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Electro-optic effect and transmission spectrum in a Fibonacci optical superlattice

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Received 9 April 1992, in final form 19 June 1992

Abstract. A new type of Fibonacci optical superlattice is analysed. Its electro-optic effect is studied. The phase-matching concept is proposed for the first time for the Fibonacci optical superlattice. The transmission spectrum is non-self-similar owing to the dispersive effect of the refractive index. An extinction phenomenon exists provided that the thicknesses of the domains are properly selected.

1. Introduction

The discovery of quasi-crystalline phases in metallic alloys has opened up a new field in solid state physics [1]. Since then, much work has been done on the light transmission properties of the one-dimensional quasi-periodic superlattice [2–5], a heterostructure with quasi-periodic ordering of multilayers. In these studies, its linear phenomena [2–4] and its third-order non-linearity [5] have been greatly emphasized and the physical parameters such as the dielectric coefficients and Kerr and electrostriction non-linearity constants were taken to be non-dispersive. Little work has been done on the phenomena associated with parameters of third-rank tensors because of lack of appropriate materials.

Recent development in technology makes it possible to prepare a Fibonacci optical superlattice (Fos) of single LiNbO₃ crystals with quasi-periodic laminar ferroelectric domains by waveguide fabrication [6]. It provides a useful tool for the study of phenomena associated with third-rank tensors. Previously we investigated second-harmonic generation both in a periodic optical superlattice and in an FOS [7-13]. Here we shall study its electro-optic effect and shall restrict ourselves to the so-called FOS.

In this paper, we shall report our theoretical results of the basic features of the transmission spectrum of an FOS, which is made from a single $LiNbO_3$ crystal with quasi-periodic ferroelectric domain structures. We shall first discuss its mathematical treatment in section 2. In section 3, we shall propose the phase-matching concept for the FOS. To our knowledge, no such concept has been suggested to date. The structure of the transmission spectrum will be described in section 4. We find that the

transmission spectrum is non-self-similar because of the dispersive effect of the refractive index. By choosing the thicknesses of the domains appropriately, an extinction phenomenon occurs.

2. Coupled-mode theory for the Fibonacci optical superlattice

The initial description of a quasi-periodic superlattice (Fibonacci superlattice) was given by Merlin *et al* [14]. Two different blocks A and B, each made up of two different materials, are layered according to a well prescribed rule, ABAABABA.... X-ray and Raman scattering measurements have revealed the special features of this non-periodic heterostructure [14].

Likewise, here we define our FOS. It also consists of two building blocks A and B that are arranged according to the Fibonacci sequence ABAABABA.... Each block consists of one positive ferroelectric domain and one negative ferroelectric domain as shown in figure 1, where l_{A1} and l_{B1} represent the thicknesses of the positive domains in blocks A and B, and l_{A2} and l_{B2} the thicknesses of the negative domains with $l_A = l_{A1} + l_{A2}$ and $l_B = l_{B1} + l_{B2}$. In our treatment, we have set $l_{A1} = l_{B1} = l$.



Figure 1. FOS made from a single $LiNbO_3$ crystal (the arrows indicate the directions of the spontaneous polarization). (a) The two building blocks of an FOS, each composed of one positive and one negative ferroelectric domain. (b) Schematic diagram of an FOS.

The positive domain and the negative domain are interrelated by a dyad axis in the x direction. In this kind of material, it is easy to prove that all the odd-rank tensors will change signs from one domain to the next, while all the even-rank tensors will remain the same. Therefore, the electro-optic tensor of the FOS, being a thirdrank tensor, will change its sign quasi-periodically and the dielectric tensor, being a second-rank tensor, will be constant through the FOS. However, under the action of an external electric field, the dielectric tensor will be modulated quasi-periodically because of the electro-optic effect. If the magnitude of the field is moderate (say about 10^6 V cm⁻¹), the change in the dielectric tensor is very small [15] and the modulation can be taken to be a perturbation. It will couple the energy of two unperturbed normal modes which are the allowed polarized states of the FOS in the absence of an electric field. Thus, the transmission spectrum of light with different polarizations should be tackled by the coupled-mode theory. Here we shall extend the coupled-mode theory, already established for the periodic superlattice [15], to the FOS.

