Improving the light-harvesting of thin-film solar cells with photochemical upconversion

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Current solar cells are fundamentally limited by their inability to harvest photons with energies less than the absorber optical threshold. Thin-film solar cells such as a-Si:H or organic solar cells, usually having band gaps above the optimum single-band gap value of 1.34 eV given by the Shockley-Queisser limit, and a much smaller volume to absorb the light, are particularly prone to this loss mechanism and thus suffer from imperfect usage of the solar spectrum. An elegant way towards overcoming this limitation and using a larger fraction of the incident light is the re-shaping of the solar spectrum by upconversion (UC) of photons. Photochemical upconversion, as realized by triplet-triplet annihilation (TTA) in organic molecules [1], operates efficiently from the near infrared and red part of the solar spectrum to the yellow or green, with measured quantum efficiencies of up to 30% [2].

Recently, we have demonstrated TTA-upconversion to improve the light harvesting of amorphous silicon and organic bulk heterojunction solar cells [3, 4, 5], cf. Fig. 1. Employing a moderate concentration of 19-30 suns, and a simple optical coupling scheme between solar cell and upconverting unit based on silvercoated glass beads acting as a back reflector, we demonstrated a relative gain of up to 12% in solar cell quantum efficiency around 700 nm and a relative overall efficiency increase of up to 0.2%. Although the demonstrated current increase is still about two orders of magnitude below device-relevant levels, a clear advantage of the TTA-UC process as



Fig. 1: (a) Spectral lineup of upconverter absorption and emission characteristics. (b) EQE and transmission of a-Si:H solar cell. (c) Upconversion signature in the spectral response of a-Si:H solar cell, and model for spectral response of upconverter.

compared to the commonly employed lanthanide-based upconversion strategies [6] was demonstrated. Thus, TTA-upconversion is proven to be a highly promising concept for the augmentation of high-bandgap thin-film solar cells.

In the present talk we will review these recent results on a-Si:H and organic solar cells, and then focus on recent strategies, aiming to increase the photocurrent gain by TTA-UC to device-relevant levels, which we are currently investigating:

The first step consists in 1) exploiting micro-optical means for the concentration of light in the upconverting layer, which we have found to increase the solar cell photocurrent due the to inherently non-linear response of the UC yield on the illumination conditions. Our strategy is based on microembossed back reflector



Fig. 2: (a) Microstructured back reflector (metal-coated hot-embossed PTFE film). (b) Focussed-ion-beam (FIB) slices verified the spherical shape of the indentations. (c) Ray tracing simulations were used to model the focusing effect, and - in combination with rate equations describing the TTA upconversion - to predict the impact on the upconversion yield.

designs facilitating focusing of light in the upconverter, and at the same time enhanced outcoupling of the upconverted light. Proof-of-principle experiments were performed with metal-coated structured PFTE films (spherical indentations as obtained by hot-embossing of silica beads into the PTFE film) and have shown an advantage of upconverting units with structured back sides as compared to flat reflectors, while simulations promise an up to 9-fold gain for an optimized structure and optical configuration.

2) The second step is the transfer of the TTA-process into a solid-state medium, which will allow for a significant increase of the TTA yield – and thus the UC efficiency – by concentrating the organic chromophores in a much smaller volume. Previously presented strategies employing blended polymer films or -nanoparticles and have shown promising upconversion yields [7, 8, 9], but no device application has been presented so far. We will report on a novel strategy employing nano-sized support structures, to which the TTA-performing molecules are attached. Recent results, employing this nanostructured upconverter in an assembly with a thin-film solar cell, will be presented, again by an *in-situ* approach measuring the impact on the spectral response of the solar cell.

REFERENCES

- [1] Baluschev, S., Miteva, T., Yakutkin, V., Nelles, G., Yasuda, A., and Wegner, G. Phys. Rev. Lett. 97, 143903 (2006).
- [2] Cheng, Y., Khoury, T., Clady, R. G. C. R., Tayebjee, M. J. Y., Ekins-Daukes, N. J., Crossley, M. J., and Schmidt, T. W. Phys. Chem. Chem. Phys. 12, 66–71 (2010).
- [3] Cheng, Y. Y., Fückel, B., MacQueen, R. W., Khoury, T., Clady, R. G. C. R., Schulze, T. F., Ekins-Daukes, N. J., Crossley, M. J., Stannowski, B., Lips, K., and Schmidt, T. W. *Energy Environ. Sci.* 5, 6953–6959 (2012).
- [4] Schulze, T. F., Czolk, J., Cheng, Y. Y., Fückel, B., MacQueen, R. W., Khoury, T., Crossley, M. J., Stannowski, B., Lips, K., Lemmer, U., Colsmann, A., and Schmidt, T. W. J. Phys. Chem. C, DOI: 10.1021/jp309636m (2012).
- Schulze, T. F., Cheng, Y. Y., Fückel, B., MacQueen, R. W., Danos, A., Davis, N. J. L. K., Tayebjee, M. J. Y., Khoury, T., Clady, R. G. C. R., Ekins-Daukes, N. J., Crossley, M. J., Stannowski, B., Lips, K., and Schmidt, T. W. Aust. J. Chem. 65, 480–485 (2012).
- [6] de Wild, J., Rath, J. K., Meijerink, A., van Sark, W. G. J. H. M., and Schropp, R. E. I. Sol. Energ. Mat. Sol. C. 94, 2395–2398 (2010).
- [7] Islangulov, R. R., Lott, J., Weder, C., and Castellano, F. N. J. Am. Chem. Soc. **129**, 12652–12653 (2007).
- [8] Monguzzi, A., Frigoli, M., Larpent, C., Tubino, R., and Meinardi, F. Adv. Funct. Mater. 22, 139 (2012).
- [9] Kim, J.-H., Deng, F., Castellano, F. N., and Kim, J.-H. Chem. Mater. 24, 2250–2252 (2012).