Helmholtz Virtual Institute for
**Mem**ory Effects in **R**esistive
**I**on-beam Modified **Ox**ides

HELMHOLTZ ZENTRUM DRESDEN ROSSENDORF
MEMRIOX

# MEMRIOX International Workshop 2016

25 – 27 September 2016

Berghotel Bastei, Germany

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HELMHOLTZ ZENTRUM DRESDEN ROSSENDORF
MEMRIOX

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# Venue and organization

## Venue

Berghotel Bastei
01847 Lohmen, Germany
Phone: +49 35024 779-0, Fax. -481
E-mail: info@bastei-berghotel.de.de
Homepage: www.bastei-berghotel.de

## Organization

Helmholtz Virtual Institute MEMRIOX
Helmholtz-Zentrum Dresden-Rossendorf
Bautzner Landstraße 400, 01328 Dresden, Germany
Phone: +49 351 260 3121
E-mail: memriox-workshop@hzdr.de
Homepage: www.hzdr.de/memriox

### Scientific Organizers

Speaker: Prof. Dr. Sibylle Gemming
Institute of Ion Beam Physics and
Materials Research
Helmholtz-Zentrum Dresden-Rossendorf
01328 Dresden, Germany
Phone: +49 351 260 2470
E-mail: s.gemming@hzdr.de

Deputy: PD Dr. Heidemarie Schmidt
Chair of Materials for Nanoelectronics
Chemnitz University of Technology
09107 Chemnitz, Germany
Phone: +49 371 531 32481
E-mail:
heidemarie.schmidt@etit.tu-chemnitz.de

### Local Organizer

PD Dr. Peter Zahn
Helmholtz-Zentrum Dresden-Rossendorf
Institute of Ion Beam Physics and Materials Research
Bautzner Landstraße 400, 01328 Dresden, Germany
Phone: +49 351 260 3121
E-mail: p.zahn@hzdr.de

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**MEMRIOX International Workshop 2016**

HELMHOLTZ ZENTRUM DRESDEN ROSSENDORF
MEMRIOX

**Agenda**

Place: Berghotel Bastei, 01847 Lohmen
updated: 21.09.2016 (PZ)

**Sunday, 25 September 2016**

| Start | Who | Durat. | Short Title (Full titles see next page) | Notes |
|---|---|---|---|---|
| 10:30 / 09:45 | | | Transfer from **Pirna** / Hiking from **Rathen** | |
| 12:00 | | | Arrival/Lunch Buffet | |
| **13:30** | S. Gemming | | **Welcome address** | Chair: C. Ronning |
| 13:40 | M. Kats | 40+10 | Tunable mid-infrared photonics with phase-transition materials | |
| 14:30 | S. Slesazeck | 30+10 | The negative aspects at the origin of hysteresis effects in FeFETs and Nb switches | |
| 15:15 | | 30 | Break | |
| 15:45 | R. Waser | 40+10 | Redox-based memristive switching in metal oxides | Chair: B. Abendroth |
| 16:40 | H. Schmidt | 30+10 | Analog resistive oxide switches for neuromorphic computing and fuzzy logics | |
| 17:30 | | | Individual discussions, e.g. follow-up projects | |
| 19:00 | | | Dinner | |

**Monday, 26 September 2016**

| | | | | |
|---|---|---|---|---|
| 6:30 | | | Sunrise Walk (Ferdinandstein, 10 min., sunrise at 6:56) individual | |
| **8:45** | M. DiVentra | 40+10 | Memcomputing: a brain-inspired topological computing paradigm | Chair: H. Schmidt |
| 9:35 | T. You | 30+10 | Non-volatile resistive switching in $BiFeO_3$-based thin films + application | |
| 10:15 | | 30 | Break | |
| 10:45 | Y.-H. Chu | 40+10 | Controllable Conduction at Complex Oxide Interfaces | Chair: S. Gemming |
| 11:35 | S. Slesazeck (2) | 30+10 | The diversity of $Nb_2O_5$-based Memristors | |
| 12:30 | | | Lunch | |
| 14:00 | | 60 | **Poster session and Coffee** | 11 Posters |
| 15:00 | | | Break | |
| 15:30 | A. McLeod | 40+10 | Insulator-metal phase coexistence probed by cryogenic near-field microscopy | |
| 16:20 | J. Rensberg | 30+10 | In-situ electrical characterization of ion beam irradiated $VO_2$ structures | |
| 17:15 | | | **Sunset Walk** (Bastei Bridge/ Neurathen castle, sunset at 18:53) indiv. | |
| 19:30 | | | Dinner | |

**Tuesday, 27 September 2016**

| | | | | |
|---|---|---|---|---|
| 8:30 | | | **Check-Out** (latest 10:30) | |
| **8:45** | B. Abendroth | 30+10 | Electronic transport properties of ALD thin films $TiO_2$-$SrTiO_3$ | Chair: H. Stöcker |
| 9:25 | S. Krüger | 20+10 | Technology Transfer, Validation and Exploitation | |
| 10:00 | | | Break | Chair: P. Zahn |
| 10:30 | M. Wuttig | 40+10 | Novel Phase Change Materials by Design: The Mystery of Resonance Bonding | |
| 11:25 | S. Gemming | 30+10 | MEMRIOX: 5$^{th}$ anniversary | |
| 12:05 | S. Gemming | 20 | Final Discussion & Closing Remarks | |
| 12:30 | | | Lunch | |
| 14:00 | | | Departure | |

Breakfast will be served from 7:00.

## Invited Talks

## Contributed Talks



## Poster Contributions

see list on page 18

# Controllable Conduction at Complex Oxide Interfaces

Ying-Hao Chu[1,2,3]

[1] Department of Materials Science & Engineering and Electrophysics, National Chiao Tung University, Hsinchu, Taiwan
[2] Institute of Physics, Academia Sinica, Taipei, Taiwan
[3] Material and Chemical Research Laboratories, Industrial Technology Research Institute, Hsinchu, Taiwan

Corresponding author: yhc@nctu.edu.tw

Complex oxide interfaces emerge as one of the most exciting subjects among condensed matters due to their unique physical properties and new possibilities for next-generation electronic devices. Three types of complex oxide interfaces have been established. Among them, the most explored interface is the artificially constructed heterointerfaces. Various interactions at the interface have resulted in a number of exciting discoveries, such as highly mobile quasi-two dimensional electron gas (2DEG) forming between two band insulators ($LaAlO_3$ and $SrTiO_3$). Moreover, in ferroic oxides, domain walls dictate natural homo-interfaces as a consequence of the minimization of electrostatic and elastic energies. Several key studies have pointed out interesting observations, such as local conduction, on domain walls in multiferroics. Recently, new tubular oxide interface has been developed in the self-assembled heterostructures and the local conduction at the tubular interfaces of $BiFeO_3$ (BFO)-$CoFe_2O_4$ (CFO) heterostructure was found. Such results create a huge playground to explore and design intriguing properties of complex oxide interfaces. However, in the push for practical applications, it is desirable to have the control of the interface functionalities through external stimulus. In this presentation, a nonvolatile modulation of the local conduction at the homo- (BFO domain walls) and heterointerfaces ($LaAlO_3$/$SrTiO_3$) will be demonstrated. By inserting a ferroelectric layer, the polarization can tune and modulate the conduction at the $LaAlO_3$/$SrTiO_3$ heterointerface. The XPS and XSTM/S results reveal the electrostatic predictions of the conducting state modulation, demonstrating the possibility of non-volatile control. In addition, we went back to the BFO-CFO tubular interfaces and showed the interface conduction can be modulated non-volatilely and reversibly via an external electric field by using conducting AFM. A memristive-like electronic conduction was observed, that is strongly correlated to the motion of oxygen vacancies (donor impurities) at the interface and in turn modifies the junction characteristics between the measurement tip and the interface. Our results complete the control of the conduction at complex oxide interfaces and suggest the possibility for new devices based on complex oxide interfaces.

