Simultaneously phase-matched enhanced second and third harmonic generation

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Second and third harmonic generation via a $\chi^{(2)}$ three-wave mixing process can occur with high conversion efficiency in a one-dimensional photonic band gap structure. We find that it is possible to simultaneously achieve enhancement and exact phase-matching conditions of second harmonic and sum frequency generation, $\omega+2\omega\rightarrow3\omega$. It is also remarkable that high conversion efficiencies persist under tuning conditions that correspond to a phase mismatch. While these conditions are quite unusual and cannot be achieved in any known bulk material, we show that they can be easily obtained in finite layered structures by using and balancing an interplay between material dispersion and the geometrical dispersion introduced by the structure.

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Simultaneous second and third harmonic generation via $\chi^{(2)}$ interactions in bulk media was first investigated theoretically by Akhmanov and Khokhlovnce [1]. After they realized the difficulty of achieving phase-matched, three-wave mixing (TWM) they concluded that: ''*successive three-frequency interactions are, for the present, not of interest for nonlinear optics*.'' They found that it was impossible to achieve simultaneous phase-matching conditions for second harmonic and sum-frequency generation in ordinary materials. During the last 10 years, interest in nonlinear frequency conversion has increased thanks to a technique referred to as quasiphase matching (QPM) [2–4]. Even though QPM was devised in the 1960s by Armstrong *et al.* [2], multicolor generation (red, green, blue) was first reported by Yamamoto, Yamamoto, and Taniuchi [5] in a proton-exchanged, MgO-doped $LiNbO_3$ waveguide. The process exploits the Cerenkov configuration [6], which provides QPM conditions over a wide range of wavelengths. These experiments demonstrated that QPM could be used to yield good conversion efficiencies for simultaneous multiwavelength generation, and reopened the challenge for the realization of compact, multicolor laser sources for laser scanners, printers, and color display devices [7–9].

It is also possible to achieve phase matching for three-wave mixing using a periodic modulation of the linear refractive index, as first proposed by Bloembergen and Sievers [2] in reflection geometry. This technique allows interesting applications of multicolor frequency generation, as recently shown by Konotop and Kuzmiak [10]. In their work, the authors show that it is possible to achieve a double phase matching condition for second and third harmonic generation using simultaneous second- and third-order nonlinearities in infinite structures, thus neglecting effects that arise from field localization. In this paper we show that a generic, *finite*, one-dimensional photonic band gap (PBG) structure can be used to achieve efficient, simultaneous, collinear, *global* (both forward and backward) second and third harmonic generation using only a second-order nonlinearity, and by judiciously taking advantage of the simultaneous availability of phase-matching conditions and field localization effects inside the finite structure. Therefore, we propose a simple way to achieve optimal conditions by tuning the fields to appropriate resonance peaks, *without* resorting to uniaxial crystals or periodic domain inversion, as required by QPM. We then test the model on a realistic AlN/SiO_2 stack, and find that large enhancements of conversion efficiencies, in excess of one order of magnitude, persist even under conditions of a slight phase mismatch.

Previously we showed that in a periodic structure phase-matching conditions for second harmonic generation (SHG) [11,12] and frequency downconversion could be fulfilled if the fields were tuned to the appropriate resonance peaks of the transmission spectrum: the effective dispersion introduced by the geometry acts to compensate material dispersion. We emphasize that the dispersive properties of finite structures are not the same as the dispersive properties of structures of infinite length. The expression for the real part of the effective index for the N period structure can be written as [11]

$$\hat{n}_{\mathrm{eff}}=\frac{C}{\omega N a}\left\{\tan^{-1}[z\tan(N\beta)\cot(\beta)]+\mathrm{Int}\left[\frac{N\beta}{\pi}+\frac{1}{2}\right]\pi\right\}, \tag{1}$$

where β is the Bloch phase for an infinite structure having the same unit cell as the finite structure in question. Eq. (1) contains all the information necessary for the phase-matching conditions of a generic multiwave mixing process. As an example, we consider a 40-period, mixed *half-wave/eighth-wave* PBG structure composed of air and a dispersive material. We choose this particular arrangement because it allows easy tuning of all the fields near their respective band edges and simultaneous access to a relatively high density of modes (DOM) [11,12].

