The sign of the Dzyaloshinskii-Moriya interaction *vs* atomic structure of weak ferromagnets: theory and experiment

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Outline

1) Symmetry of Dzyaloshinskii-Moriya interaction: relation with ***local structural chirality*** *of crystals.*

2) Interference of magnetic and non-magnetic channels in x-ray diffraction allows us to deduce ***the sign*** *of the Dzyaloshinskii-Moriya interaction even in centrosymmetric canted antiferromagnetics :* $FeBO_3$, $MnCO_3$, α-Fe_2O_3, *etc.*

Dmitrienko, Ovchinnikova et al., ***JETP Letters****,* ***92****, 383 (2010)*

3) Experiment and ***ab initio*** *simulations for* $FeBO_3$ *and* α-Fe_2O_3 *crystals (LSDA+U+SO).*

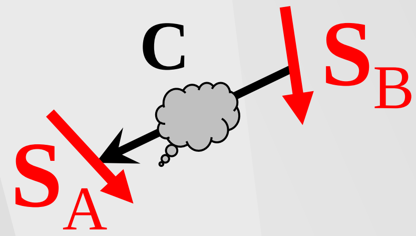
Dmitrienko, Ovchinnikova et al., ***Nature Physics****,* ***10****, 202 (2014)*

Dzyaloshinskii–Moriya interaction

Dzyaloshinskii, 1957

Moriya, 1960

Local asymmetry determines **D**:

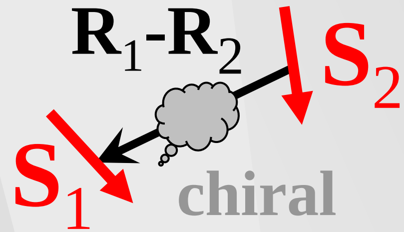


$$E_{DM} = \mathbf{D}\cdot[\mathbf{S}_A \quad \mathbf{S}_B]$$

Table 1. The Moriya Rules [1] that govern the DM vector, $\mathbf{D}$, between two spins at points A and B with a mid-point at C, and a comment on whether these are consistent with $\mathbf{D}$ transforming as a polar or axial vector

Moriya Rule	Polar?	Axial?
1. When a center of inversion is located at C, $\mathbf{D} = \mathbf{0}$	yes	no
2. When a mirror plane perpendicular to AB passes through C, $\mathbf{D} \parallel$ mirror plane or $\mathbf{D} \perp AB$	yes	no
3. When there is a mirror plane including A and B, $\mathbf{D} \perp$ mirror plane	no	yes
4. When a two-fold rotation axis perpendicular to AB passes through C, $\mathbf{D} \perp$ two-fold axis	no	no
5. When there is an n-fold axis ($n \geq 2$) along AB, $\mathbf{D} \parallel AB$	yes	yes

Dzyaloshinskii–Moriya interaction



$|\mathbf{R}_1\text{-}\mathbf{R}_2|=1$

The DM vector **D** is similar to the gyration vector **g !**

$$E_{DM} = \mathbf{D}_{12}\cdot[\mathbf{S}_1\ \ \mathbf{S}_2]$$

$$E_{DM} = T_{lkm}\,(\mathbf{S}_1)_l\,(\mathbf{S}_2)_k\,(\mathbf{R}_1\text{-}\mathbf{R}_2)_m$$

The DM tensor T_{lkm} is antisymmetric in the first two indices

$$T_{lkm} = -T_{klm}$$

It corresponds to the crystal symmetry of the midpoint ($\mathbf{R}_1+\mathbf{R}_2$)/2.

Then all the Moriya rules are obviously working!

To return to **D**: $(\mathbf{D}_{12})_n = \frac{1}{2}\,\varepsilon_{lkn}\,T_{lkm}\,(\mathbf{R}_1\text{-}\mathbf{R}_2)_m$

Magnetic structure of $MnCO_3$, $FeBO_3$, etc.

$FeBO_3$ is a weak ferromagnetic

$E_{DM}=\mathbf{D}\cdot[\mathbf{S}_1 \quad \mathbf{S}_2]$

Dzyaloshinskii, 1957

Moriya, 1960

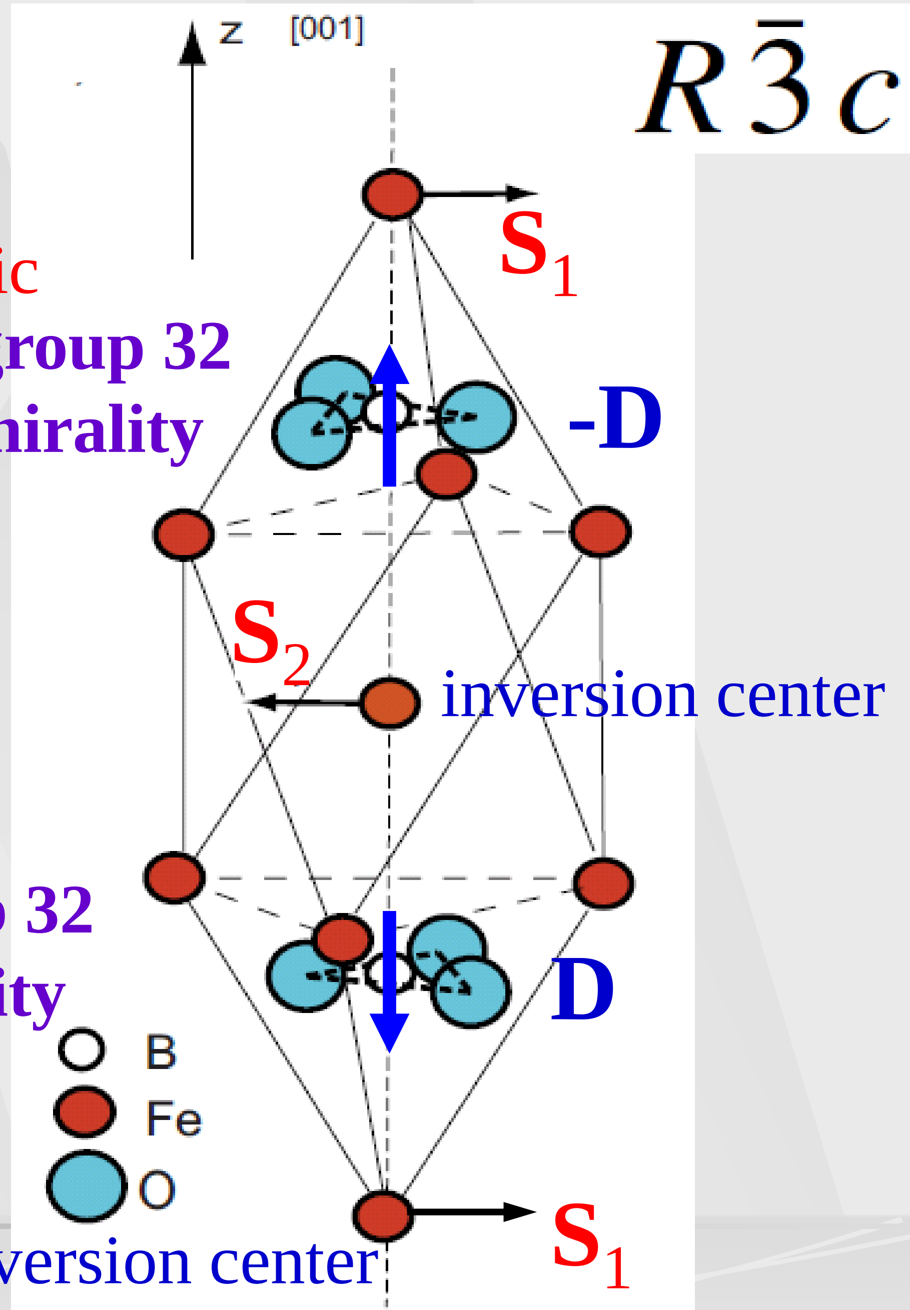
Vectors $\mathbf{S}_j$ are slightly canted, inducing a small ferromagnetic moment $\mathbf{M}\perp\mathbf{D}$ and $\mathbf{M}\perp\mathbf{S}_j$.

