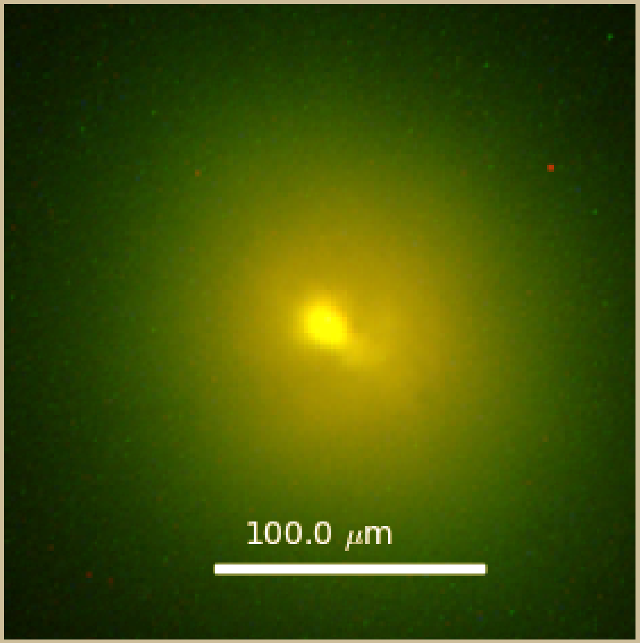
Bose-Einstein condensation of photons: dynamics and non-stationary statistics



Rob Nyman
Imperial College London

4th June 2019, KITP Santa Barbara
Open Quantum System Dynamics

What is a Bose-Einstein Condensate?

Macroscopic occupation of the quantum ground state at thermal equilibrium

- Bose-Einstein distribution: chemical potential μ

$$f(\epsilon) = \frac{1}{e^{(\epsilon - \mu)/k_B T} - 1}$$

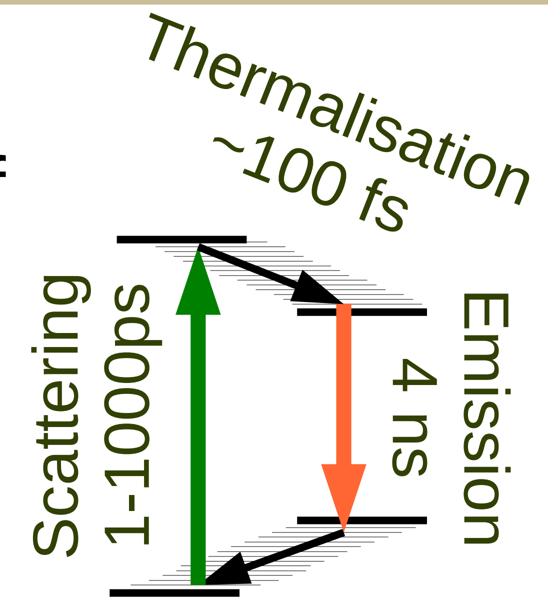
- Photons don't have a well defined μ
- Photons in a medium with a band gap do have μ

P Würfel, *J Phys C*, **15** 3967 (1982)

Photons in dye: giving light a chemical potential

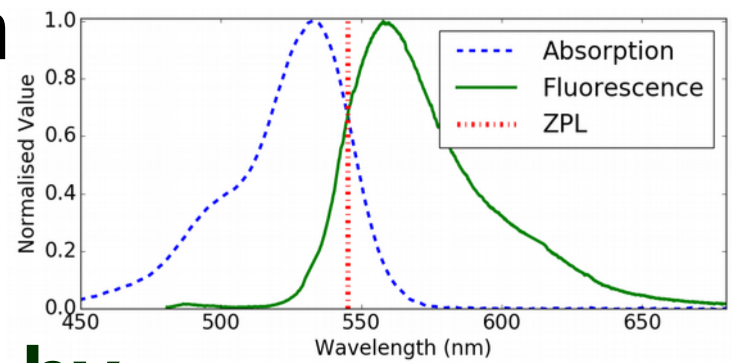
Optically pump laser dye

- Rhodamine 6G re-emits almost all of the light it absorbs



Photons excite an electron in a dye molecule

- Effective energy exchange between photon and solvent thermal bath
- Photons reach thermal equilibrium in picoseconds



Absorption and emission related by Kennard-Stepanov

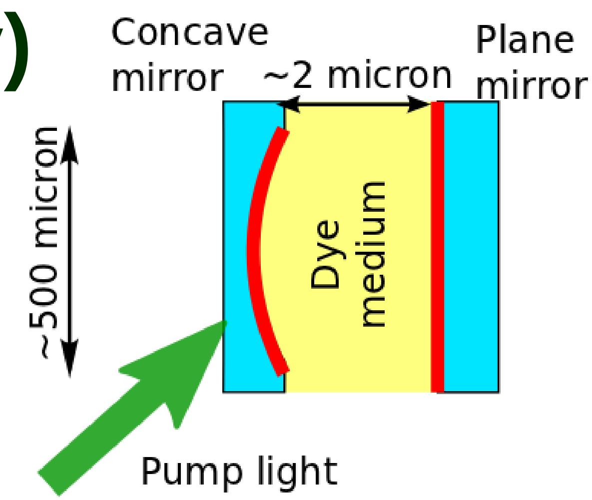
$$\mathrm{Abs}(E-E_0)=\mathrm{Fluo}(E-E_0)\,e^{(E-E_0)/k_BT}$$

Photons in a Microcavity

Mirrors trap photons long enough to reach thermal equilibrium

Free-spectral range larger than dye spectrum width (~1.5 μm-long cavity)

- Only 1 relevant longidutinal mode
- Photon dispersion relation like massive particle



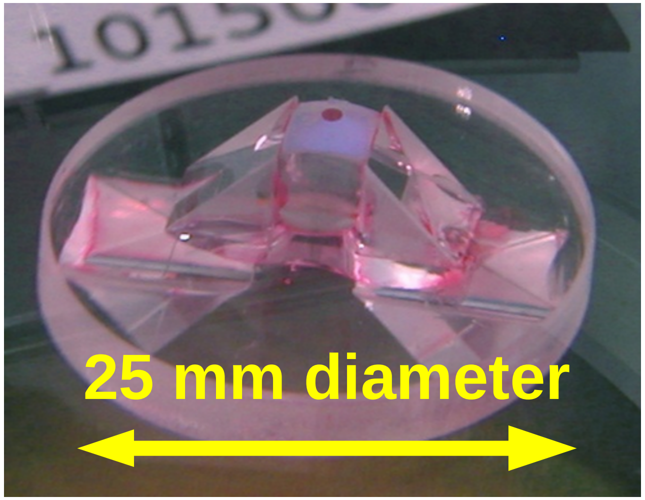
Curved mirror gives transverse modes like harmonic oscillator

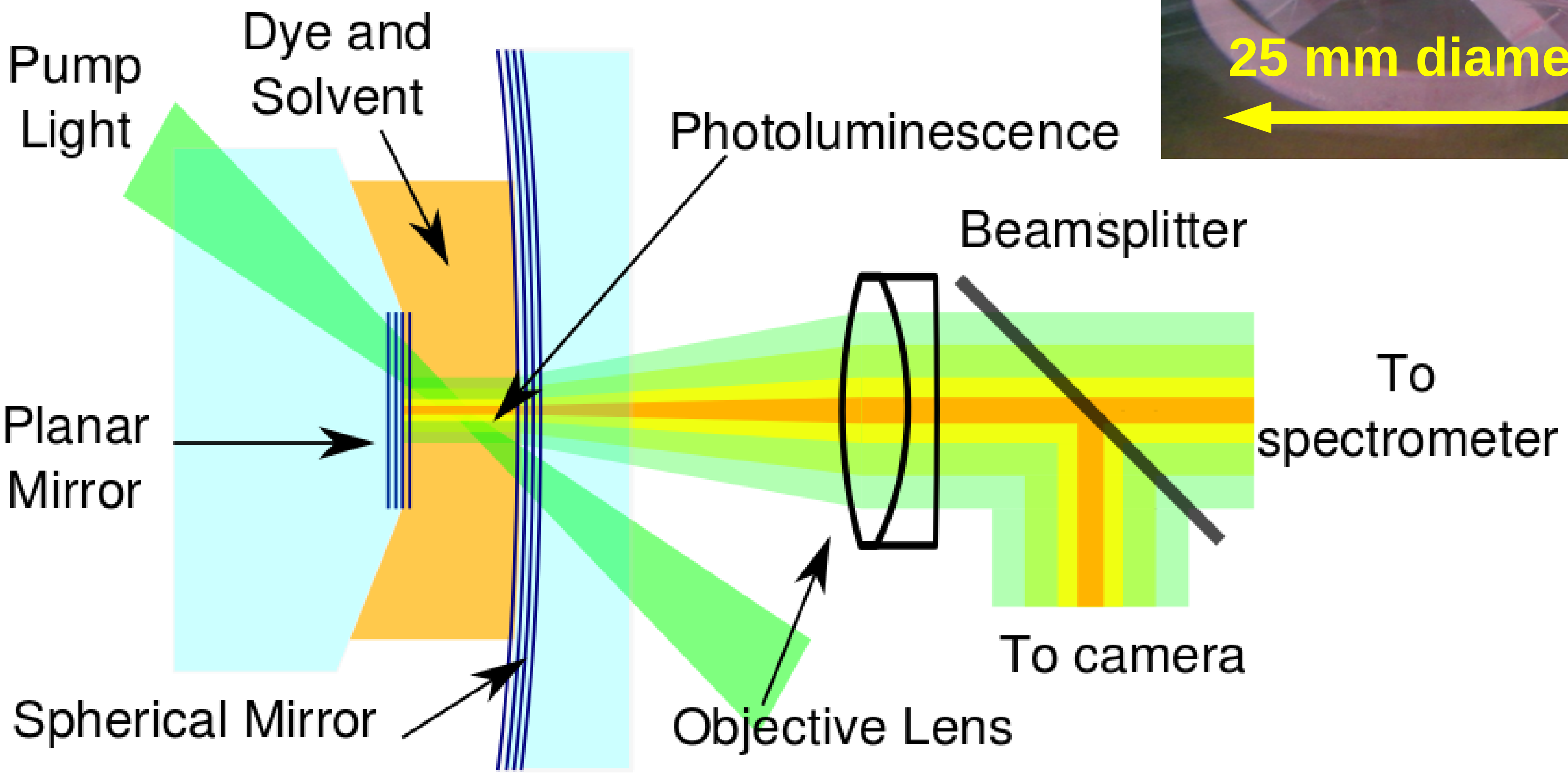
- Lowest transverse mode is ground state for thermal equilibrium

