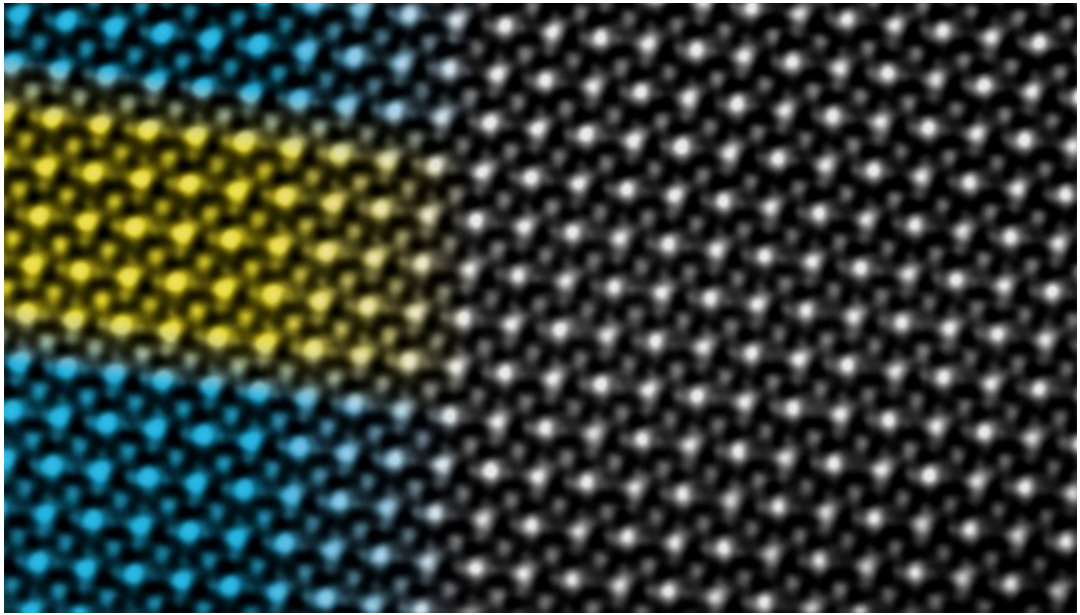
Condensed Matter Physics



Condensed matter theory

Prof. Titus Neupert

We study **topological phases of quantum matter** with numerical and analytical tools. Topological electronic states are characterized universal and robust phenomena, such as the Hall conductivity in the integer quantum Hall effect, that are of fundamental interest or promise applications in future electronics. We study and propose **concrete materials** to realize such topological effects, but are also interested in studying abstract models to understand what phases of matter can exist in principle.
Our numerical toolbox includes **neural network algorithms** to study strongly interacting quantum many-body systems. Furthermore, we work at the interface of **quantum computing** and condensed matter physics.

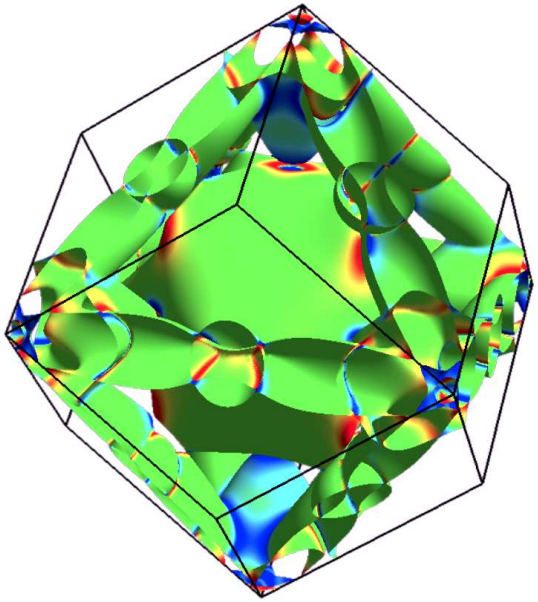
https://www.physik.uzh.ch/g/neupert

Numerical tools for first principles material modeling

Topology has become a major theme in studying quantum matter over the past decades. It is grounded in beautiful mathematical concepts and allows for the classification of phases of matter in a way that is complementary to the paradigm of symmetry breaking. Most excitingly, however, many of the theoretically conceived new phases have been found realized in actual materials over the past years. Their physics is often driven by spin-orbit coupling, with examples including two- and three-dimensional time-reversal symmetric topological insulators, Weyl and Dirac semimetals, and higher-order topological insulators. These discoveries were enabled by detailed predictions of first-principles calculations.

