Indication of current-injection lasing from an organic semiconductor

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In this study, we investigate the lasing properties of 4,4′-bis[(N-carbazole)styryl]biphenyl thin films under electrical pumping. The electro-luminescent devices incorporate a mixed-order distributed feedback SiO2 grating into an organic light-emitting diode structure and emit blue lasing. The results provide an indication of lasing by direct injection of current into an organic thin film through selection of a high-gain organic semiconductor showing clear separation of the lasing wavelength from significant triplet and polaron absorption and design of a proper feedback structure with low losses at high current densities. This study represents an important advance toward a future organic laser diode technology.

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with an area of $30 \times 90 \mu m$ [Fig. 1(b)], and organic layers and a metallic cathode were vacuum-deposited on the substrates to complete the devices. We designed the mixed-order DFB gratings to have an alternation of first- and second-order Bragg scattering regions that provide strong lateral optical feedback and efficient vertical outcoupling of the laser emission, respectively. Grating periods ($\Lambda_1$ and $\Lambda_2$) of 140 and 280 nm were chosen for the first- and second-order regions, respectively, based on the Bragg condition, $m \lambda_{Bragg} = 2n_{eff}\Lambda_m$, where $m$ is the order of diffraction, $\lambda_{Bragg}$ is the Bragg wavelength, which was set to the reported maximum-gain wavelength (477 nm) for BSBCz, and $n_{eff}$ is the effective refractive index of the structure, which was calculated to be 1.70. The lengths of the individual first- and second-order DFB grating regions were 1.12 and 1.68 $\mu m$, respectively, in the first set of devices characterized, hereafter referred to as the OSLDs.

The OSLDs fabricated in this work had an inverted OLED structure of ITO (100 nm)/20 wt% Cs:BSBCz (60 nm)/BSBCz (150 nm)/MoO3 (10 nm)/Ag (10 nm)/Al (90 nm) with the energy levels as shown in Fig. 2(a). Doping the BSBCz film with Cs in the region near the cathode improved electron injection into the organic layer, and MoO3 was used as a hole injection layer (Fig. S2, SD). While the most efficient OLEDs generally use multilayer architectures to optimize charge balance, charges can accumulate at organic hetero-interfaces at high current densities, which can be detrimental to device performance and stability. The OSLDs fabricated in this work contained only BSBCz as the organic semiconductor layer and were specifically designed to minimize the number of organic hetero-interfaces. Reference devices without DFB gratings, hereafter referred to as the OSLDs.

![Fig. 1. (Color online) (a) Schematic representation of the OSLDs (SiO2 widths of 140 and 70 nm for second- and first-order gratings, respectively). (b) Laser microscope and (c) SEM images at 5000× and 200 000× (inset) magnification of a DFB grating. (d) Cross-section SEM images of the OSLD. (e) Cross-section EDX images of the OSLD.](image)
to as the OLEDs, were also fabricated to investigate the influence of the gratings on the EL properties.

Figure 2(b) shows optical microscope images of an OSLD and a reference OLED under DC operation at 3.0 V. In addition to the previously described DFB grating, five other DFB grating geometries (Table S1 and Fig. S3, SD) were optimized and characterized in the OSLDs. While EL was emitted homogeneously from the active area of the reference OLEDs, more intense emission could be seen from the second-order DFB grating regions, which were specifically designed to promote vertical light outcoupling, in the OSLDs [Figs. 2(b) and S3 (SD)].

The maximum current densities before device breakdown of the reference OLEDs increased from 6.6 A cm\(^{-2}\) under DC operation to 5.7 kA cm\(^{-2}\) under pulse operation because of reduced Joule heating with pulse operation.\(^{7}\) Under DC operation, all of the devices exhibited maximum \(\eta_{\text{EQE}}\) higher than 2% at low current densities and strong efficiency roll-off at current densities higher than 1 A cm\(^{-2}\), which is presumably due to the thermal degradation of the devices. On the other hand, efficiency roll-off in the OLEDs under pulse operation [Figs. 2(c), 2(d)] began at current densities higher than 110 A cm\(^{-2}\), consistent with a previous report.\(^{7}\) Efficiency roll-off was further suppressed in the OSLDs under pulse operation, and \(\eta_{\text{EQE}}\) was even found to substantially increase above 200 A cm\(^{-2}\) to reach a maximum value of 2.9%. The rapid decrease in \(\eta_{\text{EQE}}\) above the current density of 2.2 kA cm\(^{-2}\) is likely due to the thermal degradation of the device.

