

Statistical Physics of Bose-Einstein-Condensed Light in a Dye Microcavity

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We theoretically analyze the temperature behavior of paraxial light in thermal equilibrium with a dye-filled optical microcavity. At low temperatures the photon gas undergoes Bose-Einstein condensation, and the photon number in the cavity ground state becomes macroscopic with respect to the total photon number. Owing to a grand-canonical excitation exchange between the photon gas and the dye molecule reservoir, a regime with unusually large fluctuations of the condensate number is predicted for this system that is not observed in present atomic physics Bose-Einstein condensation experiments.

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For many problems in statistical physics, one has the freedom of choice to use different statistical ensembles for their description, as they often predict the same physical behavior in the thermodynamic limit. An interesting counterexample (among others) is Bose-Einstein condensation [1,2], where the grand-canonical description leads to unusually large condensate number fluctuations of the order of the total particle number [3–6]—in contrast to the predictions in the (micro)canonical case that is typically realized in experiments with ultracold atomic Bose gases [7]. This peculiar behavior is known as the grand-canonical fluctuation catastrophe [5,6]. In recent work, we have observed Bose-Einstein condensation of a two-dimensional photon gas in an optical microcavity [8,9]. Here, the transversal motional degrees of freedom of the photons are thermally coupled to the cavity environment by multiple absorption-fluorescence cycles in a dye medium, with the latter serving as both a heat bath and a particle reservoir. Because of particle exchange between the photon gas and the molecular reservoir, grand-canonical experimental conditions are expected to be realized in this system.

In this Letter, we discuss the thermalization mechanism and derive statistical properties of the photon condensate, including its photon number distribution, fluctuations, and intensity correlations. The main result is that photonic Bose-Einstein condensates, owing to the grand-canonical nature of the light-matter thermalization, can show unusually large particle number fluctuations, which are not observed in present atomic Bose-Einstein condensates. Our calculations are done in the limit of a noninteracting photon gas.

The system under investigation, as shown in Fig. 1(a), consists of a microresonator formed by two perfectly reflecting spherically curved mirrors enclosing a dye medium. In the cavity, optical photons are permanently absorbed and reemitted by dye molecules. For a sufficiently small mirror spacing, this process will maintain the longitudinal mode number of the photons denoted by q ; see [8–10] for a detailed description of the experiment.

The photon gas effectively becomes two-dimensional, as only the two transversal degrees of freedom remain. Throughout this Letter, we do not consider experimental imperfections as mirror losses or nonradiative decay of dye excitations. Moreover, we do not consider coherent time evolution of the combined light-dye system (strong coupling) which would result in polaritonic eigenstates. Under the experimental conditions of Refs. [8,9], this is inhibited by a rapid, collision-induced decoherence process.

There is a close connection to the statistical physics of the atomic two-dimensional Bose gas, which can be seen by expressing the photon energy as a function of the longitudinal (k_z) and transversal wave number (k_r): $E = \hbar\tilde{c}\sqrt{k_z^2 + k_r^2}$, with \tilde{c} as the speed of light in the medium. The boundary condition in the z direction is incorporated by an ansatz $k_z(r) = q\pi/D(r)$, where $D(r) = D_0 - 2(R - \sqrt{R^2 - r^2})$ is the mirror separation at a distance r from the optical axis and R is the radius of curvature. In a paraxial approximation, with $k_r \ll k_z(0)$ and $r \ll R$, this yields [10]

$$E \simeq m\tilde{c}^2 + \frac{(\hbar k_r)^2}{2m} + \frac{m\Omega^2}{2}r^2, \quad (1)$$

which is formally equivalent to the energy of a two-dimensional harmonic oscillator when defining a photon mass $m = \hbar k_z(0)/\tilde{c}$ and a trapping frequency $\Omega = \tilde{c}/\sqrt{D_0 R}/2$. The photon gas can thus be mapped onto a (2D) gas of massive bosons in a harmonic trap—a system known to undergo Bose-Einstein condensation (BEC) at a nonzero temperature [11,12].

