

Properties of the peaks of second harmonic light through Fibonacci-class ferroelectric domains

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Abstract. After establishing the method of constructing a class of one-dimensional (1D) Fibonacci-class quasiperiodic (FC(n)) ferroelectric domains system, we have studied the properties of the electric field of the second harmonic generation (SHG) by means of the small-signal approximation in the case of the vertically transmission. It was found that only the second harmonic light (SHL) peaks which were indexed by two special integers q and p would be the brightest and the spectra whose positions were decided by successive FC(n) integers q_F and p_F were perfect self-similar without considering the dispersive effect of the refractive index on SHL. The effect of the vacancies for some special spectral lines was also studied generally. The analytic results were confirmed by the numerical simulations.

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1 Introduction

There have been a large number of studies on the properties of the quasiperiodic systems since the experimental discovery of fivefold symmetry in the diffraction pattern of metallic alloys [1]. Particularly, from this on, much attention has been paid to 1D FC(1) or the superlattice systems constructed following the FC(1) sequence because of its structure's not only having the main characteristics of the quasicrystals but also being relatively very simple. In 1985, Merlin *et al.* [2] presented the X-ray and Raman scattering measurements to the sample, consists of alternating layers of GaAs and AlAs to form FC(1) sequence in which the ratio of incommensurate periods is equal to the golden mean τ . Then Nori and Rodriguez [3] studied the acoustic and electronic properties of 1D quasicrystals. The quasiperiodic structures with any number of layers and several types of boundary conditions were studied there. Afterwards, Tamura and Wolfe [4] studied acoustic-phonon transmission through a realistic Fibonacci superlattice theoretically and obtained some results for the transmission spectra. Zhu and Ming [5] analyzed a Fibonacci optical superlattice which is made from a single crystal with quasiperiodic laminar ferroelectric domain structures and studied SHG in this system. Zhu *et al.* [6] fabricated a nonlinear optical superlattice of LiTaO₃ in which two building blocks A and B were ar-

ranged as a Fibonacci sequence and measured the quasi-phase-matched (QPM) SHG spectrum of it.

Recently Fu *et al.* [7] proposed and studied a 1D quasilattice model which was more general than 1D Fibonacci one and was called "Fibonacci-class quasilattices" (FC(n)). They proved that the electronic energy spectra of FC(n) were perfect self-similar. Based on Kohmoto *et al.*'s studies [8] on the transmission of light through dielectric multilayers arranged following 1D FC(1) chain, and based on Huang *et al.*'s results [9] in the case of the multilayers constructed following intergrowth quasiperiodic sequence (*i.e.* FC(2)), we [10] have recently studied the common situation for FC(n) in detail and have found the very useful switchlike property of the transmission coefficient.

Being the theoretical basis of the application of multi-wavelength SHL devices, naturally, it is a very interesting problem to study spectral peaks of SHL through ferroelectric domains constructed following FC(n), the general new model, sequence. We shall study here the light transmission in a FC(n) superlattice. First, we introduced the characteristic of the construction of FC(n) in Section 2. Then we studied the electric field $E_2(x)$ of SHG in Section 3 analytically. Section 4 is devoted to study the rule of SHL's brightest spectra, in which the perfect self-similarity was found, but the formula we got is different from that of Merlin [2], Tamura [4] and Zhu [5]. In Section 5, we investigated the effect of the vacancies of SHL spectral lines. A brief summary was given in Section 6.

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2 The characteristic of FC(n)'s construction

From the origin, the position of the N th atom in FC(n) sequences obtained by means of direct projection method [7] is given by

$$x_N = N \cos \theta + \sin \theta \lfloor N \operatorname{tg} \theta \rfloor, \quad (1)$$

where $\lfloor \cdot \rfloor$'s represent the greatest integer function, and where

$$\operatorname{tg} \theta = \sigma_n = \frac{\sqrt{n^2 + 4} - n}{2} \quad (2)$$

is the positive root of the characteristic equation

$$x^2 + nx - 1 = 0. \quad (3)$$

FC(n) can also be generated by the substitution rules $B \rightarrow B^{n-1}A$, $A \rightarrow B^{n-1}AB$. Starting with a B, the first three generations of FC(n) are

$$\begin{aligned} S_1 &= B, \\ S_2 &= B^{n-1}A, \\ S_3 &= \underbrace{(B^{n-1}A)(B^{n-1}A)\dots(B^{n-1}A)}_{n-1} B^{n-1}AB, \end{aligned} \quad (4)$$

