Exciton-plasmon coupling in nanoimprinted plasmonic crystals for light extraction coupling

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Abstract: Photoluminescence of dye chromophores-loaded in printable polymer has been strongly enhanced thanks to two-dimensional nanoimprinted plasmonic crystals. Comparing the out of plane emission of the spin-coated dye-doped polymer on metallic corrugated and flat surfaces, the PL emission intensity increased by a factor 26 from the area with the nanostructure. This enhancement is explained by a coupling plasmon-exciton in the vicinity of the metal layer and an efficient diffraction of the surface plasmon modes by the metallic structures.

Improving the luminous efficiency, reducing the production cost, increasing the lifetime of OLEDs are the current key issues to be solved in order to achieve widely commercialized OLEDs. Indeed only a small fraction of the total photons generated inside the film are generally usable, because of the total internal reflection and waveguiding effects. Here we investigated the exciton-field coupling of nanoimprinted plasmonic crystals, which exhibit surface plasmon polaritons (SPP), with dye chromophores-loaded. We explored: the direct coupling of the light emitted by a doped polymer to the SPP at the periodic metal layer interfaces, the enhancement of the light extraction achieved by the plasmonic crystals fabricated by nanoimprint lithography (NIL) and the out-coupling to the far field.

To realize nanoimprinted plasmonic crystals, silicon stamps with grating structures were imprinted with a 2.5 inch *Obducat* nanoimprinting tool. The dye-doped polymer was then UV cross-linked during 30 sec followed by the evaporation of 50 nm Au thin film deposited on top cured nanoimprinted polymer layer (Figure 1). An emission spectra comparison of the imprinted unpatterned dye-doped polymer on a quartz substrate without metal and on three quartz substrates coated with 50 nm of Al, Au and Ag, respectively, is presented in Figure 2b. When the plasmon resonance wavelength (controlled by the Ag islands size-Figure 2a) matches the emission wavelength of the dye, the excited electrons in the dye molecules can couple efficiently to the free charges at the metal surface. To better understand the exciton-plasmon coupling phenomena, a model taking in account the molecular excitation and relaxation near plasmonic surfaces was developed. A relatively good agreement has been obtained with simulated and experimental results showing a 9.3 fold enhancement in the PL peak intensity for controlled aggregated size silver metallic films. The rigorous coupled-wave approach has been used then to determine the periodic metallic structures to tune the surface plasmon resonance, to excite the near-field modes and diffract SP polaritons.

To furthermore increase the probability of photon extraction from the SP's energy and to recover the trapped energy, periodic metallic surfaces have been fabricated by NIL to enhance SP–photon extraction (Fig. 1b). Figure 3 presents the photoluminescence intensity of four nanoimprinted structures after metal deposition. An enhancement of a factor 26 have been measured for the structure with a pitch of 900 nm compared to the dye-doped polymer film without metallic layer, thanks to the coupling exciton-plasmons via rough and structured metallic layers patterned by NIL. Similar experiments were done by replacing the dye-doped polymer with an electroluminescent conjugated polymer to use the metallic surface as an electrical contact. Results will be presented. Our results indicate that the combination of surface plasmons and nanoimprinted plasmonic structures in an active layer can lead to a new class of cost effective and high efficiency OLEDs.

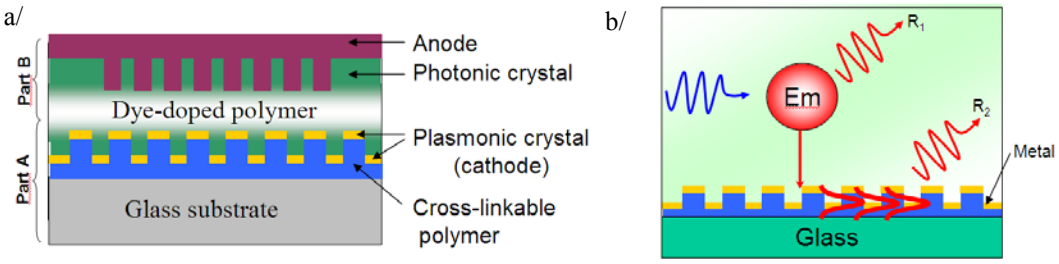

Figure 1: a/ Cross-section schematic of the system. Part A shows the studied system. Part B presents the next fabrication step for potential OLEDs application b/ Schematic of the alternative recombination path (named R_2) to the conventional radiative recombination (named R_1) in the near field of the metal layer in order to enhance light extraction.

