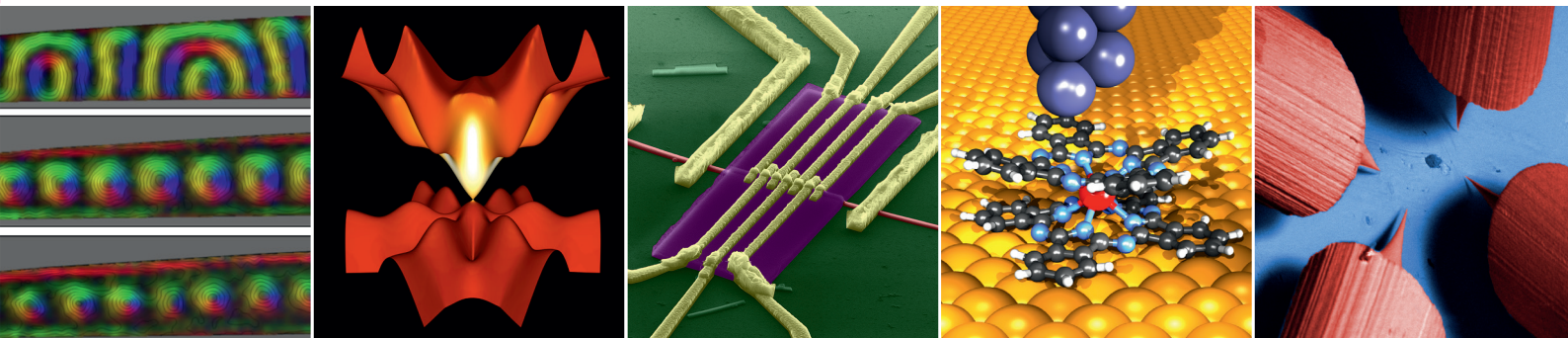


Status Report 2013–2017

Scientific Evaluation of the Research Field Key Technology

Forschungszentrum Jülich, 23–26 October 2017

Volume I – Part IV



Future Information Technology
Peter Grünberg Institute (PGI)

Research Field Key Technology

Status Report 2013 – 2017

Volume I

Part IV

Program Future Information Technology

**Research Unit:
Peter Grünberg Institute (PGI)**

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2. Research Unit: Peter Grünberg Institute (PGI) – Program Future Information Technology – Fundamentals, Novel Concepts, and Energy Efficiency

2.1 Overview

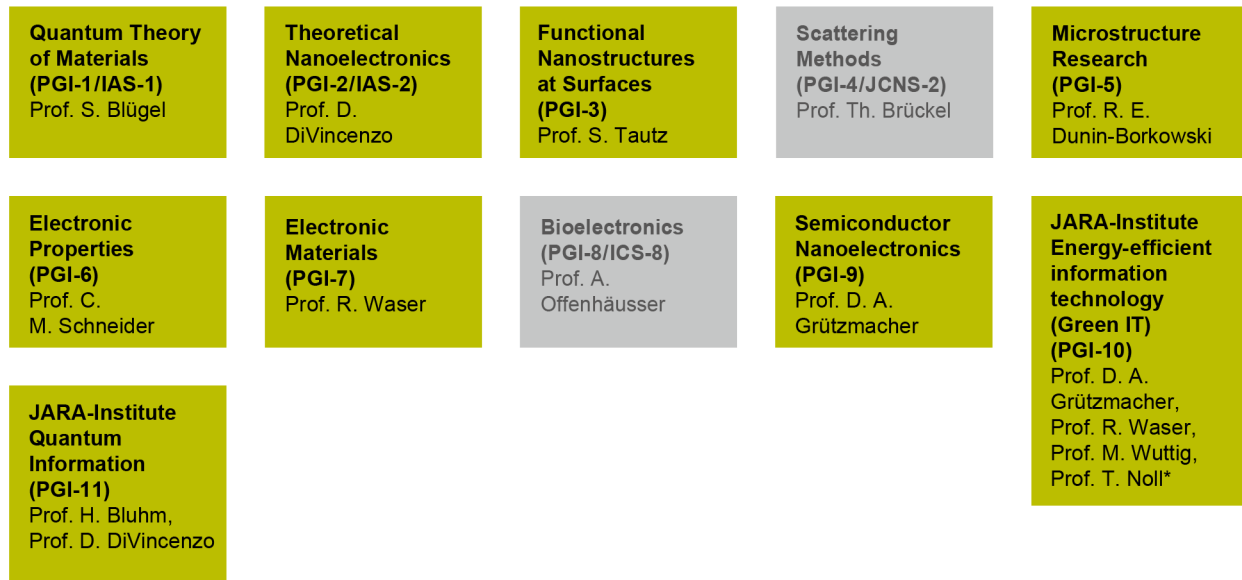


Fig. 1: Research units participating in the Program FIT in Jülich (institutes in green contribute to this review). (* Jara Senior Professor)

The Peter Grünberg Institute consists of 11 scientific departments (PGI-1 to PGI-9, and the JARA Institutes PGI-10 and PGI-11) with complementary research interests (Fig. 1) and a support unit of technical/scientific infrastructure. The head of each department holds a joint appointment with nearby universities. Several departments in PGI have double department memberships, reflecting joint activities in the fields of supercomputing (PGI-1/IAS-1, PGI-2/IAS-3), neutron science (PGI-4/JCNS-2) and biophysics (PGI-8/ICS-8). In addition, the Ernst Ruska-Centre (ER-C) and the JARA Institutes are operated jointly with RWTH Aachen University.

A key element of the PGI's strategic concept is JARA (www.jara.org), the alliance between Forschungszentrum Jülich and RWTH Aachen University. This is achieved in particular through the JARA section *Fundamentals of Future Information Technology* (JARA-FIT). Within the framework of the Jülich-Aachen Research Alliance (JARA), PGI has recently been extended by the formation of two new JARA Institutes: Green Information Technology (PGI-10) and Quantum Information (PGI-11).

For the term 2015 – 2019, the research of the Peter Grünberg Institute is defined mainly within the HGF Program *Future Information Technology* (FIT). This program is carried out in close collaboration with the Helmholtz Zentrum Berlin für Energie und Materialien (HZB) (approx. 6% of the program). It is structured into four Topics, which address relevant **state variables** in the physics of information technology and electronic phenomena: (1) Controlling Charge-Based Phenomena, (2) Controlling Spin-Based Phenomena, (3) Controlling Configuration-Based Phenomena and (4) Controlling Collective States. More detailed descriptions of the program can be found in the separate FIT research program proposal.

Fig. 2 presents the Jülich contribution of personnel funded by the Program FIT. Personnel in 2016 totaled 135 scientists in units of full-time equivalents (FTE) (neglecting Master's students, stipendiaries, and guests) at PGI. This number can be further broken down into nine W3/C4 positions for the institute directors (plus three at the JARA institutes), six W2/C3 positions, and

six W1 positions with teaching responsibilities at one of the neighboring universities (RWTH Aachen University, University Duisburg-Essen, etc.). The PGI hosts 7 Helmholtz-University Young Investigator groups and one DFG-funded Emmy-Noether group. The leaders of four Young Investigator Groups have a W1 position. The remaining groups consist of research scientists (135 FTE), engineers, technicians and administrative personnel (63). In 2016, we supervised 144 PhD students.

The institute unit PGI-8 also participates in the research program BioSoft as ICS-8 (see Part III: BioSoft). Both these institute units are presented and reviewed during the ICS review. Furthermore, the PGI-4 main focus is on the application of neutron scattering methods for solid state physics. The Institute is therefore only briefly presented in this report and will be reviewed as PGI-4 / JCNS-2 in the research field “Matter”. However, numbers given in Fig. 2 include all personnel funded by the program.

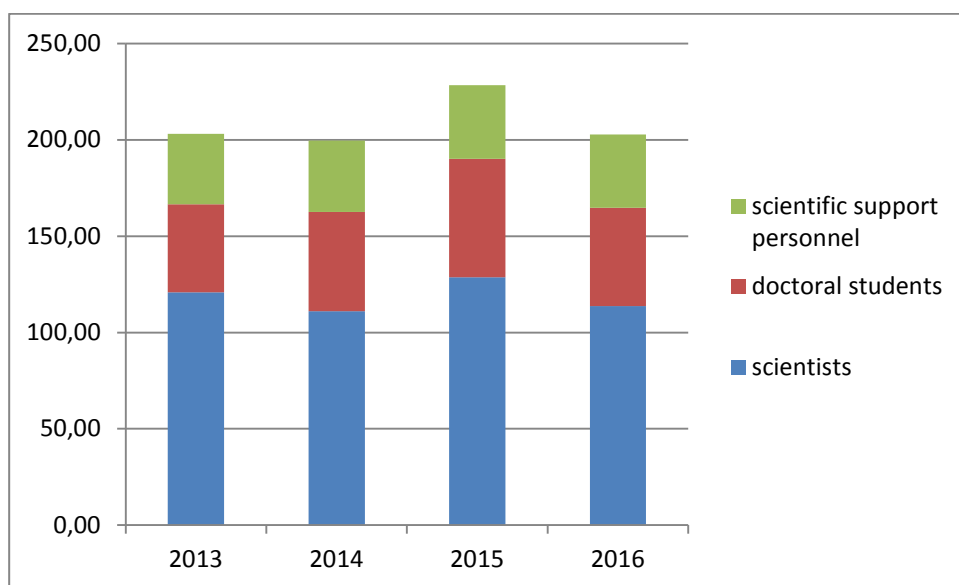


Fig. 2: Personnel funded by the Program FIT. Numbers are given in units of Full-Time Equivalents (FTE), defined as the number of total hours worked divided by the maximum number of compensable hours in a full-time schedule. Numbers do not include PGI-4 and PGI-8 personnel in the Program FIT, which are reviewed together with ICS-8 and JCNS-2 in the programs BioSoft and Matter, Materials and Life.

2.2 Introduction

The mission of the Peter Grünberg Institute (PGI) is the discovery and interpretation of new phenomena in condensed matter, the development of novel materials and functional nanostructures as well as innovation in experimental and theoretical methods, all with a special emphasis on potential long-term applications in information technology. A particular strength of the PGI is the ability to tackle complex multiscale problems by an integrated approach, involving scientists across and also beyond the boundaries of its departments. This approach is facilitated by a wide spectrum of experimental facilities and individual expertise, and by a close collaboration between experimental and theoretical groups. Fully exploiting the special infrastructure of Forschungszentrum Jülich, research focuses on the basic understanding and technical development of nanoelectronics and information technology with an emphasis on novel concepts for future energy-efficient information technology and on electronic phenomena in specific fields of energy technology.

The PGI covers important research areas in condensed matter physics, surface physics, quantum information, information technology, and energy research. Existing scientific strengths

of the PGI are based on long-standing expertise and excellence in fundamental research in physics, solid-state-based information technology, simulation sciences and supercomputing, cutting-edge method development and contributions to dedicated scientific infrastructure. These areas of expertise and strength form the necessary basis to enable PGI to be adaptable and flexible institute concerning the choice of research topics and to be an institute that always seeks out new scientific challenges. Using a comprehensive approach in experimental and theoretical science covering fundamental and applied research as well as device physics and prototyping, the PGI drives innovation from the idea through to its realisation.

The participating institutes in Jülich and Aachen are:

- Peter Grünberg Institute & Institute for Advanced Simulation – Quantum Theory of Materials (PGI-1/IAS-1), Forschungszentrum Jülich, Head: S. Blügel
- Peter Grünberg Institute & Institute for Advanced Simulation – Theoretical Nanoelectronics (PGI-2/IAS-3), Forschungszentrum Jülich & Institute for Quantum Information, RWTH Aachen, Head: D. P. DiVincenzo
- Peter Grünberg Institute – Functional Nanostructures at Surfaces (PGI-3), Forschungszentrum Jülich, Head: S. Tautz
- Peter Grünberg Institute & Jülich Centre for Neutron Science – Scattering Methods (PGI-4/JCNS-2), Forschungszentrum Jülich, Head: Th. Brückel
- Peter Grünberg Institute – Microstructure Research, Forschungszentrum Jülich & Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons (PGI-5/ER-C), Head: R. E. Dunin-Borkowski
- Peter Grünberg Institute – Electronic Properties (PGI-6), Forschungszentrum Jülich, Head: C. M. Schneider
- Peter Grünberg Institute – Electronic Materials (PGI-7), Forschungszentrum Jülich & Institut für Werkstoffe in der Elektrotechnik II, RWTH Aachen University, Head: R. Waser
- Peter Grünberg Institute & Institute of Complex Systems – Bioelectronics (PGI-8/ICS-8), Forschungszentrum Jülich, Head: A. Offenhäusser
- Peter Grünberg Institute – Semiconductor Nanoelectronics (PGI-9), Forschungszentrum Jülich, Head: D. Grützmacher
- Peter Grünberg Institute – JARA Institute Green IT (PGI-10), Forschungszentrum Jülich & RWTH Aachen University
- Peter Grünberg Institute – JARA Institute Quantum Information (PGI-11), Forschungszentrum Jülich & RWTH Aachen University

The role of the research units PGI-4/JCNS-2 and PGI-8/ICS-8 in this review has been commented on already in 2.1. For a description of the PGI-10 and -11, please see below.

Scientific expertise is also available through the close cooperation with the Institute for Advanced Simulation (IAS) and the very successful participation of RWTH Aachen University in the Program through the JARA-FIT Alliance. The PGI secures the expertise of important senior scientists through the awarding of Helmholtz Professorships (e.g., Prof. H. Katayama-Yoshida) and the JARA Senior Professorships (e.g., Prof. T. Noll).

The Peter Grünberg Institute actively contributes to two of the six sections of JARA (see Vol. I, Part I, Strategic Partners and Cooperation): JARA-FIT and JARA-HPC. In fact, JARA-FIT was founded by the Peter Grünberg Institute together with colleagues from RWTH Aachen University, shortly after the Nobel Prize was awarded to Peter Grünberg in 2007, as a strategic partnership program centered on discipline-based research. The section JARA-FIT is a collaborative and cross-disciplinary effort of 32 institutes and chairs (11 Jülich/ 21 Aachen) from

physics (8/9), chemistry (0/3), biology (1/0), mathematics (0/1), electrical engineering and electronics (2/6), mechanical engineers (0/1) as well as earth sciences and geography (0/1). The section has over time expanded in dedicated fields by means of joint strategic planning of the research, coordinated teaching activities and infrastructure development.

An integral element of the HGF Program FIT and the contribution from the Peter Grünberg Institute was the fact that its partner, RWTH Aachen University, was able to consolidate its leading position as a technical university within the Excellence Initiative of the German federal and state governments in the period 2012 to 2017 (Excellence Initiative II). This relied strongly on the well-established Jülich-Aachen Research Alliance, JARA, which served as a role model for uniting program and discipline-based research in Germany. JARA is active in joint appointment procedures, joint infrastructure planning such as synchronizing HNF and RWTH Aachen cleanroom facilities, and the establishment of joint research groups located at both sites, thus harmonizing the general strategy of both institutions in the JARA research areas.

An important example of the success of JARA-FIT is the establishment of the DFG (German Research Foundation)-funded Collaborative Research Centre (Sonderforschungsbereich) **SFB 917 “Resistively Switching Chalcogenides for Future Electronics – Structure, Kinetics and Device Scalability”**, where synergetic effects with RWTH Aachen University are further exploited. Groups from six different PGI institutes have contributed here.

Major achievements of JARA-FIT began with a dedicated seed-funding scheme which involved the evaluation and commissioning of bilateral cooperative projects. All together, five calls have been launched since 2011, with a current total of 24 projects. A milestone in the cooperative effort was the establishment of the Virtual Institute for Topological Insulators, VITI, in 2012, founded by Forschungszentrum Jülich and RWTH Aachen University together with Würzburg University and Shanghai Institute of Microsystem and Information Technology.

Other measures fostering this collaboration are jointly organized regular workshops. The biannual Science Days provide a forum for scientific exchange among JARA-FIT scientists, bringing together young scientists (PhD students and postdocs) with senior scientists from both institutions. The Nanoelectronics Days were set up as an international conference, the last being held in 2015 and devoted to green information technology.

A major development in JARA-FIT during the present period of the Excellence Initiative is the commitment of the two institutions, Jülich and Aachen, to strengthen the partnership by establishing new formats of cooperation, so-called **JARA Institutes** and **JARA Professorships**. This is accompanied by the two institutions developing a common strategy in the fields of neuromorphic computing and quantum computing. The two JARA Institutes within JARA-FIT have a specific mission in the areas of (1) energy-efficient information technology (“Green IT”) and (2) quantum information. Since the institutes were first established in 2016, the Institutes’ recent scientific work will be presented in more detail under the following subsections (PGI-10 & PGI-11).

JARA-FIT Institute Green-IT (PGI-10) The main thrust of the new institute is the development of novel devices and architecture concepts for merging logic and storage components on computer chips. Under the joint leadership of Detlev Grützmacher (FZJ), Tobias Noll (RWTH/FZJ), Rainer Waser (FZJ/RWTH), and Matthias Wuttig (RWTH/FZJ), the institute aims to bring together expertise from physics and electrical engineering in Aachen and Jülich in order to combine ultra-low power logic with novel energy-efficient memristive devices at the nanometer-scale. It is the goal of the institute, to extend the scope of our Collaborative Research Center (Sonderforschungsbereich SFB 917) *Resistively Switching Chalcogenides for Future Electronics* and to develop a prototype for a combined transistor/memristive device,

appropriate architectures, as well as suitable device and circuit models. When ultra-low energy is used on each operation, the rate of intermittent, transient faults will increase significantly. Therefore, the institute will investigate the reliability of these new systems with a particular focus on variability and fault tolerance. Moreover, the institute has the potential to support the transition from binary computing to neuromorphic information processing, inspired by the energy efficiency of the human brain. Materials already under intense investigation within JARA-FIT such as chalcogenide-based memristive systems, have the ability to adopt multinary resistive states, which may represent synaptic weights in neuromorphic concepts. The institute will take up this challenge and establish a fundamental materials basis for neuromorphic computing, making extensive use of findings and data from basic neuroscientific research present in Jülich and its strong link with the increasing efforts in brain research (Helmholtz Research Program *Decoding the Human Brain* and EU flagship project *Human Brain Project*). The backbone of the institute is the establishment of a state-of-the-art deposition cluster with in-situ characterization tools (**Nanocluster**) to efficiently combine different materials and significantly improve interface quality on the campus in Jülich (2014). With a total of 10 UHV growth chambers, the cluster offers approaches ranging from molecular beam epitaxy to atomic layer deposition and magnetron sputtering. In-situ handling between the units permits the growth of hybrid material structures incorporating metals, semiconductors, and oxides. In combination with the Helmholtz Nano Facility (HNF), this allows the construction and investigation of new devices within the “Green IT” framework as well as the broader use of JARA-FIT.

JARA-FIT Institute for Quantum Information (PGI-11) Another approach moving further beyond conventional computing is the use of quantum interference effects as a resource for information processing. The Program *FIT* has a long-standing track-record in spintronics, a field whose importance was affirmed by the Nobel Prize awarded to Peter Grünberg and Albert Fert in 2007. The Program is confident that it will establish an outstanding and broad-ranging presence in an effort to turn spin and other solid-state qubits into functional devices for quantum information processing. After successfully recruiting leading international experts in the field in both Aachen and Jülich, the two institutions will now pool their theoretical and experimental expertise in Jülich and Aachen within the joint JARA Institute for Quantum Information under the directorship of David DiVincenzo and Hendrik Bluhm. Guided by David DiVincenzo, the institute will conduct mathematical studies into the efficacy of fault-tolerant error correction in integrated circuit qubit arrays, and research on the basic solid state physics of quantum dots, quantum wires, and superconducting devices, with detailed analyses of their potential to create higher fidelity qubits. A major task will be the modelling of quantum control schemes performed in direct collaboration with experiment. The experimental effort spearheaded by Hendrik Bluhm will continue to work on highly coherent two-level quantum systems in semiconductor quantum dots. Crucial techniques in high-speed control and measurement will be further improved upon, and multi-qubit systems will be designed, fabricated, and tested. New materials and device configurations will also be examined. The institute will explore opportunities for establishing additional areas of major investigations, including studies of superconducting microwave-resonant structures and superconducting qubits.

2.3 Contribution to FIT

The HGF Program FIT is carried out jointly by Forschungszentrum Jülich and the Helmholtz-Zentrum Berlin (HZB). The program comprises four topics, which address relevant state variables in the physics of information technology and electronic phenomena. All four topics have their roots in basic research, from which novel concepts for highly energy-efficient IT and contributions to other fields will be developed.

The Peter Grünberg Institute contributes to each of the four topics of the program:

Topic 1 – Controlling electron charge-based phenomena. Today, advanced processor and memory chips are still many orders of magnitude away from the ultimate physical and technological limits of charge-based electronics. A dramatic increase in power consumption caused by data traffic as well as by the demand for mobile, grid-independent devices will shift the focus from device scaling to energy efficiency as the main technology driver. This Topic explores potential extensions of Si-CMOS and beyond-CMOS concepts on the basis of semiconductor nanowires for ultra low power transistors and novel concepts for Si based optoelectronics for the integration of optical interconnects. Moreover, alternative chip architecture concepts including energy harvesting for “zero power” mobile devices will be investigated.

Topic 2 – Controlling spin-based phenomena. Spin transport electronics – spintronics – focuses on utilizing the electron spin as an alternative state variable in classical and quantum information technology. This field is currently enjoying a sequence of exciting observations that are due to relativistic effects generating a flurry of transversal spin-currents, new types of spin-torques, and chiral spin-structures. Novel concepts such as topologically nontrivial spin textures in real and reciprocal space are opening new perspectives for dissipationless spin currents as well as large emergent magnetic and electric fields. We realize such concepts in designed nanostructures and investigate static, dynamical and excitation properties with the aim of uncovering new functionalities for future devices.

Topic 3 – Controlling configuration-based phenomena. The configuration of atoms, ions, and molecules can be used as a state variable for information processing and storage on the smallest conceivable scale, as well as in related applications in catalysis, chemical and biochemical sensing, and in specific processes in energy technology. In this Topic, we aim to elucidate the microscopic physical and chemical mechanisms of these types of configuration changes. In particular, we will study energy landscapes and the change in entropy associated with the evolution of one configuration into another, since such processes control kinetics, reversibility, and the energy efficiency of the process – specific to each application.

Topic 4 – Controlling collective states. Collective states permit the exploitation of quantum coherence both for transport beyond classical limits and for quantum information processing. Our program in this topic will comprise (a) the study of the emergent properties of highly entropic and topological states; (b) an understanding of emergent properties arising from broken symmetries found in correlated quantum systems; (c) control of multifunctional states, in bulk materials and in composite structures, towards functionalization; and (d) exploitation of quantum coherence in a comprehensive attack on quantum information processing.

The institutes and departments from HZB participating in the Program FIT are:

- Institute for Methods and Instrumentation in Synchrotron Radiation Research (G-ISRR), Head: A. Föhlisch
- Working Group Magnetisation Dynamics (M-AMD Head (acting): O. Rader
- Working Group Quantum Phenomena in Novel Materials (M-AQM), Head: B. Lake

The HZB contributes specifically to the field of topological insulators through photoemission experiments and to the field of ultrafast spintronics and magnetism using femto-second pump-probe techniques. Both activities are directly related to the research possibilities provided by the synchrotron source BESSY-II operated by HZB. These activities are part of Topic 2: Controlling Spin-Based Phenomena and are covered by 7 of 55 FTEs. The HZB contributes also to Topic 4: Controlling Collective States, through 10 of 67 FTEs, investigating highly entropic and topological states in frustrated lattices of bulk materials, making explicit use of the neutron facility at HZB.

2.4 Scientific Results

2.4.1 PGI - Quantum Theory of Materials (PGI-1 / IAS-1)

Head of Institute: Prof. Dr. Stefan Blügel

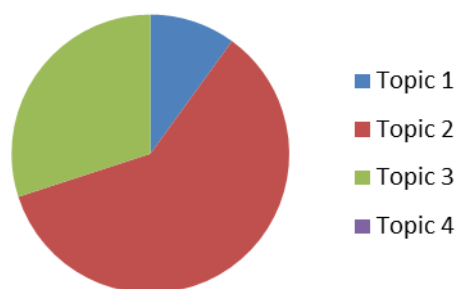
Contributing Personnel (31 Dec 2016 in FTE):

Core-funded: 14 scientists, 11 doctoral students, 1 scientific support personnel

Third-party funded: 15 scientists, 2 doctoral students, 0 scientific support personnel

Contributing Principal Investigators: Nicolae Atodiresei, Gustav Bihlmayer, Christoph Friedrich, Marjana Lezaic, Samir Lounis, Phivos Mavropoulos, Yuriy Mokrousov, Bo Persson, Daniel Wortmann

The hallmark of the research of the Peter Grünberg Institute PGI-1 / IAS-1 – *Quantum Theory of Materials* is the computation and the analysis of microscopic properties of realistic solid-state systems from the basic principles of quantum mechanics in terms of both basic science and practical applications in collaboration with experimentalists. Emphasis is on quantum materials, functionalities and effects relevant to nanoelectronics.



A major asset of our institute is the competence in developing conceptual and computational methods based on static and time-dependent density (DFT) and reduced density matrix functional theory (RDMFT), many-body perturbation theory, Kubo-type linear response and ab-initio Boltzmann transport theory as well as scale-bridging atomistic spin dynamics, paying particular attention to massively parallel high-performance computing that provides petaflop performances. Jointly with e.g., Stefano Baroni, Elisa Molinari, Nicola Mazzari, Pablo Ordejon and Thomas Schulthess we are member of the European Centre of Excellence *MaX: Materials Design at Exascale* (www.max-centre.eu). In developing these methods, we benefit from being part of the Institute for Advanced Simulation (IAS), e.g., through collaboration with **PGI-2/IAS-2** or the Jülich Exascale Labs, and from being part of the Jülich Aachen Research Alliance, section High-Performance Computing (JARA-HPC). We provide training and transfer of knowledge of our tools and methods through CECAM-workshops and interact with other PoF programs of the research field Keytechnologies.

PGI-1/IAS-1 mainly contributes to Topic 2.

PGI-1 is an international core contributor to the development of the new research field **spinorbitronics** through research on spin-orbit related phenomena of electrons such as **topological insulators**, **skyrmions** and associated **transport phenomena**. We contributed to the establishment of the National Priority Programs *DFG-SPP 1666: Topological Insulators* and *DFG-SPP 2137: Skyrmionics*, we are members of the EU-FET open program MAGicSky coordinated by Albert Fert and Vincent Cros and the DFG funded Collaborative Research Center CRC 1238: *Control and Dynamics of Quantum Materials*, CRC 917: *Nanoswitches* and hold an ERC consolidator grant (S. Lounis). The activities on topological insulators and skyrmions are in close collaboration with **PGI-3**, **PGI-5**, **PGI-6** and **PGI-9** and are central subjects of the Jülich Aachen Research Alliance, section Fundamentals of Future Information Technology (JARA-FIT). Furthermore, the **Virtual Institute for Topological Insulators** (VITI) fosters tight collaborations with RWTH Aachen University, University of Würzburg and the Shanghai Institute of Microsystem and Information Technology (SIMIT). The three Helmholtz Young Investigator groups (M. Lezaic, Y. Mokrousov, S. Lounis) and the DFG-funded Emmy

Noether group (N. Helbig) have been blessings to PGI-1. Bo Persson continued to expand on his role as a leading authority on tribology, rubber friction, adhesion, contact mechanics and seals. The knowledge gained through fundamental research at PGI-1 is transferred to many companies via [MultiscaleConsulting](#).

With this research portfolio, the researchers contribute with major results to **topic 2 Controlling spin based** and **topic 3 Controlling configuration based phenomena** of the PoF III program Future Information Technology (FIT). Some highlights and milestones are presented in the following.

Fermi surface mediated interactions and density oscillations on surfaces

Atomic-scale structures on surfaces allow very direct control of information stored in their local or collective charge or spin state. Pivotal here are long-range density oscillations that allow for both distant coupling and non-local switching. As we have found, it is possible to tailor the Fermi surface of the electron liquid by quantum confinement or doping so as to optimize these effects. In this respect, the anisotropy of the Fermi surface is essential, allowing for flat, nested regions that produce spin or charge density oscillations with almost no dephasing and with strong directional dependence around impurities. This *focusing effect* extends beyond the conventional theory of Friedel oscillations and has a number of ground breaking applications. Our theory and calculations are corroborated by experiments. This is a contribution to *Topic 2*.

Gigantic, spin-density oscillations were found in specific directions around an oxygen defect on ferromagnetic Fe ultrathin films grown on W(001) [1]. Confinement-generated quantum well states in the Fe films form flat Fermi contours, amplifying the strength of the oscillations and transmitting spin information to several tens of nm, which is very untypical for transition metals. **Topological insulator surfaces** (Bi_2Te_3) host a Fermi contour that can be tuned, by doping, to a hexagonal shape with excellent nesting conditions. Then, the focusing mechanism causes undamped directional density oscillations around magnetic defects, observed in a range of >30 nm. **The Kondo scale in interacting Fe dimers** buried under the Cu(001) surface, is directly related to long-range directional magnetic interactions, due to a manifestation of the focusing effect. We revealed that the interatomic distance and the direction of the dimer's axis are essential ingredients in enhancing or reducing the Kondo temperature [2]. **Chiral magnetic order**, driven by the Dzyaloshinskii-Moriya interaction (DMI), emerges at the atomic scale among at adatom or atoms in thin magnetic films. We explored this new paradigm together with the STM group of Roland Wiesendanger documented in about 14, partly high-impact publications between 2007 and 2017. For example, among Fe adatoms on the Pt(111) surface, we found, in agreement with experiment, that the DMI amplitude oscillates as a function of the adatoms separation. The DMI vector changes direction indicating the possibility of designing bottom-up magnets with tuneable chirality and non-collinear magnetization states [3].

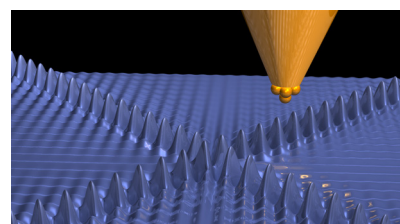


Fig. 1: Focusing of giant spin-filtered Friedel oscillations in directions defined by multiple flat energy contours in Fe films grown on W(001).

- [1] M. Bouhassoune, B. Zimmermann, Ph. Mavropoulos, D. Wortmann, P. H. Dederichs et al., *Quantum well states and amplified spin-dependent Friedel oscillations in thin films*, Nature Communications **5**, 5558 (2014) [doi:10.1038/ncomms6558](https://doi.org/10.1038/ncomms6558).
- [2] H. Prüser, D.E. Dargel, M. Bouhassoune, R.G. Ulbrich, T. Pruschke et al., *Interplay between Kondo effect and RKKY interaction*, Nature Communications **5**, 5417 (2014) [doi: 10.1038/ncomms6417](https://doi.org/10.1038/ncomms6417).

- [3] A. A. Khajetoorians, M. Steinbrecher, M. Ternes, **M. Bouhassoune, M. dos Santos Dias et al.**, *Tailoring the chiral magnetic interaction between two individual atoms*, Nature Communications **7**, 10620 (2016)
[doi: 10.1038/ncomms10620](https://doi.org/10.1038/ncomms10620).

Organic molecules at surfaces: from electronic structure engineering to functions

Hybrid molecule-surface systems obtained by combining the complementary physical properties of organic molecules and magnetic/non-magnetic surfaces offer an interesting perspective with respect to novel materials for green energy and quantum technologies. As an example, we discovered a new class of two-dimensional (2D) **molecular quasi-crystals** on a metal surface [1] as a consequence of the subtle interplay between the chemical molecule-surface vs. the van der Waals (vdW) interaction between molecules. In a different example, we explored how the interaction of π conjugated molecules with magnetic surfaces locally changes the substrate properties and proposed concepts such as the **magnetic hardening** [2] and **softening** [3] of the exchange interactions between the magnetic

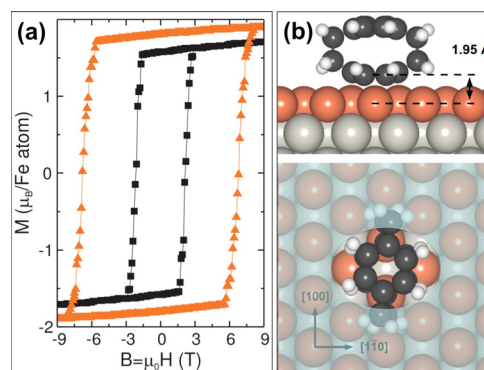


Fig 2: (a) illustrates the opening of the magnetic hysteresis curve (orange color) in comparison to the clean surface (black color) of Fe/W(110) due the interaction with a π -conjugated non-magnetic paracyclophane molecule (b).

surface atoms below molecules. Both effects have been successfully used in practice to (i) build spintronic devices with a large magneto-resistance even close to room temperature [4] and (ii) mediate a long-range interaction between molecular magnetic units embedded within a skyrmion lattice [5]. **Intercalation** of electropositive (Eu) or electronegative (O) atoms between a 2D material and a metal substrate are powerful means to modify the spatial extent of the π electronic states of a 2D π system. On this basis, we demonstrated that the strength of the vdW interactions acting between a finite π molecular system ($C_{10}H_8$) and graphene could be engineered by this doping mechanism [6]. We enabled a detection and manipulation of localized *4f* atomic-like orbitals [7] due to an efficient overlap between the large spatial extent of the π orbitals of organic molecules and graphene with the atomic states of rare-earth metals. This is a contribution to *Topic 2 and 3*.

- [1] M. Paßens, **V. Caciuc, N. Atodiresei, M. Feuerbacher, M. Moors et al.**, *Interface-driven formation of a two-dimensional dodecagonal fullerene quasicrystal*, Nature Communications **8**, 15367 (2017)
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Controlling spin-, charge- and configuration-based phenomena in oxides

In transition-metal oxides, the control of transport properties with the spin and charge degrees of freedom opens many possibilities for new applications. Understanding how we can engineer new oxides with desired charge distributions is essential for this purpose and a central issue in *Topic 3*. E.g., in **perovskite $PbXO_3$ compounds** ($X = 3d$ transition-metal element) the energy of the $6s$ levels of Pb, with respect to that of the $3d$ levels of X , determines the specific charge distribution over the cations of the compound. The series begins with $Pb^{2+}Ti^{4+}O_3$ and $Pb^{2+}V^{4+}O_3$, and ends with $Pb^{4+}Ni^{2+}O_3$, i.e., the oxidation states of the cations are interchanged along the X series. In a combined theoretical and experimental effort, we found that the members of the series with $X=Cr, Co$ induce very particular charge-distribution patterns, namely $Pb^{2+}_{0.5}Pb^{4+}_{0.5}Cr^{3+}O_3$ [1] and $Pb^{2+}Pb^{4+}_3Co^{2+}_2Co^{3+}_2O_{12}$ [2]. These charge distributions can be sensitive to pressure, e.g., a metal-insulator transition can be induced in $PbCrO_3$ in this way.

Also in **resistively switching oxides** the resistive state depends strongly on the charge state of the involved transition metal, which is controlled by the concentration of oxygen vacancies in the sample. In the forming step a high voltage is applied in order to form conductive filaments within an insulating matrix. Their resistive state can be easily switched with a small voltage afterwards. Within the *SFB917: Nanoswitches*, together with the experimental group of the PGI-7 we study the resistive-switching mechanisms in different systems. E.g., in Fe-doped $SrTiO_3$ X-ray photoemission and absorption spectra were compared to the ones obtained theoretically for different kinds of defects, yielding various conclusions on the most likely defects contained in the samples [3]. Moreover, we identified different types of extended defects in TiO_2 and simulated their electronic structure by density functional theory calculations [4].

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Topological insulators and related phases

Topological concepts have entered many areas of condensed matter theory to classify periodic solids and are a core theme of *Topic 2*. The first proposals of **topological insulators** (TIs), inspired by Haldane's work on the quantum Hall effect, predicted dissipation-less *spin* currents at the edges of two-dimensional (2D) materials. Density functional theory (DFT) calculations turned out to be an indispensable tool to detect the band inversions responsible for the non-trivial topology of the electronic structure of TIs and led to countless materials predictions in the last years. Recently, the interest extended to the **quantum anomalous Hall insulator** (QAHI) state, which shows dissipation-less *charge* currents at its edges. The TI-QAHI transition can be triggered by an external magnetic field, but in practice the necessary field strength is difficult to reach. Chemical modification of 2D TIs can be used to create intrinsic exchange fields that are sufficiently strong to realize this phase, e.g., a hydrogenated Bi(111) bilayer can be driven to that state [1]. While the topology of the TI is

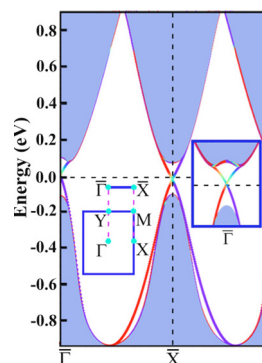


Fig. 3: Edge band structure of a TISe monolayer. The color indicates the spin direction [2].

derived from time-reversal symmetry (and consequently destroyed by magnetic fields), also crystalline symmetries can be used to define topological invariants. They characterize **topological crystalline insulators** (TCIs) that have been found in two- and three-dimensional materials. Even phase transitions between TCIs and TIs have been predicted with a corresponding change of the edge channels [2]. In a collaborative effort in the Virtual Institute for Topological Insulators (VITI), with colleagues from PGI-6 and PGI-9, we discovered a material that combines the edge states of a TCI on some surfaces with those of a weak TI on others [3]. This so called **dual topological insulator** offers the possibility to control the conductivity on different surfaces by breaking the respective symmetries (e.g., by strain or a magnetic field).

While DFT yields accurate ground-state properties, it is less reliable in the description of excited states. In TIs, strong spin-orbit coupling (SOC) causes a band inversion that leads to an energy gap of the order of a few hundred meV. Such small band gaps require a reliable description of the electronic structure **beyond DFT**, like that provided by the *GW* approach. We have implemented the **quasiparticle self-consistent GW** (QS*GW*) scheme with the SOC included throughout the full self-consistent cycle [4]. Results from this method provided a benchmark for similar implementations around the world and are a starting point for the treatment of SOC in *GW* calculations at different levels of approximations [5]. We showed that the widely used inclusion of SOC through a *posteriori* correction is not appropriate for some TIs. Our *GW* approximation has allowed us to characterize very accurately the band structures and plasmonic excitations of some of the most promising TIs.

