

Intermediate high index layer for laser mode tuning in organic semiconductor lasers

M. Stroisch¹, T. Woggon^{1*}, C. Teiwes-Morin¹, S. Klinkhammer¹, K. Forberich^{2,4}, A. Gombert^{2,5}, M. Gerken³ and U. Lemmer¹

¹Light Technology Institute (LTI) and Center for Functional Nanostructures (CFN), Universität Karlsruhe (TH), Kaiserstr. 12, 76131 Karlsruhe, Germany

²Fraunhofer Institut für Solare Energiesysteme (ISE), Heidenhofstraße 2, 79110 Freiburg, Germany²

³Lehrstuhl für Integrierte Systeme und Photonik, Technische Fakultät der Christian-Albrechts-Universität zu Kiel, Kaiserstr. 2, 24143 Kiel, Germany

⁴Present address: Konarka Austria Forschungs und Entwicklungs GmbH, Altenbergerstrasse 69, A-4040 Linz, Austria

⁵Present address: Concentrix Solar GmbH, Bötzingger Str. 31, 79111 Freiburg, Germany
*thomas.woggon@lti.uni-karlsruhe.de

Abstract: We modified the optical properties of organic semiconductor distributed feedback lasers by introducing a high refractive index layer consisting of tantalum pentoxide between the substrate and the active material layer. A thin film of tris-(8-hydroxyquinoline) aluminium doped with the laser dye 4-dicyanomethylene-2-methyl-6-(p-dimethylamino-styryl)-4H-pyran was used as the active layer. By varying the intermediate layer thickness we could change the effective refractive index of the guided laser mode and thus the laser wavelength. With this technique we were able to tune the laser emission range between 613 nm and 667 nm. For high index layer thicknesses higher than 40 nm the laser operated on the TE₁-mode rather than the fundamental TE₀-mode.

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1. Introduction

Organic semiconductor lasers are a promising approach for compact tunable laser sources for the visible spectral range. Due to their large spectral gain, the possibility of large area deposition and the applicability of diode lasers or LEDs as pump sources [1–3] they offer many advantages compared to their inorganic counterparts. A particular interesting aspect of organic semiconductor lasers is the possible integration into micro-optical systems, such as lab-on-a-chip systems [4].

This work concentrates on distributed feedback (DFB) resonators as they have been identified to be capable of low threshold single mode lasing operation. An organic DFB laser is normally based on a slab waveguide structure formed by an optical transparent substrate with an organic layer on top. The distributed optical feedback can be induced by a modulation of the real or the imaginary part of the refractive index. The refractive index can be corrugated by periodically patterning the substrate surface or the organic layer.

Using photolithographic patterning techniques for organic materials turns out to be an intricate task due to the high solubility of organic semiconductor compounds. For this reason approaches for the fabrication of organic DFB lasers are based on pre-structured substrates. For organic DFB lasers based on a structured substrate primarily e-beam lithography, laser interference lithography and nanoimprint could be demonstrated with convincing results [5–7]. Other techniques like laser interference ablation [8], UV-linked polymers [9] or nanoimprint [10] have been successfully used for corrugating the active layer.

The benefit of organic laser materials over their inorganic counterparts is the typically large spectral gain and thus a large tuning range. The emission wavelength of a distributed feedback laser can be estimated by the Bragg formula: $\lambda_{\text{Bragg}} = 2\Lambda n_{\text{eff}} / m$

Here Λ is the grating period of the corrugation or the pumping pattern, n_{eff} is the effective refractive index of the guided mode and m represents the order of Bragg reflection used for optical feedback. According to this relation the emission wavelength for a specific laser material can be tuned by adjusting Λ or n_{eff} . Different grating periods [11] are an established way for achieving such a wavelength tuning although this approach relies on elaborated electron beam lithography for defining the grating structures with sufficient precision.

Parallel methods like laser interference lithography are far more suitable. However, resonators with different periodicities on the same substrate cannot be realized as convenient as it is possible with serial techniques. Using laser interference lithography for this task for example would require multiple exposures with different periods and shadow masks. Due to diffraction at the mask edges which is difficult to avoid, this is an elaborate task.

One method for dealing with this problem is to vary the thickness of the laser film thus changing the effective refractive index of the guided mode. This was done for evaporated organic thin films on different substrates [7]. This technique, however, cannot easily be extended to solution processable organic semiconductors which are spin coated onto the substrate.

