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ELECTRO-OPTIC BEAM STEERING USING NON-LINEAR ORGANIC MATERIALS

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This report summarizes Hughes Research Laboratories efforts on evaluation of non-linear optical polymer materials supplied by State University of New York (SUNY), Buffalo, for potential application to the Hughes electro-optic beam deflector device. Evaluations include electro-optic coefficient, response time, transmission, and resistivity. Electro-optic coefficient measurements were made at 633 nm using a simple reflection technique. The samples measured in the range of 1-2.5 pm/V, which is too low to be of practical use in the Hughes beam deflector device.

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Section 1
Introduction

Electro-optical (EO) beam steering offers improved pointing performance, speed, and reliability, as well as advantages in space and power requirements, over conventional gimbal mechanical systems for laser radar and laser communications systems. Hughes Aircraft Company has previously developed a non-mechanical beam deflector device based on nematic liquid crystals. The advantage of liquid crystals in this application is that they have a large, electrically controllable birefringent anisotropy (an effective electro-optic coefficient of 5000 pm/V), and they are readily available and extensively researched. The predominant drawback is the slow time response (tens to hundreds of msec) of the liquid crystals.

The effort reported herein was aimed at evaluation of currently available non-linear organic (NLO) materials for their potential use in the Hughes beam deflector device. The polymer materials were produced at the State University of New York at Buffalo (SUNY/Buffalo), under direction of Prof. Paras Prasad and Dr. Yaroslaw Zieba. These organic and organo-metallic compounds can have extremely fast response times, ranging from microseconds down to femtoseconds, but usually have relatively small voltage-controlled index of refraction and associated electro-optic coefficient. The hope of this program, and concurrent programs within SUNY and other labs, is that material development would produce candidate samples that could be considered for this application.

It is appropriate to give a brief review of the Hughes beam deflector device. As shown in Figure 1, the device consists of a series of optical phase modulators with width equal to 5 μm (up to 20 μm) and length equal to the size of the active aperture (several mm, up to 10s of cm). Each modulator contains an electro-optic (EO) material sandwiched between a transparent counter electrode on one substrate and resistive coating on the opposing substrate. By placing voltages on the thin (0.5 to 1.0 μm) electrodes that are underneath the resistive coating, an electric field is created across the EO material which produces a change in the refractive index, resulting in a relative electro-optical path length change or phase shift in the modulator. Two electrodes per modulator are used to create a voltage gradient across the resistive coating, which produces a linear phase gradient (or blaze) across the modulator (see Figure 2). By properly selecting the voltages at each modulator, the resulting phase gradients can be coherently configured together to steer the beam in a desired (one-dimensional) direction. The optional mirror shown in Figure 1 causes the incident wave to travel through the EO material twice, resulting in a phase shift twice that of a single pass geometry. This reflective geometry reduces the required thickness of the EO

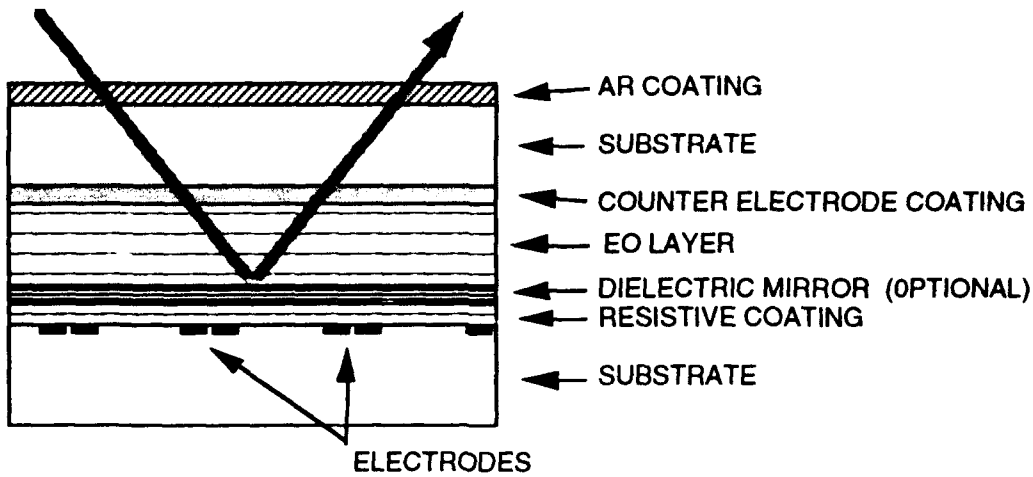


Figure 1. A cross-section of the Hughes beam deflector.

