Interaction Effects on Number Fluctuations in a Bose-Einstein Condensate of Light

E. C. I. van der Wurff,* A.-W. de Leeuw, R. A. Duine, and H. T. C. Stoof
Institute for Theoretical Physics and Center for Extreme Matter and Emergent Phenomena, Utrecht University, Leuvenlaan 4, 3584 CE Utrecht, The Netherlands
(Received 9 April 2014; published 23 September 2014)

We investigate the effect of interactions on condensate-number fluctuations in Bose-Einstein condensates. For a contact interaction we variationally obtain the equilibrium probability distribution for the number of particles in the condensate. To facilitate comparison with experiment, we also calculate the zero-time delay autocorrelation function $g^{(2)}(0)$ for different strengths of the interaction. Finally, we focus on the case of a condensate of photons and find good agreement with recent experiments.

DOI: 10.1103/PhysRevLett.113.135301 PACS numbers: 67.85.Hj, 42.50.Ar, 42.50.Lc

Introduction.—Fluctuations are ubiquitous in physics: from the primordial quantum fluctuations in the early Universe that reveal themselves as fluctuations in the cosmic microwave background, to current fluctuations in every-day conductors. For large voltages, the latter fluctuations give rise to shot noise, that is due to the discrete nature of charge [1]. As a consequence, shot noise can be used to determine the quanta of the electric charge of the current carriers in conducting materials [2]. Indeed, it has been used to characterize the nature of Cooper pairs in superconductors [3] and the fractional charge of the quasiparticles of the quantum Hall effect [4]. For low voltages, the noise in the current is thermal and is called Johnson-Nyquist noise [5,6]. Contrary to shot noise, thermal noise is always present in electrical circuits, even if no externally applied voltage is present, since it is due to thermal agitation of charge carriers, that leads to fluctuating electromotive forces in the material.

Theoretically, fluctuations in equilibrium are described by the fluctuation-dissipation theorem, as formulated by Nyquist in 1928 and proven decades later [7]. This theorem relates the response of a system to an external perturbation to the fluctuations in the system in the absence of that perturbation. Given a certain fluctuation spectrum we can reconstruct the response of the system. Therefore, this theorem is very powerful, as was fervently argued by the Japanese physicist Kubo [8].

Having stressed the importance of fluctuations in physics and the information they contain, we now zoom in on condensate-number fluctuations as our main point of interest. Traditionally, weakly interacting Bose-Einstein condensates were first observed in dilute atomic vapors [9]. For these systems, it is very difficult to measure number fluctuations because typically number measurements are destructive. Therefore, theoretical work has focused more on density-density correlation functions [10,11].

In recent years, Bose-Einstein condensates of quasiparticles have also been created, such as exciton-polariton condensates [12], magnon condensates [13], and condensates of photons [14,15]. These condensates of quasiparticles are realized under different circumstances compared to the atomic condensates. For instance, the condensates of quasiparticles are created at higher temperatures than the condensates of dilute atomic gases: from several kelvin for the exciton-polariton condensate to room temperature for the photonic condensate. Additionally, the condensates of quasiparticles are not in true equilibrium, since the steady state is a dynamical balance between particle losses and particle gain by external pumping with a laser. Due to these differences, new experimental possibilities have opened up. For example, large number fluctuations of the order of the total particle number have been observed in a condensate of photons [16].

In this Letter we investigate number fluctuations in Bose-Einstein condensates. We start by introducing an effective contact interaction into the grand-canonical Hamiltonian of a Bose gas and derive an equilibrium probability distribution for the number of particles in the condensate. Subsequently, we investigate these distributions for different condensate fractions and interaction strengths. We also calculate the zero-time delay autocorrelation function $g^{(2)}(0)$ to quantify the number fluctuations. In this manner we are able to reproduce all experimental curves of Schmitt *et al.* [16] by using the interaction strength as a single fitting parameter. Having provided this interpretation of the experimental results, we finally discuss possible mechanisms for the interactions in a condensate of light.

