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Hot carrier exploitation strategies and model for efficient solar cell applications

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Hot carriers are electrons or holes that are created in semiconductors upon the absorption of photons with energies greater than the fundamental bandgap. The excess energy of the hot carrier cools to the lattice temperature via carrier–phonon scattering and wasted as heat in [the] picoseconds timescale. The hot-carrier cooling represents a severe loss in the solar cells that have significantly limits their power conversion efficiencies. Hot carrier solar cells aim to mitigate this optical limitation by effective utilization of carriers at elevated energies. However, exploitation of hot carrier energy is extremely challenging as hot carriers rapidly lose their excess energy in phonon emission and therefore requires a substantial delay of carrier cooling in absorber material. In this paper a simple model was formulated to study the kinetic energies and hence the energy levels of the photo excited carriers in the quantum dots (QDs) whereas Schaller model was used to investigate the threshold energies of considered QDs. Results strongly indicate low threshold photon energies within the energy conservation limit for PbSe, PbTe, PbS, InAs, and InAs QDs. These materials seem to be good candidates for efficient carrier multiplication. It is found also that PbSe, PbTe, PbS, InAs, ZnS and InAs QDs exhibit promising potential for possible hot carrier absorber due to their widely spaced energy levels predicted to offer a large phononic gap between the optical and acoustic branches in the phonon dispersion. This in principle enhances phonon bottleneck effect that dramatically slows down hot carrier cooling leading to retention of hot carriers long enough to enable their exploitation. Two novel strategies were employed for the conversion of hot carriers into usable energies. The first approach involves the extraction of the energetic hot carriers while they are 'hot' to create higher photo voltage while the second approach uses the hot carrier to produce more carriers through impact ionization to create higher photo current. These mechanisms theoretically give rise to high overall conversion efficiencies of hot carrier energy well above Shockley and Queisser limit of conventional solar cells.

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1. Introduction

Hot carrier solar cell is a third generation photovoltaic specifically designed to tackle carrier thermalisation losses after absorption of supra bandgap photons with efficiencies in close proximity to the maximum thermodynamic limit. In semiconductors, a supra bandgap illumination source generates hot carriers with excessively high kinetic energy. These characteristic optically-excited hot carriers with energies well above Fermi level have very short lifetime that can be utilized for photovoltaic applications where strategies for their effective exploitation are highly desirable for greater efficiencies. Despite their potentials in driving quantitative gains in the efficiency of energy conversion [scenarios], their short lifetimes open the possibility of using them as optically-active material in devices with short response times which results in expansion of frontiers in the field of hot carrier Physics.

Understanding the hot carrier dynamics and their exploitation can considerably permits the engineering of materials and techniques to harvest what would otherwise be dissipative losses in

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the single junction semiconductor solar cells. Initial effort to explore the hot-carriers is practically impossible in relation to hot-carrier cooling that is usually much faster than the carrier transport and extraction in the conventional devices. Quantum dots (QDs) have emerged as the most attractive photo active materials for hot carrier solar cells due to their large absorption band and discrete electronic states that modulate the phonon dispersion and induce phonon bottleneck effects to slow the hot carrier cooling which provide new energy flow pathways not achievable in the bulk materials. Thus enhancing the basic photo conversion processes in a solar cells by utilizing the excess energy of the hot carrier can achieve a significant increase in the overall power conversion efficiency.

Semiconductors play a significant role in solar cells which absorb solar radiation and convert it into electrical energy [1]. In semiconductors, fundamental bandgap energy governs their optical and electronic behaviors and is arguably the most critical parameter for the range of photons solar cells can absorb and convert into electrical energy (Jara, 2014). Sunlight is a form of electromagnetic radiation, which is composed of particles called photons. Each photon is associated with a particular wavelength and energy given by:

$$E_{photon} = \frac{hc}{\lambda} \tag{1}$$

where, E_{photon} is the photon energy, h is the Planck's constant, c is the speed of light and λ is the wavelength of light. When solar energy hits a solar cell, a fraction of the energy is absorbed by the semiconductor material to generate an electron-hole pair by exciting an electron from the valence to the conductance band. The electron and hole are then separated by the electric field created by the p-n junction, which pushes the electrons to the top of the cell towards the n-type and the holes to the bottom towards the p-type. In principle, the electrons are passed through an external load as a current and then recombined with holes at the bottom of the cell [2]. These processes are depicted in Figure 1.

