Spectrally narrowed edge emission from organic light-emitting diodes

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Abstract
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Keywords
Ames Laboratory, Glass waveguides, Organic light emitting diodes, Emission spectra, Optical waveguides, Planar waveguides

Disciplines
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Comments

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A dramatic spectrally narrowed edge emission (SNEE) from small molecular organic light-emitting diodes at room temperature, with a full width at half maximum of 5–10 nm, is described. The results show that this emission is due to irregular waveguide modes that leak from the indium tin oxide anode to the glass substrate at a grazing angle. Measurements of variable stripe length devices exhibit an apparent weak optical gain, but there is no observable threshold bias associated with this SNEE. Hence this apparent “optical gain” is suspected to result from misalignment of the propagating leaky waveguide mode and the collecting optics. ©2007 American Institute of Physics.

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Unlike inorganic compound semiconductors, π-conjugated materials are a four-level system,1 rendering them very desirable as a laser medium. Optically pumped organic lasers have inspired intense efforts to achieve lasing via electrical injection in organic light-emitting diodes (OLEDs). This goal, however, has remained elusive. Under optical pumping, a singlet exciton (SE) is directly generated on a single molecule or conjugated segment of a polymer. The main optical losses competing with laser action are due to spontaneous emission, various SE quenching mechanisms, and propagation dissipation in waveguided structures. In electrical excitation, the electron and hole can recombine to either a SE or a triplet exciton (TE). Additional strong losses arise from quenching by the copious polarons, SEs, TEs, and the metal electrodes.2–6 This presents a major obstacle in reaching the lasing threshold.

This letter describes a striking spectrally narrowed edge emission (SNEE) from small molecular OLEDs (SMOLEDs) and measurements on variable stripe length devices that appear to exhibit a weak optical gain. These are two key features of amplified spontaneous emission (ASE). However, no threshold bias for this SNEE is observed. The results demonstrate that it results from irregular leaky waveguide modes propagating in the lateral direction along the OLED/glass substrate interface, which are consequently quite different from the regular waveguide modes that are confined entirely within the OLED layers. The apparent “optical gain” is suspected to result from misalignment of the leaky waveguide modes and the collecting optics.

Various multilayer SMOLEDs were fabricated on indium tin oxide (ITO)-coated glass substrates. The R(Ω) ~ 20 Ω/□, 140 nm thick ITO-coated 2 × 2 in.2 glass substrates were cleaned as described elsewhere7 and UV-ozone treated. The organic layers, CsF buffer layer,8 and Al cathode were deposited using a combinatorial sliding shutter technique described previously.7 Thus, the Al cathode was deposited through a shadow mask containing 21 × 21 1.5 mm diameter circular holes. 1 mm wide stripes, up to 25 mm long, were fabricated by evaporating the Al through an appropriate mask.

The emitting layer was N,N'-diphenyl-N,N'-bis[1-naphthylphenyl]-1,1'-biphenyl-4,4'-diamine (NPD), tris(quinolinolinate) Al (Alq3), or 4,4'-bis(2,2'-diphenylvinyl)-1,1'-biphenyl (DPVBi). Figure 1 shows the surface and edge emission spectra from a (glass)/(ITO)/(10 nm copper phthalocyanine (CuPc))/(x nm NPD)/(30 nm bathocouproine (BCP))/(1 nm CsF)/Al OLED for various x; the non-emissive BCP served as both an electron-transporting and hole-blocking layer. Thus, the recombination zone was in the NPD layer, as confirmed by the surface emission spectra. In devices with a 40 ≤ tNPD ≤ 80 nm thick NPD layer, the edge emission spectrum was rather broad, redshifted relative to that in the normal direction, and nearly purely transverse magnetic (TM). Similar edge emission spectra from similar OLED structures were previously attributed to weak microcavity effects.9 However, in devices with tNPD ≥ 80 nm, a narrow emission band with a full width at half maximum (FWHM) of 7.5 nm emerges at 410 nm, i.e., significantly blueshifted relative to the surface emission, and the broad emission collapses; the narrow band polarization is transverse electric (TE) with a polarization degree > 0.90. Finally,
FIG. 2. (a) Edge emission spectra and (b) surface emission spectra from an OLED with the structure glass/ITO/(5 nm CuPc)/(46 nm NPD)/(x nm DPVBi)/(6 nm Alq3)/(1 nm CsF)/Al, with x = 76, 91, 106, and 121 nm.

the narrow emission band redshifts with increasing NPD layer thickness.

