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Nonlinear optical characterization of the generalized Fibonacci optical superlattices and their 'isotopes'

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Abstract. Nonlinear optical characterization of superlattices generated by the production rule $S_j = mS_{j-1} + S_{j-2}$ with the initial conditions $S_1 = A$ and $S_2 = A^nB$ have been studied theoretically. For each m and n, the real space structure is quite different. However superlattices with the same m but different n have the same reciprocal space structure, thus forming an 'isotope' family. The result shows the promising applications of these superlattices in nonlinear optical frequency conversion.

Usually, efficient nonlinear optical frequency conversion can be realized by birefringence phase matching the interacting waves in uniaxial and biaxial crystals [1]. In the case where birefringence phase matching is impossible, a modulated structure can be constructed. Its period equals twice the coherence length determined by the index dispersion of the interacting waves. In this case, the phase mismatch of the interacting waves is compensated by the reciprocal vectors provided by the structure. Fourier analysis of the structure indicates that each reciprocal vector is related to an effective nonlinear optical coefficient, some of which are large and some are small. For quasi-phase-matched (QPM) efficient frequency conversion only those reciprocal vectors with large effective nonlinear optical coefficients are preferred. This method is known as quasi-phase-matching [2]. A material suitable for this purpose is one with ferroelectric domain inverted structures [3–6], which may be termed an optical superlattice (OSL). The periodic OSL is the mostly used for nonlinear optical frequency conversion, such as the second-harmonic generation (SHG), optical parametric process etc [7–16]. Recently interest has been aroused in the high-harmonic generation, such as the third-harmonic generation (THG) and the fourth-harmonic generation, which can be obtained through two nonlinear parametric processes [17–19]. For example, TH involves an SHG process and a sum frequency generation (SFG) process. Thus, in the QPM regime, two reciprocal vectors are needed: one used for QPM SHG and the other for QPM SFG. One candidate for the THG is a quasiperiodic OSL which can provide many more reciprocal vectors than the periodic OSL. Experiments for THG have been performed in a Fibonacci quasiperiodic OSL LT and SBN [18, 19]. Numerical calculations for THG have been conducted in OSLs with Thue–Morse structure, three-component Fibonacci structure, intergrowth structure etc [20–22]. However, since the THG depends not only on the QPM but also on the magnitude of the effective nonlinear optical coefficients, the fundamental wavelengths at which TH can be generated with high conversion efficiency is limited with these structures. Thus it is necessary to investigate the nonlinear optical characterization of OSLs with structures other than the structures mentioned above. By nonlinear optical characterization, here we are mainly concerned with the distribution of reciprocal vectors in reciprocal space, which is essential for the QPM and the magnitude of the related Fourier components, which is important for the conversion efficiency.

In this paper, the nonlinear optical characterization of the generalized Fibonacci superlattices has been studied theoretically. By exchanging the initial conditions, each generalized Fibonacci superlattice can be expanded into a whole family having the same reciprocal space structure. The magnitude of the Fourier components depends on which family the superlattice belongs to and on the corresponding structure parameters. This greatly facilitates the design of superlattices for applications.

The superlattices discussed in this paper can be generated by the production rule

$$S_j = mS_{j-1} + S_{j-2} \tag{1}$$

with the initial conditions: $S_1 = A$ and $S_2 = A^n B$, where $mS_{j-1} + S_{j-2}$ means a sequence of m $(j-1)$th generation followed by the $(j-2)$th generation. For each superlattice, there are two characteristic numbers. One, defined as σ, is the limit value of the ratio of the total numbers of types A and B in two subsequent generations

$$\sigma(m) = \lim_{j\to\infty} \frac{S_j}{S_{j-1}}. \tag{2}$$

From the production rule, we can obtain

$$\sigma(m) = \frac{m + \sqrt{m^2+4}}{2}. \tag{3}$$

The other one, defined as γ, is the ratio of A to B in the same generation when $j \to \infty$

$$\gamma(m,n) = \frac{1}{\sigma} + n = \delta + (n-m). \tag{4}$$

For $n = m$ the OSL becomes the generalized Fibonacci superlattice [23, 24] and γ equals σ, whereas for $n \neq m$ these two numbers are totally different. However, for a fixed value of m, for example, $m = 2$ with different values of n, σ and γ are all related to an irrational number $\sqrt{2}$. In fact, the superlattices with the same m but different n can be related to one another by the so-called deflation rule. Usually, the deflation rule is defined as AB $\to$ A and A $\to$ B. However, in this paper, for the convenience of discussion, we define the following deflation rule: AB $\to$ B and A $\to$ A. For $m = 2$ and $n = 2$, the silver Fibonacci superlattice is

AABAABAAABAABAAABAABAABAAABAABAAABAABAABA . . . .

