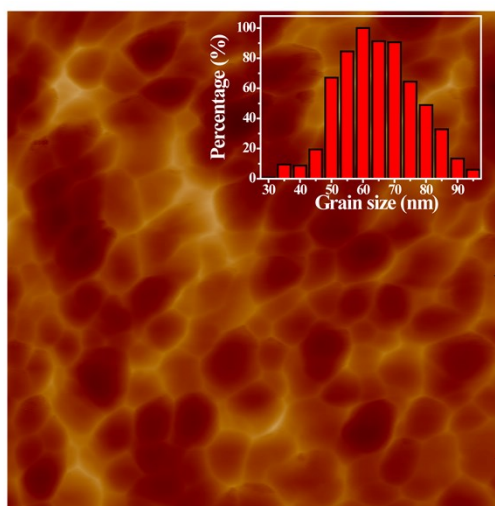


# A Fast Response, Self-Powered and Room Temperature Near Infrared-Terahertz Photodetector Based on MAPbI<sub>3</sub> /PEDOT:PSS Composite

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KEYWORDS: Perovskites, terahertz, photothermoelectric photodetector, fast response, self-powered



**Figure S1** The AFM surface morphology of MAPbI<sub>3</sub> film.

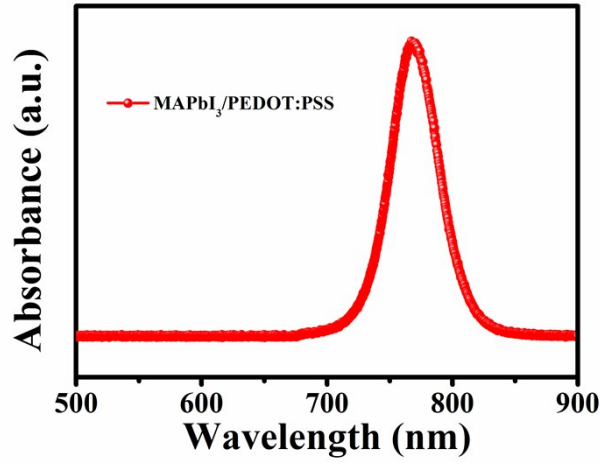


Figure S2 The PL spectra of the MAPbI<sub>3</sub>/PEDOT:PSS thin film.

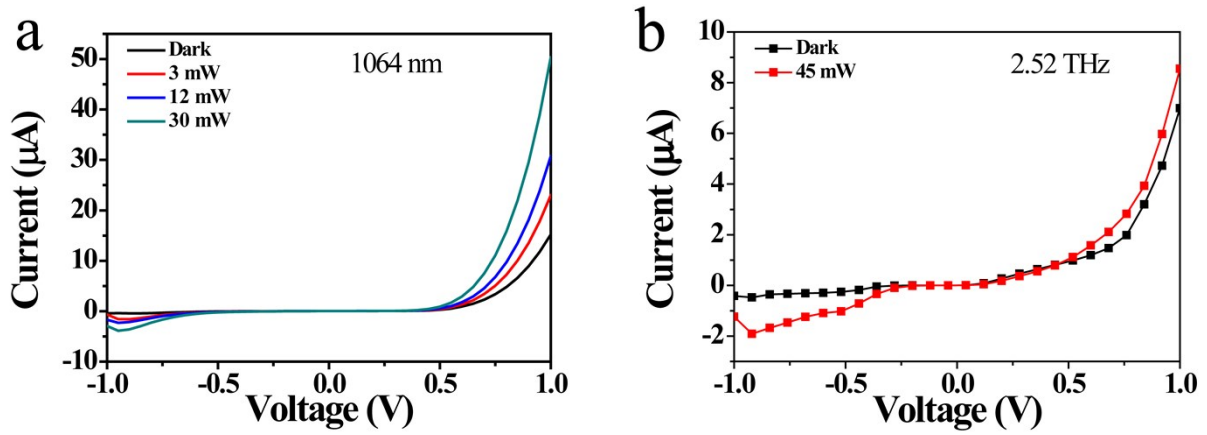


Figure S3. The  $I$ - $V$  curves in a wider range of -1 V to 1 V bias voltage.

