Spectrally narrowed edge emission from leaky waveguide modes in organic light-emitting diodes
Zhengqing Gan, Yun Tian, David W. Lynch, Ji-hun Kang, Q-Han Park, and Joseph Shinar

Citation: Journal of Applied Physics 106, 094502 (2009); doi: 10.1063/1.3253582
View online: http://dx.doi.org/10.1063/1.3253582
View Table of Contents: http://scitation.aip.org/content/aip/journal/jap/106/9?ver=pdfcov
Published by the AIP Publishing

Articles you may be interested in
Phosphorescence lifetimes of organic light-emitting diodes from two-component time-dependent density functional theory

Influence of phosphorescent dopants in organic light-emitting diodes with an organic homojunction

Coupling light from an organic light emitting diode (OLED) into a single-mode waveguide: Toward monolithically integrated optical sensors

Spectrally narrowed edge emission from organic light-emitting diodes
Appl. Phys. Lett. 91, 143504 (2007); 10.1063/1.2778358

Highly-efficient broadband waveguide outcoupling in light-emitting diodes with self-organized polymer blends
Spectrally narrowed edge emission from leaky waveguide modes in organic light-emitting diodes

Zhengqing Gan,1 Yun Tian,1 David W. Lynch,1 Ji-hun Kang,2 Q-Han Park,2 and Joseph Shinar1,4,a)
1Department of Physics and Astronomy and Ames Laboratory-USDOE, Iowa State University, Ames, Iowa 50011, USA
2Department of Physics, Korea University, Seoul 136-701, Republic of Korea

(Received 2 March 2009; accepted 24 September 2009; published online 3 November 2009)

A dramatic spectral line narrowing of the edge emission at room temperature from tris(quinolinolinate) Al (Alq3), N,N′-diphenyl-N,N′-bis(1-naphthylphenyl)-1,1′-biphenyl-4,4′-diamine (NPD), 4,4′-bis(2,2′-diphenyl-vinyl)-1′-biphenyl (DPVBi), and some guest-host small molecular organic light-emitting diodes (OLEDs), fabricated on indium tin oxide (ITO)-coated glass, is described. In all but the DPVBi OLEDs, the narrowed emission band emerges above a threshold thickness of the emitting layer, and narrows down to a full width at half maximum of only 5–10 nm. The results demonstrate that this narrowed emission is due to irregular waveguide modes that leak from the ITO to the glass substrate at a grazing angle. While measurements of variable stripe length l devices exhibit an apparent weak optical gain 0≤g≤1.86 cm−1, there is no observable threshold current or bias associated with this spectral narrowing. In addition, in the phosphorescent guest-host OLEDs, there is no decrease in the emission decay time of the narrowed edge relative to the broad surface emission. It is suspected that the apparent weak optical gain is due to misalignment of the axis of the waveguided mode and the axis of the collection lens of the probe. However, it is not clear if such a misalignment can account for all the effects of the observed evolution of the edge-emission spectra with l. © 2009 American Institute of Physics. [doi:10.1063/1.3253582]

I. INTRODUCTION

The demonstration of optically pumped organic lasers1 has 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 spontaneous emission, various SE quenching mechanisms, and propagation dissipation in waveguided structures. Electrical excitation is a different process, where the electron and hole are injected separately into the active region to recombine to either a SE or a triplet exciton (TE). Additional strong losses arise from the presence of absorbing species such as polarons,2,3 and the incorporation of absorbing metal electrodes.4 Moreover, the electroluminescence (EL) quantum efficiency is reduced substantially at high current density due to quenching of SEs by other SEs, by TEs, and by polarons.5,6 This presents a major obstacle in reaching the lasing threshold.

This paper describes striking spectrally narrowed edge emission (SNEE) from small molecular OLEDs (SMOLEDs)7,8 and measurements on these devices with variable stripe length (VSL).9,10 which exhibit an apparent weak optical gain. These are two key features of amplified spontaneous emission (ASE), i.e., mirrorless lasing. However, no threshold bias or current for this spectral narrowing is observed. The results demonstrate that the SNEE 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.

