Simultaneous generation and compression of broadband terahertz pulses in aperiodically poled crystals

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Abstract: We introduce a technique to generate compressed broadband terahertz pulses based on *cascaded difference-frequency generation*. The approach employs a non-uniform sequence of pump pulses in aperiodically poled crystals. The pump-pulse format and poling of crystals conceived are such that the emergent terahertz pulse is already compressed. The method circumvents pump-pulse distortions that result from non-collinear approaches and the need for external compression. While capable of generating even single-cycle pulses, it is particularly efficient for the generation of pulses with few to tens-of-cycles duration. For instance, calculations accounting for cascading effects predict conversion efficiencies in the few percent range for cryogenically-cooled lithium niobate. The focused electric fields are $\gg 100$ MV/m in free space.

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1. Introduction

High-field terahertz radiation are important probes for a variety of fundamental scientific investigations [1–7]. In addition, they are considered to be invaluable to future particle accelerator technology [8–13], which could in turn transform a wide swath of applications such as X-ray generation [14].

While various methods to generate terahertz (THz) radiation, ranging from photoconductive switches [15] to gyrotrons [16] and free-electron lasers [17] exist, they have been limited in one or more of frequency, peak power or bandwidth.

With the proliferation of advanced solid-state laser technology, methods based on nonlinear frequency down-conversion of high-power infrared or near-infrared (NIR) frequencies have gained ground. Due to the large difference in terahertz and NIR photon energies, the single-photon energy-conversion efficiency of this class of approaches is expected to be low. However, repeated energy down-conversion of NIR photons or cascaded difference-frequency generation (DFG) [18] may be utilized to produce optical-to-terahertz energy conversion efficiencies exceeding the percent-level [19, 20]. In addition, laser-driven approaches offer precise synchronization possibilities, which are instrumental for spectroscopic experiments.

Despite remarkable progress, drawbacks persist with various nonlinear frequency down-conversion schemes. In organic crystals, terahertz and optical waves are close to being perfectly phase matched, purely by virtue of fortuitous material properties. Therefore, crystals shorter than the coherence length maybe utilized [20, 21] to generate broadband terahertz radiation >1 THz efficiently. However, in general, phase matching needs to be enforced by developing a suitable mechanism.

A general scheme for broadband phase matching is via the use of tilted pulse fronts [22] in non-collinear geometries. This method [23–25], amenable for broadband or single-cycle terahertz pulse generation, has produced very high conversion efficiencies and pulse energies in the <1 THz regime in lithium niobate [19, 26].

However, the approach is ultimately limited by a spatio-temporal break up of the optical pump pulse upon generating terahertz radiation at high conversion efficiencies [27–30]. This is because, at high conversion efficiencies, cascading effects produce large spectral and angular broadening of the pump. This issue may be alleviated using a superposition of beamlets [31–33] and variants thereof [34, 35]. Alternatively, attempts to eliminate the drawbacks of tilted pulse fronts in lithium niobate by utilizing smaller pulse-front tilt angles are being pursued [36, 37].

Quasi phase matching based on periodic inversion of the second-order nonlinearity [38–43] addresses the break-up issues posed by non-collinear phase-matching geometries but is only suited to the generation of narrowband terahertz pulses. Recently, we suggested the use of aperiodically poled structures [44] to generate broadband terahertz pulses. Such structures are advantageous in that they eliminate limitations posed by non-collinear phase-matching techniques in conditions where dramatic cascading occurs. They also open up the possibility of re-using the optical pump pulse in subsequent stages. However, they require an external device to compress the chirped terahertz pulses, which increases the complexity of practical implementation.

In this paper, we present an approach previously introduced by us in [45, 46], which utilizes aperiodically poled nonlinear crystals to generate broadband terahertz pulses which emerge already compressed. In this regard, work by [47] similar to that initially developed in [45, 46] may be of interest.

Our proposal entails using aperiodically poled crystals in conjunction with a non-uniform sequence of short pump pulses. Such a non-uniform sequence may in turn be generated by the superposition of a pair of long pulses. The long pulses will be required to possess different bandwidths, a relative chirp rate and will need to be spectrally separated by the desired terahertz center-frequency.

Each individual pulse in this non-uniform sequence of pump pulses shall possess a different bandwidth (or duration). As a result, each pump pulse in the sequence generates terahertz radiation centered at a different frequency.

Furthermore, each pump pulse in the sequence will be phase matched for terahertz generation at a different location in the aperiodically poled crystal.

The crystal may therefore be poled in such a way that the time delay at which a particular terahertz frequency is generated is offset by its propagation time. For example, consider a material where the terahertz group velocity is smaller than the optical group velocity. Here, the first pump pulse in the sequence will generate terahertz radiation centered at frequency f_1 at the beginning of the crystal. The time it takes for this terahertz wave centered at frequency f_1 to reach the end of the crystal is designed to be synchronized with the time of arrival of the last pump pulse in the sequence. The last pump pulse in turn generates terahertz radiation centered at frequency f_2 , which is now in phase with the terahertz wave centered at f_1 . This way, every generated terahertz frequency component remains in phase to cumulatively produce a transform-limited terahertz pulse. A more detailed step-by-step explanation is provided in Section 2 following Fig. 1.

Viewed alternately, aperiodic poling imparts a chirp onto the generated terahertz pulse. However, this chirp can be exactly compensated by the relative chirp between the two aforementioned long pulses (whose superposition produces the non-uniform sequence of short pump pulses).

The above physical description is not guaranteed to hold true in the presence of cascading effects due to the spectral broadening experienced by the pump. However, in the present work, we verify the feasibility of the approach even in the presence of cascading effects. This is a key aspect of our work. In addition, it is imperative that cascading effects be considered to accurately predict energy conversion efficiencies that exceed the single-photon conversion level (or Manley-Rowe limit).

