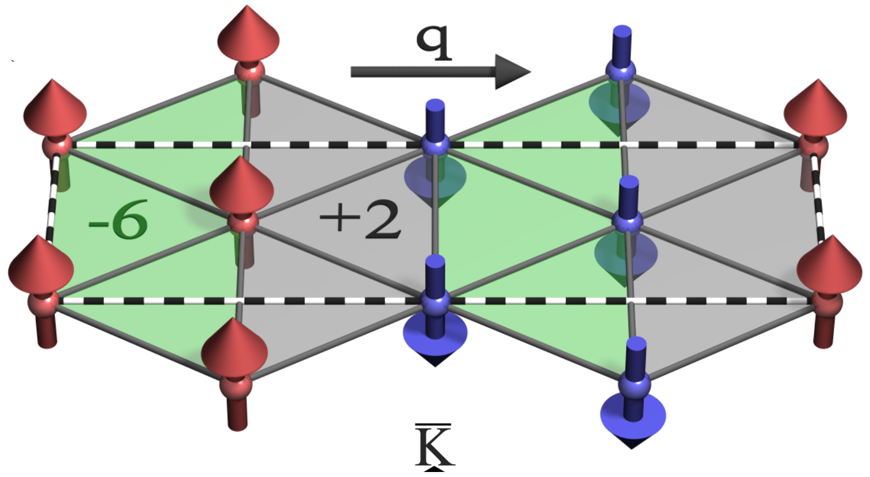
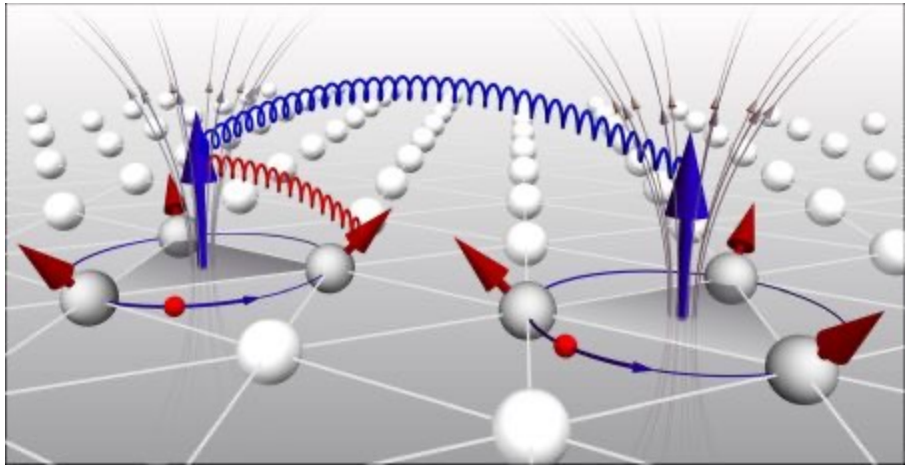




# DERIVING SPIN MODELS FROM DENSITY FUNCTIONAL THEORY: CHALLENGES AND LIMITATION

20.11.2022 I G. BIHLMAYER , D. MAZHJOO, B. ZIMMERMANN, S. GRYTSIUK, M. HOFFMANN, AND S. BLÜGEL
PETER GRÜNBERG INSTITUT AND INSTITUTE FOR ADVANCED SIMULATION
FORSCHUNGSZENTRUM JÜLICH AND JARA, 52425 JÜLICH, GERMANY

# ACKNOWLEDGEMENTS

Donya Mazhjoo

Bernd Zimmermann

Markus Hoffmann

Sergii Grytsiuk

Stefan Blügel

and the members of the Institute Quantum Theory of Materials, J. P. Hanke, S. Lounis and Y. Mokrousov, further B. Dupé, S. Heinze and his team from University Kiel and many more…

SPP2137 Skyrmionics
Topological Spin Phenomena in Real-Space for Applications

HELMHOLTZ GEMEINSCHAFT 20 Jahre

CRC1238 Control and Dynamics of Quantum Materials


DARPA
Topological Excitations in Electronics (TEE)

MAGic Sky
Funded by the Horizon2020 Framework Programme of the European Union

JARA Jülich Aachen Research Alliance

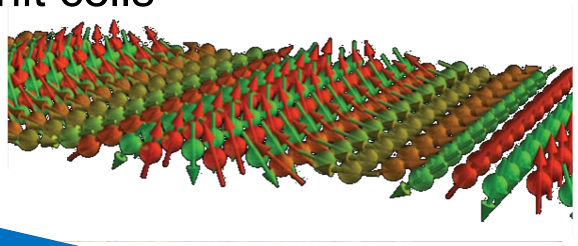
JÜLICH Forschungszentrum

# MOTIVATION(1): MAPPING TO SPIN MODELS

## strategies for finding the magnetic ground state

**Magnetic structure** ⟷ **Electronic structure**

| Magnetic structure | Electronic structure |
|---|---|
| various solutions<br>may extend over many chemical unit cells | single solution<br>symmetry of the chemical unit cell |

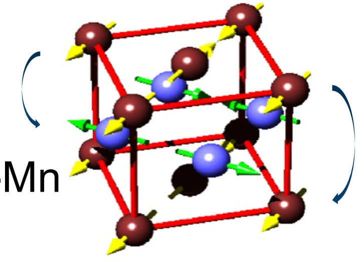


↓

similar to (structural) relaxation,
similar methods

↓

force field models
classical Heisenberg
model + extensions

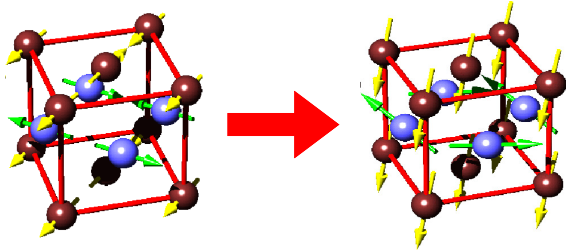


$J_{Fe\text{-}Mn}$ $J_{Fe\text{-}Fe}$

analytically

numerically

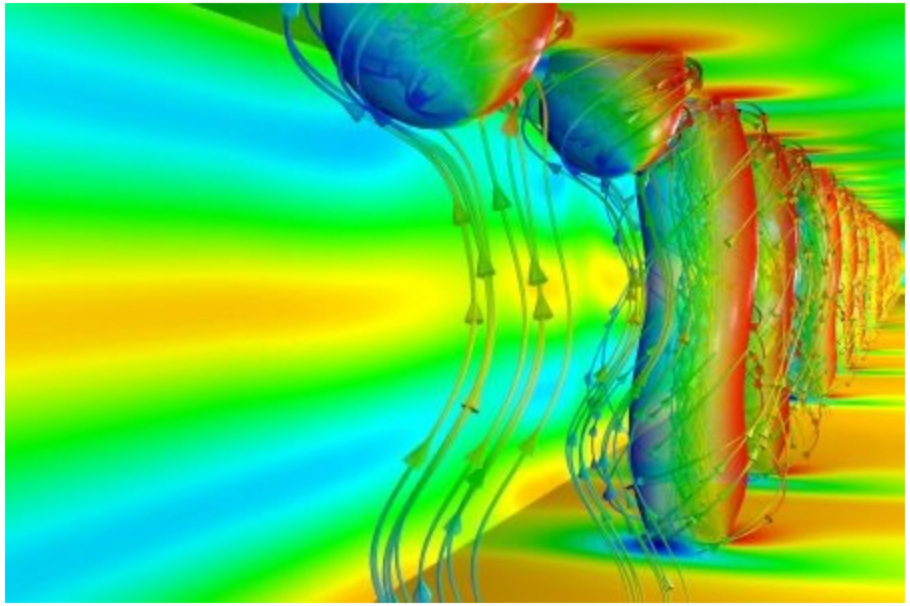
molecular dynamics
spin dynamics



large unit cells
usually too expensive

# MOTIVATION(2): MAPPING TO SPIN MODELS

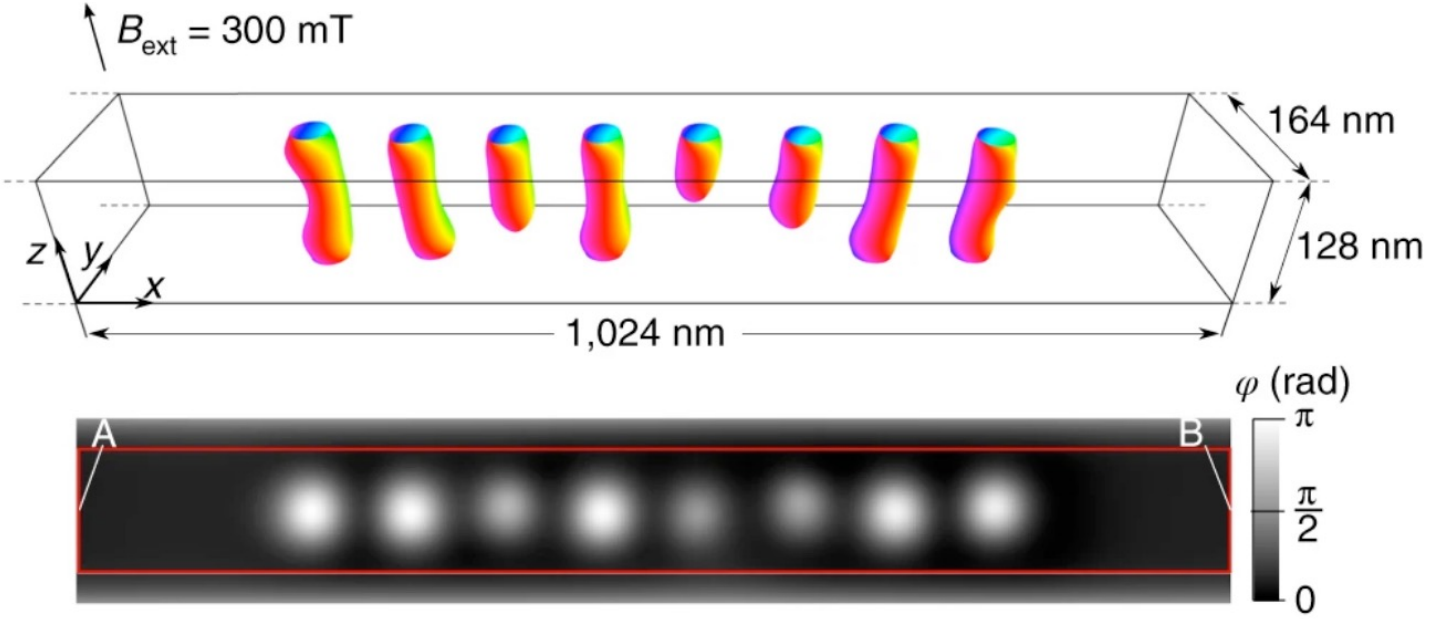
## obtain parameters for large scale simulations



Micromagnetic simulations

$$E = \int_{V_s} \{E_{ex} + E_{DMI} + E_Z + E_d\} d\mathbf{r}$$

exchar | DM | ext. | dipole interaction (M)




Experimental observation of chiral magnetic bobbers in B20-type FeGe, Zheng et al., Nature Nanotechnology **13**, 451 (2018)

# INTRODUCTION

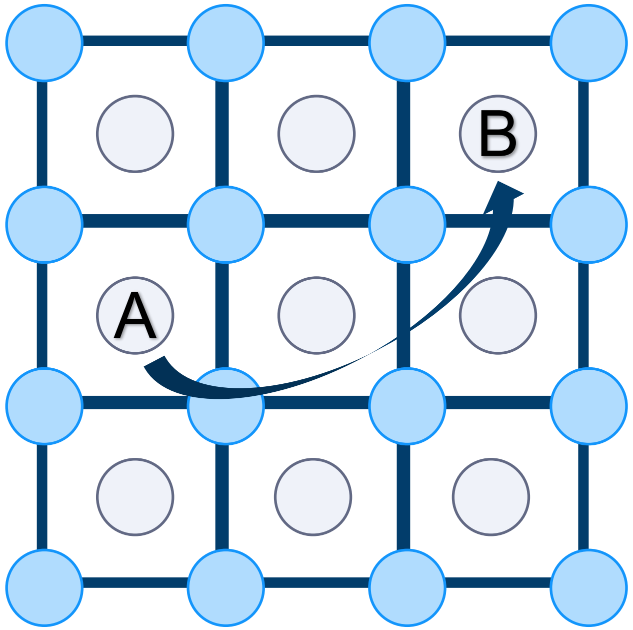
- Heisenberg-type interactions
- higher-order interactions
- relativistic interactions
- DFT tools

