Coherent detection of freely propagating terahertz radiation by electro-optic sampling

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(Received 11 September 1995; accepted for publication 31 October 1995)

We report the demonstration of an electro-optic sampling technique that allows for the detection of freely propagating terahertz radiation. Coherent sampling is performed in a poled polymer device that is physically separated from the emitter. The poling electrodes in the sampling element are found to have an integrating effect on the incident terahertz field. The shot noise limited minimum detectable field in the polymer is 100 (mV/cm)/$\sqrt{\text{Hz}}$. We discuss methods by which the sensitivity may be significantly enhanced. © *1996 American Institute of Physics.* [S0003-6951(96)03502-7]

The use of ultrafast laser sources to generate and detect freely propagating pulses of coherent far-infrared radiation has stimulated significant interest in recent years. These coherent pulses have been used to examine ultrafast carrier dynamics in semiconductors[1] and to measure the far-infrared linear optical properties of a wide range of dielectric media.[2] The terahertz (THz) bandwidth pulses are typically generated by exciting radiative current transients in photoconductive media[2–4] or producing a nonlinear polarization via difference frequency mixing in nonlinear optical media.[5] To date, the only broadly applicable technique to coherently detect this radiation requires the use of synchronously gated photoconducting dipole detectors.[3] While these detectors exhibit excellent sensitivity, there is a strong speed versus sensitivity trade-off which is determined by the photoconductive response and antenna dimensions.[2]

Electro-optic sampling[6] is an attractive alternate approach for the detection of THz radiation. Significant advantages of this technique are the capability of detection that is limited primarily by the optical bandwidth and the ability to calibrate the detected electric field. These features have been exploited in extensive characterization of ultrafast electrical pulses propagating in transmission lines and circuits.[6–8] The technique has also been utilized in measurements where freely propagating THz radiation was generated and detected within the same device. Prominent examples of this include studies of Cherenkov radiation in $LiTaO_3$ (Ref. 9) and Bloch oscillations in GaAs/AlGaAs superlattices.[10]

In this letter, we demonstrate an electro-optic sampling technique that allows for the coherent detection of freely propagating submillimeter wave radiation. In this scheme, the emitter and detector are physically separated from one another by a large distance, making the system well suited for spectroscopic applications. As an electro-optic medium, we make use of a poled polymer. In contrast to traditional electro-optic media such as $LiTaO_3$,[9] polymers do not suffer from the complication of a large difference in the optical and far-infrared refractive indexes and absorption in the far infrared. Furthermore, the optical nonlinearity in poled polymers is expected to have negligible response time. In this work, electro-optic sampling is accomplished using a thin poled polymer film in a double pass geometry. The performance of this configuration, the influence of the poling electrodes, as well as possibilities for future experiments are discussed.

We fabricated the electro-optic sampling element (EOSE), shown schematically in Fig. 1, on an *R*-plane sapphire substrate. A high reflectivity dielectric coating centered at 800 nm was first evaporated onto one side of the substrate. Two coplanar 5 mm×5 mm×2000 Å thick aluminum pads separated by 50 μm, used for poling, were then photolithographically defined onto the coating. The polymer used in this study is composed of 20 mol % 2-*N*-[4-(4-nitrophenylazo) indolino] ethyl methacrylate (MA9) and methyl methacrylate (MMA), which we designate MA9:MMA. The chemical structure, synthesis, polymer properties, and linear and nonlinear optical properties are described elsewhere.[11] The polymer film in the EOSE was 10 μm thick and poled at its glass transition temperature with a field of 1.0 MV/cm.[12] A high resistivity silicon (>10 kΩ cm) hyperhemispherical lens[2] was attached to the sapphire substrate to focus the THz radiation into the electrode gap.

The experimental setup for generating and detecting THz radiation is shown in Fig. 2. A 76 MHz mode-locked Ti:sapphire laser operating at 800 nm served to generate and detect the transient submillimeter wave pulses. A collimated 2 mm

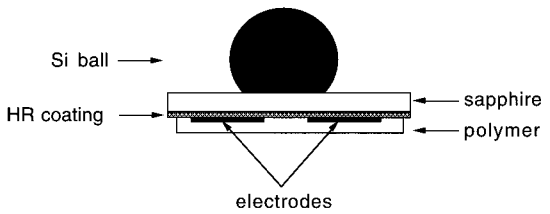

FIG. 1. Schematic drawing of the electro-optic sampling element.

