General Theory of Bose-Einstein condensation applied to an ideal gas of photons in a 2- and 3-dimensional resonator

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1. Introduction

Bose-Einstein condensation (BEC) in an ultra-cold gas of atoms has become a well known phenomenon since its first experimental demonstration in 1995 [1].

What is the scheme of Einstein's original prediction of BEC in 1924 [2]**?**

He refers to a monoatomic ideal quantum gas. There he identifies a fundamental quantum phase transition. Structurally, he compares the phase transition with the condensation of vapor. He fixes the temperature in the gas, and increases the number density of the atoms beyond the saturation value.

That means: He uses a **grand-canonical ensemble, defined by two independent variables.**

[1] M. H. Anderson et al., Observation of Bose-Einstein condensation in a dilute atomic vapor. Science 269, 198 (1995).
[2] A. Einstein, Quantentheorie des einatomigen idealen Gases. Zweite Abhandlung, Sitzungsber. Preuss. Akad. Wiss. I, 3 (1925).

Albert Einstein, 1879-1955
Satyendra Nath Bose, 1894-1974

Einstein’s approach to BEC in an ideal quantum gas presumed particles with non-zero rest mass. He extended Bose’s new statistics for Planck’s ideal photon gas to an ideal quantum gas of massive “bosons“. In this case, it is possible to control the particle density experimentally. Einstein’s hypothesis of a condensation in an ideal boson gas did not include an ideal photon gas.

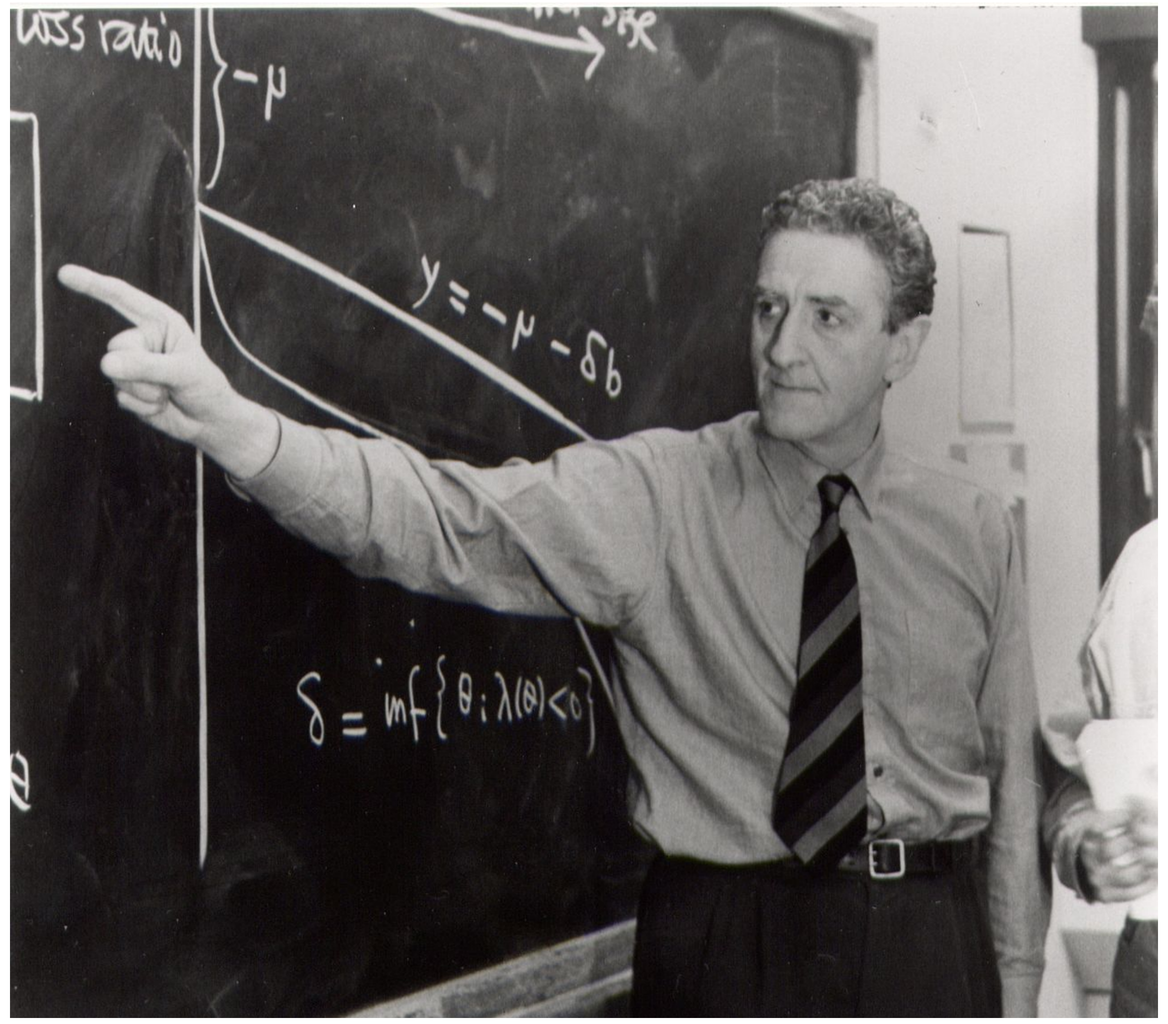
The zero rest mass of the free photons presents **a severe conceptual problem for a photon condensation**. Condensation means that, beyond the saturation value, an excess of bosons in the gas transit to a “state without kinetic energy“ [2]. **A photonic occupation of the state without kinetic energy seems to have no substance at all**.

