Coupled surface plasmon-polariton mediated photoluminescence from a top-emitting organic light-emitting structure

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We report strong photoluminescence from a top-emitting organic light-emitting structure where emission takes place through a thin (55 nm) silver film. We show that this emission is mediated via coupled surface plasmon-polariton modes. Our results show that the addition of a dielectric grating to otherwise planar structures, such as surface-emitting organic light-emitting diodes, may offer a way to increase the external efficiency of top-emitting organic light-emitting diodes. © *2004 American Institute of Physics*. [DOI: 10.1063/1.1772516]

Top-emitting organic light-emitting diodes (OLEDs)[1–3] have two advantages over substrate-emitting structures. First, the drive electronics may be integrated into an opaque silicon substrate and second, losses associated with guided modes in the substrate are eliminated. However, a top-emitting OLED does present other problems, notably losses to the surface plasmon-polariton (SPP) modes associated with the metallic cathode. Calculations show that up to 40% of the power that would otherwise be emitted may be lost via this decay channel,[4,5] thus limiting the external quantum efficiency. If some way could be found to recover some of the power lost to these SPPs to light, device efficiency could be improved. One method of coupling SPP modes to light has been the introduction of a periodic microstructure into the metal film, allowing the SPPs to Bragg scatter,[6–8] however, such an approach is demanding in terms of fabrication. Here, we present an alternative in which a microstructured dielectric overlayer is superimposed onto the completed device.

A metallic cathode supports two SPP modes, one associated with each metal surface. For a given frequency, these two modes have different in-plane wave vectors (momenta) and so do not interact. An emitter in the organic layer couples more strongly to the SPP associated with the metal/organic interface than with the SPP associated with the metal/air interface.[9] The energy coupled into the metal/organic SPP thus needs to be transported across the metal if it is to emerge as light. One way is to use grating-mediated SPP cross coupling; an appropriate microstructure enables the momentum mismatch between the SPP modes to be overcome, allowing them to interact providing a route to transport energy across the film.[9,10] However, emission mediated via SPP cross-coupling occurs only over a narrow range of emission angles and wavelengths.[9] An alternative method of coupling the SPPs is to ensure that the materials on either side of the metal have the same refractive index, the SPP wave vectors are then degenerate and may couple.[11] It is this coupled geometry that we employ here, a dielectric overlayer added to the metal acts to match the effective refractive index sampled by SPP fields. The coupled SPPs are then scattered to light by a microstructure imposed on the top surface of the dielectric layer.

Our experimental structure consisted of a silica substrate spin coated with a 160 nm film of a polymer poly(methylmethacrylate) (PMMA) doped with tris(8-hydroxyquinoline)aluminium (Alq_3) (3% Alq_3 by weight). A 55 nm thick silver film was added by thermal evaporation. To construct the corrugated dielectric overlayer (period $\lambda_g = 485$ nm), the silver was spin coated with a photoresist (PR), which was subsequently exposed and developed using standard holographic techniques. A schematic of the structure is shown in the inset in Fig. 1, we refer to it as the experimental structure. Photoluminescence (PL) emission from this structure was compared to the PL from two control samples; both were identical in construction to the experimental structure except the PR layer in the first control was planar while the second had no PMMA film.

To establish the nature of the modes supported by each structure and compare the strength of the emission they produced, measurements of the angle dependent PL were taken from each sample. The Alq_3 was pumped through the silica substrate by a 410 nm diode laser. A narrow aperture limited the collection angle to 1°, and the PL emitted through the

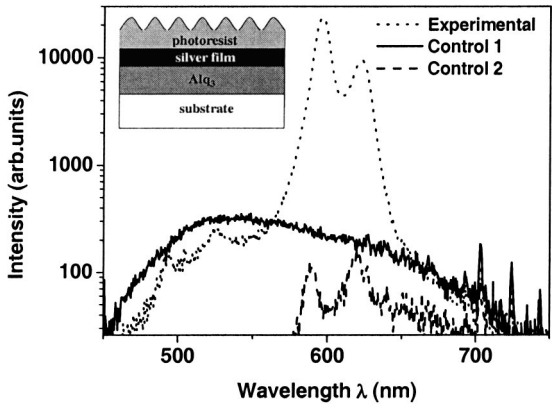

FIG. 1. PL emission spectra collected at a polar emission angle $\theta = 20°$ from the experimental structure and two control structures. Control 1 contains a planar PR layer and control 2 has a corrugated PR layer but no PMMA film.

