The mode structure of microcrystal and microdroplet lasers

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Yearbook of the Max-Planck Institute for
Physics of Complex Systems, 1996-1997

The work horse of optics is the *ray picture* – i.e., the short-wavelength limit of Maxwell's wave equations. In the form of the paraxial approximation, it is also the backbone of conventional optical resonator theory. Without such approximations, one is forced to resort to numerical solutions of the full wave equations, which in general reduces resonator design to trial-and-error. An exception are those few systems for which exact solutions can be obtained due to their symmetric, *separable* geometry.

Short-wavelength (or quasiclassical) approximations appear to be as important for light as they certainly are for electrons, and hence it is somewhat surprising that classical concepts such as "diffusion" familiar from condensed matter physics have not played a significant role in optical resonator design until recently. That is so because the engineer often has the freedom to choose geometries for which either the ray picture is simple or the wave equation is separable (up to small perturbations).

This is a luxury that we do not usually have with optical systems that occur in nature – specifically *self-assembled* dielectric microresonators formed from materials as diverse as aerosol droplets or microcrystallites. We study these optical cavities since they exhibit properties that are desirable for artificially patterned micro-optical devices as well. Because of this, the ray picture with its explanatory and predictive power is again of value. However, ray and wave properties combine in new ways when diffraction and interference compete with (a) *ray chaos*, induced by unconventional resonator geometries, and (b) the *openness* of the system due to its coupling to the environment.

Among the pioneering experiments in microcavity optics were spectroscopy and imaging of liquid **microdroplets** falling in air[2]. If they contain a suitable organic dye, excitation by a pump beam can cause these droplets to act as extremely efficient lasers. The feedback required for lasing is provided by long-lived modes whose emission properties are found to depend sensitively on the *shape* of the droplet. To understand these "morphology-dependent resonanaces", ray optics is an excellent framework: firstly, the droplets' diameter of $30 - 100\,\mu$m is much larger than optical wavelengths; furthermore, the mechanism by which the light is trapped in the droplets is *total internal reflection* at

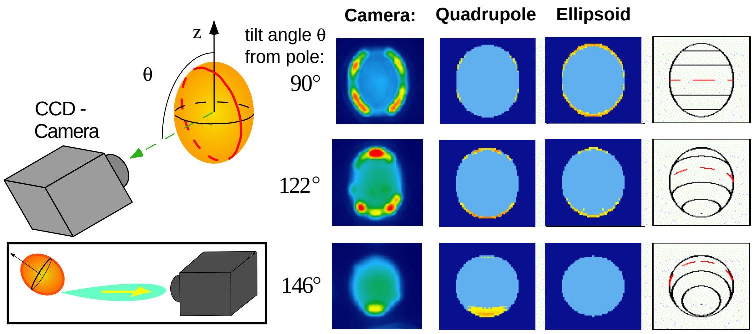

Figure 1: Left: experimental setup, showing the tilted CCD camera and the droplet it images from different directions. Right: recorded CCD images at three tilt angles θ, compared to two ray simulations, differing only in the assumed droplet shape. Although a quadrupole and ellipsoid shaoe appear very similar to the eye, only the quadrupole agrees with experiment. The latitude circles as seen by the camera are shown in the rightmost column for orientation.

the interface to the outside air – a classical phenomenon in that it occurs almost indepently of wavelength.

Whereas spherical droplets are separable systems, generic oval droplets are not. The latter exhibit highly anisotropic emission directionality, characteristic spectral line shifts and degradation of their resonance lifetimes[1]. In the ray dynamics, chaotic and non-chaotic (“regular”) types of motion coexist. An important exception are oval cavities with the shape of a mathematical *ellipsoid*, for which no chaos exists. This raises the question if chaos can be relevant at all in oval droplets, because their shape could be approximated by an ellipsoid. Possible corrections could then be obtained by a perturbation approach.

