Optical generation of narrow-band terahertz packets in periodically-inverted electro-optic crystals: conversion efficiency and optimal laser pulse format

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Abstract: We explore optical-to-terahertz conversion efficiencies which can be achieved with femto- and picosecond optical pulses in electro-optic crystals with periodically inverted sign of second-order susceptibility. Optimal crystal lengths, pulse durations, pulse formats and focusing are regarded. We show that for sufficiently short (femtosecond) optical pulses, with a pulsewidth much shorter than the inverse terahertz frequency, conversion efficiency does not depend on pulse duration. We also show that by mixing two picosecond pulses (bandwidth-limited or chirped), one can achieve conversion efficiency, which is the same as in the case of femtosecond pulse with the same pulse energy. Additionally, when the group velocity dispersion of optical pulses is small, one can substantially exceed Manley –Rowe conversion limit due to cascaded processes.

OCIS codes: (190.2620) Frequency Conversion; (190.7110) Ultrafast nonlinear optics; (260.3090) Physical optics, Infrared, far.

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1. Introduction

Optical rectification (OR) of ultrashort laser pulses is an established way of generating broad-band terahetz (THz) radiation. THz output in this case is produced in an electro-optic (EO) medium via difference-frequency mixing between Fourier components of the same optical pulse. First demonstrated with picosecond pulses in ZnTe, ZnSe, CdS and quartz [1] crystals, as well as in $LiNbO_3$ [1,2], this technique was later extended to femtosecond laser pulses [3] which allowed generation of much broader bandwidths in terahertz region, [4] and even reaching mid-IR freqiencies of 40 to 50 THz [5,6].

Typically, to generate broadband THz transients via OR, thin (~1mm or less) electro-optic crystals are used, because of phase matching constraints as well as high absorption in conventional crystals ($LiNbO_3$, ZnTe) at THz frequencies. Optical-to-THz conversion efficiencies achieved so far by optical rectification methods are low [7], typically 10^{-6} - 10^{-9}, even with femtosecond pump pulse energies as high as 10 mJ [8]. However, recently conversion efficiency of $5x10^{-4}$ was reported. The authors used OR in $LiNbO_3$ with tilted pulse front excitation and energy per optical pulse 500 μJ [9].

In order to enhance the optical-to-THz conversion efficiency, larger interaction lengths with collinear interaction of THz and optical waves is desirable. The idea of using quasi-phase-matched (QPM) materials with periodically-inverted sign of second-order susceptibility $\chi^{(2)}$ for THz optical rectification was proposed by Lee and coauthors [10]. Multi-cycle narrow-band terahertz radiation was produced in periodically poled lithium niobate (PPLN) crystal. The authors used femtosecond pulses at 800 nm and cryogenically cooled (18K)

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PPLN crystal to reduce THz absorption, and achieved 10^{-5} conversion efficiency. OR in this case gives rise to a THz waveform which corresponds to the domain structure of the PPLN. Recently, efficient narrow-band terahertz radiation was demonstrated in another QPM material, orientation-patterned gallium arsenide (OP-GaAs) [11], which is extremely promising for THz generation because of intrinsically small THz absorption in GaAs.

THz wave generation in QPM crystals is illustrated in the time-domain animation of Fig. 1. Each inverted domain of a nonlinear crystal contributes a half-cycle of the THz pulse [10] and thus the THz wave packet (provided that THz-wave attenuation is low) has as many oscillation cycles as the number of quasi-phase-matched periods over the length of the crystal.

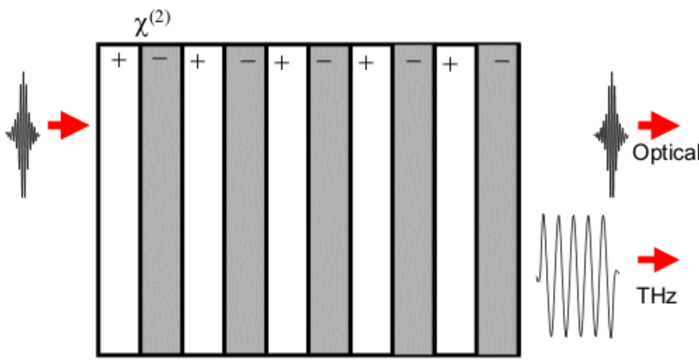

Fig. 1. Illustration of optical rectification in a media with periodically inverted $\chi^{(2)}$ sign, using femtosecond pump pulses. The static picture shows optical and THz electric fields, while the animation shows the nonlinear driving polarization and the THz electric field (779kB).

The purpose of this paper is to evaluate maximum optical-to-terahertz conversion efficiency which can be achieved by quasi-phase-matched down conversion process and to find optimal conditions in terms of pump wavelength, pump pulse format, crystal length, and focusing.

2. Plane wave analysis, femtosecond pulses (optical rectification).

Consider as an optical pump, bandwidth-limited ultrashort (e.g., femtosecond) laser pulses propagating along z in the form of infinite plane waves, with the gaussian time envelope of the electric field

$$E(t) = \mathrm{Re}\{E_0 \exp(-t^2/\tau^2)\exp[i(\omega_0 t)]\} = \frac{1}{2}\{E_0 \exp(-t^2/\tau^2)\exp[i(\omega_0 t)] + c.c.\}, \quad (1)$$

where ω_0 is the central frequency and τ is the pulsewidth.

