

Nonlinear Optical Polymers in Advanced Photonics

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SYNOPSIS

Development of advanced photonic devices requires extensive integration of several technologies. Electrooptical devices must simultaneously satisfy several materials properties including: high nonlinear optical activity, thermal stability, mechanical strength, and optical transparency. In addition, the device materials must be amenable to fabrication, and must be compatible in a multi-layered configuration. We have recently developed polymers which meet these complex requirements. Indeed, electrooptical devices have been demonstrated using these polymers. This paper is a comprehensive review of the development of such devices, from design of the molecular architecture to waveguide design and fabrication. The critical issues in each aspect of device development are discussed. © 1994 John Wiley & Sons, Inc.

NONLINEAR OPTICAL POLYMERS

Nonlinear optical materials are the basis for an array of active optical devices in development throughout several high technology industries¹⁻³ including communications, computing, and aerospace. Active optical devices are used to alter light in order to encode or direct information in a number of different ways. Although the applications of optical technology such as fiber optics, optical storage, and displays have created a demand for such devices, material properties have limited their development.

OPTICAL TECHNOLOGY

As futuristic as optical devices seem, the idea of using light for the transmission of information is actually quite old. In fact, Alexander Graham Bell, after his historic transmission of speech through wires, attempted next to transmit speech in the form of optical signals⁴ with his so-called photophone. Signals could be heard over a distance of several hundred meters. In spite of such pioneering work, until the 1940s and early 1950s, only minor efforts were in-

vested in promoting the use of light for information transmission and processing. Studies during this period concluded that practical optical transmission systems were impossible without coherent light sources.⁵ In the absence of such sources, the capacities of optical transmission systems would be considerably smaller than those of millimeter wave systems. The invention of the laser in the early 1960s changed the situation dramatically by providing a stimulus in the form of coherent light sources of optical power in spectra extending from the UV through the visible and into infrared regions. Such light sources allow conventional optical communications systems to have much greater capacity than that of microwave systems. But improved light sources alone were inadequate to make optical communications systems a large-scale reality. Early optical communications components⁶ were bulky, heavy, expensive, and sensitive to many external influences and required high electrical power. For optical systems to compete with other forms of transmission, they had to be efficient, reliable, rugged, compatible with modern electronics, and economical.

The demonstration of low-loss optical fibers⁷ in the early 1970s brought modern optical transmission systems into reality by providing a highly efficient, high-capacity, reliable, and economic medium for light transmission over very long distances. During

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the 1980s, optical technology began to expand along a much broader front. Semiconductor research resulted in a new miniaturized coherent light source: the laser diode. This compact light source further spurred the use of fiber optics for communications and created entirely new optical applications such as optical storage, the compact disk, and optical displays. Today, optical technology is using increasing numbers of active optical devices that are emerging from development. Such devices modulate light-permitting higher-density information to be transmitted by fiber optics, switch light from one fiber to another, alter the wavelength of light to increase the capacity of optical storage, or actively interconnect electronic chips within computers.

FUNDAMENTALS OF NONLINEAR OPTICS (NLO)

All active optical devices require special materials that alter their optical properties with the application of an electromagnetic field. Devices that use an externally applied electric field require electrooptical material. Other devices utilize the electric field of the light itself and work by the application of light alone. In either case, at the core of the device is a nonlinear optical material. Such a material may be simply described⁸ by its polarization behavior under the influence of an electric field:

$$P_i(E_j, E_k, E_l, \dots) = \chi_{ij}^{(1)} E_j + \chi_{ijk}^{(2)} E_j E_k + \chi_{ijkl}^{(3)} E_j E_k E_l + \dots \quad (1)$$

Above, the polarization of a material, $P_i(E_j, E_k, E_l, \dots)$, is expressed as a function of independent applied electric fields, E , which may occur at different frequencies or along different axes given by j, k, l, \dots . All materials have a component of the polarization that shows a linear relationship between the electric field, E_i , and the polarization, P_i . The coefficient of linear polarizability is given by, $\chi_{ij}^{(1)}$, which expresses the degree of polarization that is achieved in direction i by the application of an electric field in direction j . For ordinary materials, this linear component is dominant, giving rise to linear optical properties. Nonlinear materials have additional contributions to polarization as described in the above equation by the additional coefficients $\chi_{ijk}^{(2)}$ and $\chi_{ijkl}^{(3)}$. As defined by the subscripts, nonlinear components of induced polarization are contributed by two or more electric fields at once. The fields may occur in different directions and each field may have

its own time dependence (i.e., may be of a different frequency). Thus, nonlinear polarization has the ability to intermix fields at different frequencies. For example, low-frequency electric fields from 0 to 100 GHz may be coupled through nonlinear polarization to affect the behavior of much higher optical frequency electric fields as is the case for electrooptical materials.

For organic materials, the polarization response is molecular in origin and is described as follows:

$$\mu_i(E_j, E_k, E_l, \dots) = \mu_0 + \alpha_{ij} E_j^0 + \beta_{ijk} E_j^0 E_k^0 + \gamma_{ijkl} E_j^0 E_k^0 E_l^0 + \dots \quad (2)$$

Here, $\mu_i(E_j, E_k, E_l, \dots)$ represents the molecular dipole moment in the terms of the permanent ground-state dipole moment, μ_0 , and the polarizabilities α, β , and γ . The actual electric fields experienced by a molecule are modified due to the presence of neighboring molecules and are denoted by E^0 .

A nonlinear optical material must be assembled from many nonlinear molecules. This may be accomplished by several methods, and as described in the relationships above, the resulting material is polarized by the application of electric fields in a manner directly analogous to the molecular case. There is a direct relationship between each type of molecular and material coefficient of polarizability. For example, the first nonlinear terms are related as follows:

$$\chi_{ijk}^{(2)} = N f^{\omega_1} f^{\omega_2} f^{\omega_3} \langle \beta_{ijk} \rangle \quad (3)$$

In this equation, N represents the number of molecules, and the f^ω terms, correction factors due to local electric field effects that neighboring molecules induce on each other. The brackets $\langle \rangle$ indicate an average for a specific molecular packing arrangement.

Coupling of electric fields at different frequencies appears at first to be an abstract concept. In fact, this phenomenon is not only observable but is also the basis of active optical devices. For example, the coupling of low frequencies from 0 to 100 GHz with light at optical frequencies is known as electrooptics and is the basis for an important class of devices. This coupling occurs as a result of the $\chi^{(2)}$ coefficient and gives rise to an electrooptic constant, r , as follows:

$$r = - \frac{2\chi^{(2)}}{n^2 \epsilon} \quad (4)$$

Here, n is the material index of refraction, and ϵ , its dielectric constant. r is a function of molecular activity given by eq. (2) and molecular packing as indicated in eq. (3).

