Observing the Propagation of Fluorescent Light in Doped Polymer Films

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Abstract: By making use of spot excitation to generate photoluminescence, and parallel grooves to extract light, light propagation in a polymer film was quantified. Scatterings contribution in limiting the propagation distance was determined by simulation.
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1. Introduction

Maximizing device efficiency, the product of the internal quantum efficiency (IQE) and light extraction efficiency (LEE), is of crucial importance in developing solid-state lighting [1]. The first is related to material quality and the second is related primarily to geometry. In the past, considerable effort has been expended in developing efficient fluorophores resulting in IQE of devices reaching over 70% for many common dies or light emitting polymers. Recently the focus has shifted to improving the LEE of devices since total internal reflection within a device may trap up to ~75% of the generated luminescence [2,3]. In order to guide efforts in this direction it is necessary to measure light propagation within thin polymer films. We introduce a combined experiment and simulation based approach to address this issue. Spot excitation with a focused laser beam provided a localized source for photons (photoluminescence). Grooves were created within the film to extract the photoluminescence (PL) far from this point source. By varying the separation between excitation and extraction points, the propagation distance of photoluminescence was quantified. Monte Carlo ray tracing was used then to model and thus estimate the mean free distance between scattering events based on the known PL and absorption spectra of the film along with the geometry.

2. Experimental Procedures

Polystyrene was dissolved in a solvent of equal-parts toluene, tetrahydrofuran, and cyclohexanone and doped with 1% poly[2- methoxy-5-(2'-ethylhexyloxy)-p-phenylene vinylene] and spin-casted on a clean glass. The resulting weakly absorbing, weakly fluorescent ~500nm thick film was floated off on a glass surface, transferred to a copper grid and stretched uniaxially to produce localized deformation zones (crazes) perpendicular to the stress direction spaced approximately 20 μm apart in which the film thickness abruptly drops by a factor of three [4].

PL, generated by focusing the output from a cw-Argon laser (λ=488nm) the thin film, was collected and observed using an EMCCD camera. The propagation of the PL generated within the thin film was quantified by varying the separation between the excitation spot and the closest craze edge over a 20 micron range. .

Specially written Monte Carlo based raytracing was used to model the propagation and absorption of the excitation light, generation of PL, and its propagation and emission from the film.

3. Results

Figure 1(left) shows a typical EM-CCD image of the PL emitted from the film under excitation with the focused Argon laser. In addition to the high irradiance in the directly excited region (bright circular spot), emission also occurs crazes (two bright vertical lines), and as a result of scattering centers within the thin film (bright spots). The considerable light intensity emitted from the craze on the left (~16 μm away from the excitation region) indicates that PL emitted at less than the critical angle is trapped within the film and can propagate over a considerable distance.

In order to understand the propagation of PL within the film, the excitation spot was moved across the film (e.g. red line in Fig. 1 (left)) and the PL irradiance at the craze edge obtained as a function of the separation between the center of the excitation source and edge of the craze. Figure 1 (right) shows the results as black dots. The reduction in emission as a function of distance is due to the circular spreading out of PL in two dimensions, self-absorption by the dopant molecules (MEH-PPV) and, to a lesser extent, scattering within the thin film. In other words,

$$I \propto \frac{1}{r} \exp\left(-\frac{r}{r_o}\right)$$

where r is the separation between the excitation spot and the location of emission and r_o is the characteristic decay distance due to absorption and scattering. Fitting the experimental results using this formula (Fig. 1 (right), black line) resulted in a value of r_o = 40 μm for the decay distance. This suggests that, for this film, LEE can be improved by adding scatterers or structural inhomogeneity within the film with a length scale on the order of 40 μm.

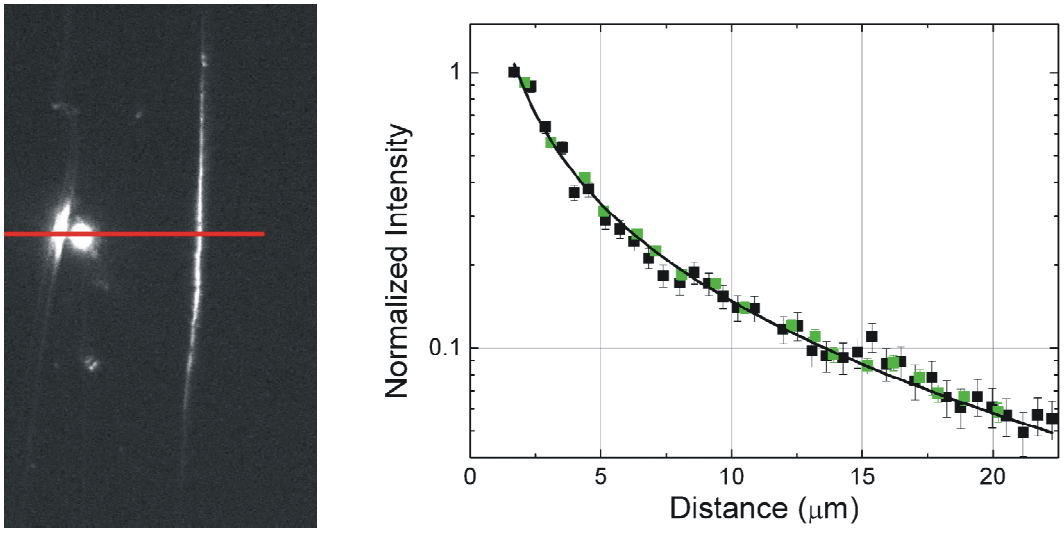

Fig. 1. (left) PL emitted from the film during spot excitation with a Gaussian beam. (right) Intensity of PL emitted from the craze edge as a function of the separation between the excitation spot and craze. Experimental (simulation) results are indicated by black (green) dots. The black line is the result of fitting the experimental results to an exponential-circular decay curve.

As it is not possible to separate the contributions of absorption and scattering to this decay distance experimentally, we iteratively employed Monte Carlo ray tracing simulation with scattering length as the sole free parameter. The green dots in Fig. 1 (right) is the best fit results with a scattering length of 100 μm.

4. Conclusions

The propagation of light within a thin polymer was observed using spot excitation to excite, and crazes to extract, photoluminescence. Ray tracing simulation was used to quantify the amount of light scattering in the thin films. As this methodology relies on relative and not absolute intensities, we believe that this methodology be helpful in observing and characterizing the propagation of light in thin films.

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4. References

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