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## IV. Photorefractive and liquid crystal materials

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### IV.1. Introduction

This panel considered two separate subject areas: photorefractive materials used for nonlinear optics and liquid crystal materials used in light valves. Two related subjects were not considered due to lack of expertise on the panel: photorefractive materials used in light valves and liquid crystal materials used in nonlinear optics. Although the inclusion of a discussion of light valves by a panel on nonlinear optical materials at first seems odd, it is logical because light valves and photorefractive materials perform common functions.

Strictly speaking, one might define a photorefractive material as a material in which light induces a refractive-index change. Over the past fifteen years, however, the term has come to be understood to refer

to a much smaller subset of such materials. We consider photorefractive materials to be those in which absorbed photons cause charge migration (drift, diffusion, hopping, etc.), the distortion of charge results in a space charge field, and the field modulates the refractive index through the electrooptic effect. All photorefractive materials must absorb light and have both mobile charges and a nonzero electrooptic coefficient.

Liquid crystal light valves consist of a layer of liquid crystal material, an electric field applied to the layer, and a means to obtain spatial modulation of the field. In optically addressed light valves the spatial modulation is obtained by shining light on a photoconductive layer in series with the liquid crystal medium. In electronically addressed light valves the current through the device is modulated spatially.

## IV.2. Motivation for Research

There appear to be two major motives for research in photorefractive materials. The first is that one can perform many different optical processing algorithms in these materials with low-power cw lasers, at room temperature, at modest cost, in simple and compact systems, and with a relatively sturdy material. The second motive is that qualitatively new processes appear in experiments that use photorefractive materials because the nonlinearity is so large.

Motivation for research with liquid crystal light valves is clear. These light valves are in commercial production and any improvements in performance or new ways of using them can be translated directly into products.

In a broader sense, the motivation for research in photorefractive materials, light valves, and many other nonlinear materials is provided by a large number of parallel computing applications that either require the manipulation of huge amounts of optical data or require more computing in a shorter time period than can be obtained digitally with current supercomputers. Examples of systems in which photorefractive materials may have realistic near-term applications include real-time optical pattern recognition, real-time command, control, and communication (C<sup>3</sup>) optical signal processing, optical computing modules, and the real-time synthetic array radar processor.

## IV.3. Review of the Field

Current photorefractive materials consist of electrooptic crystals such as BaTiO<sub>3</sub>,<sup>1</sup> KNbO<sub>3</sub>,<sup>2</sup> LiNbO<sub>3</sub>,<sup>3</sup> Sr<sub>1-x</sub>Ba<sub>x</sub>Nb<sub>2</sub>O<sub>6</sub> (SBN)<sup>4</sup> (ferroelectrics), Bi<sub>12</sub>(Si, Ge, Ti)O<sub>20</sub> (nonferroelectric oxides),<sup>5,6</sup> and GaAs,<sup>7,8</sup> InP,<sup>7</sup> CdTe (compound semiconductors). The mobile charge in these materials is provided by a donor (or acceptor) trap system such as provided by iron in two valence states (Fe<sup>2+</sup> and Fe<sup>3+</sup>) in KNbO<sub>3</sub>,<sup>2</sup> LiNbO<sub>3</sub>,<sup>9</sup> InP,<sup>7</sup> and probably BaTiO<sub>3</sub> (Ref. 10) or by the defect EL2 and EL2<sup>+</sup> in nominally undoped GaAs.<sup>7</sup>

Photorefractive materials permit construction of unique devices. Optical amplifiers with gain factors of 4000 (Ref. 11) have been constructed using photorefractive materials and cw lasers. Efficient self-pumped conjugators that are self-starting and require no external pump beams have also been constructed using cw lasers.<sup>12,13</sup> Here the only competing technology is stimulated Brillouin scattering conjugators that usually require pulsed lasers with more than a millijoule per pulse.<sup>14</sup>

A wide variety of other prototype operations have been performed in devices constructed from photorefractive materials. These operations include matrix inversion,<sup>15</sup> beam clean-up,<sup>16</sup> beam combining or locking,<sup>17</sup> real-time interferometry, associative memory,<sup>18-20</sup> threshold detection,<sup>21</sup> convolution/correlation,<sup>22</sup> edge enhancement,<sup>23</sup> differentiation/integration, holographic storage,<sup>3</sup> wavelength conversion, optical limiters,<sup>24</sup> incoherent-to-coherent conversion, and beam steering<sup>25</sup> or real-time holographic optical intercon-

nection, imaging of phase objects,<sup>26</sup> and rf signal correlation.<sup>27</sup>

A number of factors appear to be obstructing conversion of these prototype devices into marketable devices. First, not all photorefractive materials have a large enough nonlinearity to perform these operations. Second, commercial availability of materials with large nonlinearities is limited. To be specific, BaTiO<sub>3</sub>, which is available from one commercial supplier, has been used for many of these experiments. Other materials with large nonlinearities, such as KNbO<sub>3</sub>, SBN, and other tungsten bronzes, and KTa<sub>1-x</sub>Nb<sub>x</sub>O<sub>3</sub> (KTN) are not available commercially with good optical quality. Growth of these crystals of the size and optical quality needed for optical signal processing requires an investment of several years, substantial funding, and talented personnel.