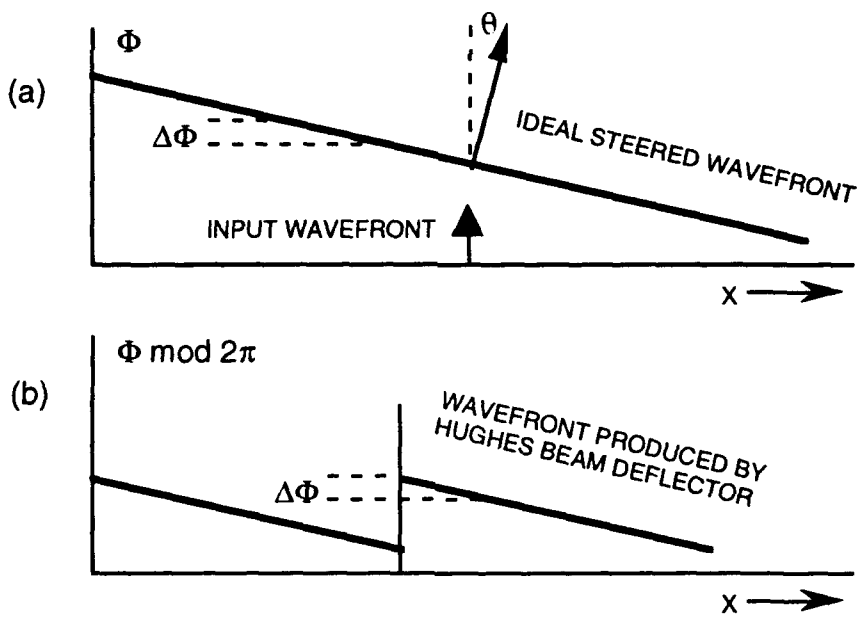


Figure 2. Wavefront of deflected beams. (a) Wavefront of an ideal plane wave propagating in the q direction. (b) Wavefront produced by the Hughes beam deflector.

material and reduces the level of the electrode voltages. Two-dimensional beam steering is accomplished by cascading two devices with orthogonal electrode geometries.

There are several requirements which an EO material must satisfy for practical use in the Hughes beam deflector. These include large voltage-controlled index of refraction, low optical absorption, fast response time, material stability, and compatibility with large scale fabrication. As seen below, it does not appear that sufficient advances have been made on high EO material development to enable practical application to the Hughes beam deflector device. The magnitude of the change in the index of refraction is very important because it affects the thickness of the EO layer, which is also related to the required applied voltage and the distribution of electric fields in the device. The amount of phase shift required for each modulator is a function of the maximum deflection angle, the width of each modulator, and the wavelength. For a maximum deflection angle of $\pm 20°$ and a wavelength of 532 nm, the phase shift required for each modulator is 8.5π for a 5 μm wide modulator, and is 28π for a 20 μm wide modulator. For example, with liquid crystals having maximum Δn of 0.2 the required thickness of the LC is in the range 5.5 to 18.5 μm for a reflective device to meet the above phase shifts.

Section 2
Non-linear Organic (NLO) Polymer Materials

Non-linear organic (NLO) materials are currently under development at the State University of New York at Buffalo (under direction of Prof. Paras Prasad, SUNY/Buffalo Chemistry Dept.). Measurement samples from synthesized material were prepared by Dr. Jarek Zieba (SUNY/Buffalo) for our evaluation of their potential use in the Hughes beam deflector device. Appendix A has a description of the polymer material developed for this program, and how it was prepared by researchers at SUNY/Buffalo. The polymer material was provided to HRL coated on 1" × 3" glass substrates with an indium tin oxide (ITO) conductive layer on the glass under the polymer. The polymer was corona poled by SUNY, and measured by HRL within 1 to 3 months after this poling (with no in-situ poling).

SUNY supplied Hughes with two separate batches of polymer material samples during this effort. Coating uniformity was fairly good, but typically thinner in the middle and thicker toward the edges. It is not clear that polymer layers with adequate uniformity over large areas (10s of cm diameter) could be practically achieved for large area beam deflectors. Our measurements were confined to the central (several mm diameter) thinner and more uniform region of the substrates.