One thrust of our research, lead by Dr. Stepan Tsirkin, is to develop tools that allow to analyze the results of such calculations and further connect them to experimentally observable quantities. Two such tools have been released in 2020.

IrRep is a python tool that calculates the irreducible representation of Bloch eigenstates in the electronic band structure of a material. Along with the refinement of topological classifications came a renaissance of band theory. It has been realized that topological properties of a material can be 'read



Fermi surface of body centered cubic iron, colored by the Berry curvature computed using WannierBerri

off' its symmetry representations in many cases, leading to so-called symmetry-indicator topological invariants. Our IrRep code allows to extract the information needed for such an analysis based on the bandstructures computed with state-of-the-art density functional theory codes such as VASP, Quantum Espresso, or Abinit, as well as any other code that has an interface to the popular format of Wannier90.

WannierBerri is a code package that allows to compute very efficiently physical properties of materials that are related to their Berry curvature. Berry curvature, often characterized as a 'magnetic field in momentum space', is an intrinsic property of many materials, in particular spin-orbit coupled ones, and is intimately related to topological characteristics. For instance, a Weyl point is a monopole of diverging Berry curvature, leading to remarkable properties of Weyl semimetals. WannierBerri is capable of computing the intrinsic anomalous Hall and Nernst conductivities, the orbital magnetization, the nonlinear Hall effect, the gyrotropic magnetoelectric effect and many other response functions. Its capabilities are constantly expanding.

Highlighted Publications:

1. IrRep: symmetry eigenvalues and irreducible representations of ab initio band structures, M. Iraola *et al.*, arXiv:2009.01764
2. High performance Wannier interpolation of Berry curvature and related quantities with WannierBerri code, S.S. Tsirkin, npj Computational Materials 7, 33 (2021), https://www.nature.com/articles/s41524-021-00498-5
3. WannierBerri https://wannier-berri.org

Superconductivity and Magnetism

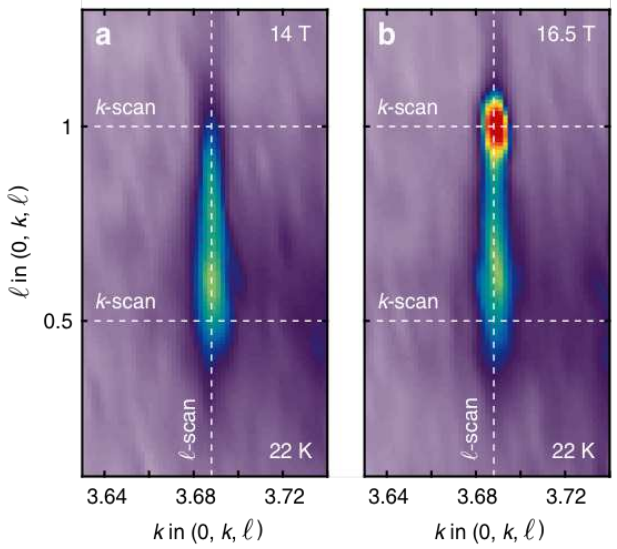
Prof. Johan Chang

We investigate **quantum matter phases emerging from strong electronic interactions**. High-temperature superconductivity, strange metals, density-wave instabilities and electronic driven metal-insulator transitions are studied by synchrotron and laboratory based experimental techniques. At international synchrotrons, we are carrying out angle-resolved photo-emission spectroscopy (ARPES) and resonant inelastic x-ray scattering (RIXS) to reveal electronic structures and properties of correlated electron systems. Quantum phase transitions tuned by magnetic field or hydrostatic pressure are furthermore explored by high-energy x-ray diffraction. Within our laboratory, similar themes are probed by electrical and thermo-electrical transport measurements. Our group also has technical initiatives to develop innovative and compact cryo-cooling methodology. Finally, we are involved in single crystal synthesis through interdisciplinary collaborations with solid state chemists.

