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Theoretic considerations for multi-mode terahertz generations in multi-periodically poled dielectric material

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Abstract
We propose a new efficient method to achieve terahertz (THz) radiation with multi-mode and equal spectral power in multi-periodically poled dielectric materials. The optimal structure consists of a uniform grating and multiple phase-reversal sequences. Single-, dual-, triple-, quartic-, and even multi-mode THz radiations can be generated flexibly using different pre-designed structures. The dependence of THz radiation on the optical pulse duration is studied and analysed; this turns out to be an important parameter for optimum THz generation.

In recent years, the advent of artificial microstructured dielectric superlattices has opened an area in the development of new synthetic materials that are of interest and importance for optical devices. Typically, the characteristics of these dielectric superlattices strongly depend on the modulation of their material parameters. The best known dielectric superlattice is the photonic crystal [1, 2], which is based on a variation of the dielectric constants. A lesser known example is the new kind of function material, called phononic crystal [3, 4], which is based on the modulation of elastic parameters. Furthermore, if the piezoelectric coefficient is modulated in a phononic crystal, the interaction between the superlattice vibrations and electro-magnetic waves may generate polaritons that are induced by both transverse and longitudinal waves [5, 6]. Besides the photonic crystal and phononic crystal mentioned above, the modulations of electro-optic and nonlinear optical coefficients have been extensively studied for more effective electro-optical switching [7, 8] and quasi-phase-matched frequency conversion [9, 10]. Quite apart from material parameters, the design of superlattices is further extended by complex structural modulation such as quasi-periodic or aperiodic structures. Much effort has been devoted to the applications of these complex structures. For example, magnetic polaritons in Fibonacci magnetic quasi-crystals and coherent acoustic phonons in Fibonacci optical superlattices have been investigated, [11, 12] respectively. The efficient direct third harmonic generation in a quasiperiodic $LiTaO_3$ optical superlattice has been reported [10].

On the other hand, narrow-band terahertz (THz) radiation is of great interest due to the abundance of excitations in molecular systems and condensed media. These frequencies can bridge the gap between optical waves and microwaves for high-frequency telecommunication. A number of approaches have been proposed for the generation of narrow-band THz radiation. Most recently, a ferroelectric superlattice (to be called an optical superlattice henceforth), in which the ferroelectric domain is modulated periodically, has been used to generate a THz wave [13]. However, only single THz frequency radiation can be generated in the periodically poled ferroelectric structure. In order to encompass a wide range of applications, it is desired to generate THz radiation with a tunable range of frequencies. Multi-channel frequency tuning by lateral spatial chirping of the domain width has been demonstrated [14, 15]. Dual- or triple-mode THz generation in a Fibonacci optical superlattice has been predicted [16]. In this paper, we report our theoretical studies on optical characterizations of THz radiation in multi-periodically poled ferroelectric material. Compared to THz radiation in multi-channel gratings and Fibonacci optical superlattices, our simulation results indicate that multi-periodically optical superlattices have the advantage of equal spectral power in a single sample; and the multi-frequency THz generation can be pre-determined flexibly according to the practical requirements.

THz radiations are generated via optical rectification in the pre-engineered domain structure of poled ferroelectric material. More accurately this effect can be described by a difference frequency generation process: when short laser pulses with a broad frequency spectrum are incident on a nonlinear material, at each point z of the medium the difference frequency mixing between the closely spaced different spectral components of the pump wave $E_P(\omega, t, z)$ induces a second-order nonlinear polarization:

$$P_2(\Omega, z) = \tfrac{1}{2}\varepsilon_0 \int \chi^{(2)}(-\Omega, \Omega+\omega, -\omega) E_P(\Omega+\omega, z) E_P^*(\omega, z)\, d\omega \tag{1}$$

where ω and Ω lie, respectively, in the optical and THz frequency ranges. This radiation at frequencies Ω contains frequencies from 0 to several THz. The amplitude of each spectral THz component $E_T(\Omega, z)$ is computed by solving the nonlinear Maxwell equation [17] in the spatial and temporal Fourier domains:

$$\frac{\partial^2 E_T(\Omega, z)}{\partial z^2} + k(\Omega)^2 E_T(\Omega, z) = H(\Omega) \exp[i\Omega(z/v_g)] \tag{2}$$

with $H(\Omega) = -(\Omega^2/c^2)\chi^{(2)}(\Omega)C(\Omega)$, where $C(\Omega)$ is the Fourier transformation of the intensity profile of the input pulse, and v_g is the group velocity of the optical pulse. The solution of the Maxwell equation yields the THz field at each point in the crystal:

$$E_T(\Omega, z) = iz \frac{1}{k(\Omega) + \Omega/v_g} \frac{\Omega^2}{c^2} \chi^{(2)}(\Omega) C(\Omega) \times \exp\left[i\left(k(\Omega) + \frac{\Omega}{v_g}\right)\frac{z}{2}\right] \sin c\left[\left(k(\Omega) - \frac{\Omega}{v_g}\right)\frac{z}{2}\right]. \tag{3}$$

