Quantum Origins of Anomalous Hall: Berry Phase & Atomic Ordering in Heusler Alloys

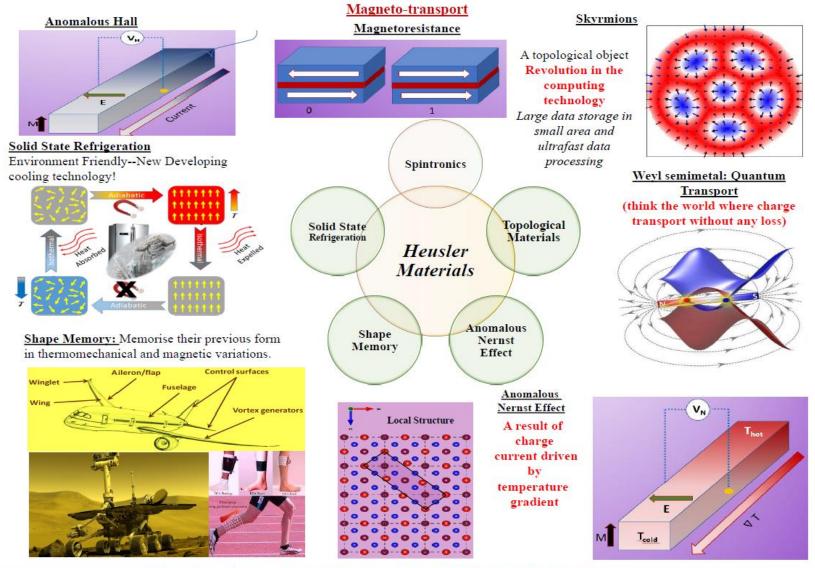


Dr. Sanjay Singh School of Materials Science and Technology Indian Institute of Technology (BHU), Varanasi ssingh.mst@iitbhu.ac.in

https://iitbhu.ac.in/dept/mst/people/sanjaymst

QIT, IIIT Allahabaad

In our lab we work on.....



On going Research work in Functional Materials Laboratory



Research @ Anomalous Hall



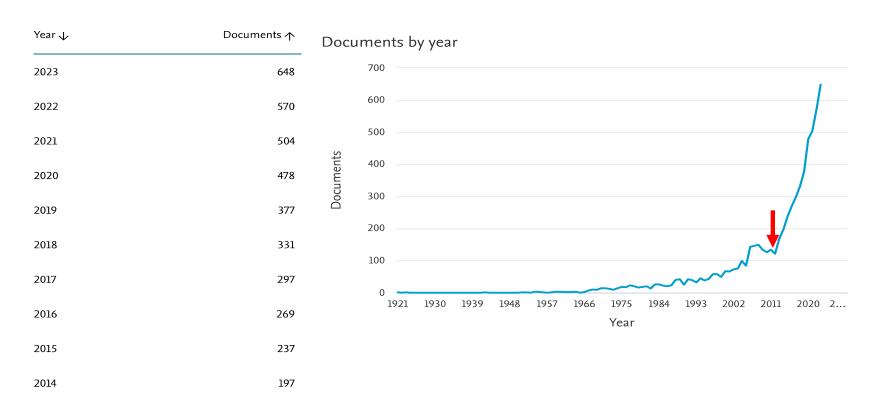
🔀 Email

→ Export 🖶 Print

Analyze search results

6,384 document results

Select year range to analyze:	1921	to	2023	~	Analyze]
Select year range to analyze:	1921	_ to	2023		Analyze	



Applications of anomalous Hall effect

- AHE depend on the electronic and magnetic structure of material has become a useful experimental tool for solid state physicist. For e.g. study of extremely thin magnetic layers and to observe the propagation of conduction electrons in a metal.
- ❖ The ability of the anomalous Hall effect to trace the magnetic impurities in metal can.
- ❖ We could study the spin-dependent properties e.g. spin-orbit coupling with the help of AHE.



Samsung Galaxy S4 s-view cover (Sensor)



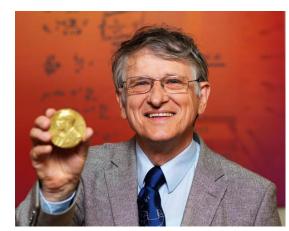
Non-volatile memory device

- * High sensitivity ($s = \frac{\partial R}{\partial H} = \frac{\mu R_{AH} \chi}{t}$, χ =magnetic susceptibility, t=thickness of the sample) far beyond the semiconductor typically 1000 ohm/T.
- Widely used in sensors and memory devices due to their linear field response, thermal stability, high-frequency operation (can operate at GHz frequency), sub-micron dimensions of samples etc.

Rev. Mod. Phys. 82, 1539 (2010), J. Appl. Phys. 122, 033901 (2017)

Nobel Prize in the area of Hall Effect

Nobel Prize in Physics (1985)
For the discovery of integer quantum Hall effect



Klaus von Klitzing

Nobel Prize in Physics (1998)

Discovery of new form of quantum fluid with fractionally charged excitations

(Associated with Fractional quantum Hall effect)



Photo from the Nobel Foundation archive. Robert B. Laughlin



Photo from the Nobel Foundation archive. Horst L. Störmer



Photo from the Nobel Foundation archive.

Daniel C. Tsui

> Outline

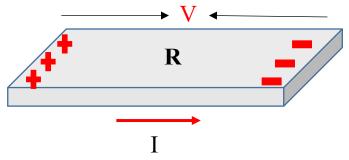
> Basics: Anomalous Hall effect

- > Results:
- ➤ Atomic disorder and Anomalous Hall in Heulser Compounds
- > Tunning with chemical disorder
- > Conclusion

Electrical transport

When an electric current flows through a conductor a longitudinal voltage develops across the conductor.

R = Resistance of the conductor



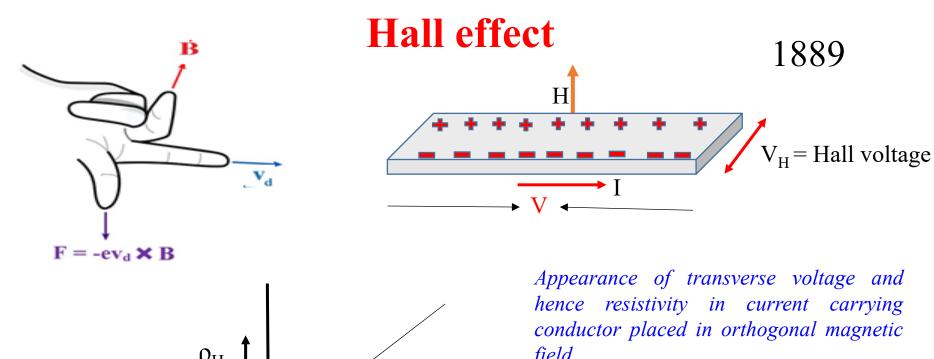
Follow ohms law

V = I*R



George Ohm

What will happen if we place the current carrying conductor in external magnetic field



 $\rho_{\boldsymbol{H}}$ Hall coefficient

$$R_0 = 1/ne$$

field.



Edwin Hall

'n' is the carrier concentration (Revolutionized the semiconductor physics & industry)

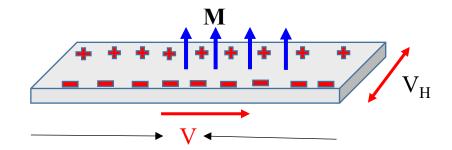
Anomalous Hall effect (AHE)

A typical curve of

Hall resistivity in

ferromagnets

In ferromagnets the Hall resistivity enhances many times in comparison to the normal metal

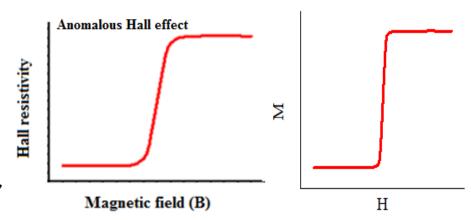


$$\rho_{\rm H} = R_0 H + R_s M$$

Anomalous Hall resistivity

$$\rho_{\rm H} = \rho_O + \rho_{AH}$$

AHE somehow looks related to the magnetization, fairly indicates that the AHE is spin dependent phenomenon.



