Creating large Fock states and massively squeezed states in optics using systems with nonlinear bound states in the continuum

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The quantization of the electromagnetic field leads directly to the existence of quantum mechanical states, called Fock states, with an exact integer number of photons. Despite these fundamental states being long-understood, and despite their many potential applications, generating them is largely an open problem. For example, at optical frequencies, it is challenging to deterministically generate Fock states of order two and beyond. Here, we predict the existence of an effect in nonlinear optics, which enables the deterministic generation of large Fock states at arbitrary frequencies. The effect, which we call an n-photon bound state in the continuum, is one in which a photonic resonance (such as a cavity mode) becomes lossless when a precise number of photons n is inside the resonance. Based on analytical theory and numerical simulations, we show that these bound states enable a remarkable phenomenon in which a coherent state of light, when injected into a system supporting this bound state, can spontaneously evolve into a Fock state of a controllable photon number. This effect is directly applicable for creating (highly) squeezed states of light, whose photon number fluctuations are (far) below the value expected from classical physics (i.e., shot noise). We suggest several examples of systems to experimentally realize the effects predicted here in nonlinear nanophotonic systems, showing examples of generating both optical Fock states with large n (n > 10), as well as more macroscopic photonic states with very large squeezing, with over 90% less noise (10 dB) than the classical value associated with shot noise.

The principle of wave-particle duality is at the core of the modern understanding of the electromagnetic (EM) field. Central to the particle side of the duality is the idea of quantization, which states that the EM field is composed of discrete packets of energy (photons). In the language of quantum electrodynamics, this quantization is expressed by the Fock states \( |n\rangle \) \((n = 0, 1, 2, \ldots)\), which are eigenstates of the photon number operator \(a^\dagger a\), where \(a\) is the annihilation operator of a quantized mode of the electromagnetic field. Being photon number eigenstates, they have an exactly defined integer photon number, with zero uncertainty. They are also eigenstates of the electromagnetic energy operator (the Hamiltonian), \(H = \hbar \omega a^\dagger a\), where \(\hbar\) is the reduced Planck constant, and \(\omega\) is the frequency of the EM mode in question. As the eigenstates of the Hamiltonian, they are also time-harmonic solutions to the Schrodinger equation for the electromagnetic field. In that sense, Fock states are the most elementary quantum states of light and, as such, have played a foundational role in our understanding of the quantum theory of light.

Accordingly, Fock states have long been identified by the quantum community as an important state to access. For example, these highly nonclassical states have long been considered for precision measurements (in the field of quantum metrology) because they have no uncertainty in their photon number (or equivalently, their intensity). These states are thus not subject to the so-called shot noise associated with the Poissonian fluctuations of photon number in classical light pulses (corresponding to quantum-mechanical coherent states) (1–3). One of the earliest proposed applications in precision measurement was to use a Fock state in an optical cavity as an extremely sensitive sensor of small vibrations (4). Beyond applications in precision measurements, they are also considered valuable for the fields of quantum simulation and quantum information processing. For example, Fock states of microwave frequency fields in LC resonators have already been used for quantum chemistry tasks, such as calculating the energy spectra of molecules (5). Another high-profile application of large Fock states along these lines is in the implementation of quantum algorithms such as boson sampling (6–13), in which the passage of a Fock state through an array of waveguides and beamsplitters can

**Significance**

The quantization of the electromagnetic field leads directly to the existence of quantum mechanical states, called Fock states, with an exact integer number of photons. Despite these fundamental states being long understood, and despite their many potential applications, generating them is largely an open problem. As a result, large Fock states are considered a type of “holy grail” for quantum science and technology. In this work, we predict the existence of an effect in nonlinear optics that enables the deterministic generation of large Fock states at arbitrary frequencies, using standard lasers and nanostructured optical components—which may enable all of the envisaged applications of these elusive states in quantum science and technology.

