n-photon bound states in the continuum for strong intensity squeezing and
deterministic stabilization of large photonic Fock states

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Abstract

Large non-Gaussian states of light are a highly coveted resource in quantum science and technology. An important example of such states are $n$-photon states of light (Fock states), which, besides being the most fundamental states of the quantized radiation field, are theoretically believed to be valuable for many tasks including metrology, communication, simulation, quantum state generation, and information processing. However, the deterministic creation and stabilization of even approximate large-number ($n \geq 2$) Fock states at optical frequencies is a long-standing open problem. Here, we present a fundamental new effect in nonlinear photonic systems — called $n$-photon bound states in the continuum — which can be applied to deterministically create large Fock states, as well as very highly intensity-squeezed states of light. The effect is one in which destructive interference gives a certain quantum state of light an infinite lifetime, despite coexisting in frequency with a radiative continuum. For Kerr nonlinear systems, that state is an $n$-photon (Fock) state of a particular and tunable $n$. We develop the theory using experimentally-realizable examples (showing examples of producing 30-photon Fock states, and states with very large intensity squeezing (10 dB)), and we describe schemes to pump and probe the generated quantum states. The effect requires only Kerr nonlinearity and linear frequency-dependent (non-Markovian) dissipation, and is in principle applicable at any frequency. The theory and concepts are also immediately applicable to nonlinear bosons besides photons, and should be implementable in many other fields (e.g., superconducting circuits, magnonics, phononics). Further, we expect the broader possibility of using engineered nonlinear photonic structures — with highly tailorable radiation loss — as a basis to create many of the other quantum photonic states that have traditionally been hard to generate.
Much of the current focus in quantum optics is on the generation and application of quantum states of light, such as single-photons, entangled photon pairs, cluster states, and quadrature-squeezed light \[1\text{–}6\]. Such states enable the extension of important applications — such as information processing, simulation, precision measurement, and communication — beyond the limits imposed by classical physics. While these states are already useful, a transformation of the quantum landscape is expected if macroscopically non-classical (e.g., non-Gaussian) states of light, which offer a unique degree of quantum advantage, can be deterministically realized and manipulated.

As a concrete example, consider the case of large optical Fock states \(|n\rangle\), which are eigenstates of the quantized radiation field with a perfectly defined number of photons. These states are — especially in optics — currently very difficult to realize, and despite this, have long been identified by the quantum community as an important state to access. For example, such states have long been considered for quantum metrology because they have a perfectly defined intensity that would enable measurements without shot noise \[7\text{–}9\]. One of the earliest proposed applications along these lines was to use an \(n\)-photon state in a cavity as an extremely sensitive sensor of small vibrations \[10\]. They are also considered valuable in simulation and information processing tasks. For example, in the microwave domain, Fock states of LC resonators have already been used for quantum chemistry tasks, such as simulating vibronic spectra of molecules \[11\]. In particular, high-order Fock states of the resonator can be mapped to highly electronically and vibrationally excited states of a molecule, enabling simulation of excited-state spectra, which is traditionally challenging. Another high-profile application of large Fock states for simulation is in the implementation of quantum algorithms such as boson sampling (or gaussian boson sampling) \[12\text{–}17\]: a “modest” (multimode) Fock state of even 100 photons enables computations of matrix permanents at least fifteen orders of magnitude larger than could be handled by even the largest supercomputers today \[18\]. Fock states are also important “resource states,” providing a vehicle to generate other desirable quantum states. For example, small Fock states can be used to generate (squeezed) Schrödinger cat states \[19\], and could further be used to generate states such as displaced Fock states, GKP states, etc. Beyond all of these “applications”-oriented arguments, there is also a clear case for the interest in these states on purely fundamental grounds.

For the reasons above and many more, the problem of generating Fock states has generated much attention at both microwave and optical frequencies. At microwave frequencies, it is currently possible to deterministically, and with high fidelities (> 90%), produce Fock states of up to
15 photons. These Fock states are generated in microwave resonators via quantum optimal control algorithms that simultaneously employ external driving of the cavity by microwave pulses and superconducting transmon qubits [20]. Fock states have also been generated in microwave cavities by strongly coupling them to transmon qubits that are repeatedly pumped to inject photons into the cavity at deterministic times [21]. Even older work made use of Rydberg atoms strongly coupled to microwave cavities in order to generate low-order Fock states using principles such as the one above, as well as quantum feedback protocols [22–24]. Such Rydberg atom-cavity interactions form the basis for new theoretical proposals to extend microwave Fock states to higher photon numbers [25, 26].

In optics, the situation is markedly different. As there is no real analogue of superconducting qubits for optics, the techniques above cannot be applied to generate photonic Fock states. In fact, it is currently a significant challenge to deterministically produce Fock states in optics even with order larger than \( n = 2 \). This lies in stark contrast to single photon Fock states, where the capabilities are much more mature [27]. In general, proposals for generating the Fock state \(|n\rangle\) \((n \geq 2)\) are either non-deterministic (meaning the state is generated with random order, or at random times [28–31]), or require intermediate creation of hard-to-generate states (such as atomic Dicke states [32–34]).

The key result of our work is a new dissipative effect for photons that can be applied to deterministically generate Fock states of arbitrary order, which can be applied at optical frequencies. More precisely:

1. We develop a general quantum theory of nonlinear leaky resonators with frequency-dependent outcoupling, describing the dynamics of dissipation in photonic structures such as those shown in Fig. 1a.

2. We present a new effect that arises from this theory: \( n \)-photon bound states in the continuum \( (\text{BICs}) \). In this effect, a photonic resonance can have \emph{infinite} lifetime, conditioned on the resonance being in the state \(|n\rangle\). This leads to a situation where a Fock state is \emph{stable and dissipationless}, even in the absence of pumping.

3. We show how these \( n \)-photon BICs enable deterministic generation of optical Fock states and highly intensity-squeezed states of light. We propose nonlinear photonic architectures to realize these \( n \)-photon BICs and develop a protocol to deterministically create Fock states using them: by injecting a short pulse from a laser into the nonlinear cavity, and letting the light decay freely from that cavity, Fock and highly squeezed states are naturally produced.
FIG. 1: Quantum nonlinear dissipation based on leaky modes with frequency-dependent outcoupling, and $n$-photon BICs. (a) Examples of photonic structures, which, when Kerr nonlinearity is present, realize a resonance with a photon-number-dependent loss rate (a nonlinear loss) [pictured generically in bottom right]. (b) Nonlinear loss for a resonance coupled to a waveguide terminated by a backreflector. Due to destructive interference between two paths for radiation (blue and orange), it is possible for the coupling of the resonator to the waveguide to vanish for a certain resonator frequency, leading to a loss minimum (a quasi-BIC) (top inset). Due to the Kerr nonlinearity, the resonator frequency is a function (linear) of the intracavity photon number (bottom inset), leading to (b) a photon-number dependent loss for the cavity. In the case of a true BIC, where the lifetime of the resonance becomes infinite, the loss becomes zero for some photon number $n_0$ — what we call a structure with an $n_0$-photon BIC. For a quasi-BIC, the loss reaches a minimum, rather than a zero, for that same photon number. Changing the detuning of the resonator from the BIC frequency changes the zero-loss-photon-number — here this is shown for three different detunings of roughly $\Delta_{1,2,3} = (0.0094, 0.043, 0.093)\gamma$. The parameters used in this figure are: $\omega_a = 1.47eV$, $\Delta_\omega = 10^{-2}\omega_a$, $T^{-1} = 10^{-3}\omega_a$, $\beta = 5 \times 10^{-6}\omega_a$. Detailed experimental justification of parameters is provided in the discussion of Fig. 3, as well as in the SI, Section IV.

1. THEORY AND NEW EFFECTS

We begin by describing the new effects and the intuition behind them. The physics we are interested in is that of radiation loss in photonic resonators with Kerr nonlinearity. We consider a special type of photonic resonator, of which Fig. 1a shows three instances. What all three structures have in common is that they can have very high $Q$-resonances due to destructive interference
between two or more “paths” for light in the resonator (labeled \(a\)) to escape to the continuum.

Such systems, where high-\(Q\) arises from destructive interference between radiation loss pathways, have been the subject of many recent works in the photonics community, under names like *bound states in the continuum* (BICs) [35], *quasi-bound states in the continuum* (quasi-BICs) [36, 37], and *Fano resonances* [38]. Characteristic to these high-\(Q\) resonances is that their \(Q\)-factor sensitively depends on geometrical and material parameters (e.g., feature size, index of refraction, photon wavevector), achieving a large maximum for some value of the parameters. This maximum occurs for the geometrical parameters which lead to opposite phases for the two leakage paths (e.g., blue and orange paths in the first and second systems of Fig. 1a). When this happens, the \(Q\) is limited only by what we’ll call “external” losses (e.g., absorption, scattering). As a point of terminology, we will generally use the term BIC to refer to “cancellation-induced” high-\(Q\) resonances, in keeping with previous usage of the phrase [36, 39, 103].

Now, we consider the quantum optics of dissipation due to light leakage in structures with BICs. At first glance, it seems that the quantum optics of radiation loss of these nonlinear high-\(Q\) resonances would just be governed by the textbook theory of dissipation in nonlinear high-\(Q\) resonators [40–42]. The conventional theory has been applied extensively for over forty years, predicting a variety of effects which have been observed, such as optical bistability (in the classical domain [43]), dissipative phase transitions [44], and modest amplitude squeezing (antibunching of light) in the cavity mode [45–47]. This arguably natural assumption, that it is only the value of \(Q\) that matters, is surprisingly not correct.

Consider what happens when we add Kerr nonlinearity to the resonance \(a\) (for example, when the cavity in Fig. 1a (left) has third-order optical nonlinearity). In that case, the refractive index depends on intensity, or equivalently, the number of photons, \(n\), in the cavity. Then, as the intracavity photon number changes, so does the resonance frequency \(\omega_a\) [48], and also the relative phases of leakage paths in Fig. 1a. In such a case, the \(Q\)-factor depends on the number of photons in the cavity: \(Q = Q(n) = Q(\omega_a(n))\); it is the composition of the dependence of \(Q = Q(\omega_a)\) on the resonator frequency (amplitude-phase coupling) and the dependence of the resonator frequency \(\omega_a = \omega_a(n)\) on the photon number (because of the Kerr nonlinearity). This system therefore has nonlinear loss: a decay rate \(\kappa(n)\) that depends on the number of photons in \(a\).

We will show that Kerr nonlinear BICs realize a very unique form of nonlinear loss for photons compared to well-known forms of nonlinear loss, like saturable or multiphoton absorption. This is illustrated in Fig. 1b, where we plot the nonlinear loss rate as a function of photon number.
(parameters will be explained in the section on proposed experimental realizations). As anticipated from the arguments above, there is a maximum in $Q$ (minimum in $\kappa$) as a function of photon number. For the case of an ideal BIC (infinite lifetime), there is a special photon number $n_0$ for which the loss is exactly zero $\kappa(n_0) = 0$. We refer to the structure as having an $n_0$-photon BIC, because when it has $n_0$ photons, the resonance lifetime is infinite.

It is clear that such a system might facilitate creation of $n_0$-photon Fock states. Suppose we populate the system with an average number of photons $\bar{n}$ greater than $n_0$ (for example, a coherent state with a mean and variance of $\bar{n}$ photons. Then the system will decay until it has $n_0$ photons exactly (with zero variance): the system will stay stuck in the state $|n_0\rangle$ because the loss rate is zero in that state, and the photons have nowhere to go at that point; the variance (fluctuations) have disappeared entirely. By continuity, even when the BIC is imperfect due to some finite external loss $2\kappa_i$ (see Fig. 1b), one expects the variance in photon number to rapidly drop (well below the mean), leading to intensity-squeezed (nonclassical) light. That said, if the background loss is too high, these quantum optical effects will be mitigated, as the nonlinear loss looks basically linear (intensity-independent, see black curves of Fig. 1b), converging to textbook dissipation theory.