In contrast to these approaches we focus on the concept of influencing n_{eff} by an extra intermediate layer between the substrate and the active material. The material of the intermediate layer needs to be optical transparent and it should have a large refractive index compared to the adjoining layer materials. Typically, the refractive index of the substrate is 1.4–1.5 and the active material is in the range of 1.6–1.8. Therefore high index materials like titanium dioxide (TiO₂) and tantalum pentoxide (Ta₂O₅) are a good choice. Depending on

the evaporation process they have a refractive index of 2.0 - 2.3. In the case of rutile, a special crystalline form of TiO_2 even up to 2.7 is possible.

Harbers et. al. investigated resonators based on columns consisting of TiO_2 and could demonstrate a higher coupling constant and lower laser thresholds [12]. Here we show the tuning of laser emission by varying the thickness of an intermediate Ta_2O_5 layer. This material has been used for various applications where dielectric waveguides are used. Losses on the order of 1 dB/mm have been reported [13]. This value is lower than the losses due the outcoupling of the laser radiation in second order distributed feedback structures used in our experiments.

2. Experimental

As a substrate we use sinusoidal gratings of ~ 95 nm depth and 400 nm in period. The gratings were formed by UV embossing into an acrylic resist [14]. The period is matched to a second order distributed feedback laser. Thus, the optical feedback is due to second order Bragg scattering and laser light is diffracted perpendicular to the device by first order scattering.

The intermediate layer of Ta_2O_5 was produced by e-beam evaporation with a rate of 6 \AA/s . Onto this layer tris-(8-hydroxyquinoline) aluminium (Alq_3) doped by 4 mol% 4-dicyanomethylene-2-methyl-6-(p-dimethylaminostyryl)-4H-pyran (DCM) was co-evaporated. The thickness of this layer was 350 nm. The scheme of the structure is depicted in Fig. 1(a).

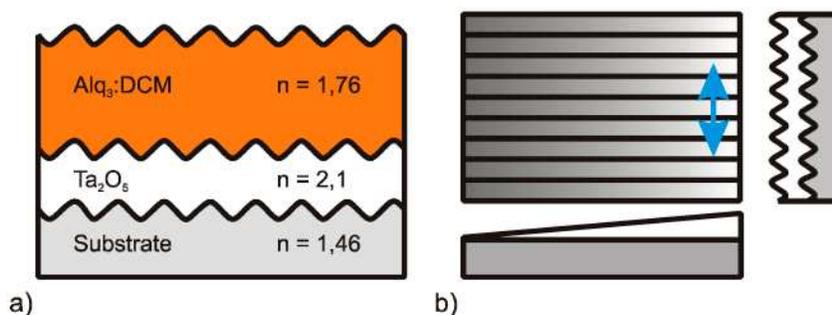


Fig. 1. (a) Scheme of the layer structure used in the experiments. (b) Scheme of the sample with a gradient layer of Ta_2O_5 . The arrow shows the direction of feedback.

We produced seven samples with a size of 2.5 cm x 2.5 cm and homogeneous Ta_2O_5 -layers up to 163 nm. To demonstrate variable wavelength tuning a sample with an area of 2.5 cm x 5 cm was evaporated with a wedge shaped layer of Ta_2O_5 . This was realized by moving a shutter in front of the sample during the evaporation process forming layer thickness between 0 nm and 80 nm. In order to minimize the influence of the wedge on the laser performance the grating was oriented with its lines parallel to it. Therefore a homogeneous stripe of high index material is in the direction of the optical feedback [see Fig. 1(b)].

For the optical characterization the devices are pumped with a frequency-tripled neodym yttrium-vanadat (Nd:YVO_4) laser which emits 500 ps long pulses at a wavelength of 355 nm. The used optical setup for characterization is described in more detail elsewhere [5].

3. Results and discussion

The thickness of the homogeneous Ta_2O_5 -layer was varied from 28 nm to 163 nm. For each sample we measured the laser threshold and emission wavelength. Additionally we characterized a substrate without a high index layer for comparison. This device emits at 659 nm. The relation between the Ta_2O_5 -layer thickness and the emission wavelength can be seen in Fig. 2(a). The wavelength increases with rising layer thickness. The peak wavelength switches down to 612 nm at a layer thickness of 44 nm, where two laser lines at 667 nm and 612 nm can be observed. For higher Ta_2O_5 layer thicknesses the emission wavelength increases again.

