Second-harmonic generation in a Fibonacci optical superlattice and the dispersive effect of the refractive index

Yong-yuan Zhu
Laboratory of Solid State Microstructures, Nanjing University, Nanjing 210 008, People's Republic of China

Nai-ben Ming*
Laboratory of Solid State Microstructures, Nanjing University, Nanjing 210 008, People's Republic of China and Center for Condensed Matter Physics and Radiation Physics, Chinese Center of Advanced Science and Technology (World Laboratory), P.O. Box 8730, Beijing 100 080, People's Republic of China

(Received 11 April 1990)

A Fibonacci optical superlattice is analyzed which is made from a single crystal with quasiperiodic laminar ferroelectric domain structures. The second-harmonic generation in this system is studied. Because of the dispersive effect of the refractive index, the second-harmonic spectrum does not reflect the symmetry of the quasiperiodic structure and thus does not exhibit self-similarity. The existence of the extinction phenomenon constitutes one major difference between our system and heterostructure systems. A general extinction rule is also obtained.

One of the most striking events in condensed-matter physics in recent years has been the discovery and development of quasiperiodic crystals which show many unused physical properties. Many subsequent researchers have focused on its linear phenomena[1-3] and its third-order nonlinearity.[4,5] In these works the physical parameters such as dielectric coefficients and elastic coefficients were taken to be nondispersive. Little has been done on the second-order nonlinear-optical phenomena because of lack of proper materials.

The Fibonacci optical superlattice (FOS) made from a single $LiNbO_3$ crystal with quasiperiodic laminar ferroelectric domain structures provides a useful tool for the study of second-order nonlinear-optical phenomena. Previously, we investigated second-harmonic generation in a periodic optical superlattice,[6,7] which is made from a single $LiNbO_3$ crystal with periodic laminar ferroelectric domain structures, and verified the theory of quasi-phase-matching proposed by Bloembergen *et al.*[8,9] In this paper we report our theoretical results of the second-harmonic generation in a FOS. Considering dispersive effects of the refractive index, we find that the spectrum of the second-harmonic intensity in a FOS doe not possess self-similarity. We also find that if the structure parameter of the FOS is properly selected, a phenomenon somewhat similar to the extinction phenomenon in solid-state physics will occur.

Traditionally, the Fibonacci superlattice is constructed as follows. First, define two building blocks A and B, each composed of two layers of different constituent materials. Then arrange them according to the concatenation rule $S_j = S_{j-1}|S_{j-2}$ for $j \geq 3$, with $S_1 = A$ and $S_2 = AB$. The Fibonacci superlattice thus formed is a heterostructure. In our case each block consists of one positive ferroelectric domain and one negative ferroelectric domain. The two domains are interrelated by a dyad axis in the x direction. Since the nonlinear-optical coefficients form a third-rank tensor, it is easy to prove that they will change their signs from positive domains to negative domains.[10] The superlattice thus formed is not a heterostructure, but still a single crystal. This kind of

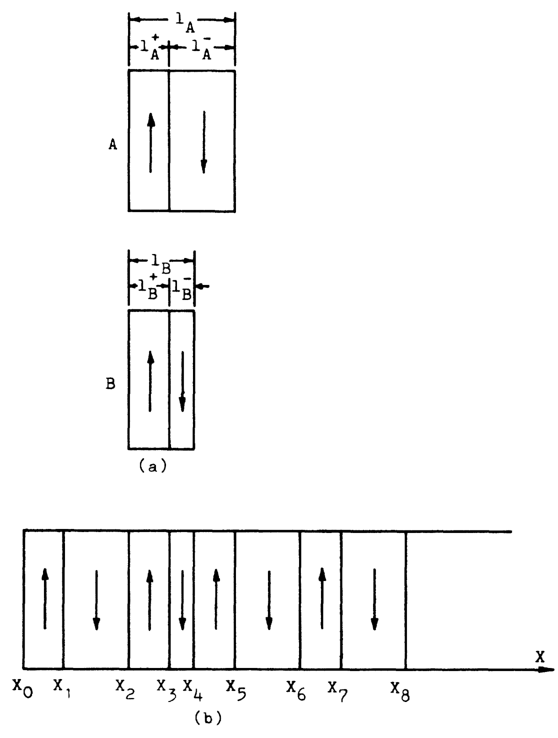

FIG. 1. FOS made of a single $LiNbO_3$ crystal (the arrows indicate the directions of the spontaneous polarization). (a) The two building blocks of a FOS, each composed of one positive and one negative, ferroelectric domain. (b) Schematic diagram of a FOS.

structure can be fabricated by a special crystal-growth technique developed by us.[11–13] Here we term this structure a Fibonacci optical superlattice, or FOS, which is shown in Fig. 1.

