

Photon Induced Electron Transfer in Quantum Dot Solar Cell

Ubani Charles Ahamefula

*Department of Physics, Universiti Kebangsaan Malaysia, Bangi, 43600 Selangor D.E., Malaysia.

Abstract- Low dimensional semiconductor crystals structures otherwise known as quantum dots are in possession of unique optoelectronic properties that allow the flow of electrons to be harvested instantaneously. The advent of quantum dot for various technological applications is motivated by their flexibility of their bandgap supporting their capability to tap power from the visible spectrum as well as the invisible infrared through absorbable wavelength ranges from about 700 nanometre to 10 micron. Besides the wide energy harvesting range, quantum dot utilizes small to area for high energy production. The emergence of quantum dot and its application in optoelectronic device such as solar is set to improve the efficiency limit of traditional devices. The configuration of quantum dot in solar cell is discussed as well as electron transfer processes. Their three unique advantages for solar-to-electric energy conversion such as provision of large surface and interfacial areas per unit volume for light absorption and charge separation. The confinement process of charge carriers in quantum dot provides the ability to tune the optical and electronic properties of materials in ways that are not possible with bulk materials. The study shows that the incorporation of quantum into solar cell could enhance the performance of devices and as well, low their cost.

Index Terms- Quantum dots, Electron transfer, optoelectronic properties, Charge carriers, Solar cell.

I. INTRODUCTION

The cost of photovoltaic (PV) power is determined by the PV module conversion efficiency as well as the capital cost of the PV system per unit area. The ratio of the capital cost per unit area (RM/m²) to the peak power (Wp) produced per unit area yields the generated power cost in the unit of RM/peak watt (RM/Wp). The cost of finite energy sources (gas and petroleum) in 2007 ranges from 0.25 – 0.35 RM/kWh while PV generated power was in the range of 28 RM/Wp (RM 17.5 for the PV module plus RM 10.5 for the balance of system cost). Owing to the increase in energy demand, active components for PV cell must be rid of the barriers to high efficiency such as the avoidance of internal losses due to non-radiative centres and the generation of excessive heat which is predominant in conventional silicon based solar cells. Presently, the market share of silicon-based solar cells comprises single crystal, polycrystal, and amorphous silicon is about 99%. Silicon-based solar cell consumes huge amount of energy, requires high temperature purification of the raw material and many processing steps and as a result, is expensive. “Photon induced charge transfer of quantum dot in solar cell” is favoured by nano-sized domain energetic semiconductors particles capable of absorbing wide electromagnetic spectrum and can be structured to avoid premature recombination of electrons-hole pairs.

To meet the global energy demand as well as encouraging wider use of solar power on terawatt scale (TW), there is need to develop low-cost PV cell with high conversion efficiency. The efficiency of semiconducting material used in solar cell for absorbing sunlight varies with respect to their energy gap while, the solar spectrum contains photon of wavelength between 700nm to 10 micron or in terms of energy between 0.5eV to 3.5eV. Absorption of solar photons larger than the bandgap create electron – hole pairs with the total energy equivalent to the difference between the photogenerated electron energy and the conduction band energy [1–4].

However, in a single-junction solar cell made of semiconductor material such as silicon, spectral losses can be as large as 50%. This is as a result of the mismatch between the incident solar spectrum and the spectral absorption properties of the material [5]. Large part of the solar spectrum is not absorbed as a result of the band gap (E_g) of the material. Photons with energy (E_{ph}) larger than the band gap are absorbed while the excess energy E_{ph}–E_g is effectively transferred to the electrons as thermal energy. Photons with E_{ph} < E_g are not absorbed. Alternatively, report [6] on CuInSe₂ (CIS) based solar cell is promising though limited by the complexity and complicated nature of the technology. In search for breakthrough on cost and efficiency, nanocrystalline dye sensitized solar cells in contrast to Si and CIS cells offer promising functional element for light absorption as well as charge separation. Currently, the chemisorption of dye on the surface of the semiconductor was developed to enhance the function of the dye [7, 8]. The concept uses dispersed particles to provide enhanced surface interface [9]. Photoelectrodes having high surface roughness were employed with TiO₂ as the choice semiconductor [10, 11]. The materials are inexpensive, non-toxic and abundant. A conversion efficiency of 7.1 % was obtained in 1991 [12] while the efficiency to date is over 18 %. In another development, quantum dot solar cells exploit quantum confinement either to tune the optical absorption spectrum of the solar cell or to generate multiple excitons (MEG). Though this has not to date been demonstrated in an actual solar cell several proposed designs have been reported [13, 14]. In these designs the dye molecules are replaced with a layer of quantum dots adsorbed onto nanowires. Such solar cell operates in a similar manner to the dye

