
Epitaxial lead-halide-perovskite microcrystal microcavity lasers



Wolfgang Heiss

Short Summary About Solar Cells and Modules Market:

The Global Solar Cells and Modules market is anticipated to rise at a considerable rate during the forecast period, between 2024 and 2032. In 2024, the market is growing at a steady rate [CAGR of 9.2%] and with the rising adoption of strategies by key players, the market is expected to rise over the projected horizon.

2020-2026 Edge-emitting lasers market revenue forecast by segment (\$B)

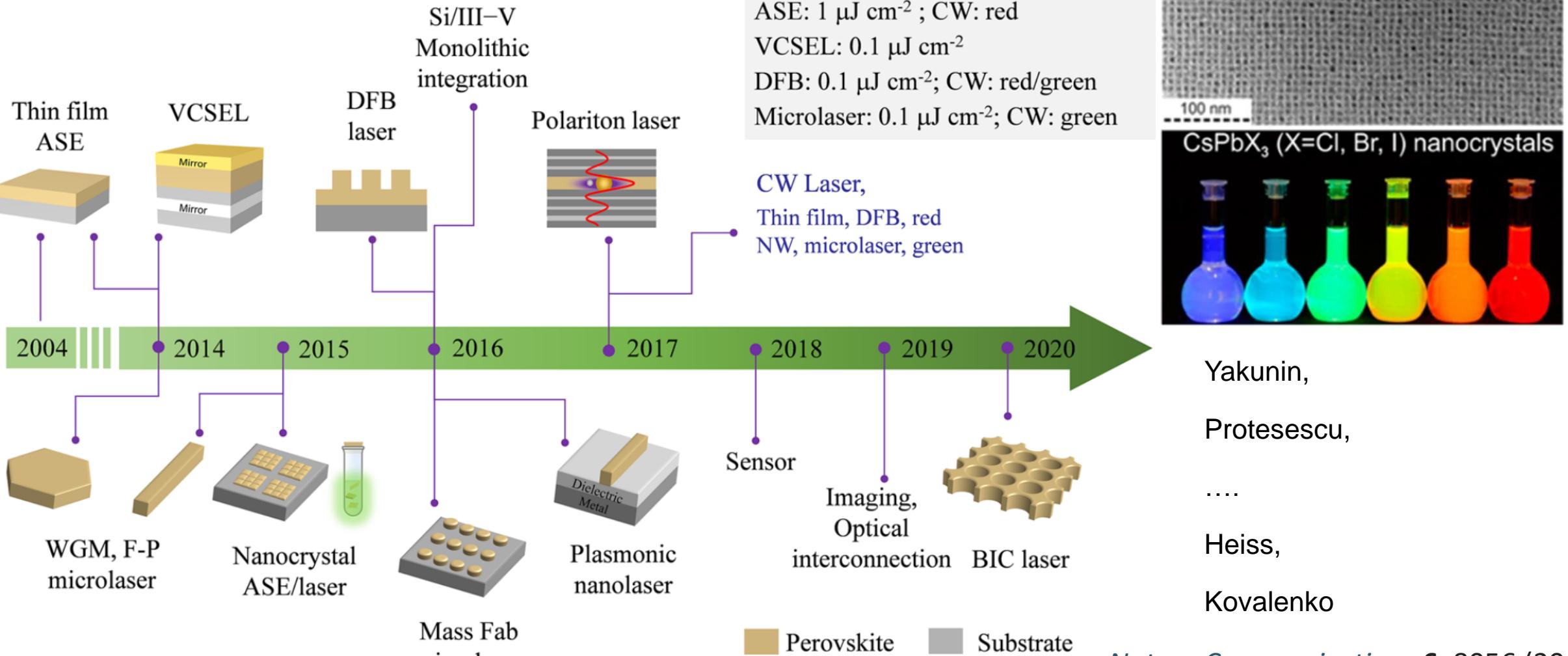
(Source: Edge Emitting Lasers - Technology and Market Trends 2021 report, Yole Développement, 2021)

- Optical communications
- Emerging applications (Displays, sensing, medical and lighting)
- Other applications¹



1. Material processing (including marking), printing, optical storage and R&D

15% compound annual growth rate



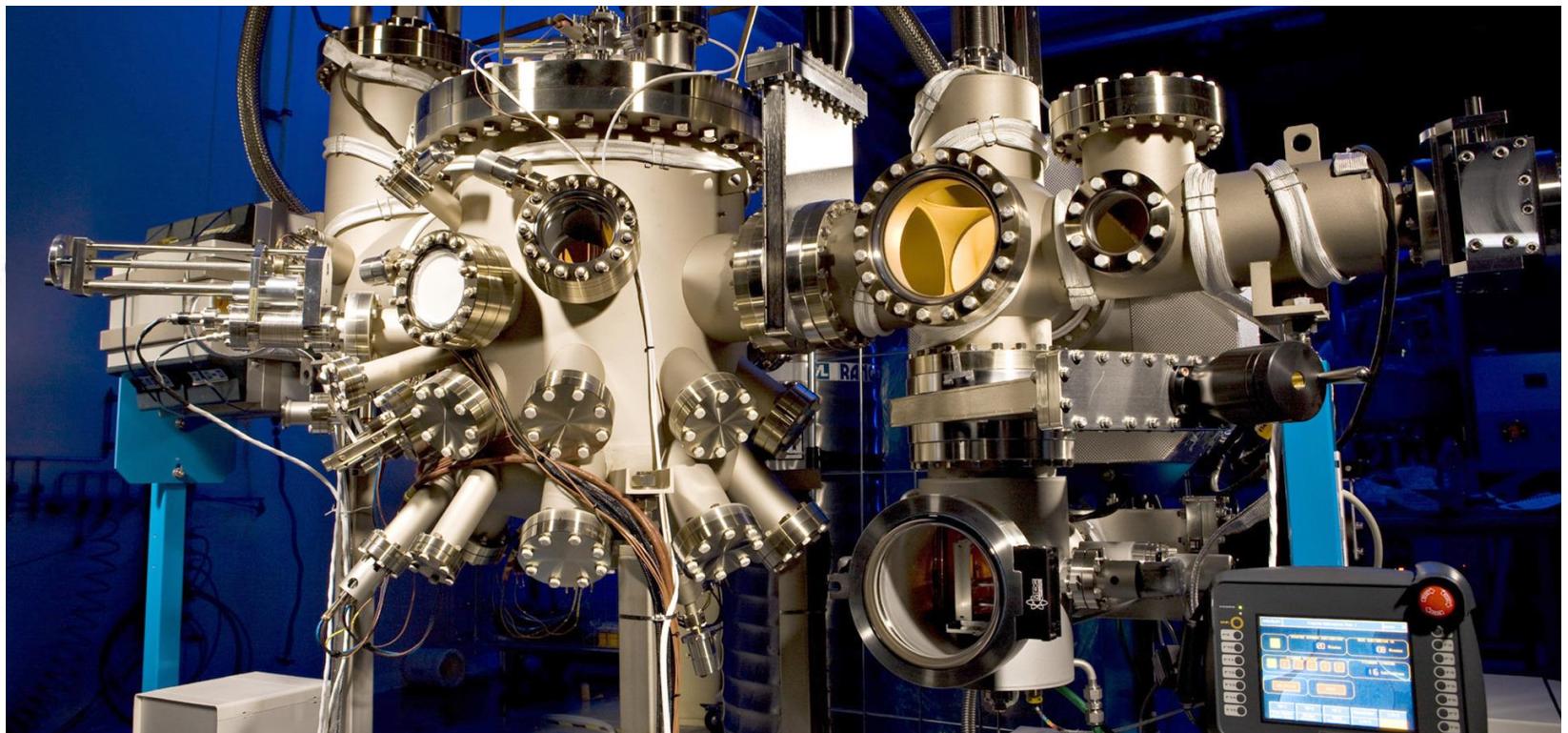
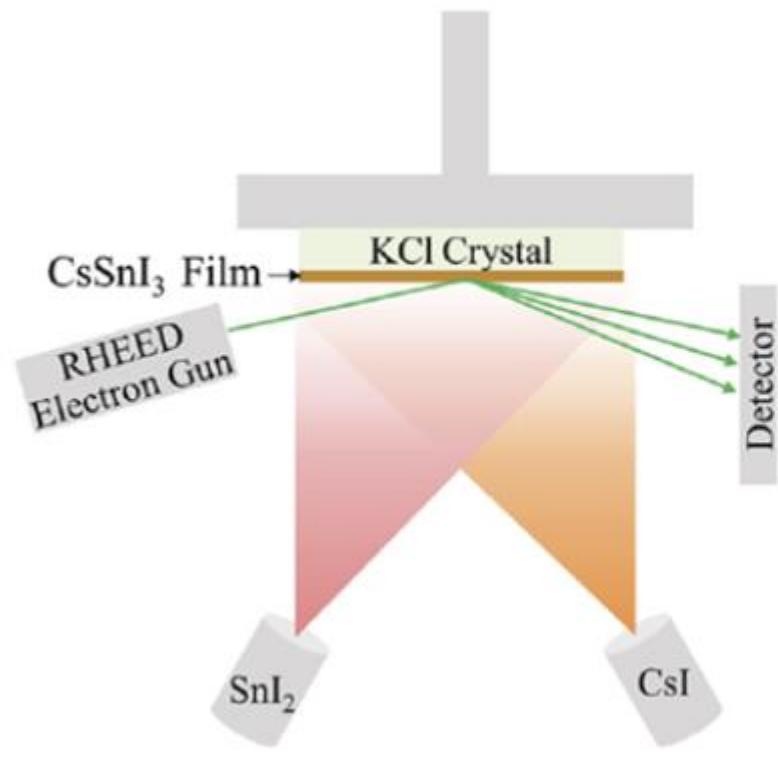
2022

