Generation of Terahertz Radiation via Nonlinear Optical Methods

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Abstract—There is presently considerable research activity in the development of the new sources in the terahertz (THz) region of the spectrum. Over the past two to three years, many new techniques were developed for generating especially short or especially powerful pulses. New techniques also allow single shot detection of the THz pulse field as well as real-time and near-field imaging. Recent experiments have exploited the unique properties of T-rays to study ultrafast dynamics in systems. In this paper, I will attempt to give a brief overview of THz generation, especially those methods based on second-order nonlinear processes. The fundamental principle of THz wave generation via optical rectification is revisited. We also present several recently developed techniques which will direct the future of THz generation.

Index Terms—Ultrafast nonlinear optics, terahertz wave generation.

I. INTRODUCTION

The terahertz region of the electromagnetic spectrum covers the frequency range from roughly 0.1 to 10THz, which is between the microwave and infrared regimes. Recently, this range have been extended to 40-50THz. Fig. 1 displays the so-called THz gap in frequency domain. Sources in this region have found many applications, such as THz spectroscopy for studies of carrier dynamics and intermolecular dynamics in liquids [1]. In terahertz spectroscopy, we use photons with energies of about 1 meV. This energy scale is important for learning about many important aspects of condensed matter physics. At the same time, this power is not strong enough to harm the organic structure, so it is a promising replacement of X ray in medical imaging.

Another key feature of THz spectroscopy is that instead of measuring the intensity of the THz pulse, we instead measure the electric field. This means that we have access to information not only about the amplitude change but also the phase change of the electric field as the pulse passes through the medium. It means that we can obtain both the real and imaginary components of the index of refraction. This information usually reflects the intrinsic structure of a crystal.

According to the THz range in electromagnetic spectrum, it can be generated from both the optical and the microwave sides. There are techniques of two categories developed for the generation THz pulses. One of them includes free electron lasers [2], photoconductive switching [3] and dipolar antennas. They are sometimes called resonant optical rectification since the THz emission results from the polarization change that follows the transport of excited carriers in an applied or surface electric field. In the far-field, the electric field is proportional to the time derivative of the surface current. The bandwidth of the generated pulse is usually limited to a few THz. The basic concept of this method is sketched in fig. 2.

Another group of methods, which is mostly interested here, is the non-resonant optical rectification based on nonlinear dielectric crystals. Already in the 1970s, it was shown [4] that picosecond pulses could be rectified in LiNbO$_3$ to produce pulses in the far infrared. In the last few years, it was discovered that optical rectification could be a very efficient method for THz wave generation [5]. Other nonlinear methods include OPO and DFG which are intrinsically of similar principle and they are discussed later.

Three nonlinear optical techniques used for generating THz radiation are discussed here: the optical rectification, the difference frequency generation and the parametric generation. They are all based on nonlinear second order processes,
which occur in all non-centrosymmetric materials. Optical rectification makes use of $\chi^{(2)}(0, -\omega, \omega)$ and the other two process make use of $\chi^{(2)}(\omega_1 - \omega_2, \omega_1, -\omega_2)$.

II. THz WAVE GENERATION VIA OPTICAL RECTIFICATION

A. basic principle

The generation of THz radiation by optical rectification is only possible by pulsed laser beams. THz pulses generated from an optoelectronic crystal may be calculated using the nonlinear Maxwell equation. The local THz field is proportional to the second-order time derivative of the second-order nonlinear Maxwell equation. The equation 2.1.11 in [6] gives us

$$\nabla^2 \mathbf{E} + \frac{1}{c^2} \frac{\partial^2 \mathbf{D}^{(1)}}{\partial t^2} = -\frac{4\pi}{c^2} \frac{\partial^2 \mathbf{P}^{(2)}}{\partial t^2}$$

(1)

where the polarization $\mathbf{P}^{(2)}$ is the product of the second-order nonlinear index $\chi^{(2)}$ and the the intensity of the pump laser pulse in spectral domain. We assume the laser pump propagates in the z-direction and the pulse width is $\tau_G$. If its temporal profile is Gaussian or $I(\omega) = I_0(\omega)\exp(-\tau_G\omega^2/2)$, we get

$$P^{(2)}(z, \omega) = \chi^{(2)}(\omega)I(\omega)\exp(i\omega z/v_g)$$

(2)

where $v_g$ is the group velocity of the optical pulse. The solution for (1) is obtained locally as

$$E^{THz}(z, \omega) = \omega^2 \chi^{(2)}(\omega)I(\omega)z \text{sinc} \left( \frac{\Delta k^{THz} z}{2} \right)$$

(3)

where $\Delta k^{THz} = k - \omega/v_g$ is the momentum mismatch between the THz pulse and the pump pulse [7]. Notice (3) is expressed in SI units system. The generated THz pulse is then the integral of local generation. If these is no momentum mismatch, the output signal follows the intensity envelope of the pump laser pulse. Usually we use ultrafast pulses on the order of 100 femtoseconds or shorter as the pump. The spectrum of the generated pulse peaks around a few terahertz. Fig.3 shows a profile of a THz-pulse and its spectrum.