II. EXPERIMENTAL PROCEDURE

Various multilayer SMOLEDs were fabricated on Colorado Concept Coatings (formerly Applied Films Corp.) indium tin oxide (ITO)-coated glass substrates. The R ⊥ ~20 Ω/□, 140 nm-thick ITO coated 2×2 inch2 glass substrates were cleaned by detergent and organic solvents and then treated in a UV/ozone oven to increase the ITO work function and facilitate hole injection, as described elsewhere.11–13 The organic layers, CsF buffer layer,14,15 and Al cathode were deposited in a thermal vacuum evaporation chamber (background pressure <5×10−6 Torr) installed in an Ar-filled glove box. The organic layers typically included a copper phthalocyanine (CuPc) hole injecting layer, an N,N′-diphenyl-N,N′-bis(1-naphthylphenyl)-1,1′-biphenyl-4,4′-diamine (NPD) hole transport layer (HTL), the emitting layer of variable thickness, and a tris(quinolinolinate) Al (Alq3) electron transport layer (ETL) or 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline [bathocuproine (BCP)] ETL and hole-blocking layer. The emitting layer was either Alq3, NPD, 4,4′-bis(2,2′-diphenylvinyl)-1,1′-biphenyl (DPVBi) or fac tris(2-phenylpyridine) iridium [Ir(ppy)3] or Ir(III)bis[4,6-di-fluorophenyl-pyridinato-

---

a)Author to whom correspondence should be addressed. Electronic mail: shinar@ameslab.gov.
$N_{C2'}$]picolinate (Flrpic)-doped 4,4'-bis(9-carbazolyl)-biphenyl (CBP). The Al cathode was deposited through a shadow mask containing 21×21 1.5-mm-diameter circular holes, yielding a 21×21 matrix array of OLED pixels. By using a combinatorial sliding-shutter technique, the thickness of two of the organic layers could be varied systematically across the 21×21 matrix array, as described elsewhere.\textsuperscript{7,11}

For VSL measurements, 1-mm-wide striped OLEDs, up to 20 mm long, were fabricated by evaporating the Al through a mask with appropriate slits. Such measurements are typically conducted to provide evidence for optical gain, and consequent ASE in inorganic semiconductors and optically pumped organic films.\textsuperscript{9,10} To our knowledge, our preceding paper\textsuperscript{8} and this paper describe the first detailed VSL measurements in electrically pumped organic devices. When conducting the VSL measurements, the length of the Al cathode was shortened for each step, by either 1 or 0.5 mm. In other words, the VSL measurements described in this work were destructive. In some cases, however, the ITO was photolithographically etched to yield stripes with lengths varying systematically from 100 to 1400 $\mu$m, i.e., the VSL measurements were carried out on a series of OLEDs.

Figure 1 shows the experimental setup for measuring the edge emission. It included an optical bench and an Ocean Optics Chem 2000 spectrometer. A metal probe installed on the platform was connected to the device cathode; 0–38 V dc was provided by a Kepco DPS-40-2M programmable power supply, and the current was measured using a Keithley 2000 multimeter. The optical fiber of the spectrometer was fixed on an xyz micropositioner and oriented perpendicular to the edge of the device, collecting $\sim$2° of the emitted light around that direction. The position of the fiber tip was adjusted to maximize the EL intensity and minimize its linewidth. For polarization-dependent measurements, a linear polarizer was placed in front of the probe. To measure surface and edge-emission spectra simultaneously, two spectrometers and two computers were used. This ensured that the surface and edge-emission spectra were monitored simultaneously at the same current density, enabling a reliable comparison between them.

To measure the angle dependence of the SNEE, instead of fixing the optical fiber on a micropositioner, it was held in a homemade holder which was fixed on a goniometer that could change the detection angle (see Fig. 2). This total assembly was fixed on the xyz micropositioner, and the OLED was fixed on the edge of a stage. The edge of the device was placed at the center of the goniometer. Hence we could ensure that regardless of the angle $\theta$, the optical fiber was always directed toward the edge of the device.

For optical pumping experiments, the setup was the same, with the exception that a laser beam was guided to the surface of the organic film. The laser intensity was by either the laser power supply or a neutral density filter. The laser beam diameter was $\sim$1 mm and its direction was controlled by a mirror.

The lifetime of the surface and edge EL of phosphorescent OLEDs were also measured. The EL was guided into a photomultiplier tube (PMT) that had a response time <10 ns.

