General theory of Bose-Einstein condensation applied to an ideal quantum gas of photons in an optical microcavity

Eberhard E. Müller[*]

Institut für Theoretische Physik, Technische Universität Berlin, Hardenberg Strasse 36, D-10623 Berlin, Germany

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This paper provides, first, a succinct theoretical derivation of Bose-Einstein condensation in a grand-canonical ideal photon gas elaborating on previous results [Müller, Ann. Phys. **184**, 219 (1988); Physica A (Amsterdam, Neth.) **139**, 165 (1986)] additionally including new results on the condensate function and, second, applies this framework to consistently explain experimental findings on Bose-Einstein condensation of photons in an optical microcavity [Klaers, Schmitt, Vewinger, and Weitz, Nature **468**, 545 (2010)]. A grand-canonical photon gas is realized by a steady state of a photon flux deviating from a canonical thermodynamic Planck equilibrium. The theoretical approach presented here invites to significantly widen the experimental framework for Bose-Einstein condensation of photons including three-dimensional photon resonators and thermalization mechanisms different from a dye medium in the cavity.

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I. INTRODUCTION

At first glance, a Bose-Einstein condensation (BEC) in an ideal photon gas seems unreasonable. Einstein's original prediction of this fundamental quantum phase transition refers to a monoatomic ideal quantum gas [1]. Using massive particles, he was able to control the number density in the gas. He fixed the temperature and increased the particle number density beyond the saturation value. Thus he deviated from a canonical gas, and passed over to a grand-canonical gas where temperature and number density were two independent thermodynamical variables. Beyond the saturation state, in the critical regime, the quantum phase transition emerged which Einstein compared to the condensation of vapor. The excess atoms transit to a "state without kinetic energy" [1]. It is this point that makes an ideal photon gas basically differ from an ideal monoatomic gas. While both photons and neutral atoms with an even number of neutrons do have integer spin, thus qualifying as bosons, their rest masses belong to different categories: In contrast to atoms, free photons are characterized by rest mass zero. The two parameters spin 1 (helicity 1) and zero rest mass label an irreducible representation of the Poincaré-group, the space-time symmetry group of the special theory of relativity, thus defining the photons among the free elementary particles [2]. The zero rest mass of the free photons presents a severe conceptual problem for a photon condensation. A photonic occupation of the state without kinetic energy seems to have no substance at all. As far as we know, Einstein never publicly touched the question of a possible photon condensation, neither in the negative nor in the affirmative respect [3,4].

In this paper, we closely inspect the zero rest mass problem for a Bose-Einstein condensation in an ideal photon gas. This problem is intrinsically related to the chemical potential of the photons. We shall provide an appropriate thermodynamic framework to resolve it. The second fundamental problem is the thermalization in an ideal photon gas. Here we recall Planck's widely accepted thermalization concept for an ideal photon gas, and we give a quantum perspective for a thermalization mechanism. Thus we are prepared to demonstrate Bose-Einstein condensation in a grand-canonical thermodynamic equilibrium of an ideal photon gas. Eventually, we calculate the form of the condensate function in the limit of an infinite number of infrared photons. The approach presented here relies on a number of mathematical derivations that are present in previous works [5,6], but are likely not common knowledge for most readers. Therefore, a comprehensive and mathematically self-contained derivation is presented in this work. The mathematical technique is inspired by [7] where the focus aimed at nonrelativistic BEC-systems.

The theoretical results of the present paper are used to consistently explain experimental findings on Bose-Einstein condensation of photons in an optical microcavity [8]. In addition, we prove that the photon condensate is accumulated in the center of the resonator. To establish this, we have to calculate the grand-canonical entropy density of the photon gas: The photon condensate does not contribute to the entropy density [9].

The theoretical approach presented here invites to significantly widen the experimental framework for BEC of photons including three-dimensional photon resonators and thermalization mechanisms different from a dye medium in the cavity.

II. RECENT APPROACHES TO REALIZE BEC IN PHYSICAL SYSTEMS WITH PARTICIPATION OF PHOTONS

Considering interacting systems avoids the conceptual problem of BEC in an ideal photon gas. Photons can be strongly coupled with solid-state excitons forming polaritons,

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[*] eberhard.mueller@campus.tu-berlin.de

i.e. bosonic quasiparticles. Such dressed photons have an effective mass and interact with each other, thus providing a thermalization mechanism in an appropriate gas configuration. Balili *et al.* [10] were the first to successfully observe substantial signatures of BEC in a polariton gas where the polaritons were confined in a trap giving rise to an effectively two-dimensional quantum gas. They followed previous work by Deng *et al.* [11], and Kasprzak *et al.* [12]. The theory of polariton BEC was introduced by Imamoglu *et al.* [13]. According to [14,15], an appropriate low-dimensional harmonic trap system qualifies for BEC.

Different from the above polariton approach, there is a possibility for the BEC of photons where the photons are weakly coupled to a thermodynamic reservoir of dye molecules [8]. The dye molecules filled into an optical microcavity repeatedly absorb and re-emit photons, thus providing the thermalization mechanism needed to realize the phase transition. If the number density of the grand-canonical photon gas [16] exceeds a critical value, the excess of photons occupies the ground state of the resonator macroscopically. The design of the optical microcavity includes a photon trap that introduces a preference direction for a standing electromagnetic wave. That fixes a longitudinal degree of freedom for the photons in the cavity (paraxial light). With respect to this longitudinal degree of freedom the photons build up a classical standing electromagnetic wave maintaining their relativistic mass. The longitudinal degree of freedom is "frozen out". With respect to the transversal degrees of freedom, the photons display their quantum character. Thus the experimental setting in [8] constitutes a two-dimensional quantum gas of photons carrying an effective mass. This view splits the three-dimensional quantum character of photons based on an irreducible representation of the Poincaré-group into a two-dimensional quantum part and a one-dimensional classical part, thereby arriving at a semiclassical approximation of the photons. To explain BEC in their photon system, Klaers *et al.* [8] relied on a two-dimensional system and referred to the results in [14,15] in a way similar to the polariton case.

