

Supplementary Information for “Nearly quantum-limited Josephson-junction Frequency Comb synthesizer”

Pinlei Lu*, Tzu-Chiao Chien, Xi Cao, Olivia Lanes, Chao Zhou, and Michael J. Hatridge

Department of Physics and Astronomy, University of Pittsburgh

Saeed Khan*, Hakan E. Türeci

Department of Electrical Engineering, Princeton University

**These authors contributed equally to this publication.*

(Dated: March 30, 2021)

CONTENTS

I. Lyapunov stability	1
II. Supplementary experimental details and results	2
A. Phase diagram for Device B (5 SQUIDs)	2
B. Kerr nonlinearity measurement	3
C. Typical Kerr nonlinearity strength of optical microresonators	5
III. Simulating stochastic differential equations	6
A. Practical computation of steady-state operator moments and correlation functions using SDEs	6
B. Calculating the filtered output temporal coherence function	7
C. Non-exponential signatures in phase decoherence	9
IV. Dephasing in the weak-driving regime	10
A. Exact dynamics	10
B. Dephasing measurement in the two-level approximation	11
C. Cavity ringdown method and theoretical simulations	12
V. Derivation of Floquet eigensystem	13
References	15

I. LYAPUNOV STABILITY

In this section we describe a standard numerical approach to the calculation of Lyapunov exponents of dynamical systems, which we use for the analysis in Appendix C of the manuscript. Employing the notation from that Appendix, consider a deterministic trajectory $\vec{\zeta}(t)$ evolving according to the nonlinear equations of motion describing the classical system, Eqs. (B2). Linearized fluctuations about this trajectory, $\vec{z}(t)$, are then propagated by the time-dependent Jacobian matrix evaluated along the deterministic trajectory:

$$\frac{d\vec{z}}{dt} = \mathbf{J}[\vec{\zeta}(t)] \cdot \vec{z}(t) \quad (1)$$

which is simply the system given by Eqs. (B11) from the manuscript, Appendix B, but with the Jacobian now evaluated along a general time-dependent trajectory $\vec{\zeta}(t)$. Such trajectories may exhibit complicated dynamics, but being governed by a linear system, this evolution is ultimately always related to characteristic exponents obtained from the dynamical matrix. The maximal Lyapunov exponent λ_M is the largest such exponent (accounting for sign, not magnitude); it plays the role that the largest eigenvalue would play in the case of a dynamical system governed by a time-*independent* dynamical matrix. The maximal Lyapunov exponent may be computed by studying the long-time dynamics of trajectories governed by Eq. (1):

$$\lambda_M = \lim_{t \rightarrow \infty} \frac{1}{t} \log \frac{\|\vec{z}(t)\|}{\|\vec{z}(0)\|} \quad (2)$$

However, since the evolution of trajectories is governed by a linear dynamical equation, these trajectories may grow unbounded exponentially with time if the Lyapunov exponent is positive. In practice this exponential growth renders the above expression intractable for numerical computations. An alternative procedure to circumvent this issue begins by separating the time evolution from $[0, t]$ into a series of N_p consecutive short-time intervals, $[\tau_0, \tau_1, \dots, \tau_{N_p}]$, where $\tau_0 = 0, \tau_{N_p} = t$, and $\tau_p - \tau_{p-1} = \Delta\tau$ is the short-time interval spacing. We then solve for $\vec{z}^{(p)}(\tau)$, $\tau \in [\tau_{p-1}, \tau_p]$ with $p = 1, \dots, N_p$, obtaining N_p individual trajectory vectors $\{\vec{z}^{(p)}(\tau)\}$. Such an evolution would be identical to the entire evolution over $[0, t]$ if we imposed $\vec{z}^{(p)}(\tau_{p-1}) = \vec{z}^{(p-1)}(\tau_{p-1})$, requiring continuity of the solution at the endpoints of each time interval. Consequently, the process would do nothing to alleviate the problem of unbounded growth. To guard against the latter, we additionally require the initial trajectory at the beginning of every evolution interval to be normalized to one:

$$\vec{z}^{(p)}(\tau_{p-1}) = \frac{\vec{z}^{(p-1)}(\tau_{p-1})}{\|\vec{z}^{(p-1)}(\tau_{p-1})\|} \implies \|\vec{z}^{(p)}(\tau_{p-1})\| = 1 \quad (3)$$

Then, for the p th iterate, we can estimate the maximal Lyapunov exponent as:

$$\lambda_M^{(p)} = \frac{1}{\Delta\tau} \log \frac{\|\vec{z}^{(p)}(\tau_p)\|}{\|\vec{z}^{(p)}(\tau_{p-1})\|} = \frac{1}{\Delta\tau} \log \|\vec{z}^{(p)}(\tau_p)\| \quad (4)$$

which simply measures the growth of the norm of the p th trajectory $\vec{z}^{(p)}(\tau)$ for $\tau \in [\tau_{p-1}, \tau_p]$.

An estimate for the actual maximal Lyapunov exponent is then finally computed by averaging over the iterations:

$$\lambda_M \approx \frac{1}{N_p} \sum_{p=1}^{N_p} \lambda_M^{(p)} \quad (5)$$

The above procedure ensures a faithful and *bounded* evolution of a single trajectory over a time $N_p \Delta\tau$, *provided* $\Delta\tau$ is chosen judiciously relative to $|1/\lambda_M|$; the latter sets the characteristic evolution timescale for the trajectories and is of course *a priori* unknown. If $\Delta\tau \gg |1/\lambda_M|$, trajectories may grow too substantially during the evolution if $\lambda_M > 0$, leading to the same numerical errors present in the original formulation, Eq. (2). If instead $\Delta\tau \ll |1/\lambda_M|$, the normalized trajectories will remain mostly unchanged from their initial values, leading to an estimate of $\lambda_M^{(p)} \rightarrow 0$ regardless of its actual value. Therefore an intermediate $\Delta\tau$ value must be employed, keeping in mind also that smaller $\Delta\tau$ values necessarily require the use of more iterations N_p for convergence. In practice, the convergence of the estimate may be evaluated by calculating λ_M as a function of increasing N_p for a fixed $\Delta\tau$, until λ_M remains approximately unchanged with further iterations. Comparing a set of such estimates for a range of $\Delta\tau$ values then ensures that a consistent estimate is obtained.

The maximal Lyapunov exponent λ_M obtained with this approach is plotted for Device A parameters in drive- Δ_{db} space in Fig. 5 of the manuscript.

II. SUPPLEMENTARY EXPERIMENTAL DETAILS AND RESULTS

In this section, we include details of additional experimental measurements, including the measured phase diagram for Device B, as well as the measurement of the Kerr nonlinearity strength.

A. Phase diagram for Device B (5 SQUIDs)

In addition to Device A, which employs a 25 SQUID array, we explore the impact of nonlinearity on comb dynamics by fabricating Device B, which employs a 5 SQUID array and therefore possesses an approximately 25-fold stronger nonlinearity. In Fig. 1 (a), we show the flux sweep of this device, indicating the polariton resonances of the two-mode system. Fitting to the avoided crossing reveals a coupling strength of $g/2\pi = 89.25$ MHz, similar to Device A (by design), and a bare linear mode linewidth of $\kappa/2\pi = 22.84$ MHz.

Having verified that the device satisfies the strong coupling condition $g > \frac{\kappa}{2}$ at resonant driving ($\Delta_{da} = 0$, see main text), we can explore the classically predicted unstable regime as was done for Device A. Fixing the driving frequency at $\omega_d/2\pi = 4.9085$ GHz, we change the external flux through the SQUIDs to sweep the nonlinear mode frequency, as shown schematically in the top panel of Fig. 1 (a). Device B has a much larger flux modulation range than Device A, enabling us to explore a wider range of drive-nonlinear mode detunings Δ_{db} . The resulting phase diagram in drive

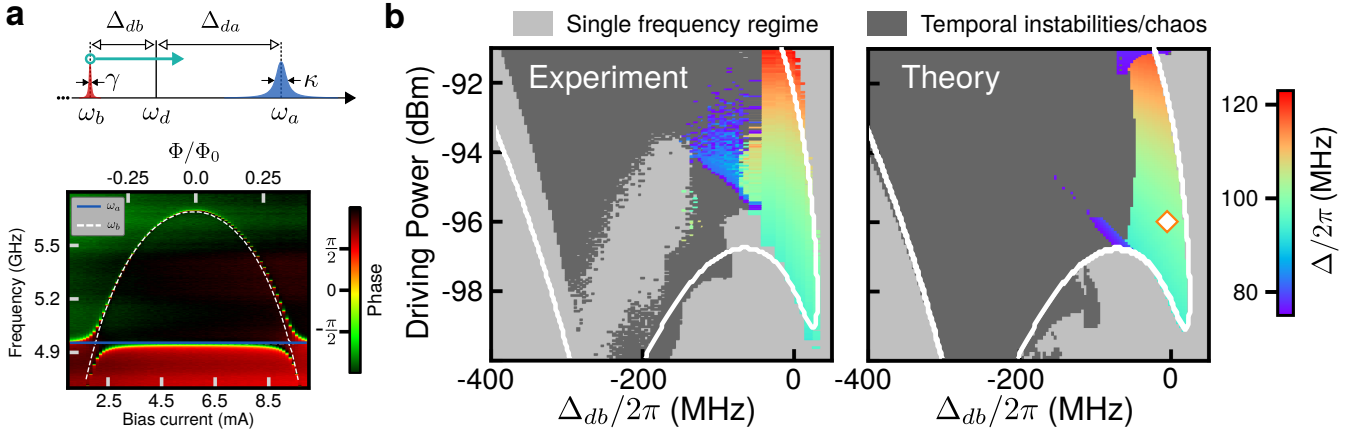


Figure 1. **Device B measurements.** (a) Flux sweep showing the system polariton modes and the avoided crossing as the nonlinear mode is swept across the linear mode resonance. The horizontal blue line indicates the bare linear mode frequency, while the white dashed line indicates the bare nonlinear mode frequency. (b) Experimental and theoretical phase diagram in Drive- Δ_{db} space, as the nonlinear mode frequency is swept (see schematic in (a)). The observed comb spacing Δ in the multifrequency regime is plotted, alongside the single frequency regime (light gray) and regime with temporal instabilities (dark gray). White curve in both phase diagrams is the analytically obtained boundary enclosing the classically unstable region. Orange diamond indicates the drive power (-96 dBm) and detuning ($\Delta_{db}/2\pi = -5.87$ MHz) for which coherence function results are plotted in Fig. 3(c) of the main text for Device B.

power- Δ_{db} space is shown in Fig. 1 (b), with the theoretically predicted phase diagram shown in the right panel, both plotted with the same axes. The light gray regions indicate the single frequency regime, which gives way to a multifrequency comb regime at appropriate drive strengths for small $|\Delta_{db}|$. The experiment and theory agree quite well both in terms of the critical detuning where the combs emerge, as well as the observed comb spacings. Finally the orange diamond in the theory plot indicates the position on the phase diagram for which coherence function results are plotted in Fig. 3 of the main text.