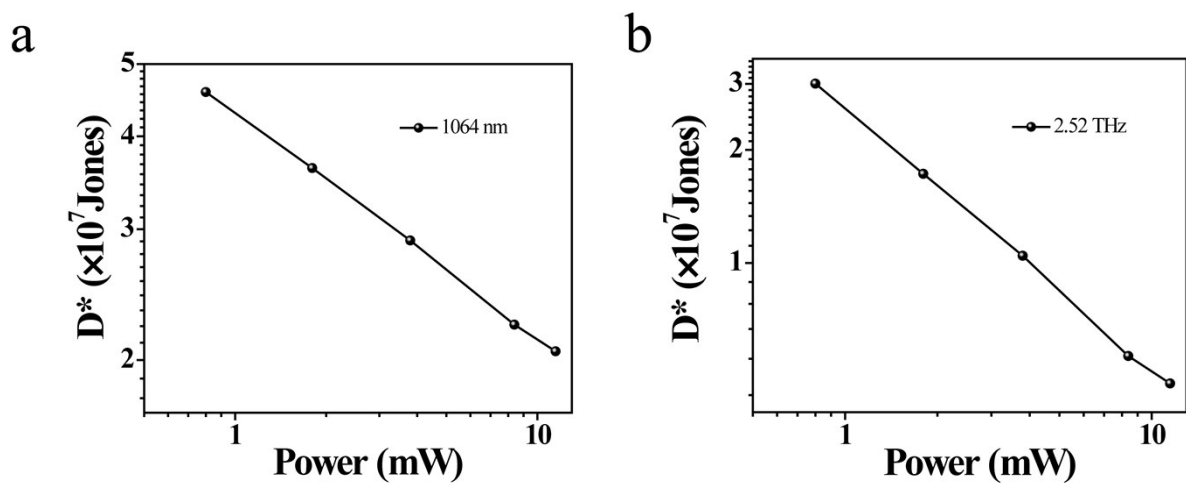
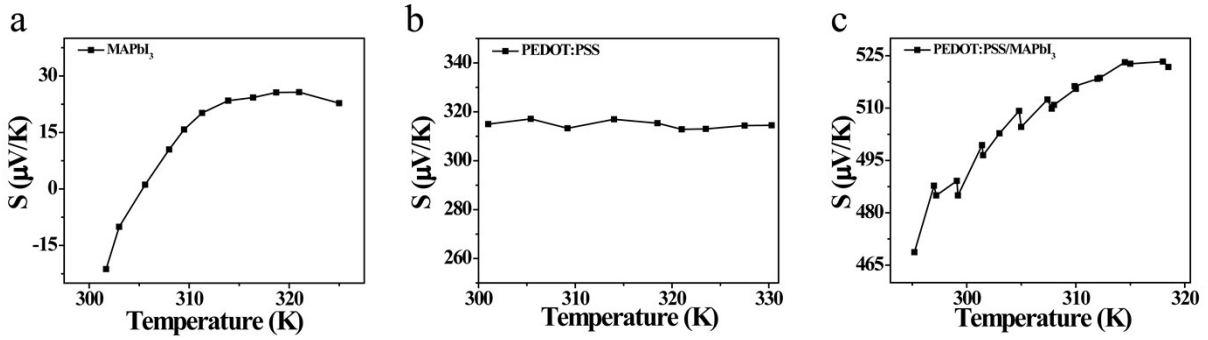


Figure S4. The  $D$  curves of the device under 1064 nm and 2.52 THz irradiation as a function of power intensity.



**Figure S5** The Seebeck coefficient of the MAPbI<sub>3</sub>, PEDOT:PSS and MAPbI<sub>3</sub>/PEDOT:PSS film.

The Seebeck coefficient of each individual component in the composite have been tested as shown in **Figure S5**. The Seebeck coefficient was measured by the self-built test system. A heating band was used to produce temperature difference of the channel. The voltage difference ( $\Delta V$ ) between the two electrodes was measured by a Keithley 2400 source meter and the temperature distribution was obtained by the Infrared thermal imager. The Seebeck coefficient was calculated as  $S = \Delta V / \Delta T$ , here  $\Delta T = T_{hot} - T_{cold}$ , meaning the temperature difference between hot side and cold side. The Seebeck coefficient of MAPbI<sub>3</sub> shows a -14~30  $\mu\text{V/K}$  with the temperature increasing from 301 to 325 K. The Seebeck coefficient of PEDOT:PSS shows a relatively high value of 316  $\mu\text{V/K}$  with the temperature increasing from 301 to 325 K. The Seebeck coefficient value of MAPbI<sub>3</sub>/PEDOT:PSS with the composite material can be attributed to the combined effect of MAPbI<sub>3</sub> and PEDOT:PSS layer and shows a high value of 465-525 $\mu\text{V/K}$ .