Interaction effects on number fluctuations.—We consider a harmonically trapped Bose gas with a fixed number of particles. Because the condensates of quasiparticles are typically confined in one direction, we specialize to the case of two dimensions. However, the following treatment is completely general and can easily be generalized to higher or lower dimensions.

To investigate the number fluctuations, we first calculate the average number of particles $\langle N_0 \rangle$ in the condensate. Because the condensates of quasiparticles allow for a free exchange of bosons with an external medium we treat the system in the grand-canonical ensemble: the probability distribution $P(N_0)$ for the number of condensed particles is of the form $P(N_0) \propto \exp[-\beta\Omega(N_0)]$, with $\Omega(N_0)$ the grand potential of the gas of bosons.

To find the grand potential we use a variational wave function approach. We note that the bosons in the condensate typically interact with each other. A reasonable first approximation for the form of this interaction is a contact interaction, as essentially every interaction is renormalized to a contact interaction at long length and time scales, independent of the precise origin of the interactions. Therefore, we consider the following energy functional for the macroscopic wave function $\phi_0(\mathbf{x})$ of the Bose-Einstein condensate [17]:

$$\Omega[\phi_0(\mathbf{x})] = \int d\mathbf{x}\left(\frac{\hbar^2}{2m}|\nabla\phi_0(\mathbf{x})|^2 + V^{\rm ex}(\mathbf{x})|\phi_0(\mathbf{x})|^2 - \mu|\phi_0(\mathbf{x})|^2 + \frac{g}{2}|\phi_0(\mathbf{x})|^4\right), \quad (1)$$

where $\mathbf{x}$ is the two-dimensional position, the first term represents the kinetic energy of the condensate, $V^{\rm ex}(\mathbf{x}) = m\omega^2|\mathbf{x}|^2/2$ is the harmonic trapping potential, μ is the chemical potential for the particles, and g is the coupling constant of the effective pointlike interaction between the particles.

We use the Bogoliubov substitution $\phi_0(\mathbf{x}) = \sqrt{N_0}\psi_q(\mathbf{x})$, with the normalized variational wave function $\psi_q(\mathbf{x})$, such that $\int d\mathbf{x}|\phi_0(\mathbf{x})|^2 = N_0$. Subsequently, we minimize the energy as a function of the variational parameter q, which describes the width of the condensate. As an ansatz we take the variational wave function to be the Gaussian $\psi_q(\mathbf{x}) = (\sqrt{\pi}q)^{-1}\exp(-|\mathbf{x}|^2/2q^2)$. Substituting this into the energy given by Eq. (1) and minimizing with respect to the variational parameter, we obtain

$$q_{\rm min} = \sqrt[4]{\frac{2\pi\hbar^2 + mN_0 g}{2\pi\omega^2 m^2}} = q_{\rm ho}\sqrt[4]{1 + \frac{\tilde{g}N_0}{2\pi}}, \quad (2)$$

where we introduced the dimensionless coupling constant $\tilde{g} := mg/\hbar^2$ and the harmonic oscillator length $q_{\rm ho} = \sqrt{\hbar/m\omega}$. Note that for a sufficiently small number of condensate particles $q_{\rm min}$ reduces to $q_{\rm ho}$. For a large number of condensate particles the Thomas-Fermi ansatz for the wave function is in principle more appropriate. However, it is well known from the atomic condensates that even in this case the Gaussian approach is rather accurate [18].

We now substitute the minimal value for the variational parameter q into the energy functional, yielding the probability distribution

$$P(N_0) \propto \exp\left[\beta N_0\left(\mu - \hbar\omega\sqrt{1 + \frac{\tilde{g}N_0}{2\pi}}\right)\right], \quad (3)$$

where the normalization is $\int_0^\infty dN_0 P(N_0) = 1$.

