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ISOE2019

5th International School of Oxide Electronics

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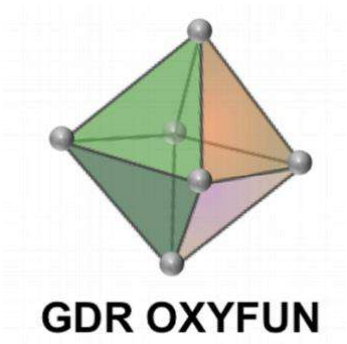

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Abstracts of lectures

A Unique Exploration

Hans Boschker
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Research in an applied and scientifically exciting field such as oxide electronics is relevant for applications as well as fundamental science. But there is even more to oxide electronics. Oxide electronics is a trailblazer in a unique adventure: the pioneering endeavor of opening the entire periodic table to applications in electronic devices. I will discuss some of the merits of the oxides, current challenges, and future limitations of this endeavor.

Relativistic oxide materials

Jeroen van den Brink
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The observed richness of topological states on the single-electron level prompts the question what kind of topological phases can develop in more strongly correlated, many-body electron systems. Correlation effects, in particular intra- and inter-orbital electron-electron interactions, are very substantial in 3d transition-metal compounds such as the copper oxides, but the spin-orbit coupling (SOC) is weak. In 4d and 5d transition-metal compounds such as iridates, the interesting situation arises that the SOC and Coulomb interactions meet on the same energy scale. The electronic structure of iridates thus depends on a strong competition between the electronic hopping amplitudes, local energy-level splittings, electron-electron interaction strengths, and the SOC of the Ir 5d electrons. The interplay of these ingredients offers the potential to stabilise relatively well-understood states such as a 2D Heisenberg-like antiferromagnet in Sr2IrO4, but in principle also far more exotic ones, such a topological Kitaev quantum spin liquid, in (hyper)honeycomb iridates and RuCl3. I will discuss the microscopic electronic structures of these materials, their proximity to idealized Heisenberg and Kitaev models and our contributions to establishing the physical factors that appear to have preempted the realization of quantum spin liquid phases so far.

Introduction to ferroelectrics

Gustau Catalan

ICREA and Institut Català de Nanociència i Nanotecnologia (ICN2), Barcelona, Catalonia

The aim of this lecture is to lay down clear concepts for understanding and manipulating ferroelectrics. Starting with basic notions and common applications, I will try to develop ferroelectricity all the way up to the point where it branches into emerging areas of research. The idea is to set the students on a robust foundation to develop their own research with a confident knowledge of what ferroelectrics are and can (or cannot) do, and the ability to identify common mistakes.

At the theoretical level, the lecture will cover the essentials of Landau theory, depolarization fields, phonon freezing. At the experimental level, we will discuss the main tools for characterizing ferroelectrics (hysteresis loops, dielectric measurements, piezoresponse force microscopy), as well as their artefacts. The lecture will end with a listing of trending topics in ferroelectrics, which will include the bulk photovoltaic effect (the ability of ferroelectrics to generate photovoltages much bigger than their bandgap), the electrocaloric effect (the ability of ferroelectrics and antiferroelectrics to heat up or cool down as a voltage is applied to them), domain wall physics, and flexoelectricity.

Introduction to Magnetic Oxides

Michel Viret

Service de Physique de l'Etat Condensé, DRF/IRAMIS/SPEC, CEA Saclay, France

In this lecture, I will review the general magnetic properties of oxide materials. I will recall the basic rules for magnetic ordering and in particular the indirect exchange interactions through the oxygen. I shall underline the role of lattice distortions and charge and orbital ordering. I will illustrate the basic rules taking as two examples the manganites and the multiferroics. In the first family, I will present the correlation between magnetic properties and electrical transport introducing the colossal magnetoresistance effect. Transport properties will be illustrated through the models of magnetic localization. The problem of phase separation in these materials will be surveyed with a critical eye. Other metal/insulator transitions will be addressed especially in the antiferromagnetic nickelates. I shall then briefly illustrate the revival of the multiferroics through the example of $BiFeO_3$. A particular emphasis will be given to its magnetic properties and their correlation to ferroelectricity. Finally, I will underline the relevance of magnetic oxides to the field of spintronics with a particular emphasis on antiferromagnets and multiferroics.

Modern theory of ferroelectrics

David Vanderbilt

Department of Physics and Astronomy, Rutgers University
Piscataway, New Jersey, USA

Ferroelectrics are materials in which electric dipole moments spontaneously appear and order to generate a macroscopic electric polarization, in analogy to the spontaneous magnetization of ferromagnets. Because ferroelectric domains can be read and written electrically, they have potential advantages for future electronic memory technologies. Ferroelectrics are typically also piezoelectrics, i.e., materials in which strains induce electric charges and vice versa; such materials are in widespread use in transducer applications ranging from medical ultrasound to marine sonar.

In this talk, I will review the computational tools that have been developed in the materials theory community for the first-principles calculations and predictions of the properties of ferroelectric materials. I will briefly review the modern theory of polarization; effective Hamiltonian and classical atomistic approaches for finite-temperature and domain-evolution behavior; the theory of piezoelectricity; and current work on the theory of flexoelectricity (polarization response to a strain gradient).

In the last part of the talk, I will discuss some recent theoretical attempts to propose or investigate new classes of ferroelectric materials.

Spinorbitronics

Nicolas Reyren

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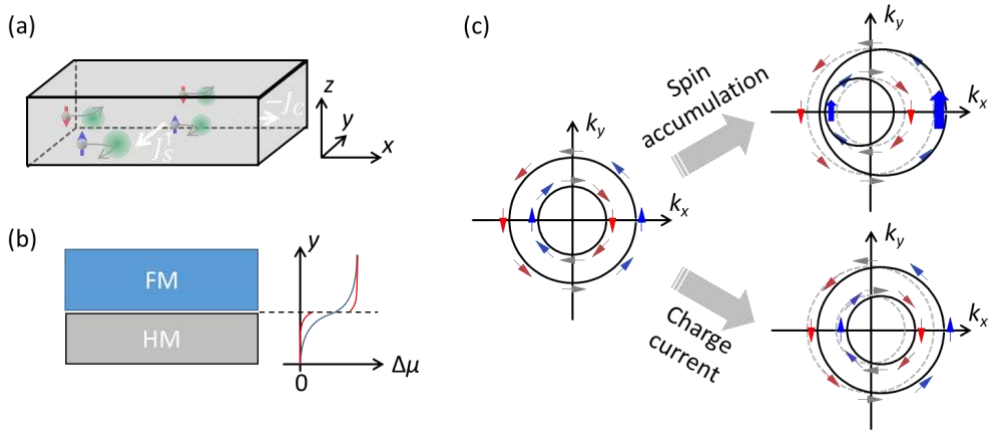
Spintronics evolves along new paths involving non-magnetic materials having large spin-obit coupling (SOC) to generate or detect (pure) spin currents, previously generated by electrical current polarization through a ferromagnetic layer. SOC is a relativistic correction of the quantum mechanics, which considers the magnetic field emerging in an electron rest frame when it moves in electric fields, effectively coupling its spin to its motion ("orbit").

