Low-temperature THz time domain waveguide spectrometer with butt-coupled emitter and detector crystal

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Abstract: A compact high-resolution THz time-domain waveguide spectrometer that is operated inside a cryostat is demonstrated. A THz photo-Dember emitter and a ZnTe electro-optic detection crystal are directly attached to a parallel copper-plate waveguide. This allows the THz beam to be excited and detected entirely inside the cryostat, obviating the need for THz-transparent windows or external THz mirrors. Since no external bias for the emitter is required, no electric feed-through into the cryostat is necessary. Using asynchronous optical sampling, high resolution THz spectra are obtained in the frequency range from 0.2 to 2.0 THz. The THz emission from the photo-Dember emitter and the absorption spectrum of 1,2-dicyanobenzene film are measured as a function of temperature. An absorption peak around 750 GHz of 1,2-dicyanobenzene displays a blue shift with increasing temperature.

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provides video rate acquisition of time-domain THz transients without a mechanical delay generated and detected via high-speed asynchronous optical sampling (ASOPS). ASOPS pump laser at the emitter in comparison to single dipole antennas. The THz signals are into the cryostat is eliminated. The large active area of the emitter simplifies positioning the [10]. Since these emitters do not require external bias, the need for electrical feed-through currents [8, 9]. These emitters have an efficiency close to biased photo-conductive emitters [8, 9, 10].

Direct guided THz wave excitation in parallel plate waveguides has been demonstrated by Cao et al [5]. In that work, a dielectric slab-waveguide embedded within a PPWG was used, the guided wave was excited by optical rectification in a nonlinear optical polymer. Coleman et al developed a parallel plate THz transmitter which excited a guided wave in a dielectric filled PPWG via a photoconductive switch and/or optical rectification [6]. However, THz radiation via unbiased optical rectification generated much weaker field amplitudes compared to the photoconductive switch. Mirrors for in and out-coupling of THz radiation are not always necessary, e.g. Knab et al has attached the detection crystal directly to the PPWG for the near-field detection of the THz radiation [7].

In this paper we describe an improved and compact THz spectroscopy scheme in which a photo-Dember emitter and ZnTe crystal detector are directly attached to the PPWG. We make use of a recently developed large area photo-Dember emitter based on lateral diffusion currents [8, 9]. These emitters have an efficiency close to biased photo-conductive emitters [10]. Since these emitters do not require external bias, the need for electrical feed-through into the cryostat is eliminated. The large active area of the emitter simplifies positioning the pump laser at the emitter in comparison to single dipole antennas. The THz signals are generated and detected via high-speed asynchronous optical sampling (ASOPS). ASOPS provides video rate acquisition of time-domain THz transients without a mechanical delay.

1. Introduction

Time-domain THz spectroscopy has stimulated research in a wide range of disciplines and has proven to be a valuable technique for the study of vibrational excitations in molecules and crystals. Of the many different geometries available, THz parallel copper-plate waveguides (PPWG) have become important because deposition of an analyte film provides a long interaction path length resulting in high sensitivity absorption measurements [1–4]. High sensitivity and resolution are necessary for performing precision absorption measurements at cryogenic temperatures in order to elucidate the physics of low lying inter-and intramolecular vibrations. Narrower linewidths at low temperature allow the identification and quantification of spectral features that may not be resolved at room temperature. The Grischkowsky group pioneered THz absorption studies of molecular films at cryogenic temperatures with PPWG [3, 4]. In their experimental setup, high resistivity silicon lenses and parabolic mirrors are used for collimating and coupling the THz signal into and out of the PPWG. The THz radiation is generated and detected by photo-conductive antennas [1–4].
stage hence allowing easy on-line alignment and optimization of the THz signal [11–13]. In order to characterize the set-up we first investigate the temperature dependence of the THz emission from the photo-Dember emitter. We then measure a thin film of 1,2-dicyanobenzene (1,2DCB) deposited on one side of the copper plate waveguide to characterize the performance of our system by comparing it with published results [3].

2. Experimental setup

ASOPS provides a platform for ultrafast spectroscopy having excellent long-term stability and sensitivity near the shot noise limit. We implement ASOPS with two mode-locked Ti:sapphire laser oscillators working at a nominal repetition rate of approximately 1 GHz. A frequency offset between the two lasers is precisely maintained at a frequency of close to 2 kHz. One laser serves as a pump for the photo-Dember emitter and the second beam probes THz-induced reflectivity changes in a ZnTe electro-optic (EO) crystal [8–10]. A time-delay between pump and probe beam of 1 ns is scanned at the offset frequency of 2 kHz with a time resolution of 50 fs [14].

The THz waveguide consists of two polished parallel copper plates (Fig. 1). Two Si-stripes with the dimensions 125 µm × 10mm × 2mm provide a separation of the waveguide of 125 µm. Since the Si spacers are prepared from a Si wafer with homogeneous thickness, the waveguide plate separation is expected to be homogeneous with deviations smaller than 1 µm. Screws provide a pressure on the waveguide plates and the Si spacers. The temperature dependence of the waveguide plate separation is hence determined by the temperature dependence of the thickness of the Si spacers (linear expansion coefficient of Si along (100) directions is \( \alpha = 2.6 \times 10^{-6}/K \) [15]).