Transmission of the polymer layer (first batch) measured in a spectrophotometer is shown in Figure 3. The yellow-orange tint of the electro-optic material is due to strong absorption at blue-green wavelengths. Note that this material is not a good candidate for use at 532 nm wavelength because of this optical absorption below about 600 nm.

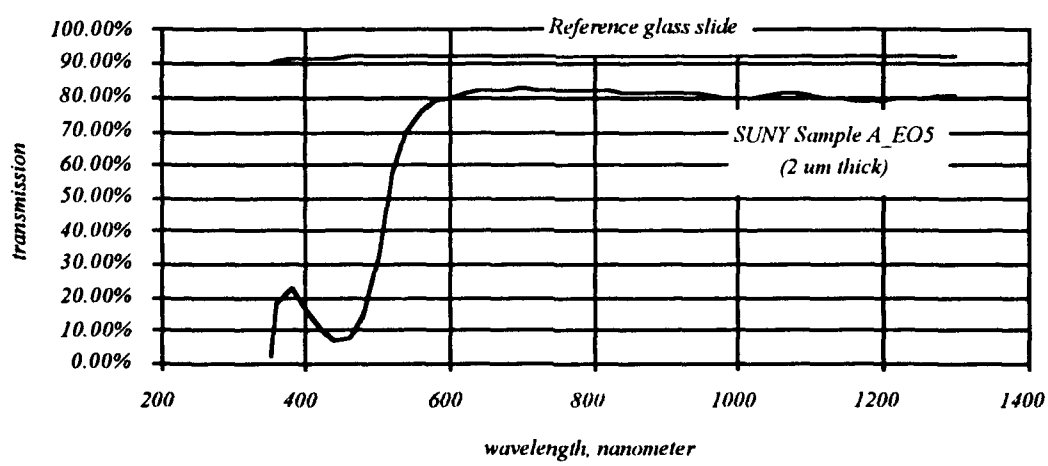


Figure 3. Spectrophotometer transmission scan of sample from first batch of SUNY non-linear organic polymer.

We encountered difficulties in making the electro-optic coefficient measurements on samples in the first batch due to unexpectedly low EO modulation. In discussions with SUNY, it was postulated that the protective polymer overcoat may have hindered the corona poling process and resulted in a weak poling with reduced stability. Although this clear layer did not attenuate the transmission, the strength of the corona poling field may have been reduced, resulting in less effective poling of the active polymer. Thus, by the time of the optical measurements the EO effect was substantially reduced.

Additional samples of the same polymer, but without using the protective overcoat, were then prepared and poled at SUNY. The samples from this second batch were successfully used for the electro-optic coefficient measurements described in the following section. The poling conditions were intentionally adjusted to achieve improved stability at the expense of reduced EO effect. The samples were corona poled under mild conditions at 140°C. The distance between corona wire and sample surface was 9.5 mm and poling voltage was 4.25 kV. Because the new samples were much thicker (measuring 28-30 μm versus the previous 2 μm) we ran another transmission scan with the result shown in Figure 4. Note the slight absorption between 600 and 900 nm which was not observed in the thinner samples. This absorption is not a hinderance to the EO measurements, but could be a factor for use in multipass optical arrangements for wavelengths in this 600-900 nm range. Again, the sample could not be used for optical wavelengths below about 550 nm.

The polymer electrical resistivity was determined by measuring the current leakage between a 41 mm^2 aluminum contact pad (the mirror/counterelectrode) on one side and the uniform ITO layer (the other electrode) bounding the polymer thin film. At 20 V_{DC} the current was approximately 10 picoamps. Thus, the polymer resistivity is estimated to be about 3×10^{14} $\Omega \cdot cm$ — a value which poses no anticipated problems for the beam deflector device application.

