

 Open access • Journal Article • DOI:10.1103/PHYSREVA.88.033829

Schwinger-Keldysh theory for Bose-Einstein condensation of photons in a dye-filled optical microcavity — [Source link](#)

[A. W. de Leeuw](#), [H. T. C. Stoof](#), [Rembert A. Duine](#)

Institutions: [Utrecht University](#)

Published on: 18 Sep 2013 - [Physical Review A](#) (American Physical Society)

Topics: [Photon gas](#), [Photon](#), [Optical microcavity](#), [Optical cavity](#) and [Laser pumping](#)

Related papers:

- [Bose–Einstein condensation of photons in an optical microcavity](#)
- [Nonequilibrium Model of Photon Condensation](#)
- [Bose-Einstein condensation of exciton polaritons](#)
- [Experimental evidence for inhomogeneous pumping and energy-dependent effects in photon Bose-Einstein condensation](#)
- [Thermalization of a two-dimensional photonic gas in a `white wall' photon box](#)

Share this paper:    

View more about this paper here: <https://typeset.io/papers/schwinger-keldysh-theory-for-bose-einstein-condensation-of-1c4mvvh9jy>

Schwinger-Keldysh theory for Bose-Einstein condensation of photons in a dye-filled optical microcavity

A.-W. de Leeuw,^{*} H. T. C. Stoof, and R. A. Duine*Institute for Theoretical Physics, Utrecht University, Leuvenlaan 4, 3584 CE Utrecht, Netherlands*

(Received 21 June 2013; published 18 September 2013)

We consider Bose-Einstein condensation of photons in an optical cavity filled with dye molecules that are excited by laser light. By using the Schwinger-Keldysh formalism we derive a Langevin field equation that describes the dynamics of the photon gas and, in particular, its equilibrium properties and relaxation towards equilibrium. Furthermore we show that the finite lifetime effects of the photons are captured in a single dimensionless damping parameter that depends on the power of the external laser pumping the dye. Finally, as applications of our theory we determine spectral functions and collective modes of the photon gas in both the normal and the Bose-Einstein condensed phases.

DOI: [10.1103/PhysRevA.88.033829](https://doi.org/10.1103/PhysRevA.88.033829)

PACS number(s): 42.50.Ct, 33.80.-b, 03.75.Hh

I. INTRODUCTION

After the theoretical prediction of Bose-Einstein condensation (BEC) in 1925 [1,2], it took until 1995 for the first direct experimental observation of this phenomenon in weakly interacting atomic vapors [3–5]. In addition to these atomic gases, BEC of bosonic quasiparticles such as magnons [6], exciton-polaritons [7,8], and photons [9] is now also observed. The Bose-Einstein condensates of these quasiparticles form a different class of condensates as they are not in true thermal equilibrium.

These nonequilibrium Bose-Einstein condensates are driven by external pumping to compensate for the particle losses and thereby to keep the average number of particles in the system at a constant level. In these systems the steady state of the Bose gas is therefore determined by interparticle interactions that lead to quasiequilibrium and by the balance between pumping and particle losses. Furthermore, contrary to dilute atomic gases, the temperature is typically constant in these experiments. Instead, one varies the strength of the external pumping power while keeping the system at a constant temperature. Above some critical value of the pumping power, the density of particles in the system is above the critical density, and the system undergoes BEC.

Another special feature of these pumped systems is the temperature at which BEC occurs. Since BEC happens when the phase-space density is of the order of unity [10], the temperature at which the magnons, exciton-polaritons, and photons condense is inversely related to their mass to the power $3/2$. Although these particles do not even always have a bare mass, they are all formally equivalent to bosons with an effective mass that is several orders of magnitude smaller than that of alkali atoms. Therefore these systems undergo BEC at temperatures in the range of 10 to 300 K instead of in the nK regime relevant for the atomic Bose-Einstein condensates.

In order to get a detailed understanding of these nonequilibrium Bose-Einstein condensates, we from now onwards focus on the photon experiment of Klaers *et al.* [9]. This experiment is concerned with a photon gas in a dye-filled

optical resonator. The distance between the cavity mirrors is chosen such that the emission and absorption of photons with a certain momentum in the longitudinal direction dominates over that of other momenta. Thus this component of the momentum of the photons is fixed, and the photon gas becomes equivalent to a Bose gas with a small effective mass. Furthermore, the gas becomes effectively two-dimensional. In general this prohibits observing BEC at nonzero temperature since a homogeneous two-dimensional Bose gas can only condense at zero temperature [10]. However, due to the curvature of the cavity mirrors there is a harmonic potential for the photons. Therefore BEC of photons is observed above some critical pumping power since a harmonically trapped two-dimensional Bose gas can exhibit BEC at a nonzero temperature [11,12].

Theoretically, a lot of progress has been made for BEC of magnons and exciton-polaritons [13–19]. Although the observation of Bose-Einstein condensation of photons is more recent, it has also motivated theoretical studies: Klaers *et al.* predicted a regime of large fluctuations of the condensate number [20]. Furthermore, the authors of Ref. [21] found that the photons cannot reach thermal equilibrium for small absorption and emission rates. The modification of the Stark shift of an atom in a Bose-Einstein condensate of photons was investigated in Ref. [22], and conditions for BEC of photons that are in thermal equilibrium with atoms of dilute gases were derived in Ref. [23].

In this article we develop a theory for the photon experiment performed by Klaers *et al.* [9]. We describe this photon system by using the Schwinger-Keldysh formalism, which is commonly used in the quantum optics community (see, for example, Ref. [24]). In Sec. II we derive an effective action for the photons. In Sec. III we use this effective action to derive a Langevin field equation for the photons including Gaussian noise, which incorporates the effect of thermal and quantum fluctuations. The main advantage of this approach is that it simultaneously treats coherent and incoherent effects. In particular, it enables us to describe the complete time evolution of the photons, including the relaxation towards equilibrium; thus equilibrium properties can also be obtained. Subsequently, we show that the finite lifetime effects of the photons can be captured in a single dimensionless parameter α that depends on the power of the external laser pumping the dye. In Sec. IV we

^{*}A.deLeeuw1@uu.nl

calculate equilibrium properties of the homogeneous photon gas in the normal and Bose-Einstein condensed phase, such as spectral functions, collective modes, and damping. We end with conclusions and outlook in Sec. V.

II. MODEL

In this section we derive an effective action for the photons by using the Schwinger-Keldysh formalism developed in Ref. [25]. In particular, the photons are coupled to a reservoir of dye molecules. The energy of the photons is given by

$$\epsilon_\gamma(\mathbf{k}) = \hbar c \sqrt{k_x^2 + k_y^2 + k_z^2}, \quad (1)$$

where \hbar is Planck's constant, c is the speed of light in the medium, and \mathbf{k} is the transverse momentum of the photon. In agreement with the experiment $k_z = \pm k_\gamma$ since the frequency of the pump laser is such that in the longitudinal direction the absorption of photons with mode number $q = 7$ dominates over other absorption processes [9]. For the molecules we take an ideal gas in a box with volume V . Since this gas is at equilibrium and at room temperature, we describe the translational motion of the molecules by a classical Maxwell-Boltzmann distribution. Furthermore, we model these molecules as a two-level system with energy difference $\Delta > 0$ between the excited and ground states. This is a simplification since these molecules have a rovibrational structure. Therefore the dye molecules have more than two levels as the rovibrational structure divides the ground and excited levels into several sublevels. However, in this section we will show that we can model this multilevel system by introducing an effective mass for the molecules in our two-level model.