Observing the photons

Light leaks through the mirrors

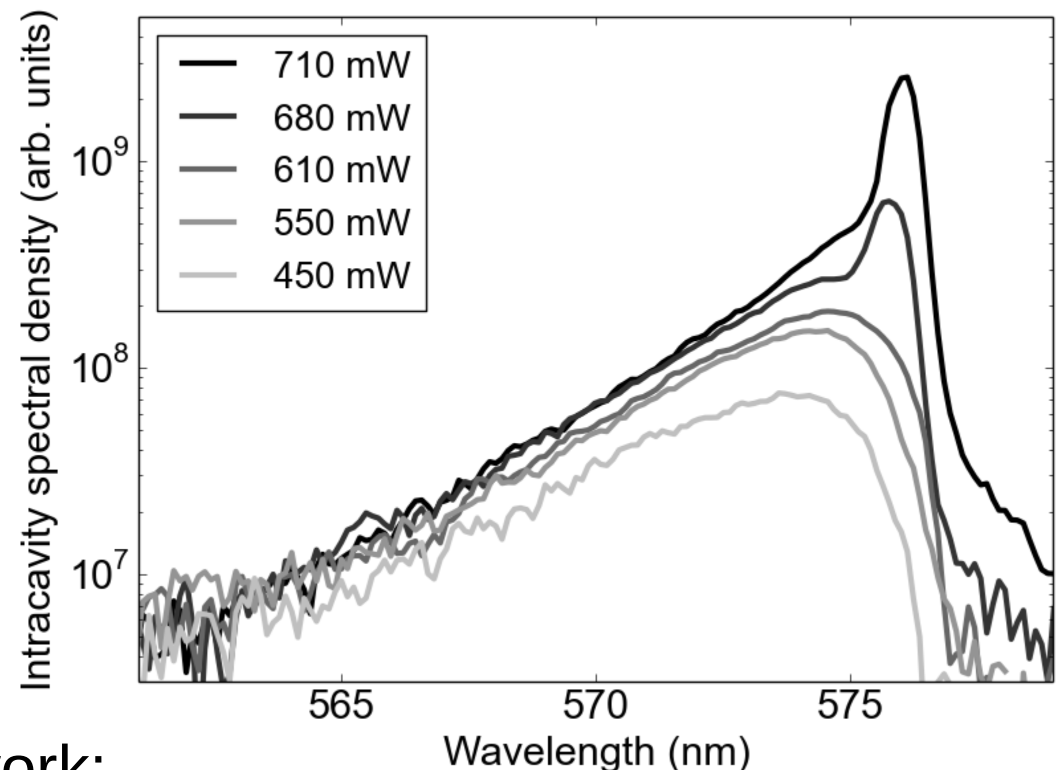
- Cavity finesse ~60 000 for nanosecond resonator lifetime

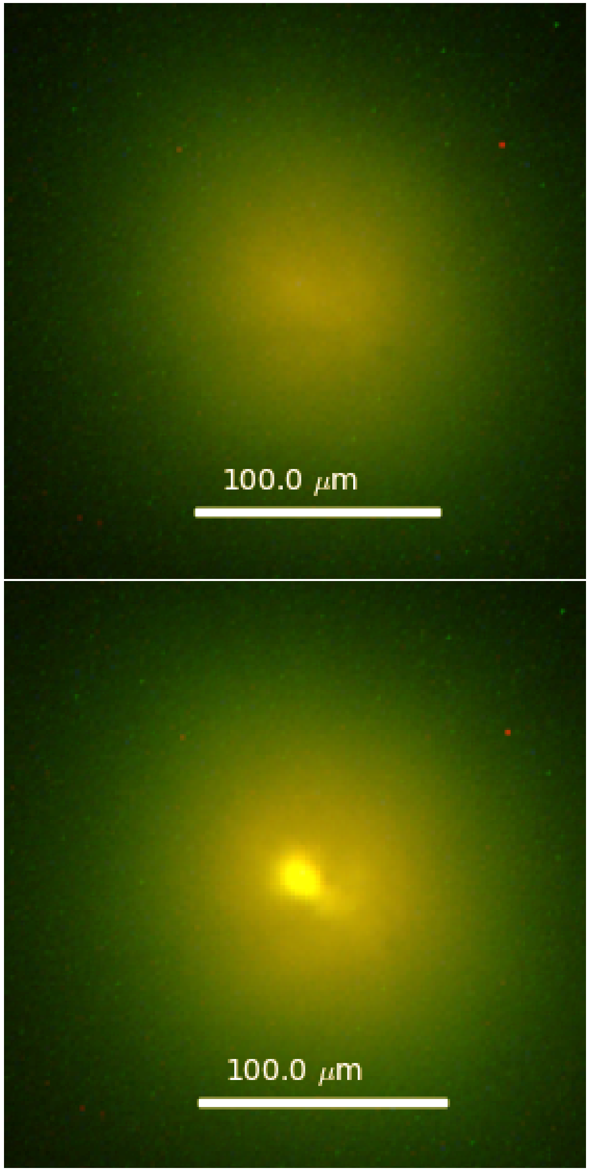




Thermal photons and BEC

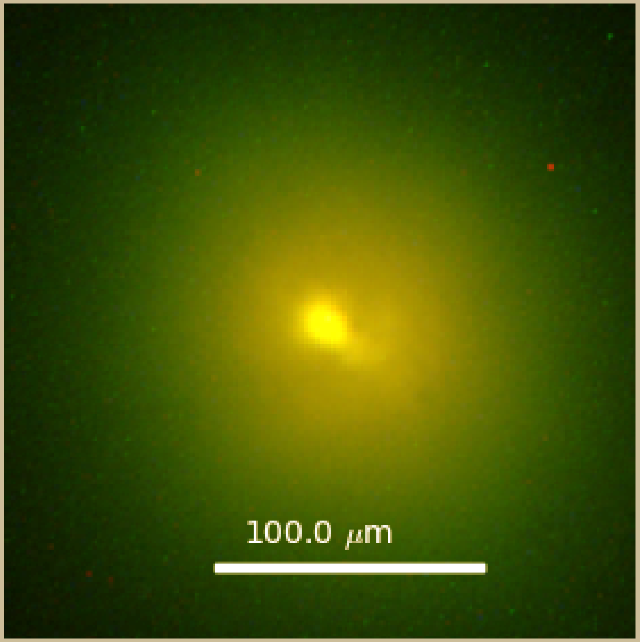
- Below threshold: thermal cloud
- Above: occupation of lowest-energy mode





Original work:
Klaers et al, *Nature* **468** p545 (2010)

Non-equilibrium behaviour



Non-equilibrium model of photon BEC

Model due to Kirton and Keeling

Keeling & Kirton, *PRA* **93**, 013829 (2016)

– Multi-mode/molecule Jaynes-Cummings hamiltonian with phonons

$$H = \sum_m \omega_m a_m^\dagger a_m + \sum_i \frac{\Delta}{2}\sigma_i^z + \Omega\left(b_i^\dagger b_i + \sqrt{S}\sigma_i^z(b_i + b_i^\dagger)\right) + g\sum_{m,i}(a_m\sigma_i^+ + a_m^\dagger\sigma_i^-)$$

Drive and dissipation

– cavity photon loss, pumping, fluorescence

$$\dot{\rho} = -i[H_0, \rho] - \sum_{i,m}\Big\{\frac{\kappa}{2}\mathcal{L}[a_m] + \frac{\Gamma_\uparrow}{2}\mathcal{L}[\sigma_i^+] + \frac{\Gamma_\downarrow}{2}\mathcal{L}[\sigma_i^-] + \frac{\Gamma(-\delta_m)}{2}\mathcal{L}[a_m^\dagger\sigma_i^-] + \frac{\Gamma(\delta_m)}{2}\mathcal{L}[a_m\sigma_i^+]\Big\}\rho.$$

Approximation: rate equation

$$\frac{\partial n_m}{\partial t} = -\kappa n_m + N\frac{\Gamma(-\delta_m)(n_m+1)\tilde{\Gamma}_\uparrow - \Gamma(\delta_m)n_m\tilde{\Gamma}_\downarrow}{\tilde{\Gamma}_\uparrow + \tilde{\Gamma}_\downarrow}$$

Extended to include inhomogeneities

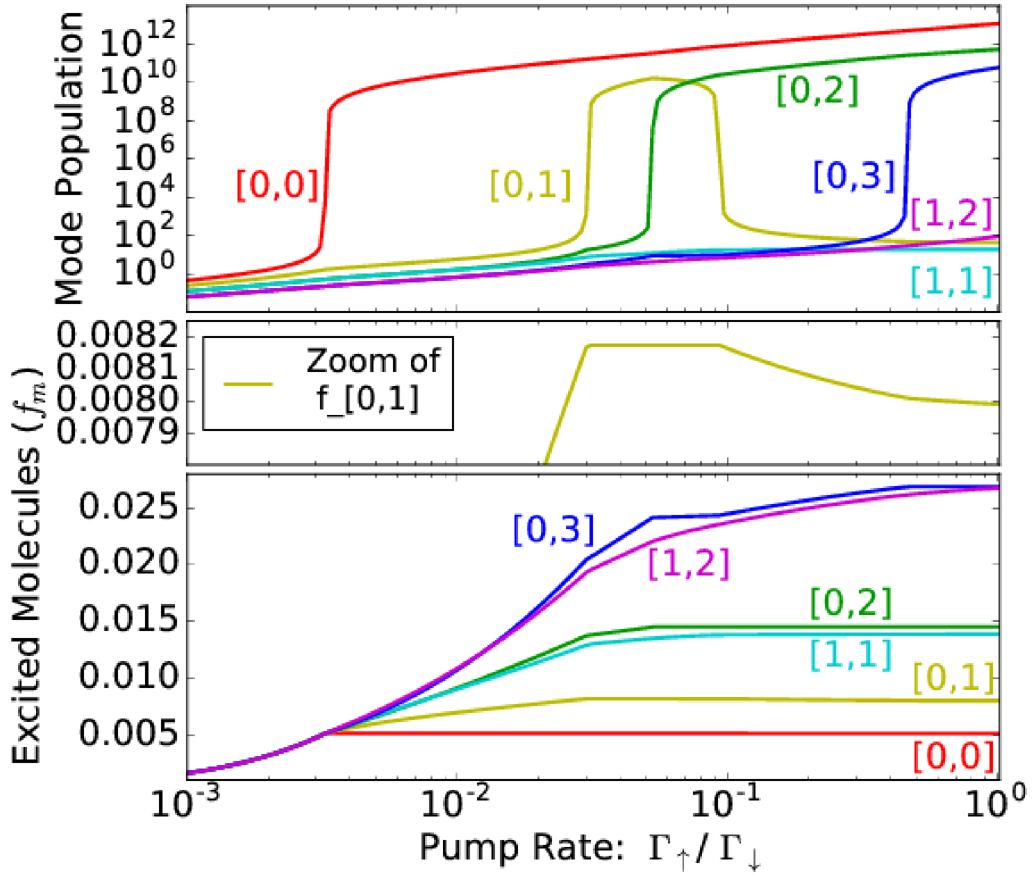
Non-equilibrium theory explains multimode behaviour

