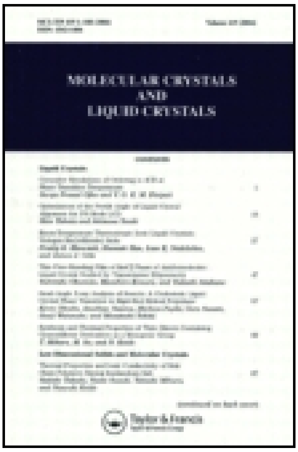

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Liquid-Crystal Materials for High Peak-Power Laser Applications

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LIQUID-CRYSTAL MATERIALS FOR HIGH PEAK-POWER LASER APPLICATIONS

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Abstract This is a brief review of materials and device-development issues related to the application of liquid-crystal technology to multikilojoule, high peak-power lasers.

INTRODUCTION

Novel applications of liquid crystals now include their use in high peak-power lasers. Multikilojoule laser systems instrumental in plasma physics and in inertial confinement fusion studies[1] require beam-control and transport elements with special sets of properties. These elements are usually required to be large in diameter (≤1 m), long-term stable at room temperature, low in loss (both in terms of scattering and absorption) at the laser operating wavelength, uniform in performance across the aperture (against systematic or random wave-front errors) and must be resistant to laser damage of multijoules/square centimeter fluence levels. Birefringent liquid crystals made their entry into the large-scale laser area when size limits among naturally occurring birefringent crystals forced the exploration of scalable, synthetic substitutes. From there, liquid crystals spread to several passive applications based on nematic or chiral nematic structures, and most recently to active electro-optic and opto-optic laser device concepts.

For the applications discussed here, monomeric and, still infrequently, side-chain polymeric liquid crystals are used. Polymers are of special interest: they promise device simplifications over monomeric systems that require high-grade, supporting substrates. However, polymer processing issues need further development before stand-alone, polymeric devices come into regular, high-peak-power laser use. Preliminary results on polymers were reported last year.[2]

We begin this review with a discussion of requirements germane to all devices, and follow with sections on active and passive devices.

GENERAL REQUIREMENTS

Both laser damage considerations and device performance uniformity mandate impurities and particulate matter be kept from the liquid-crystal layer and its associa alignment or surface anchoring structure. In practice, this necessitates frequent filtra and purification of all solvents, cleaning agents, and mesogenic materials used in the assembly process. It further requires device preparation and assembly to be carried out under cleanroom conditions. We currently operate class-100 cleanroom assembly using ultrasonic cleaning and rinsing of substrates in ultrapure water and drying in filtered nitrogen streams.

In addition to extrinsic impurities, intrinsic molecular structure may contribute to laser damage in organic media. Where highest fluence requirements must be met, mesogens must be selected that are fully saturated. How aryl groups and aromaticity phenomenologically affect laser damage at different wavelengths is published elsewhere for certain model compounds.[3,4] The detailed physical mechanisms dominating the energy transfer from the intense laser field into the molecular system needs yet to be clarified. At the pulse lengths of importance here (<1 ns to a few nanoseconds) nonlinear absorption appears a likely mechanism. In a following section on $\chi^{(3)}$ measurements of opto-optic device materials we will return to this issue.

PASSIVE DEVICES

Several passive device types such as wave plates, circular polarizers, and apodizers have been described before[5] in terms of their operational principles, liquid-crystal material properties, trade-offs, and assembly procedures. More than 60 units are now in continuous operation in the University of Rochester OMEGA laser with a remarkable reliability record. The major difference between display technology and laser technology devices based on identical operating principles lies in more stringent performance figures and tolerances for the latter. While, for instance, polarizer contrast between 20:1 and 50:1 is usually adequate for display applications, contrasts for laser circular polarizers are rarely less than 2000:1 and must be maintained to within ±3% across a 100-mm aperture. For the 24-beam OMEGA laser at Rochester, the key benefit derived from these performance figures is the beam-to-beam energy balance that now can be maintained routinely within 3% whereas without liquid-crystal optics the energy balance was at best 5%. Since irradiation uniformity and beam balance are the current *sine-qua-non* for direct-drive inertial confinement fusion, liquid-crystal technology is a direct contributor to progress in that area.