The effective mass of the harmonically trapped photons depends on the size of the cavity and vanishes for an infinitely large cavity. Thus, in the infinite volume limit, the number conservation gets unfounded, which, up to now, is commonly believed to be a prerequisite for BEC [17].

III. OPTICAL MICROCAVITY SYSTEM AS AN IDEAL PHOTON GAS

In this paper we adopt a different view on the weakly coupled photons in [8]; we demonstrate Bose-Einstein condensation in an ideal photon gas, and we thereby explain the experiment. We fully take into account the three-dimensional quantum nature of the ideal photon gas, and we shall use an asymptotic expansion including bulk contribution and surface contribution to identify the two-dimensional photon gas in the optical microcavity of [8]. Thus we can compare the experimental results from [8] to our theoretical predictions. We shall see that our theoretical approach matches the experimental results. It is important to notice that a relativistic ideal gas admits BEC in three as well as in two dimensions while BEC does not work in a nonrelativistic two-dimensional ideal gas [5,6]. Our view of an ideal photon gas in an optical microcavity with three-dimensional volume and a corresponding two-dimensional surface area disregards the specific trap construct (curvature of the enclosing mirrors) of this experiment. This is a comfortable advantage to perform the thermodynamic limit of the photon resonator.

A. Thermalization

In the case of an ideal gas, the infinite volume limit is of particular interest to understand BEC: A quantum gas in a finite container has discrete energy levels raising the question of how the necessary energy transfer between the energy levels can be achieved. However, the infinite volume limit renders the spacing between the levels infinitesimally small. This continuous spectrum idealization, suggested in 1937 [7], removes a severe obstacle for an energetic redistribution of the gas constituents. It made Uhlenbeck withdraw his early objection to Einstein's condensation hypothesis [7,18,19]. Beyond the BEC dispute, the thermodynamic limit is a well-established means to fulfill the unchallenged desire to establish the sharp manifestation of phase transitions [7].

Strictly speaking, the continuous spectrum property on its own is not sufficient to obtain thermodynamical equilibrium. However, this property can be supplemented by a nontrivial, deep result for noncommutative algebras of observables which holds in the infinite volume limit. This result is applicable for the boson structure of an ideal photon gas. To generalize the thermodynamical Gibbs equilibrium condition to include the case of the infinite volume limit, we use the Kubo-Martin-Schwinger boundary condition (KMS condition). Now the Tomita-Takesaki theorem states a correspondence between a thermodynamic KMS state and a time automorphism group which is given by a "modular operator" [20]. This modular operator measures the noncommutativity in the observable algebra; it is intimately related to quantum correlations. Thus, the Tomita-Takesaki theorem states nothing less than that in the thermodynamical equilibrium of an infinitely large ideal quantum gas, the thermal fluctuations can be reduced to quantum fluctuations. The scale factor between the thermal and the quantum fluctuations is given by the inverse temperature.

We look on the question of thermalization in an ideal photon gas in a finite cavity more explicitly. This question had already been decisive for Max Planck's reasoning on heat radiation. For an evacuated cavity with perfectly reflecting walls filled with an arbitrary composition of light rays, he symbolically postulated the presence of a minute coal dust to transform any radiation into black body radiation [21]. The heat capacity of this "minute" black dust is assumed to be negligible compared to the heat capacity of the radiation within the cavity. For a real cavity, Planck's symbolic assumption is always met by the finiteness of the quality factor of the cavity due to the finiteness of the electric conductance of the walls. The walls do the job of the coal dust by absorbing and re-emitting light rays. (It took some time for Planck to pass from the picture of light rays to Einstein's picture of free light quanta, the photons.)

There is another general argument on thermalization which deserves to be mentioned besides Planck's minute coal dust. For any cavities, ideal cavities with perfectly reflecting walls included, we draw attention to their finite size to notice the spatial uncertainty of the photons therein. This spatial uncertainty is transformed by Heisenberg's uncertainty relation into an uncertainty of the photon momentum which induces a smearing out of the photon resonances in the cavity. They are characterized by a line width which is specific for the cavity. The photon resonances overlap, and we get probabilities for transitions between the resonances. In the ideal case of a perfectly reflecting wall, all transition probabilities are nonzero, although a number of them might be extremely small. We see that the finiteness of a container provides a means to achieve thermal equilibrium of an ideal quantum gas, potentially on a large timescale. Having established the equilibrium, the spectral temperature of an ideal photon gas in a perfect resonator will be induced by the value of the photon gas energy; this can be done by the technique of the Lagrange multipliers. Planck did not have Heisenberg's uncertainty relation at hand when working out black body radiation.

In the common case of cavities with absorbing walls, the competition between the probabilities for absorption and the probabilities for transitions dominates the smearing out due to the uncertainty relation. It is decisive to deal with this competition when constructing a photon resonator for BEC. If transition probabilities are sufficiently higher than the respective absorption probabilities, then a precondition for the possibility of building up a grand-canonical thermodynamic equilibrium of photons is fulfilled.