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Skyrmions – two-dimensional topological magnetic solitons in chiral magnets

Magnetic skyrmions in chiral magnets are stable localized two-dimensional excitations with particle-like properties and are at the core of *Topic 2*. They have the great potential of becoming the smallest bit of information in a racetrack type solid-state memory. Their properties are determined by exchange interactions in competition with the **Dzyaloshinskii-Moriya interaction** (DMI) occurring in solids and interfaces with a lack of bulk or structure inversion symmetry, respectively, in combination with spin-orbit interaction. We contributed to the key challenge of the community, which is the design of small (<10 nm) skyrmions that are stabilized at the interface at room temperature by systematically investigating the DMI of all interfaces between magnetic 3*d* and spin-orbit active 5*d* metals [1], by proposing small skyrmions by a new concept focussing on chiral magnets with additional **exchange frustration** (e.g., Mn/W(100)), whose skyrmion phase is reached employing the **exchange bias** effect [2], and by design of **heterostructures** of period *n* incorporating nonmagnetic 4*d* metals (e.g., Rh) such as in [Rh|Fe|Pt]_{*n*} [3].

Detection of skyrmions is the second important issue to which we contributed. We have been very successful in the prediction and analysis of the transport properties of skyrmions from first

principles, in particular the **topological Hall effect** of ferromagnetic as well as antiferromagnetic skyrmions [4] (see also [4] of next section), and in the proposal of two protocols for the detection of skyrmions, (i) by all electrical means via the **tunnelling spin-mixing magnetoresistance** (TXMR) [5] or optically with X-ray spectroscopy exploiting the **topological orbital magnetization** (TOM) [6]. TXMR has immediate implications for device concepts, as it can reach efficiencies of 20% and can be used within a current perpendicular-to-plane geometry. X-ray spectroscopy can be used to single out the chiral contribution to the orbital magnetization of a skyrmion [6] induced by the **scalar spin chirality** $\mathbf{S}_1 \cdot (\mathbf{S}_2 \times \mathbf{S}_3)$ of non-coplanar spins, which converges in the adiabatic limit to the flux of the emergent magnetic field characterizing the magnetic skyrmion. The chiral orbital moment is thus proportional to the **topological charge** N_{Sk} . Exploiting the distinct properties of the chirality-driven and spin-orbit-coupling-driven orbital moments we defined the **topological orbital magnetization ratio** (TOMR), $\text{TOMR} \approx M_{\text{orb}}(\text{chiral})/M_{\text{orb}}(\text{F}) \propto N_{Sk}$, as a new measure accounting skyrmion matter, which is detectable by driving the sample from a reference ferromagnetic state (F), into a possibly unknown chiral state. If the TOMR is finite, there is a strong signature for the topological character of the magnetic structure.

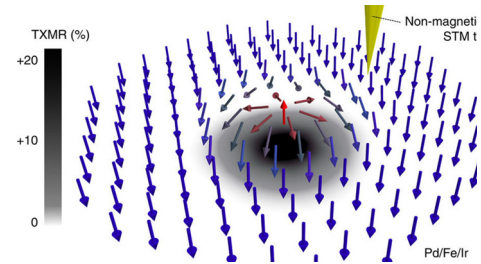


Fig 4: Expected TXMR efficiency when approaching a skyrmionic defect in Pd/Fe/Ir near the bias voltage 0.5 eV.

We have conjectured the existence of new particles, e.g., **antiskyrmions** coexisting with skyrmions in chiral magnets as well as a three-dimensionally localized thermodynamically stable magnetic hybrid particle, termed **chiral bobber** [7], which is made of a skyrmions-like soliton and an atomic scale cluster of non-collinear magnetic atoms, and which is expected to occur at the surfaces or interfaces of films made of bulk chiral magnets, e.g., the B20-material FeGe. The latter was observed recently in PGI-5 using electron holography. Both open a path to the realization of a new generation of magnetic solid-state memory.

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Topological Spinorbitronics

One of the most prominent and technologically relevant effects in modern spinorbitronics (Topic 2) is the effect of **spin-orbit torque** (SOT), which is exerted on the magnetization when an electric field is applied to a ferromagnetic system with spin-orbit interaction and broken inversion symmetry in bulk, at surfaces or interfaces, used to switch the magnetization of a ferromagnetic

layer. One of our key discoveries in this area is the strong anisotropy of the SOT and its generally complex dependence on the magnetization direction, arising as a result of intricate competing physical contributions, which at the end enhance its functionality [1]. Another breakthrough is the formulation of the SOT within the linear response formalism in terms of the Green functions of the disordered system and the velocity operator uncovering the topological origin of the SOT, which let us develop approaches to determine the SOT, based on an extrinsic (Boltzmann) and intrinsic formalism, in analogy to the Hall effects. This powerful approach, was applied to describe and understand the SOT in various interfacial and bulk materials [2], as well as to formulate and compute a whole palette of SOT-driven novel effects such as thermal, inverse, and thermal inverse SOT [3]. We have also made the discovery that the SOT is fundamentally **related to the Dzyaloshinskii-Moriya interaction**, providing thus a geometric interpretation as spin-orbit entangled phenomena taking place in collinear and chiral systems [4], as well as establishing the means for their precise evaluation in structurally and chemically complex disordered systems.

Antiferromagnetic spintronics is a potentially disruptive technology. In the past years, we have demonstrated that the antiferromagnets (AFMs) are ready to enter the realm of spinorbitronics. Firstly, we have shown that antiferromagnetic transition-metal materials are able to give rise to strong spin Hall effect and serve as a source of pure electric-field driven spin currents, which can be eventually utilized for exerting a sizeable SOT on a magnet, brought in contact to AFMs [5]. Secondly, we have made a seminal discovery that certain types of AFM materials can display a strong coupling of an electrical current with the local staggered magnetization via the SOT, which can lead to a purely **electrical switching of the AFM magnetization** [6]. These two discoveries opened the door to various prospects of the utilization of AFMs in relativistic-effect based applications.

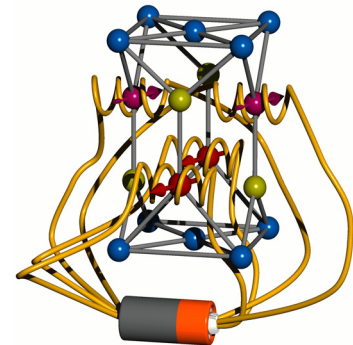


Fig 5: In an antiferromagnet, such as CuMnAs, an applied electric field via the effect of spin-orbit torque can exert effective fields which are staggered on each of the magnetic sublattices, thereby causing a reorientation of the AFM magnetization [6].

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Ultrafast Spintronics

The terahertz (THz) frequency in the electromagnetic spectrum provides a perfect window to many fundamental resonances of materials. An example are the **large intrinsic zero-point fluctuations** of the magnetic moment in single adatoms on surfaces, which are of quantum mechanical nature and can now be calculated from DFT [1]. They **overcome the magnetic anisotropy potential barrier** even at absolute zero temperature, which destabilizes the

magnetic moment (Fig. 6). We provided practical guidelines for designing magnetically stable nanomagnets with minimal quantum fluctuations. Building clusters made of strongly coupled magnetic atoms, for example, lowers the strength of the fluctuations, which explains that the smallest Fe nanostructure that is magnetically stable on Cu(111) contains five atoms [2]. Also, the concept of zero-point fluctuations unravels the striking non-observation of magnetic patterns in Mn nanowires on Ni(110) [3]. This part contributes to *Topic 2*.

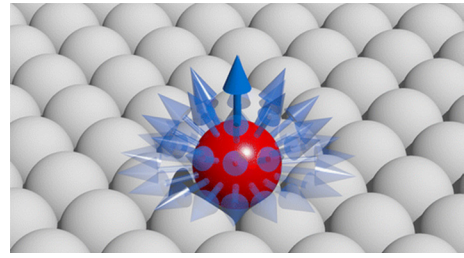


Fig. 6: A sketch of the atomic magnetic fluctuations of zero-point nature.

A novel twist for THz applications lies in the realm of spintronics, with the vision of transforming conventional GHz- into ultrafast THz spintronics. We performed first-time investigations of **dynamical magneto transport** properties, up to the THz regime and beyond, based on the realistic electronic structure of materials. We demonstrated that AC magneto-resistances together with the Hall effects could be greatly magnified or dwindled by tuning the frequency of the AC current and the applied magnetic field [4]. For example, the effective conversion from charge to spin currents, described by the spin Hall angle, can be boosted by more than two orders of magnitude. A breakthrough in the field came with our recent works on metallic interfaces of ferromagnets with non-magnets, for which the properties of generated currents and emitted THz radiation can be efficiently engineered by tuning the electronic structure of the interfacial systems. We have suggested two possible channels for the generation of ultrafast THz spin and charge currents as a result of interaction with femtosecond laser pulses. One is based on the effect of **inverse SOT** (see [3] of spinorbitronics), the other is based on the **inverse spin Hall** effect [5,6].

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Future Goals and Directions

Quantum materials and spinorbitronics are competitive fields of great scientific dynamics, sources of conceptual and methodological innovation, and areas of scientific discovery. We plan to extend the scientific investigation from our experience in electronic properties of realistic materials, of relativistic spin-orbit effects and nontrivial topologies to the seven dimensions of real and momentum space as well as time. On the agenda are two-dimensional solids, spin objects with complex gauge groups such as skyrmions in non-collinear systems, chiral and ultrafast spinorbitronics. In collaboration with scientists from RWTH Aachen and PGI-5 we search for three-dimensional magnetization solitons such as hopfions as particle for a truly three-dimensional solid-state memory and new memristive functionalities based on spin. In collaboration with the PGI-2, PGI-9 and the University of Cologne, we explore hybrid topological

materials, which are topologically non-trivial in many spaces. One example is the investigation of topological superconductors to host Majorana fermions. This research direction is routed in *Domain 1: Exploring New Paradigms and Device Concepts for Future Information Technology* of the future PoF IV *Program 2: Information Processing: Physical Systems, Biosystems, and the Brain* and contributes from the viewpoint of quantum materials to the *Domain 2: Fundamentals of Quantum Computing* and *Domain 3: Network Dynamics and Neuromorphic Computing* and interfaces strongly with the RWTH-Aachen University through JARA and the University of Cologne and Bonn.

Materials discovery lab – Computer. The discovery of new technological materials offering new functionalities is a slow and intensive process. The 21st century offers powerful computer architectures, new concepts in computer science analysing data or turning scientific texts into knowledge graphs. We take this opportunity to develop in collaboration with the newly founded *Centre for Simulation and Data Science (JARA-CSD)* of the RWTH-Aachen University and the Forschungszentrum Jülich a materials discovery lab enabling cognitive materials discovery and virtual materials design. Our focus is on electronic, magnetic and renewable energy materials. One step to achieve this goal is to extend and translate our scientific first-principles core methods (see www.juDFT.de), into stable and robust infrastructures that can be used by a wide community for the simulation of electronically and structurally increasingly complex materials. Efficiency on modern modular high-performance computers with accelerators and boosters will be achieved in collaboration with the Jülich Exascale labs. Incorporating these methods into a workflow environment such as the AiiDA framework (www.aiida.net) paves the way for automated materials screening and systematic materials to property design. This research directions connects the future PoF IV *Program 2: Information Processing: Physical Systems, Biosystems, and the Brain* with the future PoF IV *Program 1: From Data to Knowledge and Action* and *Program 3: Materials Systems Engineering 4.0* enabling hereby collaborations with many institutes of the Forschungszentrum Jülich including those in the Institute of Energy and Climate Research (IEK).

2.4.2 PGI - Theoretical Nanoelectronics (PGI-2 / IAS-3)

Head of Institute: Prof. David P. DiVincenzo

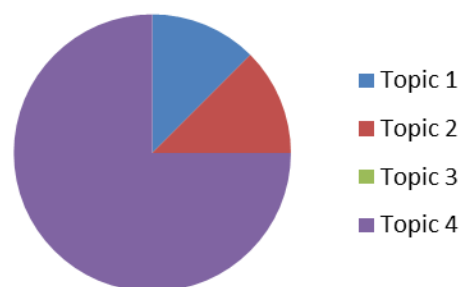
Contributing Personnel (31 Dec 2016 in FTE):

Core-funded: 13 scientists, 0 doctoral students, 1 scientific support personnel

Third-party funded: 1 scientists, 1 doctoral students, 0 scientific support personnel

Contributing Principal Investigators: Rudolph Zeller, Maarten Wegewijs, Theo Costi, Eva Pavarini, Gianluigi Catelani.

Our unit pursues a diverse set of topics that further our theoretical understanding of electronic systems and devices at nano- and atomic scales, and in the application of these small quantum systems to new forms of information processing. The quantum properties of individual entities are an established theme. Researchers have experience both in strongly- and weakly-correlated matter. Strong correlations result in the emergence of new kinds of spin- and orbital-ordering in oxide compound crystals. Fast quenches of parameters in single-impurity problems lead to new manifestations of the Kondo effect. Weakly correlated matter, including many of the materials that are vital in the information processing world today, give us the opportunity to develop computational techniques that permit simulations of unprecedented scale and realism.



PGI-2/IAS-3 mainly contributes to Topic 4.

PGI-2 has been a central participant in the development of new paradigms in quantum information science. Quantum coherence is a vital resource for accomplishing qualitatively new tasks. In spin qubits, the spin of electrons in single-electron quantum dots can serve as a platform for quantum computing, and the group studies mechanisms for effective quantum gate operations in this system. Studies are done of other prospective qubits, including those available in superconducting devices, with detailed theories of quasiparticle dynamics assisting in current experimental investigations. Concepts for multi-qubit architectural approaches are developed in close collaboration with experimentalists. Other aspects of the full-system problem in quantum computation are also research, with a strong current focus on non-reciprocal solid-state devices such as microwave circulators.

We organize this description of PGI-2 activities under the overall topics of “**correlated electron physics**” and “**quantum computing**”, with examples of selected specific accomplishments.

Correlated Electron Physics

Following on long experience in theoretical modelling of orbital- and spin-ordering phenomena in d^1 perovskites such as LaMnO_3 , CaVO_3 , YTiO_3 and YTiO_3 [1], Eva Pavarini and coworkers have now solved the correlated electronic structure of the compound Sr_2RuO_4 . This Ruthenate has been of intense interest in the exploration of topological superconductivity, with the high likelihood that it, unique among the

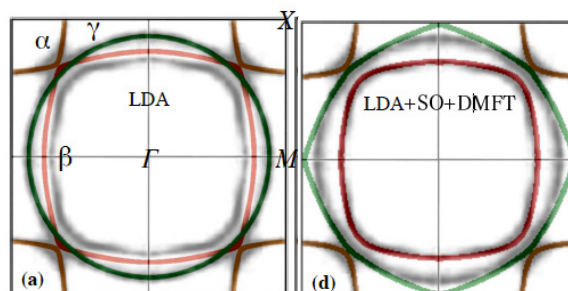


Fig 1: Fermi surfaces of Strontium Ruthenate, showing the difference between a calculation without correlations (LDA, left), and with correlation corrections (DMFT).

perovskites, realizes triplet-pairing, p-wave superconductivity, which could support fractionalized excitations and form the basis of topological quantum information processing. However, the highly correlated normal state of this material has not previously been well understood. In [2], the group finds that the Fermiology of the material is strongly influenced by spin-orbit and by electron-correlation effects. While the Local Density Approximation gives qualitatively the right fermi-sheet structure, the inclusion of spin orbit, and correlations via the Density Matrix Functional Theory (DMFT) gives quantitatively a different structure of these surfaces (Fig. 1). The computations support the point of view that the Cooper pairs in the superconducting phase have strong spin-orbital entanglement.

Strongly connected with this area of research, Pavarini and associates have run a regular Jülich Autumn School on Correlated Electrons every year for six years (2011-16), attracting a capacity group of around 70 students each year. This activity has been very successful and is expected to continue.

Rudolph Zeller continues to make unique contributions to large-scale, massively parallelizable electronic structure calculations within the KKRNano project. He has made methodological discoveries [3, 4] that decidedly improve the practical and fundamental convergence properties of the KKR method. He continues to assist in incorporate these fundamental insights into the material simulation work of PGI-1.

Theo Costi continues to expand on his role as a leading authority on all aspects of the application of Wilson's Numerical Renormalization Group (NRG) method. A primary focus in recent years has been to adapt the NRG to time dependent problems, so that systems subject to a "quantum quench" (i.e., abrupt change of the coupling parameters in a quantum impurity system) can be simulated reliably. After some years of painstaking method development and testing of approximations [5], this method is ready for application for a variety of applications in ultra-fast quantum phenomena. His work gives a clear view of the temporal formation of the Kondo screening cloud after the parameters of a quantum impurity are switched into the Kondo regime.

In a recent application, Costi's group, in collaboration with workers in the JARA partnership at RWTH Aachen University, has done a wide-ranging comparison [6] of the efficacy of different many-body methods for solving a canonical model in decoherence theory, the Ohmic spin-boson model. The methods tested, besides his time-dependent NRG, were the Density Matrix Renormalization Group technique (DMRG), the non-interacting Blip Approximation (NIBA), and the functional renormalization group method (FRG). Fig. 2 shows one of the many comparisons made, showing that for certain values of the dissipation parameter α , the TD-NRG method gives a much more accurate result for the decay of two-state coherence than the time-honored NIBA technique.

Maarten Wegewijs has extended the expertise built up by his former YIG group. Working with a Humboldt fellow, a new magnetic proximity effect of tensorial nature was predicted [7], illustrated in Fig. 3. Unlike known vectorial exchange fields, this allows for the creation of magnetic anisotropy in otherwise magnetically-uninteresting molecular transistors. Soon afterwards, effects of this kind were measured by STM in atomic-scale transistors. Entirely new directions have been explored, e.g., by establishing a framework for geometric and topological phases that are specific to open quantum systems in connection with counting statistics, as well as the incorporation of rigorous insights of quantum-information theory into diagrammatic techniques for open-system dynamics.

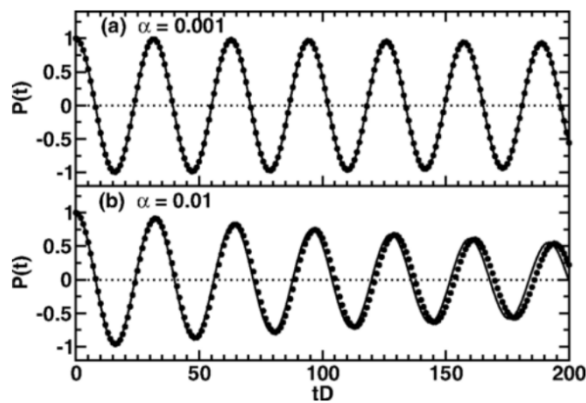


Fig 2: Decay of coherence in the spin Boson model for two different damping parameters [5]. Subtle effects at higher damping are captured accurately by the NRG technique.

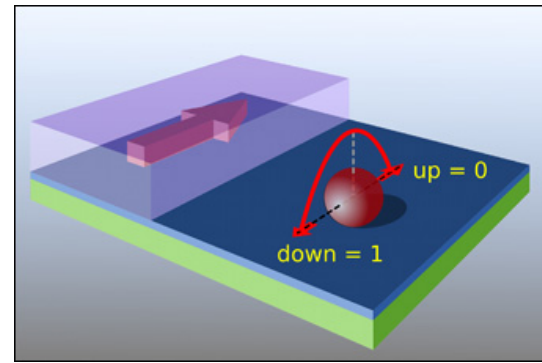


Fig 3: Pure quadrupolar spin exchange: close to a magnet, a molecular transistor develops a spin-reversal barrier (red). Surprisingly, the transistor's spin remains bistable – storing a bit of information in an unbiased way – even though the magnet's orientation seems to favor state 0.

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Quantum Computation

Problems in the theory of quantum computation and other areas of quantum information science have been increasingly central to the work of PGI-2 over the last five years. Work has included the theoretical exploration of semiconductor quantum dot qubits and their optimal functionality, superconducting qubit operation, including detailed evaluation of decoherence properties, exploration of remote entangling couplers for effective use in multi-qubit architectures, and study of crucial microwave devices for the functioning of solid-state quantum computing systems.

In the context of a joint program conducted with the renowned group of Prof. Schoelkopf at Yale University since 2014, Gianluigi Catelani and coworkers have systematically modelled and explored the effect of quasiparticles in superconducting qubits (Fig. 4). In earlier work, Catelani had shown that

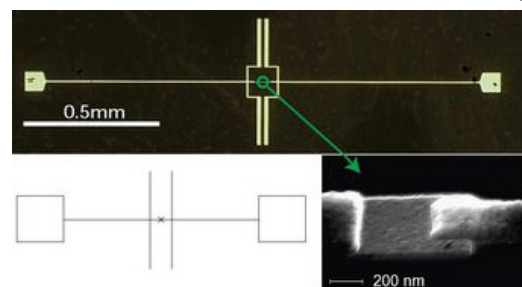


Fig 4: Quasiparticles traps positioned at the far ends of a modified 3D transmon qubit. Placement of a magnetic vortex in the end regions cause trapping of quasiparticles, keeping them away from the qubit junction (center) and improving the qubit coherence. [1,2].

quasiparticles passing through the Josephson tunnel barrier had a severe effect on the coherence of these qubits; much further work has been done to characterize this process and the devise schemes for suppressing them. In [1] and the accompanying theoretical analysis [2], progress has been made in ameliorating the quasiparticle mechanism.

This work has shown that quasiparticles can be trapped in magnetic vortices. Systematic experimental study of the positioning of the trapping region, and the presence or absence of vortices, and, more recently, of superconducting gap engineering in the device, systematically vary the coherence properties. A series of theoretical studies [2] indicate further device strategies that will extend the qubit lifetime further.

In a separate collaboration with the group of Prof. Oliver at MIT, Catelani has provided further insight [3] into quasi-particle physics by proving that they can be intentionally pumped away from regions of the device where they do the most harm (Fig. 5). The puzzling observation in the experiment of non-exponential coherence decay turns out to be attributable to switching between different (small) numbers of quasiparticles per qubit island, leading to a superposition of different exponential decay laws. When the theoretically constructed pumping sequence was devised, qubit coherence was accordingly enhanced in the experiment.

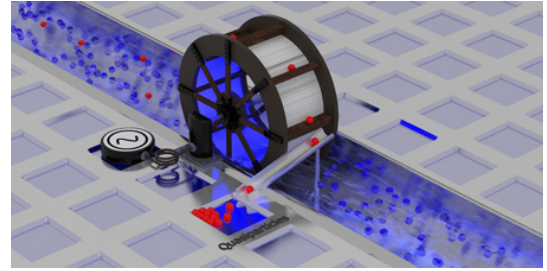


Fig 5: Artist's conception of a flux qubit subject to quasiparticle pumping.

In the work-group of DiVincenzo, an ongoing highlight has been development of theoretical tools for the development of effective Hamiltonians for qubit systems. Many analyses of models for system-environment interactions begin with a derivation of an effective Hamiltonian in a reduced Hilbert space. A variety of elimination methods have been employed for this purpose in the previous literature. The contribution here has been to make a unified procedure for performing these model reductions, based on the formalism of Schrieffer and Wolff. The final result has been a long analysis [4] giving a comprehensive review of the necessary superoperator techniques that improves the efficiency of these calculations, adaptation of the technique to multipqubit networks, and rigorous results about the convergence of the resulting expansions.

The group has made many practical, conceptual contributions to the advancement of experiments in spin qubits. Having pioneered the idea of qubits encoded in angular momentum subspaces (the “three quantum dot” qubit [5]), we have continued to make progress in having long-distance couplers between these qubits. Extensively analyzed is the “QS” (“quantum state”) structure, which may be an elongated quantum dot, with one or a small number of discrete quantum levels, that is exchanged coupled to two qubits. In [6] a full analysis of many possible coupling structures is given, showing that high fidelity gates are possible. As illustrated in Fig. 6, schemes are given for specifically achieving the CNOT quantum gate, with interaction sequences that are shorter and simpler than in any previous work.

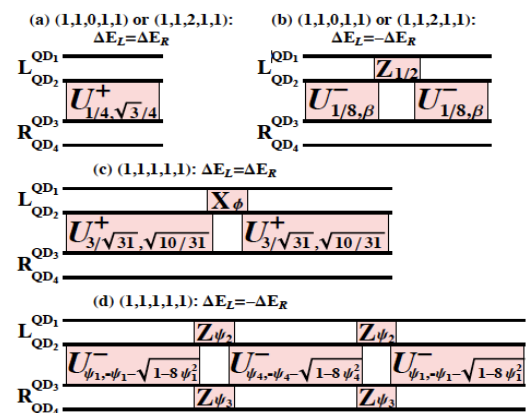


Fig 6: Compact implementation of the CNOT gate by small sequences of interactions mediated by a coupling quantum structure. For this example the quantum dot qubits are singlet-triplet qubits. See [6] for details.

The group has had a significant effort in the area of microwave devices that are essential in the control apparatus of the quantum computer. We have focused on the role of non-reciprocal devices. A new set of techniques and approaches in low-temperature physics are coming over the horizon that could eventually result in a first area of large industrial application of ultra-low temperature techniques ($<1\text{K}$), and be the route for scalable quantum computers. Several forms of solid-state qubits systems, notably single-electron quantum dots, superconducting devices based on the Josephson effect, and single-impurity spin states in semiconductors, have as a common element that they are coupled via quantum states of microwave radiation, and they are controlled and read out using high amplitude, classical radiation also in the microwave band. The handling of this classical radiation with an extraordinarily high level of precision, with the reduction of all forms of thermal and other noise, is a key enabler to using this technique to obtain high-fidelity multiqubit quantum logic operations.

The microwave circulator is an essential component in this toolkit. Presently in use is a form of circulator invented fifty years ago, based on the microwave Faraday effect. Its quasi-optical design means that it is very bulky and does its job only with marginally acceptable parameters. DiVincenzo has invented a new approach to the circulator, the capacitively coupled Hall-effect circulator [7] that uses plasmonic action to make possible the extreme miniaturization of this device, with the prospect of improved bandwidth and functional parameters. In addition to the publication, two patents have been granted in this area [8].

Finally, work has proceeded for many years, and right up to the present, on innovations in multi-qubit layout that will facilitate the most effective implementation of quantum error correction codes. In superconducting qubit systems, it has been very standard to represent the dynamics of the systems with a discrete electric circuit. But there has not previously been a systematic way to derive the appropriate circuit topology, or the component values, for an accurate circuit representation. We have done both in recent work [9], which relies on reviving decades-old electric networking theory and updating it for applications in the quantum context.

With this background, our group could confidently analyze multiqubit structures in error-correction layouts. In quantum error correction, small quantum computations are typically invoked so that the parity of a set of qubits (3 or 4) can be transferred to a single ancilla qubit, which is then read out. We have worked to replace this by a more direct quantum scattering process in which the multi-step process is replaced by a single interrogation of the system, resulting directly in the readout of a parity. Analyses of these ideas in various settings have led to promising conclusions [10, 11], and it is hoped that it will be ready for a direct experimental test in the near future.

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Planned New Directions

It is presently a very dynamical and transitional time for the PGI-2 with many new things to do:

- In the area of correlated electron physics, activities will proceed in uncovering novel orderings, spin, orbital, and other, in oxide materials. New applications for the Numerical RG method will continue to be studied.
- In the area of nanoscale transport, work will transition to applying techniques of quantum information theory to understand the physics of measurements in electronic physics. The concept of counting statistics will be given a quantum-measurement interpretation, with applications to the problems of real measurement noise for quantum dot qubits.
- Major new directions will open in the direction of quantum computation. While over the last five years that work has been carried on almost exclusively within PGI-2, new, expanded avenues of work will go on, but with a significant portion of the work being handed over to the JARA Institute for Quantum Information, PGI-11 (see PGI-11 report). The general strategy will be to leave the more speculative and longer-term activities in quantum information in the hands of PGI-2, while the activities that impinge directly on the problem of scaling up quantum computation will be transferred over to PGI-11, and built up anew.
- An activity to remain in PGI-2 is most of the work on the emerging style of qubit, topological qubits. In collaboration with workers in PGI-1 and at the University of Cologne, especially in the context of the currently proposed Excellence Cluster ML4Q (Matter and Light for Quantum Computing) in the German Excellence Initiative¹, future work will examine the topology of braided Floquet bands in periodically-driven topological structures, and will continue on the road to seek ways to make Majorana qubits more immune to various forms of environmental noise.
- A small amount of exploratory work will begin in the area of quantum communication; a current focus is to define a two-client, one-server setting in which trusted quantum oracle calculations can be done by the use of an untrusted quantum server; this work will define a possible near-term experiment in the area of three-node NV-center networks.
- Both PGI-2 and PGI-11 will participate in pan-European initiatives (QuantERA, and the Quantum Technology Flagship), again from the side of PGI-2 the contributions will tend in the direction of fundamental theoretical contributions from the quantum information direction.
- Scientists in PGI-2 will assist in the startup of more planned institutes in quantum information science, and stand ready to collaborate with new groups as they come to the Forschungszentrum.

¹ http://www.dfg.de/en/research_funding/programmes/excellence_initiative/

2.4.3 PGI - Functional Nanostructures at Surfaces (PGI-3)

Head of Institute: Prof. Frank Stefan Tautz

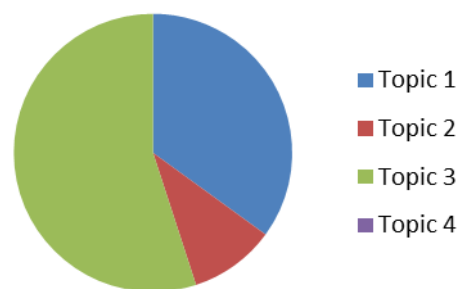
Contributing Personnel (31 Dec 2016 in FTE):

Core-funded: 10 scientists, 11 doctoral students, 5 scientific support personnel

Third-party funded: 1 scientists, 0 doctoral students, 0 scientific support personnel

Contributing Principal Investigators: Bert Voigtländer, Christian Kumpf, Sergey Subach, Vasily Cherepanov, Ruslan Temirov, Francois Posseik, Christian Wagner

The mission of PGI-3 is **exploring and extending the limits of nanoscale engineering**. The nanostructures we are concerned with are located at surfaces and interfaces. Nanoscale engineering at surfaces and interfaces is the basis for novel devices in quantum technologies, neuromorphic computing and green information technology, and hence our research contributes to the foundations of these. In particular, we investigate structural issues, interaction phenomena and charge transport at surfaces, interfaces and nanostructures of the relevant materials.



PGI-3 mainly contributes to Topics 1 and 3.

Our research covers single molecules, homo- and heteromolecular monolayers and thin films, single layers and stacks of novel two-dimensional (2D) materials, semiconductor nanostructures and topological materials. We apply well-established surface science techniques and also develop new methods such as new scanning probe techniques (STHM, SQDM, 4-tip STM, adiabatic demagnetization mk-STM), photoelectron tomography or parallel-detection electron energy loss spectroscopy.

Scanning probe manipulation of single molecules: from fundamentals to functions

(Contribution to Topics 2 and 3) The idea to freely control the atomic-scale structure of matter has intrigued scientists for many decades. Our research is centered on the controlled manipulation of molecules with low-temperature scanning probe microscopes (SPM). This technique enables us to engineer and spectroscopically characterize isolated molecules in metastable conformations [1] and supramolecular structures [2, 3]. Influential for the field was our experimental mapping of entire molecular adsorption potentials by which we revealed a superlinear increase of van der Waals interactions with molecular size due to many-body quantum effects [1]. This benchmark data demonstrates the accuracy and limitations of state-of-the-art DFT methods. To enable more complex molecular manipulation protocols and promote the intuition for molecular scale mechanics, we have developed a molecular manipulation lab (momalab.org) in which the SPM, a virtual reality interface, and a real-time capable simulation are directly connected [2, 4].

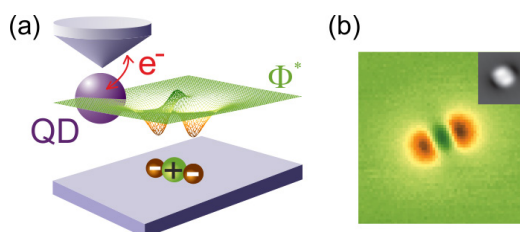


Fig. 1: (a) Operation principle of SQDM: The charge state of the tip-attached QD is determined by the potential Φ of a nanostructure. (b) SQDM image of a single PTCDA molecule (inset STM image).

A complementary benefit of our molecular manipulation approach is the development and patenting of two new SPM imaging techniques functioning according to the sensor-transducer principle [5, 6]. In

both techniques an individual molecule bound to the front tip-apex atom takes the role of a nanoscale sensor for forces [6, 7] or electric potentials [8], respectively. Here, large molecules that are weakly hybridized with the tip become molecular quantum dots (QD). Since this QD can be gated by the electrostatic field of surface-bound nanostructures, it enables imaging of such potentials with unprecedented sensitivity and resolution [8] (Fig. 1). Hence, Scanning Quantum Dot Microscopy (SQDM) is a rare example of a fully functional and readily applicable single-molecule, single-electron device.

To understand and control magnetic properties of single molecules and supramolecular structures we induced a Kondo effect in an all-organic compound by SPM manipulation or by surface chemical reaction with individual gold atoms. In the latter case, we were able to identify a new interaction channel between magnetic molecules leading to a quantum phase transition, [3]. Unlike the famous quantum critical point in the two impurity Anderson model found by Jones and Varma in the 1989, the new critical point is topologically protected, separating an underscreened *doublet* from a locally screened *singlet* ground state. It has remarkable attributes: Instead of the magnetic exchange interaction, the *non-magnetic* chemical interaction between the impurities drives the transition between distinct magnetic ground states.

For an even better access to low-energy phenomena such as molecular magnetism, we currently develop an SPM which allows measurements in magnetic fields of up to 8T and temperatures as low as 100 mK obtained without ^3He by adiabatic demagnetization.

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Metal-organic interfaces

(Contribution to Topic 3) Metal-organic interfaces are a topic of wide importance in nanoscale engineering, because at these interfaces “soft” molecular matter and crystalline condensed matter meet – the interaction between local electronic states in the molecule and extended ones in the metal leads to the emergence of interesting properties. Tailoring interface properties between these two limiting cases is a challenge, because unlike metal-on-metal epitaxy, where the building blocks are point-like single atoms, organic molecules add dimensionality, anisotropy and conformation [1]. Our research covers interfaces between metals and single-component organic layers as well as hetero-molecular systems, containing two or more molecular species. We focus on the interplay between geometric and electronic properties.

Benzene on coinage metal surfaces [2] is a recent example for the collaborative study of homomolecular metalorganic interfaces (with Professor A. Tkatchenko at the University of Luxembourg and Professor P. Tegeder at the University of Heidelberg). This study, based on normal incidence x-ray standing waves (NIXSW), temperature-programed desorption (TPD) experiments and first-principle calculations reveals universal binding energy trends for aromatic molecules. In reference [3], in collaboration with Professor Aeschlimann at the University of Kaiserslautern, we study hybridization-related modifications of the interactions across metal-organic interfaces, induced by alloying the uppermost silver substrate layer (by Pb in this case). We find that localized σ -type chemical bonds (not π -type or van der Waals bonds) are able to modify both geometric (relaxation of surface atoms) and electronic (band) structure. This provides an exciting platform for tuning the Rashba-type spin texture of surface alloys using organic molecules.

Combining different molecules (e.g., charge accepting and donating species) in the organic layers adds new design opportunities. We performed a trend-setting study for one such a heteromolecular adsorbate system, PTCDA + CuPc / Ag(111) [4]. In homomolecular layers both species are charge acceptors, but in an ordered monolayer blend CuPc changes its electronic character and donates charge to PTCDA via the metallic surface. This electronic modification goes along with at first glance counterintuitive structural changes, which however may be rationalized by lateral redistribution of charge density: The charge acceptor (PTCDA), although taking up more charge (i.e., electronically strengthening its surface bond), increases its adsorption height (i.e., the bond length) compared to a homomolecular layer, cf. Fig. 2, while the reverse happens for CuPc. Within a more comprehensive study we have also tuned the electron affinity of acceptors and donors in the molecular film by using other, similar molecular species [5, 6].

Furthermore, we have used the PTCDA+ CuPc system on Ag(111) to demonstrate an effective and generic way of steering the growth of two different planar aromatic components in the sub-monolayer coverage regime. Fig. 3 shows the first-layer phase diagram [1], compiled with our state-of-the-art aberration-corrected low energy electron microscope. The only prerequisite for this steering mechanism is the existence of mixed phases and repulsive intermolecular interactions of at least one molecular species. All other features of the phase diagram follow from standard thermodynamic considerations. Indeed, the proposed model describes fully the results for three different heteromolecular phases on highly ordered pyrolytic graphite reported in 2010 [1].

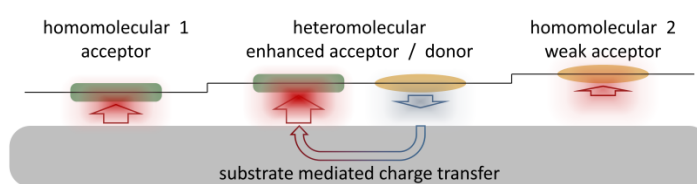


Fig. 2: Illustration of charge transfer and adsorption height alignment in PTCDA+CuPc/Ag(111).

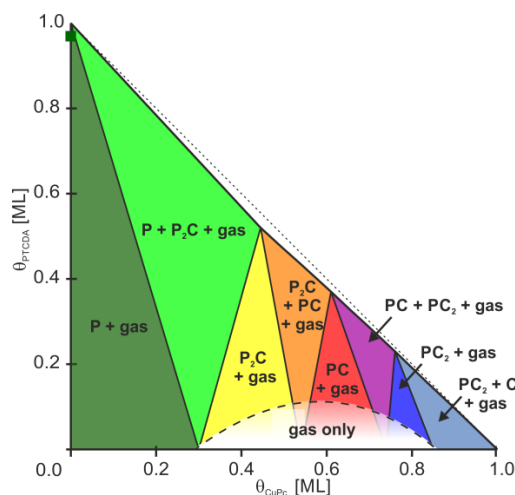


Fig. 3: Sub-monolayer phase diagram of PTCDA/CuPc/Ag(111)

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Visualization of the molecular wave-functions

(Contribution to Topic 1) Although electron wave functions are not, strictly speaking, quantum mechanical observables, we have undertaken a significant effort to observe them experimentally, in cooperation with Professors P. Puschnig and M. G. Ramsey at the University of Graz and Professor M. Richter at the Physikalisch-Technische Bundesanstalt in Berlin. In part, this was inspired by the long-standing question, raised by Nobel Laureates R. Hoffmann and K. Fukui decades ago, whether the single particle orbital, which has proved so powerful in chemistry, has a reality beside the all-important electron density that physicists tend to focus on (K. Fukui, Intern. J. Quantum Chem. 12 (Suppl. 1), 277 (1977), and R. Hoffmann, J. Am. Chem. Soc. 121, 3414 (1999)). A second, no less important motivation is provided by the rapidly developing quantum information technologies, demanding for a deeper conceptual understanding of the electron quantum state.