We have implemented Kirton and Keeling's inhomogeneous theory

– Multimode behaviour depends on rates of absorption vs loss

Decondensation

– A mode goes below threshold for increasing pump rate



Non-equilibrium phase diagram

HJ Hesten, RN, F Mintert, *PRL* 120 040601 (2018)

Condensate: re-absorption faster than cavity loss

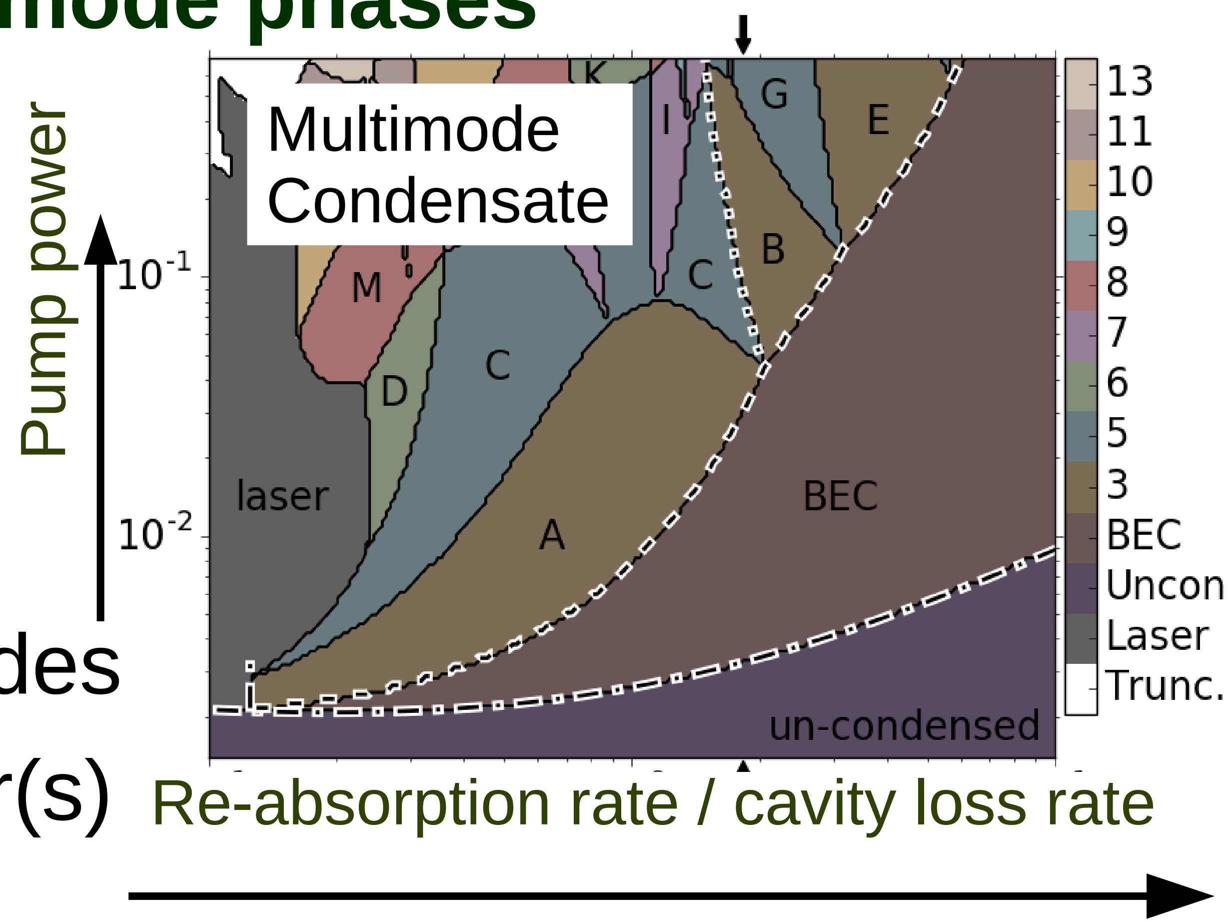
Laser: large occupation but not in lowest mode

Many possible multimode phases

- Similar to gain clamping in multimode lasers

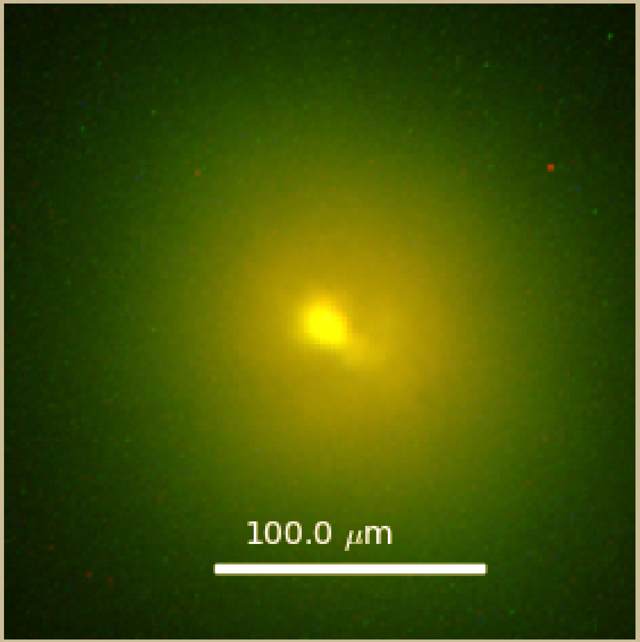
Concepts applicable to other systems

- Multiple boson modes
- Saturable reservoir(s)



Condensation of just a few photons

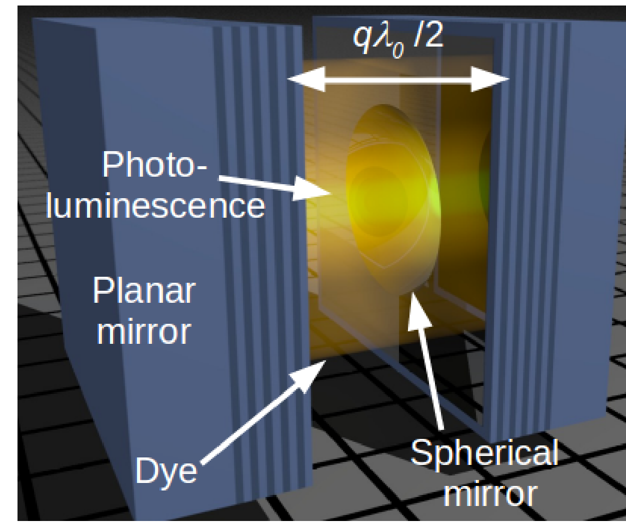
BT Walker et al (RN), *Nature Physics* 14 1173 (2018)



BEC with few photons

Threshold photon number (no spin degeneracy)

$$N_{th} = \frac{\pi^2}{6}\left(\frac{kT}{\hbar\omega}\right)^2 = \frac{nq\lambda}{12}\left(\frac{\pi kT}{\hbar c}\right)^2 \times RoC$$



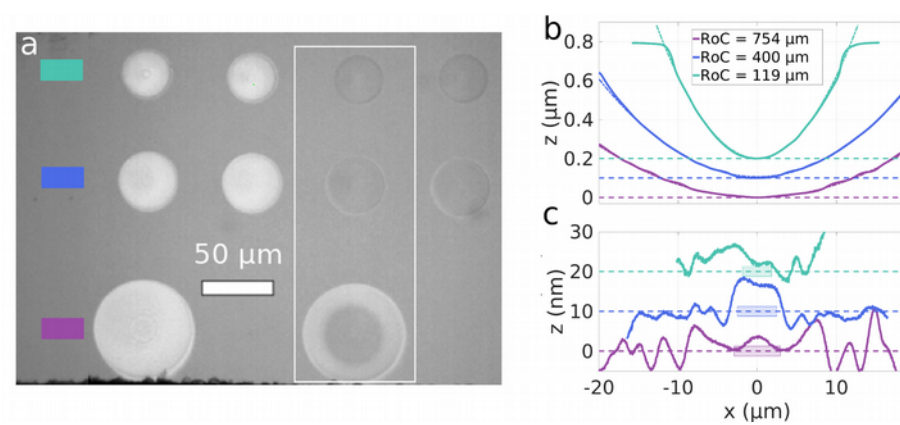
– Trapping potential depends on mirror curvature

– Previous experiments: $RoC \;\; 0.5\,\mathrm{m} \Rightarrow N_{th} \sim 50\,000$

– $RoC \;\; 400\,\mu\mathrm{m} \Rightarrow N_{th} \sim 40$

We have microfabricated mirrors for tiny BEC.

- Jason Smith group (Oxford)



Threshold behaviour for a small system

BEC: Basic equilibrium statistical mechanics

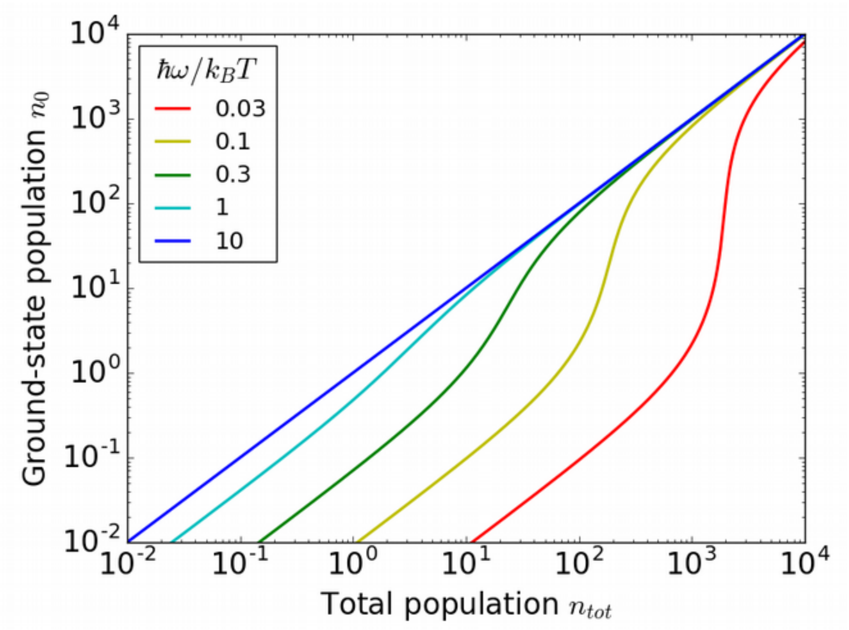
$$f(\epsilon_m|\mu) = \frac{1}{e^{(\epsilon_m - \mu)/k_B T} - 1}$$