For the sake of concreteness, we add a more familiar thermalization setting. We use an electron gas as a thermal reservoir and couple it to the photon gas. Now Compton scattering between the photons and the electrons induces an exchange of energy between the photons establishing thermal equilibrium in the photon gas [22]. This is comparable to the collision processes between the massive particles of an ideal gas with the constituents of a heat bath that is in contact with the gas to attain thermal equilibrium. Instead of an electron gas, we can employ the nonabsorbing wall of the photon resonator as an intermediate agent between the photons: The wall is exposed to the collisions of the photons which imply a rebounding of the wall to the photon gas. Thus, again, we see an energy exchange between the photons needed for thermalization.

B. Conceptual preliminaries: Choice of the thermodynamic variables; the role of the chemical potential

We shall take the term "canonical ensemble" to refer to a state in which entropy is maximized subject to the mean energy density being fixed. To this constraint there corresponds a Lagrange parameter β which stands for the inverse temperature $\frac{1}{kT}$ with T and k being the temperature and the Boltzmann constant, respectively. We shall take the term "grand-canonical" ensemble to refer to a state in which entropy is maximized subject to the mean energy density and the mean particle number density being fixed. To the additional constraint for the mean particle number density there corresponds a second Lagrange parameter μ which is associated with the chemical potential. Only one sort of particle, the photon, is considered in this paper. For a grand-canonical photon gas in a finite-size cavity with reflecting walls, any two of the four thermodynamic variables mean energy density u, mean photon number density ρ, inverse temperature β, and chemical potential μ can be used as independent thermodynamic variables.

For the thermodynamic limit of a photon gas this is no longer true. In this limit, the lowest energy value of the photons approaches zero. A finite number density of photons with infinitesimal energy cannot build up a nonzero energy density. Therefore, in the infinite volume limit, we have to carefully distinguish between the mean energy density being fixed and the photon number density being fixed. In [6], the thermodynamic limit was performed with mean number density fixed while in [5], it was the mean energy density that was fixed for the limit procedure. An essential difference shows up. In the number density limit, the photon condensate does not contribute to the mean energy density; the finite occupation number of the lowest state has to be multiplied by the respective energy eigenvalue ϵ_1^R, with R being a characteristic cavity length tending to infinite, such that ϵ_1^R tends to zero, and therefore the potential condensate contribution to the energy density tends to zero as well (see below and [6]). Physically this is not fully satisfactory, although an onset of condensation emerges. However, in the mean energy density limit, the condensate necessarily contributes to the mean energy density while the photon number density diverges. This infrared divergence is in line with physical experience and understanding. In any case the mean energy density is experimentally accessible and controllable, as well as the temperature. In this paper we therefore adopt temperature and mean energy density as the independent thermodynamic variables thus forming the grand-canonical photon gas. Consequently the photon number density and the chemical potential are dependent variables. A number-conserving thermalization is thus no longer a necessary precondition for the BEC of photons.

In the finite photon gas system, the chemical potential μ_R of the photons depends on the size of the cavity. R shall denote a characteristic length of the cavity with V_R its volume such that $R^3 = V_R$. Let the inverse temperature β and the value $\underline{u}$ of the mean energy density function u_R of the photon gas be fixed as follows:

$$u_R(\beta,\ \mu_R(\beta,\ \underline{u})) = \underline{u}. \tag{1}$$

Equation (1) defines the chemical potential μ_R as a function of β and $\underline{u}$. We shall see that, in the critical regime where the value $\underline{u}$ exceeds $u(\beta)$ which is the mean energy density of black body radiation associated with β, the convergence rate of μ_R in the infinite volume limit turns out to be

$$\mu_R \sim \frac{1}{R^4} \quad \text{for}\, R \to \infty, \tag{2a}$$

$$\lim_{R\to\infty} \mu_R(\beta, \underline{u}) = 0, \quad \text{for}\, \underline{u} \geqslant u(\beta). \tag{2b}$$

In the limit, the chemical potential of the photon condensate is zero; there are no longer single photons, there is a new collective phase. In his pioneering work [1], Einstein already presumed the chemical potential of the condensate to be zero. For nonrelativistic Bose-Einstein condensates of an ideal boson gas, this has been proved in general by Lewis *et al.* [7]. For boson systems with a persistently gapped spectrum, and for pumped laser systems in particular, the chemical potential remains nonzero [23]. The nonequilibrium laser system can be formally transformed into a grand-canonical equilibrium system, where the chemical potential $\mu \neq 0$ absorbs the pumped shift of the spectrum [24,25]: The pumping in the laser system yields a nonzero chemical potential μ of the photons. Even in a thermodynamic limit idealization of such a system, the chemical potential remains nonzero. It can be regarded as a shifted ground state of the laser-photon system. This introduces a sharp distinction between BEC, with $\mu = 0$, and laser states, with $\mu \neq 0$. Note that in both cases the systems are described via a grand-canonical ensemble.

The crucial point of our approach is the distinction between two types of limits that appear in the critical regime. There is, first, the grand-canonical limit $R \to \infty$ for the mean energy density u_R with variable $\mu_R(\beta, \underline{u})$, and, second, there is the canonical limit $R \to \infty$ for u_R putting the chemical potential zero *a priori*. We have

$$\begin{aligned}\lim_{R\to\infty} u_R(\beta, \mu_R(\beta, \underline{u})) &\neq \\ \lim_{R\to\infty} u_R(\beta, 0) &= u(\beta), \quad \text{for } \underline{u} > u(\beta). \end{aligned} \tag{3}$$

A nonzero difference $\underline{u} - u(\beta) > 0$ allows for Bose-Einstein condensation.