Large SOC can typically be found in 5*d* metals or in peculiar surface states, allowing for example large spin-to-charge current conversion (spin Hall and (Rashba-)Edelstein effects). These heavy metals or these peculiar surface states have other effects: in proximity of magnetic thin films they can burst out the Dzyaloshinskii-Moriya interaction (DMI) leading to the stabilization of chiral magnetic structures (*e.g.* Néel domain walls). Another source of recent interest relies on "non-trivial topologies" resulting from the SOC, either of the band structure (*e.g.* topological insulators), or of the spin textures (see "Skyrmion" talk on Friday 28th).

In this lecture, we will first review the different sources of pure spin current, *i.e.* a spin current which is not accompanied by a charge current (at least not in the same direction), and some of the techniques that can be used to estimate its magnitude. We will concentrate on charge to spin current conversion and discuss the efficiency of the different mechanisms.

In a second part, we will describe how spin currents can be used to move magnetization textures (*e.g.* chiral domain walls) or to switch nanomagnets (*e.g.* MRAM elements). Because the details of the spin textures are crucial to understand their motion, we will start this second part by a brief introduction about the DMI, a SOC-induced antisymmetric exchange.



Different mechanisms related to spin-orbit coupling. (a) *The extrinsic spin Hall effect generates a pure spin current (along y) perpendicular to a charge current (along x). The small grey spheres symbolize the electrons with a red/blue arrow symbolizing their spin.* ***(b)*** *SOC at interfaces is responsible for reduced spin current transmission and discontinuity of the spin accumulation (blue curve without SOC at interface, red with SOC).* ***(c)*** *In a Rashba system, spin and momentum are locked: a charge current is generated by a spin accumulation and reciprocally a spin accumulation is generated by a charge current.*

Crystal symmetry

Béatrice GRENIER
Univ. Grenoble Alpes, CEA, IRIG-DEPHY-MEM-MDN, Grenoble, France

This lecture [1,2] will focus on the description of a crystal, which relies on two fundamental symmetries: point group (i.e. orientation) symmetries and translation symmetry. The final aim will be to understand and use the information contained in the space group of a crystal, as given in the International Tables for Crystallography [3]. Throughout this lecture, the various concepts will be exemplified through a few selected examples of oxide compounds and some useful websites will be presented. This lecture will be organized in three parts, as described hereafter.

In a first part, the various elementary point group symmetries (inversion, rotation, reflection, and roto-inversion) are first recalled. Combining the point symmetries that are compatible with the translation symmetry yields the 32 crystallographic point groups, among which 11 (called the Laue classes) possess inversion. Their description and their Hermann-Mauguin symbol (international notation) are explained, and further illustrated through examples of molecules. Last, the relation between the knowledge of the point group and the prediction of physical properties is exemplified in the case of dielectric properties.

In a second part, the notion of lattice and motif, based on the translation symmetry, is recalled, together with important concepts such as that of the unit cell and its multiplicity. After a classification of the various unit cells into 6 conventional cells or 7 crystal systems, based on their point symmetry, the various manners to center a unit cell are depicted, and the associated symbol for the corresponding lattice type is given. In the end, this yields a classification into 14 Bravais lattices (6 primitive ones and 8 centered ones). Then, some concepts very useful for diffraction (topic of my second lecture) are briefly recalled: lattice directions and net planes, reciprocal lattice.

The last part is devoted to space group symmetries. In some cases, point symmetries alone do not allow to obtain the perfect coincidence of the crystalline edifice with itself, and one has to combine them with fractional translations, that is, translations acting inside the unit cell. After presenting the two kinds of such non symmorphic symmetries (glide planes and screw axes), all the existing ones and the symmorphic ones (i.e. the point group symmetries) are reviewed with the information on their graphical and printed symbols. These symmetries act inside the motif and, combined to the lattice translations, yield the space group of the crystal (there are 230 in total), for which the Hermann-Mauguin symbol is also explained. The example of space group $Pnma$ is then discussed, based on the information found in [3], with a particular focus on the asymmetric unit and the Wyckoff sites.

[1] See a transcript of a similar lecture given at the thematic school "Contribution of symmetries in condensed matter" (Giens peninsula, 2009) in: Chapter 6 of "Contribution of symmetries in condensed matter", Edited by B. Grenier, V. Simonet, and H. Schober, EPJ Web of Conferences, Volume 22 (2012),
website: https://www.epj-conferences.org/articles/epjconf/abs/2012/04/contents/contents.html

[2] See slides and video of a similar lecture (lecture I) given at a school of GDR MEETICC (Banyuls, 2018)
website: http://gdr-meeticc.cnrs.fr/ecole-du-gdr-meeticc-school_v3/

[3] International Tables For Crystallography, Volume A, Space-group symmetry (Kluwer Academic Press, 5th ed., 2002), website: http://it.iucr.org/

Oxides for integrated electronic or photonic technologies

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Research on oxide materials has been since long a field of intense investigations, in particular for Information and Communication Technologies. This area of research has been driven since the beginning from both a scientific and a technological perspective. In the initial phase 30 years ago, the scientific community nucleated around the research on high-Tc superconductivity, and since then expanded towards other materials and challenges. In a more recent second phase, substantial work has been dedicated to the exploitation of oxide's superior properties in *real* technologies

The quest for a replacement gate dielectric in field effect transistors has been a powerful driver to progress in this direction. Replacing SiO_2 at the heart of Metal-Oxide-Semiconductor Field-Effect-Transistors was a major challenge for the microelectronic industry. To solve this challenge, disruptive approaches have been considered, e.g. with single crystalline oxides directly on semiconductor surfaces. My presentation will review the incentives to develop such technologies, highlight the major challenges and achievements, and give a perspective on the need for new materials and devices for future microelectronic technologies. However, the combination of silicon microfabrication techniques with the capability to grow crystalline directly on silicon opens up perspectives for completely different fields, as for example in integrated photonics.

Photonics has been the backbone for long range data communication since many years. The need for bandwidth droves the development towards a tighter integration of electro-optical systems, and silicon photonics became the baseline technology. As a key asset for the future of information and communication technology, silicon photonic integrated circuits will greatly benefit from the integration of novel materials. Several oxide materials have very interesting optical properties and can nowadays be integrated with silicon while maintaining their superior optical characteristics. Polar materials such as ferroelectric oxides fall into this category. Such materials are extremely relevant because they are already in use as discrete components for long range communication, e.g. using $LiNbO_3$ for electro-optical modulators. However, unless one can integrate such materials into silicon photonics, modulators will be limited to the use of the plasma dispersion in silicon. I will then also review recent work accomplished to integrate $BaTiO_3$ in various photonic devices monolithically integrated with a silicon photonic platform.

