SP 13 Complex oxides, multiferroics and composite multiferroics

SP13 - Parallel Session 1

SP13 - Parallel Session 1

SP13.1.02

The Coulomb based magneto-electric coupling in multiferroic tunnel junctions and granular multiferroics

O. Udalov, I. Beloborodov

Text We study magneto-electric effect due to the Coulomb interaction in two systems: i) multiferroic tunnel junction (MFTJ) - magnetic tunnel junction with ferroelectric barrier and ii) granular multiferroic (GMF) - material with ferromagnetic (FM) metallic grains embedded into ferroelectric matrix. We show that in the absence of spin-orbit or strain-mediated coupling the Coulomb interaction influences the magnetic state of the system in several ways: i) through the spin-dependent part of the Coulomb interaction (exchange interaction); ii) due to virtual electron hopping - the Coulomb blockade effect suppresses electron hopping between grains and therefore magnetic coupling; iii) through image forces and polarization screening that modify the barrier for electrons in MFTJ and GMF - the barrier defines the hopping probability and magnetic coupling. We show that magneto-electric effect appears due to the following reason: The Coulomb interaction depends on the dielectric properties of the system. For GMF it depends on the dielectric constant of FE matrix and for MFTJ on the dielectric constant of the FE barrier. Applying external electric field one can tune the dielectric constant and the Coulomb interaction. Thus, one can control magnetic state with electric field. Here we calculate dependence of magnetic state of GMF and MFTJ on applied electric field and temperature and show that FM order can be induced with electric field.

SP 13 Complex oxides, multiferroics and composite multiferroics

SP13 - Parallel Session 1

SP13.1.03

Mechanism for reversible switching of spin polarization in Co/PZT/LSMO tunnel junction by electric field

N. Stojic, M. Imam, N. Binggeli

Text Recent experiments have shown that in the Co/PZT/LSMO magnetic tunnel junctions (MTJ) it is possible to reversibly switch the sign of the spin polarization of the tunneling electrons by an electric field, indicating technologically-promising new ways of spin transport control in spintronic devices. The spin-polarization inversion effect in this MTJ is expected to occur at the Co/PZT interface due to the half-metallic nature of LSMO. Still, the possible optimization of this effect as well as the application to other MTJ systems is hindered by the lack of understanding of the underlying mechanism. So far, despite the theoretical efforts, the spin inversion with ferroelectric switching has not been explained. We have identified a likely and realistic mechanism explaining the observed ferroelectric switching of spin polarization, using density-functional theory. The explanation is based on an O-vacancy related interface structure, which could be expected at such interfaces. This O-vacancy configuration is singled out after an extensive search of an interface PZT/Co configuration which can account for the experimentally observed trend. The mechanism we find is based on drastic and reversible changes in the reactivity and chemical binding of the interfacial O with Co upon the inversion of the ferroelectric polarization. The ferroelectric-controllable reactivity of O leads virtually to ON/OFF switchable O-Co hybridization and a swappable spin polarization in the PZT barrier.

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SP13.1.04

Low noise all-oxide magnetic tunnel junctions based on a La0.7Sr0.3MnO3 / Nb:SrTiO3 interface

G. Kurij, A. Solignac, T. Maroutian, G. Agnus, R. Guerrero, L. E. Calvet, M. Pannetier Lecoeur, P. Lecoeur

Text Functional oxides have shown a wide range of peculiar physical properties that could potentially be exploited for applications. One example is manganite La0.7Sr0.3MnO3 (LSMO) based Magnetic Tunnel Junctions (MTJs), which benefit from the full spin polarization of manganite and exhibit high tunneling magnetoresistance (TMR) ratios at low temperatures up to several hundred of percent. Such systems can be used in magnetic field sensing for high sensitivity magnetometers. Nevertheless, the noise in this system, measured by our groups, has, until now, been higher than in metallic junctions, thus limiting their usefulness for applications.
In this paper, we demonstrate how the noise level can be reduced by three orders of magnitude and now making these devices competitive with metallic systems. Our work investigates replacing the STO insulating barrier by a semiconducting Nb:STO barrier in LSMO based tunnel junctions.
High quality LSMO(30nm)/Nb:STO(3nm)/LSMO(10nm)/La0.7Sr0.3Mn0.93Ru0.07O3(20nm) junctions were grown in situ by pulsed laser and then patterned into micrometric MTJs.
We explore temperature- and voltage-dependent magneto-transport of LSMO based tunnel junctions with semiconducting Nb:STO or undoped STO barriers. We demonstrate that the junctions with Nb:STO barriers exhibit improved TMR characteristics over a wide range of temperatures, much lower noise levels and significant robustness.

Ref: G. Kurij et al., Appl. Phys. Letters 110, 082405 (2017)

SP 13 Complex oxides, multiferroics and composite multiferroics

SP13 - Parallel Session 1

SP13.1.05

Study on magnetic characteristics modulated by Ferromagnetic/Ferroelectric heterojunction

A. Yamaguchi, Y. Utsumi, K. Yamada, T. Saiki, A. Nakao

Text Control of Magnetization reversal and magnetic domain structure is significant important for application of spintronic devices and systems. One of the interesting candidates, which can achieve the control of magnetization dynamics and domain structures through spin-orbit coupling or magnetoelastic effect induced by the application of electric voltage, is multiferroic material. Heterojunction between ferromagnetic and ferroelectric materials may allow the creation of novel multiferroic material which can work above room temperature.

