Supporting Information:

Sustained photon pulse revivals from inhomogeneously broadened spin ensembles

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Supplementary Note 1. Volterra equation for the cavity amplitude

Our starting point is the Hamiltonian (1) of the main article from which we derive the equations for the cavity and spin operators, $\dot{a} = i[\mathscr{H}, a]$, $\dot{\sigma}_k^{(\mu)(-)} = i[\mathscr{H}, \dot{\sigma}_k^{(\mu)(-)}]$, respectively. Here a stands for the cavity operator and $\sigma_k^{(\mu)(-)}$ are standard Pauli operators associated with the k-th spin residing in the μ-th ensemble. (All notations are in tact with those introduced in the main article.) During the derivations we use the following simplifications and approximations valid for various experimental realizations: (i) $kT \ll \hbar\omega_c$ (the energy of photons of the external bath is substantially smaller than that of cavity photons); (ii) the number of microwave photons in the cavity remains small as compared to the total number of spins participating in the coupling (limit of low input powers of an incoming signal), so that the Holstein-Primakoff-approximation, $\langle \sigma_k^{(\mu)(z)} \rangle \approx -1$, always holds; (iii) the effective collective coupling strength of each spin ensemble, $\Omega_\mu^2 = \sum_{k=1}^{N_\mu} g_k^{(\mu)2}$, satisfies to the inequality $\Omega_\mu \ll \omega_c$, justifying the rotating-wave approximation; (iv) the spatial size of the spin ensembles is sufficiently smaller than the wavelength of a cavity mode. Having introduced all these assumptions, we derive the following system of coupled first-order linear ordinary operator equations for the cavity and spin operators in $\omega_p = \omega_c$-rotating frame

$$\dot{a}(t) = -\kappa \cdot a(t) + \sum_{\mu=1}^{M} \sum_{k=1}^{N_\mu} g_k^{(\mu)} \sigma_k^{(\mu)(-)}(t) - \eta(t), \tag{1a}$$

$$\dot{\sigma}_k^{(\mu)(-)}(t) = -\left[\gamma + i(\omega_k^{(\mu)} - \omega_c)\right] \sigma_k^{(\mu)(-)}(t) - g_k^{(\mu)} a(t), \tag{1b}$$

where κ and γ are the total dissipative cavity and individual spin losses. By formally integrating the equations (1b) for the spin operators and inserting them into Eq. (1a) for the cavity operator, we get

$$\dot{a}(t) = -\kappa \cdot a(t) + \sum_{\mu=1}^{M} \sum_{k=1}^{N_\mu} g_k^{(\mu)} \sigma_k^{(\mu)(-)}(0) e^{-i\left(\omega_k^{(\mu)} - \omega_c - i\gamma\right)t} - \Omega^2 \int_0^\infty d\omega F(\omega) \int_0^t d\tau e^{-i(\omega - \omega_c - i\gamma)(t-\tau)} a(\tau) - \eta(t),$$

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where $\sigma_k^{(\mu)(-)}(0)$ is the initial spin operator and $F(\omega)$ stands for the total spectral function which is defined as a sum over the spectral densities of each spin ensemble, $F(\omega) = \sum_{\mu=1}^{M} \Omega_\mu^2/\Omega^2 \cdot \rho_\mu(\omega)$. Here $\rho_\mu(\omega) = \sum_{k=1}^{N_\mu} g_k^{(\mu)2}\delta(\omega-\omega_k^{(i)})/\Omega_i^2$ describes the spin spectral density of the μ-th ensemble, $\Omega_\mu = (\sum_{k=1}^{N_\mu} g_k^{(\mu)2})^{1/2}$ is its effective collective coupling strength to the cavity mode and Ω stands for the coupling strength of the central ensemble.

