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Giant enhancement of second harmonic generation in multiple photonic quantum well structures made of nonlinear material

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We present a general solution of second harmonic generation (SHG) in one-dimensional (1D) inhomogeneous systems. The structure consists of 1D multiple photonic quantum wells (PQWs) made of optical nonlinear material. The optimal arrangement of the polarization directions of the ferroelectric domains in multiple PQWs is determined by a simulated annealing method. We find that the conversion efficiency of SHG can be significantly enhanced when the fundamental wave frequency is aimed at one of the defect states. By tuning the polarization directions of the domains, the conversion efficiencies of "forward" and "backward" SHGs can be flexibly changed. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2181607]

Photonic crystals (PCs) with periodic modulated dielectric function have attracted considerable interest because of many novel effects in the aspects of fundamental physics and potential applications. Previous studies on second harmonic generation (SHG) in PCs are mainly focused on the characteristics of the second harmonic wave (SHW) when frequency of the fundamental wave (FW) approaches its band edge, in which the density of electromagnetic field becomes very large, the field is highly localized, thus the conversion efficiency should be enhanced significantly.[1–3] If a defect structure made of nonlinear material is introduced into PCs, some defect states within the photonic band gap (PBG) are produced and give rise to strong localized fields. In this system, it is expected that the conversion efficiency might be increased considerably.[4] However, the conversion efficiency of "forward" or "backward" SHGs is uncontrollable.

The slowly varying amplitude approximation (SVAA) is widely adopted in nonlinear optics, however, the SVAA is invalid when the reflection of a SHW cannot be neglected.[5] For the inhomogeneous system, the reflection of a SHW is so strong that the SVAA may bring inaccurate results. Motivated by these works, in this Letter, we present a general solution of SHG in a one-dimensional (1D) inhomogeneous system. We investigate the structure composed of multiple photonic quantum wells (PQWs) made of ferroelectric nonlinear material. We find that the conversion efficiency of SHG can be remarkably enhanced when the FW frequency has taken aim at one of the defect states and that both forward and backward SHGs can be controlled by adjusting polarization directions of ferroelectric domains with the use of a simulated annealing (SA) algorithm.[6–9]

The typical sample studied is composed of alternatively stocked dielectric-dielectric or air layers with different refractive indices. We assume that interface of layers is laid on the yz plane. The incident light is normally launched upon the surface of the sample. In a direct nondepleted pump wave approximation, for the lth layer, the FW (SHW) electric field $E_l^{(1)}(x)$ $[E_l^{(2)}(x)]$ satisfies the following equations[10]:

$$\left[\frac{d^2}{dx^2}+k_l^{(1)2}\right]E_l^{(1)}(x)=0, \tag{1}$$

$$\left[\frac{d^2}{dx^2}+k_l^{(2)2}\right]E_l^{(2)}(x)=-k_{20}^2\chi_l E_l^{(1)2}(x), \tag{2}$$

where $k_l^{(1)}=n_l^{(1)}k_{10}$, $k_l^{(2)}=n_l^{(2)}k_{20}$, $k_{10}=\omega/c$, and $k_{20}=2\omega/c$. c is the light speed in vacuum and $n_l^{(1)}$ $(n_l^{(2)})$ is the refractive index of the lth layer material at the FW (SHW) frequency. χ_l is nonlinear optical coefficient of the lth layer.

The FW electrical field in the lth layer of the sample can be expressed as

$$E_l^{(1)}(x)=A_l^{(1)}e^{ik_l^{(1)}(x-x_{l-1})}+B_l^{(1)}e^{-ik_l^{(1)}(x-x_{l-1})}, \tag{3}$$

where x_1 sets 0, $x_l=x_{l-1}+d_l$ $(l=2,3,\ldots)$, and d_l is the thickness of the lth layer. $A_l^{(1)}$ and $B_l^{(1)}$, respectively, represent the amplitudes of the forward and backward FW waves at the interface. The presence of layer interfaces should cause strong scattering of light waves due to inhomogeneity of the system, different from homogeneous structures most reported in the literature.[5]

