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Enhanced third harmonic generation in periodic structures

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Abstract. We consider third harmonic generation in a periodic layered medium with alternate nonlinear media. We show enhanced third harmonic generation when the fundamental frequency matches one of the mode frequencies of the distributed feedback structure. The observed feature is explained in terms of large local field enhancement for the fundamental wave.

Keywords. Third harmonic; periodic structure.

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1. Introduction

In recent years there has been a lot of interest in harmonic generation in nonlinear periodic structures [1–4]. The theory of harmonic generation in periodic structures with undepleted pump approximation is now well understood. In the context of second harmonic generation, periodic structures have been used to achieve phase matching and quasi-phase matching [4]. There is another interesting aspect of periodic structures which can be exploited to enhance the generated harmonics. It is well-known that the linear transmission characteristics of periodic structures have the so called Bragg reflection bands [5–8]. For finite structures there are sharp resonances at the edge of the Bragg bands. The origin of these resonances are due to the excitation of modes of the periodic structure [8]. Such modes possess high quality factors and their excitation corresponds to large local field enhancements. Resonances and associated local field enhancements [9] have been utilized to lower the threshold for nonlinear processes and increase their efficiency [10]. Surface and guided mode resonances and resonances of Fabry–Perot cavities have been used for nonlinear effects like optical bistability [11–14]. The sharper the resonance, the larger is the local field enhancement and hence the nonlinear effect is higher [12]. Keeping in mind the aforesaid we study third harmonic generation in a periodic structure with alternate cubic nonlinear layers. We apply the method developed by Bethune [1] to calculate the generated third harmonic in the forward and backward directions. We show that when the fundamental wavelength is resonant with any of the modes of the periodic structure there is considerable enhancement in the generated third harmonic intensity. This is again because of the large local field enhancement in the fundamental intensity which acts as a source for the third harmonic.

The organization of the paper is as follows: In § 2, we briefly recall the method of Bethune [1] for the calculation of third harmonic intensity. In § 3, we present the results of our numerical investigation and finally in § 4, we conclude the paper.

2. Theory

We follow the analysis of Bethune [1] for evaluating the third harmonic intensity from the layered media. The technique is based on modification of the well-known optical transfer matrix method for layered media to include the nonlinear polarization. This formulation has the advantage that the total third harmonic output field is expressed as the sum of contributions from individual layers and thus the effect of any single layer can be studied separately.

Figure 1 shows the geometry of the layered structure with N periods. Each period consists of two layers A (linear) and B (nonlinear), with refractive indices n_A and n_B and thickness d_A and d_B respectively. The z-axis is normal to the layers and x–z is the plane of incidence. First the electric field at the fundamental frequency in each layer is determined. This is required for the third harmonic calculations. The analysis is presented for the s-polarization. The electric field (at ω) in any ith layer is the plane wave solution of the wave equation and can be written as

$$E_i^{\pm}(r,t) = E_i^{\pm}(r)\exp[(\pm iN_i k_0 z) + i(\kappa x - \omega t)], \tag{1}$$

where $k_0(=\omega/c)$ is the vacuum wave vector, $N_i(=k_{iz}/k_0)$ is the propagation constant and $\kappa(=k_x)$ is the x-component of the wave vector.

The forward and backward propagating amplitudes at the left and right side of the layer are related through a 2×2 matrix. This matrix is the product of interface and propagation matrices which are given as

$$G_{ij} = \frac{1}{t_{ij}}\begin{pmatrix} 1 & r_{ij} \\ r_{ij} & 1 \end{pmatrix} \tag{2}$$

and

$$\Psi_i = \begin{pmatrix} \psi_i & 0 \\ 0 & \bar{\psi}_i \end{pmatrix}, \tag{3}$$

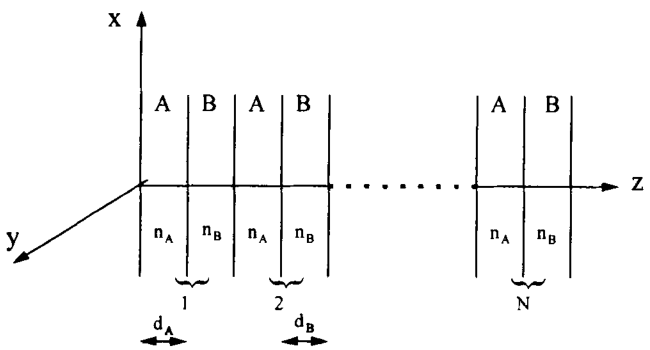

Figure 1. Schematic view of the layered structure.

