Single-shot measurement of terahertz electromagnetic pulses by use of electro-optic sampling

Jie Shan, Aniruddha S. Weling, Ernst Knoesel, and Ludwig Bartels

Departments of Physics and Electrical Engineering, Columbia University, New York, New York 10027

Mischa Bonn

Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin 14195, Germany

Ajay Nahata

AlliedSignal Corporation, Morristown, New Jersey 07962

Georg A. Reider

Abteilung Quantenelektronik und Lasertechnik, Technische Universität Wien, Vienna A-1040, Austria

Tony F. Heinz

Departments of Physics and Electrical Engineering, Columbia University, New York, New York 10027

Received November 18, 1999

We demonstrate a simple scheme for capturing the temporal waveforms of a freely propagating terahertz electromagnetic transient in a single shot. The method relies on electro-optic sampling in a noncollinear geometry for the terahertz radiation and the visible probe beam, coupled with multichannel detection. The approach provides time resolution that is comparable to that of conventional electro-optic sampling measurements.

OCIS codes: 320.7100, 250.0250.

Electro-optic (EO) sampling has proved to be a powerful technique for the measurement of electrical transients in both transmission lines[1] and free space.[2] Time resolution sufficient for measuring electric fields in the mid infrared[3,4] has been demonstrated with ultrafast pulses from a mode-locked laser in a sampling mode in which the relative delay between the electrical transient and the probe pulse is varied for each data point. In this Letter we present a new approach to such EO measurements that permits a complete temporal waveform of freely propagating THz radiation to be captured in a single shot. The scheme, based on an easily implemented noncollinear detection geometry, retains the excellent time resolution of conventional EO sampling. At the same time, the method provides the possibility of significant improvement in the signal-to-noise characteristics of THz measurements. The method should facilitate THz pump–probe measurements,[5] particularly when low pulse-repetition rates are used to attain high pump fluences or to probe samples that degrade or undergo irreversible change.

In EO detection the THz signal is effectively upconverted to the optical region by ultrafast probe-laser pulses. This permits the use of the highly developed multichannel detectors for parallel data collection. In recent experiments this approach has been applied to novel time-domain THz measurements, including the capture of spatial[6] and temporal[7] profiles of freely propagating THz electromagnetic transients. In the latter case the detection scheme relies on transcribing the temporal profile of the THz pulse on the spectral profile of a linearly chirped optical probe pulse.[7] In contrast, in the scheme presented here the temporal waveform of the THz electric field is imposed directly on the transverse *spatial* profile of the probe beam. This approach retains the exceptional temporal resolution of conventional EO sampling measurements and is free of the trade-offs inherent in combining frequency- and time-domain methods.

The basic concept of the parallel EO detection scheme is shown in the right-hand part of Fig. 1. In conventional free-space EO sampling, an optical pulse propagates collinearly with the THz electromagnetic transient. Through the EO effect, the probe pulse experiences a polarization modulation that is proportional to the strength of the THz electric field in the EO sampling medium. For the multichannel EO detection scheme presented here, we simply modify

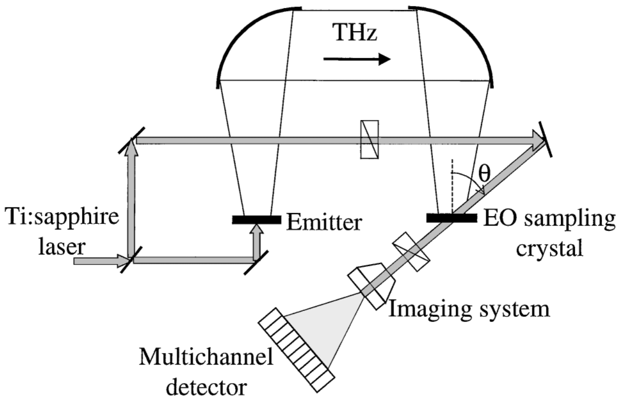

Fig. 1. Schematic drawing of the experimental setup used to capture the temporal waveforms of freely propagating THz radiation by multichannel EO sampling.

0146-9592/00/060426-03$15.00/0

this arrangement by introducing the probe-laser beam at an angle with respect to the THz radiation. In the crossed-beam geometry there is a temporal skew in the probe wave front with respect to the THz wave front. Different points across the transverse profile of the probe beam see the THz electric field at different instances in time, and the temporal image of the THz waveform is thus impressed on the transverse spatial variation of the probe beam. The mapping between the time coordinate t and the spatial coordinate x (as shown in Fig. 1) can be readily seen to be

$$t = x \tan\theta / c\,, \tag{1}$$

where θ is the angle (in air) between the two beams. The total window of time, $\Delta T = w \tan\theta/c$, probed during the measurement is determined by the width w of the incident probe beam.

