HL 37: Oxide Semiconductors

Time: Thursday 9:30–13:00 Location: EW 203

HL 37.1 Thu 9:30 EW 203

Polaronic entropy stabilizes mixed-valence compound K_4O_6 — Patrick Merz[1], Claudia Felser[1], Martin Jansen[1,2], •Christoph Freysoldt[3], and Jörg Neugebauer[3] — [1]Max-Planck-Institut für Chemische Physik fester Stoffe, Nöthnitzer Str. 40, 01187 Dresden — [2]Max-Planck-Institut für Festkörperforschung,Heisenbergstr. 1, 70569 Stuttgart — [3]Max-Planck-Institut für Eisenforschung GmbH, Max-Planck-Str. 1, 40273 Düsseldorf

Alkali sesquioxides, M_4O_6, contain peroxide O_2^{2-} and superoxide O_2^- ions and are prime examples of correlated open-shell p-electron systems. They show a rich phase diagram arising from the interplay of magnetic, orbital, and charge ordering depending on temperature and the cation size (chemical pressure). We report on the reversible endothermic formation of K_4O_6 from the separate peroxide and superoxide phases at ≈350° C. Rapid cooling leads to a metastable, charge-ordered phase, that decomposes when heated to 100° C. In order to better understand thermodynamic stability by means of density-functional theory (DFT) calculations, we have developed a scheme to include Hubbard-U corrections to account for correlations between the molecular sites. The calculations suggest that the stability at elevated temperatures is driven by the polaronic entropy associated with charge hopping between the O_2 molecular units. The ionic displacements associated with polaron hopping are complex and large, sometimes >1 Å in distance. Our findings highlight that the commonly used separation between electronic and phononic excitations in finite-temperature materials modelling may miss crucial stabilization mechanisms.

HL 37.2 Thu 9:45 EW 203

Controlling and Analyzing the Conductivity of TiO2 Nanorod Arrays — •Carola Ebenhoch, Julian Kalb, Elise Sirotti, Domenik Vögel, and Lukas Schmidt-Mende — Department of Physics, University of Konstanz, Germany

Besides being an important semiconducting material in industry for electrical components such as transistors, diodes, sensors, solar cells, and many more [1], TiO2 can easily be grown in nanometer dimensions (e.g. thin films via SALD and nanowires via hydrothermal growth), which enables the access to nanodevices. Due to an increased surface to bulk ratio for nanowires, effects from surface states become even more important for electronic properties, which still require extensive investigations. In this regard, we present conductivity measurements on nanorod arrays grown on a FTO substrate via hydrothermal growth, contacted with a gold tip as top electrode. The density of defect states at the surface, e.g. oxygen vacancies, were modified using a post annealing process in different atmospheres. With this method, temperature dependent conduction mechanisms for highly conductive nanowires, containing a large amount of oxygen vacancies at the surface [2], or rather low conductive nanowires could be identified.

[1] Yu, X., et al., Metal oxides for optoelectronic applications. Nat Mater, 2016. 15(4): p. 383-96.

[2] Folger, A., et al., Tuning the Electronic Conductivity in Hydrothermally Grown Rutile TiO(2) Nanowires: Effect of Heat Treatment in Different Environments. Nanomaterials (Basel), 2017. 7(10).

HL 37.3 Thu 10:00 EW 203

Spray Coating of Transparent Oxide Semiconductors - β-Ga_2O_3, In_2O_3-Ga_2O_3-ZnO, and ZnO Thin Films — •Constance Schmidt, Axel Fechner, and Dietrich R.T. Zahn — Semiconductor Physics, Technische Universität Chemnitz, 09107 Chemnitz, Germany

Transparent, semiconducting materials offer new opportunities for optoelectronic devices, like flat panel displays. The most remarkable property of these transparent oxide semiconductors (TOSs) is their wide band gap (>3 eV). Besides established deposition methods like MBE and CVD, cost efficient preparation methods are of great interest. Spray coating, the deposition technique of our choice, is one of these cost efficient methods to prepare TOS thin films. Samples were prepared by spray coating using a solution containing $Ga(NO_3)_3$, $Zn(NO_3)_2$, and In_2O_3, Ga_2O_3, or ZnO. For Ga_2O_3 and ZnO we solved their nitrates in H_2O and Ethanol, for In_2O_3-Ga_2O_3-ZnO thin films we used isopropyl alcohol as solvent. N_2 is always used as carrier gas. Post annealing (300 °C - 1300 °C) needs to be applied to all thin films prepared. Optical respectively vibrational properties were studied using spectroscopic ellipsometry and Raman spectroscopy. X-ray diffraction was employed to investigate structural and phase purity. Topological and morphological characterization was performed using scanning electron microscopy. Finally, the conductivity was determined by the four-point probe method. It is demonstrated that high quality TOSs thin films using cost efficient spray coating.

