Intense Squeezed Light from Lasers with Sharply Nonlinear Gain at Optical Frequencies

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Nonclassical states of light, such as number-squeezed light, with fluctuations below the classical shot noise level, have important uses in metrology, communication, quantum information processing, and quantum simulation. However, generating these nonclassical states of light, especially with high intensity and a high degree of squeezing, is challenging. To address this problem, we introduce a new concept which uses gain to generate intense sub-Poissonian light at optical frequencies. It exploits a strongly nonlinear gain for photons which arises from a combination of frequency-dependent gain and Kerr nonlinearity. In this laser architecture, the interaction between the gain medium and Kerr nonlinearity suppresses the spontaneous emission at high photon number states, leading to a strong "negative feedback" that suppresses photon-number fluctuations. We discuss realistic implementations of this concept based on the use of solid-state gain media in laser cavities with Kerr nonlinear materials, showing how 90% squeezing of photon number fluctuations below the shot noise level can be realized.

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Nonclassical states of light such as squeezed states and Fock states are famous for maintaining noise levels below the classical "shot noise" limit. Therefore, these states are considered important for applications in precision measurement [1,2], optical communication [3,4], quantum information processing, and quantum simulation [5–11]. Intense coherent light is readily produced by lasers and masers. However, generation of squeezed light necessarily relies on some types of nonlinear mechanism, which is often a form of nonlinear loss or gain.

In the microwave regime, various mechanisms of nonlinear gain have been explored as tools to modify the quantum statistics of light. For example, experiments with so-called "micromasers" pumped by excited Rydberg atoms have been used to generate sub-Poissonian light in a microwave resonator [12–16]. Similar physics can also be exploited by superconducting qubits to deterministically generate Fock states up to order 10 [17,18]. Robust effects like photon blockade have also been used as sources of single photons [19] and sub-Poissonian light [20]. Additionally, it has been shown how light-matter systems in the so-called "deep strong coupling" regime can be used to create an effective N-photon blockade which lends itself to the generation of N-photon Fock states when used as a maser gain medium [21].

Despite the effectiveness of the described tools in the microwave regime, many of these techniques do not readily extend to optical frequencies. In the optical regime, both second and third-order nonlinear interactions, as well as nonlinear dissipation mechanisms such as two-photon absorption, have been used to squeeze light below the shot noise limit [22–30], with many promising new approaches for very strong reduction of intensity noise, especially for lower intensity light [31–38]. However, these methods have typically been limited to the generation of sub-Poissonian light which is only 50% below the shot noise limit, with notable exceptions limited to specialized cases (e.g., requiring the use of solitons in fibers) [30]. Furthermore, by and large, many of these nonlinear techniques work either at very low powers (e.g., below the threshold of an optical parametric oscillator), or moderate powers. It is still an open question of how to realize strongly squeezed light at macroscopic intensities (e.g., watt scale and beyond). In general, high power optical sources are produced by amplification through gain media, which either amplify fluctuations or, at best, produce approximations to coherent states (but often have substantial excess noise). As a result, the generation of intense, and strongly sub-Poissonian optical radiation remains a great challenge.

Here, we introduce a mechanism of sharply nonlinear gain which can be used to create intense sub-Poissonian light at optical frequencies. We show how this nonlinear gain can be realized by incorporating Kerr nonlinearity into a traditional solid state laser architecture. We detail how the nonlinear gain acts as a form of "supersaturation," leading to new phenomena like nonlinear power curves and the suppression of transient relaxation oscillations. Furthermore, we show how this nonlinear gain can lead to macroscopic states of light with intensity fluctuations

