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# PPLN Crystals for Nonlinear-optical Detection of Terahertz Wave Radiation

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# Abstract

Periodically poled lithium niobate crystals can be utilized in nonlinear-optical spectral brightness detectors of terahertz range. In this paper characteristics of detectors were determined by analyzing spectra of spontaneous parametric downand up-conversion in these crystals.

#### Introduction

Apart from fundamental tasks, interest to the terahertz wave radiation arises due to variety of promising scientific and technological applications of this range radiation in spectroscopy, diagnostics, communications, biomedicine, e.g. for study of rotational transitions in organic molecules or explosives detection for security tasks.

At present, there are various techniques proposed for terahertz wave detection. Depending on the information available to obtain, one can divide these techniques into two vast groups. First group of detectors provides one with information about the detected radiation power only. It includes different-type bolometers, pyro-electric detectors, and Golay cells [1]. Operation principle here is the following: the working substance is being heated by the incident terahertz wave radiation. Afterwards, the corresponding changes are detected, e.g. the gas pressure in Golay cells or the conductivity in hotelectron bolometers. Current results on the NEP of this group of devices are  $\sim 10^{-10} W / \sqrt{Hz}$  for common devices or in 2 orders better in the recordbreaking setups. It is also worth mentioning that 'intensity' detectors are the only ones commercially available due to their relative simplicity and reliability. As of the disadvantages, these detectors are not spectral-selective enough and require terahertz range filters or other special devices for spectral analysis, and also have low temporal resolution. Moreover, the high-sensitivity detectors need cryogenic cooling.

The other group allows one to measure the amplitude and phase of the incident terahertz wave radiation. These devices are referred to as terahertz time-domain sampling. The basic idea is to scan the terahertz pulse with the optical so-called 'probe' pulse. It becomes possible due to the temporal difference between femtosecond optical and terahertz pulses:  $\tau_{THz} \ge 10 \cdot \tau_{probe}$ . Modulating the properties of the detector (photocurrent in photoconductive antenna or polarization of electrooptic crystal) with terahertz wave radiation, this change is being measured by the probe pulse [2]. In this method, the probe wave amplitude is proportional to the terahertz wave field  $E_{TH_2}$ . Nonlinear-optical devices based on Pockels effect and sum/difference-frequency generation are among the most common ways for the probe wave modulation by the incident terahertz wave. The resulting data format is the dependence of the probe wave intensity or phase from the delay time. Its Fourier-transform gives one the spectrum of the terahertz wave radiation being measured. In most cases the terahertz time-domain spectroscopy is used in the pulsed regime in order to increase the peak amplitude values. The spectral selectivity of these schemes is dependent on the pump spectrum and the nonlinear medium used for the pump beam modulation.

This article is devoted to the detection method based on non-linear optical up-conversion of the terahertz frequency to the optical range. It is not so well-known as an electro-optical sampling method commonly used in femtosecond time-domain spectroscopy schemes [3], but is applicable for detection of CW and quasi-CW nanosecond-pulsed terahertz wave radiation [4,5]. As it has been shown in our previous papers [6,7], the nonlinear-optical detection of terahertz wave radiation in periodically poled crystals yields a few advantages one can make use of. These are the spectral selectivity controlled via crystal domain structure properties, room-temperature operation, and, in a distant future, possibility of absolute measurements of spectral brightness in the terahertz range without any pre-calibrated terahertz wave sources.

### **The Nonlinear-Optical Detection Method**



The nonlinear-optical detection method is based on the parametric frequency conversion in a nonlinear medium (Fig.1), which allows one to transform THz radiation (at frequency  $\omega_{THz}$  ) into the optical wave ("signal wave", at frequency  $\omega_s$  ). The pump beam at the optical frequency  $\omega_p = \omega_s + \omega_{THz}$ is required for the method to function. LiNbO<sub>3</sub> (LN) or periodically poled LiNbO3 (PPLN) crystals are appropriate nonlinear media. By measuring the spectrum of the signal wave intensity, one can detect the spectrum of the terahertz idler wave intensity. There is a way also to calibrate the spectral brightness of the input terahertz radiation if one measures also the background "noise" signal without any THz radiation at the input of the detector. This approach is based on the idea of D.N. Klyshko [8], first proposed for the optical brightness calibration. Since the background signal appears as a result of spontaneous parametric down-

