LIGHT EMISSION FROM SEMICONDUCTING POLYMERS: LIGHT-EMITTING DIODES, LIGHT-EMITTING ELECTROCHEMICAL CELLS, LASERS AND WHITE LIGHT FOR THE FUTURE

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High performance photonic devices fabricated from conjugated polymers have been demonstrated, including diodes, light emitting diodes, photo-diodes, field-effect transistors, polymer grid triodes, light emitting electro-chemical cells, optocouplers and lasers, i.e. essentially all the categories which characterize the field of photonic devices. In many cases, semi-conducting polymer-based devices have reached performance levels comparable to their inorganic counterparts. This short review focuses on recent progress in the area of light emission from semiconducting polymers. © 1998 Elsevier Science Ltd. All rights reserved

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1. INTRODUCTION

Semiconducting (conjugated) polymers are a novel class of materials that combine the optical and electronic properties of semiconductors with the processing advantages and mechanical properties of polymers [1, 2]. Examples of polymers within this class include poly(p-phenylene vinylene) (PPV) and poly(p-phenylene) (PPP) and their derivatives; a few of the more important molecular structures are shown in Fig. 1.

Because of the sp^2p_z bonding of these planar con-jugated macromolecules, each carbon is covalently bonded to three nearest neighbors (two carbons and a hydrogen); and there is formally one unpaired electron per carbon. In the case of PPV, the repeat unit contains eight carbons; PPV is a semiconductor in which the p_z-band is split into eight sub-bands. The energy gap, the π-π^* gap, is the energy between the highest occupied and the lowest unoccupied molecular orbitals.

When functionalized with flexible side chains, for example, MEH-PPV (see Fig. 1), these materials become soluble in common organic solvents and can be processed from solution into uniform, large area, optical quality thin films. As polymers, they are flexible and easily fabricated into desired shapes. The side chains also serve to tune the energy gap (through steric hindrance) and hence the emission color.

2. POLYMER LIGHT-EMITTING DIODES

There has been significant progress since the discovery of electroluminescence (EL) from semi-conducting polymers [3] and the demonstration of high efficiency EL emission [4]. The emission color can be varied with relative ease by modification of the molecular structure (either main chain or side chain modification). Emission over the entire visible spectrum has been demonstrated with impressive efficiency and brightness. By using a metallic polymer as the hole-injecting contact, robust and mechanically flexible LEDs were demonstrated [5]. Additional advantages include the ability to generate a uniform area of light and the thin profile of the thin film ''sandwich'' architecture [see Fig. 2(a)].

Because of the ease of processing the semiconducting polymer (uniform films can be spin-cast from solution), pixelated column-row addressable displays can be fabricated by simply patterning the electrodes (the active polymer film needs no patterning); a process which is simple and which promises to be cost effective
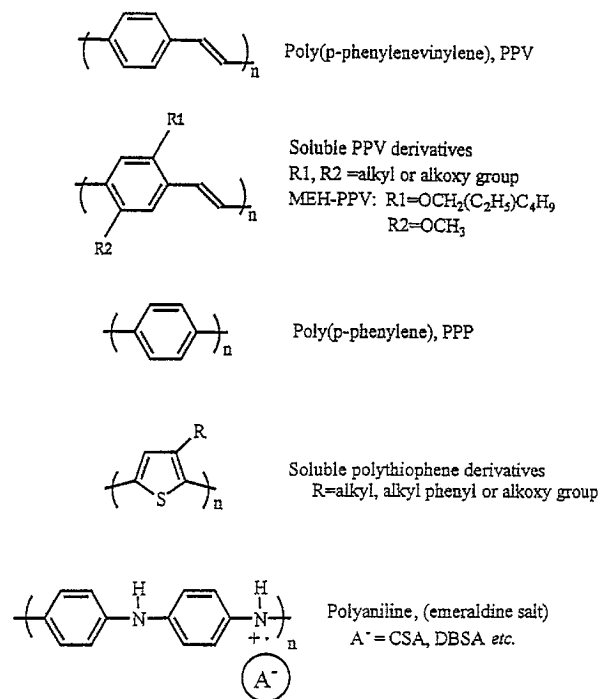
Poly(p-phenylenevinylene), PPV

Soluble PPV derivatives
R1, R2 =alkyl or alkoxy group
MEH-PPV: R1=$OCH_2(C_2H_5)C_4H_9$
R2=OCH_3

Poly(p-phenylene), PPP

Soluble polythiophene derivatives
R=alkyl, alkyl phenyl or alkoxy group

Polyaniline, (emeraldine salt)
A^- = CSA, DBSA etc.

