Investigating the Second Harmonic Generation in One-Dimensional Multi-layer Structure Consisting of Polar Ferroelectric Called LiTaO₃ Using the Transfer Matrix Method

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ABSTRACT:

The transfer matrix method is widely used to calculate the electromagnetic waves dispersion. In this paper, we have investigated the second harmonic generation in a one-dimensional structure including LiTaO₃ ferroelectric. Using the transfer matrix method, we obtain the second harmonic efficiency. In this article, in the structure of nonlinear photonic crystals, the linear and nonlinear optical parameters are changed periodically, which causes the second harmonic conversion efficiency to be several times greater than the conversion efficiency of a nonlinear structure based on conventional quasi-phase matching (QPM). The reason for this is that in the system discussed in this paper, the presence of effects of band gap edges causes the density of states of the electromagnetic fields is increased, the group speed of the electromagnetic waves is decreased (as a result, the interaction time is increased) and local field is boosted. As a result of these three factors, the nonlinear optical interaction is effectively strengthened.

Key words:

second harmonic generation, photonic band gap, conversion efficiency, quasi-phase matching conditions

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1. Introduction

In recent years, amplification and increase of second harmonic generation (SHG) efficiency based on quasi-phase matching conditions (QPM) have been investigated in nonlinear photon crystals (NPCs). In order to establish the quasi phase matching conditions in periodic structures (photonic crystals), it is used to change the nonlinear polarization sign in the layers. In this method, the length of the layers should be correspond to the coherence length of the electromagnetic wave in the desired layer [1]. The method of changing the nonlinear polarization sign in the layers is a suitable way for increasing conversion efficiency of the second harmonic [2].

The increase and amplification by several orders of magnitude of second harmonic generation efficiency in nonlinear photonic crystals has been investigated in ref [3, 4]. This increase and enhancement is due to the high density of resonant modes and the low speed of electromagnetic waves at frequencies near the photonic band gap (PBG) edge [3]. These factors are the reason for better enhancement and limitation of field (that means, for a certain range of radiation electromagnetic frequencies, the conversion efficiency is high) [5].

In recent decades, ferroelectric crystals with periodic polarization have been considered due to the unique properties that have been used to compensating for the phase mismatch in nonlinear optical interaction processes. Second-order nonlinear optical interaction, for example, second harmonic generation (SHG) and the optical parametric oscillator, has been investigated in structures composed of periodic polarized ferroelectric such as $LiNbO_3$, $KTiOPO_4$ and SBN ($Sr_xBa_{1-x}Nb_2O_6$ $0.32 \leq x \leq 0.82$) [6-10]. However, it's always difficult to achieve high-power nonlinear radiation, because there are always optical losses.

In recent years, increasing the nonlinear conversion efficiency in ferroelectric crystals with periodic polarization has been a long term objective. Lately, an increase by several orders of magnitude of second-order nonlinear conversion efficiency in these photonic crystals has been reported in [11]. This increase, as we have said, is related to the high density of resonance modes and the low velocity of electromagnetic waves at frequencies near the photonic band gap edge.

In addition, the theoretical analysis of the second harmonic generation has been investigated in multilayered structures in [12]. However, the impact of the band gap edge (high density of resonant modes and low velocity of electromagnetic waves) has not been considered in ferroelectric structures composed of $LiTaO_3$ with periodic polarization. In this paper, we consider the second harmonic generation (SHG) in nonlinear crystals including $LiTaO_3$ with periodic polarization with different refractive indices using the transfer matrix method.

In this paper, we use the transfer matrix method in order to investigate the SHG problem in nonlinear multi-layered systems in which fundamental plane-wave beams incident vertically to the surface of the structure, and so that both linear and nonlinear properties alternate throughout the system. It is assumed that nonlinear optical processes are weak so that the fundamental pump waves are not necessarily affected by the nonlinear process. Using this method, we calculate not only the second harmonic wave energy in the output of structure, but also the distribution of the pump and the second harmonic fields within the system. In our method, there is no approximation except for the non-depleted pump wave approximation. The results of this paper are different from the results of the works in which the systems is based only on the traditional quasi-phase matching (QPM). We will discuss the cause of this difference in the following.

This paper is organized as follows in sec.2 we focus on the details of the model and we derive the related transfer matrix. We also explain how we calculate the conversion efficiency. In sec.3 we present and discuss our numerical results. Finally, sec. 4 ends the paper with a summary of our results.

