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Effect of Nanostructure on Carrier Transport Mechanism of III-Nitride and Kesterite Solar Cells: A Computational Analysis

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ABSTRACT In this work, the computational analysis on use of nanostructures to both III-Nitride and Kesterite solar cell are presented and compared. The scope behind the comparative analysis of III-nitride and kesterite material based solar cells is due to their excellent material properties, which made them suitable as an absorber for solar cells. In III-nitride based solar cells, stress induced polarization charges play an important role in carrier dynamics of cell, whereas, kesterite based cells suffer from detrimental effect due to its in-house defects. Here, an analysis is carried out to understand the above mentioned important effects, i.e., carrier transport mechanisms under the influence of polarization charges in III-nitride materials and in presence of defects in kesterite materials with nanostructures. It is anticipated that, nanostructures provide lots of advantages over bulk layer in order to overcome the detrimental effects in solar cells. A detailed comparative analysis of both type of solar cells are carried out. Finally, quantum efficiency measurement is carried out in order to observe the range of light absorption in each structures. It is quite interesting to observe that nanostructure doesn't boost the performance of the solar cell unless until the device is engineered properly. It is purely depends on carrier recovery and transport from the nanostructures.

INDEX TERMS Nanostructures, quantum well, CZTS, III-nitride, CZTSe, quantum efficiency.

I. INTRODUCTION

Ranging from few watts of electricity generation to multi-megawatt power station, harnessing of energy from sunlight using semiconductor device is one of the optimum solution to fossil fuel crisis. As the dominant material used in the today's solar cell, crystalline silicon, doesn't perform well and technology has saturated, the cost has become a road-blocking factor towards generation of multi-megawatt energy generation. However, looking towards the surging global demand and compound annual growth rate of last decade, next generation such as second and third generation photovoltaic technology is given more importance in recent days [1]. One such technology to achieve high efficiency is the use of III-nitride semiconductor materials. Indium Gallium Nitride ($In_xGa_{1-x}N$) is a highly emerging material, belongs to III-nitride family with band gap ranging from 0.64 to 3.4 eV. One of the best ability of the material is to absorb nearly whole solar spectrum to increase the conversion efficiency copiously [2], [3]. Since past few years, $In_xGa_{1-x}N$ material has been showing its potential for different optoelectronic and power electronic applications [4]. This motivation is driving immense scientific interest to develop high-performance solar cells using $In_xGa_{1-x}N$ material. Additionally, this alloy has direct band gap with high carrier mobility, drift velocity, high saturation velocity, stable radiation resistance, thermal stability [5], [6] and optical absorption of 10^5 cm^1, which add more interest to explore further for next generation photovoltaic (PV) applications [2], [3], [7]. In spite of these success stories, the material suffers with a low power conversion efficiency due to several issues [8], [9]. One of the key issues is stuck of immobile charge carriers along the heterojunction interface [10], [11]. In III-nitride material, spinodal decomposition occur at higher 'In' compositional level, leading

to the formation of In-rich and Ga-rich regions in planar GaN/InGaN junction, which lead to stuck of immobile charge carriers at the interface of GaN/InGaN heterojunction, known as polarization charges (PCs) [11], [12]. Depending upon the growth quality of InGaN upon GaN substrate, the polarization charges can be of two types. Firstly, due to intrinsic symmetric of bonding among substrate and overlaying material, inherent immobile charge carriers are stuck along the interface, known as spontaneous polarization charges [12], [13]. Secondly, piezo-electric polarization charges are due to stress generated lattice deformation which takes place along the interface [12]. This polarization induced electric field changes the band bending and affect the carrier transport mechanism in solar cell. Use of nanostructure brings a new possibilities to solar cell as thin layers add low stress as well as polarization charges [7], [14], [15], [16]. Previously, authors have also studied the effect of polarization charges in GaN/InGaN solar cell, where it is observed to be detrimental for solar cell performance. The performance mainly affect due to presence of unfriendly polarization charges in space charge region, which reduces the total electric field across the depletion region [17]. Later on, nanostructure is introduced instead of bulk InGaN layer upon GaN substrate in order to reduce defect density and stress in overlying layer [18]. The analysis was also carried out by many research groups including [19], [20], [21], [22], [23]. The important findings of high performance nanostructure solar cells is drawn as compared to planar counterparts.

