Exceptional transport properties of topological semimetals and metals

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Topological materials (TMs) represent a family of new quantum materials, and the quantum Hall effect is the first realized topological phenomenon in condensed-matter physics. Band inversion occurs in topological insulators, and symmetry allows the bulk gap to fully reopen. At the surface of the three dimensional topological insulator, bands cross linearly (Dirac cone) and the crossing point is protected by time reversal symmetry. In contrast to Weyl and Dirac semimetals, the Dirac cone forms in the bulk, wherein the nodal points are two- and four-fold degenerate, respectively. Quasiparticles residing at these nodal points are equivalent of Dirac and Weyl fermions in particle physics. Recently, many other topological materials like nodal line semimetals, double Weyl semimetals, triple point Fermion metals, etc. have also been discovered. Topology in the band structure makes these materials interesting by imparting many exotic physical characteristics. Our group is involved in crystal growth and transport property measurements at very low temperatures and high magnetic fields to understand the effect of topology in materials. Among the first verified Weyl semimetals, NbP shows ultrahigh mobility of 5,000,000 cm^2 V^{-1} s^{-1}, low effective mass, and extremely large magnetoresistance (MR). We find both extremely large MR (200 million % in 63 T at 2.5 K) and ultralow resistivity (3 nΩ·cm at 2 K) simultaneously in the Weyl semimetal. In the Heusler family, the Weyl semimetal GdPtBi is significant because it exhibits a chiral anomaly, anomalous Hall conductivity (60 Ω^{-1}cm^{-1}) with a large anomalous Hall angle (23%), planar Hall effect, and linear optical conductivity in a large energy range well above the transition temperature. Triple point fermionic MoP behaves like an ultra-pure metal, wherein the long-lived electrons in MoP flow collectively like a liquid. The topological semimetal LaBi exhibits quasi-two-dimensional electron transport, and the nodal-line semimetal HfSiS exhibits a non-trivial π-Berry phase. These unusual transport properties hint at the existence of fermions as a quasiparticle in condensed matter systems.

Materials are conventionally divided into metals, semiconductors, and insulators. Through the lens of topology, materials can be reclassified as either topologically trivial or non-trivial. Depending on the inherent symmetry and touching point of bands in a particular compound, two- and four-fold degenerate points are classified as Weyl and Dirac types, respectively, which are equivalent to Dirac and Weyl fermions in particle physics. Materials possessing such fermions as quasiparticles are known as Dirac semimetal (DSM) and Weyl semimetal (WSM). In solid-state band structures, Weyl fermions exist as low-energy excitations of the Weyl semimetal, in which bands disperse linearly in three-dimensional (3D) momentum space through a node termed a Weyl point. Weyl points act as monopoles in momentum space with a fixed chirality that behave as a source ("+" chirality) or a sink ("–" chirality); a non-vanishing Berry curvature exists between them. The Berry curvature is a quantity that can be used to characterise topological entanglement between the conduction and valence bands, which is equivalent to a magnetic field in momentum space.

Due to the topology of the bulk bands, topological surface states appear on the surface and form exotic Fermi arcs in WSM. All bands in a DSM are doubly degenerate, while the degeneracy is lifted due to breaking of inversion symmetry, breaking of time-reversal symmetry, or both in the WSM. In a type-I WSM, the Fermi surface (FS) shrinks to zero at the Weyl points when the Fermi energy is sufficiently close to the Weyl points, while the Weyl point acts as the touching point between electron and hole pockets in the FS due to the strong tilting of the Weyl cones in a type-II WSM. In condensed matter systems such as non-trivial semimetals and metals exhibit novel, low-energy fermionic excitations which are associated with bands that are linearly dispersed around a crossing point.

Pnictide TaAs-family of Weyl semimetals

At the beginning of this decade, the WSM was predicted to be a class of time reversal symmetry breaking (magnetism) compounds, including pyrochlore iridates (such as $Y_2Ir_2O_7$) and $HgCr_2Se_4$. These compounds, however, have not been confirmed experimentally.

A Major breakthrough came in early 2015 when a WSM was predicted in the transition metal pnictides NbP, TaP, NbAs, and TaAs (TaAs-family). These compounds have non-centrosymmetric tetragonal crystal structure with $I4_1md$ space group (no. 109) that breaks inversion symmetry. Each of them show twelve pairs of Weyl points throughout the Brillouin zone (BZ).

