Giant Second-Harmonic Generation in Cantor-like Metamaterial Photonic Superlattices

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ABSTRACT: We present a nonlinear transfer matrix method for studying the second-harmonic generation (SHG) in nonperiodic metamaterial photonic superlattices. A large enhancement of up to 5 orders of magnitude in SHG efficiency was observed for superlattices made with a Cantor-like quasiperiodic assembly of a nonlinear material and a metamaterial. The enhancement was found to depend much more on the electric field amplitude along the structure because of self-similarity effects than on the amount of nonlinear material, which opens the way to design superlattices for tailored applications in broad-band tunable lasers.

INTRODUCTION

Photonic superlattices have become the main route for broadband second-harmonic generation (SHG) with high conversion efficiencies (η) owing to their ability to control light transmission and propagation properties upon designing suitable geometries. Dielectric superlattices, in particular, can be designed to have reciprocal lattice vectors, G_in, to compensate the phase mismatch Δk = k_2ω - 2k_ω, where k_ω and k_2ω are the wavevectors of the fundamental field (FF) and second-harmonic (SH) waves, respectively. This enhances the second-harmonic (SH) conversion efficiency in a mechanism known as the quasi-phase-matching condition.5–9 Other approaches to realize high-efficiency SHG employ the field amplification in the structure because of the slow light effect (high density of modes) at photonic band edges,10–20 or the strong light confinement in photonic cavities, defective and disordered superlattices, and plasmonic systems.21–27 The incorporation of artificial materials with a negative refractive index (n_0 = sqrt(-ε_0 - μ_0) < 0) has led to striking phenomena in photonic superlattices. The so-called metamaterials with simultaneous negative dielectric permittivity (ε_0) and magnetic permeability (μ_0)28–34 yield superlattices with a gap under oblique incident light, i.e., θ ≠ 0, known as the magnetic/electric bulklike plasmon-polariton (PP) gap. The latter originates from the resonant coupling of the magnetic/electric field component of light with the corresponding plasmonlike μ_0(ω)/ε_0(ω) effective response under transversal electric/magnetic (TE/TM) polarized light, which cannot be observed in all-dielectric superlattices. The bulklike PP gap edges and the PP defective mode were used for the giant enhancement of SHG in the microwave and terahertz regimes,35 where the intrinsic losses of metallic building blocks were shown to have a detrimental effect on the SHG in the high-frequency regime.

In this work, we present an extended version of the nonlinear transfer matrix method (TMM),34 available for dielectric and periodic bilayer systems, to consider nonperiodic metamaterial systems under oblique incidence light. TMM is notably simpler, from the analytical and computational points of view, than the Green’s functions method used earlier for periodic metamaterial bilayer systems.7–9 Through the nonlinear TMM developed here, we use the bulklike PP gap edges to numerically demonstrate that the enhancement factor of SHG has a stronger dependence on the field enhancement in the structure than on the amount of nonlinear material. For these purposes, calculations are made using Cantor-like quasiperiodic metamaterial superlattices, for which the amount of nonlinear material diminishes as (2/3)^N for each successive N-step of the Cantor series, whereas the unit cell length remains fixed. This is in contrast with Thue–Morse- and Fibonacci-like superlattices, where the unit cell thickness increases with the sequence step.34 Self-similarity properties of these nonlinear structures can also be used to extend the results to be presented for broad-band SHG applications,30–38 which cannot be reached, for example, using defect modes.35

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Figure 1. Transmission spectra of a quasiperiodic metamaterial superlattice with its unit cell made according to the (a) first, (b) second, and (c) third steps of the Cantor series. (d)–(f) show the transmission spectra of the SH wave generated in the systems in (a)–(c), respectively. All of the systems were made with periodic repetition of 10 unit cells. The angles of incidence were the same for the FF and SH waves.

THEORETICAL FRAMEWORK

We are concerned here with the SHG from finite quasiperiodic one-dimensional metamaterial superlattices. The superlattices are made as a periodic repetition of unit cells built by alternating a nonlinear dielectric LiNbO3 and a linear negative-refractive metamaterial, labeled B, and following the Cantor fractal series, as depicted in the upper chart of Figure 1. Using a specific rule to design the fractal unit cells, such as the Cantor series, allows for control of self-similarity properties, thus providing flexibility for tailoring and tuning the photonic properties. Hypothetical lossless metamaterial slabs are considered, with their permittivity and permeability given

\( \varepsilon(\nu) = 1 - \frac{\nu^2}{\nu_0^2} \)

\( \mu(\nu) = 1 - \frac{F_0^2}{\nu^2 - \nu_0^2} \)

where \( \nu_0 = 10 \text{ GHz}, F = 0.56, \nu_0 = 4 \text{ GHz, with } \nu = \omega/2\pi. \) By solving \( \mu(\nu) = 0, \) we found the magnetic bulk PP frequency \( \nu_m = 6.03 \text{ GHz}, n^{(1)}_m = 2.157 \) and \( n^{(2)}_m = 2.237. \)

