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## HIGH-FIELD STRENGTH THE SOURCES BASED ON TILTED PULSE FRONT PUMPING: OPERATION PRINCIPLE AND PERSPECTIVES

## ВИСОКОАМПЛІТУДНІ ТЕРАГЕРЦОВІ ДЖЕРЕЛА З ПОМПУВАННЯМ НАХИЛЕНИМ ФРОНТОМ: ПРИНЦИП РОБОТИ І ПЕРСПЕКТИВИ

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The terahertz wave generation using tilted pulse front pumping (TPFP) is the most advanced technique nowadays. In this paper we made a brief review of common terahertz sources. Theoretical aspects and experimental results of tilted pulse front pumping on  $\text{LiNbO}_3$  have been discussed.

**Key words:** TPFP,  $\text{LiNbO}_3$ , Terahertz wave, optical rectification.

Генерація електромагнітних хвиль терагерцового діапазону з використанням методики помпування нахиленим фронтом є найсучаснішим підходом на сьогодні. У статті зроблено огляд традиційних джерел терагерцового випромінювання. Також описано теоретичні аспекти й експериментальні результати генерації хвиль за допомогою накачки з нахиленим фронтом на кристалі  $\text{LiNbO}_3$ .

**Ключові слова:** помпування нахиленим фронтом,  $\text{LiNbO}_3$ , Терагерцові хвилі, оптичне випрямлення.

### Problem statement

The development of new terahertz sources is an extremely topical nowadays due to rapid development of new materials and nanostructures. Terahertz band of electromagnetic wave spectrum, which is typically governs frequency region from 0.3 THz up to 20 THz [1], has a lot of applications: material characterization, security systems, medical applications etc.

## Common terahertz sources

Generation of these waves is quite problematic but the modern devices greatly increased the range of frequencies which can be used [2]. One of the simplest ways, which was discovered in 1975 [3] by D. H. Auston of Bell Labs, is called “Auston switch”. The biased semiconductor surface is illuminated by femtosecond laser pulse and excites carriers in semiconductor. Thanks to these carriers, the picoseconds current is established. Since the current is time-dependent, the illuminated area classically radiates in the broad range of frequencies including terahertz. This is a classical photoconductive antenna [4]. This approach is also used for the detection of terahertz radiation and it’s called “Grischkowsky antenna” [5] and it is used in time-domain Fourier transform spectroscopy [6]. This concept of optically excited electron plasma is being extensively investigated and various approaches of wave generation have being introduced. This has been applied in optical semiconductor switches [7], semiconductor antennas [8] and plasma antennas [9]. In excited semiconductor antennas the main advantage is reconfigurability, which allows the continuous change of the resonance position of the antenna in broad interval of frequencies. Improvement of applications, which are using terahertz radiation, is mainly depends on development of continuous wave sources. For this purpose the microwave sources with harmonic generation chains are used, but their efficiency decreases with every stage of harmonic generation. Another way is use of backward wave oscillators, but they require high external magnetic field. The new method is quantum cascade laser [10, 11]. It is made of some number of quantum wells and requires quite sophisticated fabrication techniques. In such system, electrons are transferred between energy states of different wells emitting THz photons. One of the main disadvantages of this technique – extremely low temperatures is required.

In this paper we are going to describe more deeply another class of THz generators which is nonlinear optics devices.

### Terahertz generation using tilted pulse front pumping

In such devices, the main radiator is a nonlinear crystal (LiNbO<sub>3</sub>, ZnTe, LaTiO<sub>3</sub>, zinc-blende semiconductors or organic crystals), which is pumped by short-impulse laser [12]. This gives rise to nonlinear optical phenomena such as optical rectification, second harmonic generation and difference harmonic generation. These effects are mathematically described by expanding dielectric susceptibility  $\chi$  in powers of the electric field  $E$  as

$$P = c_1 E + c_2 E^2 + c_3 E^3 \dots \quad (1)$$

where  $P$  is the polarization. First term corresponds with linear response of matter and the following terms are responsible for nonlinear effects. In particular, second term  $c_2 E^2$  is responsible for difference frequency generation and optical rectification. This can be shown on example in which two electromagnetic waves  $E_1 = E_0 \cos(\omega_1 t)$  and  $E_2 = E_0 \cos(\omega_2 t)$  are interacting inside of the nonlinear crystal

$$P_2^{nl} = c_2 E_1 E_2 = c_2 \frac{E_0^2}{2} (\cos(\omega_1 t - \omega_2 t) + \cos(\omega_1 t + \omega_2 t)) \quad (2)$$

where  $P_2^{nl}$  is a second order polarization. As one can see, we have two terms: one is sum of frequencies and one is their difference. Obviously, for terahertz generation the difference frequency term relevant. In case of terahertz pulse generation by optical rectification, the frequency difference appears because of big bandwidth of pump optical pulse [13].