In Fig. 1 we show that beginning with this basic structure it is possible to tune the fields so that the frequencies correspond to the Bloch phases given by

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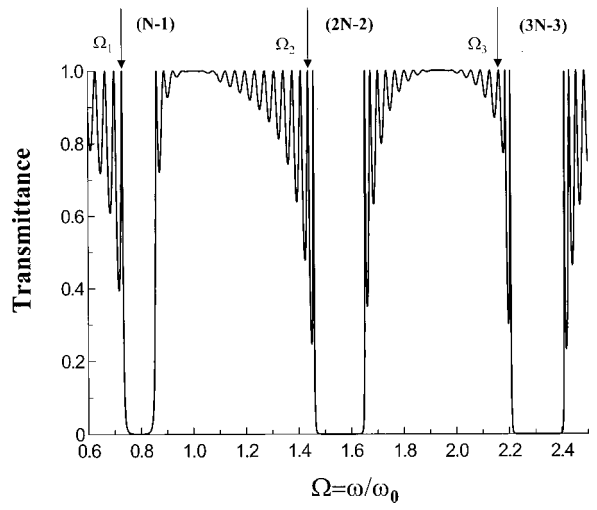

FIG. 1. Transmittance vs normalized frequency for a 20-period mixed λ/2-λ/8 structure. We use $n_1=1$ for the low index material at all frequencies. We have introduced a small amount of dispersion, typical of semiconductors, such that: $n_2(\Omega_3)=1.481$, $n_2(\Omega_2)=1.453$, and $n_2(\Omega_1)=1.428\,571\,4$. Under these conditions material and geometrical dispersion combine to provide precise phase-matching conditions at ω, 2ω, and 3ω near their respective band edges as follows: Ω_1 at the $(N-1)$th resonance, Ω_2 at the $(2N-2)$th resonance, and Ω_3 at the $(3N-2)$th resonance.

$$\beta_\omega=\frac{\pi}{N}(N-1),\quad \beta_{2\omega}=\frac{\pi}{N}(2N-2),\quad \beta_{3\omega}=\frac{\pi}{N}(3N-3). \tag{2}$$

For a reference wavelength of 1.1 μm, the frequency ω corresponds to $\lambda=1.5\,\mu$m; 2ω has $\lambda=0.75\,\mu$m; and 3ω has $\lambda=0.5\,\mu$m. The phase-matching condition for SHG is satisfied if $\beta_{2\omega}=2\beta_\omega$ [11]. Phase matching for the sum-frequency generation will also be satisfied if $\beta_{3\omega}=\beta_{2\omega}+\beta_\omega$. Our Eqs. (2), and Fig. 1, show that both these conditions are fulfilled simultaneously for our generic structure. Therefore, the description of the effective index given in Eq. (1) and Ref. [11] in terms of the Bloch phase and phase-matching conditions at resonance peaks amounts to the simple task of counting resonances, at least in the case of periodic structures [11]. For aperiodic structures the effective index formulation discussed in Ref. [11] is still valid, although an alternative to our Eq. (1) must be sought.

We assume we are operating in a region where losses can be neglected, and the nonlinear material is distributed in the high index layers only [12], with $\chi^{(2)}\approx 144$ pm/V. Although this value may seem somewhat large, the precise magnitude of $\chi^{(2)}$ is not important because we compare a PBG structure with a bulk material that has the same nonlinear coefficient and the same length, which in this case is roughly equivalent to one coherence length for the third harmonic (TH) process. Therefore, the reader should focus on the enhancement that comes about from the linear geometrical dispersion of the structure, which will hold for any $\chi^{(2)}$ value. The equations that we integrate are derived from Maxwell's equation for the field in a regime that we refer to as the slowly varying envelope approximation in time (SVEAT) [12,13]. We consider pulse propagation in the presence of large index discontinuities, and so we retain all second-order spatial derivatives. However, we assume that pulse envelopes initially have duration much greater than the optical cycle during the entire interaction. The method of integration is based on a modification of the split-step algorithm that can handle reflections, and is discussed in detail in Ref. [13]. The fields can arbitrarily and conveniently be written as