Here, we investigate the physical mechanism of magnetic anisotropy generated by the heterojunction between Ni layer and single crystalline Y-cut $LiNbO_3$ substrate using magnetoresistance measurement, magnetic imaging using Kerr microscopy, rectifying effect and X-ray photoemission spectroscopy (XPS). Electrical measurement and magnetic imaging techniques reveal the generation of uniaxial magnetic anisotropy in Ni layer on $LiNbO_3$ substrate. In the Ni wires aligned perpendicular to the X-axis of $LiNbO_3$, zebra-stripe domain structure is formed by competition between the shape magnetic anisotropy and additional magnetic anisotropy induced by the heterojunction near the absence of magnetic field. We found that the XPS peak shift of Ni near the interface from the XPS depth profile data. This indicates that the modulation of electronic state distribution near the interface of Ni/LiNbO3 system can generate the uniaxial magnetic anisotropy in the Ni layer.

SP 13 Complex oxides, multiferroics and composite multiferroics

SP13 - Parallel Session 1

SP13.1.06

Magnetoelectric coupling in a layered-perovskite thin film heterostructure

C. Wang

Text In ferroic (ferroelectric, multiferroic) materials and their heterostructures, a large reversible electric-field-driven elastic deformation can provide an effective pathway to achieve the coupling between lattice degree of freedom and other order such as spontaneous polarization, spin, orbital and so on. This is crucial for designing sensing, data-storage and magnetoelectric devices with ultralow energy consumption. Here, we report that reversible in-plane elastic switching with a non-volatile strain of approximately 0.4% can be achieved in layered perovskite Bi2WO6 (BWO) thin films. A magnetoelectric device has been further fabricated in the ferroelectric-ferromagnetic heterostructure demonstrating a new approach to control the low-dimensional spin texture by ferroelastic strain using electric field with low energy consumption.

SP 13 Complex oxides, multiferroics and composite multiferroics

SP13 - Parallel Session 1

SP13.1.07

Strain-induced magnetization control in a LSMO/BTO heterostructure

F. Motti, G. Vinai, A. Petrov, B. Davidson, B. Gobaut, A. Filippetti, G. Rossi, G. Panaccione, P. Torelli

Text Controlling magnetism by using electric fields is a goal of research towards novel spintronic devices and future nano-electronics. For this reason, multiferroic heterostructures attract much interest. Here we provide experimental evidence, and supporting DFT analysis, of magnetic transitions in a $La_{0.65}Sr_{0.35}MnO_3$ (LSMO) thin film (30 unit cells), induced by structural changes the ferroelectric $BaTiO_3$ (BTO) substrate, which can be also induced by applying external electric fields.
Two magnetic transitions versus temperature are observed in LSMO with X-ray Magnetic Circular Dichroism, corresponding to the structural phase transitions of the substrate. We also show that ferromagnetism, absent in the pristine condition at room temperature, can be established by electrically switching the BTO ferroelectric domains in the out-of-plane direction.
These observations can be explained as the effects of a strong interplay between the structural properties of the BTO substrate, resulting in variations of the LSMO thin layer strain, and the magnetic properties of the latter.
Besides these strain-driven phenomena, further experiments will be proposed, focusing on the electrostatic effects driven by the charge balance at the interface and the BTO electric polarization. Finally, limitations and advantages of the different magneto-electric coupling mechanisms in oxide multiferroic heterostructures will be discussed.

SP 13 Complex oxides, multiferroics and composite multiferroics

SP13 - Parallel Session 2

SP13 - Parallel Session 2

SP13.2.01

Magneto-ionic control of magnetism using a solid state proton pump

A. J. Tan, M. Huang, C. O. Avci, M. Mann, H. L. Tuller, G. S. D. Beach

Text In a thin film ferromagnet/oxide heterostructure, the chemical structure at the interface can substantially influence the magnetic properties. This interface can be electrochemically tuned using a gate voltage to produce large changes in the properties. The approach, dubbed magneto-ionic control of interfacial magnetism, has demonstrated modulation of magnetic anisotropy on the order of ~1erg/cm2, >10x larger than any other voltage-induced effect. However, all current magneto-ionic devices are oxygen-based devices; they rely on the redox of the ferromagnet layer to toggle the anisotropy. As a result, they have poor cyclability due to irreversible structural damage to the magnetic layer. In this work, we demonstrate the first hydrogen-based magneto-ionic devices where interfacial magnetism is modulated by hydrogen loading in an adjacent storage layer. This novel approach presents an enormous advantage in protection of the magnetic layer, and we were able to toggle the magnetic anisotropy for >2000 cycles, two orders of magnitude larger than the state-of-the-art magneto-ionic device reported in literature. Besides the cyclability, the device exhibits the fastest room temperature switching.