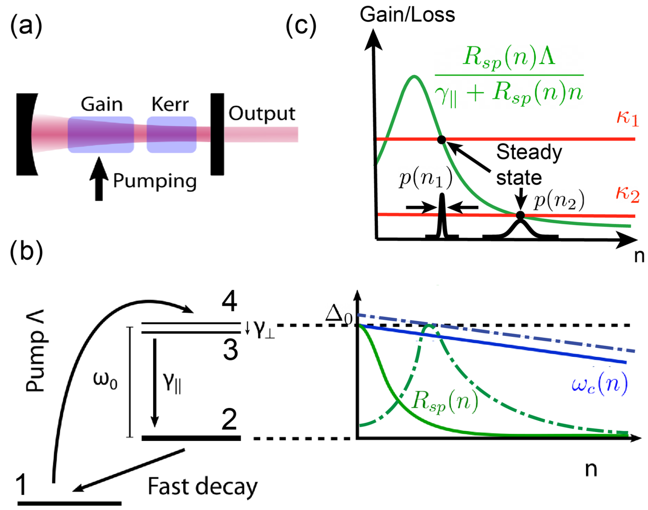

FIG. 1. A laser based on Kerr nonlinearity. (a) A pumped gain medium coupled to a cavity with Kerr nonlinearity. (b) Four-level atoms in the gain medium, with lasing transition between level 3 and 2 with frequency ω_0. The dashed lines show the dependence of the cavity resonance frequency $\omega_c(n)$ (blue) and the spontaneous emission rate $R_{\rm sp}$ (green) on the light intensity of an "off-resonant Kerr laser" ($\omega_c - \omega_0 = \Delta_0$), and the solid lines are those of "resonant Kerr laser" ($\omega_c = \omega_0$). (c) The photon number probability distribution [$p(n_1)$ and $p(n_2)$] depends on the angle between the gain ($\{R_{\rm sp}(n)\Lambda/[\gamma_\| + R_{\rm sp}(n)n]\}$) and the loss ($\kappa_1$ and κ_2) at their intersection point (or the steady state). Because the gain intersects with the loss κ_1 at intensity n_1 more steeply than with the loss κ_2 at intensity n_2, the probability distribution at n_1 is more squeezed, thus, the photon number variance is reduced.

90% below the shot noise level, which corresponds to sub-Poissonian (number-squeezed) states of light that have no classical analog [39,40]. This is in strong contrast to conventional high-power lasers, which produce coherent states or states with substantial excess intensity noise.

A laser based on Kerr nonlinearity.—The concept for the "Kerr laser" is illustrated in Fig. 1(a). The system consists of a pumped gain medium in a cavity containing a Kerr nonlinear medium. The Hamiltonian of just a cavity with an embedded Kerr medium (so no gain) is $H_{\rm Kerr}/\hbar = \omega_c \hat{a}^\dagger \hat{a} - (\beta/2)\hat{a}^{\dagger 2}\hat{a}^2 = \omega_c \hat{a}^\dagger \hat{a} + (\beta/2)\hat{a}^\dagger \hat{a} - (\beta/2)(\hat{a}^\dagger \hat{a})^2$ (The conventional expression of Kerr Hamiltonian is $H_{\rm Kerr}/\hbar = \omega_c \hat{a}^\dagger \hat{a} + (\beta/2)\hat{a}^{\dagger 2}\hat{a}^2$. However, in this Letter, we negate the sign of Kerr nonlinear strength for brevity. The justification is that, in transparent materials, the index of refraction increases as a function of intensity, which leads to a decrease in the cavity resonance frequencies. This decrease corresponds to a negative value of β.)($\hat{a}$ and $\hat{a}^\dagger$ are the annihilation and creation operators of the cavity mode, β is the Kerr nonlinear strength of a single photon, and ω_c is the cavity frequency) [40,41]. In such a system, the energy needed to add one more photon into a cavity which already contains $n-1$ photons is $\hbar(\omega_n - \omega_{n-1}) = \hbar\omega_c(n) = \hbar(\omega_c - \beta n)$. The cavity resonance frequency depends linearly on the photon number as shown in Fig. 1(b). Hence, at a certain Fock state $|n\rangle$ in the cavity, the gain medium and the cavity are resonant [Fig. 1(b)]. Therefore, the spontaneous emission rate $R_{\rm sp}$ is maximal for photon numbers where the cavity and the gain are resonant, but suppressed for photon numbers where they are detuned. As a result, the cavity "prefers" photon numbers which are close to resonance, and suppresses photon numbers away from them, leading to a reduction in the photon number fluctuations and, thus, squeezing [1]. This type of gain can be thought of as a "supersaturable" gain, which decreases more sharply with n from the equilibrium point (of balanced gain and loss), as compared to conventional saturable gain [21]. Surprisingly, this approach can lead to very large photon-number squeezing in the cavity (approaching 10 dB), as we will now show.

