Max-Planck-Institut für Chemische Physik fester Stoffe

Max Planck Institute for Chemical Physics of Solids

Nöthnitzer Str. 40, 01187 Dresden, Germany

Status Report

May 2018 – April 2021

Institute Philosophy and Strategy

The core scientific goal of our Institute is to work at the forefront of modern solid state chemistry and physics, and in particular to profit from strong interactions between departments to advance the interface of the two fields. By maintaining an open, collaborative atmosphere with minimal inter-group barriers, we profit from interdisciplinary expertise at a number of levels. Major open questions of particular interest to us include: understanding the interplay of topology and symmetry in modern materials; maximizing the level of control in material synthesis, for example to minimize defect levels; the identification and study of giant response functions at phase boundaries in materials at the borderline of standard metallic behavior; understanding the chemical nature of intermetallic compounds and related materials through experimental, theoretical and computational investigation of their chemical bonding; and high resolution measurement of chemical bonding related physical properties. Physicists and chemists are also encouraged to work together on creation of new materials and the refinement of existing materials to world-leading levels of purity at which entirely new collective phenomena can emerge. Although we always strive to advance our in-house expertise, we also aim to be outward-facing, and maintain a network of collaborator groups, of appropriate quality, throughout the world.

The cover displays an atomically resolved image of a (100) surface of SmB_6. This material is well known as a topological Kondo insulator, combining the rising field of topological insulators with the one of strongly correlated systems. The image was obtained by Scanning Tunneling Microscopy (STM) at a temperature of about 5 K. The reconstructed (100) surface is only observed on small patches of the otherwise atomically rough surface typically obtained upon *in situ* sample cleaving at about 20 K and is energetically favorable compared to polar (100) surfaces. The bright spots likely represent Sm atoms on top of a surface made up by B-octahedra, and the atomically resolved surface opens the way to scanning tunneling spectroscopy of this fascinating material.

IMPRESSUM

Herausgeber / Published by
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Executive Summary

Summary of institute scientific staff

We comprise four departments of approximately equal size. In addition to the Directors, we host two independent Max Planck Research Groups (MPRG) led by W2 level scientists, four further departmental W2 scientists, and 39 Group Leaders and Staff Scientists, 17 of whom are responsible for our Scientific Platform. We typically have approximately 65 Postdocs and 50 Masters and PhD students at any given time. Technical and administrative services are shared between the departments and provided by 87 staff members. Further details are given in Section 2.3.

Pandemic response

The work in the census period was done against the background of the Covid-19 pandemic, as a result of which we operated under restrictions of varying severity from 15 March 2020 to 30 April 2021. The measures we have taken to ensure the safety of our staff are expanded on in Section 2 of this report. We had five confirmed positive cases among our staff, corresponding to 2% compared to the Saxony average of 7%. In only one case was this the result of transmission within the workplace.

Summary of overall publication statistics

Over the three-year period covered by this report, Institute members have published 871 papers in Web of Science recognized journals. Many of our papers were published too recently for citation statistics to be a particularly useful guide, but by the end of June 2021 those published in the period 2018-2021 had received nearly 9800 citations, with 254 papers cited 10 or more times and an h-index of 46. Many of our papers appeared in leading journals, e.g. 30 in Physical Review Letters, 113 in Science or Nature group journals, 16 in the Angewandte Chemie and 16 in Proceedings of the US National Academy of Sciences. Further details of these statistics and the breakdown between departments are given in Section 2.7. In spite of the pandemic, these indicators represent a substantial advance on those given in our 2018 report. This is the case in particular for the publication in leading journals and the citations.

Invited talks

During the census period, Institute members gave approximately 380 invited talks at international conferences, workshops and individual institutions. Nearly 60% of these were given by group leaders and more junior staff. These numbers were obviously strongly affected by the pandemic, although it was still possible to give some of the talks virtually.

New senior staff and independent research groups

During the assessment period we have been working hard to attract new talent to the Institute and nominate our leading existing young scientists for independent funding. Séamus Davis of the University of Oxford and University College Cork was appointed as a Max Planck Fellow of the pan-German Graduate Center for Quantum Materials, affiliated to our Institute. Johannes Gooth won an MPRG and joined us from IBM Zürich, and Claire Donnelly of the University of Cambridge won a Lise Meitner Excellence Group and takes up post in September 2021. Existing members of our Institute Eteri Svanidze and Veronika Sunko also won competitive Minerva Fast Track Groups, and Sunko a Miller Fellowship at UC Berkeley which she is currently doing on leave from her Minerva position. Very recently Uri Vool of Harvard University has been appointed to another MPRG and will take up post in March 2022. In addition, a senior recruitment process that would result in a new department is in its late stages at time of writing, though the eventual outcome remains uncertain.

Prizes and awards

We are delighted to report that Institute scientists from several age groups have won prestigious awards. Juri Grin won the V. I. Vernadskyj Gold Medal of the National Academy of Sciences of Ukraine, and Claudia Felser won the James C. McGroddy Prize for New Materials of the American Physical Society, and was awarded International Membership of both the US National Academies of Science and Engineering, and our emeritus founding director Frank Steglich the Fritz London Memorial Prize for low temperature physics. All these are major international awards. In addition, Martin Jansen, formerly of the Max Planck Institute for Solid State Research but at the time an emeritus group leader in our Institute, won the Otto Hahn Prize of the German Chemical Society and German Research Council. At group leader level, Johannes Gooth won the Rudolf Kaiser Prize, and Philip Moll the Nicholas Kurti Prize. Last but definitely not least, our graduate students continued to excel. Veronika Sunko, Yang Zhang and Maja Bachmann each won an Otto Hahn Medaille, Mark Barber, Riccardo Freccero, Sunko and Bachmann won Springer Awards for Outstanding

Thesis Research, Freccero the Italian Chemical Society Thesis award and Sunko the Richard L. Greene Dissertation Prize of the American Physical Society and the Woodruff Thesis Prize of the UK Institute of Physics.

Doctoral training
Our doctoral training has continued to be guided by the principles of operation of our International Max Planck Research School for Chemistry and Physics of Quantum Materials (IMPRS-CPQM), which has been renewed for a further six years of funding from 2022. During the assessment period we admitted 34 and graduated 33 doctoral students, a number of whom (see above) distinguished themselves on the national and international stage. Our PhD officer, Dr. Burkhard Schmidt, co-ordinates with the PhD representatives and also holds individual meetings with each student, addressing any issues that they wish to raise.

Gender balance
We actively seek leading female scientists to build the base that our field needs for the future. We now have females in leadership positions at all levels of our Scientific and Administrative staff, and over half of IMPRS-CPQM students are women. In 2021 the Gender Equality Plan and equal opportunities website were redesigned.

A focus over the past three years has been to redress the historical gender imbalance at group leader level. In addition to the independent group leader positions won by Claire Donnelly, Eteri Svanidze and Veronika Sunko, we have appointed Haijing Zhang, Elena Gati, Ashley Cook, Iryna Antonyshyn, Kathryn Arpino and Yu Pan to departmental group leaderships, and Maia Vergniory to a departmental W2 position.

Career development
Over the assessment period a number of our group leaders and junior scientists have won academic positions across the world. For example, tenure-track positions were won by Jacob Gayles (U. South Florida), Kaustav Manna (IIT Delhi), Jayita Nayak (IIT Khanpur), Chenguang Fu (Zhejiang U.), Joyce Pham (Cal. State U. San Bernardino), Jhuma Sannigrahi (IIT Goa), Guowei Li (Ningbo Institute of Materials Technology & Engineering, Chinese Academy of Sciences), Nitesh Kumar (Bose Institute Kolkata) and Kim Modic (IST Austria). Senior appointments went to Philip Moll (Assistant Professorship EPFL in 2018 then Directorship, Max Planck Institute for Structural Dynamics in 2021), Takashi Oka (U. Tokyo), Enke Liu (IOP Beijing) Philip Hansmann (Friedrich-Alexander U. Erlangen-Nuremberg) and Clifford Hicks (U. Birmingham).

TU Dresden
As part of a formal agreement that has been in place since 1999, members of our institute contribute substantially to teaching at TU Dresden and the majority of our doctoral students graduate there. Collaborative interactions with the university have strengthened over the assessment period. In addition to the successful renewal bid for our International Max Planck Research School, we participated strongly in three substantial German grants: the successful renewals of SFB 1143 ‘Correlations and topology in frustrated magnetic systems’ and SPP 1708 'Material synthesis near room temperature' and the Dresden-Würzburg Excellence Cluster ‘Complexity and topology in quantum matter’. This last was particularly important to the university because it was one of two Excellence Clusters they were awarded, enabling their successful bid for Excellence status and ~ 90 M€ extra performance-related funding. We also successfully renewed Michael Ruck’s Max Planck Fellowship for an exceptional third term, and continue our membership of the Dresden Concept research alliance.

Civic contribution
The Helpline which we co-founded, and to which we still contribute, has become an important facility for the whole city. Its volunteers assisted with approximately 220 issues in 2020 and played a particularly important role in interfacing with the health authorities during the pandemic.

International links
Establishment and maintenance of international links has been one of the things most strongly affected by the pandemic, but we have managed to make progress on several fronts. New Max Planck Partner Groups have been established with colleagues at the University of La Plata, Argentina and the Brazilian Synchrotron Light Source.

Land and building issues
After some intense negotiation with the city of Dresden, we succeeded in reversing some planning proposals that posed a threat to our medium- to long-term ability to expand as necessary for our science. Another major development is the plan to build a new 280 m^2 clean-room and 100 m^2 office space as extra storeys above our thin film laboratories, though financial constraints at the Max Planck Society mean that this is scheduled to be finished in 2024.

Structure and Content of this Status Report

In the opening section of the Report we provide an overview of the central theme of our work: our research activities and achievements. Each of the four departments (A. Mackenzie, L.H. Tjeng, C. Felser, and J. Grin), the MPRGs of E. Hassinger and J. Gooth, the Minerva group of E. Svanidze, the work of the emeritus director F. Steglich, and the Max Planck Fellowship groups of M. Ruck, L. Molenkamp and J.C.S. Davis are described in separate sections. Collaborative (inter-departmental/group) research is strongly encouraged in our Institute, so we highlight selected joint ventures in section 1.12. In all our scientific reporting, we mark work co-authored by Institute members that was published during the census period with an asterisk (*). Other citations are either to work from other institutions or to items of our own work that were published before May 2018 or after April 2021.

In section 2 we describe the administrative, financial and organizational structures that underpin this science, and some more summary performance statistics. Our aim was to keep this as brief as possible, and cross-reference to the more extensive set of data provided in the Addendum. In addition, we will provide (confidential) 'Data, Facts, and Figures' about personnel and finances during the Science Advisory Board site visit in November 2021. We cover all the points raised in the 'Rules for Scientific Advisory Boards' document of the Max Planck Society, but have altered the order from the one given there.

We have designed our style of reporting somewhat differently to the one that we see used by other Max Planck Institutes. Rather than providing a comprehensive report on our scientific activities in a paper document running to hundreds of pages, we have striven to keep our written reporting concise and selective, concentrating on what we rate as our research highlights. The fuller background information is still available through longer reports that can be accessed from links in our main report, either on-line or using the USB drive appended to the back of the report booklet. In this way, we hope to ease the work of the readers, giving them access to the details, but only if they wish to read them.

The report is organized as follows:

1.1 Chemical Metals Science

Director: Yuri Grin[#]

Group leaders: Iryna Antonyshyn, Michael Baitinger, Horst Borrmann, Ulrich Burkhardt, Peter Höhn, Miroslav Kohout, Andreas Leithe-Jasper, Reiner Ramlau / Paul Simon, Marcus Schmidt, Ulrich Schwarz, Eteri Svanidze, Frank R. Wagner

The following basic problems were – among others -in the focus of studies at the Department of Chemical Metals Science in the reported period: What is the relationship concerning bonding and structure organization between intermetallic compounds and traditional inorganic materials? How much chemical information is given by a crystallographic description? How much information can be obtained from traditional solid-state characterization techniques? How may the current understanding regarding the nature of intermetallic compounds be used for the development of new materials?

Further progress in the understanding of intermetallic compounds was the goal and the content of the research in the Department of Chemical Metals Science in the reported period. The components of intermetallic phases mark these as a part of inorganic chemistry. The main feature distinguishing intermetallic compounds from other representatives of inorganic chemistry is the demand for valence electrons, as the usual VEC (valence electron concentration) is lower than four. In this respect, it was interesting to understand, if the formation of polycations in intermetallic substances is possible, *i.e.* how and under which conditions 'excess' electrons are formed and which structural consequences this has.

The newly discovered monogermanide of lutetium is prepared by a high-pressure-high-temperature route [1]. The electronic balance in the FeB-type structure can be described in the sense of the Zintl concept as $Lu^{3+}(2b)Ge^{2-}\times 1e^{-}$. The analysis of chemical bonding applying quantum-chemical techniques in position space reveals - beside the expected 2c-Ge–Ge bonds in the germanium polyanion - four-atomic interactions between lutetium atoms indicating the formation of a polycation by the excess electrons in the system. Despite the reduced VEC of 3.5, lutetium monogermanide follows the extended 8–*N* rule with the trend to form lutetium-lutetium bonds utilizing the electrons left after satisfying the bonding needs in the anionic Ge-Ge zigzag chain (Figure 1, left) [2, 3].

The electronic situation in $Sr_3Li_5Ga_5$ which shows an unusual bell-like $[Ga_5]$ anion can also be satisfactorily described by a Zintl-like balance $[Sr^{2+}]_3[Li^{1+}]_5[(1b)Ga^{4-}]_1[(3b)Ga^{2-}]_3[(4b)Ga^{1-}]_1$. On the other hand, the recently found pyramidal $[Ga_5]$ unit in Ba_3LiGa_5 is proven to be a *nido* Wade anion, *i.e.* it needs fewer electrons for stabilization [4]. Further bonding analysis reveals, that also the bell-like $[Ga_5]$ anion does not need all the electrons (available from the cations) for its stabilization. The excess electrons are used for the formation of the hexacations $[Sr_6]$, which are located between each of the two polyanions $[Ga_5]$ along the [001] direction (Figure 1, right) [5, 6].

Another use of the excess electrons is found in the new cage compounds Sr_8Si_{46} and $MgSi_5$, obtained by high-pressure-high-temperature preparation. Both materials belong to the clathrate family [7, 8]. Their frameworks are built by four-bonded silicon atoms, allowing the description within the Zintl-Klemm model. Because such frameworks do not require additional electrons to fulfill the 8-*N* rule, the corresponding balances show excess electrons. Especially in case of the strontium compound, their amount is quite large:

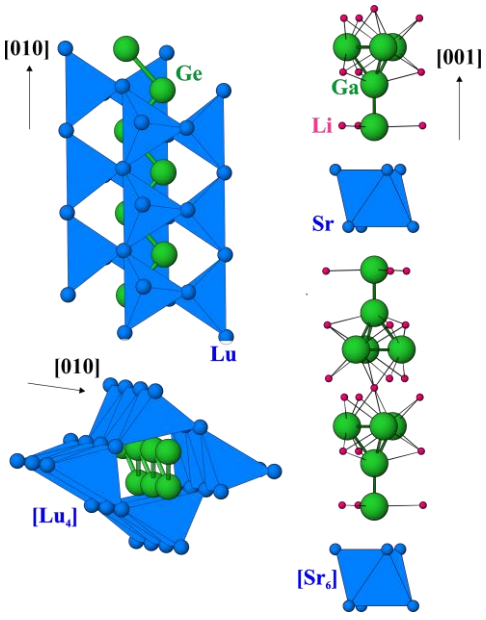

Fig. 1: Excess electrons and arrangement of polycations (blue) and polyanions (green) in the crystal structures of LuGe (left) and $Sr_3Li_5Ga_5$ (right) [2, 5].

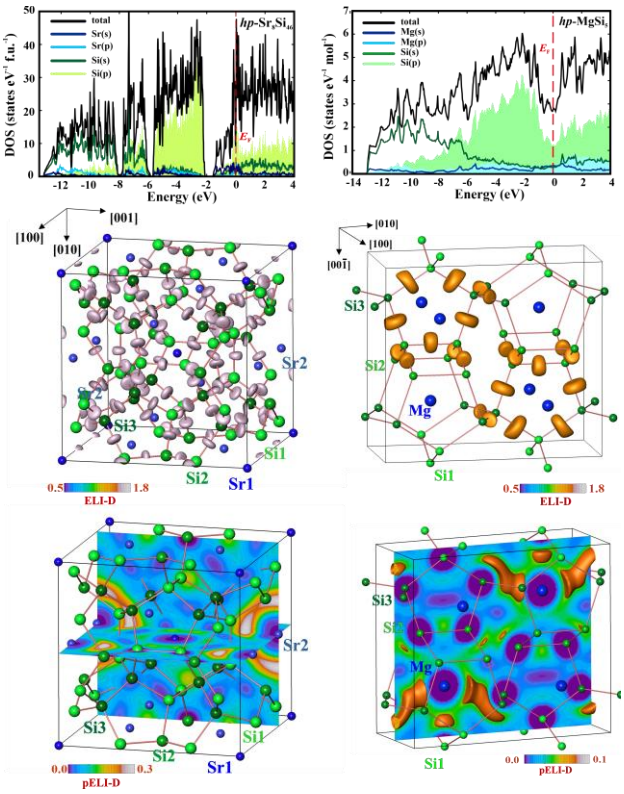

Fig. 2: Excess electrons in the cage compounds Sr_8Si_{46} (left [8]) and $MgSi_5$ (right [7]): (top) Electronic density of states (DOS) with partially filled antibonding states; (middle) Isosurfaces of ELI-D visualizing the two-atomic interactions in the silicon framework; (bottom) Distribution of partial ELI-D revealing the interaction between the fillers and the framework within the cages.

$[Mg^{2+}][(4b)Si^0]_5 \times 2e^-$ and $[Sr^{2+}]_8[(4b)Si^0]_{46} \times 16e^-$. In the DOS distribution, they are located above the (pseudo) bandgap filling antibonding states (Figure 2, top). The appearance of excess electrons in silicon cage compounds is attributed to the insufficient stability of framework defects. The two-atomic interactions within the framework are confirmed by the analysis of the electron-localizability indicator (ELI-D, Figure 2, middle). Application of the partial ELI-D technique reveals that the excess electrons are used for atomic interactions (which until now were considered as completely ionic) between filler atoms and framework within the cages [7, 8] (Figure 2, bottom).

The chemical bonding analysis in position space, used for the studies above, is a relatively young branch of quantum chemistry and is still under development. In this process, a new and unique tool for calculations of energetic parameters of *crystalline solids* – the software package ChemInt – was established. It utilizes

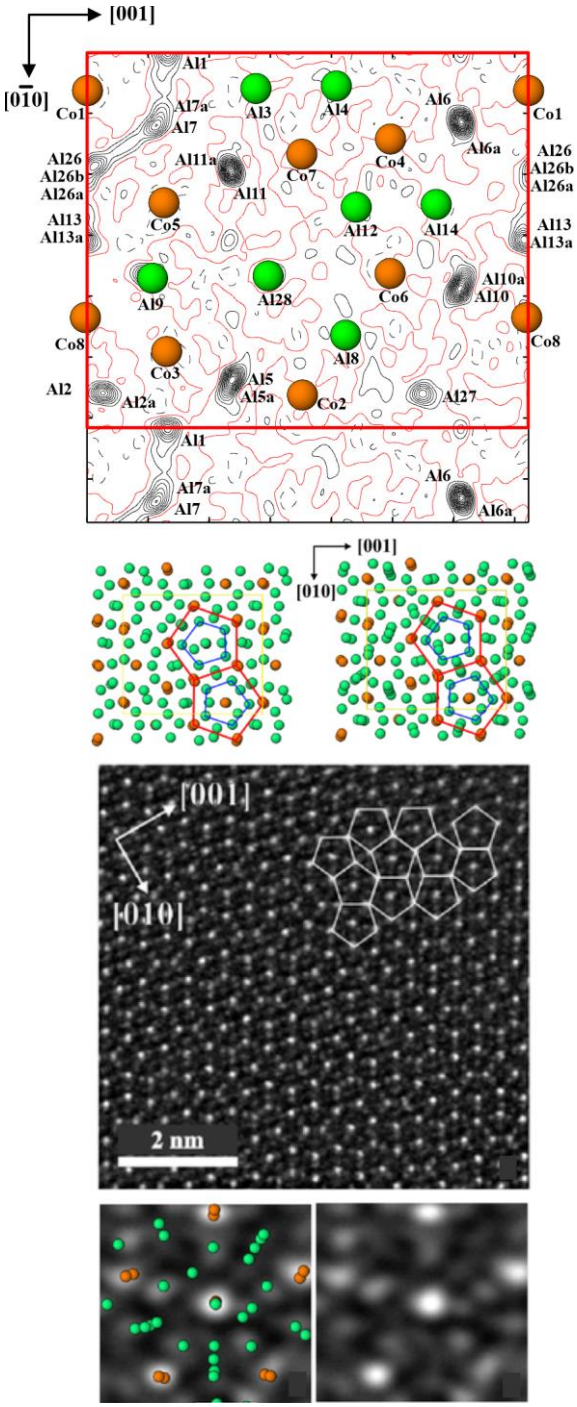

Fig. 3: Violation of translational symmetry in the real crystal structure of o-$Al_{13}Co_4$: (top) Difference electron density in the (100) plane revealing local disorder; (upper middle panel) Ordered and split-position models of the crystal structure; (lower middle panel) STEM image resembling the atomic arrangement on average; (bottom) magnified fragment of the STEM image with (left) and without (right) the projection of the available split positions.

the Interacting Quantum Atoms (IQA) approach. Employing this new tool allows to quantitatively answer questions about the relevance of atomic interactions in a given system, about the balance of

self-energy and atomic interaction energies in order to achieve a lower energy of the system, and about the contribution – and, therefore, importance – of Coulomb and exchange-correlation energy to the stability of a bond or a structure fragment [3].

The multi-atomic interactions in intermetallic structures, which were discovered and visualized by the quantum chemical position-space techniques, allow for very subtle energetic differences between atomic arrangements with similar geometric characteristics. These discrepancies may occur even within the same crystal. Thus, the crystal structure of *o*-$Al_{13}Co_4$ can – on average – be represented by a monoclinic unit cell with *ca.* 102 atoms (Figure 3). The 3D framework, bonded by multi-atomic interactions, comprises tri-atomic bent Co-Al-Co groups, which are held together by mainly two-atomic bonds. More precise analysis of the crystal structure shows a complex distribution of the difference electron density pointing at strong local disorder (Figure 3, top). An additional atomic-resolution STEM study reveals essential local deviations from translational symmetry, in particular, for aluminum (Figure 3, upper panels). With this, it becomes evident that the confinement of symmetry to one of the 230 space groups does not provide a sufficient and correct description of the real atomic arrangements, particularly in intermetallic crystals. On one hand, fine violations of the translational symmetry still allow for the determination of the average crystal structure by diffraction techniques. On the other hand, they may be one reason for the catalytic activity of this substance in the ethylene hydrogenation reaction since it provides variable local catalytic centers [9].

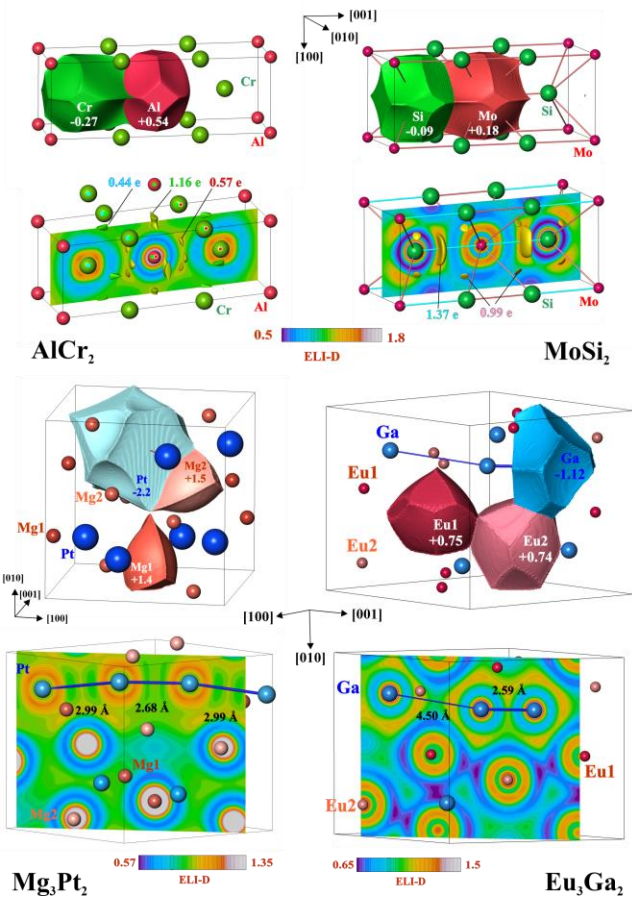

Fig. 4: Realization of the same structure type by different bonding pattern: (top) QTAIM (Quantum Theory of Atoms in Molecules) charges in isotypic compounds $AlCr_2$ and $MoSi_2$ [11]; (upper middle panel) ELI-D maps revealing multi-atomic interactions in $AlCr_2$ and mainly two-atomic ones in $MoSi_2$; (lower middle) QTAIM charges in the isotypic compounds Mg_3Pt_2 and Eu_3Ga_2; (bottom) ELI-D maps revealing the formation of Pt chains and Ga dumbbells.

The category 'structure type' is widely used in crystal chemistry of intermetallic compounds when describing close structural relationships between substances. Systematic analysis of the crystallographic characteristics leads to the conclusion that different levels of structural relations exist (isopointal, iso-configurational or isotypic structures) and that consideration of additional physical/chemical characteristics is needed to describe structural similarity. Two studies on isopointal structures of Mg_3Pt_2, Eu_3Ga_2 and K_3Bi_2, as well as $AlCr_2$ and $MoSi_2$ reveal bonding to be a reason behind the difficulties in assigning structural similarity (Figure 4). The crystal structure investigation of $AlCr_2$ – due to formation by ordering in solid state combined with a stability range below 900 °C – only became possible by applying the FIB technique to cut single-crystalline specimens from the multi-phase microstructure [10]. Both the prototype $MoSi_2$ and $AlCr_2$ belong to the same structure type. However, the crystallographically similar structural arrangement with relatively small contributions of Coulomb interactions (low QTAIM charges) is realized by different bonding patterns. While in $MoSi_2$ the mainly two-atomic Mo-Si interactions constitute the backbone of the crystal structure, the bonding in $AlCr_2$ is multi-atomic – as a consequence of the lower VEC among other reasons (Figure 4, bottom panels) [11]. The new binary compound Mg_3Pt_2 belongs to the structure type Eu_3Ga_2 [12]. The common feature of all structures in this family is the large electronegativity difference between the components, which yields essential charge transfer in the structure (Figure 4, top panels). The characteristic structural feature is the parallel linear

arrangement of the anionic component in the unit cell. Within each linear arrangement, chemical bonding analysis shows alternating two-atomic Pt-Pt interactions, stabilized by the bridging three-atomic bonds, which together yield a continuous chain anion in Mg_3Pt_2 (lower VEC). The interactions between Ga atoms in Eu_3Ga_2 and between the Bi atoms in A_3Bi_2 (*A* – alkaline metal) compounds are, however, mostly two-atomic (higher VEC). Moreover, within the linear arrangement, each so-formed dumbbell is separated from the neighboring ones by lone-pair-like interactions, *i.e.*, the linear arrangement is made of separate dumbbell anions, revealing completely different chemical entities in comparison to Mg_3Pt_2 (Figure 4, top panels) [12].

The earlier developed preparation strategy for intermetallic compounds by employing redox reactions originates from the possibility of assigning electron-localizability-based oxidation numbers for compounds where the formal assignment cannot be regularly

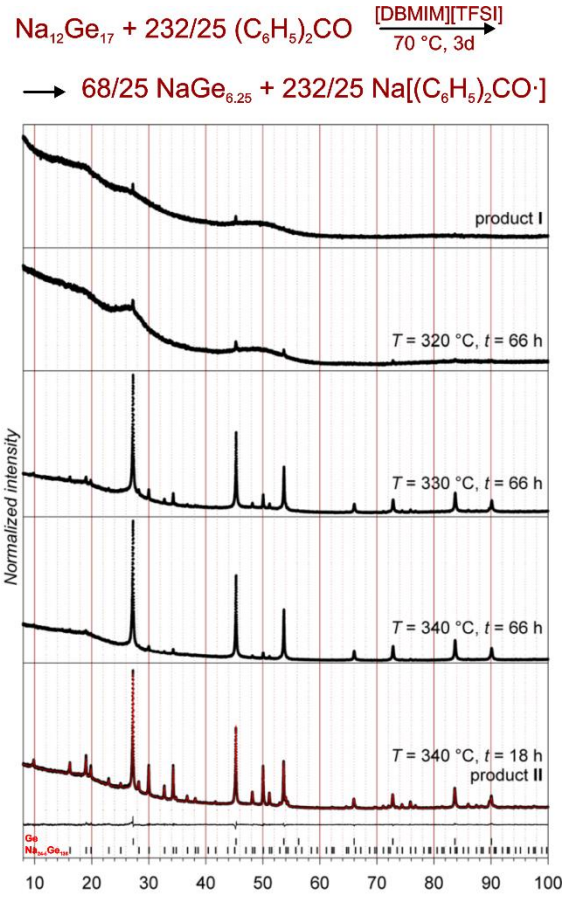

Fig. 5: Preparation of the clathrate-I $Na_{24-\delta}Ge_{136}$ in the ionic liquid [DBMIM][TFSI]: (top) Reaction scheme of the first step of the synthesis (product I – $NaGe_{6.25}$); (bottom) powder XRD patterns of the products of thermal treatment of the product I (product II – mixture of elemental Ge and $Na_{24-\delta}Ge_{136}$).

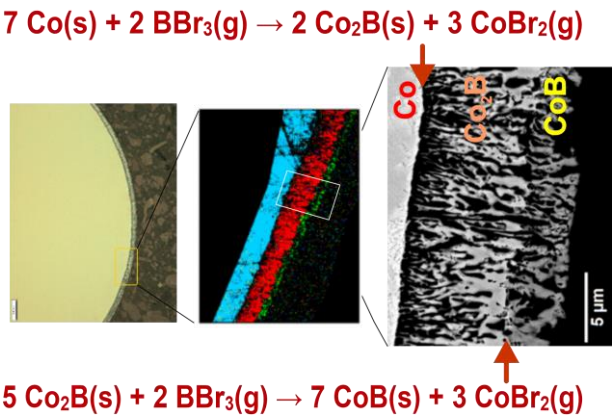

Fig. 6: Redox reactions for the preparation of borides: spatial organization of the products and mechanism of the cobalt reaction with boron tribromide [14].

applied. While most reactions realized so far were hetero-phase gas-solid processes, the newly discovered way to prepare a clathrate utilizes ionic liquids as inert reaction media. By reacting $Na_{12}Ge_{17}$ at 70 °C with benzophenone as an oxidizer and subsequent thermal treatment of the X-ray amorphous product at 340 °C, the metastable clathrate-II $Na_{24-\delta}Ge_{136}$ was obtained (Figure 5) [13]. The so-discovered reaction pathway allows for obtaining a substance, which is already unstable at room temperature in both argon atmosphere and air, and decomposes with time to a clathrate depleted in sodium.

The mechanism and the products of heterogeneous reactions yielding intermetallic compounds were studied, for example, on the interaction of cobalt with boron tribromide (Figure 6) [14]. In the first step of the reaction, Co_2B is formed, which continues to react to form cobalt monoboride. Due to the preparation setup (the reaction takes place on the surface of a heated cobalt wire), the cobalt bromide leaves the reaction space and shifts the equilibrium toward the formation of borides. The products form a 3D stable but non-dense layer, which may be useful for appropriate potential applications, *e.g.*, catalysis.

An alternative new redox route for preparing equiatomic ternary intermetallic phases is found during attempts to obtain nitridometallates of electro-negative transition metals by their reaction with binary nitrides in lithium melt supported by high-temperature centrifugation-aided filtration (HTCAF) [15].

The systematic development of vapor transport techniques provides the material basis for inter-departmental cooperation on intrinsic properties of single crystals, the results of which are published in more than 15 papers [16] (Figure 7). Particularly large

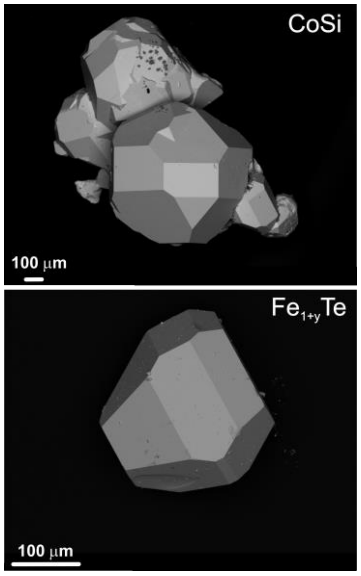

Fig. 7: Single crystals, obtained by chemical vapor transport, and used for the study on absolute structure assignment by applying EBSD technique (CoSi [17]) or on reverse phase transformations ($Fe_{1+y}Te$ [18]).

interest is paid to compounds of the FeSi type of structure. The chiral structure of this family of compounds is the origin of intriguing physical and chemical properties.

