

# Supporting Information

## Modulation of anisotropic photoluminescence and photocurrent in $\text{PEA}_2\text{PbI}_4$ single crystal thin-films

Tao Man (满涛)<sup>1</sup>, Zicheng Li (李子成)<sup>1</sup>, Xinyu Duan (段心玉)<sup>1</sup>, Zehui Zhou (周泽慧)<sup>1</sup>, Junjie Cui (崔俊杰)<sup>1</sup>, Xiangxiang Chen (陈香香)<sup>1</sup>, Beibei Xu (许贝贝)<sup>1\*</sup>, Jianrong Qiu (邱建荣)<sup>1</sup>

<sup>1</sup>Institute of Micro & Nano Photonics, State Key Laboratory of Extreme Photonics and Instrumentation, College of Optical Science and Engineering; Zhejiang University, Hangzhou 310058, China

\*Corresponding author: [bbxu2019@zju.edu.cn](mailto:bbxu2019@zju.edu.cn)

	$\text{PEA}_2\text{PbI}_4$	
Temperature:	RT	100 K
Crystal system:	Triclinic P	Triclinic P
Lattice parameters:	a = 8.725 Å, $\alpha = 95.00^\circ$ b = 8.728 Å, $\beta = 100.14^\circ$ c = 16.591 Å, $\gamma = 90.39^\circ$	a = 8.658 Å, $\alpha = 89.65^\circ$ b = 8.665 Å, $\beta = 85.26^\circ$ c = 16.240 Å, $\gamma = 89.54^\circ$
Unit cell volume:	1250 Å <sup>3</sup>	1213 Å <sup>3</sup>

Fig. S1: The lattice parameters of the crystal at room temperature and 100 K.

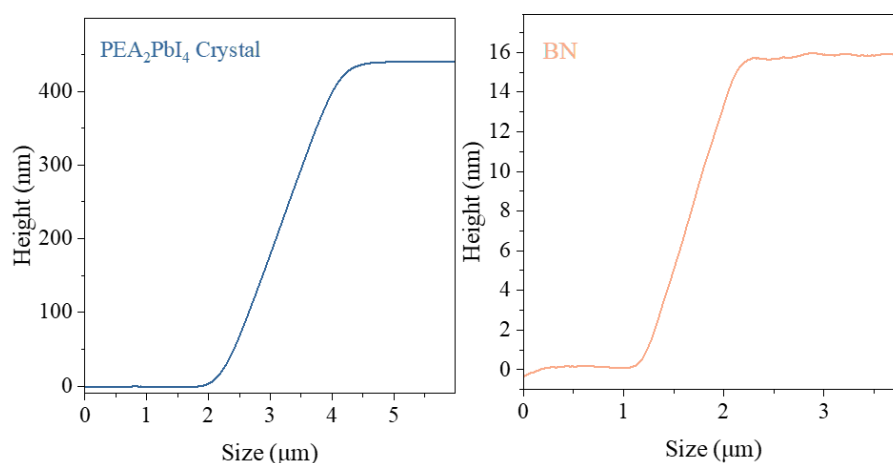


Fig. S2: The thicknesses of  $\text{PEA}_2\text{PbI}_4$  and BN.

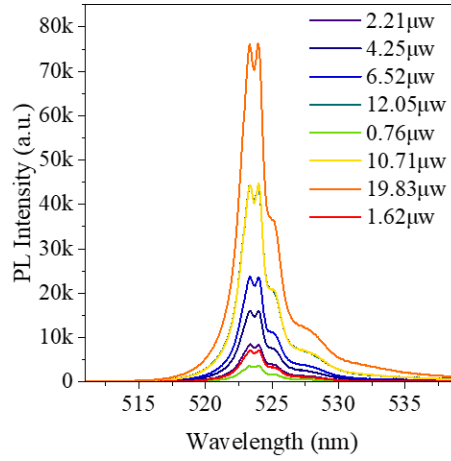


Fig. S3: The excitation power-dependent spectra of PEA<sub>2</sub>PbI<sub>4</sub> crystal.

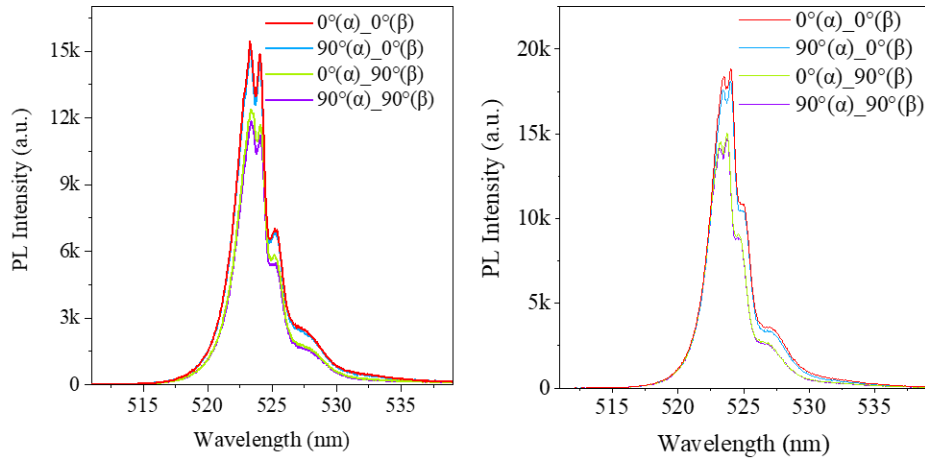


Fig. S4: Spectra of different samples at different excitation light angles (α) and detection angles (β).

