X-ray, AFM, UV–VIS–IR analysis of a-Si:H/µc-Si:H supperlattice structure

ANDRZEJ KOŁODZIEJ^{*}, WITOLD BARANOWSKI, EDWARD KUSIOR, JAROSŁAW KANAK

Faculty of Electrical Engineering, Automatics, Computer Science and Electronics, AGH University of Science and Technology, al. Mickiewicza 30, 30-059 Kraków, Poland

*Corresponding author: kolodzie@agh.edu.pl

The use of the cyclic method of the deposition of multilayer amorphous and microcrystalline silicon films, based on the knowledge of a phase transient algorithm for silicon in low temperature conditions (below 250 °C), can give a possibility of creating silicon quantum dot structures. The thickness, crystallographic structure, optical gap as well as film and interface roughnesses of the amorphous Si:H and μ c-Si:H dot layer on glass and multicrystalline substrate were systematically studied by atomic force microscopy (AFM), small angle X-ray and UV–VIS–IR technique. It was developed on the base of these measurements that there is the phase transition from amorphous a-Si:H to multinanocrystalline Si structure with 4–15 nm crystallites.

Keywords: X-ray, atomic force microscopy (AFM), UV-VIS-IR analysis, nc-Si:H multilayer structure.

1. Introduction

At present there are many researches on the application of nanotechnology in solar cells [1-3]. This study is a continuation of searching for optimal technological recipes in order to fabricate silicon standard and quantum dot thin film tandem and triple structures [3-5]. A special attention was paid to manufacturing of the layers in the cycle technique on glass and foil to make better use of phase transient algorithms for silicon, germanium and alloys in low temperature conditions (150–250 °C).

2. Experiment details

The main aim was to obtain silicon quantum dots structures to higher absorption of the material and further use of the film in solar cell applications. A special programmable option was prepared to get gases into the apparatus according to a remote profile (multistep dependence of H_2/SiH_4 flow versus time and other parameters) during manufacturing of the layers with projected thickness and structures in the multilayer



Fig. 1. Fabrication of Si quantum dots solar cell concept by the RF PECVD multistep technique.



Fig. 2. Schematic diagram of energy band of the structure shown in Fig. 1. In the case of Si dots lower than 10 nm the energy gap of nc-Si is increasing (size effect).

system. The detailed description of the technique is in the paper presented at the 25th EU PVSEC in Valence [6]. The idea of the presented experiments was a sample fabrication, shown schematically in Fig. 1, with a periodic energetic scheme shown in Fig. 2.

Different algorithms of these periodic changes decide whether the layer is amorphous or nanocrystalline. Additionally, the period of the switching decides about the size of crystals. Many systematical and methodical experiments on fabricated silicon layers using a multistep (MS) approach were conducted to obtain repeatable properties of the structures. It was found that in this system, the amorphous and nanocrystalline silicon layers with better homogeneity can be obtained, avoiding the typical growth of columnar silicon crystals through the whole layer.

Exemplary X-ray, AFM and UV–VIS–IR experiments for layers fabricated in the cyclic change technique for the period a_1 and a_2 are shown in Figs. 3–8, which can



Fig. 3. Exemplary phase AFM pictures of the TCO substrate (corresponding image gives rms 1.32 nm).

Fig. 4. Exemplary phase AFM pictures of multilayer structure 120 double a_1 cycle by MS technique a-Si:H layer. A protocrystalline structure (corresponding image gives rms 2.47 nm) can be seen.



Fig. 5. Exemplary phase AFM pictures of multilayer structure 240 double a_2 cycle by MS technique nc-Si:H layer. A stress form substrate caused by big complexes can be seen. I Inside them, small Si crystallites can be indicated on 8–12 nm (corresponding image gives rms 3.69 nm).



Fig. 6. X-ray reflectivity curve and theoretical fit for the film a_2 made during 240 cycles shown in Fig. 5, with the assumption that the substrate TCO is 150 nm and the period for Si film is 6 nm.

indicate the size of crystallites. The set of the AFM pictures shows how the TCO surface influences particularly the quantum dots structure.