SP 13 Complex oxides, multiferroics and composite multiferroics

SP13 - Parallel Session 2

SP13.2.02

Soliton excitations observed in multiferroic $LiCuVO_4$

C. Grams, P. Becker, J. Hemberger

Text $LiCuVO_4$ is a single-phase multiferroic that has an orthorhombic distorted inverse spinel structure (space group Imma) with spin ½ Cu chains along the crystallographic [010] axis. The transition into the multiferroic phase was observed at 2.3 K in zero magnetic field and can be suppressed with increasing fields down to 0 K at 7.4 T, hinting at possible quantum criticality at low temperatures. Above 41 T $LiCuVO_4$ enters a spin-nematic phase before it realizes its spin-saturated phase at approx. 44 T.
Another interesting aspect in $LiCuVO_4$ is caused by its ratio of the competing nearest- and next-nearest-neighbor interactions J_1 and J_2. It has been predicted by theory[1] that in this material nearly gapless chirality excitations exist that are described by solitons and should be visible in low frequency dielectric permittivity measurements.
To verify this we performed dielectric spectroscopy measurements with frequencies up to 5 GHz at temperatures down to 0.025 K. Our results show critical slowing down close to the phase transition above 0.4 K as was observed also e.g. in $MnWO_4$ [2]. However, at lower temperatures a slowing down is not observed; instead we see an excitation at 2.9 GHz (12.5 µeV).

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SP 13 Complex oxides, multiferroics and composite multiferroics

SP13 - Parallel Session 2

SP13.2.03

Magnetic order, hysteresis, and phase coexistence in magnetoelectric $LiCoPO_4$

E. Fogh, R. Toft-Petersen, E. Ressouche, C. Niedermayer, S. L. Holm, M. Bartkowiak, O. Prokhnenko, S. Sloth, F. W. Isaksen, D. Vaknin, N. B. Christensen

Text Materials with a coupling between magnetic and electric order parameters have been studied for decades and they offer the prospect of a wide range of technological applications. However, an all-embracing fundamental understanding of these materials is still being pieced together. Our research is centered around the lithium orthophosphates, Li*M*PO_4 (*M* = Mn, Fe, Co, Ni), which all exhibit the magnetoelectric effect in their low-temperature commensurate antiferromagnetic phase. Here,we focus on $LiCoPO_4$ with T_N = 21.7K [1] and which with its special properties stands out of this family of compounds. The magnetic phase diagram was established using neutron diffraction and magnetometry in magnetic fields up to 25.9 T applied along the easy *b* axis [2]. For fields greater than 11.9 T, a short-wavelength cycloid is formed with spins in the (*b*,*c*)-plane. Intriguingly, additional ordering vectors appear for increasing fields in the hysteresis region below the transition field. A simple mean-field model is proposed to explain this behavior. In the field interval 20.5–21.0 T, the period of the magnetic structure remains constant but the spins re-orient compared to the cycloid phase. Above 21.0 T and up until saturation, a commensurate magnetic structure exists with a ferromagnetic component along *b* and an antiferromagnetic component along *c*.

[1] D. Vaknin et al., Phys. Rev. B 65, 224414 (2002)
[2] E. Fogh et al., Phys. Rev. B 96, 104420 (2017)

SP 13 Complex oxides, multiferroics and composite multiferroics

SP13 - Parallel Session 2

SP13.2.04

Field-induced reentrant magnetoelectric phases in $LiNiPO_4$

R. Toft-Petersen, E. Fogh, T. Kihara, Y. Narumi, J. Jensen, J. C. Dyrnum, M. K. Sørensen, J. Lee, M. B Stone, G. E. Granroth, K. Fritsch, M. Bartkowiak, O. Prokhnenko, D. Vaknin, H. Nojiri, N. Bech Christensen

Text Using pulsed magnetic fields oriented along the crystallographic c-axis we have measured the bulk magnetization and electrical polarization of $LiNiPO_4$, and have studied its field-dependent magnetic structures by time-of-flight neutron Laue diffraction. Our data establish the existence of two reentrant high-field magnetoelectric phases (i) between 19 and 21T [1], and (ii) for fields greater than 39T [2], in addition to the known low-field magnetoelectric phase of this compound. Between 21T and 39T, a magnetoelectrically inactive, short-wavelength, incommensurate magnetic structure is identified. We show that we can quantitatively account for the magnetoelectric response between 19 and 21T, and qualitatively for the response above 39T using a simple model for the field-dependent structures. The model takes as its point of departure the known zero-field structure and accounts for the measured field-induced magnetization along c. This strongly suggests that the microscopic mechanism of magnetoelectricity is identical in all three phases.

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[2] E. Fogh et al, To be submitted (2018).

SP 13 Complex oxides, multiferroics and composite multiferroics

SP13 - Parallel Session 2

SP13.2.05

Transition metal valence states and magnetic structure of the frustrated $ABaM_4O_7$ (A=Y,Ca; M=Co,Fe) system

V. Galakhov, D. Turkin, V. Mesilov, S. Shamin, G. Bazuev, K. Küpper

Text The complex oxides of type $ABaM_4O_7$ (A=Y,Ca; M=Co,Fe) are of interest due to a variety of remarkable properties, e.g. the significant electrochemical activity of $YBa(Co,Fe)_4O_7$-cathodes [1,2]. Furthermore, spin assisted ferroelectricity has been found for $CaBaCo_4O_7$ [3], which has been associated with competing exchange interactions [4], which are strongly dependent on the spin and oxidation state of the Co and Fe ions in these compounds. Therefore, we employ a suitable set of structural, magnetic and soft- x-ray absorption approaches to characterize the electronic and magnetic structure of $ABaM_4O_7$ (A=Y,Ca; M=Co,Fe) [5]. We find a spin-glass behavior with negative values of the Curie-Weiss temperatures, indicating predominant antiferromagnetic exchange interactions. Furthermore, we determine the Co and Fe oxidation states from the x-ray absorption spectra.