The main challenge in working with chiral materials is the assignment of the absolute structure (enantiomorph) for a given specimen. In order to achieve this, usually the complete crystal structure determination from high-resolution X-ray diffraction data should be performed, or a complex TEM study should be done. A new alternative way is the comparison of experimental electron backscatter diffraction (EBSD) data utilizing the Kikuchi pattern intensity with calculated patterns from structural data within the dynamic scattering model. Up to now, only simple inorganic materials like quartz were studied by this technique. Cobalt monosilicide CoSi is the first intermetallic compound where the applicability of this approach has been successfully demonstrated [17]. For X-ray diffraction experiments, single-crystalline specimens were cut from the matrix using the FIB technique (Xe beam) [10]. Single-crystal X-ray diffraction generally does not allow for absolute structure assignment, if the crystal structure contains one type of atom only. One of the few known examples for such a situation is the chiral allotrope β-Mn with its two enantiomorphic forms (Figure 8, top). Application of the EBSD-based technique offers the unique possibility of assigning the absolute structure to each grain in the microstructure of polycrystalline β-Mn with µm resolution (Figure 8, bottom). *I.e.*, enantiomorph mapping is possible not only for single-crystalline but also for polycrystalline (and not necessarily single-phase) materials. This opens new avenues for systematic investigations of the influence of absolute structure on materials properties [19, 20].

New approaches in understanding of intermetallic compounds allow for realization of experimental research projects in intricate chemical systems like intermetallic compounds of beryllium. Analysis of the chemical bonding in binary compounds of beryllium and platinum reveals strong polarity of atomic interactions (ionicity), which may support the appearance of a gap in the band structure. While the gap in the previously reported superconductor $Be_{21}Pt_5$ is located below the Fermi level, the compound Be_5Pt is a quite unusual example for a semiconductor, which forms with a strong valence electron deficiency. The bonding pattern is mainly constituted by 8- and 10-atomic interactions with only a small number of two-atomic bonds between Pt and Be (Figure 9, top) [12, 21, 22]. Due to the thermodynamic difficulties in manufacturing a mm-sized single-phase specimen (Be inclusions in the microstructure), the experimental proof of semiconducting behavior was obtained from the µm-sized device approach in collaboration with the Physics of Quantum Materials department [10].

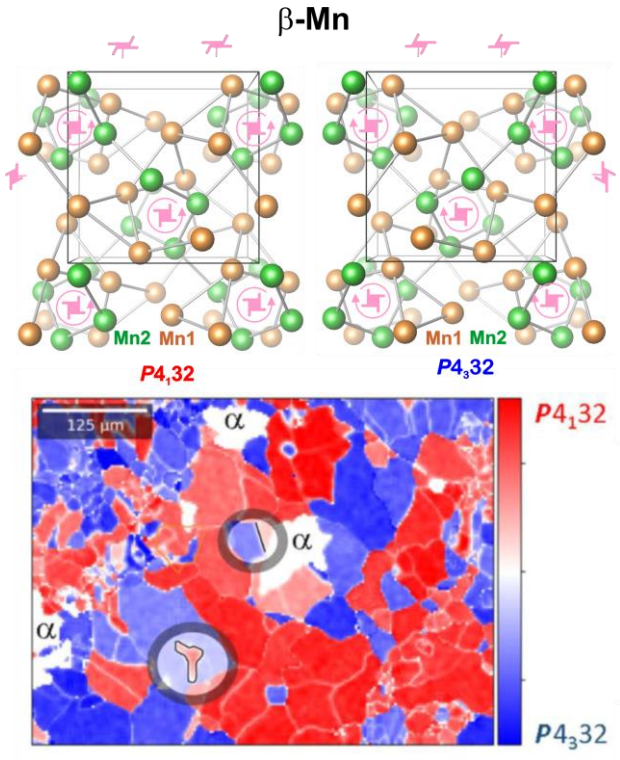

Fig. 8: Absolute structure assignment for β-manganese from EBSD [20]: (top) Enantiomorphs of β-manganese; (bottom) Enantiomorph mapping in the microstructure.

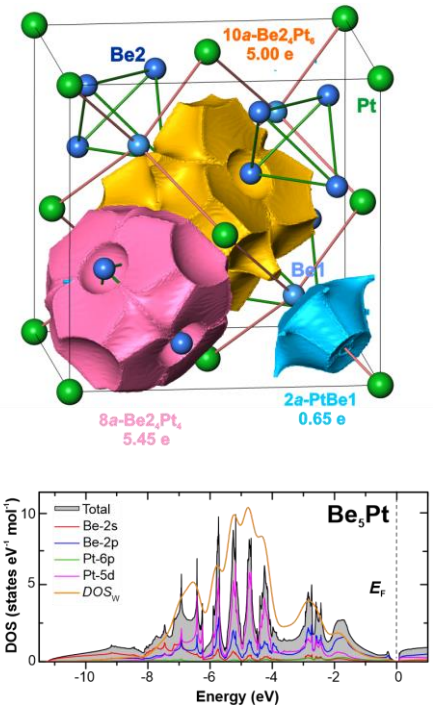

Fig. 9: Chemical bonding and properties of Be_5Pt [21]: (top) ELI-D bonding basins of two-, eight- and ten atomic interactions; (bottom) Electronic density of states with the narrow gap at the Fermi level.

Due to the high vapor pressure of beryllium at elevated temperatures, the gas-solid equilibrium has to be considered for the development of material preparation protocols. The influence of this equilibrium on the physical behavior of the heavy-fermion superconductor UBe_{13} was discussed in the previous Scientific Report. Preparative difficulties escalate when switching from binary to ternary systems. Nevertheless, the new family of compounds $R_4Be_{33}Pt_{16}$ (*R* – rare earth metals, uranium and thorium) was prepared and characterized, revealing a plethora of physical phenomena, in particular unconventional non-centrosymmetric superconductivity with Y, La, Lu and Th as *R* component [12, 22, 23].

Semiconducting states can appear in intermetallic compounds, having different bonding patterns in the background. Such a pattern, based on mainly two-atomic interactions between transition metals and multi-atomic bonding between transition metals and triels, was earlier described with the example of $FeGa_3$. Using the same approach, semiconducting behavior is predicted for *ht*-$IrGa_3$. The compound itself is a high-temperature phase and is found to be stable between 799 and 974 °C. The crystal structure at ambient conditions is strongly affected by its metastability (Figure 10, top and middle), showing unusual non-periodic modulation. The complexity of the real crystal

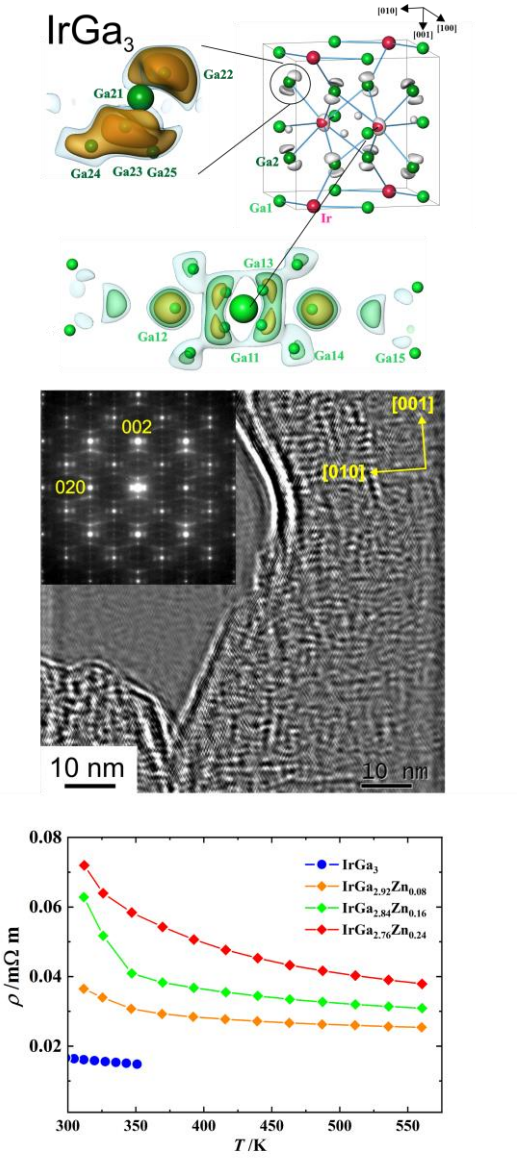

Fig. 10: Structure and properties of ht-$IrGa_3$: (top) Ideal structure arrangement of the $FeGa_3$ type and distribution of the difference electron density, obtained from single-crystal X-ray diffraction experiment; (middle) Filtered HRTEM image revealing exotic non-periodic local modulations (insert - diffuse scattering in electron diffraction); (bottom) Temperature dependence of electrical resistivity of ht-$IrGa_3$ in comparison to its substitutional derivatives $IrGa_{3-x}Zn_x$.

structure of *ht*-$IrGa_3$ influences the electronic transport and the thermal conductivity, especially its lattice part. Quite unexpectedly, both appear not to be strongly affected by the above structural features. The electrical resistivity still reflects the semiconducting character, and the lattice thermal conductivity is only slightly smaller than that of the isotypic $FeGa_3$. Moreover, the Zn-substituted materials show the expected increase of resistivity (Figure 10, bottom) [24].

The level of understanding of chemical reasons behind physical properties is crucial for the efficiency of thermoelectric materials design and development. Important thermoelectric parameters like electrical conductivity and Seebeck coefficient can already be calculated with a respectable level of reliability. Due to its multi-level nature, thermal conductivity is best to determine experimentally. Analysis of the relation between chemical composition and crystal structure on one hand, and thermoelectric properties of materials on the other, reveals the key role of chemical bonding in the chemical and structural organization of thermoelectric materials. Atomic interactions not only determine the total bonding balance in the material, regulating charge carrier concentration and transport properties, but also affect heat transport. The co-existence of different types of bonding in substances (bonding inhomogeneity) supports the reduction of lattice thermal conductivity. Finally, the spatial distribution of regions with different types of chemical bonding – bonding anisotropy – influences the direction dependence of thermal transport more than the crystallographic features [25].

The chemical behavior of intermetallic compounds is rarely discussed in literature, although their catalytic activity turns out to be one of the most attractive features of this family of substances. Applications in electrocatalysis come especially more and more into focus of research groups. The intermetallic compound $Hf_2B_{2-2\delta}Ir_{5+\delta}$ shows a chemically very unusual partial substitution of boron dumbbells by single iridium atoms. This results in different local iridium arrangements in the structure and may yield various catalytic iridium centers on the surface (Figure 11, top). The ternary substance was applied as electrocatalyst for the anodic oxygen evolution reaction in an acidic environment. The harsh oxidative conditions change the near-surface region of the material toward formation of cavities, in which $IrO_x(OH)_y(SO_4)_z$ particles are located. The latter develop their own oxygen evolution reaction (OER) activity. Such self-optimized composite material, composed of bulk $Hf_2B_{2-2\delta}Ir_{5+\delta}$ matrix and $IrO_x(OH)_y(SO_4)_z$ particles, acts as an active OER electrocatalyst within long-term experiments over 240 h at 100 mA cm^{-2} current density [26, 27].

The presented results reveal that combination of the quantum mechanical investigations of chemical bonding with the development and optimization of synthetic approaches and careful in-depth characterization is a highly productive strategy for

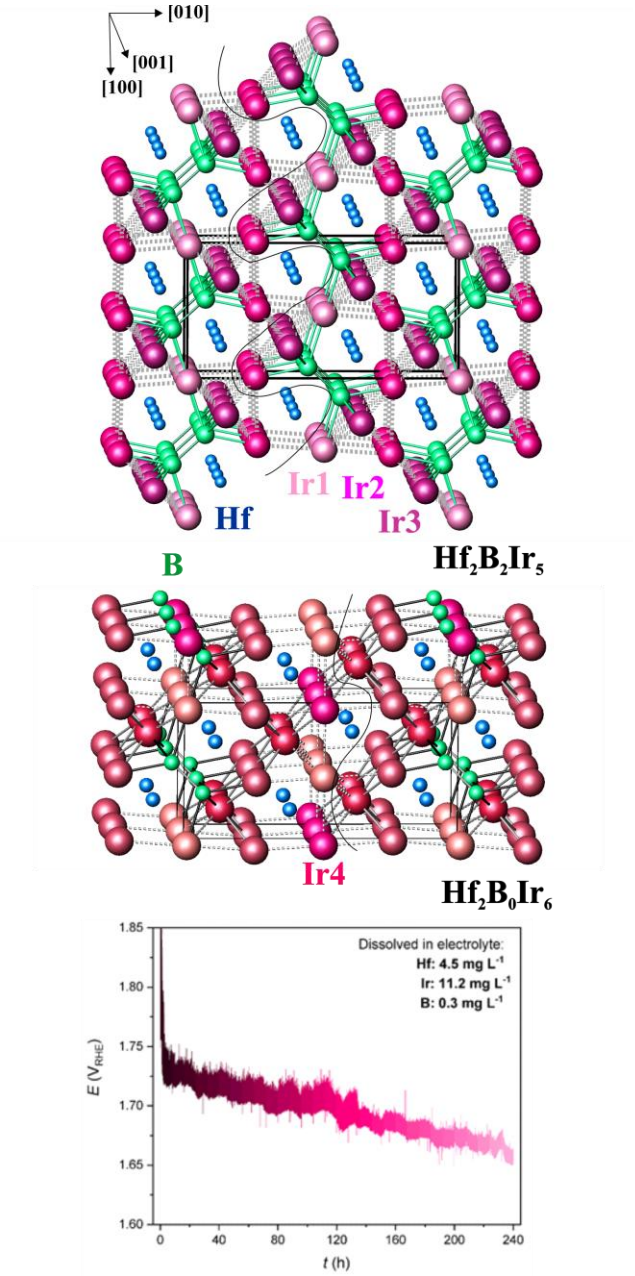

Fig. 11: Structure, bonding and chemical properties of $Hf_2B_{2-2\delta}Ir_{5+\delta}$ [27]. (top and middle) Different atomic arrangements in the crystal structure, caused by boron-pair-by-iridium replacement (curved lines show possible cleaving and atomic arrangements on the surface); (bottom) Long-term chrono-potentiometry experiment with $Hf_2B_{2-2\delta}Ir_{5+\delta}$ as anode material and its product concentrations in the electrolyte.

advancing our knowledge and understanding of intermetallic compounds. Several basic chemical problems are successfully tackled yielding essential contributions to their solution (formation of polycations, structure type *vs.* bonding pattern, multiatomic interactions, assignment of absolute structure, violation of translational symmetry).

Following the idea of chemical physics of solids, the so-cumulated understanding of the chemical nature of intermetallic compounds was used for the development of new thermoelectric and catalytic materials, in particular for creating new semiconductors containing metallic components only.

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1.2 Physics of Quantum Materials

Director: Andy Mackenzie[#]

Group leaders: Michael Baenitz, Manuel Brando, Clifford Hicks[†], Elena Gati, Michael Nicklas, Helge Rosner, Veronika Sunko[‡] and Haijing Zhang

The goal of the Physics of Quantum Materials department is to perform work of lasting significance across the interlinked themes of unconventional superconductivity, quantum criticality, strong correlations in metals and exotic magnetism, working wherever possible with materials of extremely high purity. We also aim to influence our field through technique development, designing apparatus and experimental protocols that are of utility not just for our work but for that of other groups as well. Our work is intrinsically collaborative, and as well as our department's groups collaborating with each other and across the Institute, we perform joint work with many groups from around the world.

In the following pages we will outline our main achievements in the census period of May 2018 – April 2021 inclusive, but we begin by mentioning some particular highlights:

1. Overturning twenty years of received wisdom on the unconventional superconductivity Sr_2RuO_4 by disproving one of the cornerstone results of the field and providing strong evidence for an even parity superconducting order parameter rather than the odd parity one that had widely been assumed to exist [1].

2. Experiments indicating that although of even parity, the order parameter of Sr_2RuO_4 has two components [2] (see also D. Agterberg, News & Views, Nature Physics **17** (2021) 169) and is associated with time reversal symmetry breaking [3].

3. Proving that the remarkable mean free paths of tens of microns in the so-called delafossite metals $PdCoO_2$, $PdCrO_2$ and $PtCoO_2$ arise because of extremely small intrinsic disorder in as-grown crystals. Advances that took decades to achieve in semiconductor two-dimensional electron gases and years to achieve in graphene are an automatic gift from nature in the delafossites [4, 5].

4. Observation of a new class of field-periodic resistance oscillations in microstructures of $PdCoO_2$ and $PtCoO_2$ [6].

5. Discovery, in collaboration with the independent group of Elena Hassinger, of remarkable two-phase heavy fermion superconductivity in $CeRh_2As_2$, most likely associated with local (but not global) inversion symmetry breaking [7].

6. Discovery of novel metamagnetic long-wavelength spin texturing in a second system with local inversion symmetry breaking, $Ca_3Ru_2O_7$, [8] (see also E. Blackburn, News & Views, Nature Physics **15** (2019) 625).

We will expand on each of these below, setting them in the context of our broader program of research.

1. Unconventional superconductivity

A new era for Sr_2RuO_4 research (Hicks, Nicklas, Mackenzie)

The layered perovskite Sr_2RuO_4 is a benchmark system for the broader study of unconventional superconductivity for a number of reasons. It is the cleanest unconventional superconductor ever discovered, and the metallic state from which the exotic superconductivity condenses is both well understood and relatively simple. In short, this is a superconductor which should be understandable, in detail, using the theoretical tools that have been developed over the past three decades. However, this understanding has proven to be surprisingly elusive, and in spite of the advantages listed above, there is still no consensus on the symmetry of its superconducting order parameter. Indeed, until work pioneered by our department over the past six years, the field seemed to be stagnating. That situation changed when we demonstrated during the 2015-2018 census period that the superconducting transition temperature could be increased by over a factor of two by the application of uniaxial pressure [9], stimulating intense activity in the field.

In the current census period we have made very rapid progress, building on our previous work. We proved by employing both transport measurements [10, 11] and angle-resolved photoemission spectroscopy (ARPES) [12] that the peak in the superconducting transition temperature is associated with a pressure-induced Lifshitz transition (Fig. 1).

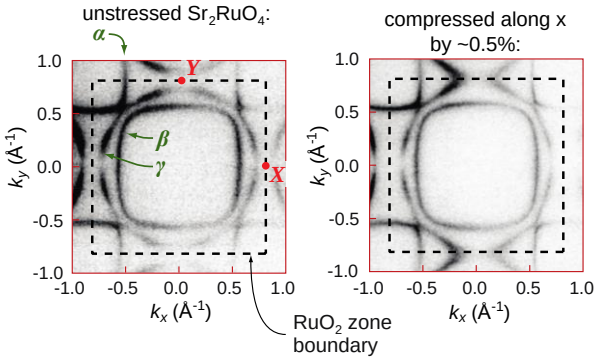

Fig. 1. A text-book spectroscopic signal from a Lifshitz transition: ARPES visualization of the Fermi surfaces of (left) unstressed Sr_2RuO_4, and (right) Sr_2RuO_4 that has been compressed uniaxially along the x axis by approximately 0.5% [11]. The γ sheet transitions from an electron-like to an open geometry. The change in Fermi surface topology occurs at a uniaxial stress of $\sigma_{xx} \approx$ -0.71 GPa. (Negative values denote compression.) The conventional labelling of Fermi sheets – α, β, and γ – is shown in the left-hand panel. Performing this experiment required the development of bespoke, passive, strain apparatus suited for challenging sample environments.

Next, a collaboration was established with the group of Stuart Brown at UCLA to study both normal- and superconducting-state nuclear magnetic resonance on uniaxially pressurized crystals. This first produced interesting results on the normal-state susceptibility and relaxation [13], and on the response of the superconducting state in highly pressurized crystals. Intriguingly, the superconducting state data looked qualitatively different to those reported by Ishida and collaborators in a 1998 Nature paper that was one of the cornerstones of the field, supposedly providing incontrovertible evidence that the order parameter was spin-triplet and hence of odd parity. Our initial thought was that there was a pressure-dependent transition between odd and even parity, but in careful follow-up experiments we discovered something even more surprising – the classic 1998 work was subject to a major systematic error whose existence we not only proved but explained to Ishida's group who quickly reproduced our result and reversed their original conclusion (Fig. 2). It is fair to say that our paper [1] completely changed the landscape of the field, stimulating intense activity which is still on-going.

It now seems as if the order parameter of Sr_2RuO_4 must have even rather than odd parity, but of what precise nature? According to resonant ultrasound and muon spin rotation experiments [2, 3] it has two components, and is associated with time reversal symmetry

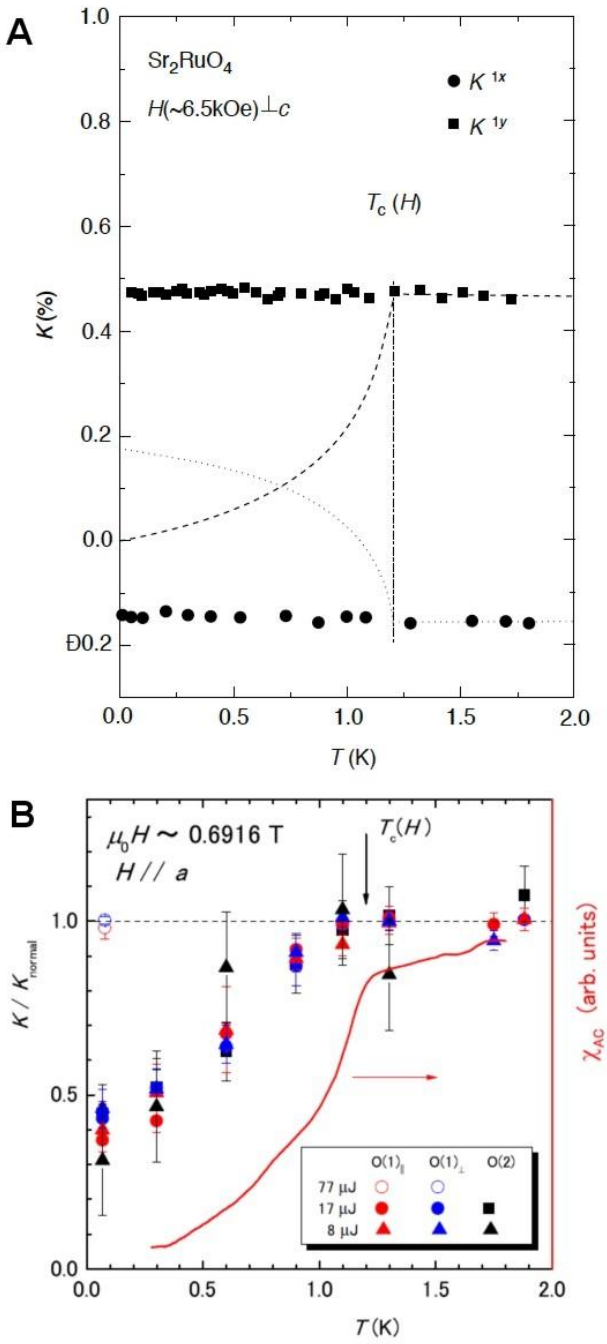

Fig. 2: A shifted paradigm. These two graphs show published data from two NMR experiments by the Ishida group in Kyoto on the same ^{17}O-enriched crystal. The first (panel A), reported in Nature in 1998, appears to show a temperature-independent Knight shift on entry to the superconducting state. Such a result could only be compatible with an odd parity, spin triplet superconducting order parameter. The completely different result shown in B is a new experiment performed in 2019 after we explained [1] how to correct a systematic error from the 1998 work.

breaking. However, there are real remaining subtleties and mysteries. In particular, we succeeded, after a four-year program of development, to measure the heat

capacity under uniaxial pressure [14]. This most fundamental of thermodynamic properties is ideally suited to searching for secondary phase transitions in the superconducting state. None are resolved within our resolution, placing strong constraints on theories of two-component order parameters; there is also increasing evidence for nodes along the Brillouin zone diagonals [15]. These observations have stimulated a host of on-going and future work which will be described in a later section of this report.

For further reading see
https://www1.cpfs.mpg.de:2443/PQM_01

Finite Q order in cuprates (Hicks, Mackenzie, collaboration with Institute Fellow Davis)

One of the advantages of uniaxial pressure is that it is likely to be a sensitive way of tuning systems in which there is modulated order such as charge or spin density waves. This realization stimulated productive collaborations with the groups of Bernhard Keimer and Matthieu Le Tacon in Stuttgart and Karlsruhe, and Hubertus Luetkens (PSI Zürich), on the $YBa_2Cu_3O_{7-x}$ and $La_{2-x}Ba_xCuO_4$ systems respectively. In $YBa_2Cu_3O_{6.67}$, non-resonant [16] and resonant [17] inelastic measurements demonstrated that long-range 3D charge density wave order can be induced along the crystallographic *b* axis following 1% compression along *a*, and that the 2D charge density wave is fundamentally uniaxial, strengthened along *b* for compression along *a* and vice-versa. While in $YBa_2Cu_3O_{6.67}$ there is good evidence for competition of the density waves with long-range superconducting order, it has long been thought that in $La_{2-x}Ba_xCuO_4$ near the '$1/8^{th}$' filling of $x = 0.125$ the superconducting order parameter is itself spatially modulated in the CuO_2 planes (a so-called 'pair density wave') and that different modulation directions in adjacent planes suppress the onset of 3D phase coherence. Considerable support for this interpretation was given by the observation that the bulk transition temperature can be increased by over a factor of three by the application of modest uniaxial pressures [18].

In both the classes of experiment described above, magnetic fields were first used to gain hints at what the underlying physics might be, with the existence of finite Q order demonstrated from analysis of signals seen in the halos of vortex cores. In contrast to magnetic field, uniaxial pressure makes its effects felt uniformly across the whole superconducting sample, leading to spectacular signal enhancement. In that context, the observation of a signal consistent with a pair density wave in STM experiments on vortex core halos in $Bi_2Sr_2CaCu_2O_8$ [19] is significant not just in its own right but as a motivation for still more ambitious future STM experiments under uniaxial pressure.

For further reading see
https://www1.cpfs.mpg.de:2443/PQM_03

Heavy fermion superconductivity (Khim, Geibel, Nicklas, Brando, Mackenzie)

Superconductivity in heavy fermion systems continues to provide benchmark data for the broader field of unconventional superconductivity, and our work in this area has resolved some long-standing issues and also yielded some welcome surprises.

Using the microstructuring capabilities provided by our department's focused ion beam facilities, we solved a long-standing mystery in $CeIrIn_5$, one of the well-known '115' family of heavy fermion superconductors. Since soon after its discovery approximately twenty years ago, there had been many observations that the temperature at which its resistivity drops to zero differs from the transition temperature identified by bulk probes like the heat capacity. Using a combination of microstructuring, transport measurements and scanning SQUID microscopy experiments (with the microscopy performed in collaboration with colleagues at Cornell University) we showed that the resistive transition is highly sensitive to local strain. In bulk crystals the strained regions typically exist at surfaces, explaining the previously mentioned transition temperature discrepancies. In microstructures, the strain fields can be tailored, spatially modulating the superconductivity and opening the way to new classes of device [20, 21].

We have also devoted effort to superconductors in which one of the active elements is Fe, but have concentrated on materials that complement main-stream research on iron pnictides. In CeFeAsO, a long programme of hydrostatic pressure measurements culminated in the construction of a definitive phase diagram for an intriguing material that in some senses provides a bridge between Kondo lattice physics and that of iron-based superconductors [22]. Another compound whose properties contain similarities with both heavy fermion and heavily hole-doped iron-based superconductors is YFe_2Ge_2. As part of a collaboration with former Institute scientist Malte Grosche, now a full professor at Cambridge, we have refined and greatly improved crystal quality of this newly-

discovered material via a systematic study of its heat capacity [23].

Arguably the highlight of our research on heavy fermion superconductors has been the discovery of a particularly intriguing class of unconventional superconductivity in $CeRh_2As_2$. A generic expectation of unconventional order parameters is degeneracy or near degeneracy, leading to non-trivial superconducting phase diagrams with multiple phases. This intuition partly stems from the superfluidity of ^3He, and observations on one of the first unconventional superconductors, UPt_3. However, in spite of the discovery of scores of new superconductors in the nearly forty years since UPt_3, there were no other thermodynamically confirmed cases of multiphase superconductivity in stoichiometric materials. The past year has seen two – UTe_2 under hydrostatic pressure, and the material discovered in this department, $CeRh_2As_2$. In a productive collaboration with the independent Max Planck Research Group of Elena Hassinger (whose article in this report describes the physics of $CeRh_2As_2$ in more detail) we have uncovered two separate superconducting phases for magnetic fields applied along the crystallographic *c*-axis [7]. We believe that the transition is between odd and even parity superconducting states, and that Rashba spin-orbit coupling plays an important role. Vitally, the crystal structure is globally inversion-symmetric but with inversion symmetry broken within the unit cell. This has two key consequences. Firstly, the Rashba-like term is allowed only in a staggered

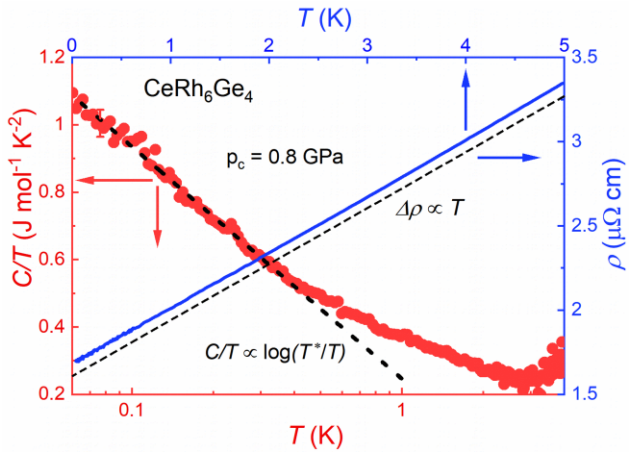

Fig. 3: Electrical resistivity ρ(T) and specific heat C(T)/T of $CeRh_6Ge_4$ at its critical pressure p_c = 0.8 GPa. ρ(T) exhibits linear behavior extending from 5 K down to at least 40 mK, whereas C(T)/T continues to increase logarithmically with decreasing temperature. Both are hallmarks of a metal tuned to a quantum critical point. Figure taken from Ref. [24].

form, changing circulation direction from layer to layer and secondly, a sharp phase transition between phases of well-defined parity is possible. If the inversion symmetry were broken globally, this would not be the case.

For further reading see
https://www1.cpfs.mpg.de:2443/PQM_04
https://www1.cpfs.mpg.de:2443/MPRG_01

2. Magnetism and magnetic quantum criticality

Although magnetism is a venerable topic in quantum materials research, its frontiers address some of the most profound modern questions in the field. Our goal is to work at those frontiers, expanding experimental knowledge and stimulating theory. In the census period our results fall into the following classes.

Ferromagnetic quantum criticality (Brando, Nicklas, Geibel, Baenitz, Steglich)

Non-thermal tuning parameters are one of the most important modern tools of quantum materials research. They can be used to traverse regions of phase space that are of particular relevance to theory, and as a tool with which to discover new phases. The uniaxial pressure tuning of unconventional superconductivity described above is one example of such a strategy; hydrostatic pressure as applied to CeFeAsO is another. Hydrostatic pressure was also used to tune the phase diagram of the heavy fermion ferromagnet $CeRh_6Ge_4$, revealing a continuous quantum phase transition that challenges the main theories of such phenomena (see Fig. 3) [24]. In another project, one of the hallmark materials of our Institute, $YbRh_2Si_2$, was tuned towards a ferromagnetic instability by the chemical substitution of Co for Rh [25].

Search for triangular lattice spin liquids (Baenitz, Brando, Rosner, Schmidt, Khim, Modic, Sichelschmidt)

At least as interesting as magnetically ordered systems are those in which some form of frustration prevents order, opening the possibility of spin liquid ground states. Our department's material work in this area has largely concentrated on triangular lattice materials. In the census period notable investigations have been performed on $NaYbS_2$ [26, 27], $NaYbSe_2$ [28], $NaYbO_2$ [29], Y_2CuTiO_6 [30] and $Sr_3CuSb_2O_9$ [31], making use in some cases of a combination of lattice frustration and spin-orbit coupling. Samples of the crystals grown for this research are shown in Fig. 4. In somewhat related work, an investigation of the low-

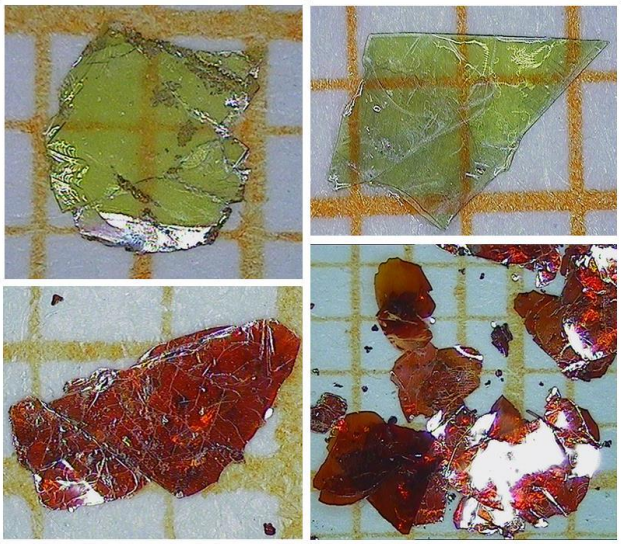

Fig. 4: Examples of plate - like $NaYbS_2$ (top row) and $NaYbSe_2$ (bottom row) single crystals used for our studies. The growth of the crystals is such that the lateral dimension corresponds to the (a,b) - plane of the rhombohedral structure of these spin-liquid candidate materials.

dimensional quasi-spin chain compound $YbAlO_3$ found evidence for deconfinement / confinement transitions of spinons [32]. In parallel with the new materials research, we have developed novel resonant torsion magnetometry techniques as a tool to measure the magnetic anisotropies that are important in understanding spin liquid candidates [33, 34].

