Progress:

In collaboration with Allied Signal Inc., we have investigated the ultrafast electro-optic response and sensitivity of a poled side chain polymer film by a femtosecond electro-optic sampling technique. A 760 fs rise-time electrical transient was observed corresponding to a 460 GHz bandwidth. The measurement was made in a high speed coplanar transmission line structure using the electro-optic polymer as the dielectric. This result has important implications for the use of electro-optic polymers for probing signals in multi-chip module interconnection circuits. A paper describing this work (preprint enclosed) has been accepted for publication in Applied Physics Letters.

A second area of activity during the past quarter has been the establishment of a new experimental facility which will be used for the high speed testing of materials and devices for applications to multichip modules. A new titanium-sapphire mode-locked laser system was installed in September and is now fully operational. It provides a source of optical pulses...
having a sub-picosecond duration and an average power of 1 Watt over a wide tuning range in the infrared region of the spectrum. Together with our high speed optoelectronics measurement capability, we will use this laser system to generate and detect the high speed electrical signals that will be used for probing multichip modules. The measurement of the response of the electro-optic polymer structure described in the previous paragraph was made with this laser in our laboratory.

We have used the funds ($100,000) provided with the first year of this contract to establish this new measurement capability. New equipment has been purchased to provide a state-of-the-art system for high speed electrical measurements. This includes a computer-controlled translation stage for precision delays, optical components for beam handling, a lock-in amplifier for sensitive detection, a microscope for sample inspection and positioning, a diamond saw for sample preparation, and other components needed for high speed measurements.

**Plans for 1992:**

Our plans for the next year include an extension of our current work on the use of electro-optic polymers for optical probing of high speed interconnection circuits. The next experiment we intend to make will be to fabricate an optical modulator from this material and evaluate its performance for possible applications to high speed optical interconnections. Following that we will be fabricating some thick polymer films which we can use for precise measurements of the dielectric constant and loss over a spectral range from dc to 500 GHz. We will also be doing some preliminary experimental measurements with photoconducting polymers to evaluate their properties for potential applications to high speed interconnection circuits. An important part of our activity next year will be to establish our fabrication and measurement capability in the areas of polymer films and interconnection circuits. We will also be working closely with our partners in industry to supply materials and test jigs for high speed testing.
Publications:


Femtosecond response of electrooptic poled polymers

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ABSTRACT

We investigate the ultrafast electrooptic response and sensitivity of a poled side chain polymer film via the electrooptic sampling technique. A 760-fs risetime electrical transient is observed corresponding to a bandwidth of 460 GHz. We believe this to be device limited and not due to limitations in the speed of response of the polymer.
The successful integration of novel polymers with semiconductor devices promises to offer a powerful new class of electrooptic devices. Polymeric materials provide for a unique set of chemically-tailorable material properties including dielectric constant, refractive index, transparency, and optical nonlinearity. The fabrication of polymeric devices such as passive waveguides and electrooptic modulators is based on spin-casting and UV photodelineation techniques.\textsuperscript{1} Nonlinear guest/host polymers are formed by introducing a small concentration of nonlinear optical moieties into a polymer solution.\textsuperscript{2} Polymers with nonlinear main chain or side group charge transfer chromophores are more stable and allow for a higher concentration of the nonlinear moiety, and thus a larger nonlinear effect. Upon electric field or corona poling to create a non-centrosymmetry in the material, optical nonlinearities on the order of those found in inorganic crystals such as LiTaO\textsubscript{3} have been demonstrated.\textsuperscript{1,3} Key interest is placed in the ability of polymer devices to provide higher bandwidths than conventional optical electronics. Electronic motions occur on a femtosecond time scale, thus extremely high bandwidths may be possible in these polymers if vibrational dynamics do not play an important role in their nonlinear response.\textsuperscript{4} Recently, Girton et al. have tested a polymer based Mach-Zehnder modulator up to 20 GHz by beating together two microwave signals and detecting modulation at the difference frequency.\textsuperscript{5} Thackara et al. have recently formed a poled guest/host polymer film on top of a planar transmission line on GaAs and shown that microwave signals of at least 20 GHz can be detected via electro-optic sampling of the polymer.\textsuperscript{6} In this letter, we demonstrate a nonlinear response bandwidth of 460 GHz in a poled sidechain polymer film. We believe this to be the highest polymeric electrooptic response bandwidth reported to date.
Our technical approach is based on the electrooptic sampling technique of Valdmanis and Mourou. These workers place an electro-optic crystal (LiTaO$_3$) above a planar transmission line, then use an ultrafast laser pulse as a narrow temporal window to sample the birefringence induced in the crystal by an electrical signal propagating along the transmission line. The fastest (< 1 ps) electrical transients are generated by illuminating a photoconductive switch integrated into a transmission line, also with an ultrafast laser pulse. Thus, a common embodiment of the electrooptic sampling technique is the use of an excitation pulse to create an electrical transient and a sampling pulse to probe the resulting birefringence induced in an electrooptic material.