References
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SP 13 Complex oxides, multiferroics and composite multiferroics

SP13 - Parallel Session 2

SP13.2.06

Correlation of crystalline and magnetic structures of barium ferrites with dual ferroic properties

V. Turchenko, A. Trukhanov, S. Trukhanov, M. Balasoiu, N. Lupu

Text The unique functional properties of barium ferrites and their solid solutions have allowed them to receive a wide technical application in magnetic recording devices, electric motors and as absorber of decimeter and centimeter electromagnetic radiation. However, recently studies of similar materials [1] have found a large ferroelectric polarization at room temperature. Taking into account their high Curie temperature it can be conclude about coexistence of ferroelectricity and ferromagnetism that allow considering these materials as potential multiferroic candidate.
The main aim of present work is ascertain reasons and microscopic mechanism of appearance of double ferroic properties in solid solutions of $BaFe_{12-x}D_xO_{19}$ partially substituted with diamagnetic ions (D= In, Ga and Sc; x= 0.1 – 1.2). The investigation of crystal and magnetic structures of polycrystalline barium ferrites was carried out by neutron di ffraction method in a wide te
The electric field-induced polarization has been found in samples $BaFe_{11.9}D_{0.1}O_{19}$ (D= In and Ga) at room temperature. The refinement of crystal structure has been carried out in frameworks of centrosymmetric SG P63/mmc and non-centrosymmetric SG P63mc. It was shown that shift of iron ion from the local center of octahedrons can be reason of appearance of total nonzero dipole electric moment in the case of non-centrosymmetric SG P63mc.

[1] A.V. Trukhanov, S.V. Trukhanov, L.V. Panina et al. JMMM 426 (2017) 487.

SP 13 Complex oxides, multiferroics and composite multiferroics

SP13 - Parallel Session 2

SP13.2.07

Nature of exchange-bias effect in ferrimagnetic orthoferrites

A. Wisniewski, I. Fita, R. Puzniak, V. Markovich

Text Exchange bias (EB) effect observed in some ferrimagnets (FIMs) exhibiting spontaneous magnetization (*M*) reversal or negative *M* is discussed. Especially interesting finding is that EB of compensated FIMs reverses its sign at crossing the compensation temperature T_{comp}. The EB in single crystal of $ErFeO_3$ emerges in the vicinity of T_{comp} = 45 K, at which the oppositely directed magnetic moments of antiferromagnetically (AFM) coupled Er and Fe sublattices are equal. The novel feature observed is the change of EB sign achieved by varying the field-cooling protocol, depending if T_{comp} is crossed with decreasing or increasing temperature. The origin of EB effect is related to the intrinsic exchange coupling within the unit cell. The negative EB effect occurs in polycrystalline $La_xBa_{1-x}FeO_3$ (x = 0.125, 0.25, 0.33). In low doped samples with $x \leq 0.25$, the EB field H_{EB} increases in absolute value rapidly at small cooling field H_{cool} but it falls abruptly at higher H_{cool}, due to the field-induced ferromagnetism (FM). In contrast, the x = 0.33 sample, with strong AFM constituent, shows no field-induced FM and the H_{EB} vs. H_{cool} dependence is reminiscent of that observed commonly for system of isolated FM clusters embedded in an AFM matrix. Reversed exchange bias effect observed in $LuCr_{0.5}Fe_{0.5}O_3$ is due to the magnetic moment reversal which results from an interplay of various Dzyaloshinskii-Moriya interactions between Fe^{3+} and Cr^{3+} ions.

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SP13 - Parallel Session 2

SP13.2.08

Epitaxial growth of vanadates by ozone assisted Molecular Beam Epitaxy

G. Masset, O. Copie, K. Dumesnil, S. Andrieu

Text Transition metal oxides (TMO) with a perovskite structure have gained much interest owing to the wealth of different functionalities that have emphasized the significant role of the interplay between the charge, spin, orbital and lattice degrees of freedom. In addition to its high technological potential, e.g. in multiferroics, this multifunctional character is a playground for fundamental research and is the basis of the rising field of oxide electronics. Among the TMO family, we study the RVO3 perovskites (R = rare earth or yttrium) that exhibit the prototypical behavior arising from these different couplings. Furthermore, the growth of RVO3/R'VO3 superlattices has been proposed to be ferroelectric by design [1].
As a first step towards the synthesis of PrVO3/LaVO3 superlattices, we investigated the growth of PrVO3 and LaVO3 on (001)-oriented SrTiO3 substrates. The R and V codeposition is performed in a MBE chamber in which a stabilized reactive gas (O3+O2) is introduced. RVO3 epitaxial films of very high structural quality and controlled stoichiometry are obtained with the clear observation of RHEED intensity oscillations during full deposition process. We investigated the structural, chemical and physical properties of our systems, by combining in situ (Auger and XPS) and ex situ (XRD, EDS and HRTEM) experiments.

