Ultra-high external quantum efficiency of photomultiplication-type organic photodiodes *via* interfacial electrostatic interactions

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An electric double layer (EDL)-embedded photomultiplication-type organic photodiode (PM-OPD) is demonstrated with an unprecedentedly high external quantum efficiency (EQE) of 2,210,000%, responsivity of 11,200 A W^{-1}, specific detectivity of 2.82×10^{14} Jones, and gain-bandwidth product of 1.92×10^{7} Hz as well as high reproducibility. A polymer electrolyte, poly(9,9-bis(3'-(N,N-dimethyl)-N-ethylammoinium-propyl-2,7-fluorene)-alt-2,7-(9,9-dioctylfluorene))dibromide (PFN-Br) is employed as a work function modifying layer of ITO to construct a Schottky junction with a donor polymer semiconductor. The EDL at Schottky interface enables the stabilization of trapped electron states within the isolated acceptor domains by electrostatic interactions between the exposed cations and trapped electrons, resulting in boosting gain generation. The numerical simulations based on drift-diffusion approximation of charge carriers clearly show the effects of the EDL on the energetics of trapped electron states. The feasibility of the fabricated ultra-high EQE PM-OPD is demonstrated *via* a 10 × 10 pixelated prototype image sensor with remarkable responsivity.