Hybrid planar microresonators with organic and InGaAs active media

J. R. Mialichi,1,* A. Camposeo,2 L. Persano,2 L. A. M. Barea,1 P. Del Carro,2 D. Pisignano,2,3 and N. C. Frateschi1,4

1 Device Research Laboratory, Applied Physics Department, “Gleb Wataghin” Physics Institute, University of Campinas-UNICAMP, 13083-970 Campinas, São Paulo, Brazil
2 National Nanotechnology Laboratory, CNR-Istituto di Nanoscienze, Università del Salento, via Arnesano, I-73100 Lecce, Italy
3 Istituto Superiore di Formazione Interdisciplinare ISUFI, Università del Salento, via Arnesano, I-73100 Lecce, Italy
4 Center for Semiconductor Compounds, University of Campinas-UNICAMP, 13083-870 Campinas, São Paulo, Brazil

* mialichi@ifi.unicamp.br

Abstract: The authors report on the fabrication of hybrid planar microresonators based on InGaAs microdisks with an evaporated organic material. Samples of InGaAs grown on InP(100) substrates are obtained by Chemical Beam Epitaxy, and microdisks of InGaAs with different diameters are fabricated by focused ion beam. The hybrid disks are obtained by the subsequent evaporation of 8-hydroxyquinoline aluminium doped with 4-Dicyanomethylene-2-methyl-6-(p-dimethylaminostyryl)-4H-pyran on the InGaAs microdisks. The devices, characterized by micro- and confocal photoluminescence imaging and spectroscopy, exhibit emission around 650 nm, from the organic material for disks with different radius. Finally, simultaneous emission in the visible and at whispering gallery resonant modes in the 1350-1450 nm range are observed due to excitation transfer to InGaAs. These devices open the possibility to combine the flexibility of organics with the high gain of III-V compounds for wavelength down conversion and telecom applications.

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1. Introduction
Microresonators of different geometries and materials are studied due to their potential applications in optical-communications systems [1–3]. In particular, microdisks are shown to produce a high quality factor $Q$ [4] which may consequently lead to low laser thresholds. Active-microdisk resonators offer a great advantage in obtaining stimulated emission in small volume [5] since they support whispering-gallery modes (WGMs), which are very confined resonances with maximum intensity near the disk edge [6]. High photonic lifetime can be achieved without complex systems for optical feedback. Moreover, the peculiar structure, with planar emission without the need of cleaved mirrors, enables easy integration with other optoelectronic devices. Two factors are fundamental for the performance of active microdisk...
resonators, namely the low roughness of the disk walls to reduce optical scattering losses [7] and the low surface recombination velocity to increase internal quantum efficiency. For these reasons, microdisks are very useful for photonic applications. On the other hand, since the discovery of stimulated emission, amplified spontaneous emission, and lasing in organic conjugated systems [8], many efforts have been directed towards the realization of single or multi-wavelength, solid-state organic-based laser devices for applications in diagnostics, displays, and data storage [9]. Compared with inorganic materials, organic low-molar mass molecules and polymers exhibit some important advantages in terms of fabrication process, often performed at ambient working temperatures, broad emission spectral range and possibility of color tunability, flexibility in terms of molecular design and synthesis, and low cost [10,11]. In the last decade, the field of conjugated molecules has soared with many applications [12–14] including optically pumped lasers [15,16]. Conjugated polymers are also employed as active layers of vertical cavity surface-emitting lasers [17] and of microdisk lasers with 20-30 µm diameter, operating at 620-630 nm when pumped by UV [18]. Furthermore, organic laser sources are integrated on chips to couple light from an active material slab which is combined with a distributed feedback grating into a polymer strip waveguide [19]. As well known, a limitation of this technology to be useful in photonics applied to telecommunication is the emission wavelength, which is mainly in the visible range for organic systems. Emission in the near infrared is reported by combining conjugate polymers and inorganic nanocrystals [20]. Hybrid structures based on organic materials deposited on InGaAs disks can be valuable candidates to extend the applicability of light-emitting molecules to near-infrared applications, since the highly efficient InGaAs material can convert the organic emission in the visible range into emission in the c-band, near 1550 nm.

Here we report on hybrid micro-resonators with simultaneous visible and infra-red emission. We fabricate microdisks with different diameters from InGaAs material grown on InP(100) substrate by Chemical Beam Epitaxy (CBE). The InGaAs layer is compressively strained by ~0.4% with respect to InP. The organic system, 8-hydroxyquinoline aluminium (Alq3) doped with 4-Dicyanomethylene-2-methyl-6-(p-dimethylaminostyryl)-4H-pyran (DCM) is deposited on the microdisks by thermal evaporation. Two cases are analyzed. In the first case (i), a bulk layer of SiO2 is deposited on the InGaAs microdisk before depositing the Alq3:DCM in order to study the effects of the cavity size on the organic emission. A blue-shift in the emission is observed as the disk size is reduced. In the second case (ii), the Alq3:DCM is deposited directly on the InGaAs microdisks to investigate the light coupling between the organic layer and the InGaAs substrate. A simultaneous emission at 660 nm and 1350-1450 nm is observed, due to the excitation transfer to the InGaAs material.