where r_{ij} and t_{ij} are the reflection and transmission amplitudes for wave incident at $i-j$ interface and $\psi_i \exp(iN_i k_0 d_i)$, $\bar{\psi}_i = \exp(-iN_i k_0 d_i)$. Thus, the electric fields at any layer can be related to those at another layer by successive product of the transfer and propagation matrices. In terms of the interface and propagation matrices of all layers, the total transfer matrix at the fundamental frequency is given as

$$T = G_{f(f-1)} \Psi_{(f-1)} G_{(f-1)(f-2)} \cdots G_{21} \tag{4}$$

and the overall reflection amplitude $r = -T(2,1)/T(2,2)$. We can now write the electric field amplitudes at the left end of each layer as

$$E_i = G_{i(i-1)} \Psi_{(i-1)} G_{(i-1)(i-2)} \cdots G_{21} \begin{pmatrix} 1 \\ r \end{pmatrix}, \tag{5}$$

where

$$E_i = \begin{pmatrix} E_{i+} \\ E_{i-} \end{pmatrix}. \tag{5a}$$

Knowing the pump field in each layer, we can now calculate the nonlinear polarization and the corresponding electric field amplitudes at 3ω. Since we are neglecting the pump depletion, the nonlinear polarization in different layers act as independent sources and the generated third harmonic field will be the sum of contributions from all the nonlinear layers. Let layer j be optically nonlinear. As the pump field is s-polarized, the nonlinear polarization generated and the corresponding third harmonic field will also be s-polarized. Under the pump depletion approximation, the third order nonlinear polarization is written as

$$P_j^{\mathrm{NL}}(3\omega) = \chi^{(3)}[E_j^+(\omega)\exp(iN_j k_0 z) + E_j^-(\omega)\exp(-iN_j k_0 z)]^3, \tag{6}$$

where $\chi^{(3)}$ is the third order susceptibility. The expansion of the RHS of eq. (6) leads to terms with exponential factors like $\exp(\pm iN_j k_0 z)$ and $\exp(\pm 3iN_j k_0 z)$ contributing to the third harmonic. The corresponding effective source dielectric constants can be defined as

$$\varepsilon_s^{(1)} = \left[\left(\frac{N_j}{3}\right)^2 + \kappa^2\right] \quad \text{and} \quad \varepsilon_s^{(3)} = (N_j^2 + \kappa^2) = \varepsilon_j(3\omega). \tag{7}$$

The total electric field (at 3ω) will be the sum of free waves (solution of homogeneous part of third harmonic wave equation) and bound waves (solution of inhomogeneous wave equation)

$$E_{sj}(3\omega) = E_j^+(3\omega)\exp(3iN_{bj}k_0 z) + E_j^-(3\omega)\exp(-3iN_{bj}k_0 z) + \frac{4\pi}{\varepsilon_s - \varepsilon_j(3\omega)} P_j^{\mathrm{NL}}, \tag{8}$$

where $N_{bj} = N_j(3\omega)$, E^+ and E^- are the forward and backward propagating amplitudes at 3ω. In every nonlinear layer there will be contribution from each of the source dielectric constants $\varepsilon_s^{(1)}$ and $\varepsilon_s^{(3)}$ to the third harmonic electric field.

The continuity of the total (bound plus free) tangential 3ω electric and magnetic fields at the $i-j$ and $j-k$ interfaces leads to

$$E_i = M_{ij}E_j + M_{is}E_s, \tag{9}$$

$$M_{kj}\Phi_j E_j + M_{ks}\Phi_s E_s = E_k, \tag{10}$$

where E_j is given by (5a) with amplitudes at 3ω, M and Φ are the interface and propagation matrices at 3ω. Using (9) and (10) we can define source vector for the jth layer as

$$S_j \equiv (\Phi_j M_{js}\Phi_s - M_{js})E_s. \tag{11}$$

In the s-subscripted matrices appropriate ε_s must be used. Now we can write the final reflected and transmitted 3ω field amplitudes $E_1^-(j)$ and $E_f^+(j)$ from the jth layer as

$$\begin{bmatrix} E_f^+(j) \\ E_1^-(j) \end{bmatrix} = \frac{1}{\tilde{T}(1,1)} \begin{pmatrix} 1 & 0 \\ \tilde{T}(2,1) & -\tilde{T}(1,1) \end{pmatrix} S_j', \tag{12}$$

where $\tilde{T} = L_{j1}^{-1}R_{jf}$ is the total right to left transfer matrix at 3ω, $S_j' = L_{j1}^{-1}S_j$, $R_{jf} = \Phi_j M_{jk} \cdots \Phi_{(f-1)} M_{(f-1)f}$, $L_{j1}^{-1} = \bar{\Phi}_1 M_{12} \cdots \bar{\Phi}_{(j-1)} M_{(j-1)j}$ and $\bar{\Phi} = \Phi^{-1}$. The total output fields can be obtained by adding the contributions from all values of $|k_{sz}|$ from all the layers.