Thermal equilibrium and condensation.—First we discuss the thermalization process in more detail. Thermal equilibrium between photons and dye molecules will be shown to arise under two conditions: (i) The Einstein coefficients of the dye medium fulfill the Kennard-Stepanov law, and (ii) chemical equilibrium between photons, excited, and ground-state molecules is achieved.

(i) The Kennard-Stepanov law [13,14] (and references therein) relates the Einstein coefficients of stimulated absorption and emission, and can be stated in the form

$$\frac{B_{21}(\omega)}{B_{12}(\omega)} = \frac{w_{\downarrow}}{w_{\uparrow}} e^{-\hbar(\omega - \omega_0)/k_B T}, \quad (2)$$

where $B_{12,21}(\omega)$ are the Einstein coefficients of absorption and emission, ω_0 is the zero-phonon line of the dye, and $w_{\downarrow,\uparrow} = \int_{\epsilon \geq 0} \mathcal{D}_{\downarrow,\uparrow}(\epsilon) \exp(-\epsilon/k_B T) d\epsilon$ are statistical weights related to the rovibronic density of states $\mathcal{D}_{\downarrow,\uparrow}(\epsilon)$ of the ground (\downarrow) and excited (\uparrow) dye state. This law is often fulfilled for dyes in liquid solution and goes back to a thermalization process of the rovibronic dye state due to frequent collisions with solvent molecules [15].

(ii) The excitation exchange between photon gas and molecules can be seen as a photochemical reaction of the type $\gamma + \downarrow \rightleftharpoons \uparrow$. Here γ stands for a photon, \downarrow for a ground-state molecule, and \uparrow for an excited molecule. The energy of optical photons is well above thermal energy at room temperature, i.e., $E \gg k_B T$, and the conversion of ground-state molecules into excited molecules by thermal fluctuations is thus negligible. In this situation, the total number of photons and excited molecules is not adjusted by temperature as for blackbody radiation but is a conserved quantity (it depends on the initial state of the photon gas and the molecular medium). The chemical potential of the photons μ_γ then can become nonzero. In equilibrium, μ_γ is related to the other chemical potentials by $\mu_\gamma + \mu_{\downarrow} = \mu_{\uparrow}$ [16]. Starting from this, the photon chemical potential μ_γ can be shown [15] to be related to the excitation level $\rho_{\uparrow}/\rho_{\downarrow}$ in the dye medium by

$$e^{\mu_\gamma/k_B T} = \frac{w_{\downarrow}}{w_{\uparrow}} \frac{\rho_{\uparrow}}{\rho_{\downarrow}} e^{\hbar\omega_0/k_B T}. \quad (3)$$

Here $\rho_{\downarrow,\uparrow}$ are the densities of ground- and excited-state molecules. The chemical potential μ_γ is spatially homogeneous in equilibrium (as is the temperature), from which follows that the excitation level of the medium $\rho_{\uparrow}/\rho_{\downarrow}$ is also position-independent.

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 $K = (n_0^K, n_1^K, n_2^K, \dots)$. The mode occupation numbers are frequently 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 \mathbf{r} , denoted by $R_{12}^{K,i}(\mathbf{r})$ and $R_{21}^{K,i}(\mathbf{r})$, respectively, have the form

$$R_{12}^{K,i}(\mathbf{r}) = B_{12}(\omega_i) u^i(\mathbf{r}) \rho_{\downarrow} n_i^K, \quad (4)$$

$$R_{21}^{K,i}(\mathbf{r}) = B_{21}(\omega_i) u^i(\mathbf{r}) \rho_{\uparrow} (n_i^K + 1), \quad (5)$$

where $u^i(\mathbf{r})$ is the spectral energy density of one photon in mode i . We first assume a grand-canonical ensemble limit;

i.e., we consider the number of dye molecules as sufficiently large that the change of the excitation level $\rho_{\uparrow}/\rho_{\downarrow}$ from photon absorption and emission can be neglected. Thus, the species densities $\rho_{\downarrow,\uparrow}$ are treated as fixed parameters.