In Fig. 1 we can see that $l_A = l_A^+ + l_A^-$ and $l_B = l_B^+ + l_B^-$. In our treatment we have set $l_A^+ = l_B^+ = l$, $l_A^- = l(1+\eta)$, and $l_B^- = l(1-\tau\eta)$ with the golden ratio $\tau \equiv (1+\sqrt{5})/2$. Here, l and η are two adjustable parameters. Through the variation of the value of η, the structure can be either a periodic superlattice or a quasiperiodic one. For example, if $\eta=0$, then the structure is a periodic one; otherwise it is a quasiperiodic one. In our system, l plays an important role, and so we call it a structure parameter.

As will be seen below, the phase-matching regime cannot be used to study the effect of the FOS on the nonlinear-optical phenomena, and so the quasi-phase-matching regime[8,9] should be used. In order to use the largest nonlinear-optical coefficient d_{33} of $LiNbO_3$ crystals, we assume that the domain boundaries are parallel to the y-z plane (Fig. 1) and that the polarizations of the electric fields are along the z axis, with their propagating directions along the x axis.

In what follows we will restrict ourselves to the case of second-harmonic generation (SHG) with a single laser beam incident onto the surface of the FOS. According to Refs. 10 and 14, these two electric fields, E_1 and E_2, satisfy the wave equation under the small-signal approximation:

$$\frac{dE_2(x)}{dx} = -i\frac{32\pi\omega^2}{k^{2\omega}c^2} d(x) E_1^2 \exp[i(k^{2\omega}-2k^{\omega})x] , \tag{1}$$

with $d(x)=d_{33}$ in positive domains and $d(x)=-d_{33}$ in negative domains, where ω and k^{ω} are the angular frequency and wave number of the fundamental beam, respectively, $k^{2\omega}$ is the wave number of the second harmonic, and c is the speed of light in vacuum.

By integrating Eq. (1), the second-harmonic electric field after passing through N blocks of the FOS can be represented as

$$E_2(N) = -\frac{64\pi\omega^2}{k^{2\omega}c^2\,\Delta k} d_{33} E_1^2 \left\{ \sum_{j=0} e^{i\Delta k\, x_{2j+1}} + \exp(i\pi) \sum_{j=0} e^{i\Delta k\, x_{2j}} \right\} , \tag{2}$$

where $\Delta k = k^{2\omega} - 2k^{\omega}$. $\{x_n\}$, $n=0,1,2,\ldots$, are the positions of the ferroelectric domain boundaries (Fig. 1).

In Eq. (2) the terms inside the curly braces comprise the structure factor, which is divided into two parts, with one part lagging behind the other by a phase $\exp[i(\Delta k\, l+\pi)]$.

For an infinite array with $l_A/l_B=\tau$, i.e., $\eta\approx 0.34$, by use of the direct[15] or the projection method,[16] Eq. (2) can be written in the form

$$E_2(\Delta k) \propto -\frac{128\pi\omega^2}{k^{2\omega}c^2\Delta k} d_{33} E_1^2 \exp(i\tfrac{1}{2}\Delta k\, l) \sin(\tfrac{1}{2}\Delta k\, l) \sum \frac{\sin X_{m,n}}{X_{m,n}} \exp(iX_{m,n}) \delta(\Delta k - 2\pi(m+n\tau)/D) . \tag{3}$$

Here, $X_{m,n} = \pi\tau(m\tau - n)/(1+\tau^2)$, $D = \tau l_A + l_B$.

