Generation of coherent terahertz radiation with multifrequency modes in a Fibonacci optical superlattice

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We investigate theoretically the generation of coherent narrow-band terahertz (THz) radiation with multifrequency modes in an optical superlattice produced by quasiperiodically poled ferroelectric crystal. The superlattice consists of two building blocks A and B, in which each containing a pair of antiparallel 180° ferroelectric domains, arranged as a Fibonacci sequence. Second-order nonlinear optical rectification of femtosecond pulses gives rise to a THz wave form whose multifrequency modes are determined by the quasiperiodic structure properties. The relationship between multifrequency modes terahertz generation and Fibonacci structure parameters is also analyzed and studied. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1598644]

Since the discovery of the icosahedral phase in Al–Mn alloys in 1984,[1] the field of quasiperiodic structures (or quasicrystals) has caught a lot of attention. Besides the studies focused on their structural and physical properties,[2,3] much effort has been devoted to the applications of quasiperiodic structures. For example, a strong suppression of the optical transmission in quasiperiodic dielectric multilayer stacks of SiO_2 and TiO_2 thin films has been observed.[4] Magnetic polaritons in Fibonacci magnetic quasicrystals and coherent acoustic phonons in Fibonacci optical superlattice have been investigated,[5,6] respectively. Study of wave packet dynamics in quasione-dimensional metal–halogen complex has been reported[7] and quasiperiodic envelope solitons has been introduced.[8] For quasiphase-matching grating, a nonlinear quasiperiodic optical superlattice, multicolor second harmonic generation with conversion efficiencies of $\sim 5\%-20\%$ has been measured.[9] The quasiperiodic optical superlattice in $LiTaO_3$ can also be used for efficient direct third harmonic generation.[10]

On the other hand, coherent terahertz (THz) radiation is of great interest for a wide variety of applications in fundamental and applied sciences such as spectroscopy, sensing, communication, medical diagnoses, as well as biomedical imaging and tomography. Many molecular excitations in condensed matter systems fall into the THz frequency range which results in the great demand for narrow-band THz sources. A number of approaches have been therefore taken to generate narrow-band THz radiation. These include difference generation,[11] photomixing,[12] optical parametric oscillation,[13] and plasma oscillation.[14]

Recently, one promising technique to generate narrow-band THz radiations has been demonstrated, using second-order optical rectification of femtosecond pulse in periodically poled $LiNbO_3$ (PPLN).[15] In the absence of absorption and domain-width fluctuation, the relative bandwidth $\Delta\nu/\nu$ of the THz field is given simply as $2/N$, where N is the number of domains in PPLN. However, it is well known that such THz generation in PPLN consists of only single frequency mode radiation, which really limits the applications of THz sources. In most of applications frequency tunability is important and essential. It is interesting and significant to investigate the THz generation with frequency tunability and multifrequency modes. In the present work, we report our studies on optical characterization of narrow-band terahertz with multifrequency modes using an optical Fibonacci superlattice, and demonstrate numerically the possibility of multifrequency modes THz radiation in wide frequency range.

Narrow-band THz wave generation can be calculated by considering the second order optical rectification induced by femotsecond pulses. Assuming the second order susceptibility to be modulated periodically or quasiperiodically, we write wave equation of frequency domain in the form

$$\frac{\partial^2 E_{\mathrm{TH}}(z,\omega)}{\partial z^2}+\varepsilon(\omega)\frac{\omega^2}{c^2}E_{\mathrm{TH}}(z,\omega)=-\omega^2\mu_0 P^{(R)}(z,\omega), \tag{1}$$

where the source term, $P^{(R)}(z,\omega) = (1/2\pi)\,\varepsilon_0 d(z)\int_{-\infty}^{+\infty}E_{\mathrm{opt}}(z,t)E^*_{\mathrm{opt}}(z,t)\exp(-i\omega t)dt$, represents optical rectification induced by the Fourier transformation of the optical pulse intensity; $\varepsilon(\omega)$ is the dielectric function and the spatial modulation of the susceptibility is described by the grating function $d(z)$. The envelope of femtosecond pulse moves at the group velocity without any distortion when the dispersion of the optical pulse in the medium is neglected. For a typical Gaussian input pulse, the analytical local solution of THz field can be integrated directly, and, at the output of the crystal, the contribution of the THz field from position z can be written in form

