Parametric stimulated two-photon emission through a biphotonic cascade

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A parametric two-photon process, involving stimulated emission of two cascaded photons from a bidoped poled material placed inside an optically pumped cavity, is proposed and assessed within the priming photon scheme. We show that, with a specific level configuration for the two dopants A and B, and an appropriate cavity design, stimulated emission from dopant A can provide the priming photons for stimulated two-photon emission from dopant B through a parametric biphotonic cascade. We discuss how this process is driven by a ratchet effect introduced by the simultaneous breakdown of both the space and time inversion symmetries. The optical Stark shifts are shown to considerably impact the efficiency of such a cascaded two-photon process.

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I. INTRODUCTION

The demonstration of a two-photon laser remains both elusive and challenging even after close to 50 years of sustained effort [1–3]. Although some of its most advertised features, such as wide tunability, have been substantially surpassed by the performance of the lasers based on one-photon stimulated emission, new features related to the nonlinear character of the two-photon emission process and the relevance of two-photon states to quantum information [3–5] are now taking the central stage and provide renewed impetus for the two-photon laser studies.

A stumbling issue in this quest is the much smaller efficiency of the two-photon emission compared with that of the competing one-photon emission in multilevel schemes. The need to achieve a critical photon density was recognized already in the 1964 proposal [1], and the use of priming photons was suggested to initiate and enhance the two-photon emission at the expense of one-photon emission. The proposed scheme [1] for the degenerate case made use of an active medium containing two species of fluorescent ions (A and B) of which species A served as the gain medium of a four-level laser emitting at a frequency ω_A, while the species B, also a four-level system, was the active site for two-photon emission at the frequency $\omega_B = \omega_A + \omega_C$, stimulated by the priming photons at ω_A. In this scheme, the population inversion at ω_A and ω_B results from the same light source, and the optical cavity is set up for stimulated emission at the frequency ω_A but blocking it at ω_B. Even in a four-level configuration, complexity of the two-photon transition probability, proceeding through multiple pathways that can interfere and connect the two states via virtual transitions through a relay state, poses some serious problems for its enhancement. Besides, it has been tacitly assumed in the majority of cases that the active B sites possess the spatial inversion symmetry and their states accordingly have definite parities.

Here we propose an approach that differs from Ref. [1] by using an effective two-level system in place of a four-level scheme. This is accomplished using a poled, bidoped, ferroelectric crystal (or a poled dielectric or polymer) that breaks down both the space inversion symmetry and the parity, and thus allows two-photon transitions even in a two-level scheme through a single pathway involving succession of an intrastate and an interstate transition. When such a crystal is pumped optically to create off-equilibrium population inversion, a *ratchet mechanism* [6], set up by the simultaneous breakdown of both the space and time inversion symmetries, further drives and sustains the two-photon emission process. We stress that it is the ratchet mechanism that avoids the multipath quantum interference inherent in the four-level scheme of Ref. [1] by imposing a single path between the two levels.

Over recent years, multiwavelength lasers and coherent sources have been developed using bidoped ferroelectrics containing two different rare-earth dopants [7–13], each providing its own characteristic laser emission upon pumping, together with the second harmonic, sum, or difference frequency generation, provided an appropriate phase-matching condition can be satisfied. Notwithstanding the interest of such schemes for multiwavelength emission, the realization and exploitation of such self-frequency conversion through *in situ* nonlinear processes depend critically on achieving phase matching, a rather formidable problem in this case even with the use of quasi-phase-matching schemes. In contrast, the parametric stimulated two-photon emission we propose here is exempt from such phase-matching restrictions and will in fact be favored over any competing three-wave mixing process that depends on a stringent phase-matching condition, as expected from optical balance considerations [14].

II. PROPOSED SCHEME

To reveal the underlying physics as simply as possible, we consider the simplified case of a conventional laser cavity shown in Fig. 1. It consists of two mirrors, M_b and M_f, and contains as active medium a ferroelectric crystal doped uniformly with two fluorescent species A and B (e.g., rare-earth ions) with strong emission bands: a broad one at Ω_A and an overlapping narrower one at Ω_B (with $\Omega_B > \Omega_A$). Alternatively, one may consider poled polymers [15] hosting two fluorescent molecular complexes or dyes that act as species A and B. The two species are represented in Fig. 1 by two-level transitions, suitably broadened by vibronic coupling with the host lattice. It is important to stress that the two species are otherwise uncoupled and only communicate radiatively through the stimulated two-photon process indicated in Fig. 1.