We then treat the problem semiclassically by introducing the cavity and spin expectation values, $A(t) = \langle a(t)\rangle$ and $B_k^{(\mu)}(t) = \langle \sigma_k^{(\mu)(-)}(t)\rangle$. For the sake of simplicity we consider the case when all spins are initially in the ground state, $B_k^{(\mu)}(0) = 0$, so that Eq. (2) reduces to the closed Volterra integro-differential equation for the cavity amplitude

$$\dot{A}(t) = -\kappa \cdot A(t) - \Omega^2 \int_0^\infty d\omega F(\omega) \int_0^t d\tau e^{-i(\omega-\omega_c-i\gamma)(t-\tau)} A(\tau) - \eta(t). \tag{2}$$

Next we formally integrate Eq. (2) in time and simplify the resulting double integral on the right-hand side by means of the partial integration method. Assuming that the cavity is initially empty, $A(0) = 0$, we finally derive the following Volterra integral equation for the cavity amplitude

$$A(t) = \int_0^t d\tau \mathscr{K}(t-\tau) A(\tau) + \mathscr{D}(t), \tag{3}$$

where $\mathscr{K}(t-\tau)$ is the kernel function

$$\mathscr{K}(t-\tau) = \Omega^2 \int d\omega \frac{F(\omega)\left[e^{-i(\omega-\omega_c-i(\gamma-\kappa))(t-\tau)} - 1\right]}{i(\omega-\omega_c-i(\gamma-\kappa))} \cdot e^{-\kappa(t-\tau)}, \tag{4}$$

and the function $\mathscr{D}(t)$ is given by

$$\mathscr{D}(t) = \int_0^t d\tau\, \eta(\tau) \cdot e^{-\kappa(t-\tau)}. \tag{5}$$

We solve then Eq. (3) numerically using the methods described in details in our recent publications [1, 2].

Supplementary Note 2. Single-photon dynamics

Here we prove that the probability for a single photon, which is populating the cavity at time $t = 0$, to stay inside the cavity at $t > 0$, reduces to $N(t) = |A(t)|^2$, where $A(t)$ is the solution of the Volterra equation (2) with the initial condition $A(0) = 1$ and $\eta(t) = 0$. By definition, this probability is nothing more than the expectation value of the number operator $\mathscr{N} = a^\dagger(t)a(t)$, i.e. $N(t) = \langle 1,\downarrow| a^\dagger(t)a(t)|1,\downarrow\rangle$. Taking into account that we deal with a single excitation in the system, we make use of the following closure relation,

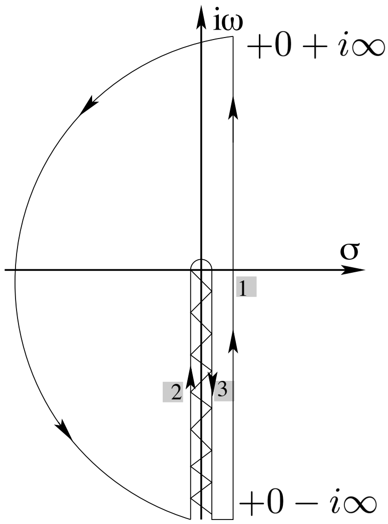
$$\mathbb{1} = |0,\downarrow\rangle\langle 0,\downarrow| + \sum_l |0,\uparrow_l\rangle\langle 0,\uparrow_l| + |1,\downarrow\rangle\langle 1,\downarrow| + \sum_l |1,\uparrow_l\rangle\langle 1,\uparrow_l|, \tag{6}$$

where for the sake of notational simplicity the index l enumerates all spins independently of the spin ensemble to which they belong to. We derive the following expression for $N(t)$

$$N(t) \equiv \langle 1,\downarrow| a^\dagger(t)\mathbb{1}a(t)|1,\downarrow\rangle = \tag{7}$$

$$|\langle 0,\downarrow|a(t)|1,\downarrow\rangle|^2 + |\langle 1,\downarrow|a(t)|1,\downarrow\rangle|^2 + \sum_l |\langle 0,\uparrow_l|a(t)|1,\downarrow\rangle|^2 + \sum_l |\langle 1,\uparrow_l|a(t)|1,\downarrow\rangle|^2.$$

We then let the operator equations (1a, 1b) from Supplementary Note 1 act on the bra- and ket-vectors which show up in Eq. (7) and derive four independent sets of coupled ODEs for the corresponding expectation values $\langle a(t)\rangle$ and $\langle \sigma_j^-(t)\rangle$.