$$n_{tot} = \sum_m f(\epsilon_m|\mu)\, g_m$$

2D Harmonic oscillator

$$g_m = m + 1 \quad ; \quad \epsilon_m = \epsilon_0 + m\hbar\omega$$

Condensation seen in ground-state population as function of total



Smallness when $\hbar\omega / kT \rightarrow \infty$

- Threshold becomes broad and shows small population jump

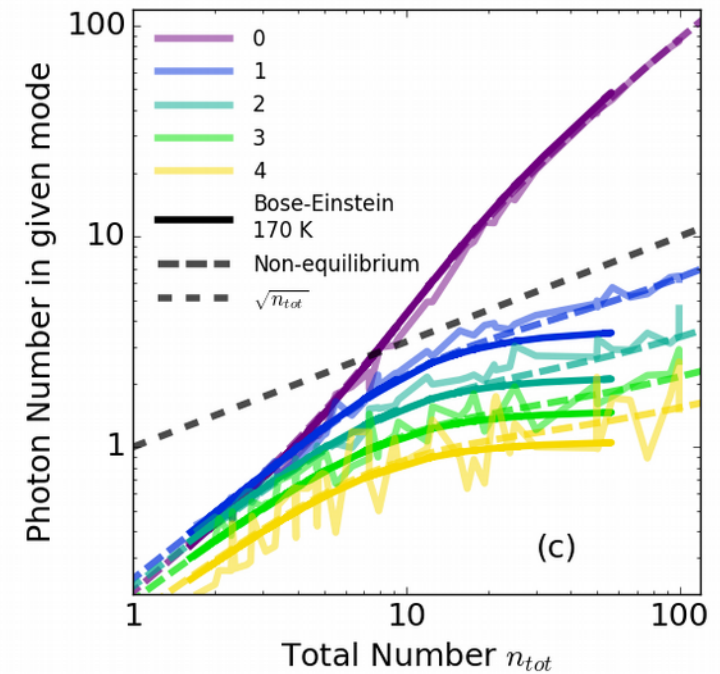
BEC phase transitions for a microscopic system

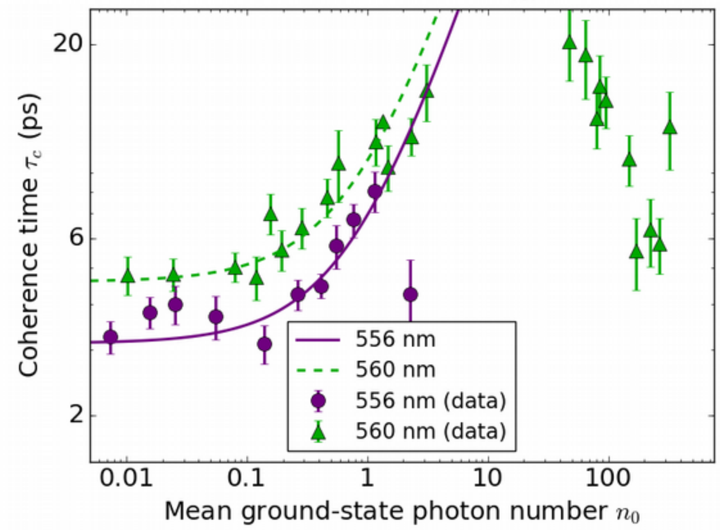
BEC: saturation of excited-state populations

- 7 ± 2 photons at phase transition (*not macroscopic!*)
- Temperature 150-170 K (*imperfect equilibrium!*)

Non-equilibrium model explains imperfect saturation

- Based on open-system light-matter interactions *(not totally coherent!)*

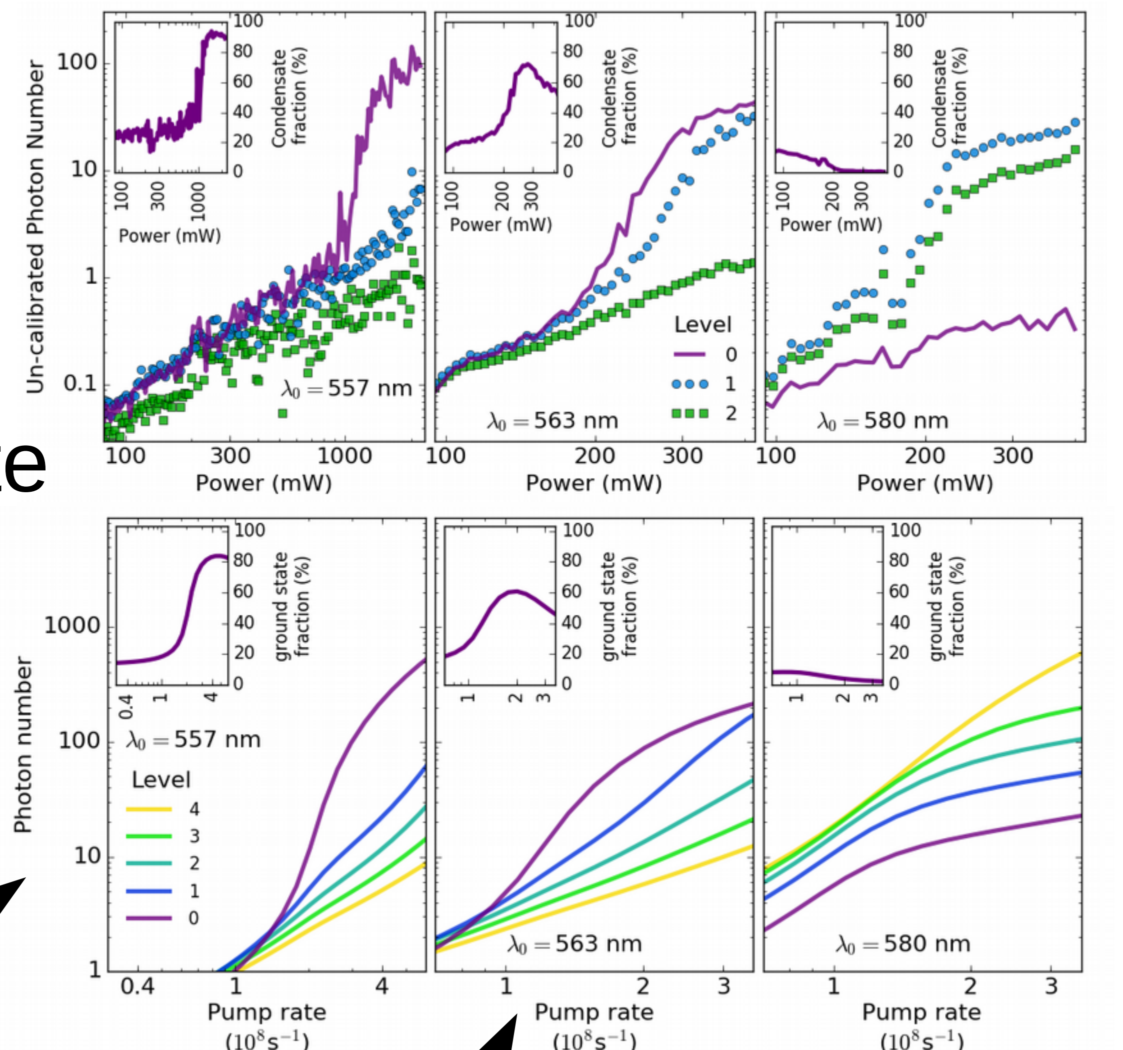




Multimode condensation

Thermalisation rate controlled by detuning from molecular resonance

- controls absorption rate
- 5 ps cavity lifetime



BEC (fast thermalisation)

Multimode condensate (some re-absorption)

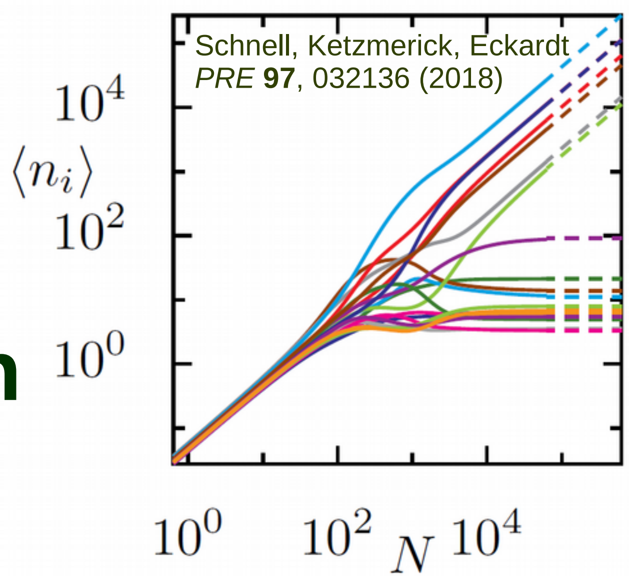
Laser (no re-absorption)

General threshold criteria: in theory

(Multimode) Condensation can occur in many systems

- driven Bose gases, evolutionary games, chemical kinetics, traffic jams
- Defined theoretically in the limit of infinite particle number
  - When a finite fraction of all particles go into some modes, but not others

J. Knebel *et al (Frey), Nat Comms* **6**, 6977 (2015)
D. Vorberg *et al* (Eckardt), *PRL* **111**, 240405 (2013)



Condensation is a phase transition

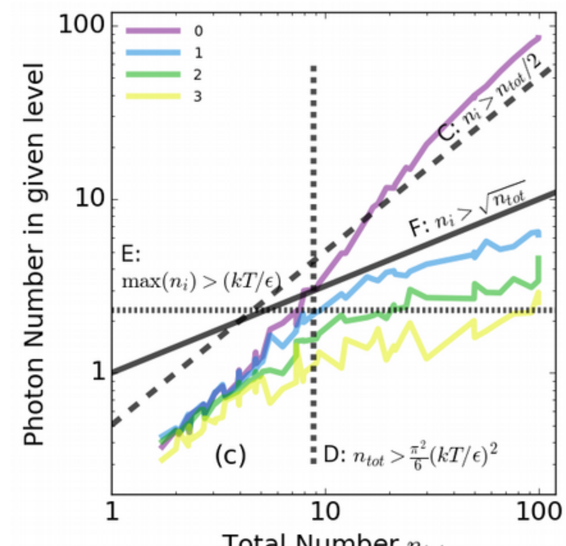
- Including laser and BEC

General threshold criteria: in experiments

Experiments need criteria applicable for particle finite number

$$N_{total} > \lim_{(N_{tot} \to \infty)} \{N_{exc}\}$$

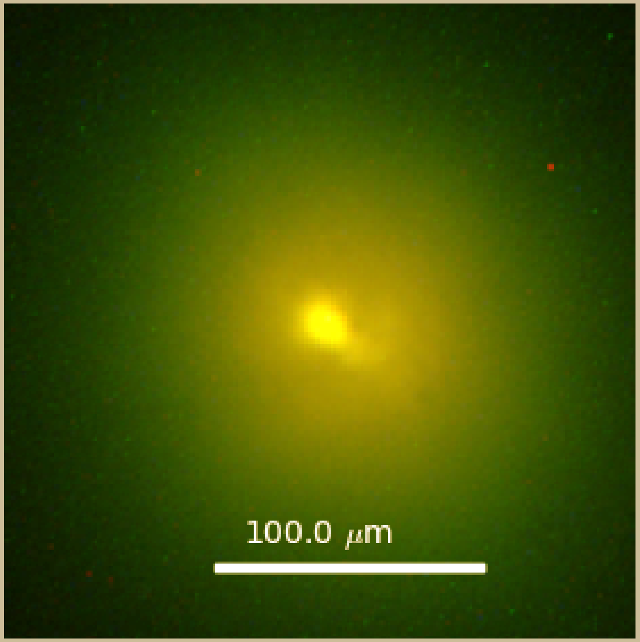
- Equilibrium definitions don’t apply out of equilibrium
- One-mode microlaser criterion (1 photon) unhelpful



