Instability of an exciton-polariton condensate

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Collaborations

Theory

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Experiment

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S. A. Maier (Imperial)
S. Kéna-Cohen (Montreal)
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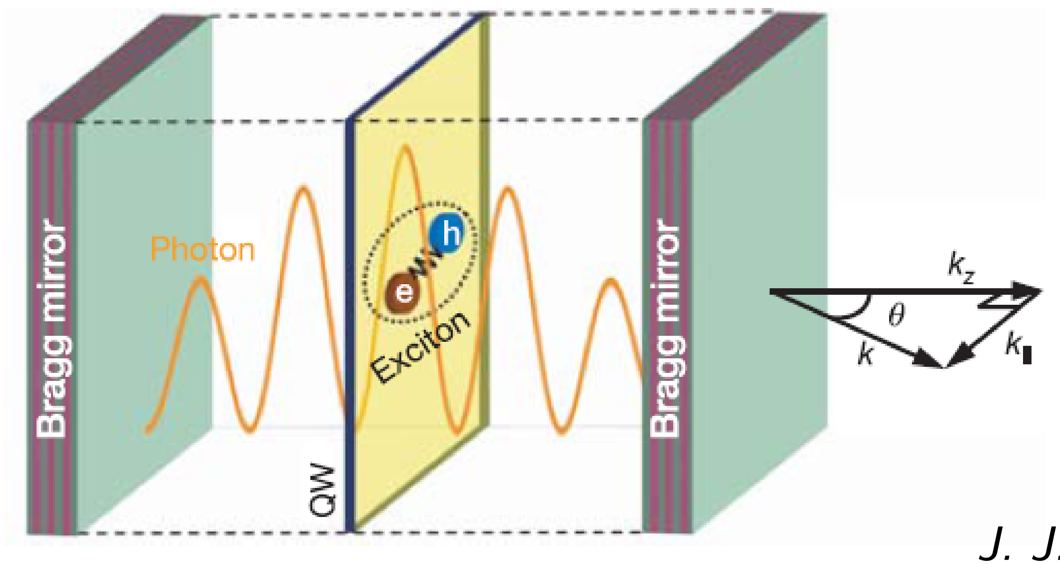
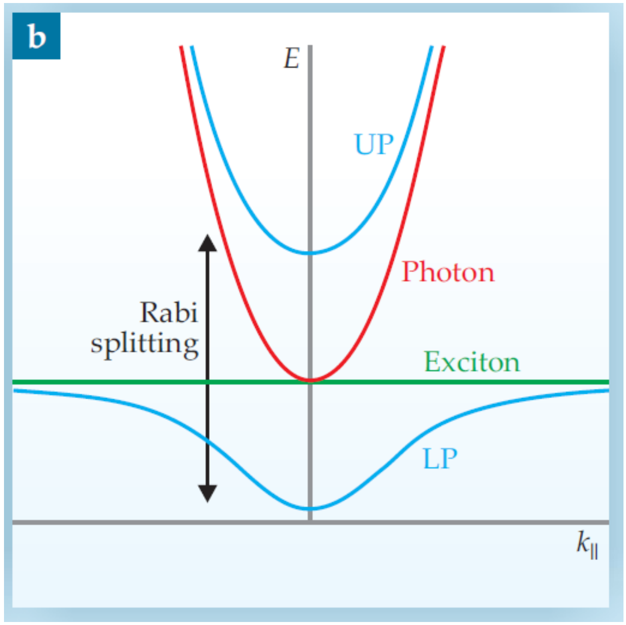
M. Steger, D. W. Snoke (Pittsburgh)
L. Pfeiffer, K. West (Princeton)

Instability of a nonresonantly pumped exciton-polariton condensate

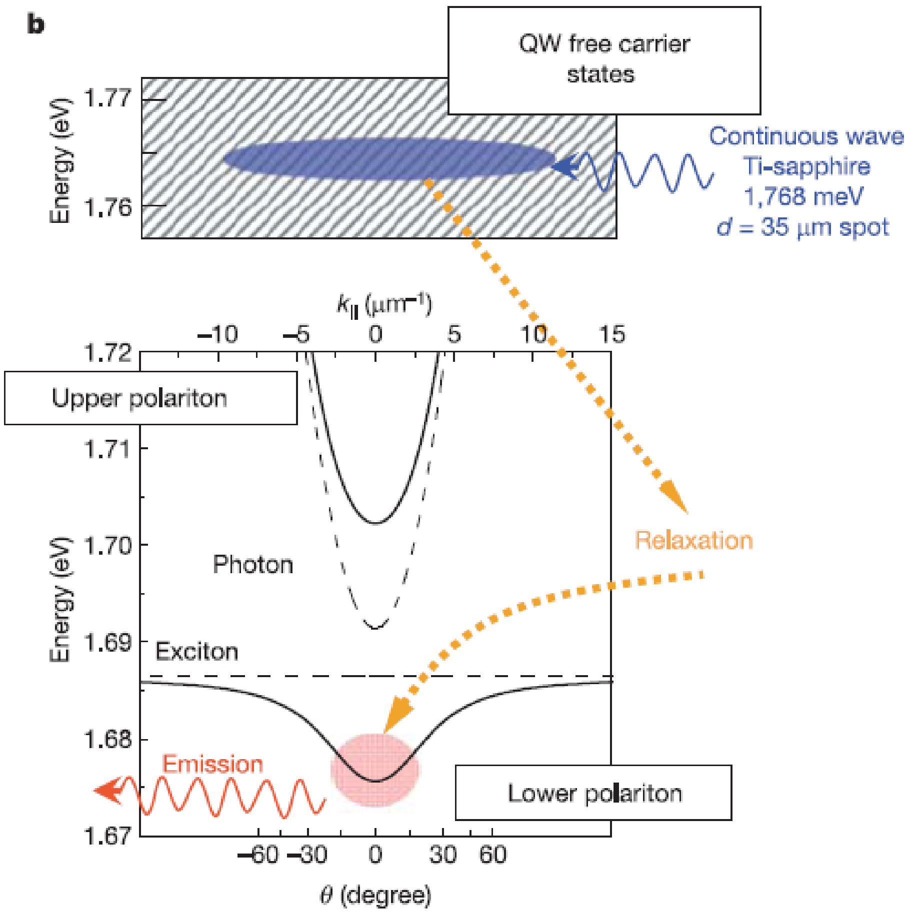
Exciton-Polaritons



J. J. Hopfield, 1958
C. Weisbuch et al., 1992
J. Kasprzak et al., 2006

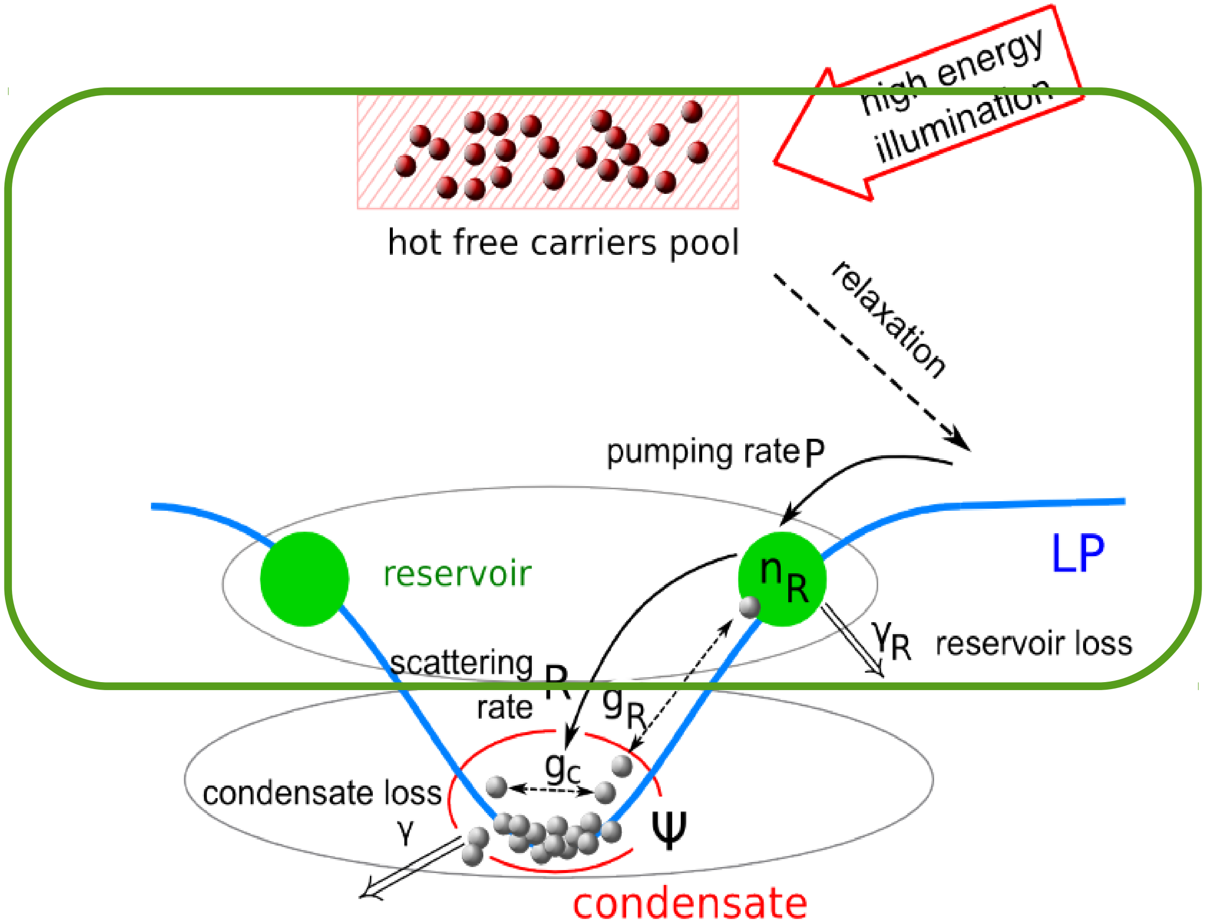


Exciton-Polariton condensation



Nonresonant excitation - theoretical description

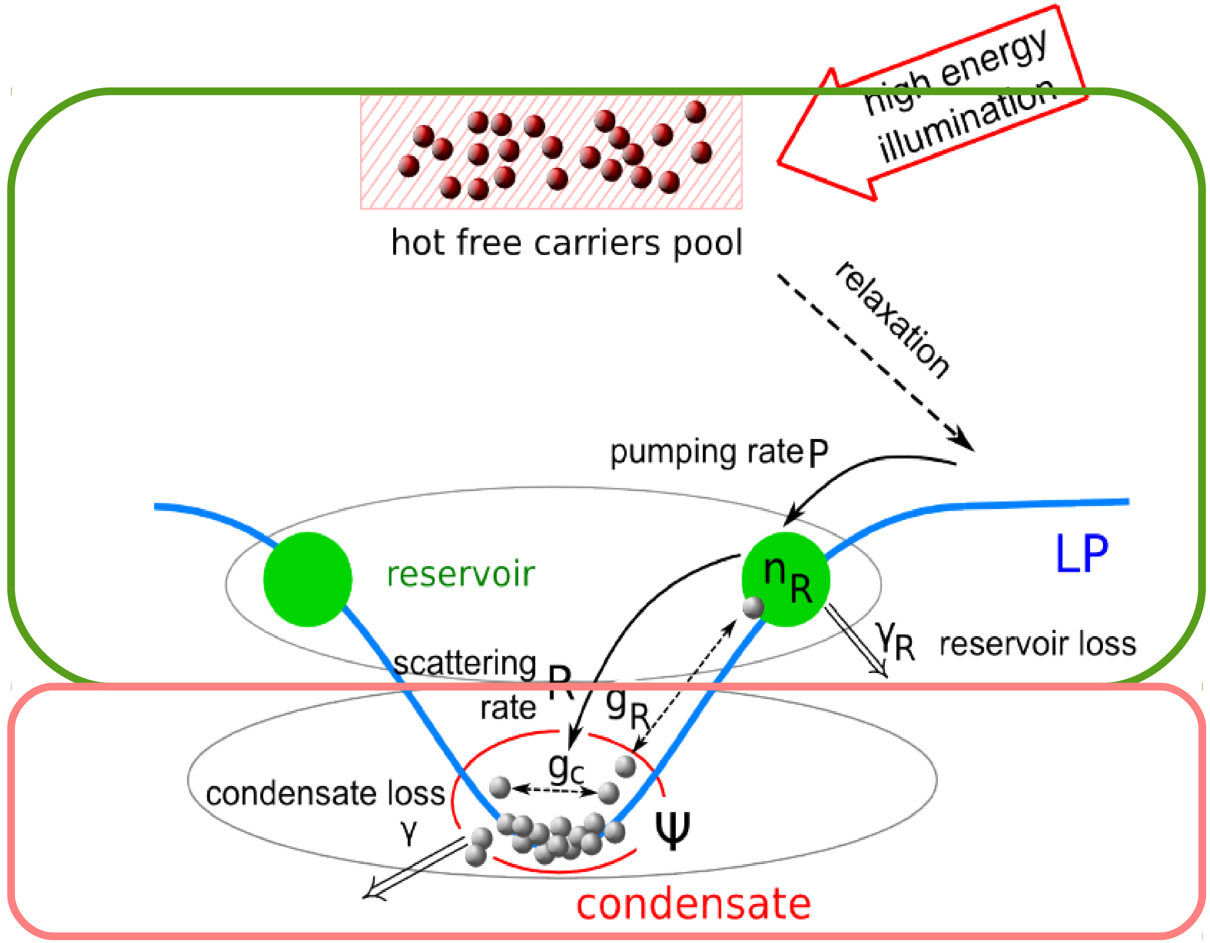
Semiclassical Boltzmann equations
– no coherence



