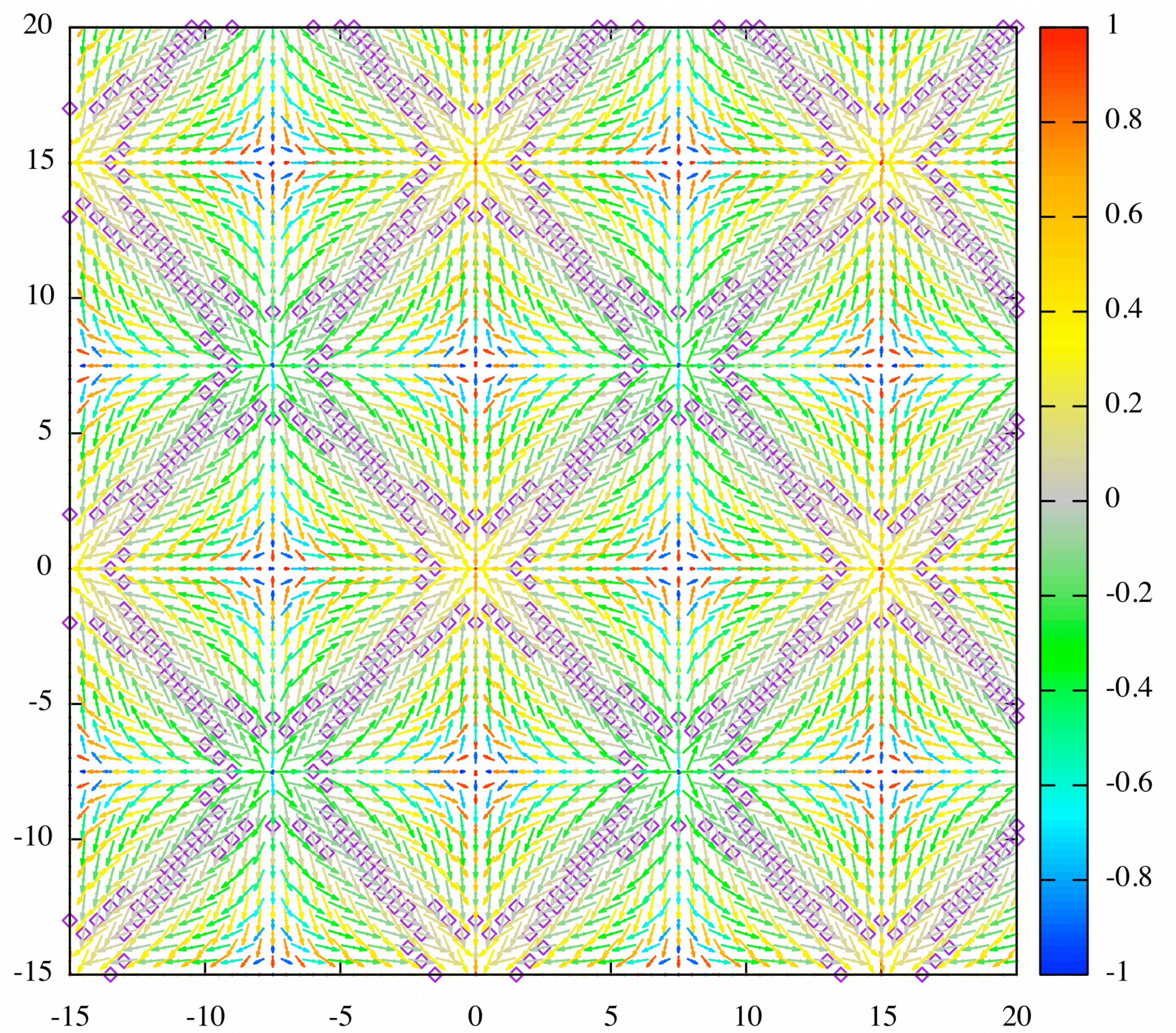
Magnetic neutron diffraction

Vladimir Pomjakushin
Laboratory for Neutron Scattering and Imaging
Paul Scherrer Institut PSI
Switzerland



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or short link http://psi.ch/node/29534

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1. Introduction to magnetic neutron diffraction (1-13)
   1.1. General. Intro. Experimental technics for magnetic diffraction
   1.2. Examples of instruments at PSI
   1.3. Literature, computer and web-resources related to magnetic diffraction
2. Basic principles of magnetic neutron diffraction. (15-32)
   2.1.Master formulae for the scattering. neutron-electron interaction Hamiltonian. Scattering Q-operator (15-20)
   2.2. Magnetic order parameters overview. Magnetic multipoles (22-27)
   2.3.Magnetic form-factors (what are neutrons sensitive to?) Expansion of Q [exp(i**kr**) series] (29-40)
      2.3.1. Dipole approximation. Examples.
      2.3.2. Multipole approximation, parity even, time odd. Symmetry of multipoles.
      2.3.3. Anapole (toroidal moment) and other parity-odd modern exotics (theory: Lovesey)
3. Description and determination of magnetic structure (41-72)
   3.1. Introduction to propagation vector(s) formalism star/arm (42,43,51)
   3.2. Magnetic structure factors. General formula 44-47
   3.3. Commensurate vs. incommensurate case's examples.
   3.4. Introduction to irreps (48-50)
   3.5. Magnetic Shubnikov groups (52-53)
4. Classification of the magnetic structures that are used in the literature, such as Shubnikov (or Magnetic) space groups, 3D+n superspace groups and irreducible representation (irrep) notations. Relation between two approaches. A bit of history. (54-59)
5. How can one construct all possible symmetry adapted magnetic structures for a given crystal structure and a propagation vector (a point on the Brillouine zone) by the combined use of irrep and the magnetic symmetry? A real case study of:
   5.1. multiferroic TmMnO3: 2D irrep k=[1/2,0,0].Ferro-electric phase polar magnetic group Pbmn21 (61-65)
   5.2. Topologically nontrivial skyrmionic incommensurate magnetic structure. Superspace. (66-72)

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Introduction to magnetic diffraction

neutron properties

mass m = 1.660 10 $^{-24}$ g = 939 MeV

spin: S=½

magnetic moment $\mu_n = \gamma\mu_N$ = -1.91 nuclear magnetons

The state of neutron is describe by its wave vector ***k***

plane wave $\psi \sim \exp(i\mathbf{kr})$

and its spin component S_z=±½

Instead of ***k*** we often find:

Energy $E = \dfrac{\hbar^2 k^2}{2m}$

momentum $\mathbf{p} = \hbar\mathbf{k}$

velocity $\mathbf{v} = \dfrac{\hbar\mathbf{k}}{m}$

wavelength $\lambda = \dfrac{2\pi}{k}$

neutron g-factor g_n=-3.8

$\mu_n = g_n S[\mu_N]$

$\gamma = g_n/2 = -1.91$

nuclear magneton

$\mu_N = e\hbar/2m_p c$

Bohr magneton

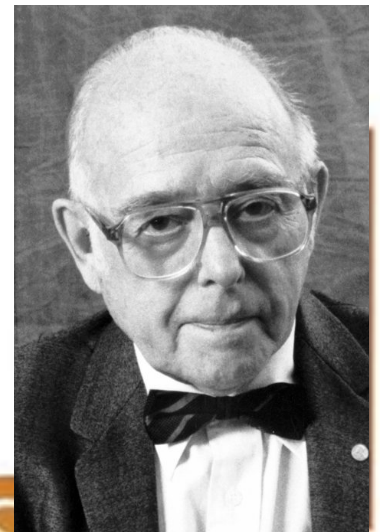
$\mu_B = e\hbar/2m_e c$

m_e=0.5 meV

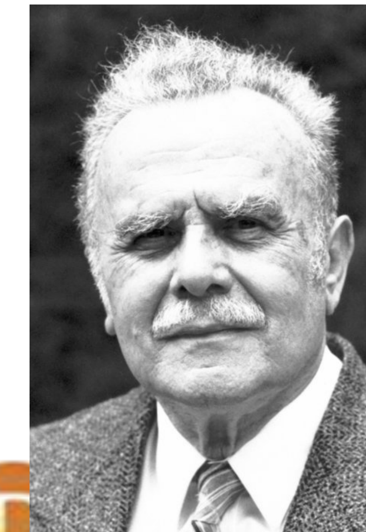
proton g_p=5.6, electron g_e=-2.0

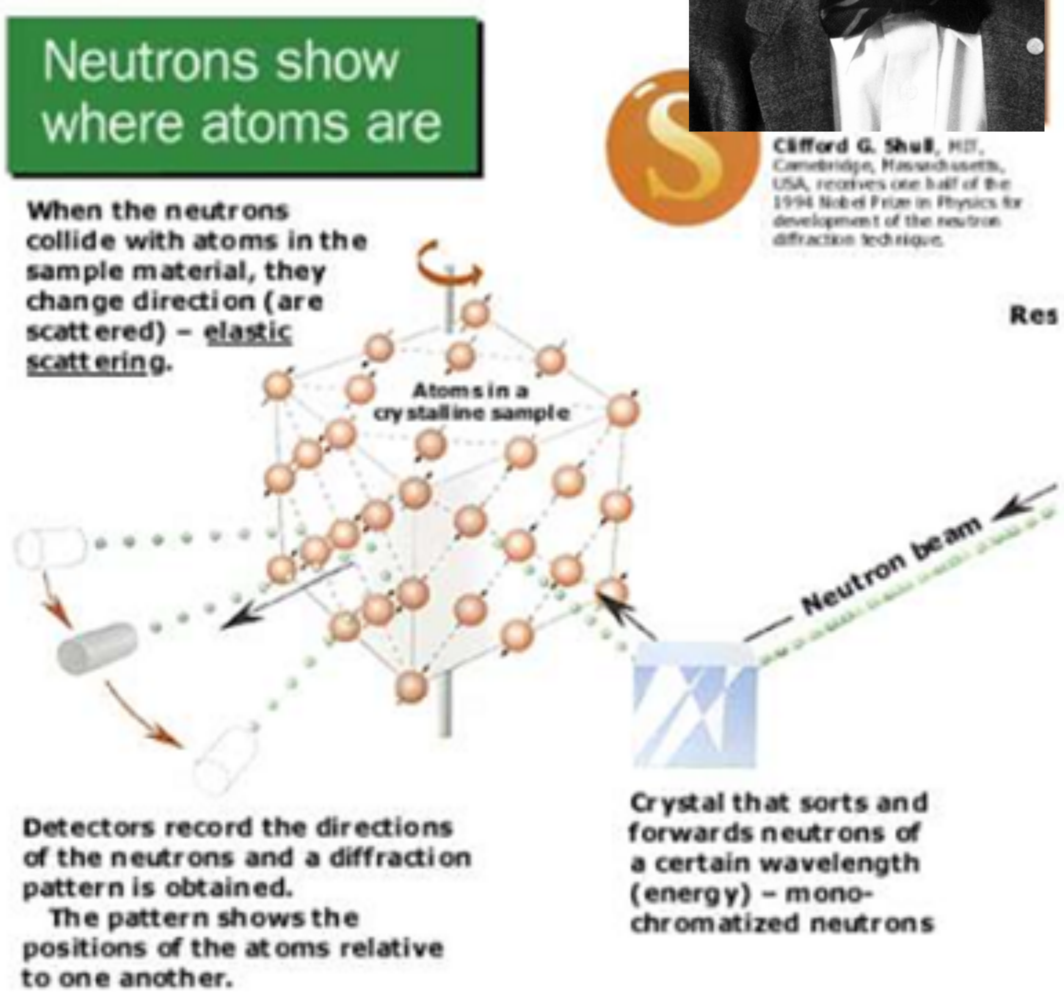
1994 Nobel Prize in Physics

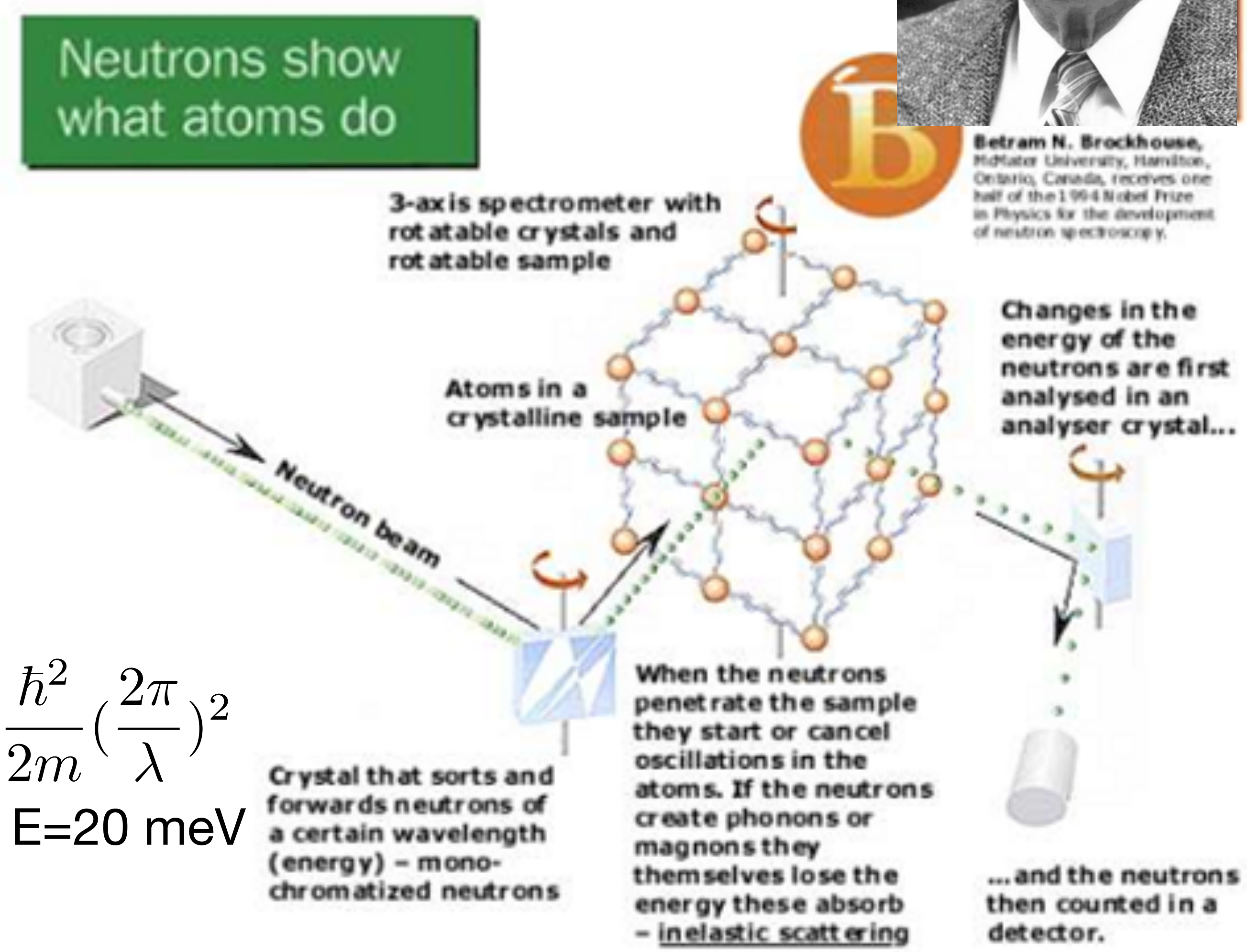
Clifford G. Shull
1915 – 2001, USA



Bertram N. Brockhouse
1918 – 2003, Canada







$$E = \frac{\hbar^2}{2m}\left(\frac{2\pi}{\lambda}\right)^2$$

λ=2Å, E=20 meV

https://www.nobelprize.org/prizes/physics/1994/summary/

Neutron Diffraction by Paramagnetic and Antiferromagnetic Substances

C. G. SHULL, W. A. STRAUSER, AND E. O. WOLLAN
Oak Ridge National Laboratory, Oak Ridge, Tennessee
(Received March 2, 1951)

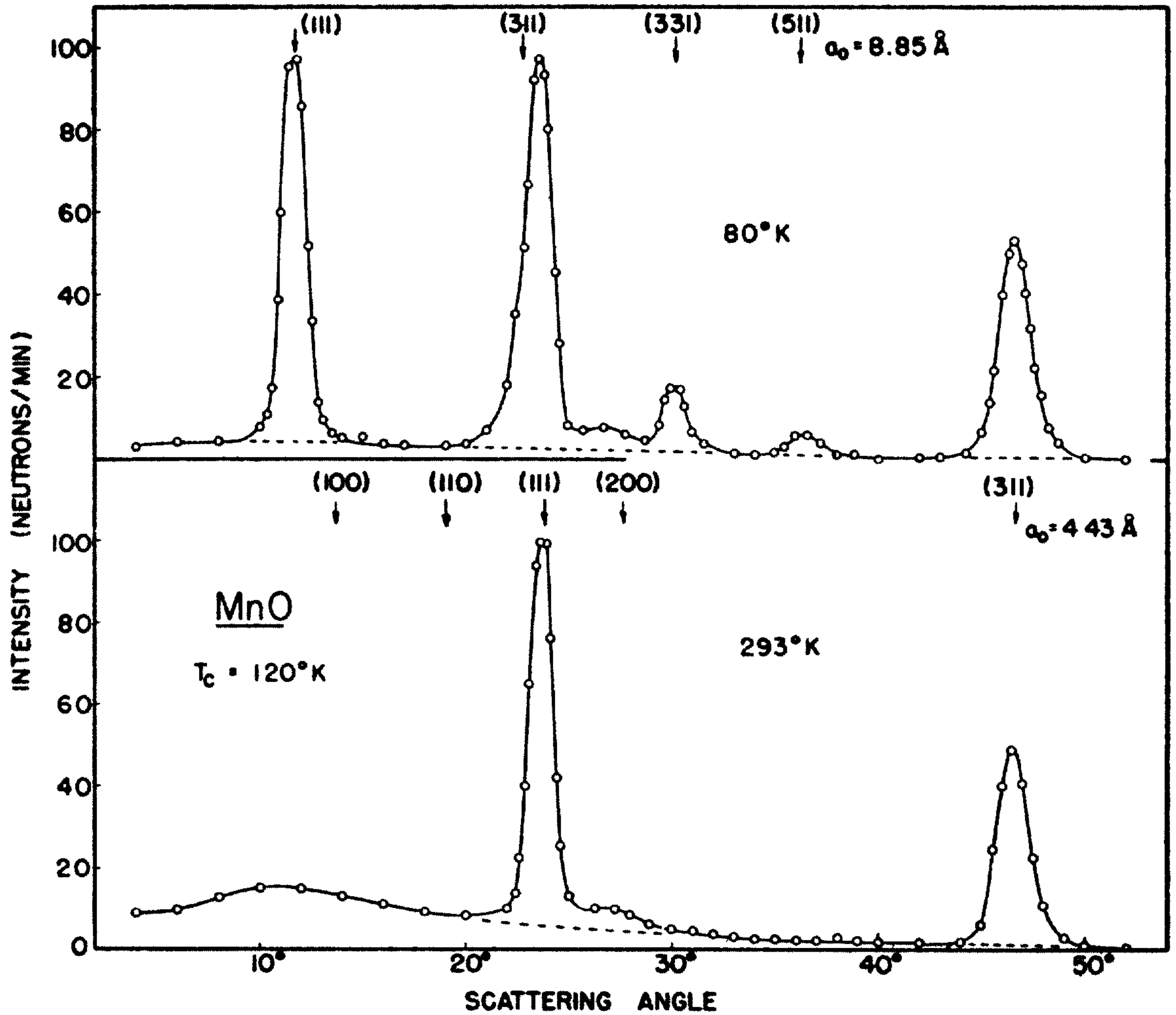


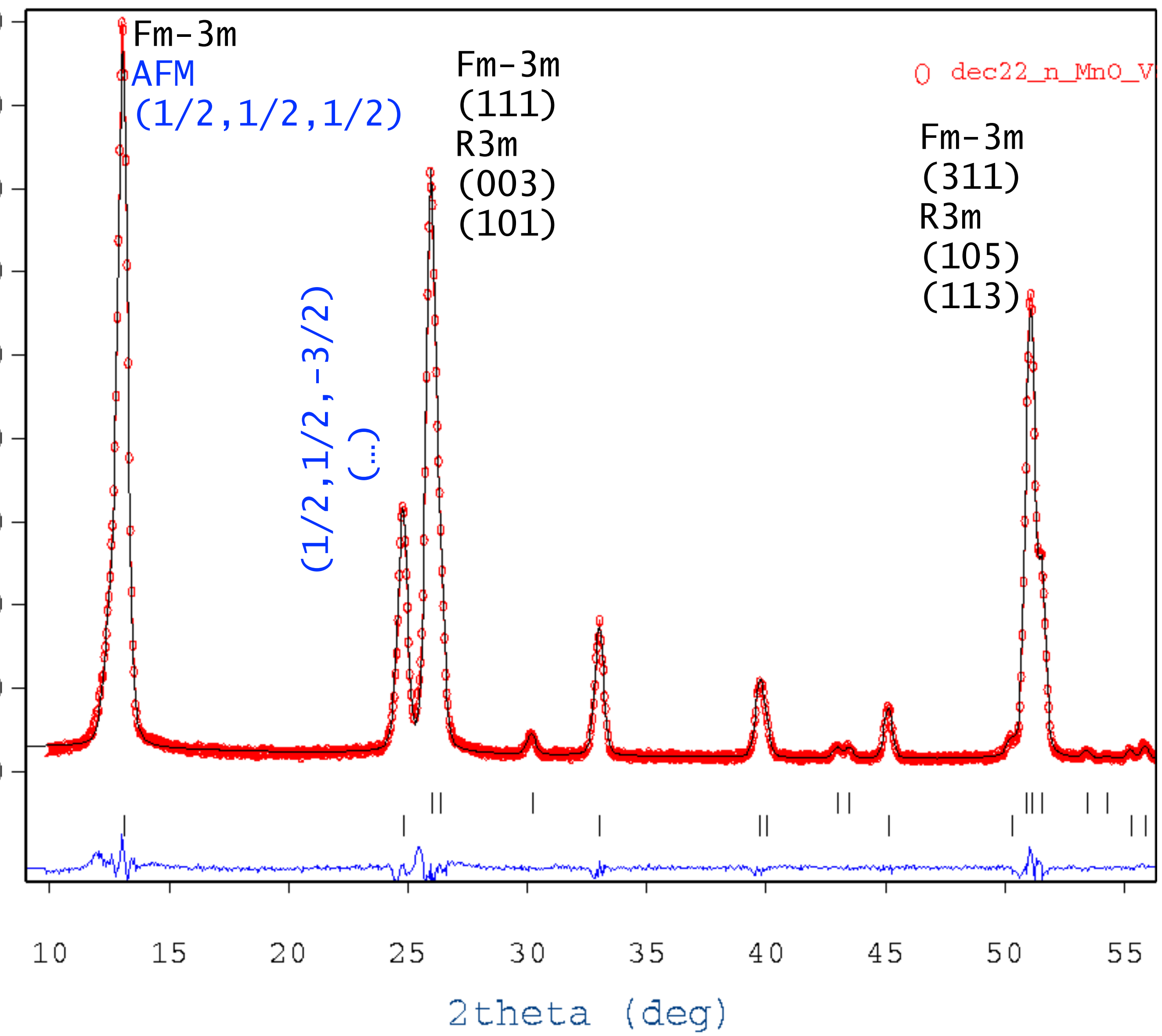
FIG. 4. Neutron diffraction patterns for MnO taken at liquid nitrogen and room temperatures. The patterns have been corrected for the various forms of extraneous, diffuse scattering mentioned in the text. Four extra antiferromagnetic reflections are to be noticed in the low temperature pattern.

VOLUME 83, NUMBER 2

HRPT/SINQ nowadays
λ=1.15Å, MnO @ 2K.

Rhombohedral distortions are explicitly seen

R-3m and k=003/2



raction by Paramagnetic and Antiferromagnetic

λ=1.057Å

C. G. SHULL, W. A. STRAUSER, AND E. O. WOLLAN
Oak Ridge National Laboratory, Oak Ridge, Tennessee

(Received March 2, 1951)

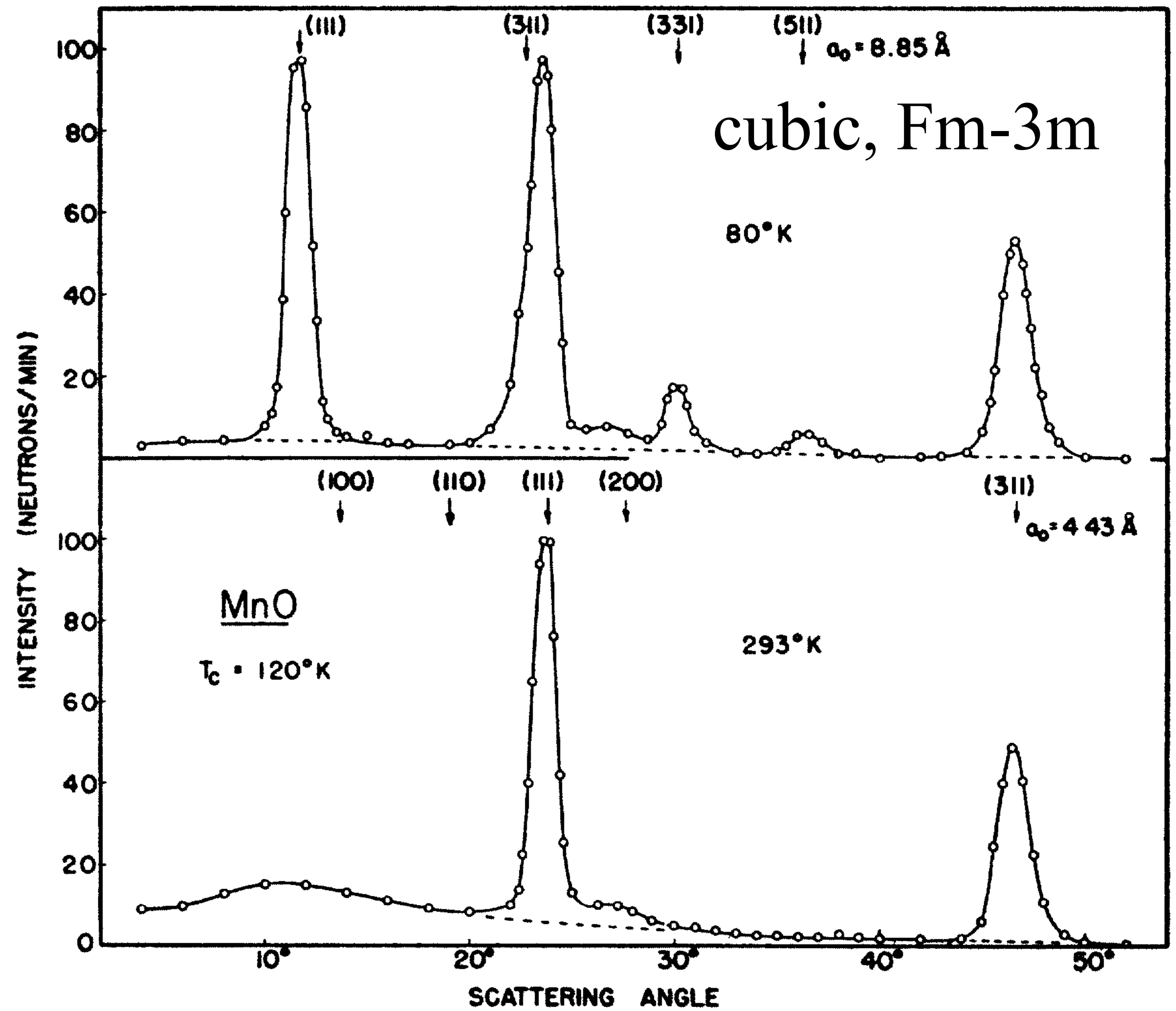


FIG. 4. Neutron diffraction patterns for MnO taken at liqui nitrogen and room temperatures. The patterns have been cor rected for the various forms of extraneous, diffuse scatterin mentioned in the text. Four extra antiferromagnetic reflection are to be noticed in the low temperature pattern.

Types of (magnetic) neutron diffraction techniques

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- spin-polarised:
  - nuclear/magnetic interference for non-spin flip. Purely magnetic for spin-flip channel.
  - Full 3D analysis of neutron polarization - spherical neutron polarimetry.