Fig. 1. Molecular structures of conjugated polymers.
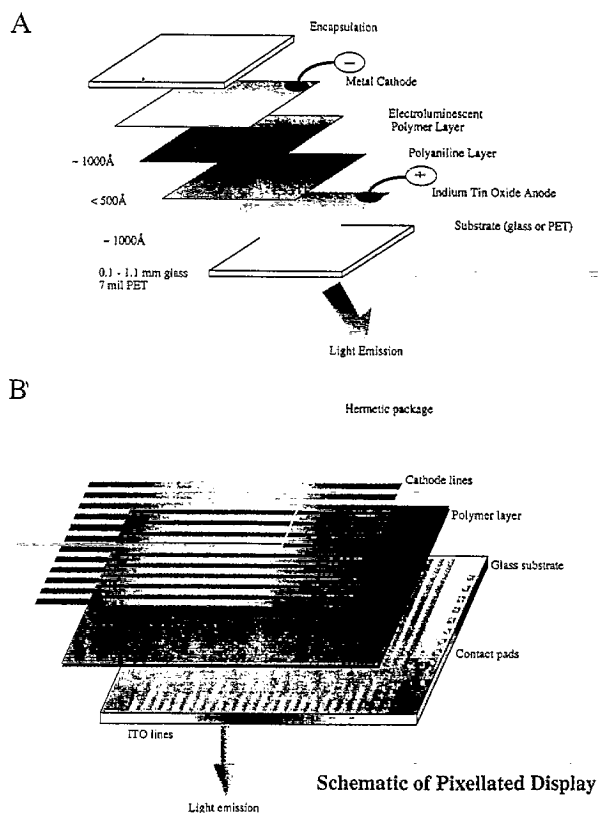
A

B

Schematic of Pixellated Display

Fig. 2. (a) The thin film "sandwich" architecture of polymer light-emitting diodes. (b) The architecture of a passively addressed (column-row) dot-matrix display fabricated with a semiconducting polymer as the light-emitting layer.

compared to the "pick and place" fabrication of inorganic semiconductor emissive displays. A sketch of the architecture of a passively addressed (column-row) dot-matrix display is shown in Fig. 2(b).

The polymer LED is a dual carrier injection device [6]. Electrons are injected from a low work function cathode into the π^*-band of the semiconducting polymer and holes are injected from a high work function electrode into the π-band. The oppositely charged carriers in the two bands meet with the polymer film and recombine radiatively to give off light. Ideally, the work functions of the cathode and anode are perfectly matched to the π^*-band and the π-band, respectively. Generally, however, the match is imperfect leading to barriers for electron and hole injection which occur via a combination of Fowler–Nordheim tunneling and thermionic emission.

To get the light out, one of the electrodes, typically the anode, must be (semi)transparent. Two types of transparent anodes are used to fabricate high performance polymer LEDs: (1) Indium/tin-oxide, ITO, (on glass or on plastic); (2) A bilayer electrode fabricated by spin-casting a layer of a semitransparent conducting polymer (e.g. polyaniline, PANI) onto the ITO-glass substrate. Although pure conducting polymer can be used as the anode, the higher sheet resistance limits the active area; hence the bilayer (e.g. PANI on ITO) is favored. Moreover, the bilayer electrode improves the LED performance, enabling lower operating voltages and longer lifetimes.

The best LED performance is obtained from alkoxy derivatives of PPV. These polymer LEDs turn on below 2 V (d.c.) and reach 100 cd/m^2 at 2.4 V (the brightness of a color TV), 4000 cd/m^2 for $V < 4$ V (the brightness of a fluorescent lamp) and over 10 000 cd/m^2 for $V > 5$ V. The luminance (L) vs voltage and current (I) vs voltage are shown in Fig. 3. The external quantum efficiency of these polymer LEDs has been improved to greater than 2% ph/el with excellent reproducibility and the luminous efficiency is 1.5–1.8 cd/A; values which are competitive with existing display technologies.