$$\begin{aligned}E_{\mathrm{TH}}(z,\omega)_{\mathrm{LOCAL}}=&A_0\exp\left[-\frac{1}{4}\tau^2\omega^2-\gamma_T\frac{(L-z)}{c}\omega\right]\\&\times\exp\left[-i\left(\frac{z}{v_O}+\frac{L-z}{v_T}\right)\omega\right]+rA_0\\&\times\exp\left[-\frac{1}{4}\tau^2\omega^2-\gamma_T\frac{(L+z)}{c}\omega\right]\\&\times\exp\left[-i\left(\frac{z}{v_O}+\frac{L+z}{v_T}\right)\omega\right],\end{aligned} \tag{2}$$

where $A_0=1/(n_{\mathrm{TH}}^2-n_{\mathrm{O}}^2)\,I_0\,(\tau\sqrt{\pi}/2\pi)\,d(z)$ and $v_O=c/n_{\mathrm{O}}$, $v_T=c/n_{\mathrm{TH}}$ and $E_{\mathrm{TH}}(z,\ \omega)$ consists of two terms correspond-

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TABLE I. The Fibonacci indices and THz frequencies in range of principal value.

m, n	0, 1	1, 0	0, 2	1, 1	0, 3	2, 0	1, 2	0, 4	2, 1	1, 3	3, 0	0, 5
$f_{m,n}$ (THz)	0.64	1.03	1.27	1.67	1.91	2.06	2.30	2.55	2.70	2.94	3.09	3.18

ing to THz radiation propagating in forward and backward direction, respectively; r is the reflection of THz wave at the crystal-air interface. Equation (2) may be Fourier transformed to get the THz field in time domain, and then spatially averaged to give the total output field.

The quasiperiodic grating used for simulations is the same as the one introduced and studied previously.[9] It has two building blocks A and B of length l_A and l_B, respectively, which are ordered in a Fibonacci sequence according to the production rule $S_j = S_{j-1}|S_{j-2}$ for $j \geqslant 2$, with $S_1 = \{A\}$ and $S_2 = \{AB\}$. Each block has a domain of length l_{A1} (l_{B1}) with positive ferroelectric domain (black) and a domain of length l_{A2} (l_{B2}) with negative ferroelectric domain. In this letter, l_{A1} is set to be equal to l_{B1}. For our simulations presented later we have chosen the ratio τ of length scales l_A and l_B as $\tau = (1+\sqrt{5})/2$, the so-called golden ratio. Furthermore, the structural parameter l can also be adjusted in our simulations. The grating function $d(z)$ varies between $+d$ and $-d$ according to the Fibonacci sequence.

As the femtosecond optical pulses propagate through the domain-reversal crystal, a THz nonlinear polarization is generated by optical rectification. Each domain in the crystal contributes a half cycle to the radiated THz field. The frequency of THz wave is determined essentially by

$$\nu_T = \frac{mc}{\Lambda(n_{TH} - n_O)}, \tag{3}$$

where c is the light velocity, for a periodic structure, Λ is the period of domain-reversal crystal. m is integer index and $m = 1$ represents principle value of THz frequencies which corresponds to the most intense mode in spectral domain. This formula can be extended from periodic structure to quasiperiodic structure. In analogy to periodic structure, it is expected that their THz radiation will reflect the quasiperiodicity of the Fibonacci sequence. The multifrequency modes THz radiations are given by quasiperiodicities

$$\nu_T = \frac{c}{\Lambda_{m,n}(n_{TH} - n_O)}, \quad \Lambda_{m,n} = \frac{D}{m + n\tau}, \tag{4}$$

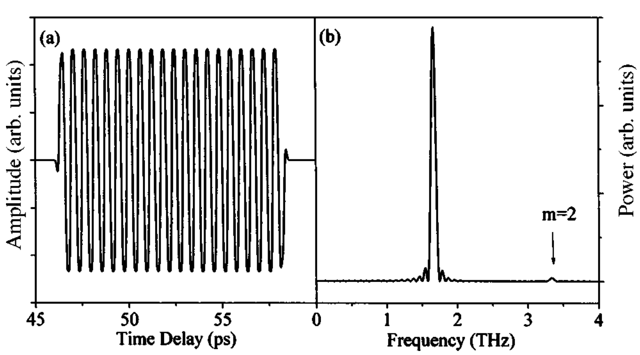

FIG. 1. Simulated THz wave forms in time domain (a) and corresponding power spectra (b). PPLN domain structure parameters are assumed as $L = 1.2$ mm and $\Lambda = 60\ \mu$m.

where m and n are integer indices of the quasiperiodicities and $D = \tau l_A + l_B$ is the average lattice parameter. THz frequencies in quasiperiodical structures are seen to confine to its principal value when $m + n\tau < 2 + 2\tau$. There are 12 possible Fibonacci indices inside the principal value presented in Table I. The main or so-called central frequency of principal values corresponds to the quasiperiodic index (1, 1).

