Photonic stopband tuning of organic semiconductor distributed feedback lasers by oblique angle deposition of an intermediate high index layer

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We modified the photonic band structure of organic distributed feedback lasers by introducing a patterned high index intermediate layer of tantalum pentoxide. This layer was oblique angle evaporated onto one dimensional surface gratings with a periodicity of 400 nm. The dielectric broadened the stopband due to its high refractive index compared to both the substrate and the active layer. By tuning the layer thickness we could increase the stopband from 3 to 16 nm. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3184591]

Organic semiconductor lasers are an interesting field of research because of the potentially large area and low-cost patterning and deposition techniques available.[1–3] In addition, organic solid state laser simplify the integration of coherent light sources for lab-on-a-chip systems and integrated photonic circuits.[4–6]

One and two dimensional photonic crystal resonators are particularly promising, because they allow low threshold distributed feedback (DFB) lasing.[7] They consist of a substrate and a thin active layer, thus forming a planar waveguide. The optical feedback is introduced by a periodic corrugation of the substrate or the active layer, which results in Bragg reflection for specific wavelengths.

Due to the small difference of the refractive indices of the substrate ($n=1.4-1.6$) and the organic material ($n=1.6-1.8$) the index contrast is typically very small. A higher contrast could favor a stronger coupling of the laser mode allowing for smaller resonator sizes in return.[8,9] Also, a higher contrast results in a larger width of the photonic stopband.[10] A width of the photonic stopband of only a few nanometers is observed in purely organic photonic crystal lasers.[11,12] A broadening of the stopband is of particular interest. For example, a wide stopband is advantageous for ensuring single mode laser operation because the wavelength at the two band edges will differ strongly in their spectral gains. Another interesting application for large band gap organic semiconductor DFB lasers could be the realization of low threshold defect mode lasers.[13]

Here we report the enhancement in the stopband from 3 to 16 nm in an organic-inorganic hybrid system. We achieved this by evaporating an additional noncontinuous layer of tantalum pentoxide onto organic laser resonator substrates mounted at an oblique angle with respect to the evaporation source.

We used linear surface gratings patterned into an acryl resist by UV-nanoimprint as resonators. The grating period is 400 nm and the depth of the sinusoidal profile is about 95 nm. Tantalum pentoxide was deposited onto the gratings using e-beam evaporation. The substrates were mounted in the holder at an angle of 75° between the evaporation direction and substrate normal. The sample was positioned with the grating lines direction being perpendicular to the evaporation direction in order to get homogenous Ta_2O_5 lines parallel to the grating. An even mounted glass substrate was evaporated simultaneous for determining the average layer thickness. By taking into account the geometry factor of cos(75°) nominal thicknesses between 12 and 28 nm are estimated.

Onto these samples a 350 nm layer of the host system tris-(8-hydroxyquinoline) aluminum (Alq_3) doped with the laser dye 4-dicyanomethylene-2-methyl-6-(*p*-dimethylaminostyryl)-4*H*-pyran (DCM) was coevaporated. The resulting asymmetric slab waveguide structure with a Bragg surface grating is shown in Fig. 1. The grating period was chosen to provide an optical feedback by second order Bragg reflection. This results in a perpendicular outcoupling of the lasing mode by first order Bragg diffraction.

For the optical investigation of the devices, we used a frequency tripled actively *Q*-switched diode pumped neodymium: yttrium orthovanadate ($Nd:YVO_4$) laser (AOT-YVO-20QSP) as pump source emitting at a wavelength of 355 nm (500 ps pulses at 1250 Hz). The pump laser beam was focused onto the sample in a slightly elliptical spot of about 10^{-3} cm^2. To protect the active organic material from degradation due to photo oxidation the samples were kept in a vacuum chamber at a pressure below 5×10^{-3} Pa. In Fig. 2 a scheme of the setup is shown. The angular dependent emission was detected with an optical fiber with a core diameter of 1300 μm. The fiber can be moved around the sample using two rotation stages while the entrance aperture is always directed toward the photoexcited area of the active layer. The angular resolution of this setup is 0.1° which was