Thus, the collinear geometry of the present approach addresses the issues of pump-pulse distortion suffered by tilted pulse fronts while circumventing the need for external compression demanded by prior proposals of aperiodic structures. The approach is also promising due to

the development of cm^2 -aperture, poled lithium niobate crystals with $< 0.1 \mu\text{m}$ control over domain periods [48]. On the other hand, the required control of poling periods in this approach are much larger, i.e. $> 1 \mu\text{m}$. Furthermore, the required pump pulses can be generated using recent developments in high-energy Ytterbium amplifiers [49].

It is worth noting that self-compressing second-harmonic generation has been previously demonstrated [50–53]. However, in contrast to second-harmonic generation where a single chirped pump pulse is sufficient, a non-uniform sequence of pump pulses is necessary for terahertz generation. Secondly, it is worth reiterating that terahertz generation with high conversion efficiency requires cascaded DFG. Such strongly phase-matched cascading effects are not considered in self-compressing second-harmonic generation systems.

It is found that the technique presented here is capable of generating even single-cycle pulses (See Fig. 2). However, it is particularly efficient for generating terahertz pulses of few to tens of cycles. As a specific example, the use of cryogenically-cooled lithium niobate at 80 K pumped by $1 \mu\text{m}$ lasers are predicted to yield optical-to-terahertz energy conversion efficiencies of a few percent with peak-field strengths of 50 MV/m for collimated beams. Similar performance with materials such as Potassium Titanyl Phosphate (KTP) may also be envisaged. The use of a long sequence of pulses spanning a ~ 100 ps full-width at half-maximum (FWHM) total duration permits pump fluences of 1 J cm^{-2} [43]. For cm^2 -aperture, poled crystals [48], this translates to pump-pulse energies of $\sim 1 \text{ J}$. For the above mentioned percent-level energy conversion efficiencies and 50 MV/m fields, this translates to terahertz pulse energies of $> 10 \text{ mJ}$ and focused field strengths in free space of $\gg 100 \text{ MV/m}$. Such high-field, few to tens-of-cycles terahertz pulses have been predicted to be useful for the acceleration of relativistic electrons [54]. In particular, such terahertz pulses could be very effective in mitigating space-charge effects. Therefore, they could produce electron bunches that are much shorter compared to those produced by terahertz pulses with hundreds of cycles [43]. Such electron bunches could in turn be disruptive for X-ray generation.

We first describe the physical principle governing the operation of the proposed method. We then proceed to analytically verify its functionality. The analysis is shown to be validated by numerical simulations. Subsequently, we present analytic expressions and perform simulations in both depleted (i.e including cascading effects) and undepleted limits to describe the behavior of the system and furnish optimal parameters.

2. Physical principle

The proposed method requires :

(i) An aperiodically poled electro-optic crystal with a second-order nonlinearity profile $\chi^{(2)}(z)$. This allows for various longitudinal positions z to be phase matched for the difference-frequency generation of different terahertz angular frequencies $\Omega_g(z)$.

(ii) A sequence of pulses with non-uniform duration, to each generate a different frequency.

The general idea is to compensate for the propagation times of terahertz radiation generated at various locations z , by the times at which their generation is initiated. This can be achieved by exploiting the difference in terahertz and optical group velocities.

Consider the typical situation when the terahertz velocity is smaller than that of the optical pump. In this case, pulses appearing earliest in the pump sequence would be phase matched for terahertz generation at the beginning of the crystal. On the other hand, pulses appearing latest in the sequence would be phase matched for terahertz generation at the end of the crystal.

The overall envelope of the optical pump would walk-off from the terahertz pulse that is generated. However, at each location z , a different pulse in the sequence would be overlapped with the terahertz pulse.

Furthermore, this pulse would generate precisely that angular frequency $\Omega_g(z)$ that is phase matched by the local poling period of $\chi^{(2)}(z)$.

Fig. 1. Schematic of outlined approach : An aperiodically poled crystal with second-order susceptibility $\chi^{(2)}(z)$ for the difference-frequency generation of different terahertz frequencies at different locations and a non-uniform sequence of pulses (blue) are required. The idea exploits the difference in optical and terahertz group velocities to offset the terahertz propagation time t_{THz} by the time at which it gets generated. (a) The pulse of duration τ_0 (green outline) is phase matched at $z = -L/2$ (green-shaded area) to generate a terahertz wavelet $E_{THz,1}$ of angular frequency $\Omega_g(z = -L/2) = 2/\tau_0$ (purple trace) at a time $t_{rel} = -L\Delta nc^{-1}/2$, relative to the center of the overall pump envelope. The terahertz wavelet born here reaches $z = 0$ at $t_{THz} = Ln_{THz}c^{-1}/2$. (b) The central pulse in the sequence with duration τ_1 (green outline) is phase matched at $z = 0$ (green-shaded area) to generate a terahertz wavelet $E_{THz,2}$ with $\Omega_g(z = 0) = 2/\tau_1$ (red trace). The previously generated terahertz radiation has reached $z = 0$ at precisely the same time as the newly generated trace, leading to coherent growth of the total field $E_{THz,total}$ (black-dashed). (c) The pulse with duration τ_2 is phase matched at the end of the crystal. While the overall pump envelope slips through the terahertz field, there is always a pulse in the sequence which is overlapped with the terahertz field. This pulse generates a terahertz angular frequency $\Omega_g(z)$ that is precisely phase-matched at z . The length of the crystal here is arbitrary, hence the terahertz pulse remains compressed throughout.

2.1. Obtaining $t_{rel}(z)$

The problem at hand is therefore to obtain expressions for $\Omega_g(z)$ and $\chi^{(2)}(z)$. This is done by first determining $t_{rel}(z)$ - the time relative to the center of the pump envelope, at which the pulse in the pump sequence, phase matched for terahertz generation at z arrives.

We proceed by examining Fig. 1. Here, snapshots of the aperiodically poled crystal between $z = \mp L/2$, the optical pump sequence (blue) and generated terahertz wavelets at different time instants t_{op} are depicted.

The time t_{op} is referenced to the center/peak of the overall pump envelope. For instance, when $t_{op} = 0$, the peak of the pump envelope is located at $z = -L/2$.