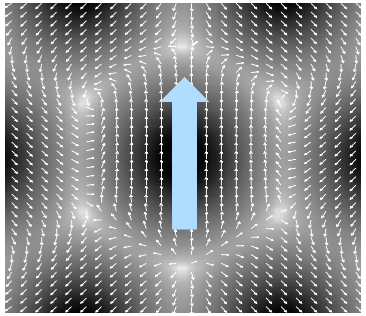
# SPIN MODELS AND DFT

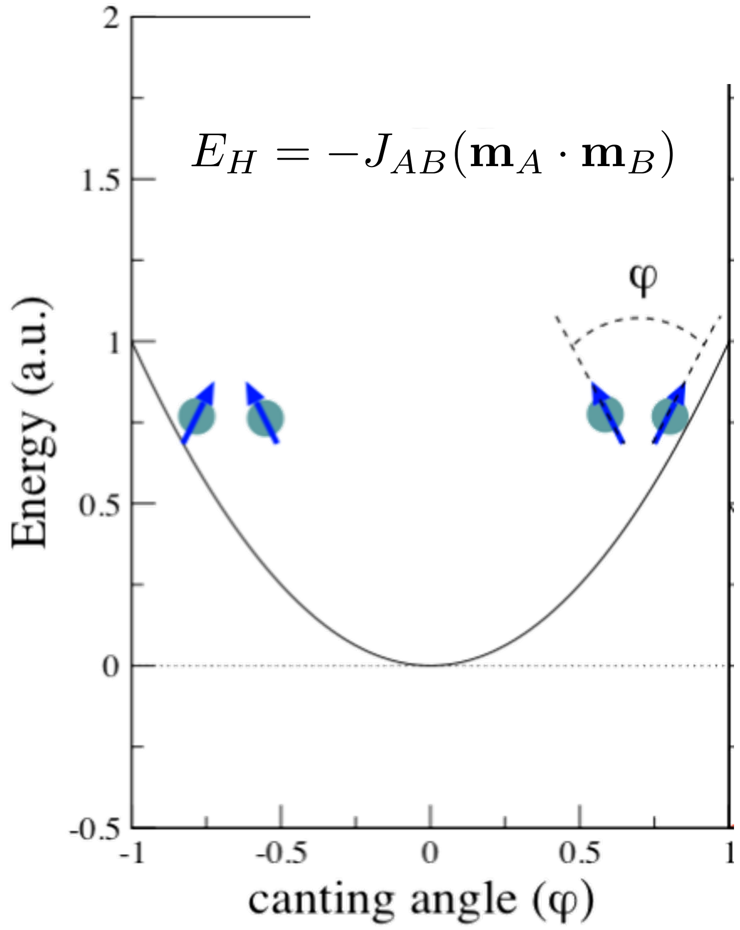
## minimal requirements

$$H = -\sum_{i,j} J_{ij}\mathbf{m}_i \cdot \mathbf{m}_j$$

- well defined magnetization directions at sites A and B
- length of magnetic moments stable against rotations
- independence of $J_{AB}$ from magnetic configuration
- independence of $J_{AB}$ from canting angle φ
- method to extract energy change (e.g. constrained DFT)







JÜLICH Forschungszentrum

# HEISENBERG MODEL

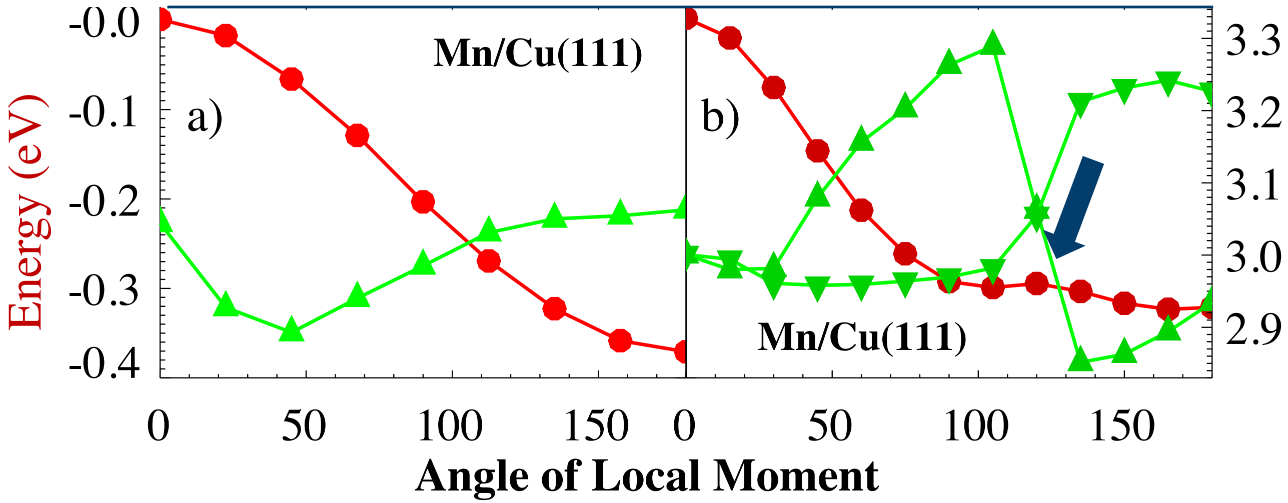
## triangular lattice

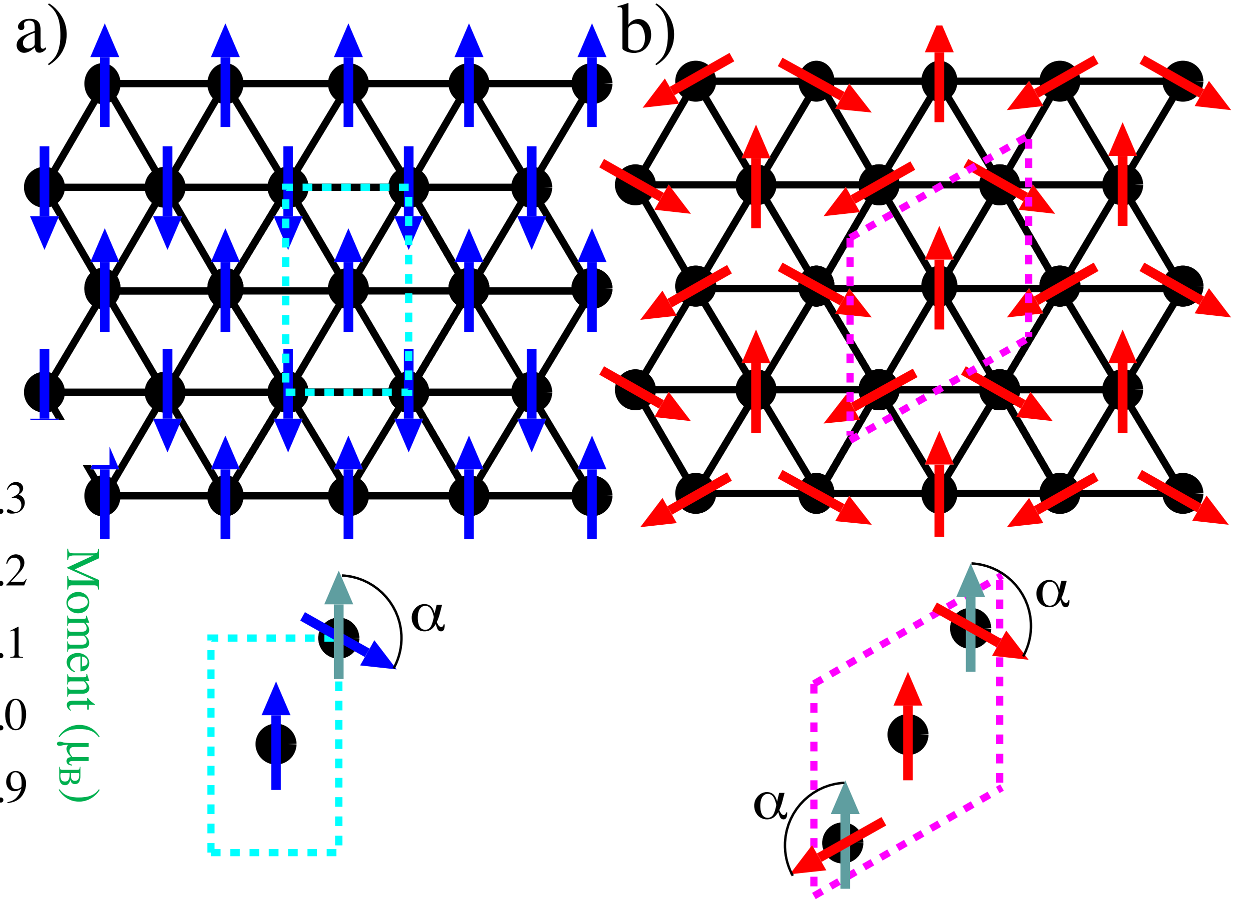
Including interactions up to $J_3$ we obtain
a):

$$E(\alpha) = -\{(J_1 + J_2)(2 + 4\cos\alpha) + 6J_3\}$$

b):

$$E(\alpha) = -\{(J_1 + J_3)(4\cos\alpha + 2\cos 2\alpha) + 6J_2\}$$




Ph. Kurz et al., J. Appl. Phys. **87**, 6101 (2000)

# HIGHER ORDER INTERACTIONS

## cyclic 4-spin term

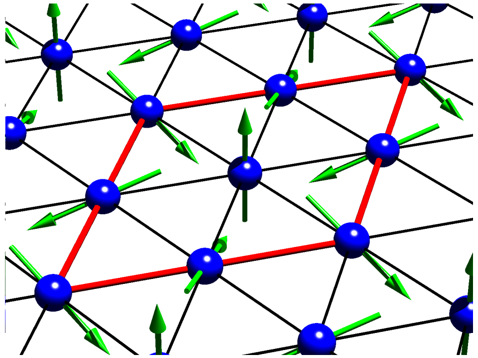
$$H_4 = -4K_1\left[(\mathbf{m}_1\cdot\mathbf{m}_2)(\mathbf{m}_3\cdot\mathbf{m}_4) + (\mathbf{m}_1\cdot\mathbf{m}_4)(\mathbf{m}_2\cdot\mathbf{m}_3) - (\mathbf{m}_1\cdot\mathbf{m}_3)(\mathbf{m}_2\cdot\mathbf{m}_4)\right]$$

this leads to

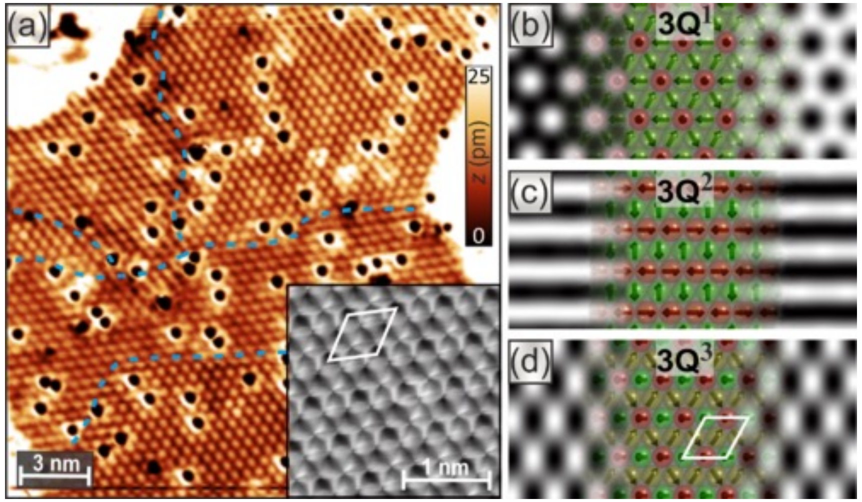
a): $E_4(\alpha) = -K_1(4\cos 2\alpha + 8)$

b): $E_4(\alpha) = -K_1(8\cos 3\alpha + 4)$

and can stabilize a new ground state (superposition of 3 states that are degenerate in the Heisenberg model)



3Q state predicted by
Ph. Kurz et al., Phys. Rev. Lett. **86**, 1106 (2001)
and confirmed in Mn/Re(0001):
J. Spethmann et al.
Phys. Rev. Lett. **124**, 227203 (2020)






JÜLICH Forschungszentrum

# TENSORIAL SPIN INTERACTIONS

## back to bilinear terms

- Interactions between two spins: $\mathbf{m}_i \underline{J}_{ij} \mathbf{m}_j$

| on-site | | inter-site | | |
|---|---|---|---|---|
| $\mathbf{m}_i J_{ii} \mathbf{m}_i$ | $\mathbf{m}_i \underline{J}_{ii} \mathbf{m}_i$ | $\mathbf{m}_i J_{ij} \mathbf{m}_j$ | $\mathbf{m}_i \underline{J}^S_{ij} \mathbf{m}_j$ | $\mathbf{m}_i \underline{J}^A_{ij} \mathbf{m}_j$ |
| scalar | traceless sym. | scalar | traceless sym. | antisymmetric |
| Stoner magnetism | magnetic anisotropy | Heisenberg interaction | (pseudo)-dipolar interaction | Dzyaloshinskii Moriya int. |

non-relativistic effects

$$\underline{J}^A_{ij} = \begin{pmatrix} 0 & D_z & -D_y \\ -D_z & 0 & D_x \\ D_y & -D_x & 0 \end{pmatrix}$$

leads to $\mathbf{D}_{ij} \cdot (\mathbf{m}_i \times \mathbf{m}_j)$

$$\underline{J}^A_{ij} \propto \left(\frac{\Delta g}{g}\right) J \quad ; \quad \underline{J}^S_{ij} \propto \left(\frac{\Delta g}{g}\right)^2 J$$

[T. Moriya, Phys. Rev. **120**, 91 (1960)]