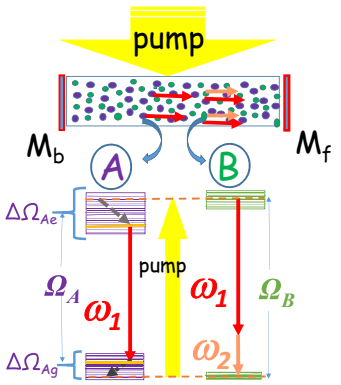

FIG. 1. (Color online) Schematic of the proposed scheme for stimulated parametric two-photon emission. The Fabry-Perot cavity (top) contains a ferroelectric medium doped with species A (purple) and B (green) that emit light when pumped optically (yellow arrow). The bottom part shows the energy bands and frequencies at which light is emitted.

Optical pumping of both species by the same light source, and subsequent fast intrasystem relaxation within the state manifolds, can invert populations of different sublevels within the transition widths. The cavity is configured to provide tunable stimulated emission from the sublevels of the broad Ω_A transition but blocks stimulated emission from those of the Ω_B transition. The photon population at one of the cavity modes, at frequency ω_1 within the bandwidth of Ω_A, will then induce cascaded two-photon emission at frequencies ω_1 and ω_2 from the species B such that $\omega_1 + \omega_2 = \Omega_B$.

The two-photon transition amplitude for such a poled, effective two-level system is dominated by two pathways involving a succession of intra- and interstate transitions shown schematically in Fig. 2. Since the cross section of the underlying two-photon process increases with increasing numbers of photons at ω_2, an avalanche process sets in. It drives the two-photon stimulated process through a *ratchet mechanism* set up by the simultaneous breakdown of spatial inversion symmetry and thermal equilibrium in the optically inverted off-equilibrium populations. One can infer from the level scheme in Fig. 1 that in general $\omega_1 > \omega_2$, and the tuning range of two-photon emission is practically fixed by that of

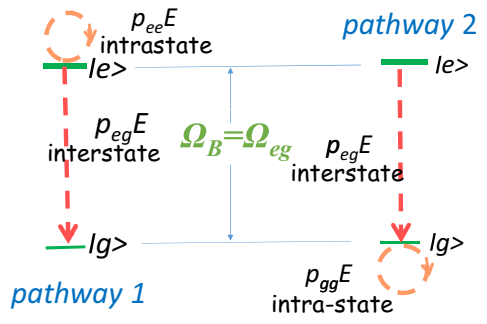

FIG. 2. (Color online) Schematic of the effective two-level system associated with species B. The parametric two-photon process is dominated by the two pathways involving a succession of an intrastate (orange circle) and an interstate (red arrow) transition.

the priming laser emitting at ω_1; in addition, over most of this range the complementary frequency ω_2 does not fall within the bandwidths $\Delta\Omega_{Ag}$ and $\Delta\Omega_{Ae}$ associated with the species A. We should stress that, although our scheme makes use of the priming photon scheme of Ref. [1], it differs from it as it is driven by a ratchet effect.

III. NONLINEAR COUPLED AMPLITUDE EQUATIONS

To quantify the buildup of the stimulated two-photon process, we consider the two-photon transition rate between the excited and ground states of species B [16,17]:

$$\frac{dP_{e\to g}}{dt} = 8\pi^3 d_{eg}\frac{\omega_1\omega_2}{n_1^2 n_2^2}\left|K_{eg}^{(2)}\right|^2 g(\Delta\omega)(\bar{N}_1+1)(\bar{N}_2+1), \quad (1)$$