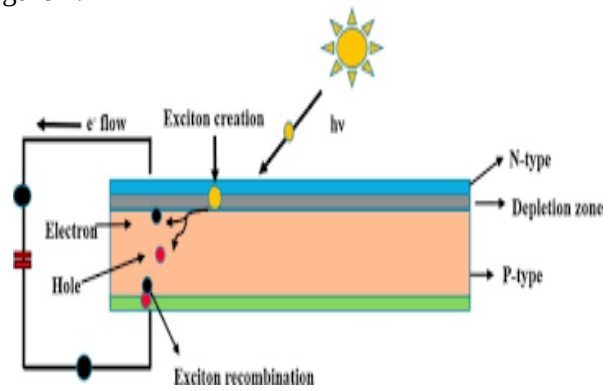

Fig. 1. Basic operation of solar cell

The excitation of charge carrier in semiconductors is possible only when incident photon corresponds to the bandgap energy of the absorbing semiconductor material [3]. Consequently, sub bandgap photons ($E_{photon} < Eg$) are unable to produce photo current thus leading to transmission loss [4] while photons having energies greater than the material band gap ($E_{photon} > Eg$) are utilized to generate electrical energy but the excess energy is given as the difference between the photon energy and the semiconductor band gap ($E_{photon} - Eg$) [5]. In the conventional single junction solar cells these excess energies of the photo generated electrons and holes are in the form of kinetic energy and the charge carriers are characterized by a temperature greater than that of the lattice and are termed "hot carriers" [6]. The electrons in the valance band will be photo-excited to the conduction band with excess energies ΔE_e given as [7]

$$\Delta E_e = (hv - E_g)\left[1 + \frac{m_e^*}{m_h^*}\right]^{-1} \tag{2}$$

while the distribution of the excess energy for the hole in the valence band ∇E_h is expressed as

$$\nabla E_h = \left(h\nu - E_g\right) - \nabla E_e \quad (3)$$

where m_e^* and m_h^* are the electron and hole effective masses respectively, ΔE_e is the energy of the electron relative to the conduction band, and ΔE_h is the hole energy relative to the valence band. Subsequent to the absorption of a photon with energy ($E_{photon} > Eg$), hot carriers lose energy by cooling down from their initial energetic ("hot") position to the band edges on a timescale of tens of picoseconds. This relaxation process is accompanied by the conversion of the excess energy ($E_{photon} - Eg$) into thermal energy via carrier-phonon scattering [8] as shown in Figure 2. This process produces carrier cooling often called thermalization [9].

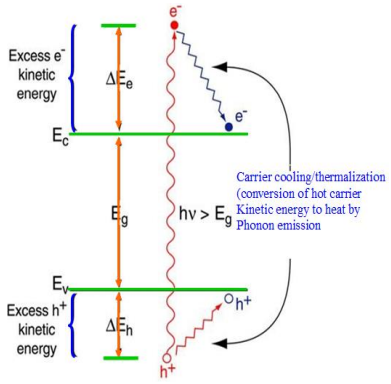

Fig. 2. Hot carrier cooling in semiconductors [10]

Thus, hot carrier populations created by photon absorption cool down and transmits their excess energy to the semiconductor lattice in the form of vibration modes – called phonons. An important mechanism for thermalisation of hot carriers is energy transfer to the host lattice by interactions between carriers and optical phonons as described by Fr¨ohlich interactions [11]. Thus hot carriers lose their energy by emission of optical phonons at high temperatures which decay into acoustic phonons at lower (energy) temperatures as shown in Figure 3

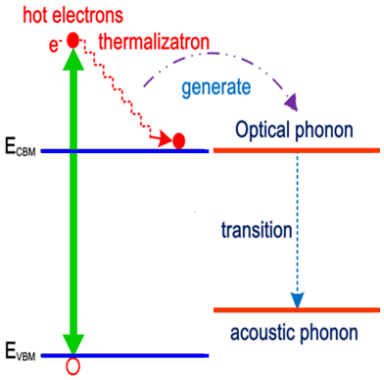

Fig. 3. Decay of high energy optical phonon to lower energy acoustic phonon.