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be used to calculate matrix permanents. For this application, a “modest” Fock state of even 100 photons can enable computations of matrix permanents at least fifteen orders of magnitude larger than could be handled by even the largest supercomputers today. Beyond this, Fock states are also important “resource states,” which enable the generation of other desirable quantum states. For example, Fock states can be used to generate Schrödinger cat states (14), displaced Fock states, and Gottesman–Kitaev–Preskill states, which all have been identified as having important applications in quantum computation.

For the reasons above and many more, the problem of generating Fock states has generated considerable attention. At microwave frequencies, it is possible to deterministically produce Fock states of modest sizes (roughly 15 photons), with a high rate of success (>90%). These Fock states are generated in microwave resonators through a combination of external driving of the cavity by microwave pulses and superconducting transmon qubits (15) (which provide a very strong nonlinearity). 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 (16). Older foundational work in the field of cavity quantum electrodynamics 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 (17–19). Such Rydberg atom–cavity interactions form the basis for new theoretical proposals to extend microwave Fock states to higher photon numbers (20, 21).

In optics, the situation is drastically different, where the techniques above cannot be applied. In fact, it is currently a significant challenge to deterministically produce Fock states in optics when \( n \geq 2 \). This lies in stark contrast to single-phonon Fock states, where the capabilities are much more mature (22). In general, proposals for generating the Fock state \( |n\rangle \) \((n \geq 2)\) are either nondeterministic meaning the state is generated with random order or at random times (23–26) or are otherwise rather complex and resource intensive (27–32), requiring highly correlated (superradiant) states of many-atom, exotic high-order photonic nonlinearities, and/or extremely complex pump protocols—all of which have thus far not been implemented.

Here, we predict a remarkable effect in nonlinear optics that enables deterministic creation of large Fock states. In particular, Fock states result from simply taking a specially designed photonic resonator with an intensity-dependent refractive index, injecting laser light into that resonator (which loads a coherent state into it), and letting the photons decay from the resonator through radiation loss (leakage of light from the resonator). Radiation loss in this special resonator will—in the ideal case of the effect we predict—convert the coherent state into a Fock state with perfect efficiency. This is in stark contrast to the typical case in resonators with loss, where classical (coherent) states stay classical upon their decay, and nonclassical states become classical. Here, a classical state, after decaying, becomes extremely nonclassical (a large Fock state). This surprising result is accounted for by the fact that the specially designed resonator we introduce supports a bound state of many photons: Namely, the resonator is dissipationless when there are \( n \) and exactly \( n \) photons in the resonator, and it is dissipative otherwise. This effect can be seen as an example of “dissipative state preparation” in which quantum states can be stabilized by dissipation. Such effects are of great interest and are being employed extensively in atomic and superconducting qubit systems (see for e.g., refs. 33–36), although relatively little of this dissipative-state engineering has been employed for photons in nonlinear optics. One of the main contributions of this work is to establish a type of nonlinear dissipation for photons that both stabilizes Fock states and promotes the creation of extremely squeezed states of a resonator, potentially beyond what has been observed or inferred thus far.

We show that the many-photon bound state, which we call an \( n \)-photon bound state in the continuum, also facilitates generating very strongly squeezed states of photons. Squeezed states, in particular photon number-squeezed states, are quantum mechanical states of the radiation field with photon number variance \((\Delta n)^2\) less than the mean \(\langle n\rangle\): \((\Delta n)^2 < \langle n\rangle\). In other words, their fluctuations are below the value associated with the shot noise typical of classical light (in quantum mechanical coherent states). Because shot noise is associated with Poissonian fluctuations of the photon number, such photon-number squeezed states with variance below the mean are often called sub-Poissonian. Fock states with \(\Delta n = 0\) are a special case of number-squeezed states, representing the ultimate degree of squeezing. Number-squeezed states are very useful and desirable in their own right for precision measurement and have been thoroughly explored in nonlinear optics (37–42). There is a large push to realize strong squeezing in nanophotonic systems, which would open up many applications in quantum sensing and precision measurement. That said, large degrees of squeezing in nanophotonics (e.g., 10 dB) have proven elusive, although impressive strides have been made recently based on the use of second-order nonlinearities (43). The effects we introduce enable very large degrees of squeezing in nanophotonic systems, and we show examples of how photon-number fluctuations can realistically be reduced by over 90% (10 dB) of the classical shot noise level.

