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Design requirements imposed on advanced solar cell concepts by sparse solar photon flux

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Third generation photovoltaics concepts such as upconversion and hot carrier solar cells aim to use more of the solar spectrum than is possible in a conventional single junction device. Upconversion generally involves two (or more) low energy excitations resulting in a single higher energy excitation via energy transfer upconversion (ETU). The limiting efficiency of a single junction solar cell with an upconverter under 1-Sun illumination is about 47%, and about 63% under full concentration [1]. Hot carrier solar cells (HCSCs), first proposed by Ross and Nozik [2], extract carriers via energy selective contacts while maintaining them above the lattice temperature. They thereby use the excess kinetic energy of charge carriers excited by photons with energy above the bandgap, extracting work from the hot electron gas akin to a Carnot engine. In this case, the limiting efficiency is about 66% under 1-Sun illumination, and 85% under full concentration [3]. These concepts are part of a group of several advanced solar cell concepts that have been an intense focus of research because their limiting efficiencies are higher than that described by detailed balance in a single junction solar cell [4].

One aspect that differentiates upconversion and hot carrier solar cells from other advanced concepts is that they involve energy transfer processes that require the interaction of multiple excitations, making them intensity dependent. Downconversion, on the other hand, only requires a single high energy excitation that is converted to two low energy excitations. While there are mechanisms to upconvert using a single species using nonlinear processes such as two photon absorption, they are very inefficient and require extremely high fluences. Incoherent processes such as ETU are more efficient but require interaction between excitations in the form of resonant energy transfers [5]. Carrier-carrier equilibriation is key for HCSC operation as it allows for energy redistribution and continual repopulation of the contact energy for extraction [6]. It involves an exchange of energy that could involve some high energy carriers transferring their energy to low energy carriers, but also several low energy carriers resulting in high energy carriers. These can be thought of as continuous downconversion and upconversion events, and the importance of each is determined by the position of the energy selective contact, with a higher energy contact requiring more upconversion events.

These energy transfer processes need to occur within an interaction range and the lifetime of the carriers. Once an excitation is produced, it needs another excitation within that spatial and temporal range for an interaction to occur. This means that the excitation rate needs to be sufficiently high enough allow for these events to occur frequently enough for efficient operation. However, if the incident photon flux is too sparse, the average number of interactions might be too low to attain the promised high efficiencies. When one considers solar irradiation, the total solar photon flux arriving on the Earth is of the order ~10^{21} photons m^{-2} s^{-1}. Even though this is a seemingly large flux, the average number of photon absorbed within the interaction volume of an excitation can be small. This average number is denoted as $\langle n_p \rangle$.

This problem is aggravated in the case of nanotechnology, where the spatial scale is limited by the physical size of the device and/or its components. Metal nanoparticle hot carrier solar cells (Metal NP HCSCs) for instance need a thermalised hot carrier population formation in individual nanoparticles. Lanthanide doped upconverting nanoparticles (Ln doped UCNP) and semiconductor upconverting quantum dots (SC UCQDs) similarly need multiple excitations in nanostructures. The absorption cross-section dictates the absorbed photon flux in such structures. This, along with the relevant timescales such as electron cooling time (for metal NP HCSCs) or decay time control the potential efficiency of these devices. This can similarly be done for bulk systems such as triplet-triplet annihilation schemes, but it is to be noted that the ability of molecules to diffuse in solution results in a larger effective length scale. We surveyed the literature [6-15] to obtain relevant length

and time scales to calculate $\langle n_p \rangle$ for these concepts. Figure 1 shows the results obtained assuming 1000-Sun illumination, taken as a practical upper limit to solar concetration.