The operating life, the time needed for light emission to degrade to half the initial value at constant current, has been a question of concern: Can lifetimes sufficient for commercial display products be achieved with polymers processed from solution? Until recently, there was skepticism that the level of purity required for semiconductor applications could be achieved.

Recent progress at UNIAX has demonstrated that high performance polymer LEDs can be fabricated with long operating life. At 400–500 cd/m^2 initial brightness, room temperature operating lifetimes of several thousand hours are obtained. The extrapolated lifetime at
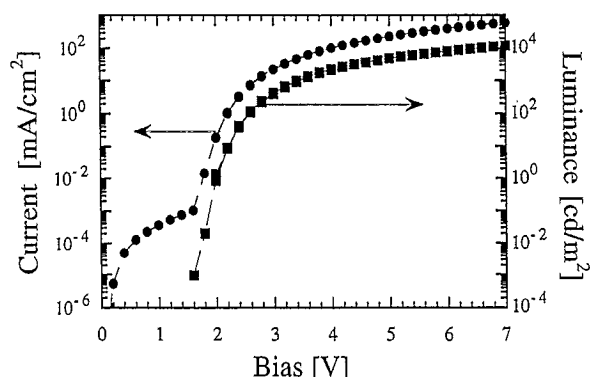
Fig. 3. Luminance (L) vs voltage and current (I) vs voltage for polymer LEDs fabricated from a soluble alkoxy derivative of PPV.
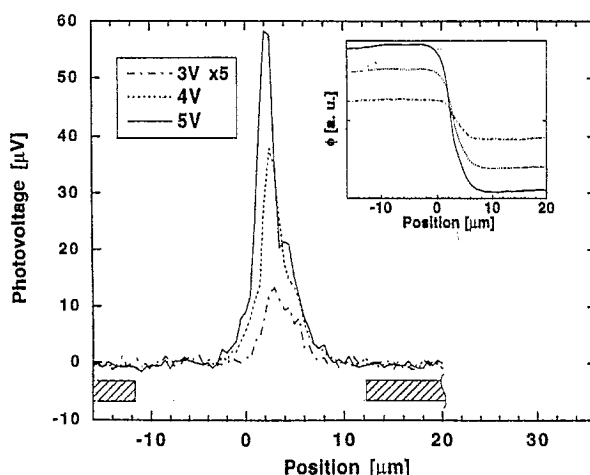
Fig. 4. OBIC scans (photovoltage vs x) showing built-in electric field across the cell. The spatial profile of the built-in potential is shown in the inset.

$100 \, cd/m^2$ is in excess of $10^4 \, h$. Accelerated lifetime studies carried out at 85°C (initial brightness of $100 \, cd/m^2$) indicate in excess of 400 h to half brightness, indicative of more than 200 000 h at room temperature (independent measurements give an acceleration factor of approximately 100 between 85°C and room temperature). Thus, polymer LEDs fabricated with materials processed from solution and spin-cast onto substrates can meet the requirements for commercial products with operating lifetimes in excess of $10^4 \, h$ at display brightness.

3. LIGHT-EMITTING ELECTROCHEMICAL CELLS (LECS)

LECs provide an alternative approach to light emission from semiconducting polymers [7]. Polymer LECs utilize a polymer blend containing semiconducting polymer and solid electrolyte. At voltages above the energy gap, electrochemical redox and the accompanying redistribution of ions from the electrolyte cause p-type doping near the anode and n-type doping near the cathode; a p-i-n junction is created in-situ. Light is emitted from the insulating (i) region between the n- and p-doped layers.

Because of the ohmic contacts between metal electrodes and the p- and n-doped regions, electron and hole injection are balanced. Thus, higher quantum efficiencies are often observed in LECs than in LEDs made with the same semiconducting polymer. Moreover, the same, stable electrodes can be used with any semiconducting polymer. By contrast, recall that for polymer LEDs, the anode and cathode metals must be matched to the π- and π^*-bands, respectively. Thus, for polymer LEDs, different electrodes much be developed to optimize emission from each new polymer (and for each new color!). Red, green and blue (and broadband white) emission from polymer LECs have been demonstrated with external efficiencies of 2–4%.

