Photon up-conversion in the presence of non-radiative recombination

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Several ideas have been presented under the heading "Third generation photovoltaics" which can, in principle, improve the efficiency of solar cells above the Shockley-Queisser limit of 30% for non-concentrated sun light. The up-conversion of useless small energy photons into useful higher energy photons is one of them[1]. In this process photons with energy smaller than the band gap are transmitted by the solar cell and are then absorbed by an up-converter placed behind the solar cell. These photons generate electron-hole pairs of nearly twice the photon energy by a two-step excitation process, with a first photon from the valence band to an intermediate level and with a second photon from the intermediate level to the conduction band. Electrons in the conduction band can recombine with the holes in the valence band to produce large energy photons which are emitted towards the solar cell where they generate additional electron-hole pairs. Recombination of the electrons and holes in the up-converter can, however, equally well proceed via the intermediate level, whereby the generation process is exactly reversed and two small energy photons, useless for the solar cell, are emitted.

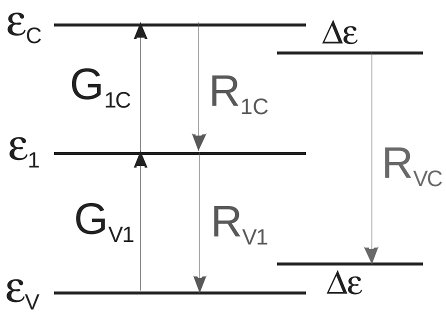

Fig.1 For photon up-conversion, electrons in the conduction band (ε_C) and holes in the valence band (ε_V) are generated by the absorption of two small energy photons in a 2-step transition. Recombination can occur via the intermediate level or after thermalization by $\Delta\varepsilon$ directly to the valence band, whereby a high energy photon is emitted.

Figure 1 shows a 3-level system which can be turned into a 5-level system by allowing electrons and holes to thermalize to an energy which is lower by $\Delta\varepsilon$. All transitions are assumed to be purely radiative. Radiative transitions between the thermalized states and the intermediate level are assumed to be forbidden for symmetry reasons. To ensure good absorption properties for the absorption of the small energy photons the level at ε_1 should be half occupied and is therefore chosen to be half donor-like and half acceptor-like. The band gap of the model system is 2.3 eV and the intermediate level is in the middle of the band gap. Figure 2 shows the quantum efficiency for the emission of high energy (thick line) and low energy photons (broken line) as a function of the incident energy current j_E of small energy photons. It is seen that an up-conversion process is not supported by a 3-level system ($\Delta\varepsilon = 0$), where nearly all electron-hole pairs recombine via the intermediate level. The emission of high energy photons becomes the dominant process after thermalization of both the electrons and the holes by $\Delta\varepsilon = 0.2$ eV.

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[1] T. Trupke, M. Green, P. Würfel, J. Appl. Phys. **92** (2002) 4117

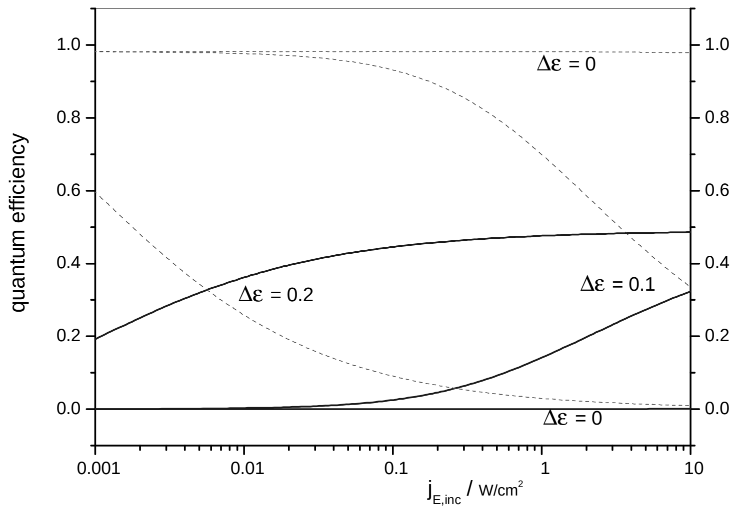

Fig.2 Quantum efficiency for the emission of high energy (thick line) and low energy photons (broken line) as a function of the intensity of incident low energy photons for systems with different degree of thermalization (**Δε**) for both, electrons and holes. All transitions are radiative.

In real devices non-radiative transitions cannot be excluded. Non-radiative transitions characterized by a cross-section are therefore added between all states of the system including transitions between the thermalized states and the intermediate level. Their effect on the quantum efficiency for the emission of low and high energy photons is shown in Figure 3. For an incident intensity of low energy photons of 0.1 W/cm^2 non-radiative transitions begin to degrade the quantum efficiencies for cross-sections as small as 10^{-22} cm^2.

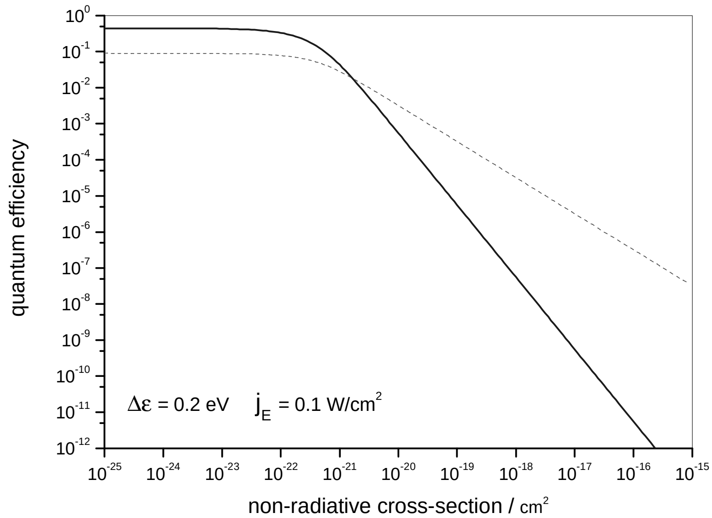

Fig.3 Quantum efficiency for the emission of high energy (thick line) and low energy photons (broken line) as a function of the cross-section for non-radiative transitions. The intensity of incident low energy photons is 0.1 W/cm^2 and the energy loss by thermalization is **Δε** = 0.2 eV.

Non-radiative transitions via an impurity state in semiconductors have cross-sections in the range of the geometrical cross-section of an atom, i.e., around 10^{-15} cm^2. For cross-sections of this magnitude, photon up-conversion is very ineffective and would not be an option for the improvement of solar cell efficiencies. A similar system, a solar cell utilizing the impurity photovoltaic effect, is less sensitive to non-radiative recombination, although cross-sections in the range of the geometrical cross-section could not be tolerated in this system either.