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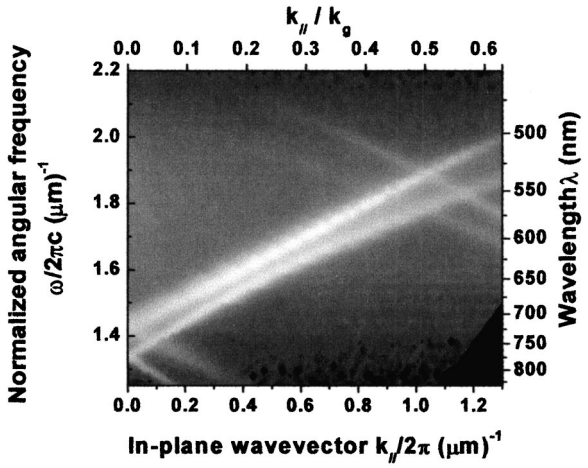

FIG. 2. Dispersion map derived from PL emission data from the experimental structure; emission intensity is shown as a function of frequency and in-plane wave vector, light regions represent areas of strong emission.

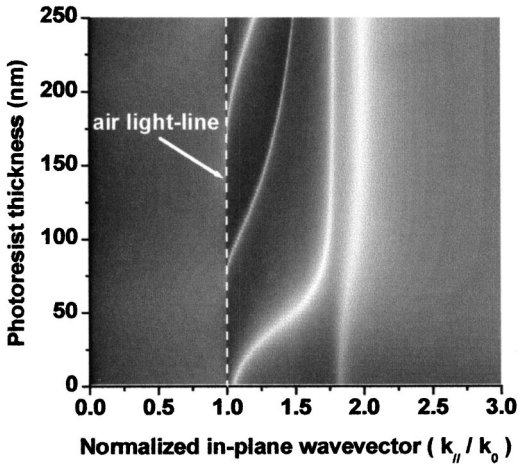

FIG. 3. Theoretically derived dispersion data showing the power dissipated from a dipole source to modes supported by the experimental structure as a function of PR layer thickness and in-plane wave vector. Light regions represent areas of strong emission. The features seen at crossing the air/light line at PR thicknesses of ~75 nm and ~175 nm are the lowest-order guided modes in the PR layer. (For clarity, the source was located in the PR layer; this does not affect the dispersion of the modes but does ensure that the source couples well to all the modes supported by the structure, thus allowing them to be easily seen.)

silver film was recorded using a spectrometer ($\Delta\lambda = 2$ nm) and charge coupled device (CCD). By rotating the sample with respect to the collection optics, emission spectra could be recorded for a range of polar emission angles θ.

Figure 1 shows the PL spectrum from the experimental structure at $\theta = 20°$ and shows two maxima at 595 nm and 620 nm. These peaks dispersed with emission angle, indicative of Bragg scattered modes, and were found to be transverse magnetic (TM) polarized. To identify the nature of these emission features, they were compared to PL from the two control samples (Fig. 1).

The intensity of the emission peaks from the experimental structure is approximately two orders of magnitude greater than those from either control sample. The PL from the control 1 sample matches the emission spectrum from a bare Alq_3 film and the shape did not vary with emission angle. This is expected as modes supported by this structure have no means to couple to light. The PL from the control 2 structure displayed angular dependent emission maxima at wavelengths equal to those seen for the experimental structure. However, these features which arise from the PR emission were weak, the PR film being on the opposite side of the metal to the pump and the pump wavelength (410 nm) being far from the peak absorption wavelength of the PR (~300 nm). The strong emission features in the spectra from the experimental structure (Fig. 1) are thus attributed to the excitation of the Alq_3 layer and not the optical excitation of the PR.