In Fig. 1(a), the first pulse in the sequence (green-outline) is located at $z = -L/2$. If this pulse occurs at a time $t_{rel}(-L/2)$ relative to the center of the envelope, then based on our prior definition, $t_{op} = t_{rel}(-L/2)$. The pulse (green-outline) has a duration τ_0 that is phase matched for the generation of terahertz radiation with angular frequency $\Omega_g = 2/\tau_0$ at $z = -L/2$ (green-shaded area). Difference-frequency generation produces a terahertz wavelet $E_{THz,1}$ (purple) as depicted below the crystal. The generated wavelet takes a time $t_{THz} = Ln_{THz}c^{-1}/2$ to reach the center of the crystal at $z = 0$. Note, that the choice of the crystal center (i.e. $z = 0$) is arbitrary and is only for illustrative purposes. We shall later find that the terahertz pulse remains compressed throughout.

In Fig. 1(b), the central pulse in the sequence of duration τ_1 (green-outline) with $t_{rel} = 0$ is depicted. It is phase matched (green-shaded area) for the generation of the corresponding angular frequency $\Omega_g = 2/\tau_1$ at $z = 0$.

For coherent growth of terahertz radiation to occur, the terahertz wavelet generated in Fig. 1(a) has to now overlap precisely with the central pulse in the envelope. This would require the time Δt_{op} elapsed between Figs. 1(a) and 1(b) to be the same as the time taken for the terahertz radiation to reach $z = 0$ from $z = -L/2$.

Thus, we have the condition that $\Delta t_{op} = Ln_g/(2c) - t_{rel}(-L/2) = Ln_{THz}/(2c)$. This yields $t_{rel}(-L/2) = -L\Delta n/(2c)$, where $\Delta n = n_{THz} - n_g$ is the difference between terahertz and optical group refractive indices.

Upon satisfying this requirement, we see in Fig. 1(b) that the terahertz wavelet $E_{THz,2}$ (red) generated at $z = 0$ overlaps precisely with $E_{THz,1}$ (purple) from Fig. 1(a) to produce coherent growth of the total field $E_{THz,total}$ (black-dashed).

Extending the above argument to general z , we obtain the following constraint for $t_{rel}(z)$ in Eq. (1).

$$\frac{(L/2 + z)n_{THz}}{c} = \frac{(L/2 + z)n_g}{c} - t_{rel}(-L/2) + t_{rel}(z) \quad (1)$$

Substituting $t_{rel}(-L/2) = -L\Delta n/(2c)$ in Eq. (1) as deduced above, we obtain the following expression for $t_{rel}(z)$ below.

$$t_{rel}(z) = \frac{z(n_{THz} - n_g)}{c} = \frac{z\Delta n}{c} \quad (2)$$

The validity of Eq. (2) is seen in Fig. 1(c) which shows the optical pulse in the pump sequence with duration τ_2 (green-outline) with $t_{rel} = +L\Delta n/(2c)$. This is phase matched (green-shaded area) for $\Omega_g = 2/\tau_2$ and the generated $E_{THz,3}$ (yellow) overlaps with the previously generated terahertz radiation at $z = L/2$. The total terahertz field has continued to grow between Figs. 1(a)–1(c), as evident in the superposition of individual wavelets $E_{THz,total}$ (black-dashed).

Thus, it has been described how a compressed terahertz pulse may be generated using the proposed method.

2.2. Generating a non-uniform sequence of pulses

Now, we prescribe a method to generate the desired non-uniform sequence of pulses. The overall pump field described by the electric field phasor $E(t)$ in time t can be generated by the superposition of two pulses $E_1(t), E_2(t)$ as shown in Eq. (3a).

In Eq. (3b), $E_1(t)$ represents a pulse centered at angular frequency ω_1 with transform-limited duration τ_{TL} , chirped at a rate $b = (\tau\tau_{TL})^{-1}$ to a duration $\tau \gg \tau_{TL}$. On the other hand, from Eq. (3c), $E_2(t)$ represents a pulse with transform-limited duration τ , centered at angular frequency ω_2 . $E_1(t)$, $E_2(t)$ and their corresponding spectra $E_1(\omega)$, $E_2(\omega)$ in angular frequency ω are delineated in Eqs. (3b) and (3c) respectively.

$$E(t) = E_1(t) + E_2(t) \quad (3a)$$

$$E_1(t) = E_1 e^{j\omega_1 t} e^{jbt^2} e^{-t^2/\tau^2} \quad (3b)$$

$$E_1(\omega) = \frac{E_1 e^{-jb^{-1}(\omega-\omega_1)^2} e^{-\frac{(\omega-\omega_1)^2 \tau_{TL}^2}{4}}}{2\sqrt{\pi}(\tau^{-2} + jb)}$$

$$b = (\tau\tau_{TL})^{-1}$$

$$E_2(t) = E_2 e^{j\omega_2 t} e^{-t^2/\tau^2} \quad (3c)$$

$$E_2(\omega) = \frac{\tau E_2 e^{-\frac{(\omega-\omega_2)^2 \tau^2}{4}}}{2\sqrt{\pi}}$$

From Eqs. (3b) and (3c), it is clear that $\Omega_0 = \omega_1 - \omega_2$ is defined as the central terahertz angular frequency, while $f_0 = \Omega_0/(2\pi)$ is the central terahertz frequency.

It can be seen that the overall intensity profile $|E(t)|^2 \propto \cos^2[(\Omega_0 + bt)t/2]$. This corresponds to a spacing between pulses given by $\approx T_0(1 - bt/\Omega_0)$ (for $t \ll \tau$) and pulse duration $\approx T_0(1 - bt/\Omega_0)/2$, where $T_0 = 2\pi/\Omega_0$ is the time period of the central terahertz frequency. Thus it is clear that for $b > 0$, earlier pulses ($t < 0$) in the sequence are longer and spaced more sparsely.

