Velocity matching by pulse front tilting for large-area THz-pulse generation

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Abstract: We propose a generally applicable velocity matching method for THz-pulse generation by optical rectification in the range below the phonon frequency of the nonlinear material. Velocity matching is based on pulse front tilting of the ultrashort excitation pulse and is able to produce a large-area THz beam. Tuning of the THz radiation by changing the tilt angle is experimentally demonstrated for a narrow line in the range between 0.8-0.97 times the phonon frequency. According to model calculations broad-band THz radiation can be generated at lower frequencies. Advantages of the new velocity matching technique in comparison to the electro-optic Cherenkov effect and non-collinear beam mixing are discussed.

OCIS codes: (190.2620) Frequency conversion; (190.7110) Ultrafast nonlinear optics

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1. Introduction

The possibilities of spectroscopy in the THz frequency range have been dramatically increased by novel generation and detection techniques for THz waves using ultrashort light pulses [1-3]. These techniques allow full characterization of the complex dielectric constant of many materials through time-domain spectroscopy [4-5]. In particular, dynamic effects can be time-resolved down to the subpicosecond time scale [6]. Coherent manipulation of quantum bits [7] and imaging [8] by THz pulses are further interesting applications with promising perspectives. The most widely used method for the generation and time-resolved detection of ultrashort THz-wave pulses utilizes [2,3] optical rectification and electro-optic sampling, respectively. The efficiency of both effects critically depends on matching between the group velocity of the ultrashort light pulse and the phase velocity of the THz radiation [2]. Velocity matching is obtained if the condition

$$v_{vis}^{gr} = v_{THz}^{ph} \tag{1}$$

is fulfilled. Recently velocity-matched THz generation in the broad range from 10 - 40 THz has been achieved [9], employing angle tuning in a birefringent semiconductor (GaSe). It should be noticed, however, that angle tuning implies a reduced effective nonlinear coefficient. Furthermore, velocity matching by birefringence is impossible below but close to the phonon frequency even in GaSe, which has an exceptionally large birefringence. This is due to the strong increase of the dielectric constant originating from phonon-polariton formation [10,11]. This fundamental problem exists for every material, and makes velocity matching impossible on the whole range below the phonon frequency in such important wide-bandgap dielectric materials as $LiNbO_3$ or $LiTaO_3$. For these materials, the refractive index in the far infrared is more than two times larger than in the visible. Instead of birefringence, Stevens et al. [12] have utilized strong dispersion in the vicinity of an electronic transition in ZnSe to obtain velocity matching between the visible excitation pulse and the generated THz radiation. The applicability of this technique is limited, it needs special pump wavelength determined by the THz frequency and the dispersion of the nonlinear crystal in the visible. The electro-optic Cherenkov effect [1,13] can produce THz radiation only if the lateral extension (beam-waist) *w* of the light pulse is considerably smaller than its axial spread:

$$w << v_{vis}^{gr} \cdot \tau , \tag{2}$$

where τ is the pulse duration, which has to be significantly shorter than the oscillation period of the THz field. The generated THz radiation propagates along a cone (see Fig. 1a) with an angle Θ_C determined by

$$\Theta_C = \cos^{-1}(\frac{v_{THz}^{ph}}{v_{vis}^{gr}}) . \tag{3}$$

Such a propagation characteristic makes it very difficult to collect the THz radiation for applications. In particular the energy of the THz emission can not be increased by increasing the cross section of the exciting laser beam since the radiated energy strongly drops when the lateral extension approaches the axial spread of the pulse [1,12,13].

Here, we suggest a very general method to reach velocity matching between visible light and THz radiation in a dielectric crystal below the phonon frequency. In comparison to the method of Stevens et al., this novel technique is distinguished by the fact that velocity matching can be achieved in wide-bandgap dielectric materials such as $LiNbO_3$ or $LiTaO_3$ and tunability of the THz frequency is attainable with a fixed-frequency fs laser. Contrary to the usual Cherenkov configuration, the excitation geometry proposed in this paper generates a collimated THz beam (see Fig. 1b) and provides high conversion efficiency for an extended ultrashort pump light beam. Therefore this technique enables scaling of the THz power by simply increasing the cross section and the power of the pump beam.

