HL 16: Functional Semiconductors for Renewable Energy Solutions I

Time: Tuesday 9:30–11:45 Location: ER 325

HL 16.1 Tue 9:30 ER 325

Metavalently bonded tellurides: the essence of improved thermoelectric performance in elemental Te — •Yuan Yu[1], Decheng An[2], and Matthias Wuttig[1] — [1]Institute of Physics (IA), RWTH Aachen University, Sommerfeldstraße 14, 52074 Aachen, Germany — [2]College of Chemistry, Taiyuan University of Technology, Taiyuan 030024, China

Doping is crucial to obtain high-efficiency elemental Te thermoelectrics. However, the remarkably low solubility of dopants such as As, Sb, and Bi in Te provides a major challenge. This also complicates the understanding of improved thermoelectric properties upon doping these elements. Here, we develop a correlative method to characterize the local microstructures and corresponding transport properties in doped Te crystals. We confirm that the conspicuous enhancement of bulk electrical conductivity and power factor stems from the dopant-induced formation of metavalently bonded precipitates, which form electrically beneficial interfaces with the Te matrix. A quantum-mechanical-derived chemical bonding map successfully uncovers efficient dopants for improving the thermoelectric performance of Te. Forming metavalently bonded tellurides, which facilitate charge transfer across the interface, becomes a key factor for property design. This nonclassical doping recipe is based on the newly developed understanding of the origins of thermoelectricity in this system and opens pathways for tailoring other complex semiconductors.

HL 16.2 Tue 9:45 ER 325

Rational Design of the CoS Co9S8@NC Composite Enabling High-Rate Sodium-Ion Storage — •Ying Qi — TU Ilmenau

Metal sulfides have been considered promising anode materials for sodium-ion batteries (SIBs) due to their high specific capacities. However, the poor electrical conductivity and sluggish electrochemical kinetics of metal sulfides are the critical factors that are limiting their applications. In this work, cobalt sulfides with heterostructures embedded in an N-doped carbon composite (CoS/Co9S8@NC) have been synthesized to further investigate Na+ diffusion in the SIBs. With contributions from the heterostructure, the N-doped carbon, and the unique morphology, the composite can deliver enhanced rate capability and cycling stability compared to Co9S8. This work depicts the change of Na+ diffusion under the influence of heterostructures, providing an effective strategy of material design for enhancing the electrochemical performance of sodium-ion storage.

HL 16.3 Tue 10:00 ER 325

Constructing metal sulfide anodes with excellent K-ion storage properties via microstructure engineering — •Kangzhe Cao[1,2], Huiqiao Liu[2], Jiahui Ma[2], and Yong Lei[1] — [1]Fachgebiet Angewandte Nanophysik, Institut für Physik & IMN MacroNano, Technische Universität Ilmenau, 98693 Ilmenau, Germany — [2]College of Chemistry and Chemical Engineering, Xinyang Normal University, Xinyang 464000, China

K-ion batteries (KIBs) feathered by abundant resources and high energy densities are considered a kind of ideal candidates for the large-scale energy storage systems. Anode materials, such as metal sulfides, with high capacity always hold more than one K ions per molecule, companying large volume expansion during cycling. This character endangers the stability of the electrode structure and the solid-electrolyte-interphase (SEI) on the active materials, resulting in low coulombic efficiency and limited cycle life. Herein, we would like to introduce a strategy to construct a series of high-performance metal sulfide anodes for KIBs via structure engineering, including conversion reaction anodes and conversion-alloy reaction anodes. We will demonstrate how the confining layer improves the coulombic efficiency of CuS anode, why the electrochemical reversibility of Sb2S3 anode is triggered by the synergistic effect of confining and catalysis, and how to improve the cycling stability of SnS2 through heterostructure designing. Our work confirms that the K-ion storage properties of metal sulfide anodes can be largely improved by microstructure engineering, offering a reliable strategy for designing high-performance KIB anodes.