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which show the following recurring relation

$$S_j = S_{j-1}^n S_{j-2} \quad (j \geq 3). \quad (5)$$

In this paper we obtained FC(n) by indirect projection method from a two-dimensional (2D) square lattice, and the position of the N th atom on the projection line from the origin is given by

$$x_N = N + \frac{1}{\varphi_n} \lfloor \frac{N}{\varphi_n} \rfloor, \quad (6)$$

where

$$\varphi_n = \frac{1}{\sigma_n} = \frac{\sqrt{n^2 + 4} + n}{2} \quad (7)$$

is an irrational quadratic number and the positive root of the characteristic equation

$$x^2 - nx - 1 = 0. \quad (8)$$

3 The electric field of SHL

In order to manufacture the LiNbO₃ sample easily and compute the characteristic of the system conveniently, we made the two ferroelectric domains A and B each composed of two layers of different properties, which were shown in Figure 1. We set that

$$\begin{aligned} l_A/l_B &= \varphi_n, \\ l_{A1} &= l, \quad l_{A2} = l(1 + \eta), \\ l_{B1} &= l, \quad l_{B2} = l(1 - \varphi_n \eta), \end{aligned} \quad (9)$$

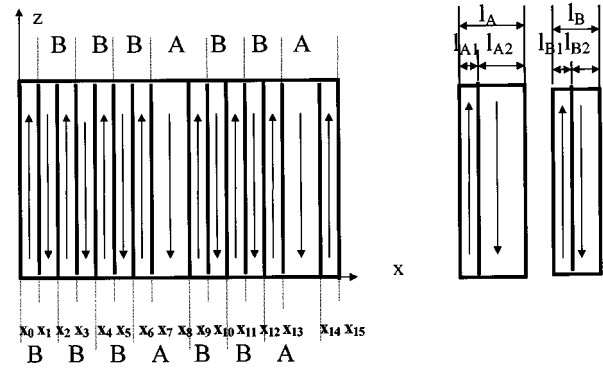


Fig. 1. The ferroelectric multilayers constitute a FC(3) system, where the directions of the arrows denote of the electric field's polarizations. (a) The schematic diagram of total ferroelectric superlattice, where the coordinates $\{x_N\}$ are the positions of the ferroelectric domain boundaries; (b) the structures of the two kinds of building blocks A and B.

where l is the structure parameter, and where η satisfies

$$\eta = \frac{2(\varphi_n - 1)}{1 + \varphi_n^2} = \frac{2(\varphi_n - 1)}{2 + n\varphi_n} = \frac{3\sqrt{n^2 + 4} + n - 6}{2n + 3}. \quad (10)$$

Generally, η is responsible for the arrangement to be periodic (when $\eta = 0$) or quasiperiodic (when $\eta \neq 0$).

In the case of SHL with a single laser beam incident onto the surface of FC(n) ferroelectric, the electric field of fundamental beam E_1 and that of SHG E_2 satisfy the wave equation under the small-signal approximation [5,11]:

$$\frac{dE_2(x)}{dx} = \frac{-i32\pi\omega^2}{k^{2\omega}c^2} d(x) E_1^2 e^{i(k^{2\omega} - 2k^\omega)x}, \quad (11)$$

where

$$d(x) = \begin{cases} d_{33}, & \text{in positive ferroelectric domains;} \\ -d_{33}, & \text{in negative ferroelectric domains;} \end{cases} \quad (12)$$

and ω is the angular frequency of the fundamental beam; and k^ω and $k^{2\omega}$ are wave numbers of fundamental beam and that of SHL, respectively; and c is the speed of light in vacuum.

