DS 6: Thin Film Properties I

Time: Tuesday 9:30–12:45 Location: A 060

Invited Talk DS 6.1 Tue 9:30 A 060

Concerted electron-nuclear motion in polaron formation and exciton transfer — •Wolf Gero Schmidt — Universität Paderborn

Ab initio molecular dynamics calculations on (excited-state) potential energy surfaces obtained from constrained density-functional theory [1] provide deep insight into the concerted electron-nuclear motion of excited systems and allow for the quantitative modelling of the excitation dynamics [2]. This is demonstrated in my talk using two intriguing examples: (i) The formation of bound polarons in lithium niobate occurs on the femtosecond timescale [3] and modifies significantly the linear and nonlinear optical response [4]. (ii) The transfer of triplet excitons resulting from singlet fission in organic overlayers into Si solar cells is shown to be greatly accelerated by dangling-bond interface defects: The vibrations of Si surface atoms hosting the dangling bonds are associated with defect state energy changes that effectively shuttle the excitons across the interface [5].

[1] O Pankratov, M Scheffler, Phys. Rev. Lett. 75, 701 (1995).
[2] T Frigge et al., Nature 544, 207 (2017); CW Nicholson et al., Science 362, 821 (2018).
[3] M Krenz, U Gerstmann, WG Schmidt, Applied Physics A 128, 480 (2022).
[4] AL Kozub, A Schindlmayr, U Gerstmann, WG Schmidt, Phys. Rev. B 104, 174110 (2021).
[5] M Krenz, Verhandl. DPG (VI) 58, 572 (3/2023).

DS 6.2 Tue 10:00 A 060

Molecule adsorption at wz-Sc(x)Ga(1-x)N surfaces investigated by photo electron spectroscopy — •Fabian Ullmann[1,2], Abdul Qadir Shahbaz[1,2], and Stefan Krischok[1,2] — [1]TU Ilmenau, Ehrenbergstraße 29, 98693 Ilmenau — [2]Zentrum für Mikro- und Nanotechnologie, Gustav-Kirchhoff-Straße 7, 98693 Ilmenau

ScGaN can occur in various crystal orientations. The most important are wurtzite and rock salt formation. Depending on the scandium concentration, a phase transition between these orientations can be found. Wz-ScGaN surfaces with different scandium concentrations were grown by molecular beam epitaxy (MBE) to investigate the near-surface electronic structure. Furthermore, in-vacuo gas interactions (hydrogen, oxygen and water molecules) were analyzed by X-ray (XPS) and ultraviolet photoelectron spectroscopy (UPS).

DS 6.3 Tue 10:15 A 060

EXAFS Analysis of GeSn heteroepitaxial layers — •Sliman Gougam[1], Francesco De Angelis[2], Carlo Meneghini[2], Giovanni Capellini[1,2], and Marvin H. Zoellner[1] — [1]IHP-Leibniz-Institut für Innovative Mikroelektronik, Im Technologiepark 25, 15236 Frankfurt (Oder), Germany — [2]Dipartimento di Scienze, Università Roma Tre, Viale G. Marconi 446 Roma 00146, Italy

There's considerable attention focused on GeSn epitaxial layers due to their promising use in advanced optoelectronic devices. GeSn epitaxial layers quality needs to be finely tuned through growth parameters, as the arrangement of Sn within the Ge crystal lattice may play a crucial role for its application. To study atomic short-range order around Sn in these films, X-ray Absorption Fine Structure (XAFS) spectroscopy is suitable due to its chemical selectivity and sensitivity to local structure. Here, we explore the short-range order of a series of Ge1-xSnx thin films grown on Ge/Si(001) virtual substrates using MBE. Sn K-edge XAFS spectra have been measured at ESRF where results show that Sn is coordinated to 4 Ge nearest neighbors. However, a distinct variation is observed in the next neighbor shell, where the analysis shows that the number of Sn next neighbors is larger than expected for a random distribution, suggesting a chemical ordering with higher Sn-Ge-Sn affinity, which in turn is influenced by growth conditions and film composition. Average modifications in local Sn arrangement among the samples revealed by XAFS, have been correlated with changes in structural properties probed by EDX analysis, which allows to reveal the dispersion of Sn throughout the layers.

15 min. break

DS 6.4 Tue 10:45 A 060

Controllable in-situ growth of nanostructured graphene on cubic-SiC/Si(001) wafers — •Victor Aristov[1,2], Olga Molodtsova[1], Sergey Babenkov[1,3], Dmitrii Potorochin[1,4], Dmitry Marchenko[5], Andrea Locatelli[6], Tevfik Onur Mentes[6], Alessandro Sala[6], and Alexander Chaika[7] — [1]Deutsches Elektronen-Synchrotron DESY, 22607 Hamburg, Germany — [2]Institut fuer Theoretische Physik, Universitaet Hamburg, 22607 Hamburg, Germany — [3]CEA-Saclay, 91190 Gif-sur-Yvette, France — [4]TU Bergakademie Freiberg, D-09599 Freiberg, Germany — [5]HZB für Materialien und Energie, D-12489 Berlin, Germany — [6]ElettraSincrotrone Trieste, I-34149 Basovizza, Trieste, Italy — [7]CRANN, School of Physics, Trinity College Dublin, Dublin 2, Ireland