The electrooptic polymer is prepared as in reference 9. Two planar electrode pads separated by a gap are fabricated on a quartz substrate. A polymer solution is applied over the electrode gap. The sample is baked to remove the residual solvent, poled at its glass transition temperature $T_g$, then cooled to room temperature. The polar axis of the polymer is normal to the electrode gap. In the presence of a voltage $V$, applied across the electrode gap, a light beam focussed through the polymer and linearly polarized at 45° to the polar axis, will experience a phase retardation, $\Gamma$. In the small angle approximation, this phase retardation can be related to the electrooptic coefficients $r_{33}$ and $r_{13}$ by

$$\Gamma = \frac{\pi d V}{\lambda d} (n_e^2 r_{33} - n_o^2 r_{13})$$

Eq. (1)

where $n_o$ and $n_e$ are the ordinary and extraordinary refractive indices of the polymer, $\lambda$ is the wavelength of the light, $l$ is the polymer thickness, and $d$ is the electrode gap width. For
relatively low poling fields \((\leq 1 \text{ MV/cm})\), \(r_{13} = r_{33}/3\), and thus, assuming
\[ n_o = n_s = n \text{, Eq. 1 reduces to} \]
\[ \Gamma = \frac{2\pi v I n^3 r_{33}}{3\lambda d} \quad \text{Eq. (2)} \]

By using a polarizer/compensator/analyzer combination, \(\Gamma\) can be converted into an intensity modulation of the beam and detected by a photodiode.

Our experiment centers around the integration of such a poled polymer thin film into a photoconductive switch - transmission line device (Figure 1). Two device configurations are used- one to achieve a subpicosecond risetime (configuration I), and the other to obtain the best possible noise floor (configuration II). In both, electrical transients are created via the excitation of a photoconductive switch gap in intrinsic GaAs using 830-nm, 100-fs exciting pulses from a hybridly mode-locked ultrafast dye laser. These transients are then propagated down a symmetrical coplanar waveguide (CPW) transmission line into the region of the transmission line containing the polymer. The polymer is oriented with its polar axis parallel to the substrate and transverse to the direction of electric field propagation. A much weaker sampling pulse is split off from and, via a translation stage, variably time-delayed relative to the excitation pulse. The sampling beam is focussed through the polymer. It is polarized at 45° to the transmission line and therefore to the polar axis of the polymer. The beam is recollimated and aligned into a Soleil-Babinet compensator, balanced to give circularly polarized light, and finally split into two orthogonally polarized beams and detected by two amplified photodiodes. In the detection optics
(polarizer/ compensator/ analyzer), this phase rotation is converted into equal intensity modulations of opposite sign in the two output beams. This scheme allows us to use differential detection of the two photodiode signals to effectively double the magnitude of the resulting signal while subtracting out common mode noise.

As the electrical waveform passes the probe beam in the polymer, the induced phase retardation is sampled at a series of delay times between the pump and probe beams. Noise reduction is achieved through lock-in detection. The bias voltage on the photoconductive switch is modulated with a 100-kHz, 13-Volt square wave, which is also used as the lock-in reference.