At time t_0 the photons are coupled to the molecules with a momentum-independent coupling constant g . To study the dynamics of the coupled system at times larger than t_0 we consider the action

$$\begin{aligned} S[a_{\mathbf{k}}, a_{\mathbf{k}}^*, b_{\mathbf{k}}, b_{\mathbf{k}}^*] &= \sum_{\mathbf{k}} \int_{G^\infty} dt a_{\mathbf{k}}^*(t) \left\{ i\hbar \frac{\partial}{\partial t} - \epsilon_\gamma(\mathbf{k}) + \mu_\gamma \right\} a_{\mathbf{k}}(t) \\ &+ \sum_{\mathbf{p}, \rho} \int_{G^\infty} dt b_{\mathbf{p}, \rho}^*(t) \left\{ i\hbar \frac{\partial}{\partial t} - \epsilon(\mathbf{p}) + \mu_\rho - K_\rho \right\} b_{\mathbf{p}, \rho}(t) \\ &- \frac{i}{\sqrt{2V}} \sum_{\mathbf{k}, \mathbf{p}} \int_{G^\infty} dt g a_{\mathbf{k}}(t) b_{\mathbf{p}, \downarrow}(t) b_{\mathbf{p}+\mathbf{k}, \uparrow}^*(t) + \text{H.c.} \\ &+ \frac{i}{\sqrt{2V}} \sum_{\mathbf{k}, \mathbf{p}} \int_{G^\infty} dt g a_{\mathbf{k}}(t) b_{\mathbf{p}, \downarrow}(t) b_{\mathbf{p}+\mathbf{k}, \uparrow}^*(t) + \text{H.c.} \quad (2) \end{aligned}$$

Here time is integrated along the Schwinger-Keldysh contour G^∞ , which is depicted in Fig. 1. The photons are described by the fields $a_{\mathbf{k}}(t)$ and $a_{\mathbf{k}}^*(t)$. Furthermore, $\epsilon_\gamma(\mathbf{k})$ is given by



FIG. 1. The Schwinger-Keldysh contour G^∞ . The integration is first from t_0 to ∞ and then back from ∞ to t_0 .

Eq. (1), and μ_γ is the chemical potential of the photons. For now we neglect the harmonic potential for the photons since this term is not important for the coupling between molecules and photons. The fields $b_{\mathbf{p}, \rho}(t)$ and $b_{\mathbf{p}, \rho}^*(t)$ describe the dye molecules, with ρ being equal to \downarrow or \uparrow , corresponding to the ground and excited states, respectively. Also, $\epsilon(\mathbf{p}) = \hbar^2 \mathbf{p}^2 / 2m_d$, with m_d being the mass of the rhodamine 6G molecule. Moreover, K_ρ accounts for the energy difference between the molecular states, and we take $K_\downarrow = 0$ and $K_\uparrow = \Delta$. The last two terms describe the processes of the absorption and emission of a photon, respectively. Here g is the coupling strength between the photons and molecules, $\mathbf{k}_+ = (k_x, k_y, k_\gamma)$, and $\mathbf{k}_- = (k_x, k_y, -k_\gamma)$. Note that the structure of the interaction terms is a consequence of the expansion of the photon field in terms of a standing wave, instead of a plane wave, in the z direction. Furthermore, the summation over \mathbf{k} is two-dimensional, whereas the summations over \mathbf{p} are three-dimensional. The latter convention will be used throughout the paper.

In this system one of the two molecular chemical potentials determines the density of molecules. The experiment of Klaers *et al.* used rhodamine 6G dye solved in methanol with a concentration of $1.5 \times 10^{-3} \text{ mol L}^{-1}$. Therefore we use a typical value of $n_m = 9 \times 10^{23} \text{ m}^{-3}$ for the density of molecules. Furthermore the value of $\Delta\mu = \mu_\uparrow - \mu_\downarrow$ determines the polarization of the molecules. This polarization is defined as

$$P(\Delta\mu) := \frac{N_\uparrow - N_\downarrow}{N_\uparrow + N_\downarrow} = \frac{e^{\beta(\Delta\mu - \Delta)} - 1}{e^{\beta(\Delta\mu - \Delta)} + 1}, \quad (3)$$

where β is the inverse of the thermal energy $k_B T$ and N_\uparrow and N_\downarrow are, respectively, the total number of excited-state and ground-state molecules. For small $\Delta\mu$ all molecules are in the ground state. By increasing the value of $\Delta\mu$, the number of molecules in the excited state increases. Since the total number of molecules is constant, the number of ground-state molecules thereby decreases. Thus for increasing $\Delta\mu$ the polarization increases. Moreover, the polarization is exactly zero for $\Delta\mu = \Delta$. A plot of the polarization as a function of $\Delta\mu$ is given in Fig. 2. The parameter $\Delta\mu$ is also important for making a

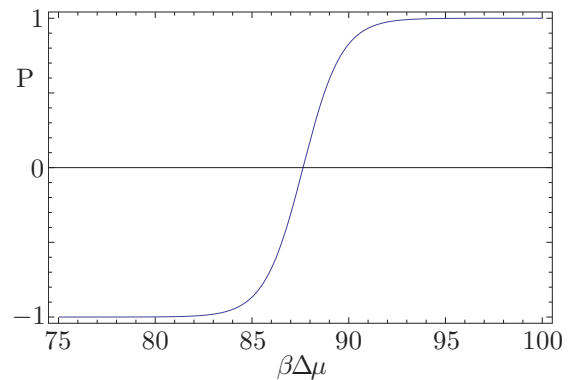


FIG. 2. (Color online) Plot of the polarization of the molecules P at room temperature $T = 300 \text{ K}$ as a function of $\beta\Delta\mu$ for a density of molecules $n_m = 9 \times 10^{23} \text{ m}^{-3}$, $\Delta = 3.63 \times 10^{-19} \text{ J}$. The polarization is exactly zero if $\Delta\mu$ is equal to the energy difference between the excited and ground states of the molecules.

connection with the experiment since the number of excited molecules and thereby the polarization are determined by the pumping power of the external laser.

In our nonequilibrium theory the chemical potential of the photons μ_γ becomes only well defined after the photon gas equilibrates by coupling to the dye molecules. Since both the sum of the number of ground-state molecules and excited-state molecules and the sum of the number of excited-state molecules and photons are constant, we have in equilibrium

$$\Delta\mu = \mu_\gamma. \quad (4)$$

To derive an effective action for the photons, we first integrate out the molecules. Next, we use perturbation theory up to second order in g to obtain

$$\begin{aligned} S^{\text{eff}}[a_{\mathbf{k}}, a_{\mathbf{k}}^*] &= \sum_{\mathbf{k}} \int_{G^\infty} dt' \int_{G^\infty} dt a_{\mathbf{k}}^*(t) \\ &\times \left(\left\{ i\hbar \frac{\partial}{\partial t} - \epsilon_\gamma(\mathbf{k}) + \mu_\gamma \right\} \delta(t, t') - \hbar \Sigma(\mathbf{k}, t, t') \right) a_{\mathbf{k}}(t'), \end{aligned} \quad (5)$$

where the photon self-energy due to coupling with the dye is given by

$$\begin{aligned} \Sigma(\mathbf{k}, t, t') &= \frac{-i|g|^2}{2\hbar^2 V} \sum_{\mathbf{p}} G_\downarrow(\mathbf{p}, t', t) \\ &\times \{G_\uparrow(\mathbf{k}_+ + \mathbf{p}, t, t') + G_\uparrow(\mathbf{k}_- + \mathbf{p}, t, t')\}. \end{aligned} \quad (6)$$

It turns out that both terms on the right-hand side are equal, and therefore we can write

$$\Sigma(\mathbf{k}, t, t') = \frac{-i|g|^2}{\hbar^2 V} \sum_{\mathbf{p}} G_\downarrow(\mathbf{p}, t', t) G_\uparrow(\mathbf{k}_+ + \mathbf{p}, t, t'). \quad (7)$$

Here the Keldysh Green's function for the dye molecules is given by

$$\begin{aligned} G_\rho(\mathbf{p}, t, t') &= i e^{-i[\epsilon(\mathbf{p}) - \mu_\rho + K_\rho](t-t')/\hbar} \\ &\times \{\Theta(t, t')[N_\rho(\mathbf{p}) - 1] + \Theta(t', t)N_\rho(\mathbf{p})\}, \end{aligned} \quad (8)$$

where $\Theta(t, t')$ and $\Theta(t', t)$ are the corresponding Heaviside functions on the Schwinger-Keldysh contour. Furthermore, the occupation numbers for the dye molecules are

$$N_\rho(\mathbf{p}) = e^{-\beta[\epsilon(\mathbf{p}) - \mu_\rho + K_\rho]}, \quad (9)$$

with $\rho \in \{\uparrow, \downarrow\}$. Since this action is defined on the Schwinger-Keldysh contour, we can only use this action to calculate quantities on the Schwinger-Keldysh contour. However, the relevant physical quantities should be calculated on the real-time axis. Therefore we need to transform this action into an action that is defined on this real-time axis. As is shown in Ref. [25], this boils down to determining the retarded, advanced, and Keldysh self-energies. Roughly speaking, the advanced and retarded self-energies determine the dynamics of the single-particle wave function in the gas, i.e., the coherent dynamics, and the Keldysh component accounts for the dynamics of their occupation numbers, i.e., the incoherent dynamics.

