

## ACKNOWLEDGMENTS

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## Nonlinear Optics and Photonic Applications of Photorefractive Polymeric Composite Materials

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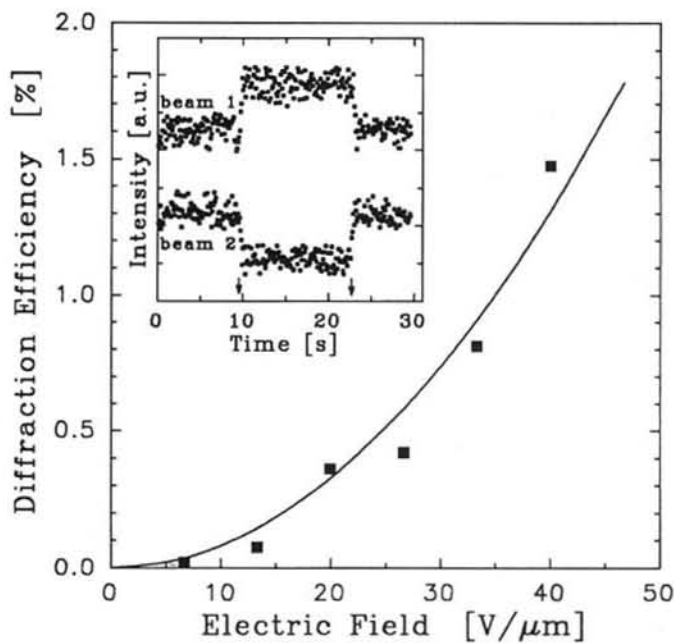
Until very recently the photorefractive effect has been observed exclusively in inorganic crystals, such as BaTiO<sub>3</sub>, doped GaAs, BSO, and others. The organic polymeric photorefractive films define an entirely new and very promising class of media exhibiting photorefractive response. The crucial functions for photorefractivity are photoconductivity and electro-optic activity. Known inorganics show high  $\chi^{(2)}$  nonlinearities and moderate to high carrier mobilities. However, there is an inherent limitation imposed on the photorefractive figure of merit,  $n^3 r_{\text{eff}} / \epsilon$  (where  $n$  is the refractive index,  $r_{\text{eff}}$  the effective electro-optic coefficient, and  $\epsilon$  the material's dielectric constant), for these systems due to the ionic polarizability origin of their nonlinearities resulting in high values of  $\epsilon$ .<sup>1</sup> Secondly, these materials are very difficult to fabricate and process. Organics differ from their inorganic counterparts in that they are easy to process and, due to the localized nature of their electronic properties, combine low dielectric constants and relatively high electro-optic coefficients.

We adopted the approach of dividing, among independent components of the system, the different functions that a photorefractive material has to fulfill.<sup>2</sup> From the perspective of engineering photonic materials, this approach has two main advantages. First, the composite can be spectrally sensitized for information writing at a desired wavelength by choosing the proper photosensitizer, the NLO-active chromophore being chosen to keep its absorption resonances away from the working wavelength and thus avoid creation of local absorption gratings (*e.g.* of photochromic origin). Second, the polymeric matrix can also be independently

optimized with respect to carrier mobility, concentration of trapping centers, and optical quality. An example of such designed material is the polymeric composite PVK:C<sub>60</sub>:DEANST, containing C<sub>60</sub> fullerene molecules responsible for photogeneration of charge carriers, diethylaminonitrostyrene (DEANST) moieties providing high electro-optic response, and poly[vinylcarbazole] (PVK) polymer as a charge transporting host matrix.

