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Backward-travelling second-harmonic generation and the quasi-phase matching in an optical superlattice

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Abstract. The second-harmonic generation in a newly developed optical superlattice is analysed. Two second harmonics are predicted: a forward-travelling harmonic and a backward-travelling harmonic. Each can be enhanced enormously by the quasi-phase-matching technique.

Shortly after the advent of laser devices, the importance of phase matching in second-harmonic generation was recognized [1]. Meanwhile the quasi-phase-matching concept was proposed [2], for which a periodic structure is a most suitable candidate. In such a periodic structure, not only a forward-travelling second harmonic but also a backward-travelling second harmonic can be enhanced greatly through the quasi-phase-matching technique. In a refractive-index-modulated structure, Bloembergen and Sievers [3] suggested the generation of a backward-travelling second harmonic due to the folded-zone effect. Tang and Bey [4] extended the analysis to include the modulation of both the refractive index and the non-linear optical coefficient. Two approximations were made: the small-signal approximation and the parabolic approximation. Van der Ziel and Ilegems [5] were the first to observe this phenomenon experimentally. In a parametric oscillation process, there also exists a backward-travelling wave [6].

In this paper, we report our theoretical studies on the process of second-harmonic generation in an optical superlattice developed by us recently [7–15]. The parabolic approximation is removed. Only the small-signal approximation is retained. The theory predicts the generation of a backward-travelling second harmonic as well as a forward-travelling second harmonic. Each can be enhanced enormously by the quasi-phase-matching method.

The optical superlattice is a single $LiNbO_3$ crystal with periodic laminar ferroelectric domains. It has the following properties. Firstly, it is a single crystal and not a heterostructure. Secondly, only 180° ferroelectric domains can exist in it according to its symmetry. These 180° ferroelectric domains, or the positive and the

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negative domains are arranged along one direction periodically. Thirdly, it is easy to prove that the physical tensors with odd rank change their signs from one domain to the next, whereas the even-rank tensors such as the dielectric constant are unchanged.

The advantage of the optical superlattice over a homogeneous medium is that it can make non-linear interactions quasi-phase matched at any desired temperature, using any non-linear coefficient and involving any wavelength within the transparency range of the structure [16].

In the early 1980s, we verified successfully the quasi-phase-matching theory with an optical superlattice [17, 18] made of a single $LiNbO_3$ crystal with periodic laminar ferroelectric domains. An important conclusion was obtained. Comparison between the second-harmonic output of crystals with an optical superlattice in the quasi-phase-matching regime $e^{\omega} \cdot e^{\omega} \to e^{2\omega}$, d_{33} is used) and that of single-domain crystals of the same length and optical quality under the phase-matching condition ($o^{\omega} \cdot o^{\omega} \to e^{2\omega}$, d_{31} is used which is much smaller than d_{33}), enhancement of the second-harmonic output for one order of magnitude was observed. In 1985, Feisst and Koidl [19] repeated the experiment with a superlattice of fewer periods. More recently, Magel *et al* [16] realized second-harmonic generation of blue light in periodically poled $LiNbO_3$. So the optical superlattice discussed here is potentially useful for practical applications.

Here we assume that the thicknesses of the positive and negative domains are the same and equal to the coherent length. In order to use the largest non-linear optical coefficient d_{33} of $LiNbO_3$ crystals, we also assume that the domain boundaries are parallel to the y–z plane and that the polarization of the electric fields, including the fundamental (E_1) and the second harmonic (E_2), are along the z axis with their propagation directions along the x axis.

Starting from the Maxwell equation

$$\nabla^2 \boldsymbol{E}(x,t) - (\epsilon_3/c^2)[\partial^2 \boldsymbol{E}(x,t)/\partial t^2] = (4\pi/c^2)(\partial^2 \boldsymbol{P}/\partial t^2) \tag{1}$$

and using the small-signal approximation, we obtain

$$\partial^2 E_2/\partial x^2 - 2\mathrm{i}k^{2\omega}(\partial E_2/\partial x) = -(64\pi\omega^2/c^2)d_{33}f(x)E_1^2 \ \exp[\mathrm{i}(k^{2\omega} - 2k^{\omega})x] \tag{2}$$

where $E_2(x,t)$ takes the form

$$E_2(x,t) = E_2(x) \ \exp[\mathrm{i}(2\omega t - k^{2\omega}x)] \tag{3}$$

and

$$f(x) = \begin{cases} +1 & \text{if } x \text{ is in the positive domain} \\ -1 & \text{if } x \text{ is in the negative domain.} \end{cases}$$

c is the speed of light in vacuum and $\boldsymbol{P}$ is a non-linear polarization vector.