After passing through N FC(n) ferroelectric layers then SHL electric field E_2 can be given as

$$E_2(N) = \frac{-64\pi\omega^2}{k^{2\omega}c^2\Delta k} d_{33} E_1^2 \left(\sum_{j=0} e^{i\Delta k x_{2j+1}} - \sum_{j=0} e^{i\Delta k x_{2j}} \right), \quad (13)$$

where

$$\Delta k = k^{2\omega} - 2k^\omega, \quad (14)$$

generally it satisfies [12]

$$|\Delta k| = |k^{2\omega} - 2k^\omega| = \frac{4\pi}{\lambda} (n_2 \cos \theta_2 - n_1 \cos \theta_1), \quad (15)$$

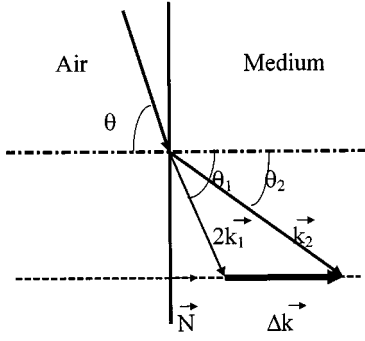


Fig. 2. Schematic diagram of the relationship between SHG wave number vector and that of fundamental beam.

shown as Figure 2, where n_2 and n_1 are the refractive indices of SHL and fundamental beam, respectively, and where θ_2 and θ_1 are the refractive angles of SHL and fundamental beam, respectively. In the vertically transmission case of $\theta = \theta_1 = \theta_2 = 0$, we have

$$\Delta k = |\Delta \mathbf{k}| = \frac{4\pi}{\lambda}(n_2 - n_1). \quad (16)$$

It was well-known that [13]

$$\lfloor x \rfloor = x - \{x\}, \quad (17)$$

where the function $\{x\}$ is periodic in x with period 1, then formula (6) can be reexpressed as

$$x_N = N + \frac{1}{\varphi_n} \left(\frac{N}{\varphi_n} - \left\{ \frac{N}{\varphi_n} \right\} \right). \quad (18)$$

From formula (7), it was obviously to see that

$$\varphi_n \left(1 + \frac{1}{\varphi_n^2} \right) = \sqrt{n^2 + 4}. \quad (19)$$

Submitting equation (19) into formula (18) one can obtain

$$\begin{aligned} x_N &= N \left(1 + \frac{1}{\varphi_n^2} \right) - \frac{1}{\varphi_n} \left\{ \frac{N(1 + \frac{1}{\varphi_n^2})}{\sqrt{n^2 + 4}} \right\} \\ &= Na + F(Na), \end{aligned} \quad (20)$$

where

$$a = 1 + \frac{1}{\varphi_n^2}, \quad F(x) = -\frac{1}{\varphi_n} \left\{ \frac{x}{\sqrt{n^2 + 4}} \right\}. \quad (21)$$

In the reciprocal space, the Fourier transform of the atomic positions defined by equation (20) will consist of Bragg peaks [14] at positions

$$k_{pq} \equiv \frac{2\pi}{1 + \frac{1}{\varphi_n^2}} \left(p + \frac{q}{\varphi_n} \right), \quad (22)$$

where p and q are integers.

For the finite FC(n) multilayers, we can conclude that the integration

$$\begin{aligned} I(x_n) &= \sum_n e^{ikx_n} \\ &\approx \sum_{pq} e^{iX_{pq}} \frac{\sin X_{pq}}{X_{pq}} \delta \left[k - \frac{2\pi(q + p\varphi_n)}{D} \right], \end{aligned} \quad (23)$$

where

$$X_{pq} = \frac{\pi\varphi_n}{1 + \varphi_n^2} (q\varphi_n - p), \quad (24)$$

and where

$$D = \varphi_n + \frac{1}{\varphi_n} \quad (25)$$

and has the dimension of a length. We define l_A as a unit length, for the symmetry of the system is not determined by the absolute thicknesses of layer A or layer B but by the order of the arrangement of ferroelectric domains, so only did the relative thicknesses between layer A and layer B which satisfy equation (9) have to be thought of, then we have

$$D = \varphi_n l_A + l_B. \quad (26)$$

Now it is easily to obtain the following result by the use of equation (23)

$$\begin{aligned} I(x_{2j+1}) &= \sum_{j=0} e^{ikx_{2j+1}} \\ &\approx \sum_{pq} e^{iX_{pq}} \frac{\sin X_{pq}}{X_{pq}} \delta \left[k - \frac{2\pi(q + p\varphi_n)}{D} \right]. \end{aligned} \quad (27)$$