The appearance of Δk is due to the energy coupling between the fundamental beam and the second harmonic through the nonlinear-optical effect. Obviously the FOS cannot be used in the study of phase-matched SHG ($\Delta k=0$). It can be only used in the study of quasi-phase-matchable SHG.

The peaks of the second-harmonic intensity can be obtained from the δ function in Eq. (3), which is

$$\Delta k_{m,n} = 2\pi(m+n\tau)/D ,$$

or (4)

$$(\Delta k\, D)_{m,n} = 2\pi(m+n\tau) .$$

The factor $(\sin X_{m,n})/X_{m,n}$ in Eq. (3) is important. It determines which peaks are stronger. Here we know that the smaller the $X_{m,n}$, the larger the value of $(\sin X_{m,n})/X_{m,n}$. This means that n/m must be close to τ. It is well known that the best rational approximants to τ occur when n and m are successive Fibonacci numbers, F_k.[17] Therefore, the sequence of the most intense peaks corresponds to $(m,n)=(F_{k-1},F_k)$, where $(F_0,F_1)=(0,1)$. Note that the second-harmonic intensity peaks are indexed by two integers, even though the structure is one dimensional. The appearance of more indices than the dimensionality is typical of incommensurate crystals and quasicrystals.

We will discuss some interesting phenomena in both real space and reciprocal space. All calculational results are valid only under room temperature and, without loss of generality, only the results with $N=100$ have been presented.

In real space we study the dependence of the second-harmonic intensity on the structure parameter l of the FOS. In this case the wavelength λ_0 is kept unchanged, as are the refractive indices n_{10} and n_{20}. Here, n_{10} and n_{20} are the refractive indices for the fundamental beam and the second harmonic, respectively. Thus the dispersion of the refractive indices has no effect on the second-harmonic spectrum. Equation (4) can be rewritten as

$$l_{m,n} = \frac{(m+n\tau)\lambda_0}{4(n_{20}-n_{10})(1+\tau)} . \tag{5}$$

Here, λ_0 represents the wavelength of the fundamental frequency in vacuum.

For those intense peaks, Eq. (5) becomes

$$l(s,p) = \frac{\lambda_0}{4(n_{20}-n_{10})(1+\tau)} s\tau^p . \tag{6}$$

Here, s and p are integers.

Obviously, here the relation $l(s,p+1)=l(s,p)+l(s,p-1)$ holds, and thus the spectrum of the second harmonic exhibits self-similarity. Figure 2 shows the relation between the second-harmonic intensity and the structure parameter l with the pump beam at wavelength $\lambda_0=1.318$ μm and $n_{10}=2.1453$ and $n_{20}=2.1970$ for $LiNbO_3$ crystals under the condition $l_A/l_B=\tau$. The result conforms to the discussion above. The intense peaks take the form of $\lambda_0 s\tau^p[4(n_{20}-n_{10})(1+\tau)]$. Under general conditions, i.e., $l_A \neq \tau l_B$, calculations have shown that the peak positions of the second-harmonic intensity remain unchanged, except for their strengths. In our case the change of the value of η does not affect the value of D. This can be seen easily from the relation $D=\tau l_A+l_B=2l(1+\tau)$. This indicates that D is a characteristic parameter of the FOS. The result is consistent with that of Merlin *et al.*[4] They found that for all $l_A \neq l_B$ the Fourier spectrum of the structure factor of a Fibonacci superlattice consists of δ-function peaks at $k=2\pi(m+n\tau)/D$, with $D=\tau l_A+l_B$. We may deduce from these results that the FOS possesses certain space symmetry. The symmetry is determined by the order of arrangement of the blocks, *not* by the block thicknesses.

In reciprocal space we study the dependence of the second-harmonic intensity on the wavelength λ. In this case, the structure parameter l is kept constant. Equation (4) can be rewritten as

$$(1/\lambda)_{m,n}=\frac{m+n\tau}{4[n_2(\lambda)-n_1(\lambda)](1+\tau)l} . \tag{7}$$

Here, $n_1(\lambda)$ and $n_2(\lambda)$ are functions of λ.[18] Equation (7) indicates that here the dispersive effect of the refractive index on the second-harmonic spectrum must be taken into account; moreover, for these intense peaks Eq. (7) becomes

$$(1/\lambda)_{s,p}=\frac{s\tau^p}{4[n_2(\lambda)-n_1(\lambda)](1+\tau)l} . \tag{8}$$

The relation $(1/\lambda)_{s,p+1}=(1/\lambda)_{s,p}+(1/\lambda)_{s,p-1}$ no longer holds because of the dispersion of the refractive index, whereas in linear phenomena and the third-order nonlinear-optical phenomena this relation is valid.

