Supporting Information

Liquid-Phase Growth and Optoelectronic Properties of Two-dimensional Hybrid Perovskites $CH_3NH_3PbX_3$ (X=Cl, Br, I)

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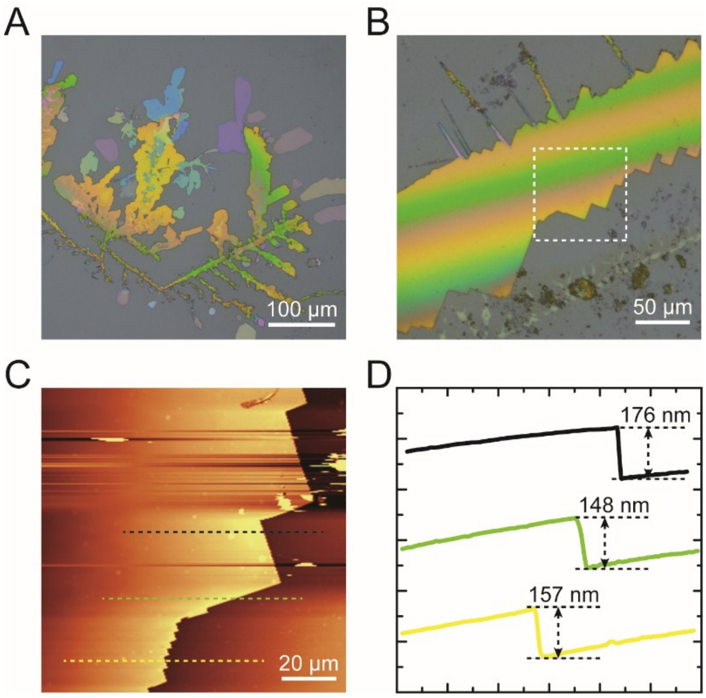

Figure S1. The $MAPbBr_3$ perovskites grown on the mica substrate without oleic acid. (**A, B**) Optical images. (**C**) AFM image of the perovskite denoted by the dotted box in (B). (**D**) High profile denoted by the black, green and yellow dash lines in (C). We note that the $MAPbBr_3$ perovskite sheets grown without oleic acid are as large as several hundreds, but it is hard to obtain the sheets thinner than 100 nm thick.

