Layered noble metal (Pd, Pt) chalcogenides with structural formulas ranging from MX_2 to MX (M: Pt or Pd, and X: Se or Te) have been proposed as active catalysts for the electrochemical hydrogen evolution reaction (HER), but the details of the catalytic action are far from being understood. This may be related in part to strong interlayer interactions and size quantization effects in these materials, which cause significant modifications of their electronic properties depending on the number of layers. At the same time, these physical effects may also open new avenues to the tailoring of catalytic properties in these materials. Moreover, controlled design of undercoordinated atoms at step edges with special electronic properties may also increase the abundance of highly active catalytic sites. In this project, we will identify the active compositional phases and how nano-structuring (number of layers and step edge density) of these noble metal chalcogenide materials may be used to boost the HER activity. We also intend to utilize the structural similarities of a wide range of transition metal dichalcogenides (TMDs) as a materials platform to investigate possible synergetic effects in TMD-phase mixtures (alloys) to enhance HER activity. We envision that the planar nature of these materials will aid the characterization of structural and electronic properties of mixed-phase materials and thus facilitate the fundamental understanding of synergetic effects in multi-component materials. To conduct these studies, we assembled a team with complementary expertise and capabilities. Planar model systems will be synthesized by van der Waals epitaxy, and their atomic structure and electronic properties will be characterized by scanning probe microscopy and photoemission spectroscopy in the Batzill's group at University of South Florida, USA. The electrochemical properties of these well-defined samples, so far poorly investigated in the electrochemistry community, will be analyzed at the TU Braunschweig in the Oezaslan's group. The experimentally determined micro-kinetics results will be rationalized through ab initio simulations to be done in Krasheninnikov's group at the Helmholtz Zentrum Dresden-Rossendorf. The theoretical predictions for alloys and dopants will also guide the experiments and help to identify promising materials combinations. This team encompasses detailed characterization of the materials so that the kinetic parameters and HER activities can be correlated with their physical and chemical properties. The outcome of this project is the identification of new potential electrocatalysts for HER and their tunability by controlled nano-structuring. These studies will not only provide fundamental knowledge on the catalytic action of advanced low-dimensional materials, but also define new pathways for the practical design of advanced electrocatalysts, and thus contribute to finding ecological energy solutions for the society.