Direct-substitution method for studying second harmonic generation in arbitrary optical superlattices

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\section*{Abstract}

In this paper, we present the direct-substitution (DS) method to study the second-harmonic generation (SHG) in arbitrary one-dimensional optical superlattices (OS). Applying this method to Fibonacci and generalized Fibonacci systems, we obtain the relative intensity of SHG and compare them with previous works. We confirmed the validity of the proposed DS method by comparing our results of SHG in quasiperiodic Fibonacci OS with previous works using analytical Fourier transform method. Furthermore, the three-dimension SHG spectra obtained by DS method present the properties of SHG in Fibonacci OS more distinctly. What’s more important, the DS method demands very few limits and can be used to compute directly and conveniently the intensity of SHG in arbitrary OS where the quasi-phase-matching (QPM) can be achieved. It shows that the DS method is powerful for the calculation of electric field and intensity of SHG and can help experimentalists conveniently to estimate the distributions of SHG in any designed polarized systems.

\section*{Introduction}

In 1961 Franken \cite{1} and co-workers observed experimentally the second-harmonic generation (SHG) at \(\sim 347.2\) nm produced upon projection of an intense beam of 694.3 nm light through crystalline quartz and the range of wavelength of laser can be broadened conveniently. It is the first report on nonlinear optical effect after the appearance of laser and from then on, people have paid much attention to the SHG in various materials and the improvement of its energy conversion efficiency.

A classical approach for phase matching uses the birefringent properties of uniaxial or biaxial crystals \cite{2}, but at a specific temperature or angle, usually only a single wave-mixing process can be phase matched out of a large number of desired nonlinear interactions \cite{3}. Using one-dimensional spatial periodic modulation of nonlinear susceptibilities, Armstrong and co-workers \cite{4} proposed a scheme for quasi-phase-matching (QPM) in 1962. The scheme of QPM may be applied to non-birefringent crystals as well as some birefringent crystals with large nonlinear optical coefficients whose phase matching condition cannot be satisfied. Feng et al. \cite{5} expected that for the commonly used nonlinear optical crystal, LiNbO\(_3\), the theoretical maximum enhancement of SHG under QPM scheme could be 23 \(\left[\frac{d_{33}}{d_{31}}\right]^2 (2/\pi)^2 \approx 23\) and observed the enhancement of SHG relative to conventionally phase-matched crystals of the same length. Since the 1990s, the stable techniques of crystal growth have been developed and the experimental difficulties in QPM technique have been overcome, SHG in nonlinear optical crystals with periodically poling \cite{6} have attracted a great deal of attention. The discovery of quasicrystals \cite{7} extends the theory of QPM from periodic optical superlattices to quasiperiodic optical superlattices (QPOS), where the latter has lower space-group symmetry, more Fourier distributions and provides more reciprocal-lattice vectors \cite{8}. An incomplete list includes Fibonacci QPOS \cite{8-10}, the three-component Fibonacci one \cite{11}, Fibonacci-class one \cite{12,13}, and the Family A of generalized Fibonacci GF (m,1) one \cite{14}, etc. Naturally, the theory of QPM was also expanded to study two-dimensional nonlinear photonic crystals \cite{15-17}. Using the means of reciprocal lattice and Fourier transform, Arie et al. \cite{18,19} systematically analyze three wave mixing processes in one-dimensional and two-dimensional nonlinear photonic crystals in which the modulation is either periodic, quasi-periodic, radially symmetric or even random.

The electromagnetic wave theory of SHG in optical superlattices

Conventionally, one can calculate the relative intensity of SHG \(I(2\omega)\) by means of the electric field of fundamental beam (FB), \(E_1\),
and that of SHG, $E_2$, under the small-signal approximation, which satisfy the following wave equation [20]:

$$\frac{dE_2(x)}{dx} = \frac{1}{k_0^2c^2}\frac{\partial^2}{\partial x^2}\left(i32\pi\omega^2E_2(x)\right),$$

(1)

where $\omega$ is the angular frequency of FB, $k_1$ and $k_2$ are wave numbers of FB and SHG, respectively, and $c$ is the speed of light in vacuum. On the other hand, the essential prerequisite for achieving QPM in optical superlattices (OS) is that the phase of nonlinear polarization should shift from one laminar to the next by a $\pi$ radian, and the parameter $d(x)$ equals to $d_{33}$ in positive domains and $-d_{33}$ in negative ones. If one orders the odd number of layers to be positive domains and the even number of layers to be negative ones, then after passing through $N$ layers of OS the electric field $E_2$ can be expressed as follows:

$$E_2(N) = \frac{64\pi\omega^2}{k_0^2c^2\Delta k}d_{33}\left(\sum_{j=0}^{\infty}e^{i\Delta k x j} - \sum_{j=0}^{\infty}e^{-i\Delta k x j}\right),$$

(2)

and $\Delta k$ satisfies the following equation [21]:

$$\Delta k = \frac{4\pi}{\lambda} \left[n_2(\lambda) \cos \theta_2 - n_1(\lambda) \cos \theta_1\right],$$

(3)

where $n_1$ and $n_2$ are the refractive indices of FB and SHG, respectively, $\theta_1$ and $\theta_2$ are the refractive angles of FB and SHG, respectively.