In order to make the normal modes coupled together under the action of an electric field, it is necessary that the electrodes be on the y surfaces or the x surfaces of the Fos [15]. Here we choose the y surfaces to be the electrodes and the x axis to be the propagating direction of the light beams.



Figure 2. Geometrical configuration of the FOS. X, Y, Z denote the principal axes of the unperturbed dielectric tensor and X, Y', Z' the principal axes of the perturbed dielectric tensor.

The geometrical arrangement of our system is as follows. The polarizer has its transmission axis parallel to the y axis, and the analyser parallel to the z axis (figure 2). In the absence of an electric field, the FOS is homogeneous to the propagation of light and the direction of its principal axes are along the x, y, z axes, respectively. The dielectric tensor in the principal coordinate of the FOS is

$$\tilde{\epsilon}_{0} = \epsilon_{0} \begin{pmatrix} n_{o}^{2} & 0 & 0\\ 0 & n_{o}^{2} & 0\\ 0 & 0 & n_{e}^{2} \end{pmatrix}$$
(1)

where ϵ_0 is the dielectric constant of the vacuum, and n_o and n_e are the refractive indices of ordinary and extraordinary light, respectively.

In the presence of an electric field, because of the electro-optic effect, the FOS becomes inhomogeneous to the propagation of light. Using the fact that the perturbed terms are much smaller than the unperturbed terms, we obtain

$$\epsilon = \tilde{\epsilon}_0 + \Delta \epsilon \tag{2}$$

with

$$\Delta \epsilon = -\epsilon_0 r_{42} E_2 n_o^2 n_e^2 \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{bmatrix} f(x)$$
(3)

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

Here r_{42} is an electro-optic constant, $\Delta \epsilon$ can be treated as a small dielectric perturbation and E_2 is the applied external electric field.

At the same time, we obtain the rotation angle of the optic axis due to the electro-optic effect (see figure 2):

$$\tan(2\alpha) = 2r_{42}E_2f(x)/[(n_o^2)^{-1} - (n_e^2)^{-1}].$$
(4)

Obviously, the angle rocks back and forth from $+\alpha$ to $-\alpha$ along the x direction from one domain to the next. In this way the function of the FOS is similar to a Solc filter [16] with quasi-periodicity.

The following derivation is much the same as the derivation used for the periodic superlattice [15].

First, we expand f(x) as a Fourier integral:

$$f(x) = \int f(k) \exp(-ikx) dk$$
(5)

and

$$f(k) = \frac{1}{2\pi} \int f(x) \exp(ikx) dx$$
$$= \frac{1}{ik\pi} \left(\sum_{j} \exp(ikx_{2j+1}) + \exp(i\pi) \sum_{j} \exp(ikx_{2j}) \right)$$
(6)

where $\{x_n\}$ are the positions of the ferroelectric domain boundaries (figure 1).

In equation (6), the summation terms comprise the structure factor, which is divided into two parts with one part lagging behind the other by a phase $\exp[i(kl + \pi)]$. Thus, equation (6) can be written as

$$f(k) = \frac{2}{\pi k} \exp(i\frac{1}{2}kl) \sin(\frac{1}{2})kl \sum_{j} \exp(ikx_{2j}).$$
 (7)