From a simple point of view, the second-order nonlinear crystal will generate polarization of both sum and difference frequencies. Since the laser pulses have large bandwidth, the frequency components are differentiated with each other and the produced signal have frequency component from 0 to the bandwidth of the pump $\Delta \omega$, which is the order of $1/\tau_G$. For femtosecond laser pulses with $\tau_G \sim 10^{-15}$s, $\Delta \omega \sim 10THz$. As rectification and electrooptic sampling are nonresonant effects, the minimum duration of the THz pulses that can be generated or detected is only limited by the thickness of the crystal scaled with the difference in phase and group velocity. Thus, with 10-15 fs exciting pulses at 800 nm, it was shown that T-rays could be generated with detectable frequencies as high as 37 and even 44 THz.

B. velocity(phase) matching

From (3), we know that the electric THz component gets maximized when its refractive index equals the group index of the generating optical pulse. After a walk-off length $l_w = c\tau_G/(n_{THz} - n_{opt})$, the optical pulse will lead the THz pulse by $\tau_G$. Newly generated THz will be totally out of phase with previous generated one so that only the THz wave generated in surface is emitted.

Unfortunately, the optical beam always travels faster than the THz beam so that the velocity matching is not achievable in many nonlinear optical materials like LiNbO$_3$. In laboratory, the velocity match is often accomplished with a ZnTe crystal, which has a strong absorption line at 3.7THz [8], and an optical pulse near 800nm [9]. The strong dispersion in the vicinity of an electronic transition in ZnSe makes the velocity match possible. Similar materials include GeSe [10] and DAST. Only in specific spectral range of pump pulse and the generated THz wave, the velocity matching is achievable.

A method called pulse front tilting is provided to compensate the velocity mismatch in other materials where angle and wavelength tuning are not useful.

THz generation via optical rectification can be accomplished...
Fig. 4. (a) Schematic diagram of the multiple THz pulse generation in PPLN. (b) THz waveform via optical rectification in PPLN.

in two different configurations characterized by slight and tight focusing of the fs laser beam. Slight focusing means that cross section of the beam is much larger than the extension of the pulse in the longitudinal direction while for tight focusing they are comparable.

The crucial feature of the novel scheme is the tilt of the pulse front of the light beam with respect to the phase front, which is perpendicular to the propagation direction of the light with $v_{opt}$. The THz radiation will propagate perpendicularly to the pulse front with $v_{THz}$. The projection velocity of the pump pulse to the direction of the THz propagation is $v_{opt} \cos \theta$. Obviously $\theta$ can be selected to be $\arccos(v_{THz}/v_{opt})$ to match this velocity $v_{THz}$. In case of tight focusing of the femtosecond pulse the THz electromagnetic wave is radiated into a Cerenkov cone with an angle of $\theta$ [11], velocity matching is achieved.

C. narrow-band THz wave generation

The single-cycle THz wave generated has broad bandwidth. In many cases, spectrally narrow and bright THz sources with frequency tuning are desired. Free electron laser produces the highest power and best coherence up to date, although a free electron laser is relatively more expensive compared to other schemes.

One of such technique has been demonstrated, using optical rectification of femtosecond pump pulses in periodically poled lithium niobate (PPLN) [12].

The technique utilizes the broad bandwidth of femtosecond optical pulses to satisfy a quasi-phase-matching condition enabled by the periodic structure. As illustrated in fig. 4(a), if the polarization domains of the PPLN reverse sign on a length scale $l_d \approx l_w$, then the polarization generated in the crystal will radiate a THz field consisting of N/2 cycles, where N is the number of domains over the length of the PPLN sample. If the domains are perfectly periodic, narrow band THz wave will be generated with a period $\Delta r_f = 2l_d(v_{THz} - n_{opt})/c$. The THz frequency can be varied by changing the domain length so that frequency tuning can be achieved. The is shown in fig. 5(a).

An example of the THz wave form is shown in fig. 4(b). A damped oscillatory wave form is observed at early times due to the imaginary part of the PPLN, followed by a more complicated beat pattern at later times. The bandwidth of the generated THz wave $\Delta \nu$ spectrum is predicted by $2\nu/N$ since it includes N/2 cycles. The time domain profile can also be calculated through integral of (3). These calculated results match the experiment results.

If the PPLN is manufactured in circular, the period of the domain can be tuned continuously by rotating the crystal. The pump beam passes through the center so that the beam direction stays constant during the rotation.