Before moving to the results, we note that these remarkable predictions are the direct consequence of a concept that we put forth in this paper: photonic structures with nonlinear radiation loss (Fig. 1), which is, in other words, photonic structures where the lifetime of the photons depends on how many photons are inside the structure. We expect this concept to have many applications even for classical optical devices such as lasers, modulators, sensors, and switches. Here, we focus on the remarkable quantum effects that light experiences when leaking out of these structures, developing the first-principles quantum optics theory of them. It must be emphasized it is precisely due to this nonlinear radiation loss that we are able to predict the possibility of directly producing Fock states by direct laser excitation of simple and existing nonlinear optical structures.

**1. Radiation Loss in Nonlinear Photonic Resonances**

We begin by describing the effects and the intuition behind them. The physics we are interested in is that of radiation loss in photonic resonators made with materials that have an intensity-dependent refractive index (Kerr nonlinearity). We consider a special type of photonic resonator, of which Fig. 1 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) (44), quasibound states in the continuum (45, 46), and Fano resonances (47). Characteristic to these high-Q resonances is that their Q-factor...
sensitively depends on the geometrical and material parameters of the resonator (e.g., feature size, index of refraction, photon wavevector), and the \( Q \) achieves a very large maximum for some value of these 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. 1). When this happens, the \( Q \) is limited only by what we will call "external" losses. These external losses can arise due to a number of factors, such as residual absorption (e.g., from impurities), roughness of the sample leading to light scattering (e.g., from structural disorder), vertical leakage (in planar, "chip-like" structures), or even finiteness of the structure. In general, even for very good BICs, there will still be some residual loss. As we will find in the examples of the main text, even with this loss, one can generate states for which the states are either of "mesoscopic" photon number (so \( \gg 1 \)), but still in the quantum regime, as would be the case for a 10-photon or 100-photon state), yet close to Fock states (with photon number uncertainty of order 1; Figs. 2–4), or of very large photon number (essentially macroscopic light states, typical of what is emitted by table-top lasers), but with photon-number fluctuations far below the shot noise level (Fig. 6). As a point of terminology, we will generally use the term BIC to refer to "cancellation-induced" high-\( Q \) resonances, in keeping with current usage of the phrase (45, 48). * Despite many of these structures being theoretically unable to achieve literally infinite quality factor, due to their finite extent (44).

Now, we consider the quantum mechanical dynamics of the intraresonator photon state, due to the leakage of light out of a resonator with a BIC and Kerr nonlinearity. At first glance, it seems that the dynamics of radiation loss from these nonlinear high-\( Q \) resonances would just be governed by the textbook theory of dissipation in nonlinear high-\( Q \) resonators (49–51). 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 (52), dissipative phase transitions (53), and modest amplitude squeezing (antibunching of light) in the cavity mode (54–56). This 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 medium of the cavity in Fig. 1 (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 \( \omega_a \) (57), and so do 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. 2, 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, by populating the resonance with a coherent state with a mean of \( \bar{n} \) photons and a 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 (as it always is) due to some finite external loss rate, denoted \( 2\kappa \) (Fig. 2), one expects the variance in photon number to rapidly drop (well below the mean), leading to the photon-number-squeezed light described earlier. Thus, even though the BIC is never perfect, we will find that impressively good approximations can still emerge in realistic settings. That said, if the background loss is too high, these quantum optical effects will be washed out, as the nonlinear loss looks linear (independent of photon number); see the black curves of Fig. 2, where one would expect the usual physics of dissipation in which nonclassical states become classical.

