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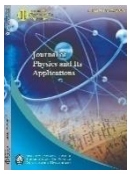

Concurrent measurement of sample and reference waveforms in an optical-pump terahertz-probe system using a controlled optical diaphragm shutter

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ABSTRACT

We present concurrent measurement of sample and reference terahertz waveforms for an optical-pump terahertz-probe spectrometer, using a controlled optical diaphragm shutter for the optical pump line. When waveforms are taken consecutively, laser power fluctuations and other experimental conditions can introduce spectral artefacts, thus a concurrent measurement is preferred. Instead of techniques based on double modulation, the use of the diaphragm shutter eliminates the need for a second lock-in amplifier and/or constricted alignment due to the use of a single chopper blade for modulating two signals, simultaneously. Drude fitting of the complex conductivity obtained for GaAs confirms that measurements obtained using our set-up agree with reported scattering times.

1. Introduction

Time resolved terahertz spectroscopy (TRTS) is a recently developed technique that can probe ultrafast carrier dynamics in the excited state[1,2]. Building upon a terahertz time domain spectrometer ("THz-TDS", Fig. 1), a third laser path is created to provide an optical pump (Fig 2). This creates an optical pump-terahertz probe (OPTP) configuration. The invention of the TRTS or OPTP has elucidated many mechanisms, such as, understanding trapping and intervalley scattering in LT-GaAs [1,3]; mobility and photoconductivity in nanowires [4,5]; exciton dynamics and conductivity in organic photovoltaics [6,7], among many others.

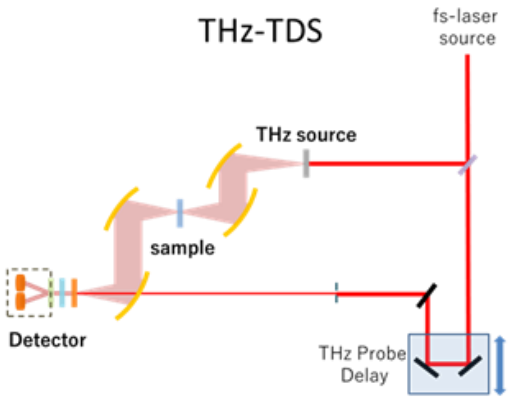

Fig. 1: Schematic of a typical THz time domain spectrometer

Since the development of THz-TDS, its use has become widespread across disciplines and materials. It provided a new way of studying properties of molecular and atomic crystals, pharmaceutical and biological samples, aqueous solutions, dielectrics and even gases [2]. The basic transmission experiment is conducted by passing a THz pulse,through a sample, $E_{smp}(t)$, and through a reference material, $E_{ref}(t)$. The ratio of their corresponding frequency spectra gives the spectral transmission $\tilde{T}(\omega)e^{i\varphi(\omega)}$:

$$\frac{\tilde{E}_{smp}(\omega)}{\tilde{E}_{ref}(\omega)} = \tilde{T}(\omega)e^{i\varphi(\omega)} \quad (1)$$

$$\tilde{T}(\omega)e^{i\varphi(\omega)} = \hat{t}_{12}\hat{t}_{23}e^{-\alpha d/2}e^{i(n-1)\omega d/c} \quad (2)$$

for a system where the THz pulse travels from medium 1, incident normally to the sample (medium 2), and exits to medium 3. Frequency (ω)-dependent functions are denoted by the tilde symbol. The spectral amplitudes and phases are compared as Eq. 1, from which Fresnel coefficients ($\hat{t}$) determine the amplitude transmitted when moving from one medium to the next, the subscripts "12" and "23" denote the movement of the THz pulse from medium 1→2 and 2→3, respectively; while thickness (d), as well as frequency-dependent absorption (α) and complex refractive index (n) information can be obtained from the phase difference (Equation 2).

For the pump-probe scheme, a third path from the laser source is created in order to provide optical

excitation (Fig.2). A delay stage is provided in order to control the pump-probe delay time (Δt), which allows for the observation of time-bound processes. The time-domain nature of detection thus allows the study of charge dynamics on the subpicosecond timescale. [1-3].