For an infinite array with $l_A/l_B = \tau$, according to [17–19], equation (7) transforms to

$$f(k) \propto \sum_{m,n} \exp[i(\frac{1}{2}kl + X_{m,n})] \frac{\sin(\frac{1}{2}kl)}{k} \frac{\sin X_{m,n}}{X_{m,n}} \delta(k - G_{m,n})$$
(8)

where

$$X_{m,n} = \pi \tau^2 (m\tau - n) / (1 + \tau^2)$$
⁽⁹⁾

$$G_{m,n} = 2\pi (m+n\tau)/D \tag{10}$$

with

$$D = \tau l_{\rm A} + l_{\rm B}.\tag{11}$$

Substituting equation (8) into equation (5), we have

$$f(x) \propto \sum_{m,n} \exp\left[i(\frac{1}{2}G_{m,n}l + X_{m,n})\right] \frac{\sin(\frac{1}{2}G_{m,n}l)}{G_{m,n}} \frac{\sin X_{m,n}}{X_{m,n}} \exp(-iG_{m,n}x)$$
(12)

where $G_{m,n}$ is the reciprocal vector of the FOS. Analogous to the periodic lattice, it is reasonable to anticipate that this lattice may play a significant role in interactions proceeding in the FOS.

Then, we derive the coupled wave equations. From equation (3) it is clear that only the y-polarized light and the z-polarized light can be coupled together. Analogous to the procedures in [15], starting from Maxwell's equations and using the parabolic approximation, we obtain

$$dA_2/dx = -iKA_3 \exp(i\Delta\beta x)$$

$$dA_3/dx = -iK^*A_2 \exp(-i\Delta\beta x)$$
(13)

with

$$\Delta \beta = \beta_2 - \beta_3 - G_{m,n} \tag{14}$$

$$K = -\frac{1}{2}(\omega/c)(n_o^2 n_e^2/\sqrt{n_o n_e})r_{42}E_2 A_{m,n}$$
(15)

$$A_{m,n} \propto \exp\left[i(\frac{1}{2}G_{m,n}l + X_{m,n})\right] \left\{ \left[\sin(\frac{1}{2}G_{m,n}l)\right]/G_{m,n} \right\} \left[(\sin X_{m,n})/X_{m,n}\right]$$
(16)

where A_2 and A_3 are the mode amplitudes for y-polarized and z-polarized light, respectively, and β_2 and β_3 are the corresponding wavenumbers.

The initial condition at x = 0 which is determined by the polarizer is given by

$$A_2(0) = 1 (17) (17)$$

The solution of the coupled equations is then

$$A_{2}(x) = \exp(i\frac{1}{2}\Delta\beta x)[\cos(sx) - i(\Delta\beta/2s)\sin(sx)]$$

$$A_{3}(x) = [\exp(-i\frac{1}{2}\Delta\beta x)(-iK^{*})\sin(sx)]/s$$
(18)

where s is given by

$$s^{2} = K^{*}K + (\Delta\beta/2)^{2}.$$
(19)

At the analyser (z polarized), x = L (which is directly related to the block number N); A_2 is extinguished. The transmission for the z-polarized light is thus given by

$$T = [|K|^2 \sin^2(sL)]/s^2.$$
⁽²⁰⁾

This is the same as the well known Ewald pendellösung or pendulum solution [20]. The energy is passed back and forth between the incident and diffracted beams as they travel in the FOS. The coupling is provided in this case by the scattering from one beam to the other owing to the existence of a perturbed dielectric tensor.

In the following, numerical calculations are made and some interesting phenomena in the FOS are discussed. In all the calculations, we have set $l_{\rm B2} = \tau(\tau l - l_{\rm A2})$, which is mainly required in the discussion of an extinction phenomenon. This expression combined with $l_{\rm A} = \tau l_{\rm B}$ gives $D = 2(1 + \tau)l$. Moreover, we have set $l = \pi/\Delta\beta_0 = 5.0441 \ \mu \text{m}$, where $\Delta\beta_0 = \beta_2 - \beta_3 = 2\pi(n_o - n_e)/\lambda_0$ with $\lambda_0 = 0.8000 \ \mu \text{m}$.

