Fluctuation-dissipation relation for a Bose-Einstein condensate of photons

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For equilibrium systems, the magnitude of thermal fluctuations is closely linked to the dissipative response to external perturbations. This fluctuation-dissipation relation has been described for material particles in a wide range of fields. Here we experimentally probe the relation between the number fluctuations and the response function for a Bose-Einstein condensate of photons coupled to a dye reservoir, demonstrating the fluctuation-dissipation relation for a quantum gas of light. The observed agreement of the scale factor with the environment temperature both directly confirms the thermal nature of the optical condensate and demonstrates the validity of the fluctuation-dissipation theorem for a Bose-Einstein condensate.

The fluctuation-dissipation theorem, relating the thermal fluctuations of a system at temperature \( T \) to its response to an external perturbation by the thermal energy \( k_B T \), is a cornerstone of statistical mechanics [1, 2]. Experimentally, it has been observed in a wide range of systems, e.g., with particles undergoing Brownian motion [3], the statistical fluctuations of electrical currents in resistors [4], and more recently also in cold atomic gas settings [5–7], including two-dimensional Bose superfluids in the strongly interacting regime [8, 9].

The relation provides an elegant approach to access microscopic properties of a system (fluctuations) by probing the response on a macroscopic level (dissipation), allowing to determine equilibrium quantities such as the structure factor, which would be difficult to access otherwise [10, 11].

For Bose-Einstein condensates, despite that this phase is one of the most thoroughly investigated quantum states of matter, the fluctuation-dissipation theorem could so far not be examined. In cold-atom condensates thermal number fluctuations are strongly suppressed [12, 13], and in optical condensates the driven-dissipative nature of such systems [14] has kept the possibility for a successful test of the fluctuation-dissipation relation an open question. Interestingly, the fluctuation-dissipation relation can be extended to non-equilibrium systems in steady state such as lasers [15], however there the scaling with temperature – a universal quantity – is replaced by system-specific two-point correlation functions [16, 17]. Along this line, theory work has recently pointed out that probing the validity of the fluctuation-dissipation relation provides a very direct and critical test of thermalization and allows one to characterize the eventual departure from equilibrium in optical quantum gases [18].

A new approach to study fluctuations and the corresponding response function in the condensed phase has emerged in quantum gases as exciton-polaritons and photons, where a coupling to reservoirs is realized [19–24]. In the latter experiments using photons, other than for the case of a blackbody gas, a thermodynamic phase transition to a Bose-Einstein condensate can be observed, e.g., in two-dimensional dye-filled optical microcavity systems [25–27]. Thermalization here is achieved by absorption-re-emission processes on dye molecules, which provide both an energy and a particle reservoir due to the possible interconversion of cavity photons and dye electronic excitations. This situation can be described by a grand canonical ensemble model, a physical setting for which unusually large fluctuations occur in the condensed phase [28–31]. Experimentally, the corresponding number fluctuations have been observed in the dye microcavity system, with the magnitude of fluctuations being tunable by adjusting the relative size of the condensate and the effective reservoir [20, 21].

In this letter, we report a measurement of both the spontaneous number fluctuations and the associated reactive response of a photon Bose-Einstein condensate coupled to a reservoir, demonstrating the validity of the fluctuation-dissipation theorem for a Bose-Einstein condensate. By tuning the reservoir size we find that the relation applies from canonical through to grand canonical conditions. Within experimental uncertainties, the observed scaling between fluctuations and the response is consistent with \( k_B T \), where \( T \approx 300 \text{ K} \) is the temperature of the reservoir. Such a critical test of the thermalized nature of an optical condensate as well as its coupling to the reservoir goes beyond earlier work that has verified, e.g., spectral properties and spatial redistribution of light in trapping potentials [25, 32–34], and is also of interest for thermometry in complex lattice or quenched systems [35–37].