sensitized solar cell but the light would be absorbed by the quantum dots instead of the dye molecules. The concept of quantum dot sensitized solar cell [15] based on CdSe quantum dots attached to ZnO nanowires was used to demonstrate the absorption of light energy to generate electrons that were then transferred to the nanowire to create electrical current. Report shows that better utilization of the solar spectrum takes place in quantum dot sensitized solar cell [16]. Besides its wide energy harvesting range, quantum dot may be painted onto walls or windows, sprayed on clothing, or printed onto rolls of paper. Owing to the design flexibility of QDSSCs, handheld devices ranging from PDAs and iPods to cell phones could be powered eliminating need for electrical cords through the use of QDSSC.

II. QUANTUM DOTS SENSITIZATION SOLAR CELL (QDSSC)

Quantum dot sensitized solar cells (QDSCs) offers an ideal approach to third generation solar cells [17-20]. The intrinsic attractive properties of quantum dots (QDs) (tunable band gap [21] high extinction coefficients, [21, 22] and large intrinsic dipole moment [23, 24]) is of advantage over metallorganic dyes as alternative light absorbing medium. Couple with the demonstration of MEG by impact ionization, QDs [25, 26] could extend the thermodynamic efficiency limit of these devices to 44% [18] ignoring 31% of the Shockley Queisser detailed balance limit. Although the conceptual efficiencies of QDSCs have not been attained [27] QD-sensitized nanostructured solar cells are rapidly been researched on as a promising alternative to conventional solar cells [28-31]. The principle of the sensitized solar cell is explained in Fig. 1.

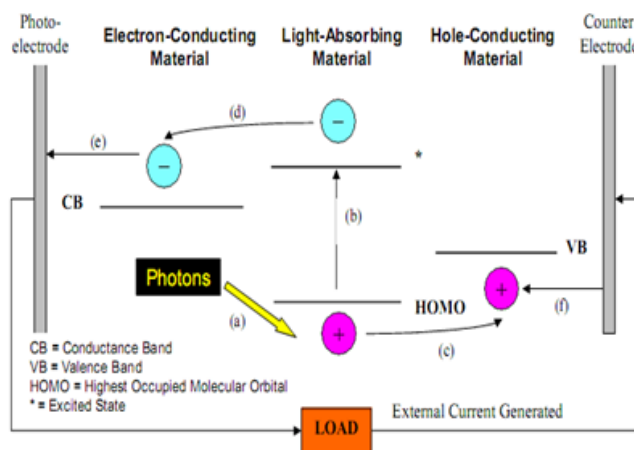


Fig. 1 Generalised Electron and Hole Orbital and Cell Movement in QDSSCs: (a) Photoexcitation of the Light-Absorbing Material; (b) Electron/Hole Pair Photogeneration; (c) Movement of the Generated Hole from the HOMO of the Light-Absorbing Material to the VB of the Hole-Conducting Material via the Transfer of an Electron from the Latter to the Former; (d) Electron Flow from the Excited State of the Light-Absorbing Material to the CB of the Electron Conducting Material; (e) Electron Flow Through the Photoelectrode and the External Circuit, Re-entering Through the Counter Electrode; (f) Regeneration of the Hole-Conducting Material

In general, sensitized solar cell is made up of three principle components: a light-absorbing material, an electron-conducting material and a hole-conducting material. A Quantum Dot Sensitized Solar Cell is shown in Fig. 2. In a QDSC, it is the QDs that absorb the light, creating an electron/hole pair. The efficiency of QDSSCs depends on the spectral absorbance of the QDs semiconductor material and the electron/hole injection efficiency, the electron transport and recombination rates, which are dependent on the electron-conducting material; and the hole transport and stability is related to the hole conductor used. The processes taken place in a QDSSC such as photoexcitation, electron injection as well as recombination of electrons and holes is shown in Fig. 2. QDSSC are sandwiched between two electrical contacts or electrodes.