Local chirality determines the sign of $\mathbf{D}$?

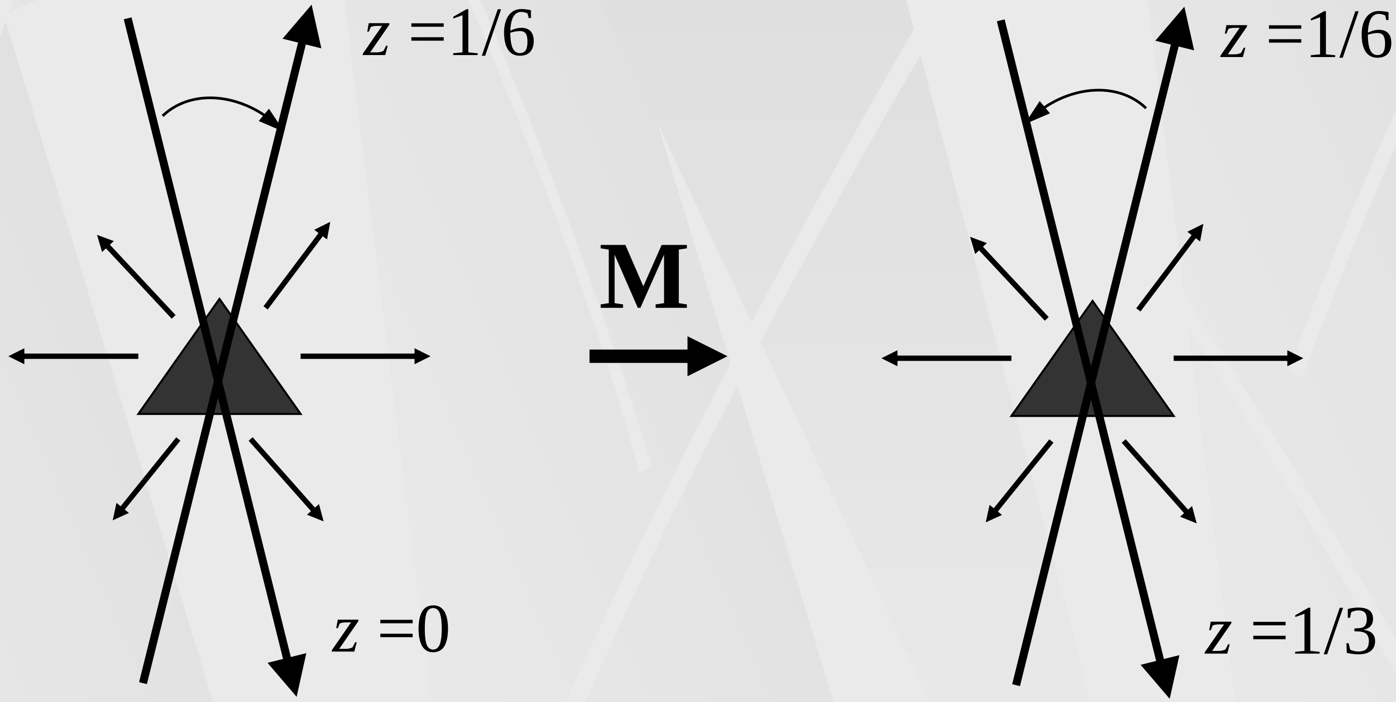
two magnetic atoms/cell; crystal is centrosymmetric

point group 32 local chirality

point group 32 local chirality



Local twist of moments from layer to layer in $FeBO_3$



left-hand twist

right-hand twist

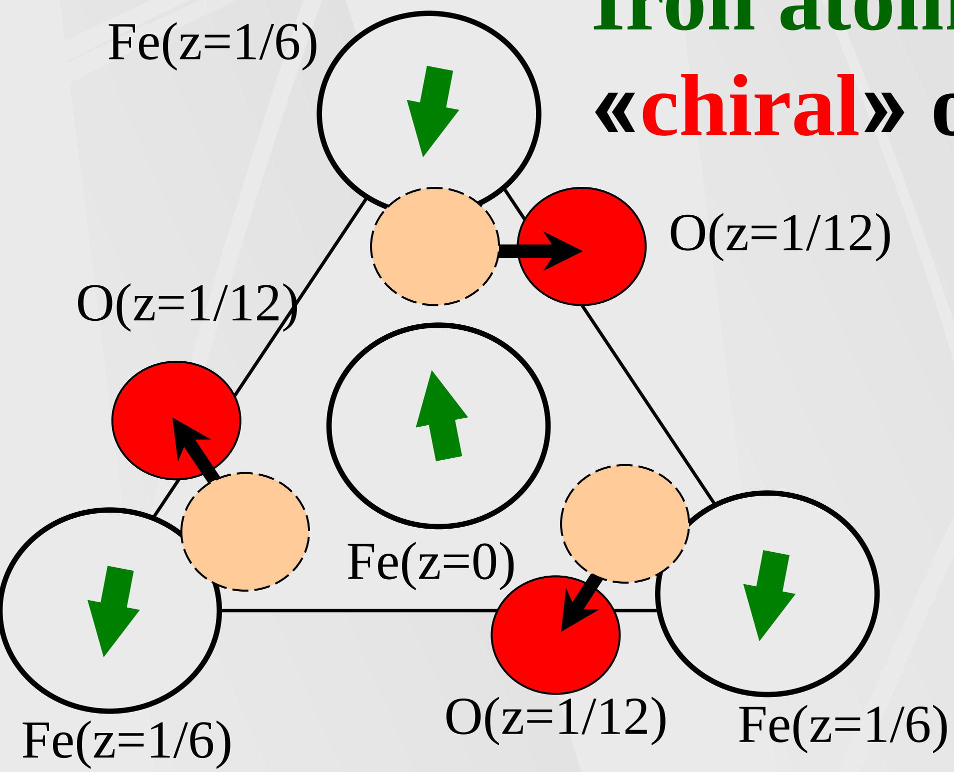
What is the reason for such alternating twist?