JÜLICH Forschungszentrum

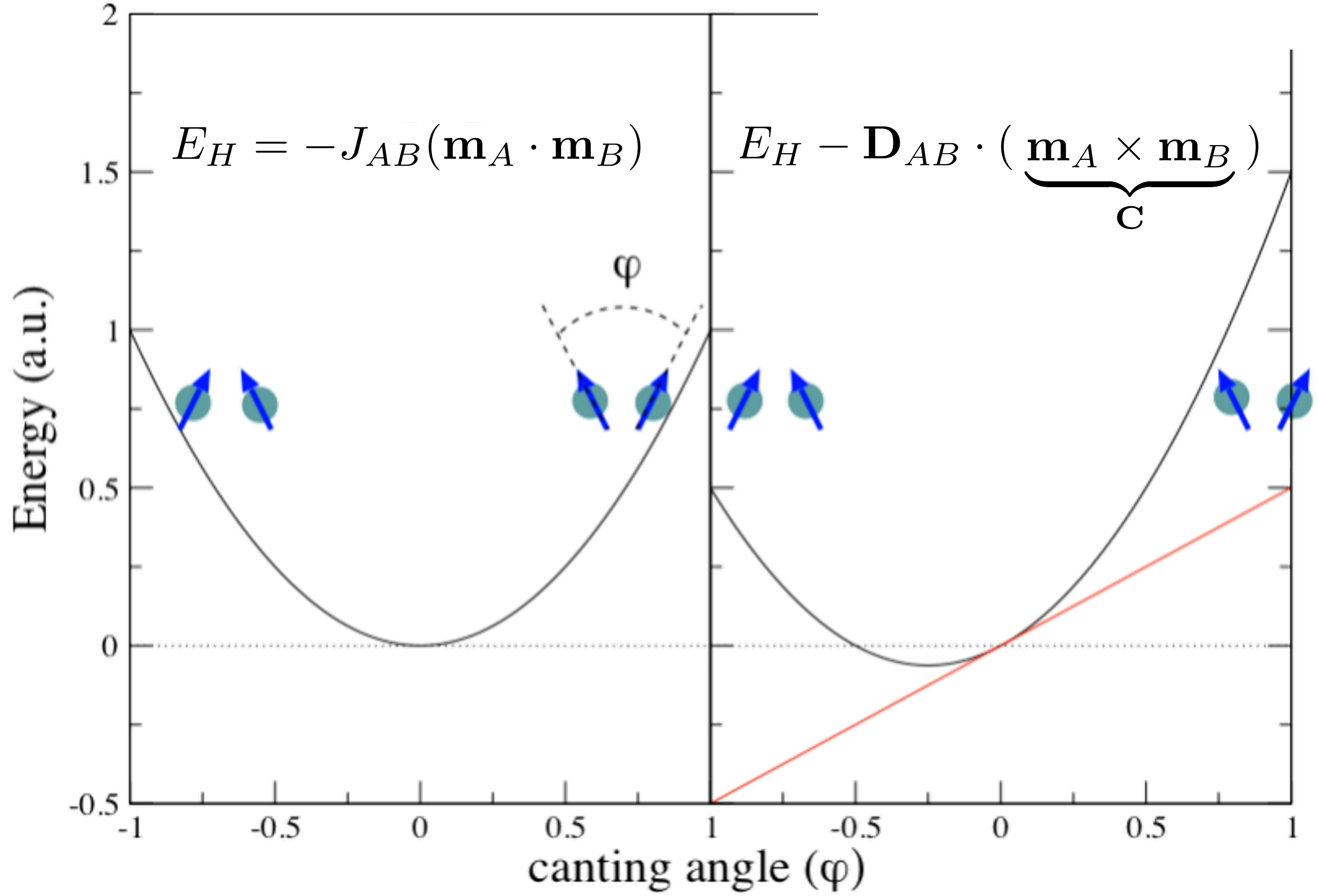
# TENSORIAL SPIN INTERACTIONS

## Antisymmetric parts: Dzyaloshinskii-Moriya Interaction (DMI)

$$\underline{J}_{ij}^{A} = \begin{pmatrix} 0 & D_z & -D_y \\ -D_z & 0 & D_x \\ D_y & -D_x & 0 \end{pmatrix}$$

leads to $\mathbf{D}_{ij} \cdot (\mathbf{m}_i \times \mathbf{m}_j)$

- requires spin-orbit interaction
- only for certain symmetries (Moriya rules)
- stabilizes canted spin-structures with preferred winding (**C**)
- can lead to chiral spin structures



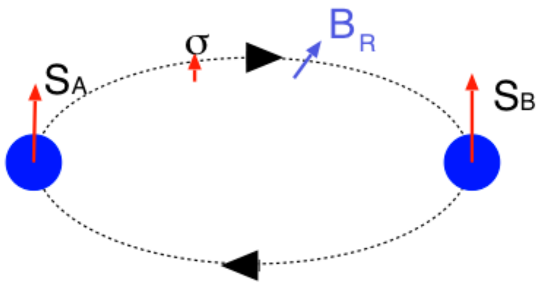


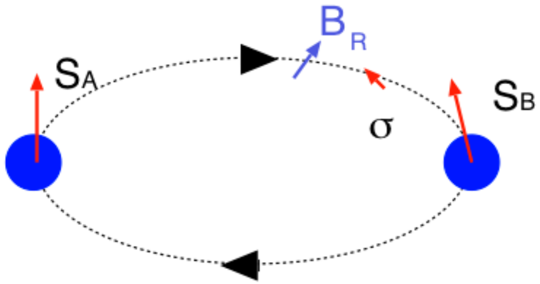

JÜLICH Forschungszentrum

# DZYALOSHINSKII-MORIYA INTERACTION

**first-order relativistic effect:**

$$H = -\sum_{i,j} J_{ij}\mathbf{m}_i \cdot \mathbf{m}_j + \sum_{i,j} \mathbf{D}_{ij} \cdot (\mathbf{m}_i \times \mathbf{m}_j)$$




$$E \propto (\mathbf{S}_A \cdot \boldsymbol{\sigma})\mathcal{G}_{A\to B}(\mathbf{S}_B \cdot \boldsymbol{\sigma})\mathcal{G}_{B\to A} \quad ; \quad \mathcal{G}_{A\to B} \approx \mathcal{G}_0 + \mathcal{G}_0 H_{\mathrm{SOC}}\mathcal{G}_0$$

$$H_{\mathrm{SOC}} = 0 : \quad E \propto \mathrm{Tr}_\sigma(\mathbf{S}_A \cdot \boldsymbol{\sigma})\mathcal{G}_0(\mathbf{S}_B \cdot \boldsymbol{\sigma})\mathcal{G}_0 = \frac{1}{2}J_{AB}\mathbf{S}_A \cdot \mathbf{S}_B$$

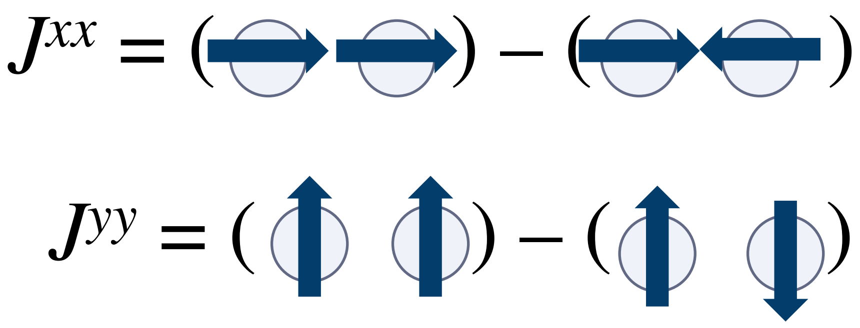
$$H_{\mathrm{SOC}} = \mathbf{B}_{\mathrm{eff}} \cdot \boldsymbol{\sigma} : \quad E_{DM} \propto \mathrm{Tr}_\sigma(\mathbf{S}_A \cdot \boldsymbol{\sigma})(\mathbf{B}_{\mathrm{eff}} \cdot \boldsymbol{\sigma})(\mathbf{S}_B \cdot \boldsymbol{\sigma})\mathcal{G}_0 \propto \mathbf{B}_{\mathrm{eff}} \cdot (\mathbf{S}_A \times \mathbf{S}_B)$$

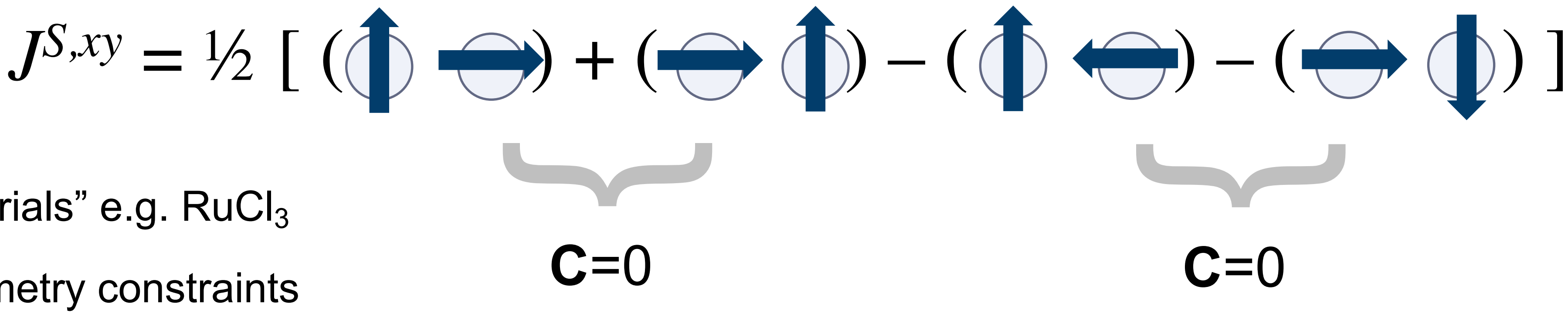
D. A. Smith, J. Magn. Magn. Mater. **1,** 214 (1976)

# TENSORIAL SPIN INTERACTIONS

## Symmetric parts

$$\underline{J}_{ij}^{S} = \begin{pmatrix} J_{ij}^{xx} - J/3 & \frac{1}{2}(J_{ij}^{xy} + J_{ij}^{yx}) & \frac{1}{2}(J_{ij}^{xz} + J_{ij}^{zx}) \\ \frac{1}{2}(J_{ij}^{xy} + J_{ij}^{yx}) & J_{ij}^{yy} - J/3 & \frac{1}{2}(J_{ij}^{yz} + J_{ij}^{zy}) \\ \frac{1}{2}(J_{ij}^{xz} + J_{ij}^{zx}) & \frac{1}{2}(J_{ij}^{yz} + J_{ij}^{zy}) & J_{ij}^{zz} - J/3 \end{pmatrix}$$




- important for "Kitaev materials" e.g. $RuCl_3$
- often suppressed by symmetry constraints
- second order in spin-orbit coupling

# DFT TOOLBOX (1)

## basic magnetic quantities

- single-particle wavefunction: $\phi(\mathbf{r}) = \begin{pmatrix} \phi^{\uparrow}(\mathbf{r}) \\ \phi^{\downarrow}(\mathbf{r}) \end{pmatrix}$
- spin density: $\mathrm{s}(\mathbf{r}) = \langle \boldsymbol{\phi}(\mathbf{r}) | \underline{\boldsymbol{\sigma}} | \boldsymbol{\phi}(\mathbf{r}) \rangle \quad ; \quad \underline{\sigma}_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \quad \underline{\sigma}_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \quad \underline{\sigma}_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$
- magnetization density: $\mathbf{m}(\mathbf{r}) = -\mu_{\mathrm{B}} g \langle \boldsymbol{\phi}(\mathbf{r}) | \underline{\mathbf{S}} | \boldsymbol{\phi}(\mathbf{r}) \rangle \quad \text{with} \quad \underline{\mathbf{S}} = \frac{1}{2} \underline{\boldsymbol{\sigma}} \quad \text{and} \quad g = 2$
- charge density: $\rho(\mathbf{r}) = e \langle \boldsymbol{\phi}(\mathbf{r}) | \underline{I} | \boldsymbol{\phi}(\mathbf{r}) \rangle$ particle density: $n(\mathbf{r}) = \langle \boldsymbol{\phi}(\mathbf{r}) | \underline{I} | \boldsymbol{\phi}(\mathbf{r}) \rangle$

density matrix: $\underline{n}(\mathbf{r}) = \frac{1}{2} \left( n(\mathbf{r}) \underline{I} + \mathbf{s}(\mathbf{r}) \cdot \underline{\boldsymbol{\sigma}} \right) = \frac{1}{2} \begin{pmatrix} n + s_z & s_x - i s_y \\ s_x + i s_y & n - s_z \end{pmatrix}$

potential matrix: $\underline{V}(\mathbf{r}) = v(\mathbf{r}) \underline{I} - \mu_{\mathrm{B}} \mathbf{B}(\mathbf{r}) \cdot \underline{\boldsymbol{\sigma}}$

# DFT TOOLBOX (1A)

## von-Barth Hedin formulation

- assume some spin-dependent external potential $\underline{V}$

$$E[\underline{n}] = T_0 + V_{\mathrm{Coul}} + E_{\mathrm{xc}} + \sum_{\sigma\sigma'} \int v_{\sigma\sigma'}(\mathbf{r}) n_{\sigma'\sigma}(\mathbf{r}) d\mathbf{r}$$

- ground state wavefunction uniquely determined by $\underline{n}(\mathbf{r})$
- $E$ is stationary w.r.t. variations of $\underline{n}(\mathbf{r})$ when $N = \sum_{\sigma} \int n_{\sigma\sigma}(\mathbf{r}) d\mathbf{r}$ is conserved.