The important condition that needs to be fulfilled in order to obtain maximum efficiency of terahertz generation is phase matching between incoming pump pulse and generated terahertz radiation inside the crystal. The formula of this condition reads as

$$\Delta k(\Omega) = k(\Omega) + k(\omega) - k(\omega + \Omega) = 0 \quad (3)$$

where  $\Omega$  is THz wave frequency and  $\omega$  is optical frequency. This condition describes the conservation of energy and momentum. Because of  $\Omega \ll \omega$  we can rewrite phase matching condition as

$$\Delta k(\Omega) = [n(\Omega) - n_g(\omega_0)]\Omega / c = 0 \quad (4)$$

where  $\omega_0$  is averaged pump frequency,  $c$  – speed of light,  $n$  and  $n_g$  is refractive indices of matter under influence of terahertz wave and pump wave, respectively.  $n_g$  is often called group index. Phase matching

condition can also be written as equality of THz phase velocity  $u(\Omega)$  and group velocity  $u_g(w_0)$  of optical excitation pulse:

$$u(\Omega) = u_g(w_0) \quad (5)$$

When velocity is matched and dispersion in terahertz range is negligibly small, the output terahertz pulse got the form of time derivative of pump pulse envelope.

Under phase matching condition the efficiency of deference frequency generation by plane wave pulses can be written as

$$h_{THz} = \frac{2\Omega^2 d_{eff}^2 L^2 I}{e_0 n_{NIR}^2 n_{THz} c^3} * e^{-\frac{a_{THz} L}{2}} * \frac{\sinh^2[\frac{a_{THz} L}{4}]}{(\frac{a_{THz} L}{4})^2} \quad (6)$$

where  $d_{eff}$  is effective nonlinear coefficient,  $a_{THz}$  is absorption coefficient of terahertz waves,  $e_0$  is the permittivity of vacuum, I is the intensity near-infrared pump pulse, L is length of the nonlinear crystal and  $n_{nir}$  and  $n_{THz}$  are the refractive indices of crystal in NIR and THz range, respectively. When choosing the crystal length it is important to remember that it should not be longer than penetration depth of THz wave and it should be smaller than coherence length in order to avoid conversion cancellation due to the phase mismatch. For the sake of convenience the two figures of merit are used in optical rectification for negligible absorption and the opposite and the defined by

$$FOM_{NA} \equiv \frac{d_{eff}^2 L^2}{n_{NIR}^2 n_{THz}} \quad (7)$$

$$FOM_A \equiv \frac{4d_{eff}^2}{a_{THz}^2 n_{NIR}^2 n_{THz}} \quad (8)$$

In table 1 the most used material for optical rectification and their parameters are presented [14].

Table 1

### Properties of the nonlinear materials which are used for optical rectification

Material	$d_{eff}(\text{pm V}^{-1})$	$n_{800nm}^g$	$n_{THz}(\text{for 1 THz, DAST 0.8 THz})$	$n_{1.55\mu m}^g$	$a_{THz}(cm^{-1})$	FOM ( $\text{pm}^2 \text{cm}^2 \text{V}^{-2}$ )
CdTe	81.8		3.24	2.81	4.8	11.0
GaAs	65.6	4.18	3.59	3.56	0.5	4.21
GaP	24.8	3.67	3.34	3.16	0.2	0.72
ZnTe	68.5	3.13	3.17	2.81	1.3	7.27
GaSe	28.0	3.13	3.27	2.82	0.5	1.18
sLn	168	2.25	4.96	2.18	17	18.2
sLn, 100K					4.8	48.6
DAST	615	3.39	2.58	2.25	50	41.5

As one can see, under room conditions the DAST organic crystal got the highest FOM value, but generation of strong pulses is limited in this one because of small damage threshold. In order to use it with more powerful pump sources, further improvement is required. CdTe also got quite high FOM in room temperature, but because of the strong absorption at 800 nm the generation of THz radiation require not common pump source. ZnTe is the most spread material in THz generation field due to its 4-th FOM value and ability to approximately fulfill velocity matching condition in collinear geometry. As down side, it has strong two-photon absorption at 800 nm which increases THz absorption due to increase free carrier density. New word in generation of THz using optical rectification is the use of stoichiometric Li NbO<sub>3</sub> (sLN). In this crystal THz waves strongly interact with optical phonons giving rise to phonon-polariton modes. These modes contribute greatly to the FOM value. Also relatively big band gap allows only three-photon absorption, which allows generation of high-intense THz pulses due to the higher possible pump intensities. Main drawback is significant difference between the group refractive index and THz refractive

index, meaning that no collinear velocity matching is possible. Improved phase matching can be reached by using poled LN.