By using the continuous condition of the fields at interface, satisfied by the electrical field and magnetic field, or the mode match technique, we derive

$$\begin{pmatrix}A_{l+1}^{(1)}\\ B_{l+1}^{(1)}\end{pmatrix}=\begin{pmatrix}t_{11}^{(l)} & t_{12}^{(l)}\\ t_{12}^{(l)*} & t_{11}^{(l)*}\end{pmatrix}\begin{pmatrix}A_l^{(1)}\\ B_l^{(1)}\end{pmatrix}=\hat{T}_l\begin{pmatrix}A_l^{(1)}\\ B_l^{(1)}\end{pmatrix} \tag{4}$$

with the elements

$$t_{11}^{(l)}=\frac{k_{l+1}^{(1)}+k_l^{(1)}}{2k_{l+1}^{(1)}}e^{ik_l^{(1)}d_l},$$

$$t_{12}^{(l)}=\frac{k_{l+1}^{(1)}-k_l^{(1)}}{2k_{l+1}^{(1)}}e^{-ik_l^{(1)}d_l}. \tag{5}$$

The overall transfer matrix is obtained from cascading product of the successive individual transfer matrix of $\hat{T}_l$. Thus, the relative amplitudes $A_l^{(1)}$ and $B_l^{(1)}$ of every layer can be completely determined.

We turn to consider the SHW electric field. In the lth layer, it can be expressed by a form as

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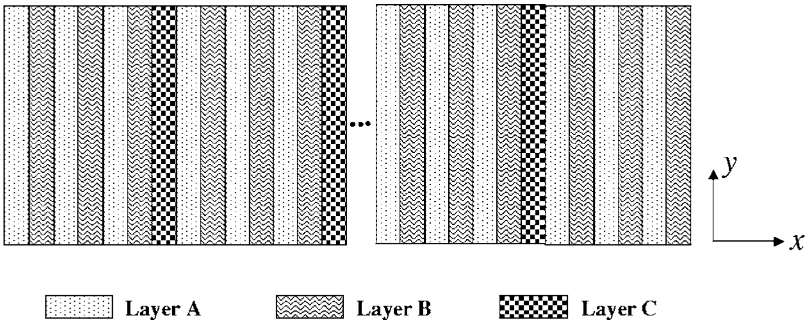

FIG. 1. Schematic of 1D multiple photonic quantum well structures made of nonlinear material. We denote it by $[(AB)_m(C)]_n(AB)m$ according to the sequence of the stacking layers. Layers A, B, and C correspond to the nonpoled $BaTiO_3$, air, and the poled $LiNbO_3$ crystal, respectively. The stacked growth direction is the x axis, and each layer lies on the yz plane. The polarization orientation of layer C ($LiNbO_3$ crystal) is parallel to the $\pm z$ axis; thus, its optical nonlinear coefficient is d_{33} or $-d_{33}$.

$$E_l^{(2)}(x) = A_l^{(2)} e^{ik_l^{(2)}(x-x_{l-1})} + B_l^{(2)} e^{-ik_l^{(2)}(x-x_{l-1})} + C_l^{+} e^{i2k_l^{(1)}(x-x_{l-1})} + C_l^{-} e^{-i2k_l^{(1)}(x-x_{l-1})} - \frac{2k_{20}^2\chi_l}{k_l^{(2)2}} A_l^{(1)} B_l^{(1)}, \tag{6}$$

where $A_l^{(2)}$, $B_l^{(2)}$ represent the amplitudes for the forward and backward SHWs, respectively. Substituting Eqs. (3) and (6) into Eq. (2), we have

$$C_l^{+} = \frac{-k_{20}^2\chi_l A_l^{(1)2}}{k_l^{(2)2} - 4k_l^{(1)2}}$$

and

$$C_l^{-} = \frac{-k_{20}^2\chi_l B_l^{(1)2}}{k_l^{(2)2} - 4k_l^{(1)2}}. \tag{7}$$