Our photon Bose-Einstein condensates are prepared in a microcavity apparatus shown in Fig. 1(a), realized by two curved mirrors filled with a dye molecule solution of refractive index \( n = 1.44 \); see refs. [25, 38] for details. The cavity length \( D_0 = q\lambda_c/2\pi \approx 1.4 \mu m \) on the order of the optical wavelength \( \lambda_c \approx 575 \text{ nm} \) at mode number \( q = 7 \) introduces a low-energy cutoff at \( h\omega_c \approx 2.1 \text{ eV} \), with \( \omega_c = 2\pi c/\lambda_c \), speed of light \( c \) and the reduced Planck’s constant \( \hbar \). In the microcavity, the photon dispersion relation becomes two-dimensional and matter-like, i.e., energy scales quadratically with the wave vector; owing to the mirror curvature, the photons are harmonically trapped with frequency \( \Omega/2\pi \approx 40 \text{ GHz} \) [25]. Other than, e.g., in cold atoms or exciton-polaritons, equilibration of the ensemble does not occur by interparticle collisions, but by contact to a heat bath of photo-excitible dye molecules at room temperature. Thermalization is effective when the contact to the dye molecules dominates over losses, e.g., from mirror transmission [33], and the aver-
excitations, see Fig. 1(b) (top), preserves the total number of excitations \( X = n + M_e \) with \( n \) condensate photons and \( M_e = \sum_{i=0}^{\infty} f_i \) dye excitations; here, \( f_i = \{0, 1\} \) refers to the \( i \)-th dye molecule being in the electronic ground (0) or excited state (1), respectively. This assumption is well justified due to the small overlap of the excited molecules in the ground mode volume with the higher-lying photon modes in the harmonic trap, such that a reservoir-mediated cross-coupling is in general weak, and due to the spatially near-uniform excitation level, such that (on average) excitations do not flow to or away from the ground mode reservoir. In thermodynamic equilibrium, one obtains the photon number probability distribution for \( n \) particles in the Bose-Einstein condensate

\[
P_n = \frac{M!}{Z(M - X + n)!(X - n)!} e^{-\frac{\Delta n}{k_B T}},
\]

where the partition function \( Z \) is determined by normalization \( \sum_n P_n = 1 \). Note that eq. (1) is derived by assuming thermal contact of the total system of photons and dye molecules to a heat bath (the solvent). Microscopically, the detuning dependence of \( P_n \) is understood from the Kennard-Stepanov relation \( B_{abs}/B_{em} = \exp(\Delta \omega/k_B T) \) between the molecular Einstein coefficients for absorption \( B_{abs} \) and emission \( B_{em} \) of condensate photons. This detailed balance condition for large negative \( \Delta \) energetically favors a relatively large photon number \( \langle n \rangle \) as compared to the (generally larger) number of excited molecules \( \langle M_e \rangle \), see the left panel in Fig. 1(b); for \( \Delta \to 0^+ \), on the other hand, \( \langle M_e \rangle \) by far exceeds \( \langle n \rangle \) and presents the dominant contribution to \( X \) (right panel). Correspondingly, we can define an effective reservoir size \( M_{eff} = M/[2 + \cosh(h\Delta/k_B T)] \) based on the condition \( P_0 = P_1 \) [20, 29]. At this point, which occurs for \( \langle n \rangle^2 = M_{eff} \), the photon number statistics changes from super-Poissonian to Poissonian, realizing a distinction point between the grand canonical and canonical statistical regimes [38].

Using eq. (1), the lowest moments \( \langle n^k \rangle = \sum_n n^k P_n \) can be expressed through first and second-order derivatives of the partition function, \( \langle n \rangle = -Z^{-1}(k_B T/h) \frac{dZ}{d\Delta} \) and \( \langle n^2 \rangle = Z^{-1}(k_B T/h)^2 \frac{d^2Z}{d\Delta^2} \). We obtain an expression forming a fluctuation-dissipation relation

\[
\langle \Delta n^2 \rangle = -\frac{k_B T}{h} \left( \frac{d\langle n \rangle}{d\Delta} \right)_{X,T},
\]

which connects the squared photon number fluctuations \( \langle \Delta n^2 \rangle = \langle n^2 \rangle - \langle n \rangle^2 \) to a reactive response function \( (d\langle n \rangle/d\Delta)_{X,T} \) by thermal energy \( k_B T \) (in angular frequency units). The response function shown in the bottom panel of Fig. 1(b) describes the susceptibility of the mean condensate population \( \langle n \rangle \) to changes of the dye-cavity detuning \( \Delta \) at constant temperature \( T \) and excitation number \( X \); in other words, it qualifies how easy it is to “compress” photons into the dye reservoir. The fluctuation-dissipation relation in eq. (2) directly translates (intrinsic) thermal energy fluctuations of the dye molecules determined by \( k_B T \) into the mag-