https://www.physik.uzh.ch/g/chang

Two-flavor superconductivity

The charge density wave (CDW) in the high-temperature superconductor $YBa_2Cu_3O_{7x}$ (YBCO) has two different ordering tendencies differentiated by their *c*-axis correlations. These correspond to ferro- (F-CDW) and antiferro- (AF-CDW) couplings between CDWs in neighbouring CuO_2 bilayers. This discovery has prompted several fundamental questions: how does superconductivity adjust to two competing orders and are either of these orders responsible for the electronic reconstruction? With a x-ray diffraction study of $YBa_2Cu_3O_{6.67}$, we showed that regions with F-CDW correlations suppress superconductivity more strongly than those with AF-CDW correlations. This implies that an inhomogeneous superconducting state exists, in which some regions show a fragile form of superconductivity. By comparison of F-CDW and AF-CDW correlation lengths, it is concluded that F-CDW ordering is sufficiently long-range to modify the electronic structure. Our study thus suggests that F-CDW correlations impact both the superconducting and normal state properties of YBCO.



Charge density wave diffraction peaks in $YBa_2Cu_3O_{6.67}$ for temperatures and magnetic field as indicated.

High-temperature charge-stripe correlations.

Unconventional superconductivity is often associated with competing intertwined order parameters. For under-doped cuprate superconductors, the omnipresence of charge ordering has been established. By differentiating elastic from inelastic x-ray scattering signals of $La_{1.675}Eu_{0.2}Sr_{0.125}CuO_4$, it is demonstrated that charge-stripe correlations precede both the structural low-temperature tetragonal phase and the transport-defined pseudogap onset. Likewise to other stripe-ordered compounds, the in-plane integrated intensity remains roughly temperature independent, so that our results are interpreted via a single scattering constituent. Due to this similarity, we provide a unifying picture for the charge-stripe ordering in La-based cuprates. As charge correlations in $La_{1.675}Eu_{0.2}Sr_{0.125}CuO_4$ extend beyond the low-temperature tetragonal and pseudogap phase, their emergence heralds a spontaneous symmetry breaking in this compound.

Highlighted Publications:

1. Spatially inhomogeneous competition between superconductivity and the charge density wave in $YBa_2Cu_3O_{6.67}$
   J. Choi *et al.*, Nature Communications **11**, 990 (2020)
2. Oxide Fermi liquid universality revealed by electron spectroscopy
   M. Horio *et al.*, Physical Review B **102**, 245153 (2020)
3. High-Temperature Charge-Stripe Correlations in $La_{1.675}Eu_{0.2}Sr_{0.125}CuO_4$
   Q. Wang *et al.*, Physical Review Letters **124**, 187002 (2020)

Oxide Interface Physics

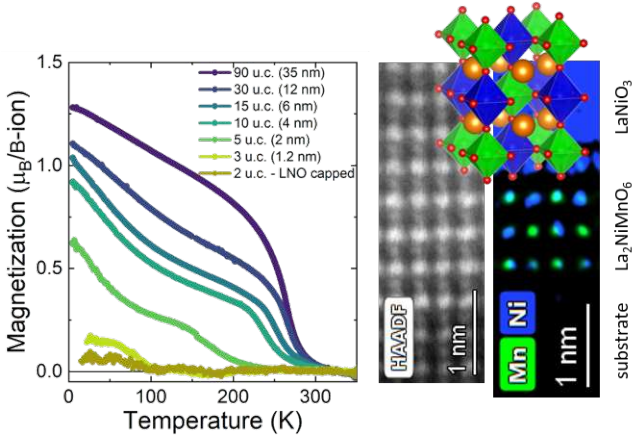
Prof. Marta Gibert

In our group, we grow transition metal oxide heterostructures (i.e. thin films, superlattices) and we investigate their functionalities. We especially focus on the study of the electronic and magnetic properties resulting from reduced dimensionalities and reconstructions occurring at oxide interfaces. Our goal is to understand the subtle atomic-scale structural and electronic mechanisms controlling interface physics in complex oxides. This knowledge is key for the rational design of materials with tailored properties. The atomic-scale precise oxide layers are grown by off-axis rf magnetron sputtering.

