

# Altermagnetism: Emerging Opportunities in a New Magnetic Phase

## Magnetic octupoles as the order parameter for unconventional antiferromagnetism

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[syantika.bhowal@mat.ethz.ch](mailto:syantika.bhowal@mat.ethz.ch)

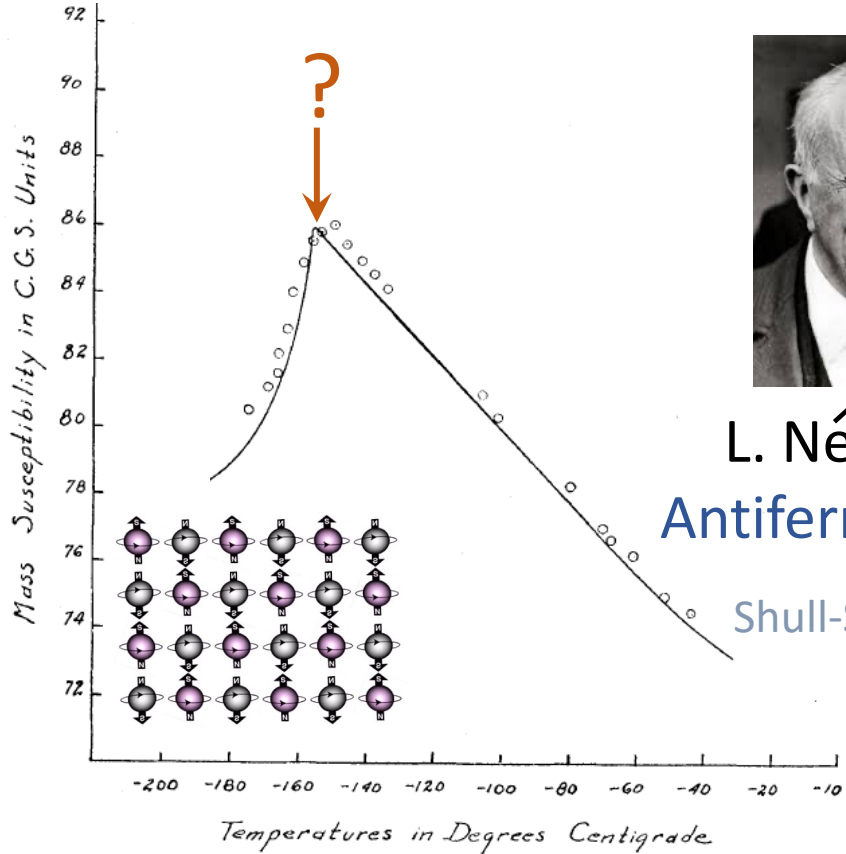
Wednesday, May 10th



# Antiferromagnetism: an early example of “hidden” magnetic order

## The Magnetic Susceptibility of MnO as a Function of the Temperature

RAYEN WELCH TYLER, *University of Illinois*  
(Received June 10, 1933)



L. Néel (1934)

Antiferromagnetism

Shull-Smart, PRB 76, 1256 (1949)

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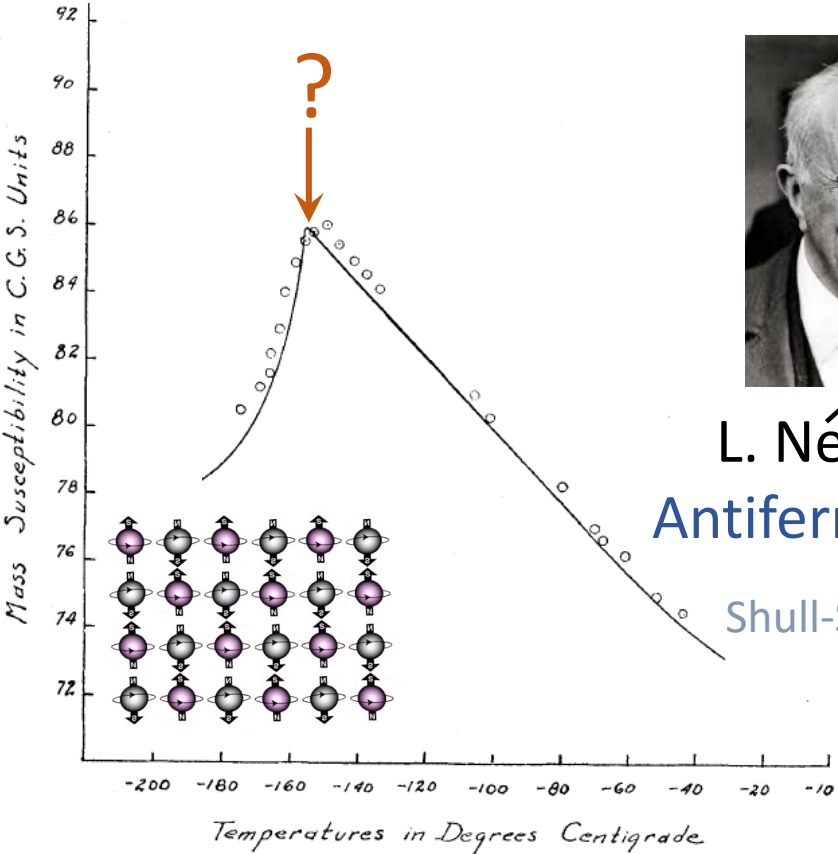
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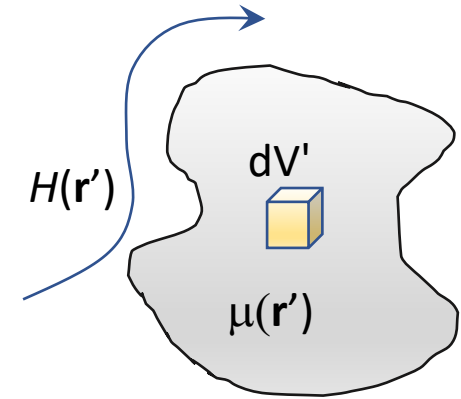
Antiferromagnetism

Shull-Smart, PRB 76, 1256 (1949)



- Inconvenience of staggered magnetization  $\mathbf{L} = \mathbf{M}_1 - \mathbf{M}_2$
- Absence of any ferroic ordering
- No information on conjugate field to select magnetic domain
- Can not distinct AFMs with & w/o broken time-reversal symmetries

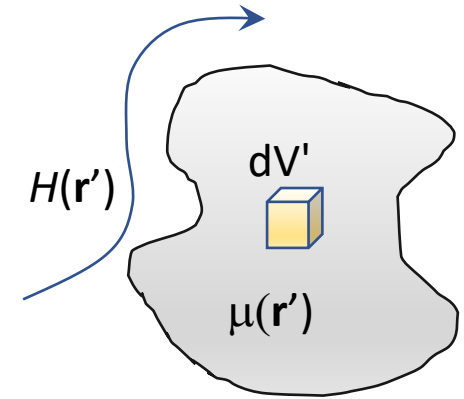
# Magnetic multipoles : Recent example of “hidden” magnetic order



$$\begin{aligned}
 \mathcal{E}_{\text{int}} &= - \int \vec{\mu}(\vec{r}') \cdot \vec{H}(\vec{r}') dV' \\
 &= -\vec{H}(0) \cdot \int \vec{\mu}(\vec{r}') dV' - \underbrace{\partial_i H_j(0)}_{\text{Magnetic dipole}} \int r'_i \mu_j(\vec{r}') dV' - \underbrace{\partial_i \partial_j H_k(0)}_{\text{Magnetolectric multipole}} \int r'_i r'_j \mu_k(\vec{r}') dV' .. \\
 &= -\vec{m} \cdot \vec{H}(0) - \mathcal{M}_{ij} \partial_i H_j(0) - \mathcal{O}_{ijk} \partial_i \partial_j H_k(0) ..
 \end{aligned}$$

