Electro-optic detection of femtosecond electromagnetic pulses by use of poled polymers

Hua Cao

NEC Research Institute, Inc., Princeton, New Jersey 08540, and
Department of Electrical Engineering, Princeton University, Princeton, New Jersey 08540

Tony F. Heinz

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

Ajay Nahata

NEC Research Institute, Inc., Princeton, New Jersey 08540

Received November 9, 2001

We report the generation and coherent detection of freely propagating ultrashort baseband electromagnetic pulses. Using optical rectification in ⟨110⟩ GaAs for wideband emission and electro-optic sampling in a poled polymer for wideband detection, we demonstrate spectral sensitivity that extends from the far infrared ($\lambda \sim 100\ \mu$m) to ~33 THz ($\lambda = 9\ \mu$m). Over a band of nearly 20 THz, a relatively flat frequency response is observed. We discuss issues that limit the response bandwidth. © 2002 Optical Society of America

OCIS codes: 320.7110, 320.7120, 190.4400, 190.7220, 250.2080.

The generation and detection of ultrashort electromagnetic pulses has become a subject of increasing interest in recent years. Potential applications that arise from this capability include the study of nonequilibrium carrier dynamics in semiconductors[1] and nonlinear spectroscopy.[2] Two prevalent techniques for detecting terahertz (THz) radiation are photoconductive sampling[3] and electro-optic sampling.[4–6] The former typically utilizes an antenna structure fabricated upon a semiconductor that has been processed to exhibit a short carrier lifetime. Although the carrier dynamics and circuit properties limit the response time, a recent study by Kono *et al.* has shown that these devices may exhibit usable response as far as ~20 THz.[7] Much of the work in developing broadband coherent detectors has focused on electro-optic sampling. This technique has been exploited to achieve responses to high frequencies.[8–10] To make full use of the essentially instantaneous (electronic) component of the material response requires transparency across the relevant spectral range and an adequate degree of phase matching. For established inorganic materials such as ZnTe, one is then constrained to using thin crystals, a procedure that is technically challenging and reduces detection sensitivity.

Poled polymers are attractive materials for broadband electro-optic detection because of their potentially large electro-optic coefficients,[11] relatively low dispersion between the optical and THz refractive indices,[12] and ease of processing for creating large-area thin films. It was demonstrated earlier that poled polymers are promising candidates for emitters and detectors of THz radiation.[6,12,13] However, the frequency response was limited to a few THz because of the relatively long laser pulses used in those studies.

In this Letter we describe the application of a poled polymer as a detection medium for ultrafast electromagnetic pulses. Using optical rectification in GaAs to generate wideband emission, we demonstrate the coherent detection in a poled polymer of radiation from the far infrared up to a frequency of ~33 THz. We accomplish this by copropagating the optical probe and baseband pulses through the poled polymer. In contrast to ZnTe, which exhibits a marked reduction in frequency response at ~17 THz for 30-μm-thick crystals owing to phase mismatch,[8] the poled polymer has shown no evidence of a similar reduction to at least 30 THz for an interaction length of ~80 μm.

The poled polymer detection medium was fabricated upon a fused silica substrate. A 200-nm-thick aluminum layer was initially deposited upon the substrate. As described below, this metal layer acted both as a poling electrode and as a mirror for the THz and optical probe beams. A 35-μm-thick copolymer that was capable of exhibiting nonlinear optical properties was then solution cast over the metal film. The copolymer used in this study was composed of 30 mol.% 2-*N*-[4-(4-nitrophenyazo) indolino] ethyl methacrylate (MA9) and methyl methacrylate. Its chemical structure, synthesis, polymer properties, and linear and nonlinear optical properties were described elsewhere.[14] In the fabrication process we deposited a top electrode over the polymer film. To effect dipolar alignment in the copolymer we placed the entire device in an oven and heated the sample to the glass-transition temperature of the copolymer (137 °C). After applying a poling field of 1.25 MV/cm between the metal electrodes we cooled the device to room temperature with the field in place. The top electrode was then removed. It should be noted that, on poling, the polymer remains completely amorphous, though it is rendered uniaxial with the *c* axis parallel to the poling field.