$$E(z,t)=\sum_1^3 \varepsilon_j(z,t)\exp[(k_jz-\omega_jt)]+\text{c.c.},$$

$$P_{NL}(z,t)=\sum_1^3 P_j(z,t)\exp[(k_jz-\omega_jt)]+\text{c.c.},$$

where ε_j and P_j are complex envelope functions. This decomposition highlights the fields' angular frequencies. We assume $\omega_1=\omega$, $\omega_2=2\omega$, and $\omega_3=\omega_2+\omega_1=3\omega$ with $k_1\equiv k=\omega/c$, $k_2=2\omega/c$, and $k_3=3\omega/c$. It can then be shown that the nonlinear polarization takes the following form:

$$\begin{aligned}P_{\rm NL}(z,t)=\chi^{(2)}E^2(z,t)&=2\chi^{(2)}\{\varepsilon_\omega^*\varepsilon_{2\omega}+\varepsilon_{2\omega}^*\varepsilon_{3\omega}\}e^{i(kz-\omega t)}\\&+2\chi^{(2)}\{\varepsilon_\omega^*\varepsilon_{3\omega}+\tfrac{1}{2}\varepsilon_\omega^2\}\exp[(2kz-2\omega t)]\\&+2\chi^{(2)}\varepsilon_\omega\varepsilon_{2\omega}\exp[i(3kz-3\omega t)]+\text{c.c.}\end{aligned} \tag{3}$$

Since we are considering harmonics of a fundamental field, the right-hand side of Eq. (3) contains multiple contributions to the nonlinear source terms that are usually neglected under the rotating wave approximation. These terms were also noted and discussed early on in Ref. [1], and to our knowledge have not been studied in this context before. The fields at 2ω and 3ω are initially zero everywhere, and are generated in the backward and forward directions. Our choice of wave vectors corresponds to conditions consistent with a pump field initially propagating in free space, located away from the structure. Any phase modulation effects that arise from multiple reflections and nonlinear interactions are fully accounted for in the dynamics of the complex field envelopes. Therefore, assuming that pulses never become so short as to violate SVEAT, we integrate the equations of motion following the numerical model set forth in Refs. [12], [13], which for the three-wave mixing process are,

$$\begin{aligned}n_{2\Omega}^2(\xi)\frac{\partial}{\partial\tau}\varepsilon_{2\Omega}(\xi,\tau)&=\frac{i}{4\pi 2\Omega}\frac{\partial^2}{\partial\xi^2}\varepsilon_{2\Omega}(\xi,\tau)-\frac{\partial}{\partial\xi}\varepsilon_{2\Omega}(\xi,\tau)\\&+i\pi[n_{2\Omega}^2(\xi)-1]2\Omega\varepsilon_{2\Omega}(\xi,\tau)\\&+i8\pi^2 2\Omega\chi^{(2)}\{\varepsilon_{3\Omega}\varepsilon_\Omega^*+\tfrac{1}{2}\varepsilon_\Omega^2\},\end{aligned} \tag{4}$$

$$\begin{aligned}n_{3\Omega}^2(\xi)\frac{\partial}{\partial\tau}\varepsilon_{3\Omega}(\xi,\tau)&=\frac{i}{4\pi 3\Omega}\frac{\partial^2}{\partial\xi^2}\varepsilon_{3\Omega}(\xi,\tau)-\frac{\partial}{\partial\xi}\varepsilon_{3\Omega}(\xi,\tau)\\&+i\pi[n_{3\Omega}^2(\xi)-1]3\Omega\varepsilon_{3\Omega}(\xi,\tau)\\&+i8\pi^2 3\Omega\chi^{(2)}\varepsilon_{2\Omega}\varepsilon_\Omega,\end{aligned} \tag{5}$$

