Bose-Einstein condensation of photons

Jan Klaers and Martin Weitz

Institut für Angewandte Physik, Universität Bonn,
Wegelerstr. 8, 53115 Bonn, Germany

We review recent work on the Bose-Einstein condensation of photons in a dye microcavity environment. Other than for material particles, as e.g. cold atomic Bose gases, photons usually do not condense at low temperatures. For Planck’s blackbody radiation, the most ubiquitous Bose gas, photon number and temperature are not independently tunable and at low temperatures the photons simply disappear in the system’s walls, instead of massively occupying the cavity ground mode. In the here described approach, this obstacle is overcome by a fluorescence-induced thermalization mechanism in a dye-filled microcavity. Experimentally, both the thermalization of the photon gas and, at high photon densities, Bose-Einstein condensation has been observed. This article describes the thermalization mechanism of the photon gas in detail and summarizes so far performed experimental work.

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I. INTRODUCTION

When a gas of particles with given density is cooled to such low temperatures that the associated de Broglie wavepackets spatially overlap, quantum statistical effects come into play. Specifically, for a gas of particles with integer spin (bosons), Bose-Einstein condensation into the ground state sets in above a critical phase space density. For dilute atomic gases, this effect has first been observed in 1995 by means of laser and subsequent evaporative cooling of alkali atoms [1–3], see also the article of F. Chevy and J. Dalibard in this volume. The signature of Bose-Einstein condensation has also been observed for several solid-state quasi-particles, as exciton-polaritons and magnons, see e.g. the contributions by Y. Yamamoto and by V. Demokritov and A. Slavin, respectively, in this volume.

Photons, the quantized particles of light also are bosons, but usually show no Bose-Einstein condensation. In a blackbody radiator, the chemical potential of photons vanishes, i.e. the average particle number does not follow a given conservation law, but adjusts itself to the available thermal energy [4]. This is the essence of the Stefan-Boltzmann law, linking the total radiation energy $U$ to the fourth power of the temperature, $U \propto T^4$. At low temperatures, the photon number simply decreases and no macroscopic occupation of the cavity ground state occurs. Thus, a necessary precondition for a Bose-Einstein condensation of photons is to find a thermalization process that allows for an independent adjustment of both photon number and temperature. The lack of such a mechanism has long prevented the realization of light sources that are capable of generating single mode light, without the necessity to be driven out of thermal equilibrium, as e.g. for a laser. Note that in a laser both the state of the light field and that of the active medium are far removed from thermal equilibrium [5]. To some extent, lasing is even a prime example for a non-equilibrium process, as only the absence of thermal equilibrium allows for inversion and optical gain. Early theoretical work has proposed to reach Bose-Einstein condensation of photons by Compton scattering off a
thermal electron gas \[6\]). Later, Chiao et al. proposed a two-dimensional photon quantum fluid in a nonlinear Fabry-Perot resonator, where thermalization was sought from photon-photon scattering \[7–9\]. This concept is similar to atom-atom scattering processes in atomic physics BEC experiments, though the limited non-linearity has so far prevented a thermalization of the photon gas \[10\]. In other work, the demonstration of (quasi-)equilibrium Bose-Einstein condensation of exciton-polaritons, mixed states of matter and light, has been reported \[11–13\]. Here interparticle collisions of the excitons, i.e. the material parts of the polaritons, act as a thermalization mechanism. In other experiments, superfluidity of polaritons has been observed \[14, 15\].

In recent experiments of our group, photon Bose-Einstein condensation is achieved in a dye-solution filled optical microresonator \[16–18\]. Thermalization of the photon gas with the dye is achieved by repeated absorption emission cycles. For such systems it is known that frequent collisions (\(\sim 10\) fs timescale) between solvent and dye molecules causes rapid transverse decoherence at room temperature, so that the condition of strong light-matter coupling is not met \[19, 20\]. The distance between the two spherically curved resonator mirrors is in the micrometer regime, which causes a large frequency spacing between the longitudinal resonator modes. The latter is of order of the emission width of the dye molecules. In combination with an intracavity modification of the spontaneous emission, preferring the emission to small volume modes (low transversal excitation), a regime is reached, where to good approximation the resonator is populated only by photons of a single longitudinal mode number, see Fig. IIa. The longitudinal modal quantum number is frozen out and the photon gas effectively becomes two-dimensional. As is indicated in Fig. IIb, the photon dispersion relation acquires quadratic, i.e. particle-like character, with the frequency of the transverse TEM\(_{00}\) mode acting as a low-frequency cutoff frequency. Furthermore, a harmonic trapping potential for the photon gas is induced by the mirrors curvature. Thermal equilibrium of the photon gas with its environment (at room temperature) is achieved as the photons are absorbed and emitted by the dye molecules repeatedly. The photon frequencies will accumulate within a spectral range of order \(\sim k_B T/\hbar\) above the low frequency cutoff. Other than in a blackbody radiator, the thermalization process allows for an independent adjustment of temperature and photon number. This becomes clear by noting that the energy of fluorescence photons, which takes values above the cutoff frequency of the resonator in energy units \((\hbar \omega_{\text{cutoff}} \simeq 2.1\, \text{eV})\), is far above thermal energy, \(k_B T \simeq 1/40\, \text{eV}\). Thus, purely thermal excitation of (optical) photons is negligible. Instead, the number of photons will be determined by the strength of the optical
pumping. One can show that the photon gas confined in the resonator is formally equivalent to a harmonically trapped two-dimensional gas of massive bosons with effective mass

$$m_{\text{eff}} = \hbar \omega_{\text{cutoff}} n_0^2 / c^2,$$

where $n_0$ denotes the refractive index of the medium and $c$ the vacuum speed of light. For such a system it is well known that a Bose-Einstein condensate exists at a finite temperature \[21, 22\]. In recent experiments, we have both observed thermalization of the photon gas in the dye-filled microcavity system \[17\] as well as Bose-Einstein condensation \[16\]. The so far observed properties of the photon Bose-Einstein condensates in many respects resemble that of atomic gases, while the approximately 10 orders of magnitude smaller effective photon mass allows for transition temperatures in the room temperature regime.

Bose-Einstein condensation and superfluidity are two closely related phenomena \[23\]. While the former is connected to equilibrium properties, the latter deals with transport properties. Bose-Einstein condensation is in principle possible with an ideal gas, while the presence of superfluidity requires interparticle interactions. In future, it remains to be experimentally verified whether the photon Bose-Einstein condensate also exhibits superfluidity.