In the theory of random walks, a well-known detailed balance criterion exists that determines if the rates given by Eqs. (4) and (5) lead to equilibrium [17,18]. Suppose that a state K' emerges from state K by the absorption of a photon in mode i , with $n_i^{K'} = n_i^K + 1$, and accordingly that K emerges from K' by an emission process into this mode. The corresponding (local) rates are $R_{12}^{K,i}(\mathbf{r})$ and $R_{21}^{K',i}(\mathbf{r})$; and with Eqs. (4) and (5), their ratio is given by $R_{12}^{K,i}(\mathbf{r})/R_{21}^{K',i}(\mathbf{r}) = B_{12}(\omega_i)\rho_{\downarrow}/B_{21}(\omega_i)\rho_{\uparrow}$. Thermal equilibrium will be reached, if this ratio is given by the Boltzmann factor of the energy difference between K and K' [18], i.e.,

$$\frac{B_{12}(\omega_i)}{B_{21}(\omega_i)} \frac{\rho_{\downarrow}}{\rho_{\uparrow}} = e^{(\hbar\omega_i - \mu_\gamma)/k_B T}. \quad (6)$$

If one now applies the Kennard-Stepanov relation, Eq. (2), and assumes chemical equilibrium, Eq. (3), the detailed balance condition Eq. (6) is indeed verified. Thus, the state of the photon gas will thermalize to the temperature of the dye solution T at a chemical potential μ_γ related to the excitation level of the dye molecules. In particular, the average occupation number of mode i , denoted by \bar{n}_i , can be determined by balancing the average absorption and emission rates at a given cavity position. This gives the expected Bose-Einstein distribution $\bar{n}_i = \{\exp[(\hbar\omega_i - \mu_\gamma)/k_B T] - 1\}^{-1}$. For atomic Bose gases, one typically omits the rest energy mc^2 from the Hamiltonian. We will adopt this convention for the photon gas by removing the ground-state energy E_{q00} from all energies including the chemical potentials. With $\mu = \mu_\gamma - E_{q00}$, the Bose-Einstein distribution becomes

$$\bar{n}(u) = \frac{g(u)}{e^{(u-\mu)/k_B T} - 1}, \quad (7)$$

where $u = E - E_{q00} = 0, \hbar\Omega, 2\hbar\Omega, \dots$ is the reduced photon energy, and we have used the degeneracy $g(u) = u/\hbar\Omega + 1$ to combine all modes of the same energy; see Fig. 1(b). For the sake of simplicity, we neglect the polarization degeneracy of each mode throughout this Letter, which would give an additional degeneracy factor of 2. This case is discussed in the Supplemental Material [15]. For this system, a phase transition is expected at a critical temperature of [11,12]

$$T_c = \frac{\sqrt{6}\hbar\Omega}{\pi k_B} \sqrt{\bar{N}} = \frac{\sqrt{12}\hbar\tilde{c}}{\pi k_B} \sqrt{\frac{1}{D_0} \frac{\bar{N}}{R}}, \quad (8)$$

at which the ground-state occupation $\bar{n}_0 \equiv \bar{n}(0)$ becomes a macroscopic fraction of the total average photon number \bar{N} .

Condensate fluctuations.—For grand-canonical Bose-Einstein condensation, i.e., in the presence of an infinitely

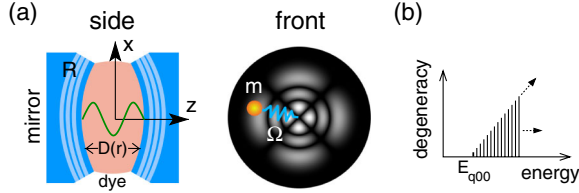


FIG. 1 (color online). (a) Optical microresonator enclosing a dye medium (left). For a fixed longitudinal mode number q , the photon gas can be mapped onto a two-dimensional gas of massive particles (effective mass m) confined in a harmonic trap with trapping frequency Ω (right). (b) Scheme of the density of states in the resonator.

large particle reservoir, condensate number fluctuations of the order of the total particle number occur [3–6]. However, no excess fluctuations occur in the (micro)canonical ensemble, which is usually realized in atomic BEC experiments [7]. For the photon Bose-Einstein condensate, the ground-state mode is coupled to the electronic transitions of a given number of dye molecules. In this way, the condensate exchanges excitations with a reservoir of size M , which is given by the product of dye concentration and ground-state mode volume.

