Hybrid optoelectronics: A polymer laser pumped by a nitride light-emitting diode

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We demonstrate indirect electrically pumped lasing in a hybrid polymer laser. The lasers comprise a corrugated fluorene copolymer waveguide on an InGaN light-emitting diode and were driven under nanosecond pulsed operation. We observe the onset of distributed feedback lasing at 568 nm for peak drive currents above 144 A. Angle-resolved photoluminescence measurements identify the lasing mechanism as band edge feedback from a photonic stopband in the TE_0 waveguide mode. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2912433]

The visible light emission of semiconducting conjugated polymers, combined with their simple fabrication as optoelectronic devices, make them attractive candidates for lasers that can access the full visible spectrum,[1–6] but so far they have been unable to realize their full potential because they require an additional laser to excite them. Although great progress has been made in reducing the size of the pump laser required,[7–10] it remains an expensive, delicate, and inconvenient component. Electrical excitation would transform the situation but has so far proved impossible because of the high current densities required, losses due to the presence of contacts, and, most seriously, absorption by the injected charge carriers.[4,6,11,12] All these issues relate to the low mobility of these disordered materials. Here, we show that this problem can be overcome by taking advantage of the complementary properties of inorganic semiconductors. We show that by separating the charge transporting and lasing regions in a compact package combining an indium gallium nitride light-emitting diode (LED) with a semiconducting polymer distributed feedback (DFB) laser, an electrically pumped hybrid polymer laser can be made. Our hybrid optoelectronic approach provides a new route to compact and low-cost visible lasers with the potential for applications in security, sensing, spectroscopy, and medical diagnostics.

To pump a laser above threshold, one must create a population inversion density of sufficient magnitude that optical gain can surpass the round-trip losses of the laser resonator. In the case of fluorescent organic lasers, the short fluorescence lifetime (typically $\leqslant$1 ns) demands very intense pumping to attain a substantial inversion density. For laser pumping, the intense excitation needed may in part be achieved by tightly focusing the pump beam through the gain medium. However, the situation is rather more challenging for LED pumping since the emission from a LED is incoherent and highly divergent, making it difficult to collect and then tightly focus the light. Historically, LEDs have also been very limited in output power and in the available emission wavelengths. However, recent developments in III-N LEDs have extended their emission into the blue and even ultraviolet.[13–15] Considerable advances in high-power LEDs, driven by solid-state lighting applications, now mean that single emitters may generate hundreds of milliwatts of light with efficiencies exceeding 10%. While the highest photometric powers (i.e., weighted by the human eye response) are in the green part of the spectrum, higher radiometric powers are possible in the blue at around 450 nm. Therefore, to achieve the goal of LED pumped organic lasers, one requires a suitable gain medium whose absorption will match the optimum output wavelengths of the LED, configured in a low-threshold laser structure that can convert the limited brightness of the LED pump into a substantial population inversion density.

For the polymer gain medium, we selected a commercially available fluorene co-polymer (ADS223YE, American Dye Source Inc.), which is shown in the inset of Fig. 1. Thin films of ADS223YE were spin cast from solution in chlorobenzene onto silica substrates, and their thicknesses were determined by using a Dektak-3 surface profiler and absorbance measurements. The absorption of this material is maximum around 370 nm with a smaller shoulder at 450 nm, overlapping well with the emission from nitride LEDs [Fig. 1(a)]. The green-yellow photoluminescence (PL) peaks at 530 nm and has a high solid-state PL quantum efficiency of 80%. The PL lifetime is 1 ns, which is relatively long for conjugated polymers and is useful for achieving a population