The two characteristic numbers are

$$\sigma(2) = \gamma(2,2) = 1 + \sqrt{2}.$$

By a deflation, it becomes $m = 2$ and $n = 1$

ABABAABABAABABABAABABAABABAB . . . .

The characteristic number γ becomes

$$\gamma(2,1) = \sigma + n - m = \sqrt{2}.$$

The second deflation changes the above sequence into a sequence with $m = 2$ and $n = 0$

BBABBABBBABBABBB . . . .

The characteristic number γ becomes

$$\gamma(2,0) = \sigma + n - m = \sqrt{2} - 1$$

which is just the reciprocal of $\gamma(2,2)$. A careful comparison reveals that the sequences with $n = 0$ and $n = 2$ are the same except that A $\to$ B and B $\to$ A. This interesting phenomenon

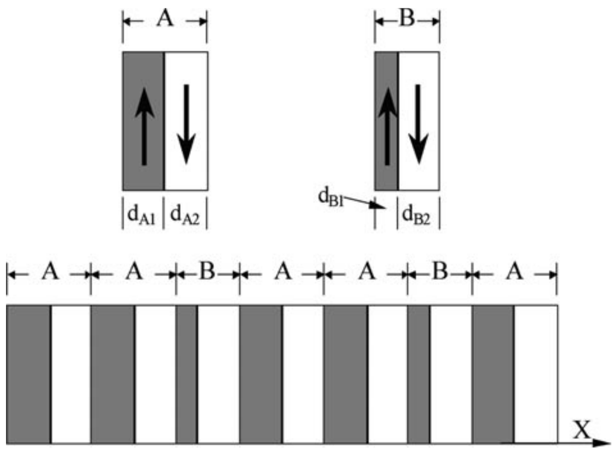

Figure 1. Silver Fibonacci quasiperiodic superlattice. (a) Two building blocks: A and B, each composed of one positive and one negative ferroelectric domain. (b) Schematic diagram of a silver QPOSL with $d_{A2} = d_{B2}$.

happens to other sequences. For example, for the golden Fibonacci superlattice, one deflation changes it to itself. The characteristic number $\gamma(1, 0) = (\sqrt{5} - 1)/2$, which is the reciprocal of $\gamma(1, 1) = (\sqrt{5} + 1)/2$. For $m = 3$ the copper Fibonacci superlattice, after three deflations, returns to itself. The characteristic number $\gamma(3, 0) = (\sqrt{13} - 3)/2$, which is the reciprocal of $\gamma(3, 3) = (\sqrt{13} + 3)/2$. The same discussion holds for $m > 3$. Thus, by successive deflation, each generalized Fibonacci superlattice forms a cycle.

In real space, each superlattice with the superlattices generated by deflation rule appears quite different. However, from the above discussion, these superlattices are not isolated but closely related to one another. Then one may ask what their reciprocal spaces look like. Are there any relations among these reciprocal spaces? These are of fundamental interest in physics and crucial to the QPM frequency conversion.

Let us start our discussion with the generalized Fibonacci quasiperiodic optical superlattices (QPOSLs) we are interested in. For QPM frequency conversion, a typical generalized Fibonacci ferroelectric QPOSL is constructed from two building blocks, A and B, each composed of a pair of oppositely polarized domains. The thicknesses of the positive ferroelectric domain in blocks A and B are d_{A1} and d_{B1}, respectively. The thickness of the negative ferroelectric domain in blocks A is d_{A2}, and that in block B is d_{B2}. Figure 1 is a schematic diagram of the silver Fibonacci superlattice. As second-order nonlinear coefficients form a third-rank tensor, they change signs from the positive domains to the negative domains. For an infinite array, the quasi-periodically modulated nonlinear coefficient $d(x)$ can be written, by use of the Fourier transform, as

$$d(x) = \sum_{l,p} d_{l,p}(m) \exp\{\mathrm{i}G_{l,p}(m)x\}$$

$$d_{l,p}(m) = \frac{2[1+\gamma(m)]d_0}{\gamma(m)d_A + d_B} \frac{\sin\{G_{l,p}(m)d_0/2\}}{\{G_{l,p}(m)d_0/2\}} \frac{\sin(X_{l,p})}{(X_{l,p})} d_{33} \tag{5}$$

where

$$G_{l,p}(m) = 2\pi[l + p\gamma(m)]/D \tag{6}$$

is the reciprocal vector and $D = \gamma(m)d_A + d_B$ is the average parameter of the QPOSL, $X_{l,p} = \pi[1+\gamma(m)](ld_A - pd_B)/D$, l and p are two integers, $d_0 = d_{A2} = d_{B2}$, $d_A = d_{A1} + d_{A2}$

