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A very compact tunable solid state laser utilizing a thin film organic semiconductor

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Abstract

Optically pumped organic semiconductor lasers are fabricated by evaporating a thin film of tris-(8-hydroxyquinoline) aluminum (Alq_3) molecularly doped with a laser dye on top of a polyester substrate with an embossed grating structure. We achieve low threshold, longitudinally monomode distributed feedback laser operation. Varying the film thickness of the organic semiconductor film the wavelength of the surface emitting laser can be tuned over 44 nm. The low laser threshold allows the use of a very compact all solid state pump laser. The concept opens up a way to inexpensive and tunable lasers in the whole visible range.

Organic semiconductors have recently attracted much interest as active electroluminescent materials in organic light-emitting diodes (OLEDs) [1]. Performances in excess of 20 Lumen/Watt for the best LEDs render such devices very interesting for many applications. The high efficiency for the conversion of electrical energy into light and the broad spectral range of available emission colors has also triggered intense research towards optically [2-9] and electrically pumped [10] organic laser devices. Optically pumped organic semiconductor lasers have been realized based on thin spin-coated conjugated polymer films as well as on evaporated thin films of small organic molecules. For both classes of materials low threshold optically pumped gain narrowing and lasing has been observed for various wavelengths ranging from blue to red [11]. Various geometries such as microcavities, external resonators, Fabry-Perot type resonators, distributed feedback lasers (DFB) along with whispering gallery mode lasers have been demonstrated. Due to the low laser threshold and the broad gain spectra organic semiconductor lasers bear a large potential for many applications, e.g. for laser-based analytical techniques. The organic semiconductor thin film lasers were pumped by rather bulky and expensive lasers such as frequency-doubled/tripled Nd:YAG-lasers, N_2-lasers and complex femtosecond laser systems. Such lasers are well suited for basic studies of the laser properties of semiconductors, however, they do not take full advantage of the low laser threshold in organic semiconductor lasers. For most applications much more compact all solid-state pump laser sources are desirable. Additionally, approaches for achieving tunable monomode laser emission have to be elaborated.

In this letter we report on DFB-lasers utilizing thin evaporated films of the organic semiconductor tris-(8-hydroxyquinoline) aluminum (Alq_3) doped with the laser dye 4-dicyanmethylene-2-methyl-6-(p-dimethylaminostyryl)-4H-pyran (DCM) as the active medium. Longitudinal monomode laser emission in the spectral range between 604 nm and 648 nm is achieved. The emission wavelength can be tuned by adjusting the film thickness of the organic semiconductor waveguide. An extremely compact all solid state laser system can

be realized using a self-pulsating passively Q-switched Nd^{3+}:YAG microchip laser[12] with a monolithically integrated frequency tripler as pumping source.

The guest-host system Alq_3:DCM is one of the most promising materials for OLED applications and was also among the first organic electroluminescent materials that were used as the active material for optically pumped organic solid state lasers[4, 14, 15]. In an OLED the Alq_3 gives rise to the transport of charge carriers, in optical experiments photons with a wavelength around 400 nm are efficiently absorbed by Alq_3. DCM dissolved in various liquid organic solvents acts as a four-level system and is a commonly used laser dye having a high photostability and a large tuning range due to its broad gain spectrum[16]. In the composite system neutral excitations are rapidly funneled from Alq_3 to the DCM-molecules (exhibiting red-shifted optical transitions) by diffusion within the Alq_3 manifold and a subsequent energy transfer. This guest-host system is a potential candidate for tunable electrically pumped as well as optically pumped organic solid state lasers.

The time-dependent optical gain spectra $\gamma(\lambda,\tau)$ of Alq_3:DCM-films are measured via differential transmission spectroscopy using a femtosecond white-light pump-probe setup[17]. For these measurements Alq_3:DCM films evaporated on plane substrates are used. In order to suppress the unwanted effect of amplified spontaneous emission we use indium tin oxide (ITO) coated glass substrates[17]. The gain spectrum can be extracted from the differential transmission spectrum $\frac{\Delta T(\lambda,\tau)}{T(\lambda)}$ by correcting for the residual absorption. The broad spectrum in Fig. 1 shows the measured gain coefficient $\gamma(\lambda)$ for an excitation density of $2\cdot10^{20}cm^{-3}$, an excitation wavelength of 400 nm and a pump-probe time delay of $\tau = 30$ ps. We observe a broad spectral range of positive gain spanning a spectral region from 585 nm to 690 nm. The maximum gain coefficient is found to be $\approx 700cm^{-1}$. Based on the assumption that all DCM-molecules are excited (justified by the fact that the density of DCM molecules