In the continuum limit the retarded self-energy becomes

$$\begin{aligned} \hbar \Sigma^{(+)}(\mathbf{k}, t - t') &= \frac{i}{\hbar} \Theta(t - t') \int \frac{d\mathbf{p}}{(2\pi)^3} |g|^2 \\ &\times e^{i[\epsilon(\mathbf{k}_+, \mathbf{p}) + \Delta\mu](t-t')/\hbar} \{N_\uparrow(\mathbf{k}_+ + \mathbf{p}) - N_\downarrow(\mathbf{p})\}. \end{aligned} \quad (10)$$

Here we used Eqs. (6) and (8) and we defined $\epsilon(\mathbf{k}_+, \mathbf{p}) = \epsilon(\mathbf{p}) - \epsilon(\mathbf{k}_+ + \mathbf{p}) - \Delta$. In Fourier space this self-energy reads

$$\begin{aligned} \hbar \Sigma^{(+)}(\mathbf{k}, \omega) &:= S(\mathbf{k}, \omega) - iR(\mathbf{k}, \omega) \\ &:= \int d(t - t') \hbar \Sigma^{(+)}(\mathbf{k}, t - t') e^{i\omega(t-t')}. \end{aligned} \quad (11)$$

Since the molecules behave as a Maxwell-Boltzmann gas at room temperature, we can find an analytical expression for $R(\mathbf{k}, \omega)$. We obtain

$$R(\mathbf{k}, \omega) = A(\mathbf{k}, \omega) \frac{|g|^2 m_d^2}{2|\mathbf{k}_+| \pi \beta \hbar^4} \sinh \left\{ \frac{\beta \hbar \omega}{2} \right\}, \quad (12)$$

with

$$\begin{aligned} A(\mathbf{k}, \omega) &= \exp\{\beta(\mu_\downarrow + \mu_\uparrow - \Delta)/2\} \\ &\times \exp \left\{ -\frac{\beta}{4} \left[\epsilon(\mathbf{k}_+) + \frac{(\Delta - \Delta\mu - \hbar\omega)^2}{\epsilon(\mathbf{k}_+)} \right] \right\}. \end{aligned} \quad (13)$$

Furthermore, in Fourier space the Keldysh self-energy is given by

$$\begin{aligned} \hbar \Sigma^K(\mathbf{k}, \omega) &= i \int \frac{d\mathbf{p}}{(2\pi)^2} \delta[\hbar\omega + \epsilon(\mathbf{k}_+, \mathbf{p}) + \Delta\mu] \\ &\times |g|^2 \{2N_\downarrow(\mathbf{p})N_\uparrow(\mathbf{k}_+ + \mathbf{p}) - N_\downarrow(\mathbf{p}) - N_\uparrow(\mathbf{k}_+ + \mathbf{p})\}. \end{aligned} \quad (14)$$

Since the dye is in quasiequilibrium, this Keldysh self-energy can be related to the imaginary part of the retarded self-energy. We find

$$\hbar \Sigma^K(\mathbf{k}, \omega) = -2i[1 + 2N(\omega)]R(\mathbf{k}, \omega), \quad (15)$$

where

$$N(\omega) = \frac{1}{e^{\beta\hbar\omega} - 1}. \quad (16)$$

This result is known as the fluctuation-dissipation theorem. As we show in the next section, this result guarantees that the photon gas relaxes towards thermal equilibrium in the limit of $t \rightarrow \infty$.

To make further progress, we have to determine typical numerical values for Δ and g appropriate for the experiment of Klaers *et al.* These values can be obtained by looking at the physical meaning of the self-energy. Consider a system of molecules that can be in either a ground state or excited state. If we apply a laser to this system, we can measure, for instance, the total number of molecules in the excited state. This number depends on the rate of photon absorption and emission and therefore on the lifetime of the photons. Since the imaginary part of the retarded self-energy is related to the lifetime of the photon, we can determine the emission and absorption spectra of the molecules with the help of our expression for $R(\mathbf{k}, \omega)$.

In order to obtain the absorption and emission spectrum separately, we take a closer look at the retarded self-energy given by Eq. (10). In this expression the factor with the Maxwell-Boltzmann distribution can be rewritten as $N_{\downarrow}(\mathbf{p})[N_{\uparrow}(\mathbf{k}_+ + \mathbf{p}) \pm 1] - N_{\uparrow}(\mathbf{k}_+ + \mathbf{p})[N_{\downarrow}(\mathbf{p}) \pm 1]$. The first term can be understood as the absorption of a photon, since this statistical factor accounts for the process where a ground-state molecule scatters into an excited state. The factor $N_{\downarrow}(\mathbf{p})$ simply is the number of molecules that can undergo the collision, and $[N_{\uparrow}(\mathbf{k}_+ + \mathbf{p}) \pm 1]$ denotes the Bose enhancement factor or Pauli blocking factor depending on the quantum statistics of the dye molecules. By using a similar reasoning the second term can be understood as the emission of a photon. Hence the part of the self-energy proportional to $N_{\downarrow}(\mathbf{p})$ is related to the absorption spectrum, and the part proportional to $N_{\uparrow}(\mathbf{k}_+ + \mathbf{p})$ is related to the emission spectrum.

The absorption and emission spectra are usually obtained in experiments where the number of photons is not conserved. So in these systems the photons have no chemical potential. To make a comparison, we therefore have to set $\mu_{\downarrow} = \mu_{\uparrow}$. Furthermore, contrary to the experiment of Klaers *et al.*, there is no restriction on the momentum of the photons. This implies that the photon field should be expanded into plane waves instead of standing waves. Therefore the fourth term on the right-hand side of the action in Eq. (2) is absent, and the prefactor of the third term is changed into $1/\sqrt{V}$. However, this modification leaves the expressions for the self-energies unchanged.

In order to get more insight into the role of the parameters of our model in the absorption and emission spectra, we first consider the experiment of Klaers *et al.*, and we keep \mathbf{k} fixed. Then, the spectra have a maximum at

$$\hbar\omega_{\pm} = \Delta \pm \frac{\hbar^2 \mathbf{k}_+^2}{2m_d}, \quad (17)$$

where we used $\Delta\mu = 0$. Here the plus sign is the maximum of the absorption spectrum, and the minus sign corresponds to the position of the maximum of the emission spectrum. So we obtain a difference in frequency between the maximum of the absorption and emission spectra. This difference is also obtained experimentally and is known as the Stokes shift. From this expression we find that the value of m_d determines the value of the Stokes shift. Furthermore we can see from Eq. (17) that we can change the position of the peaks by varying Δ .

Now we turn to more conventional experiments, where the absorption of laser light by the medium is measured as a function of frequency. To obtain the absorption and emission spectra only as a function of frequency, we have to consider the self-energy on the shell and thus replace \mathbf{k}_+ with ω/c . For the physical mass of the rhodamine 6G molecule, we obtain peaks that are too narrow and a Stokes shift that is too small. This is because we neglected the rovibrational structure of the molecules. Due to this rovibrational structure there are many possible transitions since the excited and ground levels are split into several sublevels. Therefore there is a whole range of photon energies which can be absorbed or emitted by the molecule. This causes a considerable broadening of the spectra. As mentioned before, we model this rovibrational structure by choosing an effective mass for the molecules.

