

# New Kind of Condensation of Bose Particles Through Stimulated Processes

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#### **Abstract**

We show that stimulated scattering of an isolated system of *N* Bose particles with initially broad energy distribution can yield condensation of particles into excited collective state in which most of the bosons occupy one or several modes. During condensation, the total particle number and energy are conserved, while the entropy of the system grows. Onset of condensation occurs at a critical particle occupation number when spectrum narrowing due to stimulated processes overcomes spectrum broadening due to diffusion. This differs from Bose–Einstein condensation in which particles undergo condensation into the equilibrium state due to thermalization processes.

**Keywords** Condensation of Bose particles  $\cdot$  Stimulated scattering  $\cdot$  Bose–Einstein condensation

#### 1 Introduction

We are very pleased to dedicate this paper to the low-temperature leaders, professors David Lee and John Reppy. They are low-temperature adventurers in the spirit of C.T. Lane, the pioneer of the Yale superfluid He II group. John's Bose–Einstein condensation (BEC) experiments in the porous Vycor glass [1–3] have been an inspiration to us in considering fluctuations of BEC particle number in the canonical ensemble. When we had the good fortune to convince David to come to work in Texas A&M University, we were all very excited. Indeed, it is an inspiration to see

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him (at 90 years young) still coming into the laboratory every day and publishing PRLs. As he says: "We've got a great story to tell. I just got to have another PRL!" David's genius for asking deep questions has enriched our lives in many ways. For example, his charming curiosity has led us to a fruitful study of black hole entropy from a quantum optical perspective [4].

Bose–Einstein condensation implies macroscopic accumulation of particles on the ground-state level (or in states other than the ground state, for example, BEC with quantized vortices) of a Bose system at low temperature and high density. It involves the formation of a collective quantum state composed of identical particles. Superfluids and superconductors were, for a long time, the only physical systems where the effect of BEC had been observed. In the past decades, BEC has been theoretically predicted and detected in other systems, including cold atomic gases [5–7], collective modes and bosonic quasiparticles. Examples of the latter are longitudinal electric modes [8], phonons [9], excitons [10], polaritons [11], exciton–polaritons [12, 13], photons [14], rotons [15], and magnons [16–19]. In these systems, quasiparticles are externally pumped, but they are sufficiently long-lived, so that their number N is quasi-conserved. As a result, the chemical potential  $\mu = dE/dN$  is nonzero during the lifetime of the condensate.

BEC of photons has been observed in an optical microcavity [14]. The cavity mirrors provide both a confining potential and a nonvanishing effective photon mass, making the system formally equivalent to a two-dimensional gas of trapped, massive bosons. Photon number-conserving thermalization was achieved by filling the cavity with a dye. The photons thermalize to the temperature of the dye solution by multiple scattering (absorption and re-emission) with the dye molecules. Upon increasing the photon density, spontaneous onset of a macroscopic quantum phase with massively populated ground-state mode on top of a broad thermal wing has been detected [14], which is a signature of the BEC transition.

An interesting phenomenon in open systems far from thermodynamic equilibrium is the emergence of collective behaviors and self-organization, which is the mechanism behind superfluorescence [20], synchronization in collective nonlinear dynamics [21–23]. In biological systems, many theoretical works have suggested that the collective behavior may have profound effects on the chemical and enzyme kinetics [24], and the cognitive function of brain [25]. Among these works, a widely used model is the Fröhlich condensate [8, 26, 27]. In 1968, Fröhlich showed that the energy of a driven set of oscillators would condense at the lowest vibrational mode once the external energy supply exceeds a threshold [8, 26]. The Fröhlich condensate is a nonequilibrium phenomenon.

In this paper, we study a possibility of condensation of bosons into a collective state with a macroscopic occupation of several modes with different energies in a system with a nonlinear dynamics governed by spontaneous and stimulated emission and absorption. Namely, we show that such a system, apart from thermal-like states with a broad energy distribution, can have steady states in which one or several modes are macroscopically occupied. In such condensed steady states, there is exchange of particles between the macroscopically occupied modes by means of stimulated processes. The Bose system can condense into one of these collective



steady states, provided that for the initial particle distribution the stimulated scattering dominates.

One should note that collective states into which particles condense are different from fragmented BECs. Fragmented BEC occurs into a state, which is degenerate. Then, two or more states are competing simultaneously for Bose–Einstein condensation [28]. In our case, condensation occurs into several modes with different energies.

