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Aperiodic optical superlattices engineered for optical frequency conversion

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An aperiodic optical superlattice is designed. The designing method is universal and can be applied to all frequency conversion processes by using the coupling of quasiphase matching, without any limitations to special materials and to given fundamental wavelengths. © *2001 American Institute* of Physics. [DOI: 10.1063/1.1381569]

Nonlinear optical frequency converters are attractive sources of coherent radiation in applications for which laser sources are unavailable or for which a wide tunable range is needed. In the optical frequency conversion process, the wave vector conservation involves light waves with different wavelengths. In that case, the wave vector conservation is just the so-called phase-matching condition. The phase mismatch between the interacting waves can be compensated using the birefringence phase-matching (BPM) method. Another promising technique is quasiphase-matching (QPM) realizable in periodic,¹ quasiperiodic,² and aperiodic^{3,4} superlattices.

It is well known that the key of QPM is to construct a structure that provides reciprocal vectors to compensate for the mismatch. It is easy to find the reciprocal vectors of a structure by Fourier transformation. For example, the Fourier spectrum of 1D periodic optical superlattice (POSL) is⁵

$$
F_{\text{POSL}}(x) = \sum_{m} f_m \exp(iG_m x), \tag{1}
$$

where

$$
G_m = \frac{2\pi m}{\Lambda},\tag{2}
$$

$$
f_m = \frac{2}{m \pi} \sin\left(\frac{m \pi}{2}\right). \tag{3}
$$

POSL structure can provide a series of reciprocal vectors, each of which is an integer times a primitive vector. Compared with the periodic structure, a 1D quasiperiodic optical superlattice (QPOSL) has a low space group symmetry, its Fourier spectrum is²

$$
F_{\text{QPOSL}}(x) = \sum_{m,n} f_{m,n} \exp(iG_{m,n}x), \tag{4}
$$

where

$$
G_{m,n} = \frac{2\,\pi(m+n\,\tau)}{D},\tag{5}
$$

$$
f_{m,n} = 2 \frac{(1+\tau)l}{D} \frac{\sin\left(\frac{G_{m,n}l}{2}\right)}{\frac{G_{m,n}l}{2}} \frac{\sin(X_{m,n})}{X_{m,n}},
$$
(6)

$$
X_{m,n} = \pi D^{-1} \tau^2 (m l_A - n l_B), \tag{7}
$$

$$
D = \tau l_A + l_B, \qquad (8)
$$

where *m* and *n* are integers, *D* is an average structural parameter, *l* is an important adjustable parameter, l_A and l_B are the elementary components, and τ the golden mean (1 $+\sqrt{5}/2.$

From the above analysis, for POSL and QPOSL, the reciprocal vectors are distributed discretely. Thus POSL and QPOSL can only be used for single QPM processes such as second harmonic generation (SHG) or high-harmonic generation at some specific wavelength. The aperiodic superlattice (APOSL) has the structure different from both POSL and QPOSL. Several APOSL structures have been reported, which has the advantage either for increasing the wavelength acceptance bandwidth or for pulse compression. Another advantage of APOSL is that its reciprocal vectors can be designed according to the needs. Furthermore, the relative magnitude of the Fourier coefficients can be adjusted artificially. All of these make the APOSL much more flexible for practical use. Here, in this article, a general designing method of APOSL will be proposed. As an example, an APOSL for third harmonic generation (THG) at 1.064 μ m will be discussed.

Before describing the APOSL, it should be emphasized that for any optical superlattice, its structure can be completely determined by a structure function. The structure function only takes $+1$ or -1 , representing two inverse polarization directions in ferroelectric. Two functions will be used below, which must be defined beforehand. The first is $floor(x)$, equal to the greatest integer $\leq x$, and the other is $ceil(x)$, equal to the smallest integer $\geq x$.