- a) Solution epitaxial lasers rivaling vapor phase deposited ones.
- b) Squeezing the threshold of microcrystal microcavity laser
- c) Surface passivation of epitaxial microstructures
- d) Positioning of perovskite microcrystal lasers

a) Solution epitaxial lasers rivaling vapor phase deposited ones

Molecular Beam Epitaxy (MBE)

<https://www.ribel.com/>

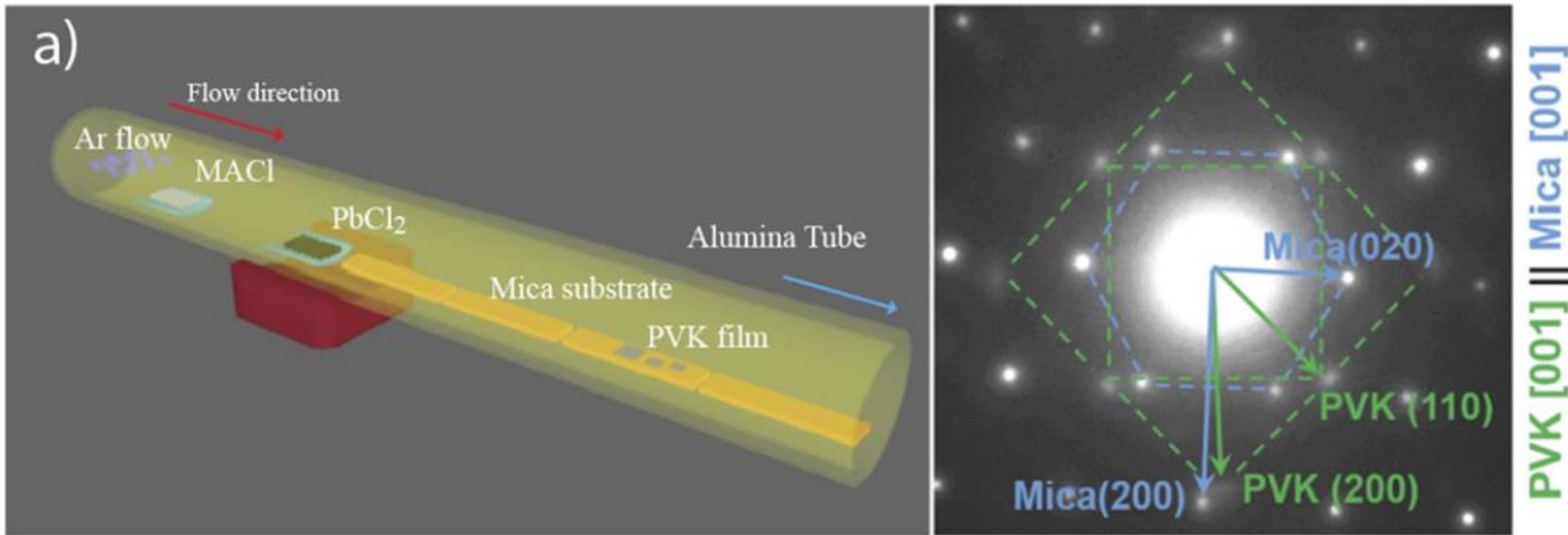


Wang et al., ACS Appl. Mater. Interfaces 11, 32076 (2019)

a) Solution epitaxial lasers rivaling vapor phase deposited ones

Chemical Vapor Deposition (CVD)