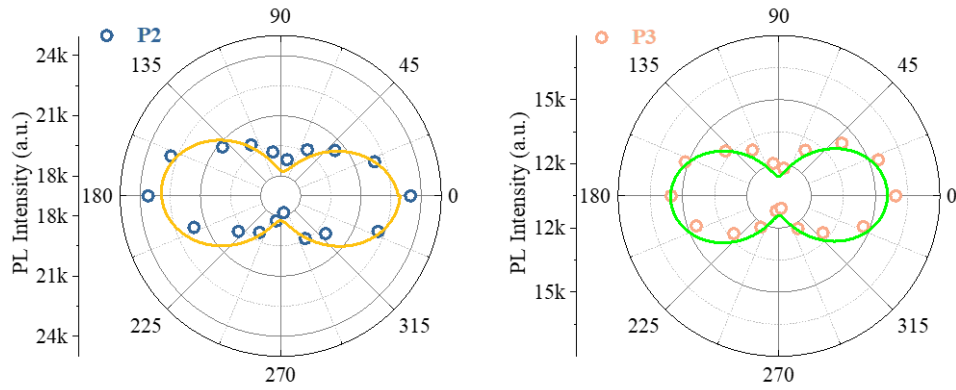


Fig. S5: Trend of P2 and P3 intensity as a function of detection angles ( $\theta$ ).

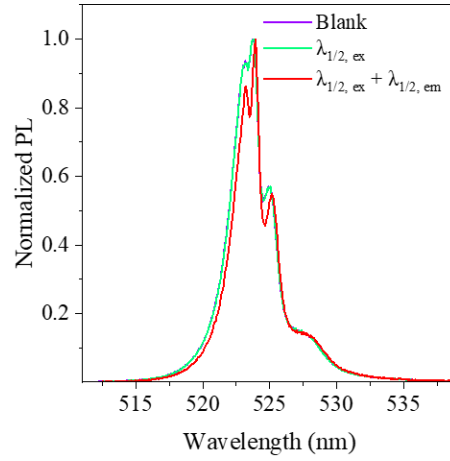


Fig. S6: PL spectrum without half-wave-plate (purple curve), PL spectrum with one half-wave-plate at the excitation light port (green curve), PL spectrum with one half-wave-plate at the excitation light port and one half-wave-plate before the emission collection system (green curve).

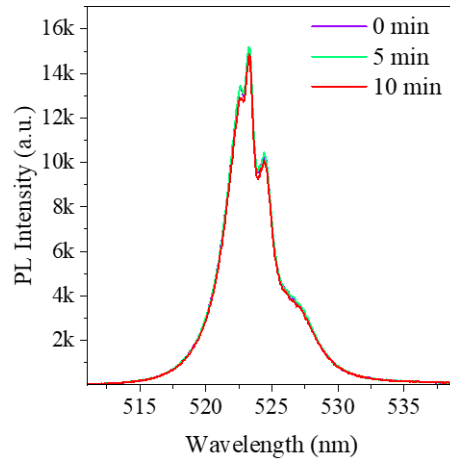


Fig. S7: The spectra changes of  $\text{PEA}_2\text{PbI}_4$  crystal in 10 min at 78 K.

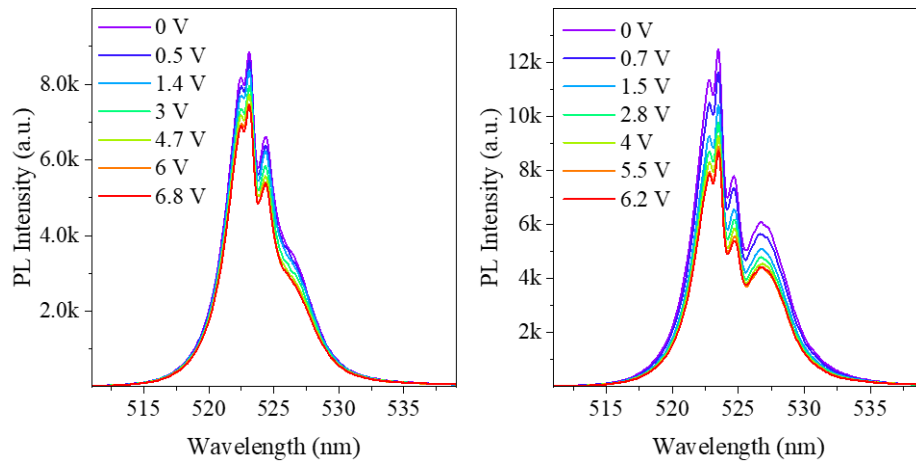


Fig. S8: Voltage-dependent PL spectra at selected detection angles  $\beta = 90^\circ$  (left) and  $\beta = 0^\circ$  (right).