The electrooptical constant may be used to illustrate just how fields of different frequencies are coupled. Under the application of an electric field, a material with $\chi^{(2)}$ (or r) activity changes its index of refraction. This is known as the electrooptical effect. Quantitatively, the change in index of refraction is given by

$$\Delta n = -\frac{1}{2} n^3 r E_{\text{applied}} \quad (5)$$

Light traveling through a material has a velocity, V , given by

$$V = \frac{c}{n} \quad (6)$$

where c is the speed of light in a vacuum. So, a change in the index of refraction due to an applied electric field produces a change in the velocity of light within the material as follows:

$$\Delta V = -\left(\frac{c}{n^2}\right) \Delta n = \frac{1}{2} n c r E_{\text{applied}} \quad (7)$$

DESIGN OF ELECTROOPTICAL POLYMERS

A key parameter characterizing an electrooptic (EO) material is its electrooptic (or Pockels) constant, r . It measures the extent to which the refractive index of the material changes with an applied electrical field [eq. (5)].

The Pockels constant⁸ of conventional EO materials ranges from 1 pm/V for GaAs to about 30 pm/V for LiNbO₃. Compared to these materials, organic NLO materials exhibit significantly improved EO performances. Single crystals of 2-methyl-4-nitroaniline (MNA) demonstrate an r value of 500 pm/V and EO polymers¹ have shown a value as high as 100 pm/V under suitable conditions. The EO performance of EO polymers is continuously improving with intense worldwide research activities.

For successful device applications, the EO materials need to possess physical properties other than just high EO activity. The EO property must be stable at high device operating temperatures (sometimes as high as 100°C) for an operational lifetime. They also must possess an optical transparency at operating laser wavelengths. Furthermore, the materials must be robust enough to withstand high

temperature, severe chemical environments, and mechanical stress of device fabrication processes. The EO polymers are advantageous not only in EO activity but also for their superior secondary properties.

The EO activity of organic NLO materials originates from the molecular electronic response to an electromagnetic field [eq. (2)]. How the NLO molecules are packed within a unit volume of the materials and well arranged in a noncentrosymmetric manner also affects the EO performance. An oft-used design rule for second-order NLO materials containing axial planar NLO chromophores can be written as

$$r = N f \beta \langle \cos^3 q \rangle \quad (8)$$

where r is the EO coefficient; N , the number density of the chromophore; and f , the local field factor. Also, β is the hyperpolarizability of the NLO chromophore, q is the angle between a chromophore and the optical axis, and the brackets imply an orientation average. The importance of the noncentrosymmetry is represented in the orientational brackets that vanish for centrosymmetric material systems. In the case of EO polymers, the design equation becomes

$$r \cong N f^3 \beta \mu_0 E / 5 k T \quad (9)$$

where μ_0 is the dipole moment; E , the poling field; k , Boltzmann's constant; and T , the poling temperature.

Using this equation as a guide, we can develop a strategy for development of polymers with high EO activities. First, we need a chromophore with large β and μ_0 . The equation clearly points to that the number density (N) of the NLO chromophores in the polymer should be maximized, as should the poling efficiency by means of high poling fields.

MOLECULAR DESIGN OF NLO CHROMOPHORES

A starting point of development of efficient organic EO materials is therefore design and synthesis of highly active NLO chromophore molecules with a large molecular hyperpolarizability β . The hyperpolarizability was defined from the molecular dipole moment equation [eq. (2)] as the coefficient of the term that is second order in E .

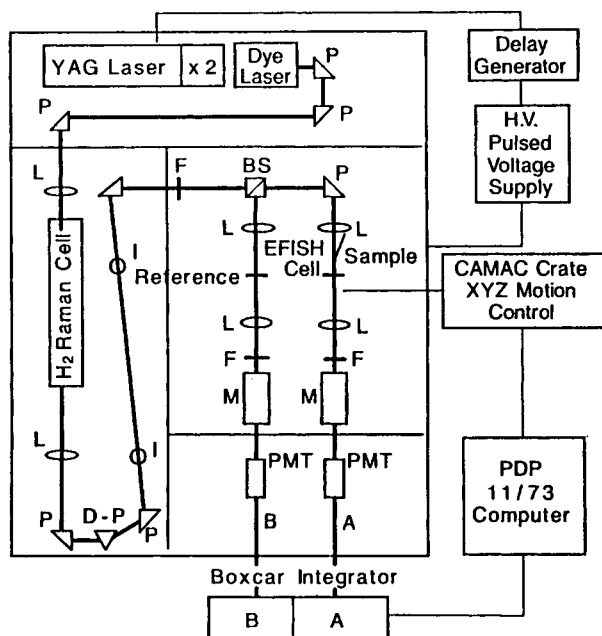
Theoretical consideration⁹ has identified structural requirements of highly active chromophore

molecules: The molecule must have an asymmetric electronic structure; the electron delocalization upon optical excitation must be large (i.e., the optically excited electron should move as large a distance as possible); and the quantum efficiency of the excitation should also be large. We studied the molecular NLO properties of many different chromophores meeting the above requirements both experimentally and using molecular modeling techniques and identified several chromophores for our polymer development effort (Table I).

For this study, there are a number of experimental techniques for molecular NLO investigation. One of the techniques¹⁰ is electric field-induced second harmonic generation (EFISH) in which chromophore molecules in solution are oriented by applying an electrical field and the intensity of second harmonic light signal is determined (Fig. 1). The molecular hyperpolarizability, β , is determined from the conversion efficiency from the fundamental to the second harmonic beams. Other techniques that we currently use include solvatochromic shift¹¹ of the

Table I Second-order Nonlinear Optical (NLO) Properties of Selected NLO Chromophores

NLO Compounds	β at 1.9 μm (10^{-30} esu)
	6
	18
	38
	22
	72
	40
	90
	100 (2800 at 1.06 μm)



Maker Fringe from Poled Polymer Film

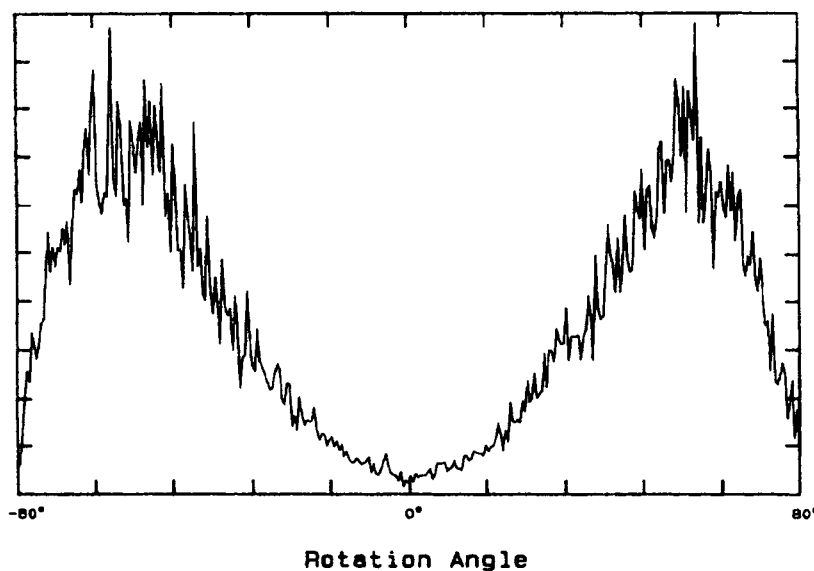


Figure 1 Second-order optical materials double the frequency of an incoming laser beam. The intensity of the doubled light is determined using an SHG unit (top). Intensity of the second harmonic changes as the angle between the sample and optical beam varies; the fringe pattern allows us to determine the second-order parameters such as χ^2 and β (bottom).

absorption spectra of NLO chromophores between polar and nonpolar solvents and EO measurement² of poled solid solution of NLO chromophores in polymer matrix. These techniques have been successfully used for rapid and reliable determination of new chromophores and establishing heuristic design rules of NLO chromophores.