Nonresonant excitation - theoretical description

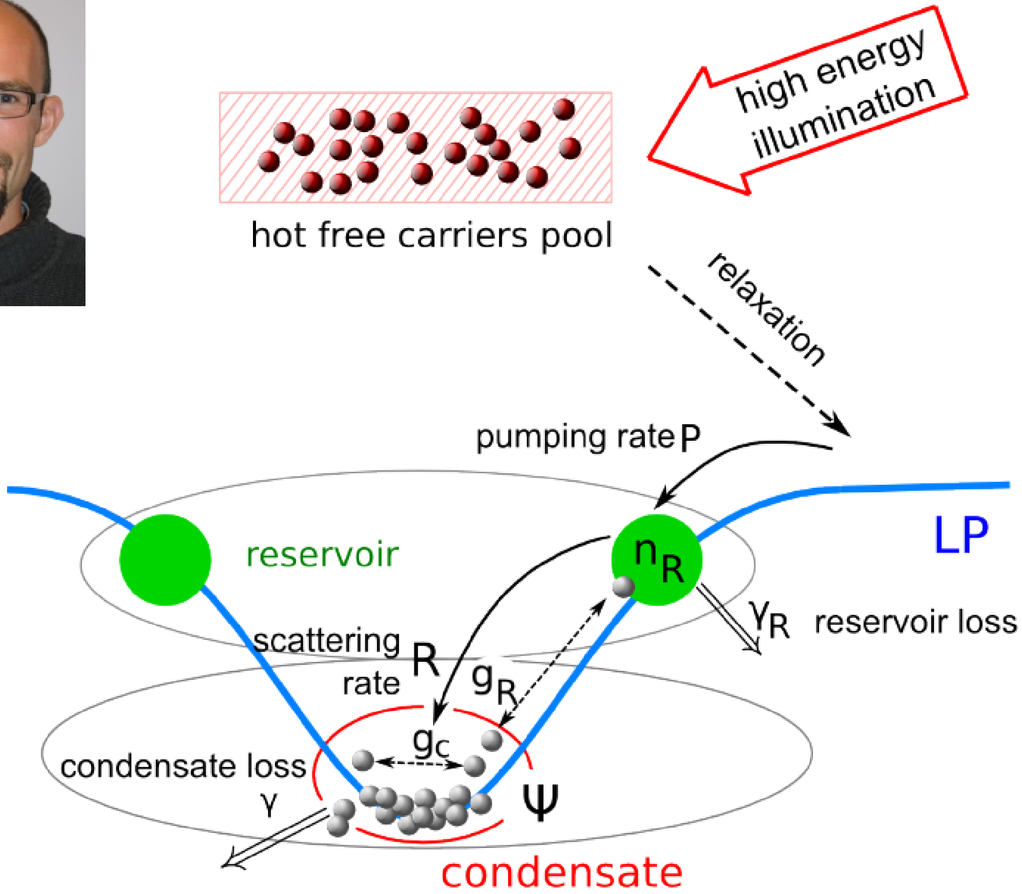
Semiclassical Boltzmann equations
– no coherence

Mean field approximation
– full coherence



$$\Psi(x_1,\dots,x_N)=\prod_{i=1}^{N}\psi(x_i)$$

Open-dissipative Gross-Pitaevskii equation (ODGPE)



Rate equation for the reservoir

$$\frac{\partial n_{\mathrm{R}}}{\partial t} = P(x) - (\gamma_{\mathrm{R}} + R^{1\mathrm{D}}|\psi|^2)n_{\mathrm{R}}$$

Gross-Pitaevskii equation describing condensate

$$i\hbar\frac{\partial\psi}{\partial t} = \left[-\frac{\hbar^2}{2m^*}\frac{\partial^2}{\partial x^2} + g_{\mathrm{C}}^{1\mathrm{D}}|\psi|^2 + g_{\mathrm{R}}^{1\mathrm{D}}n_{\mathrm{R}} + i\frac{\hbar}{2}\left(R^{1\mathrm{D}}n_{\mathrm{R}} - \gamma_{\mathrm{C}}\right)\right]\psi$$

Wouters and Carusotto, PRL 2007

Models of nonresonantly excited condensate

Mean field models neglect the effects of quantum fluctuations $\Psi = \prod_i \psi(x_i)$

Open-dissipative Gross-Pitaevskii equation

$$i\mathrm{d}\psi = \left[-\frac{\hbar}{2m^*}\frac{\partial^2}{\partial x^2} + g|\psi|^2 + g_R n_R + \right.$$
$$\left. + \frac{i}{2}\left(R n_R - \gamma\right)\right]\psi \mathrm{d}t$$
$$\frac{\partial n_R}{\partial t} = P - \left(\gamma_R + R|\psi|^2\right) n_R,$$

Includes a separate equation for the reservoir

M. Wouters, I. Carusotto, 2007

Complex Ginzburg-Landau Equation (CGLE)

$$i\mathrm{d}\phi = \left[\omega_0 - \frac{\hbar}{2m_0^*}\frac{\partial^2}{\partial x^2} + g_0|\phi|^2 + \right.$$
$$\left. + i\left(\frac{P_0}{1 + \frac{|\phi|^2}{n_s}} - \gamma_0\right)\right]\phi\,\mathrm{d}t$$

Single equation only

J. Keelling, N. Berloff, 2008

Both models were successful in modelling experiments in certain conditions

Models of nonresonantly excited condensate

Stochastic versions include the effects of quantum fluctuations

$$\Psi = \prod_i \psi(x_i) + \delta\psi$$

Open-dissipative Gross-Pitaevskii equation

$$\mathrm{i}\mathrm{d}\psi = \left[-\frac{\hbar}{2m^*}\frac{\partial^2}{\partial x^2} + g|\psi|^2 + g_R n_R + \right.$$
$$\left. + \frac{i}{2}\left(R n_R - \gamma\right)\right]\psi \mathrm{d}t + \mathrm{d}W,$$
$$\frac{\partial n_R}{\partial t} = P - \left(\gamma_R + R|\psi|^2\right) n_R,$$

Complex Ginzburg-Landau Equation (CGLE)

$$i\mathrm{d}\phi = \left[\omega_0 - \frac{\hbar}{2m_0^*}\frac{\partial^2}{\partial x^2} + g_0|\phi|^2 + \right.$$
$$\left. + i\left(\frac{P_0}{1 + \frac{|\phi|^2}{n_s}} - \gamma_0\right)\right]\phi\, \mathrm{d}t + \mathrm{d}W_{\mathrm{SGPE}}$$

Derivation using Truncated Wigner Approximation: *M. Wouters, V. Savona, 2009*

Equivalent results can be obtained within the Keldysh formalism:

M. Szymańska, J. Keeling, P. Littlewood, 2006

Models of nonresonantly excited condensate

What are the differences between the models?

Open-dissipative Gross-Pitaevskii equation

$$\mathrm{i}\mathrm{d}\psi = \left[-\frac{\hbar}{2m^*}\frac{\partial^2}{\partial x^2} + g|\psi|^2 + g_R n_R + \right.$$
$$\left. + \frac{i}{2}\left(R n_R - \gamma\right)\right]\psi \mathrm{d}t + \mathrm{d}W,$$
$$\frac{\partial n_R}{\partial t} = P - \left(\gamma_R + R|\psi|^2\right) n_R,$$

Complex Ginzburg-Landau Equation (CGLE)

$$\mathrm{i}\mathrm{d}\phi = \left[\omega_0 - \frac{\hbar}{2m_0^*}\frac{\partial^2}{\partial x^2} + g_0|\phi|^2 + \right.$$
$$\left. + \; i\left(\frac{P_0}{1+\frac{|\phi|^2}{n_s}} - \gamma_0\right)\right]\phi\,\mathrm{d}t + \mathrm{d}W_{\mathrm{SGPE}}$$

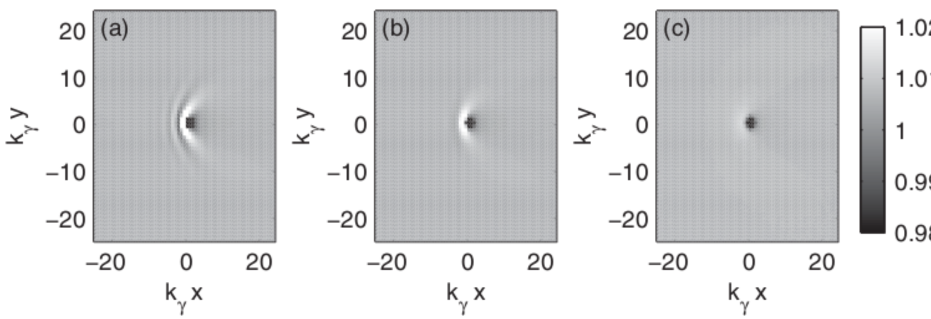
ODGPE can be reduced to CGLE under the “adiabatic approximation”: the reservoir follows.

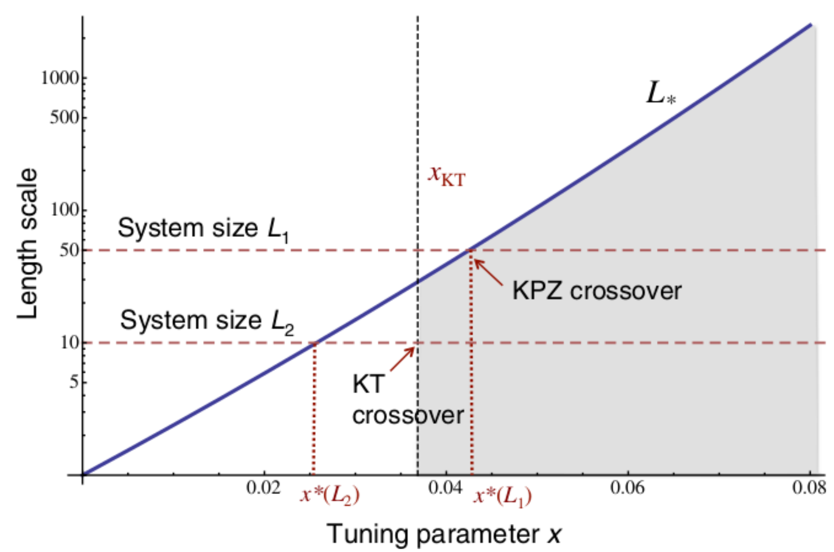
When is it a realistic assumption? What are the physical consequences?

Models of nonresonantly excited condensate

Fundamental description of polariton condensates with the CGLE-like models

- Superfluidity *(M. Wouters, I. Carusotto, PRL 2010; E. Altman et al., PRX 2015)*
- Fluctuations and correlation functions *(A. Chiocchetta, I. Carusotto, EPL 2013; V. N. Gladilin et al., PRB 2014)*
- Critical phenomena *(L.M. Sieberer et al., PRL 2013)*
- Kardar-Parisi-Zhang physics *(E. Altman et al., PRX 2015; V. N. Gladilin et al., PRB 2014; L. He et al, PRB 2015)*
- Superfluid stiffness with disorder *(A. Janot et al., PRL 2013)*





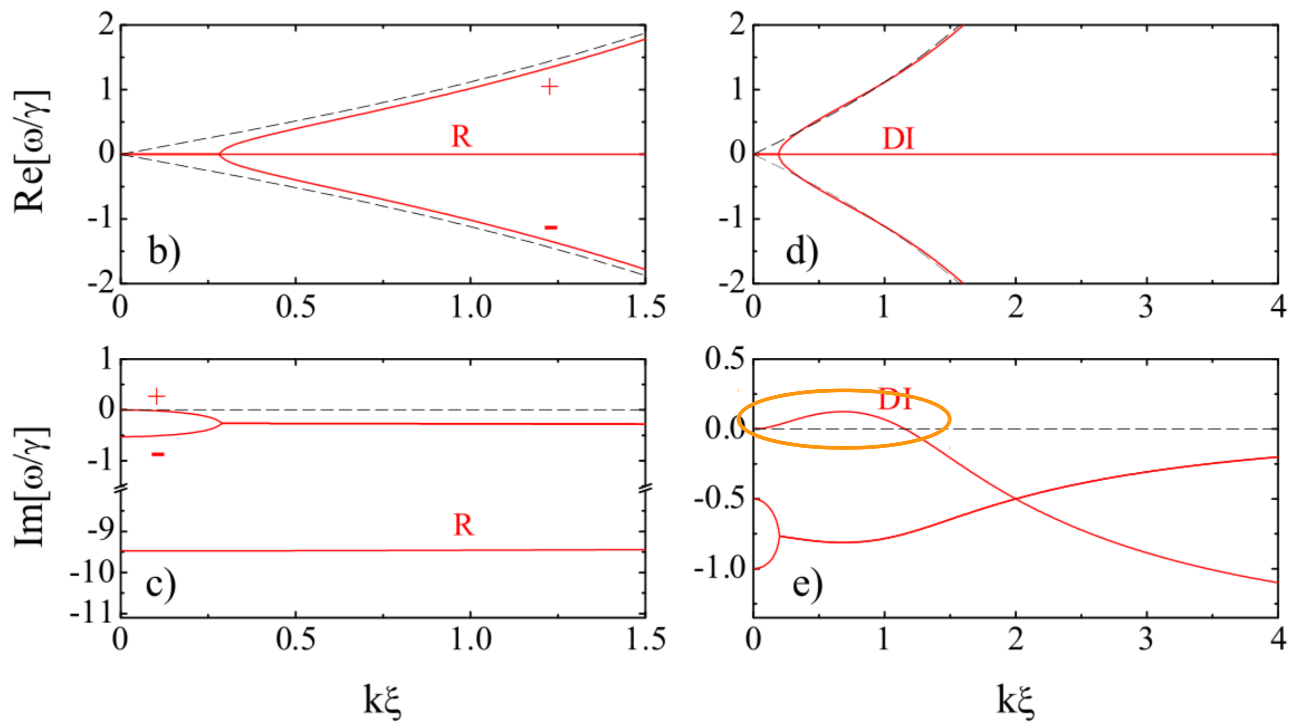
Bogoliubov approximation

Excitations of a homogeneous condensate (u, v, w small)

$$\psi=\left(\psi_0+u\,e^{ikx-i\omega t}+v^*\,e^{-ikx+i\omega^* t}\right)e^{-i\mu t},\; n_R=n_{R0}+w\cos(kx+\omega t)$$

Fluctuations with imaginary frequencies can be unstable

M. Wouters, I. Carusotto, 2007



Condensate instability

The stability condition for ODGPE is

L.A. Smirnov et al., 2014

$$\frac{P}{P_{\text{th}}} > \frac{g_R}{g_C}\frac{\gamma_C}{\gamma_R}.$$

Typically:

$g_R \approx g_C, \gamma_R \ll \gamma_C$

while the CGLE is always stable for repulsive interparticle interactions, $g_0 > 0$!