Beyond the Lorentz force

A typical M-H curve For ferromagnets

Origin of the anomalous Hall effect

Controversial over a long decades

Several experimental studies have done to understand the origin of anomalous Hall

Theories of J. Smit, R. karplus and J.M. luttinger successfully explain the origin of AHE

$$\rho_{\rm H} = \rho_O + \rho_{A\rm H}$$

$$\rho_{AH} = a \rho_{xx} + b \rho_{xx}^2$$

Skew Scattering (extrinsic)

Intrinsic mechanism + Side jump (extrinsic)

R. Karplus et.al Phys. Rev. 95, 1154 (1954)

J.M. luttinger Phys. Rev. 112, 739 (1958)

J. Smit Phys. Rev. 92, 1576 (1953)

Rev. Mod. Phys. 82, 1539 (2010)

Intrinsic (Quantum) Origin of anomalous Hall effect

Intrinsic mechanism: Purely arises from the band structure of the material

(AHC)
$$\sigma_{\alpha\beta} = -\frac{e^2}{\hbar} \sum_n \int \frac{d^3k}{(2\pi)^3} \Omega^n_{\alpha\beta}(k) f_n(k),$$

$$Berry\ curvature \qquad \Omega^n(R) = \nabla_R \times A_n(R).$$

Phase picked by the wave function in an infinite small loop around the (k_x, K_y) point of an energy band in reciprocal space

Equivalent to the magnetic field in k space and result into the anomalous Hall conductivity (AHC).

Rev. Mod. Phys. 82, 1959 (2010)

Berry Phase: A Quantum Mechanical Phenomenon

Classical Boltzmann Transport

Classical transport theory describes how electrons move in a solid under external forces (like electric or magnetic fields) without quantum effects like spin, Berry phase, or wave interference.

Group velocity of electron:

$$rac{d\langle {f r}
angle}{dt} = rac{1}{\hbar}rac{\partial E}{\partial {f k}} \; .$$

Force on electron in the presence of both E and F:

$$\hbar rac{d\mathbf{k}}{dt} = -e\left(\mathbf{E} + \mathbf{v} imes \mathbf{B}
ight)$$



explains ordinary Hall effect

as Lorentz force:

$$\mathbf{F} = -e\left(\mathbf{E} + \mathbf{v} \times \mathbf{B}\right)$$

In Steady state, electric force balances the magnetic force:

$$-e\mathbf{E}_{\mathrm{Hall}} = -e(\mathbf{v} \times \mathbf{B})$$

$$\Rightarrow \mathbf{E}_{Hall} = \mathbf{v} \times \mathbf{B}$$

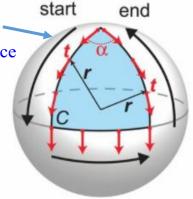
Fails to explain anomalous Hall effect due to quantum geometric phase (Berry phase).

If a quantum mechanical system is perturbed by a weak perturbation, then after a time t' (say), the system does not find a new quantum state but return to its original state with a phase factor.

classical example is the parallel transport of a vector around a path on a curved surface

$$\gamma_n(t) = \int d{m R} \cdot {m A}_n({m R}),$$
Berry Phase Berry vector potential

Rev. Mod. Phys. 82, 1959 (2010)



Science 356, 845-849 (2017)

Berry curvature

Quantum corrections: Including Quantum Berry phase

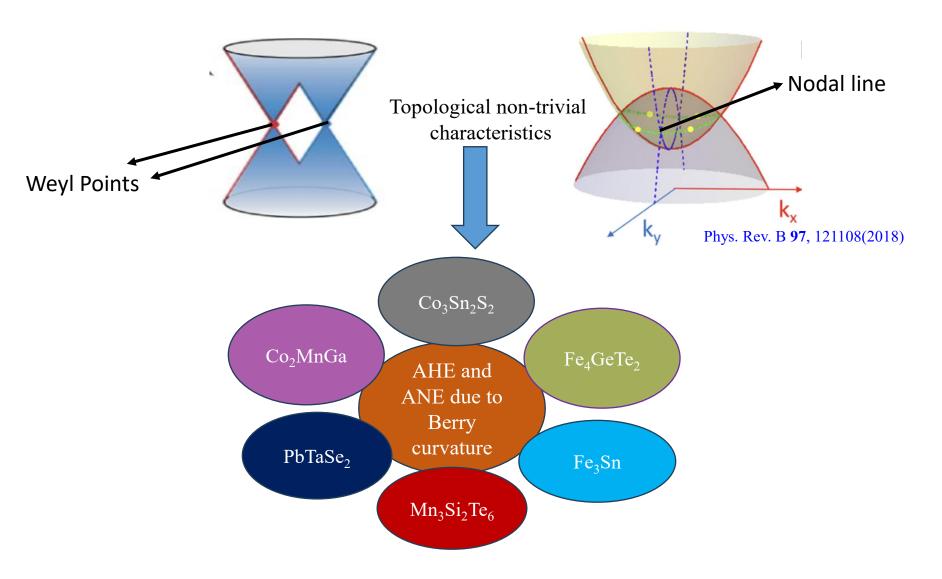
Modified group velocity:

$$rac{d\langle \mathbf{r}
angle}{dt} = rac{1}{\hbar} rac{\partial E}{\partial \mathbf{k}} + (\dot{\mathbf{k}} imes \mathbf{\Omega}_n(\mathbf{k}))$$

The curvature of Berry phase is equivalent to the magnetic field in k space and result into the additional contribution to the anomalous velocity of electrons. 12

- Classical transport also uses momentum space but considers only energy not wavefunctions.
- Berry phase comes from wavefunction geometry- not energy but how the state itself evolves as k changes.

Sources of Berry curvature



Therefore, tuning of Fermi level close to these topological non-trivial characteristics can significantly enhance AHE and ANE

Materials Selection



Research @ Heusler alloys

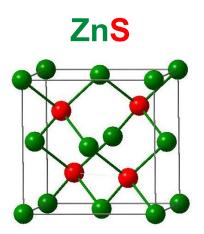


Analyze search results

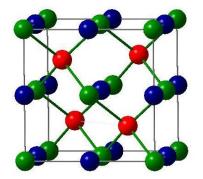


Year ↓	Documents ↑	Documents by year
2023	723	800
2022	686	600
2021	686	J
2020	648	Documents and the second of th
2019	660	å
2018	639	200
2017	524	
2016	495	1900 1911 1922 1933 1944 1955 1966 1977 1988 1999 2010 2021 2 Year

Heusler alloys







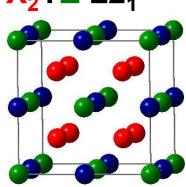
XYZ Heusler compounds

La Ce Pr Nd Pm Sm Eu Gd Tb Dy Ho Er Tm Yb Lu
1.10 1.12 1.13 1.14 1.13 1.17 1.20 1.20 1.10 1.22 1.23 1.24 1.25 1.10 1.27