Conventionally, the second-order derivative in equation (2) is neglected; this is the so-called parabolic approximation. Here we shall solve equation (2) directly.

The solution of equation (2) can be obtained in two steps. First, we solve the homogeneous equation

$$\partial^2 E_2/\partial x^2 - 2\mathrm{i}k^{2\omega}(\partial E_2/\partial x) = 0. \tag{4}$$

Its solution is

$$E_{20} = C_1 + C_2 \exp(\mathrm{i}2k^{2\omega} x). \tag{5}$$

Next, we use the method of variation of constants to obtain the solution of equation (2), which is

$$E_2 = C_1(x) + C_2(x) \exp(\mathrm{i}2k^{2\omega} x). \tag{6}$$

Through conventional procedures, we have

$$\partial C_1/\partial x = -\mathrm{i}(32\pi\omega^2/k^{2\omega}c^2)d_{33}f(x)E_1^2 \exp(\mathrm{i}\,\Delta k\, x) \tag{7}$$

$$\partial C_2/\partial x = \mathrm{i}(32\pi\omega^2/k^{2\omega}c^2)d_{33}f(x)E_1^2 \exp(-\mathrm{i}kx) \tag{8}$$

with $\Delta k = k^{2\omega} - 2k^{\omega}$ and $k = k^{2\omega} + 2k^{\omega}$.

The periodic function $f(x)$ can be written as a Fourier series [20]

$$f(x) = \sum_{m\neq 0} \frac{\mathrm{i}[1-\cos(m\pi)]}{m\pi} \exp(-\mathrm{i}G_m x) \tag{9}$$

where $G_m = 2\pi m/L_\mathrm{p}$ is the reciprocal vector of the superlattice, and L_p is its period.

The solution of equation (7) describes the forward-travelling second-harmonic wave and the solution of equation (8) describes the backward-travelling wave. The solutions of equations (7) and (8) are

$$C_1 = -\mathrm{i}\frac{64\pi\omega^2}{k^{2\omega}c^2}d_{33}E_1^2 \sum_{m\neq 0} \frac{\mathrm{i}[1-\cos(m\pi)]}{m\pi} \frac{\sin[(\Delta k - G_m)L/2]}{\Delta k - G_m} \exp[\mathrm{i}\tfrac{1}{2}(\Delta k - G_m)L] \tag{10}$$

$$C_2 = -\mathrm{i}\frac{64\pi\omega^2}{k^{2\omega}c^2}d_{33}E_1^2 \sum_{m\neq 0} \frac{\mathrm{i}[1-\cos(m\pi)]}{m\pi} \frac{\sin[(K + G_m)L/2]}{K + G_m} \exp[-\mathrm{i}\tfrac{1}{2}(K+G_m)L]. \tag{11}$$

L is the total thickness of the optical superlattice. In obtaining equations (10) and (11), the boundary conditions $C_{01}(0) = 0$ and $C_{02}(L) = 0$ have been used.

Formally, equations (10) and (11) are much the same. For the forward-travelling second harmonic, the quasi-phase-matching condition is

$$\Delta k - G_m = 0. \tag{12}$$

For the backward-travelling second harmonic, the quasi-phase-matching condition is

$$K + G_m = 0. \tag{13}$$

It is seen from equations (10) and (11) that, when these conditions are satisfied, the energy conversion will be greatly enhanced; when they are not satisfied, the energy conversion will be very low. For a given structure, only one kind of second-harmonic wave can be enhanced efficiently—either the forward-travelling wave or the backward-travelling wave.

Obviously, in any case, K cannot be equal to zero; hence the backward-travelling second harmonic can never be phase matched in a homogeneous medium. So in a homogeneous medium it is reasonable to neglect the second-order derivative in equation (2), which results in the disappearance of the backward-travelling second harmonic. However, in a periodic superlattice, the phase mismatch of the second-harmonic generation process caused by the dispersion of the refractive index can be compensated by the reciprocal vector which the structure provides. Therefore, the backward-travelling second harmonic can be quasi-phase matched and enhanced enormously.

In conclusion, theoretical studies are made on a newly developed optical superlattice. The parabolic approximation is removed; only the small-signal approximation is retained. Two second harmonics are predicted. Each can be enhanced greatly by the quasi-phase-matching technique.

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