Experimentally, the relevant parameter is the condensate fraction $x := \langle N_0\rangle/\langle N\rangle$, with N the total number of particles. Thus, to relate our results to the experiments we need a relation between $\langle N_0\rangle$ and the average total number of particles. For temperatures T below the critical temperature for Bose-Einstein condensation, the average number of particles in excited states can in a good approximation be determined from the ideal-gas result [19]. We obtain

$$\langle N_{\rm ex}(T)\rangle = \int_0^\infty \frac{g(\epsilon)d\epsilon}{\exp(\epsilon/k_B T) - 1} = \frac{N_s}{6}\left(\frac{\pi k_B T}{\hbar\omega}\right)^2, \quad (4)$$

where we used the density of states $g(\epsilon) = N_s\epsilon/(\hbar\omega)^2$ for a two-dimensional harmonic trapping potential [20]. The integer N_s denotes the number of spin components of the boson. The critical temperature T_c is defined by $\langle N\rangle = \langle N_{\rm ex}(T_c)\rangle$. With this criterion, we find

$$\langle N_0\rangle = \frac{xN_s}{6(1-x)}\left(\frac{\pi k_B T}{\hbar\omega}\right)^2. \quad (5)$$

Results.—Given an interaction strength $\tilde{g}$, we use the normalized probability distribution in Eq. (3) to calculate the chemical potential as a function of $\langle N_0\rangle$; i.e., $\mu = \mu(\langle N_0\rangle)$. Given a condensate fraction x, we then use Eq. (5) to calculate $\langle N_0\rangle$ and the corresponding μ. As an example we take $N_s = 2$ [21], which is appropriate for the Bose-Einstein condensate of photons [14–16]. Finally, we use the obtained chemical potential to plot the probability distribution at fixed x and $\tilde{g}$. Typical plots of the probability distribution for different condensate fractions are displayed in Fig. 1. Clearly, we have exponential behavior due to a

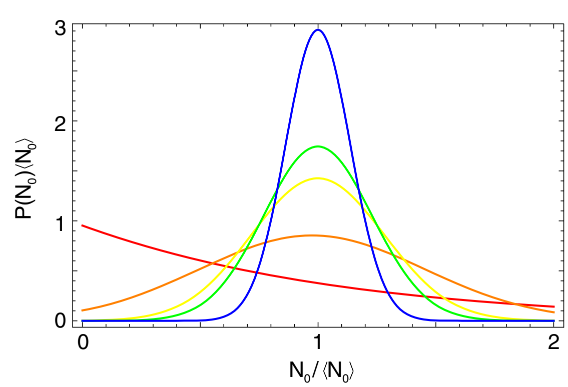

FIG. 1 (color online). Typical plot of the probability distribution for two-component bosons for a fixed interaction strength $\tilde{g} = 5\times10^{-6}$ and different condensate fractions (from bottom to top) $x_{\rm red} = 0.04$, $x_{\rm orange} = 0.28$, $x_{\rm yellow} = 0.40$, $x_{\rm green} = 0.45$, and $x_{\rm blue} = 0.58$.

Poissonian process for small condensate fractions and Gaussian behavior for larger condensate fractions. Physically, this shows that the effect of repulsive interactions is to reduce number fluctuations, as the interactions give fluctuations an energy penalty. Increasing the interaction strength yields Gaussian behavior for even smaller condensate fractions. These Gaussians are also more strongly peaked around $\langle N_0 \rangle$ for higher interaction strengths, which is expected since stronger interactions between the bosons leads to the supression of fluctuations.

Next, we obtain the second moment $\langle N_0^2 \rangle$ from the probability distribution $P(N_0)$. This gives us all the information needed to quantify the number fluctuations of the condensate. The time-averaged second-order correlation function of the light intensity is given by $g^{(2)}(\tau) := \langle I(t)I(t+\tau)\rangle / \langle I(t)\rangle\langle I(t+\tau)\rangle$, where τ is the time difference in the arrival of two beams of photons on the detectors in a Hanbury-Brown–Twiss experiment and $I(t)$ represents the intensity of those beams at time t. In fact, the corresponding zero-time delay autocorrelation function is given by