Decay of high energy optical phonons to lower energy acoustic phonons is dominated by processes known as Klemens decay and Ridley decay mechanisms which strictly adhere to conservation of energy and momentum laws. Klemens decay occurs when a phonon in the optical branch decays into two acoustic phonons with half the energy but opposite and equal momentum

[12]. Ridley decay occurs when an optical phonon decays to an optical phonon of lower energy and an acoustic phonon [13]. Carrier-phonon interaction is responsible for several phenomena including phonon replicas in the emission spectra, homogeneous broadening of the excitonic line width and the relaxation of hot carriers to the fundamental band edge [14]. A wide separation between optical and acoustic branches of the phonon dispersion can remove the dominant Klemens decay and possibly slow the carrier cooling rate [15]. The thermalization of hot carriers constitutes the major losses that have dramatically constrained the power conversion efficiency of conventional devices to 31% based on thermodynamic detailed balance analysis by Shockley and Queisser [13]. The power generated by a solar cell is determined by the product of photo current and the photo voltage and these parameters are determined by band gap of semiconductor used in the solar cells [14]. Generally, if the bandgap is higher, fewer incoming photons can be absorbed but the electrons have higher energy (higher voltage)[15]. Conversely, semiconductor absorber with narrow electronic bandgap permits absorption of a wide range of photons and hence produces a high current but the electrons have lower energy (lower voltage) and gives room for hot carriers that are usually thermerlised to their respective band edges in a very short timescales [16][17].

The efficiency of a photovoltaic device can be increased if the kinetic energy of the hot carriers is utilized to excite additional carriers across the band gap upon absorption of one energetic photon through impact ionization (an inverse Auger process) leading to an increase in the photo current [18][19]. In bulk semiconductors the threshold energy for impact ionization is extremely high for practical consideration due to the need to conserve both the energy and momentum during the generation of extra charge carrier [20][21]. Theoretical measured energy threshold for impact ionization in bulk semiconductors is 4-5 times the band gap [22]. This together with ultra fast phonon relaxation renders bulk materials incompetence for carrier multiplication solar cell applications. A second approach to boost the photon conversion efficiency requires that the kinetic energy of the hot carriers be extracted from the photo-converter to do useful electrical work before they cool to band edges leading to higher photo-voltage [23].

However, effective utilization of hot carrier energy is extremely challenging as hot carriers rapidly lose their excess energy as they tend towards thermal equilibrium with the lattice and therefore requires a substantial delay of the hot carriers thermalisation from the picoseconds timescale common in semiconductors, in addition to low threshold energy properties required for efficient impact ionization. The incorporation of QDs that simultaneously offer an improved optical absorption, photo-excited carrier collection and phonon bottleneck effects are presently being explored. QDs are semiconductors in nanometer dimensions providing strong quantum confinements on the carriers in 3D leading to discrete and size dependent energy spectrum in sharp contrast to the bulk matter whose electronic states are distributed in continuous bands as depicted in Figure 4, thus making QDs typical example of live quantum mechanical phenomenon [24]. The discrete energy level spectrum in QDs is a replica of atomic energy spectra which confers on them the moniker “artificial atoms”[25].

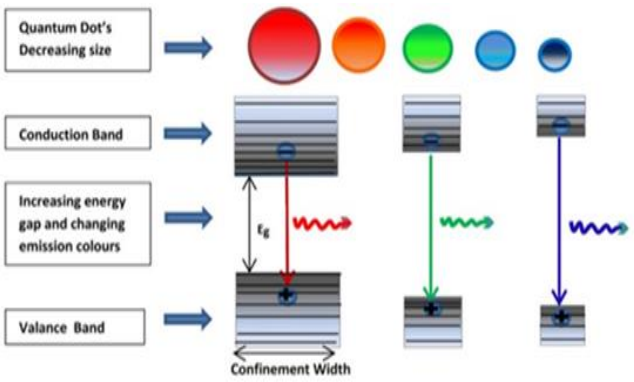

Fig. 4. Blue shift in QD electronic bandgap with decreasing size

In this research we propose quantum mechanical model and two strategies to potentially exploit the hot carriers based on quantization effects in QDs for efficient solar cell applications.