IV. PHOTON CONDENSATION IN THREE AND TWO DIMENSIONS

We consider a photon gas in a finite cavity of volume V_R with reflecting walls. The lowest eigenvalue ϵ_1^R of the photon Hamiltonian for the cavity with Dirichlet boundary conditions is strictly positive, $0 < \epsilon_1^R \leqslant \epsilon_2^R \leqslant \epsilon_3^R \leqslant \ldots$, where $\epsilon_k^R, k \geqslant 2$ denote the excited modes. Assuming temperature T and mean energy density $\underline{u}$ as independent thermodynamic variables of the photon gas implies a deviation of the thermodynamic Planck equilibrium. To establish this, photons are continuously injected into the photon gas where the frequency and the power of the radiation into the cavity is suitably adjusted. Cooling the walls of the cavity the temperature of the photon gas is fixed at a chosen value. As a result the photon flux builds up a steady state of energy with some desired value $\underline{u}$ of the mean energy of the photon gas. As an example, this setting was fulfilled in [8]. The Hamiltonian of free photons in the cavity is given by

$$\hbar c\sqrt{-\Delta_R}, \tag{4}$$

with Δ_R denoting the Dirichlet Laplacian defined in the cavity; $\hbar$ is the reduced Planck constant, c the speed of light. We switch over to an energy spectrum

$$\lambda_k^R := \epsilon_k^R - \epsilon_1^R, \tag{5a}$$

with zero as the lowest value. Accordingly, we introduce a normalized chemical potential

$$\mu_R \leqslant 0. \tag{5b}$$

The lower bound zero of the spectrum (5a) remains fixed when the thermodynamic limit process is performed. This is a mathematical preconditon for the bound to possibly act as a limit point. The integrated spectral density $F_R(\lambda)$ of the photon Hamiltonian is a function of the variable λ and counts the number of the eigenvalues $\lambda_{k,\alpha}^R$ up to the variable value λ [6]; the natural number k denotes the mode numbers, and α the respective helicity of the photons

$$\begin{aligned} F_R(\lambda) &:= \frac{1}{V_R}\#\big\{(k,\alpha) \in \mathbb{N} \times (+1,-1) : \lambda_{k,\alpha}^R \leqslant \lambda\big\} \\ &= \frac{1}{3\pi^2}\left(\frac{\lambda}{\hbar c}\right)^3 - \frac{A_R}{8\pi V_R}\left(\frac{\lambda}{\hbar c}\right)^2 + O\left(\frac{\lambda}{R^2}\right); \end{aligned} \tag{6}$$

accordingly, the spectral density is [5,26]

$$\begin{aligned} dF_R(\lambda) &= \frac{1}{\pi^2}(\hbar c)^{-3}\lambda^2 d\lambda \\ &\quad - \frac{A_R}{4\pi V_R}(\hbar c)^{-2}\lambda d\lambda + O(R^{-2})d\lambda; \end{aligned} \tag{7}$$

with $\lambda_{k,\alpha}^R := \lambda_k^R$, α counting the two helicity values of the photons; A_R denotes the surface area of the cavity. The first term gives the bulk contribution to the density and the second one the surface contribution. In the following we neglect higher orders in R^{-1}. The approximation up to second order, given in (6), is valid for a large class of sufficiently smooth geometries covering most physical applications.

The Hamiltonian H_R of the photon gas is the second quantization of the free photon Hamiltonian (4) [27]. We include the two helicity values denoted by α:

$$H_R = \sum_p \epsilon_p^R b_p^* b_p, \; p = (k,\alpha) \in \mathbb{N} \times \{+1,-1\}. \tag{8}$$

The creation and annihilation operators b_p^* and b_p for the photon field obey Bose statistics. The corresponding particle number operator is

$$N_R = \sum_p b_p^* b_p. \tag{9}$$

The grand-canonical state on observables A of the photon gas is given by their expectation value $\mathbb{E}_{\beta,\mu}^R$ which is associated with the grand-canonical density matrix

$$\begin{aligned} D_{\beta,\mu}^R &= e^{-\log Z_{\beta,\mu}^R - \beta H_R + \beta\mu N_R}, \\ Z_{\beta,\mu}^R &:= \mathrm{Tr}[e^{-\beta(H_R - \mu N_R)}]; \end{aligned} \tag{10a}$$

$$\mathbb{E}_{\beta,\mu}^R[A] := \mathrm{Tr}\big[D_{\beta,\mu}^R A\big], \tag{10b}$$

where Tr denotes the trace over the states of the photon gas.