In this context photoemission spectroscopy is the method of choice, because it provides direct access to (occupied) electron states. We employ a combined experimental/theoretical approach, the photoemission tomography (PT) technique, which enables analysis of the photoemission angular distributions in terms of the relevant molecular orbitals [1]. Indeed, it allows reconstructing wave-functions of organic molecules, including their spatial distribution [2] and phase [3], from purely experimental data (Fig. 4).

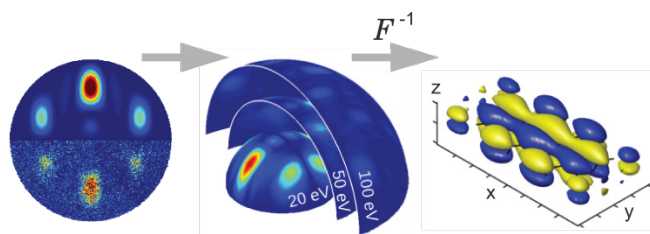


Fig. 4: Principles of a 3D orbital reconstruction: (left) Photoemission *k*-map of PTCDA LUMO (top part – theory, bottom part – ARPES), (middle) combining photoemission data measured at different photon energies, (right) spatial distribution of PTCDA LUMO, recalculated from experimental datacube using the inverse Fourier transform.

Besides its fundamental value, PT is successfully employed for application-relevant issues, too. For example, we evaluated the role of intermolecular and interfacial interactions [4, 5] at molecule/metal interfaces and resolved contributions of different species of binary molecular heterostructures [Ref. 3 of prev. sec.]. Furthermore, the precise data on energy level alignment obtained due to PT provide a stringent benchmark for ab initio electronic structure theory [1]. We have implemented the PT technique at several synchrotron light sources (Diamond, BESSY, Elettra) and at our photoemission electron energy microscope.

Further improvement of the technique aims at its broader usability for practically relevant systems, from small molecules to extended 2D materials, and on the application of alternative

excitation sources. In the framework of Sonderforschungsbereich 1083 at University of Marburg, we will utilize a high harmonic generation laser light source in the group of Professor U. Höfer for time-resolved photoemission tomography experiments.

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Interfaces of 2D materials

(Contribution to Topic 2 and 3) Internal interfaces are important for device functions in many contexts. In fact, it has been pointed out that "the interface is the device" (H. Kroemer). The growing interest in 2D-materials such as graphene, hexagonal boron nitride (hBN) and transition metal dichalcogenides (TMDC) has moved internal interfaces even more strongly into focus, because the layer-by-layer combination of different 2D sheet materials in arbitrary sequence could lead to exceptional material properties. In the extreme limit, if each sheet of such a heteroepitaxial stack of 2D materials is a different material, a new class of bulk "materials" emerges that consists exclusively of internal interfaces ("the interfaces are the material").

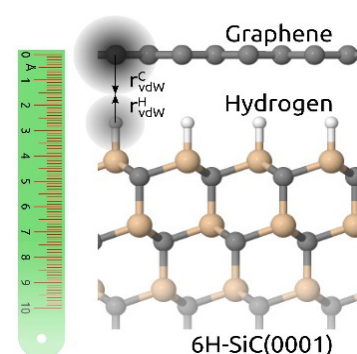


Fig. 5: Van der Waals radii of C and H are not overlapping for H-intercalated graphene on SiC.

To explore this approach to materials design, we have investigated graphene on the substrate SiC, a wide band-gap semiconductor that can readily be used to build devices. Graphene can be decoupled from its substrate by intercalation with, e.g., hydrogen. The graphene-substrate distance, obtained by the normal incidence x-ray standing wave (NIXSW) technique, is a direct measure of the interaction strength of graphene with the substrate. In collaboration with Professor U. Starke at the MPI for Solid State Research in Stuttgart we were able to show that H-intercalated graphene on SiC (Fig. 5) is, to date, the system with the weakest interaction (van der Waals) between epitaxially grown graphene and the substrate. Using DFT-vdW calculations in collaboration with Professor P. Rinke at Aalto University and Professor V. Blum at Duke University, we also found that the electron density is very close to the one of truly freestanding graphene [1].

For applications in information technology it is indispensable to dope the graphene layer in a controlled way. In particular, proximity doping is of great interest when designing materials based on atomically-thin 2D sheets. We have used the chemical sensitivity of NIXSW to show that the N-doping of graphene not only substitutes C by N in the graphene lattice, but that N atoms also partly replace the intercalated H [2, 3] (with Dr. Jelinek, Czech Academy of

Sciences). In fact, the latter contribute to the doping of graphene. We found similar results for B doping [4]. As a result, combining n-doping (N atoms) and p-doping (B atoms) allows a flexible tuning of the electronic properties of graphene [5].

Beyond structural properties, the phonon dispersion is very valuable for understanding the elastic, optical and thermodynamical properties of 2D materials. However, the bottleneck of existing electron energy loss (EELS) spectrometers is the acquisition speed. In collaboration with Professor H. Ibach (PGI-6), we have developed a new spectrometer for parallel acquisition of EELS data. This instrument represents a breakthrough in data acquisition since it allows to measure the full Brillouin zone of surface phonons within few minutes [6].

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Nanoscale charge transport studied by Multi-Tip STM

(Contribution to Topic 1) Applying Multi-Tip Scanning Tunnelling Microscopy (STM) to charge transport measurements through nanostructures is another step in the recent paradigm shift in scanning probe microscopy, transforming from “just imaging” to a tool for detailed measurements at the nanoscale [1]. Having developed a unique family of Multi-Tip STMs based on our patented Koala-Drive [2], we proceed to demonstrate that the Multi-Tip STM method provides excellent capabilities to reveal fundamental nanoscale charge transport properties. The following are a few examples from our recent work:

Mapping resistance profiles and corresponding doping profiles along freestanding GaAs nanowires. Compared to imaging, nanoscale charge transport properties of nanostructures grown by self-organization are much harder to access. Multi-Tip STM enables the use of the four-probe method (Fig. 6) with freely positionable tips in order to record resistance profiles and also doping profiles along freestanding GaAs nanowires. Important information on the dopant incorporation during the nanowire growth is obtained [3]. This research was conducted in cooperation with the group of Professor T. Hannappel at the University of Ilmenau and with Dr. W. Prost from the University Duisburg-Essen.

Disentangling surface conductivity from semi-conductor bulk conductivity. The increasing importance of the surface conductance (compared to that of the bulk) in modern nanoelectronic

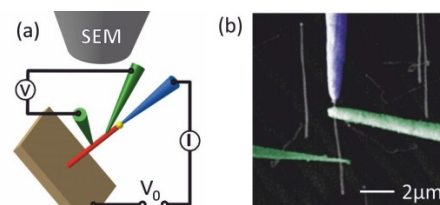


Fig. 6: (a) Schematic of a four-point measurement on a nanowire with three tips contacting a nanowire. (b) SEM image. By repositioning the left tip, a resistance profile along a nanowire is measured with high spatial resolution [2].

devices calls for a reliable determination of the surface conductivity, in order to minimize the influence of undesired leakage currents on the device performance, or in order to use surfaces as functional units. We have shown that distance-dependent electrical four-point measurements using a Multi-Tip STM together with a theoretical model of the charge transport can be used to disentangle surface conductivity from parallel conductance channels through the space charge layer and the bulk [4]. We are currently applying these methods to topological insulators [5]. This research was conducted in cooperation with the group of Professor I. Ořt’adal at the Charles University Prague, as well as Dr. G. Mussler (PGI-9), Dr. G. Bihlmayer (IAS-1), Dr. M. Luysberg (PGI-5), Dr. E. Neumann (HNF), and Dr. H. Soltner (ZEA-1).

Electrical resistance of individual defects at a topological insulator surface. Scanning tunneling potentiometry allows to map the potential landscape while a current flows through a nanostructure and has the unique potential to access the spatial distribution of resistances at the nanoscale. We use potentiometry to analyse the resistance of different kinds of defects at the surface of a $(\text{Bi}_{0.53}\text{Sb}_{0.47})_2\text{Te}_3$ topological insulator thin film [6, 7]. The largest localized voltage drop we find to be located at domain boundaries in the topological insulator film, with a resistivity about four times higher than that of a step edge. As shown in Fig. 7, we resolve resistivity dipoles located around nanoscale voids in the sample surface. The influence of such defects on the resistance of the topological surface state is analysed by means of a resistor network model [6]. Furthermore, we are able to disentangle *in situ* top and bottom conductance of a topological insulator thin film by gate dependent measurements. This research was conducted in cooperation with Dr. G. Mussler (PGI-9) and Dr. L. Plucinski (PGI-6).

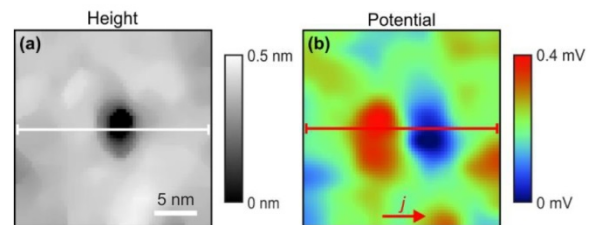


Fig. 7: Resistivity dipoles around nanoscale voids. (a) STM image of a typical void in the sample surface. (b) Corresponding potential map (acquired with scanning tunneling potentiometry) showing a dipole shaped feature centered at the defect.

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Contributions to the Program and the Center

Novel device concepts in nanoelectronics and quantum technologies (e.g., neuromorphic computing, energy-efficient “green” IT, quantum computing) require, first, interface engineering and, second, quantum-scale control, the latter in the sense of advanced diagnostics and atomic-scale manipulation capabilities, which is of strategic importance for the program and the centre.

Following our mission (“exploring and extending the limits of nanoscale engineering”), we provide important elements towards both goals: The metal-organic interface and interfaces between 2D materials belong to the class of interfaces offering the most exciting functionalities for future devices, and our work on single molecule manipulation, visualization of molecular wave functions and Multi-Tip STM are key elements of quantum-scale control.

The close integration of our research into the program and centre is illustrated by STM studies and Multi-Tip charge transport measurements on nanostructures that are fabricated by Dr. G. Mussler in PGI-9. In this unique “complete in situ” approach we transfer samples in vacuum from the growth chamber in PGI-9 to our four-tip STM for electrical measurements, in order to avoid any contamination. In this way, we study the charge transport properties of topological insulators together with our colleagues Dr. G. Mussler (PGI-9), Dr. L. Plucinski (PGI-6), Dr. G. Bihlmayer (IAS-1), Dr. M. Luysberg (PGI-5), Dr. E. Neumann (HNF), and Dr. H. Soltner (ZEA-1). This work contributes to Topics 1 and 2 of the program.

As a service to the program and the centre, PGI-3 organizes the annual JARA-FIT Lab Course Nanoelectronics² with about 30 - 50 students participating in a one-week course.

Future Goals and Directions

JARA Institute for Quantum-Scale Control. In order to further strengthen our capabilities in quantum-scale control, we will intensify our collaboration with the group of Professor M. Morgenstern at the RWTH Aachen. Specifically, we aim to establish a joint JARA Institute for Quantum-Scale Control. Taken together, PGI-3 and the Morgenstern group offer a unique portfolio of relevant expertise: time-resolved STM with world-record time resolution of 120 ps (RWTH), novel SPM imaging methods enabled by a molecular architecture of the probing tip (PGI-3), the combination of STM and multi-tip transport experiments (PGI-3), a world-wide unique Six-Probe SEM/STM (PGI-3), tip-on the chip technology for a direct processing of the atomically resolved data in μm proximity to the tip by innovative circuitry, e.g., for single electron counting (RWTH), and a dedicated molecular manipulation laboratory (www.momalab.org, PGI-3). As a first initiative we combine the STM time-resolution available at the Morgenstern group at RWTH with our Multi-Tip STM in order to enable time-resolved experiments with multiple-probes.

We also succeeded in attracting Dr. Markus Ternes, currently at the MPI for Solid State Research in Stuttgart, an internationally recognized expert in the field of single molecule spin physics. He will join the JARA Institute for Quantum-Scale Control, in the frame of a Heisenberg Professorship. He will install and run a Scanning Probe Picoscope for Qubit-Engineering in PGI-3.

Molecular 3D printing. Our experience in precise SPM-based molecular manipulation is the starting point for a much greater challenge: The additive manufacturing of molecular-scale mechanical and electronic devices. If successful, this may allow rapid prototyping at the molecular level and thus provide a new and very efficient way to test novel device concepts in quantum technologies. To achieve this goal of “3D printing” of molecular structures, we will combine computer-controlled manipulation experiments (data collection), sophisticated machine learning (ML) techniques (data structuring and analysis), and simulations (data interpretation), enabling the inference of molecular conformations in real time during manipulation. As a complementary strategy, we will use reinforcement learning to solve manipulation tasks with limited information about molecular conformations. This research is supported by a ERC

² www.fz-juelich.de/pgi/nanoelektronikpraktikum

Starting Grant *Controlled Manipulation of Molecules (CM³)* to Dr. C. Wagner of PGI-3. Regarding ML, we have established a collaboration with Professor Klaus-Robert Müller at TU Berlin.

Sonderforschungsbereich 1083 “Structure and Dynamics of Internal Interfaces” at the University of Marburg. Despite their enormous importance, the microscopic understanding of internal interfaces is lagging behind that of volume or surface properties. The main reason for this knowledge gap is the experimental difficulty to detect and isolate the weak interface signature from the signals of the dominant bulk. The objective of the collaborative research centre SFB 1083 at the University of Marburg, coordinated by Professor U. Höfer, is to close this gap. On the strength of our pioneering work in the normal incidence x-ray standing wave technique (NIXSW), carried out in collaboration with many groups world-wide (Professor M. Sokolowski at University of Bonn; Professor B. Stadtmüller at University of Kaiserslautern; Professor M. Gottfried at University of Marburg; Professor D. P. Woodruff at Warwick University; Professor P. Tegeder at University of Heidelberg, Professor S. Kera at the National Institute for Molecular Science, Japan), we have been invited to join the SFB as an external member. Our project has received excellent reviews. In future, our activities in 2D materials will be closely coordinated with the SFB. In turn, the program and the center will benefit from the world-leading research program on internal interfaces of the SFB at the University of Marburg.

Method development and technology transfer. The HGF has been repeatedly urged to intensify its technology transfer. PGI-3 has a longstanding tradition of developing of unique research instruments and is therefore in a good position to respond to this request. Examples from the past are the magnetic turbo pump, the Besocke STM, and the Ibach HREELS spectrometer. Currently, we are working on Multi-Tip STM, also at low temperatures (4K) and in high magnetic fields (8T), a parallel-detection HREELS spectrometer, and a mK-STM that is cooled by adiabatic demagnetization.

Regarding the commercialization of parallel-detection HREELS, we are currently in negotiations with a manufacturer of surface analytics, while our Multi-Tip STM is commercially available from the spin-off company mProbes (www.mprobes.com), founded by members of PGI-3. The next step is the development of a combined six-tip SEM/AFM nanoprobe (Hexaprobe) that can be used in the semiconductor industry for semiconductor failure analysis. Within a Helmholtz Validation Fund project, the advantages of our nanoprobe for the use in semiconductor failure analysis will be validated together with the semiconductor company Global Foundries (Dr. W. Horstmann). In order to bring together the Multi-Tip STM community, we have organized the first international workshop on “Charge Transport at Surfaces and Nanostructures with Multi-Probe Techniques” in Jülich.

2.4.4 PGI - Scattering Methods (PGI-4 / JCNS-2)

Head of Institute: Prof. Thomas Brückel

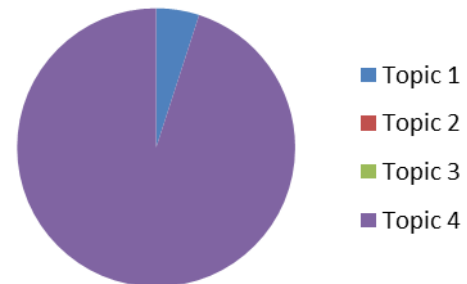
Contributing Personnel (31 Dec 2016 in FTE):

Core-funded: 3 scientists, 1 doctoral students, 0 scientific support personnel

Third-party funded: 0 scientists, 0 doctoral students, 0 scientific support personnel

Contributing Principal Investigators: Manuel Angst, Karen Friese, Oleg Petravic, Sabine Pütter, Yixi Su, Yinguo Xiao.

We focus our research on nanomagnetism and correlated electron systems. Employing state-of-the-art neutron- and synchrotron radiation- techniques, we provide crucial microscopic information on structure, magnetic correlations and elementary excitations to the FIT program. This research is conducted in the joint institutes of PGI-4 and JCNS-2 linking disciplinary research with method development. JCNS-2 is involved in the development and operation of neutron scattering instruments at some of the best sources worldwide. It also operates the JCNS-Infrastructure as an user facility.



PGI-4/JCNS-2 mainly contributes to Topic 4.

This research is embedded in the research field Matter, Program “Matter, Materials and Life”, where the institute JCNS-2 / PGI-4 will be evaluated. For information, we present here a compilation of highlights relevant for topic 4 of the FIT program.

Unconventional superconductivity in Fe based materials is of high current interest. EuFe_2As_2 is a unique representative of the 122 type family of iron based superconductors due to the two magnetic sublattices, the strong coupling between spin-, lattice- and charge degrees of freedom, and the tuneability to superconductivity by chemical doping or external pressure. We have determined the magnetic ground states in superconducting $\text{EuFe}_2(\text{As},\text{P})_2$ [1, 2], and $\text{Eu}(\text{Fe},\text{Ir})_2\text{As}_2$ [3] by single-crystal neutron diffraction and x-ray resonant magnetic scattering (XRMS). In these compounds, strong ferromagnetism from Eu can coexist with bulk superconductivity, a surprising finding for these two antagonistic phenomena. Evidence for a possible interplay between the two magnetic sublattices is obtained by XRMS measurements on the underdoped, non-superconducting $\text{Eu}(\text{Fe}_{0.94}\text{Ir}_{0.06})_2\text{As}_2$ [4].

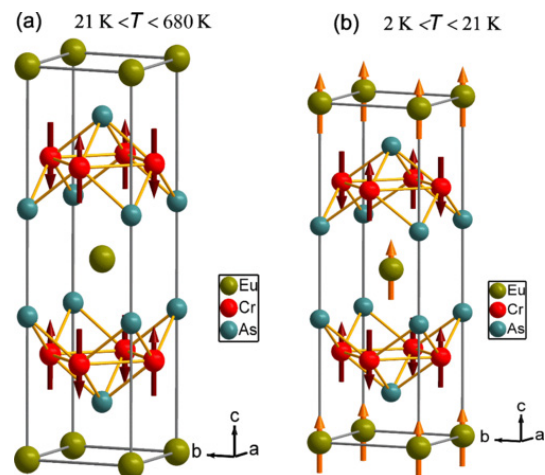


Fig. 1: The ground-state magnetic structure of EuCr_2As_2 for (a) $21\text{ K} \leq T \leq 680\text{ K}$, and (b) $2\text{ K} \leq T \leq 21\text{ K}$ as determined by single-crystal neutron diffraction [5].

- [1] S. Nandi, W. T. Jin, Y. Xiao, Y. Su, S. Price, D. K. Shukla, J. Strempfer, H. S. Jeevan, P. Gegenwart, and Th. Brückel, Coexistence of superconductivity and ferromagnetism in P-doped EuFe_2As_2 , Phys. Rev. B **89**, 014512 (2014) doi: [10.1103/PhysRevB.89.014512](https://doi.org/10.1103/PhysRevB.89.014512)
- [2] S. Nandi, W. T. Jin, Y. Xiao, Y. Su, S. Price, W. Schmidt, K. Schmalzl, T. Chatterji, H. S. Jeevan, P. Gegenwart, and Th. Brückel, Magnetic structure of the Eu^{2+} moments in superconducting $\text{EuFe}_2(\text{As}_{1-x}\text{P}_x)_2$ with $x=0.19$, Phys. Rev. B **90**, 094407 (2014) doi: [10.1103/PhysRevB.90.094407](https://doi.org/10.1103/PhysRevB.90.094407)

- [3] **W. T. Jin**, Wei Li, **Y. Su**, **S. Nandi**, **Y. Xiao**, W. H. Jiao, M. Meven, A. P. Sazonov, **E. Feng**, Yan Chen, C. S. Ting, G. H. Cao, and **Th. Brückel**, *Magnetic ground state of superconducting $\text{Eu}(\text{Fe}_{0.88}\text{Ir}_{0.12})_2\text{As}_2$: A combined neutron diffraction and first-principles calculation study*, Phys. Rev. B **91**, 064506 (2015)
[doi: 10.1103/PhysRevB.91.064506](https://doi.org/10.1103/PhysRevB.91.064506)
- [4] **W. T. Jin**, **Y. Xiao**, **Y. Su**, **S. Nandi**, W. H. Jiao, G. Nisbet, **S. Demirdis**, G. H. Cao, **Th. Brückel**, *Magnetic polarization of Ir in underdoped nonsuperconducting $\text{Eu}(\text{Fe}_{0.94}\text{Ir}_{0.06})_2\text{As}_2$* , Phys. Rev. B **93**, 024517 (2016)
[doi: 10.1103/PhysRevB.93.024517](https://doi.org/10.1103/PhysRevB.93.024517)
- [5] **S. Nandi**, **Y. Xiao**, N. Qureshi, U. B. Paramanik, **W. T. Jin**, **Y. Su**, B. Ouladdiaf, Z. Hossain, and **Th. Brückel**, *Magnetic structures of the Eu and Cr moments in EuCr_2As_2 : Neutron diffraction study*, Phys. Rev. B **94**, 094411 (2016) [doi: 10.1103/PhysRevB.94.094411](https://doi.org/10.1103/PhysRevB.94.094411)

Topological quantum materials have become a focus of intense research owing to the remarkable possibilities to realize emergent exotic quasi particles. Realization and eventual manipulation of those quasi particles can lead to potential applications for future information technology such as decoherence-free quantum computing. Frustrated magnets have been among particularly productive playgrounds in this regard. In frustrated A-site spinels, an exotic spiral spin-liquid state, where spins fluctuate collectively as spirals with propagation vectors forming a continuous surface in reciprocal space, may emerge as the consequence of the competing nearest neighbor and next-nearest neighbor exchange interactions. Predominant short-range spin correlations reminiscent of a spin liquid state have been revealed in high-quality FeAl_2O_4 [6]. Very recently, direct evidence for the spiral spin-liquid state has been obtained in single-crystal MnSc_2S_4 based on the observation of the continuous spiral surface via polarized diffuse neutron scattering [7]. Extensive studies have also been carried out on rare-earth pyrochlore oxides with the focus on the potential experimental realization of quantum spin ice. An incipient ferromagnetic correlation in $\text{Tb}_2\text{Ge}_2\text{O}_7$ with dominant antiferromagnetic exchange has been identified with polarized neutron scattering [8].

- [6] **H. S. Nair**, **Z. Fu**, **J. Voigt**, **Y. Su** and **Th. Brückel**, *Approaching the true ground state of frustrated A-site spinels: A combined magnetization and polarized neutron scattering study*, Phys. Rev. B **89**, 174431 (2014)
[doi: 10.1103/PhysRevB.89.174431](https://doi.org/10.1103/PhysRevB.89.174431)
- [7] S. Gao, O. Zaharko, V. Tsurkan, **Y. Su**, J.S. White, G.S. Tucker, B. Roessli, F. Bourdarot, R. Sibille, D. Chernyshov, T. Fennell, A. Loidl and C. Rüegg, *Spiral spin-liquid and the emergence of a vortex-like state in MnSc_2S_4* , Nat. Phys. **13**, 157 (2017) [doi: 10.1038/nphys3914](https://doi.org/10.1038/nphys3914)
- [8] A.M. Hallas, J.G. Cheng, A.M. Arevalo-Lopez, H.J. Silverstein, **Y. Su**, P.M. Sarte, H.D. Zhou, E.S. Choi, J.P. Attfield, G.M. Luke and C.R. Wiebe, *Incipient ferromagnetism in $\text{Tb}_2\text{Ge}_2\text{O}_7$: Application of chemical pressure to the enigmatic spin-liquid compound $\text{Tb}_2\text{Ti}_2\text{O}_7$* , Phys. Rev. Lett. **113**, 267205 (2014)
[doi: 10.1103/PhysRevLett.113.267205](https://doi.org/10.1103/PhysRevLett.113.267205)

Functional magnetic nanomaterials constitute a novel type of condensed matter with properties determined by the individual behavior of its nanoparticle building blocks, their mutual interactions and the embedding matrix material. For the collective magnetism to be understood and employed, highly ordered arrangements of nanoparticles are essential, achieved by assisted self-assembly. We have followed the evaporation-induced assembly process of superparamagnetic, $\gamma\text{-Fe}_2\text{O}_3$ nanospheres dispersed in toluene in real time using Grazing Incidence Small-Angle X-ray Scattering (GISAXS) [9]. Another route to the formation of a well-ordered supercrystal of particles is magnetic field-induced self-assembly, as we demonstrated by means of Small-Angle Neutron Scattering (SANS) [10].

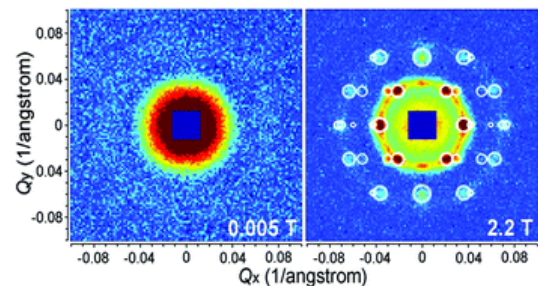


Fig. 2: Appearance of Bragg reflections, when in nanoparticle dispersion a face centred cubic superlattice of magnetic nanoparticles is formed in moderate magnetic field [10].

We have also investigated the magnetic properties of the nanoparticles themselves in detail, using neutron scattering. On MnO nanoparticles we were able to demonstrate that an apparent discrepancy between magnetometry and neutron scattering is due to the different time scales of these techniques [11]. Studying the magnetic excitations in cubic maghemite nanoparticles, we obtained a consistent view of the energy scales of superparamagnetic relaxation and collective magnetic excitations [12].

- [9] E. Josten, E. Wetterskog, A. Glavic, P. Boesecke, A. Feoktystov, E. Brauweiler-Reuters, U. Rücker, G. Salazar-Alvarez, Th. Brückel, L. Bergström, *Superlattice growth and rearrangement during evaporation-induced nanoparticle self-assembly*, Scientific Reports **7**, 2802 (2017) [doi: 10.1038/s41598-017-02121-4](https://doi.org/10.1038/s41598-017-02121-4)
- [10] Z. Fu, Y. Xiao, A. Feoktystov, V. Pipich, M.-S. Appavou, Y. Su, E. Feng, W. Jin, Th. Brückel, *Field-induced self-assembly of iron oxide nanoparticles investigated using small-angle neutron scattering*, Nanoscale **8**, 18541 (2016) [doi: 10.1039/c6nr06275j](https://doi.org/10.1039/c6nr06275j)
- [11] X. Sun, A. Klapper, Y. Su, K. Nemkovski, A. Wildes, H. Bauer, O. Köhler, A. Schilman, W. Tremel, O. Petravic, Th. Brückel, *Magnetism of monomer MnO and heterodimer FePt@MnO nanoparticles*, Phys. Rev. B **95**, 134427 (2017) [doi: 10.1103/PhysRevB.95.134427](https://doi.org/10.1103/PhysRevB.95.134427)
- [12] S. Disch, A.A. Podlesnyak, K. An, T. Hyeon, G. Salazar-Alvarez, L. Bergström, T. Brückel, R. Hermann, E. Wetterskog, *Spin excitations in cubic maghemite nanoparticles studied by time-of-flight neutron spectroscopy*, Phys. Rev. B **89**, 064402 (2014) [doi: 10.1103/PhysRevB.89.064402](https://doi.org/10.1103/PhysRevB.89.064402)

Thin-film heterostructures allow for new hybrid materials and interface effects unexpected from the bulk materials involved. We fabricate heterostructures with state-of-the-art oxide Molecular Beam Epitaxy (MBE) systems directly linked to our high flux neutron reflectometer MARIA by a versatile UHV transport and measurement chamber [13]. Mastering optimum interface preparation, we could demonstrate that the interface quality in Pt/YIG(111) has significant impact on the spin Hall magnetoresistance [14].

- [13] A. Syed Mohd, S. Pütter, S. Mattauch, A. Koutsoumpas, H. Schneider, A. Weber, T. Brückel, *A versatile UHV transport and measurement chamber for neutron reflectometry under UHV conditions*, Review of Scientific Instruments **87**, 123909 (2016) [doi: 10.1063/1.4972993](https://doi.org/10.1063/1.4972993)
- [14] S. Pütter, S. Geprägs, R. Schlitz, M. Althammer, A. Erb, R. Gross, S. T. B. Goennenwein, *Impact of the interface quality of Pt/YIG(111) hybrids on their spin Hall magnetoresistance*, Appl. Phys. Lett. **110**, 012403 (2017) [doi: 10.1063/1.4973460](https://doi.org/10.1063/1.4973460)

Magnetoelectric (ME) multiferroics have a significant applications potential, provided the ME coupling is strong. One focus of our research has been on multiferroics with ferro-electricity originating from charge ordering (CO): because the same ions, those with partially filled d-shells, are involved in both CO and spin ordering, the ME coupling may be significant. We continued our exploration of charge and magnetic order in rare earth ferrites $R\text{Fe}_2\text{O}_4$, for which the mechanism had first been proposed (for $R=\text{Lu}$). Although our previous investigations had shown a lack of ferroelectricity for $R=\text{Lu}$, tuning e.g., by varying the R ion size, could modify the orders. Growing stoichiometric single crystals with $R=\text{Y}$ and $R=\text{Yb}$, we found indeed a completely different, though also not ferroelectric, CO [15].

Type-II multiferroics, where ferroelectricity directly results from magnetic ordering, have an intrinsically large ME coupling. For most known type-II multiferroics, ferroelectricity is driven by a combination of the “inverse Dzyaloshinskii-Moriya interaction” and the “spin current mechanism” (IDM/SC). One example is MnWO_4 , for which we could improve ferroelectricity by tuning the geometrical frustration with Cu doping [16]. In this compound, we could furthermore identify,

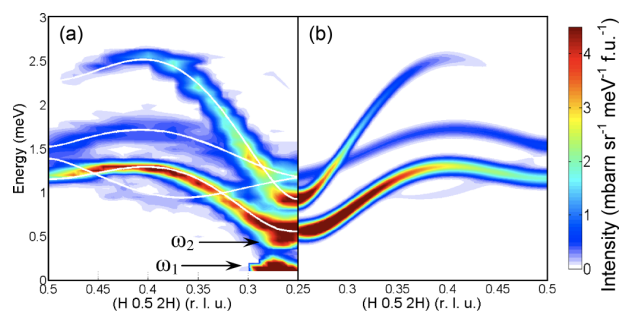


Fig. 3: Observed (left) and calculated spin wave dispersion relations of MnWO_4 at 1.5K. The excitations marked by arrows are identified as electromagnons [17].

by inelastic neutron scattering, low-energy dispersive “electromagnons”, which reflect the dynamic ME coupling and could be observed even in the paraelectric phase [17]. Detailed analysis of the dispersion suggests that IDM/SC may not be the sole source of ferroelectricity.

- [15] **T. Mueller, J. de Groot, J. Stremper, M. Angst**, *Stoichiometric $\text{YFe}_2\text{O}_{4-\delta}$ single crystals, grown by the optical floating-zone method*, J. Crystal Growth **428**, 40 (2015) [doi: 10.1016/j.jcrysgro.2015.07.015](https://doi.org/10.1016/j.jcrysgro.2015.07.015)
- [16] **C. M. N. Kumar, Y. Xiao, P. Lunkenheimer, A. Loidl, M. Ohl**, *Crystal structure, incommensurate magnetic order, and ferroelectricity in $\text{Mn}_{1-x}\text{Cu}_x\text{WO}_4$ ($0 \leq x \leq 0.19$)*, Phys. Rev. B **91**, 235149 (2015) [doi: 10.1103/PhysRevB.91.235149](https://doi.org/10.1103/PhysRevB.91.235149)
- [17] **Y. Xiao, C. M. N. Kumar, S. Nandi, Y. Su, W. T. Jin, Z. Fu, E. Faulhaber, A. Schneidewind, and Th. Brückel**, *Spin-wave and electromagnon dispersions in multiferroic MnWO_4 as observed by neutron spectroscopy: Isotropic Heisenberg exchange versus anisotropic Dzyaloshinskii-Moriya interaction*, Phys. Rev. B **93**, 214428 (2016) [doi: 10.1103/PhysRevB.93.214428](https://doi.org/10.1103/PhysRevB.93.214428)

Materials exhibiting a sizable magnetocaloric effect (MCE) form the basis of novel energy-efficient and environmentally-friendly refrigeration technologies based on the magnetocaloric cooling cycle. Our research focuses on compounds of the series $\text{Mn}_{5-x}\text{Fe}_x\text{Si}_3$ which are composed of abundant and ecologically friendly elements and exhibit a sizeable MCE at magnetic field changes below 2 T. Of special interest for us is the compound MnFe_4Si_3 [18,19], as it shows a ferromagnetic transition close to room temperature. X-ray and neutron diffraction studies have shown that Fe is incorporated into the sites which are octahedrally coordinated by silicon. A second site for the paramagnetic atoms exhibits mixed occupancy by Mn and Fe. Only for this site a significant magnetic moment exists, with the magnetic moments aligned in the *a,b*-plane of the hexagonal unit cell [18]. The magnetocaloric effect is anisotropic, being significantly higher when the magnetic field is applied in the *ab*-plane. Inelastic neutron scattering (INS) demonstrates interference between magnetism and phonons at low energies, which can pave the way for first microscopic explanation of the MCE. We also investigated the parent compound Mn_5Si_3 with INS, identifying magnetic fluctuations above the magnetocaloric transitions that may play an essential role for the MCE.

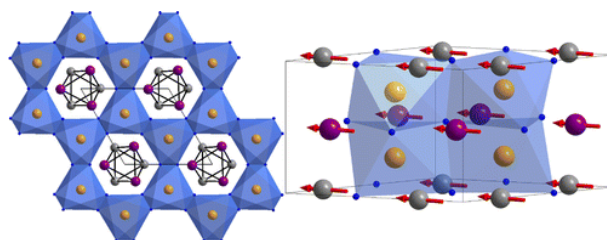


Fig. 4: crystal and magnetic structure of MnFe_4Si_3 [18].

- [18] **P. Hering, K. Friese, J. Voigt, J. Persson, N. Aliouane, A. Grzechnik, A. Senyshin, T. Brückel**, *Structure, Magnetism, and the Magnetocaloric Effect of MnFe_4Si_3 Single Crystals and Powder Samples*, Chem. Materials **27**, 7128 (2015) [doi: 10.1021/acs.chemmater.5b03123](https://doi.org/10.1021/acs.chemmater.5b03123)
- [19] **M. Herlitschke, B. Klobes, I. Sergueev, P. Hering, J. Persson, R. P. Hermann**, *Elasticity and magnetocaloric effect in MnFe_4Si_3* , Phys. Rev. B **93**, 094304 (2016) [doi: 10.1103/PhysRevB.93.094304](https://doi.org/10.1103/PhysRevB.93.094304)

2.4.5 PGI - Microstructure Research (PGI-5)

Head of Institute: Prof. Rafal E. Dunin-Borkowski

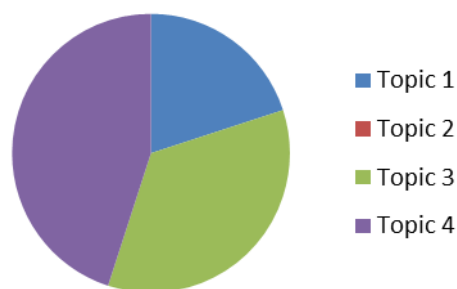
Contributing Personnel (31 Dec 2016 in FTE, excluding guests from other institutes and visitors):

Core-funded: 15 scientists, 3 doctoral students, 8 scientific support personnel

Third-party-funded: 11 scientists, 5 doctoral students, 0 scientific support personnel

Contributing Principal Investigators: Juri Barthel, Philipp Ebert, Michael Faley, Michael Feuerbacher, Marc Heggen, Chunlin Jia, Lei Jin, András Kovács, Martina Luysberg, Vadim Migunov, Michael Schnedler, Andreas Thust

The Institute for Microstructure Research (PGI-5) works on topical fields in solid state physics. Strategically, two directions are followed: first, to make key contributions to the development and application of ultra-high-resolution transmission electron microscopy (TEM), in particular to aberration-corrected electron optics for sub-Ångström structural and spectroscopic resolution and *in situ* TEM; second, to produce selected material systems, including high temperature superconductors and novel complex metallic and high entropy alloys, and to study their physical properties using TEM and scanning tunnelling microscopy and spectroscopy. The superconductors provide the basis of the institute's work on SQUID sensors and ac-Josephson effect based Hilbert spectroscopy.



PGI-5 mainly contributes to Topics 3 and 4.

The head of the institute is co-Director of the Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons (ER-C) - a user and development facility for atomic resolution microscopy and spectroscopy with electrons at the highest international level. It is operated as a joint establishment of Forschungszentrum Jülich and RWTH Aachen University (where the co-Director is Prof. Joachim Mayer). The ER-C is the first national user centre for ultra-high-resolution electron microscopy (see Part I, 4.3). It offers researchers from science and industry access to the most powerful electron microscopes that are currently available. The latest addition is the PICO microscope - a chromatically and spherically aberration-corrected TEM with a spatial information limit of 50 pm and an energy resolution of 100 meV. Selected examples of recent research are described below.