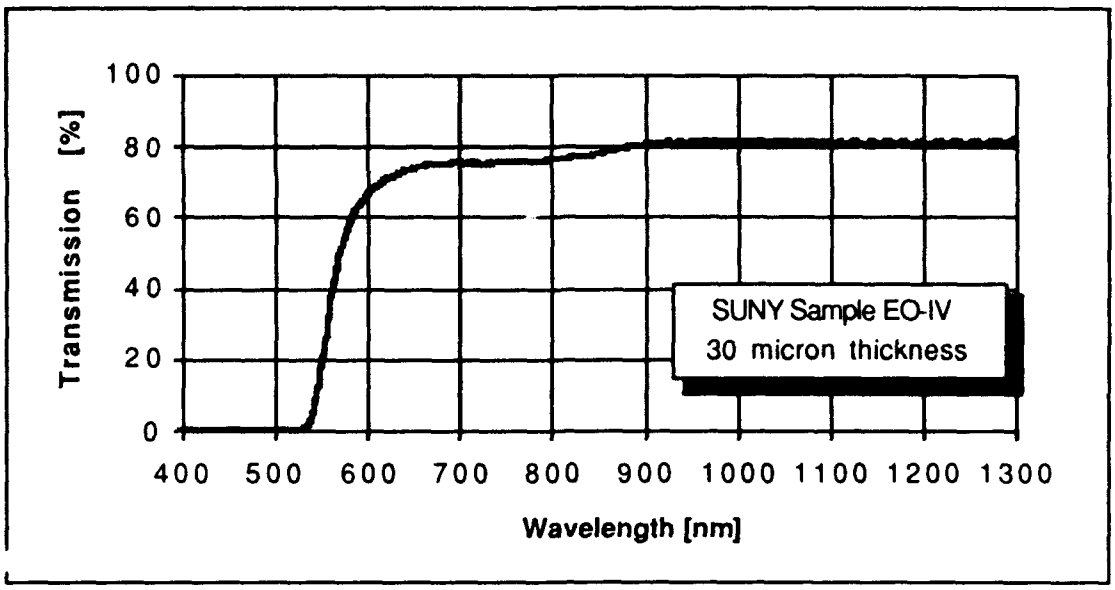


Figure 4. Spectrophotometer transmission scan of sample EO-IV from second batch of SUNY NLO polymer.

Section 3
Electro-Optic Coefficient Measurement Setup

We followed the technique described by Teng and Man[1] and Schildkraut[2] for measurements of the electro-optic coefficient, because the method is simple and the configuration is similar to our device application. The optical layout of our experimental setup is shown in Figure 5. The input laser beam (HeNe at 633 nm) is incident onto the sample at angle θ to the normal. The beam is transmitted through the glass, the transparent indium tin oxide (ITO) electrode, and the electro-optic polymer layer, and then reflected back out into air by a metal mirror (counterelectrode). The input beam polarization is oriented at 45° to the plane of incidence so that the s-wave and p-wave components are equal in magnitude. The reflected beam is passed through an adjustable wave plate and an analyzer at cross orientation to the input polarization, before entering the detector.

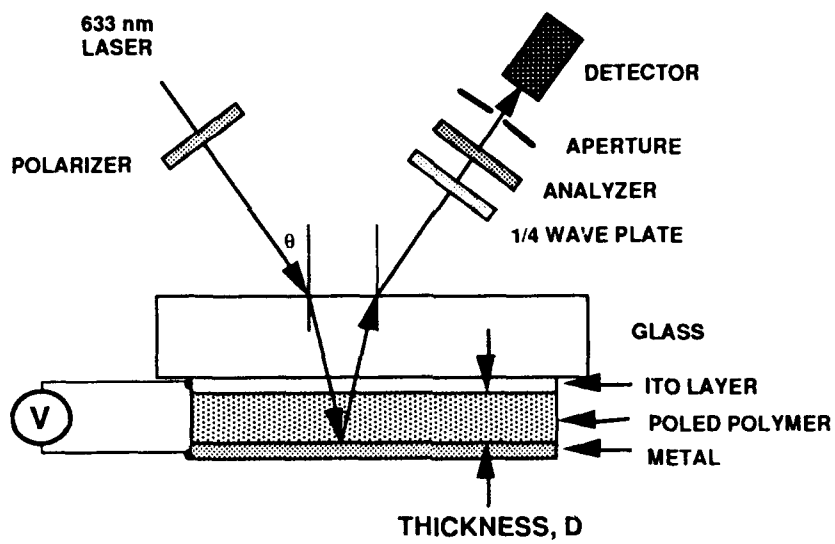


Figure 5. Optical setup of reflection technique for measuring the electro-optic coefficient.