A. Theory of Nonlinear Radiation Loss. 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. We emphasize that this analytical theory we develop, along with its remarkable consequences, has also been verified (SI Appendix) by standard numerical simulation tools for open-quantum systems. First, we state the key results of this theory. (Detailed derivations are provided in SI Appendix.) The family of systems to which our theory immediately applies is a driven Kerr nonlinear resonance, coupled to some number \(N\) of reservoirs (also called continua; they can be associated with radiation or absorption loss\(^3\)). Coupling of the resonance to the reservoir leads to dissipation of the resonance. For this class of systems, the Hamiltonian takes the form:

\[
\begin{align*}
H/\hbar &= \omega_0 a^\dagger a + \frac{\beta}{2} a^\dagger a^2 + \alpha(t) a + \alpha^*(t) a^\dagger \\
&\quad + \sum_{\sigma=1}^{N} \int \frac{d\omega}{2\pi} \omega \sigma_\sigma(\omega) a^\dagger \sigma_\omega(\omega) \\
&\quad + \sum_{\sigma=1}^{N} \int \frac{d\omega}{2\pi} i \left( K_{\sigma,\omega}(\sigma_\omega(\omega)a^\dagger - K_{\sigma,\omega}^*(\sigma_\omega^*(\omega)a) \right).
\end{align*}
\]

[1] Here, \(a\) is the annihilation operator of the resonance with frequency \(\omega_0\). The Kerr nonlinearity of the resonance manifests as the second term in the Hamiltonian \((49, 51)\), leading to a "photon-number–dependent resonance frequency." Given \(n\) photons in the resonator, the energy to add another is \(h\omega_{n+1,n} \equiv h\omega_0(1+2\beta n)\), where \(\beta\) is a (dimensionless) nonlinear coefficient. The coherent-drive strength is \(\alpha(t)\) and is not restricted to be monochromatic. This driving neglects frequency and amplitude noise, but, due to the particulars of the systems we consider, this is justifiable\(^3\). The continuum modes are described as usual by a set of harmonic oscillators \((58)\); for the \(\sigma\) reservoir, the continuum modes are labeled by their frequency \(\omega\) and annihilation operator \(\sigma_\omega(\omega)\).

The resonance couples to the continuum through the last term in the Hamiltonian with coupling coefficient \(K_{\sigma,\omega}(\omega)\). This \(K_{\sigma,\omega}\) is the in-coupling function from the \(\sigma\) reservoir: Given a monochromatic input (at frequency \(\omega\)) from reservoir \(\sigma\) into the resonator, the classical amplitude of the resonance, denoted \(a(\omega)\), is proportional to \(K_{\sigma,\omega}(\omega)\). 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 \((52, 59, 60)\), which are well known to accurately describe many widely used photonic architectures. In SI Appendix, we provide explicit forms of \(K_{\sigma,\omega}(\omega)\) for a few common photonic architectures that have BICs (the BIC is encoded in a zero/minimum of \(K_{\sigma,\omega}(\omega)\) as a function of frequency).

The Hamiltonian of Eq. 1 is exact but not generally solvable. Now, we employ the sole approximation of our theory: The bandwidth of the reservoirs, \(\Delta\omega_\sigma\), is much larger than the inverse "response" timescale of the reservoir \(T^{-1}\). The physical quantity which sets \(\Delta\omega_\sigma\) and \(T^{-1}\) varies from architecture to architecture, but it frequently holds. This approximation, which says that the system is only weakly non-Markovian, \((50, 61)\), is already satisfied by the structures of Fig. 1 for the parameters of Figs. 2–6 (more details are provided in SI Appendix, page 7). Taking this approximation yields an equation of motion for the reduced density matrix of the cavity, denoted \(\rho\). It is (SI Appendix):

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

where \(H_K + H_{\text{drive}} = \omega_0 a^\dagger a + \frac{\beta}{2} a^\dagger a^2 + \alpha(t) a + \alpha^*(t) a^\dagger\) is responsible for the conservative parts of the evolution of the resonance, and the dissipator \(D\) is defined through its matrix elements as \((m, n\text{ are Fock states}):\)

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

The function \(K_f(\omega)\) is the "loss function" or "frequency-dependent loss" of the cavity and is directly connected to the

