Graduate Student Workshop
on Transparent Conducting Oxide Semiconductors

Venue:

Humboldt-Universität zu Berlin

IRIS Building, Room 021

17-18 November 2015

Program

17. Nov (Tue)			**18. Nov** (Wed)		
Start	*End*	*Speaker*	*Start*	*End*	*Speaker*
09:30	10:00	Meet & Greet	09:00	09:15	Meet & Greet
10:00	10:15	Introduction	09:15	10:00	**Cocchi**
10:15	11:00	**Janowitz**	10:00	10:45	**Fiedler**
11:00	11:15	Coffee Break	10:45	11:00	Coffe Break
11:15	12:00	**Nazarzahdemoafi**	11:00	11:45	**Rombach**
12:00	12:30	**Müller**	11:45	12:15	**Siebert**
12:30	14:00	Lunch Break	12:15	14:00	Lunch Break
14:00	14:45	**Schewski**	14:00	14:45	**Splith**
14:45	15:30	**Schlupp**	14:45	15:30	**Handwerg**
15:30	15:45	Coffee Break	15:30	16:00	**Thielert**
15:45	16:30	**Neumann**	16:00	16:15	Closing remarks

Post-Doc
PhD Student
MSc/BSc

Welcome!

This workshop on Transparent Conducting Oxide Semiconductors intends to provide an opportunity for networking connections over group and university borders within the oxide semiconductor community. Its main focus is to open a discussion based on the knowledge, methodology, but also problems or questions of young researchers. Ideally, new collaborations and crosslinks will further strengthen the cooperation among different groups.
We hope some of these goals will be reached and are looking forward to an interesting scientific exchange.

We thank Prof. Dr. Saskia Fischer of HU Berlin for initiating this workshop.

To facilitate exchange, a list of (already known) participants is reproduced at the end in alphabetical order.

Fariba Hatami, Martin Feneberg, Mattia Mulazzi

Tue, 10:15-11:00

Electronic structure of the TCO's In_2O_3 and Ga_2O_3

Christoph Janowitz

Humboldt University Berlin

Transparent conducting oxides encompass a growing number of oxides, which combine transparency for visible light with good electrical conductivity. Although no consensus on the doping mechanism is reached, depending on the doping level these materials are often regarded as degenerate semiconductors. In this contribution the electronic structure of In_2O_3 and Ga_2O_3 is discussed with regard to common trends. ARPES on cleaved single crystals is especially used to investigate the states in the vicinity of E_F. The results disagree with the charge accumulation layer model and suggest, that electron phonon coupling leading to polaron states may be an essential ingredient to TCO physics.

Tue, 11:15-12:00

Peculiar electronic properties of metal-In_2O_3 interfaces

M. Nazarzadehmoafi,[1] A. Siebert, [1] F. Titze,[1] C. Janowitz,[1] M. Mulazzi,[1,2] Z.Galazka,[2] and R. Manzke[1]

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In_2O_3 is a promising material to be used like a true semiconductor in transparent electronic devices. To this aim, high quality single crystals with controllable surface and interface properties are of great demand. To shed light on the microscopic origin of the conduction in In_2O_3, the behavior of the electronic structure of as-cleaved melt-grown In_2O_3 (111) single crystals was studied upon noble metals, indium and tin deposition using angle-resolved photoemission spectroscopy. A small Schottky barrier was observed in Ag and Au/In_2O_3 with the contact prepared at room temperature and a larger one in Au/In_2O_3 with the contact prepared at low temperature. From these results, it can be deduced that there is no surface electron accumulation layer at as-cleaved surface of the studied crystals. In contrast to the aforementioned noble metals upon deposition of Cu, the interface shows ohmic character. Moreover, in the thick Au and Cu overlayer on In_2O_3 at room temperature, Shockley-like surface states were observed, which in the case of Au, as opposed to Cu, it shares many similarities with its single crystal. This indicates the uniform growth of this metal on In_2O_3 at this temperature. In and Sn evaporation led to the downward band bending, which was larger in the case of metallic tin deposition. The initial stage of Cu and In growth on In_2O_3 was accompanied by formation of a two dimensional electron gas (2DEG) which fades away for higher coverages. These states are not associated with the earlier-detected 2DEG at the pristine surface of In_2O_3 thin films. In spite of similar electronic properties of In and Sn, no 2D state appears by Sn deposition. By measuring work functions of In_2O_3 and the studied metals *in situ* and applying the Schottky-Mott rule, except Ag/In_2O_3, we identified a strong disagreement between the predicted barriers from this model and experimentally-derived ones. These discrepancies could be explained by the presence of the 2DEG in cu and In/In_2O_3, but not for the other presented interfaces.