2.3. Obtaining $\Omega_g(z)$

Since the generated terahertz frequency is merely the difference of the instantaneous frequencies of the two pulses, it may be deduced that $\Omega_g = \omega_1 + bt - \omega_2 = \Omega_0 + bt$. However, at a given z , only terahertz frequencies corresponding to $t_{rel}(z)$ from Eq. (2) shall be phase matched, which yields the relationship for $\Omega_g(z)$ in Eq. (4).

$$\Omega_g(z) = \Omega_0 + \frac{bz\Delta n}{c} \quad (4)$$

2.4. Poling profile

Having established the spatial distribution of terahertz frequencies by Eq. (4), it is straightforward to obtain the corresponding poling profile. By analogy to a periodically poled crystal, the required aperiodic poling profile to generate $\Omega_g(z)$ is given by Eq. (5).

$$\chi^{(2)}(z) = \chi_b^{(2)} \text{sgn} [\cos(\Omega_g(z)\Delta n z c^{-1})] \quad (5)$$

In Eq. (5), $\chi_b^{(2)}$ is the second-order nonlinear bulk susceptibility and **sgn** represents the signum function.

3. Analysis

3.1. Proof of functionality

In this section, we analytically show that the proposed approach indeed functions as suggested. A solution for the terahertz spectral component $E_{THz}(\Omega, z)$ at angular frequency Ω is obtained from the nonlinear wave equation by assuming a solution of the form $E_{THz}(\Omega, z) = A_{THz}(\Omega, z)e^{-j\Omega n_{THz} z c^{-1}}$. The evolution of the terahertz spectral envelope $A_{THz}(\Omega, z)$ in the undepleted limit then follows [43] and is reproduced in Eq. (6) below.

$$\frac{dA_{THz}(\Omega, z)}{dz} = \frac{-j\Omega}{2cn_{THz}} \chi^{(2)}(z) e^{-j\Delta k z} \mathfrak{F}(|E(t, z)|^2) - \frac{\alpha(\Omega)}{2} A_{THz}(\Omega, z) \quad (6)$$

In Eq. (6), $\Delta k = -\Delta n \Omega c^{-1}$ represents the phase mismatch between the generated terahertz radiation and incident optical pump. The total pump field $E(t)$ is obtained from Eqs. (3a)-(3c). The term $\mathfrak{F}(|E(t)|^2)$ in Eq. (6) corresponds to the spectral distribution of the nonlinear polarization due to various difference-frequency generation processes. This term is found in previous models such as [43, 44, 55]. The final term in Eq. (6) corresponds to terahertz absorption with $\alpha(\Omega)$ being the terahertz absorption coefficient.

Upon integrating Eq. (6) over longitudinal distance z , we obtain the following expression for $A_{THz}(\Omega, z)$ in Eq. (7).

$$A_{THz}(\Omega, z) = \frac{-j\Omega \mathfrak{F}(|E(t)|^2) e^{-\alpha z/2} \int_{-L/2}^z e^{(-j\Delta k + \alpha/2)z'} \chi^{(2)}(z') dz'}{2cn_{THz}} \quad (7)$$

In order to obtain a closed-form expression for $A_{THz}(\Omega, L/2)$ from Eq. (7) that illustrates the functionality of the approach, a few simplifying assumptions are made. These are without the loss of generality as validated later by detailed numerical calculations. First, absorption in Eq. (7) is ignored. Next, the expression for $\chi^{(2)}(z)$ from Eq. (5) is approximated as $\chi^{(2)}(z) \approx \chi_b^{(2)} e^{-j\Omega_g \Delta n c^{-1} z}$, where $\Omega_g(z)$ is given by Eq. (4). In addition, the limits of integration in Eq. (7) maybe set to $[-\infty, \infty]$. This approximation maybe justified by the fact that contributions outside the interval $[-L/2, L/2]$ are less significant due to the finite bandwidth of the pump pulse. Upon explicit evaluation of $\mathfrak{F}(|E(t)|^2)$ using Eqs. (3a)-(3c), we obtain the following closed-form expression for $A_{THz}(\Omega, L/2)$ in Eq. (8) below.

$$A_{THz}(\Omega, L/2) = \frac{\Omega \chi_b^{(2)} E_1 E_2 e^{-\frac{(\Omega - \Omega_0)^2 \tau_L^2}{2(4\tau_L^2 \tau^{-2} + 1)}}}{4b\Delta n n_{THz}} \left(e^{-\frac{j b(\Omega - \Omega_0)^2}{4(b^2 + 4\tau^{-4})}} e^{\frac{j(\Omega - \Omega_0)^2}{4b}} \right) \quad (8)$$

Inspecting the first exponential term in Eq. (8), we notice that $A_{THz}(\Omega, L/2)$ is centered about the terahertz angular frequency Ω_0 with bandwidth proportional to $(8\tau^{-2} + 2\tau_L^{-2})^{1/2}$. For the case when the chirped-pulse duration is much greater than the transform-limited duration, i.e. $\tau \gg \tau_L$, the terahertz spectral bandwidth is proportional to τ_L^{-1} . This corresponds to the range of possible beat frequencies between the two input pulses and is consistent with expectations.

The terms enclosed within the round brackets in Eq. (8) correspond to the various phase terms that accrue. The first phase term arises from the difference-frequency generation process, while the second term arises from the aperiodic poling profile. Notice that the two terms are of opposite signs, indicative of phase compensation.

Fig. 2. Numerical simulations of Eq. (6) of a non-uniform sequence of pulses (blue) with parameters $\tau = 150$ ps, $\tau_{TL} = 0.5$ ps and $f_0 = 0.5$ THz in an $L=2$ cm lithium niobate crystal with poling profile given by Eq. (5) at $T=80$ K. The poling profile (black) possesses a gradually reducing period from $910 \mu\text{m}$ to $100 \mu\text{m}$. (a) The lower terahertz frequencies are generated earlier, at the beginning of the crystal to minimize absorption. The overall pump envelope walks ahead of the single-cycle terahertz transient (red) due to larger group velocity. (b)–(d) The poling profile is arranged such that the terahertz frequencies are phase matched precisely at locations where the corresponding pulses in the sequence (blue) overlap with the terahertz transient (red) to produce coherent growth in the terahertz field and bandwidth.