Flexoelectricity

Gustau Catalan

ICREA and Institut Català de Nanociència i Nanotecnologia (ICN2), Barcelona, Catalonia

Flexoelectricity is a coupling between electric polarization and strain gradient. In layman's terms, it's the ability of materials to generate a voltage upon bending –or, conversely, to bend when a voltage is applied to them. By symmetry, it is allowed in ALL materials. In practice, in order to generate measurable flexoelectric effects one needs very large strain gradients and/or very large flexoelectric coefficients, which are not so easily encountered.

In recent years, flexoelectricity has gone from an obscure academic curiosity to something approaching the mainstream, thanks to the discovery that (i) enormous strain gradients can be achieved easily at the nanoscale, and (ii) besides "conventional" flexoelectricity, there are several other mechanisms that can provide flexoelectric-like responses with coefficients far exceeding the classical limits. The universality of the phenomenon is also contributing to its popularity, having now been identified in dielectrics, semiconductors, electrets and even biomaterials such as bones. A final and very important feature of flexoelectricity is that its symmetry constraints are very different from those of piezoelectricity, so it can act not just as a substitute for it but in fact it can enable new physical responses that cannot be achieved by any other means.

In this lecture, I will describe what is flexoelectricity, in what materials is it largest and why. I will also explain why the symmetry of flexoelectricity enables fundamentally new physical effects, and give real examples of such phenomena.

Pulsed Laser Deposition for Advanced Thin Film Technology

Prof. dr. Mark Huijben
MESA+ Institute for Nanotechnology, University of Twente

Epitaxial engineering has proven to be a successful tool for achieving advanced functional properties in complex oxide thin films, which cannot be obtained in single crystals or polycrystalline samples. Since the successful synthesis of a high temperature superconducting thin film in 1987, pulsed laser deposition (PLD) has emerged as a versatile technique for the deposition of high-quality (epitaxial) thin films from a wide variety of complex oxide materials, including superconductors, metals, ferroelectrics, ferromagnets, dielectrics and their multilayers. The PLD technique, used broadly in research laboratories worldwide, is conceived as relatively simple, mainly because of the fact that the heating source for evaporation (ablation), i.e. a powerful laser, is located outside the process chamber. Although PLD derives its popularity thanks to its versatility, the actual processes and mechanisms that take place are highly dynamic and non-trivial.

In this lecture I will start from the basic principles of the PLD deposition process, and subsequently go into its advanced application for the realization of highly controlled ultrathin films, multilayers and superlattices. I will describe several interesting complex oxide materials systems in which control on the atomic scale is crucial to enable interface engineering for enhanced material properties. The successful realization of reconfigurable complex oxide thin films critically depends on the ability to control the deposition with high precision. I will describe the current status of in situ diagnostic tools such as Reflective High-Energy Electron Diffraction (RHEED) and Light-Induced Fluorescence (LIF).

- G. Koster, M. Huijben, A. Janssen, G. Rijnders, Chapter '*Growth studies of heteroepitaxial oxide thin films using reflection high-energy electron diffraction (RHEED)*', Book 'Epitaxial Growth of Complex Metal Oxides', Woodhead Publishing (2015).

- G. Koster, M. Huijben, A.J.H.M. Rijnders, *Chapter 'Oxide superlattices by PLD: A practical guide*', Book 'Metal Oxide-Based Thin Film Structures', Elsevier (2018).

- G. Koster, M. Huijben, G. Rijnders, '*Perspectives for applications of ultimate (atomic) control of oxide films using PLD*', as part of M. Coll et al., 'Towards Oxide Electronics: A Roadmap', Applied Surface Science 482, 1–93 (2019).

Theory of topological materials

David Vanderbilt

Department of Physics and Astronomy, Rutgers University
Piscataway, New Jersey, USA

I will begin by introducing the basic electronic structure concepts behind topological insulators (TIs), beginning with an elementary discussion of Berry phases and curvatures. I will then discuss the quantum anomalous Hall (QAH) phase, and use this as the basic building block for arriving at an understanding of time-reversal invariant 2D ("quantum spin Hall") and 3D ("strong" and "weak") topological insulators. The connection to edge and surface states will be emphasized. I will also mention other classes of topological materials, including topological crystalline insulators and Weyl semimetals. In the last part of the talk I will review some of the experimental realizations of these topological phases, and highlight some theoretical efforts to identify new and better topological materials.

Skyrmions

Nicolas Reyren

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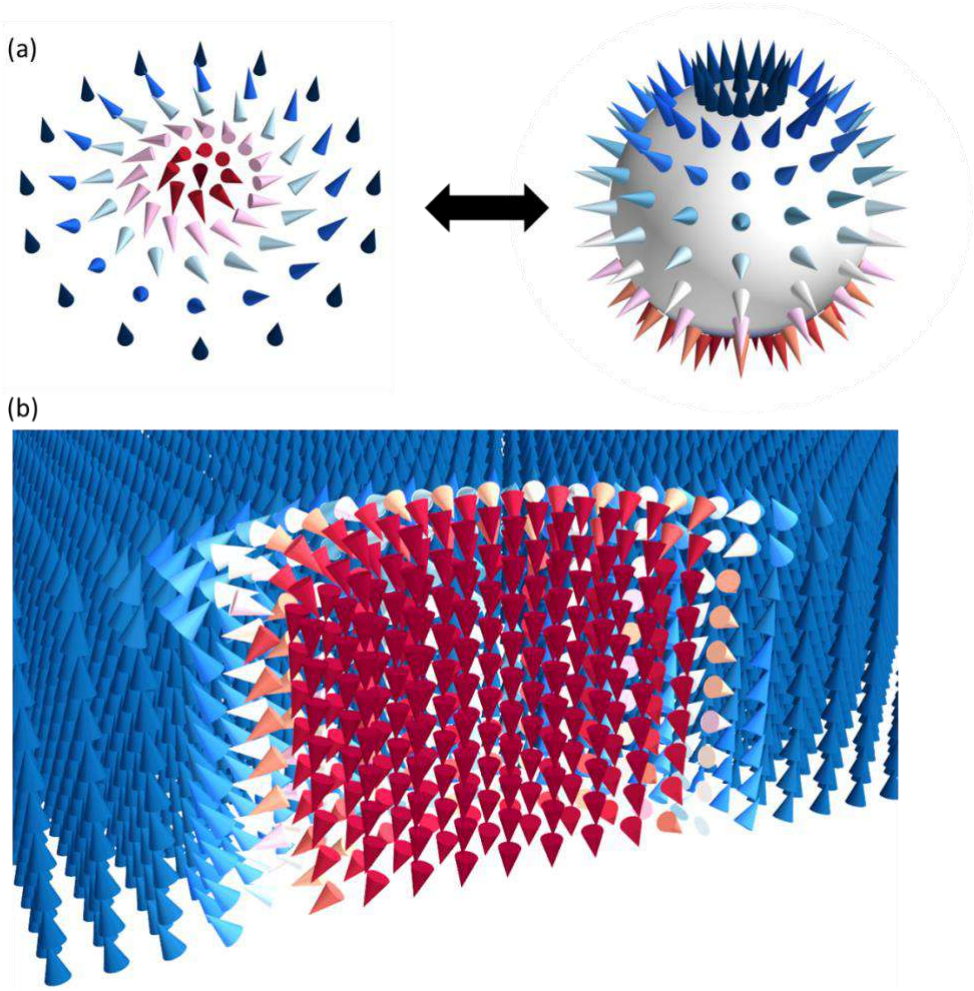
Magnetic skyrmions are magnetic solitons, with a spin texture topologically different from the uniform ferromagnetic state, holding a lot of promise for applications as well as of fundamental interest. They have been observed for the first time in magnetic multilayers at room temperature only a few years ago. The topology of these magnetic objects imposes peculiar dynamics, interesting both in fundamental and applied perspectives.

