Plastic microring lasers on fibers and wires

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(Received 11 December 1997; accepted for publication 12 February 1998)

Photopumped, pulsed, narrow line laser emission is demonstrated using cylindrical microcavities formed by π-conjugated polymer thin films wrapped around thin glass optical fibers and metal wires with various diameters D. A variety of cavity-dependent resonant laser mode structures were observed, which for $D<10$ μm contain a single resonant spectral line of less than 1 Å in width. The microring lasers are also characterized by a well-defined, very low threshold excitation intensity, at which beam directionality and polarization degree dramatically increase. These findings open up the fields of lasers and fiber optics to organic materials. © *1998 American Institute of Physics.* [S0003-6951(98)00115-6]

Low-energy excited states in luminescent conducting polymers (LCP) are excitonic in nature,[1] similar to those of common laser dyes. During the past few years, lasing has indeed been demonstrated in solutions of several LCP.[2] Unlike laser dyes, however, LCP still exhibit strong optical gain even in the solid state.[1] In fact, stimulated emission has been observed in thin LCP films at high photoexcitation densities, resulting in spectral narrowing of the polymer emission.[3–7] This spectral narrowing has been attributed to lasing,[3] amplified spontaneous emission,[4,5] and superfluorescence.[6,7] These phenomena require that the optical gain exceeds all combined optical losses, such as self-absorption, excited-state absorption, light scattering, and diffraction. Lasing, in addition, requires an optical cavity to produce optical feedback, which subsequently forms resonant laser modes. Planar microcavities[3] have been used to demonstrate photopumped laser action in poly(*p*-phenylene–vinylene) (PPV) films. Lasing has also been shown recently in various blends of small luminescent organic molecules.[8,9] Moreover, since doped π-conjugated polymers are able to efficiently conduct electrical current, then ultimately it might be possible to produce an electrically pumped, all plastic laser diode.

Successful fabrication of laser diodes from LCP requires a sufficiently low laser threshold current. Therefore, in order to achieve the lowest possible laser threshold, it is first necessary to optimize the performance of photopumped polymer lasers. Microcavities (μcavities) have been shown to furnish an excellent quality factor Q and optimal coupling between emission and cavity modes, which results in low laser thresholds.[10] Although planar μcavity LCP lasers are compatible with the present organic light-emitting-diode technology, and this is desirable for electrical pumping, they suffer from unavoidable losses due to imperfect reflections and emission leakage to the sides of the μcavity plane. Hence, the lowest laser threshold excitation intensities cannot be achieved in planar μcavities. Cylindrical or ring μcavities, on the other hand, provide superior two-dimensional optical confinement resulting in Q values in excess of 10^6,[11] and thus are much more advantageous for obtaining low laser thresholds.[12,13]

In this letter, we demonstrate that photopumped, pulsed, narrow laser emission lines with very low threshold excitation intensities can be obtained using LCP films deposited around thin optical fibers and metal wires. For the laser active material we have chosen a derivative of PPV, namely, 2,5-dioctyloxy PPV (DOO-PPV).[14] DOO-PPV has been shown to be an excellent laser-active medium in the red/yellow spectral range.[6,13] The lowest excited states in DOO-PPV are excitons with energy levels similar to those of organic laser dyes, which under optical excitation form a four-level laser system. The polymer laser transition then occurs at longer wavelengths compared to the pump wavelength, and thus, population inversion can be achieved at relatively low excitation densities.[4,6]

To form ring μcavities, optical fibers of various diameters D ranging from 5 to 200 μm were dipped into saturated chloroform solutions of DOO-PPV. The subsequent fast evaporation of the wet polymer film resulted in a self-assembly process in which a thin polymer film was uniformly coated around the fibers (Fig. 1, upper inset). Thin polymer films of about 1 μm in thickness forming a complete cylinder of 100 μm or more in length were obtained in this way. In order to produce thicker films, this self-assembly procedure could be repeated several times. Optical excitation of the ring μcavities was provided by a frequency-doubled output (532 nm) of a Nd:YAG regenerative amplifier producing 100 ps pulses at a 1 kHz repetition rate. The pump beam was focused by a cylindrical lens on one side of the polymer ring, while the polymer emission was collected on the other side by a round lens in the plane of the illuminated ring and then analyzed using a 0.6 m spectrometer and a charge-coupled device camera with ~ 1 Å spectral resolution. In order to minimize photo-oxidation, all samples were contained in a chamber with flowing N_2 gas at room temperature.