In other hand, A thin film Cu_2ZnSnS_4 (CZTS) solar cell is proposed as an emerging alternative candidate for photovoltaic applications because it has very similar properties as of Cupper Indium Gallium Selenide (CIGS) material and it also replaces the expensive material 'In' and 'Ga' with cheap Zinc and Tin leading to significant reduction in module price. The kesterite-type CZTS is a derivative of two indium cations in the tetragonal unit cell of a chalcopyrite-type lattice are substituted for two tin cations, while the other two indium cations are substituted for two zinc cations [24]. Due to easy of fabrication, thin film, high efficiency and low cost, more attention is paid to explore this material in recent years. Additionally, CZTS (Kesterite) material with large absorption coefficients ($> 10^4$ cm^{-1}), ideal direct band-gap (1.45 eV) and non-toxic elements made researchers more attractive towards achieving high efficiency [25], [26]. It's considered as one of the best alternative towards Copper Indium Gallium Selenide (CIGS) and Cadmium telluride (CdTe) solar cell due to their material scarcity and toxicity issues. However, the best CZTS solar cell (pure sulfide) so far, reaching 11% power conversion efficiency [27]. This limit results from compromise in the choice of bandgap, i.e., inability of absorbing photons with energy lower than the absorber band gap. Additionally, recombination rate are quite high in CZTS type of absorber due to higher numbers of in-house defects. Nanostructures are well known to allow the absorption of photons with energies lower than the band gap of absorber material. In fact, theoretical efficiencies of nano structured solar cell is about 42% [14]. Application of nanostructured carrier confinement techniques otherwise known as quantum wells have the potential to enhance the performance of solar cell. Quantum well is a potential well which confines photo generated carriers such as electrons and holes to two dimensions, so as to increase the gain and efficiency of solar cell. Many literature have investigated the pros and cons of incorporation of QWs in to III-nitride materials in order to reduce the stress and polarization effects [12], [28], [29]. In the other hand, nanostructured kesterite solar cell is a less explored area and reported by very few research community [30]. It's concluded from the literature that incorporation of Multiple QWs (MQWs) have two primary effects: (i) the short-circuit current (J_{sc}) increases because of the additional absorption of low-energy photons in the lower band gap QWs; (ii) the open-circuit voltage (V_{oc}) decreases because of the increased recombination of carriers trapped in the QWs. The question that still remain is whether efficiency of CZTS and GaN/InGaN based solar cell can be increased as comparable to traditional or bulk solar cell. A clear understanding on carrier dynamics, stress, possibilities of defects in both type of solar cells with nanostructure is necessary. In this article, authors have taken an attempt to computationally analyze the mechanisms happening in solar cell with nanostructure in order to conclude how QWs affect the performance of both III-nitride and kesterite solar cells. Additionally, a comparative study with bulk material is also carried out to observe the optimum performance of the QW with CZTS and GaN/InGaN materials.

II. DEVICE STRUCTURE & SIMULATION

Here, an attempt has been taken to analyze the carrier transport mechanism in both material based nanostructured solar cells. As carrier dynamics are bit different from traditional nanostructure solar cell due to polarization charges and defects, it is necessary to understand its effect on solar cell performance. Three different structures of both GaN/InGaN and kesterite solar cells are considered such as (i) QW GaN/InGaN planar solar cell (ii) QW CZTS/CZTSe solar cell (iii) Kesterite bulk solar cell. Since many literature are reported on bulk GaN/InGaN hetero-structure, here bulk III-nitride case is not taken into consideration. The planar Multiple-QWs (MQWs) structure consist of n-GaN outer layer of thickness of 40 nm followed by three GaN/i-InGaN QW of 7 nm/3 nm of thickness making a total of 30 nm, p-GaN core thickness of 100 nm, respectively as shown in Fig. 1(a). III-nitride crystallographic structure consists of complex structure with different types of planes as facet, but in this study only polar {000-1} and no-polar {0001} facets are taken into consideration because in planar growth of GaN/InGaN QWs, semipolar facets are normally silent and don't add much effect to the device. Additionally, the device structure for MQW CdS/CZTSe/CZTS/CZTSe/CZTS solar cell is shown in Fig. 1(b). The proposed structure consists of n-type CdS ($1x10^{15}$ cm^3) of 100 nm, p-type CZTS