Given that the chirp rate $b = (\tau\tau_{TL})^{-1}$ (Eq. (3b)) and that the chirped pulse duration $\tau \gg \tau_{TL}$, the $e^{\frac{-jb(\Omega-\Omega_0)^2}{4(b^2+4\tau^{-4})}}$ term is almost perfectly compensated by the $e^{\frac{jb(\Omega-\Omega_0)^2}{4b^2}}$ term. In fact, one may achieve exact compression by adjusting the chirp rate to b' defined by Eq. (9).

$$b' = b\sqrt{1 + 4\tau^{-4}b^{-2}} \quad (9)$$

Thus, it has been analytically shown in Eq. (8) that the proposed approach produces a perfectly compressed terahertz pulse in the ideal limit.

3.1.1. Numerical validation

To validate our analysis, numerical simulations employing Eq. (6) without approximations are performed for a pump pulse $E(t)$ defined by Eqs. (3a)–(3c) with parameters $\tau_{TL} = 0.5$ ps, $\tau = 150$ ps and $f_0 = 0.5$ THz. The complete material dispersion in the NIR frequency range [56], terahertz frequency range [57] and terahertz absorption [58] at $T = 80$ K were taken into account. The bulk second-order nonlinear susceptibility was assumed to be $\chi_b^{(2)} = 336$ pm/V. An $L = 2$ cm long lithium niobate crystal with a poling profile given by Eq. (5) is assumed. The poling period varies from $\Lambda = 910 \mu\text{m}$ at the beginning of the crystal to $\Lambda = 100 \mu\text{m}$ at the end of the crystal. The results are presented in Figs. 2(a)–2(d). The lowest terahertz frequencies are phase matched at the beginning of the crystal, evident by the larger poling periods (black), to minimize absorption. The long pulses in the pump sequence (blue) arrive first. They generate the low terahertz frequencies and walk-off from the generated terahertz transient (red) in Fig. 2(a) since $n_{THz} > n_g$ in lithium niobate. However, the terahertz transient is always synchronized to a latter pulse in the pump sequence. These generate terahertz frequencies $\Omega_g(z)$, which also happen to be phase matched at the same location as the terahertz transient. Thus, the terahertz transient continues to grow in strength and reduce in duration in Figs. 2(b)–2(d). Thus, a compressed single-cycle terahertz transient can be generated in a manner described at the outset.

3.2. Length and frequency behavior

In Eqs. (10a)–(10c), an analytic solution to Eq. (6) which accounts for the finite length of the crystal, absorption and terahertz dispersion is furnished using imaginary error functions (**erfi**). In comparison to a complete numerical solution of Eq. (6), these expressions only account for the principal harmonic components of the aperiodic poling profile, i.e. $\chi^{(2)}(z) \approx \chi_b^{(2)} e^{-j\Omega_g \Delta n c^{-1} z}$.

$$A_{THz}(\Omega, z) = \frac{\Omega \chi_b^{(2)} E_1 E_2 e^{-\frac{(\Omega - \Omega_0) \tau_{TL}^2}{2(4\tau_{TL}^2 \tau^{-2} + 1)}}}{8\sqrt{bb'} \Delta n n_{THz}} \left(e^{-\frac{jb(\Omega - \Omega_0)^2}{4(b^2 + 4\tau^{-4})}} e^{\frac{j(\Omega - \Omega_0)^2}{4b'}} \right) G(\gamma, \theta) \quad (10a)$$

$$G(\gamma, \theta) = e^{-\gamma\theta/2} \left(\text{erfi} \left[\frac{\sqrt{j}}{2} (j\gamma - \theta) \right] - \text{erfi} \left[\frac{\sqrt{j}}{2} (j\gamma|_{z=-\frac{L}{2}} - \theta) \right] \right) \quad (10b)$$

$$\gamma = \frac{2z}{z_0} - \frac{\Omega - \Omega_0}{b'^{1/2}}, \quad \theta = \frac{\alpha z_0}{2}, \quad z_0 = \frac{c}{b'^{1/2} \Delta n} \quad (10c)$$

In Eq. (10a), terms outside the round brackets are similar to corresponding terms from Eq. (8). The phase terms within brackets correspond to those from the difference-frequency generation process and aperiodic poling profile respectively. As already shown, these tend to compensate each other. The dependence of the terahertz spectral component $A_{THz}(\Omega, z)$ on length z , detuning $\Omega - \Omega_0$ and terahertz absorption coefficient $\alpha(\Omega)$ are captured by the function $G(\gamma, \theta)$ which is presented in Eq. (10b). In Eq. (10b), the prefactor $e^{-\gamma\theta/2}$ corresponds to the loss term. The normalized variables γ, θ and z_0 are in turn defined in Eq. (10c). Note that γ captures propagation and bandwidth effects, θ accounts for absorption and z_0 is a normalized distance parameter. Based on the properties of **erfi** functions, it can be readily shown that $|G| \leq 2$. In this limit, Eq. (10a) becomes identical to Eq. (8).

We plot $G(\gamma, \theta)$ from Eq. (10b) in Fig. 3 in the limit when $-L/2 \rightarrow -\infty$. This enables us to study key features of $A_{THz}(\Omega, z)$. Later on it will be shown that Eqs. (10a)–(10c) are quantitatively very accurate (For e.g., see Fig. 5).

From Fig. 3, it can be seen that the behavior of $|G|$ is step-like with its magnitude abruptly increasing with γ before saturating. However, since γ is proportional to z/z_0 (from Eq. (10c)), it implies that z_0 may be viewed as a critical-length parameter beyond which $|A_{THz}(\Omega, z)|$ becomes significant. From Eq. (3b) and Eq. (10c), $z_0 \approx c\Delta n^{-1} \sqrt{\tau \tau_{TL}}$. Therefore, optimal interaction lengths are expected to be shorter for smaller τ, τ_{TL} . These deductions are in agreement with the numerical simulations in the next section (See Fig. 4(d)).