where $d_{eg} = (N_e - N_g)/V$ is the population inversion density, N_g and N_e being the number of species B atoms in the ground and excited states, respectively. Further, $\bar{N}_j$ and n_j are the average photon number and refractive index at the frequency $\omega_j (j = 1,2)$, $\Delta\omega = \Omega_{eg} - \omega_1 - \omega_2$ is the detuning from resonance, and $g(\Delta\omega) = T_2/[\pi(1 + (\Delta\omega T_2)^2]$ is the density of states for the $e \Rightarrow g$ transition with the coherence relaxation time T_2. The two-photon transition amplitude $K_{eg}^{(2)}$ is given by

$$K_{eg}^{(2)} = \sum_c \left[\frac{\langle e|\hat{e}_1\cdot\boldsymbol{p}|c\rangle\langle c|\hat{e}_2\cdot\boldsymbol{p}|g\rangle}{\hbar(\omega_{cg}-\omega_2)} + \frac{\langle e|\hat{e}_2\cdot\boldsymbol{p}|c\rangle\langle c|\hat{e}_1\cdot\boldsymbol{p}|g\rangle}{\hbar(\omega_{cg}-\omega_1)}\right], \quad (2)$$

where $\hat{e}_1$ and $\hat{e}_2$ are the unit vectors along which the two electric fields are polarized and the sum extends over all states.

For an effective two-level scheme for the species B (no intermediate states), expression (2) reduces to

$$K_{eg}^{(2)} = 2\hbar\kappa = \frac{\boldsymbol{p}_{eg}\cdot\hat{e}_1}{\hbar\omega_2}(\boldsymbol{p}_{ee}-\boldsymbol{p}_{gg})\cdot\hat{e}_2 + \frac{\boldsymbol{p}_{eg}\cdot\hat{e}_2}{\hbar\omega_1}(\boldsymbol{p}_{ee}-\boldsymbol{p}_{gg})\cdot\hat{e}_1, \quad (3)$$

where $p_{eg} = \langle e|\boldsymbol{p}|g\rangle$ is the interstate (transition) dipole moment and p_{ee} and p_{gg} are the intrastate ones for the excited and ground states, respectively; we assume the same values for all sublevels within the upper and lower band states of species B. Equation (3) shows clearly that $p_{ee} - p_{gg}$ must be nonzero, which is possible only for polar materials whose energy states do not have a definite parity. On plausible physical grounds one also expects p_{ee} and p_{gg} to point in the opposite directions and $|p_{ee}| > |p_{gg}| > |p_{eg}|$, as can be inferred from charge extension considerations.

From (1) we can write the evolution of photon number in mode j ($j = 1,2$) in the form

$$\frac{d\bar{N}_j}{dz} = \frac{n_j}{c}\frac{\omega_1\omega_2}{n_1 n_2}c^2\gamma(\bar{N}_1+1)(\bar{N}_2+1)_2 - \alpha(\omega_j)\bar{N}_j, \quad (4)$$

where the two-photon gain γ is given by [16]

$$\gamma = \frac{8\pi^3}{n_1 n_2 c^2} d_{eg} \left| K_{eg}^{(2)} \right|^2 g(\Delta\omega), \tag{5}$$

and losses are included phenomenologically using the absorption coefficient $\alpha(\omega)$. This equation can be used to study the initial buildup of photon population at the frequency ω_2 from spontaneous parametric two-photon emission. As the stimulated two-photon regime is established and the photon densities in both ω_1 and ω_2 modes become large, two-photon resonant nonlinearities higher than the cubic ones considered above become relevant. In this high-intensity regime as the saturation regime sets in, quantum fluctuations in the photon states can be neglected, and one can introduce amplitudes A_1 and A_2 for the two modes and use semiclassical nonlinear amplitude equations generated by the relevant nonlinear polarization sources. The latter can be obtained with the optical Bloch equations for the coherent two-photon resonance using the adiabatic following approximation [18,19]. The field intensities I_j are related to $\bar{N}_j$ as ($j = 1,2$)

$$I_j = 2\varepsilon_0 c n_j |A_j|^2 = \frac{c}{n_j} \bar{N}_j \hbar\omega_j. \tag{6}$$

The nonlinear polarization source resulting from the two-photon resonance has the form $\tilde{P}_j^{\mathrm{NL}} = \varepsilon_0 \tilde{\chi}_{jR}^{\mathrm{NL}} A_j$ at the frequency ω_j.

