

# THz Generation and Detection by Nonlinear Optical Methods

Sourabh Mukhopadhyay

Department of Physics, Jhargram Raj College (Govt. of West Bengal), Jhargram, INDIA

\*Corresponding Author: [smukhopadhyay.phy@gmail.com](mailto:smukhopadhyay.phy@gmail.com)

Available online at: [www.isroset.org](http://www.isroset.org)

Received: 09/Mar/2018, Revised: 16/Mar/2018, Accepted: 29/Mar/2018, Online: 30/Apr/ 2018

**Abstract**—This article presents a comprehensive review on efficient THz generation by optical rectification and detection by electro-optic sampling. Starting with the basics of optical rectification and electro-optic field sensor, chronological development of the subject is presented. Performance of several zinc-blende crystals for THz generation as well as electro-optics sampling is discussed in detail and their comparative superiority relating to different crystals parameters are justified with experimental results. Organic crystals, due their higher nonlinearity, are important for THz generation by optical rectification and got special attention in this article. Phase-matching for THz generation by optical rectification is always a challenge and this issue is addressed with different schemes employed till date and optical rectification in tilted pulse front geometry is elaborated since it can dramatically increase the effective interaction length by achieving phase-matching. Material absorption in THz domain, which limits the THz generation efficiency, is however overcome by cryogenic cooling and performance of periodically poled lithium niobate under such low temperature for optical rectification in different schemes is presented in detail. The success in continuously increase of THz generation efficiency promises future high power THz devices for their application in basic science and technology. Recent propositions and theoretical models with predictions of orders of magnitude increase in the THz generation efficiency naturally pave the path with future challenges for the experimentalists. This article clearly figures out the directions of future research for THz generation by optical rectification and detection by electro-optic field sensor.

**Keywords**—*Optical rectification, Difference frequency generation, Phase-matching, Electro-optic sampling,*

## I. INTRODUCTION

There has been a consistently growing research interest for last two decades, in the field of generation and manipulation of THz wave which occupies the frequency range from 0.1 to 10 THz of the electromagnetic spectrum. This THz spectral range is of prime interest today due to its wide spread application in science and technology, for example, in studying collective phonon dynamics, excitation of spin and electronic degree of freedom [1], free carrier absorption [2], time resolved spectroscopic and carrier dynamics studies [3-5], resonant control over material parameters [6] non-perturbative intraband response in antimonide [7], electron hole re-collision in semiconductor quantum structure [8], communication, security applications [9] and many more. Being a low energy non-ionizing radiation THz radiation is promising for future medical application, for example, in biomedical imaging [10,11], cancer detection [12], DNA repair and manipulation [13] etc. THz radiation with high field strength is also believed to be very promising to develop table top, compact charge particle accelerator [14].

There has been several approach for generating either few cycle broad band or multi-cycle narrow band THz radiation. The most conventional method was to excite biased photo-

conductive switch by femtosecond (fs) laser pulse [15,16]. The photoconductive switch is essentially a semiconductor structure having two metal electrodes implanted on it. The choice of semiconductor material depends on the excitation laser wavelength. For Ti:Sapphire laser GaAs is a good choice since the band gap energy is less than the photon energy of the laser. The electrodes are made up of metals like gold or aluminum having separation  $\sim 10\mu\text{m}$  and biased with voltage 10-50V which produces an electric field  $\sim$ few kV/cm. As the fs laser pulse is incident on the switch, free carriers are generated and are accelerated by the biased field. The rapid change in the polarization induced by the ultrafast laser generates coherent THz radiation which partially propagates along the electrodes and partially in the free space. Several geometry of photo conductive switches have been tried but the scope of this technique is limited for poor conversion efficiency and for not being able to be scaled to higher energies due to THz field saturation [17].

Another promising technique is to use gas plasma for generation of coherent THz radiation. In this technique an fs laser pulse of high pulse energy ( $\sim$ mJ) and having central frequency  $\omega$  is incident on a nonlinear crystal to produce second harmonic ( $2\omega$ ) and then subsequently focussed on to a noble gas to produce gas plasma. The fundamental and the

second harmonic is mixed up in the gas plasma to produce directional electron current as well as coherent THz radiation is by breaking the symmetry of the AC field [18-20]. Free electron laser could also be a good choice for generation of high power THz radiation [21], however, is expensive and thus access to such source is limited.

Most popular and promising technique for coherent THz generation, till date, is to use nonlinear optical effect namely ‘Optical rectification’ (OR). This technique relies on series of difference frequency generation (DFG) between successive frequency components of an ultra-short broad-band optical pulse. This technique is the most emerging one because of its simplicity and capable of generating high power broad band single or multi-cycle coherent THz radiation. THz generation by OR was first demonstrated in zinc-blende crystal structure and later on many inorganic as well as organic crystals have been employed to achieve higher efficiency. Innovative approaches like tilting of incident pulse front have been tried and THz conversion efficiency was enhanced.

This paper aims to provide comprehensive review of coherent THz generation by optical rectification [22,23]. Starting with the basics of the technique, a chronological development of the subject and the state of the art scenario is presented. Some experimental results have been reported here which gives a detail understanding on the THz generation by OR. The major issues such as efficiency, choice of nonlinear crystal, excitation laser characteristics are addressed and different innovative modifications for producing high energy THz radiation have been discussed and compared in detail. Detection of THz radiation is another issue and therefore the technique of electro-optic sampling (EOS) in zinc-blende crystals has been discussed in detail [24,25]. The EOS has the potential to detect THz radiation irrespective of its pulse energy, spectrum and cycle. Theory of EOS is presented along with experimental results obtained to identify the promise, challenges and also to address several issues with this technique.

