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Third-Order Nonlinearity Enhancement in One-Dimensional Photonic Crystal Structures

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Enhancement of the effective third-order optical nonlinearity of a one-dimensional photonic crystal structure with a nonlinear defect layer is discussed by defining the figure of merit of the system. The nonlinear optical material is doped in the central defect layer of the photonic crystal structure, and it is assumed that the concentration of the nonlinear material can be controlled. It was found that the achievable figure of merit is proportional to the square of the field enhancement in the defect layer. The relationship between the figure of merit and the response time of the photonic-crystal system was obtained, and shows a trade-off between fast response and large nonlinearity. [DOI: 10.1143/JJAP.41.1349]

KEYWORDS: photonic crystal, nonlinear optics, third order, figure of merit, response time

1. Introduction

The interaction between matter and radiation can be modified significantly by using photonic crystal (PC) structures. In particular the nonlinear interaction can be greatly enhanced by the use of defect modes in a one-dimensional (1D) dielectric PC structure.[1–7] In this case, modes of light are modified by the PC structure, and the mode density is enhanced at a certain spectral range, leading to the enhancement of the interaction between matter and radiation. In our previous paper we analyzed the third-order nonlinearity of a 1D PC structure with a defect containing nonlinear optical material,[1] where general expressions for intensity-dependent transmittance and four-wave mixing intensity were derived and optical bistability was also predicted in the PC system.

In this report, we first define the figure of merit of the effective third-order nonlinearity in the PC system based on the previous model.[1] The figure of merit of third-order optical nonlinearity is often used to characterize the efficiency of nonlinear optical materials, which is defined by the third-order susceptibility divided by the absorption coefficient. Since the nonlinearity in our model is purely absorptive, the figure of merit enables us to determine the effectiveness of using the PC structure, and to discuss the dependence of the nonlinearity enhancement on various parameters. Next, we derive the temporal response of the PC system. It is shown that there is a trade-off between the effective nonlinearity and the response time.

2. Model

We assume the structure of the 1D PC with a defect layer to be that depicted in Fig. 1.[1] The structure is symmetrical, and the defect layer is sandwiched between two identical stacks of layers of high and low refractive index on a substrate. Each stack is composed of N layers of low refractive index, A layers, and N layers of high refractive index, B layers. The refractive indices of the A and B layers are denoted n_A and n_B, respectively. The optical thicknesses of the A and B layers are $\lambda/4$, where λ is the incident light wavelength. The optical thickness of the defect layer is $\lambda/2$. Nonlinear optical materials, such as semiconductor quantum dots and dyes, are assumed to be doped in the defect layer, and the material is assumed to have a purely absorptive nonlinearity. The refractive index and the extinction coefficient of the defect layer are denoted n_X and κ, respectively. The nonlinearity is introduced by the absorption saturation

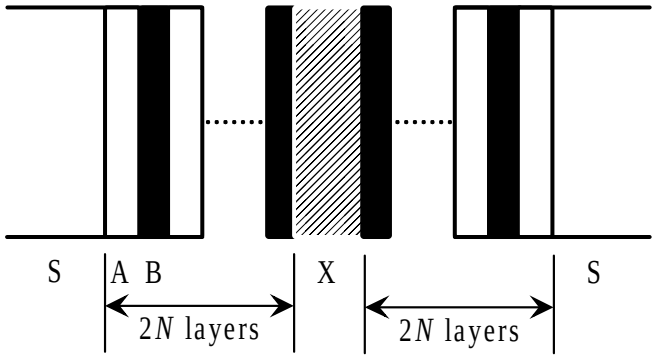

Fig. 1. Model structure of the one-dimensional photonic crystal with a defect layer at the center. S: substrate, A: low-index layer, B: high-index layer, X: defect layer.

$$\kappa = \frac{\kappa_0}{1 + I/I_S}, \tag{1}$$

or

$$\kappa = \kappa_0(1 - I/I_S), \tag{2}$$

to the lowest order of the light intensity. Here, κ_0, I, and I_S are the unsaturated extinction coefficient, the light intensity in the defect layer, and the saturation intensity. The nonlinearity assumed here is third order.