While the EL spectra of the OLEDs were similar to the steady-state photoluminescence (PL) spectrum of a neat BSBCz film (Fig. S4, SD) and did not change as a function of the current density, the EL spectra from the glass face of the OSLDs under pulse operation exhibited spectral line narrowing with increasing current density [Fig. 3(a)]. A Bragg dip corresponding to the stopband of the DFB grating was observed at 475 nm for current densities below 650 A cm\(^{-2}\) [Fig. 3(b)]. Lasing occurred at the long-wavelength band edge (480.3 nm) of the Bragg stopband, which can be explained by the difference in gain at the two band edges. As the current density increased above this value, strong spectral line narrowing occurred at 480.3 nm, suggesting the onset of lasing. The intensity of the narrow emission peak was found to increase faster than that of the EL emission background, which could be attributed to the non-linearity associated with stimulated emission.

The output EL intensity height and FWHM of an OSLD are plotted in Fig. 3(c) as a function of the current. While the FWHM of the steady-state PL spectrum of a neat BSBCz film is around 35 nm, the FWHM of the OSLD at high current densities decreased to values lower than 0.2 nm, which is close to the spectral resolution limit of our spectrometer (0.17 nm for a wavelength scan range of 57 nm). The FWHM became saturated above the lasing threshold due to the
limited resolution of our spectrometer. Note that the second apparent transition point in Fig. 3(c) observed around 2 kA cm\(^{-2}\) is presumably due to device instability at high current density. The slope efficiency of the output intensity abruptly changed with increasing current and can be used to determine a threshold of 600 A cm\(^{-2}\) (8.1 mA). Above 3.5 kA cm\(^{-2}\), we observed significant emission of broad EL in addition to the lasing emission. This might have been induced by the partial breakdown of the SiO\(_2\) grating due to the extremely high voltage and current. The output power as a function of the current density is plotted in Fig. 3(d). The maximum output power measured with a power meter placed in front of an OSLD at a distance of 3 cm from the ITO glass substrate [Fig. 3(d)] was 0.50 mW at 3.3 kA cm\(^{-2}\). Here, we note that the slope efficiencies in Figs. 3(c) and 3(d) are appreciably different. Since the fabrication batches were different in these devices, this may have come from a difference in quality of the DFB structures. In a future study, we would like to clarify this fluctuation issue. These observed EL properties strongly suggest that light amplification occurs at high current densities and indicate that electrically driven lasing is achieved above a current density threshold.

Polarization of the emitted light was characterized below and above the threshold to provide a further indication that this was lasing.\(^{28}\) As shown in Fig. S6, the output beam of an OSLD above the threshold was strongly linearly polarized along the grating pattern, which is expected for laser emission from a one-dimensional DFB. Coherence and clear observation of an output beam above the threshold are other central facets of lasing and must be carefully examined to support a claim of lasing. As shown in Figs. S6(b)–S6(c), the projection of the output beam of an OSLD on a screen resulted in a fan-shaped pattern only above the threshold, as expected for a one-dimensional DFB. Regarding the demonstration of the coherence of the output beam, it is necessary to include proof of temporal and spatial coherence. To gain insight into the temporal coherence of the OSLD output, we estimated coherence lengths (\(L\)) from the equation \(L = \lambda_{\text{peak}} / \text{FWHM}\), where \(\lambda_{\text{peak}}\) is the peak wavelength. For all of the devices, \(L\) is 1.1–1.3 mm, which is fully consistent with lasing. For comparison, it is worth noting that the coherence length of a 405 nm gallium nitride diode is around 150 \(\mu\)m.

To characterize the spatial coherence and the spatial profiles of the output beam of the organic laser diode under
electrical pumping, we developed the setup described in the SD [Fig. S7(a)]. One CCD camera was used to characterize the evolution of the beam in the far-field for two different distances between the device and the camera. Another CCD camera was used to examine the near-field pattern of the output beam. Our results about coherence are reported in the SD (Figs. S7–S8). The far-field patterns indicate the existence of a well-defined beam, which was formed only above the lasing threshold. These far-field beam cross sections are consistent with those of an OSL under optical excitation. In addition, zero-order diffracted emission could be observed above the threshold but was not present below the threshold because of the absence of spatial coherence. This can be seen as one proof of spatial coherence. However, the fact that we did not observe other diffracted orders suggests that the emission from our devices contained coherent laser emission and some incoherent EL emission. Looking at the near-field patterns of the output beam, we can see some fringes only above the threshold. This behavior is identical to that we observed in an optically pumped organic laser above the threshold and is another indication of current-injection lasing from OSLDs.