For further reading see
https://www1.cpfs.mpg.de:2443/PQM_05
https://www1.cpfs.mpg.de:2443/PQM_06
https://www1.cpfs.mpg.de:2443/PQM_07

Novel magnetic textures and mechanisms (Sokolov, Mackenzie)

Long-range spin textures such as skyrmions are both of fundamental interest and hold the possibility of novel applications, and so are an intense area of study. We identified $Ca_3Ru_2O_7$ as a possible host for novel spin texturing because it hosts two spin sub-lattices, grew large single crystals and investigated the magnetism using a range of magnetometry and neutron scattering techniques. The long-wavelength spin texture that was discovered is a novel hybrid of antiferromagnetic and ferromagnetic alignment now referred to as a metamagnetic texture [8]. Working with the same crystals, it was also possible to understand the origin of antiferromagnetic low temperature state within which this spin texture arises. Although it had previously been thought to be the consequence of a structural phase transition, it turns out to be more interesting: the driving energy scale is actually that of a staggered Rashba effect due to local inversion symmetry breaking [35], similar to the physics thought to drive the superconducting phase diagram of $CeRh_2As_2$.

3. Ultra-high purity metallicity in delafossite metals (Sunko, Mackenzie)

The layered triangular lattice metals $PdCoO_2$, $PtCoO_2$, $PdCrO_2$ and $PdRhO_2$ have been a focal point of the department's research for the past seven years, because of their astonishingly high electrical conductivity [36]. To put this in context, $PtCoO_2$ is the most highly conducting material known at room temperature – nearly three times as conducting as silver or copper, taking into account the difference in their volume carrier densities. At low temperatures, mean free paths of tens of microns have been deduced from analysis of the bulk resistivity of $PdCoO_2$, longer than those of the semiconductor two-dimensional electron gases in which the Fractional Quantum Hall Effect was observed in the 1980s. In the semiconductors the long mean free paths were the result of decades of painstaking materials research, while in the delafossites they can be observed in unrefined single crystals. This seems so amazing that it is suspicious. Could there be some protection against backscattering, for example, that leads to the measured resistivity severely underestimating the true scattering rate?

To investigate this issue, we have undertaken a series of experiments during the current census period. First, we studied a classic ballistic experiment, transverse electron focusing. With injection and probe contacts 35 μm apart, we resolve nine focusing harmonics (Fig. 5), something that would not be possible unless the true mean free path were well in excess of 10 μm [4, 20].

More surprising still was the observation of field-periodic oscillations in the *c*-axis magnetoresistance

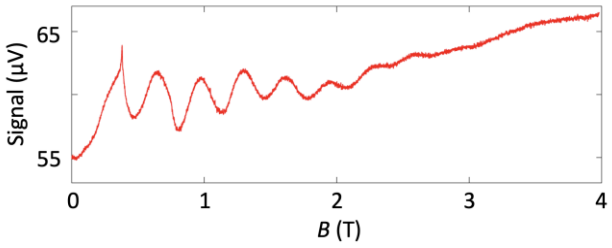

Fig. 5: The nine transverse electron focusing harmonics mentioned in the main text, which were observed at 5 K in a $PdCoO_2$ device processed from a single crystal using focused ion beam sculpting. The experiment provides a direct mesoscopic caliper demonstrating the extremely long mean free path.

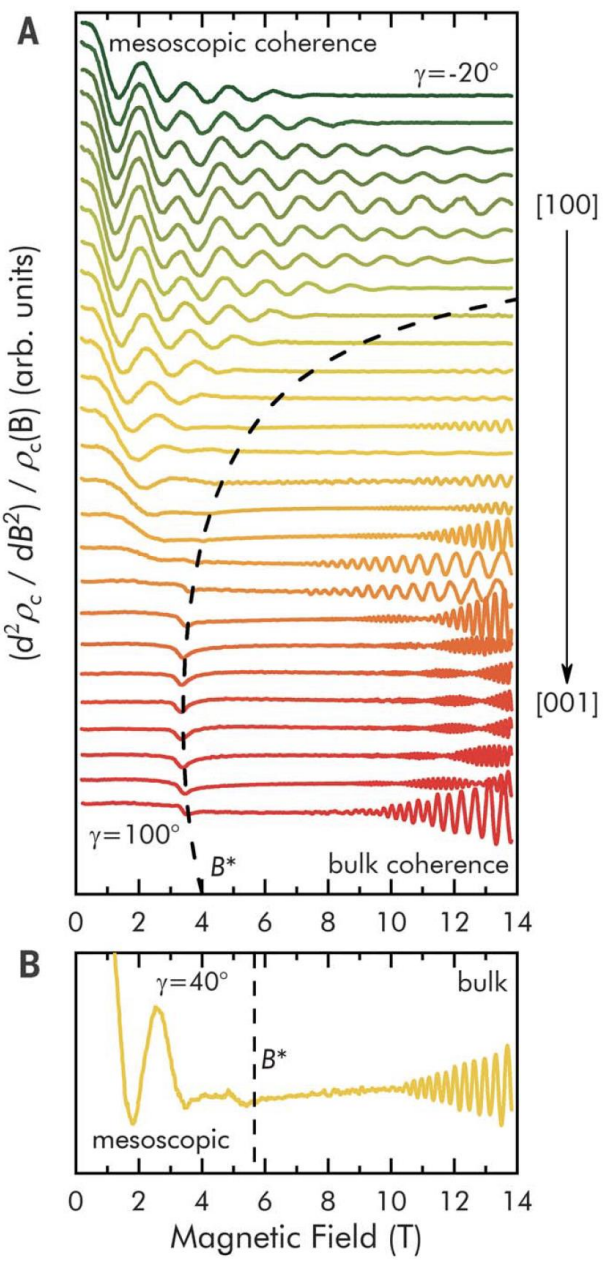

Fig. 6: Oscillations in the c-axis resistivity of a $PdCoO_2$ device give intriguing intuitive insight into the nature of magneto-oscillatory effects in metals. As a function of magnetic field angle in the same device, there is a well-defined crossover between two distinct regions. In the conventional regime of 'bulk coherence', the oscillatory period is controlled by real space flux quantisation through an area defined by a magnetic length, causing its period to be periodic in reciprocal field. As the field is rotated into the conducting planes, we enter a regime never previously observed, in which the quantization is controlled by a geometrically fixed area whose dimension is the product of one interplane spacing and the entire width of the mesoscopic channel. Figure from ref. [6].

when in-plane fields are applied (see Fig. 6). The oscillatory period corresponds to the quantization of flux through a loop comprising the microstructure width perpendicular to the field multiplied by one interplane spacing. A full explanation of the effect, including its robustness to temperature, remains challenging, but the data show that coherence must be maintained across in-plane distances of 50000 lattice spacings or more [6]. Again, this must somehow be the result of unprecedented material purity, a hypothesis that we strongly wanted to test. To do so, we deliberately introduced point defects using the unique collision kinetics of 2.5 MeV electrons with the Pd / Pt atoms in the materials. By varying the beam energy and solving for the full Mott cross-section we showed that point defects in the conducting planes scatter electrons at the unitarity limit; no suppression of backscattering is resolved. Instead, the conclusion is even more unusual. As-grown single crystals of delafossites can have fewer than one point defect in 10^5 atoms [5]. Now, the challenge is to understand why that is, and whether it can also be observed in other complex crystal structures.

While most of the above research was carried out on non-magnetic delafossite metals, magnetism is clearly present as well, on the CoO_2-terminated [37] and Pd-terminated [38] surfaces of $PdCoO_2$ or in the Mott insulating CrO_2 layers of $PdCrO_2$. In this natural Mott insulator / nearly free electron heterostructure, it was possible to observe a new class of interlayer Kondo coupling with a very strong signature in angle-resolved photoemission spectroscopy [39, 40]. We believe that these remarkable metals will continue to provide fruitful avenues of research in the coming years.

For further reading see
https://www1.cpfs.mpg.de:2443/PQM_02

4. Technique development (Hicks, Nicklas, Mackenzie)

In order to push forward our own measurement program and, ultimately, those of the broader community, we place considerable stress on the development of novel techniques and instruments, and on publishing our methods in detail. The uniaxial pressure experiments mentioned above were enabled by a number of new uniaxial pressure rigs, whose specifications are described in depth in refs. [41-45]. Many of our designs are commercialized on license by Razorbill Instruments, who sell approximately 100 rigs per year to other groups around the world.

Equally important to our work is the design of bespoke protocols for preparing microstructures for specific experimental circumstances. For the electron beam irradiation project, for example, no glues or epoxies could be used, so a method using FIB-deposited Pt to 'weld' the samples in place and provide ramps for gold contacts was developed. In our current work, mostly still unpublished, we make substantial use of FIB techniques to prepare samples for study under uniaxial pressure. Again, we publish the protocols in detail both in papers and PhD theses, hopefully to the benefit of other groups. Some examples of our development work are shown in Fig. 7.

5. Future plans

Although we thrive in the Max Planck Society environment where research decisions are taken in the present rather than having to be predicted years in advance, we have some concrete plans for future research. An obvious goal is to finally settle the issue of the Sr_2RuO_4 order parameter, something we believe we are well-placed to do. Our uniaxial pressure program will slowly change emphasis due to Clifford Hicks' career advancement to a faculty position at the University of Birmingham in the UK. However, the change will be gradual both because he retains a visiting group leader position in our department and because we have recruited an excellent group leader, Elena Gati, to lead this effort into the future. She has exciting and challenging plans to combine uniaxial and hydrostatic pressure to tune solids in entirely new ways. Another area of development will be measurement of the elasto-caloric effect, which will in principle enable simultaneous thermodynamic and spectroscopic information to be obtained from the same samples. One of the spectroscopies that we will combine it with will be spatially resolved optics, which we are in the process of establishing.

Another avenue of emphasis will be the superconductivity and normal state physics of $CeRh_2As_2$, which promises to have a particularly rich phase diagram. In the delafossite metals we will continue study of non-local transport properties, with emphasis on a new class of directional ballistics that we have recently discovered there. Some of this work will be done in collaboration with the group of Institute Fellow Laurens Molenkamp. This will be combined with a program to grow and investigate insulating delafossites with band gaps below 2 eV, which can in principle be bridged by liquid or solid ion gating, techniques being set up in the new group of Haijing Zhang. We also plan to establish apparatus for high

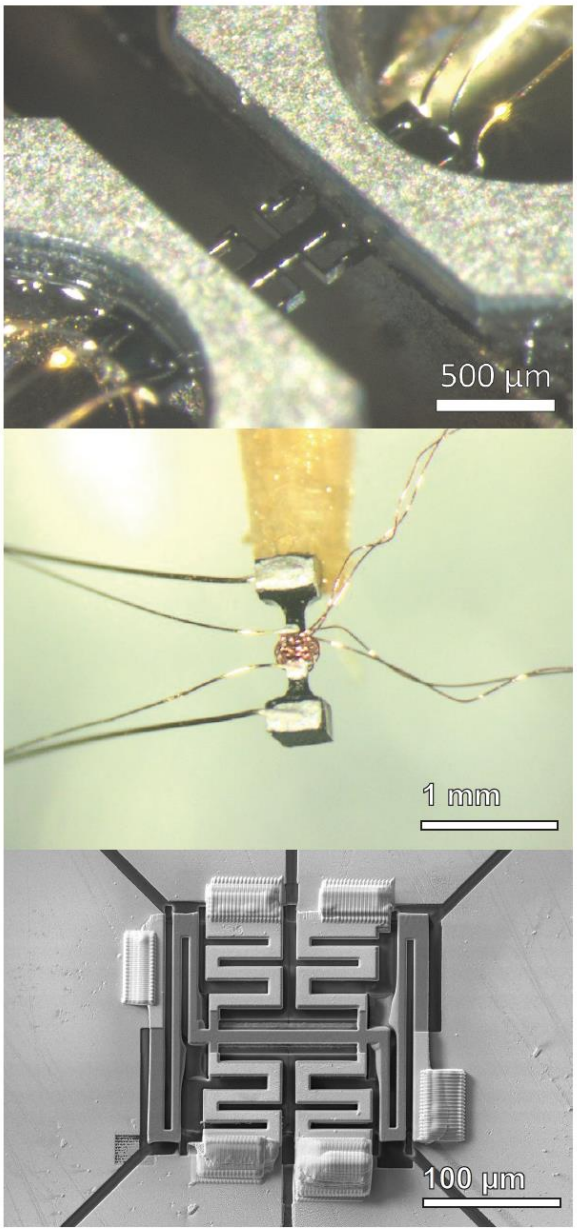

Fig. 7: Three novel sample preparation protocols specifically tailored to the needs of our experiments. In each case the starting point was a single crystal of Sr_2RuO_4 grown in our Institute. In the upper panel, a single crystalline plate has been cut into a 'spider' shape with voltage contacts sculpted from the same single crystal, and mounted in the jaws of a strain rig for magnetotransport measurements. The middle panel shows a single-crystal dumbbell whose long dimension is along the crystalline c-axis, in preparation for a magnetic susceptibility measurement under c-axis pressure. The device shown in the bottom panel is mounted, glue-free, for study while point defects are introduced by high energy electron irradiation at a special facility in Paris. Each protocol is the result of months of development, and is only possible thanks to our department's FIB facilities.

temperature measurements investigating the concept of Planckian dissipation, which hypothesizes a bound on the rate at which local equilibration can take place in many-body systems. The foundations for this work are currently being laid in the form of a review article on the topic, whose preparation is helping to sharpen our understanding of the issues that need to be addressed experimentally.

New group leader Ashley Cook (a joint appointment with the neighboring Physics of Complex Systems institute) will collaborate on appropriate topics across our program of future work.

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1.3 Solid State Chemistry

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Group leaders: Sergey Medvedev, Anastasios Markou, Satya Narayan Guin, Mengyu Yao, Kathryn Arpino, Fabian R. Menges, Yu Pan, Walter Schnelle, Chandra Shekhar, Yan Sun, Maia Garcia-Vergniory, Gudrun Auffermann, Gerhard Fecher, Peter Adler

The primary goal of the Solid State Chemistry (SSC) department is the design, synthesis, and physical characterization of novel quantum materials with topological properties. Over the past three years the main focus was on non-magnetic and magnetic topological semimetals, and chiral fermions. These materials display a plethora of novel phenomena including giant chiral Fermi arcs, giant anomalous Hall and Nernst effects, hydrodynamic flow of electrons, non-collinear spin structures, and circular photogalvanic effects. Together with the group of Johannes Gooth we have investigated quantum effects that are referred to in high energy physics as the "chiral anomaly" and in astrophysics as the "axial gravitational anomaly", as well as a phason collective mode which mimics a dynamical axion. A hallmark of many of these new quantum properties that are derived from fundamental symmetries of the bulk material is that they are topologically protected. Surprisingly, we have shown that more than 35% of all known inorganic compounds belong to a non-trivial topological materials class.

All inorganic solids can be classified through the lens of topology, using either a single electron approach [1], or by including magnetic interactions [2]: their classification is published at the internet web site https://topologicalquantumchemistry.org. Using a recently developed formalism (Bernevig, Vergniory, Felser and co-workers), that is referred to as topological quantum chemistry (TQC) and magnetic TQC, high-throughput searches of inorganic materials in well-established databases, such as the Inorganic Crystal Structure Database were carried out. More than 20,000 materials that display topological features were identified including ~100 new topological antiferromagnetic materials. Maia Vergniory has recently joined the SSC team to develop new codes for new topological materials beyond the single particle picture.
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To date, physicists have been the main contributors to the success of topology and materials. A number of solid-state chemists, particularly those who have been

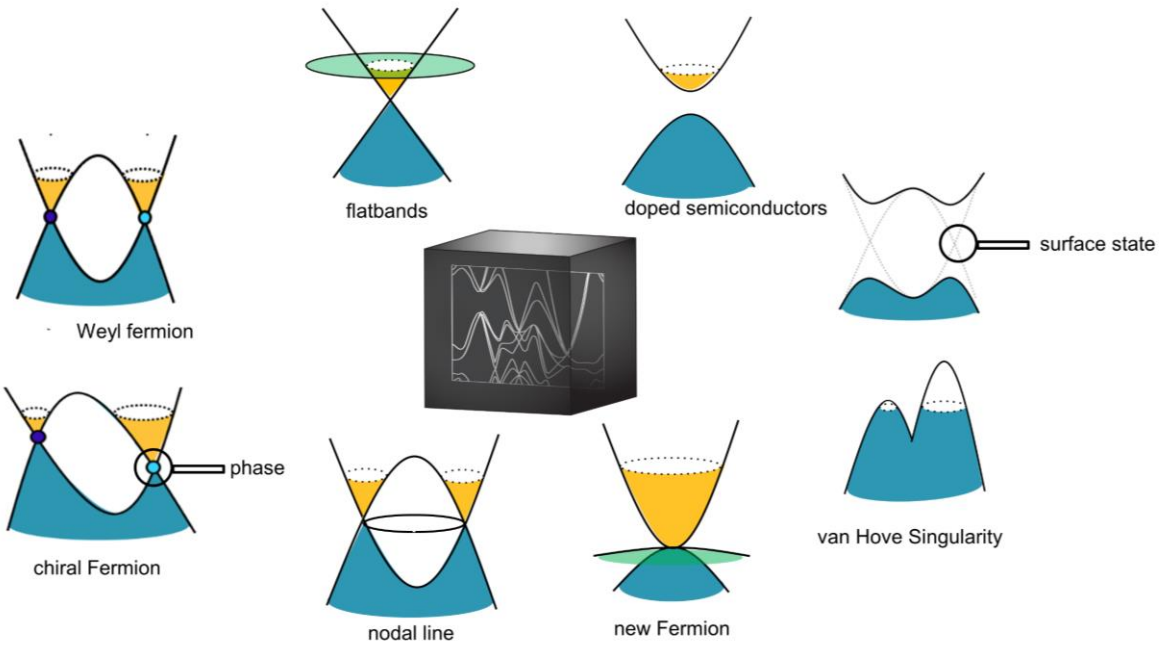

Fig. 1: Shining light on the black box of the band structure of topological materials.

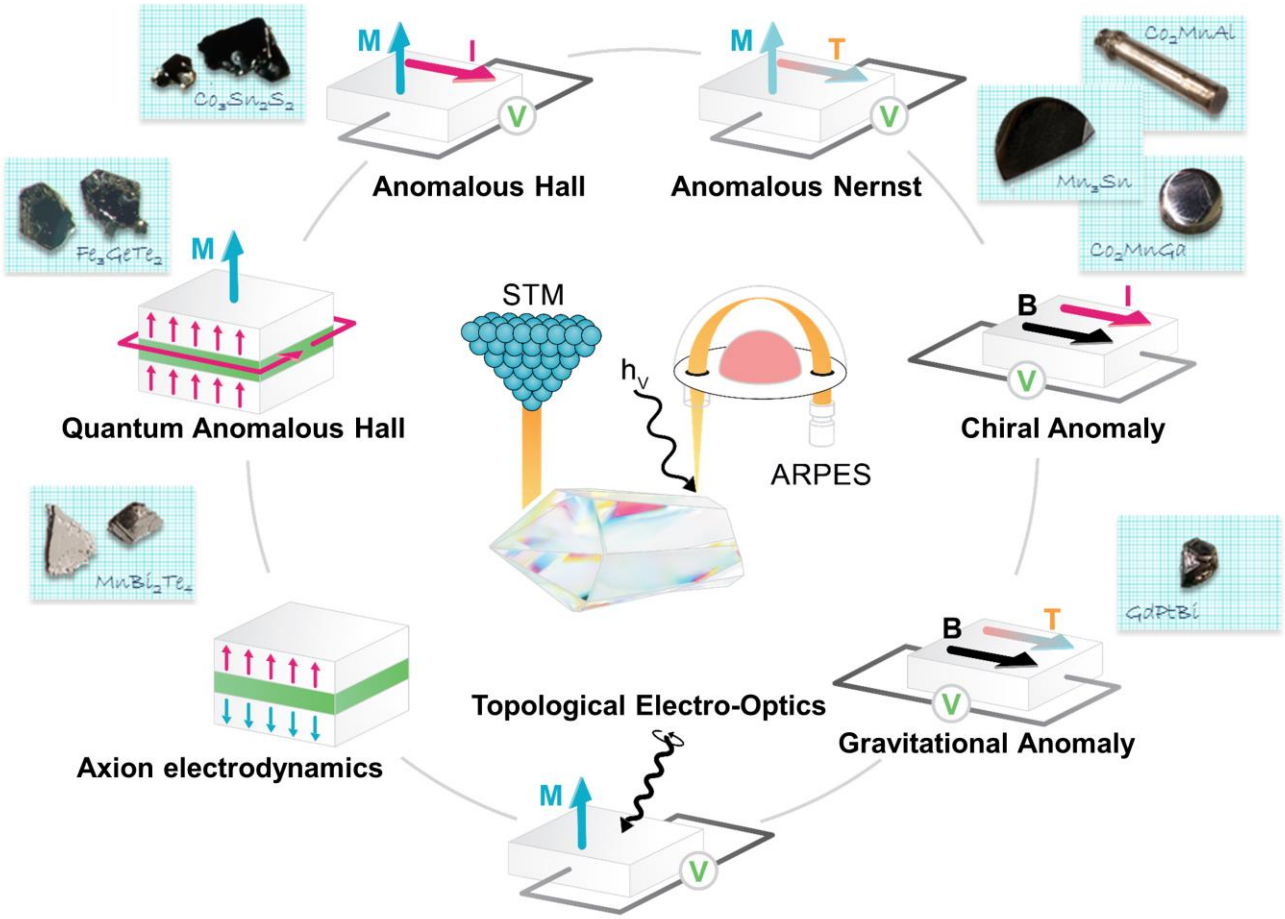

Fig. 2: Magnetic Weyl semimetals: Examples are $Co_3Sn_2S_2$, Fe_3GeTe_2, Mn_3Sn, Co_2MnAl, Co_2MnGa and GdPtBi and the magnetic topological Insulator $MnBi_2Te_4$ and their characteristic topological properties.

influenced by the works of Roald Hoffman, have joined the topological community. Band structures of solid-state crystals can be very complex. By *shedding light* on the band structure of topological materials (Figure 1), via the identification of certain signatures such as crossing points, nodal lines, flat bands, and van Hove singularities, we can identify potentially promising quantum properties of known and new materials, through high throughput calculations [3]. Besides topologically protected surface states, the phase of the wave function is important, especially for Weyl and chiral Fermions. In Weyl semimetals, nodes in the electronic structure come in pairs with opposite chiralities, or handednesses, corresponding to two forms of the Weyl Hamiltonian, and can be defined alternatively as Berry curvature monopoles and anti-monopoles, or sources and sinks, in momentum space. The projections of these nodes onto the Fermi surface are connected by topologically protected Fermi arc surface states [3]. This chiral electronic structure has led to fundamentally new properties that have been observed in electronic and thermal transport and that were originally predicted in high energy physics, as the chiral anomaly and the axial gravitational anomaly. The chiral anomaly could be responsible for the asymmetry of matter and antimatter in the universe, and the corresponding signature in a solid is the asymmetry of electron and holes. The application of a magnetic field parallel to the electric field or thermal gradient, leads, by analogy to high-energy physics, to the breaking of the law of conservation of chirality, and to a positive longitudinal magnetic field-dependent electric current.

All centrosymmetric ferromagnetic materials break time-reversal symmetry so that crossing points in the band structure are potential Weyl points or nodal lines. Several materials such as Co_2MnGa, Mn_3Ge, Mn_3Sn, GdPtBi, $Co_3Sn_2S_2$, Fe_3GeTe_2, and $MnBi_2Te_4$ were grown by the SSC team, see Figure 2. The antiferromagnetic Heusler compound GdPtBi has been shown to become a Weyl semimetal in the presence of a large external magnetic field (~2 Tesla) with two strong signatures of the chiral anomaly: a large unsaturated negative quadratic magnetoresistance for fields up to 60 T and an unusual intrinsic anomalous

Hall effect (AHE) [3, 4]. However, the potential of Heusler compounds such as PtMnSb and PtMnBi that are half-metallic ferromagnets with Curie temperatures up to 1000 K is still unexplored.

In the large family of magnetic full-Heusler compounds, Weyl nodes and nodal lines are predicted to be rather common. Co_2YZ (with Y = V, Zr, Nb, Ti, Hf; Z = Si, Ge, Sn, proposed by Bernevig and Dai 2015) and Co_2MnZ (Z=Al, Ga proposed, by Kübler and Felser 2016) have all been predicted to be Weyl semimetals. The proof that Co_2MnGa is a magnetic Weyl semimetal was obtained via angle-resolved photoemission spectroscopy (ARPES), by which nodal lines and the related drum-head surface states were observed [5]. However, the cubic structure made it difficult to observe the chiral anomaly. Co_2MnZ has Weyl nodes near the Fermi energy and, as a consequence, a giant AHE and a giant anomalous Nernst effect (ANE) that stems from the divergent Berry curvature at the Weyl nodes. Co_2MnGa exhibits one of the highest anomalous Hall conductivities, Nernst effects, and anomalous Hall angles amongst all known magnetic Heusler compounds at room temperature. Heusler compounds have natural advantages of high magnetic ordering temperatures, clearly defined topologically non-trivial band structures, low charge carrier densities, and strong electromagnetic responses, and thus show great promise for the study of quantum effects. The design of a material that exhibits a high-temperature quantum AHE (QAHE) via quantum confinement of a magnetic Weyl semimetal and its integration into quantum devices is the next step. Realization of the QAHE at room temperature would be revolutionary in overcoming the limitations of many of today's data-based technologies, which are affected by large electron scattering-induced power losses. Such a discovery could pave the way to a new generation of low-energy consuming quantum electronic and spintronic devices [3]. A precondition for the investigation of the QAHE are thin films and devices that are being grown in the SSC department.
https://www1.cpfs.mpg.de:2443/SSC_11

High throughput calculations for all known cubic magnetic Heusler compounds have identified those that should have intrinsic anomalous transport properties. The Fe_2-Heusler compounds, in particular, were identified as outperforming the best ferromagnetic Heusler materials reported to date. These results revealed the importance of symmetries, especially mirror planes, in combination with magnetism for the observation of giant anomalous Hall and Nernst effects. Simple crystal structures, such as cubic or hexagonal, with as many mirror planes as possible are the best candidates because negative Berry curvature otherwise cancels out positive Berry curvature in different parts of the Brillouin zone. MnAlGe was identified as a layered anisotropic topological nodal-line magnet, that is structurally related to cubic Heuslers. This compound exhibits a unique 2D-Berry curvature distribution that results in a large anomalous Hall conductivity.
https://www1.cpfs.mpg.de:2443/SSC_05

Another group of materials that has drawn a lot of attention are those that have a kagome lattice formed from one of the 3d-transition metal ions Iron, Manganese and Cobalt. These exhibit Weyl and Dirac fermions for both ferromagnetic and antiferromagnetic phases. The kagome lattice can host correlated topological phases and at the same time flat bands. The dimensionality of the electronic and magnetic structure varies with the material's composition and this together with the particular transition metal ion sets the magnetic interaction. The non-collinear antiferromagnetic three-dimensional Mn_3Ge and Mn_3Sn [4] and the quasi-two-dimensional ferromagnetic Shandite $Co_3Sn_2S_2$ [6] were predicted by the SSC team to have unusual properties. It was believed that, typically, antiferromagnets that have a net magnetic moment of zero, have a zero net Berry curvature because the negative Berry curvature cancels out the positive Berry curvature, although crossings are observable in the band structure of antiferromagnetic metals. In agreement with this simplified picture the AHE is absent in nearly all antiferromagnets that that have zero magnetization. Two systems, hexagonal Mn_3Sn and Mn_3Ge, have a non-collinear triangular antiferromagnetic structure that might be the origin of a non-vanishing Berry curvature that is observed in density functional theory calculations. Mn_3Sn and Mn_3Ge have Weyl points close to the Fermi energy and show the predicted properties with an AHE even at room temperature. The strong response of a Weyl semimetal to external stimuli makes these compounds promising candidates for topological antiferromagnetic spintronics. The Shandite crystals contain transition metal atoms that lie on a quasi-two-dimensional kagome lattice, and which leads to ferromagnetism with an out-of-plane orientation. One of the most interesting candidate materials is $Co_3Sn_2S_2$, which has the highest magnetic ordering temperature within this family of materials, and for which the Co magnetic moments are oriented in a direction perpendicular to the kagome

plane. Magnetic field-dependent electrical transport measurements are consistent with the chiral anomaly [6]. Angle resolved photoemission spectroscopy and scanning tunneling microscopy experiments revealed intrinsic time-reversal symmetry broken Weyl semimetal states in $Co_3Sn_2S_2$ with very long Fermi arcs [7, 8]. Owing to the large Berry curvature in $Co_3Sn_2S_2$, that stems from its Weyl nodes and weakly gapped nodal lines, and its relatively low electrical conductivity, its anomalous Hall conductivity and anomalous Hall angle have been measured experimentally to have large values of up to 1,130 S cm^{-1} and 20%, respectively [6]. This readily grown magnetic Weyl semimetal thus serves as an ideal platform for studying Weyl physics: its Berry curvature-induced intrinsic AHE makes it a candidate for the observation of the QAHE in a 2D insulating system that would thereby give rise to dissipationless edge currents [9]. This study establishes that a magnetic kagome lattice WSM is a key material for both fundamental research and applications in devices that connect topological physics and spintronics.
https://www1.cpfs.mpg.de:2443/SSC_04

Heusler compounds with non-collinear magnetic structures, in addition to *k*-space Berry curvature, also possess real-space topological states in the form of magnetic antiskyrmions, which have not yet been observed in other materials. These magnetic skyrmions are topologically protected nanoscopic vortices of magnetization that can be stabilized in a magnetic field. Magnetocrystalline anisotropy in uniaxial crystal structures is required to be strong enough to resist self-demagnetization in the shape of a thin lamella or film. Inspired by the properties of $Mn_{1.4}PtSn$, we discovered new skyrmion bubble materials with similar features of perpendicular anisotropy and spin-reorientation as those in the traditional hard magnetic materials, MnBi and $Nd_2Fe_{14}B$, which exhibit a large topological Hall effect at room temperature. However, the spin-reorientation transition is unfavorable in hard magnets. To avoid this, new very promising rare-earth-free hard magnetic materials without spin reorientation transitions, including especially Rh_2CoSb and $(Fe,Co)_2(P,Si)$, were discovered.
https://www1.cpfs.mpg.de:2443/SSC_10

Based on the relationship between topology, Berry curvature and a giant thermoelectric response, several new Mn based compounds were discovered. For a large longitudinal thermoelectric effect, i.e. a Seebeck effect, an optimal charge carrier concentration of ~10^{19} cm^{-3} near room temperature is required and, therefore, Dirac/nodal line semimetals with a small density of states were considered. Taking $YbMnSb_2$ as an example, a large Seebeck coefficient of over 160 µV K^{-1} and a large power factor of 2.1 mW m^{-1} K^{-2} at 300 K have been achieved. For the case of highly conductive topological semimetals with large Berry curvatures, the transverse thermoelectric performance, e.g. anomalous Nernst effect (ANE) was explored. $YbMnBi_2$, $EuMnBi_2$ and MnBi were found to show large ANE signals exceeding several microvolts per Kelvin.
https://www1.cpfs.mpg.de:2443/SSC_02

As already mentioned, electrons in conventional Weyl semimetals are described by relativistic equations, which originally arose in the field of high energy physics. But there are also solid-state crystals in which Weyl-like electrons have no corresponding analogues in high-energy physics and are, therefore, described as 'new fermions' [3]. These 'new fermions' have a topological order that is four times larger than that of conventional Weyl fermions. Compared to high-energy physics, symmetries can be broken in the solid, which must be preserved in a vacuum, which leads to new electron properties. One of these symmetries is inversion symmetry, which is broken in chiral crystals. In a chiral material, the atoms follow a spiral, step-like pattern as in biological systems such as DNA. While the staircase rotates clockwise in one system, it rotates counterclockwise in the opposite system. Single crystals were grown by either a self-flux method (PtAl, PtGa, PdGa) or a Bridgman technique (RhSi, CoSi), or by a chemical vapor transport method (CoSi): both enantiomers are available for PdGa. All crystals grow in the same space group (the $P2_13$ space group, no. 198), as MnSi and FeGe, which exhibit the B20 structure type and in which skyrmions were originally found.
https://www1.cpfs.mpg.de:2443/SSC_06

The "enantiomers" are mirror images of each other. While Dirac fermions are four-fold degenerate and Weyl fermions are two-fold degenerate, the new fermions can even show six and eight-fold degeneracies. The so-called Rarita–Schwinger fermion, which obeys the relativistic field equation for spin-3/2 fermions, and other multi-fold fermions resulting from three-fold and six-fold degenerate band crossings were discovered as new kinds of quasi-particles in the materials RhSi, CoSi [10], PtAl [11] PtGa [12] and PdGa [13] via ARPES. The band structures of the two enantiomers are identical while the phase of the wave function and the chirality of the

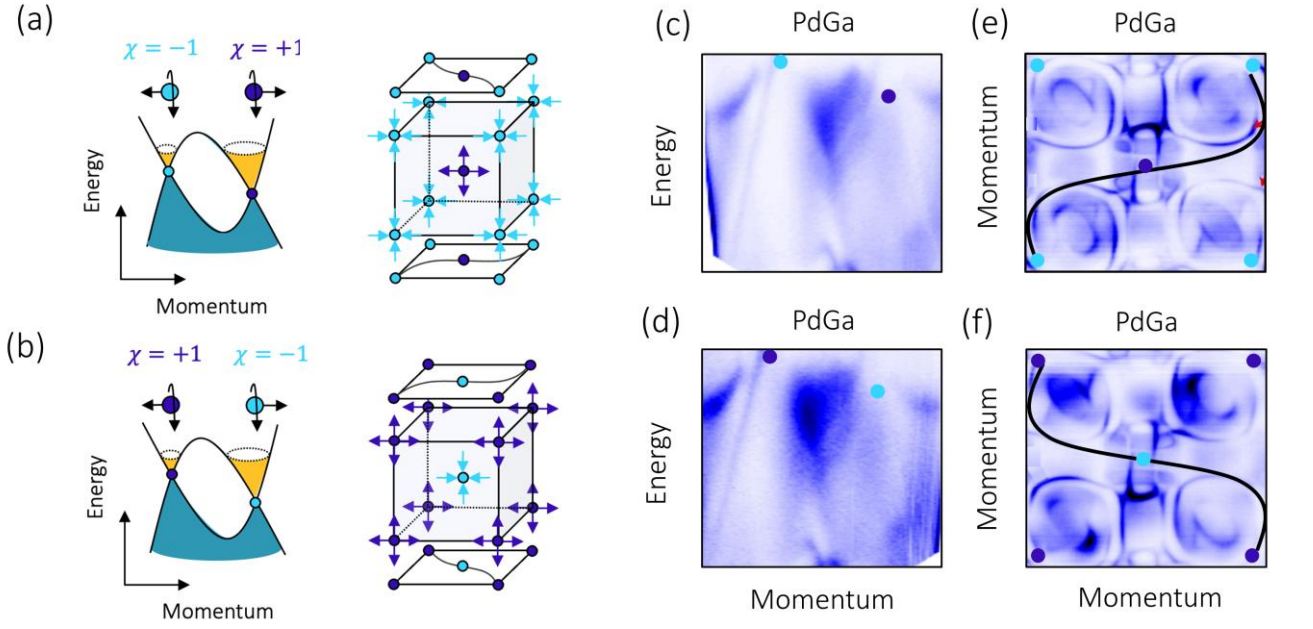

Fig. 3: Chiral semimetals. (a) Sketch of the band structure, and its distribution of the Weyl crossing points of enantiomer A, (b) its band structure, and its distribution of the Weyl crossing points of enantiomer B. The Fermi arcs on the surface of the crystal are chiral. (c) Images of the angle-resolved emission spectroscopy of the interior of PdGa enantiomer A and (d) enantiomer B. (e) Images of the angle-resolved emission spectroscopy of the surface of PdGa enantiomer A and (f) enantiomer B.

