

# Recent Progress in Inorganic Solar Cells Using Quantum Structures

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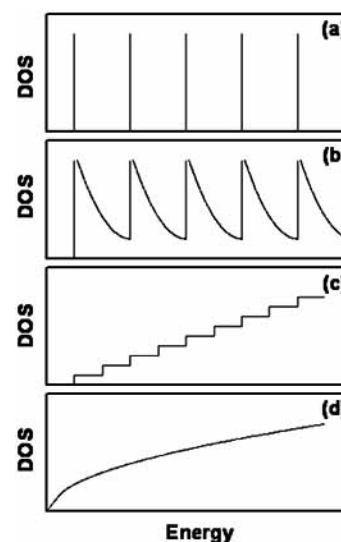
**Abstract:** Thermalization of photogenerated carriers in bulk materials is the main bottleneck for the conversion efficiency of conventional inorganic solar cells. Furthermore, despite extensive research, the achieved conversion efficiency is nearly saturated during the last decade. Therefore, new device concepts to break through the efficiency barrier are highly requested. Nanotechnologies are the building blocks for next-generation solar cells, because low-dimensional quantum structures can possibly reduce thermalization and extend the light absorption range. Hereafter, recently invented inorganic solar cells using quantum structures will be reviewed.

**Keywords:** Inorganic solar cell, multiple quantum well, superlattice, quantum dot, multilayer, nanorod.

## INTRODUCTION

Because of global warming deeply related to emission of CO<sub>2</sub> and oil crisis, energy ranks humanity's top issue for next 50 years. Accordingly, clean, renewable energy including solar, wind and hydrogen energy becomes a prime issue. The solar cells using solar light are promising candidates among renewable energy technologies, because the sun is our primary source of clean, abundant energy. Since 120,000 TW of solar radiation strikes on the surface of the Earth, 10% efficient solar conversion systems covering 0.16% of the land on the Earth would produce 20 TW of power, nearly twice the annual global energy consumption [1].

During the past 9 years, there has been an explosive, worldwide increase in solar module market (~ 40% of an annual increase). Because bulk crystalline silicon (c-Si) modules share 90% of market, the recent shortage of c-Si wafers has increased the production cost of solar modules. In addition, the conversion efficiency of conventional bulk-semiconductor-based solar cells is severely limited by thermalization of photogenerated carriers in lattice; excess photon energy larger than the optical band gap is easily converted into heat via thermal scattering of photo-generated carriers. Hence, the theoretical maximum efficiency of inorganic single-junction solar cells is limited to 30% [2]. Furthermore, the current best efficiency of inorganic single-junction solar cells is 20-25% [1] and it is almost saturated during the last decade. Therefore, new challenges to break through energy loss that stems from thermalization are highly requested. As shown in Fig. (1), low-dimensional-semiconductors can modify the density of states (DOS) by restricting their size to the order of an electron wavefunction. The quantized energy levels in quantum structures become the driving force for next-generation solar cells due to the mitigation of the thermal scattering as well as the control of the light absorption range. In this article, currently



**Fig. (1).** Density of states (DOS) in the semiconductors; (a) 0D (quantum dot), (b) 1D (quantum wire), (c) 2D (quantum well), and (d) 3D (bulk) [1].

developed inorganic solar cell technologies using quantum structures will be revisited. Moreover, the recently issued patents on the topic issued by JP6302840, JP6163962, JP11220150, JP332945, JP9237908, JP62128182, US585-1310, US5496415, US20060050288, US20036670544, US20026437233, US4688068, US20006147296, US20026372-980, and WO0147031, WO9617388 Patent Offices will be reviewed.

## MULTIPLE QUANTUM WELL & SUPERLATTICE

To achieve higher conversion efficiencies, multi-band-gap absorber systems such as multiple quantum wells (MQW) and superlattice (SL) were suggested [3]. Fig. (2a) shows the energy band diagram of a p-i-n type single-junction MQW solar cell. MQW consists of the alternate