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SP 13 Complex oxides, multiferroics and composite multiferroics

SP13 - Parallel Session 3

SP13 - Parallel Session 3

SP13.3.01

Coupled spin dimers driven by charge and orbital ordering in an open-shell *p*-electron Rb_4O_6 compound

D. Arcon

Text The Verwey transition in Fe_3O_4, a complex structural phase transition concomitant with a jump in electrical conductivity by two orders of magnitude, has been a benchmark for charge ordering (CO) phenomena in mixed-valence transition metal materials [1]. Here, we demonstrate an archetypical Verwey-type transition in an open p-shell anionic mixed-valence compounds [2]. In Cs_4O_6 a transition from a cubic structure with a single crystallographic site for the molecular O_2^{x-} units to a tetragonal structure with ordered superoxide O_2^- and peroxide O_2^{2-} entities is accompanied by a drastic drop in electronic conductivity and charge fluctuation rates. In a sister compound Rb_4O_6, the analogous CO process is accompanied by molecular *p*-orbital ordering [3,4], which establishes a network of antiferromagnetically coupled O_2^- (S = 1/2) dimers. High-field electron paramagnetic resonance detected direct singlet to triplet transitions and weaker lines attributed to multiple-triplet bound states establishing Rb_4O_6 as a new valence bond solid system. The simple CO pattern of molecular units thus suggest Cs_4O_6 and Rb_4O_6 as model systems for disentangling the complex interplay of charge, lattice, orbital, and spin degrees of freedom in Verwey-type CO processes.

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SP 13 Complex oxides, multiferroics and composite multiferroics

SP13 - Parallel Session 3

SP13.3.02

Electrical gating of GeTe towards non-volatile control of spin physics

S. Varotto, J. Sławińska, S. Cecchi, M. Asa, R. Calarco, S. Picozzi, R. Bertacco, C. Rinaldi

Text Ferroelectric Rashba Semiconductors provide a way for non-volatile fully electric control of spin currents in semiconductors like GeTe, via ferroelectric manipulation of the Rashba physics [1]. Our experimental investigation of GeTe(111) films unambiguously proved that the direction of ferroelectric polarization controls the spin texture of the bulk Rashba bands, as measured by Spin resolved ARPES on two surfaces with inward or outward remanent polarizations [2].
Here we first analyse the electrical gating of GeTe for ferroelectric switching over large areas. Despite the semiconductive behaviour of the material, ferroelectricity can be switched applying voltage pulses to a tunnelling gate electrode, and the written state can be monitored by reading the polarization-dependent resistance of the junction. We relate this dependence to the charge screening within the semiconductor at GeTe/gate interface.
Besides, Unidirectional Spin Hall magnetoresistance measurements [3] in GeTe/Fe heterostructures reveal sizeable Spin Hall Effect [4] and spin injection in the magnetic layer. The ferroelectric control of spin injection through gating would open the perspective of non-volatile control of current-induced magnetization dynamics in GeTe/Fe.

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SP13 - Parallel Session 3

SP13.3.03

Large positive and negative magnetodielectric coupling in Fe half-doped $LaMnO_3$

J. P. Palakkal, R. S. Cheriyedath, P. N. Lekshmi, M. Valant, M. V. Mihelj, M. R. Varma

Text The perovskite $LaFe_{0.5}Mn_{0.5}O_3$ is synthesized by a (citrate-nitrate) combustion method in bulk form. The material shows dielectric relaxation at two distinct temperature zones, whereas both the relaxations follow Arrhenius law. A sufficient coupling between the magnetic and dielectric order parameters in the presence of an external magnetic field offers a large positive and negative magnetodielectric effect for $LaFe_{0.5}Mn_{0.5}O_3$. In addition to the intrinsic magnetodielectric coupling of +4.5% at 85 K, a very large extrinsic magnetodielectric coupling of +45% at 122 K, -18% at 147 K and -6.8% near room temperature is observed for the material in a comparatively low magnetic field of 5 kOe. A magnetoresistance of -5.6% is also found for the material at 100 K under 90 kOe. The influence of magnetoresistance on Maxwell-Wagner-Sillars polarization is the origin of the extrinsic magnetodielectric effects in the material. In the present study, we identified the perovskite $LaFe_{0.5}Mn_{0.5}O_3$ as a candidate for the new set of materials that exhibit both the positive and negative magnetodielectric effects.

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SP13 - Parallel Session 3

SP13.3.04

Magnetic Fluctuations and the Origin of Antiferromagnetism in RENiO3 Perovskites

L. Korosec, D. Gawryluk, M. Pikulski, T. Shiroka, K. Conder, J.-A. Alonso, M. Medarde, H.-R. Ott, J. Mesot

Text The rare-earth nickelate perovskites RENiO3 are an archetypal family of strongly correlated electron systems. By using rare-earths with different ionic radii, one can tune their metal–insulator transition (MIT) temperature from 0 to 600 K while also changing the antiferromagnetic (AF) transition temperature between 0 – 220 K. For PrNiO3 and NdNiO3, the MIT and AF transition occur simultaneously, whereas an intermediate paramagnetic insulating (PI) phase exists for RE = Sm – Lu. LaNiO3 remains metallic at all temperatures. In the insulating phase, the crystal structure consists of alternating large and small octahedra. This has been interpreted as a form of charge disproportionation. The AF order with the unusual propagation vector q = (1/4, 1/4, 1/4) breaks inversion symmetry and is expected to cause ferroelectric polarisation.