In what follows, we show concrete examples and applications of this concept, showing that it remains interesting in realistic cases. The results arise from a general theory that we develop, of dissipation in nonlinear photonic structures with frequency-dependent (“non-Markovian”) outcouplings. First, we state the key results of this theory (see SI for detailed derivations). The family of systems to which our theory immediately applies is a driven Kerr nonlinear resonance, coupled to $N$ reservoirs (continuua; radiative or absorptive [104]) leading to dissipation of the resonance. For this class of systems, the Hamiltonian takes the form:

$$H/\hbar = \omega_a a\dagger a + \beta \omega_a a\dagger^2 a^2 + \alpha(t) a + \alpha^*(t) a\dagger + \sum_{i=1}^{N} \int \frac{d\omega}{2\pi} \omega s_i^\dagger(\omega)s_i(\omega)$$

$$+ \sum_{i=1}^{N} \int \frac{d\omega}{2\pi} i \left( K_{c,i}(\omega)s_i(\omega)a\dagger - K_{c,i}^\dagger(\omega)s_i^\dagger(\omega)a \right).$$

(1)

Here, $a$ is the annihilation operator of the resonance with frequency $\omega_a$. The Kerr nonlinearity of the resonance is well-known to manifest as the second term in the Hamiltonian [40, 42], leading to a “photon-number-dependent resonance frequency.” Given $n$ photons in the resonator, the energy to add another is $\hbar \omega_{n+1,n} \equiv \hbar \omega_a (1 + 2\beta n)$, where $\beta$ is a (dimensionless) nonlinear coefficient. The coherent-drive strength is $\alpha(t)$ and is not restricted to be monochromatic. The continuum modes
Dynamics of quantum statistics of a cavity undergoing nonlinear radiation loss. Dynamics of photon number mean and variance for different initial states. Poissonian states (black diagonal) flow towards heavily number-squeezed (sub-Poissonian) states with far less noise (variance) than their mean. For long times, and for a true BIC, rather than decaying to vacuum, they tend to Fock states (here, with $n_0 = 10$). For starting states with less than 10 photons, they rapidly decay to vacuum states. Initial Fock states (points on bottom $x$-axis) start to have added noise (as usual), but then eventually experience a noise decrease and tend to $n = 10$ Fock states. Parameters are the same as in Fig. 1b. The state indicated by the purple circle is a mixed state in which there is some probability of the system being in a Fock state and some of it being in vacuum, which occurs when the initial probability distribution of the cavity photon number is nonnegligible both above and below $n_0 = 10$.

are described as usual by a set of harmonic oscillators [49]: for the $i$-th reservoir, the continuum modes are labeled by their frequency $\omega$ and annihilation operator $s_i(\omega)$.

The resonance couples to the continuum through the last term in the Hamiltonian with coupling coefficient $K_{c,i}(\omega)$. This $K_{c,i}$ is the in-coupling function from the $i$-th reservoir: given a monochromatic input (at frequency $\omega$) from reservoir $i$ into the resonator, the classical amplitude of the resonance, denoted $a(\omega)$, is proportional to $K_{c,i}(\omega)$. It can be directly calculated from numerical electromagnetic simulations, for example, by launching an input wave at a resonance in a simulation and examining the frequency-domain amplitude of the resonance. This makes our theory ab initio and broadly applicable. The in-coupling function can also be extracted from
temporal coupled mode theory models \[43, 50, 51\], which are well-known to accurately describe many widely-used photonic architectures. In the SI, we provide explicit forms of \(K_c(\omega)\) for a few common photonic architectures that have BICs (the BIC is encoded in a zero/minimum of \(K_c(\omega)\) as a function of frequency).

The Hamiltonian of Eq. (1) is exact, but not generally solveable. Now, we employ the sole approximation of our theory: that the bandwidth of the reservoirs, \(\Delta \omega_i\) is much larger than the inverse “response” timescale of the reservoir \(T^{-1}\). The physical quantity which sets \(\Delta \omega_i\) and \(T^{-1}\) varies from architecture to architecture, but it very frequently holds. This “semi-Markovian” approximation \([41, 52]\) yields an equation of motion for the reduced density matrix of the cavity, denoted \(\rho\). It is (see SI):

\[
\dot{\rho} = -i[H_K + H_{\text{drive}}, \rho] + D[\rho]
\]

(2)

where \(H_K + H_{\text{drive}} = \omega_a a^\dagger a + \beta \omega_a a^\dagger a^2 + \alpha(t)a + \alpha^*(t)a^\dagger\) is responsible for the conservative parts of the evolution of the resonance, and the dissipator \(D\) is defined through its matrix elements as (\(m, n\) are Fock states):

\[
\langle m | D[\rho] | n \rangle = -(m K_l(\omega_{m,m-1}) + n K_l^*(\omega_{n,n-1})) \rho_{m,n} + \sqrt{(m + 1)(n + 1)} (K_l(\omega_{m+1,m}) + K_l^*(\omega_{n+1,n})) \rho_{m+1,n+1}.
\]

(3)

The function \(K_l(\omega)\) is the “loss function” or “frequency-dependent loss” of the cavity, and is directly connected to the incoupling function by a Kramers-Kronig relation, as:

\[
K_l(\omega) = \sum_i K_{l,i}(\omega), \text{ with } K_{l,i}(\omega) = i \int \frac{d\omega'}{2\pi} \frac{|K_{c,i}(\omega')|^2}{\omega - \omega' + i\eta},
\]

(4)

where \(\eta\) is an infinitesimal. The relation of Eq. (3) enforces the intimate connection between leakage and incoupling. Equations (2-4) summarize our quantum theory of nonlinear outcoupling: they tell us that the evolution of the quantum state of a resonance, including photon probabilities, field correlations \((g^{(1)})\), intensity correlations \((g^{(2)})\), and so on – are strongly controlled by the photon number \((n)\) dependence of quantities like: \(K_l(\omega_{n+1,n}) = K_l(\omega_n(1 + 2\beta n))\), whose form can be controlled by engineering the outcoupling of light in a cavity. The theory here differs from the standard theory of leaky resonators, which typically makes a “white noise” approximation, neglecting the frequency-dependence of the continuum coupling (e.g., by evaluating it at a single reference frequency) \([105]\).

Let us now discuss what dictates the value of \(n_0\) which stabilizes the Fock state. The \(n\)-photon
BIC condition, in the language of our theory, is

$$\text{Re } K_{l}(\omega_{n_0,n_0-1}) = 0. \quad (5)$$

Suppose the zero of the loss function $K_{l}(\omega)$ occurs at some frequency $\omega_0$ (the BIC frequency). Then, we may expand $K_{l}$ around $\omega_0$ as $\text{Re } K_{l}(\omega) \approx c_2(\omega - \omega_0)^2$ [106]. From Eq. (5), we have that

$$n_0 = \frac{\Delta_0}{2\beta} + 1, \quad (6)$$

where $\Delta_0 \equiv \omega_0 - \omega_a$ is the detuning of the linear resonance from the BIC frequency. This simple equation shows that the order of the Fock state can be controlled by simply tuning the resonator frequency (see Fig. 1b), and that there are discrete detunings that lead to Fock states. This equation also reveals that larger single-photon nonlinearities ($\beta$) and smaller detunings ($\Delta_0$) lead to smaller photon numbers, while smaller single-photon nonlinearities (characteristic of “bulk material” nonlinearities) lead in principle to Fock states at larger photon numbers. The larger Fock states are more fragile, but we will see that even so, intense and extremely squeezed light can be realized — beyond what is typically achievable in resonators through normal nonlinear loss — and thus this regime is still very interesting.

The dynamics of various quantum states undergoing the nonlinear radiation loss of Fig. 1b are illustrated in Fig. 2, in a “phase space representation” where the phase space variables are mean and variance. Each line indicates a trajectory of some initial state: we show the dynamics of a set of initially “Poissonian” states with shot noise photon number fluctuations, as well as initial Fock states. Let us consider the dynamics of the Poissonian states with mean greater than $n_0 = 10$. For a true BIC (top panel), the trajectories move towards a Fock state of order 10 (blue circle in top panel). States with mean sufficiently below $n_0 = 10$ decay to vacuum, as expected. Intriguingly, initial states that have significant probability to be both at $n < 10$ and $n \geq 10$ lead to a “mixed” state that has some probability of being in vacuum and some of being a Fock state (see trajectories terminating for example at the purple point in Fig. 2).

It is worth pausing to emphasize the striking-ness of this effect. In all known photonic systems, in the absence of a driving field, the only stable state is the vacuum state with zero photons. All finite photon-number states dissipate. In the systems examined here, all but two states dissipate: the zero photon state, and the $n_0$-photon Fock state (the “mixed” state in Fig. 2 is a manifestation of that bistability). This type of bistability differs from conventional bistability in Kerr systems in that: with no driving amplitude, there is only a single stable state (vacuum) [43]. Further, in
conventional bistability, Fock states are not steady states [40].

In the presence of background losses, the BIC becomes imperfect and the bifurcation of trajectories in the top panel of Fig. 2 softens, as the state at \( n = 10 \) is no longer stuck – it slowly leaks and progress towards the vacuum state. This is shown in the bottom panel for \( \kappa_i = 10^{-7} \omega_a \) (an external \( Q_i = 5 \times 10^6 \)). As can be seen, the blue trajectories start by initially moving well-below the black line, indicating that they become highly sub-Poissonian (number-squeezed). For example, if the initial state is Poissonian with a mean of 40 photons, at some point in time (about 300 ps in this example), the state turns into one with roughly 20 photons and an uncertainty \( \Delta n = 2 \), corresponding to a resonator state with fluctuations about 80% (about 7 dB) below the shot noise level, which is a very high degree of squeezing for a mesoscopic state like this one.

II. PROSPECTS FOR EXPERIMENTAL REALIZATION

We have thus far addressed the physical principles behind the nonlinear loss, and why this generates Fock and squeezed states. We will now discuss concrete physical systems to implement the physics (and expected numbers), a protocol to deterministically prepare these states, and experimental signatures of them. The theory developed above is quite general, being applicable to any Kerr nonlinear oscillator coupled to one or more continua with frequency-dependent couplings. As Kerr nonlinearities appear in many physical systems (photonics and beyond), the approach we take is to choose one example system, and show the expected numbers from end-to-end (from pumping to detection), and provide a schematic discussion of other systems near the end.