We will now determine the values of $A_l^{(2)}$ and $B_l^{(2)}$. We consider the continuous condition of the electric and magnetic fields at interface and that

$$\begin{pmatrix} A_{l+1}^{(2)} \\ B_{l+1}^{(2)} \end{pmatrix} = \begin{pmatrix} q_{11}^{(l)} & q_{12}^{(l)} \\ q_{12}^{(l)*} & q_{11}^{(l)*} \end{pmatrix} \begin{pmatrix} A_l^{(2)} \\ B_l^{(2)} \end{pmatrix} + \begin{pmatrix} f_{l+} \\ f_{l-} \end{pmatrix} \tag{8}$$

with the elements

$$q_{11}^{(l)} = \frac{k_{l+1}^{(2)} + k_l^{(2)}}{2k_{l+1}^{(2)}} e^{ik_l^{(2)} d_l},$$

$$q_{12}^{(l)} = \frac{k_{l+1}^{(2)} - k_l^{(2)}}{2k_{l+1}^{(2)}} e^{-ik_l^{(2)} d_l},$$

$$f_{l\pm} = -\frac{k_l^{(2)} \pm 2k_l^{(1)}}{2k_l^{(2)}} C_l^{+} - \frac{k_l^{(2)} \mp 2k_l^{(1)}}{2k_l^{(2)}} C_l^{-}$$

$$+ \frac{k_l^{(2)} \pm 2k_{l-1}^{(1)}}{2k_l^{(2)}} C_{l-1}^{+} e^{i2k_{l-1}^{(1)} d_{l-1}} + \frac{k_l^{(2)} \mp 2k_{l-1}^{(1)}}{2k_l^{(2)}} C_{l-1}^{-} e^{-i2k_{l-1}^{(1)} d_{l-1}}$$

$$+ \frac{k_{20}^2\chi_l}{k_l^{(2)}} A_l^{(1)} B_l^{(1)} - \frac{k_{20}^2\chi_{l-1}}{k_{l-1}^{(2)}} A_{l-1}^{(1)} B_{l-1}^{(1)}.$$

Considering the initial condition $A_1^{(2)}=0$ and $B_N^{(2)}=0$, the conversion efficiencies of the forward and backward waves are evaluated respectively by

$$\eta_{\text{forth}} = \frac{n_N^{(2)} |A_N^{(2)}(x_{N-1})|^2}{n_1^{(1)} |A_1^{(1)}|^2} \tag{9a}$$

and

$$\eta_{\text{back}} = \frac{|B_1^{(2)}(x_1)|^2}{|A_1^{(1)}|^2}, \tag{9b}$$

where N denotes the total number of layers in sample.

We now investigate the sample, which is composed of alternating stacked dielectric-air-nonlinear material layers with finite number and embedded in air, as shown in Fig. 1. For convenience, we denote the structural configuration as $[(AB)_m(C)]_n(AB)_m$. Layers A, B, and C possess different refractive indices. Layers $(AB)_m$ can be regarded as Bloch mirrors for the FW and C as nonlinear material defect layer. This is the so-called optical quantum wells (PQWs). It is noted that the polarization direction of ferroelectric domain of layers C may be inverse. We choose $m=4$ and $n=80$ in calculation and assume that layer A (B) is made by the nonpoled $BaTiO_3$ (air) material for determinacy. The refractive index and thickness of layer $A(B)$ are $n_A=2.4$ ($n_B=1$) and $d_A=0.360$ μm ($d_B=0.640$ μm), respectively. Layer C is chosen as the poled $LiNbO_3$ crystal with a thickness of $d_C=3.909$ μm. Its refractive index is referred to Ref. 11 and its nonlinear coefficient d_{33} is 47.0 pm/V, referred to Ref. 12. The intensity of the incident FW wave sets $I=1.328\times 10^9$ W/m^2, corresponding to $|E_1^{(1)}(x_1)|^2=1.00$ V^2/μm^2.