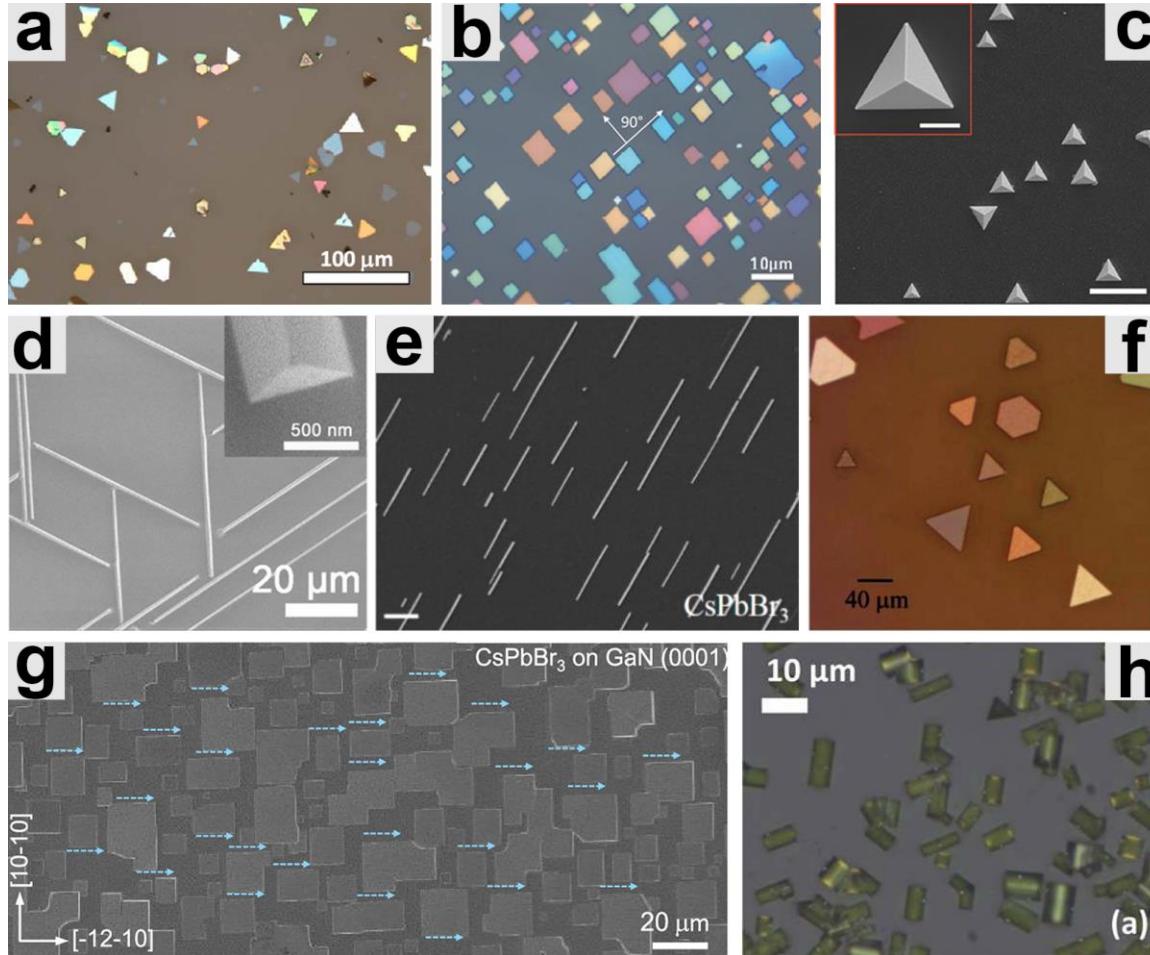
MAPbCl_3



Wang *et al.*, Cryst. Growth Des. 15, 4741 (2015)

a) Solution epitaxial lasers rivaling vapor phase deposited ones

Vapor-Phase Epitaxial Lasers



Material and cavity geometry	Lasing threshold ($\mu\text{J}/\text{cm}^2$)	Q-factor	Growth method	Substrate	Operation Temp.	Pump Laser	Year
MAPbI ₃ triangular nanoplatelet	37	650	CVD	mica	RT	400 nm, 150 fs, 1 kHz	2014 (a)
MAPbI _{3-x} Br _x triangular nanoplatelet	128	900	CVD	mica	RT	400 nm, 50 fs, 1 kHz	2016 (b)
CsPbBr ₃ square nanoplatelets	2.2	~3530	CVD	mica	RT	400 nm, 80 fs, 1 kHz	2018 (c)
MAPbBr ₃ cube-corner pyramid	92–2200	~960	CVD	mica	80–200 K	400 nm, 80 fs, 1 kHz	2018 (d)
MAPbBr ₃ cube-corner pyramid	26, 75	—	CVD	mica/Ag	80, 300 K	400 nm, 80 fs, 1 kHz	2018 (e)
Composition graded CsPbBr _x I _{3-x} triangular nanowire	16 (521 nm) 28 (556 nm)	~1737 ~1390	CVD	mica	RT	400 nm, 150 fs, 1 kHz	2018 (f)
CsPbBr ₃ triangular nanowire	4	2675	Thermal evaporation	sapphire	RT	470 nm, 100 fs, 1 kHz	2018 (g)
CsPbI ₃ triangular nanowire	21	2256				400 nm, 100 fs, 1 kHz	2019 (h)
CsPbCl ₃ triangular nanowire	11	1931					
MAPbI ₃ triangular nanoplatelet	18.7	2600	CVD	mica	RT	343 nm, 290 fs, 6 kHz	2019 (i)
CsPbBr ₃ rectangular microplatelet	700	~781	CVD	GaN	RT	400 nm, 80 fs, 1 kHz	2019 (j)
CsPbBr ₃ rectangular microrod	136	5360	CVD	mica	RT	355 nm, 350 ps, 1 kHz	2021 (k)

- a) Zhang et al. *Nano Letters* 14, 5995 (2014) b) Zhang et al. *AFM* 26, 6238 (2016) c) Mi et al. *Small* 14, 1703136 (2018) d) Huang et al. *AM* 30, 1800596 (2018)
 e) Wang et al. *ACS Nano* 12, 6170 (2018) f) Li et al. *AFM* 29, 1805553 (2019) g) Zhao et al. *ACS Nano* 13, 10085 (2019) h) Wu et al. *Opt. Express* 29, 37797 (2021)

a) Solution epitaxial lasers rivaling vapor phase deposited ones

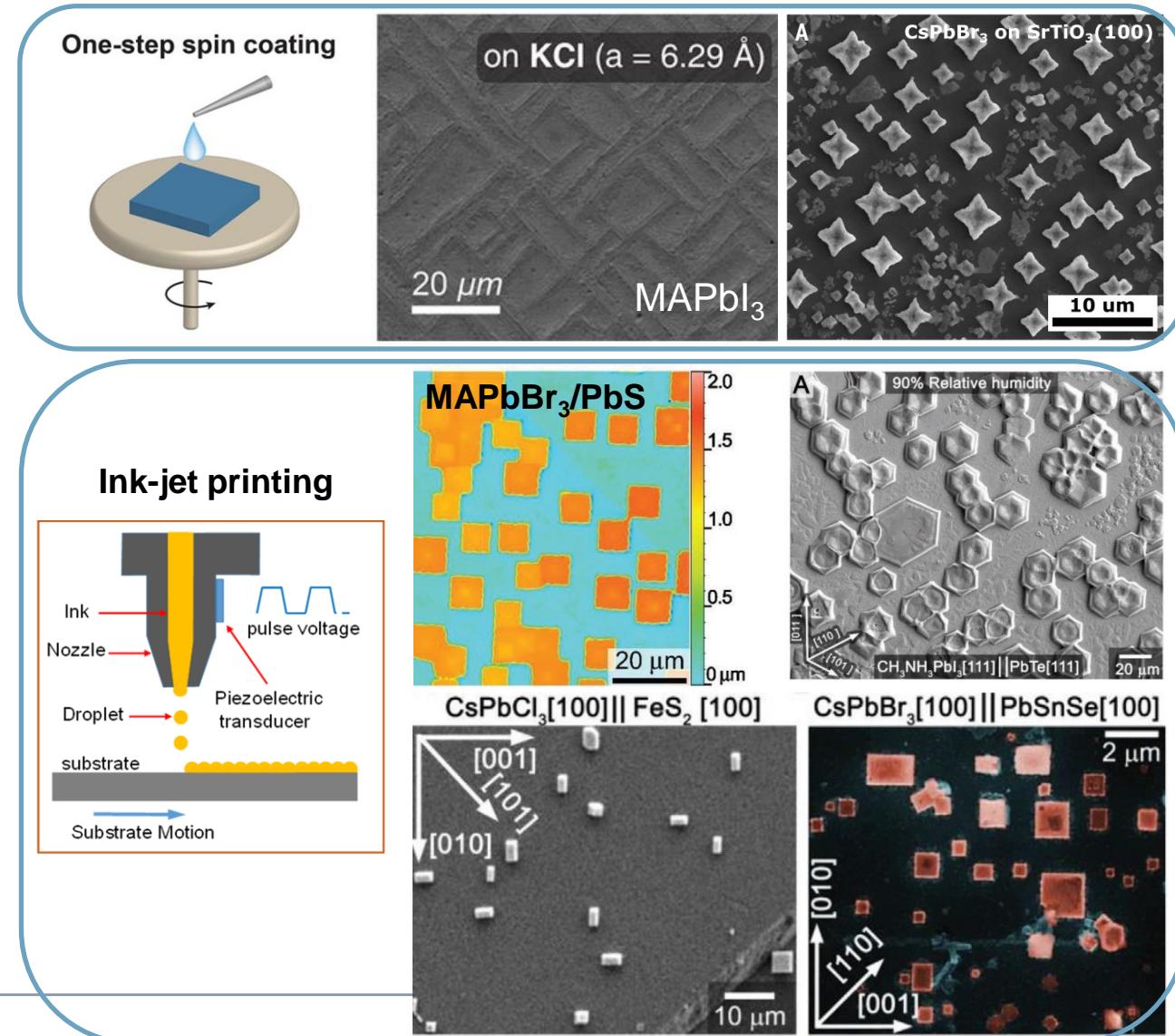
Solution–Phase Epitaxial Growth

	Solution Phase	Vapor Phase
Facile processing	✓	✗
Low temperature	✓	✗
No vacuum	✓	✗
Low-cost fabrication	✓	✗
Low-energy usage	✓	✗

