DS 2: Thin Film Characterisation I: Structure Analysis and Composition

Time: Monday 9:30–13:00 Location: H 0111

DS 2.1 Mon 9:30 H 0111

Probing the oxygen deficit in YBCO thin films with a positron beam — •Markus Reiner[1], Thomas Gigl[1], Rainer Jany[2], German Hammerl[2], and Christoph Hugenschmidt[1] — [1]Heinz Maier-Leibnitz Zentrum (MLZ) and Physik Department E21, Technische Universität München, Lichtenbergstraße 1, 85748 Garching, Germany — [2]Experimentalphysik VI, Elektronische Korrelationen und Magnetismus, Institut für Physik, Universität Augsburg, D-86135 Augsburg, Germany

High quality single-crystalline $YBa_2Cu_3O_{7-\delta}$ (YBCO) thin films were grown epitaxially by pulsed laser deposition. After tempering under different conditions they were characterized by various techniques such as electrical conductivity measurements, X-ray diffraction and electron microscopy. The highly intense reactor based positron beam NEPOMUC was applied for Doppler Broadening Spectroscopy (DBS). A strong correlation between the Doppler broadening of the positron annihilation line and the oxygen deficit was found and its origin was examined by ab-initio calculations. This correlation enabled the study of the kinetics of oxygen out diffusion from the YBCO thin film during in-situ measurements at temperatures up to $673\,K$. Furthermore, the high spatial resolution of the positron beam (up to around $0.3\,mm$) was used for mapping the oxygen deficit and hence, to probe the spatial homogeneity of the oxygen distribution in the examined YBCO films. Financial support within the Project No. 05KI0WOB by the BMBF is gratefully acknowledged.

DS 2.2 Mon 9:45 H 0111

Reference-free, depth dependent characterization of nanoscale materials by combined X-ray reflectivity and grazing incidence X-ray fluorescence analysis — •Philipp Hönicke[1], Matthias Müller[1], Blanka Detlefs[2], Claudia Fleischmann[3], and Burkhard Beckhoff[1] — [1]Physikalisch-Technische Bundesanstalt (PTB), Abbestr. 2-12, 10587 Berlin, Germany — [2]CEA-LETI, 17 rue des Martyrs, 38054 Grenoble, France — [3]imec, Kapeldreef 75, BE-3001 Leuven, Belgium

The accurate in-depth characterization of nanoscaled systems is an essential topic for todays developments in many fields of materials research, especially in the semiconductor related ones.

Synchrotron-based Grazing Incidence X-ray Fluorescence (GIXRF) analysis in combination with X-Ray Reflectometry (XRR) provides additional access to the optical properties of the sample allowing for an improved characterization reliability of GIXRF [1].

Employing the radiometrically calibrated instrumentation at the laboratory of the Physikalisch-Technische Bundesanstalt at the BESSY II synchrotron radiation facility, the combination of XRR and GIXRF allows for a more reliable reference-free quantitative in-depth analysis [1,2,3].

[1] P. Hönicke et al., JAAS 27, (2012), 1432.
[2] M. Müller et al., Materials 7(4), (2014), 3147.
[3] P. Hönicke et al., Solid State Phenomena 195, (2013), 274.

DS 2.3 Mon 10:00 H 0111

Model-based thin film characterization by confocal and total interference contrast microscopy — •Matthias Vaupel — Carl Zeiss Microscopy GmbH, 37081 Göttingen, Germany

It is investigated how optical models improve the accuracy of thickness measurements of a layer stack. Starting with an optical model for ellipsometry on thin films, we derive optical models for total interference contrast (TIC) and confocal microscopy. The confocal model of incoherent superposition of reflections is tested in case of one thin transparent passivation layer on an electronic chip. The model parameters are obtained by a cross-section with FIB-SEM. Thickness and refractive index of the layer are obtained from the confocal measurement by means of the optical model. TIC measures the phase difference of two adjacent spots on a sample. Consequently the phase difference is independent of vertical sample vibration. This allows for pm-vertical resolution of film thickness. As an example the TIC-optical model is applied to obtain the thickness profile of mono- and bilayers of graphene[1] out of the phase profile measured by means of a micrograph recorded in less than 100 ms. The advantages and restrictions of TIC in comparison to AFM, imaging ellipsometry, Mirau-type white light interferometry are discussed.