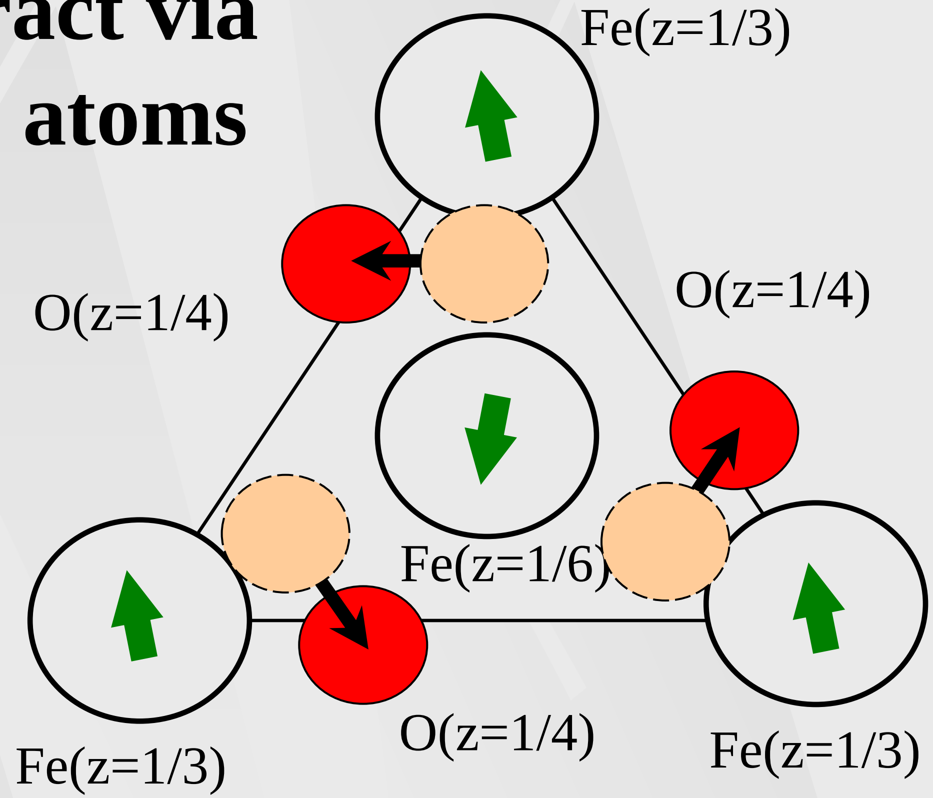
Local chirality as a reason for alternating twist of Fe spins in $FeBO_3$; space group- $R\bar{3}c$

from z=0 to z=1/6: left-hand twist of the oxygen-atom triangle and left-hand twist of the Fe spins

from z= 1/6 to z=1/3: right-hand twist of the oxygen-atom triangle and right-hand twist of the Fe spins

Iron atoms interact via «chiral» oxygen atoms





from z=0 to z=1/6: left-hand twist of of spins or magnetic moments (green) is determined by the sign of **D**

from z=1/6 to z=1/3: right-hand twist of spins or magnetic moments (green)

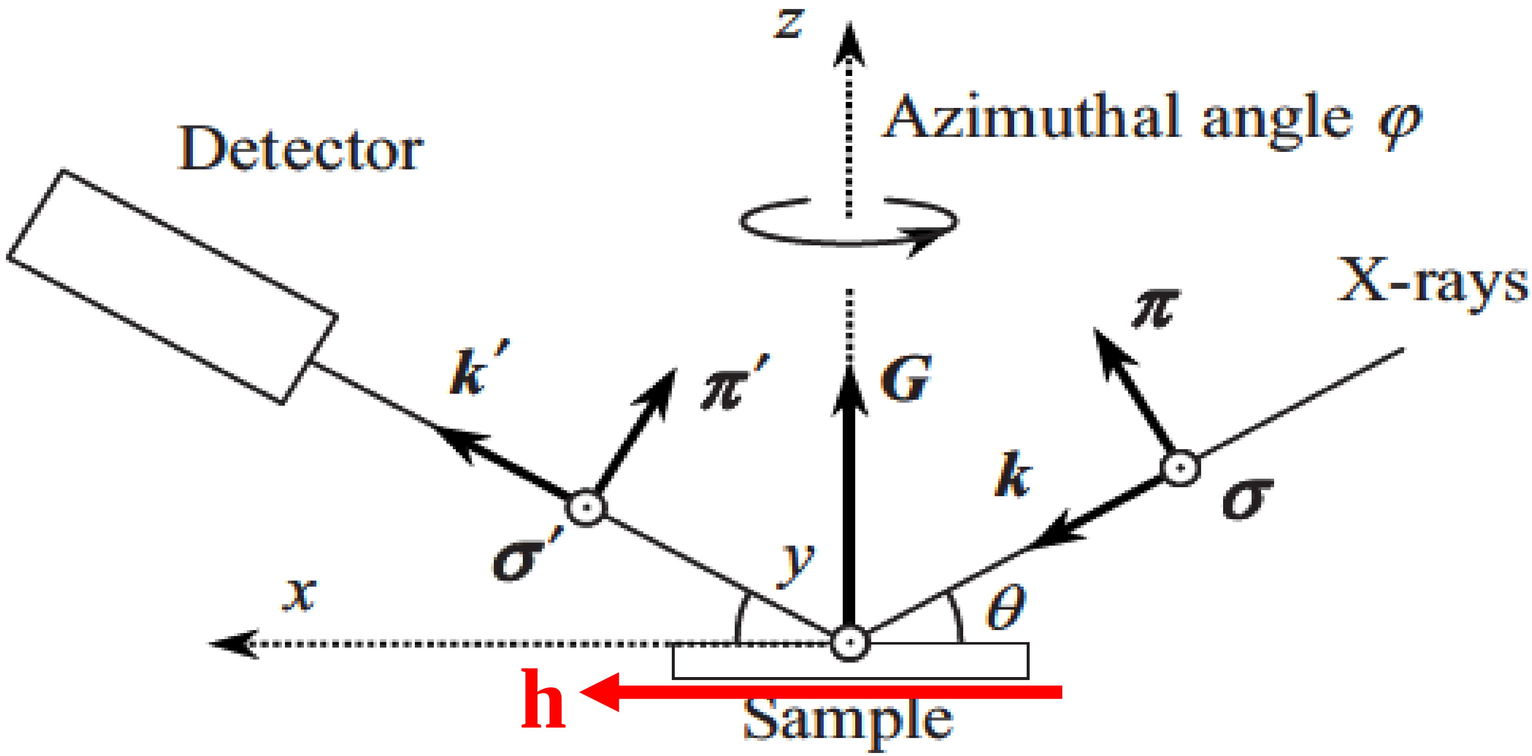
In hexagonal system: Oxygen at (0.2981, 0, 1/4)

Typical setup

Common: Interference between magnetic and structural scattering.