**Massimiliano Di Ventra**

*Department of Physics, University of California San Diego, La Jolla, CA 92093-0319 USA*

## Memcomputing: a brain-inspired topological computing paradigm

Which features make the brain such a powerful and energy-efficient computing machine? Can we reproduce them in the solid state, and if so, what type of computing paradigm would we obtain? I will show that a machine that uses memory to both process and store information, like our brain, and is endowed with intrinsic parallelism and information overhead - namely takes advantage, via its collective state, of the network topology related to the problem - has a computational power far beyond our standard digital computers [1]. We have named this novel computing paradigm "memcomputing" [2]. As an example, I will show the polynomial-time solution of prime factorization and the NP-hard version of the subset-sum problem using polynomial resources and self-organizing logic gates, namely gates that self-organize to satisfy their logical proposition [3]. I will also show that these machines are described by a Witten-type topological field theory and they compute via an instantonic phase where a transient long-range order develops due to the effective breakdown of topological supersymmetry [4]. The digital memcomputing machines that we propose are *scalable* and can be easily realized with available nanotechnology components, and may help reveal aspects of computation of the brain.

[1] F. L. Traversa and M. Di Ventra, **Universal Memcomputing Machines**, *IEEE Transactions on Neural Networks and Learning Systems*, 26, 2702 (2015).

[2] M. Di Ventra and Y.V. Pershin, **Computing: the Parallel Approach,** *Nature Physics*, **9**, 200 (2013).

[3] F. L. Traversa and M. Di Ventra, **Polynomial-time solution of prime factorization and NP-hard problems with digital memcomputing machines,** *arXiv:1512.05064.*

[4] M. Di Ventra, F. L. Traversa and I.V. Ovchinnikov, **Topological field theory and computing with instantons** (in preparation).

# Tunable mid-infrared photonics with phase-transition materials

Mikhail A. Kats[1]

[1]Departments of Electrical and Computer Engineering, Materials Science and Engineering, and Physics, University of Wisconsin – Madison, Madison, WI, USA

Corresponding author: mkats@wisc.edu

This talk will review our efforts to utilize phase-transition materials in mid-infrared optics and highlight relevant related literature. Materials like vanadium dioxide ($VO_2$) and rare-earth nickelates undergo structural and electronic phase transitions that can be driven via thermal, electrical, and optical means. These phase transitions result in dramatic changes in electronic properties, and correspond to large changes in optical properties, especially in the mid-infrared spectral range where the Drude response dominates. Previously we have demonstrated that $VO_2$ integrated into thin film structures enable temperature- and current-tunable absorbers and reflectors and plasmonic antennas, as well as thermal emitters with anomalous temperature dependence.

More recently, we looked to expand the palette of tunable optical materials by defect engineering $VO_2$ to alter its properties, and by using less-studied phase-transition materials such as samarium nickelate (SNO). By combining these approaches, thermally driven phase transitions can be found over a range of critical temperatures from below room temperature to above 100 °C. We are currently demonstrating anomalous thermal emitters using SNO, which can mask the temperatures of objects from thermal imagers, and $VO_2$-based metasurfaces with new functionalities using nanoscale defect engineering with ion irradiation.

Many thanks to the contributions of numerous students, colleagues, and collaborators, including Chenghao Wan, Patrick Roney, Jad Salman, Alireza Shahsafi, Jura Rensberg, Carsten Ronning, You Zhou, Zhen Zhang, Shriram Ramanathan, Shuyan Zhang, and Federico Capasso.

# Cryogenic nano-optical imaging of the insulator-metal transition in correlated oxides

A. S. McLeod[1*], E. van Heumen[2], J. Zhang[1], K. W. Post[1], J. G. Ramirez[1], S. Wang[1], T. Saerbeck[1],S. Guenon[1], M. Goldflam[1], M. K. Liu[1], M. Hepting[3],W. B. Wu[4], R. Averitt[1], B. Keimer[3], I. K. Schuller[1], and D. N. Basov[1]

[1] *University of California San Diego, Dept. of Physics, 9500 Gilman Dr., La Jolla, CA, USA*
[2] *University of Amsterdam, Spui 21, 1012 WX Amsterdam, Netherlands*
[3] *Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, 70569 Stuttgart, Germany*
[4] *University of Science and Technology of China, Hefei, Anhui, 230026, P.R. China*

[*] *asmcleod@ucsd.edu*

While insulator-metal transitions remain among the most studied phenomena in correlated electron physics, their interplay with spatially ordered phase coexistence at the meso- and nano-scales remains poorly understood in realistic materials. Enabled by cryogenic near-field infrared microscopy, we reveal real-space evolution of the low temperature insulator-metal transitions in thin films of the correlated transition metal oxides: $V_2O_3$, $NdNiO_3$, and $La_{2/3}Ca_{1/3}MnO_3$. The canonical Mott transition in $V_2O_3$ is observed to proceed through two real-space textures: First, spontaneous formation of insulator-metal stripes is attributed to accommodation strain from an underlying structural transition, whereas a distinct “droplet” nano-texture dominates at lower temperatures, consistent with the role of long-range Coulomb interactions. Comparison with bulk electronic and structural probes reveals an associated thermal separation of structural and Mott transitions in this epitaxial compound. Such droplet nano-texture again manifests in $NdNiO_3$ upon a first-order transition into a low-temperature charge-transfer insulator state. Nucleation and growth of the high-temperature metallic state is then clearly resolved emerging from both defects/impurities and evident magnetic domain walls. Lastly, in the canonical magnetoresistive oxide $La_{2/3}Ca_{1/3}MnO_3$ we demonstrate that low-temperature metallic ferromagnetism can be inhibited through epitaxial strain, then reversibly restored on command by ultrafast photo-excitation. Through nano-scale imaging, we reveal the process of 1) photo-patterning and 2) reconfigurably erasing the magnetic metal, as mediated through unforeseen magneto-elastic coupling. Our studies demonstrate the unmatched utility of cryogenic nano-optical imaging for unraveling the mysteries of insulator-metal transitions among correlated oxides.

# Fundamental ingredients of redox-based memristive switching in metal oxides

Rainer Waser, Felix Gunkel, Regina Dittmann, Vikas Rana, Dirk Wouters,
Ilia Valov, and Stephan Menzel

*JARA-FIT & PGI-7, Forschungszentrum Jülich, 52425 Jülich, and*
*IWE2, RWTH Aachen University, 52056 Aachen, Germany*

Redox-Based Resistive Switching Memories (ReRAM), also called *nanoionic* memories or *memristive* elements, are widely considered to provide a potential leap beyond the limits of Flash (with respect to write speed, write energies) and DRAM (with respect to scalability, retention times) as well as energy-efficient approaches to neuromorphic concepts.

In this seminar talk, fundamental aspects of the physics and chemistry (lattice disorder, ionic and electronic transport processes, and phase formation) of these elements will be presented. In particular, the ultra-high non-linearity of the switching kinetics of redox-based resistive switching devices will be discussed with an emphasis on the so-called valence change mechanism (VCM) typically encountered as a bipolar switching in metal oxides. The involved electrochemical and physical processes can be either electric field/voltage enhanced or accelerated by a local increase in temperature due to Joule heating. The analysis of the published SET switching kinetics data of VCM-type ReRAM systems showed that their nonlinearity is mainly dominated by temperature-accelerated ion hopping, controlled by the local power during the switching process. The gradual RESET transition can be explained in terms of temperature-accelerated ion movement with counter-acting ion drift and diffusion processes. It will be shown that a designated combination of oxides can significantly improve the long-term kinetics, i.e. the retention time, by tailoring the ion diffusion properties in the oxide layers.

