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## Dynamics and correlations of a Bose–Einstein condensate of photons

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## PhD Tutorial

# Dynamics and correlations of a Bose–Einstein condensate of photons

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### Abstract

The Tutorial reports recent experimental advances in studies of the dynamics as well as the number and phase correlations of a Bose–Einstein condensed photon gas confined in a high-finesse dye-filled microcavity. Repeated absorption–emission-processes of photons on dye molecules here establish a thermal coupling of the photonic quantum gas to both a heat bath and a particle reservoir comprised of dye molecules. In this way, for the first time Bose–Einstein condensation under grand-canonical statistical ensemble conditions becomes experimentally accessible. The work presented in this Tutorial was one of the four shortlisted finalists of the 2017 DPG SAMOP dissertation prize.

Keywords: Bose–Einstein condensation, quantum optics, correlations, statistical physics, laser physics

(Some figures may appear in colour only in the online journal)

### 1. Grand-canonical Bose–Einstein condensation

Large statistical number fluctuations are a fundamental property known from the thermal behaviour of bosons, as has been strikingly revealed in experiments with light and material particles [1–6]. For low temperatures or high densities, however, when a Bose gas undergoes Bose–Einstein condensation (BEC), the persistence of large particle number fluctuations can conflict with the conservation of the total particle number. Accordingly, fluctuations are damped out and second-order coherence emerges [7–9]. This notion grounds on the microcanonical or canonical statistical description of the system, which applies for systems well-isolated from their environment suppressing both energy and particle exchange with the environment as e.g. realised in ultra-cold atomic gases [9, 10]. Following the first observation of BEC in dilute atomic vapour [11, 12], evidence for the emergence of first-order coherence [13–16] and the suppression of density fluctuations [3, 17–20] have provided

hallmarks for the phase transition. More recently, BECs have also been observed in two-dimensional (2D) gases of exciton-polaritons [21–24], magnons [25] and photons [26–28]. Quintessentially, these systems are open due to their coupling to the environment for e.g. particle injection or thermalisation, which reinforces the relevance of reservoirs for their description, as for example provided by grand-canonical statistics.

In the grand-canonical ensemble, the system is subject to particle (and energy) exchange with a reservoir [29]. For bosons, the population  $n$  in each quantum state suffers large number fluctuations  $\delta n \simeq \bar{n}$ , while the fixed chemical potential (and temperature) accounts for a complete thermodynamic description of the gas. In the thermodynamic limit, all three statistical ensembles are generally expected to become equivalent due to vanishing relative fluctuations of the total particle number, i.e.  $\delta N/\bar{N} \rightarrow 0$ . Applied e.g. to the macroscopically occupied ground state in the Bose–Einstein condensed phase ( $\bar{n} \simeq \bar{N}$ ), however, in the grand-canonical ensemble large fluctuations of the total particle number,  $\delta N \simeq \bar{N}$  occur. Surprisingly, the statistical fluctuations here become enhanced as the system temperature approaches

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absolute zero instead of being frozen out. This so-called *grand-canonical fluctuation catastrophe* has been a long-standing issue in theoretical physics [9, 10, 30–37] and its observation has long remained elusive. Most notably, Ziff, Uhlenbeck and Kac altogether questioned the physical significance of the grand-canonical ensemble in the condensed phase [32]; their arguments, however, apply only for diffusive contact between a spatially separated BEC and particle reservoir.

In contrast, for a BEC of photons in a dye-filled optical microcavity genuine grand-canonical statistical conditions in the condensed phase can become relevant. Here, the coupling of the condensed particles to an effective reservoir is realised by interparticle conversion between photons, ground and excited state dye molecules [38, 39]. In this system, we have for the first time observed grand-canonical number statistics in a BEC by demonstrating its coupling to both a heat bath and a particle reservoir [40–42]. These results provided a first experimental hint at the fluctuation catastrophe. Moreover, our work revealed phase fluctuations of the condensate wave function in the wake of grand-canonical statistical number fluctuations [40, 41, 43].

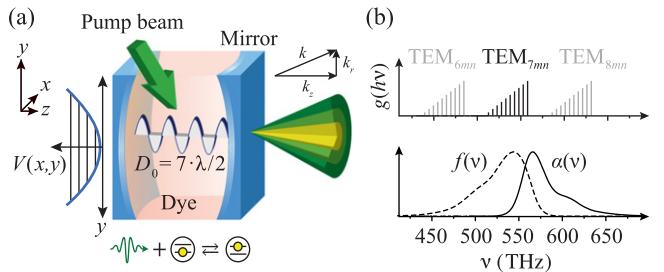
The present Tutorial contains a theoretical and experimental study of the thermalisation dynamics and first and second-order temporal correlations of a Bose–Einstein condensed photon gas under canonical and grand-canonical ensemble conditions. The Tutorial is organised as follows: section 2 introduces the concept of photon BEC, sections 3–5 give a theoretical description of the photon thermalisation process, along with the BEC number and phase correlations, while sections 6–8 describe our corresponding experiments. Finally, section 9 concludes and gives an outlook.

## 2. Bose–Einstein condensation of photons

Photons depict a prime example among the Bose gases known today and yet, it has taken almost a century to find ways to condense them—*why?* Thermal photons usually do not become quantum degenerate: in blackbody radiation, for example, the coupling of temperature and total photon number prohibits BEC at low temperatures as photons at  $T \rightarrow 0$  vanish instead of forming a condensate<sup>2</sup>. In optical gases with a conserved particle number as e.g. in nonlinear microcavities, photon–photon interactions are usually too small to achieve efficient thermalisation of the light [44, 45].

Quantum fluids of light have nevertheless emerged in recent years by synthesising dressed light–matter-states [46], such as exciton–polaritons in microcavities [24] or surface-plasmon-polaritons [47]. These platforms have provided long sought-after evidence for condensation, coherence [21–23, 48–50] and thermalisation [51–54] in optical quantum gases.

More recently, BEC of *pure* photons has also become tractable by implementing a photon thermalisation mechanism with an incoherent molecular medium that realises a non-zero chemical potential for the light [55]; see [56, 57] for



**Figure 1.** Scheme of the experiment. (a) The dye-filled optical cavity consists of two highly-reflecting mirrors separated by  $q = 7$  half-wavelengths. The spherical mirror curvature introduces a harmonic potential  $V(x, y)$  for the transverse motion of the photons. A pump laser excites the dye molecules and subsequent emission–absorption-cycles lead to a thermalisation of the photon gas at  $T = 300$  K. The cavity emission is monitored in a spatially, spectrally and time-resolving way. (b) Cavity mode spectrum of the photons (top), and spectral profiles of dye fluorescence  $f(\nu)$  and absorption  $\alpha(\nu)$  (bottom). The height of the bars indicates the degeneracy of the cavity eigenmodes. Fluorescence photons are emitted into transverse modes with fixed  $q = 7$  (black bars), making the photon gas effectively 2D. Reproduced from [64]. CC BY 4.0.

similar concepts. The first observation of photon BEC in 2010 by Klaers *et al* [26, 27, 58] has been confirmed in more recent work by Marelic *et al* [28] and Greveling *et al* [59]. In the meantime, a number of experiments have elaborated on the thermalisation [41, 60, 61], the calorimetry [62], the first-order spatial coherence [63, 64], the first- and second-order temporal correlations [40, 43], the polarisation properties [65], non-local interactions [66] and the generation of lattices and micropotentials for photon condensates [67, 68]. Key aspects that are related to the topics discussed in this Tutorial have been studied in a (non-exhaustive) series of theoretical work on photon condensation and its dynamics [69–72], on grand-canonical particle number correlations [39, 58, 73], on phase diffusion [74] and on the relation of photon condensation and lasing [75, 76].

In this section, we introduce the scheme for BEC of photons in a dye-filled optical microcavity with a focus on thermal and chemical equilibrium, the microcavity dispersion and the statistical physics of the photons.

### 2.1. Photons in a dye-filled microcavity

Figure 1 shows our microcavity experiment, which consists of two curved mirrors spaced by  $D_0 \simeq 1.4 \mu\text{m}$  (or  $1.6 \mu\text{m}$ ) and filled with a liquid dye solution. At a mirror separation  $D_0 = q\lambda/2\tilde{n}_0$  the resonator encloses  $q = 7$  (or 8) half waves, which corresponds to a free spectral range of adjacent longitudinal cavity modes  $\Delta\lambda = \lambda^2/2\tilde{n}_0D_0 \simeq 80 \text{ nm}$  ( $\Delta\nu \simeq 75 \text{ THz}$ ) comparable to the spectral width of the dye fluorescent emission (figure 1(b)). Here,  $\lambda$  denotes the optical wavelength in vacuum and  $\tilde{n}_0$  is the refractive index of the dye solution. Accordingly, photons associated with a fixed longitudinal wave number  $q$  are absorbed and emitted into the resonator differing only in their transverse quantum numbers  $m$  and  $n$ . Effectively, this reduced dimensionality introduces a low-energy ground state (‘cutoff’) for the photon gas  $\hbar\omega_c$ ,

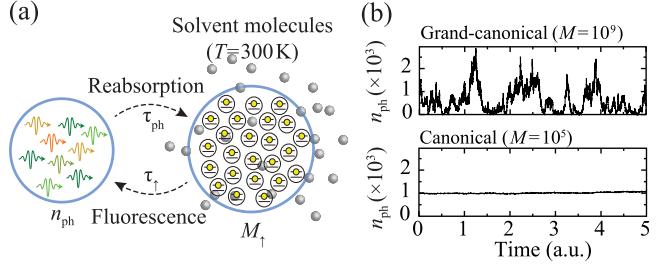
<sup>2</sup> In other words: chemical potential  $\mu = 0$ .

which is given by the  $\text{TEM}_{q00}$  cavity mode. In other words, the photons can be ascribed an effective mass  $m_{\text{ph}} = \hbar\omega_c/(c/\tilde{n}_0)^2$  and the photon kinetics is reduced to the transverse plane of the resonator. Additionally, the curvature of the cavity mirrors imposes an in-plane confinement (figure 1(a), left). The photon gas behaves formally equivalent to a 2D, harmonically trapped ideal Bose gas, for which in thermal equilibrium BEC is expected below a finite critical temperature  $T_c$  or above a critical particle number  $N_c$  [77, 78].

The solved dye molecules are optically pumped by a laser beam and the electronically excited molecules decay via emission of fluorescence photons in the cavity modes, as sketched in figure 1((b), top). Inside the high-finesse cavity, frequent absorption–emission-cycles of photons by dye molecules establish a thermal contact between both subsystems in the sense of the grand-canonical ensemble: the photon gas acquires a temperature  $T$  (room temperature) and chemical potential  $\mu$ , as determined by the much larger molecular reservoir. Firstly, for the (energy) thermalisation the spectral distributions of fluorescence  $f(\omega)$  and absorption  $\alpha(\omega)$ , see figure 1(b), are required to scale with a Boltzmann-factor  $f(\omega)/\alpha(\omega) \propto \omega^3 \exp(-\hbar\omega/k_B T)$ <sup>3</sup>. The fluorescence-induced energy exchange between the photons and the (thermal) molecular bath then translates to the spectrum of the photon gas. Secondly, the chemical (particle number) equilibration rests on the fact that the interaction between photons and molecules can be considered as a photochemical reaction, see figure 1((a), bottom). The required energy to electronically excite a dye molecule  $\hbar\omega \simeq 2.3$  eV exceeds thermal energy  $k_B T \simeq 0.025$  eV by far, which suppresses fluctuation-driven dye excitations by a factor of order  $\exp(-\hbar\omega/k_B T) \approx 10^{-37}$ . Similarly, the thermally excited emission of photons into the cavity modes ( $\hbar\omega \simeq 2$  eV) is very unlikely. An optical photon (of energy  $\hbar\omega$ ) is emitted only, if another optical photon ( $\hbar\omega'$ ) has been previously absorbed. If this condition is maintained throughout the experiments, the photon number does not decrease as the gas is cooled down, i.e.  $\mu \neq 0$ , in contrast to blackbody radiation.

The thermalisation process equilibrates photons over the set of  $\text{TEM}_{mn}$  modes, leading to an average internal energy of the photon gas  $\sim k_B T$  above  $\hbar\omega_c$ . ‘Cold’ photons propagate near the optical axis, while ‘hot’ photons exhibit large angles with respect to the optical axis. By heating up the dye solution, an enhanced population of highly excited transverse states is observed [27, 64]. The equilibrium Bose–Einstein distribution has been experimentally confirmed in the dye-microcavity experiment [26–28, 40, 41, 43, 55, 60, 62]: for small total particle numbers,  $N \leq N_c$ , the photon energies are Boltzmann-distributed, while for  $N > N_c$  adding more photons results in the accumulation of a BEC in the transverse ground state accompanied by a saturation of excited transverse modes.

The light–matter-interaction between photons and molecules at room temperature is incoherent, due to many dephasing collisions between dye and solvent molecules



**Figure 2.** Statistical ensemble and particle number fluctuations. (a) The photon gas couples to the reservoir of electronically excited molecules by reabsorption after a photon lifetime  $\tau_{\text{ph}}$ . The molecular excitations decay within  $\tau_{\uparrow}$ , establishing chemical equilibrium between the photon gas and the particle reservoir. Simultaneously, multiple collisions of dye and solvent molecules lead to a thermalisation of the rovibronic dye states at room temperature. The light–matter-interaction imprints the thermal equilibrium state onto the photon gas and the molecules can be regarded as a heat bath. (b) Temporal fluctuations of the photon number under grand-canonical ensemble conditions (large reservoir  $M = 10^9$ , top), and damped fluctuations in the canonical ensemble (small reservoir  $M = 10^5$ , bottom).

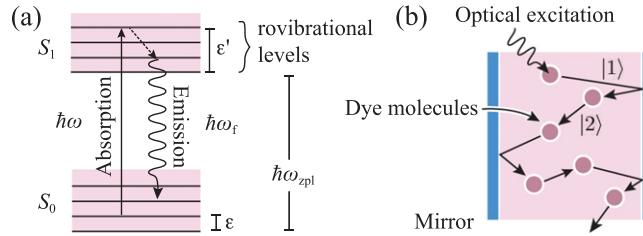
during the dye excited state lifetime [83, 84]. Consequently, the dynamics of photons and molecules can be modelled by rate equations, which also determine the mean population of photons  $\bar{n}$  and excited dye molecules  $\bar{M}_{\uparrow}$  (see figure 2(a)). As for typical experimental parameters  $\bar{M}_{\uparrow} \gg \bar{n}$ , the ensemble of excited dye molecules can be viewed as an effective particle reservoir for the photon gas. The heat energy and particle exchange with the dye reservoir paves the way for studies of the transition from canonical to grand-canonical ensemble conditions, as illustrated in figure 2(b).

In the grand-canonical ensemble, where each eigenstate suffers strong number fluctuations  $\delta n_i / \bar{n}_i \simeq 1$ , the second-order coherence of a BEC is expected to be substantially reduced, i.e.  $g^{(2)}(0) \simeq (\delta n_0 / \bar{n}_0)^2 + 1 = 2$ . Experimentally, we find evidence for large statistical intensity fluctuations in BECs, which persist up condensate fractions of  $\bar{n}_0 / \bar{N} \simeq 70\%$  as long as the particle reservoir complies with grand-canonical conditions [38, 40, 43]. This is in contrast to experiments with ultra-cold atoms, where a reduction of density fluctuations in the Bose–Einstein condensed phase has been observed [3, 5, 6, 17]. In this case, the emergence of second-order coherence is related to the isolation of the atomic ensemble from its environment, which necessitates a statistical description in the microcanonical ensemble with fixed particle number and Poissonian fluctuations  $\delta n_0 / \bar{n}_0 = 1 / \sqrt{\bar{n}_0} \xrightarrow{\bar{n}_0 \gg 1} 0$ , i.e.  $g^{(2)}(0) = 1$ . Interestingly, also the photon statistics in a laser follows a Poissonian distribution [85–87]. The fluctuation properties of photon BECs under grand-canonical conditions differ strikingly from those of both lasers and BECs in the microcanonical or canonical ensemble.

## 2.2. Thermal equilibrium

We outline the fluorescence-induced thermalisation process of the microcavity photon gas, which is based on radiative

<sup>3</sup> For many dye solutions at room temperature the scaling is based on the Kennard–Stepanov relation [79–82].



**Figure 3.** (a) Electronic and rovibrational energy levels in a dye molecule. (b) In the ‘photon box’ absorption–emission-cycles by dye molecules frequently change the configuration of the light field  $|1\rangle \rightarrow |2\rangle \rightarrow \dots$  to thermalise the photon gas.

energy exchange between photons and dye molecules by absorption–emission-processes. The latter establish a thermal contact between the system (photon gas) and a heat bath at room temperature (dye solution) by dissipating excess energy of ‘hot’ photons and providing energy for ‘colder’ photons. For this, the spectral absorption and emission profiles of the dye molecules are required to fulfil the so-called Kennard–Stepanov relation, as will be discussed in the following. A more refined derivation of the thermalisation process can be found in [27, 55, 58].

The relevant, underlying molecular processes are sketched in the simplified energy diagram of a dye molecule in figure 3(a). The electronic ground and excited singlet states  $S_{0,1}$  exhibit a (quasi-)continuous subset of rotational and vibrational modes (shaded areas), and the energy difference between the ground states in  $S_0$  and  $S_1$  is on the order of  $\hbar\omega_{zpl} \simeq 2$  eV (zero-phonon line). After a photon absorption ( $\hbar\omega_a$ ), frequent collisions between dye and solvent molecules ( $10^{-15}$  s time scale at room temperature) rapidly alter the rovibrational molecular state, resulting in a thermal distribution in the electronically excited manifold. During the relaxation, any excess energy is dissipated by the solvent bath on a  $10^{-12}$  s timescale. To this end, the subsequent fluorescence emission ( $\tau = 10^{-9}$  s,  $\hbar\omega_f$ ) occurs from a thermally equilibrated state  $S_1$  to the ground state  $S_0$ , which is subject to the same relaxation mechanism.

This insight allows us to derive a Boltzmann-type law relating the spectral absorption and emission profiles of the dye molecules, known as the Kennard–Stepanov relation [79–82, 88, 89]. We obtain the ratio of fluorescence  $f(\omega)$  and absorption  $\alpha(\omega)$  by integrating over the rovibrational energy levels

$$\frac{f(\omega)}{\alpha(\omega)} \propto \frac{\int D_{\uparrow}(\epsilon') p(\epsilon') A(\epsilon', \omega) d\epsilon'}{\int D_{\downarrow}(\epsilon) p(\epsilon) B(\epsilon, \omega) d\epsilon}, \quad (1)$$

where  $\epsilon$ ,  $\epsilon'$  denote energies and  $D_{\downarrow}(\epsilon)$ ,  $D_{\uparrow}(\epsilon')$  the rovibrational density of states in ground ( $\downarrow$ ) and excited ( $\uparrow$ ) state. Due to the collisional relaxation,  $p(\epsilon') = \exp(-\epsilon'/k_B T)$  in both states. Considering energy conservation  $\hbar\omega + \epsilon = \hbar\omega_{zpl} + \epsilon'$ , the Einstein coefficients  $A(\epsilon', \omega)$  and  $B(\epsilon, \omega)$  are related by  $D_{\uparrow}(\epsilon') A(\epsilon', \omega) d\epsilon' = \frac{2\hbar\omega^3}{\pi c^2} D_{\downarrow}(\epsilon) B(\epsilon, \omega) d\epsilon$  [88]. Assuming identical rovibrational substructures,  $D_{\downarrow}(\epsilon) = D_{\uparrow}(\epsilon')$ , (1) yields

the Kennard–Stepanov relation

$$\frac{f(\omega)}{\alpha(\omega)} \propto \frac{2\hbar\omega^3}{\pi c^2} \exp\left(-\frac{\hbar(\omega - \omega_{zpl})}{k_B T}\right). \quad (2)$$

Experimentally, the scaling has been verified in e.g. liquid dye solutions [58, 90–93], dye-doped polymers [60], semiconductors [94] or ultra-dense gases [95].

The Kennard–Stepanov relation is the key ingredient for the photon thermalisation mechanism to work. In our high-finesse ‘photon box’, see figure 3(b), the fluorescence photons undergo many absorption–emission-cycles, corresponding to a random walk of the light field configuration [58]. The ratio of the transition rates between two configurations  $|1\rangle \rightarrow |2\rangle$ , which differ from each other by the absorption of one photon with frequency  $\omega_i$  and the emission of one photon with frequency  $\omega_j$ , is

$$\frac{R_{12}}{R_{21}} = \frac{\alpha(\omega_i) f(\omega_j) \omega_j^3}{\alpha(\omega_j) f(\omega_i) \omega_i^3} = e^{-\hbar(\omega_j - \omega_i)/k_B T} \quad \forall i, j. \quad (3)$$

From the theory of Markov processes it is known that exactly such a Boltzmann-scaling of the transition rates evokes a thermal state of the master equation (detailed balance) [58, 96, 97]. The Kennard–Stepanov relation ensures that the photon gas for sufficiently long times acquires a thermal equilibrium state.

### 2.3. Chemical equilibrium

Besides energy exchange with the heat bath (temperature  $T$ ), the effective particle exchange between the photon gas and the dye reservoir assigns the photons a chemical potential  $\mu_{\gamma}$ . In the following, we will see that it is determined by the excitation level of the dye medium.

Due to  $\hbar\omega_{zpl} \gg k_B T$ , purely thermal excitation of molecules from their ground ( $\downarrow$ ) to excited electronic ( $\uparrow$ ) states is strongly suppressed, and optical photons ( $\gamma$ ) are required to drive the transition. Vice versa, the decay of a molecule results (with an efficiency of  $\simeq 95\%$ ) in the emission of a photon. Altogether, such a behaviour resembles a photochemical reaction:



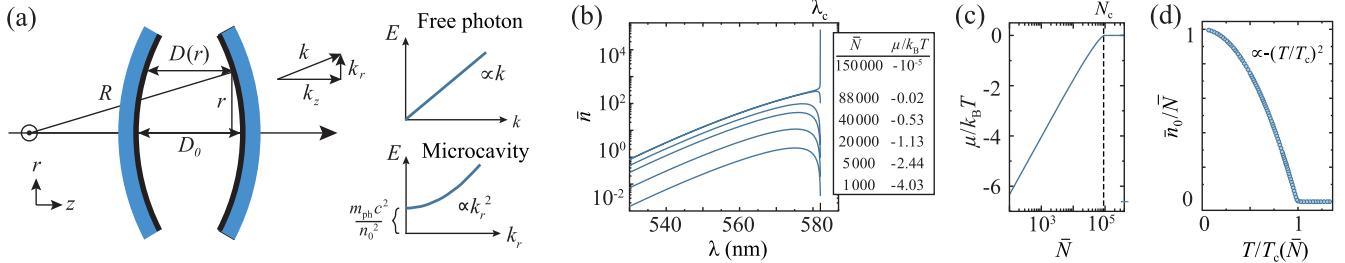
In chemical equilibrium (i.e. zero net particle flux between different species) the particular chemical potentials balance,  $\mu_{\gamma} + \mu_{\downarrow} = \mu_{\uparrow}$ . Thus, the fugacity of the photons reads

$$z = e^{\frac{\mu_{\gamma}}{k_B T}} = e^{\frac{\mu_{\uparrow}}{k_B T}} / e^{\frac{\mu_{\downarrow}}{k_B T}}. \quad (5)$$

The partition function of a dye molecule  $\mathcal{F} = w_{\downarrow} \exp(\mu_{\downarrow}/k_B T) + w_{\uparrow} \exp[(\mu_{\uparrow} - \hbar\omega_{zpl})/k_B T]$ , with the statistical weights  $w_{\downarrow, \uparrow} = \int_{\epsilon \geq 0} D_{\downarrow, \uparrow}(\epsilon) \exp[-\epsilon/k_B T] d\epsilon$ , allows one to associate the molecular chemical potential with the probability of finding a molecule in the ground or excited electronic state, respectively:

$$w_{\downarrow} \frac{e^{\frac{\mu_{\downarrow}}{k_B T}}}{\mathcal{F}} = \frac{M_{\downarrow}}{M}, \quad w_{\uparrow} \frac{e^{\frac{\mu_{\uparrow} - \hbar\omega_{zpl}}{k_B T}}}{\mathcal{F}} = \frac{M_{\uparrow}}{M}. \quad (6)$$

This probability is determined by the ratio of the number of excited and relaxed dye molecules  $M_{\uparrow, \downarrow}$  and the total number of



**Figure 4.** (a) Microcavity geometry showing mirrors (radius of curvature  $R$ ) separated by  $D(r)$  at a transverse position  $r$ . In the paraxial approximation ( $k_z \gg k_r$ ), one finds a modification of the photon dispersion relation from linear scaling  $E = \hbar c / \tilde{n}_0$  in 3D free space (top) to a quadratic scaling with small transverse momenta  $k_r$  in the microcavity (bottom), similar to the dispersion relation of a 2D massive particle. (b) Spectral occupation versus wavelength. The rest energy of the photons is determined by the cutoff wavelength  $\lambda_c = \hbar \tilde{n}_0 / (m_{ph} c)$ . Below the critical photon number  $N_c \approx 90 000$ , the spectra show a Boltzmann scaling. For  $\bar{N} > N_c$ , the ground state becomes macroscopically occupied. (c) The chemical potential grows in the classical region with increasing particle number until it saturates at  $\mu = 0$  around  $N_c$ . (d) The condensate fraction exhibits an quadratic scaling as a function of the reduced temperature. Experimentally, we adjust the reduced temperature by varying  $T_c \propto \sqrt{N}$  to match room temperature  $T = 300$  K when operating at the phase transition.

molecules  $M$ . By renormalising the chemical potential with respect to the cavity ground state energy,  $\mu = \mu_\gamma - \hbar\omega_c$ , (5) yields

$$e^{\frac{\mu}{k_B T}} = \frac{w_\downarrow}{w_\uparrow} \frac{M_\uparrow}{M_\downarrow} e^{-\frac{\hbar\Delta}{k_B T}}. \quad (7)$$

In equilibrium, the chemical potential  $\mu$  is thus determined by the dye molecular excitation level  $M_\uparrow/M_\downarrow$  and the detuning between the condensate frequency and dye resonance  $\Delta = \omega_c - \omega_{zpl}$ .