The graphene grown on low-cost cubic-SiC/Si(001) wafers usually contains nanometer-sized domains with a few different lattice orientations. Here we present the in-situ investigation of layer-by-layer graphene growth on such wafers. The measurements were performed using several methods: scanning tunneling microscopy with atomic resolution, low-energy electron microscopy (LEEM), high-resolution laterally-resolved X-ray photoelectron spectroscopy (micro-XPS), angle-resolved photoelectron spectroscopy (micro-ARPES), and micro low-energy electron diffraction (micro-LEED). The experimental data evidence the opportunity to control the local thickness of the graphene overlayer on the silicon carbide substrate in situ during UHV synthesis.

DS 6.5 Tue 11:00 A 060

Anatase-to-Rutile transformation in $CuTiO_2$ alloys — •Hao Lu[1,2], Martin Becker[1,2], Jan Luka Dornseifer[1,2], and Hao Lu[1,2] — [1]Institute of Experimental Physics I, Justus-Liebig-University, Giessen, Germany — [2]Heinrich-Buff-Ring

Alloying the TiO_2 with CuO_2 yielding $Cu_xTi_{1-x}O_2$ may provide a suitable buffer layer for optical smart windows based on VO_2. We successfully grew polycrystalline $Cu_xTi_{1-x}O_2$ alloys with x up to 31% on float glass and quartz substrates by conventional rf-sputtering employing a TiO_2 ceramic target and Cu wires as Cu source. The surface morphology was measured by SEM. Systematic variations in film morphology were observed concomitant with alterations in the Cu content. We determined the crystal phase of the deposited thin films by XRD and Raman spectroscopy and established a 2D phase map versus substrate temperature during growth and Cu content x. It shows that increasing Cu content considerably lowers the growth temperature where rutile $Cu_xTi_{1-x}O_2$ thin films can be obtained. For x = 23.5%, the minimum growth temperature for the rutile phase still can be as low as 200 °C. Transmission spectroscopy and ellipsometry reveal that the band gap of the $Cu_xTi_{1-x}O_2$ decreases with increasing x. Furthermore, we find that the morphology of the $Cu_xTi_{1-x}O_2$ thin films changes with increasing x. Currently, we are assessing the trade-off between band gap, morphology, and growth temperature required for obtaining the most suitable rutile $Cu_xTi_{1-x}O_2$ buffer layer from the viewpoints of the best materials properties as well as a suitability for future commercialization in smart windows.

DS 6.6 Tue 11:15 A 060

Anisotropic strain relaxation in epitaxially constrained α-(Al,Ga)2O3 thin films on a-plane Al2O3 — •Anna Reis, Michael Hanke, Joao Marcelo Lopes, and Achim Trampert — Paul-Drude-Institut, Hausvogteiplatz 5, 10117 Berlin

Over the past two decades Ga2O3 in its thermodynamically stable β-phase has attracted large scientific interest due to its ultra-wide bandgap enabling the implementation of high-power electronic devices. Lately also the metastable trigonal α-phase of Ga2O3 has received growing attention. Being isostructural to α-Al2O3 ternary (Al,Ga)2O3 can be alloyed across the full compositional range allowing for bandgap engineering between 5.3 eV and 8.8 eV. In order to effectively design heterostructure devices detailed knowledge about strain formation and relief is of fundamental interest.

Thin α-(Al,Ga)2O3 films were epitaxially grown on lattice-mismatched a-plane Al2O3 via molecular beam epitaxy and probed in-situ by X-ray diffraction at the PHARAO facility at BESSY II. Grazing incidence diffraction patterns of the orthogonal (00.6) and (30.0) lattice planes reveal the in-plane strain dynamics of the interface. In the first monolayers the (Al,Ga)2O3 epilayer is found to be

fully pseudomorphic whereas afterwards a partially relaxed layer is formed on top. Within deposition of the first 10-15 nm in-plane compressive strain accumulates preferably along the [100]-direction whilst along [001] strain is relieved exposing the anisotropy of the strain relaxation dynamics [A. Reis et al. Appl. Phys. Lett. 123, 122102 (2023)].

15 min. break

DS 6.7 Tue 11:45 A 060

Determination of Material Compositions from Strain Measurements in Scanning Transmission Electron Microscopy — •Frederik Otto, Laura Niermann, Tore Niermann, and Michael Lehmann — Technische Universität Berlin, Germany

Scanning Transmission Electron Microscopy (STEM) offers atomic-scale resolution for strain analysis by scanning a focused electron probe across a sample and evaluating the spacing between Bragg discs of the resulting electron diffraction patterns at each beam position. These Bragg discs contain features of multiple electron scattering, manifesting as patterns within the diffraction disc. While these patterns contain 3D scattering information, they can compromise the precise detection of diffraction discs' positions. To address this, a common strategy for achieving high-precision strain measurements involves utilizing a precessing electron beam, effectively averaging over multiple patterns.

