# Lead-free FASnI<sub>3</sub> laser amplifiers integrated in flexible waveguides

Isaac Suárez, <sup>1</sup>\* Vladimir S. Chirvony, <sup>2</sup> Jesús Sánchez-Díaz, <sup>3</sup>Rafael S. Sánchez, <sup>3</sup> Iván Mora-Seró, <sup>3</sup>\* and Juan P. Martínez-Pastor <sup>2</sup>\*

1 Escuela Técnica Superior de Ingeniería, Universidad de Valencia, Valencia 46100, Spain 2 UMDO, Instituto de Ciencia de los Materiales, Universidad de Valencia, Valencia 46980, Spain 3 Institute of Advanced Materials (INAM), Universitat Jaume I, Castelló de la Plana, Castelló 12006, Spain \*e-mail: isaac.suarez@uv.es, sero@uji.es, juan.mtnez.pastor@uv.es

**Abstract:** A FASnI<sub>3</sub> lead-free perovskite is integrated in a flexible waveguide to demonstrate Amplified Spontaneous Emission. An extremely low threshold, 1  $\mu$ J/cm<sup>2</sup>, is observed together with the formation of narrow random lasing lines (< 1 nm).

### 1. Introduction

The integration of optical functionalities in a flexible substrate has become in an important trend in the optoelectronics community [1]. However, the sophisticated technology to fabricate suitable optical architectures in a flexible substrate limits its demonstration to extremely few reports, which becomes even more challenging for active devices, where the appropriate integration of a material with high efficiency of emission is also necessary. In this work, a  $FASnI_3$  (FA, formamidinium) lead-free halide perovskite (LFP) thin films are integrated in a flexible polyethylene terephthalate (PET) substrate and cladded by a polymethylmethacrylate(PMMA) thin film [2]. The structure conforms a planar waveguide where the geometrical parameters (i.e. thicknesses of the films) are properly chosen to: (i) allow the single mode propagation at the photoluminescence (PL) wavelength, (ii) provide an optimum excitation of the FASnI<sub>3</sub> by end-fire coupling the pump beam, (iii) enhance the light-matter interaction in the semiconductor and with it the optical gain, (iv) provide preferable direction for the emitted light and a direct outcoupling [3]. As a result, amplified spontaneous emission is demonstrated with an extremely low threshold, about  $1 \mu J/cm^2$ , and a strong polarization anisotropy preferable to the transverse electric (TE) polarization. Moreover, the device exhibits narrow lasing lines (< 1 nm) caused by the formation of random cavity loops in the polycrystalline films consisting of grains [4]. The operation of the device is analyzed under bending conditions demonstrating the figures of merit can be tuned with the curvature radius. The proposed device represents an important step towards the development of future cheap and green flexible/wearable technology.



Fig. 1. (a) Structure of the waveguide. (b) PL below (black) and above (red) threshold.

## 2. Waveguide design and fabrication

High-quality FASnI<sub>3</sub> films where integrated in the waveguide architecture shown in Fig. 1a following the procedure developed elsewhere [5]. In particular, LFP thin film was deposited on a flexible PET substrate and covered by a cladding of PMMA with two purposes: (i) the PMMA acts as a protective layer for the semiconductor against air moisture, (ii) the transparency of the polymer enables the propagation of the excitation beam through high order cladding modes [3]. Thicknesses of the FASnI<sub>3</sub> and PMMA were fixed to 0.8  $\mu$ m and 0.2  $\mu$ m, respectively. At these conditions, the waveguide propagates the fundamental mode TE<sub>0</sub> at the photoluminescence (PL) wavelength (890 nm) confined in the semiconductor together with TE<sub>2</sub> cladding mode at the excitation wavelength (532 nm), see

mode distribution in Fig. 1a. The waveguides were analyzed by pumping the structures with Nd:Yag laser doubled at 532 nm (1 kHz, 1ns) and collecting the photoluminescence (PL) with a grating spectrograph (DNS-300 from DeltaNu) and detected by a back-illuminated Si CCD (DV420A-OE from Andor) at its exit. All measurements were carried out at room temperature and ambient conditions.

ITu1C.2

#### 3. Waveguide characterization.

The PL of the waveguides was initially characterized by pumping the surface of the samples and collecting the PL transmitted through the substrate. At low excitation fluences the PL shows a Gaussian PL spectra centered at 890 nm, see black line in Fig. 1b. However, above a threshold of around 40  $\mu$ J/cm<sup>2</sup> the spectrum collapses to a narrow band characteristic of Amplified Spontaneous Emission [3]. Moreover, this band is modulated by narrow lines of around 0.6 nm which are explained as random lasing action caused by the PL light scattering on the polycrystalline grains [4]. Once the basic parameters of the PL/ASE were analyzed in the through-substrate transmittance geometry, the waveguides were characterized by end-fire coupling the laser at the input edge of the sample with the aid of a microscope objective and collecting the waveguided PL at the output edge of the sample with another microscope objective. The PL decoupled from the output edge of the waveguide showed the generation of ASE above excitation fluences of  $0.5 \,\mu$ J/cm<sup>2</sup>, see Fig. 2a. The waveguides also show narrow emission lines (<1 nm) corresponding to RL. More interestingly, the RL shows a preferable transverse electric (TE) polarization, which enables practical applications where the control of the polarization becomes necessary. The RL action is also studied under bending conditions in the flexible device to stablish the dependence of the threshold, PL intensity and dichroism with the curvature radius, see Fig. 2b.



Fig. 2. Demonstration of ASE and RL observed for the waveguiding geometry. (a) PL spectra for different excitation fluences. (b) Log-Log plot of the PL intensity at 890 nm as a function of excitation fluence for different curvature radius.

#### Conclusions

In this manuscript, FASnI<sub>3</sub> films were incorporated in a flexible waveguide and demonstrated ASE and RL at room temperature and ambient conditions. This configuration imposes a preferable directionality for the RL reducing the ASE threshold and allowing the formation of localized RL loops with high Q factors (Q~1000) and narrow lasing lines (<1 nm). The stimulated emission threshold is reduced down to  $1.5 \,\mu$ J/cm<sup>2</sup> as compared to the through-substrate transmittance geometry and the structure shows a preferable TE lasing polarization. Finally, the dependence of the threshold and polarization is studied as a function of the bending radios of the PET substrate, and demonstrates that both the PL, intensity and dichroism can be tuned with the curvature radius.

#### References

[1] Y. Bonnassieux, C. J. Brabec, Y. Cao et al. "The 2021 flexible and printed electronics roadmap," Flex. Print. Electron., 6 023001 (2022)

[3] I. Suárez, E. J. Juárez-Pérez, V. S. Chirvony et al., "Mechanisms of Spontaneous and Amplified Spontaneous Emission in CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> Perovskite Thin Films Integrated in an Optical Waveguide," Phys. Rev. Appl. 13 064071 (2020)

[4] R. Sapienza, "Determining Random Lasing Action," Nat. Rev. Phys. 1, 690-695 (2019).
[5] I. M. S. Sanchez-Diaz, R. S. Sánchez, S. Masi et al., " Tin perovskite solar cells with >1300 hours of operational stability through a

synergistic chemical engineering approach. Unravelling the crucial role of halides chemistry" Joule, in press.

<sup>[2]</sup> I. Suárez, V.S. Chirvony, J. Sánchez-Díaz, R. S. Sánchez, I. Mora-Seró, and Juan P. Martínez-Pastor, "Directional and polarized lasing action on Pb-free FASnI3 integrated in flexible optical waveguides," Adv. Opt. Mater., submitted.