of $\approx 5 \cdot 10^{19} cm^{-3}$ is much lower than the excitation density) we extract a cross section for stimulated emission of $\sigma_{SE} \approx 1.3 \cdot 10^{-17} cm^2$. This is in good agreement with the value reported for DCM in liquid solution[18]. The broad gain spectrum and the high absolute amount of gain show that the good laser properties of DCM are conserved in a solid matrix of Alq_3. We do not observe a strong competing photoinduced absorption due to neutral or charged excited species in this spectral range.

The inset of Fig. 2 shows a scheme of our all organic DFB-laser. As the substrate we use a flexible poly (ethylen terephtalate) (PET) sheet nanostructured by a UV-embossing process. Among other soft-lithography methods UV-embossing has proven to be very successful for the fabrication of nanopatterned plastic substrates for all organic solid state lasers[8,13]. DFB-gratings with a periodicity Λ of 400 nm (2^{nd} order grating) and a sinusoidal height modulation of ~100 nm were prepared. The organic film is deposited on top of the PET substrate by vacuum co-evaporation of Alq_3 and DCM. The DCM concentration is adjusted to approx. 2% by weight. The resulting organic semiconductor film has a higher refractive index than the surrounding air/substrate, thus forming a waveguide. In order to exploit the broad optical gain spectrum of Alq_3:DCM we have fabricated several DFB-lasers with varying thickness of the Alq_3:DCM-film. The film thickness d_f is varied from 120 nm to 435 nm. In our case the distributed feedback is provided by second order Bragg reflection, while the laser light is coupled out perpendicular to the film via first order Bragg diffraction (see inset of Fig. 2).

The different laser lines shown in Fig. 1 demonstrate that the laser wavelength can be tuned within the spectral region of gain via control of the film thickness d_f . The laser wavelength can be tuned over a spectral range of $\Delta\lambda_{Las}$=44 nm from λ_{Las}=604 nm for a 120 nm thick film to λ_{Las}=648 nm for a 435 nm thick film, respectively[19]. The experimentally determined peak positions of the laser emission are plotted versus the film thickness in Fig. 2. The effective refractive index n_{eff} governing the propagation of light within the waveguide

depends on the film thickness. This results in a thickness dependent Bragg resonance for the guided electromagnetic waves. The solid line in Fig. 2 shows the calculated wavelength dependency for a TE_0 mode in an unperturbed planar waveguide[20]. We have calculated $\lambda_{Las}(d)$ assuming n_f=1.75 for the organic semiconductor and n_s=1.505 for the substrate, taking into account that the wavelength is linked to n_{eff} via the Bragg condition $\lambda_{Las} = n_{eff} \cdot \Lambda$. The experimental data points correspond very well to the calculated curve for higher film thicknesses. Only for the thinnest films a small deviation is observed. This might be explained by the rather strong perturbation of the planar waveguide in our structure or by a wavelength dependent refractive index. We note that the range of laser wavelengths being accessible with one single grating is limited by the range of film thicknesses that give rise to a single mode waveguide. Whereas no guided wave is supported below d_f = ~110 nm, a second allowed TE_1 mode can appear for d_f > 470 nm.

The low laser threshold of our devices allows the realization of a very compact widely tunable all solid state laser. For this purpose we use a diode pumped passively Q-switched Nd^{3+} :YAG [12] laser with a monolithically integrated frequency tripler (*uniphase NanoUV-355*) as pumping source. The inset of Fig. 3 shows a scheme of the laser configuration. The pump laser produces sub-nanosecond pulses with a pulse energy of ~100 nJ at a wavelength of 355 nm and a 10.8 kHz repetition rate. The organic laser can be pumped without need of any focussing optics. The whole optical arrangement has a length of only ~10 cm and a volume of ~70 cm^3.

Fig. 3 shows the input-output characteristics of a laser utilizing a 300 nm thick Alq_3:DCM film. The observed laser threshold amounts to E_P= 2 nJ when the pump laser is focussed to an excitation spot with a diameter of 125 µm. Under the above mentioned conditions the durability of the organic laser in ambient atmosphere is limited to some 10^5 pulses. However, the lifetime is increased by several orders of magnitude by placing the organic sample in vacuum.