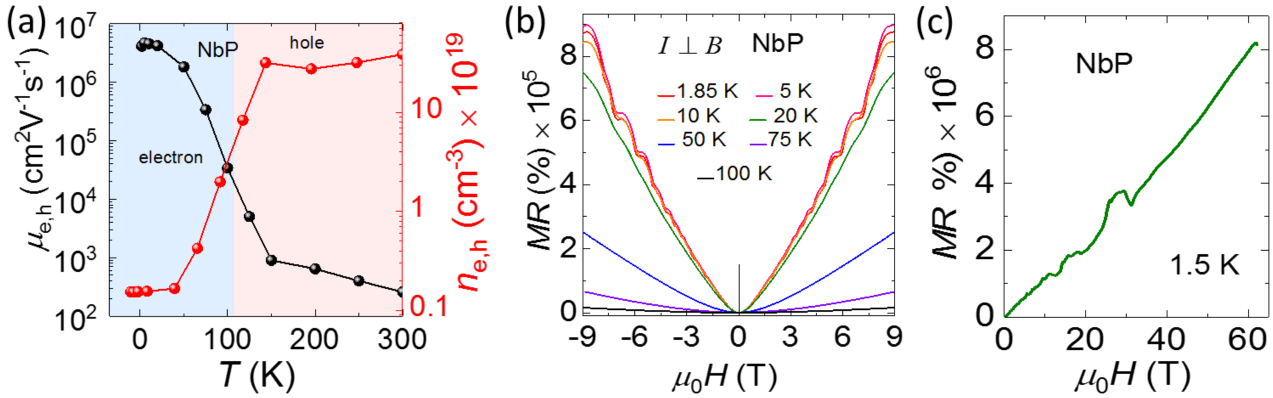

Fig.-1: (a) Temperature dependence of the mobility (left axis) and the carrier density (right axis). (b) Transverse magnetoresistance measured at different temperatures. (c) Measured MR in fields up to 62 T at 1.2 K.

Since we have vast expertise of growing single crystals, we grew large size (mm order) crystals of all the members of the TaAs-family by chemical vapor transport within a month of the original prediction. After full characterisation, we found that all these crystals were of high quality and exact composition. To observe the signature of Weyl fermions, we measured the electrical transport in extreme conditions of very low temperature and very high magnetic field for all compounds. All compounds exhibited similar transport behaviour, and the results from NbP are mentioned here as an example [1]. The field dependence of the Hall resistivity $\rho_{xy}(H)$ exhibits a nonlinear behaviour that indicates involvement of more than one type of charge carrier in the transport properties. NbP exhibits a negative Hall coefficient R_H up to 125 K, which changes sign for temperatures above 125 K. We use the single-carrier Drude model $n_{e,h}(T) = 1/[eR_H(T)]$ to calculate the carrier density, and we used $\mu_{e,h}(T) = R_H(T)/\rho_{xx}(T)$ to estimate the mobility, where $n_e(n_h)$ and $\mu_e(\mu_h)$ are the charge density and mobility of electrons (holes), respectively. The electron carrier concentration was found to be 1.5 $\times 10^{18}$ cm^{-3} at 1.85 K and increases slowly with temperature, exhibiting a semimetal-like or very small gap-like behaviour (Fig. 1a). The mobility plays a major role in charge transport in a material and, consequently, determines the efficiency of various devices. Here, NbP exhibits an ultrahigh mobility of 5×10^6 cm^2 V^{-1}s^{-1} at 1.85 K. A large MR is usually associated with high mobility. Fig. 1b shows the magnetoresistance (MR) measured in transverse magnetic fields up to 9 T at different temperatures. We find that NbP exhibits an extremely large MR 8.5×10^5% at 1.85 K. To locate the positions of Weyl points experimentally, we conducted detailed fermiology investigations and constructed the Fermi surface in TaP. We found two pairs of Weyl points (W1 and W2), which are located at +13 meV (above E_F) and -41 meV (below E_F) [2]. To explore the deep physics, we provided these transition metal pnictides to Oxford University and the Weizmann Institute of Science for angle-resolved photoemission spectroscopy (ARPES) measurements and scanning tunneling microscopy (STM) examinations, respectively. These groups independently found the Weyl points and Fermi arcs as the first experimental verification of Weyl semimetals. Besides these properties, chiral anomaly (longitudinal negative magnetoresistance) is considered clear evidence of Weyl fermions. Despite the absence of an independent Fermi-surface around the Weyl points in TaP, an apparent negative longitudinal MR is detected, which is attributed to the current jetting effect [2].