The electrodynamic solution for a polymer ring μcavity contains a set of resonant modes, of which intensity distribution depends on the thickness d of the film. At $d<5$ μm the film acts as a curved two-dimensional waveguide and the corresponding resonances in this case are waveguided modes

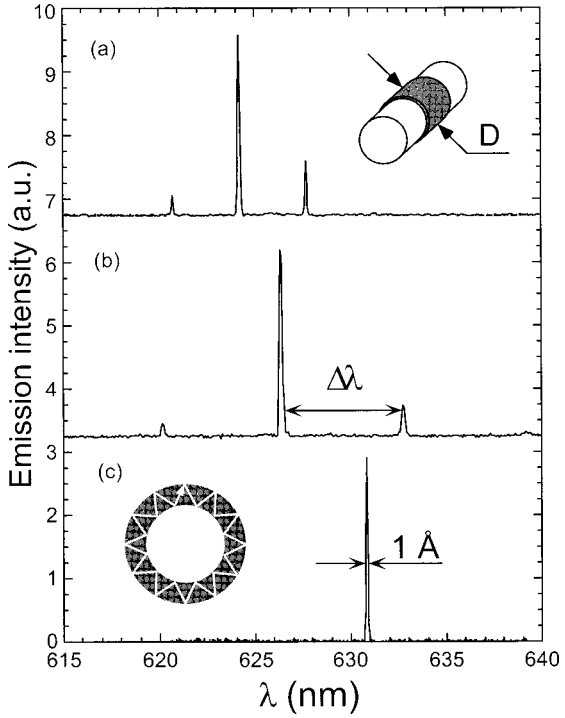

FIG. 1. Normalized emission spectra of lasing μrings formed around thin glass fibers measured at excitation intensities close to the laser threshold excitation intensity; the upper spectra are given with vertical offsets. The fiber diameters D and corresponding intermodal spacing $\Delta\lambda$ are 20 μm and 3.5 nm (a), 12 μm and 6.4 nm (b), and 5 μm and >10 nm (c), respectively. The upper inset shows the structure of the μring laser, and the lower inset illustrates schematically the propagation of waveguided modes inside the μring.

(WM) (Fig. 1, lower inset). The resonant WM wavelengths λ_m are given by[11–13]

$$m\lambda_m = \pi D n_{\text{eff}}, \tag{1}$$

where m is an integer and n_{eff} is the effective polymer refraction index, which in our case is close to that of the bulk, $n_p \sim 1.7-1.8$.[13] In thicker polymer rings ($d/D \geqslant 0.2$), however, the resonant modes are no longer waveguided.[15] In fact, these optical modes never reach the film/core interface, and therefore, are dubbed whispering gallery modes (WGM).[10,11] The light in this case is confined by total internal reflections at the film/air interface, and the mode intensity distribution is concentrated on the outer edge of the film. The WGM wavelengths are still given by Eq. (1) with different n_{eff}.[11]

Figure 1 shows the laser emission spectra obtained from DOO-PPV μrings deposited on glass fibers of three different diameters. Their characteristic feature is the narrow equidistant emission lines, which appear above I_0 of order 0.1 nJ/pulse (Fig. 2). The laser emission wavelengths are close to the maximum of the optical gain spectrum, which in DOO-PPV peaks at 630 nm.[6,13] The narrow spectral width of these laser lines, of about 1 Å, is limited by the resolution of our apparatus and is consistent with the high Q estimated previously for DOO-PPV ring μcavities.[13] The spectra in Figs. 1(a) and 1(b) are typical of multimode laser emission. The wavelength of each mode is determined by Eq. (1), and the measured intermodal spacing $\Delta\lambda$ are in excellent agreement with those calculated using $\Delta\lambda = \lambda_m - \lambda_{m-1} = \lambda^2/\pi D n_{\text{eff}}$ with $n_{\text{eff}} = n_p$. Since the spectral width Γ of the optical gain in DOO-PPV is about 10 nm,[13] and does not depend on the

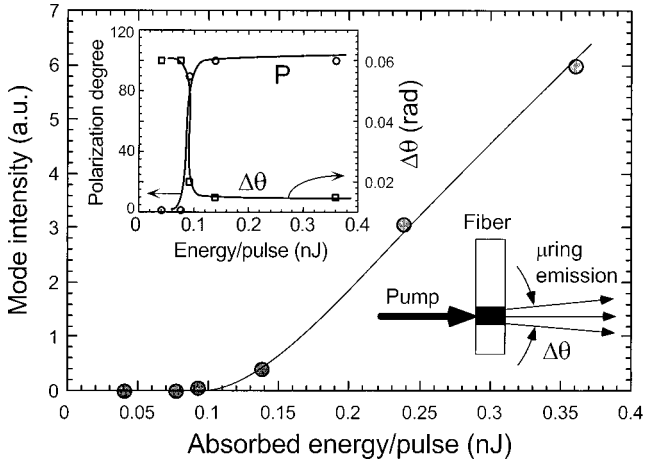

FIG. 2. Mode peak intensity dependence on the excitation intensity for a 10 μm diam ring DOO-PPV μlaser. The upper inset shows the corresponding excitation intensity dependencies of the polarization degree and the μring emission confinement angle $\Delta\theta$; the lower inset shows a schematic drawing of a photopumped ring μlaser, where $\Delta\theta$ is defined.