In summary we have realized a very compact, widely tunable all solid state laser on the basis of an organic film fabricated by vacuum co-evaporation of the organic semiconductor Alq_3 and the laser dye DCM onto a nanopatterned DFB-substrate. The substrate of the tunable organic DFB laser was fabricated by UV-embossing of a periodic structure into an acrylic-coated PET substrate. As pump laser a very compact diode pumped frequency tripled Nd^{3+}:YAG laser producing sub-nanosecond pulses at 355 nm can be used. The use of a tapered grating with spatially varying periodicity Λ and/or an evaporated film with a thickness gradient can provide a possibility for continuous tuning of the laser wavelength over the entire gain spectrum on one and the same substrate.

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Figure captions:

Fig. 1: The broad spectrum shows the spectral dependence of the gain coefficient measured via femtosecond differential transmission spectroscopy. The narrow lines are the laser spectra measured for samples with varying film thickness of the Alq_3:DCM-layer. For these measurements a regeneratively amplified Ti:Sapphire laser is used as the excitation source.

Fig. 2: Inset: Scheme of the distributed feedback laser. Films consisting of 98 % Alq_3 and 2 % DCM (by weight) are evaporated on top of a poly (ethylen terephtalate) (PET) substrate. Second order Bragg diffraction of waveguided light gives rise to the positive feedback necessary for laser operation. First order Bragg diffraction couples light out into a direction perpendicular to the substrate. Main part: The symbols represent the experimentally determined laser wavelengths for various film thicknesses. The solid lines are calculations based on the assumption of a planar waveguide.

Fig. 3: The inset shows the scheme of the very compact all solid state laser using the organic semiconductor film as active medium. The pump source is a commercially available diode-pumped, self-pulsating, frequency tripled Nd:YAG-microchip laser (*uniphase Nano UV - 355*). The organic semiconductor laser can be placed directly in front of the pump laser. Thus the total length of this very compact solid state "dye" laser is less than 10 cm. The main part of the figure shows the input-output characteristics at the laser wavelength.

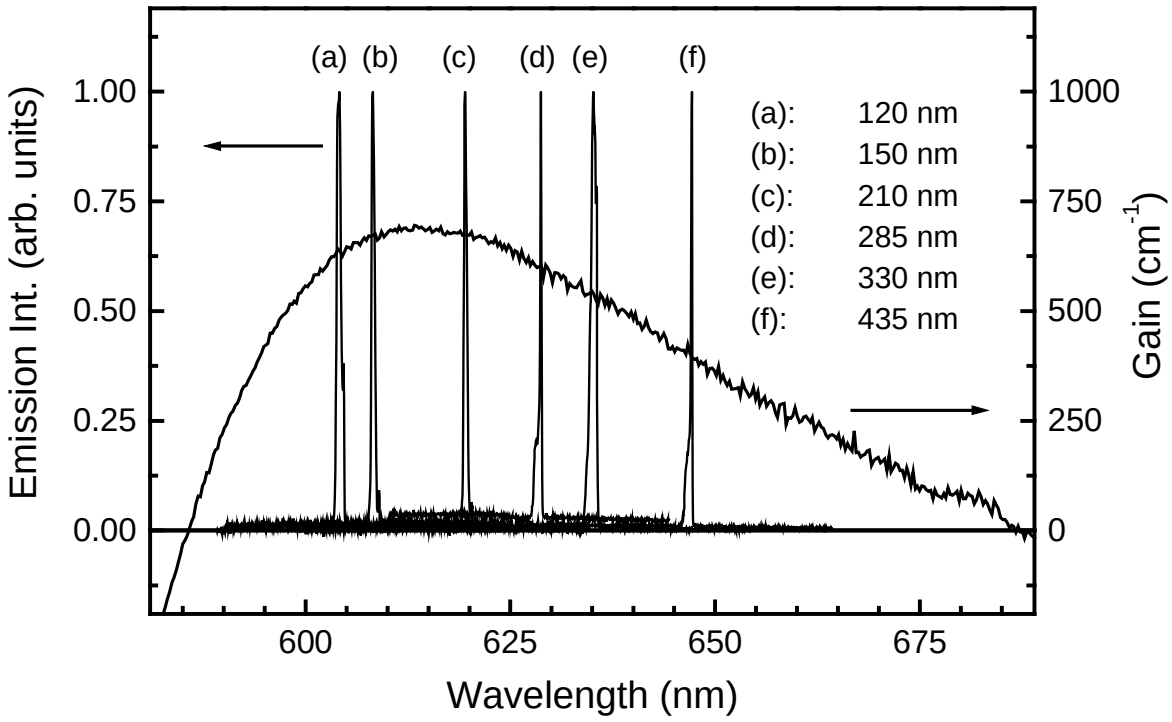
(a) (b) (c) (d) (e) (f)
(a): 120 nm
(b): 150 nm
(c): 210 nm
(d): 285 nm
(e): 330 nm
(f): 435 nm
Emission Int. (arb. units)
Gain (cm-1)
Wavelength (nm)
1.00
0.75
0.50
0.25
0.00
1000
750
500
250
0
600
625
650
675

