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Article

Nearly Single-Cycle Terahertz Pulse Generation in Aperiodically Poled Lithium Niobate

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Abstract: In present work an opportunity of nearly single-cycle THz pulse generation in aperiodically poled lithium niobate (APPLN) crystal is studied. A radiating antenna model is used to simulate the THz generation from chirped APPLN crystal pumped by a sequence of femtosecond laser pulses with chirped delays τ_m (m = 1, 2, 3 …) between adjacent pulses. It is shown that by appropriative choosing τ_m it is possible to obtain temporally overlap of all THz pulses generated from positive (or negative) domains. It results in the formation of a nearly single-cycle THz pulse, if the chirp rate of domain length δ in the crystal is sufficiently large. In opposite case, a few cycle THz pulses are generated with the number of the cycles depending on δ. The closed form expression for THz pulse form is obtained. The peak THz electric field strength of 0.3 MV/cm is predicted for APPLN crystal pumped by the sequence of laser pulses with peak intensity of the separate pulse in the sequence about 20 GW/cm^2. By focusing the THz beam and by increasing the pump power the field strength can reach values of an order of few MV/cm.

Keywords: terahertz; ultrafast photonics; nonlinear optics; lithium niobate

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1. Introduction

The area of the applications that require intense terahertz (THz) pulses has been rapidly increasing during the last years [1,2]. Optical rectification (OR) of the femtosecond laser pulses in lithium niobate (LN) crystals has emerged as the most powerful way to generate broadband nearly single-cycle THz pulses with spectrum centered at frequencies below 1 THz. The broadband single-cycle THz pulses with record energy of 0.4 mJ [3] and pump-to-THz conversion efficiency η = 3.8% [4] have been reported. To satisfy phase matching condition the pump beam is reflected off a grating to acquire a tilted-pulse-front (TPF), which is then subsequently imaged onto the crystal using a lens or a telescope. However, imaging errors limit effective length of the crystal [5,6] and, therefore, further increase in THz generation performance is challenging. The attempts to mitigate this problem by using contacting grating [7], stair-step echelons [8,9], and its combination with reflecting grating [10]) still do not result in the THz generation with energy or efficiency close to the abovementioned values. It is well established [11,12] that effective interaction length in the semiconductor materials such as ZnTe, GaP, and GaAs, can be significantly larger due to operation ability at smaller tilt angles < 30° and lower THz-wave absorption at room temperature. However, to escape 2nd- and 3rd-order multiphoton pump absorption (MPA), these materials have to be pumped at longer wavelengths $\lambda_p \geq 1.7$ μm, where it is still relatively challenging to obtain femtosecond laser pulses with required high power.

It should be noted that MgO-doped LN crystals have the advantages of large nonlinear coefficient (especially among the inorganic crystals), sufficiently wide bandgap (the lowest order effective MPA is four-photon absorption at the 1.03 μm pump wavelength), high damage threshold and, finally, ability for the spatial sign-modulation of the nonlinear coefficient. The latter is important as nowadays high-power pulses can be used to pump periodically poled lithium niobite (PPLN) crystals. The large area PPLN crystals with cross section 3 mm × 3 mm are commercially available and even a crystal with dimensions 1 cm × 1.5 cm was recently used for THz generation

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[13]. Besides, ability to significantly reduce THz-waves absorption in PPLN crystal by cryogenic cooling allows to use crystals with a length about 2 - 3 cm [13,14]. Therefore, investigations of alternative methods for nearly single-cycle broadband THz pulse generation in domain-engineered PPLN crystals are interesting.

It has been well established that the use of aperiodically poled lithium niobate (APPLN) leads to the generation of broadband THz pulses [15-18]. However, its temporal form is not a nearly single-cycle THz pulse (i.e. it is not determined by spectral bandwidth) because different spectral components are radiated from different positions of the crystal. Like in the optical region, the technique of broadband pulse compression by using Bragg grating, chirped mirror, and pairs of gratings or prisms, can be used. However, strong THz beam divergence limits its applications.

The opportunity of nearly single-cycle THz pulse generation in APPLN crystal in conjunction with THz chirped mirror has been analyzed in detail by Yahaghi and co-worker in Ref. [19]. However, the difficulties of THz chirped mirror fabrication and its application in THz generator scheme complicates practical implementation. To avoid this problem, recently [20] it was proposed and analyzed a nearly single-cycle THz pulse generation in chirped APPLN crystal using a pair of chirped optical pump pulses with various chirp rates. The relative chirp between the pump pulses compensates the temporal shift between THz frequencies generated at different locations in the crystal. As a consequence, the THz pulse emerges compressed upon exiting the crystal.