[1] M. Vaupel et al., Topography, complex refractive index, and conductivity of graphene layers measured by correlation of optical interference contrast, atomic force, and back scattered electron microscopy, J.Appl.Phys. 114, 183107 (2013)

DS 2.4 Mon 10:15 H 0111

Atom probe tomography study of the p-n junction in CIGS thin-film solar cells — •Anna Koprek[1], Oana Cojocaru-Mirédin[1], Christoph Freysoldt[1], Roland Wuerz[2], and Dierk Raabe[1] — [1]Max-Planck-Institut für Eisenforschung, Max-Planck-Straße 1, 40237 Düsseldorf, Germany. — [2]Zentrum für Sonnenenergie- und Wasserstoff-Forschung Baden-Württemberg, Industriestrasse 6, 70565 Stuttgart, Germany.

Solar cells based on $Cu(In,Ga)Se_2$ (CIGS) are one of the most promising among thin film technology. Despite high efficiency over 20%, the cells are still far below the theoretical limit of $\sim$ 30%. Recent studies indicate that the Cd, S, and Cu that interdiffuse across the p-n junction strongly affect the electrical properties of the device.

In this work atomic scale investigation combined with electrical measurements is presented for CdS/CIGS interfaces of CIGS solar cells annealed at different temperatures. The electrical characterization shows decrease of efficiency of the cells with increasing temperature of annealing. By means of atom probe tomography Cd and S diffusion is traced. Indeed, after annealing, besides the Cu-depleted and Cd-enriched CIGS surface, Cd had diffused over a long distance into the absorber. This is contrasted with an as-grown sample for which Cd-enriched and Cu,Ga-depleted zones were observed only at the CIGS surface. The Cd^{2+} ions are expected to occupy Cu rather than Ga sites as Cd^{2+} and Cu^{+} have similar ionic radii. Such Cd^{+}_{Cu} donors could change the p-n junction properties as it was suggested by "Buried homojunction" and "Type inversion of the CIGS surface region" models.

DS 2.5 Mon 10:30 H 0111

Crystallization of Ge nanoparticles in ZrO2-based dielectrics for electrical applications. — •David Lehninger[1], Larysa Khomenkova[2], Frank Schneider[1], Cameliu Himcinschi[3], Volker Klemm[4], David Rafaja[4], and Johannes Heitmann[1] — [1]Institut für Angewandte Physik, TU Bergakademie Freiberg, D-09596 Freiberg — [2]Lashkaryov Institute of Semiconductor Physics, National Academy of Sciences of Ukraine, 03028 Kiev, Ukraine — [3]Institut für Theoretische Physik, TU Bergakademie Freiberg, D-09596 Freiberg — [4]Institut für Werkstoffwissenschaft, TU Bergakademie Freiberg, D-09596 Freiberg

Crystallization of Ge was studied in high-k ZrO2 and TaZrOx host materials using Ge3.6ZrO2/ZrO2 or GeTaZrOx/TaZrOx superlattices, sputtered on Si wafers which were either covered by a SiO2 or Si3N4 starting layer. In order to achieve a better understanding of the Ge-NCs-high-k interface, the formation of Ge-NCs was analyzed by different analytical methods for different annealing temperature. In pure ZrO2, elongated non-spherical Ge-NCs with insufficient control of shape, size, and spatial distribution were observed on wafers covered by SiO2. A Si3N4 starting layer led to formation of multilayer structures with thin Ge films within the ZrO2 matrix. Using TaZrOx as matrix, Ge-NCs with spherical shape and well-defined size were formed in amorphous TaZrOx. In this system, charge trapping phenomena were investigated using MIS structures with single and double layer storage nodes. A memory window of up to 5 V together with a programming-to-flatband-voltage slope of near 1 could be observed.