A schematic diagram of the experimental setup is shown in Fig. 1(a). We used a mode-locked Ti:sapphire laser as the optical source for generating and detecting the transient baseband pulses. The laser oscillator operated at a central wavelength of 820 nm with a repetition rate of 89 MHz and produced transform-limited optical pulses with a duration of

0146-9592/02/090775-03$15.00/0

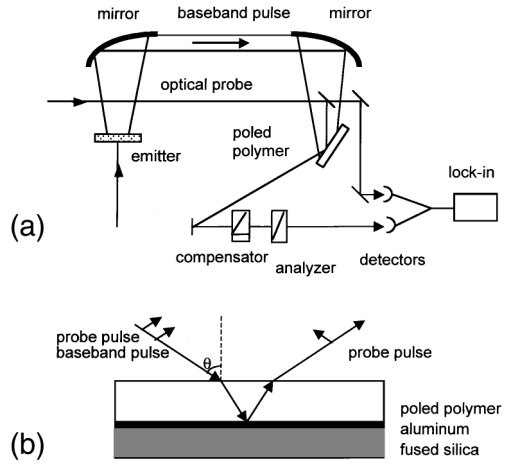

Fig. 1. (a) Schematic diagram of the experimental setup. Ultrashort baseband pulses are generated in ⟨110⟩ GaAs and detected in a poled polymer. (b) Expanded view of the poled polymer and beam propagation path. The optical probe and baseband pulses copropagate through the polymer with an external angle of incidence θ. The baseband pulse is p polarized, whereas the optical probe is polarized at 45° with respect to the plane of incidence.

23 fs. The optical pump beam, with an average power of 380 mW, was focused onto a 280-μm-thick ⟨110⟩ GaAs wafer at normal incidence. p-polarized baseband pulses were generated by optical rectification and transmitted through the wafer. They were then collected and refocused into the polymetric electro-optic medium by two off-axis paraboloidal mirrors.

For detection of the ultrashort baseband pulses we employed a reflection geometry, as shown schematically in Fig. 1(b). Inasmuch as the polymer was poled normal to the plane of the film, an oblique incidence geometry was necessary for access to the nonzero components of the electro-optic tensor. The addition of a bottom metal reflecting layer allowed us to double the interaction length for the optical probe and baseband pulses. These beams were incident upon the polymer device at an external angle of $\theta = 60°$. The polarization state of the 20-mW probe beam was altered from its initial 45° polarization. The corresponding phase retardation was measured with balanced photodiodes in a differential detection scheme.[15]

As stated above, the poled polymer exhibits the properties of a uniaxial optical medium. The magnitude of the birefringence is directly related to the dipole moment of the chromophore and the poling parameters. For the experimental parameters given above, the measured refractive indices at 820 nm were $n_e = 1.665$ and $n_o = 1.646$ for the extraordinary and the ordinary waves, respectively. The static phase shift difference Γ_{sp}, caused only by the anisotropy in the polymer between the s- and p-polarized waves, is given by

$$\Gamma_{\mathrm{sp}} = \frac{4\pi d}{\lambda}\left[(n_o{}^2 - \sin^2\theta)^{1/2} - \frac{n_o}{n_e}(n_e{}^2 - \sin^2\theta)^{1/2}\right]. \quad (1)$$

Here d is the thickness of the medium and λ is the wavelength of the probe beam. Thus, in our experiment, there is a static phase shift difference of 1.25π, which can be offset with a compensator.