After a general introduction (definition, general properties), we will explore skyrmions in more details for the case of metallic multilayers based on the Co|Pt interface. We will discuss in particular the experiments of electrical nucleation, propagation, and detection. These are essential steps for devices, but they also enlighten several fundamental aspects. We will in particular discuss the three-dimensional magnetic textures of the skyrmions in magnetic multilayers, differing for the naive two-dimensional picture, with important implications for their manipulation.



Magnetic Skyrmions. (a) *The skyrmions have a peculiar topology: their spins cover the 4π direction of space and turn with a given chirality.* ***(b)*** *Skyrmions in multilayers can have more complex textures with hybrid chirality, as illustrated by this cross-section.*

X-ray and neutron diffraction

Béatrice GRENIER
Univ. Grenoble Alpes, CEA, IRIG-DEPHY-MEM-MDN, Grenoble, France

This lecture [1] will focus on single crystal diffraction, first for the determination of a crystallographic structure, using X-rays or neutrons, second for the determination of a magnetic structure, using neutrons.

In the first part, a general introduction on X-ray and neutron diffraction is given. While X-rays interact with the electronic cloud, neutrons interact with the nucleus, yielding some significant differences between both radiations and thus making them complementary. Once this difference pointed out, both are treated together to discuss the diffraction condition (directions of the diffracted beams) and the diffracted intensities (through the structure factor). The impact of the crystal symmetries on the diffraction pattern is then focussed on: the symmetry of the diffraction pattern is that given by the Laue class of the crystal, while the use of a centered unit cell and the presence of non symmorphic space group operations both yield a systematic absence of intensity on some particular reflections, which is referred to as extinction rules.

In the second part, magnetic neutron scattering is considered. The propagation vector formalism is first explained and illustrated through various types of magnetic structures. A propagation vector $\vec{k}$ is analogous to the wave vector of a plane wave: It reflects the periodicity of the magnetic structure and the direction in which it propagates, and thus it gives the information on where the magnetic intensity is observed. The magnetic structure factor is then introduced and its peculiarities, coming from the nature of the magnetic (dipolar) interaction, are detailed.

Last, the most widely used single crystal X-ray and neutron diffractometers are presented. A few experimental examples on oxide materials are then shown, with a focus on the determination of the magnetic structure, including a brief discussion on group theory representation. At this occasion, various useful softwares and websites are also mentioned.

[1] See slides of a similar, but much more detailed, lecture (lectures II and III) given at a school of GDR MEETICC (Banyuls, 2018); website: http://gdr-meeticc.cnrs.fr/ecole-du-gdr-meeticc-school_v3/

Introduction to Multiferroics

Michel Viret

Service de Physique de l'Etat Condensé, DRF/IRAMIS/SPEC, CEA Saclay, France

Multiferroics are materials with a coexistence of magnetic and ferroelectric order. Although their discovery dates from the 1950's, they have seen an impressive revival in the past 15 years, mainly driven by the possibility of an efficient route for the control of magnetism by electric fields. In recent years, key discoveries in theory, synthesis and characterization techniques have boosted the field. In this lecture, I will present the different mechanisms of multiferroicity, such as lone-pair, geometric, charge-ordering and spin-driven effects. I will put a particular emphasis on the archetype compound $BiFeO_3$ and its relevance to different fields, including ferroelectricity, spintronics and topology. The spatial and time-scales for these properties will also be discussed to better illustrate the great potential of these materials.

Enhanced Lithium Transport in Epitaxial Thin Films for Lithium-ion Batteries

Prof. dr. Mark Huijben,
MESA+ Institute for Nanotechnology, University of Twente

Lithium-ion batteries are the most popular rechargeable batteries nowadays, as they have become the main power source for many applications, such as portable electronics, power tools, and hybrid/full electric vehicles. None of the current rechargeable batteries can fully satisfy all the challenging requirements for our current energy storage. Although tremendous research effort has been devoted to investigate the electrochemical performance of a wide variety of active lithium-based materials, current rechargeable batteries exhibit energy density, lifetime and safety still far below their theoretical capabilities.

Essential for all high-performance energy applications are processes that happen at the interfaces between the different components. Key problems include slow electrode process kinetics with high polarization and low ionic diffusion or electronic conductivity, particularly at the electrode-electrolyte interfaces. Epitaxial engineering is used to control the crystal orientation of electrode thin films, which enables a unique insight into the relation between electrochemistry and crystal directionality of such chemically complex inorganic interfaces, not obtainable in single crystals or polycrystalline samples.

In this lecture I will show the lithium diffusion behavior in $LiMn_2O_4$ cathode, and $Li_4Ti_5O_{12}$ anode, thin films, which are epitaxially grown by pulsed laser deposition on single crystalline Nb-doped $SrTiO_3$ substrates. Control over the specific crystal orientation of the full thin film enables detailed analysis of the lithium diffusion along specific crystal planes ({001}, {110} and {111}). Single phase films show enhanced cyclability and faster charging speed, as compared to studies on polycrystalline materials. The achieved capacity reached >90% of the theoretical limit and minimal capacity reduction was observed when measured over 1000 cycles.

- T.A. Hendriks, D.M. Cunha, D.P. Singh, M. Huijben, '*Enhanced Lithium Transport by Control of Crystal Orientation in Spinel $LiMn_2O_4$ Thin Film Cathodes*', ACS Applied Energy Materials 1, 7046–7051 (2018).