Nanoelectronic oxides

Recent research on nanoelectronic oxides includes a study of the influence of bulk polarization and surface polarity on surface reconstructions and local properties in multiferroic BiFeO₃ films. Perovskite oxides undergo surface relaxation and reconstruction as a result of the breaking of translational symmetry. Coupling between lattice and order parameters, such as polarization (\mathbf{P}_s) in ferroelectrics and polarity in polar structures, has a strong impact on surface relaxation and reconstruction. The resulting surface structures can affect samples with large surface to volume ratios and devices that rely on interfacial coupling. Fig. 1 shows results taken from an atomic-resolution study of multiferroic BiFeO₃ (BFO) thin films on DyScO₃ (110)_o (where o denotes orthorhombic) combined with density functional theory (DFT) calculations. (BiO)⁺ and (FeO₂)⁻ layers stack alternately along the *c*-axis, leading to the formation of a polar {001} surface, while off-centre displacements of Fe and O atoms with respect to the Bi sub-lattice

result in a large \mathbf{P}_s of approximately $0.9 - 1.0 \text{ C/m}^2$ along the $[111]$ body diagonal. \mathbf{P}_s contains an $\langle 001 \rangle$ component that interacts with charges on the surface. A 180° domain wall can be traced by mapping displacement vectors pointing from the centres of four Bi columns to FeO columns. Two surface structures are evident, depending on the \mathbf{P}_s of the underlying ferroelectric domain. On domains that have an upward polarization component (Type I), a layer with an

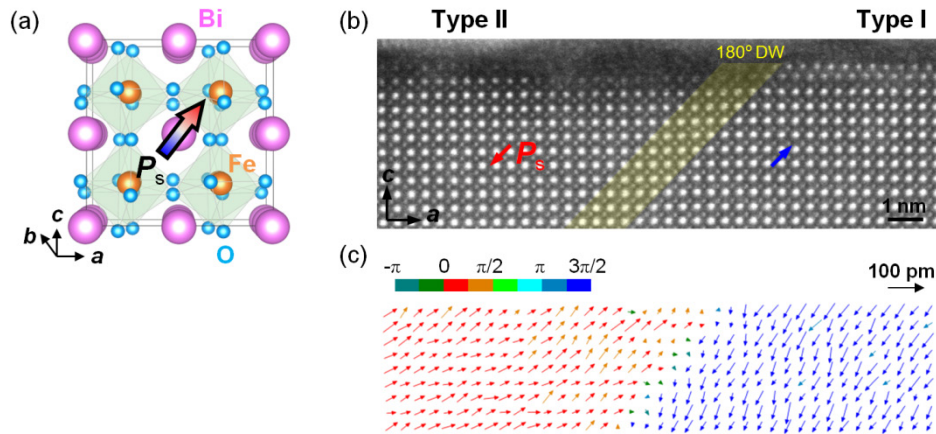


Fig. 1: (a) $2 \times 2 \times 2$ pseudocubic unit cells of room temperature BFO, showing displacements of Fe and O with respect to the Bi sub-lattice. The displacements lead to a spontaneous polarization \mathbf{P}_s pointing towards the $[111]$ body diagonal. The $(\text{BiO})^+$ and $(\text{FeO}_2)^-$ atomic planes have positive and negative charges and stack alternately along the $[001]$ axis. Four configurations of the (001) surface are expected, depending on the \mathbf{P}_s direction and the termination of the atomic planes. (b) Atomic-resolution HAADF STEM image showing two BFO ferroelectric domains separated by a 180° domain wall. The surface structure on the left domain can be distinguished from that on the right domain. (c) Displacement map between the centre of four Bi columns and FeO columns, showing a reversal in \mathbf{P}_s .

Aurivillius-Bi₂O₂-like structural unit is observed. Dramatic changes in local properties are measured below the surface layer. On domains that have a downward polarization component (Type II), no reconstructions are visible. DFT calculations show that for the type I surface the structural unit of the Aurivillius phase acts as a reservoir for negative charges from an excess of O in the outer Bi plane, which stabilizes the surface structure. In contrast, the negative surface bound charges induced by the downward component of \mathbf{P}_s on the type II surface can be self-compensated by the positive charges of the $(\text{BiO})^+$ termination layer and do not require an additional Bi₂O₂-like structure. Our results promise to stimulate interest in research on surface reconstructions in other ferroelectric oxides with odd valences of cations, such as LiNbO₃ [1].

- [1] L. Jin, P.X. Xu, Y. Zeng, L. Lu, J. Barthel, T. Schulthess, R.E. Dunin-Borkowski, H. Wang, C.L. Jia, *Surface reconstructions and related local properties of a BiFeO₃ thin film*, Sci. Rep. **7**, 39698 (2017)
[doi: 10.1038/srep39698](https://doi.org/10.1038/srep39698)

Magnetic imaging of nanostructures

We use Lorentz TEM (LTEM), off-axis electron holography and electron magnetic chiral dichroism, in combination with the development of novel inversion algorithms, to study the local magnetic properties of materials that include chiral magnets, mesocrystals and working spintronic devices. Magnetic skyrmions are nanoscale spin objects that can be stabilized in chiral magnets and bilayer films due to the Dzyaloshinskii-Moriya interaction. Interest in magnetic skyrmions has arisen in part because of the ability to move them using extremely low electrical currents. We have studied the temperature, magnetic field, field direction and angular dependence of the magnetic moments of individual skyrmions in thin films of B20-type chiral FeGe [1]. The amplitude of the magnetic moment and internal skyrmion shape are found to vary with applied magnetic field. Deviations from a circular to a hexagonal skyrmion structure are

observed in the lattice phase, in agreement with micromagnetic simulations performed in collaboration with PGI-1. Geometrically-confined skyrmions in nanostripes of FeGe are able to adopt a wide range of sizes and ellipticities that are absent in thin films and bulk materials and can be created from a helical magnetic state with a distorted edge twist [2]. We have also

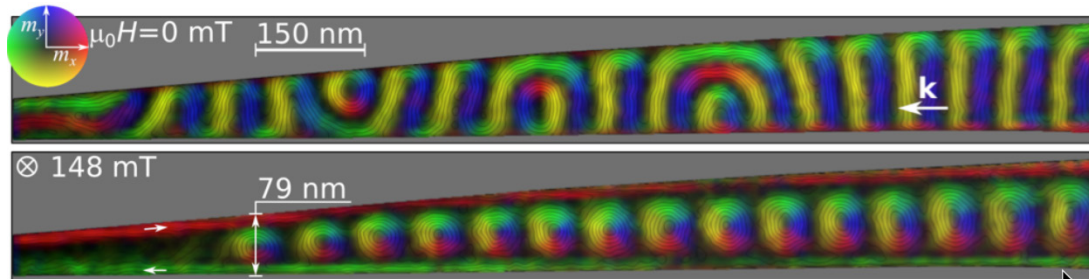


Fig. 2: Magnetic induction maps of helical spin structures and distorted skyrmions in an FeGe nanostripe at 200 K recorded using off-axis electron holography at the indicated out-of-plane magnetic fields [3].

studied skyrmion formation at lattice defects and grain boundaries in FeGe nanowires and

polycrystalline FeGe. We find inversions of crystallographic and magnetic chirality across grains, resulting in the formation of interface spin stripes at grain boundaries. In the absence of material defects, skyrmion lattices possess dislocations and domain boundaries, in analogy to atomic crystals. Distorted skyrmions can flexibly change their size and shape to accommodate local geometry (Fig. 2).

High-resolution magnetic imaging has also been used to characterize a new form of magnetic mesocrystal [3] and to understand the thermomagnetic behavior of pseudo-single-domain magnetite nanoparticles [4, 5]. Our work on electron energy-loss magnetic chiral dichroism [6] includes the development of a new approach that allows in-plane magnetic signals to be measured by selecting a specific diffraction geometry.

- [1] K. Shibata, **A. Kovács**, N. Kanazawa, **R. E. Dunin-Borkowski** and Y. Tokura, *Temperature and magnetic field dependence of the internal and lattice structures of skyrmions by off-axis electron holography*, Phys. Rev. Lett. **118**, 087202 (2017) doi: [10.1103/PhysRevLett.118.087202](https://doi.org/10.1103/PhysRevLett.118.087202)
- [2] C. Jin, Z.-A. Li, **A. Kovács**, **J. Caron**, **F. Zheng**, F. N. Rybakov, N. S. Kiselev, H. Du, S. Blügel, M. Tian, Y. Zhang, M. Farle and **R. E. Dunin-Borkowski**, *Control of morphology and formation of highly geometrically confined magnetic skyrmions*, Nature Communications **8**, 15569 (2017) doi: [10.1038/ncomms15569](https://doi.org/10.1038/ncomms15569)
- [3] V. Reichel, **A. Kovács**, M. Kumari, É. Bereczk-Tompa, E. Schneck, **P. Diehle**, M. Pósfai, A. M. Hirt, **M. Duchamp**, **R. E. Dunin-Borkowski** and D. Faivre, *Single crystalline superstructured stable single domain magnetite nanoparticles*, Scientific Reports **7**, 45484 (2017) doi: [10.1038/srep45484](https://doi.org/10.1038/srep45484)
- [4] T. P. Almeida, A. R. Muxworthy, **A. Kovács**, W. Williams, P. D. Brown and **R. E. Dunin-Borkowski**, *Direct visualization of the thermomagnetic behavior of pseudo-single-domain magnetite particles*, Science Advances **2**, e1501801 (2016) doi: [10.1126/sciadv.1501801](https://doi.org/10.1126/sciadv.1501801)
- [5] D. Song, **A. H. Tavabi**, Z.-A. Li, **A. Kovács**, J. Rusz, W. Huang, G. Richter, **R. E. Dunin-Borkowski** and J. Zhu, *An in-plane magnetic chiral dichroism approach for measurement of intrinsic magnetic signals using transmitted electrons*, Nature Communications **8**, 15367 (2017) doi: [10.1038/ncomms15367](https://doi.org/10.1038/ncomms15367)

Catalyst nanoparticles for energy conversion

The performance of proton exchange membrane (PEM) fuel cells is limited by the oxygen reduction reaction (ORR) at fuel cell cathodes. Because of their catalytically active (111) surfaces, Pt-Ni octahedra are outstanding catalysts for the ORR. We have used aberration-corrected TEM to establish that compositional segregation in shaped Pt alloy nanoparticles (Fig. 3) results in complex corrosion of the nanoparticles during the ORR [1] and to show that rapid growth of Pt-rich hexapods along <100> precedes delayed deposition of a Ni-rich phase on concave {111} sites [2]. This element-specific growth is the main reason for degradation and

loss of activity. Our current research is focused on the stabilization of the microstructures of catalyst nanoparticles using surface doping and additional surface treatments [3]. We have shown that Rh-doped Pt-Ni octahedral nanoparticles have high ORR activities combined with improved performance and shape stability compared to bimetallic Pt-Ni octahedral particles. By using small amounts of Rh, we could suppress the migration rates of Pt atoms and octahedral shape loss. We have shown that Rh-doped Pt-Ni octahedral nanoparticles have high activity and selectivity for ethanol electro oxidation at low overpotentials near the reversible potential of the anodic complete oxidation of ethanol [4]. The effect of different surface treatments under acidic and alkaline conditions was investigated [5]. In acid-chemical leaching, non-noble metals were removed from the surface, with changes in surface composition. Electrochemical measurements showed increased ORR activity compared to untreated nanoparticles and an improvement in activity by 10 compared to commercial Pt. Thermal aging on octahedral Pt-Ni catalyst nanoparticles was carried out *in situ* in the electron microscope. It was shown that thermal annealing at up to 500 °C results in surface diffusion of Pt atoms from the corners to the concave Ni-rich {111} facets, resulting in an atomically smooth facet with a Pt-rich surface [5].

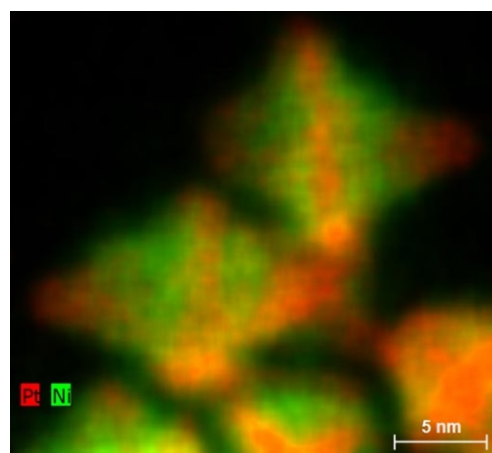


Fig. 3: STEM/EDX of octahedral Pt-Ni nanoparticles showing an inhomogeneous elemental distribution with a Pt-rich frame and segregation of Ni at {111} facets.

- [1] C. Cui, L. Gan, **M. Heggen**, S. Rudi, P. Strasser, *Compositional segregation in shaped Pt alloy nanoparticles and their structural behaviour during electrocatalysis*, Nature Materials **12**, 765–771 (2013) [doi: 10.1038/nmat3668](https://doi.org/10.1038/nmat3668)
- [2] L. Gan, C. Cui, **M. Heggen**, F. Dionigi, S. Rudi, P. Strasser, *Element-specific anisotropic growth of shaped platinum alloy nanocrystals*, Science **346**, 1502–1506 (2014) [doi: 10.1126/science.1261212](https://doi.org/10.1126/science.1261212)
- [3] **M. Heggen**, **M. Gocyla**, **R. E. Dunin-Borkowski**, *The growth and degradation of binary and ternary octahedral Pt-Ni-based fuel cell catalyst nanoparticles studied using advanced transmission electron microscopy*, Advances in Physics X **2**, 281–301 (2017) [doi: 10.1080/23746149.2017.1282834](https://doi.org/10.1080/23746149.2017.1282834)
- [4] N. Erini, V. Beermann, **M. Gocyla**, M. Gliech, **M. Heggen**, **R. E. Dunin-Borkowski**, and P. Strasser, *The Effect of Surface Site Ensembles on the Activity and Selectivity of Ethanol Electrooxidation by Octahedral PtNiRh Nanoparticles*, Angewandte Chemie **129**, 6633 (2017) [doi: 10.1002/ange.201702332](https://doi.org/10.1002/ange.201702332)
- [5] R. Arán-Ais, J. Solla-Gullon, **M. Gocyla**, **M. Heggen**, **R. Dunin-Borkowski**, P. Strasser, E. Herrero, J. Feliu, *The effect of interfacial pH on the surface atomic elemental distribution and on the catalytic reactivity of shape-selected bimetallic nanoparticles towards oxygen reduction*, Nano Energy **27**, 390–401 (2016) [doi: 10.1016/j.nanoen.2016.07.024](https://doi.org/10.1016/j.nanoen.2016.07.024)

Influence of dose rate on high-resolution TEM image contrast

The observation that image contrast in high-resolution TEM is lower than expected from simulations has previously been explained in large part by the contrast-damping characteristics of cameras in use [1]. An additional smaller effect, which is mainly relevant in the sub-Ångström resolution regime, is associated with thermal magnetic field noise [2]. By incorporating these two incoherent contrast damping effects in image simulations, a nearly perfect match between simulations and experiment could be achieved on an absolute scale [3, 4]. Recently, an alternative explanation was given for the mismatch problem [5, 6], attributing the contrast mismatch mainly to sample excitations caused by the electron beam [5]. In order to assess this hypothesis, we acquired dose-rate series of high-resolution TEM images and assessed the magnitude of the image contrast f

or Au, MgO and Ge [10]. The images were normalized to the same mean value and the contribution of noise to the image contrast, which is different for each dose-rate setting, was taken into account. Figure 4 shows a dose-rate series of [110]-oriented Ge for 500 to 87000

electrons per \AA^2 . We found no substantial difference in the magnitude of the image contrast. The same observations were made for MgO and Au. Our results indicate that the hypothesis of a dose-rate dependent contrast reduction is not correct. We also established theoretically that neither static displacements of atoms due to knock-on damage nor vibrational displacements of atoms due to phonon excitation affect the observed contrast significantly. An interpretation of the problem as an object-related phenomenon, which requires the dose rate as an ingredient, can therefore be ruled out.

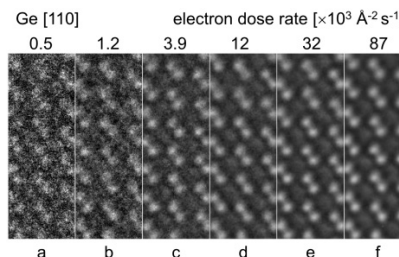


Fig. 4: HRTEM images of Ge [110] for increasing dose rates. The mean intensity of each patch is normalized to 1. The gray scale is from 0.5 (black) to 2.2 (white).

- [1] **A. Thust**, *High-Resolution Transmission Electron Microscopy on an Absolute Contrast Scale*, Phys. Rev. Lett. **102**, 220801 (2009). doi: [10.1103/PhysRevLett.102.220801](https://doi.org/10.1103/PhysRevLett.102.220801)
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- [3] **C.-L. Jia, J. Barthel, F. Gunkel, R. Dittmann, S. Hoffmann-Eifert, L. Houben, M. Lentzen, A. Thust**, *Atomic-Scale Measurement of Structure and Chemistry of a Single-Unit-Cell Layer of LaAlO₃ Embedded in SrTiO₃*, Microsc. Microanal. **19**, 310–318 (2013). doi: [10.1017/S1431927612014407](https://doi.org/10.1017/S1431927612014407)
- [4] **C.L. Jia, S.B. Mi, J. Barthel, D.W. Wang, R.E. Dunin-Borkowski, K.W. Urban, A. Thust**, *Determination of the 3D shape of a nanoscale crystal with atomic resolution from a single image*, Nat. Mater. **13**, 1044–1049 (2014). doi: [10.1038/nmat4087](https://doi.org/10.1038/nmat4087)
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Time-resolved electron holography

We are using electron holography to image dynamic electromagnetic phenomena using double exposure electron holography [1] and direct electron detectors. We have studied the dynamics of electron-beam-induced charging of SiO₂ by acquiring holograms at 400 fps using a Gatan K2 camera [2]. Charge is observed to build up with time and to saturate after ~100 ms of exposure (Fig. 5). Comparisons with theory suggest that the emission of secondary electrons leaves positive charge on the specimen. The generation of carriers by the beam is responsible for the R-C behaviour of the electrical circuit and for a decrease in charge when the beam touches a metal support. Such studies are of interest for understanding the interaction of high-energy electrons with solids. We are also measuring superparamagnetic fluctuations of magnetisation by developing new approaches for denoising and statistical detection in collaboration with T. Furnival and P. Midgley in the University of Cambridge.

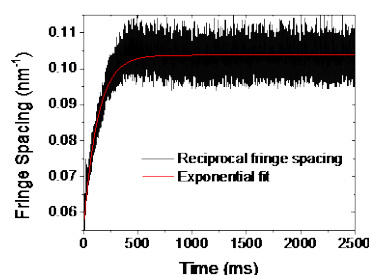


Fig. 5: Dependence of reciprocal fringe spacing (total accumulated charge) on electron irradiation for 20 nm SiO₂ at room temperature.

- [1] V. Migunov, C. Dwyer, C. B. Boothroyd, G. Pozzi, R. E. Dunin-Borkowski, R. E. Prospects for quantitative and time-resolved double and continuous exposure off-axis electron holography. *Ultramicroscopy* **178**, 48–61 (2017) doi: [10.1016/j.ultramic.2016.08.010](https://doi.org/10.1016/j.ultramic.2016.08.010)
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High entropy alloys

High entropy alloys (HEAs) have near-equiatomic compositions of principal elements (typically 5) and constitute a novel class of materials between crystals and metallic glasses. We have developed growth routes for high quality HEAs, using the Bridgman technique to grow single crystals of FeCoCrMnAl with a volume of $\sim 5 \text{ cm}^3$ [1]. This phase consists of B2 inclusions in a body-centred cubic matrix. The inclusions are {001} platelets of about 65 nm thickness with lateral extensions of $\sim 500 \text{ nm}$, predominantly containing Al and Co, while the matrix is rich in Fe and Cr. We also discovered the first hcp HEA, HoDyYGdTb, which forms a single phase with a Mg-type structure. We showed that similar formation conditions can be achieved for 11 further rare earth elements, paving the way for the production of 1486 different HEAs of 5 to 11 components. We also developed production routes for other HEAs, which were investigated in-house with respect to their mechanical properties [3] or in cooperation with external partners, leading to the discovery of the first superconducting HEA.

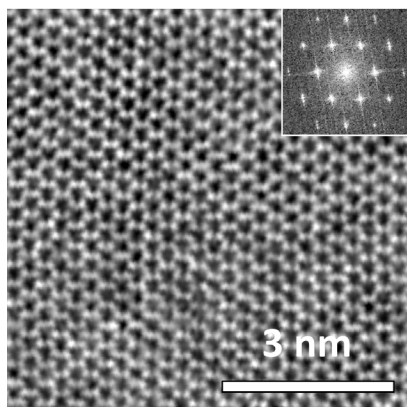


Fig. 6: High-angle annular dark-field image of the equiatomic Ho-Dy-Y-Gd-Tb HEA along [0001]. The inset displays the corresponding FFT.

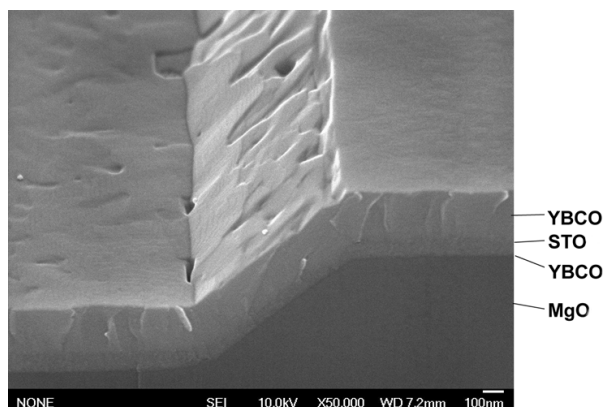


Fig. 7: SEM image of an YBCO film deposited on a double-layer-buffered 45° step on an MgO substrate. The image was obtained with the sample cut perpendicular to the step [9].

- [1] M. Feuerbacher, M. Heidelmann, C. Thomas, *Hexagonal high-entropy alloys*, *Mat. Res. Lett.* **3**, 1 (2015) doi: [10.1080/21663831.2014.951493](https://doi.org/10.1080/21663831.2014.951493)
- [2] M. Feuerbacher, E. Wuertz, A. Kovacs, C. Thomas, *Single-Crystal Growth of a FeCoCrMnAl High-Entropy Alloy*, *Mat. Res. Lett.* **2**, 128 (2017) doi: [10.1080/21663831.2016.1234516](https://doi.org/10.1080/21663831.2016.1234516)
- [3] M. Feuerbacher, *Dislocations and deformation microstructure in an Al28Cu20Cr11Fe15Ni26 high-entropy alloy*, *Scientific Reports* **6**, 29700 (2016) doi: [10.1038/srep29700](https://doi.org/10.1038/srep29700)

High- T_c thin film superconducting devices

The sensitivity of high- T_c superconducting quantum interference devices (SQUIDs) for biomedical applications is crucially dependent on the quality of the epitaxial growth of the thin film metal oxide heterostructures and their nanopatterning. We have developed novel graphoepitaxial high- T_c step edge Josephson junctions for high- T_c DC SQUIDs intended for magnetoencephalography, magnetic micro-scopy and non-destructive evaluation [1-5]. The homogeneity and quality of the superconducting $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (YBCO) films and grain boundaries (GBs) on 45° step edges on MgO substrates was improved significantly by the

introduction of a 10-nm-thick non-superconducting YBCO film as a graphoepitaxial “seed” layer that was covered by a 30-nm-thick epitaxial SrTiO₃ (STO) film as a “blocking” layer. Figure 7 shows a scanning electron microscopy (SEM) image of an YBCO film deposited on a double-layer- buffered 45° step on an MgO substrate. High-resolution TEM reveals two 45° [100]-tilted GBs in the top epitaxial YBCO layer. A normal state resistance $R_n \sim 20$ ohm, capacitance $C \sim 10$ fF and $I_c R_n$ product of 800 μ V at 77 K were measured. The resulting magnetometers achieved a noise equivalent induction of ~ 4 fT/ $\sqrt{\text{Hz}}$ at 77 K and were used for the first source localization of human brain activity using high- T_c sensors [1–4].

- [1] **M. I. Faley**, *Reproducible step-edge Josephson junction*, Patent US20150069331 A1 (published 12.03.2015), Patent DE102012006825 B4 (granted 26.02.2015) and Patent EP 2834860 B1 (granted 30.12.2015).
- [2] J. Dammers, H. Chocholacs, E. Eich, F. Boers, **M. Faley**, **R. E. Dunin-Borkowski** and N. J. Shah, *Source localization of brain activity using helium-free interferometer*, Appl. Phys. Lett. **104**, 213705 (2014), doi: [10.1063/1.4880097](https://doi.org/10.1063/1.4880097)
- [3] **M. I. Faley**, **U. Poppe** and R. L. Fagaly, *Electrical resistance elements and measuring system for measuring time-variable magnetic fields or field gradients*, Patent US9476950B2 granted 25.10.2016.
- [4] **M. I. Faley**, J. Dammers, Y. V. Maslennikov, J. F. Schneiderman, D. Winkler, V. P. Koshelets, N. J. Shah, and **R. E. Dunin-Borkowski**, *High- T_c SQUID biomagnetometers*, Superconductor Science and Technology, (2017), doi: [10.1088/1361-6668/aa73ad](https://doi.org/10.1088/1361-6668/aa73ad)

Scanning tunnelling microscopy of semiconductor surfaces

We have used scanning tunnelling microscopy (STM) to identify lazarevite-type short-range order (SRO) in InAs_{1-x}Sb_x nanowires (NWs) [1]. Fig. 8 shows an atomically-resolved STM image of a sidewall facet on an InAs_{0.90}Sb_{0.10} nanowire (NW), which contains 5–10 nm twinned domains separated by twin boundaries. The brighter atomic positions are a signature of Sb atoms on anion lattice sites. We derived a two-dimensional pair correlation function (PCF) for Sb atoms. The PCF exhibits large values above 1 along the [001] and [00 $\bar{1}$] directions, indicating Sb pairs across the atomic chains. In contrast, nearest- and second-nearest-neighbour Sb pairs within the atomic chains in $\pm[110]$ directions occur rarely. The ternary alloy exhibits SRO, defined by an ordering vector in the [001] direction and an anti-correlation vector in the [110] direction. Density functional theory calculations show an increased Sb-Sb pair interaction energy along the [110] direction. In STM images, the lazarevite ordering shows up as Sb alignment along [001]. During lateral overgrowth on {110} sidewalls, repulsion of Sb-Sb pairs along zigzag chains blocks new Sb atoms from being incorporated. Hence, Sb atoms must sidestep towards neighbouring lattice positions, creating lazarevite-type [001] ordering.

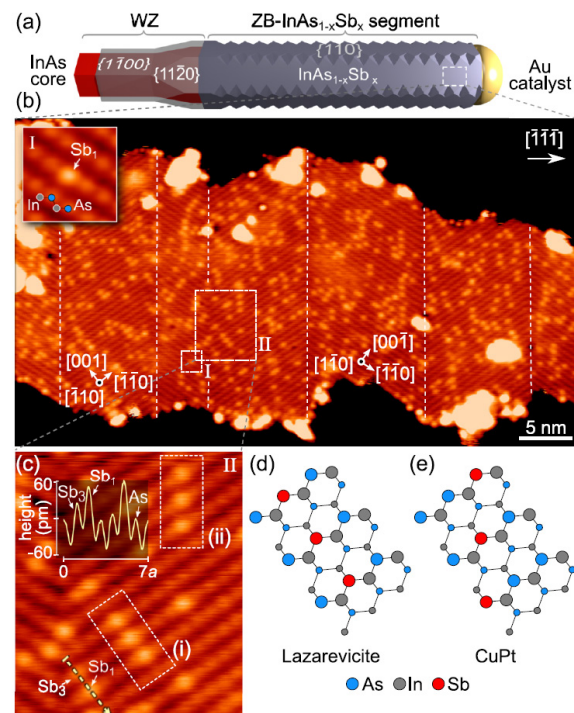


Fig. 8: (a) Schematic of an InAs_{0.9}Sb_{0.1}/InAs NW. (b) Filled state STM image of a sidewall surface at 77 K, with filled dangling bond states above surface anions. Bright atomically localized features arise from Sb atoms on anion sites. Twin boundaries are marked by dashed lines. The inset shows one Sb_{As} atom in the surface layer. (c) High-resolution STM image (~ 2 V, 700 pA) of area II in (b). Sb atoms in the first (Sb₁) and third (Sb₃) layer are visible. Lazarevite- and CuPt- type ordered areas are labeled (i) and (ii). (d) and (e) show corresponding atomic models.

- [1] **M. Schnedler**, I. Lefebvre, T. Xu, **V. Portz**, G. Patriarche, J.-P. Nys, S. R. Plissard, P. Caroff, M. Berthe, H. Eisele, **R. E. Dunin-Borkowski**, **Ph. Ebert**, and B. Grandier, *Lazareviciite-type short-range ordering in ternary III-V nanowires*, Phys. Rev. B **94**, 195306 (2016) [doi: 10.1103/PhysRevB.94.195306](https://doi.org/10.1103/PhysRevB.94.195306)

Planned New Directions

The future roadmap for PGI-5 and the ER-C contains many goals, including:

- Strengthening and expansion of the focus of PGI-5 and the ER-C in the development of new techniques in electron and scanning probe microscopy and their application to challenging problems in condensed matter physics, biology and chemistry, including *in situ* studies of the functional properties of working nanoelectronic and spintronic devices and real-time studies of chemical reactions at the highest spatial resolution, within the framework of a national user facility (compare Part I, 4.3).
- Development of quantitative transmission electron microscopy on an absolute scale through instrument characterization, development and control in collaboration with instrument manufacturers, with a focus on statistical and model-based interpretation of images, diffraction patterns and spectra recorded at both atomic resolution and medium resolution and the development of new evaluation procedures for analyzing, storing and disseminating big data.
- Application of chromatic aberration corrected TEM to the dose-efficient structural and chemical characterization of hard and soft materials with unprecedented spatial resolution.
- Correlative and time-resolved studies to combine high-resolution TEM with scanning probe techniques, atom probe tomography, neutron scattering and synchrotron-based techniques, thereby fostering synergies with a broader range of cooperation partners.

2.4.6 PGI - Electronic Properties (PGI-6)

Head of Institute: Prof. Dr. Claus M. Schneider

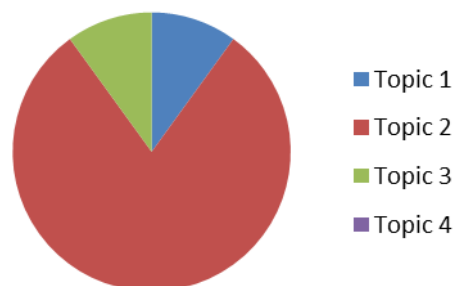
Contributing Personnel (31 Dec 2016 in FTE):

Core-funded: 19 scientists, 12 doctoral students, 7 scientific support personnel

Third-party funded: 5 scientists, 6 doctoral students, 0 scientific support personnel

Contributing Principal Investigators: Roman Adam, Daniel Bürgler, Markus Büscher, Stefan Cramm, Vitaliy Feyer, Paul Kögerler, Natalia Izarova, Frank Matthes, Ewa Mlynczak, Martina Müller, Slavomir Nemsak, Lukasz Plucinski, Carolin Schmitz-Antoniak, Carsten Wiemann

Modern condensed matter physics interfaces the most fundamental phenomena involving the structure of matter and direct applications to technology. Information technology (IT) is the most notable example being continually driven by advances in condensed matter research. Physical properties in condensed matter are intimately connected to details of the electronic structure and the underlying quantum physics. While the research at PGI-6 focuses on fundamental questions in solid state and material science, a particular emphasis lies in the experimental exploration of spin-dependent phenomena and processes, and their long-term translation into energy efficient IT. The scientific program in our “*Spin Institute*” is closely matched by a dedicated development of spectroscopy, microscopy, and scattering techniques, using both synchrotron radiation and ultrashort-pulse laser sources. In-house sample fabrication is based on a variety of thin film deposition techniques, as well as chemical synthesis of molecular structures performed by the Kögerler group.



PGI-6 mainly contributes to Topics 3 and 4.

PGI-6 pursues four closely interwoven research lines: **electronic structure of quantum materials, spintronics and nanomagnetism, ultrafast spin dynamics, and fundamentals of redox processes**. Thus, the PGI-6 activities contribute predominantly to the POF 3 **topic 2 “Controlling spin based phenomena”**, with significantly smaller activities in **topic 3 “Controlling configuration based phenomena”** and **topic 1 “Controlling electron charge-based phenomena”**.

Electronic Structure of Quantum Materials

The exploration of novel quantum effects and quantum materials is the basis for new functionalities in information technology. We have concentrated our efforts on carefully selected topological insulators, 2D materials, magnetic thin films, and molecular structures in order to elucidate the electronic states and disentangle the relevant interactions. The electronic structure studies are a backbone of the scientific work in PGI-6, thereby covering topics 1 to 3.

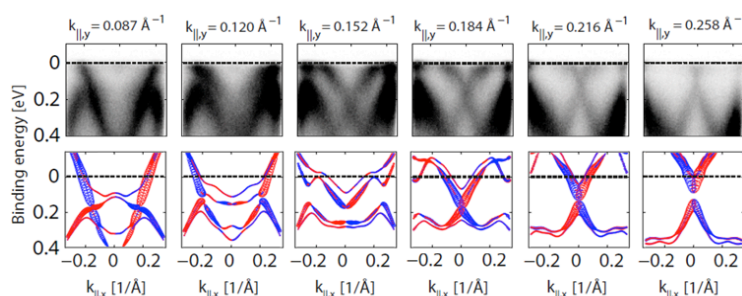


Fig. 1: Surface state crossings away from the Brillouin zone center in Bi_1Te_1 topologically protected by the crystal mirror symmetry. Top row: ARPES spectra measured at PGI-6, bottom row: DFT calculation performed at PGI-1. See Ref. [2] for details.

In case of topological insulators we focused on thin film superstructures produced by molecular beam epitaxy at PGI-9, which is the main collaboration partner in this activity. Recently, we demonstrated band structure engineering in $\text{Sb}_2\text{Te}_3/\text{Bi}_2\text{Te}_3$ bilayer p-n junctions, where the position of the Fermi level, and thus the transport properties, can be controlled by the relative layer thickness [1]. In another line of research we addressed a novel superlattice material, Bi_1Te_1 , composed of periodically stacked atomically thin layers of Bi_2Te_3 and Bi [2]. Bi_1Te_1 is predicted to host two different types of topologically protected surface states on its various surfaces, which we could experimentally verify (Fig. 1).

We have also studied spin textures in the electronic structure of MoS_2 – one of the most important 2D materials – using spin-resolved ARPES [3]. We could demonstrate that even though MoS_2 is centrosymmetric, the electronic structure within each layer is spin-polarized, and the resulting spin texture, which exhibits spin-momentum locking, can be accessed in photoemission experiments thanks to its surface sensitivity and momentum resolution. Fig. 2 shows the ARPES band maps from a MoS_2 cleaved single crystal together with corresponding density functional theory calculations.

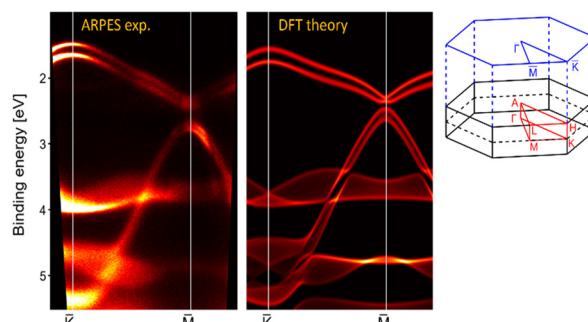


Fig. 2: Experimental band structure of MoS_2 measured at PGI-6 compared to the theoretical calculation performed at PGI-1. See Ref. [3] for details

The high energy and momentum resolution available in photoemission allowed us to map spin-orbit gaps in well-ordered Fe films on $\text{Au}(001)$ [4]. Spin-orbit interaction in transition metal ferromagnets is often considered small and negligible, but according to theoretical predictions, which are confirmed by our experiments, the magnitude of these gaps can be of the order 150 meV or more. These spin-orbit gaps are microscopic signatures of the magnetocrystalline anisotropy. They often mark “hot-spots” in the Brillouin zone, which exhibit non-trivial topologies and play a key role in modern theories of the anomalous Hall effect and ultrafast demagnetization.

Molecular orbital tomography (MOT) combines angle-resolved photoemission spectroscopy (k-PEEM) and DFT calculations within the plane wave approximation. This method allows direct imaging of the orbital structure in k-space and determines the occupation of the frontier molecular orbitals at organic-metal interfaces, which are at the heart of molecular spintronics. Using MOT, we found clear evidence of a pronounced charge transfer involving iron phthalocyanine (FePc) molecules adsorbed on low-index crystal faces of silver [5]. A comparative analysis of the k-space molecular patterns on substrates with different symmetry indicates that both of the twofold degenerate FePc’s lowest unoccupied molecular orbitals (LUMO) are filled by charge transfer from the substrate at the organic-metal interface. By combining MOT and LEED, we could reconstruct the real space adsorption geometries of FePc (Fig. 3). At

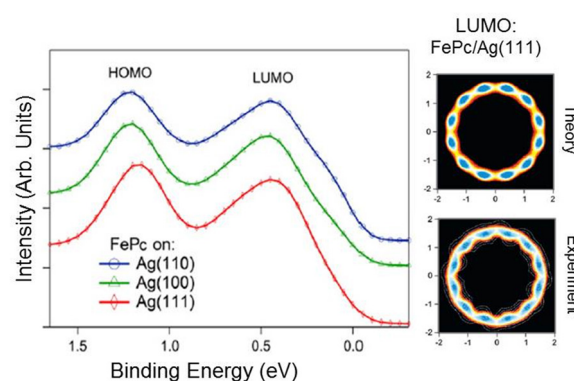


Fig. 3: Left: Photoemission spectra from FePc on Ag surfaces. Right: Comparison between measured k-space pattern and the correspondent calculated Fourier transform of the real-space LUMO orbital.

monolayer coverage ordered superstructures form on all low-index surfaces containing one molecule per unit cell for Ag(100) and Ag(111), and two molecules per unit cell for Ag(110).