#### 2.4. Microcavity dispersion relation

The microcavity photons can be formally described as a 2D harmonically trapped Bose gas. In the resonator filled with a dye medium with index of refraction  $\tilde{n}$ , the energy-momentum-relation of a photon in free space  $E = \hbar c / \tilde{n} \sqrt{k_r^2 + k_z^2}$  with  $c$  the speed of light,  $k_r$  the radial and  $k_z = \pi q / D(r)$  the longitudinal wave vector ( $q$  is an integer longitudinal wave number) component gets modified by the cavity boundary conditions. These are determined by the mirror spacing  $D(r) \simeq D_0 - r^2/R$  at a radial distance  $r$  from the optical axis, where  $D_0$  denotes the mirror separation on the optical axis. These parameters are illustrated in figure 4(a). Using the paraxial approximation ( $k_z \gg k_r$ ), the dispersion relation of the microcavity photons becomes

$$E \simeq \frac{\pi \hbar c q}{\tilde{n} D_0} + \frac{\pi \hbar c q}{\tilde{n} R D_0^2} r^2 + \frac{\hbar c D_0}{2\pi q \tilde{n}} k_r^2. \quad (8)$$

In addition, we extend (8) by accounting for a nonlinear response of the refractive index subject to changes of the 2D photon density, i.e. the intensity of the light field. The total index of refraction  $\tilde{n} = \tilde{n}_0 + \Delta\tilde{n}_r = \tilde{n}_0 + \tilde{n}_2 I(\mathbf{r})$  can be written as a sum of the linear refractive index in the absence of photons  $\tilde{n}_0$  and a nonlinear contribution  $\tilde{n}_2$ . The nonlinear term results from physical effects that lead to intensity-dependent energy shifts, as e.g. the optical Kerr effect [98], or temporally slow thermal lensing [67]. Assuming  $\tilde{n}_0 \gg \Delta\tilde{n}_r$ ,

we obtain

$$E = m_{ph} \frac{c^2}{\tilde{n}_0^2} + \frac{\hbar^2 k_r^2}{2m_{ph}} + \frac{1}{2} m_{ph} \Omega^2 r^2 - \frac{m_{ph} c^2}{\tilde{n}_0^3} \tilde{n}_2 I(\mathbf{r}). \quad (9)$$

Here, the effective photon mass  $m_{ph} = \pi \hbar q \tilde{n}_0 / (D_0 c)$  and trapping frequency  $\Omega = c / \tilde{n}_0 \sqrt{D_0 R / 2}$  have been introduced, revealing the formal equivalence of (9) with the dispersion of a massive, harmonically trapped particle moving non-relativistically in a 2D plane, see figure 4((a), right). The first term in (9) determines the effective rest energy of the photons, a global energy shift determined by the cavity boundary conditions. It corresponds to the energy of the  $q$ th longitudinal mode without any transverse excitations  $m_{ph} (c/\tilde{n}_0)^2 = E_{q00} = \hbar\omega_c$  with the cutoff frequency  $\omega_c$ . The eigenenergies in the cavity are given by 2D (isotropic) harmonic oscillator states  $E_{n_x, n_y} = m_{ph} c^2 / n_0^2 + \hbar \Omega (n_x + n_y + 1)$  with quantum numbers  $n_x$  and  $n_y$ . The eigenfunctions  $\psi_{n_x, n_y}(x, y) = \psi_{n_x}(x) \cdot \psi_{n_y}(y)$  are given by the 1D solutions  $\psi_n(x) = (\sqrt{2^n} n! \sqrt{\pi} b)^{-1} H_n(x/b) \exp[-x^2/(2b^2)]$ , where  $b = \sqrt{\hbar / m_{ph} \Omega}$  denotes the oscillator length and  $H_n(x)$  the Hermite polynomials.

#### 2.5. Statistical physics of microcavity photons

In the following, we will describe the temperature behaviour of the (ideal) 2D photon gas in a harmonic trap [77, 99–102]. We derive the critical particle number and temperature, respectively, as well as the spectral and spatial distributions for the experimentally studied photon gas. We can specify the transversal excitation energies in the harmonic trap

$$u_{n_x, n_y} = E_{n_x, n_y} - m_{ph} \frac{c^2}{\tilde{n}_0^2} - \hbar \Omega = \hbar \Omega (n_x + n_y) \quad (10)$$

with a degeneracy of the eigenstates  $g(u) = 2(u/(\hbar\Omega) + 1)$ , where the factor 2 accounts for the two-fold polarisation degeneracy of the photons. At temperature  $T$ , the average occupation of an excited state with energy  $u_{n_x, n_y}$  is given by

the Bose–Einstein distribution

$$\bar{n}_{T,\mu}(u) = \frac{g(u)}{\exp[(u - \mu)/k_B T] - 1}. \quad (11)$$

Here, we have implicitly assumed that the system is grand-canonical with a chemical potential  $\mu$  adjusting the average total particle number  $\bar{N}$  under the constraint  $\bar{N} = \sum_{u=0,\hbar\Omega,2\hbar\Omega,\dots} \bar{n}_{T,\mu}(u)$ . At high temperatures or low total photon numbers, the chemical potential obeys  $\mu/k_B T \ll 0$ , and (11) equals the classical Boltzmann distribution. In the opposite limit ( $T \rightarrow 0$  or  $N \rightarrow \infty$ ), the chemical potential converges asymptotically to the ground state energy  $\mu \rightarrow 0^-$  (figure 4(c)) and the ground state becomes macroscopically occupied. The phase transition to a BEC occurs at the critical photon number or temperature, respectively,

$$N_c = \frac{\pi^2}{3} \left( \frac{k_B T}{\hbar \Omega} \right)^2, \quad T_c = \frac{\sqrt{3}}{\pi} \frac{\hbar \Omega}{k_B} \sqrt{\bar{N}}, \quad (12)$$

and as a function of the cavity parameters  $T_c \propto (\bar{N}/R)^{1/2}$ . Notably, an equilibrium phase transition requires its critical temperature to remain finite in the thermodynamic limit ( $\bar{N}, V \rightarrow \infty$ ). It can be achieved by increasing the particle number  $\bar{N}$  and volume  $V \propto R^2$  in a way that conserves  $\bar{N}/R$ , i.e. by gradually switching off the trapping potential  $R \rightarrow \infty$ .

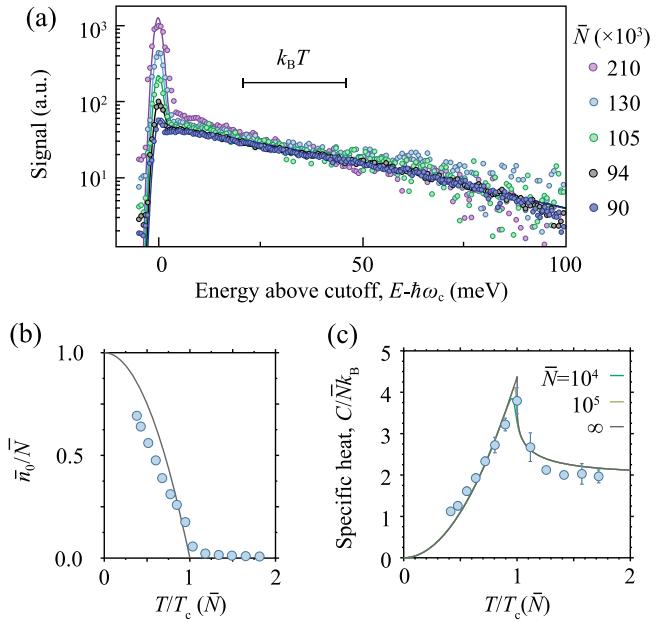
The expected spectral photon distributions for increasing chemical potentials are shown in figure 4(b). The condensation fraction scales quadratically with the reduced temperature,  $\bar{n}_0/\bar{N} = 1 - (T/T_c)^2$ , see figure 4(d), as expected for a 2D harmonically trapped ideal Bose gas [99, 102]. In this confinement, BEC occurs not only in momentum space but also in position space. The spatial intensity distribution of the condensed photon gas is the sum over all oscillator eigenfunctions weighted with the Bose–Einstein factor:

$$I_{T,\mu}(x, y) \simeq \frac{2m_{\text{ph}}c^2}{\tilde{n}_0^2 \tau_{\text{rt}}} \sum_{n_x, n_y} \frac{|\psi_{n_x, n_y}(x, y)|^2}{\exp\left(\frac{\hbar\Omega(n_x + n_y) - \mu}{k_B T}\right) - 1}. \quad (13)$$

The power per photon is accounted for by  $m_{\text{ph}}(c/\tilde{n}_0)^2/\tau_{\text{rt}}$ , where  $\tau_{\text{rt}} = 2D_0\tilde{n}_0/c$  denotes the photon round trip time of in the resonator. This approximation is valid due to  $\hbar\Omega \sim 0.1 \text{ meV}$  being much smaller than the rest energy  $m_{\text{ph}}(c/\tilde{n}_0)^2 \sim 1 \text{ eV}$ .

The spectral and spatial distributions of the photon gas have been experimentally verified for the first time for both the classical and Bose–Einstein condensed phase in pioneering work by Klaers *et al* [26, 27, 55, 58]. Subsequent studies have provided further insight into the phase transition, and revealed e.g. thermodynamic properties such as condensate fraction or specific heat [28, 40, 41, 43, 60, 62–64, 66]; see figure 5 for an overview of some experimental signatures of photon BEC.

The purpose of the present Tutorial is to elucidate the coherence properties of BECs of light. For this study, typically realised experimental parameters for the microcavity setup are  $q = 8$  at a cavity cutoff wavelength of  $\lambda_c = 580 \text{ nm}$ , which is associated with a mirror separation of  $D_0 = 1.62 \mu\text{m}$ . The refractive index of the dye medium (Rhodamine 6G solved in ethylene glycol) amounts to  $\tilde{n}_0 = 1.43$  at room



**Figure 5.** Evidence for BEC of light. (a) Photon spectra for increasing total photon numbers  $\bar{N}$ . For  $\bar{N} \geq N_c$ , the excited mode population saturates and the ground state becomes macroscopically occupied. Reprinted figure with permission from [43]. Copyright (2016) by the American Physical Society. (b) The condensate fraction and (c) the specific heat of the photon gas is derived from the spectral distributions (not shown) at various particle numbers above and below  $N_c$ . The discontinuity close to  $T = T_c(\bar{N})$  reveals the phase transition. (b) Reproduced from [62]. CC BY 4.0.

temperature  $T = 300 \text{ K}$ . Most of the experiments are conducted using mirrors with a radius of curvature of  $R = 1 \text{ m}$ . Therefore, the effective photon mass is  $m_{\text{ph}} = 7.8 \times 10^{-36} \text{ kg}$  and the frequency of the harmonic trap is  $\Omega/2\pi = 37 \text{ GHz}$ . With this one expects a critical particle number  $N_c = 93\,000$ . Due to the short resonator round trip time  $\tau_{\text{rt}} = 15 \text{ fs}$ , the average circulating optical power in the resonator at threshold becomes  $P_c \approx 2.1 \text{ W}$ . The highly-reflecting mirrors transmit a fraction  $\tilde{T} \simeq 2.5 \times 10^{-5}$  of the optical power. At criticality, the continuous power of the cavity emission is approximately 5 nW. Table 1 summarises parameter sets, which are used in the course of the discussed experimental sequences.

### 3. Multimode photon dynamics

Bose–Einstein condensation is a phase transition of the Bose gas in thermal equilibrium. The thermalisation of a nonequilibrium system can occur via different mechanisms and with characteristic dynamics. Atomic gases e.g. relax into equilibrium by contact interactions, while microcavity photons inherit their temperature solely from the thermal contact to a molecular heat bath. The atomic equilibration requires several interatomic collisions [103, 104], whereas the photons can be thermalised after only a single absorption–emission–cycle. In this section, we will theoretically investigate the photon dynamics.

**Table 1.** Photon gas parameters for some different microcavity geometries. (Refractive index  $\tilde{n}_0 = 1.43$  for ethylene glycol, temperature  $T = 300$  K.)

$q$	7	7	8	7	
$R$	1	6	1	1	m
$\lambda_c$	580	580	580	560	nm
$D_0$	1.42	1.42	1.62	1.37	$\mu\text{m}$
$m_{\text{ph}}$	7.79	7.79	7.80	8.07	$10^{-36}$ kg
$\Omega/2\pi$	39.6	16.2	37.1	40.3	GHz
$N_c$	81700	490200	93200	78800	

### 3.1. Rate equation model

We start our discussion by analysing the rate equations for absorption and emission of photons in the microcavity modes. As the dye solution is embedded into the cavity volume, fluorescence emission occurs directly into the reabsorbing medium. After an absorption process, the high collision rate between solvent and dye molecules at room temperature leads to decoherence of the molecular dipoles [83]. The photon-dye-system here correspondingly is in the weak coupling regime [84, 105]. In first-order perturbation theory, the photon dynamics can thus be adequately modelled by semi-classical rate equations, which are determined by the time evolution of the diagonal elements of the density matrix.

To begin with, we consider a configuration of the light field  $\{n_1, n_2, \dots, n_i, \dots\}$  with  $n_i$  photons in the  $i$ th cavity mode. The transition rates (per volume)

$$\begin{aligned} R_{12}^i(\mathbf{r}) &= B_{12}(\omega_i)u^i(\mathbf{r})\rho_{\downarrow}n_i \\ R_{21}^i(\mathbf{r}) &= B_{21}(\omega_i)u^i(\mathbf{r})\rho_{\uparrow}(n_i + 1), \end{aligned} \quad (14)$$

give the probability (per time) to absorb or emit a photon in mode  $i$  at position  $\mathbf{r}$  with the frequency-dependent Einstein coefficients for absorption and emission  $B_{12,21}(\omega_i)$ , the spectral energy density per photon  $u^i(\mathbf{r})$ , and the densities of ground and excited state molecules  $\rho_{\downarrow,\uparrow}$ . Due to the densities on the right-hand side in (14) one obtains rates per volume, which yield absolute rates after integrating out the resonator volume. For the transverse ground state ( $i = q00$ ) with  $\omega_i = E_{q00}/\hbar$  and  $n_i = n$ , this gives

$$R_n^{12} = B_{12}\left(\frac{E_{q00}}{\hbar}\right)u^{q00}(0)\frac{n\rho_{\downarrow}}{|f^{q00}(0)|^2}. \quad (15)$$

Here, we have expressed the energy density  $u^{q00}(\mathbf{r}) = u^{q00}(0)|f^{q00}(\mathbf{r})|^2/|f^{q00}(0)|^2$  by the normalised mode function  $f^{q00}(\mathbf{r})$ . Using the effective mode volume  $\tilde{V}_{\text{eff}}^{q00} = \int|f^{q00}(\mathbf{r})|^2/\max\{|f^{q00}(\mathbf{r})|^2\}dV = 1/|f^{q00}(0)|^2$  [106, 107], the modified Einstein coefficients  $\hat{B}_{12,21} = B_{12,21}(E_{q00}/\hbar)u^{q00}(0)$  and the number of ground and excited state molecules  $M_{\downarrow,\uparrow} = \rho_{\downarrow,\uparrow}\tilde{V}_{\text{eff}}^{q00}$ , one obtains the rate equations for the ground mode populated with  $n$  photons

$$\begin{aligned} R_n^{12} &= \hat{B}_{12}M_{\downarrow}n = \hat{B}_{12}(M - X + n)n \\ R_n^{21} &= \hat{B}_{21}M_{\uparrow}(n + 1) = \hat{B}_{21}(X - n)(n + 1). \end{aligned} \quad (16)$$

According to the photochemical reaction in (4), we have expressed the rates as a function of the sum of all molecular and

photonic excitations  $X = M_{\uparrow} + n$  and the total molecule number in the ground mode volume  $M = M_{\downarrow} + M_{\uparrow} = M_{\downarrow} + X - n$ , which we assume to be constant reservoir parameters.

The rate equations readily provide the temporal evolution of the photon number

$$\frac{\partial}{\partial t}n_i = \hat{B}_{21}M_{\uparrow}(n_i + 1) - (\hat{B}_{12}M_{\downarrow} + \gamma_{\text{ph},i})n_i \quad (17)$$

with a photon loss rate  $\gamma_{\text{ph},i}$  due to mirror transmission. To conserve the excitation number  $X$ , any loss must be compensated for by a net gain  $P$  in the molecule rate equations, which is experimentally realised by pumping with a laser beam:

$$-\frac{\partial}{\partial t}M_{\downarrow} = \frac{\partial}{\partial t}M_{\uparrow} = P - \sum_i \frac{\partial}{\partial t}n_i - \gamma_M M_{\uparrow}. \quad (18)$$

Additionally,  $P$  must balance the molecular loss rate  $\gamma_M$ , which results from non-radiative decay and fluorescence into unconfined leakage modes.

### 3.2. Steady-state photon number

The rate equation model enables a quantitative description of the photon thermalisation dynamics in the microcavity. For this, we consider a simplified model for the multimode photon gas in the uncondensed phase without spatial photon transport, losses or pumping ( $P = \gamma_M = \gamma_{\text{ph},i} = 0$ ). Here, the molecule number ( $10^8$ ) exceeds the average photon number per mode ( $10^1$ ), such that the rate equations of the dye medium in (18) can be considered as quasi-stationary with a fixed molecular excitation level  $M_{\uparrow}/M_{\downarrow}$ . A more refined model including dissipative spatial dynamics has been theoretically reported by Kirton and Keeling [69, 70]. Our own detailed numerical simulations of the spatial photon dynamics are discussed in section 6.5 [41].

For a single cavity mode (angular frequency  $\omega_i$ ), the Kennard–Stepanov relation reads  $\hat{B}_{21}^i/\hat{B}_{12}^i = w_{\downarrow}/w_{\uparrow} \exp[-\hbar(\omega_i - \omega_{\text{zpl}})/k_B T]$ . Together with (7), we obtain the average photon number in thermal and chemical equilibrium from (17):

$$\bar{n}_i = \left( \frac{\hat{B}_{12}^i M_{\downarrow}}{\hat{B}_{21}^i M_{\uparrow}} - 1 \right)^{-1} = (e^{\frac{\hbar(\omega_i - \omega_c) - \mu}{k_B T}} - 1)^{-1}. \quad (19)$$

By summing (17) over all degenerate cavity modes with the energy  $\epsilon_i = \hbar\omega$ , we obtain the rate equation for the photon

number  $n \equiv n(\omega, t) = \sum_{\epsilon_i=\hbar\omega} n_i(t)$  in the multimode cavity:

$$\begin{aligned} \frac{\partial n}{\partial t} &= \hat{B}_{21} \left[ n + \sum_{\epsilon_i=\hbar\omega} 1 \right] M_{\uparrow} - \hat{B}_{12} n M_{\downarrow} \\ &= \hat{B}_{21} n M_{\uparrow} + \hat{A}_{21} M_{\uparrow} - \hat{B}_{12} n M_{\downarrow}. \end{aligned} \quad (20)$$

The term  $\sum_{\epsilon_i=\hbar\omega} 1$  gives the energy-dependent mode density  $g(\omega) = 2[(\omega - \omega_c)/\Omega + 1]$ . In the second step, we have identified the Einstein coefficient for spontaneous emission  $\hat{A}_{21} = g(\omega) \hat{B}_{21}$ . The steady-state photon number is  $\bar{n}(\omega) = g(\omega) \{ \exp[\hbar(\omega - \omega_c) - \mu/k_B T] - 1 \}^{-1}$ .

### 3.3. Spectral photon number evolution

To determine the thermalisation time, we rephrase the single mode (17) as  $\dot{n}_i + \alpha n_i + \beta = 0$ , with the coefficients  $\alpha = \hat{B}_{12}^i M_{\downarrow} - \hat{B}_{21}^i M_{\uparrow}$  and  $\beta = -\hat{B}_{21}^i M_{\uparrow}$ . For the initial condition  $n_i(0) = 0$ , this differential equation is solved by

$$n_i(t) = -\frac{\beta}{\alpha} [1 - e^{-\alpha t}] = \bar{n}_i [1 - e^{-t/\tau_i}], \quad (21)$$

with the time constant  $\tau_i = (\bar{n}_i + 1) / (\hat{B}_{12}^i M_{\downarrow}) = \bar{n}_i / (\hat{B}_{21}^i M_{\uparrow})$ . We expand (21):

$$n_i(t) = \hat{B}_{21} M_{\uparrow} t \left[ 1 - \frac{t}{2} (\hat{B}_{12} M_{\downarrow} - \hat{B}_{21} M_{\uparrow}) \right]. \quad (22)$$

For early times, we can neglect the second-order term  $\propto t^2$ , so that the spectrum in this limit will be determined by the emission profile  $\hat{B}_{21}(\omega)$ . If we approximate (22) for the uncondensed regime with  $M_{\uparrow} \ll M_{\downarrow} \hat{B}_{12}/\hat{B}_{21}$ , we obtain the characteristic time after which the initial spectral redistribution of the photon gas occurs

$$\tau_{\text{th}} \simeq \frac{1}{\hat{B}_{12} M_{\downarrow}}. \quad (23)$$

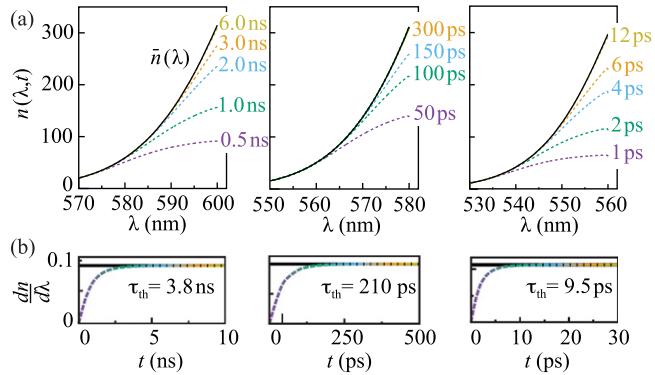
This equals the mean reabsorption time of a photon in the dye medium. The relative occupation of  $n_i$  and  $n_{i+1}$  of two neighbouring resonator modes (with frequencies  $\omega_i$ ,  $\omega_i + \Omega$ ) demonstrates, that the spectral slope is nearly thermal after  $\tau_{\text{th}}$ . Without loss of generality, we assume that the fluorescence strength into the modes is equal,  $\hat{B}_{21}^i = \hat{B}_{21}^{i+1}$ , as is indeed fulfilled for the used dyes (section 6). Using the Kennard–Stepanov relation, the absorption coefficients  $\hat{B}_{12}^i = \hat{B}_{21}^i \exp[\hbar(\omega_i - \omega_{\text{ZPL}})/k_B T]$  and  $\hat{B}_{12}^{i+1} = \hat{B}_{12}^i \exp[\hbar\Omega/k_B T]$  yield the photon dynamics

$$n_{i,i+1}(t) \simeq M_{\uparrow} \hat{B}_{21}^{i,i+1} t \left[ 1 - \frac{t}{2} M_{\downarrow} \hat{B}_{12}^{i,i+1} \right]. \quad (24)$$

An expansion in  $\hbar\Omega/k_B T$  determines the spectral population difference at the thermalisation time

$$\frac{n_{i+1} - n_i}{\Omega}(\tau_{\text{th}}) = -\frac{1}{2} \frac{\hbar}{k_B T} \frac{M_{\uparrow} \hat{B}_{21}^i}{M_{\downarrow} \hat{B}_{12}^i} = -\frac{1}{2} \frac{\hbar}{k_B T} \bar{n}_i \quad (25)$$

in the limit of a Boltzmann distribution with  $\bar{n}_i = M_{\uparrow} \hat{B}_{21}^i (M_{\downarrow} \hat{B}_{12}^i)^{-1}$ . For the equilibrium distribution in (19), the similar scaling  $(\bar{n}_{i+1} - \bar{n}_i)/\Omega = -\hbar/(k_B T) \bar{n}_i$  demonstrates



**Figure 6.** (a) Spectral thermalisation dynamics for  $\lambda_c = \{600; 580; 560\}$  nm. All shown equilibrium spectra  $\bar{n}(\lambda)$  (solid lines) are close to the condensation threshold. The wavelength-dependent dye reabsorption,  $\sigma(\lambda) \propto \exp(-\lambda)$ , leads to a slower photon number evolution (dashed) in the red ( $10^{-9}$  s) than in the green spectral region ( $10^{-11}$  s). (b) The low-wavelength spectral slope (dashed) quantifies the degree of thermalisation, which approaches the equilibrium value (solid) with advancing times. After  $\tau_{\text{th}}$  the relative difference between both curves is less than 1%. (Dye concentration 0.1 mmol l<sup>-1</sup>,  $\mu = -0.007 k_B T$ .)

that the photon spectrum agrees except for a factor 1/2 with the spectral shape of a Boltzmann distribution after the reabsorption time  $\tau_{\text{th}}$ . Accordingly, the microcavity photons have relaxed to a thermal-like equilibrium after completing approximately one emission-absorption-cycle.

