

Second Order Nonlinear Processes in Photonic Crystals

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ABSTRACT

We show that the second order parametric nonlinear interaction from waves that are counter propagating may be perfectly phase matched in the framework of a photonic crystal. We observe that the large momentum mismatch of such parametric process is compensated because of the interfaces present in the nonlinear photonic crystal considered, which separate a linear from a nonlinear material. The numerical results presented indicate that such nonlinear photonic crystals could be a very good material to consider the observation of backward parametric oscillation without mirror feedback. In the search for a material capable of holding such type of oscillation, we have performed second harmonic generation experiments using several nonlinear colloidal crystals. We compared the efficiency between such different nonlinear crystals obtained by binding different types of organic nonlinear molecules onto the surface of a polystyrene particle.

Keywords: nonlinear interaction, centrosymmetric materials, photonic crystal, second harmonic generation, counter-propagating.

1. INTRODUCTION

The second order nonlinear interaction was incorporated in the field of photonic crystals in 1995 when second harmonic generation in 3-dimensional (3-D) photonic crystals (PC) was reported for the first time [1]. In that early work and subsequent publications it was shown that the periodic distribution of dielectric material in photonic crystals was very useful to phase match or enhance such nonlinear interaction [1-9].

In the first part of the present paper, we will show, theoretically, that another very unique feature of photonic crystal structures is the capability to hold a non-degenerate parametric interaction of waves that are counter-propagating. In the bulk of the conventional nonlinear materials used for parametric generation, such counter-propagating interaction is extremely out of phase matching and, consequently, transfer of energy between the three fields involved vanishes. In fact, the fundamental nonlinear process of backward parametric oscillation without mirror feedback [10-11], which requires a phase matching mechanism capable of compensating such large mismatch [12], has never, to the best of our knowledge, been observed. In a nonlinear photonic crystal, the interfaces between the two dielectric materials, separate also a linear from a nonlinear material. At such interfaces the counter-propagating interaction may be as significant as the co-propagating one [13]. We will show that, in certain conditions, such counter-propagating interaction is perfectly phase matched and becomes the major contribution to the transfer of energy among the three fields at the three different frequencies. In the second part of the paper, we present a study on the second order nonlinear interaction from several nonlinear photonic crystals. We used three different nonlinear molecules and two types of binding to the surface, by physical adsorption or with a covalent bond to the carboxylate groups on the surface. The efficiency of second harmonic generation is measured in all cases.

2. COUNTER-PROPAGATING INTERACTION

In our theoretical model we consider a 3-D photonic crystal made of dielectric spheres of diameter D surrounded with a homogeneous material of a lower dielectric constant. The nonlinear material is assumed to be a layer of thickness δ , located only at the interfaces separating these two dielectric materials. As shown in Fig. 1, the geometry is such that the three fields are contained on the xz plane, which is perpendicular to one set of the several sets of planes present in a 3-D PC. It is assumed that there are two strong beams, one incident from the left and another incident from the right, at frequencies ω_3 and ω_2 , respectively. A third beam is generated either to the left or to the right (cf. Fig. 1) at $\omega_1 = \omega_3 + \omega_2$ when summation is considered, or at $\omega_1 = \omega_3 - \omega_2$ when we have difference generation. Because of multiple reflections within the photonic crystal, at each given plane of the set (except for the first and last) there is also a field at ω_3 incident from the right and a field at ω_2 incident from the left (cf. Fig. 1). Unless we consider a long photonic crystal one may neglect the depletion of the two pump beams.

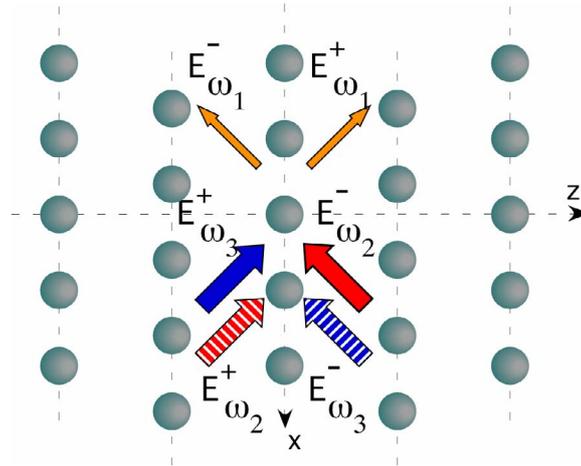


Figure 1. Schematic representation of the forward and backward propagating fields involved in the nonlinear interaction we consider. Thick solid arrows represent the incident fields, while the dashed arrows represent the fields that appear because of multiple reflections in the periodic structure. Thin arrows represent the fields generated via the nonlinear interaction.