In order to directly measure the full spatial spin distribution of magnetic molecule orbitals as well as hybrid states at organic-ferromagnetic interfaces, spin-resolved momentum microscopy is currently being developed employing a highly efficient 2-D spin filter.

- [1] M. Eschbach, **E. Mlynczak**, J. Kellner, J. Kampmeier, M. Lanius, E. Neumann, C. Weyrich, M. Gehlmann, P. Gospodaric, S. Döring, G. Mussler, N. Demarina, M. Luysberg, G. Bihlmayer, T. Schäpers, **L. Plucinski**, S. Blügel, M. Morgenstern, **C. M. Schneider**, and D. Grützmacher, *Realization of a vertical topological p-n junction in epitaxial $\text{Sb}_2\text{Te}_3/\text{Bi}_2\text{Te}_3$ heterostructures*, Nat. Commun. **6**, 8816 (2015). doi: [10.1038/ncomms9816](https://doi.org/10.1038/ncomms9816)
- [2] M. Eschbach, M. Lanius, C. Niu, **E. Mlynczak**, P. Gospodaric, J. Kellner, P. Schüffegen, M. Gehlmann, S. Döring, E. Neumann, M. Luysberg, G. Mussler, **L. Plucinski**, M. Morgenstern, D. Grützmacher, G. Bihlmayer, S. Blügel, and **C. M. Schneider**, *Bi_2Te_3 is a dual topological insulator*, Nat. Commun. **8**, 14976 (2017), doi: [10.1038/ncomms14976](https://doi.org/10.1038/ncomms14976)
- [3] M. Gehlmann, G. Bihlmayer, I. Aguilera, **E. Mlynczak**, M. Eschbach, S. Döring, P. Gospodaric, **S. Cramm**, B. Kardynal, **L. Plucinski**, S. Blügel, and C. M. Schneider, *Quasi 2D electronic states with high spin-polarization in centrosymmetric MoS_2 bulk crystals*, Sci. Rep. **6**, 26197 (2016), doi: [10.1038/srep26197](https://doi.org/10.1038/srep26197)
- [4] **E. Mlynczak**, M. Eschbach, S. Borek, J. Minár, J. Braun, I. Aguilera, G. Bihlmayer, S. Döring, M. Gehlmann, P. Gospodarič, S. Suga, **L. Plucinski**, S. Blügel, H. Ebert, and **C. M. Schneider**, *Fermi surface manipulation by external magnetic field demonstrated for a prototypical ferromagnet*, Phys. Rev. X **6**, 041048 (2016), doi: [10.1103/PhysRevX.6.041048](https://doi.org/10.1103/PhysRevX.6.041048)
- [5] **V. Feyer**, M. Graus, P. Nigge, M. Wiessner, R.G. Acres, **C. Wiemann**, **C.M. Schneider**, A. Schoell, and F. Reinert, *Adsorption geometry and electronic structure of iron phthalocyanine on Ag surfaces: A LEED and photoelectron momentum mapping study*. Surf. Sci. **621**, 64 (2014), doi: [10.1016/j.susc.2013.10.020](https://doi.org/10.1016/j.susc.2013.10.020)

Spintronics and Nanomagnetism

Nanosized magnetic structures are fundamental building blocks for spin transport devices. The research in PGI-6 therefore covers both the magnetism and spin transport on length scales down to the molecular level, leading to the field of molecular spintronics with potential applications in memory and logics. These activities are inherent to topic 2.

Molecular Spintronics

Research on molecular spintronics rests on three complementary pillars: chemical synthesis of molecules, theoretical modeling of molecule-substrate interfaces, and experimental realization and investigation of organic-inorganic hybrid systems. These are intensively pursued and closely cooperate in Jülich, targeting the development of single-molecule and hybrid spintronics as a major topic.

Magnetic molecules are potential functional units for molecular and supramolecular spintronic devices that provide a vast design space for all aspects relevant to their intrinsic quantized spin states and charge-transport characteristics. However, their magnetic and electronic properties depend critically on interactions with the metallic electrodes. Charge transfer and hybridization modify the electronic structure and thereby influence or even quench the molecular magnetic moment. Yet, detection and manipulation of the molecular spin state by means of charge

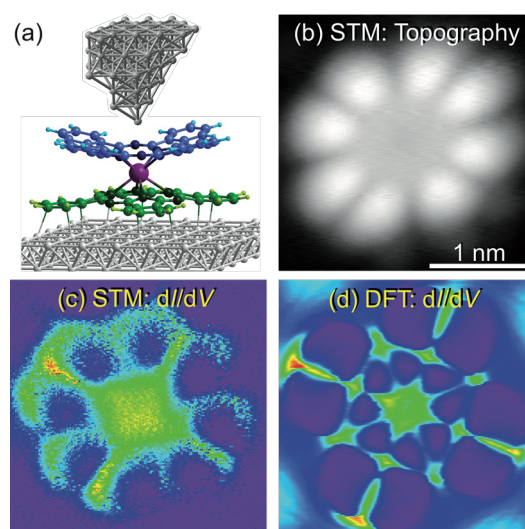


Fig. 4: (a) Calculated adsorption geometry of NdPc_2 on $\text{Cu}(100)$, (b) STM topography, (c) experimental, (d) calculated dI/dV -maps at -0.8 eV depicting the occupied Nd 4f-derived hybrid orbitals. [11]

transport, that is, spintronic functionality, mandates a certain level of hybridization of the magnetic orbitals with electrode states. In a joint effort between chemistry (Koegerler group), experiment, and theory (PGI/IAS-1) we have shown [1], how a judicious choice of the molecular spin centers allows us to control these critical molecule–electrode contact characteristics. In-house synthesized Nd double-decker phthalocyanine molecules served to study the correlation between adsorption on metal surfaces with different reactivity and intramolecular binding [2]. Combined scanning tunneling microscopy/ spectroscopy (STM/STS) experiments and DFT simulations reveal that, in contrast to the extensively studied late lanthanide analogues, the spin-polarized 4*f*-states of early lanthanides such as Nd [Figs. 4(a) and (b)] can be directly accessed by STM as evidenced by spatially and energetically resolved *dI/dV* conductivity maps of the Nd 4*f*-derived hybrid orbitals [Figs. 4(c) and (d)]. Hence, the 4*f*-orbitals contribute to charge transport, while retaining their magnetic moment as evident from comparing spectroscopic data with ab-initio calculations. These results showcase how tailoring molecular orbitals opens up prospects for electrical manipulation and detection of the molecular spin state, providing the foundation for all-electrically controlled spintronic device concepts in single-molecule spintronics.

An alternative approach to molecular spintronics offering prospects for room-temperature operation exploits interfacial phenomena in hybrid (molecule-surface) systems [3], specifically aromatic molecules chemisorbed on ferromagnetic thin films. The strong hybridization of molecular π -orbitals with *d*-orbitals of the substrate strongly enhances coercivity and blocking temperature. We have explored the site dependence of the spin-dependent molecule-substrate hybridization for polyaromatic triphenyl-triazine (TPT) molecules adsorbed on 2 ML-thin Fe films on W(110) and 2 ML-thin Co nanoislands on Cu(111). In both cases the hybridization-induced spin polarization detected by spin-polarized STM/STS revealed an asymmetric intramolecular distribution yielding different polarization values for the three phenyl groups of the TPT molecule. For the Fe surface this can be explained by a highly asymmetric chiral adsorption geometry [4]. In contrast, the adsorption geometry of TPT on Co(111) was experimentally and theoretically found to be fully symmetric. In this case, the symmetry breaking is due to the spin-dependent quantum interference pattern of the Co(111) surface state [5]. These results reveal two mechanisms for designing the intramolecular magnetic properties of molecule-surface hybrids on the nanometer scale. These measurements also lay the groundwork for hybrid spin filters such as tunneling magnetoresistors, in which the magnetic anisotropy of additional molecular components covalently linked to the chemisorbed π -groups is greatly enhanced via chemisorption-induced spin polarization effects.

In parallel, new molecular magnet materials based on nanoscaled molecular multi-metal oxides [6] have been developed that specifically address the challenge of precise, reproducible multi-terminal contacts and enable controllable coupling between the surface states and the magnetically relevant molecular orbitals [7]. With molecular charge- and spin-dependent conductivities, these systems also represent prospective molecular memristors.

Spin-Torque Oscillators

The gyrotropic rotation around the equilibrium position constitutes the fundamental excitation of magnetic vortices in nanostructures and has recently been exploited in spin-torque-driven nano-oscillators. Main advantages are the extremely narrow line width of the HF output signal and the operation without applied magnetic field. The gyrotropic mode usually varies with material and sample geometry, but is independent of the vortex vorticity and its core direction. We have demonstrated that this degeneracy is lifted in a spin-torque oscillator containing two vortices stacked on top of each other [8]. When driven by spin-polarized currents, such devices exhibit a set of dynamic modes with discretely split frequencies, each corresponding to a specific combi-

nation of vorticities and relative core polarities [Fig. 5(a)]. Micromagnetic simulations [Fig. 5(b)] and an analytical model corroborate this finding and reveal that the fine splitting arises from the Zeeman energy of the Oersted field and the hyperfine splitting from the variation of the gyration orbits due to the dependence of the vortex-vortex coupling on the relative alignment of the vortex core polarities. The resulting rich fine structure of the oscillatory modes occurs even in the absence of external fields, demonstrating that such devices can function as zero-field, multi-channel, nano-oscillators for communication technologies. It also facilitates the detection of the relative core polarization and allows for the eight non-degenerate configurations to be distinguished electrically, which may enable the design of multi-state memory devices based on double-vortex nanopillars.

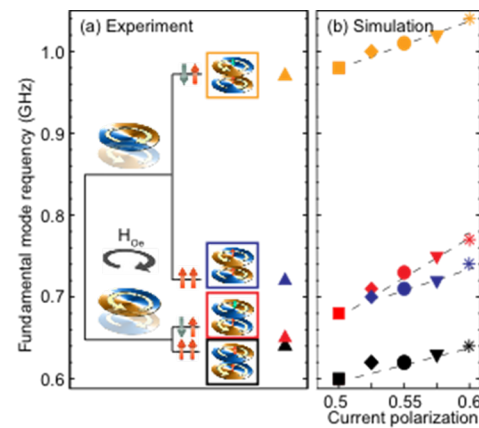


Fig. 5: (a) Measured and (b) simulated fundamental mode frequencies for different vorticity and polarity combinations of the stacked vortices exhibit a particular mode splitting. Simulations are carried out for various current polarizations.

Oxide Spintronics (Young Investigator Group M. Müller)

Functional oxides play an important role in the development of spintronics, due to their broad variety of electronic, magnetic and ferroic properties. The complexity of oxide materials, however, poses enormous challenges for synthesis, characterization and functionalization in heterostructures, at interfaces and quantum wells, and realizing spin-dependent transport. Motivated by unlocking the rich potential of 3d and 4f-based ferro(i)magnetic oxides for spintronic applications, the Young Investigator Group (YIG) explores the interrelations between magnetism, electronic structure and transport phenomena in novel heterostructures, with the goal to merge insights from materials science, synchrotron radiation-based spectroscopy and nanoscale transport. In particular, the impact of low dimensionality, strain, interface and defect chemistry is investigated – corroborated by mastering the property control of heterostructures at the atomic-layer level.

As a highlight, the YIG solved the long-standing challenge of a seamless *in-situ* integration of a spin-functional magnetic oxide on silicon wafers [9]. The inherent chemical reactivity between most (binary or more complex) magnetic oxides and Si has prevented a heteroepitaxial integration to date, which, however, is a perfect route to enrich silicon nanotechnology with spin filter functionality. According to theoretical predictions, the magnetic oxide Europium Oxide (EuO) is the only binary magnetic oxide thermodynamically stable in direct contact with silicon. As a rare material in nature, EuO unites ferromagnetic order and insulating properties. In the few nanometer regime, EuO acts as a highly efficient spin-selective tunnel barrier due to its exchange-split conduction band. Moreover, its insulating band gap matches that of silicon very well, thus overcoming the conductivity mismatch problem. Thus, ultrathin EuO films are perfectly suited as efficient spin filter contacts directly on clean silicon. Interface control was realized by applying

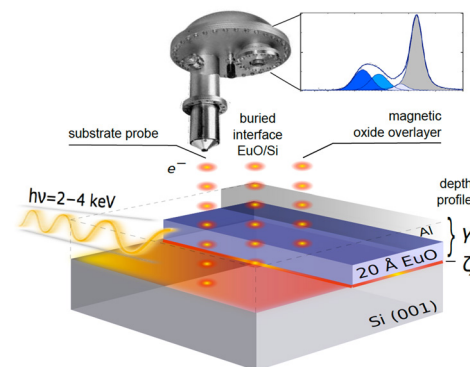


Fig. 6: Schematics of a depth-profiling HAXPES experiment in order to probe a functional buried oxide-Si interface

complementary passivation techniques for the reactive Si (001) surface, without any additional buffer layers, by employing an advanced molecular beam epitaxy growth process. By chemical depth profiling of the oxide-semiconductor interface via hard x-ray photoemission spectroscopy (HAXPES) (Fig. 6) [10], it was shown, how to systematically minimize both Eu silicide and Si oxide formation to the sub-monolayer regime – and how to ultimately interface-engineer chemically clean, heteroepitaxial and ferromagnetic EuO/Si (001) in order to create a strong spin filter contact to silicon.

Borderline Magnetism (Young Investigator Group C. Schmitz-Antoniak)

The Helmholtz Young Investigators Group "Borderline Magnetism" investigates the influence of surface modifications and tuned valences to tailor the electronic and magnetic properties of 3d and 4d transition metals, their alloys, and oxides at the nanoscale. For this purpose a UHV chamber for thin film preparation, characterisation and x-ray absorption spectroscopy was set-up and the wet-chemical synthesis of metal/oxide hybrid nanoparticles has been established in our group.

To tailor the properties of the materials for applications, a detailed understanding on a microscopic, i.e., atomic, scale is crucial. For this purpose we employ synchrotron based spectroscopy methods, in particular the analysis of x-ray absorption near-edge structure (XANES), magnetic circular and linear dichroism (XMCD, XLD), as well as the extended x-ray absorption fine structure (EXAFS).

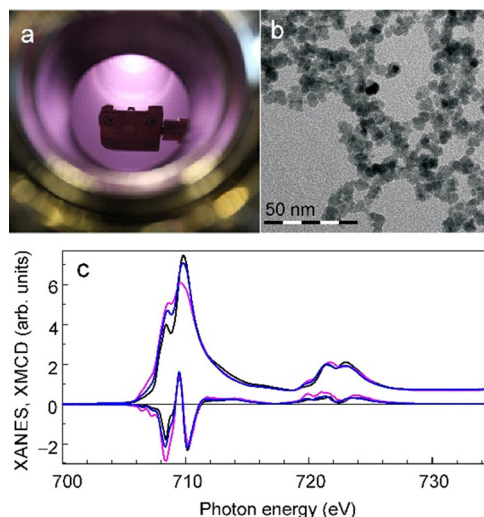


Fig. 7: An *in situ* hydrogen plasma treatment (a) improves the properties of magnetite nanoparticles (b) for applications as evidenced by XANES and XMCD (c) [11].

As a highlight of our recent research, we could fully reverse ageing effects of magnetite nanoparticles by a soft hydrogen treatment in a self-constructed portable plasma chamber [10]. Monitoring the Verwey transition by temperature dependent changes of the intra-atomic spin dipole term, we showed that structural, electronic and magnetic properties of plasma treated magnetite nanoparticles can even outperform the ones of the freshly prepared batch (Fig. 7). In addition, atomic hydrogen is known to recover the half-metallic character of the magnetite surface, which is essential for miniaturised spintronic applications.

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Ultrafast Spin Dynamics

The dynamics in magnetic systems, particularly on ultrashort time scales, currently receives strong interest. This is driven by the search for ultrafast magnetic switching in information technology, but also the need to understand the fundamental mechanisms governing energy and angular momentum transfer on femtosecond time scales. This understanding must include the complex interactions between the electron, spin, and lattice subsystems.

In the framework of an extensive collaboration with groups in Kaiserslautern, Uppsala, and JILA Boulder, we carried out several key experiments on ultrafast demagnetization processes in ferromagnets, using a pump-probe technique with high harmonic generation (HHG). The HHG technique converts the near-infrared pulses of a table-top femtosecond laser into the XUV regime, covering the M-edges of the transition metals. In this way, we are able to study magnetization dynamics element-selectively, separating the individual contributions in a layered structure. In the first key experiment, we disentangled the spin dynamics in trilayer spin-valve type systems with interlayer exchange coupling through a nonmagnetic interlayer [1]. These systems are major building blocks for spintronics devices. We could observe a quite unexpected response of the system, which could only be explained by a new demagnetization mechanism, namely the action of optically excited superdiffusive spin currents between the magnetic layers (Fig. 8). The generation of highly spin-polarized superdiffusive currents from Ni is favored by the intrinsic spin-filtering in Ni during the transport and the preservation of the spin-polarization during transfer through the nonmagnetic interlayer. This superdiffusive transport generally competes with other spin-dephasing mechanisms, such as Elliot-Yafet spin scattering, but becomes dominant in layered systems. In a subsequent set of experiments we clarified the role of the interlayer electronic structure on the transient spin currents [2]. We also showed that an insulating interlayer, which only allows a tunneling transport, strongly suppresses superdiffusive

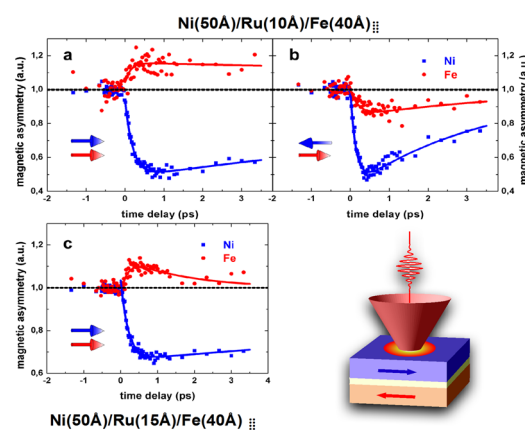


Fig. 8: Ultrafast and element-selective spin dynamics in coupled layer Ni/Ru/Fe stacks. The dynamic response caused by superdiffusive spin currents from the top Ni layer depends on the parallel (a) or antiparallel orientation (b) of the Fe and Ni magnetization. After [1].

spin transport. A potential application of superdiffusive spin currents could be an ultrafast magneto-optical switching in magnetic data storage.

Although the above experiment gives already a detailed insight into the demagnetization processes, we are still missing the direct link to the response of the electronic structures. This aspect was addressed in a second key experiment involving time, spin- and angle-resolved photoemission [3]. For the first time, we could follow the evolution of band states in the 3-*d* ferromagnet Cobalt during ultrafast laser excitation on a 100 fs time scale, mapping the response of minority and majority states separately. We find the excitation to affect the entire electronic structure and not only the states close to the Fermi level, and to be also strongly *k*-dependent. This is most likely due to correlations and many photon excitations. The response of minority and majority spins states exhibits significant differences. Overall our results are compatible with a fluctuating band model, which assumes the persistence of the exchange splitting of the bands also in the fully demagnetized state.

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Redox Processes

Redox processes play a key role in numerous scientific fields, i.e., energy conversion and storage, catalysis, environmental and earth sciences as well as the information technology. A control mechanism in redox-based devices is usually introduced at an interface of two materials - either solid-solid, or solid-liquid/solid-gas. Synchrotron based spectroscopic and microscopic techniques proved to be a valuable asset in investigating interfacial redox phenomena and, in the cases we introduce below, it is a very dynamic and fast growing field. These activities contribute to topic 3, as well as to energy research.

Resistive Switching

The semiconductor industry's roadmap lists memristors as a possible pathway towards highly scalable non-volatile memory. Although prototypes of resistive memory are already entering the market, many of the microscopic mechanisms are still poorly understood. Oxides are a dominating class of materials in this research area. In close collaboration with PGI-7 and the SFB 917 we focus on micro-spectroscopic studies of redox processes on the nanoscale. Several oxidic materials with different switching characteristics, namely Strontium Titanate, Gallium Oxide [1] and Tantalum Oxide [2] have been structured into resistive switching contacts and investigated

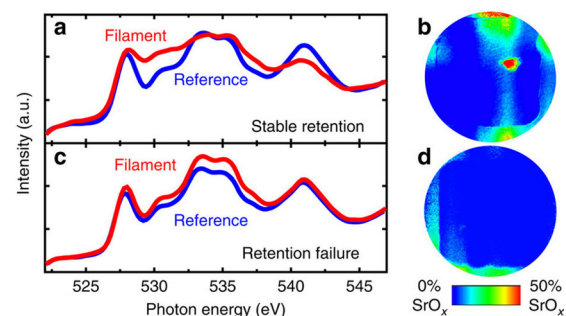


Fig. 9: Nanoscale redox processes in SrTiO_3 resistive switching devices showing the chemical state in a filament (red dot in (b)) observed in x-ray absorption spectra (a), and the lateral distribution in PEEM (b). Retention failure causes a change in the oxidation state which is reflected in the spectra (c) and in the disappearance of the filament contrast (d).

using photoemission techniques, The necessity to access interfaces below a metallic electrode stimulated the development of energy-filtered PEEM with hard x-rays (HAXPEEM). It combines the high probing depth of hard x-ray photoelectron spectroscopy (HAXPES) with the lateral resolution of photoemission microscopy (PEEM) into a method capable of imaging and analyzing buried structures and interfaces in a non-destructive manner [3]. This approach can even evaluate the inelastically scattered electrons to yield information about the depth profile of chemical constituents in the samples investigated [4].

A strong focus of our studies concerned the model system SrTiO_3 (STO). A main challenge posed by STO lies in the fact that here the resistive switching proceeds via the formation of nanoscale conductive filaments. Employing different microspectroscopy methods with soft as well as hard x-rays we were able to directly observe the relevant redox processes within the filamentary areas [5]. Moreover, we could unambiguously connect these to the local formation of oxygen vacancies and a corresponding valence change of the Titanium ions. In addition, we could make an important contribution to improving the stability and reproducibility of the switching process. A central problem of resistive memories is the volatility of the stored information. The required retention time of >10 years is still out of reach for most resistive memory concepts. In the study cited above we were able to show that the retention quality can be improved considerably by the introduction of an additional Al_2O_3 layer acting as barrier for oxygen migration (Fig. 9). In a further key experiment, using in-operando methods, we could quantitatively observe for the first time the modification of the Schottky barrier between electrode and STO during the switching process [6].

Chemistry at solid-liquid interfaces

Many of the redox applications rely on processes taking place at solid-liquid interfaces. This is especially true for energy-related devices – batteries, supercapacitors, electrolysis cells, fuel cells, etc. There is a high need for appropriate *operando* characterization techniques in the energy research area (IEK institutes at FZJ). Due to their very nature, liquid samples are incompatible with ultra-high vacuum probes unless special experimental measures are taken. On the other hand, photoemission spectroscopy is a very powerful tool for investigating interfaces, since, beyond the conventional elemental and chemical analysis, it is also sensitive to electric fields and can be used for a direct mapping of electrical potentials by observing core-electrons energy shifts. However, one missing aspect is depth selectivity, which in a traditional X-ray photoemission is only achieved via the exponential attenuation of the photoemission signal. This can be overcome by a spatially non-uniform excitation, i.e., an X-ray modulation using the standing wave approach.

Recently, PGI-6 in a collaboration with LBNL developed SWAPPS - standing-wave ambient pressure photoemission spectroscopy - a powerful tool for studying solid-liquid interfaces. The first-of-a-kind SWAPPS study was performed on a prototypical water splitting material – hematite. A Si/Mo multilayer mirror was used as an X-ray standing-wave generator (Fig. 10), with a thin hematite film grown on top of it. A 0.01M water solution of $\text{NaOH}+\text{CsOH}$ was drop-cast onto the film. Detailed parameters of the solid-liquid interface and the electrical

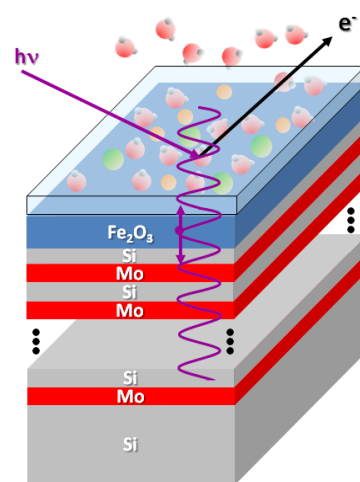


Fig. 10: A sample schematics. A thin hematite film is grown on a Si/Mo multilayer. The liquid layer on top is a droplet of 0.01M solution of $\text{NaOH}+\text{CsOH}$. Reproduced from [8].

double layer, such as depth concentration profiles of the individual ionic species as well as the interface roughness and contaminant distribution, were measured with an unprecedented accuracy [7]. The exquisite depth resolution also revealed different chemical processes taking place in different proximity of the solid-liquid and liquid-gas interfaces, providing a truly interface sensitive picture with sub-nm precision. Another application of this newly developed technique was a corrosion study of a thin nickel film in a working electrochemical cell [8].

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Collaborations

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Planned New Directions

In an effort to strengthen the coherence of the research activities in PGI-6 with respect to the future program and Center strategy towards information research, the following activities will be started, or have been initiated quite recently:

- **Molecular Spin Hybrids** – we will use the interactions between molecule and surface to create magnetic hybrid units with new magnetic properties (e.g., magnetic moment, anisotropy) and tailored spin-transport properties. This includes molecules with specifically designed metallic ligands, which serve as well-defined electrical contacts.
- **Spin Dynamics** – we will expand our studies on ultrafast spin dynamic processes to complex 3d-4f material systems and nanostructured samples to disentangle the spin

excitation/ relaxation processes, interactions between the sublattices, and effects of lateral confinement on the superdiffusive spin currents. The studies will also be extended to materials with antiferromagnetic spin order to make a link to antiferromagnetic spintronics.

- **Momentum Microscopy** – the recently established technique of momentum or k-space microscopy permits access to the electronic structure with unparalleled detail. We will exploit these capabilities in our ongoing studies of quantum materials. We will further incorporate spin and time-resolution into this approach to address electron and spin dynamics in the entire Brillouin zone during ultrafast optical excitations. This will give the long-sought interrelations between the details in the electronic structure in k-space and the ultrafast spin dynamics.
- **Resistive Switching Dynamics** – our activities on redox processes in resistive switching materials will be extended into the time domain. Employing time-resolved photoemission spectroscopy and microscopy with soft and particularly hard x-rays we want to address the dynamics of the relevant redox processes and chemical changes in real time.
- **JuSPARC** – the start of the project JuSPARC as an experimental facility for ultrashort photon pulses in the XUV and soft x-ray regime gives the ultrafast dynamics activities a strong push. Priority will be given to the construction of two experiments: spin- and time-resolved momentum microscopy and time-resolved soft x-ray scattering up to 200 eV photon energy. These will strongly improve our capabilities in dynamic electronic structure studies.

2.4.7 PGI - Electronic Materials (PGI-7)

Head of Institute: Prof. Dr. Rainer Waser

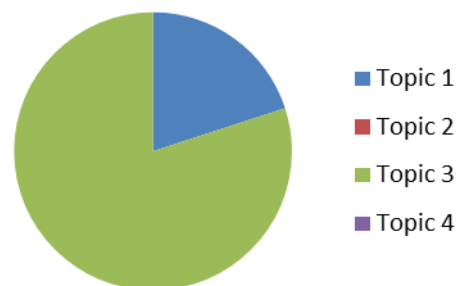
Contributing Personnel (31 Dec 2016 in FTE):

Core-funded: 13 scientists, 2 doctoral students, 7 scientific support personnel

Third-party funded: 0 scientists, 5 doctoral students, 0 scientific support personnel

Contributing Principal Investigators: Regina Dittmann, Felix Gunkel, Susanne Hoffmann-Eifert, Silvia Karthäuser, Stephan Menzel, Marco Moors, Vikas Rana, Kristof Szot, Ilia Valov, Rainer Waser

The Institute of Electronic Materials focuses on the physics and chemistry of oxides and organic molecules which are promising candidates for non-volatile memories, i.e., storage class memory, as well as beyond-von Neumann architectures such as ‘logic in memory’ concepts and brain-inspired neuromorphic computing. The research is aiming at the basic understanding of electrochemical redox-processes, space charge effects as well as ferro- and piezoelectricity in complex electronic materials. Due to the close similarity of the underlying redox- and oxygen exchange mechanisms of memristive switching to the field of catalysis and the expected cross-fertilization, we are exploring oxides’ surfaces and interfaces for sensors and energy conversion applications such as water splitting. With these activities, the PGI-7 is mainly involved in the PoF III **topic 3 “Controlling configuration based phenomena”** but furthermore contributes to **topic 1 “Controlling electron charge-based phenomena”** and **topic 4 “Controlling collective states”**.



PGI-7 mainly contributes to Topics 1 and 3.

The research activities in PGI-7 comprise fundamental research on the defect chemistry and redox-processes of binary and complex oxides in bulk and thin film form as well as at dedicated, atomically tailored surfaces and interfaces. The PGI-7 is employing a variety of thin film deposition techniques such as sputtering, atomic layer deposition and pulsed laser deposition for the growth of amorphous or polycrystalline thin films on CMOS compatible substrates as well as for the atomically tailored growth of epitaxial thin films and interfaces. We are addressing in detail the changes induced under thermal- and chemical gradients and electrical biasing by a broad variety of state-of-the-art analysis methods. For that purpose, the PGI-7 is operating the **electronic oxide UHV cluster-tool (EOC)**, which enables the *in situ* characterization and nanoscale electrical manipulation of oxide thin films and devices by scanning probe techniques and photoelectron spectroscopy and -spectromicroscopy. These investigations are complemented by high-resolution transmission electron microscopy and -spectroscopy within the **PGI-5** and **ER-C** as well by synchrotron-based spectroscopic and spectromicroscopic techniques in close collaboration with the **PGI-6**. These activities are embedded in the interdisciplinary **SFB 917 “Resistively switching chalcogenides for future electronics-Structure, Kinetics and Device Scalability”** with participating groups from different PGIs and the RWTH Aachen University.

Moreover, the **Helmholtz Nanoelectronic Facility** is employed to fabricate memristive micro- and nanostructures as well as cross-bar arrays for resistive random access memories (ReRAM) or logic-in memory applications in close cooperation with the **PGI-10**. The **Helmholtz UHV Nanocluster** is employed for *in-situ* stacking of ultrathin oxide and metal layers grown by off-

axis sputtering and atomic layer deposition. The tool is utilized for ReRAM research focusing on interfacial effects of *in-situ* stacked sub 10 nm thin metal oxide and metal layers for concepts on **three dimensional integrated ReRAM**.

Basic switching mechanisms of nanoionic memristive systems

The PGI-7 is focusing on two different types of nanoionic memristive systems that show bipolar switching, namely **electrochemical metallization** (ECM) cells and **valence change mechanism** (VCM) cells. ECM cells operate by an electrochemical dissolution of an active electrode metal such as Ag or Cu, a drift of cations through an ion conductor and a formation of a metal nanofilament. We were able to show that electrode materials believed to be inert become electrochemically active in ECMs, e.g., Pt, Ti and Ta in SiO_2 . We have furthermore demonstrated that moisture plays a crucial role in the mechanism of ECM switching [1] and that it has a strong impact on the switching kinetics. This effect is related to the electrocatalytic activity of the electrode materials used in ECM devices.

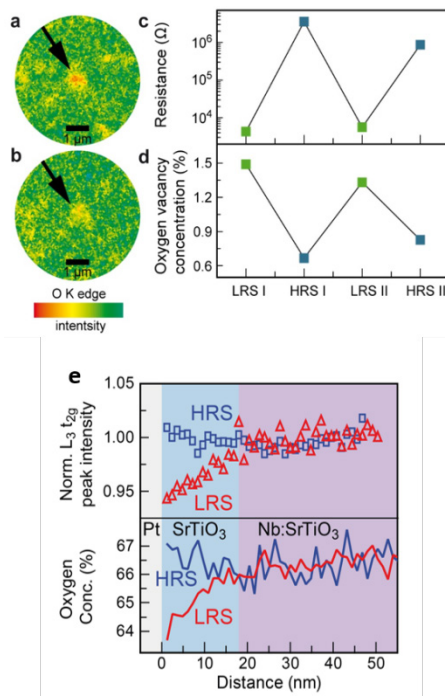


Fig. 1: *In operando* PEEM measurements on SrTiO_3 cells PEEM images of the filament region in LRS (a) and HRS (b) at the O K edge. Resistance change (c) and related change of the oxygen vacancy concentration (d) during repeated switching; [3] (e) Upper panel: Intensity of the EELS $\text{Ti } L_{3\ 23}$ edge as a function of depth into the specimen for the LRS and HRS. Lower panel: Oxygen concentration profiles acquired by integrating the oxygen EELS edge for the same region in the HRS and LRS.[6]

VCM switching is typically found in metal oxides that show sufficient ion mobility. The migration of these ions changes the local stoichiometry and, hence, was suggested to lead to a redox-reaction accompanied with a valence change of the cation sublattice and a change in the electronic conductivity. The PGI-7 gained significant knowledge about the microscopic mechanism of electroforming and resistive switching in a variety of VCM systems containing binary oxides (TiO_2 , HfO_2 , Ta_2O_5) as well as complex oxides (SrTiO_3 , $\text{Pr}_{1-x}\text{Ca}_x\text{MnO}_3$) thin film devices and about the impact of extended defects, stoichiometric variations and interface reactions.

By employing **spectroscopic methods** we could in close collaboration with the PGI-6 explicitly show that resistive switching in SrTiO_3 and Ta_2O_5 involves a **valence change** in the metal ions within nanoscale filaments. We could furthermore reveal the SrO segregation process as the underlying mechanism for the retention stabilization in memristive SrTiO_3 thin film cells and proposed based on this a strategy for a rational design of stable memristive devices [2].

By employing graphene electrodes as photoelectron transparent electrodes for the ***in-operando*** characterization of memristive devices we obtained a breakthrough in the quantification of redox-processes taking place during the operation of SrTiO_3 devices [3]. As shown in Fig. 1 we could identify an oxygen deficient filament and could determine the oxygen vacancy concentration and monitor their change during repeated switching. Using this quantitative numbers as input for nanoionic device simulations demonstrates remarkable

agreement between simulation and experiment. In cooperation with the ER-C and D. Cooper from CEA-LETI, Grenoble we utilized ***in-operando* transmission electron microscopy** (TEM) and **electron energy loss spectroscopy** to clarify the vertical changes of the oxygen vacancy

concentration during resistive switching (Fig. 1) [4]. We could thereby identify an oxygen exchange reaction at the interface as switching mechanism for an anomalous switching mode with a switching polarity that is in contradiction to an internal redistribution of oxygen vacancies.

The ohmic interface at the Ta/Ta₂O₅ accommodates electrochemical electrode reactions and allows for faster exchange of ions (oxygen and Ta ions) and is a crucial part of the switching kinetics [5,6]. By studying the set and reset dynamics in a Pt/Ta₂O₅/M device with different electrodes 'M' including Hf, Ti, Ta and W, it is demonstrated that the oxygen exchange reaction occurring at the Ta₂O₅/M interface influences the switching dynamics [7]. The studies on the importance of the ohmic interface give a further insight into the switching mechanism and open up a wider set of design parameters for tailoring ReRAM cells.

Although it is usually anticipated that oxygen anions are the mobile species in VCM cells, we could show sufficient mobility of different metal ions such as Ta in TaO_x, Ti in TiO_x and Hf in HfO_x to participate and concur with oxygen ions [5]. Suppressing the oxidation process at the interface by introducing a graphene or carbon layer, results in a **transition from VCM to ECM switching** as has been observed for Ta₂O₅ cells [6]. As determined for ECM cells, it has also been demonstrated that moisture significantly influences the electrochemical reactions and the forming process of VCM cells [6]. Thus, moisture effects appear to be a general trend rather than an exception.

Besides the observed valence changes, for some materials resistive switching is accompanied by crystallographic **phase transformations**. We analysed the surface of TiO₂ single crystals upon reduction in UHV by local-conductivity atomic force microscopy (LC-AFM). We found that using a dedicated sequence of high-temperature reduction with subsequent room temperature oxidation a quasi-homogeneous distribution of switchable nanofilaments can be generated [8]. To gain an insight in the electronic structure of the nanofilaments, ab initio calculations of these defect arrangements were performed at the **PGI-1**. These calculations show that more or less extended defect states split off the conduction band and reside in the gap. In contrast to larger defects or defect-bundles that could describe dislocations, the metallic pathways in these oxygen-vacancy rows are easily disrupted by adding oxygen atoms. Thus, these structures would be compatible with the electrically active nanofilaments. Together with the fact that the switchable filaments were found to be distributed on the surface with a density of 10¹² cm⁻² (much higher in density than observed for single crystals so far) we conclude that the observed linear defects have a promising potential for building memristive devices with high storage density.

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Memristive devices for memory and neuromorphic applications

Within the current POF period, the PGI-7 addressed key questions related to the use of memristive devices as novel **non-volatile memory elements in embedded and new storage class memory devices**, such as device scaling, reliability, switching speed, switching kinetics and power consumption. Therefore, our application oriented research has been pursued through fabrication and characterization of **nano crossbar**-type resistive switching metal/metal oxide/metal stacks. Memristive cells built from almost all technically interesting metal oxides, such as, HfO₂, Ta₂O₅, TiO₂ as well as oxide bilayers like Al₂O₃/TiO₂ and HfO₂/TiO₂ in combination with various metal electrodes have been studied. Additionally, ALD SrTiO₃ and ALD Nb₂O₅ have been provided through co-operation in an EC founded FP7 Marie Curie project.