Since the n-type doped and p-type doped regions in the LEC (under bias) have relatively low resistivity and since symmetric contact electrodes can be used, LECs can be fabricated either in the sandwich configuration [see Fig. 2(a)] or in a planar surface cell configuration with interdigitized electrodes. The latter offers potential manufacturing advantages and enables direct imaging of the junction. Electrochemical doping and junction formation were confirmed using optical beam induced current (OBIC) microscopy to probe the built-in electric field and the built-in potential (Fig. 4) [8].

Polymer LECs have typically been studied in the "dynamic junction" mode where the junction forms and decays with the external bias; in this case the device remains in the neutral form during storage. Recent results have demonstrated that polymer LECs can be operated such that the p-i-n junction is created at an elevated temperature (where ion mobility is high), while at the lower operating temperature (where ion mobility is negligible) the junction is frozen. Using an ion transport polymer with high glass transition temperature, frozen-junction LECs can be operated at or even well above room temperature. Frozen junction LECs can be used for pixelated emissive displays in which high bias voltage, low duty cycle and fast response are required. The freeze-out of ion motion eliminates electrochemical reaction, thus enabling polymer LECs to be operated at high brightness (i.e.. at voltages well outside the electrochemical stability window).

4. CONJUGATED POLYMERS AS LASER MATERIALS

Can "plastic" laser diodes be made from semiconducting polymers? This question would have been

without content little more than a year ago. However, with the recent emergence of semiconducting polymers as laser materials, an active research effort has emerged worldwide in pursuit of this goal [9–11]. Semiconducting polymers offer important advantages as potential laser materials:

- High photoluminescence efficiency;
- Low self-absorption;
- Emission wavelengths that span the visible spectrum;
- Easily processed into optical quality thin films; and
- Electrically pumped in a diode configuration.

The high PL efficiency and large cross-section for stimulated emission persist to high concentrations; in contrast to traditional laser dyes, conjugated polymers function as high gain laser materials in the solid state, neat and undiluted.

Thin semiconducting polymer films with glass ($n = 1.5$) on one side and air ($n = 1.0$) on the other, function as asymmetric planar waveguides. As waveguided light travels through the gain medium, the identity is amplified and grows as $\exp(gl)$, where g is the gain coefficient and l is the distance traveled. When $gl \gg 1$, the linewidth of the spectrum of light emitted from the waveguide is significantly narrowed ("gain narrowed") and centered about the wavelength where the net gain is maximum. Gain narrowing through amplified spontaneous emission (ASE) was observed for more than a dozen different semiconducting polymers when optically pumped in planar waveguides. Typical data for BuEH-PPV are shown in Fig. 5. PL spectra were collected by pumping with 435 nm laser pulses (pulse width ~10 ns, beam size diameter ~1 mm at the sample). Figure 5(a) shows the PL spectra at excitation energies above and below the threshold for gain narrowing. The effect of strong optical pumping is dramatic: the broad PL emission is significantly narrowed with peak centered near the gain maximum of the polymer. The evolution of the linewidth as a function of the excitation intensity is shown in Fig. 5(b), with a well defined threshold for gain narrowing at around 0.2 μJ/pulse.

Gain narrowing of BuEH-PPV and the laser dye DCM suspended in films of polystyrene were directly compared (using DCM concentration optimized for maximum PL efficiency and film thickness with optical density comparable to that of the reference BuEH-PPV film). Although the DCM concentration is more than 10^2 higher than used in typical dye lasers, the threshold for gain narrowing in BuEH-PPV is 10^3 times lower than that of the comparison DCM film [see Fig. 5(b)]. Thus, conjugated polymers emerge as a unique class of laser materials: they provide the intense absorption and
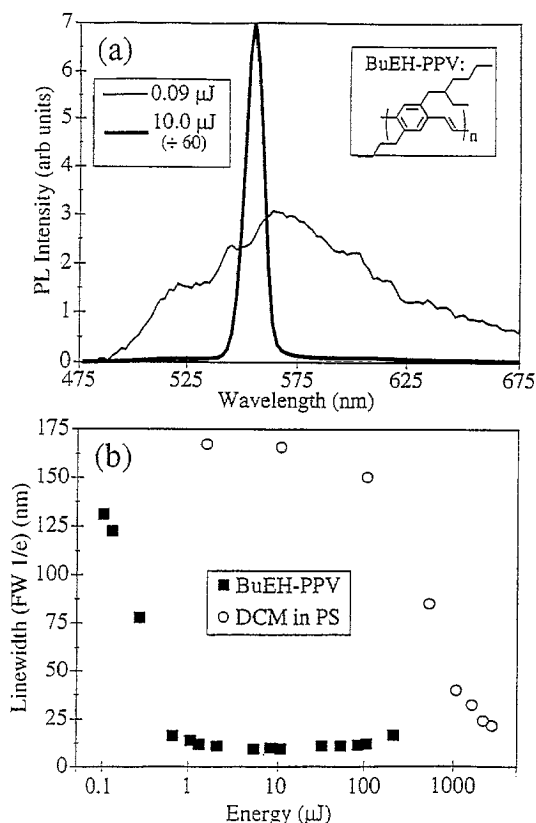

