ADVANCED OPTICAL MATERIALS

Supporting Information

for *Adv. Optical Mater.,* DOI: 10.1002/adom.202001773

Optical Gain of Lead Halide Perovskites Measured via the Variable Stripe Length Method: What We Can Learn and How to Avoid Pitfalls

*Ada Lili Alvarado-Leaños, Daniele Cortecchia, Giulia Folpini, Ajay Ram Srimath Kandada, and Annamaria Petrozza**

Material Characteristics

The result of the X-ray diffraction (XRD) measurement is presented in Figure S1a, which confirms the formation of $MAPbI_3$ with the expected tetragonal crystalline structure without the presence of PbI_2. The broad signal peaked at 25° is due to the amorphous glass substrate underneath the perovskite film (having a thickness of 141±4 nm). The absorption spectrum in Figure S1b further confirms the formation of $MAPbI_3$ with absorption edge at 780 nm.

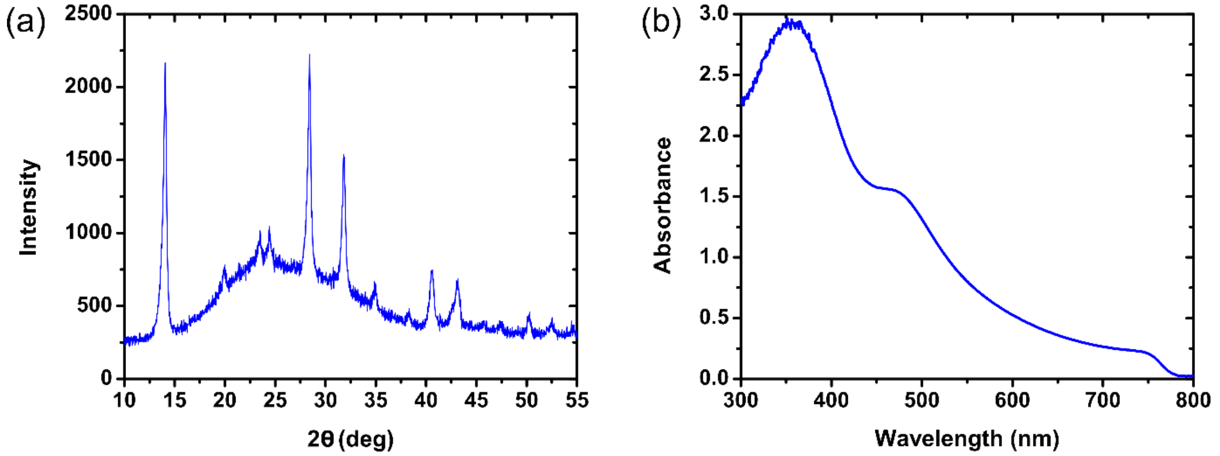

Figure S1. (a) X-ray diffraction (XRD) pattern of the studied $MAPbI_3$ sample and (b) corresponding absorption spectrum.

