Topological Magnetic Heuslers: Role of Symmetry and the Berry Phase
Kaustuv Manna*, Claudia Felser, Johannes Gooth, Satya N. Guin, Ting-Hui Kao, Jürgen Kübler, Lukas Mückler, Jonathan Noky, Chandra Shekhar, Rolf Stinshoff, Yan Sun

Topological materials are a new class of quantum materials where gapless electronic excitations are observed, which are protected by the crystal symmetry and topology of the bulk band structure. It was recently realized that the Berry phase is deeply connected with these topological states of matter and can be easily tuned via the crystal symmetry, band structure, and by engineering the number of valance electrons in topological Heuslers. We synthesized single crystals of these various topological Heuslers and investigate their magnetic, electrical, and thermal transport properties, as well as the electronic band structure with angle-resolved photoemission spectroscopy experiments. We performed topological band theory calculations in order to gain a better understanding of these materials. We found that Co$_2$MnGa, the first three-dimensional topological magnet, shows one of the highest anomalous Hall conductivities (AHC) ~ (1600 Ω$^{-1}$cm$^{-1}$) at 2 K and the highest known anomalous Hall angle (AHA) (up to 12%) of any magnetic Heuslers at room temperature. We observed a remarkably high anomalous Nernst thermopower $\gamma_N^\delta$ of ~6.0 µV K$^{-1}$ at 1 T magnetic field and at room temperature, which is beyond the magnetization-scaling relation in any compound reported so far in literature. We also show how the AHC in topological Heuslers can be tuned from zero to a colossal value that is independent of the sample’s magnetization. We also found that the metallic ferrimagnetic compound Mn$_2$CoGa compensates the AHC to zero due to topological reasons, regardless of its high magnetic moment (~2 µB). We illustrate how a metallic magnet can be converted to a topologically trivial semiconductor by symmetry engineering and extend this understanding to a series of compounds like spin-gapless semiconductors.

Topological semimetals have drawn tremendous attention in recent years due to their fascinating properties and promise for potential applications in modern technology. As a highly tunable material, Heusler compounds serve as a fertile playground where various novel topological properties, like the Weyl, Dirac, and nodal line semimetals, and spin-structured topological states like skyrmions can be investigated.

In this collaborative work, we focus to understand the physical properties of various topological magnetic Heuslers. Our investigation led to a deeper understanding of the tunability of these properties, which might result in the discovery of novel phenomenon like topological superconductors, Majorana fermions, or the quantum anomalous Hall effect at room temperature. For this investigation, we employed X-ray and Laue diffraction, magnetization, electrical and thermal transport, angle-resolved photoemission spectroscopy (ARPES) measurements, and we performed electronic band structure calculations using density functional theory.

In our study, we use the Berry curvature as one guiding principle to tune the topological properties of Heusler compounds. Berry curvature is the equivalent of the magnetic field in the parameter space that describes entanglement of the valence and conduction bands in an energy band structure. It resides as a strong value in the case of protected band crossings in topological semimetals.

**Giant anomalous Hall effect in Co$_2$MnGa single crystals**

Large anomalous Hall conductivity (AHC) in Co$_2$MnGa was predicted by Kübler et al. [1] via Berry curvature calculations. Here, the intrinsic AHC is calculated by integrating the Berry curvature for all the states up to the Fermi energy ($E_F$). Thus, for magnetic Weyl semimetals with broken time reversal symmetry, where nodal lines form very close to $E_F$, the AHC attains a colossal value [2].

Here, we have successfully synthesized Co$_2$MnGa single crystals with the Bridgman crystal growth technique for the first time (Fig. 1a). An image of a full grown crystal and a corresponding Laue pattern are shown in Figure 1a. The calculated band structure (Fig. 1b) shows the formation of three nodal lines very close to $E_F$ that are protected by mirror symmetry. Co$_2$MnGa is found to be ferromagnetic with $T_C$ ~ 686.4 K as seen from the field-cooled and zero-field-cooled magnetization in Figure 1c. The saturation magnetization is 4 µB f.u.$^{-1}$ at 2 K and slightly decreases to 3.8 µB f.u.$^{-1}$ at 300 K. The strong Berry curvature from the nodal lines induces a colossal AHC of ~1600 Ω$^{-1}$cm$^{-1}$ at 2 K (Fig. 1d), which is the highest known AHC value apart from Co$_2$MnAl in any magnetic Heuslers [3]. The AHC decreased marginally to 970 Ω$^{-1}$cm$^{-1}$ as the
temperature increased to 300 K (Fig. 1e), but the longitudinal conductivity dropped sharply [3, 4]. As a result, we observed a giant anomalous Hall angle (AHA) up to 12% at room temperature (Fig. 1f), which is maximum observed value in any magnetic Heusler reported so far in the literature [3].