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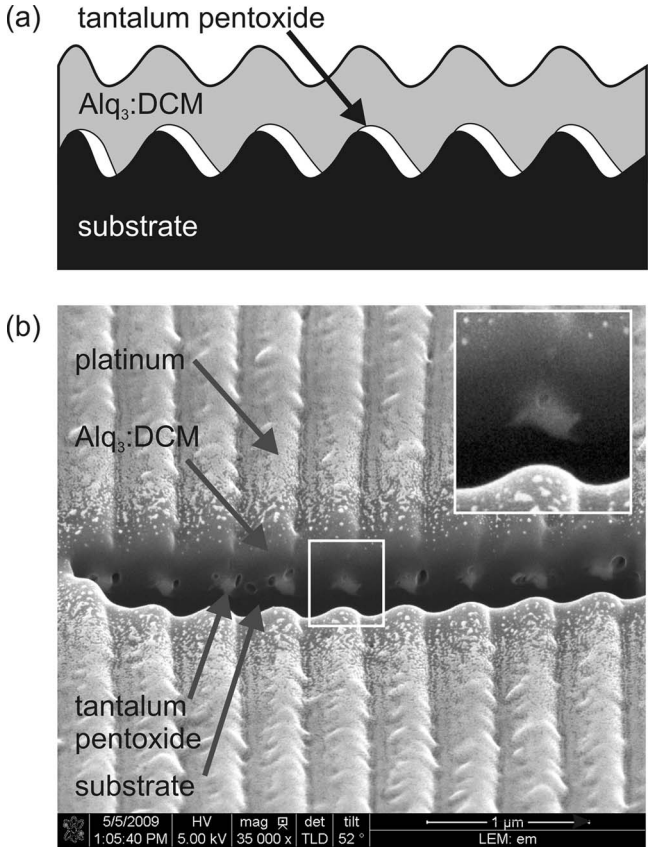

FIG. 1. (a) Scheme of the layer sequence for the devices. (b) Side-face SEM micrograph of a cut made into a sample with a focused ion beam. The structured Ta_2O_5 layer (d_{nom}=90 nm) can be identified as bright spots between the Alq_3:DCM layer and the patterned substrate.

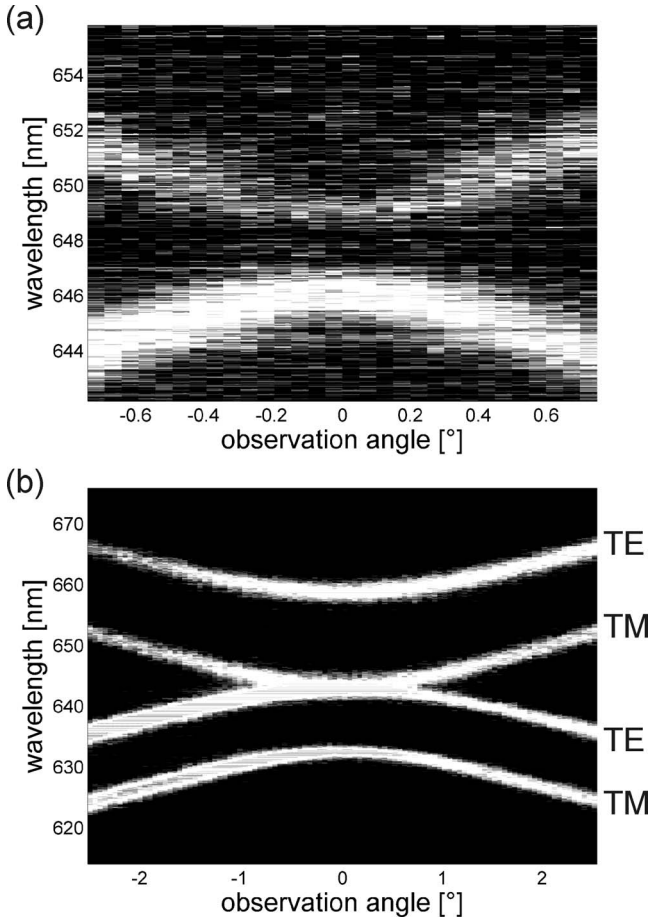

FIG. 3. Measurement data of the angular resolved emission spectra analysis for Ta_2O_5 layers of (a) d_{nom}=0 nm and (b) d_{nom}=28 nm. The TM-bands can be distinguished from the TE-bands.

determined by measuring the far field of a defined helium-neon gas laser reflex on a glass slide. We scanned the far field with a step size of 0.05°. For determining the relevant part of the photonic band structure the sample was aligned with the grating lines being perpendicular to the plane in which the fiber was moved. The sample was pumped below laser threshold. A detailed description of the setup for measuring the laser threshold was given in Ref. 14.