Ji et al., Nano Lett. 18, 994 (2018)

Kelso et al., Science 364, 166 (2019)

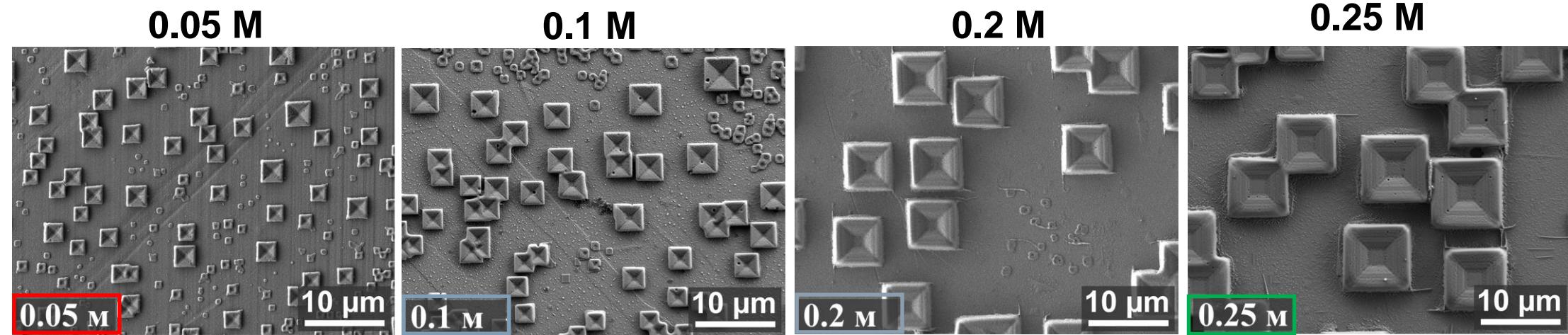
Sytnyk et al., Adv. Funct. Mater. 30, 2004612 (2020)



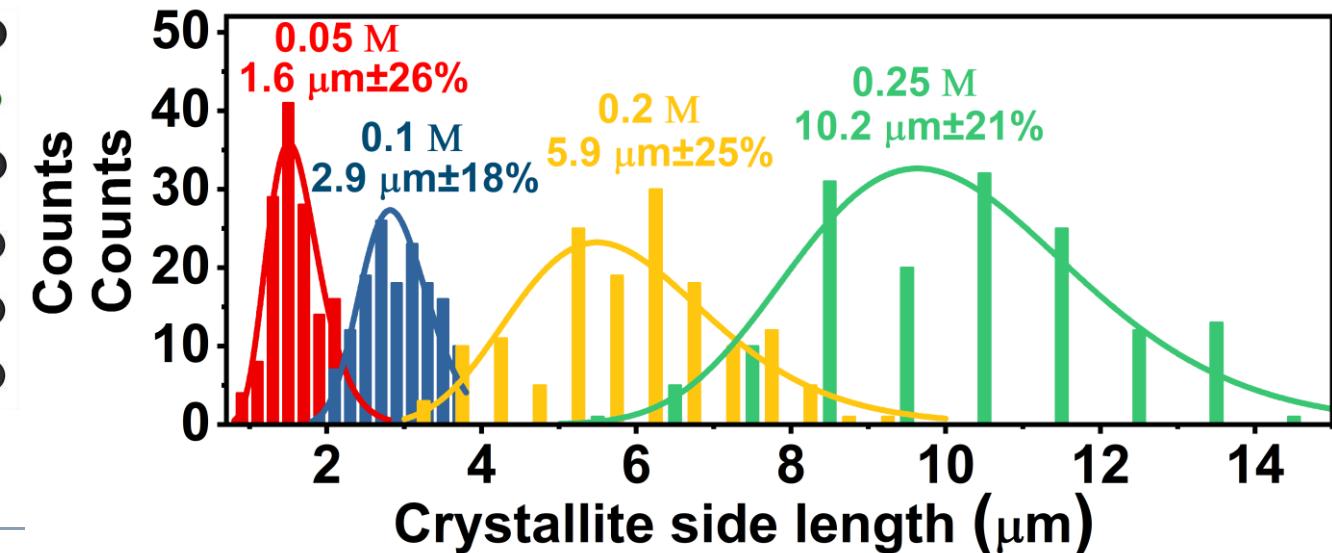
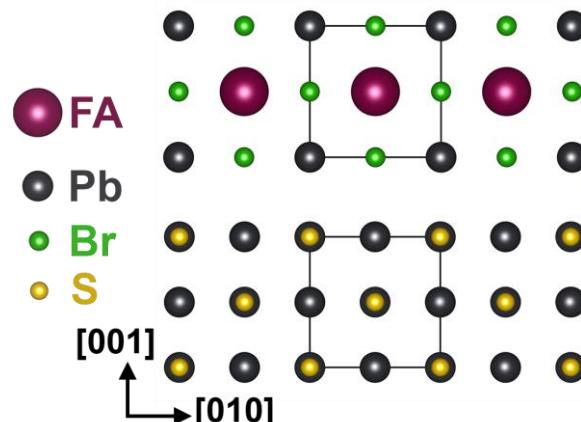
a) Solution epitaxial lasers rivaling vapor phase deposited ones

Epitaxial lasers by drop casting

Precursor Concentration →



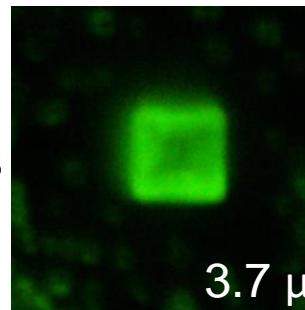
Solvent:
DMF+DMSO (3:1)
 $T_s = 80^\circ\text{C}$
Humidity = 80-90%
Bromium/Methanol
treatment



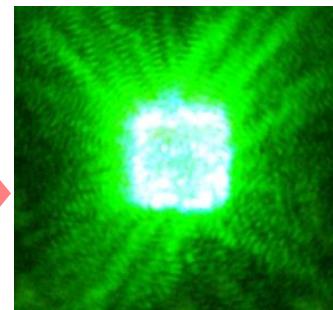
a) Solution epitaxial lasers rivaling vapor phase deposited ones