New theoretical methods and extensive use of powerful computers now make possible realistic simulations of complex molecular and electronic processes. NLO processes are primarily due to molecular electronic phenomena. Quantum mechanics calculations come into play here because they can determine the most probable locations of a mole-

cule's electrons (i.e., molecular orbitals). Visualization of the molecular electronic processes and their relationships to the chemical structure are proving extremely helpful in the design of molecules with large NLO properties. Figure 2 shows two computed molecular orbitals[†] of 4-dimethylamino-4'-nitrostilbene (DANS), a molecule that exhibits a good second-order NLO response and is similar to the chromophore used in Hoechst Celanese NLO polymers.² The orbitals containing the highest-energy electrons (HOMO) are shown in red; the lowest-energy unoccupied molecular orbital (LUMO) is colored light blue. We can see from Figure 2 that the HOMO is localized on the amino side of the molecule (electron-donating) while the LUMO is localized on the nitro side of the molecule (electron-accepting). When the molecule absorbs light, an electron moves from the HOMO to the LUMO, inducing a large change in the molecular polarization (or a large electronic delocalization). The particular chemical characteristics leading to large spatial separation of the HOMO and LUMO (and thus high NLO activity) can be determined by comparing the high-resolution three-dimensional images computed for various sets of molecules.

We also use quantum mechanics calculations to study the interactions of NLO-active molecules with electric fields. The electron-donating and -accepting characteristics of each atom within a molecule are computed and represented graphically by spheres centered at each atomic position. This provides a detailed picture of the electronic behavior of the molecule. Figure 3 displays the computed partial atomic charges for the DANS molecule interacting with several different fields. The spherical radius represents the size of the partial atomic charge. The electron-accepting behavior of the nitro group is easily visualized in the increasing electron density on the two oxygen atoms as the electric field increases.

Beyond purely visual analyses, these studies yield accurate predictions of NLO activity. Field-dependent molecular dipole moments are easily computed from molecular orbital calculations.[†] The β values are then determined from these field-dependent nonlinear polarizations. Thus, the activity of new molecules can be predicted in advance of an often lengthy and difficult chemical synthesis.

These computational and experimental studies confirm the molecular structural requirements for

high NLO activity, as illustrated in Figure 4. What is needed is an asymmetric molecular structure containing electron-donating and electron-accepting groups, separated by an electronic bridge (such as a π -electron conjugated groups); such a structure has the required separation of the HOMO and LUMO and undergoes intramolecular charge transfer in an electric field, which the DANS molecule exemplifies. The effects of substituting various donor, acceptor, and bridge groups have also been studied both experimentally and by simulation. Figure 5 highlights the results, in which the effect of the π -electron bridge length and the acceptor strength on β are assessed. The data indicate that increasing the length of the electron bridge increases NLO activity and that the nitro group (NO_2) is a very good electron acceptor. These and other similarly derived design rules provide guidelines for our synthetic effort in the development of new chromophores that can meet device requirements and materials challenges.

NEW MOLECULES TO MATERIALS

Once the chromophores are identified and synthesized, they are assembled to create an EO material. As we saw in the material design equations [eq. (8)], the material design must ensure that the number density (N) of chromophores is large and that they are spatially arranged in a highly noncentrosymmetric manner to create a highly active EO materials. Furthermore, the material design also must ensure that the materials have all the important secondary properties needed in practical applications of these materials. It means that the material must be transparent over a few centimeters of length at laser wavelengths of interest and the EO properties must be stable over the device lifetime in the operating temperature and chemical environment. It also means that the material has robust mechanical properties and processability so that we can easily fabricate optical waveguides and that it survives the strains of fabrication process.

Several materials concepts for organic EO and NLO materials have been attempted. These include organic molecular crystals,¹² Langmuir-Blodgett (L-B) films¹³ and other self-assembled structures, poled solid solutions of NLO chromophores in polymer matrix, and poled EO polymers.¹⁴ Although the molecular crystal concept can provide an EO material with a large chromophore density, it has a number of serious drawbacks. The most serious is that highly active chromophores tend to crystallize

[†] Calculations were completed using the MOPAC computer program: QCPE program 506 by the Dewar research group and J. J. P. Stewart. Obtained from QCPE, Indiana University, Bloomington, IN.

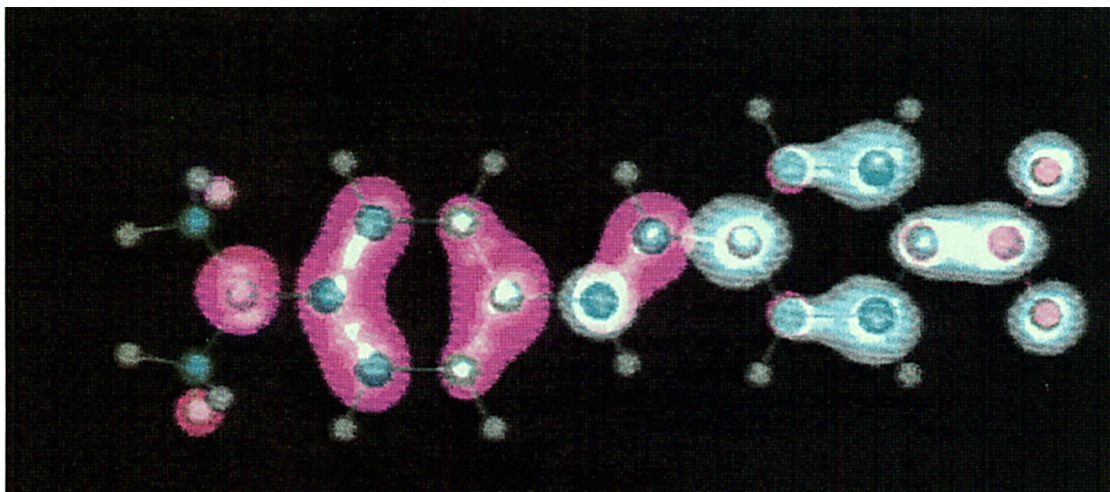


Figure 2 Computer modeling allows us to visualize the molecular orbitals of NLO chromophores such as 4-dimethylamino-4'-nitrostilbene (DANS).

in a centrosymmetric arrangement and develop a material with no macroscopic EO/NLO activity. The highly dipolar nature and a rodlike configuration of the chromophore molecules promote an antiparallel interaction and, thus, centrosymmetric crystallization. Although theoretical understanding is not complete, some NLO chromophores do crystallize in a noncentrosymmetric modification and exhibit a high NLO activity.¹⁵ However, these materials were found to be thermally unstable and weak

in mechanical properties and do not render themselves for easy device fabrication.

