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Directional emission from asymmetric microlaser resonators of π-conjugated polymers

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A π-conjugated polymer film was fabricated into an asymmetric microlaser resonator having a quadrapole deformation with irregular boundaries and a Q factor of 600. At high excitation intensities above the threshold for lasing, we observed multimode laser emission spectra and *directional emission* at four different angles. Chaotic ray dynamics explains the observed emission pattern. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1753064]

Microdisk resonators are low threshold laser devices and have been used to demonstrate lasing in a variety of materials.[1–5] Unfortunately they are of limited practical use because laser emission is isotropic in the plane of the disk. One of the more successful attempts to obtain directional emission has been to use shapes that deform the circle.[6]

We have fabricated an asymmetric microlaser resonator from a π-conjugated polymer film, poly(dioctyloxy) phenylene vinylene or DOO-PPV,[7] which has a lower refractive index of 1.8 compared to the inorganic materials, and a peak emission near 630 nm. The polymer microresonator was fabricated using photolithographic methods and is a variation of the well documented microdisk laser.[1,5] The intended boundary deformation can be described in the r,θ plane by the following equation:

$$r=\sqrt{1+2\epsilon\cos(2\theta)}. \tag{1}$$

This shape is completely convex for values of ϵ up to 0.2. After that, the perimeter near 90° and 270° pinch in and the boundary begins to resemble that of the number 8. For small values of ϵ in the range of 0.04 to 0.08, however, the shape is barely different from a circle. Though the shape of a deformed quadrapole resembles a circle, the radius at 0° is larger than that at 90° and this destroys the rotational invariance. An optical microscope image shows the boundary of the obtained cavity in Fig. 1. The boundary is far from smooth.

The polymer resonator was excited above laser threshold with the second harmonic of a Nd:YAG regenerative amplifier at 532 nm with 100 ps pulses and a repetition rate of 100 Hz. The sample was placed in a vacuum chamber to reduce photo-oxidation. The sample could be rotated 360° inside the chamber with an angular resolution of about 1°. The laser emission from the microcavity was collected with a fiber optic inside the vacuum chamber, sent to a 1/2 meter spectrometer and recorded with a charge coupled device camera. The total spectral resolution was 0.02 nm.

The laser emission spectra for two angles of the quadrapole excited above the threshold are shown in Fig. 2(a). For each angle, there are numerous modes with widths less than 1 nm. However, note the difference in scales of the two spectra. *The intensity of the largest peak at 81° is roughly ten times the intensity of the highest peak at 0°*. The modes are rather broad indicating that the overall quality factor is low. The quality factor of a resonator is $\lambda/\delta\lambda$; this is correct only if the resonator is below lasing threshold. However, the value does not change too much once the device crosses the lasing threshold.[8] Thus, from the emission spectrum, the mode width for the peak at 627 nm is 1 nm that gives a Q factor ≈ 600.

Emission spectra were recorded every 9° for the entire boundary of the quadrapole. Figure 2(b) shows the angular dependence of the integrated emission intensity. The emission intensity is normalized to that at $\theta=0$. The ratio of the integrated intensity between 81° an 0° is about 25; a strong preferred directional emission is observed. As a system check, the angular intensity of a microdisk was also measured, as shown in the inset of Fig. 2(b). For the circle, the ratio of highest to lowest intensities is roughly 2.

Previous numeric studies of an elliptical microcavity showed that maxima emission occur at the highest radius of curvature.[9] For the quadrapole, the highest curvature occurs at $\theta=90°$ and 270°; indeed there are maxima in the emission pattern at about these angles (Fig. 2). However, it also appears that the intensity is split between two neighboring angles both at "90°" and "270°." The fingerprint of a classical ray phase space would give the most intense emission at the greatest curvature of the resonator; since this is not

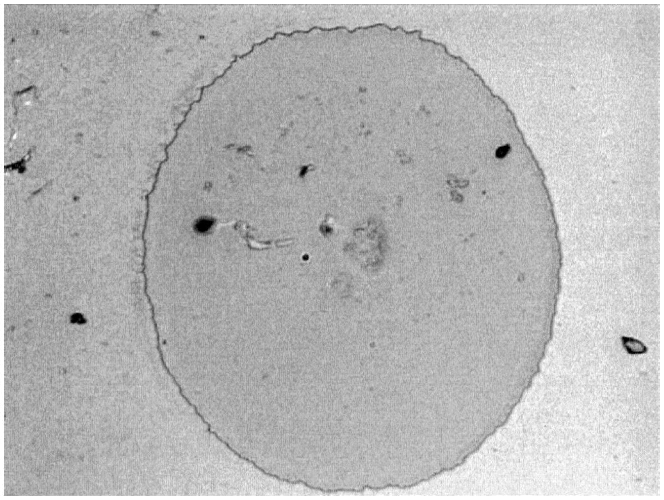

FIG. 1. Optical microscope of the quadrapole microcavity fabricated with $\epsilon=0.04$ on the DOO-PPV polymer film. The physical dimensions of the major axis are roughly 200 and 210 μm.