Epitaxial lasers by drop casting

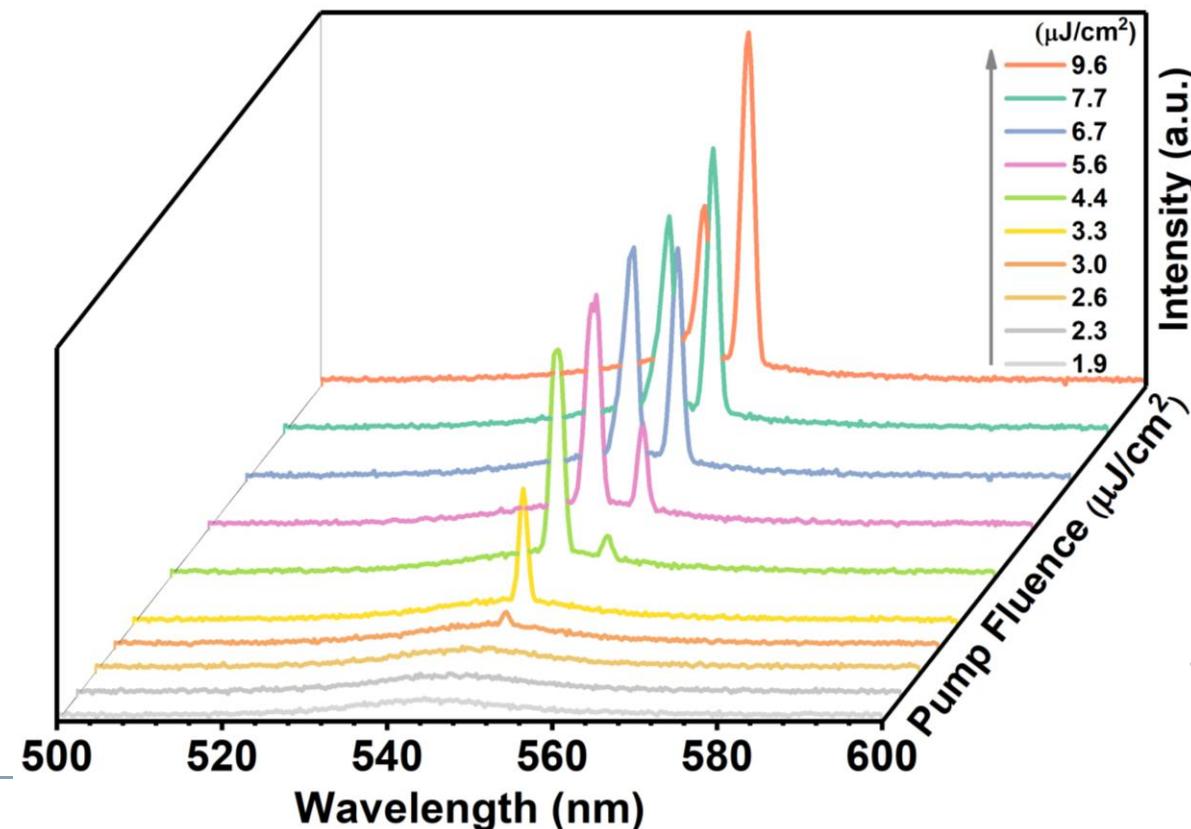
At low
pump
fluences



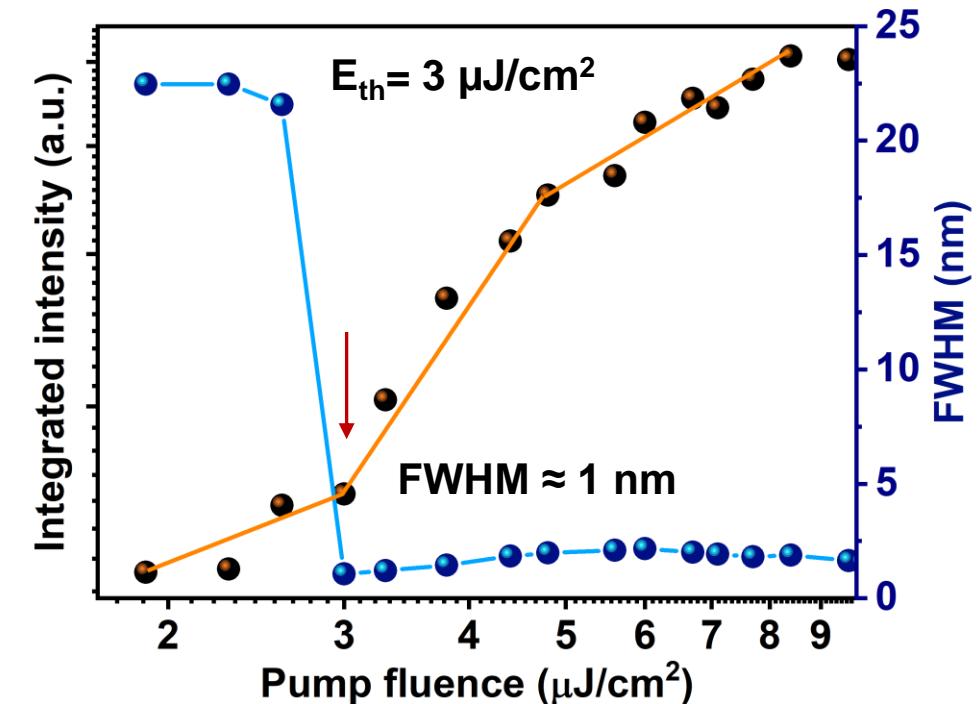
3.7 μm



Above
lasing
threshold



520 nm fs laser (>350 fs, 500 kHz)



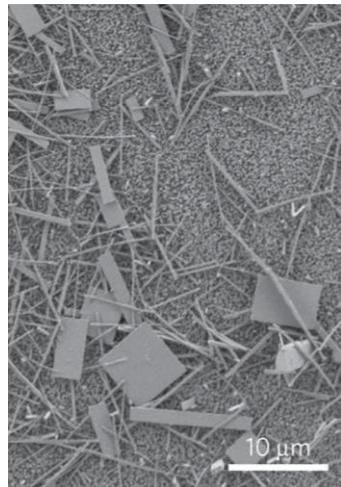
Lowest threshold for FAPbBr_3 in literature
and among lowest 20% of reported values for Pb-perovskites.

a) Solution epitaxial lasers rivaling vapor phase deposited ones

Antisolvent vapor assisted crystallization

FAU

AVC on glass



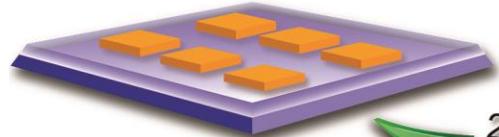
Zhu et al., Nature Materials 14, 636 (2015)

AVC on mica

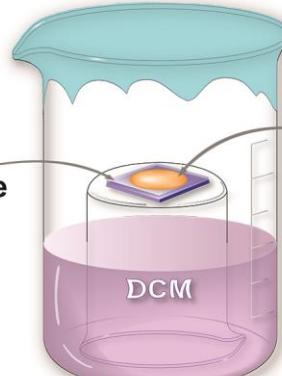
Hany Afify et al., Adv. Funct. Mater. 32, 2206790 (2022)

Micro-cuboid

MAPbBr₃/DMF
100 mL beaker
20 mL DCM



Mica Substrate



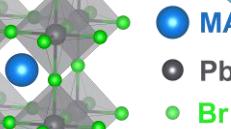
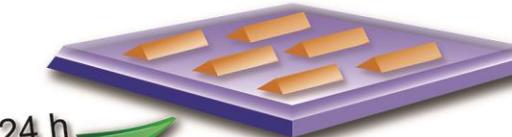
MAPbBr₃
Precursor

