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Photocarrier generation in quantum-dot sensitized solar cells using Ge nanoparticle films

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Abstract

We report, for the first time, electrical power generation of Ge quantum-dot (QD) sensitized solar cells with rectification in a TiO_2/Ge/polysulfide electrolyte system. The crystalline Ge nanoparticle films are deposited by a high pressure radio-frequency sputtering method in an Ar/H_2 gas mixture. We have measured current-voltage characteristic and incident photon-to-current conversion efficiency (IPCE) of Ge QD sensitized solar cells. The photocurrent was detected by light irradiation in a wavelength range less than 800 nm, and IPCE is 40 % at 350 nm. This device shows short-circuit current of 0.65 mA/cm^2, open-circuit voltage of 0.15 V, and fill factor of 0.37.

1. Introduction

Third generation photovoltaics have attracted considerable attention, because their unique characteristics such as tunable band gap and multiple exciton generation (MEG) are expected to enhance the efficiency above the Shockley and Queisser limit of 33% [1]. MEG is able to produce more than two excitons by one incident photon in semiconductor nano-crystals and represents a promising route to increase solar conversion efficiency in single-junction photovoltaic cells [2,3].

QD sensitized solar cells, which are assembled by replacing dye molecules with semiconductor nanoparticles, have been developed over the past two decades as low-cost solar cells [4,5]. For this type of solar cells, small band-gap semiconductors such as CdS, PbS, CdSe, InP, and InAs serve as sensitizers because electrons photogenerated within them can be transferred to large band-gap semiconductors such as TiO_2 under light excitation. Recently, Sambur, *et al*. clearly demonstrated the MEG effect in PbS QD sensitized solar cells, where they observed over 100 % internal quantum yield [6].

Our interest has been motivated by QD sensitized solar cells using Si and Ge nanoparticles [7-12], because the technologically important and abundant Si and Ge poses no environmental problems regarding toxicity, and is the backbone of the current electronics and solar cell industries. In this study, we fabricated the Ge QD sensitized solar, and succeed in carrier generation in Ge nanoparticle films.

2. Experimental Results

The schematic geometry of the Ge QD sensitized cell is depicted in Fig. 1. The Ge QD sensitized solar cells form a photoelectrochemical system with an electrolyte containing a redox mediator. In the solar cell device, photogenerated electrons are expected to be transferred from excited Ge nanoparticles into the lower-lying conduction band of the TiO_2 films. On the other hand, holes in the Ge nanoparticles are neutralized by electrons injected from the sulfide ions S^{2-} of the redox mediator.

First, TiO_2 paste was coated on an FTO electrode by the doctor-blade method and baked at 450°C in air. The size of TiO_2 nanoparticles was about 20 nm, and the thickness of the TiO_2 films was 7 μm. Then, Ge nanopaticles films were fabricated using 13.56 MHz radio-frequency magnetron sputtering process in an Ar and H_2 gas mixture under a high pressure condition of 1.5 Torr. The sputtering target was a poly-crystal Ge disk (1 inch) with a purity of 99.99 %. Substrate temperature (T_s) was room one and 180°C. The film thickness was 230 nm. As seen in Fig. 2, X-ray diffraction spectra of Ge nanoparticle films show crystalline structure, where the diffraction peaks appear at 2θ = 27°, 45°, and 53° corresponding to the (111), (220), and (311) crystal planes of Ge, respectively. We also investigated the optical property of the nanocrystalline Ge films. The absorption coefficient steeply increases below 800 nm, and is a high value of 10^5 cm^{-1} in the visible wavelength region as shown in Fig. 3.

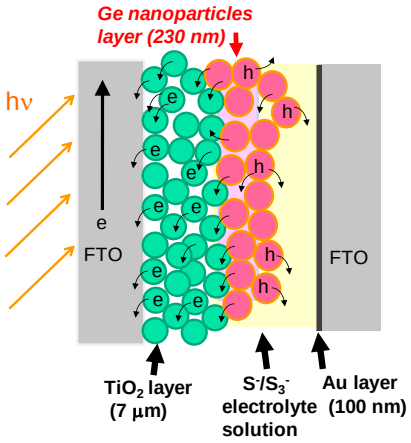

Fig. 1 Schematic of Ge quantum-dot sensitized solar cells.

