Losses compensation in metallic nanocomposite polymer by optical gain: application to nanoimprinted microcavities

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A key critical issue in practical applications of nanoplasmonics is the necessity to reduce the absorption losses by the metal nanostructures. One approach is to compensate the loss by gain in the interfacing dielectric in the mixture of nanoparticles and active media. Here, we report a strong enhancement in the spontaneous emission intensity from dye chromophores loaded in a printable polymer by coupling the dye emission to surface plasmons of metallic nanoparticles. The nanocomposite material, embossed by using nanoimprint lithography (NIL) process, shows good imprint properties.

Small amounts of Au nanorods with a double surface plasmon resonance (Figure 1a) were added to a solution of rhodamine 6G (R6G) in a PMMA-based copolymer (PMMA-DMAEMA). The photoluminescence (PL) intensity reported Fig.1b was recorded for different nanoparticle concentrations in the mixture. The measurements indicate an increase in the emission intensity of the dye. This enhancement can be attributed to an increased absorption and emission of R6G in presence of the metallic nanoparticles. In the concentration range of $2x10^{-5}$ to $1.55x10^{-5}$ M of Au nanorods, the number of absorbed R6G molecules per metallic nanoparticle is expected to exceed 1 and the gain of the dye compensates the loss of the localized surface plasmons. At higher concentration of Au nanorods in the functionalized polymer, the losses due to the localized surface plasmons are no longer compensated.

The functionalized polymer was spin-coated on a Si substrate. Figure 2 shows the fluorescence lifetime of the modified polymer with and without nanorods. A reduction of the lifetime from 3.3 ns to 2.8 ns confirmed the modification of the spontaneous emission rate of the rhodamine by the surface plasmons generated by the Au nanorods. Microcavities based on triangular lattice photonic crystal have been fabricated by NIL in the metallic nanocomposite polymer. SEM images of a defect mode nanoimprinted laser in the nanocomposite thin film are shown in Figure 3. The photonic crystal has been designed to present a photonic band gap at the emission peak of the R6G.

In conclusion, we show that the incorporation of gold nanorods in a dye doped thermoplastic polymer adds new optical properties to the material. We demonstrate that the functionalized polymer structures fabricated by NIL exhibit a surface roughness of less than 6 nm rms and that the optical properties of the polymer are hardly affected by the nanoimprint process. The increase of the polymer refractive index by the incorporation of metallic nanoparticles without additional optical loss can lead to new photonic applications in plasmonic. As an example, the metal nanoparticle doped polymer can be used to increase the photonic band gap of nanoimprinted polymer micro-cavity (Figure 3) for “ultra-low” laser applications. This application will be discussed.

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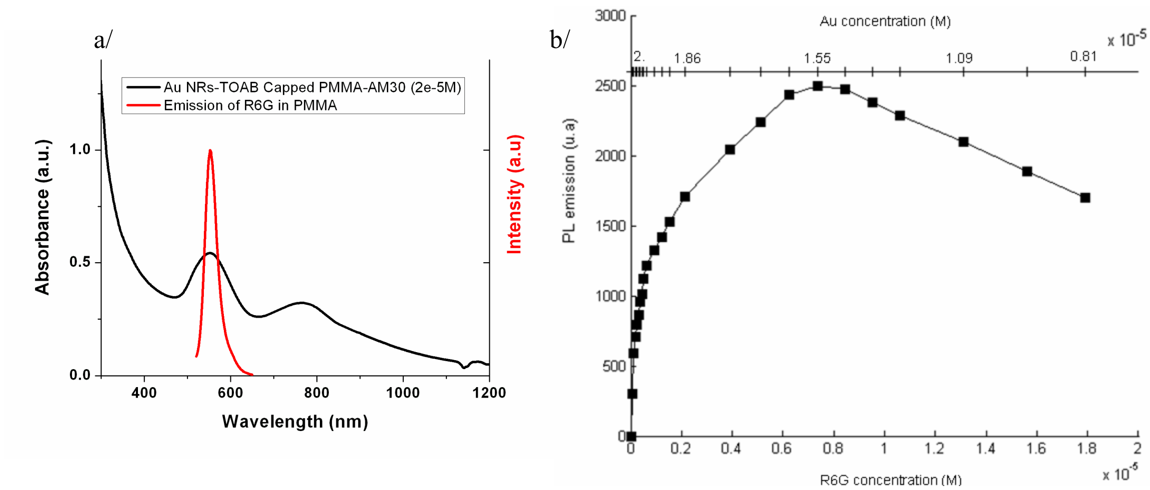

Fig 1. : a/ Absorbance spectra of TOAB capped gold nanorods in PMMA-DMAEMA (concentration: $2x10^{-5}$ M). b/ Photoluminescence intensity of the functionalized polymer versus different rhodamine 6G and the gold nanorods concentrations.

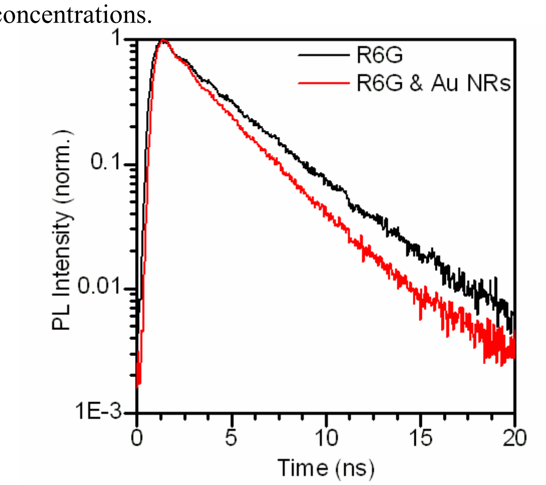

Fig. 2: Fluorescence lifetime of the nanocomposite polymer with and without Au nanorods

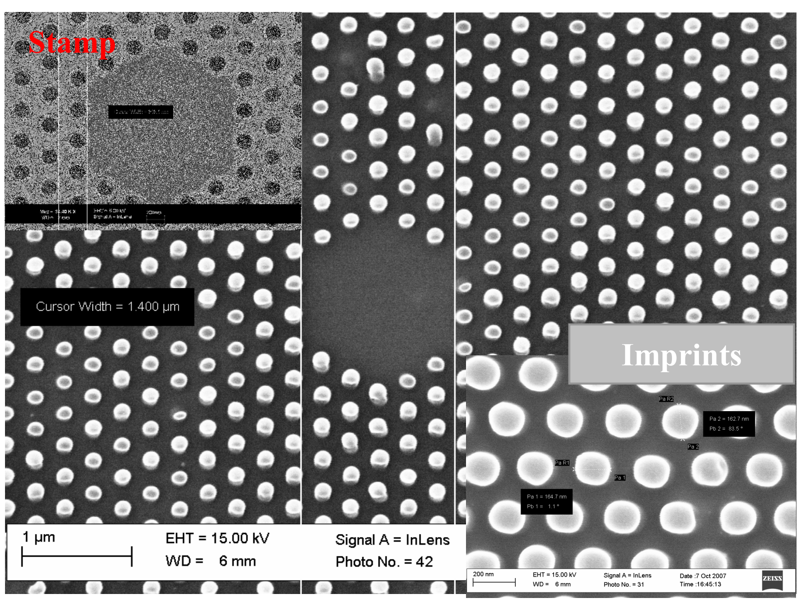

Fig. 3: SEM micrograph of a nanoimprinted micro-cavity for "ultra-low" laser applications imprinted in the metallic nanocomposite polymer