## II. OPTICAL RECTIFICATION

Optical rectification is essentially a second order nonlinear optical phenomenon first observed in 1962 [26]. If an intense laser pulse is incident on a second order nonlinear optical crystal it produces a dc polarization across the crystal by way of difference frequency generation [27] of the incident pulse spectrum. This phenomenon is exploited for generation of THz pulse from several crystals by use of a broad band optical pulse. An fs optical pulse has a broad bandwidth, however the frequency components are discrete by virtue of mode-locking. The separation between any two adjacent frequency components which lie in THz domain is constant and determined by the cavity round trip time of the laser

oscillator. These closely spaced frequencies are rectified by way of DFG and results in producing an optical polarization for THz field. The THz polarization can be written as [27,28]

$$P^{(2)}(\Omega) = \epsilon_0 \int d_{ijk}^{(2)}(\Omega: \omega + \Omega; -\omega) E_j(\omega + \Omega) E_k^*(\omega) d\omega \quad (1)$$

Here  $d^{(2)}$  represents the second order nonlinear coefficient tensor,  $E$  the electric field and  $\omega$  and  $\Omega$  the optical and THz frequency respectively. However, for a polarized input fs pulse the electric field components will have the same polarization ( $j = k$ ). THz generation efficiency will depend choosing the geometry such that maximum value of  $d$  tensor can be utilized. For zinc-blende crystals, under contracted notation of  $d$  tensor, only three elements will be non-vanishing ( $d_{14} = d_{25} = d_{36}$ ) [29]. However, the THz efficiency will also depend crucially on phase matching of the optical and the THz wave. For THz generation as in (1), the phase matching condition is

$$\Delta k = k(\omega + \Omega) - k(\omega) - k(\Omega) = 0 \quad (2)$$

Here  $k$  represents the wave vector. An equivalent quantity related to the phase matching is the coherence length, which determines the length of the crystal for which generation is possible and if we neglect the dispersion in the optical spectral range can be written as [22],

$$l_c = \frac{\pi c}{\Omega |n_{opt} - n_{THz}|} \quad (3)$$

Where  $n_{opt}$  and  $n_{THz}$  are the refractive indices at the optical and the THz frequency. Since the difference of the refractive indices at the THz and the optical frequency is very large the coherence length becomes very small. However, considering the dispersion in the optical range itself, many fold increase of the coherence length is possible. If we consider the dispersion in the optical spectrum the phase matching condition can be reduced to [22]

$$\frac{k(\Omega)}{\Omega} \approx \left( \frac{\partial k}{\partial \omega} \right)_{opt} \quad (4)$$

This implies that in modified phase matching condition, the phase velocity ( $v_p$ ) of the THz wave should be equal to the group velocity ( $v_g$ ) of the optical pulse. The coherence length in such case can be brought in a form [22]

$$l_c = \frac{\pi c}{\Omega \left| n_{opt} - \lambda_{opt} \left( \frac{dn_{opt}}{d\lambda} \right)_{\lambda_{opt}} - n_{THz} \right|} \quad (5)$$

In practice sufficient group velocity dispersion (GVD) exists in the optical domain. ZnTe, which is a very promising crystal for THz generation has sufficient GVD exists [29] around 800nm such that phase matched THz around frequency of 2 THz can be efficiently generated.

### III. ELECTRO-OPTIC SAMPLING

Electro-optic sampling is an easy and effective way for detection of THz wave [22,30,31]. The process relies on the electric field induced birefringence of optical crystals. In this process the THz and a linearly polarized fs optical probe pulse is co-propagated in electro-optic (EO) crystal. The presence of THz field instantaneously modulates the index ellipsoid of the EO crystal and introduces a phase difference between the different components of the optical probe beam. The introduced phase difference can result in a polarization change of the probe beam. Therefore, if the output optical beam is analyzed with an analyzer, the electric field strength of the THz pulse can be figured out. Let us describe the process of EOS for the zinc-blende crystals, e.g. ZnTe. Zinc-blende crystals possess  $\bar{4}3m$  point group symmetry and only three elements of the electro-optic tensor will be non-vanishing ( $r_{14} = r_{25} = r_{36}$ ). The crystal doesn't have birefringence; however, THz field can introduce birefringence in the crystal. Let us consider THz and the optical probe, both linearly polarized, are incident on the (110) plane of ZnTe crystal. The geometry is elaborated in Figure 1.  $x[1,0,0], y[0,1,0]$ , and  $z[0,0,1]$  represents the

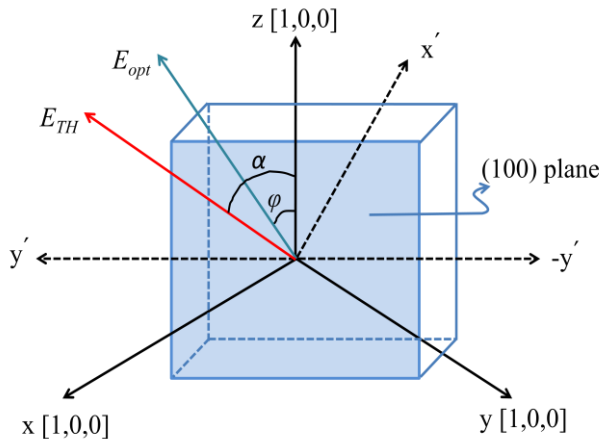


Figure 1. Geometry of (110) cut ZnTe crystal used as EOS of THz.

crystallographic axes of the ZnTe. The electric field of the THz wave ( $E_{THz}$ ) and the electric field of the optical probe ( $E_{opt}$ ) make angles  $\alpha$  and  $\varphi$  respectively with the crystallographic  $z$  axis. The non-birefringent ZnTe becomes birefringent under the influence of the THz field and the modified index ellipsoid becomes [30],

$$\frac{x^2 + y^2 + z^2}{n^2} + 2 E_{THz,x} r_{41} yz + 2 E_{THz,y} r_{41} xz + 2 E_{THz,z} r_{41} xy = 0 \tag{6}$$

