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研究課題名（和文）異種材料接合ｓｉ／ＩｎＧａＮを用いた多バンド高効率太陽電池の創製

研究課題名（英文）Fabrication of Si/InGaN hybrid solar cells

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研究成果の概要（和文）：本研究では新規デバイスコンセプトとして、p型のpoly-Siやa-Si:H薄膜をn型の高In組成InGaN薄膜上に成長させることを提案する。InGaN薄膜上にp型のa-Si:Hか多結晶Si(プラズマCVD)を堆積させる、InGaN層のp型化の難しさを克服するとともに、p型Si系材料との接合によって長波長側への感度が向上すること。長波長側への感度が向上のためにInGaN活性層に多層量子ドット構造を利用した中間バンドの形成をする。

研究成果の概要（英文）： P-Si/InGaN heterojunction structures are proposed to fabricate solar cells to achieve both the current match and overcome the difficulty of p-type doping in In-rich InGaN. This structure can extend the optical absorption to the full solar spectrum by adjusting the In composition in InGaN and/or doping in the p-Si. We have successfully deposit amorphous Si on p-type InGaN by PECVD deposition to fabricate heterojunctions. The heterojunction indicated very good characteristics, showing the p-n junction behavior. The photovoltaic properties are achieved. InGaN quantum dots structures are also successfully achieved to realize long-wavelength absorption.

研究分野：工学

科研費の分科・細目：応用物理学・工学基礎　 薄膜・表面界面物性

キーワード：薄膜

１．研究開始当初の背景

The multi-junction photovoltaic (PV) cells based on the direct-bandgap III-Nitride material $In_xGa_{1-x}N$ is the most potential candidate for the third-generation photovoltaic application to overcome the efficiency limit of the conventional Si solar cells. This is because the bandgaps of $In_xGa_{1-x}N$ exhibit perfect match to the solar spectrum by adjusting the In composition. From the balance modeling estimation, the conversion efficiency of an $In_xGa_{1-x}N$-based four-junction solar cell was expected to be more than 50%. However, the reported maximum conversion efficiency (3%) in experiments for the $In_xGa_{1-x}N$ single-junction is far behind its theoretical one (27%). Up to now, most of the research is focused on improving In composition in the active region to fabricate multi-junctions to achieve higher conversion efficiency. However, *p*-type doping in In-rich $In_xGa_{1-x}N$ is considered to be a worldwide puzzle due to the physical restriction of the surface electron accumulation. Therefore, it is necessary to develop ***novel device concepts*** to overcome the drawbacks in the $In_xGa_{1-x}N$ material system.

２．研究の目的

In the proposed device concepts, ***p-type polycrystalline Si or a-Si: H thin film*** is deposited on the high-quality *n*-type $In_xGa_{1-x}N$ film, which provides the current match and overcomes the difficulty in achieving *p*-type doping in In-rich $In_xGa_{1-x}N$. This structure extends the optical absorption to the full solar spectrum by adjusting the In composition in $In_xGa_{1-x}N$ and/or doping in the *p*-type Si, which enhances the conversion efficiency markedly. ***Intermediate-band (IB) structures*** using $In_xGa_{1-x}N$ multi-layer quantum dots (QDs) as the sub-bandgap absorption region are proposed as the active regions to further improve the overall efficiency.

３．研究の方法

(1) The high-quality $In_xGa_{1-x}N$ thin films are deposited by using metal-organic chemical vapor deposition (MOCVD) system. The growth parameters such as growth pressure, temperature, V/III ratio, and flow rate are optimized to improve the crystalline quality.

(2) P-type amorphous Si was deposited by using both sputtering and plasma enhanced chemical vapor deposition (PECVD) method.

(3) The devices are fabricated using the standard semiconductor device processing, including laser lithography, dry etching, E-gun deposition, and annealing.