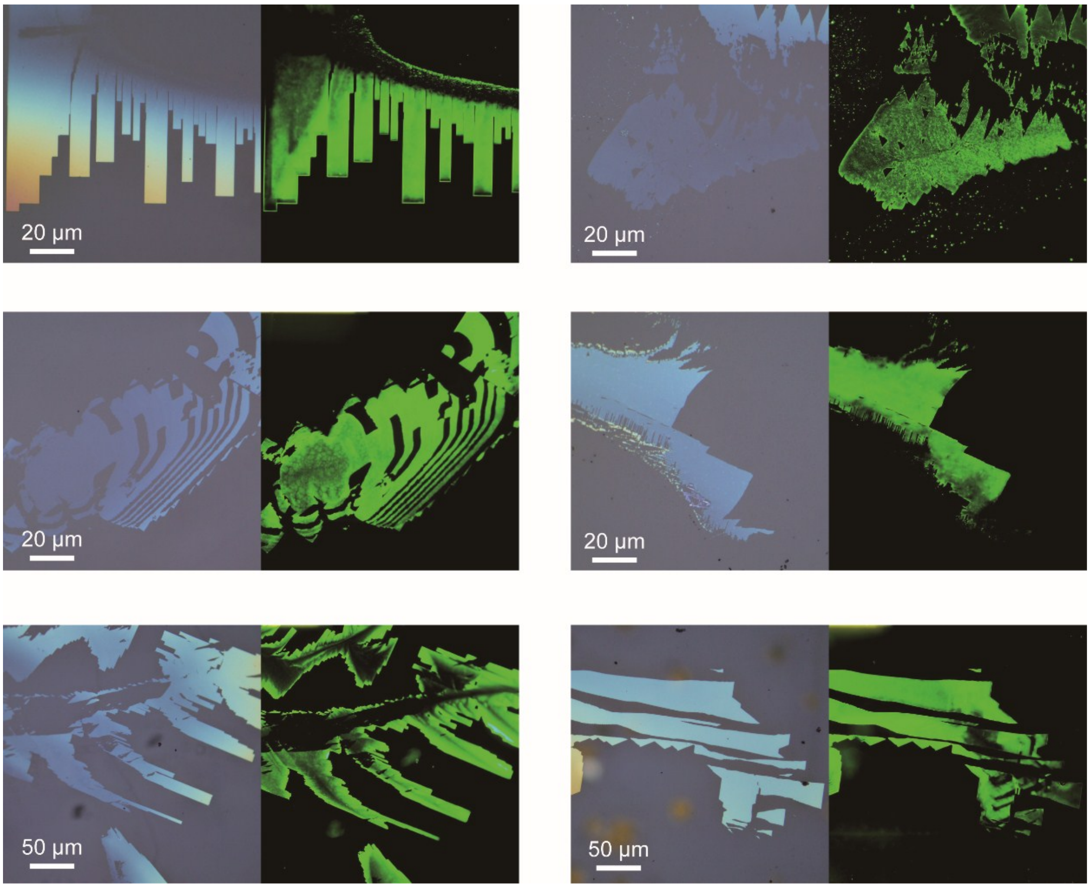

Figure S2. The optical and fluorescent images of 2D $MAPbBr_3$ perovskites grown on the mica substrate with oleic acid.

