



Organic Solid-State Lasers: Past and Future

A. Dodabalapur; E. A. Chandross; M. Berggren; R. E. Slusher

Science, New Series, Vol. 277, No. 5333. (Sep. 19, 1997), pp. 1787-1788.

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structural anisotropy on the molecular scale as well as a large flexibility in local bonding rearrangement in a disordered network. If we consider the chalcogenide glass as a randomly connected microvolume, each of which carries an independent anisotropic microstructure, polarized light can preferentially excite microvolumes having their optical axes along the electric field of the light. Thus, as has been pointed out by Fritzsche (4), a net anisotropy of the system can be realized only when hole-electron pairs recombine nonradiatively in their excited microvolumes, whereby some atomic rearrangements occur and rotate optical axes of some of the excited microvolumes. Occurrence of excitation and nonradiative recombination in the same microvolume is essential to the existence of vector effects, whereas diffusion of excited carriers out of microvolumes results in scalar effects, be-

cause carriers lose the memory of preferentially excited microvolumes.

Regarding more detailed mechanisms for light-induced optical and structural anisotropy, several structural models have been proposed, but they are still controversial. Many of them emphasize isolated short-range atomic events such as bond switching (5), modification of defects (6), or LP orbitals, whereas Lee *et al.* (7) and Tanaka *et al.* (8) have suggested more cooperative and longer range structural modification of the network. Krecmer's result favors the latter. Furthermore, quite recently, it has been reported that chalcogenide glasses behave like liquids in their local structure under photoexcitation with respect to scalar effects (9). This result suggests that a disordered network of chalcogenide glasses is considerably dynamic under photoexcitation and possibly changes its middle-range order or sometimes

long-range order through the cooperative effect of local atomic events.

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APPLIED PHYSICS

Organic Solid-State Lasers: Past and Future

A. Dodabalapur, E. A. Chandross, M. Berggren, R. E. Slusher

A world-wide race is on to achieve electrically pumped lasing in organic materials. Organic solid-state lasers have the potential to provide a compact, low-cost optical source over a broad range of wavelengths throughout the visible spectrum. Organic solid-state lasers are also an excellent example of how basic research in one area can influence many other fields, including some of considerable technological significance. Demonstrations of light amplification and laser action in photoexcited solid-state organic materials (1) rapidly followed the invention of liquid dye lasers (2). Since then, the results of organic laser research have had a substantial impact on many aspects of laser physics. The ability to define phase gratings holographically in organic materials made possible the first demonstrations of distributed feedback (DFB) and distributed Bragg reflector (DBR) lasers (3, 4) (see figure). These lasers had gain media consisting of a polymer host such as poly(methyl methacrylate) or gelatin doped with rhodamine 6G as a film on a glass substrate. Gratings that function as wavelength-selective mirrors are placed at the ends of the cavity in DBR lasers. The cavity

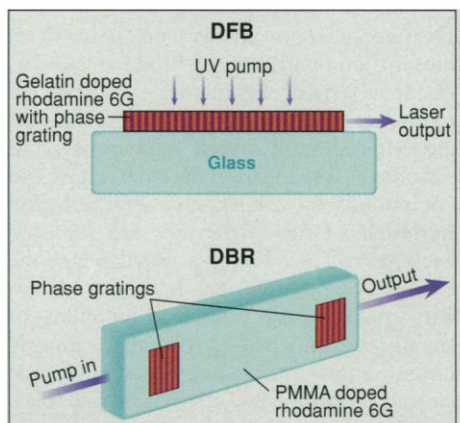
and gratings are part of a waveguide that confines light along the plane of the active material. Upon photoexcitation, some of the light emitted is amplified in the cavity, and the mode that has the highest net gain becomes the lasing mode above threshold.

The successful realization of DBR and DFB lasers with dye-doped polymers was followed by efforts to replicate these resonator

designs with III-V semiconductors such as GaAs, but success was slow to come because of processing difficulties. Eventually these difficulties were overcome, and both photopumped (5) and electrically driven DFB (6) lasers were demonstrated. Subsequent advances led to the widespread use of InP-based DFB and DBR lasers in long-haul optical communication systems. These lasers are favored because of their superior single-frequency operation and high-speed modulation characteristics compared to conventional Fabry-Perot lasers.

Research on organic gain media—liquid and solid—was useful in clarifying many aspects of the operation of vertical-cavity surface-emitting lasers (VCSELs). For example, work with organic lasers improved understanding of the influence of the spontaneous emission factor β on the threshold characteristics of planar microcavity lasers (7, 8), which are similar to VCSELs. The factor β is the fraction of the total spontaneous emission that is coupled to the lasing mode below threshold. Microlasers with β close to 1 can have very low thresholds. The VCSELs based on III-V semiconductors are technologically important because of the compact volume, low-threshold currents, and high coupling efficiency to single-mode optical fibers.

