# Picosecond pump and probe spectroscopy utilizing freely propagating terahertz radiation

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We demonstrate the feasibility of scaling up a terahertz-pulse generation scheme for use with a 10-Hz amplified femtosecond laser system. Visible pulsed excitation combined with a far-infrared probe should prove to be a powerful picosecond time-resolved technique.

The past three years have seen dramatic progress in the development and characterization of picosecond far-infrared light sources and detectors. Subpicosecond visible optical pulses have been utilized to generate essentially single-cycle radiation at approximately 1 THz (33 $cm^{-1}$). Schemes based on Hertzian dipole emission from biased ultrafast photoconductive antennas,[1-3] Čerenkov radiation in electro-optic crystals (optical rectification),[4,5] and, most recently, charge acceleration through the built-in electric fields present at semiconductor surfaces have all been reported.[6] Gated detection of such light has been performed either by electro-optic sampling or with conventional antenna structures.

While the majority of research with picosecond terahertz light pulses has concentrated on techniques of generation and detection, two notable studies have utilized this light to perform pump and probe nonlinear spectroscopy. The first generated freely propagating terahertz light by propagating a high-voltage picosecond electrical transient into a dipole antenna.[7] The emitted infrared light was in turn transmitted through a semiconductor sample (Ge or GaAs), which was excited with a time-delayable visible light pulse. Finally, the terahertz light was detected with a standard cw millimeter-wave microwave diode detector. The second study utilized Čerenkov radiation bound within a $LiTaO_3$ crystal to perform nonlinear spectroscopy on an optically contacted GaAs crystal.[5] Beam focusing and timing constraints of this latter technique dictated a specialized pump–probe geometry.

In this Letter we continue along these lines with the objective of demonstrating that freely propagating picosecond terahertz light pulses can be utilized in a totally conventional and versatile pump and probe experimental arrangement. The major advantage of our technique relies on an ability to scale up the infrared pulse energies beyond those previously obtainable. Combined with sufficiently high visible excitation pulse energies (~100 $\mu$J) available with low-repetition-rate amplified laser systems, reasonable sample excitation densities make many pump and probe experiments feasible. This holds particularly true in light of the constraint of diffraction-limited spot sizes at the sample of no less than ~1 mm for the broadband terahertz probe light.

A 10-Hz YAG-pumped, amplified colliding-pulse, mode-locked dye-laser system was used to generate approximately $2 \times 10^{-4}$ J, 70-fsec light pulses at 630 nm. The laser light was divided into three beams, all delayable with respect to one another. The largest portion of the light excited an InP wafer, which generated a terahertz light pulse. This light was focused with an off-axis paraboloid onto the sample and, in turn, onto the detector. The detector consisted of a 200-$\mu$m dipole antenna fabricated on an epilayer of low-temperature-grown GaAs.[8] The low-temperature-grown GaAs served as the high-speed photoconductive gate that had low dark current and good light sensitivity compared with that of radiation-damaged silicon.

A second portion of the 630-nm light gated the detector, and a pulsed current was detected as a function of delay time between the antenna gate and the arrival of the terahertz light pulse. With a gain of $10^8$ V/A and a $1 \times 10^{-5}$ sec bandwidth, peak signals of 200 mV were observed. A typical trace of the electric field versus the delay time is shown in Fig. 1.

Spectral analysis (not shown) reveals a peak at roughly 0.3 THz, with essentially no light beyond 0.5 THz. We believe the response to be largely antenna limited. Recent results obtained in our laboratory

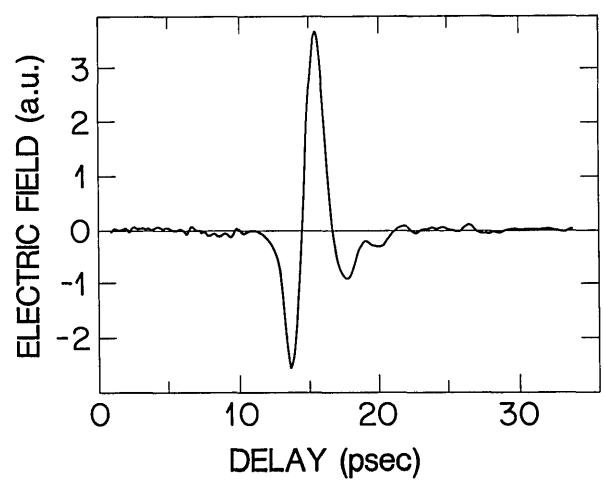


Fig. 1. Detected electric field versus the delay time between the antenna gate pulse and the terahertz light pulse.

