Experimental study on terahertz radiation

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In this letter, we describe a coherent subpicosecond terahertz (THz) spectroscopy system based on non-resonant optical rectification for the generation of THz radiation. We studied the two-photon absorption (TPA) of ZnTe induced by femtosecond laser pulses via THz generation, and its influence on the generation of THz radiation. Experimental results demonstrated that the intensity of pump beam against TPA must be traded off to get an optimum generation of THz radiation. As an example, we measured absorption spectrum of water vapor by time-domain spectroscopy (TDS) in the frequency range from 0.5 to 2.5 THz with a high overall accuracy.

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The recent increasing interest in the generation and detection of coherent subpicosecond electromagnetic pulses at terahertz (THz) frequency (0.1−tens of THz) has enabled a wide range of applications[1,2] in the long inaccessible frequency region. This electromagnetic frequency range is very attractive for scientists in various fields because the characteristic energy of many fundamental interactions in molecules and condensed matter occurs in this region. Recently, there has been also increasing interest in THz microstructured devices[3]. THz time-domain spectroscopy (TDS) can be used to directly measure amplitude and phase of the THz radiation pulse. This technique has been utilized recently to investigate the wavelength dependence of complex refractive index in semiconductors, superconductors, and dielectrics through transmitted and reflected light. The capability to measure the complex dielectric constant of materials is invaluable in condensed-matter physics. And in the future, THz-TDS will become a powerful technique for illustration and analysis of ultrafast phenomena in basic sciences such as physics, chemistry, and biology. Now, more and more people have begun to pay attention to the investigation of THz radiation and our group has detected the THz radiation waveform and its frequency spectrum[4]. We are also investigating possible applications in industrial fields such as non-destructive materials inspection.

THz generation via optical rectification can be accomplished in two different configurations characterized by slight[5] and tight[6] focusing of femtosecond laser beams. Slight focusing means that the beam size in the emitter is much larger than the extension of the pulse in the longitudinal direction while for tight focusing they are comparable. When the laser beam is slightly focused, the velocity mismatch reduces the effective thickness of the emitter contributing to THz generation. In this situation the THz radiation originates from two thin layers near the front and back surfaces of the emitter. In the case of tight focusing, the smaller beam waist of the pump, and thus the increased intensity result in high conversion efficiency of THz radiation. But the increasing intensity of the pump beam will induce nonlinearity in the emitter, such as two-photon absorption (TPA) and Kerr effect, which may influence the generation of THz radiation. Caumes *et al.*[7] have discussed Kerr-like nonlinearity induced by THz generation in zinc blende crystals. In this letter, we describe a coherent subpicosecond THz spectroscopy system based on non-resonant optical rectification for the generation of THz radiation. On the setup of THz radiation, we have experimentally studied the influence of the TPA and Kerr effect induced by pump laser pulses on the generation of THz radiation. The experimental results demonstrated that to get an optimum generation of THz radiation, one has to trade off the intensity of pump beam against TPA. As an example, we measured absorption spectrum of water vapor by TDS in the frequency range from 0.5 to 2.5 THz with overall accuracy as high as reported in literature[8].

In a wideband coherent THz radiation system using ZnTe crystals, the generation of THz radiation is based on optical rectification, and THz radiation detection is via electro-optic effects in ZnTe. Optical rectification allows to produce much broader THz bands[6], and the simplicity of the experimental realization is another advantage. Figure 1 schematically illustrates the experimental setup of THz radiation. A homemade self-mode-locked Ti:sapphire laser[9] delivered 40-fs pulses with an

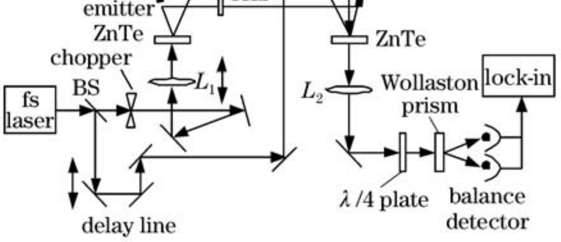

Fig. 1. Schematic drawing of the experimental setup.

