



Supplementary Materials for
**Observation of a non-Hermitian phase transition
in an optical quantum gas**

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Materials and Methods

Experimental system

The used experimental apparatus is similar to as described in earlier works, see Refs. 6,14 of the main text for details. Our optical microcavity consists of two ultra-high reflectivity mirrors with reflectivity above 99.998% of $R=1\text{m}$ curvature spaced by a distance of $D_0 \cong 1.4\mu\text{m}$. The cavity is filled with rhodamine dye dissolved in ethylene glycol (concentration: 10^{-3} mol/l). At room temperature, rapid transverse decoherence caused by collisions of solvent molecules with the dye suppresses the formation of polaritons (30,31). Importantly, this also prevents Rabi oscillations of the molecules, meaning that the oscillations of the photon number observed in the oscillatory condensate regime cannot be attributed to a coherent evolution, but are rather due to the weakly dissipative character of the dye microcavity system. The small mirror spacing causes a large frequency spacing between adjacent longitudinal optical modes that is comparable with the emission width of the dye molecules. In this regime we observe that to good accuracy the resonator is only populated with photons of a fixed longitudinal mode, $q=7$ here, and the two transverse modal degrees of freedom make the system two-dimensional. The dye microcavity is pumped with a laser beam of 532 nm wavelength at an angle near 45° with respect to the optical axis. To suppress pumping of the dye into triplet states and excessive heating, the pump pulses are acousto-optically chopped into 500ns long pulses at a 50 Hz repetition rate.

In the cavity the dispersion relation due to the short mirror spacing is modified with respect to free space and acquires quadratic, i.e. massive particle-like, character. The transverse TEM₀₀ mode

becomes the lowest populated eigenmode, which acts as a low-frequency cutoff at energy $\hbar\omega_c = hc/\lambda_c$, where λ_c is the cutoff wavelength. Further, the mirror curvature leads to harmonic confinement of the photon gas, making the photons equivalent to a two-dimensional, harmonically confined gas of massive bosons with effective mass $m_{\text{eff}} = \hbar\omega_c/(c/n)^2$, where c denotes the speed of light and $n \cong 1.43$ is the refractive index of the solvent ethylene glycol. For such a system it is known that a BEC exists at thermal equilibrium conditions (32).

Thermal equilibrium of photons in the cavity is achieved as the photons are absorbed and re-emitted many times by the dye molecules. The used rhodamine dye fulfills the Kennard-Stepanov relation to good accuracy (33). This universal thermodynamic frequency scaling between absorption and emission is well known to apply for systems with rovibrational spectra on top of both lower and upper electronic levels in equilibrium. The conversion of photons into dye electronic excitations and vice versa (right panel of Fig. 1A of the main text) can be seen as an exchange of both energy and particles with the dye which acts as a reservoir in the grand canonical sense. These multiple absorption and re-emission processes induce a thermal spectral distribution of the photon gas at the temperature of the dye rovibrational excitations, which are at room temperature. Given that thermal emission is negligible in the limit of $\hbar\omega_c (\cong 2.1\text{eV}) \gg k_B T (\cong 1/40\text{eV})$, temperature and chemical potentials are independently tunable. This is a striking difference to the usual case of black-body radiation where photons vanish in the system walls upon lowering the temperature instead of exhibiting condensation. In the dye microcavity system, both

photon gas thermalization with a freely adjustable chemical potential and Bose-Einstein condensation has been observed in earlier works, see Refs.11,23,24 for details.

To experimentally monitor the number statistics of the photon condensate, the transmission of one of the microcavity mirrors following a mode filter is directed onto a fast photomultiplier. The mode filter separates the (TEM₀₀) condensate mode from the higher transverse modes forming the thermal cloud by transmission through two optical pinholes acting as a real space and a momentum filter, respectively. In addition, a polarizer is placed into the detection path, removing the polarization degeneracy. The electronic signal of the used photomultiplier (Photek PMT 210) is analyzed with a fast oscilloscope, yielding a correlation signal from the time-resolved photomultiplier signal traces. This signal is used as a measure for the time-dependent second-order coherence $g^{(2)}(\tau)$ of the photon condensate. Experimental data for $g^{(2)}(\tau)$ was recorded in 5 different measurement runs, sampling dye microcavity emission signals acquired within $3 \cdot 10^4$ pump beam pulses each. In each of the measurement runs data for different pump beam powers was recorded so as to vary the condensate size, with calibration of the photon number obtained by simultaneously recording spectra of the dye microcavity emission. The spectra relate the condensate mode population to the photon number in the thermal cloud, the latter equaling the critical number $N_c = \frac{\pi^2}{3} \left(\frac{k_B T}{\hbar \Omega} \right)^2 \cong 80700$ for the used trap frequency $\Omega \cong 2\pi \cdot 40$ GHz. The data used for further analysis are averages of the second-order coherence functions for different mean photon numbers \bar{n} with a number bin size of 250 photons, as shown in the top panels of Fig. 1C for two different

values of \bar{n} . In preparatory measurements with a pulsed laser source, the time resolution of the detection system for measurements of the second-order coherence was determined to be $\cong 500$ ps.

