Laser oscillations of whispering gallery modes in thiophene/phenylene co-oligomer microrings

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Laser oscillations of whispering gallery modes in thiophene/phenylene co-oligomer microrings

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Laser oscillation of whispering gallery modes was observed in microring structures of semiconducting thiophene/phenylene co-oligomer (TPCO) crystals at room temperature. Microring structures were formed by dry etching from thin film crystals of TPCO. The thresholds for the laser oscillation of a microring and a thin film crystal are 200 and 1400 μJ/cm² for picosecond excitation, respectively. Therefore, the threshold for the microring was reduced to 1/7 of that for the thin film crystal. The dramatic reduction of threshold clearly demonstrates the importance of microcavity in making efficient organic semiconductor lasers. © 2007 American Institute of Physics.

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Recently, thiophene/phenylene co-oligomers (TPCOs) have attracted much attention because they show excellent optical and transport properties at room temperature (RT). Crystals of TPCO showed spectrally narrowed emissions under high density excitation at RT, and a stimulated emission including amplified spontaneous emission (ASE) was reported. Laser oscillation has been demonstrated as well. The stimulated resonance Raman scattering and pulselike emission with time delay and other optical amplification processes were observed. In addition, TPCO crystals exhibited electroluminescence and showed good performance of field-effect transistors at RT. Therefore, these materials are expected to be good candidates for organic laser materials.

Both the polymer organic materials and the aligned crystalline organic materials are good candidates for organic semiconductor lasers. Carrier mobility of crystalline organic materials is generally higher than that of polymers, and a very high mobility exceeding 10 cm²/V·s was reported in semiconducting thiophene/phenylene co-oligomer materials. So, highly crystalline organic materials are preferable to the realization of organic semiconductor lasers. Low lasing threshold is very important for the application. It is believed that confining the light into high-gain materials reduces the lasing threshold. Lasing in microcavities such as Fabry-Perot cavities, rings, disks, and spheres has been demonstrated not only in inorganic but also organic semiconductors. Microcavities provide a strong coupling of stimulated emission into lasing modes, and consequently high Q values lead to low lasing thresholds. In this letter, we reported laser oscillations of whispering gallery modes (WGMs) in TPCO microring cavities excited by a picosecond laser.

We used 2,5-bis(4-biphenylyl)thiophene (BP1T) crystals as organic high-gain media. The fundamental optical properties and the origin of the ASE observed in BP1T single crystals at low temperature were reported previously. Firstly, a buffer layer 300 nm thick of amorphous fluorocarbon polymer, Cytop CTX804.5A (ASAHI GLASS Co., Ltd., Japan), was spin coated on a SiO₂ layer 160 nm thick formed by thermal oxidation on the silicon (100) substrates. The buffer layer worked to suppress the crack formation during the crystallization of the BP1T amorphous layer. Secondarily, a BP1T amorphous layer 400 nm thick was formed on the buffer layer by the vapor deposition of BP1T microcrystals and an overlayer 300 nm thick of the CTX804.5A was made by spin coating to prevent desorption of BP1T during the course of its crystallization. Further thermal annealing by a hot plate formed BP1T thin film crystals at 220 °C. After annealing the BP1T film, long molecular axis of BP1T stood perpendicular to the SiO₂ surface, as is shown schematically in the right inset of Fig. 1, and the transition dipole moments were nearly parallel to the long molecular axes. Microring structures were made by dry etching from thin film crystals of TPCO. The thresholds for the laser oscillation of a microring and a thin film crystal are 200 and 1400 μJ/cm² for picosecond excitation, respectively. Therefore, the threshold for the microring was reduced to 1/7 of that for the thin film crystal. The dramatic reduction of threshold clearly demonstrates the importance of microcavity in making efficient organic semiconductor lasers. © 2007 American Institute of Physics.
processes. Fujiwara et al. /H20851/BP1T microrings for the efficient pumping. Light with the incident angle of 50° was used to excite each BP1T microring is aligned to a molecule length axis excited homogeneously. The transition dipole moment of larger than the areas of microrings. Thus, each microring was excited from 2.64 to 2.70 eV correspond to the 0–1 vibronic band of BP1T.20 The value of the microring was estimated to be 2200 at 2.67 eV from the linewidth of the resonator mode. The maximum

Equally spaced peaks were observed from each microring. The mode interval becomes smaller with increasing microring diameters because of the oscillations due to higher order modes. These mode intervals are calculated by the fast Fourier transformation, indicating the presence of a main peak given by the inverse of the period. The relationship between the mode interval and the resonator length is plotted in Fig. 3. Here the circumferential length of the microring was assumed to be the resonator length L. The solid line in Fig. 3 is the least-squares fitting. Luminescence peaks were separated from each other by $E_m = n hc / n L$, given by

$$\Delta E_m = E_{m+1} - E_m = \frac{hc}{n_{\text{eff}} L}, \quad n_{\text{eff}} = \left( n + E \frac{dn}{dE} \right)_{E_m},$$

where $h$ is the Planck constant, $c$ is the velocity of light in vacuum, $n_{\text{eff}}$ is the effective refraction index, and $n$ is the refraction index of the gain materials.22 Thus the inverse proportion between the mode interval $\Delta E$ and the resonator length $L$ represented in Fig. 3 is a direct consequence of the presence of cavity multimodes confined as standing waves in the resonator. Consequently, we conclude that the microring lasers reflect WGMs.

The calculated WGMs were obtained by the analysis based on the finite difference time domain method. The number of the electromagnetic field in the radial direction $n_r$ is assumed to be 1 or 2 for the microring diameter of 20 μm. Then, the node number of circumferential direction $n_	heta$ is $(n_r, n_	heta) = (1, 243–248), (2, 234–239)$ in the 0–1 amplification line. Both TE and TM modes are allowed in the WGM so
that the total number of the WGM’s should be $6 \times 4 = 24$ in the 0–1 amplification region. However, peak energies of (1, $\lambda$) on the TM mode give close agreement with that of (2, $\lambda$–8) on the TE mode. We thought that these two modes could not be resolved in the experiment. On this assumption, the total number of WGM’s should be $6 \times 3 = 18$, which gives the good agreement with the observed 17 WGM’s, as shown in Fig. 2.

Figure 4 shows the emission intensity of a BP1T thin film crystal and a microring formed on the same BP1T thin film crystal as a function of the excitation density. The typical thresholds for the laser oscillation of a BP1T microring and a BP1T thin film crystal are 200 and 1400 $\mu$J/cm$^2$, respectively. The laser oscillation threshold for the microring was reduced to 1/7 of that for the thin film crystal. Microring structure was demonstrated to be useful for the organic semiconductor laser and other optical and electronic devices.