We have studied magnetic fluctuations in RENiO3 by means of 17O-NMR relaxation measurements. The relaxation rate is strongly enhanced in the PI phase. This effect is clearly seen both by measuring samples with different RE at constant temperature and in the temperature dependence for fixed composition. This enhancement is most likely caused by fluctuations of the Ni moments at wavevector q = (1/4, 1/4, 1/4) which can occur only in the insulating phase. In LaNiO3, we find Pauli paramagnetic behaviour across all temperatures. Therefore, we conclude that the origin of AF order in RENiO3 lies in the structural and charge modulation of the insulating phase.

SP 13 Complex oxides, multiferroics and composite multiferroics

SP13 - Parallel Session 3

Sp13.3.05

Probing domain dynamics in complex oxide micromagnets using scanning transmission x-ray microscopy

E. Digernes, J. Leliaert, P. E. Vullum, B. Van Waeyenberge, E. Folven

Text Epitaxial complex oxide materials, with a strong link between atomic arrangement and functional properties, are promising candidates for spintronic applications. In a device perspective, understanding the magnetization dynamics is central. Recent development in synchrotron based x-ray transmission techniques makes it possible to assess the magnetic structure with nanometer spatial resolution and sub-nanosecond temporal resolution. However, these techniques have been limited to materials that can be directly deposited on a thin membrane, thus leaving epitaxial systems unavailable. In this study we establish proof-of-concept for scanning transmission x-ray microscopy investigations of complex oxide microstructures and present direct imaging of domain wall dynamics with nanosecond time resolution.

Our model system is micromagnets defined in ferromagnetic $La_{0.7}Sr_{0.3}MnO_3$ and antiferromagnetic $LaFeO_3$ grown on $SrTiO_3$. To enable scanning transmission x-ray microscopy, the microstructured thin films are released as roughly 150 nm thick lamellas using focused ion beam milling and placed on top a Cu strip line defined on a Si_3N_4 membrane. A Landau flux-closure ground state is distorted using field pulses delivered through the Oersted field of the Cu strip line and the relaxation of the domain structure recorded. The presented method is general and enables probing the spin dynamics in a wide group of epitaxial materials.

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SP13 - Parallel Session 3

SP13.3.06

Exploring the magnetic properties of $BiFeO_3$-$PbTiO_3$ based ceramics

A. Kumar, J. Shen, P. Sharma, H. Zhao, Q. Zhengjian, Q. Li

Text $BiFeO_3$ (BFO) is an attractive multifunctional material with novel and interesting physical phenomenon [1]. BFO shows Neel temperature at ~1103 K [2] and G-type antiferromagnetism at ~643 K [3]. BFO with insulating ABO_3 oxides, such as $PbTiO_3$ [4] and $BaTiO_3$ [5], has been proposed as one of possibility to improve the electrical resistance of the material. We report the crystallographic and magnetic behavior of $(1-x)BiFeO_3$–$(x)PbTiO_3$ (x = 0.05, 0.1) ceramics as produced by solid-state reaction route. XRD and Raman spectroscopy analysis of $(1-x)BiFeO_3$–$(x)PbTiO_3$ suggest absence of any structural phase transition and can be indexed with rhombohedral structure (*R*3*c*) and matched well with JCPDS No. 86-1518. Surface morphology was checked by scanning electron microscope (SEM) technique and it has been confirmed that there is a presence of flakes in SEM images. BFO is expected to have thirteen zone-center Raman-active phonon modes which can be summarized using the irreducible representation $\Gamma_{Raman} = 4A_1+9E$ where A_1 modes are polarized along the z-axis and E modes in the x–y plane [6,7]. In $(1-x)BiFeO_3$–$(x)PbTiO_3$ samples, there is existence of 13 Raman active modes. Magnetometry have been performed to investigate the temperature and magnetic field dependence of magnetic phenomenon. M-H loop curves show the antiferromagnetic ordering with weak ferromagnetism though M-T measurement shows a transition at temperature around 150K that need to focus more for understanding of magnetoelectricity.

SP 13 Complex oxides, multiferroics and composite multiferroics

SP13 - Parallel Session 3

SP13.3.07

Magnetoelectric properties of rare-earth langasite $Ho_{0.09}La_{2.91}Ga_5SiO_{14}$

N. Kostyuchenko, A. Mukhin, A. Pimenov, L. Weymann, A. Popov, A. Zvezdin

Text In this work, we investigate both experimentally and theoretically the holmium doped rare-earth langasite $Ho_{0.09}La_{2.91}Ga_5SiO_{14}$. The magnetic and magnetoelectric behavior in single crystalline $Ho_{0.09}La_{2.91}Ga_5SiO_{14}$ along the main crystallographic directions has been studied in the wide range magnetic fields and temperatures.
To explain the observed properties peculiarities we applied a theoretical approach successfully used previously for rare-earth borates [1-4]. Here we extend the quantum theory of magnetoelectric effect in rare-earth langasites to the holmium doped compound. From the comparison of the experimental data and theoretical results we evaluate the crystal-field and the magnetoelectric Hamiltonian parameters. Both magnetic and magnetoelectric properties of the Ho-doped langasite can be successfully explained by the suggested model.