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- powder/single crystal
- λ=const: I(2θ), Time Of Flight TOF: I(t), Laue

Diffraction instruments at swiss continuous spallation source SINQ. λ=const

- HRPT - High Resolution Powder Diffractometer for Thermal Neutrons, λ=0.94 - 2.96 Å, High Q-range ≤11Å^{-1}
- DMC – High Intensity Powder Diffractometer for Cold Neutrons, λ=2.35 - 5.4 Å, High flux and good resolution at low and moderate Q ≤4Å^{-1}
- TriCS/Zebra - Single crystal diffractometer, λ=1.18, 2.3 Å, Thermal Neutrons
- TASP (triple axes) with MuPAD for polarised ND, Cold Neutrons, λ=1.8 - 6.0 Å

Literature on (magnetic) neutron scattering

Neutron scattering (general)

S.W. Lovesey, "*Theory of Neutron Scattering from Condensed Matter*", Oxford Univ. Press, 1987. Volume 2 for magnetic scattering. Definitive formal treatment

G.L. Squires, "*Intro. to the Theory of Thermal Neutron Scattering*", C.U.P., 1978, Republished by Dover, 1996. Simpler version of Lovesey.

All you need to know about magnetic neutron diffraction. Symmetry, representation analysis

Yu. A. Izyumov, V. E. Naish and R. P. Ozerov, "*Neutron diffraction of magnetic materials*", New York [etc.]: Consultants Bureau, 1991. Obsolete with respect to magnetic space groups and magnetic (super)symmetry.

Literature on magnetic neutron scattering

Modern way of magnetic symmetry and representation analysis

"*Magnetic superspace groups and symmetry constraints in incommensurate magnetic phases*", J M Perez-Mato, J L Ribeiro, V Petricek and M I Aroyo, J. Phys.: Condens. Matter **24** (2012) 163201

"*MAGNDATA: towards a database of magnetic structures.*"
Gallego, Perez-Mato, Elcoro, Tasci, Hanson, Momma, Aroyo & Madariaga
JOURNAL OF APPLIED CRYSTALLOGRAPHY (2016) Volume: 49 Pages: 1750-1776, 1941-1956

"Tabulation of irreducible representations of the crystallographic space groups and their superspace extensions"
Harold T. Stokes, Branton J. Campbell and Ryan Cordes
Acta Cryst. (2013). A69, 388–395

Web/computer resources to perform group theory symmetry analysis, in particular magnetic structures.

General tools for representation analysis, Shubnikov groups, 3D+n, and much more...
Web sites with a collection of software which applies group theoretical methods to the analysis of phase transitions in crystalline solids.

- Harold T. Stokes, Dorian M. Hatch, and Branton J. Campbell

  ISODISTORT: ISOTROPY Software Suite, http://iso.byu.edu

ISOTROPY Software Suite

Harold T. Stokes, Dorian M. Hatch, and Branton J. Campbell, Department of Physics and Astronomy, Brigham Young University, Provo, Utah 84606, USA,

M. I. Aroyo, J. M. Perez-Mato, D. Orobengoa, E. Tasci, G. de la Flor, A. Kirov

- Bilbao Crystallographic Server

bilbao crystallographic server http://www.cryst.ehu.es/

Computer programs to construct symmetry adapted magnetic structures and fit the experimental data.

Workhorses: Computer programs for representation analysis to be used together with the diffraction data analysis programs to determine magnetic structure from neutron diffraction (ND) experiment.

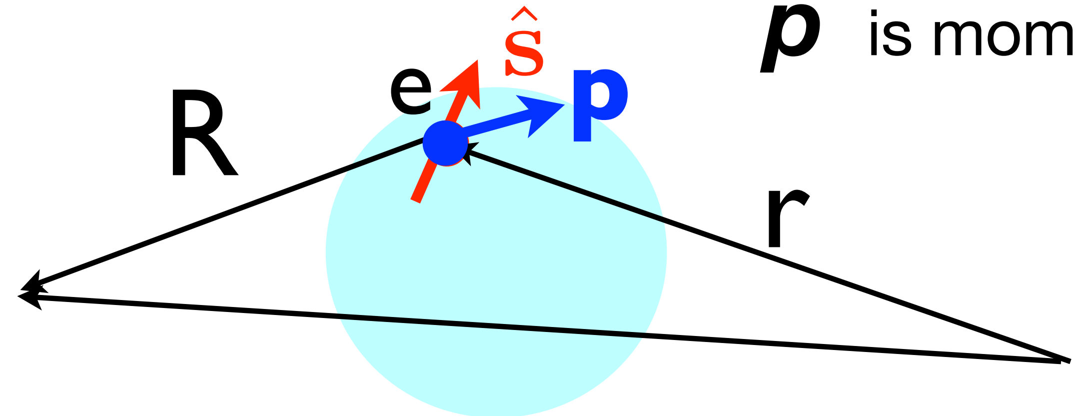
- Juan Rodríguez Carvajal (ILL) et al, http://www.ill.fr/sites/fullprof/
  `Fullprof` suite
- Vaclav Petricek, Michal Dusek (Prague) `Jana2006` `http://jana.fzu.cz/`

Basic principles of magnetic neutron diffraction

Magnetic neutron scattering on an atom

$\boldsymbol{\mu}_e = -2\mu_B \hat{\mathbf{s}}$

$\boldsymbol{p}$ is momentum



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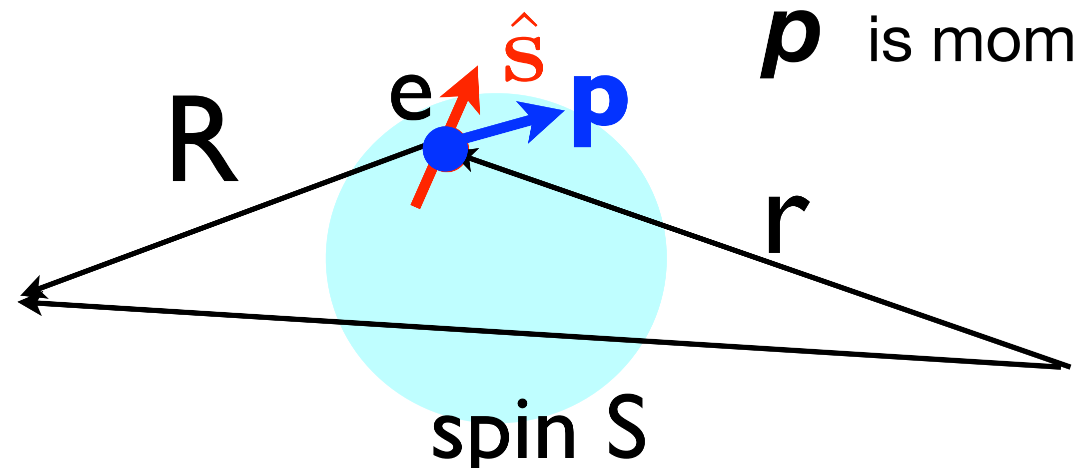

Magnetic field from an electron $$\mathbf{H}(\mathbf{R}) = \mathrm{rot}\left[\frac{\boldsymbol{\mu}_e \times \mathbf{R}}{|\mathbf{R}|^3}\right] - \frac{2\mu_B}{\hbar}\left[\frac{\mathbf{p}_e \times \mathbf{R}}{|\mathbf{R}|^3}\right]$$

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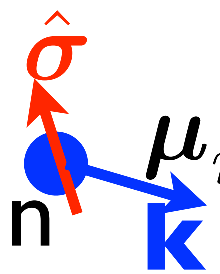

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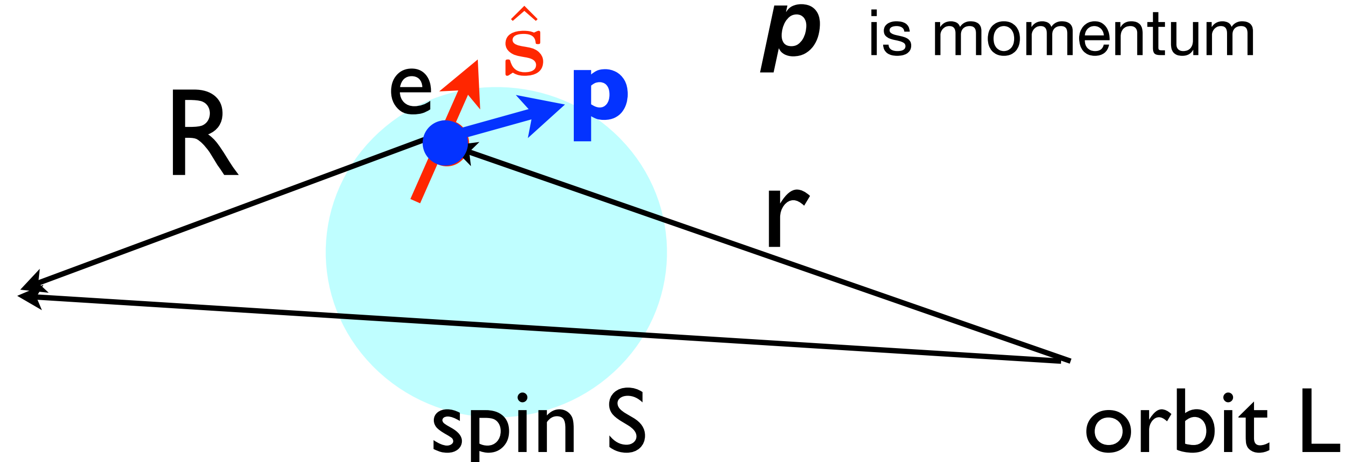

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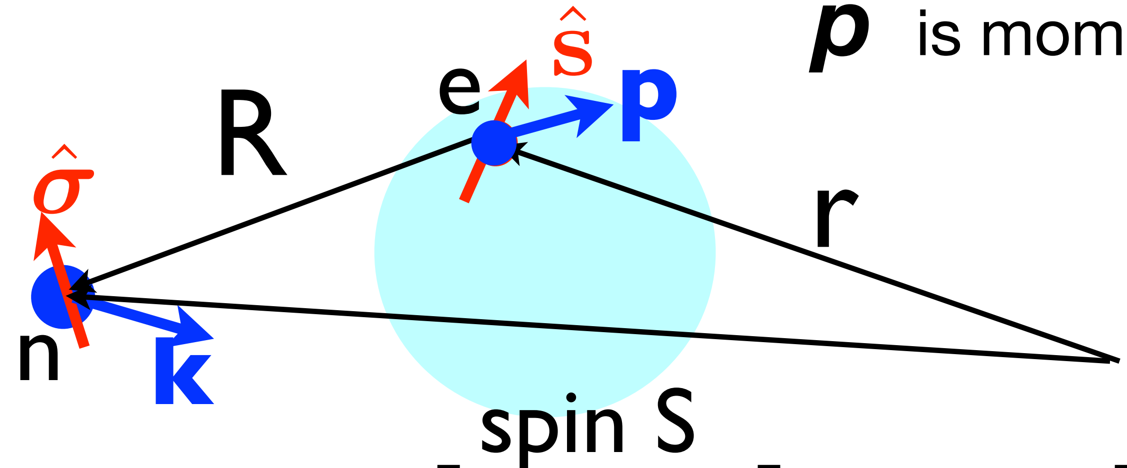

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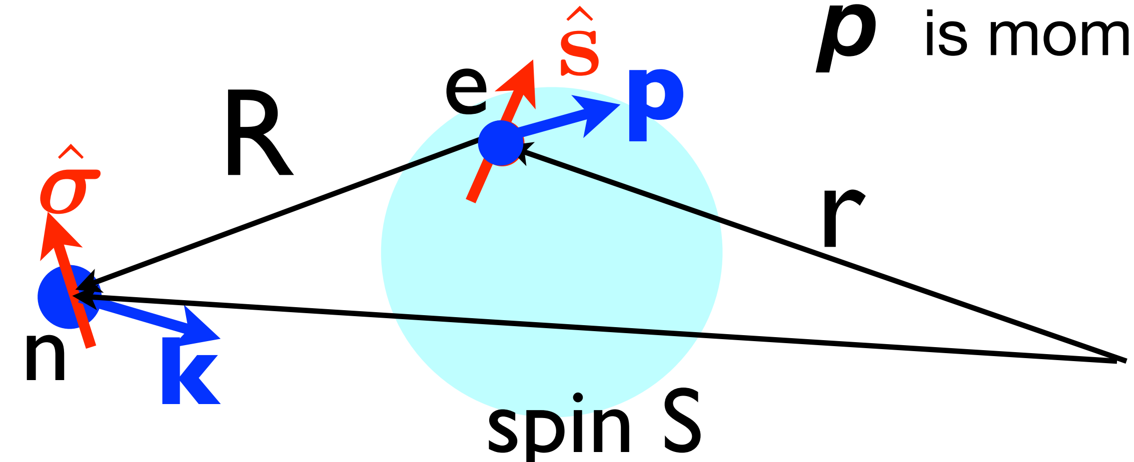

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averaging over neutron coordinates

$$\langle\mathbf{k'}|V(\mathbf{R})|\mathbf{k}\rangle = \gamma r_e\hat{\boldsymbol{\sigma}}\frac{1}{q^2}[\mathbf{q}\times[\hat{\mathbf{s}}e^{i\mathbf{q}\mathbf{r}}\times\mathbf{q}]]$$

$$\mathbf{q} = \mathbf{k}' - \mathbf{k}$$

Magnetic neutron scattering on an atom

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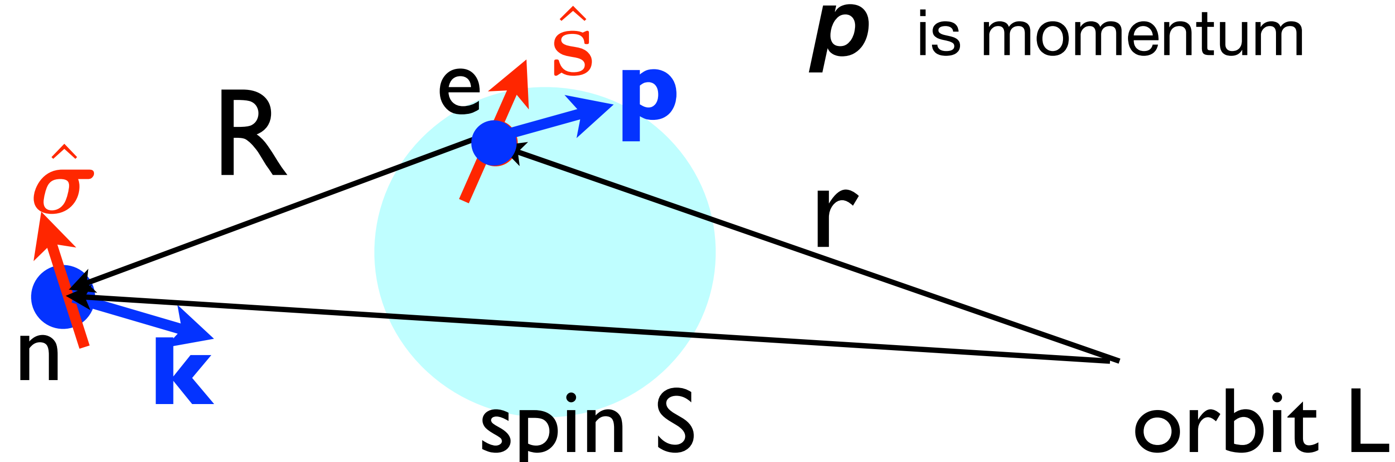

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magnetic interaction operator $\hat{\mathbf{Q}}_\perp$

Magnetic neutron scattering on an atom

$$\text{“magnetic scattering amplitude”} = \gamma r_e \left\langle \hat{\mathbf{Q}}_\perp \right\rangle ,$$

Magnetic neutron scattering on an atom

1. The size

$$\text{"magnetic scattering amplitude"} = \gamma r_e \left\langle \hat{\mathbf{Q}}_{\perp} \right\rangle ,$$

neutron magnetic moment in μ_n -1.91

classical electron radius $\frac{e^2}{mc^2}$

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$$\gamma r_e = -0.54 \cdot 10^{-12} \mathrm{cm} = -5.4\,\mathrm{fm} (\times S)$$

fm=fermi=10^{-13} cm

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x-ray scattering length: Zr_e

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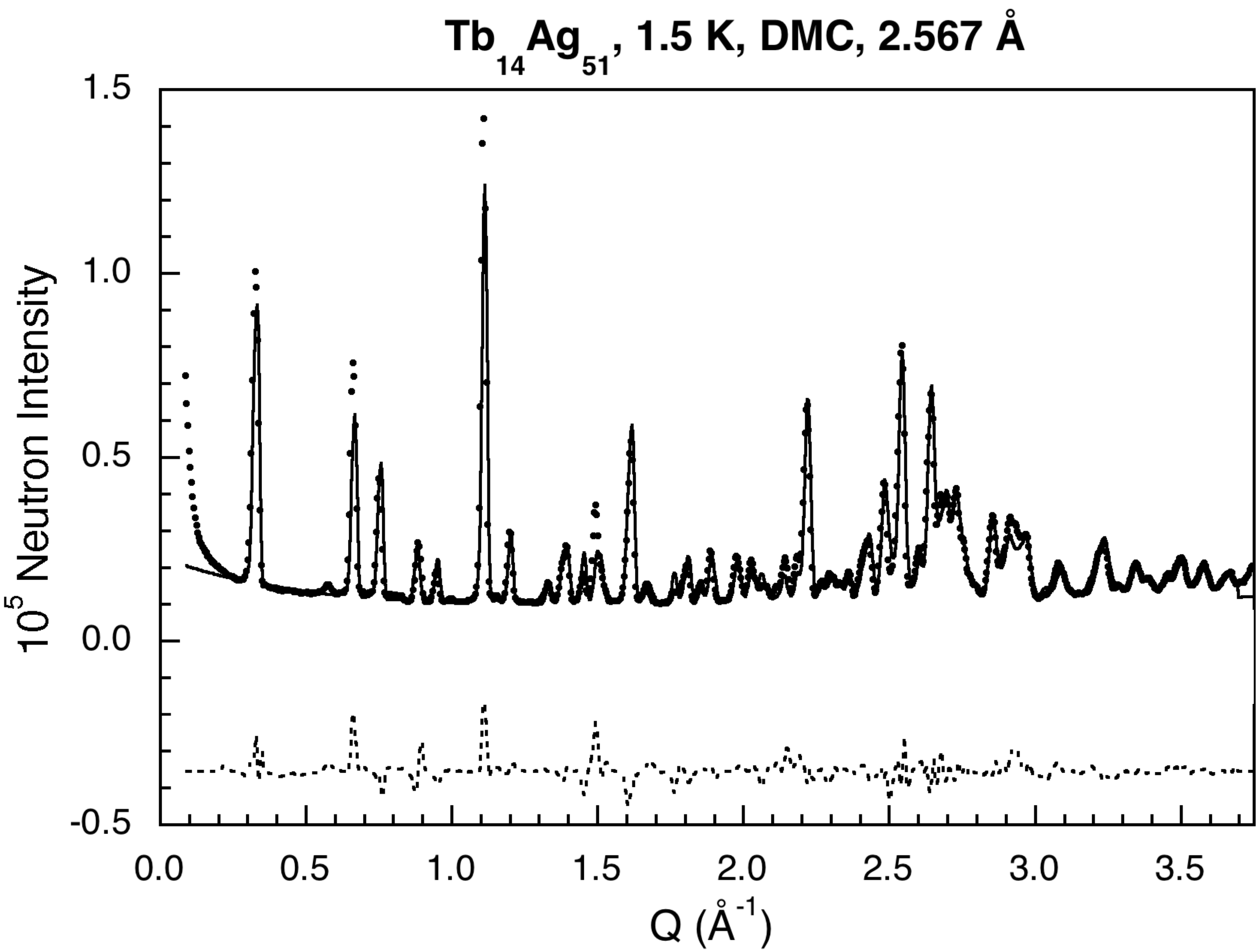
Comparison of neutron scattering lengths (fm)

magnetic

Mn^{3+} (S=2): -10.8, Cu^{2+} (S=½): -2.65

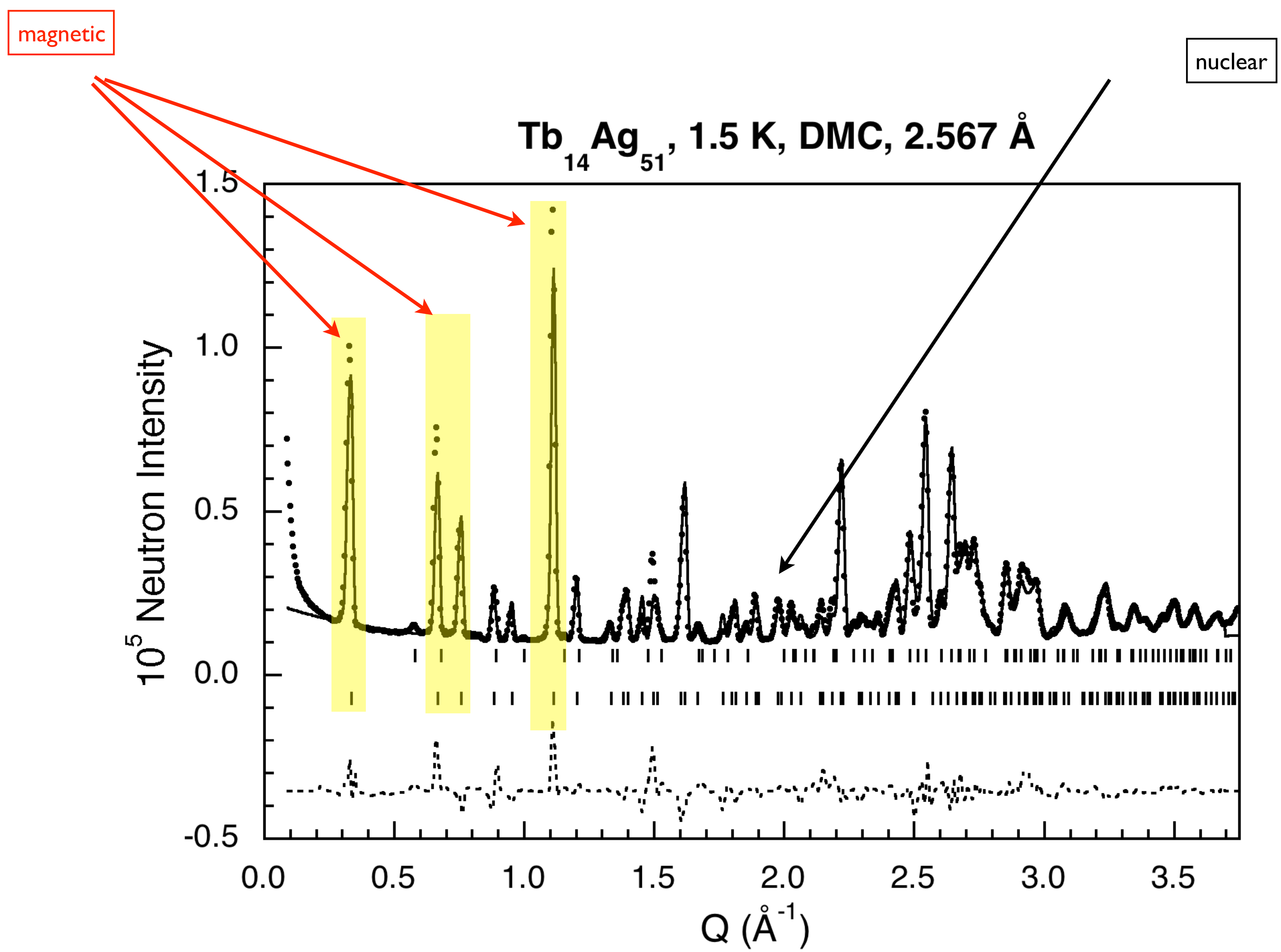
nuclear

Mn : -3.7, Cu: 7.7



Tb14Ag51, 1.5 K, DMC, 2.567 Å
10^5 Neutron Intensity
1.5
1.0
0.5
0.0
-0.5
0.0
0.5
1.0
1.5
2.0
2.5
3.0
3.5
Q (Å^-1)

magnetic scattering intensity can be larger than the nuclear one



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Magnetic neutron scattering on an atom

2. q-dependence

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$$\frac{1}{q^2}[\mathbf{q} \times \hat{\mathbf{Q}} \times \mathbf{q}]$$

Magnetic neutron scattering on an atom

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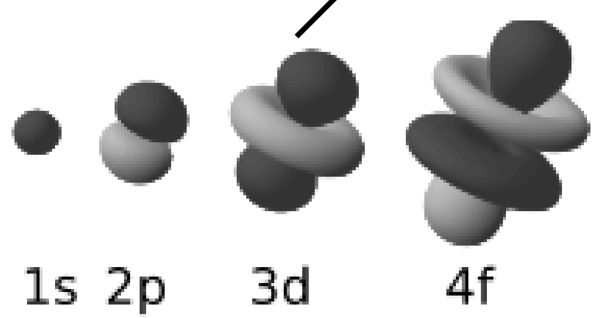
$$\frac{1}{q^2}[\mathbf{q} \times \hat{\mathbf{Q}} \times \mathbf{q}]$$

$$\langle \hat{\mathbf{Q}} \rangle = \langle \psi | \, \hat{\mathbf{s}} e^{i\mathbf{qr}} \, | \psi \rangle \simeq \mathbf{S} \int d\mathbf{r} \rho_s(\mathbf{r}) e^{i\mathbf{qr}}$$



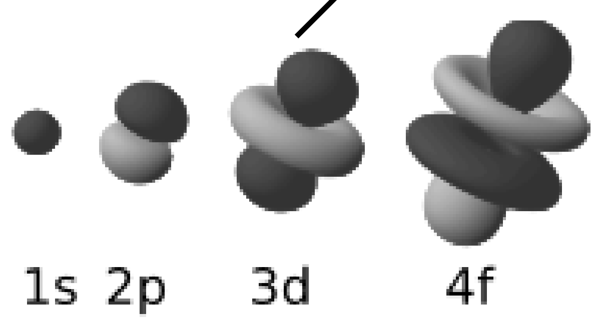
Magnetic neutron scattering on an atom

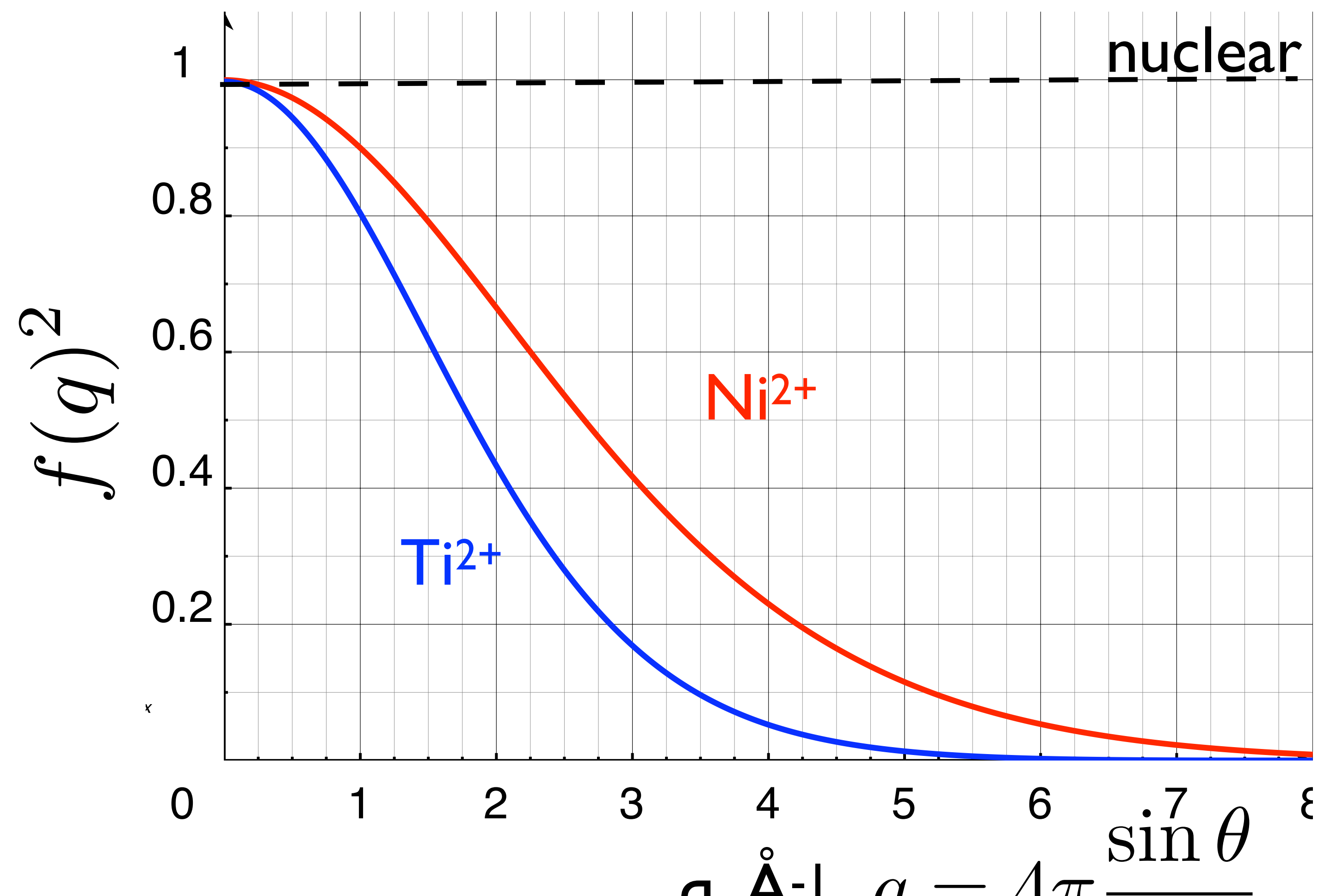
2. q-dependence

"magnetic scattering amplitude" $= \gamma r_e \left\langle \hat{\mathbf{Q}}_\perp \right\rangle$,