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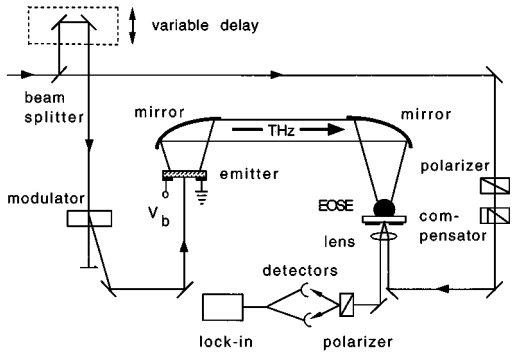

FIG. 2. Schematic drawing of the experimental setup used to detect freely propagating THz radiation.

diameter pump beam, with an average power of 400 mW, was chopped at 95 kHz and used to drive a large aperture photoconducting antenna.[4] The emitter was oriented so that the resulting THz electric field was parallel to the c axis of the poled polymer. Two off-axis paraboloidal mirrors were used to collimate and focus the THz radiation into the EOSE. The total separation between the emitter and detector was 60 cm.

The detection system employed a crossed polarizer arrangement with differential detection.[6] The 5 mW probe beam was optically biased at its quarter wave point by a Soleil–Babinet compensator and focused into the electrode gap of the EOSE. The reflected probe beam, split by the Wollaston prism, generated a quiescent current of approximately 1 mA in each detector.

In the small angle approximation, the double pass phase retardation can be related to the electro-optic coefficients r_{33} and r_{13} by

$$\Delta\Gamma = \frac{2\pi l}{\lambda}(n_e^3 r_{33} - n_0^3 r_{13})E \approx \frac{4\pi l}{3\lambda} n^3 r_{33} E, \tag{1}$$

where n_0 and n_e are the ordinary and extraordinary refractive indexes, respectively, l is the polymer thickness, λ is the wavelength of the optical probe, and E is the applied field. For relatively low poling fields ($\leqslant$1 MV/cm), $n_0 \approx n_e = n = 1.65$ and $r_{33} \approx 3r_{13}$.[13] The observed low-frequency (95 kHz) electro-optic coefficient r_{33} was 11 pm/V. This corresponds to a value of E_π, the field needed to cause a π phase retardation, of 12 mV/cm. For square wave amplitude modulation (chopping) of the electric field, it can be shown that the fundamental component of the rms signal current measured by a lock-in amplifier at the chopping frequency is $i_{\mathrm{sig}} = 2\sqrt{2} I_0 E(t)/E_\pi$, where I_0 is the quiescent current from each detector and $E(t)$ is the electric field. The total shot noise from both photodiodes is $i_{\mathrm{SN}} = \sqrt{4qI_0 B}$, where q is the electron charge and B is the detection bandwidth. In the shot noise limit, the minimum detectable field occurs when $i_{\mathrm{sig}} = i_{\mathrm{SN}}$, so that for a 1 Hz detection bandwidth

$$E_{\min} = E_\pi \sqrt{\frac{q}{2I_0}} \frac{(\mathrm{V/cm})}{\sqrt{\mathrm{Hz}}}. \tag{2}$$

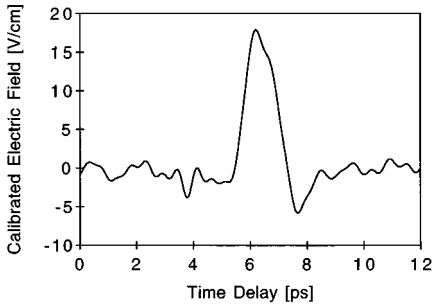

FIG. 3. Temporal wave form detected by electro-optic sampling.

Thus for a quiescent current of 1 mA, a minimum detectable field of 100 (mV/cm)/ $\sqrt{\mathrm{Hz}}$ is expected.

The temporal wave form of the THz electric field measured by the electro-optic effect in the polymer is shown in Fig. 3. Data were collected for an antenna bias of 300 V and averaged over ten scans. We calibrated the THz response of the EOSE by applying a 95 kHz, 5 V square wave across the 50 μm electrode gap. The procedure is based on the neglect of the dispersion in the electro-optic coefficients. This assumption is believed to be reasonable since the optical nonlinearity is essentially electronic in nature, as determined by comparing electro-optic and second harmonic generation measurements.[14] The peak THz field detected was thus $\sim$18 V/cm with a noise floor of 3 (V/cm)/ $\sqrt{\mathrm{Hz}}$. Our detection noise floor is thus nearly 29 dB above the shot noise limit and appears to be dominated by excess laser noise. Modulation of the pump beam at significantly higher frequencies should reduce this noise.[15]

