Free-space electro-optics sampling of mid-infrared pulses

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(Received 16 May 1997; accepted for publication 7 July 1997)

We report on the coherent detection of ultra-broadband mid-infrared electromagnetic pulses using a 30-μm-thick ZnTe electro-optic sensor. The detected frequency spectrum exceeds 37 THz, extending from microwave to the mid-infrared. The frequency response can be further improved by reducing the sensor thickness to 10 μm. © *1997 American Institute of Physics.*
[S0003-6951(97)02536-9]

Despite the rapid advance of terahertz optoelectronics in the recent decade, the frequency region above 10 THz has yet been accessible for coherent detection. Whereas Katzenellenbogen *et al.* reported a frequency response of 6 THz from their photoconducting dipole antenna (PDA),[1] the record was recently pushed to 7 THz with a much broader 3-dB bandwidth[2] in a GaP-based free-space electro-optic sampling (FS-EOS) system. However, because of the Reststrahlen region near 11 THz, a GaP sensor is unlikely to reach 10 THz. This is generally the case for most electro-optic (EO) sensor materials since the frequency of the fundamental TO phonon resonance in GaP is one of the highest known of many semiconductor EO crystals.

The possibility of extending FS-EOS into the mid-infrared region is nevertheless favored by the following factors. First, most semiconductor EO materials are transparent in the frequency region between phonon resonance (far-IR) and the electronic resonance (near-IR); Second, it is reasonable to assume an instantaneous nonlinear response and a constant electro-optic coefficient across the mid-infrared region for covalent semiconductor EO crystals, where the dominant nonlinearity is electronic in origin; And the last, 10 fs lasers, which now proliferates rapidly, can readily provide a sampling bandwidth (FWHM) of as high as 44 THz.

The generation of broadband mid-infrared radiation through optical rectification was recently demonstrated by Bonvalet *et al.*[3] In their experiment, an incoherent detector (HgCdTe) and interferometric technique were employed to characterize the mid-infrared pulses. The spectrum of the generated radiation, as they noted, spanned beyond the detection bandwidth of the HgCdTe detector. Furthermore, due to the dispersion of GaAs, which has an index of $n(5\ \mu\mathrm{m}) = 3.2978$ and $n(25\ \mu\mathrm{m}) = 3.058$,[4] the spectral phase of the mid-infrared pulse in fact cannot be assumed constant even for their 100-μm-thick GaAs emitter. Incoherent detector based linear correlation technique was therefore unable to provide an accurate characterization of the ultra-broadband radiation. FS-EOS, as a coherent detection technique, becomes attractive when combined with an ultra-broadband mid-infrared source.

In this letter, we report the use of electro-optic sensors for coherent characterization of mid-infrared terahertz beams with a bandwidth up to 37 THz. This represents a significant improvement over the previous record of 7 THz,[2] and also confirms the broadband nature of the radiation generated by optical rectification.

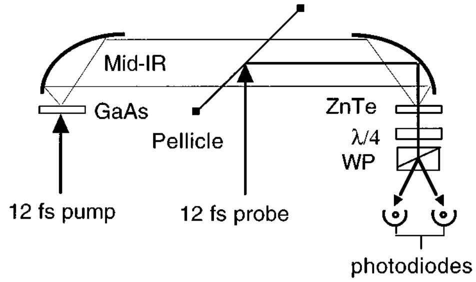

FIG. 1. Schematic of experimental setup. λ/4: achromatic quarter wave plate; WP: Wollaston prism.

Figure 1 schematically illustrates the experimental setup of the FS-EOS detection system. The 12 fs Ti:sapphire laser (Kapteyn-Murnane Labs) delivered an average power of nearly 500 mW at a center wavelength of 800 nm. A 0.45-mm-thick ⟨110⟩-oriented GaAs was used as an emitter and a 30-μm-thick ⟨110⟩ ZnTe crystal as an EO sensor. 350 mW of the laser power was focused on the GaAs emitter by a gold-coated off-axis parabolic mirror with a 5 cm effective focal length. The broadband THz radiation generated from the GaAs emitter by optical rectification[3] was collimated and then focused on the ZnTe EO sensor by a pair of $f/0.6$ off-axis parabolic mirrors. The laser probe beam was combined to collinearly travel with the THz beam through a 2-μm-thick pellicle, which had a negligible effect on laser pulse width and THz beam. The EO modulation induced by the ultrafast Pockels effect was detected by using a pair of bal-

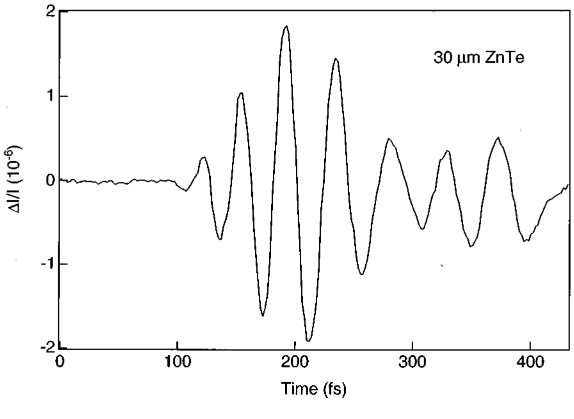

FIG. 2. Temporal waveform of the THz radiation measured by a 30 μm ZnTe sensor. The shortest oscillation period is 31 fs.