S 1 Contour completion in the complex plane $s=\sigma+i\omega$ for the calculation of the inverse Laplace transform. Those contours which give nonzero contribution are designated by numbers. The zig-zag line corresponds to the branch cut along the negative part of the imaginary axis.

Remarkably, these sets of equations look formally the same being independent of the specific bra- or ket-vector appearing on the left or right side in these operator equations i.e. they evolve in the same fashion as the corresponding operators themselves. The only difference between the resulting solutions for the expectation values appearing in Eq. (7) stems from the initial conditions which are nonzero only for the first term in the r.h.s. of Eq. (7), namely $A(0)=\langle 0,\downarrow|a(0)|1,\downarrow\rangle=1$. For all other terms the resulting expectation values are zero at $t=0$, and as a consequence, they remain zero at $t>0$ as well. Therefore, the probability for a photon to reside in the cavity at $t>0$ reduces to $N(t)=|\langle 0,\downarrow|a(t)|1,\downarrow\rangle|^2=|A(t)|^2$, where $A(t)$ is exactly given as the solution of the Volterra equation (2) in Supplementary Note 1 with the initial conditions $A(0)=1$ and $B_l(0)=0$.

Supplementary Note 3. Laplace transform of the Volterra equation

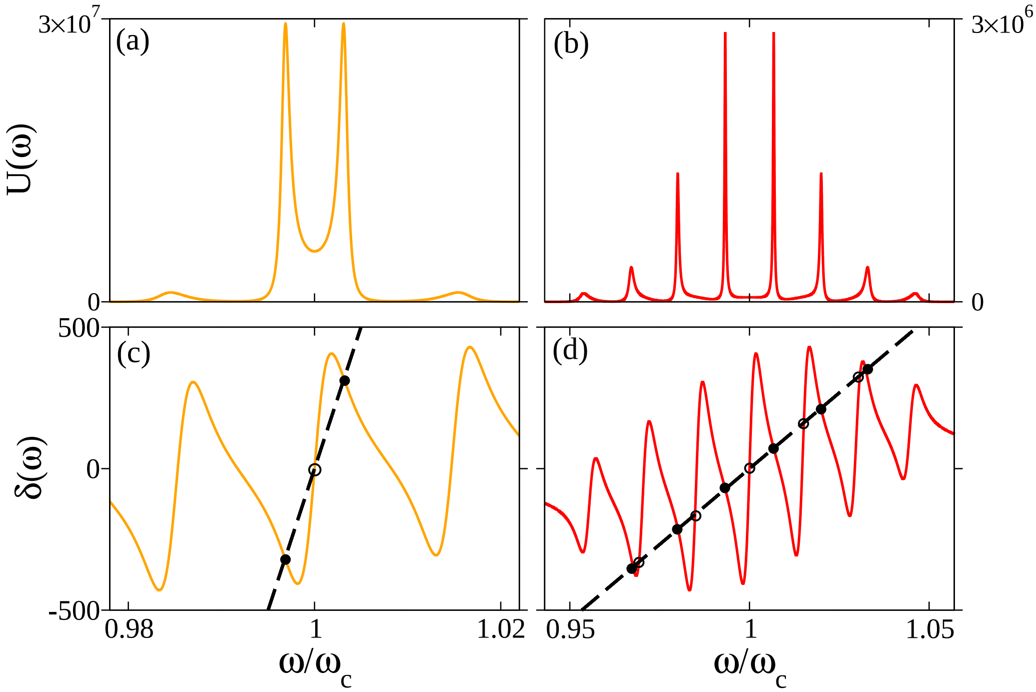
Here we sketch the derivation of the Laplace transformation of the Volterra equation (2) from Supplementary Note 1 assuming that all spins are initially in the ground state and the cavity mode a contains initially a single photon, $A(0)=1$ (the case considered in Supplementary Note 2). For that purpose we multiply Eq. (2) by e^{-st} ($s=\sigma+i\omega$ is the complex variable), integrate both sides of the equation with respect to time and finally obtain the following expression for the Laplace transform:

$$\tilde{A}(s)=\frac{1}{s+\kappa-\gamma+\Omega^2\int_0^\infty\dfrac{d\omega F(\omega)}{s+i(\omega-\omega_c)}}. \tag{8}$$