Neglecting diffraction and absorption losses and applying the slowly varying envelope approximation procedure, the nonlinear amplitude equations become

$$\frac{dA_j}{dz} = \frac{i\omega_j}{2n_j c} \tilde{\chi}_j^{\mathrm{NL}} A_j, \tag{7}$$

where $\tilde{\chi}_j^{\mathrm{NL}}$ are the effective two-photon resonant nonlinear susceptibilities with $j = 1$ and 2. Their expressions can be derived from the Bloch equations in the form [20]

$$\tilde{\chi}_j^{\mathrm{NL}} = \tilde{\chi}_0 \frac{(\Delta + i)\tilde{I}_{3-j} + \delta_j(1+\Delta^2)(T_1/T_2)}{(1 + \Delta^2 + \tilde{I}_1\tilde{I}_2)}, \tag{8}$$

with $\tilde{\chi}_0 = -(N\hbar\kappa/2\varepsilon_0)(T_2/T_1)^{1/2}$. Here $\Delta = \Delta\omega T_2 + (\delta_1\tilde{I}_1 + \delta_2\tilde{I}_2)/2$ represents detuning from the two-photon resonance, $\delta_j = (T_2/T_1)^{1/2}[\alpha_g(\omega_j) - \alpha_{e'}(\omega_j)]/2\hbar\kappa$ is related to the dynamic Stark shift ($j = 1$ or 2), and $\tilde{I}_j = |E_j|^2/I_s$ is the intensity normalized to the two-photon saturation intensity defined as $I_s = (\kappa^2 T_2 T_1)^{-1/2}$. Finally, $\alpha_g(\omega_j)$ and $\alpha_e(\omega_j)$ are the ground and excited state polarizabilities for species B at the frequency ω_j. For an effective two-level system, neglecting the intrastate contribution and keeping only the interstate one, to a good approximation we can set $\alpha_g(\omega_j) \cong -\alpha_e(\omega_j) \cong \alpha_0 = (2/\hbar)|p_{eg}|^2\Omega_{eg}/(\Omega_{eg}^2 - \omega_j^2)$. In Eq. (6), T_1 and T_2 are the population and coherence relaxation times, respectively. Their introduction allows the system to achieve a nonequilibrium steady state; they also set the saturation intensity of the underlying two-photon transition. Introducing Eq. (8) in Eq. (7), we obtain a set of coupled equations

$$\frac{dA_j}{dz} = \frac{i\omega_j\chi_0}{2n_0 c}\left[\frac{(\Delta + i)\tilde{I}_{3-j} + \delta_j(1+\Delta^2)(T_1/T_2)}{(1 + \Delta^2 + \tilde{I}_1\tilde{I}_2)}\right] A_j, \tag{9}$$

where we neglected the refractive index dispersion and set $n_1 = n_2 = n_0$. Note that χ_o is negative in the case of inverted population of species B.

IV. ANALYTIC AND NUMERICAL RESULTS

As a first approximation, we neglect the Stark shifts and set $\delta_j = 0$. Writing Eq. (9) in terms of intensities using the relation (6) we obtain

$$\frac{1}{\omega_1}\frac{d\tilde{I}_1}{dz} = \frac{1}{\omega_2}\frac{d\tilde{I}_2}{dz} = \frac{K\tilde{I}_1\tilde{I}_2}{1 + \Delta^2 + \tilde{I}_1\tilde{I}_2}, \tag{10}$$

where $K = -\chi_0/(n_0 c)$. To simplify the notation, we drop the tildes over I_1 and I_2 in what follows. It is easy to conclude that $I_1(z)/\omega_1 - I_2(z)/\omega_2 =$ constant, which expresses the fact that the two photons are produced simultaneously. The constant can be found using the initial conditions at $z = 0$ and leads to the relation

$$I_1(z) = I_{10} + r[I_2(z) - I_{20}], \tag{11}$$

where a zero subscript denotes the initial value at $z = 0$ and $r = \omega_1/\omega_2$ is the frequency ratio of the two photons involved in the parametric two-photon process. Using the relation in Eq. (11), it is easy to integrate the coupled set of two equations in Eq. (10). More specifically, the intensity I_2 is found to change with z along the medium length as

$$I_2(z) = I_{20}\left(1 + \frac{r}{I_{10}}(I_2 - I_{20})\right) \times \exp\left[\frac{I_{10}(1 + rI_{20})}{1 + \Delta^2}(K\omega_2 z - I_2 + I_{20})\right]. \tag{12}$$