In the following, section II describes the fluorescence induced thermalization mechanism

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**Figure 1:** (a) Scheme of optical resonator (top) and the cavity modes (bottom) for the case of a spacing between resonator mirrors of half an optical wavelength of the lowest optical mode. The resonator is filled with dye solution. The photon gas thermalizes to the temperature of the dye solution. (b) Photon dispersion in resonator (solid line) and the dispersion of a free photon (dashed line).
of the photon gas and section III the statistical theory of the trapped photon gas. Further, section IV reviews experiments on the thermalization process and section V corresponding results on Bose-Einstein condensation. Finally, section VI gives conclusions.

II. FLUORESCENCE INDUCED THERMALIZATION

A. Kennard-Stepanov theory of dye spectra

Early experimental work has shown that spectra of dye molecules in liquid solution show several universal properties \[24–27\]. These are for example the mirror rule, which states that the spectral absorption profile is a mirror image of the fluorescence profile; the Stokes rule, stating that the spectral centroid of fluorescence occurs at a higher wavelength than that of absorption; and Kasha’s rule \[28\], which expresses that the fluorescence does not depend on the wavelength of the exciting light. These common properties, which to good accuracy are fulfilled in many dye species, suggest that absorption and fluorescence in such systems follow a general mechanism. Most of these properties can be understood as being the consequence of a collisionally induced thermalization mechanism \[24, 25\]. For a corresponding model, consider an idealized dye molecule with an electronic ground state \(S_0\) and an electronically excited state \(S_1\), each of which are subject to additional rovibronic level splitting, as shown in Fig. 2a. The vibrational and rotational state of a dye molecule in liquid solution is permanently altered by collisions with solvent molecules, e.g. on a femtosecond timescale at room temperature. The frequent collisions lead to a thermalization of the rovibrational state within a sub-picosecond timescale, which is much faster than the electronic lifetime of the excited state, being typically of the order of nanoseconds. When a photon is absorbed, the dye molecule is likely to be transferred to a highly rovibronically excited substate of the \(S_1\) manifold, but the excessive rovibronic energy will be quickly dissipated into the solvent bath. The fluorescence photon will be emitted from a dye state that is in thermal equilibrium with the solvent bath, with typically lower rovibrational quantum number. This thermalization process, which occurs both in the electronically excited and in the ground state level, explains why fluorescence can be dissipative (Stokes shift) and why there typically is no correlation between the wavelength of the absorbed and the emitted photon (Kasha’s rule).

The thermalization of the rovibronic degrees of freedom has another, though closely related, consequence: the Einstein coefficients of absorption and emission at a certain photon energy \(\hbar \omega\) are connected by the Boltzmann factor of that energy. This relation is known as the
Kennard-Stepanov law, and can be written in the form

\[
\frac{B_{21}(\omega)}{B_{12}(\omega)} = \frac{w_\downarrow}{w_\uparrow} e^{-\frac{\hbar(\omega-\omega_0)}{k_B T}}
\]  

(1)

where \(B_{12}(\omega)\) and \(B_{21}(\omega)\) are the Einstein coefficients of absorption and stimulated emission respectively, \(\omega_0\) is the frequency of the zero-phonon line of the dye, and \(w_\downarrow, w_\uparrow\) are statistical weights related to the rovibronic density of states, which will be defined subsequently in this text. We note that the Kennard-Stepanov relation can also be stated in terms of \(A(\omega)/B_{12}(\omega)\), where \(A(\omega)\) is the Einstein coefficient for spontaneous emission - with the difference to the above definition essentially being the density of states. This relation has been discovered in the beginning of the last century \[29, 30\], and has been 'rediscovered' several times. A short historical outline can be found in \[31\]. Both theoretical and experimental investigations can be found in the literature \[31–35\].

In the following, we give a short derivation of the Kennard-Stepanov relation. As described above, we model the dye molecule by an electronic two-level system with levels \(S_0\) and \(S_1\), also denoted by \(\downarrow\) and \(\uparrow\), each of which is subject to additional rovibronic level splitting. It is important to note that the Einstein coefficients of such a medium at a given frequency \(\omega\) are an average over all pairs of individual rovibronic substates \((\alpha, \beta)\), with \(\alpha \in S_0, \beta \in S_1\) that match the transition frequency:

\[
e_\alpha + \hbar \omega = \hbar \omega_0 + e_\beta .
\]  

(2)

The latter equation expresses energy conservation, see also the Jablonski diagram of Fig. 2a. We assume that the population of rovibronic states in both lower and upper electronic states is fully thermalized from frequent collisions with solvent molecules. This assumption will be valid as long as the radiative lifetime of the electronically excited state \(S_1\) remains clearly longer than the thermalization time. We thus expect that the corresponding substates within the lower and upper electronic manifolds will be occupied with a probability given by the Boltzmann factors

\[
p_\alpha = e^{-\frac{e_\alpha}{k_B T}}/w_\downarrow \quad \text{and} \quad p_\beta = e^{-\frac{e_\beta}{k_B T}}/w_\uparrow ,
\]  

(3)

with the normalization factors

\[
w_\downarrow = \sum_{\alpha \in S_0} e^{-\frac{e_\alpha}{k_B T}} \quad \text{and} \quad w_\uparrow = \sum_{\beta \in S_1} e^{-\frac{e_\beta}{k_B T}} .
\]  

(4)

From eqn (2) and (3) one immediately obtains
Figure 2: (a) Jablonski diagram of a dye molecule with two electronic levels. Both lower and upper electronic state, $S_0$ and $S_1$, are split into a rovibronic substructure. (b) The process of absorption and fluorescence of a photon can be seen as a photochemical reaction.

\[
p_{\beta} = \frac{w_\downarrow}{w_\uparrow} e^{-\frac{\hbar(\omega-\omega_0)}{k_B T}} p_{\alpha}.
\]  