2. Fabrication of microdisks

While the most of reported microdisk structures based on organic light-emitting materials are realized by means of standard photolithography [18,21–23], we first fabricate inorganic pedestals to deposit Alq3:DCM afterwards. Samples of InGaAs are grown by CBE on InP(100) substrates using trimethylindium, triethylgallium, AsH3 and PH3 precursors. Initially, 200-nm-thick InP buffer is grown on an oriented InP(100) substrate at 520°C, followed by 1000-nm-thick InGaAs grown at 530°C. After CBE growth, disks of different diameters (38, 26 and 18 µm) are obtained. The processing steps consist essentially of an InGaAs selective wet-etching (H2SO4:H2O2:H2O) resulting in cylinders with vertical walls, followed by InP selective wet-etching (HCl: H3PO4) for the pedestal formation. Pedestal heights of 4-8 µm are achieved after 30-60s of InP selective wet-etching. Disks with diameters of 2, 4 and 8 µm are obtained by focused ion beam (FIB) at the edge of the samples.
Figures 1a and 1b show the lateral and top view of a microdisk with 18 and 26 µm of diameter, respectively. Notice that the walls of the disk are not vertical due to the InGaAs selective wet-etching that tends to stop at the (111)A planes. This certainly influences the InGaAs resonator, but does not significantly affect the organic material deposition. Improvement for the InGaAs profile can be achieved with dry etching before InP selective etching. Figures 1c and 1d show microposts with diameters of 4 µm, realized at the edge of the sample by a dual-beam FIB equipment with scanning electron microscopy capability. FIB milling is performed using a Ga\(^+\) beam current of 3 nA accelerated at 30 kV during 12 minutes. Due to the reduced size of the posts, InP selective wet-etching is not carried out on these samples.

Two cases are investigated. In the first case, a bulk layer of SiO\(_2\) (800 nm) is deposited on the InGaAs microdisk before depositing the organic thin film. The SiO\(_2\) layer is deposited by a Temescal Supersource 2 electron-beam gun system, in an oxygen atmosphere (≈2.4 × 10\(^{-4}\) mbar) using 99.9% purity SiO\(_2\) disks (Leybold, Germany) as source materials. The chamber is at room temperature (about 20 °C) at the beginning of the deposition, reaching less than 60 °C at the end of the process. The organic thin film is then deposited on SiO\(_2\) layer by thermal evaporation using a PVD75 system from Kurt J. Lesker. A 300 nm thick layer of Alq\(_3\) doped with DCM (2% in ww) is evaporated at pressure of 4.5 × 10\(^{-7}\) mbar and a constant deposition rate of 1.5 Å/s, monitored by a quartz gold-coated crystal microbalance. In the second case, a 300 nm thick film of Alq\(_3\):DCM is directly deposited onto the InGaAs microdisk under the same evaporation conditions.

3. Results and discussion

PL micrographs of the samples are obtained by using a confocal microscope (FV1000, Olympus), exciting the samples with the 488 nm line of an Ar ion laser. PL spectroscopy measurements of the samples with the SiO\(_2\) layer are performed using the third harmonic of a Q-switched neodymium-doped yttrium aluminum garnet pulsed laser (Alphalas GmbH, \(\lambda = 355\)nm) with a spotlight of about 20 µm, pulse duration of 0.6 ns and repetition rate of 100 Hz. The pulse energy is kept constant at 0.2 µJ. The disk emission is collected by an optical-fiber coupled to a monochromator (iHR320, Jobin Yvon), equipped with a Charge Coupled Device for detection (1024 × 256 pixel, Simphony, Jobin Yvon). The measurements are carried out at room temperature with a spectral resolution of 0.2 nm. As the SiO\(_2\) index refraction (1.46 at 630 nm) is lower than that of Alq\(_3\):DCM [16], there is a reduced coupling of the light emitted by the organic layer to the InGaAs disks.
Figure 2 shows confocal PL images of disks with 38, 26 and 8 µm of diameter (2R) with 300 nm-thick film of Alq₃:DCM over their surfaces. The organic film is quite homogeneous and uniform. The underneath pedestals can be visualized due to the index refraction differences of the materials. Figure 3 shows instead the normalized PL spectral emission curves of Alq₃:DCM deposited on SiO₂ layers on the top of InGaAs disks. PL measurements on the disks show peaks at 664 nm, 651 nm, and 648 nm for diameters of 38 µm, 26 µm and 8 µm, respectively. A blue-shift of the emission from the organic material is observed upon decreasing the disk diameter.

We observe that the blueshift increases as the laser spot approaches the disk diameter value of 26 µm. As the disk becomes smaller than the spot area very small change in the blueshift is observed. Therefore, pumping density must play an important rule in the blueshift. The absorbed pump energy density increases as the disk diameter decreases with a maximum when the laser spot matches the disk diameter. Higher absorbed pump energy density leads to higher population of the levels which in turn causes a larger blue-shift for the transitions. This effect is similar to band filling in semiconductors. As the disk diameter is further reduced, there is essentially no change in pump density and, therefore, negligible change in the blueshift.