We start with the master equation for the probability $p_n = p_n(t)$ to find n ($\equiv n_0$); photons in the ground state at time t . The flow of probability between the ground mode and its reservoir is governed by [19]

$$\dot{p}_n = R_{n-1}^{21} p_{n-1} - (R_n^{12} + R_n^{21}) p_n + R_{n+1}^{12} p_{n+1}, \quad (9)$$

where $R_n^{12} = \hat{B}_{12}(M - X + n)n$ is the rate of absorption and $R_n^{21} = \hat{B}_{21}(X - n)(n + 1)$ the rate of stimulated and spontaneous emission for a configuration with n photons, $X - n$ electronically excited molecules, and $M - X + n$ ground-state molecules. The excitation number X is the sum of the ground mode photon number and the number of excited molecules in the reservoir. In this calculation, X is treated as a constant [15]; i.e., it is not expected to perform large fluctuations on its own. The rates R_n^{12} and R_n^{21} follow from Eq. (4) and (5) by integrating over volume and setting $\hat{B}_{12,21} := B_{12,21}(E_{q00}/\hbar)u^{q00}(\mathbf{0})$, where $u^{q00}(\mathbf{0})$ is the spectral energy density of the ground mode on the optical axis. For large times, $p_n(t)$ is expected to become stationary, $\dot{p}_n(\infty) = 0$, and approach its equilibrium value $\mathcal{P}_n := p_n(\infty)$. In this asymptotic case, the solution of the master equation (9) is found to be $\mathcal{P}_n = \mathcal{P}_0 \prod_{k=0}^{n-1} R_{k+1}^{21}/R_k^{12}$, which is

$$\frac{\mathcal{P}_n}{\mathcal{P}_0} = \frac{(M - X)! X!}{(M - X + n)! (X - n)!} \left(\frac{\hat{B}_{21}}{\hat{B}_{12}} \right)^n. \quad (10)$$

This photon number distribution can be used to obtain both the average ground-state occupation and its fluctuations. We consider experimental conditions, where the temperature of the system is varied, with the total average photon

number \bar{N} (in all modes) being fixed. Correspondingly, upon temperature variations the excitation level ρ_1/ρ_1 of the dye medium and the total ground mode excitation parameter X have to be readjusted (see Supplemental Material [15]).

Figure 2(a) shows the ground mode occupation \bar{n}_0/\bar{N} (solid lines) as a function of the reduced temperature T/T_c for a constant average photon number of $\bar{N} = 10^4$, six reservoir sizes from $M_1 = 10^8$ over $M_2 = 10^9$ up to $M_6 = 10^{13}$, and a dye-cavity detuning of $\hbar\Delta/k_B T_c := (E_{q00} - \hbar\omega_0)/k_B T_c = -4.35$, which adjusts $\hat{B}_{21}/\hat{B}_{12}$ via Eq. (2) and is experimentally realizable [8,9]. Upon reaching criticality, the ground mode occupation becomes a macroscopic fraction of the total photon number. The occupation level closely follows the analytic solution $\bar{n}_0/\bar{N} = 1 - (T/T_c)^2$ [not shown in Fig. 2(a)], nearly independent of the reservoir size M [the solid lines for different M essentially overlay in Fig. 2(a)]. The dashed lines shows the fraction \bar{n}_0/X of ground mode occupation and total excitation number versus temperature, which reveals that the vast majority of excitations is stored as electronic excitations in the medium down to relatively low temperatures. Figure 2(b) gives the normalized ground mode fluctuations in terms of the zero-delay autocorrelation function $g^{(2)}(0) = \langle n_0(n_0 - 1) \rangle / \bar{n}_0^2$ versus temperature. For $T \geq T_c$, one finds the usual case of strong intensity fluctuations with $g^{(2)}(0) = 2$, accompanied by a Bose-Einstein-like photon number distribution. Interestingly, for large reservoir sizes M , the intensity correlation function remains at this value even at temperatures below T_c , i.e., when condensation sets in. In this regime, large condensate number fluctuations occur due to grand-canonical particle exchange with the molecular reservoir. At even lower temperatures, $T \ll T_c$, the fluctuations are damped, and one finds $g^{(2)}(0) \simeq 1$, accompanied by a