The subtle distinction between an ellipsoid and the *quadrupolar* oval leads to qualitatively different predictions for the laser emission directionality in a geometric-optics simulation, cf. Fig. 1. Comparison with recent position - and angle - resolved imaging measurements unambiguously determines that the droplets are indeed quadrupoles, and *not* ellipsoids[3]. The emission directionality is thus a sensitive probe of the internal ray dynamics. Its structure is properly revealed only if one portrays the rays not in real space, but in a *phase space* spanned by the positions and orientations at which rays impinge on the surface. The lasing modes are found to be supported by rays that *diffuse in phase space* but are guided by stable structure which arises as a non-perturbative consequence of the oval shape.

Because of their low refractive index, only a small portion of the phase space is available for long-lived modes in the droplets. The same holds for **zeolite microcrystals** which have recently been discovered as a novel class of composite laser media[4]. Encapsulated in the hexagonal nanopore matrix of the zeolite are dye molecules of the same type as in the droplet example, but the porous host material serves to impose order and stability on the fluorescent “guest” molecules. This leads to novel optical properties, among them polarized emission and lasing, in microcrystallites with hexagonal facets at less than 10 μm diameter, cf. Fig. 2 (a,b). The pump thresholds required for lasing can be as small as that of semiconductor based vertical-cavity surface-emitting lasers. The lasing modes exhibit a frequency spacing consistent with the optical paths shown in Fig. 2 (a). An infinite family of such periodic orbits with identical lengtsh exists, held in the cavity by internal reflection at the facets. Total internal reflection requires plane interfaces, which is well satisfied except at the corners. But the latter are in fact reached by a “degenerate” member of the periodic-orbit family in Fig. 2 (a).

As expected, experiment and numerical wave calculations, Fig. 2 (b) and (c), reveal that most oft the emission originates at the corners, whereas the internal intensity is distributed evenly over the whole periodic-orbit family of Fig. 2 (a). In contrast to the smooth droplets, the *directions* into which the light is radiated by these hexagonal crystals cannot be explained by geometric optics, because the radius of curvature at the corners is much shorter than the wavelength, leading to *corner diffraction.* In a simulation which approaches the limit of sharp corners starting from a shape with rounded corners via a continuous reduction of the radius of curvature ρ, one finds a crossover in the spectrum from level repulsion for $\rho \gg \lambda$ (=wavelength) to pronounced near-degeneracies which develop and stay practically unchanged as soon as $\rho < \lambda$. This qualitative change is indicative of a transition from quantum chaos to *pseudointegrability* (a nonseparable system without chaos); but it is not accompanied by a similar transition in the emission directionality: over a large range of ρ above and below λ, the light emanates from the crystal approximately parallel to its facets. Intruigingly, that is in fact what the ray picture predicts for the case of rounded corners where it is again applicable.

References

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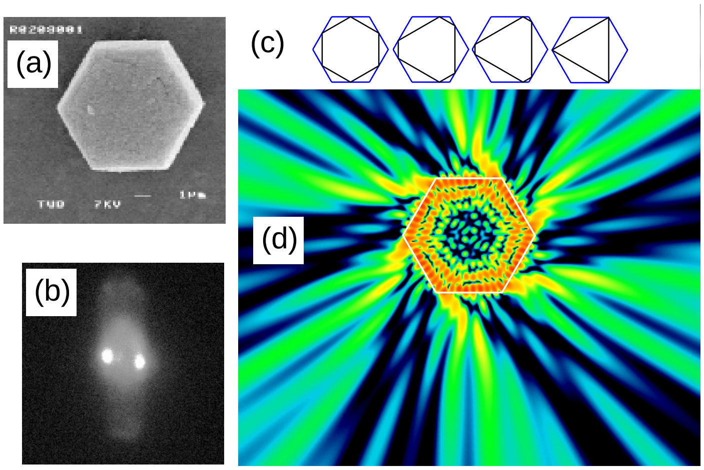

Figure 2: Hexagonal dye-doped zeolite crystals: cross section (a) and laser emission (b). Periodic ray orbits in (c) are internally reflected, except for the rightmost one entering the corners. The cross-sectional field intensity (d) from numerical solution of the wave equation shows emission predominatly from the corners.