Characterization of nc-Si / SiOx based quantum superstructures for the solar cell application by aberration-corrected high resolution electron microscopy

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Si-based nano-structures are of a large interest of the photovoltaic community due to an appealing combination of attributes and large sensitivity of the band gap energy on the dimensions of structural modulations. An enhanced photovoltaic performance of the quantum superstructures can be achieved by designing a so-called tandem solar cell, where the cell consists of nano-modulated parts to absorb different wavelength regions of visible light and UV- light [1]. Ideally, in the Si-based quantum superstructures composed from arrays of alternating conductive and isolating layers, the quantum size effect deduce strong photo-luminescence (PL) of the nano-crystalline Si (nc-Si) in the visible spectral range and leads to blue shift of the luminescence by decreasing nanocrystal size (the quantum confinement effect) [2]. That means electro-optical performance of a nano-structured functional element crucially depends on its nano-structural design. Therefore control of size, passivation, and density is mandatory [3]. As a consequence, the development of new quantum superstructures for high performance solar cells requires the thorough structural understanding by characterization of nano- and sub-nano-scale structure [4]. The size control is doable by changing the chemical stoichiometry of the films and initial film thickness and it follows by heat treatment procedure. Annealing of a deposited multilayer elevate the precipitation of silicon, diffusion of the Si and O atoms and the nucleation of spherical shaped Si nano-crystals, embedded within the SiO_2 matrix [5]. Reduction of the implanted Si dose or the O enrichment is the usual ways for a decrease in nano-crystal size. By decreasing the nano-crystal size into the desired range the density of the nanocrystals is reduced simultaneously. However for thicker samples, most of the nano-crystals touch each other and form a continuous polycrystalline layer. This clearly limits the luminescence efficiency in the visible range due to non-radiative processes at the grain boundaries.

In the frame of this work, we prepared two nano-structured Si-based quantum systems, a-Si/SiO_x and SiO_x/SiO_2 multilayer systems. First series, consisted from periodically stacked alternating amorphous Si and sub-stoichiometric amorphous $SiO_{x(x<2)}$ layers with different thickness. The second, an as-deposited $SiO_{x(x<2)}$ /SiO_2 series was composed from periodical arrangement of alternating amorphous under-stoichiometric $SiO_{x(x<2)}$ and stoichiometric SiO_2 layers. After annealing the sample at 1100 K, Si crystalline nano-crystals (nc) formed with in SiO_2 amorphous matrix. The morphology and distribution of nc-Si precipitates within a amorphous layer, their nucleation and growth kinetics, the thickness of conducting layer and diffusion of O or Si through interfaces, they all affect the optoelectronic performance of the system [4] were targeted in this study. Annealed samples were analyzed by high resolution transmission electron microscopy (HRTEM), energy filtered transmission electron microscopy (EFTEM) and electron energy loss spectroscopy (EELS). We employed aberration-corrected TEM microscopes to shift the boundary of analyzing scale tremendously. PICO, the newly installed FEI electron microscope in research center Juelich (ER-C), which is equipped with both image and probe correctors, used to reveal excellently the chemical distribution of Si element in atomic scale and crystalline structure at one single measurement (Fig. 1). Also it has been observed that the mean size of the QDs and their distribution in the dielectric matrix host, change by the thickness of the SiO_x layer and the thermal annealing process (Fig. 2). After high temperature annealing of amorphous superlattice the size of the nano-crystals is limited at least in one direction due to the Si layer nominal thickness. The kinetics of the formation of nano-

crystalline Si (nc-Si) precipitates in Si-reach layers sandwiched between amorphous barrier layers was studied as a function of stacking period and oxygen content in a system. We used EELS to clarify chemical differences between the interfaces near the Si substrate and interfaces near SiO_2 and qualitatively observed diffusion profile of Si Atoms. It has been shown that the nucleation rate of the crystalline phase increases with a decrease of the oxygen content in Si-rich layers and Oxygen also affects the precipitation morphology. Namely upon annealing, in the O-depleted superstructure, as-deposited a-Si/SiO_x multilayers, the nearly continuous Si-crystalline layer forms within the original a-Si layer.

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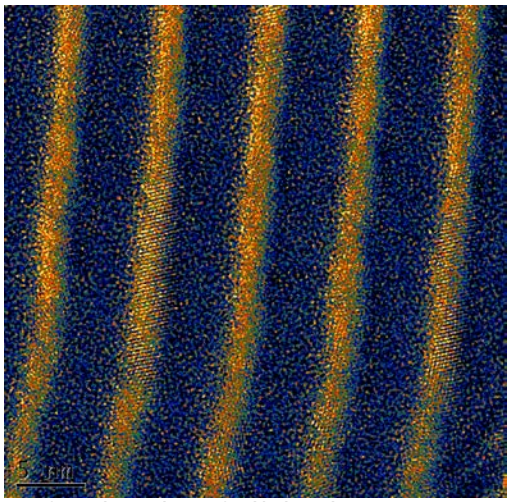

Figure 1. *High resolution EFTEM* map for Si L edge of Si/SiO_2 hetero-structure reveals clearly atomic crystalline structures beside the chemical distribution of Si atoms

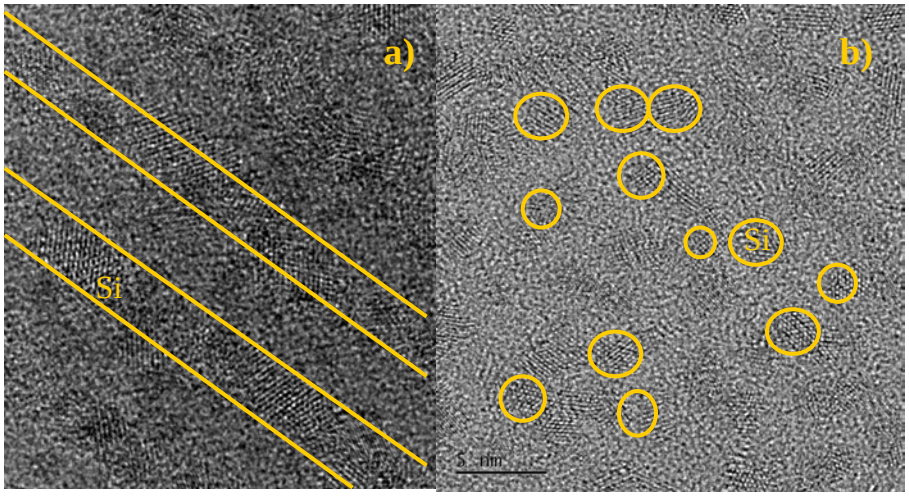

Figure 2. *HRTEM a) within a multilayers sample of 4nm SiO_x and 3nm of SiO_2 , continues polycrystalline layer of Si formed while for thinner sample, b) of 3nm SiO_x and 3nm of SiO_2 some discrete nc formed*