More robust criteria

- $n_i > N_{total}/2$ (one condensate only)
- $max(n_i) > kT/\epsilon$ (quasi-eq^bm only)
- $n_i > N_{total}^{1/\alpha}$ with $\alpha \sim 1/2$
  - Separates condensed from depleted populations
  - Can describe multimode condensation

Dynamics and Non-stationary statistics



Critical & non-critical slowing down

HJ Hesten, BT Walker, RN, F Mintert, arXiv:1809.08772 and 1809.08774

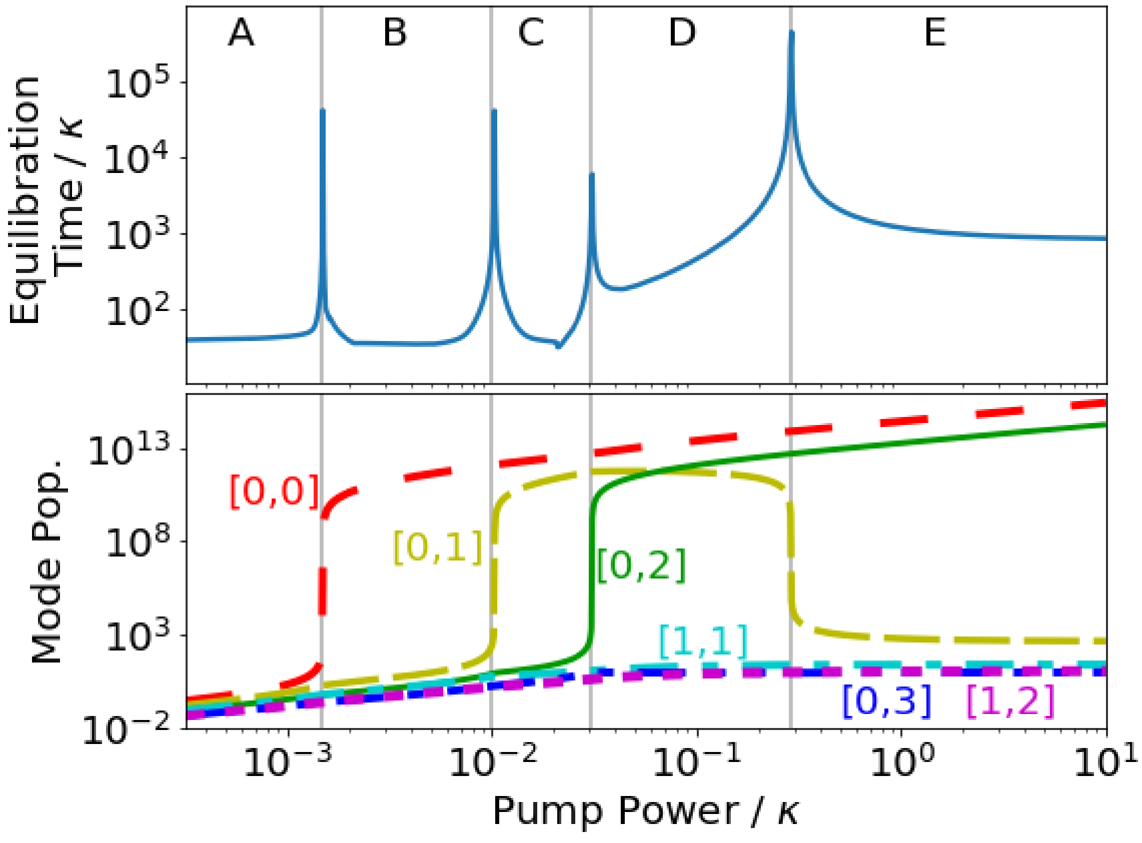
New numerical techniques for mean-field dynamics, not just steady state

Relaxation after pump rate quench is very slow near threshold

- Critical slowing down

Very slow relaxation in decondensed phase

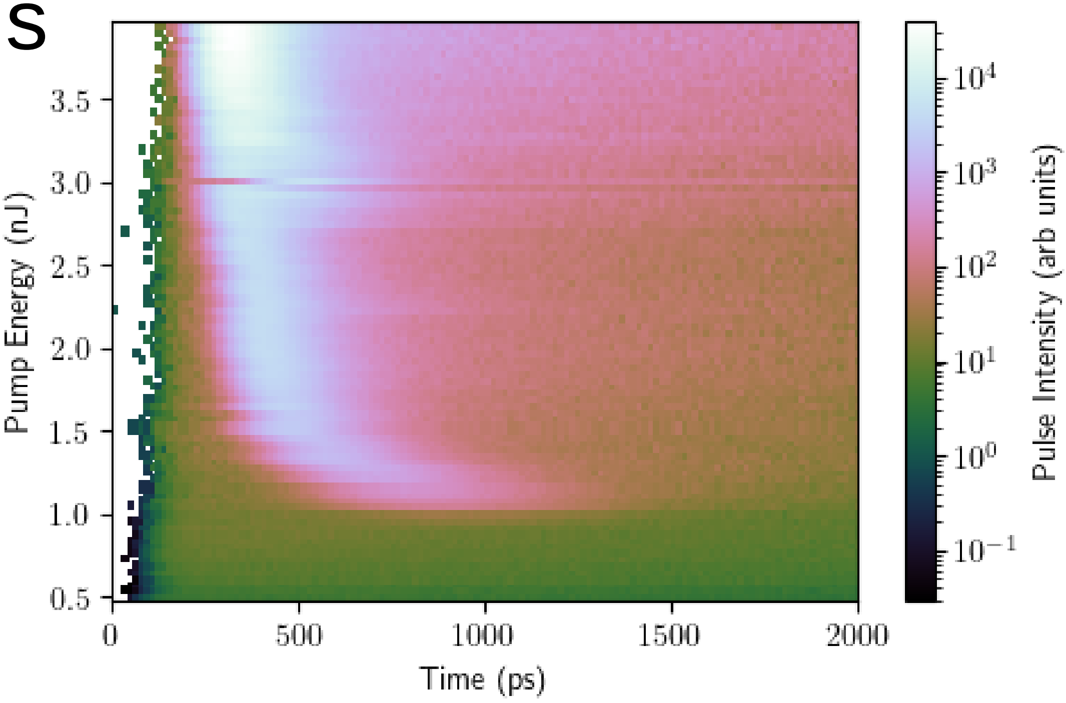
- A new non-critical slowing down phenomenon.



Dynamics (ensemble average)

Pulsed pumping, single-photon detection

- <40 ps timing resolution
- Average BEC formation as slow as 1000 ps
- Critical slowing down
- Timescale also depends on thermalisation rate



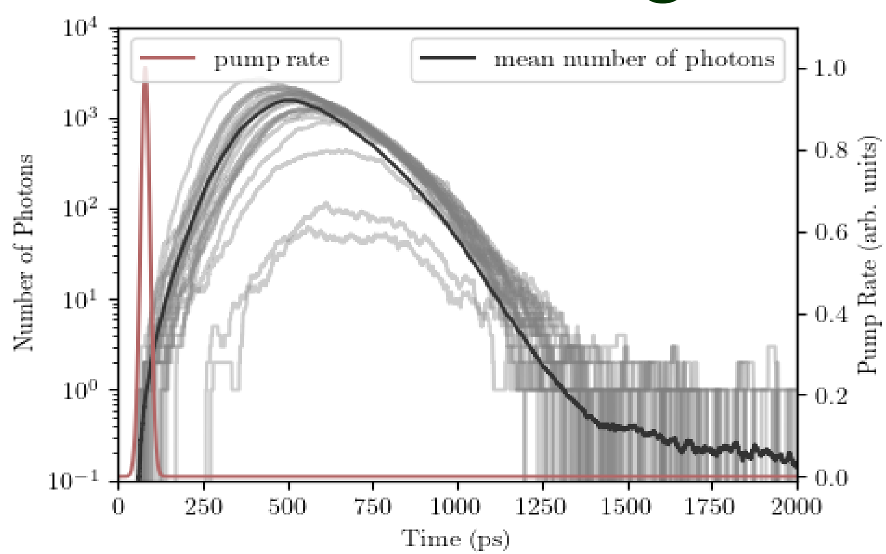
Beyond mean field: dynamics and fluctuations in BEC

BEC comes from stimulated scattering but must be seeded by spontaneous events

After quench, time for phase transition to occur depends on spontaneous events

- Slower close to threshold, with greater timing jitter

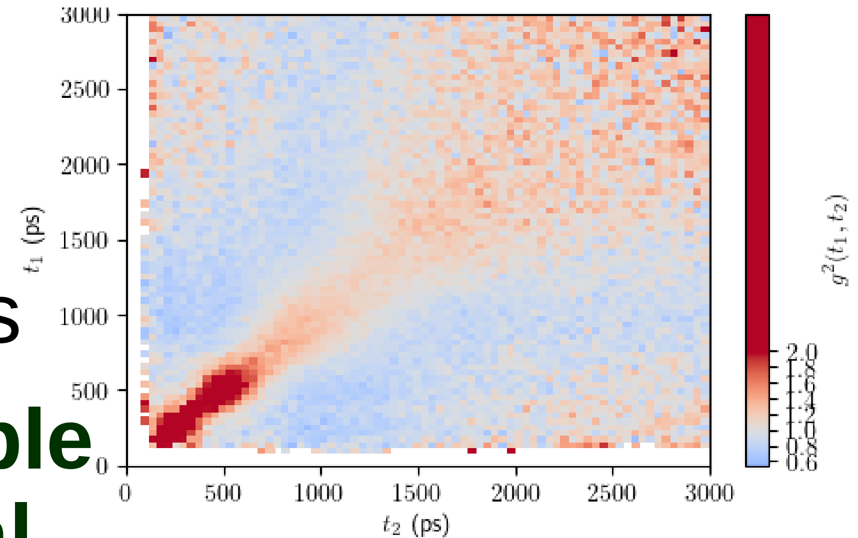
Monte Carlo simulation of single-mode system



