Photoelectrochemical tandem cells for highly efficient unassisted water splitting

T. Hannappel[1], O. Supplie[1,2], M.M. May[4], Wen-Hui Cheng[3], Matthias H. Richter[3], H.-J. Lewerenz[3], H. A. Atwater[3], F. Dimroth[5], J. Ohlmann[5], D. Lackner[5]

[1]*Ilmenau University of Technology, Institute of Physics, Dep. Photovoltaics, Germany*
[2]*Helmholtz-Zentrum Berlin für Materialien und Energie, Solar Fuels Institute, Germany*
[3]*California Institute of Technology, Joint Center for Artificial Photosynthesis, Pasadena, USA*
[4]*University of Cambridge, University of Cambridge, Dep. of Chemistry, United Kingdom*
[5]*Fraunhofer Institute for Solar Energy Systems, Freiburg, Germany*

Regarding the design of a water-splitting photoelectrochemical (PEC) cell and the given thermodynamic and kinetic energetic condition for water splitting, only absorbers with large band gaps or tandem structures with a sufficient voltage, i.e. with sufficient splitting of the quasi-Fermi level, can efficiently produce H2 [1]. As a result, the energy gap of a single junction PEC cell needs to be larger than 2 eV considering the chemical potentials of electrons and holes in the absorber layers, the anodic and cathodic redox potentials and over-potentials, which provide the driving force for the solid—liquid interfacial reactions. Hence, in a PEC cell, when utilizing single absorber device structures, the maximum solar-to-hydrogen (STH) efficiency is limited to lower values than 15% at one sun AM1.5. When using tandem cells much higher STH efficiencies can be achieved, potentially up to 25% [2,3]. On the example of a monolithic two-junction water-splitting PEC device that works close to the thermodynamically limiting efficiency and exhibits an initial STH efficiency of > 19 % in acid electrolytes and an efficiency of > 18 % [4] at neutral pH conditions (under simulated sunlight) I will discuss details on the requirements of highly efficient water splitting devices.

