

# Traveling wave electro-optic phase modulators based on intrinsically polar self-assembled chromophoric superlattices

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Traveling-wave electro-optic modulators based on chromophoric self-assembled superlattices (SASs) possessing intrinsically polar microstructures have been designed and fabricated. Although the thickness of the SAS layer is only  $\sim 150$  nm, a  $\pi$ -phase shift is clearly observed. From the measured  $V_\pi$  value, the effective electro-optic coefficient of the SAS film is determined to be  $\sim 21.8$  pm/V at an input wavelength of 1064 nm. © 2001 American Institute of Physics. [DOI: 10.1063/1.1389514]

Since the 1970's, significant advances in electro-optic (EO) modulators have attracted great interest, because of their potential applications in ultrahigh speed information transmission and processing. Over the past 30 years, much attention has been devoted to the study of various materials such as  $\text{LiNbO}_3$ ,<sup>1</sup> III-V semiconductors,<sup>2</sup> and organic polymers,<sup>3</sup> etc., for use in fabricating modulator EO-active layer. In comparison to inorganic crystal-based modulators, organic polymer devices offer potential advantages of large bandwidths and low half-wave voltages ( $V_\pi$ ). This is because organic polymers have low dielectric constants,<sup>4</sup> which permit the optical and microwaves to propagate with nearly equal phase velocities in such material. Moreover, organic polymers offer the promise of exceptionally high EO coefficients,<sup>5</sup> which are very important factors in reducing  $V_\pi$ . Organic modulators which operate at frequencies as high as 113 GHz<sup>6</sup> and with very low  $V_\pi$  (0.8 V)<sup>7</sup> have recently been demonstrated. However, most studies to date of organic modulators have used poled polymers as the EO-active layer. Such materials are challenged by loss-inducing chromophore aggregation effects, charge injection-induced degradation, and the inherently low degree of polar orientation that can be achieved by the poling process.<sup>8</sup> Additional concerns include achieving sufficiently high  $T_g$  values to stabilize polar orientations for long periods of time and the use of high poling voltages,<sup>9</sup> which introduces device design complexity and increases the volume and cost of the devices.

In this letter, we report on the first thin-film self-assembled superlattice (SAS) chromophore waveguide structure with the potential for traveling-wave EO modulator applications. Compared to poled polymers, the SAS microstructure is intrinsically polar, and the device design is therefore relatively simple. In addition, the SAS structures have high chromophore number densities and high degrees of acentric alignment, with second-order nonlinear coefficient  $d_{33}$  approaching  $\sim 700 \times 10^{-9}$  esu at a fundamental wavelength of 1064 nm for a single SAS layer.<sup>8</sup> Therefore, modu-

lators using SAS films as the guiding layer will combine the advantages of both poled-polymer and  $\text{LiNbO}_3$  devices.

The waveguide configuration employed is shown schematically in Fig. 1 and has a four-layer stack structure. The commercially available polymer Cytop™ (a fluorinated polyether) is used for the upper cladding layer, and  $\text{SiO}_2$  is used as the lower cladding layer. In this work, a 40 layer SAS combined with a layer of the polymer Cyclotene™ 3022-35 (poly bisbenzocyclobutane) served as the guiding layer. The thickness of the 40 layer SAS is  $\sim 150$  nm. The refractive index of Cyclotene™ 3022-35 ( $n = 1.56$  at a wavelength of 1064 nm) is very close to that of the SAS, and the refractive index of Cytop™ is 1.34 at a wavelength of 1064 nm. Both Cytop™ and Cyclotene™ exhibit good transparency over a wide wavelength range from the ultraviolet to infrared.<sup>10</sup> Af-

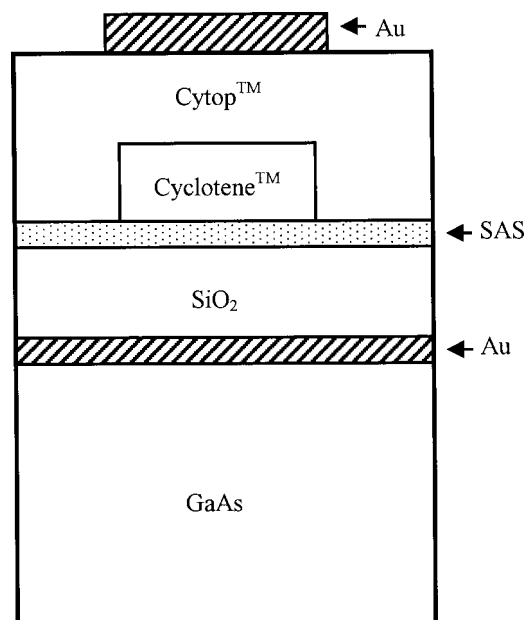


FIG. 1. Schematic cross section of the traveling wave EO phase modulator is shown.