The example we choose is meant to show how to generate close approximations of Fock states with large photon numbers (\( n_0 \) on the order of 10 or so, with \( \Delta n \lesssim 1 \)). Such platforms, as per Eq. (6), should be realized using sizeable single-photon nonlinear strengths \( \beta \). Fig. 3a illustrates such a system, formed by coupling excitons to a resonator which is then coupled to a waveguide [53]. The parameters for the resonator-waveguide system are very similar to those in [39]. The coupling between exciton and resonator leads to exciton-polaritons. Such excitations are the subject of many recent experiments (not limited to, but including [44–47, 54]). From experiments, these exciton-polaritons are known to: (a) reach strong coupling, with measured Rabi splittings exceeding the decay rate by 1-2 orders-of-magnitude (b) be well-described by a driven single-mode Kerr Hamiltonian (with damping), as in Eq. (1), with the lower polariton serving an “effective photon” (or photonic quasiparticle [55]) and (c) present the strong nonlinearities that
**FIG. 3:** **Deterministic creation of large Fock states based on a pump-and-ringdown protocol in an example system: conversion of coherent states into Fock states.**

(a) A system to realize BICs and sizeable Kerr nonlinearity simultaneously: a photonic crystal slab with a (terminated) defect waveguide and a defect resonator, coupled to a semiconductor quantum well. (b) Pump-and-ringdown protocol for deterministic creation of Fock states. By sending in a short pulse, an initial coherent state of polaritons can be loaded. After the pump pulse, the system is left to evolve without any external pumping. This natural “ring-down”, following Eq. (2), leads to the coherent state turning into a Fock state, as illustrated through the Husimi Q functions for different times. (c) Evolution of the photon probabilities in time for the same initial state (a coherent state of 50 photons). The Fock state order can be tuned by changing the resonator frequency, as can be seen from the different panels, where we show tuning the Fock state order between 10 and 30 with near unit fidelity starting from a coherent state. Note that parameters for which \( n_0 \) in Fig. 2 is not an integer lead to a “failed” Fock state. Here, \( \Delta_{1,2,3,4} \approx (0.0305, 0.0300, 0.0200, 0.0095) \gamma \). (d) Time-dependent photon-number squeezing for different levels of external linear loss. Importantly, even for realistic linear losses, it is possible to achieve substantial number squeezing, e.g., about 10 dB. For very large linear background loss (brown line), the result is indistinguishable from shot noise (0 dB squeezing). Parameters are the same as in Figs. 1b and 2. This figure further assumes an exciton-photon Rabi frequency of 1.8 meV, a nonlinear exciton-exciton energy of 20 \( \mu \text{eV} \cdot \mu \text{m}^2 \), and an exciton-photon hybridization coefficient of 0.5. The input pulse preparing the coherent state is assumed to be 10 fs in duration and contain about 1000 photons (as from an attenuated pulsed laser).
are needed. The nonlinearities already present \( (\beta \sim 10^{-5} \omega) \) are already much larger than what is available in diffraction-limited microcavities of bulk nonlinear optical materials such as GaAs and GaP. In recent experiments, exciton-polaritons in microcavities have also been shown to present the characteristic optical bistability of Kerr systems, with concomitant squeezing \([45]\). Even more recently, it has been shown that polariton-polariton interactions are now strong enough to lead to antibunching of light, with promising prospects for photon blockade (or more appropriately, polariton blockade) upon improvement of the exciton lifetime (which in those experiments, was on the order of 10 ps) \([44, 46]\). The most recent experiments have even managed to couple exciton polaritons in GaAs to optical bound states in the continuum in one-dimensionally periodic gratings, forming polariton BICs with measured lifetimes approaching 1 ns \([54]\) (similar to a lower-bound associated with exciton dispersion, discussed for a different material platform \([53]\)). All in all, this suggests the use of such exciton-polaritons as a promising platform to realize the physics we describe here — motivating its choice as our main example.

We now describe a protocol for “loading” Fock and highly squeezed states. It is illustrated in Fig. 3b: we start by injecting a short pulse through the resonator. This pulse is short compared to the timescale of the nonlinearity and the dissipation. Its purpose is to load an initial state with mean number of photons greater than \( n_0 \). The state can be somewhat arbitrary. Then, after the pulse passes, the dynamics are governed by the nonlinear dissipation of Eqs. (2-4). The simulated dynamics of the overall quantum state of the cavity, are visualized in Fig. 3b through the Husimi \( Q \) functions \( Q(\alpha) = \langle \alpha | \rho | \alpha \rangle / \pi \), where \( | \alpha \rangle \) is a coherent state. Initially, the state is vacuum. After the pump pulse, it is in a coherent state. The subsequent decay leads to a stretching of the \( Q \) function in phase (angular direction), while the overall photon number decreases (this meniscus shape is well-documented in early works on Kerr squeezing \([56]\)). Then, after long times, the system approaches a Fock state. Fig. 3(c) shows the simulated photon probabilities, affirming the intuition of Fig. 1a. The main observation is that as a function of time, the photon noise of the coherent state condenses: in other words, the nonlinear loss leads to photon number squeezing, until eventually, for longer times, the distribution converges to a Fock state (here, at times on the order of several hundred ns). But already, for shorter times (e.g., 700 ps or 7 ns), there is significant photon number squeezing (roughly 10 dB) and strong fidelity enhancement for Fock state generation. The limiting Fock state order depends on the detuning. For detunings which cause the nonlinear loss to vanish at an integer photon number, the Fock state is produced with fidelity 1. Meanwhile, for detunings for which the loss zero is non-integral, the resonator does not
reach a Fock state, as there is no photon number for the distribution to get stuck at, leading to a “failed” Fock state.

Fig. 3d shows the effect of external (linear) losses on the intensity squeezing (it is the primary limitation to consider). The external loss is taken to come from the nonradiative decay of excitons. Fig. 3d shows the expected time-dependent photon-number-squeezing in the resonance (the lower-polariton) for different levels of external nonlinear loss. The main conclusions are that for external $Q$-factors in the range of $10^5 - 10^6$, there is substantial squeezing, even beyond the “3 dB limit.” A sufficiently large linear loss will fully mask the nonlinear loss (e.g., at $Q_i = 5 \times 10^3$), such that there is no noticeable deviation from what is expected from linear loss (shot noise stays as shot noise). The peak squeezing occurs around a few 100 ps. The current state of the art in exciton polaritons is on the order of 400 ps ($Q_i \approx 10^6$ — near the green curve). This is about an order of magnitude away from a discussed bound associated with exciton dispersion (on the order of 1 ns exciton lifetime, corresponding to the orange curve), discussed for a different material platform [53]. In other words, even within optics, we already expect the new nonlinear dissipation we developed to be testable.

**Experimental signatures.** We now address the question of how to detect the generated quantum states. In Fig. 4, we show two possibilities: one standard technique, based on measuring second-order correlations, and one recently demonstrated technique which measures more directly the intracavity quantum state using a near-field probe (a free electron).

Second-order correlation measurements have been extensively used to characterize quantum statistics of a resonance, even in systems of excitons coupled to microcavities (for example, one sends the light emitted from the resonator into a Hanbury-Brown-Twiss interferometer) [44, 46, 47]. Fig. 4a shows the expected second-order correlations (at zero time-delay) as a function of the measurement time, as well as the external losses. The Fock state of a given order will lead to $g^{(2)} = 1 - 1/n$, so a Fock state of order $n = 10$ will causes $g^{(2)}$ to approach 0.9. In the presence of external loss, the minimum $g^{(2)}$ increases towards 1. The above reveals a disadvantage of second-order correlation approaches. In particular, a $g^{(2)}$ between 0.9 and 1 could indicate a possible Fock state, or some other generic antibunched light state. Such techniques also appear unsuitable for larger Fock orders and intense squeezed light, where the deviation from unity would be much smaller than one.

Recently, a technique has been demonstrated that is capable of measuring the quantum statistics of the intraresonator field with higher granularity. It is based on a new type of near-field photon
FIG. 4: **Experimental signatures of intracavity Fock states.** (a) Far-field detection based on outcoupling light from a weak Markovian output port (e.g., vertically scattered light, or an additional outcoupling waveguide) and corresponding second-order correlations at zero time-delay, as measured by a Hanbury-Brown-Twiss interferometer. Colors in plot correspond to same external losses as in Fig. 3d. (b) Near-field detection based on quantized energy exchange between a free electron and the resonance via stimulated emission and absorption. Electron energy loss and gain probabilities are measured by an energy spectrometer for the electron, leading to energy losses which differ for Fock versus coherent states. Plots show energy loss probabilities for Fock and coherent states for three values of the electron-photon coupling strength (justification for values in SI) - indicating clear differences between Fock and coherent states.

detection technique, referred to as photon-induced near-field electron microscopy (PINEM). The basic theoretical description and experimental implementations are discussed in Refs. [55, 57–60]. The idea is illustrated in Fig. 4b: an energetic electron (typical kinetic energy $E_0 \sim 100$-200 keV) grazes past the sample, interacting with the evanescent field of the resonance (it can also pass directly through the sample). The electron can then undergo absorption and stimulated emission induced by the photons in the resonance, leading to energy gain and loss. Due to the short duration of interaction (< 1 ps), the electron probes the instantaneous density matrix of the light at delay time $\tau$. The electrons which pass through the sample are then sent through an electron spectrometer, which measures the energy loss or gain of the electron [107]
This technique in principle enables extraction of all normally-ordered moments of the field \( g^{(n)} \) through the electron loss probabilities \([60]\). In this work, we are merely content to show how Fock states and coherent states (of the same average photon number) are differentiated. As one can see from Fig. 4b, there are clear differences (even for small \( g \) such as 0.1) between the energy losses for Fock states and coherent states, which are noticeable for all energy losses with significant probability. That these differences are present over multiple data points (energy losses) is precisely what enables inversion to calculate moments beyond \( g^{(2)} \), giving a characterization of the statistics directly of the cavity mode.

III. DISCUSSION AND OUTLOOK

In optics, the realization of the nonlinear loss of Fig. 1 and the \( n \)-photon BIC effect should be within reach. Already Figs. 2 and 3 indicate possibilities for combining strong exciton-polariton nonlinearities with photonic bound states in the continuum. As discussed above, and also more extensively in SI Section IV, the nonlinearities are already strong enough, and low external losses have also very recently been achieved (exciton-polaritons have even already been interfaced with BICs as of this year).

Minimally, all that is required is a BIC (or quasi-BIC) which appears for a certain index of refraction, and Kerr nonlinearity (self-phase modulation). The types of architectures to realize the former (illustrated in Fig. 1) have already been realized several times now. Fock states with extremely high fidelities would require sizable nonlinearities, but sufficiently interesting intensity squeezing should already be achievable in the macroscopic domain for bulk-type nonlinearities (e.g., in silicon or InGaAs). Nanophotonic systems more broadly (exploiting coupled cavities based on high-\( Q \) ring resonators and microspheres \([61]\), or photonic crystal cavities \([62]\)) should enable the construction of almost arbitrary nonlinear losses, and even with very little background loss (though with weaker nonlinearities, leading to high squeezing rather than Fock state generation). Typically, on-chip nanophotonic squeezing has been limited by various external losses (e.g., associated with coupling out of the chip), but as of this year, significant progress has been made along these lines, achieving strong squeezing on-chip with second-order nonlinearities \([63]\). More extreme nonlinear dissipation, enabling one- and few-photon Fock states, could be achieved by combining these resonators with matter systems supporting single-photon-scale nonlinearities (e.g., cavity QED systems with photon blockade \([64]\) or Rydberg atoms \([65]\) with BICs).
Another worthwhile platform for implementing the physics described here is in superconducting circuits. Although several techniques already exist for creating Fock states in superconducting qubits (as discussed in the introduction, and reviewed in more depth in SI Section V), the approach we pose, which makes use only of Kerr nonlinearity and linear loss engineering, is quite flexible, and may be beneficial even when implemented in superconducting qubit systems. From a nonlinearity and external loss perspective, we suspect the capabilities are more than present to demonstrate Fock-state and squeezed-state generation with \( n \)-photon BICs. A simple heuristic argument is that even ten years ago, single-photon Kerr strengths in superconducting qubits were 30-times larger than the losses, which enables even cat-state generation from coherent states, one of the most exotic predictions in the quantum physics of the Kerr effect. Beyond providing a useful proving ground for the concepts developed here, our technique does provide a path to readily tune the Fock state order, and achieve fairly high Fock-state numbers with high fidelity, and at the minimum, large squeezing beyond the so-called 3 dB limit discussed in other platforms \[66-68\].

To summarize, we have presented a fundamentally new form of nonlinear dissipative interaction for photons. At the most basic level, the nonlinear dissipation arises from combining nonlinearity and leaky modes with frequency-dependent radiation loss. When the nonlinearity is Kerr, this combination induces a decay rate for photons with an intensity-dependence qualitatively beyond what is offered by commonly employed multiphoton and saturable absorbers. When the leaky-mode is an approximate BIC, the nonlinear dissipation creates a “potential” in photon number which facilitates the generation of Fock states and highly intensity-squeezed states.

As discussed earlier, the theory developed to describe such effects is quite general, as it is applicable to any Kerr nonlinear oscillator coupled to one or more continuua with frequency-dependent couplings. The consideration of Kerr nonlinearity is not so restrictive: many systems in nature with self-interactions are described by a Kerr Hamiltonian, with some value of the \( \beta \) parameter which can be predicted from first-principles, or measured. Such systems include: bulk optical materials (where Kerr comes from \( \chi^{(3)} \) \[69\]), exciton-polaritons (where Kerr comes from Coulomb interactions \[70\]), superconducting circuits (where Kerr comes from nonlinear inductance \[71\]), magnons (where Kerr comes from magnon-magnon interaction \[72\]), Rydberg atoms (where single-photon nonlinearities arise from Rydberg blockade \[65\]), and cavity-QED systems (where single-photon nonlinearities arise from photon blockade \[40\]).