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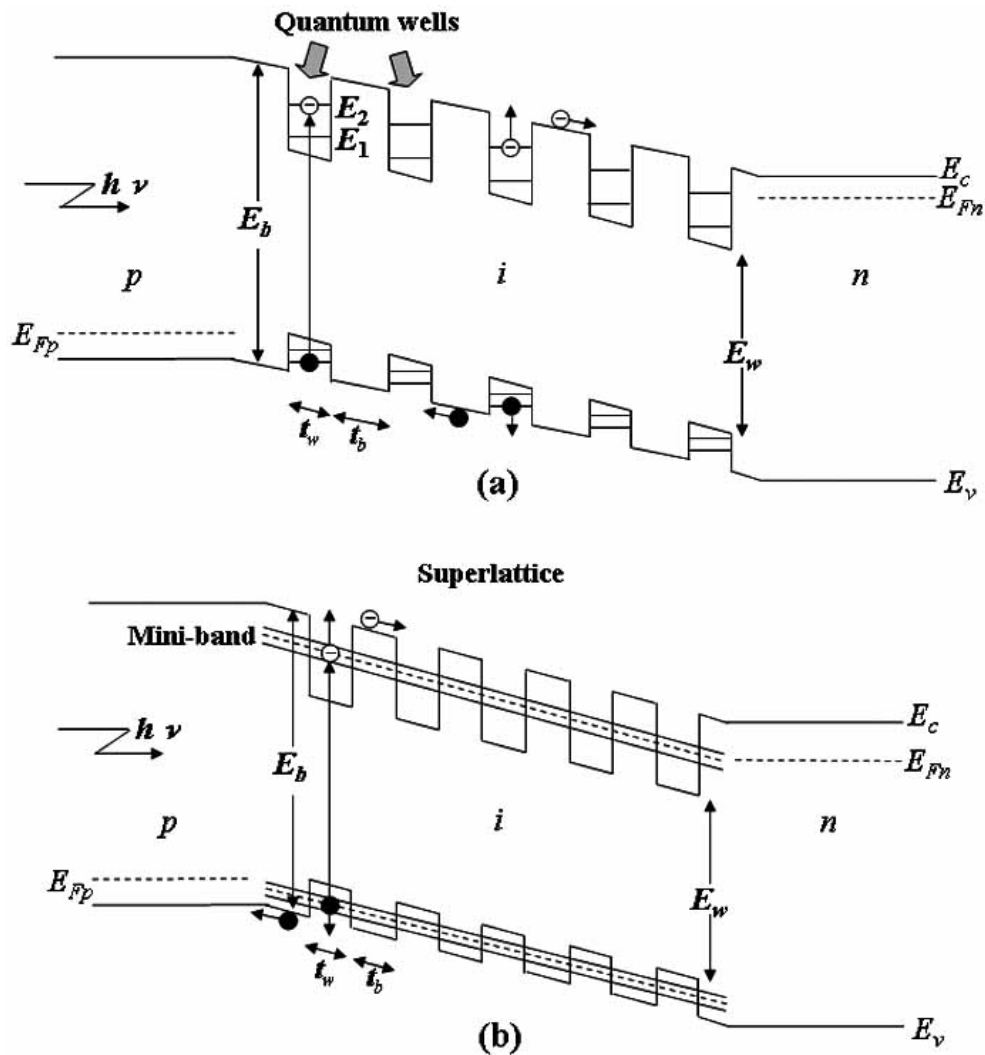


Fig. (2). Energy band diagrams for p-i-n type single-junction solar cells using (a) MQW absorber and (b) SL absorber.

layers of two (or more) different semiconductors having different band gaps; one is the host barrier layer with a wider band gap ( $E_b$ ) and the other is the well layer with a narrower band gap ( $E_w$ ) [4]. MQW is always initiated and terminated with the barrier layer. The thickness of the barrier layer ( $T_b$ ) is usually below 100 nm, but it is not so critical. However, the thickness of the well layer ( $T_w$ ) is important to form quantized energy states in the well [5]. The typical value of  $T_w$  is 5-10 nm. Due to the quantum mechanical effect, the effective band gap of MQW is lower than  $E_b$ , and thus, the MQW solar cells can expand the light absorption range to longer wavelengths. The enhanced well number can offer an improved efficiency by increasing the short-circuit current [6]. In the presence of the built-in electric field in the depletion layer, the photogenerated carriers produced by absorbing extra photons in the well escape to the adjacent barrier layer, and thus contribute extra current with a high escape efficiency at room temperature [7-9]. Accordingly, p-i-n type MQW solar cells patented by [3,10] are better than p-n junction MQW solar cells patented by [11]. The calculated maximum efficiency is about 63% [12].