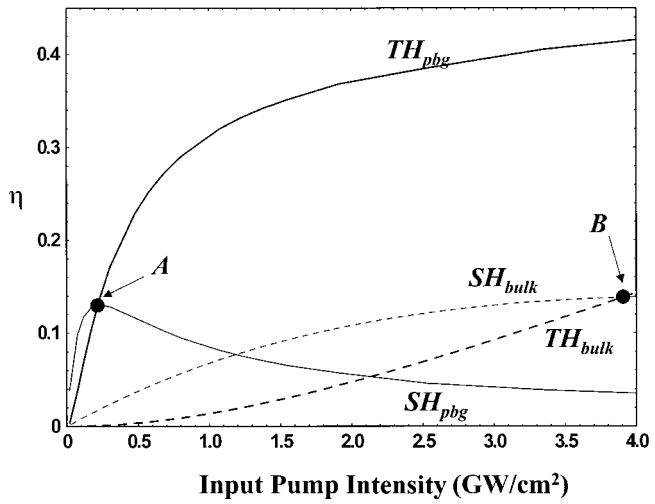

FIG. 2. SH and TH conversion efficiencies as functions of the pump peak intensity for the PBG structure (solid lines) and an ideal case of a plane wave propagating in a phase-matched bulk (dashed lines) of the same length and nonlinear coefficient as the PBG structure. Points *A* and *B* identify the peak input intensities for which the conversion efficiencies of second and third harmonic generation are equal.

$$n_\Omega^2(\xi)\frac{\partial}{\partial\tau}\varepsilon_\Omega(\xi,\tau)=\frac{i}{4\pi\Omega}\frac{\partial^2}{\partial\xi^2}\varepsilon_\Omega(\xi,\tau)-\frac{\partial}{\partial\xi}\varepsilon_\Omega(\xi,\tau)+i\pi[n_\Omega^2(\xi)-1]\Omega\varepsilon_\Omega(\xi,\tau)+i8\pi^2\Omega\chi^{(2)}\{\varepsilon_{2\Omega}^*\varepsilon_{3\Omega}+\varepsilon_{2\Omega}\varepsilon_\Omega^*\}. \quad (6)$$

The complexity of the system of equations (4–6) is immediately apparent. In Fig. 2 we depict the conversion efficiency η, defined as the ratio between the total energy of the generated field to the total energy of the input pulse, for the 2ω and 3ω fields as functions of the input peak intensities. We assume incident pump pulses are approximately 2 ps in duration, which satisfy SVEAT. We also plot η for a plane wave propagating in an exactly phase-matched bulk material that has the same $\chi^{(2)}$ and the same length as the PBG structure. *These conditions represent the best-case scenario, described in Ref. [1], for a material that in reality does not exist*. The dynamics is typical of a phase-matched interaction, though some notable differences arise. The crossing points denoted by *A* and *B* in Fig. 2 represent input peak powers that yield the same conversion efficiency for second and third harmonic generation. We note that for the ideal bulk material *B* occurs at ~4 GW/cm^2, while for the PBG structure *A* occurs at ~0.2 GW/cm^2, a factor of 20 smaller for the PBG. In addition, in the neighborhood of 0.2 GW/cm^2, the PBG conversion efficiency is at least two orders of magnitude greater than bulk's conversion efficiency. In the case of the PBG structure, using 2 ps pulses also makes a difference. The pulse is not only finite in extent, its bandwidth is also comparable to the bandwidth of the band edge resonance, and so part of the incident energy is back reflected. To some extent, TH conversion efficiency increases as pulse duration increases, as previously observed for SHG [12].