４．研究成果

1) High-quality InGaN thick film using AlN template (the lowest (002)-plane FWHM) and microstructure investigation

InGaN film is usually deposited on GaN template. As a result of lattice and thermal mismatch between GaN and InN, the critical thickness is just tens of nanometers. Therefore, high-density mismatch dislocations are usually generated if the thickness of InGaN is higher than their critical value. However, the absorption coefficient of InGaN is in the order of 105/cm. For a solar cell to absorb more than 90% solar light, the thickness of more than 250 nm InGaN is necessary. From metal-organic chemical deposition (MOCVD) growth technique, InGaN film with a thickness of 300 nm was deposited on GaN template. By optimizing the growth condition, the full-width at half maximum (FWHM) of X-ray diffraction (XRD) rocking curves around (002)-plane is 317 arcsec, and that of (101)-plane is 570 arcsec, which are the typical values for InGaN film with a In content of 11%. To further decrease the FWHM values and improve the crystalline quality, AlN template was proposed to be used as a template. Under the same growth conditions, InGaN thick film with the thickness of 300 nm is deposited. The FWHM of XRD (002)-plane was greatly reduced to 99 arcsec, which is the lowest value ever reported for InGaN thick film without using GaN substrate. The FWHM of (101)-plane is also decreased to 379 arcsec. The high-quality InGaN film will be helpful to further improve the performance of InGaN-based devices.

The microstructure of the film was investigated by transmission electron microscopy (TEM) to further investigate the nature of the threading dislocations, as shown in Fig.1. From the figures, even if AlN layer is with high-density dislocations, most of them were stopped by the sharp interface between AlN and GaN. Some of the dislocations formed loops and lead to annihilation due to their image forces. By titling the sample with different g vectors, screw-type, edge-type and mixed type dislocations were distinguished. As can be seen, the main dislocation type inside the GaN layer is mixed-type, which includes both screw- and edge-type component.

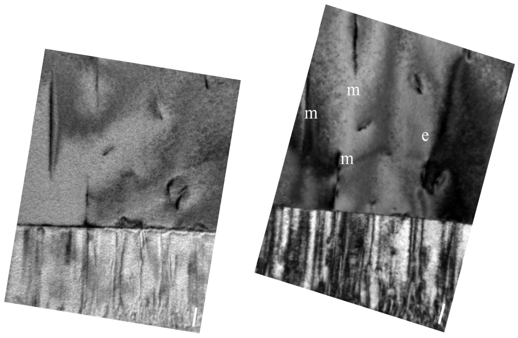

Fig. 1 TEM image with different g vectors of (a) [0002] and (b) [1-100]

3) Si/InGaN heterojunction solar cell fabrication

Although III-Nitride material InxGa1-xN is expected to be the most promising candidate for the next-generation photovoltaic applications, the inferior crystalline quality in the active region, difficulty in achieving p-type in In-rich InxGa1-xN, and poor understanding of the transport across the heterojunctions hinder the development in high conversion efficiency solar cells. On the other hand, the Si-based solar cells with crystalline, polycrystalline, or amorphous have been well developed. We propose to deposit p-type polycrystalline Si or a-Si thin film on the high-quality n-type InGaN film to develop heterojunction solar cells. This novel device concept can avoid the puzzle of p-type doping in In-rich InGaN, and extend the optical absorption by adjusting the In composition in InGaN and/or doping in the p-type Si. P-type amorphous Si was deposited by using both sputtering and plasma enhanced chemical vapor deposition (PECVD) method. We have successfully achieved the rectifying characteristics of the p-Si/n-InGaN heterojunctions, as shown in Figure. 2.

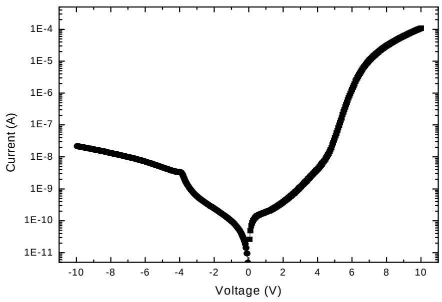

Figure 2 I-V characteristics of p-Si/n-InGaN heterojunctions

4) Introduce InGaN quantum dots to fabricate intermediate-band solar cells

The intermediate sub-bands from InGaN quantum dots (QDs) are introduced in the InGaN p-n junction as the active region. To form multi-levels, three different kinds of quantum dots with different In composition and size are introduced. The formation of the mini-bands is further confimed from both experiment and theoretical estimation. This is the first time for achieving intermediate-band transitions in the III-Nitride field.

