Terahertz Time-Domain Spectroscopy with Continuous Coverage of the Entire Terahertz Range

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***Abstract*—Using gases as the emitter and detector media in a THz system allows for good phase matching over the full terahertz range (0.3 to 10 THz), without interruptions from crystal phonon modes or other instrument artifacts. A heterodyne technique facilitates coherent measurement of the electric field waveform, enabling this technique to be used for time-resolved spectroscopy in which a single measurement can continuously cover the entire terahertz region of the electromagnetic spectrum.**

I. Introduction And Background

Terahertz time-domain spectroscopy (THz-TDS) has been established as a useful tool for the study of many different types of materials and interactions. Typically, THz-TDS systems have made use of crystalline materials for the generation and detection of the THz pulse, which have had the advantage of good efficiency and sensitivity, but spectra limited by material properties such as phonon modes or carrier lifetimes, or by resonant features of antenna structures. Since phonon resonances appear quite often in the THz range, they place limits on the continuous bandwidth of the THz pulse, either through absorption or loss of phase matching due to dispersion. If a non-absorbing gas is used as the medium, however, this problem does not appear, and the observed spectra are limited mainly by the properties of the optical pulse.

II. Results

Utilizing 80 fs, 700 µJ pulses from a Spectra-Physics Hurricane, we used dry nitrogen as the emission [1, 2] and detection [3,4] medium. Terahertz pulses were produced by focusing the (~500 µJ) pump pulse through a ß-barium borate (BBO) crystal to produce a pulse at the second harmonic, which then focused collinearly with the fundamental pulse with intensity high enough to form a laser-induced plasma. During ionization, a THz pulse is generated through a process that has been described both through perturbative nonlinear optics and semi-classical electron dynamics [2]. This pulse was collected and refocused by a set of parabolic mirrors and the pump light was filtered out using a silicon wafer. The terahertz pulse was then detected using a four-wave mixing process where it was focused collinearly with an 800 nm, ~200 µJ probe pulse, producing a second harmonic pulse. An AC bias was applied to the focus, generating a phase-modulated local oscillator through field-induced second harmonic generation. The terahertz-induced second harmonic and local oscillator were then detected using a photomultiplier tube. A lock-in amplifier isolated the desired coherent heterodyne term by being referenced to the AC bias frequency. A schematic diagram of the THz spectrometer is shown in Figure 1.

Before being used for detection, the probe pulse passes through a Keplerian beam expander, resulting in self-steepening and spectral broadening. This increases the detectable bandwidth of the THz pulse, indicating that the primary limitation on the system bandwidth is the duration of the amplified laser pulse [5].

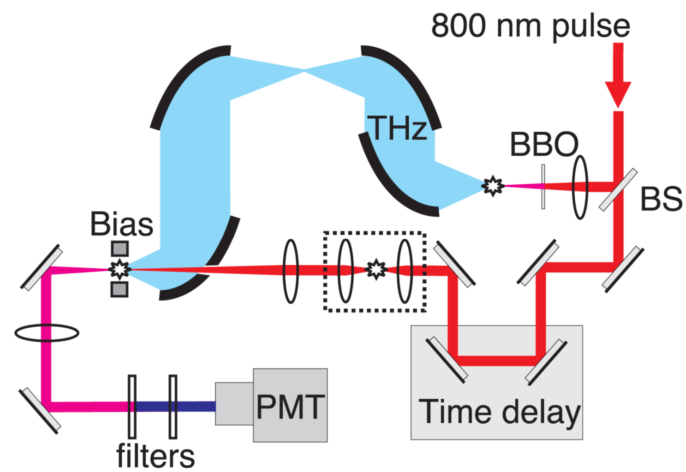

Fig. 1. Schematic diagram of the THz spectrometer. The input optical pulse is split into pump and probe beams with a beam-splitter (BS) with a ratio of 77:23. The pump beam is focused by a 125 mm focal length lens through a type-I BBO crystal, producing a second harmonic pulse. The fundamental and second harmonic pulses interact to produce a THz pulse, which is collected and focused by a pair of off-axis parabolic mirrors. Another pair of mirrors, the last of which has a small hole through the center, does the same, focusing the beam for detection. The probe pulse passes through a variable time delay and Keplerian beam expander (for spectral broadening, marked with dotted outline) and is focused by a 150 mm focal length lens through the hole in the final parabolic mirror. The probe pulse and THz pulse produce a second harmonic pulse through four-wave mixing. An AC bias placed across the focus produces a local oscillator second harmonic pulse through field-induced second harmonic emission, which is used to coherently detect the THz field by measuring their combined intensity with a photomultiplier tube (PMT).

The detected terahertz peak had a peak-to-peak temporal duration of less than 100 fs and continuous spectrum covering 0.1 THz to above 10 THz, finally reaching the noise floor of the measurement near 20 THz. The waveform and spectrum are shown in Figure 2. The peak amplitude of the THz pulse was measured to be 80 kV/cm by a simple comparison of the field-induced second harmonic and THz-induced second harmonic pulse energies.

These results establish the generation and detection of THz pulses in gases as a unique method for broadband THz-TDS capable of fully utilizing the bandwidth of ultrashort laser pulses.

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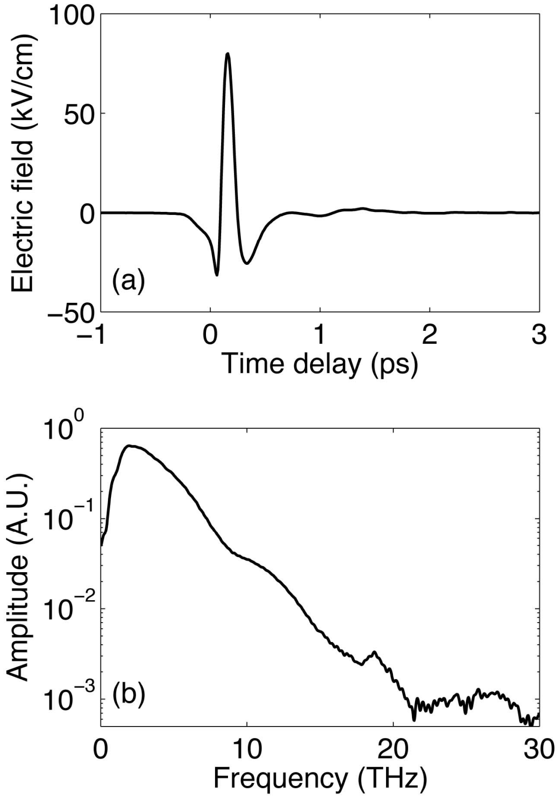

Fig. 2. (a) Waveform and (b) spectrum of the measured THz pulse. The spectrometer was purged with dry nitrogen gas for all measurements.

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