Both nematic wave plates and apodizers, as well as chiral-nematic polarizers of either handedness, are now available for different operating wavelengths, i.e., 1053 nm and 351 nm. Improved performance in the UV becomes increasingly important with more and more laser-driven inertial confinement fusion facilities opting for experiments at shorter wavelengths: the 1053-nm output of OMEGA (Rochester), NOVA (Lawrence Livermore) and GEKKOXII (Osaka) are frequency tripled to 351 nm, AURORA (Los Alamos) and the planned European Superlaser are or will be KrF systems lasing at 248 nm. All of these lasers are multibeam systems. Target irradiation uniformity can be enhanced for individual UV beams by controlling the on-target interference behavior of spatially separate portions of each beam. For this purpose, conventional liquid-crystal wave-plate concepts have been extended to one-dimensional and two-dimensional gradient devices with continuous, polarization-rotation wedge or checkerboard-style, discrete rotation properties (Figure 1) (distributed polarization rotator, DPR).

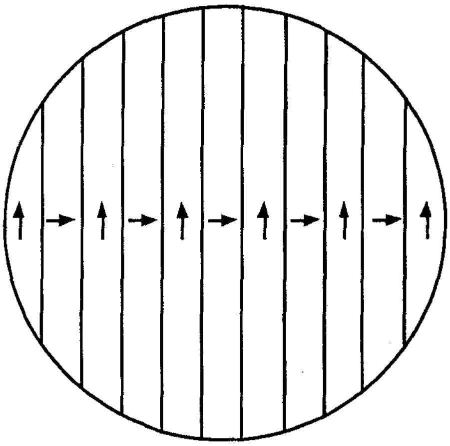

FIGURE 1 The output polarization distribution of a linearly polarized input beam after passing through a distributed polarization rotator.

Performance details for DPR's have also been reported elsewhere.[6] The major challenge in assembling the checkerboard devices lies in optimizing the photolithographic patterning process to minimize laser damage caused by photoresist residue at checkerboard-element edges. Continuous, one-dimensional wedge devices are simpler in assembly and are already deployed on the OMEGA laser. The liquid-crystal material used is a commercial mixture of fluorinated cyclohexyl-core nematics (BDH 14616/14627) with a birefringence of $\Delta n = 0.04$ at 351 nm.

ACTIVE DEVICES

Among the active liquid-crystal devices we distinguish electro-optic and opto-optic devices with the latter group further separated into slow and fast response devices. All of these devices are either at the prototype stage or under further development preceding actual, routine use in the high peak-power laser. The purpose of these devices is again in facilitating or enabling beam control and transport at 1053 nm or in enhancing irradiation uniformity on target at 351 nm. They are all based on well understood physical principles applied to untried situations. They are also largely based on commercially available materials except for fast-response, electronic-nonlinearity, opto-optic compounds that form the core of our in-house, synthesis activities.

Electro-Optic Devices

The electro-optic device to be described here is an electrically addressed wave plate that has a radially varying layer thickness. Between parallel polarizers, the radial gap gradient causes a soft-edge apodization that can be radially tuned by electric-field adjusted tilt of the nematic liquid crystal. Figure 2 shows a cross section through such a device. The 25-mm diameter prototype of this device uses indium-tin-oxide conductive coatings and Nylon 66 alignment layers together with BDH E-7 nematic mixture. The radially variable gap results from the spherical surface of a convex lens, the center of which has been polished flat. In a further iteration on this method of creating a

