DS 3: Oxide Semiconductors for Novel Devices (Focussed Session): Session I

The class of semiconducting oxides includes low temperature processed amorphous thin films for bendable electronics and display technology as well as highly crystalline materials such as the wide band group-III sesquioxides being interesting for UV and DUV photo sensors, power electronics and even memristors. This session sets a focus on physical properties of such oxides, their growth methods and heterostructures for demonstrator devices. This focus session is supported by the Leibniz ScienceCampus GraFOx.

Organized by

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Time: Monday 9:30–13:15 Location: E 020

DS 3.1 Mon 9:30 E 020

Switching kinetics of VCM-based resistive memories at ultrafast time scales — •Moritz von Witzleben[1], Viktor Havel[1], Karsten Fleck[1], Andreas Kindsmüller[1], Rainer Waser[1,2], Stephan Menzel[2], and Ulrich Böttger[1] — [1]Institut für Werkstoffe der Elektrotechnik II, RWTH Aachen University, 52064 Aachen, Germany — [2]Peter Grünberg Institut (PGI 7), Forschungszentrum Jülich, 52425 Jülich, Germany

Redox-based resistive memories (ReRAM) are likely to overcome the challenges that nowadays memories are facing as they show auspicious properties regarding switching times, scaling, retention and endurance. For their application, fast switching times are mandatory for the writing process whereas their resistive state must be retained during many read-out operations. This issue is known as voltage-time dilemma and can be overcome by an extremely nonlinear correlation between the switching time and the applied voltage. Therefore, the switching kinetics of $Pt/Ta_2O_5/Ta$ and $Pt/ZrO_2/Ta$ VCM cells are measured over almost 15 orders of magnitude between 10^4 s and 120 ps. The ReRAM cells were incorporated in coplanar waveguide (CPW) structures to provide proper impedance matching at the contact surfaces. Our results indicate that the voltage-time dilemma can be addressed with VCM cells. Furthermore, the switching kinetics at time scales below 10 ns are not limited by conduction mechanisms or electrochemical reactions in the oxide layer or at its interfaces, but RC times play a decisive role due to the capacities of the CPW structure.

DS 3.2 Mon 9:45 E 020

Memsensors: How to design devices with enhanced capabilities in neuromorphic engineering — •Alexander Vahl[1], Jürgen Carstensen[2], Sören Kaps[2], Oleg Lupan[2], Thomas Strunskus[1], Rainer Adelung[2], and Franz Faupel[1] — [1]Christian-Albrechts University at Kiel, Institute for Materials Science, Chair for Multicomponent Materials, Kaiserstr. 2, 24143, Kiel, Germany — [2]Christian-Albrechts University at Kiel, Institute for Materials Science, Chair for Functional Nano Materials, Kaiserstr. 2, 24143 Kiel, Germany

Memsensors are a class of devices combining resistive switching and sensing properties. Apart from their inherited properties, pinched I-V hysteresis and stimulus dependent resistivity, memsensors have the capability to adapt to an external stimulus. This adaptation shows striking similarities to adaptation in biological neuronal systems, making memsensors ideal candidates for applications in neuromorphic engineering. In addition, the resistive switching is strongly dependent on the external stimulus. We propose a simple equivalent circuit containing two memristors, one in parallel and one in series to a linear sensor. This model allows to understand a multitude of experimental findings and implies a large predictive power for further optimization of memsensing devices and their application in neuromorphic engineering. Based on the model and experimental findings we propose design rules for memsensors that will facilitate further research on memsensitive systems.

DS 3.3 Mon 10:00 E 020

Defect Investigation of $CuBi_2O_4$ Photocathodes for Solar Water Splitting — •Michael Sahre, Marlene Lamers, Matthias Müller, Fatwa F. Abdi, and Roel van de Krol — Institute for Solar Fuels, Helmholtz-Zentrum Berlin, Berlin, Germany

The rise of global warming requires the development of novel sustainable carbon-free energy sources. One promising method is by directly converting water into hydrogen and oxygen using sunlight; the process is usually called "solar water splitting". In such process, a semiconductor is used to absorb the light, separate the photo-generated charge carriers, and produce hydrogen and/or oxygen on its surface. A promising candidate as the semiconductor is $CuBi_2O_4$ due to its suitable bandgap of 1.8 eV (theoretical solar-to-hydrogen efficiency of 24 %) and favorable band positions.[1,2] However, poor charge carrier transport and surface charge transfer limit the photocurrent.[2]

In this work, we attempt to alleviate the above-mentioned limitations by modulating the defects in $CuBi_2O_4$ thin films through various annealing conditions. Time resolved microwave conductivity measurements show an improved charge carrier transport after the high temperature annealing. Simultaneously, this treatment leads to a surface modification and enhanced electron transfer from the semiconductor into the electrolyte, so that the AM1.5 photocurrent is significantly increased. The interplay between the annealing condition, defect formation and photoelectrochemical performance will be discussed.

