Observation of THz to near-infrared parametric conversion in $ZnGeP_2$ crystal

Yujie J. Ding and Wei Shi

Department of Electrical and Computer Engineering
Lehigh University, Bethlehem, PA 18015, USA
yud2@lehigh.edu

Abstract: We observed a THz parametric conversion in which a near-infrared laser beam was mixing with a THz wave to generate a radiation in the near-infrared region (i.e. an upconverted signal) through difference-frequency generation phase-matched in a $ZnGeP_2$ crystal. By measuring the upconverted signal, we deduced the characteristics of the THz pulses participating in the frequency mixing. Specifically, we determined the range of the incident wavelengths, the minimum detectable energy per pulse, and the average linewidth of the THz pulses as 91-139 μm, 276 pJ, and 0.36 cm^{-1}, respectively.

OCIS Codes: (190.7220) Upconversion; (190.2620) Frequency conversion; (140.3070) Infrared and far-infrared lasers.

References and links

1. W. Shi and Y. J. Ding, "Chemical identification based on direct measurement of absorption spectrum by frequency-tuning monochromatic THz source," Laser Phys. Lett. **1**, 560-564 (2004).
2. W. Shi, Y. J. Ding, and P. G. Schunemann, "Coherent terahertz waves based on difference-frequency generation in an annealed zinc-germanium phosphide crystal: Improvements on tuning ranges and peak powers," Opt. Commun. **233**, 183-189 (2004).
3. See, e.g., H. R. Fetterman, P. E. Tannenwald, B. J. Clifton, C. D. Parker, W. D. Fitzgerld, and N. R. Erickson, "Far-ir heterodyne radiometric measurements with quasioptical Schottky diode mixers," Appl. Phys. Lett. **33**, 151-154 (1978).
4. J. E. Oswald, T. Koch, I. Mehdi, A. Pease, R. J. Dengler, T. H. Lee, D. A. Humphrey, M. Kim, P. H. Siegel, M. A. Frerking, and N. R. Erickson, "Planar diode solid-state receiver for 557 GHz with state-of-the-art performance," IEEE Microwave Guid. Wave Lett. **8**, 232-234 (1998).
5. W. Shi and Y. J. Ding, "A monochromatic and high-power THz source tunable in the ranges of 2.7-38.4 µm and 58.2-3540 µm for variety of potential applications," Appl. Phys. Lett. **84**, 1635-1637 (2004).
6. R. W. Boyd, *Nonlinear Optics* (Academic, New York, 2003).
7. W. Shi, Y. J. Ding, N. Fernelius, and F. K. Hopkins, "Observation of difference-frequency generation by mixing of terahertz and near-infrared laser beams in a GaSe crystal," Appl. Phys. Lett. **88**, 101101/1-3 (2006).
8. I. B. Zotova and Y. J. Ding, "Spectral measurements of two-photon absorption coefficients for CdSe and GaSe crystals," Appl. Opt. **40**, 6654-6658 (2001).

1. Introduction

Recently, it was demonstrated that a widely-tunable monochromatic (i.e. quasi-single frequency or narrow-linewidth) THz source could be used identify rotational transitions of molecules [1]. Compared with a typical mercury lamp used in Fourier-transform infrared spectroscopy (FTIR), such a tunable source produces much higher spectral intensities in the THz region. As a result, the sensitivity for chemical sensing can be enhanced by taking advantage of such a tunable THz source. Such a tunable source could be also used to detect chemical and biological agents, to identify bioaerosols, and to take images.

Recently, based on difference-frequency generation (DFG) in a $ZnGeP_2$ crystal THz pulses were generated by mixing two near-infrared laser beams [2]. These THz pulses were generated with the pulse durations of several nanoseconds and a repetition rate of 10 Hz. The

#71880 - $15.00 USD Received 15 June 2006; revised 12 August 2006; accepted 12 August 2006

tuning ranges of 83.1 – 1642 μm and 80.2 – 1416 μm were demonstrated for two different configurations phase-matched in a $ZnGeP_2$ crystal whereas the highest peak power was measured to be 134 W at 237 μm. The DFG scheme used for efficiently generating tunable THz radiations offer advantages such as high coherence (monochromatic output), simplicity for wavelength tuning, easy alignment, and stable output powers and wavelengths.

