Perovskite Nanocrystals: an Active Material for Integrated Optics

(Student Paper)

Juan Navarro-Arenas, [1] Andrés F. Gualdrón-Reyes,[2-3] Vladimir S. Chirvony, [1] Iván Mora-Seró,[2] Juan Martínez-Pastor[1] and Isaac Suárez [4]

[1] Instituto de Ciencia de Materiales (ICMUV), Universidad de Valencia, C/ Catedrático José Beltrán, 2, 46980 Paterna, Spain.

[2] Institute of Advanced Materials (INAM), University Jaume I, Avenida de Vicent Sos Baynat, s/n, 12006 Castelló de la Plana, Castellón, Spain.

[3] Biofuels Lab-IBEAR, Faculty of Basic Sciences, University of Pamplona, Pamplona, Colombia. C. P. 543050.

[4] Escuela Técnica Superior de Ingeniería, Universidad de Valencia, C/Avenida de la Universidad s/n 46100 Burjassot, Valencia, Spain.

e-mail: juan.navarro-arenas@uv.es

ABSTRACT

Perovskite nanocrystals (PNCs) have demonstrated extraordinary capabilities to construct a new generation of optical sources and amplifiers. In this paper, we firstly analyze the optical gain properties of these materials, and then report our advances towards their integration in integrated optics devices.

Keywords: perovskite nanocrystals, optical amplification, nonlinear optical properties

1. INTRODUCTION

Traditionally, the implementation of integrated optical sources or optical amplifiers has been developed by using III-V semiconductors or glasses and ferroelectric materials doped by rare earth ions [1]. However, there is a cheaper alternative based on (nano)materials synthetized in colloidal solutions. The fabrication by chemical methods provide tailor-made optical properties (light emission, absorption, scattering) that can be engineered during the synthesis. In addition, the solution process nature allows a straightforward incorporation of the materials in optical architectures by coating techniques [2]. In this context, all-inorganic $CsPbX_3$ ($X_3 = Cl_3, Br_3, I_3$) perovskite nanocrystals (PNCs) have recently emerged as an outstanding material for integrated optics [3]. PNCs are synthetized under low cost chemical methods in cubic nanoparticles with sizes closed to the Bohr radius of the bulk material (10 nm) [4]. Their interesting optical properties include a high efficiency of absorption, a quantum yield of emission exceeding 90 % at room temperature, or a tunable band gap depending on chemical composition [4]. Consequently, during the last 3-4 years PNCs have been extensively studied as an active material, with demonstrated applications in light emitting diodes, lasers or optical amplifiers [5].

Here, we firstly analyze the formation of optical gain in $CsPbBr_3$ PNCs films and the conditions to minimize the threshold of stimulated emission. Then, PNCs films are integrated in a waveguide structure that efficiently exploits their promising light emitting properties. As a result, our waveguides demonstrate an enhancement factor of 3 for the emitted light that paves the road for future active integrated devices based on PNCs.

2. RESULTS

PNCs with three different bandgaps (see the PL spectra in Figure 1) were synthetized following the procedures explained elsewhere [4]. Then, PNCs where deposited on the appropriate substrate by a layer-by-layer technique based on a Doctor Blade applicator. This method provides the fabrication of smooth layers with an accurate thickness control (from ~10 nm to ~1 μm). First of all, the formation of ASE in these films was analyzed by pumping the active material with a pulsed (1 ns, 1 kHz) Nd:Yad laser tripled to 355 nm at cryogenic temperature in order to prevent the influence of nonradiative deactivation channels. In this way, ASE is demonstrated at 528, 600 and 705 nm by using PNC films with different halide compositions (X_3=Br_3, X_3=$Br_{1.5}I_{1.5}$, and X_3=I_3), as presented in Figure 1. Clearly, above a certain threshold the PL spectra collapse to a narrow peak (PL spectra in Figure 1) and the PL intensity grows superlinearly with the excitation fluency (insets in Figure 1), which are both signatures of optical gain. Among the possible physical mechanisms responsible for ASE (single excitions, biexcitons or free carriers) our characterization reveals that the optical gain in these materials is formed by the formation of single excitons (see a detailed explanation elsewhere [6]). In these conditions, inverted population can be reached under small thresholds (as low as 5 μJ/cm^2) only limited by the self absorption losses [6].