Two methods:

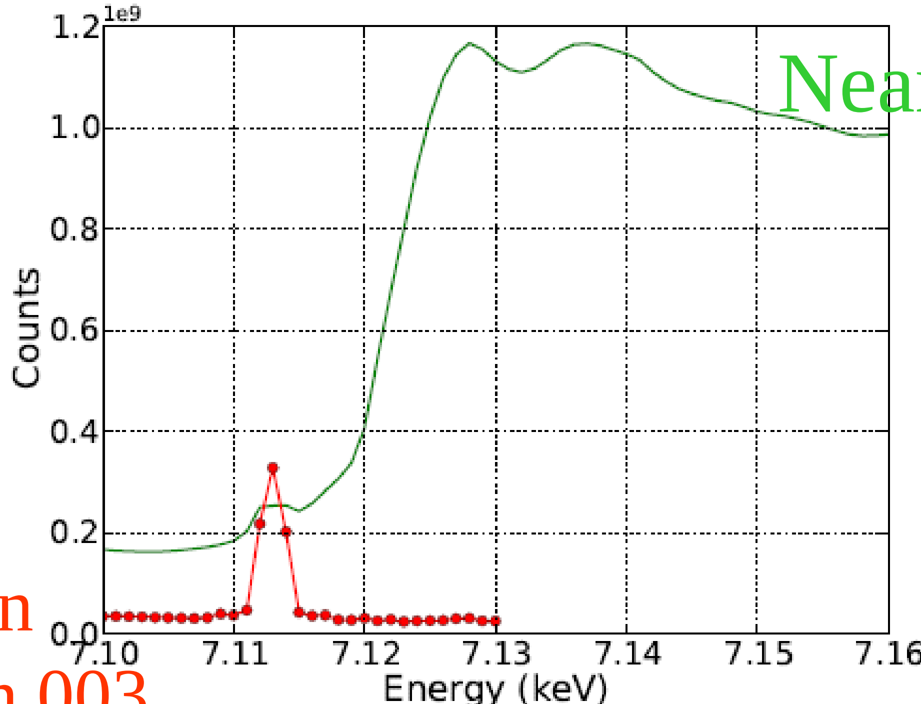
1. Resonant
2. Multiple-beam diffraction



h - weak ferromagnetic moment fixed by external magnetic field

FIG. 2. Definition of the optical setting. σ and π denote polarization vectors for the incident x rays, σ' and π' for the scattering x rays. k and k' denote the wave vectors for the incident and scattering x rays, respectively, G the scattering vector; z axis is parallel to the threefold crystal axis, and the x axis is normal to one of three vertical glide planes and parallel to the hexagonal a axis. When the azimuthal angle φ equals zero, vector $k+k'$ coincides with the x axis; just this case is shown in the figure. X-ray beam is rotated clockwise viewed from $+z$ side with the angle φ, i.e., sample crystal is rotated counterclockwise relative to the x-ray beam.

Typical experimental results (resonant method)



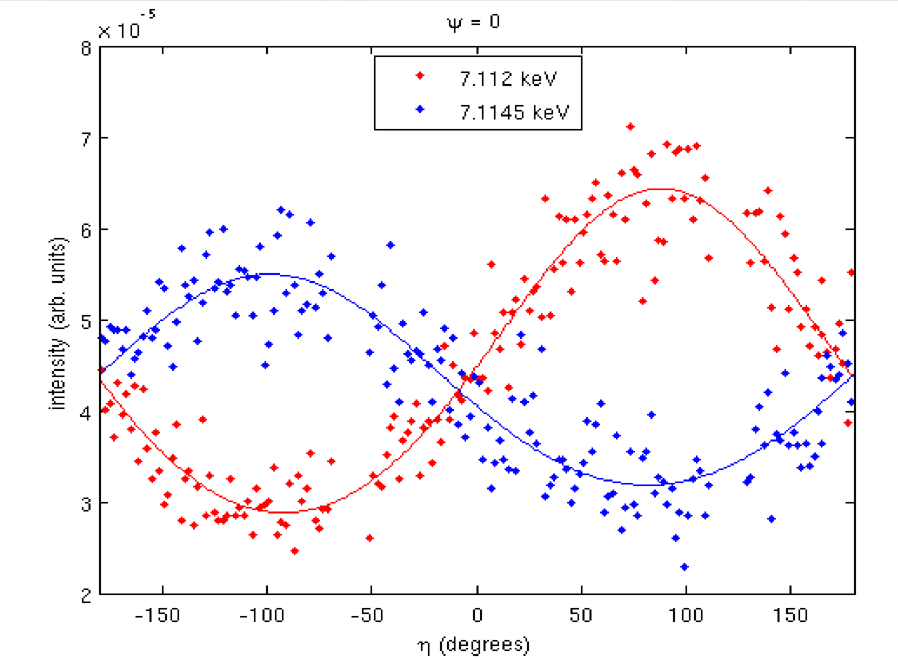
Near-edge absorption

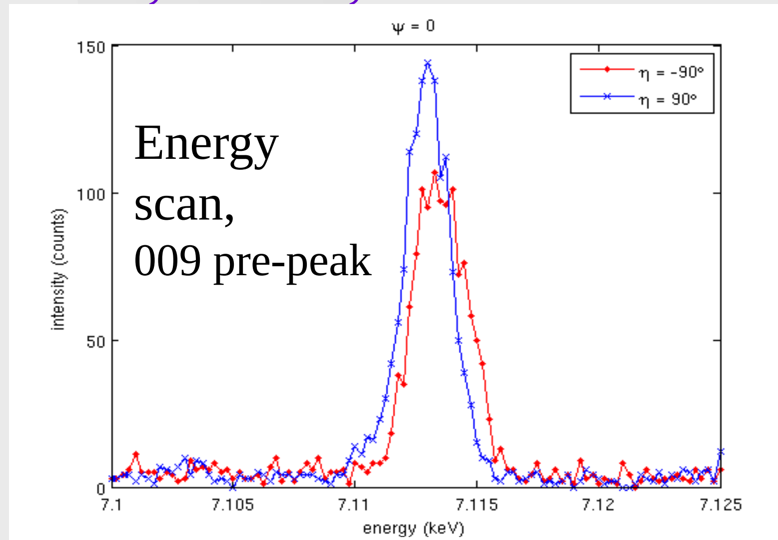
Resonant diffraction forbidden reflection 003

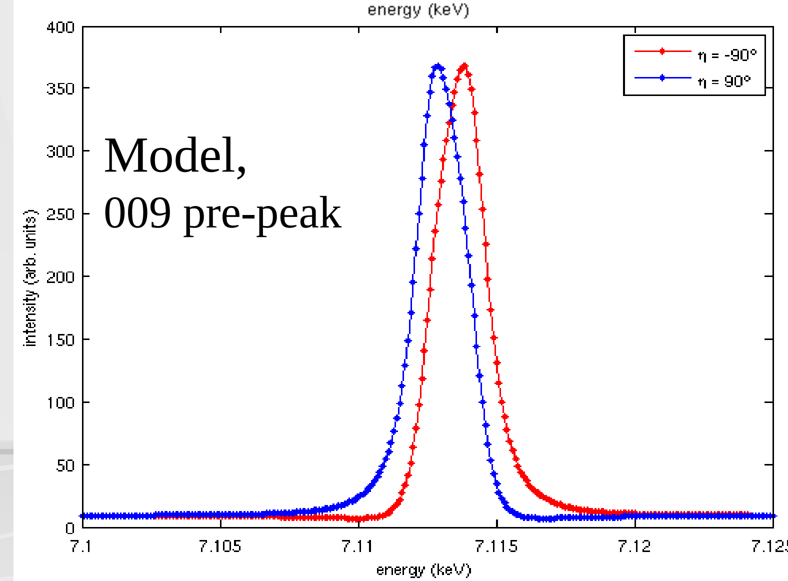
FIG. 3. The absorption (fluoresence) spectrum of $FeBO_3$ near the Fe K edge (green line), showing a weak pre-edge peak. Red: the resonant (003) forbidden scattering spectrum, showing a single sharp peak at the absorption pre-edge position.