A close inspection of the thermodynamic framework gives a solution for the conceptual problem of a BEC of photons. **The clue is an appropriate thermodynamic limit which proves the correct selection of the thermodynamic variables which are involved.**

This talk refers to my paper [3], a comprehensive and mathematically self-contained derivation of photon condensation. The mathematical technique is inspired by [4] where the focus aimed at non-relativistic BEC-systems.

The approach to a photon condensation presented here, is applied to the experimental setting for a Bose-Einstein condensation of photons in an optical microcavity realized by the group of Martin Weitz, University Bonn [5]. The theoretical predictions match the experimental results.

[3] E. E. Müller, General theory of Bose-Einstein condensation applied to an ideal quantum gas of photons in an optical microcavity. Phys. Rev. A **100**, 053837 (2019).
[4] M. Van den Berg, J. Lewis, and J. Pulè, A general theory of Bose-Einstein condensation. Helv. Phys. Acta 59, 1271 (1986).
[5] J. Klaers, J. Schmitt, F. Vewinger, and M. Weitz, Bose-Einstein condensation of photons in an optical microcavity, Nature **468**, 545 (2010).



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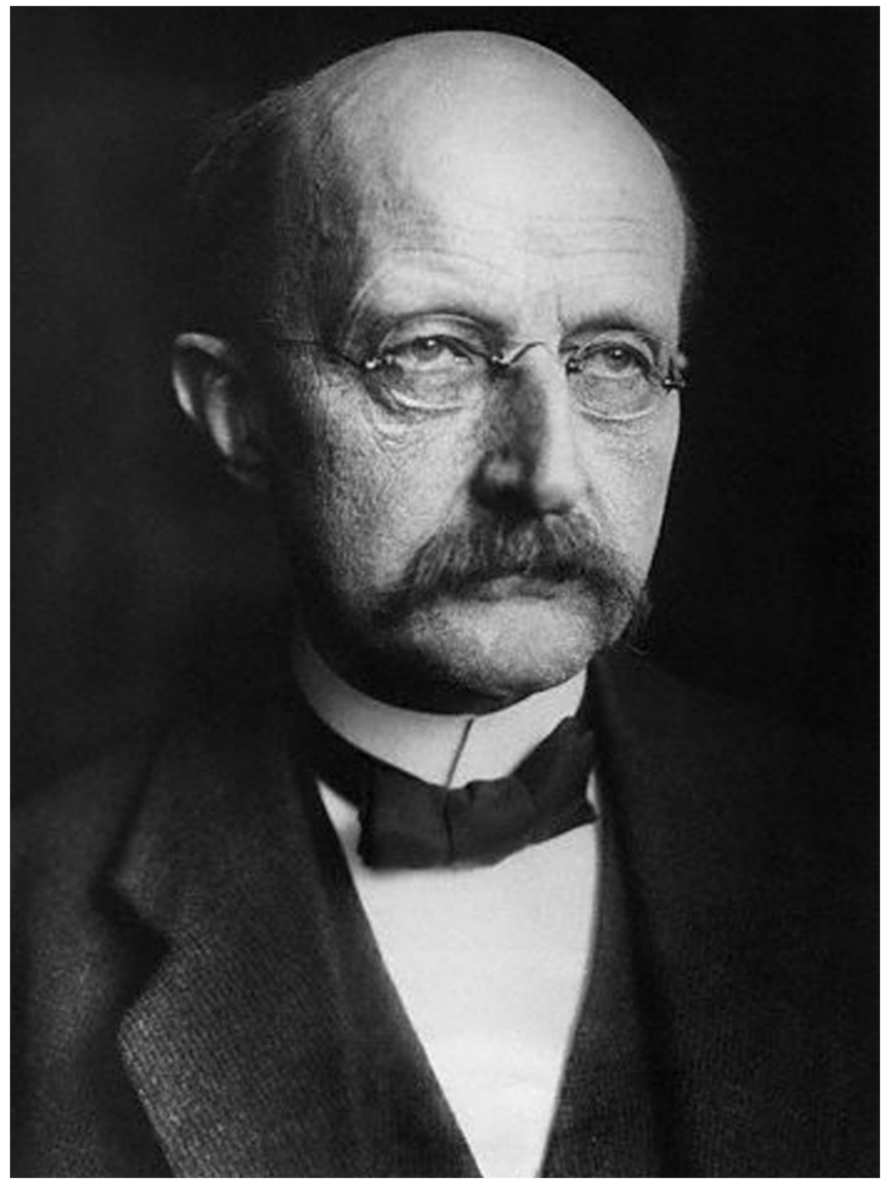
2. Thermalization in an ideal photon gas

We look for a thermalization mechanism in an ideal photon gas. Already, at the end of the 19th century, this question had been a challenge for Max Planck's reasoning on heat radiation. For an evacuated cavity with perfectly reflecting walls filled with some arbitrary composition of light rays, he symbolically postulated the presence of **a minute black dust, a coal dust,** to transform any radiation into **black body radiation** [6].



Max Planck, 1858-1947

[6] M. Planck, Vorlesungen über die Theorie der Wärmestrahlung, J. A. Barth, Leipzig, 1921; see paragraph 52.

A coal dust absorbs and re-emits radiation. It is a means to achieve thermal equilibrium.