However, the practical applications by utilizing such a widely-tunable monochromatic THz source, mentioned by us above, have been impeded by a lack of a suitable detector or detection system operating at non-cryogenic temperatures. Indeed, one of the most sensitive and common THz detectors available so far is a bolometer. However, such a detector must be cooled to 4 K or lower to significantly reduce the thermal noise. Although Schottky diodes can be used to directly measure the THz radiation at room temperature [3], these detectors are not sensitive enough at the THz frequencies beyond 900 GHz. Although these diodes can be used as photomixers [4], the incoming THz radiation needs to be modulated at a frequency much higher than 10 Hz in order to avoid high noise levels. Therefore, these detectors cannot be used to measure THz pulses with a low repetition rate. On the other hand, Keating energy meters are not sensitive enough to measure the energies per pulse produced from the parametric conversion. This is due to the fact that these energy meters have a typical noise equivalent to 1 μJ. However, for the peak power of 134 W produced by based on DFG [2], the corresponding energy per pulse is about 0.67 μJ. Furthermore, both the bolometers and Keating energy meters have suffered from rather slow responses. For example, the bandwidth for a bolometer is typically limited to 30 kHz (a response time of 15 μs). Due to such a slow response, a bolometer or Keating energy meter usually picks up a large amount of the noises integrated within an interval between the short THz pulses. Obviously a new scheme for the THz detection must be introduced and investigated in order to detect an ns-long THz pulse at a low repletion rate. A new THz detection system to be implemented must have a high sensitivity, a wide bandwidth, and an ability of operating at non-cryogenic temperatures with relatively low noises.

In this contribution, we report our results based on the observation of the frequency mixing of THz radiations with laser pulses in the near-infrared region, which is phase-matched in a $ZnGeP_2$ crystal. We demonstrate that such a parametric process can be used to measure sub-nJ energies per pulse, wavelengths, and linewidths of the THz pulses. As one can see below, using such a scheme the detection range for the THz waves is limited by the band-pass filter used to block the near-infrared Nd:YAG laser pulses. It is worth noting that the $ZnGeP_2$ crystal used in our experiment was annealed. As a result of annealing, the absorption coefficient at 1 μm was significantly reduced to 0.75 cm^{-1} [2]. However, if the $ZnGeP_2$ crystal was not annealed, the absorption coefficient at this wavelength would be as high as 5.63 cm^{-1} [2], which is too high for achieving a relatively efficient conversion from the THz pulses to the near-infrared radiation.

2. Experimental

In our experiment, we mixed Nd:YAG laser pulses emitting at the wavelength of 1.064 μm with THz radiations in a $ZnGeP_2$ crystal. Such a DFG process was phase-matched based on the *oe-e* polarization configuration where *oe* designate the polarizations of the 1.064-μm and the THz beams, respectively, and *e* denotes the polarization of the output, i.e. the upconverted signal with its wavelength in the near-infrared region. The energy of each Nd:YAG laser pulse can reach several mJ and the laser beam diameter is about 350 μm. On the other hand, the incoming THz pulses were generated by mixing two coherent near-IR radiation beams in a GaSe crystal [5]. These THz pulses were focused onto a $ZnGeP_2$ crystal by using a parabolic mirror. This $ZnGeP_2$ crystal cut at the angle of $\theta \approx 0°$ was 21 mm long. After the mixing of the THz and Nd:YAG laser pulses in the $ZnGeP_2$ crystal, the output pulses with their wavelength in the near-infrared domain, i.e. the upconverted signal, were generated. A polarizer and a band-pass filter were both inserted in the beam pass to remove the un-converted Nd:YAG laser pulses. The band-pass filter, manufactured by Semrock, typically has a transition width of < 93 cm^{-1} and a passband of 1077.8-2000 nm. The upconverted signal then passed through a monochromator, manufactured by Spectral Products, which has a maximum spectral resolution of 0.06 nm. The upconverted signal was finally measured by using an InGaAs detector or a TE-cooled photomultiplier tube.

