Hydrostatic and Uniaxial Pressure Tuning of Topological Materials and Unconventional Superconductors

Michael Nicklas#, Christoph Geibel, You-Sheng Li, Andrew P. Mackenzie, Mario M. Piva and Helge Rosner

Tuning of quantum and topological materials by external stimuli offers the potential to discover new unconventional phases, to tune a material to desired properties, or to switch between different states of matter, eventually leading to novel applications. External pressure is an excellent and clean tool for this without introducing additional complications. In our investigation of competing energy scales in $LuFe_4Ge_{12}$, we employed hydrostatic pressure as a tool to identify the underlying physical principles. The structure of this material exhibits a low-dimensional arrangement and geometric frustration of the Fe moments, which directly affect the interplay between structural and magnetic degrees of freedom. By combining the strengths of macroscopic and microscopic probes under hydrostatic pressure, we were able to gain a comprehensive understanding of the physics in $LuFe_4Ge_{12}$. Hydrostatic pressure studies have also provided insights into the importance of domain walls for understanding transport in the ferromagnetic Weyl semimetal CeAlSi, where both space-inversion and time-reversal symmetries are broken. In $HfTe_5$, compelling evidence has been found for the emergence of a quasi-quantized three-dimensional quantum Hall effect. Moreover, in $MoTe_2$, a promising candidate for the realization of topological superconductivity, our study appears to rule out this possibility. In contrast to hydrostatic pressure, the application of uniaxial pressure can also break lattice symmetries. We have extended our experimental study of the thermodynamic properties of the unconventional superconductor Sr_2RuO_4 under uniaxial stress by developing and optimizing a new setup for studying the elastocaloric effect at very low temperatures. This allowed direct access to the entropic phase diagram of Sr_2RuO_4.

The application of hydrostatic and uniaxial pressure represents a powerful tool for tuning material properties in a desired fashion. In the investigation of quantum materials, it is possible to deliberately tune competing ground states and facilitate the emergence of novel quantum phases. In topological materials, external pressure can be used to modify the band structure, for instance, to move Weyl points closer or further away from the Fermi level, or to alter the crystal structure, thereby enabling the transition between topological and conventional phases. In contrast to hydrostatic pressure, uniaxial pressure not only tunes a material, but also sometimes breaks lattice symmetries, resulting in fundamental changes in the material properties and the underlying physics. Below is an overview of our work.

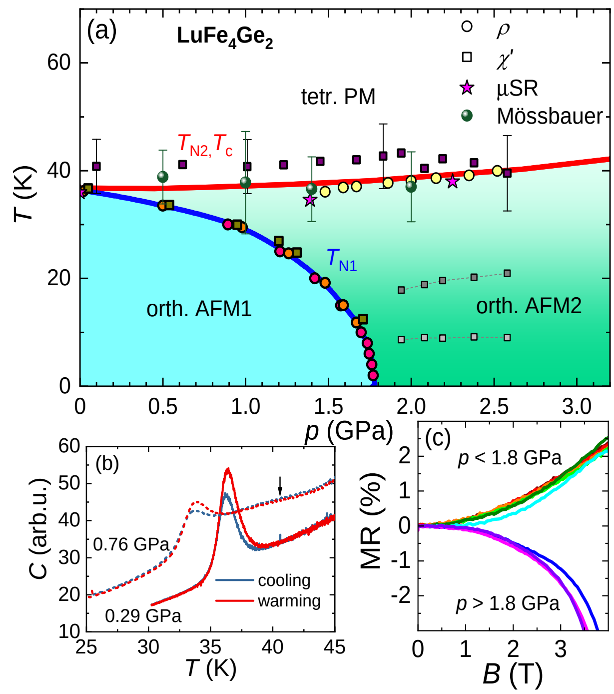

Fig. 1: (a) $T-p$ phase diagram of $LuFe_4Ge_2$. The transition temperatures obtained from electrical resitivity, magnetic susceptibility, μSR, and Mössbauer spectroscopy data are indicated. (b) Thermal hysteresis in the heat capacity confirms the first-order nature of the transition at T_{N1}. (c) Magnetoresistance (MR) measured at 2 K. Reproduced as per CC BY 4.0 [1] ©2023 The Authors, publ. by APS.