A third factor limiting application is low sensitivity. It requires typically 0.5 J/cm<sup>2</sup> to produce a phase conjugate beam with BaTiO<sub>3</sub> using self-pumping. Thus for a moderate input intensity of 1 W/cm<sup>2</sup> the response time is of the order of 0.5 s<sup>-1</sup>. At this speed photorefractive materials cannot compete with light valves or electronic computers. Finally, available crystals of BaTiO<sub>3</sub> are somewhat smaller (<5 × 5 × 5 mm) than desirable for the typical optical processing algorithms. Other photorefractive materials are faster [e.g., Bi<sub>12</sub>SiO<sub>20</sub>,<sup>28</sup> or GaAs (Ref. 7)] or available in larger pieces (GaAs, LiNbO<sub>3</sub>) but none of these is as nonlinear as BaTiO<sub>3</sub>, and LiNbO<sub>3</sub> is slower (see Ref. 29, Table II). Other materials such as semiconductors like CdTe or InP have simply not been the subject of much investigation.

The limits of performance of photorefractive materials can be assessed in a number of ways.

### A. Speed

The photorefractive effect is essentially a response to optical energy; refractive-index change per absorbed photon.<sup>30</sup> The effect scales with energy until times as short as either the time for a charge carrier to move one grating period or the time necessary for the electrooptic effect to respond to the Coulomb field of the displaced charge. The longer of these times is the fundamental limit. Diffusion times in semiconductors such as GaAs are known to be <10 ps.<sup>31</sup> The electrooptic response time is of the order of attoseconds for the electronic component of the electrooptic coefficient,  $r$ , and picoseconds for the ionic part.<sup>32</sup> The mix of these two components seems to vary from crystal to crystal, but the worst case is again in the picosecond range.

### B. Damage

Optical damage thresholds for pulsed radiation are comparable to other optical materials (50–500 MW/cm<sup>2</sup>).<sup>33</sup> Many photorefractive crystals are extremely rugged and have a long working life. One researcher reports that his barium titanate crystal has been used many thousands of hours over nine years in argon-ion laser beams (often focused at full power) and doubled Nd:YAG pulsed beams (joules per cm<sup>2</sup> per pulse) with

no sign of deterioration. Other researchers have not been so fortunate, reporting damage from pulsed lasers of 1-W/cm<sup>2</sup> average power and damage from the thermal shock of removing painted electrodes with acetone.

### C. Ultimate Size of Nonlinearity

Two approaches have been considered. In the first, the refractive-index change  $\Delta n$  in a photorefractive material is given by  $\Delta n = n^3 r E / 2$ ,<sup>30</sup> where  $n$  is the background index,  $r$  is the electrooptic coefficient, and  $E$  is the space charge field. The optimum field that can be obtained is when each absorbed photon separates one charge carrier by a grating period  $\Lambda_g$ :  $E = Ne\Lambda_g / (\epsilon\epsilon_0)$ , where  $N$  is the number density of absorbed photons,  $e$  is the charge on an electron,  $\epsilon$  is the dielectric constant, and  $\epsilon_0$  is the permittivity of free space. The resulting index change per absorbed photon density then depends only on the grating period, the index  $n$ , and the ratio  $r/\epsilon$ . For typical parameters these considerations yield  $\Delta n/N \approx 10^{-19}$  cm<sup>-3</sup>. In the second approach, the energy required to obtain a phase conjugate reflectivity per pixel of 100% was calculated. This result again depended only on the ratio  $r/\epsilon$  and  $n$  and a lower limit of  $\sim 10^{-14}$  J was obtained.