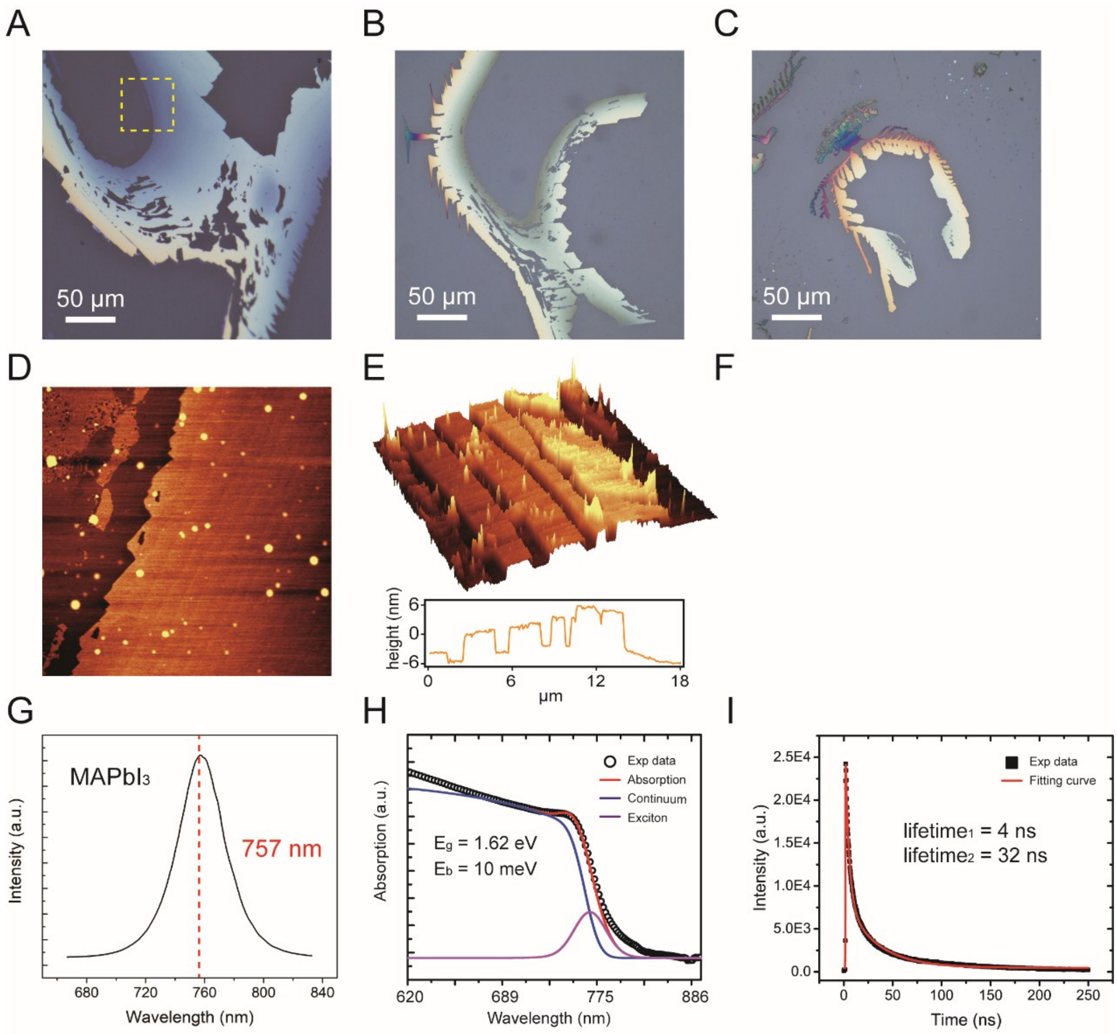

Figure S3. The liquid-phase growth of 2D $MAPbI_3$ perovskites on the mica. (**A, B** and **C**) Optical images. (**D**) AFM image of the MAPbI3 perovskites denoted by the yellow dashed box in (A). (**E**) 3D image and high profile of the grown 2D $MAPbI_3$ perovskites. The grown 2D sheet with the thickness of 6 nm, and have the very sharp edge and smoothing surface. (**G**) PL of the 2D $MAPbI_3$ perovskites, which has a blue shift to the bulk $MAPbI_3$ perovskites. (**H**) Absorption of the 2D $MAPbI_3$ perovskites. According the Eillot model, the bandgap E_g and the exciton binding energy E_b can be estimated to be 1.62 eV and 10 meV, respectively. (**I**) The lifetime of the 757 nm peak, which is estimated to be 4 ns and 32 ns.

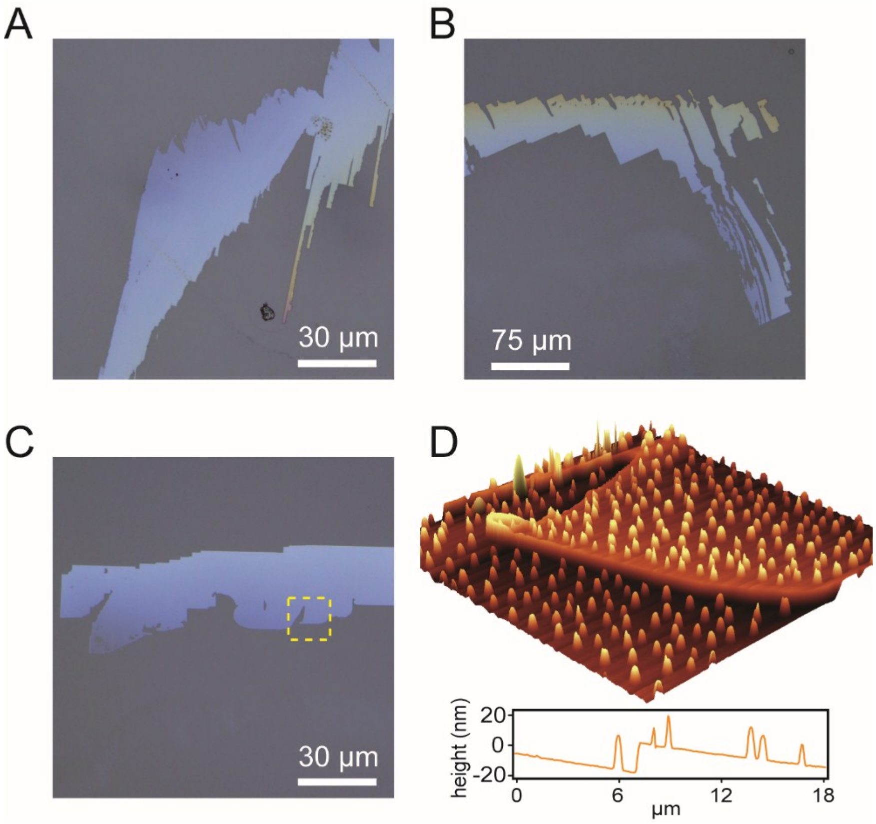

Figure S4. The liquid-phase growth of 2D $MAPbCl_3$ perovskites on the mica. (**A, B** and **C**) Optical images. (**D**) 3D AFM image and the high profile of the 2D $MAPbCl_3$ perovskite denoted by the yellow dashed box in (C).