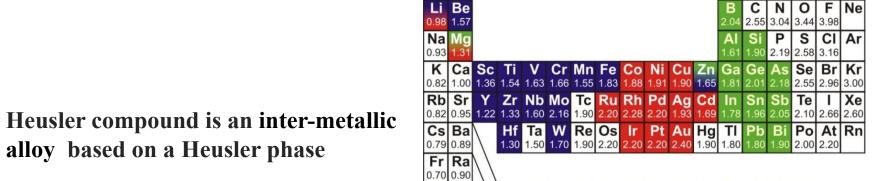
 Ac
 Th
 Pa
 U
 Np
 Pu
 Am
 Cm
 Bk
 Cf
 Es
 Fm
 Md
 No

 1.10
 1.30
 1.50
 1.70
 1.30
 1.28
 1.13
 1.28
 1.30
 1.30
 1.30
 1.30
 1.30
 1.30
 1.30





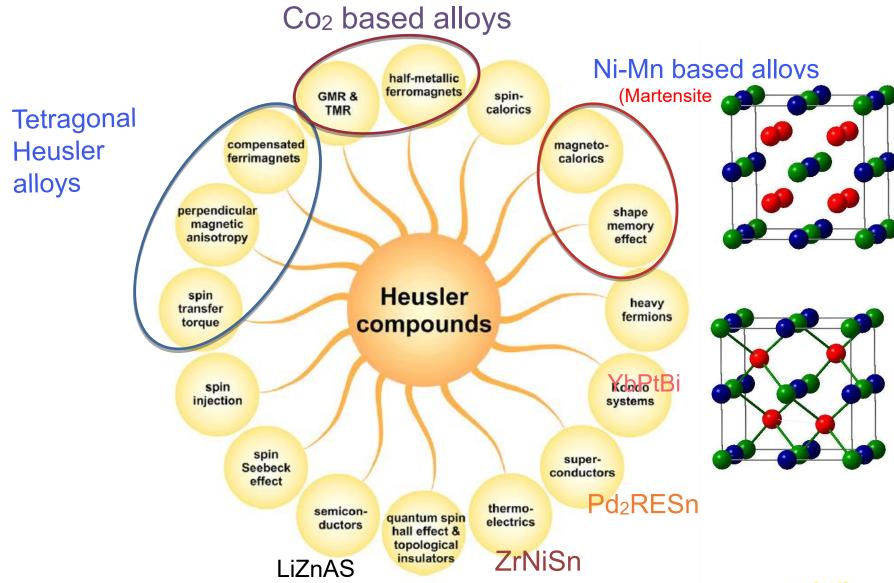
He



2.20

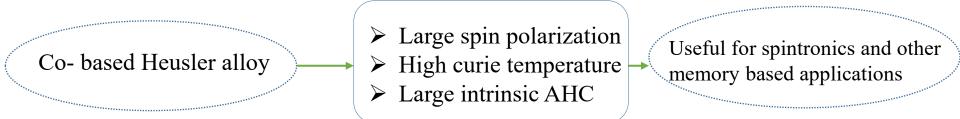
Friedrich Heusler: Cu₂MnAl (1903)

Why Heusler alloys ??





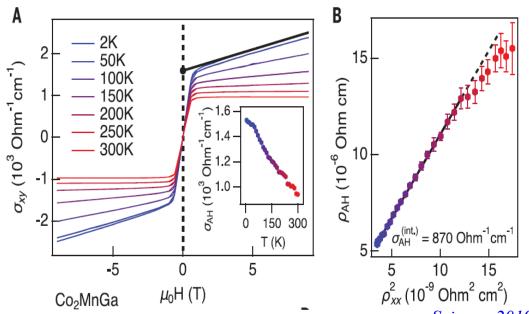
Co-based Heusler alloys



Time-Reversal-Breaking Weyl Fermions in Magnetic Heusler Alloys

Physical Rev. Letters 117, 236401 (2016).

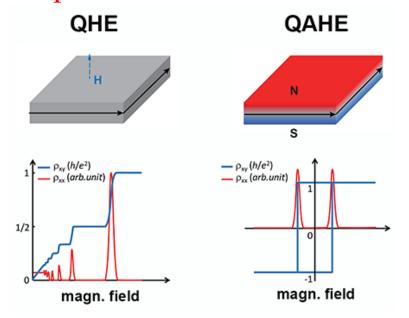
- Anomalous Hall effect and the role of Berry curvature..... Phys. Rev. B 100, 054445 (2019)
- ➤ Discovery of topological Weyl fermion lines and.... *Science-2019 DOI:* 10.1126/science.aav2327



Science-2019 DOI: 10.1126/science.aav2327

Fundamental Physics and Materials Engineering

- Theory suggest possibility of the quantized AHC, that is, a 3D quantum AHE (QAHE).
- ➤ How can be reached to the 3D QAHE or giant AHE?
- ➤ What is the recipe to find such materials?



Answers to these questions are not only of fundamental importance, but also likely leads to technological applications.

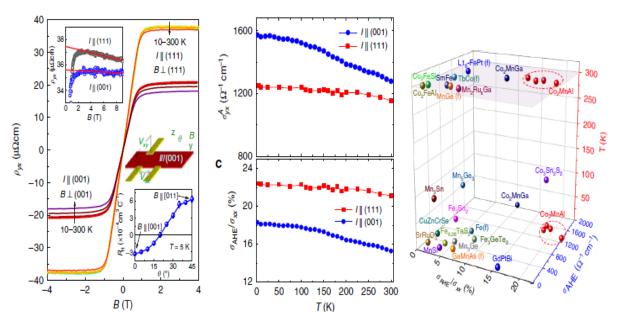
Co based Heusler: A promising class for anomalous transport



ARTICLE https://doi.org/10.1038/s41467-020-17174-9 Check for updates

Giant room temperature anomalous Hall effect and tunable topology in a ferromagnetic topological semimetal Co₂MnAl

Peigang Li^{1,6}, Jahyun Koo^{2,6}, Wei Ning^{3™}, Jinguo Li⁴, Leixin Miao⁵, Lujin Min^{3,5}, Yanglin Zhu^{1,3}, Yu Wang^{1,3}, Nasim Alem⁵, Chao-Xing Liu ⁰, Zhiqiang Mao ⁰, Shiqiang Mao ^{1,3™} & Binghai Yan ⁰, ^{2™}



All previous studies disorder B2 phase.

This study ordered L2₁ phase

Very large AHC, up to $1300 \ \Omega^{-1} \text{cm}^{-1}$ at room temperature.

Atomic ordering effect

Nat Commun 11, 3476 (2020)

Atomic ordering & AHC in Co₂-based Heusler alloys

 Co_2YZ : All atoms at their own positions: $L2_1$ ordered

Mixing of Y and Z atoms: **B2 disorder**

Mixing of Co and Z atoms: DO₃ disorder

Mixing of all atoms: A2 disorder

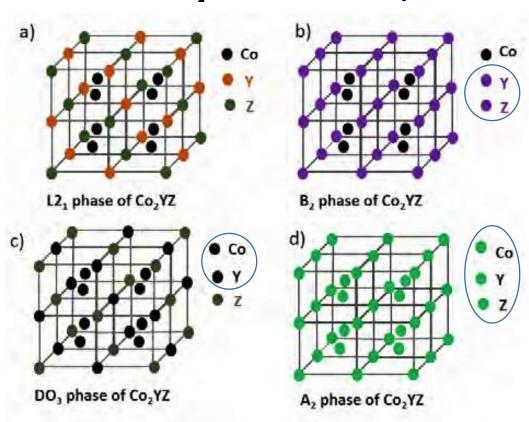
Disorder: A common phenomenon in the Co₂-based Heusler alloys

Exp.: Co₂FeSi, Co₂FeAl, Co₂MnAl (thin film) etc. show the different types of disorder