[1] Chen *et al.*, *J. Mater. Res.* 25 (2010) 3
[2] Berglund *et al.*, *Chem. Mater.* 28 (2016) 4231

DS 3.4 Mon 10:15 E 020

A pulsed laser deposition technique to control the composition of ternary thin films in growth direction demonstrated on the $Mg_xZn_{1-x}O$ alloy — •Max Kneisz, Philipp Storm, Gabriele Benndorf, Holger von Wenckstern, and Marius Grundmann — Universität Leipzig, Felix-Bloch-Institut für Festkörperphysik, Leipzig, Germany

In conventional pulsed laser deposition (PLD) a continuous variation of the composition of ternary thin films in growth direction is not possible since the number of discrete alloy combinations is limited by the amount of targets which can be mounted in the setup. We therefore propose a technique using only a single elliptically-segmented target with two regions of different composition. We control the Mg/Zn ratio of the thin films by varying the radial position of the PLD laser spot on the target and thereby changing the ratio of the path lengths of the laser spot in the different regions. In analogy to our approach for lateral continuous composition spreads [1] (lateral CCS), we call this method vertical CCS. We will show that we are able to control the composition of $Mg_xZn_{1-x}O$ thin films in growth direction quasi continuously or stepwise. Therefore films with varying single Mg-contents are grown using the new technique on highly conductive ZnO:Al buffer

layers. The Mg-content in the films is determined by low temperature photoluminescence spectroscopy. The structural and optical quality is similar to films grown via conventional PLD.
[1] H. von Wenckstern *et al.*, CrystEngComm **15**, 10020 (2013)

DS 3.5 Mon 10:30 E 020

Considerations in the Stability of Multicomponent Oxide Alloys — •Christopher Sutton[1], Robert J.N. Baldock[2], Luca M. Ghiringhelli[1], and Matthias Scheffler[1] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany — [2]Ecole Polytechnique Federale de Lausanne, Lausanne, Switzerland

Identification of stable crystalline materials for a mixture of two (three) components requires examination of the lowest free energy of approximately 2^N (3^N) configurations, where N is the number of atoms in the unit cell. Cluster expansion-based energy functions offer a numerically efficient approach for estimating the stability of new potential alloys. Combining this approach with the nested sampling algorithm, which is a Bayesian Markov chain Monte Carlo method, allows for a one-shot calculation of the phase diagram as a function of composition and temperature. Our results for stable ternary and quaternary mixtures in various crystalline symmetries of group-III oxides with the formula $(In_xGa_yAl_z)_2O_3$ where $x+y+z=1$ will be presented. The key aspects that determine the stability of these materials will be discussed. With an extensive search over configurational space, statistical learning is performed for the bandgaps and stabilities to identify structure-property relationships between the targeted properties (e.g., optical transparency) and the fundamental chemical and physical parameters that control these properties.

DS 3.6 Mon 10:45 E 020

Influence of nitrogen annealing on the properties of spray pyrolysis grown In-doped ZnO thin films — Dilawar Ali[1,2], Muhammad Z. Butt[1], David Caffrey[2], Igor V. Shvets[2], and •Karsten Fleischer[2] — [1]Department of Physics and Centre for Advanced Studies in Physics, GC University Lahore-54000, Pakistan — [2]School of Physics and Centre for Research on Adaptive Nanostructures and Nanodevices (CRANN), Trinity College Dublin, Dublin 2, Ireland

The properties of a low cost transparent conducting oxide (TCO) – spray pyrolysis grown ZnO thin films doped with indium have been investigated. We analyze the optical, electrical, and crystallographic properties as function of In content with a specific focus on post-growth heat treatment of these thin films at 320°C in an inert, nitrogen atmosphere, which remarkably improves the films electrical properties. The effect was found to be dominated by nitrogen induced grain boundary passivation, identified by a combined study using in-situ resistance measurement upon annealing, X-ray photoelectron spectroscopy, photoluminescence and X-ray diffraction studies. We also highlight the chemical mechanism of morphologic and crystallographic changes found in films with high indium content. In optimized growth and post-annealing conditions, ZnO:In with a resistivity as low as $2\times10^{-3}\,\Omega$cm and high optical quality has been obtained using low cost spray pyrolysis.