Adiabatic approximation

To reduce the ODGPE to GPE, three conditions have to be fulfilled simultaneously

$$k^2 \ll 2m^*/(\hbar\tau_R)$$

$$\frac{P_{\rm th}}{P} \gg \frac{g_C - R}{g_C},$$

$$\frac{P}{P_{\rm th}} \gg \frac{g_R}{R}\frac{\gamma_C}{\gamma_R}.$$

Bobrovska, MM PRB 2015

Which gives

$$g_0 = g_C - g_R \frac{\gamma_C^2}{RP},$$

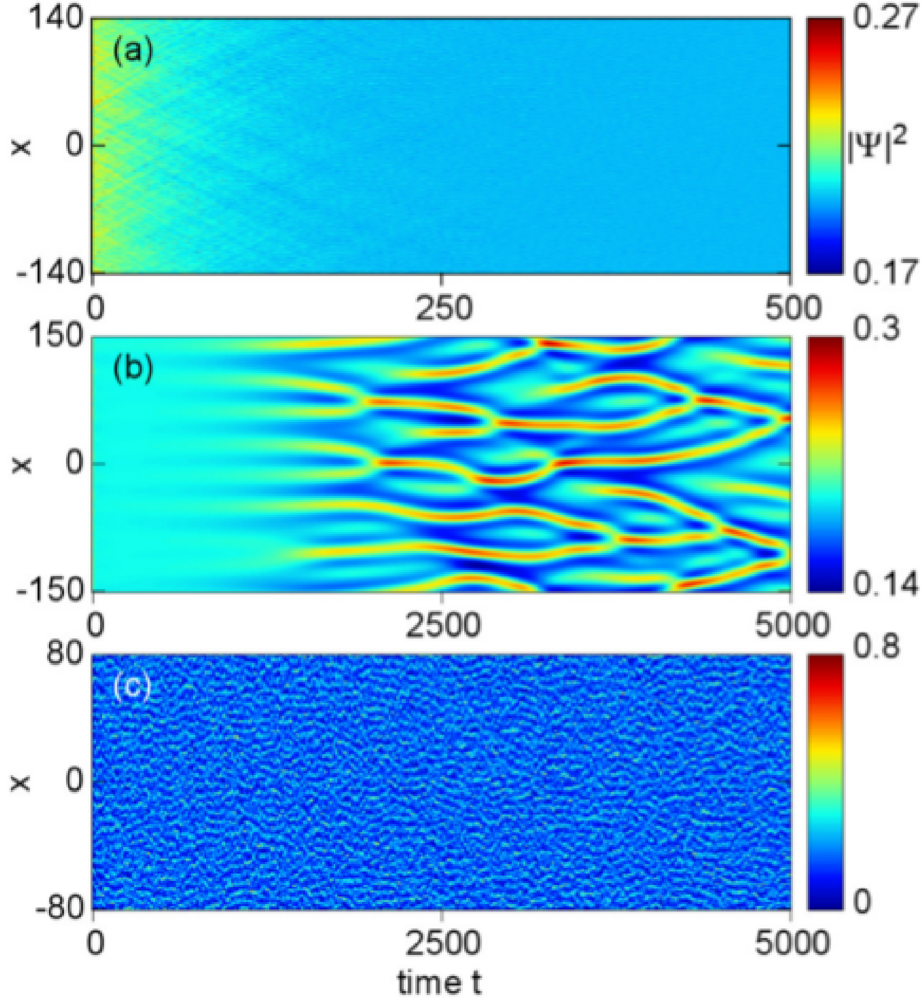
Instability of the ODGPE corresponds to the Benjamin-Feir instability at $g_0 < 0$.
Polariton interactions in the presence of reservoir become effectively attractive.

Condensate instability

No experimental evidence of this instability. Proposed explanations:

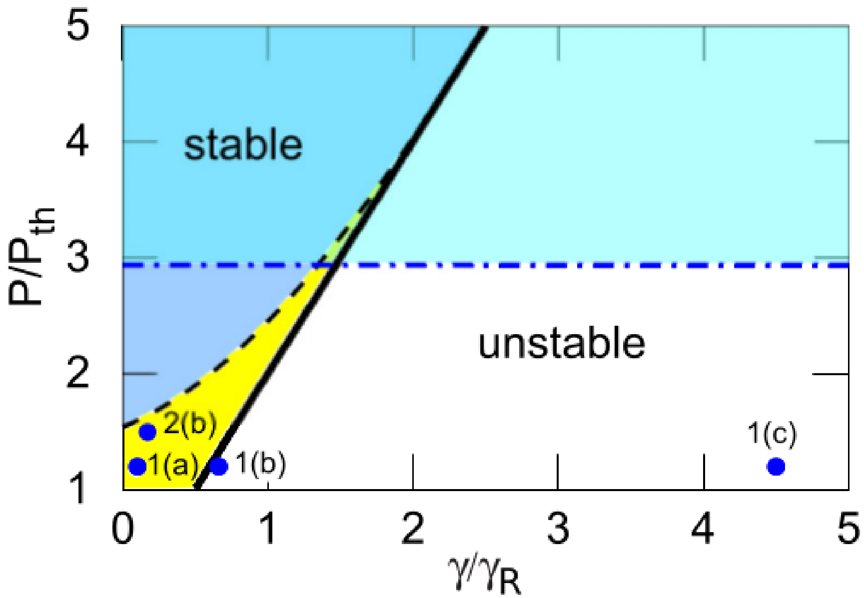
- The instability is unphysical, because the lifetime of reservoir γ_R^{-1} is in fact much shorter than the exciton lifetime (Wouters & Carusotto, 2007)
- The instability is an artifact of ODPGE, which does not take into account energy relaxation
- Or maybe the instability can exist in reality?...

Stability diagram



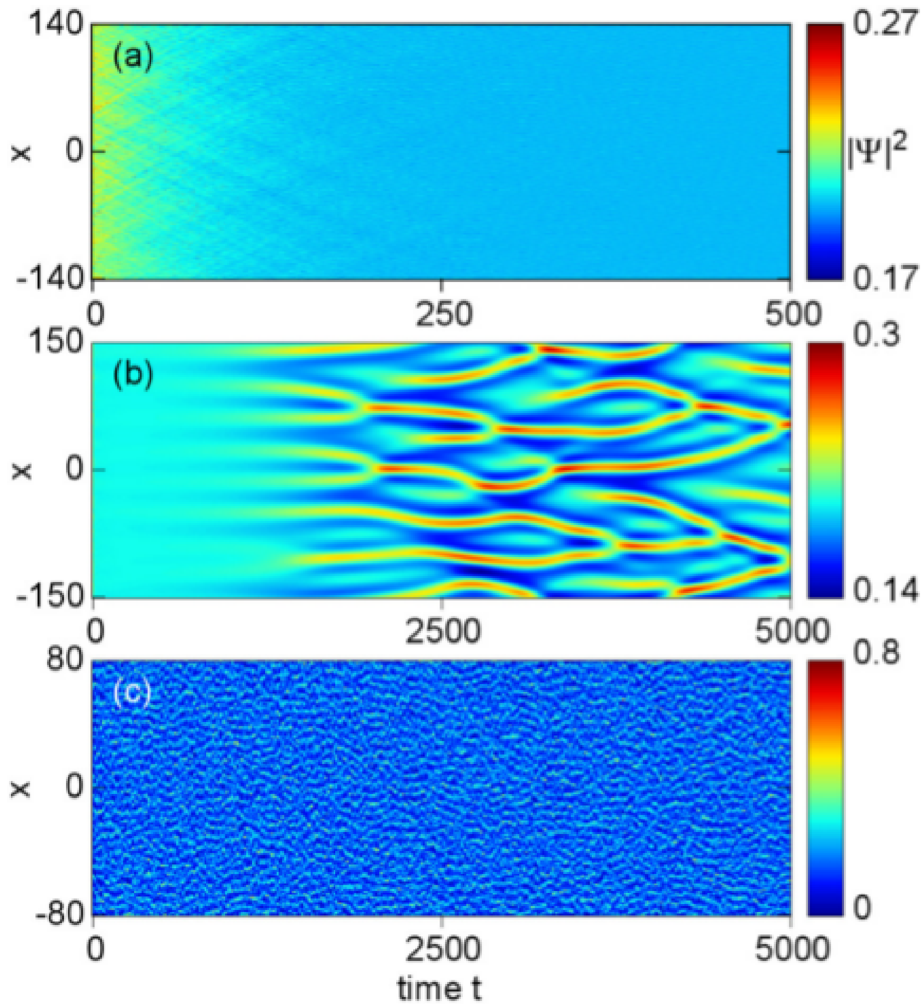
Bobrovska, Ostrovskaya, MM PRB 2014

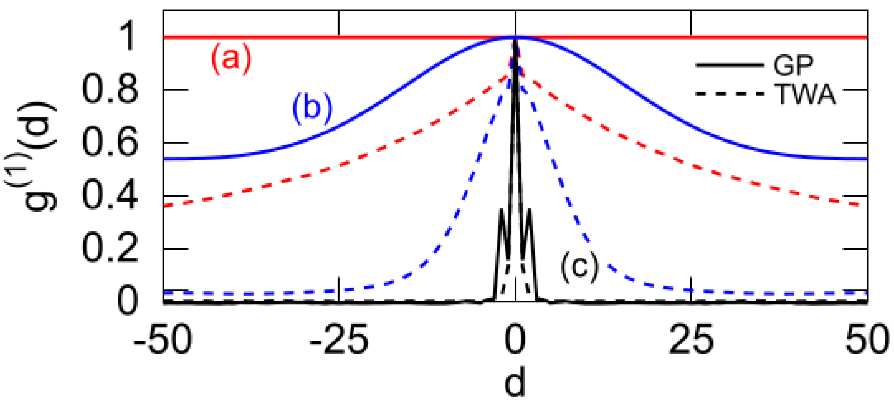
$$\frac{P}{P_{\rm th}} > \frac{g_R}{g_C}\frac{\gamma_C}{\gamma_R}.$$