Interference of non-resonant magnetic and resonant diffraction in $FeBO_3$

S.Collins, G.Beutier, G.Nisbet, XMaS experiment, ESRF, Grenoble







Azimuthal magnet scan, 009

Conclusion: in $FeBO_3$, vector **D** induces the left-hand rotation of moments between 0,0,0 and ½,½,½

FDMNES x-ray simulations

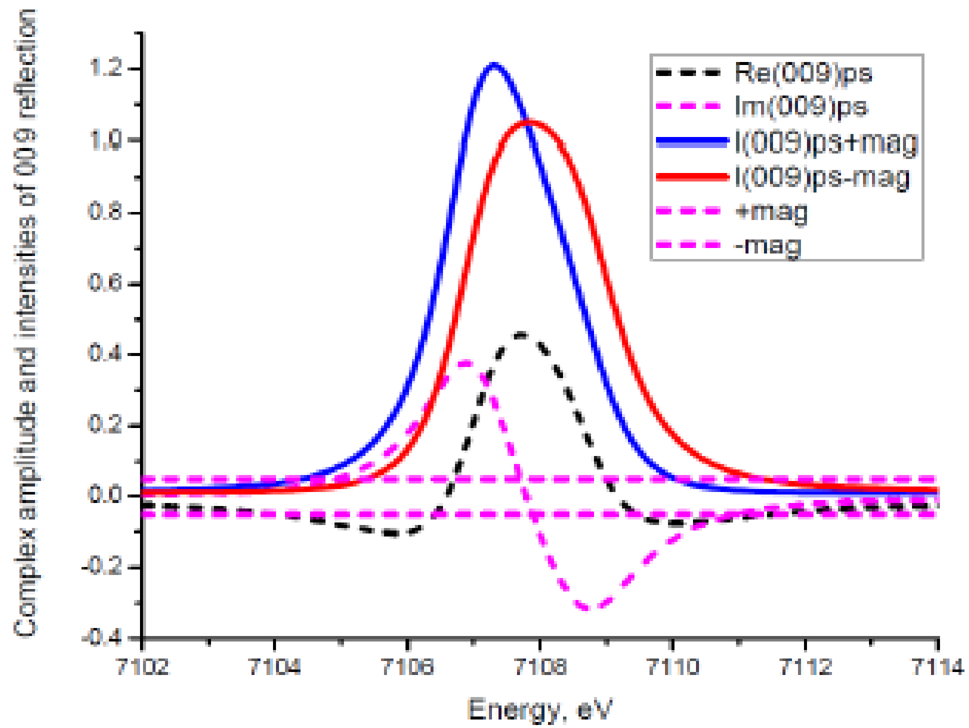


FIG. 7. Complex scattering amplitudes and intensities of the σ-to-π (009) forbidden reflection, calculated using the FDMNES program. The magenta and black dotted lines correspond to the real and imaginary parts of the resonant structure factor in the pre-edge region just below the Fe K-edge. Straight magenta lines show pure imaginary amplitudes of non resonant (energy independent) magnetic scattering amplitudes which are >0 for η=0 and <0 for η=-60°. The blue and red lines show intensities (square modulus of amplituds) for η=0 and η=-60° correspondingly; they

ab initio magnetic structure simulations

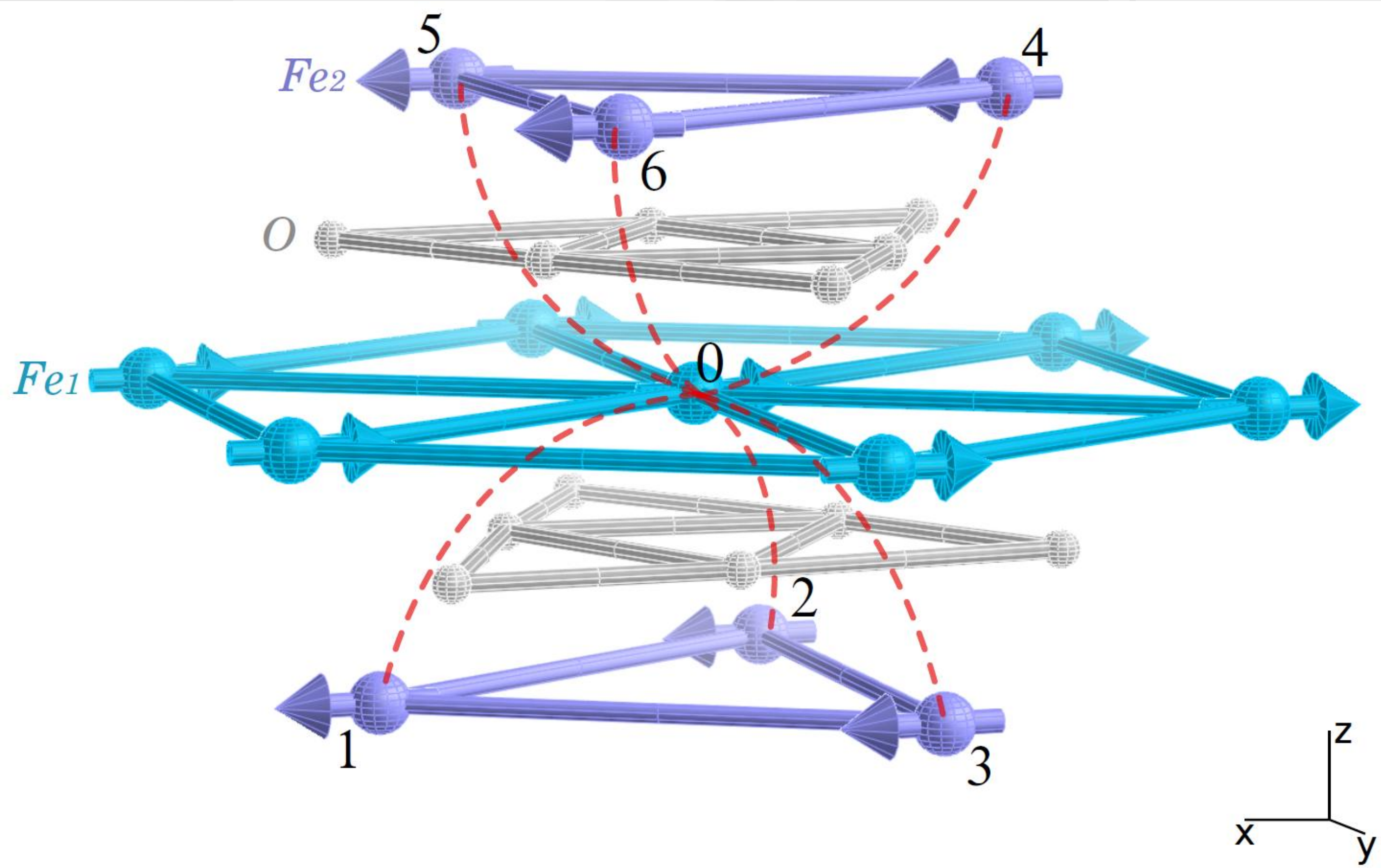


TABLE I. Calculated values of isotropic exchange interactions between magnetic moments in $FeBO_3$ (in meV). The number in parentheses denotes the coordination sphere.

