

Bragg scattering from periodically microstructured light emitting diodes

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We present a simple method of generating a periodic wavelength scale structure in the optically active layer of a light emitting diode. This is achieved by solution deposition of a light emitting polymer on top of a corrugated substrate. The periodic structure allows waveguide modes normally trapped both in the substrate and in the thin polymer film to be Bragg scattered out of the structure, thus leading to a doubling of efficiency. This scattering process gives rise to a polarization of the emission spectrum as well as angular dispersion effects. © 2000 American Institute of Physics.

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Low-dimensional photonic structures are becoming increasingly important for optical applications, in particular for optical fibers, solid-state semiconductor lasers, and light emitting diodes (LEDs).¹ Periodic structure on the scale of the wavelength of the radiation concerned has been used to control the photonic density of states and leads to intriguing modifications of the interactions of light with matter.² One significant problem with using photonic structures in practical devices lies in the fabrication process of wavelength scale periodicity with sufficient contrast in refractive index,³ as well as the actual physical modulation of the optically active material. The fact that semiconducting polymers are both soft in structure and solution processable makes them ideal for imprinting structures on them, which can be achieved through lithographic means,⁴ or by stamping.⁵ We present here an attractive and effective alternative moulding technique where one-dimensional photonic structure is induced in an optically active conjugated polymer using a high-resolution Bragg diffraction grating as the mold.⁶⁻⁸ The emitting material takes up the form of the microstructure mold when cast onto a corrugated substrate. In particular, the simple processability of conjugated polymers allows us to investigate the effect of periodicity in the emitting layer on electrically driven devices in a LED configuration. This simple photonic structure is shown to be advantageous with respect to a planar device configuration.

The relatively high refractive index of the emitting polymer results in a significant contribution of trapped waveguide modes to the total amount of light generated.⁹ We demonstrate later that the device efficiency can be increased by scattering out these modes from the device by enforcing a periodic modulation on the emitting layer. Periodic modulation of organic thin films has been investigated in a number of examples, most notably in the context of lasing.⁶⁻⁸ Although it has previously been shown that random structures and surface roughening can lead to a significant improvement in light extraction,^{10,11} the advantages of periodical modulation of the emitting layer have only been considered

in device-like configurations which do not comprise injecting contacts.¹² Some work has focused on extracting light from waveguide modes trapped in the substrate of an organic LED.¹³ However, as the refractive index of the emitting layer in an organic LED is generally much greater than that of the glass substrate, we demonstrate later that structuring the emitting layer is far more effective.

The structure of the devices studied is given in Fig. 1(a). Gratings of pitch 388 nm with depths of 10–100 nm were fabricated on a film of photoresist following an established technique¹⁴ using the interference pattern from two laser beams. The corrugation only extends across half of the substrate to allow direct comparison of corrugated and uncorrugated LEDs on one substrate. A thin strip of gold is evaporated onto the substrate, forming the anode and providing the index contrast between substrate and film necessary to achieve scattering. The semiconducting polymer poly [2-methoxy, 5-(2'-ethyl-hexyloxy) 1,4 phenylenevinylene] (MEH-PPV), [Fig. 1(b)] is spin-coated onto the gold. Calcium capped by aluminum is used as the cathode to provide sufficient electron injection. Figure 1(c) shows a photograph of an operating LED with 0.1% external efficiency and a brightness of over 1000 Cd/m², demonstrating that the grating structure allows the fabrication of efficient devices with high yields. There are two features visible in Fig. 1(c), the actual LED pixel marked A and a weaker feature B, which changes color from yellow-green to deep red. Feature B arises from the angular dispersion by the grating of modes scattered out of the *substrate*. Similar features have been observed in devices employing silica spheres to scatter out substrate modes.¹³ Our device, however, also scatters out *film* modes, which gives rise to polarization and angular peak splitting.

Devices were fabricated side by side on a substrate with both a planar and a corrugated (pitch 388 nm and groove amplitude 60 nm) region of photoresist. The electroluminescence (EL) intensity was measured by placing a large area silicon photodiode directly on top of the LED and is, hence, integrated over all emission angles in the forward direction. The current–voltage and EL intensity–voltage characteris-

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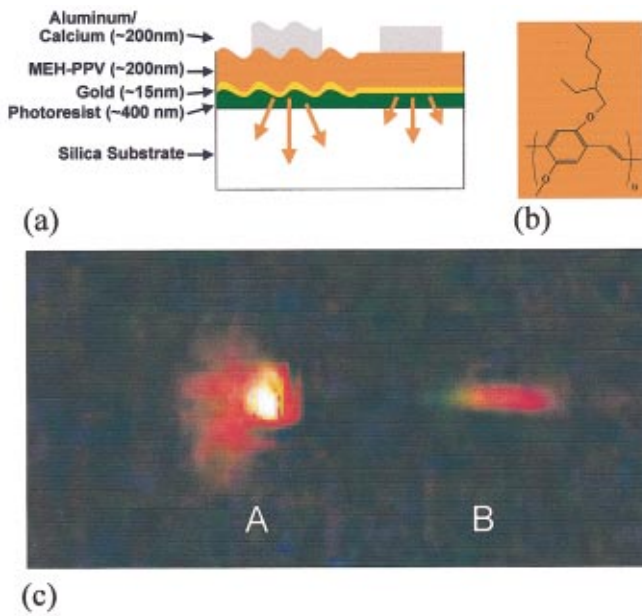