2. Theoretical Framework

Semiconductor QDs are two particle system comprising an electron and hole confined in a quantum box bounded by infinite potential walls as shown in Figure 5. Due to the opposite polarity charges that exist between these charged particles there is an attractive connection between them and the particles interact through the electrostatic potential to form a quasi bound state called exciton. Thus, a good approximation of an exciton behavior in QD is that of particle trapped in an infinite potential well. Here QD was approximated as quantum box and the exciton as a confined particle. This scenario is presented when the semiconductor is reduced to the nanoscale range where quantum confinement effects are used to describe the system behavior. Quantum confinement effects are observed when the generated excitons are restricted to smaller spatial volumes than they would occupy in the bulk matter. The natural spatial range over which the confinement emerges is described by the material natural exciton Bohr radius of the bulk matter or de Broglie wavelength of conduction band electron [26].

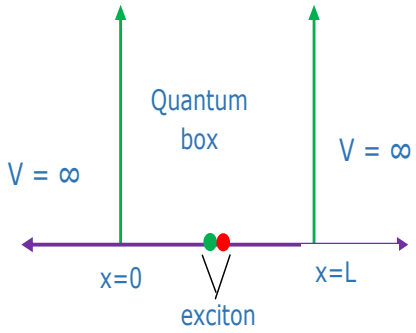

Fig. 5. Exciton in the Quantum Box Model [27]

We recall that the confinement energy of the particle confined in 1D well of width L is expressed as

$$E = \frac{n^2 h^2}{8mL^2} \tag{4}$$

In a quantum dot, the charge carriers are confined in 3D potential well and by approximation the photo-excited carriers (electron and hole) may be treated as particles in a spherical potential well. Putting $m = m_e^*$ into equation (4) for a spherical QD of radius R yields the electron confinement energy E_{en} as:

$$E = \frac{n^2 h^2}{8m_e^* R^2} \tag{5}$$

Similarly, putting $m = m_h^*$ the hole confinement energy E_{hn} is obtained as:

$$E = \frac{n^2 h^2}{8m_e^* R^2} \tag{6}$$

Therefore, the total confinement energy of the confined electron and the hole (exciton) is expressed as

$$E = \frac{n^2h^2}{8m_e^*R^2} + \frac{n^2h^2}{8m_e^*R^2} \quad (7)$$

where E is kinetic energy of exciton in the QDs, n is the quantum number that labels the different confined energy levels of exciton within QD structure, R is the dot radius, m_e^* is the mass of the electron, m_h^* is the effective mass of the hole and h is Plank's constant. The model strongly indicates that energy levels of the exciton are quantized and are limited to discrete values and this arises out of a need to fulfill the imposed boundary conditions on the system.

The threshold energy for electron-hole pair production by a carrier in a semiconductor has long been of interest in many branches of semiconductor Physics. It has recently become of importance in the interpretation of high field carrier transport phenomenon. In considerations of energy conservation, there is an energy threshold below which impact ionization cannot occur. We have made a new calculation of the threshold energy based in Schaller model. In this model the expression for threshold energy E_{th} for QDs on the basis of electron effective mass m_e^* and hole effective mass m_h^* is given as [28]

$$E_{th} = \left(2 + \frac{m_e^*}{m_h^*}\right) E_g \quad (8)$$

3. Methodology

Exciton kinetic energies as a function of sizes for CdSe, CdS, ZnS, GaAs, InAs, InSb, PbSe, PbS and PbTe QDs were computed using equation 7 for theoretical characterization of the dot materials with high kinetic energies and hence widely separated energy levels to realize the required phonon bottleneck effect to slow down the hot carrier thermalization rate. The threshold photon energies for the considered QDs were also computed analytically using Schaller model of equation 8 to investigate the QDs with low threshold energies for efficient carrier multiplication. The input parameters used in the theoretical calculation have been listed in Tables 1, 2 and 3 of the appendix.

4. Results and Discussion

Exciton kinetic energies as a function of sizes for group II-IV (CdSe, CdS and ZnS), group III-V (GaAs, InAs and InSb) and the group IV-VI (PbSe, PbS and PbTe) QDs using the equation 7 are plotted in Figures 6, 7 and 8 respectively. The results show that kinetic energy of the charge carriers in QDs exhibits quadratic inverse dependence on the confinement dimension. With decreasing dot size the kinetic energies become larger by the factor $1/R^2$ which suggests an increase in the energy levels. Thus energy level spacing increases sharply with decreasing size. It is found for the group II-IV QDs that CdSe and ZnS display high kinetic energies as shown in Figure 6.