3. Numerical results and discussion

In this section we present the results of numerical investigation for a periodic medium with alternate nonlinear layers. Silica glass was chosen as the linear medium whereas, we chose CS_2 (with $\chi^{(3)} \approx 6{\cdot}8 \times 10^{-13}$ esu) with linear refractive index $n_B = 1{\cdot}59$ as the nonlinear material. We incorporated dispersion of glass in our calculation. The dispersion data was taken from Palik [15] and they were fitted to yield

$$n_A(\lambda) = 1{\cdot}4507 - 0{\cdot}0032667\lambda^2 + \frac{0{\cdot}002998}{(\lambda^2 - 0{\cdot}0099)}. \tag{13}$$

The rms error in fitting was of the order of 10^{-11}. Due to lack of dispersion data on CS_2 we were forced to assume it to be dispersionless. However, with available data dispersion in CS_2 can trivially be incorporated. In all our calculations we used the same width of the linear layers, namely, $d_A = 0{\cdot}8\ \mu m$. We assume the pump laser to be Nd–YAG operating at the wavelength $1{\cdot}064\ \mu m$. For all our calculations we have taken a periodic system with $N = 100$ periods. It is well known that by varying the structural parameters (in our case d_B since we already fixed d_A) it is possible to make the fundamental wavelength to coincide with the center of the Bragg reflection band or lie at the edge of the Bragg band. For a finite periodic structure the edge of the Bragg band contains sharp resonances characterized by high quality factors [8]. It is well understood that such resonances are associated with large local field enhancements which is instrumental in generating large nonlinear effects. Such sharp resonances in the context of surface/guided modes were exploited to lower the threshold for various nonlinear effects [10–14]. We thus change d_B to make the pump wavelength coincide with one of these sharp resonances at the edge of the Bragg band. For example, for $d_B = 0{\cdot}944\ \mu m$,

$\lambda = 1{\cdot}064\,\mu m$ lies at the center of the rejection band, while for $d_B = 0{\cdot}937\,\mu m$ the fundamental wavelength is resonant with one of the high-Q modes of the periodic structure. We also look at the case when the fundamental wavelength is away from the Bragg bands, say, midway between two Bragg bands. In what follows we present the results for these three cases.

The results for $d_B = 0{\cdot}944\,\mu m$ (pump wavelength $1{\cdot}064\,\mu m$ coincides with the center of the rejection band) are shown in figure 2. In figure 2a we have plotted fundamental transmission coefficient T as a function of the wavelength λ. We have shown the exponential decay of fundamental intensity inside the layers at $\lambda = 1{\cdot}064$ in figure 2b. This is obvious since transmission at $\lambda = 1{\cdot}064\,\mu m$ is minimal (see figure 2a). Since fundamental field distribution (which is almost null over most of the structure) acts as the source for the third harmonic, one expects very little generated third harmonic in this case. This is shown in figure 2c where we have plotted third harmonic intensity generated in the forward and backward directions $T(3\omega)$ (solid curve) and $R(3\omega)$ (dotted curve) as functions of λ. We have multiplied the intensities by the nonlinear susceptibility $\chi^{(3)}$ in order to make $T(3\omega)$ and $R(3\omega)$ dimensionless.