In Figure 1 there exists

$$x_{2j} = x_{2j+1} - l_{A1} = x_{2j+1} - l, \quad (28)$$

we can obtain the value of the sum of x_{2j} as

$$I(x_{2j}) = e^{-ikl} I(x_{2j+1}). \quad (29)$$

By means of the integrations in equations (27, 29), then we can rewrite formula (13) as

$$\begin{aligned} E_2(N) &\approx -\frac{128\pi\omega^2}{k^2\omega c^2\Delta k} (d_{33}E_1^2) e^{\frac{i}{2}(\pi - \Delta kl)} \sin \left(\frac{\Delta kl}{2} \right) \\ &\times \sum_{pq} e^{iX_{pq}} \left(\frac{\sin X_{pq}}{X_{pq}} \right) \delta \left[\Delta k - \frac{2\pi(q + p\varphi_n)}{D} \right]. \end{aligned} \quad (30)$$

This is the analytical formula of the electric field for SHG after passing through N layers of FC(n) ferroelectric domains.

4 The rule for SHL's intense spectra

From formula (30), for the existence of δ function factor one knows that the peaks of SHG field E_2 occur for those

$$\Delta k = \frac{2\pi(q + p\varphi_n)}{D}. \quad (31)$$

In the real space we make the wavelength of fundamental beam λ_0 , the refractive indices of SHL n_{20} and that of fundamental beam n_{10} keep constant, *i.e.*, the effect of the dispersion of refractive indices on SHL was not taken into account here. By the use of equations (31, 26, 16) one can get the formula of structure parameter l of SHL peaks

$$l_{pq} = \frac{\lambda_0(q + p\varphi_n)}{4(n_{20} - n_{10})(1 + \varphi_n)}. \quad (32)$$

We order

$$f(x) = \frac{\sin x}{x}. \quad (33)$$

It was well-known that when $f^{-1}(x) = 0$ and $f^{-2}(x) < 0$, $f(x)$ gets the maximums. Then one can obtain the following conditions for the most intensive SHL peaks which were indexed by two special integers q and p , where q and p satisfy

$$X_{pq} = \text{tg}(X_{pq}); \quad (34)$$

and

$$\cos(X_{pq}) > 0, \quad (35)$$

where q and p are the integers determined by equation (31).

On the other hand, particularly,

$$\text{if } x \rightarrow 0, \text{ then } f^{-1}(x) \rightarrow 0, \quad (36)$$

some of the SHL intense peaks occur at the positions

$$X_{pq} \rightarrow 0, \quad \text{or} \quad \frac{p}{q} \rightarrow \varphi_n. \quad (37)$$

Similar to the case of FC(1) chain [15], it was demonstrated by Fu *et al.* [7] that p/q would be the best rational approximations to φ_n only when p and q were successive FC(n) integers, *i.e.*,

$$(p, q) = (p_F, q_F) = (\text{FC}(n)_h, \text{FC}(n)_{h-1}) \quad (h \geq 2), \quad (38)$$

and $(\text{FC}(n)_2, \text{FC}(n)_1) = (n, 1)$,

where h is the number of FC(n)'s generation, then it can be proved that

$$q_F + p_F\varphi_n = \varphi_n^h \quad (h \geq 2). \quad (39)$$

In the case of $n = 1$, equations (38, 39) change to

$$(p, q) = (p_F, q_F) = (F_h, F_{h-1}) \quad (h \geq 2), \quad (40)$$

and $(F_2, F_1) = (1, 1)$,

$$q_F + p_F\tau = \tau^h \quad (h \geq 2). \quad (41)$$

The formula got by Tamura *et al.* [4] is $q_{m,n} = \frac{\pi(m + n\tau)}{d} = \frac{(\text{integer}) \times \pi\tau^p}{d}$, where $(m, n) = (F_{p-1}, F_p)$, $(F_0, F_1) = (0, 1)$, *i.e.*

