Supporting Information

Introducing Cu_2O Thin-Films as a Hole-Transport Layer in Efficient Planar Perovskite Solar Cell Structures

*Soumyo Chatterjee and Amlan J. Pal**

Department of Solid State Physics, Indian Association for the Cultivation of Science, Jadavpur, Kolkata 700032, India

AUTHOR EMAIL ADDRESS: sspajp@iacs.res.in

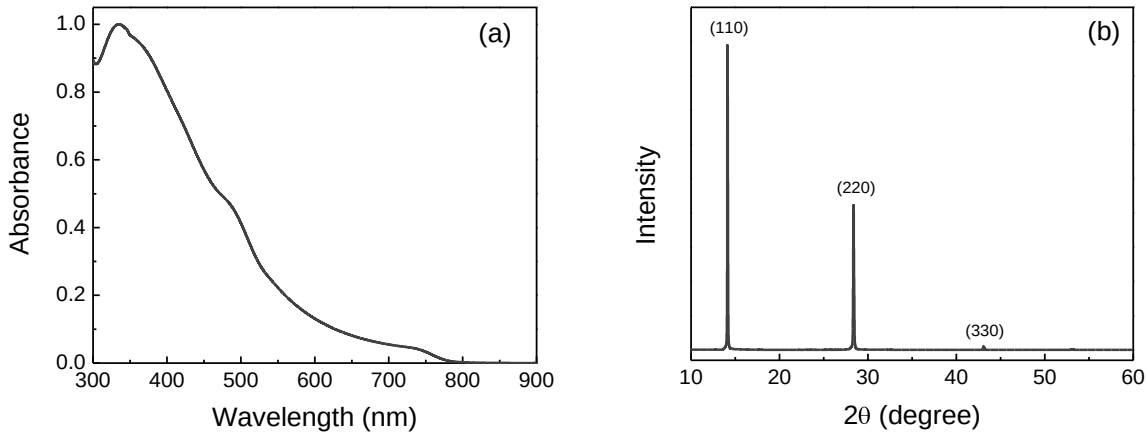

Figure S1. (a) Optical absorption spectrum and (b) XRD pattern of $CH_3NH_3PbI_{3-X}Cl_X$ perovskite thin film.

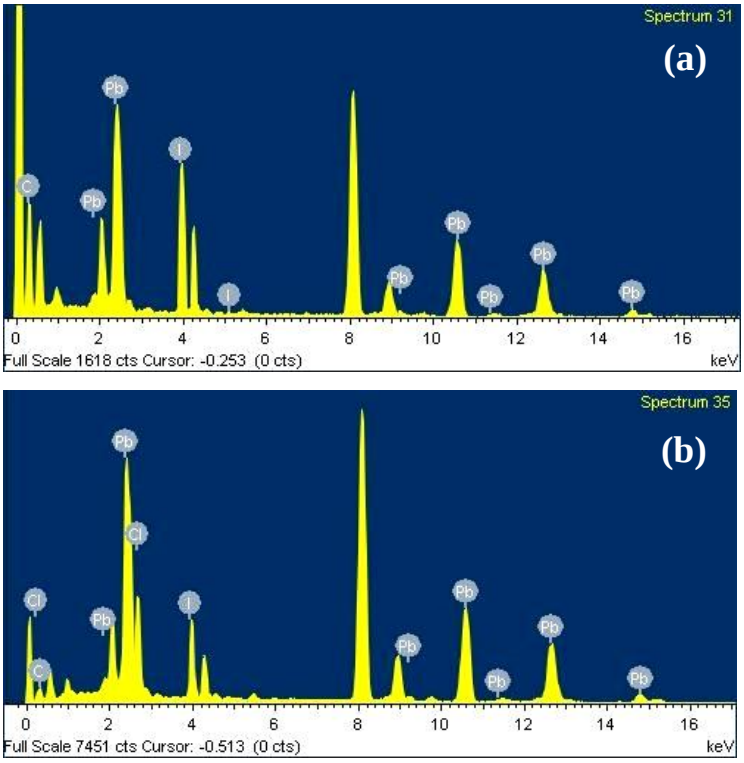

Figure S2. EDX spectra of (a) $CH_3NH_3PbI_3$ and (b) $CH_3NH_3PbI_{3-X}Cl_X$ perovskites.