•	Co ₂ MnGa	1300 S/cm
	00/11/11/04	15000

- Co₂FeGe 78 S/cm
- Co_2MnAl 1600 S/cm
- Co_2TiSn 100 S/cm
- Co₂FeAl 39 S/cm
- Co₂FeSi 189 S/cm
- Co₂FeGa 181 S/cm
- Co_2MnSi 100 S/cm
- Co_2MnSn 82 S/cm
- CO_2 IVIIISII 62 S/CIII
- Co_2CrAl 438 S/cm
- Co_2VSn 1400 S/cm
- Co₂VGa 66 S/cm

Value of AHC in different Co₂-based Heusler alloys

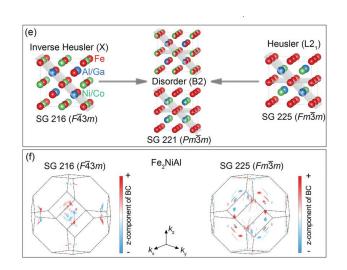


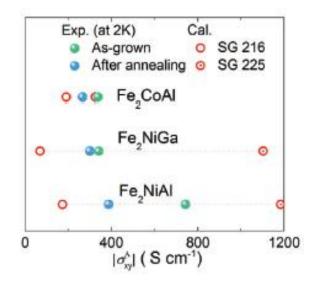
PRB 91, 134409 (2015); Phys. Rev. B **85**, 012405; Phys. Rev. B **83**, 174410 (201<u>1)</u>; Phys. Rev. B **101**, 134407 (2020); J. Magn. Mater. **448**, 371 (2018)

Effect of disorder on AHC

• Recently Fe₂-based Heusler alloys are reported to have lower AHC values with B2 type disorder in comparison to ordered structure.

Adv. Sci. 8, 2100782 (2021)





Is it always true ???



Co₂FeAl, an ideal candidate to investigate (B2 is stable structure)

Anomalous Hall effect in Co₂FeAl Heusler compound: Literature

➤ The stable structure of Co₂FeAl is B2 type.

```
J. Magn. Magn. Mater. 442, 288 (2017)
```

- ➤ The AHC in Co₂FeAl thin films shows controversial results in terms of origin and the magnitude of AHC.

 [J. Magn. Magn. Mater. 442, 288 (2017), J. Magn. Magn. Mater. 362, 52 (2014)]
- \triangleright The Berry curvature calculation gives AHC ~ 39 S/cm.

```
PRB 91, 134409 (2015)
```

Experimental AHC ~ 100 S/cm

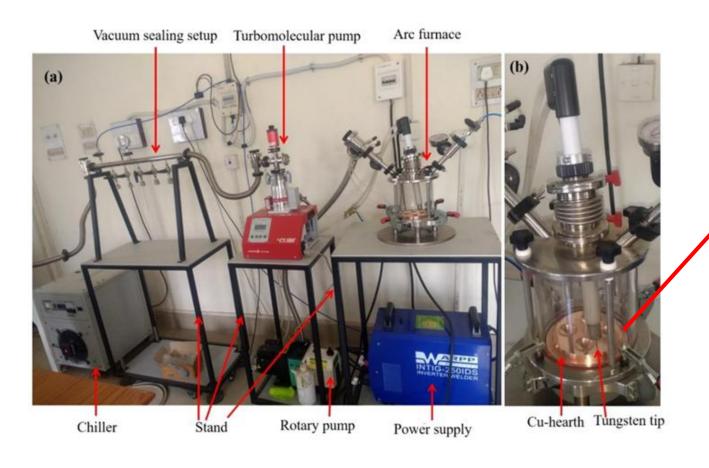
Theoretical AHC (ordered) ~39 S/cm

J. Magn. Magn. Mater. 442, 288 (2017),J. Magn. Magn. Mater. 362, 52 (2014)PRB 91, 134409 (2015)

Origin of large Anomalous Hall???

Synthesizing the material

Arc Melting Technique







Synthesis of intermetallic systems under the argon atmosphere

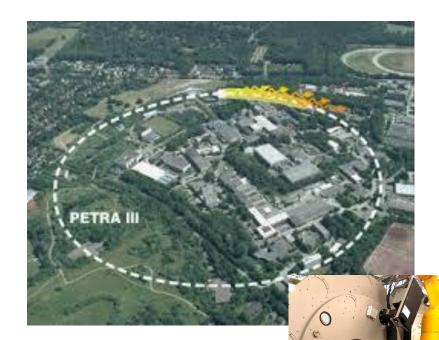
Sealed quartz tube

Co₂FeAl Heusler compound: Sample and Structure

Gaurav Shukla

- Sample preparation: Arc Melting (High Vacuum)
- •Structure: Synchrotron x-ray diffraction (DESY: GERMANY)



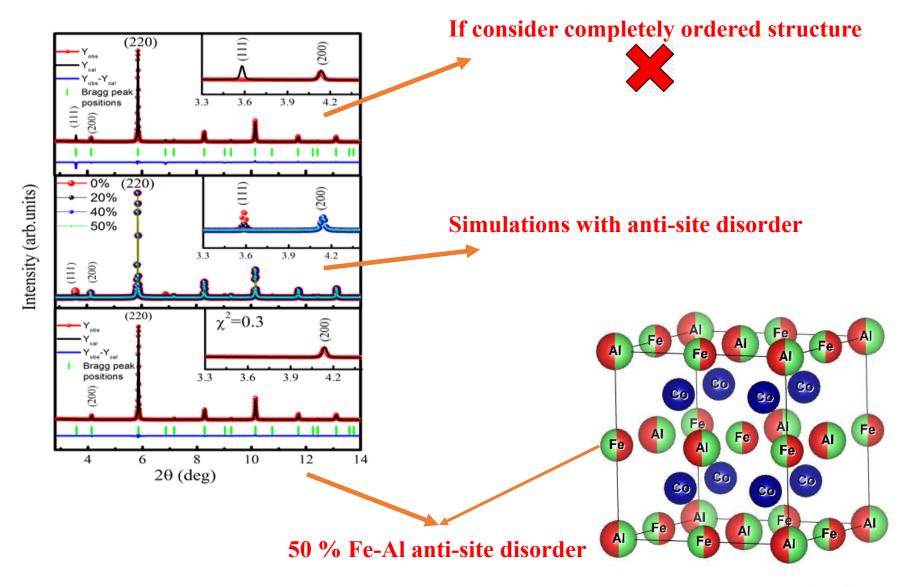




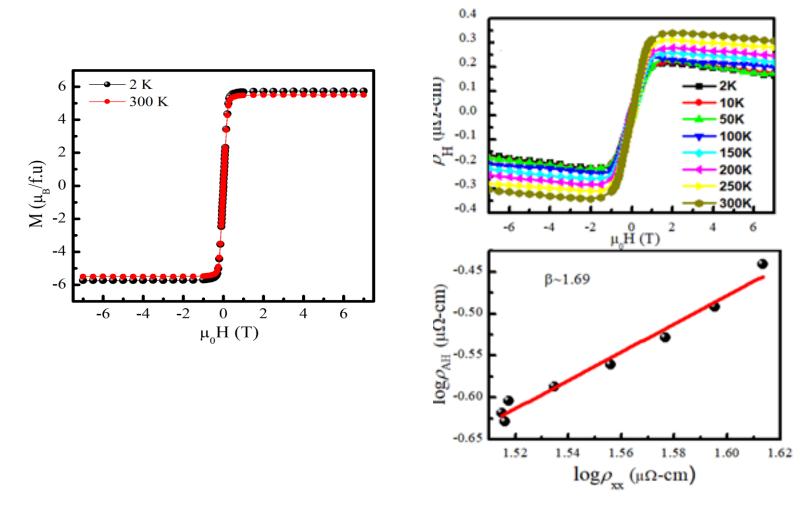
Capillary: Rotating: uniform distribution of intensities

