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Generation of broadband and multiple-peak THz radiation in aperiodically poled lithium niobate

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We theoretically analyze the generation of broadened and multi-peak terahertz (THz) radiation in aperiodically poled lithium niobate (APPLN), whose sequence of opposite domains is optimized by simulated annealing (SA) algorithm. The full-width at half maximum (FWHM) of the broadened THz radiation in our simulation is 0.26 THz. Both of the central wavelength and FWHM can be easily tuned by choosing proper objective functions. THz radiation with wider and flatter FWHM can be achieved by increasing the length of the lithium niobate crystal. The two-peak THz generation is also provided as an example of multi-peak with the central wavelengths at 1.68 and 1.80 THz, respectively.

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Terahertz (THz) regime has become the most attracting frequency range in the electromagnetic radiation in recent years[1-3]. Single-cycle THz waves intrinsically possess a broad bandwidth[4]. Unfortunately, the generated THz waveform is very weak and inhomogeneous for the THz frequency. It has been proved experimentally[5-8] that the velocity mismatch between the THz pulse and the optical femto-second pulse in the periodically poled lithium niobate (PPLN) allows the generation of ultranarrow band THz waveforms. There is a trade-off between the bandwidth and the intensity of generated THz waves. In many applications, THz waveforms with arbitrary flatten-top bandwidth will greatly extend the specified scope of the THz spectroscopy, so it is necessary to find a way by which the fixed bandwidth of THz radiation can be achieved.

In this paper, we demonstrate theoretically the generation of a THz radiation with a fixed bandwidth via optical rectification in the pre-engineered domain structure, which is optimized by the simulated annealing (SA) algorithm[9]. By setting different objective functions, we can obtain not only specific bandwidth at a fixed central wavelength, but also multi-peak THz radiation in frequency domain. These engineered THz pulses in aperiodically poled lithium niobate (APPLN) may have potential applications in THz-time domain spectroscopy and biochemistry. By the one-dimensional nonlinear wave equation simulation, we are able to obtain the pre-designed bandwidth as well as the central frequency of THz wave. The bandwidth of the THz waveform is 0.2 THz centered at 1.74 THz. And we also provide a theoretical analysis on generation of double-peaks THz radiation using SA algorithm.

A simple model is used to calculate the THz waveform. Assuming plane-wave propagation, the wave equation in the crystal is described as

\[
\frac{\partial^2 E_{\text{THz}}(z, \omega)}{\partial z^2} + \varepsilon(z) \frac{\omega^2}{c^2} E_{\text{THz}}(z, \omega) = -\frac{\varepsilon_0 \hbar}{2} P^{(R)}(z, \omega),
\]

where the source term is

\[
P^{(R)}(z, \omega) = \frac{1}{2\pi} \frac{1}{\hbar} \int F_{\text{opt}}(z, t) E_{\text{opt}}^*(z, t) \exp(-i\omega t) dt,
\]

which represents optical rectification induced by the Fourier transformation of the optical pulse intensity. \( \varepsilon(\omega) \) is the dielectric function and the special modulation of the susceptibility is described by the grating function \( d(z) \). In our simulations, the envelope of femto-second pulse transmits at the group velocity without any distortion, and the dispersion and loss of the optical pulse in the medium are neglected. With an assumption of instantaneous nonlinearity, Fourier transformation of the optical pulse \( |E(t)|^2 \) gives the source term from Eq. (2). By Eq. (1), the THz field \( E_{\text{THz}}(z, \omega) \) can be obtained. For an analytical solution, we may assume a Gaussian input pulse \( |E_{\text{opt}}(t)|^2 = I_0 \exp(-t^2/\tau^2) \). At the output of the crystal \( z = L \), the contribution of the THz field from position \( z' \) is

\[
E_{\text{THz}}(z', w) = \pm A_0 w^2 \sqrt{\pi \tau} \times \exp \left[ -\frac{\tau^2}{4} w^2 - i \left( \frac{z'}{v_{\text{opt}}} + \frac{L-z'}{v_{\text{THz}}} \right) w \right],
\]

where

\[
A_0 = 1/(n_{\text{THz}}^2 - n_{\text{opt}}^2) \frac{|I_0(\pi \sqrt{\tau}/2\pi)|}{d(z)}, \quad v_{\text{opt}} = c/\omega_{\text{opt}}, \quad v_{\text{THz}} = c/n_{\text{THz}}.
\]

The sign of the right-hand side in Eq. (3) is determined by crystal orientation of the domain at \( z' \). Equation (3) may be Fourier transformed to obtain the time-domain THz field, and then spatially integrated to give the total output field.