$$m + n\tau = (\text{integer}) \times \tau^p \quad (h \geq 1). \quad (42)$$

The equations got by Merlin *et al.* [2] are $k = 2\pi d^{-1}(m + m'\tau)$ and $k_{p,n} = 2\pi d^{-1}n\tau^p$, where n and p are integers, *i.e.*

$$p + n\tau = n\tau^p. \quad (43)$$

The results Zhu *et al.* [5] obtained are that $l_{m,n} = \frac{(m + n\tau)\lambda_0}{4(n_{20} - n_{10})(1 + \tau)}$ and $l(s, p) = \frac{\lambda_0}{4(n_{20} - n_{10})(1 + \tau)}s\tau^p$, where $(m, n) = (F_{k-1}, F_k)$, $(F_0, F_1) = (0, 1)$, *i.e.*

$$s + p\tau = s\tau^p. \quad (44)$$

Obviously, equations (41, 42, 43, 44) are not the same.

Submitting equation (39) into equation (32), we can obtain the formula of structure parameter l of the intense SHL F-peaks

$$l_{p_F q_F} = l_{F_h, F_{h-1}} = l_h = \frac{\lambda_0 \varphi_n^h}{4(n_{20} - n_{10})(1 + \varphi_n)} \quad (h \geq 2). \quad (45)$$

From equation (45), it can be demonstrated that there exist self-similarity for the intense F-spectra of the SHL fields by the rule

$$l_h + nl_{h+1} = l_{h+2} \quad (h \geq 1). \quad (46)$$

This was shown schematically in Figure 3, and can also be confirmed numerically by the data in Table 1, *e.g.*,

for FC(1) ($n = 1$), $l_{(2,3)} + l_{(3,5)} = l_4 + l_5 = l_{(5,8)} = l_6$;

for FC(4) ($n = 4$),

$$l_{(0,1)} + 4 \times l_{(1,4)} = l_1 + 4 \times l_2 = l_{(4,17)} = l_3$$

for FC(4) ($n = 4$),

$$l_{(4,17)} + 4 \times l_{(17,72)} = l_3 + 4 \times l_4 = l_{(72,305)} = l_5.$$

In the reciprocal space, similar to the case of the real space, the conclusion in equation (46) is also valid without dispersion effect or for the linear phenomena and can be rewritten as

$$\left(\frac{1}{\lambda}\right)_h + n \left(\frac{1}{\lambda}\right)_{h+1} = \left(\frac{1}{\lambda}\right)_{h+2} \quad (h \geq 1). \quad (47)$$

But if the dispersive effect of the refractive index on SHL spectra has to be considered, then $n_i = n_i(\lambda) \neq c_0 n_{i0} \neq n_{i10}$ (where c_0 and n_{i10} are all constant). By the use of