To avoid defect formation and degradation of the photovoltaic properties, lattice-matched MQW material systems are highly requested as absorbers. Based on the current technology, lattice-matched MQW absorbers with thicknesses up to 1  $\mu\text{m}$  are possible [4]. To minimize the effect of the lattice dislocations, the component materials as well as values of  $T_b$  and  $T_w$  must be selected carefully. The recent patents have claimed the lattice-matched MQW absorbers constructed with: (i) Group III-V compounds [4,11,13-15]; (ii) Group IV alloys such as C [16]; or (iii) Si/Ge [17]. The MQW absorbers can be materialized with quantum dot arrays produced by: (i) a self-assembled growth [18,19] patented by Refs. 15-17; or (ii) a patterning technique patented by [14,20]. The conventional growth techniques for MQW are molecular beam epitaxy (MBE) [9], metal-organic chemical vapor deposition (MOCVD) [21,22], metal-organic vapor phase epitaxy (MOVPE) [7,23] and chemical beam epitaxy (CBE) [8,24,25].

To absorb the incident light more effectively, graded band gap MQW solar cells have been invented by patents [26,27]. The graded band gap MQW can be achieved by changing alloy composition [26] or values of  $T_w$  [27]. The

MQW tandem cells comprised of stacked unit cells in series were claimed by patents [28-30] to provide higher conversion efficiencies by guiding incident light to the appropriate absorbers [1]. Chaffin *et al.* patented the monolithic, multilayer tandem cells alternating p-type MQW and n-type MQW [28], while Freundlich patented double-junction or triple-junction tandem cells using intrinsic InGaAs/GaAs MQW [29,30]. The strain-balanced absorbers constructed with alternate tensile and compressively strained layers have demonstrated remarkably improved photovoltaic performance, and thereby the single-junction cell with a strain-balanced GaAsP/InGaAs MQW absorber recorded the conversion efficiency of 20.0% under AM1.5 (100 mW/cm<sup>2</sup>) irradiation [23]. Furthermore, the strain-balanced cells provide good opportunities for higher efficiency MQW tandem cells [23,31,32]. Recently, it has been reported that incorporation of a distributed Bragg reflector into the strain-balanced absorber leads to the improved quantum efficiency [33]; under AM1.5 irradiation, the conversion efficiency of 21.9% was achieved by the MQW tandem cell consisting of a GaInP top cell and a strain-balanced GaAsP/InGaAs MQW bottom cell with an AlAs/AlGaAs distributed Bragg reflector [34].

In general, the conversion efficiency of inorganic solar cells declines with the increase in operating temperature, which is mainly attributed to the decrease in the optical band gap. In contrast, the MQW solar cells exhibit an excellent temperature dependence because of multi-band-gap structures [35]. Thus, Barnham patented p-i-n type single-junction concentrator solar cells having AlGaAs/InGaAs MQW or InGaAs/InP MQW [14]. The MQW solar cells are also suitable for thermophotovoltaic applications, wherein heat radiation from a combustion process is converted into electricity [36,37]. Also, the MQW solar cells have many advantages for use as space solar cells due to their radiation hardness [38,39].

On the other hand, SL is obtained by reducing  $T_b$ ; with the decrease in  $T_b$ , the tunnel probability of photogenerated carriers between adjacent wells is greatly increased and the carriers are no longer localized in individual wells [40]. Thus, carriers are easily spread throughout whole SL via continuous mini-bands [see Fig. (2b)]. This results in the high conductivity and improved performance of SL solar cells. Saif *et al.* patented the graded band gap SL by keeping  $T_b + T_w$  at the same value; progressively decreasing  $T_w$  of GaAs wells and increasing  $T_b$  of AlGaAs barriers [40]. In this patent, SL tandem cells having a AlGaAs/GaAs SL top cell and a InGaAsP/GaAs SL bottom cell was also invented. These SL solar cells are suitable for use as space solar cells.

The mini-band constructed with quantum dot arrays is often called "intermediate band" [41]. Intermediate band solar cells with many bands are very promising photovoltaic devices (theoretical maximum efficiency: 87%) [42]. However, it is very difficult to materialize such the solar cells, because the significantly high quantum dot density ( $\sim 10^{19}$  cm<sup>-2</sup>) is needed to form the intermediate band. Recently, some attempts to obtain the intermediate band structure using InAs quantum dot arrays embedded in GaAs [43,44], Si/SiO<sub>2</sub> SL [45,46], or Si quantum dots embedded in hydrogenated amorphous silicon-carbide (a-SiC:H) multilayer [47]

have been reported. Concerning the last case, Si quantum dots are formed in Si-rich a-SiC:H sublayers via post-deposition thermal annealing [48].