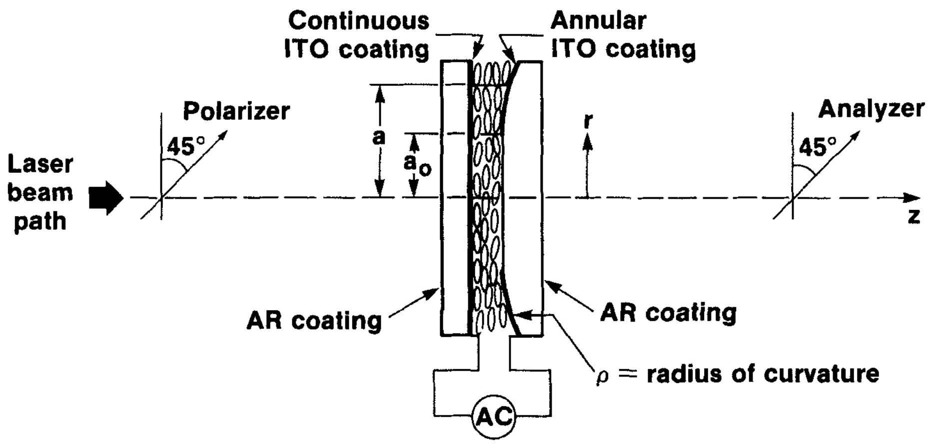

FIGURE 2 Schematic of the tunable apodizer based on the nematic wave plate concept.

prescribed apodizer edge profile, the surface profile is diamond turned from a substrate material that lends itself well to this process. Diamond turning permits more flexibility in nonspherical surface profiling than other techniques. The prototype-device clear aperture of 7.2 mm could be widened to 8.93 mm by applying a 1-kHz, sinusoidal voltage of 10 V peak-peak. The concomitant wave front error, measured by shearing interferometry, that is introduced by this switching action was limited to 0.24 waves along the ordinary direction and 1.16 waves spherical aberration at 632 nm along the extraordinary direction.

Application of the tunable apodizer in large-scale, high peak-power lasers still awaits further scale-up development work including phase-error compensation by using low-birefringence material, aperture extension to ~15 cm, etc. In rod-amplifier systems tunable apodizers promise adaptive control over edge gain. "Adaptive" here means the aperture is optimized interactively and then left at the optimum setting. It does not imply instantaneous change during the transit of a one-to-a-few nanosecond long pulse.

Opto-Optic Devices

We currently use an opto-optic device based on slow, optical-field induced molecular reorientation, and develop materials for another, fast opto-optic response device that relies on an electronic $\chi^{(3)}$ nonlinearity. The fast device is conceptualized as a dynamic focusing device that changes focal length in the subnanosecond time domain.

Bragg matching a chiral nematic structure to a circularly polarized optical field and changing the chiral pitch under the influence of the intense optical field are well-known effects.[7] Here, we combine these effects with the transverse-varying intensity distribution of the circulating field inside a laser cavity to generate a "self-adaptive" end mirror with several key advantages. The physics of spatially varying pitch dilation[8] brings about a pinholing effect and a retrofocusing effect in the reflected field, eliminating the need for an intracavity mode-selecting aperture and, at the same time, reducing diffractive cavity losses. As a result, a cavity equipped with a chiral-nematic end mirror offers a lower single transverse-mode lasing threshold than an aperture-equipped cavity with entirely dielectric mirrors. The fluid nature of the liquid crystal presents another advantage: once the retrofocusing and pinholing has been established above lasing threshold, the molecules adapt to angular mirror adjustments and maintain single-mode lasing over a mirror angular adjustment range twice that permissible for a dielectric reflector. This feature has been used with an intracavity etalon, reducing lasing to single transverse and single longitudinal modes. In this manner, monomode operation at cw output-power levels in excess of 1 W was maintained in a YAG cavity for time periods exceeding 30 min without any mechanical or thermal stabilization means.[9]

Whether similar performance can be achieved with pulsed oscillators is still under investigation.

For this power regime (≤ 1 W) a blend of a commercially available nematic (ZLI 1167) and a chiral additive CB15 has proven adequate. With the mixture tuned to selective reflection at 1064 nm, a liquid-crystal layer of 12-μm thickness produces a reflectivity of 98%. At higher powers, or at other operating wavelengths, a fully saturated chiral additive, e.g., ZLI 4447, is advisable for laser damage reasons.