DS 2.6 Mon 10:45 H 0111

First Synthesis Approach of Layered $ZrSe_2$ and New Misfit Layer Compounds $(PbSe)_{1+\delta}(ZrSe_2)_n$ Using Modulated Elemental Reactants — •Benjamin Eickmeier[1], Matti Alemayehu[2], Robert Zierold[1], Matthias Falmbigl[2], Sage Bauers[2], Johannes Gooth[1], Torben Dankwort[3], Julie Chouinard[2], Carmen Voigt[4], Carsten Ronning[4], David C Johnson[2], and Kornelius Nielsch[1] — [1]Universität Hamburg, Hamburg, Germany — [2]University of Oregon, Eugene, USA — [3]Christian-Albrechts-Universität zu Kiel, Kiel, Germany — [4]Universität Jena, Jena, Germany

A first synthesis approach of the layered dichalcogenide $ZrSe_2$ and a new family of misfit layer compounds $(PbSe)_{1+\delta}(ZrSe_2)_n$ $(n = 1, 2, 3)$

using the physical vapor deposition technique of modulated elemental reactants is reported. Thin films of about 30 nm were deposited onto Si and SiO_2 and characterized with XRD, XRR, and TEM. Composition data was acquired with electron probe microanalysis. These misfit layer compounds are metastable systems and consist of an intergrowth structure between one rock salt sublattice PbSe and n dichalcogenide sublattices $ZrSe_2$, which are alternately stacked upon each other. In the temperature range of 120–200 K hopping transport was observed on the presumable $(PbSe)_{1+\delta}(ZrSe_2)_1$, due to crystalline grains embedded in an amorphous matrix. Previous studies of misfit layer compounds report an unusual decrease up to six times of the in-plane thermal conductivity compared to single dichalcogenides. This trend makes those systems promising alternatives for e.g. thermoelectric applications.

15 min. break.

DS 2.7 Mon 11:15 H 0111

Growth and Properties of Crystalline Silicon deposited on Glass by Steady-State Solution Growth — •Christian Ehlers, Roman Bansen, Jan Schmidtbauer, Franziska Ringleb, Thomas Teubner, and Torsten Boeck — Leibniz Institute for Crystal Growth, Berlin, Germany

For a thin-film silicon solar cell, only several tens of micrometer of crystalline silicon are needed for sufficient light absorption. Therefore we have developed a two-step process for depositing thin silicon layers on inexpensive glass substrates below the softening point of glass.

In the first step a seed layer of nanocrystalline silicon is formed on the heated glass substrates by two different approaches. Either only silicon is evaporated by physical vapor deposition which subsequently forms a thick nanocrystalline layer, or a metal is co-evaporated which leads to the metal-induced crystallization of nanocrystalline silicon. In the second growth step, a subsequent silicon layer is grown on the initial seed layer by steady-state solution growth from a tin or indium solution.

In comparison to traditional liquid phase epitaxy where the supersaturation of the solution is driven by cooling of the metallic solvent, we employ a steady temperature gradient between the silicon source material and substrate. Therefore, growth conditions are similar to a commercial float glass production-process, which potentially allows for the deposition of silicon in a continuous fashion. The electrical and structural properties of the grown silicon layers are analyzed with regard to their suitability for solar cell device production.