To circumvent the difficulty in the noncentrosymmetry requirement, L-B films and other self-assembled structures have also been employed. Although the techniques produce a highly ordered, noncentrosymmetric structure, some of the practical difficulties such as building a thick film required for waveguide applications (about 10 μm thickness), high temperature stability of the structure, and ease

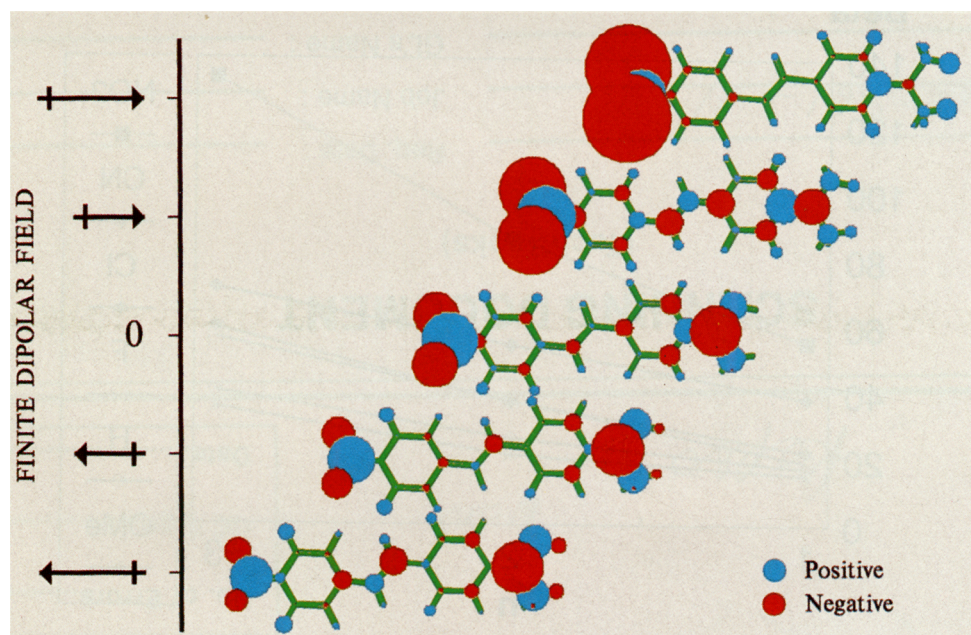


Figure 3 The partial atomic charges of an NLO chromophore such as DANS change with an external electrical field. The deformation of molecular charges reveals the origin of NLO processes.

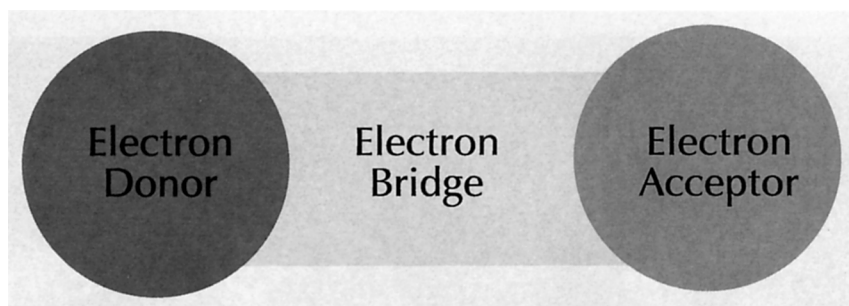


Figure 4 Molecules exhibiting a larger second-order optical response has a structure in which strong electron donor and acceptor groups are linked with a conjugated electron bridge for effective charge transfer upon optical excitation.

of waveguide definition need to be resolved for practical application of the molecular engineered materials. The solid solution approach suffers from the fact that polymers are generally a very poor solvent and, thus, the number density of chromophores that may be achieved is only about $1 \times 10^{20}/\text{cm}^3$.

EO POLYMERS AND POLING

We utilize a polymer architecture in which the polarizable NLO chromophore is attached as a side-

chain group through a hydrocarbon spacer linkage to allow the side groups enough rotational freedom for efficient poling. With a long spacer group (greater than four carbons), the polymer forms liquid crystalline phases that reduce its light-transmitting efficiency by scattering light. Isotropic glassy polymers are now used in many application development efforts. As an example of the side-chain NLO polymers, a methacrylate copolymer (P2ANS/MMA 50/50) with side chains containing 4,4'-aminonitrostilbene is shown in Figure 6. These polymers typically

Computed Beta as a Function of Bridge Length For Various Electron Acceptor Groups

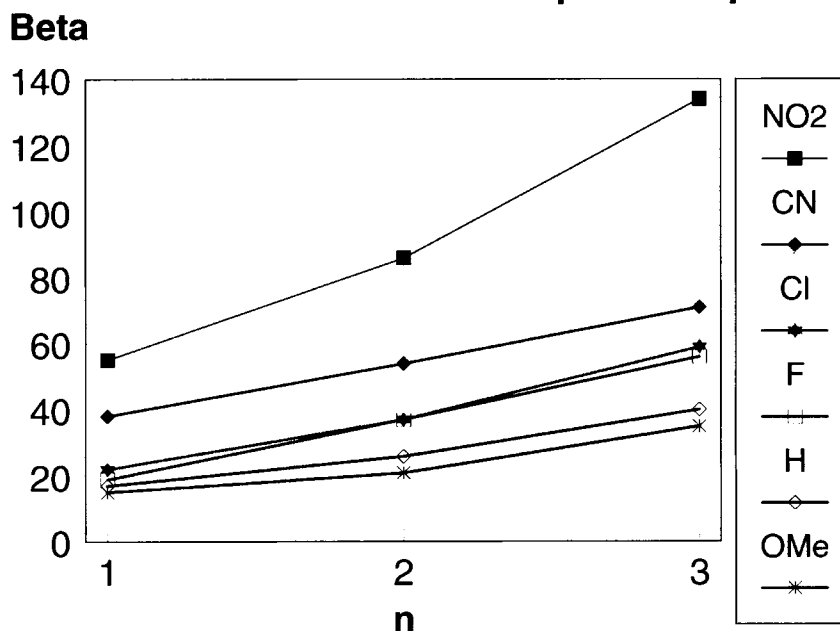


Figure 5 The molecular second-order response, β , strongly depends on the length of conjugated electron bridge and the strength of the electron-acceptor group as the molecular modeling results demonstrate.