$\chi^{(2)}(\Omega) = 2d_{33}(\Omega)$ and the spatial modulation of the susceptibility is described by the grating function, which changes sign from one domain type to the other. As previously shown [18], if the dispersion of $\chi^{(2)}(\Omega)$ is neglected, the strong modification of the THz spectrum at the exit of the crystal is basically the product of three contributions: the Ω^2 dependence of the radiative efficiency, the initial power spectrum of the rectified input laser pulse $C(\Omega)$, and the phase-matching curve represented by the sin$c$ function.

The aperiodic gratings used for simulations in our paper consist of multi-periodic structures, which provide clearer physical nature and less complication than the simulated

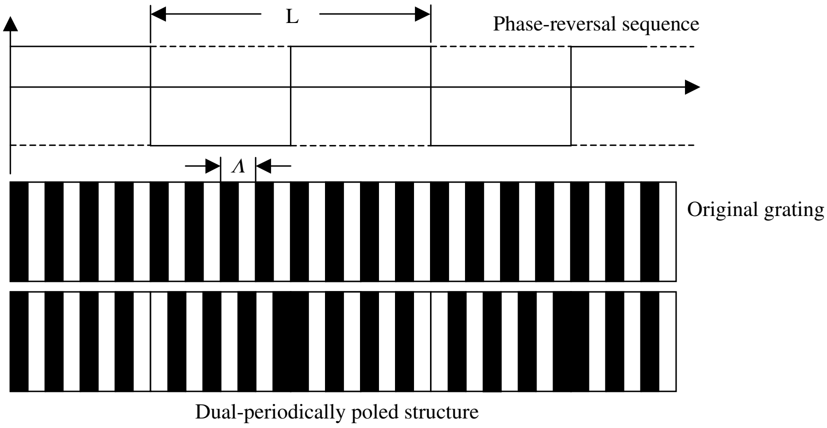

Figure 1. A schematic diagram of a uniform periodic grating superimposed by a phase-reversal sequence. Here the period ratio L/Λ is an integer.

annealing method commonly used. The multi-periodic structures are designed using multiple periodic phase-reversal sequences superimposed upon a uniform periodic grating, which are flexible to generate single-, dual-, triple-, quartic-, and even multi-mode THz radiations.

As the femtosecond optical pulses propagate through the domain-reversal crystal, a THz nonlinear polarization is generated by difference frequency generation between the spectral components of the ultrashort laser pulse. For a uniform periodic grating, by integrating equation (3) over a periodic superlattice where the second-order nonlinear susceptibility changes its sign when passing from a positive domain to a negative domain, the frequency of the THz wave is determined, essentially by [13]

$$\nu_S = \frac{mc}{\Lambda_u(n_{TH} - n_O)} \tag{4}$$

where n_O is the group index of refraction of the optical pulse; also, in general n_{TH} depends itself on the frequency ν_S. Λ_u is the period of the uniform periodic grating, m is an integer index and $m = 1$ represents the principal value of THz frequencies that corresponds to the most intense mode in the spectral domain. If the uniform periodic grating is superimposed with the phase-reversal period Λ_P, the possible THz frequencies in this structure are given by ν_u and $\nu_u \pm \nu_P$, which corresponds to THz generations with dual or triple mode. Here, we consider only the fundamental mode in detail. THz radiations with two or three peaks are implemented by controlling the duty cycle of both the uniform periodic grating and the phase-reversal sequence. A schematic diagram consisting of a uniform periodic grating and a phase-reversal sequence is shown in figure 1. Moreover, THz generations with four or more peaks can be implemented by the superimposition of another phase-reversal sequence on a two-period structure, thereby splitting the two (three) peaks into six (nine). The spectral powers of central peaks relative to the other side peaks are strongly dependent on the duty cycles of the uniform grating and the superimposed periods.