A number of significant gaps in current knowledge of photorefractive materials have been identified. First, the microscopic behavior of BaTiO<sub>3</sub> is not completely understood.<sup>10,34</sup> This includes factors such as whether one or more species is responsible for the mobile charges, the relative role of electrons and holes,<sup>35,36</sup> a model for the response time,<sup>37</sup> and measurement of the mobility, quantum efficiency, and ionization/recombination cross section. Second, a detailed understanding of the fundamental limits on the ratio  $r/\epsilon$ , which control sensitivity, is not available. The ratio  $r/\epsilon$  varies by only about an order of magnitude for all known photorefractive materials while  $r$  varies from 1 to 2000 pm/V and  $\epsilon$  varies from 10 to 4000 (Ref. 27, Table II). Third, what techniques are available for optimizing properties such as quantum efficiency, species densities, cross sections, mobilities?<sup>10,34</sup> Fourth, there are broad gaps in our knowledge of crystal chemistry and crystal growth techniques that directly affect research in photorefractive materials. Finally, are there other photorefractive materials such as organic materials, or materials sensitive in the ultraviolet or far-infrared spectral bands?

### D. Liquid Crystal Technology

Liquid crystal (LC) devices are already used for a variety of displays and optical data processing applications in the form of optically or electronically addressed 2-D spatial light modulators. Optical data processing operations performed using liquid crystal devices include the following: image processing operations such as correlation,<sup>38</sup> level slicing,<sup>39</sup> analog to digital conversion,<sup>40</sup> logarithmic filtering,<sup>41</sup> and phase conjugation<sup>42</sup>; signal processing operations such as radar range-Doppler signal processing,<sup>43</sup> feasibility of synthetic aperture radar signal processing,<sup>44</sup> and spectrum analyzers<sup>45</sup>; optical computing operations such as

logical functions,<sup>46</sup> binary operations with bistable arrays,<sup>47</sup> and residue arithmetic operations.<sup>48</sup> Finally, optical interconnects using liquid crystal light valves were also demonstrated recently.<sup>49,50</sup> The main merit of LC technology is the extremely high electrooptic coefficient resulting in high resolution, large dynamic range devices. The large spectral bandwidth of liquid crystal is another asset which allows operation ranging from near UV to the IR region.<sup>51</sup> Thus, photoactivated liquid crystal light valves<sup>52,53</sup> are available with resolution exceeding one million elements, dynamic range of >100:1 (or few wavelengths in phase shift), response times of  $\approx 10$  ms, and sensitivity of 100  $\mu$ W/cm<sup>2</sup> for full activation (contrast ratio of 100:1 or refractive-index change of  $\approx 0.1$ ). Electronically addressed devices<sup>54</sup> typically feature  $\approx 300 \times 300$  elements with dynamic range and speed similar to that of the photoactivated devices. It is important to note that photoactivated liquid crystal spatial light modulators (SLMs) perform similar functions to those of photorefractive materials. It is therefore of interest to try to compare their relative performance. Since both classes of device perform an intensity to refractive-index conversion, the photorefractive sensitivity,<sup>55</sup> expressed as the incident energy density required for unity index change, can be used for this comparison. Typical values for photorefractive materials are 1–10<sup>2</sup> cm<sup>2</sup>/J.<sup>55</sup> In the case of a typical photoactivated LC-SLM, an index change of  $\Delta n \approx 0.1$  is attained using energy density of 1  $\mu$ J/cm<sup>2</sup> ( $=100 \mu$ W/cm<sup>2</sup> at 10-ms rise time). The effective photorefractive sensitivity of a LC-SLM is therefore  $\sim 1 \times 10^5$  cm<sup>2</sup>/J, which is three orders of magnitude higher than the typical values achieved in photorefractive materials. It should be noted, however, that the resolution of liquid crystal SLMs (typically 10–50 line pairs/mm) is significantly lower than that of the photorefractive effects, which is in the 1000-line pairs/mm regime. If one redefines the photorefractive sensitivity as the incident energy required for a unity change in the refractive index per pixel of information, the above gap in the photorefractive sensitivity between liquid crystal devices and photorefractive materials will shrink considerably. Present efforts are under way to improve the relatively slow response of the nematic materials by using ferroelectric LCs.<sup>56</sup> These can be switched at typical times of 10–100  $\mu$ s, but are binary in nature. For both classes of material a trade-off exists between the dynamic range and speed of response of the devices, as both quantities are proportional to the thickness of the LC cell.<sup>57</sup> The resolution is presently limited by the driving structure, electronic driving array (CCD-addressed or MOS-matrix), or the photoconductor. Ultimately the resolution will be limited by the fringing field in the liquid crystal layers. Finally, we should mention the large optical nonlinearities which liquid crystals exhibit.<sup>58,59</sup> The molecular reorientation responsible for the large optical modulation is due to either thermal effects or to optically induced fields. Effects such as self-focusing,<sup>60</sup> optical bistability,<sup>61</sup> and wavefront conjugation at a few W/cm<sup>2</sup> (Ref. 62) were recently demonstrated.