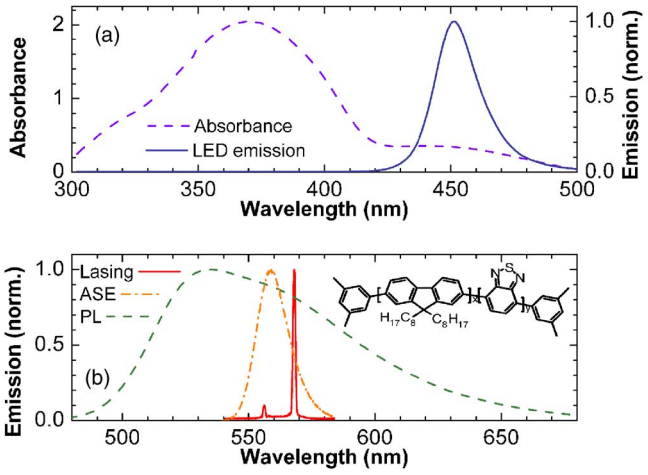

FIG. 1. (Color online) (a) Absorption spectrum of a 350 nm thick film of ADS233YE (dashed line) and emission spectrum of Luxeon K2 LED (full line). (b) PL, ASE, and DFB lasing spectra of ADS233YE. Inset: Chemical structure of ADS233YE.

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inversion with modest pumping intensity. The polymer was initially assessed for use as a laser gain material using measurements of amplified spontaneous emission (ASE). A 150 ± 10 nm thick film of the polymer was optically pumped with 4 ns pulses from an optical parametric oscillator (OPO) at 450 nm in a stripe of dimensions of 2.3 mm $\times$ 170 μm. Light emitted from the edge of the film was detected by using a fiber-coupled charge-coupled device (CCD) spectrograph. When pumped above a pulse energy of 440 nJ, the light emitted from the film edge was observed to spectrally narrow to a peak of 15 nm full width at half maximum (FWHM), centered at 559 nm, as shown in Fig. 1(b). This intensity-dependent line narrowing is attributed to the optical amplification of spontaneous emission, which is waveguided through the polymer film and indicates the optimum wavelength for lasing.

The polymer was also assessed for lasing in DFB resonators. The DFB lasers tested comprised a thin film of the ADS223YE polymer deposited on a corrugated fused silica substrate, which planarizes the corrugated surface. The period of the sinusoidal corrugation was chosen to give in-plane DFB near the ASE wavelength and a surface-emitted output coupling via second- and first-order Bragg scattering, respectively. The optical waveguide was completed by depositing a thin film of the low-index fluorinated polymer CYTOP (Asahi Glass Co. Ltd.) on top of the polymer to act as a transparent barrier layer to oxygen and water. The silica-ADS223YE-CYTOP structure formed an asymmetric slab waveguide that typically supported the lowest order transverse-electric (TE_0) and transverse-magnetic (TM_0) modes within the polymer emission band. The polymer lasers were excited with a circular beam of diameter of 1.7 mm and wavelength of 450 nm with the OPO, and the surface emission was detected by using the fiber-coupled CCD spectrograph. Lasing occurred for pumping intensities above 200 W cm^{-2}, and by double passing the pump beam through the polymer film, the threshold could be reduced to 150 W cm^{-2} Figure 1(b) shows a typical emission spectrum from a laser with a 355 ± 14 nm ADS223YE film on a silica grating of 355 nm period and 70 nm depth. Above threshold, the laser emission is characterized by the appearance of a narrow peak at 568 nm that rapidly grows at higher pumping levels to dominate the emission spectrum. At higher powers, a second weaker peak emerges at 556 nm. We identify the main peak to DFB lasing of the TE_0 mode and the subsidiary peak at 556 nm to lasing in the TM_0 mode. We observed no significant drop in output power for 1 h of operation.

For subsequent LED excitation of the polymer, we have used Luxeon emitters from Philips Lumileds. The Luxeon K2 emitter (LXK2-PR14-Q00) is specified to emit typically 620 mW at a continuous drive current of 1.5 A. The emission area of the device is 1.5×1.5 mm^2, giving a maximum possible emission intensity of 28 W cm^{-2}. This value is currently too low for pumping organic semiconductor lasers, and so we explored instead the performance of these LEDs under short pulse operation. The Luxeon K2 was driven with short current pulses from a laser diode driver (PCO-7110-120-15, Directed Energy, Inc.) at a repetition frequency of 20 Hz. Figure 2 shows a typical temporal profile of the current pulse, plus the corresponding optical pulse as measured by using a fast photodiode of 2 GHz bandwidth. The drive current pulses were triangular in shape with a FWHM of 36 ns. The optical pulses had a similar profile to the current but were slightly longer at 47 ns. The pulse duration and output energy of the LED were measured for a range of drive currents by using a calibrated optical energy detector. Figure 2 shows the variation of peak output intensity with drive current. We found that the peak output intensity linearly increased with drive currents up to 255 W/cm^2 at 160 A.