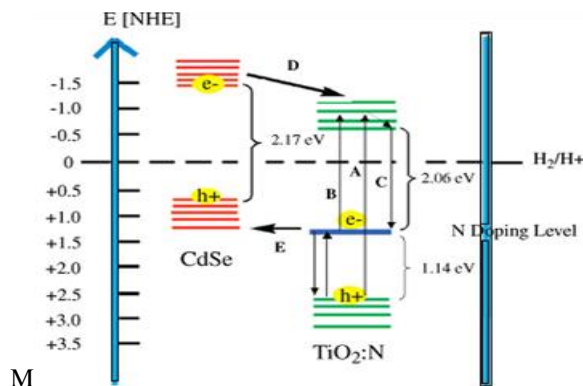


Fig. 2 Diagrammatic representation of electronic band structure of 3.5 nm CdSe with bandgap of 2.17 eV and nanocrystalline TiO₂:N having bandgap of 3.2 eV, in association with TiO₂ and a N dopant state of about 1.14 eV above the valence band. Electron and hole generation, relaxation, and recombination pathways are shown in the figure with (A) photoexcitation of an electron from the valence band (VB) to the conduction band (CB) of TiO₂, (B) transition or photoexcitation of an electron from the N energy level to the CB of TiO₂, (C) recombination of an electron in the CB of TiO₂ with a hole in the N energy level, (D) electron transfer or injection from the CB of a CdSe QD to the CB of TiO₂, and (E) hole transfer from the VB of a CdSe QD to the N energy level. Note that not all of these processes can happen simultaneously, and many of these are competing processes. [76].

On illumination and creation of electron/hole pairs, electrons injected into an electron-conducting material flow through to the anode and return through the cathode creating an external current which can be used by a load. The electron reduces the oxidised hole conductor and restores it to its original state as shown in Figure 3.1.

The sensitization of a wide gap nanostructured semiconductor electrode such as TiO₂ and ZnO with QDs can be achieved through either; (1) direct growth of the semiconductor QDs onto the electrode surface by chemical reaction of ionic species using chemical bath deposition (CBD) [30 - 33] or successive ionic layer adsorption and reaction (SILAR) [34, 35] and (2) through presynthesized colloidal QDs attached to the electrode material by a bifunctional linker molecule [36-40]. The attachment of colloidal QDs through molecular wires enhances the morphological properties of the semiconductor nanocrystals. Attaching the QDs onto the TiO₂ surface without molecular linkers is termed direct adsorption (DA) [41]. Higher surface coverage of colloidal QDs on the TiO₂ substrate improves the efficiencies of QDSSCs as well as the network configuration.

III. CONFIGURATION OF QDS IN SOLAR CELL

A. The Sensitizer

The sensitizer on the surface of the nanocrystalline semiconductor absorbs incoming photon flux. To achieve high conversion efficiencies, the photochemical, electrochemical, and chemical properties of the sensitizer play important role. Owing to the quantum size effect the edge of the absorption spectra of QD material can be tuned to different sizes. This can be done by varying the synthesizing temperature. Such an effect produces narrow size distribution [42-45] as absorption occurs throughout the visible region. To ensure a good injection rate of the photoexcited electron from the sensitizer into the TiO₂ a long excited state of the sensitizer is necessary since injection occurs on the femto-to picosecond time scale. The sensitizer performance determines the output current of the QDSSC [46]. An illustration showing the generation and flow of electron/hole pairs, through the anode to the cathode is as shown in Fig. 3. The oxidized hole restores the system to its original state.