Ederer *et. al.*, PRB **76**, 214404 (2007); Spaldin *et.al.*, PRB **88**, 094429 (2013)

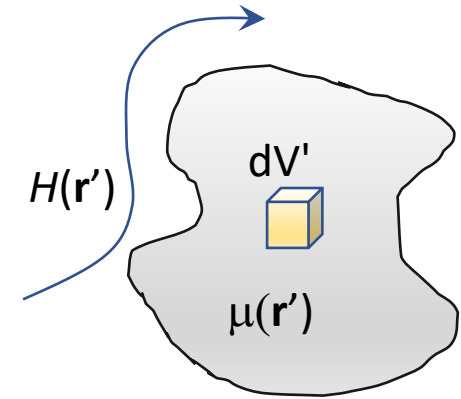
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Magnetic dipole
Magnetolectric multipole
Magnetic octupole

Ederer *et. al.*, PRB **76**, 214404 (2007); Spaldin *et.al.*, PRB **88**, 094429 (2013)

## ON THE MAGNETO-ELECTRICAL EFFECT IN ANTIFERROMAGNETS

I. E. DZIALOSHINSKIĬ

Institute for Physical Problems, Academy of Sciences, U.S.S.R.

Submitted to JETP editor June 17, 1959

J. Exptl. Theoret. Phys. (U.S.S.R.) **37**, 881-882 (September, 1959)

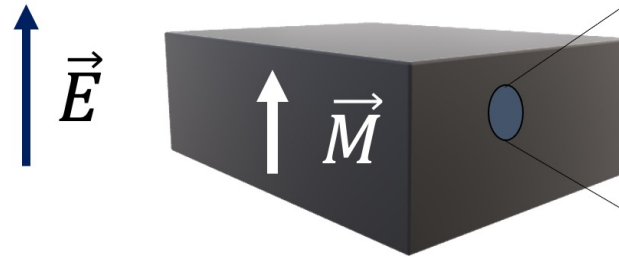
$$\begin{aligned} \mathbf{P}_i &= \alpha^{ME}_{ij} H_j \\ \mathbf{M}_i &= \alpha^{ME}_{ji} E_j \end{aligned}$$

Astrov, Sov. Phys. JETP **11**, 708 (1960)



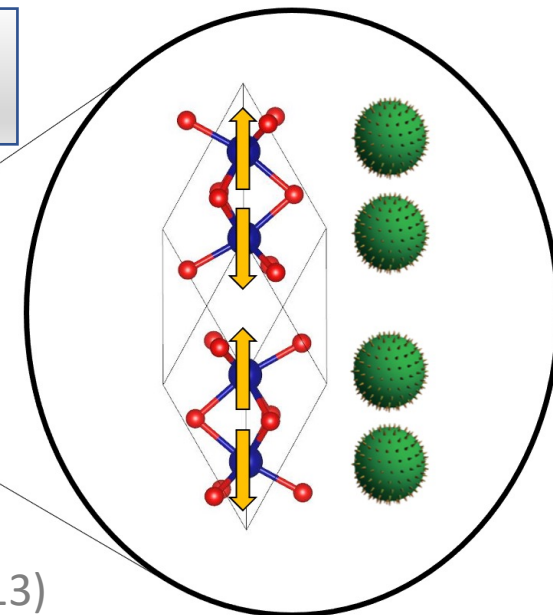
I. Dzyaloshinskii

AFMs with ferroic ordering of magnetolectric multipoles → Magnetolectric effect



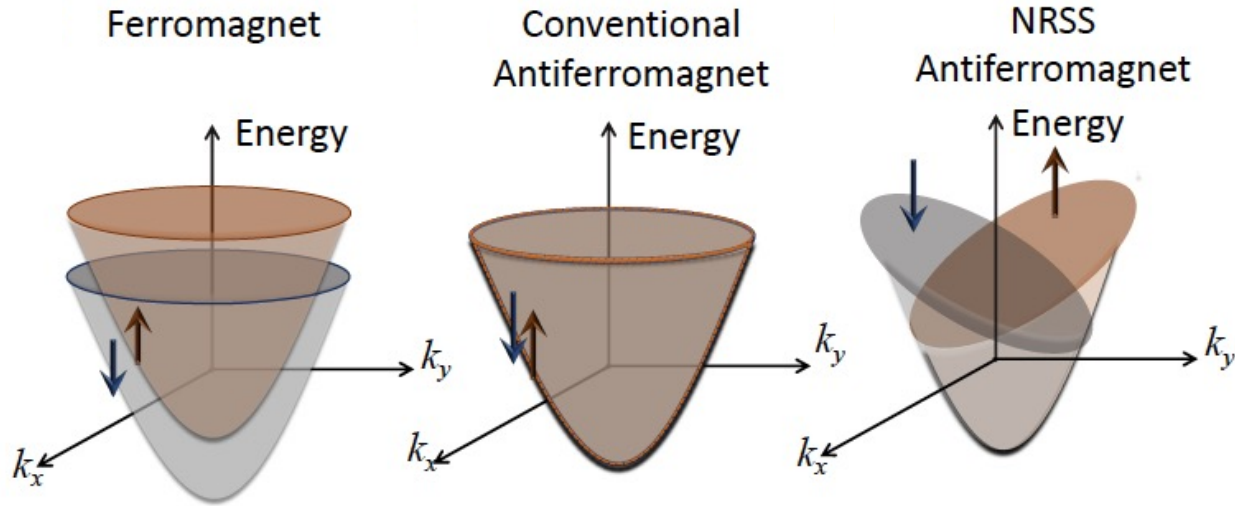
Spaldin *et. al.*, Phys. Rev. B **88**, 094429 (2013)

Verbeek *et. al.*, arXiv:2303.00513 (2023)