Note that for more negative Δ_{db} , a region (dark gray) emerges where the system exhibits temporal instabilities similar to Device A, in both the experimental and theoretical phase diagrams. Numerical simulations here indicate that the system exhibits chaotic dynamics (maximal Lyapunov exponent $\lambda_M > 0$). We also find greater disparity between experiment and theory here; in addition to possible deviations from classical predictions due to quantum effects, the dynamics exhibit temporal instabilities that require careful processing. Such dynamical regimes therefore merit further detailed investigation. The white contour in both figures depicts the analytically predicted unstable region, as determined by the linearized dynamics of Eqs. (B11) of the manuscript; it agrees well with both experiment and numerical simulations, in particular for small $|\Delta_{db}|$.

B. Kerr nonlinearity measurement

To demonstrate the dependence of comb coherence on the quantum nature of the device nonlinearity, knowledge of this engineered Kerr nonlinearity strength is of crucial importance. Typically, one would do so via a standard pump-probe measurement that measures the Kerr-induced frequency shift of the nonlinear mode as the pump power incident on it increases. However, for the two-mode system such a measurement accesses the frequency shift of the renormalized *polariton* modes of the system, which of course depends on the degree of hybridization between linear and nonlinear modes. In this section we clarify how measured polariton mode frequency shifts can be used to extract the bare nonlinear mode Kerr interaction strength.

We begin by rewriting below, for convenience, the full system Hamiltonian ($\hbar = 1$) from Eq. (1) of the main text:

$$\hat{\mathcal{H}} = -\Delta_{da}\hat{a}^\dagger\hat{a} - \Delta_{db}\hat{b}^\dagger\hat{b} - \frac{\Lambda}{2}\hat{b}^\dagger\hat{b}^\dagger\hat{b}\hat{b} + g(\hat{a}^\dagger\hat{b} + \hat{a}\hat{b}^\dagger) + \eta(\hat{a} + \hat{a}^\dagger) \quad (6)$$

Next, we consider the linear Hamiltonian $\hat{\mathcal{H}}_L$ that determines the polariton modes:

$$\hat{\mathcal{H}}_L = \omega_a\hat{a}^\dagger\hat{a} + \omega_b\hat{b}^\dagger\hat{b} + g(\hat{a}^\dagger\hat{b} + \hat{a}\hat{b}^\dagger) \equiv \begin{pmatrix} \hat{a}^\dagger & \hat{b}^\dagger \end{pmatrix} \underbrace{\begin{pmatrix} \omega_a & g \\ g & \omega_b \end{pmatrix}}_{\mathbf{H}_L} \begin{pmatrix} \hat{a} \\ \hat{b} \end{pmatrix} \quad (7)$$

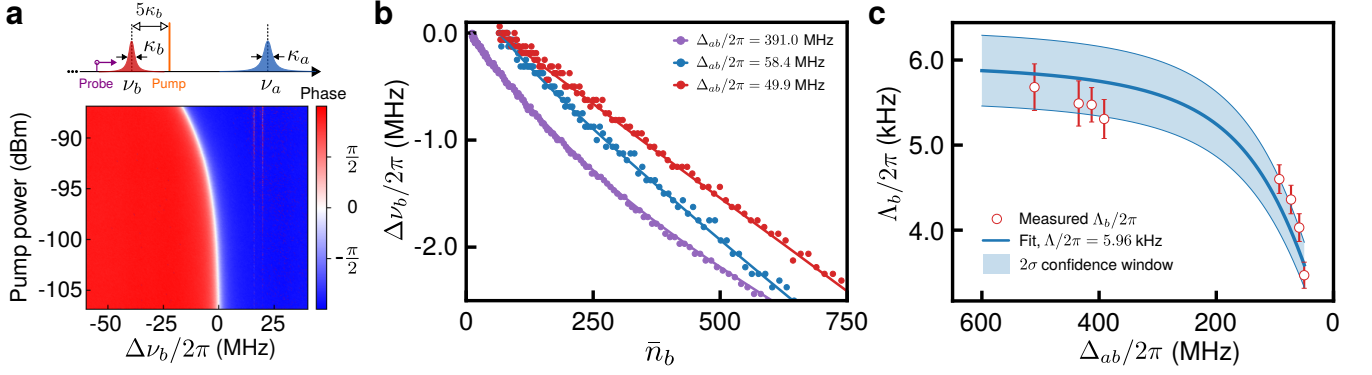


Figure 2. **Kerr nonlinearity measurement.** (a) Left column: schematic showing the pump-probe setup. Note that modes shown are *polariton* modes, not the bare modes. A strong pump tone (orange) is applied 5 linewidths positively detuned from $\hat{c}_b$ (see text), and the modified polariton resonance frequency identified using a weak probe (purple). Increasing polariton mode occupation $\bar{n}_b$ by increasing pump tone power, the frequency shift $\Delta\nu_b$ of polariton mode is measured versus $\bar{n}_b$ for various detunings between the bare linear and nonlinear modes, $\Delta_{ab} = \omega_a - \omega_b$. Lines are fits to $-\Lambda_b\bar{n}_b + \epsilon O(\bar{n}_b^2)$ where higher order terms $\propto \epsilon$ become important as pump power increases. The fits are used to extract the polariton mode nonlinearity Λ_b . With decreasing detuning Δ_{ab} , the initial slope of the fit decreases, indicating a decrease in the strength of the $\hat{c}_b$ mode nonlinearity as the hybridization increases. (b) Measured Kerr nonlinearity Λ_b of polariton mode $\hat{c}_b$ (red circles) extracted from (a), as a function of detuning Δ_{ab} . Solid blue line is the best fit to Eq. (17), with the obtained fit value of $\Lambda/2\pi = 5.96$ kHz. The shaded region indicates the 2σ confidence interval for the fit.

which is obtained from Eq. (6) by neglecting the nonlinearity and drive terms, and returning to the lab frame. The above Hamiltonian may be diagonalized by introducing the matrix of eigenvectors $\mathbf{P}$ and diagonal matrix of eigenvalues $\mathbf{D}$ for the matrix $\mathbf{H}_L$, such that $\mathbf{H}_L = \mathbf{P}\mathbf{D}\mathbf{P}^{-1}$. The Hamiltonian then becomes:

$$\hat{\mathcal{H}}_L = \nu_a \hat{c}_a^\dagger \hat{c}_a + \nu_b \hat{c}_b^\dagger \hat{c}_b, \quad \begin{pmatrix} \hat{c}_a \\ \hat{c}_b \end{pmatrix} = \mathbf{P}^{-1} \begin{pmatrix} \hat{a} \\ \hat{b} \end{pmatrix}, \quad \mathbf{D} = \begin{pmatrix} \nu_a & 0 \\ 0 & \nu_b \end{pmatrix} \quad (8)$$

which serves to define the polariton modes $\hat{c}_a$, $\hat{c}_b$, and corresponding frequencies ν_a , ν_b .

We can now rewrite the Kerr nonlinear term of the full Hamiltonian, Eq. (6), in the polariton basis. Writing the nonlinear term $\hat{\mathcal{H}}_\Lambda$ as:

$$\hat{\mathcal{H}}_\Lambda = -\frac{\Lambda}{2} \hat{b}^\dagger \hat{b}^\dagger \hat{b} \hat{b} \quad (9)$$

and noting from Eq. (8) that:

$$\hat{b} = \mathbf{P}_{21}\hat{c}_a + \mathbf{P}_{22}\hat{c}_b = \sum_n \mathbf{P}_{2n}\hat{c}_n \quad (10)$$

the nonlinear Hamiltonian in terms of polariton modes takes the form:

$$\hat{\mathcal{H}}_\Lambda = -\frac{\Lambda}{2} \sum_{nmrs} \mathbf{P}_{2n}^* \mathbf{P}_{2m}^* \mathbf{P}_{2r} \mathbf{P}_{2s} \hat{c}_n^\dagger \hat{c}_m^\dagger \hat{c}_r \hat{c}_s \equiv -\frac{\Lambda}{2} \sum_{nmrs} \mathcal{A}_{nmrs} \hat{c}_n^\dagger \hat{c}_m^\dagger \hat{c}_r \hat{c}_s \quad (11)$$

Therefore, the coupling transforms the localized nonlinearity of mode $\hat{b}$ into self- and cross-Kerr interactions between the polariton modes of the system. The Kerr-induced frequency shift observed for either polariton mode will be a combination of these terms, making it complicated to determine in general.