Tue, 12:00-12:30

Properties of heteroepitaxial grown Si-doped $\beta - Ga_2O_3$ thin films and Schottky contacts thereon

Stefan Müller[*1], Holger von Wenckstern[1], Daniel Splith[1], Heiko Frenzel[1], Florian Schmidt[1], Marius Grundmann[1]

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We report on the structural and electrical properties of Si-doped $\beta - Ga_2O_3$ thin films grown by pulsed laser deposition on c-plane sapphire substrates. The oxygen partial pressure ($p(O_2)$) was systematically varied between 3×10^{-4} and 0.024 mbar and the substrate temperature (T_G) between 400°C and 650°C. On thin films with tailored properties ($p(O_2)$ = 1×10^{-3} mbar and T_G=650°C) we fabricated Pt/PtO_x Schottky contacts (SCs) using different fabrication techniques like thermal evaporation, sputtering and long-throw sputtering. Our investigations show that the barrier formation of the gallium oxide based SCs is highly dependent on the kinetic energy of the incident metal particles. The best SCs have larger rectification ratios of 7-8 orders of magnitude, small ideality factors of about 1.2 and large barrier heights of nearly 1.5 eV.

Tue, 14:00-14:45

Group III Sesquioxides and its solid solutions studied by Transmission Electron Microscopy

R. Schewski[1], M. Albrecht[1], T. Schulz[1], G.Wagner[1], M.Baldini[1], D.Gorgova[1], Z. Glazaka[1], R.Uecker[1], H. von Wenckstern[2], O.Bierwagen[3], Patrick Vogt[3]

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In this talk we present a study by transmission electron microscopy on the growth of Ga_2O_3 and its alloys $(In_xGa_{1-x})_2O_3$. We show that the defect distribution of homoepitaxially grown Ga_2O_3 is governed by surface diffusion and two-dimensional nucleation. Layers grown on (100) substrates may suffer from a high density of planar defect with strong impact on physical properties. The crystallography, influence on the electrical properties and formation mechanism of planar defcets will be discussed.

Though alloying Ga_2O_3 with In_2O_3 offers the opportunity for band gap engineering, it is challenging since In_2O_3 and Ga_2O_3 crystallize in different phases at thermodynamical equilibrium. We show that up to 50% of Indium can be incoporated into the monoclinic phase. Beyond this phase separation into a monoclinc a hexagonal and a cubic $(InGa)_2O_3$ phase is observed. We will discuss the phase separation and solubility limits, analyse the site occupancy by Ga and In and compare our results with recently published theoretical predictions.

Room temperature fabricated unipolar and bipolar diodes comprising amorphous zinc tin oxide

Peter Schlupp, Sofie Bitter, Friedrich-Leonhard Schein, Holger von Wenckstern and Marius Grundmann
Inst. f. Exp. Physik II, Universität Leipzig, Germany

The application of amorphous oxide semiconductors (AOS) within low cost electronics is favourable because they can be fabricated at room-temperature. Additionally, this allows the usage of plastic substrates for flexible devices. Zinc tin oxide (ZTO), consisting of highly abundant materials only, is a promising n-type AOS. High electron mobilities in the order of $10\,\mathrm{cm}^2/\mathrm{Vs}$ are reported [1] rendering ZTO interesting as channel material for pixel drivers for active matrix displays. The semiconducting ZTO thin films were fabricated by pulsed laser deposition at RT. The influence of the Zn/Sn ratio on material properties will be presented. Unipolar and all-amorphous bipolar diodes were fabricated using ZTO as n-type semiconductor [2]. As substrates rigid glass and flexible polyimid were used. Diode porperties were investigated and the influence of bending on the properties will be discussed.

References

[1] Jayaraj, M. K., Saji, K. J., Nomura, K., Kamiya, T., and Hosono, H. *Journal of Vacuum Science & Technology B: Microelectronics and Nanometer Structures* **26**(2), 495 (2008).

[2] Schlupp, P., Schein, F.-L., Wenckstern, H. v., and Grundmann, M. *Advanced Electronic Materials* **1**(1-2), 1400023 (2015).

Tue, 15:45-16:30

Influence of the electron-hole interaction on the dielectric function of ZnO, MgO, and hexagonal MgZnO

M.D. Neumann

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ZnO and its ternary alloy (Zn,Mg)O are considered promising candidates for optoelectronic devices operating in the violet and ultraviolet regime. Therefore, the precise knowledge of their optical properties is mandatory. They are characterized by the dielectric function.
This talk addresses the influence of electron-hole interaction on the dielectric function of ZnO, MgO and hexagonal MgZnO in the spectral range from 2 to 20 eV. It is demonstrated, that excitonic effects must be considered in the whole spectral range, in order to correlate peculiarities in the dielectric function with transitions at critical points of the band structure. Around the fundamental bandgap excitons and their coupling to the crystal lattice are discussed in detail. Their impact on the determination of characteristic material parameters is shown. For hexagonal MgZnO the compositional dependence of the valence band ordering and its influence on the polarization properties of optoelectronic devices is presented.