- D.M. Cunha, T.A. Hendriks, A. Vasileiadis, C.M. Vos, T. Verhallen, D.P. Singh, M. Wagemaker, M. Huijben, '*Doubling Reversible Capacities in Epitaxial $Li_4Ti_5O_{12}$ Thin Film Anodes for Microbatteries*', ACS Applied Energy Materials, DOI: 10.1021/acsaem.9b00217 (2019).

Ferroelectric Domain Walls – Physics and Function

Lane W. Martin

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Domain walls are a naturally occurring, essential feature of ferroic materials. The macroscopic manifest of properties in ferroic materials is as much a function of the domain structure as it is the fundamental order parameter. Said another way, the presence of domains and domain walls can, and often does, have a profound impact on the evolution of structure and properties of ferroic materials. As such, understanding how and why these features form, how they respond under applied stimuli, and how and why they can exhibit different properties from the bulk of the material surrounding them is essential to enable the control of modern materials and next-generation devices.

In this lecture we will focus on the physics and function of domains and domain walls in ferroelectric and multiferroic materials. We will begin with an exploration of the energetics of domain-wall formation and will explore their structure and coupling with the electrostatic, elastic, etc. boundary conditions of the material. From there, we will explore how to characterize domain structures and domain walls (with, for example, scanning-probe microscopy, X-ray-based techniques, transmission electron microscopy, *etc*.). In turn, we will investigate how to engineer domain structures in as-grown materials (via approaches such as thin-film epitaxy, heterostructuring, strain gradients, electrical boundary conditions, *etc*.) and how *ex post facto* processing techniques (including, for example, scanning-probe microscopy) can be used to create domain structures in an on-demand fashion. Armed with the ability to produce and characterize such domains, we will proceed to explore how domains and domain walls can give rise to marked contributions to macroscopic responses (*e.g.*, dielectric, piezoelectric, pyroelectric, *etc*.) and exotic nanoscale function (*e.g.*, domain-wall conduction, magnetism, exchange bias, *etc*.). Finally, we will touch on the future of domain walls including aspects of probing domain-wall dynamics at the ultrafast time scale, potential for roles in devices, and much more.

Quantum mechanical shift current

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We discuss a novel manifestation of quantum mechanical current flow in solids upon photoexcitation. From old days, bulk photovoltaic effect has been known to exist in non-centrosymmetric crystals such as poled ferroelectrics exemplified by $BaTiO_3$ [1]. Naive explanation was that the drift current flows due to electric field uncompensated by insufficient formation of electric double layer on the surfaces of polar crystals. Now, it is proposed and confirmed that a quantum mechanical effect, described by the Berry's connection of Floquet bands, drives photocurrent called "shift current" as a second order optical process [2, 3]. We present experimental observations of photovoltaic effect in such polar materials systems as $LaFeO_3/SrTiO_3$ interfaces [4], a ferroelectric organic TTF-CA [5], and a polar semiconductor SbSI [6]. Ultrafast THz spectroscopy [7] and device physics [8] studies have elucidated interesting features of the shift current. This phenomenon is one of the most important topics of topological electronics [9].

[1] W. T. H. Koch et al., Ferroelectrics **13**, 305 (1976).
[2] S. M. Young, M. Rappe et al. Phys. Rev. Lett. **109**,116601 (2012).
[3] T. Morimoto, N. Nagaosa Science Advances **2**, e1501524 (2016).
[4] M. Nakamura et al. Phys. Rev. Lett. **116**, 156801 (2016).
[5] M. Nakamura et al. Nature Commun. **8**, 281 (2017).
[6] N. Ogawa et al. Phys. Rev. B (R) **91**, 241203 (2017).
[7] M. Sotome, N. Ogawa, et al. PNAS **116**, 1929 (2019).
[8] M. Nakamura et al. Appl. Phys. Lett. **113**, 232901 (2018).
[9] Y. Tokura, M. Kawasaki, N. Nagaosa Nature Physics **13**, 1056 (2017).

Exotic Polar States – Rewriting What is Possible in Ferroelectrics

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In recent years, rapid advances in the computational design of materials have been melded with innovative new synthesis approaches and ever-improving characterization methods that are rewriting the types of materials and properties we can realize. Application of these approaches to ferroelectric and multiferroic materials, for example, has resulted in a barrage of new ideas, phenomena, and exotic effects. This work has built from advances in thin-film epitaxy which have enabled the production of model versions of these complicated materials and the subsequent deterministic study of the structure and field-dependent response. But at the same time, advances in material heterostructuring, superlatticing, nanostructuring, *etc*. are giving rise to new ways to control materials which are producing novel structures with the potential to greatly enhance sought after electric field, stress, temperature, *etc*. susceptibilities (*i.e.*, dielectric, piezoelectric, pyroelectric, and electrocaloric effects).

In this lecture, we will focus on the evolution of such exotic polar order. In this process we will explore a number of routes to push the boundaries of modern thin-film strain to control materials including the use of film orientation, compositional and strain gradients, superlattice engineering, and more. In turn, we will explore how theory/modeling, synthesis, and characterization have had to evolve together to rethink what is possible and find new pathways forward in this space. We will see how such approaches can produce exotic domain structures that enable novel switching phenomena that go beyond binary function in ferroelectrics to produce deterministic multi-state function that could potentially be used to emulate neuron function. At the same time, we will explore how to produce novel domain architectures that combine aspects of ferroelectric and ferroelastic response together to enable new types of switching and electromechanical response. From there, we will explore how advances in superlattice growth are now producing novel states of matter including polar vortex and skyrmion structures. Throughout this discussion, we will highlight the need to leverage diverse computational and experimental expertise to address these challenging scientific programs and what the future holds for these systems.

Multiferroics from First Principles

Silvia Picozzi
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In this lesson, I will give an overview on the vaste phenomenology offered by multiferroics and magnetoelectrics, both in the bulk phase as well as in oxides-based junctions, focusing on the microscopic mechanisms driving multiferroicity and magnetoelectricity. In addition, I will briefly discuss how these complex materials can be modelled, showing some paradigmatic examples where theory and experiments gave a successful interpretation of the physics at play in relevant multiferroics.