The seebeck coefficient of PEDOT:PSS depends on the oxidation level of PEDOT chains. The relationship between the oxidation level of PEDOT:PSS and its TE properties was recently studied by tuning the doping level of the conjugated polymer in an organic electrochemical transistor or by controlling the oxidation level of polymer thin films.<sup>1, 2</sup> Xavier's study showed that the Seebeck coefficient of the polymer increases when it is depleted of its charge carriers. The Seebeck coefficient followed exponential growth while the

electrical conductivity continued to decrease. The Seebeck coefficient reached  $\sim 400 \mu\text{V/K}$ , where the conductivity was  $\sim 0.3 \text{ S/cm}$ .<sup>1</sup> Recently, Crispin et al. reported improved TE properties of PEDOT by controlling the oxidation level of polymer thin films. The Seebeck coefficient increases by a factor of 20 on exposing the polymer to the TDAE vapour. Initially, at an oxidation level of 36%, is found to be about  $40 \mu\text{V/K}$ , finally reaching its maximum of  $780 \mu\text{V/K}$  at the lowest considered oxidation level.<sup>2</sup> As shown in Figure S5b, the Seebeck coefficient value is about  $316 \mu\text{V/K}$ . This high value could be attributed to the high oxidation level and low charge carrier concentration.

For thermoelectric (TE) material the figure of merit (ZT) is the main indicator to evaluate its performance. The ZT expressed as:  $ZT=(\sigma \cdot S^2/\kappa) T$ . Where S, T,  $\sigma$  and  $\kappa$  are the Seebeck coefficient, absolute temperature, electrical conductivity and thermal conductivity, respectively.[S1] Therefore, possessing high electrical conductivity, high Seebeck coefficient and low thermal conductivity is an ideal material system for thermoelectric devices.  $\text{MAPbI}_3$  with high conductivity and low thermal conductivity is a promising thermal material. But, its Seebeck coefficient is generally measured in a low value of  $25\text{-}35 \mu\text{V/K}$ .<sup>3</sup>

The TE performance for PEDOT:PSS have been investigated for decades years.<sup>4, 5</sup> PEDOT:PSS as novel TE materials have been considered as potential high-ZT materials because of their acknowledged low  $\kappa$ , one to two orders of magnitude below that of inorganic TE materials, and increased electrical conductivity upon doping. In addition, for PEDOT:PSS the hole-conducting dominate the hot carriers transport.

Therefore, in our  $\text{MAPbI}_3/\text{PEDOT:PSS}$  device,  $\text{MAPbI}_3$  layer acts as the electron or hole-dominated hot carriers transport layer and PEDOT:PSS layer acts as the hole-dominated hot carriers transport layer. Due to the interaction of hot carrier at  $\text{MAPbI}_3/\text{PEDOT:PSS}$  junction, thus, the Seebeck coefficient of the whole device is improved.

The Seebeck coefficient was measured by the self-built test system. Different irradiation lasers were used to produce temperature difference of the channel. The voltage difference ( $\Delta V$ ) between the two electrodes was measured by a Keithley 2400 source meter and the temperature distribution was obtained by the Infrared thermal imager. The Seebeck coefficient was calculated as  $S = \Delta V / \Delta T$ , here  $\Delta T = T_{hot} - T_{cold}$ , meaning the temperature difference between hot side and cold side. Then Seebeck coefficients of MAPbI<sub>3</sub>, PEDOT:PSS, and MAPbI<sub>3</sub>/PEDOT:PSS under 1064 nm and 2.52 THz light illumination with 0.8 mW power are shown in Table S1.

**Table S1.** Seebeck coefficients of MAPbI<sub>3</sub>, PEDOT:PSS and MAPbI<sub>3</sub>/PEDOT:PSS under different light irradiation with 0.8 mW power.

Lasers	MAPbI <sub>3</sub>	PEDOT:PSS	MAPbI <sub>3</sub> /PEDOT:PSS
1064 nm	-18 $\mu\text{V/K}$	316 $\mu\text{V/K}$	475 $\mu\text{V/K}$
2.52 THz	-20 $\mu\text{V/K}$	315 $\mu\text{V/K}$	470 $\mu\text{V/K}$

The carrier concentration ( $n$ ), Hall mobility ( $\mu_H$ ), and calculated conductivity ( $\sigma_H$ ) of the MAPbI<sub>3</sub>/PEDOT:PSS obtained by hall test system are shown in Table S2.

**Table S2.** Carrier concentrations ( $n$ ), Hall mobility ( $\mu_H$ ), and electrical conductivity ( $\sigma_H$ ) of the MAPbI<sub>3</sub>/PEDOT:PSS.

Material	$n$ (cm <sup>-3</sup> )	$\mu_H$ (cm <sup>2</sup> /V·s)	$\sigma_H$ (S/cm)
MAPbI <sub>3</sub> /PEDOT:PSS	$8.73 \times 10^{16}$	109.4	1.5

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