We can briefly summarize some important results reported in the previous paper as follows.[1] We assume here that the refractive index of the substrate and that of the defect layer are equal, and

$$\left(\frac{n_B}{n_A}\right)^N \gg 1. \tag{3}$$

This structure has a narrow transmission peak at λ. The transmittance, T, and the spatially averaged intensity enhancement factor, G, at λ are expressed as

$$T = 4\left[2 + \frac{\pi\kappa}{n_X}\left(\frac{n_B}{n_A}\right)^{2N}\right]^{-2}, \tag{4}$$

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$$G = \frac{T}{2}\left(\frac{n_B}{n_A}\right)^{2N}. \quad (5)$$

The effective nonlinearity of the PC structure was compared with that of a naked nonlinear optical layer without the PC structure. As a measure of the effective magnitude of the nonlinearity, the following quantity was used:

$$\Delta \equiv \frac{dT}{dI_{in}} I_S. \quad (6)$$

Here, I_{in} is the incident light intensity. This quantity is proportional to the effective third-order nonlinearity of the device normalized to the figure of merit of the doped nonlinear material. For the PC structure and a naked film, they can be expressed as a function of the linear transmittance as

$$\Delta_{PC} = \frac{3}{2}\left(\frac{n_B}{n_A}\right)^{2N} T^2\left(1-\sqrt{T}\right), \quad (7)$$

$$\Delta_{naked} = T(1-T). \quad (8)$$

The derivation of eq. (7) is briefly described in the Appendix. These expressions show that the effective nonlinearity is optimized at a certain value of transmittance, or at a certain value of doping concentration of the nonlinear material. The effective nonlinearity of the PC structure has a maximum value of $\Delta_{PC} = 0.12(n_B/n_A)^{2N}$ at $T = 0.64$. An important characteristic of the doped nonlinear optical system is that the doping concentration can be changed easily, and optimized to obtain the optimum effective nonlinearity, which is in sharp contrast to the case of bulk nonlinear materials. Using eq. (4), the optimum doping concentration is obtained to be the concentration corresponding to the extinction coefficient of

$$\kappa = \frac{n_X}{2\pi}\left(\frac{n_A}{n_B}\right)^{2N}. \quad (9)$$

For a large value of N, this can be so small that negligible absorption is obtained in the system without the PC structure.

Comparison of the effective nonlinearity of the PC system with that of the naked film at a fixed doping concentration is not meaningful since the doping level can be optimized independently in each structure. Instead, comparison should be made for the optimized doping concentration in each structure, or at a fixed value of linear transmittance.[1)] The enhancement of the effective nonlinearity over that of a naked film is $0.53(n_B/n_A)^{2N}$ at the transmittance value mentioned above. Another approach for the comparison is to define a figure of merit as a measure of the magnitude of the effective nonlinearity.

3. Figure of Merit

Now, we derive the figure of merit of the effective third-order nonlinearity of the 1D PC structure. The figure of merit of third-order nonlinear optical materials is usually defined by

$$F \equiv \frac{\chi^{(3)}}{\alpha}, \quad (10)$$

where $\chi^{(3)}$ and α are the third-order nonlinear susceptibility and the linear absorption coefficient of the system, respectively.

The susceptibility of the system can be expressed as a function of the light field amplitude as[8)]

$$\chi = \chi^{(1)} + \frac{3}{4}\chi^{(3)}|E|^2, \quad (11)$$

where $\chi^{(1)}$ is the linear susceptibility, and the light field amplitude E is related to the light intensity I by

$$I = \frac{1}{2}\epsilon_0 cn|E|^2. \quad (12)$$

Here, ϵ_0 is the dielectric constant of the vacuum, c is the speed of light, and n is the refractive index of the medium. The imaginary part of the susceptibility of the nonlinear medium, which is proportional to the extinction coefficient, is expressed as

$$\Im\chi = \Im\chi^{(1)}\left(1 - \frac{I}{I_S}\right), \quad (13)$$

in the same manner as in eq. (2). Using eqs. (11–13), we can obtain the relationship between the imaginary part of $\chi^{(3)}$ and the saturation intensity as

$$\Im\chi^{(3)} = -\frac{2\epsilon_0 cn}{3}\frac{1}{I_S}\Im\chi^{(1)}. \quad (14)$$