Fourier image of the spin density in atom
or magnetic form-factor

$$\langle \hat{\mathbf{Q}} \rangle = \langle \psi | \, \hat{\mathbf{s}} e^{i\mathbf{qr}} \, | \psi \rangle \simeq \mathbf{S} \int d\mathbf{r} \rho_s(\mathbf{r}) e^{i\mathbf{qr}} = \mathbf{S} f(q)$$





$q = 4\pi \frac{\sin\theta}{\lambda}$

Magnetic neutron scattering on an atom

$$\text{"magnetic scattering amplitude"} = \gamma r_e \left\langle \hat{\mathbf{Q}}_{\perp} \right\rangle$$

$$\mathbf{Q}_{\perp} = \tilde{\mathbf{q}} \times \mathbf{Q} \times \tilde{\mathbf{q}} \quad = [\tilde{\mathbf{q}} \times \mathbf{S} \times \tilde{\mathbf{q}}] f(q)$$

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3. geometry

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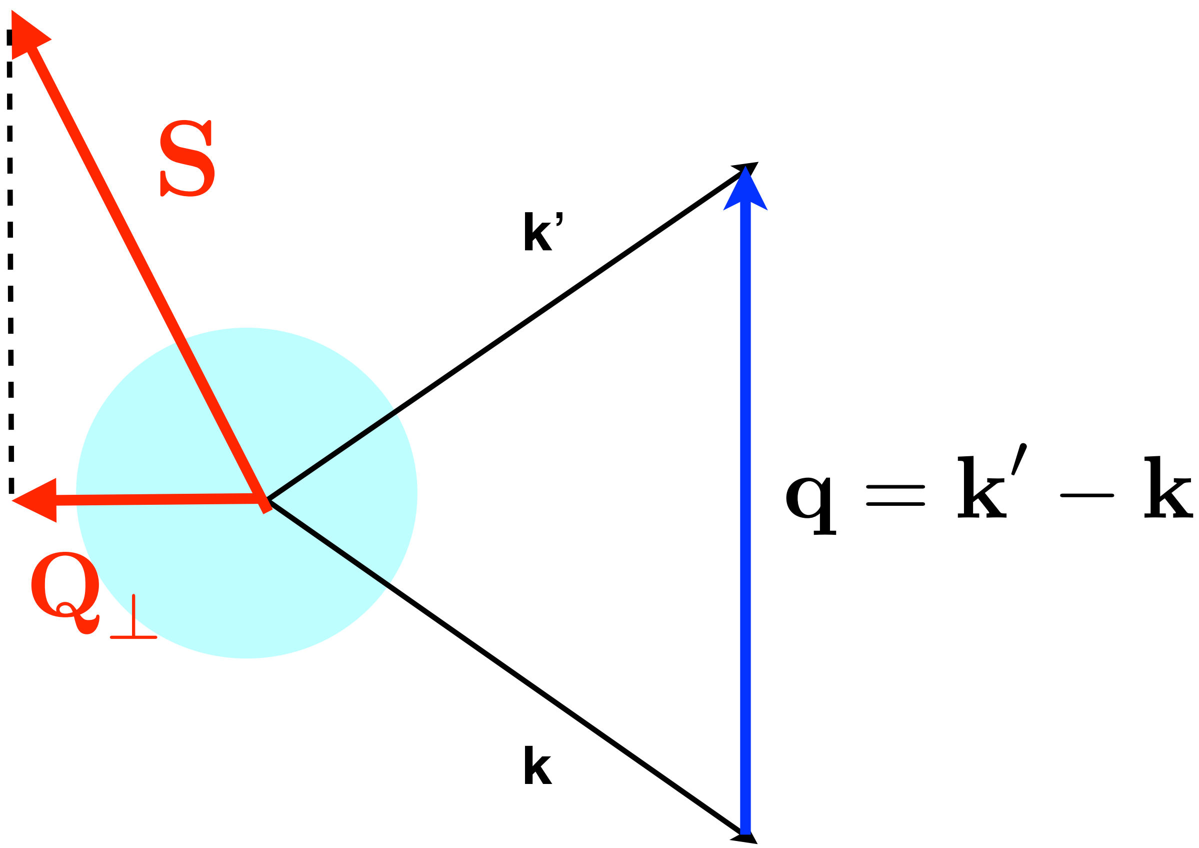

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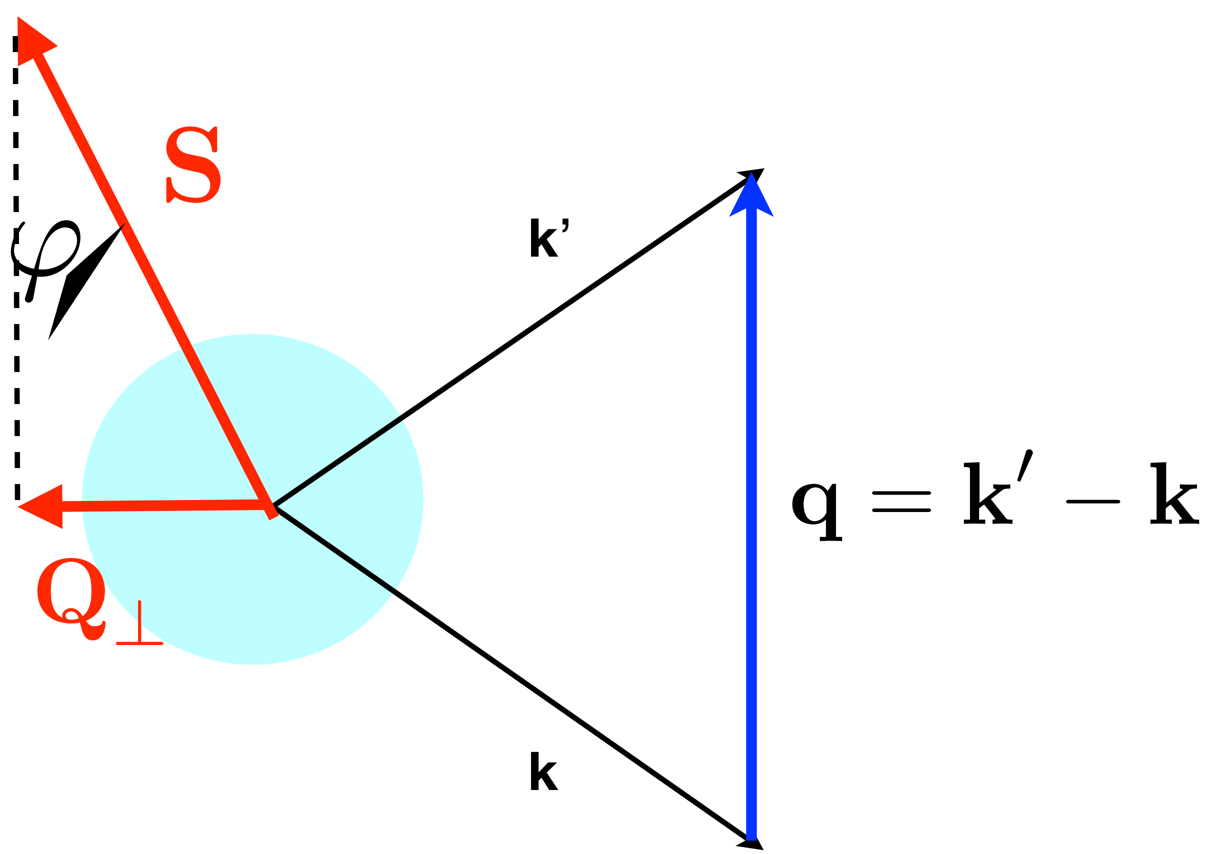

$$|\mathbf{Q}_\perp| = |\mathbf{S}| \sin(\varphi)$$

Elastic scattering intensity

Neutron scattering cross-section (for unpolarised neutron beam)

$$\frac{d\sigma}{d\Omega} \propto |\, \langle \hat{\mathbf{Q}}_{\perp} \rangle \,|^2$$

Magnetic order parameters overview

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Term **FerroMagnetism (FM)**: lodestone in Greek writings by the year 800 B.C., magnetit FeO-Fe203 used as compass. First “theory” Rene Descartes (1596-1650)

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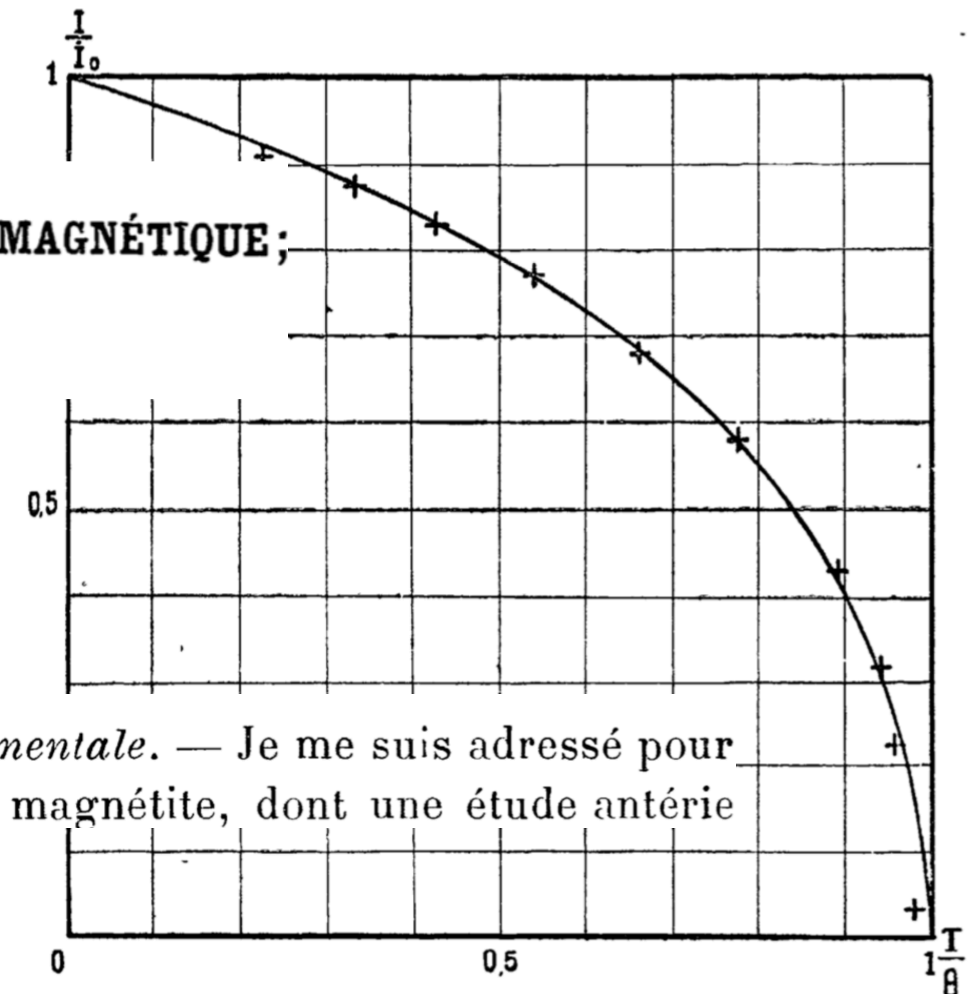
L’HYPOTHÈSE DU CHAMP MOLÉCULAIRE ET LA PROPRIÉTÉ FERROMAGNÉTIQUE;

Par M. PIERRE WEISS (1).

Submitted on 1 Jan 1907



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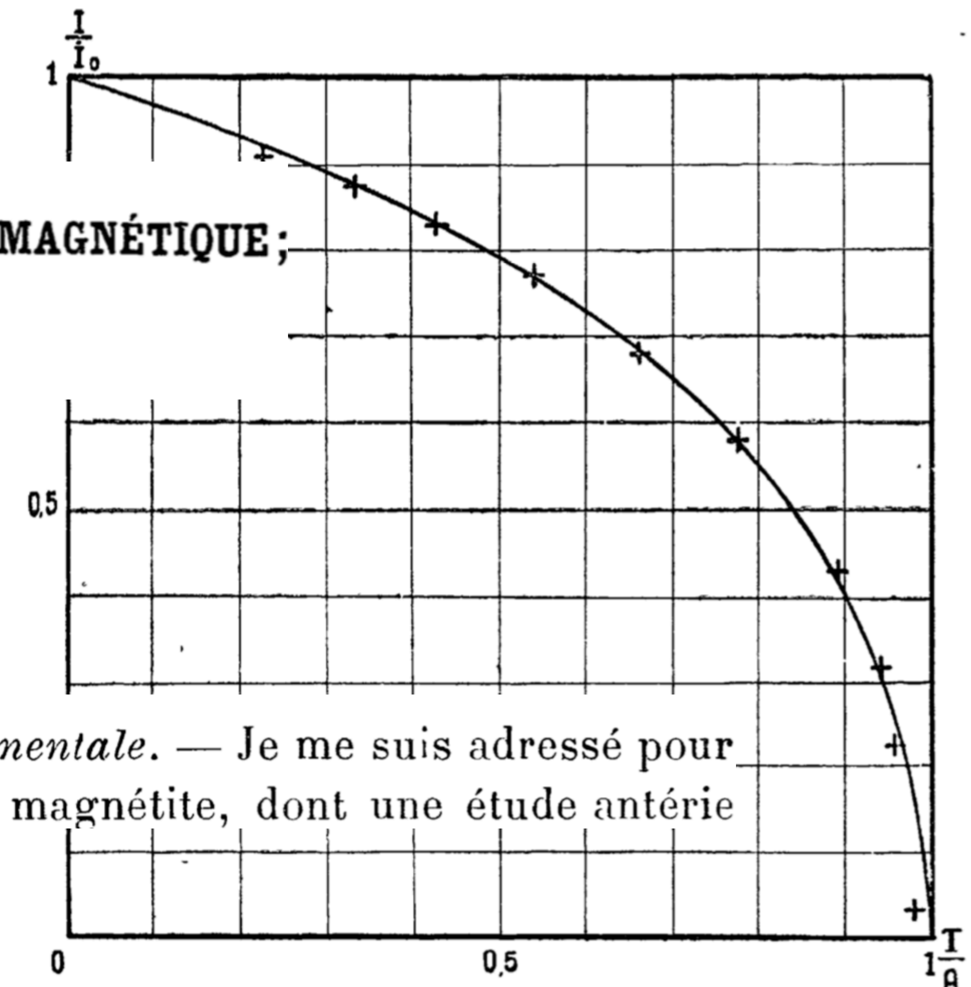

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FM is not possible in classical physics! Bohr–van Leeuwen theorem was discovered by Niels Bohr in 1911 in his doctoral dissertation

QM 1925-27

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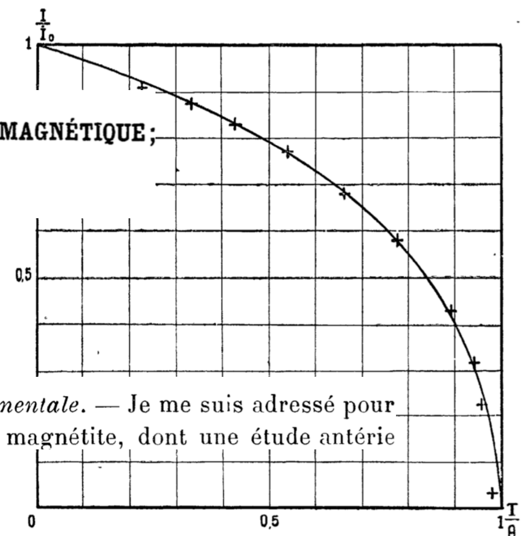

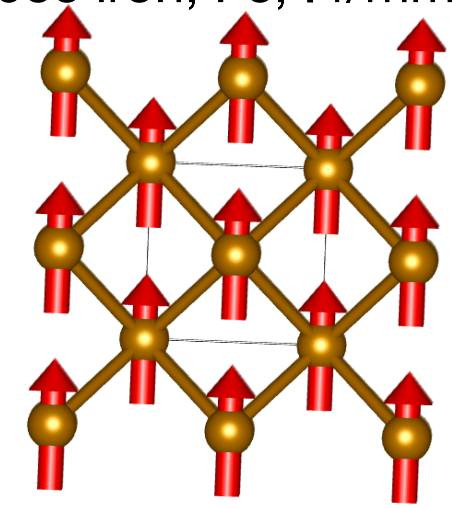

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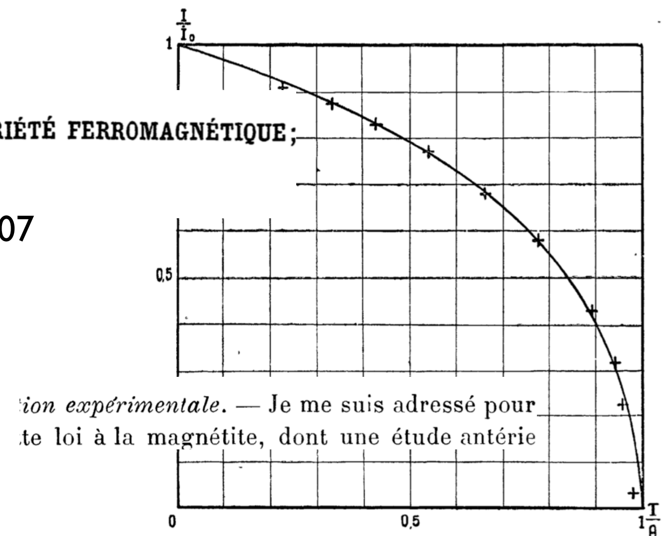

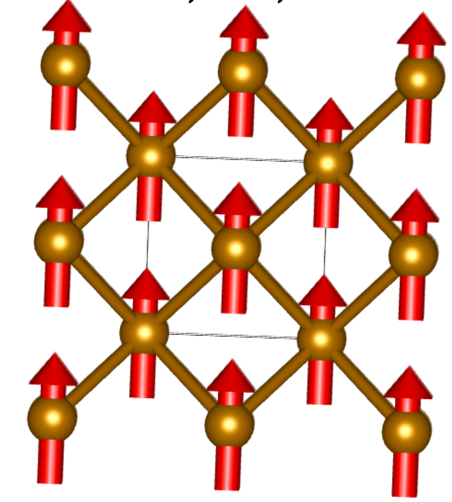

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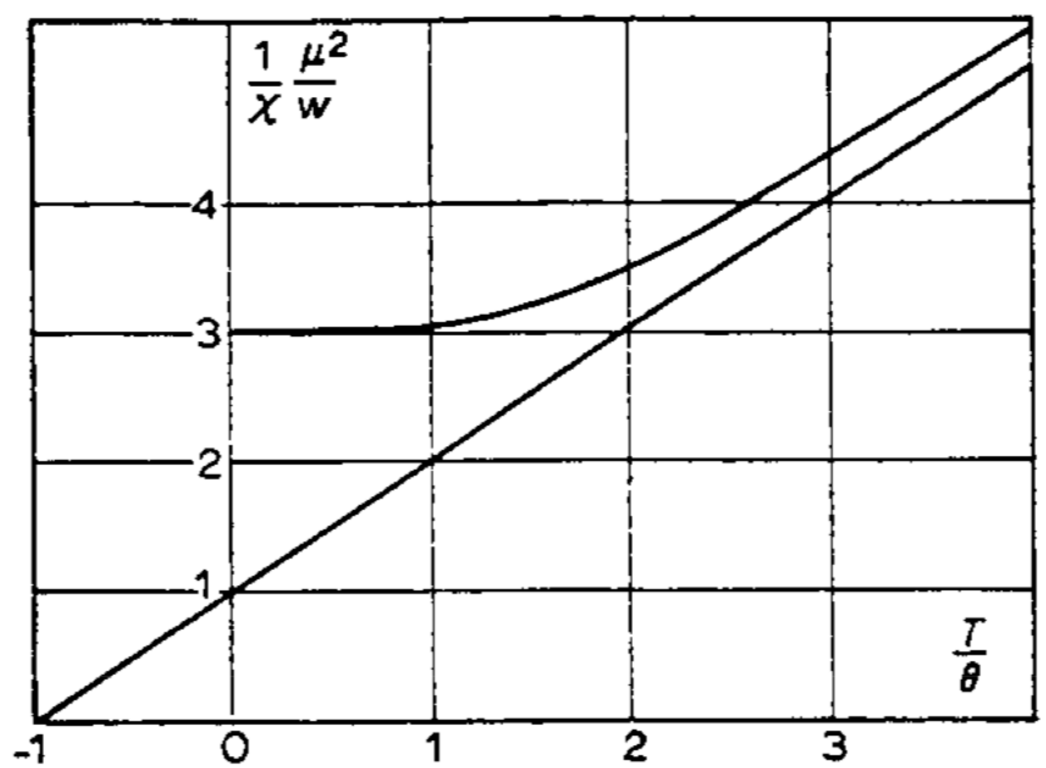

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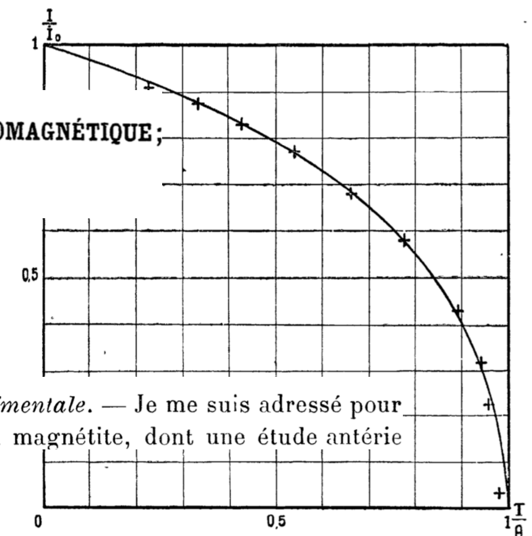

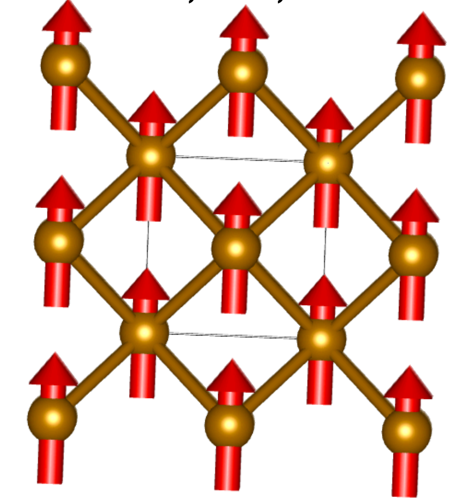

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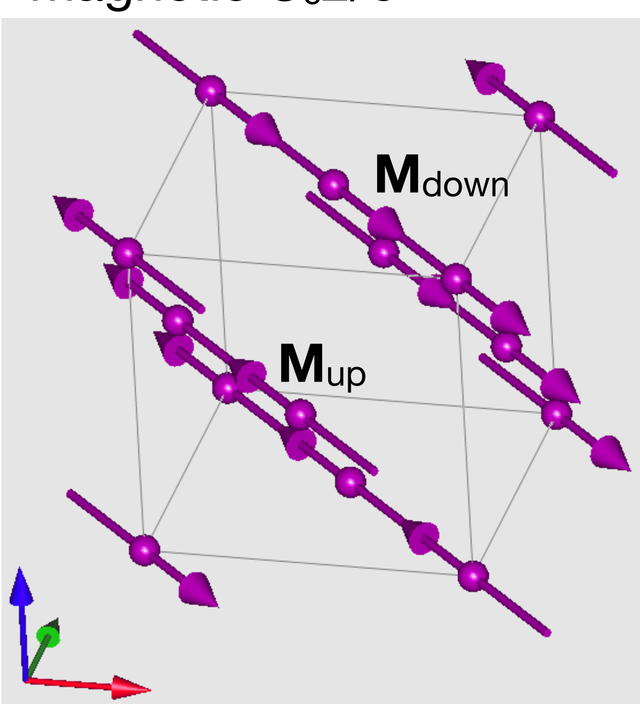
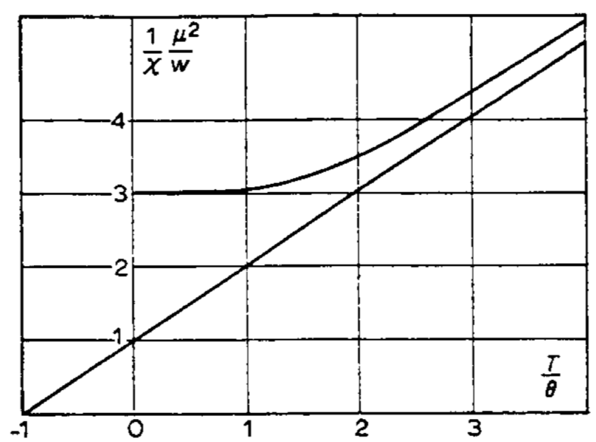


Order parameter: sub-lattice magnetisations $\mathbf{M}_{up}$ $\mathbf{M}_{down}$ are not directly seen macroscopically



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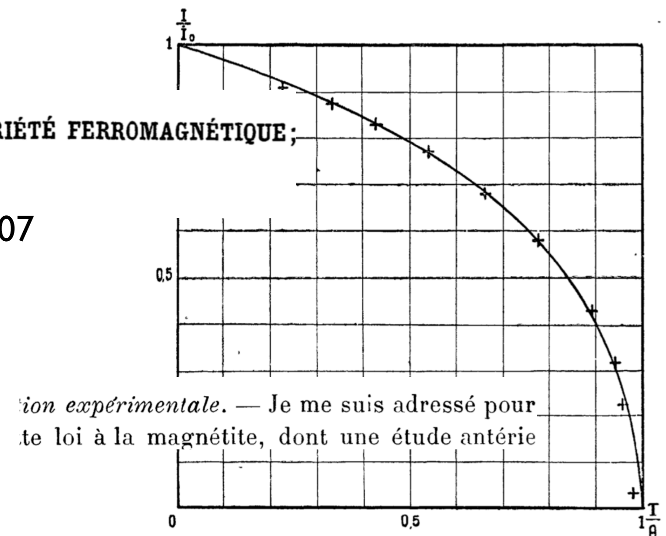

