Bose-Einstein condensation of photons in a ‘white-wall’ photon box

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Abstract. Bose-Einstein condensation, the macroscopic ground state occupation of a system of bosonic particles below a critical temperature, has been observed in cold atomic gases and solid-state physics quasiparticles. In contrast, photons do not show this phase transition usually, because in Planck’s blackbody radiation the particle number is not conserved and at low temperature the photons disappear in the walls of the system. Here we report on the realization of a photon Bose-Einstein condensate in a dye-filled optical microcavity, which acts as a “white-wall” photon box. The cavity mirrors provide a trapping potential and a non-vanishing effective photon mass, making the system formally equivalent to a two-dimensional gas of trapped massive bosons. Thermalization of the photon gas is reached in a number conserving way by multiple scattering off the dye molecules. Signatures for a BEC upon increased photon density are: a spectral distribution that shows Bose-Einstein distributed photon energies with a macroscopically populated peak on top of a broad thermal wing, the observed threshold of the phase transition showing the predicted absolute value and scaling with resonator geometry, and condensation appearing at the trap centre even for a spatially displaced pump spot.

1. Introduction

When a gas of particles is either cooled or its density is increased up to the point where the de Broglie wavepackets overlap, quantum statistical effects come into play [1]. Specifically, for material particles with integer spin (bosons), the phenomenon of Bose-Einstein condensation is expected. Experimentally, this situation was achieved in the dilute gas limit in 1995, using laser and subsequent evaporative cooling of rubidium and sodium atoms, respectively [2, 3]. The signature of Bose-Einstein condensation was also observed with solid-state physics quasiparticles [4–7]. Other than material particles, photons in black-body radiation do not show a phase transition to a Bose-Einstein condensate [8]. Thermal photons usually have no chemical potential, corresponding to a non-conserved particle number, and at low temperature the photons disappear in the cavity walls instead of macroscopically populating the ground state mode. Early theoretical work has proposed to reach a Bose-Einstein condensate of photons using Compton scattering off a thermal electron gas to allow for a particle number conserving thermalization [9, 10]. After realization of the first atomic Bose-Einstein condensates, we have witnessed an increased interest in light sources, where a macroscopically populated photon mode is not the result of laser-like amplification, but is caused by an equilibrium phase transition. In a laser, both the state of the light field and that of the amplifying, usually inverted, medium is far removed from thermal equilibrium [11]. In an interesting work, Chiao et al. proposed to achieve a photon fluid in a two-dimensional nonlinear resonator configuration [12]. Thermal equilibrium of the photon gas was sought from photon-photon scattering, similar to atom-atom scattering.
Figure 1. (a) Scheme of optical microresonator (top) and mode spectrum (bottom) for the case of a spacing between resonator mirrors of half an optical wavelength of the lowest optical mode ($q = 1$). The resonator is filled with dye solution and the photon gas thermalizes to the rovibrational temperature of the dye molecules. (b) Dispersion of a photon in the resonator (solid line) and the dispersion of a free photon (dashed line). (c) Trapping potential for the photon gas imposed by the curved mirrors.

in atomic physics BEC experiments. However the weak photon-photon interaction in available nonlinear materials has so far prevented thermalization with this method [13]. Experimentally, in a series of experiments exciton-polaritons, strongly coupled mixed states of matter and light, have been reported to allow a (quasi-) equilibrium Bose-Einstein condensation of polaritons [4–6]. The thermalization process used is based on interparticle interactions between polaritons, and in more recent experiments, superfluidity was observed [14, 15]. Our experiment described here is based on a dye molecular solution placed within an optical resonator. Thermalization of the photon gas with the dye medium is achieved by absorption and reemission of photons in the dye molecules. Rapid decoherence from frequent collisions of dye molecules with the solvent here prevents a coupling of the phases of dipole and photon [16, 17], so that we can assume the relevant particles to be well described by photons instead of polaritons. In initial experiments, a thermalized two-dimensional photon gas with freely adjustable chemical potential was realized [18]. More recently, we have observed a Bose-Einstein condensate of photons in the dye-filled optical microcavity setup [19].