2. Principle of velocity matching by pulse-front tilting

The crucial feature of the novel scheme is the tilt of the pulse front of the light beam with respect to the phase front [14,15], which is perpendicular to the propagation direction of the light. This tilted pulse front is the key for velocity matching in our set up. The THz radiation (in the material the phonon-polariton) excited impulsively along this tilted pulse front will propagate according to Huygens' principle [16] perpendicularly to this front with a velocity v_{THz}^{ph}. Although the pump pulse moves with a velocity v_{vis}^{gr}, the projection of this velocity in the propagation direction of the generated THz radiation is only $v_{vis}^{gr} \cdot \cos(\gamma)$. Obviously this projected velocity has to be equal with the velocity of the THz radiation. Therefore instead of Eq. 1, the velocity matching condition reads

$$v_{vis}^{gr} \cdot \cos\gamma = v_{THz}^{ph} . \tag{4}$$

According to Eq. 4, velocity matching is possible in a material with a significantly larger dielectric constant in the far infrared than in the visible by proper choice of the tilt angle γ.

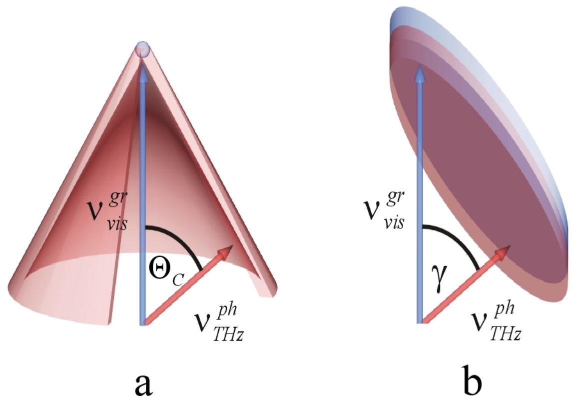

Fig.1. Two different schemes of THz excitation. For the usual Cherenkov geometry (a), the THz radiation is emitted as a cone characterized by the angle Θ_C. Velocity matching (see Eq. 4) is satisfied, but the exciting beam has to be very narrow (see Eq. 3). Velocity matching by pulse front tilting (b) creates a plane THz wave without any upper limit for the exciting beam cross-section.

Having a glance at Fig.1a and Eq.3 it is obvious that Eq. 4 is satisfied in the case of Cherenkov radiation created by a very tightly focused light pulse, too. However, THz Cherenkov radiation generated at a given time cannot interact with the radiation generated later, while for an extended beam with tilted pulse front THz photons created at different times interact with each other in the nonlinear medium resulting in enhanced conversion efficiency. A further advantage of the tilted pulse front is the generation of a plane THz wave. Its manipulation is much easier than for the cone-shaped Cherenkov emission.

3. Proof of principle experiment

In our proof of principle experiment, the exciting light pulses were obtained from a Ti:sapphire laser delivering 25 fs pulses at 810 nm with 350 mW average power at a repetition rate of 76 MHz. The phonon-polaritons generated by the pump pulse were monitored by a probe beam with variable delay. The beams were diffracted off a 1200 line/mm reflection grating in order to create the tilt of the pulse front. An f= 150 mm cylindrical lens mounted in front of the grating focused both beams to a horizontal line on its surface which was imaged into the nonlinear material (GaP) by an f = 50 mm lens. The imaging was set to create a demagnification of about 3, increasing the tilt angle by the same factor. Phonon-polaritons were excited inside a 130-μm-thick GaP crystal with a [111] surface. The transmitted probe

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beam was spectrally dispersed in a monochromator and then detected by a photodiode. The photodiode signal was fed into a computer and averaged over several thousand delay scans.

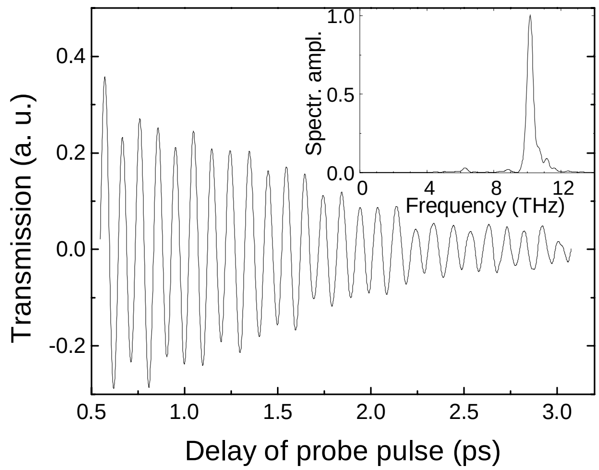

Fig. 2. Measured differential transmission versus probe delay for $\gamma = 39.3^o$ tilt angle. The inset shows the corresponding spectrum of the radiation. Its peak at 10.16 THz is in good agreement with the 10.12 THz calculated from Eq. 4. Changing γ, we were able to tune the spectrum from 9.0 -10.7 THz .