However, we can obtain a simplified expression by assuming operation near a stable fixed point and assuming a strong polariton mode occupation, both conditions that are expected to be valid for the typical pump-probe measurement scheme. The experimental scheme proceeds similarly to the case for a single nonlinear mode: a strong pump tone is applied to the system at a positive detuning of five linewidths away from *polariton* mode $\hat{c}_b$, predominantly pumping this mode, although also residually (weakly) pumping mode $\hat{c}_a$ (see schematic in Fig. 2 (a)). The resulting steady-state polariton amplitudes, and therefore occupations, can be conveniently determined by first obtaining the nonlinear and linear mode amplitudes $\bar{\beta}$, $\bar{\alpha}$ by solving Eqs. (B6) and (B8) respectively, reproduced below:

$$\left[\left(\tilde{\Delta}_{db} + \Lambda |\bar{\beta}|^2 \right)^2 + \frac{\tilde{\gamma}^2}{4} \right] |\bar{\beta}|^2 = g^2 |\chi_a|^2 \eta^2 \quad (12a)$$

$$\bar{\alpha} = -\chi_a (ig\bar{\beta} + i\eta) \quad (12b)$$

recalling the renormalized nonlinear mode detuning and damping parameters respectively:

$$\begin{aligned}\tilde{\Delta}_{db} &= \omega_d - (\omega_b + g^2|\chi_a|^2\Delta_{da}) \\ \tilde{\gamma} &= \gamma + \gamma_\varphi + g^2|\chi_a|^2\kappa\end{aligned}\quad (13)$$

where $\chi_a = (-i\Delta_{da} + \frac{\kappa}{2})^{-1}$. Then, the steady-state polariton amplitudes, $\bar{c}_a$, $\bar{c}_b$, are easily determined via the transformation matrix introduced in Eq. (8):

$$\begin{pmatrix} \bar{c}_a \\ \bar{c}_b \end{pmatrix} = \mathbf{P}^{-1} \begin{pmatrix} \bar{\alpha} \\ \bar{\beta} \end{pmatrix} \quad (14)$$

Finally, the application of a weak probe determines Kerr-mediated frequency shifts, as dictated by the nonlinear Hamiltonian, Eq. (11). We are only interested in shifts to the polariton mode $\hat{c}_b$; the corresponding terms of the nonlinear Hamiltonian are given by:

$$\hat{\mathcal{H}}_\Lambda \approx -\frac{\Lambda}{2} \left[\mathcal{A}_{2222}\hat{c}_b^\dagger\hat{c}_b + 4\mathcal{A}_{2121}\hat{c}_a^\dagger\hat{c}_a + 2\mathcal{A}_{2221}\hat{c}_b^\dagger\hat{c}_a + 2\mathcal{A}_{2122}\hat{c}_a^\dagger\hat{c}_b \right] \hat{c}_b^\dagger\hat{c}_b + (\hat{c}_a^\dagger\hat{c}_a \text{ --only and non Kerr shift terms}) \quad (15)$$

We now perform a semiclassical approximation, linearizing the above Hamiltonian around the fixed point defined by Eqs. (14), under which the effective Kerr-mediated shift $\Delta\nu_b$ of the polariton frequency ν_b is given by:

$$\Delta\nu_b = -\Lambda \left[\mathcal{A}_{2222}|\bar{c}_b|^2 + 2\mathcal{A}_{2121}|\bar{c}_a|^2 + 2\mathcal{A}_{2221}\bar{c}_b^*\bar{c}_a + \mathcal{A}_{2122}\bar{c}_a^*\bar{c}_b \right] \quad (16)$$

Finally, the effective measured Kerr constant Λ_b is obtained by determining the frequency shift per photon occupying the polariton mode, $\bar{n}_b = |\bar{c}_b|^2$:

$$\Lambda_b = -\frac{\Delta\nu_b}{\bar{n}_b} = \Lambda \left[\mathcal{A}_{2222} + 2\mathcal{A}_{2121}\frac{|\bar{c}_a|^2}{|\bar{c}_b|^2} + 2\mathcal{A}_{2221}\frac{\bar{c}_a}{\bar{c}_b} + \mathcal{A}_{2122}\frac{\bar{c}_a^*}{\bar{c}_b^*} \right] \quad (17)$$

Clearly, $\Delta\nu_b$ and the measured Kerr constant Λ_b depend on $\mathcal{A}_{nmrs}$ and consequently on the detuning between the bare linear and nonlinear modes, $\Delta_{ab} = \omega_a - \omega_b$, as well as the strength of their coupling g . As a result, both will vary as the nonlinear mode frequency ω_b is swept, even though the bare nonlinear mode Kerr constant Λ remains unchanged. In addition to this dependence on ω_b , Eq. (17) also accounts for the small but nonzero occupation of polariton mode $\hat{c}_a$ due to this mode being weakly driven, and the corresponding cross-Kerr shifts this mediates.

Experimentally, a single pump-probe measurement with pump frequency ω_P at a fixed nonlinear mode frequency populates the polariton mode $\hat{c}_b$ as the pump power P is increased. We first calibrate the polariton mode occupation with the applied pump power via $\bar{n}_b = |\bar{c}_b|^2 = \frac{\kappa_b}{\Delta_P^2 + (\kappa_b/2)^2} \frac{P}{\hbar\omega_P}$, where κ_b is the linewidth of polariton mode $\hat{c}_b$, and $\Delta_P = 5\kappa_b$ is the detuning between the pump frequency and the bare polariton mode frequency¹. The observed frequency shift $\Delta\nu_b$ as a function of $\bar{n}_b$ is shown in Fig. 2 (a) for various detunings between the bare linear and nonlinear modes Δ_{ab} . By fitting the observed frequency shift to $\bar{n}_b$, we obtain the measured polariton mode Kerr constant Λ_b . Each such measurement yields Λ_b at the given Δ_{ab} . By sweeping the nonlinear mode frequency, we obtain Λ_b as a function of Δ_{ab} , with the results plotted in red in Fig. 2 (b). Note that as the detuning Δ_{ab} decreases, the measured Kerr nonlinearity strength also decreases, since increased hybridization dilutes the nonlinearity of the originally nonlinear mode. By fitting the experimental results to Eq. (17) with the bare nonlinearity Λ as the only fitting parameter, we obtain the solid blue curve in Fig. 2 (b), with the fit value $\Lambda/2\pi = 5.96$ kHz. The shaded blue region indicates the 2σ confidence interval of the fit, which finally yields the bare nonlinearity of $\Lambda/2\pi = 5.96 \pm 0.2$ kHz for Device A.

C. Typical Kerr nonlinearity strength of optical microresonators

In this subsection we calculate the typical Kerr nonlinearity strength, or equivalently the Kerr-mediated frequency shift per photon, for nonlinear optical microresonators. For an optical microresonator with center frequency ω_{op} , refractive index n , second-order nonlinear refractive index n_2 , and mode volume V_0 , the Kerr shift per photon, Λ_{op} is given by²:

$$\Lambda_{\text{op}} = \frac{\hbar\omega_0^2 cn_2}{n^2 V_0} \quad (18)$$

where c is the speed of light in vacuum. Using parameter values for silicon nitride optical microresonators³ - a popular and successful material choice - we have: $\omega_{\text{op}}/(2\pi) = 100$ THz (equivalently, wavelength $\lambda \simeq 1.55$ μm), $n = 2$, $n_2 = 2.5 \times 10^{-19}$ $\text{m}^2 \text{W}^{-1}$, and $V_0 = (\lambda/n)^3$, we obtain:

$$\Lambda_{\text{o}}/(2\pi) \simeq 100 \text{ Hz} \quad (19)$$

which is about two orders of magnitude lower than the realized Λ for Device A. Optical microresonators are engineered to have high quality factors; we consider a large value of $Q \simeq 10^7$. For $\omega_{\text{op}}/(2\pi) = 100$ THz, this implies microresonator loss rates of $\kappa_{\text{op}}/(2\pi) \simeq 10$ MHz. As a result, the ratio of Λ_{op} to the loss rate is $\Lambda_{\text{op}}/\kappa_{\text{op}} \simeq 10^{-5}$, again about two orders of magnitude smaller than the smallest value realized by devices in our experiment.