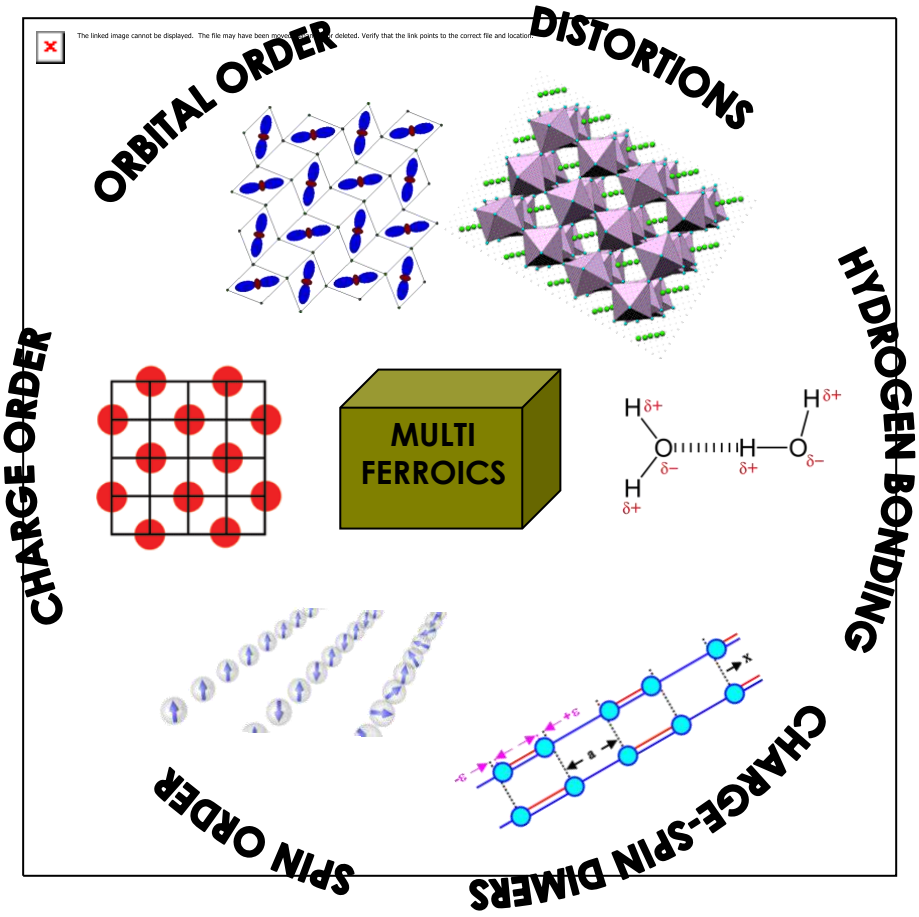

Figure 1: Pictorial representation of the different microscopic mechanisms that can lead to multiferroicity (adapted from S. Picozzi and A. Stroppa, Eur. Phys. J B 85, 240 (2012))

Unconventional Hall effects with topological origin

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Ordinary and anomalous Hall effects have been studied for long time. There has been a rapid progress in understanding the origin of anomalous Hall effect (AHE) [1]. Also to be noted are such unconventional Hall effect discovered in various compounds originating in topology of band structure (momentum space) and spin texture (real space). A pioneering example for the former was found in perovskite $SrRuO_3$ [2], where band crossing point provides magnetic monopoles in momentum space giving rise to AHE. For the latter, scaler spin chirality in non-coplanar spin structure in pyrochlore $Nd_2Mo_2O_7$ was shown to contribute huge AHE [3] that has been now called as topological Hall effect (THE). These discoveries triggered intensive research on the unconventional Hall effect, extending the materials system and variety of microscopic origins. For the former momentum space, semiconducting $EuTiO_3$ was found to be a prototypical system that has rather simple band structure with tenability of Fermi energy position relative to the band crossing point [4,5]. Such strategy to play with band crossing point (or Weyl node) has been extended to various compounds such as magnetic topological insulators, Weyl semimetals, etc. For the latter, THE emerged in spin swirling texture called as skyrmion has been intensively studied. In oxide thin films, $SrIrO_3/SrRuO_3$ [6,7] utilizing interface Dzyaloshinskii-Moriya interaction and $(LaSr)(MnRu)O_3$ [8] utilizing dipolar interaction are prototypical systems. This phenomenon is one of the most important topics of topological electronics [9].

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2D superconductivity

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Nature provides several examples of two-dimensional electron systems that are superconducting. I will discuss why interface superconductivity is a promising area of research, an historical overview of the topic, the control of superconductivity by electrostatic fields, and the possibility of novel superconducting devices.

Oxide multicalorics

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Cooling is essential for food, medicine, electronics and thermal comfort of people in houses and cars, but existing technologies for refrigeration and air-conditioning are based on the compression and expansion of gases that are harmful for the environment.

Cooling using solids is therefore attractive but thermoelectric cooling based on the Peltier effect, and optical cooling based on anti-Stokes fluorescence, are at best only ~10% efficient. By contrast, magnetocaloric, electrocaloric, and mechanocaloric cooling based on thermal changes produced in magnetically, electrically, and mechanically responsive oxides when subjected to changes in magnetic field, electric field and mechanical field promise higher efficiencies.

In this lecture, I will:

- describe the fundamentals of caloric oxides from a historical perspective
- give an overview of their measuring techniques
- present recent advances on magnetocaloric, electrocaloric and mechanocaloric oxides
- and describe recent developments on cooling devices that are based on these materials.

Optical Second Harmonic Generation

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Second harmonic generation (SHG) is the first order of various non-linear optical processes in which two photons of energy $\hbar\omega$ convert into one of energy $2\hbar\omega$. This process is allowed in media showing space and/or time symmetry breakings. The analysis (intensity, polarization dependences etc…) of SHG provides a favored access to complex and entangled ferroic orders in bulk materials as well as in thin layers. In particular, SHG is a very powerful technique to reveal particularly "silent" orders such as antiferromagnetism. In this presentation, after reviewing some fundamentals of SHG, I will provide an overview of its potential by discussing several SHG studies.

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Advanced scanning probe microscopy

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Scanning probe microscopy (**SPM**) offers a variety of different techniques using a physical probe that scans a sample to acquire surface information with nanoscale resolution. This lecture will give an overview of established methods and new developments in this field.

It started in 1981 with the invention of the scanning tunneling microscope (**STM**) by Binnig and Roher[1] (who received the Nobel Prize in Physics in 1986) allowing for the first time imaging of individual atoms. STM is based on the tunneling quantum mechanical effect between the tip and sample and works for conducting surfaces. Whenever the tip-sample distance is small enough that such a tunnel current can flow, significant forces also act between the atoms of the tip and of the surface. It was soon realized that these forces can be put to good use, and the first atomic force microscope (**AFM**) was born in 1986[2], allowing imaging of insulating samples as well.

These developments opened the route towards new "local" imaging analysis, allowing to see structure and detail with unprecedented resolution, without the need for rigorous sample preps. Further technical advances have greatly extended the capabilities of SPMs through the development of new "modes" useful in the study of functional devices, such as conductive atomic force microscopy (**C-AFM**) or Kelvin probe microscopy (**KPFM**).

The lecture will then focus on piezoresponse force microscopy (**PFM**)[3-4], a technique that has become the prevailing approach for nanoscale characterization of polar materials.