Theoretical model

The dynamics of photons coupled to the dye reservoir can be modeled with the following set of rate equations:

$$\frac{d\langle n \rangle}{dt} = [B_{em} \langle M_e \rangle (\langle n \rangle + 1) - B_{abs} \langle M_g \rangle \langle n \rangle] - \kappa \langle n \rangle, \quad (\text{S1a})$$

$$\frac{d\langle M_e \rangle}{dt} = -[B_{em} \langle M_e \rangle (\langle n \rangle + 1) - B_{abs} \langle M_g \rangle \langle n \rangle] + R_p \langle M_g \rangle, \quad (\text{S1b})$$

where $\langle n \rangle$ denotes the time-dependent expectation value of the number of photons in the condensate mode and $\langle M_g \rangle$ and $\langle M_e \rangle$ describe the time-dependent number of dye molecules in electronic ground and excited states, respectively, with $M = \langle M_e \rangle + \langle M_g \rangle$ as the total molecule number. Further, B_{abs} and B_{em} denote the Einstein coefficients for absorption and emission, respectively, which are related by the Kennard-Stepanov relation, $B_{em}/B_{abs} = \exp(-\hbar(\omega - \omega_{ZPL})/kBT)$, where $\omega_{ZPL} \cong 2\pi c/545\text{nm}$ and $T \cong 300\text{K}$ denote the frequency of the zero-phonon line for rhodamine and the temperature of the dye bath, respectively. Further, κ denotes the photon loss and R_p the pump rate, respectively. In the absence of pumping and loss, with the sum $X = \langle n \rangle + \langle M_e \rangle$ of the photon number and dye molecular electronic excitations being strictly conserved, the dynamics can be described by eq. S1a alone, predicting the fluctuation properties of grand canonical Bose-Einstein condensation, as described in previous work (34-38), see also Ref. 14. For a large relative size of the dye reservoir, that is $M_{\text{eff}} \gg \bar{n}^2$, where $M_{\text{eff}} = M/(2 + 2\cosh((\omega_c - \omega_{ZPL})/kBT))$ ($\cong \bar{M}_e$ for the detuning values chosen here) denotes the effective reservoir size, grand canonical conditions are

fulfilled, and in the condensed state, photon number fluctuations of order of the average particle number emerge. In this limit, the normalized second-order photon coherence function of the condensate reaches $g^{(2)}(0)=2$, i.e., is the same as for a thermal source. On the other hand, when the effective reservoir size is small, for $M_{\text{eff}} \ll \bar{n}^2$, the particle-number fluctuations reduce to a Poissonian level, i.e., they resemble the number statistics of a “usual” canonical ensemble condensate, or of a laser. By varying effective relative size of the reservoir, the second-order coherence of the photon condensate can be tuned between $g^{(2)}(0)=1$ and $g^{(2)}(0)=2$.

The dynamics of fluctuations of the photon number $\Delta n = \langle n \rangle - \bar{n}$ and the total number of excitations $\Delta X = \langle X \rangle - \bar{X}$ of the open system is described by eqs. 1-2 of the main text. These equations are obtained by linearizing around the average values. Remember here that \bar{n} and \bar{X} denote the average values (i.e. averaged over times longer than the correlation time) of the condensate mode photon number and sum of excitations respectively, while $\langle n \rangle$ and $\langle X \rangle$ denote the instantaneous expectation values at time t .

The response to an instantaneous fluctuation of the photon number occurring at a time t_0 around the average value \bar{n} can be written in the form

$$\Delta n(t) = e^{-\delta(t-t_0)} \left(C_1 e^{-\sqrt{\delta^2 - \omega_0^2}(t-t_0)} + C_2 e^{\sqrt{\delta^2 - \omega_0^2}(t-t_0)} + c.c. \right) \text{ where } C_1 \text{ and } C_2 \text{ are constants, with}$$

$$\omega_0 = \sqrt{\kappa(B_{em}(\bar{n}+1) + B_{abs}\bar{n})} \text{ and } \delta = \frac{1}{2}(B_{em}X/\bar{n} + (B_{em} + B_{abs})\bar{n}).$$