Based on formulae (2) and (3), people usually obtain the distribution of the Bragg peaks for SHG in reciprocal space by use of the Fourier transform of the positions of polarized domains [22]. This working route is a powerful technique and provides clear physical pictures, but unfortunately, even for the quasiperiodic sequences, there are no substitution rules on those domain positions. On this condition, how to compute the intensity of SHG? Could one plot out the SHG spectra conveniently for arbitrary OS where QPM could be achieved? In this paper, we propose the direct-substitution (DS) method to calculate the electric field of SHG directly and apply this method to two kinds of OS, Fibonacci and GF(1,4) systems. After determining the coordinates of the polarized domains' boundaries (but not the general formula of domain positions in conventional analytical method) we can calculate the intensity of SHG directly and continuously. It shows that the DS method is a convenient technique for the calculation of SHG in arbitrary OS.

**Direct-substitution method for studying SHG in arbitrary optical superlattices**

**The definition of direct-substitution method**

From formulae (2) and (3), one can see that the function $E_2(N)$ is only dependent upon two variables, $x$ and $\lambda$, so if the coordinates of the polarized laminar $x$ are fixed then $E_2[\text{and/or } l(2\omega)]$ can be obtained for each $\lambda$ uniquely. It means that any OS structure following the essential prerequisite of QPM can generate SHG and the relative intensity of SHG versus the wavelength of FB can be calculated by substituting the coordinates of the polarized domains into Eqs. (2) and (3) directly. Based on this feature we present a so-called DS method, by which one can draw the spectra of SHG in arbitrary OS directly and continuously. Comparing with the conventional method, although the DS method cannot analytically give out the distribution of the bright lines of SHG, it need not deduce not only the Fourier transform but also the general formula of the coordinates of the polarized domains and then, of course, it makes possible for the experimentalists to predict the properties of SHG in any designed OS and then choose the most suitable structure they demand. As examples, we apply the DS method to study the SHG in two kinds of representative OS, Fibonacci and GF(1,4) systems.

**The application of DS method to Fibonacci optical superlattices**

Fibonacci model is a well-known perfect quasiperiodic sequence which can be generated by the following substitution rules: $B \rightarrow A$ and $A \rightarrow AB$. Starting with a $B$, the first-three generations are

$$\begin{cases}
G_1 = B \\
G_2 = A \\
G_3 = AB
\end{cases}$$

(4)

which shows the following recursion relation:

$$G_l = G_{l-1} G_{l-2}, \quad (l \geq 3).$$

(5)

Similarly to Refs. [9,12,13], we select a LiNbO$_3$ system and make the components $A$ and $B$ each composed of two layers of different domains, whose lengths satisfy the following relations:

$$\begin{cases}
l_1 = l_A + l'_{A} \\
l_2 = l'_{B} + l_B \\
l_3 = l_B \\
l_{A} = l + (1 + \eta)l_B \\
l'_{A} = l + (1 - \eta)l_B
\end{cases}$$

(6)

where $\eta (-1 < \eta < 1/\tau)$ is an adjustable structure parameter, $\tau = (\sqrt{5} + 1)/2$ is a golden number, and the structural parameter $l$ is chosen to be 6.3733 $\mu$m which is the coherence length for the pump beam at wavelength $\lambda_0 = 1.318$ $\mu$m. By means of Eqs. (4)–(6) one can write out the coordinates of the polarized domains and calculate the relative intensity of SHG $I(l(2\omega))$ directly using the DS method. The results are shown in Fig. 1.

From Fig. 1 one can see that all the intense “mountains” of SHG are perpendicular to the $x$ axis. It means that for the Fibonacci OS, the thicknesses of domains $A$ and $B$ only influence the intensity but

![Fig. 1. The spectra of SHG in a LiNbO$_3$ Fibonacci optical OS, where the adjustable structure parameter $\eta$ is defined in Eq. (6), the coherence length for the pump beam $\lambda_0 = 1.318$ $\mu$m is $l_c = 6.373$ $\mu$m, and the corresponding refractive indices of FB and SHG are $n_0 = 2.1453$ and $n_{20} = 2.1970$.](image-url)
not the position of SHG, and $\eta$ corresponds to an adjustable parameter of the intensity of SHG. In other words, whether the SHG with certain wavelength can be generated in the OS only depends on the structure (sequence, symmetry) of the system but not the thicknesses of domains, the latter only determines its intensity. For example, Fig. 1 shows that for $\eta = 1.318 \, \mu m$, the summit of SHG appears at $\eta = 0.0$ (i.e., $l_A = l_B$). This denotes that the most effective structure for generating SHG with certain wavelength is the periodic OS whose domain thicknesses are all equal to the coherence length of the pump beam.