We can now write

\[
\frac{B_{21}(\omega)}{B_{12}(\omega)} = \frac{\sum_{(\alpha,\beta)} p_\beta B(\beta \rightarrow \alpha)}{\sum_{(\alpha,\beta)} p_\alpha B(\alpha \rightarrow \beta)},
\]

where we have introduced the Einstein coefficients $B(\alpha \rightarrow \beta)$ for transitions between the corresponding individual rovibronic states. Applying $B(\alpha \rightarrow \beta) = B(\beta \rightarrow \alpha)$ and using in addition (5), we finally obtain

\[
\frac{B_{21}(\omega)}{B_{12}(\omega)} = \frac{w_\downarrow}{w_\uparrow} e^{-\frac{\hbar(\omega-\omega_0)}{k_B T}} \frac{\sum_{(\alpha,\beta)} p_\alpha B(\alpha \rightarrow \beta)}{\sum_{(\alpha,\beta)} p_\alpha B(\alpha \rightarrow \beta)} = \frac{w_\downarrow}{w_\uparrow} e^{-\frac{\hbar(\omega-\omega_0)}{k_B T}},
\]

which is the Kennard-Stepanov law of eqn (1). As described above, the Kennard-Stepanov relation is experimentally well established. However, many dye species show smaller or even larger deviations. For a discussion of these issues, see section II E.

### B. Chemical equilibrium between photons and molecules

Molecules in the electronic ground state can be transferred to the upper electronic level by the absorption of a photon, while photon emission leads to de-excitation. These processes can be seen as a photochemical reaction of the type

\[
\gamma + \downarrow \rightleftharpoons \uparrow,
\]
see also the illustration of Fig. 2b. Here γ stands for a photon, ↓ for a molecule in the electronic ground state, and ↑ for a molecule in the electronically excited state. In chemical equilibrium the corresponding chemical potentials, with \( \mu_\downarrow \) (\( \mu_\uparrow \)) denoting the chemical potential of a ground (excited) state molecule and \( \mu_\gamma \) indicating the photon chemical potential, satisfy the relation

\[
\mu_\gamma + \mu_\downarrow = \mu_\uparrow. \tag{9}
\]

If we express this relation in terms of the photon fugacity \( z \), the equation reads

\[
z = e^{\mu_\gamma / k_B T} = e^{\mu_\uparrow / k_B T} / e^{\mu_\downarrow / k_B T}. \tag{10}\]

Next, let the partition function of a dye molecule be

\[
\mathcal{F} = w_\downarrow e^{\mu_\downarrow / k_B T} + w_\uparrow e^{-\hbar \omega_0 - \mu_\uparrow / k_B T}, \tag{11}\]

where \( w_\downarrow, w_\uparrow \) are the statistical weights of eqn (4). Further, \( \omega_0 \) is the frequency of the zero-phonon line of the dye. We can then identify

\[
w_\uparrow e^{-\hbar \omega_0 - \mu_\uparrow / k_B T} / \mathcal{F} = \frac{\rho_\uparrow}{\rho}, \tag{12}\]

\[
w_\downarrow e^{\mu_\downarrow / k_B T} / \mathcal{F} = \frac{\rho_\downarrow}{\rho}, \tag{13}\]

as the probability of finding a molecule in the excited (ground) state, where \( \rho_\uparrow \) (\( \rho_\downarrow \)) is the spatial density of the excited (ground) state dye molecules respectively, and \( \rho \) is the total dye molecular density. Using eqn (10), we find that the photon chemical potential is determined by the relation

\[
z = e^{\mu_\gamma / k_B T} = \frac{w_\downarrow \rho_\uparrow e^{-\hbar \omega_0 / \hbar}}{w_\uparrow \rho_\downarrow}. \tag{14}\]

In chemical equilibrium the photon chemical potential is thus determined by the excitation ratio \( \rho_\uparrow / \rho_\downarrow \), which corresponds to the relative number of dye molecules in the electronically excited state [18, 36].

C. Thermal equilibrium and Markov-processes

To begin the discussion of the thermalization process, let us characterize thermal equilibrium in terms of a Markov process. In general, the time evolution of a physical system coupled to a heat bath can be regarded as a random walk in configuration space [37, 38]. Such a random walk is fully characterized by the present state of the system and fixed transition
rates between configurations (process 'without memory'). Not all sets of transition rates are physically meaningful. A necessary requirement is that for a sufficiently long time evolution of the system each configuration has to occur with the statistical weight of its Boltzmann factor. In the following, we derive a condition from which one can easily decide if a given set of transition rates leads to thermal equilibrium. In the next section this will then be applied to the fluorescence induced thermalization process of a photon gas.

Let \( K \) denote a certain state of a physical system (for a photon gas this will be given by a set of mode occupation numbers). Furthermore, \( p_K(t) \) denotes the probability to find state \( K \) at time \( t \). The temporal evolution of \( p_K(t) \) is given by the master equation

\[
p_K(t + 1) - p_K(t) = \sum_{K'} p_{K'}(t) R(K' \to K) - \sum_{K'} p_K(t) R(K \to K'),
\]

where the coefficients \( R(K \to K') \) denote the transition rates between configurations. We are interested in transition rates that asymptotically bring the system into thermal equilibrium, i.e. which yield \( p_K(t \to \infty) = \exp(-E_K/k_B T)/Z \), with \( Z \) as the partition function. The asymptotic master equation for this case is given by

\[
0 = \sum_{K'} \exp(-E_{K'}/k_B T) R(K' \to K) - \sum_{K'} \exp(-E_K/k_B T) R(K \to K') .
\]

Equation (16) has many solutions. One solution stands out by showing no net probability flow between two given states (detailed balance):

\[
0 = \exp(-E_{K'}/k_B T) R(K' \to K) - \exp(-E_K/k_B T) R(K \to K') \forall K, K',
\]

(17)

or

\[
\frac{R(K \to K')}{R(K' \to K)} = \exp(-\Delta E/k_B T) \forall K, K',
\]

(18)

with an energy difference \( \Delta E = E_{K'} - E_K \). If the transition rates \( R(K \to K') \) fulfill eqn (18), this gives a sufficient condition that the Markov process drives the system into thermal equilibrium.