Here  $n$  is the unperturbed refractive index in absence of THz field. It should be noted that for the THz pulse propagating in (110) direction,  $E_{THz,x} = -E_{THz,y}$ . The presence of terms containing  $yz, xz$  and  $xy$  indicates that the principal axes of the modified index ellipsoid do not coincide with the crystallographic axes. However, new set of axes can be found so that the same would be the principal axes of the index ellipsoid. Let us make it with two successive coordinate transformations. Firstly, a rotation of axes by angle  $45^\circ$  is made with respect to the  $z$  axis so that the new set of axes ( $x', y', z'$ ) such that [31]

$$\begin{aligned} x &= \frac{x'}{\sqrt{2}} - \frac{y'}{\sqrt{2}} \\ y &= \frac{x'}{\sqrt{2}} + \frac{y'}{\sqrt{2}} \\ z &= z' \end{aligned} \tag{7}$$

With the help of the new transformed coordinate the index ellipsoid can be written as [31]

$$x'^2 \left( \frac{1}{n^2} + E_{THz,z} r_{41} \right) + y'^2 \left( \frac{1}{n^2} - E_{THz,z} r_{41} \right) + \frac{z'^2}{n^2} + 2\sqrt{2} E_{THz,x} r_{41} y' z' = 1 \tag{8}$$

Finally it is evident that a rotation around the  $x'$  axis is needed to align new coordinate system ( $x'', y'', z''$ ) with the principal axes of the ellipsoid. Designating the required angle of rotation  $\theta$ , the transformation relation takes the form [31]

$$\begin{aligned} x' &= x'' \\ y' &= y'' \cos \theta - z'' \sin \theta \\ z' &= y'' \sin \theta + z'' \cos \theta \end{aligned} \tag{9}$$

Noting that  $E_{TZ,z} = E_{THz} \cos \alpha$  and  $E_{THz,x} = -E_{THz,y} = \frac{E_{THz}}{\sqrt{2}} \sin \alpha$ ; the finally transformed index of ellipsoid can be shown to have the form [32]

$$\begin{aligned} &x''^2 \left( \frac{1}{n^2} + E_{TH} r_{41} \cos \alpha \right) \\ &+ y''^2 \left\{ \frac{1}{n^2} - E_{THz} r_{41} [\cos \alpha \sin^2 \theta + \cos(\alpha + 2\theta)] \right\} \\ &+ z''^2 \left\{ \frac{1}{n^2} - E_{THz} r_{41} [\cos \alpha \cos^2 \theta - \cos(\alpha + 2\theta)] \right\} \\ &= 1 \end{aligned} \tag{10}$$

With the condition that

$$2\theta = -\arctan(2 \tan \alpha) - n\pi$$

$$\text{and } \left(n - \frac{1}{2}\right)\pi \leq \alpha \leq \left(n + \frac{1}{2}\right)\pi, n = 0, 1, 2, \dots \quad (11)$$

If the electric field is not very strong, the refractive indices corresponding to propagation along  $y''$  and  $z''$  direction can be approximated as [31]

$$n_{y''} \approx n + \frac{n^3}{2} E_{THz} r_{41} [\cos \alpha \sin^2 \theta + \cos(\alpha + 2\theta)] \quad (12a)$$

$$n_{z''} \approx n + \frac{n^3}{2} E_{THz} r_{41} [\cos \alpha \cos^2 \theta - \cos(\alpha + 2\theta)] \quad (12b)$$

Thus it is evident that the polarization component along  $y''$  and  $z''$  of the optical probe suffers different refractive indices and there will be an accumulated phase difference while propagating through the ZnTe. The final polarization of the output optical polarization will be elliptic.

In experiment a balanced photo detector is placed after the EO crystal. The balanced photo detector comprises of a quarter-wave plate, Wollaston prism and two identical photo-diodes. In absence of the THz field the linearly polarized optical wave becomes circularly polarized by the quarter-wave plate and divided in two equal parts by the Wollaston prism. The two equal parts are fed into the two identical photo-diodes and the difference signal is monitored via Lock-in amplifier. In absence of the THz field the signal will be null. However in presence of the THz field, the incident beam to the quarter-wave plate becomes elliptical and finally the Wollaston prism splits the beam in two unequal parts, resulting in a difference signal in Lock-in amplifier. The difference signal can be shown to have the form [31]

$$\Delta S \propto I_p \frac{\omega^3 n^3 E_{THz} r_{41} L}{2c} (\cos \alpha \sin 2\varphi + 2 \sin \alpha \cos 2\varphi) \quad (13)$$

Here  $I_p$  is the probe intensity on the EO crystal and  $L$  is the EO crystal length. The difference signal is traced as function of the delay between the probe and the THz pulse and it gives the actual replica of the THz field. It is evident that the signal can be maximized by rotating the EO crystal and the maximum signal can be obtained for  $\varphi = \alpha + 90$  or  $\varphi = \alpha$ .