Fig. 5. (a) PL spectra of BuEH-PPV at excitation energies above (10 μJ per pulse) and below (0.09 μJ per pulse) the threshold for gain narrowing. (b) Gain narrowing occurs in a BuEH-PPV film above a threshold of 0.2 μJ/pulse; for comparison, a 2.6% (by weight) of DCM laser dye in polystyrene has a gain-narrowing threshold 1000 times higher.

emission characteristics of laser dyes, but with the substantial advantage of having a much higher density of chromophores in the solid-state. Consequently they offer significantly higher gain and lower threshold energies.

When waveguiding is turned off by various means (e.g. film thickness below cutoff, or use of a substrate with index of refraction greater than that of the polymer), gain narrowing is suppressed [11, 12]. Waveguiding is critical to thin film ASE: the optical path of the emitted light does not exceed the gain length without aid from the waveguide. In such waveguides, there is amplification by stimulated emission, but there is no resonant feedback mechanism for imposing a well-defined optical mode with the corresponding properties of coherence and directionality. To obtain lower thresholds and to generate coherent laser light it is necessary to have a resonant structure. "Plastic" lasers have been made in two forms; thin film microcavity lasers [10, 12] and thin film distributed feedback (DFB) lasers [13, 14].

To fabricate a microcavity laser, a thin film of semiconducting polymer was sandwiched between a highly reflective (>99%) distributed Bragg reflector (DBR) and a thin silver layer [10, 12]. Above the threshold for lasing, the emission peak nearest the gain maximum (~550 nm for BuEH-PPV) [12] grows in intensity more rapidly than the other cavity modes as the pump energy increases. Moreover, a clear increase in the directionality of the lasing mode is observed above threshold. The mode ratio, defined as the ratio of the integrated power of the lasing mode to that of one of the (non-lasing) cavity modes, increases abruptly at threshold and reaches 100 above threshold [12]. Thus, vertically emitting single mode microcavity lasers can be fabricated from semiconducting polymers. Unfortunately, because of losses from the metallic mirror etc., the threshold for lasing in microcavities is not significantly lower than the threshold for gain narrowing through ASE in simple planar waveguides.

An alternative way to create a resonant structure is to incorporate a periodic modulation of the refractive index (a diffraction grating) into the substrate so that light is Bragg reflected. DFB lasers of this type offer stable single mode emission at the Bragg wavelength, $\lambda_{Bragg} = 2n_{eff}\Lambda$, where n_{eff} is the effective refractive index of the waveguide and Λ is the period of the grating. Optically pumped DFB lasers with a conjugated polymer (BuEH-PVV) as the gain medium have recently been demonstrated [13, 14].

DFB lasers were made by spin casting 150–350 nm thick films of BuEH-PPV from xylene solutions onto gratings in 1 μm thick SiO_2 layers that were grown by plasma enhanced chemical vapor deposition (PECVD) on silicon wafers [Fig. 6(a)]. The gratings had periods of 170–185 nm and were made by holographic lithography [15] and reactive ion etching with CHF_3. The grating depths, characterized with atomic force microscopy, ranged from 15–30 nm. A 1 μm thick layer of polymethylglutarimide (PMGI) was spin-cast over the BuEH-PPV to serve as a cladding layer which reduces scattering and protects the BuEH-PPV from exposure to air. After spin-casting the polymer films, the silicon substrates were cleaved to remove the non-uniform edge bead region.