In general, the relaxation rates of individual modes depend on their frequencies. It is therefore helpful to express the Einstein coefficient for absorption  $\hat{B}_{12}(\omega)$  as a function of the experimentally accessible cross section  $\sigma(\omega)$ . By comparing the coefficients in Beer's law  $\partial n / \partial t = -M_{\downarrow} \tilde{V}_{\text{eff}}^{-1} \sigma(\omega) c n$  and the rate equation for absorption  $\partial n / \partial t = -M_{\downarrow} \hat{B}_{12}(\omega) n$ , we find the useful relation

$$\hat{B}_{12}(\omega) = \frac{\sigma(\omega) c}{\tilde{V}_{\text{eff}}}. \quad (26)$$

Figure 6(a) shows the calculated temporal evolution of the spectra in (21) for Rhodamine 6G dye (section 6). Owing to the wavelength-dependence of the absorption cross section (maximum near 530 nm), the time  $\tau_{\text{th}}$  after which the spectral distribution has relaxed to a thermal equilibrium distribution  $\bar{n}(\lambda)$  varies for different spectral regions. While the photon gas relaxation takes several nanoseconds in the red spectral region (figure 6(b), left), for the yellow-green spectral region a thermalisation time of a few picoseconds is predicted (right). For example, Rhodamine 6G absorbs photons at 580 nm wavelength with cross section  $\sigma(2\pi c/\lambda) \simeq 10^{-22}$  m<sup>2</sup>. With the mirror separation  $D_0 \simeq 1.6$   $\mu\text{m}$  and the diameter of the TEM<sub>00</sub> mode  $d_0 \simeq 12$   $\mu\text{m}$  we can estimate the effective ground mode volume  $\tilde{V}_{\text{eff},00} = \pi(d_0/2)^2 D_0 \simeq 1.8 \times 10^{-16}$  m<sup>3</sup> and the rate coefficient  $\hat{B}_{12}(\omega) \simeq 166$  s<sup>-1</sup>. For typical dye concentrations near 1 mmol l<sup>-1</sup>, around  $M_{\downarrow} \approx 10^8$  dye molecules reside in the mode volume. Therefore, the expected thermalisation time is  $\tau_{\text{th}} \approx 50$  ps.

### 3.4. Chemical equilibration time

The spectral thermalisation time of the photon gas  $\tau_{\text{th}}$  in the uncondensed phase is approximately given by the photon reabsorption time in the dye solution,  $1/\hat{B}_{21}M_{\downarrow}$ , see (23). In general, this value differs from the chemical equilibration time  $\tau_{\text{ch}}$ , which is the time after which the system has acquired its steady-state-population  $\bar{n}(\omega)$ . To see this, we extend the single mode description in (21) to the multimode system:

$$n(\omega, t) = \bar{n}(\omega)[1 - e^{-t/\tau(\omega)}], \quad (27)$$

with  $\tau(\omega) = \bar{n}(\omega)/[g(\omega)\hat{B}_{21}(\omega)M_{\uparrow}]$ . For a Boltzmann distribution  $\bar{n}(\omega) = g(\omega)\hat{B}_{21}(\omega)M_{\uparrow}/[\hat{B}_{12}(\omega)M_{\downarrow}]$ , the chemical equilibration time is the weighted average over all frequency-dependent relaxation times

$$\tau_{\text{ch}} = \frac{\int_{\omega_c}^{\infty} \tau(\omega) \bar{n}(\omega) d\omega}{\int_{\omega_c}^{\infty} \bar{n}(\omega) d\omega} \stackrel{k_B T \gg \hbar\Omega}{\simeq} \frac{\tau_{\text{th}}}{4} e^{-\frac{\hbar\Delta}{k_B T}}, \quad (28)$$

where we have assumed  $\hat{B}_{21}(\omega)$  to be independent of  $\omega$ , as is roughly fulfilled for Rhodamine 6G dye within the relevant wavelength range (540–600 nm). In our experiments, the dye-cavity detuning  $\Delta = \omega_c - \omega_{\text{zpl}}$  takes values between  $\Delta_{560\text{nm}} = -2.4k_B T/\hbar$  and  $\Delta_{605\text{nm}} = -8.7k_B T/\hbar$ , implying the chemical equilibration time to exceed the spectral relaxation time by  $\tau_{\text{ch}}/\tau_{\text{th}} \approx 3$  (560 nm) up to 1500 (605 nm). This prediction is experimentally verified (section 6). Below the critical photon number, our simplified analytical model provides an adequate description of the photon number thermalisation dynamics. It should be noted, that this model is insufficient to predict the dynamics in the Bose–Einstein condensed phase where the optical feedback onto the dye requires using the molecular rate equations. In this regime, the large photon number speeds up the dynamics by stimulated emission events and the chemical equilibration can become much faster than the spectral thermalisation, as will be discussed section 6.5 on the basis of numerical simulations.

## 4. Grand-canonical photon statistics

For BEC in the grand-canonical statistical ensemble regime, i.e. in the presence of a large particle reservoir, large statistical number fluctuations on the order of the total particle number  $N$  have been predicted [9, 10, 30–37]. In contrast to this, the (micro-)canonical statistical ensemble features Poissonian number fluctuations, i.e. a scaling with  $\sqrt{N}$ ; a situation realised in most atomic BECs [3, 5, 6, 17, 20]. In the dye-cavity system, Bose–Einstein condensed photons couple to electronic transitions of a specific number of dye molecules, which realises the repeated exchange of photon- and molecule-like excitations. The latter can be interpreted as an effective particle reservoir for the photons, with a size that depends on the molecule number and the dye-cavity detuning. We find that the photon number statistics of the ground state resembles a (nearly) Bose–Einstein distributed thermal light source, in contrast to both atomic BECs and the laser [17, 108, 109].

Under these conditions, the phase transition can be regarded as a BEC in the grand-canonical ensemble regime.

### 4.1. Photon number distribution

We start by considering the master equation for the probability  $p_n \equiv p_n(t)$  to find  $n$  photons in the ground state at time<sup>4</sup>  $t$ . The flow of probability between photons in the condensate and the reservoir is

$$\dot{p}_n = R_{n-1}^{21} p_{n-1} - (R_n^{12} + R_n^{21}) p_n + R_{n+1}^{12} p_{n+1}, \quad (29)$$

with the rates given by (16) [38, 110]. According to the experiment, we assume  $M = M_{\uparrow} + M_{\downarrow}$  and  $X = M_{\uparrow} + n$  to be constant. For  $t \rightarrow \infty$  the probability flow  $\dot{p}_n(t)$  is expected to become stationary,  $\dot{p}_n(\infty) = 0$ , and the photon number distribution converges to its equilibrium value  $\mathcal{P}_n := p_n(\infty)$ . In this limit, (29) is solved by the recursive ansatz  $\mathcal{P}_n = \mathcal{P}_0 \prod_{k=0}^{n-1} R_k^{21} / R_{k+1}^{12}$ , and one obtains the photon number statistics

$$\frac{\mathcal{P}_n}{\mathcal{P}_0} = \frac{(M - X)! X!}{(M - X + n)! (X - n)!} \left( \frac{\hat{B}_{21}}{\hat{B}_{12}} \right)^n, \quad (30)$$

which is used to calculate the average condensate number and its fluctuations. Similarly, the statistics can be derived by a entropy maximisation principle [39].

In general, (30) has to be evaluated numerically. At constant temperature  $T$ , we induce the phase transition by increasing the particle number  $\bar{N}$ , which effectively lowers the reduced temperature  $T/T_c(\bar{N})$ . For each  $\bar{N}$ , the following numerical method then computes the excitation number  $X$  that recovers the given particle number  $\bar{N}$ : for a starting value  $X$ , the average photon number in the condensate

$$\bar{n} = \sum_{n \geq 0} n \mathcal{P}_n \quad (31)$$

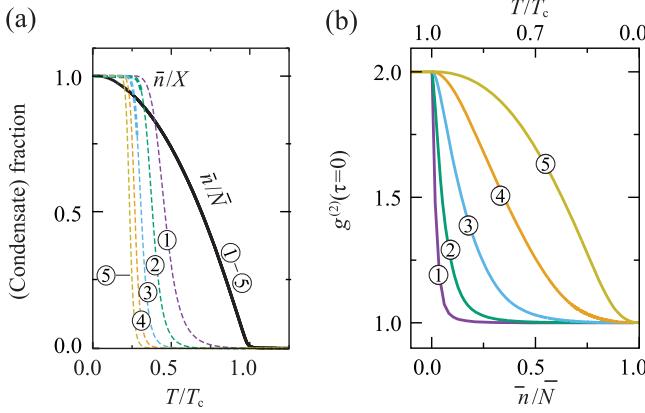
and the molecular excitation level of the medium in the ground mode volume

$$\frac{M_{\uparrow}}{M_{\downarrow}} = \frac{X - \bar{n}}{M - X + \bar{n}} \quad (32)$$

are computed. As the density of excited molecules (and thus the excitation level) is required to be spatially homogeneous in chemical equilibrium, the ratio  $M_{\uparrow}/M_{\downarrow}$  controls the chemical potential for the photon gas, see (7). Accordingly, the number of photons in excited states is  $\bar{n}_{\text{exc}} = \sum_{u>0} g(u)/(\exp[(u - \mu)/(k_B T)] - 1)$ . If there are residual deviations between  $\bar{n} + \bar{n}_{\text{exc}}$  and the target photon number  $\bar{N}$ , the numerical method is iterated with an adjusted excitation number  $X$  until a certain level of precision is reached.

Figure 7(a) shows the calculated condensate fraction  $\bar{n}/\bar{N}$  and the photonic fraction of the excitation number  $\bar{n}/X$  for five different-sized molecular reservoirs as a function of the reduced temperature. The constant dye-cavity detuning  $\hbar\Delta = -4.67k_B T$  controls the Kennard–Stepanov relation  $\hat{B}_{21}/\hat{B}_{12}$  and hence the photon statistics in (30). For all

<sup>4</sup> We will denote  $n$  (and  $\bar{n}$ ) as the (average) photon number in the BEC in sections 4 and 5.



**Figure 7.** (a) Condensate fraction  $\bar{n}/\bar{N}$  and photonic fraction of the excitation number  $\bar{n}/X$  versus reduced temperature  $T/T_c(\bar{N})$  for reservoir sizes  $M_{1-5} = \{10^8, \dots, 10^{12}\}$  and dye-cavity detuning  $\hbar\Delta = -4.67k_B T$  (adapted figure with permission from [38], copyright (2012) by the American Physical Society). (b) The autocorrelation function  $g^{(2)}(0)$  of the condensate (reservoirs as in (a)) predicts large photon number fluctuations even deep in the condensed phase. Adapted figure with permission from [38]. Copyright (2012) by the American Physical Society.

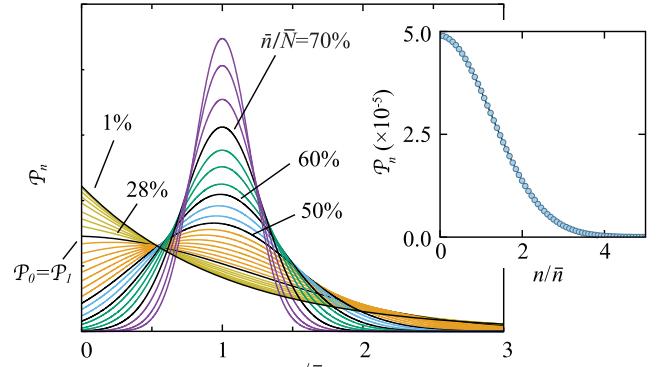
studied reservoirs, the condensate fraction follows the analytic solution  $\bar{n}/\bar{N} = 1 - (T/T_c)^2$  and the curves for  $\bar{n}/X$  reveal that a large number of excitations are present as molecular excitations down to very low temperatures. Furthermore, figure 7(b) shows the zero-delay autocorrelation function

$$g^{(2)}(\tau = 0) = \frac{\langle n(n-1) \rangle}{\bar{n}^2} = \frac{\sum_{n \geq 0} n(n-1) \mathcal{P}_n}{\left(\sum_{n \geq 0} n \mathcal{P}_n\right)^2} \quad (33)$$

for the same reservoir parameters as a function of the condensate fraction and the reduced temperature. For  $T \geq T_c$ , the ground state occupation exhibits the usual, strong intensity fluctuations in a single mode of the thermal Bose gas,  $g^{(2)}(0) = 2$ , and the photon number statistics is Bose–Einstein-distributed. In the presence of large reservoirs, the intensity correlations maintain when the temperature is lowered deep into the condensed phase, as attributed to the grand-canonical particle exchange with the dye reservoir. For  $T/T_c \ll 1$ , the statistical number fluctuations are damped out and our calculations demonstrate the emergence of second-order coherence,  $g^{(2)}(0) = 1$ , with Poissonian statistics. We do not find indications that the transition between both statistical regimes is accompanied by a discontinuity in the thermodynamic quantities, excluding a further phase transition scenario within the Bose–Einstein condensed phase. The crossover of the photon statistics in the condensed phase remains valid also in the thermodynamic limit, as will be discussed later.

#### 4.2. Asymptotic photon number distributions

We show that the photon number statistics interpolates between a Bose–Einstein- and Poissonian distribution. To



**Figure 8.** Crossover from Bose–Einstein to Poissonian statistics for increasing  $\bar{n}/\bar{N}$  at fixed reservoir size. The probability  $\mathcal{P}_n$  is plotted versus the photon number normalised with the mean condensate population  $\bar{n}$ . Up to  $\bar{n}/\bar{N} = 28.3\%$  the maximum of the distribution occurs at  $n_{\max} = 0$ . For increasing quantum degeneracy, the distribution shifts to  $n_{\max} > 0$  and the variance is reduced, demonstrating a damping of particle number fluctuations and the emergence of second-order coherence. *Inset:* photon number statistics at the crossover  $T_x = 0.847 T_c$  ( $\bar{n}/\bar{N} \approx 28.3\%$ ). The probability distribution corresponds to a Gaussian with  $n \approx 27\,000$  photons. ( $M = 10^{10}$ ,  $\hbar\Delta = -2.4k_B T$ , polarisation degeneracy neglected; the curves normalised in the  $n/\bar{n}$ -representation for clarity.)

analytically derive the limiting cases, we rewrite (30) in a recursion form:

$$\frac{\mathcal{P}_{n+1}}{\mathcal{P}_n} = \frac{X - n}{M - X + n + 1} \frac{\hat{B}_{21}}{\hat{B}_{12}}. \quad (34)$$

For *Bose–Einstein statistics* to apply,  $\mathcal{P}_n$  must follow a geometric series with a ratio  $\mathcal{P}_{n+1}/\mathcal{P}_n$  being independent of  $n$ . This is fulfilled if and only if the reservoir dimensions  $M$  and  $X$  are very large, so that the photon number on the right-hand side of (34) can be safely neglected, i.e.  $X \gg n$  and  $M - X \gg n$  ('grand-canonical limit'). With  $X \approx M_{\uparrow}$  and  $M - X \approx M_{\downarrow}$ ,

$$\frac{\mathcal{P}_{n+1}}{\mathcal{P}_n} \stackrel{\text{g.c.}}{=} \frac{M_{\uparrow} \hat{B}_{21}}{M_{\downarrow} \hat{B}_{12}} \Rightarrow \frac{\mathcal{P}_n}{\mathcal{P}_0} \stackrel{\text{g.c.}}{=} \left( \frac{M_{\uparrow} \hat{B}_{21}}{M_{\downarrow} \hat{B}_{12}} \right)^n. \quad (35)$$

Here,  $\mathcal{P}_n$  decays exponentially from its maximum at  $n = 0$ . Normalisation of (35) gives

$$\mathcal{P}_n = \left( 1 - \frac{M_{\uparrow} \hat{B}_{21}}{M_{\downarrow} \hat{B}_{12}} \right) \left( \frac{M_{\uparrow} \hat{B}_{21}}{M_{\downarrow} \hat{B}_{12}} \right)^n = \frac{\left( \frac{\bar{n}}{\bar{n} + 1} \right)^n}{\bar{n} + 1}. \quad (36)$$

In the last step, we have identified the average condensate number from (19). This result remains valid also for increased  $M$ , as long as the excitation level  $M_{\uparrow}/M_{\downarrow} \approx X/(M - X)$  (and thus  $\mu$ ,  $\bar{n}$ ,  $\bar{N}$ ) are kept constant. Equation (36) is the well-known *Bose–Einstein statistics*, see figure 8, which also applies for example for chaotic, thermal light or blackbody radiation.

In the case of *Poisson statistics*, the most probable photon number is finite,  $n_{\max} > 0$ . Under the assumption  $\mathcal{P}_{n_{\max}+1} = \mathcal{P}_{n_{\max}}$ , (34) yields  $n_{\max} = X - (M + 1)/(1 + \hat{B}_{21}/\hat{B}_{12})$ .

Expanding for  $\Delta n = n - n_{\max}$ ,

$$\frac{\mathcal{P}_{n+1}}{\mathcal{P}_n} = 1 - \frac{\Delta n}{\lambda} + \frac{1}{1 + \hat{B}_{21}/\hat{B}_{12}} \left( \frac{\Delta n}{\lambda} \right)^2 - \dots \quad (37)$$

with  $\lambda = \hat{B}_{21}/\hat{B}_{12}M + 1/(\hat{B}_{21}/\hat{B}_{12} + 1)^2$ . In the low temperature limit, the ratio of the Einstein coefficients scales with the dye-cavity detuning  $\Delta$ . For a negative detuning, as in our experiments, it diverges:

$$\frac{\hat{B}_{21}(\omega)}{\hat{B}_{12}(\omega)} = \frac{w_{\downarrow}}{w_{\uparrow}} e^{-\frac{\hbar\Delta}{k_B T}} \stackrel{T \rightarrow 0}{\simeq} \begin{cases} 0, & \Delta > 0 \\ \infty, & \Delta < 0. \end{cases} \quad (38)$$

Hence, (37) simplifies to  $\mathcal{P}_{n+1}/\mathcal{P}_n \simeq \lambda/(\lambda + \Delta n)$  (or  $\simeq (\lambda - \Delta n)/\lambda$ ) for  $\Delta > 0$  (or  $\Delta < 0$ ). This recursion formula implies the relative probability near  $\Delta n$  around the maximum  $n_{\max}$ :

$$\frac{\mathcal{P}_{n_{\max} + \Delta n}}{\mathcal{P}_{n_{\max}}} \simeq \begin{cases} \frac{(\lambda - 1)!}{(\lambda - 1 + \Delta n)!} \lambda^{\Delta n}, & \Delta > 0 \\ \frac{\lambda!}{(\lambda - \Delta n)!} \lambda^{-\Delta n}, & \Delta < 0. \end{cases} \quad (39)$$

Upon transforming  $\Delta n \rightarrow -\Delta n$ , both distributions are the same and their relative scaling is analogous to a Poisson distribution

$$\mathcal{P}_n^p = e^{-\lambda} \frac{\lambda^n}{n!} \Rightarrow \frac{\mathcal{P}_{n_{\max} + \Delta n}^p}{\mathcal{P}_{n_{\max}}^p} = \frac{\lambda!}{(\lambda + \Delta n)!} \lambda^{\Delta n} \quad (40)$$

which contains only one parameter  $\lambda$  for mean and variance. The solutions in (39) however are only Poissonian with respect to the relative photon number  $\Delta n$ , as the an additional parameter  $n_{\max}$  tunes the most probable photon number. For example, in the limit  $T \rightarrow 0$  ( $\Delta \neq 0$ ) the statistics peaks at  $n_{\max} = \bar{n} = \bar{N}$  with  $\lambda = 0$ , where all photons of the systems have condensed into the ground state and the photon number is precisely known.

#### 4.3. Statistics crossover

Figure 8 shows numerically calculated photon number distributions to find  $n$  photons in the BEC for a fixed reservoir size ( $M = 10^{10}$ ,  $\Delta = \omega_c - \omega_{\text{zpl}} = -2.4k_B T/\hbar$ ). By decreasing the reduced temperature from  $T/T_c = 1.0$  to  $0.4$ , or vice versa increasing the condensate fraction from  $\bar{n}/\bar{N} \simeq 1\%$  to  $80\%$ , one observes a continuous crossover from Bose–Einstein to Poissonian statistics.

As the crossover point between both statistical regimes, we define the reduced temperature  $T_x/T_c(\bar{N})$  when the most probable photon number ceases to be  $n_{\max} = 0$ , or in other words when the condition  $\mathcal{P}_0 = \mathcal{P}_1$  is fulfilled (figure 8, inset). Inserting into (34) yields:

$$\frac{M + 1}{X} = 1 + \frac{\hat{B}_{21}}{\hat{B}_{12}}. \quad (41)$$

Due to the large number of molecules, we assume  $M + 1 \simeq M$ . With regard to the experimental conditions, we derive  $T_x$  for fixed total numbers of molecules  $M$  and photons  $\bar{N}$ . With  $X = \bar{n} + M_{\uparrow}$  the average condensate population

**Table 2.** Numerically calculated reduced temperatures and condensate fractions, at which the crossover between Bose–Einstein and Poissonian statistics occurs for different-sized molecular particle reservoirs  $M_{\text{①–⑤}}$ . Here, the autocorrelation gives  $g^{(2)}(0) \simeq \pi/2$ , see (45). ( $\hbar\Delta = -2.4k_B T_c$ ,  $\bar{N} = 10^5$ .)

$M_{\text{①–⑤}}$	$10^8$	$10^9$	$10^{10}$	$10^{11}$	$10^{12}$
$T_x/T_c$	0.979	0.946	0.847	0.602	0.359
$\bar{n}/\bar{N}$	3.0%	9.2%	28.3%	63.1%	86.7%
$g^{(2)}(0)$	1.5706	1.5707	1.5708	1.5708	1.5708

follows

$$\bar{n} = M \frac{\frac{\hat{B}_{12}}{\hat{B}_{21}} \frac{M_{\downarrow}}{M_{\uparrow}} - 1}{\left(1 + \frac{\hat{B}_{12}}{\hat{B}_{21}}\right) \left(1 + \frac{M_{\downarrow}}{M_{\uparrow}}\right)}. \quad (42)$$

In the grand-canonical limit ( $M_{\downarrow}, M_{\uparrow} \gg \bar{n}$ ), see (36), the nominator corresponds to the inverse of the average photon number  $\bar{n} = \sum_{n=0}^{\infty} n \mathcal{P}_n = [(\hat{B}_{12}/\hat{B}_{21})(M_{\downarrow}/M_{\uparrow}) - 1]^{-1}$ . Although grand-canonical conditions do not strictly apply in the crossover region, we use this to estimate  $T_x$ . We find

$$\bar{n} = \sqrt{\frac{M}{\left[1 + \frac{w_{\uparrow}}{w_{\downarrow}} e^{\frac{\hbar\Delta}{k_B T}}\right] \left[1 + \frac{w_{\downarrow}}{w_{\uparrow}} e^{\frac{\hbar\omega_{\text{zpl}} - \mu_{\gamma}}{k_B T}}\right]}}, \quad (43)$$

where both the Kennard–Stepanov relation and chemical equilibrium have been applied. Moreover, it is safe to assume  $w_{\downarrow} = w_{\uparrow}$  for the statistical weights of ground and excited molecular states [83]. In the condensed phase, the chemical potential of the photons  $\hbar(\omega_c + \Omega) < \mu_{\gamma} < \hbar\omega_c$ , and consequently we can use  $\mu_{\gamma} \simeq \hbar\omega_c = \hbar(\Delta + \omega_{\text{zpl}})$  to simplify the second bracket term in the denominator.

