

Acknowledgment

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PHOTONIC BANDGAP STRUCTURES

Hexagonal Resonators for Molecular Sieve Microlasers

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Molecular sieves are crystalline solid-state materials with crystallographically defined nanometer-size pores. This property stimulates attempts to use the sieves as an inert ordering framework for the arrangement of optically functional molecules, to create a compound material with new optical properties. For example, many organic molecules with large nonlinear hyperpolarizability crystallize in a centrosymmetric structure, and as a result the macroscopic even-order susceptibilities vanish. It has been shown that such molecules can be arranged in molecular sieves with a noncentrosymmetric symmetry.¹

Based on this idea, we investigated the aluminophosphate sieve, $ALPO_4-5$, which exhibits channellike pores with a diameter of 0.73 nm, which is sufficiently large to consider the inclusion of some organic laser dyes. In Ref. 2 we reported laser action in the $ALPO_4-5$ molecular sieve, in which the dye 1-ethyl-4-(4-(p-dimethylaminophenyl)-1,3-butadienyl)-pyridinium perchlorate (pyridine 2) was caged. In Ref. 3 we discussed the results with derivatives of Rhodamine dyes. With an approximate diameter of 0.69 nm, pyridine 2 molecules align with the 0.73-nm-wide channel pores of the $ALPO_4-5$ molecular sieve, which acquires strong dichroic and birefringent properties. Lasing in this compound material occurs in micrometer-size ring resonators, in which the laser mode is confined by total internal reflection at the natural hexagonal crystal side faces such as in a whispering gallery mode. Inasmuch as the compounds are synthesized by the inexpensive method of hydrother-

mal growth, we have already been successful in obtaining several grams of microlasers or lasing powder.

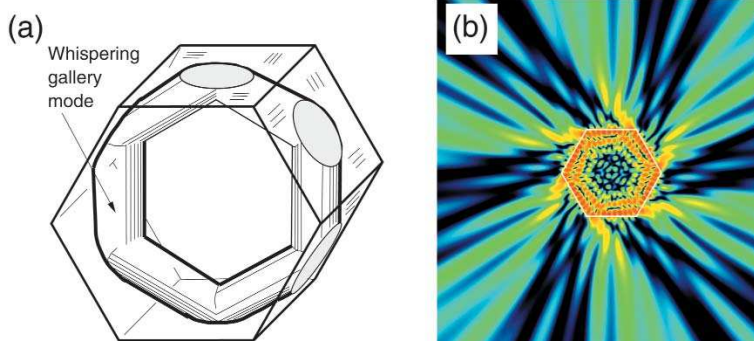
Figure 1(a) is a schematic picture of the whispering gallery lasing mode in a hexagonally bounded dielectric prism. In contrast with this picture, the experiment clearly revealed that the laser light exits from the prism resonator at the edges. The main feature that distinguishes the hexagonal resonator from other common whispering gallery type cavities, such as microdroplets or disk lasers, is that the latter do not exhibit sharp corners and flat sides. The hexagon in fact constitutes a self-assembled realization of a pseudointegrable structure.

An orthogonal coordinate system in which the wave equation can be solved by separation of variables does not exist. Figure 1(b) shows the result of a numerical simulation of the field distribution of a counterclockwise propagating mode with 685-nm wavelength in a hexagon with a 4.5- μm width over flats. The simulation correctly reproduces the outcoupling location of the field at the corners. Upon closer examination one can see that the number of ridges and nodal lines is not uniquely defined (optical defects). As a result, the modes in a hexagonal resonator cannot be labeled properly by good quantum numbers, which is a direct consequence of the nonintegrability of the problem.

The photostability of organic dyes is a critical issue. In traditional, liquid dye lasers, photobleaching is irreversible. Strong pumping bleaches the caged dye molecules as well. However, we observed that the fluorescence of caged pyridine 2 molecules recovers with a time constant of approximately 2 min. The physical mechanism behind this recovery is not yet understood.

References

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Hexagonal Resonators Figure 1. (a) Schematic representation of the lasing mode in a hexagonal ring resonator. (b) Numerical simulation of the field distribution in a hexagonal dielectric resonator with 685-nm wavelength and a 4.5- μm hexagonal width over flats.