An examination of the figure reveals that, remarkably, the PBG structure acts not only as a phase-matched bulk, a behavior to our knowledge impossible to reproduce with any known bulk material, but it also enhances the interaction due to the increased density of modes. We note that using a structure with more periods leads to further decreases in the threshold peak power, especially the power necessary to reach crossing point *A*, because in that case one can exploit even stronger field localization effects that occur near the band edge. For example, the band edge resonances discussed display a DOM about ten times higher than that of bulk. This rather small DOM can be drastically increased with very modest increases in structure length, since the DOM increases nonlinearly as N^2 [14]. This kind of interplay also strongly suggests that the geometry of the structure can effectively be used to offset small $\chi^{(2)}$ values, and we now provide an example.

Recent publications have shown reasonable nonlinear coefficients in sputtered AlN thin films (of order 10 pm/V) due to the columnar nature of film growth, which provides the anisotropy required for the onset of second-order nonlinearities [15]. Also porous silicon structures may be fabricated and engineered to the desired specifications, as recent experiments showed [16]. The nonlinearity may come as a result of pore asymmetry [16], and possibly doping. Alternatively, one may resort to materials with relatively low nonlinear coefficients, and drastically improve on the density of modes by increasing structure length. This may require further refinement of growing and deposition methods.

As an example, we follow the suggestions of Ref. [15], and consider a 30-period AlN/SiO_2 multilayer stack. As pointed out in Ref. [15], this kind of stack is particularly attractive because it might provide an alternative to more expensive molecular beam epitaxy fabrication processes. In Fig. 3(a) we show the tuning conditions on the fields; in Fig. 3(b) we show the results of the integration of equations (4–6) for this sample, and compare with SHG and third harmonic generation (THG) from one coherence length (approximately 7 μm for THG) of AlN. Since the nonlinearity is relatively small, we consider the undepleted pump regime. The parameters and the dispersion laws for both materials are given in the caption. We found that while SHG can be exactly phase matched, i.e., $\beta_{2\omega}=2\beta_\omega$, geometrical dispersion cannot fully compensate the material dispersion of both materials. That is, $\beta_{3\omega}\neq\beta_\omega+\beta_{2\omega}$, and the TWM process is slightly mismatched. Nevertheless, the TH signal is tuned to the second resonance near its band edge, as shown in Fig. 3(a), leading to a slight phase mismatch. The results depicted in Fig. 3(b) suggest that THG occurs at a rate ~300 times greater than bulk THG, while SHG is increased by at least one order of magnitude with respect to bulk.

These remarkable results show that it is not necessary to operate under perfect phase-matched conditions in order to have efficient, simultaneous second and third harmonic gen-