Electronic $\chi^{(3)}$ Materials

For the development of subnanosecond reponse-time opto-optic devices such as dynamic focusing elements or "quivering" distributed polarization rotators operating at UV wavelengths, we undertook a multiphase, materials synthesis effort of $\chi^{(3)}$ materials. In its first phase, a group of materials was synthesized and tested near 1053 nm, with the intent of identifying the extent to which electron delocalization in functional groups and in molecular linkages enhances or suppresses the nonlinear susceptibility. In later phases, UV-specific materials development and testing near 351 nm are planned.

The activity is guided by the basic tenet that large all-optical nonlinear responses under both resonance and nonresonance conditions are associated with π-bonded, electron-delocalized functional groups. A significant body of theoretical modelling of the nonlinear response in model organic systems buttresses this assumption.[10] In high peak-power, low-repetition-rate applications *resonant* nonlinear optical effects as a result of absorption of the incident light are undesirable for damage reasons. However, the concern that organic systems in general will, on account of photochemical instability, fall short in high bit-rate device applications[11] even under *nonresonant* conditions is not applicable here because of the very limited repetition rate at which large glass lasers can operate (one shot every 30 min).

Emphasizing the synthesis of thermotropic liquid crystals as $\chi^{(3)}$ materials is rationalized as follows. The macroscopic nonlinear susceptibility $\chi^{(3)}$ of a system of molecules with molecular hyperpolarizability γ is found by summing over all N molecules per unit volume and cosine product terms representing the rotation from the molecular reference frames into a macroscopic, Cartesian frame given by the respective polarization directions of the incident, optical waves driving the nonlinear response:

$$\chi^{(3)}_{ijkl}(\omega_4;\omega_1,\omega_2,\omega_3) \propto N\sum \cos(i,I)\cos(j,J) \cos(k,K)\cos(l,L)\,\gamma_{IJKL}$$

It is through these cosine terms that the order of liquid-crystal mesophase systems can help maximize $\chi^{(3)}$. In fact, by simply temperature tuning a sample from the isotropic phase into an ordered phase one can attempt to distinguish electronic contributions to $\chi^{(3)}$ from those stemming from molecular reorientation ("nuclear contribution"). Thermotropic liquid crystals offer this intrinsic advantage of order that in other media must be obtained by additional processing such as uniaxial stretching or electric-field poling.

To establish a systematic trend of third-order nonlinear optical properties with molecular configuration, a group of naphthyl-core compounds was synthesized with varying linkages or substituents. All compounds were evaluated under as close to identical conditions as possible and their relative $\chi^{(3)}$ values were determined by nearly degenerate four-wave mixing and were compared with the current, defacto $\chi^{(3)}$ standard material, CS_2. Figure 3 outlines a sequence of changing, for a given tolane or ester

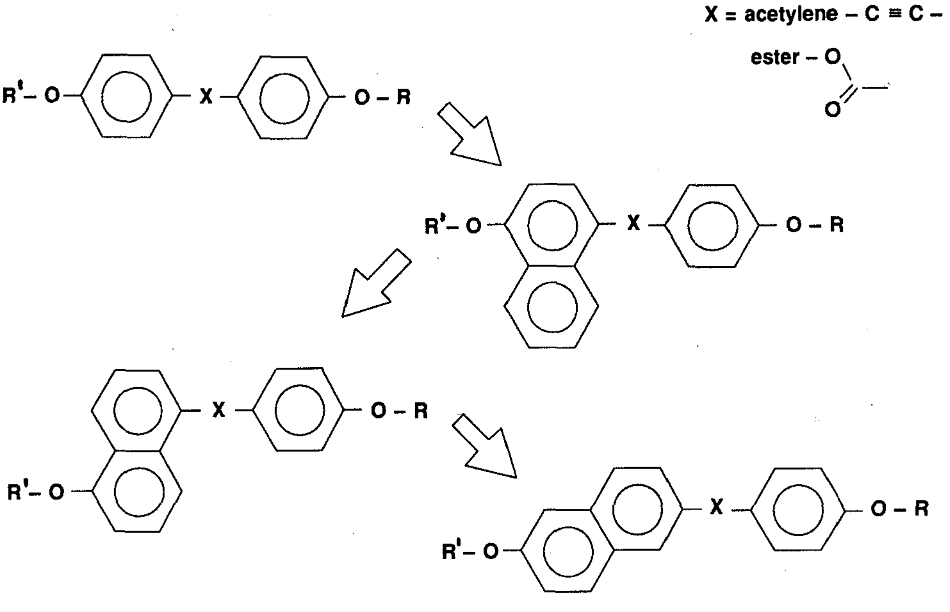

FIGURE 3 Naphthyl configuration compounds used in distinguishing π-electron delocalization effects on electronic nonlinearity. Alkyl chains $R = C_{10}H_{21}$, $R^1 = C_8H_{17}$.