This leads to the following Kohn-Sham equation:

$$\sum_{\sigma'} \left[ \left( -\frac{\hbar^2}{2m} \nabla^2 + \sum_{\sigma''} \int \frac{n_{\sigma''\sigma''}(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r}' \right) \delta_{\sigma\sigma'} + \frac{\delta E_{\mathrm{xc}}}{\delta n_{\sigma\sigma'}(\mathbf{r})} + v_{\sigma\sigma'}(\mathbf{r}) \right] \phi_{\sigma'}(\mathbf{r}) = \varepsilon_{\sigma} \phi_{\sigma}(\mathbf{r})$$

usually, the diagonal part of some LDA or GGA is used here

SOC can be included here



JÜLICH Forschungszentrum

# DFT TOOLBOX (2)

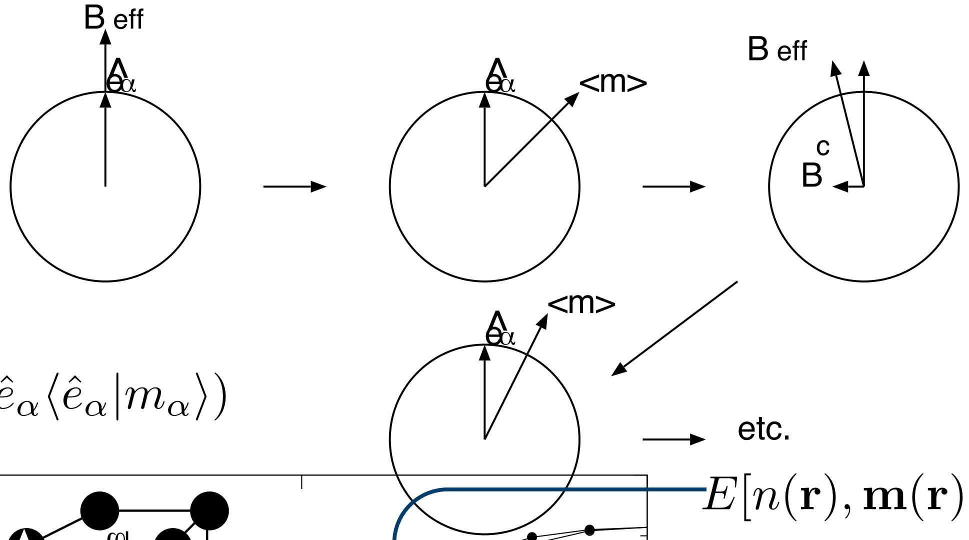
## constraints

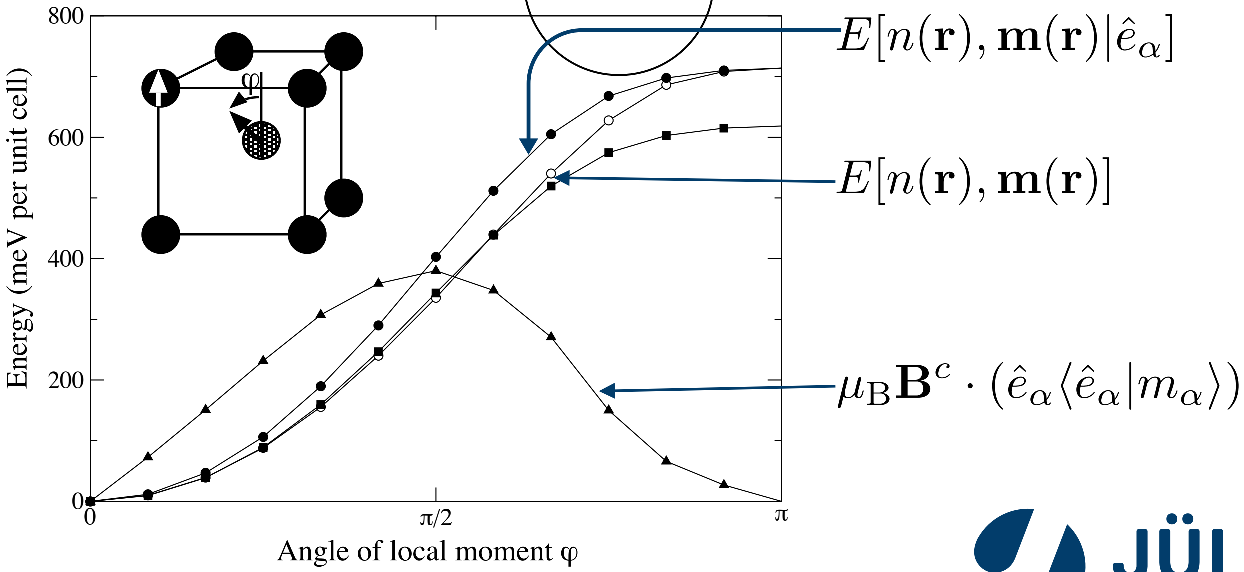
- local magnetic moment at atom $\alpha$ should point in direction $\hat{e}_\alpha$
- output magnetization $\langle m \rangle$ deviates usually from desired direction
- introduce constraint in energy functional

$$E[n(\mathbf{r}), \mathbf{m}(\mathbf{r})|\hat{e}_\alpha] = E[n(\mathbf{r}), \mathbf{m}(\mathbf{r})] + \mu_{\mathrm{B}} \mathbf{B}^c \cdot (\langle m_\alpha \rangle - \hat{e}_\alpha \langle \hat{e}_\alpha | m_\alpha \rangle)$$

- determine $\mathbf{B}^c$ self-consistently

[Kurz et al., Phys. Rev. B 69 024415 (2004)]





JÜLICH Forschungszentrum

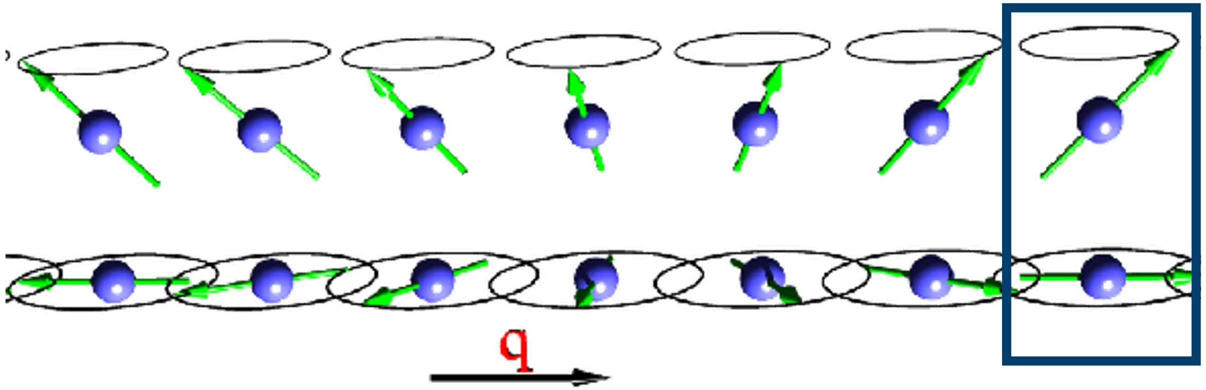
# DFT TOOLBOX (3)

## spin-spirals and the generalized Bloch theorem

Bloch theorem: Translation $\mathbf{R}_n$ : $T_n\psi_{\mathbf{k}}(\mathbf{r}) = \psi_{\mathbf{k}}(\mathbf{r}+\mathbf{R}_n) = e^{i\mathbf{k}\cdot\mathbf{R}_n}\psi_{\mathbf{k}}(\mathbf{r})$ when $\psi_{\mathbf{k}}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}}u(\mathbf{r})$

generalized Bloch theorem: add spin rotation: $U_\varphi = \begin{pmatrix} e^{-i\varphi/2} & 0 \\ 0 & e^{-i\varphi/2} \end{pmatrix}$ ; $\varphi = \mathbf{q}\cdot\mathbf{R}_n$

$$\mathcal{T}_n\boldsymbol{\psi}_{\mathbf{k}}(\mathbf{r}) = U_\varphi\boldsymbol{\psi}_{\mathbf{k}}(\mathbf{r}+\mathbf{R}_n) = e^{i\mathbf{k}\cdot\mathbf{R}_n}\boldsymbol{\psi}_{\mathbf{k}}(\mathbf{r}) \quad \text{when} \quad \boldsymbol{\psi}_{\mathbf{k}}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}}\begin{pmatrix} e^{-i\frac{\mathbf{q}\cdot\mathbf{r}}{2}}u_\uparrow(\mathbf{r}) \\ e^{i\frac{\mathbf{q}\cdot\mathbf{r}}{2}}u_\downarrow(\mathbf{r}) \end{pmatrix}$$




Spin-spirals (w/o SOC) can be described in the chemical unit cell !

www.flapw.de

Fleur



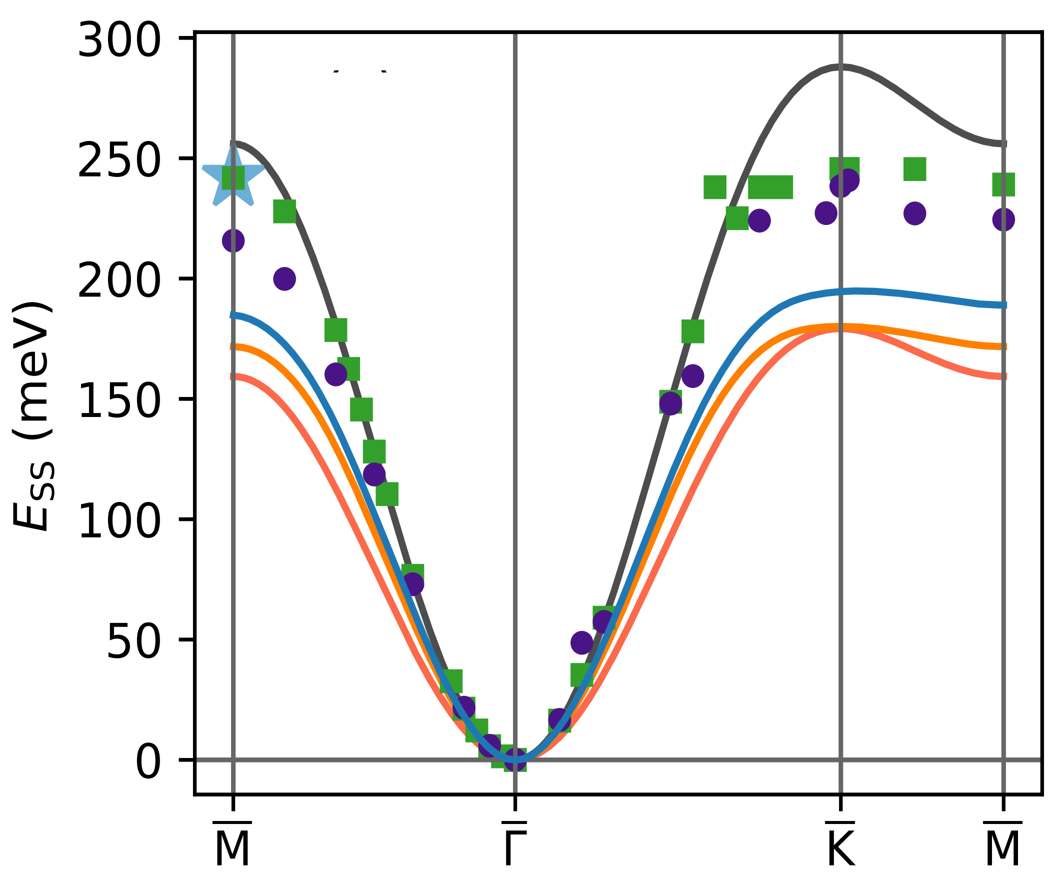
JÜLICH Forschungszentrum

# EXTRACTING INTERACTIONS FROM DFT

- spin-stiffness & DMI
- spin-spirals vs. infinitesimal rotations
- comparison to experiment

# EXAMPLE: Co/Pt(111)

## Exchange and Dzyaloshinskii-Moriya interaction (DMI)

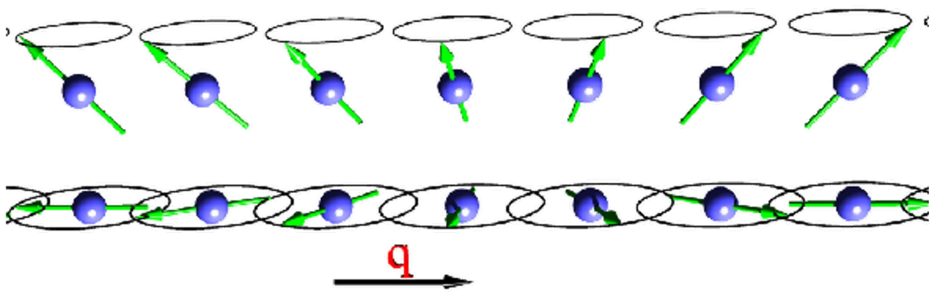



$$H = -\sum_{i,j} J_{ij}\,\mathbf{m}_i \cdot \mathbf{m}_j + \sum_{i,j} \mathbf{D}_{ij} \cdot (\mathbf{m}_i \times \mathbf{m}_j)$$

The general solutions are conical spirals with wavevector **q** and opening angle $\vartheta$: the magnetization of atom $\alpha$ in unit cell *n* is:

$$\hat{\mathbf{e}}_n^\alpha = \begin{pmatrix} \cos(\mathbf{q}\cdot(\mathbf{R}_n + \boldsymbol{\tau}^\alpha) + \zeta^\alpha)\sin(\vartheta^\alpha) \\ \sin(\mathbf{q}\cdot(\mathbf{R}_n + \boldsymbol{\tau}^\alpha) + \zeta^\alpha)\sin(\vartheta^\alpha) \\ \cos(\vartheta^\alpha) \end{pmatrix}$$

The energy of these spirals, $E_{SS}$, and the DMI contribution to it, $E_{DM}$, is calculated with DFT (FLAPW, FLEUR code).




