Amplification of the generation of the sum-frequency signal in multilayer periodic structures at the edges of the Bragg band gap

A. V. Balakin, V. A. Bushuev, B. I. Mantsyzov, P. Masselin, I. A. Ozheredov,*) and A. P. Shkurinov
M. V. Lomonosov Moscow State University, 119899 Moscow, Russia

D. Boucher
Laboratoire de PhysicoChimie de l'Atmosphère, Université du Littoral, 59140 Dunkerque, France

(Submitted 20 October 1999)
Pis'ma Zh. Éksp. Teor. Fiz. **70**, No. 11, 718–721 (10 December 1999)

A new property of a one-dimensional periodic structure — amplification of the sum-frequency signal arising under the simultaneous action of two laser pulses on this structure with radiation frequencies corresponding to the edges of the fixed Bragg band gap — is experimentally observed and described. © *1999 American Institute of Physics.* [S0021-3640(99)00323-0]

PACS numbers: 42.70.Qs, 42.65.Ky

1. Investigations of nonlinear-optical phenomena in photonic crystals have been arousing great interest in the last few years.[1] A multilayer periodic structure (MPS) is a particular case of a one-dimensional photonic crystal and is characterized by the existence of regions of forbidden frequencies where total Bragg reflection occurs.[2] In recently published investigations,[3–5] it is shown that a signal at the frequency of the second harmonic can be amplified at the edge of the region of selective Bragg reflection. One possible mechanism of this amplification, examined in Refs. 3 and 4, is due to the linear increase of the energy density of the field at the fundamental frequency.

The subject of the present letter is an investigation of the mechanism of asynchronous amplification of the signal at the sum frequency $\omega_{sf}=\omega_1+\omega_2$ in an MPS. It is established that the efficiency of the generation of the signal at the sum frequency (SF) ω_{sf} increases substantially if the frequencies ω_1 and ω_2 of the two laser pulses incident on the MPS at the same angle are chosen near the opposite edges of a given Bragg band gap. We believe that our investigations show that this asynchronous amplification is largely due to the simultaneous increase of the energy density of the fields in the structure at the frequencies ω_1 and ω_2. Indeed, under the conditions of our experient the frequency of the sum harmonic lies far from the electronic resonances of the materials used for preparing the MPS and/or Bragg band gaps. This makes it possible to rule out amplification due to synchronous mechanisms, i.e., amplification arising when additional phase-matching conditions are satisfied in the presence of spatial and frequency dispersions.

2. Let us examine the generation of a signal at the sum frequency in an MPS consisting of alternating layers with substantially different refractive indices. It is assumed that the layers with a large refractive index have a large quadratic nonlinearity.

Since the sum-harmonic signal is weak compared with the intensities of the incident waves, and taking into account the fact that the duration of the pulses is much longer than the propagation time of light through the sample, we shall seek the spatially rapidly varying amplitude of the quasistationary fields $E(\mathbf{r},t)=E(\mathbf{r})\exp(-i\omega t)$ at the fundamental frequencies, solving the linear wave equation

$$\Delta^2 E(\mathbf{r})+(\omega n(z)/c)^2 E(\mathbf{r})=0.$$

The exact solution of this equation in an arbitrary layer with number m has the form of a sum of the direct and backward (reflected) plane waves

$$E_{jm}(z)=A_{jm}\exp(ik_{j0}s_{jm}z)+B_{jm}\exp(-ik_{j0}s_{jm}z),$$

where $s_{jm}=(n_{jm}^2-\sin^2\vartheta)^{1/2}$, the indices $j=1,2$ correspond to one of the incident waves, n_{jm} is the refractive index for the j-th wave in the m-th layer, $k_{j0}=2\pi/\lambda_j$, ϑ is the angle of incidence, and z is the coordinate ''into'' the structure. To determine the amplitudes A_{jm} and B_{jm} we used the Parratt's method of recurrence relations,[6] which for the present problem yields the relations

$$A_{j,m+1}=A_{jm}Q_{jm}\frac{g_{jm}+R_{jm}g_{jm}^{-1}}{1+R_{j,m+1}}, \quad B_{jm}=R_{jm}A_{jm}, \tag{1}$$

$$R_{jm}=\frac{R_{j,m+1}+F_{jm}}{1+R_{j,m+1}F_{jm}}g_{jm}^2, \quad F_{jm}=\frac{s_{jm}-s_{j,m+1}Q_{jm}^2}{s_{jm}+s_{j,m+1}Q_{jm}^2},$$

where $g_{jm}=\exp(ik_{j0}s_{jm}d_m)$, d_m is the thickness of the m-th layer, $Q_{jm}=1$ and $Q_{jm}=n_{jm}/n_{j,m+1}$, respectively, for the s- and p-polarized waves. The recurrence relations (1) are solved for the boundary conditions $A_{j0}=E_j$; the reflection coefficient of the $N+1$ boundary is $R_{N+1}=0$; the vacuum refractive index is $n_0=1$; $g_0=1$; E_j are the amplitudes of the incident waves; and, N is the number of layers. Assuming that the intensity of the sum-frequency signal in the m-th layer is proportional to $|A_{1m}A_{2m}|^2$ for the direct and $|B_{1m}B_{2m}|^2$ for the backward waves, we shall estimate the total intensity of the SF signal as the sum of the corresponding intensities over all layers of the MPS with a large refractive index.