Intensity envelope is thus $I(t) \sim \exp(-2t^2/\tau^2)$ and the pulse duration at full width of half-maximum is $\tau_{FWHM}=(2\ln 2)^{1/2}\tau=1.18\tau$. Using Fourier transform pair in the form

$$f(t) = \int_{-\infty}^{\infty} f(\omega)\exp(i\omega t)\,d\omega \quad (2)$$

$$f(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} f(t)\exp(-i\omega t)dt\,, \tag{3}$$

and taking into account that E(t) is real, we can get (z=0) a transform of the electric field (1) in the form $\frac{1}{2}\{E(\omega)+E^*(-\omega)\}$, where E(ω) is a one-sided (ω > 0) Fourier component given by

$$E(\omega) = \frac{E_0\tau}{2\sqrt{\pi}}\exp(-\frac{\tau^2(\omega-\omega_0)^2}{4}) \tag{4}$$

Similar form of one-sided Fourier representation will be used for other fields below in the text. For arbitrary z,

$$E(\omega,z) = E(\omega)\exp[-ik(\omega)z]\,, \tag{5}$$

where k(ω) is a module of the wave vector.

The one-dimensional equation (scalar form) for the Fourier component of the THz field at the angular frequency Ω, E(Ω,z), follows directly from Maxwell's equations and is given, in the slowly varying envelope approximation and in the limit of no absorption, by [12]:

$$\frac{dE(\Omega,z)}{dz} = -\frac{i\mu_0\Omega c}{2n_1}P_{NL}(\Omega)\exp(i\Delta kz)\,, \tag{6}$$

where the Fourier component of nonlinear polarization, $P_{NL}(\Omega)$, can be expressed through the material nonlinear susceptibility $\chi^{(2)}$ as

$$P_{NL}(\Omega) = \varepsilon_0\chi^{(2)}\int_0^{\infty} E(\omega+\Omega)E^*(\omega)d\omega \tag{7}$$

Here ε_0 and μ_0 are respectively the permittivity and permeability of free space, c is the speed of light in vacuum, n_1 is the THz refractive index. From Eqs. (4) and (7) it follows that

$$P_{NL}(\Omega) = \varepsilon_0\chi^{(2)}\frac{E_0^2\tau}{2\sqrt{2\pi}}\exp(-\frac{\tau^2\Omega^2}{8}) \tag{8}$$

and (6) becomes

$$\frac{dE(\Omega,z)}{dz} = -i\frac{\Omega\chi^{(2)}E_0^2\tau}{4\sqrt{2\pi}cn_1}\exp(-\frac{\tau^2\Omega^2}{8})\exp(i\Delta kz)\,. \tag{9}$$

The k- vector mismatch Δk is given by the following relation

$$\Delta k = k(\Omega)+k(\omega)-k(\omega+\Omega)-\frac{2\pi}{\Lambda}\,. \tag{10}$$

Since Ω<< ω, we can replace $k(\omega+\Omega)-k(\omega)$ by $(\partial k/\partial\omega)_{opt}\Omega$ [1,13] and obtain Δk in the form

$$\Delta k = \frac{\Omega n_{THz}}{c}-(\frac{dk}{d\omega})_{opt}\Omega-\frac{2\pi}{\Lambda} = \frac{\Omega}{c}(n_{THz}-n_{opt}^{gr})-\frac{2\pi}{\Lambda}\,, \tag{11}$$

where n_{THz} is the phase refractive index for the THz wave, n_{opt}^{gr} is the optical group velocity refractive index, and Λ is the QPM orientation-reversal period. We assumed here that the optical group velocity dispersion $\frac{\partial^2 k}{\partial\omega^2}$ is negligible (which will be justified later). With the undepleted pump approximation, Eq. (9) can be integrated to get the power spectrum of the THz field

$$\left|E(\Omega,L)\right|^2 = \frac{\Omega^2 d_{eff}^2 E_0^4 \tau^2}{8\pi c^2 n_1^2} L^2 \exp(-\frac{\tau^2\Omega^2}{4})\mathrm{sinc}^2(\Delta k L/2) \cdot \tag{12}$$

Here we assumed $\chi^{(2)}=2d_{\mathrm{eff}}$, where $d_{\mathrm{eff}}=(2/\pi)d_{\mathrm{OR}}$ is an effective QPM nonlinear coefficient; d_{OR} corresponds to the optical rectification process $d_{\mathrm{OR}}(0=\omega-\omega)$ and is derived from electro-optic coefficient r_{ijk} using the relation [12] $d_{\mathrm{jkl}}= - n^4/4r_{\mathrm{jlk}}$, where n is the optical refractive index. In GaAs, for example, r_{14}=1.5pm/V [14], corresponding to the nonlinear coefficient d_{OR} = 47 pm/V.

If the nonlinear crystal is long enough, terahertz radiation will be emitted in the form of narrow-band spectrum centered at Ω_0, corresponding to Δk=0 condition

$$\Omega_0 = \frac{2\pi c}{\Lambda \Delta n},\ \lambda_{THz} = \Lambda\Delta n\ , \tag{13}$$

where $\Delta n = n_{THz} - n_{opt}^{gr}$ is the index mismatch. By differentiating (11) we get $\frac{d(\Delta k)}{d\Omega} = \frac{\Delta n}{c}$ and the phase-matching acceptance bandwidth based on the condition $\Delta kL/2 = \pi$

$$\Delta\Omega^{accept} = \frac{c}{\Delta n}\Delta k^{accept} = \frac{2\pi c}{L\Delta n} \tag{14}$$

We assumed that n_{THz} is nearly constant, which is true for frequencies well below the lowest phonon resonance. For GaAs, this resonance is at 8.1 THz [15], n_{THz} ≈3.6 [16], n_{opt}^{gr} =3.41 (for the 2.1-μm optical pump) [17], Δn=0.19, and acceptance bandwidth for L=1cm crystal is $\Delta\nu_{THz}^{accept}$ =5.3 cm^{-1}. In the absence of quasi phase-matching, interaction between the optical and THz waves is limited to the coherence length

$$l_c = \pi c/\Omega\Delta n\ , \tag{15}$$

corresponding to the $\Delta k l_c = \pi$ condition.