$\vartheta = \pi/4$

$\vartheta = \pi/2$

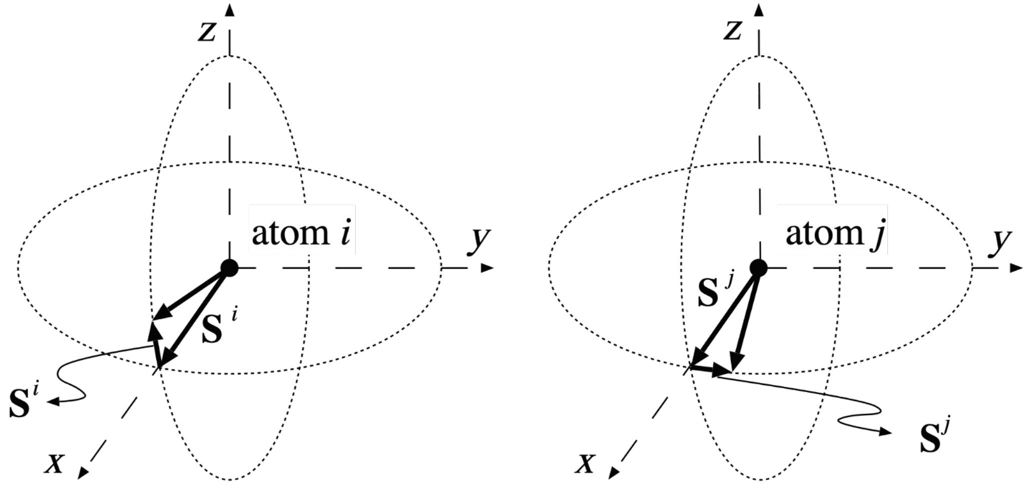


JÜLICH Forschungszentrum

# USING INFINITESIMAL ROTATIONS ...

$$H = \sum_{i<j} \mathbf{m}_i \underline{J}_{ij} \mathbf{m}_j = \sum_{i<j} m_i m_j \hat{\mathbf{e}}_i \underline{J}_{ij} \hat{\mathbf{e}}_j$$

e.g. $$J_{ij}^{xy} = \frac{\partial^2 H}{\partial x_i \partial y_j}$$




$$\underline{J}_{ij}^{A} = \begin{pmatrix} 0 & \frac{1}{2}(J_{ij}^{xy} - J_{ij}^{yx}) & \frac{1}{2}(J_{ij}^{xz} - J_{ij}^{zx}) \\ -\frac{1}{2}(J_{ij}^{xy} - J_{ij}^{yx}) & 0 & \frac{1}{2}(J_{ij}^{yz} - J_{ij}^{zy}) \\ -\frac{1}{2}(J_{ij}^{xz} - J_{ij}^{zx}) & -\frac{1}{2}(J_{ij}^{yz} - J_{ij}^{zy}) & 0 \end{pmatrix} = \begin{pmatrix} 0 & D_{ij}^{z} & -D_{ij}^{y} \\ -D_{ij}^{z} & 0 & D_{ij}^{x} \\ D_{ij}^{y} & -D_{ij}^{x} & 0 \end{pmatrix}$$

$$J_{ij}^{zy} = -\left(\frac{\partial^2 H}{\partial \theta_i \partial \phi_j}\right)_{\substack{\theta_{i,j}=\frac{\pi}{2} \\ \phi_{i,j}=0}}, \quad J_{ij}^{yz} = -\left(\frac{\partial^2 H}{\partial \phi_i \partial \theta_j}\right)_{\substack{\theta_{i,j}=\frac{\pi}{2} \\ \phi_{i,j}=0}}$$

Need to calculate total energy change due to infinitesimal rotations $\phi, \theta$

# … AND GREEN FUNCTIONS METHODS

## to calculate the DMI, e.g. with Korringa-Kohn-Rostoker (KKR) method

- We can calculate energy changes from changes of the DOS, n(E):

$$F = \int^{E_F} (E - E_F) n(E) dE = -\int^{E_F} N(E) dE \qquad ; \qquad \Delta N(E) = \frac{1}{\pi} \mathrm{Im} \mathrm{Tr} \ln \tau(E)$$

- where $\tau$ is the scattering path operator: $\tau(E) = [t^{-1}(E) - g(E)]^{-1}$
- Changes in the scattering matrix are given as: $t'^{-1}_i = \mathcal{R}(\delta\theta_i, \delta\phi_i)\, t_i^{-1}\, \mathcal{R}^\dagger(\delta\theta_i, \delta\phi_i).$

From the dependence of the scattering matrices on $\theta$,$\phi$ we get the DMI,
e.g. for the *x*-component of $\vec{D}$:

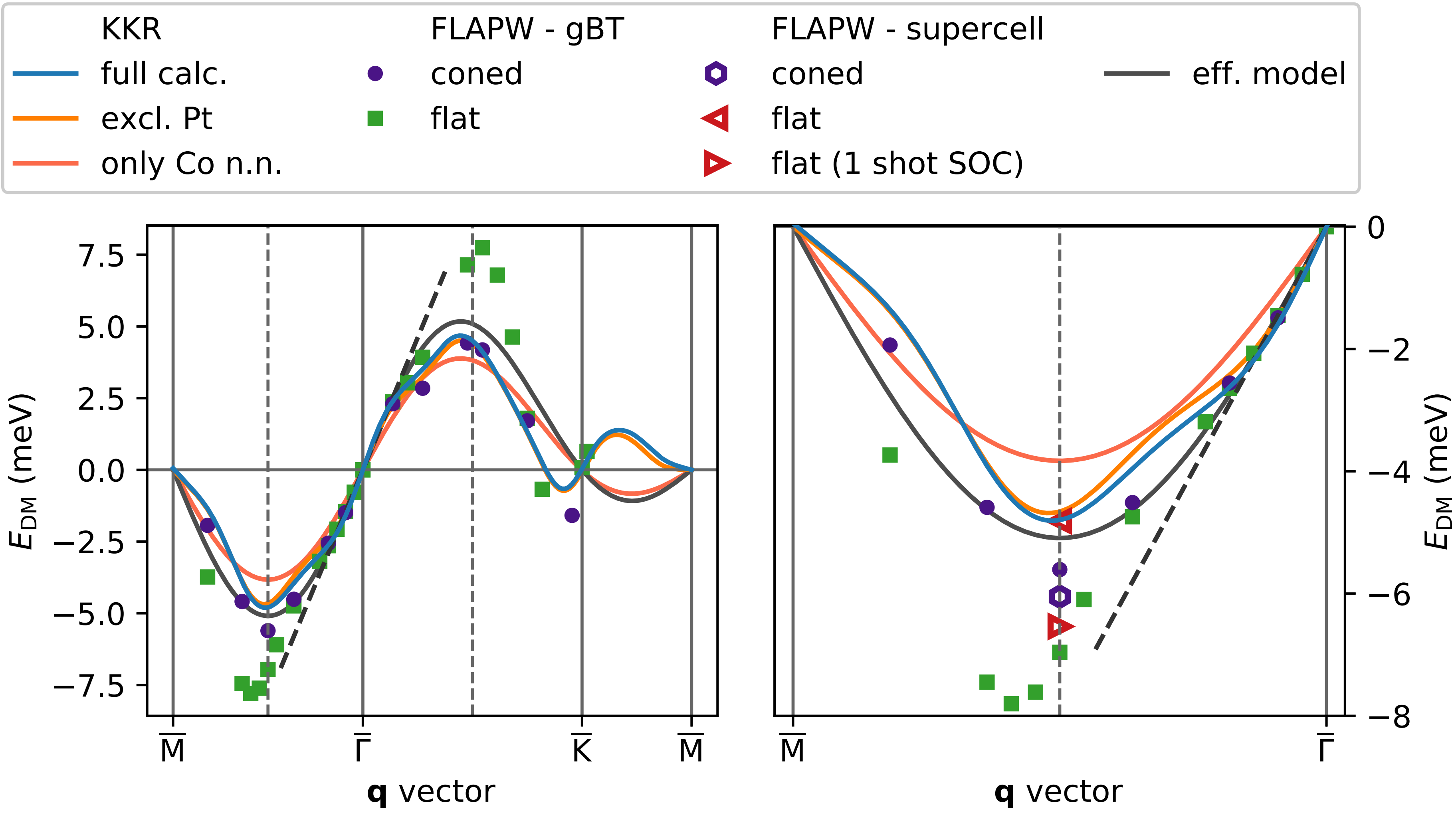
$$D^x_{ij} = -\frac{1}{2\pi} \mathrm{Im} \int^{E_F} dE\, \mathrm{Tr}_L \left[ t^{-1}_{i\,\phi}(E)\, \tau_{ij}(E)\, t^{-1}_{j\,\theta}(E)\, \tau_{ji}(E) - t^{-1}_{i\,\theta}(E)\, \tau_{ij}(E)\, t^{-1}_{j\,\phi}(E)\, \tau_{ji}(E) \right]_{\substack{\theta_{i,j}=\frac{\pi}{2} \\ \phi_{i,j}=0}}$$

Staring point is e.g. a ferromagnetic KKR calculation, SOC included in single shot.

Udvardi et al., Phys. Rev. B **68**, 104436 (2003) -- see also Psi-k Highlight No. 78 (2006)

# CALCULATING $E_{DM}$ BY DIFFERENT METHODS

## Co/Pt(111) spin-spiral with infinitesimal rotations (KKR), gen. Bloch theorem (gBT)




- inf. rotations: determine $\underline{J}_{ji}$ matrix and calculate $E_{DM}$
- gBT: calculate $E_{DM}$ and extract $J_{ij}$ and $D_{ij}$ (SOC in first order perturbation theory)
- supercell: self-consistent SOC for short range spin-spiral.

Zimmermann et al.,
Phys. Rev. B **99** 214426 (2019)



JÜLICH Forschungszentrum

# MAPPING OF THE ATOMISTIC SPIN MODEL

## to micromagnetic / effective parameters

$$H = -\sum_{i,j} J_{ij}\mathbf{m}_i \cdot \mathbf{m}_j + \sum_{i,j} \mathbf{D}_{ij} \cdot (\mathbf{m}_i \times \mathbf{m}_j)$$

Spin-stiffness tensor from spin-spiral calculation:

$$\frac{\mathcal{A}}{4\pi^2} = \frac{1}{2V_\Omega}\frac{\partial^2}{\partial \mathbf{q}^2} E_{\mathrm{SS}}(\mathrm{q})|_{\mathbf{q}_c} = \frac{1}{2V_\Omega}\sum_{i\geq 1} J_{0i}\mathbf{R}_i \otimes \mathbf{R}_i$$

$C_{3v}$ symmetry

$$A = \mathcal{A}_{11} = \mathcal{A}_{22}$$

Spiralization tensor from spin-spiral calculation:

$$\frac{\mathcal{D}}{2\pi} = \hat{\mathbf{e}}_{\mathrm{rot}} \frac{1}{V_\Omega}\frac{\partial}{\partial \mathbf{q}} E_{\mathrm{DM}}(\mathrm{q}, \hat{\mathbf{e}}_{\mathrm{rot}})|_{\mathbf{q}_c} = \frac{1}{V_\Omega}\sum_{i\geq 1} \mathbf{D}_{0i} \otimes \mathbf{R}_i$$

$$D = -\mathcal{D}_{12} = \mathcal{D}_{21}$$

Schweflinghaus et al., Phys. Rev. B **94**, 024403 (2016)

# MAPPING OF THE ATOMISTIC SPIN MODEL

## to micromagnetic / effective parameters

$$H = -\sum_{i,j} J_{ij}\mathbf{m}_i \cdot \mathbf{m}_j + \sum_{i,j} \mathbf{D}_{ij} \cdot (\mathbf{m}_i \times \mathbf{m}_j) + \sum_i K_i (\mathbf{m}_i^z)^2$$

Spin-stiffness tensor from spin-spiral calculation:

$$\frac{\mathcal{A}}{4\pi^2} = \frac{1}{2V_\Omega}\frac{\partial^2}{\partial \mathbf{q}^2} E_{\mathrm{SS}}(\mathrm{q})\big|_{\mathbf{q}_c} = \frac{1}{2V_\Omega}\sum_{i\geq 1} J_{0i}\mathbf{R}_i \otimes \mathbf{R}_i$$

$C_{3v}$ symmetry, **q** in *x*-direction:

$$A = \frac{1}{2V_\Omega}\sum_i J_{0i}(R_{0i}^x)^2 \qquad J_{\mathrm{eff}} = \frac{2}{3}\frac{V_\Omega}{a^2}A$$

Spiralization tensor from spin-spiral calculation:

$$\frac{\mathcal{D}}{2\pi} = \hat{\mathbf{e}}_{\mathrm{rot}}\frac{1}{V_\Omega}\frac{\partial}{\partial \mathbf{q}} E_{\mathrm{DM}}(\mathrm{q}, \hat{\mathbf{e}}_{\mathrm{rot}})\big|_{\mathbf{q}_c} = \frac{1}{V_\Omega}\sum_{i\geq 1} \mathbf{D}_{0i} \otimes \mathbf{R}_i$$

$$D = \frac{1}{V_\Omega}\sum_i D_{0i}^y R_{0i}^x \qquad D_{\mathrm{eff}} = \frac{1}{3}\frac{V_\Omega}{a}D$$

Schweflinghaus et al., Phys. Rev. B **94**, 024403 (2016)

# COMPARISON OF $J_{EFF}$ AND $D_{EFF}$

## infinitesimal rotations (KKR), generalized Bloch theorem (gBT) and supercells

### comparison with another KKR calculation

| | stacking position | $J_{\text{eff}}$ (meV) | $D_{\text{eff}}$ (meV) |
|---|---|---|---|
| KKR | Co(fcc) | 29.5 (27.9) | 1.75 (1.47) |
| | Co(hcp) | | |
| FLAPW-gBT (coned) | Co(fcc) | 31.7 | 1.43 |
| | Co(hcp) | | |
| FLAPW-gBT (flat) | Co(fcc) | 32.0 | 1.47 |
| Reference [16] | Co(fcc) | (27.2) | (1.43) |

[16] Simon, et al., Phys. Rev. B 97, 134405 (2018)

- fitting E(q) and extrapolating to q=0 [values in parenthesis] gives consistent $D_{\text{eff}}$.
- 10% difference in the spin-stiffness might be due to higher-order interactions.