Equation (43) resembles a boundary for the average number of condensed photons, up to which the particle number statistics can be considered Bose–Einstein-like. With  $\bar{n} = \bar{N}[1 - (T_x/T_c)^2]$ , this implicitly determines the temperature  $T_x$  for the crossover. To investigate the scaling of the reduced crossover temperature  $t = T_x/T_c$  with the system parameters, we rewrite (43):

$$1 - t^2 = \frac{\sqrt{M/2}}{\bar{N}} \left[ 1 + \cosh\left(\frac{\hbar\Delta}{k_B T_c} \frac{1}{t}\right) \right]^{-1/2}. \quad (44)$$

The temperature depends only on relative size of the subsystems  $\sqrt{M}/\bar{N}$  and the reduced detuning  $\hbar\Delta/k_B T_c$ , which plays an important role for the thermodynamic limit:  $\bar{N}, R, M \rightarrow \infty$  with  $R/\bar{N} = \text{const.}$  and  $\sqrt{M}/\bar{N} = \text{const.}$  The first condition conserves the critical temperature  $T_c$  and therefore fixes  $\hbar\Delta/k_B T_c$ , see (12). The second requirement conserves  $T_x$  (below  $T_c$ ), which rules out that the temperature difference arises from finite-size effects. Notably, both regimes, Bose–Einstein- and Poissonian statistics, exist *within* the condensed phase. While the former relates to the grand-canonical ensemble ( $M \gg \bar{n}^2$ ), the latter refers to a canonical ensemble scenario ( $M \ll \bar{n}^2$ ). The crossover between both regimes is induced by changing  $\bar{n}/\bar{N}$  or  $\Delta$ , respectively. To highlight this, table 2 summarises numerically calculated

values for  $T_x/T_c$  for different reservoirs  $M_{\text{eff}} = 10^8\text{--}10^{12}$  at fixed photon gas sizes  $\bar{N} = 10^5$  and dye-cavity-detuning  $\hbar\Delta = -2.4k_B T_c$  ( $\lambda_c = 560$  nm and  $\lambda_{\text{zpl}} = 545$  nm). For sufficiently large reservoirs, the Bose-Einstein-like grand-canonical statistics extends deep into the condensed phase.

The inset of figure 8 shows the Gaussian photon number distribution at  $T_x/T_c = 0.847$ , given by  $\mathcal{P}_n = 2/(\pi\bar{n})\exp[-(n/\bar{n})^2/\pi]$ . Accordingly, the zero-delay second-order correlation function reads

$$g^{(2)}(0) = \frac{\sum_{n \geq 0} n(n-1)\mathcal{P}_n}{\left[\sum_{n \geq 0} n\mathcal{P}_n\right]^2} = \frac{\pi}{2} - \frac{1}{\bar{n}} \stackrel{n \gg 1}{\simeq} \frac{\pi}{2}, \quad (45)$$

which analytically reproduces the numerical results in table 2. It corresponds to relative condensate number fluctuations of  $\delta n/\bar{n} = \sqrt{g^{(2)}(0) - 1} \simeq 75\%$ .

#### 4.4. Second-order time correlations

We extend our discussion of the photon statistics to the temporal dynamics of the statistical fluctuations [111]. The condensate photons are absorbed by  $M_{\downarrow}$  molecules in the electronic ground state, and  $M_{\uparrow}$  excited molecules decay by emission of photons into the condensate mode. Neglecting losses, the rate (17) becomes

$$\frac{\partial}{\partial t} n = \hat{B}_{21}(X - n)(1 + n) - \hat{B}_{12}n(M - X + n) \quad (46)$$

with  $X = M_{\uparrow} + n$ ,  $M = M_{\downarrow} + M_{\uparrow}$  and the steady-state solution  $\bar{n} = 1/(\hat{B}_{12}\bar{M}_{\downarrow}/\hat{B}_{21}\bar{M}_{\uparrow} - 1)$ . To quantify the time evolution of deviations from  $\bar{n}$ , we define  $\delta n(t) = n(t) - \bar{n}$  and obtain

$$\frac{\partial}{\partial t} \delta n(t) = -(\hat{B}_{12} + \hat{B}_{21})\delta n(t)^2 - \gamma\delta n(t), \quad (47)$$

$$\begin{aligned} \gamma &= \frac{\hat{B}_{21}X}{\bar{n}} + (\hat{B}_{12} + \hat{B}_{21})\bar{n} \\ &\simeq \frac{\hat{B}_{12}\hat{B}_{21}}{\hat{B}_{12} + \hat{B}_{21}} \frac{M}{\bar{n}} + (\hat{B}_{12} + \hat{B}_{21})\bar{n}. \end{aligned} \quad (48)$$

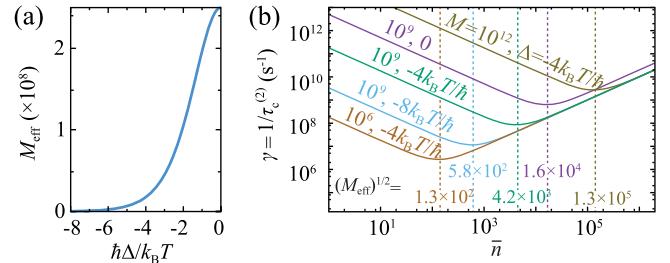
For typical experimental parameters,  $(\hat{B}_{12} + \hat{B}_{21})\delta n \simeq 10^6 \text{ s}^{-1}$  and  $\gamma \simeq 10^9 \text{ s}^{-1}$ , the coefficients in (47) comply with  $(\hat{B}_{12} + \hat{B}_{21})\bar{n}\sqrt{g^{(2)}(0) - 1} \ll \gamma$ , where  $\delta n/\bar{n} = \sqrt{g^{(2)}(0) - 1}$ . The equation of motion can thus be linearised,  $\partial_t \delta n(t) \simeq -\gamma \delta n(t)$ . The second-order autocorrelations at times  $t$  and  $t' = \tau + t$  reads

$$g^{(2)}(\tau) = \frac{\langle n(t + \tau)n(t) \rangle}{\bar{n}^2} = 1 + \frac{\langle \delta n(\tau)\delta n(0) \rangle}{\bar{n}^2}, \quad (49)$$

where  $\langle \delta n(t + \tau) \rangle = \langle \delta n(t) \rangle = 0$  has been used. Using the quantum regression theorem [111–113] allows us to trace back the dynamics  $\langle \delta n(\tau)\delta n(0) \rangle$  to the evolution of  $\delta n(t)$ :  $\partial_t \langle \delta n(\tau)\delta n(0) \rangle \simeq -\gamma \langle \delta n(\tau)\delta n(0) \rangle$ . We find

$$g^{(2)}(\tau) - 1 \simeq [g^{(2)}(0) - 1] \exp\left(-\frac{\tau}{\tau_c^{(2)}}\right), \quad (50)$$

where  $\tau_c^{(2)} = \gamma^{-1}$  denotes the second-order correlation time. With the Kennard–Stepanov relation, the inverse correlation time



**Figure 9.** (a) Effective reservoir size as a function of the dye-cavity-detuning. (b) Inverse second-order correlation time versus condensate number  $\bar{n}$  for five effective reservoir sizes  $M_{\text{eff}}$ . The curves exhibits minima at  $\bar{n}_{\text{min}} = (M_{\text{eff}})^{1/2}$ , which highlight the crossover point from grand-canonical ( $\bar{n} < \bar{n}_{\text{min}}$ ) to canonical statistics ( $\bar{n} > \bar{n}_{\text{min}}$ ). ( $\hat{B}_{21} = 10^4 \text{ s}^{-1}$ ,  $M = 10^9$ .)

can be recast as a function of experimental parameters:

$$\frac{1}{\tau_c^{(2)}} = \hat{B}_{21} \left[ \frac{M}{\bar{n}(1 + e^{-\frac{\hbar\Delta}{k_B T}})} + \bar{n}(1 + e^{\frac{\hbar\Delta}{k_B T}}) \right]. \quad (51)$$

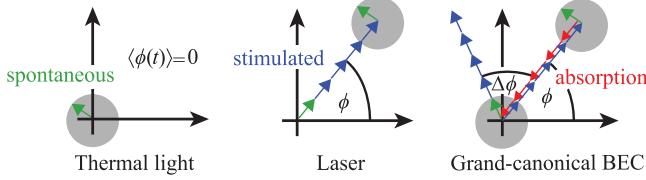
Around the crossover temperature  $T_x$  from grand-canonical to canonical ensemble conditions, the second-order correlation rate exhibits a piecewise scaling

$$\frac{1}{\tau_c^{(2)}} = \begin{cases} \frac{M}{\bar{n}(\hat{B}_{12} + \hat{B}_{21})}, & T \gg T_x \ (\bar{n}^2 \ll M_{\text{eff}}) \\ 2\bar{n}(\hat{B}_{12} + \hat{B}_{21}), & T = T_x \ (\bar{n}^2 = M_{\text{eff}}) \\ \bar{n}(\hat{B}_{12} + \hat{B}_{21}), & T \ll T_x \ (\bar{n}^2 \gg M_{\text{eff}}), \end{cases} \quad (52)$$

where we have introduced the effective reservoir size

$$M_{\text{eff}} = \frac{M}{2} \left[ 1 + \cosh\left(\frac{\hbar\Delta}{k_B T}\right) \right]^{-1}. \quad (53)$$

Figure 9(a) illustrates the variation of the effective reservoir size as a function of the dye-cavity-detuning, and figure 9(b) gives a plot of  $\gamma$  versus  $\bar{n}$  for various reservoir sizes. For a specific  $M_{\text{eff}}$  the inverse correlation time decays in the grand-canonical regime ( $g^{(2)}(0) \rightarrow 2$ ) with increasing condensate number, until it reaches a minimum at  $\bar{n}_{\text{min}} = \sqrt{M_{\text{eff}}}$ . In the canonical regime ( $g^{(2)}(0) \rightarrow 1$ ), the fluctuation rate exhibits the opposite behaviour growing linearly with increasing photon numbers. This analytic prediction is confirmed by numerical Monte Carlo simulations, see figure 12(b) in section 5. To exemplify the order of magnitude of  $\gamma$ , we give an estimate based on the typical experimental parameters discussed near the statistics crossover. For a condensate wavelength  $\lambda_c = 580$  nm, corresponding to a dye-cavity-detuning  $\hbar\Delta = -5.3k_B T$  (Rhodamine 6G) and Einstein coefficients  $\hat{B}_{12} \simeq 170 \text{ s}^{-1}$  and  $\hat{B}_{21} \simeq 3.4 \times 10^4 \text{ s}^{-1}$ , one obtains in the presence of  $M = 10^{10}$  molecules an average condensate number of  $\bar{n}_{\text{min}} = M_{\text{eff}} \simeq 7000$  photons. From (52) a time scale for the intensity fluctuations  $\tau_c^{(2)} \approx 2 \text{ ns}$  is expected, which is close to the experimental observation.



**Figure 10.** (a) Phasor model for different light sources. While spontaneous emission on average does not develop a global phase, stimulated processes in a laser result in a macroscopic phase-stable light field. For the photon BEC, similarly a global phase  $\phi$  emerges. In the presence of large reservoirs, however, the condensate vanishes due to strong dye-mediated reabsorption and subsequently emerges with a broken symmetry  $\phi + \Delta\phi$ .

## 5. Phase coherence

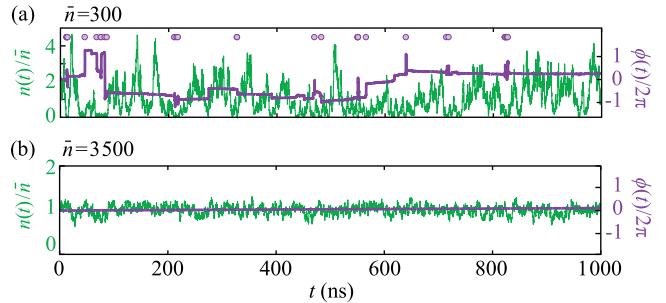
The phasor model allows a description of the temporal amplitude and phase evolution,  $\sqrt{n(t)}$  and  $\phi(t)$ , of an optical single-mode field containing  $n$  photons. As shown in figure 10, it can be a valuable tool to consider qualitative differences between light sources:

- (i) In a *thermal light source*, the superposition of spontaneously emitted photons with arbitrary phases leads to a random walk of the total phase and destructive interference prohibits the emergence of a macroscopically occupied state with a stationary phase [87]. Bose–Einstein photon statistics here gives rise to a most probable photon number  $n_{\max} = 0$ .
- (ii) Stimulated emission in a *laser* results in a macroscopic occupation of a single optical mode with a nearly stable phase. The mode selection is induced by engineering losses in all undesired modes; making laser emission in general an out-of-equilibrium phenomenon. Residual spontaneous emission into the laser mode causes an amplitude and phase uncertainty [114]. Poissonian photon number statistics with  $n_{\max} > 0$  lead to a vanishing probability to find zero photons  $\mathcal{P}_0 = 0$ .
- (iii) Finally, the phasor diagram is also helpful to illustrate the phase dynamics of a *BEC of photons*, where a reservoir induces large statistical, thermal-like (Bose–Einstein statistics) fluctuations of the condensate amplitude  $\sqrt{n(t)}$ . In this setting, the photon number eventually drops to  $n = 0$  and the subsequent spontaneous emission of a photon starts a cascade of stimulated processes forming a new macroscopically occupied ground mode. Due to the randomness of spontaneous emission we expect to observe the total phase of the wave function to change discretely in the course of time.

### 5.1. Phase dynamics of the wave function

Based on the rate (16) we perform Monte Carlo simulations of the photon number and phase evolution of the BEC based on the phasor model [97]. While stimulated absorption and emission do not alter the phase of the wave function, spontaneously emitted photons cause a Heisenberg-type phase diffusion [74, 115, 116].

The phasor of the condensate with  $n$  photons and phase  $\phi$  is described by the complex number  $\sqrt{n}e^{i\phi}$ . Following a spontaneous emission event with random phase  $\theta$  and



**Figure 11.** Simulation of condensate number  $n(t)/\bar{n}$  (green) and phase evolution  $\phi(t)$  (purple) in the presence of a particle reservoir  $M_{\text{eff}} = 2.5 \times 10^5$ . (a) Under grand-canonical conditions, large number fluctuations occur accompanied by discrete phase jumps at points, when the photon number drops to zero (top circles). (b) For larger condensate sizes (canonical), the fluctuations are damped out. No phase jumps occur as attributed to the vanishing probability to find zero photons for Poissonian statistics. ( $\Delta = 0k_{\text{B}}T$  and  $M = 10^6$ .)

amplitude  $\sqrt{1}$  the phasor is modified to

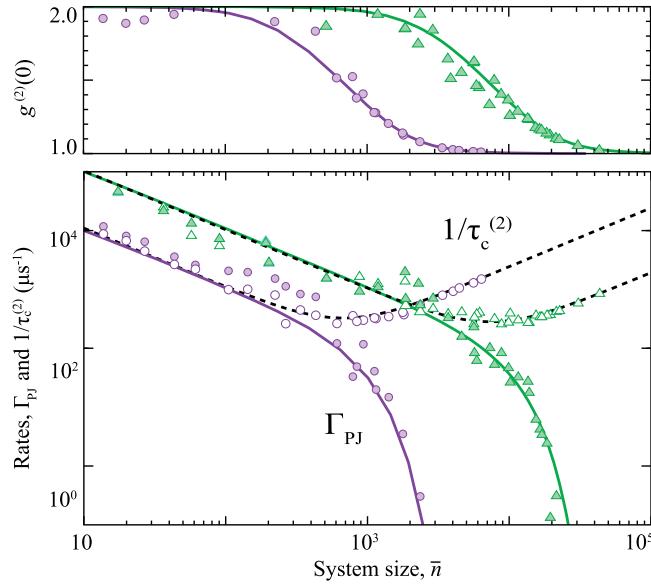
$$\sqrt{n + \Delta n} e^{i(\phi + \Delta\phi)} = \sqrt{n} e^{i\phi} + (\sqrt{n + 1} - \sqrt{n}) e^{i\theta}, \quad (54)$$

which corresponds to a length change  $\Delta n = 1 + 2\sqrt{n} \cos \theta$  and phase rotation  $\Delta\phi = \sin \theta / \sqrt{n}$ . The spontaneous phase becomes relevant for small photon numbers. Notably, the phase rotation describes only relative changes of the phase and does not apply to the case  $n = 0$ , when the randomly selected phase  $\theta$  breaks the symmetry to determine the overall phase of a re-emerging BEC.

Figure 11(a) shows a Monte Carlo simulation of the time evolution of the (normalised) occupation number and corresponding phase for a fluctuating BEC coupled to a reservoir that is compatible with grand-canonical statistics,  $\bar{n} < \sqrt{M_{\text{eff}}} = 500$ . The data reveal discrete phase jumps at points when no photons are present in the ground mode. For the same reservoir, figure 11(b) gives the number and phase dynamics of a BEC in the canonical ensemble, with  $\bar{n} \simeq 3500 > \sqrt{M_{\text{eff}}}$ . Due to its finite size the reservoir starts to saturate and the number fluctuations get damped. Notably, the zero-photon-probability  $\mathcal{P}_0$  vanishes, such that discrete phase jumps are suppressed. To quantify the temporal phase stability of the condensate, we introduce the phase jump rate  $\Gamma_{\text{PJ}} = \# \text{phase jumps} / \text{time interval}$ .

Moreover, our simulations of the photon number evolution yield the second-order correlation function  $g^{(2)}(\tau)$  and its associated timescale  $\tau_c^{(2)}$ . The phasor amplitude modulation that results from the fluctuating condensate population (time constant  $\tau_c^{(2)}$ ) is expected to affect the degree of first-order coherence  $g^{(1)}(\tau)$  via phase diffusion. Figure 11(a) (bottom), however, suggests that this effect is negligible in comparison to the large phase jumps. Experimentally, continuous phase drifts cannot be resolved with the applied interferometric method described in section 8.

Figure 12 shows  $\Gamma_{\text{PJ}}$  and  $1/\tau_c^{(2)}$  as a function of the average condensate number for two reservoirs, realised by varying  $\Delta$ . The data points are obtained from simulations similar to figure 11. In the grand-canonical regime ( $\bar{n} < \sqrt{M_{\text{eff}}}$ ), the phase jump and correlation rate decrease



**Figure 12.** Simulated autocorrelation  $g^{(2)}(0)$  (top), phase jump rates (bottom, filled symbols) and second-order correlation rates (bottom, open) versus  $\bar{n}$  for two effective reservoir sizes (violet circles and green triangles). The agreement of  $\Gamma_{\text{PJ}}$  with  $\hat{B}_{12}M\mathcal{P}_0$  (solid line) supports the assumption that phase jumps emerge when the condensate population vanishes. First-order coherence is enhanced as the photon statistics merges from being Bose–Einstein-like to Poissonian, which occurs at the  $1/\tau_c^{(2)}$  minimum at  $\bar{n} = \sqrt{M_{\text{eff}}}$ , where  $g^{(2)}(0) \simeq \pi/2$  (top) in good agreement with the analytical prediction (dashed line). ( $\hat{B}_{12} = 1000 \text{ s}^{-1}$ ,  $M = 10^9$ ,  $\hbar\Delta = \{-2.8; -7.7\}k_{\text{B}}T$ .)

simultaneously with increasing system size. At the crossover to canonical statistics ( $\bar{n} \geq \sqrt{M_{\text{eff}}}$ ), in figure 12 discernible by the autocorrelation value  $g^{(2)}(0) \approx \pi/2$  as well as the minimum of  $1/\tau_c^{(2)}$  (dashed line), both time scales separate. Beyond the minimum, we find the phase jumps to be more strongly suppressed, while an increase in the rate of second-order correlations is revealed in good agreement with the analytical prediction (48). This separation of coherence times is evident for both reservoirs in their respective crossover regions; the larger the reservoir, the further we find the phase jumps to persist in the condensed region. Strikingly, in this regime the photon condensate is expected to exhibit phase coherence despite large statistical number fluctuations characterised by  $g^{(2)}(0) > 1$ .

### 5.2. Phase jump rate

Our numerical findings suggest that the phase jump rate of a fluctuating condensate  $\Gamma_{\text{PJ}}$  correlates with the probability  $\mathcal{P}_0$  to find zero photons in the ground state. To quantify this, we assume that discrete phase changes occur only in the absence of photons from the cavity ground state at a rate  $\Gamma_{\text{PJ}}^0 = \mathcal{P}_0/\tau_0$ . Here,  $\tau_0$  labels a characteristic system time scale, i.e. the average time that a zero-photon-state exists in the cavity. Following a statistical fluctuation to a zero-photon-state, a certain time passes by until the condensate builds up with a new macroscopic phase, which is given by the inverse rate for spontaneous emission of a photon at the cutoff frequency. For

$n = 0$ , the rate (16) depends only on the Einstein coefficient  $\hat{B}_{21}$  and the number of excited dye molecules  $M_{\uparrow}$ , and thus  $1/\tau_0 = \hat{B}_{21}M_{\uparrow}$ . For the steady-state with  $\bar{n} \gg 1$ , see (19), one further obtains  $\hat{B}_{21}M_{\uparrow} \simeq \hat{B}_{12}M_{\downarrow}$ . With the typically fulfilled  $M_{\downarrow} \simeq M$ , we have

$$\Gamma_{\text{PJ}}^0 = B_{12}M\mathcal{P}_0. \quad (55)$$

As  $\mathcal{P}_0$  is determined by the photon number statistics, we consider the limiting cases: for a large reservoir (grand-canonical statistics), see (36), Bose–Einstein photon statistics gives  $\mathcal{P}_0 = 1/(\bar{n} + 1) \simeq 1/\bar{n}$ . For small reservoirs (canonical statistics), the crossover to Poissonian statistics leads to a strong suppression of the zero-photon probability:

$$\Gamma_{\text{PJ}}^0 = \frac{\hat{B}_{12}M}{\bar{n}^{\alpha}}, \quad \alpha = \begin{cases} 1, & \text{BE \& Gaussian} \\ \infty, & \text{Poisson} \end{cases} \quad (56)$$

To quantify the scaling of  $\Gamma_{\text{PJ}}^0$  with  $\bar{n}$ , we have introduced the exponent  $\alpha$ , which interpolates between 1 and  $\infty$  when connecting grand-canonical and canonical ensemble conditions. In the Poissonian limit ( $\alpha \rightarrow \infty$ ), the condensate thus exhibits the usual phase coherence. Using the above ansatz, the simulated phase jump rates can be reproduced as shown in figure 12 (solid line). The values for  $\mathcal{P}_0$  were numerically calculated (section 4).

### 5.3. Thermodynamic limit: correlation times

Figure 12 illustrates the separation of time scales for phase and intensity fluctuations, which suggests that the coherence properties of a photon BEC differ fundamentally from those of a thermal light source, i.e. violating  $g^{(2)}(\tau) = 1 + |g^{(1)}(\tau)|^2$  [87]. Therefore, the question arises whether the separation remains relevant in the thermodynamic limit. Combining (48) and (56) yields the ratio of the correlation times

$$\begin{aligned} \frac{\tau_c^{(1)}}{\tau_c^{(2)}} &= \bar{n}^{\alpha-1} \left[ \frac{\hat{B}_{12}}{\hat{B}_{12} + \hat{B}_{21}} + \frac{\hat{B}_{12} + \hat{B}_{21}\bar{n}^2}{\hat{B}_{21}M} \right] \\ &= \bar{N}^{\alpha-1} \left[ K_1(T) + K_2(T) \left( \frac{\bar{N}}{\sqrt{M}} \right)^2 \right], \end{aligned} \quad (57)$$

where we have used  $\bar{n} = \bar{N}[1 - (T/T_c)^2]$  and included the temperature dependence in the constants  $K_{1,2}(T)$ . In the thermodynamic limit,  $\bar{N} \rightarrow \infty$ ,  $T_c = \text{const.}$  and  $\bar{N}/\sqrt{M} = \text{const.}$ , one expects the relative correlation times to scale with the parameter  $\alpha$ , which (like  $\bar{N}/\sqrt{M}$ ) determines the photon number distribution  $\mathcal{P}_n$  and  $g^{(2)}(0)$ . On the one hand, for a condensate in the grand-canonical regime ( $\alpha = 1$ ) at a temperature  $T_x \leq T < T_c$ , the dependence on the total particle number  $\bar{N}$  in (57) vanishes and the correlation times coincide also in the thermodynamic limit. On the other hand, Poisson-like ( $\alpha > 1$ ) and genuine Poisson statistics ( $\alpha \rightarrow \infty$ ) are expected to cause a divergence of the first-order coherence time with respect to the second-order correlation time. Despite the here relatively large condensate fluctuations with  $g^{(2)}(0) \simeq 1.57$  ( $1 < \alpha < \infty$ ), a separation of time scales for first- and second-order is predicted for the thermodynamic limit.