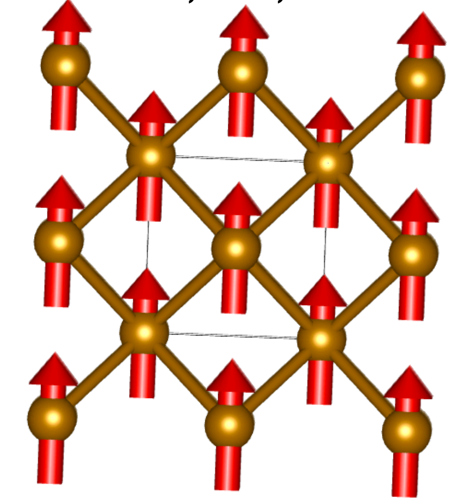

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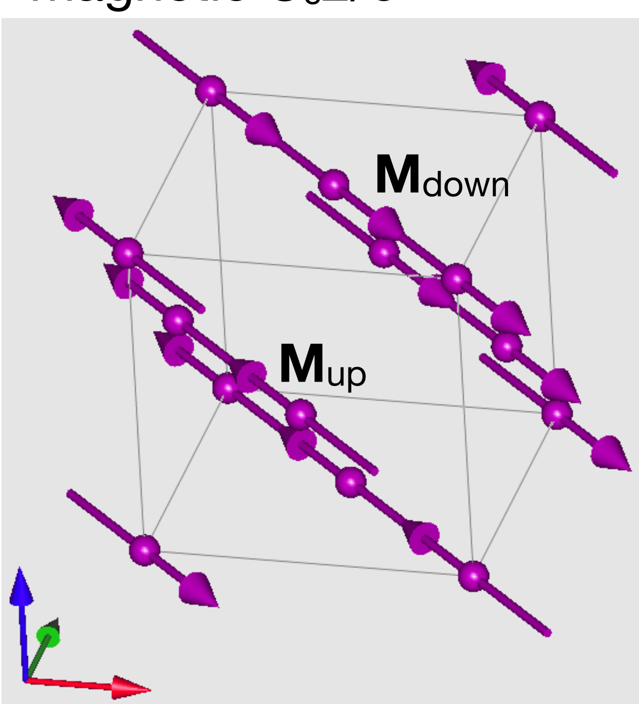
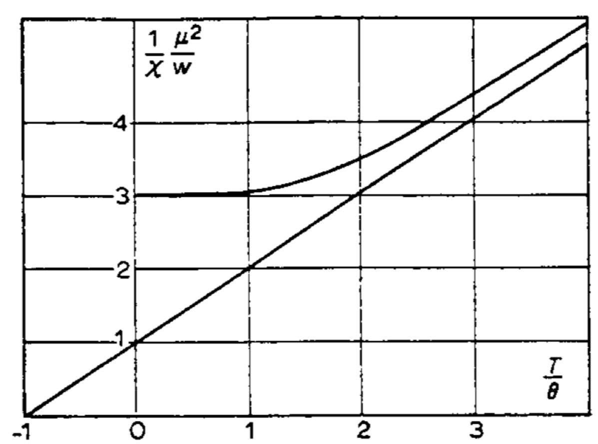
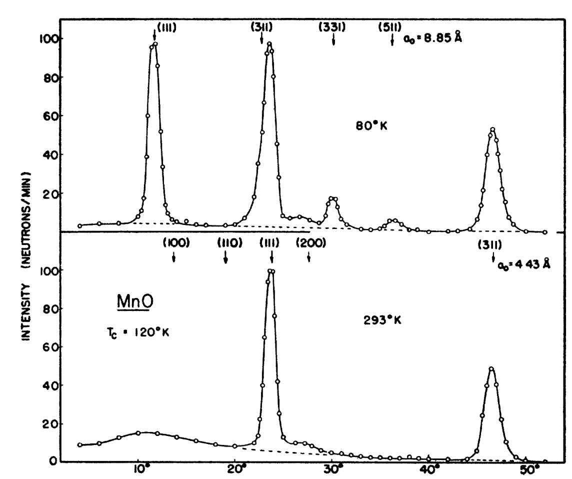
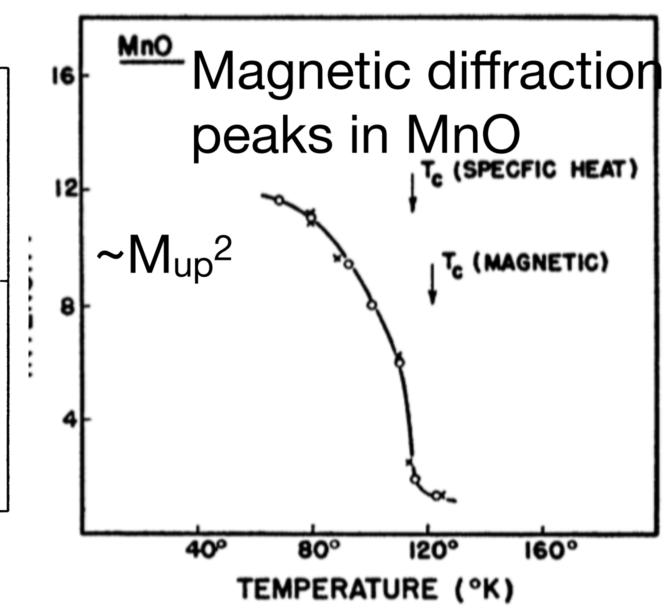
Neutron diffraction: one of the direct techniques Shull, et al 1951



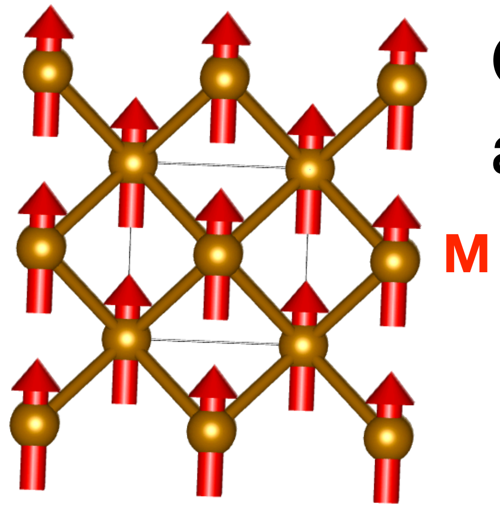






Magnetic order parameters overview

FerroMagnetism (FM), AntiFerroMagnetism (AFM), Ferrimagnetism, …



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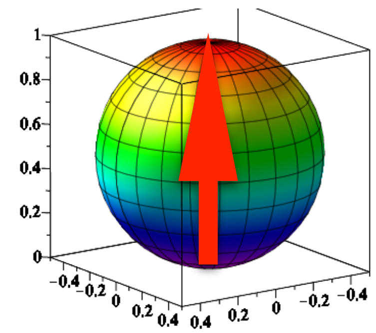

spherically symmetric distribution of moment density

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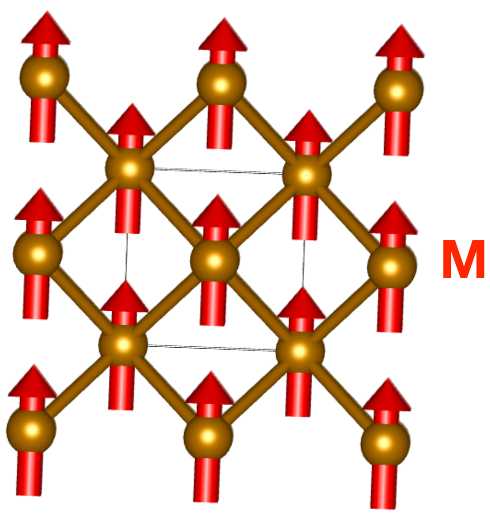

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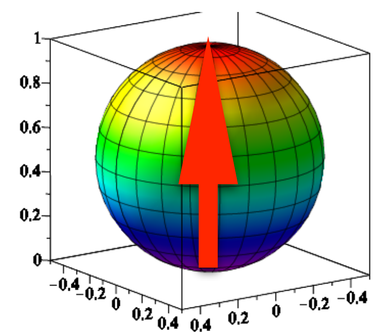

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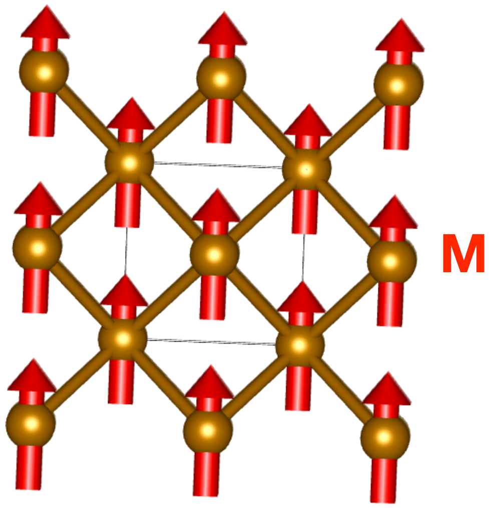

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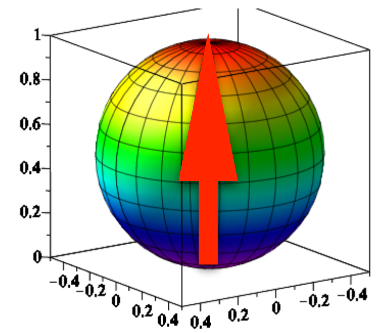

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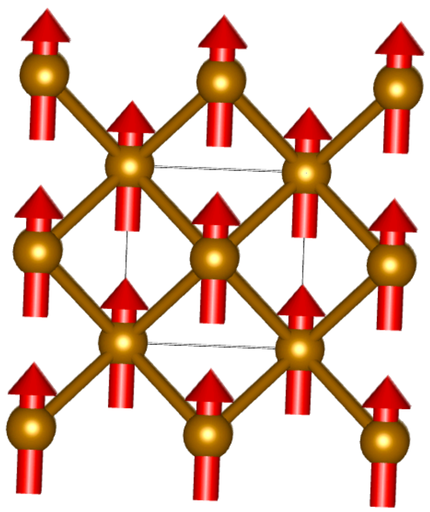

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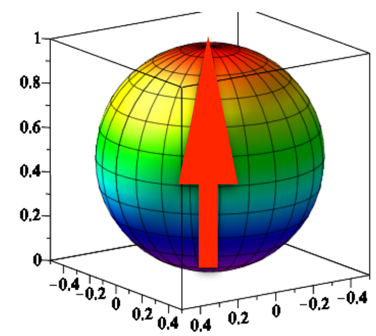

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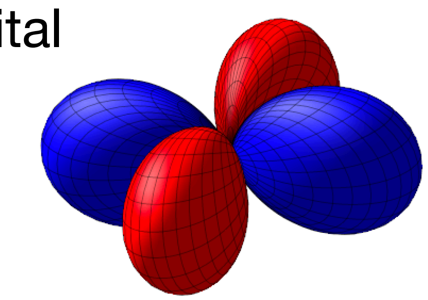
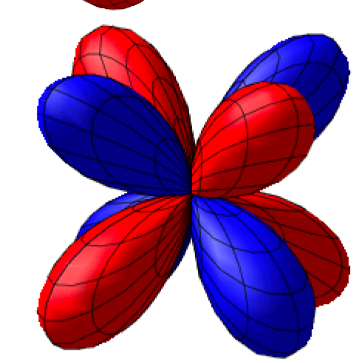
d-orbital Y_{22}

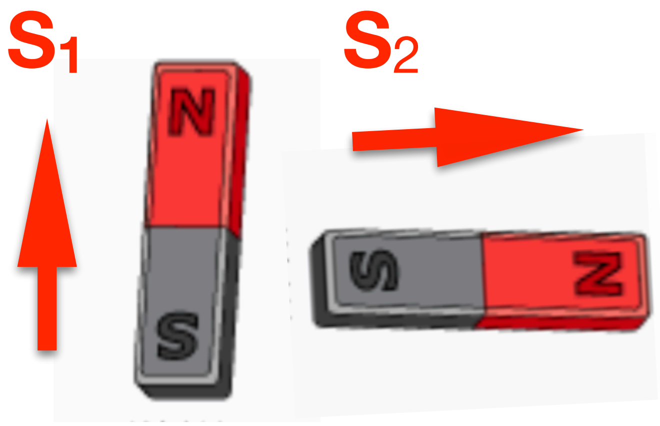


f-orbital Y_{32}



magnetic multipole moments: 3D-tensors of rank R

Dipole, tensor of rank R=1: 2^1=2 charges



... n-dipoles

$$M_i = \sum_n S_{ni} \qquad \begin{pmatrix} 1 \\ 1 \\ 0 \end{pmatrix}$$

$i, j, k... = 1, 2, 3(x, y, z)$

n runs over all dipoles

nomenclature is Greek 2^R-numbers

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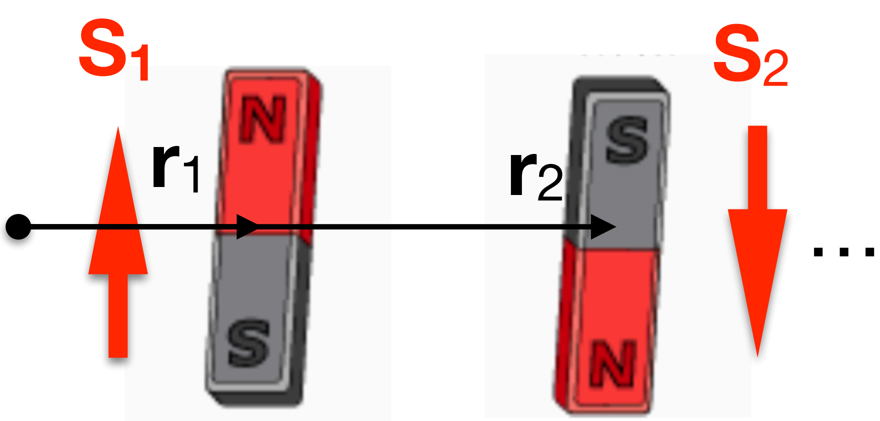
Quadrupole, tensor of rank R=2: 2^R=2^2=4 charges



$$M_{ij} = \sum_n S_{ni} r_{nj} \qquad \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

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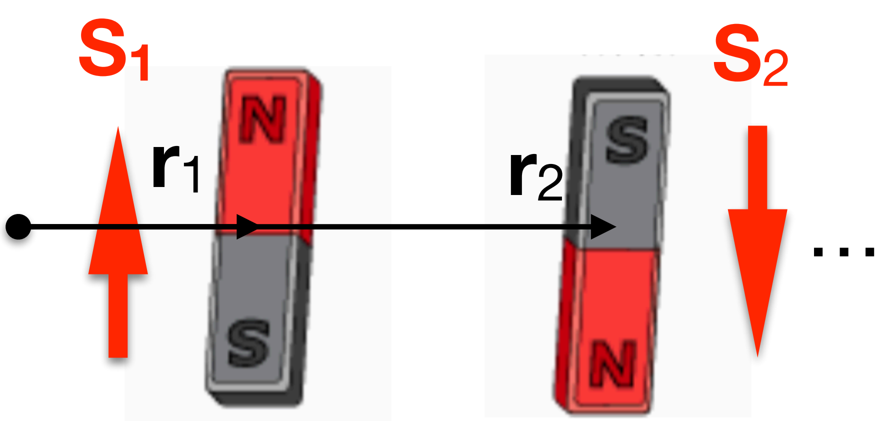

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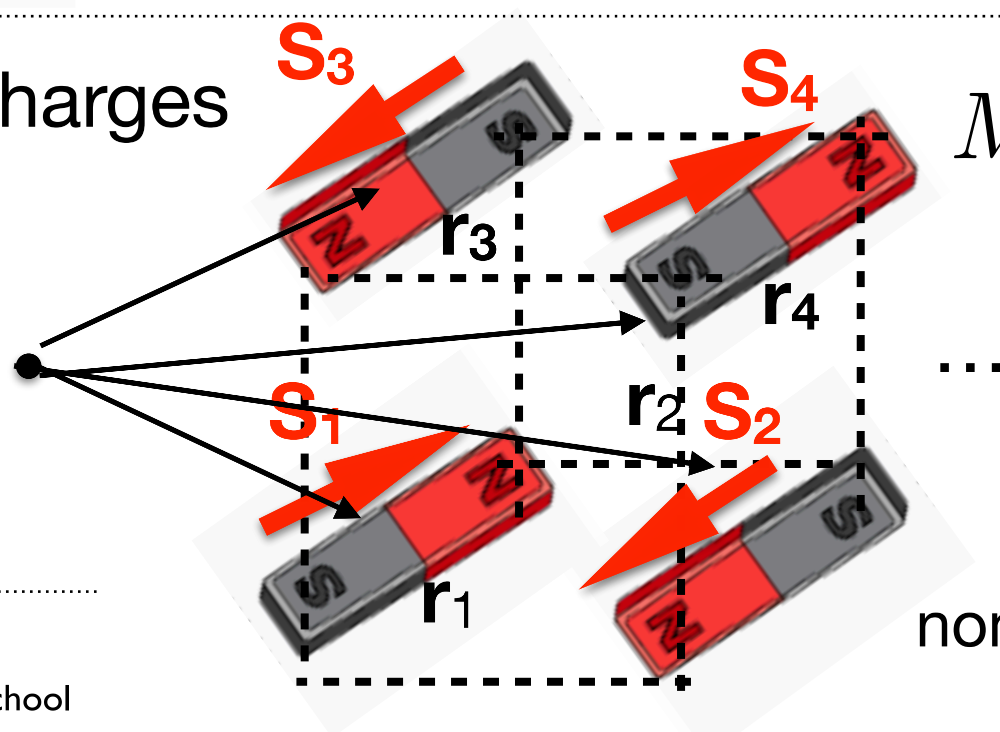
Octupole, R=3: $2^3=8$ charges



$$M_{ijk} = \sum_n S_{ni} r_{nj} r_{nk}$$

…

Hexadecapole R=4…

nomenclature is Greek 2^R-numbers

Magnetic order parameters overview

Magnetic-octupoles ordering

PHYSICAL REVIEW B 72, 144401 2005

NpO_2

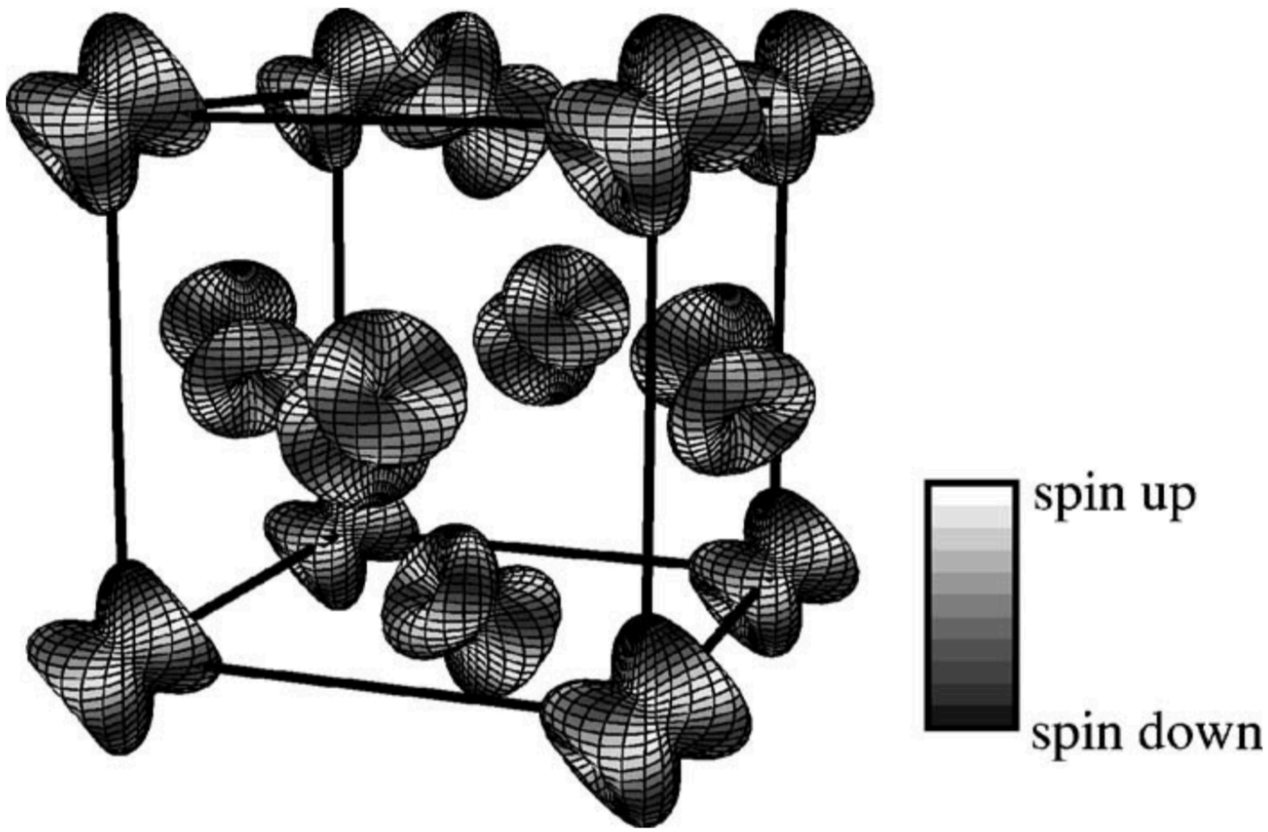


FIG. 7. The triple-**q** Γ_{5u} octupole state. The surface is defined by $=[\Sigma_\sigma|\psi(\theta,\phi,\sigma)|^2]^{1/2}$ in the polar coordinates, when the $5f$ wave unction is represented by $\Psi(r,\theta,\phi,\sigma)=R(r)\psi(\theta,\phi,\sigma)$, where σ enotes real spin. White shift of the surface indicates the increase of

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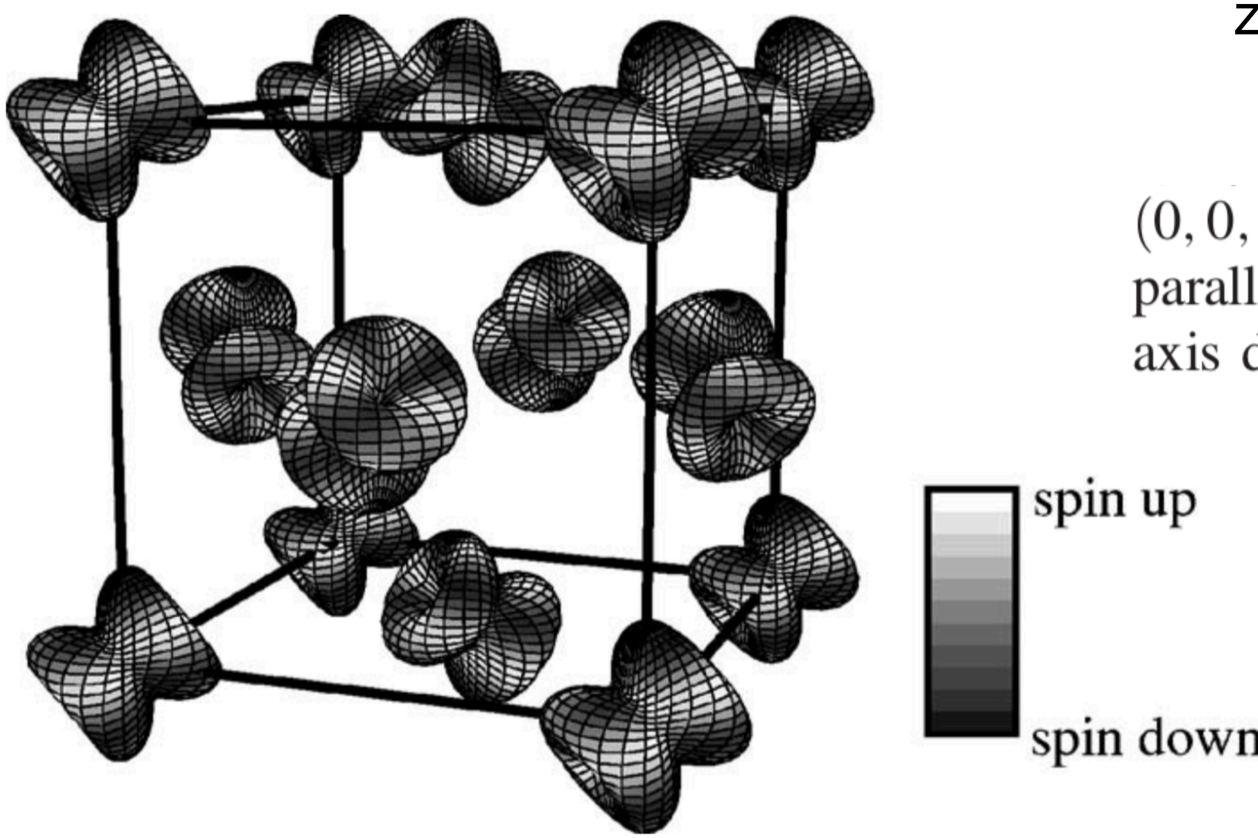
PHYSICAL REVIEW B 72, 144401 2005 NpO$_2$



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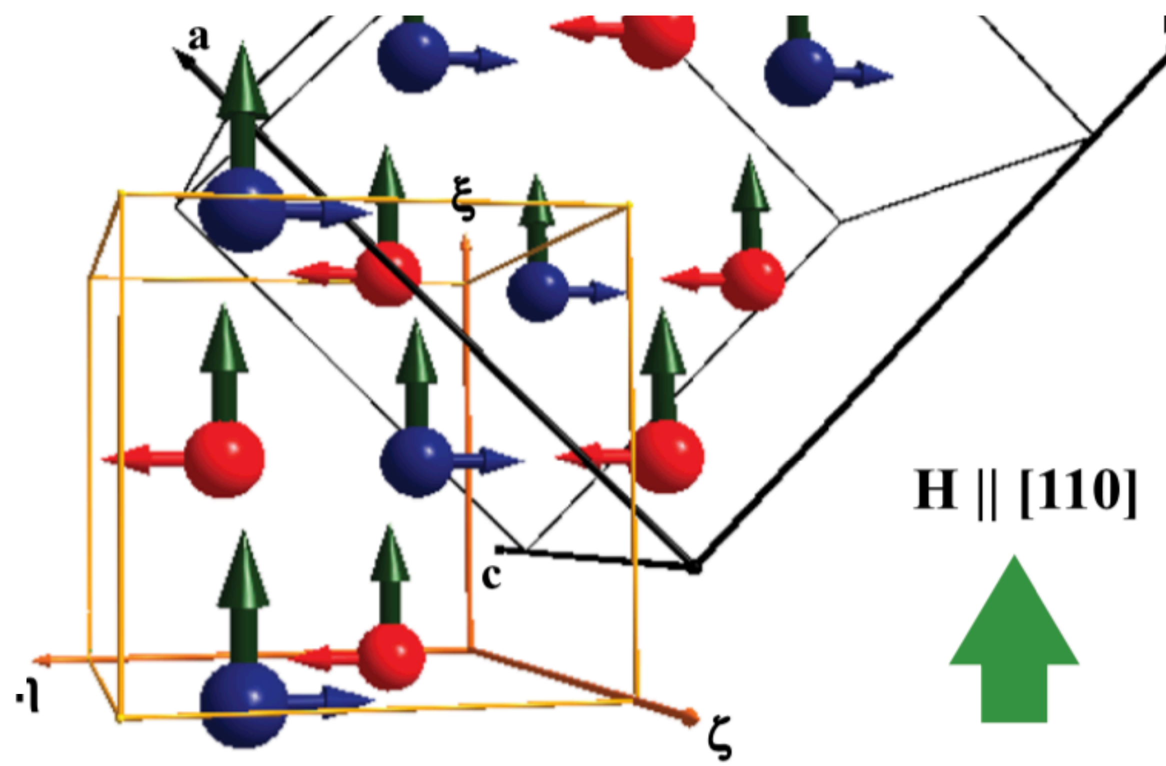
New exotics!

Dirac magnetoelectric dipole (anapole) in zero-magnetization ferromagnet Sm$_{0.97}$6Gd$_{0.02}$4Al$_2$

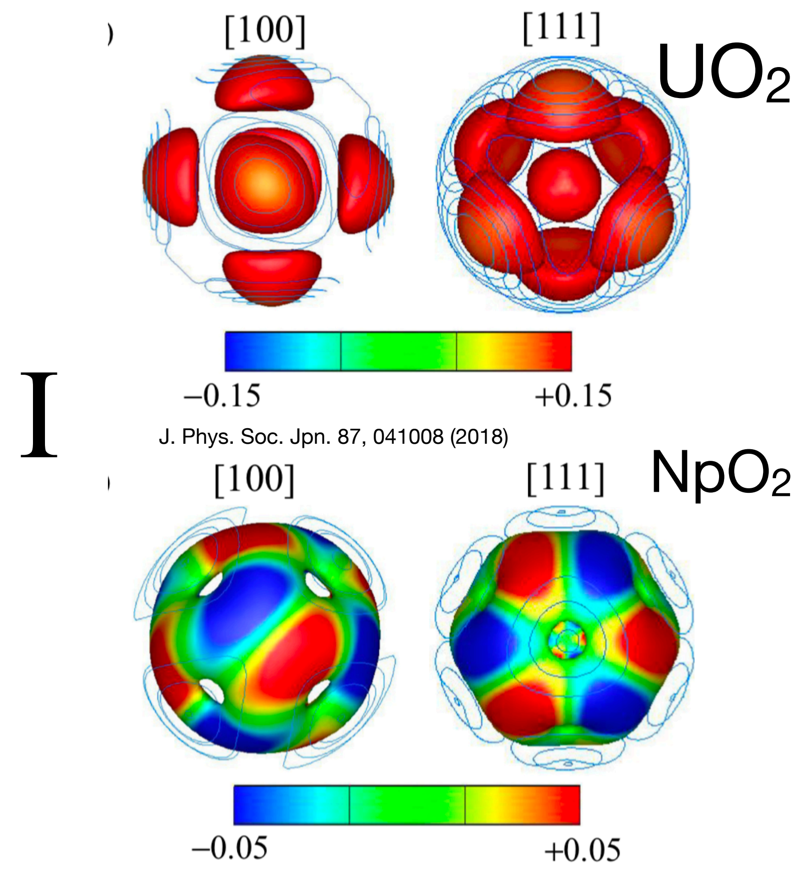
S W Lovesey et al PRL 122, 047203 (2019)

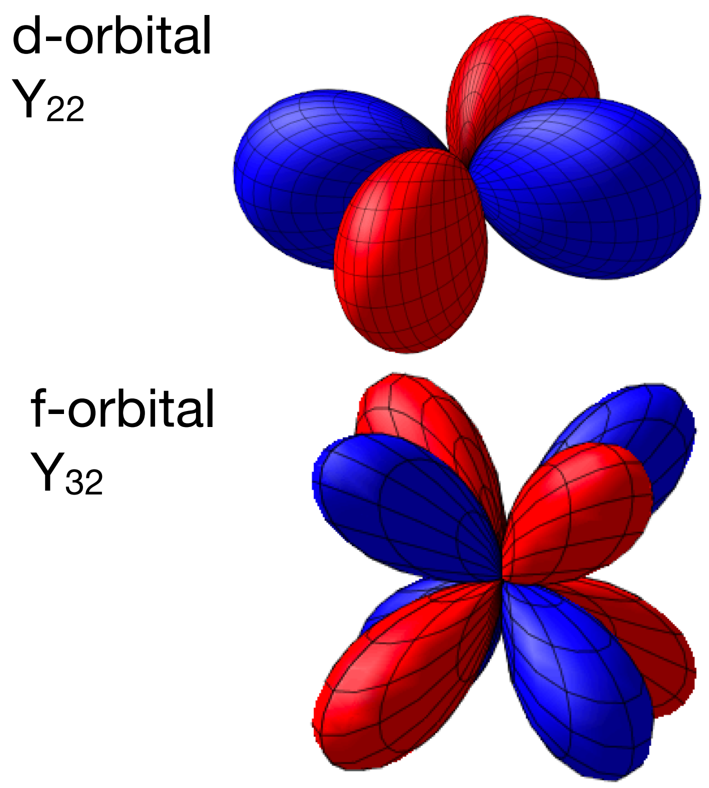
$(0,0,-1)$ outlined in yellow. Green arrows are axial dipoles parallel to the ξ axis, while blue and red arrows that lie along the η axis denote anapoles related by point inversion.