It shows an exponential growth as long as I_2 remains much less than both I_{10} and $K\omega_2 z$. The growth rate decreases when these conditions are not satisfied but never becomes negative. As an example, Fig. 3 shows how $I_2(z)$ changes with distance inside a sample of length L for three values of I_{10} in the range of 1–10 using $K\omega_2 L = 2$, $I_{20} = 1 \times 10^{-6}$, $\Delta = 0$, and $\omega_1/\omega_2 = 5$. In all cases, I_2 grows in an exponential fashion initially but begins to saturate after $z/L > 0.6$ when I_{10} is so large that I_2 approaches a value close to I_s.

As can be inferred from Eq. (9), the efficiency of the stimulated parametric two-photon emission may be affected by the Stark shift in the high-intensity regime. This is a novel feature in any coherent active two-photon process and it can be used as a control parameter. This issue was previously pointed out in the case of the passive two-photon optical bistability [20] and it was found that it could have a noticeable impact both on the contrast as well as on the threshold of optical bistability.

In our case, the situation becomes much more complex when the Stark shifts are included. We can still use Eq. (10) to obtain the following coupled intensity equations:

$$\frac{dI_j}{dz} = K\omega_j \frac{(I_{3-j} - s_j)I_j}{1 + \Delta^2 + I_1 I_2}, \tag{13}$$

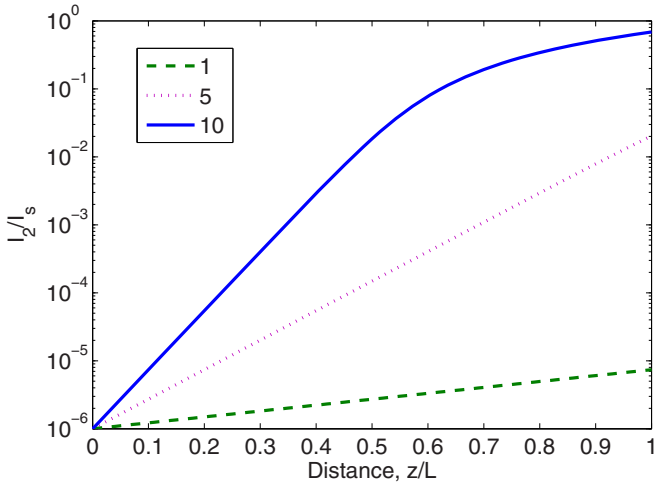

FIG. 3. (Color online) Growth of normalized intensity $I_2(z)$ associated with the parametric two-photon process plotted as a function of normalized distance z/L for three values of I_{10} using $K\omega_2 L = 2$, $I_{20} = 1 \times 10^{-6}$, $\Delta = 0$, and $\omega_1/\omega_2 = 5$. Notice the saturation occurring after $z/L > 0.6$ when $I_{10} = 10$. All intensities are normalized to the saturation intensity I_s.

where $s_j = \delta_j(1 + \Delta^2)(T_1/T_2)$ is related to the Stark shift ($j = 1$or2). Using Eq. (9) we may set $\delta_1 = \delta_2 = \delta_0$, if we neglect the frequency dependence of the polarizability, and the detuning parameter becomes $\Delta = \Delta_0 + \delta_0(I_1 + I_2)/2$ with $\Delta_0 = \Delta\omega T_2$. The solution of the coupled intensity equations still requires a numerical approach. As an example, Fig. 4 shows how $I_2(z)$ changes with distance inside a sample of length L for three values of δ_0 in the range of 0–0.2 using $K\omega_2 L = 2$, $I_{10} = 10$, $I_{20} = 1 \times 10^{-6}$, $\Delta_0 = 0$, $T_1/T_2 = 2$, and $\omega_1/\omega_2 = 5$. It is clear from this figure that any finite value of the Stark shift reduces the growth of I_2 compared to the case $\delta_0 = 0$. Although relatively small values of δ_0 (below 0.01) do not affect the intensity growth significantly, I_2 at $z = L$ is reduced by more than a factor of 100 for $\delta_0 = 0.1$.