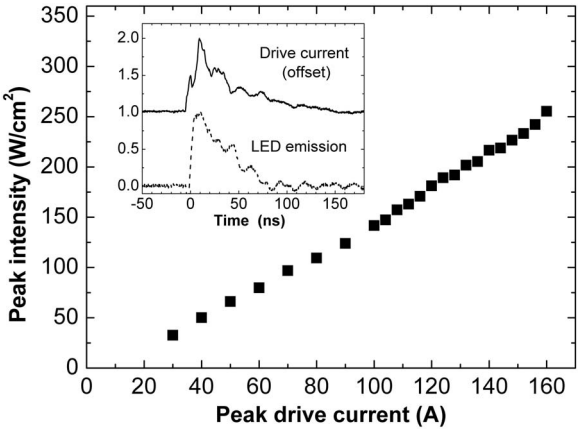
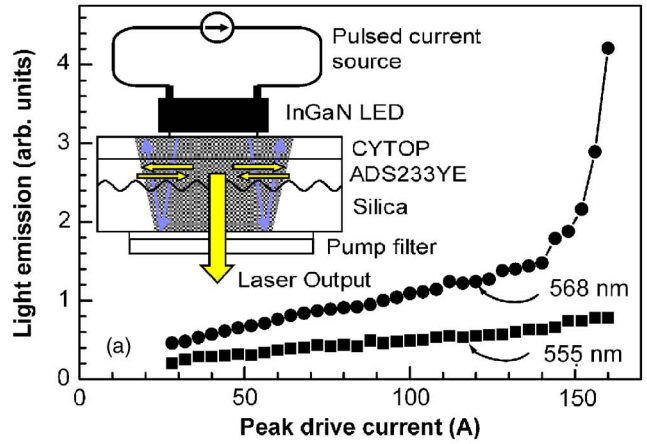


FIG. 2. Variation of the pulsed light intensity of the Luxeon K2 LED with drive current. Inset: Temporal characteristics of the drive current (top) and light output (bottom) of the LED.

The peak output from the LED exceeds the minimum threshold intensity measured for lasing when pumped by the coherent beam from the OPO, and so we went on to assemble the hybrid polymer laser shown in the inset of Fig. 3. First, the plastic lens of the Luxeon K2 LED was removed to allow close access to the LED surface. The polymer DFB laser was fabricated as above and then placed in contact with the LED, so that the CYTOP served as a low-index optical buffer layer between the higher index LED and semiconducting polymer. Finally, a dichroic mirror was mounted in contact with the rear surface of the silica substrate. This mirror had a reflectivity of 98% at 450 nm and a transmission of >90% around 570 nm, and so acted to double pass the LED pump light through the gain medium while transmitting the polymer light emission. The hybrid device was then driven with the pulsed current source and its emission was detected over an $\sim 10^\circ$ angular range about the normal to the waveguide by using a fiber-coupled CCD spectrograph.

Figure 3(b) shows the surface-emitted spectrum as a function of peak drive current. At low current densities, the emission is around 20 nm wide, with two narrow dips in the bands of emission at 557 and 567 nm. From angle-resolved PL measurements (Fig. 4),[16] we identify the surface emission mainly to arise from light initially emitted into the TE_0 and TM_0 waveguide modes, which is subsequently Bragg scattered out of the polymer waveguide by the grating. The emission dip at 557 nm corresponds to the stopband of the corrugated waveguide for the TM_0 mode [at the intersection of the bands in Fig. 4(a)], while the dip at 567 nm corresponds to the stopband for the TE_0 mode [at the intersection of the upper two bands in Fig. 4(b)].

