# Symposium Voltage Control of Functional Interfaces: Magneto-ionic Meet Memristive Systems (SYVC)

jointly organized by

the Magnetism Division (MA), the Chemical and Polymer Physics Division (CPP), the Crystalline Solids and their Microstructure Division (KFM), and the Thin Films Division (DS)

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The control of functional material properties by an external voltage, as opposed to electric current based approaches, is intensely researched as a pathway to highly energy-efficient modern device operation. The magneto-ionic/electrochemical effect is a just emerging topic in the field of voltage controlled magnetic materials. In contrast to other magnetoelectric approaches, the magneto-ionic effect is based on ionic motion and electrochemical oxidation/reduction, and thereby allows to achieve large and non-volatile changes at room-temperature. However, the unambiguous identification of the microscopic origin is still challenging. The symposia gives an overview over magneto-ionic systems by covering both solid and liquid electrolyte-based architectures. Advanced interface-sensitive and in situ techniques that enable the investigation of the microscopic origin will be presented. A comparison with memristive/resistive approaches is made to elucidate differences and similarities in the underlying ionic mechanisms. The transfer to the tuning of other functional properties and from an interfacial effect to the control of bulk materials will be addressed to initiate a fruitful discussion about future key strategies.

## **Overview of Invited Talks and Sessions**

(Lecture room H 0105)

### Invited Talks

SYVC $1.1$	Wed	15:00-15:30	H $0105$	Magneto-ionic control of interfacial magnetism — • GEOFFREY BEACH
SYVC $1.2$	Wed	15:30 - 16:00	H $0105$	Ionic Control of Materials Beyond Interfaces — • DUSTIN GILBERT
SYVC $1.3$	Wed	16:00-16:30	H $0105$	Microscopic Mechanisms of Memristive Switching in Metal Oxides
				— •Rainer Waser, Stephan Menzel, Regina Dittmann
SYVC $1.4$	Wed	17:00-17:30	H $0105$	In-situ and operando SQUID magnetometry under electrochemical
				control — •Roland Würschum, Markus Gössler, Gregor Klinser,
				Eva-Maria Steyskal, Heinz Krenn
SYVC $1.5$	Wed	17:30 - 18:00	H $0105$	Reversible chemistry as a tool for dynamic control of physical prop-
				erties — • Robert Kruk, Subho Dasgupta, Bijoy Das, Horst Hahn

#### Sessions

SYVC 1.1–1.5 Wed 15:00–18:00 H 0105

05 SYVC: Voltage Control of Functional Interfaces – Magneto-ionic Meet Memristive Systems

### SYVC 1: SYVC: Voltage Control of Functional Interfaces – Magneto-ionic Meet Memristive Systems

Time: Wednesday 15:00-18:00

Invited Talk SYVC 1.1 Wed 15:00 H 0105 Magneto-ionic control of interfacial magnetism — •GEOFFREY BEACH — Massachusetts Institute of Technology, Cambridge, Massachusetts, USA

Voltage control of magnetism has the potential to substantially reduce power consumption in spintronic devices, while offering new functionalities through field-effect operation [1-4]. Magneto-electric coupling has usually been achieved using complex oxides such as ferroelectrics, piezoelectrics, or multiferroic materials. Here I describe a new approach to voltage control of magnetism based on solid-state electrochemical switching of the interfacial oxidation state [2-4] in thin metallic ferromagnets. In ultrathin ferromagnet/oxide bilayers, perpendicular magnetic anisotropy (PMA) arises from interfacial hybridization between the ferromagnetic 3d and oxygen 2p orbitals. By using GdOx as a gate oxide with high oxygen ion mobility, we show that O2- can be reversibly displaced at Co/GdOx [4] interfaces with a small gate voltage, leading to giant changes in magnetic properties [4]. This talk will review recent results in magneto-ionic control of magnetism and give new insights into the electrochemical mechanisms that enable control not only of surface magnetism, but of a wide array of interfacial properties that manifest at metal/oxide interfaces.

U. Bauer, et al., Nano Lett. 12, 1437 (2012) [2] U. Bauer, et al., Appl. Phys. Lett. 100, 192408 (2012) [3] U. Bauer, et al., Nature Nano. 8, 411 (2013) [4] U. Bauer, et al., Nature Mater. 14, 174 (2015)

Invited TalkSYVC 1.2Wed 15:30H 0105Ionic Control of MaterialsBeyond Interfaces• DUSTINGILBERT — National Institute of Standards and Technology, Gaithersburg, MD 20899, USAUSA