Similarly, the heuristic phase jump rate becomes

$$\Gamma_{\text{PJ}}^0 = K_3(T) \frac{M}{N^2} \frac{1}{N^{\alpha-2}}, \quad (58)$$

where  $K_3(T)$  denotes a temperature-dependent parameter, which does not change with the system size. We expect phase jumps to be fully suppressed in the thermodynamic limit only for Poissonian states with  $\alpha > 2$ .

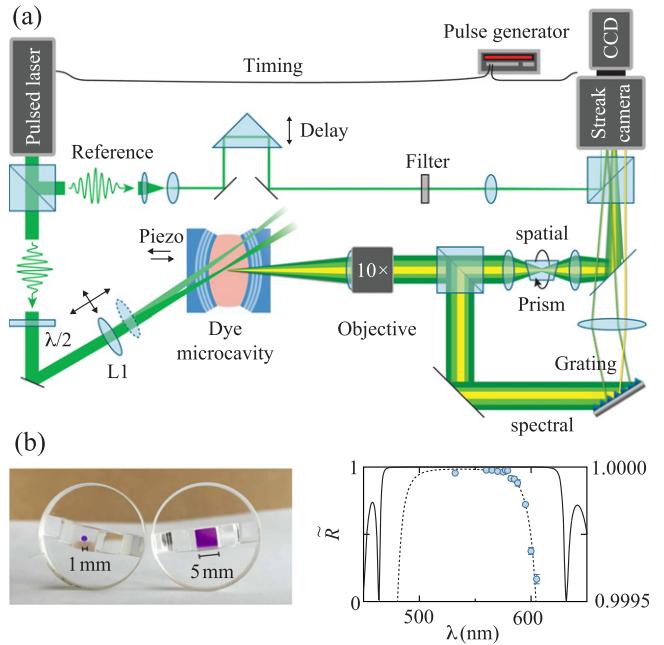
Physically, the phase jumps originate from the persistence of fluctuations to zero-photon-states also in the thermodynamic limit caused by Bose–Einstein-like statistics. Provided the particle reservoir is sufficiently large, the time scales for number and phase fluctuations remain coupled even upon extrapolation of  $\Gamma_{\text{PJ}}^0$  to the thermodynamic limit. On the one hand, the zero-photon-probability decays as  $\mathcal{P}_0 \propto \bar{n}^{-1}$  with increasing photon numbers ( $\alpha = 1$ ). On the other hand, this is counteracted by a quadratical increase of the molecule number required to conserve the photon statistics. Ultimately, this results in a larger phase jump rate.

## 6. Thermalisation dynamics

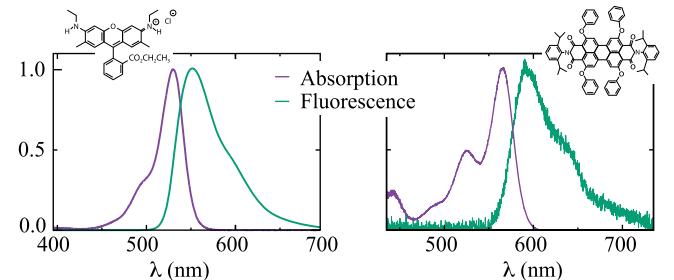
In this section, we discuss experimental results of time-resolved measurements of the spectral photon kinetics, which shed light on the thermalisation dynamics to the molecular heat bath. Our measurements are performed for photon numbers near the critical particle number  $N_c \simeq 90\,000$  ( $q = 8$ ). Moreover, the experiment enables a spatially and spectrally-resolved observation of the transition dynamics from out-of-equilibrium, laser-like states to thermal equilibrium BECs for  $\bar{N} \gg N_c$ .

### 6.1. Experimental scheme

Figure 13(a) shows a schematic of the experimental setup, which is comprised of the optical microcavity, the pump source and an analysis section [41]. For this time-resolved study, the dye-cavity is pumped under an angle of approximately  $42^\circ$  with respect to the optical axis using a picosecond pulsed laser. The cavity emission is detected by a streak camera in a spatially- and spectrally-resolved way. The microcavity is composed of highly-reflecting dielectric mirrors (CRD Optics, 901-0010-0550, radius of curvature  $R = 1\text{ m}$ ) with a maximum reflectivity  $\tilde{R} = 99.9988(2)\%$  around  $550\text{ nm}$ , while the bandwidth of a reflectivity beyond 99.98% extends over a broad range  $500\text{--}595\text{ nm}$ , see figure 13(b)<sup>5</sup>. At the maximum, the cavity finesse amounts to  $\mathcal{F} \approx 260\,000$ . To realise mirror separations in the micrometer range, the curved surface of one of the cavity mirrors is downsized in an in-house grinding process to  $\sim 1\text{ mm}$  diameter and equipped with prisms, see figure 13(b). The latter enables optical pumping of the dye reservoir under the above mentioned angle, which together with the appropriate polarisation maximises mirror transmission to approximately 80%. By adjusting the lens L1 shown in figure 13 we control both



**Figure 13.** (a) Experimental scheme for the time-resolved measurements of the spatial and spectral thermalisation dynamics. The microcavity is pumped with a pulsed laser beam and the emission imaged spatially and spectrally onto a streak-camera system. (b) Picture of the prepared cavity mirrors (left) and spectral mirror reflectivity  $\tilde{R}$  [117].



**Figure 14.** Normalised absorption and emission spectra of Rhodamine 6G and Perylene red with structure formulae.

pump spot position and diameter  $d$  in the cavity plane, in order to initially excite the dye medium in a spatially homogeneous ( $d \sim 500\text{ }\mu\text{m}$ ) or localised ( $\sim 20\text{ }\mu\text{m}$ ) way. For comparison, the spatial extent of the ground mode is  $d_0 \approx 15\text{ }\mu\text{m}$ , whereas the thermal cloud covers a region of a few hundred  $\mu\text{m}$  [55]. For a variation of the condensate wavelength, the cavity length can be piezo-tuned over a total length of  $25\text{ }\mu\text{m}$ . This allows us to actively stabilise the condensate wavelength with an accuracy  $\delta\lambda \simeq 0.2\text{ nm}$  at 10 Hz bandwidth, which compensates for long-term thermal or mechanical drifts. As dye materials we use Rhodamine 6G and Perylene red solutions of concentrations between 0.1 and  $5\text{ mmol l}^{-1}$ , see the spectra in figure 14 and table 3 for an overview of relevant properties. We expect the thermalisation time to be close to the reabsorption time and use dye concentrations of  $0.1\text{ mmol l}^{-1}$  to perform our time-resolved studies of the thermalisation dynamics.

<sup>5</sup> Obtained from cavity-ring-down measurements using a dye laser tuned to 560–605 nm [117].

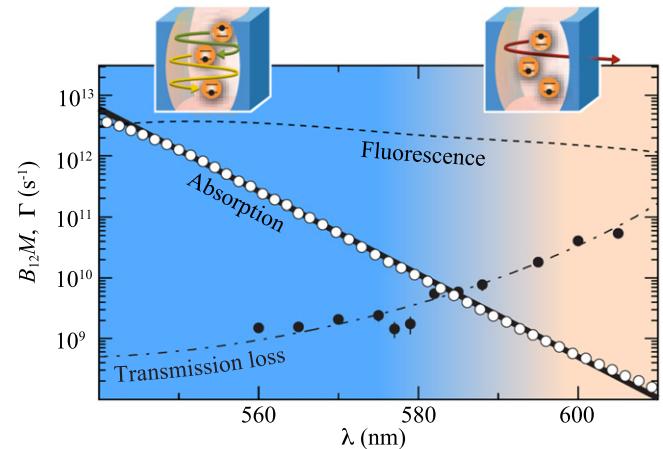
**Table 3.** Properties of the used dye media Rhodamine 6G (Rh6G) (*Radiant Dyes*) solved in ethylene glycol and Perylene (PDI) red (*Kremer Pigmente*) solved in inviscid paraffin oil. The absorption cross section follows from  $\sigma = 3.82 \times 10^{-21} \varepsilon$  (in units of  $\text{cm}^2$ ) [83] and the reabsorption time from  $\tau_{532\text{nm}} = (\rho \sigma_{532\text{nm}} c)^{-1}$ ,  $\rho = 1 \text{ mmol l}^{-1}$ .

	Rh6G	PDI red	References
$m$	479.02	1079.24	$\text{g mol}^{-1}$
$\Phi$	95	96	%
$\lambda_{\text{zpl}}$	545	585	nm
$\omega_{\text{zpl}}/2\pi$	550	513	THz
$\varepsilon_{532\text{nm}}$	114000	18000	[118]
$\sigma_{532\text{nm}}$	43	6.9	$10^{-17} \text{ cm}^2$
$\tilde{n}_0$	1.43	1.48	
$\tau_{532\text{nm}}$	0.13	0.8	ps

To initialise the dye medium in a time-resolved way a mode-locked Nd:YAG pulse laser (EKSPOLA PL2201) near 532 nm with 47  $\mu\text{J}$  pulse energy and 15 ps pulse length at 100 Hz repetition rate is at our disposal. Both its spatial and temporal intensity profile of the pump beam pulse are Gaussian. The laser system acts as the clock source for the experimental setup with electronic trigger noise around 100 ps. To obtain picosecond temporal resolution, we must therefore simultaneously detect the pump pulse and correct for its temporal jitter, which is achieved by directing part of the laser emission through a variable delay path onto the streak camera entrance slit. Subsequent to a pump pulse, the divergent microcavity emission is collimated by a 10 $\times$  long-working-distance objective (Mitutoyo M-Plan Apo 10 $\times$ ) and split into two beams. One part of the light is directed onto a diffraction grating (600 rules  $\text{mm}^{-1}$ ), and the spectrally dispersed light is focussed on the streak camera entrance slit with a width of 1.5 cm and 30  $\mu\text{m}$  height. In the second optical path, a telescope images the photon gas onto a dove prism (Thorlabs PS992M-A), which rotates the spatial ( $x$ ,  $y$ ) coordinates around the optical axis to align the emission with the entrance slit. The streak camera (Hamamatsu C10910) offers the time-resolved investigation in windows of {50; 20; 10; 5; 2; 1; 0.5; 0.2; 0.1} ns with a temporal resolution of 1% of the time range at 1% detection efficiency. The data acquisition for all measurements is performed in a photon counting mode.

## 6.2. Spectral thermalisation dynamics

First, we focus on the spectral thermalisation dynamics of the photon gas. For this, we realise different coupling strengths to the molecular heat bath and different loss rates due to mirror transmission by variation of the cutoff wavelength  $\lambda_c = \{601; 585; 577; 571\} \text{ nm}$ . Additionally, we control the reabsorption by using different dye concentrations  $\rho = \{0.1; 1\} \text{ mmol l}^{-1}$  (Rhodamine 6G). Figure 15 gives the measured spectral profiles of absorption, emission and loss rates. In the shown wavelength range, the fluorescence is approximately constant, whereas the absorption rate exhibits an exponential decay with increasing wavelength; their relative scaling confirms the validity of the Kennard–Stepanov



**Figure 15.** Rates for absorption  $\hat{B}_{12}M$ , fluorescence and photon loss  $\Gamma$  versus wavelength for Rhodamine 6G dye,  $1 \text{ mmol l}^{-1}$ , and CRD mirrors. The fluorescence (dashed line, normalised to absorption maximum) is approximately constant over the shown spectral range. For  $\lambda < 590 \text{ nm}$ , the photon dynamics is dominated by the exponential scaling of the reabsorption rate (open circles) that has been fitted with  $\propto \exp[hc(\lambda - \lambda_{\text{zpl}})/k_B T]$  yielding  $T = 308(14) \text{ K}$  (solid line). Here, a thermal equilibrium state is expected to emerge (blue region). Increased losses from mirror transmission (filled circles, dashed-dotted line) lead to dissipative dynamics for larger wavelengths (orange region).

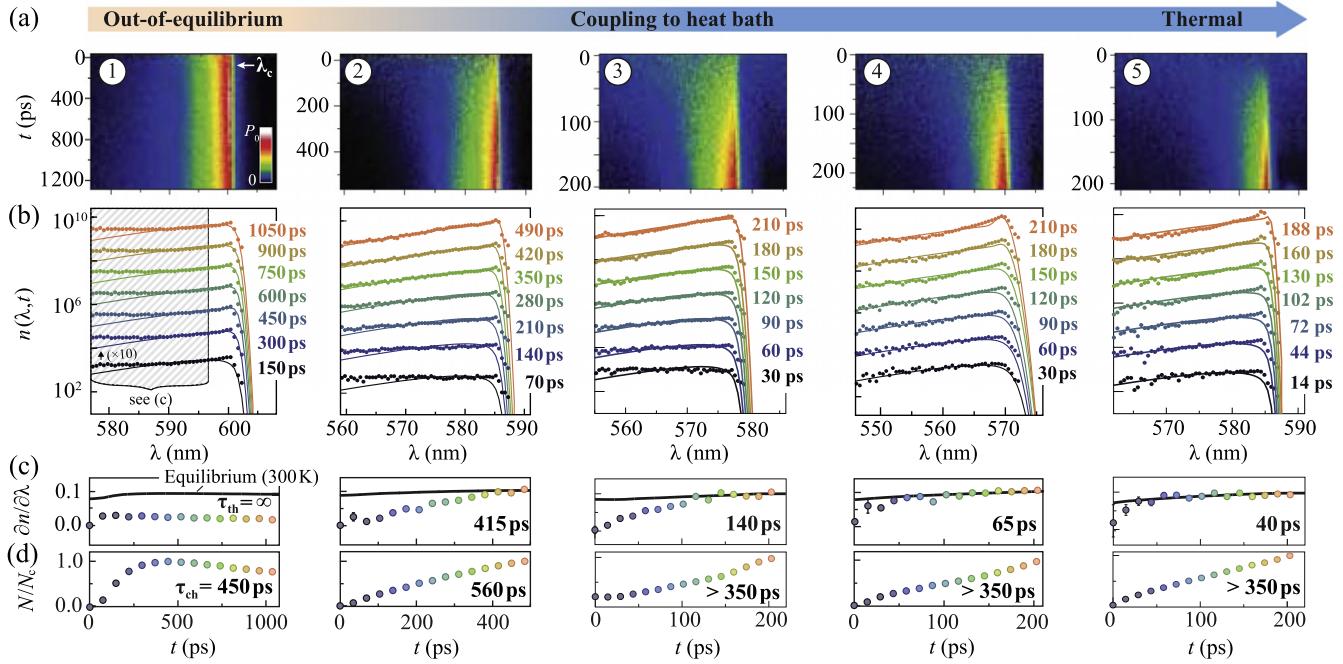
ratio for the used dye, see (2). In contrast, the photon loss by mirror transmission increases with  $\lambda$ , suggesting incomplete thermalisation for  $\lambda_c \geq 580 \text{ nm}$ . For  $\lambda_c = 580 \text{ nm}$ , however, we expect the photon gas to acquire a thermal state within its cavity lifetime.

The starting point for the measurement is a spatially homogeneous excitation of the dye medium using a broad pump beam ( $2w_0 = 500 \mu\text{m}$ ), which minimises any gradients in the excitation level of the medium at  $t = 0$ , realising well-defined initial conditions in chemical equilibrium. Figure 16(a) gives line-normalised, false-colour streak camera traces showing the evolution of the spectral mode occupation. Here, we define  $t = 0$  as the time when the first fluorescence photons are detected, see  $N/N_c(t)$  in figure 16(d). From left to right, we successively increment  $\lambda_c$  to gradually decouple the photon gas from the heat bath. All spectral distributions are weighted with the spectral mirror transmission coefficient. Individual excited modes, which are spaced by 42 pm ( $\Omega/2\pi = 37 \text{ GHz}$ ), are not resolved due to limited spectral resolution of the diffraction grating of <sup>6</sup> 1 nm. The recorded data span a total spectral range  $\Delta\lambda = 25 \text{ nm}$  ( $\Delta E = 3.5k_B T$ ), which is expected to contain the following fraction of photons:

$$\bar{N}_{\text{exp.}} = \int_0^{3.5k_B T} \frac{2(u/\hbar\Omega + 1)}{e^{(u-\mu)/k_B T} - 1} du \approx 0.93\bar{N}. \quad (59)$$

Our experimental data thus provides reliable information about the degree of thermalisation of the photon gas. In all measurements, we choose the laser power to be such that a macroscopic ground state occupation emerges at the end of the detection

<sup>6</sup> A high-resolution spectrum is shown in figure 22.



**Figure 16.** Thermalisation dynamics of the photon gas for increased heat bath coupling. (a) Streak camera traces (line normalised) and (b) extracted spectra at different times along with 300 K-Bose–Einstein distributions (solid lines). (c) By comparing the measured spectral slope (hatched area in (b)) with its equilibrium counterpart (line) the thermalisation time can be quantified. It reduces as the coupling to the heat bath is enhanced (from left to right). (d) The temporal evolution of the total power of the cavity emission (normalised to  $N_c$ ) indicates the time scale for chemical equilibration between photons and dye molecules. ( $\lambda_{c,1-5} = \{601; 585; 577; 571; 585\}$  nm, Rhodamine 6G  $\rho_{1-4} = 0.1$  mmol l<sup>-1</sup>,  $\rho_5 = 1.0$  mmol l<sup>-1</sup>). Reprinted figure with permission from [41]. Copyright (2015) by the American Physical Society.

window, which allows us to calibrate the photon number at arbitrary times  $N(t)$  with respect to its asymptotic value, i.e. the critical photon number  $N(t \rightarrow \infty) \equiv N_c \approx 90\,000$ . Therefore, we compare our spectral data with thermal equilibrium Bose–Einstein distributions at 300 K (solid lines) with a chemical potential that satisfies the total photon number.

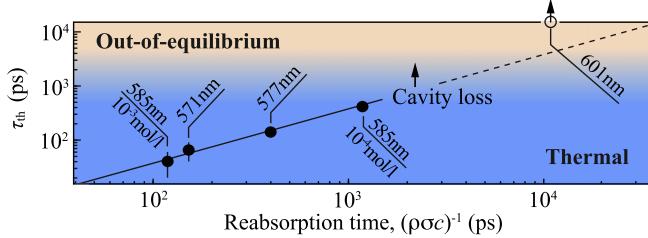
For weak dye reabsorption and large cavity losses,  $\lambda_c = 601$  nm (figure 16, left), the spectral wing (hatched) deviates from its equivalent in equilibrium for all observed times. The photon gas fails to thermalise during its microcavity lifetime. However, as the absorptive coupling rate to the molecule bath is enhanced,  $\lambda_c \leq 585$  nm, (figure 16, columns 2 to 5), we observe a thermalisation process that redistributes the photon energies, transforming the out-of-equilibrium distribution to a room temperature spectrum. The characteristic thermalisation time  $\tau_{th}$  can be quantified by the spectral slope  $\partial n(\lambda, t)/\partial \lambda$ , as illustrated in figure 16((c), circles), which in the presence of thermalisation converges to the equilibrium spectral slope (solid line). We observe  $\tau_{th} = \{415; 140; 65; 40\}$  ps defined as the time when the relative deviation between measured and equilibrium spectral slope is less than 1%. Figure 16(d) shows the temporal increase of the total cavity emission, revealing the gradual establishment of chemical equilibrium between photons and dye molecules. Notably, the time scales for either thermal or chemical equilibration differ (section 3). Even if the total photon is non-stationary, the spectral profile of the photon gas present in the microcavity can already be thermally distributed. In figure 17 we plot the measured

thermalisation times as a function of the free absorption time in the medium  $(\rho\sigma(\lambda)c)^{-1}$ , which follows a linear scaling  $\tau_{th} = 0.37(5) \cdot (\rho\sigma(\lambda)c)^{-1}$ . Indeed, the photon gas equilibrates due to an energy exchange with a heat bath at a rate that can be tuned via the reabsorption. If the thermalisation time exceeds the photon lifetime in the cavity,  $\tau_{th} > \tau_{res} \approx 500$  ps, the photons constitute an out-of-equilibrium ensemble.

### 6.3. Bose–Einstein condensation dynamics

We turn our attention to the temporal photon dynamics subject to (i) *spatially inhomogeneous* and (ii) *strongly inverting* pump excitation of the dye medium. As before, we investigate the dynamics for different coupling rates to the molecular heat bath  $\rho\sigma(\lambda)c$  and resonator losses  $\Gamma(\lambda)$ , realised by varying the cutoff wavelength. In the out-of-equilibrium regime,  $\Gamma(\lambda) > \rho\sigma(\lambda)c$ , the nonequilibrium state of the dye medium manifests itself in transient multimode laser operation. When coupling the photons to a heat bath,  $\Gamma(\lambda) < \rho\sigma(\lambda)c$  paves the way for the photon gas to thermal equilibrium and give rise to the emergence of BEC.

Figure 18(a) indicates the experimental sequence to realise initial conditions far from equilibrium. We focus the pump beam to a diameter  $2w_0 = 80 \mu\text{m}$  and position it at  $(x = 150 \mu\text{m}, y = 0)$  transversally displaced from the optical axis in the microcavity plane. As a result, the spatially inhomogeneous density  $\rho_{\uparrow}(x)$  decays by emission of (initially) spontaneous photons into excited transverse cavity

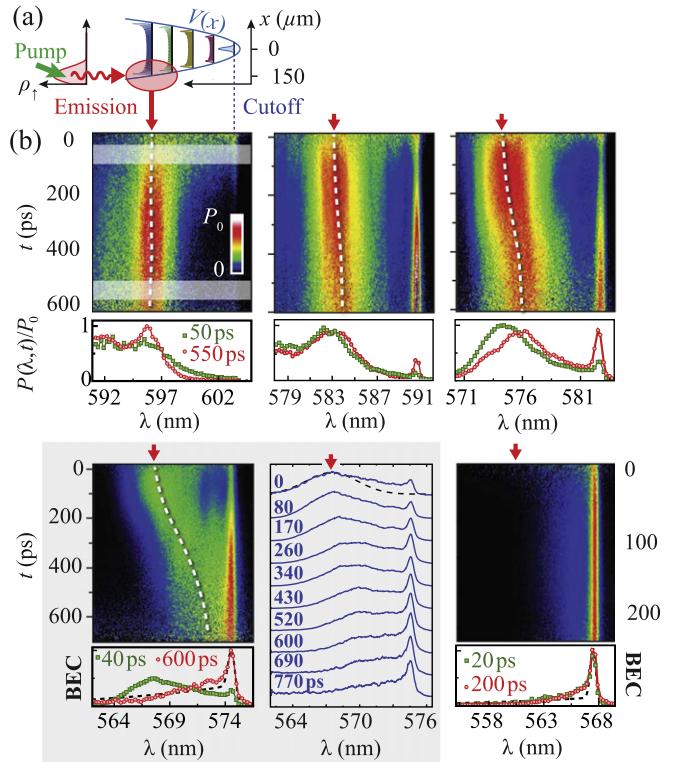


**Figure 17.** Observed thermalisation time  $\tau_{\text{th}}$  (circles) versus reabsorption time in the dye solution. For  $\tau_{\text{th}} < \tau_{\text{res}}$  the photon gas acquires a thermal state (blue); otherwise it remains an out-of-equilibrium system. Reprinted figure with permission from [41]. Copyright (2015) by the American Physical Society.

modes that overlap most with the pumped region. These eigenstates of the harmonic oscillator potential are at higher energies (lower wavelengths) than the transverse ground state  $\hbar\omega_c$  ( $\lambda_c$ ). Due to the sub-nanosecond time scales of the radiative processes, the comparatively slow effect of spatial diffusion of molecules can be safely neglected [124]. Following the initial photon emission, the photon gas kinetics depends critically on both the dye reabsorption and the cavity loss rates.