The computed dependences of the linear reflection coefficient of the MPS described below are presented in Figs. 1a, b for the two wavelengths $\lambda_1=812$ nm and $\lambda_2=733$ nm of the incident linearly p-polarized (i.e., polarized in the plane of incidence) radiation. The values chosen for λ_1 and λ_2 correspond to two edges of the Bragg band gap with angle of incidence of the radiation on the medium (MPS) $\vartheta=25°$. According to Eq. (1), near this value of the angle ϑ the amplitudes of the waves diffracted in the medium grow substantially, and the maximum localization of the energy of the incident radiation in the medium occurs at both wavelengths. This is clearly seen from the theoretical angular dependences of the total energy density of the corresponding backward waves $|B_{1m}|^2$ and $|B_{2m}|^2$ in layers with a large refractive index (Fig. 1a and 1b). For this reason, the efficiency of generation of the reflected SF signal depends on the angle ϑ, and its intensity I_{sf} has a maximum at $\vartheta=25°$ (Fig. 1c), solid line). If λ_1 and λ_2 are

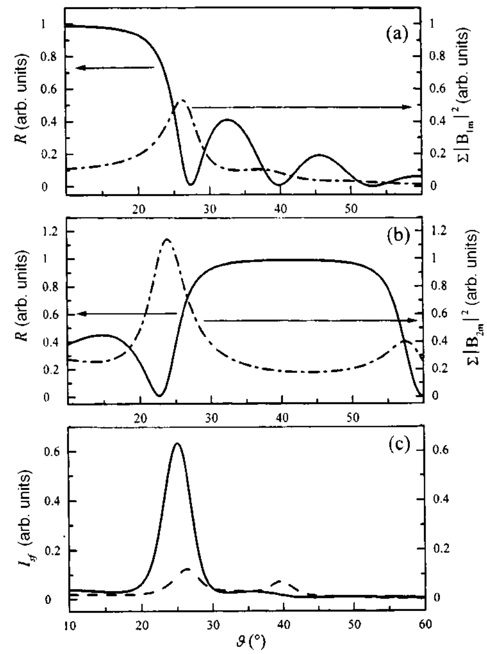

FIG. 1. a, b. Computed dependences of the reflection coefficient R versus the angle of incidence ϑ of the radiation on the MPS for two wavelengths $\lambda_1 = 812$ nm (a) and $\lambda_2 = 733$ nm (solid lines) and the angular dependences of the total energy density of the corresponding backward waves $\Sigma|B_{1m}|^2$ and $\Sigma|B_{2m}|^2$ in nonlinear layers (dot-dashed curves); c) computed angular dependences of the intensity I_{sf} of the reflected SF signal with simultaneous incidence of two waves with $\lambda_1 = 812$ nm and $\lambda_2 = 733$ nm (solid line) on the MPS at an angle ϑ and for the case where $\lambda_1 = 812$ nm and $\lambda_2 = 706$ nm (dashed line).

chosen so that the edges of the reflection curves do not intersect, for example, $\lambda_1 = 812$ nm and $\lambda_2 = 706$ nm, the intensity of the SF signal is low, and amplification does not occur (Fig. 1c, dashed line). The second maximum of the SF signal at $\vartheta = 40°$ (Fig. 1c) corresponds to the position of the edge of the reflection curve for $\lambda = 706$ nm.

In summary, for the optimal choice of wavelengths λ_1 and λ_2 amplification of the generation efficiency of the SF frequency, which does not depend on the satisfaction of phase-matching conditions, can be expected.

3. To observe the asynchronous amplification of the SF signal experimentally, we used a sample consisting of eight layers of ZnS ($n_1 = 2.316$) and seven layers of SrF_2 ($n_2 = 1.52$), deposited in the form of an MPS on a glass substrate.[4] The thickness of each layer was $d_i = 3\lambda/4n_i$ for wavelength $\lambda = 790$ nm. In all experiments λ_1 was fixed and equal to 812 nm (first channel, ω_1), and λ_2 could vary from 650 to 740 nm (second channel, ω_2); the duration of the pulses was less than 200 fs, the pulse repetition frequency was 200 kHz, and the energy could vary from 0 to 20 nJ/pulse. The synchronous detection technique was used to detect the SF signal. Both radiations at the fundamental frequencies (ω_1 and ω_2) and at the SF were linearly p-polarized.

