# Organic semiconductor distributed feedback laser fabricated by direct laser interference ablation

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**Abstract:** We use a pulsed, frequency tripled picosecond Nd:YAG laser for holographic ablation to pattern a surface relief grating into an organic semiconductor guest-host system. The resulting second order distributed feedback lasers exhibit laser action with laser thresholds being comparable to those obtained with resonators structured by standard lithographic techniques. The details of the interference ablation of tris-(8-hydroxyquinoline) aluminum ($Alq_3$) doped with the laser dye 4-dicyanomethylene-2-methyl-6-(p-dimethylaminostyryl)-4H-pyran (DCM) are presented and discussed. Lasing action is demonstrated at a wavelength of 646.6 nm, exploiting second order Bragg reflection in a relief grating with a period of 399 nm.

**OCIS codes:** (050.7330) Volume holographic gratings; (140.7300) Visible lasers; (140.3390) Laser materials processing; (140.3490) Lasers; distributed-feedback; (140.5960) Semiconductor lasers; (160.4890) Organic materials

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## 1. Introduction

The rapid development in the field of organic light-emitting diodes (OLEDs) and the progress in the achieved maximum current densities is inspiring the research on the realization of low threshold optically pumped organic semiconductor lasers as well as organic injection lasers. For these devices, a resonator providing positive optical feedback needs to be realized. One and two dimensional distributed feedback lasers have been identified as promising resonator geometries for low threshold organic lasers [1]. While large area and potentially low-cost deposition is a clear advantage of organic semiconductors, the patterning of suitable resonators comprising organic semiconductors is a severe challenge. The high solubility of most organic molecules prevents the use of conventional electron-beam- or photo-lithographic patterning techniques. Most of the organic semiconductor devices realized so far utilizes a prestructured substrate to induce optical feedback. The actual planar waveguide is formed by depositing the organic semiconductor onto the existing surface nanostructure of the substrate.

Alternatively, a surface relief grating (SRG) on top of an active layer can be used to induce the feedback. This has been realized, e.g., by using a UV-processable material on top of a dye doped polymer. However, special UV-photopolymers are needed for this purpose [2]. The incorporation of a surface relief grating (SRG) into the active layer has also been demonstrated using a dye doped polymer [3].

For future electrically pumped devices the direct incorporation of the feedback structure into the organic semiconductor might be advantageous, reducing the number of processing steps. Nanoimprinting techniques have been used to fabricate SRGs directly into semiconducting organic materials. Conjugated polymers as well as small molecular guest host systems have been patterned directly [4-6]. The easiness of fabrication by using nanoimprinting techniques can be quite attractive. However, a significant drawback is the need of a high quality master stamp.

Alternatively, holographic laser ablation (HLA) can be a versatile and high-yield patterning method. This method has been used successfully for structuring insulating polymers [3] or hard optical materials and waveguides [7].

Herein, we report the high efficiency distributed feedback laser action obtained by the host system tris-(8-hydroxyquinoline) aluminum ($Alq_3$) doped with the laser dye 4 dicyanomethylene-2-methyl-6-(p-dimethylaminostyryl)-4H-pyran (DCM). The distributed feedback action was provided by a relief Bragg grating that was inscribed into the organic semiconductor using direct 355 nm, 150 ps Nd:YAG laser holographic ablation.

## 2. Experimental setups

The active material was produced by co-evaporation of a 175-300 nm thin film of $Alq_3$:DCM on glass, in a high vacuum apparatus at a pressure of $3 \times 10^{-5}$ Pa. The doping concentration was adjusted to 5 mol% during co-evaporation of the two materials. The refractive index of this well established semiconducting host laser material is approximately 1.76 [8, 9]. Therefore, an asymmetric slab waveguide is formed in combination with the underlying glass substrate. All grating ablation experiments were performed in ambient atmosphere, keeping the laser repetition rate fixed at 10 Hz. A frequency tripled Nd:YAG-laser was used for the emission of 150 ps pulses at a wavelength of 355 nm. The pulses were injected in a simple two-mirror interferometric cavity. Figure 1 shows a scheme of the setup. A fused silica phase mask of 1050.7 nm pitch was used as a beam splitting element, while two aluminum coated mirrors were used for refolding the ±1 diffracted orders onto the sample surface. The two