By performing the inverse Laplace transformation, $A(t)=(2\pi i)^{-1}\int_{\sigma-i\infty}^{\sigma+i\infty}ds\,e^{st}\tilde{A}(s)$ (see e.g. [3] for more details), we get the formal solution for the cavity amplitude $A(t)$ which is as follows

$$A(t)=\frac{e^{i(\omega_c-i\gamma)t}}{2\pi i}\int_{\sigma-i\infty}^{\sigma+i\infty}\frac{e^{st}ds}{s+\kappa-\gamma+i\omega_c+\Omega^2\int_0^\infty\dfrac{d\omega F(\omega)}{s+i\omega}}, \tag{9}$$

where $\sigma>0$ is chosen such that the real parts of all singularities of $\tilde{A}(s)$ are smaller than σ. It turned out that the integral in the denominator of Eq. (9) has a jump when passing across the negative part of the imaginary axis leading to the branch cut in the complex plane of s (see Fig. 1). By setting the denominator of the integrand in Eq. (9) to zero, one can derive the



S 2 Route from strong coupling to multimode strong coupling regime for two different coupling strengths, $\Omega/2\pi = 8$ MHz (left column) and $\Omega/2\pi = 26$ MHz (right column). *Upper row*: Kernel function $U(\omega)$. *Lower row*: Nonlinear Lamb shift $\delta(\omega)$ for the same ω-interval as above (note the different zooms for the two columns). *Left column*: Strong coupling regime with a well-resolved Rabi splitting in $U(\omega)$ (regime of damped Rabi oscillations). *Right column*: Multimode strong coupling regime with a multi-peak structure in $U(\omega)$ when all seven spin ensembles are effectively coupled to the cavity (regime of revivals). Filled circles label resonance values ω_r of the kernel $U(\omega)$ occurring at the intersections between the Lamb shift $\delta(\omega)$ and the dashed line $(\omega-\omega_c)/\Omega^2$. At empty circles such intersections are non-resonant and do not lead to a corresponding peak in $U(\omega)$. The cavity frequency ω_c coincides with the mean frequency of the central q-Gaussian, $\omega_s = \omega_c$, shown in Fig. 1(a) of the main article.

equations for simple poles, $s_j = \sigma_j + i\omega_j$, which, however, do not appear for the spectral function shown in Fig. 1(a) of the main paper and will not be discussed here (see [2] for more details about poles' contribution).

Next, we apply Cauchy's theorem to a closed contour to evaluate the formal integral (9) taking into account that only a few paths of those shown in Fig. S1 contribute. Finally, we end up with the following expression for the cavity amplitude

$$A(t) = \Omega^2 \int_0^\infty d\omega e^{-i(\omega-\omega_c-i\gamma)t} U(\omega), \tag{10}$$

where

$$U(\omega) = \lim_{\sigma\to 0^+} \left\{ \frac{F(\omega)}{(\omega-\omega_c-\Omega^2\delta(\omega)+i(\kappa-\gamma))^2+(\pi\Omega^2F(\omega)+\sigma)^2} \right\}. \tag{11}$$

is the kernel function and

$$\delta(\omega) = \mathscr{P}\int_0^\infty \frac{d\tilde{\omega}F(\tilde{\omega})}{\omega-\tilde{\omega}} \tag{12}$$

has the meaning of the nonlinear Lamb shift of the cavity frequency ω_c, which depends on the total spectral distribution, $F(\omega)$.