In this experiment, the polymer used is a copolymer of 60 mole percent methylmethacrylate (MMA) and 40 mole percent methacrylate-bound disperse red 1 dye (MA1), which has been examined for its electrooptic effect by a number of groups. In configuration I (Figure 2a) a 6-μm thick polymer film is spin cast on a separate planar structure consisting of 0.2-μm thick, 3 mm x 5 mm aluminum electrode pads. The pads are separated by a 50 μm gap. The polymer contained in this gap is poled to 1 MV/cm at 134 °C, resulting in an \( r_{33} \) of 14 pm/volt, as measured separately using an 810 nm diode laser with detection optics described above. A small piece of this sample, centered around the poled area and containing a portion of the electrodes is cleaved out. A 0.5 μm thick aluminum CPW containing a 5-μm wide switch gap is fabricated separately on a GaAs substrate. Using a thin layer of optical epoxy, the poled polymer is positioned over the switch gap on the CPW such that the poling electrodes straddle the 20-μm center conductor and 10-μm slots, and overlap the ground planes. The weak probe beam is focussed through the polymer film with a 10 x microscope objective onto one of the transmission
line slots, reflected back off the GaAs through the objective, then steered by a prism into the
detection section of the experiment. Care is taken not to completely straddle the transmission line
slot with the probe beam as this could cause a partial short circuit. Figure 3 shows the results
of a measurement in which the excitation and sampling beams are separated by 120 μm. In this
experiment, we observe a 10% - 90% risetime in the electrical transient of 760 fs, corresponding
to an electrooptical response bandwidth of 460 GHz. This risetime is probably limited by the
device geometry rather than by the electrooptic polymer: similar experiments with a larger
geometry CPW (35-μm center conductor, 15-μm slots) yield slower risetimes of about 1.1 ps.

An estimate of an electrical transient’s magnitude is obtained by switching the modulated bias
voltage to the opposite side of the switch gap. In this way, the phase retardation induced by the
13-V, 100-kHz square wave can be measured after every adjustment of the excitation or
sampling beams. Preceding the measurement in Figure 1, we measure a 500 μradian phase
retardation signal for the 13-V square wave. We then infer that the phase retardation of 16
μradians on the positive side of the step function in Figure 1 corresponds to 0.4 V. Taking into
account the lock-in bandwidth and the signal-to-noise ratio, we calculate the noise floor of the
overall experiment with this polymer film to be 50 mV/√Hz.

In configuration II, to achieve a better noise floor, a 60-μm thick dot of polymer is formed
directly onto a CPW fabricated on a quartz substrate (Figure 2b). The polymer is dried slowly
to prevent cracking and poled on the CPW structure to 0.5 MV/cm. A CPW with a switch gap
is fabricated separately on GaAs, and the two structures are wirebonded together with 1-mil
diameter Al wire. The probe beam is focussed through the polymer and quartz substrate and is then recollimated and sent into the detectors. For this geometry, the excitation and sampling pulses are separated by about 1.5 mm and the wirebond-limited risetime is about 6 ps. Because of the increased thickness of the polymer and its intimate contact to the transmission line, an improved noise floor of 17 mV/$\sqrt{\text{Hz}}$ is observed even though the polymer is poled to only half the voltage used in configuration I.

We have successfully used a copolymer of methacrylate and methacrylate-bound red dye 1 as the electrooptic material in an ultrafast sampling experiment. We observe a device limited risetime of 760 fs which demonstrates that polymers should provide useful bandwidths of at least 460 GHz. This demonstrates the viability of using polymers as the active media in high bandwidth electrooptic devices. In electrooptic sampling, because of a polymer’s low dielectric constant, a polymer probe placed in close proximity to a device under test should cause less capacitive loading than an inorganic crystal. Also, the small thickness of a polymer film compared to an inorganic crystal allows for shorter optical transit times of the sampling beam through the electrooptic material and therefore finer temporal resolution.

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References


Physics, 183, Springer Verlag NY, 1987


FIGURE CAPTIONS

Figure 1. Experimental apparatus schematic for polymer configuration I. The sampling beam makes two passes through the polymer, as it is reflected off of the GaAs substrate. It is then analyzed in the detection optics. For polymer configuration II, the sampling beam is transmitted through the quartz substrate into the detection optics, thus making only one pass through the polymer.

Figure 2. Detail schematics of polymer configurations for electrooptic sampling. (a) Side and top views of configuration I. (b) Top view of configuration II.

Figure 3. A 760-fs risetime electrical transient measured using methacrylate bound red dye 1 copolymer in an electrooptic sampling experiment.