The instability results in the formation of polariton domains

Stability diagram

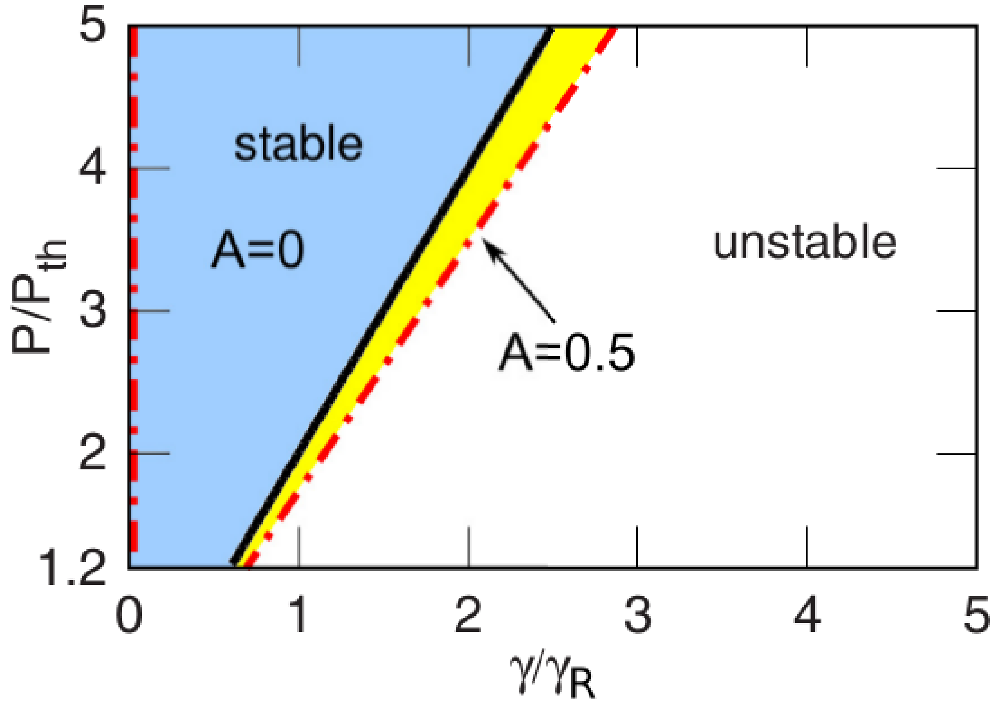




The coherence is strongly reduced, but
no signature in time-averaged intensity

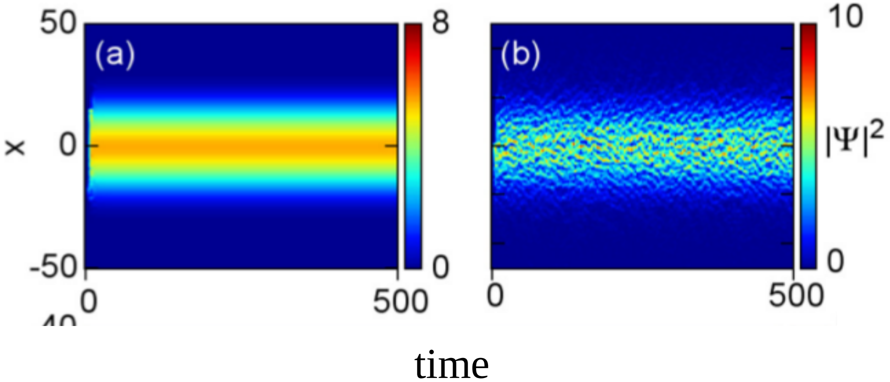
Stability diagram

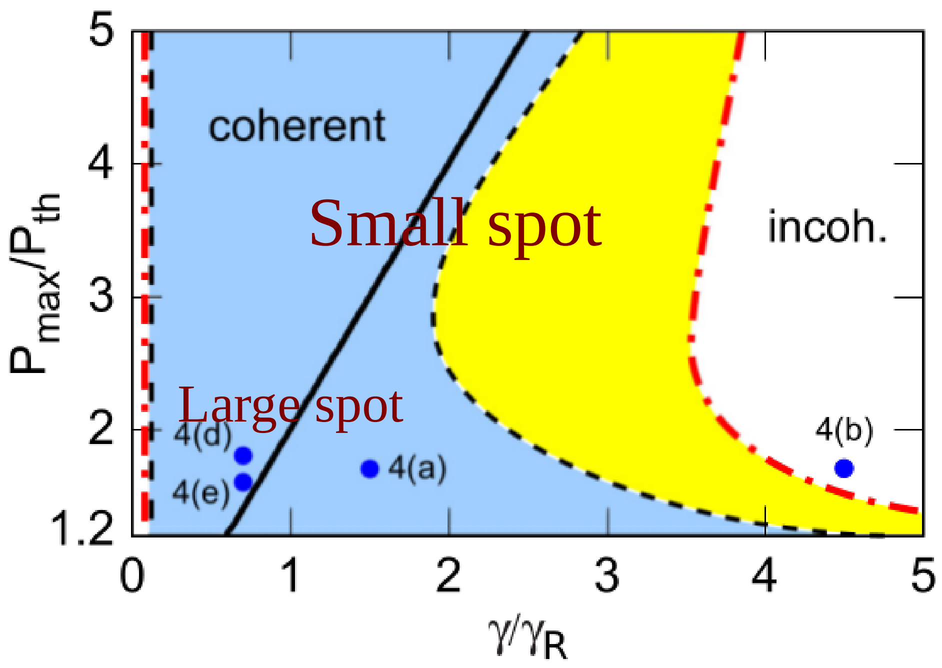
$$\frac{\partial \psi}{\partial t} = \left[(i + A)\frac{\partial^2}{\partial x^2} + \frac{1}{2}(R n_R - \gamma) - i g |\psi|^2 - i g_R n_R \right] \psi,$$



Relaxation does not help much

Finite pump spot size

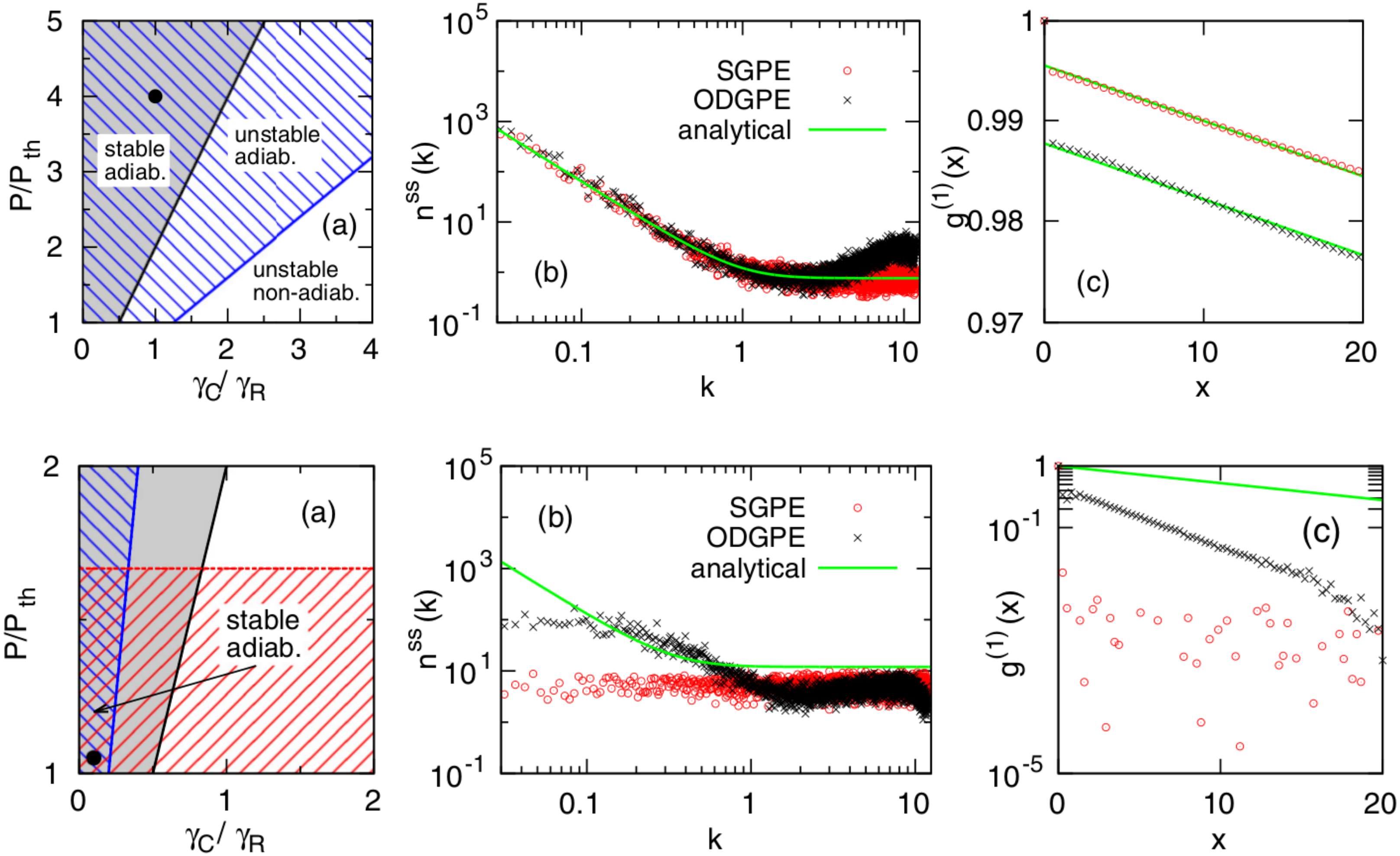




Reduction of pump spot size enlarges the stability region and improves coherence

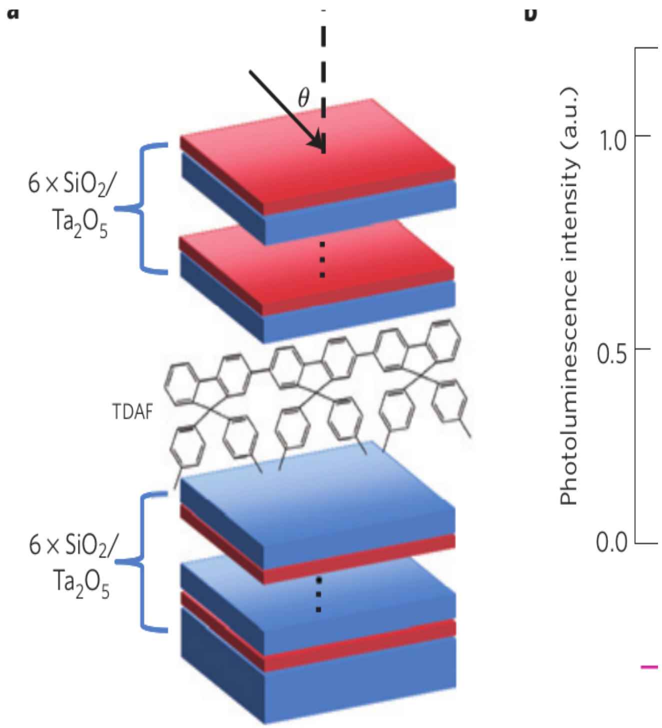
Quantum fluctuation spectrum

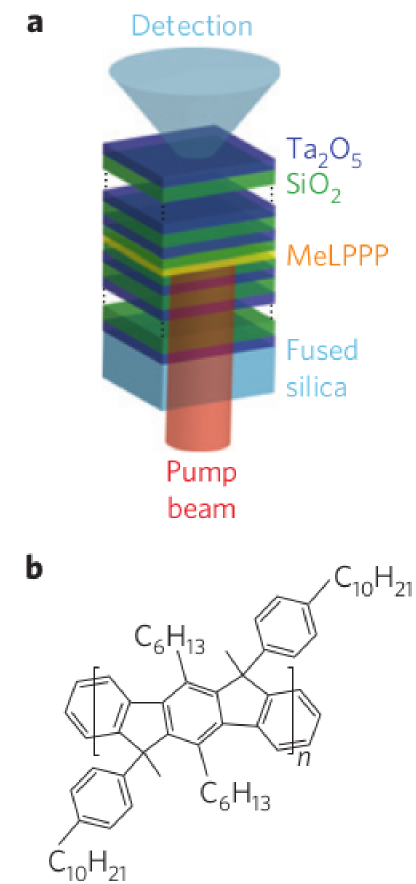
Bobrovska, MM PRB 2015



ODGPE and GPE models are equivalent only in a restricted region of parameters, which does not seem to be in the reach of current experiments!

Polaritons in organic materials





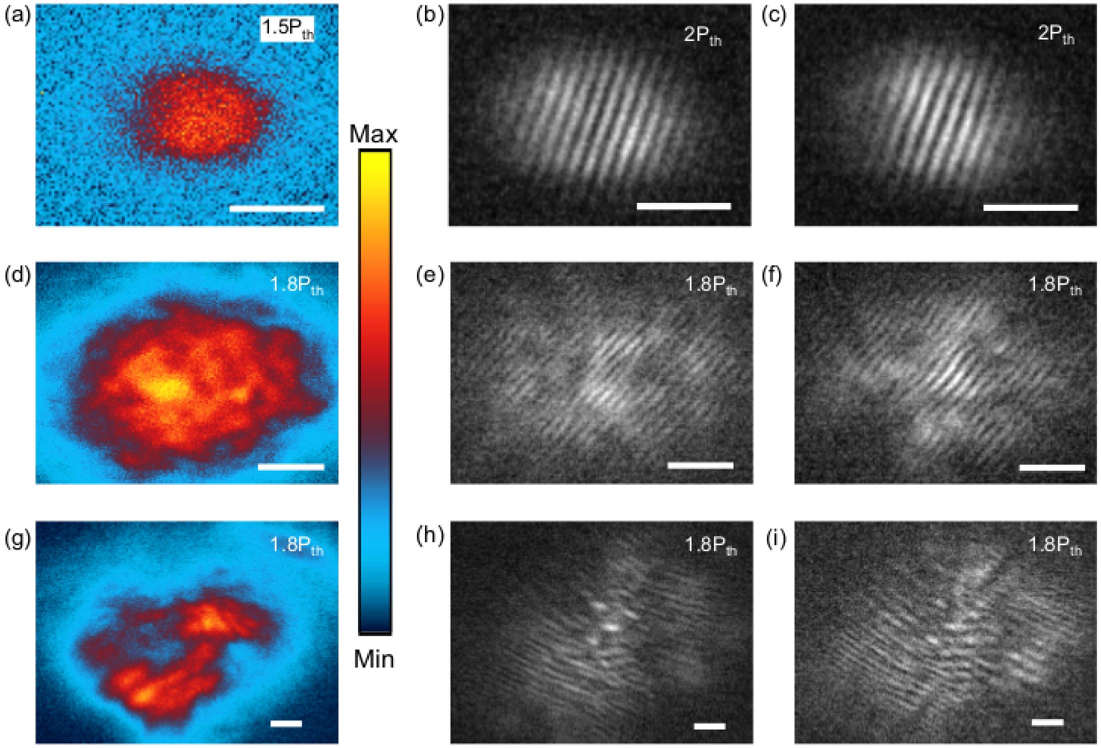
Active layers: Anthracene, MeLPP polymer, oligofluorene (TDAF)