Magnetic objects neutrons sensitive to

sketch of multipole expansion, octupoles





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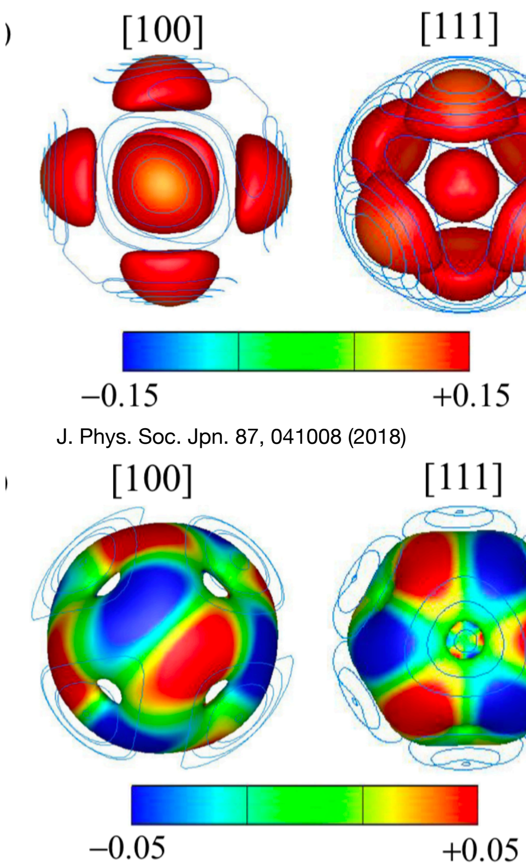
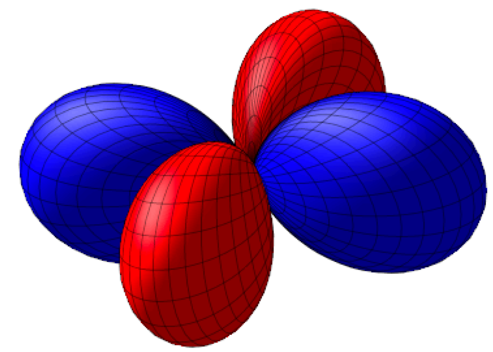


J. Phys. Soc. Jpn. 87, 041008 (2018)

UO_2

NpO_2

d-orbital
Y_{22}



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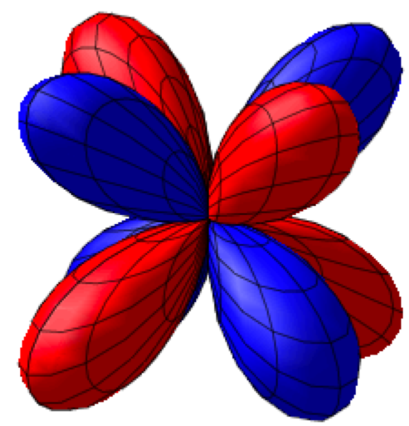
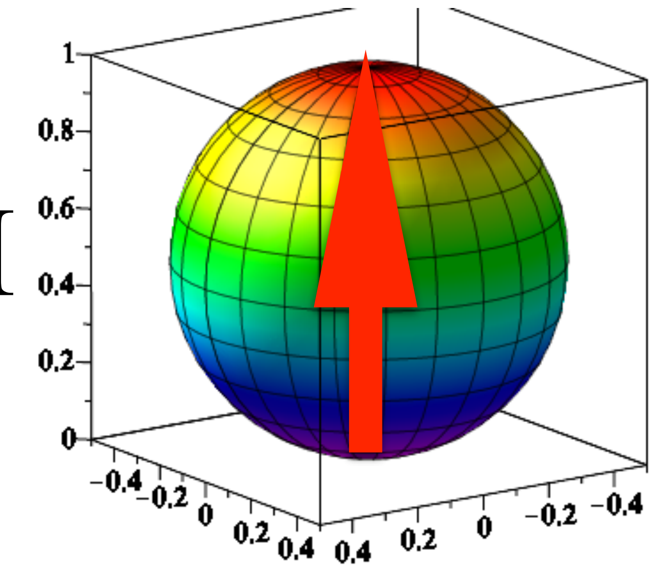


Dipole approximation

II



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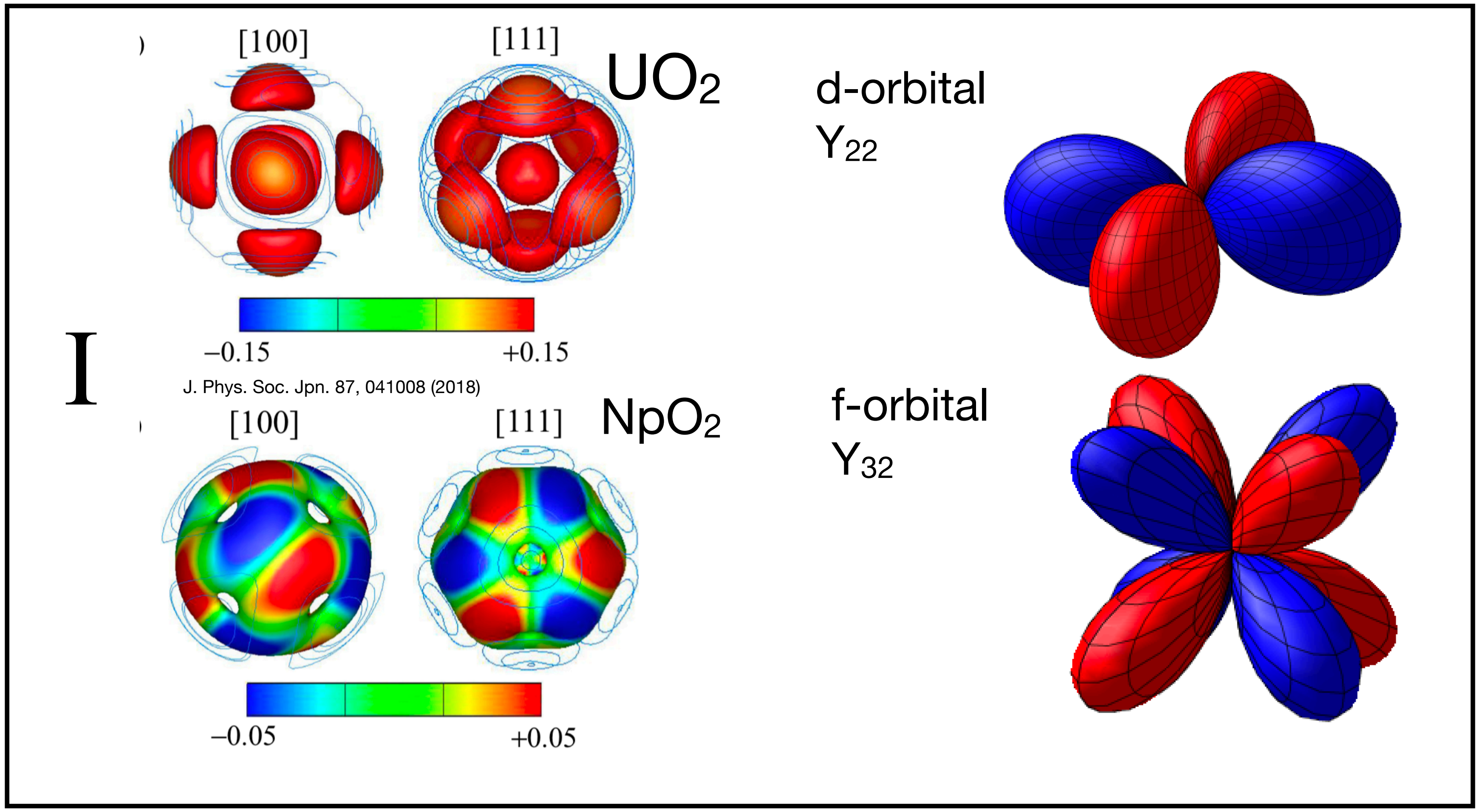

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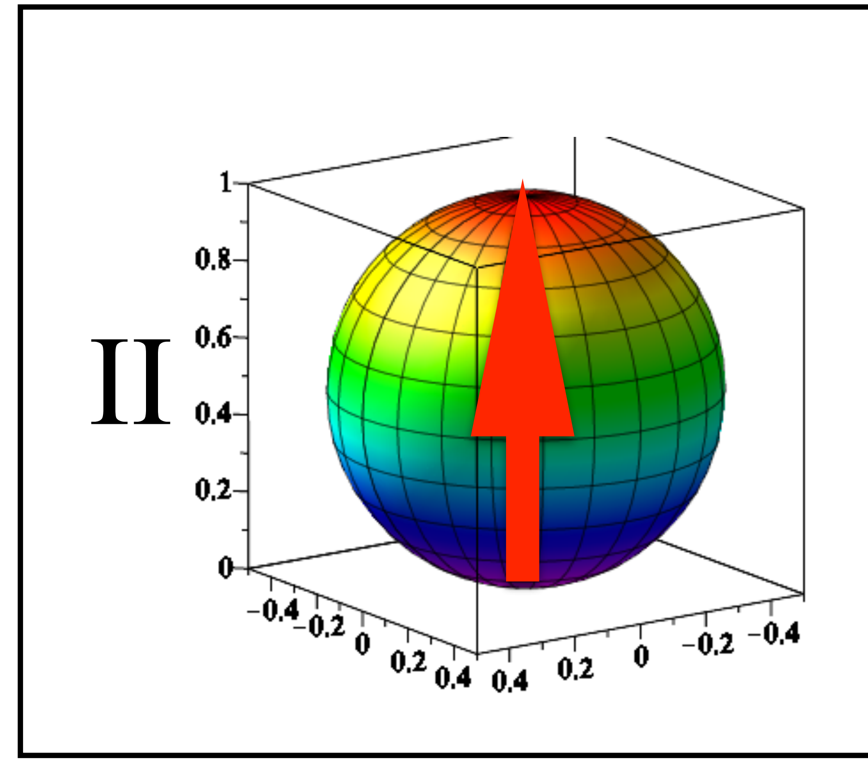
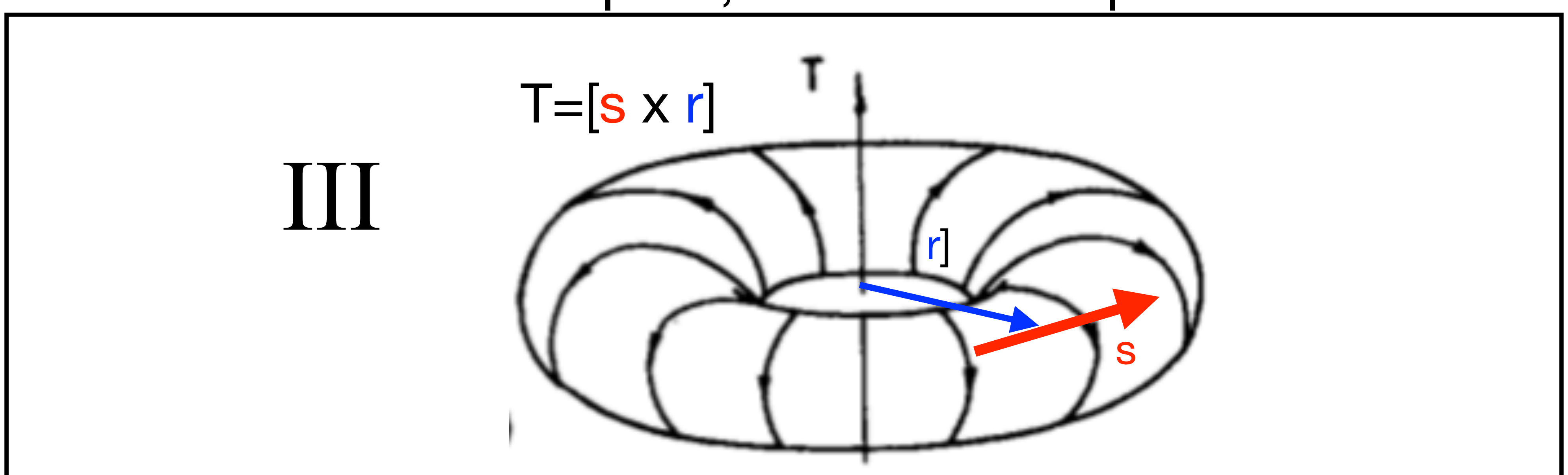


anapole, toroidal multipole



Expansion of the scattering operator Q in powers of (k.r). Splitting neutron and electron variables

We measure an expectation value of the scattering operator

$$\mathbf{Q}_{\perp} = [\tilde{\boldsymbol{\kappa}} \times \mathbf{Q} \times \tilde{\boldsymbol{\kappa}}]$$

electron neutron

(**q**==**k**)

$$\mathbf{Q} = \exp(\mathrm{i}\,\mathbf{r} \bullet \mathbf{k})[\mathbf{s} - (\mathrm{i}/\hbar k)(\tilde{\boldsymbol{\kappa}} \times \mathbf{p})],$$

$\tilde{\mathbf{k}}$=**k**/k where **k** is the neutron scattering wavevector

spin momentum -> L

expectation value of **Q** is $\langle \mathbf{Q} \rangle \equiv \langle \psi_{ATOM} | \mathbf{Q} | \psi_{ATOM} \rangle$

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electron neutron

(**q**==**k**)

$$\mathbf{Q} = \exp(\mathrm{i}\, \mathbf{r} \bullet \mathbf{k})[\mathbf{s} - (\mathrm{i}/\hbar k)(\tilde{\boldsymbol{\kappa}} \times \mathbf{p})],$$

$\tilde{\mathbf{k}}$=**k**/k where **k** is the neutron scattering wavevector

spin momentum -> L

expectation value of **Q** is $\langle \mathbf{Q} \rangle \equiv \langle \psi_{ATOM} | \mathbf{Q} | \psi_{ATOM} \rangle$

mathematical difficulty is related to the expansion of the exponent and further calculus

exp(**k r**)= $4\pi \sum_{L=0}^{\infty} \sum_{M=-L}^{L} \mathrm{i}^L j_L(\kappa r) \boxed{Y_M^L(\Omega_{\boldsymbol{r}})} Y_M^{L*}(\Omega_{\boldsymbol{\kappa}}),$

will give multipoles

use of Racah tensor-algebra, is required S.W. Lovesey, “Theory of Neutron Scattering from Condensed Matter”, Oxford Univ. Press, 1987

Expansion of the scattering operator Q in powers of (k.r). Splitting neutron and electron variables

We measure an expectation value of the scattering operator

$$\mathbf{Q}_{\perp} = [\tilde{\boldsymbol{\kappa}} \times \mathbf{Q} \times \tilde{\boldsymbol{\kappa}}]$$

electron neutron

(**q**==**k**)

$$\mathbf{Q} = \exp(\mathrm{i}\,\mathbf{r} \bullet \mathbf{k})[\mathbf{s} - (\mathrm{i}/\hbar k)(\tilde{\boldsymbol{\kappa}} \times \mathbf{p})],$$

$\tilde{\mathbf{k}}$=**k**/k where **k** is the neutron scattering wavevector

spin momentum -> L

expectation value of **Q** is $\langle \mathbf{Q} \rangle \equiv \langle \psi_{ATOM} | \mathbf{Q} | \psi_{ATOM} \rangle$

We expand in powers of (**k r**), (i.e. Y_{L0})

$$e^{-i\boldsymbol{\kappa}\cdot\mathbf{r}} = \sum_{n=0}^{\infty} (2n+1)\,(-i)^n\, j_n(\rho) P_n(\cos\theta)$$

$$\rho = \kappa r \text{ and } \cos\theta = \boldsymbol{\kappa}\cdot\mathbf{r}/\rho,$$

Some mathematics…

Legendre polynomials

$$P_{n=0,1,2,3}(x) = \left[1, x, -\frac{1}{2} + \frac{3x^2}{2}, \frac{5}{2}x^3 - \frac{3}{2}x\right]$$

Spherical Bessel functions

$$j_{n=0,1,2}(x) = \left[\frac{\sin(x)}{x}, \frac{-\cos(x)\,x + \sin(x)}{x^2}, \frac{-\sin(x)\,x^2 - 3\cos(x)\,x + 3\sin(x)}{x^3}\right]$$

$$j_n(\rho) = \rho\{j_{n-1}(\rho) + j_{n+1}(\rho)\}/(2n+1).$$

multipoles (parity even) Sketch of spin multipole expansion for **Q.**

$$e^{-i\boldsymbol{\kappa}\cdot\mathbf{r}} = \sum_{n=0}^{\infty}(2n+1)\,(-i)^n\, j_n(\rho)P_n(\cos\theta) \qquad \rho = \kappa r \text{ and } \cos\theta = \boldsymbol{\kappa}\cdot\mathbf{r}/\rho,$$

Legendre polynomia

$P_{n=0,1,2,3}(x) =$ [1, x, …

Spherical Bessel fu

$j_{n=0,1,2}(x) =$ [$\frac{\sin(x}{x}$ …

expansion $e^{-i\vec{k}\cdot\vec{r}}$ in powers of $(\vec{k}\cdot\vec{r})$

$$\langle\hat{Q}_i\rangle \simeq \left\langle \underline{\hat{S}_i}\cdot\left(\ldots j_0(\rho)+\ldots\frac{1}{\rho^2}(\underline{\vec{k}}\,\underline{\vec{r}})^2 j_2(\rho)+\ldots\frac{1}{\rho^4}(\vec{k}\vec{r})^4 j_4(\rho)\ldots\right)\right\rangle$$

ith component of atom spin

neutron

atom

$\rho = |\underline{\vec{k}}||\underline{\vec{r}}|$

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ith component of atom spin

neutron

atom

$\rho = |\underline{\vec{k}}||\underline{\vec{r}}|$

dipole

$$\langle\varphi_{atom}|\hat{\underline{S}_i}|\varphi_{atom}\rangle\langle j_0(k)\rangle$$

$$\langle j_n(\kappa)\rangle = \int_0^\infty r^2 R^2(r) j_n(\kappa r)dr$$

multipoles (parity even) Sketch of spin multipole expansion for **Q.**

$$e^{-i\boldsymbol{\kappa}\cdot\mathbf{r}} = \sum_{n=0}^{\infty}(2n+1)\,(-i)^n\, j_n(\rho) P_n(\cos\theta) \qquad \rho = \kappa r \text{ and } \cos\theta = \boldsymbol{\kappa}\cdot\mathbf{r}/\rho,$$

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ith component of atom spin

neutron atom

$\rho = |\underline{\vec{k}}||\underline{\vec{r}}|$

$$\sum_{jl} \underline{S}_i \,(\underline{\tilde{k}}_j \underline{\tilde{r}}_j)(\underline{\tilde{k}}_l \underline{\tilde{r}}_l) \cdot j_2(\rho)$$

$i, j, k = x, y, z$

$\tilde{\kappa}=\mathbf{k}/k$

$\tilde{\mathbf{r}}=\mathbf{r}/r$

we split neutron / atom

octupole

$$<Q_i> \sim \sum_{jl} (\underline{k}_j \underline{k}_l) \langle \underline{\Psi}_{atom} | \underbrace{\underline{\hat{S}}_i \underline{\tilde{r}}_j \underline{\tilde{r}}_l}\, | \underline{\Psi}_{atom} \rangle <j_2(k)>$$

$$<Q_i> \sim \sum_{jl} (\underline{\hat{k}}_j \underline{\tilde{k}}_l) \cdot < \hat{M}_{ijl} > \cdot <j_2(k)>$$

contracting by jl $\rightarrow$ $\vec{Q}(|\vec{k}|, \Theta) \leftarrow \vec{k}$ octupole

Dipole approximation

We expand in powers of (**k** r), (i.e. Y_{L0})

$$e^{-i\boldsymbol{\kappa}\cdot\mathbf{r}} = \sum_{n=0}^{\infty}(2n+1)\,(-i)^n\,j_n(\rho)P_n(\cos\theta)$$

$$\simeq j_0(\rho) - 3i\,j_1(\rho)\cos\theta = \boxed{j_0(\rho) - i\boldsymbol{\kappa}\cdot\mathbf{r}\{j_0(\rho)+j_2(\rho)\}}$$

$\rho = \kappa r$ and $\cos\theta = \boldsymbol{\kappa}\cdot\mathbf{r}/\rho$,

even S odd L

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even S odd L

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$\rho = \kappa r$ and $\cos\theta = \boldsymbol{\kappa}\cdot\mathbf{r}/\rho$,

even S odd L

electron neutron

$$\mathbf{Q} = \exp(\mathrm{i}\,\mathbf{r}\bullet\mathbf{k})[\mathbf{s} - (\mathrm{i}/\hbar k)(\tilde{\boldsymbol{\kappa}}\times\mathbf{p})],$$

spin momentum

is a term, which contains a linear combination of the spin and orbital angular moment of the magnetic ion, ***S*** and ***L***, respectively.

$$\tfrac{1}{2}\{\langle j_0(\kappa)\rangle + \langle j_2(\kappa)\rangle\}\,\mathbf{l},$$

$$\boxed{\langle\mathbf{Q}\rangle = \tfrac{1}{2}\langle j_0(\kappa)\rangle(\mathbf{l}+2\mathbf{s}) + \tfrac{1}{2}\langle j_2(\kappa)\rangle\,\mathbf{l}.}$$

$$\langle j_n(\kappa)\rangle = \int_0^{\infty} r^2R^2(r)j_n(\kappa r)dr$$

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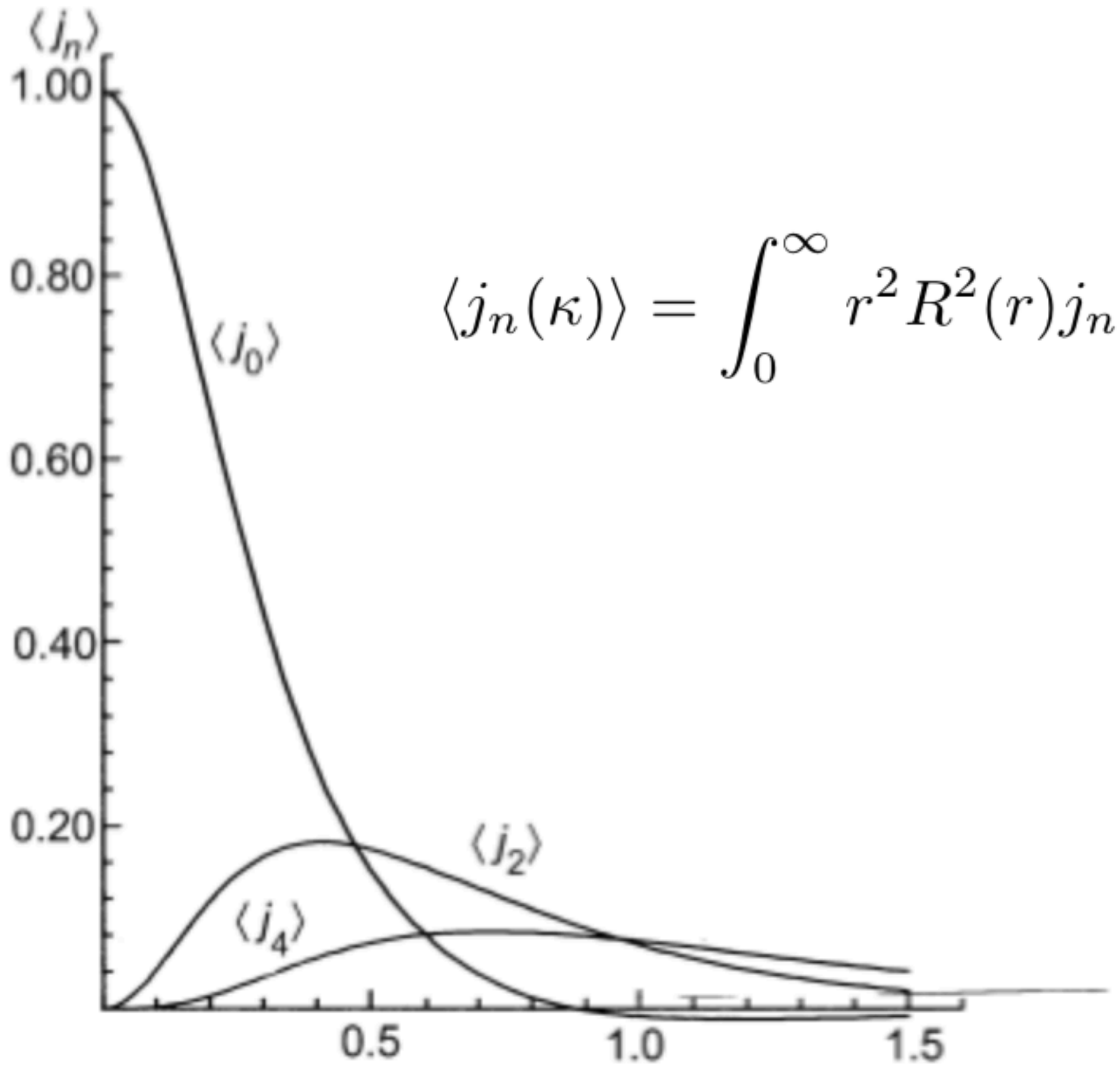
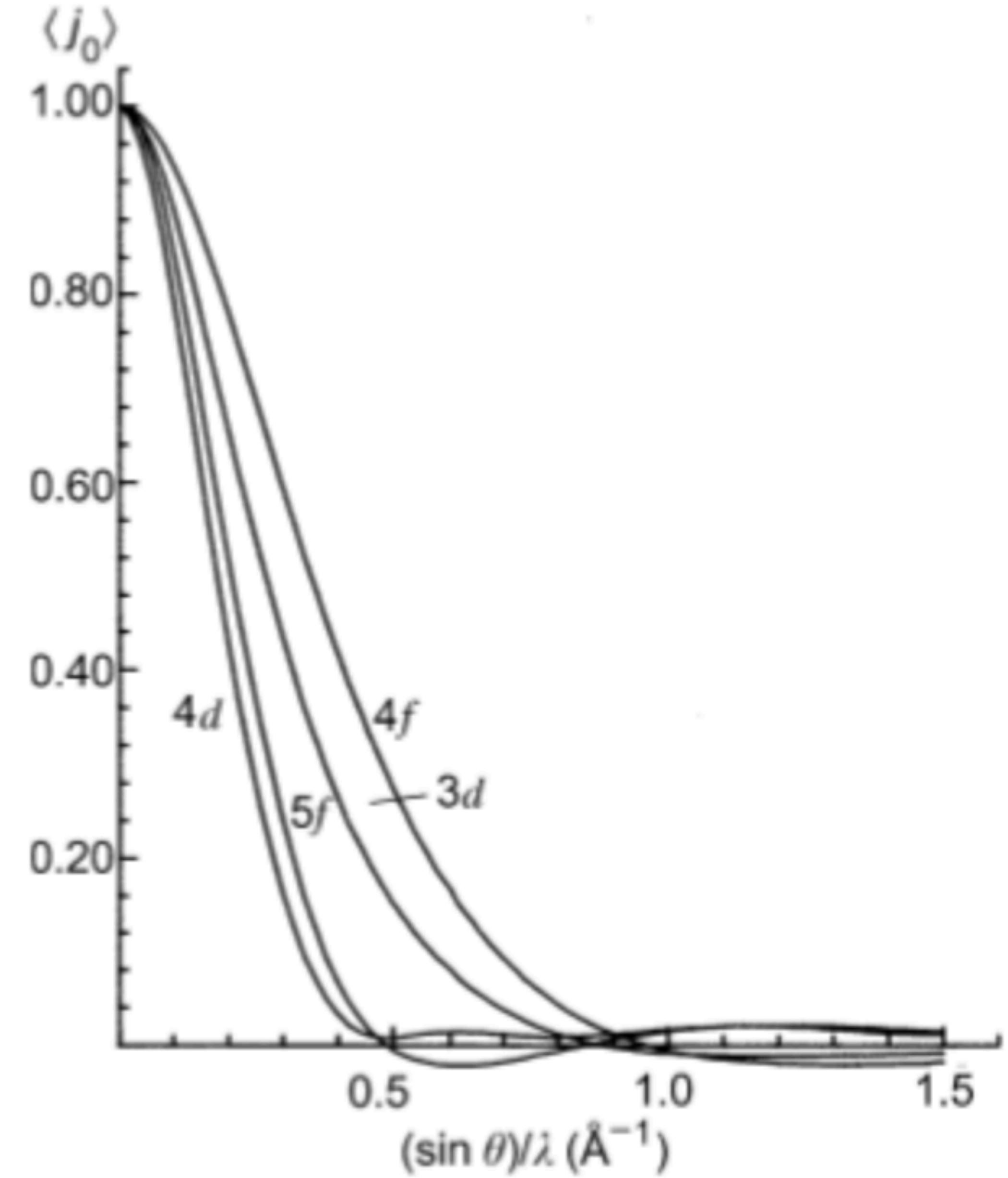
$$\left[\frac{\sin(x)}{x},\ \frac{-\cos(x)\,x+\sin(x)}{x^2},\ \frac{-\sin(x)\,x^2-3\cos(x)\,x+3\sin(x)}{x^3}\right]$$

Examples of dipole and contribution to neutron scattering

ssumed to be spin independent. In this case,



$$\langle j_n(\kappa)\rangle = \int_0^\infty r^2 R^2(r) j_n(\kappa r) dr$$



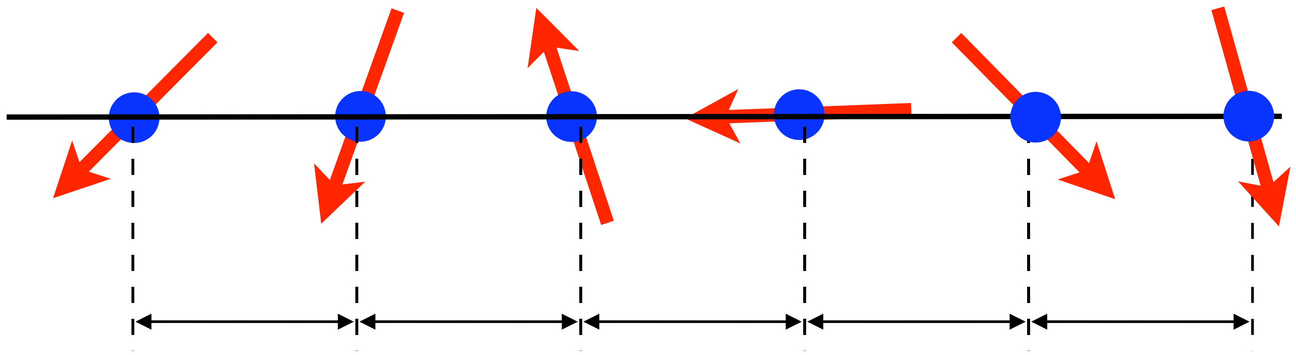
ig. 6.1.2.1. The integrals $\langle j_0\rangle$, $\langle j_2\rangle$, and $\langle j_4\rangle$ for the Fe^{2+} ion plotted against $(\sin\theta)/\lambda$. The integrals have been calculated from wavefunctions given by Clementi & Roetti (1974).