III. SIMULATING STOCHASTIC DIFFERENTIAL EQUATIONS

In this section, we provide details of the numerical simulations of the SDEs (Eqs. (3) of the manuscript), and in particular how these are used to calculate quantities of interest such as the temporal coherence functions. For convenience, we reproduce the set of SDEs below:

$$d\vec{\zeta} = \vec{A}_c(\vec{\zeta})dt + \mathbf{B}_{\text{st}}(\vec{\zeta}, \Lambda, \gamma_\varphi)d\vec{W}(t) \quad (20)$$

where $\mathbf{B}_{\text{st}}$ is the matrix square root of the diffusion matrix, defined via $\mathbf{D}_{\text{st}} = \mathbf{B}_{\text{st}}\mathbf{B}_{\text{st}}^T$. For a 4-by-4 diffusion matrix $\mathbf{D}$, the noise matrix $\mathbf{B}$ is not unique; it is in general a 4-by- k non-square matrix, with $d\vec{W}(t)$ then being a k -by-1 vector of independent Wiener increments. While this freedom of choice in the noise matrix can be used to improve SDE convergence properties⁴, we find that here a square matrix ($k = 4$) suffices. We write it in the form:

$$\mathbf{B}_{\text{st}} = \sqrt{\Gamma} \mathbf{B}_1 + \sqrt{\gamma_\varphi} \mathbf{B}_2 = \sqrt{\Gamma} \begin{pmatrix} \mathbf{0} & \mathbf{0} \\ \mathbf{b}_1 & \mathbf{0} \end{pmatrix} + \sqrt{\gamma_\varphi} \begin{pmatrix} \mathbf{0} & \mathbf{0} \\ \mathbf{0} & \mathbf{b}_2 \end{pmatrix} \quad (21)$$

where $\mathbf{0}$ is the 2-by-2 matrix of zeros as before, and $\mathbf{B}_1, \mathbf{B}_2$ are the noise matrices introduced in the main text. Here we also provide their explicit forms in terms of the 2-by-2 component matrices $\mathbf{b}_1$ and $\mathbf{b}_2$:

$$\mathbf{b}_1 = \begin{pmatrix} e^{i\theta/2}\beta & 0 \\ 0 & e^{-i\theta/2}\beta^\dagger \end{pmatrix}, \quad \mathbf{b}_2 = \sqrt{\frac{\beta^\dagger\beta}{2}} \begin{pmatrix} e^{i\pi/4} & e^{-i\pi/4} \\ e^{-i\pi/4} & e^{i\pi/4} \end{pmatrix} \quad (22)$$

Finally, we have defined the parameters Γ and θ via:

$$\Gamma e^{i\theta} \equiv i\Lambda - \gamma_\varphi \implies \Gamma = \sqrt{\Lambda^2 + \gamma_\varphi^2}, \quad \theta = \arctan\left(-\frac{\Lambda}{\gamma_\varphi}\right). \quad (23)$$

The validity of the noise matrix $\mathbf{B}_{\text{st}}$ as the square root of the diffusion matrix $\mathbf{D}_{\text{st}}$ may be easily verified by direct multiplication.

A. Practical computation of steady-state operator moments and correlation functions using SDEs

Simulations of the SDEs in Eq. (20) yield individual stochastic trajectories of the stochastic variables $\vec{\zeta}(t)$, which may then be used to compile *normal-ordered* moments and correlation functions. In what follows, we use expressions for moments and correlation functions for the linear mode as examples, since these are directly accessible via experiment. However the expressions hold equally for nonlinear mode operators by appropriate substitutions.

Suppose Eqs. (20) are solved to obtain N_s stochastic trajectories, yielding a set of stochastic trajectories $\{\vec{\zeta}_i(t)\}$ for $i = 1, \dots, N_s$. Then, first-order moments for the linear mode may be determined via stochastic averaging (indicated by notation $\langle \cdot \rangle_s$) as follows:

$$\langle \hat{a}(t) \rangle = \langle \alpha(t) \rangle_s = \lim_{N_s \rightarrow \infty} \frac{1}{N_s} \sum_{i=1}^{N_s} \alpha_i(t) \quad (24)$$

Normal-ordered two-time correlation functions follow similarly:

$$\langle \hat{a}^\dagger(t+\tau)\hat{a}(t) \rangle = \langle \alpha^\dagger(t+\tau)\alpha(t) \rangle_s = \lim_{N_s \rightarrow \infty} \frac{1}{N_s} \sum_{i=1}^{N_s} \alpha_i^\dagger(t+\tau)\alpha_i(t) \quad (25a)$$

$$\langle \hat{a}(t+\tau)\hat{a}(t) \rangle = \langle \alpha(t+\tau)\alpha(t) \rangle_s = \lim_{N_s \rightarrow \infty} \frac{1}{N_s} \sum_{i=1}^{N_s} \alpha_i(t+\tau)\alpha_i(t) \quad (25b)$$

Note that the other normal-ordered, time anti-ordered correlation functions $\langle \hat{a}^\dagger(t)\hat{a}(t+\tau) \rangle$ and $\langle \hat{a}^\dagger(t)\hat{a}^\dagger(t+\tau) \rangle$ may be obtained from the two expressions above respectively by conjugation.

While the above expressions allow access to moments and correlation functions at arbitrary times, when analyzing the long time coherence of the emergent frequency combs we will ultimately be interested in steady state quantities. The requirement of a steady state allows an alternative calculation of the above quantities. To directly acquire steady state quantities, we simulate Eqs. (20) for times $t \in [0, t_{ss} + T_W]$ with simulation time step Δt , and retain solutions in the time window $t \in [t_{ss}, t_{ss} + T_W]$ where T_W is the length of time window. The time t_{ss} is chosen long enough that the solutions $\{\vec{\zeta}_i(t)\}$ within the stored window are extracted when initial transients have decayed away; this initial t_{ss} value is verified self-consistently, as discussed at the end of this section. For simplicity, we now index the solutions in this time window by times $t_j \in [t_{ss}, t_{ss} + T_W]$, such that $t_j = t_{ss} + j\Delta t$ for $j = 0, \dots, M_1$, where $M_1 = T_W/\Delta t$.

Since in the steady state first order moments should become stationary in time, we can equivalently average moments over time, with the results being equivalent to ensemble averaging if a true steady state has been achieved. In practice, to take advantage of parallelization available with modern computing clusters, we compute moments by averaging over *both* trajectories and time:

$$\langle \hat{a} \rangle \approx \frac{1}{N_s} \sum_{i=1}^{N_s} \left[\frac{1}{M_1 + 1} \sum_{j=0}^{M_1} \alpha_i(t_j) \right] \quad (26)$$

where the term in square bracket implements time-averaging.

Similarly, two-time correlation functions reduce to single time quantities in the steady state. Suppose we wish to compute correlation functions such as $\langle \hat{a}^\dagger(\tau)\hat{a}(0) \rangle$ for $\tau \in [0, T_A]$ where $T_A \leq T_W$. The time average in this case is now performed over a subset of the total window of length T , namely over t_j where $j = 0, \dots, M_2$ where $M_2 = (T_W - T_A)/\Delta t \leq M_1$. Then, steady state correlation functions may be obtained by ensemble and time averaging via:

$$\langle \hat{a}^\dagger(\tau)\hat{a}(0) \rangle \approx \frac{1}{N_s} \sum_{i=1}^{N_s} \left[\frac{1}{M_2 + 1} \sum_{j=0}^{M_2} \alpha_i^\dagger(t_j + \tau)\alpha_i(t_j) \right] \quad (27)$$

Note that if $T_A = T_W$, the required correlation function spans the entire retained window of length T_W ; only a single correlation function is obtained ($M_2 = 0$) and thus time averaging has no effect. In practice, we retain solutions for a time window T_W that is larger than the required length of the correlation function T_A , so that time-averaging can be performed.

Finally, to verify that all averaged results computed above are truly steady state quantities, we increase the value of t_{ss} beyond its chosen initial value and recompute the results, checking to see whether the averaged quantities are unchanged. If so, they are independent of t_{ss} and we can be confident of having computed steady state quantities. Otherwise, the procedure is repeated for increasing t_{ss} values until this condition is met.

B. Calculating the filtered output temporal coherence function

For calculations of comb coherence, we introduce in the main text the first-order temporal coherence function $G^{(1)}(\tau)$; here we rewrite it in a slightly different but ultimately equivalent form:

$$G^{(1)}(\tau) = \frac{\langle \Delta \hat{I}(0) \Delta \hat{I}(\tau) \rangle}{\langle \Delta \hat{I}(0) \Delta \hat{I}(0) \rangle} = \frac{\langle \hat{I}(0) \hat{I}(\tau) \rangle - \langle \hat{I}(0) \rangle^2}{\langle \hat{I}(0) \hat{I}(0) \rangle - \langle \hat{I}(0) \rangle^2} \quad (28)$$

where $\hat{I}(t)$ is the *measured* cavity output quadrature, and we have introduced *reduced* steady-state correlation functions for arbitrary operators $\hat{o}_1, \hat{o}_2$ as:

$$\langle \Delta \hat{o}_1(0) \Delta \hat{o}_2(\tau) \rangle = \langle (\hat{o}_1(0) - \langle \hat{o}_1 \rangle)(\hat{o}_2(\tau) - \langle \hat{o}_2 \rangle) \rangle = \langle \hat{o}_1(0) \hat{o}_2(\tau) \rangle - \langle \hat{o}_1 \rangle \langle \hat{o}_2 \rangle \quad (29)$$

Note, however, that simulating the SDEs of Eqs. (20) only yields *intracavity* quantities, while we require measured cavity *output* quantities to compute $G^{(1)}(\tau)$. In this section, we show how the two can be related using quantum input-output theory⁵. We begin by analyzing the steady state output quadrature correlation function:

$$\langle \hat{i}(0)\hat{i}(\tau) \rangle = \langle \hat{i}(\tau)\hat{i}(0) \rangle^* \quad (30)$$

where $\hat{i}(t)$ is the cavity mode output quadrature *prior to any post-processing* (in particular downconversion and demodulation) carried out in the experiment. It can be written in terms of the output field non-Hermitian operators $\hat{a}_{\text{out}}(t)$:

$$\hat{i}(t) = \frac{1}{\sqrt{2}} \left(\hat{a}_{\text{out}}(t) + \hat{a}_{\text{out}}^\dagger(t) \right) \quad (31)$$

In terms of the non-Hermitian output operators, the output quadrature correlation function takes the form:

$$\langle \hat{i}(0)\hat{i}(\tau) \rangle = \frac{1}{2} \left(\langle \hat{a}_{\text{out}}(0)\hat{a}_{\text{out}}(\tau) \rangle + \langle \hat{a}_{\text{out}}^\dagger(0)\hat{a}_{\text{out}}^\dagger(\tau) \rangle + \langle \hat{a}_{\text{out}}^\dagger(0)\hat{a}_{\text{out}}(\tau) \rangle + \langle \hat{a}_{\text{out}}(0)\hat{a}_{\text{out}}^\dagger(\tau) \rangle \right) \quad (32)$$

It now proves useful to normal-order and time anti-order the individual correlation functions. This requires use of the commutation relationships between the non-Hermitian output operators⁵:

$$[\hat{a}_{\text{out}}(t), \hat{a}_{\text{out}}(t')] = 0 = [\hat{a}_{\text{out}}^\dagger(t), \hat{a}_{\text{out}}^\dagger(t')], [\hat{a}_{\text{out}}(t), \hat{a}_{\text{out}}^\dagger(t')] = \delta(t - t') \quad (33)$$

Then, the output quadrature correlation function becomes:

$$\langle \hat{i}(0)\hat{i}(\tau) \rangle = \frac{1}{2} \left(\langle \hat{a}_{\text{out}}(\tau)\hat{a}_{\text{out}}(0) \rangle + \langle \hat{a}_{\text{out}}^\dagger(0)\hat{a}_{\text{out}}^\dagger(\tau) \rangle + \langle \hat{a}_{\text{out}}^\dagger(0)\hat{a}_{\text{out}}(\tau) \rangle + \langle \hat{a}_{\text{out}}(\tau)\hat{a}_{\text{out}}(0) \rangle + \delta(\tau) \right) \quad (34)$$

Note that the second and fourth terms in the expressions are simply conjugates of the first and third terms respectively.