μcavity dimensions, we are able to control the number of resonant modes by varying D. For $\Delta\lambda > \Gamma$ only a single laser mode appears in the spectrum. Therefore, for very small fibers with D of the order of 5 μm, the μring laser contains only a single mode as shown in Fig. 1(c).[16]

The characteristic emission intensity dependence on I of the μring laser is given in Fig. 2. In addition, the Fig. 2 inset shows that the regular photoluminescence emission from the μ ring is directional,[13] concentrated in the plane of the illuminated area of the μring within an angle $\Delta\theta$ of a few hundredths of a radian. However, at I_0 $\Delta\theta$ abruptly decreases from 0.06 to 0.015 rad, showing increased directionality in the lasing regime. We note, however, that it might also be possible to couple the μring emission directly into the optical fiber using a proper built-in refraction grating. Such a μlaser would be fully compatible with the present fiber-optics technology. Moreover, the Fig. 2 inset shows that the polarization degree P (Ref. 17) of the μring emission also abruptly changes at I_0. P increases from ~ 1 for $I < I_0$ to more than 100 for $I > I_0$. We note that the μlaser emission is linearly polarized along the fiber axis regardless of the excitation beam polarization. This dramatic increase in P again shows the resonant character of the laser emission lines in the polymer μrings.

Unlike optical glass fibers, metal wires, however, are not very good substrates for a dielectric waveguide. Waveguided light on metallic surfaces experiences high absorption losses,[18] and therefore, lasing is suppressed in thin films deposited on metal wires. However, it is possible to increase d until WGM, which are confined to the outer film edge, become the dominant modes. The inner surface is no longer important in this case, and consequently, may be metallic. Indeed, when we deposited DOO-PPV films around $\varnothing 25$ μm metal fibers, we found that very thin films with $d = 1-2$ μm did not lase. However, when we increased d to values above 5 μm (Fig. 3, inset), we observed lasing WGM as shown in Fig. 3 for aluminum wires, with similar I_0 as that of a glass fiber with the same diameter. As for the glass fibers, $\Delta\lambda$ in Fig. 3 is determined by D in accordance with Eq. (1). However, from Fig. 3 we estimate n_{eff} for films

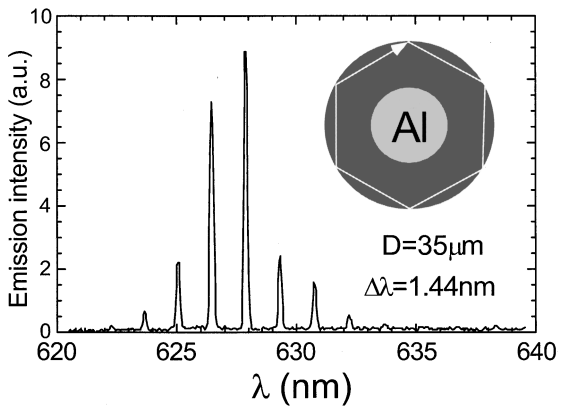

FIG. 3. Normalized emission spectra of μring lasers formed around a thin aluminum wire. The outer diameters D and intermodal spacing $\Delta\lambda$ are given; the inset shows the propagation of the whispering gallery mode inside the μring.

deposited on Al wires to be ~ 2.3. This indicates that the polymer refraction index dramatically increases when the polymer film is deposited on a metallic substrate.[19] The demonstration of ring μlasers with a metallic core is an important step in the pursuit of electrically pumped plastic lasers. It shows that future μring lasers might be electrically driven with the metal core serving as one of the electrodes.

In summary, we have achieved efficient photopumped multimode and single mode lasing in cylindrical μcavities formed by thin films of luminescent conducting polymers. The laser lines spectral width is 1 Å or less. Fabrication of such lasers is possible on thin glass and metal wires. Plastic ring μcavities are characterized by high Q and a correspondingly low lasing threshold excitation intensity, which is mainly determined by self-absorption and scattering in the polymer film. Further improvements in the polymers' quality will result in laser thresholds sufficiently low to allow electrical pumping. In this case, a ring configuration with a conducting core may be the cavity of choice for the plastic laser diode.

The authors thank M. Shkunov, Dr. W. Gellermann, and Dr. D. Chinn for assistance with the measurements. This work was done at the Utah Laser Institute and supported in part by DOE Grant No. FG-03-96-ER4549.

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