For the case of $\bar{n} \gg 1$ and a large negative dye-cavity detuning $\Delta = \omega_c - \omega_{ZPL} \ll -k_B T / \hbar$ relevant here, we have $B_{em} \gg B_{abs}$ and $M_e \gg \bar{n}$, so that the formulas reduce to the simplified forms given in the main text. In our

experiment, we monitor the grand canonical system undergoing thermally (and quantum-mechanically) driven statistical fluctuations under stationary conditions by analyzing the second-order correlation function $g^{(2)}(\tau) = \frac{\overline{n(t) \cdot n(t+\tau)}}{\overline{n(t)} \cdot \overline{n(t+\tau)}}$ of condensate mode photons. The open grand canonical system is reminiscent of a stochastically driven damped harmonic oscillator as readily seen when writing Eq. (1) of the main text as a second order differential equation, yielding the quoted eigenvalues $\lambda_{1,2}$. The expected second-order coherence function of condensate photons can be written as

$$g^{(2)}(\tau) = 1 + e^{-\delta\tau} \left[\left(Y + i\sqrt{\omega_0^2 - \delta^2} Z \right) e^{-\sqrt{\delta^2 - \omega_0^2}\tau} + \left(Y - i\sqrt{\omega_0^2 - \delta^2} Z \right) e^{\sqrt{\delta^2 - \omega_0^2}\tau} \right]. \quad (\text{S2})$$

Other than in the more compact formula given in the main text, the constants introduced in eq. S2 (Y and Z) are real numbers on both sides of the phase transition, which is helpful for the fitting procedure.

Furthermore, from eqs. S1 we can readily determine the steady state values for the photon number \bar{n} and the population of the upper electronic states \bar{M}_e . In the limit of small losses ($\kappa \ll B_{abs} \bar{M}_g$) relevant here, i.e., photons are reabsorbed (and thermalized) faster than they leave the cavity, eq. 1a in the steady state limit yields $\bar{M}_e \cong \bar{M}_g \cdot B_{abs} / B_{em}$. That is, for $B_{em} \gg B_{abs}$ we have $\bar{M}_e \ll \bar{M}_g$ so that $\bar{M}_g \cong M$. Using eq. S1b we furthermore find $\bar{n} \cong R_p M / \kappa$, which expresses that in the open system the average photon number in the cavity is tunable by the pumping strength. Typical experimental numbers for a cutoff wavelength $\lambda_c \cong 571.3$ nm, corresponding to a dye-cavity detuning $\Delta = \omega_c - \omega_{ZPL} \cong -4.1$ $k_B T / \hbar$ for rhodamine dye, are $B_{em} \cong 23.9$ kHz and $B_{abs} \cong 420$ Hz, and a cavity loss $\kappa \cong 2.3$ GHz. For a molecule number $M \cong 5 \cdot 10^9$, we have $\bar{M}_e \cong 8.8 \cdot 10^7$. For typical

condensate photon numbers, which are up to $\cong 5 \cdot 10^5$, the limits given above are fulfilled with good accuracy.

The number statistics of the photon condensate is characterized by the crossing of two eigenvalues of the matrix A (eqs. (1-2) in the main text) characterizing the fluctuations in the system. Due to the driven-dissipative nature of the system, the matrix $A=A(s)$ describing the two-component system of photons coupled to the dye reservoir is non-Hermitian. Here s denotes a set of system parameters; relevant to the described measurements are the average photon number \bar{n} , which experimentally is controlled by the pump power, the cutoff wavelength, and the dye concentration, and we have $\delta(s)$ and $\omega_0(s)$ damping rate and oscillation frequency respectively. The eigenvalues of the non-Hermitian 2×2 matrix $A(s)$ can be either both real or both complex: Two real eigenvalues mean biexponential damping of the fluctuations, while complex eigenvalues mean damped oscillatory behavior. A crossing of the two eigenvalues occurs in the complex plane for a certain value of the parameters $s=SEP$, see also the discussion in the main text. This is the exceptional point where the two eigenvalues coalesce. The gap between the eigenvalues λ_1 and λ_2 of the non-Hermitian matrix $A(s)$ opens in the real or the imaginary plane on different sides of the exceptional point.

At the exceptional point, small changes of s can lead to qualitative changes of the state one is in, i.e. biexponential or oscillatory decaying respectively. Critical behavior of common (equilibrium) phase transitions is here rather replaced by the sensitivity of the phase one is in to changes in the control parameter s , which becomes large near the exceptional point at $\delta=\omega_0$, which we understand

as the point of the phase transition. On the other hand, the condensate will remain in the biexponential or the oscillatory phase both for $\delta \gg \omega_0$ and for $\delta \ll \omega_0$ respectively upon small changes in s , i.e. when being deep in one of the corresponding phases.