The grand-canonical expectation value of the photon Hamiltonian density gives the mean energy density of the photon gas in the cavity

$$\begin{aligned} u_R(\beta,\mu_R) &= \mathbb{E}_{\beta,\mu}^R\left[\frac{H_R}{V_R}\right] \\ &= \frac{2}{V_R}\sum_{k=1}^{\infty}\big(\lambda_k^R + \epsilon_1^R\big)\big(e^{\beta(\lambda_k^R - \mu_R)} - 1\big)^{-1}. \end{aligned} \tag{11}$$

The factor 2 sums the helicities. Using the spectral density (7), the evaluation of (11) yields [5]

$$\begin{aligned} u_R(\beta,\mu_R) &= \frac{2\epsilon_1^R}{V_R}\frac{1}{e^{-\beta\mu_R} - 1} \\ &\quad + \frac{2}{V_R}\sum_{k=2}^{\infty}(\lambda_k^R + \epsilon_1^R)\frac{1}{e^{\beta(\lambda_k^R - \mu_R)} - 1} \end{aligned}$$

$$\begin{aligned}&= \frac{2\epsilon_1^R}{V_R}\frac{1}{e^{-\beta\mu_R}-1}+\frac{2}{V_R}\sum_{k=2}^{\infty}\left(\lambda_k^R+\epsilon_1^R\right)\sum_{n=1}^{\infty}e^{-n\beta(\lambda_k^R-\mu_R)}\\ &= \frac{2\epsilon_1^R}{V_R}\frac{1}{e^{-\beta\mu_R}-1}+\sum_{n=1}^{\infty}e^{n\beta\mu_R}\frac{2}{V_R}\sum_{k=2}^{\infty}\left(\lambda_k^R+\epsilon_1^R\right)e^{n\beta\lambda_k^R}\\ &= \frac{2\epsilon_1^R}{V_R}\frac{1}{e^{-\beta\mu_R}-1}+\sum_{n=1}^{\infty}e^{n\beta\mu_R}\int_{\epsilon_2^R}^{\infty}\left(\lambda+\epsilon_1^R\right)dF_R(\lambda).\end{aligned} \tag{12}$$

The second term of $u_R(\beta, \mu_R)$ represents the excited states of the photon gas; we denote it by $u_e^R(\beta, \mu_R)$. In the third equation, the two infinite summations can be rearranged since the singular ground state of $u_R(\beta, \mu_R)$ has been isolated. We evaluate the integral and, thereby, neglect the term with ϵ_1^R which, in the infinite volume limit, tends to 0; the lower integration bound ϵ_2^R also tends to 0, for $R \to \infty$. The first two terms of the asymptotic expansion of $u_e^R(\beta, \mu_R)$ with respect to R are

$$\begin{aligned}u_e^R(\beta, \mu_R) &\sim u_e(\beta, \mu)\\ &= \sum_{n=1}^{\infty}e^{n\beta\mu}\left(\frac{1}{n^4}\frac{6}{\pi^2\hbar^3c^3\beta^4}-\frac{1}{n^3}\frac{A_R}{V_R}\frac{2}{4\pi\hbar^2c^2\beta^3}\right),\end{aligned} \tag{13}$$

where $u_e^R(\beta, 0)$ is the mean energy density of black body radiation which is constituted by the excited states of the photon gas. $u_e(\beta, 0)$ turns out to be the critical mean energy density for the photon condensation. The asymptotic expansion gives the bulk contribution $u_{\text{crit}}^{\text{bulk}}(\beta)$ and the surface contribution $u_{\text{crit}}^{\text{surface}}(\beta)$

$$u_{\text{crit}}^{\text{bulk}}(\beta) := u_e^{\text{bulk}}(\beta, 0) := \frac{6}{\pi^2\hbar^3c^3\beta^4}g_4(1), \tag{14a}$$

$$u_{\text{crit}}^{\text{surface}}(\beta) := u_e^{\text{surface}}(\beta, 0) := \frac{2}{4\pi\hbar^2c^2\beta^3}g_3(1), \tag{14b}$$

where

$$g_s(z) := \sum_{n=1}^{\infty}\frac{z^n}{n^s}, \quad \zeta(s) = g_s(1). \tag{15}$$

ζ is the Riemannian zeta function; $g_4(1) = \zeta(4) = \pi^4/90; g_3(1) = \zeta(3) = 1,20206\ldots$.

Given a temperature β, and a value $\underline{u}$ of the mean energy density. Then the chemical potential μ_R is a dependent variable determined by the equation

$$u_R(\beta, \mu_R) = \underline{u}.$$

In the bulk approximation $\mu_R \to \mu$ where the thermodynamic limit μ is a unique solution [28] of

$$\begin{aligned}&u_e^{\text{bulk}}(\beta, \mu) := \frac{6}{\pi^2\hbar^3c^3\beta^4}g_4(e^{\beta\mu}) = \underline{u}\\ &\text{if } \underline{u} \leqslant u_{\text{crit}}^{\text{bulk}}(\beta),\end{aligned} \tag{16}$$

and

$$\mu = 0 \text{ if } \underline{u} > u_{\text{crit}}^{\text{bulk}}(\beta). \tag{17}$$

Equation (17) represents the condensation regime. In this regime the mean energy density of the condensate is given by

$$\begin{aligned}&u_1 := \underline{u} - u_{\text{crit}}^{\text{bulk}}(\beta) \text{ if } \underline{u} > u_{\text{crit}}^{\text{bulk}}(\beta).\\ &\text{If } \underline{u} \leqslant u_{\text{crit}}^{\text{bulk}}(\beta), \text{ we set } u_1 := 0.\end{aligned} \tag{18}$$

The excess energy u_1 (18) is absorbed by the ground state represented by the first term on the right-hand side in equation (12). This implies the convergence rate of the chemical potential μ_R when approaching zero:

$$\begin{aligned}&\frac{2\epsilon_1^R}{V_R}\frac{1}{(e^{-\beta\mu_R}-1)} = \frac{2\epsilon_1^R}{R^3}(1-\beta\mu_R+\ldots-1)^{-1} = u_1\\ &\Rightarrow \mu_R \sim \frac{2\epsilon_1^R}{\beta R^3} \sim \frac{1}{R^4}\end{aligned} \tag{19}$$

since $\epsilon_1^R \sim \frac{1}{R}$. To put the chemical potential zero before performing the infinite volume limit, or to let μ_R converge to zero in the infinite volume limit procedure (19), that makes a decisive difference giving rise to the condensation.