Wed, 9:15-10:00

X-ray spectroscopy of Ga_2O_3 from many-body perturbation theory

Caterina Cocchi* Dmitrii Nabok, and Claudia Draxl

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Group III sesquioxides are appealing materials for novel applications in optoelectronics. The properties of these systems are rather sensitive to the presence of defects or doping. Hence, a thorough characterization of the samples is mandatory. X-ray spectroscopy represents a powerful technique in this regard. A synergistic interplay between experiments and theory can provide a deep insight into the chemical composition and the structure-property relationship of these complex materials. Here, we present an in-depth *ab initio* study of the X-ray absorption properties of Ga2O3 from many-body perturbation theory, as implemented in the all-electron full-potential code **exciting** [1]. The absorption spectrum from the oxygen (O) *K* edge of the bulk material reveals strong excitonic effects in the near-edge region. For specific surface orientations, taking into account the anisotropy of the system [2], the contributions of the inequivalent O atoms present different spectra weights. Our results are in agreement with recent experiments [3].

References:

[1] A. Gulans *et al.*, J. Phys.: Condens. Matter **26**, 363202 (2014).

[2] C. Vorwerk, C. Cocchi, and C. Draxl, arXiv:1510.03306, s*ubmitted* (2015).

[3] H. Zschiesche *et al., unpublished.*

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Wed, 10:00-10:45

Electrical and optical characterization of homoepitaxial β-Ga_2O_3 layers grown by metal organic vapor epitaxy

A.Fiedler[1], K.Irmscher[1], R.Schewski[1], M.Albrecht[1], M.Baldini[1], G.Wagner[1]

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Status: PhD student

Sn and Si doped β-Ga_2O_3 homoepitaxial layers have been grown on (100) β-Ga_2O_3 substrates by metal organic vapor phase epitaxie(MOVPE). The samples are electrically characterized by temperature-dependent conductivity in Van der Pauw configuration and Hall effect measurements, which give information about resistivity, mobility, charge carrier concentration and scattering processes. Doping profiles are recorded by capacitance-voltage measurements(C-V). Concentration, ionization energy and capture cross section of defect states are measured using deep level transient spectroscopy (DLTS). Structural analysis is done using transmission electron microscopy (TEM). Growing with MOVPE on (100) β-Ga_2O_3 substrates results in layers containing incoherent grain boundaries, which form dislocation walls. The consequences on the electrical properties are discussed. Free charge carrier density is limited between $5 \times 10^{17}\,\mathrm{cm}^{-3}$ and $2 \times 10^{19}\,\mathrm{cm}^{-3}$ due to the compensation by dangling bonds in the dislocation walls, among others. Also the charge carrier mobility at room temperature is lowered from $\mu_{\mathrm{bulk}} \approx 120\,\mathrm{cm}^2\mathrm{V}^{-1}\mathrm{s}^{-1}$ to $\mu_{\mathrm{layer}} \approx 30\,\mathrm{cm}^2\mathrm{V}^{-1}\mathrm{s}^{-1}$ due to this dislocation walls.

Electrical conductivity and gas-sensing properties of single-crystalline In_2O_3 thin films: bulk vs. surface

J. Rombach[1], O. Bierwagen[1], M. Mischo[2], V. Cimalla[2], T. Berthold[3], S. Krischok[3], M. Himmerlich[3]

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3. Institut für Mikro- und Nanotechnologien, Technische Universität Ilmenau, Germany

This study aims to provide a better fundamental understanding of the gas-sensing mechanism of In_2O_3-based conductometric gas sensors. In contrast to typically used polycrystalline films, we study single crystalline In_2O_3 thin films grown by molecular beam epitaxy (MBE) as a model system with reduced complexity. Electrical conductivity of these films essentially consists of two parallel contributions: the bulk of the film and the surface electron accumulation layer (SEAL). Both these contributions are varied to understand their effect on the sensor response. Conductivity changes induced by UV illumination in air, which forces desorption of oxygen adatoms on the surface, give a measure of the sensor response and show that the sensor effect is only due to the SEAL conductivity. Therefore, a strong sensitivity increase can be expected by reducing or eliminating the bulk (or intra-grain) conductivity. Gas-response measurements in ozone atmosphere test this approach for the real application.

