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- [Article \(/doi/10.1002/adma.201405116/full\)](/doi/10.1002/adma.201405116/full)
- [References \(/doi/10.1002/adma.201405116/full#references\)](/doi/10.1002/adma.201405116/full#references)
- [Cited By \(/doi/10.1002/adma.201405116/full#footer-citing\)](/doi/10.1002/adma.201405116/full#footer-citing)

# High-Gain and Low-Driving-Voltage Photodetectors Based on Organolead Triiodide Perovskites

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Weak light sensing in the ultraviolet (UV), visible, and near-infrared (NIR) range has a wide variety of applications in fields that are of importance to both industry and defense as well as scientific research.<sup>[1,2]</sup> Solution-processable optoelectronic materials, such as organic materials, nanomaterials, and nanocomposites, have shown promise as active layers in large-area, low-cost photodetectors, because they frequently provide photoconductive gain (defined as the number of charges flowing through an external circuit per incident photon). However, further improvement of their detection performance is hindered by their poor charge carrier mobilities.<sup>[3,4]</sup> Organometal trihalide perovskites (OTPs) (with the molecular structure  $\text{CH}_3\text{NH}_3\text{PbX}_3$ , where  $\text{X} = \text{Cl}, \text{Br}, \text{I}$ , or a mixed halide) are a new family of optoelectronic materials that combine good solution processability with high Hall mobilities, i.e., comparable to that of crystalline silicon. OTP-based photodetectors potentially mark a paradigm shift in the engineering of low-cost, yet high-performance photodetectors. In contrast to silicon, solution-processed OTPs are direct bandgap semiconductors with very large absorption coefficients of up to  $\approx 10^5 \text{ cm}^{-1}$  in the UV–vis range.<sup>[5]</sup> OTP solar cells show promise for competing with commercial silicon solar cells because they achieve remarkably high power conversion efficiencies of  $\approx 15\text{--}18\%$  within only four years of development.<sup>[5–8]</sup> More and more experimental evidence suggests that defects/traps may be abundant in the OTP layer, either within the bulk or at the surface.<sup>[7–11]</sup> Although these trap states are detrimental to the performance of photovoltaic devices, we show in this paper that it is possible to exploit traps to boost the performance of perovskite photodetectors with an ingenious device design.

In this paper, we describe the fabrication and characterization of solution-processed  $\text{CH}_3\text{NH}_3\text{PbI}_3$  photodetectors that combine a high photoconductive gain with a broad spectral response, ranging from the UV to the NIR. Benefitting from the trapped-hole-induced electron injection, the  $\text{CH}_3\text{NH}_3\text{PbI}_3$  photodetector works as a photodiode in the dark and shows large photoconductive gain under illumination. The maximum device gain reached  $489 \pm 6$  at a very low driving voltage of  $-1 \text{ V}$ .

The devices studied here have a layered inverted structure (Figure 1a) where indium tin oxide (ITO) is the cathode,  $\text{CH}_3\text{NH}_3\text{PbI}_3$  is the active layer, 4,4'-bis[(p-trichlorosilylpropylphenyl)phenylamino]-biphenyl (TPD-Si<sub>2</sub>) serves as the hole transporting/electron blocking layer, molybdenum trioxide ( $\text{MoO}_3$ ) is used for anode work function modification, and silver (Ag) as the anode. The  $\text{CH}_3\text{NH}_3\text{PbI}_3$  layers were prepared by thermal-annealing induced interdiffusion of the two perovskite precursors ( $\text{PbI}_2$ ,  $\text{CH}_3\text{NH}_3\text{I}$ ) by way of a method that has recently been developed in our group to fabricate very efficient solar cells with high yield.<sup>[8]</sup> In short, lead iodide ( $\text{PbI}_2$ ) films were deposited first on ITO/glass substrates by spin-coating. A second layer consisting of methylammonium iodide ( $\text{CH}_3\text{NH}_3\text{I}$ ) (hereafter MAI) was then spin-coated on top of the dried  $\text{PbI}_2$  film, followed by thermal annealing at  $105 \text{ }^\circ\text{C}$  for 60 min. Scanning electron microscopy (SEM) measurements (Figure S1, Supporting Information) showed that the interdiffusion method allows the preparation of  $\text{CH}_3\text{NH}_3\text{PbI}_3$  films on ITO that are continuous and uniform, which is particularly important for obtaining leakage-free photodetectors. The absorption curve in Figure 1b shows that the  $\text{CH}_3\text{NH}_3\text{PbI}_3$  films have a broad absorption spectrum that ranges from 300 nm (UV) to 800 nm (NIR).