FIG. 1. (Color) (a) A typical structure of a corrugated LED. (b) Chemical structure of the emissive polymer MEH-PPV. (c) Photograph of an operating LED showing the direct emission A and a spectrally resolved faint feature B approx. 5 mm from the pixel. The color of B changes from yellow-green to deep red from left to right, which corresponds to a change in scattering angle of substrate modes.

tics are shown in Fig. 2. It is seen that the current-voltage characteristics of pixels on the corrugated and uncorrugated part of the substrate are very similar. In contrast, the corrugated LED exhibits a stronger luminescence at the same current, as is seen in the inset plot of intensity as a function of current, indicating that more light is escaping from the film. We observe an increase in the external efficiency by a factor of 2.0 ± 0.2 . The fact that the current is unchanged with the corrugation is important evidence that the injection charac-

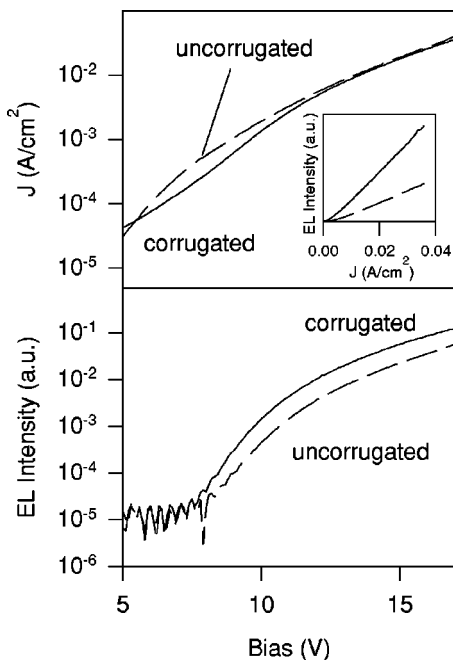


FIG. 2. Current-voltage and EL-voltage of an uncorrugated and corrugated LED. The current is very similar, whereas more light is observed from the corrugated LED. Inset is the EL intensity as a function of current.

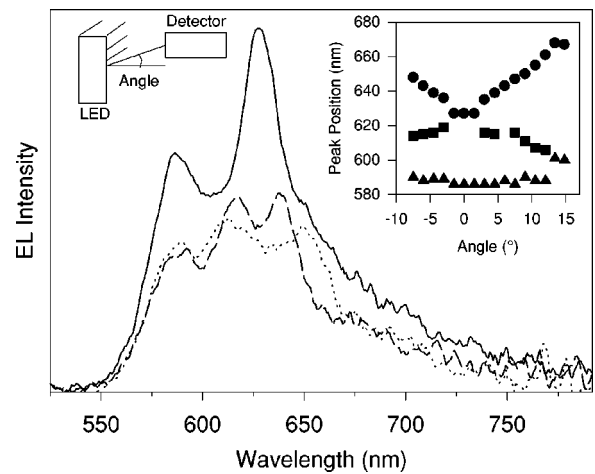


FIG. 3. Angular dependence of the emission from a corrugated LED for normal emission (solid line), emission at -4.5° (dashed line) and $+9.0^\circ$ (dotted line). The left inset shows the experimental alignment relative to the grating. The peak at 625 nm splits into two peaks as the detection moves off normal. The right inset shows the positions of the emission peaks as a function of angle: normal emission at 590 nm (triangles), split part of 625 nm peak moving to shorter wavelengths (squares), and split part of 625 nm peak moving to longer wavelengths (circles).

teristics are not modified by the grating structure. As the electric field lines are orthogonal to the surface of the metal electrodes, the charges migrate a distance through the film corresponding approximately to the film thickness. Shortening or nonuniform field distributions are, hence, not a problem in these devices. Indeed, also the light-output curves of the corrugated and uncorrugated LEDs are identical, but merely shifted in magnitude. This demonstrates that the injection processes of both electrons and holes are unchanged, which is remarkable considering the dimensions of the grating of amplitude 60 nm with respect to the film thickness of 200 nm.