To calculate the reduced correlation function, we require the steady state quantity:

$$\langle \hat{i}(0) \rangle \langle \hat{i}(\tau) \rangle = \frac{1}{2} \left(\langle \hat{a}_{\text{out}}(0) \rangle \langle \hat{a}_{\text{out}}(\tau) \rangle + \langle \hat{a}_{\text{out}}^\dagger(0) \rangle \langle \hat{a}_{\text{out}}^\dagger(\tau) \rangle + \langle \hat{a}_{\text{out}}^\dagger(0) \rangle \langle \hat{a}_{\text{out}}(\tau) \rangle + \langle \hat{a}_{\text{out}}(\tau) \rangle \langle \hat{a}_{\text{out}}(0) \rangle \right) \quad (35)$$

Using the above, we can finally write the reduced output quadrature correlation function as:

$$\langle \Delta \hat{i}(0) \Delta \hat{i}(\tau) \rangle = \frac{1}{2} \left(\langle \Delta \hat{a}_{\text{out}}(\tau) \Delta \hat{a}_{\text{out}}(0) \rangle + \langle \Delta \hat{a}_{\text{out}}^\dagger(0) \Delta \hat{a}_{\text{out}}^\dagger(\tau) \rangle + \langle \Delta \hat{a}_{\text{out}}^\dagger(0) \Delta \hat{a}_{\text{out}}(\tau) \rangle + \langle \Delta \hat{a}_{\text{out}}(\tau) \Delta \hat{a}_{\text{out}}(0) \rangle + \delta(\tau) \right) \quad (36)$$

Now we can relate the output field operators to intracavity operators via input-output theory:

$$\hat{a}_{\text{out}}(t) = \hat{a}_{\text{in}}(t) + \sqrt{\kappa} \hat{a}(t) \quad (37)$$

The two independent reduced output field correlation functions above can be related to the intracavity field reduced correlation functions (assuming zero temperature):

$$\langle \Delta \hat{a}_{\text{out}}(\tau) \Delta \hat{a}_{\text{out}}(0) \rangle = \kappa \langle \Delta \hat{a}(\tau) \Delta \hat{a}(0) \rangle \quad (38a)$$

$$\langle \Delta \hat{a}_{\text{out}}^\dagger(\tau) \Delta \hat{a}_{\text{out}}(0) \rangle = \kappa \langle \Delta \hat{a}^\dagger(\tau) \Delta \hat{a}(0) \rangle \quad (38b)$$

Finally, the reduced *output quadrature* correlation function can be related to *normal-ordered intracavity* correlation functions as:

$$\langle \Delta \hat{i}(0) \Delta \hat{i}(\tau) \rangle = \frac{1}{2} \delta(\tau) + \frac{\kappa}{2} \left(\langle \Delta \hat{a}(\tau) \Delta \hat{a}(0) \rangle + \langle \Delta \hat{a}^\dagger(\tau) \Delta \hat{a}(0) \rangle + c.c. \right) \quad (39)$$

Recall that we are ultimately interested in the correlation function for the *measured* cavity output quadrature $\hat{I}(t)$, as defined in the main text, which is related to $\hat{i}(t)$ by a downconversion and demodulation step. Fortunately, it is possible to relate measured correlation functions post-filtering directly to output correlation functions prior to filtering⁶:

$$\langle \Delta \hat{I}(0) \Delta \hat{I}(\tau) \rangle = \mathcal{F}(\tau) * \langle \Delta \hat{i}(0) \Delta \hat{i}(\tau) \rangle \quad (40)$$

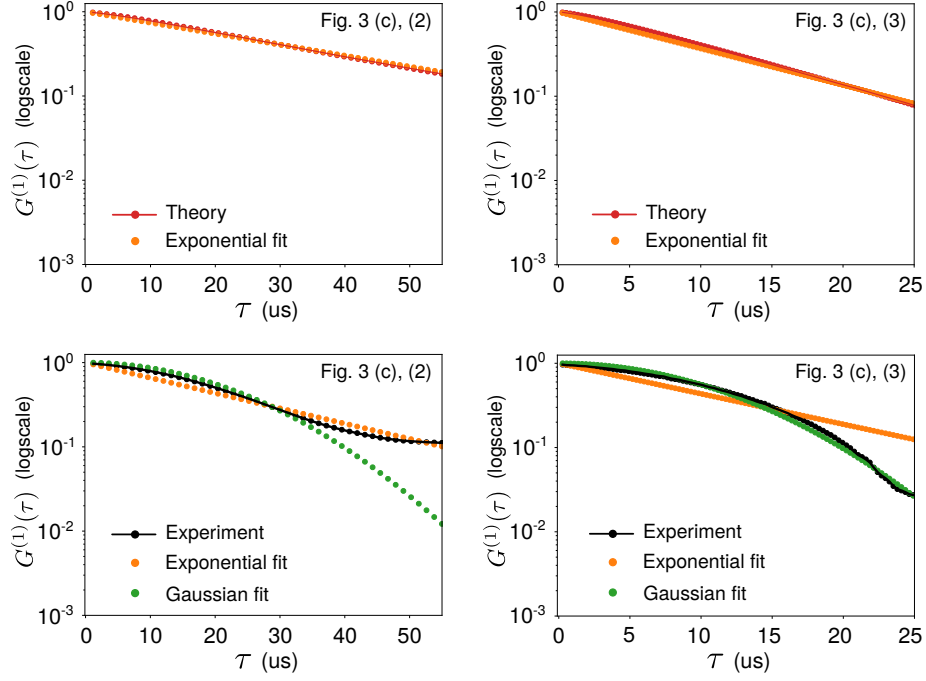


Figure 3. **Non-exponential signatures of phase decoherence.** Top panels: Theoretically calculated decay of $G^{(1)}(\tau)$ peaks as a function of time in logscale (red), and exponential fit (orange). Left and right plots correspond to $G^{(1)}(\tau)$ plotted in Fig. 3(c), positions (2) and (3) respectively of the main text. Lower panels: Same as the top panel, but now showing experimentally obtained decay of $G^{(1)}(\tau)$ peaks as a function of time (black). In addition to the exponential fit (orange), a Gaussian fit is also shown (green), corresponding to contributions from $1/f$ noise.

where $\mathcal{F}(\tau)$ is the composite filter function describing both downconversion and demodulation of the cavity output in the process of measurement, and $*$ indicates the convolution operation. Therefore, to calculate the measured $G^{(1)}(\tau)$ numerically, we first simulate Eqs. (20) and calculate the reduced intracavity correlation functions on the right hand side of Eq. (39), as described in the previous section. This enables us to obtain the output correlation function $\langle \Delta \hat{i}(0) \Delta \hat{i}(\tau) \rangle$. The resulting function is then passed through (i.e. convolved with) the composite filter $\mathcal{F}(\tau)$ to obtain the filtered correlation function, Eq. (40). Finally, employing Eq. (28) yields the required temporal coherence function numerically.

Accounting for the filtering process is important to obtain agreement between the calculated and measured coherence functions, in particular the oscillation frequency which would otherwise differ from the experiment by ~ 100 MHz, the downconversion offset implemented as part of the post-processing. This is particularly evident in comparisons of the measured and numerically calculated $G^{(1)}(\tau)$ shown in Fig. 3(c) of the main text. We also note that the filtering process replaces the somewhat unphysical δ -function contribution in Eq. (39) - arising from the abstract construct of white noise in the cavity output field - with a finite quantity, as is expected for any real detection scheme which possesses a finite bandwidth.

C. Non-exponential signatures in phase decoherence

Generally, the multiplicative nature of the noise described by Eqs. (20) allows for nonexponential decay of the phase coherence captured by $G^{(1)}(\tau)$. Signatures of this can be better seen by extracting the theoretically calculated coherence function peaks and plotting in logscale, as shown in Fig. 3, top panel. However, we find numerically that these nonexponential signatures are minor in the explored parameter regime, as can be seen by the very small deviation from a straight line in logscale. As a result the theoretical decay envelope can be considered to be exponential to a very good approximation.

For the experimentally-obtained coherence function, we have also observed that the decay envelope of the correlation function is not always perfectly exponential. We believe that the observed experimental decay is further complicated by signatures of $1/f$ noise in the system, which would lead to a Gaussian decay envelope. From typical plots of the coherence function peaks as a function of time in logscale, we find that the actual experimental decay envelope is

quite close to exponential, but with some signatures of Gaussian decay. Fig. 3(c) at position 3 is a somewhat atypical example, with more pronounced nonexponential features; we usually find decay characteristics more similar to Fig. 3(c), position 2.