Dynamics of two-time quantum correlations: experiments

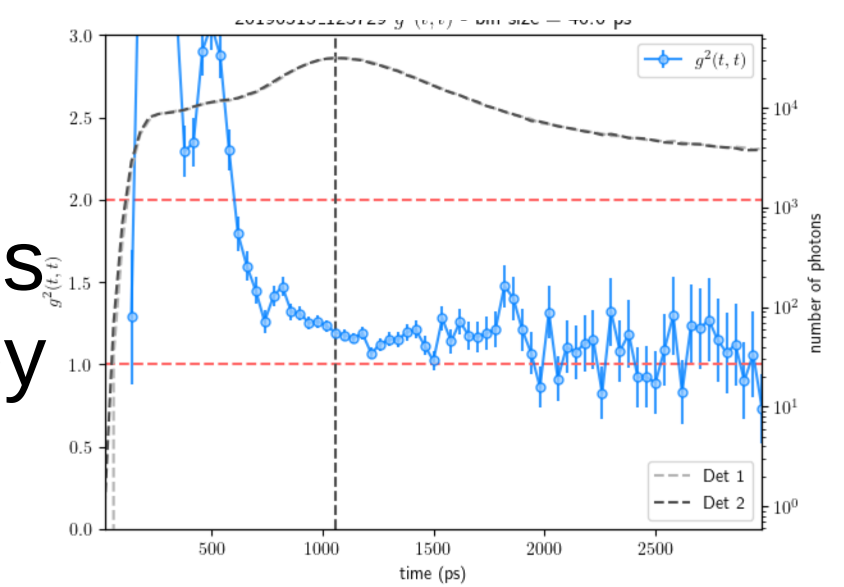
General 2-time correlation $g^{(2)}(t_1,t_2)$ shows formation jitter

- Anti-diagonal anti-correlations



Critical slowing down (ensemble average) and (one-time) critical fluctuations are linked

- Only true close to equilibrium
- Far from equilibrium, dynamics & fluctuations both revealed by non-stationary stats.



Dynamics of two-time quantum correlations: theory

Return to master equation

- Coupled rate equations including correlations

$$\frac{\partial n}{\partial t} \quad \frac{\partial m_e}{\partial t} \quad \frac{\partial \sigma_{nn}}{\partial t} \quad \frac{\partial \sigma_{mm}}{\partial t} \quad \frac{\partial \sigma_{nm}}{\partial t} \qquad \sigma_{xy} = \langle xy \rangle - \langle x \rangle \langle y \rangle$$

- Corrected ensemble-average dynamics, e.g.

$$\frac{\partial n}{\partial t} = -\kappa n + \Gamma(-\delta)\{(n+1)m_e + \sigma_{nm}\} - \Gamma(\delta)\{n(N - m_e) - \sigma_{nm}\}$$

- Zero-delay photon correlations $g_2(t,t) = 1 + \frac{\sigma_{nn}(t) - n(t)}{n(t)^2}$

2-time photon correlations via quantum regression

- Find expectations & $g^{(2)}(t_1,t_1)$, then evolve for $g^{(2)}(t_1,t_2)$

Conclusions

Rich phase diagram when thermal equilibrium breaks down

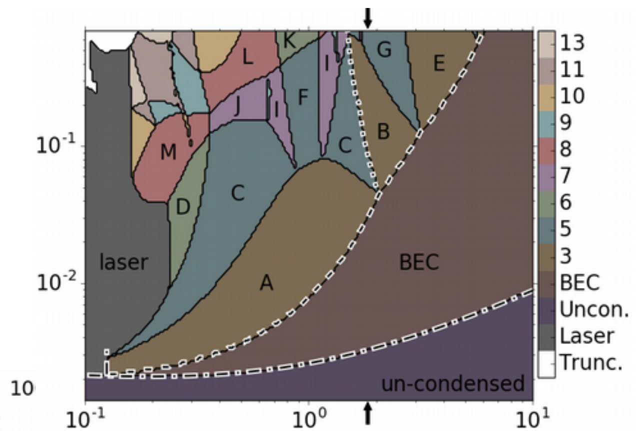
BEC phase transition extends to small numbers

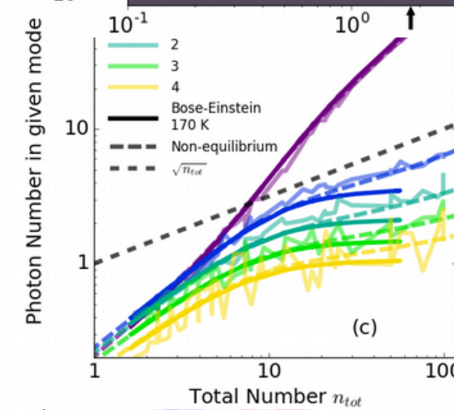
Non-stationary statistics

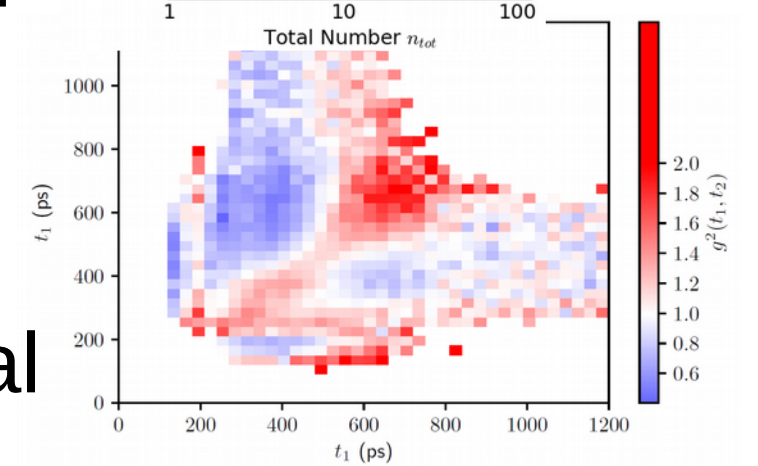
- Characterise dynamics and correlations, far from equilibrium

Next steps

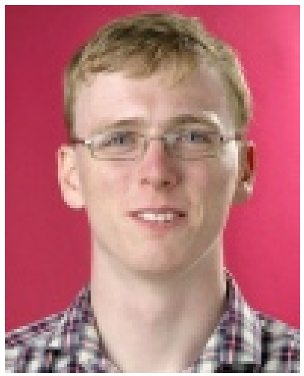
- Semiconductor photon BEC
- Replace cavity with metamaterial

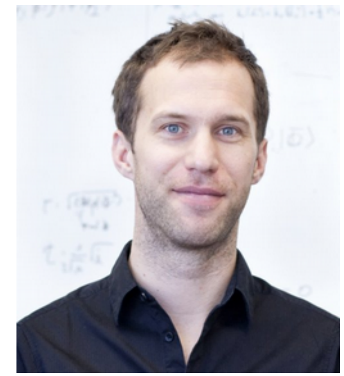






The Photon BEC team

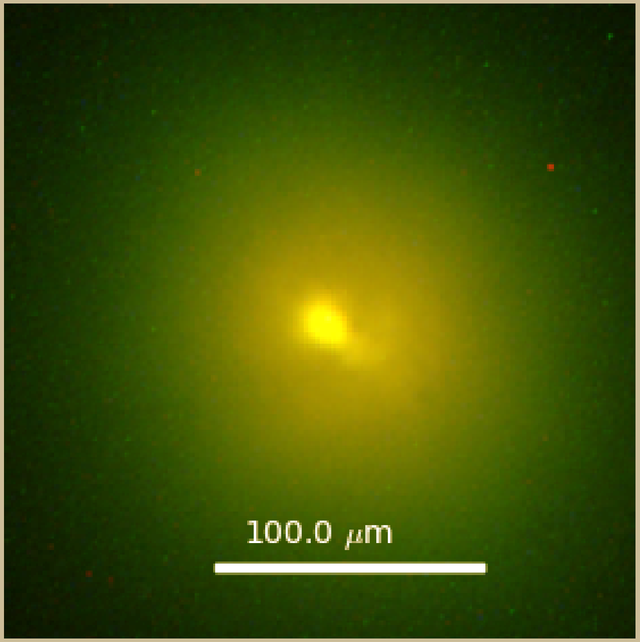

Ben Walker	Henry Hesten	João Rodrigues	Himadri Dhar	Florian Mintert	Rupert Oulton
PhD Student	PhD Student	Post-doc	Post-doc	Faculty	Faculty
Experiment	Theory	Experiment	Theory	Theory	Experiment

Former PhD student: Jakov Marelic (2013-2017)
Current Collaborators: Aurelien Trichet, Jason Smith (Oxford); Ed Clark (Sheffield); Iwan Moreels (Ghent); Robin Kaiser, Stephane Barland, Gian-Luca Lippi (Nice)

EPSRC
Engineering and Physical Sciences Research Council

PhoQuS
Photons for Quantum Simulation

BONUS MATERIAL



2-time correlations: theory

Master equation

The interaction between a single photon mode and the molecules in the cavity result in a Master equation (ME) given by:

$$\frac{d\rho}{dt} = -i[\tilde{H},\rho] + \kappa\mathcal{L}(\hat{a})\rho + \sum_i \Big\{ \Gamma_\downarrow\, \mathcal{L}(\sigma_i^-) + \Gamma_\uparrow\, \mathcal{L}(\sigma_i^+) + \Gamma(\delta)\, \mathcal{L}(\hat{a}\,\sigma_i^+) + \Gamma(-\delta)\, \mathcal{L}(\hat{a}^\dagger\, \sigma_i^-) \Big\} \rho, \quad (1)$$

where, $\mathcal{L}(\hat{K})\rho = \hat{K}\rho\hat{K}^\dagger - \frac{1}{2}\left\{\hat{K}^\dagger\hat{K},\rho\right\}$. The ME can also be used to derive the rate equations for the photon number ($n = \langle \hat{a}^\dagger \hat{a}\rangle$) and the molecular excitation ($m_e = \sum_i \langle \sigma_i^+ \sigma_i^-\rangle$).

$$\frac{\partial n}{\partial t} = -\kappa n + \Gamma(-\delta)\{m_e + \underbrace{\sum_i \langle \hat{a}^\dagger \hat{a}\, \sigma_i^+ \sigma_i^-\rangle}_{nm_e}\} - \Gamma(\delta)\{nN - \underbrace{\sum_i \langle \hat{a}^\dagger \hat{a}\, \sigma_i^+ \sigma_i^-\rangle}_{nm_e}\} \quad (2)$$

$$\frac{\partial m_e}{\partial t} = -\Gamma_\downarrow m_e - \Gamma(-\delta)\{m_e + \overbrace{\sum_i \langle \hat{a}^\dagger \hat{a}\, \sigma_i^+ \sigma_i^-\rangle}\} + \Gamma(\delta)\{nN - \overbrace{\sum_i \langle \hat{a}^\dagger \hat{a}\, \sigma_i^+ \sigma_i^-\rangle}\} + \Gamma_\uparrow(N - m_e). \quad (3)$$

To obtain a closed solution, one can ignore all correlations between the photon and molecules and make the first-order (semiclassical) approximation: $\sum_i \langle \hat{a}^\dagger \hat{a}\, \sigma_i^+ \sigma_i^-\rangle \approx \langle \hat{a}^\dagger \hat{a}\rangle \sum_i \langle \sigma_i^+ \sigma_i^-\rangle = nm_e$.