The first data set in figure 18(b) shows the spectral photon evolution in the weakly reabsorbing regime near  $\lambda_c = 603$  nm. The streak camera time traces have been line-normalised to clarify the spectral redistribution of the photons. Here, the reabsorption time  $(\rho\sigma(\lambda_c)c)^{-1} = 5.8$  ns (corresponding to  $\tau_{\text{th}} = 2.3$  ns) exceeds by far the average photon storage time in the cavity  $\tau_{\text{res}}(\lambda_c) = 18$  ps, such that no equilibrium distribution emerges. Instead, the optical feedback onto the inverted active medium causes stimulated amplification of the light field in the excited modes around  $\lambda_{\text{max}} = 595.7$  nm after approximately 100 ps and the maximum of the emission is maintained throughout the entire detection window. At  $r = 150$   $\mu\text{m}$  the resonant wavelength deviates from the cutoff wavelength by  $\Delta\lambda(r) = \lambda_c - \lambda(r) = 2nr^2/qR = 8$  nm, which agrees with the observed value of 7.3 nm. In further measurements, see figure 18(b), we successively enhance the coupling to the heat bath by reducing  $\lambda_c$ . Accordingly, we observe a more and more accelerated spectral redistribution of the light towards an equilibrium distribution. This is a consequence of fast reabsorption processes, which chemically equilibrate any gradients in the density of the ground and excited state molecules. Due to the harmonic trapping potential, this light-induced diffusion is directed towards transverse modes with lower energies than the modes overlapping with the pump beam region. Strikingly, the ground state becomes macroscopically occupied for data with shorter cutoff wavelength, and for  $\lambda_c = 574$  and 567 nm a BEC with thermally occupied excited states forms. In the case of  $\lambda_c = 567$  nm, the rapid thermalisation prevents the detection of any nonequilibrium emission at the given temporal resolution.

Using a spatially-selective photon injection technique to prepare a photon gas far from equilibrium, our measurements have demonstrated that a high-density (critical) photon gas thermalises to a Bose–Einstein condensate provided that the



**Figure 18.** Spectral condensation dynamics following an off-centre pump pulse for increasing thermal contact to the heat bath. (a) The pump pulse initialises an inhomogeneous excited molecule density  $\rho_{\uparrow}$ , which decays by stimulated emission into spatially overlapping modes. (b) Streak camera images (top) with spectral cuts (bottom) shortly after the photon injection (green squares) and at the end of the detection window (red circles). While for weak thermal contact (top, left) no thermalisation is observed, the enhanced reabsorption leads to a partial relaxation (top, middle and right) of the photon emission towards larger wavelengths. Finally, the photon gas fully thermalises to a Bose–Einstein distribution (bottom row). The grey-shaded data set highlights the thermalisation dynamics with cuts in the dynamical transition region. (Rhodamine 6G 0.25 mmol l<sup>-1</sup>,  $\lambda_c = \{603; 590; 582; 574; 567\}$  nm,  $\tau_{\text{th}} = \{2300; 388; 124; 38; 13\}$  ps, cavity lifetime  $\tau_{\text{res}} = \{18; 83; 165; 303; 500\}$  ps). Reprinted figure with permission from [41]. Copyright (2015) by the American Physical Society.

coupling to the heat bath is sufficiently strong. In the opposite limit, the high-density photon gas resembles an out-of-equilibrium state similar to a multimode laser. In contrast to the homogeneously pumped protocol (section 6.2), the photon thermalisation dynamics is not universal but depends crucially on the initial conditions of the pumped dye medium, in excellent agreement with our numerical simulations, see section 6.5.

#### 6.4. Spatial photon kinetics

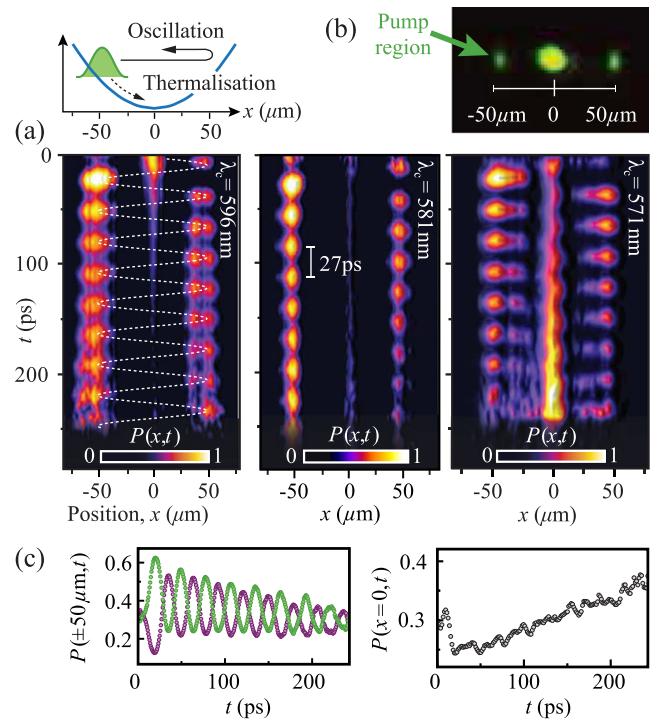
We focus on the *spatial* condensation dynamics subsequent to an inhomogeneous inversion of the dye medium. For this, a tightly focussed pump beam ( $2w_0 \simeq 27$   $\mu\text{m}$ ) irradiates the dye microcavity spatially displaced by 50  $\mu\text{m}$  from the position of the trap minimum. To analyse the spatial intensity

distribution, a real image of the cavity plane is projected onto the streak camera.

Figure 19(b) shows a typical CCD camera image of the average cavity emission. Besides the emission from the trap centre ( $x = 0, y = 0$ ), two bright spots are visible: the first one near the pumping region at  $(-50 \mu\text{m}, 0)$ , the second one at  $(50 \mu\text{m}, 0)$ , i.e. mirrored respectively to the trap centre. A time-resolved measurement (figure 19, line-normalised) of the intensity distribution along the  $x$ -axis yields an explanation for the centro-symmetric emission: following the inhomogeneous dye excitation an optical wave packet forms, which oscillates in the harmonic potential with reversal points that determine the observed emission spots. The observed oscillation period  $T = 27 \text{ ps}$  shows excellent agreement with the expected inverse trap frequency  $2\pi/\Omega \simeq (37.1 \text{ GHz})^{-1}$  (see table 1). Moreover, the wave packet emerges within only a few picoseconds. As this is considerably faster than the spontaneous decay time of the Rhodamine molecules ( $4 \text{ ns}$ ), the dynamics are driven by stimulated processes. The wave packet dynamics can be understood as a coherent superposition of adjacent transverse eigenstates spaced by  $\hbar\Omega$ , i.e. with a fixed relative phase, in close analogy to a mode-locked laser with an extremely high repetition rate. Oscillator modes that exhibit their maximum probability in proximity to the pumped region, experience maximum gain. Classically speaking, the velocity of the wave packet is minimised at the reversal points of the oscillation, maximising here the photon leakage rate out of the resonator. Quantum mechanically, this can be interpreted as constructive interference between multiple harmonic oscillator wave functions.

In the limit of weak reabsorption and large cavity losses, see figure 19((a), left) for  $\lambda_c = 596 \text{ nm}$ , the photon kinetics is determined by the highly excited oscillating out-of-equilibrium state throughout the measurement time of  $250 \text{ ps}$ . The visible residual initial population at small times is attributed to overlap of the pump beam with the ground mode at  $x = 0$ , which however quickly decays. The situation drastically changes, as the thermal contact to the heat bath is established by lowering  $\lambda_c = 581$  and  $571 \text{ nm}$  (figure 19(a), middle and right). During its oscillation, the wave packet traverses the enclosed dye volume, effectively equilibrating the initially inhomogeneous excitation level  $\rho_\uparrow/\rho_\downarrow(x)$  by multiple photon reabsorption events. Figure 20 shows corresponding numerical simulations. With advancing times, this effects a dynamical redistribution from the laser-like wave packet to a BEC. The damping of the coherent oscillations and the emergence of the macroscopic ground state in the presence of a thermal bath is shown in figure 19(c). Qualitatively, the measured photon kinetics is in good agreement with results from numerical simulations.

To conclude, our experimental study demonstrates that a thermal state of the photon gas in the dye-filled microcavity is imprinted by a molecular heat bath. In particular, we find that the efficiency of the thermal contact, i.e. the thermalisation rate, can be tuned systematically by the optical density of the dye solution. With regard to the canonical and grand-canonical statistical ensemble, the temperature of the dye solution



**Figure 19.** Mode-locked laser operation and BEC of photons. (a) Line-normalised spatial evolution of the cavity emission for different cutoff wavelengths. The pump beam excites an oscillating wave packet in the harmonic potential (top). For weak reabsorption (left), a stable mode-locked laser oscillation occurs (dashed line), where most light leaves the cavity at the reversal points. For stronger reabsorption (middle and right) the photons thermalise and a condensate emerges in the trap centre. (b) CCD camera image of the (average) emission. (c) Temporal variation of the detected relative intensity from (a, right) at the reversal points (left) and in the condensate mode (right). (Rhodamine 6G,  $\rho = 0.1 \text{ mmol l}^{-1}$ ,  $\lambda_c = \{596; 581; 571\} \text{ nm}$ ). Reprinted figure with permission from [41]. Copyright (2015) by the American Physical Society.

actually becomes an external parameter for the photon gas. Thermalisation induced by absorption and emission is a necessary prerequisite for the emergence of a photon BEC.

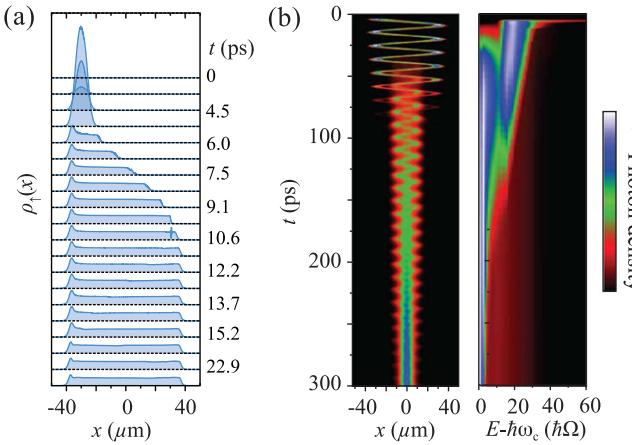
### 6.5. Numerical simulations

To gain deeper insight into the crossover from transient laser operation (*critical out-of-equilibrium gas*) to BEC (*critical equilibrium gas*), we perform numerical simulations of the photon dynamics in the microcavity [41]. Our phenomenological model relies on semiclassical one-dimensional rate equations and incorporates both the coupling of the photons to the optically active dye medium as well as their oscillatory movement in the harmonic trap. Independently, the results have been confirmed using a master equation model including coherences between photon modes [71].

Our approach is based on the equation of motion for the photon density:

$$\dot{\bar{n}}_i = \hat{B}_{21}^i \bar{\rho}_\uparrow (\bar{n}_i + \varepsilon_i) - (\hat{B}_{12}^i \bar{\rho}_\downarrow + \Gamma_i) \bar{n}_i - v_i \frac{\partial}{\partial x} \bar{n}_i. \quad (60)$$

Here,  $\bar{n}_i = \bar{n}_i(x, t)$  is the photon number density in the  $i$ th mode at position  $x$  and time  $t$  (averaged over many



**Figure 20.** Simulation of the thermalisation dynamics for off-centre pulsed excitation. (a) Density of excited molecules  $\rho_{\uparrow}(x)$  for given times after the pump pulse. The locally excited medium near  $x_0 = -30 \mu\text{m}$  induces the formation of a photon wave packet (see (b)), which homogenises  $\rho_{\uparrow}(x)$  in the course of its oscillation. (b) The temporal evolution of the spatial photon density (left) shows the oscillation of a mode-locked photon wave packet, which is damped out with time. After 250 ps, the photons have been redistributed to the cavity ground state at  $x = 0$ . Photon evolution versus photon energy  $E = (\hbar\omega - \hbar\omega_c)/\hbar\Omega$  (right), showing the emergence of a BEC with  $\bar{n}_0/\bar{N} \simeq 95\%$ . ( $\Omega/2\pi = 93 \text{ GHz}$ ,  $\lambda_c = 570 \text{ nm}$ ,  $\hbar\Delta \simeq -4k_B T$  at  $T = 300 \text{ K}$ ,  $\hat{B}_{12} = 1.3 \text{ kHz}$ ,  $\rho_{\uparrow}(x) + \rho_{\downarrow}(x) = 1.5 \times 10^8 \mu\text{m}^{-1}$ ,  $\Gamma_i = 0$ .)

realisations), the densities of molecules in ground and electronically excited state  $\bar{\rho}_{\downarrow,\uparrow} = \bar{\rho}_{\downarrow,\uparrow}(x, t)$ , and the rate coefficients for absorption and emission  $\hat{B}_{12,21}^i = \hat{B}_{12,21}(\omega_i)$  at the photon angular frequency  $\omega_i$ . Furthermore,  $\varepsilon_i = \varepsilon_i(x)$  denotes the density of a single photon in the  $i$ th mode,  $\Gamma_i = \Gamma(\omega_i)$  the cavity loss rate and  $v_i = v_i(x)$  the photon velocity field, which will be discussed in the following. Assuming a conserved excitation number  $X = n + M_{\uparrow}$ , one finds

$$-\frac{\partial}{\partial t} \bar{M}_{\downarrow} = \frac{\partial}{\partial t} \bar{M}_{\uparrow} = P - \sum_i \frac{\partial}{\partial t} \bar{n}_i, \quad (61)$$

where  $P = P(x, t)$  denotes the pump beam excitation. Heuristically, we consider a non-orthogonal set of optical modes consisting of coherent states  $|\alpha_i\rangle$  with amplitudes  $|\alpha_i| = \sqrt{u_i/\hbar\Omega}$ ,  $u_i = i \cdot \hbar\Omega$  and the trap frequency  $\Omega$ . The mode energy spectrum corresponds to the eigenenergies of the harmonic oscillator potential. In contrast to stationary eigenstates, coherent states allow us to model the oscillation of particles or wave packets in the trap. The normalised photon density  $\varepsilon_i(x)$  results from a temporal average

$$\varepsilon_i(x) = \frac{1}{T} \int_0^T |\langle x | \alpha_i(t) \rangle|^2 dt \quad (62)$$

over an oscillation period  $T = 2\pi/\Omega$ . By comparing the probability to find a particle within  $dx$ ,  $\varepsilon_i(x)dx$ , with the temporal portion of a half-period that the particle is present in this interval,  $dt/(T/2)$ , we define the photon velocity field

$$v_i(x) = dx/dt = \pm\Omega/\pi\varepsilon_i(x), \quad (63)$$

where the sign changes after each half-period. Neglecting losses,  $\Gamma_i = 0$ , the numerical results demonstrate that the

model reproduces asymptotically the analytic Bose–Einstein distributions and the critical particle number, see figure 20(b).

In analogy to the experiments described above, we simulate the photon thermalisation kinetics for initial out-of-equilibrium conditions realised by pumping the molecular medium with a Gaussian laser pulse  $P(x, t) \propto \exp[-(x - x_0)^2/2\sigma_x^2 - (t - t_0)^2/2\sigma_t^2]$  with duration  $\sigma_t = 1.5 \text{ ps}$  and waist  $\sigma_x = 7.5 \mu\text{m}$ . The pump pulse is positioned at  $x_0 = -30 \mu\text{m}$ , where it locally excites molecules within a few picoseconds, as indicated in figure 20(a). At this point, the chemical potentials of the photons and ground and excited state dye molecules exhibit strong gradients as visible in the spatially inhomogeneous dye excitation level. The cutoff wavelength  $\lambda_c = 570 \text{ nm}$  is chosen such that the photon gas couples efficiently to the molecules<sup>7</sup>.

Subsequent to the initialisation pulse, the simulations reveal the emergence of a high photon density in the pumped region, which reaches its maximum after only a few picoseconds. Owing to the trapping potential these photons are accelerated as a wave packet towards the trap minimum ( $x = 0$ ), see figure 20(b). During their oscillation the photons are quickly reabsorbed by the enclosed dye medium, which results in a homogeneous density of excited molecules within the region traversed by the wave packet after nearly half an oscillation period ( $T/2 = 5.4 \text{ ps}$ ) as visible in figure 20(a). The homogeneity of  $(M_{\uparrow}/M_{\downarrow})(x)$  is a prerequisite for chemical equilibrium among photons and molecules and the existence of a global chemical potential for the photon gas. Indeed, we find that the medium acquires a homogeneous state soon after the pump excitation, whereas the photon gas is still characterised by a non-thermal spectral distribution, see figure 20(c). It takes additional 250 ps until the laser-like wave packet has vanished and the photon energies are Bose–Einstein distributed with a macroscopic occupation of the ground mode.

## 7. Number statistics of condensed light

We describe measurements of the photon number statistics and second-order correlations of a photon BEC coupled to different-sized particle reservoirs. Our experiment gives access to canonical and grand-canonical statistical ensemble conditions, which are hallmarked by their particle number fluctuations: for small reservoirs (canonical), the photon statistics is Poissonian with small fluctuations,  $\delta n/\bar{n} = 1/\sqrt{\bar{n}} \approx 0$  (for  $\bar{n} \gg 1$ ), whereas large reservoirs (grand-canonical) support unusually large fluctuations of the condensate population,  $\delta n/\bar{n} = 1$ .

### 7.1. Experimental scheme

Figure 21 outlines the used experimental scheme. In contrast to the measurements of the thermalisation dynamics, a continuous pump and detection system is utilised. For all measurements, the microcavity is operated at  $q = 8$  and filled with either

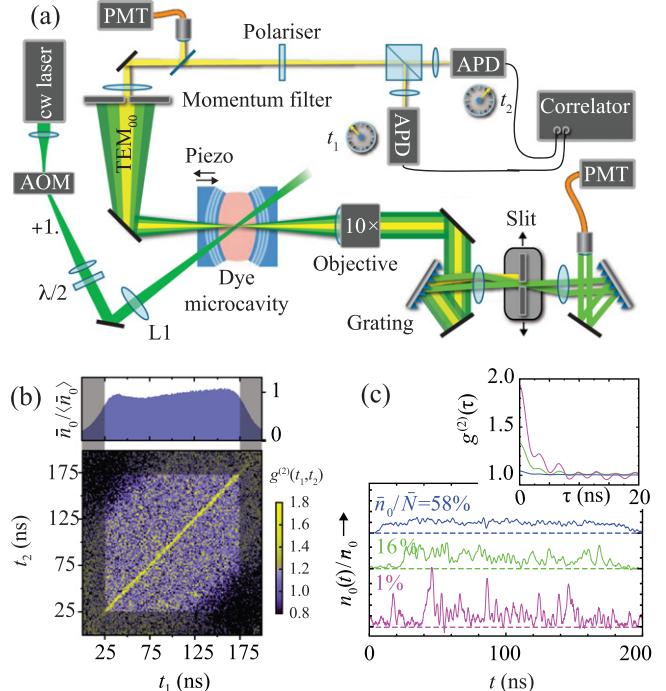
<sup>7</sup> The simulation parameters differ from experimental values for computational reasons. Cavity losses have been neglected.

Rhodamine 6G (ethylene glycol) or Perylene red (inviscid paraffin oil) solutions at varying concentrations. The dye medium is pumped by a frequency-doubled Nd:YAG laser (Coherent Verdi V8) near 532 nm, whose output power of up to 8 W is acousto-optically modulated (AOM) into 200 ns pulses at 200 Hz repetition rate, in order to reduce excitation of long-lived dye triplet states and to maintain condensate number constant throughout the pulse (figure 21(b), top). For the latter, the rf-signal driving the AOM is mixed with a temporally increasing voltage from a function generator (Tektronix AFG3252). Additionally, a voltage-controlled attenuator actively stabilises the condensate power (10 Hz bandwidth), which is separately detected by a photomultiplier. A  $f_{L1} = 400$  mm focal length lens focuses the pump beam to a diameter of  $2w_0 \simeq 150 \mu\text{m}$  into the microcavity plane to generate a photon gas. Here, the pump power controls the excitation level of the dye, as well as the chemical potential and the total number (and condensate fraction) of the photon gas. Any loss from the dye-microcavity-system is compensated by maintaining the pumping throughout the pulse.

To determine the condensate fraction  $\bar{n}_0/\bar{N}$ , we measure average photon spectra, see figure 22(a), in a 4f-spectrometer equipped with two diffraction gratings ( $2400 \text{ rules mm}^{-1}$ ) and two lenses with  $f = 100$  mm. A motion-controlled slit placed in the 2f-Fourier-plane performs a wavelength selection of the multimode light, which is detected using a photomultiplier (Hamamatsu H10721-210). Although its spectral resolution  $\Delta\lambda = 0.5 \text{ nm}$  precludes the measurement of individual transverse cavity modes spaced by  $\Delta\lambda \simeq 41 \text{ pm}$ , we confirm the solitary macroscopic occupation of the ground state with a double monochromator (LTB Demon) with 6 pm resolution, see figures 22(a) and (c).

The photon correlations of the BEC are detected in a Hanbury Brown–Twiss interferometer, while for the direct observation of the time-resolved fluctuations and photon statistics a photomultiplier is at our disposal, see figure 21. To measure the second-order correlations only of the condensate mode, the divergent cavity emission is first Fourier-filtered with a 5 mm iris in the far field approximately 850 mm behind the cavity.<sup>8</sup> The aperture acts as a transverse momentum filter to suppress contributions from excited modes: from the zero-point energy in the harmonic trapping  $\hbar\Omega$ , we can estimate the momentum uncertainty of the ground mode  $\Delta k_r = \sqrt{2m_{\text{ph}}\Omega/\hbar} \simeq 1.86 \times 10^5 \text{ m}^{-1}$ , which is much smaller than the longitudinal wave vector component  $k_z(0) = q\pi/D_0 \simeq 1.6 \times 10^7 \text{ m}^{-1}$ . Taking into account the quartz–air cavity interface ( $\bar{n}_{0,\text{Quartz}} \simeq 1.46$ ), the corresponding divergence angle  $\Theta = 0.95^\circ$  leads to a condensate diameter  $\sim 1.4 \text{ cm}$  at the momentum filter. Most of light in the first excited eigenstate ( $2\hbar\Omega$ , diameter  $\sim 2.0 \text{ cm}$ ) is expected to be blocked. After lifting the two-fold polarisation degeneracy of the photons, the transmitted light is equally split and directed onto two single-photon detectors (MPD PD5CTC, temporal resolution  $\Delta t \simeq 50 \text{ ps}$ , dead time  $\tau_{\text{PD}} \simeq 79 \text{ ns}$ ),

<sup>8</sup> The free propagation of the photon gas is equivalent to a free expansion of a harmonically trapped gas, a technique commonly used in ultra-cold atoms to infer the initial momentum distribution from the density distribution after a long time-of-flight [99].



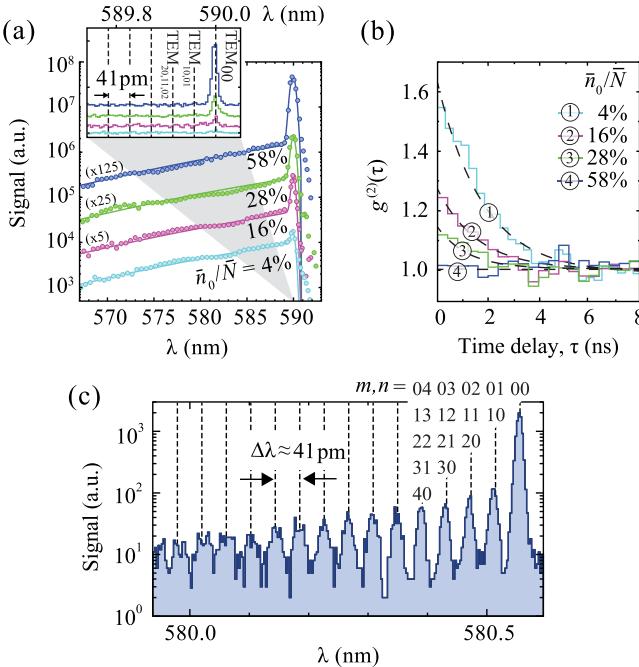
**Figure 21.** (a) Time-resolved measurements of correlations, number statistics and fluctuations of the photon BEC. The microcavity is pumped (quasi-)continuously and part of the cavity emission is spectrally analysed. In the far field, the condensate emission passes several filtering stages and its correlations are detected in a Hanbury Brown–Twiss interferometer. (b) Typical average condensate emission (top) and second-order correlation function  $g^{(2)}(t_1, t_2)$ . (c) Time evolution of the (normalised) condensate population measured with a single photomultiplier (PMT) for different condensate fractions  $\bar{n}_0/\bar{N}$ . *Inset:* corresponding autocorrelation functions with  $\tau = t_2 - t_1$ . Reprinted figure with permission from [40]. Copyright (2014) by the American Physical Society.

which are connected to an electronic correlation system (PicoQuant PicoHarp 300) that records and correlates photon detection events at times  $t_{1,2}$  with a resolution 60 ps. To avoid errors during the coincidence measurement caused by the dead time of the system ( $\tau_{\text{PicoHarp}} \simeq 90 \text{ ns}$ ), the condensate light is sufficiently attenuated to provide photon count rates around  $\sim 0.5$  photons/pulse ( $2.5 \times 10^6 \text{ photons s}^{-1}$ ) at each detector. Evaluation of the time histograms yields the second-order correlation function for the BEC

$$g^{(2)}(t_1, t_2) = \frac{\langle n_0(t_1) n_0(t_2) \rangle}{\langle n_0(t_1) \rangle \langle n_0(t_2) \rangle}, \quad (64)$$

where  $\langle \dots \rangle$  denotes a temporal average, see figure 21(b) for a typical data set. At  $t_1 = t_2$ , we find significant photon bunching,  $g^{(2)}(t_1, t_1) \approx 1.7$  (yellow diagonal), while for large time delays the photons are uncorrelated,  $g^{(2)}(t_1, t_2) \approx 1.0$ . Due to the nearly constant average photon number during the operation time (figure 21(b), top), the second-order correlations depend only on the relative time delay  $\tau = t_2 - t_1$ , and we hereafter only refer to the time-averaged correlation function  $g^{(2)}(\tau) = \langle g^{(2)}(t_1, t_2) \rangle_{t_2 - t_1 = \tau}$ .