Figure 1(a) shows the result of a calculation in time domain for 150 fs pulses, generated from a 250 kHz Ti:sapphire regenerative amplifier, incident on a PPLN crystal with $L = 1.2$ mm and $\Lambda = 60\ \mu$m, where loss is neglected and the widths of positive and negative domain are set to be equal. The refractive indices of optical pulse and THz radiation are assumed as $n_{TH} = 5.2$ and $n_O = 2.2$, respectively.[16] The corresponding single frequency mode of the power spectrum plotted in Fig. 1(b) is 1.67 THz, which is in good agreement with Eq. (3). Here the smaller frequency mode induced by "backward" wave are omitted and the higher frequency modes ($m = 2$) is also marked in Fig. 1. The multifrequency modes characteristics of THz radiation in Fibonacci grating are indicated in Figs. 2(a) and 2(b). The oscillatory properties of THz wave form in time domain are different from THz radiation in PPLN. It is obvious that some beat features and quasiperiodicities of THz wave form are observed. The frequencies of THz modes are labeled by the quasiperiodicity indices (m, n) as in Eq. (4). Here we set the average parameter of quasiperiodical Fibonacci grating as 60 μm ($\Lambda_{1,1} = 60\ \mu$m), which is same as the period Λ of PPLN, so that the main peaks of THz frequencies in the power spectrum are same for cases of Figs. 1(b) and 2(b). The ratio of length scales l_A to l_B is τ and we have chosen $l_A = 70.4\ \mu$m and $l_B = 43.2\ \mu$m, respectively. The width of positive domain in both blocks A and B is 25 μm, which can be adjustable from 0 to 43.2 μm. In comparison with situation of periodical structure, it is obvious that the fine structures of THz spectra power appear in quasiperiodical Fibonacci grating. Besides the most intense peak taken with a quasiperiodical index (1,1), some other THz frequency modes, labeled by (1,0), (2,1) and so on, are easily observed in power spectrum, cor-

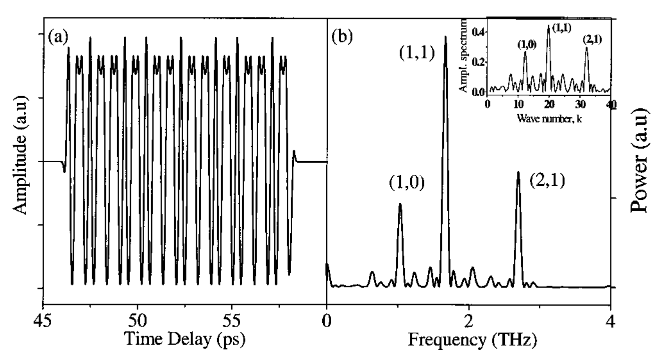

FIG. 2. Simulated THz wave forms in time domain (a) and corresponding power spectra (b). The quasiperiodic Fibonacci domain structure parameters are assumed as $l_A = 70.4\ \mu$m and $l_B = 43.2\ \mu$m ($\Lambda_{1,1} = 60\ \mu$m), respectively. The width of positive domain in block A and B is 25 μm, The structure consists of 12 building blocks A and 8 building blocks B.

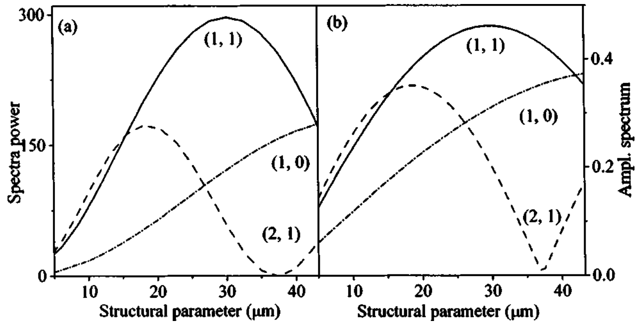

FIG. 3. Dependence of the power spectra of THz radiation (a) and Fourier transform coefficients of Fibonacci structure (b) on the structure parameter l with the fixed widths of blocks A and B ($l_A = 70.4$ μm and $l_B = 43.2$ μm). Here the three most intense modes with quasiperiodic indices (1, 0), (1, 1), and (2, 1) are calculated, respectively.