Courtesy: X. Verbeek

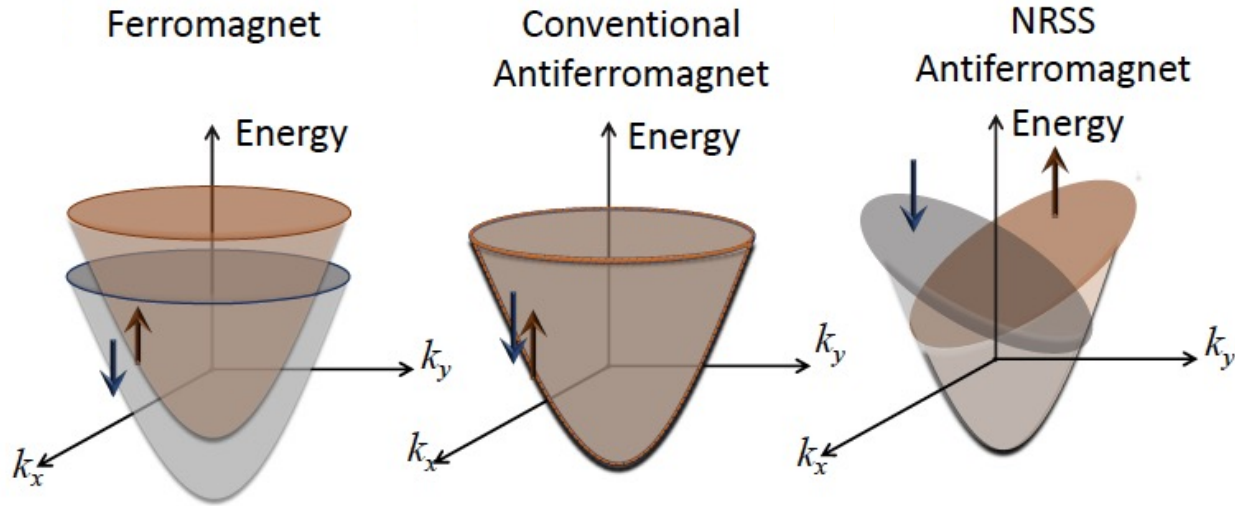
# A new type of magnetism: “Altermagnetism”



Smejkal *et al.*, *Sci. Adv.* **6**, eaaz8809 (2020)  
Naka *et al.*, *Nat. Commun.* **10**, 4305 (2019)  
Ahn *et al.*, *Phys. Rev. B* **99**, 184432 (2019)  
Hayami *et al.*, *J. Phys. Soc. Jpn.* **88**, 123702 (2019)  
Yuan *et al.*, *Phys. Rev. B* **102**, 014422 (2020); *Phys. Rev. Materials* **5**, 014409 (2021)  
Smejkal *et al.*, *Phys. Rev. X* **12**, 031042 (2022); *Phys. Rev. X* **12**, 011028 (2022)  
Mazin, Editorial: *Phys. Rev. X* **12**, 040002 (2022)  
Mazin *et al.*, *PNAS* **118**, e2108924118 (2021)  
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Emerging properties: Spontaneous Hall effect, Giant magnetoresistance, spin current generations, etc..

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Emerging properties: Spontaneous Hall effect, Giant magnetoresistance, spin current generations, etc..

Centrosymmetric “altermagnets” with non-relativistic spin splitting

$$\mathcal{E}_{\text{int}} = -\vec{m} \times \vec{H}(0) - \mathcal{M}_{ij} \partial_i H_j(0) - \mathcal{O}_{ijk} \partial_i \partial_j H_k(0) ..$$

Magnetic octupole



# Plan of talk : A classic example of “antiferromagnet”

MnF<sub>2</sub>

PHYSICAL REVIEW

VOLUME 90, NUMBER 5

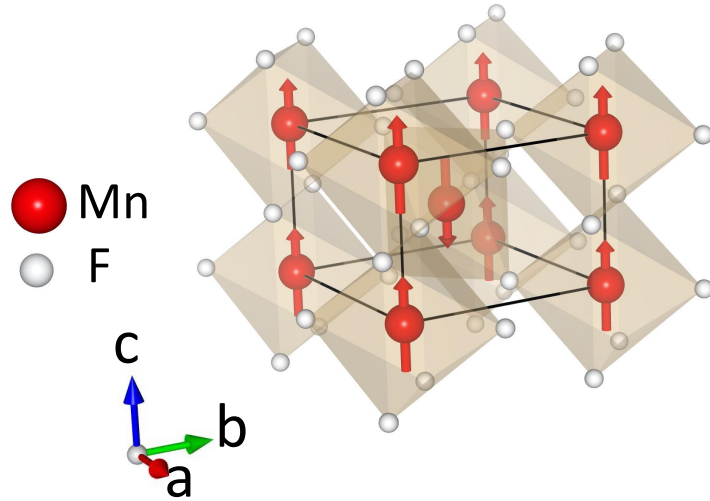
JUNE 1, 1953

## Neutron Diffraction Studies of Antiferromagnetism in Manganous Fluoride and Some Isomorphous Compounds\*

R. A. ERICKSON†,‡

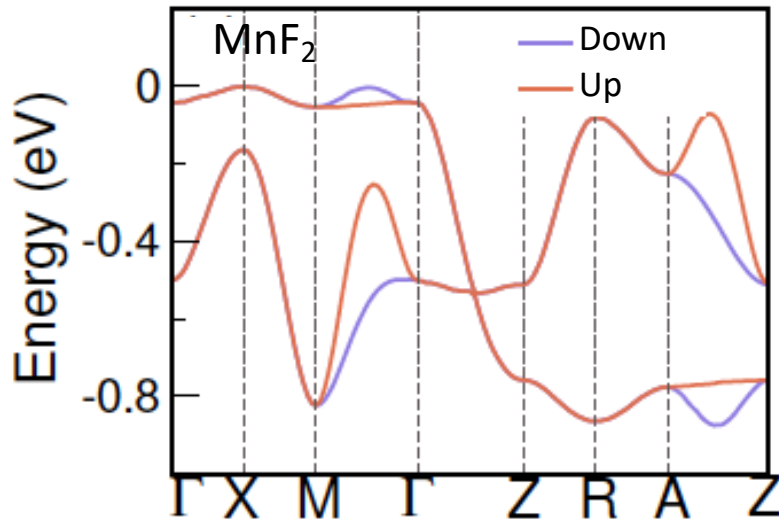
Laboratory, Oak Ridge, Tennessee and Agricultural and Mechanical College of Texas, College Station, Texas

(Received February 24, 1953)



## Magnetic octupole

- Correlation to structure and spin : octupolar domains
- Relevance to non-relativistic spin splitting