2.MODEL AND METHOD

The structure we use in this article is $(P_+P_-)^N$. $\chi_+^{(2)}$, d_+ and $n_+^{(l)}$ are, respectively, second order susceptibility, thickness and refractive index for layer P_+ (odd layer), These parameters for the P_- layer (even layer) are $\chi_-^{(2)} = -\chi_+^{(2)}$, d_- and $n_-^{(l)}$ respectively, Fig. 1. Where $l = I$ (for the fundamental waves [FW]) and $l = II$ (for second harmonic waves [SHW]). Note that the periodicity of the structure is equal to $d = d_+ + d_-$.

Fig. 1. Multi-layered nonlinear structure.

It is assumed that the common boundary of the layers is placed on the xy plane. The incoming light shines perpendicular to the surface of the device. The fundamental and second harmonic wave electric fields follow the following relationships [13]:

$$\left[\frac{d^2}{dz^2} + (k_p^{(I)})^2 \right] E_p^{(I)}(z) = 0 \quad (1)$$

$$\left[\frac{d^2}{dz^2} + (k_p^{(II)})^2 \right] E_p^{(II)}(z) = -k_0^{(II)2} \chi_p^{(2)} E_p^{(I)2}(z) \quad (2)$$

Where $k_p^{(I)} = n_p^{(I)} k_0^{(I)}$, $k_p^{(II)} = n_p^{(II)} k_0^{(II)}$, $k_0^{(I)} = \omega/c$, and $k_0^{(II)} = 2\omega/c$. and also, $n_p^{(I)}$ ($n_p^{(II)}$) show the refractive index at the FW (SHW) frequency, respectively in the p th layer (For odd layers, it becomes + for even layers, it becomes -), c is the light speed in vacuum. Using Equation (1), we have for the initial electric field:

$$E_p^{(I)}(t) = E_p^{(I)+} e^{i(k_p^{(I)}(z-z_{p-1})-\omega t)} + E_p^{(I)-} e^{-i(k_p^{(I)}(z-z_{p-1})-\omega t)} \quad (3)$$

Where z_0 is set to zero, $z_p = z_{p-1} + d_p$, and d_p is the thickness of the p th layer (For odd layers, it becomes + for even layers, it becomes -). $E_p^{(I)+}$ and $E_p^{(I)-}$ respectively, represent the magnitudes of the forward and backward primary field waves at the interface. Using boundary continuity conditions we have:

$$\begin{pmatrix} E_p^{(I)+} \\ E_p^{(I)-} \end{pmatrix} = t_p^{(1)} \begin{pmatrix} E_{p-1}^{(I)+} \\ E_{p-1}^{(I)-} \end{pmatrix} \quad (4)$$

Where $t_p^{(1)}$ is the transition matrix for the p th layer, and is calculated as follows:

$$t_p^{(1)} = D_0^{-1} D_p P_p D_p^{-1} D_0 \quad (5)$$

The related matrices are as follows:

$$D_0 = \begin{pmatrix} 1 & 1 \\ n_0 & -n_0 \end{pmatrix}, D_p = \begin{pmatrix} 1 & 1 \\ n_p^{(I)} & -n_p^{(I)} \end{pmatrix}, P_p = \begin{pmatrix} \exp(ik_p^{(I)} d_p) & 0 \\ 0 & \exp(-ik_p^{(I)} d_p) \end{pmatrix}$$

Where n_0 is the refractive index of the air. Transfer matrix of the whole system is obtained by multiplying the sequence of these transfer matrices, and connects the input and output fields of the system:

$$\begin{pmatrix} E_T^{(I)} \\ 0 \end{pmatrix} = T \begin{pmatrix} E_0 \\ E_R^{(I)} \end{pmatrix} \quad (6)$$

$$E_R^{(I)} = -(T_{21}/T_{22})E_0, E_T^{(I)} = T_{11}E_0 + T_{12}E_R^{(I)},$$

Using the equation 6, we can calculate the amplitude of the electric fields ($E_p^{(I)\pm}$) in each layer. After determining the amplitudes of the fundamental electric field in the structure, we examine the second harmonic generation. Putting Equation 3 into Equation 2, we have:

$$E_p^{(II)}(z) = E_p^{(II)+} e^{ik_p^{(II)}(z-z_{p-1})} + E_p^{(II)-} e^{-ik_p^{(II)}(z-z_{p-1})} + A_p^{(II)+} e^{i2k_p^{(II)}(z-z_{p-1})} + A_p^{(II)-} e^{-i2k_p^{(II)}(z-z_{p-1})} + C_p^{(II)} \quad (7)$$