https://www.physik.uzh.ch/g/gibert

Transition metal oxides (TMOs) are an extensive class of compounds displaying a large variety of interesting physical properties (i.e. metal-insulator transitions, magnetism, superconductivity, etc.), which makes them highly attractive candidates for next-generation electronic devices. All these functionalities stem from strong electronic correlations and a complex interplay between the charge, orbital, spin and lattice degrees of freedom. Especially attractive are not just the bulk compounds, but also the ability to create artificial layered materials by stacking different oxide compounds one on top of the other, i.e. in the so-called thin films and superlattice structure configurations.

As dimensionality is reduced, the effect of surfaces and interfaces starts to play a critical role in the behaviour of the electron system.[1] Polar/non-polar interfaces constitute a notable example, where a plethora of structural and electronic reconstructions occur leading to alterations of the respective bulk properties. This is the case of double-perovskite La_2NiMnO_6 thin films grown on (001)-oriented $SrTiO_3$ substrates. La_2NiMnO_6 is a ferromagnetic insulator with bulk Curie temperature $T_c \sim 280\,K$ provided that long-range order of Mn^{4+} - Ni^{2+} cations is achieved. By growing thin



Ferromagnetism is observed in double-perovskite La_2NiMnO_6 films as thin as 2-unit cells (0. 8nm) grown on $SrTiO_3$ substrates. By capping them with $LaNiO_3$, the undesired reconstructions associated with the polar discontinuity at film/substrate interface are overcome and the order of the Mn^{4+} - Ni^{2+} cations is recovered.
© TEM - M. Rossell, EMPA.

films, we have shown that this is a very robust ferromagnetic behaviour independent of epitaxial strain.[2] Ferromagnetism is also observed in La_2NiMnO_6 films as thin as 2-unit cells ($\sim$0.8 nm), though both the Curie temperature and the saturation magnetization are reduced compared to the bulk counterpart (see figure). Detailed x-ray absorption spectroscopy (XAS) measurements, transmission electron microscopy (TEM) studies and first-principles calculations have revealed that the polar discontinuity is counteracted by the occurrence of electron-doping, oxygen vacancies formation and antisite disorder in the first 4-5 unit cells close to the substrate interface. Interestingly, by capping the film with $LaNiO_3$ layers, these interfacial reconstructions are overcome. As a result, $LaNiO_3$-capped 2-unit cells La_2NiMnO_6 films display $T_c \sim 80$ K, which corresponds to an increase of ~ 60 K with respect to the equivalent uncapped ones. The still existing difference to the bulk-T_c is attributed to the effect of dimensionality reduction. These results provide further understanding about the interplay between size reduction and fundamental physical properties in oxides thin films. At the same time, they open new venues of research by allowing to compensate the undesired effects often associated with interfacial polar discontinuities at oxides heterointerfaces through top-layer engineering while leaving the interface pristine.

Highlighted Publication:

1. Length scales of interfacial coupling between metal and insulator phases in oxides,
C. Dominguez *et al.*, Nat. Mater. **19**, 1182 (2020)
2. Robust ferromagnetism in insulating La_2NiMnO_6 thin films,
G. De Luca *et al.*, arXiv:2101.07530 (2020)