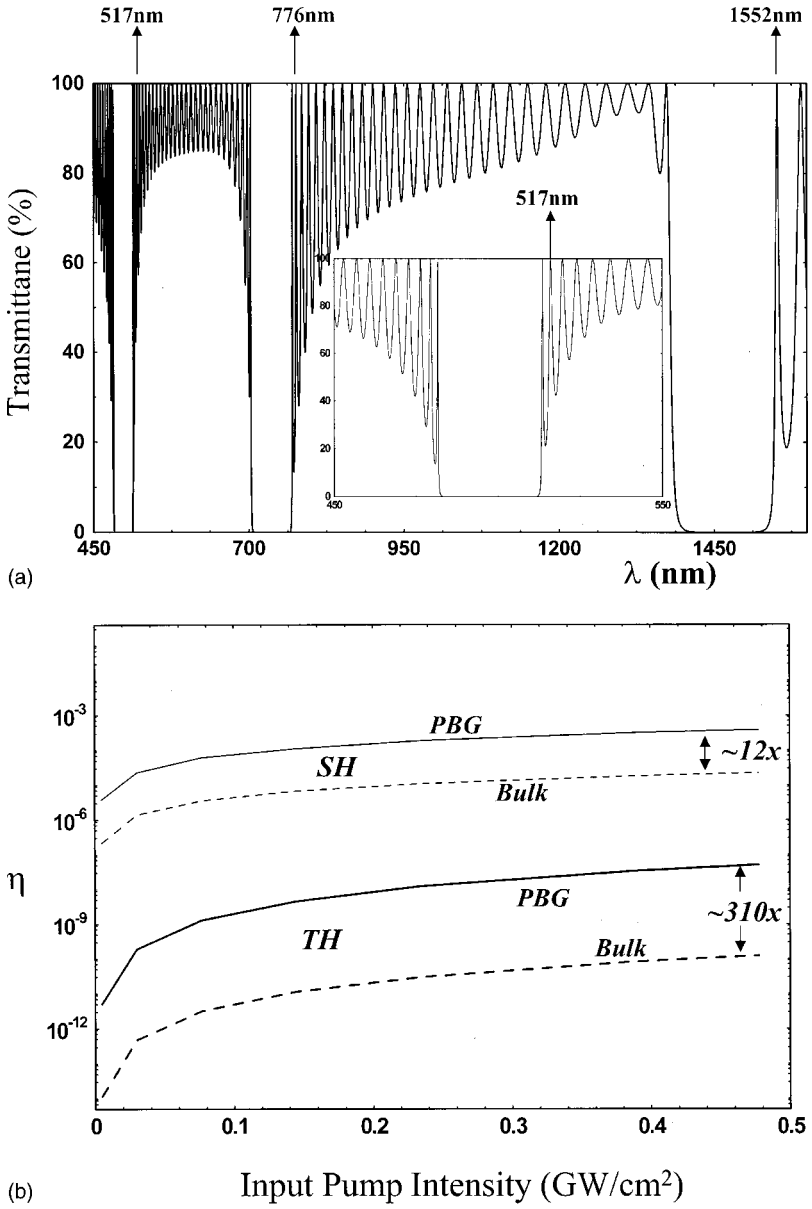

FIG. 3. (a) Linear transmission vs wavelength for a 30-period AlN(292.6 nm)/SiO_2 (91.38 nm) stack. (b) SH and TH conversion efficiencies as functions of peak input intensity for the stack of (a). The dispersion functions are as follows: $n_{\mathrm{AlN}}(\lambda)=2.0016+1.86810^{-2}/\lambda^2-1.892810^{-4}/\lambda^4$ [18] and $n_{\mathrm{SiO_2}}(\lambda)=1.5028+5.0227\times10^{-3}/\lambda^2-8.2382\times10^{-5}/\lambda^4$ (ubk7-517643. Source: Schott-Glass catalog). We compare with a bulk AlN approximately 7 μm in length.

eration. They also show that these devices are flexible, make the design process considerably easier, and relax experimental tolerances considerably while retaining relatively high conversion efficiencies. Finally we note that high conversion efficiencies in THG require a triply resonant system. However, we have seen that these conditions are not difficult to fulfill for AlN/SiO_2. In any case, the purpose of the previous example is to provide motivation for experimental work. If materials with larger nonlinearities are desired, then $Al_xGa_{1-x}N$/AlN stacks are good candidates. In fact, it can be shown that the dispersion of $Al_xGa_{1-x}N$ and AlN [17] can be almost completely overcome by the ensuing geometrical dispersion in favor of the production of efficient, phase matched, and enhanced THG at wavelengths in the 500 nm range. In short, it is possible to design structures with features similar to those described in Fig. 3 using materials that have larger nonlinearities.

In conclusion, we have presented a new way to achieve simultaneous, globally phase-matched, enhanced, collinear second and third harmonic generation in multilayer stacks having a second-order nonlinearity only. The process is based on judiciously combining and balancing geometrical and material dispersion, and does not require any complicated material processing such as poling. The process also remains efficient under a variety of tuning conditions characterized by small departures from exact phase matching. The ability to exactly phase-match multicolor interactions is quite remarkable in the face of its impossibility in ordinary materials, even for higher harmonic generation. Even though one may argue that the overall conversion efficiencies that we discussed may be obtained with current state-of-the-art devices, the gain per unit length that results from a drastic reduction in sample length due to field localization effects is several orders of magnitude larger compared to the gain per unit length of any state-of-the-art, quasi-phase matched device. For this reason we remain confident that the devices and methods we propose will become an attractive alternative as the miniaturization trend continues.

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