Then, one may write one single nonlinear wave equation for the two counter propagating beams at ω_1 . Under the assumption of harmonic solutions, and if one considers difference generation, the nonlinear polarization source term of such equation may be separated into four different contributions

$$P_{\omega_1}^{nl\pm}(r) = \varepsilon_0 \chi^{(2)}(r) : E_{\omega_3}^+(r) E_{\omega_2}^{-*}(r) \quad (1a)$$

$$P_{\omega_1}^{nl\pm}(r) = \varepsilon_0 \chi^{(2)}(r) : E_{\omega_3}^-(r) E_{\omega_2}^{+*}(r) \quad (1b)$$

$$P_{\omega_1}^{nl-}(r) = \varepsilon_0 \chi^{(2)}(r) : E_{\omega_3}^+(r) E_{\omega_2}^{+*}(r) \quad (1c)$$

$$P_{\omega_1}^{nl+}(r) = \varepsilon_0 \chi^{(2)}(r) : E_{\omega_3}^-(r) E_{\omega_2}^{-*}(r) \quad (1d)$$

Where $\chi^{(2)}(r)$ is the periodic second order nonlinear susceptibility, the $E_{\omega_i}(r)$ are the electric field components for a wave at frequency ω_i , and the “+” and “-” signs on the field amplitudes indicate z-positive and z-negative propagation directions, respectively. The “+” sign on the polarization indicates contribution to the field at ω_1 propagating in the z-positive direction, while the “-” sign indicates contribution to the same field propagating in the z-negative direction. Similar expressions may be written when one considers sum generation. Note that in a conventional nonlinear material only the source term given in equation (1a) would exist and its contribution to conversion at ω_1 would be negligible.

We may solve the nonlinear wave equation for the transverse part of the electric field extending the method developed in reference [9], including the four nonlinear source terms from equations (1a-1d). The solution is used together with the transfer matrix formalism, applied to all three frequencies, to determine the total generated field at ω_1 in the forward and backward directions. In a numerical application we consider photonic crystals made from polystyrene suspensions of microspheres surrounded with water, which exhibit a dielectric contrast

$\varepsilon_r = 1.43$. We consider a sphere diameter of 175 nm and the wavelengths of the incident fields to be $\lambda_3 = 355$ nm and $\lambda_2 = 810$ nm. As is well known, the periodicity build in a photonic crystal can be used for the phase matching of co-propagating second order nonlinear interactions in certain propagation directions. Such phase matching is achieved at the upper or lower edge of the Bragg reflection bands at each frequency involved in the interaction. This mechanism of phase matching is sufficient to compensate the dispersion of water ($\Delta n \approx 0.02$), but by no means is able to compensate a phase mismatch of the order of $\Delta k = (k_{3z} - (-k_{2z})) - k_{1z}$ equivalent to an effective index dispersion of $\Delta n \approx 2.1 \cdot \cos\theta$. The mechanism needed to compensate such large effective dispersion resulting from the counter-propagating interaction comes from an interaction at the interface. When the two mechanisms are combined one may obtain a peak of maximum conversion. In Fig. 2, where the difference generation at 632 nm is shown as a function of the angle of incidence, three of such peaks appear. In the numerical calculation the total number of planes considered is 110, while $\chi_{r'r'r'}^{(2)} E_{\omega_{2,3}} \approx 5 \cdot 10^{-3}$. The first peak at $\theta_1 \approx 19^\circ$ is located by the low frequency edge of the 810 nm band. The second peak at $\theta_2 \approx 33^\circ$ is located by the high frequency edge of the second order band at 355 nm. Finally, the last peak is located at the low frequency