Interplay of structure and magnetism in $LuFe_4Ge_{12}$

Compounds with low-dimensionality and magnetic frustration are of particular interest due to enhanced quantum fluctuations. In this respect, $LuFe_4Ge_{12}$ is a prime candidate material. It consists of edge-sharing Fe tetrahedra forming chains along the crystallographic c axis, resulting in a quasi-one-dimensional structure.

We have used a unique combination of macroscopic magnetic, thermodynamic, and electrical transport measurements in our group, together with microscopic techniques such as muon spin resonance μSR, Mössbauer spectroscopy, and X-ray diffraction in long-term collaborations with our colleagues, in particular the Max Planck Partner Group of R. D. dos Reis, to reveal the unusual phase diagram of $LuFe_4Ge_{12}$ and to gain a comprehensive understanding of the underlying physical principles. Figure 1 summarizes the results of our study in a temperature – pressure phase diagram. It also presents example data of heat capacity and magnetoresistance under applied hydrostatic pressure [1]. At ambient pressure, $LuFe_4Ge_{12}$ exhibits a magnetostructural transition at approximately 38 K. While the structural transition coupled to a magnetic transition remains nearly pressure independent, a second magnetic transition emerges and is suppressed to zero temperature at about 1.75 GPa. Our observations, together with band-structure calculations, suggest a scenario where the magnetic and structural order parameters in $LuFe_4Ge_{12}$ are linked by magnetic frustration, causing the simultaneous magnetostructural transition. The results presented in Ref. [1] reveal an intriguing and unconventional interplay between structural and magnetic properties in $LuFe_4Ge_{12}$, which differs from the observations made in the AFe_2As_2 pnictide superconductors.

Topological features and the role of domain walls in ferromagnetic Weyl semimetals

Weyl fermions can be created by breaking the space-inversion or time-reversal symmetry of a Dirac material. In ferromagnetic (FM) CeAlSi, both symmetries are broken, while in most other cases only the space-inversion symmetry is broken. In CeAlSi quantum oscillation data indicate that the FM ordering modifies the Fermi-surface topology and also leads to an unusual drop in the quantum oscillation amplitude. In the ordered phase, we observe a pressure-induced suppression of the anomalous and the loop Hall effect (Figure 2). Since we can rule out alternative explanations, we demonstrate that a simplified model describing the scattering of Weyl fermions off FM domain walls can explain the observed topological features (Figure 3). Our findings highlight the importance of topologically non-trivial domain walls in ferromagnetic Weyl semimetals and propose a novel approach to the manipulation of topological properties in these materials [2, 3].

Signatures of the quasiquantized three-dimensional quantum Hall effect in $HfTe_5$

At ambient pressure, $HfTe_5$ is a Dirac semimetal that exists between strong and weak topological insulator phases. Upon cooling, a Lifshitz transition occurs at about 70 K, driving the system through a semiconducting phase and back to a semimetallic phase at low tem-

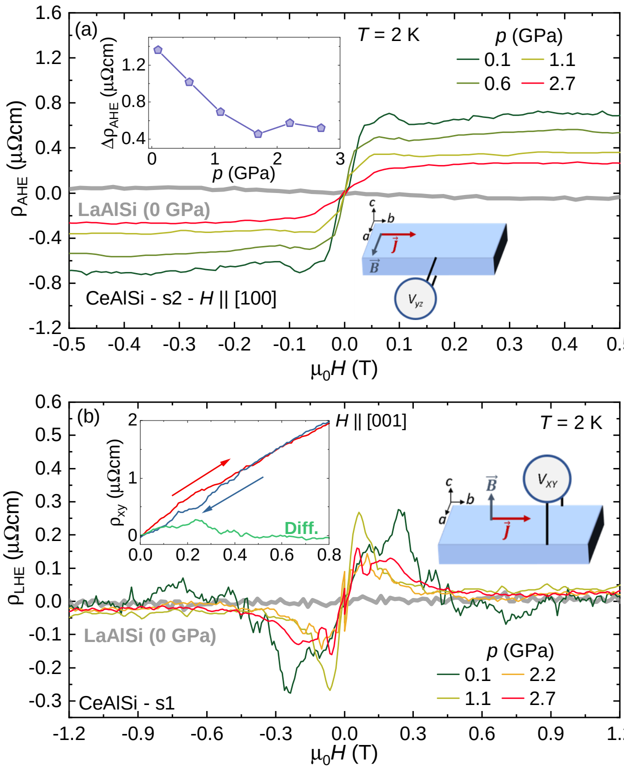

Fig. 2: (a) Anomalous Hall effect (AHE) and Loop Hall effect (LHE) of CeAlSi at 2 K vs. magnetic field $H \parallel [001]$ *for selected pressures and data of the non-magnetic reference material LaSiAl at zero pressure as gray lines. Reproduced as per CC BY 4.0 [2] ©2023 The Authors, publ. by APS.*