On the other hand, Fibonacci sequence can also be obtained by the projection method, the general formula of the domain positions and therefore its Fourier transform can be deduced easily. Using conventional method several groups have investigated the properties of SHG in these systems [8–10]. In order to compare our results with Ref. [9], we draw the sub-figure of Fig. 1 with $l_A/l_B = \tau$ (i.e., $\eta = 2(\tau - 1)/(1 + \tau^2) \approx 0.342$) in Fig. 2(a) and copy the 3rd figure of Ref. [9] in Fig. 2(b). Obviously, they are accordant with each other.

The application of DS method to GF($m,n$) optical superlattices

The second OS we are going to study are arranged in generalized Fibonacci models $[\text{GF}(m,n)]$ which are quite different from Fibonacci one. The substitution rules for $\text{GF}(m,n)$ are $B \rightarrow A$ and $A \rightarrow A^nB^n$, where $A^n$ denotes a string, $AA\ldots A$, of $n$ $A$’s and $m$ and $n$ are all positive integers. The sequences of $\text{GF}(m,1)$ (where $m > 1$) are known as the Family A of $\text{GF}(m,n)$, and $\text{GF}(1,n)$ (where $n > 2$) are known as the Family B of $\text{GF}(m,n)$. The former are a series of quasiperiodic models and have Cantor-like energy spectra with critical electronic states while the latter are non-quasiperiodic ones and have Bloch-like energy spectra with extended electronic states [23]. Meanwhile, it has been demonstrated that the former can be obtained by projection method and the general formula of the domain positions can be expressed [24], but the latter is just the reverse. Therefore, the conventional method can only be used to calculate SHG in $\text{GF}(m,1)$ systems but will be out of operation for $\text{GF}(1,n)$ ones. By means of the DS method we compute the spectrum of SHG in a $\text{GF}(1,4)$ OS, and plot the results in Fig. 3(a). In order to compare the simulation results with our previous theoretical work [14], we choose the same system parameters as those in Ref. [14] and list the spectrum of SHG in a $\text{GF}(4,1)$ OS (Fig. 4(b) in Ref. [14]).

Comparing Fig. 3(a) with 3(b), one can see that, the spectrum of SHG in $\text{GF}(1,4)$ OS is more complicated than that in $\text{GF}(4,1)$ system, the latter is composed of a series of isolated $\delta$ peaks while the former is made up of some $\delta$ peaks mixing with some quasi-consecutive “mountains”. It can be explained as follows: The reciprocal vectors of the quasiperiodic $\text{GF}(4,1)$ system can be indexed by two positive integers, i.e., its Fourier spectra are composed of a set of $\delta$ peaks. However, $\text{GF}(1,4)$ system is a non-quasiperiodic sequence and has a lower space-group symmetry. It makes $\text{GF}(1,4)$ OS provide more reciprocal-lattice vectors to compensate
for the mismatching phase among interacting waves and therefore have more plentiful Fourier distributions.

Following the same working principle and steps, one can also apply the DS method to study the properties of SHG in other arbitrary OS.

Conclusions

In conclusion, we have proposed the DS method to investigate the properties of SHG in arbitrary OS and apply this method to calculate the intensity of SHG in Fibonacci and generalized Fibonacci systems. We obtain the three-dimensional spectra of SHG intensity in Fibonacci OS versus the fundamental wavelength and the relative thickness of domains \( A \) and \( B \). It is found that the thicknesses of polarized domains of the Fibonacci OS only influence the intensity but not the position of SHG, and the SHG with certain wavelength can be generated by an OS only depending on the structure (sequence, symmetry) of the system but not the thicknesses of domains. On the other hand, we also find that the non-quasiperiodic sequences have a lower space-group symmetry and provide more plentiful distributions for the SHG spectra. Comparing with the conventional Fourier transform method, the DS method demands very few limits and can be used to compute directly and conveniently the intensity of SHG in arbitrary OS where QPM can be achieved. It shows that the DS method is powerful for the calculation of electric field and intensity of SHG and can help experimentalists conveniently to estimate the distributions of SHG in any designed polarized systems.

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