Fig.1

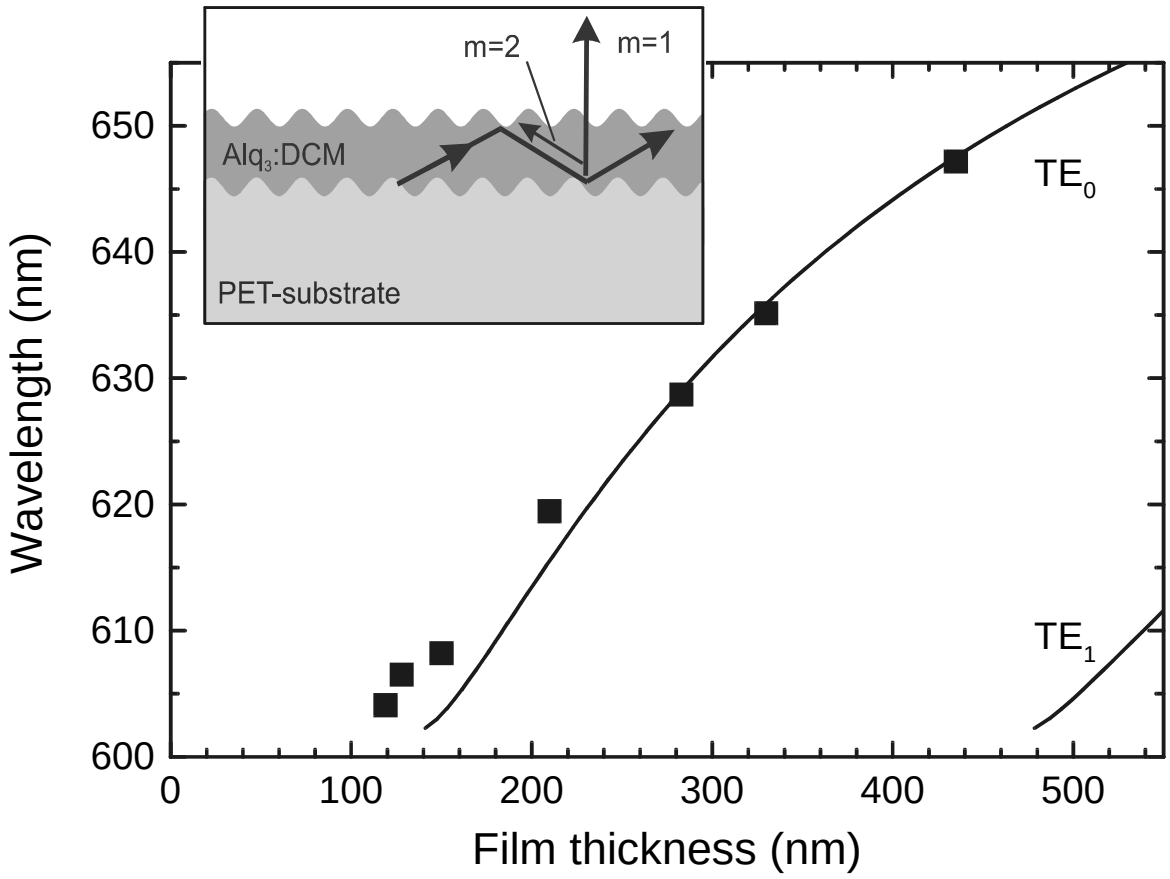
m=2
m=1
Alq3:DCM
PET-substrate
TE0
TE1
Wavelength (nm)
Film thickness (nm)
600
610
620
630
640
650
0
100
200
300
400
500