3. Results and discussion

Figure 1 illustrates our result following the peak power measured by the detector vs. the THz wavelength. One can see from Fig. 1 that the wavelengths of the incident THz pulses measured by us had covered the range of 91-139 μm. This detection range was primarily limited by the band-pass filter used in our experiment. Such a filter was necessary in order to eliminate the un-converted Nd:YAG laser pulses. However, when the wavelengths of the upconverted signal and the Nd:YAG laser pulses were too close to each other, the upconverted signal was significantly attenuated, which set the long-wavelength limit of the detection range. On the other hand, on the short-wavelength side the peak power of the THz pulses participating in the frequency mixing determined the limit. Within the detected range of the THz wavelengths, the parametric conversion from the THz radiations to near-infrared pulses was phase-matched. This was due to the fact that the phase-matching condition for such parametric conversion was exactly the same as the THz generation through the frequency mixing of two near-infrared coherent beams in the $ZnGeP_2$ crystal [2].

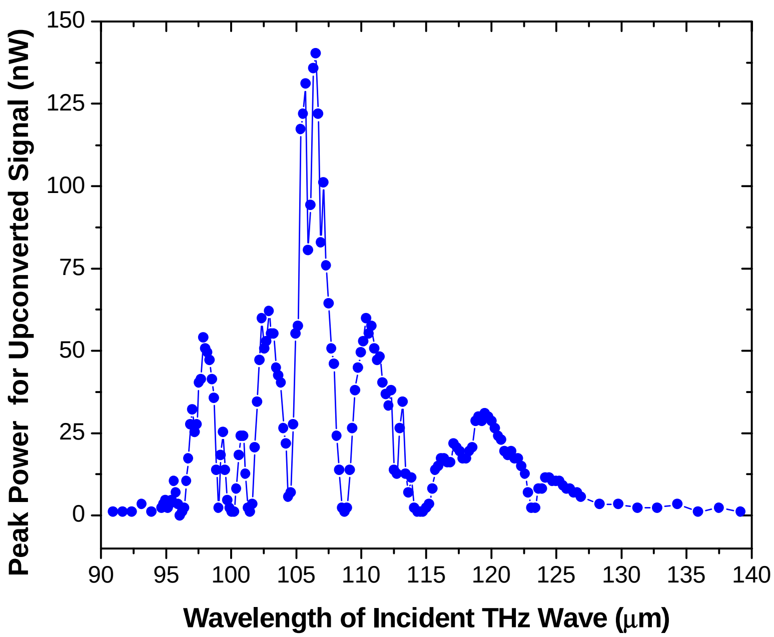

Fig. 1. Peak power for the upconverted signal measure by using an InGaAs photodiode vs. wavelength of the incident THz pulses after mixing them with Nd:YAG laser pulses. Modulation appearing in the spectrum was caused by the absorption of water vapor in the beam path.

We would like to point out that the modulations appearing in Fig. 1 were due to the absorption of the water vapor present in the beam pass. Figure 2 illustrates our result for the peak power of the upconverted signal measured by using an InGaAs photodiode vs. the incident THz power at an incident wavelength of 106 μm. One can see from Fig. 2 that the

#71880 - $15.00 USD Received 15 June 2006; revised 12 August 2006; accepted 12 August 2006

dependence of the output power on the incident THz power can be more or less fitted by a straight line. Such a behavior is consistent with the theory of DFG under the assumption of no depletion since under such a condition the power of the upconverted signal is proportional to the product of the input powers for the Nd:YAG laser and THz pulses [6]. According to our measurement, the lowest energy per pulse was 276 pJ. Such a pulse energy is translated into ≈ $0.87\ \mathrm{nW}/\sqrt{\mathrm{Hz}}$ **.** This value is three-to-four orders of magnitude more sensitive than that for a typical pyroelectric joulemeter made from lithium tantalate or Keating energy meter (≈ 5 $\mu\mathrm{W}/\sqrt{\mathrm{Hz}}$). Based on the electro-optic detection scheme one can measure a THz field with a very low magnitude. However, such a scheme is not suitable to the measurement of the electric fields of the ns THz pulses which have very narrow linewidths compared with those of the ultrafast THz pulses.