For concreteness, we consider the gain medium to consist of typical four-level atoms (with fast nonradiative decay to render it as an approximate two-level system), such as Nd:YAG. In such systems, the polarization decay rate $\gamma_\perp$ is much larger than the inversion decay rate $\gamma_\|$ and cavity leakage rate κ. Hence, we can adiabatically eliminate the polarization degree of freedom, resulting in two Heisenberg-Langevin equations of motion for the atomic inversion S and the light intensity n (See Supplemental Material [42] for detailed calculation, which includes Refs. [43–46]).

$$\dot{n} = [R_{\rm sp}(n)S - \kappa]n + F_n, \tag{1a}$$

$$\dot{S} = \Lambda - [\gamma_\| + R_{\rm sp}(n)n]S + F_S. \tag{1b}$$

The spontaneous emission rate $R_{\rm sp}(n) \equiv 2g^2\gamma_\perp/[\gamma_\perp^2 + \Delta(n)^2]$, where $\Delta(n)$ is the detuning between the atomic frequency and the (number-dependent) resonance cavity frequency, g is the coupling strength between the lasing transition and the cavity mode, and Λ is the pump rate of the gain medium. Quantum fluctuations are incorporated into the rate equations through the Langevin forces F_n and F_S whose correlation functions are $\langle F_\mu(t)F_\nu(t')\rangle = \langle 2D_{\mu\nu}\rangle\delta(t-t')$, with diffusion coefficients given by $\langle 2D_{nn}\rangle = \langle R_{\rm sp}(n)Sn + \kappa n\rangle$, $\langle 2D_{SS}\rangle = \langle \Lambda + \gamma_\| S + R_{\rm sp}(n)nS\rangle$, and $\langle 2D_{Sn}\rangle = \langle 2D_{nS}\rangle = -\langle R_{\rm sp}(n)nS\rangle$ [21,43,46].

First, we consider a resonant Kerr laser, where the empty cavity and the gain are in resonance [$\Delta(n) = \beta n$]. When the pump rate exceeds the threshold value $\Lambda_{\rm th} = \kappa\gamma_\|\gamma_\perp/(2g^2)$, the steady state cavity photon number n_s becomes nonzero, and depends on the relative pump rate $r = \Lambda/\Lambda_{\rm th}$ as shown in Fig. 2(a). Compared to the "linear laser" ($\beta = 0$), the steady-state intensity of the Kerr laser increases less rapidly with pump rate. This behavior is expected because, as the intensity increases, the cavity frequency detunes from the atomic resonance causing a sharper reduction in stimulated emission via $R_{\rm sp}(n)$ than saturable gain. The effect becomes