linkage, the naphthyl configuration and concomittant π-electron delocalization in our group of compounds. The detailed syntheses for these materials will be described separately elsewhere.[12]

Since none of these materials exhibit any measurable linear absorption near 1053 nm, the nonlinear susceptibility measurements can be safely assumed to be nonresonant measurements. Nearly degenerate four-wave mixing at this wavelength has, in principle, been shown by Adair, Chase and Payne.[13] Our approach[14] simplifies theirs and results in the nonlinear mixing of two, temporally tightly synchronized, linearly polarized pulses separated in wavelength by 8 Å. This is an inherently background-free and virtually dispersion-free approach that offers attractive advantages over degenerate, phase-conjugation or third-harmonic measurement methods. As used here, the technique is essentially that of nonresonant coherent anti-Stokes Raman scattering.[15]

Samples were assembled in 500-µm path length, spectrophotometric cells with alignment coatings prepared by dip coating in 1% wt. solutions of poly-butylene terephthalate in p-chlorophenol, baking in air at 120°C and buffing with an aramid fiber roller for planar alignment. It was one of the key challenges of this effort to prepare cells that maintained acceptable alignment over that long a path length. This unusual path-length requirement arose from the signal-to-noise demands of the optical detection setup. Cells were mounted in a Mettler programmable hot-stage for both polarizing microscope phase studies, as well as for phase control during four-wave mixing experiments. An unheated, uncoated, 500-µm cell of CS_2 served as a reference sample.

The results available to date from these measurement are summarized in Figure 4. There the relative $\chi^{(3)}/n^2$ is ranked relative to CS_2 for the different compounds identified and for their respective phases. The linear refractive index n at 1053 nm of each compound has been measured separately on a temperature-variable Abbé refractometer. The following conclusions can be drawn from these data:

- Regardless of structure and phase, none of the tested compounds possessed a $\chi^{(3)}$ exceeding that of CS_2 at the nonresonant conditions and pulse lengths employed.
- The conjugation effect of the linkage clearly has an influence on enhancing the optical nonlinearity. Tolane-linked compounds exhibit larger $\chi^{(3)}$ than ester-linked systems. There is also a subtle but distinguishable difference in response between the two, asymmetric ester-linked systems.
- Much more significant than electron delocalization in the molecular core is the presence of polarizable push-pull substituents. The halogen substitution on

the chiral center of the biphenyl 2-chlorohexanoate raises $\chi^{(3)}$ close to that of CS_2.

- Within measurement error, results from measurements in the isotropic phase do not differ from those in the respective ordered phase, indicating a dominance of the electronic contribution to $\chi^{(3)}$ with little contribution from Kerr reorientation. At longer pulse lengths this is expected to change.[16]