III. RESULTS AND DISCUSSION

A. Alq$_3$ OLEDs

Figure 3 shows the behavior of the (a) edge emission and (b) surface emission from glass/[~100 nm ITO]/[5 nm CuPc]/[45 nm NPD]/[x nm Alq$_3$]/[1 nm CsF]/Al OLEDs, fabricated combinatorially as described above. As clearly seen, the surface emission is the expected broad EL spectrum peaking at $\sim$530 nm. However, for $x$=65 and 85 nm, the edge emission exhibits a narrower emission band at 590 and 600 nm, respectively. Such a narrow transverse magnetic (TM)-polarized edge emission was described by Bulovic et
al. However, they did not describe the edge emission from devices with thicker Alq3 layers. As Fig. 3 shows, a narrow transverse electric (TE) edge-emission band peaking at ~440 nm emerges at $x = 65 \pm 85$ nm. With increasing $x$, this band intensifies and redshifts and the TM band weakens until it is no longer observable at $x = 125$ nm.

**B. DPVBi OLEDs**

Figure 4 shows the edge-emission spectrum from glass/[~100 nm ITO]/[5 nm CuPc]/[45 nm NPD]/[40 nm Alq3]/[1 nm CsF]/Al OLEDs, where $x = 65, 85, 105, 125, \text{and } 145$ nm.

**C. NPD:spiro-DPVBi OLEDs**

Figure 5 shows the edge-emission EL spectra of ITO/[5 nm CuPc]/[40 nm NPD]/[x nm 1:1 NPD:spiro-DPVBi]/[40 nm spiro-DPVBi]/[8 nm Alq3]/[1 nm CsF]/Al OLEDs, for $x = 10, 30, 50, 70, \text{and } 90$ nm. Addition of this blend layer is believed to increase the width of the recombination zone, resulting in a narrower and sharper EL spectrum.

**D. Leaky versus regular waveguide modes**

The alternating appearance and disappearance of the TE and TM modes seen in Fig. 5 is characteristic of waveguide modes. Yet, as previously shown, light that propagates in the organic+ITO waveguide, with the glass and Al cladding, would be subject to losses of the order of 1000 cm$^{-1}$, due primarily to the strong absorption of the evanescent tail of the optical field that penetrates the Al cathode. In addition, the ITO is also lossy, particularly at $\lambda < 450$ nm. Consequently, the guided light would be completely absorbed before it contributes to any far-field edge emission. 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 edge-emission 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 emergence of the waveguide modes from the edges of those layers. The resulting spectral shape, polarization, and intensity were almost identical to those of the SNEE from OLEDs, whose ITO layers remained unpatterned, demonstrating that the SNEE exits from the edge of the glass substrate and cannot be attributed to regular waveguide modes.

**E. Polarization of the edge emission**

The behavior of the SNEE clearly shows that it is related to the waveguide effect. Since the refractive indices of the organic layers $n \sim 1.7$ and the ITO’s $n \sim 1.9$ are greater than that of the glass where $n \sim 1.5$, 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.

Figure 6 demonstrates the polarization of the edge emission. The solid line is the normalized experimental spectrum; the dashed line is the $\cos^2(\theta)$ relation.
where $\theta$ is the polarization angle (the angle between the polarizer and the substrate surface). The overlap of these two curves shows that the edge emission is almost totally TE polarized. It might be argued, however, that such high TE polarization arises because the planar DPVBi and NPD molecules lie preferentially in the plane of the substrate, so more light is emitted with a TE polarization due to the preferential orientation of the SE dipoles in the plane of the substrate. Yet the SNEE from OLEDs based on spiro-DPVBi and Alq$_3$, which are intrinsically nonplanar, was also fully TE polarized. Additionally, in SMOLEDs fabricated by thermal evaporation, where the organic layers are amorphous, the dipoles are most likely randomly oriented.

**F. Photoexcitation measurements**

To better understand the nature of the SNEE, thin films and similar OLEDs structures were optically pumped by an Ar$^+$ laser at 363 nm, with the laser beam perpendicular to the organic layer surface. Because the difference in the refractive indices of most of the organic materials used in this study is marginal, a single layer, with a thickness equal to that of the multilayer OLEDs, should form a slab waveguide configuration similar to that of the OLEDs.

