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Effects of quantum dot charging on photoelectron processes and solar cell characteristics

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ABSTRACT

We present theoretical and experimental analysis of photocarrier kinetics in quantum dot (QD) solar cells. The measurements of the *J–V* characteristics reveal strong effects of QD charging by selective doping of the interdot space on the solar cell characteristics. We demonstrate that charging of QDs significantly increases electron coupling to sub-bandgap photons, provides effective harvesting of IR energy, and serve as an effective tool for manipulating the potential profile at the micro- and nanoscale. The potential well for electrons in InAs QDs is substantially deeper than that for holes and, due to major differences between the effective masses of electrons and holes, the electron level spacing is substantially larger than the level spacing for holes. Therefore, QDs act as deep traps for electrons but shallow traps for holes. Filling of QDs under illumination is determined by a condition of equality of electron and hole capture rates which is realized via strong exponential dependence of the capture rates on the potential barrier around a charged dot. Without adequate doping of the QD medium, QDs are filled by electrons from the n-doped junction area and deteriorate the solar cell performance. However, selective n-doping of the QD medium results in micro- and nanoscale potential profiles favorable for photovoltaic conversion. Potential barriers around charged QDs decrease the photoelectron capture processes and suppress recombination processes via QDs. The filling of QDs predominantly from dopants in the QD medium allows one to maintain the microscale potential profile analogous to that in the best conventional single-junction solar cells.

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1. Introduction

Photovoltaic conversion is expected to be the most cost competitive technology for off-grid (remote) commercial, industrial, and military applications. To achieve the competitive cost of a few cents per kW h, either a solar cell with moderate efficiency of $\sim 10\%$ should have a low cost of $\sim \$10/m^2$ or a solar cell with high efficiency of $\sim 50\%$ should have a moderate cost of $\$100/m^2$. Various concepts for achieving high efficiency photovoltaic conversion have attracted much attention in the scientific and engineering communities for many years. The maximum theoretical efficiency for conversion of unconcentrated solar radiation that can be achieved in conventional single-junction solar cells is given by the Shockley–Queisser limit, which is 31% for AM0 spectrum [1]. This fundamental limitation in conversion arises from the relaxation to band-edges of photocarriers that are produced by photons with energies above the bandgap and by the cut-off of all photons with energies below the bandgap (see Fig. 1). To obtain a photovoltaic efficiency above the Shockley–Queisser limit, the electron levels should be adjusted to the energy of incoming photons.

The most developed concept for high efficiency photovoltaic conversion is a multi-junction solar cell with a set of junctions having different bandgaps. The junction with the largest bandgap is placed on the top and other junctions are placed in the order of decreasing bandgaps, such that each junction absorbs and converts the photons with energies between its own bandgap and that of the previous junction. Even two–three junction cells are expected to allow for significant reduction of the thermalization losses: efficiencies of 55.9% and 63.8% can be reached, with two- and three-junction cells, respectively. For high concentration of solar energy and an infinite number of junctions the limiting efficiency is 86% [3]. However, current technology enables only triple-junction cells. This technological limitation is due to differences in the thermal expansion of materials, matching requirements of lattice constants as well as the current in a cascade of junctions [4]. The maximum conversion efficiency for unconcentrated radiation realized in triple-junction cells is $\sim 34\%$ [5], which is just slightly above the Shockley–Queisser limit for a single-junction cell.

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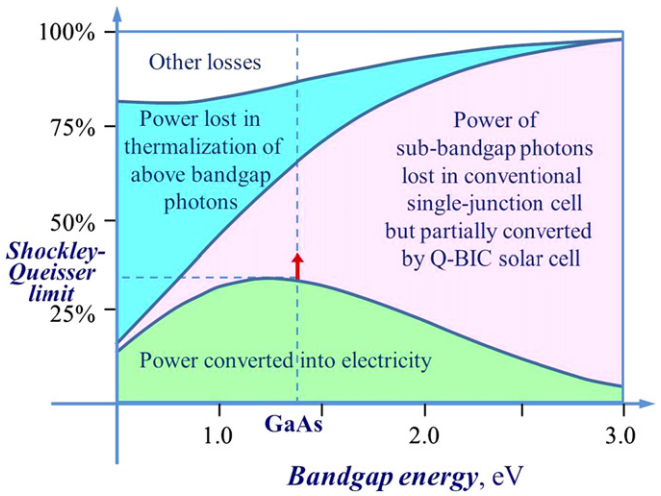

Fig. 1. Usable power, thermalization losses, and losses of sub-bandgap photons as a function of the bandgap in a single-junction solar cell. The Q-BIC technology allows for conversion of sub-bandgap photons. 5% increase (red arrow) in efficiency due to sub-bandgap photons has been already demonstrated [2]. (For interpretation of the references to color in this figure caption, the reader is referred to the web version of this article.)