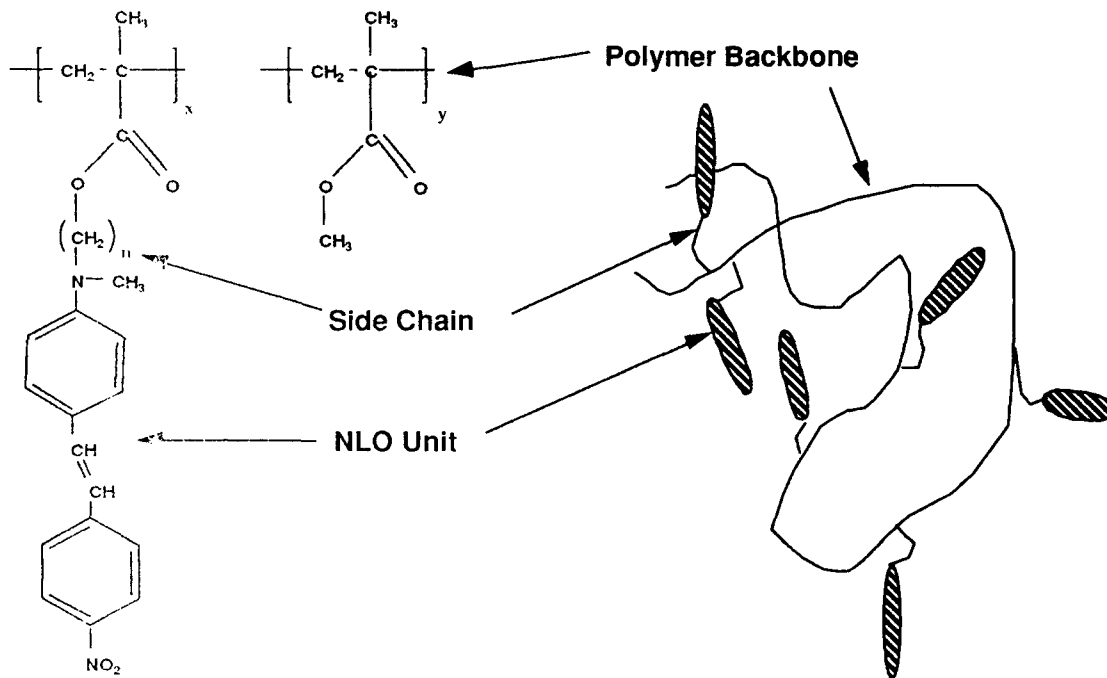


Figure 6 Inside-chain NLO polymers, the NLO chromophores are attached as a side-chain group to a polymer back through a flexible hydrocarbon spacer unit to facilitate poling process. The phase behavior of these polymers is controlled with the copolymer composition and the spacer length.

have refractive indices in the range of 1.5–1.7 and a dielectric constant of approximately 3.

The orientation distribution of chromophores in as-prepared polymer samples is symmetric and they do not exhibit an EO activity until an asymmetric orientation is developed using an electrical poling process. The process consists of heating the polymer sample to the vicinity of its T_g for sufficient molecular mobility, applying a strong field ($> 100 \text{ V}/\mu\text{m}$). During poling, the electrical field couples to the dipole moment of the NLO moieties and aligns them along the applied field direction. This alignment is counteracted by thermal randomization forces (Brownian motion). When an equilibrium is reached (which typically takes a few minutes), the polymer sample is cooled down to ambient temperatures and the electrical field is removed. At this point, the asymmetric alignment is frozen in and the sample exhibits a large EO activity.

Figure 7 shows the EO constant of P2ANS/MMA 50/50 and 5/95 as a function of poling voltage. The data include measurement results of the waveguide and reflection measurements at wavelengths of 633 nm and $1.3 \mu\text{m}$. The relative dispersion of r between the two wavelengths was calculated using a simple two-level model of chromophores. Values of the EO

constant as high as $40 \text{ pm}/\text{V}$ at $1.3 \mu\text{m}$ have been achieved with the polymer.

The side-chain polymer architecture also provides an excellent control of thermal stability of the poled structure and EO constant. By monitoring decay of $\chi^{(2)}$ at high temperatures of the polymers, it is found that the relaxation of the EO constant of poled

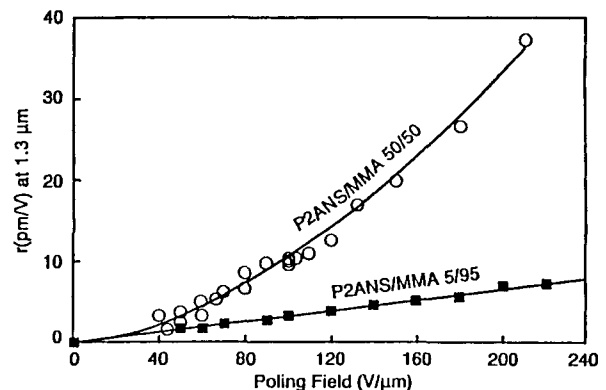


Figure 7 The EO constant, r , of NLO polymers such as P2ANS/MMA increases as the poling is accomplished at a higher field. The P2ANS/MMA polymer has already shown an r value higher than that of conventional EO materials from inorganic crystals.

polymers below T_g is closely related to the free-volume content in the sample. A modified WLF equation containing nonequilibrium free volume has been developed and the theory predicts that the poling stability at an operating temperature is directly related to the difference between the T_g of the polymer and the temperature. Quantitatively, the poled sample shows a stability longer than 10 years if the temperature difference is greater than 65°C. For example, the P2ANS/MMA 50/50 polymer has a T_g of 138°C and will be stable for 10 years at 73°C. The theory has successfully been used for design of polymers satisfying specified poling stability requirements.

ELECTROOPTIC MODULATORS

Various EO devices utilize the linear change in refractive index of an EO medium with application of an electrical signal [eq. (5)]. The index change induces a change in the phase of the optical wave propagating through the material. In this way, any information in an applied electrical signal is encoded in the phase of the optical beam that is being transmitted through the NLO material. The induced phase modulation can be further transformed into intensity or frequency modulation by appropriately designing optical circuits.