Various techniques have been developed to characterize environmental noise in nature and artificial spin systems [Nature Physics 7, 565(2011), Nature Communications 4, 2337 (2013), Nature Physics, 15, 1123 (2019)]. However, the efficacy of these methods requires sensitivity to the specific frequency distribution of the noise, e.g.: large anharmonicity, thus limiting our ability to remove this noise contribution from the experimental results, or include its effect on the theoretical calculations. Noting further that we generally find the experimental decay to be close to exponential just like the theoretical results in this parameter regime, we have used an exponentially decaying envelope to fit to both theory and experiment results and extract the coherence time. Furthermore, adding a probing system to characterize the environmental noise can also be a future research about quantum coherence in the unstable regime.

IV. DEPHASING IN THE WEAK-DRIVING REGIME

In this work we include the effects of flux noise on the tunable nonlinear mode via the pure dephasing term $\propto \gamma_\varphi$ in the system master equation. In the main text, the impact of pure dephasing on frequency comb coherence was assessed. In this appendix section we consider the influence of pure dephasing in the regime of *weak* driving, far from the instability regions where frequency combs emerge. The qualitative features of this regime can be seen by neglecting the nonlinearity, which then enables an exact analysis of the dynamics. However, being able to access the dynamics in this weak driving regime is not straightforward, as we discuss in the following sections.

A. Exact dynamics

In this linear regime, we find that the full quantum two-mode model can be reduced to a closed set of linear equations for the first and second order moments of the two modes. In particular, the linear system becomes:

$$\frac{d}{dt}\vec{v} = \mathbf{M}\vec{v} + \vec{d} \quad (41)$$

where $\vec{v}$ is the vector of first and second order moments, and $\vec{d}$ describes the drive on the linear mode:

$$\vec{v} = \begin{pmatrix} \langle \hat{a} \rangle \\ \langle \hat{a}^\dagger \rangle \\ \langle \hat{b} \rangle \\ \langle \hat{b}^\dagger \rangle \\ \langle \hat{a}^\dagger \hat{a} \rangle \\ \langle \hat{b}^\dagger \hat{b} \rangle \\ \langle \hat{a}^\dagger \hat{b} \rangle \\ \langle \hat{b}^\dagger \hat{a} \rangle \end{pmatrix}, \quad \vec{d} = \begin{pmatrix} -i\eta \\ +i\eta \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \end{pmatrix} \quad (42)$$

The dynamical matrix $\mathbf{M}$ takes the block form:

$$\mathbf{M} = \begin{bmatrix} \mathbf{M}_1 & \mathbf{0} \\ \mathbf{N} & \mathbf{M}_2 \end{bmatrix} \quad (43)$$

where:

$$\mathbf{M}_1 = \begin{pmatrix} i\Delta_{da} - \frac{\kappa}{2} & 0 & -ig & 0 \\ 0 & -i\Delta_{da} - \frac{\kappa}{2} & 0 & ig \\ -ig & 0 & i\Delta_{db} - \frac{1}{2}(\gamma + \gamma_\varphi) & 0 \\ 0 & ig & 0 & -i\Delta_{db} - \frac{1}{2}(\gamma + \gamma_\varphi) \end{pmatrix} \quad (44a)$$

$$\mathbf{M}_2 = \begin{pmatrix} -\kappa & 0 & ig & -ig \\ 0 & -\gamma & -ig & ig \\ ig & -ig & i\Delta_{da} - i\Delta_{db} - \frac{1}{2}(\kappa + \gamma + \gamma_\varphi) & 0 \\ -ig & ig & 0 & i\Delta_{db} - i\Delta_{da} - \frac{1}{2}(\kappa + \gamma + \gamma_\varphi) \end{pmatrix} \quad (44b)$$

$$\mathbf{N} = \begin{pmatrix} i\eta & -i\eta & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -i\eta \\ 0 & 0 & i\eta & 0 \end{pmatrix} \quad (44c)$$

For convenience, we define the $\hat{I}_j$ and $\hat{Q}_j$ quadratures for mode j as:

$$\hat{I}_j = \frac{1}{\sqrt{2}} (\hat{d}_j + \hat{d}_j^\dagger), \quad \hat{Q}_j = \frac{-i}{\sqrt{2}} (\hat{d}_j - \hat{d}_j^\dagger) \quad (45)$$

where $\hat{d}_j \in \{\hat{a}, \hat{b}\}$ for $j = a, b$ respectively. The above definitions also imply that:

$$\hat{A}_j^2 \equiv \hat{I}_j^2 + \hat{Q}_j^2 = \langle \hat{d}_j^\dagger \hat{d}_j \rangle \quad (46)$$

Removing the coupling ($g = 0$) renders the undriven ($\eta = 0$) dynamical matrix diagonal, and the system decay rates can simply be read off. The linear mode decay rate for $\langle \hat{a} \rangle$ is $\frac{\kappa}{2}$ and for $\langle \hat{a}^\dagger \hat{a} \rangle$ is κ , indicating that the cavity mode experiences no pure dephasing. In contrast, the nonlinear mode amplitude $\langle \hat{b} \rangle$ decays at the rate $(\gamma + \gamma_\varphi)/2$, while the nonlinear mode occupation decays at the rate γ . Therefore in this linear dynamical regime it should in principle be possible to determine the dephasing rate γ_φ by observing the decay of the nonlinear mode quadrature $\hat{I}_b$, in comparison to the decay of $\langle \hat{I}_b^2 + \hat{Q}_b^2 \rangle$.

B. Dephasing measurement in the two-level approximation

When the anharmonicity Λ of the nonlinear mode is large compared to its damping rate γ , the nonlinear mode may be accurately modeled as a two-level system. In this regime, a standard approach to measuring the two relevant decay rates has been readily employed in cQED: by tuning the resulting two-level system frequency far from the cavity mode, a dispersive coupling between the two-level system and the cavity mode is realized, which enables mapping the two-level system state to one of two cavity pointer states. Then, a measurement of the two-level system state is made via a homodyne measurement of the cavity output field. The effects of dephasing in this regime can then be recast into a form familiar in cavity QED: it leads to an additional depolarization of $\langle \hat{\sigma}_x \rangle$ (analogous to $\langle \hat{I}_b \rangle$), without affecting the relaxation rate of $\langle \hat{\sigma}_z \rangle$ (analogous to $\langle \hat{I}_b^2 + \hat{Q}_b^2 \rangle$). A standard Ramsey experiment yields the depolarization rate $(\gamma + \gamma_\varphi)/2 = (T_2^*)^{-1}$ for $\langle \hat{\sigma}_x \rangle$, and by obtaining the relaxation rate $\gamma = T_1^{-1}$ for $\langle \hat{\sigma}_z \rangle$, one can extract the pure dephasing rate $\gamma_\varphi/2 \equiv (2T_\varphi)^{-1} = (T_2^*)^{-1} - (2T_1)^{-1}$.

By mapping the two-level system state to cavity pointer states, measurements in the dispersive regime enable access to two-level system dynamics that occur on longer timescales set by $1/\gamma$, even though measurements are being made of the cavity which evolves on a much shorter time scale $1/\kappa$, since $\kappa \gg \gamma$. The latter condition is in fact necessary to ensure a measurement time that is shorter than the relaxation time of the nonlinear mode, which reduces errors due to unwanted relaxation between the end of any evolution of interest of the two-level system and the conclusion of the measurement of its state. Furthermore, since this approach measures the time-integrated homodyne current to obtain the two-level system dynamics, its temporal resolution is not limited by the DAC (digital-to-analog convertor) that determines the temporal resolution of the obtained homodyne voltage; instead, the temporal resolution is set by the degree of control over microwave pulse generation for the manipulation of the two-level system and the cavity mode.

However, for the devices under study here, we are precisely interested in the weakly-nonlinear regime, where $\Lambda/\gamma \ll 1$. While necessary for the observation of coherent frequency combs, this renders addressing just two states of the nonlinear mode unfeasible, and thus rules out making measurements of the nonlinear mode via a dispersive coupling to the cavity mode. In this case, one must resort to a direct temporal measurement of the relaxation of moments, which we discuss in the next section.

