## Tunable magnetic properties in Heusler materials

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### Plan of talk



- > A short introduction to Heusler materials
- Compensated ferrimagnets and giant exchange bias



Anomalous Hall effect in non-collinear antiferromagnets



> Non collinear spin structure and skyrmions in Heuslers



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### Structure:X<sub>2</sub>YZ



#### Designing magnetic properties from flexible structure:



*Fm*-3*m*, centrosymmetric



F-43m, noncentrosymmetric



Ζ

Х



*I4/mmm*, centrosymmetric

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*I*-4*m*2, noncentrosymmetric

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### Why Heusler materials:





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### Tetragonal Heuslers for spintronics:



### Mn<sub>3</sub>Ga: Mn<sub>2</sub>MnGa: Tetragonal material with large magnetic anisotropy



Low magnetic moment and large perpendicular magnetic anisotropy.

Ideal for spintronic application



B. Balke, C. Felser et al., APL 90, 152504 (2007).

Tetragonal structure and ferrimagnetic ordering





Jeong and Parkin et al., Nature Comm. 7, 10276 (2016).

- $\checkmark$  Extremely large perpendicular magnetic anisotropy in Mn<sub>3</sub>Ge thin films.
- ✓ TMR up to 80 % has been observed in Mn3Ge based TMR device.



# Compensated ferrimagnets and giant exchange bias



### Designing compenstad magnetic state in Mn-Pt-Ga











FM clusters in AFM background

Nayak et al., Nature Mater. 14, 679 (2015).

By combining two ferrimagnetic compounds with opposite spin alignment we can design a compensated magnetic state.

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### Compenstad magnetic state in Mn<sub>3-x</sub>Pt<sub>x</sub>Ga





A compensated magnetic state is achieved in Mn-Pt-Ga



Nayak et al., Nature Mater. 14, 679 (2015).

Large hysteresis indicates FM components inside AFM background

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### Large EB in Mn<sub>3-x</sub>Pt<sub>x</sub>Ga





EB > 3 T is achieved

- Pulsed field magnetization measurements display a close hysteresis loop with coercivity around 3.5 T.
- Field cooled MH loops measured in a dc magnetic field of 32 T confirms the presence of an extremely large EB.

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### Large EB in Mn<sub>3-x</sub>Pt<sub>x</sub>Ga





- EB monotonically decreases with temperatures.
- EB vanishes around 150 K.



EB up to room temperature in Mn-Fe-Ga system.

Nayak et al., Nature Mater. 14, 679 (2015).

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### Mn-Pt-Ga thin films:







#### Exhibit high ordering temperatures.

✓ Tunable magnetic anisotropy and compensated magnetic state is achieved.

Sahoo et al., Adv. Mater. 2016.

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### Mn-Pt-Ga bilayers:





Crossectional TEM view shows two distinct layers.



Similar compositions and identical crystal and electronic structures ensures high thermal stabilities.

Sahoo et al., Adv. Mater. 2016.

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### Anomalous Hall effect in non-collinear antiferromagnets





### Hall effect





#### Normal conductor

- ✓ Deflection of charge carriers in a presence of external magnetic field.
- ✓ Hall voltage linearly proportional to magnetic field.

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### Anomalous Hall effect





$$\rho_{xy} = \mathsf{R}_0 H_z + \mathsf{R}_s M_z,$$

- ✓ No need of magnetic fields.
- ✓ Intrinsic to all ferromagnetic materials.
- ✓  $\rho_{xy}^{A}$  roughly scale with the magnetization.







Chen et al., Phys. Rev. Lett. 112, 017205 (2014).

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A LETTERS JOURNAL EXPLORING THE FRONTIERS OF PHYSICS

December 2014

EPL, 108 (2014) 67001 doi: 10.1209/0295-5075/108/67001 www.epljournal.org

#### Non-collinear antiferromagnets and the anomalous Hall effect

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received 10 November 2014; accepted 2 December 2014 published online 10 December 2014

PACS 73.43.Cd – Theory and modeling PACS 75.50.Ee – Antiferromagnetics PACS 75.70.Tj – Spin-orbit effects

A bstract – The anomalous Hall effect is investigated theoretically by employing density functional calculations for the non-collinear antiferromagnetic order of the hexagonal compounds Mn<sub>3</sub>Ge and Mn<sub>3</sub>Sn using various planar triangular magnetic configurations as well as unexpected non-planar configurations. The former give rise to anomalous Hall conductivities (AHC) that are found to be extremely anisotropic. For the planar cases the AHC is connected with Weyl points in the energy-band structure. If this case were observable in Mn<sub>3</sub>Ge, a large AHC of about  $\sigma_{zx} \approx 900 \,(\Omega \text{cm})^{-1}$  should be expected. However, in Mn<sub>3</sub>Ge it is the non-planar configuration that is energetically favored, in which case it gives rise to an AHC of  $\sigma_{xy} \approx 100 \,(\Omega \text{cm})^{-1}$ . The non-planar configuration allows a quantitative evaluation of the topological Hall effect that is seen to determine this value of  $\sigma_{xy}$  to a large extent. For Mn<sub>3</sub>Sn it is the planar configurations that are predicted to be observable. In this case the AHC can be as large as  $\sigma_{yz} \approx 250 \,(\Omega \text{cm})^{-1}$ .

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### Anomalous Hall effect in noncollinear antiferromagnet Mn<sub>3</sub>Ge



Two layers of Mn triangles stacked along the c axis.





- ✓ Two spin triangles can be transformed into each other by a mirror reflection with respect to the xz plane.
- ✓  $\sigma^{k}_{ij}$  vanish if they align parallel to the mirror plane due to mirror symmetry.
- ✓ A non zero  $\sigma_{xz}^{y}$  ~330 (ohmcm)<sup>-1</sup> is expected.