To give a quantitative feel of the materials and device parameters involved in EO modulators, we briefly discuss the quantitative aspects of linear EO of polymeric materials. In general, the EO devices are developed for use in single-mode optical fiber network and therefore utilizes a single-mode waveguide architecture. The lateral dimensions of the waveguides are typically about 10–20 μm and one applies an electrical field less than about 0.5 V/ μm . The field then produces a refractive index change (Δn) of about 45×10^{-6} in a polymer with the refractive index of 1.65 and r of 40 pm/V through eq. (5). This creates a phase shift of about 0.7π in a laser beam of 1.3 μm wavelength when it travels through 1 cm of the waveguide. Or, it should travel through 1.4 cm to create a π radian phase shift, which is needed in destructive interference between two beams. The relationship between the material properties, device performance, and geometry is conveniently expressed by the following equation:

$$V_\pi = \lambda \left(\frac{l}{n^3 r} \right) \frac{d}{L\Gamma} \quad (10)$$

where V_π is the switching voltage; l , the wavelength;

L , the active wavelength; d , the lateral thickness of the waveguide; and Γ , the overlap factor between optical and electrical fields.

One focus device of the Hoechst Celanese program is the Mach-Zehnder interferometric modulator.^{2,8} The modulator consists of a waveguide interferometer of the Mach-Zehnder type (Fig. 8). The incoming optical beam is divided in two at the front Y-junction and then recombined in the back Y-junction. If the phase shifts of the divided beams in the both arms are identical, they constructively interfere and all the power reappears at the output. When the difference in phase shifts is π radian, the beams destructively interfere and no power comes out at the output. Thus, the optical output power can be controlled by an electrical signal applied to the modulator. Proposed applications of the modulator are a digital modulator for telecommunication and an analog modulator for an optical fiber link such as b-ISDN.

Device performance parameters important for such applications are the switching voltage, frequency bandwidth, and optical insertion loss of the devices. These parameters are closely related to the physical properties of EO materials. Compared to traditional inorganic EO materials such as LiNbO_3 , the EO polymers have significant advantages for superior devices. The switching voltage is directly related to the EO response of the material and the ever-increasing EO performance of the polymers will reduce the switching voltage to a level (< 4 V) easily accessible with semiconductor-based power supplies in the near future.

Traveling-wave Waveguide E-O Modulator

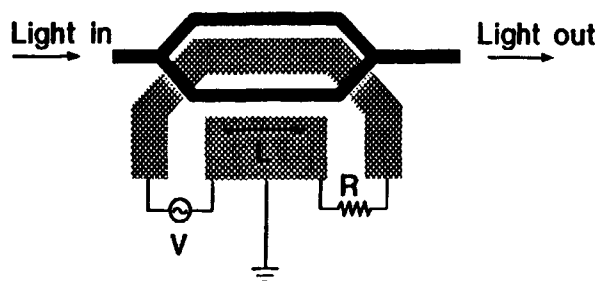


Figure 8 A Mach-Zehnder modulator operates by changing the refractive index in an EO waveguide arm and interfering beams traveled through the two waveguide arms. The extremely fast optical switch is proposed to be used as a information encoder in optical fiber telecommunication networks.

The speed (the frequency bandwidth) at which the signals can be modulated depends on the characteristics of the electrodes and the dielectric properties of the materials. In lumped-electrode modulators, the speed is limited by the capacitance of the device and, therefore, the low dielectric constant of polymers (about 3 vs. 31 of LiNbO_3) is a significant advantage over inorganic EO materials. The speed of the device can be further increased by configuring the electrodes as transmission lines and sending the microwave signals along with the optical signals, ideally at the same speed so that electrical and optical signals are synchronized; this extends the bandwidth considerably. Such an approach is only possible, however, if the optical material's refractive index (which determines the speed of light in the waveguide) and the square root of the dielectric constant (which determines the speed of the microwave signal in the transmission line) are equal or nearly equal. Here, again, NLO polymers have a significant edge over their inorganic counterparts because the polymers come much closer to meeting this velocity matching requirement. As a result, the theoretical speeds of polymeric devices (hundreds of GHz) are more than an order of magnitude greater than those in inorganic EO materials (tens of GHz).

ELECTROOPTICAL ACTIVE WAVEGUIDES

Although the physical properties of EO materials have a large influence in determining the performance of the EO modulators, the details of device structure and fabrication quality also have significant effects on the device performance. The device optical insertion loss is particularly sensitive to its geometry and quality. There are three major sources of the optical loss with a typical waveguide device: optical coupling loss, waveguide bending loss, and propagation loss. The coupling loss refers to the optical loss that occurs at the interface between a fiber optic network and the device. Several factors contribute: imperfection of the device end surface, misalignment between the two waveguides, and, most importantly, mismatch of optical field patterns in the fiber and waveguides. The bending loss occurs at the bends in the waveguide circuitry and is sensitive to the detailed geometry of the bends. The propagation loss can come from scattering and absorption of waveguides and from absorption of the evanescent optical field by metal electrodes. As such, the latter is particularly sensitive to the strength of evanescent field in the cladding layer.

The three performance parameters—switching voltage, frequency bandwidth, and optical insertion loss—are interrelated in a complex way and the optimum design of the waveguide devices is almost impossible without help of extensive computer modeling. For example, all three parameters are dependent on the device length in such a way that, with increasing device length, the switching voltage performance improves but the bandwidth and the loss characteristics degrade. The optimum design of devices must take into account the interrelationships between the performance parameters and their dependence on device structure.

Waveguide design, especially in the transverse direction, is arguably the most important component in the development of superior waveguide EO modulators. We use a step index waveguide design in which there is a small discontinuity in refractive index between waveguide and cladding materials. Inverted rib or channel architecture (Fig. 9) is used to confine the optical field in the waveguide. For the modulator application, the waveguide must be a single mode and this implies that the refractive index difference between waveguide and cladding must be less than 0.01 for any realistic waveguide lateral dimensions. To design waveguides with a superior EO performance and meeting the tight specifications, we developed a copolymer cladding materials technology that takes advantage of controllability of refractive index and EO response in the cladding region.

Because of the inherent complexity of waveguide modulator performance, design optimization and accurate performance prediction of the device must be augmented with an extensive modeling effort. We have developed and used an array of modeling tools in the development of polymer-based waveguide EO modulators. For longitudinal electrode structures such as a microstrip, a simple electrostatic analysis of the cross section of the strip yields the electric field distribution. From this distribution, the characteristic impedance and attenuation can be calculated; design parameters such as the microstrip dimensions can then be varied to achieve a specified impedance (typically 50%). The microwave field distribution over an optical waveguide also can be used in conjunction with an optical field distribution in this waveguide to predict the effectiveness of the structure in modulating light.