#### IV. THZ GENERATION AND DETECTION

The study of THz generation by OR started in 1990s. Rice *et.al.* studied THz generation in CdTe InP and GaAs crystals via OR [32]. They used Ti:Sapphire laser oscillator generating 200fs pulse train with 76 MHz repetition rate and having fluence of  $48 \text{ nJ/cm}^2$ . The laser was incident normally on (110) crystallographic plane and the efficiency was maximized by varying the azimuthal angle of the crystals. Greater efficiency was observed for GaAs and reason was dispersion of second order susceptibility. They used a dipole antenna as detector of the THz and the probe pulse from same laser was used to gate the emitted radiation (THz) from

zinc-blende crystals. The emission was detected by measuring the transient photo current but the wavelength not determined. In 1995, Bovalet *et. al.* studied THz generation exclusively in  $\langle 110 \rangle$  GaAs crystal [33]. A Ti:sapphire oscillator delivering 10–15 fs pulses at a 100 MHz repetition rate and wavelength spread from 0.72 to 0.88 nm was used to pump the GaAs crystal. Nitrogen cooled HgCdTe detector, which has a sharp cut off at 15  $\mu\text{m}$  was used to detect the rectified signal and the emitted wavelength was not conclusively determined due to detector limitation. The proper detection of THz radiation by EOS was first demonstrated in 1996 by Wu *et. al.* [34]. The THz was generated by OR from GaAs crystal which was attached with metal coated glass. They used LiTaO<sub>3</sub> crystal as electro-optic crystal for EO sampling and finally concluded that ZnTe demonstrated superior performance for free space EOS. Nahata *et.al.* described a wideband coherent THz spectroscopy system using optical rectification for the generation of THz radiation and electro-optic sampling for the coherent detection of this radiation. They used  $\langle 110 \rangle$  cut ZnTe crystal for both rectification and the EOS [22]. A mode-locked Ti:sapphire laser producing 130 fs pulses at 800 nm with 76 MHz repetition rate was used to pump 0.9mm thick ZnTe crystal for THz generation by OR. Most important part of their experiment was that they succeeded to achieve phase-matching for the OR. Although they measured the frequency range 0-4THz, the coherence length was found to exceed the crystal length of 0.9mm for  $\sim 2.5$  THz and therefore the higher frequency THz beyond 2.5 THz was shown to have attenuated significantly. A broad bandwidth of  $\sim 4$  THz and centered around 3.6 THz was generated from GaAs crystal by OR and the same was detected by free space EOS using a GaP crystal [35]. A comparison was made and concluded that  $r_{41}$  was found to be greater for ZnTe than GaP. This observation certainly indicated the superiority of ZnTe among the zinc-blend crystals. A good comparison was made between ZnTe,  $c$ -cut LiNbO<sub>3</sub>, and  $b$ -cut LiTaO<sub>3</sub> in respect of their performance to be used as electro-optic sensor and it was found that signal to noise ratio was found to be orders of magnitude high for ZnTe [24]. This observation indicated the promise of ZnTe for a good electro-optic sensor. An innovative technique of using a quarter wave plate to produce extraordinary and ordinary polarization was demonstrated and the phase-matching for OR was achieved in GaSe crystal to generate bandwidth limited THz pulse [36]. It was shown that although two strong phase-matching peaks was observed at 8 and 25 THz, the presence of lattice resonance at 7.1 THz the emission at 7.1 THz was much narrower. GaSe become important for phase-matched OR and THz generation because of the inherent birefringence present in it [37] but it was difficult to process such crystal and the use was limited. ZnTe became more attractive in terms of availability, durability and performance for both OR and EOS. Ti:Sapphire laser pumped ZnTe, proved to be very promising for broad band,

tunable source of TH radiation and with shaped pulse shape by liquid crystal modulator [38]. A typical set-up THz generation by OR and detection by EOS, based on ZnTe crystal, as we used in our experiment shown schematically in Figure 2. A Ti:Sapphire fs laser oscillator which provides

length 75 mm. The weak reflected part of the 808 nm radiation from the pellicle beam splitter is used as the probe beam. The probe beam is first passed through a computer