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# Novel Phase Change Materials by Design: The Mystery of Resonance Bonding

M. Wuttig
RWTH Aachen University of Technology, Germany

Phase change media utilize a remarkable property portfolio including the ability to rapidly switch between the amorphous and crystalline state, which differ significantly in their properties. This material combination makes them very attractive for data storage applications in rewriteable optical data storage, where the pronounced difference of optical properties between the amorphous and crystalline state is used. This unconventional class of materials is also the basis of a storage concept to replace flash memory. This talk will discuss the unique material properties, which characterize phase change materials. In particular, it will be shown that only a rather small group of materials utilizes resonance bonding, a particular flavour of covalent bonding, which can explain many of the characteristic features of phase change materials. This insight is employed to predict systematic property trends and to explore the limits in stoichiometry for such memory applications. It will be demonstrated how this concept can be used to tailor the electrical and thermal conductivity of phase change materials. Yet, the discoveries presented here also force us to revisit the concept of resonance bonding and bring back a history of vivid scientific disputes about 'the nature of the chemical bond'.

# Effect of the microstructure characteristics on the electronic transport properties of ALD thin films in the system $TiO_2$-$SrTiO_3$

Solveig Putzschke[1], Wolfram Münchgesang[1], Juliane Weise[1], Jura Rensberg[2], Carsten Ronning[2], Hartmut Stöcker[1], Barbara Abendroth[1], Dirk C. Meyer[1]

[1] Institut für Experimentelle Physik, TU Bergakademie Freiberg, Germany

[2] Institute for Solid State Physics, Friedrich-Schiller-Universität Jena, Jena, Germany

Corresponding author: Barbara.abendroth@physik.tu-freiberg.de

Transition metal oxide thin films with compositions ranging from $SrTiO_3$ to $TiO_2$ constitute a model system to study the effects of microstructure characteristics on optic and electric properties of the dielectric. In particular, this work focused on the electronic transport and resistance changes in asymmetric metal insulator metal (MIM) stacks with the dielectric embedded between a non-reactive ohmic TiN bottom electrode and a noble Au top electrode. Modification of the dielectric microstructure is achieved on the one hand by means of thin film deposition conditions and on the other hand by noble gas ion implantation.

Atomic layer deposition (ALD) has been used to deposit the dielectric thin films. Since ALD allows for independent control of the cation metal organic precursors, ternary $Sr_xTi_yO_z$ can be grown far off the thermodynamic equilibrium composition of $SrTiO_3$. ALD of ternary $Sr_xTi_yO_z$ at temperatures up to 320 °C always results in amorphous films of low leakage currents. Based on the cation stoichiometry, band gap, dielectric constant and barrier heights can be tuned between the values of $TiO_2$ and SrO. For binary $TiO_2$ the ALD temperature controls the formation of crystalline phases during growth. The presence of crystalline grains and grain boundaries affects leakage current as well as resistive switching of such layers.

Ion implantation introduces point defects such as vacancies, dangling bonds or a deviation from ideal cation coordination. Implantation parameters have been chosen to prevent preferential oxygen removal in near-surface regions as well as interface mixing with the bottom electrode. Hence the oxide stoichiometry is maintained and the effect of point defects on electronic transport properties is investigated independently from stoichiometry as a function of implantation dose.

The presentation will summarize the results accomplished during the VI Memriox PhD project of Solveig Putzschke.

# Resistive oxides and the role of defects or the 5th anniversary of MEMRIOX

S. Gemming[1,2], H. Schmidt[2], T. Mikolajick[3], D.C. Meyer[4], R. Waser[5], R. Dittmann[5], C. Ronning[6], N. Spaldin[7], and D. Basov[8]

[1] Helmholtz-Zentrum Dresden-Rossendorf, D-01314 Dresden, Germany,
[2] Technische Universität Chemnitz, D-09107 Chemnitz, Germany,
[3] Technische Universität Dresden, D-01062 Dresden, Germany,
[4] Technische Universität Bergakademie Freiberg, D-09596 Freiberg, Germany,
[5] Forschungszentrum Jülich, D-52425 Jülich, Germany,
[6] Friedrich-Schiller-Universität Jena, D-07743 Jena, Germany,
[7] ETH Zürich, CH-8092 Zürich, Switzerland,
[8] University of California, San Diego, La Jolla CA 92093-0354, U.S.A.

Corresponding author: s.gemming@hzdr.de

Transition metal oxides exhibit a wealth of physical phenomena, among them ferroic properties such as ferroelasticity, ferroelectricity and ferromagnetism, or their combination in multiferroics. Ideal multiferroics may relax back to their field-free ground state, once the external field is removed. Thus, any information encoded by the field as local change of a materials' order parameter remains volatile. However, transition metal oxides are also sensitive to further environmental changes such as temperature, pressure, irradiation with light, the external partial pressure of oxygen or – as targeted specifically in the present framework – by the bombardment with ions. Changing one or more of those extrinsic parameters induces structure changes, stoichiometry deviations, modifications of the magnetic ordering and, most importantly, local variations of the electric conductivity. Extended intrinsic imperfections such as vacancy clusters, dislocations, structural grain boundaries, ferroic domain walls or heterophase interfaces are excellent trap sites to attract mobile charged species. In this way, regions of locally enhanced conductivity can percolate to current paths in a non-volatile way. Return to the pristine state may be driven by the entropy gain, when those conductive species are released from the current-leading paths by temperature and/or field effects.

Within the MEMRIOX cooperation particular emphasis has been put on the controlled formation and operation of such reversibly conducting regions with high spatial precision. Two routes have been followed to achieve spatial resolution: the targeted local modification with the help of ion beams and the use of nanowires and very thin films. The presentation will first summarize mostly experimental results on the resistive properties of material stacks for resistive devices and the influence of ion-beam treatment on such systems. The second part will focus on the microscopic picture of materials' changes in the vicinity of structural imperfections, which are accessible to modeling from first-principles.

Mainly ternary compounds from the perovskite and spinel families will be discussed as examples, which correlate local changes due to point and planar defects with changes of the elastic, polarization and magnetic properties. The microscopic interactions are determined by density functional calculations, which yield the basis for more large-scale simulations with effective Hamiltonian approaches. Examples include the field-induced migration of oxygen vacancies in $SrTiO_3$, the complex domain wall behavior in epitaxially strained $SrMnO_3$ and the interplay between epitaxy strain, local ion-beam-induced structure changes and vacancy formation in $NiCo_2O_4$, which has a series of complex antiferromagnetic phases coexistent with ferroelectricity.

# What Researchers need to know about Technology Transfer, Validation, and Exploitation

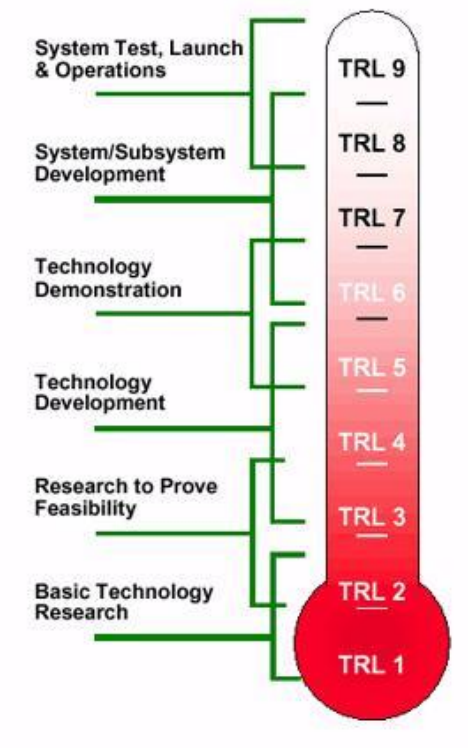
Stephan Krüger and Björn Wolf[1,2]

Department of Technology Transfer and Legal Affairs,
Helmholtz-Zentrum Dresden-Rossendorf (HZDR), 01314 Dresden, Germany

Corresponding author: s.krueger@hzdr.de

**Abstract**

Exploitation-wise, Research & Development can be grouped into different technology readiness levels (TRL). Where do you group your Memriox` results and how do you pave the way to stunning applications?