Fluorescent organic dyes have been used to study the properties of spherical and near-spherical resonators (9). The lasing modes of such resonators are called “whispering gallery” modes or morphology-dependent resonances and are the optical analogs of acoustic whispering galleries that are found in buildings such as St. Paul's Cathedral in London. Experimental work on lasing in liquid droplets containing laser dyes (9) led to re-



Schematic structures of the first DFB (top) and DBR (bottom) lasers. In the DFB laser, feedback is provided throughout the gain region by a phase grating that was defined in the gelatin by intersecting ultraviolet beams. In the DBR laser, the gratings are defined at the ends of the laser cavity. [Adapted from (3, 4)]

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cent theoretical studies of the effects of chaos, both classical and quantum, in asymmetric resonant cavities (10).

There are also examples of microlaser structures that were first demonstrated with inorganic semiconductors and were subsequently implemented with organics. The GaAs and InP microdisk lasers have the very small volumes and low-threshold power and current densities (11). The lasing modes of these thumbtack-shaped lasers are also whispering-gallery modes. Microdisk lasers with molecular doped polymer (12) and sublimed organic active layers (13) have been recently reported. These microlasers are of interest because of their simple fabrication and potential to lower threshold pump powers as a result of strong emission coupling to low-loss microcavity resonances.

In addition to being useful in investigating the properties of novel resonators, organic solid-state gain media have been considered for possible replacement of liquid dye lasers. This goal was one of the motivations for the earliest work on light amplification in organic films (1) and is still an area of active research, and some solid-state organic laser dye materials have been marketed. However, their durability has not been adequate to lead to large-scale replacement of liquid dye lasers.

Developments in organic laser dyes also influenced the design of efficient thin-film organic light-emitting diodes (LEDs) (14), in which the emissive layer consists of tris (8-hydroxyquinolinato) aluminum (Alq) doped with small amounts (~1%) of a laser dye such as DCM. The excited states created electrically in the host Alq excite the dye dopant through a process known as Förster energy transfer. The resultant emission spectrum is primarily that of the dye, and the quantum yield is enhanced with respect to that of a film of only the host Alq. Such dyedoped emissive layers are used in the most efficient and reliable organic LEDs reported to date (15). Advances in efficiency and lifetime have also been made in LEDs with conjugated-polymer emissive materials (16). These improvements in LED performance characteristics have stimulated fresh interest in organic laser research. For example, advances in conjugated-polymer design and synthesis have led to several recent reports of stimulated emission from films that can have high photoluminescence quantum yields and high optical gain (16, 17).

Electrically driven lasing in organics is being eagerly sought. Organic materials generally have emission spectra that are red-shifted with respect to their absorption spectra, which makes it easier to create population inversion in comparison with inorganic semiconductors, which must first be made transparent because the large absorption in the

unpumped state is several orders of magnitude higher than that in organics. The main difficulty in realizing organic diode lasers is that the low carrier mobilities in these materials make it difficult to create carrier densities high enough to generate the gain necessary to overcome losses in the laser cavity. There are other challenges as well: The device technology is still in its infancy. Important design issues have to be addressed and advances in fabrication technologies are needed. There are encouraging developments. Progress has been made in achieving optically pumped laser action in materials or material combinations that are capable of charge transport (13, 16, 17). Photopumped lasing has been demonstrated in resonator structures such as microdisks and planar microcavities, which are quite easily adapted for electrical injection. However, much remains to be done, and considerable effort is being expended to bridge the gap that exists between present technology and a diode laser. This laser research should help LED technology as well.

Organic laser materials, particularly the recently improved materials, will continue to be useful in researching the properties of resonators. There is currently much interest in lasers based on the photonic band-gap concept, where the separation between theory and experiment is remarkably wide (18). Organic gain media have a number of properties that are particularly suitable for these

geometries. For example, they can be quite easily made to fill the subwavelength-sized holes and crevices that are characteristic features of photonic band-gap structures. The accelerated pace of research will make the future of organic solid-state lasers at least as interesting as the past.

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BIOCHEMISTRY

Creating Isoprenoid Diversity

James C. Sacchettini and C. Dale Poulter

Nature relies on an intricate network of biosynthetic pathways to produce the cornucopia of small organic molecules needed to support life. Among these, the isoprenoids are extraordinarily diverse in chemistry and structure. Over 23,000 individual isoprenoid compounds have been characterized, and hundreds of new structures are reported each year. They serve as visual pigments, reproductive hormones, defensive agents, constituents of membranes, components of signal transduction networks, mating pheromones, and photoprotective agents, to name only a few of their many roles.

Abnormalities associated with the pathway can cause coronary heart disease and cancer, while at the same time some isoprenoid compounds, such as taxol, offer promise as potent new drugs.

Most of the molecular diversity in the isoprenoid pathway is created from the diphosphate esters of simple linear polyunsaturated allylic alcohols such as dimethylallyl alcohol (a 5-carbon molecule), geraniol (a 10-carbon molecule), farnesol (a 15-carbon molecule), and geranylgeraniol (a 20-carbon molecule). The hydrocarbon chains are constructed one isoprene unit at a time by addition of the allylic moiety to the double bond in isopentenyl diphosphate, the fundamental five-carbon building block in the pathway, to form the next higher member of the series (see the figure). Geranyl, farnesyl, and geranylgeranyl diphosphates lie at multiple branch points in the isoprenoid

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