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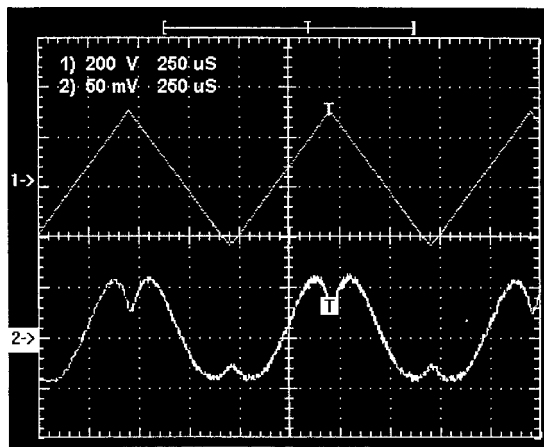


FIG. 2. The EO response measured from oscilloscope traces is shown. The upper trace is the applied electrical signal, and the lower trace is the modulator output wave form.

ter the gold layer was deposited on the GaAs substrate to form the ground electrode of the device, the lower SiO<sub>2</sub> cladding layer with a thickness of 2 μm and the SAS were grown by chemical vapor deposition and molecular self-assembly,<sup>8</sup> respectively. The details of the SAS growth method have been described elsewhere.<sup>11</sup> Next, the Cyclotene™ and Cytop™ layers were spin coated onto the SAS layer. The spinning speed and concentrations of the polymer solutions were calibrated for the desired film thickness. In the present fabrication process, the spinning conditions were adjusted for a Cytop™ layer having a thickness of 1.5 μm, and a Cyclotene™ layer having a thickness of 1.0 μm. To form uniform films, each layer was baked in an oven after spin coating (first at 70 °C for 30 min to remove the solvent, then at 130 °C for 30 min to anneal). The channel waveguide patterns were defined by conventional photolithography, and reactive ion etching (RIE) procedures were carried out to form a ridge waveguide structure. Since SAS structures proved difficult to etch by CF<sub>4</sub>, O<sub>2</sub>, and Ar in RIE, we etched the waveguide ridge height to be the same as that of the Cyclotene™ layer. The width of the resulting waveguide is 6 μm. Finally, after an additional Cytop™ layer was spin coated onto the above layers, and the upper gold electrode having 30 μm width and 0.4 μm thickness was fabricated using conventional lift-off techniques. The total thickness of the upper Cytop™ cladding layer is 2.5 μm.

The EO response of the waveguide was measured by end firing 1064 nm light from a diode pumped Nd:YAG laser, and was coupled in and out of the channel waveguide using a pair of 40× microscope objectives. The device was operated by simultaneously launching both transverse electric and transverse magnetic modes into the waveguide with light polarized at 45°. For recombining the input and output polarizations, the output light passed through a polarizer oriented at 45° and normal to the direction of the polarizer at the input. The light signal was finally coupled to a photodetector, and was measured by an oscilloscope. Figure 2 shows the typical EO response of the modulator measured from the oscilloscope traces. The upper and lower traces in Fig. 2 represent the applied electrical signal and the modulator output wave form, respectively. The half-wave voltage  $V_{\pi}$  is determined to be ~340 V.

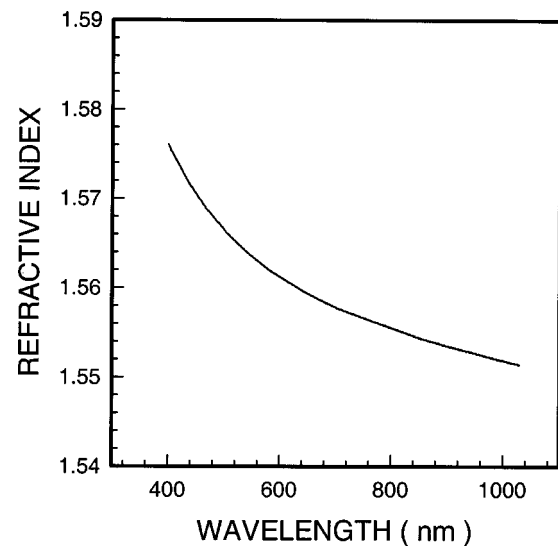


FIG. 3. Wavelength dependence of the refractive index of the self-assembled superlattice (SAS) is shown.

The large measured  $V_{\pi}$  is primarily due to the small thickness of the SAS layer, and the relatively small waveguide length. The half-wave voltage is given by,

$$V_{\pi} = \frac{\lambda G}{n^3 \Gamma r L}, \quad (1)$$

where  $\lambda$  is the wavelength of the pump laser,  $G$  is the gap between the ground electrode and upper electrode,  $n$  is the unperturbed refractive index of the material in the waveguide layer,  $L$  is the waveguide length,  $r$  is EO coefficient, and  $\Gamma$  is the spatial overlap of the optical intensity profile and the applied electric field. This is expressed as<sup>12</sup>

$$\Gamma = \frac{\iint E^2(y,z)A(y,z)F(y,z)dydz}{\iint E^2(y,z)dydz}, \quad (2)$$