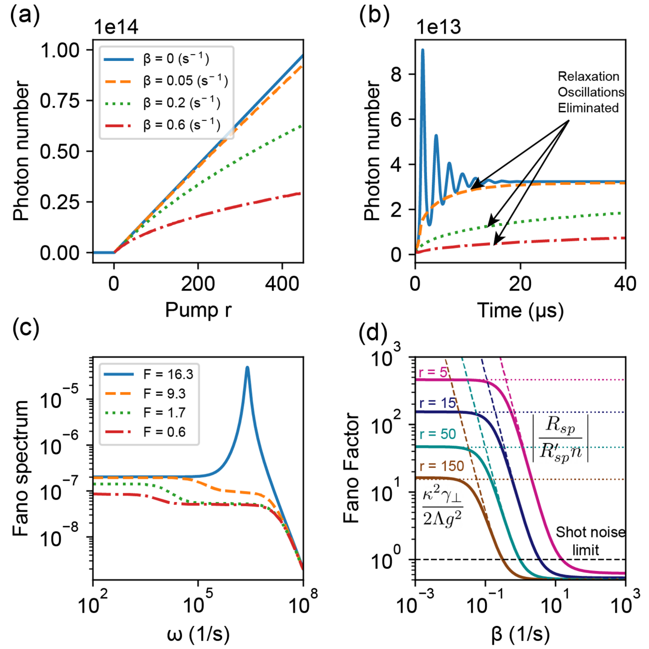

FIG. 2. Output characteristics of a resonant Kerr laser. Parameters used for an Nd:YAG based system are $\gamma_\perp = 10^{12}\ \mathrm{s}^{-1}$, $\gamma_\parallel = 4.34 \times 10^3\ \mathrm{s}^{-1}$, $\kappa = 10^7\ \mathrm{s}^{-1}$, and the coupling coefficient $g = 10^2\ \mathrm{s}^{-1}$. For figures (a), (b), and (c), lines with the same color and line style have the same β values. (a) Steady-state photon number of a Kerr laser as a function of relative pump rate r for different nonlinear coefficients. (b) Time evolution of photon number at different nonlinear coefficients and at fixed relative pump rate $r = 150$. (c) The Fano spectrum [noise spectrum in frequency domain (ω) or intensity] at different nonlinear coefficients and at fixed relative pump rate $r = 150$ and their corresponding Fano factors F. (d) Fano factor F at different pump rates as a function of nonlinear coefficient. At small β value, F can be approximated by $(\kappa^2\gamma_\perp/2\Lambda g^2)$ (dotted line). For large enough β, F approximately equals to $|R_{\rm sp}(n_s)/R'_{\rm sp}(n_s)n_s|$ (dashed line). As β becomes much larger than $\sqrt{(\kappa g^2\gamma_\perp/2\Lambda\gamma_\parallel)}$, F approaches the sub-Poissonian value of 0.5.

significant when the Kerr nonlinear shift frequency at steady state βn_s is comparable to the gain bandwidth $\gamma_\perp$.

The nonlinearity affects fluctuations from the steady state even more drastically. Small fluctuations of the intensity δn and the inversion δS are governed by the Heisenberg equations

$$\begin{pmatrix} \delta\dot{n} \\ \delta\dot{S} \end{pmatrix} = \begin{pmatrix} n_s\kappa\frac{R'_{\rm sp}(n_s)}{R_{\rm sp}(n_s)} & R_{\rm sp}(n_s)n_s \\ -n_s\kappa\frac{R'_{\rm sp}(n_s)}{R_{\rm sp}(n_s)} - \kappa & -\frac{\Lambda R_{\rm sp}(n_s)}{\kappa} \end{pmatrix} \begin{pmatrix} \delta n \\ \delta S \end{pmatrix} + \begin{pmatrix} F_n \\ F_S \end{pmatrix}, \tag{2}$$

where $R'_{\rm sp}(n_s)$ is the first derivative of $R_{\rm sp}$ evaluated at the mean photon number n_s. Transient behavior can be described by taking expectation values of this system, which is equivalent to removing the zero-mean Langevin force terms. It has long been known that most solid-state and semiconductor lasers, when perturbed from equilibrium (e.g., by gain modulation, mechanical fluctuations, etc.), exhibit decaying characteristic intensity and inversion oscillations [47], which are referred to as "relaxation oscillations" and are considered undesirable in many applications. The Kerr laser resists fluctuations from the mean, so these relaxation oscillations are suppressed [Fig. 2(b)]. Even a strength of nonlinearity which has a minimal effect on the intensity (i.e., $\beta = 0.05\ \mathrm{s}^{-1}$) leads to a complete suppression of relaxation oscillations. [The fractional change in the index of refraction in this case is on the order of 1% which, while large for continuous-wave excitation, is feasible in pulsed settings, and we expect the treatment here (which is continuous wave) to extend to that case].