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SP13 - Parallel Session 3

SP13.3.08

Ferroelectric ferrimagnetic LiFe2F6: charge ordering mediated magnetoelectricity

S. Dong

Text Trirutile-type LiFe2F6 is a charge-ordered material with an Fe2+/Fe3+ configuration. Here, its physical properties, including magnetism, electronic structure, phase transition, and charge ordering, are studied theoretically [1]. On one hand, the charge ordering leads to improper ferroelectricity with a large polarization. On the other hand, its magnetic ground state can be tuned from the antiferromagnetic to ferrimagnetic by moderate compressive strain. Thus, LiFe2F6 can be a rare multiferroic with both large magnetization and polarization. Most importantly, since the charge ordering is the common ingredient for both ferroelectricity and magnetization, the net magnetization may be fully switched by flipping the polarization, rendering intrinsically strong magnetoelectric effects and desirable functions.

Reference:
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SP13.4.02

Analysis of magneto-elastic coupling mechanisms in FePt/$BaTiO_3$ bilayers through structural and magnetic characterization

A. Román, A. López Pedroso, L. Neñer, D. Perez, M. Aguirre, M. Soares, M. Sirena, A. Butera, L. Steren

Text The design and fabrication of multiferroics (MF) is one of the main challenges for the development of oxide spintronics. Tuning the magnetic state by electric field or strains appears to be the key for low-energy devices. Magneto-electric and magneto-elastic coupling at interfaces between ferromagnetic (FM) and ferroelectric (FE) layers are at the origin of these phenomena [1]. First-principle calculations revealed recently that important changes of magnetic anisotropy should be observed on FePt/$BaTiO_3$ structures when $BaTiO_3$(BTO) is poled [2]. Moreover, BTO piezoelectric and FePt magnetostrictive properties make FePt/BTO heterostructure a highly tunable multiferoic. To study coupling mechanisms in the FePt/BTO structures, we grew a series of FePt/BTO bilayers by sputtering onto (001) $SrTiO_3$ substrates. The characterization of the FePt/BTO magnetic anisotropy was performed by magnetization measurements, while the domain structure was observed by magnetic force microscopy. The crystalline structure of each constituents was investigated in different geometries and temperatures by X-ray Diffraction using synchrotron radiation at LNLS-Campinas. The analysis of the results, in particular the correlation between crystalline and magnetic properties allows us to get a deep insight into the magneto-elastic coupling mechanisms in this system.
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SP13 - Parallel Session 4

SP13.4.03

Study of the magnetic order in epitaxially strained thin films of multiferroic perovskite Sr1-xBaxMnO3 by using low energy muon spin spectroscopy

P. A. Algarabel, L. Maurel, N. Marcano, E. Langenberg, R. Guzman, R. Guzman, T. Prokscha, C. Magen, J. A. Pardo

Text Perovskite $Sr_{1-x}Ba_xMnO_3$ (SBMO) stands out as a promising candidate for multiferroics due to the expected strong coupling between polar instability, spin order and lattice in these compounds. Moreover, epitaxial films of perovskite SBMO are ideal candidates for tailoring magnetoelectric coupling through the control of Ba-content and epitaxial strain. However, the strong metastable character of the perovskite phase in the SBMO system, relative to its hexagonal phase, has prevented epitaxial SBMO thin films from being synthesized and these properties have remained unexplored. Recently, we have synthesized the perovskite phase in SBMO films with Ba-contents in the range $0.2 \leq x \leq 0.5$ grown on different perovskite substrates. We have used low-energy muon spectroscopy (LE-μSR) to study the dependence on strain and Ba content of the magnetic order temperatures of some selected SBMO thin films, deposited using the pulsed laser deposition technique. By combining the LE-μSR and atomic resolution scanning transmission electron microscopy (STEM) experiments we have related the broadness of the magnetic transition to the existance of a Mn-O-Mn angle gradient. The occurrence of a multiferroic (ferroelectric/ferromagnetic) ground state at high strain levels is proposed for $Sr_{0.8}Ba_{0.2}MnO_3$ thin film deposited on $TbScO_3$. Finally, we demonstrate that the unit cell volume is the key parameter to determine the Néel temperature in $S_{r1-x}Ba_xMnO_3$ thin films showing G-type antiferromagnetic order.

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SP13 - Parallel Session 4

SP13.4.04

Enhanced interface magnetism by structural reconstruction in $La_{0.7}Sr_{0.3}MnO_3$ /$SrTiO_3$ heterostructures

J. I. Beltrán, J. Grandal, J. Tornos, C. León, J. Santamaría, M. Varela, M. C. Muñoz

Text Magnetism in $La_{0.7}Sr_{0.3}MnO_3$/$SrTiO_3$ (LSTO/STO) heterojunctions is intricately linked to the lattice, charge, orbital and spin coupling at the interface. Here we report on the mechanisms underlying interfacial magnetism. We combine first-principles density functional theory (DFT) with advanced electron microscopy techniques to show that the structural reconstruction is an effective mechanism to accommodate the polar, lattice and symmetry discontinuities present at the interface. DFT calculations reveal that the ideal interface shows local structural distortions but no charge transfer to the STO. The out-of-plane lattice constant expands at the interface, contrary to what is expected from elasticity theory for LSMO under biaxial tensile strain, and MnO_6 octahedral rotations are reduced. As a result, the e_g orbital ordering is modified, there is a preferential occupation of $3z^2$-r^2 orbitals and holes deplete at the Mn interface planes, which enhances interface magnetism. Partial occupancy of the Ti d-orbitals is only induced by oxygen vacancies. Atomic resolution images of LSMO/STO epitaxial interfaces grown by high-pressure O_2 sputtering, indeed exhibit an elongated out-of-plane lattice parameter and energy-loss magnetic chiral dichroism evidences the local increase of Mn and Ti interface magnetic moments, in good agreement with calculations. These results point to the structural reconstruction as the significant pathway to compensate the interface discontinuities.