In this configuration, the output beam intensity through the cross analyzer is a sinusoidal function of the phase retardation Ψ_{sp} between the s and p waves:

$$I = 2I_o \sin^2\left(\Psi_{sp}/2\right),$$

where I_0 is the mid-intensity level. The beam polarization as it enters the analyzer goes from linear polarization at $\Psi_{sp} = 0°$ to elliptical at $\Psi_{sp} < 90°$ to circular at $\Psi_{sp} = 90°$. The output

intensity is maximum for $\Psi_{sp} = 180°$, where the input beam polarization is rotated parallel to the transmission axis of the analyzer.

When a modulation voltage $V = V_m \sin(\omega t)$ is applied across the poled polymer layer there is a concomitant modulation in the phase retardation between the s and p waves $\delta\Psi_{sp}$ due to the linear electrooptic effect:[3]

$$\delta I \equiv I_m = I_o \sin(\Psi_{sp})\delta\Psi_{sp},$$
$$\delta\Psi_{sp} = \delta\Psi_p - \delta\Psi_s \equiv \Gamma_m \sin(\omega t).$$

A $\lambda/4$ plate is used to bias the total phase retardation to $\Psi_{sp} = 90°$, so that there is an approximate linear relationship (for small modulation, as in our case) between the measured intensities and the EO modulated phase:

$$I_m / I_o \cong \delta\Psi_{sp}.$$

The amplitude of the EO modulated retardation, Γ_m, is related to the input beam angle of incidence θ, the polymer film thickness d, and the poled polymer principal indices of refraction $n_\perp$ and $n_\parallel$ (referenced to the poling direction). In the linear Pockels effect the modulated indices are related to the elements of the electrooptic tensor r_{ij} by

$$\delta n_\perp = \frac{1}{2} n_\perp^3 r_{13} \frac{V_m}{d},$$
$$\delta n_\parallel = \frac{1}{2} n_\parallel^3 r_{33} \frac{V_m}{d}.$$

The derivation of Γ_m requires careful evaluation of the optical properties and reflection and refraction of light incident on uniaxial absorbing media.[4] This derivation has been discussed in previous publication.[5]

The experimental values of the electro-optic coefficients, r_{33}, are calculated from the following equation using the measured beam intensity and modulation, the incident optical beam angle and the applied modulation voltage:[6]

$$r_{33} = \frac{3\lambda I_m}{4\pi n^2 V_m I_o} \frac{(n^2 - \sin^2\theta)^{3/2}}{(n^2 - 2\sin^2\theta)} \frac{1}{\sin^2\theta},$$

λ = measurement wavelength (633 nm);
n = index of refraction (1.9);
θ = angle of incidence (45°);
V_m = modulation voltage;
I_m = modulation intensity;
I_o = midpoint intensity.

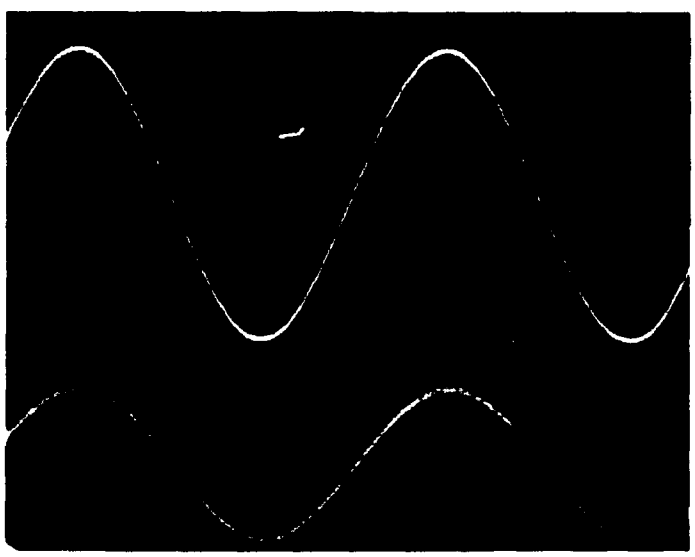
This formula uses the assumptions that $n_\perp \approx n_\parallel \equiv n$, and that $r_{33} = 3 \cdot r_{13}$. The neglect of poling birefringence[7] is reasonable in this case because of the mild poling and small induced birefringence. The commonly assumed relation between the electro-optic coefficients is also justified here due to the weak poling.