D. Light-matter thermalization process

To begin with, consider an experimental scenario as depicted in Fig. 3, consisting of a macroscopic box with reflecting walls that is filled with a dye solution. Light, emitted e.g. by a spectrally narrow optical source, is irradiated at point A. What is the spectrum of the radiation leaving the dye container at point B? If the optical density of the dye is sufficiently
Figure 3: Multiple absorption-fluorescence cycles in a macroscopic dye-filled photon box. Asymptotically, the photons entering the box at point A would be transferred to blackbody radiation at the temperature of the environment. However, without a low-frequency cutoff the thermalization process remains incomplete, as photons leave the absorption bandwidth of the dye.

large, multiple absorption-emission fluorescence cycles will occur. One then experimentally finds that the spectrum at point B is red shifted with respect to that obtained for a single fluorescence event. In the context of fluorescence spectroscopy, this effect is known as the inner filter effect \([24, 25]\). Interestingly, this red shift of the fluorescence can be understood in terms of a partial thermalization process. By fluorescence and reabsorption, the photons exchange energy with the dye solution. This energy exchange establishes a thermal contact between the light and the dye medium. In principle, one would expect that the state of the light field asymptotically relaxes towards a thermal state at the temperature of the dye solution, i.e. after many complete cycles the spectrum of blackbody radiation at room temperature would be obtained. However, owing to the successive red shift, along with the limited spectral bandwidth of the dye, the reabsorption probability rapidly decreases, and the dye solution gradually becomes transparent. The thermal contact soon breaks down and the process remains incomplete. Such an incomplete thermalization occurs e.g. in some solar light concentrators \([39]\).

Regarding photon number conservation, it on the other hand is not desirable to transfer the irradiated light into blackbody radiation. As soon as the photon energy becomes comparable to thermal energy, one would lose the ability to experimentally tune the photon number. In equilibrium, the photon number would then readjust following the available thermal energy, independently of the optical pumping power irradiated at point A (provided that the pumping does not significantly heat up the dye solution). However, in a microcavity environment, as it is used in our experiment, both a full thermalization and an experimentally tunable photon number can be achieved. This becomes clear by noting that the low-frequency cutoff, imposed
by the resonator, prevents a successive red-shift of the photon gas. The thermalization by repeated absorption-emission cycles can now proceed until the photon gas is fully thermalized, i.e. thermal contact to the dye solution is not lost. Owing to the low-frequency cutoff, the photon energy can remain far above the thermal energy. With $\hbar \omega_{\text{cutoff}} \approx 2.1 \text{eV} \gg 1/40 \text{eV} \simeq k_B T$ this limit is fulfilled in our experiment. In this situation, no photons are created or destroyed on average. The thermalization process here conserves the photon number, with the latter being determined by the strength of the optical pumping.

In the remainder of this section, we present a more rigorous treatment of the light-matter thermalization [17, 36]. The thermalization process is considered as a random walk in the configuration space of all allowed light field states. Here a state $K$ is given by the cavity mode occupation numbers $K = (n^K_0, n^K_1, n^K_2, \ldots)$. The mode occupation numbers are permanently altered by photon absorption and emission processes. In first order perturbation theory, the rates (per volume) for absorption and emission of one photon in mode $i$ at cavity position $r$, denoted by $R^{K,i}_{12}(r)$ and $R^{K,i}_{21}(r)$, have the form

$$R^{K,i}_{12}(r) = B_{12}(\omega_i) u_i(r) \rho_\downarrow n^K_i$$

$$R^{K,i}_{21}(r) = B_{21}(\omega_i) u_i(r) \rho_\uparrow (n^K_i + 1)$$

where $u_i(r)$ is the spectral energy density of one photon in mode $i$. We assume that the number of dye molecules is sufficiently large, such that photon absorption and emission leave the densities of ground state ($\rho_\downarrow$) and excited state ($\rho_\uparrow$) molecules unchanged, i.e. $\rho_\uparrow, \downarrow$ can be treated as fixed parameters. This assumption corresponds a grandcanonical limit, see also section III C.

Suppose that a state $K'$ emerges from state $K$ by the absorption of a photon in mode $i$, with $n^K_{i'} = n^K_i - 1$, and accordingly, that $K$ emerges from $K'$ by an emission process into this mode. The corresponding rates are $R^{K,i}_{12}(r)$ and $R^{K',i}_{21}(r)$, and with eqn (19) and (20), their ratio is given by $R^{K,i}_{12}(r)/R^{K',i}_{21}(r) = B_{12}(\omega_i) \rho_\downarrow / B_{21}(\omega_i) \rho_\uparrow$. In section III C it was shown that the random walk given by the transition rates of eqn (19) and (20) will lead to thermal equilibrium, if the ratio $R^{K,i}_{12}(r)/R^{K',i}_{21}(r)$ is given by the Boltzmann factor of the energy difference between $K$ and $K'$

$$\frac{B_{12}(\omega_i) \rho_\downarrow}{B_{21}(\omega_i) \rho_\uparrow} \equiv e^{-\frac{\hbar \omega_i - \mu_i}{k_B T}},$$

see also [36, 40, 41]. If we now apply the Kennard-Stepanov relation, eqn (1), and assume chemical equilibrium, eqn (14), the detailed balance condition of eqn (21) is indeed verified.
This means that multiple absorption-emission cycles drive the photon gas into thermal equilibrium with the dye solution at temperature $T$, and with a photon chemical potential $\mu_\gamma$ determined by the molecular excitation ratio. The average occupation number $\bar{n}_i$ of mode $i$ can be determined by balancing the average absorption and emission rates at a given cavity position. As expected, this gives a Bose-Einstein distribution for the average occupation number, with $\bar{n}_i = (\exp[(\hbar \omega_i - \mu_\gamma)/k_B T] - 1)^{-1}$.

### E. Spectral temperature

From the Kennard-Stepanov theory of dye spectra, as presented in section II A, one expects that the spectra of absorption and emission are interlinked by a Boltzmann factor. In general, this law is empirically well confirmed. However, it is known that many real dyes show more or less pronounced deviations. Before choosing a particular dye medium, it is thus necessary to check whether it fulfills the Kennard-Stepanov relation. For this it proves helpful to introduce a spectral temperature, which is derived using a formal solution of eqn (1). Consider the Kennard-Stepanov relation for two different frequencies $\omega$ and $\omega + \delta \omega$. From eqn (1), we obtain

$$\frac{B_{21}(\omega)}{B_{12}(\omega)} \cdot \frac{B_{12}(\omega + \delta \omega)}{B_{21}(\omega + \delta \omega)} = \exp\left(\frac{\hbar \delta \omega}{k_B T}\right).$$

We can now solve this equation for the frequency dependent spectral temperature $T_{\text{spec}}(\omega)$:

$$T_{\text{spec}}(\omega) = \frac{\hbar \delta \omega}{k_B \ln\left(\frac{B_{21}(\omega)}{B_{12}(\omega)} \cdot \frac{B_{12}(\omega + \delta \omega)}{B_{21}(\omega + \delta \omega)}\right)},$$

and eventually perform the limit $\delta \omega \rightarrow 0$. Thus, if the spectral profiles of absorption and emission are known, one can easily calculate the corresponding spectral temperature. We note that the particular form of $T_{\text{spec}}$ given by eqn (23) and (24) has the advantage of being independent of the statistical weights $w_{\downarrow, \uparrow}$ and the frequency of the zero-phonon line $\omega_0$, which might not be known in all cases. Moreover, in this form it is sufficient to know the coefficients $B_{12,21}(\omega)$ only up to a proportionality factor.

