Non-phase-matched enhancement of second-harmonic generation in multilayer nonlinear structures with internal reflections

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Traditional notions of second-harmonic generation rely on phase matching or quasi phase matching to achieve good conversion efficiencies. We present an entirely new concept for efficient second-harmonic generation that is based on the interference of counterpropagating waves in multilayer structures. Conversion efficiencies are an order of magnitude larger than with phase-matched second-harmonic generation in similar multilayer structures. © 2004 Optical Society of America

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Phase matching (PM) or quasi phase matching has always been central to any method of generating an efficient second harmonic (SH).[1] Developments in the field have shown that it is possible to achieve PM in infinite[2] and finite[3] multilayer stacks, also called one-dimensional photonic crystals (1D-PCs). Finite structures may also provide reduced group velocities, resulting in large field enhancements of the fundamental field (FF) and the SH fields. Recent experiments on PM AlGaAs/AlO_x multilayers have confirmed that the conversion efficiency scales as L^6, where L is the length of the 1D-PC.[4] Dumeige *et al.*[4] predicted that a 55-period, $L = 15\ \mu$m, AlGaAs/AlO_x structure will have a 10% conversion efficiency for a fundamental peak power of 1 kW. The spectral bandwidth of the enhancement is 0.25 nm and is comparable with pulse durations of 15 ps or longer. Bandwidths can be expanded by cascading several 1D-PCs in series.[5]

In Ref. 3 it was shown that PM could be achieved with alternating layers of optical thickness $\lambda/2$ and $\lambda/4$ with the nonlinearity in the high-refractive-index, half-wave layers. Optimum PM occurs for the FF tuned to the first transmission resonance near the lowest energy stop band and the SH field at the second transmission resonance next to the second-order stop band (solid arrows in Fig. 1). The PM conditions for a 1D-PC force the SH to be at the second transmission resonance, where the mode density is not at its highest value. As was shown earlier,[3] detuning from the PM condition degrades the conversion efficiency. However, we show here that non-phase-matched 1D-PC designs can yield a further order of magnitude of improvement in the conversion efficiency. In analyzing these new designs we find that the enhancement is due to a combination of high photon-mode density and a contribution to fast-varying interference terms that average to near zero if the thickness of the nonlinear layer is a half-wave or greater. This enhancement is unique to a 1D-PC, since neither bulk material nor the smallest cavity, i.e., $\lambda/2$, will display this type of contribution to nonlinear dynamics from the interference of counterpropagating waves.

In the study reported in Ref. 6 a multiple-scale approach was used to derive an analytical expression for the conversion efficiency for a generic layered structure of finite length composed of nonabsorbing media. It was found that the conversion efficiency in the undepleted pump approximation is proportional to the square modulus of an effective coupling coefficient, defined as

$$\tilde{d}_{\mathrm{eff}} = \frac{1}{L}\int_0^L \chi^{(2)}(z)\Phi_\omega{}^2(z)\Phi_{2\omega}{}^*(z)\mathrm{d}z\,. \tag{1}$$

Here L is the length of the structure, and $\Phi_\omega(z)$ and $\Phi_{2\omega}(z)$ are the complex, linear field profiles normalized

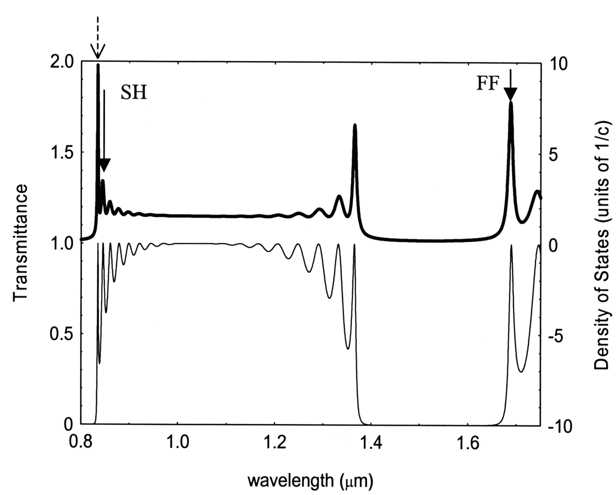

Fig. 1. Transmission spectrum and density of states (thick curve) versus wavelength at normal incidence for a 20-period mixed half-wave–quarter-wave stack. FF and SH tuning are indicated by the arrows.