DS 3.7 Mon 11:00 E 020

Modulation of the In_2O_3 surface electron transport properties by acceptor doping — •Alexandra Papadogianni[1], Julius Rombach[1], Theresa Berthold[2], Stefan Krischok[2], Marcel Himmerlich[2], and Oliver Bierwagen[1] — [1]Paul-Drude-Institut für Festkörperelektronik, Hausvogteiplatz 5–7, 10117 Berlin, Germany — [2]Institut für Physik und Institut für Mikro- und Nanotechnologien, Technische Universität Ilmenau, PF 100565, 98684 Ilmenau, Germany

In_2O_3 is a natively *n*-type transparent semiconducting oxide possessing a surface electron accumulation layer (SEAL) like several other relevant oxides, such as SnO_2 and ZnO. While the SEAL is within the core of In_2O_3-based conductometric gas sensors, it hinders numerous applications of In_2O_3 in electronic devices that require the formation of Schottky contacts. Tunability of the SEAL is hence necessary to unlock the entire spectrum of possible device applications of In_2O_3.
Oxygen plasma treatment of the In_2O_3 surface has been previously shown to effectively deplete surface electrons. Annealing the material, however, has been proven to reverse this effect, which renders this solution unsuitable for devices operating at elevated temperatures. As an alternative, we demonstrate strong reduction of the SEAL by doping with the deep compensating acceptors Mg and Ni performing X-Ray Photoelectron Spectroscopy (XPS) and transport measurements on high quality single-crystalline In_2O_3(111) films grown by plasma-assisted molecular beam epitaxy (PA-MBE). This method also allows for fine-tuning of the electrical transport properties of the SEAL through controllably selecting its degree of depletion.

15 min. break.

DS 3.8 Mon 11:30 E 020

Confining memristive filaments in TiO_2 thin films by Au nanoparticles — •Christian Rodenbücher[1], Nabeel Aslam[1], Dominik Wrana[1,2], Hehe Zhang[1], Hongchu Du[1], Michael Prömpers[1], Dirk Mayer[1], and Susanne Hoffmann-Eifert[1] — [1]Forschungszentrum Jülich, ER-C, ICS, PGI, and JARA-FIT — [2]Jagiellonian University, Institute of Physics, Krakow

The memristive effect in transition metal oxides has attracted much attention promising the design of fast non-volatile and energy-efficient memory devices which additionally would offer the opportunity of hardware-based neuromorphic computing. On the nanoscale, the underlying resistive switching effect was found to be related to conducting filaments evolving during an initial electroforming step. In order to improve the performance of the memristive devices, a control of the filament formation is of high importance. Here, we follow the approach of introducing Au nanoparticles at the Pt/TiO_2 interface of ALD-grown films. Using local-conductivity atomic force microscopy (LC-AFM) we show that the conductivity through the TiO_2 above the nanoparticles is significantly higher than above the surrounding Pt-coated substrate which can be attributed to the different material properties of the two metals (here Au and Pt) forming the interface to the oxide layer. Hence, Au nanoparticles can be used to determine the position of conducting paths as origin for filament formation on the nanoscale. We employ this effect to build up Pt/TiO_2/Pt nanocrossbar devices with embedded Au nanoparticles showing enhanced switching characteristics with significantly reduced forming voltage.

DS 3.9 Mon 11:45 E 020

Epitaxial Stabilization of NbO_2 on TiO_2(110) — •Jos Emiel Boschker[1], Saud Bin Anooz[1,2], Benjamin Kalas[3], Toni Markurt[1], Manfred Ramsteiner[4], Sverre Vegard Pettersen[5], Jostein Kvaal Grepstad[5], Martin Albrecht[1], Péter Petrik[3], and Jutta Schwarzkopf[1] — [1]Leibniz Institute for Crystal Growth, Max-Born-Str. 2, Berlin, D-12489, Germany — [2]Physics Department, Faculty of Science, Hadhramout University, Mukalla, 50511, Yemen — [3]Institute of Technical Physics and Materials Science, Konkoly-Thege Rd. 29-33, 1121 Budapest, Hungary — [4]Paul-Drude-Institut für Festkörperelektronik, Hausvogteiplatz 5-7, Berlin, D-10117, Germany — [5]Department of Electronic Systems, Norwegian University of Science and Technology, Trondheim, Norway