High flux: Low intensity peaks

Co₂FeAl Heusler compound: Crystal Structure



Magnetization and Hall results



Linear Fitting is employed using relation $\rho_{AH} \propto \rho_{xx}^{\beta}$. β was (taking log both side in $\rho_{AH} \propto \rho_{xx}^{\beta}$) found 1.69 which indicates intrinsic contribution. [$\beta = 1$ for skew scattering and $\beta = 2$ for the intrinsic origin]

Hall analysis

$\rho_{AH} = a^{skew} \rho_{xx} + \sigma^{int} \rho_{xx}^2$

Intrinsic origin

skew scattering Intrinsic AHC parameter

$$a^{skew} = 0.002$$

$$\sigma^{int} = 155 \text{ S/cm}$$

*** We ignore the saturation magnetization in fitting because the change of M with T is very small.

If M changes significantly with T:

$$ho_{AH} = (a^{skew}
ho_{xx} + \sigma^{int} \rho_{xx}^2) M_s$$
 or
 $ho_{AH} M_s
ho_{xx} = a^{skew} + \sigma^{int} \rho_{xx}$ (Linear equation)

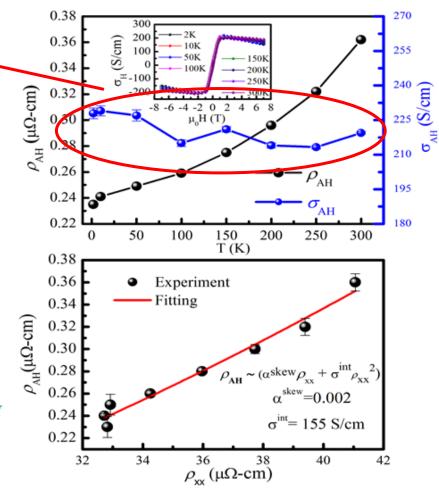
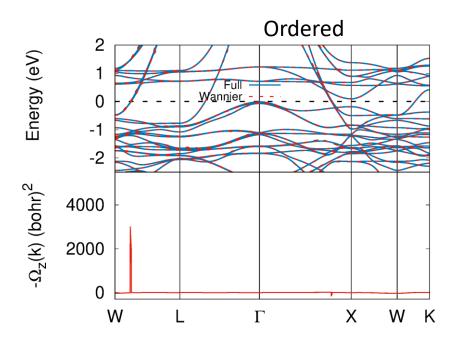
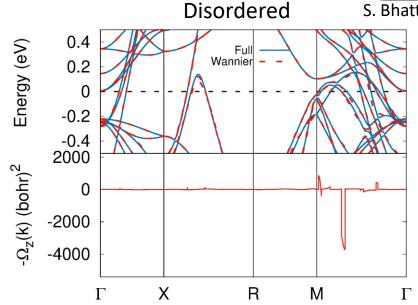


Fig. Temperature variation of AHC and the scaling analysis

First principles calculation







The calculated AHC value = 42 S/cm

Lower than the Experimental intrinsic AHC = 155 S/cm

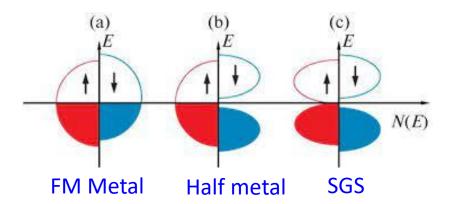
The AHC value for the 50 % Fe/Al disorder came out = 63 S/cm

Conclusion: disorder in the system can enhance the Berry curvature-induced intrinsic AHC. It depends upon the disorder-induced change in the electronic band structure.

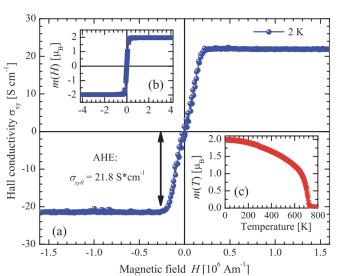
Is increasing AHC in Co₂FeAl due to antisite disorder just an incident ???

Spin-gapless semiconducting Heusler compound: Mn₂CoAl

- ➤ Spingapless semiconductors exhibit a band gap for one spin channel and a zero band gap for another spin channel.
- The first experimental evidence of spingapless semiconductor behavior was observed in the Mn₂CoAl compound.



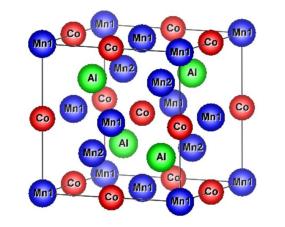
Unresolved issue ???



The experimental anomalous Hall conductivity

 $(AHC) \sim 22 \text{ S/cm}$

Theory ~ 3 S/cm.



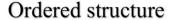
Inverse Heusler (F-43m)

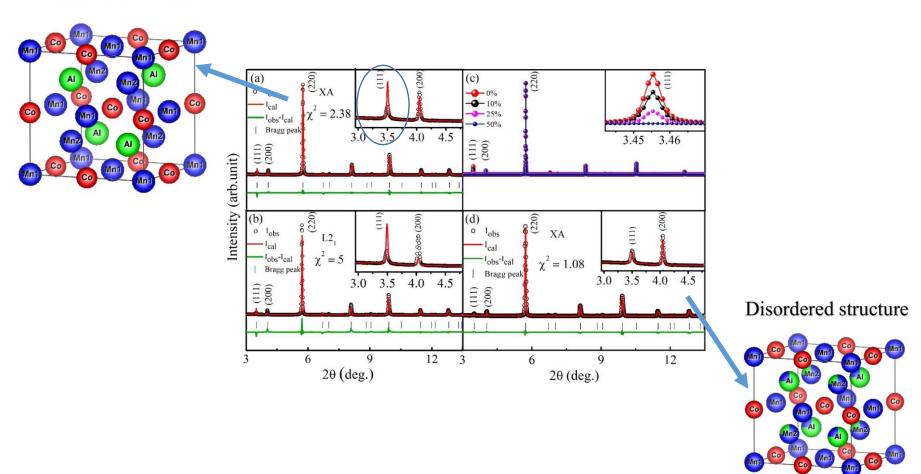
31

Crystal structure (our result): Synchrotron XRD



Nisha





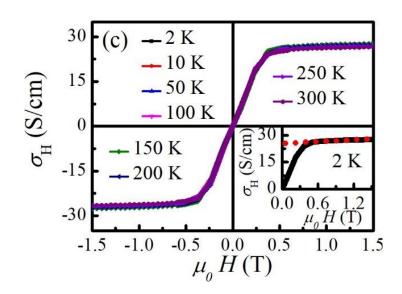
XRD analysis: 25% anti-site disorder between Mn and Al atoms.