Low dimensional systems

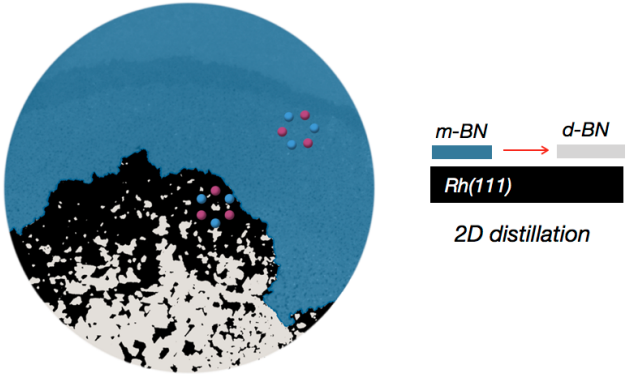
Prof. Thomas Greber

We study objects like **zero dimensional endofullerene** molecules and **two dimensional (2D) boron nitride** layers in view of their functionality as nano-materials. Single molecule magnetism is the focus in the fullerene research, where we apply bulk sensitive x-ray absorption and a sub-Kelvin superconducting quantum interference device for the investigation of the materials that are obtained from collaborations with synthesis groups. In the 2D materials activity we aim to grow highest quality boron nitride on substrates up to the four inch wafer scale with chemical vapour deposition and subsequent exfoliation. For this purposes we use a dedicated clean room, optical microscopy, transmission electron microscopy and surface science tools such as low energy electron diffraction, photoemission and scanning tunneling microscopy.

https://www.physik.uzh.ch/g/osterwalder

Surface distillation of hexagonal boron nitride

Production of high-quality two-dimensional (2D) materials is critical to the full exploitation of their single layer properties and combination in hybrids. With respect to 2D electronic device performance hexagonal boron nitride (*h*-BN) is foreseen to become the key packaging material since it is atomically thin, impermeable, flat, transparent and chemically inert. Surface distillation is a new scheme to produce *h*-BN that is superior to material from chemical vapor deposition (CVD). Single layer CVD *h*-BN is delaminated from Rh(111) and transferred back to a clean Rh(111) surface. The twisting angle between BN and the new substrate yields metastable moiré structures (*m*-BN). Annealing above 1000 K leads to 2D distillation, i.e., catalyst-assisted BN sublimation from the edges of the transferred layer and subsequent condensation into superior quality *d*-BN. It is a low-cost way of high-quality 2D material production remote from CVD instrumentation [1]. The project is a collaboration with the Hong Kong University of Sci-



Illustrated frame from a movie of the 2D distillation process. Left: Colored LEEM image with a field of view of 23 μm at a process temperature of 910 °C. The blue regions are covered with misaligned single layer boron nitride m-BN that has been back-transferred onto the Rh(111) substrate. The m-BN sublimates onto the Rh(111) substrate (black) where boron (pink) and nitrogen (light-blue) diffuse to the light grey regions where they condense into highly ordered d-BN, which is aligned to the substrate. The driving force for surface distillation is the lower chemical potential of d-BN. Right: Corresponding sketch of the 2D distillation process. (LEEM image Zichun Miao, HKUST).

ence and Technology (HKUST) where low energy electron microscopy (LEEM) experiments on our samples were performed (see Figure) and the UZH department of chemistry. The results lead to a joint UZH/HKUST patent application "A method for on-surface synthesis of a hexagonal boron nitride monolayer" that protects the production of single layer h-BN with high lattice quality. The method of surface distillation does not directly involve the use of precursor molecules from the gas phase, but the transfer of a single layer h-BN on a catalyst surface where this h-BN can be cleaned by mild thermal annealing leading to m-BN and further distilled to d-BN.

This activity is supported by the European Future and Emerging Technology flagship graphene.

Highlighted Publications:

1. High-Quality Hexagonal Boron Nitride from 2D Distillation
   H. Cun *et al.*, ACS Nano **15** 1351 (2020)
2. Gadolinium as an accelerator for reaching thermal equilibrium and its influence on the ground state of $Dy_2GdN@C_{80}$ single-molecule magnets
   A. Kostanyan *et al.*, Phys. Rev. B **103** 014404 (2021)
3. Sub-Kelvin hysteresis of the dilanthanide single-molecule magnet $Tb_2ScN@C_{80}$
   A. Kostanyan *et al.*, Phys. Rev. B **101** 134429 (2020)

Quantum Matter

Prof. Fabian Natterer

Our group investigates the properties of two dimensional quantum matter. We explore how materials receive their properties from the interaction between individual atoms that we can control with atomic precision. We furthermore study 2D van der Waal materials and develop new measurement protocols for advanced scanning probe microscopy investigations, such as electron spin resonance, nonlinear spectroscopy, and compressed sensing for quasiparticle interference imaging.