S. Kena-Cohen et al., Nat. Photonics 2010, Nat. Materials 2014
R. Mahrt et al., Nat. Materials 2013

Experiments

Daskalakis, Maier and Kena-Cohen PRL 2015

Single-shot imaging



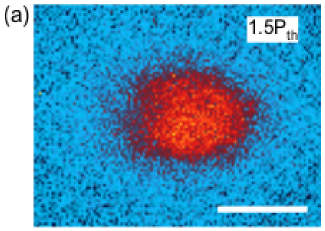
Stephane Kéna-Cohen

Small Gaussian spot

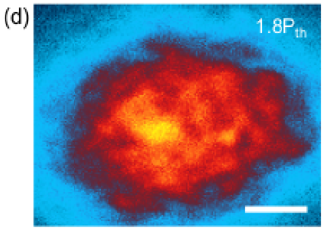
Large Gaussian spot

Large flat-top spot

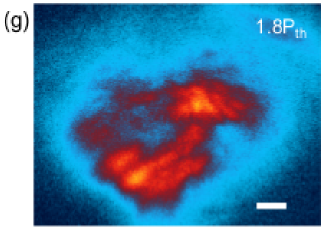
Experiments



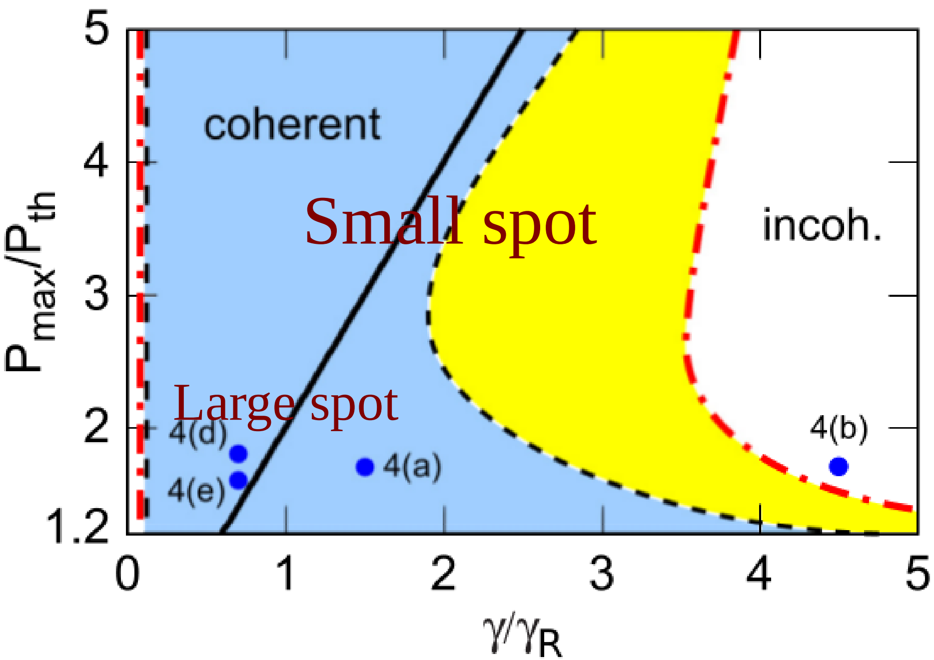
Small Gaussian spot



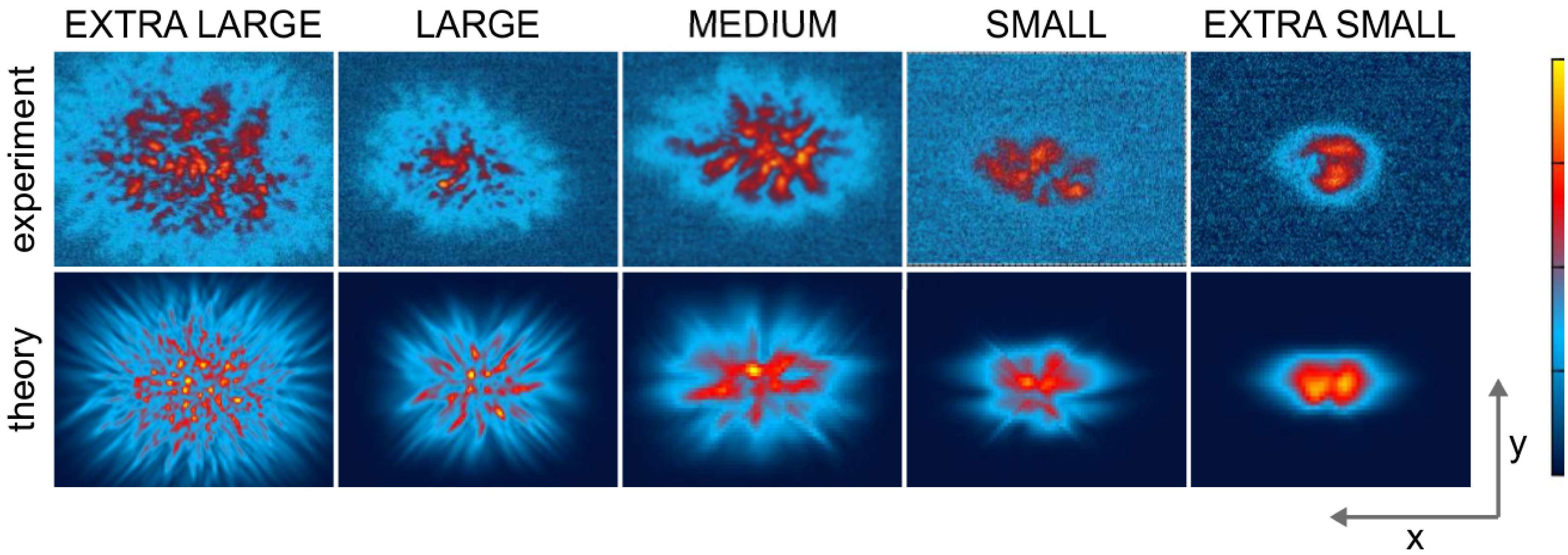
Large Gaussian spot



Large flat-top spot

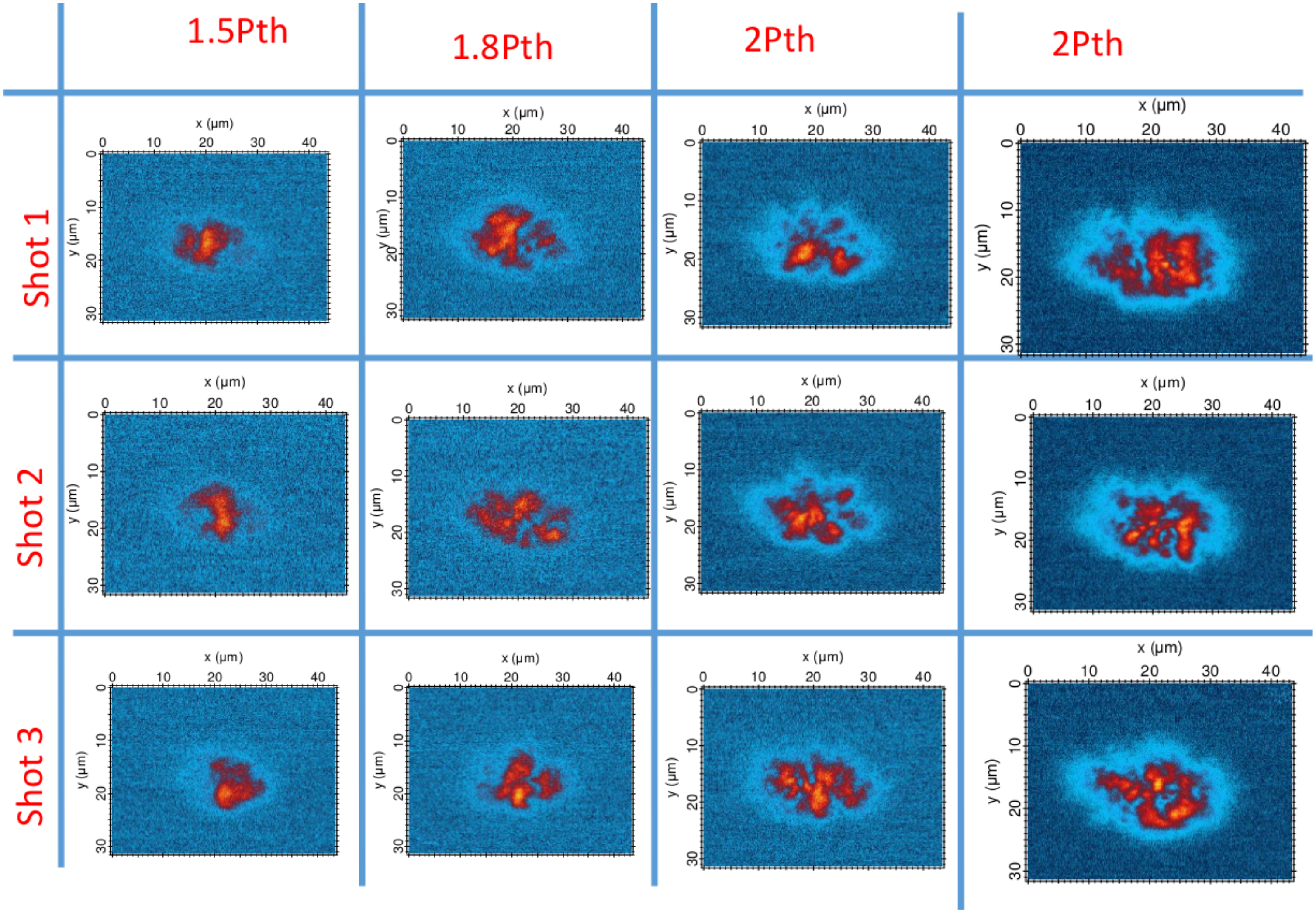


Experiments vs. theory



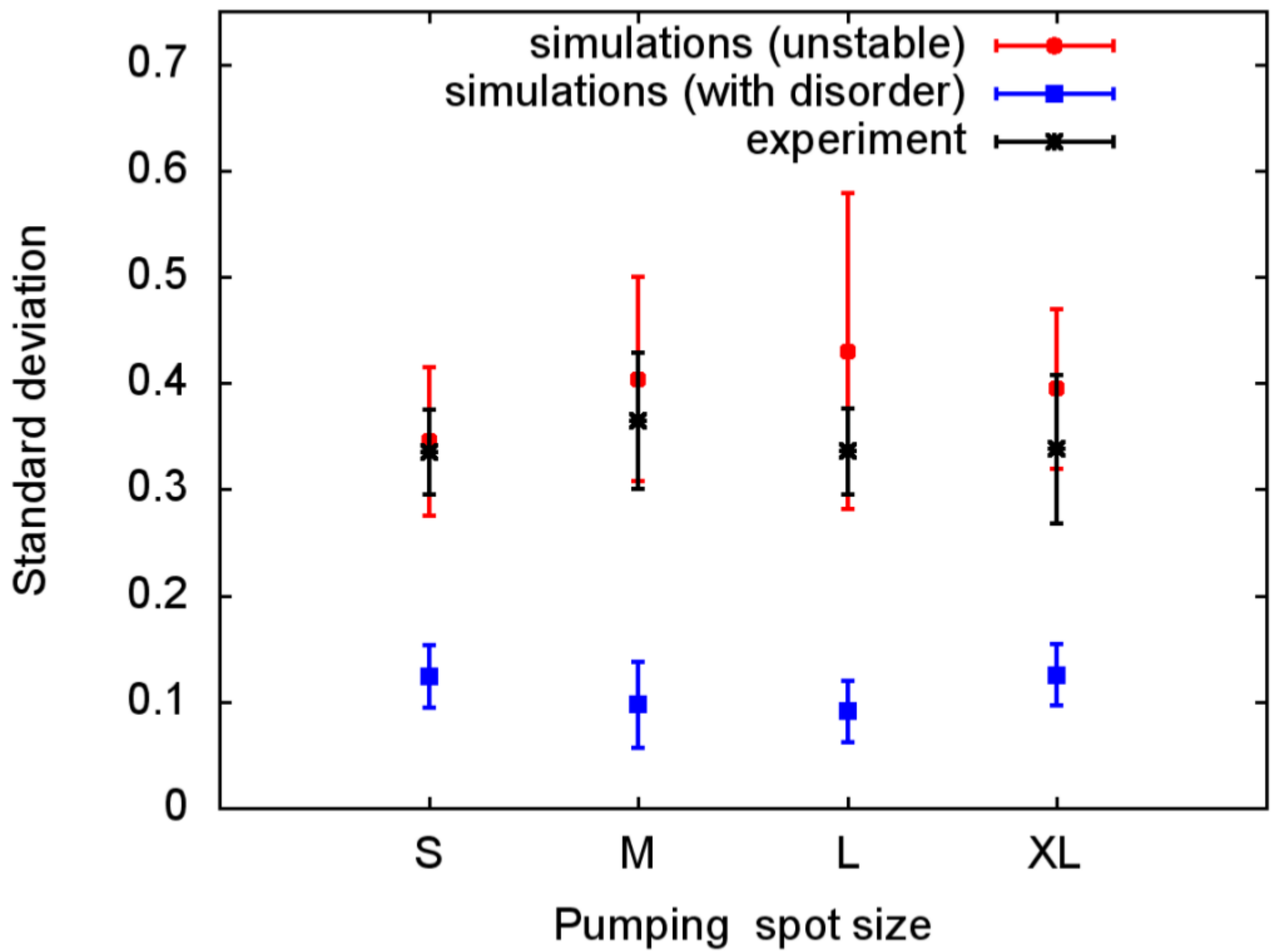
Excellent agreement **without fitting parameters**. All parameters were taken from independent studies on the same sample.