2. Experimental Scheme and Setup
Our experiment traps photons in a dye solution filled optical microresonator, see Figure 1a for a schematic picture of the experiment. The distance between the two high reflecting curved resonator mirrors is in the wavelength regime which leads to a large frequency spacing between longitudinal resonator modes - comparable with the emission width of the dye molecules - so that only photons of a fixed longitudinal mode number $q$ can be found in the resonator. Then the longitudinal mode number is frozen out and the two remaining transverse modal quantum
Figure 2. (a) Thermal emission in a blackbody radiator, for which the transition energy of the atoms in the cavity walls is of order of the thermal energy $k_B T$. Processes such as a thermal excitation of a wall atom followed by photon emission occur. The effect of this combined process is to create one photon. Clearly, the photon number here is not conserved. (b) In our experiment the photon energy is separated from the thermal energy. The used molecules have a quantized electronic structure with transition energy near or above the cavity cutoff. Thermal excitation of an electronically excited molecular state is suppressed in the limit of $\hbar \omega_{cutoff} \gg k_B T$, and the (average) photon number is conserved.

numbers make the photon gas effectively two-dimensional. Moreover, the dispersion relation becomes modified with respect to free space, and acquires particle-like, i.e., quadratic, character. This is indicated in Figure 1b. The frequency of the transverse TEM$_{00}$-mode here becomes the lowest populated eigenfrequency, which acts as a cutoff frequency. Further, the mirror curvature leads to a harmonic confinement of the photon gas (Figure 1c). Thermal equilibrium of the photon gas is reached by repeated absorption and emission processes in the dye molecules. The photon gas thermalizes to the rovibrational temperatures of the molecules, which are at room temperature. We expect that the frequency distribution of the photon gas extends over a width of $k_B T/h$ above the cutoff frequency. Essential to the experiment is that the thermalization process conserves the particle number, because thermal emission of photons is negligible in the limit of a cutoff frequency ($\hbar \omega_{cutoff} \approx 2.1$ eV for light in the green spectral region) being much above the thermal energy (1/40 eV at room temperature). The dye molecules have quantized electronic excitation levels, with transition energy near or above the cutoff, whose thermal excitation is suppressed by a factor of order exp$(-\hbar \omega_{cutoff}/k_B T) \approx 10^{-36}$; see also Figure 2.

One can show that the photon gas confined in the resonator is formally equivalent to a harmonically confined two-dimensional gas of massive photons with effective mass $m_{eff} = \hbar \omega_{cutoff}/c^2 = \hbar k_s/c$, where $\omega_{cutoff}$ denotes the cutoff frequency [18]. The photon energy in the paraxial limit and in the limit of negligible interaction is

$$E \approx m_{eff} c^2 + \frac{\hbar^2 k_t^2}{2m_{eff}} + \frac{1}{2} m_{eff} \Omega^2 r^2,$$  

where $k_t$ and $k_s$ denote the transversal and longitudinal wavenumbers of the photon, $c$ the speed of light in the medium and $\Omega = c/\sqrt{D_0 R/2}$ is the trap vibrational frequency, with $R$ the mirror radius and $D_0$ the mirror spacing. For such a two-dimensional, harmonically confined system it is known that a Bose-Einstein condensate exists [20, 21]. This is expected if the particle number $N$ exceeds a critical particle number

$$N_c = \frac{\pi^2}{3} \left(\frac{k_B T}{\hbar \Omega}\right)^2.$$
Figure 3. Experimental setup for observation of Bose-Einstein condensation of photons in a dye-filled optical microcavity. The typically used spacing between the mirrors is $D_0 \approx 1.46 \mu m$, corresponding to the $q = 7$ longitudinal mode, and the TEM$_{00}$ transverse mode of this manifold acts as a low-frequency cutoff for the two-dimensional photon gas. The dye-filled resonator is pumped with radiation near 532 nm wavelength.

The typical trap vibrational frequency is $\Omega \approx 2\pi \cdot 4 \times 10^{10}$ Hz, and for $T = 300$ K we arrive at an expected critical particle number $N_c \approx 77,000$. The possibility to observe a BEC at room temperature can be understood from the effective mass $m_{\text{eff}} = \hbar \omega_{\text{cutoff}} / c^2 \approx 4 \times 10^{-36}$ kg being 10 orders of magnitude smaller than the mass of the rubidium atom, which significantly increases the phase transition temperature with respect to the atomic gas case.

Our experimental setup is shown in Figure 3. The optical resonator consists of two high reflecting dielectric mirrors with spherical curvature ($R = 1$ m). One of the mirrors is cut to a 1 mm $\times$ 1 mm surface size to allow for a distance between mirrors in the micrometer regime ($D_0 \approx 1.46 \mu m$), as measured from the cavity free spectral range, despite the mirrors’ curvature. The resonator is filled with a drop of liquid dye (rhodamine 6G or PDI) dissolved in an organic solvent. The resonator setup is pumped with a laser beam near 532 nm wavelength inclined at 45° to the cavity axis. The pumping is performed with an external laser beam because, other than in a perfect “photon box” [22], losses from coupling to optical modes not confined in the cavity, non-radiative decay and finite cavity finesse occur, which must be compensated for. We model the pumping as filling a reservoir of electronic excitations in the dye that can exchange particles with the photon gas. Thus the photon gas is understood as an open system, in the sense of a grand-canonical ensemble, with the thermalization originating from particle exchange with a reservoir of dye excitations being in equilibrium. The latter is characterized by rovibrational molecular states that are highly equilibrated both in the lower and in the upper electronic level due to subpicosecond relaxation from frequent collisions with solvent molecules [23]. This process efficiently decorrelates the states of absorbed and emitted photons within the nanosecond electronic lifetime of the dye, and leads to a temperature dependent absorption and emission spectral profile that is responsible for the thermalization. To relax both spatially and spectrally to an equilibrium distribution, a photon has to scatter several times off molecules before being lost.