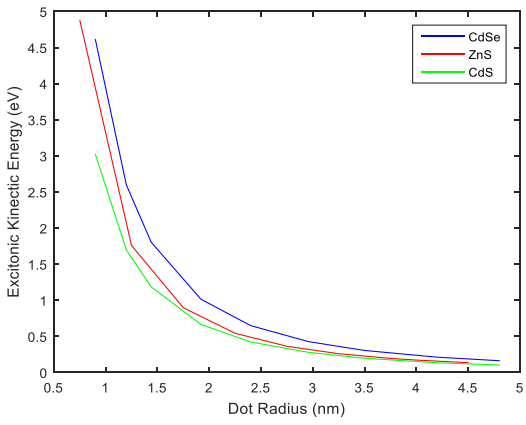

Fig. 6: Excitonic Kinetic Energy versus Dot Radius for CdSe, ZnS and CdS QDs

However, for the group III-V QDs only InAs displays high kinetic energies as shown in Figure 7

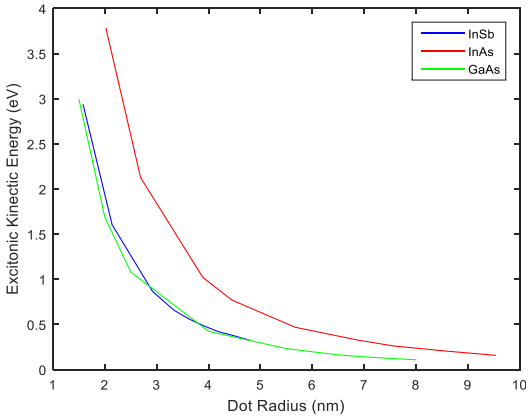

Fig. 7. Excitonic Kinetic Energy versus Dot Radius for InSb, InAs and GaAs QDs

Finally, for the group IV-VI QDs, both PbS, PbSe and PbTe display high kinetic energies as shown in Figure 8.

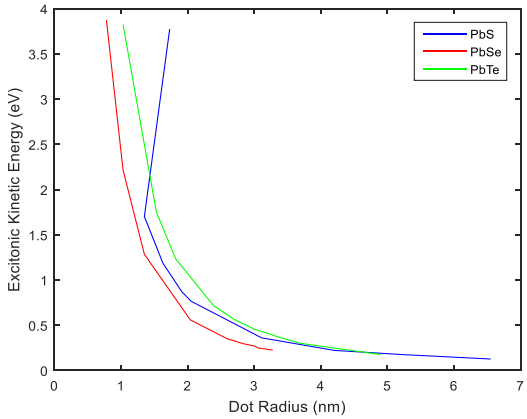

Fig. 8. Excitonic Kinetic Energy versus Dot Radius for PbS, PbSe and PbTe QDs

The high kinetic energies observed in some QD materials imply higher energy levels sufficient enough to inhibit hot carrier cooling rate common in semiconductors and therefore can be considered as new materials to realize the efficient conversion of hot carrier energies. Thus materials which exhibit extremely slow carrier thermalisation can be used for the realization of the hot carrier solar cell which can theoretically achieve very high power conversion efficiencies. The non-zero kinetic energies observed from the Figures above means that charge carriers in the QDs can never be in a state of rest and even in the lowest energy state they possess a non-zero minimum kinetic energy in a manner analogous to particle in a potential well. From the classical point of view charge carriers are said to be in state of constant motion. However, as QDs sizes are further increase the quantization effects on the carrier kinetic energies become smaller. This is because as QDs size is increased towards the weak confinement regime their energy levels move closer leading to near continuum band distribution. If the dot size is increased further towards infinity the kinetic energy will tend to zero and the energy spectrum will deviate from discrete spectrum into a continuous band spectrum and the carrier behavior becomes classical and obeys Newtonian laws.

Also, dependence of kinetic energy on the effective mass strongly indicate that lighter carriers exhibit large energy levels and will have longer lifetime than heavy charge carriers. This in turn means that light and heavy holes possess different energy levels and will therefore thermalise at different rate. For many semiconductor materials the hot carrier population of photo-excited carriers has most of the absorbed photons in the hot electrons due to their small effective mass compared to holes. Consequently, the lower energies associated with holes makes them less significant for the overall conversion of hot carrier energy. In addition, confinement also increases the proximity of electron and hole charge distribution and hence Coulomb interaction which favors the multiple exciton generation is enhanced.