Next, in the inset (i) of Fig. 3, we plot the phase $\angle G$. It can be seen that both magnitude and phase of G show oscillations for small values of γ , before stabilizing at larger values. Since γ is also a function of frequency, ripples in $|G|$ and $\angle G$ translate to ripples in the spectral magnitude and phase of $A_{THz}(\Omega, z)$.

Finally, for the same z, γ values decrease with increasing Ω . Therefore, in conjunction with the prefactor Ω in Eq. (10a), the spectral magnitude $A_{THz}(\Omega, z)$ can be expected to show a ramp-like shape, i.e. increasing with frequency for $\Omega < \Omega_0$ and then rapidly falling off for $\Omega > \Omega_0$. This can be confirmed by inspecting the terahertz spectra in Fig. 5.

3.3. Efficiency trends

The conversion efficiency η is obtained in general using Eq. (11a). It was previously shown that the upper limit of $|A_{THz}(\Omega, z)|$ from Eq. (10a) is given by Eq. (8). Employing this fact, an

Fig. 3. Magnitude and phase of G (Eq. (10b)) with $-L/2 \rightarrow -\infty$ as a function of normalized variables γ , θ and z_0 (Eq. (10c)) which simultaneously account for the effects of τ_{TL} , τ , propagation distance z and absorption coefficient $\alpha(\Omega)$. The phase and magnitude oscillations in G manifest as spectral magnitude and phase oscillations in $A_{THz}(\Omega, z)$.

upper-bound for η based on an undepleted approximation is presented in Eq. (11b).

$$\eta(z) = \pi c \epsilon_0 \int_0^\infty n_{THz} |A_{THz}(\Omega, z)|^2 d\Omega / F_{pump} \quad (11a)$$

$$\eta \leq \frac{\pi \chi_b^2 F_{pump} \Omega_0^2 \tau_{TL}}{2\sqrt{2} c \epsilon_0 n_{THz} \Delta n^2 n_g^2} \quad (11b)$$

From Eq. (11b), the proportionality of η to τ_{TL} delineates that the conversion efficiency is inversely proportional to the generated terahertz bandwidth. This is understandable given that a larger bandwidth, when apportioned over the same length, translates to less interaction length per generated terahertz frequency. Thus, it is presumable that the conversion efficiency that is generated from such structures would be lesser than that of multi-cycle generation with periodically-poled structures. On the other hand, it can be seen that despite this limitation, the conversion efficiency for moderate terahertz pulse durations breaches the percent level. Furthermore, the conversion efficiency is inversely proportional to the difference in terahertz phase and optical group refractive index Δn . In light of this, KTP with smaller values of Δn and potentially larger damage fluence thresholds may offer similar if not superior performance in relation to lithium niobate.

4. Simulation results

In this section, we present simulation results incorporating the full effects of dispersion, absorption, exact spatial variation of $\chi^{(2)}(z)$ (Eq. (5)) as well as pump depletion (or cascading effects). We first obtain optimal parameters via a numerical solution in Eq. (6) in the undepleted limit

Fig. 4. (a) Conversion efficiency η as a function of chirped-pulse duration τ and transform-limited duration τ_{TL} for 0.5 THz in lithium niobate at $T=80$ K. Increasing η for narrower terahertz bandwidths is predicted, reaching about 4% for $\tau_{TL}=50$ ps. (b) Corresponding electric field strengths in free space as a function of τ , τ_{TL} , showing collimated field strengths on the order of 50 MV/m. Focused field strengths are then expected to be $\gg 100$ MV/m for $\sim \text{cm}^2$ crystal apertures. (c) Optimal η and peak electric field strengths as a function of terahertz frequency, depicting an increasing trend and proximity of the peak fields to the corresponding transform-limited values. The optimal values occur for $\tau_{TL}=50$ ps and $\tau \approx 2\Delta n c^{-1} \alpha^{-1} \approx 150$ ps. (d) Optimal interaction lengths decrease with frequency, in agreement with increasing absorption coefficients with terahertz frequency.

and then verify functionality of the approach in the presence of cascading effects (or depleted calculations). Conversion efficiencies η are evaluated using Eq. (11a). While the simulation results are carried out for cryogenically-cooled lithium niobate ($T = 80$ K), the trends can be considered to apply to other material systems. Once the pump field is defined by Eqs. (3a)–(3c), the poling profile is uniquely defined by Eq. (5). The bulk second-order nonlinear susceptibility is assumed to be $\chi_b^{(2)} = 336$ pm/V. The material dispersion in the NIR region is obtained from [56]. The material dispersion for lithium niobate in the terahertz region is obtained from [57] while terahertz absorption is obtained from [58]. The total pump fluence that may be impinging on the crystals, is assumed to be limited by laser-induced damage according to the following expression for fluence, $F_{\text{pump}} = 0.85(\tau/100\text{ps})^{0.5} \text{ Jcm}^{-2}$ [43], where τ is the e^{-2} pulse duration.

4.1. Efficiency and peak fields

We study the dependence of the conversion efficiency (η) and peak electric field (E_{max}) on the pump parameters τ , τ_{TL} for $f_0 = 0.5$ THz in Figs. 4(a) and 4(b) respectively.

Firstly, in Fig. 4(a), in line with Eq. (11b), we see an increase in conversion efficiency η with increasing τ_{TL} . As previously discussed, this may be understood by noting that when the same length is apportioned over a larger bandwidth, the coherent build up of each terahertz spectral component is limited. Therefore for small τ_{TL} , η is limited to a fraction of a percent but for larger τ_{TL} , the conversion efficiencies breach the percent mark comfortably. However, contrary to Eq. (11b), the increase in conversion efficiency with τ_{TL} is not linear as Eq. (11b) represents the ideal limit.