Weyl points are mirror images of each other in the two enantiomers. The two Weyl crossing points (light blue and dark blue) lie at different energies in these systems (Figure 3). This leads to several remarkable properties, including a giant circular photogalvanic current [14], a chiral magnetic effect, and other transport and optical effects that are forbidden in Weyl semimetals. In addition, the electrons on the surfaces of these crystals exhibit a highly unusual helicoid structure that spirals around two high-symmetry momenta, and the complex band topology results in very long Fermi arcs (over the whole Brillouin zone), which are orders of magnitude larger and more robust than those of any known Weyl semimetals. As for Weyl metals, the complex band topology of the two enantiomers leads to Fermi arcs, which, however, are also mirror images of each other (Figure 3e and f). These investigations also find very large topologically non-trivial energy windows, where no trivial bands intersect with the Weyl cones.
https://www1.cpfs.mpg.de:2443/SSC_03

Topological materials exhibit symmetry protected metallic surface states and massless high-mobility electrons, making them ideal materials for designing heterogeneous catalysts. However, there is limited information available on how topological materials with specific properties interact with reaction intermediates. Therefore, understanding the role of electrons and surface structures in topological materials is crucial for designing highly efficient heterogeneous catalysts for use in electrochemical water splitting and fuel cells. High-quality topological bulk single crystals were used to confirm the direct relationship between topological properties and surface redox reactions by experiment and theoretical analysis. It was observed that the catalytic reaction efficiency could be altered by external stimuli such as magnetic and strain fields. The successful absorption of chiral molecules on chiral topological surfaces is an extremely promising direction which we would particularly like to study in the future. We believe that the manipulation of topological electronic structures by external stimuli, including chiral light, could be highly useful for designing highly-efficient catalysts, which could be used in the future for asymmetric synthesis and for studying the origin of life.
https://www1.cpfs.mpg.de:2443/SSC_01

Outlook

So far, we have only touched “the tip of the iceberg”. New theoretical concepts about topological properties and materials appear again and again. In particular, many predicted materials are not yet available as single crystals of sufficiently high quality to be useful for experiments. Even if at first glance all inorganic

materials have been cataloged into trivial and topological compounds, this was largely carried out under the assumption of non-interacting electrons. Only in 2019 was the SSC department and the Nanostructured Quantum Matter Group successful in measuring a large positive contribution to the magnetic field-dependent electrical conductivity of a phason [15, 16]. Weyl-charge density insulators were proposed to be correlated axions (Shoucheng Zhang). The prediction of a Weyl-CDW (charge density wave) phase in $(TaSe_4)_2I$ was confirmed this past year through ARPES measurements of high-temperature Weyl points [16] and transport experiments demonstrating axionic responses in the low-temperature CDW phase [15]. We fully expect that there are many more axion systems to be discovered. Predictions of these CDW systems, magnetic materials and so-called correlated materials, such as rare earth compounds and transition metal oxides and nitrides, must include interactions. Some examples predicted by us of so-called correlated topological materials are rare earth and actinide pnictides, filled skutterudites and $BaBiO_3$. For the theoretical prediction of oxides, fluorides and nitrides, more complex methods and, thus, longer computing times are required. However, since these materials have larger band dispersions and larger band gaps, we expect stable topological properties. The SSC department now includes a new research direction namely the investigation of topology of covalently bonded oxides and chalcogenides with the aim of melding the traditional chemical understanding of bonding with the possibilities of topology. It is likely that topology and correlations are deeply intertwined due to complex interactions of orbital parities, structural symmetries, spin texture, and electron–electron interactions. These materials can be extremely useful for future topological catalysis. It is also time to consider new avenues beyond those of condensed matter physics, for example, for topological catalysis, thermoelectric applications, and the investigation of topology in chemistry beyond the solid state by including chiral molecules.
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Moreover, we expect further novel topological phases due to correlation effects in materials which do not exist in solids with free electrons. These include, among others, Kondo-Weyl systems, Weyl super-conductors and three-dimensional quantum Hall phases. Furthermore, it would be expedient to grow even better single crystals with Dirac, Weyl and new fermions with very low defect densities to investigate hydrodynamic effects in these materials. We have made significant progress in the understanding of the hydrodynamic flow of electrons in WTe_2 [17] and WP_2 [18] and are looking forward to collaborating with the new MPRG of Uri Vool. After the promising results that have been obtained so far on topological semi-metals, the question now remains as to whether there are also measurable topological effects in metals.

The application of an external pressure to drive topological quantum-phase transitions is an effective method for obtaining a better understanding of topologically non-trivial phases by elucidating the interaction between different ground states. Achieving a superconducting state in topological materials is of particular interest as an important step toward topological superconductors. The high-pressure group maps the electronic and structural phase diagrams of topologically nontrivial materials by evaluating the electronic transport properties (electrical resistivity, Hall effect, and magnetoresistance), and chemical structure (by Raman spectroscopy and synchrotron x-ray diffraction) at high pressures. To gain insight into pressure-driven electronic transitions, experimental studies are accompanied by theoretical calculations of the electronic band structure.
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Topologically non-trivial materials also offer a rich material phase space for extending nonlinear transport and optical effects, especially through the engineering of their topology and Berry connection and curvature. Many predictions of theory are not realized experimentally. Inversion-symmetry breaking Weyl semimetals are of particular interest, as they allow for the generation of even-order nonlinear responses which can be enhanced by divergent Berry curvatures at the Weyl nodes. Topologically non-trivial materials thus offer substantial improvements over existing nonlinear materials, though the precise combination of topologically non-trivial properties that would produce the strongest nonlinear effects remains an open question.
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Only a few groups world-wide are focusing on light matter interactions of topological materials. By inviting Fabian Menges to found a new group in the SSC department we want to join forces in this exciting scientific direction. Fabian Menges will advance our understanding of the fundamental electromagnetic properties and functionalities of material systems with quantum phases and topological surface states.
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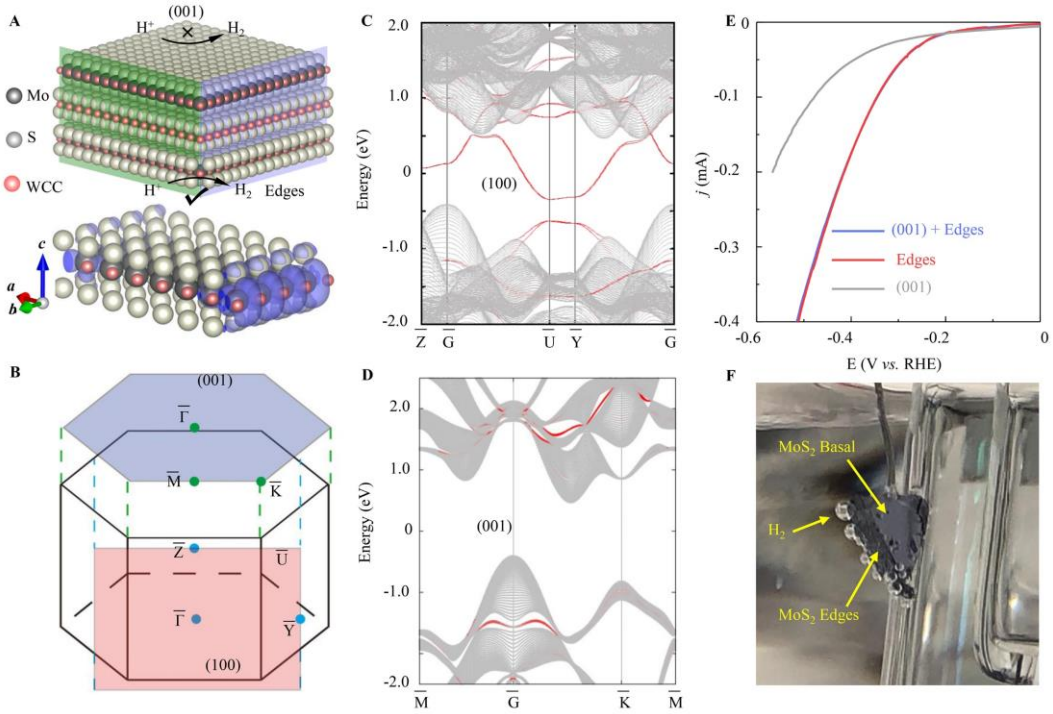

Fig. 4. The role of obstructed surface states in 2H-MoS_2 for hydrogen evolution. (A) Position of Wannier Charge Centers and Obstructed Surface States in 2H-MoS_2 (up), and the charge density states at the edges and basal plane (bottom). (B) Conventional hexagonal Brillouin zone of 2H-MoS_2, showing the (001) basal plane and the (100) edges. Calculated band structures of 2H-MoS_2 at the (C) (100) and (D) (001) surfaces, respectively. (E) Linear sweep voltammetry curves of the whole crystal, edges, and (001) basal plane, respectively. (F) Photo of the hydrogen evolution reaction process which clearly shows the production of hydrogen bubbles at the edges.

The new concept of TQC has helped us to identify diverse topological compounds and their topological protected surface states. An important question is whether we can go beyond topology to identify other novel surface states. With TQC it is also possible to identify the particular crystal surfaces of all trivial materials, both non-magnetic and magnetic, that display trivial surface states. There are three distinct classes of compounds, filling enforced obstructed insulators (feOAIs) [19], obstructed atomic insulators (OAI) [20], and orbital obstructed atomic insulators (OOAI). The feOAIs are trivial insulators which can be identified only by the number of electrons and their Wyckoff positions: examples are GaS, ZnSb and CuP_2 which can be partially connected to the Zintl and Zintl-Klemm concepts and the Pearson's (8-N) rule, including anion-anion and cation-cation bonding. More general are OAIs, which have a center of charge not localized on the atoms. In the case of OOAIs, the surface states are formed from otherwise empty orbitals at atoms at the occupied Wykoff centers. The corresponding obstructed surface states (OSSs) are defined by the bulk symmetry of the respective compound and, in real space, are centered at crystallographic sites where there are no atoms, namely Wannier charge centers. Using high-throughput computational methods applied to a database of 85,700 inorganic compounds, we have identified more than 200 such "obstructed insulator" catalysts. Characteristics of these OSSs are high stability, good conductivity, and high charge carrier density near the Fermi level. We have verified this approach for the well-known 2H-MoS_2, whose active sites are consistent with our calculations of OSSs, see Figure 4. This new methodology will facilitate and accelerate the discovery of new catalysts for a wide range of heterogeneous redox reactions, where sustainability, toxicity, and cost can be considered.

External Cooperation Partners

Andrei Bernevig, M. Zahid Hasan (Princeton); Yulin Chen (Oxford); Haim Beidenkopf, Binghai Yan (Weizmann); Joel Moore, Joe Orenstein (UC Berkeley); Prineha Narang, Amir Yacoby (Harvard); Vidya Madhavan (University of Illinois); Stuart Parkin, Niels Schröter (MPI Microstructure Physics, Halle).

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[18]* *G. B. Osterhoudt et al., Physical Review X **11** (2021) 011017, doi.org/10.1103/PhysRevX.11.011017*

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[#] Claudia.Felser@cpfs.mpg.de

1.4 Physics of Correlated Matter

Director: Liu Hao Tjeng[#]

Group leaders: Simone Altendorf, Chun-Fu Chang, Zhiwei Hu, Alexander Komarek, Oliver Stockert, Liu Hao Tjeng (Director) and Steffen Wirth

The activities of our department are centered around seven researchers with specific and complementary expertise. They lead groups and follow their own scientific interests both independently and as part of an overall team that carries out projects in a collaborative manner on various aspects of correlated materials. Highlights of their research activities will be described below. Philipp Hansmann (group leader) has left us in early 2020 for a W2 professorship in theoretical physics at the University of Erlangen-Nuremberg.

1. Introduction

The spectacular physical properties often observed in transition metal and rare-earth compounds challenge our understanding of solid-state physics. These properties include metal-insulator and spin-state transitions, frustration and multiferroicity, heavy-fermion behavior and superconductivity, as well as phenomena involving topologically protected surface states. We would like to understand how the electrons in such materials interact with each other to generate those unusual quantum phenomena.

The research activities of our department are experimental in nature and are focused on the investigation of the electronic and magnetic structure of the materials. We use various forms of synchrotron radiation and neutron-based spectroscopies, as well as in-house XPS and STM/STS. Our experimental activities have also a strong material development component: new materials, both in bulk and in thin film form, are being synthesized.

Members of our Department coauthored 224 publications in the May 2018 – April 2021 census period (187 in 2015–2018, and 161 in 2012–2015). About 75 out of the 224 publications have appeared in journals with an IF ≥ 8 (PRL=8) and 48 with IF ≥ 12 (PRX, Angew. Chemie=12, IF as of June 1, 2021).

2. Selected research highlights

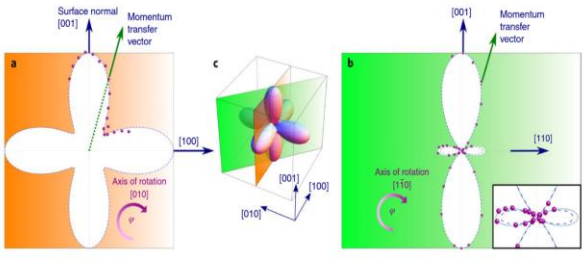
In the following we will present highlights that are representative for our research activities.

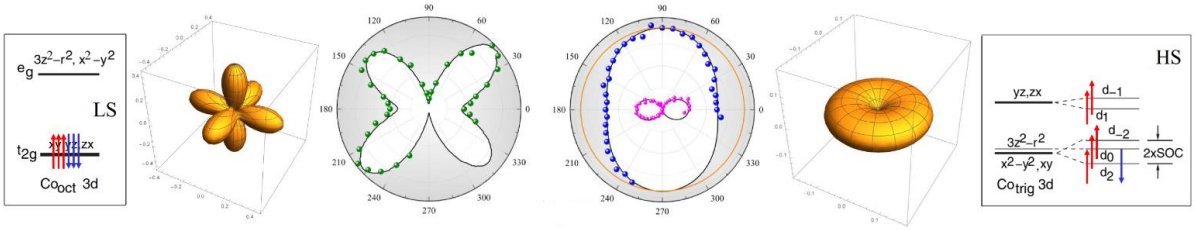
Orbital imaging and spectroscopy using s-NIXS (Tjeng):

The search for new strongly correlated quantum materials is often focused on materials containing transition-metal, rare-earth and/or actinide elements. Understanding the behavior of the *d* and *f* electrons involved is hereby essential. The identification of the active *d* or *f* orbitals has so far been mostly deduced from optical, X-ray and neutron spectroscopies in which spectra must be analyzed using theory or modelling. This, however, is also a challenge in itself, since *ab-initio* calculations hit their limits due to the many-body nature of the problem.

Here we developed a new experimental method that circumvents the need for complex analysis and instead provides the information as measured. With this technique, we can make a *direct image* of the active orbital and determine what the atomic-like object looks like in a real solid [1, 2]. The method, *s*-core-level non-resonant inelastic X-ray scattering (*s*-NIXS), relies on high momentum transfer in the inelastic scattering process, which is necessary for dipole-forbidden terms ($s \rightarrow d$, $s \rightarrow f$) to gain spectral weight. Moreover, we are able to utilize *s*-NIXS for the study of the excited states [3]: by obtaining images we can identify the orbital character of those excited states, which in turn facilitates the extraction of important energy parameters in strongly correlated materials.

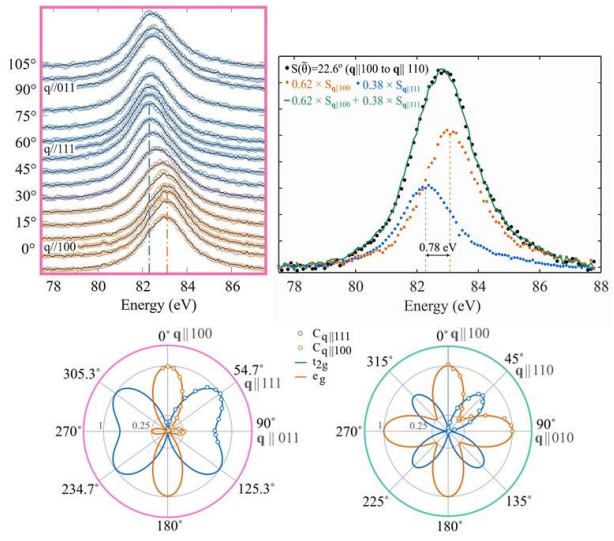
The figure below [1] displays a polar plot of the integrated intensity of the M_1 ($3s \rightarrow 3d$) NIXS spectra of a NiO single crystal as a function of the orientation of the crystal with respect to the direction of the transferred momentum. The data fall on top of the projections of the orbital shape of the Ni^{2+} ion which has the $^3A_2$ $3d_{(x2-y2)}3d_{(3z2-r2)}$ hole density, thereby demonstrating the accuracy and power of the new experimental method.





We have applied *s*-NIXS to resolve the long-standing discussion about the origin of the Ising magnetism in the frustrated $Ca_3Co_2O_6$ antiferromagnet and about the charge and spin states of its two Co sites. The *s*-NIXS data as shown in the figure above [2] reveal unambiguously that the Co in the octahedral site has all of its holes in the e_g shell and is thus in the non-magnetic low-spin 3+ configuration. The Co in the trigonal prismatic site carries all the magnetism of the material and its high-spin 3+ state has the peculiar characteristic that the sole minority-spin electron occupies the d_2 orbital, which then naturally explains the very strong Ising magnetism since the d_2 orbital has the largest orbital moment possible for a *d* system, namely 2 μ_B.

In terms of spectroscopy, we have been able to disentangle the *s*-NIXS spectra into their components and identify their orbital character by our imaging technique [3]. The figure below shows for α-MnS that the low energy component has the t_{2g} shape and the high energy e_g. The *s*-core-hole final states thus carry the $^5T_{2g}$ and ^5E_g multiplet constituents of the $3d^6$ configuration that can be reached from the high spin Mn $3d^5$ ground state. The energy splitting directly reflects the octahedral crystal field splitting 10 Dq that the Mn ion experiences. We were also able to show that we can make use of the well-established Sugano-Tanabe-Kamimura diagrams for a quantitative analysis of the spectra, since the *s*-core-hole does not add an extra orbital angular momentum component. This constitutes another major simplification in comparison to the analysis of $L_{2,3}$ and $M_{4,5}$ spectra which are the standard in transition-metal and rare-earth research.



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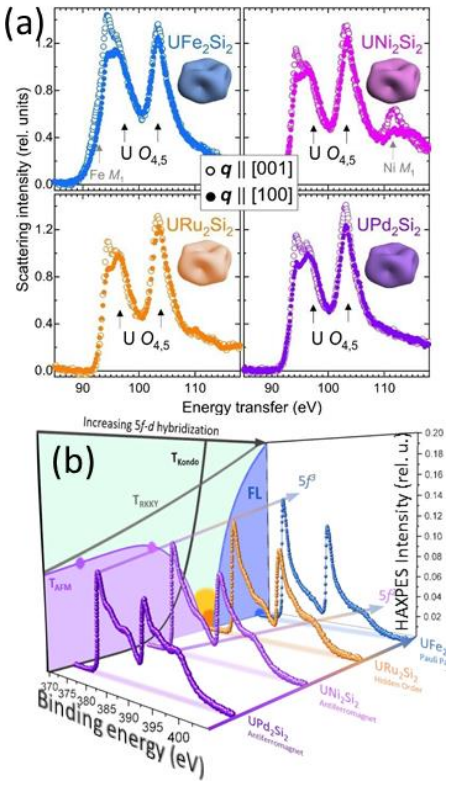
See also: https://www1.cpfs.mpg.de:2443/PCM_01 for a more comprehensive explanation.

Orbital physics in f-electron systems (Tjeng):

In close collaboration with Andrea Severing from the University of Cologne, we have continued our study of the electronic structure of 4*f* and 5*f* electron materials. We have utilized NIXS beyond-the-dipole-limit, XAS down to mK (!) temperatures, and high-resolution RIXS to unveil the orbital degrees of freedom in the *f* systems.

The interplay of band-formation and electron-correlation effects in uranium heavy fermion compounds is an intellectual challenge. The question is, should the modelling start with band theory or a local atomic approach? This has been particularly troublesome because neither the presence nor the symmetry of the active atomic-like states was so far experimentally accessible nor are there generally accepted numbers for the filling of the 5*f* shell. In the present study unexpected insight has been gained from advanced spectroscopies on isostructural members of the UM_2Si_2 family with different properties. We studied the Pauli paramagnet UFe_2Si_2, the hidden order compound URu_2Si_2, and the large-moment antiferromagnets UNi_2Si_2 and UPd_2Si_2.

We applied beyond-dipole NIXS since it is the only experimental method that has the ability to unambiguously detect (if present) atomic-like multiplet states in U intermetallics. This is related to the fact that beyond-dipole transitions reach final states with higher quantum numbers and thus lower energies so that they are more excitonic and suffer less from hybridization with continuum states. What we found [4] is that all four compounds share the same spectral features, see figure on the top of next page, leading us



to conclude that they all are in a singlet ground state of either the Γ_2 symmetry or the Γ_1 of majority $J_z = |+4\rangle \pm |-4\rangle$. We can also show that a quasi-doublet made of the two singlet states can induce the large-moment magnetism as in UNi_2Si_2 and UPd_2Si_2, as well as the hidden order in URu_2Si_2. Using hard X-ray photoelectron spectroscopy (HAXPES), see figure above, we observe a systematic increase of the U $5f^3$ weight from Pd→Ni→Ru→Fe, indicating increasing itineracy. Combining the NIXS and HAXPES, we can propose an effective Doniach phase diagram which provides a systematic understanding of the four compounds and also why the Ru compound becomes antiferromagnetic with large moments under pressure.

Using beyond-dipole NIXS we were also able to pinpoint the orientation of the ground state orbital in $CeCoIn_5$ [5], which is relevant to determine which In site plays the key role for the superconductivity.

Extending the measurement temperature range towards the mK range, we have studied the orbital ground state in $CeCu_2Si_2$ [6] with M-edge XAS and found that very little changes from well below the superconducting transition at T_c = 600 mK to well above the Kondo temperature T_K = 20 K, thus making any scenario of orbital switching (as proposed in the literature) highly unlikely. Here we also access the possibility for a multiorbital ground state in connection with the ongoing debate about the symmetry of the superconducting order parameter.

Using high-resolution RIXS we determined the local excitations in the Kondo insulators $CeRu_4Sn_6$ [7] and SmB_6 [8]. With RIXS the Sm^{2+} intensity of Sm in intermediate valent SmB_6 can be suppressed and an almost pure Sm^{3+} RIXS signal can be obtained. We further took advantage of the large energy range of RIXS and the large number of Sm multiplets. The information about the $^5H_{5/2}$ Hund's rule ground state splitting was obtained by analyzing the $^4G_{5/2}$ multiplet at about 2.3 eV. It has the same total obital moment as the ground state multiplet but a larger splitting due to a larger Stevens factor. We found a splitting of about 20 meV between the quarted Γ_8 ground state and the doublet Γ_7 of the $^5H_{5/2}$. This result suggests that the 4*f* band width cannot be much larger than 20 meV (consistent with fractional parentage considerations) and that the search for topological surface states should not focus only on bands with strong dispersions.

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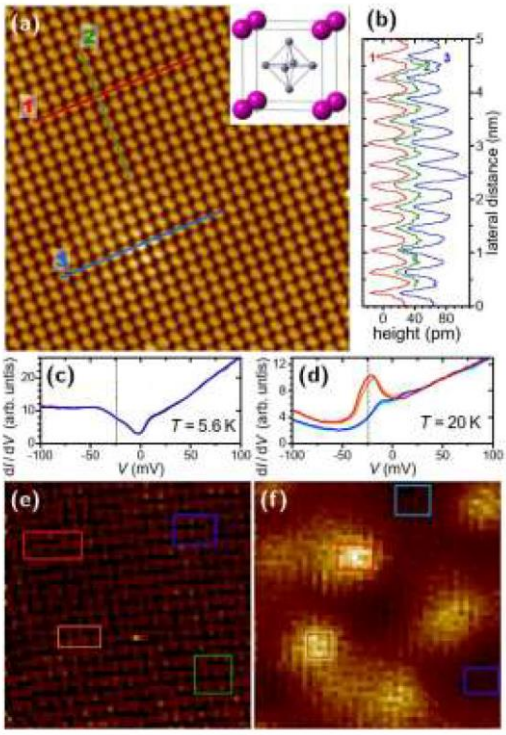
See also: https://www1.cpfs.mpg.de:2443/PCM_07 for more about our activities on *f*-electron systems.

Hexaborides: materials of diverse physics (Wirth):

SmB_6 has become of enormous topical interest because it is a candidate to be recognized as the first strongly correlated material with non-trivial topology. Here we continued our efforts towards establishing the topological nature of the surface states by studying magnetically substituted SmB_6 [9], and by employing spin-polarized tunneling. Using tunneling spectroscopy, the formation of magnetic polarons in EuB_6 [10] could be visualized for the first time. In all these studies, atomically flat surface areas and their unambiguous assignment of the surface termination on in situ cleaved samples are an indispensable prerequisite.

To scrutinize the topological nature of the surface states, we set out to study the impact of magnetic impurities in SmB_6 [9]. Such impurities are expected to break time reversal symmetry, which should strongly suppress the surface states if they are topological in nature. We investigated lightly Gd-substituted and, for comparison with a non-magnetic analogue, Y-substituted SmB_6. We found that in pure SmB_6, the characteristic STS signal for the surface



state is only slightly reduced at the impurity site, and somewhat more at an Y atom in Y-SmB_6. In contrast, there is no signature of such a peak left at the Gd impurity in Gd-SmB_6. Moreover, the suppression of this peak is very large in spatial extent, larger than in pure SmB_6 and Y-SmB_6. This has also direct implications for the resistivity: the larger areas of suppressed surface states cluster to one another already at 3% Gd substitution and impede a percolating surface conduction path. In turn, the low-T resistivity plateau – the hallmark of the metallic surface state – does not form. This clearly supports our interpretation of suppressed topological surface states through magnetic exchange interaction.

In an effort to directly visualize the electronic phase separation involved in the polaron formation, and find evidence for magnetic cluster formation in finite magnetic fields, we have investigated [10] the ferromagnetic semimetal EuB_6. It exhibits a strong CMR effect near the ferromagnetic transition temperature T_{c1} = 15.3 K, at which the magnetic polarons are believed to percolate. The magnetic polarons start forming at around T^* ~ 35 – 40 K, and finally merge at T_{c2} = 12.6 K. A clean and atomically flat surface area of EuB_6 is presented in the figure above and corresponds to a B-termination. The corrugations along the various scan lines are consistent with the lattice constant. The tunneling conductance d$I(V)$/dV is proportional to the local DOS, and at T = 5.6 K, i.e. well below T_{c2}, it does not indicate any electronic inhomogeneity. In contrast, in the paramagnetic regime at 20 K, areas of enhanced local DOS of about 3 – 4 nm in extent can clearly be recognized. This enhancement occurs at energies $V_b \approx$ -24 mV and these areas are found to coalesce at T = 15 K, i.e. between T_{c1} and T_{c2}. This clearly supports, and provides direct visualization of, the magnetic polaron scenario for the magnetic and electronic transport behavior of prototypical EuB_6.

[9]* *L. Jiao et al, Sci. Adv.* ***4*** *(2018) eaau4886.*
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See also https://www1.cpfs.mpg.de:2443/PCM_02 for more on our activities on this topic.

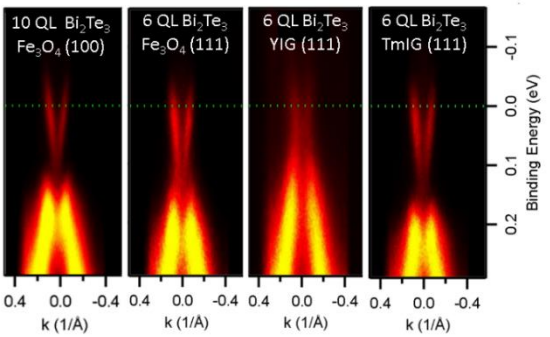
All in-situ studies of topological insulator hetero-structures (Altendorf):

Experimental research on topological insulator (TI) systems is challenging due to the inherently small number of topologically relevant charge carriers (few 10^{12}cm^{-2}). The TI properties are easily masked or even overwhelmed by topologically trivial conduction due to defects in the bulk or at the surface. Extremely high purity of the materials with less than 1% surface defects and a bulk defect concentration of well below parts per million level are required.

Our state-of-art in-house ultra-high vacuum system allows for the growth and in-situ characterization of high-quality topological insulator thin films and heterostructures. In our previous studies, we have successfully optimized the MBE growth conditions for the preparation of Bi_2Te_3 topological insulator thin films on Al_2O_3 and BaF_2 substrates that show truly bulk insulating behavior with only the characteristic Dirac-like surface states intersecting the Fermi level. Moreover, we have established a routine for sample characterization under cleanest conditions by *in-situ* RHEED, LEED, XPS, ARPES, and *in-situ* temperature-dependent resistivity measurements. In addition, we developed a reliable capping procedure with crystalline tellurium for complementary *ex-situ* measurements.

The focus of our recent work is now to investigate the influence of interfacing Bi_2Te_3 with the magnetic insulators Fe_3O_4, $Y_3Fe_5O_{12}$, and $Tm_3Fe_5O_{12}$, as well as with the topological insulator Sb_2Te_3 [11, 12, 13]. We also optimize the growth and composition of FeTe thin films [14], which build - in proximity to Te-based TIs like Bi_2Te_3 - a promising platform to study the physics of superconductor / TI heterostructures.

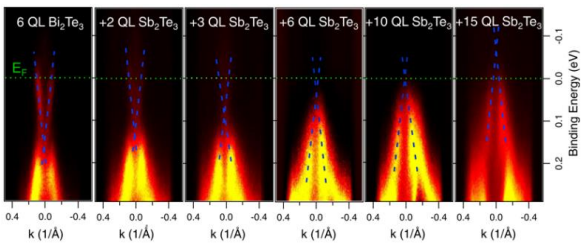
The ARPES figure on the top of the next page illustrates one of our successes, namely the epitaxial growth of Bi_2Te_3 thin films on the different



ferrimagnetic oxide substrates for which the films show systematically (1) good crystallinity, (2) insulating bulk (i.e. the top of the valence band and the bottom of the conduction band do not cross the Fermi level), and (3) well developed TI surface states that straddle the Fermi level [11, 12]. These hetero-structures also show the proximity effect of the magnetic substrate on the transport properties of the TI.

To have direct access to the TI properties by transport measurements – to observe for example the QAHE, the TI materials should ideally have a highly insulating bulk and an exposed Dirac point at or close to the Fermi level – characteristics that most TIs unfortunately do not have naturally. Interfacing different TI materials can therefore be an interesting route to engineer the band structure at the surface to the desired position. Although reports exist in the literature about such experimental approaches, it was not clear how the process of "tuning the Dirac point" actually takes place. We have therefore carried out a detailed study of the electronic structure of Sb_2Te_3/Bi_2Te_3 heterostructures from very thin (1 QL) to very thick (40 QLs) Sb_2Te_3 adlayers using *in-situ* ARPES [13].

The figure below displays a selection of the ARPES spectra, showing the top of the valence band and the surface states for different Sb_2Te_3 overlayers on Bi_2Te_3. A detailed analysis reveals that changes are taking place in the electronic structure of both the bulk and the surface, and that (uncontrolled) doping causes a rigid shift of the chemical potential. Correcting for this chemical potential shift, we found that the important modulation of the surface states with respect to the bulk states happens very rapidly for the first 3



QLs of Sb_2Te_3. For larger thicknesses, the changes are modest. Thus, the band structure engineering in Sb_2Te_3/Bi_2Te_3 can be summarized as follows: Only the very first adlayers are essential for freeing of the Dirac point from the valence band into the bulk band gap.