average power of 300 mW at 810 nm and repetition rate of 100 MHz. The linearly polarized femtosecond laser beam with polarization direction along the table plane was split into two parts, in which 70% was used as the pump beam, and the remains 30% was used as the probe beam. The pump beam chopped into 1050 Hz was focused by lens L_1 ($f = 5$ cm) on the ZnTe emitter. Lens L_1 was mounted on a translation stage, which was carefully set so that it could be moved along the pump beam direction. Two 1-mm-thick ⟨110⟩ ZnTe crystals were used as the emitter and the detector of the THz radiation, respectively. The polarization of both the THz and the optical probe beams were aligned parallel to the [1, −1, 0] direction of the ZnTe detector. The THz radiation was collimated with two off-axis parabolic mirrors and focused on the detector for electro-optic sampling of the THz radiation. A teflon filter was used to remove the residual pump beams. The probe beam modulated by THz radiation was collimated with lens L_2 and optically biased at a quarter-wave plate, which allow the system to be operated in the linear range. A Wollaston polarizer was used to convert the THz field induced phase retardation of the probe beam into an intensity modulation between the mutually orthogonal linearly polarized beams. A balanced photoelectric detector was used to detect the optical intensity modulation, which was wired to give zero current for balanced power. The signal current was sent to a lock-in amplifier (Sanford Research system SR830) for amplifying the signal and filtering noise. The optical delay line worked through a computer-controlled translation stage (Physikinstrumente, M-405.DG) with a resolution of 0.1 μm. Figure 2 is the temporal waveform of the THz radiation pulses. The two traces are for two measurements under the same operation conditions. Excellent overlap of the two traces indicates that our experimental system possesses very good reproducibility. Figure 3 shows the THz temporal signal and the corresponding frequency spectrum (inset) obtained from the ZnTe emitter beyond focus.

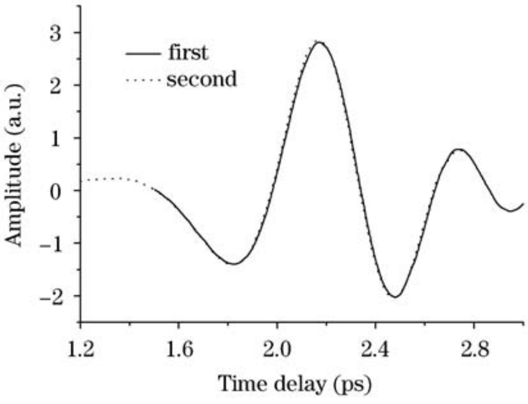

Fig. 2. Excellently overlapped temporal waveforms of the THz radiation pulses.

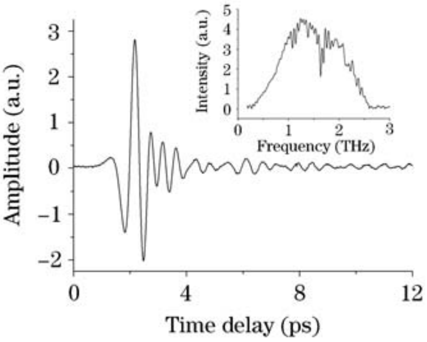

Fig. 3. Typical THz waveform and its frequency spectrum.

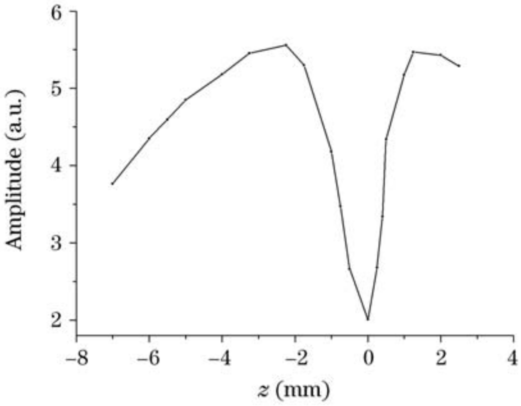

Fig. 4. Peak amplitude of THz radiation versus position z of ZnTe emitter.