Thus, bandgap which is proportional to the kinetic energy of charge carrier are tunable by varying the confinement dimension of the dot materials. This results in significant increase in energy of band-to-band excitation and hence a blue shift in the absorption and luminescence energies with decreasing size as shown in Figure 9.

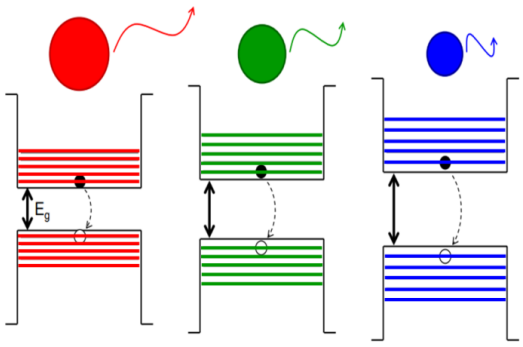

Fig. 9. Blue shift in the absorption and luminescence energies with decreasing

The bandgap modulation by quantum confinement effect is a very unique characteristic that makes QDs a perfect candidate for the high efficiency solar cells, because in principle it is possible to use QDs to build absorber with any desired electronic band-gap to better match different parts of the sunlight spectrum as shown in Figure 10

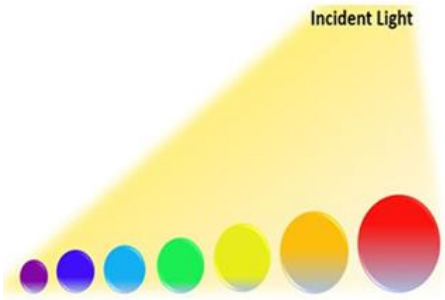

Fig. 10. Absorption of solar spectrum with different size QDs

The result has shown evidence of long carrier lifetime in QDs due to the widely separated energy levels leading to a large gap between the optical and acoustic branches in the phonon dispersion. The observed quantization of energy bands signifies a shift from continuous band distribution common in bulk to discrete energy levels that increases with decreasing dot sizes as illustrated in Figure 11.

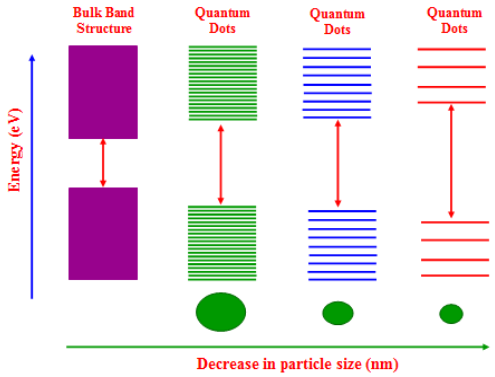

Fig. 11. Energy Levels of QDs with Decreasing Size

The gap in the phonon dispersion in QDs reduces carrier thermalisation rates by removing phonon decay pathways thus creating a phonon bottleneck effects. Consequently, substantial increase in the optically excited hot exciton cooling time is expected in QDs in contrast to sub-picoseconds carrier relaxation timescale reported for the parental bulk. The slow hot carrier cooling dynamics called phonon bottleneck is attributed to the potential mismatch between the wide electronic energy states and phonon energy contrary to the bulk whose energy level coincides with longitudinal optical phonon energy as illustrated in Figure 12.

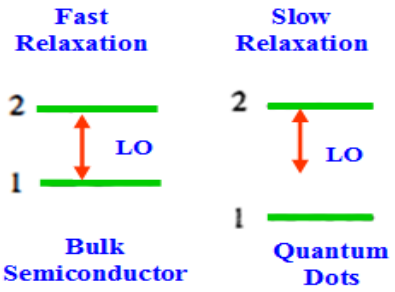

Fig. 12. Optical phonon scattering process in bulk and the quantum dot

Thus, phonon mediated hot carrier cooling can be dramatically suppressed in the QDs, making it possible for the rate of impact ionization (inverse Auger effect) and hot carrier extraction to compete favorably with the rate of hot carrier cooling. The hot carrier thus has life time that is strongly dependent on the QD size, the smaller the size the higher the energy levels and the longer the thermalization time constant. These novel optical properties allows for exploitation of hot carriers that are loss by phonon emissions in conventional devices in which huge amount of high energy photons are wasted as heat. The emergence of QDs opens a lot of opportunities for new design architecture such as hot carrier and the multi exciton generation solar cells that leverage on the size quantization to minimize the thermalization losses and remarkably improve solar to electricity conversion efficiencies.