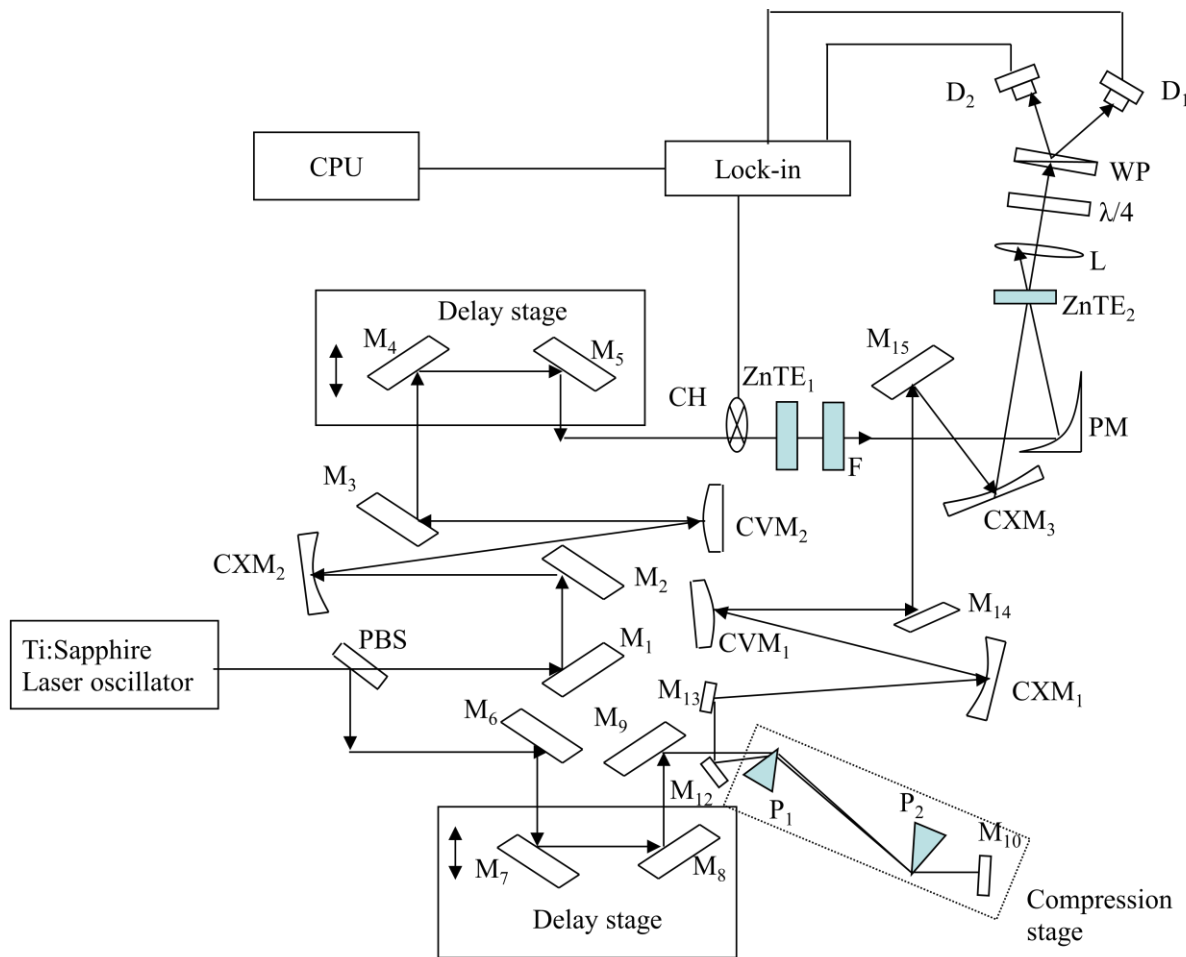


Figure 2. THz wave generation by nonlinear optical pulse rectification, and detection of the same by electro optic field sensor. PBS, Pellicle beam splitter; M<sub>1</sub> ... M<sub>15</sub>, gold plated flat mirror. CXM<sub>1</sub> and CXM<sub>2</sub>, plano-concave gold mirror of focal length 600mm; CVM<sub>1</sub> and CVM<sub>2</sub>, plano-convex gold mirror of focal length 250 mm; P<sub>1</sub> and P<sub>2</sub> right equilateral fused silica prism; ZnTe 1 & ZnTe 2, <110> cut Zinc Telluride crystals; F, Teflon filter; CH, chopper; λ/4, quarter-wave plate; WP, Walston prism; PM, L, Convex lens of 50 mm focal length Gold coated off-axis parabola; D1 and D2 Si photo-detector.

45 fs pulses at 808 nm with 100 MHz repetition rate was used in this experiment. The average laser power of 40 mW was available for the experiment. A Pellicle beam splitter of R:T of 8:92 is used to split the beam in two parts. The transmitted stronger part is collimated by using a concave and a convex mirror of focal length 600mm and 250 mm respectively and it is used as the pump for THz generation. A <110> cut ZnTe crystal of thickness 500 μm is pumped by the pump beam to generate THz wave after. The residual 808 nm radiation is filtered out for the THz wave using a teflon filter and the THz wave is then focused on a ZnTe crystal (ZnTe<sub>2</sub>) by using a gold plated off-axis parabola of focal

controlled delay stage, and then a compression stage formed by two equilateral prisms made of fused silica. The compressed pulse width as measured by second order autocorrelation using two-photon absorption diode is found to be 24 fs. The compressed probe pulse is then collimated by using a concave and a convex mirror of focal length 600mm and 250 mm respectively and finally focused on the ZnTe crystal. The angle between the pump and the probe beam is kept under 5 degree. The spatial overlapping between pump and the probe beam on the ZnTe crystal is assured by using a pinhole. The THz wave modifies the refractive indicatrix of the ZnTe crystal. The refractive index

value along the axis parallel to the THz electric field is modified due to the Pockel's effect, while the refractive index along the axis perpendicular to the THz electric field remains unaltered. As such, under the influence of the THz field the ZnTe crystal becomes birefringent, which changes the polarization state of the probe beam. First the THz wave is blocked, and the linearly polarized probe beam, after passing through the ZnTe crystal is incident on a quarter wave plate, followed by a Wollaston prism. The quarter wave plate makes the probe beam to be circularly polarized. The circularly polarized beam is then split in two equal parts of linearly polarized beam by a Wollaston prism and is detected by two Si photo-diodes connected to lock-in amplifier in differential mode. When THz wave is not

temporally overlapped with the probe beam by using the

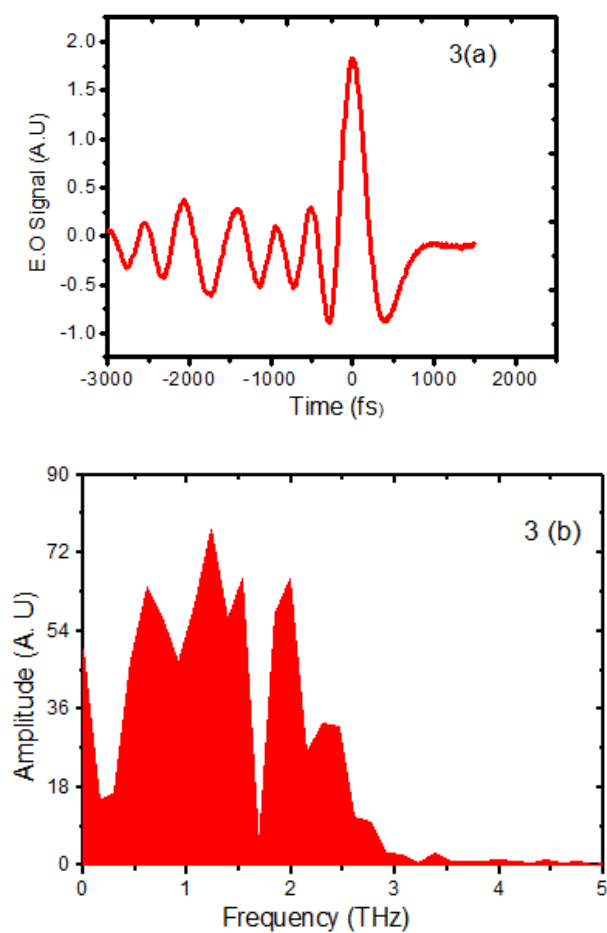


Figure 3. (a) electro-optic signal detected by 200µm thick (110) ZnTe crystal; (b) Corresponding THz spectrum obtained by FFT of (a)

present, the two photo-diodes are balanced. Now the THz wave is made to incident on the ZnTe sensor and is