For efficient electron injection into the TiO₂, the energy level of the sensitizer excited state must be higher than the energy of the lower edge of the conduction band of the semiconductor as the oxidation potential of the sensitizer excited state increases negatively than the oxidation potential of the TiO₂ semiconductor. On the other hand, the energy level of the oxidized sensitizer has to be lower than the energy level of the reduced hole-conductor to ensure an efficient regeneration of the sensitizer. For the QD size distribution, a size limit exists for the nanoparticles to inject electrons into the TiO₂ [47].

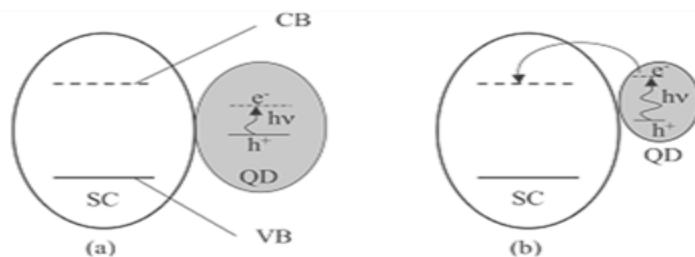


Fig. 3 Electronic levels in semiconductor (SC) and QD particles showing the valence band (VB), conduction band (CB) in (a) Large CdSe particle, (b) Small CdSe particle

Illustration in (Fig 3.(a) is dominated by large QD particles which is adsorbed onto the semiconductor; its small band gap is as a result of weak quantum confinement. However, energy of the photogenerated electron in this particle is weaker resulting in poor injection of electron into the semiconductor to affect the process. However, smaller QD as in Fig. 3. (b) depicted that the energy of the photogenerated electron is greater than that of the lowest unoccupied molecular orbital (LUMO) of the semiconductor resulting in rapid electron tunneling to the TiO₂ particle. To achieve good adsorption of the sensitizer on the semiconductor, chemical bonds have to exist between both which however, depend on the synthetic route. For QD grown directly on the surface of the semiconductor the anchoring is due to bonds appearing between the deposited metal ions and the oxygen atom at the surface. QD may also be formed in the bulk of the deposition solution before migrating to the surface of the semiconductor. The adsorption of the particle may not be due to metal/oxygen bonds in this case, as no metal valences are available. For metal selenide QD, there is tendency for a hydrogen bond to form between the selenide atom and the oxygen at the surface.

B. Electron-Conducting Material

Electrons generated in QDSSCs flow through the conduction band of TiO₂ to an electrode. The conduction band energy electron must be efficient for electronic coupling with the sensitizer energy levels and charge separation to minimize the recombining of the electron with the sensitizer. An electron-conductor with low level of recombination and good electrical connectivity to the electrode/substrate is preferred. [48] A situation where the injection is greater than the rate of recombination is termed kinetic redundancy. [49] The kinetic redundancy can be modified by changing the sensitizer-electron-conductor linker groups. [49] Usually anatase and rutile are used as the linkers. Anatase is preferred to rutile because it is transparent, colourless and stable at low-temperature. [50] Besides, it has a better light scattering property when compared to rutile, and its efficiency is higher by a factor of five. [51]

The success of QDSSCs has been partly attributed to the fast electron injection from the QDs to the TiO₂ and a slower reversal process [52]. High speed electron injection is consistent with near 100 % quantum efficiency [51]. Alternative wide band gap materials such as ZnO [53, 48, 54, 55, 56], Nb₂O₅ [53, 57], SnO₂ [53] and In₂O₃ [53] are insensitive to the visible spectrum and absorb high energy UV wavelengths. Such qualities are desirable as they imply strong chemical bonds, stable material [58] and are transparent to visible light as a result and are therefore more efficient at creating stable electron/hole pairs.