To distinguish between the leaky and regular waveguide modes, we note that the two have a different dependence on the refractive index difference $\Delta n$ at the surface. $\Delta n$ can be changed by placing drops of various liquids between a photoexcited spot of the emitting film and the edge shown in Fig. 7. If the edge emission is due to waveguide modes in the organic and ITO layers, then the change in the cladding layer index will strongly distort the edge-emission spectrum, since reflections on the upper boundary control the phase condition for mode selection. If the edge emission is due to the leaky modes in the glass substrate, then the change in the cladding layer index will have no effect on the mode, because all energy flux has leaked into the substrate.

emission changes significantly, since at the excitation region, most of the energy is still confined in the core layer, and the light still interacts with the upper boundary.

Next, DPVBi films with varying thicknesses were deposited on ITO-coated glass, and their edge emission was compared with those of similar films on which an Al layer was also deposited. Figure 8 shows the edge-emission spectra of these photoexcited DPVBi layers; the spectra shown in Fig. 8(b) are consistent with the SNEE from the OLEDs shown in Secs. III A–III C above. As clearly seen, in the 61 nm layer without Al, the emission is rather broad, but it narrows with increasing $x$, implying that the waveguide core thickness plays a crucial role. In the samples with Al, the edge emission is quite different. The emission from the 61-nm-thick layer is broad and redshifted relative to the spectrum from

\[ I = I_0 \cos^2(\theta), \]  

(1)

**FIG. 6.** (Color online) The edge-emission intensity vs polarization angle in polar coordinates: the solid line is the experimental data, the dashed line is the theoretical data fitting by Eq. (1).

**FIG. 7.** A drop of various liquids is placed between a photoexcited spot of the emitting film and the edge. If the edge emission is due to waveguide modes in the organic and ITO layers (top figure), then the change in the index of the cladding layer will strongly distort the edge-emission spectrum, since reflections on the upper boundary control the phase condition for mode selection. If the edge emission is due to the leaky modes in the glass substrate (bottom figure), the change in the cladding layer index will have no effect on the mode, because all energy flux has leaked into the substrate.

**FIG. 8.** PL Edge-emission spectra of (a) glass/ITO/[x nm DPVBi] and (b) glass/ITO/[x nm DPVBi]/Al, with $x=61$, 76, 91, 106, and 121 nm, photoexcited at 363 nm (see the small peak at that wavelength) by an Ar$^+$ laser.
the layer without Al. This is indicative of the additional absorption induced by the Al layer. Indeed, the Al layer clearly increases the threshold thickness for the SNEE, and it quenches the TM-polarized modes in the spectra of the 106- and 121-nm-thick layers, in agreement with the preferential absorption of TM modes in the metal-clad optical waveguides noted above. In addition, the FWHM of the narrowed spectra with Al is slightly larger than that without Al, providing additional evidence for the absorption by Al.

G. SNEE from OLEDs with a Ag cathode

To confirm the role of absorption by the metal cathode, DPVBi OLEDs with Al cathodes were compared with those with Ag cathodes, as the latter’s absorption coefficient is much smaller. Figure 9 shows that for 20 mm long striped OLEDs, the FWHM of the Al devices is 25.3 nm, while that of the Ag devices is only 9.7 nm. In addition, the TM mode of the Ag devices is stronger than that of the Al cathode, as expected since the TM mode is more strongly affected by the metal cathode than the TE mode.

H. Leaky waveguide mode simulations

To confirm the leaky waveguide mode nature of the SNEE, we now describe their simulation, which assumed that the indices of the refraction of the glass, ITO, and organic layers are \( n_G \sim 1.44 \), \( n_{ITO} \sim 1.90 \), and \( n_O \sim 1.7 \), respectively. To simplify the simulation, the ITO and organic layers were treated as a single layer (I) with \( n_I \sim 1.9 \). Thus, the glass and air are the cladding layers of the waveguide, and the organic+ITO is the core layer.

Figure 10 shows a schematic drawing of the leaky waveguide modes. The light is emitted from the organic film, and reflected or refracted at the film/air (IA) and film/glass (IG) interfaces. The light that is confined in the core layer is subject to the large losses mentioned above, and does not reach the edge. However, the light that is refracted into the glass propagates in the glass with very low losses since the glass’s absorption coefficient is negligible. As a consequence, the constructive and destructive interference among the rays that are refracted into the glass at grazing angles results in the observed SNEE.