### CARRIER MULTIPLICATION & HOT-CARRIER

The efficiency barrier of the conventional solar cells caused by thermalization may be overcome by direct carrier multiplication [49-58]. In bulk semiconductor crystals, impact ionization produces carrier multiplication, however, the quantum efficiency is low (1.3 electron-hole pairs per absorbed photon) [50]. In contrast, recent experimental results have reported significantly higher quantum efficiency of carrier multiplication (or multiple exciton generation) in quantum dots [54-57], i.e., 7 excitons per absorbed high energy photon in PbSe colloidal nanocrystals [55]. Quantum confinement in the nano-sized quantum dot quantizes the energy levels for electrons and holes. This discrete energy levels retard hot exciton cooling and improve multiple exciton generation [1]. The calculated maximum efficiency for nanocrystal-quantum-dot solar cells in the presence of carrier multiplication is 42% [59].

The hot-carrier solar cell using selective energy contacts has also been suggested in order to reduce the energy loss due to thermalization [60-62]. The selective energy contacts must extract photogenerated hot-carriers from the absorber before the thermal scattering of excess energy. Thus, the low-dimensional materials such as quantum dots are promising as the selective energy contacts. The single-junction hot-carrier solar cell theoretically provides the maximum conversion efficiency of 66% under the AM1.5 illumination [60].

Although there have been intense attempts, both the concepts of carrier multiplication and hot-carrier solar cell have not been materialized in devices yet. In addition, most of related technologies have only been published in academic journals.

### PROTCRYSTALLINE SILICON MULTILAYER

Thin-film Si solar cells using hydrogenated amorphous silicon (a-Si:H)-based absorbers are promising, because of remarkably low consumption of raw Si material, large-scale deposition and low-temperature production. Despite high interest in thin-film Si solar modules, a major obstacle remains to their commercialization. Specifically, light-induced degradation, which originates from the photo-creation of dangling bonds (DBs) accomplished by the nonradiative recombination of photogenerated electron-hole pairs (Staebler-Wronski effect) [63] limits the efficiency of a-Si:H-based solar cells. During the past 30 years, there have been extensive investigations aiming to reduce the Staebler-Wronski effect. The hydrogenated protocrystalline silicon (pc-Si:H) material is an unique hydrogen-diluted a-Si:H material existing just below the threshold of the a-Si:H-to-hydrogenated microcrystalline silicon ( $\mu$ c-Si:H) transition. The pc-Si:H is very stable compared to conventional a-Si:H. However, it is very difficult to control the deposition of a pc-Si:H single-layer due to its thickness and substrate dependence of microstructure.

To improve the controllability for the pc-Si:H material, novel alternately hydrogen-diluted pc-Si:H multilayer

absorbers possessing quantum dots have been developed [64-71]. Using a photo-assisted chemical vapor deposition (photo-CVD) technique, alternately hydrogen-diluted i-pc-Si:H multilayers was prepared by modulating the mass flow control of the hydrogen dilution ratio ( $H_2/SiH_4$ ) between 0 and  $R$  under continuous UV light irradiation [65]. During a gas control of  $H_2/SiH_4 = 0$ , a low hydrogen-diluted a-Si:H sublayer ( $S_L$ ) is deposited because of the residual gas in the reaction chamber. During a gas control of  $H_2/SiH_4 = R$ , a highly hydrogen-diluted sublayer ( $S_H$ ) is deposited. Under the optimized condition ( $H_2/SiH_4 = 20$ ), the thickness ratio of  $S_L$  to  $S_H$  measured by spectroscopic ellipsometry is 10 nm to 32 nm. Due to the alternate hydrogen dilution, the pc-Si:H multilayer is remarkably more controllable than pc-Si:H single-layers. In addition, the i-pc-Si:H multilayer preparation via the modulation of hydrogen dilution can be easily realizable via the conventional plasma-enhanced chemical vapor deposition (PECVD) technique [69].