Moreover, we monitor the time evolution of the condensate intensity in the same optical path, see figure 21 (top), relying on a photomultiplier (Hamamatsu H9305-01,



**Figure 22.** (a) The measured spectra for increasing condensate fractions (circles) are well described by 300 K Bose–Einstein distributions (solid lines). *Inset:* (Linear) high-resolution spectrum demonstrating the macroscopic occupation of the ground state only. All spectra have been vertically shifted for clarity. (b) Second-order correlation functions  $g^{(2)}(\tau)$  (①–③) exhibit photon bunching at short time delays  $\tau$ . ( $\lambda_c = 590$  nm,  $\hbar\Delta = -6.7k_B T$ ,  $\rho = 1$  mmol l<sup>-1</sup>, Rhodamine 6G) (c) mode-resolved spectrum of a Bose–Einstein condensed photon gas measured using a double monochromator. Reprinted figure with permission from [40]. Copyright (2014) by the American Physical Society.

$\Delta t \simeq 1.4$  ns, quantum efficiency  $\approx 10\%$ ) and a fast oscilloscope (Lecroy DDA 5005A, 5 GHz bandwidth). This allows us to resolve the number fluctuations, which occur on time scales around 2 ns; examples are given in figure 21(c). From the intensity traces  $I_0(t)$  we can equally reconstruct the second-order correlation function

$$g^{(2)}(\tau) = \frac{\langle I_0(t + \tau)I_0(t) \rangle_t}{\langle I_0(t) \rangle_t \langle I_0(t + \tau) \rangle_t}, \quad (65)$$

where  $\langle \dots \rangle_t = (T - \tau)^{-1} \int_0^{T-\tau} (\dots) dt$  denotes the temporal average of the pulse of duration  $T$ . We note, that despite consistent results for  $g^{(2)}(0)$ , the Hanbury Brown–Twiss interferometer is considered as the more reliable detection scheme for our purposes due to its high temporal resolution.

## 7.2. Time-resolved photon correlations

In a first step, we study the number correlations of different-sized photon BECs coupled to a particle reservoir of constant size by fixing the dye concentration  $\rho = 1$  mmol l<sup>-1</sup> (Rhodamine 6G) and the dye-cavity detuning  $\hbar\Delta = hc(\lambda_c^{-1} - \lambda_{\text{zpl}}^{-1}) = -6.7k_B T$  ( $\lambda_c = 590$  nm).

Figure 22(a) shows spectral distributions in the Bose–Einstein condensed phase hallmark by the macroscopically occupied ground mode and thermally populated excited states.

All condensate fractions  $\bar{n}_0/\bar{N} = \{4\%; 16\%; 28\%; 58\%\}$  and reduced temperatures  $T/T_c = \{0.98; 0.92; 0.85; 0.65\}$ , respectively, are obtained from fitting the data with  $T = 300$  K Bose–Einstein distributions. This corresponds to absolute photon numbers  $\bar{n}_0 \simeq \{4; 19; 37; 120\} \times 10^3$  and  $\bar{N} \simeq \{100; 119; 132; 207\} \times 10^3$ . To confirm the single-mode property of the condensate, we show corresponding spectra (figure 22(a), inset) with a 9 pm-resolution which is below the transverse mode spacing  $\Delta\lambda = 41$  pm. By measuring the entire cavity emission the full periodic mode structure is revealed, see figure 22(c).

The second-order correlation functions  $g^{(2)}(\tau)$  shown in figure 22(b) exhibit zero-delay autocorrelations  $g^{(2)}(0)_{\text{①–③}} = \{1.64(2); 1.30(2); 1.15(2); 1.01(1)\}$  followed by an exponential decay to  $g^{(2)}(\tau) \simeq 1$  at larger time delays (dashed lines). According to (50), we fit  $g^{(2)}(\tau) = 1 + [g^{(2)}(0) - 1]\exp(-\tau/\tau_{\text{c,exp}}^{(2)})$  to the data sets ①–③ and obtain  $\tau_{\text{c,exp}}^{(2)} \simeq \{1.75(5); 1.56(8); 1.18(3)\}$  ns. For the largest condensate fraction ④ the photon bunching vanishes,  $g^{(2)}(0) \simeq 1$ , such that we cannot determine the correlation time. Our observations reveal strikingly: above  $N_c$ , the number correlations do not rapidly drop to  $g^{(2)}(0) = 1$  as one would anticipate for a system with strictly conserved particle number [7, 38]. Indeed, the observed behaviour provides a first evidence for grand-canonical particle exchange with an effective reservoir. The bunching amplitude  $g^{(2)}(0) > 1$ , however, persists only up to a specific condensate fraction, where grand-canonical conditions cease to be applicable: the finite-size reservoir saturates and canonical ensemble conditions start to prevail in the system. According to  $\delta n_0/\bar{n}_0 = \sqrt{g^{(2)}(0) - 1}$ , the zero-delay autocorrelation  $g^{(2)}(0)$  is directly associated to the relative condensate fluctuations. For the data shown in figure 22(b), this gives  $\delta n_0/\bar{n}_0 = \{80(1); 55(2); 39(3); 10(5)\}\%$ .

## 7.3. Grand-canonical condensate correlations

We systematically demonstrate the genuine grand-canonical nature of the dye-photon-system in the Bose–Einstein condensed phase by engineering different-sized reservoirs. According to (53), the effective reservoir size is increased for high dye concentration and reduced dye-cavity-detunings.

Figure 23 shows zero-delay autocorrelations  $g^{(2)}(0)$  and the fluctuation level, respectively, as a function of  $\bar{n}_0/\bar{N}$  for five different combinations of dye concentration and detuning (R1–R3: Rhodamine 6G; R4–R5: PDI red). The main advantage of PDI red is the ability to implement small (absolute) dye-cavity-detunings  $\hbar\Delta > -2.5k_B T$  with high reabsorption rates in a spectral region (585–605 nm), where the mirrors transmit a sufficient amount of light to be measured. In order to quantify the effective reservoir size (relative to R1), we introduce

$$\varepsilon = \frac{M_{\text{eff},R_i}}{M_{\text{eff},R1}} = \frac{\rho_{R_i}}{\rho_{R1}} \times \frac{1 + \cosh(\hbar\Delta_{R1}/k_B T)}{1 + \cosh(\hbar\Delta_{R_i}/k_B T)}, \quad (66)$$

see the table in figure 23. For the case of the smallest reservoir (R1) the number fluctuations are quickly damped as the photons undergo BEC. Upon increasing the effective reservoir size (R1

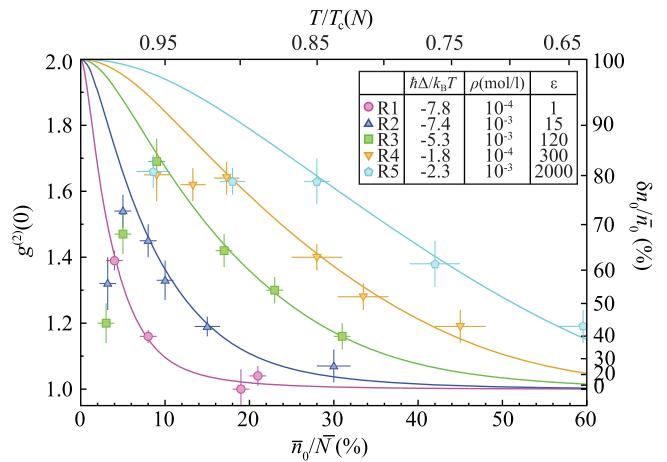
→ R5), we observe that the region with statistical fluctuations can be systematically extended to larger condensate fractions. For the largest implemented reservoir (R5), we find  $g^{(2)}(0) \simeq 1.2$  at  $\bar{n}_0/\bar{N} \simeq 60\%$ . At this point, the photon condensate performs number fluctuations  $\delta n_0/\bar{n}_0 = (g^{(2)}(0) - 1)^{1/2} \simeq 45\%$ , although its occupation  $\bar{n}_0 \approx 144\,000$  is similar to the total number  $\bar{N} \approx 240\,000$ . Our findings provide strong evidence for the photon statistics to be controlled by grand-canonical particle exchange [9, 33, 38].

The experimental results are recovered by our theoretical model (solid lines in figure 23), except for condensate fractions below 5%. This is attributed to imperfect mode filtering that leads to an effective averaging of uncorrelated photons from a few equally populated transverse modes (at  $\bar{N} \simeq N_c$ ) and suppresses the bunching amplitude. If the ground state contribution dominates ( $\bar{n}_0/\bar{N} \geq 5\%$ ), the effect becomes negligible. Furthermore, the largest detectable autocorrelation value is clamped at  $g^{(2)}(0) \simeq 1.6$ –1.7. Both issues can be resolved when the correlations are measured with a streak camera system [125]. To fit our data with the theory curves, the molecule number  $M$  is treated as a free parameter and good agreement is obtained when we choose  $10^9$ – $10^{10}$  molecules, see the caption of figure 23. The large  $M$ -values suggest that not only molecules located in the ground mode volume ( $\approx 10^8$  for  $\rho = 1 \text{ mmol l}^{-1}$ ) contribute to the effective reservoir. A possible explanation is the residual overlap between the excited TEM<sub>mn</sub> modes and the TEM<sub>00</sub> ground mode that couples molecules in both volumes by absorption and emission of ‘secondary’ photons, effectively increasing the reservoir size for the BEC. Alternatively, a modification of the autocorrelations could also be caused by photon–photon interactions [126]. To this date, the role of interactions and the origin of photon nonlinearities in the optical condensate have not been fully resolved. Previous work has identified thermal lensing to cause effective (non-local) photon–photon-interactions associated with a dimensionless interaction parameter  $\tilde{g} \simeq 10^{-5}$ – $10^{-2}$  [26, 61, 66, 67, 127, 128]. Promising candidates for the implementation of genuine quantum nonlinearities include e.g. polaritons of strongly interacting atomic Rydberg states [129, 130] or coupled cavity arrays [131]. In combination with these concepts photon BEC holds prospects for the realisation of strongly correlated many-body states of light.

#### 7.4. Intensity fluctuations and photon statistics

We have seen that the second-order correlation time ( $\tau_c^{(2)} \simeq 2 \text{ ns}$ ) of the Bose–Einstein condensed ground state is sufficiently slow to directly monitor the temporal number evolution with a fast photomultiplier.

Figure 24(a) shows the time evolution of the (normalised) photon number  $n_0(t)/\bar{n}_0(t)$  for a fixed reservoir size with parameters as in figure 23. Close to the condensation threshold, the BEC exhibits large number fluctuations, which are gradually damped out as the condensate fraction is increased. By evaluating histograms of roughly 50 traces per condensate fraction, we reconstruct the underlying photon statistics  $\mathcal{P}_n$ , i.e. the probability to find  $n$  photons in the condensate, see figure 24(b). As the fluctuations are reduced, the distributions reveal a crossover from exponentially decaying Bose–Einstein towards Poissonian



**Figure 23.** Second-order autocorrelations  $g^{(2)}(0)$  and relative condensate fluctuations  $\delta n_0/\bar{n}_0$  as a function of the condensate fraction  $\bar{n}_0/\bar{N}$  (reduced temperature  $T/T_c$ ), for different reservoir sizes R1–R5. The increasing effective reservoir size is quantified by  $\epsilon$ . For large concentrations  $\rho$  and reduced dye-cavity-detunings  $\Delta$  (R5), the fluctuations of the ground mode populations persist deep into the condensate phase. The solid lines indicate the prediction from the theoretical model discussed in section 4. Error bars give statistical uncertainties. ( $\lambda_c = \{598; 595; 580; 598; 602\} \text{ nm}$  fr R1–R5;  $\rho = \{0.1; 1.0; 1.0\} \text{ mmol l}^{-1}$  fr R1–R3 (Rhodamine 6G) and  $\rho = \{0.1; 1.0\} \text{ mmol l}^{-1}$  fr R4,R5 (PDI). For the theory curves, we use  $M = \{5.5(22); 20.5(71); 16.0(57); 2.1(4); 10.8(37)\} \times 10^9$  for R1–R5). Reprinted figure with permission from [40]. Copyright (2014) by the American Physical Society.

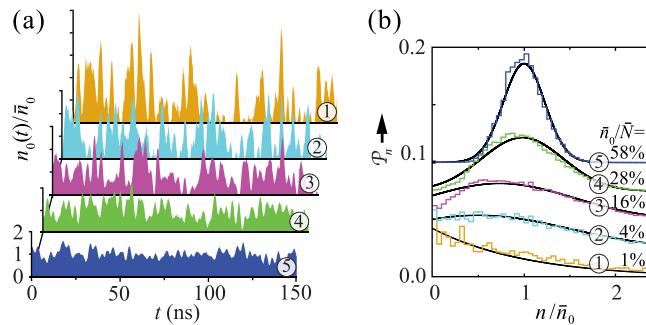
statistics, with a width  $\delta n_0$  that measures the relative degree of fluctuations  $\delta n_0/\bar{n}_0$ . Our results are in excellent agreement with theory curves (solid lines) from (30), confirming the predicted crossover from grand-canonical to canonical statistical conditions (section 4).

## 8. Phase coherence of the condensate

In the presence of large reservoirs, even strongly occupied BECs that contain thousands of photons on average display a finite probability  $\mathcal{P}_0 > 0$  to produce states without a single photon. Naturally, the question arises: how do such statistical (amplitude) fluctuations affect the temporal phase stability of the BEC? In comparison, the Poissonian statistics in the limit of small reservoirs causes the zero-photon probability to vanish  $\mathcal{P}_0 = 0$ , such that—despite residual phase diffusion [74, 115, 116, 132]—a well-defined phase is expected. Similar observations with (micro)canonical atomic BECs prompt the emergence of phase coherence for the condensate wave function [14, 16, 17, 20]. In the last section of this Tutorial, we describe an experimental measurement of the temporal phase coherence for a BEC of light.

### 8.1. Experimental scheme

To study the phase evolution, we rely on time-resolved heterodyne interference signals between the condensate emission superimposed with a dye laser acting as a phase reference, see figure 25 [74]. From a separate detection of the intensity of the



**Figure 24.** (a) Temporal evolution of the normalised condensate population  $n_0(t)/\bar{n}_0$  ( $\simeq 1.4$  ns temporal resolution). For increasing condensate fractions (①→⑤) a damping of the fluctuations is observed. (b) The photon number distributions (vertically shifted) exhibit a crossover from Bose–Einstein-like to Poissonian statistics in agreement with theory (solid lines), see also figure 8. (Parameters as in figure 22.) Reprinted figure with permission from [40].

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condensate in the interferometer (blocked dye laser), we obtain the degree of second-order coherence  $g^{(2)}(0)$  and the correlation time  $\tau_c^{(2)}$ . The experiments are performed for longitudinal wave number  $q = 7$  in the microcavity, which is filled with a Rhodamine 6G solution ( $\rho = 3$  mmol l<sup>-1</sup>). The microcavity is pumped with continuous laser light, which is here chopped into 600 ns pulses at 40 Hz repetition rate by an AOM.

As a local oscillator for the heterodyne interferometry we use a cw dye laser (Sirah Matisse), which offers a tuneable emission between  $\lambda_L = 560$ –605 nm. Analogue to the condensate operation cycle, the dye laser is acousto-optically chopped into 800 ns pulses at the same repetition rate, while the zeroth diffraction order allows to measure  $\lambda_L$  with a resolution of approximately 10 pm, see figure 25(c). The relatively long pulse duration is required in order to observe sufficiently long beatings between the condensate and dye laser emission, as will be elaborated in more detail later in this section. To obtain high-contrast interference signals, we use half-wave plates to project the polarisation axes of the momentum-filtered photon condensate and dye laser on top of each other, and combine both beams after passing a non-polarising beamsplitter (90% transmission) in a single mode fibre (Thorlabs P1-488PM)<sup>9</sup>. The temporal interference traces are detected by a fast photomultiplier tube (Hamamatsu H10721-20,  $\Delta t \simeq 0.57$  ns) and recorded with a digital oscilloscope (Tektronix DPO7000,  $\Delta\nu \simeq 3.5$  GHz). A typical time-resolved interference signal, where condensate and dye laser wavelength have been matched, is shown in figure 25(b). The superposition of Bose–Einstein condensed light field,  $\psi_c(t) = \sqrt{n_c(t)} \exp\{i[\omega_c(t)t + \varphi(t)]\}$ , and dye

laser field,  $\psi_L(t) = \sqrt{n_L(t)} \exp(i\omega_L t)$ , gives a beat signal

$$|\psi_c(t) + \psi_L(t)|^2 = n_c(t) + n_L(t) + 2\sqrt{n_c(t)n_L(t)} \times \cos\{[\omega_c(t) - \omega_L]t + \varphi(t)\}, \quad (67)$$

where  $\varphi(t)$  denotes the time-dependent condensate phase. Notably, we here have explicitly maintained the time dependence of the condensate frequency  $\omega_c(t)$ , for reasons that will be discussed in the following.

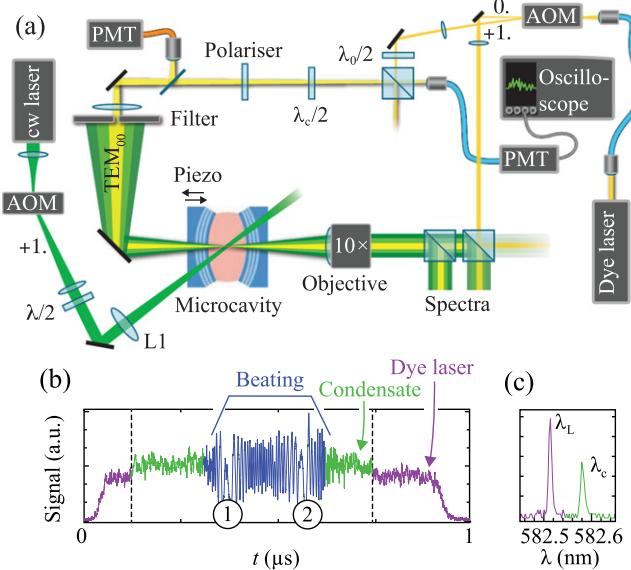
The thermodynamic state of the photon gas is obtained from a spectroscopic measurement of the energy distributions with a 4f-spectrometer (section 7). The spectra cover a wavelength (energy) range of 30 nm ( $\simeq 4k_B T$ ) and provide the ground state population  $\bar{n}_0$  and total photon number  $\bar{N}$ , being calibrated with reference spectra at  $N_c \simeq 79\,000$ . Moreover, a part of the transmitted cavity emission is injected into the high-resolution double monochromator together with the aforementioned dye laser to monitor the relative spectral position of condensate and dye laser wavelength, see figure 25(c). At the smallest achievable cavity lengths  $D_0 \approx 1.4$  μm, the curved mirrors are firmly pressed together, effectively reducing residual mechanical resonator drifts and vibrations. Under these conditions, minute piezo-tuning of the cavity length allows us to actively match the condensate with the dye laser wavelength with a spectral precision  $\Delta\lambda \approx 10$  pm. At  $\lambda = 580$  nm, the mirror separation can thus be tuned with an accuracy of  $\Delta D_0 = D_0 \Delta\lambda/\lambda \approx 24.5$  pm.

## 8.2. Modulation of the condensate frequency

Despite the mechanical stability of the microcavity, we have already seen in figure 25 that the measured intensity traces reveal a frequency modulation of the beating signal. Figure 26(a) shows the observed temporal variation of the beat signal for different initial cavity lengths, the latter modifying  $\lambda_c$ . For an average condensate wavelength blue-detuned with respect to the dye laser ( $\lambda_c < \lambda_L$ , insets of figure 26(a)), no beating signal is observed. For red-detuned light ( $\lambda_c > \lambda_L$ ) however, the occurring beating signal shows two resonances that exhibit an increased temporal separation as the condensate is further detuned, with the beating frequency in between exceeding the detector bandwidth. The interference data allows us to reconstruct the frequency drift  $\nu_c(t)$  of the condensate emission, which is shown in figure 26(b) for various cavity lengths.

The frequency drift in figure 26(b) has been recorded for temporally equidistant pump pulse excitation (pulse length  $\Delta t = 1.5$  μs every  $T_p = 25$  ms), leading to beating signals that occur in every subsequent condensate pulse with nearly the same shape (due to mechanical shot-to-shot stability). However, when irradiating the dye-microcavity with a quick sequence of 4 pump pulses ( $\Delta t = T_p = 600$  ns, followed by 100 ms dark time), see figure 26(c), only one of the four produced condensate pulses exhibits a beating with the laser, yet with the same characteristic, nearly parabolic ‘fast’ frequency drift observed previously. We attribute the ‘slow’ global frequency drift to modulation of the index of refraction of the dye medium that is caused by effectively heating the

<sup>9</sup> The optical phase is commonly retrieved in a balanced heterodyne detection scheme [133], by subtracting the interference signals at both output ports of a symmetric (50:50) beamsplitter exploiting their  $\pi$ -phase difference. For low condensate powers, however, the usage of an asymmetric (90:10) beamsplitter turned out to enhance the signal-to-noise ratio of the observed beating signals.



**Figure 25.** Heterodyne interferometry to study the phase evolution of the photon condensate. The momentum-filtered ground mode emission (left cavity mirror) is superimposed with a dye laser in a single mode fibre. (b) A typical resulting interference signal recorded with a fast photomultiplier. (c) The emission transmitted through the right cavity mirror is used to monitor the wavelengths of BEC and dye laser,  $\lambda_c$  and  $\lambda_L$ , in a double monochromator and measure the condensate fraction  $\tilde{n}_0/\bar{N}$ .

solution with the pump laser. The relaxation timescale is approximately 20 ms, similar to timescales of thermal lensing effects in our system [26, 58, 67]. The fast sloshing of the condensate eigenfrequency during a single pulse may be caused by the steep rising slope of the pump pulse itself, as it occurs in each pulse of the fast sequence scheme shown in figure 26(c). Furthermore, the behaviour is observed only for longitudinal wave numbers  $q \leq 10$  when the dye film in between the cavity mirrors becomes kinematically 2D. Both observations give reason to conclude that the parabolic contribution to the frequency modulation  $\nu_c(t)$  is based on a refractive index change, which originates from an over-damped density oscillation in the dye film. Due to the cavity length stability within a single pulse,

$$D_0 = q \frac{\lambda_c(t)}{2\tilde{n}_0(t)} = q \frac{c}{2\tilde{n}_0(t)\nu_c(t)} \stackrel{!}{=} \text{const.} \quad (68)$$

Therefore, the density of the dye solution in the ground mode volume reads

$$\rho(t) \propto \tilde{n}_0(t) = q \frac{c}{2D_0\nu_c(t)} = q \frac{1}{\tau_{\text{rt}}\nu_c(t)}, \quad (69)$$

where the (vacuum) resonator round trip time  $\tau_{\text{rt}} = 2D_0/c$  has been inserted. According to (69), the density scales inversely with the frequency drift from figure 26(b), corresponding to a compression of the solvent in the area of the pump beam. Presumably, this could be caused by an initial localised heating and dilution of the medium due to the pump pulse, see the illustrated sequence in figure 26(d). The resulting density hole leads to a reflow and densification of ethylene glycol molecules until the medium is finally

homogenised. The observed time scale of the over-damped density modulation is consistent with an estimate based on the propagation time of a sound wave through the ground mode area of diameter  $d_0 \approx 15 \mu\text{m}$ ,  $t_s = d_0/v_s \approx 10^{-8} \text{ s}$ , where  $v_s = 1688 \text{ m s}^{-1}$  is the speed of sound in ethylene glycol at 300 K [134], and it occurs on a considerably shorter time scale than thermal lensing ( $10^{-3} \text{ s}$ ). Moreover, our interpretation is affirmed by the notion that the pump beam geometry affects the condensate frequency modulation: for a larger pump beam waist, the dynamics becomes slower and the maximum of the compression is postponed to later times, see figure 26(d)<sup>10</sup>. For all subsequently discussed measurements, we use a fixed pumping geometry with a beam diameter  $2w_{01} = 2\lambda f_{L1}/\pi w_0 \simeq 140 \mu\text{m}$  (beam waist  $w_0 = 1 \text{ mm}$  and  $f_{L1} = 40 \text{ cm}$ , see figure 25).