Beam Profile

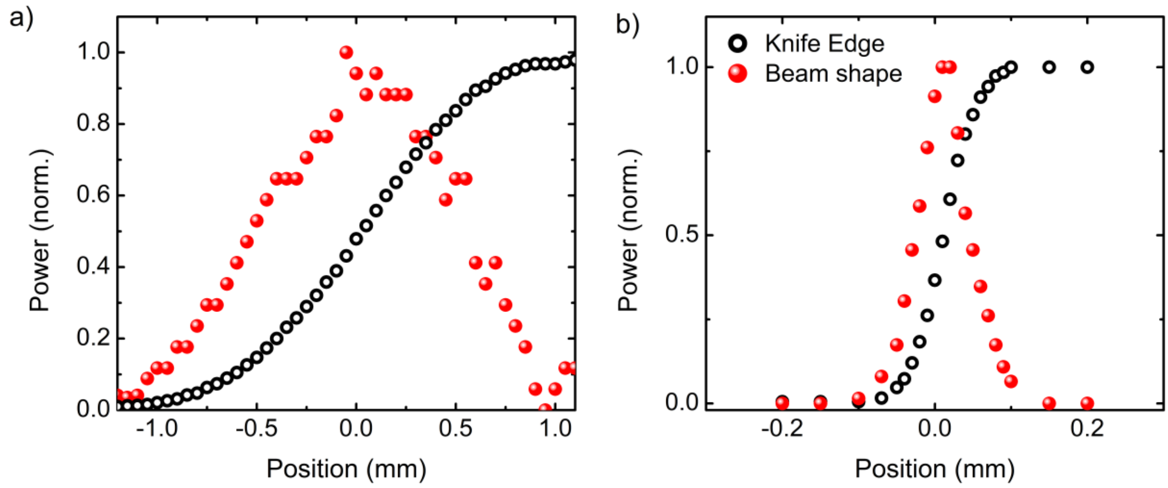

Figure S2. Beam shape without beam expander along the a) horizontal and b) vertical direction. The figures show the cumulative intensity (open black symbols) and the beam profile, i.e. the derivative of the measured intensity (closed red symbols). Both profiles are Gaussian, the horizontal profile has a radius of $r_{1/e}$ = 650 μm and the vertical profile a radius of $r_{1/e}$ = 35 μm.

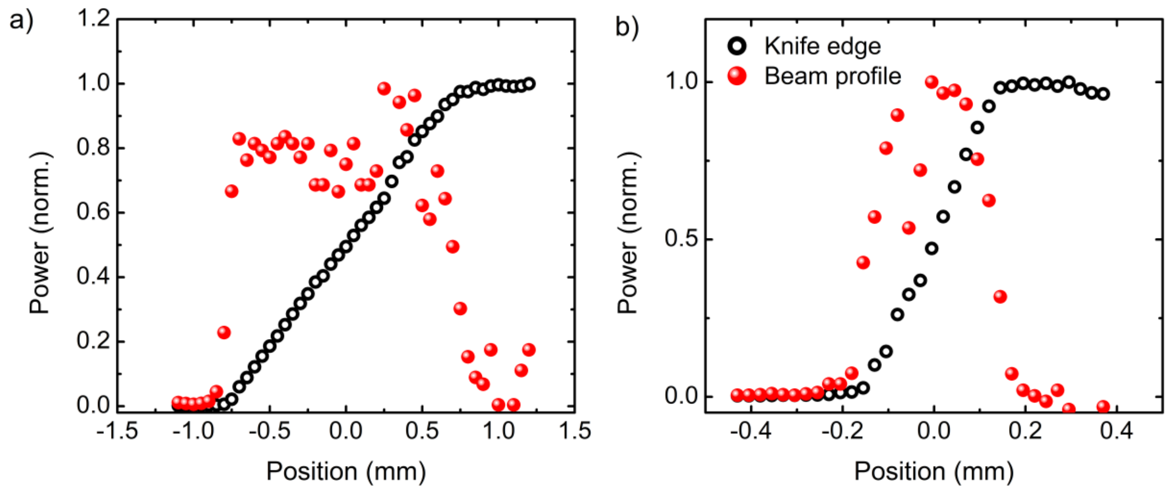

Figure S3. Typical beam shape used during for the VSL measurements, as obtained from the knife edge technique along the a) horizontal and b) vertical direction. The cumulative intensity (open black symbols) and the beam profile (closed red symbols) are shown. With the help of a beam expander and a fixed slit, the resulting horizontal profile is uniform and possesses a stripe length of z = 1.5 mm, while the vertical profile is Gaussian with $r_{1/e}$ = 145 μm.