Obviously, the relevant frequency components contributing to the dynamics of $A(t)$ are those which are resonant in the kernel function $U(\omega)$. As it can be deduced from the structure of $U(\omega)$ given by Eq. (11), a necessary condition for such resonances to show up strongly depends on the structure of the Lamb shift and the value of the coupling strength. Namely, it is given by the following approximate formula, $(\omega_r-\omega_c)/\Omega^2 \approx \delta(\omega_r)$. At small values for the coupling strength Ω the straight line $(\omega_r-\omega_c)/\Omega^2$ becomes very steep and thus leads just to a single intersection with $\delta(\omega_r)$. As a result, a single resonance occurs at $\omega_r \approx \omega_c$, so that only the central spin ensemble contributes to the coupling with the cavity, whereas the others yield a negligible contribution. In this case the kernel function $U(\omega)$ can be well approximated by a Lorentzian centered around the slightly shifted cavity frequency $\omega_c + \Omega^2\delta(\omega_c)$. We will thus deal with the exponential decay of the cavity amplitude $A(t)$ in the time domain with a decay rate depending on Ω. Actually this regime is very similar to the Purcell enhancement of the spontaneous emission rate of a single emitter inside a cavity [4]. As the coupling strength reaches a certain critical value, the straight line intersects the nonlinear Lamb shift at three points from which only two give rise to the resonances in $U(\omega)$. As a consequence, the kernel function $U(\omega)$ consists of two well-separated polaritonic peaks, which is the hallmark of the strong coupling regime of cavity QED (see the left column in Fig. S2). Note that these two resonances still reside in the vicinity of the cavity frequency and the contribution of all but the central ensemble is rather small.

The situation changes qualitatively at higher values of the coupling strength Ω, when the straight line also intersects the other distant resonances of the Lamb shift, as is seen from the right column in Fig. S2. As a result, the kernel function $U(\omega)$ forms a comb-shaped structure with almost equally spaced polaritonic peaks at frequencies which are shifted with respect to the resonances of the spectral function $F(\omega)$. It is worth noting, that such a communication of the cavity mode with distant resonances of $F(\omega)$ would never take place if the Lamb shift were approximated by its value at the cavity frequency, $\delta(\omega_c)$.

Supplementary Note 4. Eigenvalue problem

To solve the eigenvalue problem we first discretize the spectral function $F(\omega)$ (see Fig. 1 in the main article) by performing the following transformation:

$$g_l = \left[F(\omega_l) \cdot \left(\sum_{\mu=1}^{M} \Omega_\mu^2 \right) / \sum_m F(\omega_m) \right]^{1/2}. \tag{13}$$

Since in total we deal with a sizeable number of spins, we make our problem numerically tractable by dividing spins into many subgroups with approximately the same coupling strengths, so that g_l in Eq. (13) represents a coupling strength within each subgroup rather than an individual coupling strength. Note also that once a shape of the spectral function $F(\omega)$ is defined, it is not relevant anymore to which ensemble an individual spin belongs to. By doing so we get the following linear set of first-order ODEs with respect to the cavity and spin amplitudes from Eqs. (1a,1b) ($\eta(t) = 0$)

$$\dot{A}(t) = -\kappa \cdot A(t) + \sum_l g_l B_l(t) \tag{14a}$$

$$\dot{B}_l(t) = -\left[\gamma + i(\omega_l - \omega_c)\right] B_l(t) - g_l A(t), \tag{14b}$$

where $A(t) \equiv \langle a(t) \rangle$ and $B_l(t) \equiv \langle \sigma_l^-(t) \rangle$. After substituting $A(t) = A \cdot \exp(-\lambda t)$ and $B_l(t) = B_l \cdot \exp(-\lambda t)$ into Eqs. (14a, 14b), we derive the complex eigenvalue problem for λ, which can be represented as, $\mathscr{L}\psi = \lambda\psi$, where

$$\mathscr{L} = \begin{pmatrix} \kappa & -g_1 & -g_2 & \dots & -g_N \\ g_1 & \gamma + i(\omega_1 - \omega_c) & 0 & \dots & 0 \\ g_2 & 0 & \gamma + i(\omega_2 - \omega_c) & \dots & 0 \\ \dots & \dots & \dots & \dots & \dots \\ g_N & 0 & 0 & \dots & \gamma + i(\omega_N - \omega_c) \end{pmatrix},$$

and $\psi = (A \;\; B_1 \;\; B_2 \dots B_N)^T$.

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