DS 2.8 Mon 11:30 H 0111

Comparative Studies in the Physical and Optical Properties of Indium Tin Oxide Films by Various Fabrication Processes — •Akemi Tamanai and Annemarie Pucci — Kirchhoff-Institut für Physik der Universität Heidelberg, Im Neuenheimer Feld 227, 69120 Heidelberg

Tin-doped indium oxide (ITO) is a highly transparent n-type semiconductor material in VIS\UV region caused by its wide band gap. For improving the resistivity and high optical transmittance performances of ITO films, various ITO fabrication processes have been introduced so far. Since their physical and optical properties are strongly influenced by fabrication processes such as sputtering, electron beam evaporation, and pulsed-laser deposition, changes in, for instance, the annealing temperature and environment, the surface resistance, and its morphology directly affect both properties. Hence, in order to enhance and deepen the understanding for the dielectric properties of various ITO films, spectroscopic IR ellipsometry (Woollam IR-VASE) measurements have been carried out for determining the dielectric constants (ε_1 and ε_2), the degree of electric scattering time (τ), the plasma frequency (ω_p), and the damping constant (γ) defined in the Drude model which describes free-carrier contribution to dielectric properties.

DS 2.9 Mon 11:45 H 0111

Microstructure and mechanical properties of Pd-Si amorphous thin films — •Sree Harsha Nandam[1], Di Wang[1], Ruth Schwaiger[2], Zbigniew Sniadecki[1], Yulia Ivanisenko[1], Herbert Gleiter[1], and Horst Hahn[1] — [1]Institute for Nanotechnology, Karlsruhe Institute of Technology, D-76344 Eggenstein-Leopoldshafen, Germany. — [2]Institute for Applied Materials, Karlsruhe Institute of Technology, D-76344, Eggenstein-Leopoldshafen, Germany.

Amorphous thin films of Pd80Si20 were co-sputtered using Pd and Si targets in a direct current sputtering system. X-ray diffraction of the thin films clearly revealed the amorphous nature and the microstructure of the films showed a morphology consisting of nanograins. Such amorphous nanograined thin films are termed as "nanoglasses" in the present day literature. Transmission electron microscopic studies on these films confirmed the amorphous nature of the films with little variation of the composition (segregation) on the scale of the nanograined substructure. Annealing of the thin films below 200°C (for 24h) caused no big change of the nanostructure, at temperatures around 250°C (for 24h) crystallization of the films was observed. Nanoindentation of the films showed a slightly higher Young's modulus compared to the melt-spun counterparts reported in the literature. Possible reasons for the appearance of a "nanoglass" structure and for the increased Young's modulus of the Pd-Si amorphous thin films are discussed.

DS 2.10 Mon 12:00 H 0111

Cu-poor/Cu-rich transition of co-evaporated CuInSe2: Na prevents annihilation of planar defects — •Helena Stange[1,2], Stephan Brunken[2], Humberto Rodriguez-Alvarez[2], Dieter Greiner[2], Christian Alexander Kaufmann[2], Anja Scheu[2], Jakob Lauche[2], Norbert Schäfer[2], Daniel Abou-Ras[2], and Roland Mainz[2] — [1]TU Berlin, Institut für Werkstoffwissenschaften — [2]Helmholtz-Zentrum-Berlin für Energie und Materialien

Highest efficiencies of Cu(In,Ga)Se2 solar cells have been achieved with absorbers deposited by a 3-stage-based co-evaporation. The Cu-poor/Cu-rich transition during the growth process and the presence of small amounts of Na and K are known to be favorable for solar cell performance. We investigate the influence of Na on the structural evolution of CuInSe2 by interrupting the growth process at overall Cu-poor and Cu-rich film compositions. CuInSe2 layers without and with NaF precursor are prepared at growth temperatures below 400°C and analyzed by XRD, XRF, EDX and GDOES. For the samples without NaF a XRD signal characteristic for planar defects are observed only for the Cu-poor samples, while for the Cu-rich samples the signal vanishes. In contrast, for the Na-containing absorbers the characteristic signal are present also for the Cu-rich samples. For Cu(In,Ga)Se2 layers without NaF precursor, real-time XRD shows annihilation of planar defects coinciding with the Cu-poor/Cu-rich transition. We conclude that the presence of Na during growth impedes annihilation of planar defects during the Cu-poor/Cu-rich transition in CuInSe2 at low growth temperatures.