$$g^{(2)}(0) = \frac{\langle N_0^2 \rangle}{\langle N_0 \rangle^2}. \tag{6}$$

A plot of $g^{(2)}(0)$ against the condensate fraction is displayed in Fig. 2 for different interaction strengths $\tilde{g}$. We note that bunching of bosons takes place for all interactions at small condensate fractions. Theoretically, we know that for thermal photons $g^{(2)}(0) = 2$ [22], which is exactly what we observe in our plots for the corresponding case $x = 0$. For larger condensate fractions $g^{(2)}(0) \to 1$. The interpretation is as follows. Suppose we fix the condensate fraction x. At small interactions the quartic term in the energy in Eq. (1) is small and the minima of the energy are small and broad, yielding large number fluctuations. If we increase the interaction, the minima become deeper and more narrow, effectively reducing the fluctuations. The same reasoning holds for a fixed interaction strength and increasing condensate fractions, as we can also see in Fig. 1.

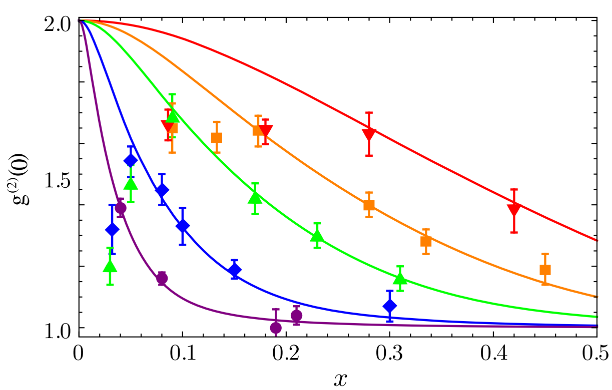

FIG. 2 (color online). Plot of the zero-time delay autocorrelation function $g^{(2)}(0)$ against the condensate fraction x for $\omega = 8\pi \times 10^{10}$ Hz and $T = 300$ K. The different curves correspond to different interaction strengths (from top to bottom): $\tilde{g}_{\rm red} = 5 \times 10^{-7}$, $\tilde{g}_{\rm orange} = 2 \times 10^{-6}$, $\tilde{g}_{\rm green} = 5 \times 10^{-6}$, $\tilde{g}_{\rm blue} = 3 \times 10^{-5}$, $\tilde{g}_{\rm purple} = 2 \times 10^{-4}$. All curves are compared to the included experimental points from Schmitt *et al.* [16]. The experimental results for small condensate fraction x are unreliable due to systematic measurement errors. Indeed, theoretically we have $\lim_{x\to 0} g^{(2)}(0) = 2$.

Comparison with experiments on a condensate of light.—The results in the previous sections were quite generic for a two-dimensional, harmonically trapped gas of bosons with two possible polarizations. In fact, measurements of $g^{(2)}(0)$ have been performed recently [16] in a Bose-Einstein condensate of photons, enabling us to compare our theory with experiments. In this experiment photons are confined in a dye-filled cavity, providing a harmonic potential and giving the photons an effective mass m by fixing their longitudinal momentum k_z [14]. The photons thermalize to the temperature of the dye solution by scattering of the dye molecules. Additionally, photon losses from the cavity are compensated by external pumping, yielding a constant average number of photons.

In Fig. 2 we plot the experimental data points of Ref. [16]. We are able to reproduce all data sets by tuning the interaction parameter $\tilde{g}$. Unfortunately, only one experimental value for $\tilde{g}$ is known. By measuring the size of the condensate for different condensate fractions, it was experimentally found that $\tilde{g} = (7 \pm 3) \times 10^{-4}$ [14], which only differs a factor of two with our result for the purple curve $g_{\rm purple} = 2 \times 10^{-4}$. However, we note that the trapping potential, concentration of dye molecules and effective photon mass were somewhat different for the purple data points and the measurement of the interaction strength. We expect the interaction strength to vary smoothly with variations in the experimental parameters. Hence the agreement is remarkable and points to the important role of interactions on number fluctuations in these experiments.