The third-order susceptibility only has an imaginary part in the present system because the nonlinearity is assumed to be purely absorptive. Since $\Im\chi^{(1)}$ is related to the linear absorption coefficient by

$$\alpha = \frac{2\pi}{n\lambda}\Im\chi^{(1)}, \quad (15)$$

we obtain the figure of merit expressed in terms of the saturation intensity as

$$\frac{\Im\chi^{(3)}}{\alpha} = -\frac{\epsilon_0 cn^2\lambda}{3\pi}\cdot\frac{1}{I_S}. \quad (16)$$

This expression shows that I_S is a measure of the figure of merit of the doped nonlinear material, or, in other words, that of the naked film. The negative sign here indicates that the absorption is decreased when the light intensity is increased. The intensity dependent absorption coefficient is expressed as

$$\alpha(I) = \frac{\alpha}{1 + I/I_S}. \quad (17)$$

Using this and eq. (16), we obtain

$$\frac{d\alpha}{dI} = \frac{3\pi}{\epsilon_0 cn^2\lambda}\Im\chi^{(3)}. \quad (18)$$

Now we proceed to the figure of merit of the *effective* nonlinearity of the PC structure. We introduce here the *effective* absorption coefficient of the device with the PC structure, α_{eff}, by

$$T = \exp(-\alpha_{eff} d). \quad (19)$$

Here, d is the thickness of the nonlinear device. Using a relation equivalent to eq. (18), the imaginary part of the effective third-order susceptibility $\chi^{(3)}_{eff}$ is introduced by

$$\frac{d\alpha_{\mathrm{eff}}}{dI_{\mathrm{in}}} = \frac{3\pi}{\epsilon_0 cn^2\lambda}\,\Im\chi^{(3)}_{\mathrm{eff}}. \tag{20}$$

By differentiating eq. (19) by the incident light intensity, we obtain the expression of the effective nonlinear susceptibility in terms of Δ as defined by eq. (6):

$$\Im\chi^{(3)}_{\mathrm{eff}} = -\frac{\epsilon_0 cn^2\lambda}{3\pi dI_{\mathrm{S}}T}\cdot\Delta. \tag{21}$$

Using eq. (7), the effective nonlinear susceptibility for the PC structure is obtained as

$$\Im\chi^{(3)}_{\mathrm{eff}} = -\frac{\epsilon_0 cn^2\lambda}{2\pi dI_{\mathrm{S}}}\left(\frac{n_{\mathrm{B}}}{n_{\mathrm{A}}}\right)^{2N} T(1-\sqrt{T}). \tag{22}$$

In order to compare the figure of merit of the PC structure with that of the naked film, the limit of $T \to 1$, or small absorption, is taken, which leads to the expression of the effective figure of merit as

$$\frac{\Im\chi^{(3)}_{\mathrm{eff}}}{\alpha_{\mathrm{eff}}} = -\frac{\epsilon_0 cn^2\lambda}{4\pi}\left(\frac{n_{\mathrm{B}}}{n_{\mathrm{A}}}\right)^{2N}\cdot\frac{1}{I_{\mathrm{S}}}. \tag{23}$$

By comparing eqs. (16) and (23), the enhancement of the effective figure of merit of the PC structure over that of the naked film is obtained as

$$\mathcal{F} = \frac{3}{4}\left(\frac{n_{\mathrm{B}}}{n_{\mathrm{A}}}\right)^{2N}. \tag{24}$$

Since the optical field is enhanced by the order of $(n_{\mathrm{B}}/n_{\mathrm{A}})^N$ in the defect layer,[1] the factor above is proportional to the square of the field enhancement. This can be explained as follows. The nonlinear optical process described by the third-order nonlinearity is a four-wave mixing process, where three incident waves are mixed to generate the fourth wave. The radiation and the matter interact with each other four times in this process, leading to the enhancement of the interaction strength which is proportional to the fourth power of the field enhancement. On the other hand, they interact two times in the linear absorption process, leading to the enhancement of the linear absorption which is proportional to the square of the field enhancement, as shown in eq. (4). Thus, the figure of merit of the third-order nonlinearity, which is the magnitude of the third-order susceptibility divided by the linear absorbance, is enhanced by a factor proportional to the square of the field enhancement.