The Fresnel equations for the amplitudes of the reflected TE and TM waves \( r^T \) and \( r^M \), respectively, and transmitted TE and TM waves \( t^T \) and \( t^M \), respectively, at the IG interface are

\[

t_{g} = \frac{n_i \cos(\phi_i) - n_g \cos(\phi_g)}{n_i \cos(\phi_i) + n_g \cos(\phi_g)},
\]

\[
	r_{g} = \frac{n_g \cos(\phi_i) - n_i \cos(\phi_g)}{n_g \cos(\phi_i) + n_i \cos(\phi_g)},
\]

\[

t_{g} = \frac{2n_g \cos(\phi_i)}{n_i \cos(\phi_i) + n_g \cos(\phi_g)},
\]

\[
	r_{g} = \frac{2n_i \cos(\phi_i)}{n_i \cos(\phi_i) + n_g \cos(\phi_g)},
\]

\[
\phi_{path} = \frac{2d}{\cos(\phi_i)} \times \frac{2\pi n_i}{\lambda} - 2d \tan(\phi_i) \sin(\phi_i) \frac{2\pi n_g}{\lambda},
\]

where \( \phi_{path} \) is the phase change between wave fronts of a directly refracted ray at the IG interface, and a ray refracted at the IG interface after reflections at the IA and IG interfaces (see Fig. 10).

The total electric field transmission at the IG interface is

\[
\tau = \sum_{m=0}^{\infty} t_{g}^{m} \left[ r_{g}^{m} r_{a}^{m} \exp(i \phi_{path}) \right] = \frac{t_{g}^{m}}{1 - \exp(i \phi_{path}) r_{g}^{m} r_{a}^{m}}.
\]

Therefore, the transmitted intensity from core to glass is

\[
T = \frac{n_g \cos(\phi_i)}{n_i \cos(\phi_i)} |\tau|^2.
\]

Figure 11 compares the simulated spectrum with the SNEE.
spectrum of glass/[150 nm ITO]/[5nm CuPc]/[45 nm NPD]/[76 nm DPVBi]/[6 nm Alq3]/[2 nm CsF]/Al, assuming a grazing angle of 89°, a device length of 1 mm, and a film thickness of 284 nm. As clearly seen, the agreement is very good; the observed SNEE is slightly broader than the simulated spectrum, likely due to absorption and scattering by the Al cathode. In addition, we note that the simulations also yield the redshift of the peak emission with increasing film thickness. We note that the simulated spectrum broadens with device lengths below 50 μm.

I. VSL OLED measurements of optical gain

Since no threshold bias or current for the SNEE could be detected, VSL measurements\(^{9,10}\) of the emission were conducted on various DPVBi OLEDs, where the DPVBi layer was sandwiched between 45 nm NPD as the HTL and 8 nm Alq3 as the ETL, to determine whether the SNEE exhibited any optical gain with increasing stripe length \(l\). The Al stripe, which determined the dimensions of the OLED, was 1 mm wide and initially 11 mm long. It was subsequently cut repeatedly by 0.5 mm, and the SNEE spectrum was measured at a constant voltage after each cut. Figure 12 shows all the spectra, on a normalized (left) and un-normalized (right) scale. As seen, the FWHM increases from 20 nm at 11 mm stripe length, to 122 nm at 1.5 mm stripe length. In addition, the spectrum of long stripes is much more polarized than that of short stripes, since the short-wavelength TM mode intensity increases relative to the long-wavelength TE mode as the stripe length decreases.

Figure 13 shows the peak intensity versus \(l\). The observed agreement with the optical gain equation

\[
I = \frac{A(\lambda)}{g(\lambda)} (e^{g l} - 1),
\]

where \(A\) is a constant related to the spontaneous emission coefficient. Fitting Eq. (5) to the data in Fig. 13 yields a small gain factor \(g = 1.86 \text{ cm}^{-1}\). This small value of \(g\) is expected since in a low mobility device the charge-induced absorption dominates the spectral range of the optical gain profile.\(^4\) In addition, losses due to the metal cathode also reduce the net gain.\(^{21}\)

In comparing the observed VSL results to those expected under ASE, we note that the latter will saturate at long \(l\), because when the coherent photon density increases, the excited state population becomes increasingly exhausted and the intensity ultimately varies linearly with the stripe length.\(^{10}\) Yet Fig. 13 does not exhibit this saturation.