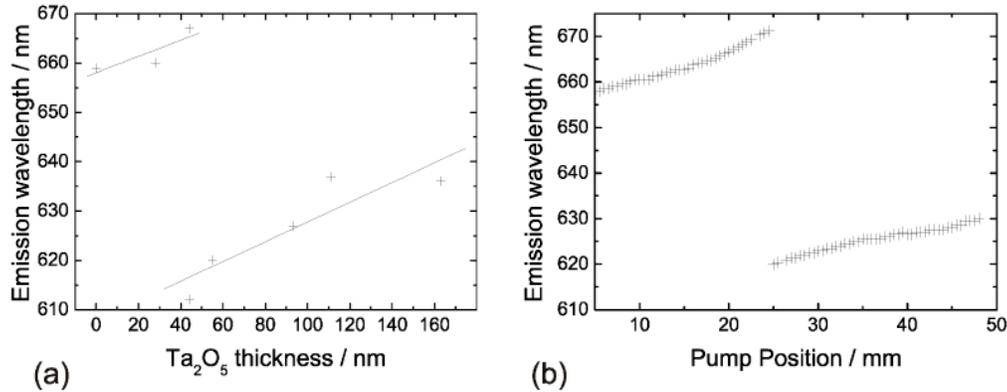


Fig. 2. (a) Relation between thickness of the Ta₂O₅ layer and the laser wavelength for substrates with a homogenous layer of the high index material. (b) Laser wavelength dependency of the pump position on a substrate with a Ta₂O₅ wedge.

Figure 2(b) shows the relation between pump position and laser wavelength for the substrate with the tantalum pentoxide wedge. The pumping spot was moved by 500 μm between each measurement. The laser wavelength displays a very similar behavior as in the case of the substrates with different intermediate layer thicknesses.

In both cases the change of the laser wavelength depends on an increase of the effective refractive index of the guided mode. Figure 3(a) shows the intensity profile of the TE₀-mode for a Ta₂O₅-layer thickness of 44 nm. A thicker Ta₂O₅-layer results in a higher effective refractive index for the TE₀-mode. At this Ta₂O₅-layer thickness, however, the waveguide structure allows a second TE-mode. Figure 3(b) shows the corresponding intensity profile. The TE₁-mode has a lower n_{eff} than the TE₀-mode. The simulation gives an index of 1.49 for the TE₁-mode instead of 1.7 for the fundamental mode. The corresponding optical confinement factors $\Gamma(\text{Alq}_3)$ are 0.76 and 0.51. The switching to another wavelength, however, is determined by the strong decrease of the material gain for the long wavelength side.

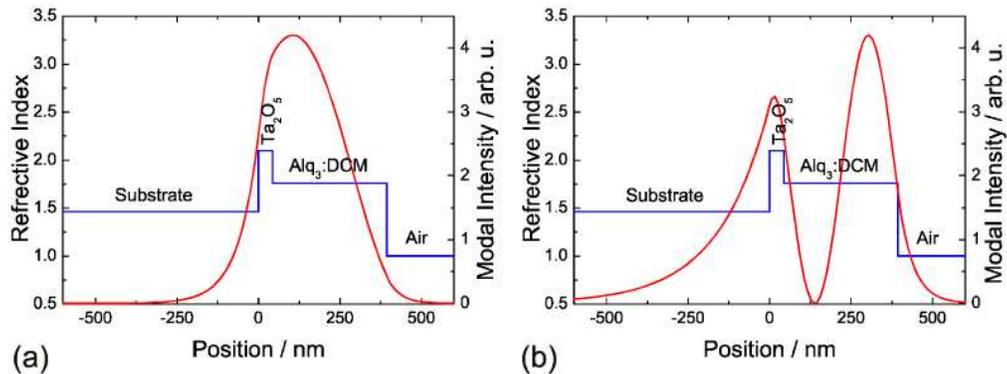


Fig. 3. Simulated intensity profile of (a) the TE₀ and (b) the TE₁ mode of for an 44 nm thick intermediate high index layer.