4. Temporal Response

Now we will discuss the factors limiting the enhancement. Equation (24) shows that the figure of merit can be enhanced as much as is desired by choosing a sufficiently large number of layers. In reality, however, any loss of light by intrinsic absorption of the host medium or by randomness can limit the enhancement. From eq. (5), it can be shown that for a given κ the light intensity enhancement factor has a maximum value of

$$G_{\max} = \frac{n_X}{4\pi\kappa} \tag{25}$$

at

$$N = \frac{\ln\dfrac{2n_X}{\pi\kappa}}{2\ln\dfrac{n_{\mathrm{B}}}{n_{\mathrm{A}}}}. \tag{26}$$

This shows that any loss of light in the defect layer can limit the maximum achievable field enhancement.

The number of layers in the stack can be limited by the manufacturing process of the stack. It can also be effectively limited by fluctuation in layer thickness. However, stacks with more than one hundred layers of good optical quality can be manufactured using commercial systems. When assuming typical values of the refractive indices of SiO_2 for A layers ($n_{\mathrm{A}} = 1.46$) and TiO_2 for B layers ($n_{\mathrm{B}} = 2.35$), we obtain $(n_{\mathrm{B}}/n_{\mathrm{A}})^{2N} = 2\times10^{10}$ with $N = 50$. A more realistic limit of the enhancement comes from another factor, the response time of the PC structure.

The finite width of the transmission peak necessarily leads to a finite response time of the PC structure. We derive the time response of the PC structure as follows. The present PC structure with a defect layer at the center can be regarded as a Fabry–Perot interferometer, where the transmittance of the mirrors is

$$\mathcal{T} = 4\left(\frac{n_{\mathrm{A}}}{n_{\mathrm{B}}}\right)^{2N} \tag{27}$$

in the limit of $(n_{\mathrm{B}}/n_{\mathrm{A}})^{2N} \gg 1$. The transmission coefficient of the Fabry-Perot interferometer is[9]

$$t_{\mathrm{e}} = \frac{\mathcal{T}}{1-(1-\mathcal{T})\exp(i\delta)}. \tag{28}$$

Here, δ is the detuning angle:

$$\delta = 2\pi\frac{\nu-\nu_0}{\nu_0}, \tag{29}$$

with ν_0 being the center frequency. When $\mathcal{T} \ll 1$, this expression can be approximated by

$$t_{\mathrm{e}} = \frac{1}{1-i\delta/\mathcal{T}} \tag{30}$$

near the transmission peak. Since the transmittance is

$$T = |t_{\mathrm{e}}|^2 = \frac{1}{1+\delta^2/\mathcal{T}^2}, \tag{31}$$

the transmission peak has a Lorentzian shape and the full-width at half maximum of the peak is

$$\Delta\nu = \frac{\mathcal{T}}{\pi}\nu_0, \tag{32}$$

or

$$\frac{\Delta\nu}{\nu_0} = \frac{4}{\pi}\left(\frac{n_{\mathrm{A}}}{n_{\mathrm{B}}}\right)^{2N}. \tag{33}$$

It is of interest that this factor is inversely proportional to the enhancement factor of the figure of merit of the PC structure as

$$\mathcal{F} = \frac{3}{\pi}\cdot\frac{\nu_0}{\Delta\nu}. \tag{34}$$

Since the incident light always has a finite spectrum width, $\Delta\nu$ should not be narrower than that spectrum width for effective use of the light power. The expression above shows

that the figure of merit is limited by the spectrum purity of the light used in the system.