Fig. 6.1.2.2. Comparison of 3*d*, 4*d*, 4*f*, and 5*f* form factors. The 3*d* form factor is for Co, and the 4*d* for Rh, both calculated from wavefunctions given by Clementi & Roetti (1974). The 4*f* form factor is for Gd^{3+} calculated by Freeman & Desclaux (1972) and the 5*f* is that for U^{3+} given by Desclaux & Freeman (1978).

Intensity ~ $|\frac{1}{2}\langle j_0(\kappa)\rangle(\mathbf{l}+2\mathbf{s}) + \frac{1}{2}\langle j_2(\kappa)\rangle\,\mathbf{l}\,|^2$

Experimental example of incoherent and coherent dipole magnetic scattering on a lattice of spins

incoherent $I \sim \left\langle \hat{S}^2 \right\rangle = S(S+1)$



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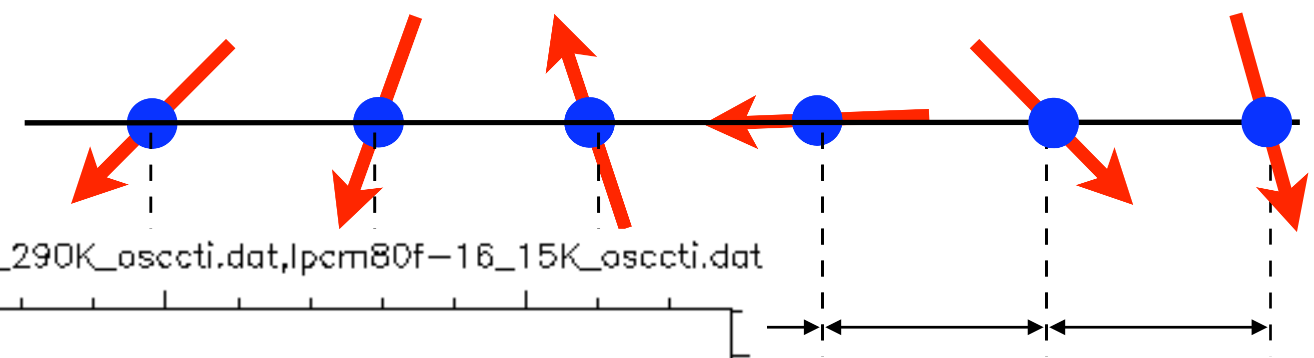

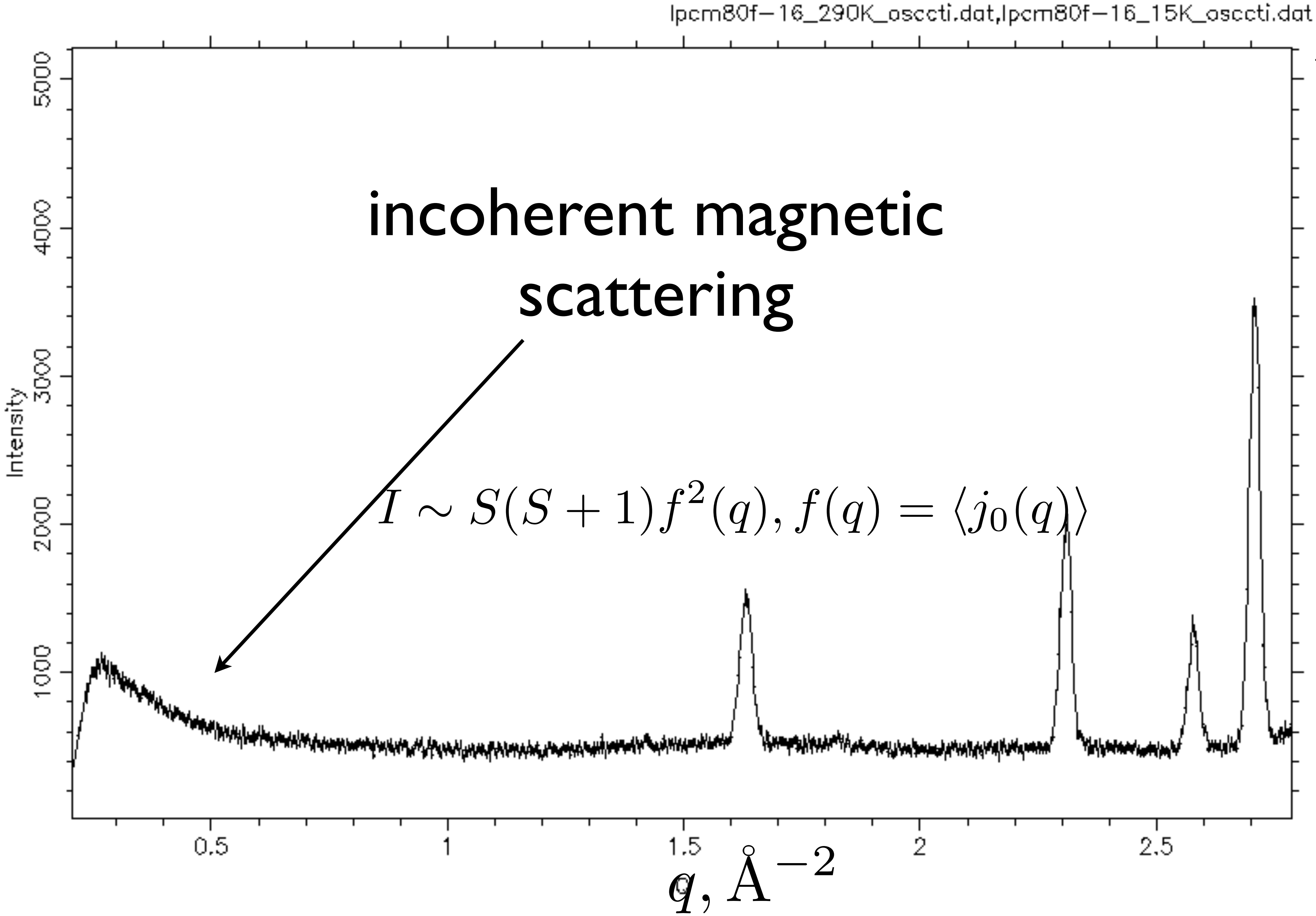

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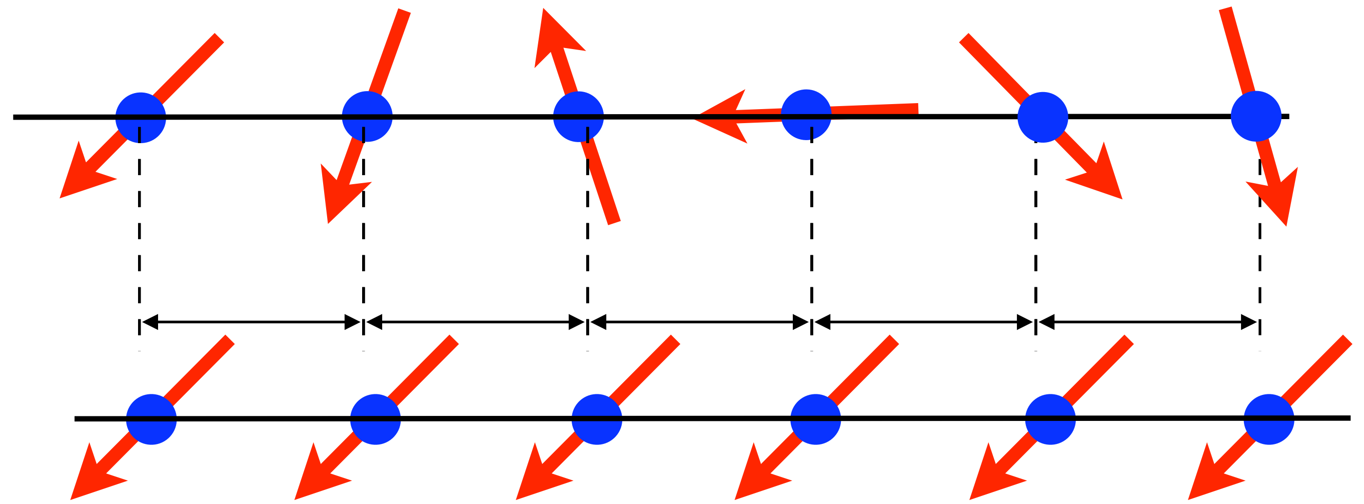
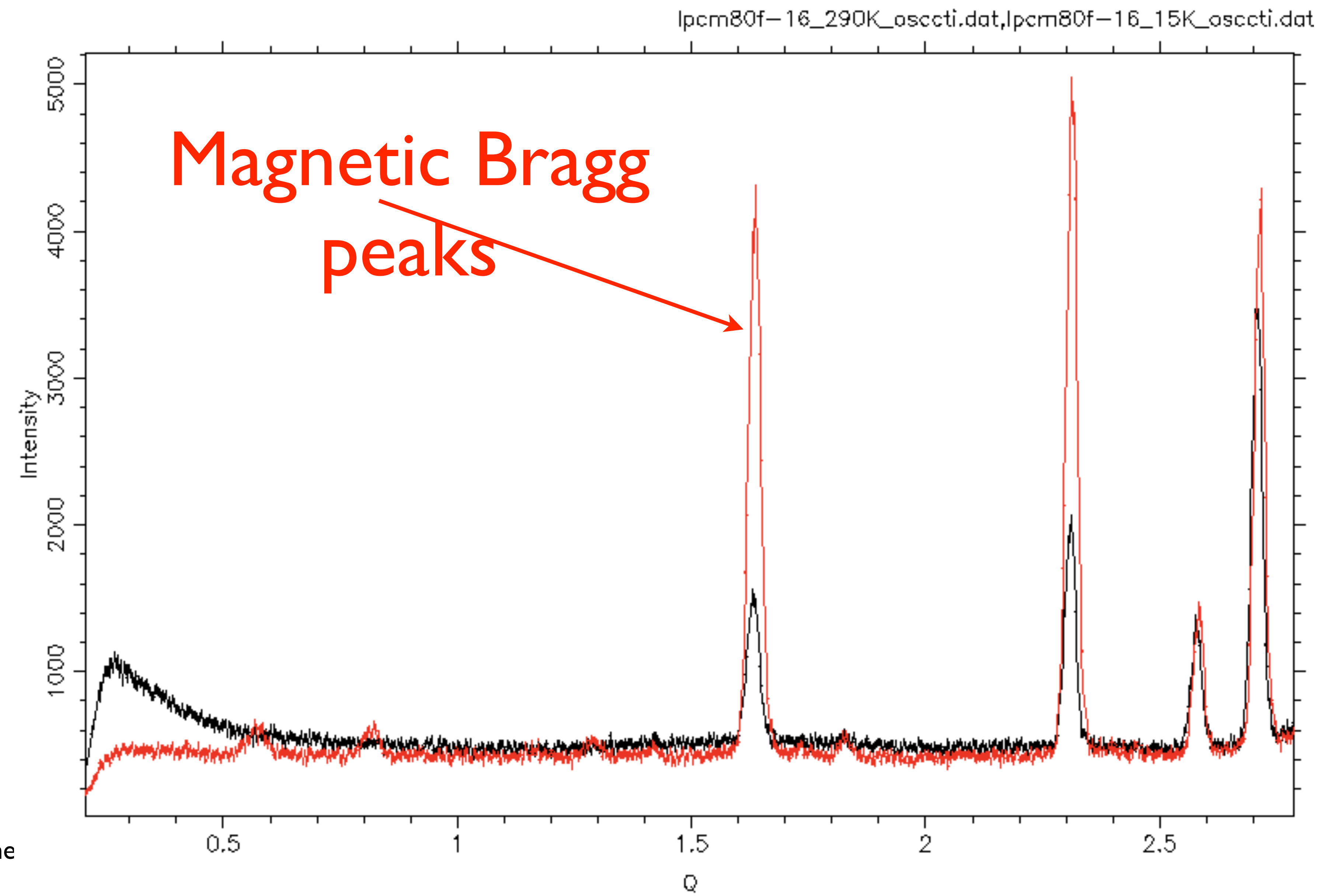
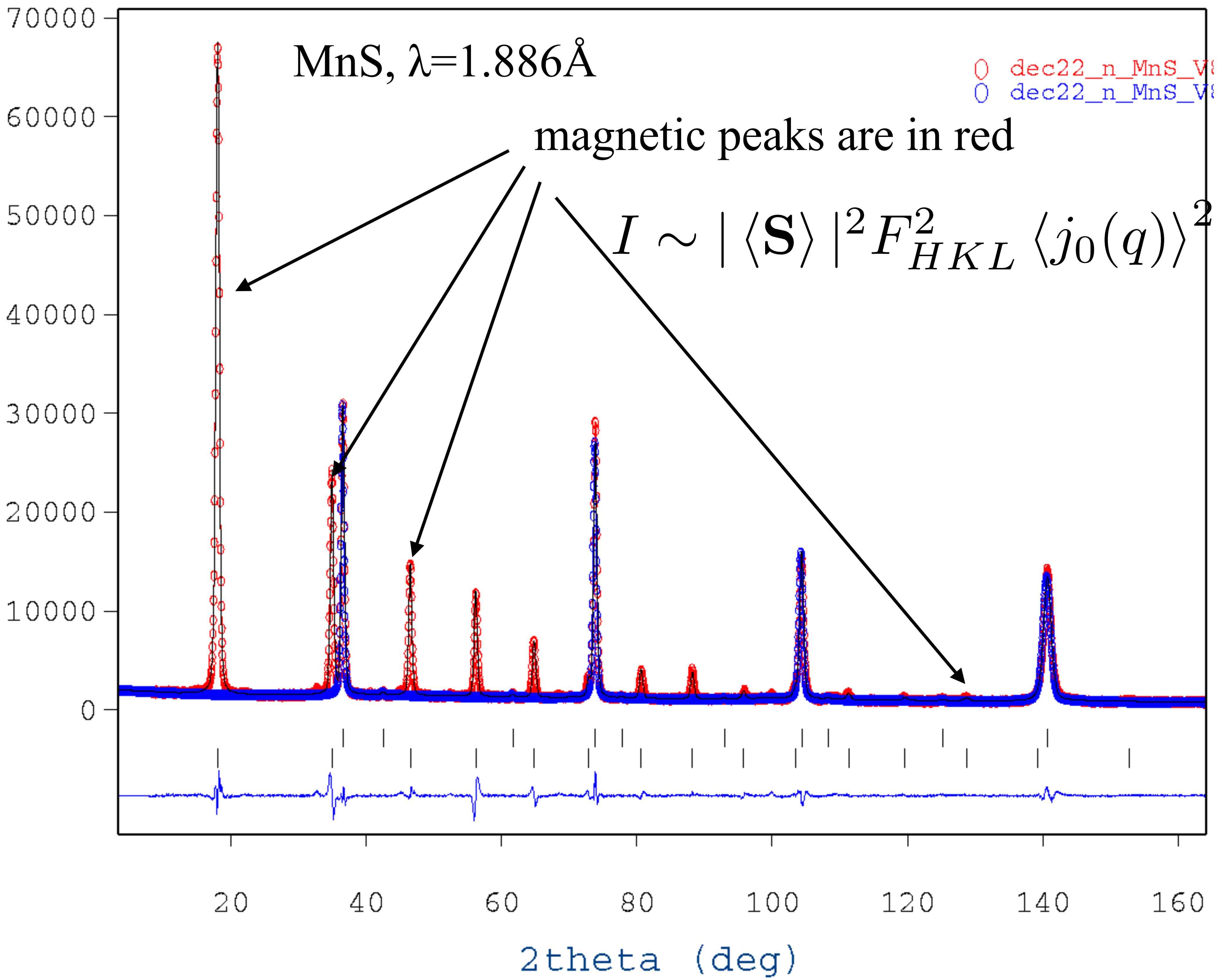


coherent Bragg scattering

$$I \sim |\langle \mathbf{S} \rangle|^2 F_{HKL}^2 \langle j_0(q) \rangle^2$$



Experimental example of coherent dipole magnetic scattering MnS/MnO



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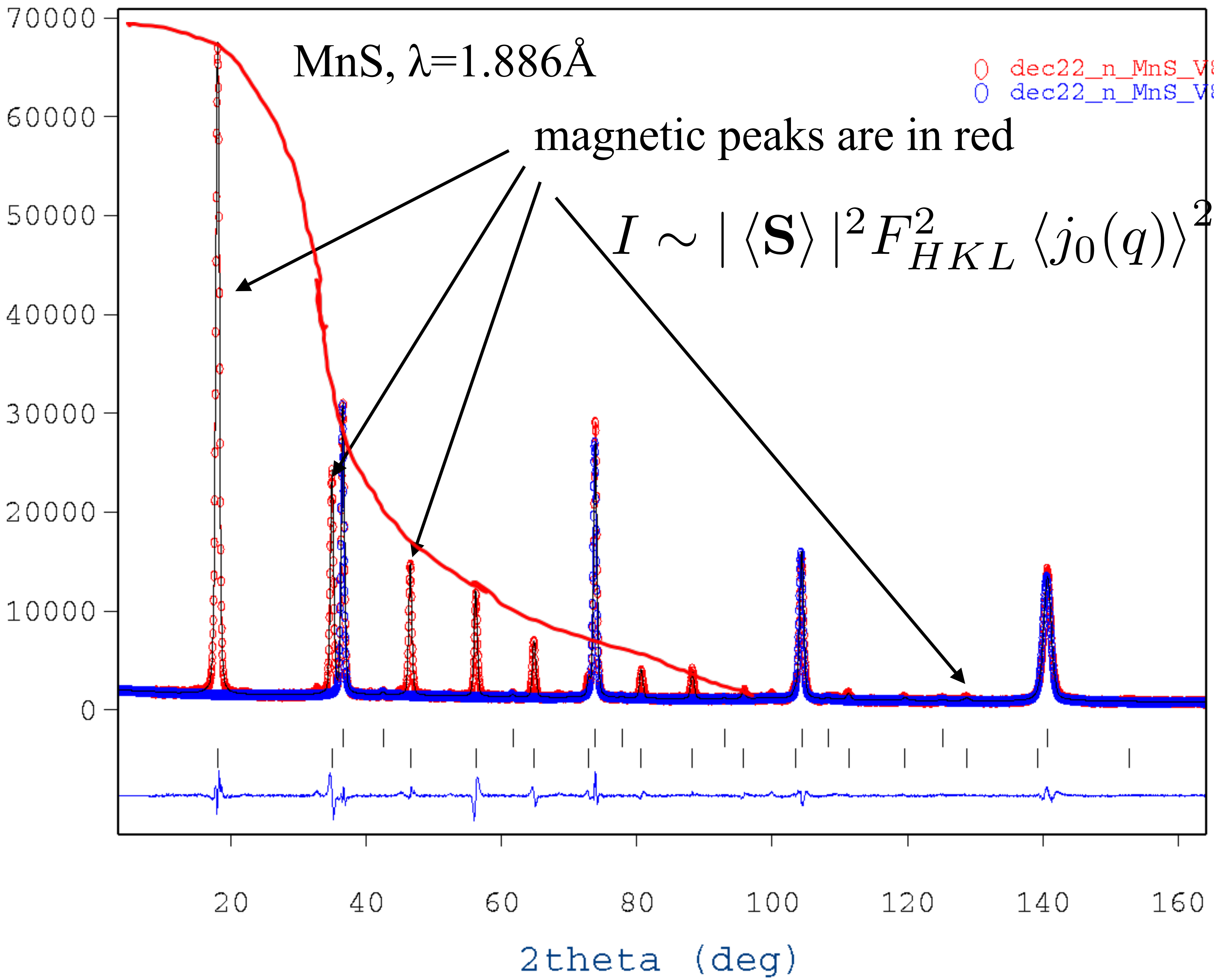


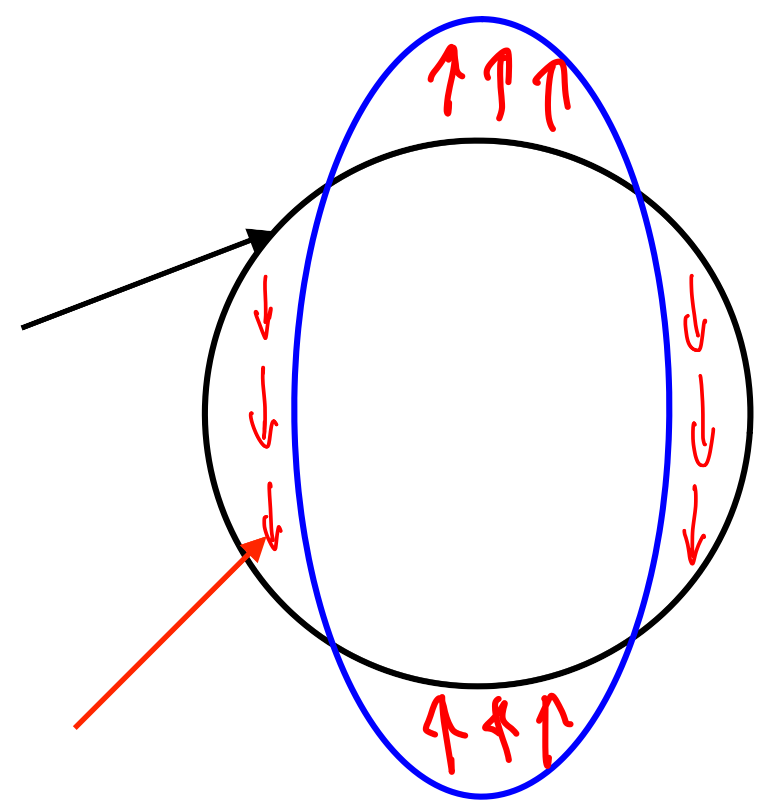
Conventional multipoles. Naive visualisations of octopole

magnetic multipoles == tensors of rank R

$$\hat{M}_{ijk\ldots} = \hat{S}_i \underbrace{r_j r_k \ldots}_{R} \qquad i,j,k\ldots = 1,2,3(x,y,z)$$

electron spin or total **J**

electron spin coordinates



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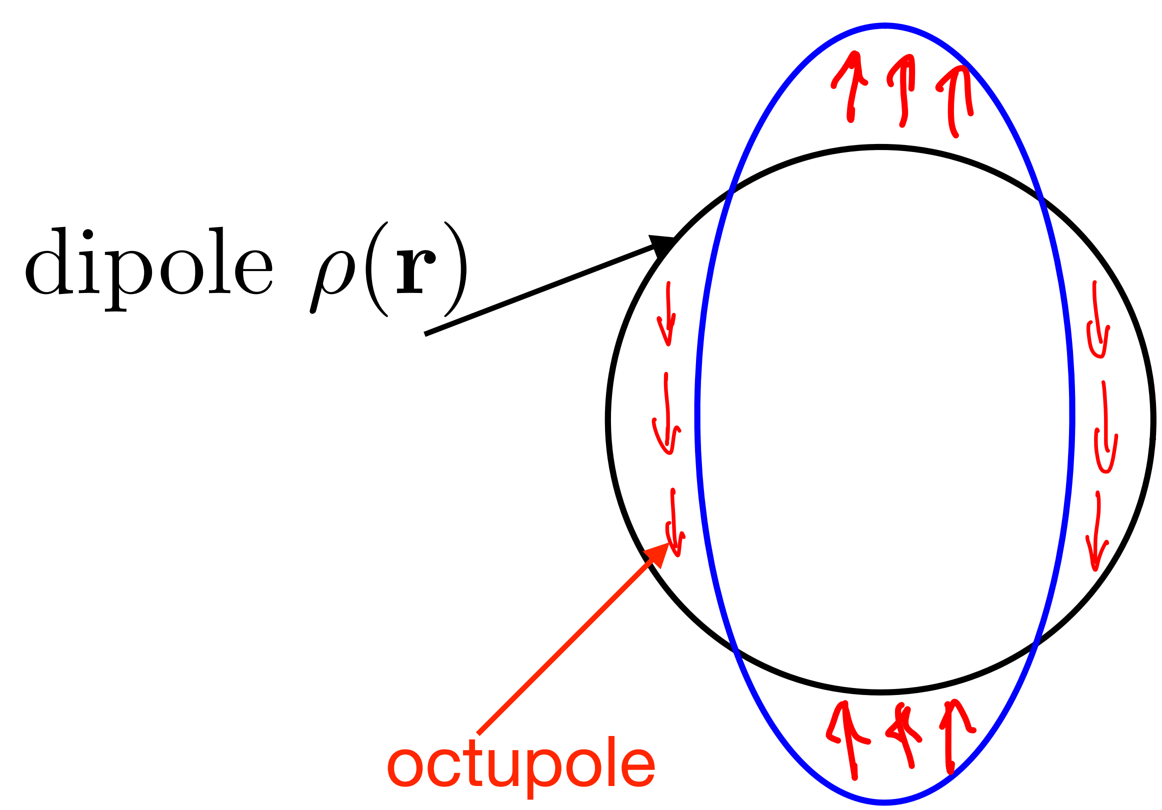
magnetic octopole: rank R=3 - operator in QM and has classical counterpart

$$\hat{O}_{ijk} = \hat{S}_i r_j r_k$$

Exp. value $\langle Q_{ijk} \rangle = \langle \psi | \hat{O}_{ijk} | \psi \rangle \longleftrightarrow O^c_{ijk} = \int M_i(\vec{r}) \cdot \rho(\vec{r})\, r_j r_k \cdot d\vec{r}$