Figure 14 shows the results from another DPVBi device which was 500 μm wide and 9 mm long, and was shortened stepwise. Figure 14(a) shows the normalized edge-emission spectrum for 1 mm = \(l\) = 9 mm. The TM mode weakens relative to the TE mode as the stripe length increases from 1 to 9 mm. Figure 14(b) shows the TE mode intensity versus \(l\). Fitting Eq. (5) to the data results in \(g\) values of 1.105, 1.596, and 1.587 cm\(^{-1}\) at 11, 12, and 13 V, respectively. Figure 14(c) shows the dependence of the FWHM of the TE mode on \(l\). The FWHM decreased from 54.5 nm at a stripe length of 1 mm to 23.4 nm at a stripe length of 9 mm.

Although the saturation effect was not observed in the two OLED batches described above, that was probably due to an insufficient \(l\). Figure 15 shows the TE mode intensity in a third OLED batch versus \(l\) up to \(l\) = 14 mm. A clear saturation phenomenon is exhibited for \(l\) > 9 mm. Fitting Eq. (5) to the data results in \(g\) values of 0.403, 0.4, and 0.41 cm\(^{-1}\) at 8, 9, and 10 V, respectively. Figure 16 shows the dependence of the FWHM on \(l\). As clearly seen, the FWHM ~ 43 nm when the stripe length is 1 mm, and it decreases to 17.5 nm when \(l\) increases to 14 mm.

We also patterned the ITO layer into a staircase-type pattern in 100 μm steps shown in Fig. 17 by photolithography and wet etching. The top Al cathode was deposited through a shadow mask, which produces a rectangular Al film that overlaps with the patterned ITO to define an OLED with a certain stripe length. Figure 18 shows the edge-emission spectra of the DPVBi OLEDs, where \(l\) is varied systematically from 100 to 600 μm. The \(l\)-dependence of these spectra is in good agreement with ASE theory in that the spectra are broad when \(l\) ≤ 400 μm, but the FWHM decreases rapidly to only 6–7 nm at higher \(l\).

J. Angle dependent VSL measurement

We observed the weak gain in the lateral direction. For ASE, the light emitted from the facet should be very directional, as it is from inorganic semiconductor lasers. In a vertical microcavity laser the light is confined in less than 1°; in a conventional diode laser, it radiates an elliptical beam, with
divergence angles of $30^\circ \times 10^\circ$. Hence, these angle dependent VSL measurements were used to check the directionality of the light.

The OLEDs’ structure was glass/[~100 nm ITO]/[5 nm CuPc]/[45 nm NPD]/[50 nm DPVBi]/[7 nm Alq3]/[1 nm CsF]/Al. The detection setup was shown in Fig. 2; the resolution of the goniometer was ~$0.1^\circ$. Figure 19 shows that at $0^\circ$, a weak gain still appears in the superlinear curve. When the detector rotates to $-5^\circ$, the intensity saturates at about 6 mm, and at $l<6$ mm, the curve appears nearly linear. At $-10^\circ$, saturation occurs at shorter $l$. In other words, the detection method and detector position affect the resulting apparent gain.

Figure 20 shows the FWHM of the edge-emission spectrum at detection angles from $25^\circ$ to $60^\circ$. We note that the light emitted at a viewing angle close to $0^\circ$ has a narrowed spectrum; it is attributed either to cavity effects, a leaky waveguide mode, or a cutoff waveguide mode.