To consider a two-dimensional ideal photon gas, we refer to the asymptotic expansion of the mean energy density (13). We drop the bulk term and focus on the two-dimensional surface term. Also, higher orders in R^{-1} are neglected. The independent thermodynamic variables are the mean energy surface density $\underline{u}^s$ (with the unit J/m^2), and the inverse temperature β. We follow the reasoning as in the bulk case. The chemical potential μ is a unique solution of

$$\begin{aligned}u_e^{\text{surface}}(\beta, \mu) &:= \frac{2}{4\pi\hbar^2c^2\beta^3}g_3(e^{\beta\mu})\\ &= \underline{u}^s \text{ if } \underline{u}^s \leqslant u_{\text{crit}}^{\text{surface}}(\beta)\end{aligned} \tag{20}$$

and

$$\mu = 0 \text{ if } \underline{u}^s > u_{\text{crit}}^{\text{surface}}(\beta). \tag{21}$$

If the value $\underline{u}^s$ lies in the critical regime (21), the mean energy surface density of the condensate emerges spontaneously and is given by

$$\begin{aligned}&u_1^s := \underline{u}^s - u_{\text{crit}}^{\text{surface}}(\beta).\\ &\text{If } \underline{u}^s \leqslant u_{\text{crit}}^{\text{surface}}(\beta), \text{ we set } u_1^s := 0.\end{aligned} \tag{22}$$

The excess energy (22) occupies the ground state.

The asymptotic expansion of the critical mean energy density $u_e^R(\beta, 0)$ up to second order can be read off from (13) and is given by

$$\begin{aligned}u_{\text{crit}}(\beta) &= u_{\text{crit}}^{\text{bulk}}(\beta) - \frac{A_R}{V_R}u_{\text{crit}}^{\text{surface}}(\beta)\\ &= \frac{6}{\pi^2\hbar^3c^3\beta^4}g_4(1) - \frac{A_R}{V_R}\frac{2}{4\pi\hbar^2c^2\beta^3}g_3(1).\end{aligned} \tag{23}$$

Accordingly the total critical energy of the finite photon gas up to second order is

$$U_{\text{crit}}(\beta) = V_R\frac{6}{\pi^2\hbar^3c^3\beta^4}g_4(1) - A_R\frac{2}{4\pi\hbar^2c^2\beta^3}g_3(1). \tag{24}$$

V. APPLICATION TO AN OPTICAL MICROCAVITY

Applying this formalism to the case of the two-dimensional optical microcavity, it is possible to calculate the critical power of radiation inside the cavity to induce condensation. Referring to the paper of Klaers *et al.* [8], two curved mirrors, with radius of curvature $R_{\mathrm{curv}} = 1$ m and central distance $D_0 = 1.46\,\mu$m, define the geometry of the optical microcavity. The data imply a volume $V_R = \pi D_0{}^2(R_{\mathrm{curv}} - D_0/3)/2 = 3.35 \times 10^{-12}\ \mathrm{m}^3$, and a surface area $A_R = 2\ \pi\ R_{\mathrm{curv}} D_0 = 9.17 \times 10^{-6}\ \mathrm{m}^2$. For room temperature of 300 K, the numerical value of the total critical energy (25) is

$$\begin{aligned} &U_{\mathrm{crit}}(300\ \mathrm{K}) \\ &= V_R \times 6.1282 \times 10^{-6}\ \mathrm{J/m^3} - \mathrm{A_R} \times 1.3601 \times 10^{-11}\ \mathrm{J/m^2} \\ &= 2.05 \times 10^{-17}\ \mathrm{J} - 12.47 \times 10^{-17}\ \mathrm{J} \\ &= -10.42 \times 10^{-17}\ \mathrm{J}. \end{aligned} \tag{25}$$

The surface term dominates the bulk term by a factor of 6 qualifying this microcavity as an approximately two-dimensional system. The minus sign of the surface-energy formally results from the Dirichlet boundary conditions [26,29]. (In the Dirichlet case, the eigenfunctions of the Laplacian are assumed to vanish at the boundary; in the boundary area, a surplus due to the extrapolated bulk value thus has to be subtracted.) More generally, the signs of the different terms in the asymptotical expansion regulate the contribution of the respective terms. Due to its dominant contribution and our interest in describing the cavity as two dimensional, we focus on the surface term contribution to the critical energy. As a physical quantity we take its positive value.

To compare the theoretical values (25) to the experimental results from [8], the critical energy of the photon gas has to be related with the critical power inside the cavity. The power inside the cavity consists of the contribution from the photon gas, and the contribution from the pumped dye molecules forming a thermodynamic reservoir. This last part is about 50 times larger than the contribution from the photon gas (see [8], same notation):

$$N_{\mathrm{exc}}/N_{\mathrm{ph}} = \tau_{\mathrm{exc}}/\tau_{\mathrm{ph}} = 1\ \mathrm{ns}/20\ \mathrm{ps} = 50, \tag{26}$$

where N_{ph}, τ_{ph} denote the average number of the photons in the resonator and the average time between emission and absorption, respectively, N_{exc} and τ_{exc} the number of molecular excitations and their electronic lifetime in the resonator, respectively. A characteristic length of the microcavity is given by the ratio of volume to surface, $l_0 = V_R/A_R \approx D_0$. As mentioned, we want to describe the photon gas in the microcavity as a purely two-dimensional system. So we decide to neglect the bulk contribution, instead of taking the whole expression from (25). Focussing on the surface term in (25), $U_{\mathrm{crit}}^{\mathrm{surface}}(300\,\mathrm{K}) = 12.47 \times\ 10^{-17}$ J, we get the critical power of the radiation in the two-dimensional photon gas

$$P_{\mathrm{crit}}(300\,\mathrm{K}) = (1 + 50)U_{\mathrm{crit}}^{\mathrm{surface}}(300\,\mathrm{K})/(l_0/c) = 1.31W. \tag{27}$$