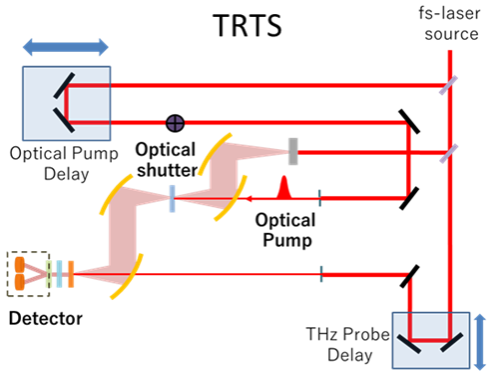

Fig. 2 : Schematic of time resolved terahertz spectrometer, with an optical shutter on the optical pump line

In the configuration where the optical pump arm and the THz detection arm are equipped with their own delay stages, the transmitting THz radiation (probe) monitors the sample's response to the optical excitation (pump). This optical pump-terahertz probe configuration has been used to study semiconductors, nanostructures, dielectrics and even semi-metals. In general, for thin conducting films, the THz pulse passing through the photoexcited layer gives sample spectra $E_{smp}(t)$, and the reference $E_{ref}(t)$, is the THz pulse passing through the unexcited material (instead of air, or a reference medium). In a similar fashion, the transmission $\tilde{T}(\omega,\Delta t)$ is obtained from the ratio [2]:

$$\frac{\tilde{E}_{smp}(\omega)}{\tilde{E}_{ref}(\omega)} \Rightarrow \frac{\tilde{E}_{exc}(\omega,\Delta t)}{\tilde{E}_{ref}(\omega)} = \left|\tilde{T}\right| e^{i\varphi} \qquad (3)$$

And the complex transmission can be obtained [2,8]:

$$\tilde{T} = \frac{n+1}{n+1+Z_0\tilde{\sigma}(\omega,t)d} \qquad (4)$$

For this configuration, instead of taking transmission measurements through a sample medium and a reference medium, we are comparing transmission with or without optical excitation. Thus, any fluctuation in laser power, or changes in the experimental environment can introduce false information to the measurement. It becomes favorable to take the reference and sample spectra simultaneously, or rather concurrently, in order to keep the experimental conditions between the two spectra as similar as possible. In this work, we report how the use of an optical shutter allows for the concurrent measurement of the photoexcited and unexcited THz waveforms.

2. Methodology

For OPTP set-up, we useda Ti:Sapphire regenerative amplifier system, with a repetition rate of 1 kHz, pulse duration of ~50 fs at 800 nm, and output power of 3 mJ. The laser path is split into the optical pump, the THz probe and the THz detector, such as in the configuration shown in Fig. 2. For the THz source, the laser is focused tightly, and made to pass through a BBO crystal to create a two-color air plasma [9].The THz radiation transmitting through the sample is then detected by means of electro-optic sampling[10],using a 1 mm-thick ZnTe (110) crystal, and a pair of balanced photodiodes. The optical pump pulse and the THz probe pulse are aligned such that they are incident normal to the sample surface.The sample is placed on a mount with a 2.5 mm diameter pinhole; this diameter is smaller than both the incident pump beam (~3 mm) and the THz beam waist (~3.2 mm) in order to ensure complete overlap.The optical pump fluence is controlled by a pair of polarizer-analyzer films.

The two methods by which an optical pump and probe set-up is used are so-called "1D" and "2D" scans, following the number of stages involved in data acquisition. The "1D scan" involves a fixed THz probe delay at the terahertz peak, and the optical pump stage timing is scanned. When the timing between the pump and the probe coincide, the change in transmission can be observed within the measurement time frame.

The second method is obtaining the full terahertz waveform as it passes through the sample under the conditions that the sample is excited and unexcited. This requires the two stages to move synchronously in order preserve the correct pump-probe delay time. When measuring a differential waveform, a concurrent method is recommended – points on the waveform are acquired in an alternating manner, and at the end of the scan two waveforms are acquired. This keeps error caused by laser position and power fluctuations minimal between reference and sample waveforms.