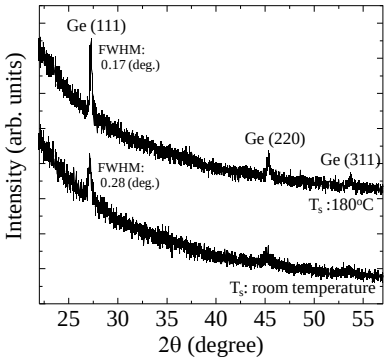

Fig. 2 Absorbance coefficient of Ge nanoparticle films deposited at substrate temperature of room temperature and 180°C.

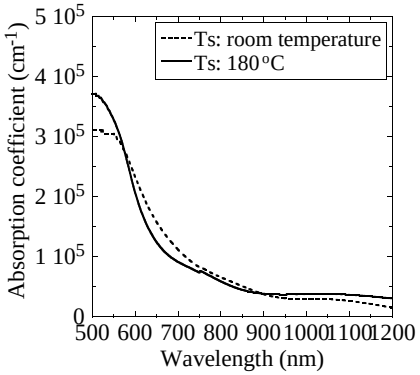

Fig. 3 Absorption coefficient of crystalline Ge nanoparticle films deposited at substrate temperature of room temperature and 180°C.

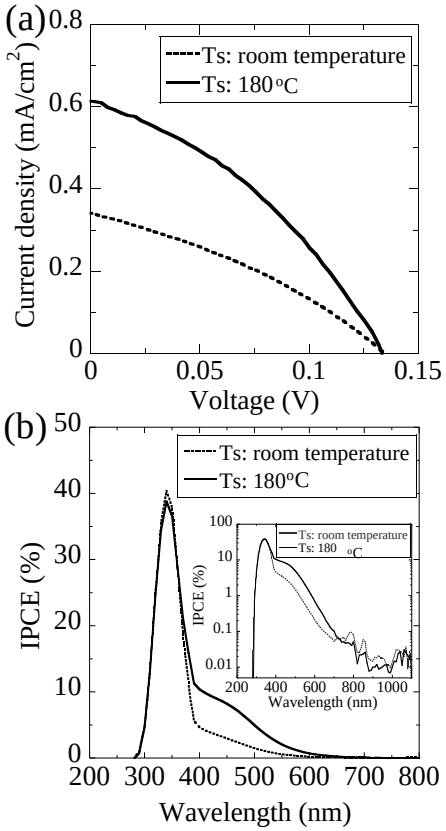

Fig. 4 (a) Current density-voltage characteristics of the Ge quantum-dots/TiO_2 solar cells. (b) Incident photon-to-current conversion efficiency spectra of Ge quantum-dots sensitized solar cells.

Finally, the Ge QD sensitized TiO_2 electrode and Au-coated FTO counterelectrode were sandwiched using 25-μm-thick spacers, and a polysulfide electrolyte solution was filled between these electrodes. The active area of the cells was 0.25 cm^2.

Figure 4(a) shows the current density-voltage (J-V) characteristics of the Ge QDs cells under AM 1.5, 100 mW/cm^2 (1 SUN) light irradiation with a solar simulator. The device, which consists of Ge nanoparticle films deposited at T_s = 180°C, shows short-circuit current of 0.65 mA/cm^2, open-circuit voltage of 0.15 V, and fill factor of 0.37. Figure 4(b) represents the incident photon-to-current conversion efficiency (IPCE) in the Ge QD sensitized solar cells. The photocurrent was detected by light irradiation in a wavelength range less than 800 nm, which is well explained by light absorption characteristics of nanocrystalline Ge films in Fig. 3. IPCE increases with decreasing wavelength, and is 40 % at 350 nm. The results indicate that excitons generated in crystalline Ge nanoparticles are separated into electrons and holes and such carriers are successfully extracted to the outer circuit. It is worth noting that this is the first report regarding electrical power generation of Ge quantum dot-sensitized solar cells to our best knowledge.

3. Conclusions

We successfully developed Ge QD sensitized solar cells using crystalline Ge nanoparticle films, which show high absorption coefficient of 10^5 cm^{-1} in visible wavelength region. IPCE of Ge QD sensitized solar cell steeply increased below 800 nm, and achieved a high value of 40 % at 350 nm.

Acknowledgements

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