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**Fig. 1.** (a) Experimental scheme to measure the number fluctuations and the response function of a photon Bose-Einstein condensate coupled to a reservoir inside a dye-microcavity. Part of the cavity emission recorded with a photomultiplier (PMT) yields the mean condensate population \( \langle n \rangle \); the other part is dispersed on a grating, and the spectrally filtered condensate evolution is recorded with a streak camera, giving \( g^{(\alpha)}(\tau) \) and the dye-cavity detuning \( \Delta \). Bottom: time-integrated spectrum showing the condensate mode at \( \omega_c \), well separated from the first excited states in the harmonic potential with trapping frequency \( \Omega \). (b) The reservoir is realized by \( M_e \) excited molecules (energy per molecule \( \approx h\omega_0 \)), coupled to \( n \) condensate photons (energy per photon \( h\omega_c \)) by interconversion (top). The cavity length controls \( \Delta \), adjusting the ratio between photons and excited molecules (middle). Bottom: When detuning from resonance, the predicted photon number and the response (red) sharply increase; at this point the system starts to minimize its free energy by creating photons instead of maximizing the entropy in the molecular reservoir (see text). At large negative detunings \( \Delta \to -\infty \), where \( \langle n \rangle \to X \), the response gradually falls off to 0. (c) Spectrally resolved streak camera traces showing random arrival times of coherent photons for large \( |\Delta| \) (top), and bunched photons at small \( |\Delta| \) (bottom).
Fig. 2. Number fluctuations in the condensate mode from the canonical to the grand canonical ensemble for a fixed dye-cavity detuning $\Delta = -4.571 k_B T / h$. (a) Second-order correlation functions $g^{(2)}(\tau)$ for decreasing photon numbers $\langle n \rangle$ along with fits (solid). For the largest $\langle n \rangle \approx 9140$ (top panel), the condensate is second-order coherent with $g^{(2)}(0) = 1.02 \pm 0.08$, indicating Poissonian number statistics under canonical conditions. For smaller $\langle n \rangle \approx (330, 26)$, photon bunching in the grand canonical condensate emission is observed with $g^{(2)}(0) = (1.79 \pm 0.20, 1.96 \pm 0.09)$. (b) The zero-delay $g^{(2)}(0)$ as a function of $\langle n \rangle$ shows the crossover of the number statistics from the grand canonical regime with $\sqrt{(\Delta n^2)/(\langle n \rangle)} = 1$ to the canonical one with $\sqrt{(\Delta n^2)/(\langle n \rangle)} = 0$, along with theory for $\langle n \rangle = 1.8 \cdot 10^6$ (solid line). Fluctuation levels are indicated by dashed lines, and error bars are calculated from the uncertainties of the fit parameters. The fit function $f(\tau) = 1 + C_1 e^{\lambda_1 \tau} + C_2 e^{\lambda_2 \tau}$ [39], where $\lambda_1, 2 = -\delta \pm \sqrt{\delta^2 - \omega_0^2}$ and $C_1, 2 = Y \pm i \sqrt{\omega_0^2 - \delta^2} Z$, gives the fit parameters $\delta \approx \{0.38, 0.42, 0.45\}$ ns$^{-1}$, $\omega_0 \approx \{0.43, 0.41, 0.10\}$ ns$^{-1}$, $Y \approx \{0.01, 0.39, 0.48\}$, $Z \approx \{0.15, 5.63, 1.09\}$ for the three cases in (a).

To determine the reactive response function of the condensate, see the right-hand side of eq. (2), requires the derivative $(d\langle n \rangle / d\Delta)_X$ to be evaluated at constant temperature $T$ and excitation number $X = n + M_e$. The molecular part $M_e$ is not directly accessible in our experiments, we must therefore determine $X$ indirectly from the precisely measured $\langle n \rangle$ and $\Delta$. We reconstruct $X$ by fitting $\langle n \rangle$, recorded at different pump powers and dye-cavity detunings, with theory $\sum_n n P_n$ based on eq. (1); here we use only $X$ as a fit parameter, while the molecule number $M$ and $\Delta$ are fixed. Each measurement thus yields four-fold information $\{(g^{(2)}(0), \langle n \rangle, \Delta, X)\}$, from which only the data closest to the target value $X = 1.538 \cdot 10^6$ are retained for further analysis. Note that we have also examined other target values of $X$, but find better statistics at the selected one (see the top panel in Fig. 4(b)). Figure 3(a) shows the obtained first and second moment of the photon number as a function of the detuning for the corresponding, fixed excitation number. For large negative detunings, strongly occupied condensates with suppressed fluctuations indicate the canonical statistical regime. In the opposite limit of detunings closer to resonance, a large fraction of $X$ consists of dye electronic excitations, forming the particle reservoir; here the reduced condensate occupancy with significant fluctuations signals the
onset of grand canonical statistics. The interesting range of detunings, where the fluctuations are significantly varying for constant $X$ (Fig. 3(a), inset), is spectrally narrow and covers only $0.004k_B T / h \approx 25$ GHz, as well understood from the large number of excitations $X$ stored in the system.