$Fe^{(1)}$	$Fe^{(2)}$	$Fe^{(3)}$	$Fe^{(4)}$	$Fe^{(5)}$	$Fe^{(6)}$	$Fe^{(7)}$
10.28	0.21	0	0.54	-0.08	0	0.02

DM local vectors in $FeBO_3$

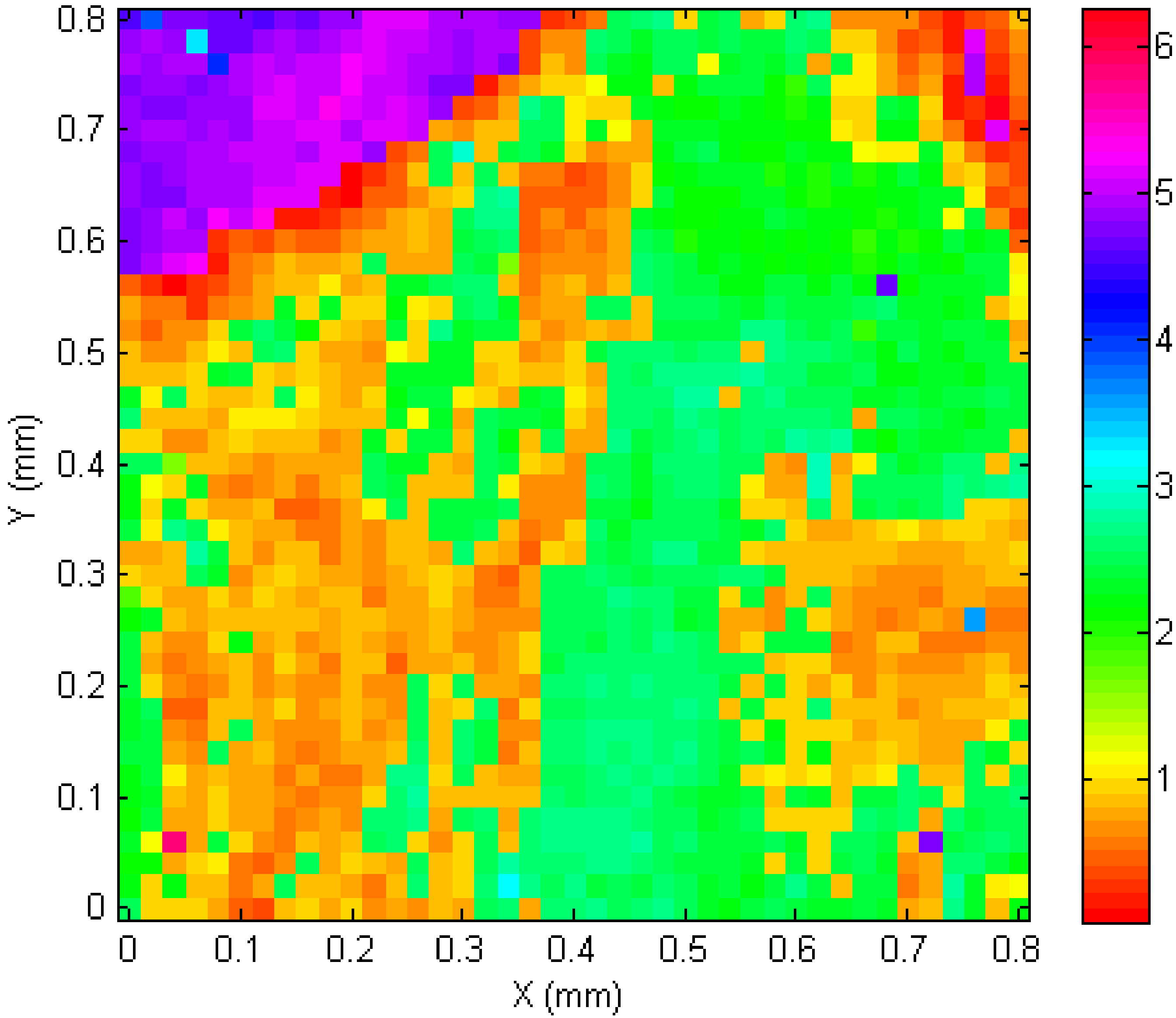
The six first neighbours are most important!

Parameters of Dzyaloshinskii-Moriya interaction (in meV) calculated *ab initio* (LDA+U+SpinOrbit)

Bond $i - j$	$\mathbf{R}_{ij}$	$\mathbf{D}_{ij}$ (meV)
0-1	(1.0 ; 0.0 ; -0.9044)	(-0.249; 0.0; -0.240)
0-2	(-0.5 ; $-\sqrt{3}/2$; -0.9044)	(0.124 ; 0.216 ; -0.240)
0-3	(-0.5 ; $\sqrt{3}/2$; -0.9044)	(0.124 ; -0.216 ; -0.240)
0-4	(-1.0 ; 0.0 ; 0.9044)	(-0.249; 0.0 ; -0.240)
0-5	(0.5 ; $-\sqrt{3}/2$; 0.9044)	(0.124 ; -0.216 ; -0.240)
0-6	(0.5 ; $\sqrt{3}/2$; 0.9044)	(0.124 ; 0.216 ; -0.240)

Beutier, Collins et al. *Acta Cryst.* **A70**, C1356 (2014)

Mapping **domains** in the weak ferromagnet $CoCO_3$



antiferromagnetic **domains**

20 microns resolution!!!