In addition to the crossed-beam EO sampling arrangement discussed above, Fig. 1 displays the generation of the freely propagating THz radiation by means of optical rectification of the laser pulses. The laser source for these measurements was a Kerr-lens mode-locked Ti:sapphire laser that we amplified regeneratively to produce pulses of ~150-fs duration at a 1-kHz repetition rate and a wavelength of 810 nm. As a nonlinear material for both optical rectification and EO sampling, we made use of ZnTe crystals that were cut in a $\langle 110 \rangle$ orientation and were 1 mm thick. For THz generation we irradiated the ZnTe crystal at normal incidence with 20-μJ laser pulses polarized along the $[1\bar{1}0]$ direction. For detection the THz beam was directed onto the ZnTe sampling crystal at normal incidence and polarized along the $[1\bar{1}0]$ direction. The THz beam had a diameter of 3 mm. The optical probe beam was polarized perpendicularly to the THz beam and was incident on the EO crystal at an angle of 31°. The probe beam had a diameter of 2.5 mm, which yields, according to Eq. (1), a time window for capture of the THz waveform of ~5 ps. One can increase this time window by choosing a larger angle of incidence or probe-beam diameter. The transmitted probe beam passed through a polarizer and was imaged on a 1024-element Si photodiode array through an optical system providing 8× magnification. The EO sampling was performed in a configuration with crossed polarizers near the null in the optical transmission. To operate in the region of a linear variation of the transmitted beam with the strength of the THz electric field, one needs a slight static birefringence. This was provided by the weak residual birefringence in the nominally isotropic ZnTe crystal.[6–8] One could also introduce a wave plate to increase and control the static birefringence, if desired.

A representative waveform of a single THz transient is displayed as the top curve of Fig. 2. The waveform was recorded with a single probe pulse by use of the multichannel detection scheme described above. The data were corrected for the spatial inhomogeneity of the probe beam by normalization of the signal in each channel to that measured in the absence of the THz electric field. The calibration of the time axis in these measurements can be obtained from Eq. (1) with knowledge of the experimental geometry. A simpler scheme, however, is to record the same THz waveform with probe pulses arriving at different delay times introduced by a conventional optical delay line. The time difference between signals recorded in adjacent Si photodiodes in the multichannel array corresponds to 4.9 fs. To realize this time resolution would obviously require far shorter probe pulses than those of 150-fs duration used in the present measurement.

The solid curve at the bottom of Fig. 2 displays the corresponding THz waveform collected by a conventional sampling measurement. We recorded the data with a single photodiode (number 500 in the array) while scanning a delay line. The resulting trace agrees reasonably well with the waveform acquired in a single shot by the parallel detection scheme. We attribute the difference to the influence of shot-to-shot fluctuations in the pump laser. As a comparison, we also show (dotted curve) averaged data for multichannel detection. The waveforms collected by the two methods are clearly consistent and show a comparable signal-to-noise ratio (SNR). For each of the measurement schemes the displayed data correspond to an average of 2500 laser shots for each time interval in the THz waveform. Since the results for the time scan were recorded by sampling with 200 distinct delay times, the corresponding measurement took precisely 200 times longer than the corresponding measurement with multichannel detection. This result illustrates the significant improvement in SNR that is attainable by use of multichannel detection. Indeed, if the primary source of noise lies in laser fluctuations, the speed of data collection at a specified SNR will scale

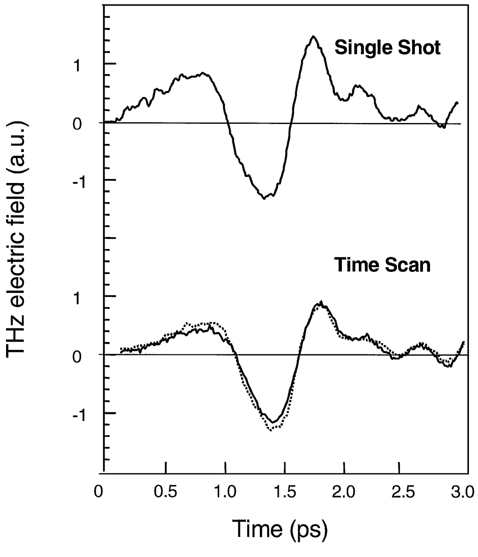

Fig. 2. Electric field waveforms for THz pulses generated by optical rectification. The figure shows a single-shot measurement of a THz pulse (top), a THz waveform obtained by averaging of single-shot data (dotted curve, bottom), and a waveform obtained in the conventional fashion with a scanning delay line (solid curve, bottom). The bottom curves both correspond to averaging of 2500 laser pulses for each data point. The total data-collection time of the sampling measurement exceeds that of the multichannel detection by a factor of 200.

simply as the number of distinct data points in the time-delay scan. Note that the SNR for multichannel detection can be enhanced through application of balanced detection with two diode arrays, by analogy with the scheme for balanced detection in the conventional EO sampling.