HL 16.4 Tue 10:15 ER 325

Effects of Defects on the Optoelectronic Properties of Ta_3N_5 Thin Films — •Lukas M. Wolz[1,2], Gabriel Grötzner[1,2], Laura I. Wagner[1,2], Tim Rieth[1,2], Matthias Kuhl[1,2], Guanda Zhou[1,2], Verena Streibel[1,2], Saswati Santra[1,2], Ian D. Sharp[1,2], and Johanna Eichhorn[1,2] — [1]Walter Schottky Institute, Technische Universität München — [2]Physics Department, TUM School of Natural Sciences, Technische Universität München

Photoelectrochemical (PEC) energy conversion is a promising approach for efficient solar-to-fuel conversion. In this context, transition metal nitride semiconductors have recently emerged as an interesting class of materials for overcoming the limitations associated with commonly studied metal oxides. While the impact of oxygen impurities (O_N) and nitrogen vacancies (v_N) on the PEC activity has already been explored, the understanding of how these defects impact stability is still lacking. Here, we utilize a controllable synthesis approach to independently tune O_N and v_N concentrations in Ta_3N_5 and systematically investigate structural, compositional, optoelectronic, and PEC properties to establish the relationship between atomic-scale defects and macroscale PEC stability. Low oxygen concentrations facilitate the formation of deep v_N defects, leading to charge recombination and limiting the PEC stability. Increasing oxygen incorporation leads to the passivation of v_N defects, resulting in reduced defect absorption, improved charge separation, and enhanced material stability. Overall, these results show the detrimental role of v_N and the beneficial impact of O_N on the stability of Ta_3N_5.

15 min. break

HL 16.5 Tue 10:45 ER 325

Oxygen incorporation as a route to non-degenerate zinc nitride — •Elise Sirotti, Bianca Scaparra, Stefan Böhm, Felix Rauh, and Ian D. Sharp — Walter Schottky Institute and TUM School of Natural Sciences, Technical University of Munich

Zinc nitride (Zn_3N_2) comprises earth-abundant elements, possesses a small direct bandgap, and is characterized by high electron mobilities. These characteristics make it, in principle, a promising compound semiconductor for photovoltaic and thin-film transistor applications. However, in practice, the unintentional degenerate n-type doping that is commonly reported and often assigned to oxygen impurity doping significantly impedes its development for technological applications. To systematically investigate this doping effect, epitaxial Zn_3N_2 were grown by plasma-assisted molecular beam epitaxy with controlled oxygen content up to 20 at.% on a-plane sapphire, resulting in single phase (222)-oriented Zn_3N_2 with variable oxygen content. A combination of structural, electrical, and optical measurements reveals how, contrary to expectations, oxygen incorporation can lift the degenerate conductivity and achieve films with charge carrier concentrations in the $10^{17}\,cm^{-3}$ range. Indeed, both electrical and optical measurements confirm that the material changes from a degenerate metallic to semiconductor behavior as the oxygen content increases, suggesting the formation of charge-compensating defect complexes. The understanding of the beneficial role of oxygen thus provides a route to bring Zn_3N_2 into reach for technological applications.

HL 16.6 Tue 11:00 ER 325

Cobalt nitride thin films for stabilizing nitride-based photoelectrodes — •Matthias Kuhl[1,2], Lukas Kohlmaier[1,2], Ian D. Sharp[1,2], and Johanna Eichhorn[1,2] — [1]Walter Schottky Institute, Technische Universität München, Germany — [2]Physics Department, TUM School of Natural Sciences, Technische Universität München, Germany