Two key defining parameters for photorefractives are their diffraction efficiency and two beam coupling gain. The figure below shows the dependence of diffraction efficiency on applied dc electric field, measured in a non-degenerate four-wave mixing geometry. Values of diffraction efficiency for a ~300  $\mu\text{m}$ -thick film at applied field of 40 V/ $\mu\text{m}$  reach 1.5%.<sup>4</sup>

The nonlocal photorefractive nature of the effect finds its confirmation in an asymmetric two-beam coupling experiment<sup>4</sup> (see the Inset in the figure), where the gain coefficient measured at  $\lambda = 645$  nm is  $\sim 4$  cm<sup>-1</sup>. This effect allows for a controllable stationary energy transfer between the interacting beams—the phenomenon of considerable photonic significance, *e.g.*, for all-optical parallel image processing.



The dependence of diffraction efficiency on applied dc electric field for a PVK:C<sub>60</sub>:DEANST composite film ( $\lambda_{\text{write}} = 645$  nm,  $\lambda_{\text{read}} = 632.8$  nm). The solid line is calculated according to the model describing formation of space-charge gratings in photoconducting polymers.<sup>3</sup> The Inset shows the beam intensity changes in an asymmetric two-beam coupling process ( $\lambda = 645$  nm). The arrows denote the moments of switching the electric field "on" and "off."

The role of the applied dc field for these materials is crucial and multi-functional. The field: (i) enhances charge carrier photogeneration quantum yield; (ii) stimulates transport of the photogenerated carriers; and (iii) forces a noncentrosymmetric alignment of  $\chi^{(2)}$  chromophores (*in situ* poling). Indeed, as we found in our studies, a very efficient switching of the photorefractive response between "0" and "1" states of the diffraction efficiency can be effected in the millisecond time regime with the use of external field.<sup>5</sup>

The obtained photorefractive efficiencies compare well to the corresponding values for some of the known inorganic photorefractive crystals and, as research progresses, the efficiencies should scale up by a meaningful factor. Two areas of effort are important here; the use of better  $\chi^{(2)}$  chromophores as well as more efficient poling techniques, and better control over the carrier trapping centers to produce higher amplitude space-charge gratings. Emphasis will also be put on increasing the beam interaction path length by using a guided wave geometry of the device.

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## Advanced Modeling and Simulation of Self-electro-optic Effect Devices

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Optical computing and photonic switching are new and emerging technologies for future parallel processing systems. Optically controllable switching elements based on multiple quantum wells (MQW), especially the self-electro-optic effect devices (SEEDs), are well-suited for applications in these areas and are now developed in different variants for mass fabrication of GaAs/AlGaAs integrated circuits (for an up-to-date overview, see Ref. 1).

As a basis for circuit simulations, we derived a new approximation of the photocurrent-to-voltage characteristics of the MQW diode.<sup>2</sup> The major mechanism behind this device is a clearly resolved exciton resonance at the absorption edge causing a strong nonlinearity in optical absorption. The exciton resonance shifts and broadens with electric field. This effect is used for modulating the reflectivity of MQW diodes and to obtain electrically and optically bistable circuits. Our approximation is the first that reflects the main nonlinearity and also the light-hole and heavy-hole resonance peaks. Henceforth, our approximation can be used to obtain more accurate dynamic models of the device for circuit simulations. These simulations are especially useful in designing of F-SEEDs<sup>1</sup> in which MQW diodes are integrated together with field-effect transistors to obtain circuits with greater logical functionality per optically controlled element ("smart pixels").

To simulate larger systems as a means of design verification and system performance evaluation, a generalized structure of an symmetric SEED with all (theoretically) possible input and output signals has been considered. This circuit can function as either a latch or as a logic gate performing the AND, OR, NAND, or NOR functions depending on the optical input signals used and on the preset pulse used to control the function. The behavior of this circuit has been described logically, and a logic model has been developed.<sup>3</sup> The logic model of the S-SEED has been used to study different system design approaches like the programmable logic array design and a newly developed method to design more compact S-SEED systems.<sup>4</sup>

Since simulations at different levels are of great importance for device development and system design, we hope that our work will contribute to the progress in the interesting area of optical computing and photonic switching.

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