The refractive indices for the dielectric nonlinear layers at their FF and SH waves, respectively. At the microwave frequency regime, the assumption of negligible losses is well supported by previous works.32–34 We use \( E_0 = 10^7 \text{ V/m (intensity } \sim 13.3 \text{ MW/cm}^2) \) as the incident electric field amplitude.3 The superlattices are taken as grown along the z-axis. The propagation of a transversal electric (TE) incident wave, i.e., the electric field \( E \) is perpendicular to the plane of polarization (xz-plane) and the magnetic field \( H = H_{TE,j} + H_{TE,l} = iH\cos \theta + iH \sin \theta \) is along that plane, can be described within the nondepleted pump approximation as

\[
\left( \frac{d^2}{dz^2} + (k_c^{(1)})^2 \right) E^{(1)} = 0
\]

\[
\left( \frac{d^2}{dz^2} + (k_c^{(2)})^2 \right) E^{(2)} = - \left( \frac{k_0}{\nu} \right)^2 \left( E^{(1)} \right)^2
\]

with \( \chi^{(2)} = n^{(2)}_B k_0^2 \cos(\theta_0^2), \) \( k_0^2 = \frac{n_0^2}{\nu_0^2}, \) \( k_0^2 = k_0^2 \cos(\theta_0^2), \) and \( \theta_0^2 \) denoting the corresponding second-order nonlinear susceptibility, wavevectors, and propagation angles for FF (\( j = 1 \)) and SH (\( j = 2 \)) waves in the \( i \)th layer, where \( \chi^{(2)} = 0 \) for linear metamaterial slabs, and \( k_0^2 = 6.7 \text{ pm/V for nonlinear LiNbO}_3 \) slabs at microwave frequencies.39,40
\[ A_j(\omega) = -\left(\frac{2\omega}{c}\right)^2 \frac{\mu_1^{(2)}k_{1z}^{(2)}}{\mu_1^{(2)}k_{0z}^{(2)}} \]

\[ C_j(\omega) = -\left(\frac{2\omega}{c}\right)^2 \frac{\mu_1^{(2)}k_{1z}^{(2)}}{\mu_1^{(2)}k_{0z}^{(2)}} I \]

\[ B_i = \begin{pmatrix} 1 & 1 \\ \frac{2k_{1z}^{(1)}}{\mu_1^{(2)}k_{0z}^{(2)}} & \frac{k_{1z}^{(1)}}{\mu_1^{(2)}k_{0z}^{(2)}} \end{pmatrix} \]

\[ F_i = \begin{pmatrix} e^{2k_{1z}^{(1)}d} & 0 \\ 0 & e^{-2k_{1z}^{(1)}d} \end{pmatrix} \]

\[ G_i = \begin{pmatrix} 1 & 1 \\ \frac{k_{1z}^{(2)}}{\mu_1^{(2)}k_{0z}^{(2)}} & \frac{k_{1z}^{(2)}}{\mu_1^{(2)}k_{0z}^{(2)}} \end{pmatrix} \]

\[ Q_i = \begin{pmatrix} e^{k_{1z}^{(2)}d} & 0 \\ 0 & e^{-k_{1z}^{(2)}d} \end{pmatrix} \]

For simplicity, we define

\[ D_m = G_0^{-1}S^K G_0 = \begin{pmatrix} D_{11}^{(2)} & D_{12}^{(2)} \\ D_{21}^{(2)} & D_{22}^{(2)} \end{pmatrix} \]

to write eq 5 as

\[ \begin{pmatrix} E_i^{(2)+} \\ 0 \end{pmatrix} = \begin{pmatrix} D_{11}^{(2)}E_0^{(2)-} + T_1^{(2)} \\ D_{22}^{(2)}E_0^{(2)-} + T_2^{(2)} \end{pmatrix} \]

where

\[ T_m = \begin{pmatrix} T_1^{(2)} \\ T_2^{(2)} \end{pmatrix} = G_0^{-1}\sum_{j=1}^{K} S^{r-j} \sum_{l=1}^{L} Z^{r}(M_l J_{l-j-1}-(L-\delta)} + J_0^{(1)+} E_{l-j-1}^{(1)+} E_{l-j-1}^{(1)-} \]