### 8.3. Phase jump detection algorithm

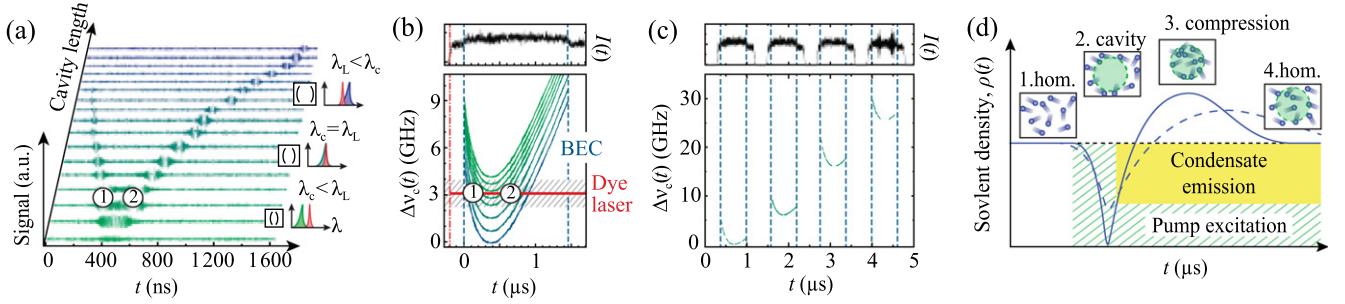
The microcavity frequency drift prohibits a temporally stable resonance condition to be fulfilled between photon condensate and dye laser, making a direct observation of the BEC phase evolution difficult. However, discrete phase jumps of the condensate can be easily unveiled if the recorded chirped interference signals are examined for irregularities in their oscillatory behaviour. For an automated analysis, we develop a phase jump detection algorithm that we benchmark with Monte Carlo-simulated data (section 5).

Figure 27 shows the simulated (a) intensity and (b) phase evolution of a BEC under grand-canonical statistics, and (c) depicts the corresponding simulated beating signal between the photon BEC and a dye laser. In the first step of the analysis, the analogue interference signal is digitalised (red). Subsequently, the procedure evaluates the digital square-signal for irregularities in the (i) width and (ii) central position of adjacent high- or low-valued segments. If the irregularities exceed predefined limits, the algorithm flags these points in time (grey shaded). The low-frequency region near the resonance (hatched) is excluded from the detection. The simulated data confirms the operability of the algorithm, as demonstrated by coincidences of grey regions with zero- or one-photon-states in the ground state (figure 27(a), top, dots). It enables the detection of discrete phase rotations between  $[0.2\pi, 1.8\pi]$ . As the analysis is based on the detection of relative irregularities, the temporal resolution is limited by the beating oscillation period.

### 8.4. Phase evolution of the photon condensate

Figure 28 shows the time evolution of the interference between photon BEC and dye laser for three different cases of photon statistics at a fixed reservoir size, starting from a strongly occupied second-order coherent condensate in figure 28(a) towards a strongly fluctuating population in (c). The left column gives the time of the

<sup>10</sup> In principle, defocusing allows one to observe temporally extended beating signals. However, this is limited by the required increased length of the pump pulses, which inevitably leads to a breakdown of the condensate operation due to photodegradation and triplet-state pumping of the dye.



**Figure 26.** (a) Interference signals upon tuning  $\lambda_c$  at fixed dye laser wavelength  $\lambda_L$  by increasing the cavity length. Both a frequency modulation and a relative temporal separation of two resonance crossings (①,②) are observed. (b) Non-resonant intensity trace (top) and relative drift of the condensate frequency  $\Delta\nu_c(t)$ , as reconstructed from (a) (bottom). Vertically shifted curves correspond to increased cavity lengths (from top to bottom), and the hatched area indicates the PMT detection bandwidth  $\approx 1.75$  GHz. (c) Combined intensity (top) and  $\Delta\nu_c(t)$  (bottom) upon excitation with a sequence of four pulses showing a beating within the last pulse. We deduce a global slow linear frequency drift over several pulses and a parabolic frequency modulation is reproduced within each individual pulse, possibly caused by a (d) temporal density modulation of the dye medium following the pump excitation (hatched). A tight pump beam focus (solid line) leads to an enhanced amplitude and speed of the modulation during the BEC emission (yellow), compared to a weak focusing scenario (dashed line).

(normalised) condensate number  $n(t)/\bar{n}$ , which is recorded after each interference measurement by blocking the dye laser. From this, the autocorrelation function  $g^{(2)}(\tau)$  is computed (middle), implicitly providing a measure of the fluctuation level  $\delta n = \bar{n}\sqrt{g^{(2)}(0) - 1}$  and the second-order correlation time  $\tau_c^{(2)}$ . In all measurements with a significant bunching amplitude ( $g^{(2)}(0) > 1$ ), an exponential fit to the autocorrelation data yields  $\tau_c^{(2)} \approx 2$  ns.

For canonical ensemble conditions with Poissonian number statistics, see figure 28(a) with  $g^{(2)}(0) = 1.01(2)$ , the beating oscillates regularly, which demonstrates the temporal coherence of the BEC throughout 120 ns<sup>11</sup>. As the condensate fraction is reduced, the reservoir size becomes sufficiently large to realise grand-canonical statistical conditions, which is hallmark by the occurrence of intensity fluctuations in figures 28(b) with  $g^{(2)}(0) = 1.33(4)$  and (c) with  $g^{(2)}(0) = 1.93(13)$ , respectively. This is accompanied by a discontinuous phase behaviour manifested in the beating signals, which for increased fluctuations shows a reduction of the time separation between adjacent phase jumps  $\Gamma_{PJ}^{-1} \approx 21.3$  ns in (b) and  $\Gamma_{PJ}^{-1} \approx 5.3$  ns in (c). In the vicinity of the detected phase jumps (grey shaded) a fit yields the magnitude of the imparted phase shift, see the inset of figure 28(b). To good approximation, the phase rotation angles are evenly distributed within the detection range  $[0.2\pi, 1.8\pi]$ , as indicated by the histogram in figure 28(d). The random distribution gives evidence for the  $U(1)$  symmetry of the infinitely phase-degenerate ground state. Physically, this equipartition is attributed to the intrinsic randomness of a spontaneous emission event, which is expected to trigger the emergence of a condensate after a previous fluctuation to low photon numbers.

### 8.5. First- and second-order coherence times

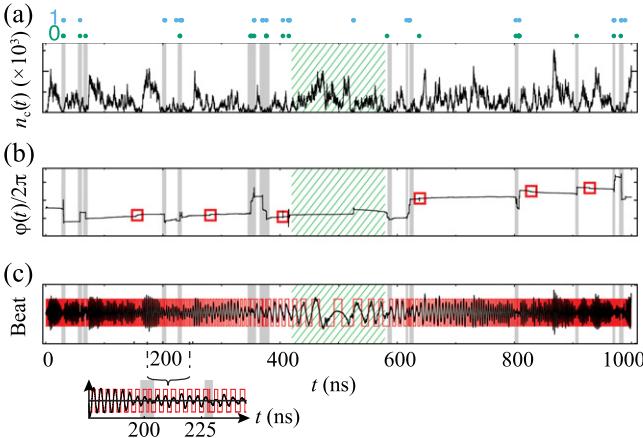
As previously discussed, figure 28 indicates a separation of the dynamics for number and phase fluctuations: while  $\tau_c^{(2)}$

<sup>11</sup> For large waists of the pump beam, the longest recorded time span without phase jumps was 1  $\mu$ s (300 m coherence length).

remains nearly constant, the measured values for  $\Gamma_{PJ}^{-1}$  change by 2 orders of magnitude and seem to depend on the choice of the statistical ensemble and its associated zero-photon-probability  $\mathcal{P}_0$ .

For a quantitative analysis of the time scale separation, figure 29 summarises experimental results of the phase jump rates  $\Gamma_{PJ}$  and inverse second-order correlation times  $1/\tau_c^{(2)}$  as a function of the average photon number in the condensate for three different-sized particle reservoirs. The phase jump rates (filled symbols) increase strongly for both growing reservoir size as well as decreasing condensate photon number ('system size') based on the here enhanced probability to have a low photon number given the increased fluctuation level (inset), which reduces the phase stability. The rates deduced from the zero-photon-probabilities  $\Gamma_{PJ}^0 = \hat{B}_{12}M\mathcal{P}_0$  (solid lines) show an excellent agreement with the experimental data. This suggests that a drop of the condensate population to zero followed by a spontaneous emission process is physically responsible for the observed phase jumps. Similarly, the inverse second-order correlation times  $1/\tau_c^{(2)}$  (open symbols) present a good agreement with theory curves (dashed lines) based on (51).

For all three configurations, a separation of the time scales for first- and second-order coherence is visible in the statistics crossover region, i.e. near  $\bar{n} = \sqrt{M_{eff}}$ . What is its physical origin? On the one hand, spontaneous emission events can cause arbitrary phase fluctuations. However, these matter only when a few photons occupy the ground state with a likelihood given by the photon statistics, which therefore dominates the first-order phase jump dynamics, see (56). On the other hand, the dynamics of particle number fluctuations is subject to absorption and emission rates of photons by the dye medium, according to (48). Although for increased condensate populations (at a fixed reservoir) the relative fluctuations  $\delta n/\bar{n}$  are reduced, the fluctuation time scale is still controlled by the Einstein coefficients. In fact, even larger condensate populations lead to a reduction of the second-order correlation time, in stark contrast to the increased first-order correlation time. Although our analysis does not account for diffusive contributions to the temporal phase



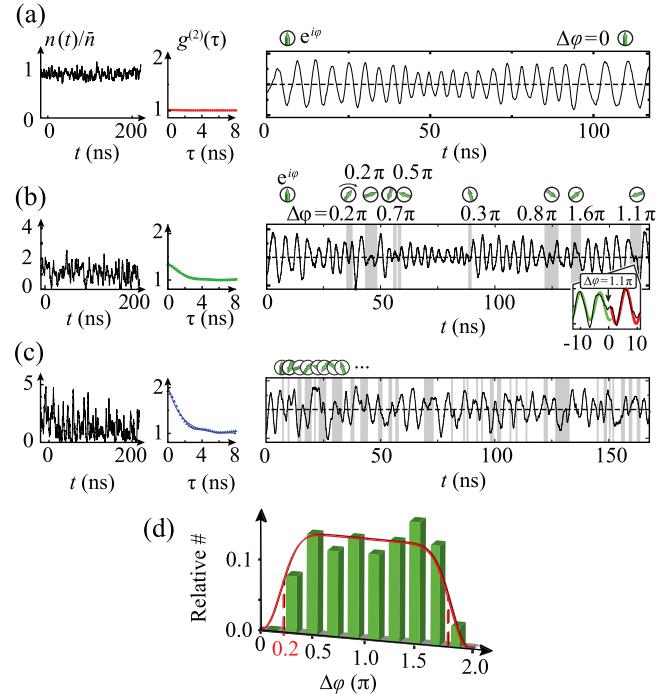
**Figure 27.** Phase jump detection benchmark with simulated interference signals. (a) Number evolution of a fluctuating BEC and points in time with zero or one photon in the ground mode (dots, top). (b) Corresponding phase evolution (units of  $2\pi$ ) with a phase jump detection resolution  $\Delta\varphi > 0.2\pi$  (red boxes indicate missed events). (c) Beating between photon condensate and reference laser and digitalised signal (red). Irregularities in the periodicity of subsequent square signals selected by our algorithm (grey shaded, see also zoomed time trace) coincide with actual phase jumps in (b). The resonance is excluded from the detection (hatched). ( $M = 10^6$ ,  $\bar{n}_c = 280$  with  $g_c^{(2)}(0) = 1.67$ ,  $\hbar\Delta/k_B T = 0$ , wavelength drift  $\lambda_c(t) = \lambda_L[1 - (0.5 - t/T)/20]$  with  $T = 1\ \mu\text{s}$ . Reference laser:  $\bar{n}_L = 1000$  with  $g_L^{(2)}(0) = 1.0$ )

coherence [74], it conveys the unusual properties of Bose-Einstein condensed light: a light source comprised of a single macroscopically occupied emitter that exhibits statistical intensity fluctuations as large as in a thermal source. The relation between first and second-order coherence for thermal emitters,  $g^{(2)}(\tau) = 1 + |g^{(1)}(\tau)|^2$ , is however expected to hold only in the extreme grand-canonical regime with  $\bar{n} \geq \sqrt{M_{\text{eff}}}$  [87, 135].

#### 8.6. Extrapolation to the thermodynamic limit

Finally, we discuss the physical significance of statistical number fluctuations and phase coherence for a photon BEC in the thermodynamic limit. For this, we study the phase jump rate for enlarged system sizes. Importantly, we ensure to increase the sizes of both condensate  $\bar{n}$  and effective particle reservoir  $M_{\text{eff}}$  in a way that conserves the statistical ensemble conditions.

Figure 30(a) shows the reservoir-system-ratio  $\sqrt{M_{\text{eff}}}/\bar{n}$  as a function of  $\bar{n}$  for different values of  $g^{(2)}(0)$  obtained from numerical calculations. For a given photon number  $\bar{n}$ , the reservoir size  $M_{\text{eff}}$  is adjusted iteratively until the corresponding photon number distribution  $\mathcal{P}_n$  reproduces one of the target values  $g^{(2)}(0) = \{1.10; \dots; 1.90\}$ . Subsequently, the procedure is repeated for larger condensate populations to yield further data points at the same fluctuation level. Our numerical results indicate that conserving the the statistical ensemble conditions, i.e.  $\sqrt{M_{\text{eff}}}/\bar{n}$ , is equivalent to a constant zero-delay autocorrelation. This suggests that the phase coherence may be extrapolated towards the thermodynamic limit ( $\bar{n}, \bar{N}, M \rightarrow \infty$ ,  $\sqrt{M_{\text{eff}}}/\bar{n} = \text{const.}$ ), provided that one



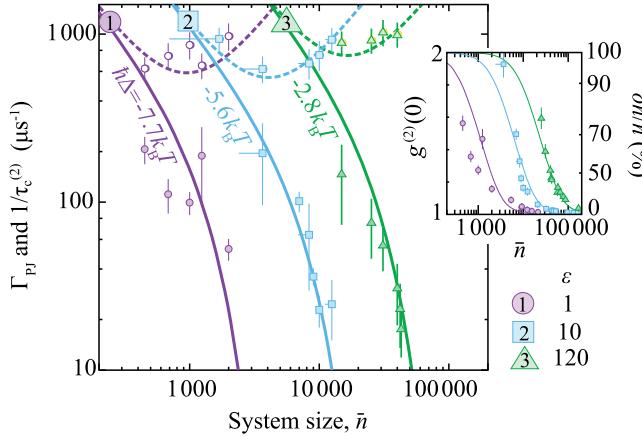
**Figure 28.** Interference between photon BEC and dye laser (right) for average photon numbers (a)  $\bar{n} = 114\ 000$  ( $\bar{n}/\bar{N} = 57\%$ ) (b) 8300 (10%) and (c) 3700 (5%), which realises different levels of statistical number fluctuations, as visible in the normalised photon number evolution (left) and the autocorrelation (middle). Regions that have been identified by our detection algorithm (grey shaded) indicate phase jumps at increasing rates from (a) to (c). The magnitude of the phase rotations is obtained from a fit as shown in the inset in (b). (d) Histogram of the phase rotation angles  $\Delta\varphi$  for 108 fitted phase jumps in signals as in (b). Within the detection window  $[0.2\pi, 1.8\pi]$  (red line), the random distribution reflects the  $U(1)$  symmetry of the ground state, which is broken upon condensation. (Rhodamine 6G,  $\rho = 3\ \text{mmol l}^{-1}$ ,  $\lambda_c = 582\ \text{nm}$ ). Reprinted figure with permission from [43]. Copyright (2016) by the American Physical Society.

does maintain the fluctuation level  $g^{(2)}(0)$ . Strictly speaking, an extrapolation also requires the critical temperature  $T_c(\bar{N}) \propto \sqrt{\bar{N}/R}$  to be constant. This could be achieved by increasing the radius of curvature of the cavity mirrors  $R \rightarrow \infty$  proportional to  $\bar{N}$ . Experimentally, this compensation is unfeasible with the described setup, such that we only comply with the requirements for a fixed statistical ensemble<sup>12</sup>.

Figure 30(b) gives the phase jump rate versus autocorrelation for three reservoirs. For all values of  $g^{(2)}(0)$ , we observe that photon condensates coupled to the smallest reservoir ( $\hbar\Delta = -7.7k_B T$ ) exhibit shorter coherence times than condensates coupled to the medium-sized ( $-5.6k_B T$ ) and largest ( $-2.3k_B T$ ) reservoir. This meets our expectations: for the same level of fluctuations, i.e. the same statistical ensemble, an increased condensate population should reduce the zero-photon-probability  $\mathcal{P}_0$ , see (58).

From our data, we extract three sets of phase jump rates for selected zero-delay autocorrelations  $g^{(2)}(0) = 1.59(18)$ ,  $1.18(9)$  and  $1.02(1)$ , which are shown in

<sup>12</sup> Recently realised photon gases in variable micropotentials might however render a conservation of  $T_c$  tractable [67, 68].

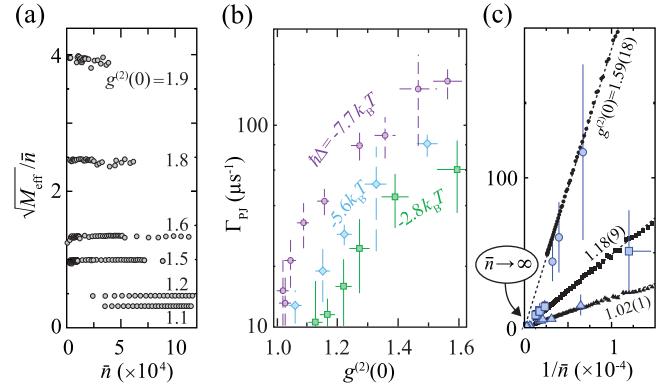


**Figure 29.** Inverse second-order correlation time  $1/\tau_c^{(2)}$  (open symbols) and phase jump rates  $\Gamma_{\text{PJ}}$  (filled symbols) versus condensate number for three different-sized reservoirs, realised by  $\hbar\Delta = -\{7.7; 5.6; 2.8\} k_B T$  and quantified by  $\varepsilon$ , see (66). Solid lines give theory curves for  $\Gamma_{\text{PJ}}^0$  with  $M = \{2.0; 4.5; 5.0\} \times 10^9$ ,  $\hat{B}_{12} = \{140; 250; 1\,300\} \text{ s}^{-1}$ . In the region accessed by our experiments, the phase jump rates are significantly smaller than the inverse second-order correlation times. The inset gives the corresponding zero-delay autocorrelation  $g^{(2)}(0)$  along with numerical calculations (solid lines). ( $\lambda_c = \{597; 582; 563\} \text{ nm}$ ,  $\rho = 3 \text{ mmol l}^{-1}$ ,  $\bar{n} = \sqrt{M_{\text{eff}}} \simeq \{1.0; 4.1; 16.5\} \times 10^3$ , error bars are statistical uncertainties). Reprinted figure with permission from [43]. Copyright (2016) by the American Physical Society.

figure 30(c) versus the inverse condensate population  $1/\bar{n}$ . All data sets lie in the range  $\bar{n} \geq \sqrt{M_{\text{eff}}}$ , for which a separation of  $\Gamma_{\text{PJ}}$  and  $1/\tau_c^{(2)}$  has been observed. A linear extrapolation of the data towards an infinitely large condensate ( $1/\bar{n} \rightarrow 0$ ) is consistent with a full suppression of discrete phase jumps in the thermodynamic limit, in spite of the absence of second-order coherence. Numerical calculations (black symbols) for  $g^{(2)}(0) = 1.50, 1.18$  and  $1.05$  support this conclusion. The largest realised fluctuation level comes close to the photon statistics crossover,  $g^{(2)}(0) = \pi/2$ , with a zero-photon-probability  $\mathcal{P}_0 \simeq 0.64 \bar{n}^{-1}$ , see (45). Under the assumption that phase jumps occur due to vanishing photon numbers, a fit to the data in figure 30(c) yields  $\Gamma_{\text{PJ}}/\hat{B}_{12}M = 0.51(14)\bar{n}^{-1}$  reproducing the expected slope within the quoted uncertainty. For lower fluctuation levels, the exact scaling of  $\mathcal{P}_0$  with  $\bar{n}$  remains elusive and we therefore compare our data only with numerical results, which similarly demonstrate a linear scaling of  $\Gamma$  with the inverse photon number ( $\mathcal{P}_0^{1.18} = 0.13(5)\bar{n}^{-1}$  and  $\mathcal{P}_0^{1.02} = 0.06(2)\bar{n}^{-1}$ ). Although the presence of amplitude fluctuations of the condensate wave function  $\sqrt{n(t)} \exp(i\phi(t))$  reduces the degree of first-order coherence, we expect that in the investigated parameter regime discrete phase jumps will be fully suppressed in the thermodynamic limit.

## 9. Conclusions and outlook

This Tutorial has presented a study of the thermalisation dynamics and temporal coherence properties of a Bose–Einstein condensed photon gas in the grand-canonical statistical



**Figure 30.** (a) Numerical scaling of effective reservoir and condensate size versus  $\bar{n}$  for various fluctuation levels  $g^{(2)}(0)$ , demonstrating the conservation of  $\sqrt{M_{\text{eff}}/\bar{n}}$  as the autocorrelation remains fixed. ( $\hbar\Delta = 0$ ,  $M = 0.5\text{--}2.0 \times 10^{10}$ ). (b) Measured  $\Gamma_{\text{PJ}}$  versus autocorrelation for three effective reservoirs. For each fluctuation level, the larger system exhibits an enhanced phase stability. (c) Extrapolation of phase coherence for large photon-reservoir-systems at three fixed degrees of fluctuations  $g^{(2)}(0)$  (filled symbols). By extrapolating  $\bar{n} \rightarrow \infty$  (dashed), all curves indicate a full suppression of phase jumps in the thermodynamic limit. Black symbols are numerical results for  $\Gamma_{\text{PJ}}^0 = \hat{B}_{12}M\mathcal{P}_0$  at corresponding  $g^{(2)}(0)$  and  $M = 4 \times 10^9$ ,  $\hat{B}_{12} = 1000 \text{ s}^{-1}$ . (Parameters as in figure 29.) Reprinted figure with permission from [43]. Copyright (2016) by the American Physical Society.

ensemble. Key evidences are provided by measurements of (i) the spectral photon dynamics, which demonstrates the thermalisation of the photons due to reabsorptive coupling to a dye heat bath, (ii) the large (grand-canonical) statistical number fluctuations at significant condensate fractions, and (iii) the observed variation of the temporal phase coherence of the condensate wave function. An extrapolation to the thermodynamic limit gives BECs with super-Poissonian number statistics despite suppressed phase jumps.

The realisation of BEC in the grand-canonical ensemble has for the first time shed light on the long-discussed *grand-canonical fluctuation catastrophe* [9, 30–36]. The observation of extremely large, statistically fluctuating condensate populations demonstrates the physical significance of the grand-canonical ensemble for the Bose–Einstein condensed phase. Moreover, the results provide the fundamental insight that BEC does not strictly imply first- or second-order coherence.

For the future, it will be exciting to study phase diffusive contributions to the condensate linewidth, as has been theoretically predicted but remains elusive in any Bose-condensed system to date [74, 115, 116, 132]. A major experimental challenge here depicts the required frequency stability of the photon BEC to observe minute phase drifts over long measurement durations. Moreover, it is expected that in-depth studies of the thermal character of the grand-canonical statistical fluctuations may reveal unusual fluctuation–dissipation-relations in the ideal Bose gas, associated with macroscopic thermodynamic quantities as e.g. a generalised statistical compressibility imposed by the particle reservoir. From a technical point of view, macroscopically occupied, but incoherent photon condensates under grand-canonical

conditions could pose interesting novel light sources for speckle-free imaging applications due to their high directional brilliance and (tunable) low degree of coherence.

Further exciting research directions for grand-canonical BECs might be pursued in conjunction with variable potentials for thermalised light and coupled condensates, as has been demonstrated in microstructured optical cavities [67, 68]. Phase-stable, macroscopically occupied condensates arranged in a lattice are expected to constitute a realisation of the XY model of 2D interacting spins, that could provide a fruitful platform to address complex optimisation problems [136, 137]. In this regard, the phase jumps associated with grand-canonical statistical fluctuations could mimic spin fluctuations at an effective temperature: at sufficiently low ‘temperatures’ one expects the emergence of the BKT phase associated with algebraic long-range spin order [138–141].

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