Our work establishes a new connection between two highly active fields: (1) radiation loss engineering, which has primarily been explored in classical optics in the context of BICs, excep-
tional points, and non-Hermitian photonics [35, 38, 73–75] and (2) quantum-state engineering, where the use of nonlinear dissipation to engineer quantum states is well-appreciated (see e.g., Refs. [76–78]). In doing so, our work points to a new line of questions that we expect to be interesting in photonics and beyond. For example, beyond systems with BICs explored here, a natural subject to investigate would be quantum nonlinear systems with exceptional points, which are known to be sensitive to small changes in the refractive index [79]. Moreover, the general platform introduced here (nonlinearity plus frequency-dependent radiation loss) suggests the possibility of using second-order nonlinearity instead of Kerr. Since second-order nonlinearities enable phase-sensitive loss (and gain), it is clear that such systems enable qualitatively different opportunities. Such nonlinear losses, arising from second- and third-order nonlinearities might very well give paths towards stabilizing other quantum optical states that are of interest to the community (Schrodinger cat states, GKP states, cluster states, and the like).

Given the generality of the effects introduced here, we expect that the theoretical and experimental development of physical platforms to realize them will provide a great deal of exciting new areas for discovery.

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Supplementary Information for:

*n*-photon bound states in the continuum for strong intensity squeezing and deterministic stabilization of large photonic Fock states

S1. GENERAL THEORY

In this Supplement, we develop the theory of nonlinear open systems with frequency-dependent outcouplings. We show that from these systems arises a unique new form of nonlinear dissipation which is capable of deterministically stabilizing large Fock states of light, as well as highly intensity-squeezed states with much reduced intensity fluctuations compared to the classical shot noise level. In what follows, we develop a set of equations that will prescribe the evolution of the quantum state (density matrix) of a bosonic resonance — including for example photon probabilities ( photon statistics), field correlations ($g^{(1)}$), intensity correlations ($g^{(2)}$), and so on, in the presence of nonlinearity and a weakly non-Markovian (frequency-dependent) coupling to the continuum. Our theory is quite general, insofar as it can be applied immediately to any system for which one knows the frequency-dependent linear response of a resonance to incident far-field light (in other words, if one knows the frequency-dependent in-coupling of light to the resonance of interest). This frequency-dependent in-coupling can readily be calculated from time-domain electromagnetic simulations, but it can also be written analytically for several experimentally relevant photonic architectures based for example on temporal coupled mode theory (we present two examples).

The family of systems we consider is a driven Kerr nonlinear resonator, coupled to $N$ reservoirs (continuua) responsible for dissipation. We also allow the system to be pumped by a coherent state injected into one or more of the reservoirs. While the modes of the continuua can be of arbitrary origin, we will mostly focus on the case in which the resonator is coupled to a continuum of far-field radiation modes, but the theory accounts for absorptive reservoirs just as well. The Hamiltonian of this family of systems is:

$$H/\hbar = \omega_a a^\dagger a + \beta \omega_a a^\dagger a^2 + \sum_{k,i} \omega_{k,i} s_{k,i}^\dagger s_{k,i} + \sum_{k,i} g_{k,i} s_{k,i}^\dagger a + g_{k,i} a^\dagger s_{k,i}. \quad (S1)$$

Here, $a$ is the annihilation operator of the resonance with frequency $\omega_a$. The Kerr nonlinearity of the resonance is well-known to manifest as the second term in the Hamiltonian [40,42], leading to
a “photon-number-dependent resonance frequency.” The coupling of the resonance to the \( k \) mode of the \( i \) port (with frequency \( \omega_{k,i} \)) is \( g_{k,i} \).

To make contact with the main useful forms of the equations later used, we introduce the continuous operator versions of the Hamiltonian above. Denoting the density of states of the \( i \)th reservoir as \( \rho_i(\omega) \), where \( \omega \) is the frequency corresponding to the continuum label \( k \) (in other words, \( \omega_{k,i} \rightarrow \omega \)), we may define the continuous operators \( s_i(\omega) = \sqrt{2\pi \rho_i(\omega)} s_{k,i} \), defined such that \([s_i(\omega), s_{j}^\dagger(\omega')] = 2\pi \delta_{ij} \delta(\omega - \omega')\). We may also define a continuous version of the coupling: \( K_{c,i}(\omega) = -i\sqrt{2\pi \rho_i(\omega)} g_{k,i}^* \). We may then express the Hamiltonian equivalently as:

\[
\frac{H}{\hbar} = \omega_a a^\dagger a + \beta \omega_a a^2 + 2 \sum_{i=1}^{N} \int \frac{d\omega}{2\pi} \omega s_{i}^\dagger(\omega)s_{i}(\omega)
\sum_{i=1}^{N} \int \frac{d\omega}{2\pi} i \left( K_{c,i}(\omega)s_{i}(\omega)a^\dagger - K_{c,i}^*(\omega)s_{i}^\dagger(\omega)a \right).
\] (S2)

Now we develop the open-systems theory for this class of systems. We start by developing the Heisenberg equations of motion for this system, making direct contact with the classical limit, corresponding to temporal coupled mode theory. The Heisenberg theory also enables us to develop approximate quantum Langevin equations for this class of nonlinear dissipative systems. Then, we develop an approximate master equation for this class of systems, coinciding with Eq. (2) of the main text (we also arise at equivalent equations from the Heisenberg treatment).

**Heisenberg equations of motion**

Let us derive Heisenberg equations for \( a \), and for the reservoir operators \( s_{k,i} \). They follow from the Hamiltonian in Eq. (1) as:

\[
\dot{a} = -i\omega_a a - 2i\beta \omega_a a^2 + i \sum_{k,i} g_{k,i}^* s_{k,i}
\]

\[
\dot{s}_{k,i} = -i\omega_{k,i} s_{k,i} - ig_{k,i,a}.
\] (S3)

We use the “discrete” operators to derive the equations more simply, and then transform the equations into the form with the continuous operators and couplings afterwards. We may derive a closed equation for \( a \) by expressing \( s_{k,i} \) as:

\[
s_{k,i}(t) = s_{k,i}(0)e^{-i\omega_{k,i}t} - ig_{k,i} \int_{0}^{t} dt' e^{-i\omega_{k,i}(t-t')} a(t'),
\] (S4)
and plugging it into the equation for \( a \). The resulting equation of motion for \( a \) is:

\[
\dot{a} = -i\omega a - 2i\beta \omega a^\dagger a^2 - \int dt' \left( \sum_{k,i} \theta(t-t')|g_{k,i}|^2 e^{-i\omega k,i (t-t')} \right) a(t') - i \sum_{k,i} g_{k,i}^* s_{k,i}(0)e^{-i\omega k,i t}.
\]  

(S5)

In this equation, we have extended the upper limit of integration to \( t \to \infty \) by introducing a heaviside function: this proves more convenient in subsequent manipulations. To proceed, we introduce the function:

\[
K_{l,i}(\tau) = \theta(\tau) \sum_{k,i} |g_{k,i}|^2 e^{-i\omega k,i \tau}.
\]  

(S6)

Defining the input field operator of the \( i \) port as

\[
s_i(t) = \int \frac{d\omega}{2\pi} e^{-i\omega t} s_i(\omega),
\]  

(S7)

with \( s_{in,i}(\omega) = \sqrt{2\pi} \rho_i(\omega) s_{k,i} \), we express the equation of motion for \( a \) in the resulting form:

\[
\dot{a} = -i\omega a - 2i\beta \omega a^\dagger a^2 - \int dt' K_{l,i}(t-t') a(t') + \sum_i \int dt' K_{c,i}(t-t') s_i(t'),
\]  

(S8)

where \( K_l = \sum_i K_{l,i} \).

Note that these definitions imply that \( K_{l,i} \) and \( K_{c,i} \) are not independent. They are related in frequency-domain by

\[
K_{l,i}(\omega) = i \int \frac{d\omega'}{2\pi} \frac{|K_{c,i}(\omega')|^2}{\omega - \omega' + i\eta},
\]  

(S9)

with \( \eta \) infinitesimal. This relation enforces the connection between incoupling and loss (outcoupling).

**Equation of motion under coherent driving**

An important class of problems relates to the driving of \( a \) by a coherent pump (e.g., an external laser field sent through one of the ports). In this case, we can express the input field operator as

\[
s_i(t) = S_i(t) + \delta s_i(t),
\]  

(S10)

where \( S_i(t) \equiv \langle s_i(t) \rangle \) is the mean value of the pump field and \( \delta s_i(t) \) are the residual (operator-valued) fluctuations of the input. For a coherent drive, the initial state for which expectation values are taken with respect to (in the Heisenberg picture) is the vacuum state.
Classical limit

It is worth pausing to derive the classical limit of these equations. They provide a general theory of Kerr nonlinear resonators with frequency-dependent outcouplings. The classical limit is obtained by taking all operators to $c$-numbers. Denoting $\alpha = \langle a \rangle$ and $S_i = \langle s_i \rangle$, we have:

$$\dot{\alpha} = -i\omega_\alpha \alpha - 2i\beta |\alpha|^2 \alpha - \int dt' K_I(t-t')\alpha(t') + \sum_i \int dt' K_{c,i}(t-t') S_i(t'). \tag{S11}$$

Note that in the linear case, we may write the intracavity mean amplitude as (in frequency-domain)

$$\alpha(\omega) = \sum_i \frac{K_{c,i}(\omega)S_i(\omega)}{i(\omega_\alpha - \omega) + K_I(\omega)}. \tag{S12}$$

Approximate master equation for a nonlinear resonator coupled to a weakly non-Markovian reservoir

Here, we derive a master equation for the resonator mode corresponding to the Hamiltonian of Eq. (1). We derive it in two ways. The first way, presented in this section, goes through the standard master-equation derivation in the Born-Markov approximation. The second way derives an equation of motion for the reduced density matrix of $a$ in the Heisenberg picture. The Heisenberg approach is somewhat more general, requiring fewer assumptions, and also provides Langevin equations for the resonator mode which are useful starting points for other analyses (such as the analysis of lasers with nonlinear dissipation, to be presented in subsequent papers).

Derivation 1: Density matrix approach

The starting point for our density matrix approach is the equation of motion in the interaction picture for the reduced density matrix of $a$ (tracing out the reservoir), defined as $\rho_{a,I}(t) = \text{tr}_r \rho_I(t)$:

$$\dot{\rho}_{a,I} = -i\hbar \text{tr}_r ([V_I(t), \rho(0)]) - \frac{1}{\hbar^2} \int_0^t dt' \text{tr}_r ([V_I(t), [V_I(t'), \rho_I(t')]]) \tag{S13},$$

where

$$V_I(t)/\hbar = \sum_{k,i} g_{k,i} s_k^\dagger a_I(t) e^{i\omega_{k,i} t} + g_{k,i}^* a_1^\dagger(t) s_k e^{-i\omega_{k,i} t}. \tag{S14}$$

For the special case of the Kerr resonator,

$$a_I(t) = e^{-i\omega_\alpha (1+2\beta a^\dagger a) t} a, \quad a_1^\dagger(t) = a^\dagger e^{i\omega_\alpha (1+2\beta a^\dagger a) t}, \tag{S15}$$

S4
enabling us to write the interaction Hamiltonian as:

\[ V_I(t) / \hbar = \sum_{k,i} g_{k,i} e^{i(\omega_{k,i} - \omega_a(1+2\alpha^2))t} a_i^+ a + g_{k,i}^* e^{i(\omega_{k,i} - \omega_a(1+2\alpha^2))t}. \]  

(S16)

In what follows, we make the standard Born-Markov approximation, which stipulates that the reservoir state changes negligibly, and has no entanglement with the resonator. In that case, we write \( \rho_I(t') = \rho_{a,I}(t') \rho_r(0) \), with \( \rho_r(0) \) the initial density matrix of the reservoir.