We can also see explicitly from Eqs. (12) and (13) that decreasing the value of m_d will indeed broaden the peaks. For $\Delta = 3.63 \times 10^{-19}$ J and $m_d = 9.3 \times 10^{-35}$ kg we recover in good approximation the normalized absorption and emission spectra given in Ref. [9].

Up to now, we have considered the relative absorption and emission spectra. To obtain the correct height of these spectra, we have to find an appropriate value for g . By using Ref. [26], we can actually compare our results to the experimentally obtained absorption and emission spectra. However, to calculate the emission spectrum for this particular experiment within our formalism, we have to take into account that the emission of a photon can be in an arbitrary direction. Thus to obtain the emission spectrum we have to perform an integral which averages over all possible emission directions. However, the absorption spectrum can be obtained without performing additional integrals, and therefore we focus on this spectrum to obtain a numerical value for g . Then, as a consequence of our formalism, for our purposes the correct emission spectrum is also incorporated.

Before we can fit g , we have to relate our calculated decay rates to the measured absorption cross sections. We have

$$\frac{dN}{dx} = -n_{\downarrow} \sigma_{\text{abs}}(\omega), \quad (18)$$

where the left hand-side is the number of absorbed photons dN at a distance dx along the path of a beam. Furthermore, n_{\downarrow} is the density of ground-state molecules, and σ_{abs} is the absorption cross section. By using Fermi's golden rule, we obtain that dN/dt is equal to $-2R_{\text{abs}}(\omega)/\hbar$. Here $R_{\text{abs}}(\omega)$ denotes the absorption term in the imaginary part of the self-energy. Hence

$$\sigma_{\text{abs}}(\omega) = \frac{2R_{\text{abs}}(\omega)}{c\hbar n_{\downarrow}}. \quad (19)$$

Since the molecules behave as a classical Maxwell-Boltzmann gas,

$$n_{\downarrow} = \left(\frac{m_{d,\text{real}}}{2\pi\hbar^2\beta} \right)^{3/2} e^{\beta\mu_{\downarrow}}. \quad (20)$$

Note that contrary to the mass of the dye molecules used in the self-energies, we here use the real mass of the dye molecules to obtain the correct densities. Thus m_d is the effective mass to model the rovibrational structure of the molecules, and $m_{d,\text{real}} \simeq 7.95 \times 10^{-25}$ kg is the physical mass of a rhodamine 6G molecule. By using our expression for $R(\mathbf{k},\omega)$ we observe that the absorption cross section is independent of μ_{\downarrow} . Therefore we do not need to specify the number of molecules to obtain a numerical estimate for g . Furthermore we can relate the absorption cross section given by Eq. (19) to the molecular extinction coefficient obtained in Ref. [26]. According to Ref. [27],

$$\sigma = (3.82 \times 10^{-21} \text{ cm}^3 \text{ mol}^{-1} \text{ L}^{-1})\epsilon, \quad (21)$$

where $\epsilon = 1.16 \times 10^5 \text{ L mol}^{-1} \text{ cm}^{-1}$ is the molar extinction coefficient. This results in $g \simeq 6.08 \times 10^{-26} \text{ J m}^{3/2}$. A plot of the absorption and emission cross sections for the obtained numerical values for Δ , m_d , and g is given in Fig. 3. The shown emission cross section is obtained from Eq. (19) by replacing $R_{\text{abs}}(\omega)$ with $R_{\text{emis}}(\omega)$. As mentioned before, this is

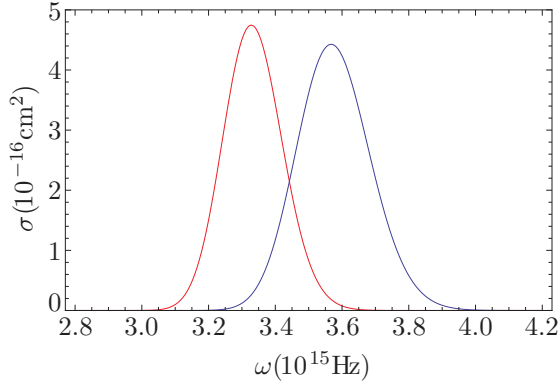


FIG. 3. (Color online) The absorption and emission cross sections of the photons obtained from the imaginary part of the retarded self-energy for $m_d = 9.3 \times 10^{-35}$ kg, $\Delta = 3.63 \times 10^{-19}$ J, and $g = 6.08 \times 10^{-26}$ J m^{3/2}. The red (left) curve corresponds to the emission, and the blue (right) curve denotes the absorption of photons. The absorption cross section is given by Eq. (19), and the plotted emission cross section is obtained from the same equation by replacing $R_{\text{abs}}(\omega)$ with $R_{\text{emis}}(\omega)$.

not the physical emission cross section since that can only be obtained by integrating over all directions of emission.

III. NONEQUILIBRIUM PHYSICS

We introduce a complex field $\phi(\mathbf{x}, t)$ for the photons such that

$$\langle |\phi(\mathbf{k}, t)|^2 \rangle = N(\mathbf{k}, t) + \frac{1}{2}, \quad (22)$$

where $N(\mathbf{k}, t)$ corresponds to the average occupation number of the single-particle state with momentum \mathbf{k} at time t . As is shown in Ref. [25], $\phi(\mathbf{x}, t)$ obeys a Langevin field equation for describing the dynamics of the photon gas. This equation ultimately reads

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} \phi(\mathbf{x}, t) &= [H(\mathbf{x}) + T|\phi(\mathbf{x}, t)|^2] \phi(\mathbf{x}, t) \\ &+ \int d\mathbf{x}' dt' \hbar \Sigma^{(+)}(\mathbf{x} - \mathbf{x}', t - t') \phi(\mathbf{x}', t') + \eta(\mathbf{x}, t), \end{aligned} \quad (23)$$

where the Hamiltonian

$$H(\mathbf{x}) = -\frac{\hbar^2 \nabla^2}{2m_{\text{ph}}} - \mu_\gamma + \hbar c k_\gamma + \frac{1}{2} m_{\text{ph}} \Omega^2 |\mathbf{x}|^2. \quad (24)$$

Here $\mathbf{x} = (x, y)$ and the field $\phi^*(\mathbf{x}, t)$ satisfies the complex conjugate equation. Furthermore, to obtain this equation, we expanded Eq. (1) for small transverse momenta, using that $k_\gamma(\mathbf{x})$ is position dependent due to the curvature of the cavity mirrors. In this equation $m_{\text{ph}} \simeq 6.7 \times 10^{-36}$ kg is the effective mass of the photons, and $\Omega \simeq 2.6 \times 10^{11}$ Hz is the trapping frequency of the harmonic potential. We also introduced a self-interaction term with strength $T \simeq 1.2 \times 10^{-36}$ J m². According to Ref. [9], this self-interaction of the photons arises from Kerr nonlinearity or thermal lensing in the dye. This self-interaction is an effective interaction and therefore also incorporates renormalization from interactions at high momenta and energies. Finally, the Gaussian noise $\eta(\mathbf{x}, t)$

satisfies

$$\langle \eta(\mathbf{x}, t) \eta^*(\mathbf{x}', t') \rangle = \frac{i\hbar}{2} \hbar \Sigma^K(\mathbf{x} - \mathbf{x}', t - t'). \quad (25)$$

Here, the brackets denote averaging over different realizations of the noise. In general it is difficult to determine correlation functions from these equations, especially because of the nonlocality of the retarded self-energy. However, since we are interested in Bose-Einstein condensation of photons we focus on the low-energy behavior of this self-energy. In the low-energy regime we are interested in $k\xi$ of the order of unity and ω around $\omega_B(\mathbf{k})$, where

$$\hbar \omega_B(\mathbf{k}) = \sqrt{\left(\frac{\hbar^2 \mathbf{k}^2}{2m_{\text{ph}}} \right)^2 + 2n_0 T \left(\frac{\hbar^2 \mathbf{k}^2}{2m_{\text{ph}}} \right)}, \quad (26)$$

is the Bogoliubov dispersion and

$$\xi = \frac{\hbar}{2\sqrt{m_{\text{ph}} n_0 T}} \quad (27)$$

is the coherence length, with n_0 being the density of condensed photons. Note that k is the norm of the two-dimensional momentum vector $\mathbf{k} = (k_x, k_y)$. Since for the experiment of Ref. [9] the critical number of photons $N_c \simeq 77\,000$ and the diameter of the condensate is measured as a function of the condensate fraction, we can make an estimate of n_0 . We obtain condensate densities in the range of at least 10^{12} – 10^{13} m⁻².