where  $E(y,z)$  is the optical field distribution in the waveguide,  $F(y,z)$  is a step function representing the  $r$  distribution in the waveguide, and  $A(y,z)$  is the applied electrical field distribution. The refractive index of the SAS as a function of the wavelength was determined using a F50 advanced thin-film measurement system, and is plotted in Fig. 3. The measurement method and the wavelength dependence of refractive indices of Cyclotene™ and Cytop™ have been published elsewhere.<sup>10</sup> The measured waveguide length is 5 mm. Using Eq. (2),  $\Gamma$  is calculated to be ~0.045. From Eq. (1), the EO coefficient is thus estimated to be ~21.8 pm/V. At room temperature, the modulation characteristics of this device were unchanged over the course of many weeks. As mentioned, the response after the high temperature bake for the samples shows that the SAS film has a good thermal stability.

Figure 4 shows the linear optical absorption spectrum of the SAS film with 40 layers. The second-order nonlinear optical coefficient  $\chi^{(2)}$  of the 40 layer SAS has also been determined to be ~120 pm/V at the pump laser wavelength of 1064 nm. The relationship between the EO coefficient and  $\chi^{(2)}$  is given by<sup>13</sup>

$$r = -\frac{2\chi^{(2)}}{n^4} \frac{f^0}{f^2\omega} \frac{(3\omega_0^2 - \omega^2)(\omega_0^2 - 4\omega^2)}{3\omega_0^2(\omega_0^2 - \omega^2)}, \quad (3)$$

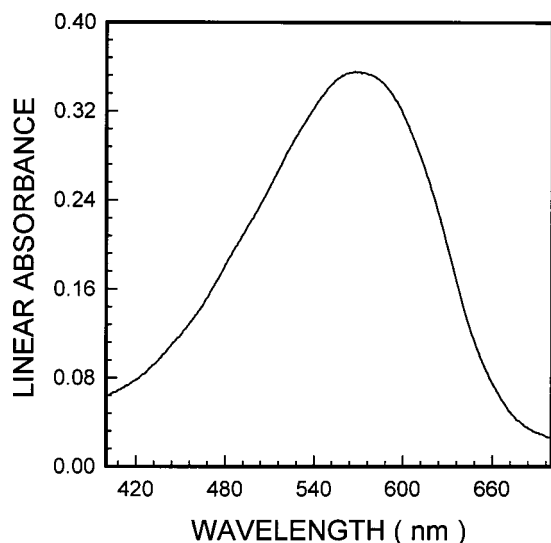


FIG. 4. Measured linear absorption spectrum of a SAS film having 40 layers is shown.

where  $\omega_0$  is the frequency of the absorption peak of the SAS,  $\omega$  is pump laser wavelength,  $f^0$  and  $f^{2\omega}$  are local field factors at the indicated frequencies. They can be written by<sup>14</sup>

$$f^0 = \frac{\epsilon(n^2(0) + 2)}{n^2(0) + 2\epsilon}, \quad (4)$$

$$f^{2\omega} = \frac{n(2\omega) + 2}{3}, \quad (5)$$

where  $\epsilon$  is the static dielectric constant, which we calculated from the measured refractive index and absorption spectrum data and  $n(0)$  and  $n(2\omega)$  are the optical indices of refraction. Using Eq. (3) and the measured  $\chi^{(2)}$  value, EO coefficient is calculated to be  $\sim 28.2$  pm/V, which is very close to the result calculated from the measured  $V_\pi$  value.

As a control experiment, we have fabricated a waveguide, the structure of which is similar to that in Fig. (1), but without the SAS layer. No EO response is observed with modulating voltages as high as 1000 V. This fact clearly shows the contribution of the SAS layer to the EO response. We have also fabricated a waveguide with the SAS layer, but with the ground electrode located under the substrate. The gap between two electrodes is then  $\sim 0.2$  mm. Measurement of the EO response shows that the output optical intensity increases with applied voltage. However,  $V_\pi$  is not observed up to the maximum voltage of the power supply (1000 V). This is because the gap between two electrodes is now much larger than that shown in Fig. 1. Assuming a SAS film was used exclusively as the guiding layer and the SAS film thickness was  $1.0 \mu\text{m}$ , then  $\Gamma$  would be  $\sim 0.7$ – $0.8$ . Furthermore,

if the length of the waveguide was 2.0 cm, then  $V_\pi$  would be reduced to below  $\sim 4.7$  V at the input wavelength of 1064 nm.

In conclusion, we have designed and fabricated the traveling-wave EO phase modulators based on a SAS material. There is no need to pole the device at high voltages. Although the film thickness of the SAS layer is only  $\sim 150$  nm, the EO response clearly exhibits a  $\pi$ -phase shift in the waveguide. We estimate that a SAS structure of  $\sim 1 \mu\text{m}$  thickness as the guiding layer will yield a small  $V_\pi$ . This is therefore a very promising modulator for applications.

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