HL 37.4 Thu 10:15 EW 203

Excitonic absorption and optical properties of ZnO films — •Stefan Zollner[1], Nuwanjula Samarasingha[1], Zachary Yoder[1], Dipayan Pal[2], Aakash Mathur[2], Ajaib Singh[2], Rinki Singh[2], and Sudeshna Chattopadhyay[2] — [1]New Mexico State University, Las Cruces, NM, USA — [2]IIT Indore, Indore, India

Using spectroscopic ellipsometry from 0.5 to 6.5 eV, we have investigated the thickness dependence of the optical constants (complex refractive index, dielectric function) of zinc oxide thin films grown on Si and SiO2 by atomic layer deposition. Independent characterization of the films was carried out using powder x-ray diffraction (texture, grain size), x-ray reflectance (density, thickness, roughness), and atomic force microscopy (roughness). The validity of the ellipsometry results was verified with careful error analysis, including a uniqueness fit for the thickness parameter, and comparison of the thickness with XRR. The dielectric function of ZnO layers was described with an oscillator model guided by measurements for bulk ZnO. Our results show convincingly that both the real and imaginary part of the dielectric function of ZnO on Si decrease with decreasing film thickness. We attribute this thickness dependence to the optical transition matrix element (electron-hole overlap) modified by the interface. The results are not so clear for ZnO on SiO2.

HL 37.5 Thu 10:30 EW 203

Tunable intersubband transitions in ZnO/ZnMgO multiple quantum wells in the mid infrared spectral range — •Laura Orphal, Sascha Kalusniak, Oliver Benson, and Sergey Sadofev — Institut für Physik, Humboldt-Universität zu Berlin, Newtonstr. 15, 12489 Berlin, Germany

ZnO is considered as a promising material for mid IR intersubband devices, and possibly also for the THz regime [1]. We demonstrate the ability to control intersubband transition (ISBT) energies in O-polar $ZnO/Zn_{0.60}Mg_{0.40}O$ multiple quantum well structures grown by molecular beam epitaxy on sapphire [2]. The ISBT between the first and the second electronic energy state within the conduction band are observed by infrared spectroscopy at room temperature. Absorption features due to the ISBT occur only for light polarized normal to the quantum well plane (p-polarized) according to the polarization selection rules. By variation of the quantum well width the ISBT energies can be tuned from 290 to 370 meV. The experimental results are in good agreement with theoretical calculations assuming the presence of internal electric fields of 2 MV/cm.

[1] E. Bellotti, K. Driscoll, T. D. Moustakas, and R. Paiella, Journal of Applied Physics **105**, 113103 (2009).

[2] L. Orphal, S. Kalusniak, O. Benson, S. Sadofev, AIP Advances **7**, 115309 (2017).

HL 37.6 Thu 10:45 EW 203

Towards high-performance printed in-plane and vertical MOSFETs — •Felix Neuper, Robert Kruk, Horst Hahn, and Ben Breitung — Institute of Nanotechnology, Karlsruhe Institute of Technology, Hermann-von-Helmholtz-Platz 1, 76344 Eggenstein-Leopoldshafen, Germany

Integrating printed MOSFETs in complex circuits has been successfully demonstrated recently by our group [1] and novel transistor architectures are under development to further improve their performance. As one way to optimize device behaviour a printed vertical porous channel approach has been shown [2], enhancing key parameters such as channel length, on-off-ratio, and high current densities.

As Atomic layer deposition (ALD) is able to coat complex porous structures with high aspect ratios, in this presentation we show how changing from electrolyte gating to ALD-processed dielectric gating in printed MOSFETs vastly increases switching speeds and reduces leakage currents, as well as providing long-term reliability and stability towards environmental influences. Despite the usage of dielectrics, the

devices are fully operational at low voltages (<= 1V) allowing applications such as smart packaging and wearable devices.

[1] Gabriel C M et al.Appl. Phys. Lett. 111, 102103 (2017) [2] Tessy. Baby et al., Advanced Materials, 29 (4), 1603858 (2017)

15 min. break.