For drive currents below 140 A, the emission spectra in Fig. 3(b) are independent of current, but above 144 A, a narrow peak emerges at 568 nm on the long wavelength edge of the TE_0 mode stopband and grows more rapidly than the surrounding PL. The appearance of this narrow peak, whose linewidth is limited by the resolution of the spectrograph, indicates the onset of lasing in the hybrid device at a wave-



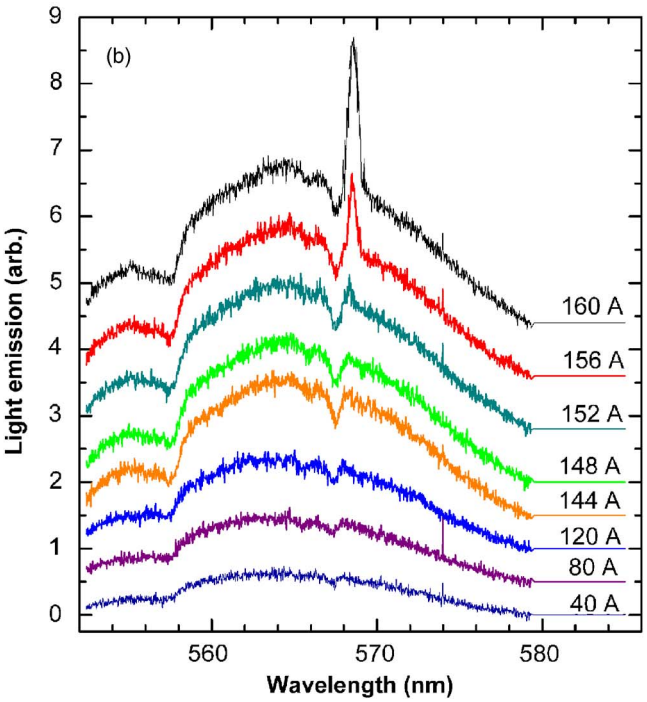


FIG. 3. (Color online) (a) Light emission at 568 and 555 nm as a function of peak drive current. Inset: Schematic diagram of the hybrid InGaN-semiconducting polymer laser. (b) Surface emission spectra from the hybrid electrically pumped polymer laser for different drive currents.

length identical to the one observed when the device is pumped with the OPO. Figure 3(a) shows the surface emission detected at 568 nm (from the TE_0 mode) and at 555 nm (from the TM_0 mode) as a function of drive current. For low drive currents, the emission at both wavelengths linearly increases, but above 140 A (a pump intensity of 217 W cm^{-2}), the emission at 568 nm rapidly increases. This sudden change of slope together with the abrupt narrowing of the spectrum is a clear signature of lasing. In contrast, at 555 nm, neither effect is observed. In other hybrid devices tested, a lasing threshold as low as 180 W/cm^2 was measured.

The integration of polymer lasers with high-power nitride LEDs provides a compact and convenient electrically pumped laser package, which bypasses the considerable challenges in achieving direct electrical pumping of an organic semiconductor laser. Compared to laser pump sources for polymer lasers, the InGaN LEDs are electronically and physically more robust and much lower cost. In our current system, the Luxeon LEDs have not been electronically designed for short-pulsed operation, and with further improvements in the LED design, drive electronics, and polymer laser resonators, we expect considerable improvements in performance to be possible.

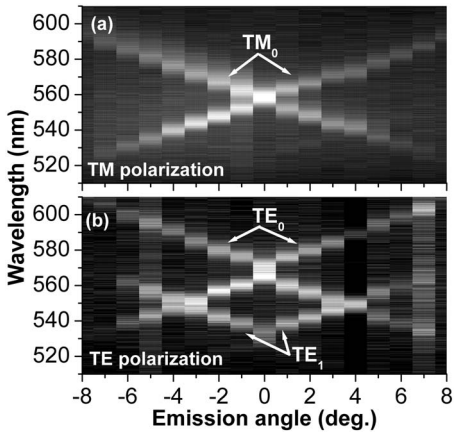


FIG. 4. Angle-resolved PL spectra from the polymer DFB laser polarized (a) parallel and (b) perpendicular to the grating grooves. Observation angles are relative to the normal to the waveguide plane. White bands show strong emission from waveguide modes that have been diffracted out of the polymer film; dark regions correspond to weak emission.

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