Finally, combinations of different SPM-based techniques will be discussed, as well as future challenges for SPM-based techniques

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Ab-initio Spin-orbitronics

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In this lesson, I will give an overview on the role of spin-orbit coupling (SOC) in complex oxides, starting from atomic magnetism and then moving to relativistic phenomena induced by SOC in both magnetic and non-magnetic compounds. I will then focus on the interplay between spin and dipolar degrees of freedom via spin-orbit interaction in ferroelectric semiconductors, by addressing the recently found electrically-controllable Rashba splitting in different materials.

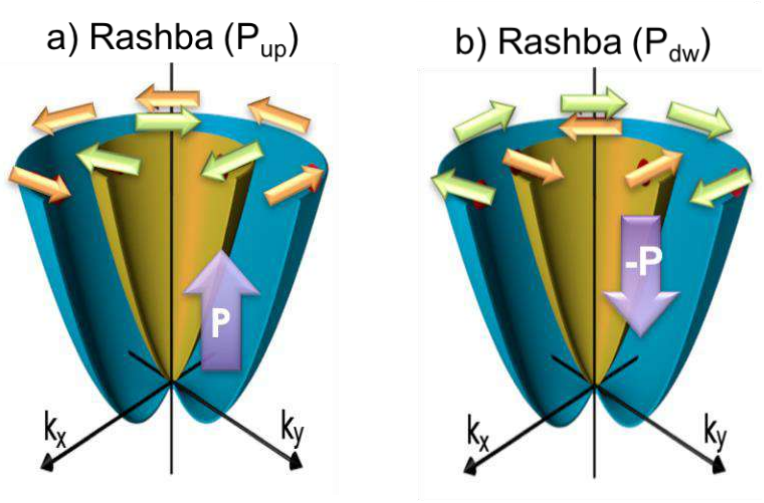

Figure 1. Dependence of spin-texture upon ferroelectric polarization due to the Rashba spin coupling.

MOCVD growth of oxide thin films

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Thin film technology, based on different chemical and physical methods, enabled miniaturization, co-integration, and amelioration of the performance of the devices. Chemical vapor deposition (CVD) systems ensure high productivity and demonstrate excellent film uniformity (up to 12-inch wafers) and repeatability with high throughput for a variety of different films of oxides, nitrides, metals, chalcogenides, etc. This tutorial aims to give a comprehensive overview and basics of application of metal-organic chemical vapor deposition (MOCVD) for synthesis of functional oxide films.

A particular attention will be given to direct liquid injection (DLI)-CVD enabling the usage of solid and liquid precursors, which has proven to be one of the most versatile CVD processes to meet industrial requirements. The requirements to the precursors suitable for DLI-CVD, different classes of available precursors, and models used to describe the evaporation will be overviewed. Then, different liquid delivery devices used in DLI-CVD such as capillary tubes, syringes, aerosol delivery systems, and valves will be introduced. Furthermore, the basic principles of crystallization and epitaxial/textured film growth used to control the film quality, morphology, roughness and orientation will be introduced. The deposition techniques enabling the control of film composition/nonstoichiometry will be compared. Future prospects and commercial aspects of MOCVD are overviewed, as well.

Oxide interfaces

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The rich physics of transition metal oxides resulting in this wide variety of properties is related to the delicate balance between charge, spin and orbital degrees of freedom. This large diversity is observed in materials, mainly of the perovskite family having similar structure and lattice constant, allowing for the growth of heterostructures with very high structural quality. Using modern synthesis methods, it is now possible to engineer interfaces between complex transition metal oxides with an atomic-scale precision. Interfaces break the symmetry, induce stresses, consequently altering the distances and bonds between the ions and giving rise to changes in bandwidths, interactions and in energy levels degeneracy, therefore possibly modifying the electronic phase of these materials. Tailoring and controlling (taking advantage of the sensitivity of these new phases to external stimuli) the physical properties at these interfaces between different oxide materials thus provides a new playground for researchers and offers a new nanoelectronics fabrication platform for future electronics and spintronics.

In this lecture I will introduce the topic of oxide interfaces and the different mechanisms for interface reconstruction. I will then describe its most emblematic member, $LaAlO_3/SrTiO_3$. I will discuss the mechanism for the formation of the two-dimensional electron gas (2DEG) at this interface and present its transport properties (in the normal and superconducting regime) and notably its unconventional Rashba spin-orbit coupling. Finally, I will present other types of oxide interfaces with charge reconstruction, and their resulting novel transport magnetic properties.

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M. Bibes, J.E. Villegas and A. Barthélémy, *Ultrathin oxide films and interfaces for electronics and spintronics* Adv. Mater. 60, 5 (2011)

This work received support from ERC CoG MINT (contract number 615759)

Oxide magnetoelectrics

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The magnetic properties of epitaxial thin films can be electrically controlled via piezoelectric strain from ferroelectric substrates. The resulting magnetoelectric effects promise energy efficient data storage.

In this lecture, I will:

- describe the fundamentals of magnetoelectric oxides
- give an overview of their (microscopic and macroscopic) measuring techniques
- and present recent advances on magnetoelectric oxides and devices.

Oxide spintronics

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Spintronics is a branch of electronics in which transport phenomena are dependent on the electron spin. Future spintronics devices will be built from elemental blocks allowing the electrical injection, propagation, manipulation and detection of spin-based information. Owing to their remarkable multi-functional and strongly correlated character, oxide materials already provide building blocks for charge-based devices such as ferroelectric field effect transistors (FETs), as well as for spin-based two-terminal devices such as magnetic tunnel junctions.

In this lecture, I will first present results obtained on such oxide-based tunnel junctions using half-metallic electrodes of e.g. manganese perovskites. Then, I will discuss the spin-filtering effect by which highly spin-polarized currents can be generated through tunneling across a thin ferromagnetic or ferrimagnetic insulator (EuO, $BiMnO_3$, spinel ferrites), useable to obtain tunnel magnetoresistance. In a second part, I will review how non-magnetic oxide heterostructures can be designed to generate and detect spin currents through the direct and inverse spin Hall effect and the direct and inverse Rashba-Edelstein effects, in particular using oxide 2DEGs.

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Introduction to Transmission Electron Microscopy

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Scanning Transmission Microscopy (STEM) and its related structural and spectroscopic imaging techniques is a very versatile technique for providing structural, chemical and electronic information from materials at very high spatial resolution. Over the last decade, the development of lens aberration correctors has allowed a real breakthrough towards sub-angstrom resolution. Different types of atomic resolution imaging modes are now available allowing access to the crystallography of materials in real space. These new imaging modes, coupled with new possibilities in spectromicroscopy at the individual atomic column scale, provide particularly relevant information channels for the exploration of physics at strongly correlated oxide interfaces.