The Raman spectrum for the multilayer deposited on quartz does not exhibit any distinct fraction of  $\mu$ c-Si:H phase (transverse optical mode of c-Si near  $520\text{ cm}^{-1}$ ), because  $S_L$ s interrupt the columnar growth of  $\mu$ c-Si:H [71]. Hence the i-pc-Si:H multilayer displays a Raman spectrum similar to that for an undiluted i-a-Si:H single-layer. These spectra can be deconvoluted to four Gaussian peaks corresponding to a-Si:H networks. The detailed shape analysis shows the considerably improved medium-range-order in the a-Si:H matrix via alternate hydrogen dilution [69,71].

Photoluminescence (PL) and Fourier transform infrared (FTIR) spectroscopy measured at room temperature [67,68] as well as the transmission electron microscopy (TEM) image [70] produce strong evidence that isolated nano-sized (3-5 nm in diameter) crystalline Si (nc-Si) grains are embedded in  $S_H$  [see Fig. (3)]. An hydrogen (H)-rich a-Si:H grain boundary layer wraps each nc-Si grain. The grain-boundary layers are the most defective component in the multilayer.  $S_L$ s that hinder the percolation path of  $\mu$ c-Si:H possess larger disorder in the a-Si:H matrix than that of  $S_H$ s. All interfaces are graded by hydrogen.

Since the pc-Si:H multilayer exhibits a fast light-induced metastability (stabilization time = 12 h) with a low

degradation (degradation ratio = 13.4%), highly stabilized p-i-n type single-junction solar cells (stabilized efficiency = 9.0%) were achieved without using any back reflector [67,71] via standard 1-sun ( $AM1.5$ ,  $100\text{ mW/cm}^2$ ) light-soaking at  $50^\circ\text{C}$ . The key technologies to success were the good p/i interface formed using a p-type a-SiC:H double layer structure [66] and the good light-induced metastability of the optimized pc-Si:H multilayer absorber [68]. Also, the pc-Si:H multilayer solar cells provide a fast, perfect recovery via thermal annealing [69,71]. Taking into account the thickness of  $S_L$  ( $\sim 10\text{ nm}$ ) and  $S_H$  ( $\sim 32\text{ nm}$ ), the average distance between nc-Si grains in the multilayer should be much shorter than the ambipolar carrier diffusion length in a-Si:H ( $> 100\text{ nm}$ ). Thus, some photoexcited carriers generated in the a-Si:H matrix of all sublayers can diffuse to grain-boundary regions surrounding nc-Si grains. Several recent reports have suggested "fast" and "slow" metastable defects in the a-Si:H matrix [72-74]. From the fast metastability and annealing behaviors of pc-Si:H multilayer solar cells, the vertically regular distribution of the isolated nc-Si grains and the improved medium-range-order in the a-Si:H matrix are considered to localize the photocreation near the grain boundary regions, and thereby suppress photocreations of slow metastable defects in the pc-Si:H multilayer. The isolated nc-Si grains tend to act as radiative recombination centers of captured carriers, which may contribute to the good stability.

In the case of thin-film Si solar cells, tandem structures provide a high stabilized efficiency; the reduced thickness of a-Si:H-based absorber in the top cell for current-matching effectively suppresses the Staebler-Wronski effect [75]. Because the pc-Si:H multilayer has a slightly wider optical band gap (effective band gap  $> 1.7\text{ eV}$ ) than conventional a-Si:H layers ( $\sim 1.7\text{ eV}$ ), the pc-Si:H multilayer solar cell is promising as a top cell for a high efficiency tandem cell [71]. The pc-Si:H multilayer/ $\mu$ c-Si:H ( $\sim 1.1\text{ eV}$ ) double-junction tandem structure provides the high stabilized efficiency over 11% with the very low degradation ratio ( $< 4\%$ ) [76].