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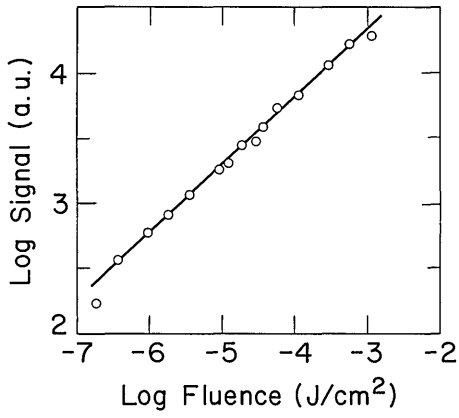


Fig. 2. Peak signal at the detector versus 630-nm pulse fluence onto an InP wafer.

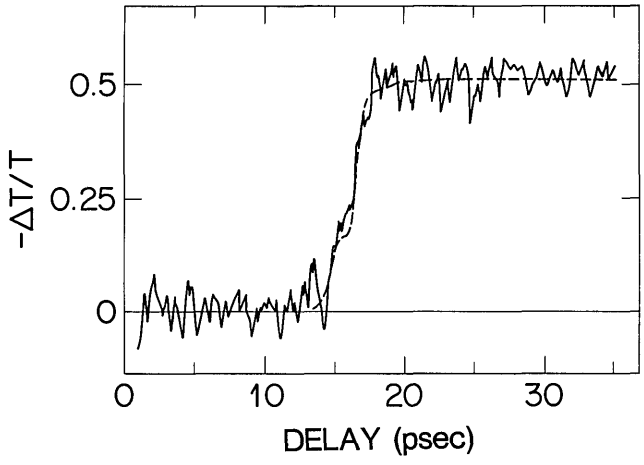


Fig. 3. Transmission of terahertz light through a 0.5-mm-thick GaAs wafer versus the visible excitation delay (solid curve) and the instrument response (dashed curve).

with a broadband antenna design reveal significant spectral power out past 1.0 THz. A plot of the peak signal at the detector versus the 630-nm pulse fluence onto the InP wafer is shown in Fig. 2. The terahertz light was observed not to be strongly saturated. At the lowest pulse fluence, the signal was approximately linear. From $10^{-6}$ to $10^{-3}$ J/cm$^2$ the signal varied as the square root of the fluence. Significant saturation was only observed at the highest fluence levels ($\gtrsim 10^{-3}$ J/cm$^2$). We believe the square-root dependence to be a consequence of the photoinjected carriers' screening the surface field. Depletion layer widths and resultant field strengths are expected to vary as $1/\sqrt{N}$, where $N$ is the carrier density.

The final portion of the visible laser light was used to excite the sample, in this case a 0.5-mm-thick wafer of semi-insulating GaAs. The sample was placed at Brewster's angle for the visible light, and a 0.7-cm aperture was attached to ensure uniform excitation and pump–probe overlap.

Absorption of the visible excitation light is expected to create free electrons and holes, resulting in infrared Drude absorption. A trace of the sample transmission versus the excitation delay is shown in Fig. 3, which shows an ~1.5-psec rise time. While full $E(t)$ traces were measured and analyzed, the primary transient effect was in the magnitude of $E$. The data in Fig. 3 are a continuous trace of the peak $E(t = \text{peak})$ versus visible excitation delay. The carrier density was estimated to be $6 \times 10^{18}$ cm$^{-3}$. Also shown in Fig. 3 is the time integral of $E^2$, which should correspond closely to the instrument response of our apparatus. The rise of infrared absorption in GaAs was therefore observed to occur in less than roughly 1 psec. Previous studies performed on GaAs with Čerenkov terahertz light pulses reported time-dependent mobility data derived from far-infrared transient reflectivity measurements.[5] More complete temperature-dependent studies of the far-infrared optical constants of GaAs utilizing transient transmission data obtained with our apparatus are under way.

We conclude that picosecond pump and probe spectroscopy utilizing visible excitation pulses and terahertz probe pulses is a feasible and versatile experimental technique. An entirely new wavelength region should now become routinely accessible to picosecond kinetic spectroscopy. It seems reasonable to assume that a wealth of new experimental research will ensue.

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