The assembly is attached to the temperature-controlled coldfinger of a helium flow cryostat. Near-infrared ultrashort pump pulses are focused with a cylindrical lens onto the photo-Dember emitter located directly on the PPWG entrance aperture slit. The generated THz pulses propagate through the PPWG and are detected with an EO sampling scheme [16]. The THz field induces small polarization changes in the reflected probe beam from a wedged ZnTe crystal. We carefully isolated the probe beam reflected from the inner surface of the crystal (i.e. the laser light reflected from the front surface is blocked) and focused it onto a single detector after passing through a \( \lambda/4 \) wave plate and polarizing beam splitter. The Dember structure of our emitter is fabricated on a 1 µm thick film of In\(_{0.5}Ga_{0.4}\)As grown on 514 µm thick InP wafer with an active area of 1mm x 1mm [8]. Thin films of 1,2DCB are made by dropping ~30 ml of 20mg/ml solution of 1,2DCB in toluene on one plate of the PPWG. The mass of the film is estimated to be 25-30 µg on an area of 10 x 13mm.
3. Temperature dependence of the PPWG THz spectrometer

Since the THz emitter and THz detection crystal as well as the copper waveguide are placed inside the cryostat, it is necessary to characterize the temperature-dependent response. The THz emission from the PD emitter after passing through the bare copper PPWG is shown for different temperatures in Fig. 2. The spectra in Fig. 2(b) are obtained by Fourier transformation of the time-domain wave forms in Fig. 2(a). Compared with a Dember emitter in a conventional free-space set-up with 4 paraboloids [8] the detected spectrum at 300K is strongly reduced in bandwidth in the waveguide. We attribute this effect to the less efficient coupling, transmission and detection of higher frequencies in the PPWG [1]. As the temperature decreases from 300 K to 5 K, the peak-to-peak amplitudes of THz pulses show...
an increase by more than a factor of 2. The useful frequency range of the detected THz spectra extends from 0.1 THz to almost 3 THz.

Fig. 2. (a) THz transients for the uncoated PPWG measured from 5 K to 300 K at intervals of 50 K. (b) Corresponding Fourier transform spectra (For comparison, the dotted line is the spectrum of the Dember emitter THz pulse in a free space set-up at room temperature).

First we consider the thermal effects on the copper waveguide: for a given waveguide separation and a free-space wavelength the absorption coefficient of an air-spaced PPWG is determined by the conductivity of the waveguide plates [17]. Previous studies on the PPWGs constructed of bulk copper have shown that the conductivity did not show an increase upon reducing the temperature [18]. Therefore, the temperature dependence of the amplitudes of THz pulses resulting from thermal effects of the copper waveguide can be neglected.

EO detection is a nonlinear $\chi^{(2)}$ mixing process where the temperature-dependence of the refractive index and the EO-coefficient $r_{41}$ have to be considered. The measured polarization change of the probe pulse is proportional to the anisotropic change of refractive index induced by the THz field $E_{THz}$ in the ZnTe crystal [19], i.e. it is proportional to $n_0 r_{41} E_{THz}$, where $n_0$ is the refractive index at the frequency of the probe pulse. Since the energy of the probe pulse (1.55 eV) is well below the bandgap of ZnTe (2.26 eV at 300 K) $n_0$ is expected to decrease as the band gap increases at low temperature (2.38 eV, 10 K) [20]. In addition, the frequency of the THz radiation is well below the TO phonon resonance in ZnTe (5.32 THz) [19, 21], so we also expect a weak influence of temperature on the dispersion of the THz wave. The EO coefficient $r_{41}$ is also assumed to be weakly temperature-dependent away from electronic resonances [22]. From the dispersion of $r_{41}$ measured at room temperature [23], we expect that $r_{41}$ will decrease when the band gap shifts toward higher energy at lower temperatures. All these factors lead to an anticipated decrease of the detected polarization changes when the temperature drops from 300 K to 5 K, yet we observe an increase by more than a factor of 2.
Fig. 3. Sketch of Dember emitter generating THz through lateral diffusion currents. Two gold stripes (yellow) and photoresist bars (green) are shown. The excitation spot covers more than 10 of these gold stripes. A strong carrier gradient is formed at the edge of the photoresist bars due to the ambipolar diffusion of electrons (blue dots) and holes (red dots) [8]. The faster diffusion of electrons leads to the rapid build-up of a polarization parallel to the surface with preferred THz emission into forward direction.

