Low-threshold polymeric distributed feedback lasers with metallic contacts

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Optical losses in waveguides comprising metallic contacts are thought to be a major hurdle to the realization of organic laser diodes. We demonstrate here that careful tuning of the waveguide mode in flexible distributed feedback lasers can allow lasing action to occur in organic thin films in the presence of contacting electrodes with virtually no difference when compared to metal free devices. A metallic electrode is most suited as the bottom contact between the polymer and the substrate as it reduces mode leakage into the substrate and enhances modal gain. In contrast, a thin transparent electrode such as a metal oxide is preferable for the top electrode, where confinement is not a problem. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1712029]

Organic semiconductors are attractive gain media for novel wide gap tunable lasers.[1–6] Substantial effort has been directed toward improving the tunability and thresholds of optically pumped lasers, but thus far it has not been possible to obtain stimulated emission under electrical pumping. A major obstacle to achieving this is believed to be the interaction of excited state species in the gain medium with the electrodes, which leads to quenching of the excited state. As disordered organic semiconductors such as evaporated small molecules or solution processable conjugated polymers have very low mobilities, the high current densities required for potentially achieving population inversion can only be reached at large fields in thin planar sandwich configurations.[4] Previous attempts to study laser action in organic materials in the presence of an electrode layer have shown a substantial rise of the lasing threshold if not a total absence of the lasing mode.[4–7] We show here that careful tuning of the actual thin film waveguide properties can minimize the interaction between the gain mode and the quenching channel of the electrodes. We are able to achieve a fully contacted laser structure with a mere 30% increase in laser threshold, and demonstrate that metallic layers can even lead to a *reduction* in threshold by enhancing the confinement factor and thus the modal gain of the laser.

When considering the interaction between the gain material and the electrode it is helpful to distinguish between two mechanisms. The obvious source of loss is absorption by the electrode. However, an electrode inserted between the polymer and the distributed feedback (DFB) substrate or placed on top of the polymer layer will also modify the waveguiding properties of the DFB laser structure. Figure 1(a) indicates the profile of the TE_0 mode for a 300-nm-thick polymer film, which was calculated using the procedure described in Ref. 8 for a planar substrate. The profile is projected on top of a grating structure to provide a rough guide to the laser mode. The mode penetrates considerably into the substrate, but displays a better confinement at the polymer/vacuum interface. Mode penetration results in an increase in lasing threshold, as it reduces the modal gain which is the product of confinement factor (here 0.83) and material gain. Inserting a low index material in between the polymer and the grating substrate should improve the mode confinement. The real part of the refractive index of metals is generally small and often <1, so that insertion of a metal layer is expected to improve confinement. Figure 1(b) shows the calculated waveguide mode for the case where a 150-nm-thick silver layer is inserted between the gain material and the substrate. In this case the confinement of the mode to the gain medium is improved (confinement factor of 0.91), thereby reducing the potentially detrimental effect of mode

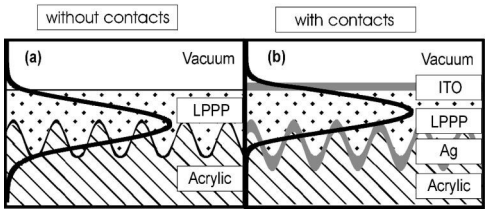

FIG. 1. Calculated TE_0 mode of a 300-nm-thick polymer film superimposed on a DFB structure. (a) without contacts; (b) with ITO and silver contacts.

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leakage into the substrate. However, the absorptive loss of a metal layer should generally exceed the benefit in mode confinement. Also included in the calculation in Fig. 1(b) is a 20-nm-thin indium tin oxide (ITO) layer on top of the polymer. A comparison with Fig. 1(a) shows that the mode is not perturbed by this top electrode layer.

In order to test whether we can differentiate between the effect of mode confinement and absorptive losses in polymer DFB lasers we fabricated flexible DFB lasers with a range of contacts. One-dimensional DFB gratings were formed by nanopatterning an acrylic coating on a polyester substrate by UV embossing, which leaves an almost sinusoidal height variation of ~330 nm with a pitch of 300 nm. We chose a ladder-type poly(para-phenylene) (LPPP) as the gain medium, which is known to exhibit excellent lasing characteristics.[3,9] The polymer layer was deposited on top of the corrugated plastic substrate by spin coating. The grating induces optical feedback of the waveguided modes via second order Bragg scattering. Atomic force microscopy measurements of the surface of the laser as well as scanning electron microscopy on fractured devices showed that the polymer levels the deep corrugation of the grating, leaving a virtually flat surface at the top. Film thicknesses were measured by absorption, which provides a spatial average over the deep corrugation.