The data points in Fig. 2 were obtained for different dye molecule densities $n_{\rm mol}$ and detunings δ, which is roughly the difference between the cavity frequency and a dye-specific frequency related to the effective absorption threshold of the dye molecules. Within our theory, the dependence of number fluctuations on these parameters can be incorporated via their influence on the interactions. Therefore, it would be useful to perform systematic measurements of $\tilde{g}$ for different detunings and molecule concentrations, as is also proposed in Ref. [23]. With this information, we would be able to directly compare all experimental results with our theoretical predictions for the number fluctuations [24].

Summarizing, we have developed a general framework to calculate $g^{(2)}(0)$ for trapped Bose-Einstein condensates. Applying this to a condensate of light we have found good agreement with recent experiments. Note that an alternative explanation for the experimental results of Fig. 2 is discussed in Refs. [16,25] and is based on a master equation for the probability p_n to find n photons in the condensate ground state. The dye molecules are modeled as two-level systems without a center-of-mass degree of freedom. As there are many momentum modes available to the dye molecules at room temperature, we believe this is not an appropriate statistical description. Nevertheless, similar fits to ours in Fig. 2 can be obtained in this manner by using the number of dye molecules as a fitting parameter. The experimental data are then interpreted as signaling a transition from the canonical to the grand-canonical regime. However, the required size of the molecular heat bath varies from 10^9 to 10^{10} dye molecules, which usually is sufficiently large for there to be no difference in the choice of ensemble.

Discussion.—The question remains: what mechanism can cause an interaction that depends on both n_{mol} and the detuning δ? In fact, we conclude from the experimental data in Ref. [16] that the interaction behaves counter-intuitively: it decreases both for an increasing molecule density and for a decreasing detuning. Three different mechanisms are expected to play a role [23,26].

Thermal lensing is the phenomenon that the index of refraction n depends on the temperature of the medium. In the experiment of interest to us, nonradiative decay of the dye molecules, local fluctuations in the photon number, and the external pumping with a laser lead to temperature fluctuations around the average temperature T_0. For a homogenous temperature distribution this implies, to lowest order, that $n(T) = n(T_0) + \alpha(T - T_0)$. As the photon energy depends on the index of refraction, these temperature fluctuations couple to the photons. This leads to a photon-photon interaction as displayed in the Feynman diagram in Fig. 3. By assuming that the temperature fluctuations behave diffusively, we derive in the Supplemental Material [27] that the interaction strength due to this effect is given by

$$\tilde{g} = \frac{4m^3 c^4 \alpha^2 T_0}{3D_0 \hbar^2 n^6(T_0) c_p}, \tag{7}$$

where $D_0 = 8\pi/k_z$ is the length scale associated with the fixed longitudinal momentum k_z of the photons and c_p is the heat capacity of the solution. Note that this interaction has no explicit dependence on the detuning δ, or on the concentration of dye molecules n_{mol}, as it is fully determined by the properties of the solution. Using typical numerical values [31], we obtain an estimate for the interaction strength which is several orders of magnitude below the only experimental result $\tilde{g} \sim 10^{-4}$, suggesting that this is not the dominant interaction effect.

The other possible photon-photon interaction is due to the Kerr effect; i.e., the index of refraction is intensity dependent due to the properties of the solvent molecules, or due to photon-photon scattering mediated by the dye molecules. The former effect has been investigated in Refs. [23,32] and turns out to be negligible. We therefore consider the latter effect by means of a Feynman diagram in the form of a box, see Fig. 4.

We adopt a simplified description of the complex rovibrational structure of the dye molecules by describing them as an effective two-level system with an excited state that has a finite lifetime Γ. This leads to

$$\tilde{g}(\mu) = \frac{m g_{\mathrm{mol}}^4 \beta n_{\mathrm{mol}}}{\hbar^4 \Gamma^2 D_0} f(\beta\mu - \beta\delta), \tag{8}$$

where g_{mol} is the coupling strength of the photons to the molecules and $f(\beta\mu - \beta\delta)$ is a smooth dimensionless function peaked around zero. By fitting the self-energy of the photons to the experimental absorption spectrum of the dye, we obtain numerical values for g_{mol}, Γ and δ [33].