Niobium oxides have promising physical properties that can be exploited in electronic devices, such as selector and memory devices. Moreover, NbO_2 has attracted considerable scientific interest, as it exhibits a semiconductor-metal transition at 1080K. However, the lack of high quality material complicates fundamental studies of their properties. In this paper, we report on the epitaxial stabilization of NbO_2 thin films grown on TiO_2 substrates by pulsed laser deposition. The anisotropic properties of films grown at different substrate temperatures were determined by various structural and optical methods. The results indicate a phase transition from the rutile structure to a distorted rutile structure with decreasing substrate temperature.

DS 3.10 Mon 12:00 E 020

Characterization of the dielectric function of $RScO_3$ type scandates — •Sergey Kuznetsov[1,2], Martin Feneberg[1], and Rüdiger Goldhahn[1] — [1]Institute of Experimental Physics, Otto-von-Guericke University Magdeburg, Germany — [2]Drexel University Department of Physics, Philadelphia, USA

Due to their high energy band gap and their structural stability towards temperature differences, wide-gap Perovskite structured oxides of type $RScO_3$ are promising candidates for replacing SiO_2 as high-k gate dielectrics. These Sc-based crystals are also among best available bulk substrates for the epitaxial growth of high-quality ferroelectric thin films, however, only limited data on their optical properties is available. Here, we present the results of spectroscopic ellipsometry from the infrared (0.04eV) up to the ultraviolet (6.6eV) at room temperature on $SmScO_3$, $GdScO_3$, $TbScO_3$, and $DyScO_3$ bulk single-crystals. The purpose of the experiments was to determine the in-plane components of the real and imaginary dielectric functions (DF). The analysis revealed 4 phonon modes in the extraordinary DF, and 8 to

9 modes in the ordinary DF. The phonon frequencies are observed to shift systematically with the atomic mass of the rare earth component of the perovskites. This effect was observed in the ordinary and extraordinary DF. Data obtained also indicate the onset of interband absorption at 6 ± 0.1 eV for $GdScO_3$ and $DyScO_3$, and at 5 ± 0.1 eV in the $SmScO_3$ and $TbScO_3$, with a local maximum at 5.4 eV prior to exhibiting similar behavior to the other two samples. A comprehensive overview of the anisotropic dielectric functions will be shown.

DS 3.11 Mon 12:15 E 020

Ferro- and antiferroelectricity in oxygen defficient hafnia and zirconia — •Konstantin Z. Rushchanskii, Stefan Blügel, and Marjana Ležaić — Peter Grünberg Institut, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany

Conventional perovskite ferroelectrics as materials for ferroelectric RAM (FeRAM) suffer from poor CMOS-compatibility and limited scalability. In this sense, the discovery of ferroelectricity in Si:HfO_2 thin films [1] opens new prospectives for HfO_2-based materials because they are CMOS-compatible, and the robust ferroelectric behavior scales down to a film thickness below 10 nm. Recent discovery of ferroelectricity in undoped HfO_2 [2] emphasizes the importance of oxygen deficiency for the desired ferroelectric properties and the quest for a better microscopic understanding.

We present results of our Density Functional Theory combined with an evolutionary algorithm [3] based study of metastable structures in Hf-O and Zr-O solid solutions. We will show suboxides of HfO_2 and ZrO_2, which simultaneously possess a resistive switching capability and (anti)ferroelectricity, i.e. both properties originate from the same structures. We will discuss a possible origin of the ferro-antiferroelectric crossover, observed in $Hf_xZr_{1-x}O_2$ thin films [4].

We acknowledge the support by DFG via SFB 917 "Nanoswitches".
[1] T. Böscke et al, Appl. Phys. Lett. **99**, 102903 (2011); [2] P. Polakowski, J. Müller, Appl. Phys. Lett. **106**, 232905 (2015); [3] http://uspex.stonybrook.edu; [4] J. Müller et al, Nano Lett. **12**, 4318 (2012).