In what follows, we will confine our explorations to the very broadly applicable case (especially in optics where thermal fluctuations are negligible) in which the reservoir in the vacuum state. In that case, the first term vanishes. The second term then evaluates to

\[
\dot{\rho}_a = -\int dt' \theta(t - t') \sum_{k,i} |g_{k,i}|^2 e^{-i\omega_{k,i}(t-t')} a_i^+(t)a_I(t') \rho_a(t') + \int dt' \theta(t - t') \sum_{k,i} |g_{k,i}|^2 e^{i\omega_{k,i}(t-t')} a_I(t) \rho_a(t') a_i^+(t') \\
+ \int dt' \theta(t - t') \sum_{k,i} |g_{k,i}|^2 e^{-i\omega_{k,i}(t-t')} a_I(t') \rho_a(t') a_i^+(t) \\
- \int dt' \theta(t - t') \sum_{k,i} |g_{k,i}|^2 e^{i\omega_{k,i}(t-t')} \rho_a(t') a_i^+(t') a_I(t).
\]

(S17)

Expressed in terms of the \( K_i \) defined earlier, this may be written as

\[
\dot{\rho}_a = -\int dt' \left( K_i(t-t') a_i^+(t)a_I(t') + K_i^*(t-t') a_I(t) a_i^+(t') \right) \\
+ \int dt' \left( K_i(t-t') a_I(t') \rho_a(t') a_i^+(t) + K_i^*(t-t') a_I(t) \rho_a(t') a_i^+(t') \right),
\]

(S18)

with \( K_i = \sum_i K_{i,i} \). To proceed to get an equation which is tractable, we make an adiabatic approximation (which holds very well in all the architectures we consider for Fock state generation). The adiabatic condition is a condition on the timescale of the variation the density matrix, denoted \( t_a \), and the timescale (memory time) of the kernel \( K_i \), denoted \( (\Delta \omega)^{-1} \). If \( t_a > (\Delta \omega)^{-1} \), then we may approximate: \( \rho_a(t') \approx \rho_a(t) \), yielding an \textit{time-local} equation for \( a \). To cast the resulting equation into a more explicit form suitable for numerical implementation and analysis, we write the equation of motion for the density matrix elements \( \rho_{m,n;I} \equiv \langle m | \rho_{a,I} | n \rangle \) (we have omitted the ‘\( a \)’ subscript for brevity). Plugging in the form of the interaction picture operators for the Kerr resonator, we arrive at

\[
\dot{\rho}_{m,n;I} = -m K_i(\omega_{m,m-1} + n K_i^*(\omega_{n,n-1})) \rho_{m,n;I} \\
+ \sqrt{(m+1)(n+1)} e^{i(\omega_{m+1,n} - \omega_{m+1,m})t} \left( K_i(\omega_{m+1,m}) + K_i^*(\omega_{n+1,n}) \right) \rho_{m+1,n+1;I},
\]

(S19)
where $\omega_{m,m-1} = \omega_a (1 + 2\beta (m-1))$.

In the Schrödinger picture, where $\rho_{m,n} = \rho_{m,n; t} e^{i\omega_{nm} t}$, we have

$$\dot{\rho} = -i[H_K, \rho] + D[\rho],$$

where $H_K = \omega_a a^\dagger a + \beta \omega_a a^\dagger a^2$ is the Kerr Hamiltonian, and the dissipator $D$ is defined through its matrix elements as

$$\langle m | D[\rho] | n \rangle = -(mK_l(\omega_{m,m-1}) + nK_l^*(\omega_{n,n-1})) \rho_{m,n}$$

$$+ \sqrt{(m+1)(n+1)} (K_l(\omega_{m+1,m}) + K_l^*(\omega_{n+1,n})) \rho_{m+1,n+1}.$$  

This is the general form of the master equation for a Kerr nonlinear resonance coupled to an arbitrary number of frequency-dependent reservoirs, assuming that the reservoirs have a bandwidth which is large compared to the characteristic decay time of $a$. This is different from the standard treatment of dissipation of a Kerr resonator in that in nearly all cases considered, the reservoir is assumed to be perfectly white, with no frequency dependence in its coupling to the resonance. Here, we preserve the effect of the frequency-dependence, and the new effects related to Fock state generation shown in the main text depend precisely on this loss.

**Addition of a coherent drive.** To accommodate a coherent driving field in this theory is straightforward. If the reservoir operators have a mean amplitude (corresponding to a coherent shift), then this leads to a driving term in the Hamiltonian of the form

$$H_{\text{drive}}/\hbar = \alpha(t) a + \alpha^*(t) a^\dagger,$$

where $\alpha(t)$ can be exactly specified in terms of the parameters of the Hamiltonian as

$$\alpha(t) = \sum_{k,i} g_{k,i} S_{k,i}^* e^{i\omega_{k,i} t} = -i \sum_{i} \int \frac{d\omega}{2\pi} K_{c,i}(\omega) S_i(\omega),$$

where $S_{k,i} = \langle s_{k,i}(0) \rangle$. Following the standard prescription, the equation of motion for the density matrix then becomes

$$\dot{\rho} = -\frac{i}{\hbar}[H_K + H_{\text{drive}}, \rho] + D[\rho].$$

**Derivation 2: Heisenberg approach**

In this subsection, we develop Heisenberg equations of motion for the resonator field. We write an equation of motion for $T_{n,m} \equiv |n\rangle \langle m|$ and for $s_q$. These operators are directly related
to the density matrix elements, as $\langle T_{n,m} \rangle = \text{tr}[\rho|n\rangle\langle m|] = \rho_{m,n}$. Typically, in quantum optical treatments of resonances, one writes an equation of motion for the annihilation operator $a$ directly, as we did in the previous section. However, due to the nonlinearity, the transition frequency varies as a function of photon number, rendering $a$ a polychromatic quantity in the interaction picture. We have found that the projector technique we develop here enables us to get analytical results beyond the mean-field approximation for nonlinear cavities. The projector technique simplifies matters because it breaks the field up into simple monochromatic parts. This projection operator lowers the cavity state by $m - n$ photons.

Using the commutators:

$$ [a, T_{n,m}] = \sqrt{n}T_{n-1,m} - \sqrt{m+1}T_{n,m+1}, \quad (S25) $$

$$ [a^\dagger, T_{n,m}] = \sqrt{n+1}T_{n+1,m} - \sqrt{m}T_{n,m-1}, \quad (S26) $$

$T_{n,m}$ evolves according to

$$ \dot{T}_{n,m} = -i\omega_{m,n}T_{n,m} $$

$$ + i \sum_{k,i} g_{k,i}s_{k,i}^\dagger \left( \sqrt{n}T_{n-1,m} - \sqrt{m+1}T_{n,m+1} \right) $$

$$ + i \sum_{k,i} g_{k,i}^* \left( \sqrt{n+1}T_{n+1,m} - \sqrt{m}T_{n,m-1} \right) s_{k,i} $$

$$ \dot{s}_{k,i} = -i\omega_{k,i}s_{k,i} - ig_{k,i}a. \quad (S27) $$

Eliminating $s_{k,i}$ as in Eq. (4), we have

$$ \dot{T}_{n,m} = -i\omega_{m,n}T_{n,m} $$

$$ - \left( \int dt' K_i(t-t')a^\dagger(t') \right) \left( \sqrt{n}T_{n-1,m} - \sqrt{m+1}T_{n,m+1} \right) $$

$$ + \left( \sqrt{n+1}T_{n+1,m} - \sqrt{m}T_{n,m-1} \right) \left( \int dt' K_i(t-t')a(t') \right) $$

$$ + F_{n,m}, \quad (S28) $$

where $F_{n,m}$ is an (operator-valued) Langevin force, defined by

$$ F_{n,m} = i \sum_{k,i} g_{k,i}s_{k,i}^\dagger(0)e^{i\omega_{k,i}t} \left( \sqrt{n}T_{n-1,m} - \sqrt{m+1}T_{n,m+1} \right) $$

$$ + i \sum_{k,i} g_{k,i}^* \left( \sqrt{n+1}T_{n+1,m} - \sqrt{m}T_{n,m-1} \right) s_{k,i}(0)e^{-i\omega_{k,i}t}. \quad (S29) $$
The Langevin force may be expressed in terms of the $K_{c,i}$ as where $F_{n,m}$ is an (operator-valued) Langevin force, defined by

$$F_{n,m} = \left( \sum_i K_{c,i}^* (t - t') s_i^\dagger (t') \right) \left( \sqrt{n} T_{n-1,m} - \sqrt{m + 1} T_{n,m+1} \right)$$

$$- \left( \sqrt{n + 1} T_{n+1,m} - \sqrt{m} T_{n,m-1} \right) \left( \sum_i K_{c,i} (t - t') s_i (t') \right).$$

(S30)

Note that, for a vacuum or multi-mode coherent-state reservoir, $\langle F_{n,m} \rangle = 0$ (which holds due to our choice of normal ordering).

To proceed, we must simplify integrals of the form:

$$\int dt' K_i(t-t') a(t') \equiv \sum_m \sqrt{m} e^{-i\omega_{m,m-1}t} I_m \equiv \sum_m \sqrt{m} e^{-i\omega_{m,m-1}t} \int dt' K_i(t-t') \tilde{T}_{m-1,m}(t') e^{i\omega_{m,m-1}(t-t')},$$

(S31)

where we have defined envelope quantities with a tilde as $\tilde{T}_{m-1,m}(t) = T_{m-1,m}(t)e^{-i\omega_{m,m-1}t}$. To simplify the integrals, we will consider a set of reservoirs for which the correlation time of the reservoir is much smaller than the intrinsic timescale of $\tilde{T}_{m-1,m}$, as we did in the density matrix derivation. In such a case, we get:

$$I_m \approx K_i(\omega_{m,m-1}) \tilde{T}_{m-1,m}(t).$$

(S32)

Plugging this result back into Eq. (S28), we arrive at

$$\dot{T}_{n,m} = -i\omega_{n,n} T_{n,m} - (mK_i(\omega_{m,m-1}) + nK_i^* (\omega_{n,n-1})) T_{n,m}$$

$$+ \sqrt{(m + 1)(n + 1)} (K_i(\omega_{m+1,m}) + K_i^* (\omega_{n+1,n})) T_{n+1,m+1} + F_{n,m}.$$  

(S33)

Upon noting that $\dot{\rho}_{m,n} = \langle \dot{T}_{n,m} \rangle$ and $\langle F_{n,m} \rangle = 0$, it becomes immediately clear that this equation is identical to Eq. (S20), indicating the equivalence of the density matrix and Heisenberg approaches.

**Addition of a coherent drive.** To accommodate a coherent driving field in the Heisenberg approach is also straightforward. If the reservoir operators have a mean amplitude (corresponding to a coherent shift) $S_{k,i} = \langle s_{k,i}(0) \rangle$, then in the force term $F_{n,m}$, we make the substitution

$$s_{k,i}(0) \rightarrow s_{k,i}(0) + S_{k,i},$$

(S34)

where the new $s_{k,i}(0)$ is has zero mean and standard vacuum fluctuations. Defining, as we did in the density matrix derivation $\alpha(t) = \sum_{k,i} g_{k,i} S_{k,i}^* e^{i\omega_{k,i} t}$, we see that

$$F_{n,m} \rightarrow F_{n,m} + i\alpha(t) \left( \sqrt{n} T_{n-1,m} - \sqrt{m + 1} T_{n,m+1} \right) + i\alpha^*(t) \left( \sqrt{n + 1} T_{n+1,m} - \sqrt{m} T_{n,m-1} \right).$$

(S35)
Using the commutation relations of Eq. (S25), the two terms may be written as $i[\alpha(t)a + \alpha^*(t)a^\dagger, T_{n,m}]$. Therefore, we see that

$$
\dot{T}_{n,m} = \frac{i}{\hbar} [H_K + H_{drive}, T_{n,m}] - (mK_1(\omega_{m,m-1}) + nK_1^*(\omega_{n,n-1})) T_{n,m}
+ \sqrt{(m+1)(n+1)} (K_1(\omega_{m+1,m}) + K_1^*(\omega_{n+1,n})) T_{n+1,m+1} + F_{n,m},
$$

which is in direct correspondence with the density matrix equation of motion.