To establish the nature of these emission peaks, spectra obtained for a range of emission angles were built up to form a dispersion map, constructed by converting PL data from intensity as a function of the emission wavelength and angle to angular frequency and in-plane wave vector ($k_{\|}$); (in plane being in the plane of incidence and parallel to the grating vector). Figure 2 shows such a dispersion map, from this a number of PL maximum may be seen including features that cross at a $k_{\|} = k_g/2$ indicating that these features may be attributed to different orders of scattering by the grating structure.

To study how the modes supported by the experimental structure varied with PR thickness, we numerically modeled the modal dispersion by calculating the power dissipated by an emissive source located in one of the layers as a function of PR thickness and in-plane wave vector,[12] Fig. 3. We see that a metal with no PR overlayer supports two SPP modes, at $k_{\|}/k_0 \approx 1.0$ and 1.75. These modes are associated with the metal/air and the metal/organic interfaces, respectively. As the PR thickness increases, the in-plane wave vector of the SPP mode, associated with the metal/air interface, increases rapidly. For a thick metal film, where the fields associated with the two SPPs do not overlap, the in-plane wave vector of this mode would continue to rise with PR thickness toward an asymptotic limit. This limit is greater than the wave vector of the SPP associated with the metal/organic interface since the PR has a higher refractive index (~1.68) than the organic layer (~1.51); for a particular PR thickness the two SPP modes would thus cross. However, the metal in the experimental structure was thin enough (55 nm) that the fields associated with each of the two SPP modes could overlap. Thus, when the effective refractive indices of the media bounding the metal film are matched, the SPP modes may interact. This interaction is seen as an anticrossing between the SPP modes associated with each metal/dielectric interface, for PR thickness between 65 nm and 100 nm (Fig. 3).

To ascertain if the increased emission from our experimental structure (Fig. 1) was due to coupled SPP modes, we had to first establish the thickness of the PR layer in this structure, something we accomplished by looking at the transverse electric (TE) polarized PL. A weak TE polarized feature was also observed but was approximately 100 times weaker than the TM emission features. This TE feature was also seen in the PL from the control 2 structure, indicating that this emission was associated with a guided mode in the PR layer. By comparing this emission with theoretically derived data, we estimate the thickness of the PR layer of the experimental structure to be ~95 nm. This PR thickness falls in the regime where the SPP modes are coupled, confirming that the emission features seen in Figs. 1 and 2 are mediated via coupled SPP modes. These coupled SPPs, in turn, couple to light by scattering from the dielectric grating.

The strength of the emission mediated via the coupled SPP route is remarkable considering the separation, of order 150 nm, between the emissive layer and the PR/air scattering interface. The strength of this emission may be attributed to two factors. First, the metal in the experimental structure is thin enough to allow the fields of the SPP modes associated with each of the metal surfaces to overlap. The dielectric overlayer matches the wave vectors of each of the SPP modes so that they may interact and couple, thus allowing the transfer of energy across the metal. The second factor is the way in which the SPPs outcouple to light. Previous studies have used metal films containing a microstructure to couple the SPP mode to light.[10] In such structures, the SPP mode associated with the metal/organic boundary may couple to light by scattering from either of the metal surfaces. It has been shown that the emission mediated via these routes are out of phase with each other,[9,13] reducing the strength of emission. As the experimental structure contained only one scattering surface, at the PR/air boundary, no such detrimental cancellation occurs.

We have shown that by patterning an appropriate microstructure into a dielectric overlayer, the emission of light through a thin metal film may be increased by some two orders of magnitude over a similar planar structure. The dielectric layer acts to couple the surface plasmon-polariton modes on the two metal surfaces, while its corrugated morphology allows the modes to scatter to light. These results indicate that the addition of a microstructured dielectric overlayer to the cathode of top-emitting OLED, perhaps by soft embossing, should increase light emission from these structures.

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