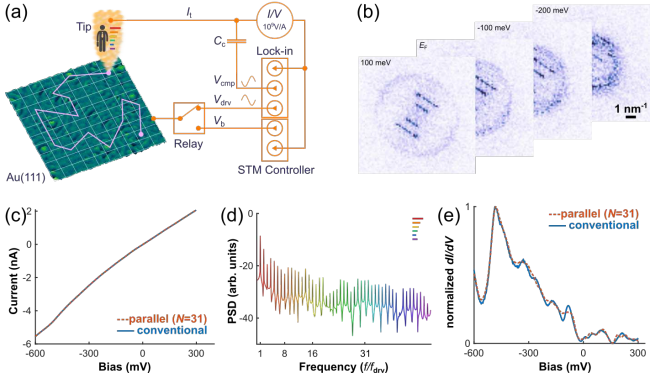
https://www.physik.uzh.ch/g/natterer

Fast Spectroscopic Mapping of Two Dimensional Material

The band structure of two dimensional quantum materials can be inferred from SPM measurements with quasiparticle interference imaging. It works by measuring the point spectroscopy (local density of states) at every topographic location. To speed up this traditionally slow technique, we introduce compressed sensing and nonlinear spectroscopy. While the former enables the measurement of fewer locations, the latter speeds up point spectroscopy. In combination, we achieve several orders of magnitude faster mapping. Measurements that would have lasted a week, can be done within minutes to few hours, enabling novel measurement concepts that were previously inconceivable. The time savings can be used to improve our spectroscopic resolution, to explore a vaster parameter space, or enable a faster measurement throughput, all promoting the discovery of novel quantum materials.

Scanning Probe Microscopy based ESR

Our second main projects is the development of novel spin sensors for electron spin resonance (ESR) with a scanning probe microscope. Our goal is to embed a molecular qubit into the SPM tip and use it as a detector for magnetic signatures at the atomic scale. This sensor will shed light on the spin texture of radicals, it can be used to scrutinize artificially



Combination of sparse sampling and parallel spectroscopy. (a) Experimental Setup. (b) QPI maps. (c) Point spectrum with nonlinearities that generate the harmonics shown in d. (d) Harmonics created by application of a harmonic drive that are used to measure the point spectrum much faster than conventionally. (e) Comparison of conductance obtained with conventional and parallel spectroscopy, showing great agreement at much faster pace.

built quantum matter and in the exploration of noncolinear magnetic structures. Our sensor would provide an alternative to nitrogen vacancy centers that have a limited spatial resolution.

Highlighted Publications:

1. Sparse sampling for fast quasiparticle-interference mapping, J. Oppliger and F. D. Natterer, Physical Review Research **2**, 023117 (2020)
2. Fast spectroscopic mapping of two-dimensional quantum materials, B. Zengin, J. Oppliger, D. Liu, L. Niggli, T. Kurosawa, F. D. Natterer, arXiv:2102.00054