In order to achieve high conversion efficiencies with LN the best way is to use the most progressive technique which is called “tilted-pulse-front pumping”. It was proposed in 2002 and the idea is to tilt the pumping intensity front with the goal to achieve the velocity matching inside the crystal. This is illustrated on figure 1.

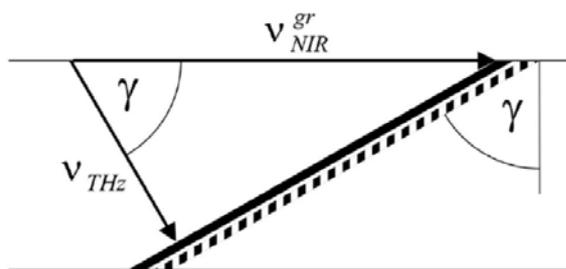


Fig. 1. Illustration of pump and THz wave fronts (bold and dashed lines, respectively) and their velocity directions [13]

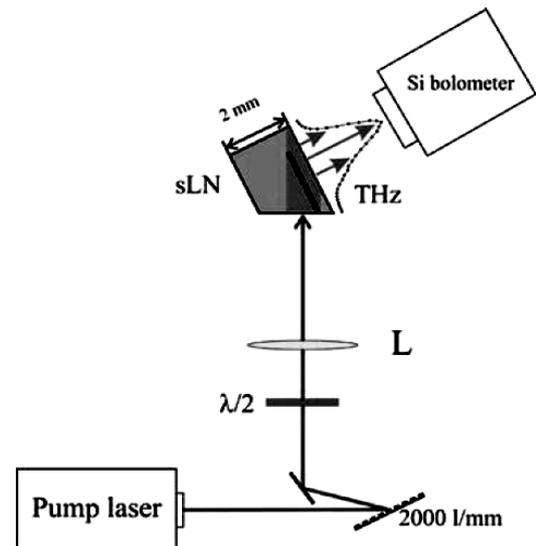


Fig. 2. Experimental setup for THz generation using TFPF [4]

As one can see from this figure, the fronts of THz pulse and pumped pulse are parallel. This is achieved by tilting the pump wave front with the use of, for example, diffraction grating. Also it is shown that THz wave propagates perpendicularly to the pump pulse front that corresponds to Huygens principle. The tilt angle  $\gamma$  here is the angle between pump group velocity  $V_{NIR}^{gr}$  and terahertz phase velocity  $V_{THz}$  which plays the main role in the phase matching. This angle needs to satisfy condition

$$V_{NIR}^{gr} \cos g = V_{THz} \quad (9)$$

In case of LN with large difference between refractive indices  $n_{nir}$  and  $n_{THz}$  ( $\sim 2$  and  $\sim 5$ , respectively) the required tilt angle is as large as  $63^\circ$ – $65^\circ$ . On figure 2 we have simple setup to generate THz waves using nonlinear sLN crystal.

In this setup, the tilt of the pump pulse front is achieved by the grating. Half-wave plate is used for polarization of the pulse and the lens is for focusing. Elements such as grating, lenses and other which are on the way of a pump pulse introducing the angle dispersion which affects the tilt angle and cause the decrease of conversion efficiency of the setup. Lenses can also introduce aberration which can cause the deformation of THz front.

On figure 3 depicted results of real TFPF THz setup. The Ti:sapphire laser was used as pump source (pulse energy 4 mJ, FWHM 85 fs, central wave length 780 nm, repetition rate 1kHz). The peak value of electric field was 1.2 MV/cm, total pulse energy  $\sim 2 \mu\text{J}$  which means that conversion efficiency is  $\sim 10^{-3}$  [14].