with respect to a unitary input field, so that the electric field inside the structure can be written as $E_{i\omega}(z) = E_{i\omega}{}^0[\Phi_{i\omega}(z) + \text{c.c.}]$, where $i = 1, 2$ and $E_{i\omega}{}^0$ is the amplitude of the input field. We emphasize that, unlike d_{eff} defined for studying nonlinear propagation in waveguides,[7,8] our $\tilde{d}_{\text{eff}}$ is a complex quantity that contains information regarding field distribution and localization, as well as contributions to the conversion efficiency coming from the PM conditions. In particular, it is easy to show that for a simple case of SH generation in an infinite periodic structure the expression of the coupling coefficient becomes $\tilde{d}_{\text{eff}} = I_{\text{u.c.}} \sum_m \delta(\Delta k_\beta - {}^{2\pi}/_{\Lambda}\, m)$, $m = 0, 1, 2, \ldots$, where Λ is the thickness of the unit cell, $\Delta k_\beta = k_\beta(2\omega) - 2k_\beta(\omega)$ is the Bloch wave vector mismatch and $I_{\text{u.c.}}$ is the overlap integral calculated over the unit cell by the formula

$$I_{\text{u.c.}} = \frac{1}{\Lambda}\int_0^\Lambda \chi^{(2)}(z) f_\omega{}^2(z) f_{2\omega}{}^*(z)\exp(i\Delta k_\beta z)\mathrm{d}z\,, \quad (2)$$

which is obtained by writing the fields as $\Phi_{i\omega}(z) = f_{i\omega}(z)\exp[ik_\beta(i\omega)z]$, where $i = 1, 2$ and $f_{i\omega}(z + \Lambda) = f_{i\omega}(z)$. $I_{\text{u.c.}}$ is a form factor and contains all the information about the geometry such as layer thickness, refractive-index contrast, amount and position of nonlinear material inside the unit cell, and overlap of the fields. In the case of finite structures the break of the translational symmetry is responsible for field localization effects; $f_{i\omega}(z)$ are no longer periodic over unit cell thickness Λ. For a finite structure the correct expression that replaces Eq. (1) becomes

$$\begin{aligned}\tilde{d}_{\text{eff}} &= \frac{1}{N}\sum_{j=0}^{N-1}\left[\frac{1}{\Lambda}\int_0^\Lambda \chi^{(2)}(z) f_\omega{}^2(j\Lambda + z) f_{2\omega}{}^*(j\Lambda + z) \right.\\ &\quad \left.\times\ \exp(i\Delta k_\beta z)\mathrm{d}z\right]\exp[i(\Delta k_\beta\Lambda)j]\\ &= \frac{1}{N}\sum_{j=0}^{N-1} I_j{}^{\text{u.c.}}\exp[i(\Delta k_\beta\Lambda)j]\,. \quad (3)\end{aligned}$$