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SP13 - Parallel Session 4

SP13.4.05

Pulsed Laser deposition of oxide thin films on buffered Si(100) substrates

A. Carrero, A. Lo Giudice, A. Roman, S. Carreira, A. Lopez Pedroso, M. Aguirre, L. Steren

Text Nowadays, a huge effort is being made in the integration of oxides to silicon waffers with the aim of improving existing electronic devices or introducing new ones. In parallel, there has been an increasing interest in developing artificial multiferroics, built of ferromagnets and ferroelectrics, to add original functionalities to advanced materials,taking advantages of interfacial couplings. A model system for oxide-based multiferroics is the $La_{0,67}Sr_{0,33}MnO_3/BaTiO_3$ (LSMO/BTO) structure. On one hand, the LSMO is ferromagnet and half-metal while the BTO is ferroelectric at room temperature, having both a perovskite-like structure. However, oxide/silicon structures are still a challenging goal due to the high reactivity of the silicon to oxygen, cationic interdifusion and mismatch of thermal expansion coefficients and lattices. In our work, we show the successfully growth of LSMO and BTO crystalline films and bilayers made by PLD onto Si(100) substrates, using different buffers. An exhaustive structural and morphological characterization of the samples was carried out to analyze their properties. The films´growth was monitored by RHEED while the lattice parameters were determined by XRD. High-resolution TEM was used to examine in detail the quality of the interfaces regarding strains, stacking and composition of the constituents. A correlation of structural, magnetic and ferroelectric properties of the different structures will be discussed in this presentation.

SP 13 Complex oxides, multiferroics and composite multiferroics

SP13 - Parallel Session 4

SP13.4.06

Electrically modulated magnetoelectric composites for sensing small magnetic fields

P. Hayes, S. Salzer, D. Burdin, V. Schell, A. Piorra, Y. Fetisov, R. Knöchel, E. Quandt

Text Magnetoelectric (ME) cantilever-type composites consisting of a piezoelectric (PE) and a magnetostrictive (MS) layer are usually employed for measurements of magnetic fields passively, i.e. an AC magnetic field directly generates a ME voltage by coupling a MS deformation to the PE phase. In order to achieve high sensitivities, mechanical resonances are exploited along with a magnetic bias field, necessary to operate at the maximum piezomagnetic coefficient of the MS phase. Despite being able to detect very small field amplitudes within biomagnetic requirements, one is limited to high frequency magnetic signals coinciding to mechanical resonances.
In the presented work the PE phase is actively excited, similar to [1]. In this case using a high mode mechanical resonance, which leads to voltage induction in a mechanically decoupled pickup coil around the sensor composite. The induced voltage proves to be very sensitive to small DC and AC magnetic fields exceeding 2200 V/T. This entirely passive readout makes ME sensors suitable for integration into arrays.
We demonstrate the performance of electrically modulated thin film ME composites, using 2 µm sputtered AlN and 2 µm amorphous MS film, in conjunction with passive readout using a pickup coil. This strategy enables for DC as well as AC magnetic field detection without the need of an external magnetic AC or DC bias field.
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SP13 - Parallel Session 4

SP13.4.07

Correlation of Oxygen Vacancies and Magnetism in $La_{0.7}Sr_{0.3}MnO_3$ Thin Films in Vertical Memristive Devices

M. Wilhelm, M. Moors, C. Bäumer, C. Wiemann, R. Dittmann, C. M. Schneider, M. Müller

Text Transition metal oxides, like perovskite manganites, are extensively investigated due to their richness of underlying physics and potential technology applications. Special attention is paid to the resistive switching oxide $La_{0.7}Sr_{0.3}MnO_3$ (LSMO), which exhibits a large spin polarization (~95 %) and a temperature (370 K), which makes it a promising candidate for future non-volatile memory applications (MRAM).
In our project we investigate the electronic switching behavior and the correlated magnetism of LSMO using spatially resolved X-ray absorption (XAS) and magnetic circular dichroism (XMCD). The switching process from a high- to a low resistance state is supposed to be driven by a migration of oxygen vacancies (V_O) under high electric fields and temperature gradients. Depending on the applied voltage polarity an accumulation/depletion of oxygen vacancies at the electrode/oxide interface is expected. Since the presence of V_O disturbs the double-exchange mechanism, which is responsible for the conductivity and ferromagnetic exchange of LSMO, this behavior goes along with a local decrease/increase in conductivity. The formation of V_O can be directly derived from the O K-Edge, and indirectly from valence changes apparent in the Mn $L_{3,2}$ absorption spectrum in LSMO.