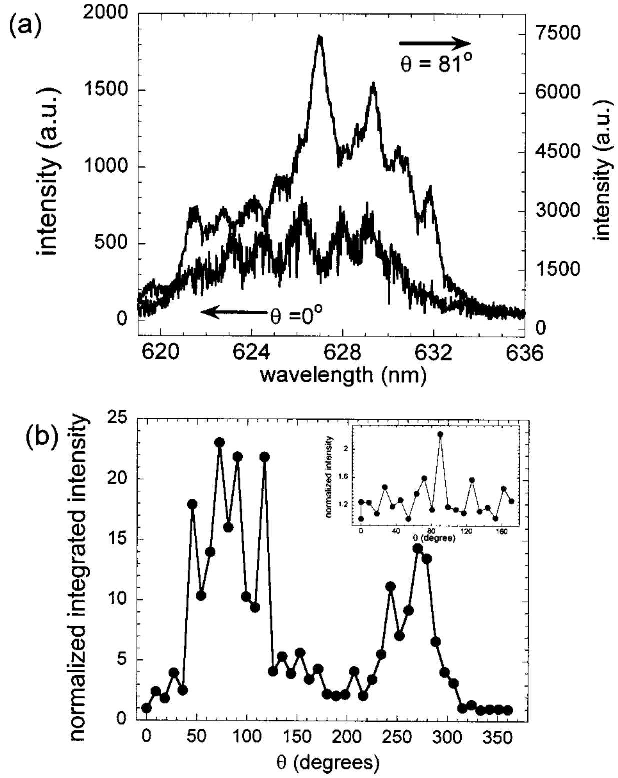

FIG. 2. (a) Laser emission spectra for two angles of the polymer quadrapole resonator with $\epsilon=0.04$; notice the different scales for the two angles. (b) Angular intensity dependence of the assymetric microlaser normalized to the intensity at $\theta=0°$. The inset is for a microdisk with a circular boundary.

quite the case for our asymmetric resonator, a possibility for explaning this is chaotic ray motion.

The chaotic variable is the light ray's angle of incidence upon the boundary. A simple billards model can be used to investigate the ray motion upon the boundary. For a microdisk with a completely circular boundary, the ray hits the boundary at the same angle everywhere. Since the circular boundary is rotationally invariant, if the ray hits the boundary in a slightly different location, the resulting angle of incident would still be the same. For the asymetric resonator, however neither of these is true. A ray typically hits the boundary at different angles depending on the location at the boundary. A small difference along the boundary can lead to a very large difference in the ray path. Figure 3 shows the angle of incidence of a ray on a circular boundary and for a one of numerous possible paths on deformed quadrapole. For the circle, the angle of incidence is always the same, whereas for the quadrapole it varies greatly.

Microdisks with a circular boundary lead to laser action by confining the light due to total internal reflection. The disk thickness is typically 1 to 2 wavelengths thick, lateral confinement is a waveguide mode between the polymer film, the surrounding air and lower index substrate. If a ray strikes the circular boundary at an angle greater than the critical angle, it is totally reflected. The types of rays paths that are favored are whispering gallery modes, where most of the field is concentrated near the outer 5% of the circular radius.[10] Surface roughness and edge irregularities adversely effect both the lasing threshold and light confinement. The rough boundary of the asymetric microresonator device in Fig. 1 should scatter so much light that no whispering gallery

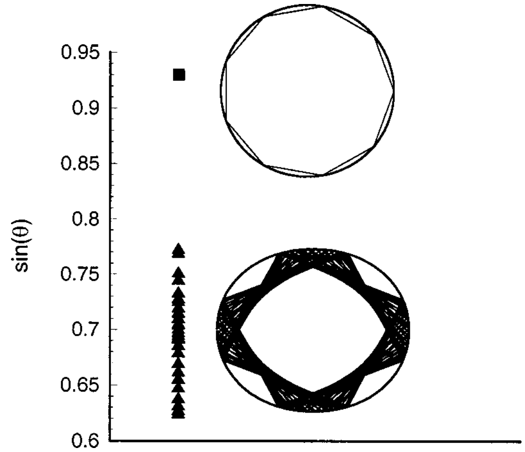

FIG. 3. Ray bounce plots of circle, top, and quadrapole, bottom. The angle the ray hits the circluar boundary is always the same; $\sin(\theta)$ is constant (square). One path is shown for the quadrapole; the angle the ray hits the boundary changes over a wide range (triangles).

modes can form. Even with a small periodic variation, the cavity quality factor steadily decreases for higher-order modes.[11] In Fig. 1, the amplitude of the edge variation is greater than the wavelength, and thus strong scattering is expected. However, since we observe laser emission, another ray motion must be occuring. Chaotic ray motion becomes the most likely candidate. The basic shape of this resonator is a deformed quadrapole, a well studied chaotic resonator.

Certain two- or three-dimensional shapes can lead to chaotic ray dynamics. Nonchaotic high-quality factor cavities have been observed with perfect disks and spheres,[1,12] where the light is confined by total internal reflection and propagates near the surface in a whispering gallery-type mode. However, experimental studies of oscillating laser dye droplets showed lasing even when the shapes were *highly distorted* from spherical.[13] These *deformed* shapes are a class of resonators known as asymmetric resonators, which have the properties of directional emission and chaotic ray dynamics inside the resonator.[9,14,15] Experimental studies of lithographic asymmetric microresonators were reported on semiconductor materials with emission at 5.1 μm,[6] and at a longer wavelength of 10 μm.[16] Both of these reports showed directional emission that indicated chaotic behavior.

For a whispering gallery mode, the ray does not penetrate very far into the bulk of the resonator. In a chatoic resonator, the ray passes much further into the main body of the resonator. The rays experience more gain and are able to cross the lasing threshold before being scattered by the rough boundary. The combination of chaotic ray motion and irregular boundary selects some complicated ray motions that experience enough gain to lase.

In conclusion, we have fabricated a π-conjugated polymer asymmetric resonator that shows directional emission and chaotic ray dynamics. The lower index of refraction of 1.8 for the polymer gain medium allows the observation of these effects at lower deformations than previous works with index of refraction near 3.[6]

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