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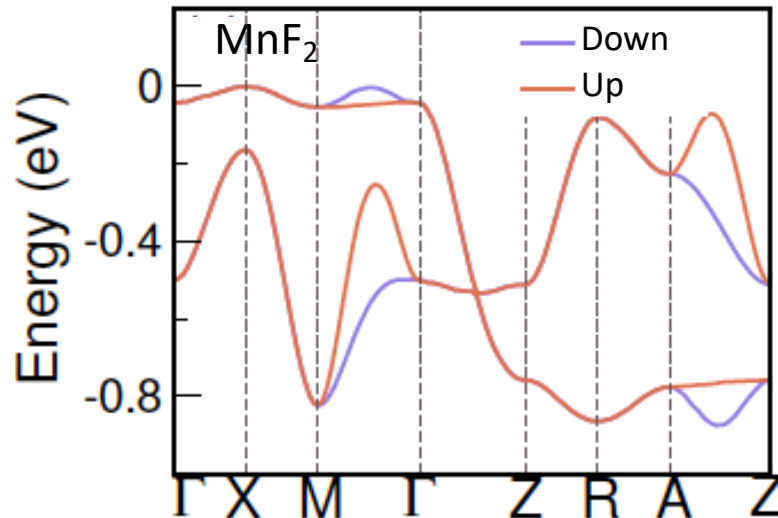
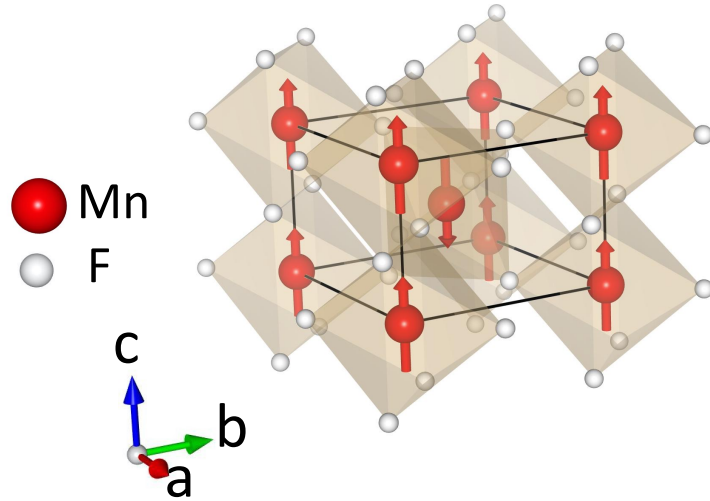
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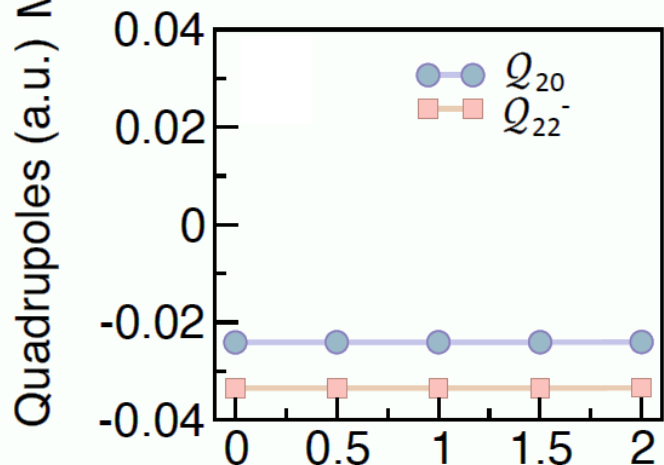
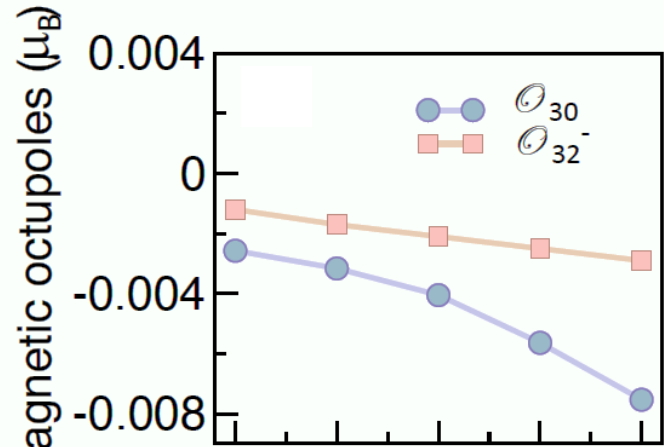
## Implication of magnetic octupoles

- Piezo & antipiezo-magnetic effects
- Magnetic Compton profile

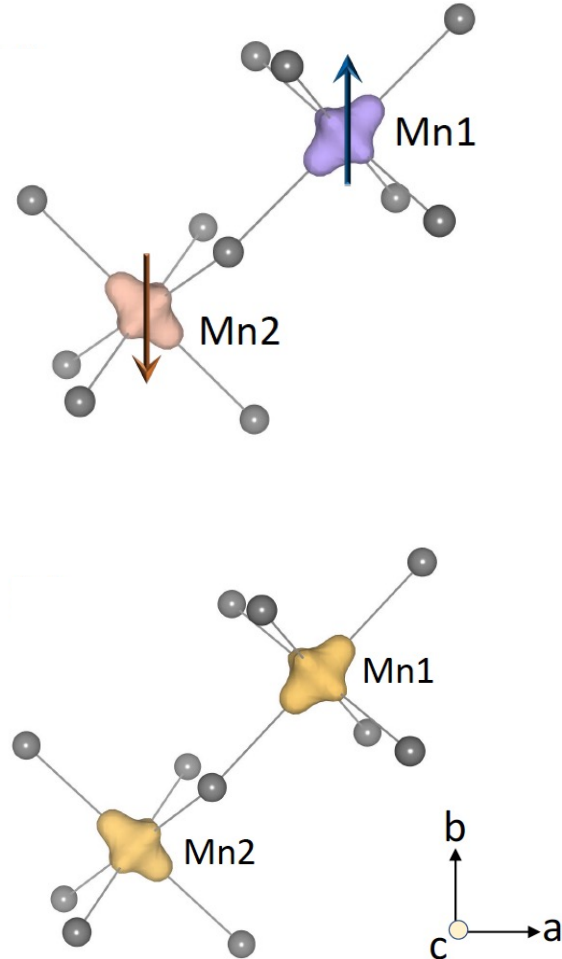
# Magnetic octupoles

Magnetic octupoles: Ferro-type ordering :  $\mathcal{O}_{32^-} (xym_z)$ ,  $Q_{x^2-y^2}^{(\tau)}$ ; Anti-ferro-type ordering :  $\mathcal{O}_{30} [(3z^2-r^2)m_z]$ ,  $t_z^{(\tau)}$

Charge quadrupoles:  $Q_{20}$  (Ferro),  $Q_{22^-}$  (AF)



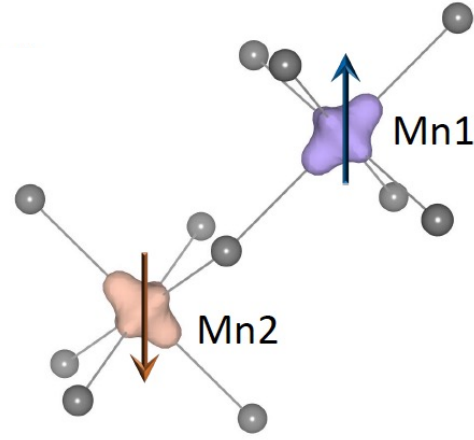
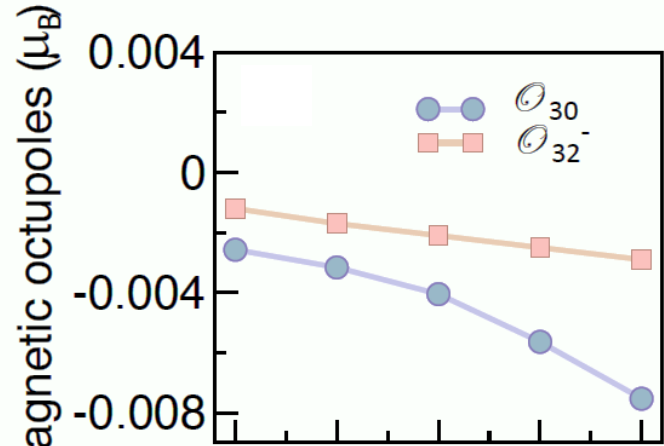
Relative SOC strength



