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Efficient THz generation and frequency upconversion in GaP crystals

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Abstract

We have developed a widely-tunable monochromatic THz source based on phase-matched difference-frequency generation (DFG) in a GaP crystal. By using a 20-mm-long GaP crystal the output wavelength was tuned in the range of 71.1–2830 μm whereas the highest peak power was 15.6 W. The advantage of using GaP over birefringent non-linear-optical crystals is obvious: crystal rotation is no longer required for wavelength tuning. Instead, one just needs to tune the wavelength of one mixing beam within a bandwidth of as narrow as 15.3 nm. Most recently, we observed THz frequency upconversion by mixing a THz wave with an infrared laser beam in another GaP crystal. The lowest energy per pulse measured by us was 780 pJ whereas the wavelength range of the detectable THz waves was 113.6–128.3 μm. This scheme allows us to measure the pulse energy density, wavelength, linewidth, and pulse width of a THz beam at non-cryogenic temperatures. We have also discussed a few mechanisms for further enhancing the conversion efficiencies of the two parametric processes.

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1. Introduction

Since the frequency spacings among rotational states of gases are within the THz region [1], a relatively compact THz spectrometer can be used to fingerprint molecular gases. Because the linewidth for the rotational transitions can be 50 MHz (0.0017 cm^{-1}) or narrower for low-pressure gases, such a spectrometer must have a high spectral resolution in order to resolve the spectrum of each rotational transition. In the past, researchers believed that it might be possible for them to develop the spectrometers working in the domains from visible to mid-infrared for fingerprinting molecules. Unfortunately, most molecules in those domains exhibit the congested and unresolved ro-vibrational spectra [2]. Therefore, truly fingerprinting molecules may not be feasible in the regions from the visible to the mid-IR. On the other hand, bio-medical and bio-chemical sensing has recently become increasingly important. However, these applications have been impeded by the strong absorption of the water present in the bio samples as well as the scattering of THz waves by the bio samples. When bio particles become airborne, THz waves could be still used to fingerprint the airborne bio particles. Since almost all the aerosol particles have diameters $\leqslant 10$ μm [3], the scattering of the THz waves by the bio-aerosol particles suspended in the air could be greatly reduced. Due to the presence of the atmospheric windows (mainly due to the absorption dips of the water vapor [4]), THz waves could be eventually used for probing as well as imaging. Based on our analysis made above, a THz system is mostly needed. It should be capable of producing the images of

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targets and simultaneously analyzing the targets as well as the chemical vapors present in the beam path with ultra-high spectral resolutions.

In the past, THz pulses were generated by optical rectifications [5], photoconduction [6], and Cherenkov radiation [7]. For example, based on optical rectification THz waves were generated in GaAs and CdTe crystals [8]. Moreover, by utilizing electro-optic effect, GaP [9] and ZnTe [10] crystals were used to detect THz pulses. It is worth noting that when the phase-matching condition was satisfied the bandwidth for the THz output could be significantly widened [10]. These schemes all require an ultrafast laser as the pump. In order to detect chemicals, THz time-domain spectroscopy was developed [11]. Such a technique can be used, e.g., for combustion diagnostics [12].

Another route to construct a THz system is to take advantage of the recent development on a current-injection coherent THz emitter based on semiconductor heterostructures. Since the first report of such an emitter based on GaAs/AlGaAs superlattices [13], several groups have been working on such a type of the emitters. However, this approach has its fundamental limit. Due to the thermal population of the carriers, such emitters with the output frequencies in the range of 1–3 THz, narrow linewidths ($\leqslant$50 MHz), and reasonably high powers (several mW) cannot be operated at the non-cryogenic temperatures. Furthermore, it is quite challenging for anyone to develop a tunable THz emitter based on the semiconductor heterostructures. Due to the relatively low frequencies of the THz radiation, it may be even more challenging to develop the sensitive room-temperature THz detectors based on semiconductor heterostructures. Although Schottky diodes can be used to measure THz radiation at room temperature [14], most Schottky diodes have low responsivities near 1 THz and above it. On the other hand, a Schottky mixer requires the presence of a rather clean local oscillator near the frequency to be measured. By mixing the THz signal with the local-oscillator output in the mixer one can then measure the beat frequency, and therefore, determine the signal frequency in turn. Obviously, it would be quite challenging to measure the output of a tunable THz source using such a method.