Where

$$A_p^{(II)+} = -k_0^{(II)2} \chi_p^{(II)} [E_p^{(I)+}]^2 / (k_p^{(II)2} - 4k_p^{(I)2}),$$

$$A_p^{(II)-} = -k_0^{(II)2} \chi_p^{(II)} [E_p^{(I)-}]^2 / (k_p^{(II)2} - 4k_p^{(I)2})$$

$$C_p^{(II)} = -2k_0^{(II)2} \chi_p^{(II)} E_p^{(I)+} E_p^{(I)-} / k_p^{(II)2}$$

Using the Maxwell equation $\nabla \times E_p^{(II)}(z) = ik_0^{(II)2} H_p^{(II)}(z)$, the magnetic field $\mathbf{H}$ is as follows:

$$H_p^{(II)}(z) = n_p^{(II)} [E_p^{(II)+} e^{ik_p^{(II)}(z-z_{p-1})} - E_p^{(II)-} e^{-ik_p^{(II)}(z-z_{p-1})}] + n_p^{(I)} [A_p^{(II)+} e^{i2k_p^{(II)}(z-z_{p-1})} - A_p^{(II)-} e^{-i2k_p^{(II)}(z-z_{p-1})}] \quad (8)$$

Using the continuous conditions of the electric and magnetic fields at the boundaries of the p th layer ($z = z_p$ and $z = z_{p-1}$), we have:

$$\begin{pmatrix} E_p^{(II)+} \\ E_p^{(II)-} \end{pmatrix} = t_p^{(II)} \begin{pmatrix} E_{p-1}^{(II)+} \\ E_{p-1}^{(II)-} \end{pmatrix} + \begin{pmatrix} r_p^+ \\ r_p^- \end{pmatrix} \quad (9)$$

The matrix $t_p^{(II)}$ is given below:

$$t_p^{(II)} = G_0^{-1} N_p G_0, \quad N_p = G_p Q_p G_p^{-1}$$

$$\begin{pmatrix} r_p^+ \\ r_p^- \end{pmatrix} = G_0^{-1} \left[(B_p F_p - N_p B_p) \begin{pmatrix} A_p^{(II)+} \\ A_p^{(II)-} \end{pmatrix} + (1 - N_p) \begin{pmatrix} C_p^{(II)} \\ 0 \end{pmatrix} \right] \quad (10)$$

$$G_0 = \begin{pmatrix} 1 & 1 \\ n_0 & -n_0 \end{pmatrix}, \quad G_p = \begin{pmatrix} 1 & 1 \\ \frac{k_p^{(II)}}{k_0^{(II)}} & -\frac{k_p^{(II)}}{k_0^{(II)}} \end{pmatrix}, \quad Q_p = \begin{pmatrix} \exp(i k_p^{(II)} d_p) & 0 \\ 0 & \exp(-i k_p^{(II)} d_p) \end{pmatrix}$$

$$F_p = \begin{pmatrix} \exp(i 2 k_p^{(I)} d_p) & 0 \\ 0 & \exp(-i 2 k_p^{(I)} d_p) \end{pmatrix}, \quad B_p = \begin{pmatrix} 1 & 1 \\ \frac{2 k_p^{(I)}}{k_0^{(II)}} & -\frac{2 k_p^{(I)}}{k_0^{(II)}} \end{pmatrix}$$

Using Equation (6), we obtain the following recursive relation for the whole structure:

$$\begin{pmatrix} E_N^{(II)+} \\ E_N^{(II)-} \end{pmatrix} = T_N^{(II)} \begin{pmatrix} E_0^{(II)+} \\ E_0^{(II)-} \end{pmatrix} + \begin{pmatrix} R_p^+ \\ R_p^- \end{pmatrix} \quad (11)$$

$$T_N^{(II)} = t_N^{(II)} T_{N-1}^{(II)}, \quad \begin{pmatrix} R_p^+ \\ R_p^- \end{pmatrix} = t_N^{(II)} \begin{pmatrix} R_{p-1}^+ \\ R_{p-1}^- \end{pmatrix} + \begin{pmatrix} r_p^+ \\ r_p^- \end{pmatrix} \quad (12)$$