3. Phase-matching concept and dynamical effect

When one discusses interactions between waves in a dielectric medium, one often encounters the phase-matching concept. The phase matching plays an important role in many processes such as the non-linear optic, electro-optic and acousto-optic processes. If the phase-matching condition is satisfied, energy conversion can be complete; if not, the conversion will be inefficient. In homogeneous dielectric media and periodic dielectric media, such a concept has already been established [15, 21]. In recent years, research on quasi-periodic superlattices has made much progress but, to our knowledge, no such concept has been proposed for quasi-periodic superlattices up to now.

From equation (20), we find that, for significant mode coupling to take place between modes 2 and 3, three conditions must be fulfilled. The first is a kinematic condition which is

$$\Delta \beta = 0. \tag{21}$$

Equation (21) will be referred to as the phase-matching condition. This condition is a counterpart of that for periodic structures [15]. In both cases, the reciprocal vector plays an important role. It is the reciprocal vector that compensates the birefringence and makes the energy-coupling process proceed efficiently.

We have calculated the dependence of transmission on the block number N of the FOS for some $G_{m,n}$. We find that, for a particular wavelength, there exists a corresponding reciprocal vector $G_{m,n}$ by which the phase-matching condition is satisfied. For example, for $\lambda = 0.8000 \ \mu m$ and $\lambda = 0.5478 \ \mu m$, the corresponding reciprocal vectors are $G_{1,1}$ and $G_{1,2}$, respectively, which satisfy the phase-matching condition. Figure 3 shows the results. For comparison, the transmission of y-polarized light is also shown. It can be seen that, when equation (21) is valid, the energy conversion can be complete. In figure 4, a wavelength is chosen such that the phasematching condition is not satisfied; clearly the energy conversion is very low.

The second condition is

$$|K|L = (2u+1)\pi/2$$
 $u = 0, 1, 2, ...$ (22)

which is a dynamical condition. If m and n are fixed, then K is a constant. Equation (22) thus indicates that for some value of L, i.e. for some value of the block number N, the transmission will be at its maximum. For some other value of N, the transmission will be zero if

$$[K]L = v\pi$$
 $v = 1, 2, 3, ...$ (23)

is satisfied. It is this dynamical effect that makes the transmission oscillate almost sinusoidally with the block number N, as can be seen in figure 3. Analogous to x-ray diffraction and electron diffraction [20, 22], here $|K|^{-1}$ can be defined as the extinction distance.

The third is that K must not vanish. If it does, then the transmission will be zero. We call it the extinction phenomenon, which will be discussed in section 4. Here we point out that the extinction phenomenon is totally determined by the thicknesses of the domains and not by the block number N, whereas the extinction distance is determined by N and is a dynamical effect. These two different phenomena should not be confused.



Figure 3. Dependence of the transmission on the block number N with phase matching and $l = 5.0441 \ \mu m$ (curves A) and, for comparison, the transmission of the ypolarized light is also shown (curves B): (a) $\lambda = 0.8000 \ \mu m$, $\beta_2 - \beta_3 - G_{1,1} = 0$; (b) $\lambda = 0.5478 \ \mu m$, $\beta_2 - \beta_3 - G_{1,2} = 0$.

4. Transmission spectrum

 $\Delta\beta$ in equation (20) (included in s) is of much significance not only because it determines the degree of energy conversion but also because it determines the characteristic of the transmission spectrum. Let us rewrite equation (21) in its explicit form

$$(1/\lambda)(n_{\rm o} - n_{\rm e}) = (m + n\tau)/2(1 + \tau)l.$$
⁽²⁴⁾

Equation (24) is very similar to the equation obtained for the second-harmonic generation in an FOS [9]. Both come from the energy coupling between waves and in both cases the dispersion of the refractive indices must be taken into account.

In order to gain insight into the main features of the spectrum, numerical calculations have been made which are valid only at room temperature. In addition, the dispersion of the electro-optic coefficient is not considered because of lack of



Figure 4. Dependence of the transmission on the block number N with no phase matching, $l = 5.0441 \ \mu m$ and $\lambda = 0.7980 \ \mu m$ (curve A). For comparison, the transmission of y-polarized light is also shown (curve B).