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\(^3\) Most driving lasers have some "added noise" in amplitude (as well as frequency). For the particular physics we propose in our manuscript, the narrow-band resonator is illuminated by a short pulse (for the purpose of loading a coherent state of many photons over a short period of time). Due to the large bandwidth of the pulse compared to any resonances, frequency noise is not particularly detrimental. Similarly, amplitude noise is also not particularly detrimental. This is because in the systems we introduce in this work there is a remarkable feature: Any coherent state with more photons than the Fock-order \(n_0\) will eventually turn into a Fock state of that order, regardless of its initial value—so it is as if the system is immune to noise in the initial amplitude. More broadly, it is in fact quite possible to get existing pulsed lasers of the relevant energy operate at near shot-noise–limited values, either by active stabilization or even by attenuation.
incoupling function by a Kramers–Kronig relation, as:

$$K_i(\omega) = \sum_\sigma K_{i,\sigma}(\omega), \text{ with } K_{i,\sigma}(\omega) \equiv i \int \frac{d\omega'}{2\pi} \frac{|K_{\sigma,\sigma}(\omega')|^2}{\omega' - \omega - \eta}.$$  \[4\]

where $\eta$ is an infinitesimal. The function $K_i$ is related to the “resonance-frequency-dependent” factor of Eq. 2 by $Q^{-1}(\omega) = 2\text{Re}K_i(\omega)/\omega$. The relation of Eq. 4 enforces the intimate connection between leakage and incoupling. Eqs. 2–4 summarize our quantum theory of nonlinear outcoupling: They tell us that the evolution of the quantum state of a resonance—incorporating photon probabilities, field correlations ($\langle q(1) \rangle$), intensity correlations ($\langle q_i^2 \rangle$), and so on—are strongly controlled by the photon number ($n$) dependence of quantities like: $K_i(\omega_{\text{in}} + \omega_n) = K_i(\omega_n + 1/2\Delta_n)$, 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 an approximation called a “white noise” approximation. In this approximation, one neglects the frequency-dependence of the continuum coupling, thus neglecting the possibility that the resonator $Q$ depends on its frequency, as it does for the resonators of Fig. 1.5.

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

$$\text{Re} K_i(\omega_{n_0,n_0-1}) = 0. \tag{5}$$

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

$$n_0 = \frac{\Delta_0}{2\beta \omega_0} + 1, \quad \tag{6}$$

where $\Delta_0 \equiv \omega_0 - \omega_0$ 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 (Fig. 2) 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 in particular, many squeezing architectures tend to be limited to 3 dB squeezing for the intracavity state (62)—and thus this regime is still very interesting.

The dynamics of various quantum states undergoing the nonlinear radiation loss of Fig. 2 are illustrated in Fig. 3, in a “phase space plot” where the two variables being plotted 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 toward 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. 3).

It is worth pausing to emphasize the strikingness 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 this 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) (52). Further, in conventional bistability, Fock states do not arise (49).

In the presence of background losses, the BIC becomes imperfect and the bifurcation trajectories in the top panel of Fig. 2 softens, as the state at $n = 10$ is no longer stuck—it slowly leaks and progresses toward the vacuum state. This is shown in the Bottom Panel for $K_i = 10^{-7}\omega_0$ (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 very strongly 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 80% (about 7 dB) below the shot noise level, which is a very high degree of squeezing.

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5Our theory is mapped to the standard theory of damping of a resonator with the identification $K_i(\omega) = \Delta a^2$, with $\Delta$ the amplitude decay rate yielding $K_i = \Delta$ and reducing Eq. 2 to the standard master equation for a damped cavity as in ref. 58.

6There is no linear component because loss has to be nonnegative.
especially for a state with a mesoscopic photon number like this one.

2. 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 (n0 on the order of 10 or so, with Δn ≤ 1). Such platforms, as per Eq. 6, should be realized using sizeable single-photon nonlinear strengths β. Fig. 4A illustrates such a system, formed by coupling excitons to a resonator which is then coupled to a waveguide (63). The parameters for the resonator–waveguide system are very similar to those in ref. 48. The coupling between exciton and resonator leads to exciton–polaritons. Such excitations are the subject of many recent experiments not limited to, but including and resonator leads to exciton–polaritons. Such excitations are the very similar to those in ref. 48. The coupling between exciton (63). The parameters for the resonator–waveguide system are splittings exceeding the decay rate by 1 to 2 orders of magnitude, especially for a state with a mesoscopic photon number like this one.