### and with another supercell calculation

| | | $D$ (m/m$^2$) | $D_{\text{eff}}$ (meV) |
|---|---|---|---|
| coned | scSOC | 20.3 | 2.02 |
| flat | scSOC | 16.2 | 1.60 |
| | one-shot SOC | 21.9 | 2.18 |
| Reference [10] | | 19.0 | 2.17 |

[10] Yang et al., Phys. Rev. Lett. **115**, 267210 (2015)

- consistent values obtained by different DFT codes
- higher $D_{\text{eff}}$ (compared to left column) due to nearest-neighbor model
- significant effects from self-consistency in SOC

# COMPARISON TO EXPERIMENTAL VALUES

| A | Ref. |
|---|---|
| 30 pJ/m (bulk hcp Co) | K. Hüller, J. Magn. Magn. Mater. **61**, 347 (1986) |
| 21 pJ/m (10 nm Co film) | C. Eyrich et al., J. Appl. Phys. **111**, 07C919 (2012) |
| 27.5 pJ/m (< 1nm Co film) | O. Boulle et al., Nat. Nanotechnol. **11**, 449 (2016) |
| 41-44 pJ/m (1 ML Co/Pt(111), DFT calculations) | B. Zimmermann et al., Phys. Rev. B **99** 214426 (2019) |

finite temperature renormalization by $[m(T)/m(T=0)]^2$ gives a theoretical result of 29.7 pJ/m

# POSSIBLE REASONS FOR DISCREPANCIES

## numerical (e.g. truncation) or methodological (different models)

- geometrical (lattice parameters, relaxation etc.)
- cutoffs (k-point sampling, Fermi broadening, basis set)
- exchange correlation functional (LDA, GGA, hybrid functionals, …)
- method (spin-spirals, infinitesimal rotations, Berry phase, spin current / susc.)
- initial magnetic configuration (ferromagnet, DLM state…)
- mapping on spin model (# nearest neighbors, types of interactions)
- …

# HIGHER-ORDER INTERACTIONS

- biquadratic interactions
- 3-site 4-spin interactions
- chiral-chiral and spin-chiral interactions

# EXCHANGE INTERACTIONS & HUBBARD MODEL

**nonrelativistic spin Hamiltonian:**

2 sites, 1 orbital, S= ½ :

$$H = -\sum_{i<j,\sigma} t_{ij}(c^\dagger_{i,\sigma} c_{j,\sigma} + \text{h.c.}) + \sum_i U_i n_{i,\uparrow} n_{i,\downarrow} + \ldots$$

downfolding →

$$\frac{4t^2}{U} \sum_{i\neq j} c^\dagger_{i,\uparrow} c_{i,\downarrow} c^\dagger_{j,\downarrow} c_{j,\uparrow} - n_{i,\uparrow} n_{j,\downarrow}$$

spin Hamiltonian →

$$H_{2o} = \frac{4t^2}{U} \sum_{i<j} (\mathbf{S}_i \cdot \mathbf{S}_j - \frac{n_i n_j}{4}) \qquad H_{4o} = -\frac{4t^2}{U^2} H_{2o}$$

Heisenberg model

4 sites, S= ½ :

terms like $c^\dagger_{4,\downarrow} c^\dagger_{3,\uparrow} c^\dagger_{2,\uparrow} c^\dagger_{1,\downarrow} c_{1,\uparrow} c_{2,\downarrow} c_{3,\downarrow} c_{4,\uparrow}$ lead to

$$H_{4o} = \frac{10t^4}{U^3} \sum_{ijkl} (\mathbf{S}_i \cdot \mathbf{S}_j - \frac{n_i n_j}{4})(\mathbf{S}_k \cdot \mathbf{S}_l - \frac{n_k n_l}{4})$$

4-spin interaction

Model Hamiltonian:

$$H = -{\sum_{ij}}' J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - {\sum_{ijkl}}' K_{ijkl} \left[ (\mathbf{S}_i \cdot \mathbf{S}_j)(\mathbf{S}_k \cdot \mathbf{S}_l) + (\mathbf{S}_j \cdot \mathbf{S}_k)(\mathbf{S}_l \cdot \mathbf{S}_i) - (\mathbf{S}_i \cdot \mathbf{S}_k)(\mathbf{S}_j \cdot \mathbf{S}_l) \right] + \ldots$$

# SPIN INTERACTIONS FOR S > ½

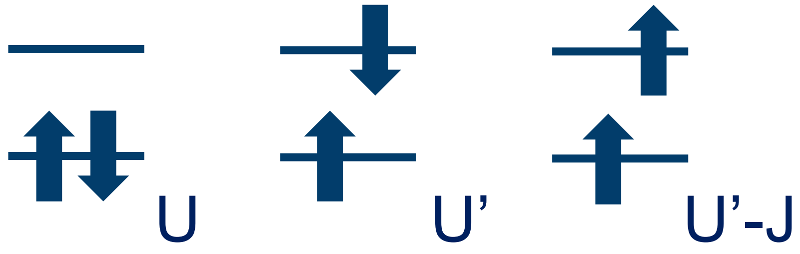
## Hubbard model, downfolding, 4th order perturbation theory

2 sites, 2 orbitals, S=1:

terms like $c^\dagger_{2,2,\downarrow}c^\dagger_{2,1,\uparrow}c^\dagger_{1,2,\uparrow}c^\dagger_{1,1,\downarrow}c_{1,1,\uparrow}c_{1,2,\downarrow}c_{2,1,\downarrow}c_{2,2,\uparrow}$

spin Hamiltonian →

$$H_{4o} = -\frac{20t^4}{(U+J_\mathrm{H})^3}\sum_{ij}(\mathbf{S}_i\cdot\mathbf{S}_j - \frac{n_i n_j}{4})^2$$




… biquadratic interaction

3 sites, 2 orbitals, S=1:

terms like $c^\dagger_{3,2,\downarrow}c^\dagger_{2,2,\uparrow}c^\dagger_{2,1,\uparrow}c^\dagger_{1,1,\downarrow}c_{1,1,\uparrow}c_{2,1,\downarrow}c_{2,2,\downarrow}c_{3,2,\uparrow}n_{3,1,\uparrow}n_{1,2,\downarrow}$

$$H_{4o} = -\frac{40t^4}{(U+J_\mathrm{H})^3}\sum_{ijk}(\mathbf{S}_i\cdot\mathbf{S}_j - \frac{n_i n_j}{4})(\mathbf{S}_j\cdot\mathbf{S}_k - \frac{n_j n_k}{4})$$

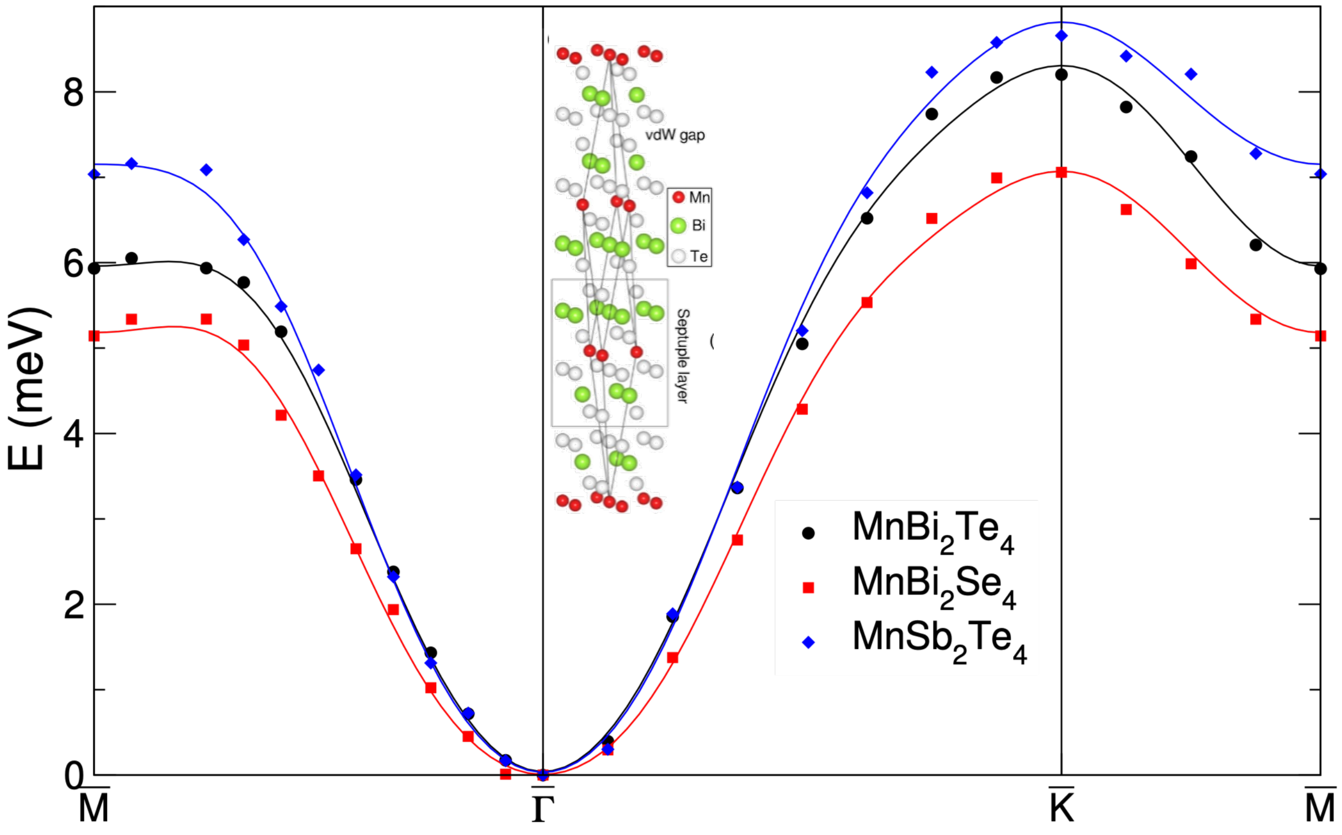
3-site 4-spin interaction

$$H = -\sum_{ij}{}' J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j - \sum_{ijkl}{}' K_{ijkl}\left[(\mathbf{S}_i\cdot\mathbf{S}_j)(\mathbf{S}_k\cdot\mathbf{S}_l) + ...\right] - \sum_{ij}{}' B_{ij}(\mathbf{S}_i\cdot\mathbf{S}_j)^2 - \sum_{ijk}{}' Y_{ijk}\left[(\mathbf{S}_i\cdot\mathbf{S}_j)(\mathbf{S}_j\cdot\mathbf{S}_k) + ...\right]$$

Hoffmann and Blügel, Phys. Rev. B **101**, 024418 (2020)

# EXAMPLE : MnBi$_2$Te$_4$ THIN FILMS

**extracting J$_{ij}$s from spin-spiral calculations**




$$E(q) = -S^2 \left(2J_2 + 2J_6 + \cos(\pi q)\left[4J_1 + 4J_5\right] + \cos(2\pi q)\left[2J_1 + 4J_3\right] + \cos(3\pi q)\left[4J_2 + 4J_5\right] + \cos(4\pi q)\left[2J_3 + 4J_4\right] + \ldots\right)$$

fit along $\Gamma$–K line (meV)

| | $J_1$ | $J_2$ | $J_3$ | $J_4$ | $J_5$ |
|---|---|---|---|---|---|
| MBT | -1.56 | -0.03 | -0.35 | 0.03 | 0.04 |
| MBS | -1.39 | -0.05 | -0.26 | 0.04 | 0.06 |
| MST | -1.79 | -0.10 | -0.21 | 0.02 | 0.06 |