[11]* *V. M. Pereira et al., APL Mater.* ***8*** *(2020) 071114.*
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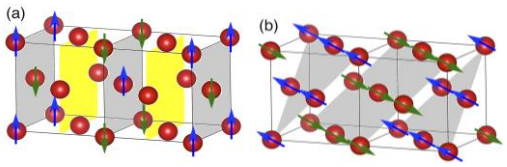
[14]* *V. M. Pereira et al., Phys. Rev. Mater.* ***4*** *(2020) 023405.*

See also https://www1.cpfs.mpg.de:2443/PCM_03 for more on the intricacies of TI heterostructures.

Magnetic frustration and quantum criticality in 4f materials (Stockert):

The influence of magnetic frustration on magnetic ordering phenomena and more generally on the ground state properties of compounds continues to be of special interest. This not only holds for finite temperature phase transitions, but frustration is also discussed as a parameter tuning continuous $T = 0$ phase transitions, i.e., quantum critical points. Combining spectroscopic methods with bulk techniques we studied a variety of strongly correlated electron systems close to quantum criticality paying special attention to frustration and the local energy scales, the latter in particular given by the crystalline electric field excitations. We investigated, e.g., the new compounds $YbNi_4P_2$ [15] and CeCoSi [16], but also studied classical heavy-fermion systems as $CeCu_{6-x}Au_x$ [17-20] or $CeCu_2Si_2$ [21]. Here we focus on the effect of geometrical frustration on the magnetism of a 4*f*-based intermetallic compound with stable 4*f* moments, namely $HoInCu_4$ [22].

Based on our previous work on the heavy-fermion compound CePdAl where frustration arises within the distorted kagome planes of this hexagonal compound, we now extended our studies to frustration in three dimensional configurations of magnetic moments, namely the face-centered cubic (fcc) lattice with a corner-sharing tetrahedral arrangement of the moments. $HoInCu_4$ crystallizes in a cubic crystal structure with an fcc arrangement of the magnetic Ho moments. We thoroughly characterized the compound by extensive neutron scattering experiments which we complemented by heat capacity and magnetization measurements [22]. Despite the large Ho moment of several μ_B, $HoInCu_4$ orders antiferromagnetically only below the very low Neel temperature $T_N = 0.76$ K. Our neutron diffraction data revealed in $HoInCu_4$ an antiferromagnetic ground state with partial frustration where half of the Ho moments remain disordered well



below T_N. This is visualized in panel (a) of the figure above, where planes of antiferromagnetically ordered Ho moments (displayed in gray) are separated by planes of disordered, frustrated Ho atoms (in yellow).

From fits an ordered magnetic 4*f* moment of 4.6 μ_B/Ho could be extracted. Such a reduced value of the ordered Ho moment agrees well with expectations for the ground state as revealed by calculations of the crystalline-electric-field (CEF) level scheme and is further corroborated by our heat capacity measurements. The disordered Ho moments show up in our neutron experiments through enhanced diffuse scattering even at lowest temperatures.

Substituting Cd for In results in $HoCdCu_4$ with antiferromagnetic order occurring at much higher temperature, T_N = 7 K, and in a fully ordered magnetic structure without any signs of frustration. While $HoInCu_4$ orders in a so-called partially frustrated type-III antiferromagnetic structure, $HoCdCu_4$ adopts a type-II antiferromagnetic structure with an antiferromagnetic alignment of the moments along [111] as shown in panel (b) of the figure above. The ordered moments in $HoCdCu_4$ are considerably enhanced with 9.5 μ_B/Ho.

The combination of heat capacity, magnetization and neutron scattering yields a consistent picture of the magnetic properties of $HoInCu_4$ and $HoCdCu_4$, in particular the characteristic energy scales like the CEF level scheme and the magnetic structures. Furthermore, our measurements give direct evidence for magnetic frustration in $HoInCu_4$. Calculations of the electronic density of states indicate their importance for tuning the ratio of nearest-neighbor to next-nearest-neighbor interactions and hence for determining the degree of frustration.

[15]* *Z. Huesges et al., K. New. J. Phys.* ***20*** *(2018) 073021.*
[16]* *S. E. Nikitin et al., Phys. Rev. B* ***101*** *(2020) 214426.*
[17]* *C. Wetli et al., Nature Physics* ***14*** *(2018) 1103.*
[18]* *S. Pal et al., Phys. Rev. Lett.* ***122*** *(2019) 096401.*
[19]* *C.-J. Yang et al., Phys. Rev. Research* ***2*** *(2020) 033296.*
[20]* *K. Grube et al., Phys. Rev. Lett.* ***121*** *(2018) 087203.*
[21]* *Z. Huesges et al., Phys. Rev. B* ***98*** *(2018) 134425.*
[22]* *O. Stockert et al., Phys. Rev. Research* ***2*** *(2020) 013183.*

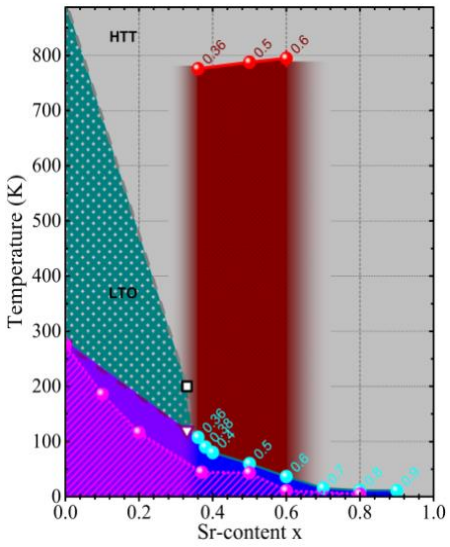
See also https://www1.cpfs.mpg.de:2443/PCM_05 for more information about our spectroscopic methods and studies on other quantum critical materials [15-22].

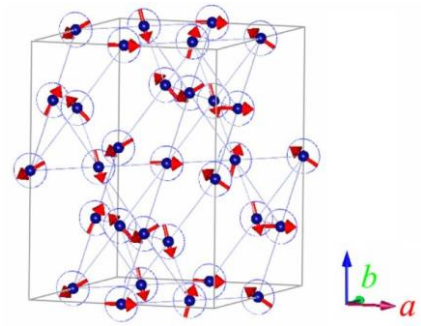
Frustration in transition metal compounds (Komarek):

As just discussed, frustrated magnetism continues to be a very active research field in solid state physics. For example, hampering the formation of simple antiferromagnetic ground states at low temperature may enable the formation of new states of matter.

In this respect it is interesting to reconsider the appearance of the so-called hour-glass magnetic excitation spectrum in high-T_c cuprate materials. Fermi surface effects or charge stripes were proposed as a possible explanation for these famous excitation spectra. However, it was discovered in the recent past that isostructural cobaltates also show hour-glass spectra although there is no Fermi surface or stripe phases in these cobaltates. We proposed an alternative scenario which involves magnetic frustration on the nano scale. We now have carried out a study on $La_{2-x}Sr_xCoO_4$ in which we address the suppression of the outwards-dispersing branches of the magnetic excitation spectra [23], a phenomenon that is not well understood on a microscopic level. Utilizing comprehensive neutron, muon and X-ray experiments, we were able to quantitatively describe the scenario in which undoped islands (with strong nearest neighbor exchange interactions J) are doped into a checkerboard charge ordered matrix (with weaker exchange interactions J' across a hole).

We also obtain the phase diagram as shown in the figure below, in which the magenta (cyan) dots indicate the magnetic ordering onset temperatures from the muon (neutron) measurements, and the red dots the onset of the checkerboard charge ordering.





In the isostructural nickelate system $RSrNiO_4$ (R = La/Nd/Y) we found quarter-integer magnetic peaks and a magnetic excitation spectrum with a steep upwards dispersion [24]. Our interpretation is that charge disproportionation occurs in this formally Ni^{3+} system and that also here disorder and frustration at the nano scale play an important role.

Recently we observed dielectric anomalies and the occurrence of a pyroelectric current at $T_N \sim 70$ K in the oxychloride Cu_2OCl_2 (melanothallite). Regarding the magnetic ions, the melanothallite crystal structure resembles the pyrochlore structure. Hence, a high degree of frustration can be expected in this system. We were now able to confirm the multiferroic properties by revealing a hysteresis within P-E-loop measurements which disappears at T_N. Moreover, we co-aligned about a dozen Cu_2OCl_2 crystals for polarized neutron diffraction and were able to show that it is a type-II multiferroic material with quite a high critical temperature of 70 K.

The magnetic structure of this novel multiferroic material is shown above. It has a cycloidal spin structure with moments spiraling in the ac plane. The electric polarization $P \propto e_{ij} \times (S_i \times S_j)$ that can be calculated for this magnetic structure according to the inverse Dzyaloshinskii-Moriya mechanism is non-zero and points in the c-direction as observed experimentally. The cycloidal magnetic structure induced by frustration is thus the driving force for the multiferroicity in Cu_2OCl_2.

We also would like to report that we continue to search for new materials with geometric frustration, and have identified $Ba_{26}Ru_{12}O_{57}$ as a promising example [26]. Currently, we are examining its physical properties.

[23]* *H. Guo et al., Phys. Rev. B* ***100*** *(2019) 014411.*
[24]* *H. Guo et al., Sci Rep.* ***10*** *(2020) 18012.*
[25]* *H. Guo et al., Phys. Rev. Materials* ***3*** *(2019) 124405.*
[26]* *J.-E. Lee et al., Crystals* ***10*** *(5) (2020) 355.*

See also https://www1.cpfs.mpg.de:2443/PCM_04 for a more detailed description.

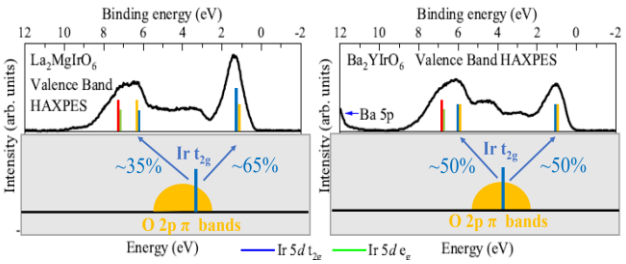
Spin-orbit interaction in 5d transition metal oxides (Chang, Hu, Tjeng):

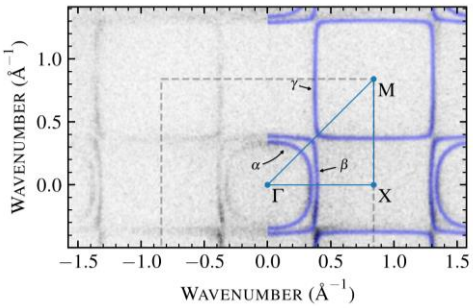
The study of transition metal oxides continues to be one of our major research activities. We co-authored in more than 75 papers in this particular research field in the 2018-2021 census period; 10 of them in journals with IF ≥ 12. These numbers do not include our papers on oxide catalysts and battery materials; those will be described in the next subsection.

In this subsection we would like to describe part of our spectroscopic work on the 5*d* transition metal oxides, and present work that has brought some nice results or unexpected insights [27-29].

The class of iridium-based oxides has attracted tremendous attention in recent years. The presence of strong spin-orbit coupling (SOC) in the 5*d* shell may lead to unexpected exotic electronic states. We focus on the layered $Sr_2Co_{0.5}Ir_{0.5}O_4$ [27], a material which we were able to synthesize in single phase and stoichiometric form without oxygen deficiency. It has very different properties than the two parent compounds Sr_2IrO_4 and Sr_2CoO_4. We found, using XAS, that the Co/Ir valences are 3+/5+, meaning that the Ir should be $J_{eff} = 0$. The analysis of XMCD data revealed, however, a deviation from the pure $J_{eff} = 0$ with an anisotropic orbital-to-spin moment ratio. It turned out that this is due to multiplet interactions being not small compared to the cubic crystal field and due to the presence of a large tetragonal crystal field. Nevertheless, we can show that the energy gap between the singlet ground state and the triplet excited state is still large and that the magnetic properties of the Ir^{5+} can be described in terms of singlet Van Vleck paramagnetism.

We have studied the electronic structure of iridates in the double perovskite crystal structure containing Ir^{4+} or Ir^{5+} using HAXPES [28]. The valence band spectra can be well reproduced using tight-binding calculations including only the Ir 5*d*, O 2*p*, and O 2*s* orbitals with parameters based on the downfolding of the density-functional band structure results. We found that, regardless of the A and B cations, the A_2BIrO_6





iridates have essentially zero O 2*p* to Ir 5*d* charge-transfer energies, see figure on the bottom of the previous page.

The consequence of this extreme covalency is that the magnetic exchange interactions become very long ranged, and thus hampering the materialization of the long-sought Kitaev physics. Nevertheless, it still would be possible to realize a spin-liquid system using the iridates with a proper tuning of the various competing exchange interactions.

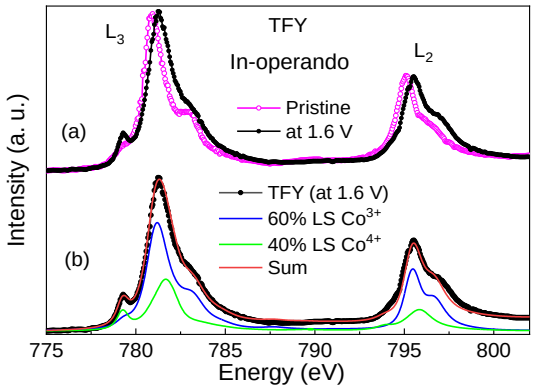
The oxide ReO_3 is quite atypical for a transition metal oxide: it is nonmagnetic, despite its *d* shell being partially filled, and it is highly metallic. In fact, it has one of the highest conductivity of all oxides, comparable with that of copper or silver. To our surprise no ARPES data have been reported in the literature so far. We suspect that the surface termination of this material is causing problems and we have therefore carried out our ARPES measurements using soft-X-rays in order to gain sufficient bulk sensitivity. Indeed, we were able to observe clear dispersions of the Re 5*d*- and O 2*p*-derived bands as well as the momentum splitting of the Fermi surface due to Re 5*d* spin-orbit interaction, see figure above.

We also found that density-functional-based band structure methods can provide an accurate description of the observed electronic states, provided that hybrid functionals are used.

[27]* *S. Agrestini et al., Phys. Rev. B* ***97*** *(2018) 214436.*
[28]* *D. Takegami et al., Phys. Rev. B* ***102*** *(2020) 045119.*
[29]* *J. Falke et al., Phys. Rev. B* ***103*** *(2021) 115125.*

X-ray spectroscopies on transition metal oxide-based catalysts and battery materials (Hu, Tjeng):

During the last three years our department has started a substantial effort to study transition metal oxide based catalysts and battery materials using synchrotron radiation. Our objective is to determine the local electronic states of the transition metal ions during the various steps of the operational process. Here we made use of our long-time expertise in soft and hard X-ray



absorption spectroscopy: expertise not only in how to experimentally obtain reliable data but especially in how to analyze the spectra in terms of configuration-interaction calculations that include the full atomic multiplet theory, in combination also with our extensive database of reference spectra. We collaborated closely with external partners who are specialists in catalyst and battery research. We selected the systems based on our experience and expectations with transition metal compounds, and in most cases we were indeed able to extract detailed information concerning the charge (valence), orbital, and/or spin state of the ions. Our research efforts in this field have resulted in more than 40 publications in the 2018-2021 census period; 26 of them are in journals with IF ≥ 12. One illustrative study is Ref. [30].

The ability to determine the electronic structure of catalysts during electrochemical reactions is highly important for identification of the active sites and the reaction mechanism. Here we successfully applied soft XAS to follow in operando the valence and spin state of the Co ions in $Li_2Co_2O_4$ under oxygen evolution reaction (OER) conditions. We have observed that a substantial fraction of the Co ions undergoes a voltage-dependent and time-dependent valence state transition from Co^{3+} to Co^{4+} accompanied by spontaneous delithiation, whereas the edge-shared Co–O network and spin state of the Co ions remain unchanged. See figure above [30]. We infer that the highly oxidized Co^{4+} site, rather than the Co^{3+} or the oxygen vacancy, is mainly responsible for the high OER activity.

[30]* *J. Zhou et al., Nature Commun.* ***11*** *(2020) 1984.*

See also https://www1.cpfs.mpg.de:2443/PCM_08 for a more comprehensive explanation and the full list of publications therein, including the 26 with IF ≥ 12.

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1.5 MPRG: Physics of Unconventional Metals and Superconductors

Group Leader: Elena Hassinger[#]

My group's motivation for research is to understand how electrons in quantum materials interact and thereby form fascinating new states of matter at low temperature. Examples of such states are topological states, unconventional superconductivity or unconventional metallic states such as non-Fermi liquids. State-of-the-art theory is not able to predict the appearance of such exotic ground states in strongly correlated systems reliably. Our aim is consequently to probe the electronic properties experimentally. In topological systems, the low energy excitations behave like relativistic particles and hence allow to investigate predictions from high energy physics. We aim at finding bulk experimental signatures for topological states of matter and connecting them to experimentally obtained information on the band structure. In heavy fermion systems, superconductivity evolves out of unconventional ground states. In these materials we look for non-Fermi liquid behavior near quantum critical points, where the quasiparticle picture should break down. We detect the electron spectrum experimentally and address the question how the unconventional normal states influence and foster the emergence of unconventional superconducting states.

A powerful and well-established way to measure the energy eigenstates of electrons near the Fermi energy is the detection of quantum oscillations in thermodynamic and transport properties [1]. Compared to newer methods like scanning tunneling microscopy and angle resolved photo emission spectroscopy, quantum oscillations provide an exquisitely sensitive spectroscopy. However, their measurement is notoriously difficult in materials with strong electron correlations, basically because the signal decreases exponentially with impurities and very low temperatures are needed when the effective masses are large. Hence high signal to noise ratios and very low temperatures are needed, as well as samples of very high quality.

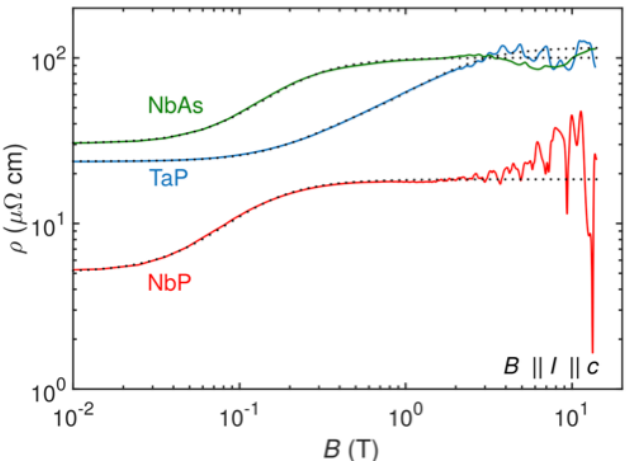

Fig. 1: Colored lines show the longitudinal magnetoresistance of the Weyl semimetals NbAs, TaP and NbP for magnetic field B and current I along the crystallographic c-axis at T = 2 K. The chiral anomaly would lead to a negative magnetoresistance, which is not observed. Instead, the data are perfectly explained by the expected orbital magnetoresistance (dotted black lines). [8]

Therefore, our research group has developed high-sensitivity techniques for the detection of quantum oscillations: magnetic susceptibility, torque, and resistivity at very low temperatures. Additionally, we are one of the few groups in the world with a setup to measure thermal conductivity down to 30 mK. We are currently running three low-temperature instruments: 1. A dilution refrigerator with a 15 T/17 T magnet with new high-sensitivity torque, resistivity or susceptibility setups (group investment in 2015), 2. a He3 system with a 16 T magnet for electrical and thermal transport (group investment in the end of 2020, in build-up phase) and 3. a dilution refrigerator with a 8 T magnet for millikelvin thermal conductivity (equipment from the PQM department run by my group since 2015).

In the last years, we have addressed scientific questions mainly in three areas. References to recent papers of our group (2018-2021) are highlighted by a * in the list of references.

Topological semimetals

https://www1.cpfs.mpg.de:2443/MPRG_02

In the area of topological semimetals [2], our main objective is to find bulk experimental evidence for Weyl fermions. Because such signatures are only expected if the Fermi energy is close to the topologically protected band crossing points [3], the so-called Weyl nodes, we first established the Fermi surface of Weyl semimetals TaAs and TaP via quantum oscillations. The results show that in TaAs, Fermi surface pockets are penetrated by an integer flux

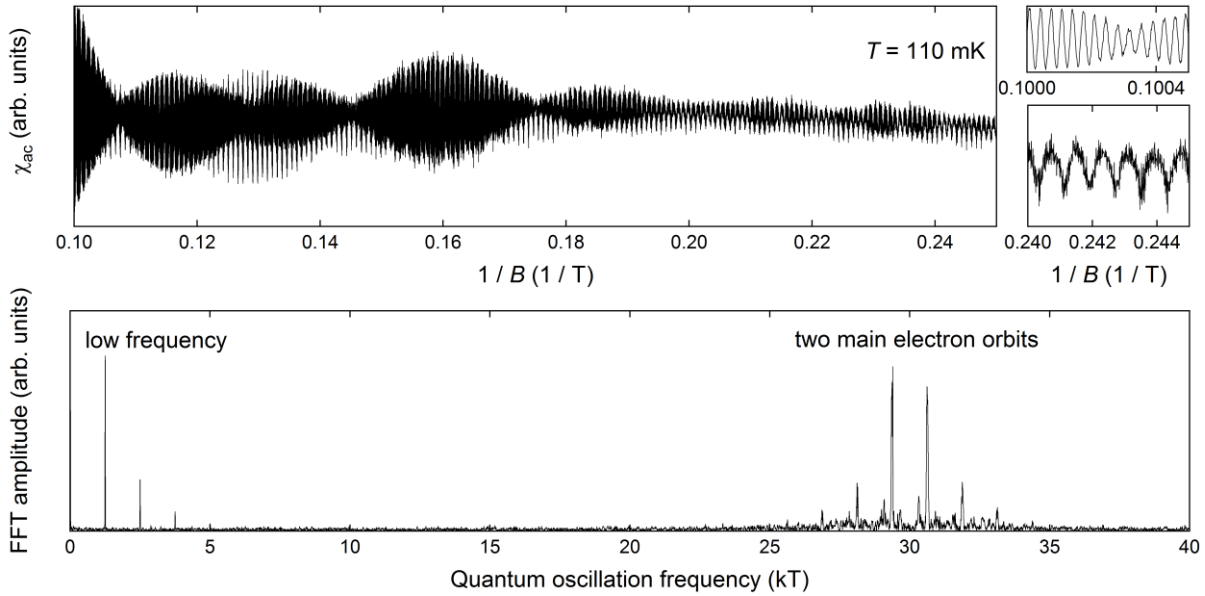

Fig. 2: Top: Quantum oscillations in $PdCoO_2$ measured by the modulation field technique and zoom on two regions highlighting the fast and slow oscillations. Bottom: Fast Fourier transform of the signal between 4 T and 10 T. Only the two peaks around 30 kT correspond to belly and neck electron orbits of the quasi-two-dimensional Fermi surface. The low frequency is suggested to be due to electron correlations. [17]

of Berry curvature, while in TaP only trivial Fermi surfaces occur [4, 5]. Recently, we submitted a Fermi surface study of the last compound of this family where this was missing, NbAs, where we discovered previously unknown Fermi surface sheets [6]. We also showed that the negative magnetoresistance discovered in this family of compounds and associated with the chiral anomaly by many authors actually is an artefact from inhomogeneous currents in longitudinal magnetoresistance measurements [5, 7]. Based on this, the first PhD student graduating from my group investigated the longitudinal magnetoresistance in these compounds achieving a homogeneous measurement current [8]. As depicted in Fig. 1, we found that the magnetoresistance actually does not show signs of the chiral anomaly. A reason might be that the dominance of the chiral contribution to conductivity is only expected when the Fermi energy is within 5 meV from the Weyl nodes [3]. Hence, it is desirable to tune the Fermi energy closer to the nodes. In a subsequent study for which supporting theoretical calculations are in progress, we could show that pressure tunes the Fermi energy towards the Weyl nodes in TaAs, i.e. in the right direction to observe the chiral anomaly. Another material where topological states are predicted to occur is $CoSb_3$ under strain. We were able to confirm the band-structure calculations underlying this prediction and therefore pave the way for measurements under strain [9]. Our plan for the future is to investigate topological semimetals at high magnetic field in the magnetic quantum limit [10]. There, the effective dimensionality is reduced and electrons become highly correlated. Field-induced ordered phases have been discovered in graphite [11] and occur in topological semimetals [12]. Magnetization and specific heat measurements in TaAs in collaboration with the high-magnetic-field lab in Grenoble (France) are ongoing but have suffered from delays due to the pandemic. As well, we plan to tune the Fermi energy through the Weyl nodes while observing the longitudinal magnetoresistance. This can be achieved by applying pressure or substituting P to As in TaAs and NbAs or As to P in TaP or NbP.

Unconventional metals

We investigated the unconventional metallic state in the high-conductivity metallic delafossites $PdRhO_2$ and $PtCoO_2$ via quantum oscillations [13, 14, 15]. We could establish the Fermi-surface topography of $PtCoO_2$ up to high-order warping parameters [15]. One of the interesting aspects in $PtCoO_2$ was the appearance of a low quantum-oscillation frequency already observed in the sister compound $PdCoO_2$ [16] but not in $PdRhO_2$ [13]. This work has triggered interest by theorist colleagues and seems to be evidence for strong electron correlations in quasi-two-dimensional metals [17]. This will be investigated further by measuring the precise temperature dependence making use of the enhanced resolution of our modulation technique setup. An example of current data on $PdCoO_2$ from this technique is given in Fig. 2.

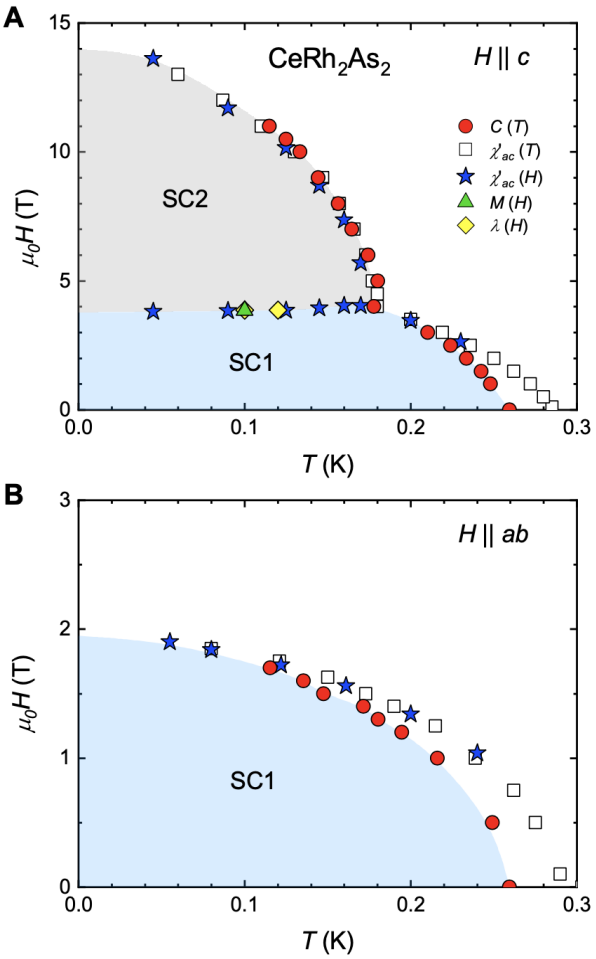

*Fig. 3: Superconducting phase diagram of $CeRh_2As_2$ for magnetic field applied along the c-axis (**A**) and in the plane (**B**). [21]*

We have also studied the thermopower of two Eu intermediate valence systems, which reveals a new mechanism for thermopower from a valence gradient across the sample induced by a temperature gradient [18]. More details on this are given in the highlight https://www1.cpfs.mpg.de:2443/MPRG_03.

Unconventional superconductors

Unconventional superconductivity is one of the most intriguing phenomena in condensed matter physics and it is at the core of our group's interest. New pairing mechanisms and unconventional pairing symmetries are still actively sought. We have contributed to this area through the study of three different materials. Primarily, an outstanding candidate for unconventional superconductivity is the material UBe_{13} since the superconducting state evolves out of a non-Fermi liquid state. However, crystal quality in this compound has always hindered a determination of the intrinsic properties and hence the physical understanding of this superconductor. In a first step, we contributed to a project in Juri Grin's department to characterize how aluminium impurities and annealing affect the physical properties of single crystals, namely the specific heat, the lattice parameter and the lattice itself [19]. Now, the plan is to collaborate with the group of E. Svanidze to work on polycrystals that do not contain Al, using the microstroucturing techniques outlined in her report (section 1.7) and the collaborative activity on microstructuring described in section 1.12.

We also contributed to a study led by the group of Philip Moll which shows that strain in micro-structured $CeIrIn_5$ single crystals can be used to control superconductivity [20]. Due to the small size of the samples, low-noise measurements in our cryostat were necessary to exclude a shift of T_c caused by noise-induced heating.

The new superconductor $CeRh_2As_2$ that was recently discovered at our institute by Seunghyun Khim of the Physics of Quantum Materials department represents a large part of our current activity. We revealed that this compound has an extraordinary phase diagram showing a huge critical field and a phase transition inside the superconducting state for a magnetic field applied along the crystallographic c-axis (see Fig. 3A). For fields in the plane, the phase diagram is more conventional but with an enhanced critical field (Fig. 3B). The submitted manuscript [21] is under consideration at *Science*, where it was recommended for publication. Such multiphase superconductivity is extremely rare in nature. In collaboration with Daniel Agterberg (U. Wisconsin) and Philip Brydon (U. Otago), leading theorists in Rashba superconductivity, we could identify the possibility that this transition is one from an even parity to an odd parity superconducting state, induced by spin-orbit coupling due to local symmetry. As such, it appears to be the first compound where an odd parity superconducting state can be realized in a spin singlet pairing channel. More details on this work can be found in this web report: https://www1.cpfs.mpg.de:2443/MPRG_01. Although this is unpublished work, I have been invited to 5 colloquia and seminars to present it, and 4 theoretical publications motivated by our findings have already appeared on the arXiv. Additionally to the extraordinary superconducting state, the compound also shows signatures of quantum criticality and multipolar order, two other fascinating phenomena. To explore these further, thermodynamic signatures in the full magnetic field-temperature space were measured and are being written up in collaboration with Seunghyun Khim and Manuel Brando (https://www1.cpfs.mpg.de:2443/PQM_04).

Our group aims at using $CeRh_2As_2$ as a new platform to study the interplay of strong correlations with locally broken inversion symmetry and the subsequent Rashba interaction and their effect on superconductivity and other orders. In the near future, we plan to investigate the angular dependence of the phase diagram. Since the phase diagrams for the two field directions are so different, it is of crucial importance to see how this anisotropic behavior connects for intermediate angles. At higher fields, an additional unidentified phase transition occurs which we will also follow as a function of field angle. The influence of impurities on the superconductivity is a parameter that gives information on the pairing mechanism and will be investigated. On top of that we have ongoing collaborations on NMR and band structure calculations on this compound.

Technical developments

The next technical development, the integration of the susceptibility/modulation-field setup into a piston-cylinder pressure cell, has been delayed by the number of exciting ambient pressure projects, but is now one of the next steps. Pressure opens up a wide parameter space and will for example enable the determination of the Fermi surface of MnSi over its pressure-induced transition from unconventional magnetism to paramagnetism [22]. We also plan to implement uniaxial strain in our cryostat for quantum-oscillation detection and magnetic susceptibility.

Thermal transport at Millikelvin temperatures

Recently, a new focus of our group has been to use thermal conductivity down to mK temperatures for the investigation of electronic, magnetic and possibly topological excitations in superconductors and metals but also in magnetic insulators. Thermal transport is a means to probe excitations that do not contribute to the electric transport. After a longer repair phase, we now have results of thermal conductivity down to 50 mK in the Tomonaga-Luttinger liquid candidate material $YbAlO_3$ [23]. We find that in zero field, the thermal conductivity is dominated by phonon conduction, but phonons are scattered by magnetic excitations near the magnetic ordering temperature. In the intermediate field range, where deconfined spinons are expected, an additional conduction channel seems to appear, probably from magnetic excitations that might be bespoke spinons. For the highest fields and temperatures, spins are polarized and the scattering of the phonons is drastically reduced, leading to an increase in the thermal conductivity. Overall, the measurements show an extremely strong influence of magnetic excitations on the thermal conductivity, both as a source of additional scattering for phonons as well as source of an additional conductivity contribution. These data will be completed and analyzed in detail in the coming year.

In a nutshell, my group uses extreme conditions such as low temperature, high magnetic fields and high pressure to investigate emergent ground states of correlated and topological electron systems.

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1.6 MPRG: Nanostructured Quantum Matter

Group Leader: Johannes Gooth[#]

Our group's main research interest is to experimentally explore quantum matter with and without interactions from the macro to the nanoscale, where quantum effects give rise to unusual electronic, thermoelectric and thermal transport properties. Examples are quantum-Hall physics in three-dimensional metals and table-top experiments for testing relativistic quantum field theories in topological materials. In the following, we highlight these two research directions and also give a perspective on future ideas.

Quantum-Hall physics in three dimensions (in collaboration with Solid State Chemistry)

When a two-dimensional (2D) electron system is exposed to a strong magnetic field B, its longitudinal resistance R_{xx} vanishes and its Hall conductance G_{xy} is given precisely by a combination of fundamental constants: $G_{xy} = e^2/h$ (where e is the elementary charge and h is Planck's constant). This is the quantum Hall effect (QHE). Its discovery in 1980 marked a turning point in condensed-matter physics. The quest for understanding the precise quantization of G_{xy}, even in a macroscopic, disordered, and irregularly shaped sample has elucidated many important and fundamental aspects of quantum physics, and has deepened our understanding of interacting electron systems. These efforts culminated in a new international system of units based on fundamental constants, and has given topology a central role in condensed matter physics. However, despite enormous theoretical and experimental efforts, one of the central questions has remained open ever since.