(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 (65), and (C) present the strong nonlinearities that are needed. The nonlinearities already present (β ∼ 10−5ω) 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 (54). 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) (53, 55). 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 (64) similar to a lower-bound associated with exciton dispersion, discussed for a different material platform (63). 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. 4B. 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 n0. For example, in this case, due to the strong nonlinearities
of exciton–polaritons, the pulse energies incident on the system are quite small, corresponding to subfemt joule energies. 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. 4D through the Husimi Q functions \( Q(\alpha) = \langle \alpha | \rho(\alpha) | \alpha \rangle / \pi \), 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 (42). Then, after long times, the system approaches a Fock state. Fig. 4C shows the simulated photon probabilities, affirming the intuition of Fig. 1A (assuming no external loss). 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 nanoseconds). 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 nonintegral, 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. 4D 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. 4D 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 \( Q = 5 \times 10^5 \)), 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 (\( Q \approx 10^6 \)—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 (63). In other words, even within optics, we already expect the nonlinear dissipation we developed to be testable.

**Experimental Signatures.** We now address the question of how to detect the generated quantum states. In Fig. 5, 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) (53, 55, 56). Fig. 5A 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 \( g^{(2)}(0) = (\langle a^\dagger a^\dagger a a \rangle / \langle a a \rangle)^2 = 1 - 1/n \), 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 toward 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. That said, the issue of limited contrast described here can be ameliorated by measuring higher-order correlation functions, such as \( g^{(4)} \) and \( g^{(6)} \), which have previously been demonstrated (see for example refs. 66 and 67). For example, fourth-order correlation functions for an \( n \)-photon Fock state would yield \( g^{(4)}(0) = (1 - 1/n)(1 - 2/n)(1 - 3/n) \), which would be 0.5 for the 10-photon Fock state.

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 detection technique, referred to as photon-induced near-field electron microscopy (PINEM). The basic theoretical description and experimental implementations are discussed in refs. 65, 68–71. The idea is illustrated in Fig. 5B: an energetic electron (typical kinetic energy \( E_0 \sim 100 \) to 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
3. Discussion and Outlook

In optics, the realization of the nonlinear loss of Fig. 1 and the \(n\)-photon BIC effect predicted in Fig. 2 should already be within reach. Already Figs. 2 and 3 indicate possibilities for combining strong exciton–polariton nonlinearities with photonic bound states in the continuum to realize mesoscopic light states with \(\Delta n \sim 1\). As discussed above, and also more extensively in SI Appendix, 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 a good approximation of a BIC) which appears for a certain index of refraction and Kerr nonlinearity (third-order optical nonlinearity). The types of architectures to realize the former (illustrated in Fig. 1) have already been realized several times now (without Kerr nonlinearity). 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 (74) or photonic crystal cavities (75) 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). Moreover, such systems have the advantage of being implementable even at room temperature, due to the optical photon frequency being large compared to thermal energies, and the material losses being low even at room temperature.

In Fig. 6, we back up this assertion by showing how existing Kerr nonlinear nanophotonic systems (where strong Kerr effects have been experimentally observed, see refs. 72, 73), when illuminated by existing optical pulses, can lead to very large squeezings even for modest limiting quality factors. Here, the pulse energies employed to generate the initial state and achieve large squeezing are in the range of 10 to 20 \(\mu\)J (and the pulse durations in this example are on the order of 150 fs), which is well within the range of existing table-top pulsed lasers. For example, if the limiting quality factor due to external loss is \(10^5\), roughly 7 dB squeezing is possible, which would exceed any direct observation of nanophotonic squeezing if measured. More strikingly, for an attainable limiting \(Q\), like \(10^6\), over 10 dB is possible.