Figure 3 shows the relation between the second-harmonic intensity and the wavelength with $l=l_c=\pi/\Delta k_0$ and $l_A=\tau l_B$. Here, $\Delta k_0=4\pi(n_{20}-n_{10})/\lambda_0$. As expected, the intense peaks occur at $(m,n)=(F_{k-1},F_k)$, but their positions have shifted markedly. For example, in Fig. 3 we can see three intense peaks occurring at $\lambda_{s,p}$, as indicated by τ^p. They are $\lambda_{1,2}=1.318$ μm, $\lambda_{1,3}=1.115$ μm, and $\lambda_{1,4}=0.960$ μm. Obviously, $(1/\lambda)_{1,4} \neq (1/\lambda)_{1,3}+(1/\lambda)_{1,2}$, and thus the spectrum of the second-harmonic intensity in reciprocal space does not exhibit self-similarity.

Here, another interesting phenomenon should be noted, one somewhat similar to the extinction phenomenon in solid-state physics. In both Figs. 2 and 3 the mode (2,2) does not appear. As discussed above, after the fundamental light passing through the entire superlattice, the resultant second-harmonic light can be viewed as being composed of two parts with a phase difference of $\exp[i(\Delta k\, l+\pi)]$. When the two parts of second-harmonic light satisfy the condition $(\Delta k\, l+\pi)=(2j+1)\pi$ or

$$l=2j(\pi/\Delta k) , \tag{9}$$

they interfere destructively. Here, $\pi/\Delta k$ is the coherence length for SHG.[10] That is to say, when the structure parameter l equals an even number times the coherence length, the corresponding SHG will disappear. This can be also deduced from Eq. (3) easily. In Eq. (3), for mode (2,2), we can obtain $\Delta k\, l=2\pi$ from the δ function, but then the factor $\sin(\Delta k\, l/2)=0$. Thus the second-harmonic intensity is zero. By substituting for $\Delta k\, l$ from Eq. (9) into the δ function in Eq. (3), we can obtain the general extinction rule, which is

$$(m,n)=(2j,2j) . \tag{10}$$

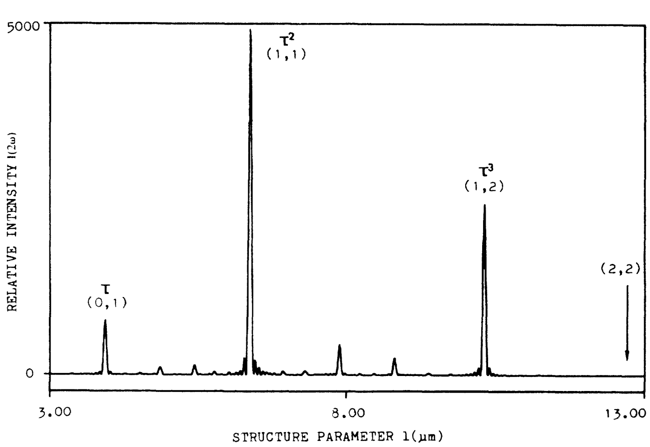

FIG. 2. Dependence of the second-harmonic intensity on the structure parameter l in real space. Note that $l(s,p+1)=l(s,p)+l(s,p-1)$.