Intensity. There is a way also to calibrate the spectral brightness of the input terahertz radiation if one measures also the background "noise" signal without any THz radiation at the input of the detector. This approach is based on the idea of D.N. Klyshko [8], first proposed for the optical brightness calibration. Since the background signal appears as a result of spontaneous parametric down-conversion (SPDC), its intensity in case of optical idler waves is proportional the effective brightness of quantum fluctuations of electromagnetic field at the idler frequency. Being equal to 1 photon per mode in quantum units, this effective spectral brightness corresponds to  $B_{\omega} = (\hbar \omega^3 / 8\pi^3 c^2) \times 1$  in radiometric units and serves as a natural brightness standard.

The advantages of this approach can be used in the terahertz range, but the calibration procedure has to be modified, taking into account the thermal field fluctuations at the terahertz idler frequencies. In this case the background signal includes both SPDC signal and the signal of parametric conversion of thermal fluctuations. Thus, the reference background is proportional  $B_{\omega} = (\hbar \omega_{THz}^3 / 8\pi^3 c^2) \times (1 + N_T)$ , where  $N_T$  is the Plank's occupation number taken at the local crystal temperature T and terahertz frequency  $\mathcal{O}_{TH_7}$ . It was shown in our previous works [6,7], that  $N_T$  can be determined exactly if one measures not only signals in the Stokes range, at frequencies  $\omega_s = \omega_n - \omega_{TH_2}$ (difference-frequency generation), but, also, at frequencies  $\omega_{as} = \omega_p + \omega_{THz}$ , that fit the anti-Stokes range and correspond to sum-frequency generation of the optical signal. High level of phase-matching between the optical and terahertz waves both in the Stokes and anti-Stokes ranges simultaneously can be achieved if one uses periodically or aperiodically poled nonlinear crystals.

# Crystals for Nonlinear-Optical Terahertz wave Detectors



Schematic diagram for calculation of the phase mismatch.

The detectors described above utilize nonlinearoptical crystals with a periodical or specially designed aperiodical domain structures that provide the frequencies of signal and idler waves to fall into the range required. The usage of the periodically poled crystals allows one to increase the length of the crystal that generates the signal wave radiation constructively. The phenomenological approach for this is the second-order nonlinear susceptibility  $\chi^{(2)}$  sign change between any two adjacent domains.

Under a parametric frequency conversion process between pump and idler waves, the signal wave brightness  $B_{\omega} = (\hbar \omega_s^3 / 8 \pi^3 c^2) \times N_s$  has the following dependency from the detector crystal parameters ( $N_s$  is the number of photons per mode of signal radiation) [7]:

$$N_{s} = \frac{4\pi^{2} E_{0}^{2}}{c^{2}} \frac{\omega_{s} \omega_{THz}}{n_{s} n_{THz}} L^{2} e^{-\alpha L/2} \left| T(\Delta) \right|^{2} N_{THz}$$
(1)

Here,  $N_s$  and  $N_{THz}$  are the numbers of photons per mode of signal and terahertz-wave radiation correspondingly.  $n_s$  and  $n_{THz}$  stand for the refractive indices,  $\alpha$  is the absorption coefficient at the terahertz frequency, L is the crystal length,  $E_0$  is the pump wave amplitude.  $T(\Delta)$  is a nonlinear transfer function (T-function) [9], which is dependent on the domain structure of the crystal and the mismatch  $\Delta(\omega_{TH_2})$  between optical and terahertz wave vectors (Fig.2). All the influence of the spatial modulation of the second-order nonlinear susceptibility  $\chi^{(2)}$  on the spectral shape of the parametric signal can be described by this parameter. Nonlinear transfer function in a wide range of  $\Delta(\omega_{THz})$  can be measured directly via studying the spontaneous parametric downconversion spectra corresponding to the THz frequency range of the idler waves. Effective

population of all possible idle modes by thermal and quantum fluctuations provide a non-zero optical signal in the widest spectral range. The spectral characteristics of the detector can be determined after that using Eq.(1). In what follows we discuss the experimental procedure of SPDCbased characterization of PPLN and aperiodically poled lithium niobate crystals, which implies measuring the *T*-function and comparing crystal samples of different domain structure periods and quality.