Thus every unit cell gives a different contribution to the overlap integral. These results suggest that we have N summation terms of different amplitudes and phases. In other words, standard theories of PM commonly used to find the optimum conditions may fail in a wide range of cases. As an example we choose a structure with a mixed quarter-wave–half-wave geometry and 20 periods. The nonlinear material ($\lambda/4$ optical thickness) has a refractive index $n_2(\omega_{\text{FF}}) = 1.428$ at the FF frequency; for simplicity, the linear material ($\lambda/2$ optical thickness) is assumed to be air, with $n_1 = 1$. The reference wavelength used to calculate the optical paths of the layers is 1 μm, corresponding to angular frequency $\omega_0 = 1.88 \times 10^{15}\ \text{s}^{-1}$. Assuming normal incidence, this simple geometrical arrangement allows us to rather easily tune the FF and SH fields to the two resonance peaks, each of which is located near two consecutive bandgaps of the transmission spectrum, where field localization effects are maximized. We tune the FF at the first-order band-edge resonance ($\lambda_{\text{FF}} = 1.69\ \mu$m, which corresponds to $\omega_{\text{FF}} = 0.592\omega_0$, as labeled by the arrow labeled FF in Fig. 1). Once the layer thicknesses have been chosen, one may add dispersion by varying the index of refraction at the SH frequency ($\omega_{\text{SH}} = 1.184\omega_0$) to tune the field to any desired resonance near the band edge. By increasing the value of $n_2(\omega_{\text{SH}})$, it is possible to tune the SH closer to the band edge. In Fig. 2(a) we depict the polar plots containing the modulus and the angular phase of the N terms of Eq. (3) when the high-index material is given suitable dispersion (asterisks). In particular, we have chosen $n_2(\omega_{\text{SH}}) = 1.676$, which tunes the SH to the first band-edge resonance (see the dashed arrow in Fig. 1). Then we compare these results with those obtained when effective PM conditions are achieved [$n_2(\omega_{\text{SH}}) = 1.616$; SH is tuned at the second resonance peak]. We note that in this case (circles in Fig. 2) the amplitude of the sources is smaller because the density of modes for the SH field is smaller; i.e., localization of the fields is weaker. Moreover, the phases of the summation terms are spread over a wider angular range and the overall sum is not maximized. Indeed, the square modulus of the sum

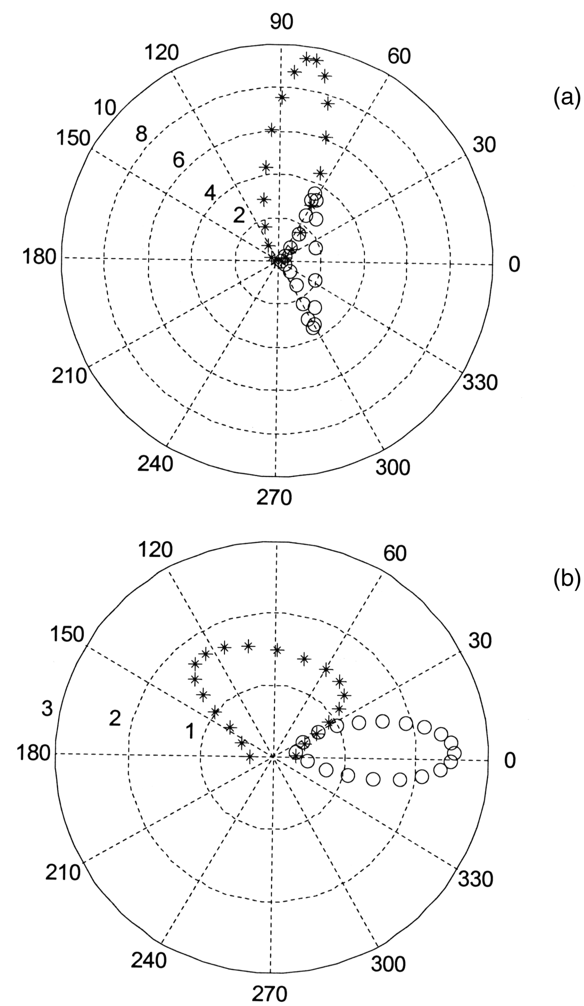

Fig. 2. Polar plots of the N summation terms of Eq. (3) when PM conditions are achieved (circles) and when both the FF and the SH fields are tuned at the band-edge resonance (asterisks). The optical thickness of the nonlinear material layers is (a) $\lambda_0/4$, (b) $\lambda_0/2$, with $\lambda_0 = 1\ \mu$m.

is one order of magnitude smaller than that calculated for the previous case. Therefore, according to our model the expected conversion efficiency when PM conditions are achieved is one order of magnitude lower than in the non-phase-matched case.