Antiferromagnetic structure of Fe_2O_3 in magnetic field $\mathbf{H} \perp z$

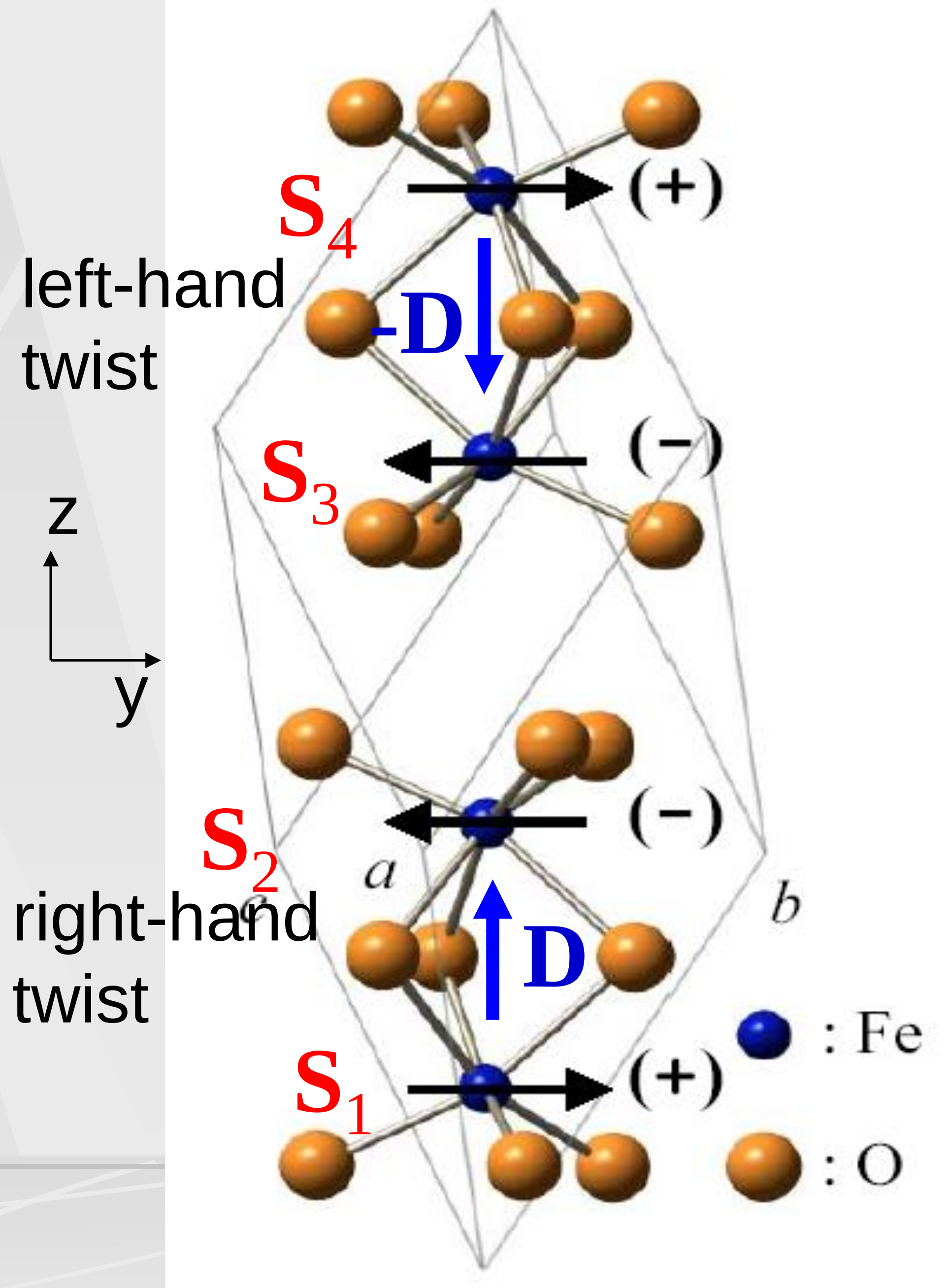
If the field is directed to us ($\mathbf{H} \parallel x$):

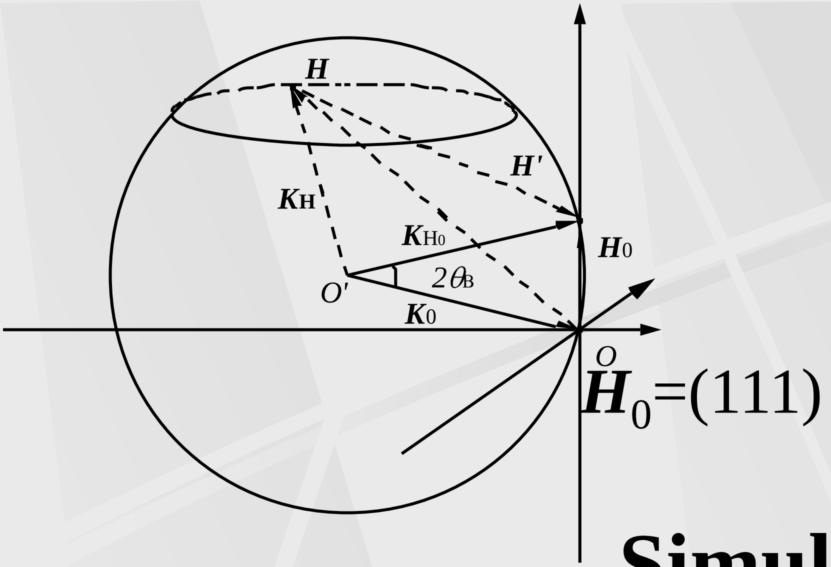
$$E_{DM} = -D_z\,(S_{1y} - S_{2y})S_{jx}$$

The phase of the spin alternation depends on the sign of vector **D**.

We have measured this phase via interference with the **Renninger** reflections.

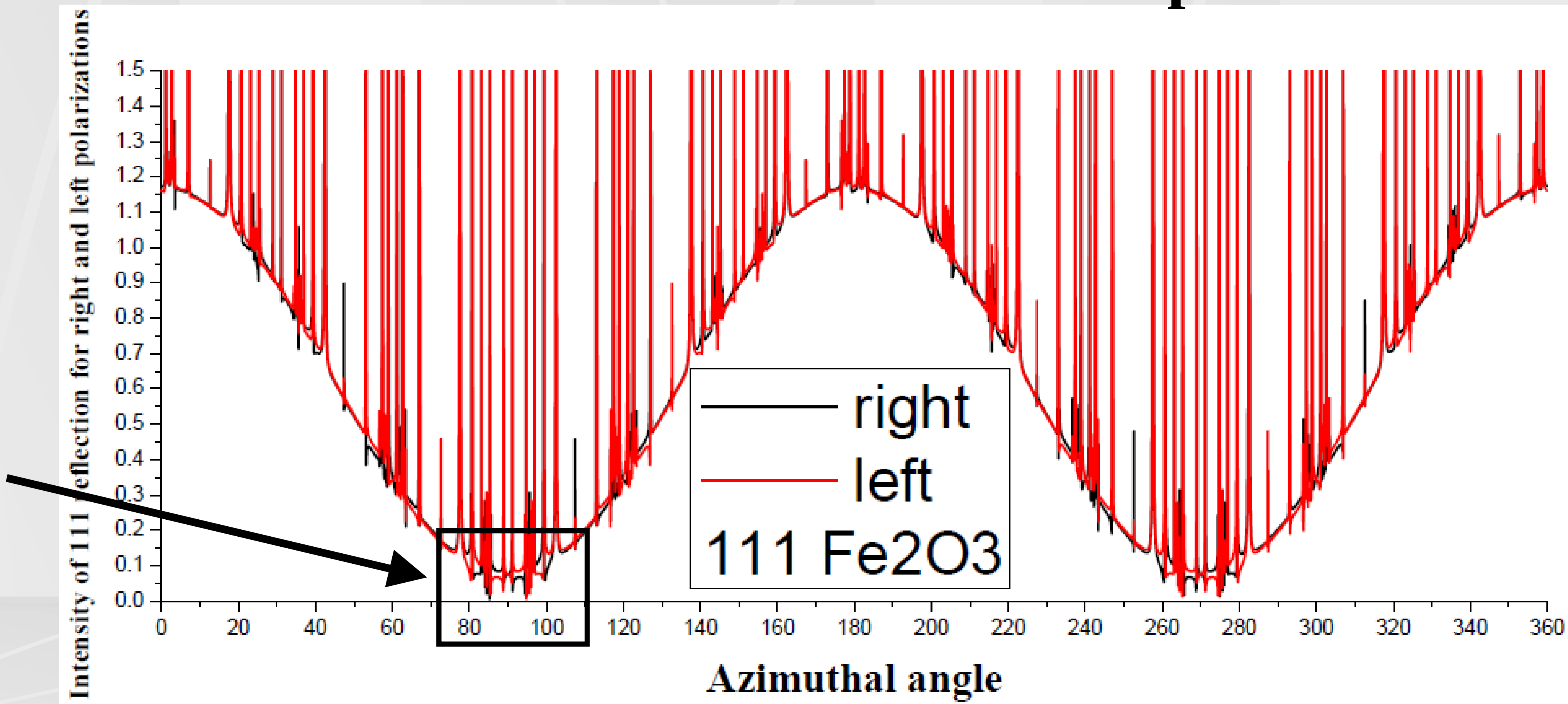
1. Far from absorption edge.
2. Circular polarizations.