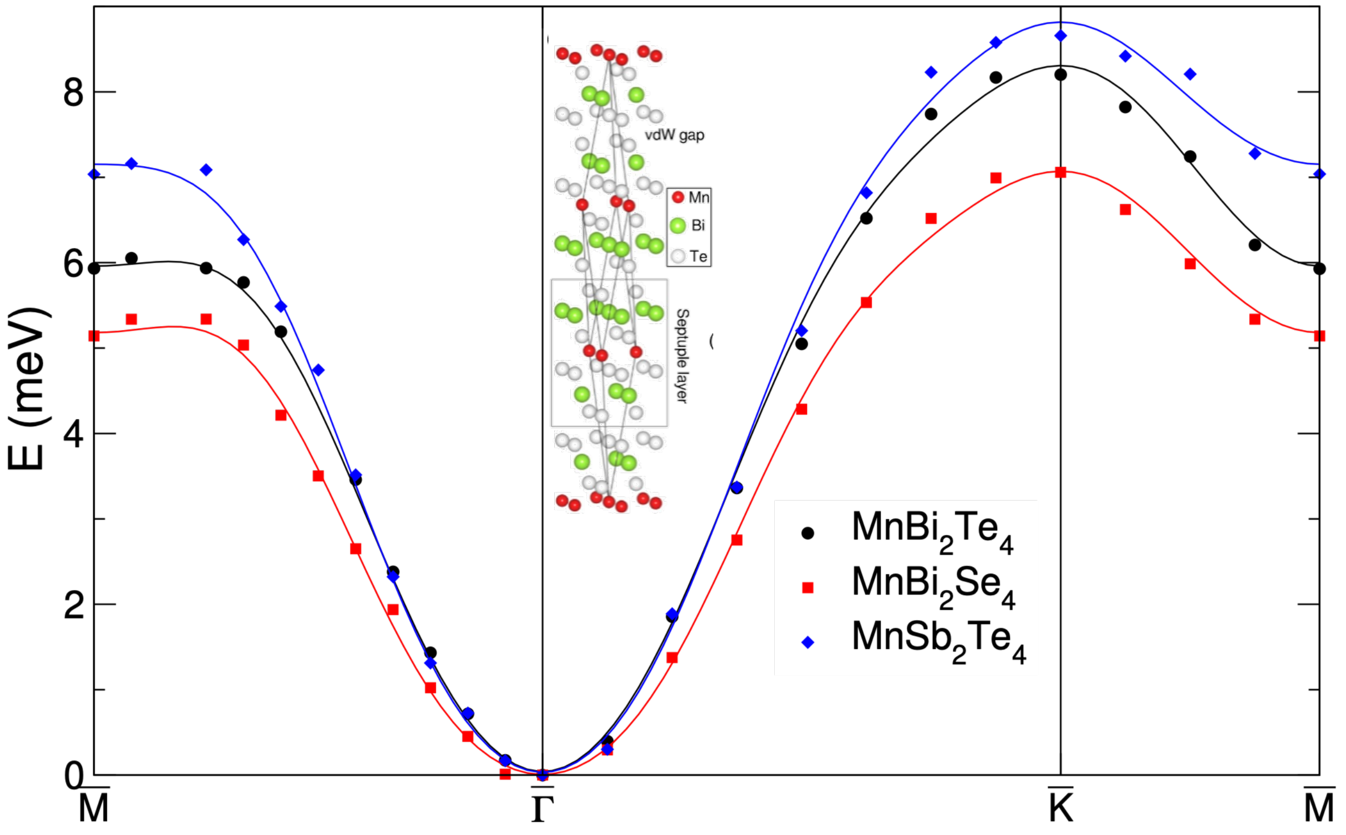


JÜLICH Forschungszentrum

# EXAMPLE : $MnBi_2Te_4$ THIN FILMS

## extracting $J_{ij}$s from spin-spiral calculations

$$E(q) = -S^2 \big(2J_2 + 2J_6 + \cos(\pi q)\,[4J_1 + 4J_5] + \cos(2\pi q)\,[2J_1 + 2(2J_3 + S^2 B_1)] + \cos(3\pi q)\,[4J_2 + 4J_5] + \cos(4\pi q)\,[2J_3 + S^2 B_1 + 4J_4] + \ldots\big)$$



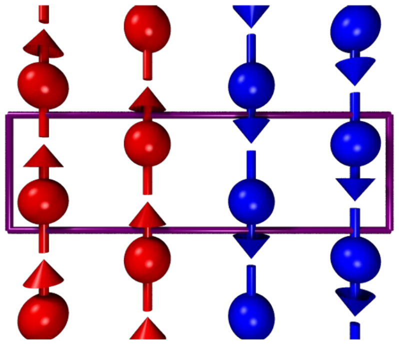

fit along Γ–K line (meV)

| | $J_1$ | $J_2$ | $J_3+B_1/2$ | 4 | $J_5$ |
|---|---|---|---|---|---|
| MBT | -1.56 | -0.03 | -0.35 | 0.03 | 0.04 |
| MBS | -1.39 | -0.05 | -0.26 | 0.04 | 0.06 |
| MST | -1.79 | -0.10 | -0.21 | 0.02 | 0.06 |

# $MnBi_2Te_4$ Thin Films: Biquadratic Interaction

## determining higher-order interactions

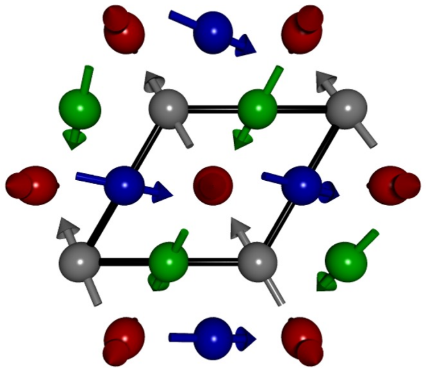
superpositions of spin-spirals (1Q) lead to multi-Q states that are degenerate in the Heisenberg model:

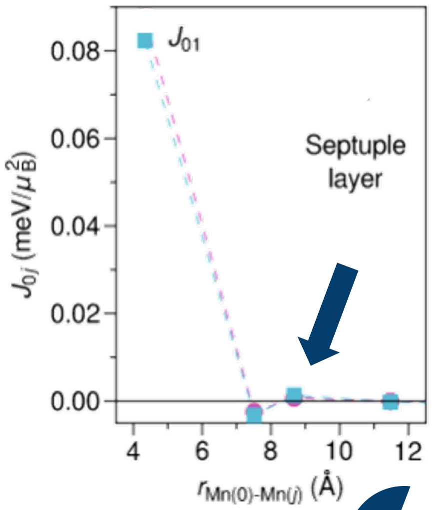


$$E_{2Q} - E_{1Q} = -4S^4(2K_1 - B_1) \qquad Q = \overline{\Gamma M}/2$$

$$E_{3Q} - E_{1Q} = -\frac{16}{3}S^4(2K_1 + B_1) \qquad Q = \overline{M}$$

$$S^4K_1 = -0.04 \text{ meV} \qquad S^4B_1 = -0.36 \text{ meV}$$






M.Otrokov et al., Phys. Rev. Lett. **122**,107202 (2019)

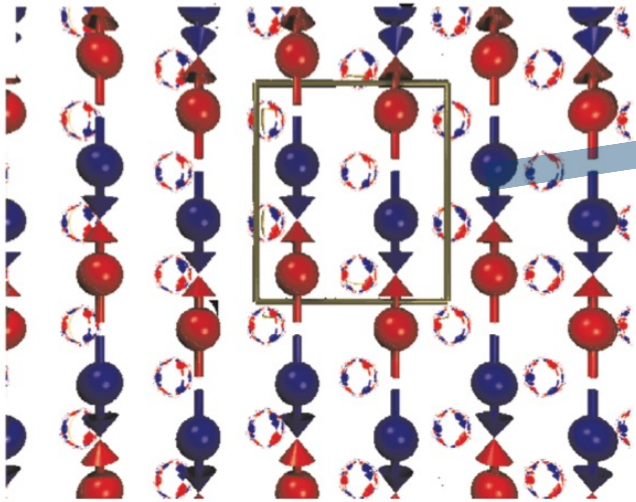


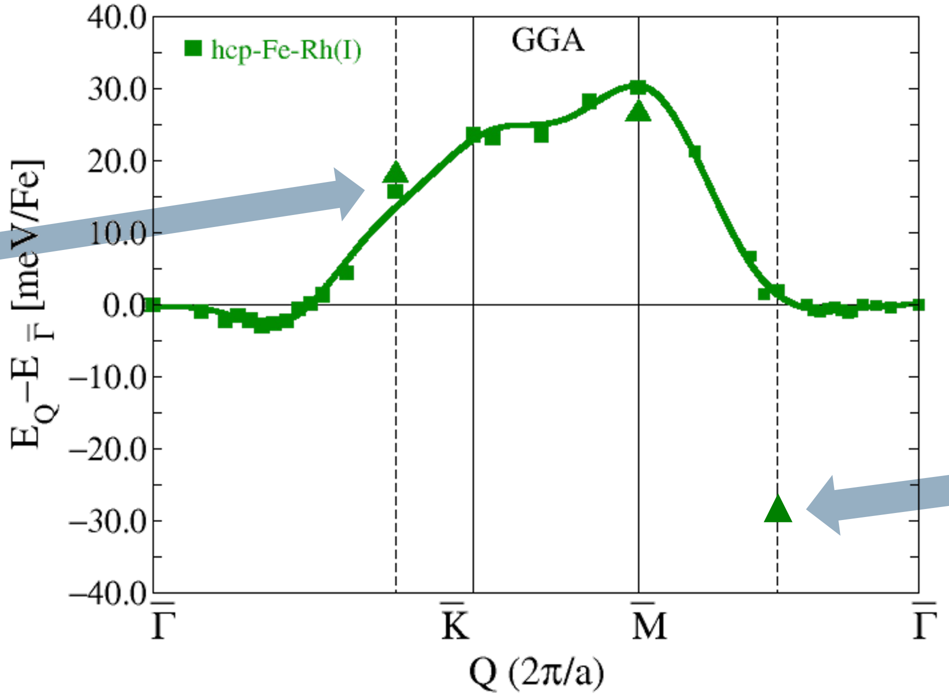
JÜLICH Forschungszentrum

# Fe/Rh(111)

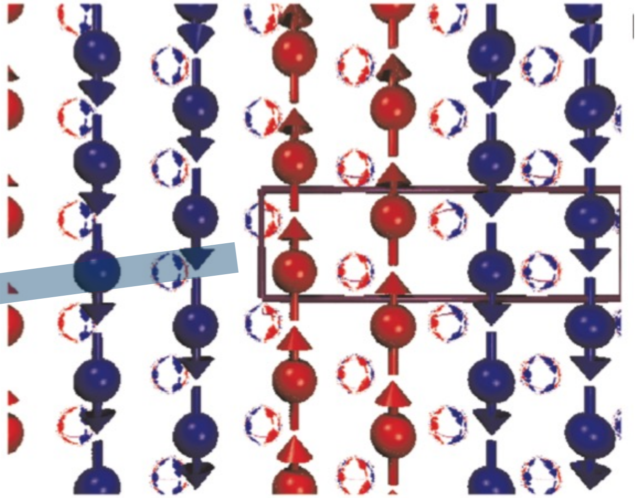
## magnetic ground state determined by higher-order interactions

$$E_{2Q} - E_{1Q} = -4S^4(2K_1 - B_1)$$






$$E_{2Q} - E_{1Q} = -4S^4(2K_1 - B_1)$$



A. Al-Zubi et al., Phys. Status Solidi B **248,** 2242 (2011)

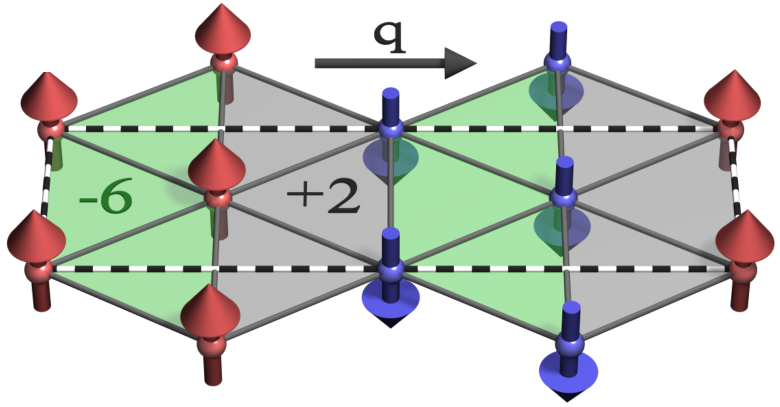


JÜLICH
Forschungszentrum

# 3 SITE – 4 SPIN INTERACTION

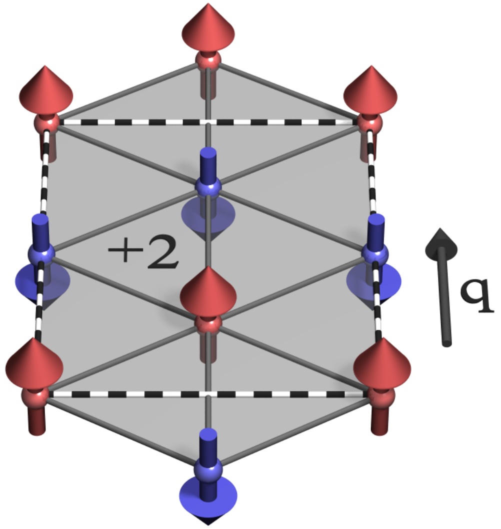
## and biquadratic term determine the ground state in Fe/Rh(111)

$$H_3 = -2 \sum_{<ijk>} Y_{ijk} \left[ (\mathbf{S}_i \cdot \mathbf{S}_j)(\mathbf{S}_j \cdot \mathbf{S}_k) + (\mathbf{S}_j \cdot \mathbf{S}_k)(\mathbf{S}_k \cdot \mathbf{S}_i) + (\mathbf{S}_k \cdot \mathbf{S}_i)(\mathbf{S}_i \cdot \mathbf{S}_j) \right]$$

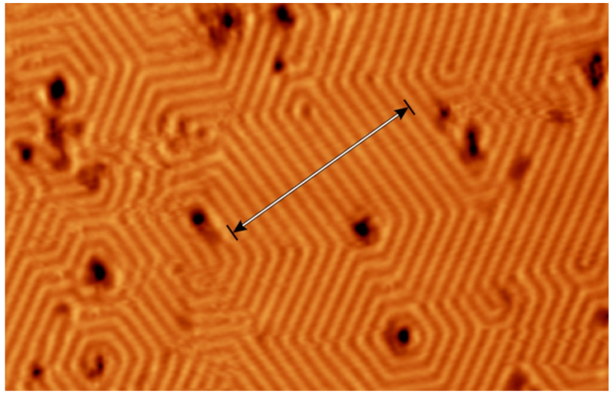



$$E_{2Q,\frac{\overline{\mathrm{M}}}{2}} - E_{\frac{\overline{\mathrm{M}}}{2}} = 4(2K_1 - B_1 - Y_1)$$

$$E_{2Q,\frac{3\overline{\mathrm{K}}}{4}} - E_{\frac{3\overline{\mathrm{K}}}{4}} = 4(2K_1 - B_1 + Y_1)$$