Subsequently, we have to solve $\tilde{g}(\mu)$ self-consistently with the Gross-Pitaevskii equation. For typical

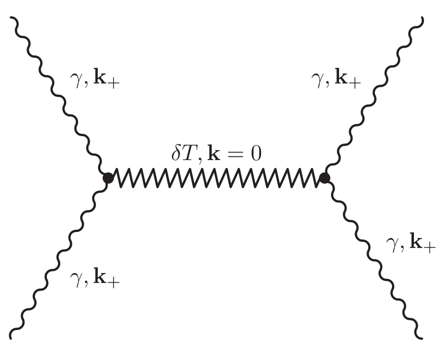

FIG. 3. Feynman diagram for the photon-photon interaction due to exchange of temperature fluctuations δT (zigzag propagator). The photons γ (wiggly propagator) are considered to be part of the condensate and are thus at zero frequency and at momentum $\mathbf{k}_+ = (0, 0, k_z)$, as their z-component momentum is fixed and $k_x = k_y = 0$ for the condensate of the homogeneous photon gas.

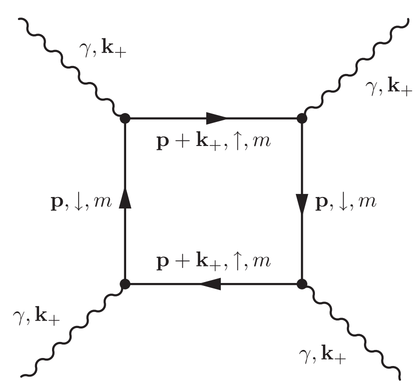

FIG. 4. Feynman diagram for the photon-photon interaction mediated by the dye molecules. Again we take the photons to be in the condensate: $\mathbf{k}_+ = (0, 0, k_z)$ and the frequency is zero. The molecule (straight line propagator) forms a closed loop of ground ($\downarrow$) and excited ($\uparrow$) states, with momentum $\mathbf{p}$ and Matsubara frequency ω_m.

experimental parameters we obtain a value for the interaction strength $\tilde{g}$ which is also small compared to the experimental value. However, the magnitude of $\tilde{g}$ is rather uncertain due to the simplification of the rovibrational energy spectrum of the dye molecules to a two-level system. Interestingly, we see from Eq. (8) that this interaction depends both on the detuning δ and the density of molecules n_{mol}. We show in the Supplemental Material [27] that a self-consistent solution can give rise to an interaction that decreases both for decreasing δ and increasing n_{mol}, which is precisely the counterintuitive behavior the experimental results exhibit.

In conclusion, we have calculated the effect of self-interactions on number fluctuations in Bose-Einstein condensates. Comparing our results with recent experiments on a condensate of light, we find good agreement. However, systematic measurements of the interaction strength are necessary to understand the true nature of the interaction. If the interaction is indeed a contact interaction at long wavelengths, then this would imply that the photon condensate is also a superfluid.

It is a pleasure to thank Dries van Oosten, Jan Klaers, and Martin Weitz for useful discussions and the latter two also for providing experimental data. This work is supported by the Stichting voor Fundamenteel Onderzoek der Materie (FOM) and is part of the D-ITP consortium, a program of the Netherlands Organisation for Scientific Research (NWO) that is funded by the Dutch Ministry of Education, Culture, and Science (OCW).