Recently, by mixing two coherent near-infrared laser beams in GaSe [15–17] and $ZnGeP_2$ [18–20] crystals, we efficiently generated the THz waves with extremely-wide tuning ranges and very high peak powers as well as high conversion efficiencies. The widest tuning range achieved by us is from 66.5 μm to 5.66 mm (i.e. from 4.51 THz down to 53 GHz) based on the type-*oe–e* phase-matched difference-frequency generation (DFG). On the high-frequency side of the reststrahlen band, we were able to generate the radiation tunable in the range of 2.7–28.7 μm using a different phase-matching configuration [21]. One can see that the tuning range achieved by us almost covers the entire THz region (0.1–10 THz) and part of the microwave or millimeter-wave domain, i.e. from 1 mm to 5.66 mm (53–300 GHz). Before this result was reported, such a wide tuning range was only produced by bulky free-electron lasers. The highest output peak power measured by us was 389 W at 203 μm (1.48 THz) and 4.7 W at the wavelength of 1 mm (300 GHz), corresponding to the conversion efficiencies of 0.098% and 0.0012% in terms of the pulse energies, respectively.

By using our widely-tunable THz sources we have explored a few applications. In particular, by measuring the absorption spectra of three families of the commonly-used chemicals in the vapor phase, we could fingerprint certain molecules [22]. We also measured the absorption spectra of DNA and proteins [16]. Moreover, we characterized the Bragg reflectors and 2-D photonic crystals fabricated on Si wafers [23].

In this article, we summarize our previous results on the efficient generation of the widely-tunable monochromatic THz waves by mixing two near-IR laser beams with slightly different frequencies in a GaP crystal. In addition, we present our new result on the observation of THz frequency upconversion in another GaP crystal. Furthermore, we discuss a few schemes for further improving the conversion efficiencies of the two THz parametric processes.

2. THz generation in GaP crystal

As discussed above, widely-tunable monochromatic THz waves were efficiently generated by mixing two coherent infrared beams in GaSe and $ZnGeP_2$ crystals. Birefringence for these non-linear crystals was absolutely necessary in order to achieve phase-matching for DFG. In order to tune the output wavelength one must rotate each of these crystals. In general, in order to achieve the efficient conversion for the DFG the non-linear coefficients for non-linear crystals must be large. Furthermore, non-linear crystals must have low absorption coefficients in the THz region. Besides GaSe and $ZnGeP_2$, zincblende crystals such as GaAs and GaP also satisfy these two requirements [24]. Even though they do not possess any birefringence, phase-matching for the THz conversion can be still satisfied due to the presence of the reststrahlen bands in the THz region [24,25]. According to the calculations [24,25] GaAs and GaP could be used to generate tunable THz beams by tuning the mixing wavelengths in the ranges of 1.247–1.333 μm and 0.9958–1.034 μm, respectively.

A GaP crystal was previously used to achieve upconversion, i.e. the generation of a visible output by mixing a visible input with each of five far-infrared waves resonant to the broad reststrahlen band of the crystal [26]. Recently, this crystal was also used for generating the tunable THz beam based on non-collinear phase-matched DFG [27]. As a result, a tuning range of 42.8–600 μm and a peak power of 480 mW were obtained. Such a tuning range was achieved by changing the incident angle formed between the propagation directions of the two mixing beams. However, under such a configuration the spatial overlap between the THz beam and the two mixing beams inside the crystal was sacrificed due to the large angle