Figure 3(b) shows the behavior of the relative effective reservoir size $M_{\text{eff}} / \langle n \rangle^2$, which takes values below to well above unity as the detuning is tuned closer to resonance. The inset of Fig. 3(b) gives the photon chemical potential $\mu = k_B T \ln [(X - \langle n \rangle)/(M - X + \langle n \rangle)] - \hbar \Delta$, which directly depends on $X$ via the ratio between excited and ground state molecules [29, 38]. At large negative detunings, $\mu \approx 0$, as expected from Bose-Einstein statistics. Notably, in the opposite, grand canonical limit, the chemical potential becomes finite; nevertheless, $|\mu| < \hbar \Omega$ indicates that despite fluctuations the system remains condensed.

Next we examine the validity of the fluctuation-dissipation relation in eq. (2) by directly comparing the condensate number fluctuations (see Fig. 3) to the response function for varied detuning. Figure 4(a) shows the resulting squared number fluctuations $\langle \Delta n^2 \rangle$ (red circles) and the scaled response function $-k_B T / \hbar \langle d(n)/d\Delta \rangle_{X,T}$ (blue diamonds) as a function of the detuning $\Delta$, here for fixed $T = 300$ K. The good agreement between both data sets, and with theory (solid line), gives evidence for the fluctuation-dissipation relation to be well fulfilled in our system both in the canonical and grand canonical regime. We note that in the latter case the fluctuation-dissipation relation can be written in terms of the isothermal compressibility $\kappa_T = \langle \mu / d(n) \rangle_{X,T}$ [40]. We use the derivative $\langle d\mu / d(n) \rangle_{X,T} \approx -\hbar \langle (\Delta/d(n)) \rangle_{X,T}$ to rewrite eq. (2) and obtain

$$\langle \Delta n^2 \rangle = \frac{1}{2M_{\text{eff}}} + \frac{1}{k_B T} \left( \frac{1}{M_{\text{eff}}} \right)_{X,T}$$

which for large reservoirs $M_{\text{eff}}$, corresponding to the grand canonical regime, approaches the "textbook" form of the fluctuation-dissipation relation [41]. The data (green squares) in Fig. 4(a) show that the correction term $1/(2M_{\text{eff}})$ can be neglected only deep in the grand canonical regime, while for large negative detunings the compressibility diverges.

Our measurements confirm the thermal nature of the room-temperature Bose-Einstein condensate of photons in a rigorous way. Figure 4(b) shows the deduced temperature $T = -\hbar / k_B \langle \Delta n^2 \rangle / \langle d(n)/d\Delta \rangle_{X,T}$ as a function of the dye-cavity detuning, which agrees with room temperature over the investigated range of detunings, and we find $T = 271(30)$ K. Physically, the results give evidence that independent of the detuning the statistical number fluctuations are driven by thermal energy, with correspondingly varying "stiffness" of the response to perturbations.

In conclusion, we have demonstrated a fluctuation-dissipation relation connecting the statistical number fluctuations of a photon Bose-Einstein condensate coupled to a dye reservoir with the reactive response of the condensate population to variations of the relative energy scale of system and...
reservoir constituents. The ratio between the independently measured fluctuations and response function agrees with thermal energy, confirming the thermalized nature of the optical condensate. In the same breath, the findings also give evidence for the thermal equilibrium character of the molecular reservoir. For the future, probing the dynamical response of the system after a fast perturbation of the reservoir can allow us to extend the studies of the fluctuation-dissipation relation to the time-dependent regime. Other prospects include studies of fluctuations and susceptibilities associated with photon transport, e.g., in lattice or box geometries [31, 40], and their applicability to open quantum systems, which become accessible when tuning thermalization and photon loss [39].

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