We make a low-energy approximation to the imaginary part of the retarded self-energy. As we see in Fig. 4, this is a good approximation in the low-energy regime. Furthermore, the real part of the retarded self-energy is small, and as a zeroth-order approximation we neglect this contribution. Thus we approximate

$$\hbar \Sigma^{(+)}(\mathbf{k}, \omega) = -i\alpha \hbar \omega, \quad (28)$$

and we can write for the Langevin field equation that

$$i\hbar(1 + i\alpha) \frac{\partial}{\partial t} \phi(\mathbf{x}, t) = [H(\mathbf{x}) + T|\phi(\mathbf{x}, t)|^2] \phi(\mathbf{x}, t) + \eta(\mathbf{x}, t). \quad (29)$$

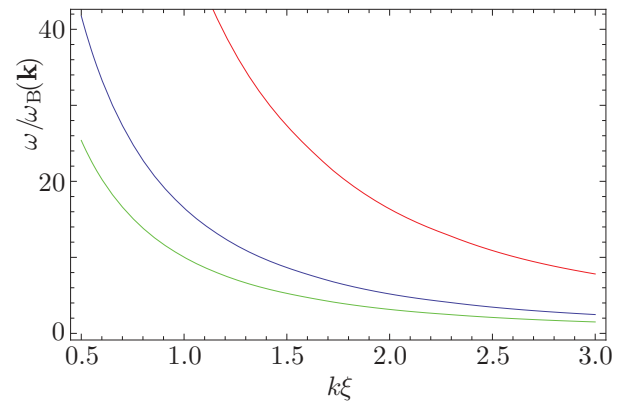


FIG. 4. (Color online) Plot of the validity of the linear approximation of the retarded self-energy for $n_0 = 10^{12}$ m⁻² and certain values of ω and k . The blue (middle), red (top), and green (bottom) curves are for, respectively, $\Delta\mu$ equal to 3.4×10^{-19} , 3.7×10^{-19} , and 4.0×10^{-19} J. Below the curves is the region where the linear approximation is within 1% of the actual value of the self-energy.

This is the equation which determines the complete dynamics of the photon gas. The finite lifetime effects are captured by the single dimensionless parameter α , which depends on the difference between the chemical potentials of the excited-state and ground-state molecules. Furthermore the noise $\eta(\mathbf{x}, t)$ is related to the Keldysh self-energy via Eq. (25), and in this approximation

$$\begin{aligned} \hbar \Sigma^K(\mathbf{x}' - \mathbf{x}, t' - t) &= -i\delta(\mathbf{x} - \mathbf{x}') \\ &\times 2\alpha\hbar \int \frac{d\omega}{2\pi} [1 + 2N(\omega)] \omega e^{-i\omega(t'-t)}. \end{aligned} \quad (30)$$

The explicit dependence of α on $\Delta\mu$ is given by

$$\alpha = \alpha_{\max} n_m \frac{e^{-C(\Delta\mu - \Delta)^2}}{\cosh\left\{\frac{1}{2}\beta(\Delta\mu - \Delta)\right\}}, \quad (31)$$

where n_m is the density of dye molecules,

$$C = \frac{\beta m_d}{2\hbar^2 |k_\gamma|^2}, \quad (32)$$

and

$$\alpha_{\max} = \sqrt{\frac{\pi m_{d,\text{real}}}{8\beta\hbar^2 k_\gamma^2}} \left(\frac{\beta |g| m_d}{m_{d,\text{real}}}\right)^2 e^{-\beta\hbar^2 k_\gamma^2 / 8m_d}. \quad (33)$$

The damping parameter α is inversely proportional to the photon lifetime and accounts for the decay of photons due to the interaction with the dye molecules. The emission and absorption of photons are equally important for the photon equilibration. Therefore α has a maximum when there is an equal amount of excited-state and ground-state molecules, i.e., for $\Delta\mu = \Delta$ where $P = 0$. This also explains the symmetric form of α around $\Delta\mu = \Delta$. Namely, α should be symmetric while changing the sign of the polarization as this only switches the excited-state and ground-state molecule densities. A plot of α as a function of $\Delta\mu$ is shown in Fig. 5.

The Langevin field equation given by Eq. (29) incorporates the complete dynamics of the photons. However, we still need to check that for large times the photon distribution function relaxes to the correct equilibrium. For this it suffices to consider the homogeneous case and to neglect the self-interaction of

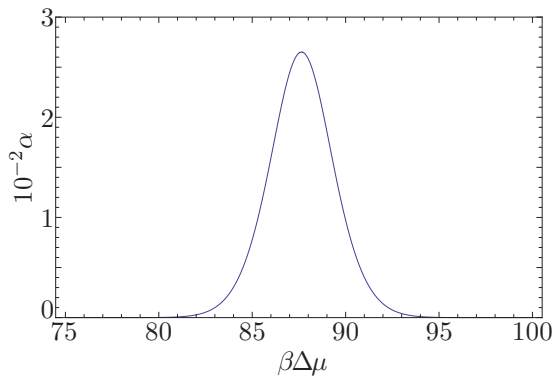


FIG. 5. (Color online) Plot of the dimensionless damping parameter α as a function of $\beta\Delta\mu$. For this plot we used $n_m = 9 \times 10^{23} \text{ m}^{-3}$. The parameter α has a maximum value of about 2.65×10^{-2} at $\Delta\mu$ equal to Δ .

the photons. For purposes of generality, we do not make a low-energy approximation to the self-energy, and we Fourier transform the Langevin field equation into

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} \phi(\mathbf{k}, t) &= (\epsilon_\gamma(\mathbf{k}) - \mu_\gamma) \phi(\mathbf{k}, t) + \eta(\mathbf{k}, t) \\ &+ \int_{t_0}^{\infty} dt' \hbar \Sigma^{(+)}(\mathbf{k}, t - t') \phi(\mathbf{k}, t'). \end{aligned} \quad (34)$$

As mentioned in the previous section, the fluctuation-dissipation theorem given by Eq. (15) should ensure that the gas relaxes towards thermal equilibrium. To check that this formalism contains this correct equilibrium, we assume that $\langle \phi(\mathbf{k}, t) \phi^*(\mathbf{k}, t') \rangle$ only depends on the difference $t - t'$ and write

$$\langle \phi(\mathbf{k}, t) \phi^*(\mathbf{k}, t') \rangle = \int \frac{d\omega}{2\pi} G(\mathbf{k}, \omega) e^{-i\omega(t-t')}. \quad (35)$$

Then

$$i\hbar \frac{d}{d(t+t')} \langle \phi(\mathbf{k}, t) \phi^*(\mathbf{k}, t') \rangle = 0, \quad (36)$$

and for $t' = t$ we obtain the equilibrium value for $\langle \phi(\mathbf{k}, t) \phi^*(\mathbf{k}, t) \rangle$. Since we are interested in the equilibrium, we consider times much larger than t_0 . Therefore we are allowed to take the limit of $t_0 \rightarrow -\infty$. Now Eq. (36) can be rewritten as

$$\begin{aligned} &\langle \eta(\mathbf{k}, t) \phi^*(\mathbf{k}, t') \rangle - \langle \phi(\mathbf{k}, t) \eta^*(\mathbf{k}, t') \rangle \\ &= \int_{-\infty}^{\infty} dt'' \langle \phi(\mathbf{k}, t) \phi^*(\mathbf{k}, t'') \rangle \hbar \Sigma^{(-)}(\mathbf{k}, t'' - t') \\ &\quad - \int_{-\infty}^{\infty} dt'' \hbar \Sigma^{(+)}(\mathbf{k}, t - t'') \langle \phi(\mathbf{k}, t'') \phi^*(\mathbf{k}, t') \rangle. \end{aligned} \quad (37)$$

Here we used $\hbar \Sigma^{(-)}(\mathbf{k}, t' - t) = [\hbar \Sigma^{(+)}(\mathbf{k}, t' - t)]^*$. Furthermore, since the field $\phi(\mathbf{k}, t)$ and its complex conjugate depend on the noise, we have a nonzero value for $\langle \eta(\mathbf{k}, t) \phi^*(\mathbf{k}, t') \rangle$, which can be determined by formally integrating Eq. (34) and using Eq. (25).

