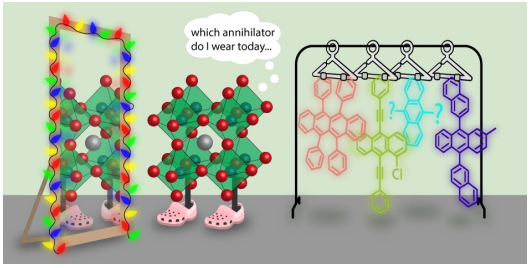
Generating Spin-Triplets at the Perovskite/Organic Interface

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Triplet generation at a hybrid inorganic/organic semiconductor interface is a very promising approach to increase the (photo-)excited state recombination lifetime, and thus, facilitate solar energy harvesting. One of the possible applications using the generated spin-triplet excitons is photon upconversion. Photon upconversion describes the process of shortening the wavelength of the light emitted after irradiation, resulting in a net gain in photon energy. Here, upconversion occurs by combining multiple low energy photons to a single high energy photon through triplet-triplet annihilation. Since direct optical excitation of triplet states is 'spin-forbidden', sensitizers are required to indirectly populate the triplet state by energy or charge transfer.

Currently, triplet sensitizers span a broad range of material classes including metal-organic complexes, nanomaterials, and bulk perovskite films. Understanding the fundamental energy transfer mechanism is crucial for the advancement of optoelectronic devices based on this process. The exact triplet sensitization mechanism varies depending on several factors including: (i) the absolute alignments of the sensitizer and acceptor energy levels. (ii) The exciton binding energy in the sensitizer, resulting in excited states in form of excitons or free carriers. (iii) The local trap density. Here, I will present the current understanding of charge transfer at the perovskite/organic interface resulting in triplet generation interrogated by a combination of scanning probe microscopy and optical spectroscopy. Of particular interest are the nanoscale properties of the perovskite and how surface treatments or added stressors influence the fundamental optoelectronic properties of the perovskite both on the ensemble and at the nanoscale. [1,2]



[1] Sullivan, C.M.; Bieber, A.S.; Drozdick, H.K.; Moller, G.; Kusyznski, J.E.; VanOrman, Z.A.; Wieghold, S.; Strouse, G.F.; Nienhaus, L., Surface Doping Boosts Triplet Generation Yield in Perovskite-Sensitized Upconversion. *Adv. Opt. Mater.*, **2022**, 11, 2201921.

[2] Sullivan, C.M.; Nienhaus, L., Generating Spin-Triplet States at the Bulk Perovskite/Organic Interface for Photon Upconversion. *Nanoscale*, **2023**, 15, 998-1013