Shot-to-shot fluctuations



The shot-to-shot fluctuations exclude the possibility that the domains are solely due to the sample disorder

Experiments vs. theory



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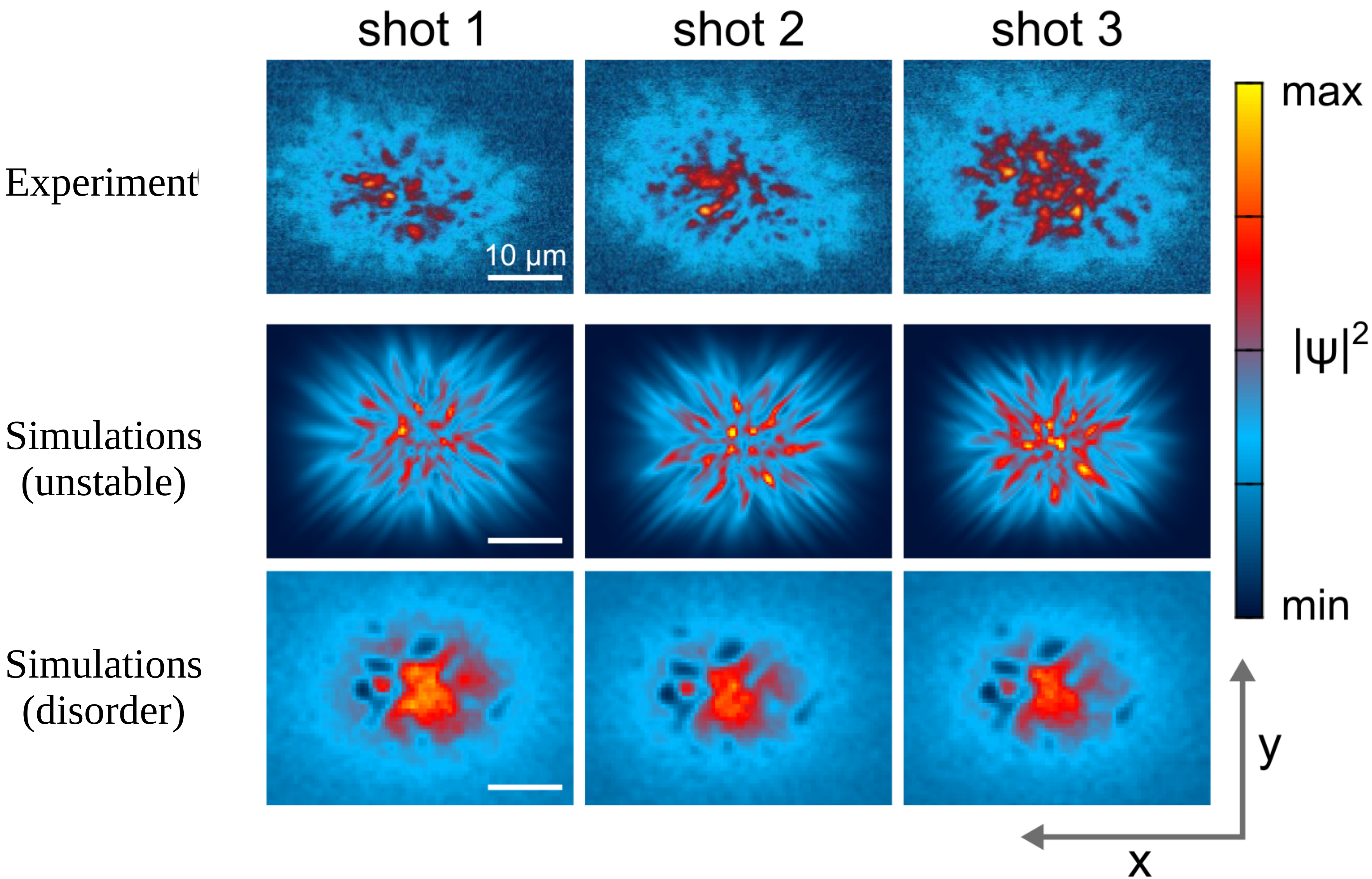
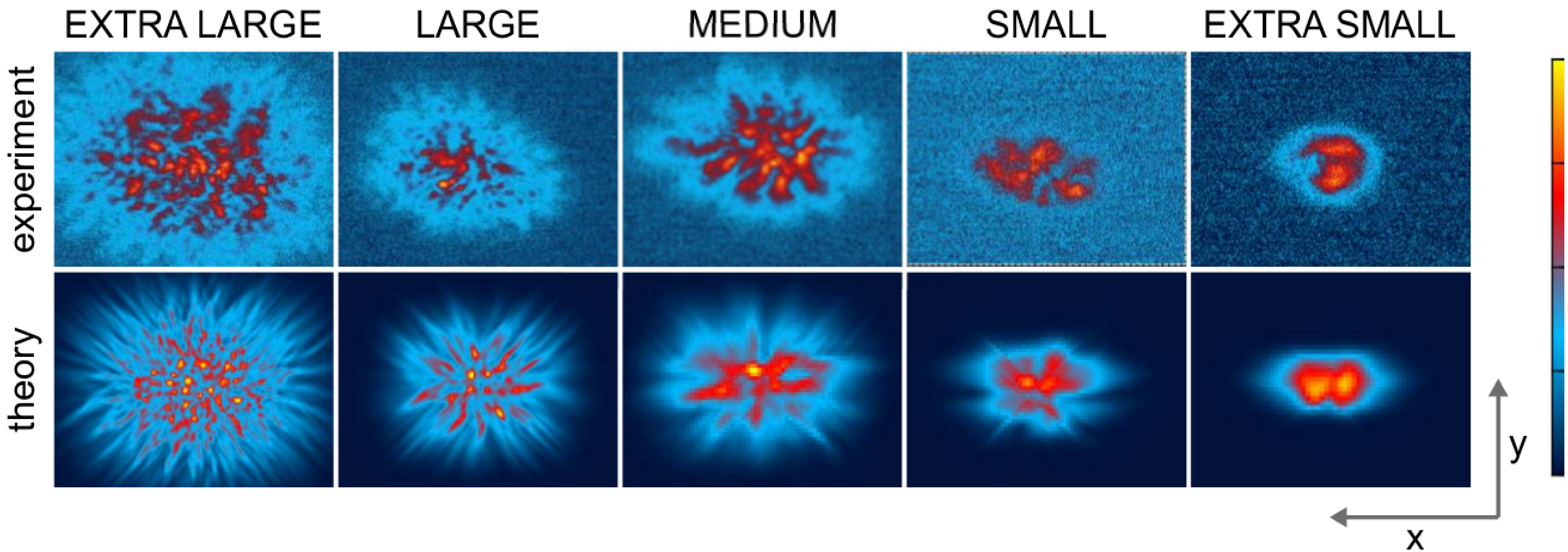


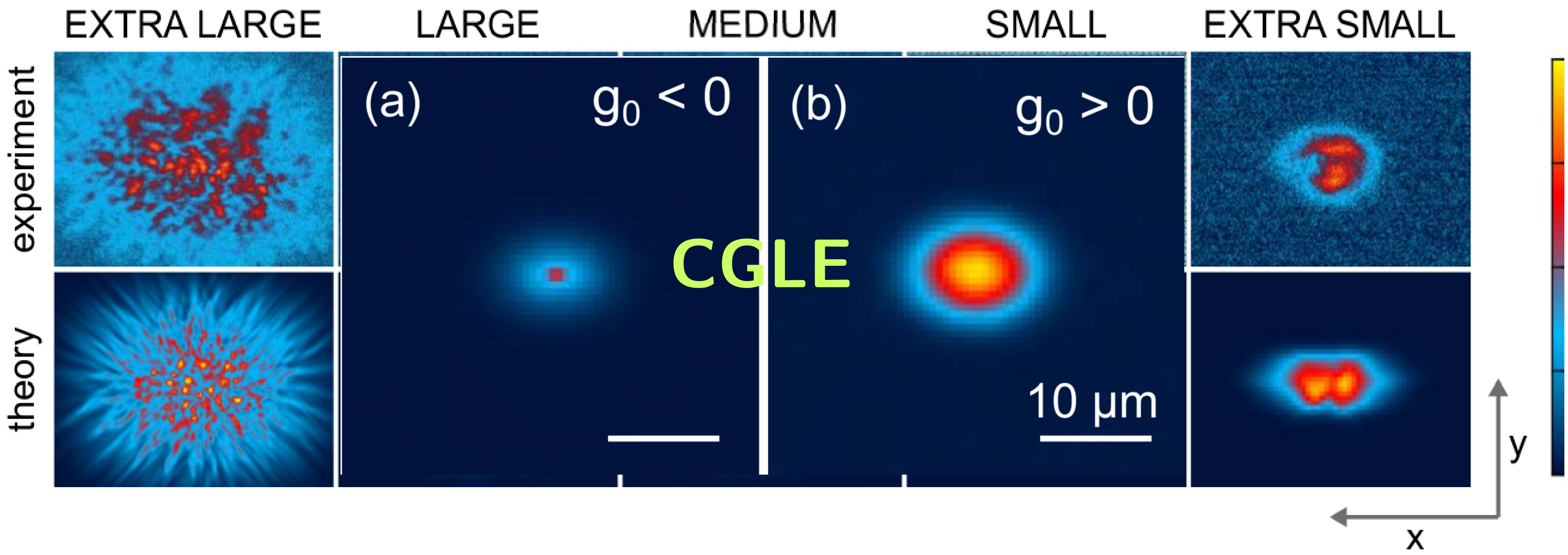
shot 1
shot 2
shot 3
Experiment
Simulations
(unstable)
Simulations
(disorder)
10 μm
max
$|\psi|^2$
min
y
x

Experiments vs. theory



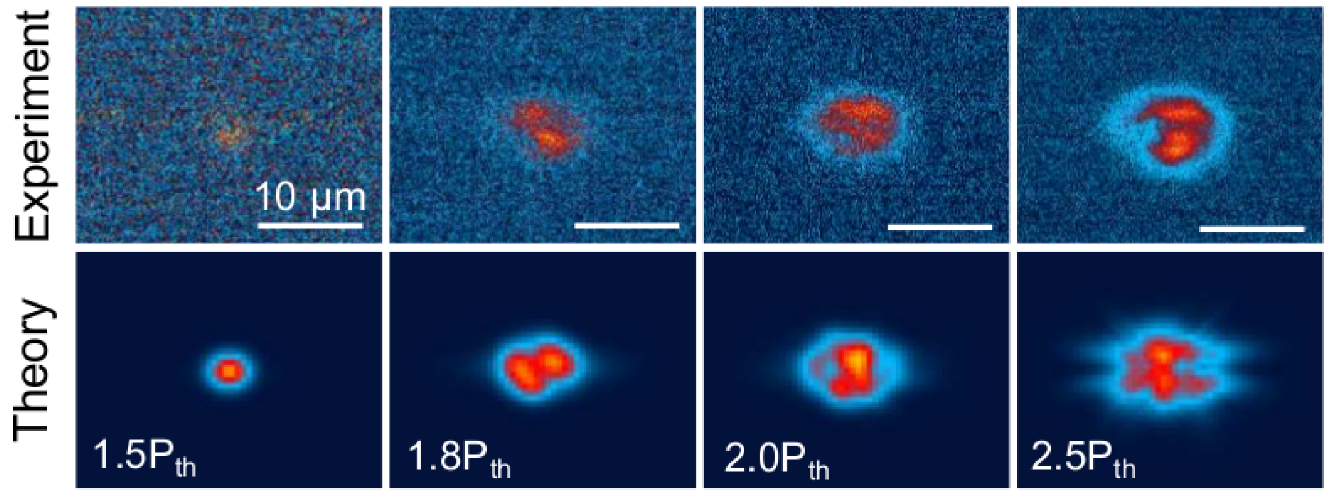
The parameters correspond to a strongly nonadiabatic regime.
Simple CGLE models should not be used!