III. THz WAVE GENERATION VIA DFG AND TPO

One of the requirements for the generation of THz pulses by optical rectification is a fs laser source, which is still very expensive. In this section we discuss THz generation by difference frequency generation and parametric oscillation with ns laser pulses or even with normal cw lasers. Besides, THz generation by optical rectification has two obvious disadvantages: the broad linewidths and the low output power or pulse energies.

Continuously tunable and coherent radiation in the wide range has been achieved as a novel and promising terahertz source based on collinear phase-matched difference frequency
**Fig. 6.** Schematic of the 1064nm-pumped LiNbO$_3$ frequency THz-wave parametric oscillator.

THz-wave parametric oscillators, defined similar to OPO, are also reported [14] based on stimulated polarization scattering in a LiNbO$_3$ crystal pumped by a Q-switched Nd:YAG laser operated at a repetition rate of 50 Hz. A possible experimental setup is shown in fig. 6. The LiNbO$_3$ crystal was placed inside a Fabry-Perot cavity that consisted of mirrors M$_1$ and M$_2$, which reflected the idler wave at 1.07$\mu$m and let the pump beam pass. The THz wave exits the LiNbO$_3$ through the arrayed Si-prism coupler. The linewidth of the output THz wave is about 100GHz and the tuning rate is in the kHz range.

The advantages of TPO method include design simplicity and room temperature operation. The tunable range is from 1THz to 3THz, and TPO cavity must be rotated to meet the phase matching angle. This limits the tuning speed.

**IV. TECHNIQUES RELATED: THZ WAVE DETECTION AND SYNTHESIS**

The reverse of optical rectification is electrooptic sampling: A THz pulse incident on an electrooptic crystal such as ZnTe will induce a birefringence through the Pockels effect. An ultrafast visible probe pulse with a variable delay co-propagating through the same crystal will experience a retardation that can be retrieved with balanced detection. Scanning the relative time-delay of the probe pulse, one can record a time-domain trace of the electric-field of the THz pulse.

For many applications, the radiations in a variety of formats are required. A schematic diagram of an experimental setup for THz wave synthesis is shown in Fig. 7. A grating and a lens are used to image the spectrum of an ultrashort pulse onto a spatially varying mask, which can amplitude modulate or phase shift different components. A subsequent lens and grating are used to recombine the spectrally filtered components, producing a shaped waveform in the time domain. Since a spatial light modulator (SLM) can be used as a programmable mask, the optical waveform generation can be computer controlled, using adaptive feedback if necessary.

**V. CONCLUSION AND FUTURE WORK**

This paper has presented a general introduction of THz wave generation via nonlinear optical rectification. Most of the THz generation studies are carried in laboratory. The research concentrates on the several topics, like increasing the signal power, spectral tunability and extending the frequency range. As stated above, the basic limitation of THz generation by optical rectification is the velocity mismatch. Three different methods for the generation of THz radiation have been discussed. All of them have different advantages and disadvantages which are shown and listed in Table I in brief.

To date, all the THz nonlinear frequency mixing processes were done in bulk nonlinear optical materials. One way to greatly enhance the nonlinear frequency conversion efficiency is to perform parametric generation or oscillation in a nonlinear optical waveguide. The high efficiency results from tight confinement of mixing waves in a waveguide area. For LiNbO$_3$ crystals, the thickness is usually in the range of 500-1000 mm, forming a waveguide at THz wavelengths. The waveguide confinement for the THz radiation could greatly enhance the efficiency of the nonlinear frequency conversion.

Ideally, new generation THz sources should have the advantages of compactness, broad tunability, simple alignment, and stable THz output. Coherent THz emission based on intersubband transitions has not yet been implemented. There are many problems need to be solved.

**ACKNOWLEDGEMENT**

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TABLE I
ADVANTAGES AND DISADVANTAGES OF DIFFERENT NLO METHODS

<table>
<thead>
<tr>
<th>Generation by</th>
<th>Advantages</th>
<th>Disadvantages</th>
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<tbody>
<tr>
<td>optical rectification</td>
<td>- easy alignment</td>
<td>- fs laser needed</td>
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<tr>
<td></td>
<td>- waveform synthesis</td>
<td>- limited output power</td>
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<tr>
<td></td>
<td>- broadband THz spectrum</td>
<td>- difficult phase matching</td>
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<tr>
<td></td>
<td>- high time resolution</td>
<td>- broadband linewidths</td>
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<tr>
<td>difference frequency generation</td>
<td>- ns laser needed</td>
<td>- difficult alignment</td>
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<tr>
<td></td>
<td>- narrow linewidths</td>
<td>- two lasers needed</td>
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<td></td>
<td>- cw possible</td>
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<tr>
<td>parametric oscillation</td>
<td>- fast tuning possible</td>
<td>- limited output power</td>
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REFERENCES