The photon gas assumes the temperature of the black dust.

In a real cavity, the walls are not perfectly reflecting. They take over the role of a coal dust.
Eventually, the photon gas in the cavity will reach thermal equilibrium with the walls.

A **thermalization mechanism in an ideal quantum gas of photons** is essential
- not only to establish thermal equilibrium, but, in addition,
- to realize **a possible phase transition**.

A quantum view to thermalization

We use the very quantum nature of an ideal photon gas to get around Planck's knack of a coal dust. Given a finite size cavity, with perfectly reflecting walls. Let R be a typical length of the cavity. The localization within the cavity implies a spatial uncertainty for the photons of

$$\Delta x \ < \ R$$

The Heisenberg uncertainty relation implies an uncertainty Δp for the photon momentum $p = h\nu/c$.

$$\Delta p \ \geq \ (h/4\pi) \ / \ \Delta x \ > \ (h/4\pi) \ / \ R$$

$$\Delta \nu \ \geq \ (c/4\pi) \ / \ \Delta x \ > \ (c/4\pi) \ / \ R$$

The uncertainty of the frequency ν, $\Delta\nu$, induces a smearing out of the resonances. They overlap. Thus, we get nonzero transition probabilities between the resonances, implying a thermalization mechanism.

So far, the use of the uncertainty relation depends on the finiteness of the cavity.
Since we shall use a thermodynamic limit, we need a procedure for **infinite quantum systems** to establish a thermal equilibrium.

We know, that the uncertainty relation results from the non-commutativity of the observable algebra.
In the Tomita-Takesaki theory there is an operator on a von Neumann algebra, the so-called "modular operator" Δ, which measures the non-commutativity in the observable algebra.
In the ideal quantum gas, Δ induces a time evolution:

$$x \rightarrow \Delta^{it}\, x\, \Delta^{-it}, \quad x \in \text{von Neumann algebra of observables.}$$

This holds for infinite large systems, too.