What happens to the QHE in three dimensions?

While the QHE is traditionally considered to be a purely 2D phenomenon, experiments over the past 40 years on different types of three-dimensional (3D) samples, ranging from coupled stacks of 2D electron systems to isotropic 3D metals, have also shown some of the remarkable aspects associated with the QHE, in particular plateau-like features in the Hall conductivity σ_{xy} accompanied by minima in the longitudinal resistivity ρ_{xx}. However, since ρ_{xx} remains always finite in these systems and a quantitative relation between σ_{xy} and e^2/h could not be established, the role of quantum-Hall physics in three dimensions is still unclear to date. Our group sets out to give a new perspective on this long-standing puzzle and to settle the role of quantum-Hall physics in three dimensions on the basis of our recent experiments.

In simple terms, electrons in 2D metals subject to a strong magnetic field B are forced to move on curved orbits with a discrete set of energy eigenvalues - the Landau levels (LLs). At sufficiently large B, where only a few LLs are occupied, 2D electron systems enter the quantum Hall regime. When the Fermi level E_F of the system lies between two LLs, the samples are characterized by a fully gapped electronic spectrum in the bulk and dissipationless current-carrying one-dimensional (1D) edge states, leading to a quantized Hall conductance $G_{xy} = N \cdot e^2/h$ and a vanishing longitudinal resistance $R_{xx} \approx 0$. Here, the LL index N counts the number of LLs below E_F. By increasing B, the LLs shift through E_F one after the other, crossing from one distinct insulating phase to another, which gives rise to the characteristic plateaus in $G_{xy}(B)$.

The crossover between insulating phases in the quantum-Hall regime does not break any additional symmetries and can hence not be understood within Landau's traditional view of phase transitions. Instead, the different insulating phases in a 2D system in a strong magnetic field define topologically distinct states. Each of these states is characterized by the value of its topological invariant, the Chern number. The latter is defined as the integral across the whole Brilloin zone (BZ) over the Berry curvature Ω of each occupied LL: $C = N \cdot 1/(2\pi)^2 \cdot \iint_{BZ} \Omega \, dk_x dk_y = 0, 1, 2, 3, \ldots$, where k_x, k_y are the x and y components

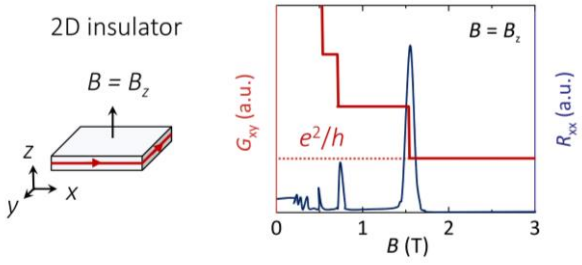

Fig. 1: Quantum-Hall physics in two dimensions. The left panel displays a 2D electron system in a magnetic field applied in the z-direction ($B = B_z$). The right panel shows a sketch of the Hall conductance G_{xy} (left axis, red) and longitudinal resistance R_{xx} (right axis, blue) as a function of B_z. Here, e is the elementary charge and h the Planck constant.

of momentum. The significance of these considerations for the interpretation of the QHE is that G_{xy} is, in fact, directly linked to the Berry curvature of the Bloch states, specifically as $G_{xy} = e^2/h \cdot \sum_{occ} 1/(2\pi)^2 \iint_{BZ} \Omega \, dk_x dk_y$, and, hence, to their topological properties. Consequently, the transitions between the different insulating phases in the quantum Hall regime have to be understood in terms of topological quantum phase transitions.

Even electron-electron interactions do not destroy the topological character of the QHE. When strong electron-electron interactions become important, the bulk develops additional correlation gaps that can be viewed as rational fractional fillings of a LL that manifest themselves in the fractional quantum Hall effect (FQHE).

In three dimensions, however, the situation is different. Instead of fully gapping the bulk energy bands of a 3D metal, high B confines the electron motion on spiral trajectories leading to a set of Landau bands (LBs) that still disperse in the field direction. Consequently, the system remains a metal, which spoils the topological argument, i.e. the quantization of G_{xy}. This observation has led to the general conjecture that quantum-Hall physics is exclusive to two dimensions.

Efforts to extend the QHE to 3D systems, therefore, usually involve the introduction of a characteristic length scale λ that transforms the original 3D system into a stack of spatially separated 2D quantum-Hall layers, stacked along the magnetic field direction either in real space or in momentum space. This effectively reduces the problem to the parallel conduction of decoupled 2D electron systems, each of them being in the quantum-Hall regime. In a seminal work, Halperin proposed that the signatures for the QHE in such a multilayer system are a quantized 3D Hall conductivity $\sigma_{xy} = N \cdot e^2/h \cdot K_z/2\pi$ and a vanishing longitudinal resistivity $\rho_{xx} \approx 0$, where K_z is the reciprocal lattice vector corresponding to the period λ of the multilayer stack along B. Importantly, for this generalization of the QHE to be observable, the system must be fully gapped in all directions.

Much effort has been devoted to search for this generalized version of the QHE, for example, in semiconductor multilayer superlattices, organic Bechgaard salts, η-Mo_4O_{11}, n-doped Bi_2Se_3, graphite, $EuMnBi_2$ - including the search for correlated Hall states. While all of these systems show plateaus in σ_{xy} accompanied by minima in ρ_{xx}, the essential signatures for the multilayer QHE – a *quantized* σ_{xy} that scales with K_z and a *vanishing* longitudinal resistivity ρ_{xx} – have not been observed in any of them. Plateau-like features in σ_{xy} accompanied by minima in ρ_{xx} alone are, however, not exclusive characteristics of layered materials and have, in fact, also been observed in 3D materials without any additional superstructure. These include also signatures of fractional LL fillings in some of them. Examples are small-gap semiconductors, such as InAs and InSb, or semimetals such as Bi, NbP, HgSe as well as high-T_C superconductors such as YBCO. While ρ_{xx} in these materials can be explained in terms of dispersive 3D LB and the Shubnikov-de Haas (SdH) effect, the observed σ_{xy} reminiscent of the QHE is quantitatively not understood to date.

Very recently, such signatures have also been observed in the 3D Dirac semimetals $ZrTe_5$ and $HfTe_5$, where the Hall plateaus were originally believed to arise from a charge density wave. This scenario is, however, in contrast with thermodynamic and thermoelectric experiments on $ZrTe_5$ by our group [1] that did not reveal any signatures of a field induced charge-density wave transition. Instead, we showed that the plateaus in σ_{xy} in $ZrTe_5$ scale with $e^2/h \cdot 2k_{F,z}/2\pi$ in the quantum limit, where $2k_{F,z}$ is twice the Fermi wavevector of the electrons in the lowest LB along B, which relates to the characteristic length scale $\lambda = 1/(k_F/\pi) = \lambda_F/2$. In such a scenario, $K_z = 2k_{F,z}$ is given by the electronic band structure rather than by a spatial extent of the system's dimensional reduction. Importantly, this indicates that σ_{xy} deeply roots in physics intrinsic to the bare electron system. The observed scaling relation can be understood considering a simple model [2]: Analogous to two dimensions, σ_{xy} of a 3D material is linked to the

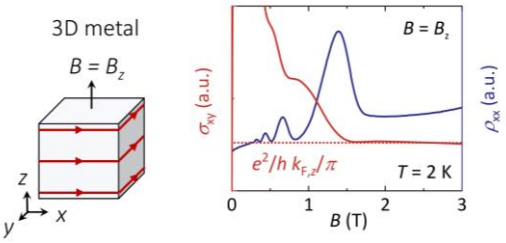

Fig. 2: Quantum-Hall physics in three dimensions. The left panel displays a 3D electron system in a magnetic field applied in the z-direction ($B = B_z$). The right panel shows σ_{xy} (left axis, red) and longitudinal resistivity ρ_{xx} (right axis, blue) of a $ZrTe_5$ sample as a function of B applied the z-direction at 2 K. The data is taken from Ref. [1]. Here, $\lambda_{F,z}$ denotes the Fermi wave vector of the 3D system along B. The main goal of this perspective is to establish this version of the Hall effect in generic 3D metals as a close 3D relative of the QHE in 2D systems.

Berry curvature as $\sigma_{xy} = e^2/h \cdot \sum_{occ} 1/(2\pi)^3 \iiint_{BZ} \Omega \, dk_x dk_y dk_z$. The results of the $dk_y dk_z$-integrals, obtained in the 2D case, can then directly be inserted into this equation, providing a direct link to the QHE. Consequently, the Hall conductivity in three dimensions is the sum of conductance quanta over all occupied LBs and wave numbers along k_z, $\sigma_{xy} = e^2/h \cdot \sum_{occ} 1/(2\pi) \cdot C \cdot \int_{BZ} \Omega \, dk_z = e^2/h \cdot C \cdot \sum_{occ} 2k_{F,z,N}$, where $k_{F,z,N}$ is the momentum vector in the magnetic field direction of the Nth LB. While $k_{F,N}$ for each LB at E_F will in general be different, specifically, in the quantum limit, the 3D Hall conductivity always becomes $\sigma_{xy} = e^2/h \cdot k_{F,z}/\pi$. While $k_{F,z}$ usually depends on B, a characteristic feature of Dirac systems is that the lowest LB does not shift with the magnetic field, leading to a B-independent Hall plateau in the quantum limit. Importantly, the model also captures SdH oscillations with minima in $\rho_{xx}(B)$ at the center of the Hall plateaus and a *finite* $\rho_{xx}(B)$ for all magnetic fields, due to the gapless LB structure along k_z.

Our recent experiments on $ZrTe_5$ [1], $HfTe_5$ [3], InAs [4], graphite (unpublished), $SrSi_2$ (unpublished) and GdPtBi (unpublished) provide evidence for that the obtained scaling relation of σ_{xy} does hold for generic 3D metallic materials and, hence, for quantum-Hall physics in 3D electron systems – including correlated states related to the FQHE in $HfTe_5$ [3].

Because our model itself does not include any specific assumptions about the band structure, a natural question to ask in the future is, whether other remarkable aspects of the QHE in two dimensions also manifest themselves in 3D systems [5]. For example, are there 2D surface states parallel to B, analogous to the 1D edge states of a 2D system? In which sense can the LB crossings with E_F in 3D electron systems be connected to quantum phase transitions despite their overall metallic nature? And what happens to the QHE at the transition between two and three dimensions? All of these questions will be addressed by our group in the next few years.

Table-top experiments for relativistic quantum field theories (in collaboration with Solid State Chemistry)

Topological materials have been essential to discover analogs of fermionic elementary particles and to test fundamental laws predicted by quantum field theory in the context of high-energy physics. Considering the richness of physics that electrical transport in topological bulk materials have recently been provided, our group has continued to unravel signatures of field theoretical predictions in topological semimetals.

In a first experiment, we were searching for signatures of axions. [6] Axions are elementary particles that have long been known in quantum field theory, but have not yet been observed in nature. However, it has been recently understood that axions can emerge as collective electronic excitations in certain crystals, the so-called axion insulators. An axion insulator is a correlated topological phase, which is predicted to arise from the formation of a charge-density wave in a Weyl semimetal—that is, a material in which electrons behave as massless chiral fermions. The accompanying sliding mode in the charge-density-wave phase – the phason – is an axion and is expected to cause anomalous magnetoelectric transport effects. Still, such an axionic charge-density wave had not been experimentally detected until very recently.

We observed a large positive contribution to the magnetoconductance in the sliding mode of the charge-density-wave Weyl semimetal Ta_2Se_8I for collinear electric and magnetic fields. The positive contribution to the magnetoconductance originates from the anomalous axionic contribution of the chiral anomaly to the phason current, and is locked to the parallel alignment of the electric and magnetic fields. By rotating the magnetic field, we show that the angular dependence of the magnetoconductance is consistent with the anomalous transport of an axionic charge-density wave. Our results show that it is possible to find experimental evidence for axions in strongly correlated topological condensed matter systems, which have so far been elusive in any other context [7]. The next step is to study the thermodynamics of the

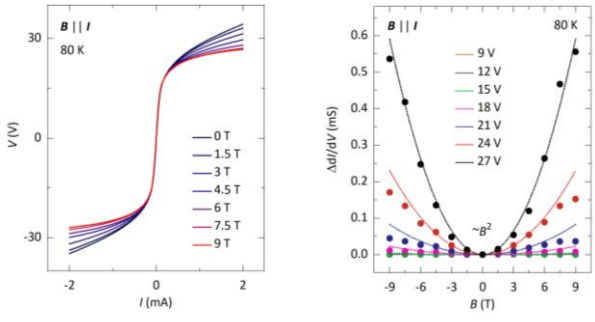

Fig. 3: Signature of an axionic charge-density wave in Ta_2Se_8I. The left panel shows the voltage–current (V–I) curve for magnetic fields parallel to the depinning field. The right panel shows the quadratic dependence of the magneto-conductance on the external field for various voltages in agreement with then prediction for an axionic charge density wave.

phase transition of the Weyl semimetal into the axion insulator in more detail.

In parallel to the search for axions, we started working on the "metric" of the electronic quasiparticles in Weyl semimetals. In other words, we studied the interplay between the electronic quasiparticles and the crystal lattice in topological semimetals. In particular, we followed two routes in preliminary works on the Weyl semimetal NbP: In a first experiment, we studied the evolution of the band structure – in particularly of the Weyl points – of NbP under the application of uniaxial strain [8]. And in a second experiment, we studied ultrasound attenuation and velocity dampening in strong magnetic fields [9]. The results show that the crystal lattice and the electronic quasiparticles in these materials are inherently connected, providing a huge playground for testing gravitational field theories in these materials in the future. Next steps are the application of uniaxial strain in $ZrTe_5$ and $HfTe_5$ as well as twisting and bending topological semimetals.

In addition, we also continued our research on the connection between temperature gradients and gravitational fields. Following our experiments on the mixed axial-gravitational anomaly in thermoelectric transport experiments, we were specifically looking for signatures of the gravitational anomaly alone in thermal transport measurements of $ZrTe_5$. We have found that a positive magneto-thermal conductivity in magnetic fields parallel to the applied temperature gradient, which diminishes when the magnetic field is applied perpendicular to the temperature gradient. This observation is consistent with the prediction of the gravitational anomaly.

Future directions

One of our main research directions in the future will be to expand our experimental activities beyond electrically charged quasi-particles. Among others, we are currently looking into neutral metallic states in excitonic insulators and quantum anomalies in spin-ladder compounds.

A second main focus in the future will be topological nanostructures. Here, we strongly collaborate with the Departments of Solid State Chemistry and Chemical Metals Science to transfer their expertise in chemical vapor transport of bulk single crystals to the vapor-liquid-solid growth of nanowires and nanoplates. The current status of this project is that we have already successfully transferred the thermodynamic model for the CVD-growth to the VLS-mechanism, have built three ovens accordingly and have grown the first Dirac semimetal Ca_3As_2 nanowires as a first test example. The nanowire device fabrication has also been developed in parallel and our cryostats are modified such that transport characterization of the nanowire devices is now possible. Next steps are to expand the VLS growth to a variety of material systems and to perform first transport experiments on the Ca_3As_2 nanowires.

Fig. 4: Furnaces for the topological nanowire growth.

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1.7 Minerva Group: Research of Exotic Actinide and Lanthanide Materials (REALM)

Group Leader: Eteri Svanidze[#]

Materials containing elements from the bottom of the periodic table often exhibit peculiar chemical and physical properties. The goal of the REALM group is to obtain a deeper understanding of *4f*- and *5f*- based compounds and alloys, which can be gained by, on one hand, a discovery of new classes of materials and, on the other hand, a comprehensive assessment of the relation between chemistry and physics in these systems.

Over the last century, both fundamental and applied research on actinide- and lanthanide-containing materials has resulted in an immense wealth of knowledge. The persistent curiosity in *4f*- and *5f*-based compounds and alloys is justified: From peculiar chemical bonding properties to unprecedented ground states – these systems still pose many open questions. Breakthroughs in all areas of actinide and lanthanide research rely on two complementary approaches – a comprehensive understanding of existing systems and the discovery of new materials.

The primary interest of the REALM group lies in a systematic search for new solid-state compounds and alloys that exhibit peculiar physical properties. For *4f*- and *5f*-based materials, a quantitative theoretical assessment, and, therefore, prediction of new materials, is often impossible, given the complexity of the relevant orbitals. One approach to finding new materials is a thorough analysis of existing systems and consequent, by identification of chemical features that are likely to result in the desired physical properties [1, 2]. By finding new materials of interest, the parameters can be refined, producing a more efficient method for the discovery of new compounds and alloys. In parallel, by pinpointing the key features, insights for theoretical research can be provided. This type of an empirical approach was successful in finding one of the heaviest uranium-based heavy-fermion systems – $U_{23}Hg_{88}$ [1] and was also applied to uranium-based superconductors [2]. However, the limited number of known uranium-based superconductors hinders its efficiency in this instance.

Another avenue in the discovery of new materials is to target specific structural motifs. In particular, we have previously shown that a beryllium-containing compound (BeAu) with a non-centrosymmetric crystal structure exhibits unconventional superconductivity [3-5]. More specifically, the two-band features of BeAu were found to be similar to MgB_2. Motivated by these results, we were interested in the design of non-centrosymmetric compounds containing beryllium and lanthanide or actinide elements.

This search resulted in the discovery of a new structure type based on the cage compound $Y_4Be_{33}Pt_{16}$ [6]. For an in-depth description of its structural and bonding features, see https://www1.cpfs.mpg.de:2443/CMS_06.

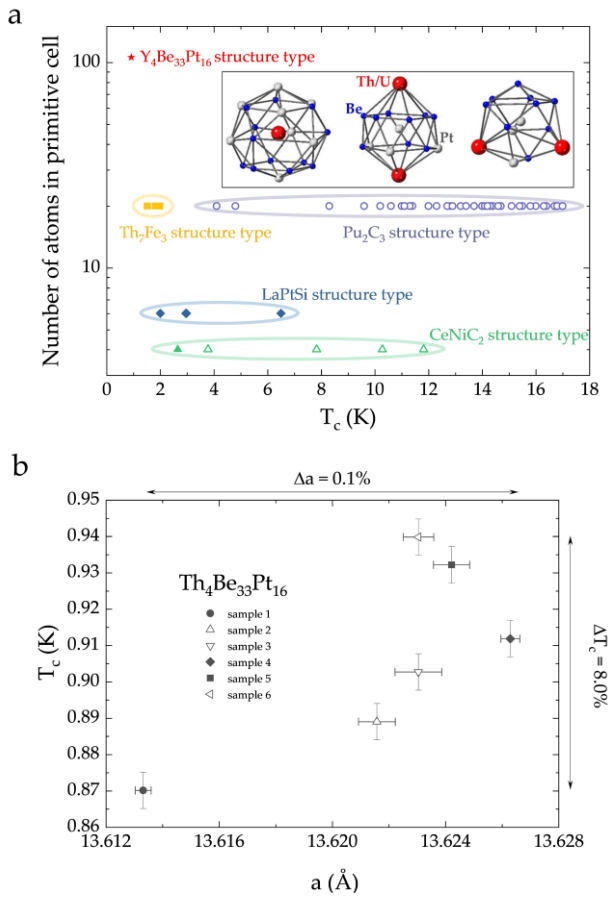

Fig. 1: (a) The number of atoms per unit cell versus the critical temperature T_c of Th-based non-centrosymmetric superconductors. The $Th_4Be_{33}Pt_{16}$ compound (red star) is the most crystallographically complex representative of this class, discovered to date. (b) $Th_4Be_{33}Pt_{16}$ exhibits a strong variation of the superconducting temperature T_c, while corresponding change in the lattice parameter a is rather modest [7].

Overall, fifteen lanthanide- and two actinide-based compounds were found to crystallize in the $Y_4Be_{33}Pt_{16}$ structure type. Their physical properties are highly diverse, ranging from superconductors to ferro- and antiferromagnets [6-8]. The $U_4Be_{33}Pt_{16}$ compound was found to exhibit an enhanced effective electron mass and hints of quantum critical behavior [8]. With 212 atoms per unit cell, $Th_4Be_{33}Pt_{16}$ is the most complex non-centrosymmetric thorium-based superconductor discovered to date [8], see Fig. 1a. There appears to be a very unusual relation between the crystallographic features and critical temperature, as summarized in Fig. 1b, with the nature of its superconducting pairing mechanism yet to be established conclusively.

While the possibility of using uranium-based materials for thermoelectric applications was proposed over half a century ago, very little work has been conducted in this direction in the meantime. In our recent investigation [9], we were able to show that uranium-based materials are suitable candidates for the development of thermoelectric components. In particular, toxicity and radiation hazard of these materials can be circumvented if they are used for aerospace applications, for which their operating temperature range of -100 °C – 100 °C seems well suited. Given low cost and abundance of depleted uranium, development of functional uranium-based materials is not only of fundamental interest; it can perhaps contribute to the solution of the nuclear waste problem as well [10].

The second effort of the REALM group is directed towards the development of new ways to synthesize and characterize solid-state materials. Rather frequently, the synthesis of novel systems is hindered by elemental immiscibility, high melting temperatures, as well as toxicity and reactivity of comprising elements. We address this issue by implementing novel experimental techniques, such as, for example, stabilization of metastable compounds by means of simultaneous high-temperature, high-pressure synthesis. However, for some compounds and alloys, even months and years of synthesis optimization do not result in samples of sufficient quality. We have recently shown that it is feasible to study these systems by isolating micro-scale pieces from polycrystalline bulk material, which allows to obtain their intrinsic physical properties. This novel approach allows for a more efficient classification and, consequently, a much faster identification of most promising compounds and alloys that have desired physical properties, and will be useful in elucidating the

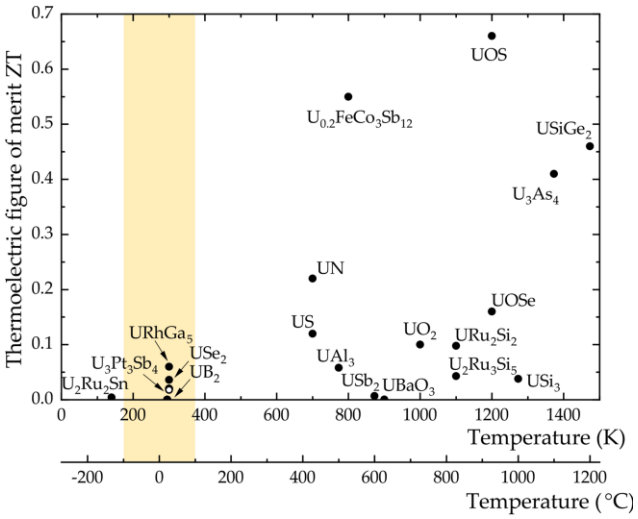

Fig. 2. Maximum value of the thermoelectric figure of merit ZT as a function of temperature for uranium-based materials. The yellow region highlights the optimal temperature range for applications of these compounds in the aerospace industry [10].

properties of UBe_{13} [11]. For more detailed information, see https://www1.cpfs.mpg.de:2443/COLL_02.

By understanding the relationship between chemical and physical properties of *4f*- and *5f*-based materials, the REALM group aims to deepen our fundamental understanding of these peculiar systems while simultaneously unveiling their potential for innovative applications.

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1.8 Emeritus Research Group: Solid State Physics

Director Emeritus: Frank Steglich

Within the last period, research was done jointly with colleagues from MPI CPfS, CCM/ZJU, IOP/CAS, LANL, Rice U., Rutgers U., Kyoto U., Toyama U., U. Augsburg, HKUST and Princeton U.

The collaboration with colleagues from chemistry has been an important part of our research. A new example is the comprehensive study of the vibrational dynamics in a family of type-I clathrates performed by the group of J. Grin, M. Baitinger et al. [1]. In the following some joint results on further materials will be discussed; for more information, see the online report at https://www1.cpfs.mpg.de:2443/EMR_01.

Kondo screening *vs* valence fluctuations. ARPES and resonant ARPES measurements by Y. Liu et al. (CCM/ZJU, Hangzhou) on the Kondo-lattice system CeBi with low carrier density indicated a well-developed $J = 7/2$ spin-orbit satellite, pointing to the action of Kondo screening [2, 3]. Yet, the $J = 5/2$ 'Kondo resonance' at the Fermi level was found to be very weak, indicating an extremely low Kondo temperature due to the low concentration of charge carriers. Upon cooling CeBi from about 35 to 10 K, a significant temperature-dependent intensity transfer was observed from the 4*f*- into the conduction-band states, accompanied by a 40 % expansion of an electron pocket of the Fermi-surface (FS) referring to the 5*d*-electrons of Ce. Within Luttinger's theorem this can be understood by a charge transfer from the Ce-4*f*-states into the 5*d*-states on neighboring Ce-sites, resulting in an increase of the Ce valency by only 1 % [2].

Destruction of the Kondo screening at magnetic quantum critical points. $CeRhIn_5$ exhibits an abrupt change of its FS volume at the quantum critical magnetic field $B_{cr}(T\rightarrow 0) \approx 30$ T, inside the antiferromagnetically ordered phase. A surprisingly wide range of FS fluctuations down to 17 T was inferred from low-temperature specific-heat results by H.Q. Yuan's group at CCM [4]. For **$CeCu_2Si_2$**, with a recently proposed modulated superconducting phase [5], a new QCP at $B = 17$ T has been explored by F. Weickert (LANL) et al. [6]. As discovered through scanning tunneling spectroscopy by S. Wirth and collaborators, for **$YbRh_2Si_2$** a moderately enhanced Fermi liquid forms at about 3 K, before at lower temperature quantum critical phenomena become visible [7]. The latter are taken as a signature of the breakdown of the Kondo effect in the zero-temperature limit. The impact of Co substitution for Rh was studied on both magnetic correlations by M. Brando's group [8] and the Seebeck effect by U. Stockert et al. [9]. P. Sun and his team at IOP/CAS (Beijing), in collaboration with Q. Si (Rice. U.) and P. Gegenwart's group (U. Augsburg) have established [10] an extended *quantum critical phase* in the low-temperature pressure-magnetic field phase diagram inside the paramagnetic phase of the distorted kagome-lattice compound **CePdAl** [11]. This quantum critical phase is sandwiched between local-moment antiferromagnetic (AF) order with *small* FS and a heavy Fermi liquid phase with *large* FS. The boundary between small and large FS is a 'Mott line' of *continuous quantum phase transitions* (or *QCPs*) at low pressure and high magnetic field and of *quantum crossovers* at higher pressure and lower field [10]. QCPs and the associated strange-metal behavior (SMB) caused by the entanglement of electron spins are frequently found in AF metals – but are commonly absent in their ferromagnetic (FM) counterparts because of the lack of spin entanglement. However, as shown for the local-moment ferromagnet **$CeRh_6Ge_4$** by H.Q. Yuan's group, along with M. Nicklas and R. Borth (MPI CPfS) as well as P. Coleman and Y. Komijani (Rutgers U.), a suitable magnetic anisotropy [12], intimately related to the pronounced anisotropy of the 4*f*-conduction electron hybridization [13] indeed causes partial spin entanglement. It paves the way for a FM QCP at moderate pressure and SMB in wide parts of the phase diagram [14]. This FM QCP appears to be of the local type as substantiated [15] by recent dHvA measurements.

Heavy-fermion superconductivity. Inelastic neutron scattering measurements performed by O. Stockert and coworkers revealed magnon excitations in non-superconducting, antiferromagnetically ordered $CeCu_2Si_2$ which are very similar to the paramagnon excitations reported earlier in superconducting samples. This observation substantiates the notion that superconductivity in $CeCu_2Si_2$ is driven magnetically [16]. Recent ARPES measurements on $CeCu_2Si_2$ by Y. Liu and collaborators (CCM, ZJU) have revealed a heavy electron band near the *X*-point and other bands with appreciable 4*f*-contribution, mainly of hole type, near the *Z*-point [17]. While the heavy electron band apparently plays the key role in forming the

superconducting condensate by singlet pairing across the nesting wavevector of its warped part [18], the scatterings between this heavy electron band and the hole bands at the *Z* point provide the inter-band contribution required by the '*d*+*d*' band-mixing pairing model introduced by Q. Si et al. [19]. This model naturally explains that in thermodynamic properties, the superconducting order parameter remains finite in the low-temperature limit, while a nodal gap exists at slightly elevated temperatures [20]. The '*d*+*d*' model relies on a multiorbital ground state, i.e., containing crystal-field states of both Γ_7 and Γ_6 symmetry. This is supported by recent measurements of the linear dichroism of linearly polarized X-ray absorption spectroscopy by A. Severing, L.H. Tjeng and coworkers which indeed suggest a ground state for $CeCu_2Si_2$ consisting of 88 % $\Gamma_7^{(1)}$ and both 6 % Γ_6 and $\Gamma_7^{(2)}$ [21]. $CeCu_2Si_2$ thus behaves as a fully gapped *d*-wave superconductor [22].

Topological effects in strongly as well as weakly correlated materials. The impact of nonmagnetic and magnetic impurities on the metallic surface states of the topological Kondo insulator **SmB_6** has been studied by S. Wirth and coworkers by combining scanning tunneling spectroscopy and transport measurements [23]. It was found that, compared to the nonmagnetic impurities, the magnetic ones are considerably more destructive to these surface states, which can be related to the different strengths of the exchange interaction. The prototypical Kondo semimetal **YbPtBi** was chosen by H.Q. Yuan et al. to demonstrate a potential modification of topological effects under the emergence of strong electron-electron correlations [24]. At elevated temperatures of order 200 K, where the 4*f*-electrons are almost stable (i.e., only very weakly Kondo screened by the conduction electrons), both a 'chiral anomaly' in the magnetoresistance as well as a topological Hall effect were detected. These observations are reminiscent of what had been published earlier for the Gd homologue where the 4*f*-electrons are stable at any temperature. In contrast, upon cooling YbPtBi, the 4*f*-electrons become unstable by the Kondo screening which leads to extremely heavy charge carriers and, correspondingly, a very small renormalized Fermi velocity v_F. Whereas at sufficiently low temperature the topological Hall effect persists, the strength of the 'chiral anomaly' ceases as it is proportional to v_F^3. Instead, a huge T^3 term ($\sim v_F^{-3}$) develops in the specific heat which reflects the flattening of the linear dispersion relation at the Weyl nodes. For the weakly correlated Weyl semimetal **TaAs**, P. Sun's group, collaborating with X. Dai (HKUST) and Z. Song (Princeton U.), observed gigantic oscillations in the longitudinal heat conductivity κ driven by the magnetic field *B* [25]. They are similar to, but more pronounced than, those found earlier by U. Stockert for NbP [26] and in antiphase with the SdH-oscillations in the electrical conductivity σ. Furthermore, they have an amplitude which exceeds that expected from σ via the Lorenz/WF relation by two orders of magnitude. These $\kappa(B)$ oscillations cannot be accounted for by any common mechanism for heat transport. Rather, they highlight a new heat-transport channel, namely by *chiral zero sound*, a collective bosonic excitation of the Weyl fermions which propagates along the field direction [25]. For the Dirac semimetal **Cd_3As_2**, the same group has found large values of the *transverse* thermoelectric figure of merit invoking the Nernst effect, which exceed significantly the values of the ordinary *longitudinal* figure of merit relying on the Seebeck effect. It is argued that the transverse figure of merit could be further optimized by tuning the Fermi energy closer to the Dirac node. For, this should cause not only an enhanced bipolar transport of compensated electronic and hole-type carriers but also an anomalous Nernst effect due to an enhanced Berry curvature [27].

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1.9 Max Planck Fellow Group: Molenkamp Group

Group Leader: Laurens Molenkamp[#]

In this report we show the current status of the projects from the Molenkamp group that are done in collaboration with groups at MPI-CPfS. These projects combine the expertise of the Molenkamp group in low temperature transport, thermoelectric transport, and molecular beam epitaxy with the wide range of expertise at MPI-CPfS and address the properties of HgTe based topological materials, delafossites, and Heusler materials.

1. Thermoelectric Properties of HgTe Weyl Semimetals:

Dirac and Weyl semimetals have attracted a lot of attention recently as they provide a linear band dispersion in three dimensions, providing us with the opportunity to study Weyl and Dirac quasiparticles that are predicted for a long time but not yet observed in nature. In this project we study the thermoelectric transport properties of Weyl semimetal realized in compressively strained MBE-grown HgTe films. Due to the outstanding quality of our HgTe layers, the charge carrier density can be controlled by gating in such a way that the Fermi energy is located in the vicinity of the Weyl point, a situation which is hardly to be realized in other topological semimetals [1].

This allows us to study predictions related to the gravitational anomaly [2]. In a first step we have successfully measured the Seebeck coefficient in a HgTe Weyl semimetals and verified the validity of Mott's relation. Fig. 1 shows a comparison between the measured and the extracted thermovoltage, using Mott's relation applied to the conductance data.

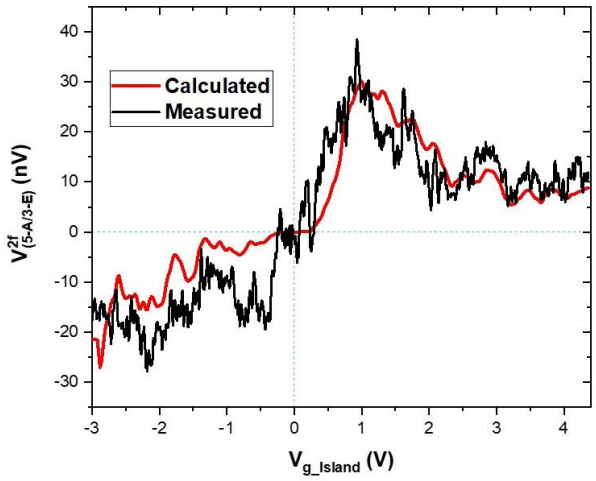

Fig. 1: A comparison between the measured and calculated thermovoltage in our samples, demonstrating the validity of Mott's relation in our system.