Although ideal optical waveguides such as optical fibers or rectangular waveguides can be studied with simple personal computer programs, real waveguides do not have these ideal cross sections. To see the optical field distribution in such a waveguide, we use

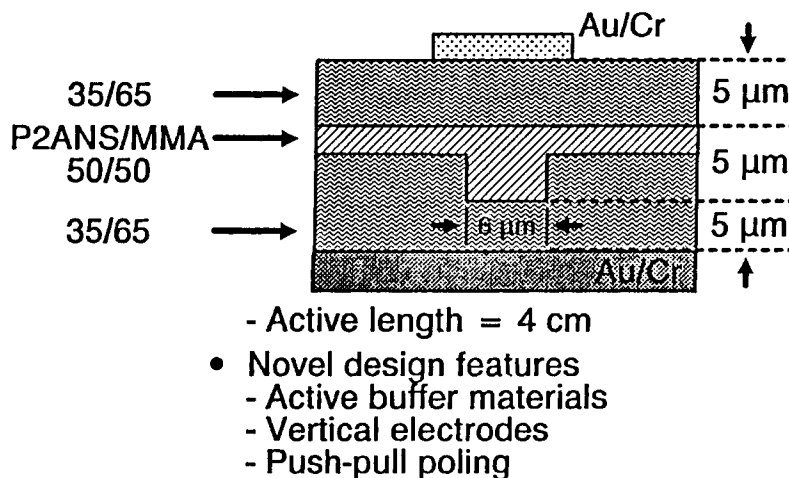


Figure 9 EO polymer waveguides requires a careful selection of the materials and precise control of the dimensions. Here, the cross-sectional view of channel waveguides shows the design features.

a finite element method (FEM).¹⁶ The FEM breaks down complex geometries into small pieces composed of simple shapes such as triangles or quadrilaterals. Then, it solves the electromagnetic field equation in each of the elements using a simple set of basis functions and recombines them into a full solution. We have used the FEM program to solve many complex waveguide problems. An example is shown in Figure 10, where the cross section of a channel waveguide can be seen, with the core outlined in yellow. The colors represent the electric field amplitude distribution of the first eigenmode of this waveguide. The optical field pattern is then convoluted with the electrical field pattern generated by the microstrip electrodes (which is computed using a numerical electrostatic modeling program) to determine the overlap parameter, Γ , of eq. (10) of the EO modulator and, thus, to accurately predict the switching voltage performance.

We use another set of tools to model how an optical beam travels down a complex waveguide circuit and how much light is preserved in the circuit. A key goal here is to design waveguides minimizing the amount of light lost in the device and the overall size of the device. For this purpose, we use a beam propagation method (BPM).¹⁷ Figure 11 shows a Mach-Zehnder modulator modeled with the BPM. In Figure 11(a), the indices of refraction of both arms are identical, leading to constructive interference at the output; thus, the device is "on." In Figure 11(b), the index of the upper arm is slightly changed, as it would be if modulated by an electric signal, so the two beams meet in phase opposition. The result is no output beam; the device is now "off." A more rigorous modeling tool than BPM is the fi-

nite difference time domain (FDTD)¹⁸ and it is used in accurate calculation of the optical loss around a critical waveguide component such as a Y-junction or bend. Figure 12(a)–(c) show an optical wave propagating through a right-angle bend at three different times; the beam deflection is produced by an imbedded air cavity at a 45° angle.

DEVICE CONSTRUCTION

Once the modeling and analyses are finished, the optical waveguide devices are then built. The first step is usually to deposit three layers of films of different polymers onto a substrate by spin-coating. Definition of the waveguide patterns and their two-dimensional confinement is accomplished by lithographic or other writing or machining methods, including reactive-ion-etching (RIE), photobleaching, and laser ablation. Through appropriate patterning and lithography, various optical waveguide circuits can be generated, depending on the device functions and requirements. Converting the circuits into working polymeric waveguide EO devices requires a poling operation. The poling electric field, which ranges from 100 to 200 V/ μm , is applied by two metallic electrodes on the top and bottom surfaces of the layered structure. After poling, an electrode pattern is deposited, aligned, and delineated on the top surface of the layered film. The electrode is for application of external electronic signals, which modify the material's refractive index through the EO effect. The information in the electronic signals is thus modulated onto the optical beam passing through the active portion of the device. The elec-

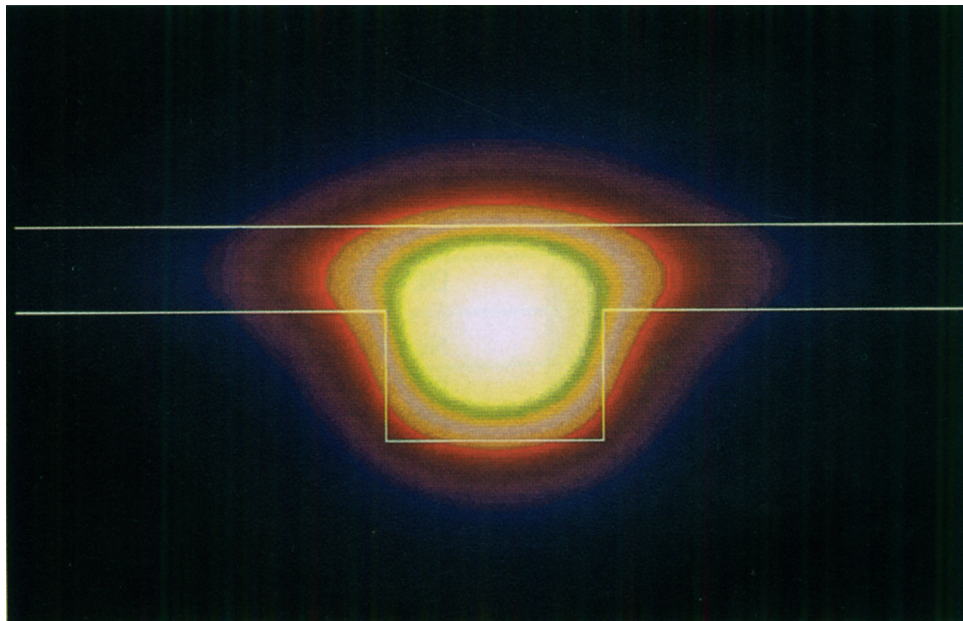


Figure 10 Lateral confinement of an optical beam in a channel waveguides enables EO manipulation of the beam in the waveguide over a few centimeters length.

trode design must be compatible with operational requirements such as speed, switching voltage, and power.

Fabrication of a functioning device also requires preparation of the high-quality end surfaces of the device for efficient coupling of light in and out of

