Weyl fermion monopnictides: from local quadrupole resonance and optical spectroscopy to bulk magneto thermal and magneto electrical transport

M. Baenitz#, M. Nicklas##, J. Sichelschmidt, U. Stockert, M. O. Ajeesh, C. Felser, E. Hassinger, D. Kasinathan, K. M. Ranjith, R. dos Reis, T. Kubo, A. P. Mackenzie, M. Schmidt, D. A. Sokolov, C. Shekhar, B. Yan, H. Yasuoka

The past years have seen an enormous rise of interest in the role of topology. Major discoveries have included two-dimensional graphene and the topological insulators (e.g. HgTe or Bi_2Se_3), whose topological properties require the existence of gapless surface states. Arguably, the most topical of the new classes of materials are Dirac and Weyl semimetals (DSM, WSM), which are predicted to host topologically protected states in the bulk. In the DSM (e.g., Cd_2As_3 or Na_3Bi), each node contains fermions of two opposite chiralities, whereas in the WSM a combination of non - centrosymmetric crystal structure and sizable spin-orbit coupling (SOC) causes the nodes to split into pairs of opposite chirality (Weyl points) [1]. Nb- and Ta-monopnictides are the ideal platform to study Weyl physics because they are completely stoichiometric and high quality single crystals can be grown.

In ideal Weyl semimetals, the Weyl points sit at the Fermi level (E_F) and the Weyl fermions are massless. In most of the materials, E_F does not exactly coincide with the Weyl nodes but if the nodes sit close enough to E_F, in a region of linear dispersion ($E \propto k$), the Weyl physics can still be observed in the excitations in the energy window k_BT. A key issue in our studies of the monopnictides is therefore to establish how close to the Fermi level the Weyl points sit and to estimate the range of energy over which the linear dispersion exists. This presents a considerable experimental challenge. The nodes appear in the electronic structure of the bulk, and the materials are fully three dimensional, so the surface-sensitive techniques that have yielded immense insight into other topological physics are not ideally suited to studying the Weyl points. Primarily, one would like to identify a probe that can excite the Weyl fermions and probe the linear dispersion $E \propto k$ indirectly via the energy dependence of the density of states DOS around the Fermi level, which is $N(E) \propto E^2$ for a Weyl node.

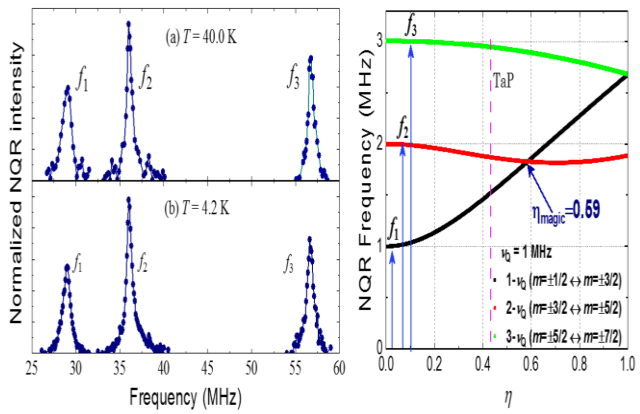

Fig.-1: Ta NQR lines at 40 K and 4.2 K in TaP (left), and the theoretical NQR frequencies (ν_Q) as a function of the asymmetry parameter of EFG (η) for I=7/2 (right). From the experimental frequencies (f_1, f_2 and f_3) ν_Q and η are determined as 19.265 MHz and 0.42, respectively, in rather good agreement with our DFT calculation [2].

Quadrupole resonance on TaP

The magnetic resonance method, in general, has the ability to probe N(E) and was applied successfully to systems like unconventional superconductors (e.g. UPt_3) or correlated magnetic semimetals (e.g. SmB_6 or $CeRu_4Sn_6$). In particular, for unconventional superconductors, the nuclear quadrupole resonance (NQR) spin-lattice relaxation (SLR) provides information about N(E) around E_F and allows us to distinguish between point nodes [$N(E) \propto E^2$] and line nodes [$N(E) \propto E$]. Therefore we applied NQR to TaP and established NQR as a novel probe for the bulk Weyl fermions and their low energy excitations [2]. In order