This relation is of greater importance when the time response of the system is fully considered. The finite spectral width of the transmission peak limits the response time of the PC structure,[3] even if the material response is faster. In other words, in order to obtain a certain response time, the system should have a sufficiently broad transmission peak. The waveform of the transmitted light field of the PC system to a delta function input can be derived by the Fourier transformation of eq. (30) as

$$E_{\mathrm{out}}(t) = \int_{-\infty}^{\infty} t_{\mathrm{e}}(\nu) e^{-i2\pi\nu t} d\nu = \mathcal{T}\nu_0 e^{-\mathcal{T}\nu_0 t} e^{-i2\pi\nu_0 t} \qquad (t > 0). \tag{35}$$

This shows that the output field has a center frequency of ν_0, and decays exponentially with a time constant of $1/\mathcal{T}\nu_0$. Exponential decay of the transmitted field has been observed previously using an interferometric technique.[3,10] When the response time of the system, τ, is defined by the decay time of the light power, it becomes

$$\tau = \frac{1}{2\mathcal{T}\nu_0} = \frac{1}{2\pi\Delta\nu} = \frac{1}{8\nu_0}\left(\frac{n_{\mathrm{B}}}{n_{\mathrm{A}}}\right)^{2N}. \tag{36}$$

By combining eqs. (24) and (36), the enhancement factor of the figure of merit of the PC structure is related to its response time as

$$\mathcal{F} = 6\nu_0\tau. \tag{37}$$

This equation shows that the enhancement of the figure of merit of the third-order optical nonlinearity achievable by using the PC structure is inherently limited by the response time of the system. When one requires a faster response of the system, one obtains smaller nonlinearity. It has already been shown that the magnitude of resonant optical nonlinearity of bulk materials is also limited by the material response time in a similar way.[11] This comes from the material resonance. The enhancement discussed in the present study arises from the modification of the radiation modes by the PC structure. It is of importance that the two mechanisms work independently. Thus, one can utilize both of them to obtain maximum nonlinearity. Optimum nonlinearity can be obtained by tuning the response time of the material and that of the radiation modes at the same time. The resultant effective nonlinearity of the system is that of the raw material multiplied by the enhancement factor of the PC structure. On the other hand, the total response time of the combined system is only limited by the sum of the material response time and that of the PC structure.

5. Conclusion

The magnitude of effective third-order optical nonlinearity of a one-dimensional photonic-crystal structure with doped nonlinear material was discussed by defining the figure of merit of the system. The figure of merit was found to be proportional to the square of the field enhancement in the defect layer. It was also found that the achievable enhancement factor is inversely proportional to the response time of the PC structure.

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Appendix

Here, the derivation of eq. (7) is briefly described.[1] Transmittance of the model PC structure used in the present study can be calculated using the transfer matrix method.[9] For large N such that

$$(n_{\mathrm{B}}/n_{\mathrm{A}})^N \gg 1, \tag{A·1}$$

the transmittance obtained can be approximated as

$$T = \frac{4}{\left[2 + \dfrac{\pi\kappa}{n_X}\left(\dfrac{n_{\mathrm{B}}}{n_{\mathrm{A}}}\right)^{2N}\right]^2}. \tag{A·2}$$

The local light intensity in the defect layer can be obtained for large N by applying the same method as a function of position z as

$$I(z) = \frac{4I_{\mathrm{in}}\left(\dfrac{n_{\mathrm{B}}}{n_{\mathrm{A}}}\right)^{2N}\cos^2(\pi z/d_X)}{\left[2 + \dfrac{\pi\kappa}{n_X}\left(\dfrac{n_{\mathrm{B}}}{n_{\mathrm{A}}}\right)^{2N}\right]^2} \qquad (-d_X/2 < z < d_X/2). \tag{A·3}$$