As discussed in the main text, Fig. 2B represents the expected phase diagram in a three-dimensional plot, showing the biexponential and the oscillatory condensate phases and, for large losses (near $\kappa \cong B_{abs} M$, see Ref. 28), the crossover to lasing. One may write the condition $\omega_0 = \delta$

for the position of the exceptional point between the two dissipative phases in the form

$$\kappa / B_{em} \sqrt{M_e} = \frac{1}{4} \left[\left(\frac{\sqrt{M_e}}{\bar{n}} \right)^{3/2} + \left(\frac{\bar{n}}{\sqrt{M_e}} \right)^{1/2} \right].$$

When expressed in terms of the scaled parameters $\kappa' := \kappa / B_{em} \sqrt{M_e}$ (scaled loss rate) and $\alpha := \bar{n} / \sqrt{M_e}$ (scaled photon number), this formula

reduces to the universal form $\kappa' = \frac{1}{4} \left[\frac{1}{\alpha^{3/2}} + \alpha^{1/2} \right]$. The expected phase diagram thus collapses to

the two-dimensional form used in Fig. 4A of the main text. In terms of the scaled variables, the theory values for the undamped oscillation frequency and damping constant can be expressed as

$$\omega_0 / B_{em} \sqrt{M_e} \cong \sqrt{\kappa' \alpha} \quad \text{and} \quad \delta / B_{em} \sqrt{M_e} \cong \frac{1}{2} \left(\frac{1}{\alpha} + \alpha \right).$$

Data analysis

The experimental data for the second-order coherence function has been fitted with the theory prediction of eq. S2, which is valid on both sides of the phase transition, using ω_0 , δ , Y , and Z (all being real numbers) as fit parameters. Depending on the difference $\delta - \omega_0$ being positive or negative, the resulting second-order coherence function is biexponential or oscillatory, respectively. Experimental data for the difference $\delta - \omega_0$, along with corresponding uncertainties, is shown in Fig. 3A of the main text for different average photon numbers for one of the five recorded

data sets, (see Table S1). The black lines in the top panels of Fig. 1C of the main text show the fit results for two individual correlation data measurements.

In the oscillatory phase ($\omega_0 > \delta$) the second-order coherence of eq. S2 takes the explicit form $g^{(2)}(\tau) = 1 + e^{-\tau/\tau_c} (C_1' \sin(\Omega_0 \tau) + C_2' \cos(\Omega_0 \tau))$, with a decay time $\tau_c = 1/\delta$ and an oscillation frequency $\Omega_0 = \sqrt{\omega_0^2 - \delta^2}$, while in the biexponential phase ($\omega_0 < \delta$) it takes the form $g^{(2)}(\tau) = 1 + (C_1'' e^{-\tau/\tau_{c1}} + C_2'' e^{-\tau/\tau_{c2}})$, with the two decay rates $1/\tau_{c1,2} = \delta \pm \sqrt{\delta^2 - \omega_0^2}$. Here, the constants C_1', C_2', C_1'' and C_2'' here again are real numbers.

Figures 3B,C of the main text show the variation of the observed decay times τ_{ci} and, for the case of the oscillatory phase, the oscillation frequency Ω_0 on the average photon number (data set #1). For a fitting of such data, the expected variation of the undamped oscillation frequency $\omega_0 \cong \sqrt{\kappa B_{em} \bar{n}}$ and damping constant $\delta \cong \frac{1}{2}(B_{abs} M / \bar{n} + B_{em} \bar{n})$ on the average photon number \bar{n} was assumed, where the molecule number M and the loss κ are free fit parameters. For the form of the damping constant δ given here, we have used the relation $\bar{M}_e \cong M \cdot B_{abs} / B_{em}$ for the population of the upper electronic state of the dye molecules. The Einstein coefficients B_{abs} and B_{em} for the corresponding wavelength have been determined by extracting the coefficient B_{abs} from absorption measurements in an independent spectroscopic measurement performed on a macroscopic dye cuvette (37), and determining B_{em} for the corresponding wavelength using the Kennard-Stepanov relation $B_{em} = B_{abs} \exp(-\hbar(\omega - \omega_{ZPL})/kBT)$.

The data points in Fig. 4A of the main text summarize the results of correlation data recorded in all five measurement data sets, and Extended Data Table 1 gives the obtained fit results. In Fig. 4 of the main text, the experimental data is plotted for the scaled variables described in the previous section, allowing to combine results recorded at different cavity cutoff wavelengths in the single diagrams shown. Note that, given that both δ and ω_0 are proportional to $B_{em} \sqrt{M_e}$, this scaling also holds for the oscillation frequency Ω_0 and the inverse damping rates $1/\tau_{ci}$.

Supplementary Text

Microscopic derivation of the rate equations

The derivation of the rate equations S1 is detailed in Ref. 6. Briefly, the rate equations describing the dynamics of the photon condensate can be obtained via a Lindblad master equation that follows from a microscopic quantum-mechanical model (6,39,40) describing the dye molecules, their vibrational modes or phonons, and the coupling to the photon modes of the cavity. Due to fast collisions of the dye with solvent molecules, the phonon excitations may be considered to be in thermal equilibrium at ambient temperature and treated as a reservoir to be integrated out. The Hamiltonian (also known as the Tavis-Cummings-Holstein model) for a collection of M molecules and multiple cavity modes reads

$$H = \sum_k (\omega_k - \omega_{ZPL}) a_k^\dagger a_k + \sum_{m=1}^M \left[\Omega b_m^\dagger b_m + \Omega \sqrt{S} \sigma_m^z (b_m + b_m^\dagger) + g \sum_k (a_k \sigma_m^+ + a_k^\dagger \sigma_m^-) \right], \quad (\text{S3})$$

