Type of presentation: Poster

IT-6-P-3009 Multi-slice simulations for in-situ HRTEM studies of nanostructured magnesium hydride at elevated hydrogen pressures of 1 bar

Surrey A.[1,2], Schultz L.[1,2], Rellinghaus B.[1]

[1]IFW Dresden, Dresden, Germany, [2]TU Dresden, Institut für Festkörperphysik, Dresden, Germany

Email of the presenting author: b.rellinghaus@ifw-dresden.de

Nanostructuring of many hydrides has been shown to reveal improved thermodynamic and kinetic properties, which are needed for both mobile or stationary applications of solid-state hydrogen storage materials. During structural characterization utilizing conventional (HR)TEM, however, hydrides such as MgH_2 degrade fast upon the irradiation with the imaging electron beam due to radiolysis in vacuum and as a consequence, the hydride phase cannot be studied at highest resolution. This problem can be overcome using a novel nanoreactor recently developed by H. Zandbergen (TU Delft) that allows for in-situ TEM studies at elevated H_2 pressures (up to 4.5 bar) and temperatures (up to 500°C) [1]. A point resolution of 0.18 nm has already been demonstrated experimentally for Cu nanocrystals [2].

We have studied the feasibility of HRTEM investigations of light weight metals such as Mg and its hydride phases with the nanoreactor by means of multi-slice HRTEM contrast simulations. Such a setup provides the general opportunity to fundamentally study the dehydrogenation and hydrogenation reactions at the nanoscale under realistic working conditions. We analyze the dependence of both the spatial resolution and the HRTEM image contrast on parameters such as the defocus, the metal/hydride thickness, the hydrogen pressure and the nanoreactor geometry in order to explore the possibilities and limitations of in-situ experiments with this reactor. Such simulations may be highly valuable to pre-evaluate future experimental studies.

Fig. 1 shows schematically the details of the nanoreactor as it was implemented in a super cell used for the multi-slice simulations. The hydrogen is encapsulated between two 20 nm thin α-Si_3N_4 windows with the metal/hydryde positioned on top of the the bottom window. First simulations were conducted for a metallic Mg film of varying thickness oriented with its [001] direction parallel to the electron beam. The slicing was chosen to account for the varying density of atomic scatterers along the beam direction. While the slice thickness was reduced to contain only a single layer of scatterers within the Mg layer, it was increased to 1 nm and 100 nm in Si_3N_4 and in the hydrogen containing volume, respectively. Fig. 2 shows as an example the simulated Weber contrast of a Mg column (averaged over 611 individual columns) with respect to the background due to the Si_3N_4 windows as a function of the Mg thickness and the defocus. (Simulation conditions: Linear imaging. g_{max} = 20/nm. Imaging parameters match a FEI Titan microscope at 300 kV for NCSI imaging. Absorption, the MTF of the CCD, and a noise level of 3% were included in the simulations.)

[1] T. Yokosawa, Ultramicroscopy 112 (2012) 47.

[2] J.F. Creemer et al., Ultramicroscopy 108 (2008) 993.