#### NANOROD-SHAPED HETEROJUNCTION

Donor-acceptor solar cells composed entirely of inorganic nanocrystals spin-cast from solution have been

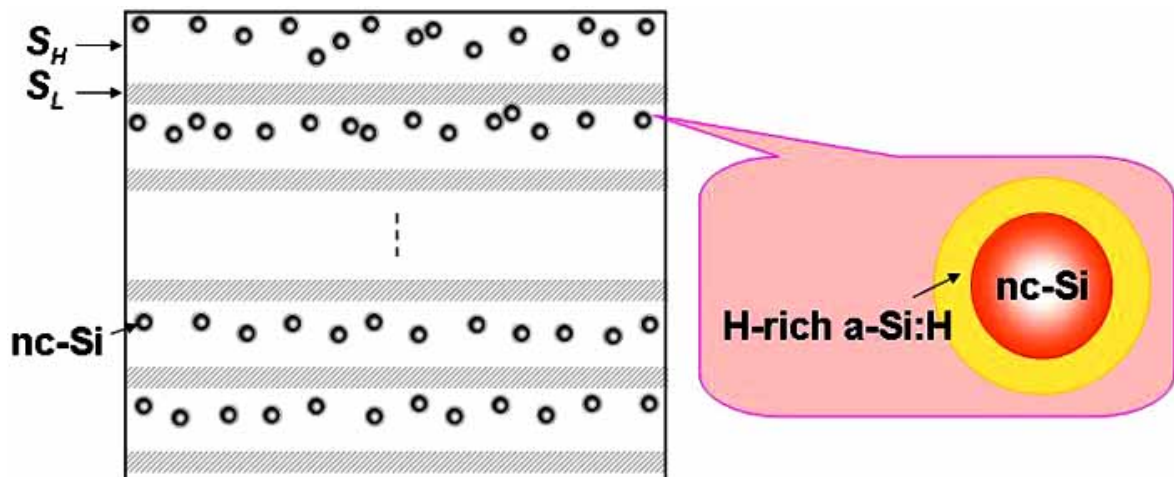


Fig. (3). Proposed structure of the pc-Si:H multilayer [68].

developed [77]. The solar cells use the nanorod-shaped CdTe/CdSe nanocrystal heterojunction. Each ultrathin (~ 100 nm) nanocrystal was spin-cast from a filtered pyridine solution. This technology provides large area, flexible thin films of densely packed nanocrystal on virtually any substrate. High quality bilayer structure was formed via brief annealing of the CdTe films for 15 min at 200°C before the subsequent deposition of the CdSe film. Fig. (4) displays the energy band diagram for the nanorod-shaped CdTe and CdSe films. From this figure, photogenerated electrons find lower energy states in the CdSe films, while holes find lower energy states in CdTe. Charge transfer in CdTe/CdSe heterojunction is analogous to that in organic donor-acceptor solar cells; carrier extraction is not driven by the built-in electric field formed at the depletion region, but it is driven rather by the breaking apart of a diffused bound exciton (coupled electron-hole pairs with no net electrical charge) at type II heterojunction [78,79]. Post-deposition sintering enhances the photovoltaic performance, allowing for an air-stable power conversion efficiency up to 2.9% under AM1.5 irradiation.

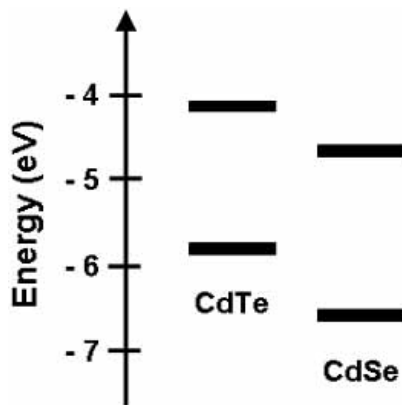


Fig. (4). Energy diagram of valence and conduction band levels for nanorod-shaped CdTe and CdSe films [77].

## CURRENTS & FUTURE DEVELOPMENTS

The recent progress in inorganic solar cells using quantum structures is reviewed. Incorporation of MQW, SL, and quantum dots into photovoltaic devices leads to a spectacular improvement of the theoretical maximum efficiency, compared to the conventional bulk-semiconductor-based solar cells. In addition, it is experimentally proved that the inclusion of isolated quantum dots in the pc-Si:H multilayer dramatically reduces the Staebler-Wronski effect in thin-film Si solar cells. The nanorod-shaped donor-acceptor solar cells also exhibit the stable performance in air.

In point of fact, the currently achieved efficiency from the solar cells using the quantum structures cannot come up with the theoretical maximum value, but it is rather lower than the efficiency of the conventional bulk-semiconductor-based solar cells. Therefore, grand challenges to reduce the gap between the ideal and real values of the conversion efficiency are requested. Besides, large-area, cost-effective, and highly reproducible fabrication processes should be developed for mass production.

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