Two other effects which are not accounted for in the above expression are interfacial reflections[8] at the air-glass and glass-polymer interfaces, and electrochromic effects[9] associated with electronic-state absorption resonances. The interference from unwanted reflections is largely minimized by use of an aperture at the detector to block these contributions. The error due to neglecting interference from the reflection at the glass-polymer interface is not significant for the purpose of this study. Electrochromic effects are not expected to be important at the 633 nm measurement wavelength, but should be a consideration at wavelengths below 600 nm where material optical absorption is significant.

Section 4
Electro-Optic Coefficient Measurement Data

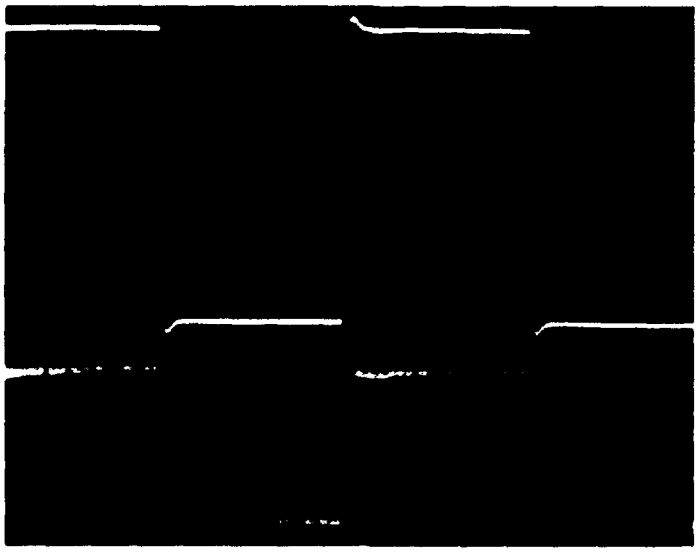
The samples were measured at two modulation voltages (200 Vp-p and 400 Vp-p) and at four modulation frequencies: 0.5 KHz, 1 KHz, 10 KHz, and 100 KHz. Measured r_{33} values were independent of the modulation voltage as expected, and only diminished slightly with increasing modulation frequency. Three separate locations (labeled a, b, and c) were tested on each 1" × 3" sample.

The optical modulation signal from sample E0-IV displayed on an oscilloscope is shown in Figures. 6(a,b). In both figures the upper scope trace is the 200 V_{p-p} modulation voltage at 10 kHz, and the lower trace is the corresponding modulated optical signal. The optical response time is estimated to be less than a few microseconds. At a modulation frequency of 100 kHz, a measureable phase shift (of the order 40 to 50 degrees) is observed between the drive voltage waveform and the modulated optical signal. It was not determined whether this phase delay is an artifact of the experimental setup or a real effect introduced by the polymer material response.



Sin wave modulation voltage [20 μs/div]

Figure 6(a). SUNY Sample E0-IV. Upper trace: 200 V_{p-p} @10 KHz modulation voltage. Lower trace: modulated optical signal (2.2 pm/V @ 633 nm).



Square wave modulation voltage [20 μs/div]

Figure 6(b). SUNY Sample E0-IV. Upper trace: 200 V_{p-p} @10 KHz modulation voltage. Lower trace: modulated optical signal (2.2 pm/V @ 633 nm).

Electro-optic coefficient measurements are summarized in Table 1. Measured values of r_{33} were in the range of 1 to 2.5 pm/V. These small values may be attributed to the relatively mild poling conditions used in this sample preparation. Dr. Jarek Zieba (of SUNY) has recently indicated that an electro-optic coefficient of 50 pm/V has been measured in their laboratory after much stronger poling. (The poling stability may be in question in this case.) Even at this higher value the material is not practical for application in the Hughes beam deflector. For comparison, liquid crystals can achieve an effective electro-optic coefficient of 5000 picometer/volt at voltages of only 10 V.

As a check on our results, we requested that SUNY provide measurement data from their lab setup. Their apparatus utilizes a similar reflection technique to ours. In addition, both real and imaginary indexes of refraction are derived from their measurements. They also have the capability to scan across a waveband to obtain r_{33} versus wavelength. The measured characteristic for sample E0-IV from SUNY is shown in Figure 7. Using the relation $r_{33} = 3 \cdot r_{13}$, the value of r_{33} measured at 633-nm wavelength is approximately 3 pm/V, compared to the 2 to 2.5 pm/V measured at HRL.

Table 1. Electro-optic coefficient, r_{33}, measured from SUNY non-linear polymer.