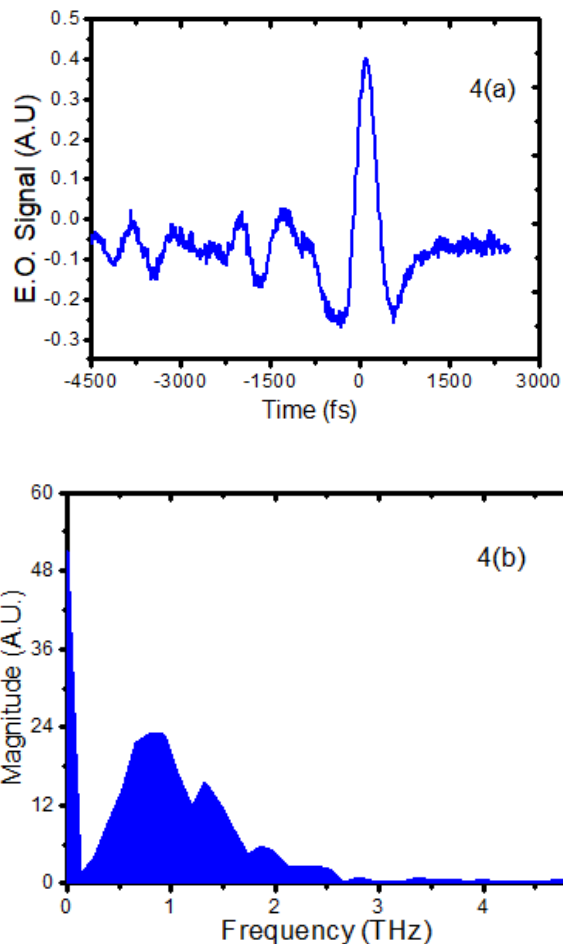


Figure 4. (a) electro-optic signal detected by 20µm thick (110) ZnTe crystal; (b) Corresponding THz spectrum obtained by FFT of (a)

delay stage. Under the action of the THz field the polarization of the probe beam becomes slightly elliptical and the two photo-diodes become unbalanced. The unbalanced signal is detected through a lock-in amplifier, which is triggered by the signal from chopper used on the pump beam. The cross-correlation signal is thus obtained as a function of time as the probe is scanned over the THz beam in temporal domain. Two detection crystals have been used to detect the electro-optic signal. Figure 3(a) shows the electro-optic signal detected for a ZnTe (ZnTe<sub>2</sub>) crystal of thickness 200 µm. The Fourier spectrums of the THz field envelop which shows the frequency spectrum of the THz radiation is shown in Figure 3(b). Electro-optic signal for 10 µm ZnTe sampling crystal is shown in Figure. 4 (a) and corresponding frequency spectrum is shown in Figure 4(b). The reduction in sampling crystal thickness increases the signal to noise ratio. It also helps reducing the group velocity mismatch between the THz and the probe pulse and thus the



larger bandwidth of the THz pulse can be detected. Since the cross-correlation signal is taken as the ensemble average over a large number of pulses the reconstruction of the replica of the THz electric field is possible only when the carrier envelope phase of the pulse stable from pulse to pulse. It thus proves the stability of the carrier envelope phase in a rectification process and is very useful to study ultrafast charge carrier dynamics and generation. The carrier envelope phase can also be measured precisely by the charge symmetry produced air plasma [39]. The choice of thickness of the crystal is very important for generating efficient THz by OR. Very thin crystal can reduce the efficiency and on the other hand a thick crystal can reduce the phase-matching band-width, truncate the spectrum and even can split the THz pulse in time domain [40]. However, more recently, in another report it was shown that thick crystals exhibited a flatter frequency response and since they are mechanically more stable, cheaper, and easier to provide and to handle, Thick crystal could be suitable for EO sensor [41]. Kapfarth *et al.* showed that 250 $\mu$ m thick (110) cut ZnTe is as efficient as 10  $\mu$ m for electro-optic detection of 0.3 to 50 THz domain [41].

Increase in the efficiency of THz generation became a great concern and several methods, crystals and engineered crystals also came in use. Imeshev *et al.* used quasi-phase-matching (QPM) to increase the interaction length and thereby to increase the efficiency of the THz generation [42]. They generated 3.3 $\mu$ W average power, 100-MHz repetition rate source in the range of 1.78 - 2.49 THz from orientation patterned GaAs pumped by a compact all-fiber femtosecond laser at the wavelength of 2 $\mu$ m. Lee *et al.* demonstrated the generation of multi-cycle narrow-bandwidth terahertz radiation based optical rectification in periodically inverted GaAs structures. Three different structures: optically contacted multilayer, orientation-patterned, and diffusion-bonded GaAs were used and it was possible to generate tunable 0.8 to 3 THz radiation by use of different structure periods [43]. Large aperture ZnTe was also used to increase the THz generation efficiency energies up to 1.5  $\mu$ J per pulse and a spectral range extending to 3 THz were obtained using a 100 Hz Ti:sapphire laser source and a 75-mm diameter, 0.5-mm-thick, (110) cut ZnTe crystal, corresponding to an average power of 150  $\mu$ W and an energy conversion efficiency of  $3.1 \times 10^{-5}$  [44]. Another very popular crystal Lithium niobate came up to produce pulses of 125  $\mu$ J energy and with the 0.25% efficiency were generated by OR of 1.3 ps pulses [45]. Experimental findings together with theoretical predictions indicated the feasibility of efficient THz pulse generation with mJ-level output energy by using optimal pump pulse duration, cooling the Lithium niobate crystal, and using a contact grating.