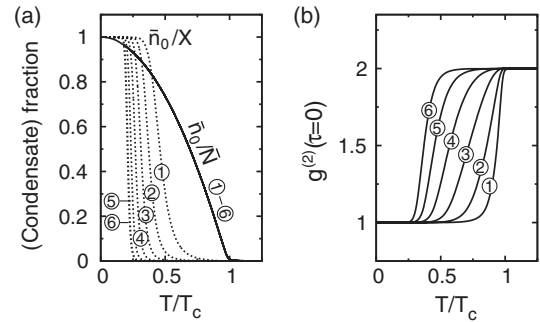


FIG. 2. (a) Condensate fraction \bar{n}_0/\bar{N} (solid lines) and fraction \bar{n}_0/X (dashed lines) as a function of the reduced temperature T/T_c for a photon number of $\bar{N} = 10^4$, six reservoir sizes from $M_1 = 10^8$, $M_2 = 10^9$ up to $M_6 = 10^{13}$, and a dye-cavity detuning of $\hbar\Delta/k_B T_c = -4.35$. (b) Normalized condensate fluctuations $g^{(2)}(0) = \langle n_0(n_0 - 1) \rangle / \bar{n}_0^2$ as a function of T/T_c for various M [as in (a)], revealing large number fluctuations even far below T_c .

Poissonian photon statistics. We have no indication that the crossover from Bose-Einstein to Poissonian statistics is accompanied by an additional phase transition, i.e., a non-analytic behavior of thermodynamic functions.

In general, the photon number distribution Eq. (10) can be either Bose-Einstein-like, Poisson-like, or an intermediate case. It will be Bose-Einstein-like if the molecular reservoir is so large that the ratio $\mathcal{P}_{n+1}/\mathcal{P}_n = (\hat{B}_{21}/\hat{B}_{12}) \times (X - n)/(M - X + n + 1)$ is approximately constant for all relevant photon numbers n . Then \mathcal{P}_n has its peak value at $n = 0$ and decays exponentially like a geometric series, with $\mathcal{P}_n/\mathcal{P}_0 \approx [(\hat{B}_{21}/\hat{B}_{12})X/(M - X)]^n$. Thus, the most probable event is to find no photons at all. Note that this can be achieved by increasing M while keeping the excitation level $\rho_1/\rho_0 \approx X/(M - X)$ fixed (which maintains μ , \bar{n}_0 , and \bar{N}). For Poissonian statistics, \mathcal{P}_n has its maximum at a nonzero n . Because of the smooth crossover, the distinction between these two statistical regimes is not unambiguous. However, a natural choice is to consider the point at which “finding zero photons” ceases to be the most probable event. This occurs at $\mathcal{P}_0 = \mathcal{P}_1$ (compare also to the laser threshold described in Ref. [20]) and with Eq. (10) corresponds to $(M + 1)/X = 1 + \hat{B}_{21}/\hat{B}_{12}$. The temperature T_x at which this occurs, for fixed \bar{N} and M , can be shown to fulfill the transcendental equation

$$\bar{N} - \frac{\pi}{6} \left(\frac{k_B T_x}{\hbar \Omega} \right)^2 \approx \sqrt{\frac{M}{(1 + e^{\hbar \Delta/k_B T_x})(1 + e^{-\hbar \Delta/k_B T_x})}}. \quad (11)$$