At November 21st /22nd, 2016 there will be an Innovation Forum, held in Dresden, concerning one of the essential parts of Memriox – the BFO topic. The Innovation Forum aims to attract investors and industry: www.charged-oxides.com

# In-situ electrical characterization of ion beam irradiated wire- and layer-like $VO_2$ structures

Jura Rensberg[1], Alexander Tille[1], Clemens Anschütz[1],
Tommy Schönherr[2], Barbara Abendroth[3], Arthur Erbe[2], Carsten Ronning[1]

[1]Institute of Solid State Physics, Friedrich Schiller University Jena, Germany
[2]Institute of Ion Beam Physics and Materials Research, Helmholtz-Zentrum Dresden-Rossendorf, Germany
[3]Institute of Experimental Physics, TU Bergakademie Freiberg, Germany

Corresponding authors: jura.rensberg@uni-jena.de & carsten.ronning@uni-jena.de

One of the most interesting and widely studied phase-transition materials is vanadium dioxide ($VO_2$), which exhibits an insulator to metal transition (IMT) upon heating above a critical temperature of $T_C \sim 68$ °C. The high-speed reversible transition in the order of nanoseconds makes this material very promising for integrated devices like ultra-fast switches, sensors, or memories [1]. The growth of high-quality $VO_2$ thin films with bulk-like electronic properties and high crystalline quality is still challenging. Enormous strains exist at the interfaces, mainly due to the differences in the crystal lattice parameters and thermal expansion coefficients between film and substrate, resulting in a broadening of the phase transition region. In contrast, stress can easily relax during the growth of nanostructures via unconstrained surfaces, making $VO_2$ nano-/microwires an ideal model system with very high crystal quality and sharp phase transitions.

Here, we compare in-situ temperature dependent electrical characterization of $VO_2$ thin films and microwire structures during ion irradiation. The experiment is complimented by temperature dependent optical and structural characterization using reflectance measurements, Raman spectroscopy, and X-Ray diffraction. We demonstrate how - independent on the geometry - ion beam irradiation can be used to modify and engineer the IMT via the intentional creation of defects and lattice damage.

Unlike existing means to modify the IMT via doping during growth, ion beam irradiation can be combined with lithographic patterning to create complex electrical and optical devices on a subwavelength scale [2]. Thermal annealing shows, that this type of defect engineering is a robust method to engineer the phase transition and greatly expands the utility of phase-change materials.

[1] M. Soltani *et al.*, Applied Physics in the 21st Century (2008).
[2] J. Rensberg *et al.*, Nano Letters **16**,1055 (2016).

# Ion irradiated, non-volatile, analog resistive oxide switches for neuromorphic computing and fuzzy logics

Heidemarie Schmidt[1]

[1]Technische Universität Chemnitz, Fakultät für Elektrotechnik und Informationstechnik
Nano-Spintronics group, Reichenhainer Straße 39
09126 Chemnitz, GERMANY

Corresponding author: Heidemarie.Schmidt@etit.tu-chemnitz.de

The ongoing research towards non-volatile memory devices has inspired many researchers to investigate different switching mechanisms, e.g. resistive switching [1] and capacitive switching [2], in oxide thin films. Here we present nonvolatile resistive switching without an electroforming process in two-terminal Ti-doped bismuth ferrite ($BiFeO_3$) thin film structures [1]. Fixed Ti donors can effectively trap mobile donors in $BiFeO_3$ if a writing voltage is applied [3]. The concentration of mobile and fixed donors is controlled during the $BiFeO_3$ thin film synthesis, e.g. by $Ar^+$ [4] and $Ti^+$ ion irradiation. In a proof-of-principle experiment for $BiFeO_3$ structures with one flexible and one unchangeable electrode we have demonstrated fast and energy-efficient spike-timing dependent plasticity (STDP) for learning [5] and higher harmonics generation for hardware-based cryptography [6]. For $BiFeO_3$ structures with two flexible electrodes we have demonstrated the nonvolatile reconfiguration of all 16 Boolean logic gates for application in binary logics [7] and in fuzzy logics. Furthermore, four learning curves have been realized with the same analog resistive oxide switch and application for complementary learning is discussed.

[1] Y. Shuai et al., Appl. Phys. Exp. 4 (2011); J. Appl. Phys. 109 (2011)
[2] T. You et al., Adv. Electron. Mater. 2 (2016)
[3] T. You et al., ACS Applied Materials & Interfaces 6 (2014);
[4] X. Ou et al., ACS Appl. Mater. Interfaces 5 (2013)
[5] N. Du et al., Front Neurosci. 9 (2015)
[6] N. Du et al., J. Appl. Phys. 115 (2014)
[7] T. You et al., Adv. Funct. Mater. 24 (2014)

# The negative aspects at the origin of hysteresis effects in FeFETs and niobium threshold switches

Stefan Slesazeck[1]

[1]NaMLab gGmbH, Dresden, Germany

Corresponding author: stefan.slesazeck@namlab.com

Recent discovery of ferroelectricity in thin hafnium oxide films has led to a resurgence of interest in ferroelectric memory devices. The electrical characteristic of these ferroelectric capacitors with hysteretic capacitance can be described as a kind of memcapacitor. However, besides the multi-domain characteristic typically measured in MFM structures, first evidence for single domain switching in ultra-scaled ferroelectric field effect transistors (FeFET) was revealed. Currently the underlying physical mechanisms are still under debate. The occurrence of a negative differential capacitance effect might explain the hysteretic switching behavior, yielding the memory window in FeFETs.

A completely different kind of device are the niobium oxide based volatile threshold switching memristors, which have been gaining interest due to their potential application as ReRAM selector elements. This devices exhibit a switching hysteresis in their IV-characteristic originating from the emergence of a negative differential resistance effect. Even though, negative resistance and negative capacitance effects stem from completely different physical effects, the characterization and modeling approaches have some commonalities. In my talk I will discuss our approaches to characterize these devices. Further, I will shed some light on new opportunities which might arise from the infinitesimal gain of energy in the 'negative differential' regions, such as the realization of steep-sub-threshold devices and self-sustaining oscillators.

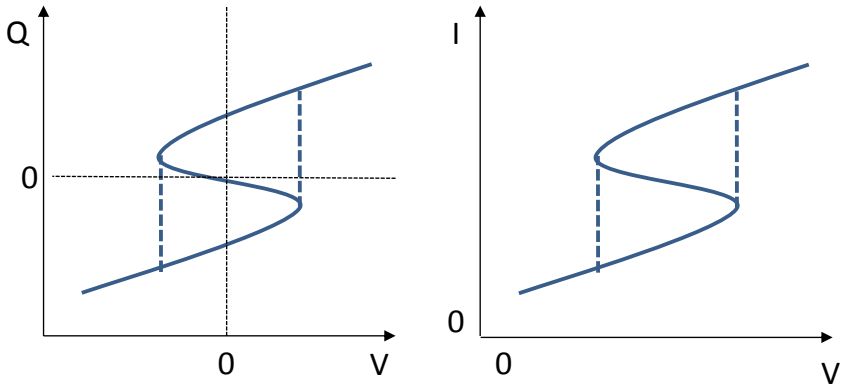



Fig. 1: Illustration of negative differential capacitance and negative differential resistance

# The diversity of Nb2O5-based Memristors

Stefan Slesazeck[1], Helge Wylezich, Melanie Herzig, Thomas Mikolajick[1,2]

[1]NaMLab gGmbH, Dresden, Germany
[2]Faculty of Electrical Engineering and Information Technology, TU-Dresden, Dresden, Germany

Corresponding author: Stefan.Slesazeck@namlab.com

Niobium oxide based memristor devices exhibit various electrical characteristics such as non-volatile analog or more digital abrupt memory switching and the volatile threshold switching. The electrical characteristic of the resistance switching devices depends not only on the device configuration, such as the oxide stoichiometry and crystallinity or the electrode material, but also on the electrical excitation. Furthermore, there are different manufacturing processes to realize the different device types, including ion implantation to locally change the oxides stoichiometry. In our talk we will give an overview on the different memristor characteristics which we realized in course of our Memriox activities. From the results we will emphasize that the niobium oxide system is a suitable model system to analyze different switching mechanisms which might occur in other material systems as well. In particular, the threshold switching effect in Niobium oxide is found to be independent of the material specific effect of metal-insulator-transition at 1070 K, but can be explained by a trap-assisted Frenkel-Poole like conduction mechanism in combination with a moderate temperature increase by only 150 K due to Joule heating. A physics based model of pure electrical nature explains the occurrence of the threshold effect as well as the negative-differential resistance behavior observed in Niobium oxide. The threshold switching might be at the origin of abrupt memory switching effects which are observed in many other commonly used material systems such as the Tantalum or Hafnium oxide devices.