where the cavity modes, a_k, a_k^\dagger , have dispersion ω_k , and the zero-phonon line of the dye molecules is defined by ω_{ZPL} . σ_m^z and σ_m^\pm are the Pauli z-matrix and the raising/lowering operators in the two-

dimensional space of electronic ground and excited states of dye molecule m , respectively. The vibrational frequency is Ω , and S describes the coupling of the phononic oscillator potential to the electronic transition. Since the phonon position operators are $\hat{x}_m \sim (b_m + b_m^\dagger)$, we understand from $\sigma_m^z = \sigma_m^+ \sigma_m^- - \sigma_m^- \sigma_m^+$ that the effect of this term is a displacement of the center of the phononic oscillator, with the sign depending on whether the molecule is in the electronic ground or excited state. Finally, a Jaynes-Cummings coupling between photons and dye is included via the small parameter g .

The molecular part of the Hamiltonian can be diagonalized by means of a polaron transformation, which leads to an effective, nonlinear coupling between photons and molecules, mediated by the phonon excitations of the dye. It is then possible to bring H into a form suitable for a perturbative expansion in g while treating the molecular vibrations as a reservoir. After expanding to second order in g , and discarding inter-molecular correlations as well as the coherent, first-order contribution (6), by making the well-known Born-Markov and secular approximations, one arrives at the master equation (6,27,40).

$$\begin{aligned}
\dot{\rho}(t) = & -i[H_S, \rho(t)] + \sum_k \frac{\kappa}{2} \mathcal{L}[a_k] \rho(t) + \sum_{m=1}^M \left[\frac{R_p}{2} \mathcal{L}[\sigma_m^+] + \frac{\gamma}{2} \mathcal{L}[\sigma_m^-] \right] \rho(t) \\
& + \sum_k \sum_{m=1}^M \left[\frac{\Gamma_k^-}{2} \mathcal{L}[a_k^\dagger \sigma_m^-] + \frac{\Gamma_k^+}{2} \mathcal{L}[a_k \sigma_m^+] \right] \rho(t),
\end{aligned} \tag{S4}$$

where we have also included the photon resonator loss κ , the external pump rate R_p , and the imperfect quantum efficiency γ of the molecules. The frequency-dependent, incoherent coupling coefficients $\Gamma_k^\pm = 2\text{Re}K(\pm[\omega_k - \omega_{\text{ZPL}}])$, which give rise to the photon absorption and emission processes, respectively, are defined as

$$K(\omega) = g^2 \int_0^\infty d\tau e^{-(R_p + \gamma)\tau/2} e^{i\omega\tau} \left(\exp \left\{ -4S \left[(1 - \cos \Omega\tau) \coth \frac{\beta\Omega}{2} + i \sin \Omega\tau \right] \right\} - \exp \left\{ -4S \coth \frac{\beta\Omega}{2} \right\} \right). \tag{S5}$$

Note that for the weak external pumping employed in our experiments, the influence of the rapid relaxation of the reservoir on the broadening of the molecular spectrum described by $K(\omega)$ is much larger than any additional broadening due to R_p and γ . It is, therefore, valid to assume that Γ_k^\pm remain constant over the experimentally explored range of the external pumping strength. Alternatively, a quantum Langevin equation can be derived at the level of the master equation (S4).

The rate equations (cf. Eqs. 1) can now be obtained from the master equation (S4) by calculating the equations of motion for the time-dependent expectation values of the photon number in cavity mode m , $\langle n_k \rangle(t)$, and the number of excited molecules, $\langle M_e \rangle(t)$. In terms of operators these equations read,

$$\partial_t \langle a_k^\dagger a_k \rangle = -\kappa \langle a_k^\dagger a_k \rangle - \frac{1}{2} \sum_{m=1}^M \left[\Gamma_k^+ \langle a_k^\dagger a_k (1 - \sigma_m^z) \rangle - \Gamma_k^- \langle a_k a_k^\dagger (1 + \sigma_m^z) \rangle \right], \quad (\text{S6})$$

$$\partial_t \langle \sigma_m^z \rangle = R_p (1 - \langle \sigma_m^z \rangle) - \gamma (1 + \langle \sigma_m^z \rangle) + \sum_k \left[\Gamma_k^+ \langle a_k^\dagger a_k (1 - \sigma_m^z) \rangle - \Gamma_k^- \langle a_k a_k^\dagger (1 + \sigma_m^z) \rangle \right], \quad (\text{S7})$$

where the average is $\langle A \rangle (t) = \text{Tr}\{A\rho(t)\}$. Assuming that the molecules are all identical, one can drop the molecular index and replace the sum over the M molecules in Eq. (S6) by a factor of M .