C. Photoelectrode

The type of electrodes use in QDSSCs affects the overall cell performance and marketability of a solar cell. Pt-free counter-electrodes use in QDSSCs with Au and Cu₂S electrodes are expensive [59, 60]. However, carbon electrode has emerge as a better alternative to Pt electrode owing to their low cost, high electric conductivity, heat resistance and corrosion-inert. Activated carbon-based electrode assembles onto CdS QDSSCs yielded conversion efficiency of 1.47%. [61]. Photoelectrode fabricated by sequential assembly of CdS and CdSe QDs can effectively be adsorbed on ZnO nanowire array. The electrodes with nanospheroidal morphology have shown better performance compared to P25 nanoparticle electrodes owing to their connectivity among nanospheroidal TiO₂ particles and deeper penetration of the electrolyte in QDSSC [62].

D. The Photoelectrode-Anode

The anode is the electron-conducting electrode termed photoelectrode as it is the illuminated electrode. Electron-conducting materials are usually placed onto the anode to maximize contact by doctor blading, [63, 53, 64-66] screen printing, [63, 67, 50, 64, 68], spin coating, [69] or sol-gel deposition of dissolved electron-conducting material with a binder.[63, 70] The contact and electron-conducting material could be fired in air to about 450°C to 500°C in a case where TiO₂ is used to aid the removal of the binder and solvent [71] as well as sintering the electron-conducting particles to the substrate. [50, 72, 73].

E. The Counter Electrode (Cathode)

Interaction between the hole conductor and the anode results in recombination [74]. The interaction at the cathode must be fast to be kinetically favourable. Base on this reason, the cathode for instance, (usually ITO glass) is often coated with carbon [75], platinum [53, 73, 48, 54] or gold. [67, 65, 69]. Poly (3,4-ethylenedioxythiophene) (PEDOT) coated Fx•SnO₂ glass could be used as counter electrode in TiO₂ QDSSCs. [74]. In addition, coating the back electrode of QDSSCs with platinum could improve cell performance. [51] The cathode can as well be adjusted to accommodate equilibrium potential close to the hole conductor's, as the anode (the photoelectrode) is predominantly far from the equilibrium potential. Such coating would not be necessary for cells without iodide/triiodide based hole conductors. Electron transport in nanoparticle QDSSCs such as in TiO₂ occur either between the counter electrode and the working electrode by hopping [75] or through extended diffusion states [48]. Owing to the high electrons mobility in

nanoparticle, the disorder of the nanoparticle slows down the electron transport. Electron hopping in the nanoparticle network is efficient. Electron transport and recombination are interconnected in such a manner that any increase in the transport rate increases the recombination rate as well; keeping the QDSSC performance stable.[76] The ZnO nanowires with electron diffusivity of 0.05 to 0.5 cm²s⁻¹ could be an alternative to TiO₂ nanoparticle. ZnO nanoparticle provide direct path from photogeneration to conducting substrate and improve electron transport irrespective of its current densities which is the same with TiO₂ nanoparticle [77]. Every electron injected is connected to the substrate [78] as the coiled wires carry the injected electron to the anode. [48]

F. Electrolyte

Electrolytes in QDSSC contain the electrically conductive charges. The electrochemical potential of the interface between a semiconductor electrode and an electrolyte solution must be the same for the two phases to be in equilibrium. The excess of charge on the semiconductor due to low density of charge carriers could extend for 10-100 nm into the electrode and forms the space charge region. The space charge region is associated with positive charge. A p-type semiconductor usually has a Fermi level that is lower than the redox potential of the electrolyte and as a result, electron flows from the solution to the electrode to generate negatively charged space charge which causes downward bending of the bands. This phenomenon is as illustrated in Fig. 4

