Multi-exciton generation via Singlet Fission – In search of charge separation in Tetracene/c-Si hybrid solar cells

Martin Liebhaber[a], Rowan W. MacQueen[b,c], Murad J.Y. Tayebjee[d], Tim F. Schulze[a], Jens Niederhausen[a], Clemens Gersmann[a], Jan Behrends[e], Timothy W. Schmidt[b], and Klaus Lips[a,e]

[a]*Helmholtz-Zentrum Berlin, Energy Materials In-Situ Laboratory Berlin (EMIL), Institute for Nanospectroscopy, Albert-Einstein-Straße 15, 12489 Berlin, Germany*

[b]*School of Chemistry, The University of Sydney, Sydney, NSW 2006, Australia*

[c]*School of Chemistry, University of New South Wales, Sydney, NSW 2052, Australia*

[d]*School of Photovoltaic and Renewable Energy Engineering, University of New South Wales, Sydney, NSW 2052, Australia*

[e]*Berlin Joint EPR Lab (BeJEL) Fachbereich Physik, Freie Universität Berlin, Arnimallee 14, 14195 Berlin, Germany*

Email: lips@helmholtz-berlin.de

Fundamental losses of conventional single band gap crystalline silicon (c-Si) solar cells restrain their theoretical conversion efficiency to ~29%[1] (Shockley & Queisser). A promising multi-exciton generating device concept to overcome this limit is to reduce thermalization losses by taking advantage of the singlet fission (SF) process in organic tetracene (Tc) layers implemented on top of conventional c-Si absorbers. In such a device structure, high energy photons deposit their energy in the Tc film as singlet excitons. Immediately, with an efficiency close to 100%, one singlet is then converted via SF into two triplets of lower energy.[2] Subsequent to their generation, the triplet excitons have to be dissociated in order to contribute to the overall current and therefore significantly enhance the efficiency of the device.

Triplet exciton dissociation has been shown at organic-organic interfaces between SF-materials and C_{60},[3,4] as well as at organic-inorganic interfaces between SF-materials and Pb nanocrystals.[5,6] No triplet dissociation at the hybrid interface to c-Si has been reported to date.

We could successfully integrate thermally evaporated Tc thin films in (n-doped) c-Si based solar cell structures (efficiency of ~10%), allowing to combine quantum efficiency results on device level with triplet dynamic studies of the functional interfaces by means of ultrafast photoluminescence. In the delayed fluorescence we clearly observe long lived triplet states, although no significant quenching due to triplet exciton dissociation and charge separation at the interface is detected. However, these long-lived triplet states cannot be detected in transient or cw electron paramagnetic resonance (EPR) experiments. Nevertheless, a strong light-induced triplet EPR signal is observed when Tc is incorporated in low concentration in a solid matrix. This indicates that the triplet sublevels of both singlet-born triplet-pair constituents are populated symmetrically and that the triplets quickly dissociate in the Tc film.

During quantum efficiency measurements various device conditions (e.g. temperature, external bias) and additional interlayers were tested to further assess whether the dissociation of geminate triplet excitons in the Tc layer and a charge transfer across the hybrid interface into c-Si is a feasible scheme to enhance the conversion efficiency of silicon based photovoltaics.
With our results, we will discuss reasons, which hamper an efficient charge separation at the Tc/c-Si hybrid interface. Possible alterative device designs will be discussed.

References:

[1]W. Shockley, and H. J. J. Queisser, Appl. Phys. **32**, 510 (1961);

[2]S. Singh *et al.*, J. Chem. Phys. **42**, 330 (1965);

[3]D. N. Congreve *et al.*, Science **340**, 334 (2013);

[4]P. J. Jadhav *et al.*, Nano Lett. **11**, 1495 (2011);

[5]N. J. Thomson *et al.*, Nat. Mater. **13**, 1039 (2014);

[6]M. Tabachnyk *et al.*, Nat. Mater. **13**, 1033 (2014).