Shifting Excitation Spot

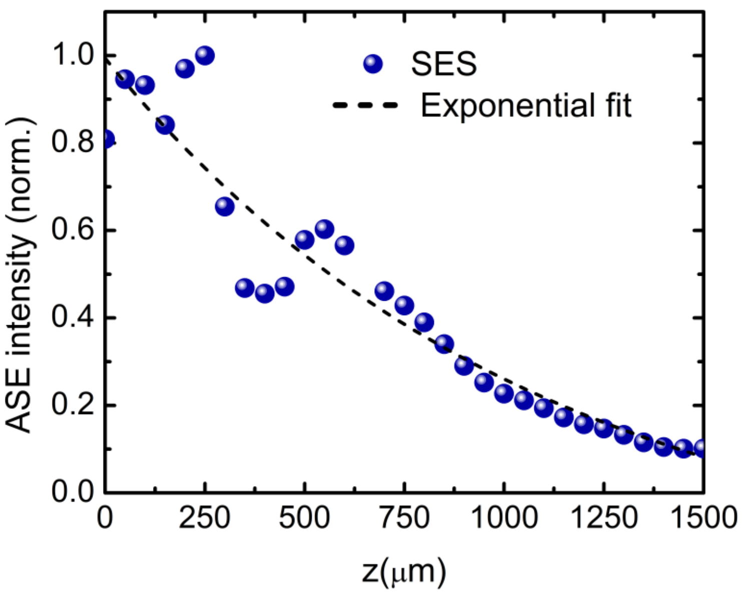

Figure S4. The shift excitation spot technique (SES) is used to evaluate the outcoupling of ASE from the side of the sample into the collection line. In this plot the blue symbols represent the ASE intensity (integrated between 785 nm – 805 nm), collected while shifting the excitation spot along the length of the stripe (z_{max} = 1.5 mm). The measurement was conducted by moving the illuminated area from the sample edge (z=0) to the center. The pump fluence was set at 200 μJ/cm^2. By fitting the SES curve to an exponential decay (dashed black line), the absorption losses in the sample can be estimated from the Beer-Lambert law: $I_{SES} = I_0 e^{-\alpha z}$, resulting in $\alpha = 9.6 \pm 3.3$ cm^{-1}.

Fitting the VSL data

For the smallest input fluence, the calculation of the optical gain coefficient started by fitting the VSL data to the small signal gain model. Many fits were performed for progressively increasing stripe lengths, all starting from a stripe length of 0.02 cm. The best fit was chosen to be the last gain value before the calculated gain started to decrease. From the best fit, the values of the optical gain coefficient, g, and the parameter A_{sp} were obtained. Afterwards, using as initial parameters the results from the small-signal gain model fit, the VSL data was fitted to the saturation model. The parameter A_{sp} was restricted to vary within $1x10^4$ and $4x10^4$, following the results obtained from the best small-signal gain fit.

The VSL spectra over a wide range of input fluences

VSL measurements were carried out for a wide range of input fluences, both above and below threshold. Below threshold, for an input fluence of 7 μJ/cm^2, the VSL spectra (Figure S5.a) show a dominant emission at ~770 nm. However, for the smallest stripe lengths, the dominant emission is observed at around 800 nm. For the higher input fluence of 17 μJ/cm^2 (Figure S5.b), it can clearly be seen that an increase of the stripe length, results in a shift from a dominant peak at 800 nm to a dominant peak at 770 nm. When the input fluence is 33 μJ/cm^2 (Figure S5.c), both emissions (at 770nm and 800 nm) play an important role, especially for the largest stripe lengths. Finally, at an input fluence of 67 μJ/cm^2 (Figure S5.d), ASE becomes the more relevant process, and the second peak remains, but becomes less significant.