Surface physics

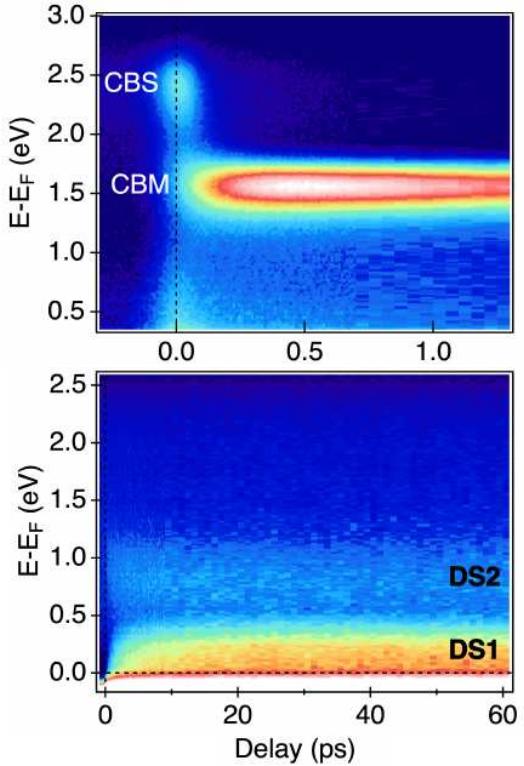
Prof. Jürg Osterwalder

We study processes at surfaces such as molecule adsorption and self assembly, charge and energy transport as well as fundamental aspects of **light-matter interaction** and associated electron dynamics. Our laboratory is equipped with a toolbox of surface science methods for the preparation and characterization of clean single-crystalline surfaces that can be used to investigate such phenomena **at the atomic and molecular level** and **at the femtosecond time scale**. Specific research projects include the structure and function of oxide semiconductor surfaces and interfaces that serve as model electrodes in water splitting devices, as well as the study of their interaction with small molecules at **solid-gas and solid-liquid interfaces**, using *in situ* **ambient pressure x-ray photoelectron spectroscopy** and x-ray absorption spectroscopy using our own endstation at the Swiss Light Source at PSI.

https://www.physik.uzh.ch/g/osterwalder

Carrier dynamics in a cuprous oxide photoelectrode

The direct splitting of water with solar light to form hydrogen gas as an energy carrier is a viable scenario for sustainable energy conversion and storage. Cuprous oxide (Cu_2O) is a promising material for use as a photocathode in photoelectrochemical cells for solar water splitting. It has a direct band gap of just the right size and a high absorbance for solar light, and it is Earth abundant and thus cheap. The relevant processes within the electrode involve the creation of electron-hole pairs, charge separation and transport to the surface. A downward band bending guides electrons through a protective layer to the surface, where a catalyst promotes the reduction of protons to form hydrogen molecules. The electron energy needs to exceed the redox potential for the water splitting reaction, and the transport within the conduction band of Cu_2O to the interface without energy loss is thus essential for an efficient device. Within the University Research Priority Program *LightChEC* we have studied these processes by means of time-resolved two-photon photoemission (2PPE).



2PPE spectra as a function of delay time after a pump laser pulse ($h\nu$ = 3 eV) has hit a $Cu_2O(111)$ sample. Data are shown for a defect-free surface (top), and for one on which a third of the surface oxygen atoms was missing (bottom).

Measurements on a defect-free $Cu_2O(111)$ surface show the ultrafast relaxation of photoexcited electrons (CBS) to the bottom of the conduction band minimum (CBM) with a decay time τ of 30 fs, where a strong population remains at the surface with τ = 10 ps. The same sample, but with a reconstructed surface missing one third of the oxygen atoms in the top layer, shows no trace of population in the conduction band. Instead, a long-lived occupation of defect states DS1 and DS2 is observed, with no decay measurable on a picosecond time scale. These results prove that surface and interface defects rather than bulk defects limit the performance of Cu_2O photocathodes.

Highlighted Publications:

1. Influence of surface defect density on the ultrafast hot carrier relaxation and transport in Cu_2O photoelectrodes
   L. Grad *et al.*, Scientific Reports **10**, 10686 (2020)
2. Dynamics of excited interlayer states in hexagonal boron nitride monolayers
   M. Hengsberger *et al.*, J. Phys. D: Appl. Phys. **53**, 203001 (2020)
3. Probing the solid-liquid interface with tender x rays: A new ambient-pressure x-ray photoelectron spectroscopy endstation at the Swiss Light Source
   Z. Novotny *et al.*, Rev. Sci. Instrum. **91**, 023103 (2020)

Phase Transitions, Materials and Applications

Prof. Andreas Schilling

We are interested in selected topics in materials research, spanning the entire spectrum from **searching new materials**, their **characterization**, and corresponding **applications**. We have been particularly active in **superconductivity, magnetism and thermodynamics**. Our laboratory is equipped with modern furnaces for material synthesis, various ^{4}He/^{3}He cryostats and a dilution cryostat, all with superconducting magnets.