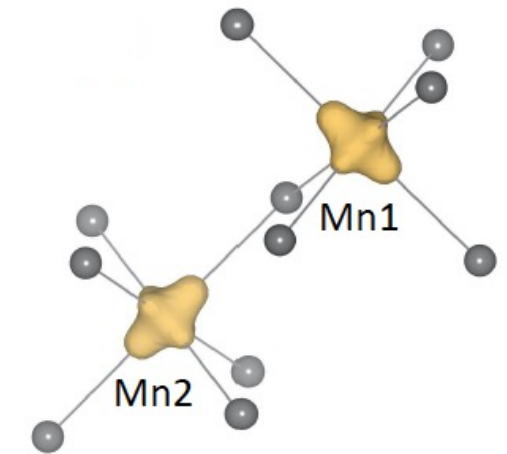
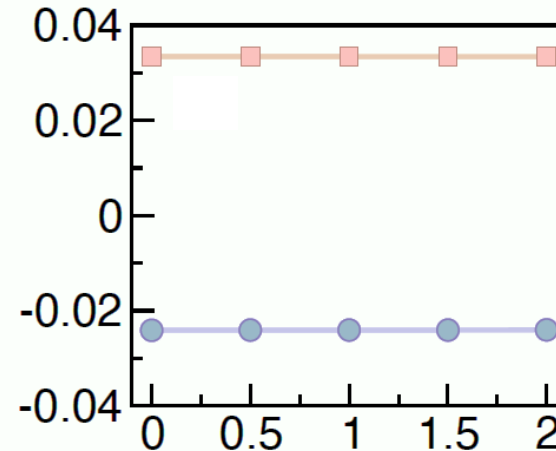
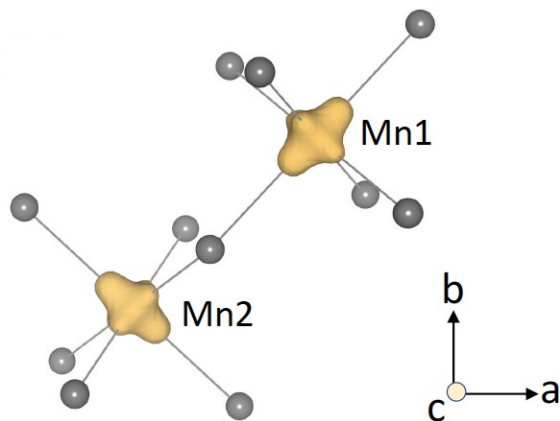
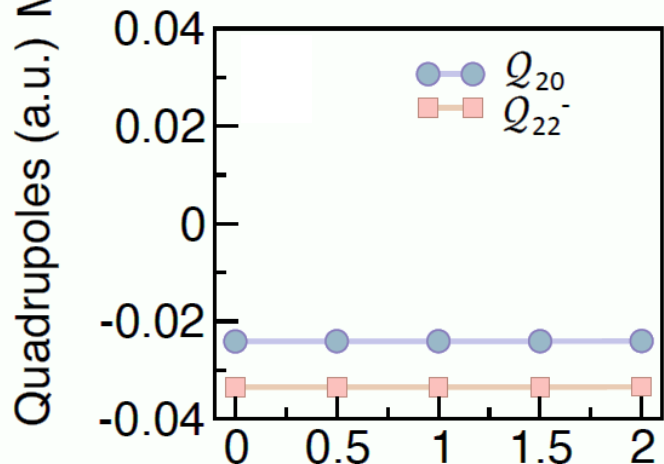
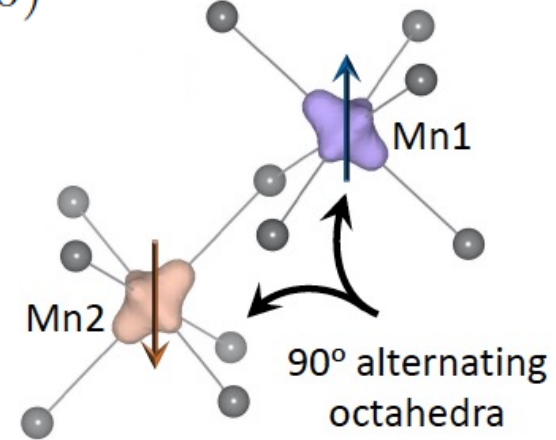
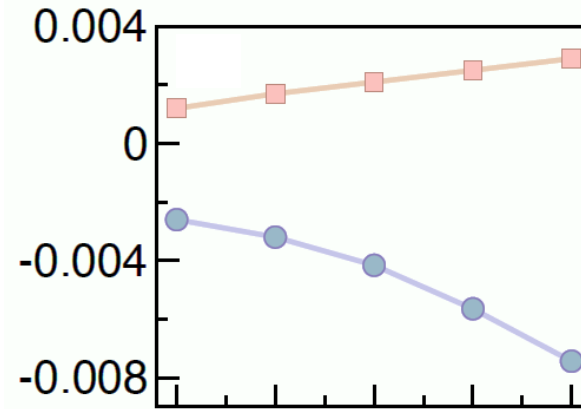
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Charge quadrupoles:  $Q_{20}$  (Ferro),  $Q_{22^-}$  (AF)