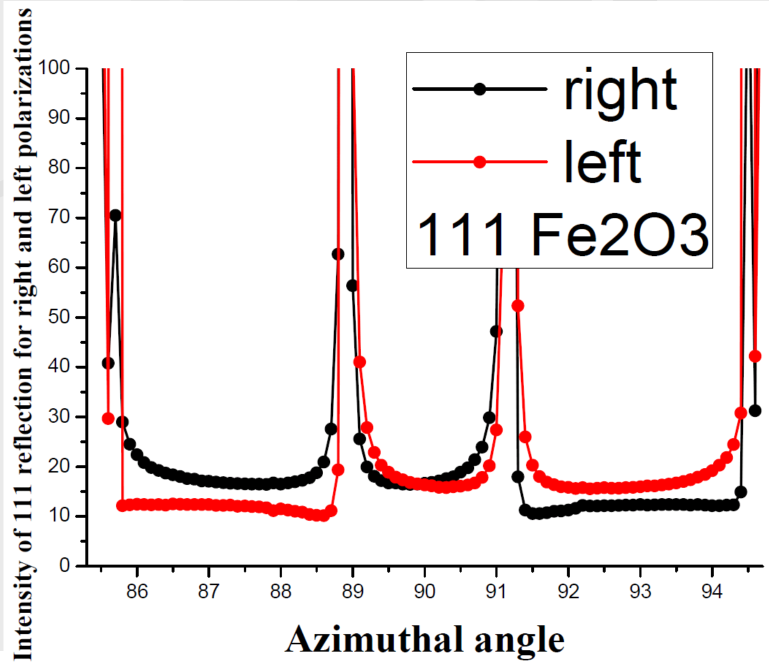
Interference between magnetic and multiple-beam diffraction in hematite α-Fe_2O_3

Simulations for two circular polarizations

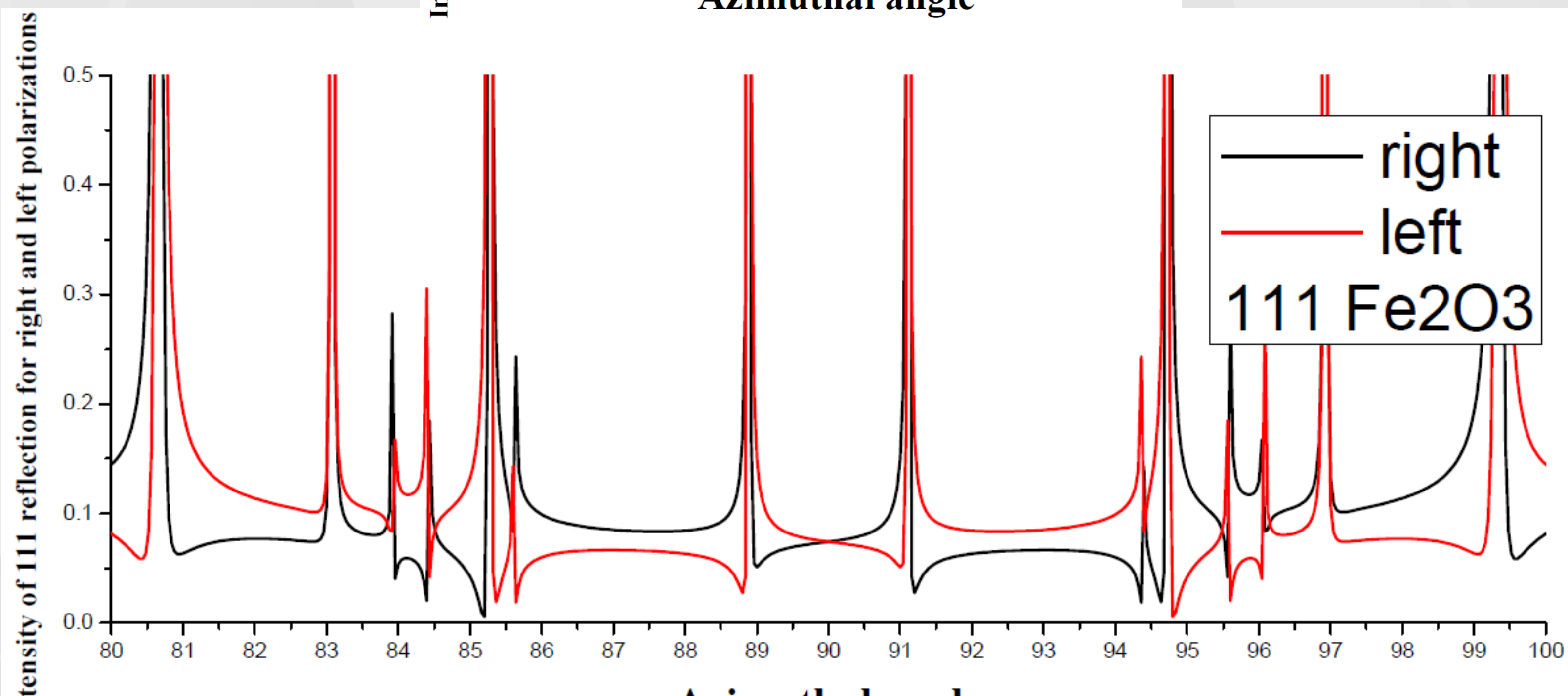


Interference with multiple-beam diffraction

Rogalev,
de Bergevin,
Kokubun,
et al.,
ID12, ESRF



measured



simulated

Conclusions

- The sign of the Dzyaloshinskii-Moriya interaction in centrosymmetric crystals is determined by ***local chirality*** of their atomic structure
- For sign measurements in $FeBO_3$ and Fe_2O_3 we used:
  1) interference between magnetic and structural diffraction;

  2) the single-domain magnetic state fixed by an external magnetic field;

  3) simulations both for x-ray diffraction and for the Dzyaloshinskii-Moriya interaction.

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