K. Finite difference time domain (FDTD) simulations of VSL measurements

To assess the possibility of ASE in the edge emission of the foregoing OLEDs, FDTD simulations of VSL measurements were conducted. ASE theory mandates that the spectra should be broad at short $l$ and become narrower with increasing $l$, and that the intensity versus $l$ should exhibit the superlinear behavior given by Eq. (5). 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 $l$. Notwithstanding the effects of the emission angle demonstrated above, the observed behavior is
very intriguing, since it suggests that the SNEE cannot be attributed to interference or waveguiding effects alone. The FDTD method was used to solve Maxwell’s equations directly in the time domain for the OLED waveguide structure. The geometry used in the FDTD calculations was [glass ($n = 1.52, 0.7$ mm thick)]/[ITO ($n = 1.9, 140$ nm thick)]/[organic layers ($n = 1.8, 180$ nm thick)]/[Al cathode ($40$ nm thick)]. Since the thickness of the glass substrate was $\sim 0.7$ mm, in the simulations we attached a perfectly matched layer below a thin (360 nm) glass to completely absorb any incident light so that the glass is effectively infinitely thick. We used a single TE polarized (E field pointing out of the sheet) dipole source, and pulsed excitation, and collected the time trace of the edge emission, to obtain the emission spectrum by taking the fast Fourier transform of the time trace. The simulation focused on the edge emission spectrum by taking the fast Fourier transform of the electric field pointing from the glass edge near the ITO/glass interface. We calculated the edge-emission spectrum, assuming no optical gain, by setting the distance between the dipole source and the measuring position to be 25, 50, 75, and 100 $\mu$m. Figure 21 clearly demonstrates that without optical gain, the behavior is sublinear. The observed behavior also rules out the possibility of superfluorescence and biexcitonic spontaneous emission. To uncover the nature of the emission spectra, we demonstrated optical gain, some critical points must be considered.\(^2\)\(^6\) When $g$ is low, this method is valid only if (a) the gain and pump intensity are homogeneous over $l$ and (b) the detector collection efficiency $\eta$ is independent of $l$. While condition (a) was likely satisfied in these VSL measurements, it is not clear if condition (b) was satisfied as well. In this work, $\eta$ of the emitted light was limited by the numerical aperture of the optical fiber.\(^2\)\(^6\) It can be argued that when the optical axis of the waveguide is not aligned with respect to the optical axis of the collecting lens, or when the focal plane of the objective is positioned well within the sample, a supralinear curve can be observed in VSL measurements without an actual optical gain. As we saw in Sec. III J, the direction angle of the detector does have an effect on the gain. Moreover, since we cannot determine if the axes were adequately aligned, it is not clear that the strong dependence of the TE and TM modes (see Fig. 12) on $l$ is or is not due to misaligned axes.\(^2\)\(^6\)

L. Issues related to the analysis of VSL measurements

Although VSL measurements are a powerful method to demonstrate optical gain, some critical points must be considered.\(^2\)\(^6\) When $g$ is low, this method is valid only if (a) the gain and pump intensity are homogeneous over $l$ and (b) the detector collection efficiency $\eta$ is independent of $l$. While condition (a) was likely satisfied in these VSL measurements, it is not clear if condition (b) was satisfied as well. In this work, $\eta$ of the emitted light was limited by the numerical aperture of the optical fiber.\(^2\)\(^6\) It can be argued that when the optical axis of the waveguide is not aligned with respect to the optical axis of the collecting lens, or when the focal plane of the objective is positioned well within the sample, a supralinear curve can be observed in VSL measurements without an actual optical gain. As we saw in Sec. III J, the direction angle of the detector does have an effect on the gain. Moreover, since we cannot determine if the axes were adequately aligned, it is not clear that the strong dependence of the TE and TM modes (see Fig. 12) on $l$ is or is not due to misaligned axes.\(^2\)\(^6\)
M. EL decay time $\tau$ measurements of phosphorescent OLEDs

In a laser gain medium, amplification is associated with the population of an excited state, from which stimulated emission can occur. Without the stimulated emission, the lifetime of the upper level population is finite due to spontaneous emission or additional quenching effects. Typically the upper state population decays exponentially with a certain decay time $\tau$, and $\tau$ will decrease significantly if stimulated emission occurs. Thus a decrease in $\tau$ provides strong evidence for ASE.

For fluorescent materials and OLEDs, $\tau \sim 1$ ns, so the stimulated emission decay time will be shorter. Since our temporal resolution was $\geq 20$ ns, we fabricated phosphorescent OLEDs, where $\tau$ is much longer. For example, for PtOEP $\tau \sim 100$ $\mu$s; for Ir(ppy)$_3$ $\tau \sim 0.5$ $\mu$s when doped into 4,4′-bis(9-carbazolyl)biphenyl (CBP); for other phosphorescent dyes, $\tau$ is generally in the $\sim 0.1$ to $\sim 1000$ $\mu$s range.

In light of the foregoing considerations, an Ir(ppy)$_3$ phosphorescent OLEDs were fabricated and studied. Their structure was glass/[150 nm ITO]/[5nm CuPc]/[60 nm NPD]/[90, 110, 130, 150, or 170 nm CBP doped with Ir(ppy)$_3$]/[50 nm BCP]/[1 nm CsF]/Al. These devices were relatively thick, because, as mentioned above, the critical thickness for a device emitting at longer wavelength is thicker than for that emitting at shorter wavelength.