In Fourier space Eq. (37) is given by

$$-\frac{1}{2i} \Sigma^K(\mathbf{k}, \omega) G^{(+)}(\mathbf{k}, \omega) G^{(-)}(\mathbf{k}, \omega) = G(\mathbf{k}, \omega), \quad (38)$$

where the retarded (+) and advanced (-) photon Green's functions are determined by

$$\hbar G^{(\pm), -1}(\mathbf{k}, \omega) = \hbar \omega^\pm - \epsilon_\gamma(\mathbf{k}) + \mu_\gamma - \hbar \Sigma^{(\pm)}(\mathbf{k}, \omega). \quad (39)$$

To make further progress, we introduce the spectral function

$$\begin{aligned} \rho(\mathbf{k}, \omega) &= -\frac{1}{\pi\hbar} \text{Im}[G^{(+)}(\mathbf{k}, \omega)] \\ &= \frac{1}{\pi} \frac{R(\mathbf{k}, \omega)}{[\hbar\omega - \epsilon_\gamma(\mathbf{k}) + \mu_\gamma - S(\mathbf{k}, \omega)]^2 + [R(\mathbf{k}, \omega)]^2} \\ &= \frac{1}{\pi\hbar^2} R(\mathbf{k}, \omega) G^{(+)}(\mathbf{k}, \omega) G^{(-)}(\mathbf{k}, \omega). \end{aligned} \quad (40)$$

This spectral function $\rho(\mathbf{k}, \omega)$ can be interpreted as a single-particle density of states. Therefore we can calculate densities in equilibrium by multiplying this spectral function with the Bose-distribution function $N(\omega)$ and then integrating over $\hbar\omega$.

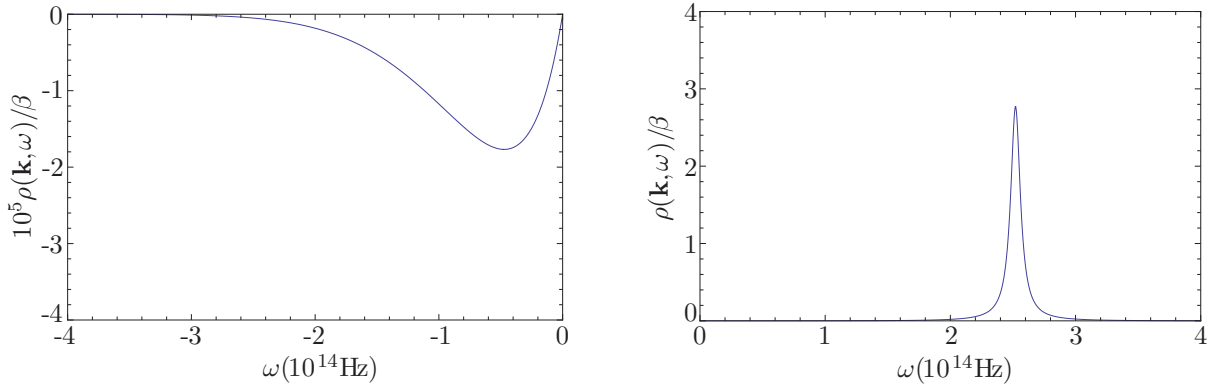


FIG. 6. (Color online) The spectral function as a function of the frequency ω for $k_y = k_x = 0$ for $n_m = 9 \times 10^{23} \text{ m}^{-3}$ and $\mu_\gamma = 3.5 \times 10^{-19} \text{ J}$. The negative (positive) contribution for negative (positive) frequencies is shown on the left (right) side.

Hence

$$N(\mathbf{k}) = \int d(\hbar\omega) N(\omega) \rho(\mathbf{k}, \omega), \quad (41)$$

where $N(\mathbf{k})$ is the number of photons in a state with momentum \mathbf{k} . Thus in equilibrium

$$G(\mathbf{k}, \omega) = 2\pi\hbar \left(\frac{1}{2} + N(\omega)\right) \rho(\mathbf{k}, \omega), \quad (42)$$

where we used the fluctuation-dissipation theorem in Eq. (15). Hence

$$\langle \phi(\mathbf{k}, t) \phi^*(\mathbf{k}, t) \rangle = N(\mathbf{k}) + \frac{1}{2}. \quad (43)$$

By comparing this result to Eq. (22), we find that the average occupation numbers $N(\mathbf{k}, t)$ relax to $N(\mathbf{k})$. During this calculation we did not use an approximation for the imaginary part of the retarded self-energy. However, we can do the same calculation for $R(\mathbf{k}, \omega)$ given by $\alpha\hbar\omega$. This approximation will directly manifest itself in the fluctuation-dissipation theorem and ultimately in the spectral function. Therefore in this approximation the equilibrium occupation numbers are also given by Bose-Einstein distribution functions.

IV. EQUILIBRIUM

In the previous section we have shown that the complete dynamics of the photon gas can be obtained from a Langevin field equation for a complex field $\phi(\mathbf{x}, t)$. On top of this nonequilibrium physics, we have demonstrated the relaxation of the photons towards the correct equilibrium. In this section we discuss equilibrium properties of the photon gas, and we therefore set $\Delta\mu = \mu_\gamma$ according to Eq. (4). We perform calculations in both the normal and the Bose-Einstein condensed states.

A. Normal state

We first consider the spectral function of the photons defined in Eq. (40). The spectral function should satisfy two conditions. First of all, because we are dealing with bosons, the spectral function should be positive (negative) for positive (negative) frequencies. From Eq. (12) it is clear that $R(\mathbf{k}, \omega)$ has this property, and therefore this condition is indeed satisfied by the spectral function. Second, the spectral function should

satisfy the zeroth-frequency sum rule

$$\int d(\hbar\omega) \rho(\mathbf{k}, \omega) = 1. \quad (44)$$

By numerically integrating this spectral function, we check that we satisfy the sum rule for all chemical potentials smaller than the lowest energy of the photons.

As we can see from Fig. 6, the spectral function consists of a Lorentzian-like peak for positive frequencies and a continuum for negative frequencies. The latter is roughly five orders of magnitude smaller than the positive contribution. Since the positive contribution is approximately a Lorentzian, we can determine the lifetime of the photons by looking at the width of these peaks [28]. This lifetime is defined as the time for which a photon in a certain momentum state \mathbf{k} goes into another state with momentum \mathbf{k}' due to absorption and reemission by the molecules.

Numerically, we obtained for small momenta and $\beta\mu_\gamma$ up to roughly 87 a lifetime on the order of 10^{-13} s . If we increase μ_γ even further, the lifetime of the photons increases rapidly. Because for larger values of μ_γ the peaks of the spectral function are at smaller frequencies, we can also show this fact analytically. Since we know that the lifetime of the photon is related to the imaginary part of this pole, we need to determine the poles of Eq. (39).