$4f : (x, x, 0) \rightarrow 4g : (x, -x, 0)$

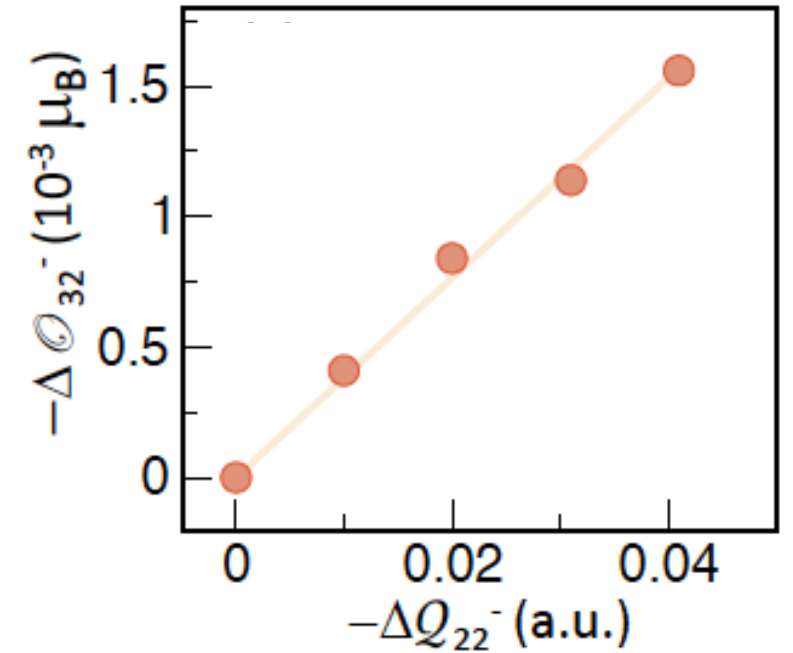
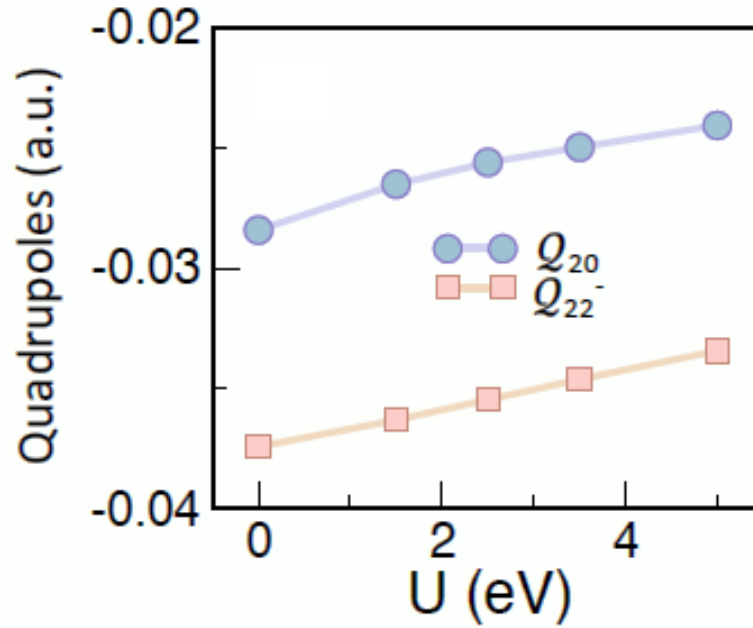
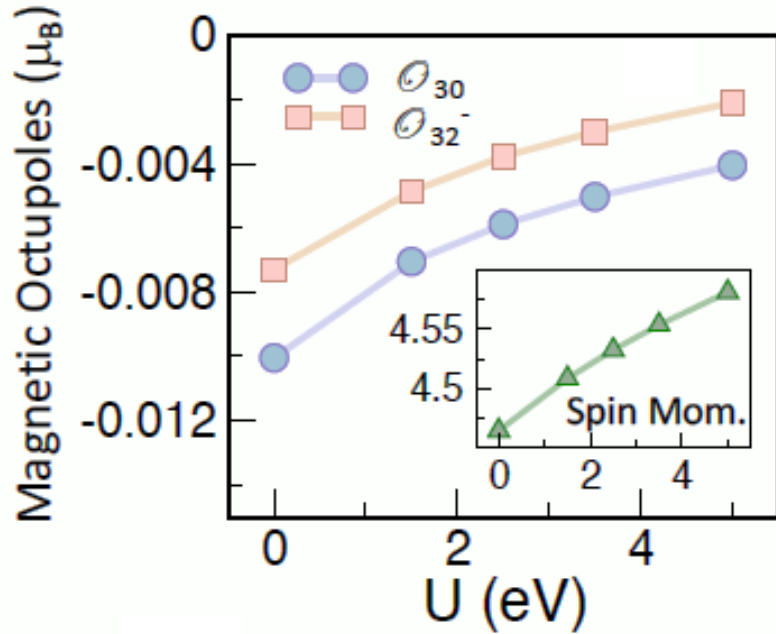


Relative SOC strength

Relative SOC strength



# Quantify the effect of neighboring nonmagnetic ions

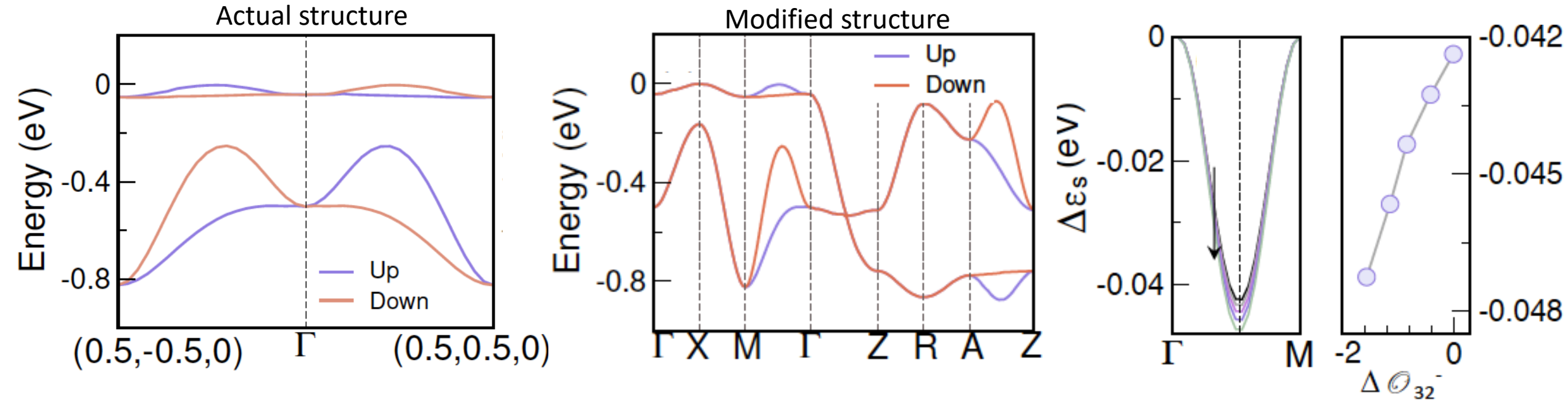


- Variation with Hubbard U : similar behavior for quadrupoles and octupoles
- Quantify the effect of neighboring nonmagnetic F environment via electric quadrupoles
- Systematic increase in the octupoles as the quadrupole moment increases

L. Schaufelberger, M. E. Merkel, A. M. Tehrani, N. A. Spaldin, and C. Ederer (to be published)

<https://github.com/materialtheory/multipyles>

# Spin splitting and its tuning



$$xym_z \rightarrow k_x k_y m_z$$

Watanabe-Yanase, PRB **98**, 245129 (2018)

- Reversal of spin splitting with **90° rotation**
- **Reversal of spin splitting** for the opposite sign of magnetic octupoles
- **Controlling spin splitting** via tuning the strength of the magnetic octupole
- Crucial insight into **conjugate fields** for the formation of magnetic domain



Product of stress (rank-2, even under inversion) and magnetic field (odd under TR)

Baruchel *et. al.*, J. Phys. Colloques **49**, C8 (1988)