It is worth mentioning that *backward emission* of THz wave is also possible. In this case, the phase-matching condition becomes

$$\frac{\Omega}{c}(n_{THz} + n_{opt}^{gr}) - \frac{2\pi}{\Lambda} = 0 \tag{16}$$

Optical-to-THz energy fluence efficiency for plane waves (PW) is

$$\eta_{THz}^{PW} = \frac{Fluence(THz)}{Fluence(pump)} \cdot \tag{17}$$

The pump fluence is given by

$$F_{pu} = \frac{c\varepsilon_0 n_2}{2}\int_{-\infty}^{\infty}\left|E(t,0)\right|^2 dt = \sqrt{\frac{\pi}{2}}\frac{c\varepsilon_0 n_2}{2}E_0^2\tau\ , \tag{18}$$

where n_2 is the optical refractive index. The THz fluence is

$$F_{THz} = \frac{c\varepsilon_0 n_1}{2}\int_{-\infty}^{\infty}\left|E_{THz}(t,L)\right|^2 dt = \frac{c\varepsilon_0 n_1}{2}2\pi\int_0^{\infty}\left|E(\Omega,L)\right|^2 d\Omega\ , \tag{19}$$

where we used Parseval's theorem: $\int_{-\infty}^{\infty}\left|f(t)\right|^2 dt = 2\pi\int_{-\infty}^{\infty}\left|f(\Omega)\right|^2 d\Omega \cdot$

From Eqs. (12) and (18-19) we get

$$\eta_{THz}^{PW} = \frac{\Omega^2 d_{eff}^2 E_0^2 \tau}{2\sqrt{2\pi} c^2 n_1 n_2} L^2 \int_0^\infty \exp(-\frac{\tau^2 \Omega^2}{4}) \mathrm{sinc}^2 [\frac{\Delta n(\Omega - \Omega_0) L}{2c}] d\Omega, \tag{20}$$

where we replaced $\Delta kL/2$ with $\Delta n(\Omega - \Omega_0)L/2c$. For $L >> l_c$, which is equivalent to the condition $\Delta\Omega^{accept} << \Omega_0$, sinc^2 function under the integral dominates and we obtain

$$\eta_{THz}^{PW} = g_1 \frac{2\Omega_0^3 d_{eff}^2}{\pi \varepsilon_0 c^3 n_1 n_2^2} L l_c F_{pu} = g_1 \frac{2\Omega_0^2 d_{eff}^2 L}{\varepsilon_0 c^2 n_1 n_2^2 \Delta n} F_{pu} \tag{21}$$

where Ω_0 is given by Eq. (13). The reduction factor

$$g_1 = \exp(-(\tau\Omega_0/2)^2) = \exp(-(\pi \nu_{THz} \tau)^2) \tag{22}$$

reflects the fact that the optical pulse should be short enough, so that its spectrum span is larger than the THz frequency Ω_0. For very short optical pulses, $\tau\Omega_0 < 1$, $g_1 \approx 1$ and optical-to-THz conversion efficiency depends *on pulse fluence only*, not intensity. Figure 2 shows the reduction factor g_1 as a function of the product $\nu_{THz}\tau$. For example at $\nu_{THz}\tau = 0.1$, $g_1 = 0.91$, close to unity, and experiences little change as the pulse duration is further decreased.

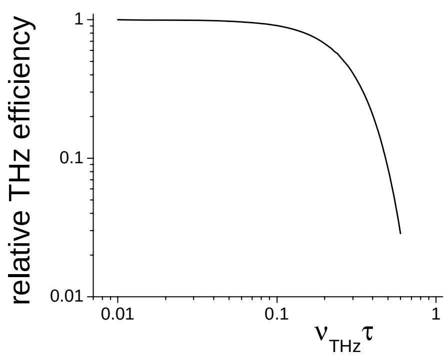

Fig. 2. Reduction factor g_1 as a function of $\nu_{THz}\tau$ for the case of femtosecond pump pulses ($\nu_{THz} = \Omega/2\pi$).

3. Plane wave analysis, picosecond pulses

Let us now regard as a pump, bandwidth-limited pulses with longer (pico- or nanosecond) duration τ, such that $\nu_{THz}\tau > 1$. In this case, the spectrum of a single pulse is narrow and to generate THz output, two different pump pulses need to be mixed to achieve difference frequency generation (DFG). Assume that two gaussian bandwidth-limited optical pulses (plane waves) at frequencies ω_2 and ω_3 with equal pulse widths propagate collinearly and generate THz wave centered at $\Omega_0 = \omega_3 - \omega_2$. Assume that the electric fields (i=2,3) are in the form

$$E_i(t) = \mathrm{Re}\{E_i \exp(-t^2/\tau^2) \exp[i(\omega_i t)]\}, \tag{23}$$

then, similar to Eq. (4), Fourier transform is given ($\omega > 0$) by

$$E_i(\omega) = \frac{E_i \tau}{2\sqrt{\pi}} \exp(-\frac{\tau^2(\omega - \omega_i)^2}{4}). \tag{24}$$

The Fourier component of nonlinear polarization at THz frequency Ω is

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$$P_{NL}(\Omega)=\varepsilon_0\chi^{(2)}\int_0^\infty E_3(\omega+\Omega)E_2^*(\omega)d\omega=\varepsilon_0\chi^{(2)}\frac{E_2E_3\tau}{2\sqrt{2\pi}}\exp(-\frac{\tau^2(\Omega-\Omega_0)^2}{8}). \quad (25)$$

Integrating Eq. (6) in the limit of no absorption and no pump depletion, we get the power spectrum of the THz field

$$\left|E(\Omega,L)\right|^2=\frac{\Omega^2 d_{eff}^2E_2^2E_3^2\tau^2}{8\pi c^2n_1^2}L^2\exp(-\frac{\tau^2(\Omega-\Omega_0)^2}{4})\mathrm{sinc}^2(\Delta kL/2), \quad (26)$$

where Δk is given by (11).