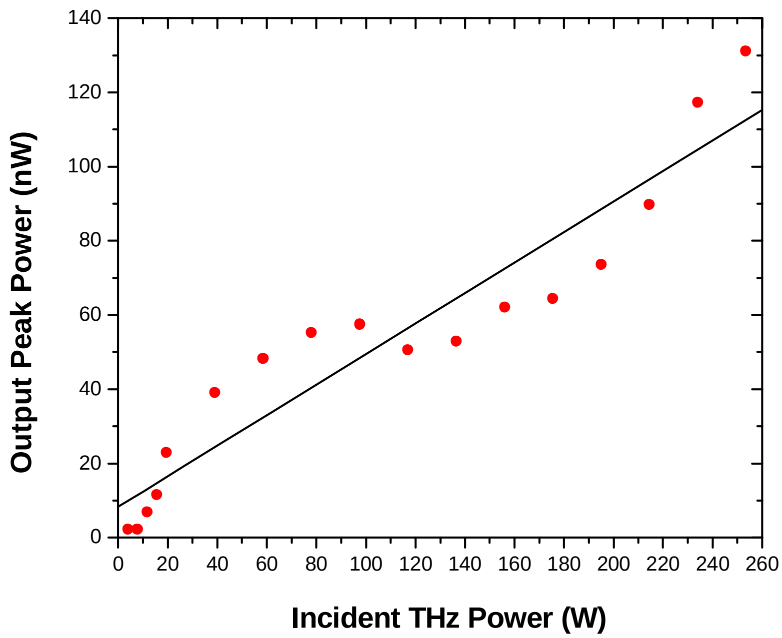

Fig. 2. Output power for the upconverted signal measured by using an InGaAs detector vs. incident THz power. Dots – data; solid line – linear fit.

Using a TE-cooled photomultiplier tube we also measured the spectra of the upconverted signals for different central frequencies of the incident THz pulses by using an infrared spectrometer. Based on the conservation of the photon energies, we then determined the corresponding frequencies across the spectrum of the THz pulses for each fixed central frequency. As a result, we constructed the spectra of the incident THz pulses participating in the frequency mixing, see Fig. 3. As we decreased the width of the entrance and exit slits for the spectrometer, we measured the linewidths of the incident THz pulses, see Fig. 4. As one can see from Fig. 4, the ranges of the linewdiths for the two lowest slit widths, measured in our experiment, significantly overlapped with each other. Therefore, we determined the average linewidths of the incoming THz pulses to be as narrow as 11 GHz (0.36 cm^{-1}). This value is quite close to the linewidth of one of the coherent near-infrared beams used for the THz generation, i.e. 0.2 cm^{-1} [5].

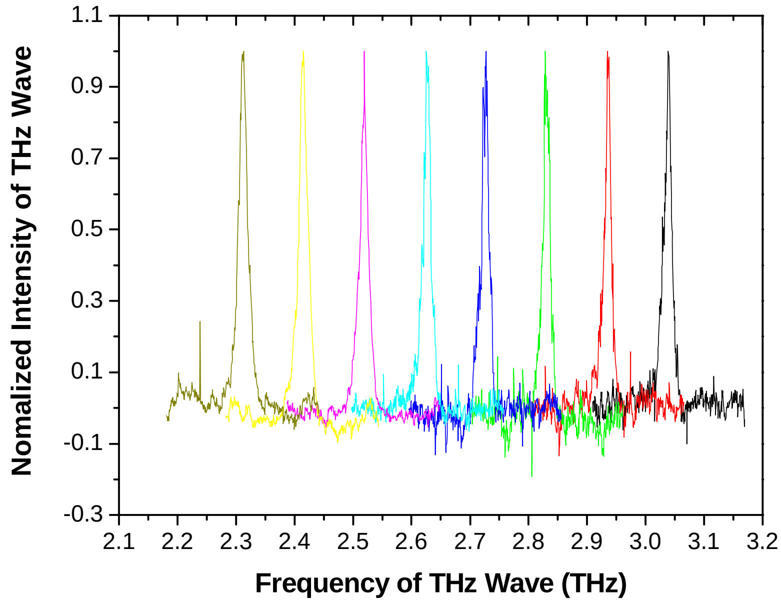

Fig. 3. Spectra of THz pulses measured by a TE-cooled photomultiplier tube at different central frequencies by using the frequency mixing process.