The laser structures were characterized in vacuum by exciting at 400 nm using 110-fs light pulses from a 1 kHz frequency-doubled regeneratively pumped titanium-sapphire laser focused to a spot of 95 μm diam. The emission was detected through a scattering plate to ensure isotropic collection and recorded using a CCD and grating monochromator. It is not straightforward to compare the laser thresholds between different structures, as metallic layers modify the reflection and transmission of the samples. To deal with this, we compared a number of different device structures with and without contacts on a single substrate and measured both the transmission and reflection. It is helpful to state the actual excitation density in the gain medium rather than the excitation intensity used. We estimated the excitation density in the film by considering the total amount of absorbed excitation pulse energy in the active layer corrected for reflection and transmission losses (the absorption coefficient of LPPP is 4.2×10^4 cm^{-1} at 400 nm). Also, it should be noted that the material gain $\gamma(\lambda)=n_{\mathrm{Exc}}\times\sigma_{\mathrm{SE}}(\lambda)$ depends strongly on wavelength,[9] where n_{Exc} is the excitation density and σ_{SE} is the cross section for stimulated emission. Inclusion of different contacts modifies the waveguiding properties slightly so that lasing occurs at marginally different wavelengths for each geometry, thereby experiencing a different net gain. All lasing modes were observed within 2 nm of the gain maximum, so that σ_{SE} can be treated as a constant thus allowing a direct comparison of the thresholds of different device structures. As seen in the following the corrections accounted for relative shifts of the threshold curves of at most 50% with respect to each other.

We investigated the effect of both metallic and ITO contacts. Metallic layers were thermally evaporated under vacuum whereas ITO was carefully rf-magnetron sputtered at a low rate of 11 nm/min under a pure argon atmosphere of 5 mbar. The deposition was carried out at room temperature

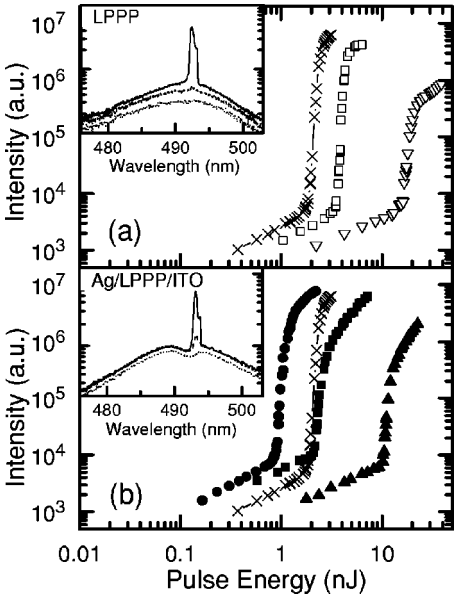

FIG. 2. Laser characteristics of different LPPP based DFB lasers. (a) Effect of different top electrodes: (×) neat LPPP (240 nm thick); (□): LPPP/ITO (285 nm); (▽): LPPP/Au (350 nm). (b) Effect of a silver bottom electrode: (×) neat LPPP (240 nm thick) (for comparison); (●): Ag/LPPP (350 nm); (■): Ag/LPPP/ITO (285 nm); (▲): Ag/LPPP/Au (350 nm). The insets show emission spectra recorded below and above threshold at (a) 1.22 nJ, 1.71 nJ, 2.02 nJ and (b) 1.85 nJ, 2.15 nJ, 2.3 nJ.

from ceramic targets (90% In_2O_3/10% SnO_2, with 99.99% purity). For each configuration we fabricated several samples with different film thicknesses. The lowest laser thresholds were found for a film thickness of 240 nm in the absence of any contacts, for 350 nm when using either a silver bottom contact or a gold top contact and for 285 nm when using silver and ITO together. It is known that using typical ITO layer thicknesses of 100 nm strongly suppresses optical amplification in the gain medium due to efficient waveguiding in the ITO layer[5,6,10,11] resulting from the relatively high index of refraction of ITO. In our studies we chose a layer thickness of 20 nm as a compromise between high electrical conductivity and low optical loss. The comparatively high sheet resistance of 500 Ω/□ poses no intrinsic limit to achieving high current densities providing that laterally patterned highly conducting leads are included.[6,11]

Figure 2(a) compares the laser characteristics for different structures. All lasers exhibit a well-pronounced threshold behavior, which is shifted to higher pulse energies upon deposition of a top ITO or gold contact. In contrast, a bottom silver contact reduces the threshold, as shown in panel (b). A sample with both a bottom silver and a top ITO contact (squares) exhibits virtually the same threshold as a device with no contacts (crosses). The threshold is only increased substantially when a gold top electrode is used together with a silver bottom electrode (triangles). The insets show emission spectra recorded slightly above and below threshold. A very clear threshold behavior is observed with the appearance of a narrow laser peak over a pulse energy change of only 10%. For both metal free and contacted devices, the laser line is merely 0.5 nm wide. The only apparent difference between the two structures is the increase in the Bragg dip at 493 nm in the presence of the metal grating.