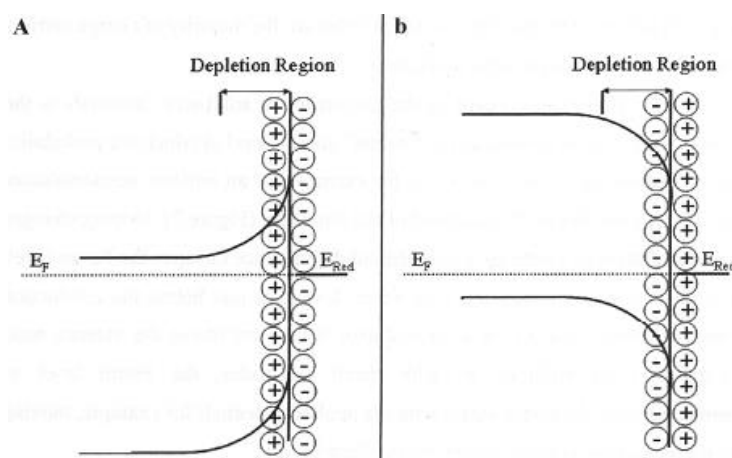


Fig. 4 Band bending for an n-type semiconductor (a) and a p-type semiconductor (b) in equilibrium with an electrolyte.

Exposing semiconductor electrode to radiations of sufficient energy, could promote electrons to the conduction band. This process results in recombination with the production of heat. If it occurs in the interior of the semiconductor the electrical field in the space charge region will promote spatial charge separation.

G. Liquid Electrolytes

Liquid electrolytes based on iodide/triiodide redox couple [64] prevents recombination with the electron by donating electron to the hole in the QDs [78] and oxidises to triiodide. Iodide is regenerated by the reduction of triiodide ion at the counter-electrode. The iodide/triiodide couple is ideal for QDSSCs application owing to the irreversible nature of the iodide to triiodide transition. However; the reverse process is kinetically unfavorable [79]. Replacing electrolyte-based components in QDSSC could enhance the cell performance. Gels [80-82] can replace liquid electrolyte in QDSSC such as Iodide-based (I⁻/I₃⁻) electrolyte and Pt electrode with CdS as sensitizer [83]. Hara et al. [68] found that increasing the concentration of the I⁻ source from 0.007 to 3mol/dm³ affects the short circuit current (I_{sc}) and open circuit voltage (V_{oc}). Increasing the concentrations of I⁻, I_{sc} and conductivity of the electrolyte (I⁻ and its cation) increase [68]. However, increasing the concentration of I⁻ beyond the maximum point decreases I_{sc} because of an increase in viscosity of the solution, which decreases ion mobility [68].

Charge separation is the building block of electron generation and takes place at the interface of the solar cell. The optimization of nanocrystalline QDSSC interface is investigated to minimize the losses resulting from direct recombination of the electron-hole pair after excitation (fig. 3.4 (K_{des})). Charge separation is efficient in QDs due to ultra-fast electron injection from the sensitizer to the semiconductor (fig. 3.4 (K_{inj})). However, interfacial charge recombination leads to high losses in the QDSSC as a result of the mesoporous network and the small distance between the electron and the holes [84].

The recombination of the injected electron with the oxidized sensitizer (fig. 3.4 (K_{reg})) is shown to be in the millisecond time range, which implies that the short-circuit potential will not suffer from recombination [84, 85]. To prevent the recombination of electrons collected from the conducting SnO₂ layer, the oxidized hole-conductor, a compact TiO₂ layer is usually deposited between the

conducting glass and the nanocrystalline TiO₂ [86]. The recombination process between the injected electron in the TiO₂ and the oxidized hole-conductor (Fig. 5 (K_{rec1})) is the major electron injection leading to the enhancement of the dark current.