Robust Weyl semimetals WP_2 and MoP_2

After observing excellent properties in transition metal pnictides, we searched for other simple compounds. Recently, we found that WP_2 and MoP_2 could host robust type-II Weyl points since even parity Weyl points are located closer together and remain far from the odd ones in the Brillouin zone (Fig. 2a). These compounds crystallise in a non-symmorphic orthorhombic structure $Cmc2_1$ space group (no. 36). Single crystals were grown using the vapor transport method. All the crystals grow with needle-shape along the *a*-axis (inset of Fig. 2b). Both compounds exhibit similar properties, but WP_2 is superior to MoP_2 [3]. The resistivity of WP_2 in the absence of a magnetic field is linear in temperature from high temperature like a metal, and the resistivity eventually reaches the smallest value of 3 nΩcm at 2 K. The residual resistivity ratio of 25,000 and residual resistivity of 3 nΩ cm in WP_2 are the most extreme recorded values

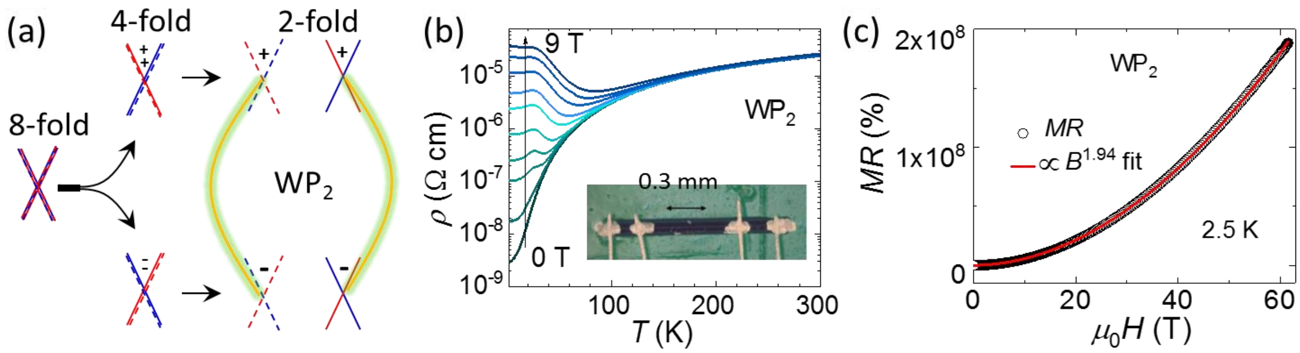

Fig.-2: (a) Evolution of Weyl points in WP_2 upon symmetry breaking. (b) Temperature dependence of the resistivity of WP_2 at various magnetic fields. (c) Measured and fitted MR in fields up to 63 T at 2.5 K.

found in any binary compound. Despite a very high conductivity, we observe extremely high MR and a typical semimetal-like upturn in resistivity in WP_2 (Fig. 2c). A large suppression of charge carrier back-scattering leads to MR of 200 million % in 63 T at 2.5 K, which is highly anisotropic due to the anisotropic Fermi surface. The large suppression of charge carrier backscattering also gives rise to an extraordinarily large mean free path of 0.5 mm. These properties are likely a consequence of the novel Weyl fermions expressed in this compound.