Taking advantage of the above idea, here we introduce a new method to generate THz pulses with controllable number of THz field oscillations, from nearly single- to multi-cycles and correspondingly on the spectral bandwidth from broadband to narrowband. The sequence of laser pulses with chirped delay between adjacent pulses (so-called pulse-position modulated (PPM) signal in optical communication) is used to pump chirped APPLN crystal. Such sequence of the laser pulses can be obtained by using birefringent crystal array [21] or modern pulse shaping techniques [22]. It is known [16-18] that in the case of crystal pumping by single fs-pulse the generated THz waveform corresponds to domain structure of the APPLN crystal. More exactly, it corresponds to spatial reversal replica (or so-called phase-conjugate replica) of the APPLN domain lengths distribution, considering that the exciting laser pulse propagates faster than the radiated THz-wave. The set of these temporally shifted replicas is originated, when pump wave is the sequence of laser pulses with chirped delays τ_m (m = 1, 2, 3…). By appropriate choosing the τ_m it is possible to obtain a temporary overlap of all generated frequencies (in the range of $\Delta\omega_{THz}$) that results in formation of a nearly single-cycle THz pulse in the case of sufficiently large both M and $\Delta\omega_{THz}$, where $\Delta\omega_{THz}$ is the bandwidth of THz generation in APPLN by separate laser pulse.

This paper provides a theoretical model for efficient generation of nearly single-cycle THz pulse in the chirped APPLN crystal pumped by a sequence of laser pulses with chirped delay times. Closed form expression for temporal shape of the generated THz pulse is obtained. The peak THz electric field strength of 0.3 MV/cm is predicted for APPLN crystal pumped by the sequence of laser pulses with peak intensity of the separate pulse in the sequence about 20 GW/cm^2.

2. Theoretical model

Let us consider a transform-limited optical pulse with Gaussian temporal and spatial profiles that propagates along the x-axis of APPLN crystal having a nonuniform distribution of the nonlinear coefficient $d_{33} = d_{33}(x)$. The APPLN crystal consists of N number domains having lengths of l_k (k = 1, 2…N), which are reduced along the direction of the pump beam propagation, i.e. the lengths l_k are reduced with increasing index m. In every adjacent domain the sign of the nonlinear coefficient d_{33} is opposite (see Figure 1).

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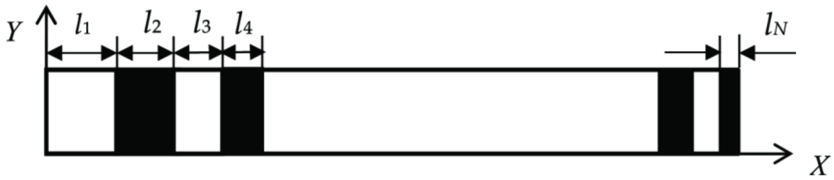

Figure 1. The schematic illustration of APPLN crystal, where white and dark colors are used for regions with opposite sign of nonlinear coefficient d_{33}.

To calculate the electric field of THz generation the radiating antenna model [23,24] is used. For simplicity, pump pulse depletion and its spatial and temporal distortions during propagation in the crystal are neglected. To avoid optical and THz waves' reflections at the interfaces of the crystal, we assume that it is incorporated in a linear medium having the same refractive index. With these assumptions the instantaneous intensity of pump pulse is given by

$$I(t,\vec{r}) = I_m \exp\left[-\left(\frac{y^2+z^2}{a^2}\right)\right]\exp\left[-\left(\frac{t-xn_g/c}{\sigma}\right)^2\right], \quad (1)$$

where a is the beam waist radius, n_g is the group refractive index, 2σ is the pulse duration at the $1/e$ level that is related to the full width at half maximum by $\sigma_{FWHM} = 2(\ln 2)^{1/2}\sigma$, and I_0 is determined by the pulse energy $\Im$ in the form: $I_m = \Im/S\pi^{1/2}\sigma$ with $S = \pi a^2$.

It is known [25] that in the frequency domain low-frequency nonlinear polarization P_{NL} is related to the pump intensity by the following expression

$$P_{NL}(\omega,\vec{r}) = \frac{2d_{33}(x)}{n_0 c} f(y,z) I(\omega) e^{-ik_g x}, \quad (2)$$

where $f(y,z) = \exp\left[-(y^2+z^2)/a^2\right]$, $I(\omega) = \Im\{I_0(t)\} = I_p\sqrt{\pi}\sigma\exp\left(-\omega^2\sigma^2/4\right)$ is the Fourier transform of the pump intensity $I_0(t) = I_p\exp(-t^2/\sigma^2)$ at entrance of the crystal $x = 0$, n_0 is the refractive index at the laser frequency, ε_0 is the electric permittivity of free space, $k_g = \omega n_g/c$.

