

Electronic Supplementary Information to

Nanocrystalline solar cells with an antimony sulfide solid absorber by atomic layer deposition

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Experimental methods

All preparative steps apart from the coating with Sb_2S_3 were performed according to previously published procedures.^{1,2} The active surface area of the TiO_2 nanocrystals was measured by gas physisorption in a Micromeritics ASAP 2020. A fit of the experimental data to the Brunauer-Emmett-Teller adsorption isotherm yielded a specific surface area of $16 \text{ m}^2/\text{g}$. The TiO_2 nanocrystals were deposited in layers of $\sim 2 \mu\text{m}$ thickness onto commercial fluorine-doped SnO_2 (TCO) transparent conducting glass slides from Asahi Glass previously coated with a 40-nm thick compact TiO_2 film by spray pyrolysis at 420°C in air,³ and annealed to 450°C . Subsequently, the ALD of amorphous antimony sulfide ($\alpha\text{-Sb}_2\text{S}_3$) was performed in exposure mode at 120°C in a home-made hot-wall reactor from antimony tris(dimethylamide) and hydrogen sulfide, both purchased from Sigma Aldrich.⁴ *Caution:* hydrogen sulfide is highly toxic! Hydrogen sulfide gas was delivered at room temperature from a lecture bottle equipped with a pressure regulator designed for handling corrosive gases. Antimony tris(dimethylamide) was kept as a liquid at 40°C in a high-vacuum steel cylinder from Swagelok. The pulse, exposure, and purge times were 1.5 s, 60 s, and 60 s for $\text{Sb}(\text{NMe}_2)_3$ and 0.2 s, 60 s, and 60 s for H_2S , respectively. A chemically resistant four-stage membrane pump MV10C by Vacuubrand was used, and unreacted H_2S was removed from the exhaust by a bed of Sulfatreat Select HP pebbles purchased from MI Swaco. After annealing at 315°C for 30 minutes on a hotplate in a glovebox under nitrogen, a LiSCN treatment was performed from 0.5 M LiSCN aqueous solution, after which the CuSCN hole conductor was infiltrated from dipropylsulfide solution according to the published procedure.⁵ Finally, 40 nm thick gold contacts were evaporated onto the backside of the system. The cells were characterized electrically with on a home-made device based on a Potentiostat type DT-2101 from Hi-Tek Instruments, England and Yokogawa 7651 as a DC source in the dark and under an AM1.5 solar simulator (sulfur lamp type Fusion Lightning S1000) at 1000 W m^{-2} , after 2 hours of irradiation. For the photovoltaic characterization the irradiated area was defined by a mask of area $9 \text{ mm} \times 6 \text{ mm}$. Spectroscopic ellipsometry was performed on a SENpro from Sentech using the (n,k) spectra previously published.^{4a} Transmission electron micrographs were recorded on a Jeol 2100F at 200 kV. Diffuse optical absorption spectroscopy was measured using a DH-2000-L light source, an HR4000 spectrometer, and an ISP-50-8-R integrating sphere from OceanOptics. For each sample, a diffuse transmission spectrum was recorded with respect to a blank taken without sample, then a diffuse reflectance spectrum was recorded with respect to a blank taken on a white standard (smooth Teflon sample, WS-1-SS from OceanOptics). In both measurements, the front face of the photovoltaic sample was presented facing the light source. The diffuse absorption spectra displayed in the article were obtained by subtracting the transmitted and reflected intensities from the incident intensity.

- 1 G. Larramona, C. Choné, A. Jacob, D. Sakakura, B. Delatouche, D. Péré, X. Cieren, M. Nagino, R. Bayón, *Chem. Mater.* **2006**, *18*, 1688-1696.
- 2 S.; Nezu, G. Larramona, C. Choné, A. Jacob, B. Delatouche, D. Péré, C. Moisan, *J. Phys. Chem. C* **2010**, *114*, 6854-6859.
- 3 C. J. Barbé, F. Arendse, P. Comte, M. Jirousek, F. Lenzmann, V. Shklover, M. Grätzel, *J. Am. Ceram. Soc.* **1997**, *80*, 3157.
- 4 (a) R. B. Yang, J. Bachmann, M. Reiche, J. W. Gerlach, U. Gösele, K. Nielsch *Chem. Mater.* **2009**, *21*, 2586-2588; (b) R. B. Yang, N. Zakharov, O. Moutanabbir, K. Scheerschmidt, L.-M. Wu, U. Gösele, J. Bachmann, K. Nielsch *J. Am. Chem. Soc.* **2010**, *132*, 7592-7594.
- 5 B. O'Regan, F. Lenzmann, R. Muis, J. Wienke, *Chem. Mater.* **2002**, *14*, 5023-5029.