Figure 3 illustrates the experimental demonstration of intermediate sub-bands including cathodoluminescence (CL), external quantum efficiency (EQE), and two-photon excitation measurement (TPE). Figure 3a displays the CL spectra of the sample at the electron beam energy of 7 kV at RT. A reference solar cell (RS cell) using InGaN thin film as the active region is also illustrated. Different from the single emission for the RS cell, several emissions are present in the CL spectra. In addition of the luminescence from GaN (363 nm), an emission located at 408 nm from the $In_{0.1}Ga_{0.9}N$/GaN pre-strained layers is observed, whose In composition (9.96%) is the same as the experimental design. The strongest peak at 465 nm (2.67 eV) is originated from InGaN/GaN host material, which is usually stronger and has a higher energy bandgap than those of QDs emissions. The luminescences located at the lower energies (517, 541, 630 nm) are attributed to the emissions from the triple QDs with different In compositions. Shown in Fig. 3d are the normalized CL spectra of MIB sample at electron beam energies ranging from 2 to 9 kV. As can be seen, at the lowest accelerating voltage of 2 kV, the strongest emission appears at 630 nm, corresponding to a photon energy of 1.97 eV. It records the information on the top region of the SLs (QDs 3). When the accelerating voltage increases to 3 kV, the luminescences related with another two QDs from 541 and 517 nm start to appear, corresponding to the photon energies of 2.29 (QDs 2) and 2.40 eV (QDs 1), respectively. With further increasing the accelerating voltages, the emissions from the host materials become stronger and the luminescence from the pre-strained layers appears. Above 7 kV, the whole luminescence information of the sample is displayed with the dominant peak from the GaN.

The photocurrent spectrum from the MIBs exhibits the widest range up to now from UV to near infrared regions in the InGaN system. The EQE (number of electrons produced as photocurrent per incident photon for the different wavelength) of the MIB solar cell measured at RT from 800 to 210 nm is shown in Fig. 3b. As expected, besides the bandgap edge absorption from VB to CB at around 465 nm, low-energy absorptions with its onset from 478 nm approaching as long as 750 nm are observed for the MIB cell. These sub-bandgap absorptions are originated from the MIB transitions from the triple QDs, which is consistent with the CL spectra.

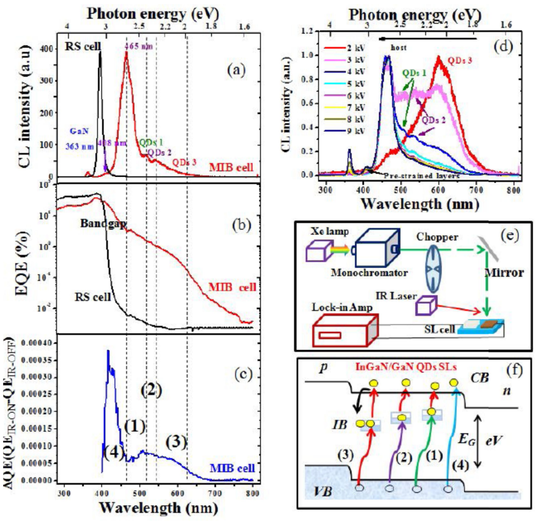

Figure 3.

５．主な発表論文等

（研究代表者、研究分担者及び連携研究者には下線）

〔雑誌論文〕（計 6 件）

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2. **<u>Liwen Sang</u>**, Meiyong Liao, Qifeng Liang, Masaki Takeguchi, Benjamin Dierre, Takashi Sekiguchi,Yasuo Koide, and Masatomo Sumiya, Multilevel intermediate-band solar cells based on III-Nitrides, 2013 **年秋季第 74 回応用物理学会学術講演会**, **同志社大学京田辺キャンパス**, 2013 年 9 月 16 日(月) ～ 20 日(金)
3. **<u>Liwen Sang</u>**, Meiyong Liao, Qifeng Liang, Masaki Takeguchi, Benjamin Dierre, Takashi Sekiguchi,Yasuo Koide, and Masatomo Sumiya, Multilevel intermediate-band solar cells based on III-Nitrides, received by 10th International Conference on Nitride Semiconductors as the ***Oral Presentation***, 2013 (Washiongton D.C. USA, 25th-30th, Aug, 2013)

〔図書〕（計 0 件）

〔産業財産権〕

○出願状況（計　　件）

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権利者：

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番号：

出願年月日：

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名称：

発明者：

権利者：

種類：

番号：

取得年月日：

国内外の別：

〔その他〕

ホームページ等

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