24 h RT

RT 24 h

Micro-prism

MAPbBr₃/(DMF+DMSO)
150 mL beaker
30 mL DCM



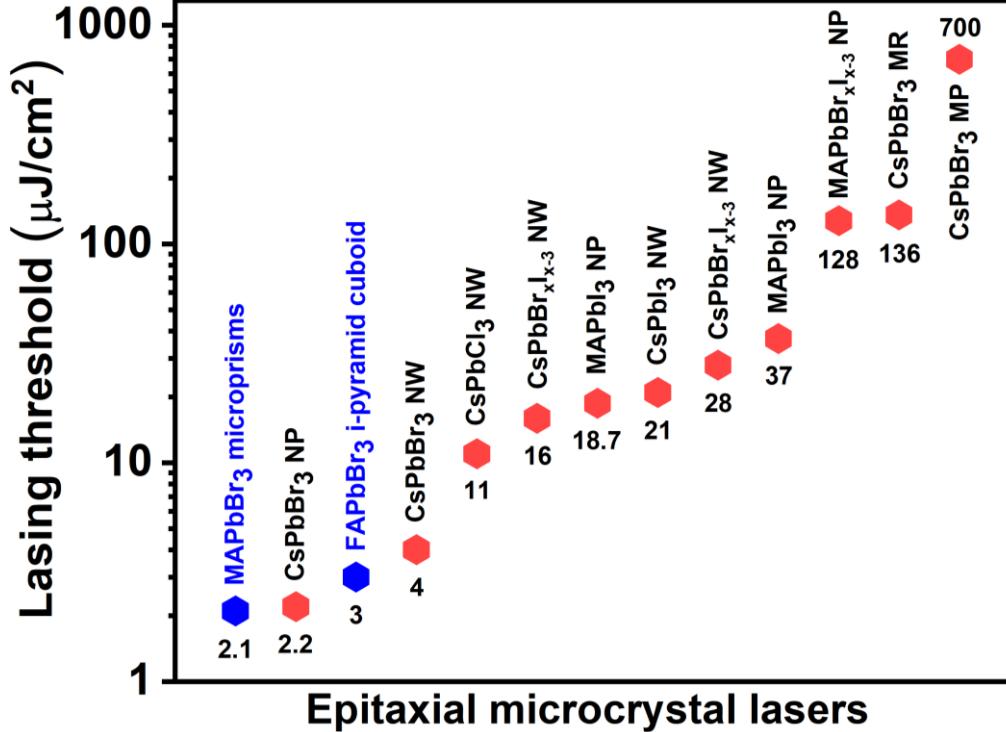
MAPbBr₃:
CH₃NH₃PbBr₃

1 μm

10 μm

10 μm

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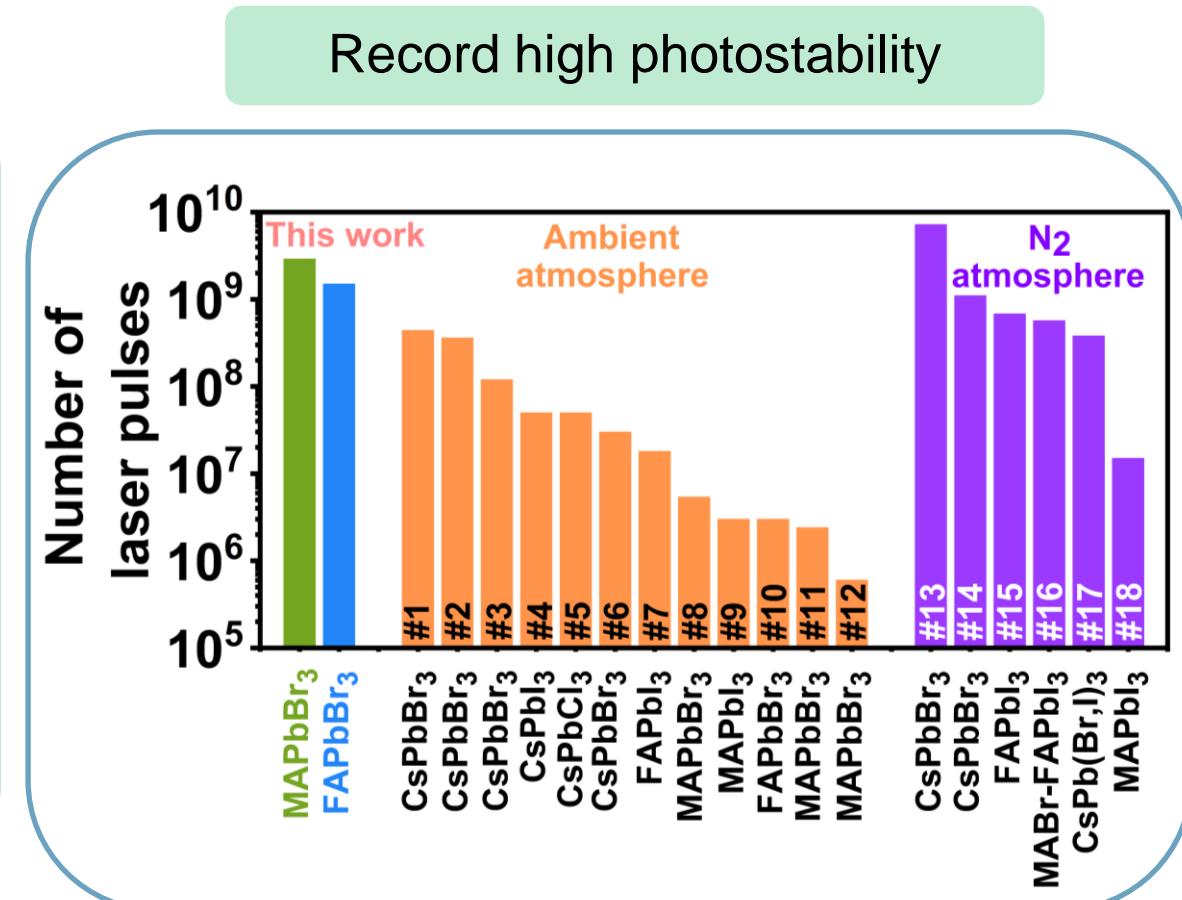
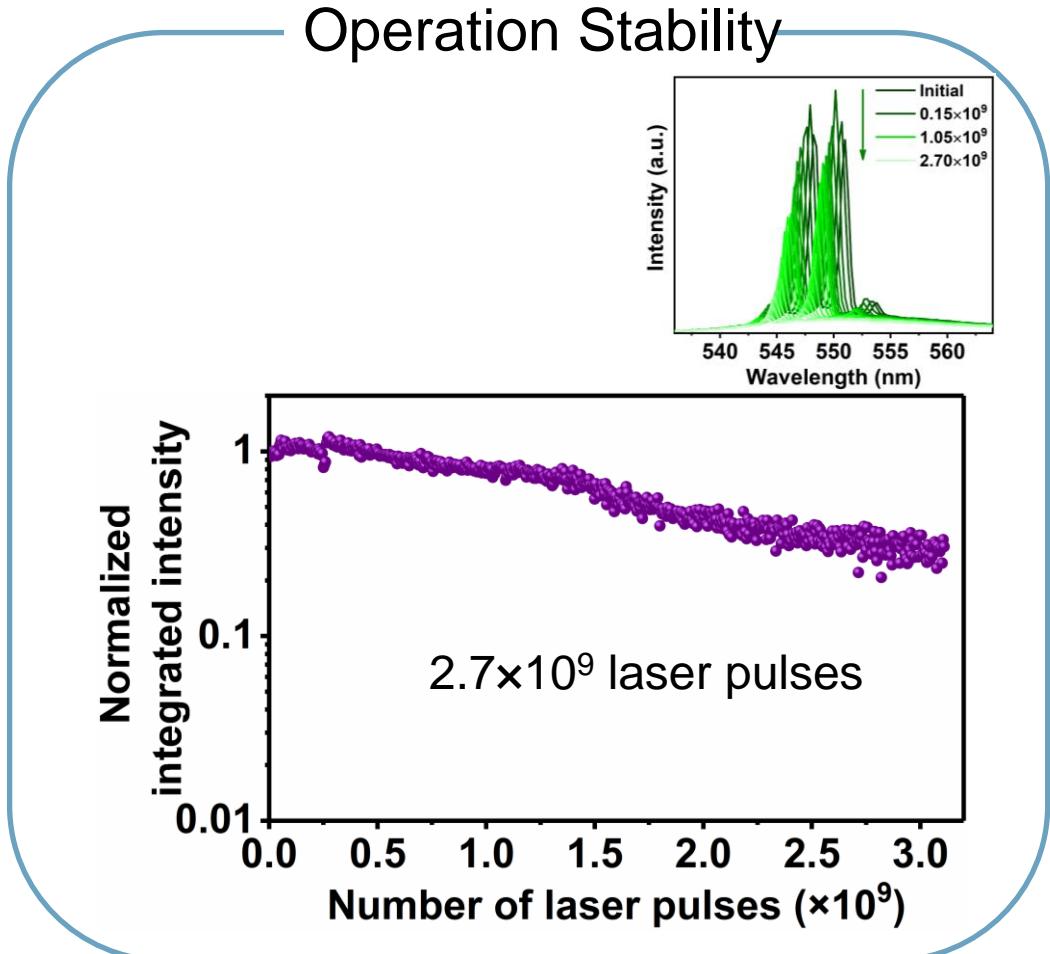