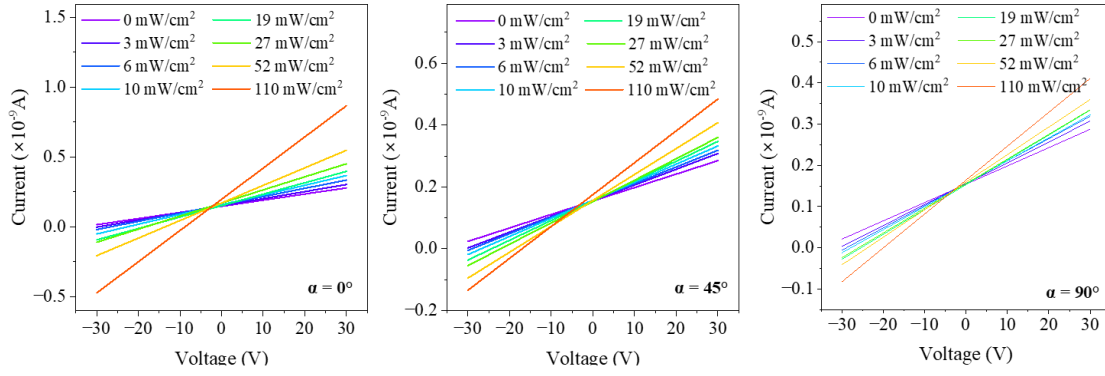


Fig. S9: Under different light intensities, the changes in photocurrent at different voltages when changing the polarization angle ( $\alpha$ ) of the excitation laser from  $0^\circ$  to  $90^\circ$ .

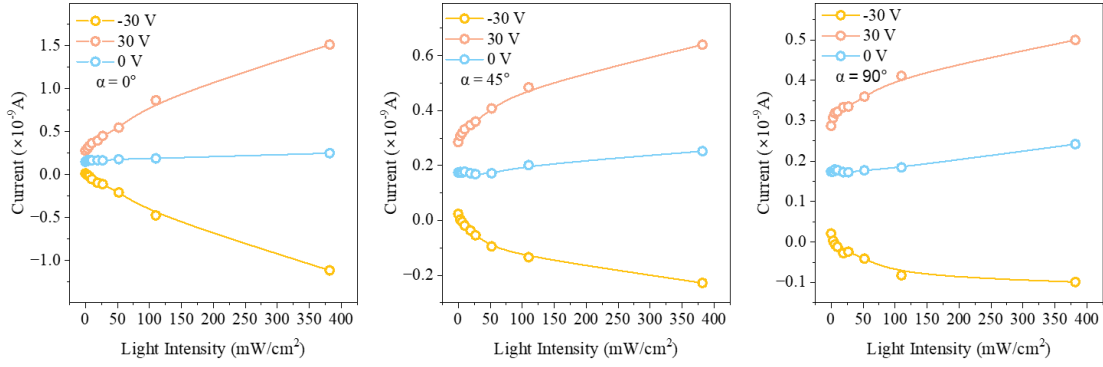


Fig. S10: Under selected voltages ( $-30$  V,  $30$  V,  $0$  V), the changes in photocurrent at different light intensities with the detection angles set as  $0^\circ$ ,  $45^\circ$ , and  $90^\circ$  (from left to right).

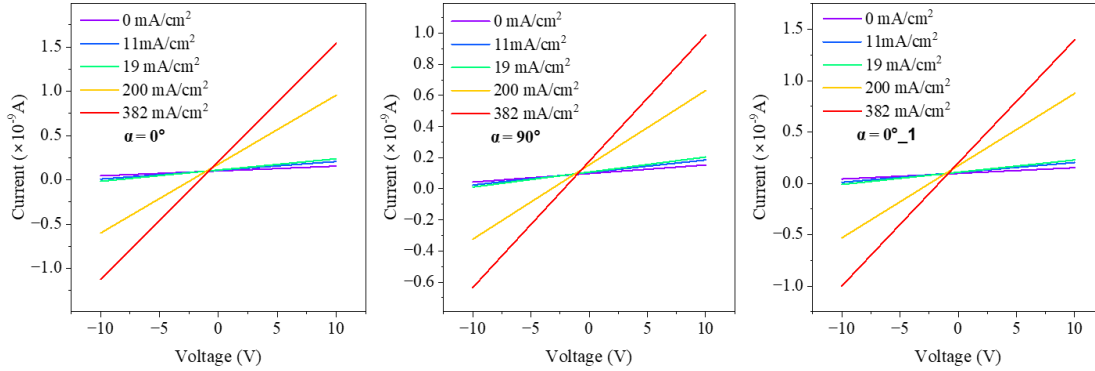


Fig. S11: Under different light intensities, the changes in photocurrent at different voltages with the detection angles set as  $0^\circ$ ,  $90^\circ$ , and  $0^\circ$  in turn (from left to right).