#### Science Advances 2, e1501870 (2016).

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### Anomalous Hall effect in noncollinear antiferromagnet Mn<sub>3</sub>Ge



✓ A large anomalous Hall conductivity is found in the xz plane.



- ✓ A small residual in-plane moment can perturb the mirror symmetry.
- ✓ As predicted by theory the  $\sigma_{xy}$  is all most zero.

Science Advances 2, e1501870 (2016).

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### Anomalous Hall effect in noncollinear antiferromagnet Mn<sub>3</sub>Ge



- ✓ Angle dependent measurements show that even when field is applied along the ab-plane,  $\sigma_{xy}$  is almost zero.
- ✓ No effect of magnetic field on the observed AHE.
- Magnetic field only helps to change the sign of the AHE.



Science Advances 2, e1501870 (2016).

18.08.16

 $\theta$ [degree]

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### Anomalous Hall effect in noncollinear antiferromagnet Mn<sub>3</sub>Sn

# LETTER

doi:10.1038/nature15723

# Large anomalous Hall effect in a non-collinear antiferromagnet at room temperature

Satoru Nakatsuji<sup>1,2</sup>, Naoki Kiyohara<sup>1</sup> & Tomoya Higo<sup>1</sup>







### Non collinear spin structure and skyrmions in Heuslers





### Why non-collinear magnetic structure:





- ✓ A magnetic skyrmion is a vortex like topological object with a circular chiral spin configuration.
- ✓ Breaking of the inversion symmetry and D-M interaction for skyrmion.
- ✓ The competition between the direct exchange (-J<sub>ij</sub> s<sub>i</sub>.s<sub>j</sub>) and the Dzyaloshinskii-Moriya exchange (D<sub>ij</sub>. (s<sub>i</sub>×s<sub>j</sub>)) gives rise to skyrmion.
- ✓ Good candidates for future data storage.



#### Nature 465, 901 (2010).

✓ Lorentz TEM image of skyrmions

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### Non-collinear magnetic structure in Mn<sub>2</sub>RhSn





- First time observed a spin-reorientaion transition in the Heusler tetragonal compound Mn<sub>2</sub>RhSn.
- Total magnetic moment of 2μB/f.u indicates a noncollinear magnetic structure in Mn<sub>2</sub>RhSn.

Meshcheriakova et al., Phys. Rev. Lett. 113, 087203 (2014).

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### Non-collinear magnetic structure in Mn<sub>2</sub>RhSn



 $M = 3.5 + 3.8 \mu_B = 6.5 \mu_B$ 



FI ordering does not hold with experimental result

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 $M = MnI_z - MnII_z = 3.5 - 1.5 \mu_B = 2\mu_B$ 

Noncollinear ordering due to competing FM and AFM interactions.

Meshcheriakova et al., Phys. Rev. Lett. 113, 087203 (2014).





FM ordering does not hold with experimental result

### Non-collinear magnetic structure in Mn<sub>2</sub>RhSn





- Neutron diffraction study confirms the noncollinear ordering.
- In-plane component of the Mn moment completely disappears above the spin-reorientation transition.

Meshcheriakova et al., Phys. Rev. Lett. 113, 087203 (2014).

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### Topological Hall effect in Mn<sub>2</sub>RhSn thin films





Existence of topological Hall effect below the spin-reorientation transition indicates presence of skyrmion like structure.

Rana et al., New J. Phys. 2016.

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### Non-collinear magnetic structure in Mn-Pt-Sn





### Skyrmions in Pd-doped Mn-Pt-Sn



The skyrmion phase can be stabilized up to 400 K, which is the  $T_{\rm C}$  of the present material.

### Skyrmions in Pd-doped Mn-Pt-Sn



Skyrmion phase can be stabilized in zero magnetic field at low temperatures.



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120 nm

### Skyrmions in Pd-doped Mn-Pt-Sn





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#### ✓ Designing compensated magnetic state for antiferromagnetic spintronics.

We have successfully designed compensated magnetic state in Mn-Pt-Ga system and use this to achieve a large exchange bias.

#### ✓ AHE in non-collinear antiferromagnets.

We have shown that Mn<sub>3</sub>Ge, which has a non-collinear antiferromagnetic spin structure can give a large AHE at room temperature.

#### ✓ Heuslers in terms of non-collinear magnetism.

Existence of both non-collinear magnetic states and skyrmions have been shown in the Heusler system.

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### What is exchange bias?







#### Exchange-biased hysteresis loop

#### Technological importance:

- Extensively used in all GMR and TMR based devices
- Permanent magnet



Interfacial exchange interaction between FM and AFM layers

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### Mn<sub>2</sub>PtGa: ZFC exchange bias





MnI=3.65 $\mu_{B}$ , MnII=3.1 $\mu_{B}$ M=3.65-3.1=0.55 $\mu_{B}$ 

Tetragonal crystal structure and ferrimagnetic ordering



✓ A large EB of 0.2T both in ZFC and FC modes.

Nayak et al., Phys. Rev. Lett. 110, 127204 (2013).

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Non-collinear magnetic structure in Mn-Pt-Sn





- A spin-reorientation transition is observed around 150 K in Mn-Pt-Sn.
- The ac-susceptibility measurements show the existence of a dip (in zero magnetic field) just below the spin-reorientation transition.

Nayak et al., Under review.

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### Non-collinear magnetic structure in Mn-Pt-Sn



These stripes may correspond to the helicoid/cycloid magnetic structure with periods nearly 110 nm.

Nayak et al., Under review.

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