There also exists an optimal value of τ for a given τ_{TL} in Fig. 4(a). This may be understood as

follows. Firstly, the bandwidth of the generated terahertz pulse is limited by τ_{TL}^{-1} , or the range of beat frequencies between the two optical pump pulses. The length of the aperiodically poled crystal is optimized when the phase-matched bandwidth is similar to the bandwidth of the pump pulse. Based on Eq. (4), this translates to $b\Delta nc^{-1}L_{opt} \approx \tau_{TL}^{-1}$. However, since $b = (\tau\tau_{TL})^{-1}$ from Eq. (3b), one obtains a condition for the optimal crystal length as $L_{opt} \approx c\tau\Delta n^{-1}$ for a given chirped-pulse duration τ .

The maximum interaction length is in turn limited by the terahertz absorption coefficient. Thus, $L_{opt} \approx 2\alpha^{-1}$ and consequently $\tau_{opt} \approx 2\alpha^{-1}\Delta nc^{-1}$. As the pump bandwidth τ_{TL} increases, the absorption length reduces by virtue of spanning larger frequencies and hence both the optimal chirped-pulse duration and crystal length drop. The first trend is evident in Fig. 4(a).

In Fig. 4(b), the peak electric field strengths as a function of τ, τ_{TL} are plotted. Since the emergent terahertz pulses are compressed, the peak-field strengths are proportional to η/τ_{TL} . Therefore, for small τ_{TL} , the peak electric field strengths are large despite low conversion efficiencies. After a minor drop for intermediate values of τ_{TL} , the peak-field strengths tend to stabilize or saturate. The maximum field strengths for collimated beams in free space are on the order of 50 MV/m. However, if one uses cm^2 -aperture crystals, focused field strengths of several hundred MV/m are indeed feasible.

Figure 4(c) delineates the global optima of conversion efficiencies and peak-field strengths (w.r.t to parameters τ, τ_{TL}) for various central terahertz frequencies f_0 . Figure 4(d) depicts the corresponding optimal crystal lengths. Based on the trends in Fig. 4(a), the optimal value of τ_{TL} occurs in the many-cycle limit, i.e. for $\tau_{TL} = 50$ ps or at the edge of the parameter space used for τ_{TL} . The corresponding optimal value of $\tau \approx 2\alpha^{-1}\Delta nc^{-1} \approx 150$ ps.

In Fig. 4(c), the conversion efficiency (green) shows an increasing trend with frequency, albeit only linearly and not quadratically owing to the effects of increasing absorption with terahertz frequency. The maximum electric field strengths (black) also increase with frequency and are very close to the transform-limited values (black-dashed), further depicting the efficacy of the present mechanism in achieving compression. Figure 4(d) depicts the optimal interaction lengths as a function of frequency. Here, the trends of decreasing optimal lengths are consistent with increasing absorption coefficients with terahertz frequency.

4.2. Spectra, phase and temporal formats

In this section, we present terahertz spectra, phase and temporal electric field traces for various values of τ_{TL} . In each case, the terahertz frequency was 0.5 THz for optimal values of $\tau \approx 150$ ps obtained from the previous section.

In Fig. 5(a), we present the spectrum for $\tau_{TL} = 0.5$ ps. The spectral profile in Fig. 5(a) is clearly broadband spanning 0.2 to 0.8 THz. The increasing ramp-like shape and ripples are in line with the discussion following Fig. 3 as seen in the analytic calculations (red-dashed) from Eq. (10a). Furthermore, the analytic calculations from Eqs. (10a)–(10c) are in agreement with numerical simulations based on Eq. (6).

In Fig. 5(b), the group delay (GD) of the terahertz pulse is plotted as obtained from analysis (Eqs. (10a–10c)) as well as simulations (Eq. (6)). In addition, we plot the uncompensated group delay $(\Omega - \Omega_0)/(2b + 8\tau^{-4}/b)$ (See derivative of the first phase term within brackets of Eq. (8)) to delineate the extent of phase compensation. As evident from Fig. 5(b), the linear group delay (black-dashed) has been roughly flattened (save for ripples, which are expected based on Eq. (10a)).

As a result, in Fig. 5(c), we find that the transform-limited terahertz transient (red) is very close to the output from the structure (green) and constitutes a single-cycle terahertz pulse with a duration proportional to $\tau_{TL} = 0.5$ ps. Similarly, Figs. 5(d)–5(f) and Figs. 5(g)–5(i) represent the corresponding properties for $\tau_{TL} = 4$ ps and $\tau_{TL} = 25$ ps respectively, which both produce terahertz pulses (green) close to transform-limited values (red).

Fig. 5. Terahertz spectra and group delay (GD) obtained from simulations (Eq. (6), green) and analytic calculations (Eq. (10a), red-dashed) along with electric field output traces obtained from simulations only (green) and corresponding transform-limited values (red-solid). The uncompensated GD due to the relative chirp between pump pulses is also shown (black-dashed). For (a)-(c) $\tau_{TL} = 0.5$ ps, (d)-(f) $\tau_{TL} = 4$ ps and for (g)-(i) $\tau_{TL} = 25$ ps. In all cases, the generated pulses are very close to the transform-limited output. The ripples in phase and spectra may be rectified by apodization.

4.3. Cascading effects

In this section, we resort to simulations in the depleted limit, i.e. including cascading effects based on the numerical model presented in [43]. The calculations consider cascading effects, full material dispersion in the NIR frequency range [56], terahertz frequency range [57] as well as terahertz absorption [58] at $T = 80$ K. The remaining parameters are the same as in previous sections and are taken from [43]. We find that despite drastic spectral broadening of the pump, the efficacy of compression is still maintained and efficiencies are in agreement with calculations in previous sections.

In Fig. 6(a), the optical spectrum corresponding to two pulses separated by a frequency of 0.5 THz, with parameters $\tau_{TL} = 25$ ps, and $\tau = 150$ ps is depicted. The choice of these parameters is based on the fact that the expected conversion efficiencies fall in a regime where cascading effects are expected to be significant due to large values of η (See Fig. 4(a)). For shorter τ_{TL} , the similarity to undepleted calculations would increase owing to lower expectations of conversion efficiency (See Fig. 4(a)). The extreme values of the poling period for this structure range between $241.8 \mu\text{m}$ at the beginning of the crystal to $238.3 \mu\text{m}$ at the end. This is within the tolerance offered by current technology [48], which is $< 0.1 \mu\text{m}$.