In addition to changing the mean field properties of the laser, the Kerr nonlinearity also influences the quantum statistics of the laser steady state. By solving Eq. (2) in the frequency domain ω, we obtain the frequency dependent spectrum of the intensity noise

$$\langle \delta n^\dagger(\omega)\delta n(\omega)\rangle = \frac{(\omega^2 + \Gamma_0^2)2\kappa n_s}{(\Omega^2 + \Gamma^2/4 - \omega^2)^2 + \Gamma^2\omega^2}. \tag{3}$$

The relaxation oscillation frequency Ω is defined by $\Omega^2 \equiv R_{\rm sp}(n_s)n_s\kappa\{n_s[R'_{\rm sp}(n_s)/R_{\rm sp}(n_s)] + 1\} - \frac{1}{4}\{[\Lambda R_{\rm sp}(n_s)/\kappa] + n_s\kappa[R'_{\rm sp}(n_s)/R_{\rm sp}(n_s)]\}^2$, and $\Gamma \equiv \Gamma_0 - [R'_{\rm sp}(n)/R_{\rm sp}(n)]n\kappa$ is the relaxation oscillation decay rate, where $\Gamma_0 \equiv \Lambda R_{\rm sp}(n_s)/\kappa$ is the decay rate without nonlinearity. This noise spectrum represents the intensity fluctuations associated with individual Fourier components of the laser field which fluctuates about its steady state. Such fluctuations may be studied with a photodiode and electronic spectrum analyzer. For the linear resonator, the relative intensity noise spectrum sharply peaks around Ω, indicating relaxation oscillations driven by quantum noise. As the nonlinear strength increases, this oscillation peak is suppressed and eventually eliminated, consistent with the transient behaviors shown for the same parameters [Fig. 2(c)].

The steady-state photon number variance $(\Delta n)^2$ can be obtained by integrating the noise spectrum: $(\Delta n)^2 = (1/\pi)\int_0^\infty d\omega \langle \delta n^\dagger(\omega)\delta n(\omega)\rangle$. The nonlinear gain leads to sizable reductions in the intensity noise, which we characterize by the Fano factor $F = (\Delta n)^2/n_s$ ($F = 1$ for Poissonian light). When βn_s is comparable to $\gamma_\perp$, F is suppressed by 2 orders of magnitude compared to that of a linear laser, and less than 1 [Fig. 2(c)].

Analytically, $F = (\kappa/\Gamma)\{[\Gamma_0^2/(\Gamma^2/4 + \Omega^2)] + 1\}$, which approximately equals to κ/Γ over a large range of pump rates ($\Lambda_{\rm th} < \Lambda < 1000\Lambda_{\rm th}$). The Kerr nonlinearity only affects F significantly when βn_s is comparable to $\gamma_\perp$. Thus, under that condition,

$$F \approx \left| \frac{R_{\rm sp}(n_s)}{R'_{\rm sp}(n_s) n_s} \right|. \quad (4)$$

F can be decreased by making both n_s and $|R'_{\rm sp}(n_s)/R_{\rm sp}(n_s)|$ as large as possible. Physically, this corresponds to situations in which $R_{\rm sp}$ (and, thus, gain) decreases as sharply as possible with an increasing photon number. In the case of a resonant Kerr laser, as β increases, $R_{\rm sp}$ will fall more sharply with higher intensity, but the sharp falling gain will intersect the loss at lower intensity. This behavior causes F to saturate at a sub-Poissionian value of 0.5.

Further noise suppression with detuning between the gain medium and zero-photon cavity.—As we have discussed above, the minimum value of the Fano factor is dictated by both the steepness of $R_{\rm sp}$ and the mean photon number. To further suppress the intensity noise, the steep region of $R_{\rm sp}$ should ideally occur at the highest photon number possible. This effect can be realized by setting the $n = 0$ cavity resonance frequency to be higher than the atomic frequency (off-resonant Kerr laser). Because of the Kerr effect, the detuning value decreases with light intensity, so the cavity and the gain medium become resonant at photon number $n_{\rm res} = (\omega_c - \omega_0)/\beta$. At $n_{\rm res}$, the spontaneous emission rate is also maximal [Fig. 1(b)]. The higher the detuning value Δ_0, the more $R_{\rm sp}$ is shifted. We set Δ_0 to be of 3% of the 1064 nm wavelength gain frequency (about ten times the gain bandwidth, which, of course, leads to a much larger threshold). The cavity can be further detuned from the gain at the expense of a higher pump rate to ensure that the gain of the empty cavity exceeds the cavity leakage rate.

