Si nanocrystals for solar shapers

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The challenge of efficient photovoltaic conversion follows directly from the “asymmetry” of the process: on the *photonic* side, the solar spectrum presents a broad range of energies, while on the *electronic* side, electrons extracted from a (single-junction) solar cell have all the same specific potential, defined by the energy gap of the active layer. Therefore the absorbed photons convert on a one-photon-to-one-electron-hole-pair basis, regardless of their initial energy, while the low-energy solar photons escape absorption and are lost entirely. In order to realize a substantial efficiency enhancement in photovoltaics, it is necessary to develop schemes addressing the part of energy which is lost in “conventional” PV conversion. A particularly attractive option with potential to realize a significant efficiency enhancement of solar cells is offered by “shaping” of the solar spectrum, so as to make it fit better with solar cell characteristics. In order to realize that, procedures for efficient down-conversion (“cutting”) of high-energy photons from the UV range of the solar spectrum, and upconversion (“pasting” together) of the low-energy photons in the near IR, need to be developed. For that purpose nanostructured materials are frequently investigated. Compared to bulk materials, nanostructures pose interesting properties induced by quantum confinement, such as (i) bandgap tuning, (ii) lower density of states with the consequent discretization of energy bands, (iii) reduction of hot carrier cooling by phonon scattering (“phonon bottleneck”), (iv) enhanced surface-related effects, (v) enhanced Coulomb interaction between carriers, and (vi) relaxation of momentum conservation, among others. The latter is especially impactful in case of Si due to its indirect bandgap. Since Si is the major PV material, this makes also Si nanostructures interesting for PV applications [1]. In particular, it turns out that layers of high-quality Si nanocrystals (Si NCs) in SiO_2 offer attractive opportunities for photon conversion in solar shapers.

Photon down-conversion

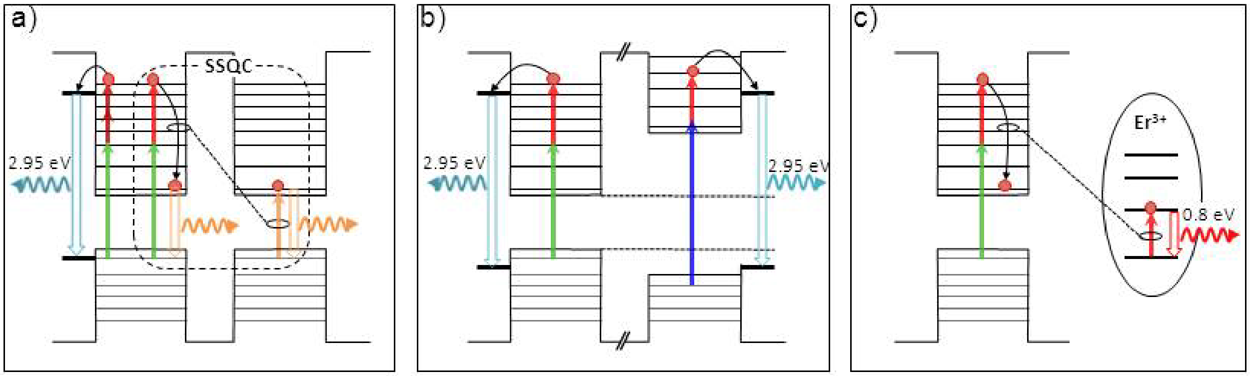
Originally, photon cutting has been demonstrated in rare-earth systems and finds wide-spread application in, e.g., fluorescent tubes. A similar idea involves carrier multiplication by generation of multiple electron-hole pairs by a single high-energy photon absorbed in a NC, where its efficiency is enhanced by a stronger Coulomb interaction. This process of *multiple exciton generation* (MEG) has been observed for NCs of different semiconductor materials. In the past, we have reported on an interesting variation of the MEG process for Si NCs – the space-separated quantum cutting (SSQC). In this case multiple excitons generated by a single high-energy photon reach their ground states not in the same, but in neighboring NCs [2,3]. The SSQC effect adds an additional virtue to the MEG: It considerably increases the excitonic lifetime above the picosecond range characteristic for multiexcitonic Auger processes, and allows for radiative recombination, and hence emission of photons with a lower energy. In that way photon “cutting” is realized, with a potential for application in solar shapers for the high energy end of the solar spectrum.

Photon up-conversion

In addition to the SSQC, our investigations of optical properties of Si NCs/SiO_2 revealed also features which open perspectives for “pasting” of IR photons with sub-bandgap energies, which are lost in a standard PV cell. The most significant of these are:

- Direct bandgap-related “hot” emission [4].
- Prominent “blue” emission band at ~420 nm, possibly (interface-) defect related.
- Efficient Auger interaction, with the characteristic time constant of ~20 ps for two excitons in a Si NC of 4.5 nm diameter [5].
- Increased excited state absorption in the near-infrared range, exceeding the linear band-to-band absorption [6].
- Reduction of hot carrier cooling rate and effective Auger “recycling” of hot carrier energy [4].

Based on the above findings, specific schemes towards up-conversion of low-energy photons using thin layers of Si NCs in SiO_2 can be proposed – see figure below. In panel (a) initial absorption of a photon with an over-bandgap energy creates free carriers. Subsequently these can be excited into higher-lying states undergoing intraband transitions induced by (multiple) absorption of low-energy photons. In this way sequential photon absorption creates hot carriers. Generation of hot carriers could then be followed by carrier multiplication through SSQC process and emission at the bandgap energy, or trapping at a defect state and the “blue” emission by the defect-related band at ~420 nm. This system could form the physical basis of a solar shaper streaming the incoming solar photons into two channels defined by the two emission bands – the excitonic one and the defect-related. Moreover, since the 420 nm wavelength of the hot emission is fixed, then the total absorption spectrum of the layer can be tailored by changing the NC size – see panel (b). A different approach is illustrated in panel (c), where Er^{3+} dopants are introduced into the system. In this case, the proposed scheme makes use of efficient excitation of Er^{3+} ions by intraband cooling of hot carriers [7]; in that way the hot carrier excess energy is converted into a monochromatic stream of photons at ~1.5 μm.



The concept of solar shapers based on thin layers of solid dispersions of Si NCs in SiO_2 offers an attractive alternative approach for reaching high-efficiencies in photovoltaics, while building upon the solid foundation of highly developed Si-cased photovoltaics.

References:

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