Summing over m also in Eq. (S7), we arrive at

$$\partial_t \langle n_k \rangle = -\kappa \langle n_k \rangle - \Gamma_k^+ \langle n_k (M - M_e) \rangle + \Gamma_k^- \langle (n_k + 1) M_e \rangle, \quad (\text{S8})$$

$$\partial_t \langle M_e \rangle = R_p (M - \langle M_e \rangle) - \gamma \langle M_e \rangle + \sum_k \left[\Gamma_k^+ \langle n_k (M - M_e) \rangle - \Gamma_k^- \langle (n_k + 1) M_e \rangle \right]. \quad (\text{S9})$$

A further simplification arises from the fact that for large dye reservoir, the occupation of a given cavity mode m and the total molecule excitation number are uncorrelated, i.e. their correlation factorizes,

$$\langle n_k M_e \rangle \approx \langle n_k \rangle \langle M_e \rangle. \quad (\text{S10})$$

This closes the set of equations S8. For a single photon mode $\langle n \rangle = \langle a_0^\dagger a_0 \rangle$, and with the ground-mode emission and absorption coefficients

$$\begin{aligned} B_{\text{em}} &= \Gamma_0^-, \\ B_{\text{abs}} &= \Gamma_0^+, \end{aligned} \quad (\text{S11})$$

this represents the rate equations 1 and completes the derivation of the rate equations from the microscopic quantum mechanical model. Two further remarks are at place here. First, observe that

the total excitation number $\langle X \rangle := \langle n \rangle + \langle M_e \rangle$, conserved without external driving or loss, obeys the much simpler equation

$$\begin{aligned}\partial_t \langle X \rangle &= -\kappa \langle n \rangle + R_p (M - \langle M_e \rangle) - \gamma \langle M_e \rangle \\ &= -(\kappa - (R_p + \gamma)) \langle n \rangle - (R_p + \gamma) \langle X \rangle + R_p M,\end{aligned}\tag{S12}$$

where the emission and absorption terms present in Eqs. (S8) and (S9) have canceled and only drive and loss terms remain. The fact that $\partial_t \langle X \rangle \neq 0$ distinguishes the driven-dissipative photon-dye system from a perfectly closed system and is ultimately responsible for the oscillatory second-order coherence dynamics. Second, in the closed limit $\kappa, R_p, \gamma \rightarrow 0$, the rate equations predict a steady-state Bose-Einstein distribution in the long-time limit (41)

$$\frac{\langle n_k + 1 \rangle}{\langle n_k \rangle} = \frac{\Gamma_k^+ \langle M - M_e \rangle}{\Gamma_k^- \langle M_e \rangle} = e^{\beta(\omega_k - \omega_{ZPL})} \frac{\sum_l \Gamma_l^- \langle n_l + 1 \rangle}{\sum_l \Gamma_l^+ \langle n_l \rangle},\tag{S13}$$

where we have used Eq. (S.6) for the first, and Eq. (S.7) and the Kennard-Stepanov relation $\Gamma_k^+ / \Gamma_k^- = e^{\beta(\omega_k - \omega_{ZPL})}$ for the second equality. The first equality is important for the description of the phase diagram, as we will see in the following note.

Fluctuations around the steady state

Here we address the derivation of the dynamics of the photon-number fluctuations Δn and the excitation-number fluctuations ΔX , as described by Eqs. (1) and (2) in the main text. Defining

the deviations from the steady state as $\Delta n(t) := \langle n \rangle(t) - \bar{n}$, $\Delta M_e(t) := \langle M_e \rangle(t) - \overline{M_e}$ and $\Delta X := \langle X \rangle(t) - \bar{X}$, the photon-number fluctuations obey the regression law

$$\begin{aligned} \partial_t \Delta n &= (-\kappa - B_{\text{abs}}(M - \overline{M_e}) + B_{\text{em}} \overline{M_e}) \Delta n + (B_{\text{abs}} \bar{n} + B_{\text{em}}(\bar{n} + 1)) \Delta M_e \\ &\approx -(B_{\text{abs}}(M - \overline{M_e}) - B_{\text{em}} \overline{M_e} + \omega_0^2/\kappa) \Delta n + (\omega_0^2/\kappa) \Delta X, \end{aligned} \quad (\text{S14})$$

where we have introduced $\omega_0^2/\kappa = B_{\text{abs}} \bar{n} + B_{\text{em}}(\bar{n} + 1) \approx B_{\text{em}} \bar{n}$. The excitation-number fluctuations follow analogously,

$$\partial_t \Delta X = -(\kappa - (R_p + \gamma)) \Delta n - (R_p + \gamma) \Delta X \approx -\kappa \Delta n. \quad (\text{S15})$$