edge of the band at 632 nm at $\theta_3 \approx 43.25^\circ$. Now, if one considers contribution from all source terms but the term given in equation (1a), difference generation is remarkably reduced for the two largest peaks of difference generation as seen in Fig. 2. This last result indicates that the largest contribution to difference generation comes directly from a counter-propagating interaction. Note that contribution from the source terms in equations (1b-d), which are present because of multiple reflections inside the structure of both incoming beams, is not significant for these two largest peaks. In other words, at θ_1 and θ_2 the nonlinear interaction is a truly counter-propagating interaction. The difference generation in either direction cannot be attributed to a cavity effect or to the co-propagating interaction that one gets from the incident field at ω_3 and the reflected field at ω_2 . This is not the case for the peak at θ_3 where phase matching is achieved via a noncollinear mechanism, which was already considered in the past.[14]

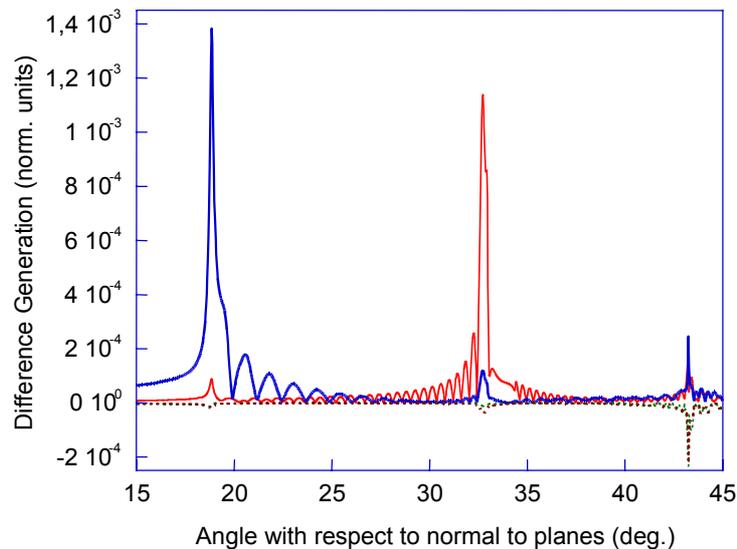


Figure 2. Difference frequency generation at ω_1 as a function of the angle with respect to the z -axis, for a 3-D PC similar to the one schematically shown in Fig. 1. The electric field amplitude is shown when all source contributions are considered (solid lines) in the forward direction (thick solid line), and in the backward direction (thin solid line). This electric field amplitude (for clarity of the figure this amplitude is multiplied by a minus sign) is also shown when the contribution from the source term given in equation (1a) is eliminated (dashed lines), in the forward direction (thick dotted line) and in the backward direction (thin dotted line).

3. EFFICIENCY MESUREMENTS ON SHG IN NONLINEAR COLLOIDAL CRYSTALS

Several 3-D photonic crystals were fabricated from commercial sulfate and carboxyl latex of monodisperse polystyrene nanospheres (refractive index $n = 1.59$) of 147 nm in diameter, suspended in water. The photonic crystals were grown in $200 \mu\text{m}$ thick precision cells that contained, in the bottom, a mixed bed ion exchange resin. To tune the position of the Bragg reflection band, of the (111) planes, the concentration of the colloidal suspension was accurately controlled before being introduced into the cell. These microspheres self organize in an fcc lattice as stray ions in the water solution diffuse towards the ion exchange resin. Different photonic crystals were fabricated; in some samples conveniently modified Methyl Orange was used to form a covalent bond with the carboxyl surface groups, while in other crystals, we choose Crystal Violet to be physically adsorbed onto the surface. Methyl Orange, commonly used organic dye in nonlinear optics, could be easily modified to be covalently coupled to the polystyrene spheres surfaces. Since Methyl Orange can not be adsorbed to the sulfate surface groups, we choose Crystal Violet, which is known to have a high second order nonlinear molecular polarizability, as the nonlinear molecule to be physically adsorbed onto the surface of the spheres.