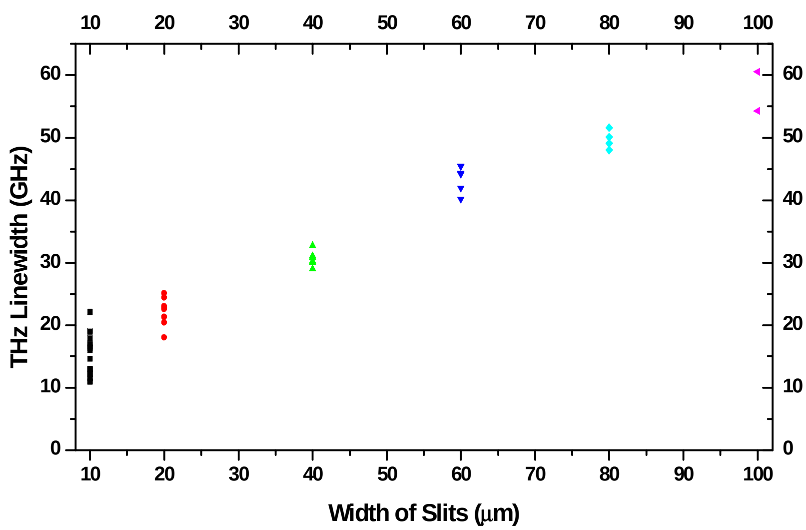

Fig. 4. Linewidth of THz pulses were measured by a TE-cooled photomultiplier tube vs. slit width of spectrometer. At each slit width, linewidth was measured for several times. It is obvious that for the two lowest slit widths the ranges of the linewidths for the THz pulses measured by us significantly overlapped with each other.

Previously [7], we observed the upconversion by using a GaSe crystal. Compared with the results obtained using the two different crystals, the noise equivalent power for the detection of THz pulses using the $ZnGeP_2$ crystal is slightly higher (276 pJ vs. 245 pJ). Also, the highest power for the upconverted beam occurred at the incident wavelength of 106.5 µm for

Received 15 June 2006; revised 12 August 2006; accepted 12 August 2006

the $ZnGeP_2$ crystal, see Fig. 1. On the other hand, for the GaSe crystal [7] the corresponding wavelength was 92.4 µm. Such a difference can be attributed to the different dependences of the effective nonlinear coefficients on the phase-matching angles for two different crystals. Besides these differences, we measured the spectra of the THz pulses using the $ZnGeP_2$ crystal and then deduced the linewidths of the incoming THz pulses. In general, the optimum conversion efficiency for the upconversion depends on the absorption coefficient of the THz pulses. For example, at 200 µm the absorption coefficients are about 0.37 cm^{-1} and 0.14 cm^{-1} for the $ZnGeP_2$ and GaSe crystals, respectively. If the incoming radiation in the near-infrared region used for upconverting THz pulses to near-infrared radiation has relatively high intensities, a GaSe crystal may be preferred since its two-photon absorption coefficient is much lower in the near-infrared region [8]. However, unlike the GaSe crystal, the $ZnGeP_2$ crystal can be mechanically polished. In addition, the polished surface of such a crystal can be covered by anti-reflection coatings such that the reflection for the near-infrared incident radiation can be almost eliminated. Furthermore, a Bragg reflector and perhaps even a band-pass filter can be eventually fabricated directly inside this crystal such that the conversion efficiency for the upconversion can be significantly increased. On the other hand, since a GaSe crystal is rather soft, it would not be possible for us to incorporate these structures into this crystal.

4. Conclusion

In conclusion, by mixing the THz radiations with the near-infrared laser pulses in a $ZnGeP_2$ crystal, we have observed the phase-matched THz frequency upconversion, i.e. DFG. Through such parametric conversion, we have determined the energies per pulse, wavelengths, and linewidths of the ns THz pulses having a repetition rate of 10 Hz, at room or a non-cryogenic temperature. The lowest energy per pulse, average linewidth, and detectable wavelength range for the THz pulses were measured by us to be 276 pJ, 0.36 cm^{-1}, and 91-139 µm, respectively. Based on the experimental results presented here, we believe that such a frequency-mixing process could be eventually developed into an effective technique for measuring short THz pulses with an ns temporal resolution. In order to use the upconversion process as a practical method for the detection of THz pulses, further work must be carried out such as the optimization of the conversion efficiency and the incorporation of waveguide structures.

Acknowledgment

This work has been supported by U.S. AFOSR.

#71880 - $15.00 USD Received 15 June 2006; revised 12 August 2006; accepted 12 August 2006