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interfering beam paths were precisely aligned in terms of temporal and spatial overlap for achieving maximum recording contrast onto the exposed sample. The constructive and destructive interference of the two beams defined a pattern of dark and bright lines. By adjusting the relative angle between the two interfering beams, the recording periodicity was set to be 399 nm. The grating period was tuned at that wavelength for optimal phase matching with the peak of the amplified spontaneous emission (ASE) spectrum of the $Alq_3$:DCM waveguide, located at ≈ 643 nm. For second order Bragg reflection the wavelength is at $\lambda_{\mathrm{Bragg}} = \Lambda \times n_{\mathrm{eff}}$, where $n_{\mathrm{eff}}$ is the effective refractive index of the waveguided mode and $\Lambda$ is the grating period. An effective refraction index $n_{\mathrm{eff}}$ of 1.61 was assumed. This Bragg reflection is used for inducing optical feedback.

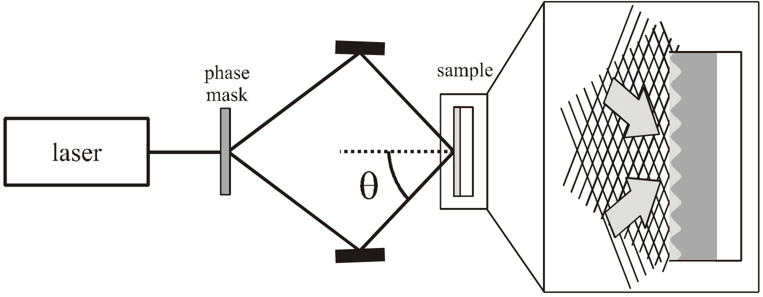

Fig. 1. Scheme of the ablation set up. The enlarged part visualizes the definition of the pattern onto the substrate.

The distributed feedback region was optically inspected by a Littrow-geometry experiment. The diameter of the ablation spot during the interference ablation was about 3.6 mm. Investigations on the (non-interferometric) ablation process were done using a rectangular ablation area of 36 mm².

The setup for the investigation of the laser characteristics is shown in Fig. 2. A frequency tripled Nd:$YVO_4$ laser as a pump source emitting at 355 nm was used (500 ps pulses at 1250 Hz). The size of the slightly elliptical spot was measured to be 298 µm × 264 µm by the knife-edge method. In order to prevent degradation of the organic layer during the lasing experiments the samples were kept in a dynamic vacuum at a pressure below $5 \times 10^{-3}$ Pa. The pump beam was reflected at an angle of 45° by a dichroic mirror, which is reflective for wavelengths below 400 nm. As we used second order distributed feedback lasers, the laser emission is coupled out perpendicular to the waveguide due to first order Bragg diffraction. This emission was collected, passed the dichroic mirror and was focused onto an optical fiber and detected in a subsequent spectrometer.

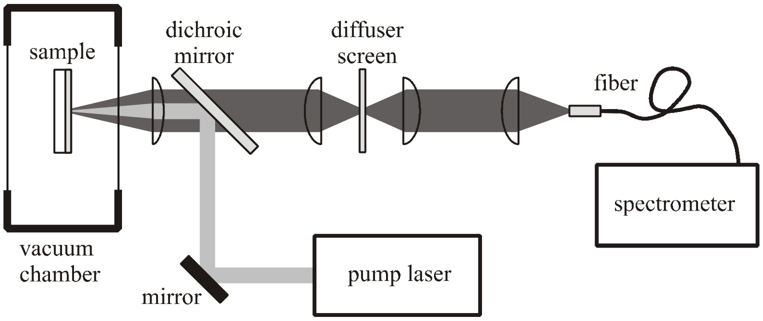

Fig. 2. Scheme of the laser characterization set up. For minimizing angle dependent effects, the collected light is focused onto a diffuser screen intermediately.