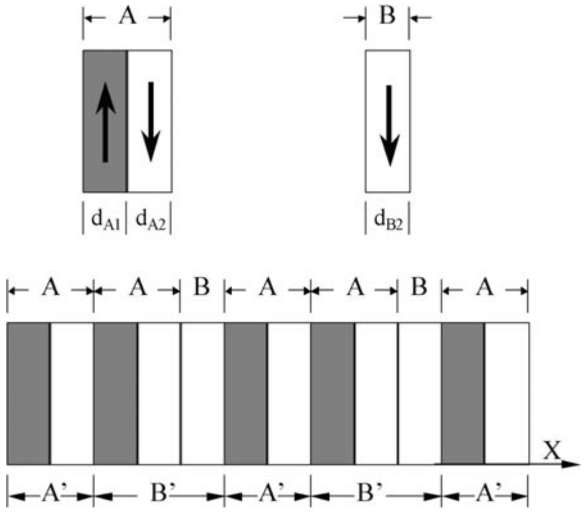

Figure 2. The limit case of the silver Fibonacci superlattice with $d_{B1} = 0$, which corresponds to the deflated silver Fibonacci quasiperiodic superlattice. (a) Two building blocks: A composed of one positive and one negative ferroelectric domain; B composed of only one negative domain. (b) Schematic diagram of the deflated silver QPOSL with $d_{A'1} = d_{A1}$, $d_{A'2} = d_{A2}$, $d_{B'1} = d_{A'1}$ and $d_{B'2} = 2d_{B2}$.

and $d_B = d_{B1} + d_{B2}$. It can be seen that in reciprocal space, the irrational number $\gamma(m)$ plays an important role, which results in a densely filled reciprocal space. It can also be seen that the reciprocal vector is dependent on the average parameter D. When the value of D is fixed, the distribution of $G_{l,p}$ in reciprocal space is determined. However, even if D is fixed, d_0, d_A and d_B can still be varied in a certain range, which results in the variation of $d_{l,p}$. Because of this, it is more flexible to use the generalized Fibonacci superlattice for high-harmonic generation where more than one reciprocal vectors are needed for the QPM conditions.

In figure 1, if we set $d_{B1} = 0$, then each block B will be merged with a block A just ahead of it to form a new one, which can be seen in figure 2. Now the structure parameters become $d_{A'1} = d_{A1}$, $d_{A'2} = d_{A2}$, $d_{B'1} = d_{A'1}$ and $d_{B'2} = 2d_{B2}$. This is equivalent to the deflation. That is, figure 2 is just the limiting case of figure 1. In view of this, equations (5) and (6) can be used for both cases. In fact, the structure shown in figure 2 is the so-called 'intergrowth structure' [25] which can be obtained by the projection method. According to Zia and Dallas [26], the Fourier transformation for those structures obtainable from the projection method can be expressed as equation (5). For the deflated silver superlattice or the intergrowth structure, $\gamma(2, 1) = \sqrt{2}$ and $D = \sqrt{2}d_{A'} + d_{B'}$. Note that $d_{B'} = d_A + d_B$, the reciprocal vectors and the magnitude of the Fourier components for these two structures can be expressed by the same equation with the following substitution: $d_{l,p}(m) \rightarrow d_{l,p}(m, n)$, $\gamma(m) \rightarrow \gamma(m, n)$ and $G_{l,p}(m) \rightarrow G_{l,p}(m, n)$. Here, by deflation, n is always less than m. However, the results can be extended to superlattices with n larger than m. These superlattices can be generated by the inflation rule: A $\rightarrow$ A an B $\rightarrow$ AB. Obviously, the superlattice with $n = m$ is the limiting case of the superlattice with $n = m + 1$, and $n = m + 1$ is the limiting case of $n = m + 2$, and so on. Therefore, superlattices with the same m can be classified into one family. Those with larger n may be termed the ancestors and those with smaller n the descendants. The reciprocal space of one family has the same structure, each related to an irrational number. For the gold family, this number is $(1 + \sqrt{5})/2$; for the silver family, $1 + \sqrt{2}$, for the copper family, $(3 + \sqrt{13})/2$,