We first calculate the transmission probability spectrum, as shown in Fig. 2: (a) for the perfect truncated (AB) PC, i.e., $d_C=0.0$ μm; and (b) for the multiple photonic quantum well structures made of nonlinear material. It is clearly seen from Fig. 2(a) that photonic band gaps (PBGs) exist in the range of 0.590–0.615, 0.712–0.793, and 0.942–1.08 μm. It is seen from Fig. 2(b) that there are three strong peaks appearing within the PBG of 0.942–1.080 μm. They correspond to defect modes. One of the defect modes is just assigned to 1.064 μm under the chosen parameters. To obtain high conversion efficiency, we select the FW wavelength 1.064 μm, assigning to one of the defect states. The polarization of domains is parallel to the z axis, the nonlinear coefficient takes d_{33} or $-d_{33}$, corresponding to the positive or negative polarization of domain of layer C. The arrangement of polarization directions of layers C should be optimized with the use of the SA method for implementing SHG with highest conversion efficiency. The objective function used in the SA is chosen as

$$D = |\eta_1^0 - \eta_{\text{forth}}(\lambda)| + |\eta_2^0 - \eta_{\text{back}}(\lambda)|, \tag{10}$$

where $\lambda=1.064$ μm and $\eta_{1,2}^0$ the preset constant. By choosing different values of η_1^0 and η_2^0, the conversion efficiencies

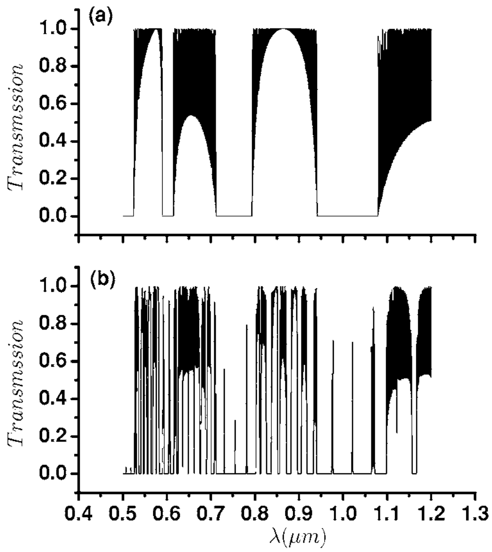

FIG. 2. Transmission spectrum for two samples: (a) the perfect truncated (AB) PC, (b) the multiple photonic quantum well structures made of nonlinear material, i.e., $[(AB)_4C]_{80}(AB)_4$.

of the forward and backward waves can be tailored.

We first set $\eta_1^0=0$ and $\eta_2^0=1$, which corresponds to the sample producing dominant backward SHG. The obtained conversion efficiencies are $\eta_{\mathrm{forth}}=3.04\times 10^{-4}$ and $\eta_{\mathrm{back}}=2.20\times 10^{-2}$. It is apparent that the η_{back} is 72 times that of η_{forth}. When setting $\eta_1^0=1$ and $\eta_2^0=0$, we can construct a sample for implementing dominant forward SHG. The calculated conversion efficiencies are $\eta_{\mathrm{forth}}=4.18\times 10^{-2}$ and $\eta_{\mathrm{back}}=1.11\times 10^{-3}$. We also can obtain optimal structure for performing nearly identical conversion efficiency of both the forward and backward SHGs. For instance, by changing the values of η_1^0 and η_2^0, we can design the sample with $\eta_{\mathrm{forth}}=2.56\times 10^{-2}$ and $\eta_{\mathrm{back}}=2.58\times 10^{-2}$. Note that in this configuration, the conversion efficiency is enhanced two orders of magnitude, compared to the prefect optical superlattice made by the poled $LiNbO_3$ crystal of 80 layers of alternatively inverting poled ferroelectric domains and each layer thickness is 3.428 μm for matching the quantum-phase-matching (QPM) condition.

In summary, we present a general solution of SHG in a 1D inhomogeneous system and apply it to evaluate the SHG conversion efficiency in the multiple PQWs structures made of nonlinear material. We find that the conversion efficiency of SHG can be greatly enhanced when the FW frequency is assigned to one of the defect states, and both forward and backward SHGs can be changed by appropriately adjusting the polarization configuration of the ferroelectric domains in sample.

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