Figure 23(a) shows the surface emission spectra of Ir(ppy)$_3$ OLEDs. The peak positions and the shape of the spectra are almost the same regardless of thickness. The minor changes in the shape result from the weak vertical microcavity effect as discussed above.

Figure 23(b) shows the edge-emission spectra. As clearly seen, their behavior is similar to those from the fluorescent OLEDs. Their spectral narrowing, the alternation of the TE TM modes, and the redshift of the narrow peak are all similar to those of the fluorescent devices.

Figure 23(c) is a magnification of Fig. 23(b). The redshift of the narrowed peak above the critical thickness is clear, and the FWHM of the peaks is $\sim 20$ nm. Although the narrowed emission bands are not as narrow as fluorescent OLEDs, their FWHM is only about half of the surface emission bands.

The phosphorescent OLEDs’ $\tau$ was measured using a PMT. Figure 24(a) shows $\tau$ of the surface emission from 35 to 55 V; it clearly “oscillates” around 1 $\mu$s. Figure 24(b) shows $\tau$ of the edge emission, and it is also $\sim 1$ $\mu$s. In other words, no clear reduction in $\tau$ from the surface to the edge emission, or versus voltage, is observed. However, in the devices with 90, 110, and 150 nm emission layer, $\tau$ slightly increases with increasing voltage, i.e., contrary to the behavior expected with the onset of ASE.

Iridium(III)bis[4,6-di-fluorophenyl - pyridinato - N, C$^2$] picolinate (Flrpic) is another widely used phosphorescent material. Its $\tau$ is $\sim 1$ $\mu$s, and its emission peaks at $\sim 470$ nm, i.e., close to that of the fluorescent material.
DPVBi. We therefore fabricated Firpic-based OLEDs as well, with the structure Glass/[150 nm ITO]/[5nm CuPc]/[35 nm NPD]/[50, 60, 70, and 80 nm Firpic-doped CBP]/[30 nm BCP]/[6 nm Alq3]/[1 nm CsF]/Al.

Figure 25(a) shows the surface emission of the devices. It peaks at 480 nm, i.e., slightly longer than the intrinsic peak of the material; this is due to a weak microcavity effect. Figure 25(b) shows the edge emission of these devices. The critical thickness required for the narrow edge emission is \(\sim 70 \text{ nm}\). The FWHM of the narrow edge emission is \(\sim 15 \text{ nm}\), which is less than half of the surface emission.

Figure 25(c) shows the \(\tau\) of the devices. For all of them, \(\tau\) decreases with increasing voltage, and the values for the edge emission are slightly lower than those for the surface emission. While this behavior is consistent with an ASE contribution, it should not occur in the surface emission. However, since we were not able to measure the \(\tau\) of the surface and edge emission simultaneously (the edge emission was measured after the surface emission), the degradation of the devices may have affected \(\tau\). Thus, a new batch of OLEDs with the same structure was fabricated, and this time the edge emission was measured first. No significant difference was observed between the \(\tau\)’s, and in most of them, the edge emission \(\tau\) was actually slightly longer than that of the surface emission. This confirms that the differences in \(\tau\) are due to the degradation of the devices.

**IV. SUMMARY**

In summary, the SNEE from SMOLEDs was described, and shown to result from leaky waveguide modes that leak into the glass substrate and propagate out of the glass edge at a grazing angle; the threshold thickness for the SNEE was shown to depend on absorption by the metal cathode. VSL measurements of the SNEE yielded some key features of ASE, but no threshold bias or current. Interference and FDTD simulations confirmed the leaky waveguide mode nature of the SNEE, but could not account for the apparent weak optical gain. The possibility that the apparent weak optical gain is due to misalignment between the optical axes of the waveguide and that of the collection lens of the detector was also considered, but it is not clear if it can account for the observed evolution of the SNEE spectra with \(l\).
EL decay times $\tau$ of the surface and edge emission of phosphorescent OLEDs were also measured, and were also consistent with the absence of any significant ASE.

ACKNOWLEDGMENTS

Ames Laboratory is operated by Iowa State University for the U.S. Department of Energy (USDOE) under Contract No. DE-AC 02-07CH11358. The work in Ames was supported by the Director for Energy Research, Office of Basic Energy Sciences, USDOE. The work in Korea was supported in part by q-Psi/KOSEF and the Seoul R&BD Program.