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PARAMETRIC STIMULATED TWO-PHOTON EMISSION THROUGH BI-PHOTONIC CASCADE

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Abstract. A parametric two-photon process, involving stimulated emission of two cascaded photons from a bi-doped poled material placed inside an optically pumped cavity, is proposed and assessed within the priming photon scheme. 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 demonstration of a two-photon laser remains both elusive and challenging even after close to fifty years of sustained effort [1–3]. 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] provides renewed impetus and interest for the two-photon laser studies.

Stumbling issues in achieving the critical photon density for TP laser operation are the much smaller efficiency of the two-photon emission compared with that of the competing one-photon emission and the concomitant complexity of the two-photon transition probability, proceeding through multiple pathways that can interfere and reduce or even suppress the process. Besides, it is being tacitly assumed in the majority of cases that the active sites possess the spatial inversion symmetry and their states accordingly have definite parities. Alternative schemes as the use of priming photons [1] to initiate and enhance the TP-emission on the expense of the one-photon emission didn't either reach the expectations.

Here we outline an approach [6] that builds on the proposal of priming photons in a poled, bi-doped, ferroelectric crystal [7, 8] (or a poled dielectric or polymer [9]) 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 transitions. When such a crystal is pumped optically to create population inversion, a *ratchet* *mechanism* [10] set up by the simultaneous breakdown of both the space and time inversion symmetries further drives and sustains the two-photon emission process.

The basic scheme and underlying physics are depicted with the simplified case 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 [7, 8] (*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 [9] 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. Optical pumping of both species by the same light source, and subsequent fast intra-system 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 $\omega_1 + \omega_2 = \Omega_B$.

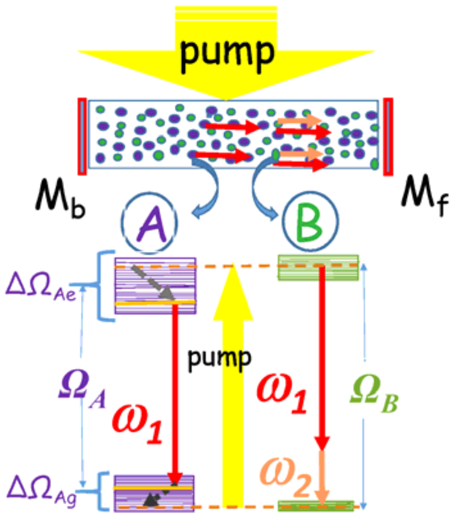

Fig. 1 – Schematic of the proposed scheme for parametric stimulated two-photon emission. The Fabry-Perot cavity contains a ferroelectric medium doped with two species A (purple) and B (green) that emit light when pumped optically (yellow arrows). The bottom part shows the energy bands and frequencies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

We describe [6] the evolution of the average photon number N_j in the mode j (j = 1, 2) in the form [11]

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

where 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. In (1) the two-photon gain

$$\gamma = \frac{8\pi^3}{n_1 n_2 c^2} d_{eg} \mid K_{eg}^{(2)} \mid^2 \; g(\Delta\omega) \tag{2}$$

is expressed in terms of the population difference $d_{eg} = (N_e - N_g)/V$ between states e and g, the joint density of states $g(\Delta\omega)$ and the two-photon transition amplitude $K_{eg}^{(2)}$ for the species B. For such a poled, effective two-level system the later is dominated by two pathways involving a succession of intra- and inter-state transitions shown schematically in Fig. 2. For an effective two-level scheme for the species B (no intermediate states), reduces to

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

where $p_{eg} = < e \mid \underline{p} \mid g >$ 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. One can show on general grounds that p_{ee} and p_{gg} point in opposite directions and in general $\mid p_{ee} \mid > |p_{gg}| > \mid p_{eg}$ as can be inferred from charge extension considerations and all terms in (3) are additive and contribute to the enhancement of $K_{eg}^{(2)}$.

Since the cross section of the underlying two-photon process increases with increasing number of photons at $\omega_{2,}$ 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 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 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 introduced by the simultaneous breakdown of both the space and time inversion symmetries.

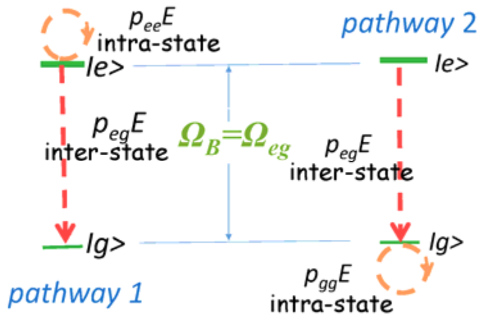

Fig. 2 – Schematic of the effective two-level system associated with species B. The parametric two-photon process is dominated by the two pathways involving a succession of an intra-state (orange circle) and an inter-state (red arrow) transition.

As the stimulated two-photon regime is established and the photon densities N_j (j = 1, 2) in both ω_1 and ω_2 modes become large, 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

$$\frac{\mathrm{d}A_j}{\mathrm{d}z} = \frac{i\omega_j}{2n_j c}\tilde{\chi}_j^{NL} A_j, \tag{4}$$

in the SVE-approximation [11] with the relevant nonlinear polarization sources resulting from the two-photon resonance $\tilde{P}_j^{NL} = \varepsilon_o \tilde{\chi}_{jR}^{NL} A_j$, where $\tilde{\chi}_j^{NL}$ are the effective two-photon resonant nonlinear susceptibilities, j = 1, 2. The later can be obtained [12] with the optical Bloch equations for the coherent two-photon resonance using the adiabatic following approximation [13]. An important issue here is the inclusion [12] of the Stark shifts which are absent in equs (1) which are effectively restricted to cubic nonlinear polarization sources.