DS 3.12 Mon 12:30 E 020

Direct imaging of reversible massive oxygen transport induced by ionic liquid gating and creation of meso-structures — •Bin Cui, Peter Werner, Tianping Ma, and Stuart Parkin — Max Planck Institute for Microstructure Physics, 06120 Halle, Germany

Ionic liquid (IL) gate induced oxygen migration at the surface of oxide thin films has been proposed to be a powerful tool for manipulating their bulk electronic properties and structures. Advancements in this direction requires an in-depth understanding of the correlation between oxygen transport manner, atomic-scale structural transition, and macroscopic physical responses under IL gating.

Here we directly image the processes of oxygen transport, using in-situ gating, within a TEM, as we induce the transformation between brownmillerite SrCoO2.5 and perovskite SrCoO3. A massive ~0.5 oxygen per formula unit, is observed to be reversibly extracted or injected over several minutes. These changes are consistent with vertical oxygen transport perpendicular to the thin film surface on nanoscale, and lateral transport parallel to the surface on microscale.

Using IL gating through lithographically patterned orifices in resist at the surface of various oxide films with different anisotropic oxygen transport, we show that various three-dimensional metallic structures such as cylinders and rings can be realized.

Our results not only reveal the oxygen transport manner in IL gating, but also provide a roadmap to the complex meso-structures construction in ion-transporting materials from their exterior surfaces.

DS 3.13 Mon 12:45 E 020

Room temperature fabricated all-oxide junction field-effect transistors and inverters on rigid and flexible substrates — •Peter Schlupp, Sofie Vogt, Holger von Wenckstern, and Marius Grundmann — Universität Leipzig, Felix-Bloch-Institut für Festkörperphysik, Leipzig, Germany

Room temperature (RT) fabrication of electronic devices saves energy within the fabrication process and enables the usage of thermally unstable but flexible substrates. One material class that can be fabricated at RT showing promising properties are amorphous (or nanocrystalline) oxide semiconductors (AOS). The most commonly used n-type AOS is indium gallium zinc oxide which is already successfully used as channel material in pixel drivers for active matrix displays [1]. However, because indium is rare and expensive we use n-type zinc tin oxide (ZTO) as channel layer material which shows easily controllable electrical properties [2,3].

We present junction field-effect transistors (JFETs) using p-type amorphous zinc cobalt oxide and nanocrystalline nickel oxide as gate diodes. The ZTO films were grown by magnetron sputtering on glass and flexible polyimide while the p-type layers were grown by pulsed laser deposition. The characteristics of the JFETs and inverters based on them fabricated on both substrates will be discussed. Additionally, their changes after bending the flexible samples will be presented.

[1] Wellenius *et al.*: J. Display Technol. **5**, 438 (2009)
[2] Jayaraj *et al.*: J. Vac. Sci. Technol. B, **26(2)**, 495 (2008)
[3] Schlupp *et al.*: MRS Proceedings **1633**, 101 (2014)

DS 3.14 Mon 13:00 E 020

Surface hole accumulation layer in NiO created by oxygen plasma treatment — •Melanie Budde[1], Carsten Tschammer[1], Theresa Berthold[2], Marcel Himmerlich[2], Stefan Krischok[2], and Oliver Bierwagen[1] — [1]Paul-Drude-Institut für Festkörperelektronik, Hausvogteiplatz 5-7, 10117 Berlin, Germany. — [2]Institut für Physik und Institut für Mikro- und Nanotechnologien MacroNano, Technische Universität Ilmenau, PF 100565, 98684 Ilmenau, Germany.

Nickel oxide (NiO) is a transparent and semiconducting p-type oxide, which is interesting for various applications, for example photovoltaics, diodes or other electronic devices. However, our high quality NiO layers grown by plasma-assisted MBE are semi-insulating with sheet resistances above 109Ω. Doping of NiO, for example with Lithium, is known to increase its p-type bulk conductivity. In this contribution, we demonstrate the formation of a surface hole accumulation layer generated by oxygen plasma treatment and characterize its conductivity. For this purpose, MBE grown NiO layers on MgO(100) are modified by an oxygen plasma process after growth. The sheet resistance was measured by Van-der-Pauw measurements and a p-type conductivity was confirmed by Seebeck measurements. Furthermore, the results allowed to determine the approximate hole concentration. X-ray photoelectron spectroscopy (XPS) demonstrates a shift of the surface Fermi level towards the valence band maximum, corroborating the formation of a surface hole accumulation layer. The results are compared to published data on p-doped NiO.