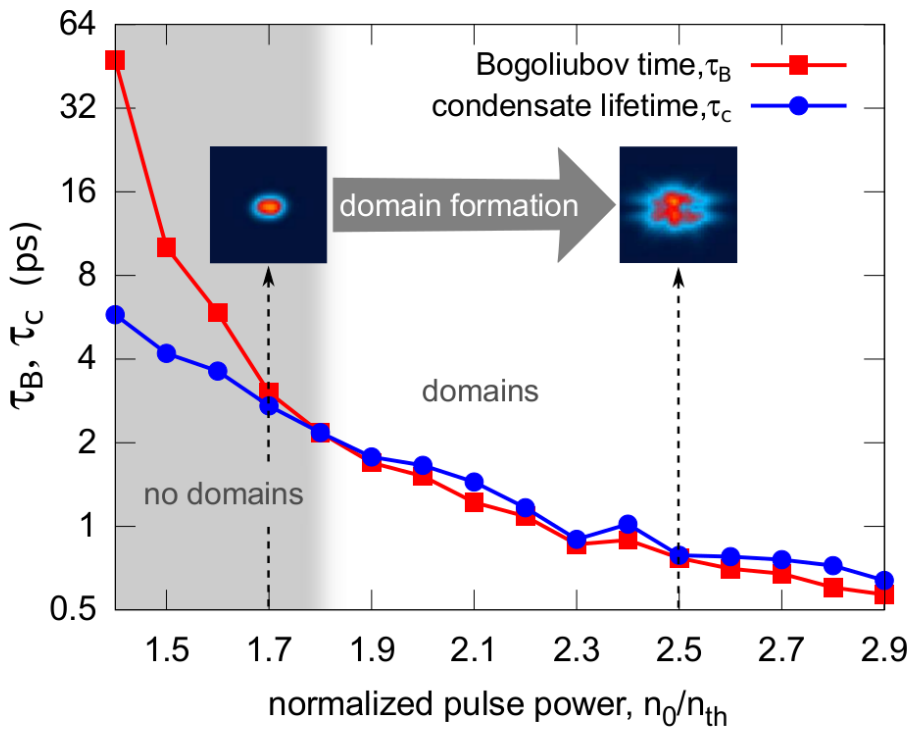
Experiments vs. theory



The parameters correspond to a strongly nonadiabatic regime.
Simple CGLE models should not be used!

Experiments vs. theory



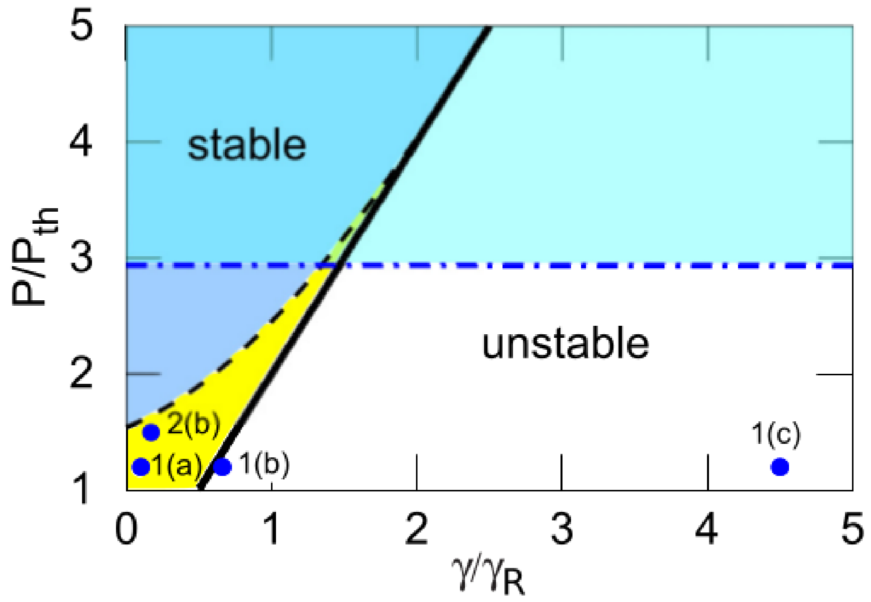


Time-dependent Bogoliubov spectrum

In the case of constant, homogeneous pumping, stability can be investigated by considering small fluctuations (Bogoliubov-de Gennes modes)

$$\psi = \left(\psi_0 + u\, e^{ikx - i\omega t} + v^* e^{-ikx + i\omega^* t}\right) e^{-i\mu t}, \quad n_R = n_{R0} + w \cos(kx + \omega t)$$

Which leads to a simple analytical condition for stability $\frac{P}{P_{\text{th}}} > \frac{g_R}{g_C} \frac{\gamma_C}{\gamma_R}.$



L.A. Smirnov et al., 2014

Time-dependent Bogoliubov spectrum

In the case time-dependent (pulsed pumping), this description fails.

Time-dependent Bogliubov modes can be considered

$$\psi = \psi_0 e^{-\frac{i\mu t}{\hbar} + \frac{\beta_C t}{2}} \left(1 + \sum_{\mathbf{k}} \left[u_{\mathbf{k}} e^{-i(\omega_{\mathbf{k}} t - \mathbf{k}\mathbf{r})} + v_{\mathbf{k}}^* e^{i(\omega_{\mathbf{k}}^* t - \mathbf{k}\mathbf{r})} \right] \right),$$

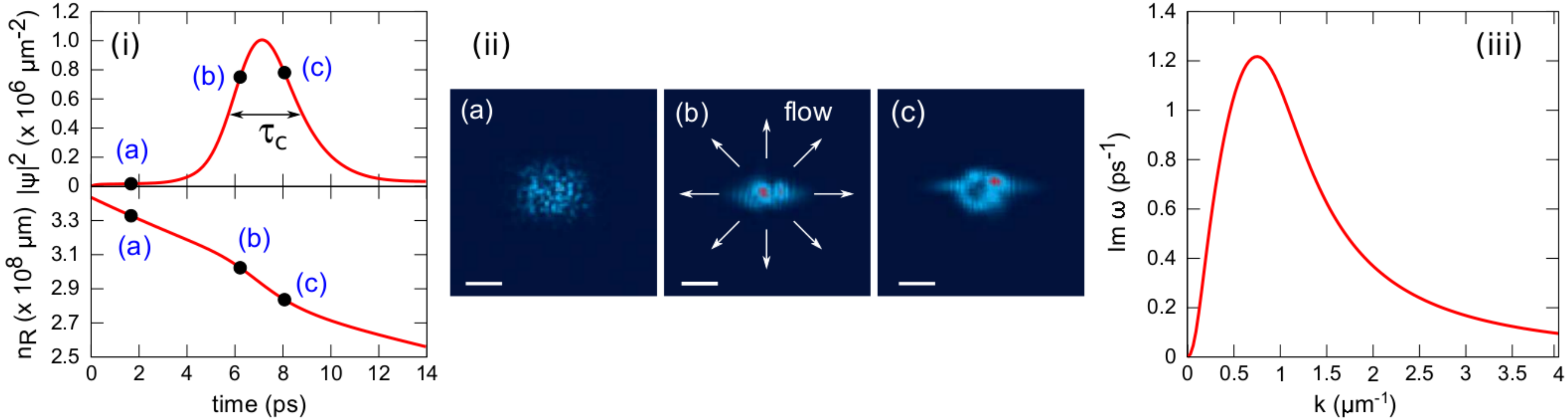
$$n_R = n_R^0 e^{\beta_R t} \left(1 + \sum_{\mathbf{k}} \left[w_{\mathbf{k}} e^{-i(\omega_{\mathbf{k}} t - \mathbf{k}\mathbf{r})} + \text{c.c.} \right] \right),$$

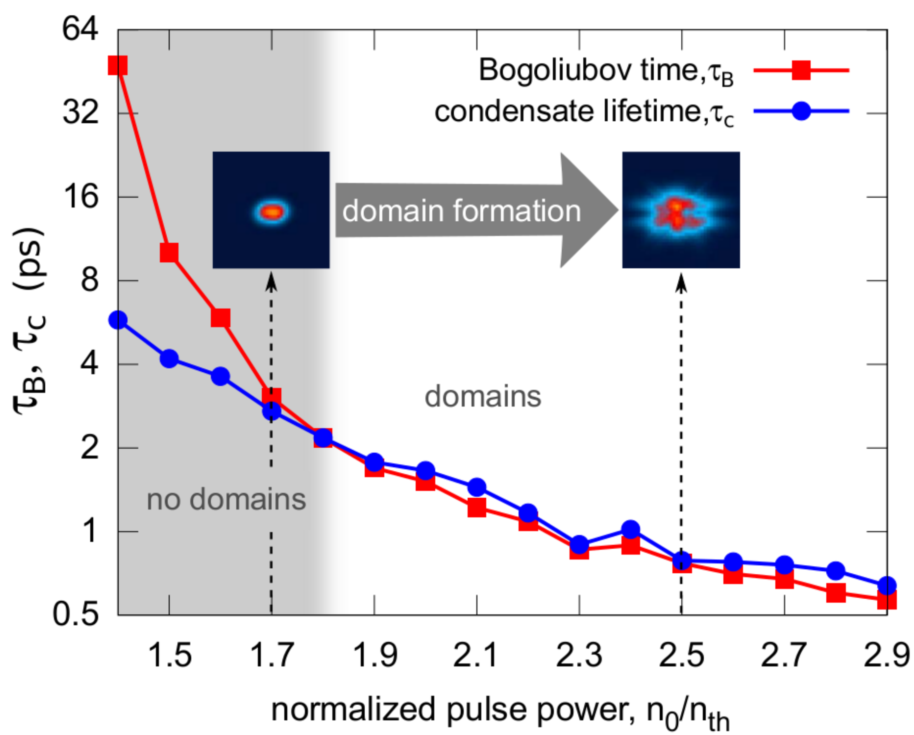
β_C, β_R – **instantaneous** condensate and reservoir density growth rates.

Ważewski inequality (T. Ważewski, 1948): The soultion to the time-dependent Bogoliubov problem is bounded from below and above by the integrated infimum and maximum of imaginary part of $\omega_{\mathbf{k}}$.

Result for $P=P_0\,\delta(t)$: system is always unstable! (Im $\omega_{\mathbf{k}}>0$ for all **k**).

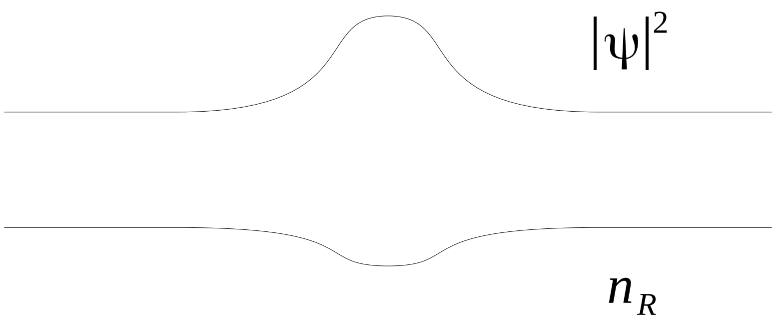
Time-dependent Bogoliubov spectrum





Instability – intuitive explanation

Consider a small density fluctuation in the continuous pumping case:



$$n_R = \frac{P}{\gamma_R + R|\psi|^2} \quad \text{in a steady state}$$

$$\mu = g_c|\psi|^2 + g_R n_R$$

$$\frac{\delta\mu}{\delta|\psi|^2} > 0 \;\Rightarrow\; \frac{P}{P_{\text{th}}} > \frac{g_R}{g_C}\frac{\gamma_C}{\gamma_R}. \qquad \text{stability condition}$$

In the pulsed pumping case, with $P=P_0\,\delta(t)$, number of particles is conserved in the process of scattering to the condensate for t>0

$$\delta|\psi|^2 + \delta n_R = 0 \;\Rightarrow\; \frac{\delta\mu}{\delta|\psi|^2} = g_C - g_R < 0$$

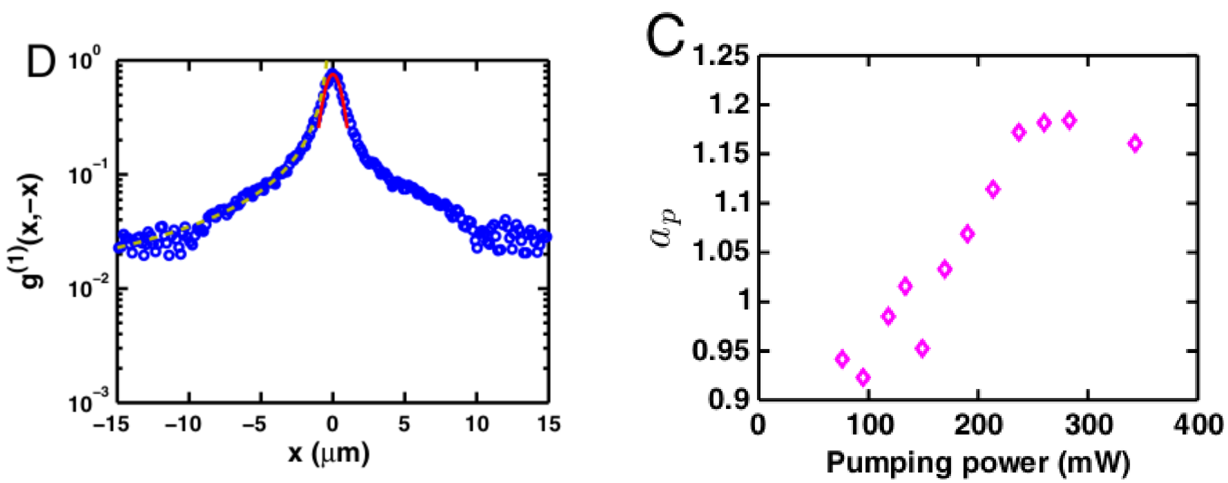
Hence the condensate is always unstable, regardless of other parameters.

Fragmentation in inorganic condensates

Relevance to inorganic polaritons (GaAs...)