*e.c.i.vanderwurff@students.uu.nl

[1] C. Beenakker and C. Schönenberger, Phys. Today **56**, No. 5, 37 (2003).
[2] W. Schottky, Ann. Phys. (Berlin) **362**, 541 (1918).
[3] F. Lefloch, C. Hoffmann, M. Sanquer, and D. Quirion, Phys. Rev. Lett. **90**, 067002 (2003).
[4] R. de Picciotto, M. Reznikov, M. Heiblum, V. Umansky, G. Bunin, and D. Mahalu, Nature (London) **389**, 162 (1997).
[5] H. Nyquist, Phys. Rev. **32**, 110 (1928).
[6] J. Johnson, Phys. Rev. **32**, 97 (1928).
[7] H. B. Callen and T. A. Welton, Phys. Rev. **83**, 34 (1951).
[8] R. Kubo, Rep. Prog. Phys. **29**, 255 (1966).
[9] M. H. Anderson, J. R. Ensher, M. R. Matthews, C. E. Wieman, and E. A. Cornell, Science **269**, 198 (1995).
[10] E. Altman, E. Demler, and M. Lukin, Phys. Rev. A **70**, 013603 (2004).
[11] N. Cherroret and S. E. Skipetrov, Phys. Rev. Lett. **101**, 190406 (2008).
[12] J. Kasprzak *et al.*, Nature (London) **443**, 409 (2006).
[13] S. O. Demokritov, V. E. Demidov, O. Dzyapko, G. A. Melkov, A. A. Serga, B. Hillebrands, and A. N. Slavin, Nature (London) **443**, 430 (2006).
[14] J. Klaers, J. Schmitt, F. Vewinger, and M. Weitz, Nature (London) **468**, 545 (2010).
[15] J. Klaers, J. Schmitt, T. Damm, F. Vewinger, and M. Weitz, Appl. Phys. B **105**, 17 (2011).
[16] J. Schmitt, T. Damm, D. Dung, F. Vewinger, J. Klaers, and M. Weitz, Phys. Rev. Lett. **112**, 030401 (2014).
[17] H. T. C. Stoof, K. B. Gubbels, and D. B. M. Dickerscheid, *Ultracold Quantum Fields* (Springer, New York, 2009).
[18] G. Baym and C. J. Pethick, Phys. Rev. Lett. **76**, 6 (1996).
[19] Note that we have $k_B T \gg \mu$. This implies that the thermal cloud around the condensate can be accurately described by a noninteracting thermal gas of bosons.
[20] H. Smith and C. J. Pethick, *Bose-Einstein Condensation in Dilute Gases*, 2nd ed. (Cambridge University Press, Cambridge, England, 2008).
[21] The photons are described with an effective mass m, associated with their fixed longitudinal momentum. However, the effective nonrelativistic form of the Hamiltonian does not change the spin degeneracy for these photons inside the cavity.
[22] C. C. Gerry and P. L. Knight, *Introductory Quantum Optics* (Cambridge University Press, Cambridge, England, 2005).
[23] R. A. Nyman and M. H. Szymanska, Phys. Rev. A **89**, 033844 (2014).
[24] Should accurate experiments of $\tilde{g}$ be performed and call for more detailed quantitative comparison, we can improve our theory by numerically solving the Gross-Pitaevskii equation for the condensate wave function to calculate $\tilde{g}$ to high accuracy.
[25] J. Klaers, J. Schmitt, T. Damm, F. Vewinger, and M. Weitz, Phys. Rev. Lett. **108**, 160403 (2012).
[26] J. Klaers (private communication).
[27] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.113.135301, which includes Refs. [28–30], for a detailed derivation of the dimensionless interaction strength due to thermal lensing and dye-mediated photon-photon scattering.
[28] S. Yaltkaya and R. Aydin, Turk. J. Phys. **26**, 41 (2002).
[29] R. R. Birge, Kodak Report No. JJ-169.
[30] J. R. Lakowicz, *Principles of Fluorescence Spectroscopy* (Springer, New York, 2006).
[31] *Handbook of Chemistry and Physics*, 91st ed. (CRC Press, Boca Raton, 2009).
[32] R. Y. Chiao, T. H. Hansson, J. M. Leinaas, and S. Viefers, Phys. Rev. A **69**, 063816 (2004).
[33] A.-W. de Leeuw, H. T. C. Stoof, and R. A. Duine, Phys. Rev. A **88**, 033829 (2013).