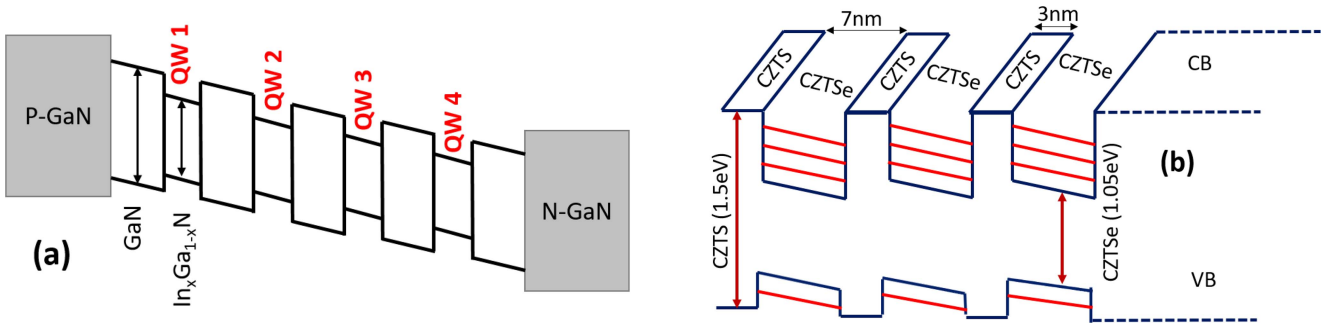

FIGURE 1. (a) QW GaN/InGaN planar solar cell (b) QW CZTS/CZTSe solar cell.

($5x10^{18}$ cm^3) bulk of 500 nm and the intrinsic region containing 'n' numbers of CZTS/CZTSe layers stacked to form QWs. Subsequently, a 200 nm thick zinc oxide (ZnO) is used as a window layer to improve the light scattering. CZTS is an intrinsic p-type semiconductor and this native p-type conductivity was attributed to the low formation energy of acceptor defects. The traditional bulk structure of ZnO/CdS/CZTS and ZnO/CdS/CZTSe [31], [32], [33], [34] have been considered for a comparison study with nanostructure solar cell. The thickness of CZTS and CZTSe bulk layer is considered as 2 micrometer, whereas, CdS and ZnO layer for both the structure is 0.1 micrometer. All the structures are simulated using ATLAS TCAD Tool [35]. The Poisson and carrier diffusion equations are solved using bi-conjugate gradient stabilized iterative method (BiCGST) considering the composition dependent material parameters calculated by linear interpolation between GaN and InN parameters [36], [37]. The simulator employs 6x6 k.p model, which was developed for wurtzite semiconductor material to calculate energy band structure of solar cell.

III. RESULT AND ANALYSIS

Fig. 2 shows the polarization charges along the GaN/InGaN hetero-interfaces in both {0001} and {000-1} crystallographic orientation. The detail derivation and theory of polarization charges can be found in our earlier study [12], [28]. It is very clear that polarization charges have a significant effect on the carrier dynamics of solar cell. Hence, in this investigation, both polar and reversed polar effect are considered in order to get a clear observation. Fig. 2(a) Shows the polarization charges along {0001}, which is detrimental for carrier collection [28]. However, polarization charges along {000-1} orientation is quite useful for carrier separation from each side of the junction. These polarization charges are added to MQW based solar cell of GaN/InGaN materials. The effect of polarization charges can be seen from the energy band diagram shown in Fig. 3.