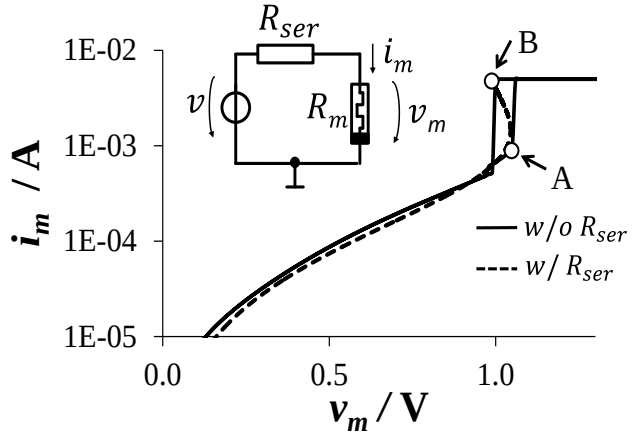



Fig. 1 Quasi-static *im-vm* characteristic of the threshold switching device with and without a stabilizing resistor of value *Rser* = 330Ω. The inset shows the measurement circuit with series resistor.

# Non-volatile resistive switching in $BiFeO_3$-based thin films and the reconfigurable logic application

Tiangui You,[1] Nan Du,[1] Danilo Bürger,[1] Ilona Skorupa,[1,2] Oliver G. Schmidt,[1,3] Heidemarie Schmidt[1]

*1. Materials systems for Nanoelectronics, Chemnitz University of Technology, 09126 Chemnitz, Germany*

*2. Institute of Ion Beam Physics and Materials Research, Helmholtz-Zentrum Dresden-Rossendorf, Dresden, Germany*

*3. Institute for Integrative Nanosciences, IFW Dresden, Dresden, Germany*

Resistive switching devices are considered as one of the most promising candidates for the next generation memories and nonvolatile logic applications. This provides the possibility to carry out the processing and storage simultaneously and at the same resistive switching devices, which could offer very significant performance benefits and overcome the von-Neumann bottleneck.[1,2] In this work, bipolar resistive switching was observed in polycrystalline $BiFeO_3$ (BFO), $BiFeO_3$:Ti (BFTO), and BFO/BFTO thin films which were fabricated by pulsed laser deposition. The resistive switching mechanism is understood by a model of complementary tunable height of top and bottom Schottky-like barrier due to the mobile and fixed donors in BFO-based thin films.[3-5] It is expected that the oxygen vacancies act as the mobile donors which can be redistributed under the writing bias to change the Schottky barrier heights, and Ti atoms act as the fixed donors which can effectively trap and release oxygen vacancies and consequently stabilize the resistive switching in BFO-based thin films. The BFO/BFTO bilayer structures show stable and nonvolatile resistive switching behavior in both positive and negative bias.[4] Different from the conventional bipolar resistive switching, the resistance state of BFTO/BFO bilayer structures not only depends on the writing bias, but also on the polarity of reading bias. For the reconfigurable logic applications, the polarity of the reading bias can be used as an additional logic variable, which makes it feasible to program and store all 16 Boolean logic functions simultaneously and into a same single cell of BFTO/BFO bilayer structure in three logic cycles. This promises a most efficient and effective means for implementing beyond von-Neumann computing.

[1] J. Bachus, *Commun. ACM*, 1978, **21**, 613

[2] T. Hasegawa, et al., *Adv. Mater.*, 2010, **22**, 1831

[3] T. You, et al., *ACS Appl. Mater. Interfaces*, 2014, **6**, 19758

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## Your Notes

**MEMRIOX International Workshop 2016, Berghotel Bastei**

## Posters

updated: 14.09.2016 (PZ)

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2) Please, remove your poster on Tuesday at latest 12:00.

# Structural and electrical characterization of ALD grown $HfO_2$ thin films

Daniel Blaschke[1,2], Frans Munnik[1], Vitaly Zviagin[3], Rüdiger Schmidt-Grund[3], Marius Grundmann[3], Agnieszka Bogusz[1,4], René Hübner[1], Andrea Scholz[1], Heidemarie Schmidt[4], Peter Zahn[1], and Sibylle Gemming[1,2]

[1)] Institute of Ion Beam Physics and Materials Research, Helmholtz-Zentrum Dresden-Rossendorf, Germany

[2)] Chair of Scale-bridging Materials Modeling, Faculty of Natural Science, Chemnitz University of Technology, Germany

[3)] Chair of Semiconductor Physics, Faculty of Physics and Earth Science, Universität Leipzig, Germany

[4)] Chair of Material Systems for Nanoelectronics, Faculty of Electrical Engineering and Information Technology, Chemnitz University of Technology, Germany

Corresponding author: d.blaschke@hzdr.de

Abstract

$HfO_2$ films were grown on 4-inch native $SiO_2$/Si wafers by Atomic Layer Deposition (ALD) from tetrakis(dimethylamido)hafnium and deionized water in a Savannah S100 system. The temperature was varied from 100°C to 350°C in steps of 50 K. All other ALD process parameters were fixed. The resulting $HfO_2$ layers were characterized in terms of thickness homogeneity, growth rate per cycle, surface roughness, crystal structure, stoichiometry, mass density, optical bandgapm, and index of refraction. Based on the obtained growth rate of $HfO_2$ on $SiO_2$/Si, 25 nm thick $HfO_2$ layers were grown on Pt/Ti/$SiO_2$/Si substrates for electrical characterization. Furthermore, the most important structural properties were compared for the growth of $HfO_2$ on Pt/Ti/$SiO_2$/Si and $SiO_2$/Si substrates.
$HfO_2$ breakdown voltages show a clear decrease with increasing growth temperature, which was correlated to the crystallinity of the films due to the preferred breakdown along grain boundaries. With respect to resistive switching, an amorphous $HfO_2$ layer grown at 150 °C was compared to a crystalline one grown at 300 °C. Furthermore, resistive switching characteristics were tuned by the use of two different top electrode materials, namely an inert Pt or a reactive Ti/Pt electrode. The contact diameter was 50 µm. In the majority of cases, the resistive switching mode was found to be unipolar. Only the combination of a crystalline $HfO_2$ layer with an inert Pt bottom and a reactive Ti/Pt top electrode led to a “pseudo-bipolar” switching mode with a convertible SET and RESET polarity.

The project is funded by the Initiative and Networking Fund of the Helmholtz Association (Virtual Institute Memriox, VH-VI-422).

# Giant resistive switching by conductive domain walls in exfoliated thin-film lithium niobate

Thomas Kämpfe[1], Alexander Haußmann[1], Scott Johnston[2], Eric Yue Ma[2], Zhi-Xun Shen[2], Hui Hu[3], Lukas M. Eng[1]

[1]Institut für Angewandte Physik, Technische Universität Dresden, 01069 Dresden, Germany
[2]Department of Physics, Stanford University, CA-94305 Stanford, USA
[3]School of Physics, Shandong University, Jinan 250100, China

Corresponding author: thomas.kaempfe@tu-dresden.de

We investigate the formation of conductive domain walls (CDWs) in exfoliated thin-film lithium niobate ($d$ = 600 nm) on a global Pt back-electrode under both Cr/Au plane electrodes ($A$ ~ 200µm$^2$) as well as sharp Pt-AFM probes ($A$ ~ 300nm$^2$). We observe a very distinct and abrupt increase in the conductivity over at least 5 orders of magnitude at $V_{set}$ = 21.05 V. The set voltage $V_{set}$ is as precisely defined as $\Delta V_{set}/V_{set} < 10^{-3}$. The set-voltage was sufficiently correlated with the coercive voltage in the material by switching-spectroscopy (ss) PFM. Plane electrode and AFM-based resistive switching were sufficiently found to be correlated to the formation of CDWs both by local cAFM and PFM measurements. ssPFM shows the coercive voltage to be very homogeneously distributed in the single crystalline film ($\Delta V_c$ < 1V) with a small internal field. Scanning microwave impedance microscopy (sMIM) and comparative FEM simulation reveal a clear increase in conductivity at the domain wall from $10^{-8}$ S/m to $10^3$ S/m, which gives a significantly larger ratio of $10^{11}$ as the background is distinctly measurable.