peratures. Furthermore, the quantum limit is reached at modest applied magnetic fields of only 1.4 T. In particular, plateaus in the Hall resistivity and minima in the longitudinal resistivity are observed, hallmarks of the quantum Hall effect (QHE). The origin of the quasi-three-dimensional (3D) QHE remains a subject of active debate, and two possible scenarios have been proposed.

In our study we investigate which of the two scenarios explains the quasi-3D QHE in $HfTe_5$. We do this by tuning $HfTe_5$ from a semimetallic ground state to an insulating state using hydrostatic pressure [5]. Our experimental data convincingly show that the QHE in $HfTe_5$ exists only in the semimetallic state. Both the Hall plateaus and the resistivity minima disappear before the material enters the pressure-induced insulating state. This supports the scenario based on a 3D metal with low carrier density and closed Fermi surfaces leading to quantized Hall plateaus and longitudinal resistivity minima in the quantum limit of 3D metals, as opposed to a scenario where a stack of 2D layers leads to a Fermi surface instability such as a charge density wave order, which would open a gap allowing the quantum Hall effect to take place.

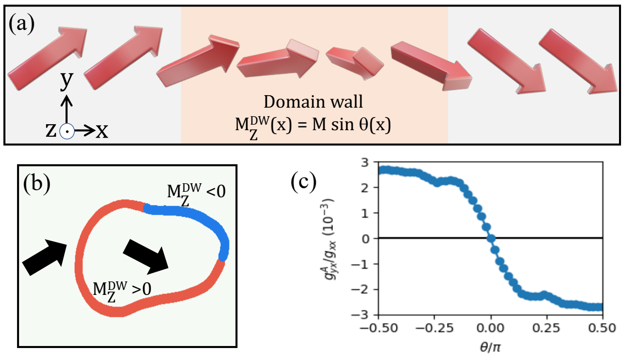

Fig. 3: (a) Domain wall between two magnetic domains showing twisted magnetization configuration. (b) Skew scattering of Weyl fermions off hysteretic domain wall loops. Black arrows represent Weyl fermion trajectories. (c) Calculated longitudinal Hall effect vs. out-of-plane tilt angle θ of the domain wall magnetization for CeAlSi. Reproduced as per CC BY 4.0 [2] ©2023 The Authors, publ. by APS.

Topological or conventional superconducting pairing symmetry in $MoTe_2$?

$MoTe_2$ has attracted significant interest as a promising candidate for the realization of a topological superconducting phase. At ambient pressure, $MoTe_2$ exhibits a first-order structural transition from a centrosymmetric $1T'$ to a non-centrosymmetric T_d phase at 250 K with temperature reduction. In the T_d phase, $MoTe_2$ is a type-II time-reversal invariant Weyl semimetal and shows low-temperature superconductivity. The presence of both non-trivial topology and superconductivity has led to extensive studies investigating the possibility of topological superconductivity. A crucial point for topological superconductivity is the presence of p-wave or s^{+-} pairing symmetry, which is extremely sensitive to disorder. Consequently, considerable research has been undertaken to investigate the pairing symmetry of $MoTe_2$ superconductivity.

By performing electrical transport experiments with small pressure steps under hydrostatic pressure, we have constructed the most comprehensive temperature-pressure phase diagram for $MoTe_2$ to date (see Figure 4). This allows us to draw robust conclusions regarding the possible superconducting order parameter(s). We find a sharp superconducting transition throughout phase diagram, especially in the mixed phase where the transition between the T_d and $1T'$ phases is suppressed to zero temperature. In this region, the T_c and the disorder scattering are significantly enhanced. Since conventional s^{++} superconductivity is robust to disorder and topological s^{+-} is strongly suppressed by disorder, it can be concluded that the superconducting pairing mechanism is conventional throughout the phase diagram of $MoTe_2$ [5].