Growth of NiO and Cu@NiO thin-films

To grow NiO thin films, 0.4 M solution of nickel acetate tetrahydrate in 20 mL methanol was prepared. Diethanolamine (0.4 M) was then added drop-wise to this light-green colored homogeneous nickel acetate solution. The color of the solution change form to deep-green and finally a blue tint appeared when no more diethanolamine was added ensuring formation of $Ni(OH)_2$. This solution was spun on a substrate to form thin-films which were annealed at 425 °C for 15 min to form uniform and strongly adherent NiO thin film.

To introduce copper ions in NiO structure, a stoichiometric (5 at%) amount of copper chloride dihydrate was added in the nickel acetate tetrahydrate solution in methanol. The rest of the procedure was kept unaltered.

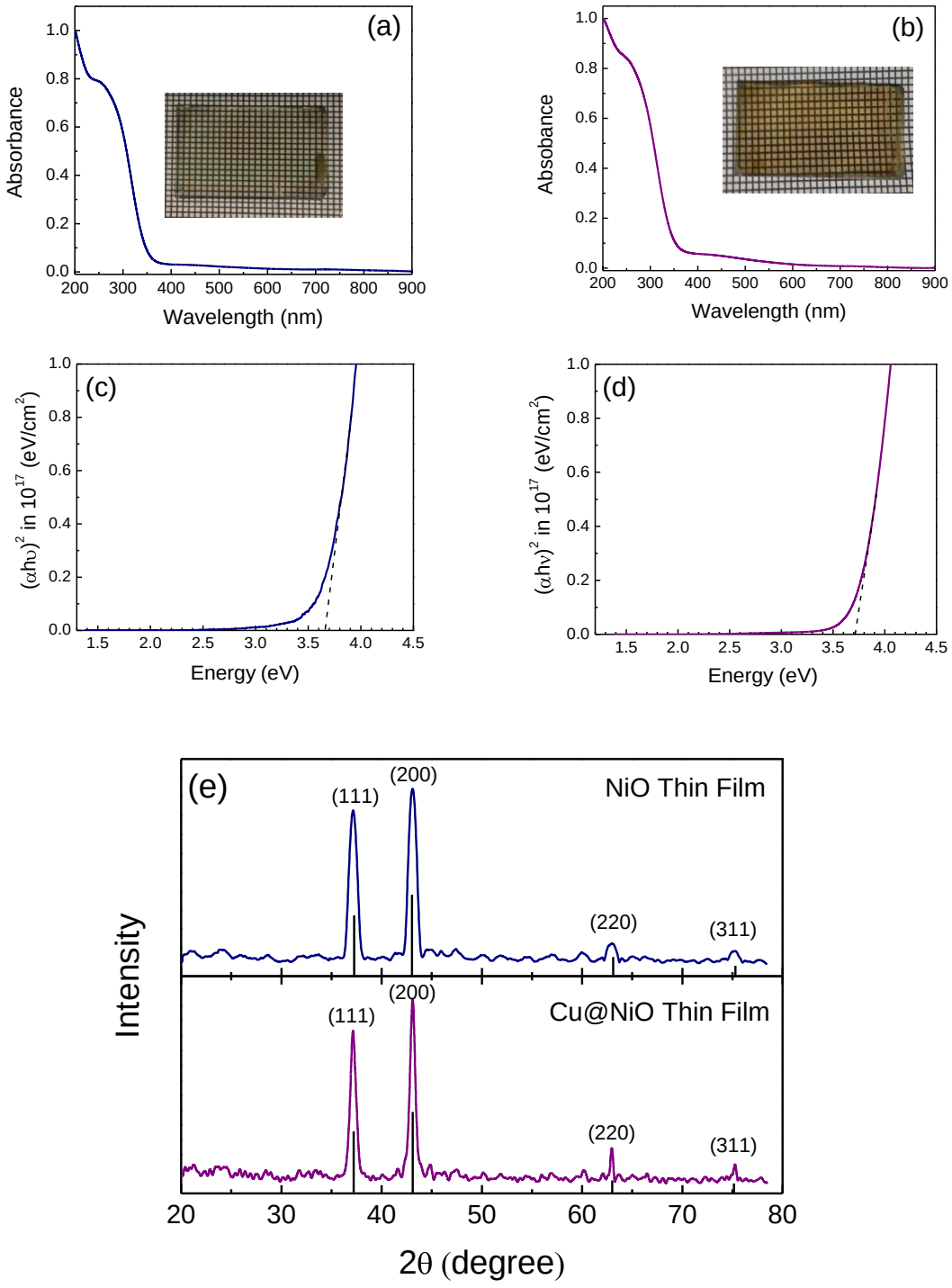

Figure S3. (a & b) Optical absorption spectra and (c & d) Tauc plot of NiO and Cu@NiO thin films. (e) XRD pattern of NiO and Cu@NiO thin films. Inset of (a) and of (b) show photograph of the respective films.

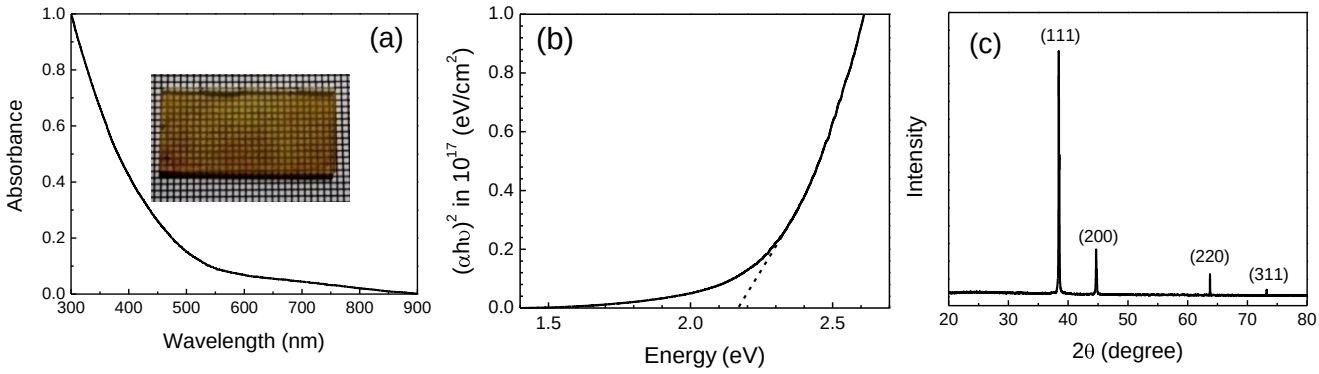

Figure S4. (a) Optical absorption spectrum, (b) Tauc plot, and (c) XRD pattern of Cu_2O thin film formed through SILAR technique. Inset of (a) shows a photograph of the film.

