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Thermally induced whispering gallery mode laser in MEH-PPV solutions

ABSTRACT

Fernando Lahoz*

Physics Department, Universidad de La Laguna, 38206 La Laguna, Spain

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We report on thermally activated whispering gallery laser modes of a solution of MEH-PPV conjugated polymer supported by a silica optical fiber. The viscosity of the polymer solution gives place to a thin shell of the gain solution around the fiber driven by capillary action. Whispering gallery modes (WGMs) are thermally induced by a decrease in the refractive index of the polymer solution under intense optical pumping. The laser emission is produced because the evanescent waves of the WGMs couple the surrounding gain medium. These results support the use of conjugated polymers in optofluidic laser systems and highlight the importance of physicochemical properties, such as viscosity and optically induced heating, on the performance of the devices.

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1. Introduction

Optofluidics has emerged in the last years as a fast growing research field with interests both in fundamental science as well as in technological applications [1,2]. In particular, optofluidic lasers stand as the combination of a liquid gain medium that interacts with a solid structure to provide the necessary optical feedback to obtain laser emission. In this sense, whispering gallery modes (WGMs) are formed in optofluidic ring resonators by total internal reflection of the light emitted by the gain medium along the curved interface between the high refractive index (RI) material, which is frequently a glass disk, or capillary, and the low RI surrounding environment, normally formed by a dye solution [3-10]. Recently, an all-optical gain switch based on semiconducting (conjugated) polymer solutions has been demonstrated, suggesting the use of conjugated polymers in optofluidic devices [11].

Semiconducting polymers have been widely investigated since the first report of organic light emitting diodes

http://dx.doi.org/10.1016/j.orgel.2014.05.038 1566-1199/© 2014 Elsevier B.V. All rights reserved. [12]. The demonstration of laser emission under optical pump in a semiconducting polymer solution arrived few years later [13]. A practical limitation of these solutions is that conjugated polymers need organic solvents to dissolve and the solutions do not conduct electricity. Nevertheless, semiconducting polymer solutions are good candidates for optofluidic laser systems due to their high fluorescence efficiency and gain values [13].

A very simple scheme to obtain an optofluidic laser uses a microcylinder as a ring resonator surrounded by a lower RI liquid gain medium. The evanescent waves of the WGMs that resonate close to the curved interface of the microcylinder overlap with the gain solution around it and produce WGM laser emission [6]. For this process to occur, the RI of the microcylinder must be higher than that of the surrounding medium. Frequently, silica capillars or silica fibers (RI = 1.458 in the visible range) are used as the ring resonator [3–6]. However, the RI of conjugated polymers is relatively high. For instance, it is about 1.8 for (poly(2methoxy-5-(2'-ethyl-hexyloxy)-1,4 phenylene vinylene (MEH-PPV) in its solid form [14]. Consequently, relatively high RI solutions are obtained when the concentration of the conjugated polymer is increased. This avoids the use







^{*} Tel.: +34 922316502x6034; fax: +34 922318228. E-mail address: flahoz@ull.es

of silica capillars with high concentrated semiconducting polymer solutions.

MEH-PPV is one of the most frequently investigated semiconducting polymers. Laser emissions of MEH-PPV in solid thin films have been widely reported using different resonance configurations [15]. However, further research on the optical properties of conjugated polymer solutions is required for their application in optofluidics. In this paper, we report an unexpected result such as the formation of WGM laser at the interface of silica optical fiber and a relatively high RI MEH-PPV solution under transversal pumping. Moreover, due to the viscosity of the polymer solution a column of the liquid grows up around the optical fiber. The establishment of WGMs is discussed in terms of thermal effects produced by the pump laser and its influence on the RI of the solution. These results demonstrate that WGM laser emission can be switched on/off by heating/cooling external processes.

2. Experimental section

Semiconducting polymer solutions of MEH-PPV from American Dye Source at a concentration of 1 mg/ml were prepared in tetrahydrofuran (THF) solvent. RI of the solvent is 1.407. A guartz cuvette was filled with the solutions. In order to observe WGM a segment of silica 125 µm diameter optical fiber was dipped into the solution, resting vertically against the cuvette front wall. The concentration of the polymer increases the viscosity of the solution in such a way that when the optical fiber was introduced in the liquid, against the front cuvette wall, a column of the solution grew up along the fiber driven by capillary action (Fig. 1). Moreover, if the optical fiber is softly removed from the cuvette wall the surface of the optical fiber is completely clean and not a trace of thin film polymer deposition is observed. This evidences that there is no solid deposition of the polymer on the optical fiber surface due to evaporation of the solvent, at least during the whole duration of the experiments.

A tunable optical parametric oscillator pulsed laser was used as the excitation source (pulse duration about 8 ns, repetition rate 10 Hz) and tuned at the ground state absorption band of the solution at 490 nm. A cylindrical

optical

(b)

(a)

fiber

pump beam

(b)

(a)

lens focused the pump beam, with its polarization parallel to the fiber axis, on the cuvette front face to form a horizontal line (\sim 1 mm long by 300 µm wide), which homogeneously illuminated the glass fiber and the gain medium around it for transversal side-pumping. A convergence lens was used to collect the light scattered from the pumped region and focus it in a high numerical aperture optical fiber (400 µm diameter) connected to a CCD spectrometer. Two different pumped areas were studied. First, the region around the optical fiber when it is immersed in the solution (position (a) in Fig. 1). Second, the section of the polymer solution that stays around the fiber by capillarity (position (b) in Fig. 1).

3. Results and discussion

The ground state absorption (GSA) of the MEH-PPV solution is given in Fig. 2. The absorption coefficient is an important parameter for lasing under transversal pumping, because it determines the absorption length of the solution, which is about 80 µm at the maximum of the absorption band. Consequently, the pump beam is almost completely absorbed in a liquid volume close to the optical fiber section. The emission of the sample was measured when the excitation stripe line was focused on the cuvette front face around the optical fiber, in the region immersed in the liquid solution (position (a) in Fig. 1). At low pump power, the typical photoluminescence (PL) emission spectrum of MEH-PPV is recorded. When the pump power is increased above a certain pump threshold, a narrowing of the emission is observed, which is associated to amplified spontaneous emission (ASE) [16-18]. Two ASE bands are observed at about 560 and 595 nm (Fig. 2). They correspond, approximately, to the spectral positions of the radiative relaxation of the π^* excited electronic state to the ground and first vibronic states of the ground level. 0-0 and 0-1, respectively. Although the intensity of the PL 0-0 transition is higher than the 0-1 one, the ASE band at 595 nm is more intense because of lower absorption losses at 595 nm than at 560 nm. The recorded signal does not change with the number of excitation pulses as far as the pump intensity is kept constant. As the pump power is further increased, the intensity of ASE evolves with a



Fig. 1. Picture and scheme of the experimental setup.



Fig. 2. Absorption coefficient, normalized PL and ASE spectra of MEH-PPV solutions.

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