# Plan of talk : A classic example of “antiferromagnet”

MnF<sub>2</sub>

PHYSICAL REVIEW

VOLUME 90, NUMBER 5

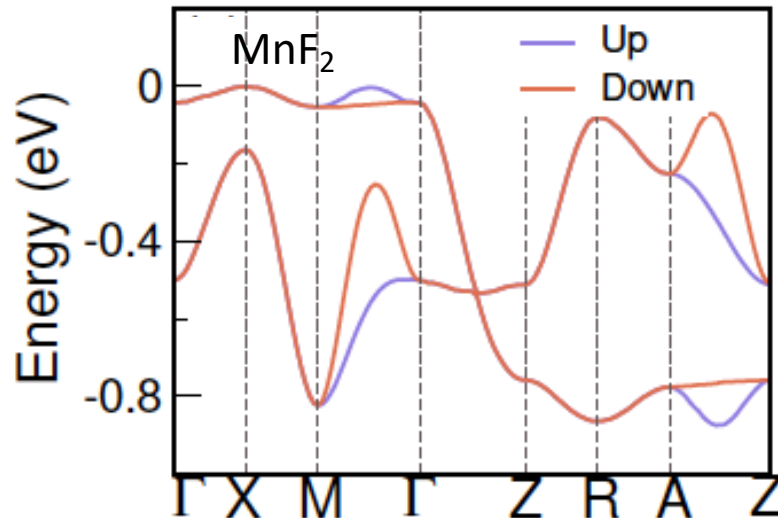
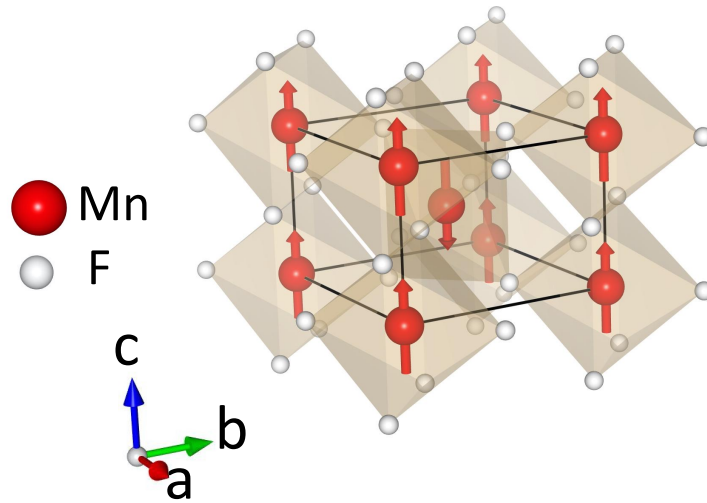
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## Magnetic octupole

- Correlation to structure and spin: octupolar domains
- Relevance to non-relativistic spin splitting

## Implication of magnetic octupoles

- Piezo & antipiezo-magnetic effects
- Magnetic Compton profile

# Implications of magnetic octupoles: Piezo and anti-piezomagnetic effects

Piezomagnetic effect: Application of stress generates a change in net magnetization

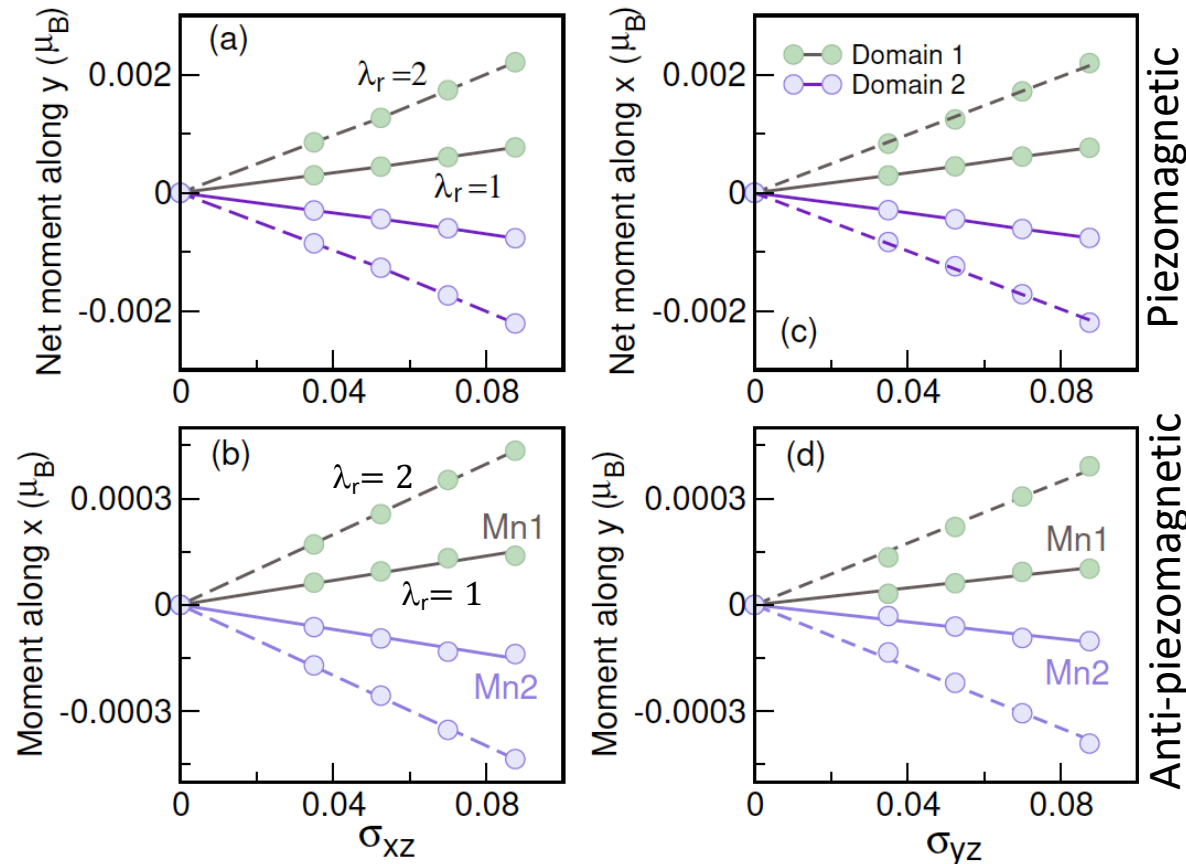
Allowed components in MnF<sub>2</sub>:

$$\mathcal{M}_x = \Lambda_{xyz}\sigma_{yz}, \mathcal{M}_y = \Lambda_{yxz}\sigma_{xz}, \mathcal{M}_z = \Lambda_{zxy}\sigma_{xy}$$

Baruchel *et. al.*, JMMM 15-18, 1510 (1980); Borovik-Romanov J. Exptl. Theoret. Phys. **38**, 1088 (1960)

Magnetic Octupole tensor  $\mathcal{O}_{ijk} \rightarrow$  Piezomagnetic response  $\Lambda_{ijk}$

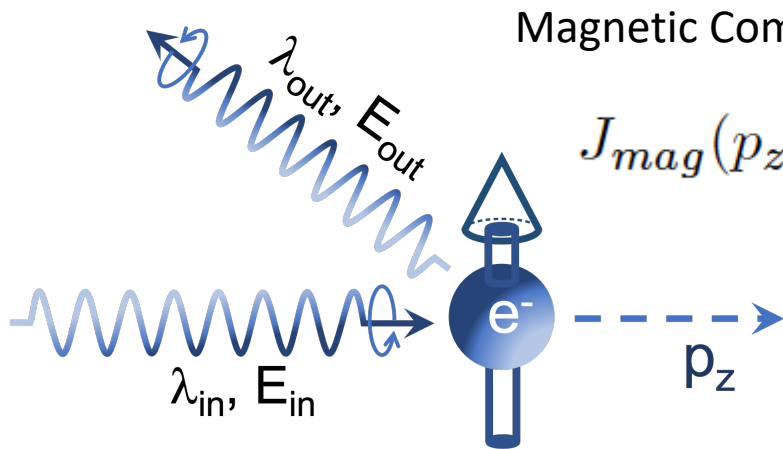
Urru-Spaldin, Ann. Phys. **447**, 168964 (2022)