### S2. APPLICATION OF THE GENERAL THEORY TO DIFFERENT NONLINEAR PHOTONIC ARCHITECTURES

The formalism developed in the previous sections represents a general theory of dissipation of a Kerr nonlinear resonator coupled to frequency-dependent reservoirs in the limit where the bandwidth of the reservoir is large compared to the intrinsic timescale of the mode $a$: a limit that holds under a wide variety of cases. A key strength of the theory derived here is that the theory applies to arbitrary reservoirs, and, as a result, is capable of effectively describing many nonlinear photonic architectures.

As a case-in-point, let us now show for several physical architectures how the functions $K_c$ and $K_l$ can be found, thus fully specifying our theory. We will show how these functions may be immediately “read off” from temporal coupled mode theories which have successfully been used to describe the architectures we consider (in the classical linear and nonlinear regimes; the addition of quantum fluctuations is standard). We note that since the nonlinearity is independent of the reservoirs (depending only on the magnitude of the intracavity field), we may find the $K_l$ and $K_c$ functions from a linear model.

**Application to a quantum nonlinear Fano mirror**

Consider the system shown in Figure 1b (left) of the main text: it consists of a nonlinear resonance (of amplitude $a$) coupled to two ports of radiation (right, denoted 1, and left, denoted 2). The incoming and outgoing waves (into and out of the resonator) in each port are denoted $s_{1,2\pm}$. The left port is terminated by a back-reflector.

We begin by writing down the temporal coupled mode (TCMT) equations and boundary conditions associated with Figure 1. The models are equivalent to those of [80-82] (only the last
includes Kerr, but is not quantized). The resonance evolves according to
\[ \dot{a} = -i\omega a - (\kappa_1 + \kappa_2)a - i\omega a^\dagger a^2 + \sqrt{2\kappa_1}e^{i\theta_1}s_{1+} + \sqrt{2\kappa_2}e^{i\theta_2}s_{2+}. \] (S37)

The waves in the two ports are connected to each other, and the resonance, by the following input-output relation
\[ \begin{pmatrix} s_{1-} \\ s_{2-} \end{pmatrix} = C \begin{pmatrix} s_{1+} \\ s_{2+} \end{pmatrix} + \begin{pmatrix} \sqrt{2\kappa_1}e^{i\theta_1} \\ \sqrt{2\kappa_2}e^{i\theta_2} \end{pmatrix} a, \] (S38)
with \( C \) the direct scattering matrix describing direct transmission between waves on the left and right of the resonator. \( C \) can be parameterized as
\[ C = \begin{pmatrix} -r_d & it_d \\ it_d & -r_d \end{pmatrix}, \] (S39)
with \( r_d, t_d \) being the (direct) reflection and transmission coefficients. Furthermore, because of the back-reflector, we have that the incoming and outgoing waves in 2 are related by
\[ s_{2+}(\omega) = -s_{2-}(\omega)e^{i\omega T} \leftrightarrow s_{2+}(t) = -s_{2-}(t - T), \] (S40)
where \( T = 2L/v \) is the round-trip time for light between the backreflector and the resonator (with \( v \) the group velocity). The phases \( \theta_{1,2} \) are not arbitrary, but are rather determined by the usual constraint
\[ C \begin{pmatrix} \sqrt{2\kappa_1}e^{-i\theta_1} \\ \sqrt{2\kappa_2}e^{-i\theta_2} \end{pmatrix} = -\begin{pmatrix} \sqrt{2\kappa_1}e^{i\theta_1} \\ \sqrt{2\kappa_2}e^{i\theta_2} \end{pmatrix}. \] (S41)

In what follows, we will consider a symmetric case in which \( \kappa_1 = \kappa_2 \) (this is the most common case). In this case, we can derive expressions for the phases \( e^{i\theta_{1,2}} \). For example, we have that
\[ e^{i(\theta_2 - \theta_1)} = \pm 1 \equiv \sigma. \] (S42)

Using this result, we may also find
\[ r_d - it_d\sigma = e^{2i\theta_1} = e^{2i\theta_2}. \] (S43)

Fourier transforming the TCMT equation for the resonance yields:
\[ [i(\omega_a - \omega) + 2\kappa] a(\omega) = \sqrt{2\kappa_1}e^{i\theta_1}s_{1+}(\omega) + \sqrt{2\kappa_2}e^{i\theta_2}s_{2+}(\omega). \] (S44)

The wave \( s_{2+} \), unlike \( s_{1+} \) is not an independent degree of freedom. In particular, we have:
\[ s_{2+}(\omega) = \frac{it_d s_{1+}(\omega) + \sqrt{2\kappa_2}e^{i\theta_2}a(\omega)}{r_d - e^{-i\omega T}}. \] (S45)
Plugging this into the TCMT equation yields:

\[
[i(\omega_a - \omega) + K_I(\omega)] a(\omega) = K_c(\omega) s_{1+}(\omega),
\]

where:

\[
K_I(\omega) = 2\kappa \left( 1 - \frac{e^{2i\theta_2}}{r_d - e^{-i\omega T}} \right) = 2\kappa \left( 1 - \frac{r_d - itd\sigma}{r_d - e^{-i\omega T}} \right)
\]

and:

\[
K_c(\omega) = \sqrt{2\kappa} e^{i\theta_1} \left[ 1 + \frac{itd e^{i(\theta_2 - \theta_1)}}{r_d - e^{-i\omega T}} \right].
\]

Application to two resonances coupled to a common waveguide

Let us now consider the second system shown in Fig. 1 of the main text, in which one mode (labeled by its annihilation operator \(a\), with anharmonic Hamiltonian \(H_a\)) is coupled to a second linear resonance, of frequency \(\omega_d\) (labeled by annihilation operator \(d\)). In many cases, this second resonance \(d\) can be thought of as the resonance of an end-mirror of the cavity, and we will occasionally refer to \(d\) as the mirror. We take the \(d\)-resonance to be linear, with Hamiltonian \(H_d = \hbar \omega_d d^\dagger d\). The two modes in general can be coupled by a (beam-splitter) interaction \(\hbar(\lambda ad^\dagger + \lambda^* a^\dagger d)\). Both \(a\) and \(d\) are also coupled to the continuum of far-field modes \(s_k\) outside of the cavity, where \(k\) enumerates the continuum of outside modes. Taking \(g_k\) and \(v_k\) to respectively be the coupling of \(s_k\) to \(a\) and \(d\), the total Hamiltonian of the system and reservoir may thus be expressed as:

\[
\frac{H}{\hbar} = \omega_a a^\dagger a + \beta \omega_a a^\dagger a^2 + \omega_d d^\dagger d + (\lambda ad^\dagger + \lambda^* a^\dagger d) + \sum_k \omega_k s_k^\dagger s_k + \sum_k (g_k a s_k^\dagger + g_k^* a^\dagger s_k) + \sum_k (v_k d s_k^\dagger + v_k^* d^\dagger s_k).
\]

To find \(K_I\) and \(K_c\), we derive the Heisenberg equations of motion for \(a\) and \(d\) in the absence of nonlinearity. They are:

\[
\dot{a} = -i\omega_a a - i\lambda^* d - i \sum_k g_k^* s_k
\]

\[
\dot{d} = -i\omega_d d - i\lambda a - i \sum_k v_k^* s_k
\]

\[
\dot{s}_k = -i\omega_k s_k - i(g_k a + v_k d).
\]

Eliminating \(s_k\) as

\[
s_k(t) = s_k(0) e^{-i\omega_k t} - i \int_0^t dt' e^{-i\omega_k (t-t')} (g_k a(t') + v_k d(t')).
\]
plugging it into the equation of motion for $a$, and making a white-noise approximation on $g, v$, we have:

\[
\dot{a} = -i\omega_a - \kappa a - i\lambda^* d - \sqrt{\kappa\gamma} e^{i\phi} d - i \sum_k g^* s_k(0) e^{-i\omega_k t}
\]

\[
\dot{d} = -i\omega_d d - \gamma d - i\lambda a - \sqrt{\kappa\gamma} e^{-i\phi} a - i \sum_k v^* s_k(0) e^{-i\omega_k t},
\]

(S52)

with $\kappa = \pi\rho|g|^2$, $\gamma = \pi\rho|v|^2$, and $\phi = \arg[g^*v] = \arg[g^*] + \arg[v] \equiv \phi_v - \phi_g$ ($\rho$ being the continuum density of states). To get $K_c, K_l$ we now Fourier transform and write the equation for $a(\omega)$. It is:

\[
-i\omega a = -i\omega a - \kappa a + \frac{(i\lambda^* + \sqrt{\kappa\gamma} e^{i\phi})(i\lambda + \sqrt{\kappa\gamma} e^{-i\phi})}{i(\omega_d - \omega) + \gamma} a - ie^{-i\phi} \left[\frac{\sqrt{2\kappa} - \sqrt{2\gamma} e^{-i\phi}(i\lambda^* + \sqrt{\kappa\gamma} e^{i\phi})}{i(\omega_d - \omega) + \gamma}\right] s(\omega),
\]

(yielding

\[
K_l(\omega) = \kappa - \frac{(i\lambda^* + \sqrt{\kappa\gamma} e^{i\phi})(i\lambda + \sqrt{\kappa\gamma} e^{-i\phi})}{i(\omega_d - \omega) + \gamma},
\]

(S53)

and

\[
K_c(\omega) = -ie^{-i\phi} \left[\frac{\sqrt{2\kappa} - \sqrt{2\gamma} e^{-i\phi}(i\lambda^* + \sqrt{\kappa\gamma} e^{i\phi})}{i(\omega_d - \omega) + \gamma}\right].
\]

(S55)

To reveal the similarity between this case and that of the Fano resonator-waveguide, let us consider the a specific case in which direct coupling between the reservoirs can be neglected ($\lambda = 0$). In that case, we get:

\[
K_l(\omega) = \kappa \left(1 - \frac{\gamma}{i(\omega_d - \omega) + \gamma}\right),
\]

(S56)

and

\[
K_c(\omega) = -i\sqrt{2\kappa} e^{-i\phi} \left(1 - \frac{\gamma}{i(\omega_d - \omega) + \gamma}\right),
\]

(S57)

which are both zero when $\omega = \omega_d$. In this case, $\omega_d$ is the BIC frequency, $\gamma$ is the filter bandwidth dictating the response time of the filter, and $\kappa$ is the intrinsic timescale of the resonator. We should note that this case has exactly the same physics as Eqs. (47) and (48): taking $r_d = 0$, and performing a quadratic Taylor expansion in $\omega$ about a zero of Eqs. (47) and (48) gives a quadratically increasing loss and outcoupling from the BIC point, with a bandwidth set by $\gamma$ (so $\gamma$ is playing the role of $1/T$ in the previous case).

We should also point out that besides the two-resonator+continuum system being another way to implement the nonlinear dissipation introduced here, it also serves as a basis to numerically validate our analytical theory from first principles. In particular, because this non-Markovian
system arises from two systems coupled to a continuum via white-noise coupling, it is amenable to standard open quantum systems simulations, where the density matrix of a system is propagated in time given some jump operators. For the two-resonator system here, one can readily find the equation of motion for the system density matrix $\rho$ (the system referring to $a + d$):

$$\dot{\rho} = -i[H_{ad}, \rho] - 2\pi \rho_0 \left( X^\dagger X \rho + \rho X^\dagger X - 2X \rho X^\dagger \right), \quad (S58)$$

where $H_{ad}$ represents the first line of Eq. (49), and $\rho_0$ is the continuum density of states. The operator $X$ is given by

$$X = ga + vd, \quad (S59)$$

indicating that the continuum coupling arises from the sum of the emitted fields from the two resonators (consistently with the possibility of destructive interference). Upon simulating these open-system dynamics for the two-resonator system, we find all of the effects (Fock and squeezed state generation, dependence on detuning, bifurcation of trajectories, etc.) that are found in the analytical theory.