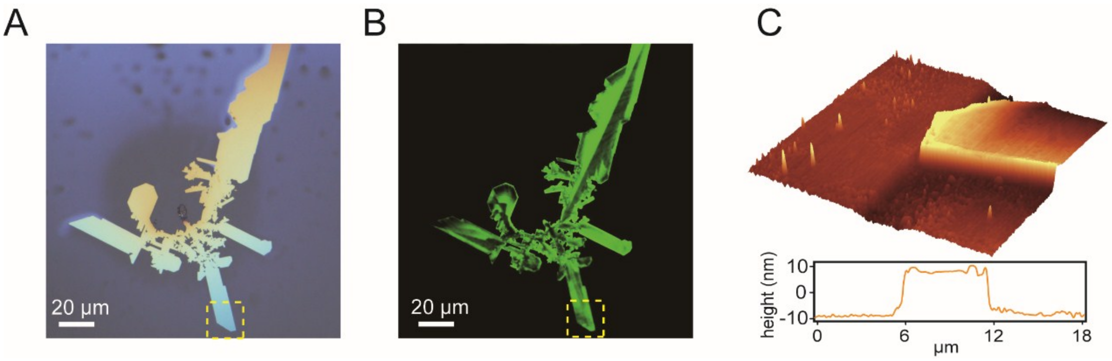

Figure S5. The liquid-phase growth of 2D $CsPbBr_3$ perovskites on the mica. (**A, B**) Optical and fluorescent images. (**C**) 3D AFM image and the high profile of the 2D $CsPbBr_3$ perovskite denoted by the yellow dashed box in (A, B).

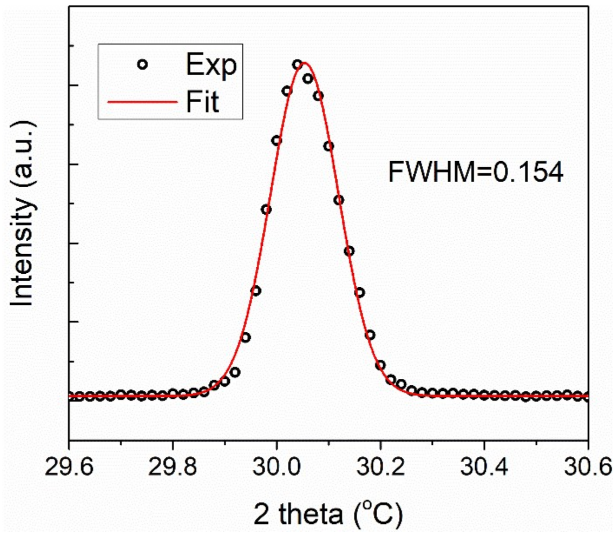

Figure S6. Rocking-curve spectrum for the (002) face of the 2D $MAPbBr_3$ perovskite on mica.

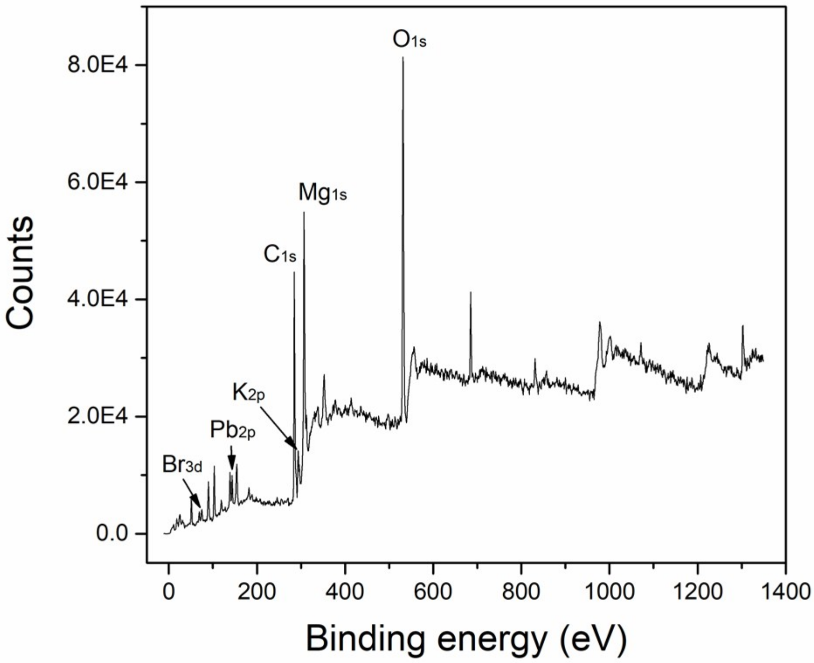

Figure S7. XPS spectrum of the 2D $MAPbBr_3$ perovskite on mica.