Alq₃ doped with DCM provides an excellent light-emitting and gain media [24], in which the maximum absorbance of the DCM is centered at 530 nm [25]. The organic layer thickness was designed to absorb almost all pumping light. We measure an absorption coefficient of 2 × 10⁵ cm⁻¹ for a 300 nm-thick Alq₃:DCM film at the excitation wavelength of 533 nm (the organic film transmits 0.2% of the incident light). In this way, 300-nm-thick films of Alq₃:DCM may avoid a significant direct excitation of the InGaAs layer. Thicker layers will result in non efficient pumping of the organic layer. Thinner layer obviously would allow
direct pumping of the InGaAs disk. PL measurements of samples without SiO$_2$ are performed by exciting sample by a diode laser at 533 nm and these measurements, shown in the Fig. 4, are collected at room temperature, by means of two different spectrometers, i.e. a Ocean Optics, model (HR2000) with resolution of 1 nm in the visible region, together with an Optical-Spectrum Analyzer, model HP 70950B with resolution of 0.8 nm in the infrared. The laser spotlight used in this set-up is approximately 25 µm. Disks with diameter smaller than 25 µm could allow lateral pumping of the InGaAs material and provides no increase in pumping density. Disks of 26 and 38 µm diameters were measured. PL spectra, shown in Fig. 4b of the 38-µm-diameter disk shows a broad emission centered at 1430 nm with no sign of resonant modes. This is expected since there is a highly absorbing unpumped 6 µm wide ring near the edge of the disk where the WGM modes should be a maximum. Therefore, in the following, we restricted our study to the 26 µm disk.

Figure 4a shows the normalized PL emission curve of the organics deposited on a 26-µm-diameter InGaAs disk, with maximum emission at 660 nm. We use a laser beam power of 1.2 mW. We use a multimode fiber to collect the light emitted from the organic material in the visible region. Figure 4b shows the measurements carried out in the infrared region (1300-1500 nm) using a single-mode fiber to collect the InGaAs disks emission. Since the index refraction of InGaAs (3.91) is quite larger than that of Alq$_3$:DCM (1.77) at 630 nm, we expect a significant coupling between the organic layer and the underneath InGaAs substrate. Such a coupling is the most probable reason of providing a spectral emission in the infrared region. The interaction between inorganics and organics within the fabricated heterostructure can be mediated by radiative or non-radiative energy transfer, or by both the mechanisms [26,27]. In particular, non-radiative energy transfer is utilized as strategy for the non-optical excitation in hybrid inorganic-organic composite systems, whose phenomenon has been predicted and observed in inorganic-organic heterostructures, constituted by inorganic quantum wells interacting with amorphous conjugated polymer films [27–29], and in nanocomposites [30–33]. Furthermore, one see clearly in the inset figure some emission peaks at 1361, 1369, 1379, 1399, 1408, 1417, 1424 and 1434 nm. Using Fast Fourier Transform (FFT) to analyze the spectrum of the inset figure, we find a 10.08 nm periodicity. Considering the effective index of refraction $n_{\text{eff}} = 2.5$ for InGaAs structure, the expected free spectral range for a 13 µm radius disk, is $\Delta \lambda \approx \lambda^2 / 2 \pi R n_{\text{eff}} \sim 10$ nm. Therefore, we believe these peaks are indeed whispering gallery modes (WGMs) of the InGaAs disk.

Based on the absorption coefficient of the polymer we estimate that 1.7 µW of the incident light excites the InGaAs disk for the 1.2 mW pumping power. The measured integrated power
irradiated by the InGaAs disk of 26 µm was $6.7 \times 10^{-2}$ µW. Our single mode fiber collects radial emission from approximately 2 µm length of the disk perimeter. Therefore, we can estimate approximately 2.8 µW of radial emission. Therefore, even assuming only radial emission and 100% efficiency of the entire system, it is impossible to neglect radiative and/or non-radiative pumping from the organic layer. In terms of the ratio between non-radiative and radiative pumping, we believe that the last is more important. If non-radiative pumping was more important we should observe no considerable difference between the InGaAs emission from the disks with 26 µm and 38 µm diameters since carrier diffusion in the InGaAs layer would lead to more uniform pumping at the edge.

4. Conclusion

In conclusion, we obtain samples of InGaAs layers grown on InP(100) substrate by CBE. InGaAs microdisks with different diameters are fabricated by photolithography and FIB. Hybrid disks are obtained by the deposition of Alq3:DCM by evaporation on the InGaAs disks. Several whispering gallery resonant modes are observed in the 26-µm-diameter InGaAs disk spectrum emission curve, with average spacing of about 10 nm obtained by FFT analysis. The developed microdisk devices are fully compatible with state-of-the-art technologies for the production of organic light-emitting diodes and potentially provide effective lateral confinement for resonant lasing modes, being therefore promising for applications in highly-integrated, hybrid organic-inorganic optoelectronics.

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