Ideal half Heusler Weyl semimetals

A large pool of Heusler compounds provides a unique platform to discover topological materials since these materials possess a wide range of tuneable structural and physical functionalities. Thanks to our theoretical group, we predicted the existence of materials with topological properties ranging from topological insulators to unconventional fermions in half Heuslers. We recently demonstrated topological surface states and Dirac points in several such Heusler compounds through our ARPES investigations [4]. This family of compounds shows topological surface states and Dirac cones, as well as their locations with respect to the Fermi level within the Heusler family. After deep theoretical and experimental investigations on various half Heusler compounds, we found that GdPtBi and NdPtBi are the strongest Weyl fermion candidates. These compounds are members of the RPtBi series, where R is a lanthanide or Y. These compounds have asymmetric inversion crystal structure ($F\bar{4}3m$ space group, No. 216), and their unit cell is composed of

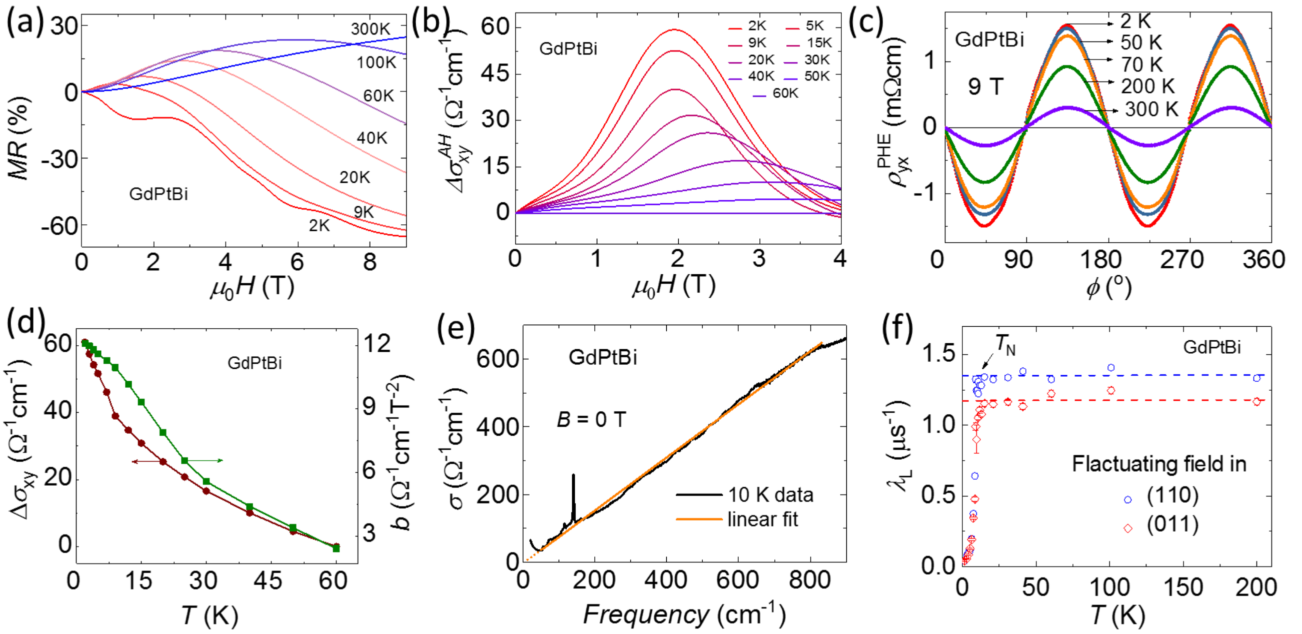

Fig.-3: Different properties of GdPtBi. (a) Negative magnetoresistance (MR) (chiral anomaly). (b) Anomalous Hall effect (AHE). (c) Planar Hall effect (PHE). (d) Correlation of negative MR and AHE. (e) Linear optical conductivity at 10 K. (f) Longitudinal zero-field relaxation rate measured for the muon spin polarization components without noticeable correlation effects above T_N.