Fig.2

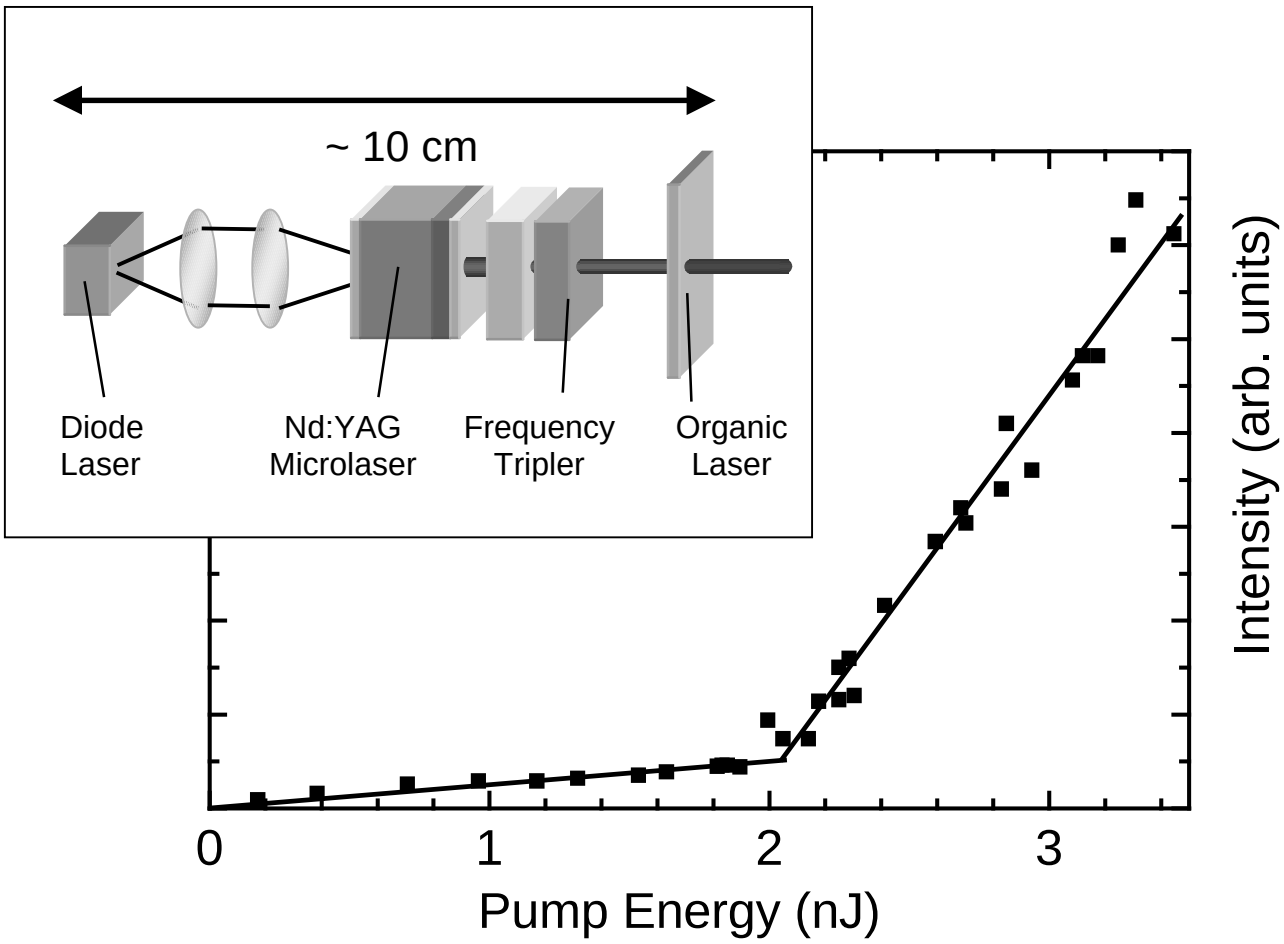
~ 10 cm
Diode Laser
Nd:YAG Microlaser
Frequency Tripler
Organic Laser
Intensity (arb. units)
0
1
2
3
Pump Energy (nJ)

Fig. 3