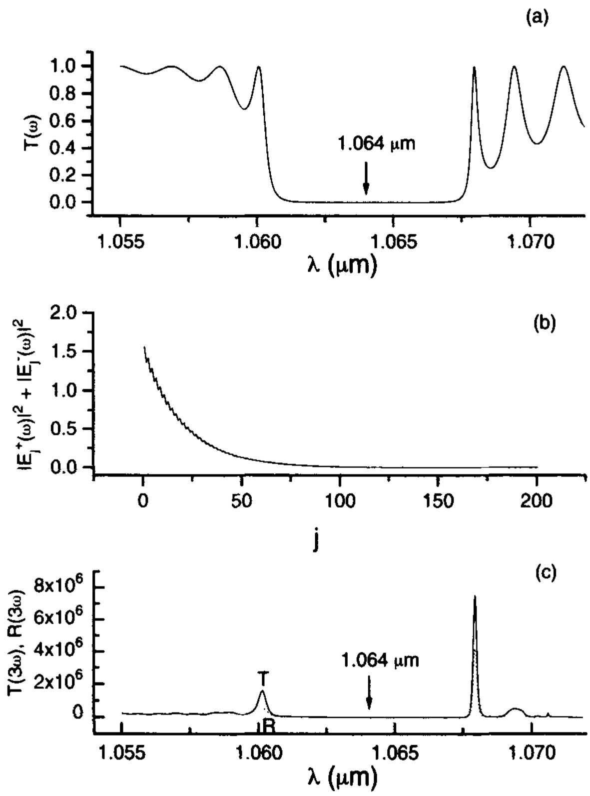

Figure 2. (**a**) Transmission coefficient $T(\omega)$ as a function of λ with $d_A = 0{\cdot}8\,\mu m$, $d_B = 0{\cdot}944\,\mu m$, $n_A = 1{\cdot}59$, n_B is calculated using eq. (13) given in the text, $n_i = n_f = 1{\cdot}0$. (**b**) Plot of fundamental intensity (in arbitrary units) inside the layers at pump wavelength $\lambda = 1{\cdot}064\,\mu m$. (**c**) Third harmonic intensity in the forward (solid) and backward (dotted curve) directions as a function of λ.

The situation is different when d_B is changed to $d_B = 0{\cdot}937\,\mu m$ so as to make the pump wavelength coincide with one of the mode frequencies of the periodic structure. The results for fundamental transmission coefficient as a function of λ for $d_B = 0{\cdot}937\,\mu m$ is shown in figure 3a. Different resonances corresponding to the different modes of the structure are labeled by ± 1, ± 2, etc. The $\pm 1 (\pm 2)$ resonances correspond to single (double) hump field distributions in the layered medium (see figure 3b). Also note that the $+1$ resonance corresponds to the largest enhancements in the local field. Thus if the fundamental wavelength is made to coincide with the $+1$ mode frequency (which is the case for $d_B = 0{\cdot}937\,\mu m$) one expects a large enhancement in the generated third harmonic. This is shown in figure 3c. It is also clear from figure 3c that the generated third harmonic decreases from $+1$ to -1 and from $+2$ to -2 mode resonances. This is explained in terms of the field distributions in figure 3b where the local field enhancement is maximum for the $+1$ mode and minimum for the -2 mode. It is thus clear that by making the fundamental laser resonant with the high quality factor modes of the periodic structure it is possible to enhance the generated third harmonic.

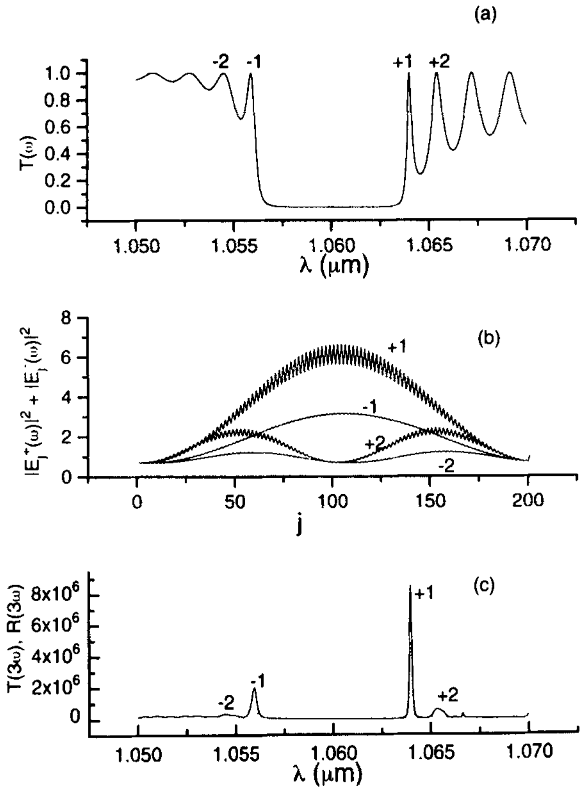

Figure 3. (**a**) Transmission coefficient $T(\omega)$ as a function of λ with $d_B = 0{\cdot}937\,\mu m$, other parameters are same as in figure 2. (**b**) Fundamental intensity (in arbitrary units) distribution when its wavelength coincides with different mode resonances. Different curves are labeled by the mode numbers ± 1 and ± 2. For example curve labeled by $+1$ is for $\lambda = 1{\cdot}064\,\mu m$. (**c**) Third harmonic generated in the forward (solid) and backward (dotted curve) directions as a function of λ.