Figure 2 shows a THz waveform with a uniform grating and the corresponding spectra from poled $LiNbO_3$ with 30 domains and $\Lambda_u = 50\ \mu$m. The optical pulse duration is set to 150 fs and a typical Gaussian beam is applied. The corresponding single-frequency mode of the power spectrum shown in figure 2(B) is 2.0 THz, which is in good agreement with equation (4). In figure 3, a dual-peaked THz waveform is demonstrated. The uniform 50 μm domain structure is superimposed with a 100 μm phase-reversal period structure, resulting in two peaks of THz

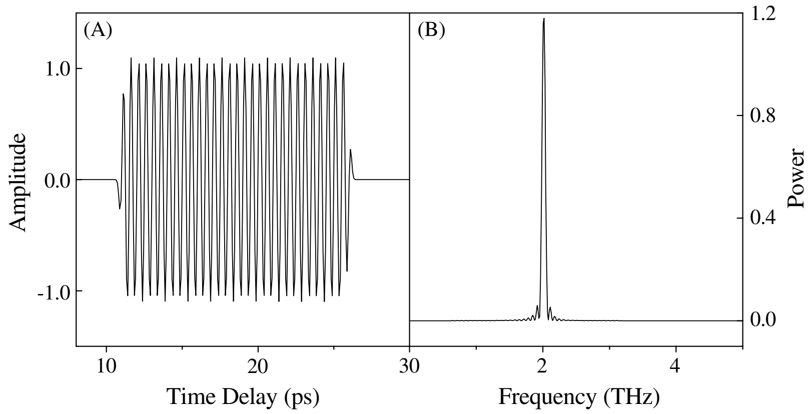

Figure 2. Simulated THz wave form in the time domain (A) and corresponding spectral power with single-mode 2.0 THz (B). The structural period of a uniform periodic grating is assumed as $\Lambda_u = 50\ \mu$m and $L = 1.5$ mm is the total length of the superlattice. Both positive and negative domains are set to be 25 μm.

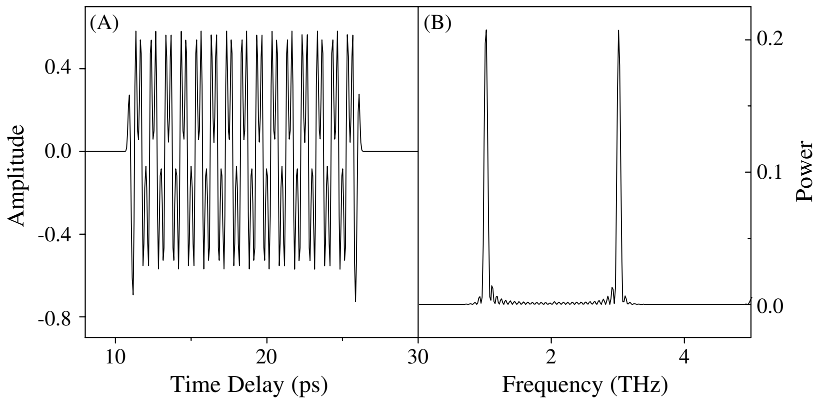

Figure 3. Simulated THz wave form in the time domain (A) and corresponding spectral power with dual-mode 1.0 and 2.0 THz, respectively (B). The structural periods of a uniform periodic grating and the phase-reversal sequence are assumed as $\Lambda_u = 50\ \mu$m and $\Lambda_P = 100\ \mu$m, respectively. The lengths of the positive and negative domains are set to be equal in a uniform periodic grating and adjusted slightly in the superimposed phase-reversal sequence.

generation in the frequency spectrum corresponding to 1.0 and 3.0 THz, respectively. In order to obtain equal spectral power, here, the duty cycle of the phase-reversal sequence is slightly adjusted.

The triple-mode THz radiation with equal spectral power is indicated in figure 4(A). By controlling the duty cycle of the phase-reversal sequence, three peaks separated by 1.0 THz are observed in the frequency spectrum. Figure 4(B) shows the THz generations with four peaks in a three-period structure. The periods of uniform grating and two phase-reversal sequences are 50, 100, 200 μm, respectively. At most seven THz peaks can be obtained in such a structure, corresponding to 0.5, 1.0, 1.5, 2.0, 2.5, 3.0, 3.5 THz. By adjusting the duty cycles of a uniform grating and phase-reversal sequences, four peaks are obtained in the spectral domain. Here an optical pulse duration of 120 fs is employed for the quartic-mode THz generation. Interestingly it is seen that in the THz generation of quartic mode the spectral power is larger