To calculate the THz far-field produced by the nonlinear polarization, the PPLN crystal is considered as an antenna fed by the z-oriented current given by $j_z(\omega,\vec{r}) = i\omega P_{NL}(\omega,\vec{r})$. In antenna theory the calculation of the radiation field for a given distribution of excitation currents leads to the integration in the so-called radiation integral over spatial coordinates $\vec{r} \equiv (x,y,z)$ [26]. To simplify integration over transverse coordinates y and z, it is assumed that pump beam is completely inside the crystal. Finally, by presenting THz field as the superposition of field radiated by the separate crystal domains, we obtain that THz field is given by

$$E(\omega) = A_0 \exp\left[-i(\omega n_{THz}c^{-1}R)\right]\omega^2 I(\omega)\sum_{m=1}^{N}(-1)^m F_m(\omega)\int_0^{l_m}\exp\left[i\left(\omega\Delta n c^{-1}\right)(x+x_m)\right]dx, \quad (3)$$

where $$A_0 = \frac{\mu_0 d_{33} S}{2\pi R n_0 c},\; F_m(\omega) = e^{-i\omega T_m},\; T_m = \sum_{k=m+1}^{N}\left(\frac{n_{THz}}{c}\right)l_k,\; x_m = \sum_{k=1}^{m-1} l_k \quad \text{with } x_1 \equiv 0, \quad (4)$$

$\Delta n = n_{THz} - n_g$, n_{THz} is the refractive index of the crystal at THz frequencies, μ_0 is the permeability of vacuum, R is the distance between point of THz field observation and the exit surface of the APPLN crystal. Here T_m is the time required for THz-wave propagation from the generation location (m-domain) to the exit surface of the crystal (N-domain).

After integration in Equation (3) the THz field is given by

$$E(\omega) = B_0 e^{-i\omega T_R}\omega I(\omega)\sum_{m=1}^{N}(-1)^m\left[e^{i\omega(\tau_{m+1}-T_m)} - e^{i\omega(\tau_m - T_m)}\right], \quad (5)$$

where $\tau_m = \Delta n x_m/c$, $T_R = n_{THz}R/c$, and $B_0 = A_0 c/\Delta n$.

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From above it follows that temporal form of the radiated THz pulse can be easily calculated. Indeed, factor $\omega I(\omega)$ is the Fourier transform of temporal derivative of the pump intensity, $\dot{I}(t) = (dI/dt)$, the exponential factors in the sum represent temporal shifts of $\dot{I}(t)$ on the corresponding values, and the exponential factor before the summation symbol is related to the time of THz-wave propagation from the crystal's exit surface to the field observation point. Using it the THz electric field in time domain is given by

$$E(t_R) = B_0 \sum_{m=1}^{N} (-1)^m \left[\dot{I}(t - T_m + \tau_{m+1}) - \dot{I}(t - T_m + \tau_m) \right], \quad (6)$$

where $t_R = t - T_R$ is the retarded time.

2. Results and discussions

Here we consider the linearly chirped APPLN, in which the domains lengths are decreasing from the beginning to the end of the crystal as $l_k = [(l_1 - \delta(k-1))]$, where δ represents the gradient of changing. Note that central frequency radiated from k-domain is proportional to $1/l_k$ and, therefore higher THz frequencies are generated close to the exit surface of the crystal. Because THz absorption at higher frequencies is larger, such design of the crystal is favorable to mitigate THz-wave damping.

The result of calculated THz-pulse form for pump pulse duration of $\sigma = 0.8$ ps, the number of domains $N = 32$, $l_1 = 80$ μm, and $\delta = 1.6$ is presented in Figure 2. In the same figure we present the THz pulses forms, generated by laser pulses having delays $\Delta\tau_m = T_1 - t_1 - T_{2m-1} - t_{2m-1}$ for the cases $m = 2$ and $m = 3$. It is seen that there is temporal overlap of the fields generated from first positive domain $k = 1$ and neighboring positive domains ($k = 3$ and 5).

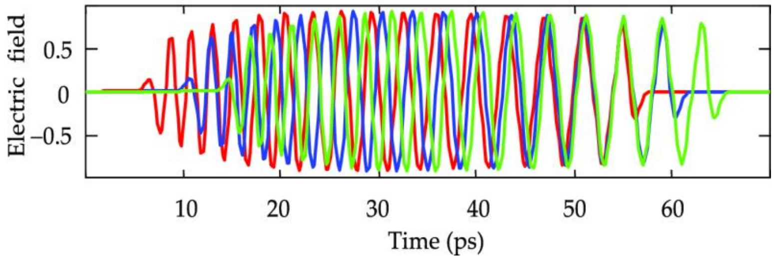

Figure 2. The temporal forms of THz pulses generated in the APPLN crystal by original laser pulse (red line) and its replicas delayed at $\Delta\tau_2$ (blue curve) and $\Delta\tau_3$ (green curve).