For zero dye-cavity detuning $\Delta = 0$, one finds the analytic solution $T_{x,\Delta=0} \approx T_c \sqrt{1 - \sqrt{M}/2\bar{N}}$, provided that $\sqrt{M}/2\bar{N} < 1$, while for general detunings Δ , Eq. (11) has to be solved numerically. Figure 3 gives a phase diagram, where solutions for three different cases $\sqrt{M}/2\bar{N} = \sqrt{0.1}$, 1, and $\sqrt{10}$ are marked as dashed lines, which separate two regimes of the condensate, denoted by C(I) with Bose-Einstein-like photon statistics and C(II) with Poisson statistics, respectively. In terms of second-order correlations, the dashed lines correspond to $g^{(2)}(0) \approx 1.571$. Note that both T_c and T_x are conserved in a thermodynamic limit $\bar{N}, M, R \rightarrow \infty$ that includes $\bar{N}/R = \text{const}$ and $\sqrt{M}/\bar{N} = \text{const}$. For a fluctuating condensate in the C(I) regime, the time-dependent intensity correlations $g^{(2)}(\tau) = \langle n_0(t)[n_0(t + \tau) - 1] \rangle / \bar{n}_0^2$ decay like $g^{(2)}(\tau) = 1 + \exp(-\tau/\tau_c^{(2)})$, with $\tau_c^{(2)} = \bar{n}_0/\hat{B}_{21}X$ as the second-order correlation time [15], similar to the photon bunching of thermal emitters.

In conclusion, we have studied the thermalization and condensation of photons in equilibrium with a dye microcavity. Our analysis predicts a regime with unusually large condensate fluctuations, not observed in present atomic BEC experiments. Moreover, it adds to a classification of photonic BEC in relation to other light sources. In general, Bose-Einstein condensation applies to a system in thermo-

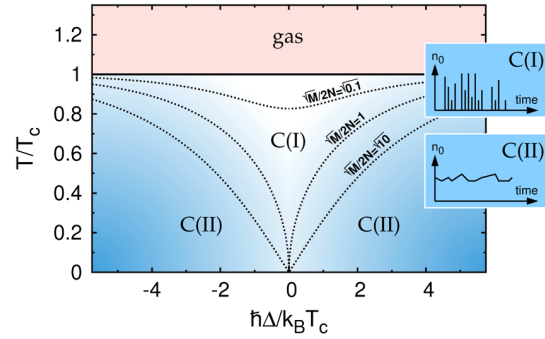


FIG. 3 (color online). Phase diagram of the two-dimensional photon gas for fixed average photon number \bar{N} , as a function of the reduced temperature T/T_c and the dye-cavity detuning $\hbar\Delta/k_B T_c$. The solid line marks the BEC phase transition. The dashed lines (three cases are shown) separate two regimes: a condensate with large number fluctuations and a BE-like photon number distribution C(I), and a nonfluctuating condensate obeying Poisson statistics C(II). The temperature of the crossover C(I)-C(II) depends on the ratio \sqrt{M}/\bar{N} , with the reservoir size M as the number of dye molecules in the mode volume of the ground state. The insets give a sketch of the corresponding temporal evolution of the condensate photon number $n_0(t)$.

dynamic equilibrium, in contrast to lasing which normally occurs under nonequilibrium conditions. The latter is brought about by a violation of chemical equilibrium which occurs if photons are lost (e.g., transmitted) instead of being reabsorbed by the medium. Additionally, BEC shows a pronounced dependence on the density of states of a system intrinsically requiring a statistical multimode treatment, whereas single-mode treatments are fully sufficient to describe the (single-mode) behavior of a wide class of lasers. If one considers a zero-delay autocorrelation function of $g^{(2)}(0) = 1$ to be an essential ingredient of lasing, as is common in laser literature [21], the intensity fluctuations (photon statistics) can give a further distinguishing feature between lasing and photonic BEC, as the latter is not necessarily accompanied by a damping of fluctuations. We note that, for other lasing threshold definitions based on the internal operation of the device, also laser light with non-Poissonian photon statistics can occur [22,23]. This ambiguity does not occur in the case of photonic BEC, where the phase transition is uniquely defined.

For the future, it will be important to study the statistics of an interacting photon gas. In general, interactions can suppress above-Poissonian intensity fluctuations [6]. One here has to distinguish between thermo-optically induced interactions [8], which are comparatively slow, and ultrafast Kerr lensing. Another line of research could also be dedicated to the (interaction-induced) superfluidity of the photon condensate.

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