#### 2 Model Evolution Equations for the Particle Distribution Function

We consider N noninteracting Bose particles trapped in a potential well with equidistant energy levels  $E_n = nE_0$ . The system is isolated. Distribution of particles over the levels is described by the distribution function  $f_n$ , where  $f_n$  is the average number of particles with energy  $E_n$ . We turn on a  $\delta$ -function (in time) interaction with an external system, which makes particles jump into the adjacent lower energy levels with some probability. We assume that during such event the distribution function changes into:

$$f_n \to f_n + \kappa [f_{n+1}(f_n+1) - f_n(f_{n-1}+1)],$$
 (1)

where  $\kappa$  is a dimensionless rate coefficient. Figure 1 explains the probability flow of particles into the level n. Occupation number of level n increases because particles jump from the level n+1 into the level n with a probability proportional to  $f_{n+1}(f_n+1)$  and decreases because particles go from the level n into the level n-1 with a probability  $\propto f_n(f_{n-1}+1)$ . Bosonic stimulation is included in the factors  $f_n+1$  and  $f_{n-1}+1$ , respectively. Those factors make the system's evolution nonlinear.

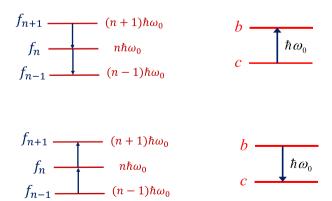


Fig. 1 (Color figure online) Probability flow diagram for photon frequency during excitation (deexcitation) of the two-level system



Then, we turn on another  $\delta$ -function interaction, which promotes particles into the adjacent higher energy levels and results in the change of the distribution function into

$$f_n \to f_n + \kappa [f_{n-1}(f_n + 1) - f_n(f_{n+1} + 1)].$$
 (2)

During consecutive scattering events (1) and (2), the total number of particles  $N = \sum_n f_n$  and their net energy  $E = E_0 \sum_n n f_n$  are conserved.

Our model, for example, can describe photons in a microcavity, which are analogous to massive particles in a harmonic trap. Radiation with a broadband spectrum is trapped inside an ideal cavity. Inside the cavity there is a two-level system with energy spacing  $E_0$ . The system can inelastically scatter light changing photon frequency by  $\pm E_0/\hbar$ . This is, e.g., the case in Raman scattering. Raman scattering is a process in which an atom or a molecule absorbs a photon by undergoing a transition to a virtual state and then decays to a real state by emitting a photon of lower (Stokes Raman scattering) or higher (anti-Stokes Raman scattering) energy [29]. During such scattering, an atom or a molecule gives to or takes away energy from the field by going to a higher or a lower energy state, while the number of photons is conserved. We assume that there is no energy dissipation, that is if the two-level system acquires energy  $E_0$  from the radiation field, then it will give this energy back in the next scattering event.

The question we are interested in here is how the distribution function evolves if the consecutive scattering events (1) and (2) occur many times. One would expect that the distribution function will evolve into  $f_n$  =const, which is a stationary state of Eqs. (1) and (2). However, we will show that nonlinearity due to bosonic stimulation can yield condensation of most of the particles into a single level if the initial particle occupation numbers are large. This is similar to the line narrowing in lasers. Photon condensation occurs into localized stationary states, which we will discuss next.

### 3 Localized Stationary States

Here we obtain stationary states of Eqs. (1) and (2) for which distribution function  $f_n$  is narrow (localized). According to Eq. (1), after scattering event in which the two-level system goes to the excited state the distribution function becomes

$$\tilde{f}_n = f_n + \kappa \left[ f_{n+1}(f_n + 1) - f_n(f_{n-1} + 1) \right]. \tag{3}$$

In the next scattering event, the two-level system goes back to the ground state and the photon distribution function, according to Eq. (2), changes to

$$\bar{f}_n = \tilde{f}_n + \kappa \left[ \tilde{f}_{n-1}(\tilde{f}_n + 1) - \tilde{f}_n(\tilde{f}_{n+1} + 1) \right]. \tag{4}$$

For stationary state



$$\bar{f}_n = f_n. \tag{5}$$

This equation has the following stationary solution

$$f_n = \frac{1}{\kappa} - 1, \quad f_{n+1} = 1 - \kappa,$$
 (6)

and other occupation numbers are equal to zero. Total photon number in this state is  $1/\kappa - \kappa$ .