Samples were photopumped with 10 ns pulses of the first and anti-Stokes line (435 nm) from a high pressure H_2 cell which was pumped by 532 nm light from a frequency doubled, 10 Hz, Q-switched Nd : YAG laser; the energy per pulse was controlled with calibrated neutral density filters. An adjustable slit and a cylindrical lens were used to shape the beam into a 200 μm $\times$ 1 mm stripe. The samples were pumped at normal incidence with the long direction of the stripe perpendicular to the grooves in the substrate grating [Fig. 6(a)]. A strong
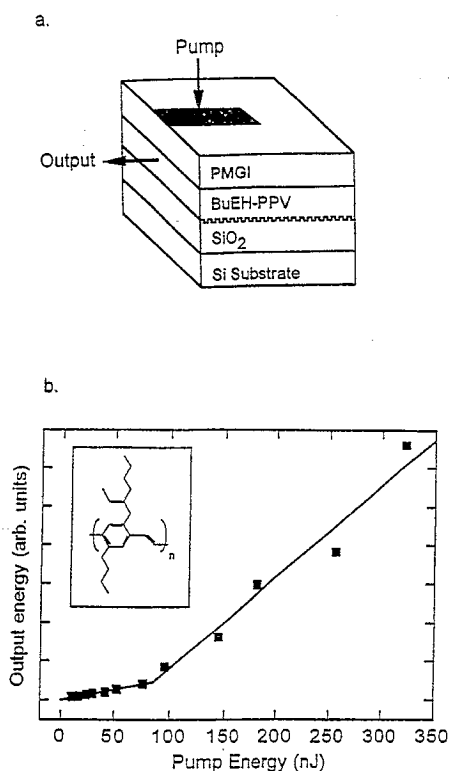

Fig. 6. (a) Schematic of the DFB laser structure and pumping configuration. (b) Total output power at all wavelengths from the edge as a function of pump energy.

beam of green light was emitted from the edge of the sample. Since the light was being emitted from a region having dimensions of approximately 0.2 $\times$ 200 μm, the output beam was highly divergent in the direction of the substrate normal due to diffraction, but only slightly divergent in the substrate plane. The beam was collected by a 10 cm focal length lens, dispersed with a 0.15 m focal length monochromater using either a 300 or 2400 lines/mm grating and detected with a thermoelectfically cooled CCD camera.

The dependence of output energy emitted from the edge vs pump energy clearly exhibits a lasing threshold at 60 nJ/pulse [Fig. 6(b)]. Below threshold the spectrum is quite broad (and arises from spontaneous emission) while above threshold the spectrum is dominated by a very narrow line and is from DFB lasing (Fig. 7). Higher resolution spectra show that the narrow line is actually two lines, each of which has a linewidth of 0.2 nm (Fig. 7).

The observation of two narrow lines is strong evidence for DFB lasing and rules out ASE, which results in peaks with linewidths of 6 nm or more. Further evidence for DFB lasing was obtained by studying a collection of samples with different grating periods (and with no grating). Above a threshold pump energy of 130 nJ/pulse, samples without gratings all exhibited a
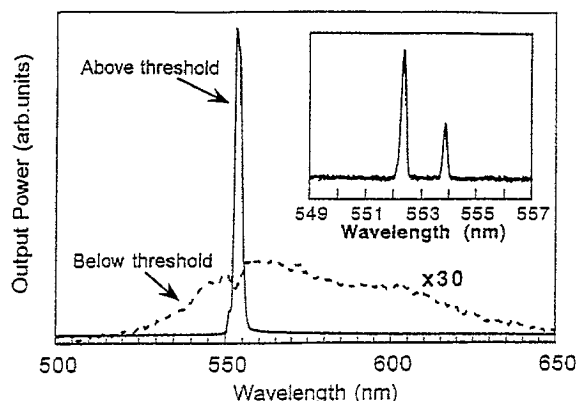
Fig. 7. Spectra taken below (dashed line) and above (solid line) lasing threshold. The below threshold spectrum was multiplied by a factor of 30. The grating period was 170 ± 1 nm, the BuEH-PPV film thickness was 200 ± 200 nm, the calculated neff was 1.62 ± 0.02 and the calculated lBragg was 551 ± 6 nm. Inset: A high resolution spectrum taken above threshold showing two narrow laser emission lines (full width at half maximum of 0 2 nm).