Taking the equilibrium distribution from (S13), we can write

$$B_{\text{abs}}(M - \overline{M_e}) = B_{\text{em}} \overline{M_e} + B_{\text{em}} \frac{\overline{M_e}}{\bar{n}}, \quad (\text{S16})$$

from which we find

$$\partial_t \Delta n \approx -2\delta \Delta n + (\omega_0^2/\kappa) \Delta X, \quad (\text{S17})$$

with $2\delta = B_{\text{em}} \overline{M_e}/\bar{n} + \omega_0^2/\kappa$. As stated in the main text, Eq. (2), we therefore have in matrix form,

$$\partial_t \begin{pmatrix} \Delta n \\ \Delta X \end{pmatrix} = \begin{pmatrix} -2\delta & \omega_0^2/\kappa \\ -\kappa & 0 \end{pmatrix} \begin{pmatrix} \Delta n \\ \Delta X \end{pmatrix}. \quad (\text{S18})$$

The eigenvalues of this non-Hermitian matrix determine the behaviour of the second-order coherence function via the quantum regression theorem, which in this case states that the density-density correlations of spontaneous fluctuations obey the same regression law as the relaxation

towards the steady state after a small perturbation, as given by Eq. (S18). For completeness, a detailed proof of this statement is given in the subsequent section.

Second order coherence dynamics

Here we give the formal proof that Eq. (S18) also applies to the dynamics of the second-order correlation function. The time-dependent photon density-density or second-order correlation function is defined as (41)

$$g^{(2)}(\tau) = \frac{\langle n(t+\tau)n(t) \rangle}{\langle n(t) \rangle^2} \Big|_{t \rightarrow \infty} = \frac{\text{Tr} \left[a^\dagger a e^{\hat{\mathcal{L}}\tau} \tilde{\rho}_\infty \right]}{\text{Tr} [a^\dagger a \rho_\infty]^2}, \quad (\text{S19})$$

where $\hat{\mathcal{L}}$ is the total superoperator defined by the master equation (S4), the steady-state density matrix is defined as $\rho_\infty = \lim_{t \rightarrow \infty} \rho(t)$, and we introduced an effective density operator defined by $\tilde{\rho}_\infty := a^\dagger a \rho_\infty$. We note in passing that, for the normal-ordered second-order correlation function, one would need to set $\tilde{\rho}_\infty = a \rho_\infty a^\dagger$. Defining also the corresponding effective average $\langle\langle A \rangle\rangle(\tau) := \text{Tr} A e^{\hat{\mathcal{L}}\tau} \tilde{\rho}_\infty$, the second-order coherence becomes

$$g^{(2)}(\tau) = \frac{\langle\langle n \rangle\rangle(\tau)}{\bar{n}^2}. \quad (\text{S20})$$

The effective averages $\langle\langle n \rangle\rangle$ and $\langle\langle M_e \rangle\rangle$ obey almost the same equations as $\langle n \rangle$ and $\langle M_e \rangle$, the only difference arising from the fact that the trace of the effective density matrix does not vanish, $\text{Tr} \tilde{\rho}_\infty$

$= \bar{n}$, which gives $R_p M \text{Tr}[\sigma^- \sigma^+ \tilde{\rho}_\infty] = R_p (\bar{n} M - \langle \langle M_e \rangle \rangle)$. Thus, one finds equations of motion formally analogous to the rate equations,

$$\partial_\tau \langle \langle n \rangle \rangle = -\kappa \langle \langle n \rangle \rangle - B_{\text{abs}} \langle \langle n(M - M_e) \rangle \rangle + B_{\text{em}} \langle \langle (n+1)M_e \rangle \rangle, \quad (\text{S21})$$

$$\partial_\tau \langle \langle M_e \rangle \rangle = R_p (\bar{n} M - \langle \langle M_e \rangle \rangle) - \gamma \langle \langle M_e \rangle \rangle + B_{\text{abs}} \langle \langle n(M - M_e) \rangle \rangle - B_{\text{em}} \langle \langle (n+1)M_e \rangle \rangle. \quad (\text{S22})$$

It is important to note that the form of these equations is independent of the operator ordering in the definition of $g^{(2)}(\tau)$ in Eq. (S19). Their structure is determined by $\hat{\mathcal{L}}$ rather than the *per se* arbitrary effective density matrix. What does change are the initial conditions. This, however, is negligible for the relatively large photon numbers relevant here, with the difference being on the order of $1/\bar{n}$. The relation between the rate equations and (S21), (S22) may also be understood as the density-matrix ansatz. If ρ_∞ is represented as a diagonal matrix with elements $P_{nM_e}^\infty$, then $\tilde{\rho}_\infty$ will also be diagonal with elements $\tilde{P}_{nM_e}^\infty(\tau=0) = nP_{nM_e}^\infty$. These provide the initial conditions for a system of equations formally identical to that for P_{nM_e} .