The cell area has been **scaled to 60 x 60 nm²** and the thickness of the metal oxide layer has been reduced to about 5 nm enabled by continuous improvement of nanolitho-

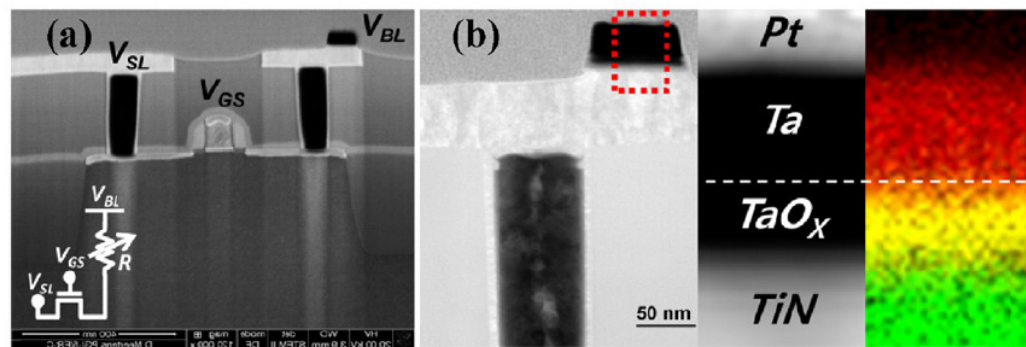


Fig. 2: Cross-sectional TEM image of ReRAM device in the 1T-1R configuration. (b) Enlarged TEM cross-section of the ReRAM device stacks (TiN/TaO_x/Ta/Pt) and energy-dispersive X-ray (EDX) analysis.

graphy and etching techniques as well as atomic layer deposition processes. We have furthermore fabricated and tested resistive random access memory cells (ReRAM) on top of CMOS transistors (1T-1R configuration) as depicted in Fig. 2. The set switching kinetics of VCM cells was investigated over 14 orders of magnitude in switching time, which extended earlier studies by several orders of magnitude. In search for the **ultimate switching speed** a dedicated coplanar wave guide Ta₂O₅-based VCM cell has been integrated. Using this kind of cell switching below **300 ps** could be demonstrated.

The forming process in resistive switching devices is a **high-power and additional-time consuming step**. The required forming voltage is generally much higher than the switching voltage, making the resistive switching devices incompatible to low-voltage CMOS technology. We therefore optimized the sputter deposition process for Ta₂O₅ thin films and obtained ReRAM device with a reasonable low forming voltage (1.8 V) with large memory window ($R_{OFF}/R_{ON} > 300$), and stable endurance up to 10⁶ cycles with good retention of 10⁴ s at 125 °C. By employing an optimal bilayer (Ta₂O₅/TaO_x) combination we succeeded to reduce the switching currents to 50 μA.

Forming-free switching cells were realized for different materials' systems such as for TiO_2 by control of the ALD process [1] and for Ta_2O_5 and HfO_2 by introducing a CMOS compatible ion (O_2 and N_2) implantation step.

One of the key issues of the use of passive cross-bar arrays for ReRAM is the problem of selecting a designated cell without interference from sneak-path currents through neighboring cells. One approach to solve this problem is the use of selector devices with a high non-linearity in the current-voltage curves. We succeeded to show NbO_2 cells with **selector function** by integrating 10 nm thin amorphous ALD Nb_2O_5 films with Pt and Ti electrodes. For the volatile threshold-type selector function in these devices we could overcome the shortcomings of the established 'metal-insulator transition' model by an alternative explanation based on an 'electric field induced thermal runaway effect' [2].

We furthermore started within this POF period to explore the properties of memristive devices for brain inspired neuromorphic computing. For example, by using the **multi-level switching capability** of Ta_2O_5 ReRAM device we succeeded to demonstrate a ternary number based modular arithmetic function [3]. Interlinked with **PGI-10**, we realized ReRAM-based **fuzzy logic gates** with quasi-static and transient pulse measurements, enabling analogous minimum and maximum operation and **logic-in-memory** [4] functionality in passive crossbar arrays. These approaches could be the basis for realizing **synaptic functionality in brain inspired computing**.

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Modelling of memristive devices

For filamentary switching VCM cells one goal of the PGI-7 was the verification of local Joule heating being the origin for the **ultra-nonlinear switching kinetics**. The analyses of the data revealed that the origin of the nonlinear switching kinetics is mainly determined by the temperature-accelerated migration of ionic defects. By a careful analysis of the current transients during the application of a set voltage pulse it was found that the current first gradually increases, which finally leads to an abrupt current jump setting the device to the low resistance state (LRS) [1]. By comparing these data to the simulation results of a compact model, a thermal runaway process induced by the positive feedback of increasing current and Joule heating could be identified as its origin.

The understanding of the dynamics of the resistive switching phenomenon was put forward by the **development of simulation models** and the comparison to experimental data in particular to the obtained switching kinetics data. In the last period simulation models for ECM and VCM cells have been developed. For ECM cells, a physics-based 1D model based on the growth/dissolution of a metallic filament was proposed that includes nucleation, electron-transfer processes, ion migration and electron tunnelling. With this model it could be shown that the set switching speed of AgI-based ECM cells are limited by nucleation at low voltages, electron-transfer processes at intermediate voltages and ion migration at high voltages. A deeper understanding of the filamentary growth was achieved by the development of a two-dimensional

kinetic Monte Carlo Model for ECM cells [2]. By including the mechanical stress induced by the growing filament into the model, it could be shown that a wire-like filament evolves for materials with a high Young's modulus. For materials with a low Young's modulus a wider filament grows or even a dendritic structure evolves during set switching.

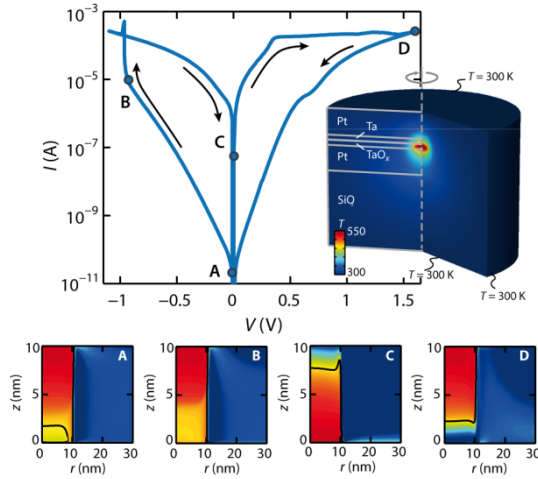
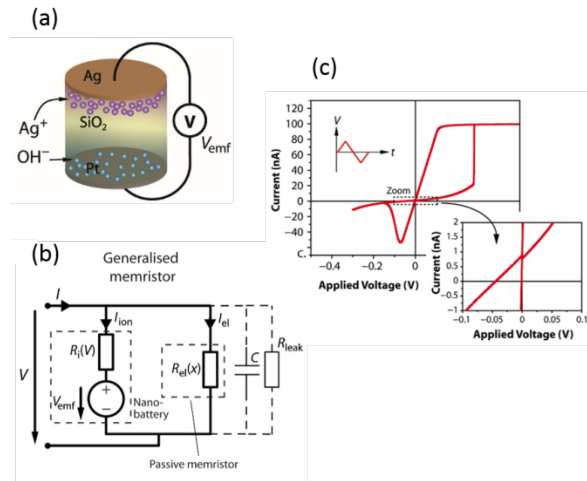


Fig. 3: Simulated quasi-static I-V characteristic of a Ta_2O_5 -based VCM cell [3]. The inset shows the 2D axisymmetric computational domain used for the temperature calculation along with the boundary conditions for the heat equation. The color map shows a typical temperature distribution as example. A to D show the oxygen vacancy concentration maps at the center of the Ta_2O_5 layer illustrating the redistribution of the oxygen vacancies during resistive switching at the points marked in the I-V characteristics.

Fig. 4: (a) Sketch of an ECM cell nanobattery. (b) Equivalent circuit of the extended memristive element. The ionic current is defined by the nanobattery, which controls the state-dependent resistor representing the electronic current path. (c) Simulated I-V characteristic of the extended memristor. [4]



For VCM cells a multi-dimensional model of nonisothermal drift-diffusion transport including the effect of contact potential barriers was developed [3] (see Fig. 3). The model accurately describes the experimental reset switching data of Ta_2O_5 -based cells and provides insights into the processes determining the gradual reset. The gradual nature can be attributed to the temperature-accelerated oxygen-vacancy motion being governed by drift and diffusion processes approaching an equilibrium situation. Furthermore, the simulation model also reproduces important switching phenomena like the I-V characteristics, the occurrence of complementary resistive switching and the set kinetics.

We have furthermore shown for the first time on a theoretical and experimental level that both ECM and VCM type ReRAM cells are inherently in a non-equilibrium state and behave as **nanobatteries**. The non-equilibrium states are controlled by thermodynamics (Nernst and/or Gibbs-Thomson potentials) and kinetic factors (diffusion potentials). The nanobattery effect makes a major amendment in the memristor theory, demonstrating that ReRAMs are active (instead of as postulated - passive) circuit elements [4]. We were able to propose a generalized memristor model by implementing the nanobattery effect (Fig. 4). We have furthermore shown

that the nanobattery effect influences the switching kinetics as well as the device failure such as retention and endurance [5, 6,].

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Thermodynamics and kinetics of redox-processes at surfaces and interfaces

Defect chemistry and thermodynamics on the nanoscale is at the heart of fundamental understanding of oxide thin film properties, their surfaces and interfaces. In particular, the nanoscale confinement of defects at surfaces and interfaces relates to a variety of applications of functional oxides such as memristive devices, all-oxide transistors, or gas sensors.

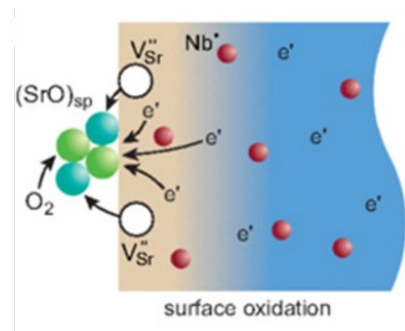


Fig. 5: Formation of electron depletion layer at Nb:SrTiO₃ surfaces. [1]

In a series of fundamental studies, we have developed nanoscale thermodynamic models that address electric fields, broken symmetry, undercoordination, and kinetic limitations occurring at surfaces and interfaces of complex oxides. Classical thermodynamic lattice disorder models need to be substantially expanded in order to embrace the nanometer length scale of modern oxide electronic devices.

In one approach, we used SrTiO₃ as model system, similar to our model material being explored for epitaxial resistive switching devices. For example, we discovered that an inherent electron depletion layer forms at the surface of donor-doped SrTiO₃ (and other oxides), due to the accumulation of cation vacancy defects that form in the very surface layer as a natural result of oxidation as illustrated in Fig. 5. Due to their slow kinetics, these vacancy defects are **kinetically confined** to the surface region on moderate time scales. Surface space charge formation explains the weak surface conductivity often observed even in highly *n*-type doped SrTiO₃ crystals, the thickness-dependent lack of carriers in *n*-type doped oxide thin films, as well as the unexpected band bending observed in resistive switching cells using Nb:SrTiO₃ bottom electrodes.

In a recent work we were able to show that the electron depletion layer depends on the ambient atmosphere, making space-charge-control in these materials a potential tool to realize oxygen sensors. Generating polar discontinuity at oxide interfaces in atomically defined heterostructures can result in a charge-transfer-based space charge region. Such space-charges can infer the formation of **2-dimensional electron gases (2DEG) at complex oxide interfaces**, which have evolved as a hot research topic within the last decade. These electron

systems are considered for multi-functional all-oxide transistors and gas sensors, thus complementing our research on memory devices.

The properties and performance of such 2DEG-devices depends on delicate control of the defect structure, which we addressed in our research activity. During the current POF period, we were able to reveal the fundamental thermodynamic processes determining the defect formation, taking place mainly during the fabrication process of such 2DEG structures. We furthermore succeeded to relate the limited electron mobility observed in these 2DEGs to strontium vacancy formation and demonstrated means to manage and suppress the formation mechanism based on our approach [2]. Furthermore, we revealed a relation of emerging magnetism at these interfaces and their defect structure [3].

Redox reactions and thermodynamic stability of surfaces and interfaces have also been studied at the metal/metal oxide interfaces of memristive devices consisting of polycrystalline or amorphous binary oxides. We have shown by X-ray absorption spectroscopy and cyclic voltammetry that intermediate oxides typically of different short range order (compared to main oxide matrix) always form at the metal-metal oxide interface irrespective of the predicted thermodynamic driving force. Such intermediate layers were observed for Me/MO_x with Me = Cu, Ta, Hf, Ti and M = Si, HfO₂, Ta₂O₅ interfaces [4]. We found that only the thickness of this intermediate layer depends on the thermodynamics.

We furthermore demonstrated that scanning tunneling microscopy can be used as a powerful tool for studying surface redox reactions. Chemical changes at the surfaces have been induced and verified by scanning tunneling spectroscopy for TaO_x and SrRuO_{3-x}[5]. Thus, we demonstrated the highest limit of lateral and mass/charge resolution for measurements of redox processes, allowing studies and analysis independent on the electronic conductivity.

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Interfaces in carbon based electronics

Besides nanoelectronic devices based on oxidic components also novel device concepts using carbon based materials (**Topic 1 and Topic 3**) are investigated in the PGI-7. Here the focus is on the design and efficiency of intermolecular contacts and of organic-metal interfaces, since one critical point with respect to device performance is the effective charge transfer between molecular components as well as between the molecular layer and the metal electrodes.

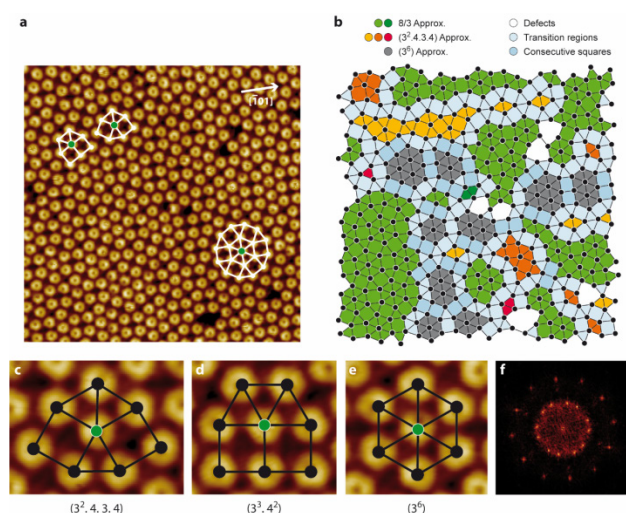


Fig. 6: Dodecagonal square-triangle tiling, as measured by STM. (a) High-resolution UHV-STM image of C_{60} on Pt-terminated $Pt_3Ti(111)$ (image size: $20 \times 20 \text{ nm}^2$, $U_{set} = -2.03 \text{ V}$, $I_{set} = 0.47 \text{ nA}$, 77 K). One dodecagon and the local structures reproduced in c) and d) are indicated in white. (b) Square-triangle tiling extracted from a), with color-coded decomposition into different types of approximants. (c)-(e), Three types of local vertex configurations around one fullerene (marked green): (c) $(3^2.4.3.4)$, (d) $(3^3.4^2)$, (e) (3^6) . (f) FFT of the STM image.

The LT-STM images with highly resolved molecular orbital structure allow a detailed assignment of the C_{60} adsorption geometry with respect to the underlying substrate, whereas STS reveals the electronic molecular-metal interaction. Most interestingly, in the case of a Pt_3Ti -single crystal alloy the surface termination, which is correlated to a certain catalytic activity, can be controlled by the preparation procedure [1]. Using this substrate third layer Ti-atoms provoke an adsorption energy landscape of the single crystal alloy so that the C_{60} /alloy interfacial interactions result in the creation of a two-dimensional dodecagonal quasi crystal, which can be described in terms of a square-triangle tiling (Fig. 6) [2]. These results are the basis for a controlled synthesis of a wide variety of quasicrystalline structures whose physical properties are tunable by the choice of substrate or adsorbed molecules.

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International Cooperation of PGI-7: R. De Souza (IPC, RWTH Aachen University); C. Jungemann (ITHE, RWTH Aachen University); U. Simon (IAC, RWTH Aachen University); Kirill Monakhov (IAC, RWTH Aachen); T. J. J. Müller (HHU Düsseldorf); R. Tetzlaff (TU Dresden); W.M.M. Kessels (Eindhoven University, Netherlands); A. Devi (Ruhr-University Bochum); Xavier Lopez (URV Tarragona); U. Celano (IMEC, Belgium); D. Cooper (CEA-LETI, Grenoble, France); D. Mc Laren (University Glasgow, UK); F. Borgatti (CNR Bologna, Italy); P. Dimitrakakis (NRSC Demokritos, Athen, Griechenland); Z. Klusek (University Lodz, Poland); J. Szade (University Katowice); V. Matolin (Charles University Prague, Czech Republic); M. Ritala, M. Leskelae (University of Helsinki, Finland); N. Pryds (DTU, Roskilde, Danmark); H. Hwang (Stanford University); Wei Lu (University Michigan, USA); B. Yildiz (MIT, Boston, USA); M. Shim (University of Illinois, Urbana, USA); M. Lanza (Soochow University, Suzhou, China); D.-Y. Cho (Chonbuk Univeristy, Korea); C.T. Hasegawa (Waseda University, Tokio, Japan); S. Hwang (Seoul National University, Korea); A. Chattopadhyay (NTU Singapore)

Planned New Directions

In line with the Helmholtz strategy in the rearch field “Information”, we are striving towards the application of memristives devices for novel types of computing such as energy-efficient memory or logic devices and brain-inspired computing. PGI-7 is furthermore involved in a multidisciplinary DFG excellence initiative “Neuro-inspired computing” within a large number of groups from RWTH Aachen University and FZ-Jülich. One prerequisite for these activities is that

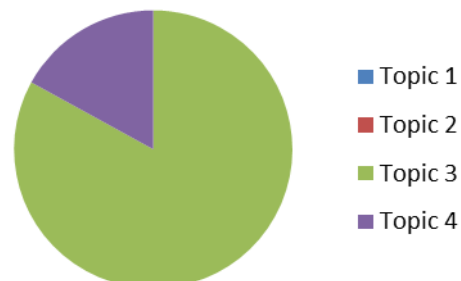
we will also further intensify our basic understanding of different types of memristive systems. In particular, the following activities will be started, or have been initiated quite recently:

- We will focus on the physics and chemistry of processes limiting the reliability of ReRAM cells (in particular endurance and retention). We will strive for a microscopic understanding of these processes by detailed studies of the electrical properties complemented by *in operando* analysis techniques and fundamental studies of the surface redox-behaviour. These activities will be accompanied by extending our simulation models in order to describe the observed phenomena accurately. Based on the experimental findings and device simulation, we develop strategies for a rational design of memristive devices with tailored properties for memory, logic and neuromorphic computing applications.
- We will investigate strategies to explore multilevel switching, the non-linearity of the switching kinetics as well as the stochasticity of memristive devices in artificial synapses of neuromorphic circuits. We aim at the demonstration of synaptic learning rules and the realization of neuromorphic concepts based on associative networks with binary inputs and outputs. This research will be conducted in close cooperation with T. Noll from PGI-10 and with M. Diesmann from IMN-6.
- One of the future challenges is the high density integration of ReRAM cells into three dimensional arrays. We will explore vertical concepts (VRRAM) which bear the greatest potential but require appropriate materials' selection and a deeper understanding and control of interface reactions. Furthermore, we will address polarity independent selector elements required for the highest integration in passive cross-bar arrays.
- In order to strive towards the ultimate scaling limits we will address the memristive properties of redox active molecules, such as donor-acceptor dyads and polyoxovanadate clusters. Furthermore, we will explore the potential of redox active molecules to build up neuromorphic devices.

2.4.8 PGI - Bioelectronics (PGI-8 / ICS-8)

Head of Institute: Prof. Dr. Andreas Offenhäusser

The PGI-8 Institute also participates in the Research Program BioSoft as ICS-8 (see Part III: Program BioSoft). Both these institute parts are presented and reviewed in the ICS-review.



PGI-8 mainly contributes to Topics 3 and 4.

2.4.9 PGI - Semiconductor Nanoelectronics (PGI-9)

Head of Institute: Prof. Dr. Detlev Grützmacher

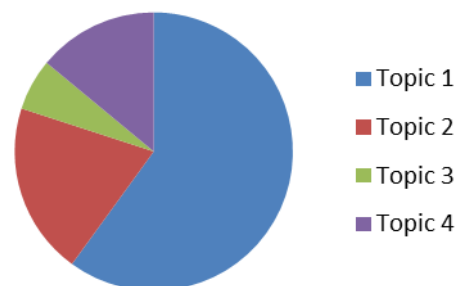
Contributing Personnel (31 Dec 2016 in FTE):

Core-funded: 18 scientists, 7 doctoral students, 12 scientific support personnel

Third-party funded: 3 scientists, 14 doctoral students, 2 scientific support personnel

Contributing Principal Investigators: Dan Buca, Hilde Hardtdegen, Larissa Juschkina, Beata Kardynal, Siegfried Mantl, Alexander Pawlis, Thomas Schäpers, Jürgen Schubert, Qing-Tai Zhao.

The research activities of the Peter Grünberg Institute “**Semiconductor Nanoelectronics**” are based on its competence in semiconductor heterostructure and hybrid nanostructure research, both in fundamental and device physics as well as in material and process development. They address four major fields. (1) Si based technology: Preparation of group IV alloys, in particular SiGeSn/GeSn hetero- and multiple quantum wells structures for Si photonics and ultra-low power, high speed device concepts. (2) III-V semiconductor as well as semiconductor/superconductor hybrid nanowires: exploring spintronic applications, helical transport and transmon qubits. (3) Hybrid nanostructures composed of topological insulators and superconductors: Exploring Majorana physics with the goal to demonstrate a qubit-based on braiding of Majorana fermions. (4) Nanostructures for quantum photonics: Single photon devices and quantum repeater for secure data transfer and quantum networks. A smaller activity concentrates on the deposition of the phase change material GeSbTe by MOVPE. This research topic is embedded in the SFB 917 nanoswitches. Moreover, the above mentioned activities are supported by exploring the opportunities of EUV light for lithography and element selective diagnostics on the nanoscale.



PGI-9 mainly contributes to Topic 1.

The research portfolio of PGI-9 contributes to all 4 topics of the program Future Information Technology (FIT) within PoF III, putting special emphasis on controlling charge based (Topic 1) and spin based (Topic 3) phenomena. The activities on topological insulators are in close collaboration with PGI-1, PGI-3, PGI-5 and PGI-6. Furthermore the Virtual Institute for Topological Insulators (VITI) fosters tight collaborations with RWTH Aachen, University of Würzburg and the Shanghai Institute of Microsystem and Information Technology (SIMIT).

It is noteworthy to point out that PGI-9 was and is still active in numerous research projects funded by the DFG, BMBF, EU and VW-Stiftung, which allowed to build up a network of research partners in universities, governmental institutes and industrial entities on national and international level. The following pages summarize selected highlights of the research performed over the last 5 years by PGI-9 in close collaboration with its research partners.

Group IV alloys as a versatile platform for electronic photonic integrated circuitry (EPIC)

Group IV alloys provide a platform for combination of silicon photonics and CMOS in which the active devices are fabricated based on alloys of group IV elements. Here we focus on SiGe, SiSn, GeSn and SiGeSn. The Sn-based epitaxy activity at PGI started 2013 and using a patented growth methodology [Patent ref.: DE 10 2014 108 352 A1, WO 2015/189004 A1, US 2017/0121845 A1, CN 106414816 A, EP 3 155 145 A1] was in short time able to grow high

crystalline quality GeSn alloys with high Sn content of about 14% [1], high above the Sn solubility limit of 1%. The detailed and temperature dependence photoluminescence (PL) allowed to map the transition from an indirect to a direct semiconductor of GeSn alloys [2, 3] and gave valuable input to bandstructure calculations of this artificial material. The direct bandgap, low effective masses for electrons and holes as well as the compatibility to Si technology makes this material a strong contender for Si based EPICs, combining monolithically near to mid-IR electrooptical devices and ultra low power electronics.

Group IV photonics (GFP): PGI-9 activities span from fundamental research on epitaxial growth of advanced GeSn/SiGeSn heterostructures accompanied by advance characterization to applied studies on photonic components - such as the laser, photo-detectors and integrated waveguides - aiming at specific applications. The latter are in collaboration with partner institutions and SMEs from Germany and EU.

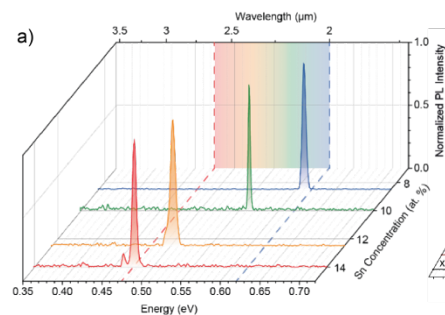


Fig. 1: a) Spectral lasing emission tuning in GeSn Fabry-Perot cavities.

The immediate demonstration of the first optically pumped laser action in GeSn alloys [2] was completed by the study of laser emission tuning by the Sn content in the alloys. The initial prediction of the indirect-to-direct transition based on PL and modelling was additionally proved by laser emission. In Fig. 1 lasing spectra of different GeSn layers shows lasing emission at 20 K from Fabry-Perot cavities, tunable in wavelength by Sn incorporation. This study showed that lasing emission can be obtained as soon as the energy of the direct Γ -valley becomes equal or smaller than the energy of the L-valleys. The Γ -L energy difference is zero for the 8.5% Sn alloy and of about -50 meV for the 12.5% Sn alloy. The maximum operation temperature of the laser, depends on this energy difference, being 60 K for 8.5% and 140 K for 12.5% Sn [4].

A room temperature operation of efficient electrically pumped optical devices requires the development of double hetero- (DHS) and multiple-quantum-well (MQW) SiGeSn/GeSn structures, in which the carriers can be confined in the GeSn active well and effectively contribute to radiative emission [5,6]. LED emission at injection currents below 1 A/cm² has been demonstrated at low T and at about 30 A/cm² at 300 K, as shown in Fig. 2a. The LED studies proved the necessity of SiGeSn barriers, in order to assure a type I band alignment in the heterostructure. Moreover, the complex development of an epitaxial process for the growth of SiGeSn alloys with high (Si, Sn) concentrations led to the first formation of direct bandgap GeSn/SiGeSn MQW. Detailed adjustments of the Sn content in the GeSn buffer and the MQW active region enables dislocation and bandgap engineering. The efforts gave rise to a substantial reduction of the lasing threshold and an increase in the operation temperature of optically pumped laser.

Microdisks cavities were built on both DHS and MQW structures and optically pumped lasing was studied looking especially at the laser threshold. The use of MQW leads to a decrease of the threshold pump power by a factor of 8 -10 compared with SiGeSn/GeSn DHS. A high resolution spectrum of a MQW microdisk laser and the lasing threshold evolution with temperature is shown in Fig. 2c and 2d.

The exciting optical properties of GeSn draw attention from many industrial and scientific entities, several close collaborations have been started to analyse the material properties and develop applications towards Si photonics. These partners are: Dr. Christian Forchel (nanoplus), Dr. Alexandra Enuica, (OPTOELECTRONICA- 2001 S.A), Dr. Jean Michel

Hartmann, Dr. Alexei Tchelnokov (CEA-LETI), Prof. Thomas Schröder, Prof. Giovanni Capellini (IHP Frankfurt Oder), Dr. Hans Sigg, (PSI, Villigen); Marianna Brais (INOE 2000), Dr. Zoran Ikonc (Univ. of Leeds), Prof. Douglas Paul (Univ. of Glasgow), Prof. Professor Goran Mashanovich (Univ. of Southampton), Prof. Jörg Schultze (Univ. Stuttgart), Prof. Stefano Chiusi (Univ. of Vigo), Prof. Dr. Philippe Boucaud (Univ. Paris-Sud), Dr. Toma Stoica, Magdalena Ciurea (NIMP, Bucharest), Prof. Shigeaki Zaima, Prof. Osamu Nakatsuka (Univ. of Nagoya), Prof. Shinichi Takagi, Mitsuru Takenaka (Tokyo Univ.), Prof. Nakashima (Kyushu Univ.)

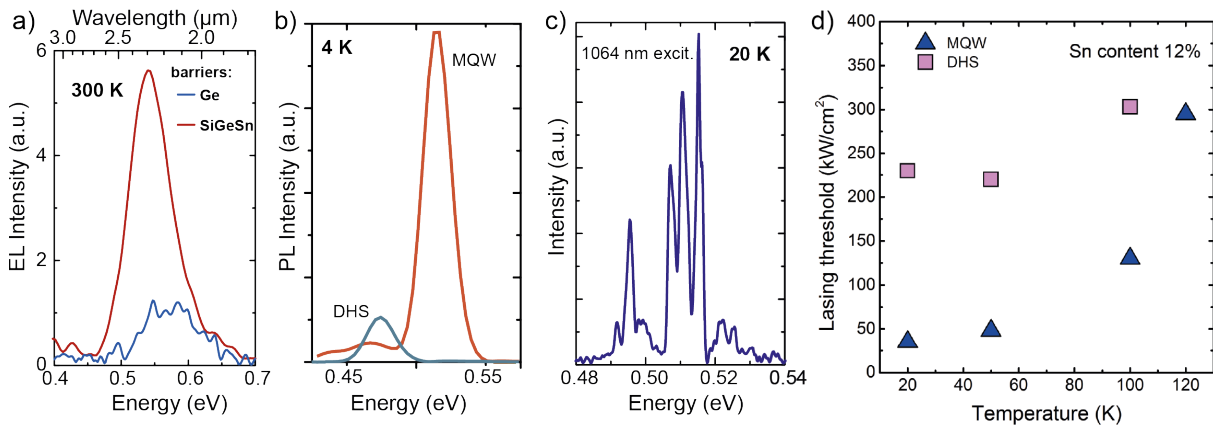


Fig. 2: (a) Comparison of GeSn MQW LED emission employing Ge and SiGeSn barriers. b) PL emission between DHS and MQW layers. c) High resolution spectrum of a MQW microdisk laser. d) Temperature dependence of lasing threshold for DHS and MQW laser structures.

Group IV Electronics: The activities of PGI 9 for electronics mainly focus on ultra low power devices with steep slope transistors like tunnel FETs (TFETs) and negative capacitance FETs (NC FETs). The band-to-band tunnelling current and the inverse subthreshold swing (SS) for a TFET depends on the properties of the semiconductor material, device geometry, tunnelling junction and the density of traps. Collaborated with EU “E2SWITCH” Project partners and BMBF Project “UltraLowPower” partners, we focus mainly on Group IV semiconductor TFETs

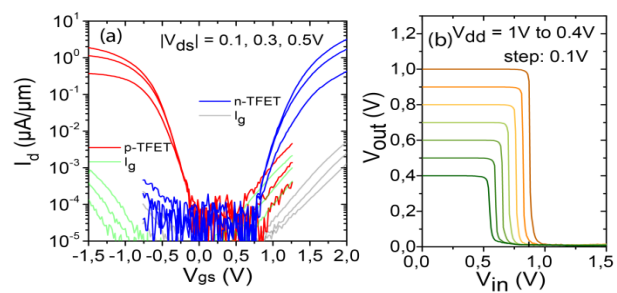
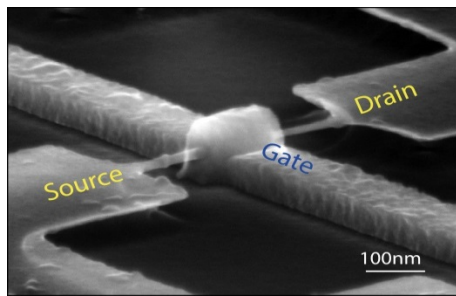


Fig. 3 GAA Si nanowire TFET structure with Si nanowire diameter <10nm. The complementary TFETs showed almost symmetric performance, allowing sharp transition of TFET inverters with good noise margin.

which offer flexible and compatible designs with Si CMOS technology. Comparing Si TFETs with single and triple gates as well as gate-all-around (GAA) geometries the improved electrostatics of Si nanowires (10 nm diameter) with GAA geometry manifests in the increased tunnelling currents and decreased subthreshold slope (Fig. 3). Using NiSi_2 at source/drain contacts and strained Si nanowire TFETs with a $\text{SS} < 60 \text{ mV/dec}$ at room temperature were achieved. For the first time an inverter and a NAND logic based on TFETs were operated at low voltages of 0.2V [7].

SiGe and GeSn alloys offering a smaller and in case of GeSn direct band gap will increase device performance, increasing tunnel currents and suppressing ambipolar switching. [8, Patent PCT/DE2015/000531]. In particular, TFETs with a direct bandgap (GeSn) gain direct tunneling

and thus high currents. Negative differential resistance was experimentally observed in GeSn p-i-n diodes, demonstrating high direct tunneling probabilities in GeSn materials [9]. The study of Ge/GeSn TFETs in collaboration with Notre Dame University in US showed that high performance TFETs can be achieved from lower defect density GeSn materials.

Negative capacitance MOSFET (NC FETs) in which a ferroelectric layer is connected in series with a normal MOSFET is another concept for steep slope devices. Here, the ferroelectric capacitor can work as negative capacitor and amplify the gate voltage. NC FETs with doped HfO_2 as ferroelectric layer in combination with FDSOI and nanowire FETs are studied in collaboration with Prof. Thomas Mikolajick at the NamLab (Dresden).

During the course of the research on group IV nanoelectronics the PGI-9 collaborated with the following partners: Thomas Mikolajick (NamLab), Dr. Heike Riel, Dr. Kirsten Moselund (IBM Zürich), Prof. Adrian Ionescu (EPFL), Prof. Giorgio Baccarani (Bologna University), Prof. Elena Gnani (Bologna University), Prof. David Esseni (Udine University), Dr. Jean-Michel Hartmann (Leti), Prof. Ru Huang (Peking University), Prof. Jyi-Tsong Lin (National Sun Yat Sen University), Prof. Andreas Schenk (ETH Zürich), Prof. Luca Selmi (Udine University), Prof. Lars-Erik Wernersson (Lund University), Prof. Dr. Yuehui Yu (SIMIT, China), Prof. Shinichi Takagi (Tokyo University), Prof. J.-P. Raskin (Univ. Catholique de Louvain).

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Quantum Phenomena of the Electron Transport in III-V nanowires

Semiconductor nanowires are very versatile objects to study fundamental quantum effects and are considered as building blocks for applications in quantum information. InAs and InSb nanowires are in the focus of interest because of high electron mobilities and g-factors. In InAs nanowires phase-coherent transport was investigated at low temperatures. From weak antilocalization measurements information on spin-orbit coupling was gained. In a joint project with a theory group at Regensburg University (Paul Wenk, John Schiemann), the strength of

the Rashba as well as the Dresselhaus contribution was extracted. In InAs nanowires with very low background doping ballistic transport was observed in samples comprising a set of top gate fingers [1]. The conductance as a function of gate voltage revealed quantized conductance (Fig. 4a). From measurements at various magnetic fields the g-factor was determined for the different conductance channels. Furthermore, in quantum point contacts connected in series, adiabatic transport was found, i.e., the overall conductance was governed by the constriction with the lowest conductance. In a regime with two almost pinched-off quantum point contacts Aharonov-Bohm type oscillations were observed, which were attributed transport in closed-loop snake-orbits. A closer inspection of the first quantized conductance step revealed a re-entrant feature, e.g., a conductance dip [2]. This phenomena could be explained in the framework of spin-helical transport, an important prerequisite for the realization of Majorana fermions in semiconductor nanowires. Majorana fermions are of major interest because of potential applications in fault-tolerant topological quantum computing. A dip feature was observed even at zero magnetic fields. In collaboration with the theory group of Prof. Björn Trauzettel at Würzburg University, within the framework of the Helmholtz Virtual Institute for Topological Insulators, this feature could be explained by spin-flipping two-particle backscattering. In order to perform braiding schemes in topological quantum computation, nanowire networks are required. By epitaxial growth on textured substrates we were able to realize nanowire networks of different complexity (Fig. 4b) [3].

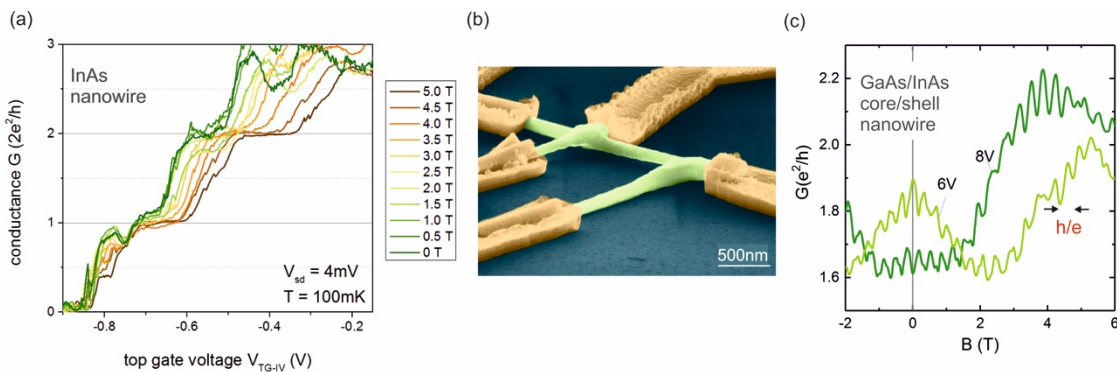


Fig. 4: (a) Quantized conductance in InAs with a dip feature at first plateau. (b) Contacted InAs nanowire network (c) Flux periodic conductance oscillations in GaAs/InAs core/shell nanowires.

By employing heterostructure nanowires, their transport properties can be tailored. GaAs/InAs core/shell nanowires, where a conductive InAs shell is wrapped around an insulating GaAs core [4] were subject of detailed investigations. In an external magnetic field along the nanowire axis, h/e magnetic flux periodic conductance oscillations were observed (Fig. 4c). The oscillations could be explained in the framework of closed-loop angular momentum states. The conduction oscillations were found to be very robust, being resolved even at temperatures as high as 50 K. By employing an advanced alignment scheme Hall contacts at the side facets of these nanowires were prepared [5] and the carrier concentration and the mobility could be extracted. Furthermore, in a magnetic field aligned parallel to the nanowire axis Aharonov-Bohm oscillations were detected using these Hall contacts. In core/shell nanowires contacted with superconducting electrodes, giant conductance oscillations with half-period compared to the case of normal metal contacts were found [6]. The origin of these oscillations was attributed to phase-coherent Andreev reflection in connection with reflectionless tunnelling. In a joint project with Prof. Martin Weides (Karlsruhe Institute of Technology), nanowire-based Josephson junctions are employed for transmon qubit applications.