In the previous section we found that for small frequencies we can use an approximation for the imaginary part of the self-energy in which it is linear in frequency. Within this approximation, the Green's function given by Eq. (39) has a pole at

$$\hbar\omega^{\text{pole}}(\mathbf{k}) = \frac{1 - i\alpha}{1 + \alpha^2} [\epsilon_\gamma(\mathbf{k}) - \mu_\gamma]. \quad (45)$$

Since $\alpha^2 \ll 1$, a typical lifetime of the photons in the normal state is given by

$$\tau(\mathbf{k}) = \frac{\hbar}{2\alpha(\epsilon_\gamma(\mathbf{k}) - \mu_\gamma)} \sim \frac{1}{\alpha\Omega}, \quad (46)$$

where α is given by Eq. (31), $\Delta\mu$ is sufficiently large, and Ω is the trap frequency of the photons. In the last step we used the fact that the photons are trapped in a harmonic potential, and therefore the typical energy of the photons is proportional to $\hbar\Omega$. From Fig. 5 we know that for the relevant values of

μ_γ , α is in the range of 10^{-3} – 10^{-2} . Therefore the lifetime of the photons is in the nanosecond regime, which agrees with Ref. [9]. We also note that the smallness of α implies that the collective-mode dynamics of the gas is underdamped as the ratio between the damping and frequency of the collective modes is precisely α .

B. Condensed state

In this section we consider the homogeneous two-dimensional photon gas below the critical temperature for Bose-Einstein condensation. To describe the condensate of photons we start from the following two-dimensional action:

$$S^{\text{eff}}[a^*, a] = \sum_{\mathbf{k}, n} \hbar G^{-1}(\mathbf{k}, i\omega_n) a_{\mathbf{k}, n}^* a_{\mathbf{k}, n} + \frac{T}{2} \sum_{\mathbf{K}, \mathbf{k}, \mathbf{q}, n, m, l} a_{\mathbf{K}-\mathbf{k}, n-m}^* a_{\mathbf{k}, m}^* a_{\mathbf{K}-\mathbf{q}, n-l} a_{\mathbf{q}, l}. \quad (47)$$

Here

$$\hbar G^{-1}(\mathbf{k}, i\omega_n) = i\hbar\omega_n - \epsilon_\gamma(\mathbf{k}) + \mu_\gamma - \hbar\Sigma(\mathbf{k}, i\omega_n), \quad (48)$$

and $\hbar\Sigma(\mathbf{k}, i\omega_n)$ follows from the retarded self-energy by Wick rotation of the real frequency to Matsubara frequencies $i\omega_n$. This action describes the same equilibrium physics as coming from the Langevin field equation in Eq. (34) since after a Wick rotation the equations of motion for the field $a_{\mathbf{k}, n}$ are determined by the average of the Langevin equations. Substituting $a_{0,0} \rightarrow a_{0,0} + \phi$ and requiring that the terms linear in the fluctuations vanish leads to the equation

$$\mu_\gamma = \hbar ck_\gamma + S(\mathbf{0}, 0) + Tn_0, \quad (49)$$

where n_0 is the density of condensed photons. This equation determines the chemical potential of the Bose-Einstein condensate of photons. We obtain $\beta\mu_\gamma \simeq 90.9$, and according to Eq. (3), we have a corresponding polarization of roughly 0.93. Therefore almost all molecules are in the excited state.

To determine the collective excitations of the condensate over the ground state we consider the action up to second order in the fluctuations. This is the so-called Bogoliubov approximation. So

$$S^{\text{Bog}}[a^*, a] = -\frac{1}{2} \sum_{\mathbf{k}, n} u_{\mathbf{k}, n}^\dagger \hbar G_B^{-1}(\mathbf{k}, i\omega_n) u_{\mathbf{k}, n}, \quad (50)$$

where

$$u_{\mathbf{k}, n} := \begin{bmatrix} a_{\mathbf{k}, n} \\ a_{-\mathbf{k}, -n}^* \end{bmatrix} \quad (51)$$

and

$$-\hbar G_B^{-1}(\mathbf{k}, i\omega_n) = \begin{bmatrix} 2Tn_0 & Tn_0 \\ Tn_0 & 2Tn_0 \end{bmatrix} - \begin{bmatrix} \hbar G^{-1}(\mathbf{k}, i\omega_n) & 0 \\ 0 & \hbar G^{-1}(\mathbf{k}, -i\omega_n) \end{bmatrix}. \quad (52)$$

Since μ_γ is given by Eq. (49), we obtain that $\text{Det}[G_B^{-1}(\mathbf{0}, 0)] = 0$. Therefore we have a gapless excitation, which agrees with Goldstone's theorem. By Wick rotating and solving for which

ω the determinant of this matrix vanishes, we can determine the dispersions. Since we are interested in the low-energy behavior, we can use Eq. (28) for the self-energy. In this approximation the dispersions are given by

$$(1 + \alpha^2)\hbar\omega(\mathbf{k}) = -i\alpha(\tilde{\epsilon}_\gamma(\mathbf{k}) + Tn_0) \pm \sqrt{-(\alpha Tn_0)^2 + \tilde{\epsilon}_\gamma(\mathbf{k})(\tilde{\epsilon}_\gamma(\mathbf{k}) + 2Tn_0)}, \quad (53)$$

with $\tilde{\epsilon}_\gamma(\mathbf{k}) = \epsilon_\gamma(\mathbf{k}) - \hbar ck_\gamma$. The imaginary part of the dispersion relations is always negative, and we find a lifetime in the nanosecond regime for n_0 in the range of 10^{12} – 10^{13} m^{-2} and excitations for which $k\xi < 0.2$, with $k = |(k_x, k_y)|$. For excitations with larger momentum the lifetime decreases until it approaches zero in the limit of $k \rightarrow \infty$.

Furthermore, we have the same behavior as was first shown in Ref. [29] for a nonequilibrium Bose-Einstein condensate of exciton-polaritons. Also in this case the dispersions become purely imaginary for small momenta. For the numerical values of the experiment and $n_0 = 10^{12}$ m^{-2} , we have purely imaginary dispersions for $k\xi < 2.2 \times 10^{-3}$. However, this does not imply that for small momenta there are only decaying quasiparticles at zero energy. This can be seen in the spectral function, which in this case corresponds to the imaginary part of $G_{B,11}(\mathbf{k}, \omega^+)$. In Figs. 7 and 8 we can see the two qualitatively different forms of the spectral function. For relatively large momenta, we have two peaks at the real part of the dispersions, and the width of the peaks is determined by the imaginary part of the dispersions. In the small-momenta region where both dispersions are purely imaginary, we have a continuum for both negative and positive frequencies. Still, the spectral function has a maximum and a minimum. Therefore in agreement with the large-momentum case we can also define the position of these extrema as the dispersion. So, contrary to what the analytical dispersion given by Eq. (53) suggests, for small momenta the spectral function also has a maximum and minimum at nonzero energy.

Finally, we check if the spectral function satisfies the sum rule given by Eq. (44). In the low-frequency approximation for the retarded self-energy we can integrate the spectral function

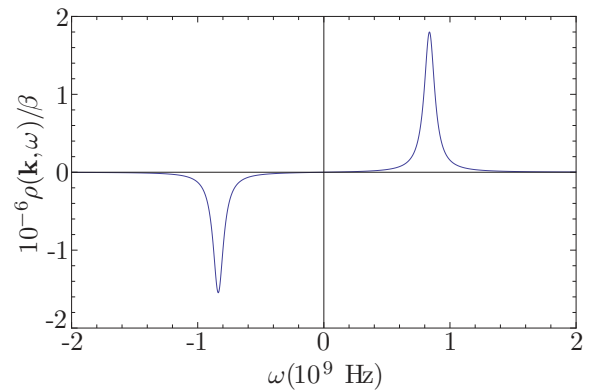


FIG. 7. (Color online) A plot of the spectral function as a function of ω for $n_m = 9 \times 10^{23}$ m^{-3} and $n_0 = 10^{12}$ m^{-2} . In this plot $k\xi \simeq 3.8 \times 10^{-2}$.