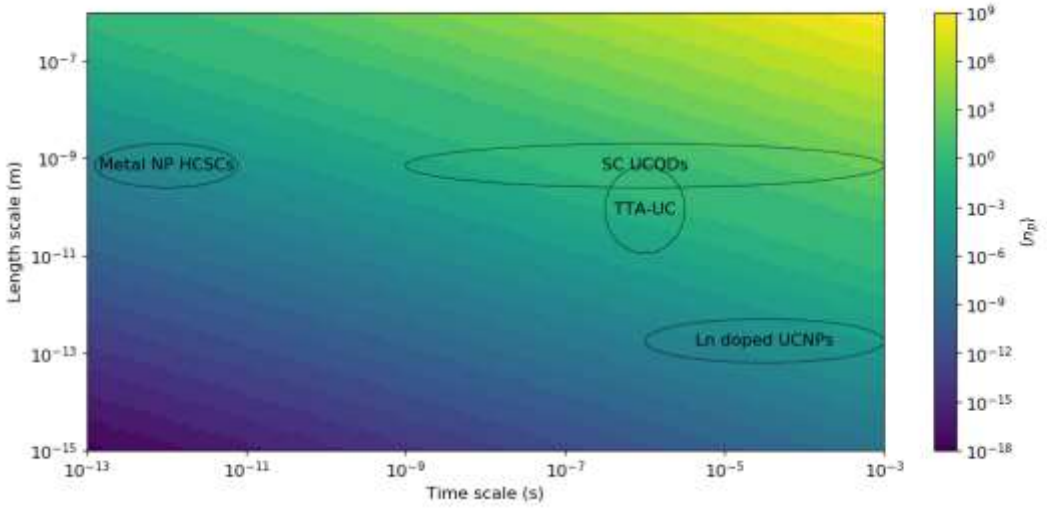

Figure 1. Average number of photons absrobed in interaction volume, $\langle n_p \rangle$, for the concepts investigated: metal nanoparticle hot carrier solar cells (Metal NP HCSCs), semiconductor upconverting quantum dots (SC UCQDs), Lanthanide doped upconverting nanoparticles (Ln doped UCNPs), and triplet-triplet annihilation upconversion (TTA-UC) under 1000-Sun illumination. Note that the x-y scales and colourbar are all logarithmic. The coloured bands are divided according to order of magnitude.

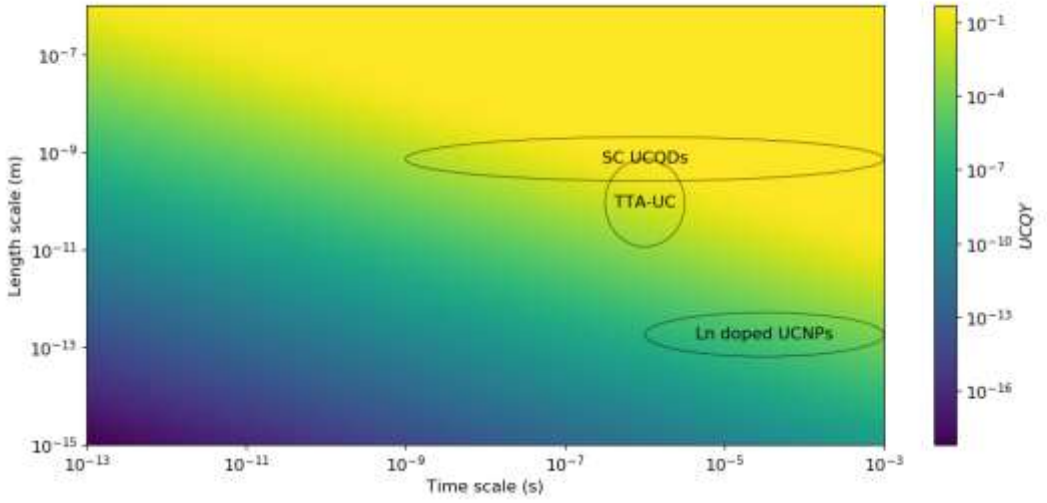

Figure 2. Limiting Upconversion Quantum Yield (UCQY) for the upconversion concepts investigated: semiconductor upconverting quantum dots (SC UCQDs), Lanthanide doped upconverting nanoparticles (Ln doped UCNPs), and triplet-triplet annihilation upconversion (TTA-UC) under 1000-Sun illumination. Note that the x-y scales and colourbar are all logarithmic.

We use an idealised upconversion model to estimate the quantum yield of the concepts considered. The model assumes that any intermediate energy transfer processes involved are instantaneous and are 100% efficient. The Sun is taken to be a blackbody at temperature 5800 K and the entire solar spectrum is considered to be able to contribute to upconversion. The

absorption cross-sections are also assumed to be independent of wavelength. While this is not true in most real systems, the model is used as an indication for where the upper limits of such a system lie. These assumptions enable a simple calculation of the upconversion quantum yield for a given excitation rate and decay rate. The excitation rate is determined by the photon flux and the absorption cross section, while the decay rate is the inverse of the lifetime. This information is used to calculate limiting upconversion quantum yields (UCQYs). UCQY is the ratio of the average number of high energy photons emitted over the number of low energy photons absorbed. The results are plotted in Figure 2 for 1000-Sun illumination. The UCQY can be seen to saturate to its maximum possible value of 0.5 (since at least 2 low energy photons are required to produce a high energy photon) for long time and length scales. Our results from Figures 1 and 2 indicate that Lanthanide-doped upconverting nanoparticles and metal nanoparticle HCSCs present challenges for practical application across all solar concentrations. Lanthanide ions suffer from having low absorption cross sections, while the rate at which electrons cool in metal nanoparticles is very high. These results emphasise the importance of sensitiers with high absorption coefficients and impose some restrictions on the maximum required decay times for efficient operation of upconverting nanotechnologies.

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