Using the initial conditions, $E_0^{(II)+} = E_N^{(II)-} = 0$ (since there is no second harmonic field to enter the structure), we calculate the fields $E_0^{(II)-}$ (backward second harmonic wave=The SH wave generated at the beginning of the system that propagates in the opposite direction to z.) and $E_N^{(II)+}$ (forward second harmonic wave=The SH wave generated at the end of the system that propagates in the z direction.). The forward and backward conversion efficiency are calculated as follows:

$$\eta_f = \frac{I_t^{(II)}}{I_{pump}}, \quad \eta_b = \frac{I_r^{(II)}}{I_{pump}} \quad (13)$$

Where $I_{pump} \propto |E_0|^2$, $I_r^{(II)} \propto |E_0^{(II)-}|^2$ and $I_t^{(II)} \propto |E_N^{(II)+}|^2$ are the intensities of the pump, reflected and transmitted SH fields, respectively.

The structure we examine is composed of periodically poled lithium tantalite (LiTaO₃) crystal with a nonlinear coefficient $\chi^{(2)} = 13.8$ pm/V. The refractive index of this ferroelectric is obtained from the following formula [14]:

$$n(\lambda, T) = \sqrt{U + \frac{V + v(T)}{\lambda^2 - [W + w(T)]^2} + \frac{Y}{\lambda^2 - Z^2} + X\lambda^2} \quad (14)$$

Where $U = 4.5284$, $V = 7.2449 \times 10^{-3}$, $W = 0.2453$, $X = -2.3670 \times 10^{-2}$, $Y = 7.7690 \times 10^{-2}$, $Z = 0.1838$, $v(T) = 2.6794 \times 10^{-8}(T + 273.15)^2$, $w(T) = 1.6234 \times 10^{-8}(T + 273.15)^2$.

In the studied structure, the refractive index of the odd layers is $n_+^{(l)} = n(\lambda^{(l)}) + \Delta n_+^{(l)} = n(\lambda^{(l)}) - \frac{1}{2}(n(\lambda^{(l)}))^3 R_{33} E$ and the even layers is $n_-^{(l)} = n(\lambda^{(l)}) + \Delta n_-^{(l)} = n(\lambda^{(l)}) + \frac{1}{2}(n(\lambda^{(l)}))^3 R_{33} E$. Where $\lambda^{(l)}$ ($l = I$, for the FW waves), $l = II$, for SHW waves), E is the electric field magnitude and R_{33} is the electro-optics factor of the ferroelectric material.

3. NUMERICAL RESULTS

From the mathematical calculations of Sec.II, it is clear that the existence of a strong light scattering effect in the structure of the photonic crystal complicates the analysis of the second harmonic generation problem. It is obvious that the conventional analytical solution is used to the ordinary quasi-phase matching (QPM) problem is no longer applicable. Therefore, numerical simulation is used to establish QPM conditions in the nonlinear multi-layer structure. We do this by adjusting the thickness of the two constituent parts of the structure. Quasi-phase matching conditions and optimal structure occur at the maximum conversion efficiency. In Fig. 2, the frequency conversion efficiency of SH as a function of the thickness of each part (d) is plotted so that the width of the positive region (The positive region is a region in which $\chi^{(2)}$ is positive) is $d_+ = 3000nm$.

Fig. 2. Conversion efficiency of SH wave in terms of thickness of each part, d the peak of conversion efficiency happens when $d = 6158.7nm$ and $d_+ = 3000nm$. The wavelength of the fundamental wave is $\lambda = 1064 nm$.

The structure consists of 35 pairs of layers and the initial wavelength (pump) is 1064 nm. In this wavelength, the refractive indices of the positive and negative regions are $n_+^{(I)} = 1.0846$ and $n_-^{(I)} = 1.8193$, respectively. The intensity of the external electric field is of the order of tens of kV/mm . The refractive indices for the SH wave in the positive and negative regions are $n_+^{(II)} = 1.0387$ and $n_-^{(II)} = 1.8653$, respectively. According to Fig. 2, the maximum conversion efficiency occurs at $d = 6158.7nm$.

As we know, the second harmonic conversion efficiency, in addition to being dependent on the power of the pump light, also strongly depend on intensity of the pump light inside the structure. So, in addition to the QPM conditions, the properties of the photonic band edges in amplifying and increasing the SH conversion efficiency are very effective. Figure 3 shows the SH conversion efficiency as a function of fundamental field (FF) Intensity.