2-time correlations: theory

Second-order rate equations

$$\frac{\partial n}{\partial t} = -\kappa n + \Gamma(-\delta)\{(n+1)m_e + \sigma_{nm}\} - \Gamma(\delta)\{n(N-m_e) - \sigma_{nm}\} \quad (4)$$

$$\frac{\partial m_e}{\partial t} = -\Gamma_{\downarrow} m_e - \Gamma(-\delta)\{(n+1)m_e + \sigma_{nm}\} + \Gamma(\delta)\{n(N-m_e) - \sigma_{nm}\} + \Gamma_{\uparrow}(N-m_e) \quad (5)$$

where, $\sigma_{xy} = \langle xy \rangle - \langle x \rangle \langle y \rangle$ is the covariance function. The covariance functions here provide the second-order corrections to the semiclassical rate equations.

$$\begin{aligned}
\frac{\partial \sigma_{nn}}{\partial t} &= -\kappa(n + 2\sigma_{nn}) + \Gamma(-\delta)\{(n+1)m_e + 2\sigma_{nn}m_e + \sigma_{nm}(2n+1)\} \\
&- \Gamma(\delta)\{n(N-m_e) + 2\sigma_{nn}(N-m_e) - \sigma_{nm}(2n_e - 1)\} \quad (6) \\
\frac{\partial \sigma_{mm}}{\partial t} &= -\Gamma_{\downarrow}(m_e + 2\sigma_{mm}) - \Gamma(-\delta)\{-(n+1)m_e + 2\sigma_{mm}(n+1) + \sigma_{nm}(2m_e - 1)\} \\
&+ \Gamma(\delta)\{n(N-m_e) - 2\sigma_{mm}n + \sigma_{nm}(-2m_e + 2N - 1)\} + \Gamma_{\uparrow}(N - m_e - 2\sigma_{mm}) \quad (7) \\
\frac{\partial \sigma_{nm}}{\partial t} &= -(\kappa + \Gamma_{\downarrow} + \Gamma_{\uparrow})\sigma_{nm} + \Gamma(-\delta)\{(n+1)(-m_e + \sigma_{mm}) - \sigma_{nn}m_e + \sigma_{mn}(m_e - n - 2)\} \\
&+ \Gamma(\delta)\{-n(N-m_e) + \sigma_{mm}n + \sigma_{nn}(N-m_e) + \sigma_{nm}(m_e - n + 1 - N)\} \quad (8)
\end{aligned}$$

Now, the second-order correlation function, with zero time-delay, can be calculated using the above set of equations, using the relation:

$$g_2(t,t) = 1 + \frac{\sigma_{nn}(t) - n(t)}{n(t)^2} \quad (9)$$

2-time correlations: theory

Two-time second order correlation function

The two-time second-order correlation function is given by,

$$g_2(t_1, t_2) = \frac{\langle \hat{a}^\dagger(t_1)\hat{a}^\dagger(t_2)\hat{a}(t_2)\hat{a}(t_1)\rangle}{\langle \hat{a}^\dagger(t_1)\hat{a}(t_1)\rangle \, \langle \hat{a}^\dagger(t_2)\hat{a}(t_2)\rangle}. \quad (10)$$

To solve this we make use of the quantum regression theorem, which allows us to map the problem to two separate time evolutions. First, we use the second-order rate equation to evolve the system ρ from $t = 0$ to t_1. Thus, we obtain the expectation values of $n(t)$, $m_e(t)$, $\sigma_{nn}(t)$, $\sigma_{mm}(t)$, $\sigma_{nm}(t)$ and $g_2(t, t)$, using Eqs.(4-9). Second, at $t = t_1$, we define a new state

$$\tilde{\rho} = \frac{\hat{a}(t_1)\rho\hat{a}^\dagger(t_1)}{\langle \hat{a}^\dagger(t_1)\hat{a}(t_1)\rangle}, \quad (11)$$

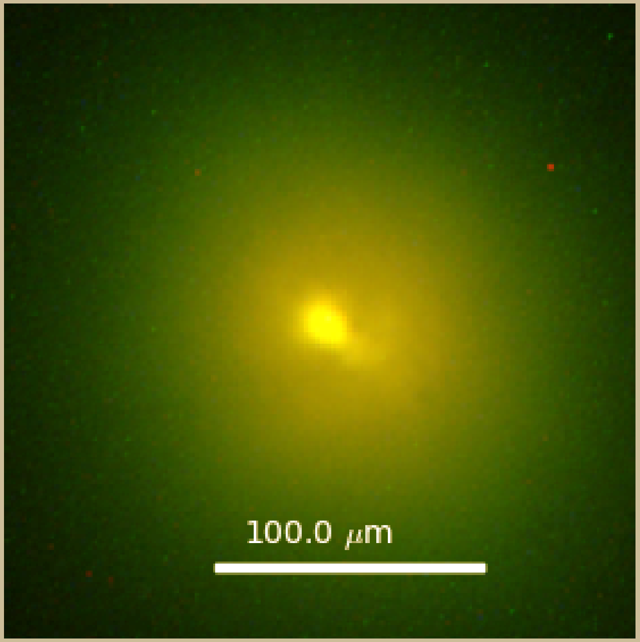
and use the first-order rate equations to solve for the expectation value $\langle\langle n(t)\rangle\rangle$ and $\langle\langle m_e(t)\rangle\rangle$, from $t = t_1$ to t_2, where $\langle\langle \hat{X}\rangle\rangle = \mathrm{Tr}[\hat{X}\tilde{\rho}]$. So we obtain at time $t = t_2$,

$$\langle\langle n(t_2)\rangle\rangle = \mathrm{Tr}[\hat{a}^\dagger(t_2)\hat{a}(t_2)\tilde{\rho}] = \frac{\langle \hat{a}^\dagger(t_1)\hat{a}^\dagger(t_2)\hat{a}(t_2)\hat{a}(t_1)\rangle}{\langle \hat{a}^\dagger(t_1)\hat{a}(t_1)\rangle} = g_2(t_1, t_2) \times \langle \hat{a}^\dagger(t_2)\hat{a}(t_2)\rangle. \quad (12)$$

The initial values for $\langle\langle n(t)\rangle\rangle$ and $\langle\langle m_e(t)\rangle\rangle$ at $t = t_1$ are known from the first time evolution,

$$\langle\langle n(t_1)\rangle\rangle = g_2(t_1, t_1)n(t_1), \quad \text{and} \quad \langle\langle m_e(t)\rangle\rangle = \frac{\sigma_{mn}(t_1) - n(t_1)m_e(t_1)}{n(t_1)}. \quad (13)$$

UNUSED SLIDES



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http://www.oxfordhighq.com/

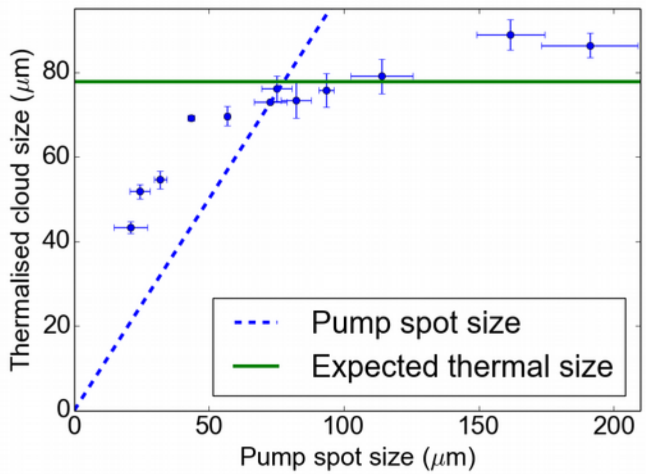
- Core technology: open optical microcavities
  - Spunout from Oxford Uni. Materials Dept. July 2018
  - High sensitivity over small sample volumes (≈fL)
- Nanoparticle analysis
  - Cavity resonance shift proportional to particle polarisability
  - Size determined from dynamics
- Chemical analysis
  - Absorption spectroscopy
  - Sensitivity to just a few 10's of molecules
- Open microcavity fabrication
  - Nanometric topographic control of non-conductive materials
  - Cavity finesse > 10 000 after coating
  - Custom shapes can be supplied

Inhomogeneous pumping

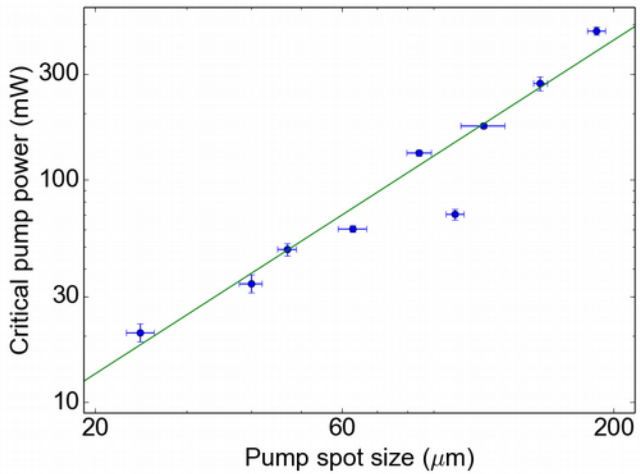
J. Marelic and R.A. Nyman, *PRA* **91,** 033813 **(2015)**

Strong dependences with pump-spot size

Below threshold, cloud size varies with pump-spot size



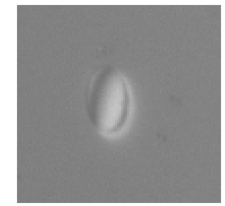
Threshold pump power varies with pump-spot size

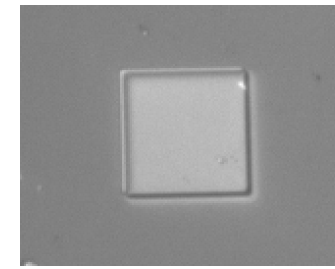


- Size at equilibrium function of temperature and confinement: observations disagree
- Threshold at equilibrium independent of pump spot: observations disagree
- Theory with inhomogeneities explains out data