In the thermal equilibrium, the time evolution with Hamiltonian *H* is given by the above modular time evolution, up to a scaling factor [7, 8]. This scaling factor is the inverse temperature.

$$H = -\beta^{-1} \log \Delta$$

The modular operator is intimately related to quantum correlations. So, t**he thermal fluctuations can be reduced to quantum fluctuations**.

Thus, we have a thermalization mechanism for an ideal quantum gas of photons that does not rely on a black dust of coal. The system may be finite, or infinite.

Loosely spoken: *We decarbonize thermalization*.

[7] M. Takesaki, Theory of Operator Algebras II, Encyclopaedia of Mathematical Sciences, Vol 125, Springer, N. Y., 2003, p. 92.
[8] R. Haag, Local Quantum Physics, 2nd ed., Springer, N. Y., 1996, p. 216 ff.

3. Particles of zero rest mass present a problem for Bose-Einstein condensation

We consider a photon gas in a finite cavity of volume V_R and surface area A_R with reflecting walls. The photon Hamiltonian for the cavity with Dirichlet boundary conditions is

(1) $$\hbar c\sqrt{-\Delta_R}$$

with strictly positive eigenvalues

(2) $$0 < \epsilon_1^R \leq \epsilon_2^R \leq \epsilon_3^R \leq \dots$$

Chemical potential μ_R* of the photon gas:

(3) $$0 < \mu_R^* \leq \epsilon_1^R$$

The eigenvalues depend on the inverse of R:

(4) $$\epsilon_1^R \sim R^{-1}, \dots$$

The chemical potential of a photon gas in a resonator of finite size is non-zero. The photons within the resonator propagate with the speed of light, and their mass is given by the fundamental relation

(5) $$mc^2 = h\nu.$$

The eigenfrequencies in the cavity are reciprocal proportional to R (3). In particular,

(6) $$\nu_1 \sim R^{-1}$$

For the sake of conceptual clarity on Bose-Einstein condensation of photons, we employ the infinite volume limit of an ideal photon gas in the cavity. It is well known that a thermodynamic limit presents a sharp manifestation of a phase transition. In the case of BEC, we have to look at the occupation of the ground state. Relation (6) shows that the energy of the photons in the ground state tend to zero. So, in the thermodynamic limit, the ground state energy seems to be zero.

Except in the case of an infinite number of photons with infinitesimally small energy!

4. Choosing the independent variables

The **“canonical ensemble“** maximizes the entropy subject to the mean energy density being fixed. The respective Lagrange parameter β stands for the inverse temperature. A canonical ideal quantum gas excludes a phase transition.

The **“grand-canonical ensemble“** maximizes the entropy subject to the mean energy density and the mean particle density being fixed. The additional constraint is dealt with a second Lagrange parameter μ, associated with the chemical potential. A grand-canonical ideal quantum gas admits a phase transition.

We use a grand-canonical ensemble to describe the ideal quantum photon gas under consideration, with the following thermodynamic variables:
- mean energy density u
- mean photon number density ρ
- inverse temperature β
- chemical potential μ of a photon in the photon gas

We allow a possibly infinite number of infrared photons with infinitesimally small energy. In this case, it is impossible to control experimentally the number density as a thermodynamic variable. This problem translates to the chemical potential. Therefore, we choose the mean energy density and the temperature as independent variables of the photon gas. They are experimentally accessible and controllable.

This decision has a consequence for the thermodynamic limit. It is very common to fix the particle number density in the infinite volume limit. Not so in this approach. Here, we shall fix the mean energy density when performing the limit.

5. Photon condensation in three and two dimensions

The lower bound of the spectrum of the photon Hamiltonian is not invariant under the procedure of the infinite volume limit. This is mathematically unsatisfactory. Therefore we rewrite the energy spectrum to get its lower bound fixed at zero. Accordingly, we introduce a normalized chemical potential μ.