PPLN, with periodic domain inverted structures, has been widely used as an effective laser frequency converter, based on quasi-phase-matching (QPM) technique owing to its outstanding nonlinear optical properties[6-8]. Subsequently, aperiodic optical superlattice (AOS)[9-11]...
with aperiodic opposite ferroelectric domains was theoretically and experimentally demonstrated to achieve broadband or multiple wavelengths conversion because it can provide plenty of reciprocal vectors to compensate for the wave-vector mismatches between the interacting waves. As for THz field generation via optical rectification in a PPLN crystal, because the positive and negative domains change alternatively, the amplitude sign of THz field in time domain are different in positive and negative domains, so that that ultra-short bandwidth THz field can be generated. As an example, the indexes of refraction of pump beam and THz wave are $n_{\text{opt}} = 2.3$ at 800 nm and $n_{\text{THz}} = 5.2$ at 1.7 THz. The domain length is determined by the walk-off length $d = c\tau/[n_{\text{THz}} - n_{\text{opt}}] = 30 \mu m$, and the period is 60 $\mu m$. The central frequency of THz generated from the PPLN is $\nu = c/\lambda(n_{\text{THz}} - n_{\text{opt}})$ and the bandwidth of the spectrum is given essentially by $\delta \nu/\nu = 2/N = 0.05$. So the full-maximum of the peak in the power spectrum is 0.077 THz at 1.7 THz which is rather small. Practically, due to the large absorption in THz domain for lithium niobate, the bandwidth of generated THz is often broadens and the band-top is inhomogeneous\cite{3,4}.

Compared with the broadband frequency conversion by nonlinear optics process in PPLN, the broadband with homogeneous band-top or multiple-peak THz field can be achieved by choosing proper aperiodically poled domain-inverted structure.

The domain orientation of each block is determined by an optimized method, simulated annealing (SA) algorithm. SA algorithm is employed to optimize sequence of the positive and negative blocks until the transmissivities of the prescribed THz frequencies are equal, reaching maximum values at the same time. Here, we chose the objective function in the SA algorithm

$$F = \sum_a T(\omega_a) - \{\max[T(\omega_a)] - \min[T(\omega_a)]\},$$  \hspace{1cm} (4)

where the symbols $\max[\cdots]$ and $\min[\cdots]$ take their maximum and minimum values along all the quantities into $[\cdots]$. $\omega_a$ is the transmission frequencies selected as the objective frequencies.

In our simulations, the pump laser is Ti:sapphire regenerative amplifier with 800-nm wavelength, 100-fs pulse duration, and 250-kHz repetition rate. The fs laser passes through an APPLN crystal, which is 1.2 mm in length, with domain length of 30 $\mu m$.

In the case of APPLN, an objective function to obtain full-width at half-maximum (FWHM) of 0.20 THz at 1.74 THz is set. The simulation result by optimized SA algorithm is shown in Fig. 1. The FWHM of the generated THz field is about 0.26 THz, which is a little smaller than we expected. Compared with the other simulation of broadened THz radiation\cite{12} the calculation results shows better features of shape. Both the central wavelength and width of FWHM can be tuned by choosing different objective functions. Because of the small number of blocks in the optimization calculations, some noises below 1.5 THz can be found. These noises can be suppressed by using longer crystal.

In order to obtain the two-peak THz radiation in APPLN, we choose the proper objective function in the SA method

$$F = [T(\omega_1) + T(\omega_2)] - \max[T(\omega_a)],$$  \hspace{1cm} (5)

$\omega_1$ and $\omega_2$ are the two selected frequencies in objective function, and $\omega_1 < \omega_a < \omega_2$.

As shown in Fig. 2, two THz fields can be generated simultaneously in the designed APPLN. The central wavelengths of the two peaks are 1.68 and 1.80 THz respectively, and the FWHMs are 0.12 and 0.1 THz. This method can be employed to the generation of multiple (> 2) THz field if proper objective function is chosen in the optimization algorithm.

In our calculations, because the transmission loss of THz field in lithium niobate is rather large, the length of APPLN crystal is as short as 1.2 mm with domain length of 30 $\mu m$. We cannot obtain even wider bandwidth because only 40 domains are used in the SA searching process. If the transmission loss of the crystal, which can be domain-inverted poled, is rather small, the wider bandwidth or multiple THz radiations can be easily achieved in aperiodically poled domain-inverted structure.

In summary, by using SA optimal algorithm, we demonstrate a theoretical analysis on generating a THz radiation with a fixed bandwidth via optical rectification in the pre-engineered domain structure. The FWHM of the broadband THz radiation achieved in this paper is 0.26 THz. The two-peak THz generation is provided as an example of multi-peak with the central wavelengths at 1.68 and 1.80 THz, respectively.

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