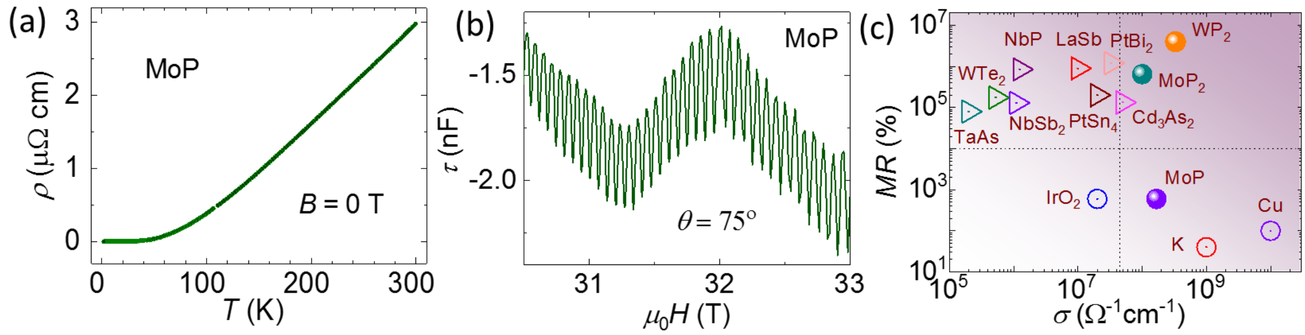

Fig.-4: (a) Temperature dependent resistivity of MoP. (b) Magnetic torque signal nearly along c-axis of MoP showing very dense dHvA oscillations with frequency of 14.5 kT. (c) A comparison of MR and conductivity of some well-known topologically trivial and nontrivial metals.

three interpenetrating *fcc* lattices. The structure can be described as a metallic multilayer formed from successive atomic layers of rare-earth, platinum, and bismuth along the [111] direction. GdPtBi and NdPtBi are antiferromagnetic (AFM) metals at temperatures below their corresponding Néel temperatures, where T_N = 9.0 K and 2.1 K, respectively. We grew large size crystals of these compounds using the self-flux method, which normally grow along the [111] direction. We gathered measurements of the field-dependent Hall resistivity ρ_{xy} and the longitudinal resistivity ρ_{xx} for the well-oriented crystal GdPtBi at various temperatures. Transport through the Fermi arc is another hallmark of the so-called chiral anomaly (negative MR) in a WSM, which arises from topological charge pumping within a pair of Weyl points when electric and magnetic fields are applied in parallel. This gives rise to several unusual properties, including a longitudinal negative MR which is clearly observed in GdPtBi (Fig. 3a) [5]. WSMs are anticipated to exhibit an anomalous Hall effect (AHE) due to the net Berry flux that is proportional to the separation between Weyl points of opposite chirality. We observed a large anomalous Hall conductivity of 60 $\Omega^{-1}cm^{-1}$ at 2 K (Fig. 3b) with an anomalous Hall angle up to 23% [5]. We find that the AHE and chiral anomaly have a similar temperature dependence that indicates their common origin (Fig. 3d). Furthermore, Fig. 3c shows a planar Hall signal dominated by chiral anomaly [6]. Interestingly, we observed all these properties well above T_N and, hence, their origin cannot be attributed to magnetism. This is clear from our μSR analyses in the paramagnetic state above T_N. The muon spin depolarization rate λ_L is temperature-independent and isotropic for two orthogonal muon spin polarization components. The overall behaviour suggests the absence of any significant magnetic correlations in GdPtBi above T_N (Fig. 3f). Furthermore, GdPtBi exhibits a linear optical conductivity over a broad range of frequency at 10 K (Fig. 3g) [7]. The aforementioned electrical properties were also observed in its sister compound NdPtBi, while its nonmagnetic variant YPtBi does not show such properties. We conclude here that parabolic bands split due to exchange coupling/ Zeeman splitting leads to a Weyl point close to the Fermi energy, which is the origin of these exotic transport properties in GdPtBi and NdPtBi.

Triple point topological metal MoP

Depending on the symmetry of a particular compound, the crossing points can be several-fold degenerate. It is not restricted to two- and four-fold degeneracy because Poincare symmetry is not restricted in condensed matter systems. Therefore, new materials going beyond Weyl and Dirac semimetals with higher band degeneracies that have no high-energy counterpart have been proposed. We grew the MoP crystals as an example of such a new, unconventional fermion compound, which is a member of the triple-point fermion family. This compound shows highly metallic characteristics with remarkably low resistivity (6 nΩ·cm) and high mobility (2.4 × 10^4 $cm^2V^{-1}s^{-1}$) at 2 K (Fig. 4a) [8]. The measured de-Haas van-Alphen oscillations in MoP exhibit beating patterns due to spin-orbit coupling-induced band splitting. We detected a large oscillation frequency of 14.6 kT the field is close to the *c*-axis (Fig. 4b). The effective mass *m** values for different bands show strong variation (from 0.23m_0 to 1.12m_0), which underlines a complex band structure. A large difference between classical and quantum carrier lifetimes, high carrier charge density accompanied by large mobility, and low resistivity make MoP an exotic nontrivial topological metal. We realized that with a similar order of conductivity in conventional metals, topological