First, the structure function of POSL is given as

$$
F_{\Lambda}(x) = 1 - 2floor\left(\frac{2x}{\Lambda}\right) + 4floor\left(\frac{x}{\Lambda}\right),\tag{9}
$$

where Λ is the period. $G_1 = 2\pi/\Lambda$ is one of its reciprocal vector. Then, an aperiodic superlattice is defined as

$$
H(x) = f_{\Lambda} \left(\frac{floor\left(\frac{x}{d}\right) +ceil \cdot (x)}{2} \cdot d \right), \tag{10}
$$

with the width of its smallest domain being,

$$
d = \sigma \frac{\Lambda}{2},
$$
 (11)

FIG. 1. Comparison between the Fourier spectra of POSL and APOSL. (a) The Fourier spectrum of $F_{\Lambda}(x)$ and (b) the Fourier spectrum of $H(x)$.

where σ is an adjustable parameter. The spectra of POSL and APOSL are compared in Fig. 1, where Λ = 7.839 μ m, σ $=\sqrt{2}/2$. It can be seen that the spectrum of APOSL thus defined contains the reciprocal vector $G_1 = 2\pi/\Lambda$.

The Fourier coefficient f of G_1 is determined by the structure parameter σ . The function $f = f(\sigma)$ is shown in Fig. 2. When neither σ nor $1/\sigma$ is an integer,

$$
f(\sigma) = \frac{4}{\sigma \pi^2} \sin\left(\frac{\sigma \pi}{2}\right) \tag{12}
$$

otherwise,

$$
f(\sigma) = \begin{cases} \frac{2}{\sigma \pi} \sin\left(\frac{\sigma \pi}{2}\right), & \sigma \ge 1\\ \frac{2}{\pi}, & \sigma < 1. \end{cases}
$$
 (13)

The APOSL structure is degenerated to POSL structure, as can be seen from Eq. (11) , and the Fourier coefficient related are singularities as shown in Fig. 2.

FIG. 2. Relation between the structure parameter σ and the Fourier coefficient $|f(\sigma)|$.

FIG. 3. Comparison of the Fourier spectra of three APOSLs. (a) $H_1(x)$, (b) $H_2(x)$, and (c) $H_3(x)$.

The APOSL thus defined can only be used for single parametric processes such as SHG, sum frequency generation (SFG), and optical parametric oscillation (OPO). For frequency conversion involving two parametric processes such as THG, the situation becomes more complicated. THG is generated through two processes: the first one the SHG and the second the SFG. The wave equations describing the THG is given by $⁶$ </sup>

$$
\frac{dA_1}{dx} = -i\kappa_2 A_3 A_2^* \exp(-i\Delta k_2 x)
$$

$$
-i2\kappa_1 A_2 A_1^* \exp(-i\Delta k_1 x),
$$

$$
\frac{dA_2}{dx} = -i\kappa_2 A_3 A_1^* \exp(-i\Delta k_2 x) - i\kappa_1 A_1^2 \exp(i\Delta k_1 x),
$$
(14)

$$
\frac{dA_3}{dx} = -i\kappa_2 A_2 A_1 \exp(i\Delta k_2 x),
$$

where κ_1 , κ_2 are two coupling coefficients

$$
\kappa_1 = \frac{d_{33}|f_1|}{2c} \sqrt{\frac{\omega_1^2 \omega_2}{n_1^2 n_2}},\tag{15}
$$

$$
\kappa_2 = \frac{d_{33}|f_2|}{c} \sqrt{\frac{\omega_1 \omega_2 \omega_3}{n_1 n_2 n_3}}
$$
(16)

and

$$
A_i = \sqrt{\frac{n_i}{\omega_i}} E_i, \quad i = 1, 2, 3,
$$
\n(17)

$$
\Delta k_1 = k_2 - 2k_1 - G_1, \tag{18}
$$

$$
\Delta k_2 = k_3 - k_2 - k_1 - G_2. \tag{19}
$$

Where n_1 , n_2 , and n_3 are the refractive indices of the fundamental, SHG and THG, respectively, ω_1 , ω_2 , and ω_3 are the angular frequencies of the fundamental, SHG, THG, and *c* is the speed of light in the vacuum. k_1 , k_2 , and k_3 represent the wave vector of the fundament, SHG and THG, respectively. Δk_1 , and Δk_2 are the phase mismatches in SHG and the sum frequency generation process, respectively. *G*¹

and $G₂$ are the reciprocal vector being used to compensate the mismatch of SHG and SFG, and f_1 and f_2 are the related Fourier coefficients.