As a final point we consider briefly two propagation effects that can influence the time resolution of multichannel detection: phase matching in the noncollinear geometry and diffraction of the probe beam as it travels through the EO crystal. The first issue has been recognized as a critical factor in achieving optimal time resolution in conventional EO sampling measurements.[9–11] One can obtain the relevant phase-matching criterion by recognizing the EO coupling between the probe and the THz waves as sum- and difference-frequency generation processes. Assuming weak dispersion in the refractive index n of the EO crystal and ω_{THz} (a frequency in the THz bandwidth)/ω_{opt} (central frequency of the optical beam) $\ll 1$, we obtain the phase-matching condition for an isotropic medium:

$$N_{\mathrm{opt}} = n_{\mathrm{THz}} \cos \theta_i \,, \tag{2}$$

where $N_{\mathrm{opt}} = n + \omega(\partial n/\partial \omega)|_{\omega_{\mathrm{opt}}}$ is the group index at the optical frequency and θ_i is the internal angle between the optical and the THz beams. The phase-matching condition of Eq. (2) can be understood as stipulating that the component of the group velocity of the optical pulse along the direction of propagation of the THz pulse should match the relevant THz phase velocity. Recently, it was shown that by exploiting the dispersion in the optical refractive index of cubic semiconductors such as ZnTe and GaP one may achieve collinear phase-matched EO sampling over a considerable range of THz frequencies.[9,10] For a noncollinear geometry, satisfying the phase-matching criterion will typically be easier than in the collinear geometry, since the THz refractive index n_{THz} generally exceeds N_{opt} away from the region of strong dispersion.

The second propagation effect that can limit the achievable time resolution of the method is diffraction of the probe beam as it passes through the EO crystal. One can estimate this limit by determining the smallest width of a segment of the incident probe beam that will undergo negligible spreading in propagating through the crystal. Fresnel diffraction theory and Eq. (1) yield an estimate for the limitation of the time resolution of $\sim$(29 fs)$\sqrt{d}$ in our setup, where d is the thickness of the EO crystal in millimeters. Although this effect is slight, it could clearly be reduced, if desired, by a decrease in the thickness of the crystal.

In conclusion, we have demonstrated an adaptation of the electro-optic sampling scheme for the single-shot acquisition of temporal waveforms of free-space THz electromagnetic pulses. The method involves transcribing the temporal profile of the THz pulse onto the transverse spatial profile of the optical probe beam through the use of a noncollinear geometry. This approach shares with conventional EO sampling the possibility of extremely high time resolution but improves the rate of data collection by means of parallel detection.

We acknowledge helpful discussions with Kolo Wamba and Jerry Dadap. This work was supported by National Science Foundation grant CHE-96-12294, U.S. Air Force Office of Scientific Research grant F49620-98-1-0137, and the Joint Services Electronics Program (DAAG55-97-1-0166). E. Knoesel and L. Bartels thank the Humboldt and Leopoldina Foundations, respectively, for postdoctoral fellowships. T. F. Heinz's e-mail address is tony.heinz@columbia.edu.

References

1. J. A. Valdmanis and G. Mourou, IEEE J. Quantum Electron. **22,** 69 (1986).
2. Q. Wu and X.-C. Zhang, Appl. Phys. Lett. **67,** 3523 (1995); A. Nahata, D. H. Auston, T. F. Heinz, and C. Wu, Appl. Phys. Lett. **68,** 150 (1996); P. Uhd Jepsen, C. Winnewisser, M. Schall, V. Schya, S. R. Keiding, and H. Helm, Phys. Rev. E **53,** 3052 (1996).
3. Q. Wu and X.-C. Zhang, Appl. Phys. Lett. **71,** 1285 (1997).
4. A. Leitenstorfer, S. Hunsche, J. Shah, M. C. Nuss, and W. H. Knox, Appl. Phys. Lett. **74,** 1516 (1999).
5. See, for example, R. H. M. Groeneveld and D. Grischkowsky, J. Opt. Soc. Am. B **11,** 2502 (1994); G. Haran, W. Sun, K. Wynne, and R. M. Hochstrasser, Chem. Phys. Lett. **274,** 365 (1997); S. S. Prabhu, S. E. Ralph, M. R. Melloch, and E. S. Harmon, Appl. Phys. Lett. **70,** 2419 (1997).
6. Q. Wu, T. D. Hewitt, and X.-C. Zhang, Appl. Phys. Lett. **69,** 1026 (1996).
7. Z. Jiang and X.-C. Zhang, Appl. Phys. Lett. **72,** 1945 (1998).
8. Z. Jiang, F. G. Sun, Q. Chen, and X.-C. Zhang, Appl. Phys. Lett. **74,** 1191 (1999).
9. A. Nahata, A. S. Weling, and T. F. Heinz, Appl. Phys. Lett. **69,** 2321 (1996).
10. H. J. Bakker, G. C. Co, H. Kurz, Q. Wu, and X.-C. Zhang, J. Opt. Soc. Am. B **15,** 1795 (1998).
11. G. Gallot and D. Grischkowsky, J. Opt. Soc. Am. B **16,** 1211 (1999).