Suppose that the QPM orientation-reversal period Λ is such that the sinc^2 peak is centered exactly at $\Omega_0=\omega_3-\omega_2$. In this case $\Delta kL/2$ can be replaced by $(\Omega-\Omega_0)\Delta nL/2c$. Using Parseval's theorem, we find the optical-to-THz energy fluence conversion efficiency with respect to one of the two pump pulses (at ω_2)

$$\eta_{THz}^{PW}=\frac{Fluence(THz)}{Fluence(\omega_2)}=\frac{\Omega^2d_{eff}^2E_3^2\tau}{2\sqrt{2\pi}c^2n_1n_2}L^2\int_0^\infty\exp(-\frac{\tau^2\Omega'^2}{4})\mathrm{sinc}^2[\frac{\Delta n\Omega' L}{2c}]d\Omega' \quad (27)$$

where $\Omega'=\Omega-\Omega_0$

The temporal walk-off length between the optical and THz pulses can be introduced as

$$l_w=\sqrt{\pi}c\tau/\Delta n, \quad (28)$$

which is proportional to the length in a crystal at which the optical pulse walks away in time, with respect to THz wave, by its width τ, due to difference in propagation velocities. It is analogous to the Boyd-Kleinmann's [18] birefringent aperture length in the theory of second harmonic generation $l_a=\sqrt{\pi}w_0/\rho$ (w_0 is beam radius, and ρ is the birefringent walk-off angle). Now (27) can be expressed as

$$\eta_{THz}^{PW}=\frac{\Omega^2d_{eff}^2E_3^2\tau}{2\sqrt{2\pi}c^2n_1n_2}L^2\int_0^\infty\exp(-\frac{\tau^2\Omega'^2}{4})\mathrm{sinc}^2[\frac{\sqrt{\pi}}{2}(\frac{L}{l_w})\tau\Omega']d\Omega' \quad (29)$$

In the limit of long pulses, $l_w>>L$, exponential function under the integral dominates and (29) becomes

$$\eta_{THz}=\frac{\Omega^2d_{eff}^2E_3^2L^2}{\sqrt{2}c^2n_1n_2}=\frac{2\Omega^2d_{eff}^2L^2}{\varepsilon_0c^3n_1n_2n_3}\frac{I_3}{\sqrt{2}}, \quad (30)$$

where n_2 and n_3 are refractive indices at ω_2 and ω_3 and I_3 is the peak pump intensity. This formula is similar to the well-know expression [19] for the CW difference frequency generation, with $I_3/\sqrt{2}$ playing the role of time-averaged pump intensity.

In the limit of short pulses, $l_w<<L$, (29) becomes

$$\eta_{THz}=\frac{\Omega^2d_{eff}^2E_3^2Ll_w}{\sqrt{2}c^2n_1n_2}=\frac{2\Omega^2d_{eff}^2Ll_w}{\varepsilon_0c^3n_1n_2n_3}\frac{I_3}{\sqrt{2}}, \quad (31)$$

Thus, for $l_w<<L$, the L^2 term is replaced by Ll_w, in full analogy with the case of second harmonic generation with the spatial walk-off [18], where L^2 is replaced by Ll_a for $l_a<<L$. Also, conversion efficiency can be rewritten in terms of coherence length $l_c=\pi c/\Omega\Delta n$ and energy fluence F_3

$$\eta_{THz}=\frac{2\Omega^3d_{eff}^2Ll_c}{\pi\varepsilon_0c^3n_1n_2n_3}F_3, \quad (32)$$

This shows that at $l_w<<L$, terahertz conversion efficiency is a function of fluence only. Besides, it is equal to the conversion efficiency for the case of a femtosecond pulses with $\nu_{THz}\tau<<1$. The only difference is that in a two-color picosecond case, in order to get the same energy per THz pulse, one needs to have twice total energy (U_0 in each of the beams), as compared to U_0 in the femtosecond case.

In the case of intermediate pulse durations, we can use (32) with a reduction factor $g_2(l_w/L)$,

$$\eta_{THz} = g_2 \frac{2\Omega^3 d_{eff}^2 L l_c}{\pi \varepsilon_0 c^3 n_1 n_2 n_3} F_3 = g_2 \frac{2\Omega^2 d_{eff}^2 L}{\varepsilon_0 c^2 n_1 n_2 n_3 \Delta n} F_3 \ , \qquad (33)$$

where

$$g_2(x) = \frac{1}{\pi} \int_{-\infty}^{\infty} \exp(-x^2 \mu^2 / \pi) \mathrm{sinc}^2(\mu) d\mu \cdot \qquad (34)$$

Figure 3 is the plot of the reduction factor g_2 as a function of l_w/L. In many cases, it is desirable to have longer pulses to suppress high-order nonlinear optical effects, even at the expense of some loss in efficiency. Thus setting l_w/L=1 (g_2=0.69) might be a good compromise between efficiency and pump intensity. For an L = 1cm GaAs and pump at 2.1 µm, the l_w/L =1 condition corresponds to the pulse duration of 3.6 ps. At longer pulses, the THz efficiency will decline; however it will not be improved dramatically if the pulses are made shorter.