Since the energy in the narrowband pulse and broadband chirped pulse are equal, their relative strengths in the spectral domain are dissimilar as is evident from the inset on the right in Fig. 6(a). Note, how this inset shows differing linewidths. The inset on the left plots conversion efficiency versus length and shows that the conversion efficiency is similar, albeit higher compared to that obtained via undepleted calculations. The higher conversion efficiency of 4% is due to the increase in pump intensity resulting from dramatic spectral broadening.

In Figs. 6(b)–6(d), we observe a significant red-shift of the optical spectrum due to cascading effects, which is responsible for the high conversion efficiency in the first place. However, due to

Fig. 6. Evolution of the optical pump spectrum as a function of distance. (a) Two 'lines' separated by 0.5 THz (see inset on right), one of which is broader due to a linewidth defined by $\tau_{TL} = 25$ ps. The inset on the left shows conversion efficiency approaching 4 % at $T = 80$ K. The efficiency is larger than the corresponding undepleted simulation due to an increase in optical pulse intensity which arises from spectral broadening. (b)–(d) A continuous red-shift is obtained due to the beating of the lines. This red-shift is responsible for the high conversion efficiencies that are observed. Blue-shift is also obtained since sum and difference frequency processes are both phase matched due to the small terahertz frequency.

the collinear geometry of this system, a spatio-temporal break-up of the pulse does not occur. This is contrary to the case of tilted pulse fronts.

Since, the strength of the two input lines differ, the situation is similar to that of a cascaded terahertz parametric amplifier [59]. Comparing Figs. 6(a)–6(d), the spectrum shows an increasing red-shift with distance, accompanied also with some blue-shift due to symmetry in the phase matching of sum and difference frequency generation that results from the small value of the terahertz frequency. This is similar to that observed in [59, 60].

In Fig. 7(a), we plot the simulated terahertz field output and compare it to the transform-limited terahertz field (obtained by assuming a flat phase across the simulated spectrum). The peak-field strength of the transform-limited field (red) is about 75 MV/m (slightly larger than the transform-limited value of 60 MV/m in Fig. 5(i) due to the increase in terahertz efficiency by cascading effects), while the output from the structure (green) is close but smaller at 55 MV/m (similar to peak-field strengths obtained from undepleted calculations in Fig. 5(i)). The duration of the two pulses are quite similar, indicating effective compression.

Figure 7(b) depicts the group delay obtained numerically while considering cascading effects (green) and analytically under undepleted approximations (Eq. (10a), red-dashed) in comparison to the uncompensated group delay of the pump pulse input (black-dashed). Clearly, the chirp on the pump input has been significantly reduced by the structure (shown by brown arrows) as evident in both simulations and analytic calculations. However, for frequencies > 0.502 THz, the undepleted calculations suggest better compression.

However, since most of the energy in the spectrum is contained within the region where undepleted and depleted calculations agree (red, inset of Fig. 7(b)), effective compression is nevertheless obtained. To delineate how broadband the generated terahertz spectrum is, we also plot the spectrum of a narrowband terahertz pulse corresponding to $\tau = 150$ ps (blue) in the inset.

The difference in group delay between depleted and undepleted calculations is due to the

Fig. 7. (a) Traces of the terahertz electric field obtained from depleted simulations accounting for cascading (green) and the corresponding transform-limited terahertz pulse (red). The output from the structure is similar to the transform-limited value and has a peak-field strength similar to that predicted from undepleted calculations. (b) Group delay of compressed pulses obtained from depleted simulations (green) and undepleted analysis (red-dashed) show effects of phase compensation in relation to the chirp present in the pump input (black-dashed). The analytic calculations show superior compression for $> .502$ THz, due to the effects of dispersion arising from cascading. Most of the generated spectrum (inset, red-solid) lies in a range where the undepleted and depleted calculations are in agreement, which results in effective compression. The spectrum of a narrowband pulse corresponding to $\tau = 150$ ps (inset, blue-solid) is shown to delineate the large bandwidth of the generated terahertz pulse. For lower η , cascading effects would be less important and the efficacy of compression would approach that predicted by undepleted calculations.

change in optical group refractive index encountered by the broadened pump spectrum. For such broadband spectra, higher-order dispersion is also significant. This renders the generated poling profile less effective. Since, the extent of broadening is efficiency dependent, the disparity between depleted and undepleted calculations can be expected to reduce for more broadband terahertz generation (e.g Fig. 5(a)), where the efficiencies are anticipated to be lower.

In the high efficiency regime delineated in Figs. 6 and 7, the compression may be further improved by numerical optimization of the poling profile. This could also be useful in mitigating effects of higher-order dispersion and would be the scope of future work.

In any case, it is worthwhile to note that the poling profile conceived analytically in the absence of depletion is highly effective, even in the presence of cascading effects, producing conversion efficiencies $\approx 4\%$. This also suggests that the device may be robust to spurious higher-order dispersion.

5. Conclusion

In conclusion, a new method to generate broadband terahertz pulses using a combination of a non-uniform sequence of pulses and aperiodically poled crystals was shown to produce compressed pulses with large peak-field strengths and high efficiencies. The non-uniform sequence of pulses could be generated by the superposition of a chirped and transform-limited pulse separated by the desired terahertz frequency. The approach is particularly efficacious for generating few to tens-of-cycles terahertz pulses, yielding conversion efficiencies of $\approx 4\%$. Analysis in the undepleted limit along with simulations in undepleted and depleted limits were presented. The simulations validated the efficacy of the approach. In the presence of dramatic cascading, the efficacy of compression only reduces marginally but this may be further improved by numerical optimization of the poling profile. The presented work could potentially be of high value for terahertz-driven electron acceleration and coherent X-ray generation.

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