The conversion efficiency of TFPF setup can be improved by cooling down the sLN crystal to cryogenic temperatures. The cooled sLN got the best FOM of all materials, as one can see from table 1. In [15] they used congruent LN because of the large crystal size of the crystal they required. Dependence of THz energy enhancement on temperature is shown on Fig 4. One can see that they reached the saturation of conversion at temperature about 150 K which corresponds to conversion efficiency equal to 3.8 %. They also made the prediction that further decrease of temperature down to 10 K will increase the conversion efficiency up to 13 %.

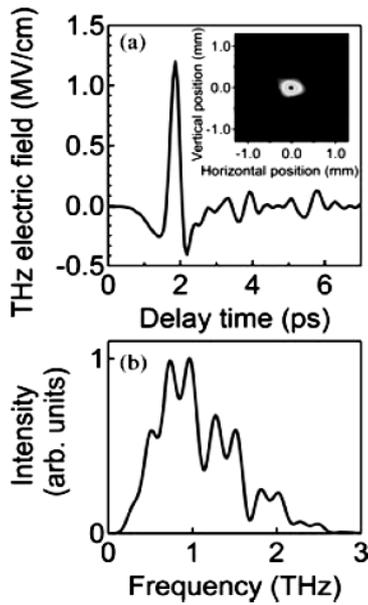


Fig. 3. Measured time profile of THz pulse and its Fourier transform

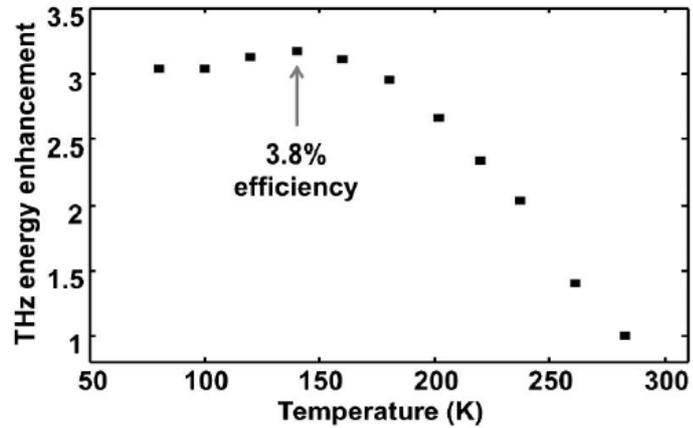


Fig. 4. THz energy enhancement with respect to temperature with fixed pump energy 1.2 mJ

High energy terahertz pulse from tilted pulse front pumping setups causing many interesting phenomena which might be used for new applications as well as improve the older ones. Most common application of THz waves is Time-dependent terahertz spectroscopy which is a very efficient tool for material characterization. Strong THz absorption of polar molecules generates distinct peaks that allow accurate recognition of them. Biological tissues and biological materials, such as DNA, can also be efficiently detected and studied. The use of more powerful sources will make the multidimensional THz spectroscopy possible. High field THz pulses open way for nonlinear THz spectroscopy of semiconductors and superconductors. Many electron properties, such as density, mobility, effective mass affects material response to THz waves. This method can be used to determine impedance of semiconducting material which is important property in design of electronic devices. Resonant interaction of THz field with semiconductors and gases can generate many nonlinear phenomena such as Stark effect, Rabi oscillations, Autler-Townes splitting and induced transparency. Main concept which describes these phenomena is ponderomotive energy

$$U_p = \frac{e^2 E^2}{4m^* \omega^2} \quad (10)$$

where E is electric field amplitude of incoming THz pulse, e is electron charge,  $m^*$  is effective mass of electron and  $\omega$  is the angular frequency of incoming pulse.

Phonon properties can be also investigated by THz wave emission spectroscopy which is generated from photon-induced carriers and interacts with optical phonons of semiconductor. Electrons in semiconductor can be accelerated into ballistic regime which can drive Frohlich polaron into highly nonlinear regime which can be used to investigate lattice distortions.

Huge electric field around 1 MV/cm can cause impact ionization effect. Such amplitude of the field can be achieved by 1THz pulses. Impact ionization causes increase of carrier density due to the impact of high energetic conduction band electrons with valance band electrons. Results in [14] shown the increase of free carrier density in GaAs by 3 orders of magnitude. This means that strong THz wave field can be used for temporary modification of semiconductor surface as in case of photoactivation. Since TFP technique corresponds to those requirements it is reasonable to use it as basic approach for THz source design.

## Conclusions

In this paper, we made the review of common terahertz wave generation techniques and described the basic principles of tilted pulse front pumping as the method of terahertz electromagnetic wave generation. This method is the most effective approach to generate such waves and further development can increase the conversion efficiency even more.

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