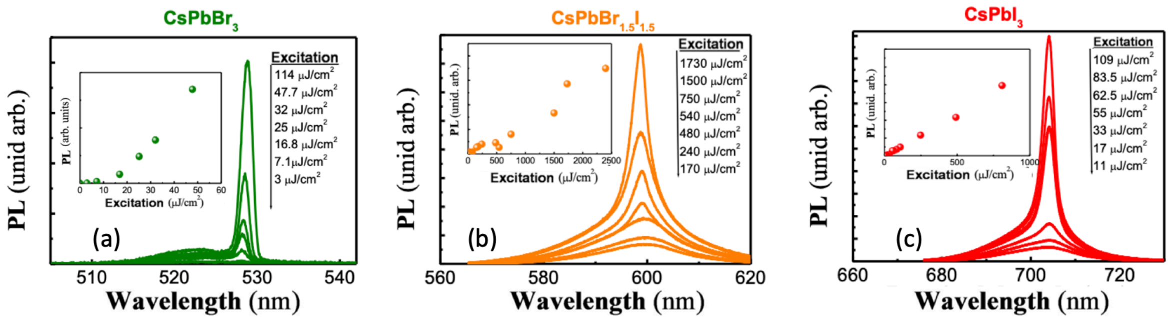

Figure 1. PL spectra of $CsPbX_3$ films at cryogenic temperatures demonstrates the generation of ASE above a certain threshold. (a) $CsPbBr_3$ (b) $CsPb(BrI)_3$ (c)$CsPbI_3$

Once the optical gain is demonstrated, PNC films are integrated in a waveguide which efficiently enhances the light. In particular, a 100 nm film of PNCs is sandwiched between two 0.5 μm thick layers of PMMA and deposited on a commercial SiO_2(2 μm)/Si substrate (see Figure 2(a)). The high refractive index contrast between the different materials results in the propagation of a fundamental TE_0 (TM_0) mode highly confined in the active layer. This strong overlap between the propagating mode and the active film enhances the reabsorption and reemission processes responsible for stimulated emission. In addition, the top and bottom claddings reduce the losses of the light travelling along the waveguide, hence allows the propagation of the light along the whole length of the structure [7]. Indeed, we showed an optical amplification in a similar configuration containing an organic compound as an active material [8]. Here, we evaluate the generation of photons emitted by the reabsoption of the light as the basis of optical amplification mechanism. For this purpose, the sample is illuminated with a stripe line at 450 nm. Then, PL collected at the output edge of the structure is analyzed as a function of the separation of the stripe and the edge of the sample (see Figure 2(b)). At these conditions, PL spectra red-shift for long separations (Figure 2(c)) due to the influence of the photons reemitted along the propagation path. Indeed, we evaluate by the Monte Carlo simulations that the generation of these secondary photons enhances the light emitted by the structure by a factor 3, indicating that this is a promising configuration for optical amplification. Future experiments will include the demonstration of optical amplification by stimulated emission or other nonlinear optical processes that can be enhanced by this configuration. For example, we recently demonstrated that a hollow-core optical fiber filled by $CsPbBr_3$ nanocrystals shows an amplification of 3 dB explained by the nonlinear properties of the nanocrystals [9].

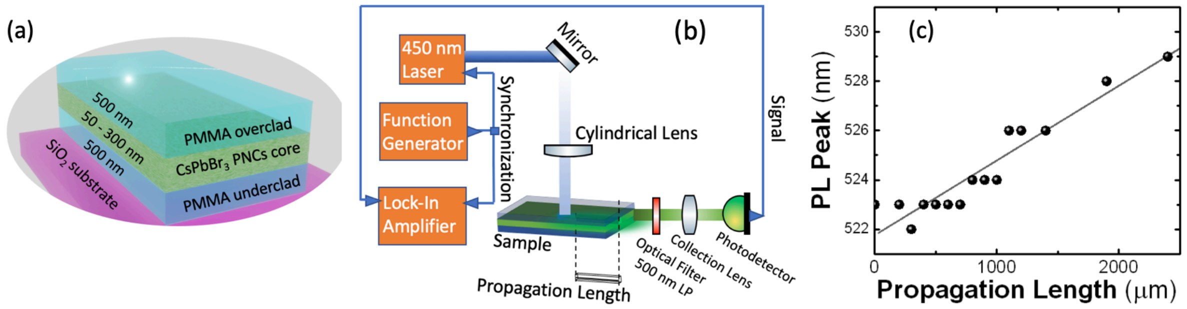

Figure 2. (a) Waveguide structure. (b) Set-up (c) Red shift with the propagation length

3. CONCLUSIONS

In this work the optical gain of perovskite nanocrystal films is thoroughly analyzed. Since these nanoparticles present a low threshold (5 μJ/cm^2 for the best film) of stimulated emission together with a straightforward integration in optical architectures, we believe that they are promising candidates for future integrated sources or amplifiers. In particular, we propose a waveguide configuration that enhances the absorption and generation of light by a high confinement of the propagating mode within the active layer. As a result, we demonstrate that the photons reemitted in the layer enhance the signal by a factor of 3 indicating that this is a promising approach for future active devices.

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