After scattering event in which the two-level system goes to the excited state, the distribution (6) changes to

$$f_n = \frac{1}{\kappa} - 1, \quad f_{n-1} = 1 - \kappa.$$
 (7)

In the next scattering event, the distribution changes back to (6). One should note that for  $\kappa \ll 1$  only one mode in state (6) is macroscopically occupied:  $f_n \gg 1$ .

Another stationary solution is

$$f_{n-1} = \frac{1}{2} \left( \frac{1}{\kappa} - 1 \right), \quad f_n = \frac{1}{\kappa} - 1, \quad f_{n+1} = \frac{1}{2} (1 - \kappa),$$
 (8)

and the other occupation numbers are equal to zero. The total number of photons in this state is  $3/2\kappa - \kappa/2 - 1$ .

After scattering event in which the two-level system goes to the excited state, distribution (8) changes to

$$\tilde{f}_{n-2} = \frac{1}{2}(1-\kappa), \quad \tilde{f}_{n-1} = \frac{1}{\kappa} - 1, \quad \tilde{f}_n = \frac{1}{2}(\frac{1}{\kappa} - 1).$$
 (9)

In the next scattering event, the distribution changes back to (8). For  $\kappa \ll 1$ , the two modes in state (8) are macroscopically occupied:  $f_n, f_{n-1} \gg 1$ .

In next sections, we demonstrate numerically that evolution Eqs. (1) and (2) can yield condensation of photons with initially broad spectrum into localized stationary states and show the importance of the stimulated scattering for condensation.

## 4 Photon Condensation Via Stimulated Raman Scattering

Nonlinear processes can result in condensation of photons into the localized stationary states. As a consequence, an initially broad spectrum can evolve into essentially delta function distribution conserving the net photon energy and the total photon number. As a demonstration, we consider a Gaussian initial photon energy distribution of the form



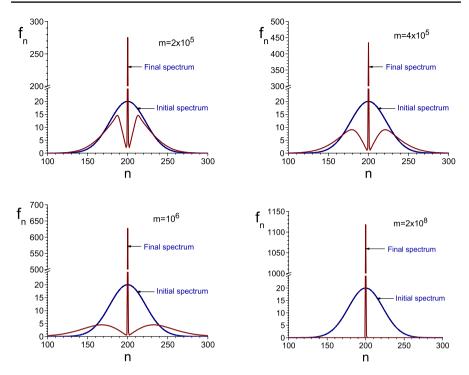


Fig. 2 (Color figure online) Initial and final radiation spectrum in the cavity. Initial spectrum is given by the Gaussian distribution (10) with  $f_0 = 20$ ,  $n_0 = 200$  and  $\sigma = 22.36$ . The final spectrum is obtained by letting the system evolve according to Eqs. (1) and (2) with  $\kappa = 0.000892$  after different numbers of scattering events  $m = 2 \times 10^5$ ,  $4 \times 10^5$ ,  $10^6$  and  $2 \times 10^8$ 

$$f_n = f_0 \exp\left(-\frac{\left(n - n_0\right)^2}{2\sigma^2}\right). \tag{10}$$

The total number of photons in the spectrum is:

$$N = f_0 \sum_{n=0}^{\infty} \exp\left(-\frac{\left(n - n_0\right)^2}{2\sigma^2}\right) \approx \sqrt{2\pi} f_0 \sigma. \tag{11}$$

Next we take  $f_0 = 20$ ,  $n_0 = 200$  and  $\sigma = \sqrt{500} = 22.36$ . Then, Eq. (11) yields that the total number of photons in the spectrum is N = 1121. We want to condense these photons into a single localized state (6). Such localized state has  $1/\kappa - \kappa$  photons. We take  $\kappa = 0.000892$ ; then,  $1/\kappa - \kappa = 1121$ , that is number of photons in the localized state (6) is equal to the total number of photons in the cavity. Under such conditions, the initially broad spectrum with width  $2\sigma = 45$  (that is spectrum covers 45 modes) condenses into a single mode by means of the nonlinear Raman scattering mechanism.