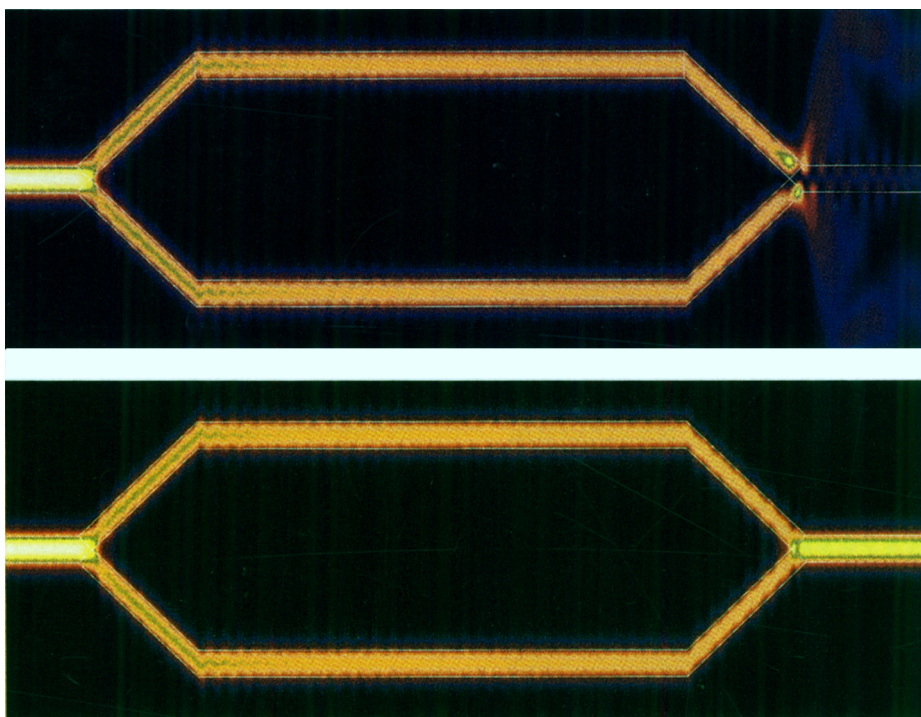
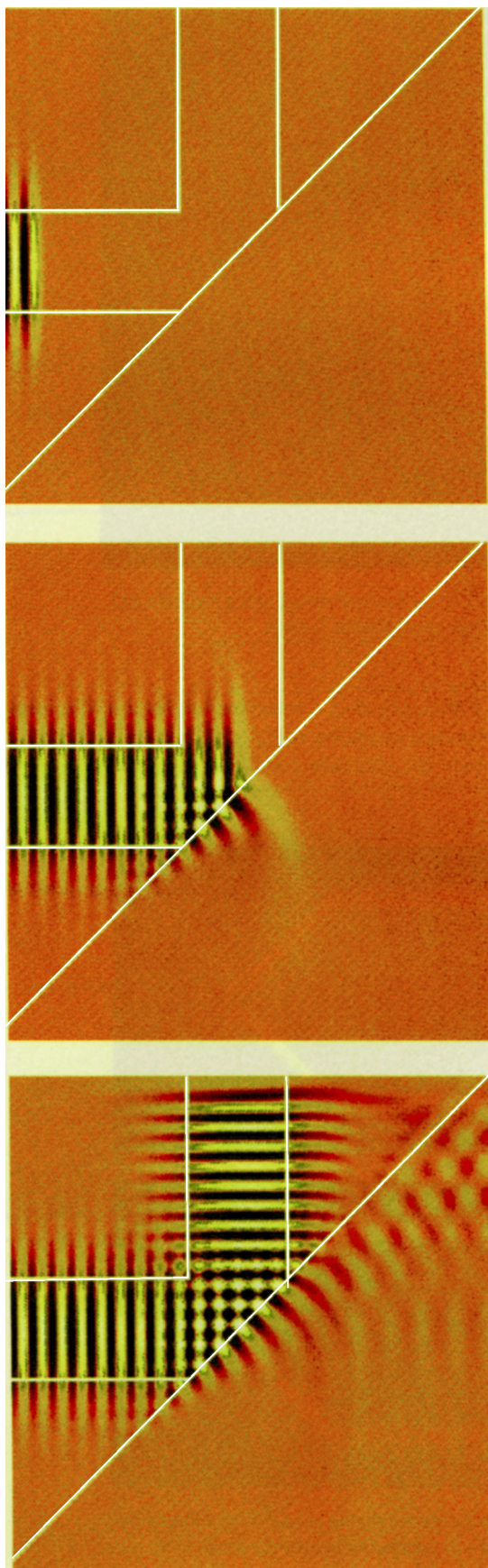


Figure 11 The optical switching in a Mach-Zehnder modulator is modeled and visualized with the beam propagation model (BPM). With such computer modeling tools, complex device design and operation are optimized.



the device via optical fibers. Polishing, cleaving, or other machining methods may be used to prepare sharp, smooth, and defect-free ends. Optical fibers are then precisely aligned and permanently pigtailed to the device for reliable interface with optical transmission systems.

POLYMERS FOR ACTIVE OPTICAL DEVICES

Nonlinear optical materials made of organic polymers are likely to be the centerpieces of tomorrow's high-speed devices. Although optimizing the properties of these materials and designing efficient devices will be formidable tasks, organic polymers have a number of unique properties that make them materials of choice for optical devices. We have demonstrated that

- Organics are intrinsically fast (femtosecond response) due to the lossless process of polarizing delocalized electrons within an organic molecule and have low dielectric constants that permit low capacitances, resulting in very high-speed EO devices.
- Organics have exceptionally high nonlinear activity, resulting in improved device performance.
- Organic polymers may be tailored by conventional synthesis methods to meet specific device requirements (e.g., transparency at particular wavelengths or stability at particular temperatures).
- Organic polymers may be formed into thin films ideal for the waveguide format of most active optical devices and offer a number of processing options to reduce the cost of active devices.

These materials can be incorporated into advanced EO devices, suggesting that they will play a role in affordable fiber-optic links to the home and a host of other key device applications for advanced optical systems.

CONCLUSIONS

The world of optics continues to expand at an ever-accelerating rate along a broader front affecting all

Figure 12 Detailed investigations of critical optical components such as a 90° waveguide bend shown here are carried out using the finite difference time domain model and a high-speed computing facility.

high-technology industries. This expansion is driven by technology advancements within optics including fiber optics, laser diodes, and, finally, active optical devices. Increasingly, however, this expansion is driven by events in related technologies such as ultrahigh-speed electronics, smart system designs, and structural materials that have created a market demand for a host of active optical devices.

Optical storage is now a reality that is in use throughout industry. The need to further increase storage capacity has created a demand for shorter wavelength blue light sources. This need is being met by coupling laser diodes to active optical devices made from NLO material capable of doubling the frequency of light from the laser diode and thereby halving the wavelength.

Advances in high-speed electronics through sub-micron definition of electrical circuits has led to a speed approaching 1 GHz for a single chip. These chips are being assembled into multichip modules, which then are capable of producing information at speeds far exceeding 1 GHz. The need to interconnect chips within the module or to interconnect one module to another has created a demand for an active optical solution that is perceived as the only viable method to switch such high-density information traffic. Such systems are key to advances in computation by parallel processing, aerospace, and a number of other high-technology industries.

Advancement of communications technology is accelerating, so that within the next decade, fiber optics will penetrate the home, bringing with it high-definition, large-screen displays, links to very smart home computers, and access to large random-access data bases. Moreover, in business and industry, a truly global communications network is emerging, unleashing a flood of information that will have to be managed and controlled. Optical communication systems, with their enormous speed, huge capacity, reliability, and compatibility with existing electronic devices, will play a pivotal role in meeting these communications needs of tomorrow.

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