We now turn our attention to the phase retardation induced in the optical probe beam by the ultrafast baseband pulses. In the limit of low poling fields for the polymer film we make the simplifying approximations that $n = n_e \approx n_o$ and $r_{33} \approx 3r_{13}$, where r_{13} and r_{33} are components of the electro-optic tensor.[16] It can then be shown that the field-induced phase retardation, $\Delta\Gamma_{\mathrm{sp}}$, is given by

$$\Delta\Gamma_{\mathrm{sp}} = \frac{4\pi d r_{33}}{3\lambda n_T}\,\frac{n^2 \sin^3\theta}{(n^2 - \sin^2\theta)^{1/2}}\,E_T\,, \quad (2)$$

where n_T is the refractive index of the polymer at THz frequencies and E_T is the magnitude of the baseband pulse.

The temporal waveform of the baseband electric field generated by optical rectification in GaAs and detected by the electro-optic effect in the poled polymer is shown in Fig. 2. The waveform exhibits a chirped oscillatory tail that is similar to that observed in previous studies with GaAs THz emitters.[8] The corresponding amplitude spectrum, shown in Fig. 3, demonstrates a detection capability of the poled polymer that extends beyond 30 THz. The decrease in the amplitude spectrum from 7 to 10 THz arises from absorption of the optically rectified field by the reststrahlen band of GaAs. We attribute the attenuated response of the spectrum beyond ~33 THz

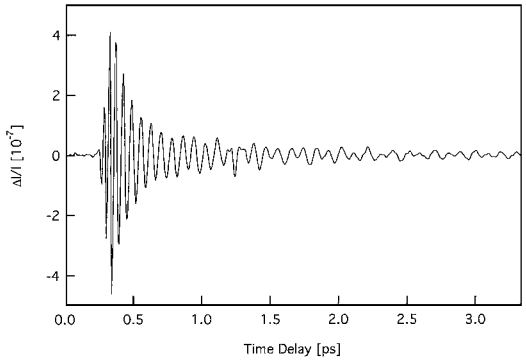

Fig. 2. Temporal waveform detected by electro-optic sampling. The shortest oscillation period is ~33 fs.

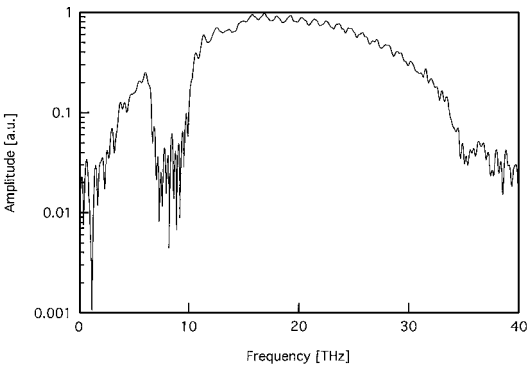

Fig. 3. Amplitude spectrum of the temporal waveform shown in Fig. 2.

largely to absorption in the poled polymer. We do not have infrared absorption data on the MA9 chromophore. However, it is similar chemically to Disperse Red Dye #1, which has been well studied. The latter chromophore, as well as methyl methacrylate, has numerous absorption lines from ~32 to ~48 THz.[17] Also, a spectral modulation with a period of ~1.1 THz is evident in Fig. 3. It is due to multiple internal reflections of the baseband pulse within the polymer. It is important to note that, for frequencies between the two absorption bands mentioned above in a band that is nearly 20 THz wide, the amplitude spectrum is relatively flat. In terms of the detector response, the flat response can be assumed to extend from low frequencies to beyond 30 THz.

The issue of phase matching is critically important for obtaining broad-bandwidth detection capability.[18] Specifically, it is necessary to match the optical group index ($n_g = c/v_g$, where v_g is the group velocity and c is the speed of light) to the refractive index of the baseband pulse. This issue is complicated by anisotropy in the dielectric properties of the medium. Thus we need to ensure that the baseband pulse travels in phase with both components of the probe beam. Although we have not been able to measure the linear optical properties of the poled polymer at THz frequencies, the chemical properties of this polymer are similar to those of a previously investigated material.[12] That material was found to have a refractive index of ~1.75 at 0.5 THz. Experimentally, the measured group indices for the s- and p-polarized components of the probe beams are $n_g{}^s = 1.751$ and $n_g{}^p = 1.756$, respectively, at 820 nm. Because these group indices are similar to the THz index, we would expect only a small phase mismatch. In addition, the anisotropy in the group indices leads to a temporal shift between the two polarization components of ~1.3 fs after the ~80-μm traversal through the polymer. Thus the group-velocity anisotropy, by itself, does not limit the detection bandwidth.