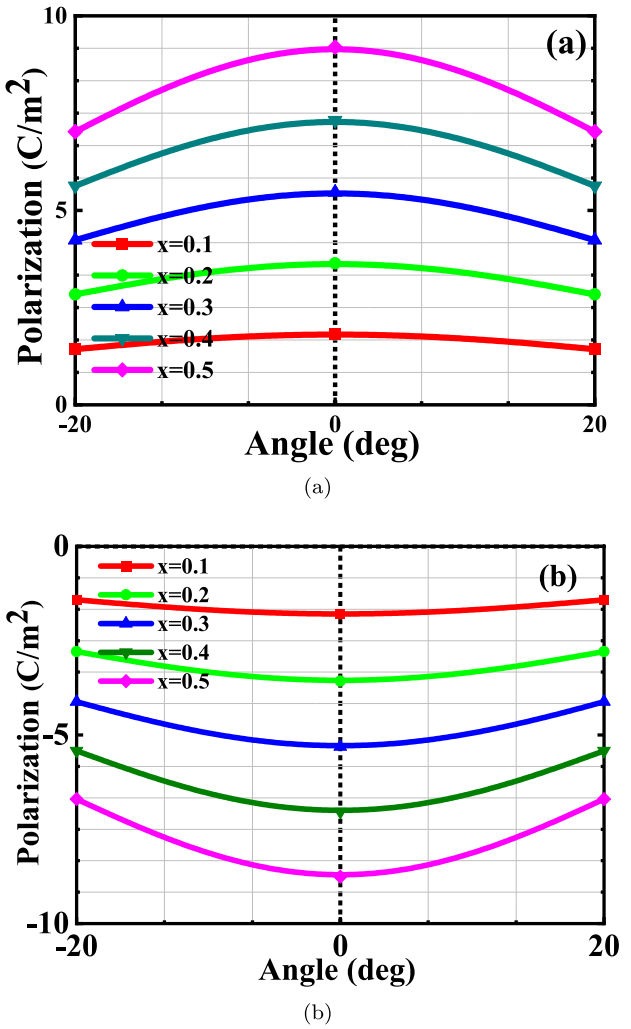

FIGURE 2. Polarization charges along (a) {0001} and (b) {000-1} interface of GaN/InGaN.

Fig. 3(a) shows the energy band diagram of 4 QW GaN/InGaN solar cell along {0001} crystal orientation. The band bending of all 4 QWs are going downwards because of applied bias. Three different Indium (In) composition is taken in to consideration and it is observed that higher 'In' content leads to lower in band bending which affects the carrier separation from the QWs. Additionally, lower 'In' content ensure lower band gap and absorbs a broad spectrum of photons. The band bending of the individual QWs is shown in the Fig. 3(a) insets, which confirms the detrimental effect of electric field. The origin of this detrimental electric field is reversed polarization charges. The polarization

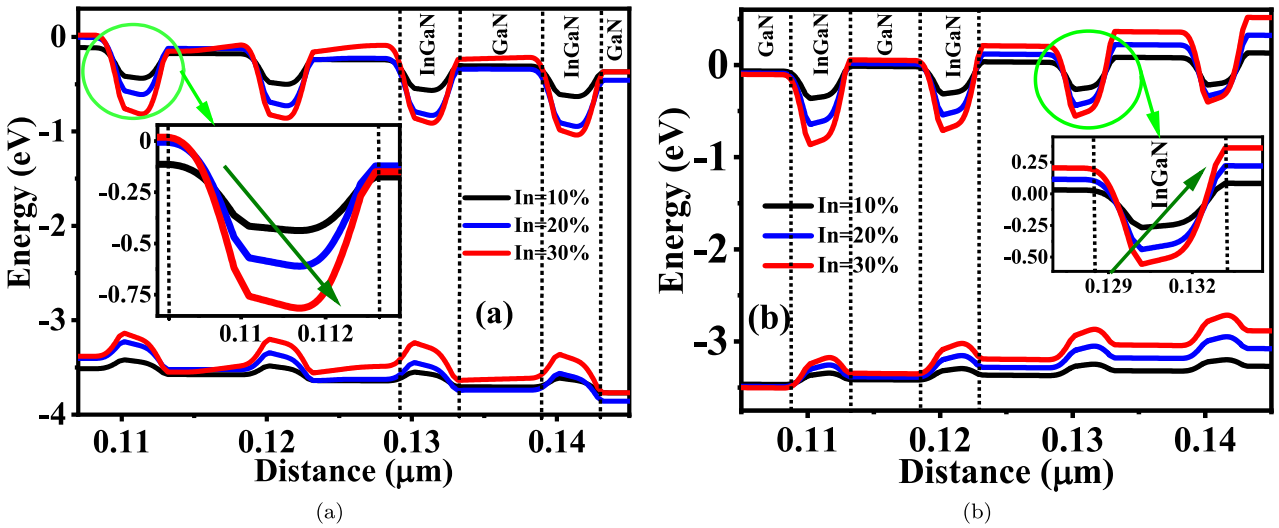

FIGURE 3. Energy band diagram of 4 QW based GaN/InGaN solar cell along (a) {0001} and (b) {000-1} growth direction.

charges along {0001} orientation is exactly opposite to in-built electric field of space charge region and sits along the interfaces also. Hence, overall immobile charge carriers are degrading due to neutralization of depletion charges by polarization charges. Hence, electric field along the interface degrading drastically and affect the carriers separation in QWs. The opposite effect of polarization charges can be seen from Fig. 3(b.) It shows the energy band diagram of 4 QW GaN/InGaN solar cell along {000-1} orientation. From the individual band bending of QW, it can be observed that the band bends towards up (shown in arrow mark), which enhance the carrier separation process due to high electric field. The physics behind this phenomenon is that polarization charges is in the same polarity with the depletion region charges. Hence, overall accumulation of charges are higher as compared to the space charge region, which increases the electric field along the interface and swift out more carriers to other side. The arrow mark in the inset of Fig. 3(a) and (b) show the detrimental and incremental effect of band bending. This type of analysis can also be found out from our previous report [22], [38].