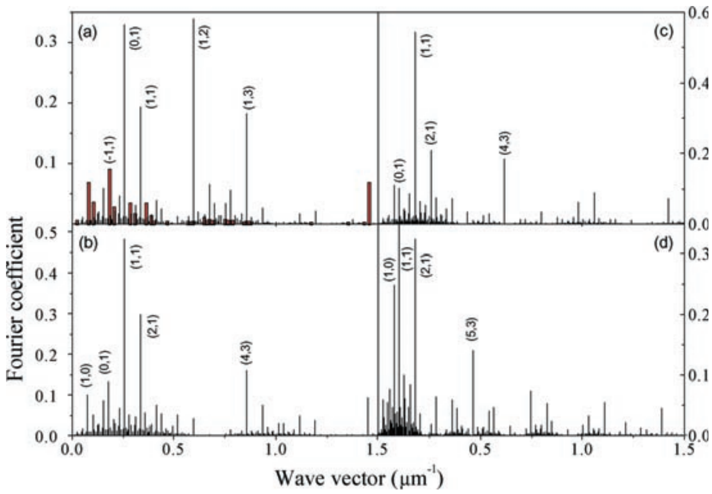

Figure 3. Dependence of the effective nonlinear coefficients on reciprocal vectors for (a) copper Fibonacci QPOSL with $d_A = 21.5$ μm, $d_B = 9.0$ μm and $d_0 = 5.0$ μm; (b) the first-order deflated QPOSL with $d_A = 21.5$ μm, $d_B = 30.49$ μm and $d_0 = 10.0$ μm; (c) the second-order deflated QPOSL with $d_A = 30.5$ μm, $d_B = 40.27$ μm and $d_0 = 15.0$ μm; (d) the third-order deflated QPOSL with $d_A = 40.5$ μm, $d_B = 67.74$ μm and $d_0 = 20.0$ μm.

and so on. This implies that the structure of the reciprocal space is wholly determined by equation (1), the production rule, whereas the initial conditions influence the index of the reciprocal vectors.

It is well known that for the gold Fibonacci superlattice, its reciprocal space exhibits self-similarity. Thus all the superlattices belonging to the gold family have the same self-similarity, whereas for the generalized Fibonacci superlattices there exists generalized self-similarity [23]. Likewise, all the superlattices belonging to the generalized Fibonacci families also possess generalized self-similarity. Figure 3 is the Fourier transformation of the copper Fibonacci superlattices with $n = 3, 2, 1, 0$. It can be seen clearly that the structures of the reciprocal spaces for these four different superlattices are the same. However the difference of the real space structure and the different structure parameters selected make the magnitude of the Fourier components quite different. This greatly facilitates the design of nonlinear optical frequency conversion devices.

Based on the above discussion, a special copper Fibonacci QPOSL has been designed for the THG at 1.7734 μm (here the refractive index data given by Bond have been used [27]), where the reciprocal vector $G_{0,1}$ is used for the QPM SHG process and the reciprocal vector $G_{1,2}$ is used for the QPM SFG process. Theoretical analysis indicates that each process relates to a coupling coefficient (K_j), which is a function of the nonlinear optical coefficient and the Fourier component of the structure. Let K_1 be for the SHG and K_2 for the SFG. The high conversion efficiency depends not only on the magnitude of the coupling coefficients but also on their ratio. The optimum ratio has been determined to be [28]

$$\frac{\tau}{2} = \cos\left(\frac{\sqrt{1-(\tau/2)^2}}{\tau}\ln 3\right) \tag{7}$$

where $\tau = K_1/K_2$, the ratio of the two coupling coefficients. Figure 4 shows the result of maximum conversion efficiency in which the ratio approximately equals τ. In this case, almost all of the fundamental energy can be transferred to the TH.

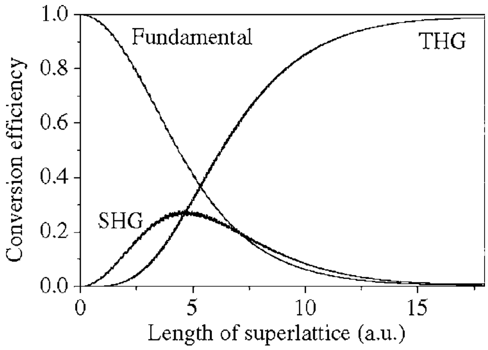

Figure 4. Dependence of the conversion efficiency on $K_2A_{10}L$ at fundamental wavelength 1.7734 μm with the structure parameters: $d_A = 14.0$ μm, $d_B = 29.4$ μm and $d_0 = 9.0$ μm. Where A_{10} is the amplitude of the fundamental wave at the input, L is the total length of the superlattice.

In conclusion, we investigated the nonlinear optical characterization of the generalized Fibonacci superlattices. By the deflation and inflation rule, each generalized Fibonacci superlattice was expanded into a large family. Theoretical analysis and numerical calculation of Fourier transform indicate that superlattices with the same m but different n have the same reciprocal space structure, which plays a significant role in QPM frequency conversion. This reminds us of the isotope, which is any of two or more forms of an element having the same or very closely related chemical properties and the same atomic number but different atomic weights.

Acknowledgments

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