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Signatures of Majorana Bound States in Topological Insulators

Triggered by the promise of dissipationless surface transport, three-dimensional topological insulators (TIs) attracted a lot of attention in applied as well as in basic physics. The current-carrying surface states are topologically protected by time reversal symmetry, i.e., the spin orientation is locked to the direction of motion. This specific property makes TIs particularly interesting for spintronics as well as for quantum computation. Typical three-dimensional TI materials, namely Bi_2Te_3 , Sb_2Te_3 , and Bi_2Se_3 , were grown by molecular beam epitaxy (MBE).

TI/superconductor (SC) hybrid devices are predicted to be a suitable platform for the detection of Majorana excitations (MEs) and the realization of topological quantum bits (qubits). In order to verify the existence of MEs topological Josephson devices have been measured at cryogenic temperatures. In order to resolve the response from so called Majorana Bound States (MBSs), which result from two opposing MEs at the two SC/TI interfaces, from conventional Andreev

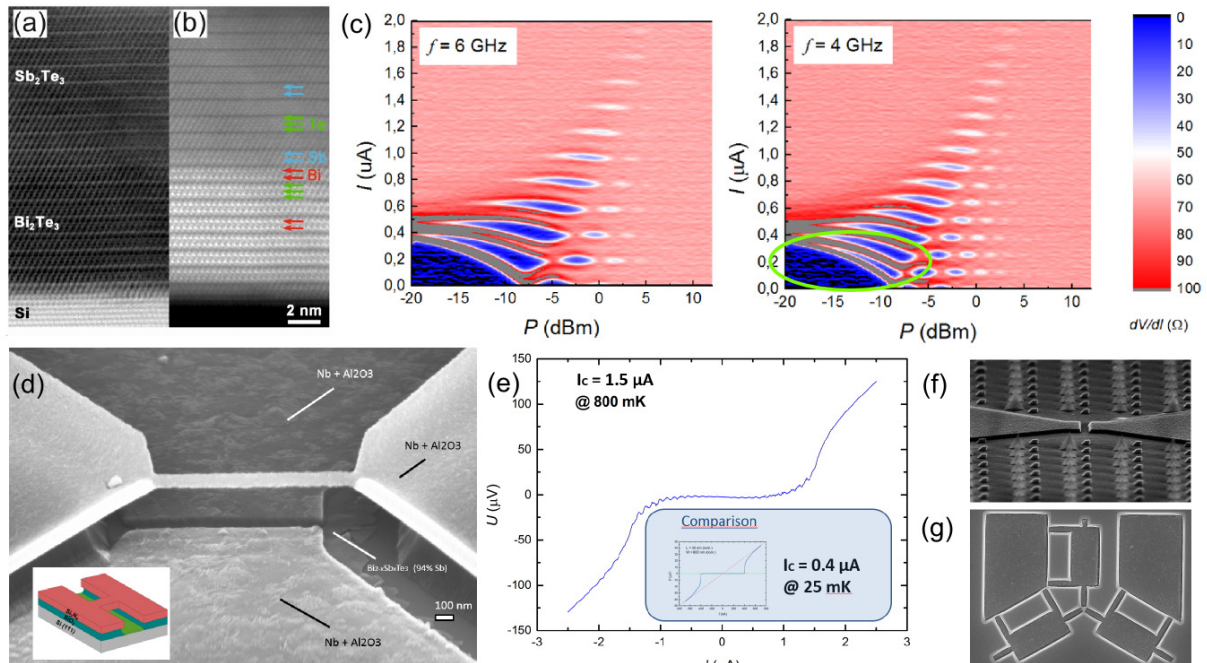


Fig. 5: TEM images of a $\text{n-Bi}_2\text{Te}_3/\text{p-Sb}_2\text{Te}_3$ heterostructure (a,b). Shapiro step response of a topological Josephson junction for 6 GHz and 4 GHz (c). In-situ grown TI/SC junction by means of the Si_3N_4 nanobridges (d). Current-voltage characteristics of in-situ grown TI/SC junction improving the critical current (ex-situ grown TI/SC junction depicted in the inset (e)). In-situ grown superconducting electrodes after lifting of Si_3N_4 mask (f). Structured superconducting thin film in the form of a tri-junction with attached SQUIDs on top of $(\text{Bi}_{0.06}\text{Sb}_{0.94})_2\text{Te}_3$. The stencil mask is still in contact with the substrate. The shown layout has been proposed for demonstrating braiding statistics of Majorana Zero Modes.

Bound States (ABSs) the Fermi level (E_F) of the TI has to be in close proximity to the Dirac point (DP). One severe problem here is related to the aforementioned MBE grown TI materials as they are intrinsically doped. Bi_2Te_3 is intrinsically n-type doped, whereas Sb_2Te_3 is usually p-type doped. In order to compensate the n- and p-type doping p-n junctions of n-type Bi_2Te_3 and p-type Sb_2Te_3 (Fig. 5 a, b) [1] as well as $(\text{Sb}_x\text{Bi}_{1-x})_2\text{Te}_3$ ternary alloys with reduced bulk carrier concentration have been realized.

Attenuated odd number Shapiro steps are first signatures of MBSs across topological Josephson devices with $(\text{Sb}_{0.94}\text{Bi}_{0.06})_2\text{Te}_3$ interlayer and an in-situ applied aluminum oxide capping. We found an attenuated first Shapiro step in our devices, when exiting the junction with the characteristic frequency of 4 GHz (green ellipse in Fig. 5 c). For higher frequencies (e.g., 6 GHz) this feature disappears. To increase the Shapiro response in TI/SC Josephson devices a high quality interface between the TI and the SC has been established by using an all in-situ process for the growth of TI/SC Josephson devices, which utilize niobium SC contacts and a stoichiometric Al_2O_3 capping layer (Fig. 5c). Therefore defined Si_3N_4 nanobridges serve as shadow mask for the deposition of SC contacts, which are in close proximity (300nm) to the silicon surface. First transport experiments at cryogenic temperatures revealed a critical current of 1.5 μA , which is a fourfold increase compared to conventionally deposited SC contacts by ex-situ processing measured before (Fig. 5e). We have developed a way to gently remove the Si_3N_4 nanobridges after growth of TI and SC (Fig. 5f). This is especially important as soon as the devices are getting more complex (Fig. 5g). Future experiments will employ the combination of pre-defined nanotrenches for selective growth of TI nanowires as well as nanobridges for the in-situ deposition of SC contacts for realization of scalable and high-throughput TI/SC hybrid devices. These include high quality Josephson devices for further Shapiro response measurements as well as first topological qubit designs.

The science on TI has been performed within the virtual Institute of topological insulators (VITI) teaming up with the Univ. of Würzburg (Hartmut Buhmann), the RWTH Aachen (Christoph Stampfer) and the SIMIT, Shanghai (Shumin Wang) as well as collaboration partners at the Univ. of Twente (Alexander Brinkmann) and Regensburg (Sergey Ganichev).

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New Materials for Quantum Photonics

Activities in quantum photonics based on two-dimensional materials started with a completion of the optical laboratory in the second half of 2014. We investigate direct band gap semiconducting monolayers (MLs) of transition metal dichalcogenides (TMDs) with the chemical formula $(\text{Mo,W})(\text{S,Se})_2$ for non-classical light sources. The heavy metal ions of the lattice, weak Coulomb screening and the lack of inversion symmetry in ML-TMDs result in a unique combination of giant exciton binding energies (exceeding 200meV), large spin-orbit coupling, as well as the optically accessible locked spin - valley degrees of freedom at the inequivalent K-valleys. This makes ML-TMDs very interesting for single and entangled photon sources or spin-photon entanglement sources. Reported single photon emission (SPE) was not reproducible and did not preserve the band structure of the ML_TMDs. We develop

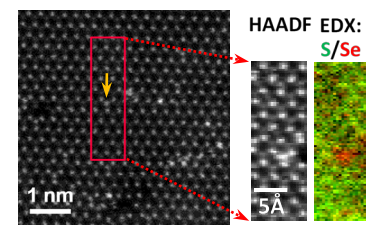


Fig. 6: Scanning transmission electron microscopy image of s-Se implanted MoS_2 . Yellow arrow marks a position of an implanted Se atom.

methods to confine excitons in a controlled way in potentials that preserve spin-valley physics and can be used for practical devices. We explore two methods: ion implantation and the modulation of the electrostatic screening. Ion implantation into 2-D materials requires ion energies below 50eV and we collaborate with University of Göttingen, where implantation at such energies is uniquely possible. Initially, we chose to study the process of implantation with ^{80}Se ions and MoS_2 target. Clear photoluminescence from trions was measured with up to 1-2% of S sub-lattice replaced with Se when using 10 and 20eV ion energy. We found that lower ion energies required higher ion doses and resulted in a higher density of S-vacancies. Our optical studies of the implanted materials (micro-Raman, micro-photoluminescence based spectroscopies) are supported by structural and chemical characterisation using scanning transmission electron microscopy (in collaboration with Univ. of Limerick in Ireland, see Fig. 6).

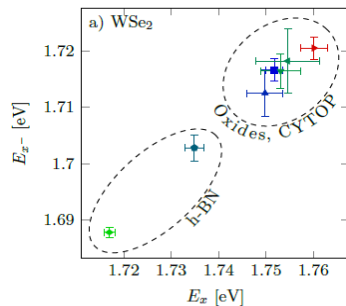


Fig. 7: Energies of exciton (E_x) and trion (E_{x-}) for WSe_2 on different substrates. The lowest energies (green point) are measured for WSe_2 embedded in h-BN. To be published.

hydrophobic h-BN, amorphous fluoropolymers or SiO_2 can define quantum dots by shifting the exciton energy by 40 meV (Fig. 7) and we are working on methods to pattern suitable substrates.

Optically active devices based on $\text{ZnMgSe}/\text{ZnSe}/\text{CdSe}$ heterostructures are in principle capable to cover the whole visible spectrum via a variation of the dimensions. However, the large lattice mismatch between ZnSe and limits the thickness of coherently grown CdSe layers on ZnSe to ~ 2 monolayers (MLs). Plastic relaxation takes place for larger thicknesses and leads to dominant non-radiative recombination in the devices. We developed a dedicated strain compensation technique involving alternately strained ZnSe and CdSe layers on InGaAs [2]. Then, the ZnSe is tensile and the CdSe compressively strained. Taking into account a specific thickness ratio between the ZnSe and the CdSe layers, this leads to an overall minimised strain energy in the heterostructure and enabled us to fabricate high quality CdSe QWs with a thickness of 1-6 MLs. Figure 8a) shows high-resolution transmission electron microscope (TEM) images of the cross-section of such a CdSe/ZnSe QW indicating the obtained high-quality interfaces between the layers. Detailed TEM investigations, strain analysis and corresponding molecular statics simulations verified that ZnSe/CdSe QWs with CdSe thicknesses up to 5 MLs grow fully coherent due to the dedicated strain compensation technique. The underlying mechanism and our tailored theoretical

In addition, in collaboration with Ernst Ruska Centre at FZJ and support from PGI-1/IAS-1, we develop quantitative electron holography to study individual defects in ML-TMDs. We have shown that one can reach fully quantitative agreement between experiments and density functional theory based simulations or pristine materials [1]. SPE in semiconductors can be achieved from quantum dots. Those can be defined in ML-TMDs in laterally modulated dielectric environment since both the electronic and optical band gaps of ML-TMDs depend on dielectric properties of the cladding materials. Due to the short lifetime of the excitons and their small Bohr radius it is not obvious what frequency dielectric constant is relevant for the optical band gap so we have performed comprehensive studies of a variety of dielectrics in order to identify suitable ones. We found that very flat, chemically stable,

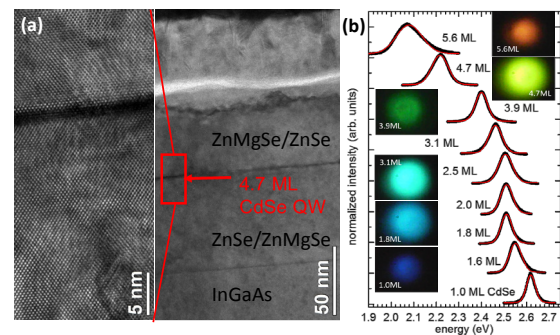


Fig. 8: (a) Cross.sction TEM of a strain-compensated $\text{ZnSe}/\text{CdSe}/\text{ZnSe}$ quantum structure. (b) Room-temperature PL spectra versus CdSe thickness.

modelling can be easily applied to other highly lattice mismatched semiconductor heterostructures. Our innovative strain-compensation technique allows us to tune the photoluminescence (PL) emission from CdSe QWs in a large spectral range between 450-550 nm wavelength. Figure 8b) shows room-temperature PL spectra of selected CdSe/ZnSe QWs covering a large range of the visible spectrum. High quantum efficiency of the PL was achieved even at room temperature. Moreover we fabricated sophisticated microdisk resonators from the CdSe QWs [3]. These results reveal the potential of our nanostructures for high-brightness light emitters or lasers covering the whole visible spectrum.

The research is supported by a transregio SFB as well the Volkswagenstiftung involving several academic partners: Prof. Manfred Bayer, Dmitri Yakovlev (TU Dortmund), Prof. Jürgen Gutowski (Univ. Bremen) Prof. Andreas Wieck (Ruhr Univ. Bochum), Prof. Dirk Reuter, Prof. Donat As (Univ. Paderborn)

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MOVPE of Ge₁Sb₂Te₄ layers

Phase change materials are currently under investigation for memory applications based on resistive switching. The switching is induced by crystallization and amorphization processes. Recently, a new field induced switching mechanism was proposed for monocrystalline compound memories consisting of rhombohedral Sb₂Te₃ and GeTe multi-layer structures - so-called interfacial phase change memories (IPCM). Here, switching consumes only a fraction of the energy needed for melting and recrystallization.

Lately, we were able to deposit trigonal Ge₁Sb₂Te₄ layers epitaxially by the industrially relevant method MOVPE on Si (111) substrates. First, precursors with matching stability were searched for and subsequently the growth conditions were optimized. It was found that a sufficiently low reactor pressure (50 hPa) and the exact control of the growth temperature (475°C) are the key to epitaxial layers of one composition [1]. The structural properties of the epitaxial layers were characterized in detail and the similarities to IPCM structures were disclosed (Fig.9) [2]. The alloy has therefore great promise for low energy non-volatile memory applications. The research is performed within the framework of the SFB 917 nanoswitches in close collaboration with Prof. Matthias Wuttig (RWTH).

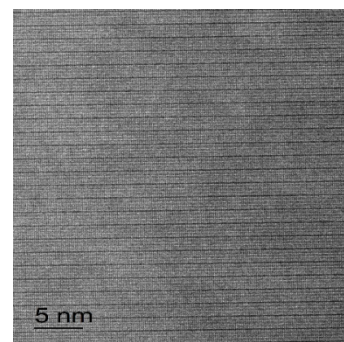


Fig. 9: HR-TEM of trigonal epitaxial Ge₁Sb₂Te₄ layers. Alternating cation / anion 7 - layer stacks are visible which are separated by v.d. Waals gaps (dark lines) [7].

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Development of a "laboratory synchrotron" for PES and PEEM

The method of chemical surface analysis, which is very widespread in basic and applied research, is photoelectron spectroscopy (PES). A further development of this technique, the

photoemission electron microscopy (PEEM), allows for chemically resolved analysis of the microscopic surface structure. PES and PEEM instruments require a light source in the ultra-ultraviolet (EUV) and X-ray spectral range, which is intense and tunable in the photon energy and has a high monochromaticity. In a proof-of-principle experiment, we combined a system consisting of a compact, very intense plasma-based EUV light source and simple EUV optics with a PEEM and demonstrated the general feasibility of a "laboratory synchrotron" for PES and PEEM. For the proof-of-principle experiment, we used an oxygen plasma. The spectral line of O^{5+} was used with a photon energy of 71.7 eV. With this design, we successfully demonstrated the following PES / PEEM operating modes: Photoemission electron microscopy for structural analysis without energy filtering of the electrons, photoelectron spectroscopy for element determination, energy-filtered photoemission electron microscopy for the combination of structural analysis and spatially resolved element determination, k-space mapping for determining the electronic band structure (Fig.10). The proof-of-principle experiment was conducted within the framework of a seed fund project collaborating with Prof. Tautz (PGI-3), Prof. Schneider (PGI-6) and Prof. Poprawe, Prof. Loosen (ILT, Fraunhofer) within the JARA. Here, the photoelectron-based spectro-microscopy has been applied to phase-change materials e.g., $Ge_2Sb_2Te_5$ (GST). The photoemission electron microscope combined with an energy analyzer is capable of imaging energy-filtered electrons, which enables spectro-microscopy in the first place. Additionally, the high surface sensitivity of the EUV light was used to study the surface oxidation on islands of the phase-change material $Ge_1Sb_2Te_4$. The plasma-based light source allows the extension of spectro-microscopy, previously only feasible at synchrotron beamlines, to laboratory-based work.

The development of intense plasma based EUV sources open pathways for multiple applications in analysis and microscopy as well as in fabrication technology, i.e., lithography (see report of PGI 10). Consequently, also this activity is well received by the academic and industry, reflected in multiple partnerships and collaborations, namely: Prof. Wim J. van der Zande, Prof. Wim M. J. Coene, Prof. Vadim Y. Banine, (ASML BV) Dr. Torsten Feigl (Optixfab GmbH), Dr. Yusuke Teramoto, (BLV Licht- und Vakuumtechnik GmbH), Prof. Mario C. Marconi, Prof. Carmen S. Menoni, Prof. Jorge J. Rocca, (Colorado State Univ.), Prof. Jianwei Miao (UCLA), Prof. William S. Brocklesby (Univ. of Southampton), Prof. Gerry O'Sullivan, Dr. Fergal O'Reilly (Univ. College Dublin), Prof. Piergiorgio Nicolosi (University of Padua), Prof. Konstantin Koshelev (Institute for

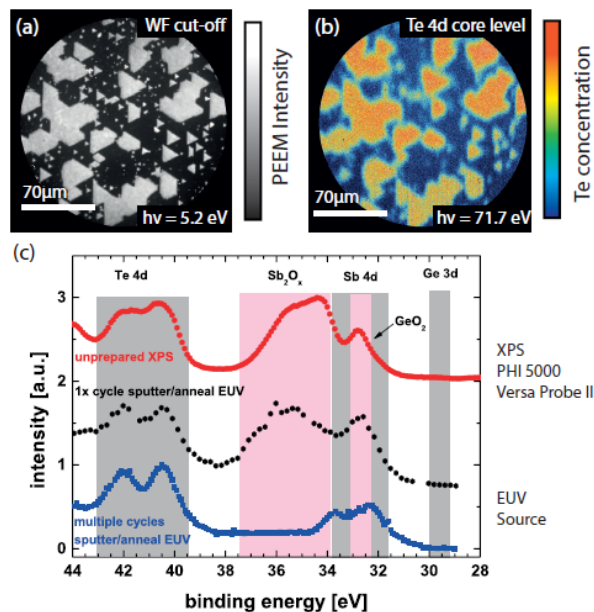


Fig. 10: EF-PEEM image of GST islands recorded at $h\nu=5.2$ eV (a) and at $h\nu=71.7$ eV (b) photon energy. Image (a) was acquired at the work function (WF) cut-off. Image (b) consists of integrated images around the Te 4d core level binding energies (39-41 eV). In panel (c), a binding energy spectrum of gently sputtered GST islands acquired with the EUV source (black) is compared to XPS reference data of a pristine sample (red). The oxide components of Ge and Sb are clearly visible. Additional preparation cycles lead to a purely metallic Sb/Te termination (blue).

Spectroscopy, Russian Academy of Sciences) Dr. Frank Scholze (PTB at BESSY).

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Future developments

The PGI 9 will foster the activity on group-IV alloys towards electrically pumped laser, by combining doping and contacting technology, as used for LEDs and the optimized MQW heterostructures. Moreover, PGI 9 develops in parallel, together with the SME Nanoplus GmbH, SiGeSn integrated detectors, as well as GeSn modulators with RWTH Aachen and University of Southampton. All this knowledge and components will be monolithically integrated on a Ge platform aiming for the realization of GeSn photonics chip. This is part of an ERC grant application together with RWTH Aachen and complemented by a DFG project "SiGeSn laser". The photonic activity also touches plasmonic studies looking for tuning of plasmon frequency by n-type doping of GeSn alloys in collaboration with Glasgow University. Recently, together with CEA LETI in Grenoble and the companies MirSense (France), Nanoplus GmbH (Germany) and Optoelectronica SA (Romania) PGI 9 as the leading partner has submitted a ICT 30 KET photonics EU H2020 project. The proposal targets the use of GeSn emitters and detectors in MIR gas sensing and skin medical treatment by MIR irradiation. Concerning electronic devices, we collaborate with CEA LETI for a new designs of SiGe/Si FETs. Moreover, using Si/SiGe/Si/SiGe(Sn) structures enables flexible design of tunnelling areas. Ferroelectric HfO₂ layer with different doping elements like Zr, Lu, Y, Gd etc. will be studied. Efforts will be put to match the negative capacitance to the MOS capacitance in order to achieve a steep slope (SS<60mV/dec) and to realize devices with steep SS but without hysteresis. Finally the research on Si-Ge-Sn alloys is within the core activities of JARA-FIT collaborating with Prof Witzens and Prof. Knoch (RWTH-Aachen) as well as PGI-5 and PGI-10, in particular as a partner in the Excellence Eluster proposal Neuro-inspired Computing (Neuro-IC) of the RWTH-Aachen submitted to the German Excellence Strategy.

It is planned to lift the research on topological insulators and quantum photonics to the next level within the proposed Excellence Cluster Matter and Light for Quantum Computing (ML4Q) in collaboration with the Universities of Aachen, Bonn and Cologne submitted to the German Excellence Strategy. Near to midterm goals are the improvement of the transparency of SC/TI contacts to obtain clear signatures of Majorana bound states and to demonstrate a qubit based on braiding of these states. The activities on spintronics and quantum photonics are embedded in the research strategy on quantum information in close cooperation with PGI-2 and PGI-11. Within the ML4Q cluster close collaboration with the University of Bonn are put in place to establish quantum optical links for qubit data transfer. Research on Josephson junction of SC and III/V nanowire will be employed in a joint project with Karlsruhe Institute of Technology, for transmon qubit applications. Strategically, the research of PGI-9 is embedded in the efforts for neuroinspired as well quantum computing within PGI and JARA-FIT as well as with neighboring universities. To this end PGI-9 plans to strengthen his efforts to be valuable partner in these complex and interdisciplinary areas of modern information technology.

2.4.10 PGI – Energy Efficient (PGI-10, JARA-Institute Green IT)

Head of Institute: Prof. Dr. Detlev Grützmacher (managing director), Prof. Dr. Rainer Waser, Prof. Dr. Matthias Wuttig, Prof. Dr. Tobias Noll, Prof. Dr. Jia Grace Lu

Contributing Personnel (31 Dec 2016 in FTE):

The JARA Institute was founded in 2016. Despite longstanding and ongoing collaborative work, the institute is still in the buildup phase and not all personnel have yet been formally assigned to the institute. Therefore no reasonable numbers can be given.

Contributing Principal Investigators: Arne Heitmann, Susanne Hoffmann Eiffert, Larissa Juschkin, Mihail Lepsa, Vikas Rana.

The research activities of the Peter Grünberg Institute “ **Energy-efficient information technology (Green IT)**” are centred around the innovative concepts enabling a drastic reduction of power consumption in future information circuitry. The ongoing increase in the volume of data and in consequence the increase in computer capacity requires innovation to improve the energy efficiency substantially. With the economy of scale running out of power, energy efficiency will be the technology driver of the future. It is envisioned that these concepts will be boosted by the discovery of new materials and material combinations, called hybrid structures, enabling memory based computing architectures. Combining those bio- or brain inspired neuromorphic chips with high rate data transmission provided by silicon photonics and large external memories may provide a versatile architecture for ultra-low energy computing. The science of PGI-10 is directed towards the development of building blocks for those systems in the near- to midterm timeline with the perspective to further integrate these building blocks in conceptual device circuitry in a longer time frame. These goals will be pursued in close collaboration with its national and international partners, such as the SNU/DTFL (Prof. C.S. Hwang) *Seoul National University*, the *University of Nagoya* (Prof. Zaima) and Dr. L. Perniola, *CEA-LETI, Grenoble*.

The NanoCluster tool

A state of the art multi-material cluster (NanoCluster) tool has been installed successfully and is operated by institutes from Forschungszentrum Jülich (PGI-10, PGI-9, PGI-7, PGI-6) and RWTH Aachen (IP 1A) in Jülich. The tool provides a unique combination of nine UHV epitaxial/deposition systems including MBE, ALD, and sputtering. It offers the possibility for *in-situ* growth of complex nanostructures from III-V and II-VI (ZnSe based) semiconductors, metals, oxides, magnetic and phase change materials. The *in-situ* growth can reduce significantly the interface density of states resulting in improved (opto-) electronic nanodevices and offering the chance to study new quantum phenomena. The cluster tool is potentially an ‘alembic’ for developing new materials and hybrid structures in ultra clean conditions.

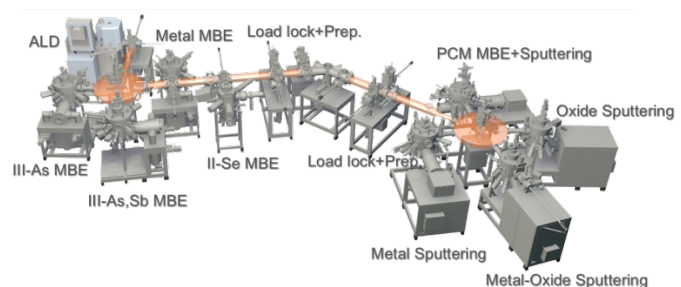


Fig. 1: CAD image of the NanoCluster

Due to UHV transfer enabled between the deposition systems, like for example off-axis metal and oxide sputtering and atomic layer deposition, this tool is utilized for RRAM research

focusing on interfacial effects of in-situ stacked sub 10 nm thin metal oxide and metal layers for concepts on three dimensional integrated RRAM.

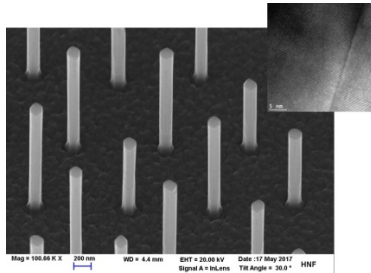


Fig. 2: InAs/GaSb core-shell NW array. The inset shows a HRTEM image of the InAs-GaSb interface.

growth/deposition processes which finally will permit us to get the suitable *in-situ* grown structures to reach our goals. Successful experiments have been carried out for the growth of Te doped InAs NWs. Good quality InAs/GaSb NW arrays have been grown with undoped and C doped GaSb shells. The deposition of TaN layer for metal gates has been studied and optimized in the metal MBE system attached to the cluster tool. Hybrid superconductor/semiconductor NW structures combining Al or Nb with InAs were in situ obtained for basic research of novel quantum phenomena, e.g., Majorana fermions, coupling of spin qubits, transmon-like devices, to this end the facility also supporting work pursued by PGI-11.

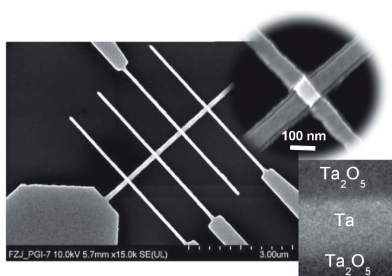


Fig. 3: SEM image of the 1×4 nanocrossbar array without access to the ME (structure B). The width of the BE varies from 40 nm to 100 nm, whereas the TE width was fixed to 80 nm. The as deposited Ta₂O₅ layers are amorphous.[3]

the implementation of memristive fuzzy logic gates in future. With highly reliable (10^6 cycles) and multi-level Pt/W/Ta₂O₅/Pt Resistive RAM devices, 3-bit modular arithmetic operation is performed. This will reduce the computational complexity by decreasing the number of needed digits for high radix number system. Thus the number of calculation operations in an addition and the number of logic devices can directly be reduced. Furthermore, these devices in crossbar arrays enable implementation of multi-parallel search algorithms for pattern recognition tasks, which are widely required for neuromorphic applications in a very efficient manner.

A further application of resistive switching cells is their use in associative capacitive networks (ACN). An ACN, as a content-addressable memory, is able to detect the Hamming distance between search and stored patterns. Such an ACN can be implemented by an architecture consisting of nanocrossbar array of complementary resistive switching (CRS) devices and using

Moreover, the growth of nanowire (NW) hetero- and hybrid-structures for the processing of TFETs is employed. Processing and characterization of devices from the grown structures complete the activity. In a first stage, we have successfully transferred our knowledge for the MBE growth of III-V heterostructures layers and (NWs) and deposition of high-k dielectrics using ALD. The potential of the new MBE systems have been demonstrated by the relative fast progress in the growth of high mobility GaAs/AlGaAs heterostructures and the growth control of single InAs NWs on SiO_x/Si (111) substrates processed with an original recipe. Step by step, we started to study the

Logic and memory based on memristive circuits

Prior to the foundation of the PGI-10, previous JARA-FIT activities in the field of on Green-, we explored the potential of memristive devices and memristive crossbar arrays for memory-intensive computing paradigms. The non-volatility of the devices enables logic-in-memory operations [1]. The logic operations are directly processed in the memory and arithmetic tasks, e.g., additions, are carried out within the array [2]. Thus, by blurring the boundaries between memory and arithmetic logic units the von-Neumann-bottleneck can be eliminated.

The realization of minimum and maximum gate function through resistive RAM devices can enable the implementation of memristive fuzzy logic gates in future. With highly reliable (10^6 cycles) and multi-level Pt/W/Ta₂O₅/Pt Resistive RAM devices, 3-bit modular arithmetic operation is performed. This will reduce the computational complexity by decreasing the number of needed digits for high radix number system. Thus the number of calculation operations in an addition and the number of logic devices can directly be reduced. Furthermore, these devices in crossbar arrays enable implementation of multi-parallel search algorithms for pattern recognition tasks, which are widely required for neuromorphic applications in a very efficient manner.

the nondestructive capacitive readout of the CRS device [4]. As compared with conventional CAMs the fully passive network gets along without MOSFETs leading to a significant area and energy efficiency. A technical realization of such an ACN has been demonstrated using preprogrammed test arrays [5].

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- [5] L. Nielen, A. Siemon, S. Tappertzhofen, **R. Waser, S. Menzel, E. Linn**, *Study of Memristive Associative Capacitive Networks for CAM Applications*, IEEE Journal on Emerging and Selected Topics in Circuits and Systems, vol. **5**, pp. 153–161 (2015) [doi: 10.1109/JETCAS.2015.2426491](https://doi.org/10.1109/JETCAS.2015.2426491)

Merging resistive switches and CMOS for functional circuits

The focus of research at EECS was given in the field of circuits based on resistive switches (RS) and nano-scaled CMOS devices. Particular application domains cover nonvolatile distributed memories, reconfigurable logic, and artificial neural networks. One of the most striking challenges in the design of such circuits is to tackle the significant impact of variability on circuit reliability, notably in consideration of reduced signal energies, speed requirements, and demands for minimal area occupation. In contrast to standard methods, focusing on functional aspects of systems a-priori, concerns about reliability were seen as a premise here while the realization of a particular function gets constrained by reliability demands. In order to develop effective means to preserve robustness against various physical sources of perturbations, the research program is based on three pillars: (i) Preparation of realistic models

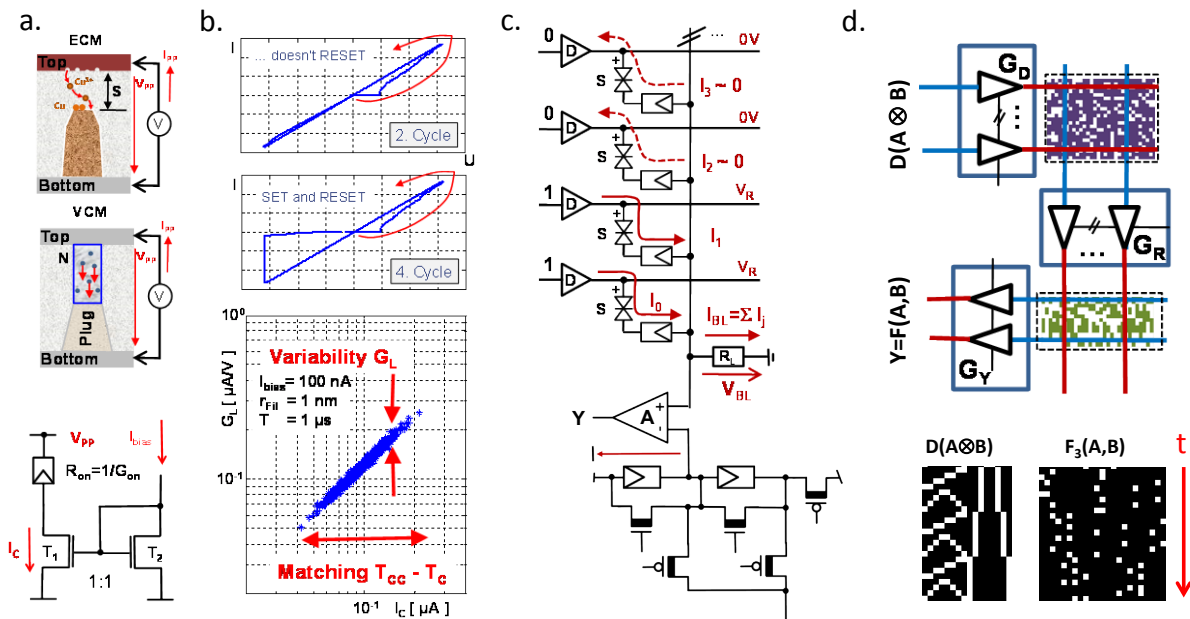


Fig. 4: a. Nano-scaled resistive switches and CMOS devices; b. variability of switching kinetics; c. detailed modeling of fundamental circuits with high fan-in; d. realization of complex logic functions by dense interconnect patterns, associative mapping of sparse activity codes.

of devices (RS, CMOS) and interconnect as well for circuit simulation. At this, typical influence factors are even by static parameter variability caused by process variability, random telegraph noise and electronic traps, random movement of charged ions, and cross-coupling of signals by electromagnetic fields. (ii) Identification of suitable functional circuits which could replace the gates used in standard CMOS technology. A particular focus was set on circuit concepts which allow for a significant increase in the gate fan-in (associative mapping of operands). Here, the degree of parallelism could be increased while the speed of individual gates could be lowered to meet particular reliability demands. In doing so, extensive Monte-Carlo simulations of fundamental circuits were carried out to characterize their performance in regard to power consumption, bit error rate, and speed. (iii) Identification of elementary passive and active measures to enhance the systems robustness dependent on the considered level (device, circuit, architecture, system). Concepts were evaluated which include feedback loops, non-volatile circuit calibration and adaptation as well as the use of particular error detecting codes.

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Method Development: EUV interference lithography

The research is focused on the investigation of resolution limits of interference and proximity lithography with EUV radiation. Various types of laboratory EUV sources were examined with regard to yielding the highest pattern density and quality. In particular, these are Xenon gas-discharge plasma source (broadband radiation near 11 nm wavelength), Ne-like Ar-plasma-based EUV laser (coherent narrow-band radiation at 46.9 nm), and high-harmonic generation (HHG) source (at 29 nm). Also different interference lithography approaches are implemented and investigated experimentally. These included reflective interference lithography with Lloyd's mirror, proximity lithography, and different types of Talbot lithography (see Fig. 5). [1-4] This is complemented by numerical simulations of optical wave propagation of EUV radiation to predict limits of the resolution and contrast for these approaches. General research objectives are analysis of optical characteristics of particular EUV sources with respect to the patterning capabilities, optimization of the optical design of interference lithography system for the use of EUV radiation, and understanding of limits of different methods.

Large-area arrays of complex nano-antennes using laboratory-based EUV proximity lithography were fabricated in cooperation with RWTH-TOS and RWTH-Infrared- Nano-Optics institutes. Precise control over the mask-wafer distance and the exposure dose increases the flexibility of

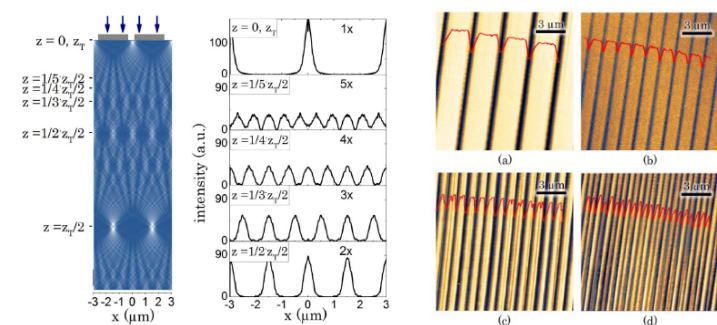


Fig. 5: (Left) Talbot pattern behind the grating with narrow slits (500 nm per 3 μm pitch) as a function of distance from a grating. (Middle) Intensity profiles at several distances indicating M_{sf} – note that the cross-section data are averaged with 10 pixels. (Right) AFM measurements of the lithography results, which are showing (a) $M_{sf} = 1$, (b) $M_{sf} = 2$, (c) $M_{sf} = 3$, and (d) $M_{sf} = 5$ from the parent mask of 3 μm pitch.

the diffraction-assisted EUV lithography and allows for generation of a variety of different nanostructures using one and the same mask [2].

For the non-paraxial case we theoretically investigated the scalability of Talbot EUV lithography. The FDTD simulation has shown that achromatic and fractional Talbot lithography with amplitude transmission masks can be used effectively for nanostructuring and pitch demagnification. [1,4] The practical resolution limit in the achromatic Talbot regime is 7.5 nm

half-pitch using TE polarized light.

