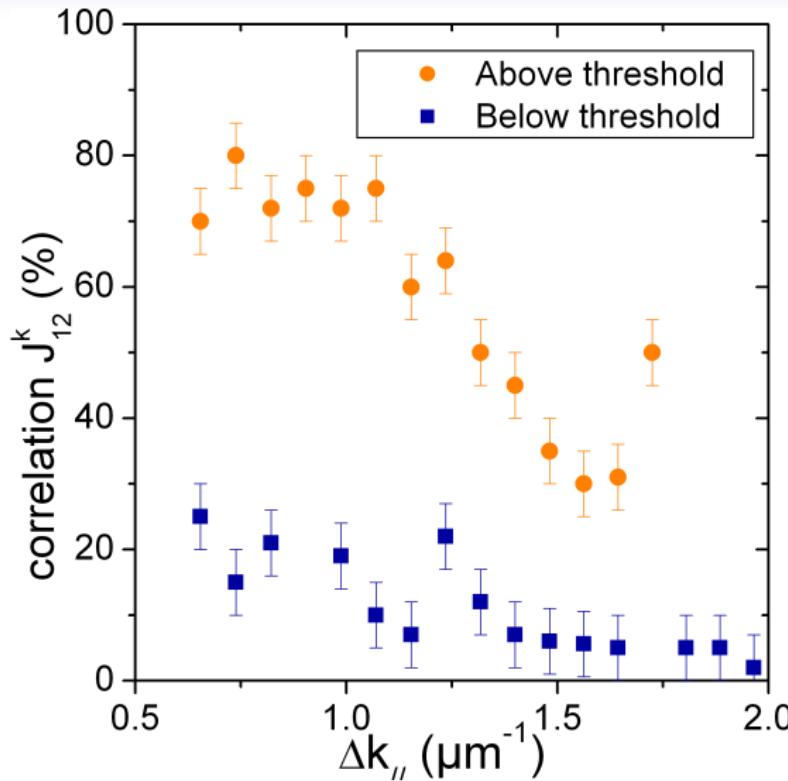
Build-up of Coherence - Further Proofs

Interference Pattern



Build-up of Coherence - Further Proofs

Enhancement of Contrast $\times 4$



Stimulation on the Ring of Excited States

Spotsize Dependence, explained in Phys. Rev. B **77**, 115340 (2008)