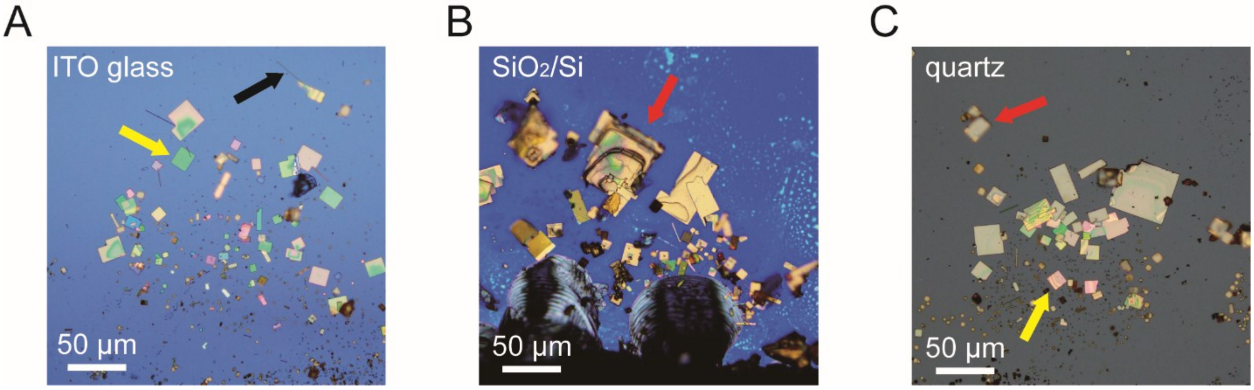

Figure S8. Perovskites grown on the ITO, SiO_2/Si and quartz substrate. (**A**) The $MAPbBr_3$ perovskites grown on the ITO glass. (**B**) The $MAPbBr_3$ perovskites grown on the SiO_2/Si. (**C**) The $MAPbBr_3$ perovskites grown on the quartz. We note that the $MAPbBr_3$ perovskites prefer to crystallize in the solution with the substrates of ITO, SiO_2/Si and quartz, which is different from the liquid-phase growth on the mica surface. The $MAPbBr_3$ perovskites with different morphologies, such as thin $MAPbBr_3$ perovskite sheets (denoted by the yellow arrows), the $MAPbBr_3$ perovskite nanowires (denoted by the black arrows), the bulk $MAPbBr_3$ crystal (denoted by the red arrows), can be found.