For completeness, we performed the same calculation for a structure designed so that PM conditions dominate. It is enough to invert the geometry, i.e., take the nonlinear layer to have optical thickness $\lambda/2$ and the linear layer to have $\lambda/4$. Once again the asterisks in Fig. 2(b) represent the case in which the SH field is tuned at the first resonance and the circles represent the phase-matched case. We note that for this structure fulfillment of PM conditions leads to optimum conversion efficiency, although the efficiency is one order of magnitude lower than that obtained with the previous structure in the non-phase-matched regime. Thus we switched from a regime in which PM conditions rule the nonlinear dynamics to a regime where the overlap of the fields dominates by simply changing the filling ratio of the nonlinear layer inside the unit cell.

To provide a qualitative interpretation of this phenomenon we decompose the complex linear field profiles as a superposition of forward and backward waves in each layer:

$$\Phi_{\omega}{}^{j,m}(z) = \{A_{j,m} \exp[ik_0 n_m{}^{\omega}(z - z_{j,m})] + B_{j,m} \exp[-ik_0 n_m{}^{\omega}(z - z_{j,m})]\}, \quad (4a)$$

$$\Phi_{2\omega}{}^{j,m}(z) = \{C_{j,m} \exp[i2k_0 n_m{}^{2\omega}(z - z_{j,m})] + D_{j,m} \exp[-i2k_0 n_m{}^{2\omega}(z - z_{j,m})]\}, \quad (4b)$$

where $j = 1, 2, \ldots N$; $m = 1, 2$; and A, B, C, and D, are constants that can be calculated by imposing boundary conditions at every interface. Substituting Eqs. (4) into the expression for the coupling coefficient, taking $\chi^{(2)}(z) = 0$ everywhere except within the nonlinear layers, and performing the integral in each layer, we obtain

$$\tilde{d}_{\text{eff}} = \frac{\chi^{(2)}dh}{L}\Bigg(\text{sinc}(k_0 \Delta n_2 dh) \times \sum_{j=0}^{N}[A_{j,2}{}^2 C_{j,2}{}^* \exp(ik_0 \Delta n_2 dh) + B_{j,2}{}^2 D_{j,2}{}^* \times \exp(-ik_0 \Delta n_2 dh)] + 2\,\text{sinc}(k_0 n_2^{2\omega} dh)\sum_{j=0}^{N} A_{j,2}B_{j,2} \times [C_{j,2}{}^* \exp(ik_0 n_2{}^{2\omega} dh) + D_{j,2}{}^* \exp(-ik_0 n_2{}^{2\omega} dh)] + \text{sinc}\left[2k_0\left(n_2^{2\omega} - \frac{\Delta n_2}{2}\right)dh\right]\sum_{j=0}^{N}\left\{A_{j,2}{}^2 D_{j,2}{}^* \times \exp\left[-i2k_0\left(n_2{}^{2\omega} - \frac{\Delta n_2}{2}\right)dh\right] + B_{j,2}{}^2 C_{j,2}{}^* \times \exp\left[i2k_0\left(n_2{}^{2\omega} - \frac{\Delta n_2}{2}\right)dh\right]\right\}\Bigg), \quad (5)$$

where $\Delta n_2 = n_2{}^{2\omega} - n_2{}^{\omega}$. We note that the relevance of the three terms on the right-hand side of Eq. (5) depends on the nonlinear layer thickness dh because of the sinc functions. Since there is no local PM because of the natural material dispersion of the high-index layers, the overlap of the fields cannot be maximized over an arbitrary nonlinear layer of length dh. In particular, the first term is related to the material index mismatch, and the second and third terms give almost zero contributions if the optical thicknesses of the nonlinear layer are longer than approximately $\lambda/2$ and $\lambda/4$, respectively. Those fast-varying terms arise from interference of counterpropagating waves inside the structure generated by multiple reflections at the interfaces and are generally neglected in bulk or microcavity theories. Nevertheless, in the case that we showed, the terms in Eq. (5) cannot generally be neglected.

In conclusion, it is possible to take full advantage of field localization effects in such a way as to weaken the role played by effective PM conditions in the nonlinear dynamics. We have shown that higher SH generation conversion efficiency is achieved when the fast-varying terms in the nonlinear polarization related to the presence of counterpropagating waves are not negligible and by properly tailoring the size and distribution of the nonlinear layers in spite of fulfilling PM conditions.

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