**Solution epitaxy
outperforms vacuum
epitaxy**

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a) Solution epitaxial lasers rivaling vapor phase deposited ones



2024

- a) Solution epitaxial lasers rivaling vapor phase deposited ones.
- b) Squeezing the threshold of microcrysal microcavity laser
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b) Squeezing the threshold of microcrysal microcavity laser

What to squeeze - **threshold power** (and not threshold energy) -> change from fs to ns excitation

FAPbBr₃

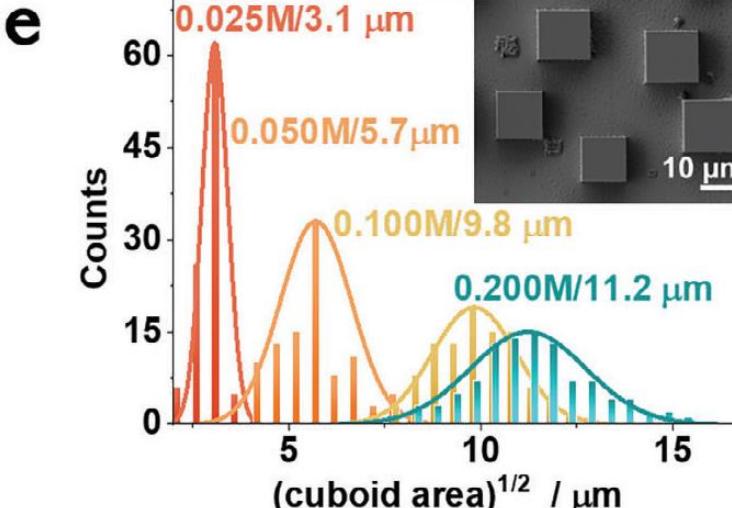
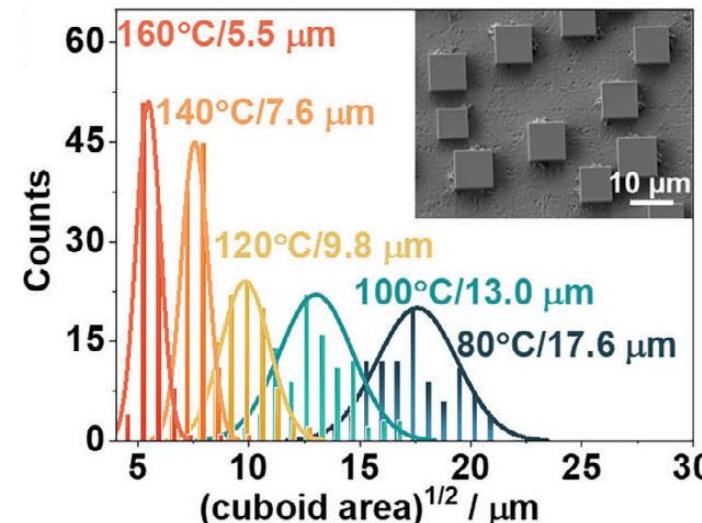
How to squeeze - optimize microresonator dimensions

- optimize perovskite material
- optimize excitation conditions (repetition rate)

Size control

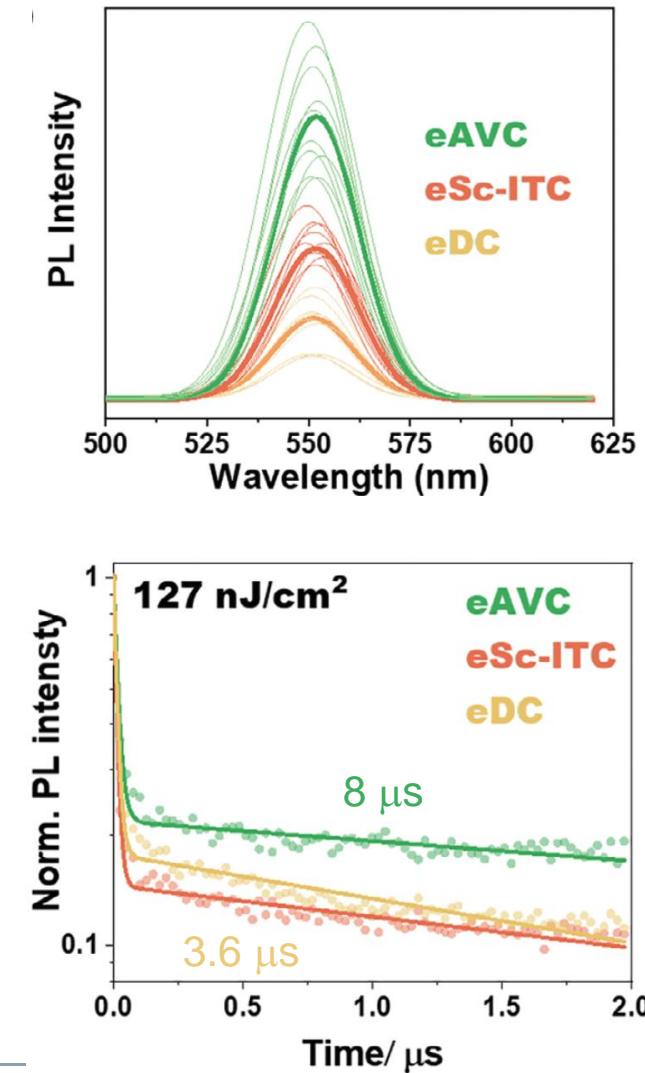
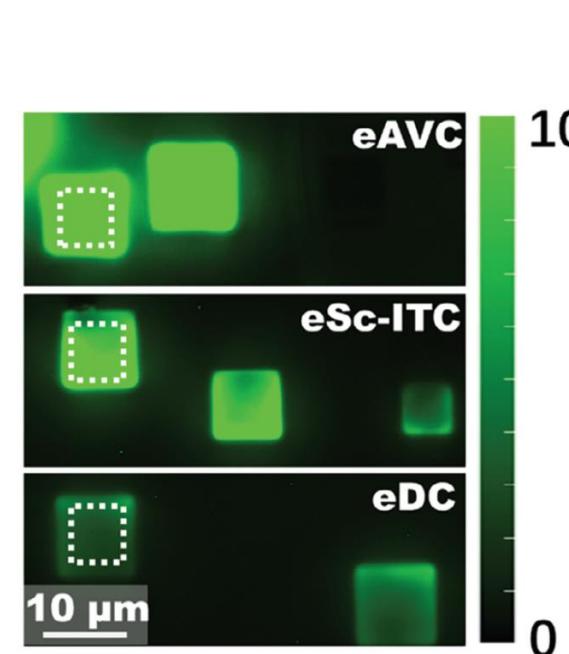
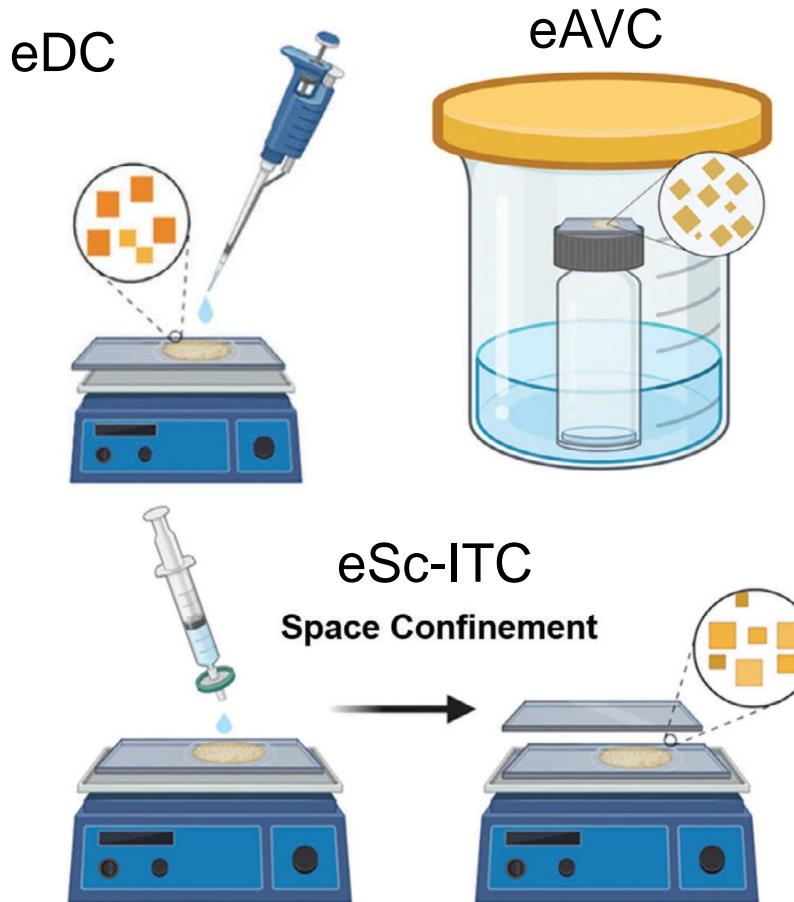
Drop casting from DMF/GBL