Quantum mechanical consistency

We should note that the Langevin type equation, Eq. (8), needs to satisfy a certain constraint in order to preserve the commutation relation between $a$ and $a^\dagger$, $[a, a^\dagger] = 1$. Solving for $a$ in frequency-domain, that constraint is:

$$\int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \left| \frac{K_c(\omega)}{i(\omega_a - \omega) + K_l(\omega)} \right|^2 = 1. \quad (S60)$$

Note that we have not included the nonlinearity here, as it is a conservative term, and is guaranteed not to lead to any change in the commutation relations over time (similar to the Markovian case). We should note that the two models presented in this section were found (according to numerical evaluation) to satisfy this constraint, as well as that of Eq. (9).

S3. NONLINEAR LOSS AND $N$-PHOTON BOUND STATES IN THE CONTINUUM

The diagonal components of the density matrix $\rho_{n,n}$ correspond to the probability $p_n$ of there being $n$ photons in $a$. As the main text is primarily focused on realizing Fock and macroscopic sub-Poissonian states of light (with probability distributions more tightly concentrated than Poisson),
the equation of motion for the photon probabilities plays a central role. In this section, we develop equations of motion for the probabilities, as well as equations of motion for the mean and variance of the intraresonator photon number — showing the condition for spontaneous squeezing due to nonlinear loss.

**Dynamics of the populations**

The dynamics of the photon probabilities \( p_n = \rho_{n,n} \) evolve according to

\[
\dot{p}_n = -L_n p_n + L_{n+1} p_{n+1},
\]

(S61)

where the \( n \)-dependent (nonlinear) loss is given as:

\[
L_n = 2n \text{Re } K_1(\omega_{n,n-1}).
\]

(S62)

This equation takes the form of a rate-equation and can be readily solved numerically. A useful analytical result is for the dynamics of the mean photon number \( \bar{n} \) and the variance \((\Delta n)^2\) of the photon number. Assuming (as is typically the case) that the photon probability distribution distribution is sharply peaked compared to the scale of variation of \( L(n) \) (where \( L(n) \) is the continuous function version of \( L_n \)), we have that

\[
\dot{\bar{n}} = -L(\bar{n})
\]

\[
(\Delta \dot{n})^2 = L(\bar{n}) - 2L'(\bar{n})(\Delta n)^2.
\]

(S63)

**Derivation of equation of motion for cumulants**

The solution of Eq. (S61) provides the time-dependent probability distribution of \( a \), giving access to all moments of the photon number operator. In many cases, we are primarily only interested in the dynamics of the mean and the variance. Thus, it is useful to derive a specific equation of motion for the mean and variance of the probability distribution. We shall do so in the approximation that the uncertainty \( \Delta n \) is small compared to the mean \( \bar{n} \), a statement which is almost always valid for states we consider, including Poissonian states (where \( \Delta n = \sqrt{\bar{n}} \ll \bar{n} \) provided \( \bar{n} \gg 1 \)). As a result of Eq. (S61), a general moment of the distribution \( \langle n^k \rangle \) evolves according to

\[
\langle n^k \rangle = -\sum_{n=0}^{\infty} n^k L_n p_n + \sum_{n=0}^{\infty} n^k L_{n+1} p_{n+1}.
\]

(S64)
Shifting the index of the second term from \( n + 1 \rightarrow n \) and making use of the fact that \( L_0 = 0 \), we find
\[
\langle n^k \rangle = \langle ((n-1)^k - n^k) L(n) \rangle,
\]
(S65)

Thus, the mean evolves according to:
\[
\dot{n} = -\langle L(n) \rangle,
\]
(S66)
where we have denoted the mean as \( \bar{n} \) to make contact with notations from the main text (other average quantities in this section will not get a bar). The second moment evolves according to:
\[
\langle n^2 \rangle = -\langle (2n-1) L(n) \rangle.
\]
(S67)

The variance satisfies the equation of motion \( (\Delta n)^2 = \dot{n}^2 - 2\bar{n} \dot{n} \). To proceed, we will consider distributions for which the distribution is sharply peaked about mean \( \bar{n} \) (and is singly-peaked), such that \( \Delta n \ll \bar{n} \). In this case, we make a continuous approximation for the probability distribution: \( p_n \rightarrow p(n) \), with averages given by \( \langle f(n) \rangle = \int_0^\infty dn f(n)p(n) \). Since the distribution is sharply peaked compared to the scale of variation of \( L(n) \), we may Taylor expand the loss about the mean:
\[
L(n) \approx L(\bar{n}) + (n - \bar{n})L'(\bar{n}) + \frac{1}{2}L''(\bar{n})(n - \bar{n})^2.
\]
To lowest order, the mean simply evolves according to
\[
\dot{\bar{n}} = -L(\bar{n}).
\]
(S68)

Meanwhile, the variance is found as:
\[
(\Delta n)^2 = -\int_0^\infty dn p(n)(2n - \bar{n} - 1)L(n)
\]
\[
= -\int_0^\infty dn p(n)(2n - \bar{n} - 1) \left( L(\bar{n}) + (n - \bar{n})L'(\bar{n}) + \frac{1}{2}L''(\bar{n})(n - \bar{n})^2 \right)
\]
\[
= L(\bar{n}) - \left( 2L'(\bar{n}) - \frac{1}{2}L''(\bar{n}) \right) (\Delta n)^2 + O((\Delta n)^3)
\]
\[
\approx L(\bar{n}) - 2L'(\bar{n})(\Delta n)^2.
\]
(S69)

Here, we have used the simplification that \( \langle n - \bar{n} \rangle = 0 \). We have also ignored higher order variations in the distribution, and made a somewhat crude approximation that \( 4L' \gg L'' \), which occurs when the distribution varies over a scale large compared to 1 (and hence is not perfectly accurate in the Fock-state regime). Still, the approximate equations capture the dynamics of the first two cumulants fairly well.
The so-called quantum nonlinear BIC condition is

\[ L_{n_0} \equiv \text{Re} \ K_1(\omega_{n_0,n_0-1}) = 0. \tag{S70} \]

Physically, it means that there is a special photon number \( n_0 \) for which its outcoupling to the far-field vanishes (thus becoming lossless). In linear optics, such lossless states are referred to as bound states in the continuum (BICs), as they are fully bound (with no leakage into the outside world), despite in principle having coupling to the continuum.

Suppose the zero of the function \( K_1(\omega) \) occurs at some frequency \( \omega_0 \). Then, we may expand \( K_1 \) around \( \omega_0 \) as \( \text{Re} \ K_1(\omega) \approx c_2(\omega - \omega_0)^2 \) (there is no linear component because loss has to be non-negative). Substituting the nonlinear resonance frequency: \( \omega_{n_0,n_0-1} = \omega(n=0) + 2\beta(n_0-1) \), we have that

\[ c_2[(\omega(n=0) - \omega_0) + 2\beta(n_0 - 1)]^2 = 0 \implies n_0 = \frac{\Delta_0}{2\beta} + 1, \tag{S71} \]

where \( \Delta_0 \equiv \omega_0 - \omega(n=0) \) is the detuning of the linear resonance from the BIC frequency. This simple equation reveals a key consideration: larger single-photon nonlinearities (\( \beta \)) and smaller detunings (\( \Delta_0 \)) lead to smaller photon numbers, while smaller single-photon nonlinearities (characteristic of “bulk material” nonlinearities) lead in principle to Fock states at larger photon numbers.

**S4. FURTHER DISCUSSION ON EXPERIMENTAL IMPLEMENTATION AND RELEVANCE OF CHOSEN PARAMETERS**

In this section, we provide some justification for the choice of parameters in the main text examples. We should first note however that the effects describe obtain for a fairly broad range of parameters (as the effect is related to interference), and so the parameters taken in the text should really be viewed as an chosen for concreteness, rather than viewed as necessary to optimize the effect.

An important upshot of the fairly detailed discussion below is that the magnitudes of the nonlinearities, as well as the ability to achieve BICs, and achieving the needed \( Q \)-factors is all within reach, and that the main parameter to focus on in experimental implementation is the magnitude of the background linear losses. The dependence of our effect on all other parameters is sufficiently
flexible that one should be able to start with the requirement of sufficiently low background linear loss, and design from there.

**Fano resonant photonic crystal waveguide:** The structure and the corresponding theoretical description of the classical optical response are based off of [39]. In Yu et al., they consider the photonic crystal cavity-waveguide structure illustrated in Fig. 3. As noted in the manuscript, the dynamics of the quasi-BIC are well-described by the temporal coupled mode theory that we describe in Sec. IV. A (excluding the gain medium, which we do not have in our work). The temporal coupled mode theory is one including: a resonator coupled to a waveguide terminated with a back-reflector, as in Fig. 2a of the manuscript. This same system-type was also considered in Refs. [80–83] (the last study even considers Kerr nonlinearity, but no quantum statistical effects).

From these works, it is known that the overall system behavior (including the Q-factors) are well-described by a few parameters: (a) the coupling strength of the cavity to the left- and right-going waveguide fields (denoted $\sqrt{2\kappa_1,2}e^{i\theta_1,2}$ in our model of Sec. IV. A) (b) the round-trip time of light propagation in the waveguide, denoted $T$ (and equal to the inverse bandwidth $\delta \omega$ defined in the text), (c) the frequency of the cavity mode (denoted $\omega_a$), (d) the direct transmission and reflection coefficients ($r_d,t_d$), and (e) the external losses (associated with a frequency-independent loss-rate $2\kappa_i$), associated for example with the cavity (due to say, vertical scatter, or some other non-radiative mechanism). The termination of the photonic crystal waveguide on the left side ensures negligible leakage from the left side, and so following earlier work, we do not take it into account. As described in Sec. IV. A., we note that this temporal coupled mode theory correctly predicts (a) a highly frequency-dependent Q factor with (b) a minimum limited by external loss $2\kappa_i$, (c) an effective Fano reflection coefficient associated with the end mirror (matching what is in Ref. [81]), and (d) a large ratio of photons in the cavity to photons in the waveguide of $1/(2\kappa T)$ [39].

We should note that the main “relevant” parameters for the $N$-photon BIC are $\kappa, T$, and $\kappa_i$, as well as the nonlinearity $\beta$ (the actual frequency of operation of the BIC has been demonstrated from visible to telecommunication wavelengths (see [39, 54, 84])). Reserving the nonlinearity discussion for the discussion below on exciton polaritons, we mention that a wide range of values for $\kappa$ and $T$ have been achieved in different works (in Ref. [83] the coupling $Q$ is 3000; in Ref. [81] it is 800; in [82] it is about 20000), and we take a $\kappa$ of $10^{-3} \omega_a$. It may readily be adjusted in the PhC architecture of Fig. 3 by creating the L7 cavity further from the waveguide with little change in the external loss. The round-trip time is controlled by the waveguide length and is
therefore readily adjustable. We take the “round-trip $Q$” to be 100, which again falls in the range of previous work (for example in all of Refs. [39] [81] [83], we back out values from $< 100 - 300$). The external loss in the figures is taken as a variable parameter (as it is the main limiting factor on the magnitude of our effect), and is sensitively dependent on the actual system design: however, it is experimentally known to be able to exceed $10^6$ and approach $10^7$ [85].

**Exciton polaritons:** As the source of the Kerr nonlinear medium, we take as an example the use of exciton polaritons in semiconductor quantum wells. Extensive work has been done probing the strong Kerr nonlinearities associated with these exciton polaritons, and their use for generating quantum light. Already, Kerr bistability has been observed in such systems (with intensity squeezing), as well as antibunching of light due to partial polariton blockade (the exciton nonlinearities observed in [46, 47] are within an order of magnitude of the non-radiative exciton losses in [46, 47], and are in fact larger than or similar to the losses in [54]).