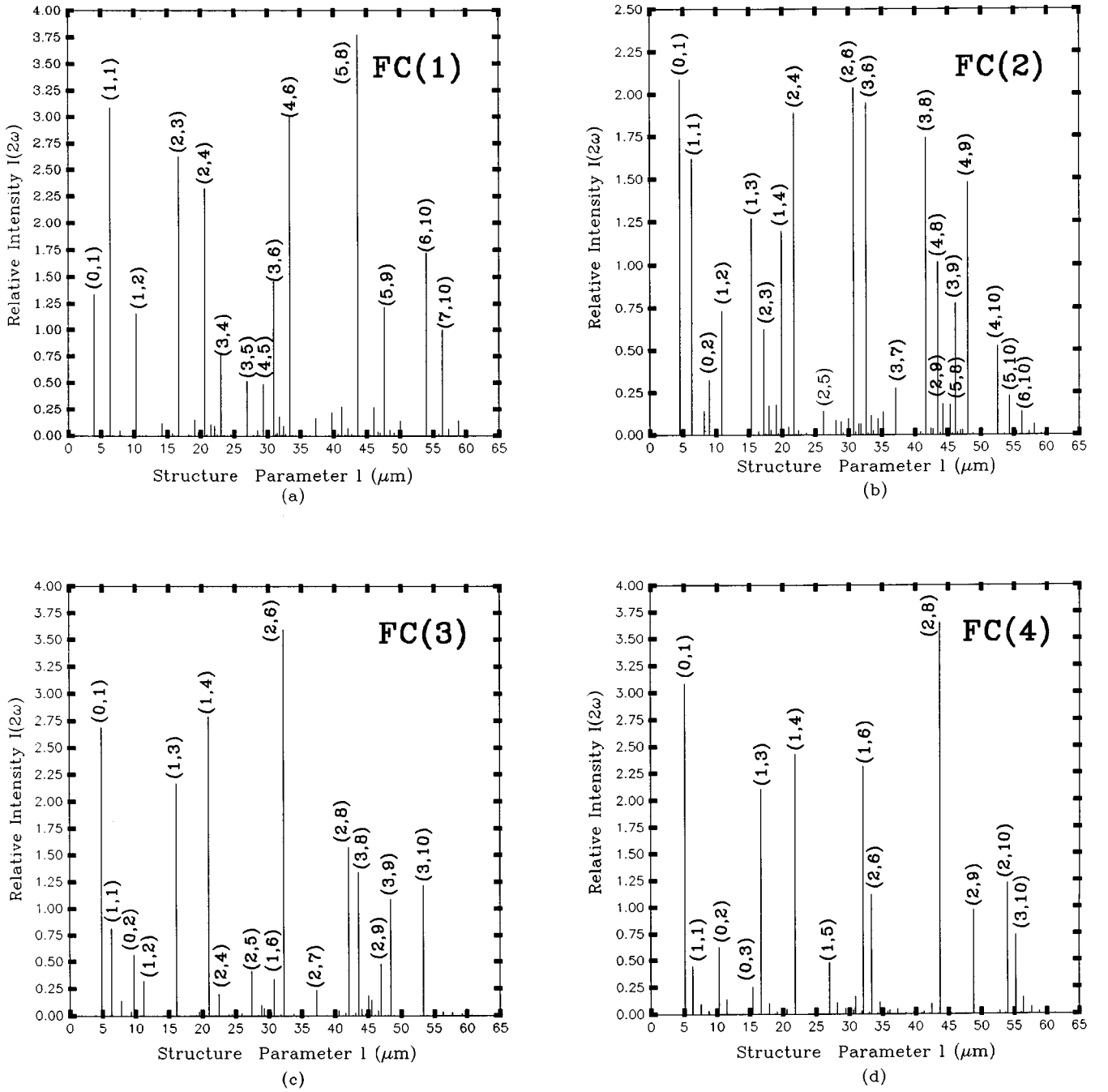


Fig. 3. In real space the figure of SHG relative intensity $I(2\omega)$ vs. structure parameter l , where for LiNbO_3 $\lambda_0 = 1.318 \mu\text{m}$, $n_{10} = 2.1453$, and $n_{20} = 2.1970$.

equation (45) it is not very difficult to obtain the following two formulae:

$$l_h + nl_{h+1} \neq l_{h+2} \quad (h \geq 1), \quad (46a)$$

$$\left(\frac{1}{\lambda}\right)_h + n\left(\frac{1}{\lambda}\right)_{h+1} \neq \left(\frac{1}{\lambda}\right)_{h+2} \quad (h \geq 1), \quad (47a)$$

i.e., the self-similarity will be absent either in the real space or in the reciprocal one.