However, the potential to increase the photovoltaic efficiency by adding extra energy levels in a single-junction device gained footing since the early sixties when Wolf proposed to use impurities to create a midgap subband for collecting long-wavelength radiation via two-step electron transitions [6]. In addition to band–band transitions in conventional solar cell, the electron–hole pairs in this device may be generated in a two-step process. Electron is excited from the valence band to the midgap subband by the first photon and another carrier is excited from the subband to the conduction band by a second photon. A maximum efficiency of 62.3% can be achieved with an optimized position of the impurity subband which is almost identical to the theoretically predicted efficiency of a three-junction tandem solar cell. However, formation of the impurity subband has undesirable and inevitable consequences. The presence of impurities drastically enhances the Shockley–Read–Hall recombination, which deteriorates the device performance. Keevers and Green performed detailed calculations and concluded that in the optimized impurity solar cell the conversion efficiency may increase by 1–2% absolute [7]. They highlighted that the impurities can provide substantial harvesting of sub-bandgap photons and increase the short circuit current by ~ 5 mA/cm^2. However, at high voltages impurities become effective recombination centers and always reduce the open-circuit voltage. Trade-off between IR energy harvesting and recombination losses due to impurities is a long-term problem studied without noticeable success in a number of theoretical and experimental investigations. To date, no laboratory cell that improves efficiency due to impurities has been shown and confirmed.

The modern version of the impurity solar cell is the quantum dot (QD) intermediate band solar cell. In this device the intermediate band is formed from discrete QD levels due to strong tunnel coupling between QDs [8,9]. Improvement in the photovoltaic conversion in QD intermediate band solar cell was expected due to specific photocarrier kinetics with the multiple exciton generation, which may reduce the relaxation losses related to electron–phonon processes [10,11]. To put this concept into practice, a number of technological problems should be solved. Formation of the intermediate band from discrete QD levels requires QDs of the same size and shape. Also, QD layers should be placed at regular positions in the structure. The correlated positions of QDs are realized due to local stress transferred from one QD layer to another. However, increasing the number of closely placed QD layers above 10–15 may degrade the performance of the device due to critical accumulation of strain which in turn reduces the carrier mobility. Thus far, intensive technological efforts to improve intermediate band solar cell show very limited success and an increase in the photovoltaic efficiency due to addition of QDs does not exceed 1% [12].

2. Photocarrier kinetics in Q-BIC structures

Quantum dots are multi-functional and scalable nanoblocks, which allow for fabrication of nanomaterials with specific optical and electrical properties favorable for photovoltaic conversion [13,14]. Is the formation of the intermediate band necessary to employ unique opportunities of QD nanomaterials for photovoltaic harvesting and conversion? Not at all! Besides band engineering, quantum dots provide effective ways for the engineering of 3D nanoscale potential profile to control photocarrier processes. To form the nanoscale potential profile favorable for photovoltaic applications, we propose to employ *q*uantum dots with *b*uilt-*in* *c*harge (Q-BIC), where QDs are charged via selective doping of the interdot space [2,15]. Charged dots create local potential barriers around single dots and collective potential barriers around dot clusters, rows, and other dot groups. Such potential barriers effectively separate QDs or QD areas, where harvesting of sub-bandgap photons is realized, from conducting channels where the photovoltaic conversion takes place. As in the case of impurity solar cells, the key problem of conventional QD photovoltaic materials is the enhanced recombination via additional energy levels which are introduced by QDs. In Q-BIC nanomaterials, the capture of photoelectrons into QDs is strongly suppressed by charging of QDs. Suppression of photoelectron capture directly increases the photoelectron lifetime and decreases the recombination losses. To suppress the photocarrier capture by potential barriers, the barrier height should be 2–3 times larger than k_BT. Therefore, at room temperatures, the local barriers should be at least 0.05 eV and QDs comprise at least 5–10 electrons. This requires relatively large dots and substantial doping of the interdot space.

To apply the Q-BIC technology for managing bipolar kinetics of photoelectrons and holes, it is important to determine which carriers are captured first and suppress these fast capture processes. The difference between electron and hole capture processes is mainly determined by the structures of electron and hole energy levels. The level structure in InAs/GaAs QD materials has been investigated in numerous photoluminescence measurements. All data show practically equidistant level positions of electrons and holes [15–20], as shown in Fig. 2. The total level spacing, $\Delta E = \Delta E_e + \Delta E_h$, which is directly determined in photoluminescence experiments, was found to be 60–80 meV [15–20]. The ratio, $\Delta E_e/\Delta E_h$, is evaluated indirectly from various measurements and found to be from 2 to 8 [15–17]. In our opinion, the specific equidistant positions of energy levels may be associated with the quasi-parabolic form of the confinement potential in InAs/GaAs QDs. In this model, the spacing ratio, $\Delta E_e/\Delta E_h$, is given by $(m_h/m_e)^{1/2} \approx 4$, which is in reasonable agreement with the scope of the experimental results. Using this model we obtained: $\Delta E_e \approx 55$ meV and $\Delta E_h \approx 14$ meV. Therefore, the electron transitions in QDs significantly exceed the thermal energy and cannot be induced by thermal phonons, while the hole transitions are induced by acoustic thermal phonons. Thus, to stimulate electron transitions by IR radiation for photovoltaic applications, n-doping is strongly preferred.

Fig. 3 shows the IR-assisted photogeneration of electron and hole pairs in undoped (Fig. 3(a)) and n-doped InAs/GaAs QD

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