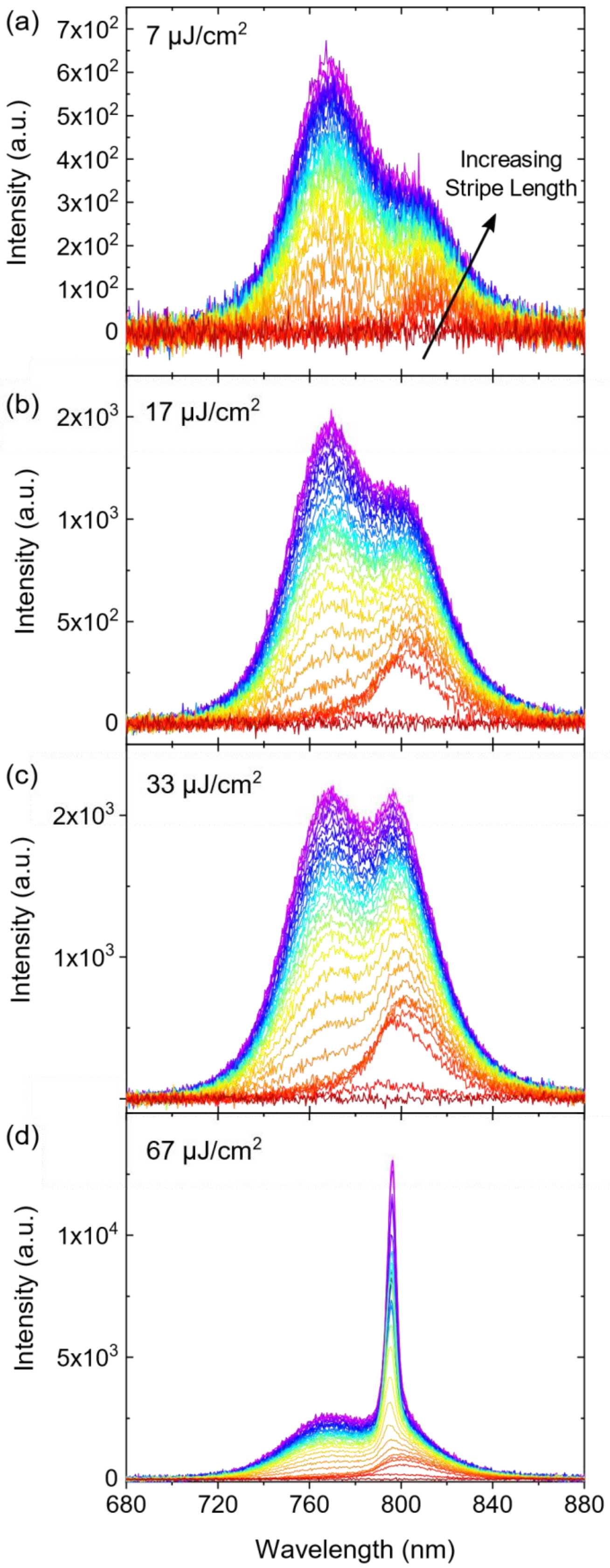

Figure S5. VSL spectra for a set of different input powers: a) 7, b) 17, c) 33 and d) 67 μJ/cm^2.

The gain spectrum over a wide range of wavelengths

For three input powers, 100, 67 and 50 μJ/cm^2, each gain spectrum was analysed over a wide range of wavelength between 700 and 860 nm (Figure S6.d). The set of spectra (Figure S6.a,b,c) indicate that the ASE peak is located around 790 nm, which is confirmed by the narrow maximum observed in the gain spectrum of Figure S6.d. However, a second peak of positive gain values is observed for shorter wavelengths next to the ASE peak. The gain values only become negative after ~710 nm. The second maximum on the gain spectrum is more significant for smaller input fluences.