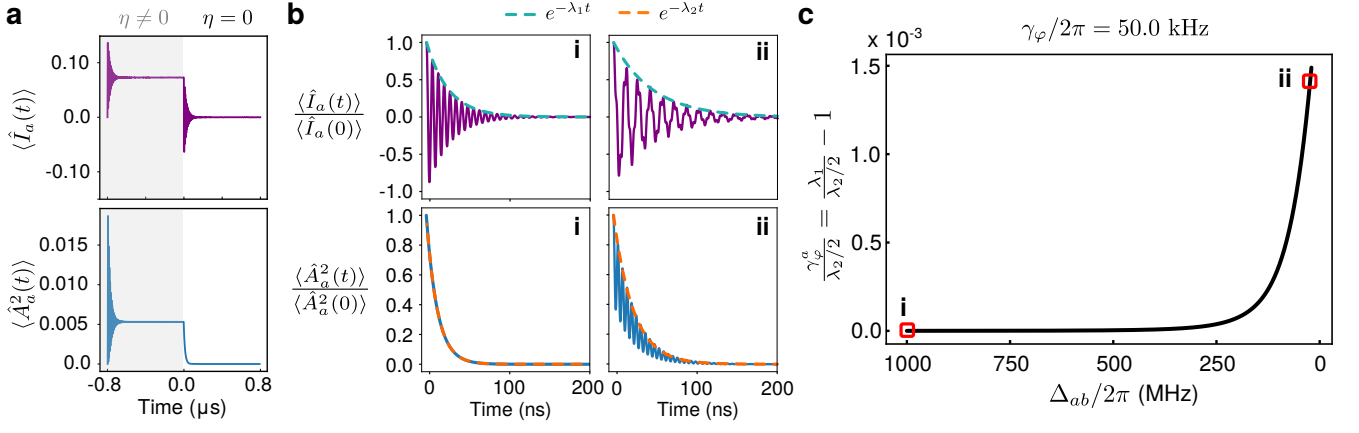


Figure 4. **Theoretical analysis of dephasing in the linear regime via cavity ringdown.** (a) Typical dynamics of cavity quadrature moments under the initialization drive (gray region) and when the drive is turned off (blank region), displaying ringdown. (b) (i) and (ii) show ringdown dynamics for $\langle \hat{I}_a(t) \rangle$ (top panel) and $\langle \hat{A}_a(t) \rangle$ (bottom panel) at the indicated values of detuning Δ_{ab} between the linear and nonlinear mode in (c). Also shown are fits to exponential decays; the top (bottom) panel shows fits with rates determined by the eigenvalue λ_1 (λ_2) for $\langle \hat{I}_a(t) \rangle$ ($\langle \hat{A}_a(t) \rangle$), computed from the dynamical matrix $\mathbf{M}_1$ ($\mathbf{M}_2$) in the ringdown regime ($\eta = 0$). (c) Extracted decay rates from ringdown dynamics as a function of Δ_{ab} . For (b), (c), we choose the actual nonlinear mode dephasing rate $\gamma_\varphi/2\pi = 50.0$ kHz, which is larger than the estimated dephasing rates in the comb generation regimes for both experimental devices.

C. Cavity ringdown method and theoretical simulations

Even when a two-level description of the nonlinear mode is not feasible, Eqs. (41) indicate that under weak driving it should still be possible to observe the effect of pure dephasing on the nonlinear mode moments. To do so, one would ideally like to probe the nonlinear mode dynamics directly, without having to observe the linear mode. This requires effectively decoupling the nonlinear mode from the linear mode (by being detuned far away) while still retaining a coupling to the outside world. However, the 3-D transmon design isolates the nonlinear mode from a direct coupling to the environment, successfully allowing for a much higher- Q nonlinear mode than lumped-element or coplanar waveguide architectures. While this design usefully reduces both the relaxation rate γ and pure dephasing rate γ_φ^7 , it also means that we only have direct access to linear mode quadratures, $\hat{I}_a$, $\hat{Q}_a$.

As such, one is restricted to determining the dephasing rate by monitoring moments of cavity quadratures. The approach one would employ is a ringdown setup⁸: a coherent drive is placed on the system to initialize it to a nontrivial state in phase space, following which the drive is turned off and the resulting ringdown dynamics of the measured first and second order cavity moments recorded as the two-mode system returns to the undriven steady-state. Comparing the rates of relaxation for first and second order moments then enables a calculation of the pure dephasing rate γ_φ .

To explore the feasibility of such an approach, we perform numerical simulations of Eqs. (41) under this ringdown setup. We assume a much larger pure dephasing rate $\gamma_\varphi/2\pi = 50.0$ kHz than estimated for either of our devices, for reasons that will become clear shortly. The typical initialization and ringdown evolution is shown in Fig. 4 (a). The drive η is turned on at $t = -0.8$ μ s, and then turned off at $t = 0$, following which the cavity undergoes relaxation to return to the undriven steady-state. The ringdown dynamics are shown in Fig. 4 (b) for two different detunings between the linear and nonlinear modes, Δ_{ab} . We fit exponentials with decay constants λ_1 , λ_2 to the moments $\langle \hat{I}_a(t) \rangle$, $\langle \hat{A}_a^2(t) \rangle$ (see Eq. (46) respectively, and define the *dephasing rate experienced by the linear mode* as $\gamma_\varphi^a = \lambda_1 - \lambda_2/2$. When the detuning is large compared to the coupling (i), the linear and nonlinear modes are effectively decoupled, so that the linear mode should experience no pure dephasing and γ_φ^a is vanishingly small. With decreasing detuning (ii), the linear and nonlinear modes hybridizes, and the linear mode inherits some dephasing, so that γ_φ^a increases.

The dephasing experienced by the linear mode γ_φ^a as a function of Δ_{ab} is plotted in Fig. 4 (c), scaled by $\lambda_2/2$. While in principle such an approach may be used to extract the pure dephasing rate γ_φ , Fig. 4 (c) brings to light a number of technical difficulties. Firstly, the variation due to pure dephasing is superimposed on the very fast cavity decay rate; the *relative* difference in decay rates $\sim \gamma_\varphi/\kappa$ is therefore very small and difficult to extract experimentally, even though we have assumed a dephasing rate here much larger than those obtained in the main text. In contrast, spectroscopy of the two-level system compares γ_φ directly to γ . Secondly, since this is a direct temporal measurement, its accuracy is limited by the DAC resolution. Small changes in the very short cavity relaxation time are therefore more uncertain.

Both these issues mean that obtaining the pure dephasing rate from direct cavity ringdown measurements under moderate to strong hybridization is likely to be inaccurate. As a result, we instead employ the strategy of obtaining γ_φ in the *nonlinear* regime, in particular within the frequency comb regime. Here, the effect of the bare mode decay rates γ and κ is overcome since the system starts to undergo self-oscillation, as discussed in the main text. Then, the comb coherence is limited entirely by the nonlinearity strength and the pure dephasing rate. By measuring the nonlinearity strength via a pump-probe measurement of the hybridized system, as discussed in Section II B, we are able to use SDE simulations of comb coherence to obtain an estimate of γ_φ .

V. DERIVATION OF FLOQUET EIGENSYSTEM

In this subsection we provide a detailed derivation of the Floquet eigensystem, consisting of the Floquet exponents and left/right eigenvectors, which are employed in the analysis of limit cycle diffusion. We consider the system of N linear first order ODEs given by Eq. (E5) of the manuscript:

$$\dot{\vec{z}} = \mathbf{J}(t)\vec{z} \quad (47)$$

A key constraint of the problem is that the dynamical matrix $\mathbf{J}(t)$ is periodic: $\mathbf{J}(t) = \mathbf{J}(t + T)$. Being a system of N ODEs, it admits N linearly independent solutions which we label $\{\vec{z}_1(t), \vec{z}_2(t), \dots, \vec{z}_N(t)\}$. We can construct a matrix $\mathbf{R}(t)$ with linearly independent columns $\{\vec{z}_i(t)\}$; the resulting matrix also satisfies:

$$\dot{\mathbf{R}}(t) = \mathbf{J}(t)\mathbf{R}(t) \quad (48)$$

Being a linear system, multiplying $\mathbf{R}(t)$ by a constant matrix $\mathbf{K}$ also satisfies the system. In particular, if we define the matrix $\mathbf{V}(t)$ as:

$$\mathbf{V}(t) = \mathbf{R}(t)\mathbf{K} \quad (49)$$

then:

$$\dot{\mathbf{V}}(t) = \dot{\mathbf{R}}(t)\mathbf{K} = \mathbf{J}(t)\mathbf{R}(t)\mathbf{K} = \mathbf{J}(t)\mathbf{V}(t) \quad (50)$$

so that $\mathbf{V}(t)$ is also a solution of the Floquet system. Since $\mathbf{J}(t + T) = \mathbf{J}(t)$, we find:

$$\dot{\mathbf{R}}(t + T) = \mathbf{J}(t)\mathbf{R}(t + T) \quad (51)$$

so that the matrix $\mathbf{R}(t + T)$ also solves the linear system. Combining the above two results, we can relate $\mathbf{R}(t + T)$ to $\mathbf{R}(t)$:

$$\mathbf{R}(t + T) = \mathbf{R}(t)\mathbf{K} \quad (52)$$

Since $\mathbf{K}$ is a constant matrix, it can be obtained from the above relation by setting $t = 0$:

$$\mathbf{K} = \mathbf{R}^{-1}(0)\mathbf{R}(T) \quad (53)$$

By choosing initial conditions such that $\mathbf{R}(0) = \mathbf{I}$, then we simply obtain $\mathbf{K} = \mathbf{R}(T)$, which is a constant matrix referred to as the *fundamental matrix* of the Floquet system. It is obtained by solving Eq. (48) for $\mathbf{R}(t)$ as a function of time $t \in [0, T]$ over a single period T of the classical solution, with the aforementioned initial condition.