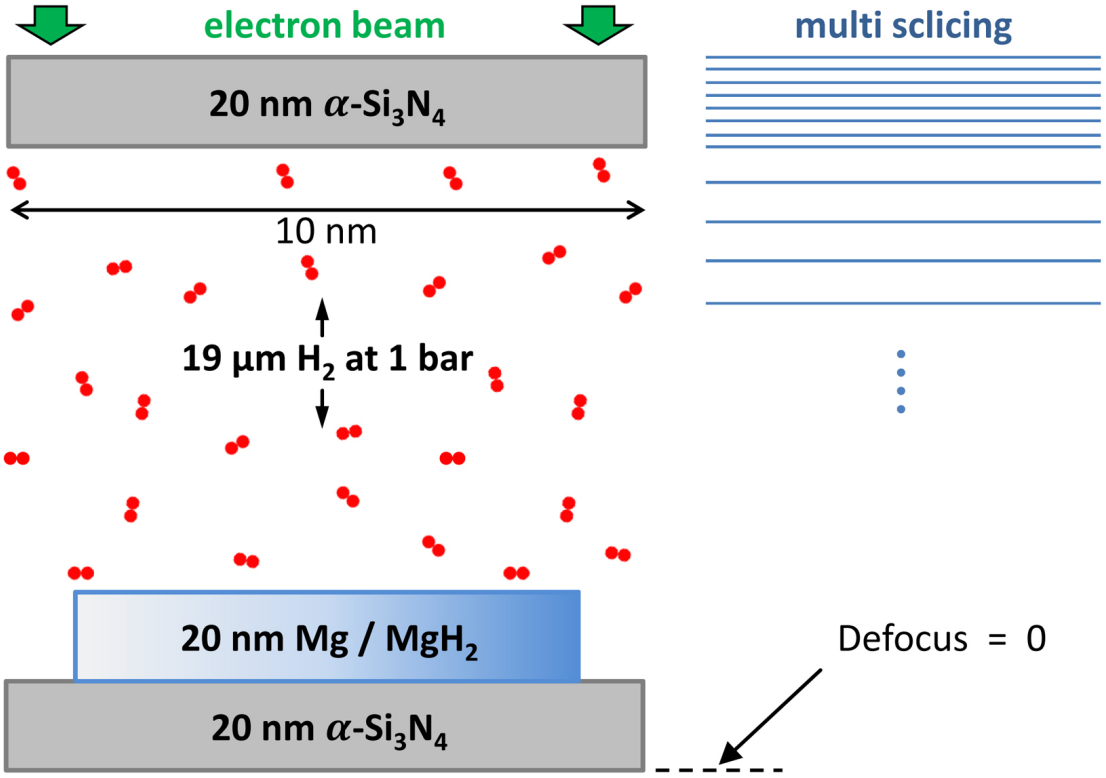

Fig. 1: Schematic illustration of the simulated super cell representing the nanoreactor used for in-situ HRTEM investigations of the (de)hydrogenation of Mg(H_2).

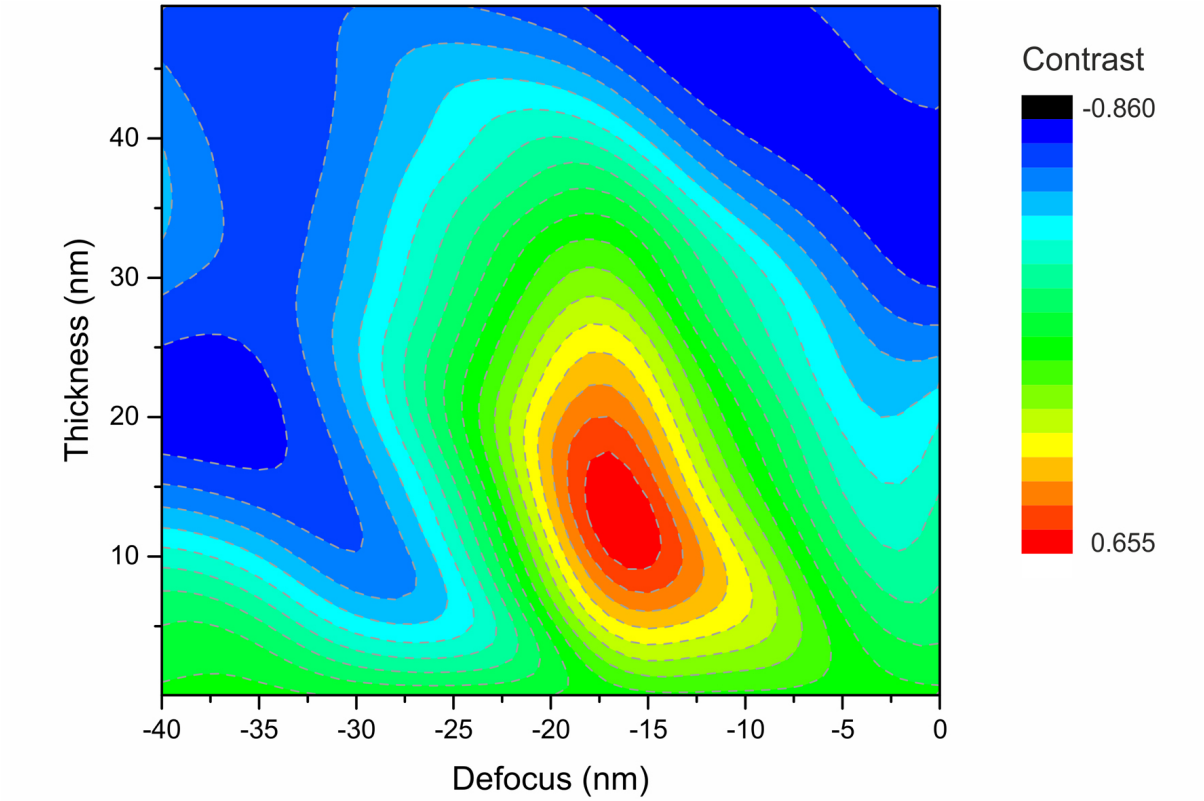

Fig. 2: Mean Weber-type image contrast of a column of Mg atoms as obtained from averaging over 611 individual atomic rows along the [001] direction of a Mg film with varying thickness. Despite the two Si_3N_4 windows and some scattering from the hydrogen atoms the individual Mg columns can be clearly imaged for thicknesses below some 30 nm.