Colossal anomalous Nernst effect beyond the magnetization scaling relation in Co$_2$MnGa single crystals

The anomalous Nernst effect (ANE) is analogous to the electrical anomalous Hall effect, where a temperature gradient is applied in a magnetic material and a Hall voltage is generated that is perpendicular to both the heat flow and the magnetization of the sample. For a long time it was believed that the value of ANE is proportional to the net magnetic moment of the sample. However, recent investigation shows that the ANE actually originates from a net Berry curvature of all bands near $E_F$. Conversely, a large anomalous Nernst thermopower ($\Delta \sigma_{xy}/\sigma_{xx}$) can be observed in topological materials with no net magnetization but large net Berry curvature $\Omega_{o}(k)$ near $E_F$. A typical example is non-collinear antiferromagnetic system, such as Mn$_3$Ge and Mn$_3$Sn. Clearly this falls outside the scope of the conventional magnetization-model of ANE, but a worthy question remains: Can the value of ANE in topological ferromagnets exceed the highest values observed in conventional ferromagnets? Recently, we observed a remarkably high value of $\sim 6.0$ $\mu$V K$^{-1}$ at 1 T magnetic field (Fig. 2a) in ferromagnetic topological Heusler Co$_2$MnGa single crystals at room temperature [4]. This is nearly a factor 7 larger than any anomalous Nernst thermopower value ever reported for any conventional ferromagnet (Fig. 2b). Combined electrical, thermoelectric, and first-principles calculations reveal that this high ANE value arises from a large net Berry curvature near the Fermi level associated with the nodal lines and Weyl points.

Discovery of the first three-dimensional topological ferromagnet

We discovered the first three-dimensional topological magnetic phase in our Co$_2$MnGa single crystals in collaboration with Prof. M. Zahid Hasan from Princeton [5]. A nodal line is formed when two bands of opposite mirror eigenvalues cross (Fig. 3a). We observe a pair of bulk bands crossing along the curves...
in the bulk Brillouin zone, which is the topological line nodes associated with π Berry phase and protected by mirror symmetry. All the line node features of the measured Fermi surface (Fig. 3b) matches very well with the ab initio constant energy surface (Fig. 3c).

Considering a series of $E_B - k_y$ ARPES cuttings at different $k_y$ values (Fig. 3d-f), we located band crossing over a range of $k_y$, where $E_B$ is the binding energy. This is the distinctive feature for observation of a line node and hence is different from the conventional Dirac or Weyl semimetals where band crossing occurs at one particular $k_y$ value. To further confirm the line nodes, we plot the constant energy surfaces as a function of $E_B$. As we move from $E_F$ to a deeper $E_B$ value, we see that the line node feature changes from a ‘<’ shape to a ‘>’ shape (Fig. 3g-i), forming electron and hole pockets accordingly. For energies which cross the line node, we find electron and hole pockets intersecting at a point (Fig. 3h). All of these results clearly demonstrate the first experimental proof for the formation of topological line nodes in Co$_2$MnGa single crystals [5].

**Fig.-2:** (a) Magnetic field-dependent Nernst thermopower ($S_{xy}$) in Co$_2$MnGa single crystals at temperatures ranging from 340 to 60 K. The inset shows the temperature variation of the extracted anomalous Nernst thermopower ($S_{xy}^A$) after extrapolation from the data gathered at high field (above 1 T). (b) A comparison plot of the magnetization-dependent anomalous Nernst thermopower ($S_{yx}^A$) in various ferromagnetic metals. Co$_2$MnGa single crystals show the highest value beyond any magnetization scaling relation, as highlighted by the blue shaded region.

**Fig.-3:** (a) Band degenerate line node (yellow curve) as a closed curve in the Brillouin zone. (b) The Fermi surface matches very well with (c) the ab initio constant energy surface at $E_B = 0.08$ eV below $E_F$, reflecting hole doping in the sample. (d-f) $E_B - k_y$ spectra cuttings at different $k_y$, and (g-i) $E_F$ at various $E_B$ values mapping the dispersive evolution of the line node in Co$_2$MnGa.