Here, I_{in} is the light intensity incident on the PC structure and d_X is the thickness of the defect layer. The extinction coefficient of the medium in the defect layer is saturated depending on the position as

$$\kappa(z) = \kappa_0[1 - I(z)/I_{\mathrm{S}}] \tag{A·4}$$

to the lowest order of the light intensity. The effective average extinction coefficient of the entire defect layer to the first order of the incident intensity can be calculated by taking the average with weight of the local light intensity as

$$\bar{\kappa} = \frac{\int_{-d_X/2}^{d_X/2}\kappa(z)I(z)dz}{\int_{-d_X/2}^{d_X/2} I(z)dz} = \kappa_0\left\{1 - \frac{1}{I_{\mathrm{S}}}\cdot\frac{3I_{\mathrm{in}}\left(\dfrac{n_{\mathrm{B}}}{n_{\mathrm{A}}}\right)^{2N}}{\left[2 + \dfrac{\pi\kappa_0}{n_X}\left(\dfrac{n_{\mathrm{B}}}{n_{\mathrm{A}}}\right)^{2N}\right]^2}\right\}. \tag{A·5}$$

By substituting κ in eq. (A·2) by $\bar{\kappa}$, the incident light intensity dependence of the transmittance is obtained as

$$\frac{dT}{dI_{\mathrm{in}}} = \frac{dT}{d\bar{\kappa}}\frac{d\bar{\kappa}}{dI_{\mathrm{in}}} = \frac{1}{I_{\mathrm{S}}}\frac{24\pi\kappa}{n_X}\left(\frac{n_{\mathrm{B}}}{n_{\mathrm{A}}}\right)^{4N}\left[2 + \frac{\pi\kappa}{n_X}\left(\frac{n_{\mathrm{B}}}{n_{\mathrm{A}}}\right)^{2N}\right]^{-5}. \tag{A·6}$$

In the derivation of this expression, κ_0 and $\bar{\kappa}$ are replaced by

κ since in this case we are studying the weak-field response. Using the relationship between κ and T, eq. (A·2), the equation above can be expressed in terms of the transmittance as

$$\frac{dT}{dI_{\mathrm{in}}} = \frac{3}{2}\frac{1}{I_{\mathrm{S}}}\left(\frac{n_{\mathrm{B}}}{n_{\mathrm{A}}}\right)^{2N} T^2(1-\sqrt{T}). \qquad (\mathrm{A}\cdot 7)$$

Finally, eq. (7) is obtained using the definition of Δ in eq. (6).

1) T. Hattori, N. Tsurumachi and H. Nakatsuka: J. Opt. Soc. Am. B **14** (1997) 348.
2) N. Tsurumachi, T. Hattori and H. Nakatsuka: Nonlinear Opt. **22** (1999) 405.
3) N. Tsurumachi, S. Yamashita, N. Muroi, T. Fuji, T. Hattori and H. Nakatsuka: Jpn. J. Appl. Phys. **38** (1999) 6302.
4) N. Tsurumachi, M. Abe, M. Arakawa, T. Yoda, T. Hattori, J. Qi, Y. Masumoto and H. Nakatsuka: Jpn. J. Appl. Phys. **38** (1999) L1400.
5) R. Shimano, S. Inouye, M. Kuwata-Gonokami, T. Nakamura, M. Yamanishi and I. Ogura: Jpn. J. Appl. Phys. **34** (1995) L817.
6) T. Akiyama, M. Tsuchiya and T. Kamiya: Jpn. J. Appl. Phys. **36** (1997) 2701.
7) E. Lidorikis and C. M. Soukoulis: Phys. Rev. E **61** (2000) 5825.
8) P. N. Butcher and D. Cotter: *The Element of Nonlinear Optics* (Cambridge University Press, Cambridge, 1990).
9) M. Born and E. Wolf: *Principles of Optics* (Cambridge University Press, Cambridge, 1999) 7th ed.
10) T. Fuji, M. Arakawa, T. Hattori and H. Nakatsuka: Rev. Sci. Instrum. **69** (1998) 2854.
11) D. H. Auston, T. K. Gustafson, A. E. Kaplan, P. L. Kelly and Y.-R. Shen: Appl. Opt. **26** (1987) 231.