We fabricated six samples with nominal Ta_2O_5 layer thicknesses between 12 and 28 nm. To have a reference we fabricated one sample without a Ta_2O_5 layer. In the following we will refer to it as a sample with zero layer thickness. In Fig. 3 the angle dependent measurement of the emission spectra for the two limiting cases (d_{nom}=0 nm and d_{nom} =28 nm) are shown.

Our reference sample exhibits a stopband of about 3 nm. For a nominal thickness of 28 nm the resulting stopband is enhanced by more than a factor of 5 and amounts to 16 nm. The samples with an intermediate Ta_2O_5 layer show an increase in the band gap. Additionally in the band structure for the device with the thickest Ta_2O_5 layer (d_{nom}=28 nm) the TM-bands could be distinguished from the TE-bands as they are shifted about 15 nm toward shorter wavelengths. This could be observed for the other samples too.

The variation in the stopband as a function of the dielectric layer thickness is depicted in Fig. 4. A systematic in-

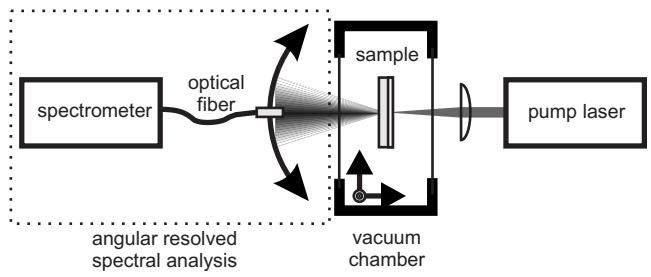

FIG. 2. Scheme of the measurement setup used for the angular resolved emission spectra analysis.

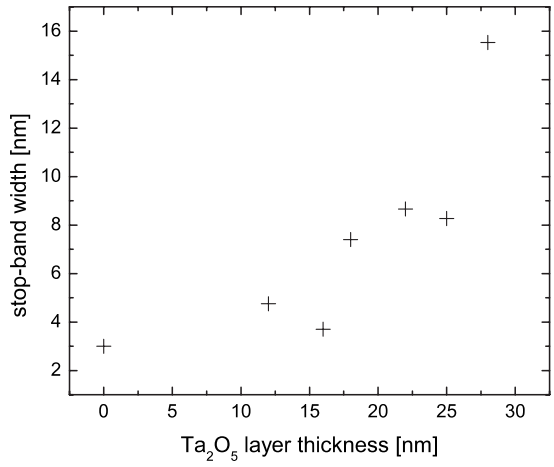

FIG. 4. Plot of the measured stopband width against the Ta_2O_5 layer thickness d_{nom}.

crease in the stopband for thicker Ta_2O_5 layers is observed.

To verify that these effects are caused by the inhomogeneous Ta_2O_5 layer we prepared an additional set of samples with a homogenous Ta_2O_5 layer with thicknesses up to 163 nm. Here we could not see any change in the stopband.

The measured laser spectra showed a peak wavelength between 641 and 653 nm and a spectral full width at half maximum (FWHM) of less than 0.3 nm (limited by the resolution of the spectrometer). The samples exhibit laser operation at the long wavelength side of the TE-stopband. The corresponding lasing thresholds were about 3 $\mu J/cm^2$. We did not see a significant dependence of the intermediate layer on the laser threshold.

In conclusion, we have modified the stopband of organic semiconductor DFB lasers by introducing a periodically patterned intermediate layer of Ta_2O_5. This layer was fabricated by oblique angle depositing the material onto a prestructured substrate. Using this technique we could enhance the stopband from 3 nm for the uncoated sample for a nominal dielectric layer thickness of 16 nm. This result could enable the exploitation of defect modes for low threshold organic semiconductor lasers where a broad band gap is essential.

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