To understand the scale of this result, it is worth describing some of the recent progress in nanophotonic squeezing of light. We should mention that almost all squeezing in nonlinear optics proceeds not by nonlinear loss engineering, as we have proposed here, but by techniques such as optical parametric amplification and four-wave mixing in second-order and third-order nonlinear media. Further, states produced by these techniques tend to not be photon-number, or intensity squeezed, as we consider here, but quadrature-squeezed, producing squeezed vacuum states (in other words, they are a different type of state, and so it is not a direct comparison). And of course, these more “conventional” nonlinear optics approaches do not produce Fock states (and to our knowledge, the approach posed in this work is a purely nonlinear optics approach that could produce multiphoton Fock states, deterministically). Those caveats aside, let us compare the squeezing magnitudes predicted here to what has been observed. The predicted squeezing magnitudes in Fig. 6 for the lowest levels of external loss feasible in silicon (76) correspond to 13 dB of squeezing. The largest “inferred” squeezings that have been measured are all very recent and are between 8 and 11 dB using these standard approaches (43, 77, 78) (the observed squeezings

8Assuming 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):

\[
P_m = |g^{(n)} - m\hbar\omega|/|g^{(0)}|S_{E}\left|\langle E^{(2)}\rangle/\langle E^{(2)}\rangle\right|\sqrt{S_{E}^{(2)}}|g^{(0)} - m\hbar\omega|,
\]

where \(S_{E}\) is the evolution operator describing the electron–photon interaction. It is given by \(S_{E} = \exp\left[\int d\nu_{E}\nu_{E}\right]\), where \(\nu_{E}\) is an operator lowering (raising) the electron energy by one unit. Namely, \(E^{(0)} - \hbar\omega = |g^{(0)} - (m + 1)\hbar\omega\) with \(bb^{\dagger} = b^{\dagger}b = 1\). 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 \(m\hbar\omega\). 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 \(m\hbar\omega\)).

9In principle, it should be possible to take approaches outside of nonlinear optics, such as through cavity QED, and extend approaches used at microwave frequencies, such as ref. 16, but to our knowledge, proposals along these lines have not been substantially pursued.
are less than that, due to outcoupling losses, but presumably, the on-chip squeezing is close to the inferred squeezing).

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 (79) or Rydberg atoms (80) 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 Appendix, section VI), 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—it would enable for example the direct conversion of a microwave probe into a Fock state of a Kerr nonlinear microwave resonator (formed by coupling a Josephson junction with modest anharmonicity to a linear microwave resonator). From a nonlinearity and external loss perspective, we suspect that the capabilities are more than present to demonstrate Fock-state and extreme squeezing 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 (51). Beyond providing a useful proving ground for the concepts developed here, our technique does provide a path to easily tune the Fock state order (just change the detuning) and achieve fairly high Fock-state numbers with high fidelity.

To summarize, we have introduced a form of nonlinear dissipation that induces many-photon bound states and enables deterministic generation of large photonic Fock states. 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 continua 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 $\chi^{(3)}$ (81), exciton–polaritons, where Kerr comes from Coulomb interactions (82), superconducting circuits, where Kerr comes from nonlinear inductance (83), magnons, where Kerr comes from magnon–magnon interaction (84), Rydberg atoms, where single-photon nonlinearities arise from Rydberg blockade (80), and cavity-QED systems, where single-photon nonlinearities arise from photon blockade (49).

Our work establishes a connection between two highly active fields: 1) radiation loss engineering, which has primarily been explored in classical optics in the context of BICs, exceptional points, and non-Hermitian photonics (44, 47, 85–87) and 2) quantum-state engineering, where the use of nonlinear dissipation to engineer quantum states is well appreciated (see, e.g., refs. 34–36). In doing so, our work points to a 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 (88). 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 toward stabilizing other quantum optical states that are of interest to the community (Schrödinger cat states, GKP states, cluster states, and the like).

Given the generality and rich variety of effects introduced here, we expect that the development of physical platforms to realize them may provide many exciting areas for discovery in quantum optics, nonlinear optics, nanophotonics, and beyond.

Data, Materials, and Software Availability. Code data have been deposited in GitHub, https://doi.org/10.5281/zenodo.7310927.

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