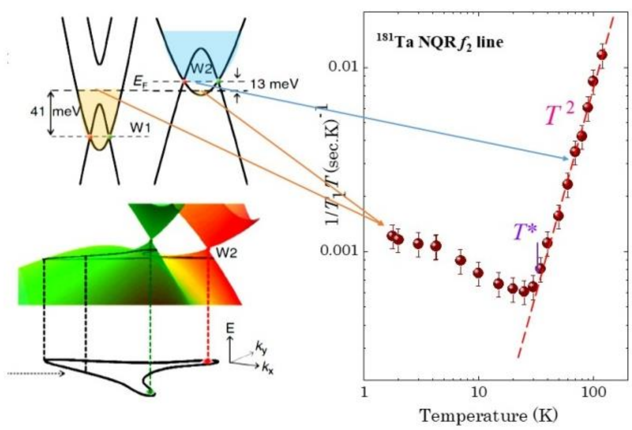

Fig.-2: (right) T- dependence of $1/T_1T$ obtained from Ta NQR in TaP. Above $T^* \sim 30$ K characteristic Weyl fermion excitations from the W2 points (dashed line) with $1/T_1T \propto T^2$ behavior could be observed. For $T < T^*$ the Korringa relaxation with a temperature dependent chemical potential is valid [2].

to examine the magnetic excitations, the T- dependence of the SLR rate ($1/T_1T$) is measured for the NQR-f_2 line ($\pm5/2 \leftrightarrow \pm3/2$ transition, Fig. 1). We find that there exist two regimes with quite different relaxation processes (Fig. 2, right). Above $T^* \approx 30$ K, a pronounced $1/T_1T \propto T^2$ behavior is found, which is attributed to the magnetic excitations at the Weyl nodes with temperature-dependent orbital hyperfine coupling (Fig. 2). It should be mentioned that the T-dependent hyperfine interaction is rather unique and directly related to the Weyl fermions and their strong orbital magnetism. Below T^*, the relaxation could be described by an extension of the Korringa theory in which a T-dependence of chemical potential is taken into account which finally leads to a $T^{-1/2}$ type dependence of $1/T_1T$ [private communication B. Dora (University Budapest) and M. Ogata (University Tokyo)] [2].

Optical spectroscopy on Ta-monopnictides

Another rather powerful probe for the bulk electronic states is the optical conductivity. We performed optical spectroscopy and band structure calculations of the typical Weyl semimetals TaP and TaAs [3]. The difference in the strongly temperature-dependent Drude weight (Fig. 3) revealed that the Weyl points of TaP are located relatively far from the Fermi level E_F, whereas those of TaAs are closer to E_F. Moreover, we

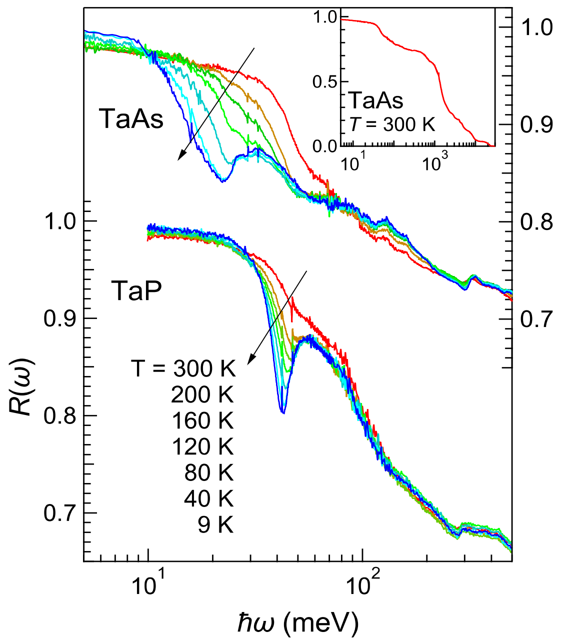

Fig.-3: Temperature-dependent reflectivity [R(ω)] spectra of TaP and TaAs in the photon-energy region of 5–500 meV. The two compounds both show a Drude behavior at the lowest-energies with clearly different pronounced temperature dependence. The inset shows the R(ω) spectrum of TaAs at 300 K in the whole measured region up to 30 eV [3].

could observe a ω-linear dependence of the optical conductivity spectra in both materials, which indicates Dirac-like linear band dispersions. These observations, and the overall agreement with our band calculations, provide important insights into the electronic structure of typical Weyl states in TaP and TaAs.