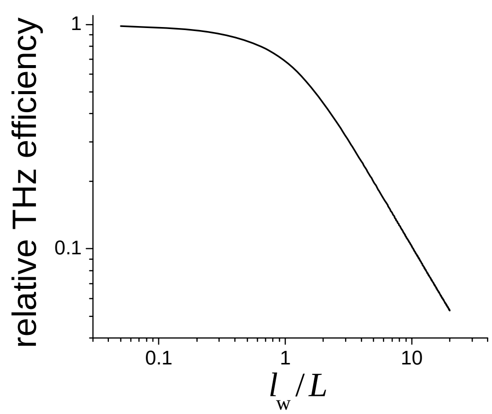

Fig. 3. Reduction factor g_2 as a function of l_w/L for the case of picosecond pump pulses.

4. Chirped stretched femtosecond pulses

Narrow-band THz radiation can also be generated by mixing two linearly chirped optical pulses (Fig. 4). Overlap of two relatively delayed optical pulses produces a constant beat frequency, which is proportional to the time delay and the amount of frequency chirp. Weling *et al*. [20] demonstrated this with photoconducting antennas and 23-ps-long pulses and showed that the generated THz frequency exhibited a linear dependence on the time delay. The same technique can be applied to THz generation using optical rectification.

Assume that the two pulses are of equal intensities. The electric field of the first optical pulse is in the form

$$E_2(t) = \mathrm{Re}\{E_0 \exp(-t^2/\tau^2) \exp[i(\omega_0 t + bt^2)]\} \qquad (35)$$

and that of the second (delayed) pulse is

$$E_3(t) = E_2(t+\Delta t) = \mathrm{Re}\{E_0 \exp(-(t+\Delta t)^2/\tau^2) \exp[i(\omega_0(t+\Delta t) + b(t+\Delta t)^2)]\} \quad (36)$$

Then the nonlinear polarization at the THz beat frequency is

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$$P_{NL}(t) = \mathrm{Re}\{\varepsilon_0\chi^{(2)}(E_2(t)+E_3(t))^2\big|_{\omega\approx 0}\}$$
$$= \mathrm{Re}\{\varepsilon_0\chi^{(2)}E_0^2 \exp[-\frac{2}{\tau^2}(t^2+\Delta t t+\frac{\Delta t^2}{2})+i(2b\Delta t t+b\Delta t^2)]\} \quad (37)$$

Its Fourier transform (Ω>0)

$$P_{NL}(\Omega) = \frac{\varepsilon_0\chi^{(2)}E_0^2}{2\pi}\int_{-\infty}^{\infty}\exp[-\frac{2}{\tau^2}(t^2+\Delta t t+\frac{\Delta t^2}{2})+i(2b\Delta t t+b\Delta t^2-\Omega t)]dt$$
$$= \varepsilon_0\chi^{(2)}\frac{E_0^2\tau}{2\sqrt{2\pi}}\exp(-\frac{\tau^2\Omega'^2}{8}-\frac{1}{2}\frac{\Delta t^2}{\tau^2})\exp(i\varphi_1) \quad (38)$$

where $\Omega' = 2b\Delta t - \Omega$ and $\varphi_1 = -\Delta t\Omega'/2 + b\Delta t^2$.

Similar to Eq. (29), we get THz fluence conversion efficiency with respect to one of the two pump pulses

$$\eta_{THz}^{PW} = \frac{\Omega^2 d_{eff}^2 E_0^2\tau}{2\sqrt{2\pi}c^2 n_1 n_2}L^2\int_0^{\infty}\exp(-\frac{\tau^2\Omega'^2}{4}-\frac{\Delta t^2}{\tau^2})\mathrm{sinc}^2[\frac{\Delta n(\Omega'+\Omega'')L}{2c}]d\Omega' \quad (39)$$

Exponential function here peaks at Ω'=0, that is when $\Omega=2b\Delta t$. If we assume that the sinc^2 function peaks at the same frequency (Ω"=0), then Eq. (39) becomes

$$\eta_{THz} = g_2\exp(-\frac{\Delta t^2}{\tau^2})\frac{2\Omega^3 d_{eff}^2 L l_c}{\pi\varepsilon_0 c^3 n_1 n_2 n_3}F_3\ , \quad (40)$$

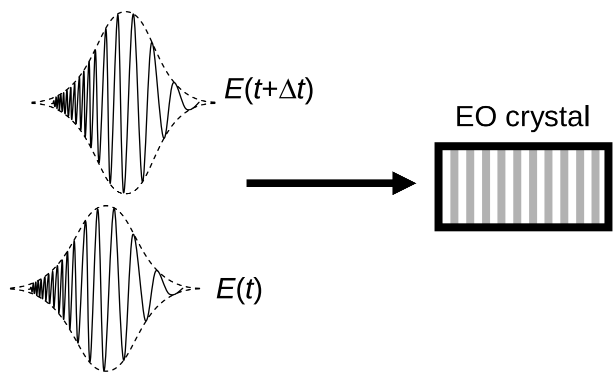

Fig. 4. Scheme to generate tunable THZ radiation from the overlap of two linearly chirped pulses.