(7) $$\lambda_k^R := \epsilon_k^R - \epsilon_1^R$$

(8) $$\mu_R \leq 0, \quad \mu_R = \mu_R{}^* - \epsilon_1^R$$

We observe

(9) $$\epsilon_k^R - \mu_R{}^* = \lambda_k^R - \mu_R$$

Our task is to find the function of the **mean energy density of the grand-canonical photon gas** in the finite size resonator. With an **asymptotic expansion** for $R \to \infty$, we get the bulk term and the surface term.

Integrated spectral density of the photon Hamiltonian that counts the eigenvalues $\lambda^R_{k,\alpha} := \lambda^R_k$ up to the variable λ [9]; k denotes the mode number, α the helicity:

(10)
$$F_R(\lambda) := \frac{1}{V_R}\,\#\left\{(k,\ \alpha) \in \mathbb{N} \times \{+1,\ -1\} : \lambda^R_{k,\alpha} \leqslant \lambda\right\}$$
$$= \frac{1}{3\pi^2}\left(\frac{\lambda}{\hbar c}\right)^3 - \frac{A_R}{8\pi V_R}\left(\frac{\lambda}{\hbar c}\right)^2 + \mathcal{O}\left(\frac{\lambda}{R^2}\right)$$

Spectral density:

(11)
$$dF_R(\lambda) = \frac{1}{\pi^2}(\hbar c)^{-3}\lambda^2\, d\lambda - \frac{A_R}{4\pi V_R}(\hbar c)^{-2}\lambda\, d\lambda + \mathcal{O}\left(R^{-2}\right) d\lambda$$

[9] M. Van den Berg, On the asymptotics of the heat equation and bounds on traces associated with the Dirichlet Laplacian, J. Funct. Anal. **71**, 279 (1987)

The Hamiltonian $\boldsymbol{H_R}$ of the photon gas is the well known second quantization of the single photon Hamiltonian (1). The helicity contributes a factor 2. The zero point energy in $\boldsymbol{H_R}$ will vanish in the thermodynamic limit of the mean energy density, so we omit it.

The mean energy density of the photon gas is the grand-canonical expectation value of $\boldsymbol{H_R / V}$.

(12) $$u_R(\beta,\ \mu_R) = \frac{2}{V_R} \sum_{k=1}^{\infty} \left(\lambda_k^R + \epsilon_1^R\right)\left(e^{\beta\left(\lambda_k^R - \mu_R\right)} - 1\right)^{-1}$$

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

Asymptotic expansion of the sum of the excited states, up to second order:

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

Critical energy density in 3 dimensions, with μ = 0 (black body radiation in 3 dimensions):

(15) $$u_{crit}^{bulk}(\beta) = \frac{6}{\pi^2 \hbar^3 c^3 \beta^4} g_4(1) ; \qquad g_4(1) = \pi^4 / 90$$

Critical energy density in 2 dimensions, with μ = 0 (black body radiation in 2 dimensions):

(16) $$u_{crit}^{surface}(\beta) = \frac{2}{4\pi\hbar^2 c^2 \beta^3} g_3(1) ; \qquad g_3(1) = 1.20206 \ldots$$

Photon condensation in 3 dimensions

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

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

If $\underline{u} \leq u_{crit}^{bulk}(\beta)$, $\mu_R \to \mu$ for $R \to \infty$ where μ is the unique solution of

(18) $$\frac{6}{\pi^2 \hbar^3 c^3 \beta^4} g_4\left(e^{\beta\mu}\right) = \underline{u}.$$

If $\underline{u} > u_{crit}^{bulk}(\beta)$, $\mu = 0$.

The **excess energy** $u_1 := \underline{u} - u_{crit}^{bulk}(\beta)$ occupies the ground state.
This is the photon condensate.

Photon condensation in 2 dimensions

Given a temperature β, and a value $\underline{u}^s$ of the mean energy surface density.

(19) If $\underline{u}^s \leq u_{crit}^{surface}(\beta)$, μ is the unique solution of

(20) $$\frac{2}{4\pi\hbar^2 c^2 \beta^3} g_3(e^{\beta\mu}) = \underline{u}^s.$$

If $\underline{u}^s > u_{crit}^{surface}(\beta)$, $\mu = 0$.