For the off-resonant Kerr laser, $R_{\rm sp}(n) \equiv 2g^2\gamma_\perp/[\gamma_\perp^2 + (-\Delta_0 + \beta n)^2]$. The gain of the cavity, defined as $G(n) = R_{\rm sp}(n)\Lambda/[\gamma_\parallel + R_{\rm sp}(n)n]$, depends nonmonotonically on the light intensity [Fig. 3(a)]. Therefore, the gain can intersect with the loss at one, two, or no point [with conditions for each scenario shown in Fig. 3(b)]. In the case of two intersecting points, only the solution at the negative differential gain region is stable [Fig. 3(a)]. The other solution is unstable under small perturbations because the positive differential gain will push the photon number further from its mean value. As such, we consider only the stable solution when analyzing the intensity noise.

The noise behavior of the off-resonant Kerr laser is shown in Fig. 3(c). When β is negligible, the Fano factor is $\kappa^2\Delta_0^2/(2\Lambda g^2\gamma_\perp)$, which is larger than the one of the resonant linear laser with the same parameters even though the spontaneous emission rate in the off-resonant laser is suppressed. This effect can be explained by the overwhelming reduction of the mean photon number in the off-resonant linear laser due to the large detuning Δ_0. As β increases, there is a region where $R'_{\rm sp}$ is positive, unlike in the resonant case [Fig. 2(c)]. Therefore, the noise first increases, reaching a value several orders of magnitude higher than the shot noise limit. In the sharply changing $R_{\rm sp}$ region, we can again estimate the noise intensity by Eq. (4). When the cavity and atoms are in resonance, or $\beta n_s - \Delta_0 = 0$, $R'_{\rm sp}$ is zero, thus, we expect a local maximum value of F. When the total frequency shift is equal to the gain bandwidth ($|\beta n_s - \Delta_0| = \gamma_\perp$), F is minimized: at this point, the detuning value is small enough that it does not significantly reduce the steady-state light intensity. The minimum obtainable Fano factor in this laser architecture is $\gamma_\perp/(\gamma_\perp + \Delta_0)$. For the parameters we consider, this gives $F \lesssim 0.1$ (over 10 dB noise reduction). This minimum is realized when β is set such that the cavity at equilibrium is detuned $\gamma_\perp$ lower than the atomic frequency. Such parameters can produce light which is more than 90% below the shot noise limit, but at a photon number of 10^{12}, which is clearly macroscopic.