Wed, 11:45-12:15

STM investigations of in situ cleaved In_2O_3(111) and β-Ga_2O_3(100)

Andreas Siebert, Mattia Mulazzi
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In this contribution I present our results on the structural and electronic properties of In_2O_3(111) and β-Ga_2O_3(100) measured with scanning tunnel microscopy. The measurements have been taken on UHV-cleaved single-crystals provided by the group of Dr. Z. Galazka of the IKZ. All the measurements have been taken in constant current mode with currents of ~0.5 nA and bias voltages ranging from -1.5 V to +1.5 V.
On both materials we found terraces as wide as 500 nm with roughness attributable to step edges and measure the crystal unit cell, but were unable to resolve the atomic structure. At mesoscopic lengthscales we detected point defects appearing as dips of about ~100 pm depth, much smaller than any structural feature.

Rectifying contacts on In_2O_3 thin films

Daniel Splith, Florian Schmidt, Steffen Lanzinger, Stefan Müller, Holger von Wenckstern and Marius Grundmann

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While the electrical properties of highly conducting, tin-doped In_2O_3 are well investigated, interest in the semiconducting properties of In_2O_3 arose only recently. The realization of rectifying contacts on In_2O_3 is crucial for the electrical characterisation by space-charge region based measurements like thermal admittance spectroscopy (TAS) or deep-level transient spectroscopy (DLTS). Further, such contacts are the key elements of electronic devices.

The realization of rectifying contacts on In_2O_3 is challenging due to the formation of a surface electron accumulation layer (SEAL) [1]. The formation of a SEAL was also reported for ZnO, where two approaches for the fabrication of rectifying contacts succeeded: On the one hand, a reactive sputtering process was capable of removing the SEAL on ZnO [2]. On the other hand, amorphous *p*-type oxide semiconductors like $ZnCo_2O_4$ or NiO form *pn*-heterojunctions on ZnO [3].

We employed both of these techniques to successfully fabricate rectifying contacts on In_2O_3 thin films. The electrical properties of these contacts will be discussed. Further, the contacts were used for first TAS and DLTS measurements on semiconducting In_2O_3 thin films.

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[2] A. Lajn *et al.*, J. Vac. Sci. Technol. B 27, 1769-1773 (2009)
[3] M. Grundmann *et al.*, Adv. Sci. Technol., 93: 252-259 (2014)

Wed, 14:45-15:30

Temperature-dependent thermal properties of Mg-doped insulating β-Ga_2O_3 bulk along [100] and [001]: thermal diffusivity, thermal conductivity and specific heat capacity.

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The thermal conductivity λ, diffusivity D, specific heat capacity C_V and the Debye temperature θ_D have been investigated for insulating monoclinic β-Ga_2O_3.
Here, we apply AC current heating techniques and determine the thermal diffusivity and specific heat capacity along the [100] and [001] directions from the so-called 3ω and 2ω voltage signals, respectively.
At low temperatures we find an anisotropy of the thermal conductivity for the [100] and [001] directions ($\lambda_{[100]}$(109K) = 54 ± 5 $Wm^{-1}K^{-1}$; $\lambda_{[100]}$(109K) = 67 ± 5 $Wm^{-1}K^{-1}$).

The anisotropy diminishes for increasing temperature which is reflected by the diffusivity ratio of $\frac{D_{[001]}}{D_{[100]}} = 1.40 \pm 0.08$ at 109 K and $\frac{D_{[001]}}{D_{[100]}} = 1.05 \pm 0.07$ at 197 K.
This may find its explanation in a decreased phonon mean free path for higher temperatures. This is confirmed by the temperature dependence of the thermal conductivity which agrees well with the phonon-phonon Umklapp scattering processes.
The temperature dependence of the specific heat is in general accordance with the Debye model.

Wed, 15:30-16:00

Noise spectroscopy on In_2O_3 films

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How good is the quality of In_2O_3 epitaxial layers prepared in different condition? In order to answer this question, several In_2O_3 samples were investigated by means of low-frequency noise spectroscopy and hall measurements. The samples were grown at the Paul-Drude-Institut using molecular-beam epitaxy with different doping; unintentionally doped, tin doped and magnesium-doped. After growth the samples were thermally annealed in vacuum and in oxygen environment.

Our results show that the samples annealed in the vacuum have 1/f noise with higher magnitude compared to the samples annealed in the oxygen environment. The estimated Hooge-factor [1] was between 10^{-2} and 10^{2} depending on the type of the doping and the annealing conditions. The Mg-doped annealed in oxygen environment has the lowest Hooge-factor of 10^{-2}, and hence the best quality. The value is more than one order lower than the reported value for the amorphous In_2O_3 [2]. The Sn-doped sample annealed in vacuum, on the other hand, has the highest Hooge-factor.

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