The Kennard-Stepanov relation holds for a given dye, if its spectral temperature coincides with the thermodynamic temperature of the solution, $T_{\text{spec}}(\omega) = T$, independent of the frequency $\omega$. Figure 4 shows the spectral temperatures, here as function of the wavelength, for eight different dye species. They were calculated with eqn. (23) using the corresponding spectra of the database of Ref. [42]. The dyes on the left hand side of the figure fulfill
the condition $T_{\text{spec}}(\omega) \simeq T$ to good approximation, while the dyes on the right hand side show significant deviations. Noteworthy, the spectral temperature here has a tendency to be higher than the ambient temperature, see also [35, 43]. As explanation for the observed deviations, one mainly finds two lines of reasoning in the literature, either based on incomplete rovibronic thermalization or on inhomogeneous broadening [35, 44–46]. Our experiment uses either perylene-dimide or rhodamine 6G dye, for both of which the quantum efficiency is 95% or above, and the Kennard-Stepanov relation is well fulfilled.

III. THERMODYNAMICS OF THE TWO-DIMENSIONAL PHOTON GAS

Formally, the photon gas confined in the resonator is equivalent to a two-dimensional gas of massive bosonic particles. This can be seen by investigating the energy momentum relation. We will derive the equilibrium properties of such a gas, and discuss the expected condensate fluctuations.

A. Cavity photon dispersion

We begin by expressing the energy of a photon in the microcavity as a function of longitudinal (i.e. along the optical axis) wavenumber $k_z$ and transverse wavenumber $k_r$,

$$E = \frac{\hbar c}{n_0} |k| = \frac{\hbar c}{n_0} \sqrt{k_z^2 + k_r^2},$$

(25)

where $n_0$ is the refractive index of the medium. Let us denote the spacing between the two cavity mirrors on the optical axis as $D_0$ ($\approx 1.46 \mu m$ in our experiment), and the radii of the two curved mirrors, which we assume to be identical, as $R$ ($\approx 1 m$ typically). The boundary conditions generated by the mirrors imply a resonance condition

$$k_z(r) = q\pi/D(r),$$

(26)

with $q$ as the longitudinal mode number and $D(r) = D_0 - 2(R - \sqrt{R^2 - r^2})$ as the mirror spacing at a distance $r$ from the optical axis. In a paraxial limit, with $k_r \ll k_z$ and $r \ll R$, we can approximate the photon energy by the expansion

$$E \simeq \frac{m_{\text{eff}} c^2}{n_0^2} + \frac{(\hbar k_r)^2}{2m_{\text{eff}}} + \frac{1}{2}m_{\text{eff}} \Omega^2 r^2,$$

(27)

with $m_{\text{eff}} = \hbar k_z(0)n_0/c = \hbar \omega_{\text{cutoff}} n_0^2/c^2$ and $\Omega = (c/n_0)/\sqrt{D_0 R / 2}$. This yields the dispersion relation of a particle with mass $m_{\text{eff}}$ moving in the transverse resonator that is subject to
Figure 4: Spectral temperatures $T_{\text{spec}}(\lambda)$ for different dyes at room temperature, derived from the spectra of the database of Ref. [42]. This numerical determination is most precise in the Stokes region, where both absorption and fluorescence are sufficiently strong and thus most precisely known. We nevertheless estimate an error of the given spectral temperatures of at least 15%. The corresponding quantum efficiencies are: perylene-diimide: $\Phi = 0.97$, rhodamine 6G: $\Phi = 0.95$, squarylium: $\Phi = 0.65$, coumarin 314: $\Phi = 0.68$, phthalocyanine: $\Phi = 0.60$, PPO: $\Phi \approx 1$, coumarin 343: $\Phi = 0.63$, chlorophyll B: $\Phi = 0.117$.

additional harmonic confinement with trapping frequency $\Omega$. If we in addition account for a nonlinear self-interaction of photons that modifies the refractive index following $n_0 \rightarrow n_0 + n_2 I(r)$, where $n_2$ is a nonlinear index of refraction, and $I(r)$ is the optical intensity, we
find that the photon energy is shifted by an amount

$$E_{\text{int}} \simeq -m_{\text{eff}} c^2 \frac{n_2}{n_0^3} f(r).$$  \hspace{1cm} (28)

A non-zero value of $n_2$ can arise e.g. from a Kerr nonlinearity or thermal lensing. Since the confined photons have the quadratic dispersion relation of massive particles, we can define a thermal de Broglie wavelength by $\lambda_{\text{th}} = \hbar / \sqrt{2 \pi m_{\text{eff}} k_B T}$, in direct analogy to e.g. a gas of atoms. Essentially, $\lambda_{\text{th}}$ is inversely proportional to the average transversal wavenumber $\sqrt{\langle k_z^2 \rangle}$ of the photons, and its value at room temperature is $\simeq 1.58 \mu$m. Bose-Einstein condensation is expected when the phase space density $n \lambda_{\text{th}}^2$ exceeds a value near unity, where $n$ denotes the two-dimensional number density.