- **Universal** to all materials with non-zero magnetic octupoles
- **Reversal** for opposite magnetic domains
- Depends on **SOC** effect
- Prediction of **anti-piezomagnetic effect**
- Same dependence on spin-orbit coupling strength and magnetic domain

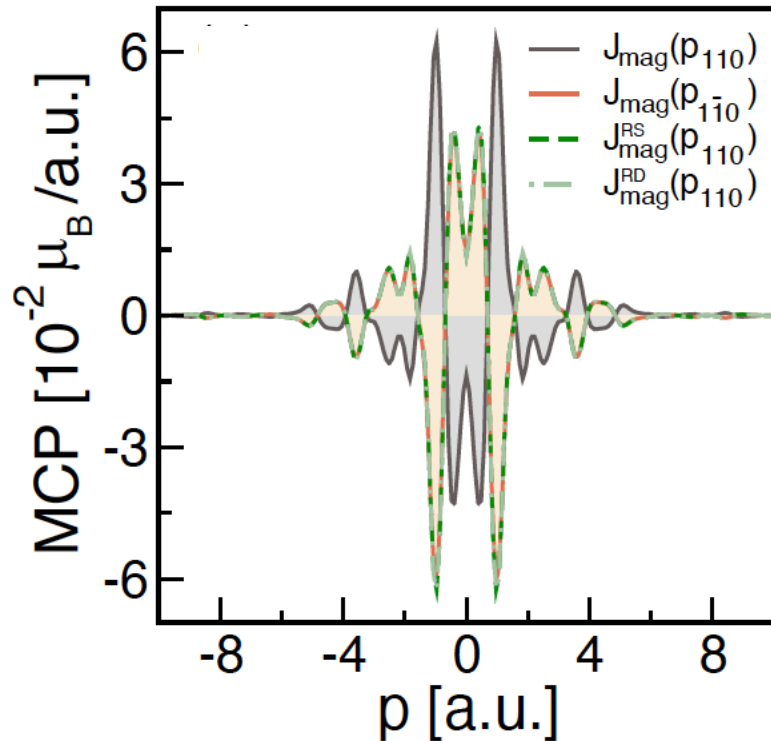


# Direct detection of magnetic octupole via magnetic Compton scattering



$$J_{mag}(p_z) = \int \int [\rho^\uparrow(\vec{p}) - \rho^\downarrow(\vec{p})] dp_x dp_y$$

Platzman-Tzoar, PRB **2**, 3556 (1970)

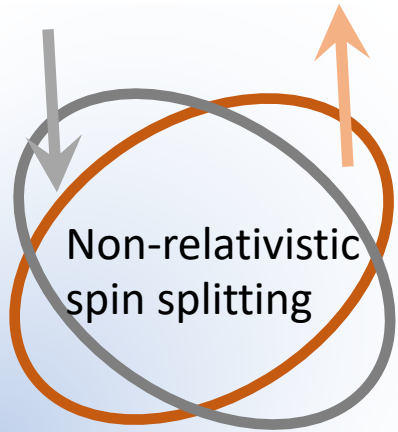


- Non-zero MCP, unusual for conventional antiferromagnets
- MCP is symmetric in  $p$
- Occurs w/o SOC
- Much larger magnitude than ferroelectrics

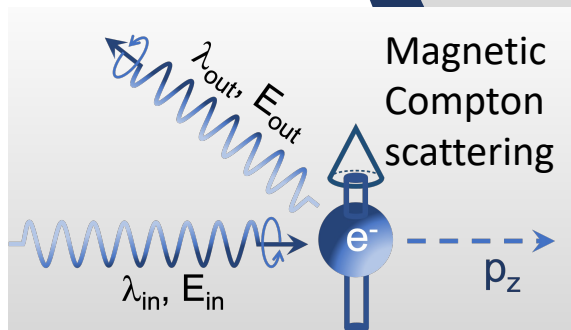
Bhowal-Collins-Spaldin, PRL **128**, 116402 (2022)

- The integral of the MCP is zero  $\rightarrow$  zero net moment
- MCPs along (110) and (1-10) have opposite signs
- Experiments require single magnetic domain (piezomagnetic annealing)

# Summary and outlook



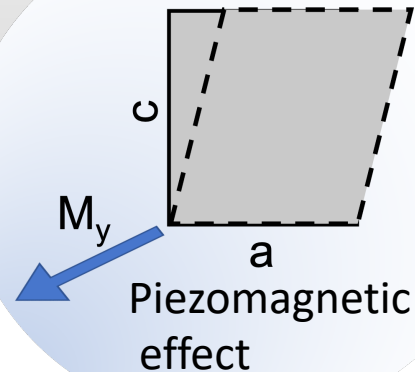
## Magnetic octupole



Plethora of intriguing physical properties and insights:

- Broken time-reversal symmetry
- Large NRSS
- Magnetic domains & conjugate field
- (anti) Piezomagnetic effect
- Magnetic Compton profile in antiferromagnets

Bhowal-Spaldin, arXiv:2212.03756 (2022)



- Second-order magnetoelectric effect
- Surface magnetism
- Spin-phonon interactions

Connection between sub-fields of physics → New avenues for future exploration

# Acknowledgements

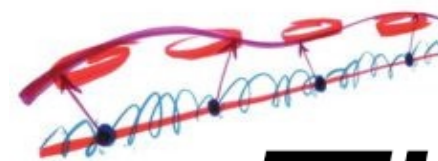
**ETH** zürich



Prof. Nicola Spaldin

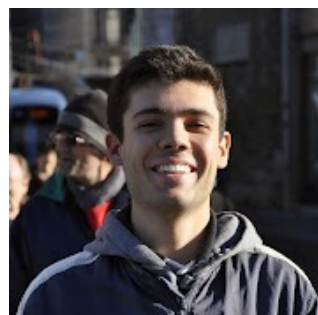


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Andrea Urru



Sophie Weber



Maximilian Ernest Merkel



Group members at ETH



Prof. Steve Collins



Dr. Jon Duffy



Dr. Urs Staub





# Acknowledgements

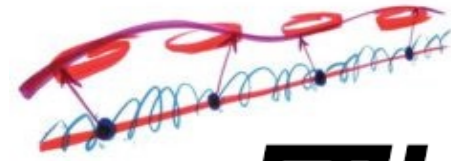
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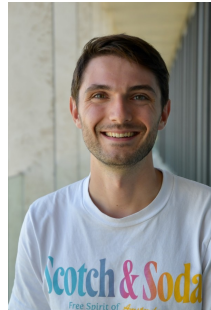
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