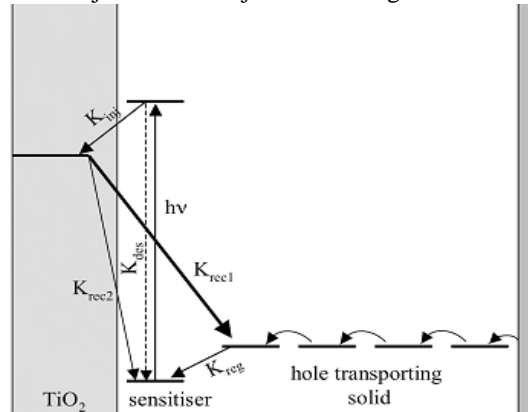


Fig. 5 Possible recombination pathway in QDSSC

Recombination occurs as a result of electron trapping in the surface states. The nanocrystalline (TiO₂/ZnO) used in the QDSSC has large surface area. The deposition of a thin semiconductor layer covering the TiO₂ to preventing contact between the oxidized hole-conductor and the TiO₂ could increase the efficiency. Similar measure has been used to increase the photovoltaic device performances using ZnO [87], Al₂O₃ [88], Y₂O₃ [89] and SrTiO₃ [90]. However, long chain alkyls could be used to passivate the surface of semiconductors such as SiO₂ [91, 92]. The phosphonic or carboxylic groups help efficient adsorption at the surface. However, the long alkyl chain (Fig. 6) increases the distance between the surface and the electrolyte [93, 94] and enhances the short-circuit current while the open-circuit voltage could be increased with hexadecylmalonic acid (HDMA).



Fig. 6 Molecules of hexadecylmalonic acid (HDMA) used for the surface modification in QDSSC

IV. INTERACTION OF LIGHT WITH SEMICONDUCTORS

When a monochromatic incident light (I) hits a semiconductor material, a certain amount of the light energy is refracted (R) and certain proportion is transmitted (T). The transmitted light energy is absorbed by the semiconductor material to excite the electrons. The light energy enables the electrons to jump from one energy state to the other. When the photon energy is greater than the bandgap energy of the semiconductor, the absorption in the material occurs to excite the electrons from the highly occupied valance band towards the empty conduction band [95-96]. Excitations of electron and electron irradiation may lead to the generation of charge carriers.

A. Recombination Process:

As semiconductor is illuminated with light of appropriate wavelength, electron-hole pairs are formed. This increases the concentration of the charge carriers in the illuminated state compared to the dark state. As light is discontinued, the charge carries decay back to the equilibrium values. The process of decaying is refer to as recombination [97, 98]. The recombination process occurs in one of a few possible ways; a) an electron releases its energy through irradiative recombination which involves the emission of energetic photon; b) the electron losses its energy through Auger recombination by transferring energy to neighboring electrons [99] phonon is generated by losing the energy of the electron [98, 100]. It becomes clear that defects in semiconductor nanocrystals give rise to the differences in the energy level which is borne out of difference in bandgap energy. At the emergence of this phenomena, recombination processes slows down. As illustrated in Fig. 7, electron move from the conduction band to the defect energy level and then moves to the valence band annihilating the hole at the valence band. This process is known as trap.

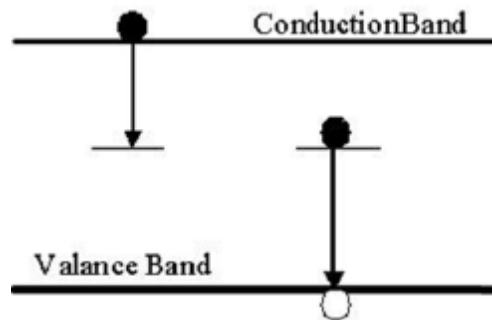


Fig. 7 Two-step recombination process at trapping level within the bandgap

The devices losses energy as result of recombination process. However, as recombination when the photon energy is discontinued from striking the semiconductor, the flow of current stop producing zero current output. When external trap is incorporated into the system, recombination slows down and allows the charges to flow. With the two-step recombination, the devices has higher chances of the charge carriers moving around the system. In application, this behaviour reduces the efficiency of a solar cell and can only be improved by introducing QDs with other semiconductor material [101] such as CdSe and PbSe QDs [102].

B. Transport Process

The charge in semiconductor materials are initiated by the movement of electrons throughout the delocalized band states [98]. In order to clearly explain charge transport process, transport of charge by the movement of electrons in the conduction band and movement of holes in the valance band is used in this paper. During the transport process of charges throughout the semiconductor material, phonons are created and this result to the increase in the temperature of the material and thus, introduces thermal conduction [103]. At thermal equilibrium, the carriers follow Boltzmann distribution depicting a function of temperature of the system [104]. Hence, hot carriers causes the decrease in the efficiency of the photovoltaic cells. The hot carriers exist in the system for a few hundred picoseconds before recombination. The hot carriers produce electric current. However, if the energy can all be captured from the hot carrier and transfer to electric energy, the efficiency of the solar cell would increase to about 66% [105] as illustrated in Fig. 8.