Here g_2 is the reduction factor given by Eq. (34); the exponential term can be regarded as another reduction factor, associated with the temporal overlap of the two pulses, one of which is delayed by Δt. Suppose that the two chirped pulses with the pulsewidth $\tau=\tau_2$ are created by stretching in time a much shorter (femtosecond) pulse with the pulsewidth τ_1. It is easy to show [21] that in this case $b=1/\tau_1\tau_2$ and $\exp(-(\Delta t/\tau_2)^2) = \exp(-(\Omega\tau_1/2)^2)$. Interestingly, this reduction factor is the same as $g_1(\tau_1)$ in (22); thus (40) becomes

$$\eta_{THz} = g_1 g_2\frac{2\Omega^3 d_{eff}^2 L l_c}{\pi\varepsilon_0 c^3 n_1 n_2 n_3}F_3\ . \quad (41)$$

We see that in the case of linearly chirped optical pulses, THz conversion efficiency is, again, a function of fluence only, provided that pulse durations τ_1 and τ_2 are short enough.

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5. Optimal length of the EO crystal

From the simple plane-wave analysis above, we see that the optical-to-THz conversion efficiency, in the optimized case, is proportional to L. If we take into account THz absorption in the crystal (which is usually much larger than the optical absorption) but still neglect pump depletion, we get

$$\eta_{THz}(L) \propto \frac{1}{\alpha}[1-\exp(-\alpha L)] = L_{eff} \quad , \tag{42}$$

where α is the THz intensity absorption coefficient and L_{eff} is an effective length. When $L\rightarrow\infty$, $L_{\mathrm{eff}}\rightarrow 1/\alpha$. Choosing $L=1/\alpha$ will give us $L_{\mathrm{eff}}=0.63/\alpha$. In general, one can introduce another reduction factor, associated with the absorption and write

$$\eta_{THz}(L) \propto g_3 L\,; \quad g_3 = \frac{1}{\alpha L}[1-\exp(-\alpha L)] \tag{43}$$

For $L=1/\alpha$, $g_3=0.63$.

6. Optimal focusing

To maximize the THz efficiency, one needs to focus pump beams. In the near field approximation, when the focusing is loose and diffraction can be neglected, the optical-to-THz conversion efficiency (OR, femtosecond pulses) with respect to the pump pulse energy U_{pu} can be obtained by integrating Eq. (21) over the transverse coordinate r ($E_{pu}(r) \sim \exp(-r^2/w^2)$)

$$\eta_{THz} = \frac{U_{THz}}{U_{pu}} = g_1 g_3 \frac{2\Omega^2 d_{eff}^2 L}{\varepsilon_0 c^2 n_1 n_2^2 \Delta n} \frac{U_{pu}}{\pi w^2} \quad , \tag{44}$$

where w is the gaussian pump beam size. Following Ch.5 of [18], which considers the DFG case, we can characterize the focusing strength by a focusing parameter $\xi = L/k_1 w^2 = \lambda_1 L/2\pi n_1 w^2$, where λ_1 is the THz wavelength and n_1 is the THz refractive index (Boyd-Kleinman's theory itself [18] is not applicable here since THz field is not a resonant field and its distribution is not defined a priori by an optical cavity). Morris and Shen [22] developed a theory of far-infrared generation by optical mixing of focused laser beams, based on Fourier analysis with respect to transverse k-vector components, and have found that focusing of the pump beams appreciably enhances the far-infrared output despite the strong far-infrared diffraction. For example in a 1-cm-long GaAs crystal and an output wavelength 100μm, the optimal focal-spot size (for the optimized phase-matching condition) was found to be around w=20 μm (ξ=110), that is less than the THz wavelength.

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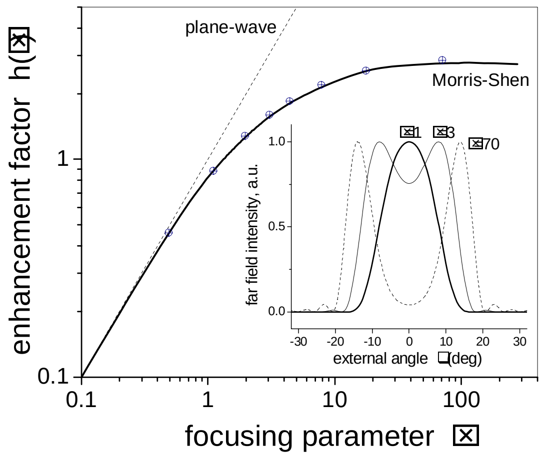

Fig. 5. Enhancement factor *h* as a function of the focusing parameter ξ. Solid curve is based on ref. 22. Dashed curve – plane-wave approximation. Dots represent our calculations based on the Green's function method. Inset: far field THz intensity profiles at different ξ for a 1-cm-long GaAs.

THz conversion efficiency for OR with femtosecond pulses and focused beams can be expressed in the form

$$\eta_{THz} = \frac{U_{THz}}{U_{pu}} = g_1 g_3 \frac{2\Omega^3 d_{eff}^2}{\pi\varepsilon_0 c^3 n_2^2 \Delta n} U_{pu} h(\xi)\,, \tag{45}$$

and, similarly, for DFG with picosecond pulses, by

$$\eta_{THz} = \frac{U_{THz}}{U_2} = g_2 g_3 \frac{2\Omega^3 d_{eff}^2}{\pi\varepsilon_0 c^3 n_2 n_3 \Delta n} U_3 h(\xi)\,. \tag{46}$$

Here $h(\xi)$, by analogy with [18], is an enhancement factor due to pump focusing, such that $h(\xi)=\xi$ at $\xi<<1$ (plane waves). Enhancement factor $h(\xi)$ obtained from the calculations of Morris and Shen [22] is shown on Fig. 5, along with the enhancement factor for plane-waves. Fig.5 (dots) also shows results of our calculations of the enhancement factor, based on the Green's function method [23,24], where the far field solution was obtained by integration over the nonlinear polarization distribution in real space; these results are in excellent agreement with the Morris-Shen curve.