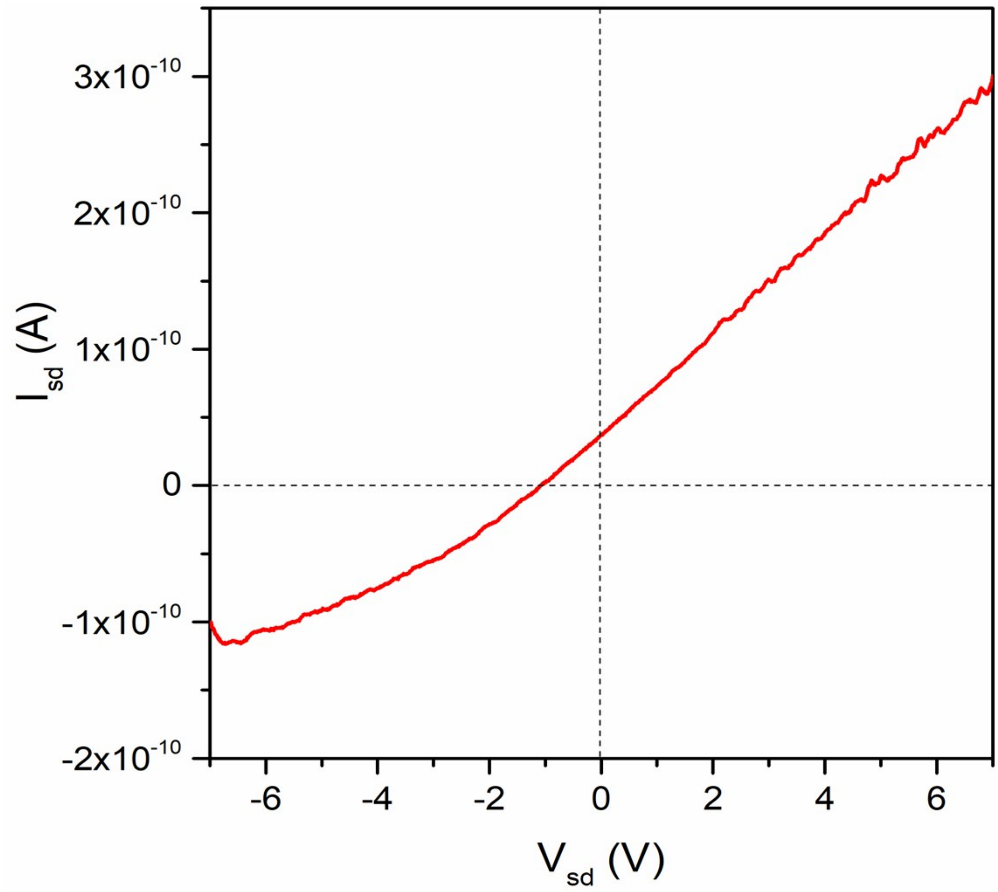

Figure S9. The *I-V* curve of a photodetector fabricated using 2D MAPbBr3 crystal grown by liquid-phase growth on mica without the OA addition.

	$MAPbBr_3$-cubic-single layer	$MAPbBr_3$-orth-single Layer	MAPbBr3-orth-single Layer
Perovskite interface absorbed with one K ion			
Interface without K ion	-121.77247161 eV		
Interface absorbed with one K ions	-121.28567324 eV		
Energy of single K ion	1.1606341 eV		
Formation energy of interface	-19.819 meV/atom		
Perovskite interface absorbed with two K ions			
Interface without K ion	-121.77247161 eV	-224.67433 eV	-244.6944
Interface absorbed with two K ions	-120.96163574 eV	-243.75209 eV	-243.74841
Energy of single K ion	1.1606341 eV	1.1606341 eV	1.1606341 eV
Formation energy of interface	-43.155 meV/atom	-20.574 meV/atom	-20.225 meV/atom

Table S1. The formation energy of the K/perovskite interface calculated by DFT

Device type	Material	Synthesis method	Responsivity (AW^{-1})	Rise/Decay time (μs)	Voltage (V)	reference
Non-layered perovskite	$MAPbBr_3$	Liquid growth	126	5.0/4.1	5	This work
	$MAPbI_3$	Liquid epitaxy	20.7	<17/-	5	[1]
	$MAPbI_3$	Two-steps	22	$<2\times10^4/<4\times10^4$	4	[2]
	$MAPbBr_3$	Solution growth	1.6×10^7	81/892	0	[3]
Layered perovskite	$(CH_3(CH_2)_3NH_3)_2(CH_3NH_3)_{n-1}PbnI_{3n+1}$	exfoliation	7.19×10^{-2}	-/-	3	[4]
	$(C_4H_9NH_3)_2(CH_3NH_3)_2Pb_3I_{10}$	Solution synthesis	1.278×10^{-2}	$1\times10^4/7.5\times10^3$	30	[5]
	$(C_4H_9NH_3)_2PbBr_4$	Solution synthesis	2.3×10^{-5}	$3.1\times10^6/3.3\times10^6$	5	[6]
	$(iBA)_2(MA)_{n-1}Pb_nI_{3n+1}$	Solution synthesis	0.117	$2\times10^4/1.7\times10^5$	1.5	[7]
Other 2D materials	MoS_2	exfoliation	7.5×10^{-3}	$<5\times10^4/<5\times10^4$	1	[8]
	MoS_2	exfoliation	880	$4\times10^6/9\times10^6$	8	[9]
	MoS_2	CVD	1.1×10^{-3}	-/-	1.5	[10]
	WS2	CVD	20	-/-	4	[11]
	Graphene	exfoliation	8.61	$>1\times10^6/>1\times10^6$	1	[12]

Table S2. Device performance comparisons of 2D $MAPbBr_3$ perovskite photodetector in this work with the reported perovskite-based photodetectors.

References

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