**B. Statistical theory of two-dimensional Bose gas in trap**

For an exact determination of the phase transition, a statistical multimode treatment is necessary. The average photon number at a transversal energy $u = \hbar (\omega - \omega_{\text{cutoff}})$ is given by a Bose-Einstein distribution

$$n_{T,\mu}(u) = \frac{g(u)}{e^{\frac{u}{k_B T}} - 1},$$  \hspace{1cm} (29)

where $g(u) = 2(u/\hbar \Omega + 1)$ is the degeneracy factor (which here increases linearly with energy), and the factor 2 accounts for the two possible polarizations. A macroscopic occupation of the ground state mode at $u = 0$ sets in when the particle number $N$ reaches a critical value of $N_c = \sum_{u>0} n_{T,\mu=0}(u)$. We find

$$N_c = \frac{\pi^2}{3} \left( \frac{k_B T}{\hbar \Omega} \right)^2.$$  \hspace{1cm} (30)

The typical photon trapping frequency in our setup is $\Omega \simeq 2 \pi \cdot 4.1 \times 10^{10}$ Hz. For room temperature, $T = 300$ K, we arrive at $N_c \simeq 77000$, which is experimentally feasible. If this critical value is reached, the occupation of transversally excited modes is expected to saturate and the ground mode starts to become macroscopically populated. The possibility to observe a Bose-Einstein condensation at room temperature can be understood from the extremely small effective photon mass $m_{\text{eff}} = \hbar \omega_{\text{cutoff}} n_0^2 / c^2$, which in our case corresponds to $2.1 \text{eV} n_0^2 / c^2 \simeq 7 \cdot 10^{-36}$ kg. This is 10 orders of magnitude below the mass of alkali atoms, which extremely enhances the phase transition temperature with respect to atomic systems.
Figure 5: Schematic illustration of the excitation exchange between the photon gas and a reservoir of electronically excited dye molecules. The photon gas can be seen as an open system, in the sense of grandcanonical experimental conditions.

C. Condensate fluctuations

In our experiment, photons are initially brought into the system by pumping the dye medium with a laser. The pumping is maintained throughout the measurement to compensate for photon losses due to unconfined optical modes, finite dye quantum efficiency and mirror losses. Despite pumping, the photon gas is expected to both spectrally and spatially relax to thermal equilibrium, provided that a photon scatters several times off a dye molecule before being lost [17]. By optical pumping we generate and uphold a reservoir of electronic excitations in the dye medium that can exchange particles with the photon gas. This experimental situation is well described by a grandcanonical statistical ensemble, see Fig. 5 in which the photon gas acquires the temperature of the dye solution and the photon chemical potential is directly related to the excitation level in the medium, as given by eqn (14).

The particle exchange with the reservoir of excited state dye molecules is of special relevance for the expected photon statistics and second order coherence of the condensate. It is well known that there is no ensemble equivalence for Bose-Einstein condensation [47–49]. In particular, the particle number fluctuations of the condensate strongly depend on the underlying statistical ensemble. For a (micro-)canonical ensemble, which is typically realized in atomic BEC experiments, the particle number distribution of the ground state changes from Bose-Einstein- to Poisson-like when the condensation sets in. This is accompanied by a damping of the number fluctuations. On the other hand, the particle number distribution for a grandcanonical ensemble remains Bose-Einstein-like and the number fluctuations remain of
the order of the average occupation number, \( \Delta n_0 = \sqrt{\langle n_0^2 \rangle - \bar{n}_0^2} \simeq \bar{n}_0 \). This can also be seen in the second order coherence of the condensate. The zero-delay autocorrelation function \( g^{(2)}(0) = \langle n_0(n_0 - 1) \rangle / \langle n_0 \rangle^2 \) is not expected to drop off to \( g^{(2)}(0) = 1 \), as it does for (micro-)

ical BECs. Instead, a bunching behavior with \( g^{(2)}(0) = 2 \) is expected even below the critical temperature [36]. In the future, it will be very interesting to test for such unusually large condensate fluctuations. This regime is not observed in Bose-Einstein condensates of ultracold atomic gases, but likely to occur for photonic BECs.

IV. EXPERIMENTS ON PHOTON GAS THERMALIZATION

A. Experimental setup

We begin by describing measurements on the thermalization of the photon gas in the dye-filled microresonator, which have been carried out at comparatively small photon numbers, i.e. far below the critical photon number. A scheme of the experimental setup used in the experiments of Refs. [16, 17] of our group is shown in Fig. 6. The optical resonator consists of two highly reflecting mirrors with reflectivity above 99.997% in the wavelength region 500 – 590 nm. This reflectivity translates to a finesse of > 100000 for the empty cavity. The mirrors are spherically curved with a typical radius of curvature of \( R = 1 \) m. To allow for a cavity length in the micrometer regime despite the curvature, one of the mirrors is cut to 1×1 mm surface size. Both resonator mirrors are glued onto glass substrates. Additional glass prisms are attached to the side to allow for a pumping under an angle of 45°, see Fig. 6. The typical (effective) cavity length of \( D_0 \simeq 1.46 \mu m \) is determined from the resonator free spectral range, and corresponds to the \( q = 7 \) longitudinal mode. The used dyes are either rhodamine 6G or perylenedimide (PDI), whose absorption and fluorescence spectra are shown in Fig. 7. The dyes have a high quantum efficiency between 95% and 97% [50], and spectral temperatures close to the thermodynamic temperature of the dye solution. We use organic solvent, e.g. methanol and ethylene glycol, and typical dye concentrations of \( 1.5 \times 10^{-3} \text{ Mol/l} \) for rhodamine 6G. A careful filtering of the dye solution is necessary before placing it between the mirrors. The microcavity is pumped with a laser beam at a wavelength of 532 nm derived from a frequency doubled Nd:YAG laser. The light transmitted through one of the cavity mirrors is split into two partial beams by a non-polarizing beamsplitter. One beam is imaged onto a CCD-camera for spatial analysis, while the other is sent into a spectrometer for a monitoring of the emitted spectrum. We use different types of spectrometers, both commercially available
Figure 6: Schematic set-up of the microcavity experiment. The dye-filled microresonator is pumped at an angle of 45° to the optical axis. The radiation emerging from the microresonator is examined both spatially and spectrally.

(a) rhodamine 6G and (b) perylene diimide.