For artificial photosynthesis, metal nitride semiconductors evolved over recent years as a promising material class, offering narrow bandgaps that are ideal for visible light absorption and pronounced bond covalency that facilitates long range charge transport. However, these materials suffer from poor stability under realistic operation conditions. One strategy to improve stability is to interface the semiconductor light absorber with conformal and ultra-thin catalytic layers that still permit interfacial charge transport and minimize losses due to parasitic light absorption. In contrast to typically studied oxide protection layers, we use plasma-enhanced atomic layer deposition (PE-ALD) to deposit ultra-thin cobalt nitride films as conformal, stable, and catalytically active coatings. Different characterization techniques are combined to reveal the effect of varying synthesis parameters on the material properties. Explicitly, we demonstrate that the deposition temperature can be employed to tune the film composition and nitrogen content, thereby controlling the optoelectronic properties and catalytic activity. Overall, this work highlights the use of PE-ALD as a promising approach for engineering pure nitride catalyst/semiconductor interfaces to create efficient and stable photoelectrodes.

HL 16.7 Tue 11:15 ER 325

Low temperature photoluminescence investigation of boron doped and quenched silicon — •Kevin Lauer[1,2], Robin Müller[1], Zia Ul-Islam[1], Katharina Peh[1], Dirk Schulze[1], and Stefan Krischok[1] — [1]Technische Universität Ilmenau, Institut für Physik, Ilmenau, Germany — [2]CiS Forschungsinstitut für Mikrosensorik GmbH, Erfurt, Germany

The linking of light-induced degradation (LID) with photoluminescence (PL) peaks has been shown to be very constructive in defect identification in case of indium doped silicon.[1] For boron doped silicon the search after PL peaks, which follow LID treatments, was not successful so far.[2] By applying quenching treatments[3] we were now able to discover a PL peak, which is impacted by LID treatments.[4] The relation of this discovered peak to closely neighbored known peaks is discussed.

[1] K. Lauer, K. Peh, D. Schulze, T. Ortlepp, E. Runge, and S. Krischok, Phys. Status Solidi A, vol. 219, no. 19, p. 2200099, 2022 [2] K. Peh, K. Lauer, A. Flötotto, D. Schulze, and S. Krischok, Phys. Status Solidi A, vol. 219, no. 17, p. 2200180, 2022 [3] M. L. W. Thewalt, U. O. Ziemelis, and R. R. Parsons, Phys. Rev. B, vol. 24, no. 6, p. 3655, 1981. [4] K. Lauer et al., arXiv, Nov. 13, 2023. doi: 10.48550/arXiv.2311.07280.

HL 16.8 Tue 11:30 ER 325

$CeTa(O,N)_3$ and $CeNb(O,N)_3$ perovskite oxynitrides for photoelectrochemical energy conversion — •Gabriel Grötzner[1,2], Laura I. Wagner[1,2], Lukas Wolz[1,2], Verena Streibel[1,2], and Ian D. Sharp[1,2] — [1]Walter Schottky Institute, Technische Universität München, Germany — [2]Physics Department, TUM School of Natural Sciences, Technische Universität München, Germany

Photoelectrochemical (PEC) water splitting presents a promising avenue for the efficient storage of solar energy in chemical bonds. Perovskite oxynitrides, emerging as a new class of materials for PEC water splitting, hold potential to overcome certain limitations associated with more extensively studied metal oxides, particularly as many perovskite oxynitrides have a band gap of ∼2 eV. However, the optoelectronic properties of many perovskite oxynitrides are not well understood and investigated. This study introduces a novel method for synthesizing thin films of the perovskite oxynitrides $CeTa(O,N)_3$ and $CeNb(O,N)_3$ through a spin coating process followed by a two-step annealing procedure in ambient air and ammonia. The investigation explores the structural, compositional and optoelectronic properties of the thin films and furthermore compares the results between $CeTa(O,N)_3$ and $CeNb(O,N)_3$. Additionally, the influence of the annealing conditions in ammonia on the material properties is evaluated. The experimental findings unveil bandgaps in the visible range and n-type conductivity for both materials. As these results suggest use cases for both materials in PEC watersplitting, their viability as photoanodes is also evaluated.