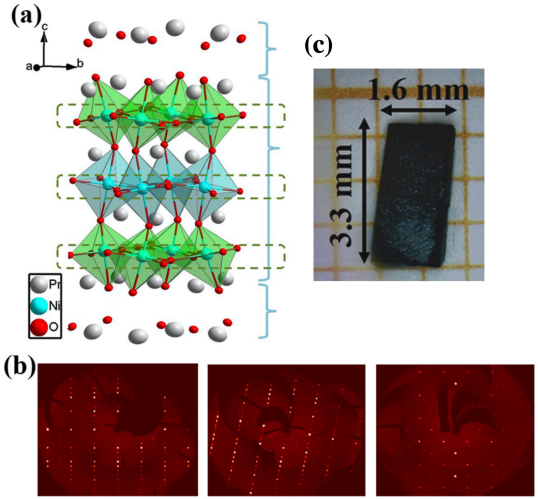
We are structuring thin superconducting films at the FIRST Center for Micro- and Nanoscience at ETHZ and are using them both for basic research and applications. While the physics of thin-film superconductors is a fascinating research topic by itself, corresponding nanostructures may serve as ultrafast single-photon detectors in the infrared, visible and X-ray range.

https://www.physik.uzh.ch/g/schilling

Infinite three-layer nickel-oxides as candidates for high-temperature superconductors

As a member of the Ruddlesden-Popper $Ln_{n+1}\ Ni_nO_{3n+1}$ series rare-earth-nickelates, $Pr_4Ni_3O_{10}$ consists of infinite quasi-two- dimensional perovskite-like Ni-O based layers (Fig. a). Although a metal-to-metal phase transition at $T_{pt} \approx 157$ K has been revealed by previous studies, a comprehensive study of physical properties associated with this transition has not yet been performed. We have grown single crystals of $Pr_4Ni_3O_{10}$ with an optical-image floating-zone furnace at high oxygen pressure in collaboration with PSI Villigen (Figs. b-c). The resistivity data indicate a metal-to-metal transition at T_{pt} within the $a-b$ plane and a metal-to-insulator-like transition along the c-axis upon cooling. The magneto-resistance (MR) is enhanced at T_{pt} and exhibits a sign change, which we attribute to a suppression of the first-order phase transition by a magnetic field.

We have furthermore successfully synthesized the series $Ln_{4-\ x}Ln'_xNi_3O_{10}$ (Ln and Ln′ = La, Pr and Nd). The



Crystal structure (a), Laue photographs (b), and a picture (c) of the grown $Pr_4Ni_3O_{10}$ single crystal

temperatures T_{pt} and the room-temperature resistivities vary systematically with the Goldschmidt tolerance factor t. With increasing t, the compounds become more conducting, which we attribute to a successively diminishing distortion of the NiO_6 octahedra which are responsible for the electron transport in these materials. While the bond lengths and Ni-O-Ni bond angles play a crucial role for the electronic structure, the magnetism of the Ln^{3+} ions is not a decisive factor.

We have synthesized $Pr_4Ni_3O_8$ by topotactic reduction of $Pr_4Ni_3O_{10}$. This compound features quasi-two-dimensional layers consisting of square-planar NiO_2 planes, in a similar way to the well-known T′-type cuprate superconductors. While $Pr_4Ni_3O_8$ is known to be metallic, little is known about the magnetic behaviour of this compound. A series of powder neutron diffraction, magnetization and μSR measurements on $Pr_4Ni_3O_8$ powders all show no evidence for long-range magnetic order in $Pr_4Ni_3O_8$. Our magnetization data clearly demonstrate a spin-glass behaviour with a freezing temperature $T_s \approx 68$ K and a distinct magnetic memory effect. The μSR data reveal two magnetic processes: a slowly-relaxing signal, resulting from paramagnetic fluctuations present at all temperatures, and a rapidly growing fast-relaxing signal due to the presence of short-range correlated regions in the glassy state below T_s.

Highlighted Publications:

1. Anisotropic character of the metal-to-metal transition in $Pr_4Ni_3O_{10}$, Sh. Huangfu *et al.*, Phys. Rev. B **101**, 104104 (2020)
2. Correlation between the tolerance factor and the phase transition in $Ln_{4-x}Ln'_xNi_3O_{10}$, Sh. Huangfu *et al.*, Phys. Rev. Research **2**, 033247 (2020)
3. Short-range magnetic interactions and spin-glass behaviour in the quasi-2D nickelate $Pr_4Ni_3O_8$, Sh. Huangfu *et al.*, Phys. Rev. B **102**, 054423 (2020)