3. Experimental results

In initial experiments with the dye-filled optical microresonator, we have tested for thermalization of the two-dimensional photon gas [18]. The observed photon spectra could be well described by Bose-Einstein distributed photon energies, showing that photon loss is sufficiently slow and equilibrium is reached. The chemical potential in this low density regime (well below the threshold to a BEC) is strongly negative. Moreover, a spatial concentration of light into the centre was observed, which is a consequence of the thermalization within the effective photon trapping potential, formed by curved mirrors. This behaviour is evident from the analogy with a trapped gas of material particles of finite temperature, and we expect that the effect can have prospects in the concentration of diffuse solar light [24].
Figure 4. (a) The connected circles show measured spectral intensity distributions for different pump powers. The legend gives the optical intracavity power, determining the photon number. On top of a broad thermal wing, a spectrally sharp condensate peak at the position of the cavity cutoff is visible above a critical power $P_{c, \text{exp}}$. The observed peak width is limited by the spectrometer resolution. The inset gives theoretical spectra based on Bose-Einstein distributed transverse excitations. (b) Images of the radiation emitted along the cavity axis, below (top) and above (bottom) the critical power. In the latter case, a condensate peak is visible in the centre.

More recently, we have operated the dye-filled microcavity in the high photon density regime, for which the optical pump beam was acoustooptically chopped to 0.5 µs long pulses (at least two orders of magnitude longer than the thermalization time) to avoid excessive population in the dye molecule triplet states. The repetition rate was 8 ms. Figure 4a shows typical spectra of the photon gas for different pumping rates $[19]$. While at small pumping rates the observed spectrum resembles a Boltzmann distribution above the cutoff frequency, near the expected phase transition a small shift of the spectral maximum towards the cutoff frequency is observed, and the spectrum resembles more that of a Bose-Einstein distribution. At optical intracavity powers above the critical value, in addition a spectrally sharp peak at the position of the cutoff frequency is visible, the Bose-Einstein condensate. The observed frequency width of the condensate peak is limited by the spectral resolution of our spectrometer. The experimental results are in good agreement with theoretical spectra based on a Bose-Einstein distribution of photon energies, see the inset of the figure. The observed critical photon number is $(6.3 \pm 2.4) \times 10^4$. Figure 4b shows spatial images of the photon gas below (top) and above (bottom) the critical power. Both images show a shift from the yellow spectral regime for light near the axis towards the green (with higher photon energy) for the radiation emitted off-axis. This is due to the higher energy of cavity resonances with large transverse momentum and correspondingly large mode diameter. Above the critical power, a bright spot is visible in the centre, indicating a macroscopically populated ground state (TEM$_{00}$) mode. The diameter of this condensate peak is $(14 \pm 3)$ µm, which is to be compared to an expected width of 12.2 µm for a noninteracting gas. Furthermore, we observe that the width of the condensate peak increases for larger photon numbers $[19]$, which is attributed to a weak repulsive interaction.
Figure 5. The data points give the optical intracavity power at criticality for three different curvatures of the cavity mirrors, and the line is the theoretical prediction based on Eq. 2. (b) Intracavity power at criticality (lower panel) versus the mirror separation, measured in units of the longitudinal mode number $q$. The data points in the upper panel give the required optical pump power (circles) along with a fit assuming an inverse proportionality to the absorption length in the dye.

Figure 6. Spatial intensity profiles recorded with a pump beam spot spatially displaced by 50 $\mu$m from the trap centre, for different cutoff wavelengths. For data recorded with a cutoff wavelength of 610 nm (bottom profile), where reabsorption is weak, the emitted radiation is centred near the pump spot. The pump beam profile is shown by the dashed line. When tuning the cutoff to shorter wavelength values, where the reabsorption is increased, light is redistributed towards the trap centre. For data recorded with 570 nm cutoff wavelength (top profile), a sharpened peak appears, attributed to a partly condensed state with a BEC fraction of 1%.