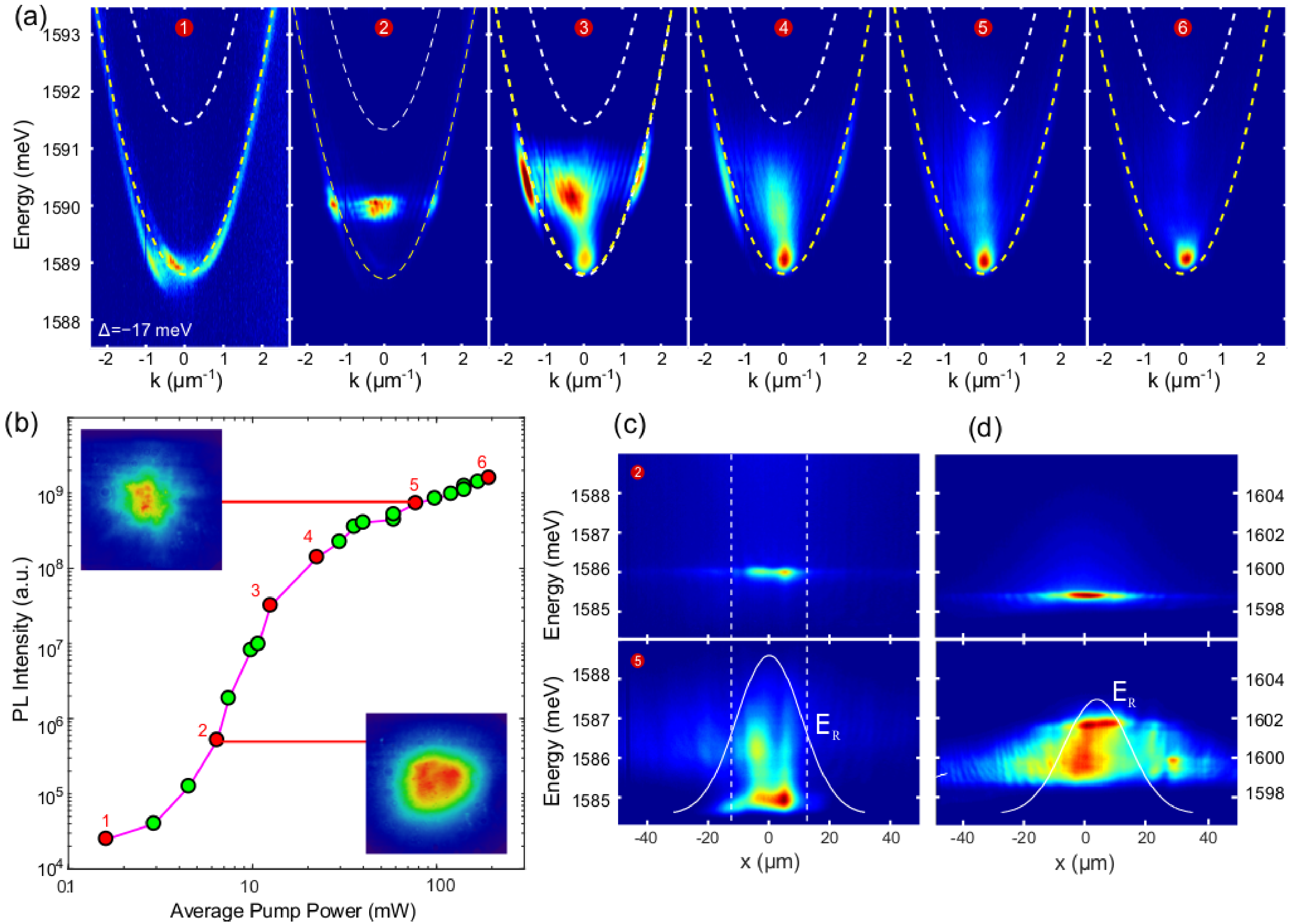
Condensates in inorganic semiconductors seem to be stable. However:

- Measurements of reservoir lifetime in GaAs samples suggest that parameters correspond to unstable regime (J. Bloch et al., 2006...)
- Number of emitted photons is typically not sufficient for single shot imaging, averaging over many shots would erase any fluctuations
- Long-range spatial coherence displays decay on a short length scale (Yamamoto et al, PNAS 2012,...)



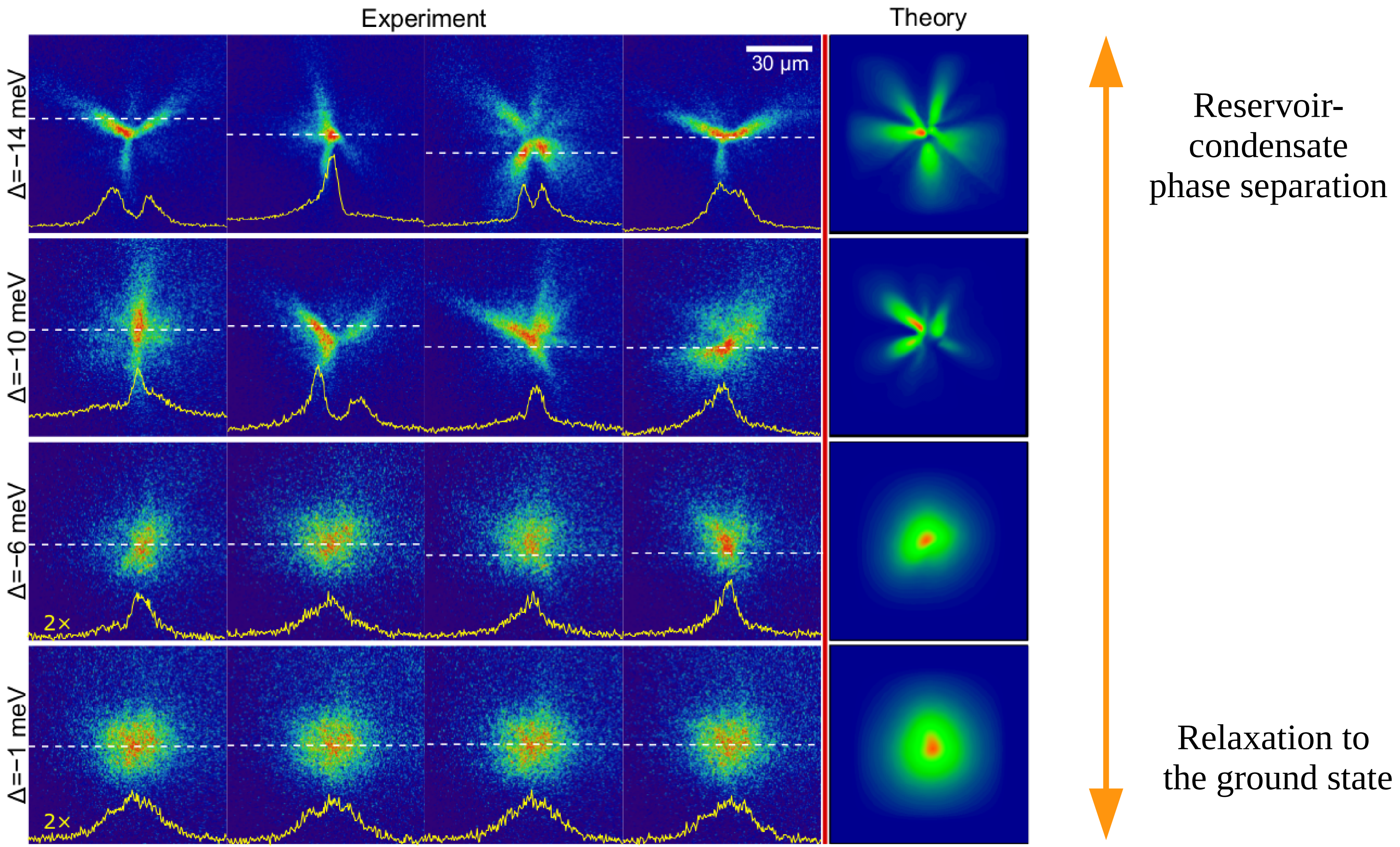
GaAs samples	Reservoir lifetime γ_R^{-1} [ps]
PHYSICAL REVIEW LETTERS 109, 216404 (2012)	300
PHYSICAL REVIEW B 88, 245307 (2013)	100
PHYSICAL REVIEW B 94, 045315 (2016)	10
PHYSICAL REVIEW LETTERS 109, 036404 (2012)	0.5
NATURE PHYSICS 7, 129 (2011)	2
PHYSICAL REVIEW B 86, 195313 (2012)	500
CdTe samples	
NATURE PHYSICS 4, 706 (2008)	0.4
PHYSICAL REVIEW LETTERS 103, 256402 (2009)	4 000
PHYSICAL REVIEW LETTERS 105, 120403 (2010)	900
PHYSICAL REVIEW LETTERS 106, 115301 (2011)	0.4
Organic samples	
NATURE PHOTONICS 4, 371 (2010)	1000
NATURE MATERIALS 13, 271 (2014)	570
NATURE MATERIALS 13, 247 (2014)	40

Long-lifetime polaritons in GaAs



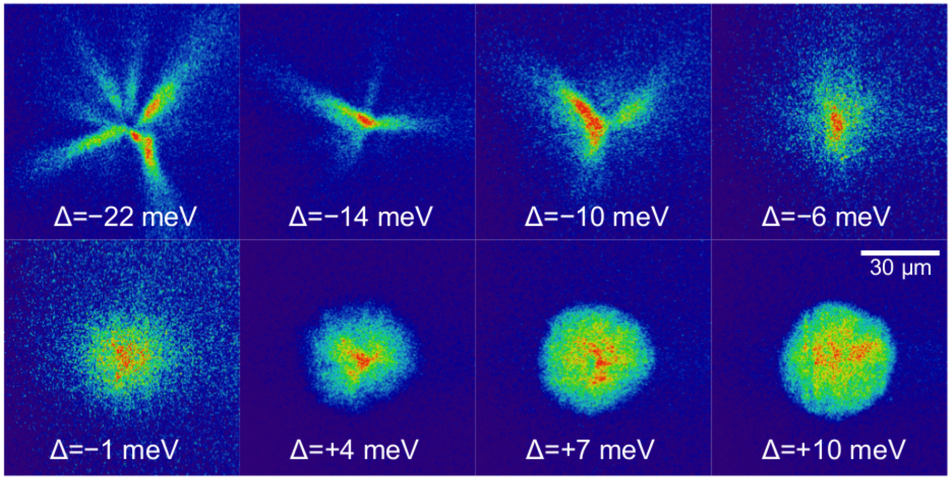
Experiment: Elena Ostrovskaya's ANU group lifetime: ~200 ps

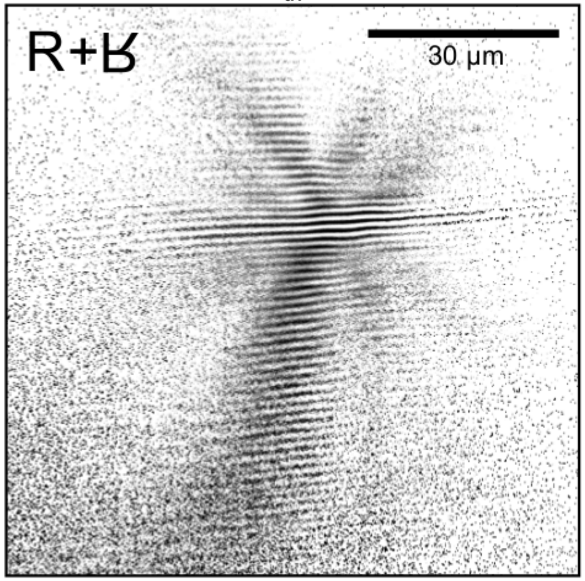
Long-lifetime polaritons in GaAs: single shot measurements

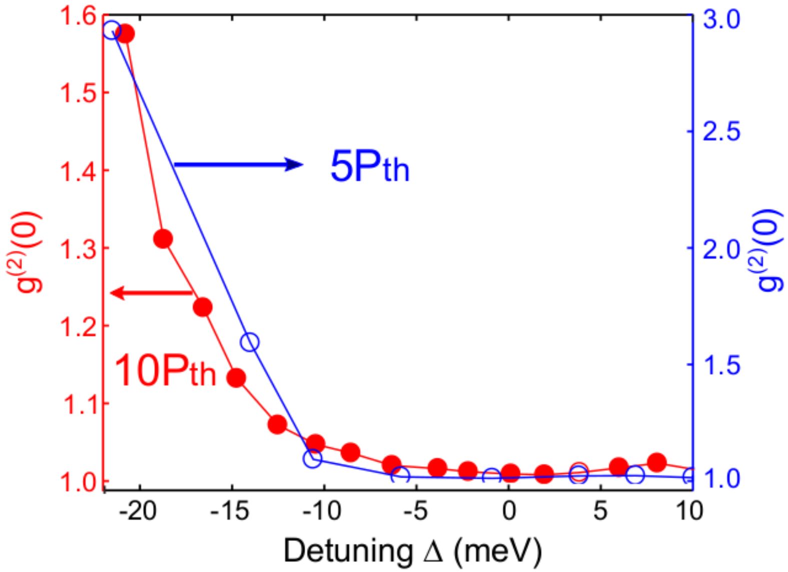


Two processes contribute to fragmentation: incomplete relaxation to the ground state and modulational instability leading to symmetry breaking and phase separation

Long-lifetime polaritons in GaAs: correlations







Fragmentation of the condensate is revealed in density-density correlations
Phase coherence is maintained across the condensate

Conclusions

- Instability due to effectively attractive interactions can occur in polariton condensates excited non-resonantly, due to the incoherent reservoir

- Excellent agreement with single-shot experiments in organic condensates suggests that the open-dissipative Gross-Pitaevskii equation is the right model to use

- Instability can be suppressed by energy relaxation in long-lifetime condensates

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