4.1. Hot Carrier Solar Cell

The hot carrier solar cell is a solar energy converter that utilizes the excess thermal energy of hot carriers that is conventionally wasted as heat to achieve very high power conversion efficiencies in a device that is actually a single junction. Thus making it possible to harvest carriers with a higher energy and subsequently create a higher voltage in the solar cell. Hot carrier solar

cells depend on absorption of energetic photons in an absorber where photo generated hot carriers are extracted through energy selective contacts before they are thermalise with the lattice by phonon emission. The design of hot carrier solar cell requires the absorber material with these properties. Firstly, an absorber with a narrow bandgap semiconductor is required. This allows absorption of a wide range of solar spectrum and hence a high photo current for the device. Secondly, an absorber with extremely slow cooling rate of hot carriers to maintain a hot carrier population for a long time for the hot carriers to be extracted. lastly, an absorber with a selective energy contacts (narrow band electronic contacts) is used which allows only carriers with a specific energy to be extracted in order to prevent heat flow when the hot carrier reservoir comes in contact with a cold one in the metallic electrode as shown in Figure 13.

In this way the increase in entropy on the carrier is drastically minimized. The results have shown that QDs incorporated into absorber material can dramatically inhibit the picoseconds timescale of hot carrier cooling. The slowed of hot carrier relaxation provided by the QD absorber material allows the hot carriers to be extracted while they are still hot leading to actualization of higher photo voltage and hence a higher DC output power.

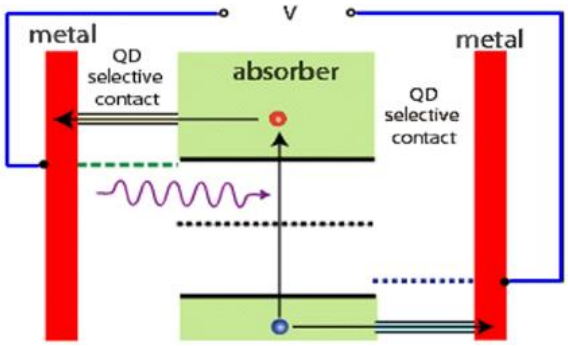

Fig. 13. Hot carrier solar cells

The increase in the output voltage of the solar cell by increasing the extracted work per absorbed photon will results in high solar to electricity efficiency. In this way excess carrier energies above the respective band edges thus contribute to higher conversion efficiencies in relation to that of a conventional cell, which is constrained by the detailed-balance Shockley-Queisser limit.

4.2. Multiple Exciton Generation Solar Cell

The carrier multiplication solar cells are photovoltaic devices designed to enhance solar to electricity conversion efficiencies by utilizing the excess energy in the absorbed energetic photons that would otherwise fizzle out as heat. The results obtained strongly reveal that multiple exciton generation is more efficient in QDs specifically because of the enhanced Coulomb interaction between the carriers and the possibilities of achieving phonon bottleneck effect. Carrier multiplication in QD is known as multiple exciton generation (MEG) because electron–hole pairs exist as exciton. MEG is essentially a process in which hot carrier created by absorption of solar photons higher than the fundamental bandgap energy is utilized to activate two or more charge carriers before they are cool to band edge through phonon emission. Thus an electron from valence band, promoted by absorption of energetic photon makes a transition to a higher energy level in the conduction band and generates one charge carrier but the excess energy would posses high kinetic energy that can be released to excite a second charge carrier through impact ionization. In this way one photon produces two charge carriers as shown in Figure 14. The creation of additional carriers significantly mitigates the spectral losses due to thermalization that characterized the conventional device thus making it possible for the exploitation of supra band photon energies. In this way the excess kinetic energy thus contribute to conversion efficiencies through generation of more carrier rather than heating up the cells. This produces significant

increase in solar to electricity conversion efficiencies in the form of increased photo generated current.