HL 37.7 Thu 11:15 EW 203

Influence of parameters on the properties of pulsed radiofrequency magnetron sputtered Ga_2O_3 — •Philipp Schurig, Fabian Michel, Martin Becker, Angelika Polity, and Peter Klar — 1. Physikalisches Insitut und Zentrum für Materialforschung (LaMa), Justus-Liebig-Universität Giessen, Giessen, Deutschland

The advantages of high power impulse magnetron sputtering (HiPIMS) on the deposition of materials is well known, but a combining an impulse method with radio frequency sputtering is not straightforward. With Ga_2O_3 as a transparent oxide with a band gap of about 4.9 eV the influence of the duty cycle or the sputter power on the layer properties of pulsed radio frequency magnetron assisted sputtered films is investigated. The idea behind this transfer is to be able to lower the growth temperature of about 650 °C for good quality Ga_2O_3 by increasing the particle energy. For industrial applications and flexible substrates high substrate temperatures are a drawback. HiPIMS studies [1] have shown that pulsed sputter deposition allows an increase of the coupled power and at the same time a decrease of the growth temperature without severe structural degradation of the material. However, post growth annealing was still necessary and was executed at a temperature of 1000 °C in ambient atmosphere. Optical, crystallographic and compositional analysis was performed after deposition and thermal treatment.

[1] E. Nakamura et al.: Appl. Phys. Lett. 104, (2014) 051121

HL 37.8 Thu 11:30 EW 203

Optical properties of amorphous Zn-Sn-Ti oxides: A combined molecular dynamics and density functional theory study — •Daniel Fritsch — Department of Chemistry, University of Bath, BA2 7AY Bath, UK

Amorphous transparent conductive oxides (TCOs) are widely used in technological applications due to their high transparancies, charge carrier mobilities, and flexibility. Active research is under way to find materials to replace the up-to-now industry standard amorphous In-Ga-Zn oxide (a-IGZO). A promising material combination has been identified within the $(ZnO)_x(SnO_2)_y(TiO_2)_z$ solid-solution. The immense parameter space for fabrication of this system (x:y:z ratio) makes computational screening and insights highly desirable.

Here, we report on our combined ab initio molecular dynamics (MD) and density functional theory (DFT) study of amorphous Zn-Sn-Ti oxide solid-solutions. Covering the whole parameter space (x:y:z ratio) we generated amorphous structures using a well-established melt and quench approach. Based on the resultant structures, structural, electronic, and optical properties are analysed and compared with available experimental and theoretical data. The electronic properties are calculated using a self-consistent hybrid functional, which has been proven to yield improved results for bulk oxide semiconductors [1].

[1] D. Fritsch, B. J. Morgan, and A. Walsh, Nanoscale Research Letters 12, 19 (2017).

HL 37.9 Thu 11:45 EW 203

Interactions of Rydberg Excitons in Cu_2O — •Felix Föst[1], Julian Heckötter[1], Rico Schwartz[2,1], Marc Assmann[1], Dietmar Fröhlich[1], and Manfred Bayer[1] — [1]Experimentelle Physik 2, TU Dortmund, Germany — [2]Institut für Physik, Universität Rostock, Germany

In this work we present absorption measurements on highly excited Rydberg excitons in cuprous oxide with principal quantum numbers n up to 25 and odd angular momentum (p, f,..., in analogy to the hydrogen atom). These excitons have a large orbital extension of up to 2 microns [1], covering over 10^9 unit cells, which leads to a strong dipole moment and therefore a giant dipole-dipole interaction.

We investigate the interaction of these highly excited Rydberg states with other exciton states, e.g. p-excitons with low n or the 1s exciton. To accomplish this, we use 2 CW lasers in a conventional two-colour pump-probe setup to pump different fixed exciton resonances and simultaneously probe the absorption spectrum. With increasing pump laser power density, we find a decrease of absorption efficiency for large quantum numbers n in accordance with theory [1]. In addition, we also see an initial increase in absorption for small n for both p- and f-excitons, which is larger for higher pump energies.

[1] T. Kazimierczuk et al., Nature **514**, (2014), p. 343

HL 37.10 Thu 12:00 EW 203

A Unifying Perspective on Oxygen Vacancies in Wide Band Gap Oxides — •Christopher Linderälv, Anders Lindman, and Paul Erhart — Chalmers University of Technology, Gothenburg, Sweden

Wide band gap oxides are versatile materials with numerous applications in research and technology. Many properties of these materials are intimately related to defects with the most important defect being the oxygen vacancy. Here, using electronic structure calculations, we show that the charge transition level (CTL) and the eigenstates associated with oxygen vacancies, which to a large extent determine their electronic properties, are confined to a rather narrow energy range, even while band gap and the electronic structure of the conduction band vary substantially. The vacancies are classified according to their character (deep and shallow), which shows that the alignment of electronic eigenenergies and CTL can be understood in terms of the transition between cavity-like localized levels in the large band gap limit and strong coupling between conduction band and vacancy states for small to medium band gaps. We consider a semi-local as well as a hybrid functional and, complementing earlier work, demonstrate that the former yields results in very good agreement with the latter provided that band edge alignment is taken into account.