Because of higher nonlinear coefficient, organic crystal attracted attention in the field of THz generation by OR. Broadband terahertz pulses in the frequency range 0.5 and 2.5 THz, have been generated in 2-cyclooctylamino-5-

nitropyridine (COANP) single crystals by optical rectification of 150 fs laser pulses from an amplified Ti:Sapphire laser [23]. Fabian *et al.* claimed COANP to be more efficient than ZnTe for THz generation by OR. THz radiation is generated by OR in an organic salt crystal 4-N,N-dimethylamino-4'-N-methyl stilbazolium tosylate, called DAST, pumped with the signal wavelength of a powerful optical parametric amplifier [46]. Conversion efficiencies up to 2% are reported and carrier envelope phase (CEP) stabilized THz pulses at a center frequency of 2.1 THz with a bandwidth of 2 THz was generated. 1.5% pump to THz energy conversion efficiency was reported and single-cycle broadband terahertz pulses with a central frequency of 1.5 THz was generated by OR from organic salt crystal 2-[3-(4-hydroxystyryl)-5,5-dimethyl cyclohex-2-enylidene] malononitrile or OH1 [47]. High-field THz single-cycle pulses with 1:5 MV/cm are generated by optical rectification in the stilbazolium salt crystal 4-N,N-dimethylamino-4'-N'-methyl-stilbazolium 2,4,6-trimethyl benzenesulfonate (DSTMS) with conversion efficiency of 0.8% [48]. In another report THz conversion efficiency of 3.2 % and THz pulse energy of 270  $\mu$ J claimed to be obtained from OH1 crystal [49].

In order to select proper crystal for THz generation by OR a figure of merit (FOM) was introduced. The FOM of a crystal for THz generation without considering the absorption loss is defined as [50]

$$FOM \equiv \frac{d_{eff}^2}{n_{opt}^2 n_{THz}} \quad (14)$$

And the same but considering the absorption loss is defined as

$$FOM_A \equiv \frac{d_{eff}^2}{n_{opt}^2 n_{THz} \alpha_{THz}^2} \quad (15)$$

Here  $\alpha_{THz}$  represents the absorption coefficient in THz domain. Table 1 summarizes crystal parameters and FOM of some selected crystals corresponding to pump wavelength of 800nm [50].

Here  $r$  represents the electro-optic coefficient,  $n_{opt}$ ; the refractive index at optical wavelength and  $n_{opt}^{gr}$ ; the group refractive index of optical pulse.

The organic crystals, due to their higher nonlinearity seemed to be attractive however is not always suitable for nonlinear optics in general. The crystals are fragile, susceptible to atmosphere and therefore difficult to handle in experimental conditions. LiNbO<sub>3</sub>, on the other hand is very popular due to its robustness, higher nonlinearity, availability of good quality large crystals and high damage threshold. The challenges of THz generation in LiNbO<sub>3</sub> are two-fold; THz absorption and difficulty to achieve phase-matching. The refractive indices of LiNbO<sub>3</sub> at optical frequencies is  $\sim 5$

Table1. Properties of some crystals and their merit for THz generation by OR

Crystal	$r$ (pm/V)	$d$ (pm/V)	$n_{opt}$	$n_{opt}^{gr}$	$n_{THz}$	$\alpha_{THz}$ (cm <sup>-1</sup> )	$FOM \left( \frac{pm^2}{V^2} \right)$	$FOM_A \left( \frac{pm^2 cm^2}{V^2} \right)$
GaAs	1.43	65.6	3.68	4.18	3.61	0.5	87.9	352
GaP	0.97	24.8	3.18	3.57	3.34	1.9	18.2	5.0
ZnTe	4.04	68.5	2.87	3.31	3.17	1.3	180	106
GaSe	1.7	28.0	2.85	3.13	3.72	0.07	25.9	5300
LiTaO <sub>3</sub>	30.5	161	2.145	2.22	6.42	46	882	0.4
LiNbO <sub>3</sub>	30.9	168	2.159	2.23	5.16	16	1170	4.6
DAST	77	618	2.38	3.31	2.4	150	28000	1.2

while it is ~2 for THz frequency range. The large difference in the refractive indices makes it difficult to achieve birefringence phase-matching for THz generation in LiNbO<sub>3</sub>. To counter this problem, an innovative technique of OR using tilted pulse-front (TPF) of the optical pump pulse was introduced and significant enhancement in the THz

phase-matching is achieved. The velocity matching condition under TPF geometry can be written as

$$v_{opt}^g \cos \gamma = v_{THz}^p \tag{16}$$

It should be remembered that in LiNbO<sub>3</sub>  $v_{opt}^g > v_{THz}^p$ , but presence of the tilt in the intensity front of the optical pulse makes it possible to achieve phase-matching. The experimentally observed THz generation efficiency was far below that predicted theoretically in case of TPF geometry. The reason of such difference in theoretical and experimentally observed efficiency is the following. There is an spectral broadening of the optical pulse associated with the THz generation and this happens due to cascaded down frequency shift of the optical pulse. Due to this large spectral broadening dispersive effect gets emphasized and the conversion efficiency drops with the propagation [58,59]. However, in comparison to the traditional OR, the THz efficiency was increased notably in TPF geometry. THz generation efficiency varying from 0.25% to 1.15% was observed at room temperature by OR in TPF geometry [54-57,60]. The THz absorption was almost eliminated by cryogenically cooling and a conversion efficiency of 3.7% was recorded in congruent LiNbO<sub>3</sub> [61].

