Graded bandgap perovskite solar cells

Onur Ergen, S.Matt Gilbert, Thang Pham, Sally J. Turner, Mark Tian Zhi Tan, Marcus A. Worsley and Alex Zettl

1Department of Physics, University of California at Berkeley, Berkeley, California 94720, USA
2Department of Chemistry, University of California at Berkeley, Berkeley, California 94720, USA
3Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA
4Kavli Energy Nanosciences Institute at the University of California, Berkeley, and the Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA
5Physical and Life Sciences Directorate, Lawrence Livermore National Laboratory, Livermore, CA 94550, USA

Corresponding Author:
Prof. Alex Zettl (azettl@berkeley.edu)

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A) Material Characterization

Concentration variation

Figure S 1 (a) UV-visible light absorption spectra of CH$_3$NH$_3$Sn(I$_3$-x)Br$_x$ and CH$_3$NH$_3$Pb(I$_1$-xBr$_x$)$_3$, with varying iodide concentration “x”, b) Photoluminescence (PL), spectra of perovskite cells, CH$_3$NH$_3$Sn(I$_3$-x)Br$_x$ and CH$_3$NH$_3$Pb(I$_1$-xBr$_x$)$_3$, by varying iodide concentration “x”. 

Figure S 2 (a) Top view SEM image of a perovskite sample after peeling off GA layer. The line formations arise due to interfacial adhesion. (b) Top view SEM image of samples without GA and b) W/O GA, (c) Bandgap changing by time with and without GA layer. (d) EDAX line mapping for oxygen signature of a perovskite with and without GA modification.
The role of the graphene aerogels

**Figure S 2** Photoluminescence analysis of perovskite cells in air (only \(\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Br}_x\)). (a) without GA. (b) with GA modification. (c) Bandgap changing by time with and without GA layer. (d) EDAX line mapping for oxygen signature of a perovskite with and without GA modification. Graphene aerogel encapsulation acts as a barrier for oxygen penetration and moisture ingress.

**Figure S 3** (a) Top view SEM image of a perovskite sample after peeling off GA layer. The line formations arise due to interfacial adhesion. (b) Top view SEM image of samples without GA improvement. The scale bars in the SEM images are 5µm.
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Figure S 4
XRD diffraction patterns of the perovskite layers of a) W/ GA and b) W/O GA.

Figure S 5
Cross sectional SEM-EDAX analysis of perovskite cells (a) EDAX signal for cell with h-BN, over the area outlined by red box in the inset SEM image. (b) Line mapping of cell with h-BN modification (dashed line indicates the position of h-BN). The scan is along the vertical red line (from top to bottom) shown in the inset SEM image. (c) Line mapping of cell without h-BN modification. The scan is along the red vertical line (from top to bottom) shown in the inset SEM image. Scale bar for inset of a) and b) is 200nm; scale bar for inset of c) is 100nm.
The role of the h-BN

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B) Electrical Characterization

Stability under illumination

Figure S 6 a) Time dependent Current density (solid lines) and Voc (dashed lines) is shown. The cells without h-BN and GA exhibit faster degradation under constant illumination compared to the complete cell with h-BN and GA. (solid lines is J_sc and dashed lines is V_oc ) b) Power conversion efficiency of the cells with h-BN and GA (red), cell w/h-BN and W/O GA (black), W/O h-BN and W/O GA (green). Complete cells show a very stable behavior under constant illumination. Even though a decrease was observed in the current density, there is a constant increase in open circuit voltage indicating that efficiency becomes stable with time.
The role of the graphene aerogels on mobility

Figure S 7 Hall effect measurement. The mobility plotted against the annealing temperature of double layered perovskite cells (re-crystallization temperatures).
**Figure S 8** J-V curves for 21.7% PCE graded band gap perovskite cell with (red) and without (blue) light illumination.
**Figure S 9** (a) Reverse and forward sweep (<0.01 V/s) J-V for a typical graded band gap perovskite device. (b) Histogram of solar cell efficiencies with reverse and forward sweep, after 1h illumination in air.
Figure S 10 Ohmic contact behavior illustrated by current-voltage (I-V) plots. The GaN contact paths are made from Ti/Al/Ni/Au (30/100/20/150 nm).
Figure S 11 Near infrared photoluminescence (NIR-PL) spectra of graded band gap perovskite solar cells, with both h-BN and GA modifications. Under constant illumination an additional PL peak forms near 1300nm and grows with increasing light intensity.
Figure S12 (a) J-V measurement of MASnI₃ based solar cells with and without GA. Devices prepared with GA show better stability in air. (All devices prepared in air). (b) J-V measurement of MASnI₃ based devices which are fabricated by following the same procedures as shown in refs. [5] and [13], Type I and Type II respectively. Type I and Type II cells have the similar architecture (FTO/d-TiO₂/mp-TiO₂/MASnI₃/spiro-OMeTAD/Au), but different ETL, HTL and Au thicknesses. The table shows the detailed comparison of our cells prepared and cells reported in the literature.

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<th>Name</th>
<th>Jsc (mA/cm²)</th>
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<td>49</td>
<td>5.36</td>
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**Figure S 13** (a) External quantum efficiency of the champion cell with integrated photocurrent (thick black line). Maximum possible $J_{sc}$, if the QE is 100% over the spectrum, is 49.4mA/cm$^2$ and the expected $J_{sc}$ is 42.32mA/cm$^2$. The EQE spectrum for reference silicon cells is also shown under A.M 1.5. (b) The plot of reflective absorption and internal quantum efficiency (IQE) versus wavelength. The inset shows the composition profile and approximate band diagram of the cell.
**Figure S 14** Back surface pits on the GaN surface after etching. The cells display excellent light trapping properties due to these textured surface properties.
**Figure S 15** Mott–Schottky analysis for the GaN/CH$_3$NH$_3$SnI$_3$ interface. The dotted line is the linear fit to experimental data. The doping density of the perovskite film is found to be $1.4 \times 10^{17}$ cm$^{-3}$. The inset shows the cross sectional SEM image of the GaN/CH$_3$NH$_3$SnI$_3$ device (scale bar is 50nm). The depletion width within the perovskite is calculated to be ~115nm.