The **excess energy** $u_1^s := \underline{u}^s - u_{crit}^{surface}(\beta)$ **is the photon condensate on the surface**.

6. Application to a two-dimensional optical microcavity

Experimental setting in [5]: Two curved mirrors form the optical cavity. The cavity is filled with a dye solution. The dye is pumped with a laser beam.

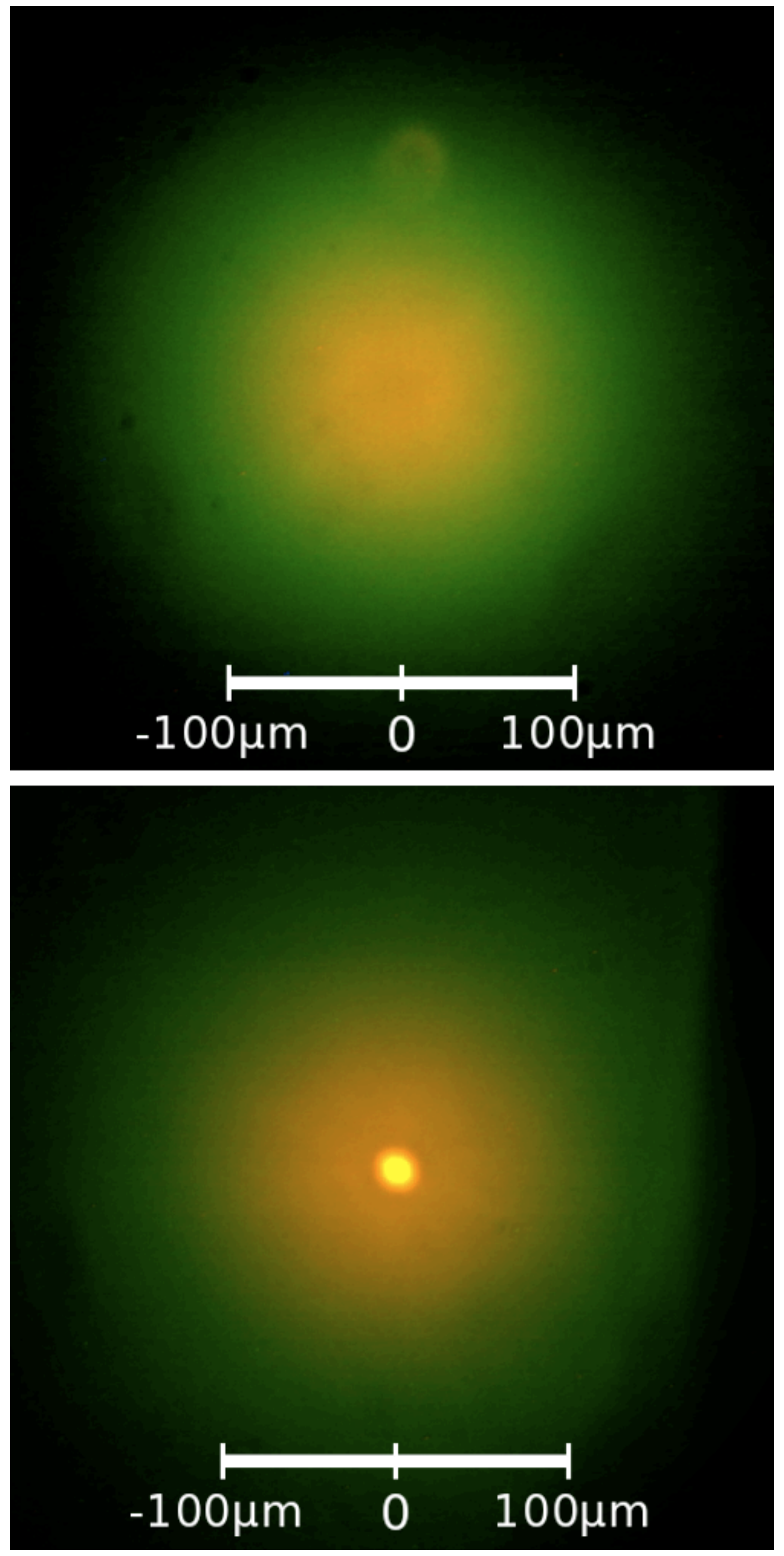
Radius of curvature $R_{curve} = 1$ m, central distance $D_0 = 1.46$ µm
Reflectivity of the dielectric mirrors: > 99.997 %.