The application of CDWs for resistive switching devices was investigated. We were able to measure a stable on- and off-current over $10^4$ s in the metal-ferroelectric-metal stack at read-voltages <10 V. It was possible to cycle the device for as much as $10^5$ cycles with a resistance window of $10^4$. The read-current in such a device was tunable both by writing voltage and time with a variation of $10^2$. The transport mechanism was found to be space-charge limited current both by direct $I$-$V$ curve analysis as well as by $dlog(I)/dV$.

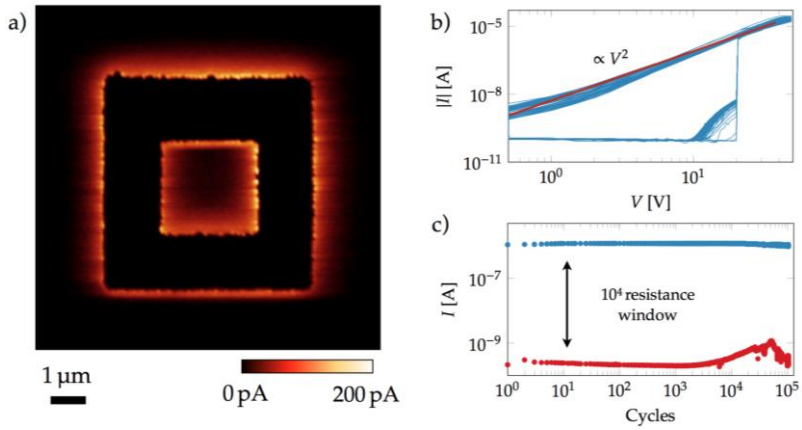



Fig. 1: Resistive switching by ferroelectric domain walls in lithium niobate a) conductive AFM measurement at $V$ = 3 V, b) 100 cycles of current-voltage ($I$−$V$) curves on metal- ferroelectric-metal stack c) cyclic resistive switching with a read-out voltage $V_{read}$ = 10 V.

# Open volume defects in annealed and ion irradiated $SrTiO_3$ systems

Maciej Oskar Liedke[1], Kay Potzger[2], Naren Srinivasan[1,2], and Andreas Wagner[1]

[1]Institute of Radiation Physics, Helmholtz-Zentrum Dresden-Rossendorf, Dresden, Germany
[2]Institute of Ion Beam Physics and Materials Research, Helmholtz-Zentrum Dresden-Rossendorf, Dresden, Germany

Corresponding author: m.liedke@hzdr.de

The formation of vacancies in $SrTiO_3$ due to annealing in vacuum [1] or low-energy ion irradiation [2] also leads to the modification of electronic properties that could be exploited in applications like resistivity switching. Here, we employ both these modification methods combined with in-situ defects evolution analysis by means of Doppler Broadening Positron Annihilation Spectroscopy (DB-PAS). Two reference $SrTiO_3$ single crystals, undoped and Nb-doped have been investigated. Vacuum annealing of undoped $SrTiO_3$ increases the defect concentration which can be attributed to the creation of oxygen and strontium vacancies close to the sample surface and deeper inside the substrate, respectively. Variable energy DB-PAS reveals the depth-dependent open volume defects concentration. Second, in-situ room and low temperature (about 165K) $Ar^+$ irradiation has been utilized as a preliminary attempt to understand a vacancy migration process beneath the kinetical ion penetration depth. For that purpose, a frozen vacancy state is required, which could be achieved by lowering the system temperature below the vacancy activation energy. Although, that part of the experiment has been not entirely successful due to not low enough temperature realized during ion irradiation, however, a strong conclusion has been drawn about necessity of performing such type of experiments *in-situ*. A clear indication of oxygen deficiency at the sample surface due to ion irradiation, as found from DB-PAS is diminished after repeated *ex-situ* measurements.

[1] T. Nestler, *et al*., Appl. Phys. A, **105**, 103 (2011)
[2] G. Herranz, *et al.*, J. Appl. Phys. **107**, 103704 (2010)

# Domain walls in $SrMnO_3$ thin films under epitaxial tensile strain

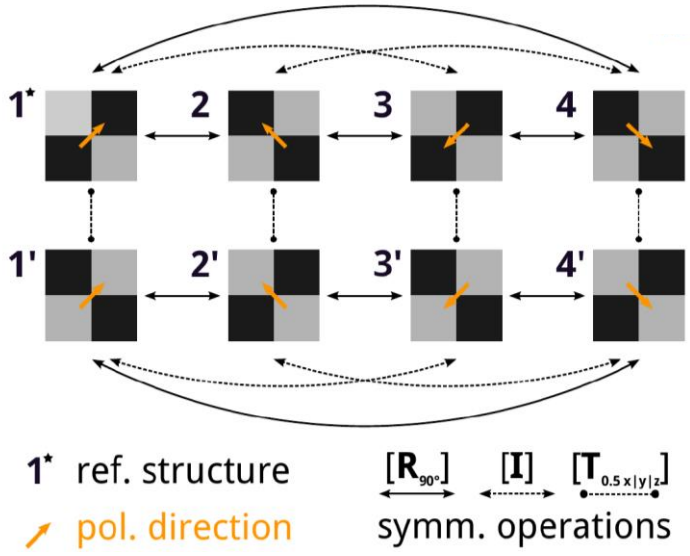
Lokamani[1], C. Faber[3], P. Zahn[1], N. Spaldin[3], and S. Gemming[1,2]

[1]Institute of Ion Beam Physics and Materials Research, HZDR, 01314 Dresden, Germany

[2]Institute of Physics, Technische Universität, 09107 Chemnitz, Germany

[3]Materials Theory, ETH, 8093 Zürich, Switzerland

Strontium manganate ($SrMnO_3$), a perovskite polymorph, exhibits cubic structure at low temperatures, which transforms into a hexagonal one at high temperatures. Density-functional calculations showed earlier, that under tensile strain the ground state of bulk $SrMnO_3$ corresponds to a G-type-antiferromagnetic (G-AFM) cubic structure. If deposited as epitaxially strained thin film a rearrangement of the $MnO_6$ coordination polyhedra was calculated, which is antiferrodistortive in the plane parallel to the substrate [1]. Recently, ferroelectric domains have been observed experimentally in thin films of $SrMnO_3$ (20 nm) on (001)-oriented LSAT with a 1.7 % tensile strain [2]. Strikingly, the domain walls were found to be electrically insulating, rendering the domains to form stable nano-capacitor.




The local polarization direction in the domains and polarization profile across the domain walls is not known experimentally. Given the fact that the ground state building block for the supercell possesses four rotational and two translation degrees of freedom, about twenty-eight different configurations for the supercell structure are possible. We employ group theoretical analysis to filter out the at-most unique configurations of the domain walls with both SrO2 and MnO2 termination planes. Concurrently, we present a first-principle investigation of the domain wall formation energies and the structural and electronic properties in these unique configurations.

As outlook, we present first results on the effect of vacancies and defects on the energetic and conductance properties of such domain walls.

1) J. H. Lee *et al.*, PRL **104**, 207204 (2010).
2) C. Becher *et al.*, Nature Nanotechnology **10**, 661 (2015).