In order to reduce costs and to realize competitive tandem device structures without employing concentrator technology, the concept of III-V semiconductor integration into silicon technology refers to one of the most topical and generic issues. Low-defect III-V growth on Si relates to the step structure of the substrate: single layer steps on the substrate initiate anti-phase disorder in the III-V material, while a double-layer stepped substrate in principle enables anti-phase-free III-V growth. We control atomic scale formation of suitable Si(100) surfaces *in situ* under CVD preparation conditions, i.e. preferentially double-atomic stepped surfaces in a complex chemical environment. We directly observe the domain formation in dependence of the preparation route With in situ reflection anisotropy spectroscopy (RAS). Oscillations in transient RA measurements indicate layer-by-layer Si removal during annealing in hydrogen. We conclude that vacancy island formation and anisotropic expansion preferentially in parallel to the dimer rows of the terraces explains the layer-by-layer Si removal process. Afterwards the nucleation of an almost lattice-matched GaP film is initiated. From such a III-V quasi (or virtual) III-V substrate different routes can be envisaged. Fig. 1 juxtaposes RA spectra of differently terminated Si(100) surfaces directly prior GaP nucleation and RA spectra of P-rich GaP/Si(100) obtained after GaP growth on the corresponding Si(100) surface. It can clearly be seen that the sublattice orientation of the majority domain in the GaP layer is determined by the majority dimer orientation on the Si(100). Fig. 1 displays the preparation of the Arsenic-terminated Si(100):As 0.1° surface with evenly spaced double-layer steps and with an exiguous presence of minority domains reflected by a high intensity of peaks in the RA spectra [5-8].
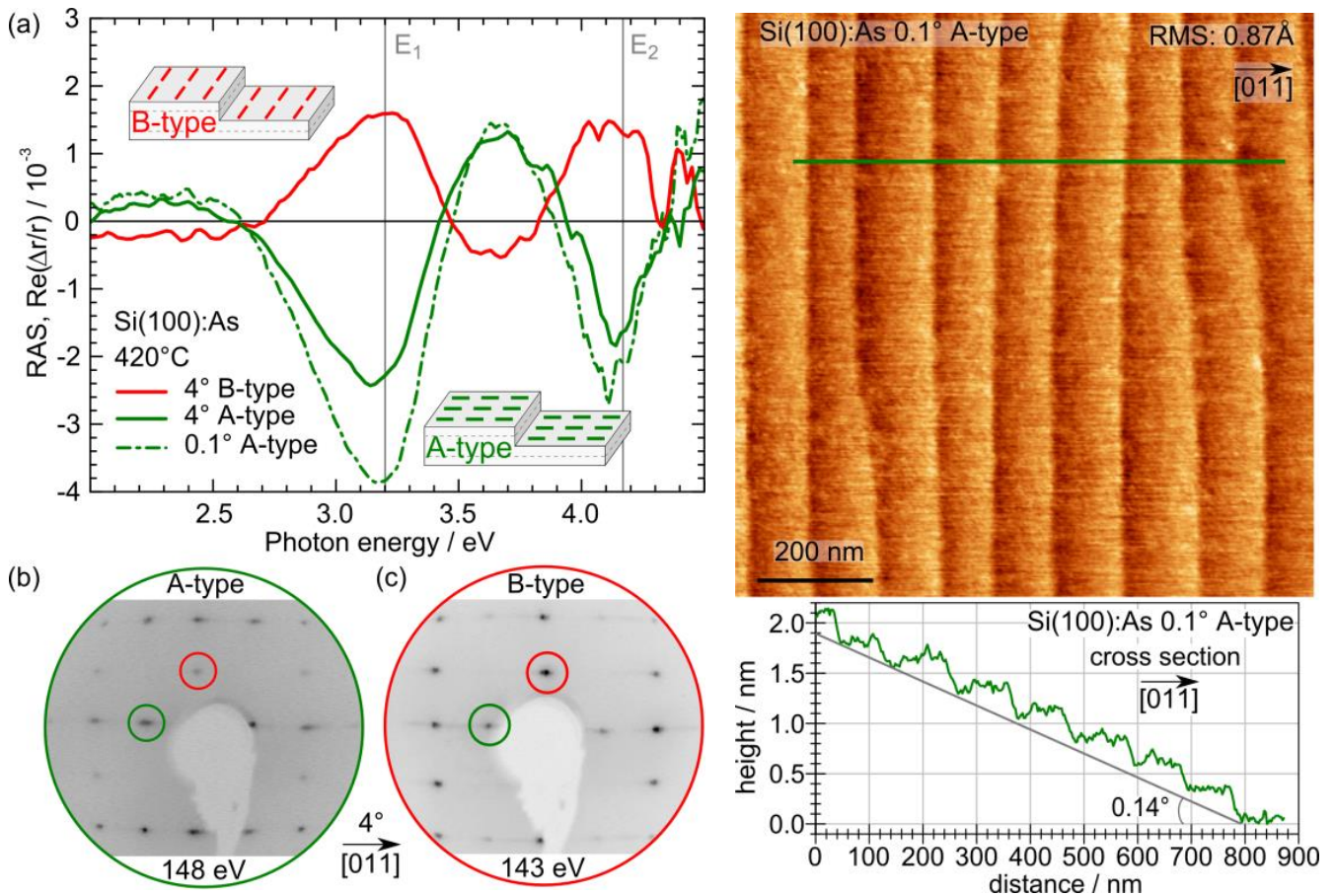
Fig. 1. (a) RAS of Si(100):As with 4° (dashed line) and 0.1° (solid line), change in the RAS sign corresponds to the predominant surface reconstruction on the Si surface (see insets); (b,c) LEED patterns of Si(100):As 4° A-type (b) and B-type (c). (d) AFM of Si(100):As 0.1° (A-type) 1 × 1 µm² image with a cross section (plane tilt corrected) of a line scan (green line).

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