Fig. 3. The SH wave efficiency vs fundamental wave intensity of the pump. The squares represent the calculated values and the red line represents the linear fit of the points. The structure is the same as the structure of Fig. 2.

As we said, the conversion efficiency is defined as the ratio of the second harmonic intensity to the pump intensity. For comparison, the SH conversion efficiency of a uniform lithium tantalite (LiTaO_3) crystal (the crystal with fixed second-order susceptibility coefficient $\chi^{(2)}$) with quasi-matching conditions, which is longitudinally equal to the length of the investigated structure in this paper, as shown in Figure 4. The order of magnitude of the convergence efficiency in Fig. 4 is about 10^{-4} , which is three times smaller than the conversion efficiency in the designed structure (Fig. 3).

Fig. 4. The SH wave efficiency vs fundamental wave intensity of the pump for uniform lithium tantalite (LiTaO_3) crystal. The squares represent the calculated values and the green line represents the linear fit of the points.

The calculated transmission spectrum for $d = 6158.7\text{nm}$ is shown in Fig. 5. By selecting these parameters (refractive index of layers and width of layers) for the nonlinear system, the FF and SH signals are all located at the photonic band gap edges. At the edge of the gap, the density of the electromagnetic waves is large and the group velocity of the wave is small. As a result, the domain of the field increases by several order of magnitude, and the time of nonlinear interaction becomes much larger. The combination of these factors is the main reason for the significant improvement in SH conversion efficiency.

Fig. 5. The transmission spectrum in the vicinity of the pump wavelength (red) and the second harmonic wavelength (blue) when $d = 6158.7\text{nm}$ for the optimal structure that produces the second harmonic waves is shown in Fig.2

In Figure 6, the SHG conversion efficiency is plotted in terms of wavelength in the vicinity of the pump wavelength (1064nm). The photonic crystal structure of Figure 6 is similar to Fig. 5. The upper and lower shapes indicate the intensity of the backward (reflective) and forward (passing) output, respectively. The conversion efficiency values in Fig. 6 are normalized to unity. It is clear from Figure 6 that the desired structure effectively radiates the second harmonic signals in both directions (forward and backward). The intensity of the backward SH signal is large and is approximately equal to the intensity of the forward SH signal. But in a bulk nonlinear optical material with only phase-matching conditions, which is longitudinally equal with the structure studied in this paper, the backward signal is smaller one or more orders of magnitude than the forward signal. The wavelength of the maximum signal SH, in Fig. 6, corresponds to the wavelength of the band gap edge in Figure 5.

Fig. 6. SH signal (normalized) in terms of initial wavelength. The parameters of the system with the parameters of the structure of Figure 5 are the same.

The distribution of the pump field inside the nonlinear system is calculated by the transfer matrix method (TMM) and is shown in Fig. 7. Regarding the shape, due to the interference induced by multiple reflections, we can observe the pump field that oscillates within the structure, the maximum intensity is increased by one or more order of magnitude than the intensity of the collision field (the field at the beginning of the structure).

Fig. 7. Distribution of pump wave intensity within nonlinear structure at wavelength 1064 nm. The intensity of the pump field doubles as compared with the input wave at the beginning of the structure ($z = 0$).

4. Conclusions

In summary, we used the transfer matrix method for analyzing the SHG problem in a multi-layered nonlinear optical device. To obtain the transmission matrix for fundamental and second harmonic waves, multiple reflections and all interference effects are considered. The method used in this paper do not use the approximations used in previous research on nonlinear optical devices based on the conventional QPM. Using the method of this paper, accurate results are obtained. We have investigated the radiation of SH waves on both sides of the nonlinear optical system and obtained the distribution of the harmonic field within the desired structure. For example, we have investigated the second harmonic generation (SHG) in a one-dimensional nonlinear photonic crystal composed of lithium tantalite (LiTaO_3). We have done a comparative analysis of the structure designed in this article and the conventional QPM structure. We found that in the investigated structure, the second harmonic conversion efficiency is several orders of magnitude greater than the conversion efficiency of nonlinear conventional QPM structure. This is while the length of the two structures is equal. The reason for this is that, at wavelengths associated with the band gap edges, the density of electromagnetic modes is large, the group velocity is small and the local fields are strengthened. These three factors give rise to an effective enhancement in the nonlinear optical interaction. In the structure studied in this paper, in addition to the forward SH signal (also exists in previous structures, and propagates in the direction of the pump waves), there is also a backward SH signal that propagates in the opposite direction of the pump waves.