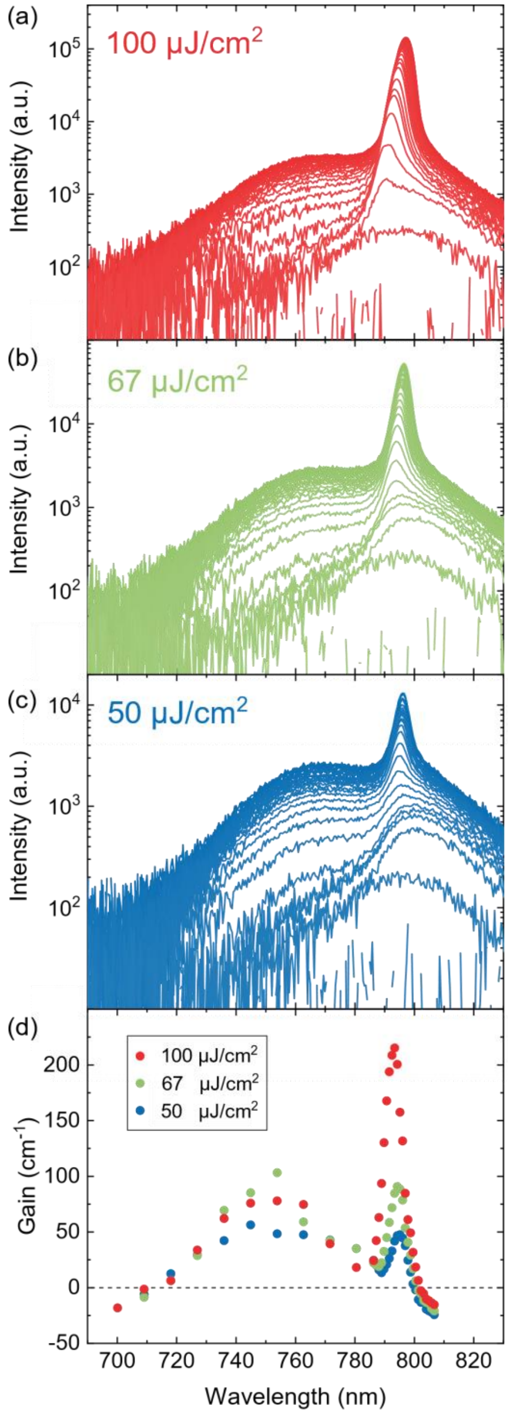

Figure S6. VSL spectra for three different input fluence: (a) 100, (b) 67 and (c) 50 μJ/cm^2, respectively. d) Gain spectrum calculated using the small-signal model and the spectra of (a), (b) and (c).

Forwards and Reverse VSL curves

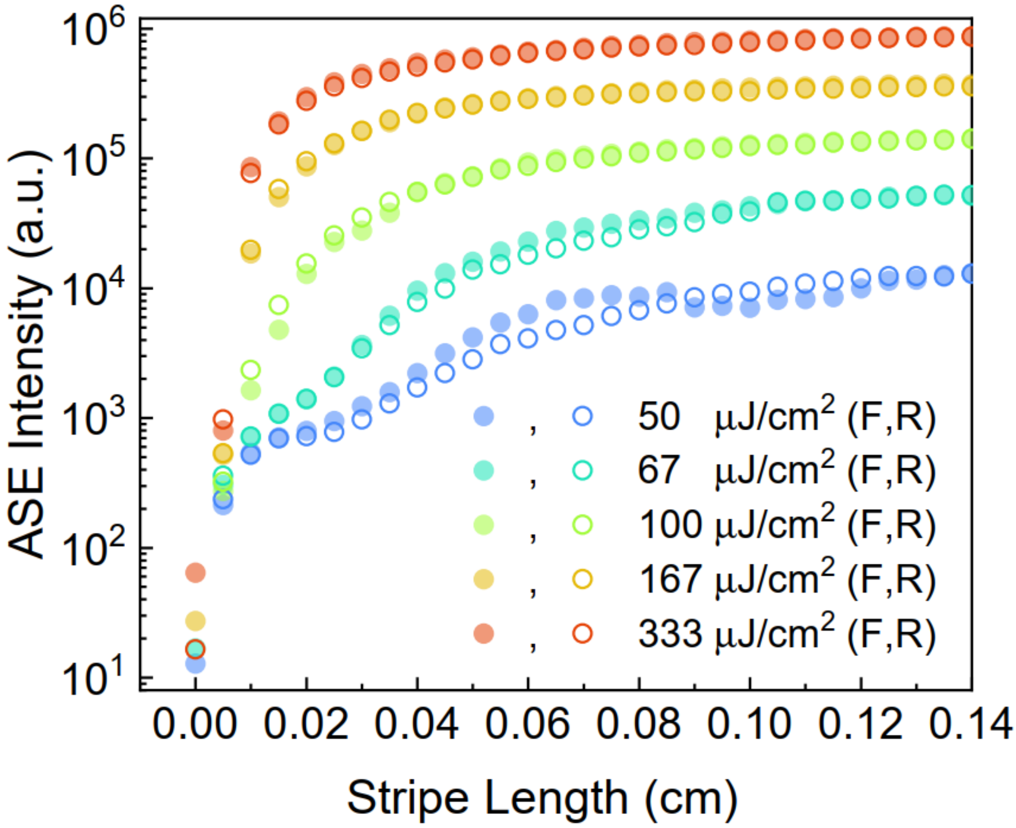

Figure S7. Fluence dependence of the VSL curve from 50 to 333 μJ/cm^2, for forwards (filled circle) and reverse (open circle) measurements.