Figure 7: Relative strengths of absorption, $B_{12}(\lambda)/B_{12}^{\text{max}}$, and emission, $B_{21}(\lambda)/B_{21}^{\text{max}}$, for the dyes (a) rhodamine 6G and (b) perylene diimide.
Figure 8: (a) Spectral distribution of the radiation emitted from the microcavity far below the critical photon number at $T = 300\,\text{K}$ and $T = 365\,\text{K}$ (circles). The spectra are in good agreement with the Boltzmann-distributed photon energies (lines). For comparison, a $T = 300\,\text{K}$ Boltzmann-distribution is additionally plotted in the bottom graph (dashed line). (Rhodamine 6G in ethylene glycol, concentration $5 \times 10^{-4}\,\text{mol/l}$, mirror curvatures $R_1 = R_2 = 1\,\text{m}$, cavity order $q = 7$) (b) Normalized spectral distribution of the radiation emitted from the microcavity (connected circles) for 4 different cutoff wavelengths $\lambda_c \simeq \{570, 590, 615, 630\}\,\text{nm}$, from top to bottom. In addition, Boltzmann-distributed photon energies for $T = 300\,\text{K}$ are plotted (lines).

and self-built devices. Care has to be taken when the spectrometer makes use of an entrance slit. The coupling efficiency to the spectrometer will be different for the various transversally excited cavity modes. This problem can be overcome by placing a diffusing plate in front of the entrance slit. However, a more advantageous method is to remove the entrance slit altogether. This is possible because the microresonator fluorescence can be sufficiently well collimated even without an additional spatial filtering. In Fig. 6, a self-built version of such a device is shown. The shown spectrometer uses a rotating grating and a photomultiplier tube.

B. Spectral and spatial intensity distribution

In initial experiments, we have tested for the thermalization of the two-dimensional photon gas in the dye-filled resonator [17]. Figure 8a shows spectra of light transmitted through one of the cavity mirrors for two different temperatures of the resonator setup (top: room temperature, $T = 300\,\text{K}$, bottom: $T = 365\,\text{K}$). The output power for these measurements was $(50 \pm 5\,\text{nW})$, which corresponds to an average photon number in the cavity of $N_{\text{ph}} = 60 \pm 10$. 
From this, the chemical potential can be determined by numerical solving \( \sum_{u \geq 0} n_{T,\mu}(u) = N_{ph} \), with \( n_{T,\mu}(u) \) given by eqn (29). For the two data sets in Fig. 8, we obtain \( \mu/k_B T = -6.76 \pm 0.17 \) \( (T = 300 \text{ K}) \) and \( \mu/k_B T = -7.16 \pm 0.17 \) \( (T = 365 \text{ K}) \), respectively. Clearly, both measurements are performed far below the phase transition, which for this finite size system is expected to occur once the chemical potential becomes comparable to the trap level spacing (i.e. \( \mu \approx -\hbar \Omega \)). Because of \( -\mu/k_B T \gg 1 \), the term \(-1\) in the denominator of eqn (29) can be neglected and the distribution becomes Boltzmann-like. As can be seen from the figure, besides a derivation near 532 nm from residual pump light, the measured spectra are in good agreement with theoretical expectations over a wide spectral range for the two temperatures, respectively. We interpret this as evidence for the photon gas to be in thermal equilibrium with the dye solution. In another set of measurement, the cutoff wavelength \( \lambda_{\text{cutoff}} = 2\pi c/\omega_{\text{cutoff}} \) was varied, as can easily be achieved in our setup by a piezo tuning of the cavity length. Corresponding spectra are shown in Fig. 8b, along with theory spectra obtained by assuming a Boltzmann distribution of the transversally excited modes. Satisfactory agreement between theory and experiment is only obtained for the two upper spectra with cutoff wavelengths near 570 nm and 590 nm. For the two lower spectra with longer cutoff wavelength, no satisfactory thermalization is present, which can be attributed to the weak dye reabsorption in this spectral regime. These observations illustrate the importance of both emission and reabsorption for the thermalization process.

We have also monitored the spatial distribution of the photon gas at similarly low pumping power. As before, the experimental data can be well explained by assuming a Boltzmann-like population of the transversally excited cavity modes [17]. Furthermore, a spatial concentration of light into the center was observed, which can be seen as consequence of minimizing the photon energy in the effective trapping potential induced by the mirror curvature. Technical applications of this light concentration effect could include the collection of diffuse solar light to a central spot [39, 51].

V. EXPERIMENTS ON PHOTON BOSE-EINSTEIN CONDENSATION

In subsequent experiments, the dye microresonator was operated at higher pump powers, allowing for photon numbers near and above the critical regime. To avoid population of triplet states and excessive heat deposition, the pump beam in these measurements was acousto-optically chopped to 0.5 \( \mu s \) long pulses with 8 ms repetition time. Since the pulses are two orders of magnitude longer than the lifetime of the excited dye molecules and four
orders of magnitude longer than the lifetime of the photons (average time between emission and reabsorption), the experimental conditions can be considered as quasi-static.

Typical experimental spectra of the photon gas are shown in Fig. 9 for different optical pumping powers [16]. At low pumping and correspondingly low intracavity optical power, a broad spectral distribution resembling a Boltzmann-like occupation of modes above the cavity cutoff is seen. Near the phase transition, the spectral maximum shifts towards the cutoff. When exceeding the critical photon number, a spectrally sharp peak at the position of the cavity cutoff is visible. The observed spectral width is limited by the resolution of the used spectrometer. The experimental results are in good agreement with theoretical spectra based on a Bose-Einstein-like population of cavity modes, see the inset of the figure. The observed imperfect saturation of the transversally excited modes for even higher photon numbers is attributed to an interaction-induced deformation of the effective trapping potential [52]. At the phase transition, the optical intracavity power is $P_{c,\text{exp}} = (1.55 \pm 0.6) \text{ W}$, which corresponds to a photon number of $(6.3 \pm 2.4) \times 10^4$. This value is in good agreement with the
theoretical prediction of eqn (30). Notably, a similar measurement with PDI gives the same result within the experimental uncertainties. Thus, the obtained results seem to be largely independent of the used dye.