The THz electric field radiated by the large aperture photoconducting antenna is expected to have a bipolar shape, such that total time integral is zero.[3] The observed shape of the detected wave form in Fig. 3, however, is unipolar. This difference arises from the influence of the electrode structure of the EOSE on the incident field. A similar effect was observed in the first description of photoconducting Hertzian dipole detectors.[3] The measured electric field in the polymer may be modeled by classical electromagnetic diffraction through a two-dimensional conducting screen with a narrow slit.[16] Such a model is appropriate since the electrodes are significantly larger than the wavelengths of interest. To first

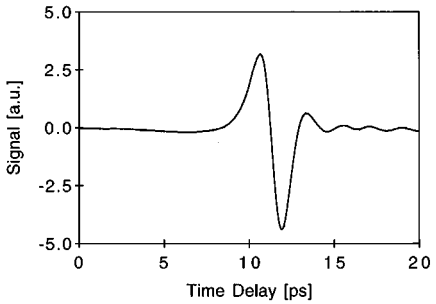

FIG. 4. Temporal wave form obtained with a 100 μm Hertzian dipole detector that preserves the bipolar nature of the incident THz field. The experimental conditions were identical to those used for the electro-optic sampling measurement.

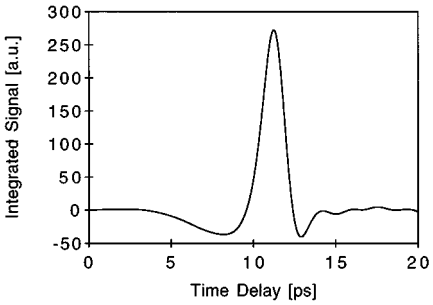

FIG. 5. Temporal integration of the wave form shown in Fig. 4.

order, the diffracted field (and corresponding electrode voltage) is proportional to the time integral $[\omega^{-1}E(\omega)]$ of the incident field. This is equivalent to the charging of the gap capacitance by the incident THz radiation. A detailed analysis of the frequency-dependent electrode gap voltage is beyond the scope of this letter and will be discussed in a future publication. However, an approximate expression for the voltage across the slit can be written as[3]

$$v_s(t)=\frac{1}{Z_0C_s}\int_{-\infty}^{t}dt'\ E_i(t'), \tag{3}$$

where Z_0 is the characteristic impedance of the medium and C_s, which has a weak frequency dependence, is the static capacitance of the electrode structure per unit length.

We demonstrate the integrating nature of the EOSE by using a photoconductive 100 μm Hertzian dipole detector that is known to preserve the bipolar nature of the incident field.[2,17] Figure 4 displays the detected wave form with the dipole detector substituted for the EOSE with all other experimental parameters unchanged. The time integral of this bipolar wave form is shown in Fig. 5. While the shape of this wave form and that from the EOSE (Fig. 3) are very similar, the FWHM time of the latter signal is smaller. To understand this, we consider the detector response of the EOSE (neglecting its integrating nature). The detection response time of the sampling system can be written as $\tau_s=\sqrt{\tau_{\rm pw}^2+\tau_{\rm it}^2+\tau_{\rm mr}^2}$. Here, $\tau_{\rm pw}$ is the FWHM optical pulse width of the probe beam and is equal to 160 fs. The interaction time of the optical pulse through the nonlinear medium $\tau_{\rm it}=2nl/c$ is 110 fs and the material response time $\tau_{\rm mr}$ is $<$10 fs.[9] Thus the overall FWHM detection response time $\tau_s<$200 fs, which is less than that of our dipole detector.

It should be possible to modify the sampling element so that the incident field is not perturbed. This can be accomplished by removing the electrodes after poling or by using an appropriate organic single crystal. It should be possible to obtain a large enhancement in sensitivity by performing the experiment in a transmission geometry. In this case, the THz field would again be normally incident on the EOSE. The probe beam would be incident at the phase velocity matching angle to permit copropagation with the THz beam, thus allowing for longer interaction lengths. Since the dispersion in the refractive index is low for these polymers, the velocity matching angle is relatively small.[18] We are currently investigating these ideas.

In conclusion, we have demonstrated an electro-optic sampling technique that allows for the coherent detection of freely propagating THz radiation. This geometry is amenable to applications such as THz time-domain spectroscopy. The shot noise limited minimum detectable field in the polymer is 100 (mV/cm)/ $\sqrt{\rm Hz}$. With appropriate changes in the sampling element and geometry, it should be possible to significantly enhance the detection sensitivity while maintaining an extremely fast response.

The authors are grateful to Binbin Hu of AT&T for suggesting the use of poled polymers for this application. The authors also thank Aniruddha Weling for fabrication of the dipole detector and many helpful discussions. This research was supported by the Air Force Office of Scientific Research under Grant No. F49620-92-J-0036.

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