The ZnTe crystal exhibits strong Kerr effect and TPA for femtosecond laser pulse at 810 nm[10]. In the case of smaller pump beam size in the ZnTe crystal, the stronger pump beam intensity results in the stronger TPA and thus more largely reduces the THz emission intensity. Figure 4 shows that the peak amplitude of THz radiation pulses decreases with the ZnTe crystal closing to focal point of the pump beam. In the experiments, we changed the pump beam size in the ZnTe crystal by moving focusing lens L_1 along the pump beam axis and keeping a constant distance between the collimating mirror and the ZnTe emitter crystal. The zero position of the ZnTe along the pump beam is defined as the focal point of pump beam at which THz radiation is minimum. The position is negative when ZnTe is closer to the focusing lens L_1 from the focal point of the pump beam, and the position is positive when ZnTe is farther away from L_1. The shape in Fig. 4 is similar to the TPA feature of ZnTe measured by Z-scan technique shown in Ref. [10]. The slope of falling edge of the valley is slower than that of the rising edge. We also can see that the peak amplitude of THz radiation is going down when the position of emitter is far away from the focus plane. Therefore there is an optimum position of THz emitter relative to the focal point of the pump beam for high efficiency of THz generation. Figure 5 shows the frequency spectra of THz radiation at three different positions ($z = 0$, $z = 1.25$ mm, $z = -2.25$ mm) of the emitter. All the frequency spectra are almost the same expect for relatively tiny shift from each other. It is clearly found that the frequency spectrum of THz radiation has a blue shift corresponding to strong TPA in ZnTe, which maybe arise from Kerr effect in ZeTe emitter induced by the pump beam and a finite aperture of the off-axis parabolic mirrors which cut off a part of long wavelength components of THz radiation.

As an application example of our THz system to TDS, we measured the absorption spectrum of water vapor by testing THz radiation pulse signal through water vapor.

Table 1. Absorption Spectrum of Water Vapor (Unit: THz)

D1	1.09922	1.1156964	1.16257	1.20938	1.23055	1.41094	1.60323	1.67109
D2	1.09367	1.1256175	1.16686	1.20078	1.23272	1.41312	1.60480	1.66869
D1	1.71808	1.79766	1.87031	1.91965	2.04147	2.16563	2.19860	2.25703
D2	1.72131	1.79272	1.86788	1.92801	2.04452	2.16479	2.19861	2.26251

*D1: absorption spectrum of water vapor in Ref. [8], D2: in our experiments.

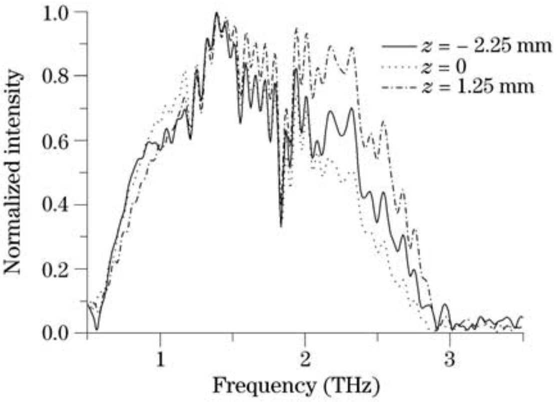

Fig. 5. Frequency spectra of THz radiation at different positions of ZnTe.

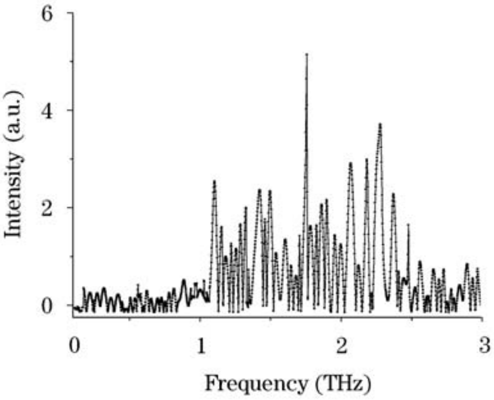

Fig. 6. Absorption spectrum of the water vapor.

The experiments were performed as follows. First, we detected THz radiation pulses as reference background after propagating only through atmosphere in the lab. And then by putting a cup of hot water under the THz radiation beam between two collimating mirrors, we measured THz radiation pulses through water vapor. The amplitude absorption spectra of the atmosphere and the water vapor are given from Fourier transform corresponding to the THz time-domain signals. By subtracting the amplitude absorption spectrum of the atmosphere from that of sample with water vapor, the amplitude absorption spectrum of water vapor in the frequency range from 0.5 to 2.5 THz is shown in Fig. 6, which is as accurate as reported in the Ref. [8]. The data in Table 1 indicate the comparison of the water vapor spectrum in our experiments with that in Ref. [8].

We built a THz-TDS system, on which we studied the TPA in the ZnTe emitter crystal induced by pump source via THz radiation generation and its influence on the generation efficiency of THz radiation. Experimental results demonstrated that we have to trade off the intensity of pump source in the ZnTe emitter crystal against TPA to get an optimum generation of THz radiation. As an application example of our system to TDS, we measured the amplitude absorption spectrum of water vapor in the frequency range from 0.5 to 2.5 THz. The measured spectrum data showed that our system possesses a high measuring accuracy.

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