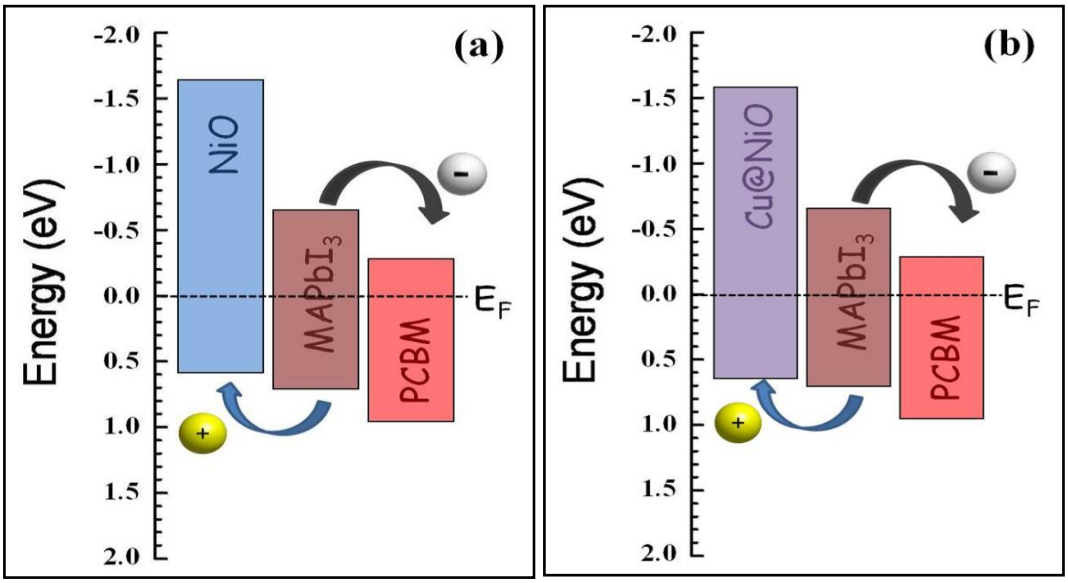

Figure S5. Energy band-diagrams of the (a) NiO/$MAPbI_3$/PCBM and (b) Cu@NiO/$MAPbI_3$/PCBM device architectures. The dashed line represents the Fermi energy after contact.

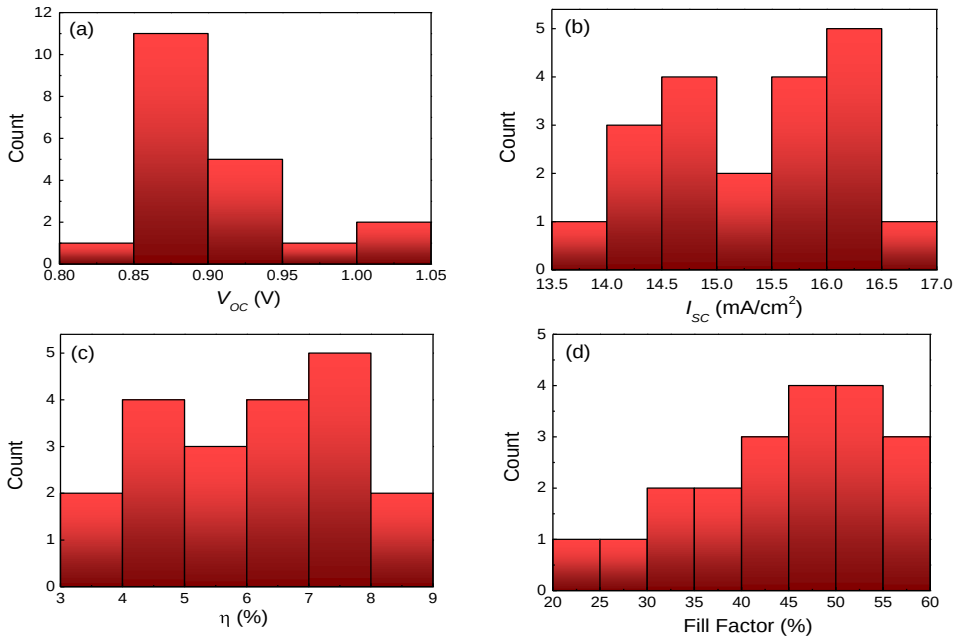

Figure S6. Histogram of (a) V_{OC}, (b) I_{SC}, (c) η, and (d) fill-factor of $Cu_2O/MAPbI_3/PCBM$ solar cells.