Thermal and electrical magneto transport studies

We contributed to the discovery of the extremely large non-saturating magnetoresistance in NbP by the Felser department [4], which attracted enormous attention in the condensed-matter physics community. NbP is a particular interesting material since it has been predicted to combine the hallmarks of a Weyl semimetal and a classical semimetal. Due to its ultrahigh mobility quantum oscillations are visible in different physical probes down to very small magnetic fields of only about 1 T [4]. This allows for a detailed and comprehensive study of the Fermi-surface topology in this family of materials at relatively low magnetic fields. The knowledge of the Fermi-surface topology and the electronic band structure is at the basis of the understanding of the physics in Weyl semimetals since it allows to determine the position of the Weyl points which is essential for the possible observation of Weyl physics. We determined the Fermi-surface topology of TaP [5] and NbP [6] in collaboration with the Felser department, the MPRG of E. Hassinger and the Dresden High Magnetic Field Laboratory. For NbP the data show that the W1-type Weyl points lie deep below the Fermi-surface, while the W2-type Weyl points are supposed to be only 5meV above.

Thermal transport study in NbP

Thermal measurements as a function of temperature and magnetic field are useful to study semimetals with light carriers and multiband contributions to the DOS. The specific heat is a direct probe for the total DOS. On the other hand the transport properties thermos-power S(T) and thermal conductivity κ(T) are determined by both the DOS and the mobilities of the relevant carriers (or in general the scattering processes). For NbP single crystals we found huge quantum oscillations in all three properties as a function of magnetic field reflecting the presence of rather light carriers in the clean semimetal. The oscillation frequencies are in rather good agreement with deHaas van Alphen measurements. Furthermore, the thermopower exhibits a huge field dependence (Fig. 4) and the Wiedemann Franz law relating thermal and electrical transport is strongly violated in NbP. These observations could be caused by the strong field dependence of the charge carrier mobility and/or a renormalization of electron-phonon scattering due to the chirality of the Weyl fermions. Our thermal transport results, which are among the first ones on Weyl semimetals, demonstrate that thermopower and thermal conductivity are highly interesting probes for this class of materials and stimulate further studies [8].

Pressure tuning the Fermi surface topology of NbP

We used Shubnikov–de Haas (SdH) oscillations in the magnetoresistance combined with ab initio calculations to investigate the pressure variations of the electronic band structure in NbP [7].

Our results indicate a robust electronic band structure without characteristic changes in the topology in the pressure range up to 2.8 GPa. Strong changes in the amplitudes of the SdH oscillations indicate subtle changes in the Fermi-surface topography in the vicinity of the extremal areas. Figure 5b and 5c display the FFT of the SdH oscillations for ambient pressure and 2.47 GPa, respectively. Furthermore, we found that the W2-type Weyl points move toward the Fermi energy upon increasing pressure, while the energy difference between the W1-type Weyl points and the Fermi energy is almost unaffected. This led us to speculate that the chiral anomaly effect may be established at higher pressures. In summary, our study demonstrated the power of combining quantum-oscillation studies and band-structure calculations to investigate pressure effects on the Fermi surface topology in Weyl semimetals under hydrostatic pressure.

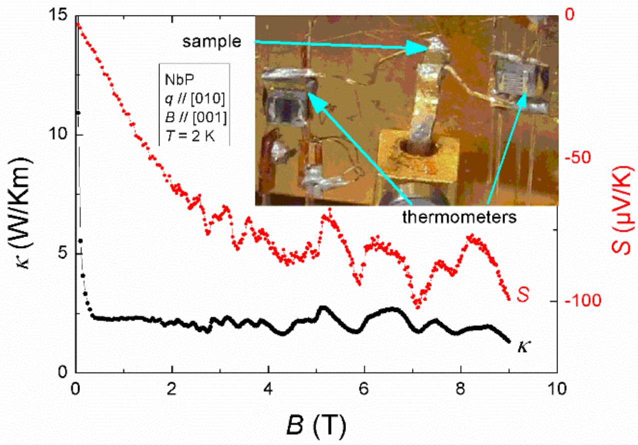

Fig.-4: Field dependence of the adiabatic thermos-power (right scale) and the thermal conductivity (left scale) in NbP single crystal [8].

Origin of the negative longitudinal magneto-resistance in Weyl semimetals

A property of Weyl fermions is the chiral or Adler-Bell–Jackiw anomaly, a chirality imbalance in the presence of parallel magnetic and electric fields, which leads to one of the characteristic fingerprints of Weyl semimetals, the negative longitudinal magneto-resistance (MR). A negative longitudinal MR has been reported in a variety of Weyl semimetals. However, its origin does not necessarily need to be related to the chiral anomaly [5, 9].