In conclusion, we have demonstrated the broadband detection of baseband pulses by electro-optic detection in a poled polymer. The frequency response of this detection scheme extends from the far infrared through much of the mid infrared (~30 THz). This high frequency limit appears to be restricted primarily by absorption in the poled polymer and the duration of the optical excitation pulses used in this study. Unlike inorganic crystals, however, polymers can be chemically modified to alter their linear and nonlinear dielectric properties. Thus we expect that a judicious choice of chromophore and host matrix will significantly extend the detection bandwidth.

H. Cao thanks W. Warren and R. A. Linke for financial support. T. F. Heinz acknowledges support from the U.S. Air Force Office of Scientific Research. A. Nahata thanks R. A. Linke for many stimulating discussions. A. Nahata's e-mail address is ajay@research.nj.nec.com.

References

1. A. Leitenstorfer, S. Hunsche, J. Shah, M. C. Nuss, and W. H. Knox, Phys. Rev. B **61,** 16642 (2000).
2. R. J. Richter, T. P. Petralli-Mallow, and J. C. Stephenson, Opt. Lett. **23,** 1594 (1998).
3. P. R. Smith, D. H. Auston, and M. C. Nuss, IEEE J. Quantum Electron. **24,** 255 (1988).
4. Q. Wu and X.-C. Zhang, Appl. Phys. Lett. **67,** 3523 (1995).
5. P. U. Jepsen, C. Winnewisser, M. Schall, V. Schyja, S. R. Keiding, and H. Helm, Phys. Rev. E **53,** R3052 (1996).
6. A. Nahata, D. H. Auston, T. F. Heinz, and C. Wu, Appl. Phys. Lett. **68,** 150 (1996).
7. S. Kono, M. Tani, P. Gu, and K. Sakai, Appl. Phys. Lett. **77,** 4104 (2000).
8. Q. Wu and X.-C. Zhang, Appl. Phys. Lett. **71,** 1285 (1997).
9. A. Leitenstorfer, S. Hunsche, J. Shah, M. C. Nuss, and W. H. Knox, Appl. Phys. Lett. **74,** 1516 (1999).
10. R. Huber, A. Brodschelm, F. Tauser, and A. Leitenstorfer, Appl. Phys. Lett. **76,** 3191 (2000).
11. Y. Q. Shi, W. P. Lin, D. J. Olson, J. H. Bechtel, H. Zhang, W. H. Steier, C. Zhang, and L. R. Dalton, Appl. Phys. Lett. **77,** 1 (2000).
12. A. Nahata, D. H. Auston, C. Wu, and J. T. Yardley, Appl. Phys. Lett. **67,** 1358 (1995).
13. A. M. Sinyukov and L. M. Hayden, Opt. Lett. **27,** 55 (2002).
14. C. Wu, K. Beeson, P. Ferm, C. Knapp, M. McFarland, A. Nahata, J. Shan, and J. T. Yardley, Mater. Res. Soc. Symp. Proc. **328,** 477 (1994).
15. J. A. Valdmanis, G. Mourou, and C. W. Gabel, Appl. Phys. Lett. **41,** 211 (1982).
16. K. D. Singer, M. G. Kuzyk, and J. E. Sohn, J. Opt. Soc. Am. B **4,** 968 (1987).
17. These data were provided by Sigma-Aldrich, Inc., Milwaukee, Wisc.
18. A. Nahata, A. S. Weling, and T. F. Heinz, Appl. Phys. Lett. **69,** 2321 (1996).