To show this, we perform numerical simulations in which we let the initial Gaussian spectrum evolve according to Eqs. (1) and (2) and take the number of the scattering events to be  $m = 2 \times 10^5$ ,  $4 \times 10^5$ ,  $10^6$  and  $2 \times 10^8$ . Figure 2 presents the results. The initial broad spectrum gradually evolves into the single localized state in which  $f_{200} = 1118.07$  and  $f_{201} \approx 1$ , while all other  $f_n$  are essentially equal to zero. The final localized state contains 99.8% of the total field energy. The obtained numerically steady state is well described by the analytical solution (6).

According to the second law of thermodynamics, entropy S of the isolated particle system should grow during the condensation process. To show that this is indeed the case, we calculate S as a function of the number of the scattering events m. In terms of  $f_n$ , the entropy reads [30]

$$S = k_B \sum_{n} \left[ (1 + f_n) \ln(1 + f_n) - f_n \ln(f_n) \right]. \tag{12}$$

In Fig. 3, we plot S(m) for spectrum evolution of Fig. 2. The total entropy of particles indeed increases with time even though most of the particles condense into the same state. For  $\kappa = 0.000892$ , the entropy of the localized state (6) into which photons are condensing is equal to  $9.41k_B$ , which is much smaller than the entropy of the initial photon distribution  $372k_B$  (see Fig. 3). Nevertheless, during condensation process the net entropy increases because a small fraction of bosons undergoes very large spectral broadening, which overcomes the entropy decrease caused by the spectrum narrowing.

To demonstrate the crucial role of nonlinearity in spectrum narrowing (stimulated scattering), we take the same initial Gaussian spectrum but now with  $f_0 = 2$ . For such initial state, the stimulated scattering is not dominant. For  $f_0 = 2$ ,  $n_0 = 200$  and  $\sigma = \sqrt{500}$  Eq. (11) gives that the total number of photons in the spectrum is N = 112. To match this number with the number of photons in the localized state (6), we take  $\kappa = 0.00892$ ; then,  $1/\kappa - \kappa = 112$ . Next we let the initial Gaussian

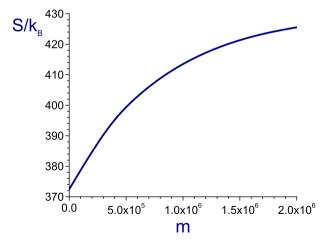
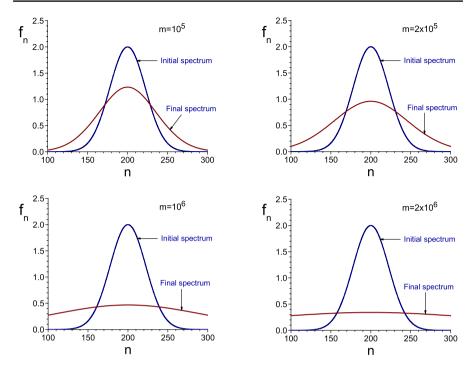


Fig. 3 (Color figure online) Entropy of the system of particles as a number of scattering events m for spectrum evolution of Fig. 2





**Fig. 4** (Color figure online) Initial and final radiation spectrum in the cavity. Initial spectrum is given by the Gaussian distribution (10) with  $f_0 = 2$ ,  $n_0 = 200$  and  $\sigma = 22.36$ . The final spectrum is obtained by letting the system evolve according to Eqs. (1) and (2) with  $\kappa = 0.00892$  after  $10^5$ ,  $2 \times 10^5$ ,  $10^6$  and  $2 \times 10^6$  scattering events

spectrum evolve according to Eqs. (1) and (2) and take the number of the scattering events  $m = 10^5$ ,  $2 \times 10^5$ ,  $10^6$  and  $2 \times 10^6$ . Figure 4 shows the result of numerical simulation. Raman-like scattering in the linear regime yields broadening of the initial spectrum. After a large number of scattering, the spectrum becomes constant, which is another steady-state solution of Eqs. (1) and (2).

## 5 Narrowing of Spectrum with Small Photon Occupation Number Using High-Intensity Seed

Occupation number of photons near the maxima of Planck distribution is about 0.06. For such low occupation numbers, the Raman-like scattering yields spectrum broadening. To condense the thermal solar spectrum into a narrow frequency interval, one can use a laser-like seed source, which has a high photon occupation number.