# A new ion-beam laboratory at the Slovak University of Technology

Pavol Noga[1], Jozef Dobrovodský[1], Stanislav Minárik[1], Anna Závacká[1], Martin Muška[1], Matúš Beňo[1], Dušan Vaňa[1], Radoslav Halgaš[1], Róbert Riedlmajer[1]

[1]Advanced Technologies Research Department, Faculty of Materials Science and Technology in Trnava, Slovak University of Technology in Bratislava, Trnava, Slovakia

Corresponding author: pavol.noga@stuba.sk

An ion beam laboratory has been commissioned recently at the Slovak University of Technology within the University Science Park CAMBO located in Trnava. The facility will serve research in the field of materials science, physical engineering and nanotechnology. Two systems have been put into operation. First, a 6MV Tandetron electrostatic tandem accelerator equipped with Cs-sputtering and Duoplasmatron ion sources and end-stations for high energy ion-beam implantation as well as ion-beam analysis with RBS, PIXE, ERDA analytical systems. Second, a 500kV implanter equipped with a Bernas type ion source and two experimental wafer processing end-stations. The facility, first experiments and near future development plans are presented.

# High Magnetic Moment in Ferrimagnetic $NiCo_2O_4$ films via He Ion Irradiation

Parul Pandey[1], Yugandhar Bitla[2], M. Zschornak[1], M. Wang[1], C. Xu[1], J. Grenzer[1], S. Gemming[1], M. Helm[1], Ying-Hao Chu[2,3], S. Zhou[1]

[1]Helmholtz-Zentrum-Dresden-Rossendorf, Institute of Ion Beam Physics and Materials Research, Dresden, Germany
[2]Department of Materials Science and Engineering, National Chiao Tung University, Hsinchu, Taiwan
[3]Institute of Physics, Academia Sinica, Taipei, Taiwan

Corresponding author: p.pandey@hzdr.de

The spinel $NiCo_2O_4$ (NCO) exhibits unique combination of electrical conductivity, infrared transparency, electro catalytic activity, and ferrimagnetic order, which makes it an attractive material for spintronic applications.[1] The NCO thin-films electrical and magnetic properties can be manipulated from high temperature ferrimagnetic and metallic to low temperature ferromagnetic and insulating by changing the growth temperature.[2] The high quality epitaxial NCO films were grown on $MgAl_2O_4$ (100) substrate at ~ 400°C exhibits metallic behavior accompanied by ferrimagnetic order with moment ~ 2 $\mu_B$/fu.[1,2] Here, we report the impact of He-ion irradiation with fluence ranging from $5\times10^{15}$/cm$^2$ – $3\times10^{16}$/cm$^2$ on these metallic NCO films. The use of He-ion irradiation results in the coherent control of out-of-plane lattice parameter of these films without changing its in-plane lattice parameter (Figure 1). The comprehensive study of magnetization data reveals the magnetic moment increases drastically to ~ 4 $\mu_B$/fu (Figure 1). The X-ray absorption spectroscopic study also suggests the possible charge redistribution within the octahedral sites of the NCO films which corroborates well with the increase in the magnetic moment.

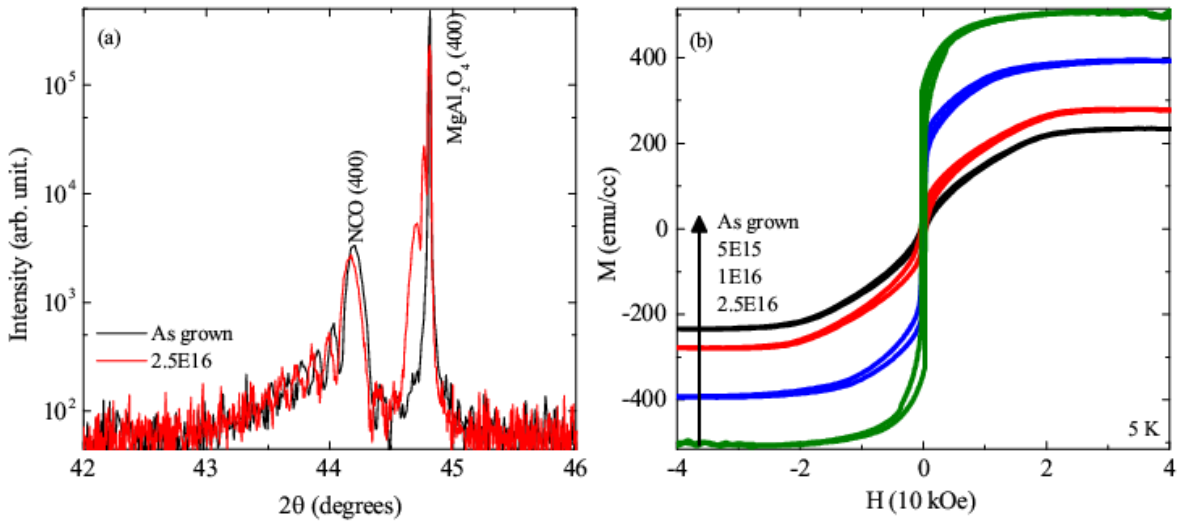



Figure1: (a) *θ-2θ* scan and (b) field (H) dependent magnetization (M) of as grown and the He-ion irradiated NCO films.

1. Punam Silwal *et al.*; *Appl. Phys. Lett.* **100**, 032102 (2012)
2. Yugandhar Bitla *et al.*; *Sci. Rep.* **5**,15201 (2015)

# Nonvolatile resistive switching to $10^5$ ohms OFF/ON resistance ratio in yttrium manganite thin films with downscaled top electrodes

Venkata Rao Rayapati[1], Agnieszka Bogusz[1,2], Nan Du[1], Danilo Bürger[1], Ilona Skorupa[1,2], Oliver G. Schmidt[1,3], Heidemarie Schmidt[1]

[1]Materials systems for Nanoelectronics, Chemnitz University of Technology, 09126 Chemnitz, Germany
[2] Helmholtz-Zentrum Dresden-Rossendorf, Institute of Ion Beam Physics and Materials Research, Bautzner Landstraße 400, 01328 Dresden, Germany
[3]Institute for Integrative Nanosciences, IFW Dresden, Dresden, Germany

Corresponding author: venkata-rao.rayapati@s2013.tu-chemnitz.de

Polycrystalline, hexagonal $YMnO_3$ (YMO) thin films grown by pulsed laser deposition (PLD) with the use of stoichiometric, ceramic YMO target, exhibit a nonvolatile unipolar resistive switching [1, 2]. In this work, we investigate the resistive switching properties of YMO thin films prepared by PLD with the use of ceramic YMO target of different chemical compositions on a large-scale Pt and Pt/Ti bottom electrode on $SiO_2$/Si substrates. Circular Au and Al top electrodes of different size have been prepared by magnetron sputtering and e-beam evaporation, respectively. All tested $YMnO_3$-based structures show unipolar resistive switching (URS). URS depends on the ceramic target and bottom electrode. Endurance of YMO with Al top electrodes is improved in comparison to YMO with Au top electrodes. In addition, for large Al top electrodes and Pt and Pt/Ti bottom electrode URS with $10^2$-$10^4$ and $10^1$-$10^3$ OFF/ON resistance ratio has been realized, respectively. The phenomenon of increasing OFF/ON resistance ratio with decreasing size of Al top electrodes can be related with the dependence of $R_{OFF}$ on electrode size, namely $R_{OFF}=10^6$ Ω for electrode diameter 260 μm and $10^4$ Ω for electrode diameter 800 μm. $R_{OFF}$ reflects decreased tendency for local shunting of YMO thin films in HRS.

[1] A. Bogusz et al., AIP Advances 4 (2014)
[2] A. Bogusz et al., Adv. Mater. Res. 1101 (2015)

# Spatially selective ion irradiation-
# Tuning thin films into inherently flat metadevices

Jura Rensberg[1], Mikhail A. Kats[2,3], Carsten Ronning[1]

[1] Institute of Solid State Physics, Friedrich Schiller University Jena, Germany
[2] Department of Electrical and Computer Engineering, University of Wisconsin - Madison, USA
[3] Materials Science Program, University of Wisconsin - Madison, USA

Corresponding authors: jura.rensberg@uni-jena.de & carsten.ronning@uni-jena.de

Traditional plasmonic metamaterials comprising metallic structures exhibit unique electromagnetic properties not found in natural materials, such as the ability to efficiently harvest light, extraordinary transmission beyond the diffraction limit, and the ability to control the reflection or transmission direction of light beams. However, they still require complex designs and sophisticated fabrication procedures, and their bandwidth is fixed to narrow resonances and they exhibit low optical efficiency due to the inherent metal absorption losses.