Anomalous Hall

Experiment:

$$\sigma_{\mathrm{H}} = rac{
ho_{\mathrm{H}}}{(
ho_{\mathrm{H}}^2 +
ho_{xx}^2)}$$

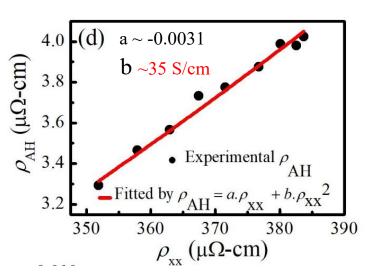
➤ Total Hall conductivity ~27 S/cm



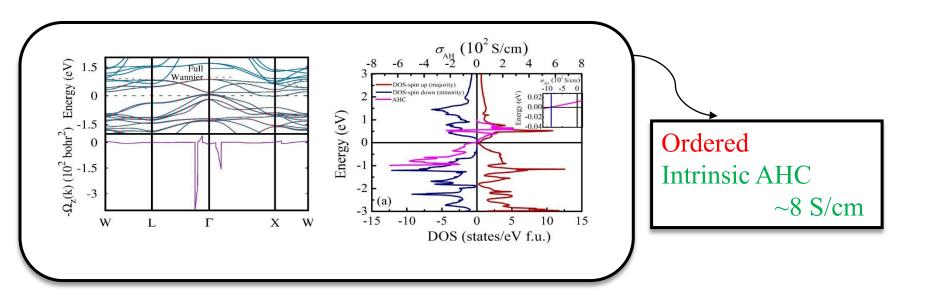
> By using the scaling relation-

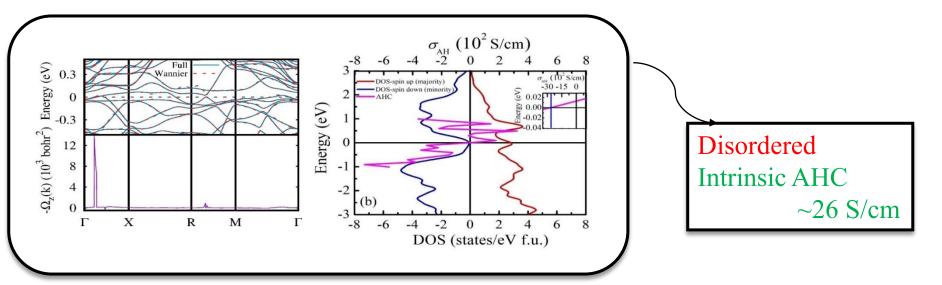
$$\rho_{\rm AH} = a\rho_{\rm xx} + b\rho_{\rm xx}^2$$

Intrinsic ~35 S/cm



Anomalous Hall effect: Theoretical study





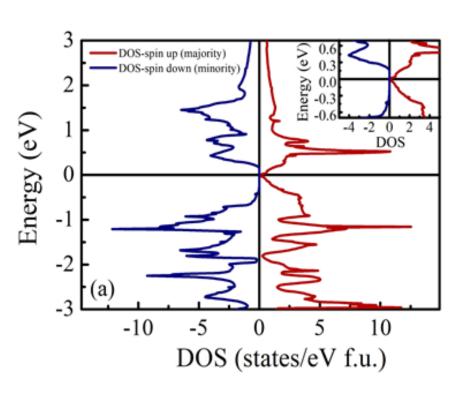
What does disorder actually do here??

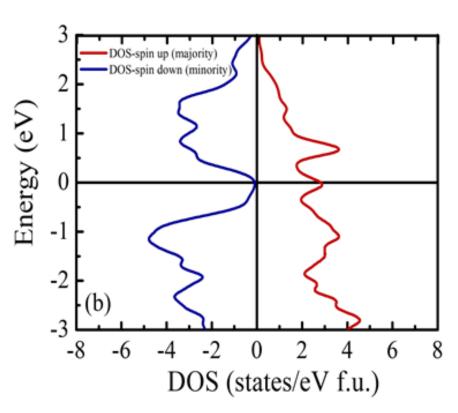


 \sim 7 S/cm

Disordered

~30 S/cm





Disordered enhances half metallic behavior and AHC

Controlling anti-site disorder is not

easy....What is an alternative way to enhance AHC? Chemical disorder is in our hands....



Payal

PHYSICAL REVIEW MATERIALS 5, 124201 (2021)

Role of chemical disorder in tuning the Weyl points in vanadium doped Co₂TiSn

Payal Chaudhary , Krishna Kant Dubey, Gaurav K. Shukla, and Sanjay Singh **
School of Materials Science and Technology, Indian Institute of Technology (Banaras Hindu University), Varanasi 221005, India

Surasree Sadhukhan and Sudipta Kanungo School of Physical Sciences, Indian Institute of Technology Goa, Goa 403401, India

Ajit K. Jena, S.-C. Lee, and S. Bhattacharjee *Indo-Korea Science and Technology Center (IKST), Bangalore 560065, India*

Jan Minár and Sunil Wilfred D'Souza New Technologies Research Centre, University of West Bohemia, Univerzitní 8, CZ-306 14 Pilsen, Czech Republic

(Received 23 December 2020; revised 12 October 2021; accepted 8 November 2021; published 3 December 2021)

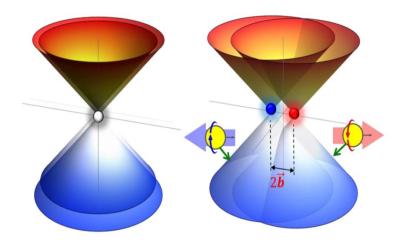
Weyl point and Anomalous Hall conductivity (AHC)

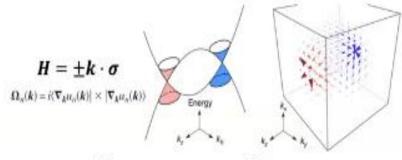
Broken time-reversal and/or Inversion symmetry

- ➤ A Weyl semimetal is a solid-state crystal whose low energy excitations are Weyl fermions
- ➤ Weyl points of opposite chirality act either as source or sink of Berry curvature.
- > Berry curvature: Equivalent to fictitious magnetic field in momentum space

Dirac semimetal

Weyl semimetal





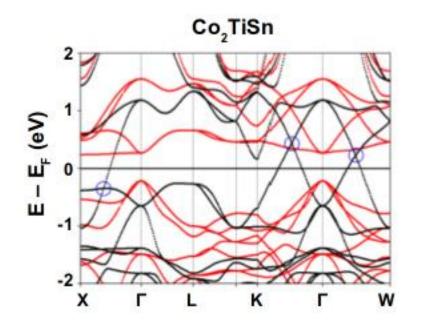
$$\sigma_{xy} = -\frac{e^2}{h} \int f_0 \Omega(k) dk = -\frac{e^2}{h} \frac{k_0}{2\pi}$$

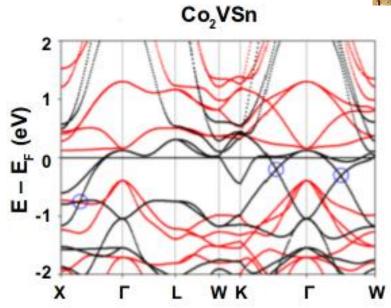
Weyl points — Large Berry curvature — Large AHC

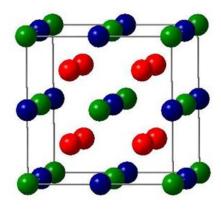
The problem



Payal







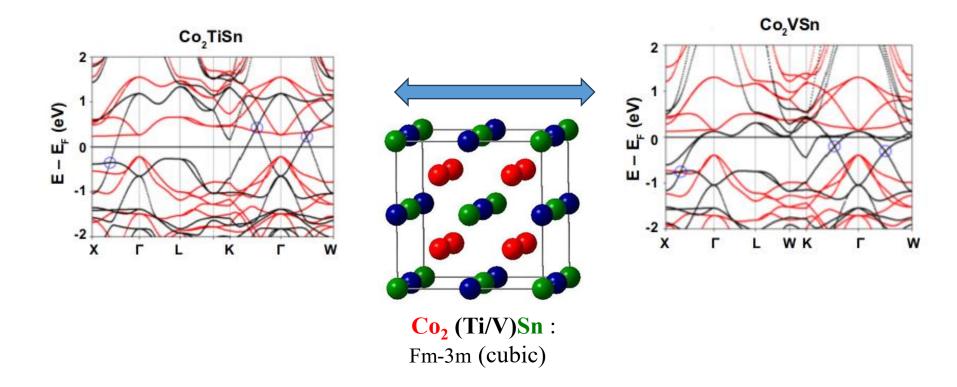
Co₂ (Ti/V)Sn: Fm-3m (cubic)