Electro-optic coefficient, r_{33} (picometer/volt)						
		Modulation Frequency =	0.5 KHz	1 KHz	10 KHz	100 KHz
# E0-IV	(a)	$V_m = 200\ V_{p\text{-}p}$	2.5	2.5	2.3	2.2
		$= 400\ V_{p\text{-}p}$	2.5	2.5	2.3	2.2
	(b)	$V_m = 200\ V_{p\text{-}p}$	2.3	2.3	2.2	2.1
		$= 400\ V_{p\text{-}p}$	2.3	2.3	2.2	2.1
	(c)	$V_m = 200\ V_{p\text{-}p}$	2.0	2.0	2.0	1.9
		$= 400\ V_{p\text{-}p}$	2.1	2.1	2.0	2.0
# E0-II	(a)	$V_m = 200\ V_{p\text{-}p}$	1.1	1.0	1.0	1.0
		$= 400\ V_{p\text{-}p}$	1.1	1.1	1.0	1.0
	(b)	$V_m = 200\ V_{p\text{-}p}$	1.1	1.1	1.0	1.0
		$= 400\ V_{p\text{-}p}$	1.1	1.1	1.0	1.0
	(c)	$V_m = 200\ V_{p\text{-}p}$	1.3	1.3	1.2	1.1
		$= 400\ V_{p\text{-}p}$	1.3	1.3	1.2	1.1

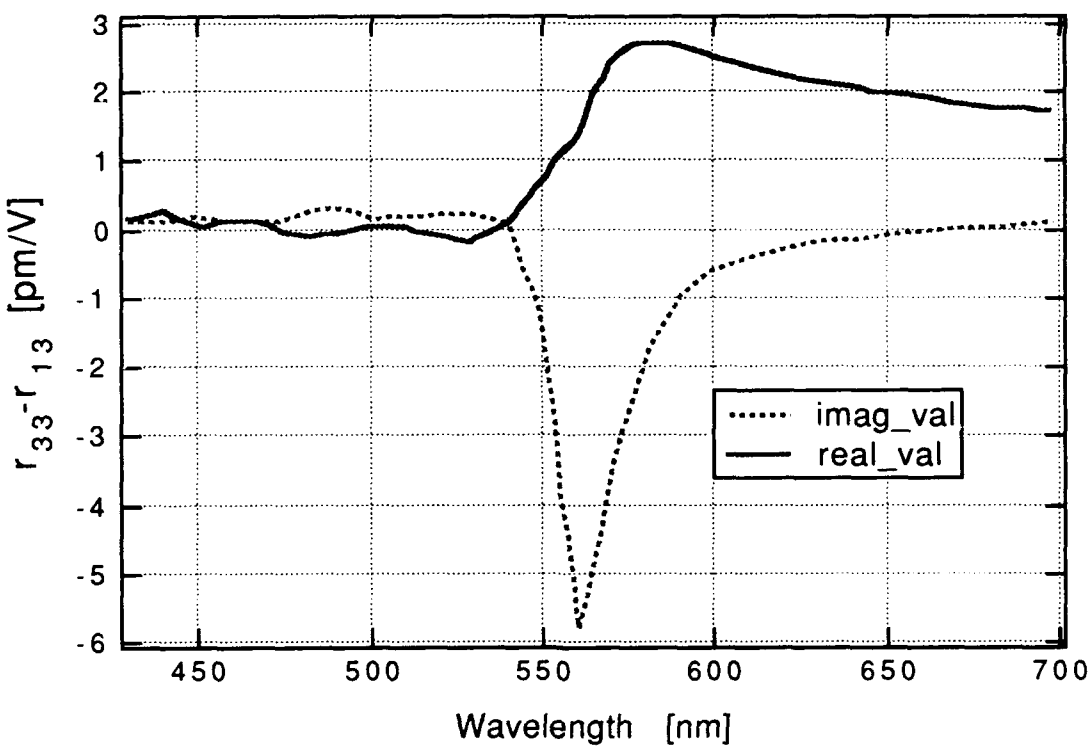


Figure 7. Electro-optic coefficient, Sample E0-IV, measured at SUNY.

Section 5
Summary and Conclusions

Electro-optical polymer materials developed at SUNY/Buffalo were supplied to Hughes for the purpose of evaluation for consideration in application to the Hughes beam deflector device. These materials were supplied as thin films on ITO-coated glass slides. They were corona poled and cross-linked for stability by SUNY before being sent to Hughes.