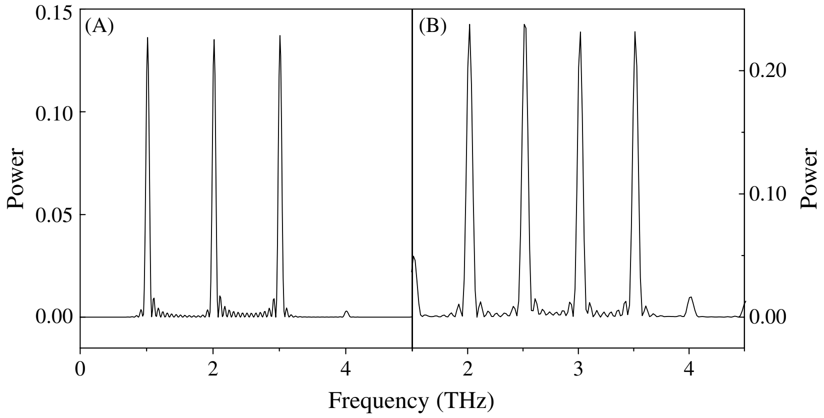

Figure 4. (A) Simulated THz spectral power with triple-mode 1.0, 2.0 and 3.0 THz, respectively. The structural periods of a uniform periodic grating and the phase-reversal sequences are assumed as $\Lambda_u = 50\ \mu$m and $\Lambda_{p1} = 100\ \mu$m, respectively. The duty cycle of the phase-reversal sequence is 38–62 μm. (B) Simulated THz spectral power with quartic-mode 2.0, 2.5, 3.0, 3.5 THz, respectively. The structural periods of a uniform periodic grating and the phase-reversal sequences are assumed as $\Lambda_u = 50\ \mu$m, $\Lambda_{p1} = 100\ \mu$m and $\Lambda_{p2} = 200\ \mu$m, respectively. The duty cycles of the uniform periodic grating and two phase-reversal sequences are 31–19 μm, 25–75 μm and 64–136 μm, respectively.

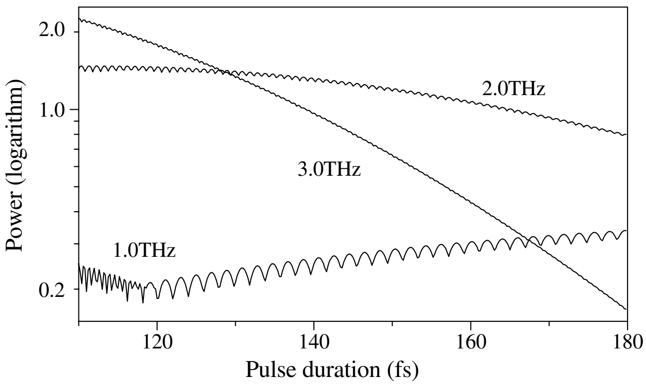

Figure 5. Dependence of the spectral power of THz radiation on the optical pulse duration. Three curves corresponding to different frequencies 1.0, 2.0 and 3.0 THz, respectively, are plotted.

than that of THz radiations with dual and triple mode. This implies that the shorter the duration of the femtosecond pulse the higher the spectral power is generated in THz radiation. The result stems from the fact that for shorter pulse durations, the spectral components with larger intensities dominate the difference frequency mixing process. More dependence of spectral power on pulse duration is shown in figure 5. The spectral power strongly depends on the pulse duration even under the quasi-phase matching operation. Both of the two radiations at 2.0 and 3.0 THz are seen to decrease monotonically in the pulse duration range of 110–180 fs with convex and concave variations, respectively. For the lower 1.0 THz radiation, the minimum spectral power appears at around the pulse duration 120 fs and is accompanied by local oscillations. Moreover, the spectral powers of THz radiations are more intense with increasing frequency. Qualitatively these phenomena can be understood from the point of view of interferences between the interacting spectral components. For THz radiations with lower frequencies, it is clearly observed in figure 5 that the interference stripes are more intense, which leads in turn to lower corresponding spectral power.

In summary, we have analysed and investigated THz generation with multi-mode in a multi-periodically poled dielectric material. The multiple frequencies are determined by the periods of a uniform grating and the phase-reversal sequences. The spectral powers are dependent on both the duty cycles of the structural periods and the incident optical pulse duration. Here the physical nature is different from the traditional optical difference frequency mixing process. Such a THz source with multi-mode can provide flexible choices of pre-designed THz frequencies and equal spectral power for different applications.

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