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

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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.

Introduction

In 1961 Franken [1] and co-workers observed experimentally the second-harmonic generation (SHG) at ~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 [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 [3]. Using one-dimensional spatial periodic modulation of nonlinear susceptibilities, Armstrong and co-workers [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. [5] expected that for the commonly used nonlinear optical crystal, $LiNbO_3$, the theoretical maximum enhancement of SHG under QPM scheme could be 23 [i.e., $(d_{33}/d_{31})^2(2/\pi)^2 \simeq 23$] 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 [6] have attracted a great deal of attention. The discovery of quasicrystals [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 [8]. An incomplete list includes Fibonacci QPOS [8–10], the three-component Fibonacci one [11], Fibonacci-class one [12,13], and the Family A of generalized Fibonacci [GF $(m,1)$] one [14], etc. Naturally, the theory of QPM was also expanded to study two-dimensional nonlinear photonic crystals [15–17]. Using the means of reciprocal lattice and Fourier transform, Arie et al. [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,

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and that of SHG, E_2, under the small-signal approximation, which satisfy the following wave equation [20]:

$$\frac{dE_2(x)}{dx} = \frac{i32\pi\omega^2}{k_2c^2} d(x)E_1^2 e^{i(k_2-2k_1)x}, \quad (1)$$

where ω 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 π 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:

$$E_2(N) = -\frac{64\pi\omega^2}{k_2c^2\Delta k} d_{33}E_1^2 \left(\sum_{j=0} e^{i\Delta k x_{2j+1}} - \sum_{j=0} e^{i\Delta k x_{2j}} \right), \quad (2)$$

and Δk satisfies the following equation [21]:

$$\Delta k = |k_2 - 2k_1| = \frac{4\pi}{\lambda}[n_2(\lambda)\cos\theta_2 - n_1(\lambda)\cos\theta_1], \quad (3)$$

where n_1 and n_2 are the refractive indices of FB and SHG, respectively. θ_1 and θ_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, it is always very difficult to conclude the general formula of those domain positions not to speak of the Fourier transform. Furthermore, for arbitrary OS arranged as random sequences, there are even 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 λ, so if the coordinates of the polarized laminar x are fixed then E_2[and/or $I(2\omega)$] can be obtained for each λ 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: $B \to A$ and $A \to AB$. Starting with a B, the first-three generations are

$$\begin{cases} G_1 = B \\ G_2 = A \\ G_3 = AB \end{cases}, \quad (4)$$

which shows the following recursion relation:

$$G_l = G_{l-1}G_{l-2}, \quad (l \geqslant 3). \quad (5)$$

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:

$$\begin{cases} l_A = l_A^+ + l_A^-, & l_B = l_B^+ + l_B^- \\ l_A^+ = l, & l_A^- = l(1+\eta) \\ l_B^+ = l, & l_B^- = l(1-\tau\eta) \end{cases}, \quad (6)$$

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 μm which is the coherence length for the pump beam at wavelength $\lambda_0 = 1.318$ μm. By means of Eqs. (4)–(6) one can write out the coordinates of the polarized domains and calculate the relative intensity of SHG $I(2\omega)$ 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 λ axis. It means that for the Fibonacci OS, the thicknesses of domains A and B only influence the intensity but

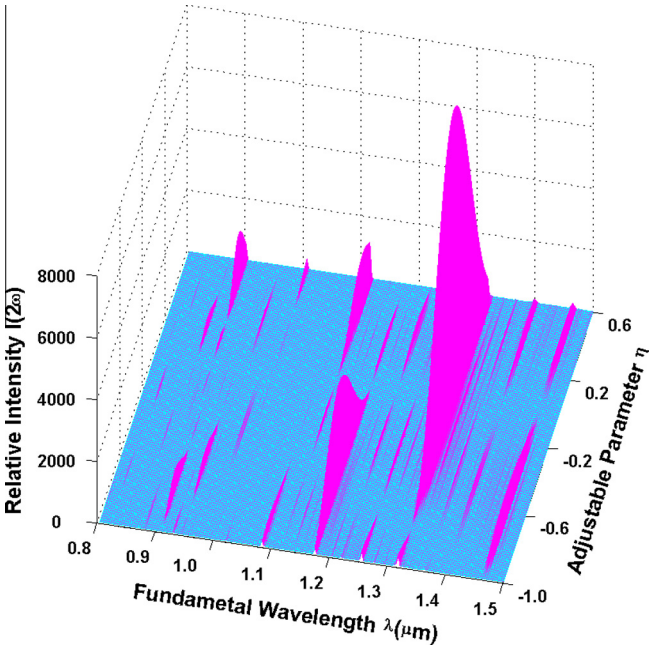

Fig. 1. The spectra of SHG in a $LiNbO_3$ Fibonacci optical OS, where the adjustable structure parameter η is defined in Eq. (6), the coherence length for the pump beam $\lambda_0 = 1.318$ μm is $l_c = 6.373$ μm, and the corresponding refractive indices of FB and SHG are $n_{10} = 2.1453$ and $n_{20} = 2.1970$.