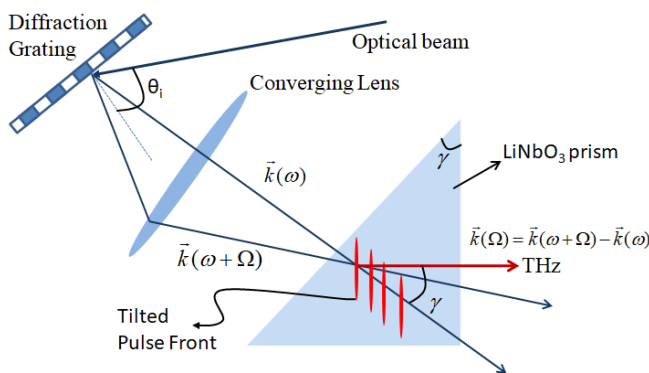


Figure 5. Geometry and phase-matching scheme in Tilted pulse front geometry in LiNbO<sub>3</sub>

generation efficiency was achieved [50-57]. The principle of OR using TPF is depicted in Figure 5. In this approach the broad band optical pump pulse is incident on a reflection grating and gets angularly dispersed. Different frequencies of the optical pulse would be dispersed in different angles and in Figure 5 two such wave vectors  $\vec{k}(\omega)$  and  $\vec{k}(\omega + \Omega)$  are shown. The angularly dispersed optical beam is imaged on to a prism shaped LiNbO<sub>3</sub> crystal by use of a converging lens. The dispersed beam has the phase front normal to the propagation direction, however, the intensity front gets tilted with respect to the phase front by an angle say  $\gamma$ . The generated THz beam emerges at an angle  $\gamma$  with respect to the direction of propagation of the optical beam. The apex angle of the prism is chosen close  $\gamma$  so that THz exits normally from the face of the prism. The phase-matching condition in LiNbO<sub>3</sub> prism for OR remains same as (2), however, since the optical beam and the THz travels different distance in LiNbO<sub>3</sub> crystal within same time,

Different schemes are coming up today to increase the THz generation efficiency. Recent theoretical study based on cascaded parametric amplification predicts conversion efficiency more than 8.0% is possible in cryogenically cooled periodically poled LiNbO<sub>3</sub> [62]. This scheme relies on multi cycle low bandwidth pump and seed optical pulse, shifted in frequency by 0.5 THz. The pump and the seed initiate the narrow band THz generation in periodically poled LiNbO<sub>3</sub> by DFG and the generated THz then simultaneously drives the repeated generation of optical lines. The cascaded generation of the optical lines are red shifted by the THz frequency and self start consecutive dramatic cascading process. This process shows exponential growth of THz energy growth over a single pass through a single crystal. In another more practically viable approach uses laser pulse sequences to drive the cascaded difference frequency generation of high energy, high peak-power and multi-cycle terahertz pulses in cryogenically cooled



periodically poled lithium niobate [63]. Conversion efficiency more than 10% has been projected to achieve through this approach. However, both the approaches need experimental verification in future days.

## V. CONCLUSION AND FUTURE SCOPE

Optical rectification has been shown to be an efficient way for generating either single cycle broad band or multi-cycle narrow band THz radiation. This process generates carrier envelope phase stable THz and therefore electro-optic sampling is the best technique for detection of such THz pulse. In optical rectification, optical frequency components get rectified by way of DFG in a nonlinear optical crystal and a low frequency THz field is generated. In the initial stages zinc-blende crystals, like ZnTe, GaAs have been used for THz generation by OR, however getting notable THz generation efficiency and achieving phase-matching were the challenges to overcome. The phase-matching, which in this case implies the matching of the phase velocity of the THz pulse and the group velocity of the optical pump pulse, was difficult to achieve due to large difference of the refractive indices in optical and THz domain. This limits the effective crystal length over which the nonlinear interaction can take place. Consideration of the dispersion of the optical spectrum itself could increase the interaction length and the method OR became promising for efficient THz generation. Several zinc-blende crystals and different pump pulse width have been tried in many innovative experimental configurations, but the efficiency of THz generation was limited to a maximum value ~0.1%. However, large bandwidth of the THz pulse was possible to generate. The employment of organic crystals, owing to their large nonlinearity, could push up the conversion efficiency up to ~0.5%. An innovative idea of tilting the intensity front of the optical pump pulse by way of angular dispersion through the use of a grating and then imaging the same on a LiNbO<sub>3</sub> prism became a very effective and efficient way to achieve phase-matching. This tilted pulse front geometry was able to generate high energy THz pulse with efficiency ~1%. LiNbO<sub>3</sub> became a promising candidate for THz generation because of its high nonlinearity, durability, availability of large crystals with good optical quality and high damage threshold, however, the absorption in the THz spectral domain restricted the effective crystal length and the efficiency of THz conversion. Cryogenic cooling of LiNbO<sub>3</sub> gave a breakthrough to suppress the absorption and an efficiency more 3% has been recorded. New propositions like cascaded parametric amplification and use of laser pulse sequences for cascaded DFG in periodically poled Lithium niobate, are coming up which claims the more than 10% THz generation efficiency is possible in near future. The field of high energy THz generation is becoming more vibrant owing to its tremendous versatile use in basic science and in technology.

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#### AUTHORS PROFILE



Dr. Sourabh Mukhopadhyay did his B. Sc.(H) and M.Sc. in Physics from University of Calcutta and subsequently obtained Ph. D. in Physics from IIT Kharagpur in 2006. He was post doctoral fellow at University of Oxford during 2007-2008. Dr. Mukhopadhyay is working in West Bengal Education Service since 2006 and presently posted as assistant Professor at Jhargram Raj College. He has published more than 15 publications in reputed international journals and numerous international and national conference proceedings including CLEO USA. Present research interest of Dr. Mukhopadhyay includes ultrafast optics, THz sources and devices.