In this introductory course, the physical basics of image formation in STEM will be introduced and some different types of image modes and contrasts will be reviewed. As a few applications, we will show, for example, how it is possible to probe the role of structural distortions or to quantify in real space the atomic displacements with a few picometers in nanostructured oxide materials.

Polarization dependent X-ray absorption

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X-ray absorption is a local probe for studying magnetic, electronic and structural properties of a variety of materials. Its main appeal is the element selectivity, which makes it a unique tool for some applications. The technique was developed and significantly improved with the advent of synchrotron sources, since it requires a bright source with tunable energy. X-ray absorption is used in widely different research topics such as catalysis, organic and inorganic systems, biochemistry, magnetism. The lecture will focus on the use of x-ray absorption in complex oxides, starting from basic concepts and relating to examples from the literature.

In this lecture the students will learn about the basics of x-ray absorption spectroscopy and how the use of the x-ray polarization can probe different quantities, such as charge distribution or magnetic moment.

The main goal of the lecture is that students learn how to apply x-ray absorption for studying certain materials and to critically think whether this technique can be used in their research project.

The lecture is divided in two parts. During these two lectures the following topics will be covered:

- Production of x-rays in synchrotron sources and its polarization properties.
- Interaction of x-rays with matter. The dipole approximation of the transition operator will be discussed and its consequence, namely the dipole selection rules. It will be shown how the x-ray absorption cross section relates to the density of electronic states.
- It will be shown how x-ray absorption by different electronic core levels probes different states and therefore gives different types of information. It will be discussed how electronic and local structural information can be obtained from K-edge (transition $1s \rightarrow np$) spectra of transition metals. Focus will be given then to $L_{2,3}$-edges (transition $2p \rightarrow nd$), where the valence band of the 3d transition metal is probed. In this context we will discuss the spectral differences between metals and oxides and its relation to electronic correlations.
- In order to further understand the information contained in a spectrum, theoretical simulations are often needed. It will be discussed the different theoretical approaches available and which one is more appropriate to which measurement.
- Examples of experimental setup will be shown and how that differs depending on the energy scale of the x-rays. Various detection methods and their different probing depths will be discussed.
- Finally, the use of x-ray polarization in absorption will be included, which gives rise to linear and circular dichroism techniques. We will discuss which quantities are probed when linear or circularly polarized x-rays are used, which quantitative or qualitative information can be obtained and what are the limitations. Examples from the literature will be given in order to relate to real problems.

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Piezoelectric oxide thin films for acoustic applications

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The next generation of high-frequency (5 - 6 GHz) wide-band RF filters or frequency-agile filters are urgently needed for the development of 5G infrastructures/networks/communications. A further increase in frequency of conventional RF surface acoustic wave (SAW) filters, mainly fabricated from $LiNbO_3$ and $LiTaO_3$ single crystals, is limited by the period of interdigited electrodes (limit of deep UV or electron lithography) and the phase velocity or electromechanical coupling coefficient of available materials. Generally, such piezoelectric RF filters operating at high frequencies are based on thin film bulk acoustic wave resonators (TFBARs) exploiting thickness mode resonances in a thin AlN piezoelectric film. AlN films are limited by their low electromechanical coupling (K^2), which restricts the bandwidth that can be achieved and hence limits the frequency. New suitable low loss materials with larger K^2 and high acoustic velocities are needed to achieve filters at high frequencies. This motivates further development of acoustic wave devices based on piezoelectric thin films, adapted to the high-frequency applications. In this tutorial, the requirements, challenges and achievements in the piezoelectric thin films and their integration with silicon technology and with acoustic devices will be presented in detail. The top-down and bottom-up techniques enabling the implementation of piezoelectric film to the acoustic devices will be compared. Future prospects of potential applications and the expected performances of thin film devices are overviewed, as well.

Nanoscale ferroelectrics

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The depolarization field arising from imperfectly screened bound charges may dramatically alter the properties of ferroelectric ultrathin films, resulting in some cases in the formation of domains of opposite polarization[1]. The film properties are also modified by the presence of a built-in voltage, resulting in a shifted P-E hysteresis loop and affecting the intrinsic polarization configuration and the stability of reversed domains. From the perspective of applications, the interest to understand and quantify these effects is driven by device concepts such as ferroelectric random-access memories, ferroelectric gate transistors, ferroelectric tunnel junctions and memristors[2-10].

This lecture will present a brief overview of ferroelectric nanostructures fabrication methods and investigation thereof by means of scanning probe microscopy – presented in the previous lecture – as well as other techniques such as XRD[11], TEM[12], or second harmonic generation microscopy SHG[13].

This lecture will then focus on the tailoring of the desired electrical properties and dynamics of domains and domain walls for nanoelectronics applications, before discussing some future trends, such as two-dimensional van der Waals ferroelectrics[14] or HfO_2-based films for memory applications[15].

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Antiferromagnetic spintronics

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One of the main features of antiferromagnets (AF) lies in the peculiar antiparallel alignment of their spin textures. Despite a lack of net magnetization and therefore a fairly challenging access to their magnetic distributions, AF are currently in the limelight thanks to recent breakthroughs which have demonstrated the efficient interplay between spin currents and the AF order parameter [1,2], leading to the emergence of a new field of research focused on antiferromagnetic spintronics [3]. Besides, the intrinsic AF dynamics, unlike its ferromagnetic counterpart, lies directly in the terahertz range. Consequently, current-induced AF control also opens new perspectives in ultrafast magnetization dynamics. On the material side, antiferromagnets represent a large majority of magnetic materials. In this presentation after reviewing the history of antiferromagnets and their main physical characteristics, we will discuss in detail their ultrafast dynamics. On this basis, the interplay between spin currents and AF distribution will be tackled. Finally, we will envision what could be achieved in the future with AF distributions.

References:

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Energy-Loss Spectroscopy in a STEM (STEM-EELS)

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In this second lecture, we will discuss the basics of Electron Energy-Loss Spectroscopy (EELS), a preferred spectroscopic technique in STEM. We will show how EELS, when spatially resolved at the atomic column scale, makes it possible to study structural reconstructions at interfaces such as the nature of the termination planes or cations inter-diffusion. In addition, we will show how this technique can probe the electronic structure at the local scale for quantifying the number of electrons in the 3d bands of transition metals. It becomes possible, for example, to image charge orders or to map the charge distribution associated with interface reconstructions. We will end this lecture by presenting some new directions in transmission electron microscopy for the study of oxide heterostructures: in-situ experiments (temperature change or application of electrical voltage during the observation) or spectromicroscopy with an energy resolution of about ten meV opening access to a new spectral range of interest for the physics of strongly correlated oxides (dd type intra-band transitions, spectral signatures of energy gaps or plasmon modes involved in metal-insulator transitions)…