of the photons. The origin of this is most likely thermal lensing in the dye, but in principle also a Kerr-nonlinearity would cause the same effect. Both effects can be modelled by a mean-field interaction, and from the observed increase of mode diameter, we can estimate a dimensionless interaction parameter of $\tilde{g} \approx (7 \pm 3) \times 10^{-4}$. This is much below the values $\tilde{g} \approx 10^{-2} \ldots 10^{-3}$ reported for two-dimensional atomic physics quantum gas experiments [25, 26] and also below the value $\tilde{g} \approx 10^{-3}$ given in Ref. [27], above which Kosterlitz-Thouless physics can be expected to be relevant in the harmonically trapped case. Experimentally, when directing the observed peak through a Michelson interferometer, we have not seen signatures for the phase blurring that was reported in two-dimensional atomic physics experiments [25]. This supports the conclusion that indeed a BEC is observed in the present experiment.
To test for the photon density dependence of the phase transition threshold, we have varied the resonator geometry. From Eq. 2 we arrive at an expected critical optical power $P_c = N_c \hbar \omega^2 / 2 \pi q = (\pi^2 / 12) (k_B T)^2 (\omega / \hbar c) R$, which grows linearly with the mirror radius $R$ and is independent of the number of longitudinal modes $q$ in the resonator. Figure 5a and the lower panel of Figure 5b show corresponding measurements of the critical power, with results in good agreement with the expected scaling. The upper panel of Figure 5b gives the required optical pump power to achieve the phase transition versus the number of longitudinal modes, which decreases for larger mirror spacings, as is expected from the stronger absorption of the pumping beam power in a dye film of larger thickness. The shown data is again in excellent agreement with a model assuming that the phase transition occurs at a constant value of the two-dimensional photon density, as anticipated for a photon BEC. This is in strong contrast to results from ‘thresholdless’ dye-based optical microlasers, for which an increase of the threshold with mirror separation was reported [28, 29].

In further measurements, we have investigated the phase transition for a spatially mismatched pumping geometry to study the efficiency of the thermalization, which is expected to drive the photon gas into the trap centre. This effect is not known in laser models, but has been seen in the context of polariton condensation [6]. For our measurement, the pump beam was displaced 50 µm from the trap centre. Figure 6 shows a series of spatial intensity profiles recorded for a pumping power slightly above threshold, for different values of the cavity cutoff wavelength $\lambda_{\text{cutoff}} = 2\pi c / \omega_{\text{cutoff}}$, which tunes the efficiency of thermalization. The lower graph gives results recorded with 610 nm cutoff wavelength, for which the maximum of the fluorescence is centred at the position of the pump spot (shown by a dashed line). The weak reabsorption in this wavelength range prevents efficient photon thermalization into the trap centre. When the cutoff is moved to shorter wavelengths, the profiles become more symmetric around the trap centre, as expected from the increased reabsorption in this wavelength range. For a cavity cutoff near 570 nm, the photon gas appears fully thermalized and we observe a narrow peak at the position of the TEM$_{00}$ mode on top of the thermal wing, which is attributed to a BEC with a 1% condensate fraction. The measurements indicate that, due to the photon thermalization process, Bose-Einstein condensation can be achieved even when the pumping intensity at the position of the ground state mode is essentially zero.

4. Conclusions
In summary, evidence for a Bose-Einstein condensation of photons was obtained from (i) Bose-Einstein distributed photon energies with a massively populated ground state mode on top of a broad thermal wing, (ii) the phase transition occurring both at the expected photon density and with the predicted cavity geometry dependence, and (iii) the ground state mode emerging even for a spatially displaced pump spot. It is instructive to discuss the relation of a photon BEC to optical microlasers, which also use high finesse cavities to capture the emission of excited state atoms and molecules in a small volume [28, 29]. The low lasing thresholds and the even possible inversion-less oscillation of microlasers, however, result from the coupling of spontaneous photons into a single cavity mode - which is not the case in the work reported here. This is evident from the observed highly multimodal emission below the phase transition. The main difference between a laser and Bose-Einstein condensation remains that the latter device in contrast to the former operates in thermal equilibrium. Note that the usual condition for the emergence of a coherent peak in a laser, of the gain coefficient exceeding the loss, is here replaced by Eq. (2), which essentially is equivalent to the condition $n\lambda_{th}^2 \approx 1$, i.e. that the de Broglie wavepackets of the photon gas spatially overlap. Here $n$ denotes the two-dimensional photon density, and $\lambda_{th}$ is the de Broglie wavelength of the photon associated with the thermal
motion in the resonator plane.

An intriguing possible consequence of the particle exchange between photons and the reservoir of excited state dye molecules is, in the limit of suitable relaxation times and small photon-photon interactions, a predicted realisation of a grand-canonical ensemble, which can give rise to unusually large number fluctuations in the condensed phase [30]. We expect that the concept of coherent light generation through condensation holds promise for new light sources, including coherent sources in wavelength regions where suitable laser media are rare or unavailable, as in the EUV regime [31]. A possible suitable medium for photon thermalization in this spectral region could be high pressure noble buffer gas samples, media that our group has recently started to investigate in the context of novel redistribution laser cooling techniques [32].

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