The solution

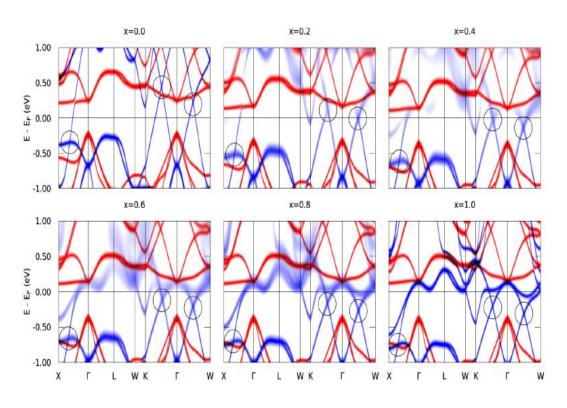


Payal

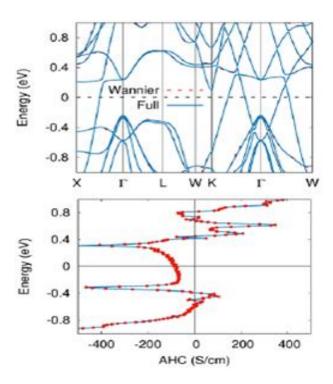
Valence electron Substituting V (5) in place of Ti (4 VE)??



Material engineering: Tuning of AHE by Chemical disorder



Band structure of $Co_2Ti_{(1-x)}V_xSn$.



Band structure (electronic + Wannier) plots and energy dependence AHC in Co_2TiSn .

AHC for x = 0.5 is 196.84 S/cm two times of x = 0 (~ 99 S/cm)

Controlling AHC..... Chemical disorder

PHYSICAL REVIEW MATERIALS 8, 034203 (2024)

Theoretical Investigation



Ujjawal

Tuning of nodal line states via chemical alloying in Co_2CrX (X = Ga, Ge) Heusler compounds for a large anomalous Hall effect

Ujjawal Modanwal, Gaurav K. Shukla, Ajit K. Jena, Satadeep Bhattacharjee, Sunil Wilfred D'Souza, Jan Minár, and Sanjay Singh 1,*

¹School of Materials Science and Technology, Indian Institute of Technology (Banaras Hindu University), Varanasi 221005, India
²Indo-Korea Science and Technology Center (IKST), Bangalore 560065, India

³New Technologies Research Centre, University of West Bohemia, Univerzitní 8, CZ-306 14 Pilsen, Czech Republic

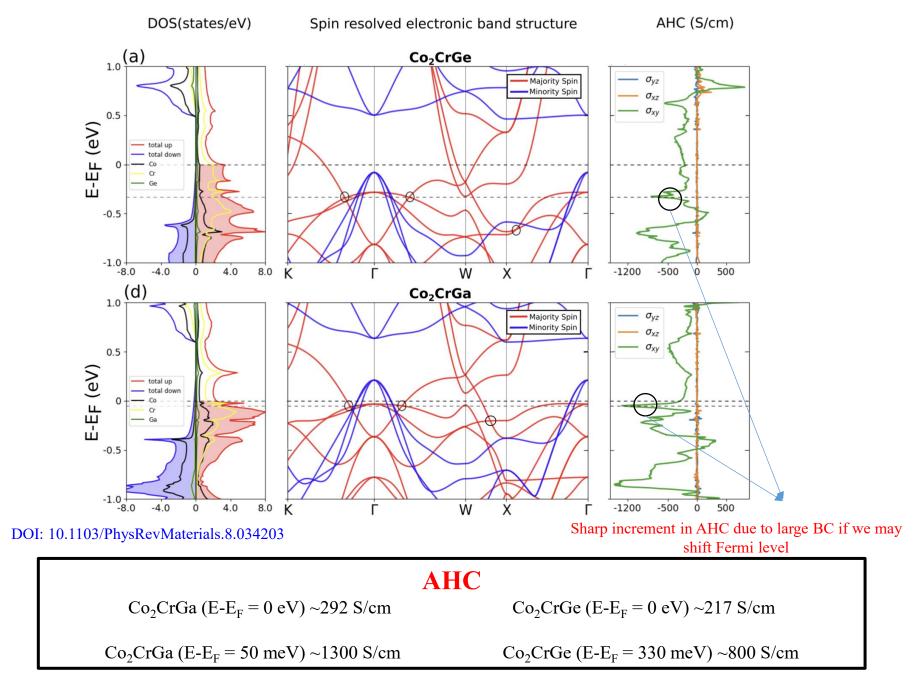


(Received 27 June 2023; revised 23 January 2024; accepted 28 February 2024; published 22 March 2024)

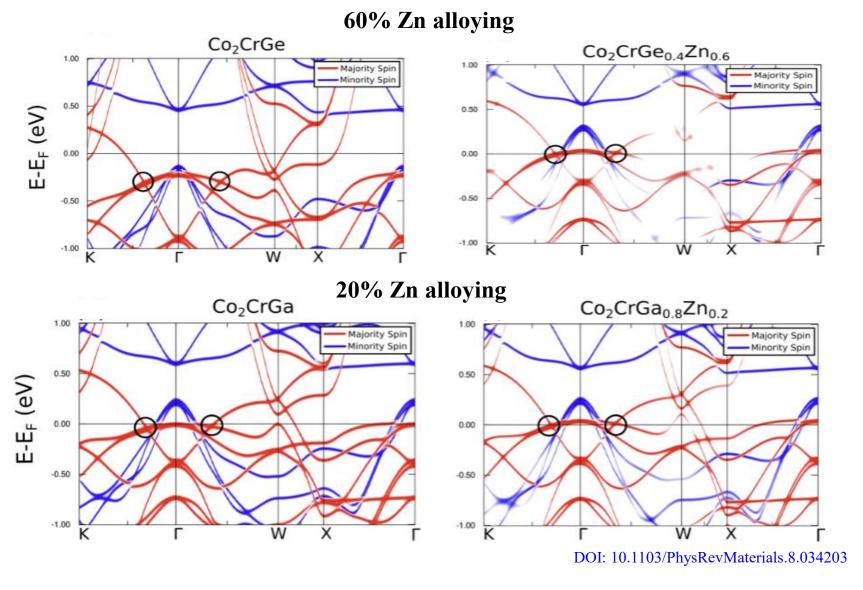
Topological materials have attracted significant interest in condensed matter physics for their unique topological properties leading to potential technological applications. Topological nodal line semimetals, a subclass of topological materials, exhibit symmetry-protected nodal lines, where band crossings occur along closed curves in the three-dimensional Brillouin zone. When the nodal lines are gapped out due to perturbation in the Hamiltonian, a large Berry curvature (BC) arises in the surrounding area of the gapped nodal line, leading to exotic anomalous transport responses. In this paper, we studied the Co₂CrX (X=Ga, Ge) Heusler compounds that exhibit mirror symmetry-protected nodal line states below the Fermi level. The BC calculation yields anomalous Hall conductivity (AHC) of about 292 and 217 S/cm for Co₂CrX (X=Ga, Ge), respectively, at the Fermi level, which increases by up to 400% at the nodal line energy level. We theoretically analyzed that 20% and 60% zinc (Zn) alloying in Co₂CrX (X=Ga, Ge) effectively lowers the Fermi level by 50 meV and 330 meV, respectively, aligning with the protected crossings. Consequently, we identified Co₂CrGe_{0.4}Zn_{0.6} and Co₂CrGa_{0.8}Zn_{0.2} as compositions to achieve the significant AHC of 800 and 1300 S/cm, respectively. The explicit AHC calculation for these alloyed compositions is in good agreement with our predictions. Our findings highlight that chemical alloying is an efficient way to enhance AHC in nodal line hosting materials.