1.5 µl drop 3 min drying



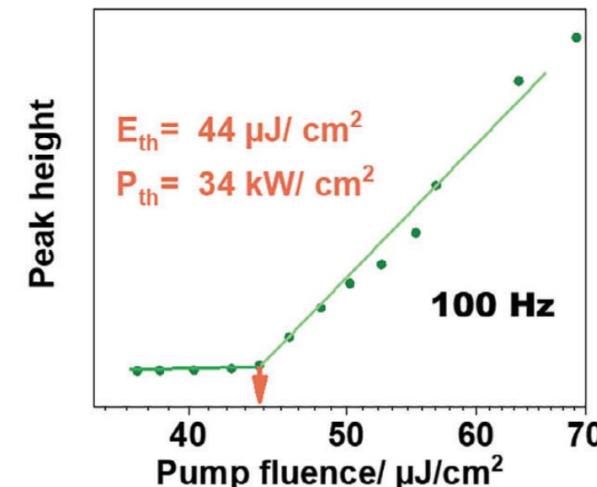
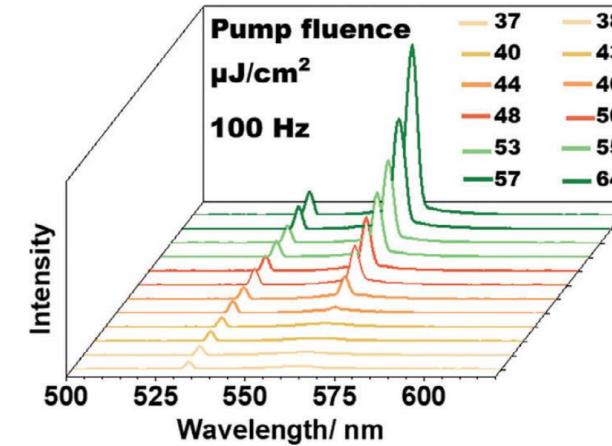
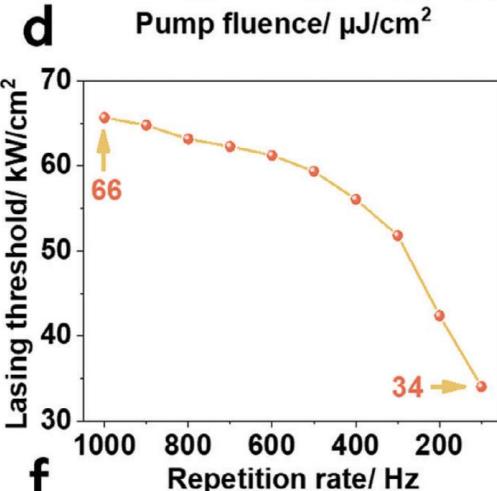
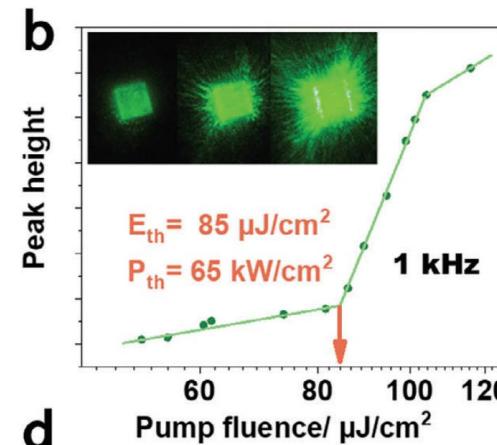
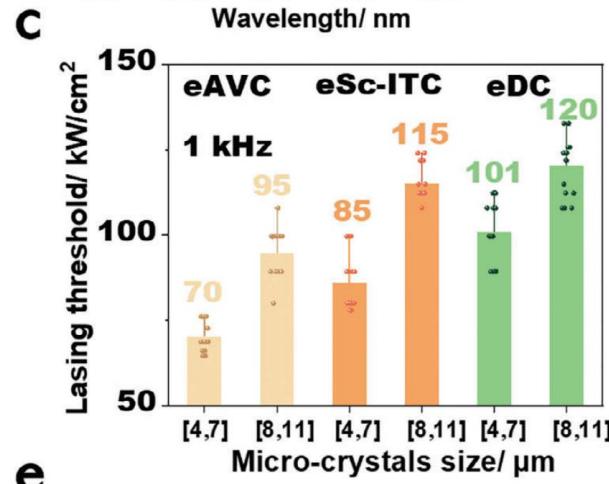
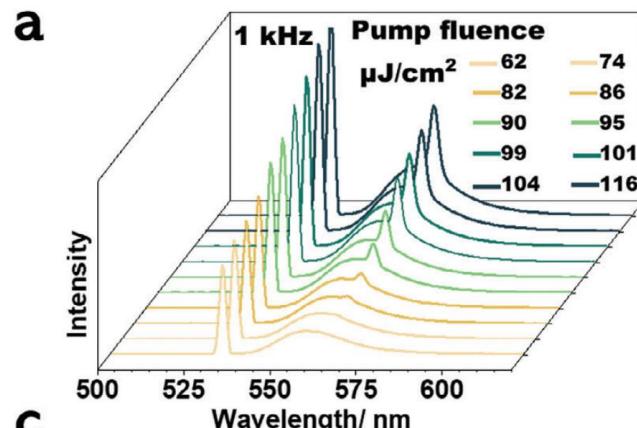
b) Squeezing the threshold of microcrysal microcavity laser

Quality optimization: epitaxial growth by



b) Squeezing the threshold of microcrysal microcavity laser

Optimize the excitation conditions



This is the new threshold power for epitaxial perovskite lasers, independent of material.

Solution epitaxial perovskite micro-crystal lasers exhibit similar performance as vacuum deposition ones, but are much easier and cheaper in preparation.

Optimizing microcrystal lasers includes size control, material improvement and selection of laser parameters.

Surface passivation is important for any epitaxial microstructures.
They can be positioned by seeds.

Principle investigators

Principle investigators

M. Sytnyk

H. Afify

S. Zhou

Y. Han

R. Grizfeld

V. Rehm

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DFG Deutsche
Forschungsgemeinschaft