Fig. 4 shows the electric field in 4 QWs in GaN/InGaN solar cell along {000-1} direction. Since {0001} orientation is detrimental for the solar cell physics, analysis on the same is not shown here and similar analysis on {0001} orientation can be found from [23]. The highlighted area shows the high electric field along all QWs, which shows the special effect of nanostructure design in III-nitride materials. This high electric field is originated from the incremental polarization charges, which comes from strain and presence of polar molecules along the GaN/InGaN interface. This type of effect is detrimental in bulk case, where as it's beneficial if nanostructure is introduced instead of bulk material. Additionally, similar type of effect can be found only from III-nitride based devices due to the presence of polar molecules. Hence, designing nanostructure based solar

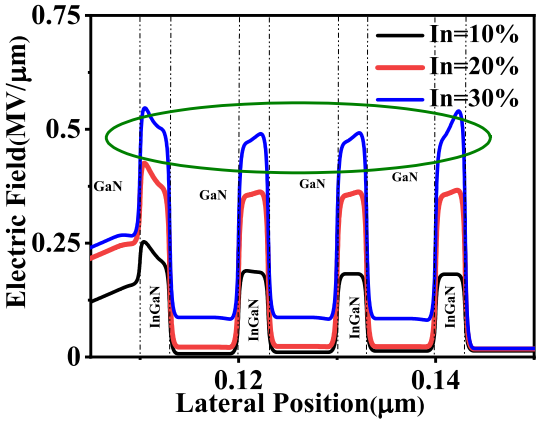

FIGURE 4. Electric field in 4 QW GaN/InGaN solar cell along {000-1} growth orientation.

cell with III-nitride material will provide extra mileage for solar cell towards achieving higher efficiency. In the other hand, kesterite materials are cost effective as compared to III-nitride solar cell as well as environment friendly. The dimension of the QWs are maintained same as GaN/InGaN QWs in order to compare the performance of the device. All above performance measuring parameters of CZTS/CZTSe solar cell are discussed and compared with GaN/InGaN as below.

Fig. 5(a) shows the energy band diagram of CZTS/CZTSe solar cell. Since kesterite materials don't contain polar molecules, effect of polarization charges are negligible here and not taken into consideration. Hence, band bending is due to only space charge region molecules comparatively less to GaN/InGaN band bending. In case of GaN/InGaN QWs, presence of polarization charges enhances the electric field and band bending is more which in turn enhances the carrier separation and collection. Fig. 5(b) shows the electric field along the QWs of CZTS/CZTSe solar cell. It