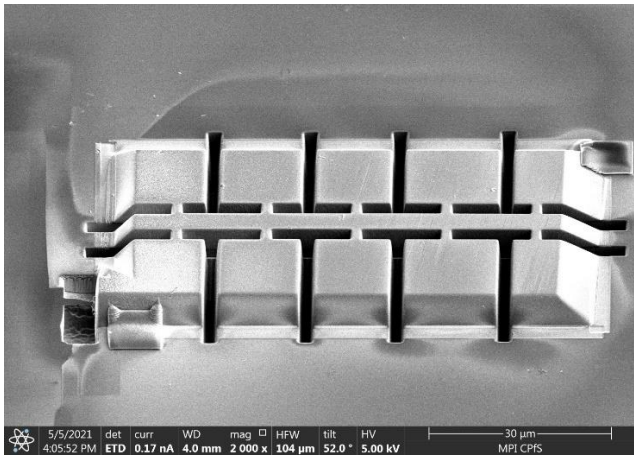

Fig. 2: SEM image of the Hall bar after FIB patterning.

This project is in cooperation with the Moessner group in MPI-PKS.

2. Thermoelectric transport in Delafossites:

We concentrate in this study on $PdCoO_2$, which is a quasi-two-dimensional delafossite metal that shows unexpected exceptional electrical properties, including ultra-high in-plane conductance [3], unconventional magnetoresistance [4], and indications for hydrodynamic electron flow. Here, we use our expertise in measuring thermoelectric transport and electron hydrodynamics in collaboration with the Mackenzie group's expertise in delafossites. The aim is to quantify hydrodynamic electron flow in this material, first reported by Moll et al. [5].

$PdCoO_2$ crystals are grown and transport devices are structured at MPI-CPfS, involving focused ion beam (FIB) techniques. Fig. 2 shows a Scanning Electron Microscopy (SEM) image of the FIB patterned device. These devices are transferred to Würzburg where gold contacts are added, using photolithography techniques, and the transport properties are measured. So far, three samples have already been fabricated, and measurement characterized successfully.

This project is carried out in collaboration with the Mackenzie group.

3. Effects of uniaxial strain on HgTe based topological materials:

Previously we have demonstrated that HgTe films can be grown to exhibit different topological states depending on the strain imposed by the substrate. Three-dimensional topological insulators (3D TI) and Weyl semimetals are created by either tensile or compressive strain, respectively. Now we are exploring the possibility to introduce the topological phase transition by applying external uniaxial strain.

The pressure cell, shown in Fig. 3, is developed at MPI-CPfS and uses piezoelectric actuators. In Würzburg this pressure cell is mounted in a flow cryostat, which reaches a base temperature of 3 K. Thus, we are able to perform low temperature transport experiments while the applied strain is changed continuously. Simultaneously, a Raman spectroscopy setup allows to monitor the applied strain directly.

This project is a collaboration with the Mackenzie group.

4. Molecular beam epitaxy of the topological superconductor YPtBi:

In this project we develop thin film growth of the half-Heusler semimetal YPtBi by molecular beam epitaxy. YPtBi is a non-centrosymmetric, topologically nontrivial superconductor [6] with strong spin orbit coupling and a critical temperature of 0.8 K [7]. Thin films of single crystal quality will allow the fabrication of lateral devices for electronic transport studies.

A dedicated molecular beam epitaxy system for YPtBi has been installed in Würzburg (see Fig. 4) and first

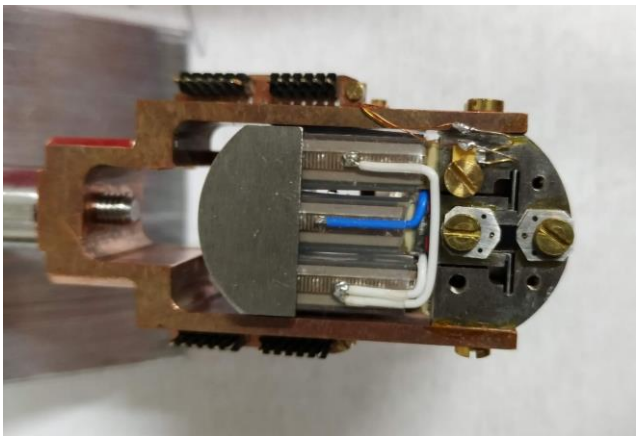

Fig. 3: Photograph of the pressure cell used to apply uniaxial strain.

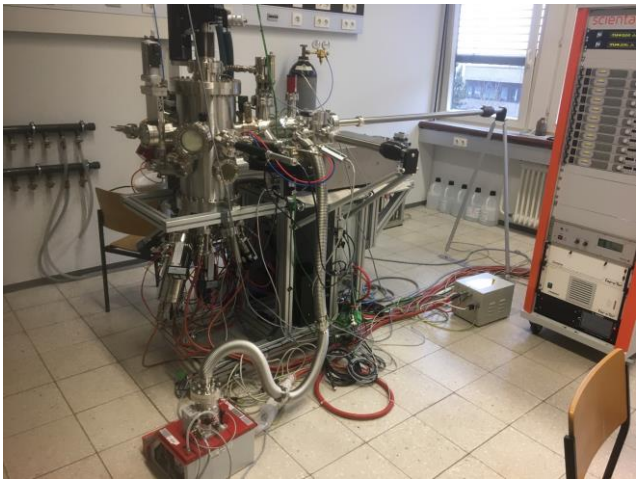

Fig. 4: Photograph of the YPtBi molecular beam epitaxy system.

samples are being grown. Since there are no well-matching substrates available for ternary YPtBi (lattice parameter 6.65 Å), we use potassium bromide (001) substrates (lattice mismatch ~0.8%). We cleave the substrates directly from bulk crystals to ensure atomically flat surfaces over distances of several µm, as confirmed by atomic force microscopy. Given the expertise of the Würzburg group with epitaxial growth of half-Heusler materials, we expect fast progress from this point.

This project is a collaboration with the Felser group.

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1.10 Max Planck Fellow Group: Ruck Group

Group Leader: Michael Ruck[#]

It is time to question the established methods of material synthesis! Is it possible to replace the abundantly used high-temperature processes with alternative low-temperature syntheses that are more energy-efficient and more controllable? Can ecological and economic aspects be combined in this way? What new (scientific) opportunities do such innovative approaches offer? We have been pursuing these challenges during the past decade and created new solutions. One of them is the so-called hydroflux synthesis.

The group of Michael Ruck at TU Dresden is working in various fields of inorganic solid-state chemistry. Besides our long-term chemical interest in low-valence compounds, clusters and coordination polymers, we are exploring new materials with interesting and potentially useful physical properties, such as low-dimensional metals, superconductors, topological insulators, frustrated magnets, or ion conductors. In part hand in hand with these topics, but also beyond, go our activities on sustainable materials synthesis. Our goal is the substitution of conventional processes by new approaches that consume less energy, are more resource-efficient and produce less waste. Besides developing the microwave-assisted polyol process for the production of active nanoscale intermetallics, we are exploring the options for "ionometallurgy". The latter envisions the access to metals or valuable chemicals in a one-pot process directly from their natural resources at temperatures below 200 °C, of course followed by the recycling of the ionic liquid that is used as reaction medium. In the following, however, we will report on our recent progress in utilizing ultra-alkaline aqueous solutions for fast and high-yield materials synthesis of (not only) metal oxides and hydroxides at moderate temperatures.

Hydroflux – Efficient Synthesis in Ultra-Alkaline Medium

The hydroflux is a unique reaction environment and stands between hydrothermal conditions and water-free salt flux, which is reflected in its name [1]. The reaction medium is an approximately equimolar mixture of water and an alkali, usually NaOH or KOH. This corresponds to a 40 to 50 molar solution. In practice, the water is completely absorbed by the solid alkali. Thereby, the melting point is dramatically reduced from 318 °C for NaOH or 360 °C for KOH to about 80 to 140 °C, depending on the specific concentration. In contrast to much lower concentrated solutions used in conventional hydrothermal synthesis, the water in the hydroflux has a very low activity and thus the pressure evolving at a typical reaction temperature of 200 °C is negligible. That we use PTFE-lined stainless steel autoclaves is not due to pressure, but to the corrosive medium and the need to prevent water loss during the reaction. At reaction temperature, the hydroflux readily dissolves many solids, including laboratory glass ware. Thus, reactions proceed rather fast. We typically use one night as reaction time. After optimization of the reaction parameters, we regularly achieve single-phase products in almost quantitative yield. The products are crystalline and contain single-crystals with sizes that can exceed one millimeter but are at least suitable for structure determination by X-ray diffraction. Some results shall demonstrate exemplarily the unexpectedly high potential of syntheses in hydroflux.

Potassium ion conductors

Oxohydroxoferrates(III) $A_2[Fe_2O_3(OH)_2]$ (A = K, Rb, Cs) were synthesized under hydroflux conditions [2]. Their structures resemble ß-alumina (Fig. 1), and they all show mobility of potassium cations at room temperature. By partial substitution of iron against iridium and thermal or mechanical treatment, the ion conductivity of a pressed pellet could be increased to the exceptionally high value of $5 \cdot 10^{-3}$ S cm^{-1} at room temperature [3]. The ion conductivity in the chiral

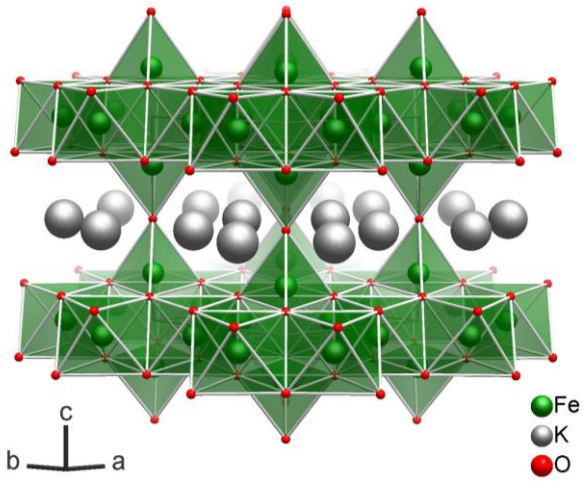

Fig. 1: Crystal structure of $K_{2-x}Fe_4O_7H_x$.

cubic labyrinth of the oxometalate $K_{12+6x}Fe_6Te_{4-x}O_{27}$ proved to be lower, despite its three-dimensional connectivity, because of a stable coordination site for potassium cations that blocks the channels.

Carbon-free precursors for functional oxides

We crystallized about 20 (new) hydroxometalates of *d* or *f* transition metals from hydroflux [4–10], among them many hydrogarnets (Fig. 2) but also layered structures. They are interesting for themselves but can also serve as carbon-free precursors to functional oxides. For example, pure Na_2IrO_3 is obtained through the thermal decomposition of $Na_2[Ir(OH)_6]$ in air, which starts already at about 220 °C [4]. Na_2IrO_3 is antiferromagnetic and shows strong magnetic frustration following the Kitaev model, which has a quantum spin liquid as its ground state [11, 12]. Similarly, $Ba_3[Rh(OH)_6]_2{\cdot}H_2O$ decomposes to the 2H hexagonal perovskite $Ba_9Rh_8O_{24}$ [5]. $CaNa[Cr(OH)_6]$ transforms via the isolable chromium(VI) intermediates $CaCrO_4$ and Na_2CrO_4 into the oxochromates(V) $Ca_5(CrO_4)_3O_{0.5}$ and $Ca_3(CrO_4)_2$ [6]. From the decomposition of rare-earth hydrogarnets $(EA)_3[RE(OH)_6]_2$ (*EA* = Sr, Ba; *RE* = Sc, Y, Ho–Lu) magnetic oxides $(EA)RE_2O_4$ were obtained [7, 8].

Redox chemistry far away from standard potentials

The parameters of the hydroflux (esp. concentrations and temperature) can be adjusted to stabilize unusual oxidation states, as the example of the manganate(V) $KSrMnO_4$ demonstrates [13]. In general, the hydroflux can be considered as an oxidizing medium. Thus, we were highly surprised to obtain large crystals of trichalcogenides K_2Se_3, K_2Te_3, or $K_2(TeSe_2)$ from SeO_2 and TeO_2 without an obvious reducing agent (Fig. 3). In this case, As_2O_3, which had initially been added for other reasons, acted as the reducing agent and was

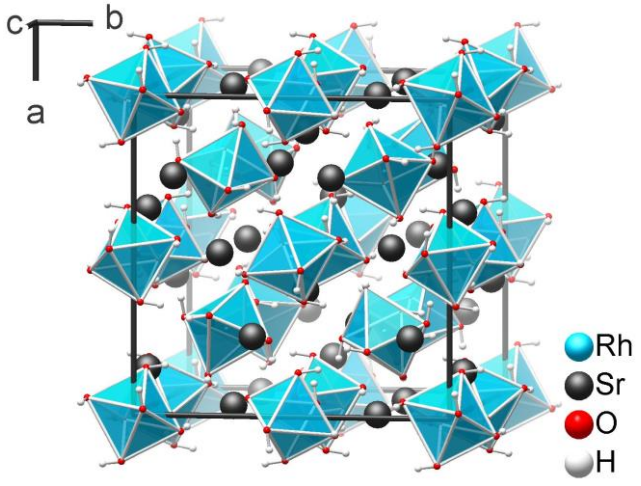

Fig. 2: Structure of the hydrogarnet $Sr_3[Rh(OH)_6]_2$.

Fig. 3: Crystals of K_2Te_3 obtained from hydroflux.

itself oxidized to arsenic(V). Arsenic is not only the electron donor but binds the oxygen atoms provided by the chalcogen dioxides in AsO_4^{3-} anions. This redox chemistry is far from what the standard potentials led us to expect, but can be rationalized by the applied ultra-alkaline conditions according to the equation 1:

$$7\ As_2O_3 + 6\ SeO_2 + 4\ K^+ + 46\ OH^- \rightarrow 2\ K_2Se_3 + 14\ AsO_4^{3-} + 23\ H_2O \quad (1)$$

The hydroflux strongly promotes the redox reaction through its high OH^- concentration on the left side of the equation as well as through its hygroscopy on the right side. The water is strongly bonded to hydroxide ions, which considerably reduces its activity. This effect also prevents the hydrolysis of the highly water sensitive trichalcogenides.

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1.11 Max Planck Fellow Group: Davis Group

Group Leader: J. C. Séamus Davis[#]

Professor Davis joined the Max Planck Graduate Center for Quantum Materials as a Fellow in November 2019, affiliated to our Institute. His primary affiliations are the University of Oxford, University College Cork, and Cornell University. He will perform collaborative work with several of the Institutes affiliated to the Graduate Center. The first planned joint work with our Institute is a project to build a spatially resolved noise microscope using broad-bandwidth SQUID readout. Although this has been far from an ideal period in which to run an international collaboration, Dr. Kent Shirer joined his project for a year in April 2020 and succeeded in fabricating a number of superconducting micro-coils which will be the primary sensors of the new microscope. Dr. Shirer will be succeeded from January 2022 by Dr. Fabian Jerzembeck. A graduate student, Hiroto Takahashi, is working in Oxford on the design and construction of the instrument itself, staying in regular contact with the Dresden part of the group by videoconference. This has been satisfactory so far, but we look forward to commencing, as soon as is practical, the physical movement of personnel between the two sites that was a key feature of the original project plan.

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1.12 Collaborative (inter-departmental/group) research activities

Although most of our projects are collaborative to some degree because the combination of physics and chemistry expertise is core to our mission and operation, a few projects are so fundamentally based on joint work that it is more appropriate to report them in this special section of the report than in the contributions from individual departments or Max Planck Research Groups.

Magnetoelasticity of $Fe_{1-y}Te$ (Chemical Metals Science and Physics of Correlated Matter)

https://www1.cpfs.mpg.de:2443/COLL_01

A theme of today's correlated materials is that they simultaneously feature many types of strong interactions and coupling, most notably electron-electron correlation and electron-lattice coupling. A natural consequence of this in certain magnetic systems is a very strong magnetoelastic coupling in which structural properties are can be tuned by the application of magnetic fields. A particularly interesting example of a system in which all three couplings are observed to play a major role is the ternary $Fe_{1+y}Te_{1-x}Se_x$, which shows an extremely rich phase diagram including superconductivity and a range of charge and spin textures. A subset of this is the binary $Fe_{1-y}Te$, in which a very large range of Fe occupation, from iron-rich to highly iron-poor is possible. After solving the chemical challenge of growing crystals across the range $Fe_{1.12}Te$ to $Fe_{0.62}Te$ and solving their structure under ambient conditions [1], we have studied the effects of their strong magneto-elastic coupling via magnetization in very high applied fields and STM tomography, revealing magnetic shape memory effects and a field-induced structural phase transition [2]. Some of the results are summarized in Fig. 1.

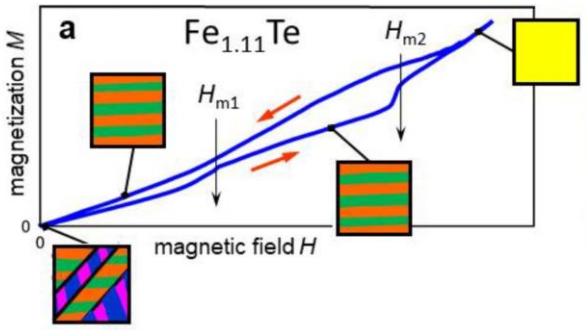

Fig. 1: Magnetic control of structural domains and an eventual field-induced phase transiton to a tetragonal structure in $Fe_{1.11}Te$.

Tailoring microstructures to reveal intrinsic chemistry and physics (Physics of Quantum Materials, Chemical Metals Science, Solid State Chemistry and Quantum Devices)

https://www1.cpfs.mpg.de:2443/COLL_02

One of the most significant changes in technical capability of the Institute over the past decade has been the establishment of a focused ion beam microstructuring laboratory in the Physics of Quantum Materials department. While microstructuring plays a key role in many of the projects described in that department's research, it has also been used in a number of collaborative projects with the chemistry departments and the Max Planck Fellow group of Professor Molenkamp.

Our critical mass of FIB expertise has proved to be useful in several ways. One highlight of the joint work was the isolation and study of single crystal microstructures from polyphase material of Be_5Pt [3] and TaGeIr [4], revealing the intrinsic properties of each material. In both cases no consensus had previously been reached in the literature, because different experiments picked up different features of the polyphase mixtures that were being studied. A second important collaborative application was the use of the FIB to isolate single-phase grains for diffraction studies by both backscattered electrons and x-rays [5], in a process outlined in Fig. 2. Among other things, this enabled the identification of the enantiomorphs of the chiral allotrope β-Mn [6]. We also performed FIB lamella preparation for both structural and Magnetic Force Microscopy [7] studies of the antiskyrmion compound $Mn_{1.4}PtSn$.

In a final project of note, we use our FIB techniques extensively for preparing devices of high-purity delafossite metals for study at Würzburg in the group of Laurens Molenkamp. Those projects are still in progress at time of writing.

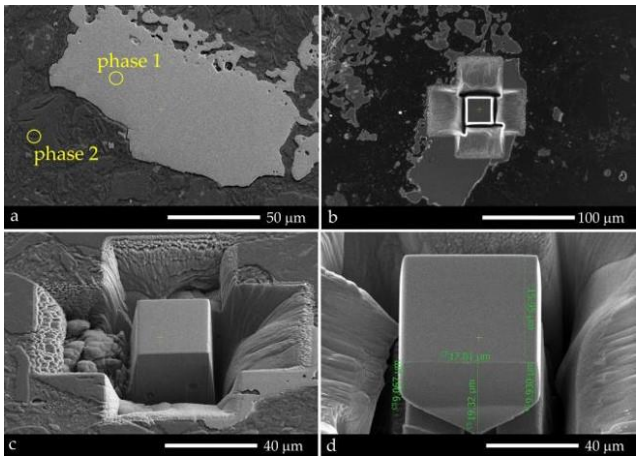

Fig. 2: Extraction of a micro-scale single crystal specimen for crystallographic investigations: (a) The areas of interest are identified, using complementary techniques such as electron backscatter diffraction (EBSD) or energy-dispersive x-ray spectroscopy (EDX); (b)-(c) a vertical column is isolated; (d) the top of the column is separated from the rest of the material and is now ready to be transferred onto a capillary for single crystal x-ray diffraction experiments.

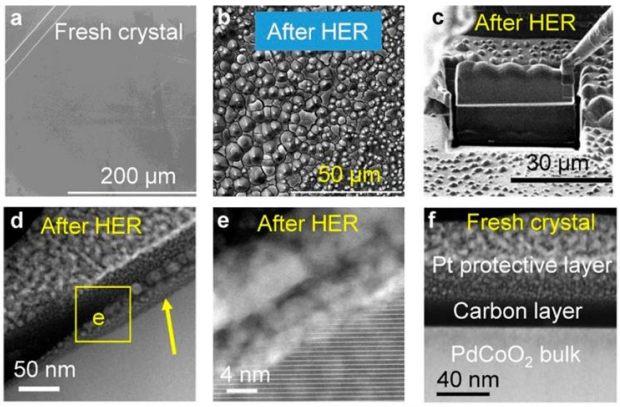

Fig. 3: (a) SEM image of a fresh $PdCoO_2$ single-crystal surface. (b) SEM image of the crystal surface after the hydrogen evolution reaction (HER). (c) A thin lamella including three islands is cut out by FIB. (d) A scanning transmission microscope (STEM) image of the single-crystal surface after the HER. A new surface layer is observed, as indicated by the yellow arrow. (e) Magnified STEM image of the interface indicated by the square in (d). (f) STEM image of a fresh single-crystal surface before HER, confirming the absence of surface layer. From [7].

High-performance metallic oxide electrocatalysts (Solid State Chemistry and Physics of Quantum Materials)

https://www1.cpfs.mpg.de:2443/COLL_03

The significance of the extremely high in-plane conductivity of the delafossite and ruthenate metals was described from a physics point of view in the report from the Physics of Quantum Materials department. It has also caught the attention of the chemistry community because of the potential of such metals to be good electrocatalysts for water splitting reactions. Experiments in the Solid State Chemistry department quickly established that this is indeed the case. $PdCoO_2$ single crystals showed a remarkably high electrocatalytic activity with an extremely low overpotential, low Tafel slope, and good stability, outperforming the state-of-the-art Pt catalyst. The reason for this proved to be that the reaction substantially changes the surface of the crystal, producing a natural hybrid system of Pd nanoclusters and cobalt oxide on a highly conducting substrate (the remainder of the crystal). Details are shown in Fig. 3. The conditions for efficient catalysis (a large number of active sites for hydrogen access and an expressway for electron transfer) are therefore both achieved and we observe reversible hydrogen sorption and desorption with the highly favorable parameters described above [8]. Ongoing studies of Sr_2RuO_4 and $Sr_3Ru_2O_7$ also look promising, with broadly similar chemical and physical processes at work.

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[6] *U. Burkhardt et al., Sci. Adv.* ***7*** *(2021) eabg0868, doi.org/10.1126/sciadv.abg0868*

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2. Structure and Organization of the Institute

2.1 Structural Summary

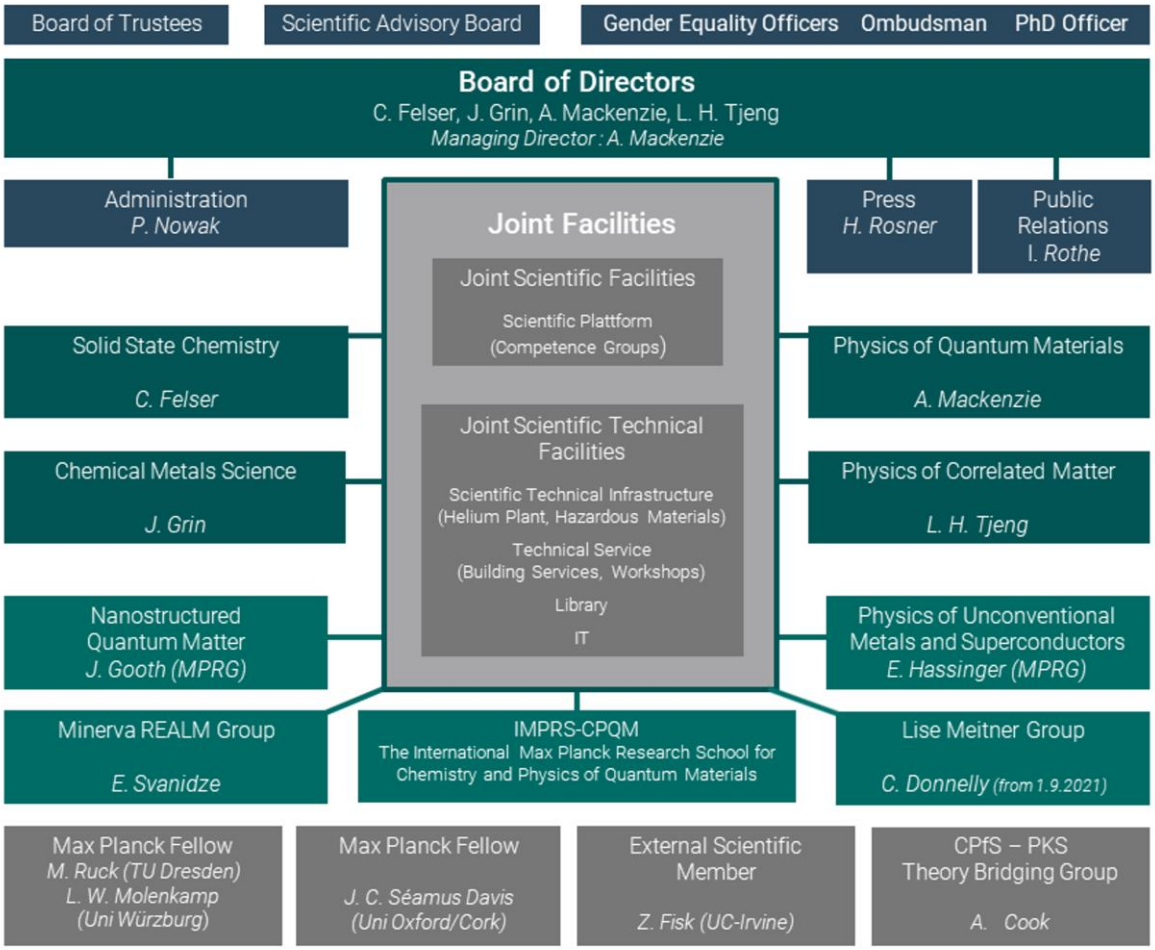
The structure of our Institute, largely unchanged since the last review period except for the staffing developments outlined in the Executive Summary, can now be conveniently summarized in the diagram below.

The scientific activities of the Institute are led by the Departments for Solid State Chemistry (SSC; Prof. Claudia Felser), Physics of Quantum Materials (PQM; Prof. Andy Mackenzie), Chemical Metal Science (CMS; Prof. Juri Grin), and Physics of Correlated Matter (PCM; Prof. Liu Hao Tjeng), as well as by the Max Planck Research Groups (MPRG) for Physics of Unconventional Metals and Superconductors (Prof. Elena Hassinger) and Nanostructured Quantum Matter (Dr. Johannes Gooth). From September 2021 we will also be hosting a Lise Meitner Excellence Group (Dr. Claire Donnelly).

The Board of Directors receives advice and recommendations from the Scientific Advisory Board, the Board of Trustees, the Ombudsman (Dr. Peter Höhn), the Gender Equality Officers (Dr. Gudrun Auffermann and Ms. Renate Hempel-Weber), the PhD Officer (Dr. Burkhard Schmidt) and the Representative of the Scientists in the CPT section of the MPG (Dr. Steffen Wirth). As a temporary appointment during the pandemic, Dr. Oliver Stockert is serving as Home Office Advisor. Dr. Marcus Schmidt's role as Safety Officer has considerably expanded to include occupational health during the pandemic.

The Board of Directors receives assistance in its decision-making from the Head of the Administration (Mrs. Petra Nowak) and Institute Commissions on finances, technical services, computer services, library, and safety issues. A Pandemic Crisis team comprises the Managing Director and his Deputy plus the Safety Officer, Gender Equality Officer and heads of key technical facilities.

The Scientific Platform, consisting of senior scientists from the four Departments, provides the long-term, stable expertise that we need in the fields of structure,



metallography, chemical analysis, material development, extreme conditions, neutron/electron spectroscopies, and theory. Its membership is flexible, which will be important as we adapt over the coming few years to the arrival of a least two new Directors.

Close scientific collaboration with the Technical University Dresden is anchored by the Max Planck Fellowship of Prof. Michael Ruck from the Chemistry Department. Our scientific collaboration with the German university sector is further enhanced by Max Planck Fellowship of Prof. Laurens Molenkamp of the Julius-Maximilians-Universität (JMU) Würzburg, with an added international dimension provided by the recent appointment to a Max Planck Fellowship of Prof. J.C. Séamus Davis of the University of Oxford and University College Cork. Common research activities with the neighboring Max Planck Institute for Physics of Complex Systems (PKS) are carried out by the CPfS-PKS bridging group. Following the departure of Dr. Takashi Oka to take up a Professorship at the University of Tokyo, we have recruited Dr. Ashley Cook to head this group. She has been in post since September 2020. Impulses for new scientific projects are provided by the External Scientific Member Prof. Zacharias Fisk (UC-Irvine). One of the saddest events of the last assessment period was the untimely death of our second External Scientific Member Prof. Shoucheng Zhang (Stanford).

The Institute also receives support from Technical Services (headed by Mr. Andreas Schwoboda), Computer and IT Services (headed by Mr. Jens Gerlach) and the Librarian Mrs Ina Werner.

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2.2 Pandemic Response

The twin goals of our pandemic response were to maintain core operations as far as possible, while giving ultimate priority to the safety of our staff. In March 2020 a crisis team was established to develop appropriate rules of operation, in close cooperation with the central crisis team of the MPG. This process has been on-going since then, with the rules adapted to best suit the local and international circumstances of the pandemic. Unlike many scientific institutions world-wide, we never had to enforce a complete shut-down. Considerable credit for this has to go to our staff, who exercised care both in their personal lives and, especially, in the work-place: as far as we know, we have had only one suspected case of transmission within the Institute.

Although it was possible to maintain running experiments that could largely be computer-controlled, all other aspects of our operation suffered considerable disruption. Visits and foreign travel basically ceased, and laboratory training of new scientific staff and graduate students was substantially hindered. Home office was widely practiced by scientific and administrative staff, and vital technical and service staff were split into two teams working alternate weeks, to avoid all having to be sent home as a result of contact tracing.

In the early part of the crisis, we were hampered while contact tracing by instructions from the central MPG crisis team that potentially infected people should not be named. Following widespread criticism of this in an anonymous staff questionnaire about the handling of the crisis we initiated a system in which people could volunteer to be named; this has worked well.

New forms of communication have played an essential role in this process. To this end, we implemented a new intranet, MAX-Net, with virtual team rooms, and both video conferencing technology and the bandwidth of our VPN system were expanded. Technical help was made available to those who required it in order to set up home office.

Attention was also paid to monitoring the mental health of those confined for most of their time at home. Via organization at department and group levels, the goal was to have video meeting contact with everyone at least twice per week. In the early stages of the pandemic we also engaged in community initiatives such as fund-raising and purchase of masks from contacts in China which we donated to the medical profession in Dresden.

There is certainly no perfect response to an unprecedented crisis of this kind, and like the staff of any organization, ours have a spectrum of opinion on how to strike the correct balance between safety and unrestricted work. However, we believe that our combined infection and productivity statistics over the assessment period show that the compromises we chose were reasonable.

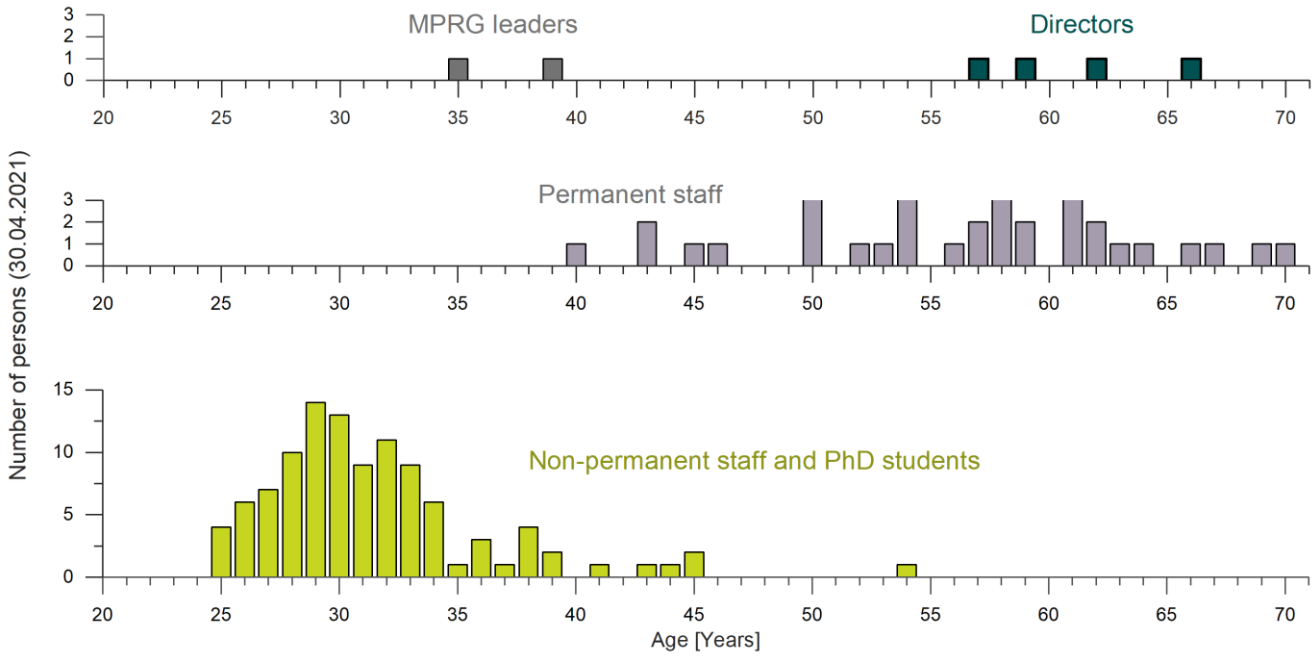
2.3 Personnel Structure

The overall structure of the scientific personnel of the Institute is most compactly summarized in the following graph showing senior staff, permanent non-directorial staff scientists and those on temporary positions either as graduate students or as post-docs.