The filled circles in Fig. 4(a) show the spectral gain of Alq₃:DCM (as taken from Ref. 7) and the calculated modal gain $\Gamma \cdot g(\lambda)$ for the TE₀-mode and the TE₁-mode for the different emission wavelengths. Starting with a laser wavelength of 659 nm the modal gain gets lower for higher Ta₂O₅-layer thicknesses and thus resulting in higher wavelengths. The laser mode then changes to the TE₁-mode with an emission wavelength at shorter wavelengths due the higher modal gain. The calculated modal gain is also qualitatively in agreement with the

experimentally observed laser threshold depicted in Fig. 4(b). A broad minimum with threshold values around 12 mJ/cm^2 could be observed for the central wavelengths where the modal spectral gain has a maximum. The threshold values observed for layer thicknesses higher than 44 nm are comparable to the threshold observed in the absence of Ta_2O_5 indicating the high quality of our dielectric films.

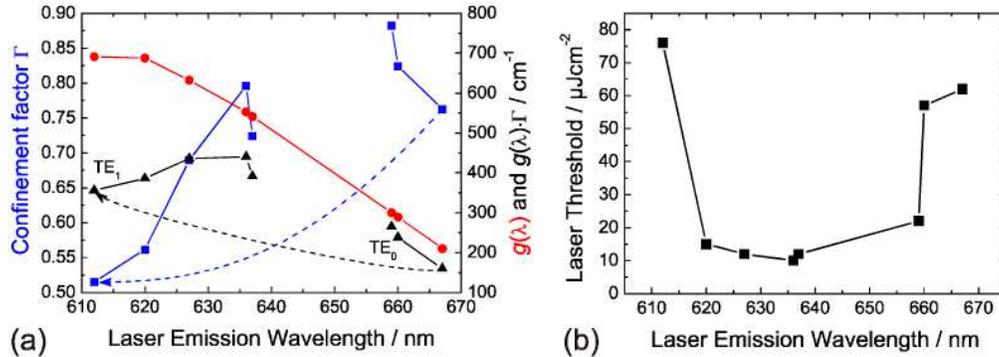


Fig. 4. (a) Relation between the laser emission wavelength and the confinement factor (squares), spectral gain $g(\lambda)$ (circles), modal gain $\Gamma \cdot g(\lambda)$ (triangles) for different laser modes. The dashed arrow and the sequence of the data points correspond to an increasing high index layer thickness. Despite the lower confinement factor the laser mode switches from TE_0 to a TE_1 -mode due to the larger optical gain at lower wavelengths. (b) Laser wavelength dependence of the laser threshold.

To demonstrate the versatility of this tuning method also for the case of solution processable materials we fabricated a continuously tunable DFB laser device with a 190 nm thick spin coated layer of the conjugated polymer F8BT (poly(9,9'-dioctylfluorene-co-benzothiadiazole)). The active material was spin coated on top of a Ta_2O_5 wedge with the layer thickness ranging from 0 to 40 nm on a resonator with a grating period of 350 nm . Again spectral tuning by moving the spatial position of the optical pump beam on the substrate is possible. Figure 5 shows the resulting excitation position dependent laser emission wavelength. We achieved a tuning range of 10 nm .

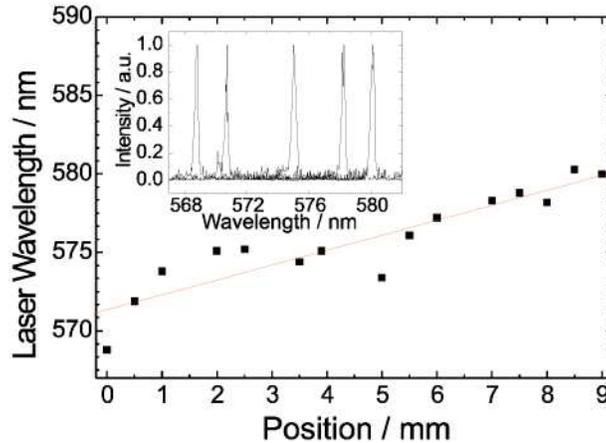


Fig. 5. Relation between the spatial pump position and the laser emission wavelength on a substrate with a Ta_2O_5 wedge and a spin coated F8BT conjugated polymer active layer. The Ta_2O_5 layer thickness was varied over a gradient ranging from 0 to 40 nm .

4. Conclusion

We presented organic semiconductor lasers mode tuning by an intermediate high index layer. Tuning of the emission wavelength between 612 nm and 670 nm could be demonstrated in Alq₃:DCM. The observed change of the laser operation from a TE₀ to a TE₁-mode at a layer thickness of 44 nm could be explained with a higher modal gain according to our calculations. Additionally we showed that our concept can be used for a continuously tunable solution processed organic DFB lasers without the need of a chirped grating.

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