With the expressions above, the forward/backward second-harmonic amplitudes can be written in a very simplified way as

\[ E_{0}^{(2)-} = -T_2^{(2)} D_{22}^{(2)} + T_1^{(2)} \]

\[ E_{i}^{(2)+} = -T_2^{(2)} D_{22}^{(2)} + T_1^{(2)} \]

from where the total conversion efficiency is calculated by \( \eta = \eta_f + \eta_b \) with \( \eta_f = \frac{|T_f^{(2)+}|^2}{|T_f^{(2)+}|^2} \) and \( \eta_b = \frac{|T_b^{(2)+}|^2}{|T_b^{(2)+}|^2} \) being the efficiencies for the forward and backward SH waves, respectively. The total length of the unit cell for each Cantor step \( N \) can be written as \( L_{N} = \left(\frac{2}{3}\right)^{N} I + \sum_{j=0}^{N-1} \left(\frac{2}{3}\right)^{j} \left(\frac{2}{3}\right)^{j} = I \) \((0 \leq j < N)\), in contrast to other fractal structures, like Thue–Morse or Fibonacci, where the unit cell lengths increase with the sequence step. The first (second) term in \( L_{N} \) corresponds to the amount of nonlinear material (metamaterial) in the unit cell. For simplicity, we use \( I = 27 \) mm for all of the calculations in this work.
RESULTS AND DISCUSSION

Transmittance spectra of the FF and SH waves associated with the first three Cantor steps, $N = 1, 2,$ and $3$, are shown in Figure 1a–f. The corresponding unit cells are illustrated in the upper chart of Figure 1. All quasiperiodic superlattices were taken as made by periodic repetition of 10 unit cells. For $\theta \neq 0$, there is a gap broadening around $\nu_p = 6.03$ GHz with increasing $\theta$. This gap is known as the PP gap, and is due to the coupling of the longitudinal magnetic field component of light, $H_{TE,LL}$, with the magnetic plasmonlike effective response of metamaterial layers. Figure 1a–c displays several sharp PP gaps for $N > 1$, because of self-similarity effects of the quasiperiodic structure, as predicted in ref 42. We tuned the angles of incidence as $\theta = 27.08^\circ$ (for $N = 1$), $\theta = 46.45^\circ$ (for $N = 2$), and $\theta = 24.69^\circ$ (for $N = 3$) to have both the FF and SH frequencies placed at gap edges to obtain the highest values of the electric field amplitudes inside the system. FF (SH) frequencies were selected as $\nu_{FF} = 5.8986$ GHz ($\nu_{SH} = 11.7972$ GHz), $\nu_{FF} = 5.9205$ GHz ($\nu_{SH} = 11.841$ GHz), and $\nu_{FF} = 6.0258$ GHz ($\nu_{SH} = 12.0516$ GHz), for $N = 1, 2$, and $3$, respectively. The use of strong light–matter interaction in the bulk PP gap combined with nonlinear properties of photonic superlattices to produce giant enhancements of SHG efficiency has been already demonstrated.\textsuperscript{35} Here, we are interested in the analysis of two competing effects: first, increasing $N$ diminishes the amount of LiNbO$_3$ material in the unit cell as $(\frac{z}{l})^N$, which we may expect to be reflected in a reduction of the nonlinear effects. Second, localized modes with enhanced amplitudes are excited by increasing the self-similarity properties of the unit cell, because of the amount of disorder introduced by the fractal aspects, which must improve the second-order nonlinear interaction in the right-hand side of eq 4. This electromagnetic field enhancement can be noted from the electric field profiles in Figures 2a,b, 2c,d, and 2e,f for $N = 1, 2$, and $3$, respectively, for the FF and SH waves. To confirm that the maximum electric field amplitudes for FF occur inside the LiNbO$_3$ material slabs or at their boundaries, in Figure 3 we replotted these results for the two unit cells around the center ($z = 135$ mm) of the corresponding superlattices. From Figure 3a (for $N = 1$), Figure 3c (for $N = 2$), and Figure 3e (for $N = 3$), we observe that the electric field profiles for the plasmonic FFs follow the self-similarity properties of the Cantor-like superlattices, as expected. In contrast, the corresponding dielectric SH field profiles in Figure 3b,d,f do not exhibit self-similarity because the dispersive effect in the structure is stronger.\textsuperscript{3}

SH conversion efficiencies for these quasiperiodic systems are presented in Figure 4. Results for superlattices are compared with the ones corresponding to a slab of LiNbO$_3$ with the same total length, under the same light incidence conditions. It is clear that increasing the field intensity inside the system is more important than the amount of nonlinear metamaterial in the structure, thus indicating a way to improve the efficiency of this nonlinear process without requiring large amounts of nonlinear material. Note also that for $N = 1$ the enhancement of $\eta$ is less than 1 order of magnitude compared to a slab of LiNbO$_3$ with the same total length. However, enhancements of up to 5 orders of magnitude are observed for superlattices made by following the Cantor steps $N = 2$ and $3$.

CONCLUSIONS

We have extended the nonlinear TMM for dielectric periodic superlattices to treat nonperiodic metamaterial superlattices, and demonstrated large enhancement factors for SH conversion efficiency in the bulklike PP gap edges of Cantor-
like quasiperiodic superlattices. Significantly, the electromagnetic field enhancement along the structure is much more important than the amount of nonlinear material in the superlattice to enhance conversion efficiency. This opens an avenue to develop systems with small amounts of nonlinear material and high conversion efficiencies; hence, our results may stimulate the design and development of photonic platforms inspired by (and/or beyond) the one considered here.

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