The efficiency of generation of radiation at the SF as a function of the angle ϑ of incidence on the MPS was measured in the experiments.

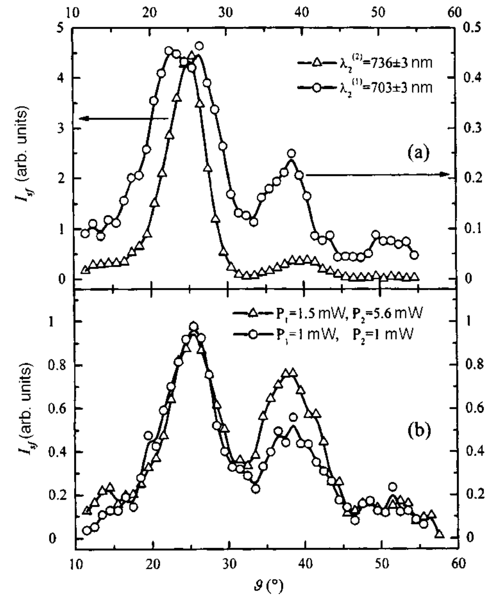

FIG. 2. a) Experimental dependences of the SF signal intensity I_{sf} versus the angle of incidence ϑ on the MPS for two different wavelengths $\lambda_2^{(1)}$ and $\lambda_2^{(2)}$ in the second channel; b) Angular dependence of the SF signal intensity I_{sf} for two different values of the average radiation power (P_1 and P_2) in the channels. The plots are normalized to the signal maximum for $\vartheta = 24°$.

The experimental dependences of the SF signal intensity on the angle of incidence on the MPS are shown in Fig. 2a for two values of the wavelengths of the second channel $\lambda_2^{(1)}$ and $\lambda_2^{(2)}$. The wavelengths $\lambda_2^{(1)} = 736$ nm and $\lambda_2^{(2)} = 703$ nm were chosen so that the edges of the reflection curves corresponding to them and $\lambda_1 = 812$ nm would intersect in one case ($\lambda_2^{(1)}$) and not in the other ($\lambda_2^{(2)}$). For the wavelength $\lambda_2^{(1)}$, an at least ten-fold amplification of the SF signal compared with the case $\lambda_2^{(2)}$ was observed experimentally; this agrees well with the theoretical results (Fig. 1c). The dependence of the SF signal amplitude on the angle of incidence on the MPS is presented in Fig. 2b for various values of the power of the incident radiation. It is evident from the figure that the maximum near $\vartheta = 40°$, corresponding to an increase in the energy density of the field near the edge of the reflection curve for radiation at the wavelength $\lambda_2^{(2)} = 703$ nm, increases with the power of the radiation at the fundamental frequency in the second channel.

4. The good agreement between the theoretical and experimental results shows that we have observed experimentally the asynchronous amplification of the generation of the SF signal in an MPS. The mechanism of such amplification is an increase in the energy density of the localized fields at two frequencies (ω_1, ω_2) near the opposite edges of a fixed Bragg band gap.

In closing, this work was partially supported by the Russian Fund for Fundamental Research (Project No. 98-02-17544), the program ''Universities of Russia'' and the Learning-Science Center for Fundamental Optics and Spectroscopy. Laboratoire de PhysicoChimie de l'Atmosphère is a member of Center d'Etude et de Recherche Lasers et Applications, which is supported by Ministère de la Recherche, the Région Nord/Pas de Calais, and Fond Européen de Développement Economique des Régions.

*)e-mail: ilya@lasmed.ilc.msu.su

[1] *Photonic Band Gap Materials*, edited by C. M. Soukoulis (Kluwer Academic, Dordrecht, 1996); E. Yablonovitch, J. Mod. Opt. **41**, 173 (1994).

[2] A. Yariv and P. Yen, *Optical Waves in Crystals, Propagation and Control of Laser Radiation*, John Wiley & Sons, New York, 1984) [Russian translation, Mir, Moscow, 1987, p. 616].

[3] M. Scalora, M. J. Bloemer, A. S. Manka *et al.*, Phys. Rev. A **56**, 3166 (1997); J. W. Haus, R. Viswanathan, M. Scalora *et al.*, Phys. Rev. A **57**, 2120 (1998).

[4] A. V. Balakin, D. Boucher, V. A. Bushuev *et al.*, Opt. Lett. **24**, 793 (1999).

[5] A. V. Andreev, O. A. Andreeva, A. V. Balakin *et al.*, Kvant. Élektron. **28**, 75 (1999) [Quantum Electron. **29**, 632 (1999)].

[6] L. G. Parratt, Phys. Rev. **95**, 359 (1954).

Translated by M. E. Alferieff