3. Results and discussion

The evidence of nanocrystallite is very difficult to obtain. Most methods are indirect. Among others, the lowering of hydrogen content in the film indicates the crystallization



Fig. 7. Small angle X diffraction a, a_1 and a_2 diagrams are shown for the films in Figs. 3, 4 and 5, respectively. The comparison gives information that only the layer in Fig. 6 grows like nanocrystalline.



Fig. 8. Exemplary shown reflection for multilayered silicon a-Si:H/ μ c-Si:H films deposited with various periodicity on multicrystalline wafer. The cycels a_3 and a_4 are too short.

process. Apparently, in first example $(a_1 - \text{Fig. 4})$ the films seem to be amorphous and in the second $(a_2 - \text{Fig. 5})$ – nanocrystalline.

The aim of this research was not only to confirm that the multistep (MS) method can be used to make homogenous protocrystalline films or homogenous nanocrystalline films with the required size of crystallites but also to fabricate solar cells with these layers. Those probes were shown in the paper [6, 7]. The exemplary data in Figs. 3–8 allow for the approximation of the crystallite size for "case a_2 " of about 8 nm.

The dot layers characterize lower resistivity in relation to standard microcrystalline films. The carried out optical studies allow not only for approximation of energy gap of the dot multilayer structure, but also for prediction of optimal thickness of the structure with respect to light absorption and confinement.

A similar analysis was conducted for a cycle a_6 sample. Four nanometre size nanocrystallites where indicated. Finally, the authors estimated the energy gap E_g for a 8 nm sample and 4 nm sample on 1.15 eV and 1.42 eV. In the second case, it is a thin size effect.

4. Summary

The application of MS technology guarantees, among others, better structural homogeneity of crystallized films and leads to multiexcyton absorption process, if the crystallites reach 5 nm size. It increases the final efficiency of fabricated cells. There are many factors which have an impact on manufacturing the nanolayers that need fast switching, for instance flow controller resolution, RF power supply with its matching time, speed of stabilization of process parameters in the technological chamber and many others. The idea of the developed method of deposition may be noticed in Fig. 2. The studies indicate that it opens a new way to manufacturing high efficiency quantum dot solar cell structures with the efficiency greater than 20%.

Acknowledgments - The work was based on the contract AGH GUZ 21.10.120.962.

References

- GREEN M., WENHA S.R., 2009 Annual Report, ARC Photovoltaics Centre of Excellence at the University of New South Wales, 2009.
- [2] SEUNG Y. MYONG, Recent Progress in Inorganic Solar Cells Using Quantum Structures, Recent Patents on Nanotechnology 1(1), 2007, pp. 67–73.
- [3] WRONSKI C.R., VON ROEDERN B., KOHODZIEJ A., *Thin-film Si:H-based solar cells*, Vacuum 82(10), 2008, pp. 1145–1150.
- [4] KOŁODZIEJ A., KREWNIAK P., BARANOWSKI W., KOŁODZIEJ T., Fabrication, characterization and modeling of thin silicon tandem cell on foil in four terminal configuration, Conference Proceedings of 34th IEEE Photovoltaic Specialists Conference (PVSC), Philadelphia, PA, June 7–12, 2009, p. 001742.

- [5] KOLODZIEJ A., Selected quantum solutions in the thin film silicon solar cell, Elektronika: konstrukcje, technologie, zastosowania (Warszawa) 51(5), 2010, pp. 87–91 (in Polish).
- [6] KOŁODZIEJ A., KREWNIAK P., BARANOWSKI W., KOŁODZIEJ T., Deposition, Characterization and Modeling of Thin Silicon Tandem Cell on Glass and Foil, Conference Proceedings of 25th European Photovoltaic Solar Energy Conference and Exhibition/5th World Conference on Photovoltaic Energy Conversion, September 6–10, 2010, Valencia, Spain, 2010, pp. 3056–3059.
- [7] FURUKAWA S., MIYASATO T., *Quantum size effects on the optical band gap of microcrystalline Si:H*, Physical Review B **38**(8), 1988, pp. 5726–5729.

Received September 25, 2010