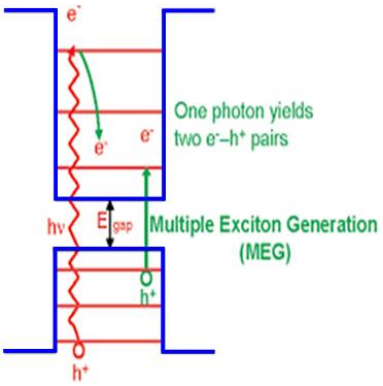

Fig. 14. Multi Exciton Generation Mechanism

In addition, quantization of energy levels in QDs relaxes the crystal momentum conservation. This reduces the threshold energy required for multiple exciton generation process in QDs to the energy conservation limit ($2E_g$) compared to that of the bulk that is too high for practical considerations. The result in Figure 15 shows that QDs posses potential for multiple exciton generation process with reduced threshold energy photons as calculated with Schaller model. An efficient exciton multiplication potential is theoretically observed in PbSe, PbTe, PbS, InAs, and InAs QDs with the threshold photon energy within the energy conservation limit of $2E_g$. Despite being an approximate expression, Figure 15 agrees well with observation by other researchers. Thus the predicted threshold for PbSe is $\sim 3E_g$ as compared to measurements of onset between values of $2E_g$ and $3E_g$ [29] while for InAs it is $\sim 2.05Eg$ as compared to measured values of 2Eg [30].

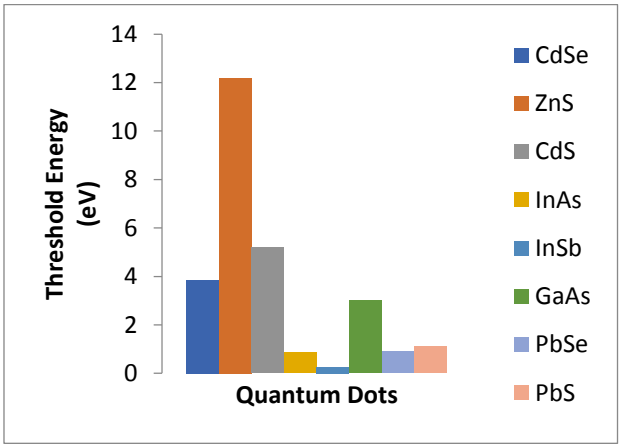

Fig. 15. Threshold Energies for different Quantum Dots

5. Conclusion

QDs exhibit large intra band energy levels that should inhibits the decay of optical phonon and thus promote phonon bottleneck effects due to size quantization effects. This mechanism offers slowed hot carrier cooling and reduces the loss of kinetic energy of the hot carrier into phonon modes and hence offer lifetimes long enough to enable proper exploitation hot carriers. This provides optical pathways that induce significant photocurrent and photo voltage and hence an increase in power conversion efficiencies beyond Shockley and Queisser established limit. It

was theoretically observed that PbSe, PbTe, PbS, InAs, and InSb QDs exhibit exceptional low threshold and high kinetic energies to be considered as new absorber materials for efficient multiple exciton generation and hot carrier solar cell applications that can drive overall conversion efficiency of the hot carrier energy.

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Appendix

Table 1.The Group II-IV QDs material parameters used for the study.

Quantum dots	M_e^x	M_h^x	**$Eg_{(bulk)}$ at 300k**
CdSe	0.13mo	0.45mo	1.74Ev
ZnS	0.34mo	0.23mo	3.68eV
CdS	0.21mo	0.80mo	2.42eV

Table 2.The Group III-V QDs experimental parameters used for the study.

Quantum dots	M_e^x	M_h^x	**$Eg_{(bulk)}$ at 300k**
InAs	$0.02m_o$	$0.40m_o$	0.36eV
InSb	$0.02m_o$	$0.40m_o$	0.17eV
GaAs	$0.06m_o$	$0.51m_o$	1.42eV

Table 3.The Group IV-VI QDs experimental parameters used for the study.

Quantum dots	M_e^x	M_h^x	**$Eg_{(bulk)}$ at 300k**
PbSe	$0.05m_o$	$0.04m_o$	0.27eV
PbS	$0.25m_o$	$0.25m_o$	0.37eV
PbTe	$0.17m_o$	$0.20m_o$	0.32eV