In Figure 3 we show the temporal forms of THz pulse generated by sequence of the laser pulses having delays $\Delta\tau_m$ ($m = 1, 2, 3\ldots N/2$) for two cases of the lengths gradient $\delta = 1.6$ and $\delta = 0.8$.

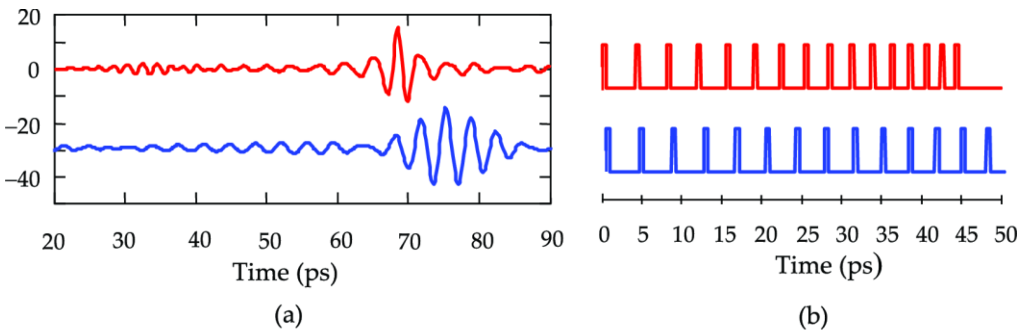

Figure 3. **(a)** Temporal forms of THz pulses generated in APPLN structures having different gradients of domain lengths $\delta = 1.6$ (red curve) and $\delta = 0.8$ (blue curve). In both cases the crystal is pumped by sequence of the laser pulses having delays $\Delta\tau_m$ with $m = 1, 2, 3,\ldots 16$. **(b)** Format of the pump pulses used for cases $\delta = 1.6$ (red line) and $\delta = 0.8$ (blue line).

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It is seen that nearly single-cycle THz pulse is formed, if sufficiently wide THz spectrum is generated in the chirped APPLN. In the case of relatively narrowband THz-wave generation $\delta = 0.8$, a few cycle THz pulses are generated with the number of the cycles depending on δ.

Let us now estimate the electric field of THz pulse generation in APPLN crystal when pulse-position modulated pump wave is applied. We start from the calculation of electric field E_1 generated by the first domain (m = 1) of the crystal. Using expansion into a Taylor series, the first term of Equation (6) can be rewritten as

$$E_1(t_R) \approx -A_0 l_1 \ddot{I}(t+T_1) \quad (7)$$

where $\ddot{I}(t)$ is the second order derivative of the pump intensity.

As it was already mentioned, the THz pulses emitted from domains having numbers $2m$ - 1 after delay at $\Delta\tau_m = T_1 - t_1 - T_{2m-1} - t_{2m-1}$ (m = 2, 3) are temporally overlapped with the pulse generated from the first domain E_1. Therefore, the electric field generated by sequence of the pump pulses having delay $\Delta\tau_m$ with m = 2, 3…N/2 can be approximately presented as $E_\Sigma(t_R) \approx NE_1(t_R)/2$. Using it, the maximal electric field strength $|E_\Sigma(t_R)|_{max}$ is estimated as 298 kV/cm, if each single optical pulse intensity in the pulse sequence is 20 GW/cm^2, the beam area is $S = \pi a^2 = 7$ mm^2, the nonlinear coefficient is d_{33} = 168 pm/V, the number of domains in APPLN crystal is N = 32, and the distance from the exit surface is R = 20 mm. This distance R well satisfies the well-known condition of the far-field approximation [26] even at lowest THz generation frequency. Obviously, the estimated value can be increased by enhancing the pump power and by focusing the THz beam with a short focus lens.

5. Conclusions

Based on radiating antenna model it is shown that chirped APPLN crystal is capable generating a nearly single-cycle THz pulse, if pumped by a sequence of femtosecond laser pulses with appropriately chosen delays between adjacent pulses. According to our estimation, peak electric field strength of 0.3 MV/cm is expected for APPLN crystal pumped by the sequence of laser pulses with peak intensity of the separate pulses about 20 GW/cm^2. By focusing the THz beam and by increasing the pump power the field strength can reach values of an order of few MV/cm. Such high-field THz radiation having spectrum centered at frequencies below 1 THz is important for many applications.

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