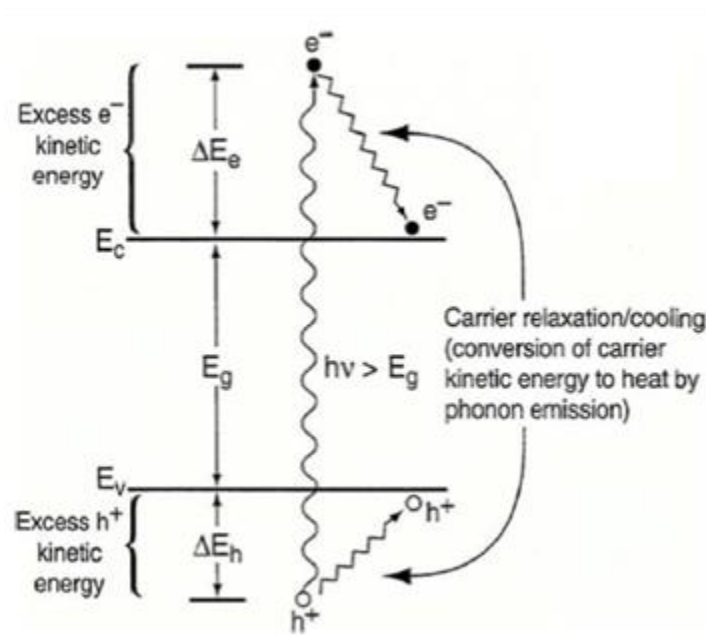


Fig. 8 Hot carrier relaxation/cooling dynamics in semiconductor [106]

V. CONCLUSION

High cost and low efficiency of conventional single junction solar cells resulting from the inability of their to absorb and convert all the photon energy absorbed by free electrons and holes in the photon has retarded competitive use of photovoltaic cell. The flexibility and lower production cost afforded by quantum dot is promising to a wider range of applications especially for solar cells enhancement. The usefulness of quantum dot continues to strive as their application becomes more viable. Nanostructures present three unique advantages for solar-to-electric energy conversion which includes first, the provision of large surface and interfacial areas per unit volume for light absorption and charge separation. Second, confinement of charge carriers in nanometer size particles provides the ability to tune the optical and electronic properties of materials in ways that are not possible with bulk materials. Lastly, nanostructured materials could help reduce solar cell manufacturing costs by providing the means to mass produce thin films inexpensively through roll-to-roll coating or printing approach. Besides, the synthesis techniques could enable the design of sophisticated nanostructures. Besides, the design flexibility of QDSSCs could be utilized in powering handheld devices extending from PDAs and iPods to cell phones.

QDs provides a low surface coverage of about 14%. This paper discusses the structure of QDs devices and their optoelectrical properties capable of being manipulated by changing their particle sizes. The colloidal dispersion with particle size range of QDs improved the efficiency of device by absorbing wider electromagnetic spectrum. This process provided interconnections between the particles with electrical contact between them. The final film is mechanically stable, transparent and has a thickness of a few microns. Fabrication at the nano-scale provides a remarkable increase in the precision and level of control that can be obtained in solar cell development. The configuration of quantum dot in solar cell supports electron transfer processes besides their unique advantages that enhances solar-to-electric energy conversion such as large surface and interfacial areas per unit volume for light absorption and charge separation. In addition, the confinement process of charge carriers enables the tuning of the optical and electronic properties in such a way that support their use in different applications. It becomes clear that the incorporation of quantum in solar cell provides enhancement platform for devices performance.

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AUTHORS

Correspondence Author – Dr. Charles Ahamefula Ubani, decharlington@yahoo.com or Charles_4_christ@yahoo.com; contact: +60166974183