To truncate and close the hierarchy of expectation values in Eqs. (S.19) and (S.20), the higher-order moments $\langle \langle M_e \rangle \rangle$ need to be factorized, which can be done using a Gaussian identity for P_{nM_e} applied to the steady-state expectation values, i.e.

$$\overline{n^2 M_e} = 2\bar{n}\overline{nM_e} + \overline{M_e} \overline{n^2} - 2\bar{n}^2 \overline{M_e}, \quad (\text{S23})$$

where the average is

$$\bar{A} = \sum_{n=0}^{\infty} \sum_{m=0}^M AP_{nM_e}^{\infty}. \quad (\text{S24})$$

However, since

$$\overline{nA} = \sum_{n=0}^{\infty} \sum_{m=0}^M nAP_{nM_e}^{\infty} = \sum_{n=1}^{\infty} \sum_{m=0}^M A\tilde{P}_{nM_e}^{\infty} = \langle\langle A \rangle\rangle, \quad (\text{S25})$$

Eq. (S23) also implies,

$$\langle\langle nM_e \rangle\rangle = \bar{n}\langle\langle M_e \rangle\rangle + \bar{n}\overline{nM_e} + \overline{M_e}\langle\langle n \rangle\rangle - 2\bar{n}^2\overline{M_e}. \quad (\text{S26})$$

Introducing furthermore the vector

$$\mathbf{g}(\tau) = \begin{pmatrix} \Delta g_n^{(2)} \\ \Delta g_{n, M_e}^{(2)} \end{pmatrix} = \begin{pmatrix} \langle\langle n \rangle\rangle - \bar{n}^2 \\ \langle\langle M_e \rangle\rangle - \bar{n}\overline{M_e} \end{pmatrix}, \quad (\text{S27})$$

Eqs. (S21) and (S22) become

$$\begin{aligned} \partial_{\tau} \Delta g_n^{(2)} = & \bar{n} \left\{ -\kappa\bar{n} - B_{\text{abs}}\overline{(M - M_e)} + B_{\text{em}}\overline{(n + 1)M_e} \right\} \\ & - \kappa\Delta g_n^{(2)} - B_{\text{abs}} \left[(M - \overline{M_e})\Delta g_n^{(2)} - \bar{n}\Delta g_{n, M_e}^{(2)} \right] + B_{\text{em}} \left[(\bar{n} + 1)\Delta g_{n, M_e}^{(2)} + \overline{M_e}\Delta g_n^{(2)} \right], \end{aligned} \quad (\text{S28a})$$

$$\begin{aligned} \partial_{\tau} \Delta g_{n, M_e}^{(2)} = & \bar{n} \left\{ R_p(M - \overline{M_e}) - \gamma\overline{M_e} + B_{\text{abs}}\overline{(M - M_e)} - B_{\text{em}}\overline{(n + 1)M_e} \right\} \\ & - (R_p + \gamma)\Delta g_{n, M_e}^{(2)} + B_{\text{abs}} \left[(M - \overline{M_e})\Delta g_n^{(2)} - \bar{n}\Delta g_{n, M_e}^{(2)} \right] - B_{\text{em}} \left[(\bar{n} + 1)\Delta g_{n, M_e}^{(2)} + \overline{M_e}\Delta g_n^{(2)} \right]. \end{aligned} \quad (\text{S28b})$$

Using the steady-state solutions of the rate equations to eliminate the curly brackets, one arrives at the linear system

$$\partial_{\tau} \mathbf{g}(\tau) = \begin{pmatrix} -\kappa - B_{\text{abs}}(M - \overline{M_e}) + B_{\text{em}}\overline{M_e} & \omega_0^2/\kappa \\ B_{\text{abs}}(M - \overline{M_e}) - B_{\text{em}}\overline{M_e} & -(R_p + \gamma) - \omega_0^2/\kappa \end{pmatrix} \mathbf{g}(\tau). \quad (\text{S29})$$

Under the various simplifying assumptions made in the main text ($B_{\text{em}} \gg B_{\text{abs}}$, validity of Eq. S13), this general set of equations is equivalent to Eq. (S18). In this way, we have formally rederived that, for the assumptions made, the steady-state density-density correlations indeed follow the same dynamics as small fluctuations around the steady state.

Data set	$\lambda_c(\text{nm})$	$B_{\text{abs}}(\text{s}^{-1})$	$\kappa (\text{ns}^{-1})$	M
1	571.3	420	2.2(2)	$4.76(3) \cdot 10^9$
2	570.4	490	1.7(1)	$6.53(1) \cdot 10^9$
3	570.4	490	2.6(1)	$4.08(14) \cdot 10^9$
4	570.4	490	2.5(1)	$2.04(3) \cdot 10^9$
5	575	219	2.7(3)	$1.83(2) \cdot 10^9$

Table S1: Summary of experimental parameters and fit results for the five different data sets, recorded with different cavity cutoff wavelengths λ_c (see second column) and dye concentrations. Both cavity loss κ and dye molecular number M are fit results (final two columns). The Einstein coefficient for emission B_{em} can be determined from the quoted Einstein coefficient for absorption B_{abs} for the corresponding wavelength using the Kennard–Stepanov relation.

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