Hughes made measurements of transmission, resistivity, and electro-optic coefficient at various modulation frequencies. Because of absorption resonances below about 600 nm it is not possible to consider these materials for use at these wavelengths. Polymer resistivity values are so high that it is not anticipated to be a problem in beam deflector device applicaion. The EO measurements were made months after the poling was completed, and with no in-situ poling field, so the results give an indication of the moderate stability of the residual EO effect in these samples.

The measured EO coefficients are far below what is needed for practical use in the Hughes beam deflector. The amount of phase shift required for each modulator is a function of the maximum deflection angle, the width of each modulator, and the wavelength. With a typical index of refraction of 0.2 for liquid crystals, a thickness of 8 μm driven by 10 V can provide up to 10π phase shift at 633-nm wavelength. The 30 μm thick polymer E-O material we measured with 2.5 pm/V provides an index of refraction change of only 6.9×10^{-5}, or $1.3 \times 10^{-3}\pi$ phase shift at the 633-nm wavelength.

Further development of EO polymers could still produce much better results. Very recent measurements at the USC Department of Chemistry indicate that EO coefficients over 100 and potentially as high as 1000 pm/V can be achieved. At this level the materials would be much closer to performance of LCs in terms of potential phase retardation, indicating that there is potential for future use of non-linear organic materials for beam deflection and other spatial light modulation applications.

Appendix A
SUNY/Buffalo Polymer Material

This appendix provides a brief statement supplied by SUNY regarding the preparation of the polymer material used in this study. It is provided here for completeness and reference. Additional information on the design, fabrication, and evaluation of these materials is contained in Ref. [10] and citations therein.

In the material preparation for this study, special attention was paid to the purity of all the composite components. Polyvinyl (9-N-carbazole) PVK (Aldrich #18,260-5) was purified by adding dropwise a 8 wt.% solution of PVK in toluene to boiling ethanol. This procedure was repeated twice. The glass transition temperature of PVK after purification was 207°C. DEANST was synthesized according to the method of Kurihara, et al[11] and purified by triple recrystalization from ethanol, followed by column chromatography over neutral silica gel, eluting with 1:10 (by volume) mixture of ethyl acetate and n-hexane. Finally, it was sublimated at 85°C under vacuum of 40 mTorr. After sublimation the melting point of DEANST was 104.1°C.

For the preparation of films, a 10% solution of PVK (by weight) in 50:50 mixture of toluene/cyclohexanone was used for dissolving DEANST. Films with thicknesses of about 10 to 15 μm were spin casted onto ITO-covered glass substrates and dried at room temperature for 2-3 hours. Subsequently, the films were dried at an elevated temperature in order to expel the residual solvents. The films had the DEANST doping level of 28.5% (by weight). Because of the plasticizing effect of the dopant the T_g of PVK/DEANST (154-153°C) was significantly lower than for pure PVK.

References and Footnotes

1. C.C. Teng and H.T. Man, "Simple reflection technique for measuring the electro-optic coefficient of poled polymers," Appl. Phys. Lett. **56**,1734 (1990).

2. J.S. Schildkraut, "Determination of the electrooptic coefficient of a poled polymer film," Appl. Opt. **29**, 2839 (1990).

3. It was discovered that sample vibration due to piezoelectric effects can cause a modulation frequency dependent resonance in the EO optical effect. This spurious effect is eliminated by appropriate substrate mounting.

4. L.P. Mosteller, Jr and F. Wooten, "Optical Properties and Reflectance of Uniaxial Absorbing Crystals," J. of the Opt. Soc. of Am. **58**, 511 (1968).

5. See Refs. 1 and 2.

6. This formula is taken from Teng and Man, Ref. 1. It is noted that this result differs slightly from that in Schildkraut, Ref. 2, possibly due to a difference in derivation of the phase retardation Ψ_{sp}. Since the difference amounts to less than 20% variation in the interpreted r coefficients we did not attempt to resolve this conflict.

7. Same as Ref. 2.

8. Same as Ref. 2.

9. K. Clays and J.S. Schildkraut, "Dispersion of the complex electro-optic coefficient and electrochromic effects in poled polymer films," J. Opt. Soc. Am. B **9**, 2274 (1992).

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