Since there are two parametric processes involved in THG, two APOSLs should be defined

$$
H_1(x) = F_{\Lambda_1}\left(\frac{floor\left(\frac{x}{d_1}\right) +ceil\left(\frac{x}{d_1}\right)}{2}d_1\right) \quad \text{for SHG,}
$$
\n(20)

$$
H_2(x) = F_{\Lambda_2}\left(\frac{floor\left(\frac{x}{d_2}\right) +ceil\left(\frac{x}{d_2}\right)}{2}d_2\right) \quad \text{for SFG}
$$
\n(21)

with

$$
d_1 = \sigma_1 \frac{\Lambda_1}{2}, \quad \Lambda_1 = \frac{2\pi}{k_2 - 2k_1},\tag{22}
$$

and

$$
d_2 = \sigma_2 \frac{\Lambda_2}{2}, \quad \Lambda_2 = \frac{2\pi}{k_3 - k_2 - k_1}.
$$
 (23)

For LiTaO₃ with the fundamental wavelength at λ = 1.064 μ m and at room temperature, Λ_1 and Λ_2 can be calculated, Λ_1 =7.839 μ m, Λ_2 =2.174 μ m. If *d* is chosen to be 3.3 μ m, then $\sigma_1 = 0.842$, and $\sigma_2 = 3.036$.

Based on the above definition, an aperiodic structure $H_3(x)$ for THG can be defined, which is

$$
H_3(x) = H_2(x) + \left(floor\left(floor\left(\frac{x}{d}\right)\gamma\right)\right)
$$

$$
-floor\left(ceil\left(\frac{x}{d}\right)\gamma\right)\right)(H_2(x) - H_1(x)) \qquad (24)
$$

here, γ is a very important structure parameter, its range is $0 < \gamma < 1$.

By Fourier transform, two vectors, G_1 for SHG and G_2 for SFG can be found with the related coefficients being f_1 $= \gamma f(\sigma_1)$ and $f_2 = (1 - \gamma)f(\sigma_2)$, respectively. From Eqs. (21) and (22) , we can get

$$
\kappa_1 = \frac{d_{33}\gamma f(\sigma_1)}{2c} \sqrt{\frac{\omega_1^2 \omega_2}{n_1^2 n_2}},
$$
\n(25)

FIG. 4. Conversion efficiency of THG in APOSL. FIG. 5. Conversion efficiency of THG in QPOSL.

$$
\kappa_2 = \frac{d_{33}(1-\gamma)f(\sigma_2)}{c} \sqrt{\frac{\omega_1 \omega_2 \omega_3}{n_1 n_2 n_3}}.
$$
 (26)

Changing γ , we can change the relative magnitude of κ_1 and κ_2 . Accordingly, the ratio of coupling coefficient δ $= \kappa_1 / \kappa_2$ is changed. The relation between δ and γ is

$$
\gamma = \frac{2 \delta \sigma_1 \left| \sin \left(\frac{\pi \sigma_2}{2} \right) \right| \sqrt{3 n_1}}{2 \delta \sigma_1 \left| \sin \left(\frac{\pi \sigma_2}{2} \right) \right| \sqrt{3 n_1} + \sigma_2 \left| \sin \left(\frac{\pi \sigma_1}{2} \right) \right| \sqrt{n_3}}.
$$
 (27)

As is reported, when the ratio of coupling coefficient δ ≈ 0.4429 , the conversion efficiency of THG is highest.⁶ Thus, γ is calculated to be 0.295. Figure 3 shows the comparison of Fourier spectrum of $H_1(x)$, $H_2(x)$, and $H_3(x)$. Compared with those of $H_1(x)$ and $H_2(x)$, the Fourier spectrum of $H_3(x)$ is more complex, which includes all the reciprocal vectors emerged in the two formers. Figure 4 shows the relation of energy conversion of three waves in APOSL. The conversion efficiency of THG can be above 90%. Comparatively, the THG conversion efficiency of a QPOSL has been shown in Fig. 5, which is about 60%.⁷

What has been described above is taken an aperiod $LiTaO₃$ superlattice as an example for direct THG. It must be emphasized that the method is universal, without considering special material and incidental wavelengths. Equations (20) – (24) , and (27) are applicable to all two-frequency conversion processes.

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