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## 3. Results and discussion

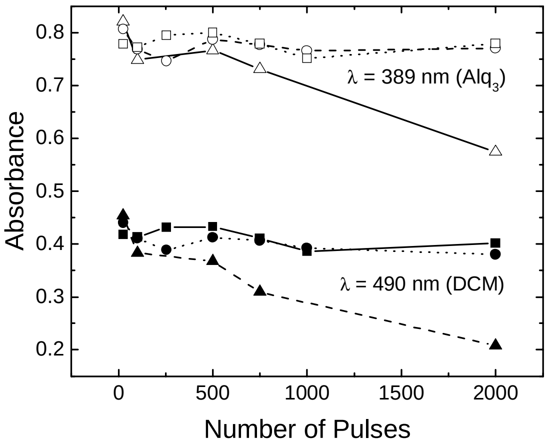

Fig. 3. Homogenous ablation of 300 nm thick $Alq_3$:DCM layers. The absorption coefficients at the absorption maximum of $Alq_3$ (open symbols) and of DCM (filled symbols) are plotted for ablation fluences of 5 mJ/cm² (squares), 7 mJ/cm² (circles) and 13 mJ/cm² (triangles) in dependence of the number of pulses.

For the determination of the optimum ablation conditions the samples were ablated using a variety of exposure conditions. Energy densities of 5, 7, 13 and 20 mJ/cm² were applied for quantifying the ablation yield. We have measured the absorption at the main absorption bands of $Alq_3$ (at $\lambda = 389$ nm) and of DCM (at $\lambda = 490$ nm), respectively, as a function of the number of pulses. For fluences larger than 20 mJ/cm² the material was completely ablated after only few pulses. In Fig. 3 the results of the absorbance measurements are presented. Using fluences of 5 and 7 mJ/cm$^2$ no significant absorption changes are detected. Thus we conclude a negligible ablation for these conditions. For a fluence of 13 mJ/cm² the measurements show a significant change of absorption. We conclude that approximately 10 mJ/cm² are needed for a controlled laser ablation. The slightly stronger decrease of the DCM absorption as compared to the $Alq_3$ absorption is attributed to a stronger photoinduced damage of the DCM molecules during the ablation process.

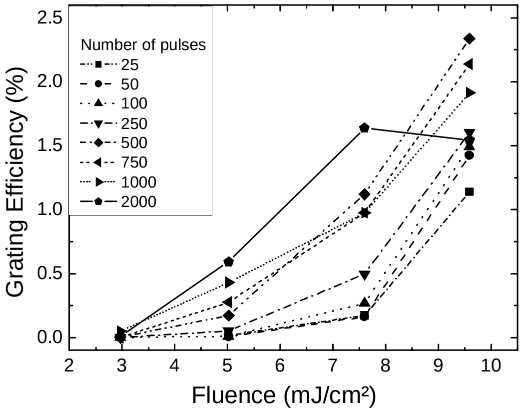

Fig. 4. Measured grating diffraction efficiencies of 175 nm thick $Alq_3$:DCM layers versus energy density of the exposure, for different number of pulses.

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For studying the interference ablation effect we have measured the diffraction efficiencies of the resulting grating structure as a function of the exposure parameters. Figure 4 shows the relation between grating efficiency and fluence for several numbers of pulses. For a fluence of 3 mJ/cm² no diffraction was measured for exposures up to 2000 pulses. Furthermore, even after 12500 pulses exposure an efficiency of only 0.15 % was measured (not shown in Fig. 4).