Photo-Dember emitters are based on ultrafast lateral diffusion currents in a semiconductor [24], hence temperature dependence of carrier transport has to be considered. The principle of THz pulse generation in such a device is shown in Fig. 3. A metalized Au stripe blocks a portion of the absorbed pump beam, producing a steep photo-carrier concentration gradient immediately beneath the stripes. The diffusion rate of electrons is much higher than that of holes, resulting in a lateral charge separation parallel to the surface. The acceleration of photo-electrons relative to holes leads to the emission of a THz pulse transient in forward direction. The gold layer has a gradually decreasing thickness that substantially weakens the photocarrier gradient in the direction away from the edge. This strongly reduces the radiation dipole vector in the reverse direction, which is necessary to build up a significant radiation signal in the far-field. The structure depicted in Fig. 3 is repeated with a period of 5 µm to form a large area emitter. More than 10 of these stripes are covered by the spot of the pump laser.

A significant difference in diffusion constants for electrons and holes is important for the THz generation via the photo-Dember effect. The diffusion constant of charge carriers given by the Einstein relation $D_i = \mu_i K_B T_i / e$, where $\mu_i$ is the mobility, $K_B$ is the Boltzmann constant, $T_i$ is the temperature of the carrier distribution, and the index i denotes electrons or holes. The mobility ratio for electrons and heavy holes in undoped $In_{0.53}Ga_{0.47}As$ at 300K is about 40 (12000 cm$^2$/Vs and 300 cm$^2$/Vs, respectively) [8], suggesting that electrons have a 40 times higher diffusion constant compared to holes. Since the diffusion length $l$ is proportional to $\sqrt{D_i t}$, it is a factor of 6 larger for electrons compared to holes in a given time $t$. The application of the Einstein relation has to be considered carefully, since the photocarriers are described by two different temperatures in the first picoseconds after excitation [25]. The effective temperature of the electrons will be higher than that of the holes because of the dispersion of the valance and conduction band. The higher electron temperature will further increase the transient diffusivity of electrons compared to the holes.

The most important temperature-dependent carrier scattering process in polar semiconductors is the interaction with LO phonons described by the Fröhlich potential. The population of LO phonons decreases as the temperature drops from 300 K to 70 K, leading to a concomitant increase of the mobility and diffusivity. This explains the temperature trend of THz peak signal that is clearly visible in Fig. 4: from 300 K down to 5 K the THz peak
amplitude increases by a factor of 2.2. One would also expect an increase in bandwidth of the detected spectrum of lower temperature when the mobility increases. However, since the detected bandwidth is limited by the PPWG set-up and not by the Dember emitter, this effect is strongly suppressed. The full description of the diffusion dynamics including nonequilibrium effects requires a Monte-Carlo simulation from which the THz radiation can be calculated [26, 27].

![Experimental data](image-url)

**Fig. 4.** THz emission (peak to peak amplitude) from Dember emitter at different temperatures with error bars.

4. 1,2DCB absorption measured in the PPWG

We made absorption measurements of 1,2DCB film at different temperatures to characterize the capabilities of our waveguide THz-TDS. To eliminate distortion from the PPWG, we recorded signals with and without the film present. Figure 5(a) shows room temperature THz transients upon propagation through the bare PPWG (blue curve) and after coating one plate with 1,2DCB film (red curve). Compared with the bare PPWG, the field amplitude of THz pulse with the 1,2DCB film is strongly reduced. The 1,2DCB film also induces a time delay of ~0.4 ps and additional ringing after the main pulse. The corresponding Fourier spectra are presented in Fig. 5(b). An absorption line at 750 GHz is in close proximity to a previously reported absorption feature at 760 GHz [3]. At frequencies above 1 THz the THz amplitude is strongly decreased due to a higher lying absorption line at 1.4 THz that is much stronger than the one at 750 GHz [3].
Figure 6 shows THz transmission spectra of 1,2DCB measured over the temperature range from 40 K to 300 K. Two trends are evident as the temperature increases: i) the high frequency cut-off of the spectra moves to lower frequencies (from around 1.4 THz at 40 K to 1.2 THz at 300 K). This is explained by the narrowing of the strong absorption peak at 1.4 THz at low temperatures as reported in [3]; ii) a blue-shift of the absorption peak near 750 GHz. The line width (90 GHz FWHM) of this peak is mainly limited by the time window used for the Fourier transform of the data. The shift of the peak around 750 GHz as a function of temperature is plotted in Fig. 7. We can rule out temperature-induced changes to the EO-detection [28]. We attribute this shift to the anharmonicity of inter-molecular vibrations resulting from a steeper inter-molecular potential at higher temperatures. The observed blue shift at increased temperatures is inconsistent with Ref [3], where a temperature-dependent red-shift of the same absorption peak is reported. We like to note that the observed shift is
consistent with experiments on 4INBP [4]. However, this discrepancy requires further studies and underscores the importance of calculating and assigning the low energy modes.

![Graph showing temperature dependence of the frequency of the absorption peak around 750 GHz with error bars.](image)

**Fig. 7.** Temperature dependence of the frequency of the absorption peak around 750 GHz with error bars.

**5. Summary**

We built and tested a compact temperature-tunable THz time domain waveguide spectrometer based on an unbiased photo-Dember emitter, electro-optic detection, and asynchronous optical sampling. We measured the far-infrared transmission of a 1,2-dicyanobenzene film in the temperature range 40—300K and found subtle changes in the absorption spectra.

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