There are however limitations on the very tight focusing. (i) When pump beam waist w is too small, THz output extends over a large span of angles θ to the normal of the crystal; when θ exceeds the total internal reflection angle θ_{max} of the material, THz transmission falls to zero. This leads to the condition $\sqrt{2}\lambda / \pi n_1 w < 2\theta_{max}$. For GaAs, for example, θ_{max} = 16°, and for 1.5 THz frequency, this corresponds to w > 45 µm. (ii) In the case of tight focusing, different Fourier components of the transverse *k*- vector have different phase-matching conditions and it appears [22] that the THz output is maximized when the far field distribution is in the form of a hollow cone. The phase-matching condition for each angle θ is

$$\Delta k_z = k_{1z} - (k_3 - k_2) = k_1 \cos\theta - (k_3 - k_2) \approx \Delta k_0 - \frac{k_1}{2}\theta^2 = 0\ , \tag{47}$$

where Δk_0 is a collinear wave-vector mismatch. For the high beam quality (solid vs hollow cone) we can require that variation of Δk_z due to variation of θ is small, so that $\delta(\Delta k_z)L<\pi$

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(corresponding to half of the QPM acceptance bandwidth). This leads to the condition $k_1\theta_d^2 L/2 \le \pi$ where θ_d is given by the diffraction: $\theta_d = \sqrt{2}\lambda_1 / 2\pi m w$. Thus we get

$$\xi = \frac{\lambda_1 L}{2\pi n_1 w^2} < \pi \,. \tag{48}$$

The far field intensity profiles of THz radiation at different ξ (GaAs, L=1cm) based on our calculations using the Green's function method is shown in the inset to Fig. 5 and is consistent with Eq. (48); ξ≈1 can be regarded as a good compromise between conversion efficiency and the beam quality.

From Eqs. (45-46) one can see that the THz conversion efficiency does not depend on the crystal length L. The length affects however the spectral width of the THz pulse; also, it is more advantageous to have longer crystals (on the order of 1/α), since in this case the beam waist will be larger and the peak intensity smaller. This is important from the viewpoint of reducing unwanted higher order nonlinear effects (see below).

As a numerical example, regard a 100-fs-long optical pump pulse at 2.1 μm, 1.5 THz output frequency, GaAs crystal length L=1/α =9.5 mm (α=1.05 cm^{-1}), and pump beam waist w=290 μm (ξ=1). In this case reduction factors are g_1=0.8, g_3=0.63, h(ξ)=0.82 and from (45) we obtain η_{THz}= 3.95×10^{-4} / μJ. For the 2.5 THz output (α=4.3 cm^{-1}), for L =2.3 mm and w=110 μm (ξ=1), g_1=0.54, g_3=0.63, h(ξ)=0.82, and η_{THz}=1.23×10^{-3} / μJ.

7. Cascading and red shift

THz wave generation via OR is a parametric process of self-mixing in which a photon from the blue (high-frequency) wing of the femtosecond optical pulse decays into a THz photon plus a red-shifted photon, corresponding to the low-frequency wing of the same optical pulse. From the photon energy conservation argument, it follows (if we neglect losses) that the center of weight of the optical pulse spectrum will be red-shifted by $\Delta\omega / \omega_0 \sim \eta_{THz}$, where ω_0 is the central optical frequency. When optical-to-THz photon conversion efficiency approaches 100%, the red shift will be on the order of THz frequency Ω. Once the optical pulse becomes red-shifted, it can still contribute to THz generation – the same process of cascaded optical down-conversion continues to transfer optical energy to lower frequencies, as long as the phase mismatch Δk in Eq. (11) is small. Accordingly, cascaded down-conversion will be the most efficient when the pump wavelength is close to the point of zero group velocity dispersion. Quantitatively, the number of cascading cycles can be expressed as N= ½ (acceptance bandwidth) / (teraherz frequency). Here acceptance bandwidth is with respect to the pump frequency and can be found by differentiating (11): $d(\Delta k)/d\omega_{pump} = (\Omega / c) dn_{gr} / d\omega$,
from which we get

$$\Delta\omega_{pump}^{accept} = \frac{2\pi c}{L}\frac{\omega_{pump}}{\Omega}\left(\lambda \frac{dn_{gr}}{d\lambda}\right)^{-1} \tag{49}$$

From Fig. 6 we can see that the number of THz cascading cycles in GaAs at 2-3.5 μm pump can be > 10. At pump wavelength near 6.6 μm, where the group velocity dispersion reaches zero, N can be even higher. Thus, THz conversion efficiency can be significantly above the Manley-Rowe limit. The possibility of overcoming quantum-defect-related limitations on the efficiency of THz wave difference frequency generation, for the case of mixing two discrete near infrared frequencies, was suggested by Cronin-Golomb [25].