#### IV.4. Conclusions

Expand availability of photorefractive materials. Materials that are known to be interesting for the photorefractive effect such as  $\text{KNbO}_3$ ,  $\text{Bi}_{12}\text{TiO}_{20}$ , and a variety of mixed crystals ( $\text{KTa}_{1-x}\text{Nb}_x\text{O}_3$ ,  $\text{Sr}_{1-x}\text{Ba}_x\text{Nb}_2\text{O}_6$ ,  $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ ,  $\text{Ba}_{2-x}\text{Sr}_x\text{K}_{1-y}\text{Na}_y\text{Nb}_5\text{O}_{15}$ ,  $\text{Pb}_{1-x}\text{Ba}_x\text{Nb}_2\text{O}_6$ ) are unavailable to most researchers. The waiting time to obtain  $\text{BaTiO}_3$  ranges up to two years.

Encourage (i.e., fund) collaborative research efforts between researchers in crystal growth, in fundamental studies of defects, transport, etc., in development of exploratory devices, and ultimately in development of real systems. The benefits of strong interactions between these four groups have not been realized to a great extent.

Search for and investigate the properties of other photorefractive materials (e.g., semiconductors and organic materials if any are photorefractive). Emphasize molecular engineering of liquid crystals. In particular, this should result in improving the understanding of the relationship between molecular properties (e.g., polarizability) and the macroscopic properties of the LC material (e.g., birefringence). The development of such theory will assist in optimizing LC properties by synthesizing materials according to the guidelines developed by the theory. In particular, one would hope to optimize the dynamic range-speed trade-off in LC materials.

Develop a better understanding of the physics of ferroelectric liquid crystals (FLCs). In particular, use these studies to try to develop a gray-scale operation of FLCs.

Photorefractive and liquid crystal materials have occupied a unique position in research in nonlinear optics for the past ten years. The high nonlinearity that can be obtained in photorefractive materials using low power cw lasers has led to new and unexpected effects and has permitted construction of a wide variety of exploratory devices for optical processing and real-time holography. Key questions for the next ten years concern details of charge transport, achievable speed of response, sensitivity, and crystal availability. New photorefractive materials continue to be identified. In the domain of liquid crystal devices the critical issues are those of optimizing the dynamic range/speed trade-offs through molecular engineering in nematic materials and the development of gray-scale operation in ferroelectric liquid crystals. If these questions can be successfully answered, it seems likely that ten years from now photorefractive and, perhaps even earlier, liquid crystal materials will be used in a wide range of commercial and military products.

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## V. Inorganic nonlinear materials for frequency conversion

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### V.1. Introduction

Inorganic nonlinear optical materials are used mostly in high-power laser applications for extending the range of wavelengths available at high power. In the past, applications have mostly involved harmonic generation, but recently there has been considerable progress in other types of frequency conversion, such as optical parametric amplifiers. With a single frequency, efficient laser source, these devices promise wide wavelength flexibility at high power and conversion efficiency. As solid state devices they offer some advantages over other options for achieving wavelength flexibility.

The panel believes that these devices have great potential and deserve serious consideration in both military and civilian contexts. Development of a national capability to address our perceived future national requirements a decade from now requires a sustained, cogent national effort in crystal growth and a broad-based systematic materials research program. Because the lead time in this area of research and development program is long, it is important to begin now to have results in the 1990s. The panel believes that applications should provide both impetus and focus for this national effort, but the program should not be devoted totally to applications. The goal is to develop a national capability in this area spanning all its major aspects to meet needs ten years from now.

### V.2. Review of the Field

#### A. Applications

The principal uses of inorganic nonlinear optical materials are:

#### 1. Commercial Lasers

Frequency conversion devices are usually offered as accessories for short-pulse (10-ns) high-power lasers available commercially. These are typically harmonic generators for Nd:YAG lasers. While harmonic generators for other lasers are available, they are not routinely available commercially.

#### 2. Low Average Power Devices

These are miniaturized or portable devices used in information processing, medical instrumentation, xerography, etc.

#### 3. High Average Power Devices

This includes future defense needs in a variety of scenarios, generally summarized as control of high pulse energy, high average power electromagnetic radiation through modulation, deflection, and frequency conversion. Examples include battlefield lasers, submarine communications, countermeasures, and optical radar. A number of industrial processes also require high average power, such as chemical processing, materials processing, and x-ray lithography. The average power required can be up to several kilowatts.

#### 4. Fusion

Because of the high energy per pulse (up to 10 MJ) the primary requirement is for an inexpensive material for frequency conversion to the near UV, with high damage threshold.

#### B. Materials Requirements

To be useful, it is not sufficient that a nonlinear device work efficiently. The device must also survive