DOI: 10.1103/PhysRevMaterials.8.034203

Very large: 1300 S/cm



> Thus, AHE can be increased by coinciding these band crossings with Fermi level



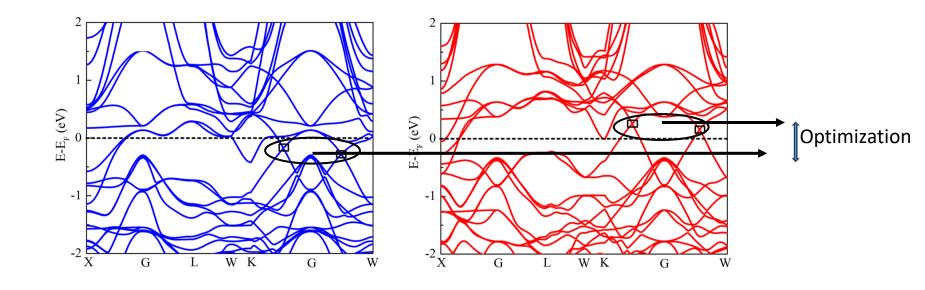
AHC calculated for close compositions $\text{Co}_2\text{CrGa}_{0.75}\text{Zn}_{0.25}$ and $\text{Co}_2\text{CrGe}_{0.5}\text{Zn}_{0.5}$ aligned with the previously predicated value at nodal line positions

Experimental proof?????

Controlling AHC..... Chemical disorder Experimental Investigation



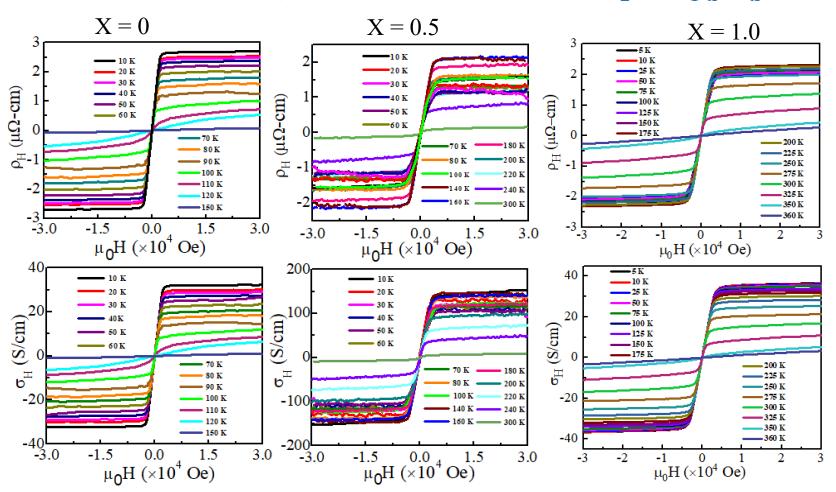
Co₂VSn_{1-x}Al_x Weyl semimetal



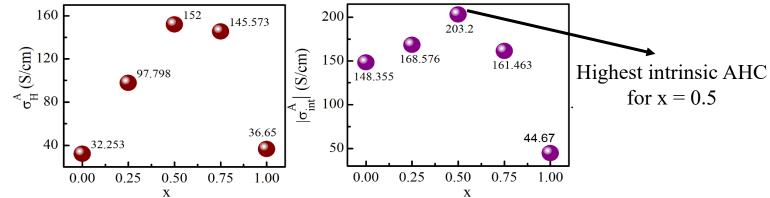
Valence electron: Substituting Al (3 VE) in place of Sn (4 VE) VE)??

Controlling AHC..... Chemical disorder Experimental Investigation

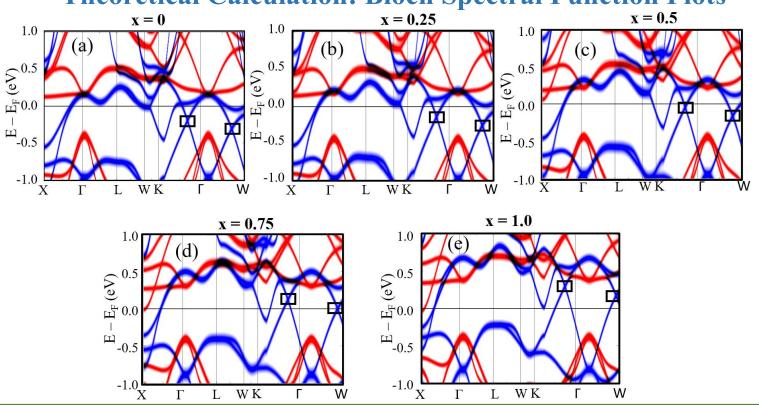
Hall Resistivity and Conductivity of Co₂VSn_{1-x}Al_x



Summary of Experimental AHC



Theoretical Calculation: Bloch Spectral Function Plots



Shifting of nodal line upwards with Al doping. Nodal line nearly coincides with E_F for x = 0.5

Conclusions

- Resolved the controversy about Anomalous Hall effect in Co₂FeAl and Mn₂CoAl Heusler compound and established it quantum origin
- ➤ Our work provides a basic understanding that, how the atomic ordering influence the Berry curvature and give a path to create large AHC due to modification of the Berry curvature induced by atomic/chemical ordering
- Large (quantum) Anomalous Hall effect may put forward for zero loss and clean energy devices.

Thank you!



VISIT OUR LAB @





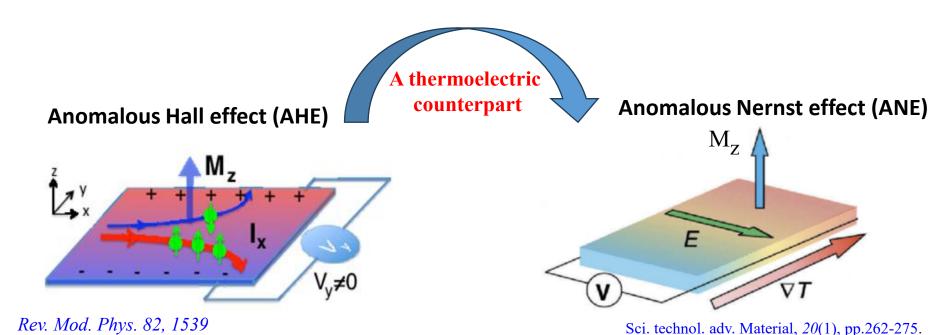


Science and Engineering Research Board (SERB)
Department of Science and Technology (DST)
Govt. of India





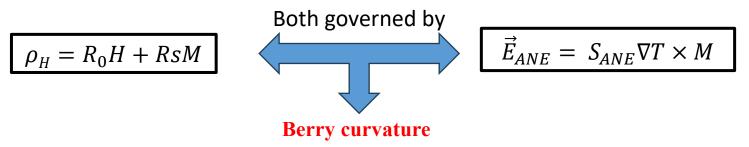
Anomalous transverse transport phenomena for clean energy



Generation of transverse voltage perpendicular to current and applied magnetic field

Generation of transverse voltage perpendicular to Temp. gradient and magnetic field

Recycling waste heat into electricity



AHE is governed by sum of Berry curvature over all occupied states and ANE is determined by the Berry curvature at Fermi level.