- [1] S. Brose, J. Tempeler, S. Danylyuk, P. Loosen, and L. Juschkin, Achromatic Talbot lithography with partially coherent EUV radiation - process window analysis, *J. Micro/Nanolith. MEMS MOEMS* 15 (4), 043502 (2016) [doi: 10.1117/1.JMM.15.4.043502](https://doi.org/10.1117/1.JMM.15.4.043502)
- [2] G. Kunkemöller, T. W. W. Maß, A.-K. U. Michel, H. Kim, S. Brose, S. Danylyuk, T. Taubner, and L. Juschkin, Extreme ultraviolet proximity lithography for fast, flexible and parallel fabrication of infrared antennas, *Opt. Express* 23 (20), 25487-25495 (2015) [doi: 10.1364/OE.23.025487](https://doi.org/10.1364/OE.23.025487)
- [3] H. Kim, W. Li, S. Danylyuk, W. S. Brocklesby, M. Marconi, and L. Juschkin, Optical properties of 2D fractional Talbot patterns under coherent EUV illumination, *J. Phys. D: Appl. Phys.* 48, 375101 (2015) [doi: 10.1088/0022-3727/48/37/375101](https://doi.org/10.1088/0022-3727/48/37/375101)
- [4] H.-S. Kim, W. Li, S. Danylyuk, W. S. Brocklesby, M. C. Marconi, and L. Juschkin, Fractional Talbot lithography with extreme ultraviolet light, *Opt. Lett.* 39, 6969-6972 (2014) [doi: 10.1364/OL.39.006969](https://doi.org/10.1364/OL.39.006969)

Future developments

On the NanoCluster tool, our efforts will be mainly focused to really demonstrate high quality *in-situ* grown nanostructures combining III-V semiconductors, high-k dielectrics and metal gates. In this sense, vertical InAs/GaSb NW TFETs will be processed and characterized. Additionally, low charge noise GaAs/AlGaAs based nanostructures to support qubit fabrication in PGI-11 will be studied. We shall continue the work regarding the superconductor/semiconductor core-shell NWs and start to investigate the *in-situ* growth hybrid semiconductor/phase change material NW structures for neuromorphic and quantum computing. In this respect it has to be highlighted that the Forschungszentrum Jülich is currently negotiating with Prof. Jia Grace Lu (currently at USC) to join the PGI-10 as codirector in autumn of 2017. Her research will focus on the development of hybrid structures composed from semiconductors and memresistive materials to explore novel building blocks for memory based logic circuitry. The design rules for materials, which will be exploited in novel memories, require the ability to identify compounds, whose resistance can be tailored. We intend to devise an advanced scheme to identify such materials within the next three years. The resulting understanding should provide a significant breakthrough in material optimization for novel resistive memories.

Within the JARA-FIT consortia and support from the department of computer science the research of the JARA Green-IT institute will aim to the development of brain-inspired computing architectures. Next to the development of neuromorphic logic and memory, this activity will involve the integration of optical interposer/interconnects in collaboration with the activities in PGI-9 and the Department of Electrical Engineering at RWTH (Prof. J. Witzens). It is envisioned that this will require highly complex integration processes, involving 3-D architectures. The technology will require hybrid structures merging the materials mentioned above as well as new paths for nano-lithography. To this end, to obtain patterns of desired shape with nm-resolution and sufficient contrast by the use of diffraction and interference of EUV radiation in combination with the proximity printing. The development of algorithms based on inverse propagation of the radiation field between mask and wafer for designing the mask layout for generating the desired photoresist patterns is essential. The elaboration of architectures making use of self-assembly methods to produce hybrid nanostructures in 2- and 3-dimensional arrays in combination with nanolithography, material development and integration of logic, memory as well as optical components will be explored to gather insights into the feasibility of brain inspired architectures for future information technology.

2.4.11 PGI – Quantum Information (PGI-11, JARA-Institute QI)

Heads of Institute: Prof. Hendrik Bluhm and Prof. David P. DiVincenzo

Contributing Personnel (31 Dec 2016 in FTE):

The JARA Institute was founded in 2016. Despite longstanding and ongoing collaborative work, the institute is still in the buildup phase and not all personnel have yet been formally assigned to the institute. Therefore no reasonable numbers can be given.

Contributing Principal Investigators: Lars Schreiber, Barbara Terhal, Fabian Hassler

Conceived in the context of the joint proposal for the Excellence Initiative, the JARA Institute for Quantum Information was founded by a joint act of Forschungszentrum Jülich and RWTH Aachen University on 1. April 2016 (Fig. 1). It institutionalizes the strong research linkage in the area of quantum computing, and other areas of quantum information, that already existed between the two organizations. Quantum Information started as a small institute at Aachen in 2011, coinciding with the arrival of DiVincenzo there as a von Humboldt Professor; soon after, Bluhm joined as Professor in the II. Physics Institute at Aachen. These units always cooperated with their counterparts at Forschungszentrum Jülich under the JARA umbrella, but it was decided to make this cooperation more concrete, and pointed, by the founding of joint institutes; PGI-10 and PGI-11 were the first instances of these.



Fig. 1: Founding ceremony of the JARA Institute for Quantum Information, RWTH Aachen University, 1. April 2016.

Most of the scientific output of PGI-11 lies in the future. However, we mention here a selection of scientific accomplishments from recent years from the predecessor Aachen institutes, and of scientists from PGI-2 who have now joined the PGI-11 effort; these accomplishments are particularly pertinent to the near-term plans for further research in this institute.

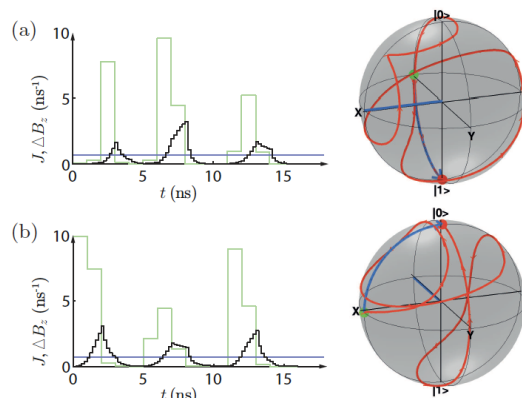


Fig. 2: Control sequences that result in high-fidelity single-triplet quantum gates.

High-Fidelity Single Qubit Gates. The group of Bluhm has built on his long experience with GaAs quantum dot qubits, and has set as a definite objective the achievement of the world's best fidelity quantum gates in this system. A large step in this direction was accomplished for the singlet-triplet qubit [1] with the help of a very extensive initial theoretical exploration of the space of possible driving-pulse trajectories. These were implemented in the experiment in an adaptive way. Theory says that 99.9% fidelity for quantum gates will be feasible, and these first experiments did not fall far short of this goal.

Real-world Quantum Hall Circulators Work started in the Aachen University group of David DiVincenzo continues under the umbrella of PGI-11, in an effort to take the basic idea of the Hall circulator and adapt it further to real-world conditions. A problem with the original proposal is that it would produce a very high impedance

device, i.e., one that would not match well to the 50-ohm microwave world. While conventional impedance matching is possible, current work [2] shows that resonant behaviour intrinsic to the capacitively-coupled Hall device itself can cause the device to self-impedance match, which could result in a profoundly reduced size of the on-chip circulating device. The group currently has a cooperation with a group at the University of Sydney that is carrying out experiments on these concepts.

Longitudinal Coupling Structures A large burden of small but not negligible stray couplings is troublesome for many multi-qubit layout concepts, particularly those involving superconducting qubits. Our investigations have recently uncovered [3] the possibility of altering the basic form of the qubit-bus coupling; the Jaynes-Cummings, or “transverse” form of coupling has clear disadvantages compared with a “longitudinal” form. Longitudinal coupling avoids many potential long-distance effective couplings that are seen with the systematic application of perturbation (Schrieffer-Wolff) theory to the existing transverse-coupling model. The longitudinal-coupling approach has the potential of substantially removing crosstalk between neighboring qubits, allowing them to be fabricated exactly at the same frequency.

- [1] Pascal Cerfontaine, **Tim Botzem**, **David P. DiVincenzo**, **Hendrik Bluhm**, *High-Fidelity Single-Qubit Gates for Two-Electron Spin Qubits in GaAs*, Phys Rev Lett. **113**.150501(2014) [doi: 10.1103/PhysRevLett.113.150501](https://doi.org/10.1103/PhysRevLett.113.150501).
- [2] S. Bosco, **F. Haupt**, and **D. P. DiVincenzo**, *Self impedance matched Hall-effect gyrators and circulators*, Phys. Rev. Applied **7**, 024030 (2017) [doi: 10.1103/PhysRevApplied.7.024030](https://doi.org/10.1103/PhysRevApplied.7.024030).
- [3] Susanne Richer and **David P. DiVincenzo**, *Circuit design featuring longitudinal coupling: a scalable scheme for superconducting qubits*, Phys. Rev. B **93**, 134501 (2016) [doi: 10.1103/PhysRevB.93.134501](https://doi.org/10.1103/PhysRevB.93.134501).

Planned New Directions

Most of the work of PGI-11 lies in the future. But it is already the focus of many new initiatives in the direction of the physical realization of quantum computing. Here is a selection of the plans for the near future:

Many of these plans are driven by the project spearheaded by PGI-11, “Scalable Solid State Quantum Computing”, a multi-million euro, 3 year initiative of the Helmholtz Association in its IVF program (Impulse and Networking Funds), approved in November of 2016. The scientific thrusts associated with these are

- achievement of the control system for a 50-qubit quantum computer. The concept for this is driven from within PGI-11, but will be carried out mostly at the ZEA-2 (Central Electronics Institute) of the Forschungszentrum.
- devise high-fidelity, long range couplers for spin qubits. This is a coordinated theory-experiment project, looking at both Si and GaAs quantum dot qubits. The technique involving an electron conveyor – a voltage-array structure for adiabatically moving an electron on chip from one quantum-dot area to another – will be the focus of the first phase of research.
- put the longitudinal-coupling schemes into practice. The theory work on this will continue to be led by PGI-11, but our Helmholtz partners at the Karlsruhe Institute of Technology (KIT) plan experiments with their superconducting qubits to experimentally explore this concept.
- Further developing of the theory and investigating the potential for implementation of the quantum surface code will go on. This will be focused on the spin qubit problem, but will cross-fertilize with the work that will go on in PGI-2 on the Majorana-qubit surface code.

In parallel with, or beyond, the IVF project activities, the plans for PGI-11 include

- Further work on a multi-year project in cooperation with IBM, in the framework of the LOGIQ program of the US IARPA agency. Work here identifies theoretically the

problems of scaling up a transmon-qubit chip to achieve a fully error-corrected qubit (involving tens of physical qubits).

- develop participation with many bilateral partners, including at TU Delft, LETI, and others, and work towards multilateral cooperations in the context of QuantERA and the Quantum Technology Flagship, focusing always on the physical implementation of the scalable solid-state quantum computer.

2.5 PoF III Recommendations of the Helmholtz Senate

- In view of the perfect mix of theory and experiment in topic 2, it is recommended to reinforce **theory in topics 1, 3, and 4**, accordingly.

In Topic 1, the theory support in the field of new laser materials was strengthened. A close collaboration with Dr. Zoran Ikonic from the University of Leeds was established. As an expert in the parameterization of band structures, he develops model parameters for the application of the materials in devices. This liaison has already led to numerous joint publications.

In program Topic 3, the different theoretical approaches for the description of memristic elements (phase-change or nanoionic cells) were summarized and overlapping bridges between quantum mechanics, density functional theory (DFT), mesoscopic kinetic Monte Carlo (kMC) and macroscopic finite element approaches (FEM) were created. This was made possible by the addition in the second application period of an accompanying DFG-funded Collaborative Research Center SFB-917: Nanoswitches, a pure theory project (B8: Theory and modulation of change-based resistive switching).

In order to strengthen the theoretical efforts in Topic 4, the HZB appointed Prof. Johannes Reuther as junior professor at the Freie Universität Berlin as part of the "Berlin Joint Lab for Quantum Magnetism (BerQuaM)". Prof. Reuther is from Caltech, Pasadena, and is concerned with the effects of magnetic frustration, which may lead to new quantum materials - so-called "spin liquids".

- The already strong coherence of the program should be intensified by appropriate measures. Therefore, it is recommended to exploit existing **synergies between topic 2 and topic 4** to a larger extent, in particular between molecular magnetism and quantum magnetism, and between oxide spintronics and multiferroics.

An initial workshop has assessed the existing collaborations between the two topics and the potential to exploit the existing synergies more efficiently. The existing cooperation agreements are to be further strengthened and new collaborations are to be initiated. The workshop focused on "Molecular Magnetism" (Bürgler, FZJ) and "Quantum Magnetism" (Lake, HZB) as well as the first results of the successful cooperation between F. Gunkel ("Oxide Spintronics", Topic 2, FZJ) and E. Schierle ("Multiferroics", Topic 4, HZB).

On 12-13 September 2016, we organized a second intensive and stimulating workshop discussing topics 2 and 4 with sessions of research talks, 2-minute speed talks and posters on the topics of functional nanostructures, topological insulators, spintronics, quantum information, quantum computing, magnetization and spin dynamics, oxide spintronics and quantum magnets. This workshop provided insight into ongoing collaborations between the HZB and FZJ and between the two program topics, along with how better to exploit existing synergies.

An inherent aspect of Topic 2 is the functionality of a materials set-up, i.e., the control of the property-to-external-parameter relation. This concept has been transferred to materials of Topic 4. Examples include the compounds CaIrO_3 and SrIrO_3 , where we plan to manipulate conductivity by the application of a magnetic field or an interface to a ferroelectric material, or the translation of Rashba physics at metal surfaces to the physics of the two-dimensional electron gas at the interface of BiAlO_3 -based oxide heterostructures.

Another example refers to magnetic skyrmions, where we translated the concept of the Fe-on-Ir(111) interface to the all-perovskite interface $\text{SrRuO}_3/\text{SrIrO}_3$, where SrRuO_3 takes on the role of the ferromagnet and SrIrO_3 the role of the spin-orbit material. This is a collaboration between PGI-1, PGI-7 and the University of Cologne.

A final example in this area is the current collaboration between F. Gunkel at PGI-7 and E. Schierle from the HZB on the subject of magnetic effects in two-dimensional electron gases at oxide heterointerfaces. Two joint beamtimes have been performed where we succeeded in detecting magnetic moments at the Ti site of $\text{SrTiO}_3/\text{LaAlO}_3$ interfaces by XMCD. A related publication is in preparation.

In total, we have so far produced 37 joint publications (FZJ and HZB) within the funding period.

- The program is strongly encouraged to ensure closer collaborations **of the programs' work on catalysis** with activities inside and outside of Helmholtz.

The advice from the referees and the HGF Senate was well accepted. Due to the close similarity of redox-processes and oxygen exchange mechanisms in memristive switching oxides to the field of catalysis and the expected cross-fertilization, the Institutes PGI-1, PGI-5, PGI-6, and PGI-7 collaborated in various ways within Forschungszentrum Jülich, in particular with the different divisions of the Institute for Energy and Climate Research (IEK) and its activities in the field of energy research. Further collaborations also ensued with various institutes at RWTH-Aachen University in the context of JARA, and also outside Jülich, with the TU Berlin, the TU Chemnitz and the Chinese Academy of Science, to name but a few. For example, PGI-1 suggested a layering of zirconium and yttrium in the fluorite structure of zirconia as a new solid state electrolyte, PGI-5, together with the ER-C, investigated the role of nanoparticles as catalysts, in particular for fuel cells, and PGI-6 and PGI-7 are jointly exploring oxide surfaces and interfaces for sensors and energy conversion applications such as water splitting. This work has been conducted within JARA FIT and various partners from RWTH Aachen University are involved, such as R. de Souza from the Institute of Physical Chemistry and T. Schneller from IWE II at the electrical engineering department.

In total we have published more than 20 papers in this field during the funding period, including papers in high-impact journals such as *Science* and the *Nature Publishing Group*.

The increased activities for quantum information processing will also have an impact on the possibilities of understanding the molecular structures of materials for catalysis and medicine. By developing new simulations of the quantum properties of materials using more efficient algorithms, a deeper understanding of the mechanisms is possible.

- The program is strongly encouraged to pursue an even **more systematic approach for using the results for innovation and for transferring the results to industry**, through patents and more connections with existing companies as well as start-ups.

To stimulate the transfer of technology and possible spin-offs, the program organized a three-day workshop on Identifying Commercial Opportunities from Emergence Science and Technology at the PGI in September 2015. The invitation went to all JARA-FIT groups. The workshop was conducted by Prof. Angus Kingon, Brown University, USA. Angus Kingon is Professor of Engineering and Professor of Entrepreneurship and Organizational Studies and has many years of experience in the initiation of successful spin-offs from universities in America and Europe.

The potential and risks of possible "private-public-partnerships" (PPPs) in the areas relevant to the PGI need to be discussed in more detail.

One development in this direction is the project "Nanoprober for the semiconductor industry". This has been selected for funding by the Helmholtz Validation Fund, as it shows particular promise in terms of the transfer of research results to market-ready products. Within this project, a unique, combined AFM/SEM nanoprober developed at PGI-3 will be validated for use in

semiconductor failure analysis. Together with the project partner Global Foundries (Dresden), the probing of individual transistors from current technology nodes will be performed. After the work has been successfully validated, the aim is to establish a spin-off company.

2.6 Future Program Development

The rapidly growing importance of information and the ubiquitous use of data in all areas of society generate new challenges for next-generation information technology. Key aspects are data and communication security, energy consumption, storage capabilities and computing capacities. As a consequence, scientific research must shift its focus from specific technologies to a global view on information and to more general concepts of information processing. In order to meet the future challenges, we need information-oriented research to take a much more comprehensive approach ranging from the understanding of information processing in biological systems through the exploration of new materials and fundamental concepts, up to the development of strategies on how to exploit all this knowledge for and with the next-generation of extremely powerful computers.

The Helmholtz Association is responding to these developments by reorganizing the entire research field *Key Technologies* into the research field *Information (Part I, 3.3)* as part of the call of the next generation of Helmholtz programs (PoF IV). The research field will be partitioned into three programs. In the process, the current program FIT will evolve and expand into a new program *Information Processing: Physical Systems, Biosystems and the Brain*, which creates the missing link between research on key principles of information processing and the successful implementation of such knowledge for novel technologies. It seeks to achieve a comprehensive understanding of the fundamental rules of information processing connecting both inanimate matter and biological systems including the human brain. It contributes to the development of “beyond von-Neumann computing”, novel computer architectures for data science, and provides the basis for future hardware solutions, e.g., for secure communication and cognitive computing.

Such a broad scope requires first and foremost a highly interdisciplinary approach, integrating different research communities and targets across the borders of several Helmholtz Centers to access the available synergies. Accordingly, these activities will be borne by Forschungszentrum Jülich, the Karlsruhe Institute of Technology, and the Helmholtz Zentrum Berlin. In order to promote rapid and lasting progress in the fields of quantum information and neuromorphic computing, Jülich will further support this development by founding several new institutes in PGI.

The result of these measures will be a strategic alignment in a new program, which will contain 6 interconnected topical domains:

- *Domain 1: Exploring New Paradigms and Device Concepts for Future Information Technology* – Information processing and communication technology in charge-based nanoelectronics is approaching its limits in terms of capacity, speed, and energy consumption. We therefore explore the use of new paradigms arising in physical systems, based on spin, topology, configurations and correlations as a future means to store and process information. Strong emphasis is put on energy efficiency by exploiting the full potential of multifunctional materials and photonics, and by developing architectures and interfaces for complex hybrid structures. The activities will encompass a broad range of engineered materials ranging from molecules and molecular structures all the way up to complex solid state systems, using a wide portfolio of highly specialized

experimental and theoretical approaches. The topic will have strong bilateral links to the topics *Fundamentals of Quantum Computing* and *Neuromorphic Computing and Network Information Processing*.

- *Domain 2: Fundamentals of Quantum Computing* – Quantum information processing promises an exponential acceleration when dealing with a number of high-impact computational problems and physically secured communication channels. While many of the basic requirements for realizing these goals have been demonstrated, there remains an immense scientific and technological interdisciplinary challenge to be addressed. This topic brings together quantum scientists and engineers with complimentary expertise at both FZJ and KIT. The ultimate overarching scientific goal is to scale up quantum information processing devices and to develop qubit control electronics. At the same time, we will continue exploring different physical realization of the qubits, ranging from established approaches such as superconducting qubits and spins in semiconductors, to potentially superior but less developed alternatives such as molecular spins, topological qubits, or quantum phase slips.
- *Domain 3: Network Dynamics and Neuromorphic Computing* – This topic investigates the brain at the level of resolution of individual nerve cells and their interactions. Brain activity is observed using electro- and optophysiological methods to acquire data on fundamental dynamical properties and the relationship between network activity and behavior. Statistical methods and neuroinformatics tools are developed and maintained for the time-resolved analysis of massively parallel data. The findings are interpreted with the help of full density network models at cellular resolution. This requires the construction and maintenance of corresponding simulation technology for supercomputers. Finally, the understanding of complex network phenomena requires the advancement of analytical theory. The fundamental neuroscience knowledge gained is used to guide the development of new neuromorphic hardware. Emphasis is put on the problems of communication and network instantiation, flexibility in neuronal dynamics, and accelerated computation. The architecture is developed in co-design with progress in the construction of large-scale neuronal network models. All levels of neuromorphic computing are addressed, from materials, circuit design and system integration, through to community building.
- *Domain 4: Molecular and Cellular Information Processing* – Information processing is key to the development, maintenance and function of biological systems. The efficient integration of a multitude of extremely diverse external and internal signals is essential for all living systems. This relies on exquisitely complex multi-scale processing systems of intracellular pathways which are extensively interconnected, creating complex networks and patterns of intracellular signals that combine to determine the cell's physiology and pathophysiology. A key challenge is to gain a comprehensive understanding of the architecture and dynamics of these networks. One long-term vision is to harness knowledge of the molecular computing capacity of cellular systems to inspire the design of a totally new generation of computing systems.
- *Domain 5: Brain Organization and Connectivity* – The human brain is organized on multiple levels both in space and time. Connectivity is key to understanding how the different levels interact, i.e., how information on a molecular and cellular level is translated into action and more complex behavior, and how, vice-versa, external information modulates signal processing, but also brain structure down to the genome. Regional segregation and integration are not mutually exclusive but rather strongly intertwined. Whole-brain models and multi-modal brain atlases are mandatory to integrate the different levels of brain organization, and to study inter-subject variability

(see “Decoding Brain Function, Dysfunction and Plasticity”). Due to the complexity and sheer size of the human brain, big data analytics and novel approaches for analytics (e.g., deep learning) are mandatory. Cross-species comparisons elucidate relationships inaccessible in the human brain. The challenges of addressing the multiple levels of brain organization are being met by linking research with Program 1 of the research field in terms of an associated partnership, which was successfully established in PoF III, and by embedding activities into the European context, the EU flagship “The Human Brain Project”.

- **Domain 6: Decoding Brain Function, Dysfunction and Plasticity** – This topic is concerned with decoding how the healthy (developing and ageing) brain processes information and how these processes are affected by neurological and psychiatric disorders. Emphasis is put on information processing at the level of both small- and large-scale networks and how plasticity of information processing is enabled. To this end, high-end neuroimaging techniques, neuromodulation methods, and computational neuroscience will be applied to investigate cognitive, motor, and affective systems of the human brain. Results will not only contribute to decoding the human brain but also enable the development of novel diagnostics in neurological and psychiatric disorders and of model-based therapeutic approaches to reduce network pathology and thereby to ameliorate information processing deficits in the diseased human brain.

It is the vision of the PGI to perform and take the lead in fundamental research on quantum phenomena for the next paradigm shifts in IT. This includes new functionalities beyond the CMOS technology today, paths towards a sustainable improvement of the integration density, novel solutions in the area of cyber security, machine learning and radically new concepts based on quantum and neuromorphic computing and, last but not least, a leap in energy efficiency of IT concepts, devices, and systems. Thus, the PGI will contribute primarily to the topical domains 1-3 listed above, as explained in more detail in the following:

Exploring New Paradigms and Device Concepts for Future Information Technology: This vision takes into account the international consensus that Si CMOS technology will remain the platform for nanoelectronics of high integration density for a much longer period than the next decade. While there will be no paradigm change in the platform, the number of materials integrated into this platform will vastly increase and this will offer revolutionary new functionalities. Promising are quantum materials. They have the potential to revolutionize energy and energy-related technologies, as well as the storage and processing of data. A stream of new quantum materials are emerging that feature unprecedented capabilities. The fabrication of materials in complex structures on an atomic scale and their characterization by highly specialized microscopes and spectroscopies go hand-in-hand and is a strength of the PGI. Integration of all these materials is a new mega-trend in nanoelectronics, called *hybrid integration*. PGI is particularly well equipped for future research in hybrid integration as it has just installed a state-of-the-art multi-materials nanocluster at the HNF; this is soon to deliver the first exciting results.

Network Dynamics and Neuromorphic Computing: One direction for this type of hybrid integration is the integration of memristive functions based on metal oxides and higher chalcogenides. This will lead to new concepts in memory, logic, and neuromorphic computing. In order to develop the scientific foundations for future neuromorphic computing systems, PGI is planning a multidisciplinary research effort within a large number of groups from RWTH Aachen University and Forschungszentrum Jülich as part of the DFG excellence initiative. We propose that novel memristor-based devices and circuits have the potential to overcome the severe limitations of classical CMOS transistor-based technology. In close collaboration with

neuroscientists from INM-6 and RWTH Aachen University we will explore the design space of possible memristor-synapse and neuron-connectivity characteristics. Through our pioneering work and the SFB 917, JARA-FIT was able to assume a leading position, and we plan to continue to set the pace in this area on an international level. We strive to develop and implement novel paradigms of computing in both hardware and software inspired by recent insights gained by neuroscience. We will elucidate how the device characteristics can be optimized and exploited with novel learning schemes. The challenges to which the PGI will contribute are the possible implementation of short time plasticity and comparison of stochastic binary switching versus multilevel switching, requiring the development of memristive elements with controlled retention times and more analog programming characteristics.

We will develop hardware architectures and software frameworks for a neuromorphic computing demonstrator. It will be mandatory to co-integrate these devices with CMOS, since a hybrid architecture combines the advantages of conventional computing components with novel neuromorphic hardware. Thereby we avoid the loss of general-purpose computing capabilities while enabling generic neuromorphic functionalities.

Fundamentals of Quantum Computing: PGI research will also be at the heart of efforts to crack the problem of the physical implementation of quantum computation. We bring to bear both world-leading theoretical and experimental capabilities in this area, and expect to partner with the strongest colleagues in Europe and elsewhere in the world. A major scientific effort in the coming time will be achieving the control system for a 50-qubit quantum computer. Together with the ZEA-2 (Central Electronics Institute) of the Forschungszentrum, we will establish that the control apparatus can be created in a scalable manner, in a way that is consistent with the constraints of the cryogenic environment of solid state qubits. In parallel with this, we will devise high-fidelity, long-range couplers for spin qubits. This will be a coordinated theory-experiment project, looking at both Si and GaAs quantum dot qubits. The technique involving an electron conveyor – a voltage-array structure for adiabatically moving an electron on a chip from one quantum-dot area to another – will be the primary focus. We will also undertake research with our Helmholtz partners at the Karlsruhe Institute of Technology (KIT), experts in superconducting qubits, aimed at putting new, alternative longitudinal-coupling schemes into practice. Last but not least, work will go on to determine the feasibility of topological qubit systems, in particular those based on topological materials and Majorana excitations.

It is envisaged that the existing expertise will be complemented by at least two or even three new institutes. These are intended to emphasize a system perspective with the goal of making major contributions to the advancement of scalable neuromorphic computers and error-corrected quantum computers in Europe. Thus, PGI's competence will span a whole range of issues from material physics to devices and also to systems. A possible outcome of these developments will be the establishment of a development and exploration facility that hosts and operates experimental quantum processors to integrate contributions from the community and allow their exploration and improvement via remote access.

2.7 Cross Cutting Section

2.7.1 Cooperation

Interdisciplinary cooperation is unquestionably an essential prerequisite for success in our research field, involving disciplines such as solid-state and surface physics, inorganic and organic chemistry, cellular and molecular biology, as well as electrical engineering and information technology. On a **local scale**, expertise beyond the PGI and other Jülich institutes,

(INM, ICS, IEK, JSC, ZEA) is not only provided by our partner institutes at the RWTH Aachen University within the framework of JARA-FIT, and it is planned to expand this alliance-type partnership to other universities and research institutions in the region. Thus, the strategy involves both strengthening the JARA institutes, but also expanding selected partnerships to other NRW universities with strong partners in focused research areas such as the University of Cologne in the area of “Quantum Matter and Materials (QM2)” supported by the German Excellence Initiative. In future, it is planned to include, together with the universities of Aachen, Bonn and Cologne, the University of Duisburg-Essen in the area of matter and light for quantum computing, the Johannes Gutenberg Universität Mainz in the area of organic electronics, nanomagnetism and spintronics, the University of Dortmund in the areas of QI, ultrafast dynamics and strongly correlated electron systems, the Max-Planck-Institute in Düsseldorf in the area of atom probe tomography, and the University of Bonn in the field of molecular self-assembly.

On a **national and international scale**, the research areas of the PGI are embedded into an extensive network of well-established cooperations with leading institutions. These cooperations are frequently revised and updated. For example, in the area of hybrid integration we plan to strive for strategic cooperations which complement our physics-based activities by 3-D integration and packaging technology through collaborations with, e.g., IMEC and the newly founded CHIPS (CMOS Hybrid Integration Processing and Systems) Institute at UCLA. We are also partners in the Joint Laboratory on Superconductivity and Bioelectronics, SIMIT, Shanghai, China.

In the field of ReRAM, we aspire to reach the limits of ReRAM scaling and concepts for 3D ReRAM integration by partnering with prestigious European institutions in the field of microelectronics, such as CEA-LETI Grenoble, Tyndall National Research Institute, MD Lab Italy, Picosun Oy Finland, within the framework of a H2020 ICT31-2017 call. We are furthermore in close contact with several national and international electronics companies (e.g., Infineon, Global Foundries, Intel, HP, Samsung, Western Digital) partially on the basis of defined research contracts or former joint projects. In the flourishing field of neuromorphic computing we are currently fostering collaborations with major partners at ETH Zürich, the University of Massachusetts and CEA-LETI.

Due to the lack of a photon source in Jülich, collaborations on both a national and international scale are the central element in J-SRL (see Part I, 4.3.). They enable us to establish and operate our instruments at external light sources in Dortmund, Berlin, and Triest (Italy) for our internal research, but also to the benefit of external users. At all of our instruments, a share of the beamtime (typically 30%) is given to external users through a peer-review process undertaken by the respective facility, with the J-SRL supplying the technical and personnel support. Currently the J-SRL is expanding its activities to PETRA-III in Hamburg, where an unique hard x-ray photoemission microscopy is being set up at the new beamline P22.

The field of skyrmionics is being investigated with European partners within the H2020 FET OPEN project MAGicSky). To develop density functional theory methods for high-performance computing, PGI and JSC scientists joined the MaX – Materials Design at the Exascale, one of the nine ‘European Centers of Excellence for HPC applications’ supported by the EU under its H2020 [e-INFRA-2015](#) call.

2.7.2 Research Infrastructures

The scientific work in the Peter Grünberg Institute makes full use of an extensive research infrastructure within the FZJ. The major infrastructure units closely connected to PGI are:

- Ernst-Ruska-Centre (Electron Microscopy)
- Helmholtz Nano Facility (HNF – Cleanroom Facility)
- Neutron Sources and Instruments (JCNS)
- Jülich Synchrotron Radiation Lab (JSRL)
- Supercomputer (JSC)
- Jülich Short-Pulsed Particle and Radiation Centre (JuSPARC, under construction)

They have already been described in Part I, Chapter 4.3.

Within the institutes in PGI, we operate individual research infrastructures adapted to specific research topics. They may be divided into sample preparation and characterization approaches.

The sample preparation is performed by a full range of physical and chemical deposition techniques, which are used to grow thin film systems, nanowires, and nanodots. A particular facility for the preparation of hybrid systems is the NanoCluster located in the HNF building, where also most of the nanostructuring activities (electron beam lithography) take place. For the synthesis of molecules and molecular structures we operate a number of specialized chemistry laboratories in PGI.

With respect to sample characterization, PGI operates a wide variety of spectroscopy, microscopy and scattering approaches, some of which are specifically developed in-house. A suite of low-temperature scanning tunneling microscopes both spin-integrated and spin-resolved is employed for atomic resolution studies of electronic and spin configurations at surfaces and molecular structures. Various kinds of scanning and full field electron microscopes address phenomena on mesoscopic length scales. Dedicated x-ray scattering instruments are employed for structural analyses. A wide variety of photon and electron spectroscopies is used to address electron states in condensed matter systems in great detail down to the level of the electron spin. Electrical characteristics can be measured in transport down to very low temperatures and in high magnetic fields. An electronic oxide UHV cluster-tool (EOC) is in operation, which enables the *in situ* characterization and nanoscale electrical manipulation of oxide thin films and devices by using scanning probe techniques and photoelectron spectroscopy and -spectromicroscopy.

2.7.3 Career development and gender equality

The PGI regards the recruitment and promotion of young scientists through excellent training at all stages of their career as one of its major tasks and responsibilities, and is indeed important in terms of sustainability.

This starts with the offer of internships and the attraction of a growing number of students doing their Bachelor and Master theses who contribute to the Program. Moreover, as the Program has close ties with national and international universities, the recruitment of PhD students has highest priority. Subsequently, during the scientific career of young scientists, international exchange and collaboration are both strongly encouraged and promoted by joint workshops, summer schools and visits abroad in competitive research groups. Cooperation takes place at group level, as well as through signed agreements between participating research centers in the Program and external university partners or research infrastructures. An example is the Center for Doctoral Studies at RWTH Aachen University, which provides continuing education and

transferable skill training opportunities for doctoral candidates. Another example is that PhD students of PGI are embedded in the “Integrated Graduate School” of the SFB 917. Those of the theory departments PGI-1 and PGI-2 participate in the DFG funded Research Training Group (RTG 1995) “Quantum many-body methods in condensed matter systems”. To foster the career development of PhD students, Forschungszentrum Jülich has begun to build up a general platform to achieve this goal (see Part I, 4.4).

The PGI secures high-potential candidates in research through the establishment of Helmholtz-University Young Investigator groups, with a joint appointment typically receiving a junior professorship at RWTH Aachen University. Between 2013 and 2017, the PGI hosted seven Helmholtz-University Young Investigator groups and one DFG-funded Emmy-Noether group.

The Forschungszentrum Jülich attaches particular significance to promoting equal opportunities for women and men. One major focus concerns the implementation of the cascade model described in the German Research Foundation’s (DFG) “Research-Oriented Standards on Gender Equality”, which is currently being adopted and realized in different ways at various universities and research organizations in Germany. The starting point is the observation that the careers of women and men with similar starting positions develop differently: although slightly more women than men successfully complete tertiary education, the proportion of women employed in research and teaching decreases successively from one career stage to the next. In order to counteract this trend, Forschungszentrum Jülich has introduced flexible target quotas. This means that the quota for a given career level is based on the proportion of women in the level below. In addition, the HGF offers programs such as the recruiting initiative to attract female professors on grade W2 and W3. Here the PGI was successful in hiring Prof. Larissa Juschkin and Prof. Regina Dittmann, and currently Forschungszentrum Jülich is negotiating with Prof. Dr. Jia Grace Lu from the University of Southern California to attract her to the PGI, as co-director of PGI-10.

2.8 List of Abbreviations

2DEG: 2-dimensional Electron Gas

AFM: Antiferromagnet

ARPES: Angle-resolved photoelectron spectroscopy

EXAFS: Extended x-ray absorption fine structure

BESSY: Berliner Elektronenspeicherring – Gesellschaft für Synchrotronstrahlung m.b.H.

CMOS: Complementary Metal Oxide Semiconductor

CRC: Collaborative Research Center

DFG: Deutsche Forschungsgemeinschaft

DFT: Density functional theory

DMI: Dzyaloshinskii-Moriya interaction

ECM Electrochemical metallization mechanism

EELS: Electron Energy Loss Spectroscopy

ERC: European Research Council

ER-C: Ernst Ruska-Centre for atomic resolution microscopy and spectroscopy with electrons,
FZ Jülich and RWTH Aachen University

FET: Field Effect Transistor

FZJ: Forschungszentrum Jülich GmbH

HAXPES: Hard x-ray photoelectron spectroscopy

HAXPEEM: Hard x-ray photoelectron emission microscopy

HF: High frequency

HGF: Hermann von Helmholtz - Gemeinschaft Deutscher Forschungszentren (Helmholtz
Association of German Research Centres)

HHG: Higher harmonic generation

HNF: Helmholtz Nanoelectronic Facility

HOMO: Highest occupied molecular orbital

HRTEM: High Resolution Transmission Electron Microscopy

HZB: Helmholtz Zentrum Berlin

IAS: Institute for Advance Simulation, FZ Jülich

ICS: Institute for Complex Systems, FZ Jülich

IEEE: Institute of Electrical and Electronics Engineers, Inc., USA

IEK: Institute of Energy and Climate Research, FZ Jülich

INM: Institute of Neuroscience and Medicine, FZ Jülich

JARA: Jülich Aachen Research Alliance

JARA-FIT: Jülich Aachen Research Alliance, section: Fundamentals of Future Information
Technology

JCNS: Jülich Centre Neutron Science, FZ Jülich
JSC: Jülich Supercomputing Centre, FZ Jülich
LEED: Low-energy electron diffraction
MOT: Molecular orbital tomography
ORR: Oxygen Reduction Reaction
PGI: Peter Grünberg Institut, FZ Jülich
PEM: proton exchange membrane
PEEM: Photoelectron emission microscopy
PoF: Program oriented Funding in the Helmholtz Association of German Research Centres (HGF)
QAHl: Quantum anomalous Hall insulator
RDMFT: reduced density matrix functional theory
ReRAM: Redox-based resistive random-access memory
RWTH: Rheinisch-Westfälische Technische Hochschule Aachen
SFB: Sonderforschungsbereich der DFG
SOT: Spin-orbit torque
SQUID: Superconducting Quantum Interference Device
STM: Scanning tunneling microscopy
STS: Scanning tunneling spectroscopy
SWAPPS: Standing-wave ambient pressure photoemission spectroscopy
TEM: Transmission Electron Microscopy
TFET: Tunnel Field Effect Transistor
TI: Topological insulator
TOM: Topological orbital magnetization
TPT: Tri-pheny triazine
TXMR: tunnelling spin-mixing magnetoresistance
VCM: Valence Change mechanism
vDW: van der Waals interaction
XANES: X-ray absorption near-edge spectroscopy
XLD: X-ray linear dichroism
XMCD: X-ray magnetic circular dichroism
XUV: Extreme ultraviolet light