Metasurfaces can also be created by using plane, ultrathin films on reflective substrates. By using the controllable abrupt phase shifts associated with reflection or transmission of light waves at the interfaces, such metasurfaces operate like optically thin cavities that strongly modify the light spectrum [1].

Here, we demonstrate that spatially selective defect engineering on the nanometer scale can transform thin films into optical metasurfaces. Using ion irradiation through nanometer-scale masks, we selectively defect-engineered the insulator-metal transition of vanadium dioxide, a prototypical correlated phase-transition material whose optical properties change dramatically depending on its state. We demonstrated several optical metasurfaces, including tunable absorbers with artificially induced phase co-existence and tunable polarizers based on thermally triggered dichroism.

[1] N. Yu *et al.*, Nature Mat. **13**, 139 (2014)
[2] J. Rensberg *et al.*, Nano Letters **16**,1055 (2016).

# Avalanche-discharge-induced electrical forming in $Ta_2O_5$ based MIM structures

Katharina Skaja[1], Christoph Bäumer[1], Oliver Peters[1], Stephan Menzel[1], Marco Moors[1], Hongshu Du[1,2], Manuel Bornhöfft[1,2,3], Chun-Lin Jia[1,2], Joachim Mayer[2,3], Rainer Waser[1,4], and Regina Dittmann[1]

[1] Peter Grünberg Institut, Forschungszentrum Jülich, 52425 Jülich, Germany
[2] Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons, Forschungszentrum Jülich, 52425 Jülich, Germany
[3] Central Facility for Electron Microscopy, RWTH Aachen University, 52056 Aachen, Germany
[4] Institute of Materials in Electrical Engineering and Information Technology II, RWTH Aachen University, 52056 Aachen, Germany

Corresponding author: k.skaja@fz-juelich.de

We investigated the resistive switching characteristics of Pt/ $Ta_2O_5$/Ta cells prepared by sputtering. Structural changes in the top electrode develops during the electrical forming process, which can be correlated to the formation of a dendrite-like conductive structure, which is induced by an avalanche discharge between the top electrode and the $Ta_2O_5$ layer, which occurs instead of a local breakdown between top and bottom electrode. The dendrite-like structure evolves primarily at structures with a pronounced interface adsorbate layer. Local conductive atomic force microscopy reveals that the entire dendrite region becomes conductive. We demonstrate by in-situ spectromicroscopy that the subsequent switching is caused by a valence change between $Ta^{4+}$ and $Ta^{5+}$, which takes place over the entire former Pt/ $Ta_2O_5$ interface of the dendrite-like structure.

# Defect-induced magnetism in SiC probed by nuclear magnetic resonance

Z. T. Zhang,[1,2,3] D. Dmytriieva,[1,4] S. Molatta,[1,4] Yutian Wang,[2] Shengqiang Zhou,[2] Zhaorong Yang[3], Manfred Helm[2, 4], J. Wosnitza,[1,4] and H. Kühne[1,*]

[1] Hochfeld-Magnetlabor Dresden (HLD-EMFL), Helmholtz-Zentrum Dresden-Rossendorf, D-01314 Dresden, Germany
[2] Institute of Ion Beam Physics and Materials Research, Helmholtz-Zentrum Dresden-Rossendorf, D-01314 Dresden, Germany
[3] Key Laboratory of Materials Physics, Institute of Solid State Physics, Chinese Academy of Sciences, Hefei 230031, People's Republic of China
[4] TU Dresden, D-01062 Dresden, Germany

Corresponding author: h.kuehne@hzdr.de

It is still an open question whether a material with only ***s*** or ***p*** electrons can be magnetic. Recently, we showed that the defects in SiC, generated by neutron irradiation, introduce paramagnetism, the amplitude of which scales with the defect concentration [Phys. Rev. B **92**, 174409]. Here, we report on a **$^{13}$C** and **$^{29}$Si** nuclear magnetic resonance (NMR) investigation of the defect-induced magnetism in SiC. Consistent with magnetization measurements, the temperature dependence of the NMR shift can well be described by a Curie-Weiss behavior for both nuclear isotopes, allowing for a detailed study of the electronic paramagnetism in SiC from a local probe point of view. The increasing line width of the NMR spectra upon cooling indicates a growing amplitude of the internal dipole fields stemming from the local moments. Simulations based on a local dipole field model were performed, and are compared to the amplitude of the experimental **$^{13}$C** and **$^{29}$Si** NMR shifts and line widths. For comparison, NMR measurements were performed on a virgin sample of SiC without defects, and, as expected, no NMR signal was observed due to its gapped, non-magnetic nature. Our study provides clear indications of defect-induced magnetism in SiC.

The project is supported by Helmholtz-Association (VH-PD-146). Z. T. Zhang was financially supported by the National Nature Science Foundation of China (Grant No. 11304321) and by the International Postdoctoral Exchange Fellowship Program 2013 (Grant No. 20130025).

# Defect induced magnetism in SiC

**Yutian Wang, Yu Liu, Sybille Gemming, Manfred Helm, Shengqiang Zhou**

*Helmholtz-Zentrum Dresden-Rossendorf, Dresden, 01328, Germany*

Defect-induced magnetism is attracting intensive research interest. It not only challenges the traditional opinions about magnetism, but also has some potential applications in spin-electronics. SiC is a new candidate for the investigation of defect-induced ferromagnetism after graphitic materials and oxides due to its high material purity and crystalline quality [1, 2]. In this contribution, we made a comprehensive investigation on the structural and magnetic properties of ion implanted and neutron irradiated SiC sample. In combination with X-ray absorption spectroscopy and first-principles calculations, we try to understand the mechanism in a microscopic picture.

For neon or xenon ion implanted SiC, we identify a multi-magnetic-phase nature [3]. The magnetization of SiC can be decomposed into paramagnetic, superparamagnetic and ferromagnetic contributions. The ferromagnetic contribution persists well above room temperature and exhibits a pronounced magnetic anisotropy. By combining X-ray magnetic circular dichroism and first-principles calculations, we clarify that p-electrons of the nearest-neighbor carbon atoms around divacancies are mainly responsible for the long-range ferromagnetic coupling [4]. Thus, we provide a correlation between the collective magnetic phenomena and the specific electrons/orbitals.

With the aim to verify if a sample containing defects through its bulk volume can persist ferromagnetic coupling, we applied neutron irradiation to introduce defects into SiC [5]. Besides a weak ferromagnetic contribution, we observe a strong paramagnetism, scaling up with the neutron fluence. The ferromagnetic contribution only occurs in a narrow fluence window or after annealing. We speculate that defect-induced ferromagnetism rather locally appears in particular regions, like surface/interface/grain boundaries.

[1] L. Li, et al., *Appl. Phys. Lett.* 98, 222508 (2011).
[2] Y. Wang, et al., *Phys. Rev. B* 90, 214435 (2014).
[3] Y. Wang, et al., *Phys. Rev. B* 89, 014417 (2014).
[4] Y. Wang, et al., *Scientific Reports*, 5, 8999 (2015).
[5] Y. Wang, et al., *Phys. Rev. B* 92, 174409 (2015).

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| Di Ventra | Massimiliano | Mr. | Prof. Dr. | Univ. California San Diego |
| Gemming | Sibylle | Mrs. | Prof. Dr. | HZDR |
| Hafermann | Martin | Mr. | | FSU Jena |
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| Kämpfe | Thomas | Mr. | | IAPP, TU Dresden |
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| Zahn | Peter | Mr. | PD Dr. | HZDR |
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