Triplet-Triplet Annihilation Mediated Photon Upconversion in Confined Nano-domains of Biopolymer-Surfactants-Chromophores Co-Assembly

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Triplet-triplet annihilation mediated photon upconversion (TTA-UC) materials are emerging for applications in biology to drive non-invasive low energy excitation based biochemical processes, and in photovoltaics (PV) to avoid transmission losses.[1] However, realization of efficient TTA-UC materials in aqueous environments and solid-state faces issues of chromophores aggregation and deactivation of excited triplets by dissolved oxygen molecules. We overcame these issues by developing a generalized approach of biopolymer surfactant-chromophore coassembly where TTA-UC chromophores are confined inside surfactant nano-domains coated with thick fiber networks of biopolymers in hydrogels or bioplastic frameworks (Fig. 1). Consequently, air-stable green to blue TTA-UC with high UC efficiency of 13.5% and a long annihilator triplet lifetime of 4.9 ms was achieved in the hydrogel. [2] Interestingly, drying of this hydrogel in air resulted in phase separation of liquid surfactant domains containing chromophores inside the transparent solid bioplastic film. The film showed even higher UC efficiency of 15.6 % in air with unprecedented durability of two years. [3] Further, to expand the photon harvesting window of bioplastics, red to blue TTA-UC bioplastics with record UC efficiency of 17.6 % and proof-of-concept Far-red to blue bioplastics were developed. [4] The key is two-fold. First, biopolymer and the surfactant self-assemble in water to give a developed hierarchical structure with hydrophobic domains which accommodate chromophores up to high concentrations to drive efficient triplet energy transfer and TTA. Second, thick hydrogen-bonding networks of biopolymer backbone prevent O_2 inflow to the interior chromophores region, as evidenced by long triplet lifetimes both in hydrogels (4.9 ms) and bioplastic films (1.6 to 2.5 ms). Later, the developed NIR to blue hydrogel was successfully applied for optogenetic genome engineering of light responsive hippocampal gene to induce neuronal dendrite growth with unconverted blue light upon excitation with NIR light in vitro. [5]

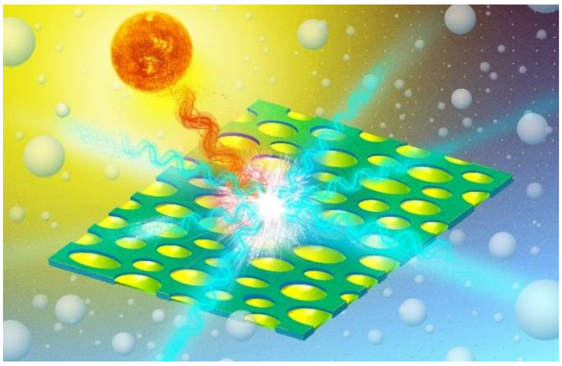

Fig. 1. Air Stable triplet-triplet annihilation mediated photon upconversion in confined nano-domains

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