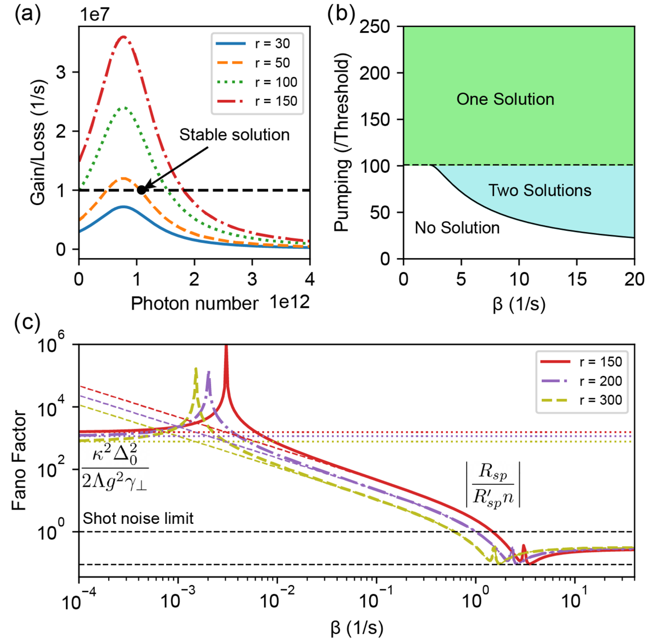

FIG. 3. Off resonant nonlinear Kerr laser with Nd:YAG gain medium and the detuning value between the gain medium and the zero photon cavity being $\Delta_0 = 10^{13}$ s^{-1}. (a) The gain and loss rate as functions of light intensity at different pump rates and at fixed nonlinear coefficient $\beta = 10$ s^{-1}. (b) The number of steady solutions depends on both the nonlinear coefficient and pump rate. (c) Fano factor F at different pump rates as a function of nonlinear coefficient. F reaches a global minimum value of $[\gamma_\perp/(\gamma_\perp + \Delta_0)]$ when the detuning value between the cavity and the gain medium $-\Delta_0 + \beta n_s$ equals the gain bandwidth $\gamma_\perp$. At small β value, $F \approx \kappa^2\Delta_0^2/(2\Lambda g^2\gamma_\perp)$ (dotted lines). For large enough β value, $F \approx |R_{\rm sp}(n_s)/R'_{\rm sp}(n_s)n_s|$ (dashed lines).

Discussion and outlook.—Here, we discuss some of the most important considerations relevant to the experimental observation of the effect we proposed. The most important element is that the per-photon nonlinear frequency shift β

be as large as possible compared to the polarization decay rate $\gamma_\perp$. This ratio of parameters dictates the effective sharpness with which the gain drops off at higher photon numbers, leading to steady-state intensity noise reduction. Gain mediums Nd:YAG, Nd:YAP, and Tm:YAG are all strong candidates due to their relatively narrow gain bandwidths set by $\gamma_\perp$. To maximize the nonlinear interaction β, one should use a material with low absorption and high $\chi^{(3)}$ such as GaP, and consider using a small cavity with a high filling fraction of nonlinear material so that the intensity-dependent frequency shift is as large as possible. Note that, while in principle, some of these materials (e.g., GaP) have two-photon absorption, it is quite weak, and may not be detrimental as such effects can lead to further number squeezing [23,24,48]. If two-photon absorption is not desired, then one can use a gain medium such as Er:YAG, Tm:YAG, or Tm:YAP, which have lasing wavelengths below the two-photon absorption cutoffs of Kerr materials such as GaAs and GaP. It is also worth pointing out that stronger Kerr shifts can be achieved at higher intensities. Thus, it is desirable to design a laser cavity that has highly reflective mirrors so that the intracavity power is orders of magnitude higher than the out-coupled power. The cavity parameters used in this Letter are consistent with those which can be realized experimentally.

While, for the sake of concreteness, we have based our discussions around a well-established solid-state laser geometry using a free space optical cavity, the concepts developed here should readily extend to other laser platforms, so long as the gain medium has a low bandwidth and an appropriate amount of Kerr nonlinearity. Additional candidates for narrow bandwidth gain media could include gasses, engineered semiconductor, or quantum well transitions. On-chip platforms have the additional advantage that nanophotonic techniques can be used to engineer small mode volumes and efficient Kerr nonlinearity.

In summary, we have proposed a laser architecture that uses sharply nonlinear gain to generate macroscopic sub-Poissonian light. We have discussed how this architecture can be implemented at optical frequencies using conventional solid state gain materials in conjunction with Kerr nonlinear materials. The parameters we have considered are experimentally accessible and can lead to intracavity light which is 90% below the shot noise limit. Importantly, the minimum achievable noise reduction is set purely by the parameters of the implementation (as opposed to mechanisms such as two-photon absorption, which have an intrinsic squeezing limit). Thus, with further optimization of the gain line shape and Kerr nonlinearity on potentially different platforms, it may be possible to go far below the 10 dB reduction we discussed here, eventually leading to the generation of macroscopic near-Fock states. More broadly, this Letter points toward the further investigation of novel optical nonlinearities as a means to create quantum states of light.

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