Designing ionic distributions in materials is one of the most promising approaches for future nanotechnology devices, enabling control of virtually every property of that material, ranging from electrical and thermal resistivity to superconductivity, or even optical, mechanical, and magnetic properties. Control of (quasi-)bulk ionic distributions is of particular interest to us, as some material properties are most useful, or even require, lengthscales larger than what is typically considered interfacial. In this talk, I will present our works controlling ionic distributions and consequentially material properties within metal(-oxide) thin films. First I will discuss our recent investigations into control of magnetism, resistivity, structure and superconductivity in magnetic heterostructures and perovskites (La,Sr)(Co,Mn)O3, and YBa2Cu3O7. We use a solid-state redox approach to leach oxygen from our films, thus continuously controlling our materials. I will show that perovskites present particular promise for ionic technologies due to their high ion mobility and sensitivity to chemical stoichiometry. In the second part of the talk I will discuss our work designing magnetism in Co thin-films with electric fields. Our work demonstrates critical limits and identifies details crucial to control of magnetism in metallic systems particularly in the quasi-bulk lengthscale. These works show that control of ionic distributions is achievable even beyond the interface and this presents fantastic opportunities for next generation technologies.

Invited TalkSYVC 1.3Wed 16:00H 0105Microscopic Mechanisms of Memristive Switching in MetalOxides — •RAINER WASER<sup>1,2</sup>, STEPHAN MENZEL<sup>2</sup>, and REGINADITTMANN<sup>2</sup> — <sup>1</sup>IWE2 & JARA, RWTH Aachen, Aachen, Germany— <sup>2</sup>PGI-7, FZ Jülich, Germany

Redox-based memristive switching devices (ReRAM), also called nanoionic memories, are widely considered as energy-efficient approaches to non-volatile memories and neuromorphic concepts. In this talk, the ultra-high non-linearity of the switching kinetics of redoxbased 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 VCMtype 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. The relevance of these aspects for neuromorphic applications will be worked out.

#### 30 minutes break

Invited Talk SYVC 1.4 Wed 17:00 H 0105 In-situ and operando SQUID magnetometry under electrochemical control — ●ROLAND WÜRSCHUM<sup>1</sup>, MARKUS GÖSSLER<sup>1</sup>, GREGOR KLINSER<sup>1</sup>, EVA-MARIA STEYSKAL<sup>1</sup>, and HEINZ KRENN<sup>2</sup> — <sup>1</sup>Inst. of Mater. Phys., TU Graz, Austria — <sup>2</sup>Inst. of Phys., Univ. Graz

In the research field of voltage-controlled functional materials, an attractive pathway is currently opening up with the the development of in-situ electrochemical techniques for SQUID magnetometry [1]. This combines the high SQUID sensitivity with the electrochemical tools for electronic and chemical controlling of surfaces by means of double layer charging, oxidation and reduction, electrochemical loading and extraction, as well as thin-film deposition. An overview will be given on our recent studies in this field comprising, on the one hand, the insitu electrodeposition of thin Co films [2] as well as charging-induced variations of the magnetic moment of nanophase metals [3]. Moreover, reduction and oxidation processes of transistion metal oxides were insitu monitored upon electrochemical insertion and extraction of Li, serving as highly sensitive fingerprint for the charge state of electrode materials [4]. Finally, the electrochemical charging-induced variation of the electrical resistance of nanophase and nanoporous metals will be addressed [5]. Acknowledgment: This work is financially supported by the Austrian Science Fund (FWF): P30070-N36.

S. Topolovec et al., Rev.Sci.Instr. 86 (2015) 063903. [2] S.
Topolovec et al., JMMM 397 (2016) 96. [3] E.-M. Steyskal et al.,
Beilst.J.Nanotech. 4 (2013) 394. [4] G. Klinser et al., APL 109 (2016) 213901. [5] E.-M. Steyskal et al., Langmuir 32 (2016) 7757.

Invited TalkSYVC 1.5Wed 17:30H 0105Reversible chemistry as a tool for dynamic control of physicalproperties- • ROBERT KRUK, SUBHO DASGUPTA, BLJOY DAS, andHORST HAHN- 1 Institute of Nanotechnology, Karlsruhe Institute ofTechnology, 76344 Eggenstein-Leopoldshafen, Germany

The idea of control of magnetic properties with reversible chemistry rests on a non-destructive, cyclic insertion (and extraction) of mobile ions into a magnetic lattice followed by a reversible chemical reaction with the magnetic ions, thus resulting in a radical change in the magnetic state of the material. This new approach to control magnetic response in a material via electrochemistry goes beyond surface and interface effects and applies to any volume of magnetic materials. Moreover, aiming at a complete on-and-off magnetic switching, reversible ion-exchange processes can be utilized as a tool to annihilate magnetism in bulk ferromagnets or to create magnetism in non-magnetic systems. This presentation provides background and discusses key issues in practical realization of electrochemically tunable systems. Selected examples of successful applications of the ion- insertion control principle are presented, including reversible control of magnetic states in spinels [1].

References

[1] Dasgupta, S.; Das, B.; Li, Q.; Wang, D.; Baby, T. T.; Indris, S.; Knapp, M.; Ehrenberg, H.; Fink, K.; Kruk, R.; Hahn, H.; Advanced functional materials, 26 (41), 7507 (2016).

Location: H 0105