Alq3-based OLEDs exhibited similar behavior above the threshold thickness $t_{\text{Alq3}} \approx 100$ nm.

Figure 2 shows the surface and edge emission spectra of a DPVBi OLED. The vertical microcavity effects induced by the metal cathode are evident from the redshift of the surface peak emission wavelength, from 443 to 470 nm, as the DPVBi thickness increases from 76 to 121 nm. As in the NPD OLEDs, the edge emission is dramatically narrower, with $\sim 6.5$ nm FWHM, and redshifts as well, from 429 to 443 nm. Since the ratio of the broad-to-narrow peak emission wavelengths remains constant, we conclude that the vertical cavity resonance, to some extent, may contribute to the edge emission as well. Moreover, the sharp edge emission peak is TE with a polarization degree as high as 0.94.

The behavior of the SNEE indicates that it is related to a waveguiding effect. Since the refractive indices of the organic ($n \approx 1.7$) and ITO ($n \approx 1.9$) layers are greater than that of the glass ($n \approx 1.5$), the combined organic and ITO layers constitute a dielectric slab waveguide core, with the glass and Al as the waveguide claddings, resulting in a three-slab asymmetric waveguide structure. This structure preferentially supports the TE polarization.

To explore the nature of the SNEE, we begin by noting that the optical propagation loss in a metal-clad waveguide is primarily caused by the strong absorption, estimated at $\sim 10^3$ cm$^{-1}$ of the evanescent tail of the optical field that penetrates the metal electrode. The ITO is also lossy, particularly at $\lambda < 450$ nm. Hence, the waveguided light would be completely absorbed before it contributes to any far-field edge emission (see below). Therefore the SNEE is not due solely to the regular waveguide modes, which are trapped in the organic and ITO layers, and cannot escape unless there is enough optical gain. This conclusion is supported by measurements on patterned OLEDs in which the organic and ITO layers at the edge of the glass substrate were etched off and replaced by a tape, thus blocking the emission from the organic and ITO edges. The resulting SNEE was almost identical to that from OLEDs whose ITO layers remained unpatterned, showing that the SNEE exits from the edge of the glass substrate and cannot be attributed to a regular waveguiding effect.

To clarify any geometric optics effects, a study of modes in the Al/organic/ITO/glass waveguide was conducted. We required that the wave in the ITO not be totally reflected at the organic interface, but that it be totally internally reflected at the ITO/glass interface. This limited the range of propagation angles inside the waveguide. Phase shifts due to propagation and reflections were required to total an integral multiple of $2\pi$ per repeat distance. For the 10–200 nm thicknesses used, many TE and TM modes were found, alternating in vacuum wavelength. Several were within the envelope of the luminescence spectrum of the emitter. Losses occurred at each reflection; they were larger for TM modes, and upon propagation in the ITO when a complex refractive index $n = 1.90 + 0.04i$ was introduced. However, these losses did not preclude propagation distances $d \sim 100$ $\mu$m without optical gain. Yet for $d \gg 1$ mm, some gain was required. However, we could not determine if the SNEE was due to a waveguide mode itself or an evanescent wave in the glass. By decreasing the angle of incidence by 0.6° (1.6°) below the critical ITO/glass interface angle, a “leaky waveguide” mode (so called because it leaks into the glass substrate) was simulated. Its wavelength blueshifted by 16 (28) nm, and it propagated in the glass at grazing angles of 5.3° (8.5°). The transmission coefficients were 0.38 (0.54), leading to observable intensity from the edge of the glass, but significant loss from the waveguide mode.