In our longitudinal MR setup, we observed different magnetic-field dependences for different voltage contacts in NbP as presented in Fig. 6. This directly hints at an inhomogeneous current distribution in the sample. We have even observed a negative voltage for small misalignments between the current and magnetic-field directions. With help of a detailed finite element

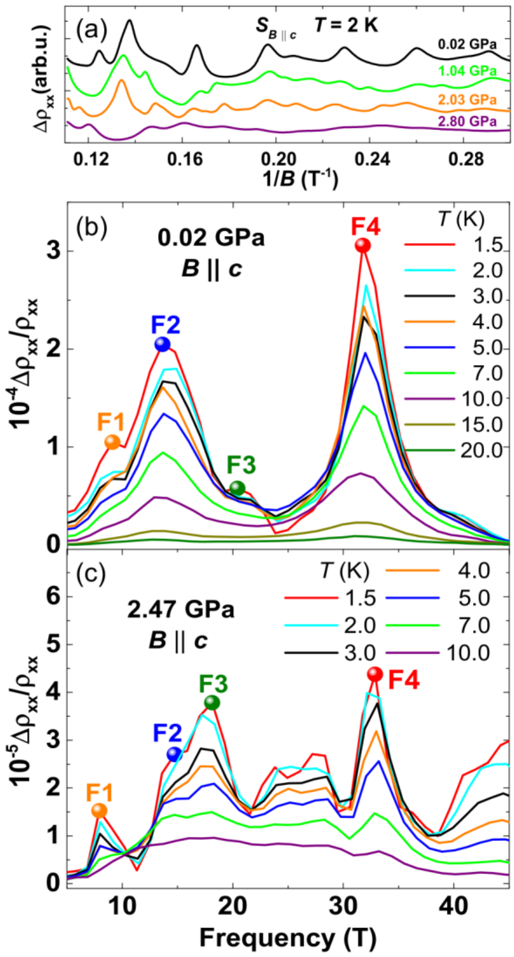

Fig.-5: (a) Shubnikov–de Haas oscillations as a function of the inverse magnetic field taken at different pressures for B||c. (b) and (c) Temperature dependence of the fast Fourier transform of the SdH oscillations (taken from Ref. 7).

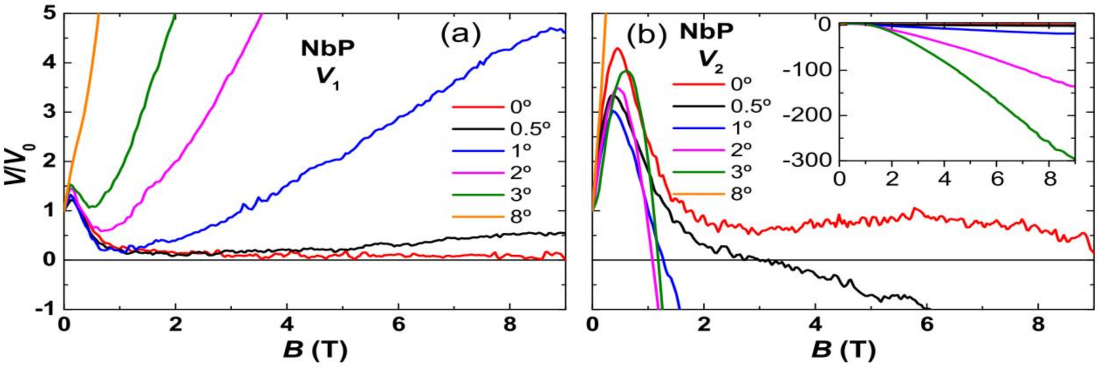

Fig.-6: (a) and (b) magnetic field dependence of the voltage for different angles θ between the magnetic field and the electrical current for the two contact pairs V1 and V2, respectively. The inset in (b) displays the region where a negative voltage is observed for the contact pair V2 (taken from Ref. 9).

simulation of the current distribution in the sample (Fig. 7), we strongly suggested that a current-jetting effect based on the large field-induced resistance anisotropy causes the observed negative longitudinal MR in NbP, NbAs, TaP and TaAs and not the chiral anomaly [9].

We learned that measurements of the longitudinal MR in materials with field-induced resistance anisotropy is not straightforward and careful analysis is required before intrinsic physical properties, such as the chiral anomaly in Weyl semimetals, are extracted.