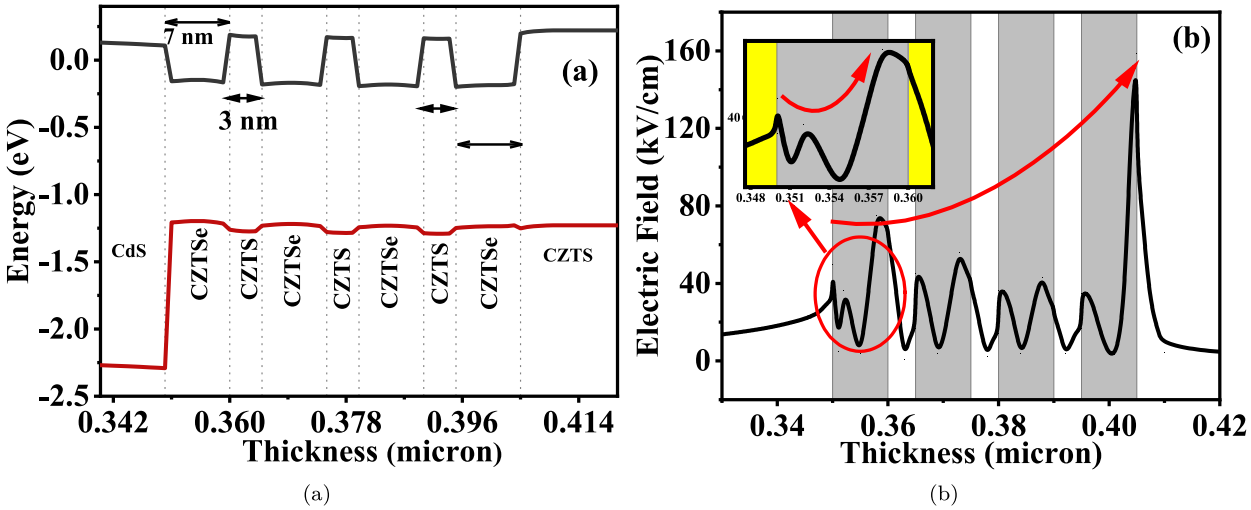

FIGURE 5. (a) Energy band diagram and (b) electric field of 4 QW CZTS/CZTSe solar cell.

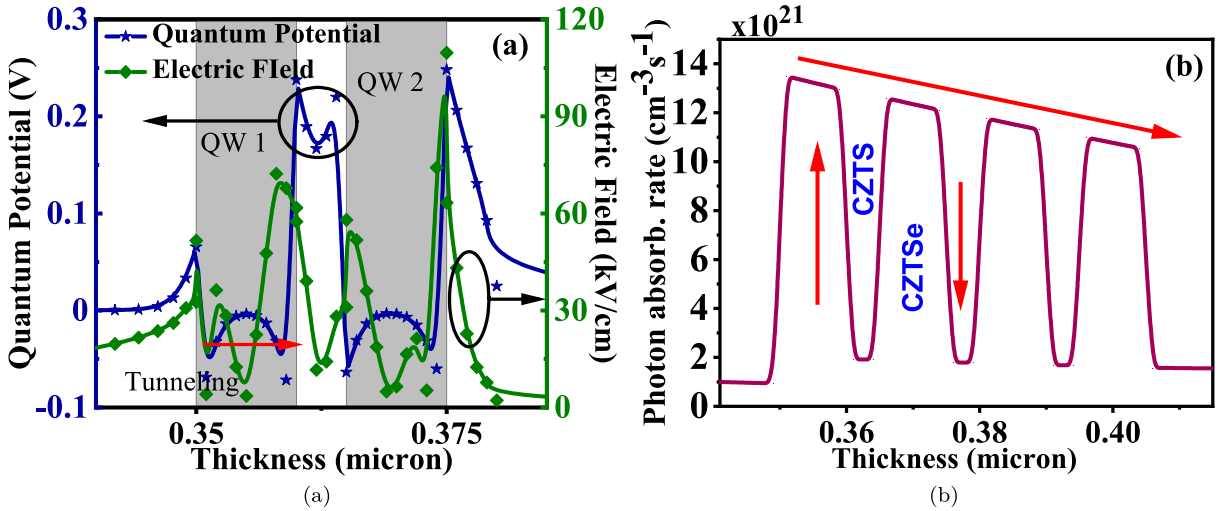

FIGURE 6. (a) Quantum Potential and Electric field (b) photon absorption rate in CZTS/CZTSe 4 QW solar cell.

is observed that the shape of electric field of CZTS/CZTSe is quite similar with GaN/InGaN solar cell and only differs with magnitude and stiffness (shown in red arrow mark). The magnitude of electric field in GaN/InGaN solar cell is higher than the CZTS/CZTSe solar cell. Hence, it can be concluded that carrier separation and collection is quite efficient in case of GaN/InGaN solar cell as compared to CZTS/CZTSe solar cell.

Fig. 6(a) shows the pictorial representation of quantum potential and electric field inside 2 QWs. Whereas, Fig. 6(b) shows the photon absorption rate along the QWs of CZTS/CZTSe solar cell. It is clear that photon absorption rate is always high inside the QWs and decreases with increase in number of QWs. The quantum potential curve shows an additional mechanism of tunneling inside the QWs, where electric field is high.

Fig. 7(a & b) show the energy band diagram of CZTS and CZTSe bulk solar cell. In case of nanostructure solar cell, the thickness of CZTS and CZTSe layer is 7 nm and 3 nm respectively, whereas in case of bulk solar cell the absorber layer thickness is taken as 2 micrometer. The energy band diagram is taken with different applied voltage to observe the conduction band bending. It is clear that conduction band bending is suitable for carrier transport at higher bias voltage.