Evidence that the enhancement in emission is due to Bragg scattering comes from the emission spectra. Figure 3 displays the angular dependence of the emission spectrum of a strongly corrugated LED, recorded using a fiber coupled charge coupled device spectrometer in a plane perpendicular to the grating grooves. As the angle at which emission spectra are recorded is increased, the middle peak at 625 nm splits symmetrically into two peaks which move to higher and lower wavelengths, respectively, as seen in the inset. We have observed a similar angular dependence in photoluminescence (PL) spectra of MEH-PPV spun on photoresist gratings¹⁵ and also on gratings covered with a nontransparent metal layer.¹⁶ A future publication will address the origin of the angular dependence, which is due to scattering of modes out of the polymer film and the strong wavelength dependence of the refractive index. Qualitatively, the angular dependence is in agreement with Bragg-scattering theory. The angle of emission, θ , is related to the wave vector of the guided mode, \mathbf{k}_g , and the Bragg vector of the grating, \mathbf{G} , by

$$\mathbf{k}_0 \sin(\theta) = \pm \mathbf{k}_g \pm n \mathbf{G},$$

where n is an integer, and \mathbf{k}_0 is the wave vector of the emitted light. There are, hence, four possibilities to add or subtract the in-plane wave vector and the Bragg vector, which give rise to the cross shape observed in the inset of Fig. 3.

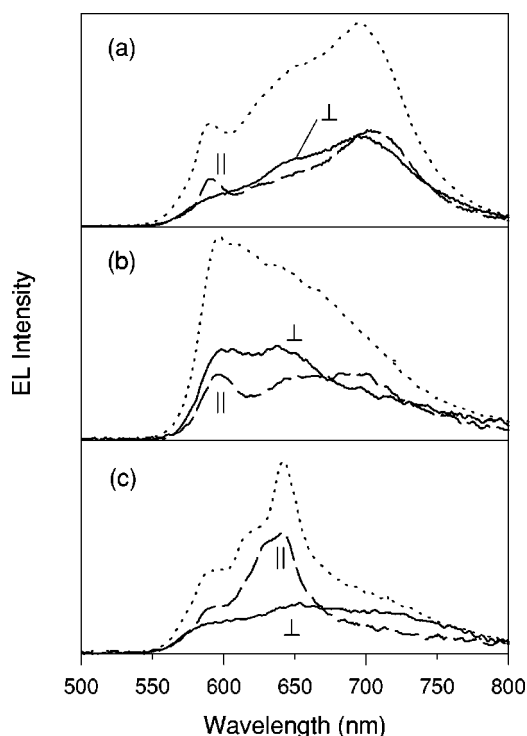


FIG. 4. Emission spectra in the forward direction, unpolarized (dotted line), polarized parallel to the direction of the grating (dashed line), and orthogonal to the direction of the grating (solid line). (a) Thin film (120 nm) on shallow grating (10 nm). (b) Thin film (120 nm) on deep grating (80 nm). (c) Thick film (200 nm) on deep grating (70 nm).

As substrate modes are suppressed in the PL measurement by the metal film between polymer and grating,¹⁶ the peak splitting is attributed unambiguously to scattering of film modes. We have investigated the concern that the angular dispersion may be detrimental to display applications by calculating the color coordinates of the extreme spectra. Due to the broad emission band of the polymer, however, we find such dispersion has a negligible effect on the color coordinates.

It has previously been demonstrated that the emitting dipoles in spin-coated MEH-PPV lie preferentially in the plane of the substrate.¹⁷ Their random distribution in this plane results in unpolarized emission in the forward direction. In contrast, waveguided light in the plane of the substrate is expected to be strongly polarized due to the presence of oriented dipoles. This effect is highlighted in Fig. 4 for LEDs fabricated with different film thicknesses on gratings of various depths. The unpolarized EL is shown together with the polarized EL, which is of lower intensity due to the absorption of the polarization filter. For all devices, the emitted light exhibits a degree of polarization, which increases with increasing film thickness and grating depth. Also, the position of the polarized feature appears to move to longer wavelengths with increasing film thickness. A detailed analysis of this effect is complicated by the presence of two metal electrodes, one of which is semitransparent, resulting in contributions from surface plasmons and interference within the cavity. However, the polarization effect does demonstrate that there is a contribution in the forward emission from scattered light, which gives rise to the observed increase in efficiency. It is also remarkable that scattering fea-

tures are seen with gratings as shallow as 10 nm.

The simple processing of organic materials makes them well suited to being formed as corrugated structures, and we have demonstrated that such structures may provide a significant contribution to increasing the overall device efficiency. In view of applications to backlights and displays, the use of a gold electrode has previously been shown to be preferable as a hole injecting contact due to the higher work function with respect to conventional indium tin oxide.¹⁸ Also, gold electrodes are readily evaporated and are ideally suited for shadow masked electrodes in matrix displays. As up to 80% of the generated light is believed to be trapped in the polymer film,⁹ a suitable structure could be fabricated which extracts most of this light polarized in the plane of the film. Such a device would be ideally suited as a backlight for monochromatic liquid crystal displays, where conventional devices rely on polarization filters to achieve the desired polarization. Although we have implemented the microstructure concept in the context of organic systems, it should also be applicable to a wider selection of materials.

In summary, we have demonstrated that photonic structures can be readily applied to organic LEDs by imposing a Bragg grating structure on a polymer film. We observe a doubling of external efficiency with no effect of the microstructure on the electrical characteristics of the diode. The increase in efficiency is due to the scattering out of waveguided light trapped inside the polymer film.

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