The theoretical value lies within the tolerance of the experimental value $P_{c,\mathrm{exp}} = (1.55 \pm 0{,}60)$ W of Klaers *et al.* [8].

VI. LOCALIZATION OF THE CONDENSATE

Now we determine the explicit form of the condensation state function. The states of an ideal grand-canonical photon gas in a resonator are given by the Hilbert space vectors of the symmetric Fock space over the single-photon Hilbert space $\mathfrak{H}$,

$$\mathfrak{F}(\mathfrak{H}) = \oplus_{n=0}^{N}\mathfrak{H}^n, \tag{28}$$

with N denoting the total number of photons, and $\mathfrak{H}^n$ the symmetrized n-fold direct product of $\mathfrak{H}$. To derive the condensation state of the photon gas, we refer to a three-dimensional parallelepiped with edges L_1, L_2, L_3:

$$\frac{-L_i}{2} \leqslant x_i \leqslant \frac{L_i}{2}, i = 1, 2, 3; \tag{29}$$

the two-dimensional case, and the case of cavity geometries different from a parallelepiped follow accordingly. The ground-state function of the photon gas occupied by N_1 photons with energy ϵ_1^R, the lowest-energy eigenvalue of the cavity (29) with Dirichlet boundaries supposed, is given by

$$\prod_{i=1}^{3} \cos\left(\frac{\pi}{L_i}x_{1,i}\right) \ldots\ \cos\left(\frac{\pi}{L_i}x_{N_1,i}\right) \in \mathfrak{H}^{N_1}. \tag{30}$$

To evaluate (30) in the condensation regime, it is crucial to observe that the condensate does not contribute to the grand-canonical entropy density s_R. The entropy density s_R is given by the energy density u_R, the photon number density ρ_R multiplied by the chemical potential $\mu_R{}^*$, and the radiation pressure p_R

$$\begin{aligned} s_R(\beta, \mu_R{}^*) = \frac{2}{V_R}\sum_{k=1}^{\infty} k_B\Big\{&\big(\beta\epsilon_k^R - \beta\mu_R{}^*\big)\big(e^{\beta(\epsilon_k^R - \mu_R{}^*)} - 1\big)^{-1} \\ &+ \ln\big(e^{\beta(\epsilon_k^R - \mu_R{}^*)} - 1\big)^{-1}\Big\} \\ = k_B\beta\{&u_R(\beta, \mu_R{}^*) - \mu_R{}^*\rho_R(\beta, \mu_R{}^*) \\ &+ p_R(\beta, \mu_R{}^*)\big\}. \end{aligned} \tag{31}$$

In this thermodynamic relation for the entropy density the nonnormalized chemical potential $\mu_R{}^* \leqslant \epsilon_1$ has to be used, and the nonnormalized energy spectrum as well. The nonnormalized $\mu_R{}^*$ and the normalized μ_R (5b) are connected as follows:

$$\mu_R = \mu_R{}^* - \epsilon_1^R.\ \text{We observe}\ \epsilon_k^R - \mu_R{}^* = \lambda_k^R - \mu_R.$$

Therefore (31) can be written as

$$\begin{aligned} s_R(\beta, \mu_R) = k_B\beta\big\{&u_R(\beta, \mu_R) - \big(\mu_R + \epsilon_1^R\big)\rho_R(\beta, \mu_R) \\ &+\ p_R(\beta, \mu_R)\big\}. \end{aligned} \tag{31$'$}$$

The term $\epsilon_1^R\rho_R(\beta,\ \mu_R)$ subtracts the ground-state contribution in $u_R(\beta,\ \mu_R)$ which is relevant for the thermodynamic limit with β and $\underline{u}$ as independent variables. That part of $\epsilon_1^R\rho_R(\beta,\ \mu_R)$, which comprises the excited states tends to zero, for $R \to\ \infty$. Also $\mu_R\rho_R(\beta,\ \mu_R)$ tends to zero, since $\mu_R\ \sim\ R^{-4}$ [see (19)], and $\rho_R(\beta, \mu_R) \sim R$.