HL 37.11 Thu 12:15 EW 203

Defect induced magnetism and polaronic states in ZnO — Sanjeev K. Nayak[1], •Waheed A. Adeagbo[2], Martin Hoffmann[3], Matthias Geilhufe[4], Hichem Ben Hamed[2], Arthur Ernst[3,5], and Wolfram Hergert[2] — [1]Department of Materials Science & Engineering, University of Connecticut, USA — [2]Institute of Physics, Martin Luther University Halle-Wittenberg, Germany — [3]Institute for Theoretical Physics, Johannes Kepler University Linz, Austria — [4]Nordita, Center for Quantum Materials, KTH Royal Institute of Technology and Stockholm University, Sweden — [5]Max Planck Institute of Microstructure Physics, Halle, Germany

Crucial issue on magnetism in ZnO in scientific literature with transition metal doped ZnO has been to identify the mechanism for magnetic interaction. However, recent reports on ferromagnetism in pure defected ZnO and Li doped ZnO have brought back our attention to examine the electronic structure of doped ZnO more carefully in order to justify the origin of magnetic moment in p-orbital system. Present understanding goes in the line that localization of holes created from some point defects, such, as Zn-vacancy V_{Zn}, is responsible for the magnetic moment. The question about the interpretation of first-principles calculations for such a problem is the interest of the present work. We propose that the hole localization in V_{Zn}, could be achieved by treating the $V_{Zn}-O_4$, as a single complex unit of point defect, where the four O are treated separately through a Hubbard-U correction. The treatment gives identical results as in hybrid-functional treatment and non-local external potential functional and is more intuitive.

HL 37.12 Thu 12:30 EW 203

Identification of switching modes in Y_2O_3 transition metal oxide RRAM devices — •Eszter Piros[1], Stefan Petzold[1], Benjamin Krah[1], Sharath Sankaramangalam Ulhas[1], Tom Blomberg[2], Marko Tuominen[2], Hessel Sprey[3], Christian Wenger[5], Eric Jalaguier[4], Sophie Bernasconi[4], Etienne Nowak[4], Erwin Hildebrandt[1], and Lambert Alff[1] — [1]Institute for Materials Science, Advanced Thin Film Technology, Technische Universität Darmstadt, Alarich-Weiss-Str. 2 D-64287 Darmstadt, Germany — [2]ASM Microchemistry Ltd. Väinö Auerin katu 12 A, 00560 Helsinki, Finland — [3]ASM Belgium NV Kapeldreef 7, 3001 Leuven, Belgium — [4]CEA Leti 17 avenue des Martyrs, 38054 Grenoble, France — [5]IHP Im Technologiepark 25, 15236 Frankfurt (Oder), Germany

Resistive random access memory (RRAM) devices offer a new solution to the current problems of scalable non-volatile data storage. Here we report the stabilized resistive switching modes [1] in yttrium oxide-based RRAM devices. In the bipolar device, stable intermediate resistance states are achieved by varying either the voltage sweep values during reset or by varying the compliance current in the set process. This opens up the possibility of multibit programming and neuromorphic applications. The conduction mechanism of bipolar and unipolar devices was also investigated. The high endurance that exceeds several hundred DC switching cycles and very good data retention (extrapo-

lated to 10 years at 85°C) make both bi- and unipolar devices suitable candidates for next generation non-volatile memory devices. [1] S. U. Sharath, Adv. Funct. Mater. 27, 1700432 (2017)

HL 37.13 Thu 12:45 EW 203

Solution processed hybrid field effect transistors based on graphene electrodes — •Surya Abhishek Singaraju[1], Tessy Theres Baby[1], Jasmin Aghassi-Hagmann[1,2], Horst Hahn[1], and Ben Breitung[1] — [1]Institue of Nanotechnology, Karlsruhe Institute of Technology, Eggenstein-Leopoldshafen 76344, Germany — [2]Department of Electrical Engineering and Information Technology, Offenburg University of Applied Sciences, Offenburg 77652, Germany

In our work we target solution-processable, electrolyte-gated field-effect transistors (EGFETs) based on an In2O3 precursor channel material and graphene for the passive electrodes. The EGFETs were prepared on glass substrates by printing crystalline In2O3 as active material, graphene ink for passive structures and a composite solid polymer electrolyte (LiClO4/PC/PVA/DMSO) as gating compound. Good printing resolution could be achieved by ink-jet printing the In2O3 precursor and the electrolyte and by microplotting the graphene electrodes using an ultrasonic-controlled fluid dispensing microplotter. The chemical properties and the interaction of different transistor components was analyzed using methods like XPS and cyclic voltammetry. The interface between source/drain and the channel plays a major role in the device performance and was investigated regarding the electrical contact. We observed that the devices exhibited relatively high output current values for a fully printed transistor. Also, the on/off ratio from the transfer curve was measured to be in the range of 105-107.