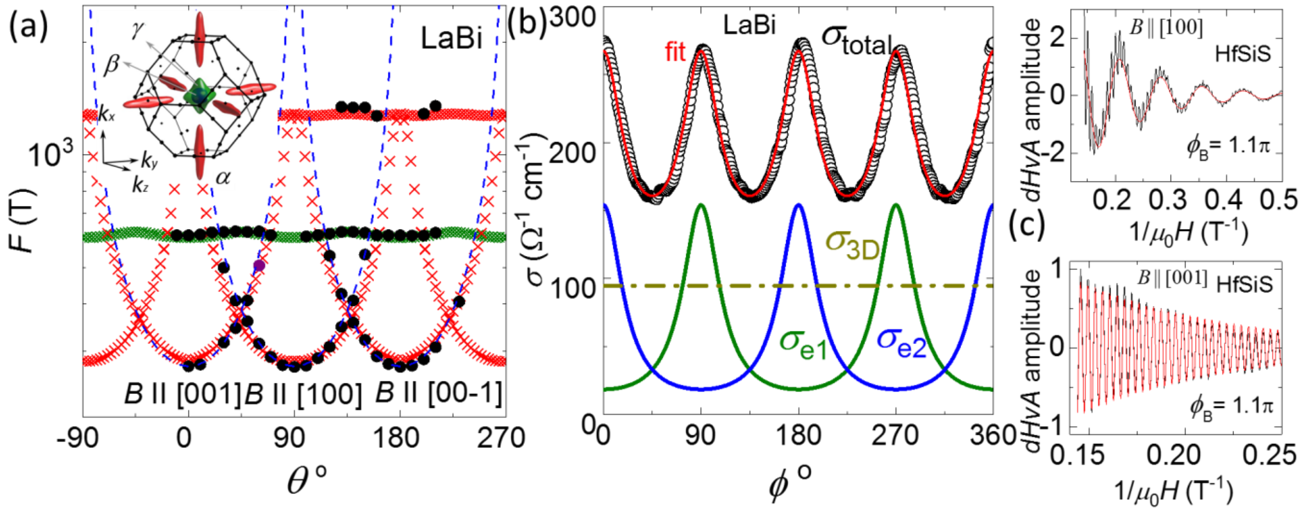

Fig.-5: Topological metals. Angular dependence of the SdH oscillation frequencies (experimental-solid circles and calculated-cross) in LaBi. (b) Conductivity of LaBi at 2 K and 9 T as a function of φ, thus inferring the contributions from different Fermi pockets. (c) Fitting of the dHvA oscillations in HfSiS by the LK formula to determine the Berry phase at different frequencies, where the black line is the measured data and the red line is a fit to theory.

materials show extremely high MR that hints at the role of their high mobility.

Topological metals

LaBi is a semimetal and contains an inverted band gap below the Fermi level. It exhibits pseudo-two-dimensional transport [9] originating from bulk states rather than the topological surface states. We observe a magnetic field-induced electronic valley polarization of 60% at 2 K and 9 T (Fig. 5a), which is attributed to the highly anisotropic electron Fermi surfaces (Fig. 5b) [10].

HfSiS is a nodal line semimetal which also host several Dirac cones near the Fermi level. It contains a large carrier density that is higher than most of the known semimetals and exhibits massive amplitudes of dHvA oscillations up to 20 K in 7 T (Fig. 5c). This assists with estimation of the Berry phase. We observe a nontrivial π-Berry phase in HfSiS depicting topological Dirac-type band dispersion, which forms the hybridization of p_x + p_y and $d_{x^2-y^2}$ orbitals in a square-net plane consisting of Si and Hf atoms, respectively [11].

Outlook

By growing high quality single crystals and gathering electrical transport measurements, we unveil topological characteristics of exotic topological materials. We further extend our expertise towards understanding various topological phases and their mutual correlation by employing methods like the laser assisted floating zone crystal growth technique and transport measurements under hydrostatic pressure.

External Cooperation Partners

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