The last convergence rate stands for the infrared catastrophe [5] which is well known in quantum electrodynamics. The limit of the radiation pressure p_R is given by (26) in [5]. Thus we get

$$\lim_{R\to\infty} s_R(\beta, \mu_R(\beta, \underline{u}))\ := s(\beta, \mu(\beta, \underline{u}))\ = s_e(\beta, \mu(\beta, \underline{u})), \tag{32}$$

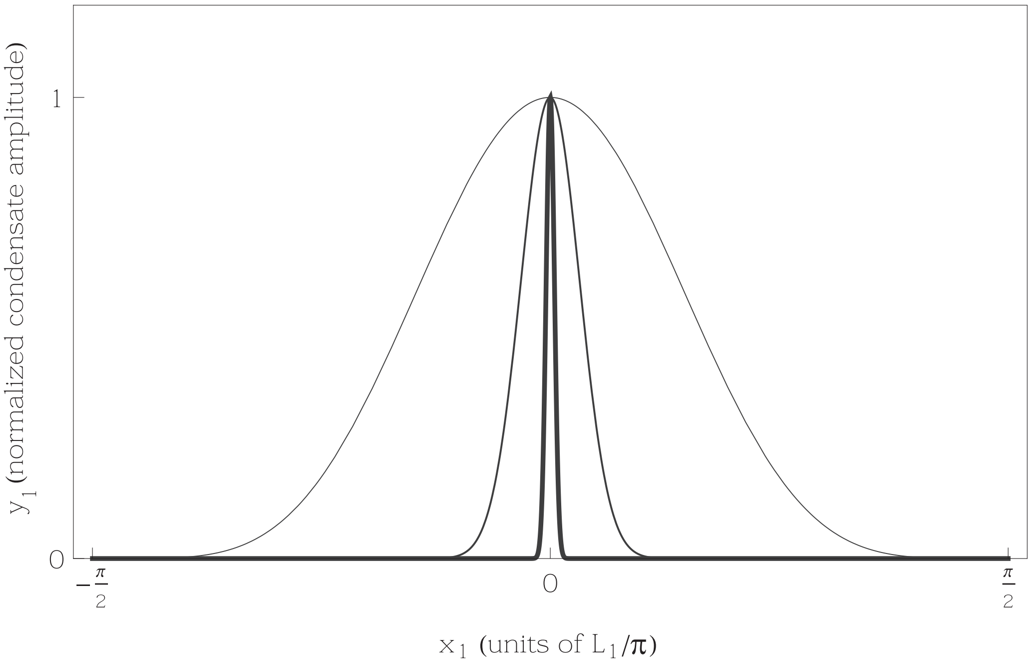

FIG. 1. x_1 component of the spatial condensate distribution $f_1^{N_1}$, for N_1= 5, 100, 5000 (starting from left).

where s_e comprises the thermodynamic limit of the sum of the excited modes $k \geqslant 2$ in (31). The result (32) includes the condensation regime: For the condensation regime $u \geqslant u_{\text{crit}}^{\text{bulk}}(\beta)$, the chemical potential is zero, and we get

$$s(\beta, \mu(\beta, \underline{u})) = s_e(\beta, 0) = \frac{4}{3} k_B \beta u_{\text{crit}}^{\text{bulk}}(\beta), \qquad \text{for}\ \underline{u} \geqslant u_{\text{crit}}^{\text{bulk}}(\beta). \tag{33}$$

Equation (31′) corrects equation (10) in [5]. Consequently equation (27) in [5] has to be replaced by equation (32) above, and equation (40) in [5] by equation (32) above [9,28]. Beyond this correction, details of the above calculations can be taken from [5].

If we increase $\underline{u}$ beyond $u_{\text{crit}}^{\text{bulk}}(\beta)$, the entropy density (33) remains constant while the energy increase builds up the condensate. The condensate does not contribute to the entropy, the entropy of the condensate is zero. This means that, in the condensation regime, the ground state is not a mixture of random phases (30) but a pure state, with identical phases for the cosine functions. This observation implies the following evaluation of (30):

$$\prod_{i=1}^{3} \left(\cos\left(\frac{\pi}{L_i} x_i \right) \right)^{N_1} \in \mathfrak{H}^{N_1}. \tag{34}$$

Expression (34), denoted by $f_1^{N_1}(x_1, x_2, x_3)$, gives the spatial condensate distribution. In the idealization $N_1 \to \infty$, the spatial distribution f_1 of the condensate is

$$f_1(x_1, x_2, x_3) = \begin{cases} 1 & \text{for}\ x_i = 0,\ i = 1,\ 2,\ 3, \\ 0 & \text{for}\ 0 < |x_i| \leqslant \frac{L_i}{2},\ i = 1,\ 2,\ 3. \end{cases} \tag{35}$$

At any point (x_1, x_2, x_3) outside the center, $f_1^{N_1}(x_1, x_2, x_3)$ forms a bounded number sequence strictly monotonic decreasing with respect to N_1 which implies the result (35). (Compare, e.g., to [30].) Figure 1 (see above) visualizes the convergence rate of (34) for the x_1 component for N_1= 5, 100, 5000; the last case shows the sharpest distribution.

VII. DISCUSSION

Bose-Einstein condensation in an ideal photon gas can be realized in three and in two dimensions. The analytical framework developed above allows a formally consistent and quantitatively accurate description of the experimental results of Klaers, Schmitt, Vewinger, and Weitz for a two-dimensional microcavity [8]. In particular, it can be proved that the photon condensate is localized at the center of the cavity, in line with the observation. The proof explains the robustness of the central localization of the condensate against a spatially displacement of the pump beam, as noticed in [8]. The central localization of the photon condensate makes clear that, in the idealization of an infinite number of (infrared) photons with infinitesimally small energy, there is no contribution of the condensate to the radiation pressure, in accordance with the corresponding proof in [5].

Photon condensation transforms photons from higher frequencies to lower frequencies. At the same time the condensate builds up a state of high order. This offers technical applications for photovoltaic energy conversion and energy storage, for new electromagnetic radiation sources, and for photonics.

On the most fundamental level, as described in this paper, the condensate represents stationary energy. Hence, according to Einstein's equivalence of energy and mass, it has to be associated with a nonzero rest mass; the fundamental criterion for matter.

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