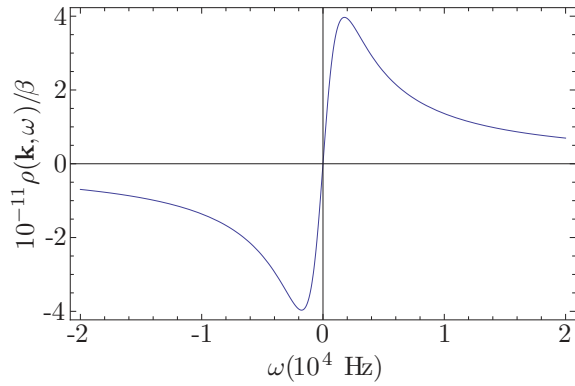


FIG. 8. (Color online) Plot of the spectral function as a function of ω for $n_m = 9 \times 10^{23} \text{ m}^{-3}$ and $n_0 = 10^{12} \text{ m}^{-2}$. In this plot $k\xi \simeq 1.9 \times 10^{-5}$.

analytically, and we obtain

$$\int d(\hbar\omega) \rho(\mathbf{k}, \omega) = \frac{1}{1 + \alpha^2}. \quad (54)$$

Since we are in the Bose-Einstein condensed phase $\alpha \simeq 4.5 \times 10^{-3}$, and we satisfy in very good approximation the sum rule. Note that this small deviation from the sum rule is a consequence of making the low-energy approximation to the self-energy. Namely, this approximation for the self-energy is only valid for small energies, and therefore the high-frequency behavior is not incorporated correctly. However, without this low-energy approximation the self-energy has the correct low-energy and high-energy limits, and we indeed find that the spectral function with the full self-energy satisfies the sum rule.

V. CONCLUSION AND OUTLOOK

In this work we constructed a theory for Bose-Einstein condensation of photons in a dye-filled cavity. By using the Schwinger-Keldysh formalism, we obtained a Langevin field equation that describes the complete dynamics of the photons.

In particular, it incorporates both the coherent and incoherent dynamics of the gas. Furthermore we found that the finite lifetime of the photons can be captured in a single parameter α , which depends on the external laser pumping the dye. In addition, we also found an analytic expression for this parameter. In the homogeneous case we have shown that our theory incorporates the correct equilibrium properties of the gas.

Subsequently, we calculated the collective modes and spectral functions of the homogeneous photon gas in the normal and Bose-Einstein condensed state. In both phases we found that the lifetime of the photons in the cavity is in the nanosecond regime, which is the same regime as found experimentally in Ref. [9]. Moreover, we obtained that the dynamics of the collective modes is underdamped. Furthermore, in agreement with the results of Ref. [29] for exciton-polaritons, we found in the Bose-Einstein condensed phase that dispersions become formally purely imaginary for small momentum. Nevertheless, for small momentum the spectral function also has qualitatively a maximum and minimum at nonzero energy. Finally, in both phases the spectral function is well behaved and satisfies the sum rule.

In future research we will consider in detail the fluctuations, in particular the phase fluctuations, of the photon Bose-Einstein condensate. For a condensate density of $n_0 \simeq 10^{12} \text{ m}^{-2}$, the trap length $l = \sqrt{\hbar/m_{\text{ph}}\Omega} \simeq 7.8 \times 10^{-6} \text{ m}$ is about 2 times smaller than the coherence length ξ . However, a condensate density of $n_0 \simeq 10^{13} \text{ m}^{-2}$ results in a trap length l that is about 2 times larger than the coherence length ξ . Both condensate densities are accessible experimentally [9], and therefore we intend to explore both the regime of Bose-Einstein condensation and the quasicondensate regime.

ACKNOWLEDGMENTS

It is a pleasure to thank Dries van Oosten for useful discussions. This work was supported by the Stichting voor Fundamenteel Onderzoek der Materie (FOM), the Netherlands Organization for Scientific Research (NWO), and the European Research Council (ERC).

-
- [1] S. Bose, *Z. Phys.* **26**, 178 (1924).
 - [2] A. Einstein, *Sitzungsber. Preuss. Akad. Wiss. Bericht* **3**, 18 (1925).
 - [3] M. H. Anderson, J. R. Ensher, M. R. Matthews, C. E. Wieman, and E. A. Cornell, *Science* **269**, 198 (1995).
 - [4] C. C. Bradley, C. A. Sackett, J. J. Tollett, and R. G. Hulet, *Phys. Rev. Lett.* **75**, 1687 (1995).
 - [5] K. B. Davis, M.-O. Mewes, M. R. Andrews, N. J. van Druten, D. S. Durfee, D. M. Kurn, and W. Ketterle, *Phys. Rev. Lett.* **75**, 3969 (1995).
 - [6] S. O. Demokritov, V. E. Demidov, O. Dzyapko, G. A. Melkov, A. A. Serga, B. Hillebrands, A. N. Slavin, *Nature (London)* **443**, 430 (2006).
 - [7] J. Kasprzak *et al.*, *Nature (London)* **443**, 409 (2006).
 - [8] R. Balili, V. Hartwell, D. Snoke, L. Pfeiffer, and K. West, *Science* **316**, 1007 (2007).
 - [9] J. Klaers, J. Schmitt, F. Verwinger, and M. Weitz, *Nature (London)* **468**, 545 (2010).
 - [10] C. J. Pethick and H. Smith, *Bose-Einstein Condensation in Dilute Gases* (Cambridge University Press, Cambridge, 2008).
 - [11] V. Bagnato and D. Kleppner, *Phys. Rev. A* **44**, 7439 (1991).
 - [12] W. J. Mullin, *J. Low Temp. Phys.* **106**, 615 (1997).
 - [13] S. M. Rezende, *Phys. Rev. B* **79**, 174411 (2009).
 - [14] S. A. Bender, R. A. Duine, and Y. Tserkovnyak, *Phys. Rev. Lett.* **108**, 246601 (2012).
 - [15] I. G. Savenko, T. C. H. Liew, and I. A. Shelykh, *Phys. Rev. Lett.* **110**, 127402 (2013).
 - [16] A. Amo *et al.*, *Nature (London)* **457**, 291 (2009).
 - [17] R. T. Brierley, P. B. Littlewood, and P. R. Eastham, *Phys. Rev. Lett.* **107**, 040401 (2011).
 - [18] M. Wouters, I. Carusotto, and C. Ciuti, *Phys. Rev. B* **77**, 115340 (2008).

- [19] D. Read, T. C. H. Liew, Y. G. Rubo, and A. V. Kavokin, *Phys. Rev. B* **80**, 195309 (2009).
- [20] J. Klaers, J. Schmitt, T. Damm, F. Vewinger, and M. Weitz, *Phys. Rev. Lett.* **108**, 160403 (2012).
- [21] P. Kirton and J. Keeling, *Phys. Rev. Lett.* **111**, 100404 (2013).
- [22] W. Fan, M. Yin, and Z. Cheng, arXiv:1301.7136.
- [23] A. Kruchkov and Y. Slyusarenko, *Phys. Rev. A* **88**, 013615 (2013).
- [24] E. G. Dalla Torre, S. Diehl, M. D. Lukin, S. Sachdev, and P. Strack, *Phys. Rev. A* **87**, 023831 (2013).
- [25] H. T. C. Stoof, *J. Low Temp. Phys* **114**, 11 (1999).
- [26] R. R. Birge, *KODAK Laser Dyes, Kodak publication JJ-169* (Eastman Kodak Company, Rochester, NY, 1987).
- [27] J. R. Lakowicz, *Principles of Fluorescence Spectroscopy* (Springer, New York, 2006).
- [28] H. T. C. Stoof, K. B. Gubbels, and D. B. M. Dickerscheid, *Ultracold Quantum Fields* (Springer, Dordrecht, 2009).
- [29] M. Wouters and I. Carusotto, *Phys. Rev. Lett.* **99**, 140402 (2007).