Critical surface energy [3]:

(21) $$U_{crit\,surf}(\beta) = A_R\, u_{crit}^{surface}(\beta)$$

$$U_{crit\,surf}(300\text{ K}) = 9.17 \times 10^{-6}\text{ m}^2 \times 1.3601 \times 10^{-11}\text{ J/m}^2 = 12.47 \times 10^{-17}\text{ J}$$

(22) $P_{crit}(300\text{ K}) =$ **1.31 W**; $P_{c,\,exp} =$ **(1.55 ± 0.60) W** [5].



Martin Weitz, University of Bonn.
BEC of photons in an optical microcavity.

7. Spatial localization of the photon condensate

We shall determine the explicit form of the condensate [3]. We demonstrate it for the case of a three-dimensional parallelepiped with edges L_1, L_2, L_3:

(23) $$\frac{-L_i}{2} \leq x_i \leq \frac{L_i}{2}, \quad i = 1, 2, 3$$

Ground state of the photon gas occupied by N_1 photons with energy ϵ_1^R:

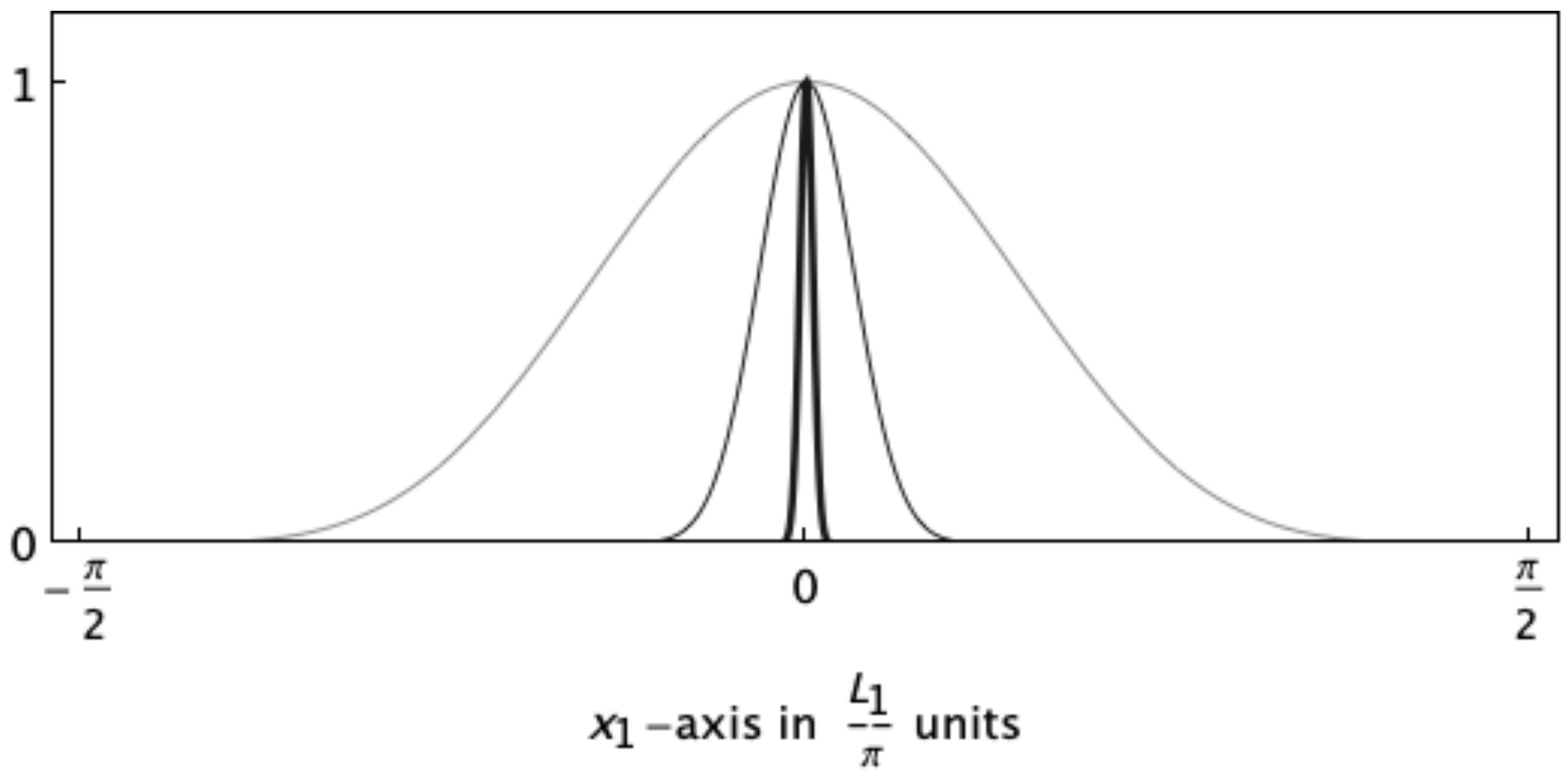
(24) $$\prod_{i=1}^{3} cos\left(\frac{\pi}{L_i} x_{1,i}\right) \ldots cos\left(\frac{\pi}{L_i} x_{N_1,i}\right)$$