In order to obtain the full THz waveform, several groups [4,11] implemented a double modulation scheme to TRTS – however, this technique requires two optical choppers – for the pump and the probe – as well as two lock in amplifiers. The double modulation scheme, normally used to reduce noise, was also proposed by Iwaszczuk, et al [12] in 2009, using a single dual frequency chopper blade and thus requiring only one lock-in amplifier. The THz generation arm and the optical pump arm pass through a 2-frequency chopper blade, one on the outer slots and the other through the inner slots.

Our method only requires placing an optical diaphragm shutter in the optical pump path (Fig. 2). A 1-in. diameter optical diaphragm shutter with a programmable controller was used. The diaphragm shutter was set-up such that it interfaces with our home-built data acquisition program. To take concurrent measurements, data points tracing the THz waveforms are taken alternately, with the shutter open and then with the shutter closed. The sequence is shown in Fig.3. At every time delay, the pump (and probe) stage(s) take (synchronous) time steps while the shutter is open. A data point (i.e. intensity) is acquired after a variable waiting time; then, the shutter is closed and a data point is

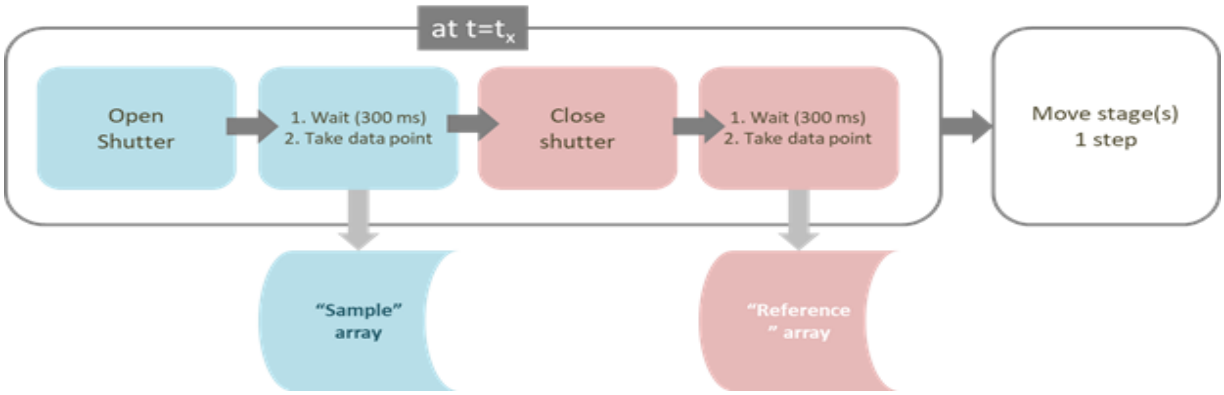

Fig. 3: Process flow for the concurrent measurement of sample and reference data point at a time $t=t_x$

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acquired after the same amount waiting time. After the stage(s) takes a step forward, the sequence is repeated.

The open and close times, quoted from the manufacturer, are typically 8 ms and 9 ms, respectively. All the timings for data acquisition can easily be edited via the user interface. Naturally, the data acquisition program is synchronized with the lock-in amplifier. Under normal operation, the lock-in amplifier time constant is 100 ms, and a waiting time is digitally introduced into our program, which we typically set to 300 ms. Thus, the effect of a < 10 ms open or close time can be compensated. The balanced photodiodes used in this set-up have an effective decay time of ~390 μs, taking account for the circuit resistance and capacitance of the detector unit. For detectors with longer response times, lock-in amplifier time constants and data acquisition waiting time should be adjusted, accordingly.

3. Results and Discussion

GaAs has well known Drude conductivity response [3,13], and is thus a good material for checking the system. The waveforms obtained concurrently for a semi-insulating GaAs substrate using the diaphragm shutter and time delay system is shown in Fig. 4.

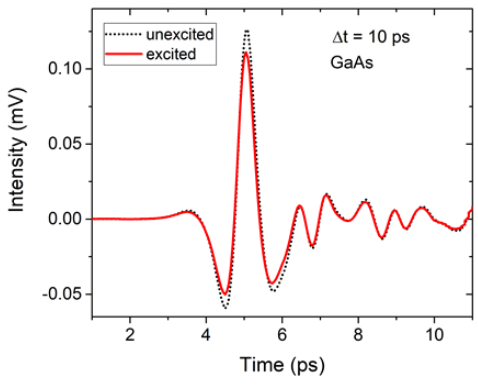

Fig. 4: Concurrently measured reference and sample waveforms for GaAs, excited at 4 μJ/cm², with a fixed pump-probe time delay of Δt = 10 ps.