The permanent staff are each members of, or overseen by, one of our departments, Scientific Platform included. Physics of Quantum Materials has 7 permanent staff members. Physics of Correlated Matter has 6, Solid State Chemistry has 8 and Chemical Metals Science 12 members. Taking person-month totals and averaging over the assessment period, we had 49 registered doctoral students at any time (20 in Physics of Quantum Materials, 5 in Physics of Correlated Matter, 11 in Solid State Chemistry, 11 in Chemical Metals Science and 2 in our MPRGs). Using the same procedure, the number of postdocs and group leaders without permanent contracts is 65 (17 in Physics of Quantum Materials, 9 in Physics of Correlated Matter, 27 in Solid State Chemistry, 8 in Chemical Metals Science and 4 in our MPRGs). In addition, we have 3 international undergraduate interns from the Max Planck Centers with Korea/Taiwan and UBC Canada, and several undergraduate and masters students doing their thesis work with our groups.

The scientific work of the Institute is supported by 58 technical and engineering staff and 29 administrative staff.

Further details of all these statistics can be found in the Addendum and in the 'Data, Facts, and Figures' sheets which will be provided during the Scientific Advisory Board site visit in November 2021.



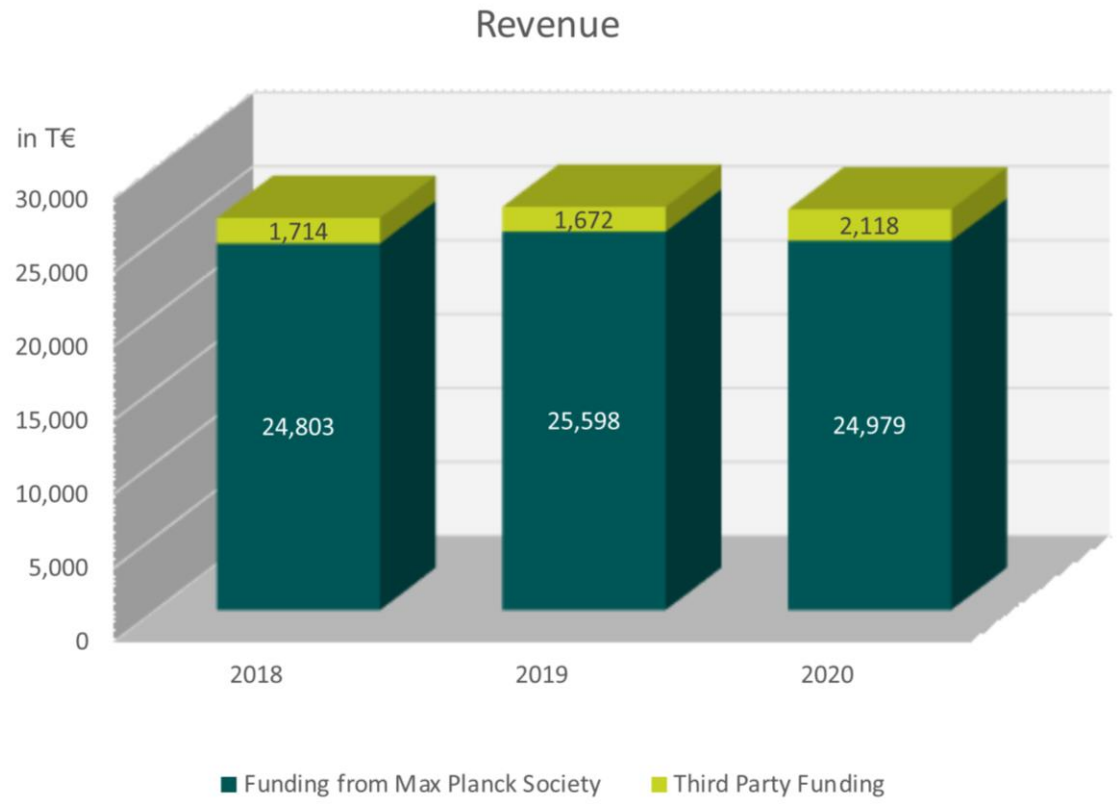
2.4 Structure of the Institute's Financing, Material Resources, Equipment and Spatial Arrangements

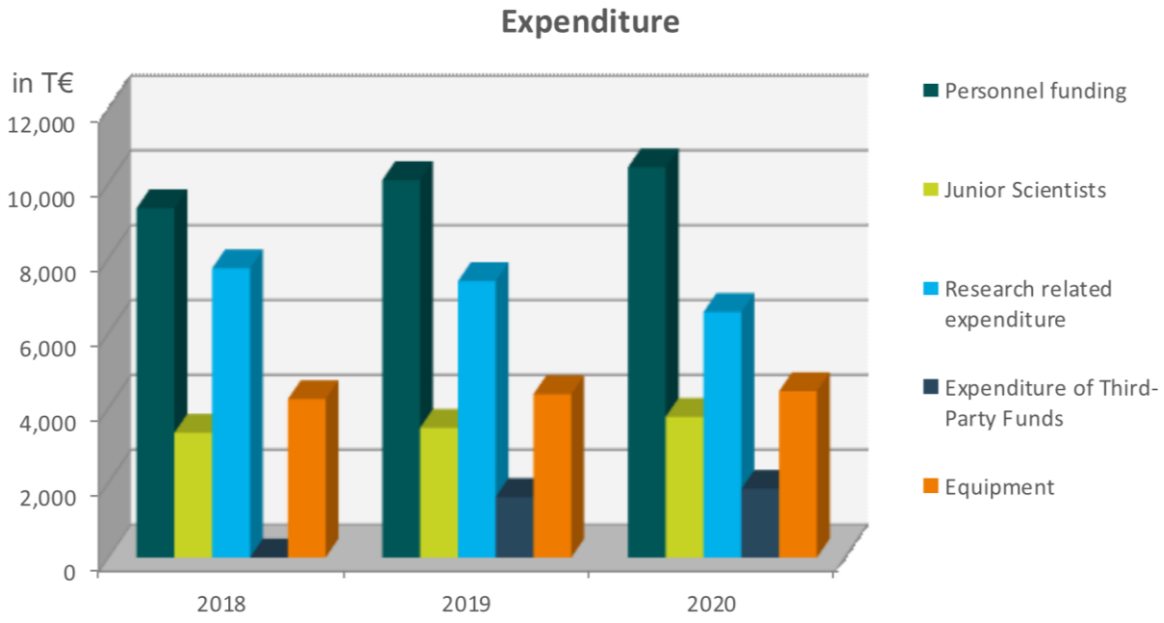
2.4.1 Institute financing

We summarize our budget structure in the enclosed graphs, and provide extensive statistical details in the Addendum and in the 'Data, Facts, and Figures' sheets which will be provided during the Scientific Advisory Board site visit in November 2021.

2.4.2 Building projects

The assessment period has seen no large-scale building projects, except for the rebuild of the kitchen for our cafeteria. Originally planned for 2020, this was postponed for a year but commenced in 2021. The main item of future planning was for the new clean room and office build mentioned in the Executive Summary and now scheduled for completion in 2024.





2.5 Junior and Guest Scientists and Career Development

At any one time we host over 100 graduate students and post-docs, and have devoted considerable time over the past years to providing them with a good environment in which to work.

2.5.1 Graduate student education and supervision

The assessment period has seen the International Max Planck Research School for Chemistry and Physics of Quantum Materials (IMPRS-CPQM; https://imprs-cpqm.mpg.de), started in January 2016, mature as the framework within which we recruit and train our doctoral students. A significant milestone was its positive review in 2020, leading to the extension of its central funding until the end of 2027. Overall, we received very positive comments both from reviewers and existing students (in an anonymous questionnaire), along with a few pointers for improvement which will be implemented in the coming years.

Although IMPRS-CPQM is the main route for student recruitment to the Institute, it is not the only one. Students can also be paid solely from departmental funds or from external grant money. We decided, however, that there should be as little distinction as possible between the two classes of student. IMPRS-CPQM courses are available to everyone, and, importantly, the same guidelines for supervision and monitoring are also standard across the Institute. At the time of our last report the new Thesis Advisory Committee system was unproven; we can now see that it is working well overall. More details can be found at https://imprs-cpqm.mpg.de/45308/supervision.

Similarly, the Institute PhD Officer, Dr. Burkhard Schmidt, is independent of IMPRS-CPQM and monitors the welfare and progress of all the students, holding regular meetings with PhD representatives, as well as individual interviews with each student. His work has been particularly valued during the pandemic.

Since 2019 we have added a new graduate student recruitment channel to our portfolio through our membership of the Max Planck Graduate Center for Quantum Materials. This is an elite venture that offers extra, high-level courses to its members, who are also by default part of IMPRS-CPQM. Those courses have been offered on-line during the pandemic, and used as a learning resource by Graduate Center students and others.

2.5.2 Community-building among post-docs

Through the PhD Officer, IMPRS co-ordinator Markus Koenig and their team of elected representatives (https://www.cpfs.mpg.de/2735818/phd_representatives), the doctoral students have a collective sense of identity. The post-doc community covers a wider range of age, family circumstances and contract length than that of the students, so establishing something similar there is more of a challenge. We are pleased to report that they have now established representatives of their own (https://www.cpfs.mpg.de/2958299/postdocs) and we will continue to offer help and encouragement with their community-building efforts. The career pressures associated with being a post-doc mean, unfortunately, that the pandemic has likely been particularly difficult for our post-docs. The main thing we can do to help is to offer them contract extensions with us to allow the job market and international mobility to free up; we are trying to take all reasonable steps in that direction. In parallel, we offer a suite of skills courses and events, summarized in section 2.6.

2.5.3 Graduate employability

We believe that the employability of our graduates is excellent: Of the 33 PhD students graduating from the groups of Institute staff in the assessment period 22 found employment in academic research science and 10 in industry (one is on post-defense travel).

2.5.4 Academic career advancement

The assessment period has seen strong performance from research staff wishing to further their academic careers. Those moving on to established positions elsewhere include:

Chenguang Fu took up a ZJU100 Young Professor position at Zhejiang University, Hangzhou, PR China.

Jacob Gayles was appointed to the Department of Physics at the University of South Florida, USA, as an Assistant Professor.

Roman Gumeniuk was promoted to a full Professorship at TU Bergakademie Freiberg (October 2020).

Philipp Hansmann moved to a Professorship position (W2) for Theoretical Physics at the Friedrich-Alexander University of Erlangen-Nuremberg.

Clifford Hicks was appointed as a Reader at the University of Birmingham.

Nitesh Kumar moved to an Assistant Professorship at the Bose Institute, Kolkata.

Guowei Li moved to an Assistant Professorship at Ningbo Institute of Materials Technology & Engineering, Chinese Academy of Sciences.

Enke Liu was promoted to a full Professorship at the Chinese Academy of Sciences Institute of Physics, Beijing.

Kaustuv Manna joined the Indian Institute of Technology Delhi as an Assistant Professor.

Philip Moll moved from his MPRG in our Institute to an Assistant Professorship at EPFL in 2018, and has subsequently been appointed to a Directorship at the Max Planck Institute for Structural Dynamics in Hamburg.

Kimberley Modic was appointed as an Assistant Professor at IST Austria.

Jayita Nayak took up an Assistant Professorship at the Indian Insitute of Technology, Khanpur.

Takashi Oka was recruited back to Japan, to a full Professorship at the University of Tokyo.

Joyce Pham was appointed as an Assistant Professor at California State University, San Bernardino.

Ricardo D. dos Reis will head the newly established Max Planck Partner Group based at SIRIUS, the Brazilian Synchrotron Light Laboratory, LNLS, in Campinas, Brazil.

Jhuma Sannigrahi was appointed to an Assistant Professorship at the Indian Institute of Technology, Goa.

Jakub Żelezny moved to the Institute of Physics, Czech Academy of Sciences as a Head of Group.

2.5.5 Helpline Dresden

Happily, we know of no incidents of racial harassment of our staff during the assessment period, but the Helpline that we co-established in 2017 has been widely used elsewhere in Dresden, aiding victims, police and medical services with cases of racial and domestic assault, medical emergencies and even being used to assist with an anti-drugs operation.

2.6 Equal Opportunities

One of our foremost priorities is to strengthen the awareness of equal opportunity and to achieve good gender balance in our institute. Therefore, we attempt to provide gender-fair working conditions. We support our employees in their different career paths, ensuring work-life balance. In collaboration with our Gender Equality Officers Gudrun Auffermann and Renate Hempel-Weber, we have pursued our equal opportunity concept and fixed the most important items in our gender equality plan. The Gender Equality Officer Team regularly participates in workshops organized by the Central Gender Equality Officer of the MPG, is embedded in several gender equality networks and takes care of gender-fair language in the institute. It also works with the local and central administration and the PhD and post-doc representatives to provide a suite of training courses, most of which are available to all genders. Activities in the assessment period include:

2018: 28th – 29th August: Workshop "Career development and third-party funding" for group leaders and advanced postdocs. Coach Dr. Beate Scholz, CTC, Bonn/Trier.

13rd – 14th November: Workshop "Stress management: Own change planning through reflection and practical exercises". Coach Ilona Vogel, Flieden.

December: Joyce Pham was selected for the Sign Up! Career Building 2019, which pursues the goal of supporting female Postdocs and scientists in their career development and to prepare them for leadership roles.

2019: 8th February: Workshop on talk technique by fellows of the Elisabeth Schiemann College.

18th – 19th March: "Effective Proposal Writing".

9th – 10th April: "Faculty recruitment at German Universities".

4th – 5th June: "Heading for New Horizons".

August: Renate Hempel-Weber qualifies as a certified mediator for organizational and intercultural affairs for all institute members.

27th – 28th August: Workshop "Developing your research brand" for group leaders and advanced Postdocs. Coach Dr. Beate Scholz, CTC, Bonn/Trier.

7th October: Workshop "Horse supported team building seminar". Coach Anja Heine, Pulsnitz.

October: The Institute successfully applied to the Max Planck Society BOOST program for salary enhancement of three female group leaders.

2020: 1st December: Reselection of the Gender Equality Officer Team (12/2020-11/2024).

December: Hilary Noad was selected for the Sign Up! Career Building 2021.

2020/2021: Redesign of the gender equality plan of the institute by mutual consent of the Board of Directors, the Head of Administration and the Gender Equality Officer Team.

2020/2021: Redesign of the web pages "Equal Opportunity" in the new MAX Intranet by the Gender Equality Officer Team.

Gudrun Auffermann and Petra Nowak are the contact persons of our institute in the framework of the Career Steps Network of the MPG and continuously provide our scientists with information to support them at different steps of their career.

In addition to these local activities, Gudrun Auffermann (Deputy Gender Equality Officer since 2015) was elected as Gender Equality Officer of the CPT-Section for 2019 – 2023.

The current gender situation in our institute (30.04.2021) is majority female for the PhD students (56%) but rather imbalanced at the Postdoc stage (19% female). Among group leaders and staff scientists with permanent contracts the percentage of female staff is 26%, at W2 33% and at W3 25%. For context, all these numbers except that for post-docs are substantially above the average for the 33 Max Planck Institutes in the Chemistry, Physics and Technology Section of the Max Planck Society. As described in the Executive Summary, we have been aggressively tackling the imbalance at group leader level, appointing eight female group leaders and two senior female W2 scientists during the assessment period, one of whom is not included in the 30.04.2021 census because she is not yet in post. Finally, we are pleased to note that over 50% of the prizes and awards detailed in section 2.9 went to female staff and graduate students.

2.7 Cooperation with National and International Research Institutes and Companies

In keeping with the nature of our field, a large proportion of our work is in collaboration with partners outside our Institute. We prefer to keep these collaborations informal and therefore administratively light-touch wherever possible, but we also run a number of formalized collaborations where the links are stronger and long-term, and involve regular travel of personnel. Here we provide a brief summary; extensive details are provided in the Addendum.

2.7.1 Major international collaborations involving formal agreements

The Physics of Correlated Matter department led by Hao Tjeng has strong links with the scientific communities of Taiwan and Korea. These have been formalized in Memoranda of Understanding with the NSRRC, NCTU and NHTU in Taiwan, and via the Max Planck Center of Complex Phase Materials with POSTECH (Korea) and NSRRC-NCTU-NHTU (Taiwan). Our Institute continues to be an active partner within the Max Planck Center with UBC (Canada) on Quantum Materials. This Center has also been extended recently to include the University of Tokyo. Juri Grin leads a collaboration with National University of Lviv in Ukraine and Claudia Felser two further collaborations, with the Czech Academy of Sciences and the Weizmann Institute respectively. In the assessment period new Max Planck Partner groups have been established with the Brazilian Synchrotron Radiation Laboratory and the University of La Plata, both led from the Max Planck side by Andy Mackenzie. A formal agreement has also been made regarding the participation of TU Dresden and the University of St Andrews in IMPRS-CPQM, along with a broader agreement with St Andrews regarding shared doctorates.

2.7.2 Scientific partners (from publications)

The most concrete evidence of our collaborative science comes from an analysis of our publications. During the assessment period, we jointly authored papers with more than 150 institutions elsewhere in Germany and beyond. A full list in alphabetical order can be found in the Addendum.

2.7.3 Industrial partners and patents

Our main industrial collaborations are with major international companies such as Western Digital (Felser), Intel (Felser), and BASF (Grin), but we also have links with smaller companies spawned from research by Institute members such as Razorbill and Innovative Measurement Technology.

In the census period we have applied for twelve patents. The inventors and the working titles are:

(i) Prof. Dr. Claudia Felser, Dr. Rolf Stinshoff, Dr. Roshnee Sahoo
Rare Earth Metal-free Hard Magnets

(ii) Prof. Dr. Claudia Felser, Dr. Kaustuv Manna, Joseph Heremans, Sarah Watzmann, Nadini Trivedi, Tim McCormick
Thermoelectric Device using Weyl Semimetal

(iii) Prof. Dr. Claudia Felser, Dr. Guowei Li, Prof. Dr. Andrew Mackenzie, Dr. Seunghyun Khim
$PdCoO_2$ als HER Katalysator

(iv) Dr. Clifford Hicks
Piezoelectric-driven uniaxial pressure cell for transmission experiments

(v) Prof. Dr. Claudia Felser
Method of identifying topological compounds

(vi) Prof. Dr. Claudia Felser, Dr. Yan Sun
Bulk band structure effect for HER catalyst
(vii) Prof. Dr. Claudia Felser, Dr. Guowei Li
OER in a low magnetic field
(viii) Prof. Dr. Claudia Felser, Dr. Guowei Li
HER in a low magnetic field
(ix) Dr. Thomas Gruner, Dr. Manuel Brando, Jacintha Banda, Prof. Friedrich Malte Grosche, Jiasheng Chen
Kühlung durch adiabatische Entmagnetisierung intermetallischer Systeme
(x) Dr. Guowei Li, Dr. Kaustuv Manna, Dr. Yan Sun, Prof. Dr. Claudia Felser
Chiral crystals for Water electrolysis and fuel cells
(xi) Prof. Dr. Claudia Felser, Dr. Guowei Li, Dr. Chenguang Fu, Dr. Yan Sun, Dr. Andrei B. Bernevig, Zhida Song, Yuanfeng Xu
Heterogenous catalysts with metallic surface states
(xii) Prof. Dr. Claudia Felser, Yangkun He
Magnetic Bubble Memory

2.7.4 Research grants and national collaborations

Our members are partners in two Deutsche Forschungsgemeinschaft (DFG) Collaborative Research Centers (SFB) with TU Dresden, one DFG Collaborative Research Center (SFB Transregio) with the Goethe-Universität Frankfurt a.M., Karlsruhe Institute of Technology and Johannes-Gutenberg-Universität Mainz, one DFG Cluster of Excellence, ct.qmat, with TU Dresden and Julius-Maximilians-Universität Würzburg, one DFG Research Unit, four DFG Priority Programmes and fifteen DFG Individual Grants. In addition, we are partners in six EU and international grants including one ERC Advanced Grant and we host two EU Marie Skłodowska-Curie Individual Fellowships. Finally, as mentioned in section 2.5.1, we are funded members of the Max Planck Graduate Center for Quantum Materials.

2.8 Statistical Summary of Publications and Invited Talks; Open Access and Archiving Policy

In this section we provide a brief statistical analysis of our primary forms of disseminating our results, namely our publications and the talks that we give at conferences. We mention talks in this section because they are important both for increasing the visibility of our Institute and of our non-Directorial staff. All departments have the default policy that any junior scientist or PhD student attending a conference should present either a contributed talk or a poster on their research.

2.8.1 Publication statistics

In section 1 of this Status Report and the related web-based research summaries we discussed research highlights and associated publications. In Table 1 on the following page, we provide statistics on our volume of publication from May 2018 to April 2021, in a format that shows the extent of inter-departmental publication. The institute's total numbers of publications for the period are shown in bold. The total outputs of the individual departments and the collaborative work within the institute are specified in detail.

2.8.2 Conference talks

During the assessment period, Directors gave approximately 160 plenary, keynote and invited talks at international conferences and workshops and individual institutions. The MPRG leaders gave approximately 20 such talks, and non-directorial staff (staff scientists, post-docs and PhD students) a further 200. Due to the pandemic, 76 of these talks were given virtually, and over 20 further invitations that had been issued were cancelled due to the relevant conferences not taking place. Details can be found in the Addendum.

2.8.3 Open access

Materials physics and parts of materials chemistry have for a long time operated a major open access vehicle not available to many other fields – the preprint archive ArXiv, to which the Max Planck Society makes a financial contribution. Almost all journals now allow posting of a late-stage manuscript on this site, containing both all the information in the final published version and a reference to that publication. We already make extensive use of this facility, and our policy is to make it a systematic requirement for all submissions on topics covered by it. In physics, it has become a far more widely-used vehicle for the dissemination of results than open access journals, and has the advantage of not being subject to the large paper-by-paper fees charged for Open Access by many journals. In some cases, it is still appropriate to opt for journal-based Open Access and in those cases our staff are encouraged to do so. For all publications, we make systematic use of the publication repository of the Max Planck Society.

Year	***2018***	***2019***	***2020***	***2021***	
Months	*05-12*	*01-12*	*01-12*	*01-04*	
					Sum
Publications total	**170**	**294**	**291**	**116**	**871**
CMS	19	36	31	18	104
PCM	24	54	59	39	176
PQM	34	68	59	15	176
SSC	55	76	84	23	238
CMS/PCM	2	1	3	3	9
CMS/PQM	3	4	8	2	17
CMS/SSC	7	9	9	2	27
PCM/PQM	7	4	4	0	15
PCM/SSC	2	7	3	2	14
PQM/SSC	3	5	1	1	10
CMS/PCM/PQM	0	2	3	0	5
CMS/PCM/SSC	1	0	2	0	3
CMS/PQM/SSC	3	7	5	0	15
PCM/PQM/SSC	1	0	0	0	1
CMS/PCM/PQM/SSC	0	1	0	0	1
CMS*	35	60	61	25	181
PCM*	37	69	74	44	224
PQM*	51	91	80	18	240
SSC*	72	105	104	28	309
MPRG Hassinger	3	3	4	2	12
MPRG Moll	7	8	8	0	23
MPRG Gooth	6	10	7	3	26
MP Fellow Ruck	5	14	17	10	46
MP Fellow Molenkamp	0	1	0	0	1
MP Fellow Davis	0	0	1	1	2
Open Access	31	68	162	57	318

Table 1: The first row of the table (bold) provides the total number of publications per year for the institute. In the following, this is subdivided into contributions from the individual departments and collaborations of two, three or all departments. In addition, the contribution of the MPRG's and the fellows are given. Since there are common publications with the departments as well, a small double counting of about 10 papers occurs. The departments, MPRGs and fellows are labeled as follows: CMS: Chemical Metal Science (Grin), PCM: Physics of Correlated Matter (Tjeng), PQM: Physics of Quantum Materials (Mackenzie), SSC: Solid State Chemistry (Felser), Max Planck Research Groups Hassinger, Moll and Gooth, Max Planck Fellows Ruck, Molenkamp and Davis.

**publications that are authored or co-authored by an author from this department*

2.8.4 Long-term archiving of research findings

We comply with the "Rules of Good Scientific Practice" adopted by the Senate of the Max Planck Society on November 24, 2000 and amended on March 20, 2009.

2.9 Recognition: Scientific Awards, Fellowships and Memberships

The assessment period has seen a number of awards for our Directors, Emeritus Directors and, pleasingly, our junior staff and graduate students.

At Director level, Claudia Felser shared the 2019 James C. McGroddy Prize for New Materials of the American Physical Society, Frank Steglich shared the 2020 Fritz London Memorial Prize for Low Temperature Physics, Juri Grin won the 2020 V.I. Vernadskyj Gold Medal of the National Academy of Sciences of Ukraine. Claudia was also elected to the US National Academy of Science and the US National Academy of Engineering and the German National Academy for Science and Engineering. Emeritus Director Martin Jansen, then leading an emeritus group at our Institute, won the 2019 Otto Hahn Prize of the German Chemical Society and German Research Council and Andy Mackenzie was awarded a Hanna Visiting Professorship to Stanford University in 2019.

Major mid-career prizes also went to Max Planck Research Group leaders. Philip Moll was awarded the 2018 Nicholas Kurti Prize, and Johannes Gooth the 2020 Rudolf Kaiser Prize.

Demonstrating that the future of the field is in good hands, our graduate students picked up a large number of well-deserved awards and prizes.

Mark Barber received a Springer Thesis Award in 2018, Riccardo Freccero won the Best Thesis Prize of the Inorganic Chemistry Division of the Italian Chemical Society in 2019 and a Springer Thesis Award in 2020. Yang Zhang was awarded an Otto Hahn Medaille of the Max Planck Society in 2020 and Maja Bachmann a Springer Thesis Award in 2019 and an Otto Hahn Medaille in 2021. Veronika Sunko won a Springer Thesis Award in 2019, and in 2020 an Otto Hahn Medaille, the Richard L. Greene Dissertation Prize of the American Physical Society and the Woodruff Thesis Prize of the UK Institute of Physics. Conference talk and poster prizes were won by M. O. Ajeesh, Andrea Amorsese, Iryna Antonyshyn, Maja Bachmann, Natalia Gloriozova, Julia Hübner, Felix Kaiser, Brett Leedahl, Eteri Svanidze and Elina Zhakina.

2.10 Service: Scientific Members' Committee Work and Institute Members' Teaching

We are conscious of the privileged position that we hold as a well-funded research-intensive Institute, and therefore encourage our staff to retain strong links with the rest of the community through appropriate service on external committees and university teaching.

2.10.1 Committee work

The Directors and several senior scientists of the Institute are members of numerous national and international committees and panels.

In particular, staff members are actively involved in review panels for beam time allocation at large scale facilities. A list is included in the Addendum.

2.10.2 Teaching

Members of our Institute carry out teaching activities on a regular basis. This is not only service to the partner University (TU Dresden) but forms also an excellent opportunity for our non-director scientists to improve and demonstrate their capabilities in teaching and educating students, which is of utmost importance for the advancement of their careers in academia. Obviously, our teaching is also an opportunity for students to get acquainted to our research field and our Institute, which may motivate them to do a Bachelor, Master or PhD project with us.

In the period April 2018 – March period (semesters at TU Dresden start in April and October), Institute members have given more than 27 lecture courses and 27 teaching seminars with a total teaching load of more than 60 SWS (Semester Wochen Stunde = hours per week per semester). This corresponds to approximately five different lecture courses per semester with 2 SWS per course on average.

The lectures range from basic courses to courses on specialized topics, and are given not only to Physics and Chemistry students, but also to students from the Engineering, Business-Engineering, Materials Science, Geography and Teaching departments of TU Dresden.

A detailed list of our teaching activities is included in the Addendum.

2.11 Conferences, Workshops, and Seminars

Using a combination of our own venue and, for physics, collaboration with our colleagues in the neighboring Physics of Complex Systems Institute, we believe that we have established Dresden as a known global focal point of our field. We strive to maintain this position by organizing large numbers of conferences and workshops for up to 100 each (consistent with our capacity), as well as occasional larger meetings held elsewhere in the city. We also participate actively in the Program and Advisory Committees of the majority of the large international conferences in our field. This aspect of our activities was hit particularly hard by the pandemic, but we still hosted 31 workshops and meetings, and 62 seminars and colloquia from external scientists in the assessment period, making use of virtual events as appropriate. A list can be found in the Addendum.

2.12 Public Relations Work

Reaching out to the public with current research topics on the local, national and international level is the main task of the public relations team.

One of our highlights is the Long Night of Science – "Lange Nacht der Wissenschaften" (LNdW). The LNdW is *the* local science event in which the major research institutions of the city open their doors to the public and show their research through experimental shows, interactive games and popular talks (see Fig. 1). For many years and again in 2018, our Institute participated and gained the attention of several thousand visitors. Due to the Covid-19 pandemic, the activities planned for 2020 were postponed to July 2021, when we participated in a virtual event.

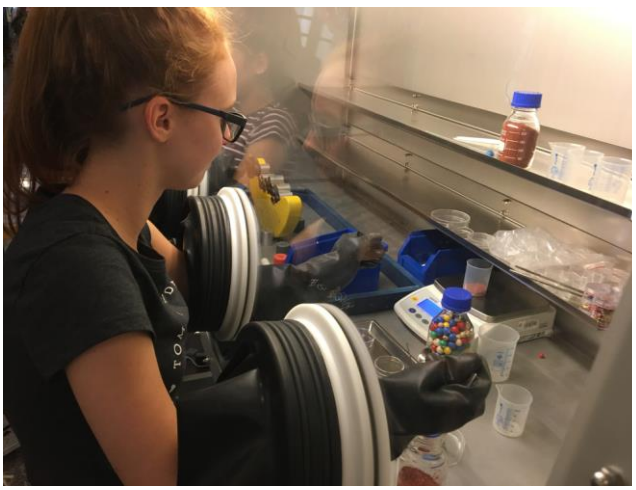


Fig. 1: Experiments by visitors at the Long Night of Science.

Fig. 2: Max Planck Day – Panel discussion with Saxon State Minister of Science, Dr. Eva-Maria Stange, MP directors Grin, Huttner, Rost, and journalists.

In addition to our usual commitment of organizing events, workshops and lab tours, our Institute and the two other Dresden Max Planck Institutes hosted the "Max Planck Day" in Dresden Town Hall on the 14th September 2018. On that day, most of the Max Planck Institutes offered insight into their research with special programmes. The hall where the mayor and the city council usually hold their debates was in the hands of researchers and citizens of Dresden and its surroundings. Diverse topics were on display in a new movie, in an exhibition which laid the background to fascinating experiments, talks by young scientists and a panel discussion with Max Planck directors, the Saxonian Minister of Science, the mayor, and journalists (see Fig. 2). The day closed with relaxed chats in the science café.

We also participate in public life in Dresden through our membership in several networks such as

Fig. 3: Science Tram – PhD student Peter Swekis explaining his research to "passengers".

Fig. 4: Ana Barrios, Felix Kaiser, Maja Bachmann, and Philippa McGuiness (f.r.t.l.) with PhD officer Burkhard Schmidt (back), at the Famous Female Researchers exhibition which they helped organize.

"DRESDEN-concept" (DDc). On 29th January 2019, scientists of our Institute joined the DDc "Science Tram" (see Fig. 3). People aboard were able to follow highlights of the research of several scientists and ask questions on that evening ride through Dresden.

The International Women's Day 2019 acquired particular attention in our institute and beyond: PhD student Ana Barrios had encouraged her peers to set up an exhibition about famous female researchers which was shown in our institute for several weeks and later in the MPI of Molecular Cell Biology and Genetics (see Fig. 4).

Throughout every year, we welcome international groups for fruitful scientific exchange, especially among young researchers. In this way, one of our PhD students, Philippa McGuinness, hosted two selected participants of the Lindau Nobel Laureate Meeting for a few days in July 2019 and brought them together with her colleagues and their research fields.

Fig. 5: Guided tour through the 'Chemistry meets Art' exhibition with the artist Sabine Wrabetz.

From September through November 2019, we had the pleasure to host the public exhibition "Chemistry Meets Art" of the chemist and artist Sabine C. Wrabetz. This exhibition was sponsored by the Wolfgang Johannes Hönle Foundation Art and Chemistry (see Fig. 5).

Meanwhile our membership in the jury of the Saxon Award for Democracy has become a tradition since we find it very important to support awardable initiatives to combat racism and discrimination and to promote a tolerant society.

http://www.wissenschaftsnacht-dresden.de/

https://www.mpg.de/12251976/max-planck-tag-2018

https://www.cpfs.mpg.de/2974909/20190125

https://www.cpfs.mpg.de/3087011/20190902

http://www.demokratiepreis-sachsen.de/der-foerderpreis-2020/