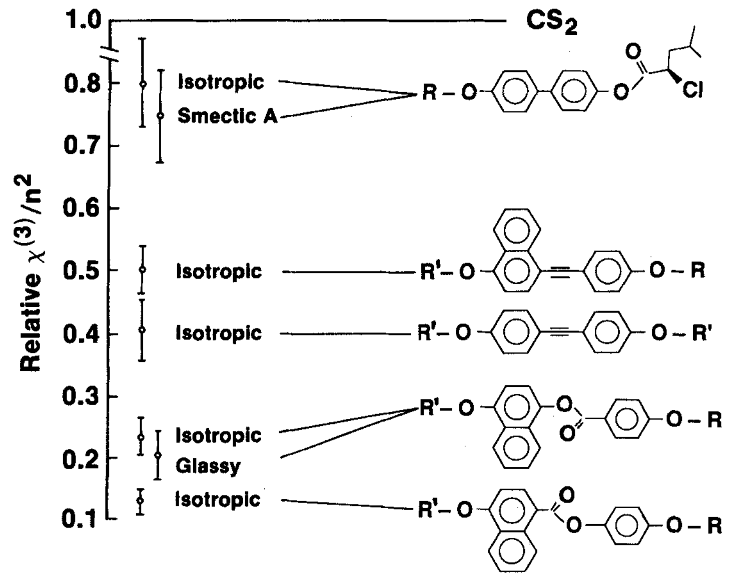

FIGURE 4 Ranking of measured $\chi^{(3)}/n^2$ relative to CS_2 for compounds listed in Figure 3 and decyloxybiphenyl-2-chlorohexanoate included to demonstrate the effect of a halogen push-pull substituent.

SUMMARY

The objective of this paper has been to describe development and performance issues of liquid-crystal applications in large laser systems. Among organic materials, liquid crystal monomers and polymers offer unique opportunities wherever scale, function, or other performance limits of conventional materials pose challenges.

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REFERENCES

1. R. S. Craxton, R. L. McCrory, and J. M. Soures, Sci. Amer. 255, 68 (1986).
2. S. H. Chen, M. L. Tsai, and S. D. Jacobs, in Materials Research Society Symposium Proceedings, A. Buckley, G. Gallagher-Daggitt, F. E. Karasz, and D. R. Ulrich, Eds. (Materials Research Society, Pittsburgh, PA, 1990), Vol. 175, p. 271.
3. M. Guardalben, A. Bevin, K. Marshall, and A. Schmid, Proceedings of the Twentieth Annual Symposium on Optical Materials for High Power Lasers, NIST Spec. Publ. 775 (Government Printing Office, Washington, DC, 1988), p. 462.
4. S. Papernov, K. Marshall, M. Guardalben, A. Schmid, and S. D. Jacobs, "351-nm, 0.7 μs Laser Damage Thresholds of Monomeric Liquid-Crystalline Systems," Liq. Cryst. (1990) (accepted for publication).
5. S. D. Jacobs, K. A. Cerqua, K. L. Marshall, A. Schmid, M. J. Guardalben, and K. J. Skerrett, J. Opt. Soc. Am. B 5, 1962 (1988).
6. T. E. Gunderman, J. C. Lee, T. Kessler, S. D. Jacobs, D. J. Smith, and S. Skupsky, Proceedings of the 1990 Conference on Lasers and Electro-Optics (Optical Society of America, Washington, DC, 1990), p. 354.
7. H. G. Winful, Phys. Rev. Lett. 49, 1179 (1982).
8. J. C. Lee, S. D. Jacobs, and A. Schmid, Mol. Cryst. Liq. Cryst. 150b, 617 (1987).
9. J. C. Lee, S. D. Jacobs, T. Gunderman, A. Schmid, T. J. Kessler, and M. D. Skeldon, Opt. Lett. 15, 959 (1990).
10. D. S. Chemla and J. Zyss, Eds., Nonlinear Optical Properties of Organic Molecules and Crystals (Academic Press, Orlando, FL, 1987).
11. B. I. Greene, J. Orenstein, and S. Schmitt-Rink, Science 247, 679 (1990).
12. K. Marshall, B. Puchebner, and C. LaPree, "The Effect of Positional Isomerism on the Liquid Crystalline Properties of 1,4-, 1,5-, and 2,6-Disubstituted Naphthalene Derivatives" (to be published).
13. R. Adair, L. L. Chase, and S. A. Payne, J. Opt. Soc. Am. B 4, 875 (1987).
14. Y. H. Chuang, Z. W. Li, D. D. Meyerhofer, and A. Schmid, "Nonresonant $\chi^{(3)}_{1111}$ Using Chirped-Pulse Technology" (submitted for publication).
15. Y. R. Shen, The Principles of Nonlinear Optics (John Wiley and Sons, New York, 1984), Chap. 15.
16. F. W. Deeg and M. D. Fayer, J. Mol. Liq. 45, 19 (1990).