Supplementary document with supporting information

Engineering Band Gap and Electronic Transport in Organic-Inorganic Halide Perovskites by Superlattices

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1. Characterization of superlattice thin films

We synthesize additional samples of the superlattice structures in layers, each of different length, on a glass film. We characterize each layer separately, as presented in Figures S-1 and S-2. Figure S-1 shows the SEM illustration of each layer including the surface and cross-sectional details. The cross-sectional images show different thicknesses for Figures S-1 (c), (f) and (i). Figure S-1(c) has a thickness of \~250 nm and it shows only layer 1. Figure S-1(f) has a thickness of \~350 nm, which is a combined thickness of layers 1 and 2. Similarly, Figure S-1(i) represents the combined thickness of layers 1, 2 and 3. Energy dispersive X-ray plots as presented in Figure S-2 show the atomic composition of each layer: (i) the presence of only Pb atoms in the first layer confirms the existence of only MAPbI\textsubscript{3} there, (ii) the second layer shows the same composition for Pb but peaks start emerging for Sn, indicating presence of MASnI\textsubscript{3} in the second layer, and (iii) the third layer has the same composition of Sn but the fraction of Pb increases, confirming that

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each layer is a perovskite thin film and they indeed form superlattices using the synthesis technique that we have described in the manuscript.

**Figure S-1:** Scanning electron microscopy (SEM) images of the thin films for the three layers of MAPbI$_3$-MASnI$_3$-MAPbI$_3$. A schematic of the sample showing the three separate layers of the superlattice is presented. The first layer is of MAPbI$_3$ thin film, layer 2 is made up of MASnI$_3$ and layer 3 is again MAPbI$_3$. (a), (d) and (g) show the surface morphology of the three layers at 5000X magnification. Similarly, (b), (e) and (h) show the surface morphology of the three layers at 15000X magnification. Cross-sectional representation for the three layers is shown in (c), (f) and (i).
2. Optical band gap of superlattice structures

We determine the optical band gap for two samples, a simple perovskite MAPbI$_3$ and the superlattice MAPbI$_3$-MASnI$_3$-MAPbI$_3$. As shown in Figure S-3, the optical band gap of the superlattice structure is smaller than that of the simple perovskite, which agrees with the computational estimate of \(~1.6\text{ eV}\). While the 1.31 eV band gap for superlattice case differs from the computational prediction of 0.91 eV, the results do confirm a qualitative decrease in the band gap of superlattice geometry as obtained by the simulations.

Figure S-3: Absorption characteristics of two thin films. The red curve shows the variation of the band gap as a function of the absorption coefficient for the superlattice, MAPbI$_3$-MASnI$_3$-MAPbI$_3$ and the blue curve represents the same for the...
MAPbI\textsubscript{3} perovskite structure. MAPbI\textsubscript{3}-MASnI\textsubscript{3}-MAPbI\textsubscript{3} assumes a band gap of 1.31 eV while MAPbI\textsubscript{3} shows a band gap of 1.6 eV.

3. Details on the two probe method of measurement

We employ a two-probe method using Keithley 230 programmable voltage source and Keithley 617 programmable electrometer for our experiments, which were performed at constant voltage source and recorded the current for voltages between 1-10 V. A schematic of the measurement device is illustrated in Figure S-4. The sample has two aluminum contacts to apply the voltage and measure the current that will flow between the contacts. Thus, the length (L) for the current flow and the corresponding orthogonal separation (W) are required for the calculations. The aspect ratio of (W/L) is set to be 10, a rather high value that ensures the perpendicular flow of current and a uniform electric field.

![Figure S-4: A schematic illustrating the measurement setup and device.](image)