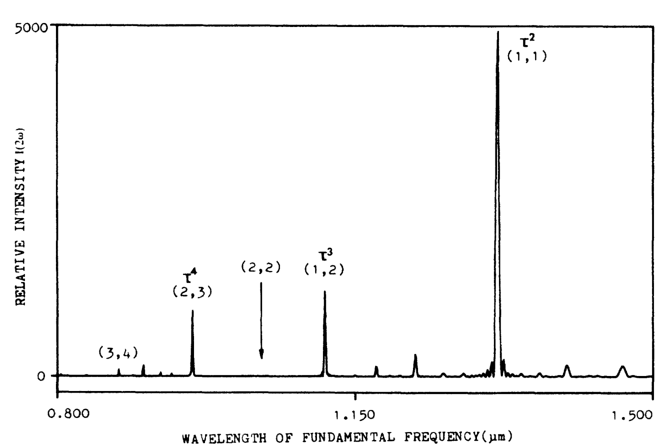

FIG. 3. Dependence of the second-harmonic intensity on the fundamental wavelength in reciprocal space. Note that $(1/\lambda)_{s,p+1} \neq (1/\lambda)_{s,p}+(1/\lambda)_{s,p-1}$.

Namely, all peaks with indices $(m,n)=(2j,2j)$ are absent in the spectrum of the second-harmonic intensity. This property may not be restricted to the nonlinear-optical effect exclusively. In our previous study[3] on ultrasonic excitation in a Fibonacci superlattice, which is the same as the one discussed here (in this case, the linear effect is discussed), the extinction phenomenon also exists, provided the structure parameter is selected properly. Nevertheless, in a conventional heterostructure superlattice this phenomenon does not exist. This constitutes one major difference between the FOS system and conventional heterostructure superlattices.

In conclusion, we have presented a FOS made of a single $LiNbO_3$ crystal and theoretically second-harmonic generation in the FOS. In our system we find that because of the dispersive effect of the refractive index, the spectrum of the second-harmonic intensity in reciprocal space does not reflect the symmetry of the quasiperiodic structure, and thus does not exhibit self-similarity. Also found is an extinction phenomenon which constitutes one major difference between our system and conventional heterostructure systems. A general extinction rule has also been obtained.

This work was supported by the National Natural Science Foundation of China.

*To whom all communications regarding this paper should be addressed.

[1]F. Nori and J. P. Rodriguez, Phys. Rev. B **34**, 2207 (1986).

[2]S. Tamura and J. P. Wolfe, Phys. Rev. B **36**, 3491 (1987).

[3]Y. Y. Zhu, N. B. Ming, and W. H. Jiang, Phys. Rev. B **40**, 8536 (1989).

[4]R. Merlin, K. Bajema, R. Clarke, F. Y. Juang, and P. K. Bhattacharya, Phys. Rev. Lett. **55**, 1768 (1985).

[5]X. K. Zhang, H. Xia, G. X. Cheng, A. Hu, and D. Feng, Phys. Lett. A **136**, 312 (1989).

[6]D. Feng, N. B. Ming, J. H. Hong, Y. S. Yang, J. S. Zhu, Z. Yang, and Y. N. Wang, Appl. Phys. Lett. **37**, 607 (1980).

[7]Y. H. Xue, N. B. Ming, J. S. Zhu, and D. Feng, Acta Phys. Sin. **32**, 1515 (1983) [Chin. Phys. **4**, 554 (1984)].

[8]J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Phys. Rev. **127**, 1918 (1962).

[9]N. Bloembergen and A. J. Sievers, Appl. Phys. Lett. **17**, 483 (1970).

[10]A. Yariv and P. Yeh, *Optical Waves in Crystals* (Wiley, New York, 1984).

[11]N. B. Ming, J. F. Hong, Z. M. Sung, and Y. S. Yang, Acta Opt. Sin. **4**, 821 (1981).

[12]N. B. Ming, J. F. Hong, and D. Feng, J. Mater. Sci. **27**, 1663 (1982).

[13]J. F. Hong and Y. S. Yang, Opt. Sin. **4**, 821 (1986).

[14]F. Zernike and J. M. Midwinter, *Applied Nonlinear Optics* (Wiley, New York, 1973).

[15]D. Levine and P. J. Steinhardt, Phys. Rev. B **34**, 596 (1986).

[16]R. K. P. Zia and W. J. Dallas, J. Phys. A **18**, L341 (1985).

[17]V. Hoggatt, *Fibonacci and Lucas Numbers* (Houghton-Mifflin, Boston, 1969).

[18]Smith, Opt. Commun. **17**, 332 (1976).