Figure 2 depicts the measured differential transmission of the probe beam for $\gamma = 39.3^o$. The clearly observable oscillation with a period of 98.4 fs is caused by the generated TO-phonon-polariton. The frequency of 10.16 THz agrees fairly well with the value of 10.12 THz calculated from Eq. 4 for $\gamma = 39.3^o$ using the well-known [17] dispersion formula of phonon-polaritons in GaP. The inset displays the Fourier transform of the transmission curve. It is dominated by the excited polariton with a spectral width of 0.44 THz. We were able to tune the frequency of the generated polariton from 9.0 - 10.7 THz by varying γ between 30 - 65^o. For this purpose, we changed the demagnification of the imaging of the grating surface into the GaP crystal by the lens. For $\gamma = 0^o$ (untilted pulse front of the pump beam), we observed only weak oscillations due to LO phonons.

4. Results of calculations

The bandwidth of the emitted THz radiation is usually relatively narrow, but its center frequency is easily tunable in the range of 0.6 - 0.95 times the TO phonon frequency. The high frequency limit of the tuning range is set by the very rapid increase of the polariton decay rate for frequencies close to the TO phonon frequency. At the low frequency edge, the polariton velocity becomes nearly independent of frequency and therefore the THz frequency cannot be tuned. Instead the spectrum becomes very broad as demonstrated in Fig. 3.

These plots display simulation results of polariton generation in GaP obtained by the simple model of Ref. 10. For velocity matching close to the TO phonon frequency of GaP at 9.0 THz (see Fig. 3a), a spectrally narrow distribution develops along the first few ten micrometers. The center frequency of this spectrum can be tuned by varying γ as predicted by the calculations and demonstrated by the experiments. Adjusting the velocity matching for frequencies lower than approximately 0.6 times the phonon frequency (see Fig. 3b), the generated THz-spectra become very broad.

Such broad THz pulses are well suited for time domain THz measurements. Tunable narrow band THz pulses are expected to become useful for spectrally and temporally resolved THz imaging.

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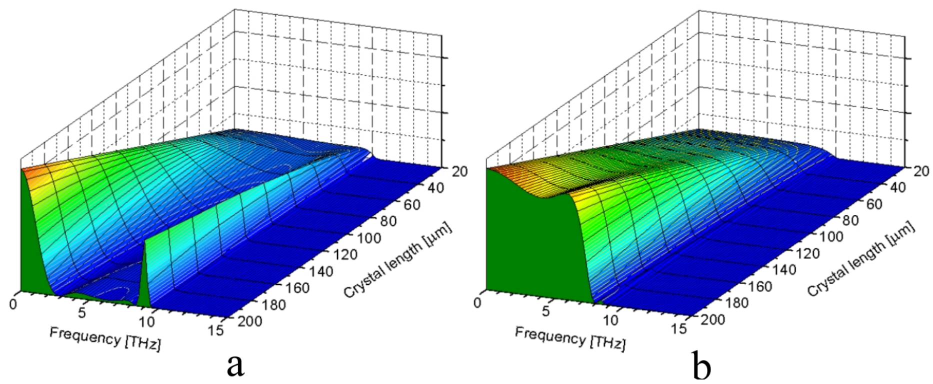

Fig. 3. Calculated dependence of the THz field amplitude spectrum on the GaP crystal length for velocities adjusted to generate a narrow spectrum at 9 THz (a), and 6 THz (b), respectively. During the calculations the excitation pulse duration and the polariton linewidth were supposed to be 30 fs and 1.1 cm^{-1}, respectively.

5. Discussion

THz generation by optical rectification using tilted pulse front excitation as proposed and demonstrated above has the advantage that the power is scalable. For higher pump power, the excited area can be increased in order to avoid damage of the crystal. This scaling possibility does not exist for the usual Cherenkov-like THz excitation, as pointed out in connection with Eq. 2.

It should be emphasized that this large-area velocity-matched interaction between a visible light pulse and a THz field offers similar advantages for time-resolved THz imaging using the electro-optic effect. It enables simultaneous imaging for a large number of parallel channels instead of the point-by-point image scanning employed so far. The high-frequency limit of THz wave generation in GaP using our technique is approximately 11 THz. Most likely, this limit can be as high as 22 THz if BN instead of GaP is employed as the nonlinear material. $LiNbO_3$, or $LiTaO_3$ on he other hand should provide a large increase of the THz conversion efficiency, since the nonlinear coefficient of these materials is almost 20 times larger than that of GaP. In a preliminary experiment, using a $LiNbO_3$ crystal and the pulses of a Ti:Sapphire laser amplified in a Coherent REGA 900 delivering 200 mW average power at 200 kHz repetition rate, we were able to detect 1.1 μW THz radiation using a He-cooled bolometer.