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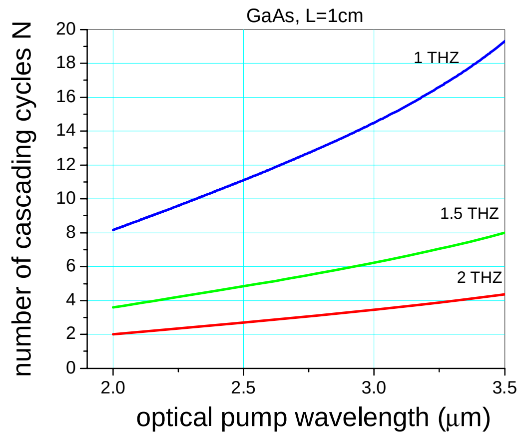

Fig. 6. Number of THz cascading cycles as a function of THz frequency and pump wavelength for GaAs, L=1cm.

The red shift of the optical pump pulse can also be understood from a different standpoint. Gustafson *et al*. [26] have pointed out that rectified field which is generated during the propagation of optical pulses through electro-optic crystals can produce considerable phase modulation of the optical pulse itself, especially when the rectified pulse travels at the same speed as the optical pulse within the crystal. This cascaded Kerr-like nonlinearity occurs because of back action of the optical rectified field upon the pump wave through the electro-optical effect [27,28].

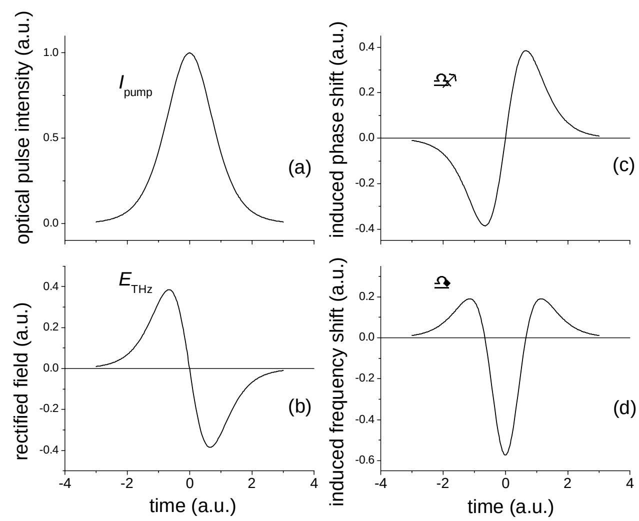

Fig. 7. (a) The optical pulse intensity profile, (b) rectified field profile, (c) phase profile across the optical pulse after traveling the crystal, and (d) frequency shift of the optical pulse.

Figure 7 illustrates the process of red shift for the case of single-cycle terahertz pulse generation. Through the EO effect, rectified field (Fig. 7(b)) produces a change in the refractive index of the medium $\delta n \sim r_{ij} E_{THz}(t,z)$ which causes the time-dependent phase shift $\delta\phi(t,z)$ (Fig. 7(c)). The amplitude of this phase shift is proportional to the integral of the rectified field over the crystal length; since the rectified field grows linearly with the length, the phase shift is proportional to the crystal length squared [26]. The frequency shift $\delta\omega=-\partial\delta\phi(t,z)/\partial t$ (Fig. 7(d)) is also proportional to the crystal length squared. The central portion of the optical pulse experiences red shift, independent on the sign of EO coefficient. This 'microscopic' consideration of the red shift gives the same dependence as before: it scales as THz conversion efficiency (proportional to the EO coefficient squared and crystal length squared) for the index-matched single-cycle terahertz pulse generation. In the case of multi-cycle THz generation with periodically inverted crystals, the optical pulse will see in average the same induced phase shift as shown in Fig. 7(b), since the relative phase between the optical and THz pulse is corrected at regular intervals via a structural periodicity of the nonlinear medium.

8. The role of higher order nonlinear effects

Two-photon absorption (2PA), three-photon absorption (3PA), and nonlinear refraction index (NRI), n_2, can severely limit the maximum THz conversion efficiency in the OR or DFG process: in the case of 2PA and 3PA, the pump beam will create free carriers which strongly absorb THz radiation; NRI, on the other hand, affects the pump pulse itself via self-phase modulation and self-focusing. As a necessary condition for achieving cascading effects in THz generation, one needs to be close to 100% in a single-stage optical-to-THz photon efficiency, which requires for GaAs pump intensities on the order of 100-1000 GW/cm^2 (fs pulses) and 1-100 GW/cm^2 (ps pulses). It is very advantageous to use longer pump wavelengths to avoid 2PA effects which are usually strong in semiconductors (e.g. for GaAs, β=26 cm/GW at 1.06 μm [29]). This dictates that pump wavelength should be above 1.74 μm (GaAs) in which case the dominant nonlinearities are 3PA and NRI (theoretically predicted values for GaAs are correspondingly $\gamma\approx0.2\ cm^3/GW^2$ [30] and $n_2\approx2\cdot10^{-4}\ cm^2/GW$ [31]). Also, it is more likely that the highest THz conversion efficiencies (> 100% by photons, with cascading) will be achieved in the picosecond, rather than femtosecond regime: since THz conversion efficiency is fluence-sensitive, peak intensity in the former case will be much smaller, and 3PA and NRI effects less pronounced.

9. Conclusion

We show that optically pumped periodically-inverted EO crystals have potential for generating narrow-bandwidth THz wave packets with high conversion efficiency. For sufficiently short (femtosecond) single-beam optical pulses with $\tau\Omega<1$ (optical rectification), THz conversion efficiency scales with the pulse energy and does not depend on pulse duration. In the case of mixing two picosecond pulses (bandwidth-limited or chirped), with pulse durations such that the temporal walk-off length is smaller than the length of the crystal. again, conversion efficiency is pulse-energy dependent, and has the same scaling factor as in the case of femtosecond pulses. Also, cascaded THz frequency down conversion is possible, allowing achieving > 100% optical-to-THz photon conversion efficiency.

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