Designing trapping potentials through mirror shapes

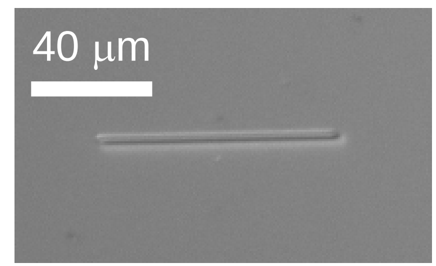
Anisotropic shapes

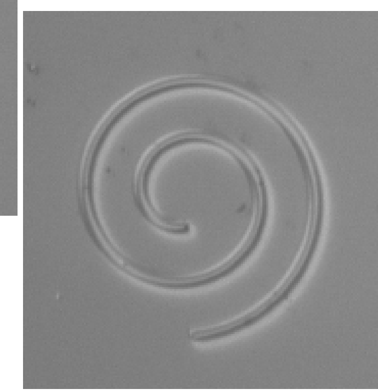




– For measuring interactions

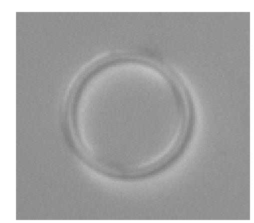
1D geometries

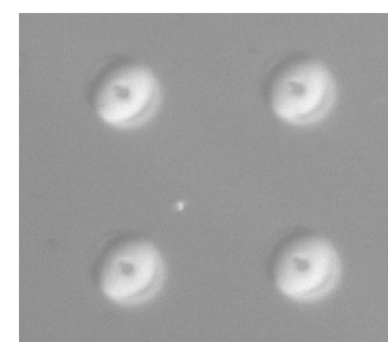




– Transport, low-D Bose gases

Non-trivial topology





– Mexican-hat potentials and 1D rings

These samples await mirror coating

Threshold behaviour: BEC vs microlaser

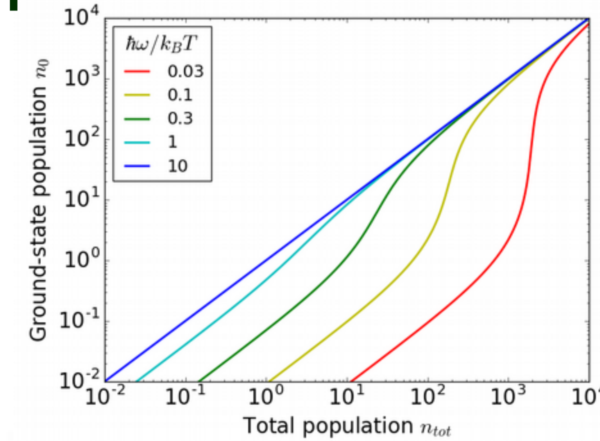
BEC: Basic equilibrium statistical mechanics

$$f(\epsilon_m|\mu) = \frac{1}{e^{(\epsilon_m - \mu)/k_B T} - 1} \qquad n_{tot} = \sum_m f(\epsilon_m|\mu)\, g_m$$

2D Harmonic oscillator

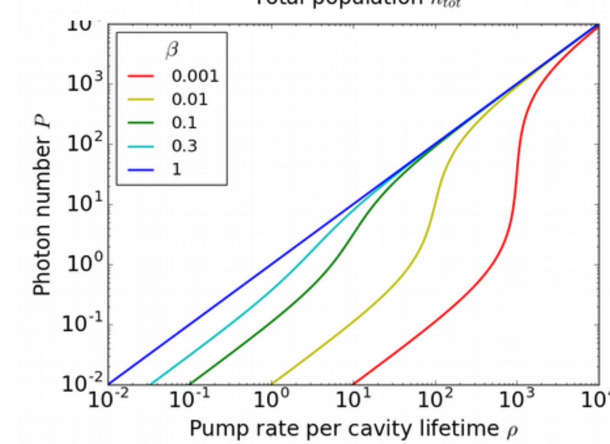
$$g_m = m+1 \;\; ; \;\; \epsilon_m = \epsilon_0 + m\hbar\omega$$

Condensation seen in ground-state population



Microlaser

- Fraction β of spontaneous emission into cavity
- Simplified rate-equation model



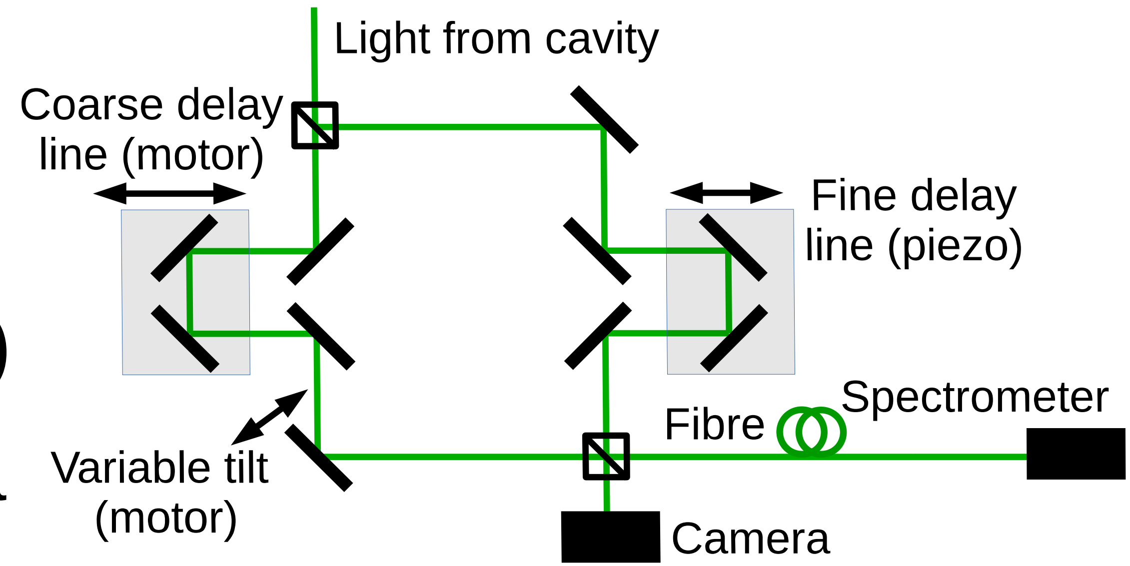
Smallness: $\beta \to 1$ vs. $\hbar\omega / kT \to \infty$

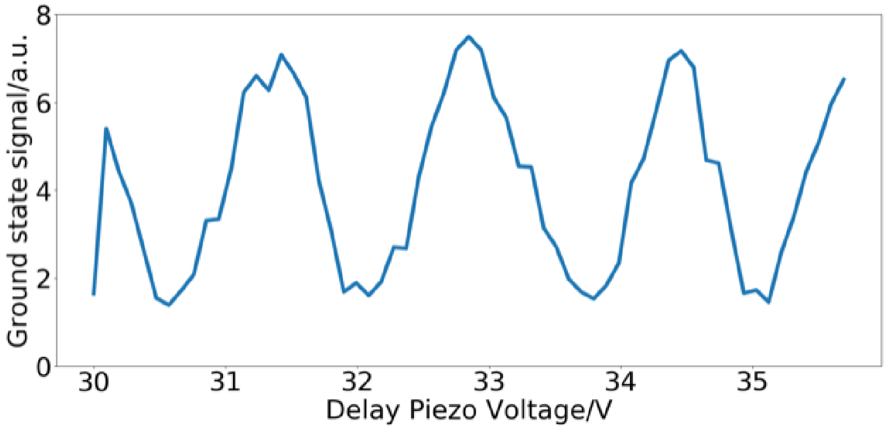
- Threshold becomes broad and shows small population jump

Coherence

Mach-Zehnder interferometer

- Shift images in time and space: $g^{(1)}(\mathbf{r},\mathbf{r}',t-t')$
- Spectrometer for data acquisition
- Can resolve individual cavity modes if needed



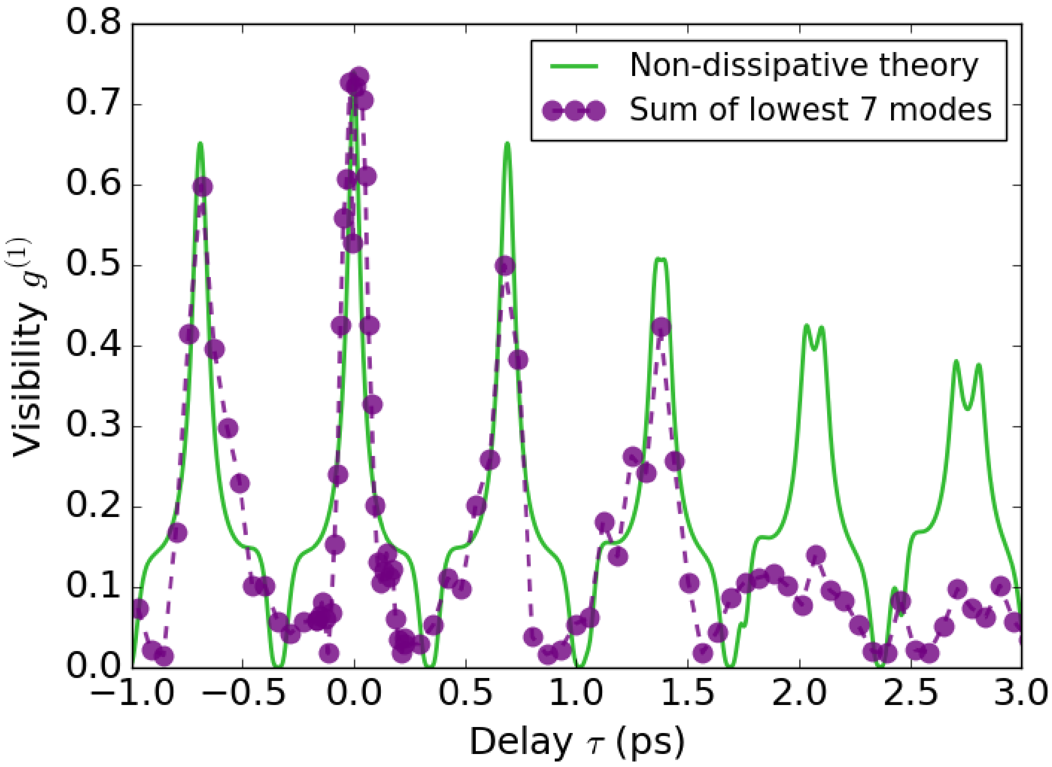


Coherence of all modes: revivals

Coarse scan delay τ for $g^{(1)}(\tau)$

Measure $g^{(1)}(\tau)$ summed over whole spectrum

- Decay time ~$\hbar / kT$ = 25 fs
- Revivals with trapping period 0.6 ps (1/trap frequency)
- Partial revivals show misalignment and anisotropy
- Fits non-dissipative Bose-gas theory



Coherence of ground mode alone

Bi-exponential decay (approx)

Coherence increases with photon number n_0

- Above threshold, $\tau_{coh} \propto n_0$
  Schawlow-Townes limit
- Below threshold $\frac{1}{\tau_c} = \frac{1}{2}\left[\kappa + n_{mol}\sigma(\lambda)c^*\right]$
  - Decoherence from loss and re-absorption (thermalisation)

Breaks down near multimode condensation threshold