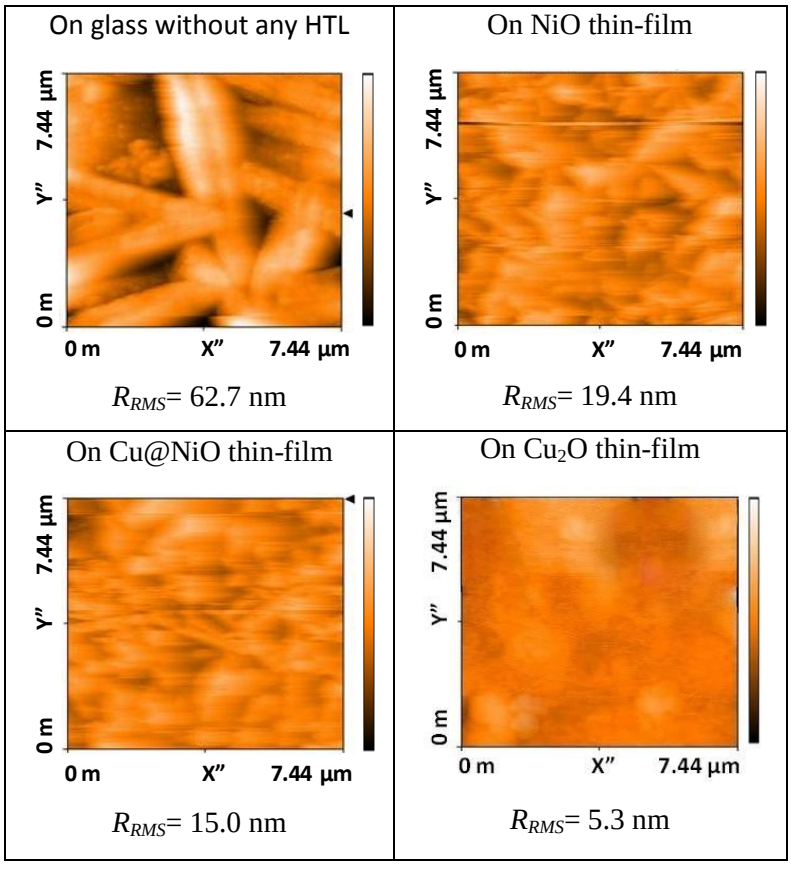

Figure S7. Surface morphologies of the $MAPbI_3$ perovskite films deposited on different HTLs along with their RMS roughnesses.

The complete author list of some references:

(9) Wang, H.; Sheikh, A. D.; Feng, Q. Y.; Li, F.; Chen, Y.; Yu, W. L.; Alarousu, E.; Ma, C.; Haque, M. A.; Shi, D.; Wang, Z. S.; Mohammed, O. F.; Bakr, O. M.; Wu, T. Facile Synthesis and High Performance of a New Carbazole-Based Hole-Transporting Material for Hybrid Perovskite Solar Cells. *ACS Photonics* **2015**, *2*, 849-855.

(13) Hu, L.; Peng, J.; Wang, W. W.; Xia, Z.; Yuan, J. Y.; Lu, J. L.; Huang, X. D.; Ma, W. L.; Song, H. B.; Chen, W.; Cheng, Y. B.; Tang, J. Sequential Deposition of $CH_3NH_3PbI_3$ on Planar NiO Film for Efficient Planar Perovskite Solar Cells. *ACS Photonics* **2014**, *1*, 547-553.

(23) Kim, H. S.; Lee, C. R.; Im, J. H.; Lee, K. B.; Moehl, T.; Marchioro, A.; Moon, S. J.; Humphry-Baker, R.; Yum, J. H.; Moser, J. E.; Gratzel, M.; Park, N. G. Lead Iodide Perovskite Sensitized All-Solid-State Submicron Thin Film Mesoscopic Solar Cell with Efficiency Exceeding 9%. *Sci Rep* **2012**, *2*, 591.