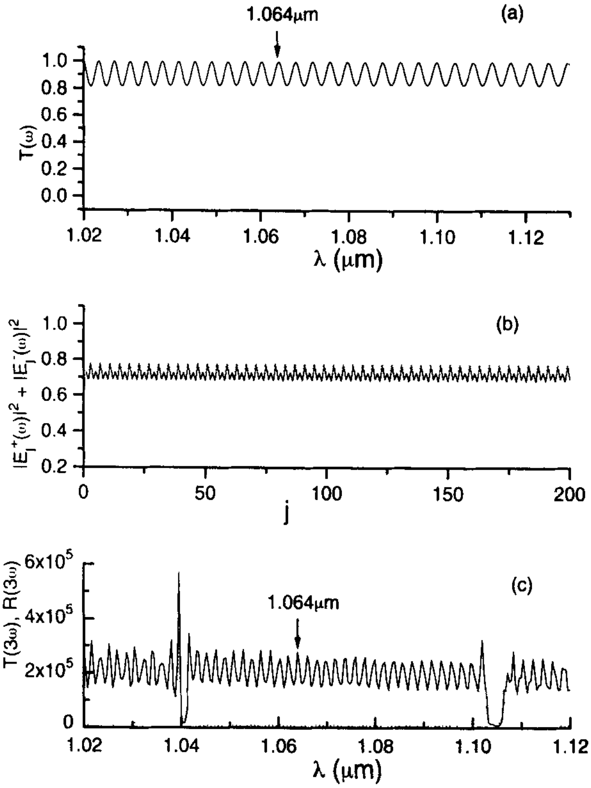

Figure 4. (**a**) Variation of transmission coefficient $T(\omega)$ with λ for $d_B = 1{\cdot}11$ μm, $n_A = 1{\cdot}59$, other parameters are the same as in figure 2. (**b**) Plot of fundamental intensity (in arbitrary units) inside the layers at pump wavelength $\lambda = 1{\cdot}064$ μm. (**c**) Plot of third harmonic intensity in the forward (solid) and backward (dotted curve) directions as a function of λ.

In order to complete the above discussion, we present the results for $d_B = 1{\cdot}11$ μm, when the pump laser wavelength 1·064 μm is midway between two Bragg reflection bands. The results for $T(\omega)$ and the field distribution are shown in figures 4a and 4b respectively. It is clear from figure 4b that the distribution lacks any local enhancements and hence the generated third harmonic (figure 4c) is lower, by an order of magnitude, than the one with enhancement (compare with figure 3c). The gaps close to 1·04 μm and 1·10 μm, which are due to the transmission characteristics of the distributed feedback structures at the third harmonic frequency.

4. Conclusion

In conclusion we studied a periodic layered medium with alternate cubic nonlinear layers for third harmonic generation. Thus one can have enhanced third harmonic generation from the layered medium if the fundamental frequency coincides with one of the mode frequencies of the periodic structure. The high yield of the third harmonic is explained in terms of the local field enhancements associated with the excitation of the modes at the fundamental frequency.

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References

[1] Bethune, *J. Opt. Soc. Am.* **B6**, 910 (1989)
[2] Bethune, *J. Opt. Soc. Am.* **B8**, 367 (1990)
[3] N Hashizume, M Ohashi, T Kondo and R Ito, *J. Opt. Soc. Am.* **B12**, 1894 (1995)
[4] G I Stegeman and C T Seaton, *J. Appl. Phys.* **58**, R57 (1985)
[5] P Yeh, *Optical waves in layered media* (New York, Wiley, 1988) pp. 118
[6] A Yariv and P Yeh, *Optical waves in crystals* (New York, Wiley, 1984) pp. 155
[7] P Yeh, *J. Opt. Soc. Am.* **69**, 742 (1979)
[8] S Dutta Gupta and G S Agarwal, *Opt. Commun.* **103**, 122 (1993)
[9] D Sarid, *Phys. Rev. Lett.* **47**, 1927 (1981)
[10] See, for example, S Sarid, R T Deck, A E Crag, R K Hickernell, R S Jameson and J J Fasano, *Appl. Opt.* **21**, 3993 (1982)
G S Agarwal, *Phys. Rev.* **B31**, 3534 (1985)
[11] G S Agarwal and S Dutta Gupta, *Phys. Rev.* **B34**, 5239 (1986)
[12] S Dutta Gupta and G S Agarwal, *J. Opt. Soc. Am.* **B4**, 691 (1987)
[13] M B Pande and S Dutta Gupta, *Opt. Lett.* **15**, 944 (1990)
[14] S Dutta Gupta, *J. Opt. Soc. Am.* **B10**, 1927 (1989)
[15] E D Palik, *Handbook of optical constants of solids* (Academic Press, New York, 1985) pp. 749