In this study, rather than disregarding the effects of multiple electron scattering, we focus on carefully evaluating variations in the patterns. A comparative analysis of measured diffraction discs and simulations reveal that these variations originate from deformations appearing at the TEM lamella's surface. These deformations arise as a consequence of stress relaxation in a strained sample during the preparation process. Therefore, this effect is indicative of the strain in the sample (here: the (Al,Ga)N/GaN interface) and thus provides insights into the material's composition. Consequently, we demonstrate a novel method for deducing the composition of layered structures through a single STEM measurement of the strain at the interfaces.

DS 6.8 Tue 12:00 A 060

Precision Through Precession: Enhanced Accuracy of Strain Investigations in Scanning Transmission Electron Microscopy — •Rahel Specht, Frederik Otto, Laura Niermann, Tore Niermann, and Michael Lehmann — Technische Universität Berlin, Germany

In semiconductor development, strain engineering plays a pivotal role, for example in enhancing piezoelectrical effects in AlGaN quantum wells. With ever decreasing device sizes, knowledge of interacial strain at high spatial resolution is an important feedback for the manufacturing process. Scanning Transmission Electron Microscopy (STEM) is capable of resolving the local lattice spacing with nanometer resolution by scanning a focused electron probe over the sample and evaluating the distance of diffraction discs at each respective beam position. However, due to multiple electron scattering and subsequent interference, intensity variations appear in the resulting diffraction discs. These variations hinder the precise detection of the disc's position in the diffraction pattern. In this work, we employ a precessing electron beam to map the position-dependent measurement of lattice spacing of (Al,Ga)N in GaN. Precessing the electron beam effectively averages multiple diffraction patterns, resulting in a more uniform intensity distribution in the diffraction discs. While larger precession angles enhance the precision of disc detection, due to microscope aberrations, higher precession angles also lead to a reduction in spatial resolution. Therefore, we aim to outline a pathway to determining optimal settings for high-resolution strain measurements at heterointerfaces in STEM.

DS 6.9 Tue 12:15 A 060

Spiral and pyramid like structures in solvent prepared crystalline organic C13-BTBT thin films — •Fabian Streller[1], Manuel Johnson[1], Mingjian Wu[2], Erdmann Spiecker[2], and Rainer Fink[1] — [1]Friedrich Alexander Universität Erlangen Nürnberg (FAU), Department Chemistry & Pharmacy — [2]Friedrich Alexander Universität Erlangen Nürnberg (FAU), Department Materials Science und Engineering

The demand for high quality organic thin films for electronic applications is steadily increasing. In OLEDs, OFETs, or sensorics devices, their light weight, flexibility, chemical tunability, and large area preparation makes organic semiconductors valuable resources. Saturated solutions of many aromatic molecules may lead to the formation of large-area crystalline 2D organic film at the solvent-water interface driven by the π-π-interactions for molecular self-organization. In rare cases we observe spiral and pyramidal structures. Excitingly, 3D structures in solvent prepared α-,ω-Hex-6T-Hex show uniform azimuthal rotations in subsequent layers. Here we extend these studies onto mono-substituted C13-BTBT. The interaction towards the water and thus the ratio of intra- vs. interlayer interactions was modified by water surfactants. Compared to previous results, the 3D structures are more extended. Complementary microscopic (AFM, KPFM, TEM, 4D-SCEM) and spectroscopic probes were applied to gain further insight into structure-property relationships and the origin of specific 3D structures. The research is funded by the BMBF (contract 05K22WE2).

DS 6.10 Tue 12:30 A 060

Detailed Microstructure and the Influence of Post-Treatment on CVD TiAlN Wear-Resistant Coatings — •Monica Mead[1], Olof Bäcke[2], Dirk Stiens[3], and Mats Halvarsson[2] — [1]Institute for Materials Science, University of Stuttgart, Germany — [2]Department of Physics, Chalmers University of Technology, Gothenburg, Sweden — [3]Walter AG, Tübingen, Germany

The suitable properties of cubic TiAlN have led to its importance as wear-resistant coating for cutting tools. Preparation by chemical vapour deposition (CVD) has enabled deposition with Al contents above 90 at.% while limiting formation of undesired hexagonal TiAlN. Despite intensive research on the growth of CVD TiAlN coatings, there is no comprehensive understanding of the growth mechanism and intricate microstructure. In addition, research on the effect of blasting on the stress state of wear-resistant coatings is considerable, while the effect on the microstructure is less well studied.

In this work, the microstructure of nano-lamellar low-pressure CVD TiAlN coatings on cemented carbide substrates and the influence of blasting is investigated by scanning electron microscopy (SEM), scanning transmission electron microscopy (STEM) and transmission Kikuchi diffraction (TKD). Two distinct morphologies are observed and connected to specific grain orientations. Furthermore, the proposed growth mechanism suggests an influence of the detailed microstructure on the surface reaction kinetics leading to varying Al/Ti ratios. Blast-treatment of the hard TiAlN coatings introduces plastic deformation, where an influence of the grain orientation is observed.