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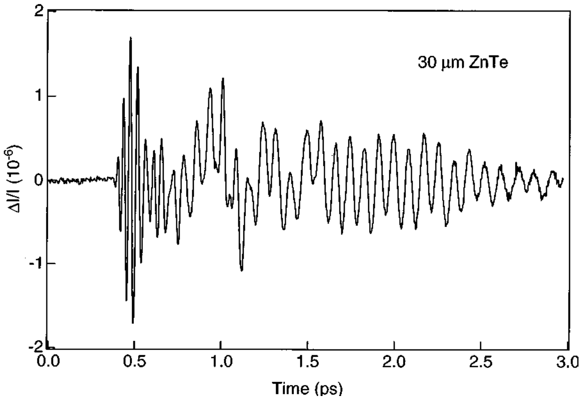

FIG. 3. Temporal waveform of the THz radiation with a long scan.

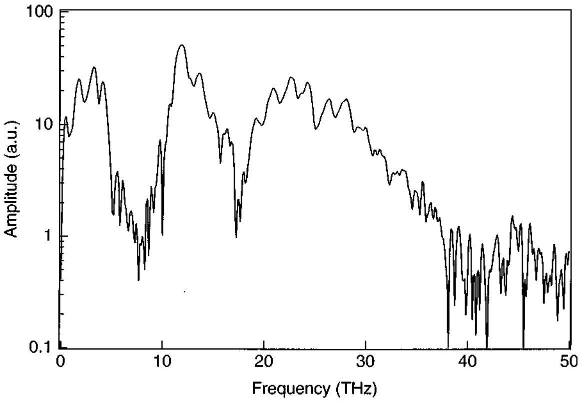

FIG. 4. Amplitude spectrum of the waveform in Fig. 3.

anced photodiodes, where a shot-noise limited detection sensitivity of photomodulation ($\Delta I/I$) as small as 2×10^{-8} Hz$^{-1/2}$ was routinely achieved. By varying the time delay between the optical pump and probe pulses, the temporal waveform of the mid-infrared transient was sampled.

Figure 2 shows a typical waveform obtained in a single scan with a 300 ms lock-in time constant. The shortest period of the oscillation is 31 fs. Figure 3 shows a long scan and the Fourier transform of the waveform is shown in Fig. 4. The spectral modulation period of about 2 THz in Fig. 4 is due to the multiple reflection of the THz field in the ZnTe. The modulation period in turn can be used to measure the thickness of the sensor crystal. The highest frequency response reaches 37.3 THz, which corresponds to a wavelength of 8 μm. The absorption gap between 5 and 10 THz is due to the Reststrahlen bands of ZnTe and GaAs. The dip around 17 THz in the spectrum can be understood in terms of the response cutoff due to velocity mismatching. The index difference between optical group index and mid-infrared index in ZnTe is 0.6, which corresponds to a group-velocity mismatch (GVM) of 60 fs in a 30-μm-thick sensor. According to the response function modeling,[2,5] the first frequency cutoff point occurs at $(\mathrm{GVM})^{-1}=16.7$ THz, which matches the experimental results very well. The chirping shown in the time-domain waveform of Fig. 3 is also attributable to the 17 THz dip, which in principle can be shifted to 51 THz by using a 10-μm-thick sensor.

Although a thinner sensor will provide a flatter frequency response, polishing crystals as soft as ZnTe to below 20 μm with optical quality is technically challenging. We are currently investigating sensor processing, as well as other EO sensor materials. The study of various wide band mid-infrared emitters is also underway.

In conclusion, we demonstrated the first coherent detection technique in mid-infrared region up to 8 μm. This technique opens the doors in mid-infrared region for coherent emission spectroscopy, time-domain spectroscopy,[6] ultrafast pump-probe spectroscopy, and THz imaging with a better spatial resolution.[7]

This work was supported by the Army Research Office and the National Science Foundation Career Award. The authors thank L. Wang and G. A. Wagoner for their technical support and discussion. ZnTe crystal supply from Chris Ard (II–VI Inc.) and technical assistance in the crystal preparation from Molecular OptoElectronics Corporation (MOEC) are acknowledged.

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