Finally it should be mentioned that the generation of polaritons by a pump beam with a tilted pulse front can be also well described in a framework different from the model used above which is based on velocity matching and Huygens' principle. The ultrashort pump pulse contains components covering a wide frequency range. Pulse front tilting of a light beam necessarily leads to angular dispersion [15]: components with different frequencies propagate in slightly different directions. Considering two frequency components $\omega_{vis+THz}$ and ω_{vis}, efficient generation of radiation with ω_{THz} by difference frequency mixing requires fulfillment of the well known phase matching condition:

$$\Delta\mathbf{k} = \mathbf{k}_{vis+THz} - \mathbf{k}_{vis} - \mathbf{k}_{THz} = 0\,, \tag{5}$$

where the **k**`s are the wave vectors of the corresponding frequency components. The wave vector diagram is depicted in Fig. 4a. The connection between the pulse front tilt γ and the angular dispersion $d\Theta/d\omega$ for a material with refraction index n is given by [15]:

$$\tan\gamma = -\frac{n}{n_{gr}} \cdot \omega \cdot \frac{d\Theta}{d\omega}\,, \text{ where } n_{gr} = n - \omega \cdot \frac{dn}{d\omega} = \frac{c}{v_{vis}^{gr}} \tag{6}$$

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Using Eq. 6, it is easy to show that for small values of $\Delta\Theta$ the inclination angle of $\mathbf{k}_{\mathrm{THz}}$ from the average direction of $\mathbf{k}_{\mathrm{vis}}$ and $\mathbf{k}_{\mathrm{vis+THz}}$ is the same γ as the pulse front tilt angle. Therefore the two pictures used in this paper to describe polariton excitation by ultrashort pulses with a tilted pulsefront, based on Huygens' principle or wave-vector conservation, respectively, predict the same propagation direction for the created THz beam. In our experiments, the full angular spread of the light beam $\Delta\Theta$ was about 2-3^{o}, while γ was 35-65^{o}.

For our pump pulses with tilted pulse front and angular dispersion, the average of the momentum points upwards in Figs. 1b and 4a, but the wave packet propagates in the same direction as the polariton generated by the tilted visible pulse (see Fig. 1b) according to the definition given in Ref. 18. This makes the matching of the velocities possible.

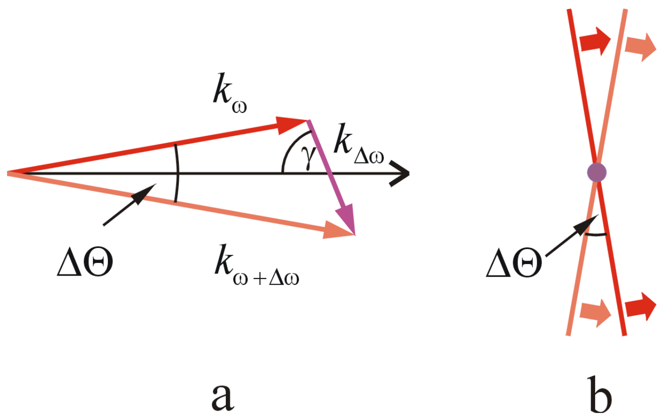

Fig. 4. Illustration of phase-matching (wave-vector conservation) for THz generation (a). Application of two ultrashort pulses without pulse front tilt for mixing achieves only partial overlap of the beam cross-sections, (b). For a beam with tilted pulse front (see Fig. 1b), the spectral components overlap across the whole cross-section of the beam, resulting in efficient THz generation.

Considering Fig. 4a, one could argue that application of a pump beam with a tilted pulse front for excitation of THz waves is equivalent to the use of two beams (without pulse front tilt) differing in their central frequencies by ω_{THz} and crossed at an angle $\Delta\Theta$. This presumption however is only correct for long pulses. Fig. 4b illustrates schematically the interaction of two ultrashort pulses crossing under an angle $\Delta\Theta = \frac{d\Theta}{d\omega}\cdot\omega_{THz}$. It is important to notice that spatial and temporal overlap is attainable only for a small part of the extended beam cross section. Thus the THz generation efficiency remains small. For tilted pulse front excitation in contrast, the components with different frequencies overlap across the whole diameter of the beam, implying efficient THz generation.

6. Conclusion

A new velocity matching technique based on tilting the front of the ultrashort excitation pulse has been demonstrated for THz pulse generation by optical rectification. Contrary to the electro-optic Cherenkov effect and noncollinear two-beam mixing, this method does not suffer from an upper limit for the cross-section of the excitation beam. According to proof of principle experiment, tunable ultrashort THz pulses can be generated in this way. It is expected that this new technique enables efficient generation of high energy THz pulses.

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