**Tuning the anomalous Hall conductivity from colossal to zero in topological magnetic Heuslers**

The Hall resistivity is commonly expressed as: $\rho_{xy} = R_0B + (\alpha\rho_{xx}^2 + \beta\rho_{xx})M + \rho_{topo}$, where $R_0$, $\alpha$, and $\beta$ are constants. The first term is the linear Hall coefficient due to the Lorentz force, and the last term is the contribution from the Berry curvature in real space (spin texture) due to skyrmion formation, etc. Evidently, the second term gives a finite Hall contribution and AHC $\neq 0$ for a ferromagnetic metal ($M, \rho \neq 0$). Interestingly, all the magnetic Heusler compounds we investigated here are metallic in nature. Since the discovery of AHE, it has been considered that AHE is a characteristic feature of finite magnetization in a compound. But is it a universal law? Our recent
investigations on transport measurement in topological Heuslers indicate otherwise.

In topological semimetals like Co$_2$MnGa, three nodal lines form near the $\Gamma$ point in the $k_x$, $k_y$, and $k_z$ planes and are protected by the mirror symmetries $M_x$, $M_y$, and $M_z$ in the $Fm\overline{3}m$ space group. Upon incorporating SOC, the crystal symmetry changes depending on the magnetization direction. For example, the $M_x$ and $M_y$ mirror symmetries are broken if a sample is magnetized along the [001] direction. Therefore, the nodal lines will open up unless certain symmetries remain to protect the band crossings away from $E_F$. As a consequence, at least two Weyl points form along the $k_z$ axis, leading to a finite AHC [2, 3]. Now, what happens if we break this mirror symmetry that protects the nodal line in the $k_z$ plane?

We reduce the crystal symmetry in such a way that the inversion and mirror symmetries are broken. An easy example is to form an inverse-Heusler structure with $F\overline{4}3m$ space group, such as occurs in Mn$_2$CoGa. Because of the non-centrosymmetric crystal structure, the mirror planes $M_x$, $M_y$, and $M_z$ of the full-Heusler no longer exist. Naturally, a gap appears between the nodal lines, and no Weyl points form upon incorporating SOC. Our DFT calculations show that the Berry curvature attains a positive and negative hot spots in $k$-space resulting zero AHC upon integration, which is independent of the sample’s magnetization.

We investigated ferromagnetic Mn$_2$CoGa single crystals ($T_C$ of 687 K with a saturation magnetization of $\sim 2 \mu_B$ and electrically metallic with finite DOS at $E_F$ (Fig. 4a). However, the AHC is zero (Fig. 4b), which breaks the classical law established so far in the literature. From the theoretical calculations, we understand that both spin channels in Mn$_2$CoGa contribute a finite and opposite Hall contributions due to the special crystalline symmetry and band structure (Fig. 4c) as well as both positive and negative hot spots in the Berry curvature (Fig. 4d). Thus, the overall integration results in a zero AHC for Mn$_2$CoGa [3]. We arrive at a situation where the compound is topologically semiconducting, but electrically metallic (Fig. 4g).

We realize that the classical law governing AHC breaks down in a series of compounds like the spin-gapless semiconductors [3]. Here, the AHC can be compensated, irrespective of finite magnetization in the sample. Our theoretical calculation also predict that the converse effect is also true, i.e., a large AHE can be observed in a compensated ferrimagnet as well. Here, one can break the glide symmetry that prevents any operation to reverse the sign of Berry curvature of the inverse-Heusler structure via atomic substitution,
hence resulting in a non-zero AHC. We predict Ti$_2$MnAl as a candidate compound, which is a compensated ferrimagnetic Weyl semimetal and might show a strong AHE ($\sim$500 S/cm) [6]. Therefore, through simple materials engineering via manipulation of the crystal symmetry and $N_\gamma$, we present a design scheme that allows the the details of band structure to be tuned so as to change the AHC from colossal to zero, independent from sample’s magnetization, which is suitable for next generation topo-spintronic applications (Fig. 4e,f).

**Outlook**

This year, we are about to receive a laser based-optical floating zone instrument and a high powder rod casting machine. This extends our flexibility and capacity for growing single crystals of various compounds beyond our halogen-based optical floating zone, Bridgman, or flux-growth techniques. In the future, we are interested to investigate various exotic compounds with multiple-topological order and in exploring how the real space topological effect like skyrmion formation can influence the $k$-space topological effects, such as the Dirac or Weyl points.

**External Cooperation Partners**

M. Z. Hasan (Princeton University, USA); Y. Chen (Physics Department, Oxford University, Oxford, UK); J. Heremans (The Ohio State University, USA)

**References**


# Kaustuv.Manna@cpfs.mpg.de