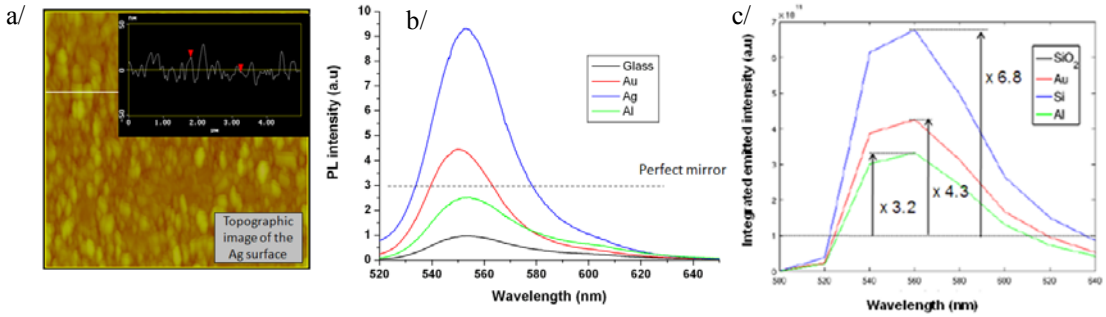

Figure 2: a/ SEM images of a nanoimprinted metallic structure. b/ Photoluminescence spectra of a nanoimprinted unpatterned dye-doped polymer film on a quartz substrate (black line), on a 50 nm Al quartz substrate (green line), on a 50 nm Ag quartz substrate (blue line), on a 50 nm Au quartz substrate (red line), c/ Simulated results of photoluminescence intensity emitted by a dipole (rhodamine 6G) integrated on the thickness of the polymer from three metallic surfaces.

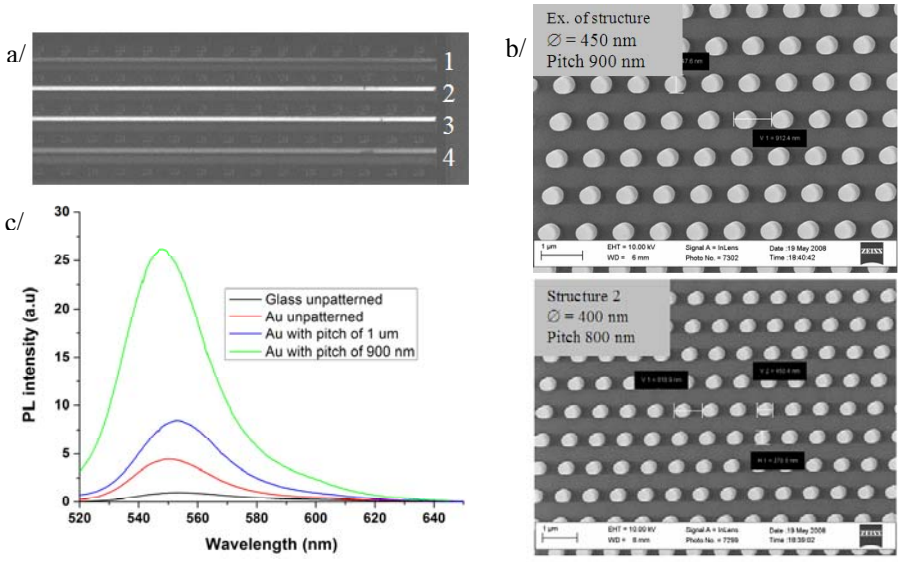

Figure 3: a/ CDD image of imprinted structures through an epifluorescence microscope, excitation between 390-420 nm, emission collection from 450 nm, b/ SEM images of nanoimprinted metallic structures with their associated critical dimension, c/ Intensity of the four structures, c/ Photoluminescence spectra of a dye-doped polymer film on a quartz substrate (black line), on a unpatterned 50 nm Au quartz substrate (redline), on a patterned (pitch of 1 µm) 50 nm Au quartz substrate (blue line), on a patterned (pitch of 900 nm) 50 nm quartz substrate (red line).

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