Several phenomena that have been misinterpreted as lasing in the past must be ruled out as the cause of the observed behavior.28 The emission from our OSLDs was detected in the direction normal to the substrate plane and showed clear threshold behavior, so line narrowing arising from the edge emission of waveguide modes without laser amplification can be dismissed.29 ASE can appear similar to lasing, but the FWHM of our OSLDs (~0.2 nm) is much narrower than the typical ASE linewidth of an organic thin film (a few nanometers) and is consistent with the typical FWHM of optically pumped organic DFB lasers (~1 nm).30 A very narrow emission spectrum obtained by inadvertently exciting an atomic transition in ITO has also been mistaken for emission from an organic layer.30 However, the emission peak wavelength of the OSLD in Fig. 3(a) is 480.3 nm and cannot be attributed to emission from ITO, which has atomic spectral lines at 410.3, 451.3, and 468.5 nm.31

If this is lasing from a DFB structure, then the emission of the OSLD should be characteristic of the resonator and the output should be very sensitive to any modifications of the laser cavity. Thus, OSLDs with different DFB geometries, labeled OSLD-1 through OSLD-7 and OSL (Table S1, SD), were fabricated and characterized (Fig. S3, SD). The emission peaks were nearly the same for OSL, OSLD-1, OSLD-2, and OSLD-3 (480.3 nm, 479.6 nm, 480.5 nm, and 478.5 nm, respectively), which had the same DFB grating periods. Furthermore, OSLD-1, OSLD-2, and OSLD-3 all had low minimum FWHMs (0.20 nm, 0.20 nm, and 0.21 nm, respectively) and clear thresholds (1.2 kA cm$^{-2}$, 0.8 kA cm$^{-2}$, and 1.1 kA cm$^{-2}$, respectively). On the other hand, OSLD-4 and OSLD-5, which had different DFB grating periods, exhibited an emission peak at 459.0 nm with an FWHM of 0.25 nm and a threshold of 1.2 kA cm$^{-2}$ (OSLD-4) and at 501.7 nm with an FWHM of 0.38 nm and a threshold of 1.4 kA cm$^{-2}$ (OSLD-5). These results clearly demonstrate that the emission peak wavelength and the threshold are controlled by the DFB resonator architecture. The resonant wavelengths, the Q-factors and the confinement factors were calculated for the different OSLDs (Table S1). The confinement factor was similar for all the geometries. The calculated resonant wavelengths agree well with the experimental lasing wavelengths. OSLD-1, OSLD-3 and OSL showed higher Q-factors than the other OSLDs (Table S1). The calculated Q-factors for OSL and the OSLDs are to some extent consistent with the experimentally estimated Q-factor (Fig. S5). In further work, we will carefully examine the influence of the DFB structure on OSLD performance and expect to see a clear trend in the relationship between the threshold current density and Q-factor.

Overall, as supported by the additional data in Figs. S9–S11, SD, laser emission under current injection was observed in this study due to (1) the clear separation of the lasing wavelength from significant absorption due to charge carriers and triplet excitons; (2) the reduction of optical losses due to electrical contacts because of the DFB architecture; and (3) the low lasing threshold of the BSBCz films allowing us to observe current-injection lasing before device degradation took place.

In conclusion, this study provides an indication that lasing from a current-injection organic semiconductor is possible through the proper design and choice of resonator and organic semiconductor to suppress losses and enhance coupling. The narrow emission demonstrated here has been reproduced in multiple devices, and clear thresholds in both input–output characteristics and strong polarization were observed, thereby excluding other phenomena that could be mistaken for lasing. The low losses in BSBCz are integral to enabling lasing, so the development of strategies to design laser molecules with similar or improved properties is an important next step. This report opens up opportunities in organic photonics and serves as a basis for the future development of an organic semiconductor laser diode technology that is simple, cheap, and tunable and can enable fully and directly integrated organic-based optoelectronic platforms.

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