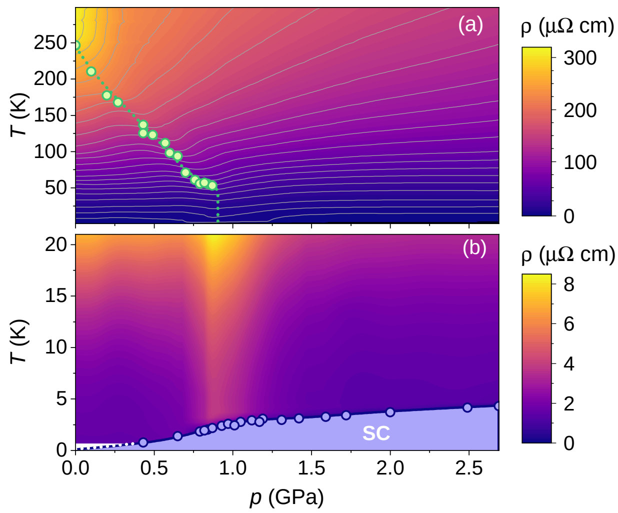

Fig. 4: Color maps of the electrical resistivity of $MoTe_2$. Circles denote phase transitions, gray lines are equipotentials, dotted lines are guides for the eye. Reproduced as per CC BY 4.0 [4] ©2023 The Authors, publ. by APS.

Phase diagram of Sr_2RuO_4: Insights from low temperature thermodynamic measurements

The application of uniaxial pressure not only tunes a material, but can also break lattice symmetries and provide access to energy scales that are not easily reached by applying hydrostatic pressure. In particular, uniaxial pressure combined with thermodynamic probes is a powerful tool to gain a deeper understanding of the unconventional superconductor Sr_2RuO_4. In the previous report we presented heat capacity results allowing us to establish the thermodynamic phase diagram and already put stringent constraints on theories of superconductivity in Sr_2RuO_4 [6]. We extended the heat capacity experiments to moderate magnetic fields and found a change of $H_{c2,\parallel c}(T)$ from a convex function of temperature to a concave function close to the Van Hove strain [9]. This finding is not well understood and will certainly motivate future work. Recently we were able to perform a high precision measurement of the elastocaloric effect (ECE) on strained Sr_2RuO_4 (see Figure 5), giving us access to the strain dependence of the entropy [7]. We observe a strong entropy quench upon entry into the superconducting state, in excellent agreement with a model calculation for pairing at the Van Hove point. We also obtain a quantitative estimate of the entropy change associated with entry into a magnetic state, which is observed in the vicinity of superconductivity. Our data together with sophisticated theoretical work of our collaborators allow to place a strong constraint on the possible superconducting order parameter, being a strong step towards finally identifying the order parameter in Sr_2RuO_4 [7, 8].

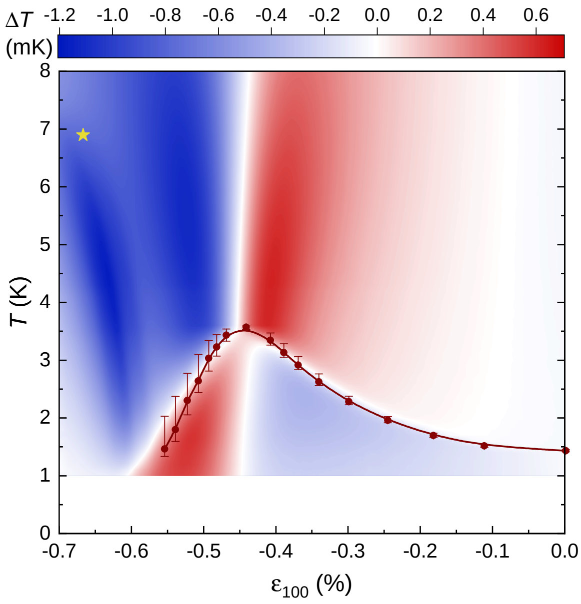

Fig. 5: Color map of the elastocaloric effect signal from Sr_2RuO_4. The circles correspond to T_c from heat capacity experiments [6]. Adapted as per CC BY 4.0 [7] ©2023 The Authors, publ. by Springer Nature.

External Cooperation Partners

R. D. dos Reis (Max Planck Partner Group; Brazilian Synchrotron Light Laboratory), C. Adriano and P. G. Pagliuso (UNICAMP, Brazil), M. Côté (Université de Montréal, Canada), H. H. Klauss (TU Dresden, Germany), A. Paramekanti (University of Toronto, Canada), C. Petrovic (Brookhaven National Laboratory, USA), A. W. Rost (University of St Andrews, UK), M. Garst and J. Schmalian (Karlsruher Institut für Technologie, Germany).