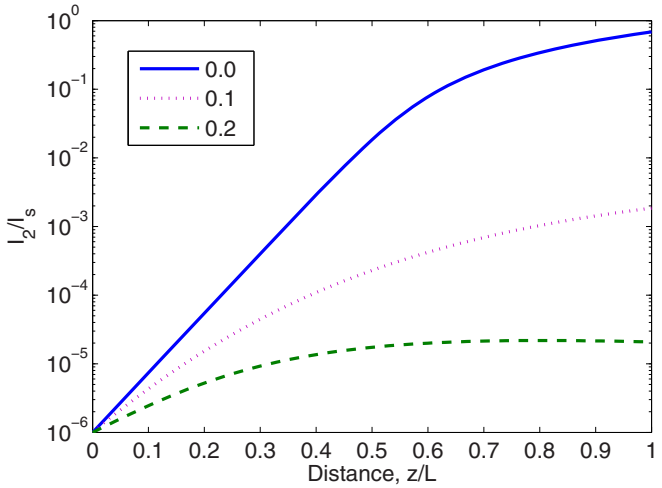

FIG. 4. (Color online) Same as Fig. 3 except that three curves correspond to different values of the Stark shift parameter δ_0 in the range of 0–0.2 at fixed values of $I_{10} = 10$ using $\Delta_0 = 0$, $T_1/T_2 = 2$. All other parameters and normalizations remain the same.

V. DISCUSSION AND CONCLUSIONS

To evaluate the practical significance of the scheme in Fig. 1, we need realistic values of material parameters such as K and δ_0. Notwithstanding the growing interest and activity in the spectroscopy of rare-earth ions in ferroelectrics [7–13], systematic data are still not available that will allow one to deduce the relevant parameter values for the rare-earth impurities in ferroelectrics. This hinders even an order-of-magnitude estimate of the parameters involved in the above scheme. Still, we can infer from the simplified models that the permanent dipole moments of the ground and excited states of species B, being determined by the unit-cell dimension, are almost an order of magnitude larger than the transition dipole moments between the ground and excited states, the latter being at best determined by the impurity extension which is smaller than that of the unit cell. This clearly leads to a sizable enhancement of the two-photon transition probability in Eq. (3) and the related gain in Eq. (4). On the other hand, one can expect complications from the crystal field effects and concomitant inhomogeneous broadening of the transitions, as well as problems related to thermal fluctuations in the ferroelectric microdomains.

In spite of these difficulties, our analysis here shows that the scheme first proposed in Ref. [1] has the potential of exhibiting stimulated parametric two-photon emission efficiently when two suitable rare-earth materials are used for doping a ferroelectric crystal. The effective gain coefficient of this parametric process for the species B can be estimated from Eq. (12) and is given by $g = K\omega_2 I_{10}/I_s$, where I_{10} is the intensity of the laser beam produced by pumping of species A. It shows that the parametric gain is enhanced by the flux of the priming photons contained within the laser beam. Since I_{10} can be made quite large through a suitable pumping scheme, the parametric gain is enhanced by a large factor, and the two-photon parametric process associated with species B becomes much more efficient.

The proposed scheme builds on the idea of priming photons in Ref. [1] but differs from it by using a poled, bidoped, ferroelectric crystal (or a poled dielectric or polymer) that breaks down both the space inversion symmetry and the parity of the energy states. This sets the stage of (i) two-photon transitions even in a two-level scheme through a single pathway involving succession of an intrastate and an interstate transition and (ii) a ratchet mechanism, set up by the simultaneous breakdown of both the space and time inversion symmetries, that drives and sustains a parametric two-photon emission process whose gain is enhanced by a large factor related to the number of priming photons. The approach outlined here can be used to describe other nonlinear processes in poled media with distributed gain, without requiring a cavity or phase matching. In particular, it may be applicable to the rectification of quantum optical noise.

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