Neglecting the Stark shifts and recasting (4) in terms of light intensities we obtain

$$\frac{1}{\omega_1}\frac{\mathrm{d}\tilde{I}_1}{\mathrm{d}z} = \frac{1}{\omega_2}\frac{\mathrm{d}\tilde{I}_2}{\mathrm{d}z} = K\frac{\tilde{I}_1\tilde{I}_2}{1+\Delta^2+\tilde{I}_1\tilde{I}_2}, \tag{5}$$

with

$$I_j = 2\varepsilon_0 c n_j |A_j|^2 = \frac{c}{n_j}\bar{N}_j\,\hbar\omega_j, \quad K = (\hbar\kappa\Delta N_{eg}/2\varepsilon_o)(T_2/T_1)^{1/2}, \; I_s = 1/(\kappa^2 T_2 T_1)^{1/2}.$$

I_s is the two photon saturation intensity, T_1 and T_2 are the phenomenological population and coherence relaxation times and a tilde denotes that all intensities are normalized to I_s. Note that K reverses sign with inverted population ΔN_{eg} of species B.

It is easy to conclude from (5) that $I_1(z) = I_{10} + r[I_2(z) - I_{20}]$, expressing the fact that the two photons are produced simultaneously; a zero subscript denotes the initial value at $z = 0$ and $r = \omega_1/\omega_2$. Since the initial population of ω_2 photons has its origin in quantum noise, we assume $I_{20} << 1$ and neglect it in comparison of I_1, I_2 , I_{10} then at distances $z > 0$ we obtain

$$I_2(z) = I_{20}\left(1 + \frac{r(I_2 - I_{20})}{I_{10}}\right)\exp\left[\frac{I_{10}(1 + rI_{20})}{1 + \Delta^2}(K\omega_2 z - I_2 + I_{20})\right]. \qquad (6)$$

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. 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 to 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.

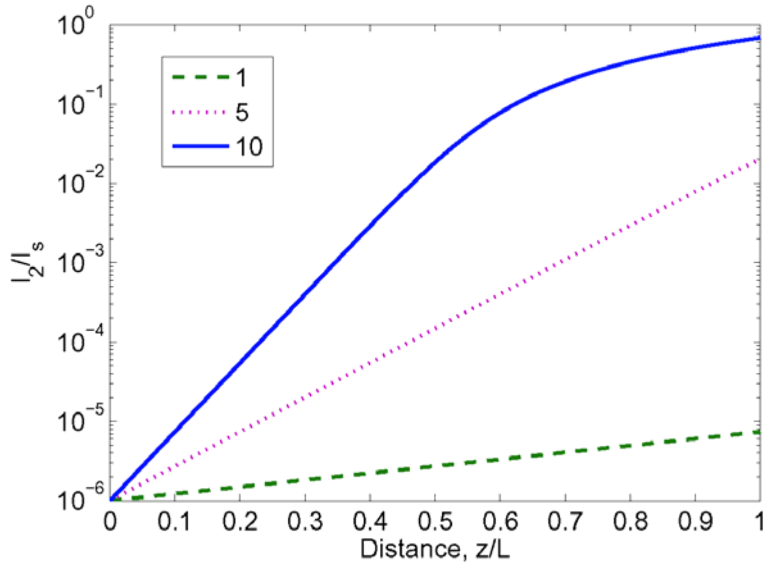

Fig. 3 – Growth of intensity I_2(z) associated with the parametric two-photon process plotted as a function of z/L for three values of the intensity I_{10} (associated with the primed photons) using $K\omega_2 L = 2$, $I_{20} = 1 \times 10^{-6}$, $\Delta = 0$, and $\omega_1/\omega_2 = 5$. Notice the saturation occurring after $z/L > 0.6$ when $I_{10} = 10$.

The situation becomes much more complex when the Stark shifts are included and the relevant equations can only be solved numerically [6]. The key result there is the impact of the Stark shifts on the efficiency of the processes and their use as control parameter.

To evaluate the practical significance of the scheme shown in Fig. 1, we need realistic values of material parameters such as K and δ_0 which are still scarce although over the recent years multi-wavelength lasers and coherent sources have been developed [7–13] by using bi-doped ferroelectrics containing two different rare-earth dopants, each providing its own characteristic laser emission upon pumping, together with second harmonic, sum or difference frequency generation if appropriate phase matching (PM) can be achieved. Notwithstanding the interest of such schemes for multi-wavelength 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 subject to a stringent phase-matching condition, as expected from optical balance considerations [14].

There are also complications from the crystal field effects and concomitant inhomogeneous broadening of the transitions, as well as problems related to thermal fluctuations in the ferroelectric micro-domains dimer formation of the RE-impurities to form dimers and related energy transfers which are recurrent also in the other systems.

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. (2) 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. Alternatively the whole scheme can also be recast in the form of cascade of two stimulated processes as can be inferred from the coupled equations (4) extending the class [15] of such nonlinear optical processes.

In conclusion, we have discussed a novel approach for enhancing two-photon emission that may prove useful for demonstrating a true two-photon laser. It builds on the idea of priming photons of Ref. [1] but differs from it by using a poled, bi-doped, ferroelectric crystal (or a poled dielectric or polymer) that breaks down both the space inversion symmetry and the parity of the energy states.

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