5 The effect of the vacancies of SHL spectral lines

From equation (30) it can be easily deduced that some SHL spectral lines would be disappeared when

$$\sin \frac{\Delta kl}{2} = 0$$

i.e.

$$\frac{\Delta kl}{2} = m\pi, \quad (48)$$

Table 1. The following are the data of the disappeared spectra (DS) and SHG's intense peaks (IP), where h is the number of FC(n)'s generation, and where I_{E_2} is the relative intensity of SHG electric field E_2 , and where the brackets in the third and sixth column mean "quasi".

Fibonacci-class quasilattices	(q, p)	h	l	I_{E_2}	characteristic
FC(1)	(0,1)	1	3.9389	1.34276	IP
	(1,1)	2	6.3733	3.09050	IP
	(1,2)	3	10.3122	1.15743	IP
	(2,1)		8.8077	0.00000	DS
	(2,2)		12.7466	0.00000	DS
	(2,3)	4	16.6855	2.62629	IP
	(2,4)	(3)	20.6244	2.32939	(IP)
	(3,3)	(2)	19.1199	0.15408	(IP)
	(3,5)	5	26.9977	0.51795	IP
	(3,6)	3	30.9366	1.46137	IP
	(4,2)		17.6153	0.00000	DS
	(4,4)	(2)	25.4932	0.00000	(IP),DS
	(4,6)	(4)	33.3710	2.99407	(IP)
	(4,8)	(3)	41.2488	0.27866	(IP)
	(5,5)	(2)	31.8665	0.18434	(IP)
	(5,8)	6	43.6832	3.77331	IP
	(5,10)	(3)	51.5610	0.00217	(IP)
	(6,3)		26.4230	0.00000	DS
	(6,6)	(2)	38.2398	0.00000	(IP),DS
	(6,10)	(5)	53.9954	1.72516	(IP)
	(7,7)	2	44.6131	0.00449	IP
	(8,4)		35.2307	0.00000	DS
	(8,8)	(2)	50.9863	0.00000	(IP),DS
	(10,5)		44.0384	0.00000	DS
	(10,10)	(2)	63.7329	0.00000	(IP),DS
	FC(4)	(0,1)	1	5.1561	3.08832
(1,4)		2	21.8416	2.43193	IP
(2,2)			12.7466	0.00000	DS
(2,4)			23.0588	0.00000	DS
(2,8)		(2)	43.6832	3.65649	(IP)
(4,4)			25.4932	0.00000	DS
(4,8)			46.1176	0.00000	DS
(4,17)		3	92.5225	2.10708	IP
(6,6)			38.2398	0.00000	DS
(8,3)			25.2058	0.00000	DS
(8,8)			50.9864	0.00000	DS
(10,10)			63.7329	0.00000	DS
(17,72)		4	391.9316	2.02549	IP
(72,305)	5	1660.2489	2.00608	IP	

where m is integer. Submitting equation (31) into equation (48), then one can get

$$q + p\varphi_n = 2m(1 + \varphi_n). \quad (49)$$

For the irrational quadratic number φ_n and the integers q and p , we have

$$q = p = 2m, \quad (50)$$

where m is integer. Therefore SHL spectral lines satisfying equation (50) will be disappeared, this is the effect of the vacancies of SHL spectral lines. But for the intense F-peaks there exist the relation of equation (39), and for arbitrary integer m

$$\varphi_n^h - 2m(1 + \varphi_n) \neq 0, \quad (h \geq 2) \quad (51)$$

so the phenomena of the effect of SHL spectral lines' vacancies only occur at the positions of SHL weak peaks

($2m, 2m$) ($m = 0, \pm 1, \pm 2, \dots$), the numerical results were partly shown in Table 1.

6 Brief summary

In Section 2, we introduced the characteristic of the construction of FC(n). Then we have studied the properties of SHL's electric field by means of the small-signal approximation in the case of the vertically transmission in detail. It was found that only SHL peaks which were indexed by two special integers q and p , where q and p satisfy equations(34, 35, 31), would be the most intensive ones and the spectra whose positions were decided by successive Fibonacci-class integers q_F and p_F were perfect self-similar without considering the dispersive effect of the refractive index on SHL or for the linear optical phenomena. The effect of the vacancies for some special spectral lines was also studied generally. The analytic results were confirmed by the numerical simulations.

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