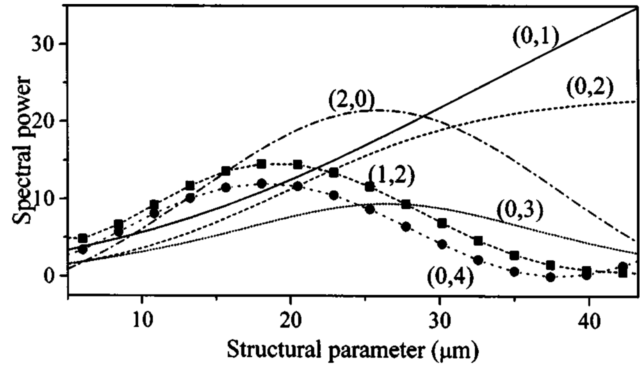

FIG. 4. Multimodes THz generation as a function of the structure parameter l with the fixed widths of blocks A and B, here some moderate modes with quasiperiodic indices (0, 1), (0, 2), (2, 0), (1, 2), (0, 3), and (0, 4) are considered. The corresponding parameters are same as those in Fig. 3. Different curves correspond to different quasiperiodic indices.

responding to the THz frequencies 1.03, 2.70 THz, and so on, respectively. Inset of Fig. 2(b) plots the calculated Fourier amplitude spectrum of quasiperiodical Fibonacci structure. The peak intensities of THz radiation are in accordance with amplitude spectrum of grating function. It is also known that the amplitude spectrum of quasiperiodical structure strongly depends on the width of positive domain in block A and B. The dependences of the generated THz power spectra [Fig. 3(a)] and Fourier transform spectrum of grating function [Fig. 3(b)] on width of positive domain (adjustable structure parameter) of quasiperiodic Fibonacci grating are illustrated, respectively. Several main intense and moderate quasiperiodic indices are calculated inside principle values.

From these figures, it can be seen that the variation of THz power with the structure parameter has the same tendency as Fourier amplitude spectrum of grating function for main indices (1, 0), (1, 1), as well as (2, 1). That means, the nonlinear processes of THz generations strongly relate to the reciprocal vectors induced by quasiperiodic structures. Meantime, the optimum conditions for the multimodes THz can be determined. Two frequency modes with same power amplitude at 1.03 and 1.67 THz, respectively, can be realized for structure parameter $l = 16$ μm. It is also clearly seen that the two THz frequencies with equal power for 1.67 and 2.70 THz emerge when positive and negative domains almost merge in building block B.

There are more freedoms for generation of THz waves, for example, when the structure parameter l equals 30 μm, the most intense THz radiation with index (1, 1) takes the maxima. Whereas for THz generation with index (2, 1), the largest value occurs at the structure parameter close to 20 μm and THz wave with index (1, 0) increases monotonously in whole regain of structure parameters.

Moreover, some calculated results with respect to lower THz spectral powers are illustrated in Fig. 4. For Fibonacci indices (0, 3), (2, 0), (1, 2), and (0, 4), the THz spectral powers oscillate with corresponding frequencies 1.91, 2.06, 2.30, and 2.55 THz, respectively, whereas the spectral powers, with Fibonacci indices (0, 1) and (0, 2), increase steadily with corresponding frequencies 0.64 and 1.27 THz. The power amplitude of all modes inside principal value with practical potential can be generated when the structure parameters located in the region of 20–30 μm. Further calculations indicate that the spectral power with zero mode (0, 0) can be restrained for structure parameters between 20 and 30 μm.

In conclusion, we have analyzed and investigated THz generation with multifrequency modes in an optical Fibonacci superlattice. The dependence of THz radiation on quasiperiodicities and reciprocal vectors of Fibonacci grating have been obtained. In comparison with THz generation in PPLN we have predicted that a significant source of coherent THz radiation with multifrequency modes will be achieved using quasiperiodic optical superlattices. Such THz source with multifrequency modes can provide THz radiation in a wide range of frequencies whose spectral power can be adjusted by structure parameters. The results presented here would be beneficial and flexible to the design of a practical THz generator with multifrequency modes.

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