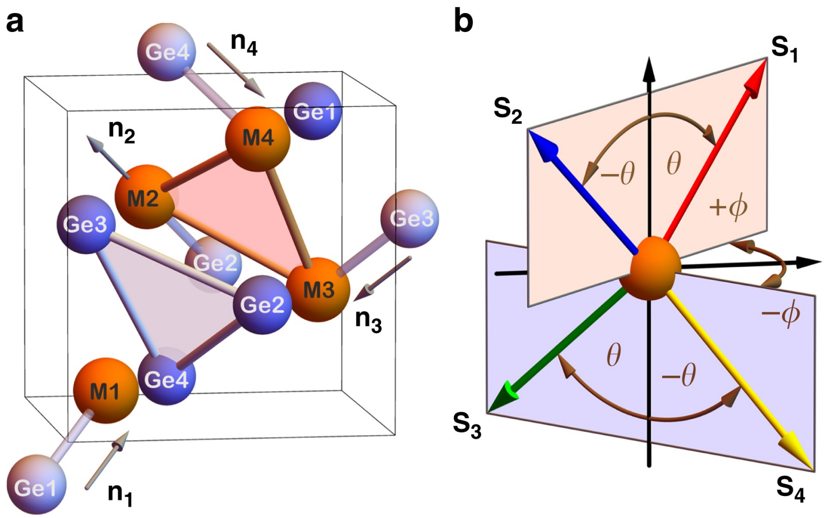

| | |
|---|---|
| $B_1$ | 3.39 meV |
| $Y_1$ | 4.00 meV |
| $K_1$ | 0.07 meV |

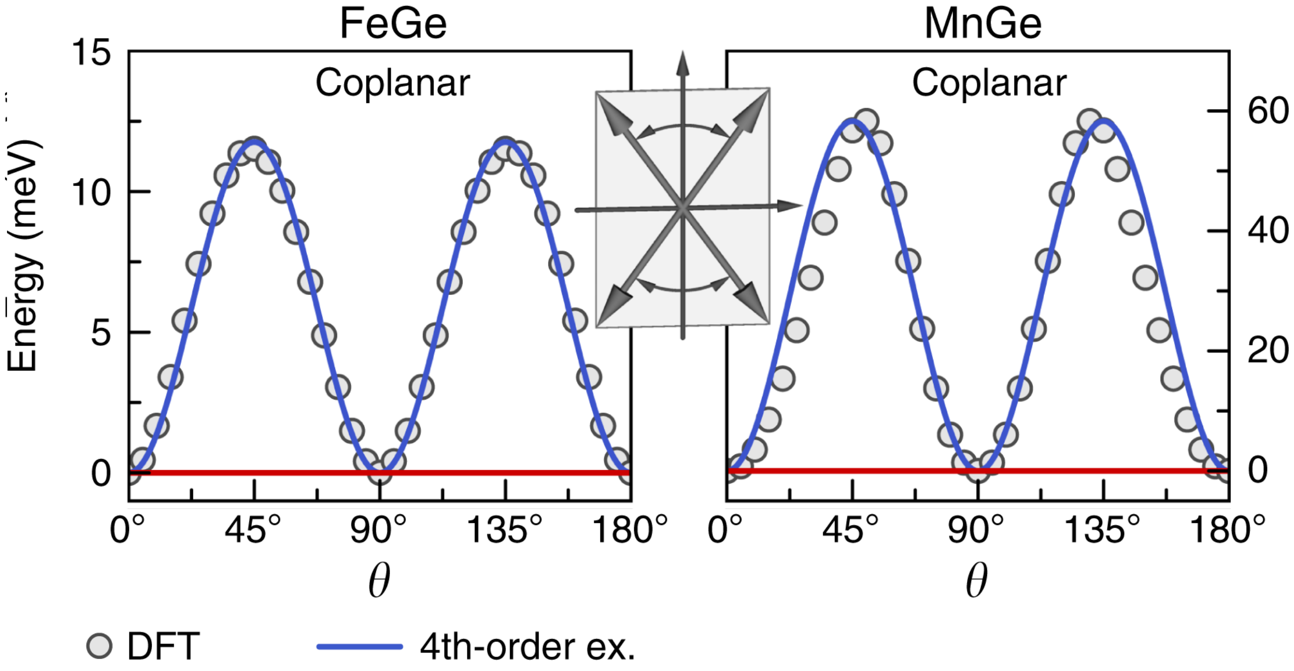


Krönlein et al., Phys. Rev. Lett. **120**, 207202 (2018)

# FeGe / MnGe - MORE EXOTIC INTERACTIONS

## determine higher order interactions selecting special spin configurations




varying θ and ϕ gives no contribution from Heisenberg terms

$$H = -\sum_{ij}{}' J_{ij}\mathbf{S}_i \cdot \mathbf{S}_j - \sum_{ijkl}{}' K_{ijkl}\left[(\mathbf{S}_i \cdot \mathbf{S}_j)(\mathbf{S}_k \cdot \mathbf{S}_l) + (\mathbf{S}_j \cdot \mathbf{S}_k)(\mathbf{S}_l \cdot \mathbf{S}_i) - (\mathbf{S}_i \cdot \mathbf{S}_k)(\mathbf{S}_j \cdot \mathbf{S}_l)\right] + \ldots$$

Grytsiuk et al., Nature Commun. **11**, 511 (2020)

# TOPOLOGICAL ORBITAL MOMENTS

## in non-collinear magnetic structures

non-coplanar spin structures with finite chirality:

$$\chi_{ijk} = \mathbf{S}_i \cdot (\mathbf{S}_j \times \mathbf{S}_k)$$

gives rise to an emergent magnetic field:

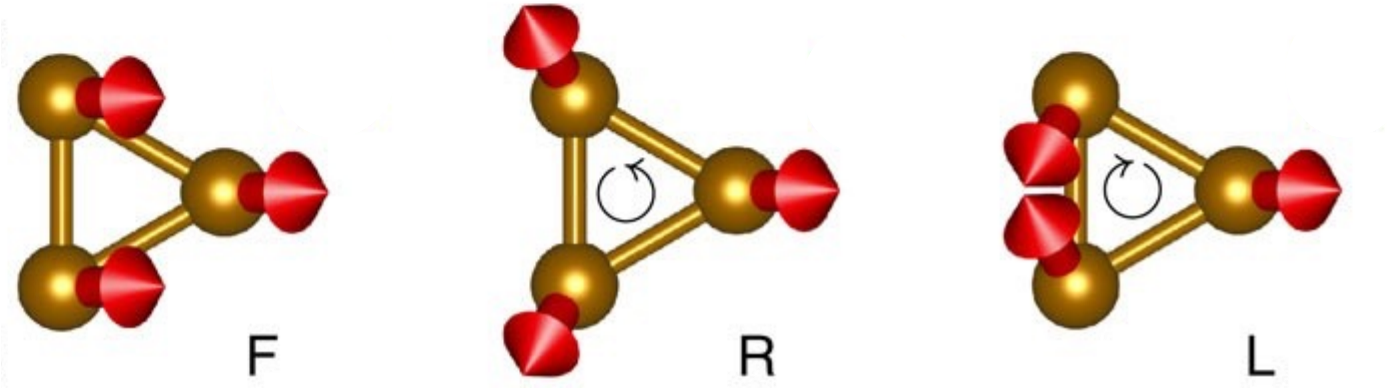
$$\mathbf{B}^{\text{eff}} \propto \chi_{ijk}\, \boldsymbol{\tau}_{ijk}$$

in the direction normal to the triangle <i,j,k>.

This leads to a topological orbital moment:

$$\mathbf{L}_{ijk}^{\text{TO}} = \kappa_{ijk}^{\text{TO}}\, \chi_{ijk}\, \boldsymbol{\tau}_{ijk}$$

proportional to the orbital susceptibility, $\kappa^{\text{TO}}$




dos Santos Dias et al., Nature Commun. **7**, 13613 (2016)

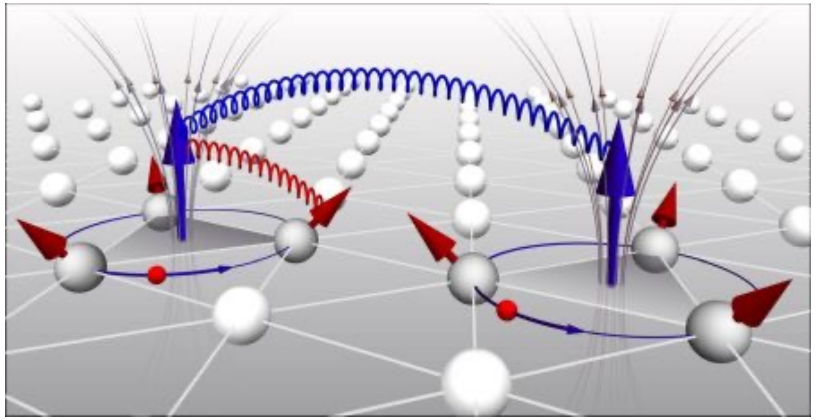


JÜLICH Forschungszentrum

# FeGe/ MnGe - MORE EXOTIC INTERACTIONS

## chiral-chiral and spin-chiral interactions

Chiral-chiral (CC) interaction between **B**$^{\text{eff}}$ and **L**$^{\text{TO}}$ located at the same triangle:



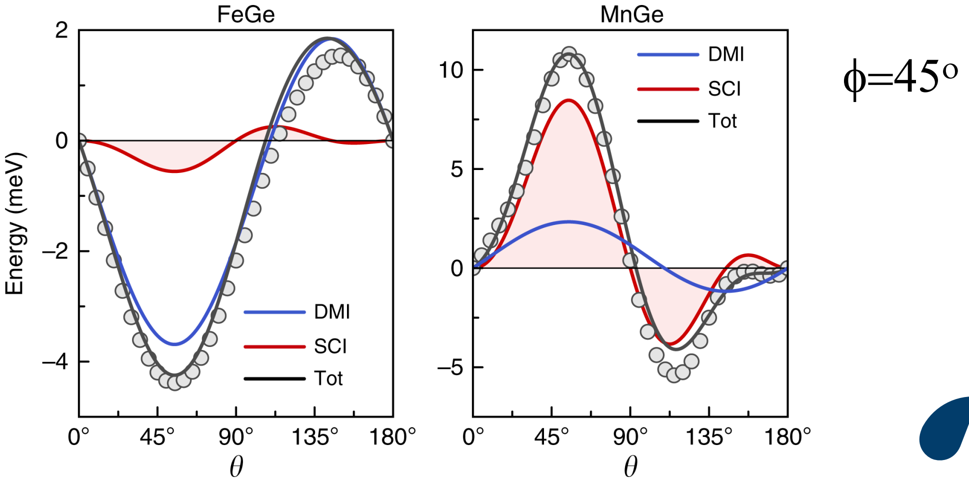
$$E^{\text{CC}} = -\frac{1}{2}\sum_{i\langle jk\rangle} \kappa^{\text{CC}}_{ijk}\,\chi^2_{ijk} = -\frac{1}{2}\sum_{i\langle jk\rangle} \kappa^{\text{CC}}_{ijk}\,(\mathbf{S}_i\cdot(\mathbf{S}_j\times\mathbf{S}_k))^2$$

Spin-chiral (SC) interaction between **L**$^{\text{TO}}$ and **S**:

$$E^{\text{SC}} = -\frac{1}{2}\sum_{i} \kappa^{\text{SC}}_{i}\,\mathbf{L}^{\text{TO}}_i\cdot\mathbf{S}_i = -\frac{1}{2}\sum_{i\langle jk\rangle} \kappa^{\text{SC}}_{ijk}\,(\boldsymbol{\tau}_{ijk}\cdot\mathbf{S}_i)(\mathbf{S}_i\cdot(\mathbf{S}_j\times\mathbf{S}_k))$$

is a relativistic (spin-orbit coupling) effect.

The SC interaction can account for effects ascribed to the DMI, but is intrinsically related to 3-D spin structures (like in MnGe).



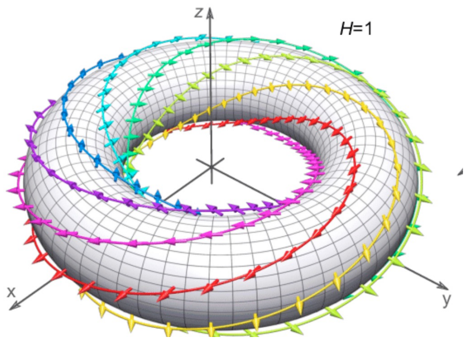



JÜLICH Forschungszentrum

# CONCLUSIONS

- DFT calculations can be converged to consistent results for exchange constants (comparison to experiment might need corrections)
- Selection of the spin-model is crucial (Heisenberg and DMI are just spin ½ models, usually higher orders are important)
- Higher order interactions can lead to new ground states and influence excited states (finite T)
- Chiral interactions open new possibilities to realize 3-dimensionally modulated magnetic structures.

**Thank you for your attention!**




Rybakov et al., preprint (2019)