The nonlinearity arises through the Coulomb interactions between excitons. The magnitude of the nonlinear interaction between excitons depends on the area over which the excitons are confined, and is characterized by an interaction constant $U$. Typical experimental values are roughly $U = 20 \mu$eV $\cdot \mu$m$^2$ (taking in mind our convention for the factor of 2 in the definition of the Kerr coefficient). The nonlinearities in principle can be made even higher by considering for example dipolar polaritons, as mentioned in [46]. Taking the photonic crystal (PhC) photonic structure in [39], and assuming a layer of InGaAs can be deposited within a sub-wavelength of the PhC (e.g., within a few 10s of nm), the interaction area is roughly $1.2 \ \mu$m$^2$. This leads to an exciton-exciton Kerr coefficient of $20 \ \mu$eV. Supposing the excitons strongly couple to the resonance (which we’ll address shortly), this leads to a polariton-polariton interaction strength of $\beta = 5 \ \mu$eV when the exciton and resonator are on-resonance (giving exciton content of 0.5 for the polariton). This ultimately assumes that the exciton strongly couples to the photon. When the in-plane extent of the quantum well covers the mode area of the resonance, the Rabi splitting $\Omega$ is given by $\Omega = \sqrt{\frac{\hbar^2}{4\pi\epsilon_0} \frac{2\pi e^2 f}{m L_{\text{eff}}}}$ [86], where $f$ is the exciton oscillator strength per unit area, and $L_{\text{eff}}$ is an effective mode length. Taking a typical oscillator strength density of $5 \times 10^{12}$ cm$^{-2}$ and estimating from Ref. [39] a mode length of 4$\mu$m (the mode length takes into account the squared index of refraction), we estimate a Rabi splitting of 1.3 meV. This magnitude is very much in line with the literature on exciton-polaritons (including the above-references experiments): this is unsurprising in light of the modal dimensions of the photonic structure here being somewhat similar to that of the other platforms. Notably, it is substantially larger than the photonic and excitonic damping rates, indicating strong coupling (as
is consistent with many of the experiments cited above).

**Pump scheme:** In our scheme to deterministically create Fock states, we consider initially loading the cavity with a coherent pump pulse (basically to create an initial coherent state that then relaxes to the Fock or squeezed state).

Simulating the quantum state dynamics of the resonance under this type of pumping, we find that a 30 fs pulse containing roughly 100 photons can load the 50-photon coherent state used in the simulations of Figs. 2 and 3. Such pulse durations and pulse energies are well-within what is possible with pulsed lasers at 1.5 eV, the exciton energy (e.g. Ti:Sapphire).

**Free electron probing:** In the section on detection, we discuss the possibility of detecting the exciton polariton quantum state with a flying free-electron passing over the cavity. The detection is based on the electron energy loss and gain resulting from the quantized interaction of the electron probe with the optical field. As mentioned in the text, the electron loss and gain probabilities are essentially dictated by a single parameter, $g$, which is typically estimated as $g = eL\sqrt{\frac{1}{2\epsilon_0\hbar\omega V}}$, with $L$ the length of interaction (essentially the cavity length) and $V$ the mode volume. Taking from [39] a length of 3 $\mu$m and a mode volume of 0.26 $\mu$m$^3$, we get a $g$ of roughly 0.1, which is very much in line with several estimates for an efficient single-photon coupling with the electron (see e.g., [58]).

**S5. RELATION TO THE LITERATURE**

The new effect that we describe ($n$-photon bound states in the continuum) is the only one we are aware of that can potentially allow for deterministic creation of large multiphoton Fock states in optics. That said, let us use this section to discuss in some depth the previous theoretical and experimental work on implementing nonlinear loss in optics to discuss some works that at least establish a precedent for the ideas here.

**Nonlinear dissipation**

Let us connect this nonlinear dissipation to the state-of-the-art in quantum-state engineering. In that field, it is well-appreciated that a nonlinear loss is a very valuable resource, and has been explored as the basis for generating cat states, error correction, and more [76–78]. The value derives from the fact that nonlinear loss can actually reduce quantum fluctuations of light, rather
than add them, as linear loss does.

**Nonlinear absorbers in optics.** An example of a nonlinear dissipation is simple $n$-photon absorption (two-photon absorption, three-photon absorption, four-wave mixing, etc.). Such nonlinear absorbers had been extensively studied, as early as four decades ago, in the context of lasers and harmonic generation for reducing the quantum fluctuations of light: that said, previously, only simple nonlinear absorbers such as two-, three-, and multi-photon absorbers (arising either from literal absorption, or harmonic generation) had been considered, and the noise reductions were fairly modest — typically not exceeding 50% reduction of photon number fluctuations below the classical shot noise (Poisson) limit [87–91]. let alone the close approximations to resonator Fock states that we shall show here. Unlike simple multi-photon absorption, the new nonlinear loss that we present here is not well-described by a Taylor expansion about zero photons, and, in that precise sense, can be considered as an emergent *non-perturbative* nonlinear loss — which leads to the stabilization of states not usually accessible through simpler nonlinear loss like multiphoton absorption. Phrased in the language of nonlinear dissipators, a typical $k$-photon absorption process might have a jump operator of the form $J_{k \text{photon}} = a^k$. On the other hand, the nonlinear loss $L_n$ associated with Eq. 4, arises effectively from an effectively high-order nonlinear dissipator as:

$$L_n = 2 \langle n | a^\dagger \text{Re} \ K_1(a^\dagger a) a | n \rangle,$$

which, for $n \approx n_0$, could be written as $L_n \sim \langle n | a^\dagger (a^\dagger a - n_0)^2 a | n \rangle$, which clearly does not admit a low-order Taylor expansion about $n = 0$, as simple multiphoton absorbers do.

**Trap state physics.** Another important class of absorption mechanisms, which has extensively been considered in the microwave domain (for superconducting qubits, as well as Rydberg atoms) are “trap states.” Trap states are states to which a pump cannot add more photons. This is in general due to a vanishing matrix element to add a photon, requiring a highly nonlinear pump mechanism. While in optics, such highly nonlinear pumping mechanisms have not been realized, in both microwave frequencies, the very strong and unique couplings of Rydberg atoms and superconducting qubits to microwave cavities enable further possibilities. As mentioned in the introduction, for Rydberg atoms, for very precisely controlled Rabi frequencies and interaction times between the atom and cavity, there can be a vanishing matrix element from $n$ to $n + 1$ photons [22, 23, 49]. Such mechanisms are also possible with superconducting qubits, although they make use of a Hamiltonian unique to the coupling of Josephson junctions to cavities [92, 93]. Generally, such “trap state” approaches have proven challenging to scale up to sizeable Fock states.
(e.g., $n > 10$), often because they depend on a fairly precise zeroing of the matrix element, thus being unstable with respect to perturbations. Nevertheless, a clear “advantage” of the $n$-photon bound state in the continuum is that it requires only Kerr nonlinear and tuning of optical phases (to achieve frequency-dependent loss) and is thus extremely versatile, and can be applied to any system with intensity-dependent nonlinearity (in other words, there is no requirement for strong coupling, or for specialized coupling Hamiltonians that are not generic that currently do not exist in optics.) We should note that the $n$-photon BIC approach also relies on a cancellation, but that fairly strong cancellations of the loss have been demonstrated [39, 84]. Even more recently (as of this year), it has been demonstrated that ultra-high-$Q$ BICs can be realized (approaching $Q = 10^7$) with very low mode volumes (enhancing nonlinearity [94]); undoubtedly useful for realizing the effects proposed here. Moreover, as shown in Figs. 2, 3, our technique also can be used for generating extremely intensity squeezed light, even when the condition for exact Fock states is not met. Importantly, the technique presented here is also versatile enough to be used even in the macroscopic optics regime to generate highly intensity squeezed light with large numbers of photons.

**Kerr squeezing.** One of the last related pieces of physics that we mention here is the fact that Kerr nonlinearities had, even three decades ago, been known to readily generate squeezing (typically squeezed vacuum states) [56, 95–98]. This effect arises from the fact that Kerr nonlinearity leads to distortion of quantum statistics (similar to the third $Q$-function plot in Fig. 3b). The resulting state, when destructively interfered with itself, can produce vacuum squeezed light (similar to what second-order nonlinearities can do but typically with higher powers compared to the second-order case). Intriguingly, it was also shown that solitons could in fact undergo intensity squeezing. In particular, by taking a soliton and sending it through a simple optical bandpass filter, some squeezing could be generated (roughly 2-4 dB) [99]. And in fact, that effect is understandable as a special case of the general quantum optical theory of radiation outcoupling developed here. As described on page 17, a loss that increases with intensity leads to intensity squeezing, and such a loss (which leads to flattened input-output relations) is what was effectively employed in the soliton work. Thus, our work can be seen as introducing a concept that takes photon-number squeezing to its logical extreme, introducing the type of loss (and ways to realize it) that create Fock states.
Proposals to generate Fock states applicable to optics

Let us discuss the current status of theoretical and experimental methods to generate “large” Fock states. A large class of methods to generate such states are measurement-based, and are accordingly non-deterministic, meaning that the order of the Fock state is often random, as is the time in which a target Fock state is generated. Such measurement-based protocols have been applied in practice to light generated by spontaneous parametric down-conversion [28–30]. Such “measurement-based” techniques have also been proposed in the context of quantum non-demolition measurements [95], as well as in the quantum interactions of free electrons with light [31]. On the other hand, there is another broad class of methods to generate $n$-photon states, such as via superradiance, and techniques such as the $n$-photon gun (the latter of which produces correlated streams of photons, but not directly the state $|n\rangle$, which we are interested in, as mentioned in [100]). In superradiant proposals, one uses a so-called Dicke state of $s$ atoms, which emit $s$ photons into free-space [32 33] or a waveguide [34]. However, such Dicke states are themselves extremely challenging to produce (as they are essentially atomic Fock states), and the emission, basically being spontaneous emission from $s$ atoms, is into a large number of modes, therefore not being emitted into a useful temporal mode (in contrast to a resonator).

For single photon Fock states on the other hand, the capabilities are much more mature. One-photon Fock states can be deterministically created using e.g., heralded parametric down conversion, photon blockade, or quantum emitters (see e.g. [27] for a review). One-photon Fock states are already heavily applied in quantum technologies today: especially in the domain of photonic quantum computing, communication, and information processing. As a result, there is still even today great experimental and theoretical interest in finding new ways to produce one photon Fock states (and realize concomitant single-photon nonlinearities), with emerging techniques being proposed such as nonlinear excitonic mirrors [53]. Rydberg blockade with cold atoms [65], polariton blockade with strongly interacting exciton polaritons [46 47], “unconventional” photon blockade [101], engineered driving terms in the Hamiltonian [102]).

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Despite many of these structures being theoretically unable to achieve literally infinite quality factor, due to their finite extent [35])

While the origin of the continuum modes can be arbitrary, we will focus on the case in which the resonator is coupled to a continuum of far-field radiation modes.

Our theory is mapped to the standard theory of damping of a resonator with the identification $K_c(\omega) = \sqrt{2\kappa}$, with $\kappa$ the amplitude decay rate (yielding $K_l = \kappa$ and reducing Eq. (2) to the standard master equation for a damped cavity as in [49]).

There is no linear component because loss has to be non-negative.

Assuming an initially monochromatic electron incident on the photonic structure, the probability of energy loss of $m$ photons for a given quantum state is (negative $m$ denotes photon absorption):

$$P_m = \langle E_0 - m\hbar\omega \mid \text{tr}_{ph}[S(|E_0\rangle\langle E_0| \otimes \rho_{ph})S^\dagger]\rangle |E_0 - m\hbar\omega \rangle,$$

where $S$ is the evolution operator describing the electron-photon interaction. It is given by $S = \exp [ga^\dagger b - g^*b^\dagger a]$, where $b$ ($b^\dagger$) is an operator lowering (raising) the electron energy by one unit. Namely: $b|E_0 - m\hbar\omega\rangle = |E_0 - (m + 1)\hbar\omega\rangle$ with $bb^\dagger = b^\dagger b = I$. Note that there are slight differences in the photon transition frequency due to Kerr nonlinearity. These are negligible from the perspective of the electron interaction, and we can take the electron energy change to be approximately $\hbar\omega_0$. This is valid insofar as we are mostly interested in the number of photon exchanges, rather than the exact
energy change of the electron per photon exchange (which will be tightly centered around $\hbar \omega_a$).