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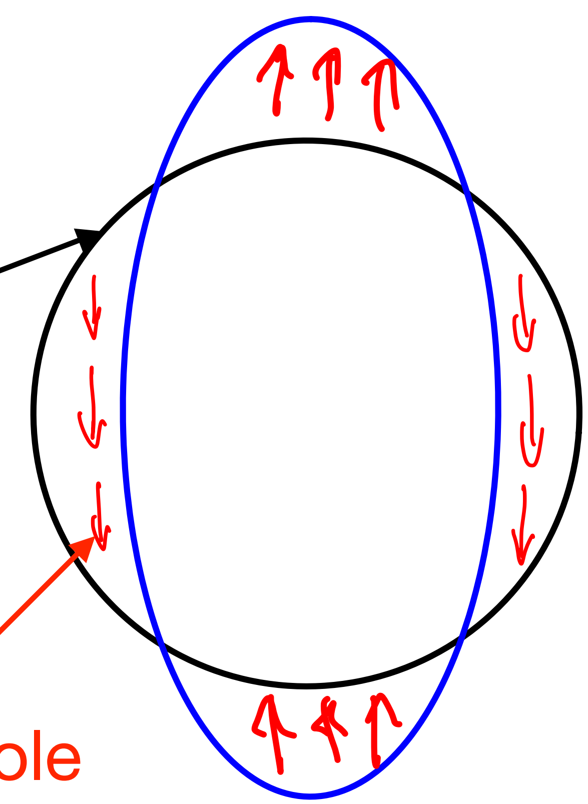
dipole $\rho(\mathbf{r})$



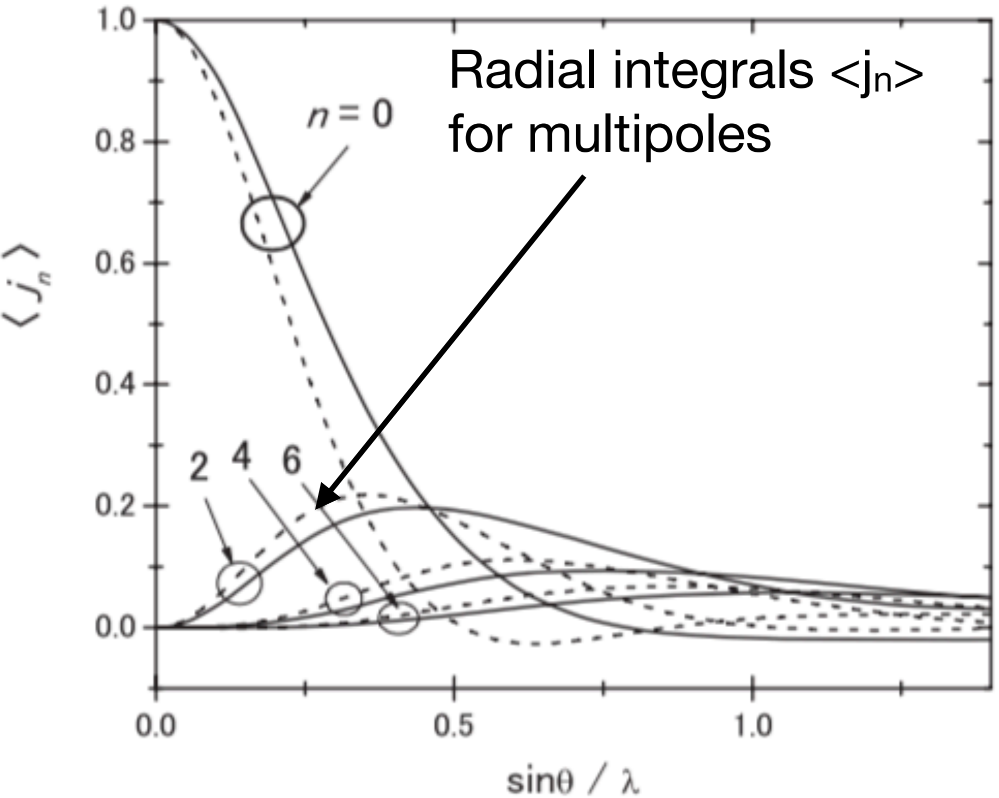
octupole

$$\mathbf{L} + \mathbf{S} \sim [\mathbf{p} \times \mathbf{r}] = m[\frac{d\mathbf{r}}{dt} \times \mathbf{r}]$$

spin is **axial vector** = product of two polar vectors
time inversion (1’) **S** = -**S**
space inversion (-1) **S** = **S**
conventional odd-rank magnetic multipoles fulfil this: dipole (vector), octupole (rank-3 tensor),…

R=1,3,… 2n+1

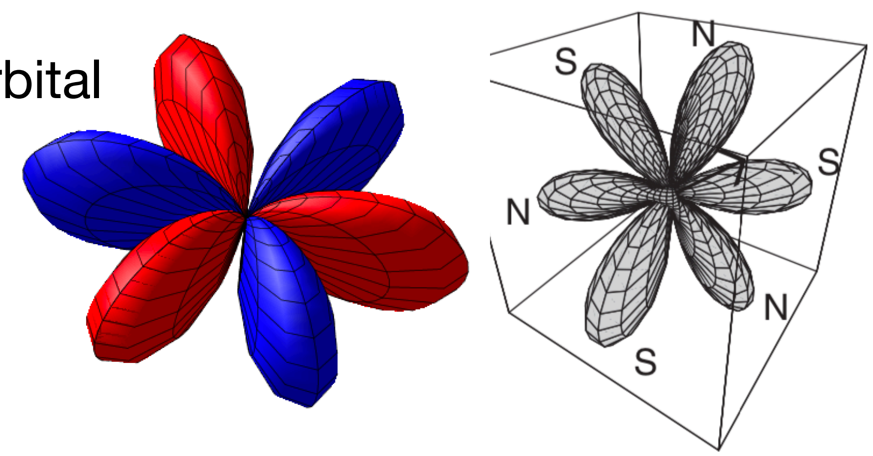
Multipole moments visualisation (qualitative, symmetry properties) and their q-dependence



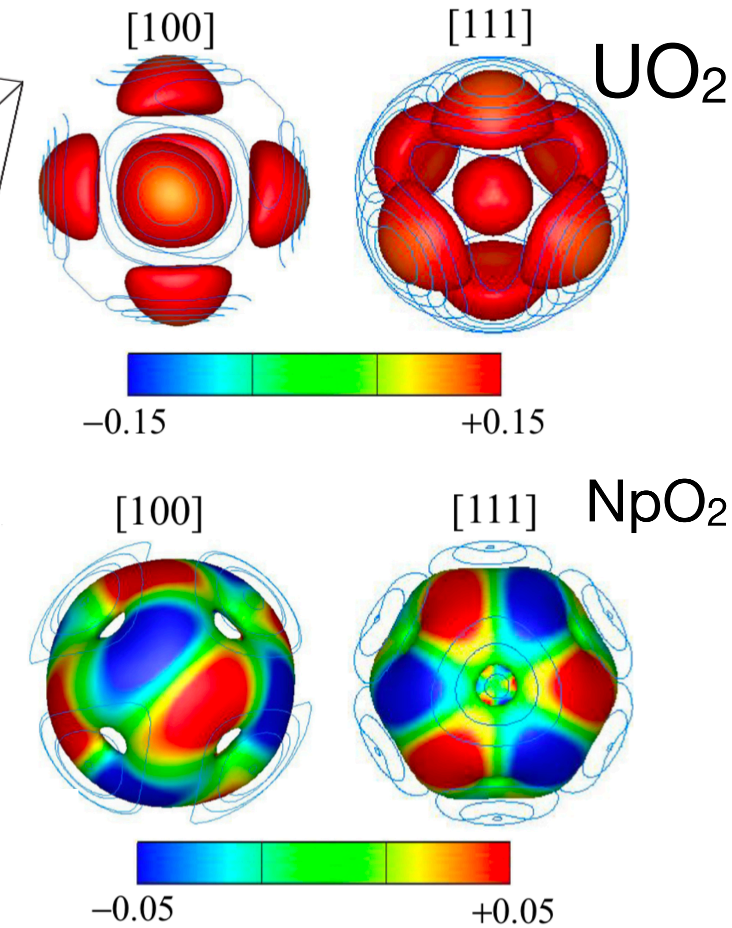
Radial integrations of spherical Bessel function $\langle j_n \rangle$ as fun
neutron momentum transfer $\sin\theta/\lambda$,
and broken lines represent the re

J. Phys. Soc. Jpn. 76, 094702 (2007)
J. Phys. Soc. Jpn. 80 (2011) SB008

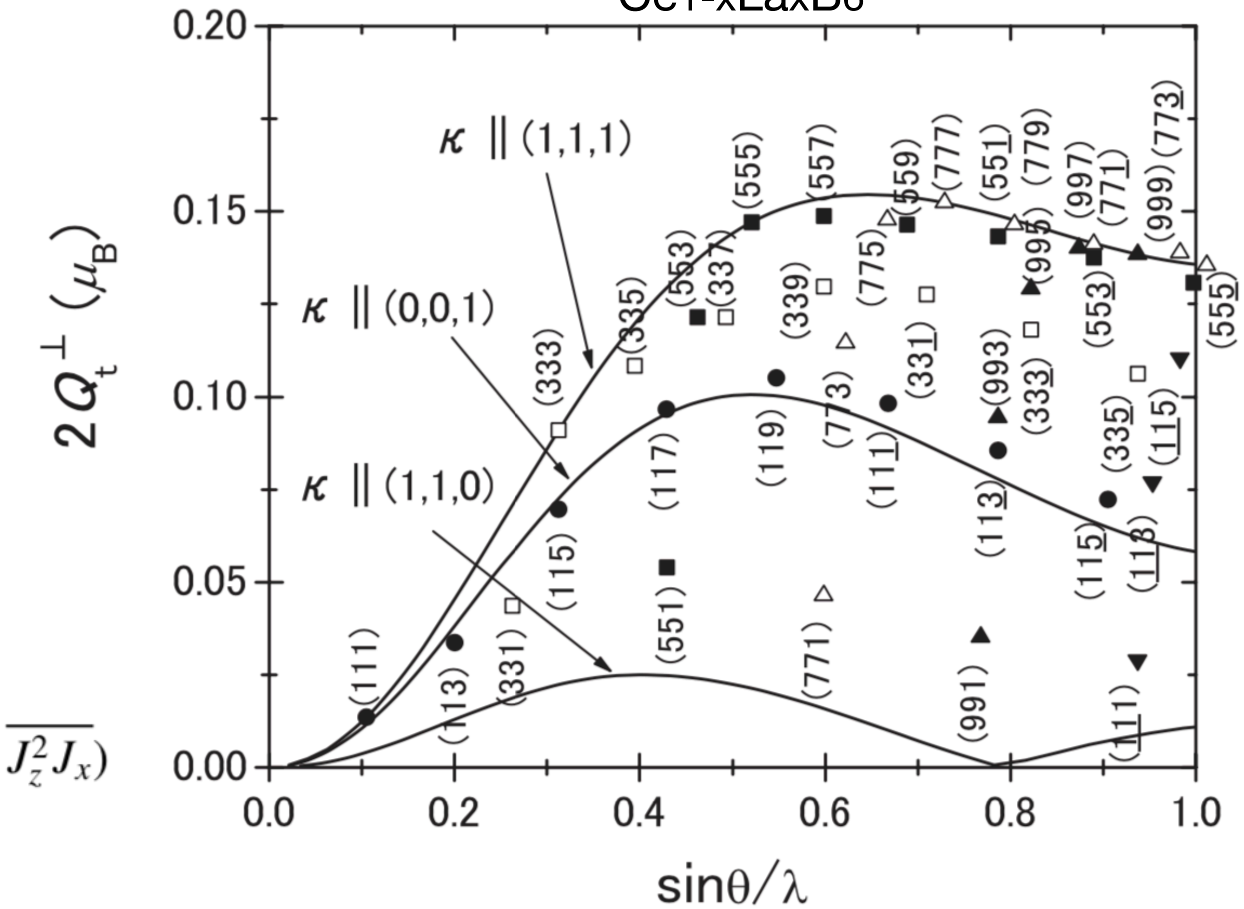
f-orbital
Y_{33}



J. Phys. Soc. Jpn. 87, 041008 (2018)



$Ce_{1-x}La_xB_6$



$$T_x^{\beta} = \frac{\sqrt{15}}{6}(\overline{J_x J_z^2} - \overline{J_z^2 J_x})$$

Symmetry of multipoles

magnetic multipoles == tensors of rank R

$$\hat{M}_{ijk\ldots} = \hat{S}_i \underbrace{r_j r_k \ldots}_{R}$$

$i, j, k... = 1, 2, 3(x, y, z)$

electron spin or total **J**

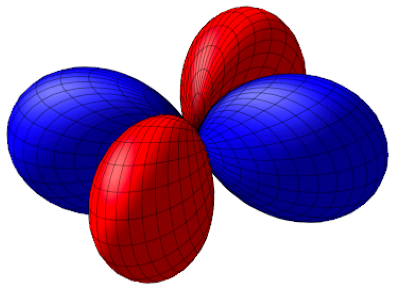
electron spin coordinates

1. Only **time reversal odd** multipoles because of n-e interaction Hamiltonian

$$1' \cdot \hat{M}_{ijk\ldots} = -\hat{M}_{ijk\ldots}$$

one can construct multipoles from pure *J*$_x$, *J*$_y$, *J*$_z$, angular operators -> can be mapped to **Q**

$$\mathbf{Q} = \exp(\mathrm{i}\mathbf{R}_j \bullet \mathbf{k})[\mathbf{s}_j - (\mathrm{i}/\hbar k)(\boldsymbol{\kappa} \times \mathbf{p}_j)]$$



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$$\mathbf{Q} = \exp(\mathrm{i}\mathbf{R}_j \bullet \mathbf{k})[\mathbf{s}_j - (\mathrm{i}/\hbar k)(\boldsymbol{\kappa} \times \mathbf{p}_j)]$$

2. If wave function of unpaired electrons has definitive parity, i.e. under space inversion

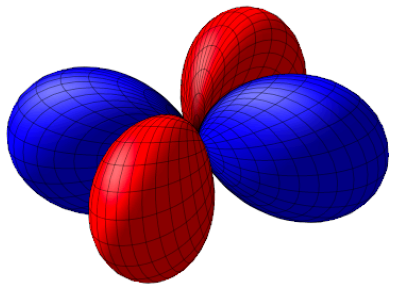
$$\bar{1}|\psi> = \pm|\psi>$$

$$<\psi|\hat{M}_{ijk\ldots}|\psi> \neq 0$$

$$\bar{1} \cdot \hat{M}_{ijk\ldots} = +\hat{M}_{ijk\ldots}$$

conventional multipoles R=1,3,… 2n+1

we can have only **parity even** multipoles -> rank R -odd, e.g. no conventional quadrupoles



What are the magnetic objects neutrons sensitive to?

3. Dirac dipoles (anapoles) that are polar (parity odd) and magnetic (time odd).

Literature on neutron scattering on Dirac multipoles

S W Lovesey, "Theory of neutron scattering by electrons in magnetic materials", Phys. Scr. 90 (2015) 108011. Main paper

S W Lovesey, "Magneto-electric operators in neutron scattering from electrons" J. Phys.: Condens. Matter 26 (2014) 356001

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What are the magnetic objects neutrons sensitive to?

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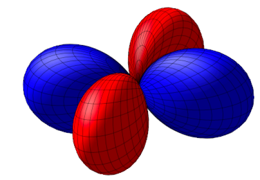
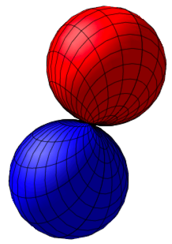
If wave function of unpaired electrons has no parity we can have parity odd multipoles





spinor spinor

$$|\psi >= (a \left|s, +\frac{1}{2}\right\rangle |3d\rangle + b| \left|s, -\frac{1}{2}\right\rangle |2p\rangle)$$

orbitals with opposite parities

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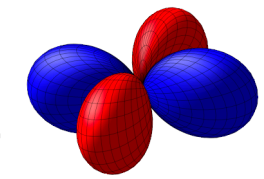

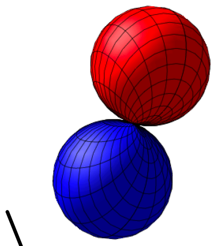

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$$|\psi> = (a\left|s,+\frac{1}{2}\right\rangle|3d\rangle + b|\left|s,-\frac{1}{2}\right\rangle|2p\rangle)$$

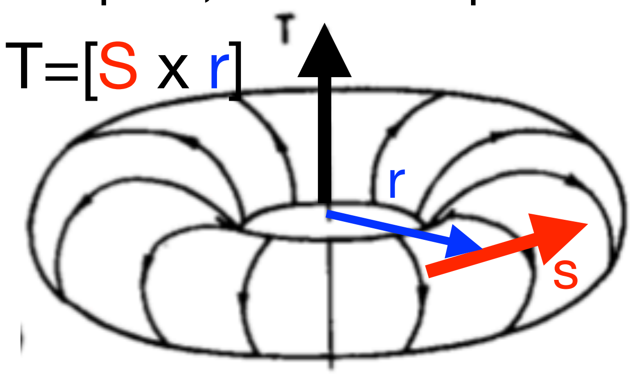
orbitals with opposite parities

If wave function of unpaired electrons has no parity we can have parity odd multipoles

$$<\psi|[\mathbf{S}\times\mathbf{n}]|\psi>\neq 0, \mathbf{n}=\mathbf{r}/r$$

anapole, toroidal dipole moment

T=[S x r]



$$\boldsymbol{T}(\boldsymbol{\mu}) = \frac{1}{2}\int \boldsymbol{r}\times\boldsymbol{\mu}_{\perp}\,\mathrm{d}^3\boldsymbol{r}\,.$$

V.M. Dubovik and V.V. Tugushev, Toroid moments in electrodynamics and solid-state physics

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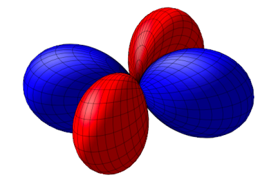
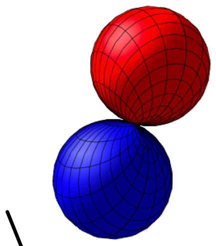
S W Lovesey and D D Khalyavin "Neutron scattering by Dirac multipoles", J. Phys.: Condens. Matter 29 (2017) 215603

What are the magnetic objects neutrons sensitive to?

3. Dirac dipoles (anapoles) that are polar (parity odd) and magnetic (time odd).

If wave function of unpaired electrons has no parity we can have parity odd multipoles

$$< \psi|[\mathbf{S}\times\mathbf{n}]|\psi > \neq 0, \mathbf{n} = \mathbf{r}/r$$

spinor   spinor

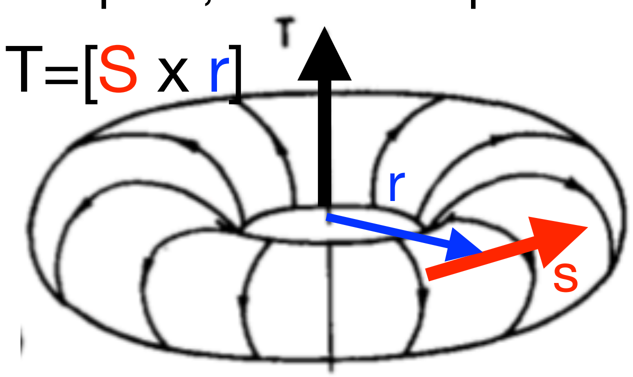
$$|\psi> = (a\left|s,+\frac{1}{2}\right\rangle|3d\rangle + b|\left|s,-\frac{1}{2}\right\rangle|2p\rangle)$$

orbitals with opposite parities

$$\langle 2,0|_d \left\langle +\frac{1}{2}\right|(S_y n_z)_x\left|-\frac{1}{2}\right\rangle|1,0\rangle_p \neq 0$$

anapole, toroidal dipole moment

T=[S x r]



$$\boldsymbol{T}(\boldsymbol{\mu}) = \frac{1}{2}\int \boldsymbol{r}\times\boldsymbol{\mu}_\perp\, \mathrm{d}^3\boldsymbol{r}\,.$$

V.M. Dubovik and V.V. Tugushev, Toroid moments in electrodynamics and solid-state physics

Literature on neutron scattering on Dirac multipoles

S W Lovesey, “Theory of neutron scattering by electrons in magnetic materials”, Phys. Scr. 90 (2015) 108011. Main paper

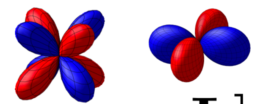
S W Lovesey, “Magneto-electric operators in neutron scattering from electrons” J. Phys.: Condens. Matter 26 (2014) 356001

S W Lovesey and D D Khalyavin “Neutron scattering by Dirac multipoles”, J. Phys.: Condens. Matter 29 (2017) 215603

A zero-magnetization ferromagnet $Sm_{0.976}Gd_{0.024}Al_2$

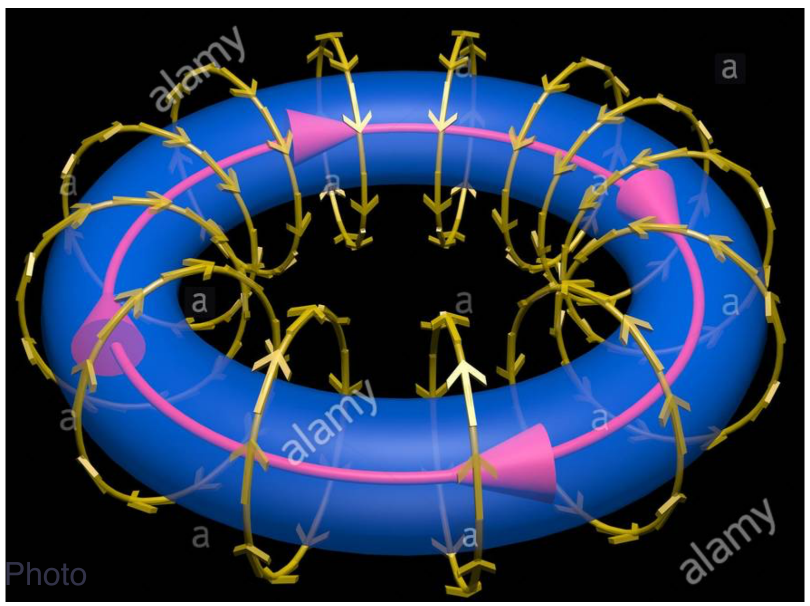
S W Lovesey et al PRL 122, 047203 (2019). “Direct Observation of Anapoles by Neutron Diffraction”: Experiment & theory

Atomic wave functions are $4f^5$-$5d^1$



$$\boldsymbol{\Omega}_S = [\mathbf{S} \times \mathbf{n}], \boldsymbol{\Omega}_L = [\mathbf{L} \times \mathbf{n}] - [\mathbf{n} \times \mathbf{L}]$$

toroidal magnetic field is localised



Alamy Stock Photo

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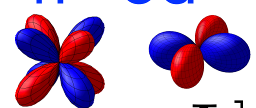

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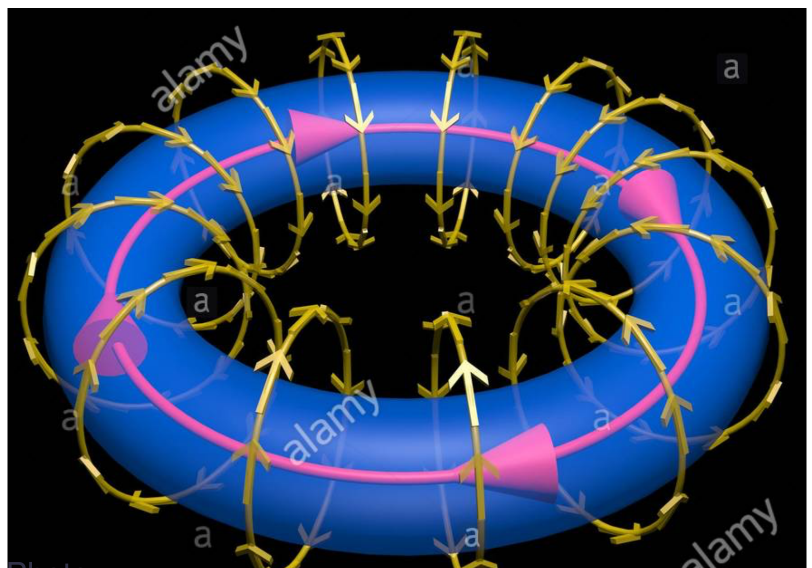
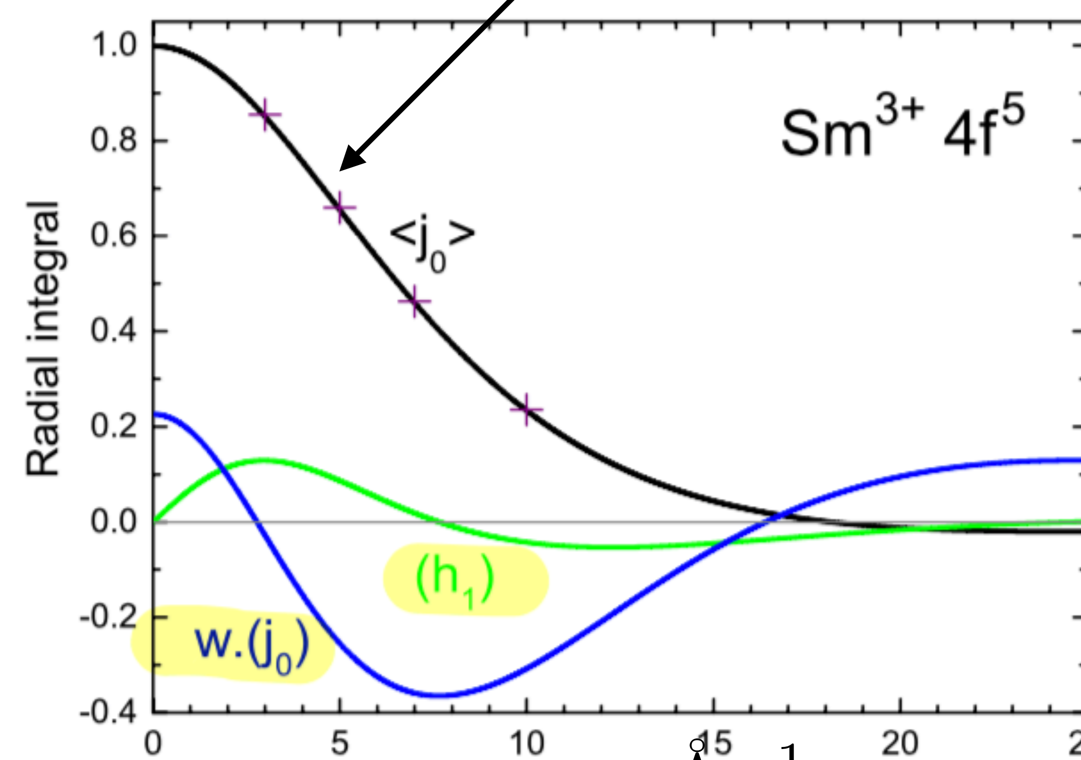


Alamy Stock Photo

magnetic dipole

$$\langle \mathbf{Q} \rangle = \frac{1}{2}\langle j_0(\kappa)\rangle(\mathbf{l} + 2\mathbf{s}) + \frac{1}{2}\langle j_2(\kappa)\rangle\, \mathbf{l}.$$



$\sim \boldsymbol{\kappa}, \mathrm{\AA}^{-1}$

FIG. 3. Radial integrals for Dirac multipoles that appear in Eq. (2) derived from an atomic code due to R. D. Cowan [22]. Dimensionless variable $w = 12\pi a_o s$, where a_o is the Bohr radius, while the standard variable for radial integrals s is derived from the Bragg angle and neutron wavelength $s = \sin\theta/\lambda$. Green curve shows (h_1) and blue shows $[w \times (j_0)]$. Note that (j_0) is proportional to $1/w$ as the wavevector approaches zero. Atomic wavefunctions are $4f^5$–$5d^1$. Also included in the figure is the standard radial integral $\langle j_0 \rangle$ that appears in the so-called dipole-approximation (Eq. 1) for diffraction by axial dipole moments. Results obtained with our Sm^{3+} $(4f^5)$ wavefunction are denoted by the continuous black curve, to which we added for comparison four values (+) derived from the standard interpolation formula [23].