The 3-D photonic crystals obtained were pumped with a 6 ns laser pulse at 1064 nm from a Nd:YAG laser and placed in a rotating mount with the rotating axis perpendicular to the laser beam. The laser pulses with an average energy per pulse of 10 mJ , were focused down to a spot of 1.0 mm in diameter. Using a photomultiplier tube we performed measurements of the generated second harmonic light in transmission as a function of the angle of the normal to the crystal face with respect to the incident beam for different samples. In each case, we clearly observe a narrow peak of SHG at the upper edge of the first order Bragg reflection band at 532 nm , as it can be seen in Fig. 3, where a SHG peak corresponding to the sample with adsorbed Crystal Violet is shown.

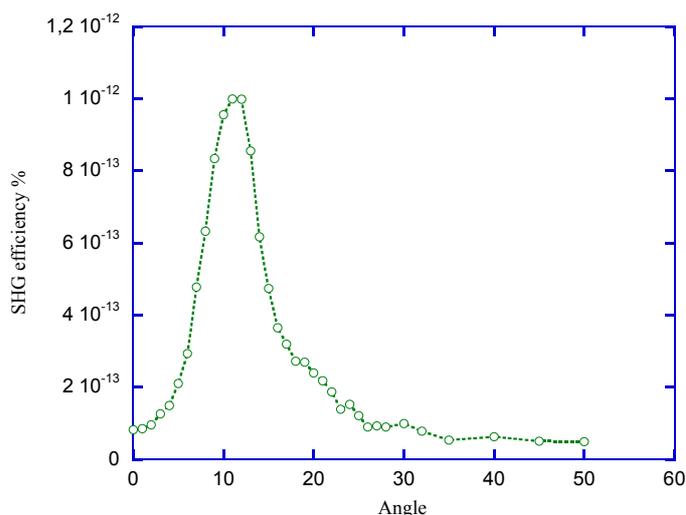


Figure 3. SHG peaks when physically adsorbed Crystal Violet is used as nonlinear molecule.

The energy conversion efficiencies from the fundamental light to second harmonic generated light in each sample are shown in Table 1. Previously reported results where Malachite Green was adsorbed onto the surfaces of the latex spheres [16] are also shown in Table 1 for comparison.

Table 1. SHG efficiency results.

Nonlinear Dye	Surface-molecule link	SHG efficiency
Malachite Green	physical adsorption [16]	$1.0 \cdot 10^{-13}$ %
Crystal Violet	physical adsorption	$1.0 \cdot 10^{-12}$ %
Methyl Orange	covalent bond	$2.0 \cdot 10^{-13}$ %

Note that the nonlinearity of Methyl Orange is 10 times less than the nonlinearity of Crystal Violet. Then, conversion efficiency should have been at least 100 times less than one obtained from adsorbed Crystal Violet. From such results, one may conclude that in the case of covalent bond, a larger number of molecules are bound to the surface.

4. CONCLUSIONS

We have demonstrated that a perfectly phase matched interaction of counter propagating waves is possible in nonlinear three-dimensional photonic crystals. We have shown that this effect is the result of a nonlinear interaction at the interface. The efficiency of such backward difference generation at other wavelengths is determined mostly by the particular 3-D geometry of the basis of the lattice. In this or any other geometry the efficiency of the process could be increased by simply considering layers of nonlinear material with a thickness comparable to the wavelength of the light. If one of these photonic materials with a sufficiently high nonlinearity is fabricated, one should be able to observe the reflectorless backward parametric oscillation predicted by S.E. Harris thirty-seven years ago [10].

We have experimentally observed that conveniently modified nonlinear molecules as Methyl Orange can be covalently bound to the multiple surfaces of a photonic crystal, and an efficiency comparable to the other two molecules is obtained. We show that using Crystal Violet as the nonlinear molecule the energy conversion efficiency increases by a factor of ten with respect to the Malachite Green efficiency. At present, work is underway to modify Crystal Violet in order to covalently bind a large number of such molecules onto the surface of the spheres. In such conditions we expect to see a significant increase in the conversion efficiency.

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