References

[1]* *Interplay of structure and magnetism in $LuFe_4Ge_2$ tuned by hydrostatic pressure, M. O. Ajeesh, P. Materne, R. D. dos Reis, K. Weber, S. Dengre, R. Sarkar, R. Khasanov, I. Kraft, A. M. Leon, W. Bi, J. Zhao, E. E. Alp, S. Medvedev, V. Ksenofontov, H. Rosner, H. H. Klauss, C. Geibel, and M. Nicklas, Phys. Rev. B **107** (2023) 125136 (Editors' suggestion),* https://dx.doi.org/10.1103/PhysRevB.107.125136

[2]* *Topological features in the ferromagnetic Weyl semimetal CeAlSi: Role of domain walls, M. M. Piva, J. C. Souza, V. Brousseau-Couture, S. Sorn, K. R. Pakuszewski, J. K. John, C. Adriano, M. Côté, P. G. Pagliuso, A. Paramekanti, and M. Nicklas, Phys. Rev. Res. **5** (2023) 013068,* https://dx.doi.org/10.1103/PhysRevResearch.5.013068

[3]* *Topological Hall effect in CeAlGe, M. M. Piva, J. C. Souza, G. A. Lombardi, K. R. Pakuszewski, C. Adriano, P. G. Pagliuso, and M. Nicklas, Phys. Rev. Matter. **7** (2023) 074204,* https://dx.doi.org/10.1103/PhysRevMaterials.7.074204

[4]* *Superconducting pairing symmetry in $MoTe_2$, M. M. Piva, R. Wawrzyńczak, Nitesh Kumar, L. O. Kutelak, G. A. Lombardi, R. D. dos Reis, C. Felser, and M. Nicklas, Phys. Rev. Matter. **8** (2024) L041202,* https://dx.doi.org/10.1103/PhysRevMaterials.8.L041202

[5]* *Importance of the semimetallic state for the quantum Hall effect in $HfTe_5$, M. M. Piva, L. O. Kutelak, R. Borth, Y. Liu, C. Petrovic, R. D. dos Reis, and M. Nicklas, Phys. Rev. Matter. **7** (2023) L111801,* https://dx.doi.org/10.1103/PhysRevMaterials.7.L111801

[6] *High-sensitivity heat-capacity measurements on Sr_2RuO_4 under uniaxial pressure, Y.-S. Li, N. Kikugawa, D. A. Sokolov, F. Jerzembeck, A. S. Gibbs, Y. Maeno, C. W. Hicks, J. Schmalian, M. Nicklas, and A. P. Mackenzie, High-sensitivity heat-capacity measurements on Sr_2RuO_4 under uniaxial pressure, Proc. Natl. Acad. Sci. USA **118** e2020492118, (2021).* https://dx.doi.org/10.1073/pnas.2020492118

[7]* *Elastocaloric determination of the phase diagram of Sr_2RuO_4, Li, Y. S., M. Garst, J. Schmalian, S. Ghosh, N. Kikugawa, D. A. Sokolov, C. W. Hicks, F. Jerzembeck, M. S. Ikeda, Z. H. Hu, B. J. Ramshaw, A. W. Rost, M. Nicklas, and A. P. Mackenzie, Nature **607** (2022) 276,* https://dx.doi.org/10.1038/s41586-022-04820-z

[8]* *Constraints on the superconducting state of Sr_2RuO_4 from elastocaloric measurements, G. Palle, C. Hicks, R. Valentí, Z. H. Hu, Y. S. Li, A. Rost, M. Nicklas, A. P. Mackenzie, and J. Schmalian, Phys. Rev. B **108** (2023) 094516 (Editors' suggestion),* https://dx.doi.org/10.1103/PhysRevB.108.094516

[9]* *Upper critical field of Sr_2RuO_4 under in-plane uniaxial pressure, F. Jerzembeck, A. Steppke, A. Pustogow, Y. K. Luo, A. Chronister, D. A. Sokolov, N. Kikugawa, Y. S. Li, M. Nicklas, S. E. Brown, A. P. Mackenzie, and C. W. Hicks, Phys. Rev. B **107** (2023) 064509,* https://dx.doi.org/10.1103/PhysRevB.107.064509

#michael.nicklas@cpfs.mpg.de