The external quantum efficiency or spectral response (EQE) of a solar cell is defined as the ratio of the number of electrons generated from the solar cell by an incident photon of a given wavelength. Here three different case is considered to analyze the photon absorption rate and recombination rate of each solar cell. Fig. 8 shows the EQE of 4 QWs GaN/InGaN, 4QW CZTS/CZTSe, Bulk CZTS and Bulk CZTSe solar cell. It is observed that EQE is high up to 0.78 micron then slowly degrades to zero in case of GaN/InGaN solar cell due to high 'In' content of 30% . As 'In' content increases, bandgap of InGaN layer

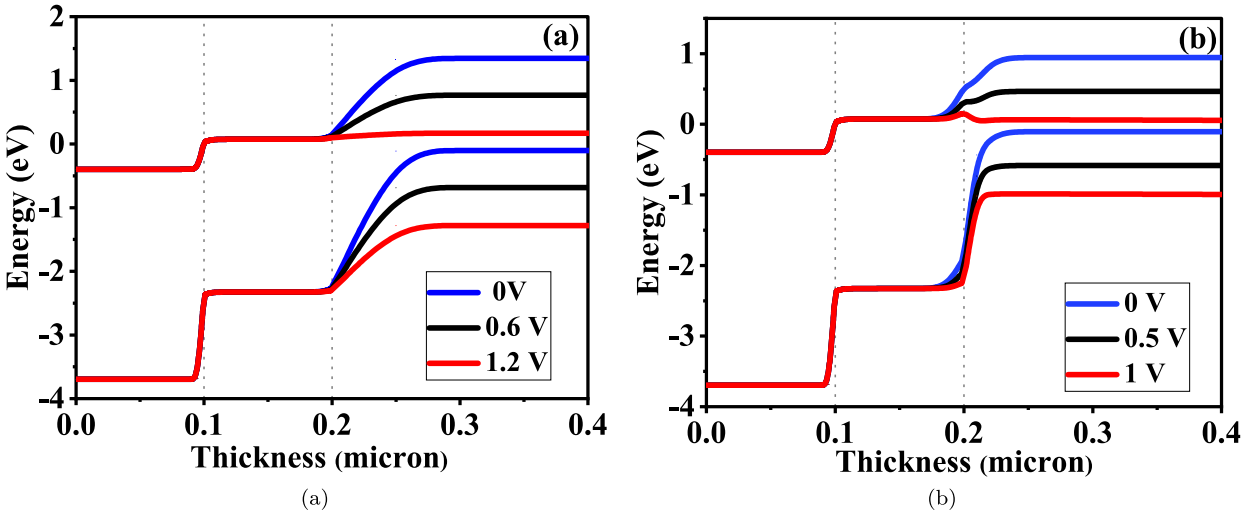

FIGURE 7. Energy band diagram of bulk (a) CZTS (b) CZTSe with different bias voltage.

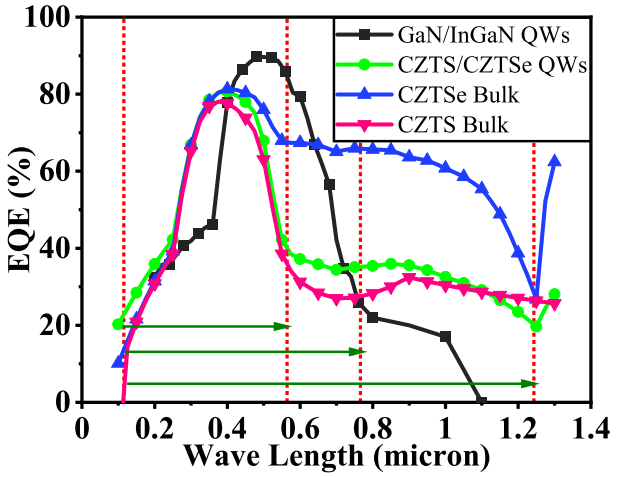

FIGURE 8. External Quantum Efficiency curve of GaN/InGaN QWs, CZTS/CZTSe QWs, CZTS Bulk, and CZTSe Bulk solar cell.