In order to properly utilize the thin film approximation, the photo-induced change in transmission must be kept below 20% [13, 14]. For our GaAs films, a pump fluence of 4 μJ/cm² induces a change in transmission of approximately 20%. Representative data shown in Fig. 4 was measured at a pump-probe delay of Δt = 10 ps. The dashed curve shows the unexcited waveform; when the sample is excited, free carriers are able to absorb the incoming THz radiation, reducing the transmission intensity and creating a slight temporal shift, as shown by the red curve. The complex conductivity spectra were then extracted in the following manner [2]:

$$\sigma'(\omega,t)=\frac{n+1}{Z_0 d}\left(\frac{1}{|\tilde{T}|}\cos\varphi-1\right) \qquad (5a)$$

$$\sigma''(\omega,t)=\frac{n+1}{Z_0 d}\left(\frac{1}{|\tilde{T}|}\sin\varphi\right) \qquad (5b)$$

where n is the index of refraction of the material at THz frequencies (n_{GaAs}=3.6), $\tilde{T}$ is the transmission described by Eqtn. 4, Z_0 is the impedance of free space, and d is the thickness of the excited layer (in the case of GaAs at 800 nm, this is ~1 μm). The corresponding complex conductivity dispersion is shown in Fig. 5. Due to water absorption, artefacts in the dispersion curves can be seen at ~1.7 THz.

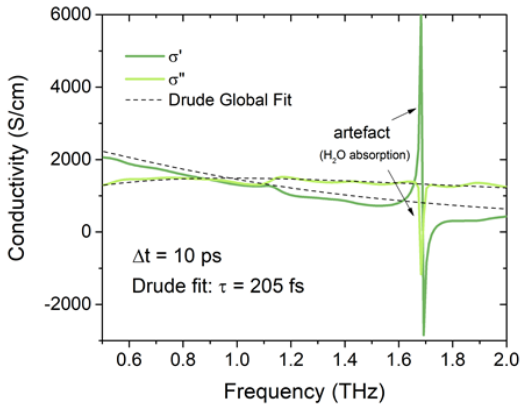

Fig. 5 :Drude fitting at a pump-probe delay time of 10 ps. The Drude scattering time is 205 fs, in agreement with other measurements as well as literature.

Subsequently, the complex conductivity dispersion is globally fitted using the Drude model [15], where the Drude scattering time and plasma frequency are free fitting parameters. Table 1 shows the comparison of Drude fitting parameters obtained for the same sample using a different OPTP set-up [16]. The values in both systems are in agreement with reported values under similar conditions [13].

Table 1 Comparison of Drude fits with a double-modulation based OPTP system

Set-up	Detector	Range for Drude Fitting	Δt=10ps		(Δt=100ps)	
			Scattering time T	Plasma frequency ω	Scattering time τ	Plasma frequency ω
Optical Shutter	ZnTe (EO-sampling)	0.5 – 2.0 THz	205 fs	5.88 THz	220 fs	5.41 THz
Double-modulation	Air-biased coherent detection	2.5 – 4.0 THz	208 fs	7.48 THz	226 fs	5.88 Thz

For the two systems, the detection schemes are different, and are thus sensitive at different THz frequencies. Furthermore, the system with double modulation is within a dry air environment, and is free of water vapor absorption artefacts. Bandwidth for the air-biased coherent detection [9] is broader. But despite the difference in the frequency range used, the results of the fitting are still quite close to each other.

4. Conclusions

The integration of an optical diaphragm shutter as a method to concurrently obtain sample and reference spectra for time-resolved spectroscopy measurements has been demonstrated. Compared to double-modulation based solutions, the use of a diaphragm shutter eliminates the need to use extra optical choppers and/or lock-in amplifiers. The diaphragm shutter open and close times can easily be controlled through the data acquisition program, which also creates flexibility for the user to choose the sampling rate. Photoconductivity measurements done on a GaAs substrate show Drude scattering times that agree with previous measurements and other published results.

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