To demonstrate that this is possible, we take an initial spectrum given by the Gaussian distribution (10) with  $f_0 = 0.2$ ,  $n_0 = 1200$  and  $\sigma = \sqrt{50000} = 223.6$ . The total number of photons in the spectrum is N = 112, and photon occupation number in each mode is  $\leq 0.2$ , which alone yields no condensation. In addition,



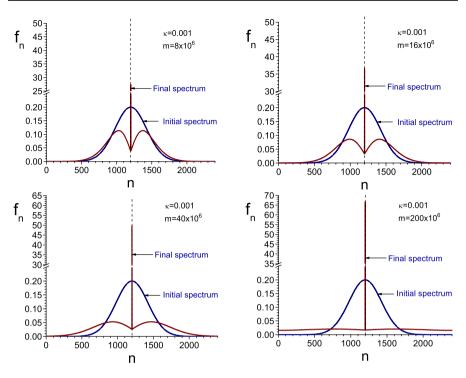


Fig. 5 (Color figure online) Initial and final radiation spectrum in the cavity. Initial spectrum is given by the Gaussian distribution (10) with  $f_0 = 0.2$ ,  $n_0 = 1200$  and  $\sigma = 223.6$ . In addition, there is a  $\delta$ -like seed pulse with  $f_{1200} = 900$  (shown by dash line). The final spectrum is obtained by letting the system to evolve according to Eqs. (1) and (2) with  $\kappa = 0.001$  after different numbers of scattering events  $m = 8 \times 10^6$ ,  $16 \times 10^6$ ,  $40 \times 10^6$  and  $200 \times 10^6$ 

we add a  $\delta$ -like seed pulse with  $f_{1200} = 900$  and let the system evolve according to Eqs. (1) and (2) with  $\kappa = 0.001$ . Figure 5 shows the spectrum after different numbers of the scattering events  $m = 8 \times 10^6$ ,  $16 \times 10^6$ ,  $40 \times 10^6$  and  $200 \times 10^6$ . After  $200 \times 10^6$  scattering events, 67 (out of 112) photons in the initial broad spectrum (about 60%) condense into the seed mode, while the rest of the spectrum broadens out. Thus, narrow-band sources with high photon occupation number can be used for condensation of thermal radiation.

## 6 Discussion and Summary

Steady state of quantum systems can be non-thermal and have a narrow spectral width. For example, if inverted atoms are injected sequentially into a cavity, this can yield lasing in a cavity mode with the highest gain, and the steady state of light in the cavity is non-thermal [31, 32]. The laser linewidth can be very narrow, much narrower than the natural linewidth of the atom's emission line [33]. In this example,



the energy flows from the atoms (active medium) into the cavity field, which is balanced by the field leakage out of the cavity.

In this paper, we consider a model in which photons in an ideal cavity interact with a two-level atom. The atom can inelastically scatter light changing the photon frequency by  $\pm E_0/\hbar$  (Raman process). During such scattering, the number of photons is conserved. As the ground-state two-level atom acquires energy  $E_0$  from the radiation field, it becomes excited and gives the energy  $E_0$  back to the field by going to the ground state in the next scattering event. Our system is analogous to a system of N Bose particles in the microcanonical ensemble.

We find that apart from a thermal-like steady state with a broad energy distribution, the system has steady states in which one or several modes are macroscopically occupied. The system can evolve into the state with the narrow energy distribution if initially the stimulated scattering dominates. Thus, Raman scattering, combined with photon stimulated emission and absorption, can result in condensation of a broad spectrum radiation into a narrow frequency interval conserving the total photon number and energy. We demonstrate the crucial role of stimulated processes (which make equations nonlinear) in photon condensation and show that condensation can also occur for weak (solar-like) radiation in the presence of intense laser beam acting as a seed.

In our system, the spectrum narrowing occurs due to stimulated processes, which is analogous to a laser. However, in contrast to the laser, there is no need for an external energy source (active medium). The present effect also differs from BEC because the latter occurs due to thermalization, with the macroscopically populated mode being a consequence of equilibrium Bose statistics.

Our findings could have practical applications in solar energy conversion into electric power. Efficiency of semiconductor solar cells is low mostly because they convert the broad solar spectrum into electric energy [34, 35]. If it would be possible first to convert the broad spectrum into a narrow one without changing the total radiation energy, this could enhance the efficiency of a single pn junction cell by a factor of two [36].

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