Figure 5. Dependence of the transmission on the wavelength. Note that $(1/\lambda)_{1,p+1} \neq (1/\lambda)_{1,p} + (1/\lambda)_{1,p-1}$. Here $l = 5.0441 \ \mu \text{m}$.

sufficient data. This will only affect the peak heights and does not affect the peak positions as can be seen in equations (15), (20) and (21).

Taking into account the dispersion of the refractive indices [22], we rewrite equation (24) as follows:

$$(1/\lambda)_{m,n} = (m+n\tau)/2[n_{\rm o}(\lambda) - n_{\rm e}(\lambda)](1+\tau)l$$
⁽²⁵⁾

where $n_o(\lambda)$ and $n_e(\lambda)$ are functions of λ [23]. In this case, the transmission spectrum will be affected by the dispersion. For those peaks where n, m are successive Fibonacci numbers, equation (25) becomes

$$(1/\lambda)_{1,p} = \tau^p / 2[n_o(\lambda) - n_e(\lambda)](1+\tau)l.$$
 (26)

The relation

$$(1/\lambda)_{1,p+1} = (1/\lambda)_{1,p} + (1/\lambda)_{1,p-1}$$
(27)

is no longer valid because of the dispersion.

Figure 5 shows the dependence of the transmission on the wavelength. As usual, the intense peaks occur at $(m, n) = (F_{k-1}, F_k)$, i.e. m, n being successive Fibonacci numbers, but their positions shift markedly. For example, in figure 5, we can see four intense peaks occurring at $\lambda_{s,p}$, as indicated by τ^p . They are $\lambda_{1,1} = 1.2240 \ \mu\text{m}, \ \lambda_{1,2} = 0.8000 \ \mu\text{m}, \ \lambda_{1,3} = 0.5478 \ \mu\text{m}$ and $\lambda_{1,4} = 0.4062 \ \mu\text{m}$. Obviously, $(1/\lambda)_{1,3} \neq (1/\lambda)_{1,2} + (1/\lambda)_{1,1}$, so the spectrum is non-self-similar. In other words, the spectrum does not reflect the self-similarity of the reciprocal space and does not reflect the symmetry of the quasi-periodic structure.

In the second-harmonic generation of an FOS, we have discussed the extinction phenomenon. Here this phenomenon also exists. In section 3, we have already mentioned this phenomenon. The general extinction rule can be derived directly from equations (15), (16) and (20). We can see from them that the transmission depends on K as well as on $\Delta\beta$. The extinction occurs when K = 0. From equations (15) and (16), this happens when

$$\sin(\frac{1}{2}G_{m,n}l) = 0 \tag{28}$$

i.e.

$$l = 2j(\pi/G_{m,n}) = jD/(m+n\tau)$$
(29)

namely all peaks satisfying equation (29) are absent in the spectrum. According to equation (29), some peaks can be purposely made to be absent provided that the thicknesses of the domains are appropriately selected. For instance, if we want the peaks with their indices (m, n) = (j, 2j) to be extinguished, after substituting (m, n) = (j, 2j) into equation (29), we have

$$l_{\rm B2} = \tau (l - l_{\rm A2}). \tag{30}$$

If we set

$$l_{\rm B2} = \tau (\tau l - l_{\rm A2}) \tag{31}$$

then peaks with the indices (m, n) = (2j, 2j) are absent in the spectrum; this can be seen in figure 5, where the peak with its index (m, n) = (2, 2) is missing.

5. Conclusion

We have analysed theoretically the electro-optic effect in an FOS. The phase-matching concept has been presented for the first time for the FOS. The transmission spectrum shows non-self-similarity due to the dispersion of the refractive indices. An extinction phenomenon has been discussed.

Acknowledgment

This work was supported by a grant for the Key Research Project from the State Science and Technology Commission of China.

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