DS 2.11 Mon 12:15 H 0111

In situ X- Rays measurements of lattice expansion on metallic superlattices. — •Sotirios A. Droulias, Lennard Mooij, Gunnar K. Palsson, Xin Xiao, Vassilios Kapaklis, Bjorgvin Hjorvarson, and Max Wolff — Materials Physics, Division of Physics and Astronomy, Uppsala University, Sweden

Transition metals are exceptional candidates for hydrogen storage applications since large H quantities can be effectively absorbed. By growing superlattices of such materials, finite size and proximity effects can be investigated. In our work, we present expansion measurements of Cr-V and Fe-V superlattices by using in situ X-Ray diffraction. From the expansion measurements the elastic constants of the material can be extracted and related to the hydrogen interaction which is mediated by elastic forces. A complete and accurate description of the host lattice under various H pressures (concentrations) and different temperatures provide us with a unique tool for understanding the system*s thermodynamics. From the volume expansion one can derive the site occupancy of H in the host material and if combined with other techniques, such as optical transmition, and curvature measurements a complete image of the system can be drawn. The different techniques can be correlated directly through resistivity measurements. This forms the basis for understanding the observed phase transitions in such systems and the influence of finite size and proximity. Furthermore, characterization of the host lattice under various conditions is a detailed quality description after repeated loading and unloading of the hydrogen containing material.

DS 2.12 Mon 12:30 H 0111

GISAXS investigation of growth of Ag nanoparticles from atomic deposition: experiment and simulation — Celine Durniak, •Marina Ganeva, Gennady Pospelov, Walter Van Herck, and Joachim Wuttke — Jülich Centre for Neutron Science, Forschungszentrum Jülich, Outstation at MLZ, Garching, Germany

Grazing-Incidence Small-Angle X-ray Scattering (GISAXS) is a powerful tool for the investigation of the growth of noble metal particles on various substrates. Understanding of the underlying growth mech-

anisms allows for a control of the properties of the nanocoatings. Due to the complexity of the GISAXS data analysis, appropriate software is required to deduce the structural information from the measured GISAXS pattern. Here we present our software BornAgain [1]. BornAgain is a multi-platform open-source project that aims at supporting scientists in the analysis and fitting of their GISAS data, both for synchrotron (GISAXS) and neutron (GISANS) facilities. As a case study, the growth of Ag nanoparticles on Si and PTFE substrates during DC magnetron sputtering of Ag is taken. The GISAXS investigation of structural properties was performed at the high-resolution diffractometer GALAXI (Jülich).

[1] http://bornagainproject.org

DS 2.13 Mon 12:45 H 0111

In-situ GIXD/GISAXS investigation of intercalation processes in bulk-heterojunction organic solar cells — •Marvin Berlinghof[1], Thaer Kassar[1], Nusret Sena Güldal[2], Christoph Brabec[2], and Tobias Unruh[1] — [1]Chair for Crystallography and Structural Physics, Friedrich-Alexander-Universität Erlangen-Nürnberg — [2]Chair for Materials for Electronics and Energy Technology, Friedrich-Alexander-Universität Erlangen-Nürnberg

Organic solar cells are a promising technology for cost efficient energy production, because – compared to traditional anorganic solar cells – they only need low temperatures during production, are cheaper to produce in industrial-scales and are flexible. Doctor-bladed thin film solar cells were investigated in-situ during drying by GIXD and GISAXS using our in-house X-ray source and synchrotron radiation. Using different polymers (pBTTT, P3HT, PQT) as organic semiconductors and a wide variety of fullerenes, we observed a change in a different behavior of the d-spacing of pure polymers and polymer-fullerene-mixtures during drying. While the pure polymers showed a decreasing of the d-spacing of the (100)-polymer-peaks during drying, caused of the evaporation of solvent. In the mixtures this d-spacing stayed constant during drying, which is a indicator for intercalation of fullerenes in-between the polymer sheets, which directly influences the efficiency of the organic solar cells. Furthermore the fullerenes hinder the presence of higher diffraction orders and with that decreases the crystallinity of the films. In addition the results of our GISAXS investigation and depth-dependend measurements will be presented.