# SPDC-based Characterization of Crystals for Nonlinear-Optical Terahertz Wave Detectors

The example of the SPDC wavelength-angular spectra is given in Fig.3. The following typical areas are presented here: the horizontal line in the center of the picture corresponds to the elastically scattered pump wave; the 'Stokes' SPDC signals are located below it, the 'anti-Stokes' parametric signals are observed above. On the left, the signal wavelength is presented, at the bottom, the scattering angle is. Two horizontal lines at the upper and lower parts of the spectrum correspond to scattering by the E-phonon resonance in PPLN at approximately 152 cm<sup>-1</sup>.



The typical example of the SPDC spectrum in geometry *ooe* obtained for a PPLN crystal.

Crystals with post-growth polarization often have unpredictable properties (e.g. domain structure period). It was shown in [9] that all of the spatial modulation of  $\chi^{(2)}$  and phase mismatch can be taken into account by single T-function (or nonlinear transfer function). In case of the collinear interaction, T-function is defined as:

$$T(\omega_{THz}) = \sum_{m = -\infty} \chi_m \cdot f(\Delta kL + 2\pi m),$$
  

$$\chi_m = \frac{1}{L} \int_{-L/2}^{+L/2} \chi^{(2)}(x) \cdot e^{i\frac{2\pi m}{L}x} dx.$$
(2)

In these terms, intensities of signal waves in processes of (a) spontaneous parametric downconversion and (b) parametric frequency downconversion take forms:

$$I(\omega_s, L) = \frac{\hbar \omega_s^4 (\omega_p - \omega_s)^2 L^2}{n_s n_p k_{THz} c^6} \left| T(\omega_{THz}) \right|^2 I_p \quad (3a)$$

$$I(\omega_{s},L) = \frac{4\pi^{2}\omega_{s}^{2}L^{2}}{n_{s}^{2}c^{2}}e^{-\alpha L/2} \left|T(\omega_{THz})\right|^{2}I_{P} \qquad (3b)$$

First process (a) takes place in the detector crystal even without any incident terahertz wave radiation. When there is a terahertz input, parametric frequency conversion (b) takes place also. For the detected (optical) signal wave is described by the equation (b), all the influence of the detector crystal unique parameters can be taken into account by analyzing spectra of spontaneous parametric downconversion.

The spectrum processing procedure is the following. First, the zero angle line being cleared from single-pixel noise (small white dots in Fig. 3) and is averaged in a small range of angles to decrease the NEP. The resulted signal wave intensity profile is obtained by converting signal wavelength into the terahertz frequency. The typical resulting graph is given in Fig. 4 ('Stokes' region, 0.5-2.5 THz). Here, the gray stripe indicates the CCD-camera readout noise, and the dashed black line indicates the constant "sun" noise. In a case of the same pump wavelength for the detector crystal, this graph gives one the spectral range of the detector directly, as a full width at half magnitude (FWHM) in THz. E.g. Fig. 4 stands for a detector with the spectral range of approximately 1.1-1.7 THz. In case the different pump wavelength will be used to operate the detector crystal, the recalculation should be performed using (3a) and (3b). In this case one should have accurate information on the absorption coefficient in the terahertz range, which is taken into account by the  $e^{-\alpha L/2}$ factor.



Fig. 4 The intensity profile ('Stokes' region @0.5-2.5 THz) at zero angle for the SPDC spectrum in Fig. 3.

### Summary

The nonlinear-optical method for calibration of the terahertz wave spectral brightness has been discussed. For the method operation, the specially manufactured QPM crystals with periodic and aperiodic domain structures are necessary, providing detection of the parametric-conversion signals in both Stokes and anti-Stokes ranges simultaneously. Experimental results have been provided for the detector crystal characterization procedure. It has been shown that one can obtain the spectral range of the detector crystal studying the spectrum of parametric up/down-conversion in this crystal. Recalculation can be performed for the detector usage at a different pump wavelength.

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