also changes, which further absorb only higher energy photons. But, EQE reaches peak at 0.5 micron to 90% due to incremental effect of polarizatrion charges along {0001} orientation. Additionally, it is also observed that the EQE curve for CZTS/CZTSe 4QW solar cell goes up to 1.2 micron which is more than GaN/InGaN case. It is anticipated that, the enhance of photon absorption may be due to the use of low band gap materials such as CZTS and CZTSe. However, peak of CZTS/CZTSe QW solar cell reaches only up to 80% at 0.4 micron. It is also seen that CZTS/CZTSe QW solar cell maintains high EQE for less wavelength as compared to GaN/InGaN solar cell. More than 50% EQE is maintained from 0.4 to 0.7 micron in GaN/InGaN QWs whereas it is maintained from 0.3 to 0.5 micron in case of CZTS/CZTSe QW solar cell. EQE for CZTS solar cell is quite similar to CZTS/CZTSe QW solar cell. However, EQE for CZTSe solar cell is very high as compared to all above mentioned cases. It maintains more than 50% from 0.3 to 1.1 micron wavelength. Hence, QE is quite significant in case of bulk CZTSe solar cell. It may be concluded that inclusion of nanostructure reduces QE which may be due to high recombination rate inside the QWs.

A summarized table of performance is given in Table 1 for all 4 structures. It is quite clear that {0001} orientation based solar cell shows a poor performance as compared to {000-1} plane. The reason for the same is discussed in the Fig. 3 and 4. It is also observed that J_{sc} is very high in case of CZTS/CZTSe solar cell as compared to GaN/InGaN solar cell. However, V_{oc} for CZTS/CZTSe solar cell is less than GaN/InGaN solar cell. It is anticipated that due to low bandgap, V_{oc} is lesser in case of kesterite solar cell. Similarly, fill factor (FF) of the CZTS/CZTSe solar cell is higher than III-nitride based solar cell. It is due to higher J_{sc} value for the kesterite material. However, efficiency of the CZTS/CZTSe QW solar cell is quite low as compared to GaN/InGaN solar cell, which is due to high recombination rate and low EQE. Hence, it can be concluded that QW based GaN/InGaN solar cell is better as compared to CZTS/CZTSe QW solar cell. Additionally, taking bulk CZTS and CZTSe solar cell with GaN/InGaN QW solar cell in to consideration, it can be observed that both J_{sc} and FF are high in case of bulk material. Hence efficiency is high in case of CZTSe solar cell as compared to GaN/InGaN QW solar cell. Comparing, CZTS and CZTSe bulk solar cell, it is concluded that due to high absorption coefficient and low defect density, efficiency of bulk CZTSe solar cell is optimum among all. Additionally, CZTSe bulk solar cell is also more cost effective compared to III-nitride materials. Use of nanostructure in kesterite materials don't improve much performance, whereas as nanostructure improves efficiency of III-nitride materials. It can be anticipated that due to presence of polarization charges and control over growth plane nanostructure play an important role in GaN/InGaN QW solar cell, which is not possible in case of kesterite QWs.

TABLE 1. Performance measuring parameters of all types GaN/InGaN, CZTS/CZTSe, CZTS, and CZTSe bulk solar cell.

Parameters	GaN/InGaN QWs						CZTS/CZTSe QWs	CZTS Bulk	CZTSe Bulk
	{0001} Plane			{000-1} Plane					
	10%	20%	30%	10%	20%	30%			
J_{sc} (mA/cm^2)	1.36	1.31	1.25	1.29	1.14	0.8	21.9	29.38	54.58
V_{oc} (V)	1.013	0.93	0.89	0.97	0.83	0.76	0.75	1.06	0.66
FF (%)	71.1	66	52.3	67	59	51	85.1	88.75	84.3
Efficiency (%)	9.2	8.4	7.1	8.13	7.6	4.2	15.18	27.7	30.5
EQE (%)	89	88	79	62	60	56	80	80	90

IV. CONCLUSION

Use of nanostructure in GaN/InGaN QW solar cell reveal that detrimental effect of bulk III-nitride material can be overcome by using proper design of thin film layers. However, incorporation of nanostructure in kesterite material improves the efficiency as compared to bulk one. Effect of PCs is more pronounced in planar solar cell instead of nano enabled structures. This physical advantages brings a new aspect of nanotechnology into solar cell. An interesting power conversion efficiency of 9.2% with 71.1% FF is observed from only 4 QWs of GaN/InGaN solar cell. Further improvement in QW numbers and optimization may lead to more efficiency and performance. CZTSe bulk material shows a remarkable efficiency of 30% with 84% FF from 2 micron absorber layer thickness. A comparative analysis with all other possibilities are also discussed in detail. Hence, it is concluded that nanostructure boosts the efficiency to certain kind of materials, which can properly use the advantages of nonolayers such as CZTS/CZTSe. These findings may provide clear insight on potential of III-nitride and kesterite solar cell and can be further optimized for better improvement in efficiency.

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