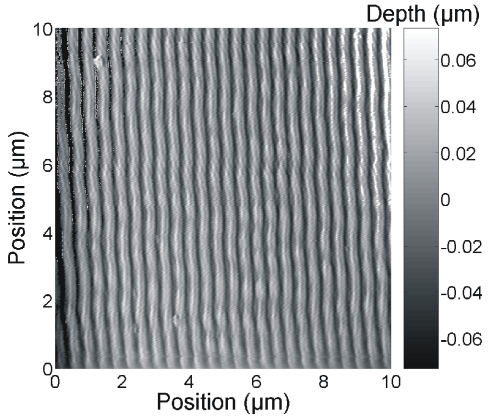

Fig. 5. AFM picture of a relief Bragg reflector ablated using 500 pulses and a fluence of 5 mJ/cm².

Energy densities in the range between 5-10 mJ/cm$^2$ are of higher yield. The previous is in agreement with the findings for the homogeneous ablation mentioned above. Due to the twofold intensity enhancement in the interference maxima, local laser ablation took place at approximately 50 % of the ablation threshold for homogeneous exposure. For the highest dose used in our experiments (2000 pulses with a fluence of 10 mJ/cm$^2$) the diffraction efficiency exhibits a non-monotonic behavior, starting to decrease at a certain point of the exposure. Prolonged exposures result in a damage of the grooves by exfoliations or ablation in the dark fringes [10]. An AFM-image of a relief grating structure ablated using 500 pulses of 5 mJ/cm² energy density is shown in Fig. 5. The grating structure appears to be smooth, free of redeposition debris and groove defects, effects that can result in the generation of parasitic spectral features when such periodic structures are used as Bragg reflectors.

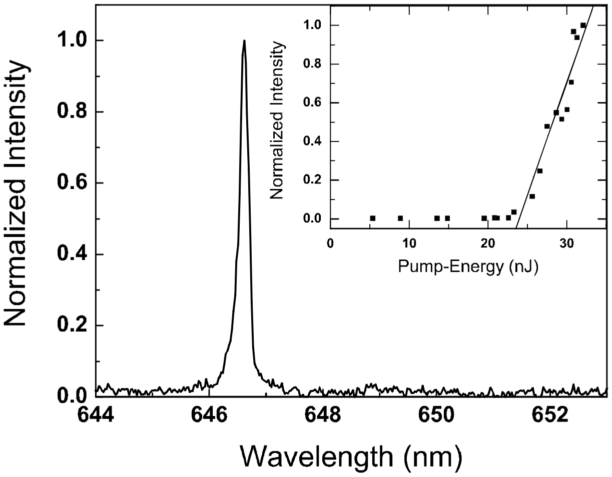

Fig. 6. Laser spectrum measured on a 300 nm Alq$_3$:DCM layer with a 399 nm grating ablated with 500 pulses and fluence 5 mJ/cm² and. Inset: Laser line. The threshold can be identified to 24 nJ.

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Lasing activity was observed for a wide range of ablation parameters of the processed films. Namely, spectrally narrow laser lines, indicative of distributed feedback, were measured for the majority of the processed samples, confirming the low-optical and structural damage effect of the ablation method. A resulting laser spectrum and the corresponding input-output characteristic for a sample that has been patterned with 500 pulses and fluence of 5 mJ/cm² is shown in Fig. 6. A laser line is emitted at the wavelength of 646.6 nm (indicating an effective refractive index of 1.62 of the waveguided mode) exhibiting a spectral width of 0.2 nm (FWHM). The inset of Fig. 6 shows the input-output characteristic of the corresponding laser. The laser threshold is determined to 24 nJ or 41 µJ/cm². This lasing threshold is slightly higher than previously reported results for $Alq_3$:DCM films on pre-patterned substrates [9].

## 4. Conclusion

In conclusion, we have shown lasing in organic semiconductor distributed feedback lasers being prepared by 355 nm, 150 ps Nd:YAG laser holographic ablation. The laser threshold and the emission behavior are comparable to the case of lasers using a pre-patterned substrate. The results presented may constitute a promising starting point for the exploitation of direct ultraviolet laser ablation approaches for the straightforward patterning of electrically pumped organic semiconductor lasers.

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