Figure 9b shows spatial images of the radiation emitted by the dye microcavity (real image onto a color CCD camera) both below (top) and above (bottom) the critical power. Both pictures show a shift from the yellow spectral regime for the transversally low excited cavity modes located near the trap center to the green for transversally higher excited modes appearing at the outer trap regions. In the lower image, a bright spot is visible in the center with a measured FWHM diameter of \((14 \pm 2) \mu m\). Within the quoted uncertainties, this agrees with the expected diameter of the TEM\(_{00}\) mode, \(d = 2 \sqrt{\hbar \ln 2 / m_{\text{eff}} \Omega} \approx 12.2 \mu m\), yielding clear evidence for a macroscopic population of the transversal ground mode. Further, we observe that not only the height of the condensate peak increases for higher photon numbers, but also its width. This mode diameter increase suggests a weak repulsive self-interaction mediated by the dye solution. The origin of this is most likely thermal lensing, but in principle it could also be due to a microscopic Kerr-nonlinearity in the dye medium. Both effects contribute to the nonlinear index of refraction of eqn (28), if they are treated on a mean-field level. By comparing the observed increase of the mode diameter with numerical solutions of the two-dimensional Gross-Pitaevskii equation, we estimate a dimensionless interaction parameter \(\tilde{g} = (7 \pm 3) \times 10^{-4}\). This is significantly smaller than the values in the range \(10^{-2} \ldots 10^{-1}\) reported for two-dimensional atomic gas experiments, and also below the regime in which Kosterlitz-Thouless physics can be expected to be relevant. An indication for the latter would be the loss of long-range order. Experimentally, when directing the condensate peak through a sheering interferometer we have not seen any signatures for such a spatially varying phase. Thus, the observed condensation does not seem to deviate from the BEC scenario of an ideal or weakly interacting Bose gas.

In further experiments, we have investigated the influence of the resonator geometry on the observed critical particle number. For that we measured the critical circulating power for various mirror radii and different mirror spacings. From eqn (30) we expect that the critical power \(P_c = N_c \hbar \omega^2 / 2\pi q = (\pi^2 / 12)(k_B T)^2(\omega / hc)R\) should increase linearly with the mirror radius \(R\) and have no dependence on the cavity order \(q\) (at least as long as the two-dimensionality of photon gas holds). Figure 10a and the lower graph of Fig. 10b give the corresponding experimental results, which confirm the theoretical predictions, both the scaling and the absolute values. The upper graph of Fig. 10b shows the pump power required to achieve criticality as...
function of the longitudinal mode number \( q \). Here, a decrease is observed for larger mirror spacings. This becomes clear by noting that the pump power absorption, which is of order 1% of the incident light, increases with the thickness of the dye film. Therefore, less pump power is necessary to reach a given circulating power. The experimental data can be well modeled by assuming that the absorbed pump power at criticality remains constant when increasing \( q \) (solid line). Note that a quite contrary scaling was observed in early work on ‘thresholdless’ microlasers, for which an increase of the pump threshold with mirror spacing has been reported [56, 57].

As already discussed, for lower intracavity powers the thermalization process is accompanied by a spatial redistribution of photons towards the trap center. This effect is also observed for higher pump powers, and we expect it to provide a critical photon density in the center of the cavity even for a displaced pumping spot. We note that this effect has been observed in polariton BEC experiments [13], but is unobserved in lasers. For a corresponding measurement, the pump beam was displaced 50 \( \mu \)m away from the trap center. A set of spatial intensity profiles for different values of the cavity cutoff wavelength is shown in Fig. [11]. By a tuning of the cavity cutoff one can vary the reabsorption probability, which alters the degree of thermalization. The lowest shown profile displays results recorded with a cutoff wavelength near 610 \( \mu \)m. The low dye reabsorption in this wavelength range inhibits an effi-
Figure 11: Intensity distribution of the photon gas along an axis intersecting the trap center for different cut-off wavelengths $\lambda_c$ (indicated on the left hand side). The pump beam (dashed line) is located outside the trap center and its position as well as its power are kept fixed during the course of the measurement. The top curve shows a photon gas at criticality with a ground state population of $N_0/N \lesssim 1\%$ (Rhodamine 6G in methanol, concentration $1.5 \times 10^{-3}$ Mol/l, cavity order $q = 7$, mirror curvatures $R_1 = R_2 = 1$ m, pulse duration $0.5\,\mu$s).

cient thermalization, and spatial accumulation at the trap center. Thus, the maximum of the fluorescence remains at the location of the pump spot (shown by a dashed line). If the cutoff wavelength is decreased, the reabsorption probability increases and the intensity profiles get more symmetrically distributed around the trap center. The top profile shows data recorded for a cutoff near $570\,\text{nm}$. Here, the photon gas here seems fully thermalized. The spatial intensity profile shows a cusp at the trap center, which indicates criticality (condensate fraction $\lesssim 1\%$). As the pump beam intensity at the trap center is almost negligible, we conclude that the critical photon density is not directly generated by pumping, but is a consequence of the spatial relaxation towards the cavity ground state connected to the thermalization.

VI. CONCLUSIONS

We have investigated thermodynamic properties of a two-dimensional photon gas in a dye-filled optical microcavity. A fluorescence induced thermalization process leads to a thermal photon gas at room temperature with freely adjustable chemical potential. Evidence for a Bose-Einstein condensation of photons was obtained from Bose-Einstein distributed photon energies including a massively populated ground state mode, the phase transition occurring at
the expected critical photon number and exhibiting the predicted dependence on the cavity geometry, and a spatial relaxation process leading to a condensation even for a displaced pump spot.

An interesting question is, in what respect does photon Bose-Einstein condensation relate to lasing. In general, the main borderline between a laser and a photonic Bose-Einstein condensate is that lasing is a non-equilibrium phenomenon, while Bose-Einstein condensation occurs in thermal equilibrium. Thermalization of the photon gas above a cavity low-frequency cutoff can be achieved in media fulfilling the Kennard-Stepanov relation, when recapturing (spontaneous) emission in one of the many transversal cavity modes, and tuning to a regime with strong reabsorption. In such a system, the condition for the emergence of a macroscopically occupied photon mode, which for a laser usually is given by a (small-signal) gain condition in an inverted medium, is replaced by a Bose-Einstein criticality condition, as given in eqn (30) for the case of a harmonically trapped two-dimensional photon gas. In essence, this is equivalent to the condition of the phase space density exceeding a value near unity. The thermalization process will select the cavity ground state to have the biggest statistical weight, which even becomes macroscopic when condensation sets in.

For the future, we plan to investigate condensate properties in more detail. For example, an intriguing question is whether the observed self-interaction is sufficient to cause superfluidity. Also, the coherence properties of the photon condensate should be further explored. Initial interferometric measurements have already delivered a lower bound for the first order coherence length in the centimeter regime [18, 41]. It will be important to also investigate the second-order coherence of the photon Bose-Einstein condensate. This is of particular interest because of the unusually large intensity fluctuations that are expected for grandcanonical Bose-Einstein condensates.

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