In order to be able to compare the threshold behavior

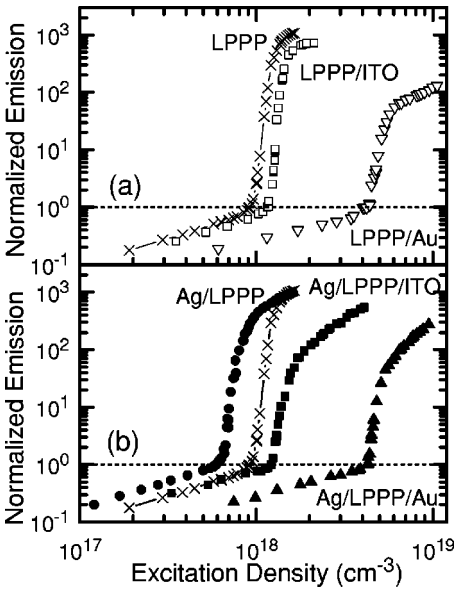

FIG. 3. Corrected laser characteristics in terms of excitation density. (a) Effect of different top electrodes: (×) neat LPPP; (□): LPPP/ITO; (▽): LPPP/Au. (b) Effect of a silver bottom electrode: (×) neat LPPP; (●): Ag/LPPP; (■): Ag/LPPP/ITO; (▲): Ag/LPPP/Au.

quantitatively, we estimated the actual excitation density for every structure as stated above. Figure 3(a) shows the laser threshold characteristics obtained for two different top contacts compared to a neat polymer film (crosses). The neat polymer structure exhibits lasing behavior above an excitation density of 9×10^{17} cm^{-3} corresponding to an energy of 2 nJ per pulse. Deposition of a 20 nm layer of ITO on top of the polymer only slightly raises the laser threshold and does not modify the lasing characteristics. ITO introduces only very little absorption loss and the thin layer does not significantly modify the confinement of the waveguide mode. In contrast, deposition of a thin (10 nm) semitransparent layer of gold as the top electrode raises the threshold by a factor of 4 due to strong absorption.

The case is very different when one considers the bottom contact of the diode structure at the grating-polymer interface. Figure 3(b) shows gain characteristics for a silver bottom contact with different top contacts compared to a metal free structure. Surprisingly, inserting a 150-nm-thick opaque silver layer between the polymer and the substrate results in a 30% *decrease* in laser threshold (circles) when compared to a polymer only structure (crosses). The benefit in confining the waveguide mode achieved by the bottom metal contact thus outweighs the absorptive loss inherent to the metallic layer. Subsequent deposition of a top ITO contact (squares) almost doubles the laser threshold, whereas a top gold contact gives rise to a sevenfold rise. The threshold characteristic of the Ag/LPPP/Au device is virtually identical to that of the LPPP/Au device, showing that the performance is limited by the gold absorption. However, the effect of enhanced mode confinement with the silver layer becomes apparent above the threshold, as the Ag/LPPP/Au device characteristic (filled triangles) exhibits a steeper slope than the LPPP/Au device (open triangles).

We should stress that in a previous similar experiment an increase by a factor of 150 of the lasing threshold was observed for a silver backed polymer laser.[7] In contrast to our work, however, the structure in Ref. 7 also exhibited only weak threshold behavior and spectrally rather broad emission. Our results demonstrate that the common notion that contacting a thin film polymer laser structure by including a metallic layer be detrimental to operation due to dominating absorptive losses does not necessarily hold. For sufficiently thick polymer films, insertion of a metallic bottom contact actually results in a *reduction* in threshold. This can be due either to a reduction in loss, an increase in gain, or a confinement enhancement. As it is highly unlikely that the losses are reduced or even the gain enhanced by the metal, we conclude that the confinement is increased, and thus the modal gain. Covering the polymer with a thin transparent electrode barely modifies the confinement and introduces almost no additional absorption losses. A polymer laser structure may therefore comprise a bottom metal electrode for electron injection and a top, hole injecting ITO electrode. As LPPP has an exceptionally high charge carrier mobility together with rather narrow polaronic and excitonic absorption features as well as very low trap densities this material should be a suitable candidate for achieving electrically pumped lasing. Also, wavelength scale microstructure does not modify the electrical properties of polymer sandwich-type structures and thus poses no limitation to carrier injection.[12] DFB-type sandwich structures therefore appear to be the most promising avenue to pursue.

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