Pauchard et al. described an irregular mode that is confined at the polymer/SiO2 interface in a multilayer field-effect transistor structure, with a propagation loss of only 9 cm$^{-1}$, thus leading to a considerably reduced optical gain threshold compared to the regular guided mode. Recently, an optically pumped irregular waveguide mode with a narrowed spectrum was identified and characterized through grazing-angle edge emission measurements. Such a mode is generally identified with a leaky waveguide mode, which emerges from the substrate edge nearly parallel to the plane of the organic layer. Its spectral narrowing is believed to be due to the interference between the multiple reflections within the organic layer or between the multiple leaked beams along the organic/glass interface, quite similar to a Fabry-Perot-like microcavity behavior. Indeed, Li et al. observed a low gain threshold for optically pumped grazing-angle edge emission from a three-slab asymmetric waveguide structure, which was attributed to a cavity enhancement effect. More recently, Blinov et al. showed that under certain conditions, leaky-mode lasing has a lower threshold than regular waveguide lasing, and it suppresses the efficient regular waveguide laser action.

The observed SNEE is therefore most likely due to a leaky waveguide mode or perhaps other unknown irregular waveguide modes, propagating along the ITO layer/glass substrate interface. Using a luminance meter, the SNEE was observed to occupy a stripelike bright area at the edge of the glass substrate whose position is consistent with the irregular leaky waveguide modes.

To explore the possible presence of optical gain in the edge emission, we measured the edge emission from stripe-shaped SMOLEDs of variable length $l$. Figure 3(a) shows the normalized edge emission spectra for $l = 12$ mm and (a fixed 1 mm stripe width) at $V = 10$ V and $J = 1$ mA/cm$^2$. As $l$ increases from 1 to 5 mm the TE blueshifts from 472 to 470 nm and the TM mode weakens relative to the TE mode.

Figure 3(b) shows the TE mode intensity versus $l$. We fit the data to $I = (A I_p / g \exp(g l - 1))$ for ASE, where $I_p$ is the threshold intensity.
pump intensity, and $g$ is the net gain coefficient. The excellent match of the data points with the fitting curve is evident; the values of $g$ that resulted from this fitting are 1.58, 1.56, and 1.69 cm$^{-1}$ at 8, 9, and 10 V, respectively. The values of $A_{l_p}$ increase with $V$. Above a certain $l$, the intensity is expected to level off due to a saturation effect. Figure 3(b) exhibits such a saturation clearly for $l > 9$ mm.

Figure 3(c) shows the dependence of the FWHM of the TE mode on $l$. The decreasing FWHM approaches an asymptotic value of $14$ nm at long $l$ ($l > 9$ mm).

ASE theory mandates that the spectra should be broad at short $l$ and become narrower with increasing $l$, and that the output intensity versus $l$ should exhibit supralinear behavior. In contrast, if a mechanism other than ASE dominates, the emission spectrum should not be affected by $l$ and the intensity should increase linearly or sublinearly with $l$. The observed behavior is very intriguing, since it suggests that the SNEE cannot be attributed to interference or waveguiding effects alone. The finite-difference time-domain (FDTD) method was used to solve Maxwell’s equations directly in the time domain for the OLED waveguide structure. We calculated the edge emission spectrum (assuming no optical gain) by setting the distances between the dipole source and the measuring position to be 25, 50, 75, and 100 μm. The FDTD results clearly demonstrate that without optical gain, the behavior is sublinear.

In summary, we have shown that the SNEE from SMOLEDs arises from an irregular leaky waveguide mode that propagates in the organic and ITO layers, until, at some point, it leaks into the glass and then propagates out the edge of the glass at a grazing angle. The nature of the leaky waveguide mode in terms of its propagating pathways and characteristics is still not clear. Although leaky waveguide modes may share some common attributes with the regular waveguide modes, they are different in nature due to different mode field distributions and spatial confinement within the OLED structures. VSL measurements yielded some features of ASE, but no threshold bias or current was observed for the SNEE. We therefore conclude that the apparent weak “optical gain” was probably due to misalignment of the propagating leaky waveguide mode and the collecting optics.

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