Note that the fundamental matrix is in general non-Hermitian; as such we need to consider its complex eigenvalues ρ_i and left/right eigenvectors $\vec{b}_i, \vec{c}_i$ respectively:

$$\begin{aligned} \mathbf{K}\vec{b}_i &= \rho_i\vec{b}_i \\ \vec{c}_i^\dagger \mathbf{K} &= \rho_i^*\vec{c}_i^\dagger \end{aligned} \quad (54)$$

which satisfy the orthogonality relation:

$$\vec{c}_i^\dagger \vec{b}_j = \delta_{ij} \quad (55)$$

If we now define the set of vectors $\{\vec{y}_i(t)\}$:

$$\vec{y}_i(t) = \mathbf{R}(t)\vec{b}_i \quad (56)$$

Substituting the above into Eq. (47), we find:

$$\dot{\vec{y}}_i(t) = \dot{\mathbf{R}}(t)\vec{b}_i = \mathbf{J}(t)\mathbf{R}(t)\vec{b}_i = \mathbf{J}\vec{y}_i(t) \quad (57)$$

which means $\{\vec{y}_i(t)\}$ are solutions to the Floquet system, Eq. (47), as well. This decomposition of the solutions in terms of the eigenvectors of the fundamental matrix also implies:

$$\begin{aligned} \vec{y}_i(t+T) &= \mathbf{R}(t+T)\vec{b}_i = \mathbf{R}(t)\mathbf{K}\vec{b}_i = \rho_i\mathbf{R}(t)\vec{b}_i \\ &= \rho_i\vec{y}_i(t) \end{aligned} \quad (58)$$

Therefore solutions to the Floquet system are in general *not* periodic, unless $\rho_i = 0$. The set of $\{\rho_i\}$ are referred to as *Floquet multipliers*. However, the solutions separated by a period are simply related by a constant. In particular, this enables writing them in the form:

$$\vec{y}_i(t) = e^{\mu_i t} \vec{p}_i(t) \quad (59)$$

where we introduce a set of periodic vectors $\{\vec{p}_i(t)\}$, such that:

$$\vec{p}_i(t+T) = \vec{p}_i(t) \quad (60)$$

Then,

$$\vec{y}_i(t+T) = e^{\mu_i T} e^{\mu_i t} \vec{p}_i(t+T) = e^{\mu_i T} e^{\mu_i t} \vec{p}_i(t) = e^{\mu_i T} \vec{y}_i(t) \equiv \rho_i \vec{y}_i(t) \quad (61)$$

This enables a parameterization of the Floquet multipliers in terms of *Floquet exponents* $\{\mu_i\}$:

$$\rho_i = e^{\mu_i T} \quad (62)$$

We define the periodic vectors $\{\vec{p}_i(t)\}$ as the Floquet right eigenvectors. They are completely determined by the eigenvectors $\{\vec{b}_i\}$ and eigenvalues $\{\rho_i\}$ of the fundamental matrix via:

$$\vec{y}_i(t) = \mathbf{R}(t)\vec{b}_i = e^{\mu_i t} \vec{p}_i(t) \implies \vec{p}_i(t) = e^{-\mu_i t} \mathbf{R}(t)\vec{b}_i \quad (63)$$

To find the equation of motion for the Floquet right eigenvectors $\vec{p}_i(t)$, we can simply take the time derivative of the above, which then yields the equation of motion:

$$\begin{aligned} \dot{\vec{p}}_i(t) &= [-\mu_i + \mathbf{J}(t)] e^{-\mu_i t} \mathbf{R}(t)\vec{b}_i \\ \implies \dot{\vec{p}}_i(t) &= [\mathbf{J}(t) - \mu_i] \vec{p}_i(t) \end{aligned} \quad (64)$$

Similar to the definition of $\vec{y}_i(t)$, we can define solutions $\{\vec{w}_i(t)\}$ in terms of the left eigenvectors of the fundamental matrix $\mathbf{R}(t)$:

$$\vec{w}_i^\dagger(t) = \vec{c}_i^\dagger \mathbf{R}^{-1}(t) \quad (65)$$

Clearly, we have:

$$\vec{w}_i(t+T) = \vec{c}_i^\dagger \mathbf{R}^{-1}(t+T) = \vec{c}_i^\dagger \mathbf{K}^{-1} \mathbf{R}^{-1}(t) = \rho_i^{-1} \vec{c}_i^\dagger \mathbf{R}^{-1}(t) = \rho_i^{-1} \vec{w}_i(t) \quad (66)$$

where we have used the relationship:

$$\vec{c}_i^\dagger \mathbf{K} = \rho_i \vec{c}_i^\dagger \implies \vec{c}_i^\dagger = \rho_i \vec{c}_i^\dagger \mathbf{K}^{-1} \implies \vec{c}_i^\dagger \mathbf{K}^{-1} = \rho_i^{-1} \vec{c}_i^\dagger \quad (67)$$

Then, since $\rho_i = e^{\mu_i T}$, we can write $\vec{w}_i(t)$ in terms of a periodic vector $\vec{q}_i(t) = \vec{q}_i(t+T)$:

$$\vec{w}_i(t) = e^{-\mu_i t} \vec{q}_i^\dagger(t) \quad (68)$$

We analogously define the set of periodic vectors $\{\vec{q}_i(t)\}$ as the left Floquet eigenvectors, which are again completely determined by the eigenvalues and eigenvectors of the fundamental matrix as:

$$\vec{q}_i^\dagger(t) = e^{\mu_i t} \vec{c}_i^\dagger \mathbf{R}^{-1}(t) \quad (69)$$

We can also determine an equation of motion for the Floquet left eigenvectors by taking the time derivative of the above relation. This requires the time derivative of the inverse of $\mathbf{R}(t)$:

$$\begin{aligned}\frac{d}{dt}(\mathbf{R}\mathbf{R}^{-1}) &= \frac{d}{dt}\mathbf{I} = 0 = \dot{\mathbf{R}}\mathbf{R}^{-1} + \mathbf{R}\dot{\mathbf{R}}^{-1} \\ \implies \dot{\mathbf{R}}^{-1} &= -\mathbf{R}^{-1}\dot{\mathbf{R}}\mathbf{R}^{-1} \\ \implies \dot{\mathbf{R}}^{-1} &= -\mathbf{R}^{-1}\mathbf{J}\end{aligned}\tag{70}$$

which then yields the equation of motion:

$$\begin{aligned}\dot{\vec{q}}_i^\dagger(t) &= e^{\mu_i t} \vec{c}_i^\dagger \mathbf{R}^{-1}(t) [\mu_i - \mathbf{J}] \\ \implies \dot{\vec{q}}_i^\dagger(t) &= \vec{q}_i^\dagger(t) [\mu_i - \mathbf{J}(t)]\end{aligned}\tag{71}$$

Finally, we note that the right and left Floquet eigenvectors satisfy the orthogonality relationship:

$$\vec{q}_j^\dagger(t) \vec{p}_i(t) = e^{(\mu_j - \mu_i)t} \vec{c}_j^\dagger \vec{b}_i = \delta_{ij}\tag{72}$$

at all times $t \in [0, T]$, as can be easily found from the definitions of the Floquet eigenvectors, Eqs. (63), (69).

Finally, we show here that provided the Floquet system admits a periodic solution, at least one of the Floquet exponents vanishes⁹. The corresponding Floquet eigenvector is then proportional to the velocity vector of the limit cycle solution. To do so, we begin with Eq. (E2) from the manuscript that describes the limit cycle velocity:

$$\vec{v} = \frac{d\vec{\zeta}_c}{dt} = \vec{A}_c[\vec{\zeta}_c(t)]\tag{73}$$

Differentiating the above with respect to time, we obtain:

$$\dot{\vec{v}} = \sum_j \partial_j \vec{A}_c^i[\vec{\zeta}_c(t)] \vec{v}^j \implies \dot{\vec{v}} = \mathbf{J}[\vec{\zeta}_c(t)] \cdot \vec{v} \implies \dot{\vec{v}} = [\mathbf{J}(t) - 0] \vec{v}\tag{74}$$

where we have used the chain rule since $\vec{A}_c[\vec{\zeta}_c(t)]$ depends on time only via its dependence on $\vec{\zeta}_c(t)$, and we recall that the Jacobian matrix elements are defined via $J_{ij} = \partial_j A_c^i$. When written as the third term, it becomes clear that $\vec{v}$ satisfies the equation of motion for the Floquet eigenvector $\vec{p}_i(t)$ with $\mu_i = 0$, Eq. (63). We label this Floquet exponent with index 0, $\mu_0 = 0$. Clearly, the corresponding eigenvector $\vec{p}_0$ is then proportional to the tangential velocity $\vec{v}$, differing only by a constant that is set by the normalization requirement for the left and right eigenvectors, Eqs. (72).

REFERENCES

- ¹M. Aspelmeyer, T. J. Kippenberg, and F. Marquardt, “Cavity optomechanics,” *Reviews of Modern Physics* **86**, 1391–1452 (2014).
- ²T. J. Kippenberg, A. L. Gaeta, M. Lipson, and M. L. Gorodetsky, “Dissipative Kerr solitons in optical microresonators,” *Science* **361** (2018), 10.1126/science.aan8083.
- ³A. L. Gaeta, M. Lipson, and T. J. Kippenberg, “Photonic-chip-based frequency combs,” *Nature Photonics* **13**, 158–169 (2019).
- ⁴P. D. Drummond and P. Deuar, “Quantum dynamics with stochastic gauge simulations,” *J. Opt. B: Quantum Semiclass. Opt.* **5**, S281–S289 (2003).
- ⁵C. W. Gardiner and M. J. Collett, “Input and output in damped quantum systems: Quantum stochastic differential equations and the master equation,” *Physical Review A* **31**, 3761–3774 (1985).
- ⁶M. P. da Silva, D. Bozyigit, A. Wallraff, and A. Blais, “Schemes for the observation of photon correlation functions in circuit QED with linear detectors,” *Physical Review A* **82**, 043804 (2010).
- ⁷H. Paik, D. I. Schuster, L. S. Bishop, G. Kirchmair, G. Catelani, A. P. Sears, B. R. Johnson, M. J. Reagor, L. Frunzio, L. I. Glazman, S. M. Girvin, M. H. Devoret, and R. J. Schoelkopf, “Observation of high coherence in josephson junction qubits measured in a three-dimensional circuit qed architecture,” *Phys. Rev. Lett.* **107**, 240501 (2011).
- ⁸O. Maillet, F. Vavrek, A. D. Fefferman, O. Bourgeois, and E. Collin, “Classical decoherence in a nanomechanical resonator,” *New Journal of Physics* **18**, 073022 (2016).
- ⁹H. Haken, “At least one Lyapunov exponent vanishes if the trajectory of an attractor does not contain a fixed point,” *Physics Letters A* **94**, 71–72 (1983).