The condensate does not contribute to the grand-canonical entropy density. **The entropy of the condensate is zero** [3]. This means that, in the condensation regime, the ground state is not a mixture of random phases (24) but a pure state, with identical phases for the cosine functions. This implies the following evaluation of (24):

$$\prod_{i=1}^{3}\left(\cos\left(\frac{\pi}{L_i}x_i\right)\right)^{N_1} =: f_1^{N_1}(x_1, x_2, x_3) \quad (25)$$

In the idealization $N_1 \rightarrow \infty$ the spatial distribution f_1 of the condensate is

$$f_1(x_1, x_2, x_3) = \begin{cases} 1 & \text{for } x_i = 0,\ i = 1, 2, 3 \\ 0 & \text{for } 0 < |x_i| \leq \frac{L_i}{2},\ i = 1, 2, 3 \end{cases} \quad (26)$$



The figure shows the convergence rate of (25) for the x_1 component for N_1 = 5, 100, 5000.

8. Visions for technical applications of photon condensation

1. Photon condensation **transforms** photons from higher **frequencies** to lower frequencies.

2. At the same time the condensate builds up a state of high order, of **strong coherence**.

3. The **radiation pressure of the condensate is zero**, in the thermodynamic limit.

4. Photon condensation in **resonators with fractal dimension** larger than 1 might be an intriguing option.

Ad 1 Photovoltaic energy conversion
New electromagnetic radiation sources

Ad 2 Coherence in biological systems: Microtubuli as resonators, photosynthesis
Photonic BEC chips for quantum computing and all optical networks, at room temperature
BEC with a low number of photons

Ad 3 **Energy storage**

9. Photon condensation makes non-zero rest mass emerge

The photon condensate represents stationary energy.
The Einstein equivalence of energy and mass implies, in this case, a non-zero rest mass.
Non-zero rest mass is the fundamental criterion for matter.

Hence we have a phase transition from light to mass.
This is relevant for the understanding of cosmological evolution.
And possibly for elementary particle physics.

Photon condensation matters.

Convergence rate of the chemical potential in the critical regime

μ_R is determined by (17). In the critcal regime, the excess energy u_1 is absorbed by the ground state.

$$\frac{2\,\epsilon_1^R}{V_R}\left(e^{-\beta\mu_R}-1\right)^{-1} \sim \frac{2\,\epsilon_1^R}{R^3}\left(1-\beta\mu_R+\ldots-1\right)^{-1} = u_1$$

(i) $$\Rightarrow \quad \mu_R \sim \frac{2\,\beta\epsilon_1^R}{R^3} \sim \frac{1}{R^4}$$

This displays the following inequality which is essential of our approach:

(ii) $$\lim_{R\to\infty} u_R\,(\beta,\ \mu_R(\beta,\ \underline{u}\,)) \neq \lim_{R\to\infty} u_R\,(\beta,\ 0\,) = u\,(\beta),\quad \text{for } \underline{\boldsymbol{u}} > \boldsymbol{u}\,(\boldsymbol{\beta})$$