narrow peak centered at 560 nm which was abrupted to ASE. The wavelength of ASE was always 560 nm because the luminescence spectrum peaks near there; 560 nm is the wavelength where the net gain is highest. Samples with gratings all lased in two closely spaced modes with linewidths of 0.1–0.3 nm (FWHM) at the wavelength predicted by Bragg equation. By adjusting the grating period and BuEH-PPV film thickness, the lasing wavelength was systematically varied from 540 to 583 nm, thus proving that the lasing results from the distributed feedback and demonstrating that conjugated polymer lasers can be tuned to lase over a wide range of wavelengths. The threshold for lasing can be lowered substantially by improving the quality of the waveguide to reduce losses (both scattering losses and residual self-absorption).

5. PROSPECTS FOR ELECTRICALLY PUMPED LASERS

The observation of optically pumped lasing in undiluted thin films of conjugated polymers offers the promise of diode lasers with such polymers as gain media. To realize this promise, however, carrier concentrations sufficient to produce lasing must be demonstrated by electrical pumping. Based on the photon densities at threshold with optical pumping and assuming an electroluminescence quantum efficiency of a few percent, the transient current densities necessary to reach the threshold for gain narrowing in planar waveguides are estimated to be a few thousand A/cm^2 (depending on the quantum efficiency for EL). Current

densities of 100 A/cm^2 were reported at the MRS meeting (Boston, 1996) in electrically pulsed polymer LEDs when operated in pulsed mode with low duty cycle; the corresponding peak brightness was 10^6 cd/m^2. Similar results were obtained for PPV. Construction of the polymer diode within a high-Q resonant structure should provide the required reduction in the threshold necessary to achieve electrically pumped lasing.

The features that make semiconducting polymers attractive as laser materials have wide applicability. For example, recent progress in GaN technology has enabled the development of compact, bright and highly efficient blue LEDs and blue laser diodes using InGaN quantum well structures [16]. By combining the high efficiency PL from semiconducting polymers with the emission from InGaN LEDs, hybrid LEDs can be fabricated which emit white light or light of any color; the InGaN LED provides the blue component and, simultaneously, serves as the short wavelength pump source for exciting the PL of the polymer film(s) [17]. White, green, green-yellow and yellow emitting InGaN/ polymer hybrid LED prototypes have been demonstrated simply by dip-coating pre-packaged blue LEDs in the appropriate polymer solutions.

The achievement of compact hybrid InGaN/polymer electrically pumped laser diodes with colors that span the visible spectrum is an attractive possibility. By improving materials and resonant structures and or by using InGaN *laser* diodes (already demonstrated and expected to be commercially available in the near future) [16] electrically pumped hybrid lasers with a full range of colors can be anticipated.

6. CONCLUSION

Since the discovery that semiconducting polymers can be doped over the full range from insulator to metal [18, 19] processable conjugated polymers have been extensively developed. Scientific progress has made available semiconducting and metallic polymers which are both stable and processable. This progress has enabled applications in two principal areas:

(1) Processable conducting polymers and conducting polymer blends with relatively high electrical conductivity [20] (the counter-ion induced processability of polyaniline [21]);

(2) Device applications with semiconducting (undoped) conjugated polymers.

Conducting polymers and conducting polymer blends are targeted for bulk applications (e.g. electrostatic charge dissipation and electromagnetic shielding) and for use as transparent electrodes.

As summarized in this short review, light-emitting devices fabricated with semiconducting polymers have been demonstrated and the performance parameters of these polymer devices have been improved to levels comparable to, or in some cases even better than, their inorganic counterparts. There has also been significant progress in polymer based electronic devices such as polymer field effect transistors [22, 23] and polymer grid triodes [24]. Logic circuits and oscillators have been successfully fabricated from polymer FETs [25]. The "plastic retina", a device that provides local contrast enhancement for image processing [26] is currently under development.

The field of plastic electronics is moving rapidly from fundamental research to industrial R&D with many interesting and novel opportunities. The dream of using semiconducting and metallic polymers for fabricating novel photonic devices, electronic devices and optical devices is becoming reality.

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