External Cooperation Partners

B. Dora & F. Simon (Budapest University of Technology and Economics, Hungary); J. P. Heremans (Ohio State University, USA); M. Ogata (University of Tokyo, Japan) Shin – ichi Kimura (Osaka University, Japan); H. Tou (Kobe University, Japan); J. Wosnitza (Helmholtz-Zentrum Dresden-Rossendorf, Germany)

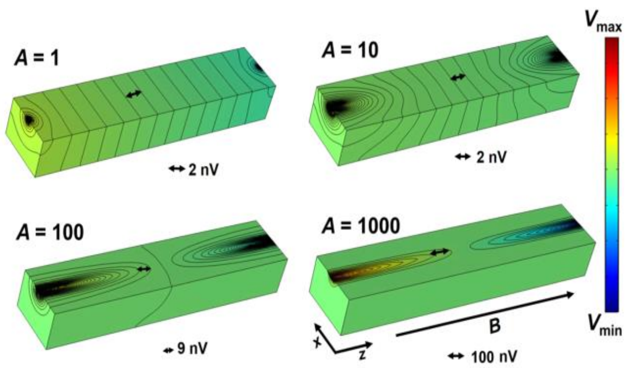

Fig.-7: Calculated potential distribution for different resistance anisotropies A = ρ_{xx}/ρ_{zz} = 1, 10, 100, and 1000 for a sample with point-like current electrodes. The current contacts are in the middle of the upper edge of the face sides. B is aligned with the current electrodes, ρ_{zz} (ρ_{xx}) is the resistance parallel (perpendicular) to B. The lines are contour lines of the equipotential surfaces (taken from Ref. 9).

References

[1] *Topological Materials:Weyl Semimetals, B. Yan and C. Felser, Annu. Rev. Condens. Matter Phys.* ***8*** *(2017) 337–54.*

[2]* *Emergent Weyl Fermion Excitations in TaP Explored by* ^{181}*Ta Quadrupole Resonance, H. Yasuoka, T. Kubo, Y. Kishimoto, D. Kasinathan, M. Schmidt, B. Yan, Y. Zhang, H. Tou, C. Felser, A. P. Mackenzie and M. Baenitz, Phys. Rev. Lett.* ***118*** *(2017) 236403.*

[3]* *Optical signature of Weyl electronic structures in tantalum pnictides TaPn (Pn = P, As), S. Kimura, H. Yokoyama, H. Watanabe, J. Sichelschmidt, V. Süß, M. Schmidt and C. Felser, Phys. Rev. B.* ***96*** *(2017) 075119.*

[4]* *Extremely large magnetoresistance and ultra high mobility in the topological Weyl semimetal candidate NbP, C. Shekhar, A. K. Nayak, Y. Sun, M. Schmidt, M. Nicklas, I. Leermakers, U. Zeitler, Y. Skourski, J. Wosnitza, Z. Liu, Y. Chen, W. Schnelle, H. Borrmann, Y. Grin, C. Felser and B. Yan, Nature Phys.* ***11*** *(2015) 645.*

[5]* *Negative magnetoresistance without well-defined chirality in the Weyl semimetal TaP, F. Arnold, C. Shekhar, S.-C. Wu, Y. Sun, R. D. dos Reis, N. Kumar, M. Naumann, M. O. Ajeesh, M. Schmidt, A. G. Grushin, J. H. Bardarson, M. Baenitz, D. Sokolov, H. Borrmann, M. Nicklas, C. Felser, E. Hassinger and B. Yan, Nature Comm.* ***7*** *(2016) 11615.*

[6]* *Quantum oscillations and the Fermi surface topology of the Weyl semimetal NbP, J. Klotz, S.-C. Wu, C. Shekhar, Y. Sun, M. Schmidt, M. Nicklas, M. Baenitz, M. Uhlarz, J. Wosnitza, C. Felser and B. Yan, Phys. Rev. B* ***93*** *(2016) 121105(R).*

[7]* *Pressure tuning the Fermi surface topology of the Weyl semimetal NbP, R. D. dos Reis, S. C. Wu, Y. Sun, M. O. Ajeesh, C. Shekhar, M. Schmidt, C. Felser, B. Yan and M. Nicklas, Phys. Rev. B* ***93*** *(2016) 205102.*

[8]* *Thermopower and thermal conductivity in the Weyl semimetal NbP, U. Stockert, R. D. dos Reis, M. O. Ajeesh, S. J. Watzman, M. Schmidt, C. Shekhar, J. P. Heremans, C. Felser, M. Baenitz and M. Nicklas, J. Phys.: Condens. Matter* ***29*** *(2017) 325701.*

[9]* *On the search for the chiral anomaly in Weyl semimetals: the negative longitudinal magnetoresistance,*

PHYSICS OF QUANTUM MATERIALS

R. D. dos Reis, M. O. Ajeesh, N. Kumar, F. Arnold, C. Shekhar, M. Naumann, M. Schmidt, M. Nicklas and E. Hassinger, New J. Phys. ***18*** *(2016) 085006.*

[#] Michael.Baenitz@cpfs.mpg.de
[##] Nicklas@cpfs.mpg.de