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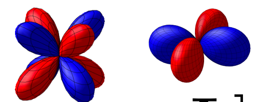

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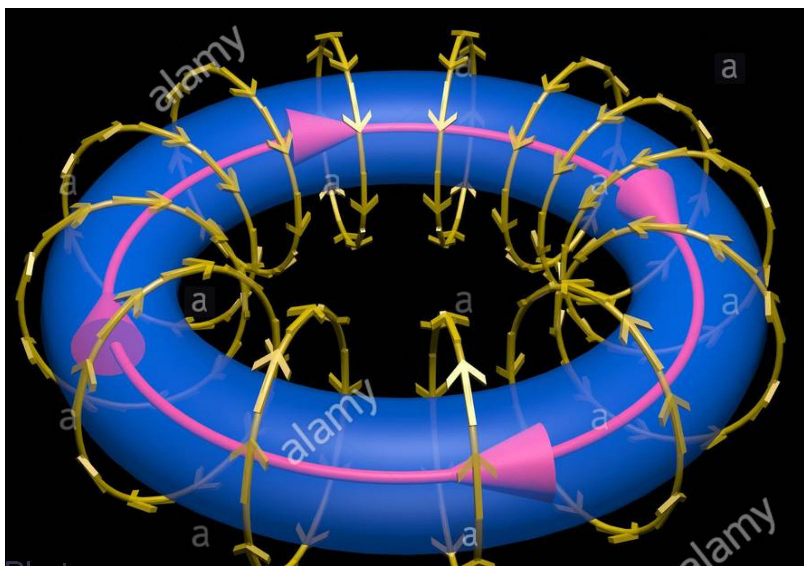

Alamy Stock Photo

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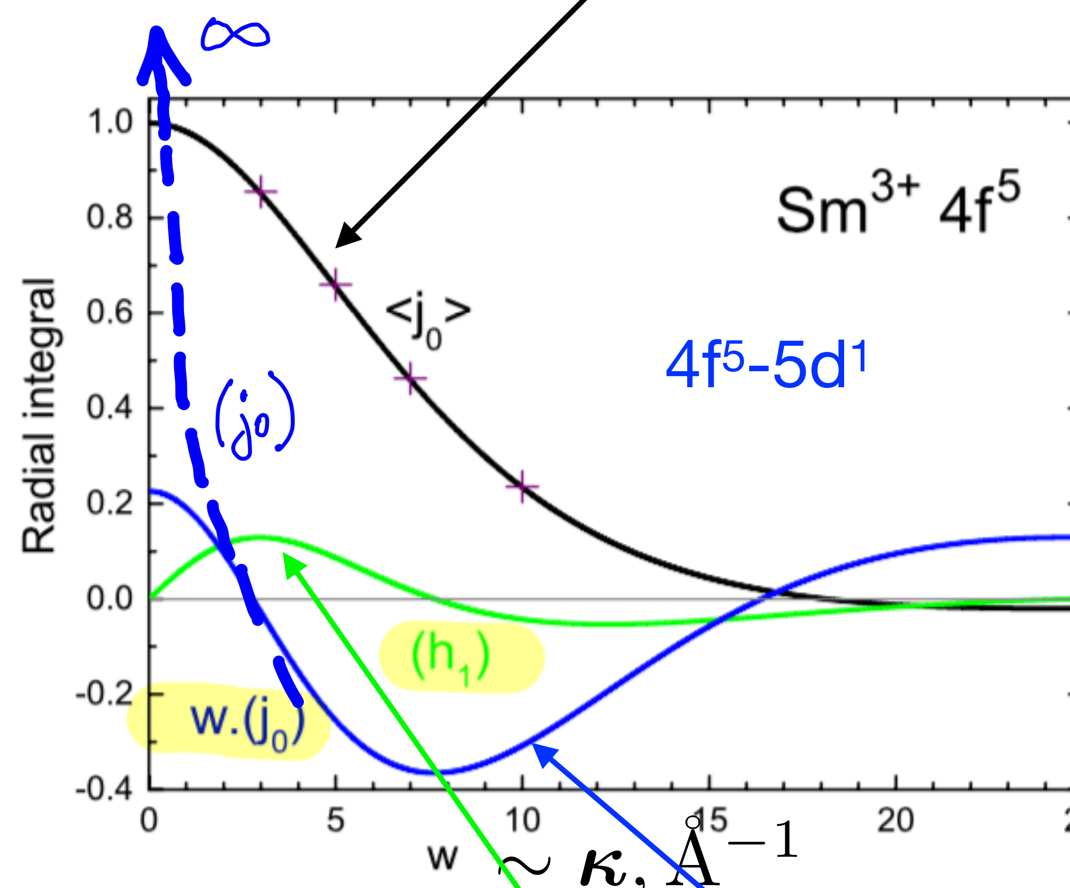


magnetic structure factor for ordered anapoles

$$F^{(-)} \approx -i \exp[\frac{i\pi}{4}(H_o - K_o - L_o)] \times \kappa_\zeta\, [3\langle \Omega_\eta \rangle_S\, (h_1) - \langle \Omega_\eta \rangle_L \boxed{(j_0)}],$$

1. Description of magnetic structures

1.1 How do we describe/classify/predict magnetic symmetries and structures?

1.2 How do we construct all symmetry allowed magnetic structures for a given crystal structure?

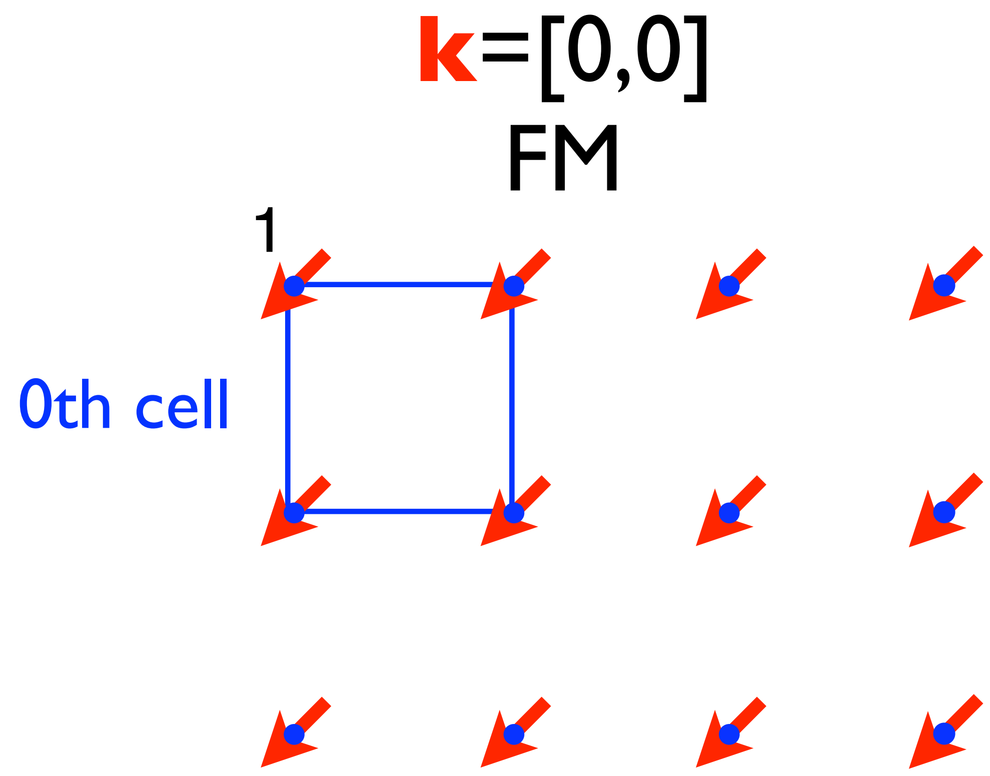
and

2. their determination by neutron diffraction

Magnetic structure factors, practical applications…

Magnetic structure

Examples

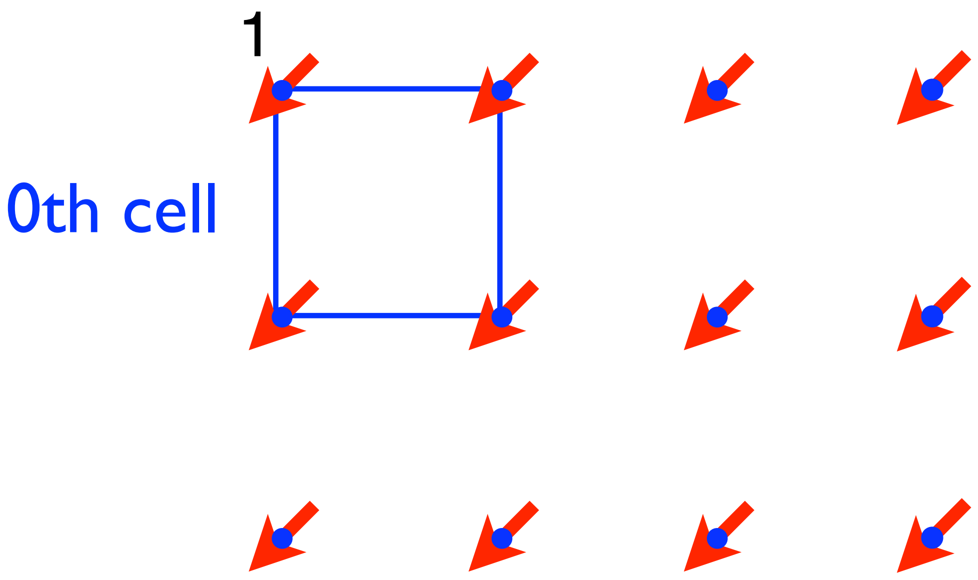


$$\mathbf{S}_{01} = \mathbf{S}_x + \mathbf{S}_y$$

Magnetic structure

Examples

k=[0,0]
FM



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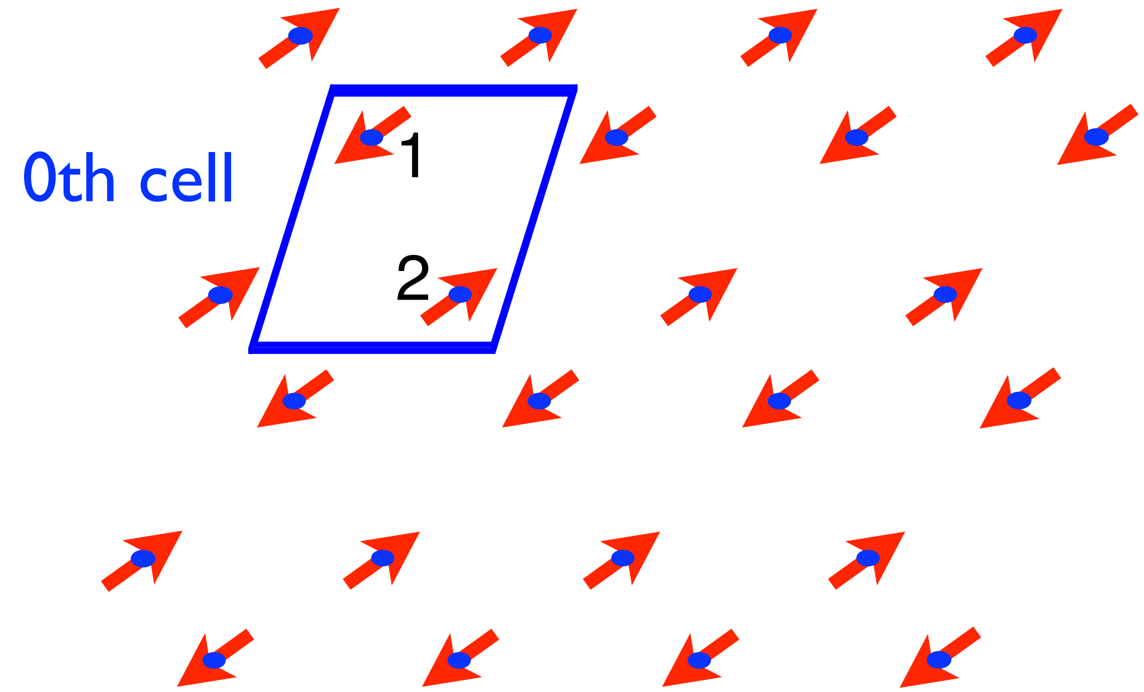
AFM



$$\mathbf{S}_{01} = \mathbf{S}_x + \mathbf{S}_y$$

$$\mathbf{S}_{02} = -\mathbf{S}_{01}$$

Examples of magnetic structures. Propagation vector formalism k≠0.

position of spin in the lattice

Magnetic moment is a real quantity

$$\mathbf{S}(\mathbf{t}_n) = \frac{1}{2}(\mathbf{S}_0 e^{+2\pi i \mathbf{t}_n \mathbf{k}} + \mathbf{S}_0^* e^{-2\pi i \mathbf{t}_n \mathbf{k}}) \equiv |S_{0\alpha}| \cos(2\pi \mathbf{t}_n \mathbf{k} + \phi_\alpha)$$

$$\alpha = x, y, z$$

Fourie amplitude is complex (one can not avoid this)

$$\mathbf{S}_0 = \mathbf{S}_x e^{i\phi_x} + \mathbf{S}_y e^{i\phi_y} + \mathbf{S}_z e^{i\phi_z}$$

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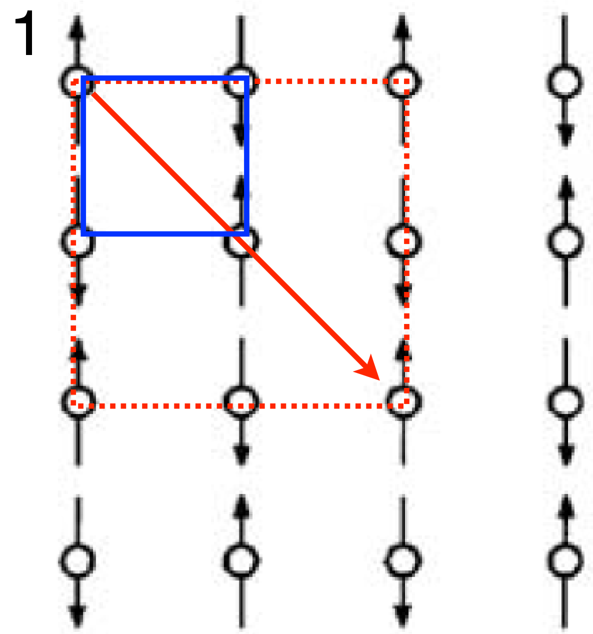
k=[1/2,1/2] AFM



$$\mathbf{S}_{01} = \mathbf{S}_y$$

$$\mathbf{S}(\mathbf{t}_n) = \mathbf{S}_y \sin(2\pi \mathbf{t}_n \mathbf{k})$$
$$= \mathbf{S}_y \sin(\pi(t_{nx} + t_{ny}))$$

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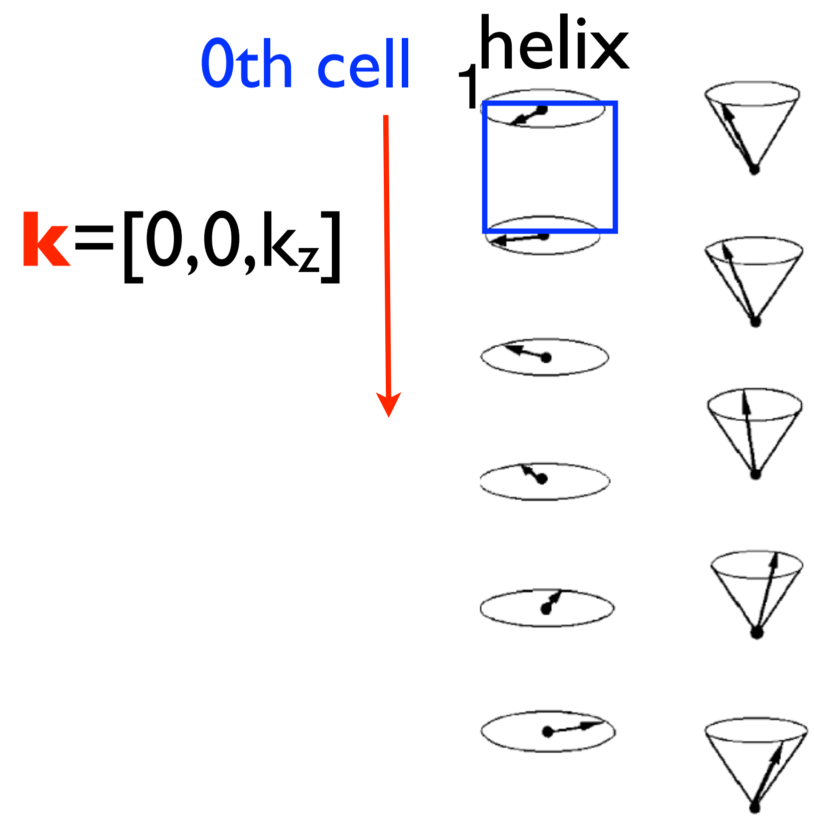
commensurate: k=m/n, m,n: integers

modulated (in)commensurate



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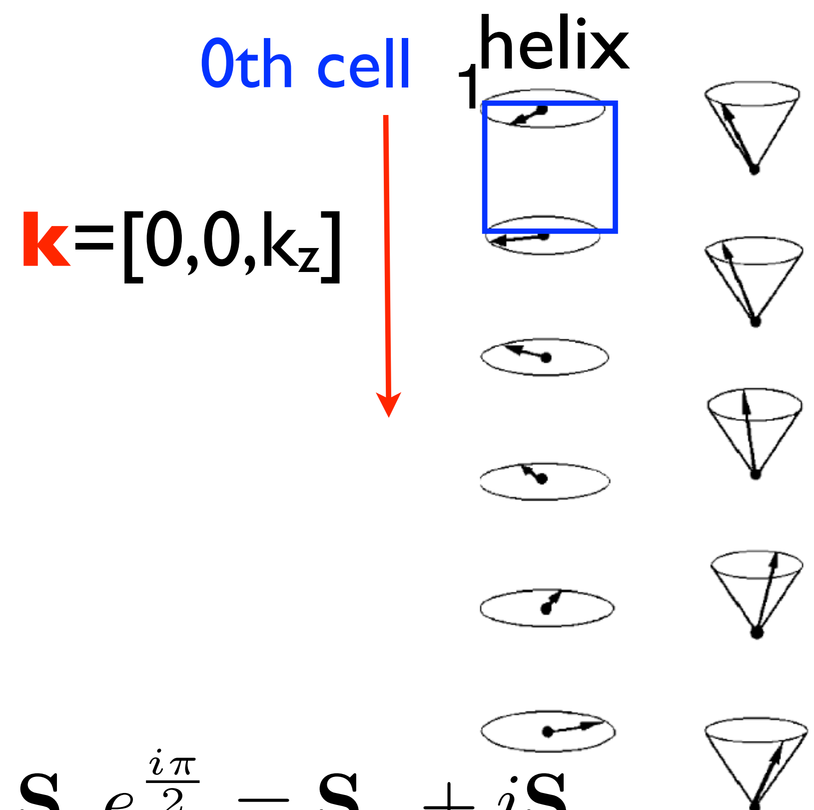

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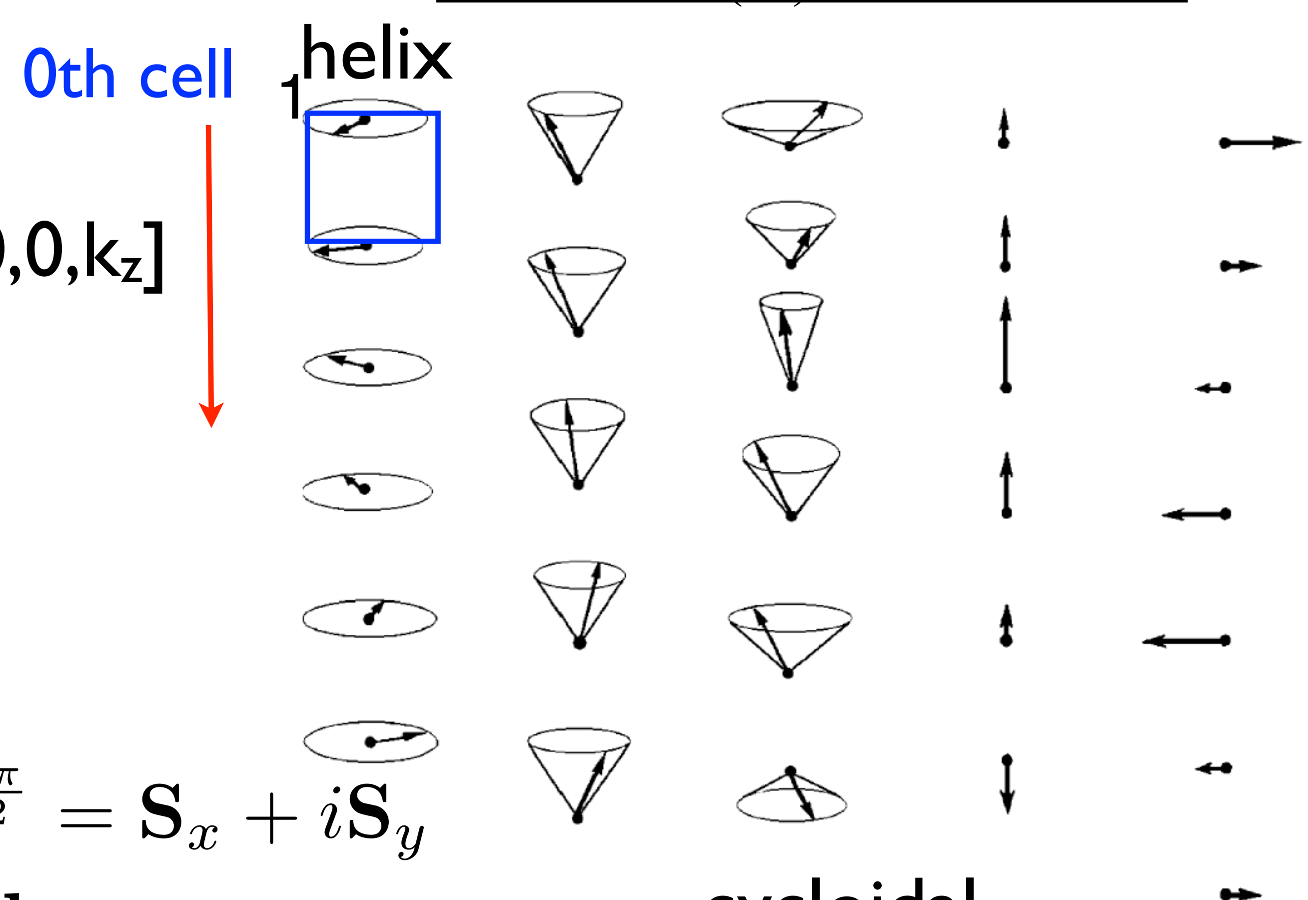
commensurate: k=m/n, m,n: integers

<u>modulated (in)commensurate</u>

k=[0,0,k_z]



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Scattering from the lattice of spins. Magnetic structure factor F(q)

In ND experiment we measure correlators of Fourier transform of magnetic lattice

$$\frac{d\sigma}{d\Omega} \propto \left(\underbrace{\mathbf{Q}_\perp(\mathbf{q}) \cdot \mathbf{Q}_\perp^*(\mathbf{q})}_{\text{structure factor}} + \underbrace{i\mathbf{P} \cdot [\mathbf{Q}_\perp(\mathbf{q}) \times \mathbf{Q}_\perp^*(\mathbf{q})]}_{\text{polarized neutron (chiral) term.}}\right) \cdot \delta(\mathbf{H} \pm \mathbf{k} - \mathbf{q})$$

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structure factor

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Bragg peak at

$$\mathbf{q} = \mathbf{H} \mp \mathbf{k}$$

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Bragg peak at

$$\mathbf{q} = \mathbf{H} \mp \mathbf{k}$$

Sum runs over all atoms in zeroth cell

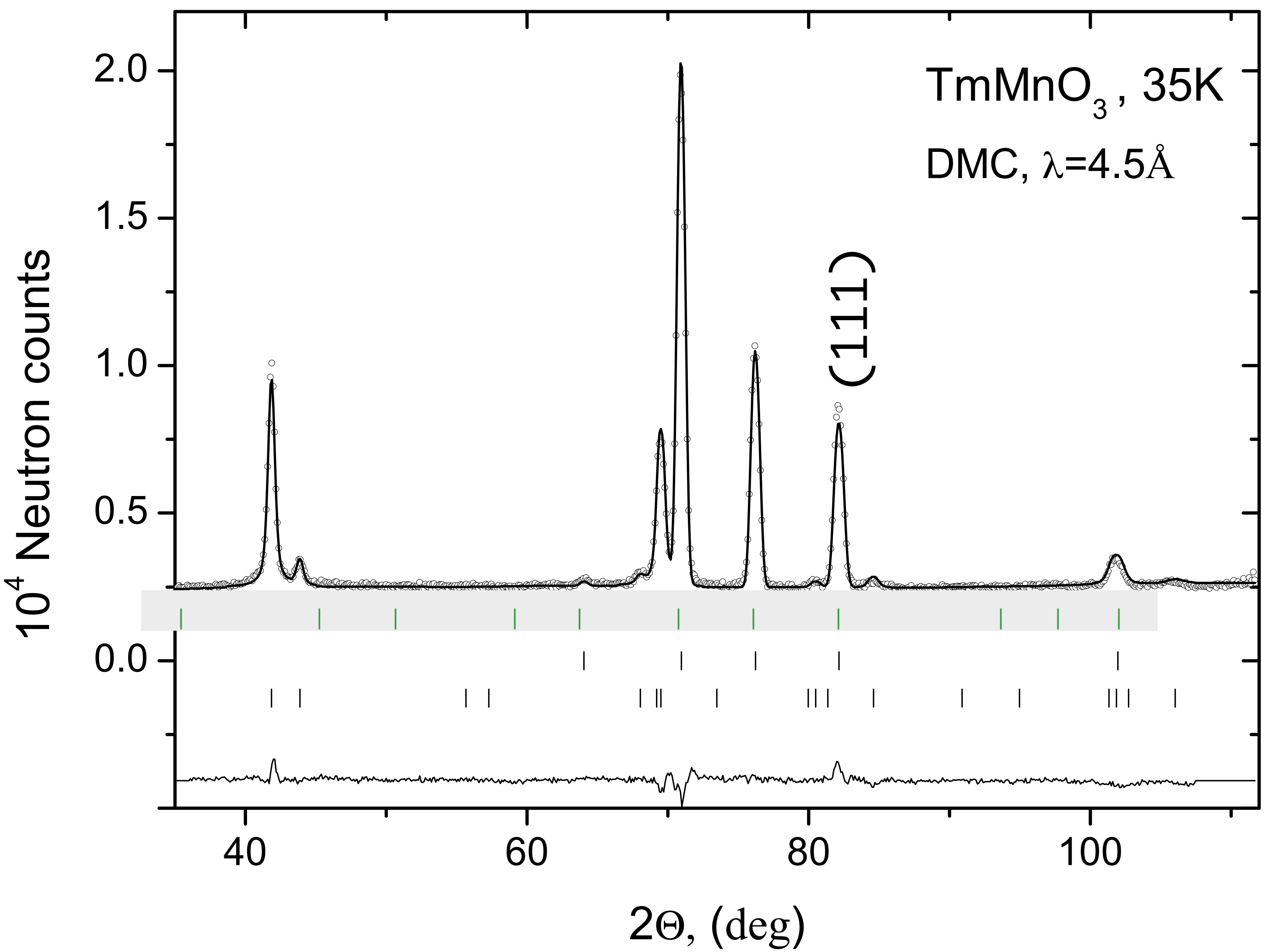
$$\mathbf{Q}_\perp(\mathbf{q})_{-\boldsymbol{k}} = \sum_j \frac{1}{2}\mathbf{S}_{0j\perp} \cdot \exp(i\mathbf{r}_j\mathbf{q}) \qquad \mathbf{Q}_\perp(\mathbf{q})_{+\boldsymbol{k}} = \sum_j \frac{1}{2}\mathbf{S}^*_{0j\perp} \exp(i\mathbf{r}_j\mathbf{q})$$

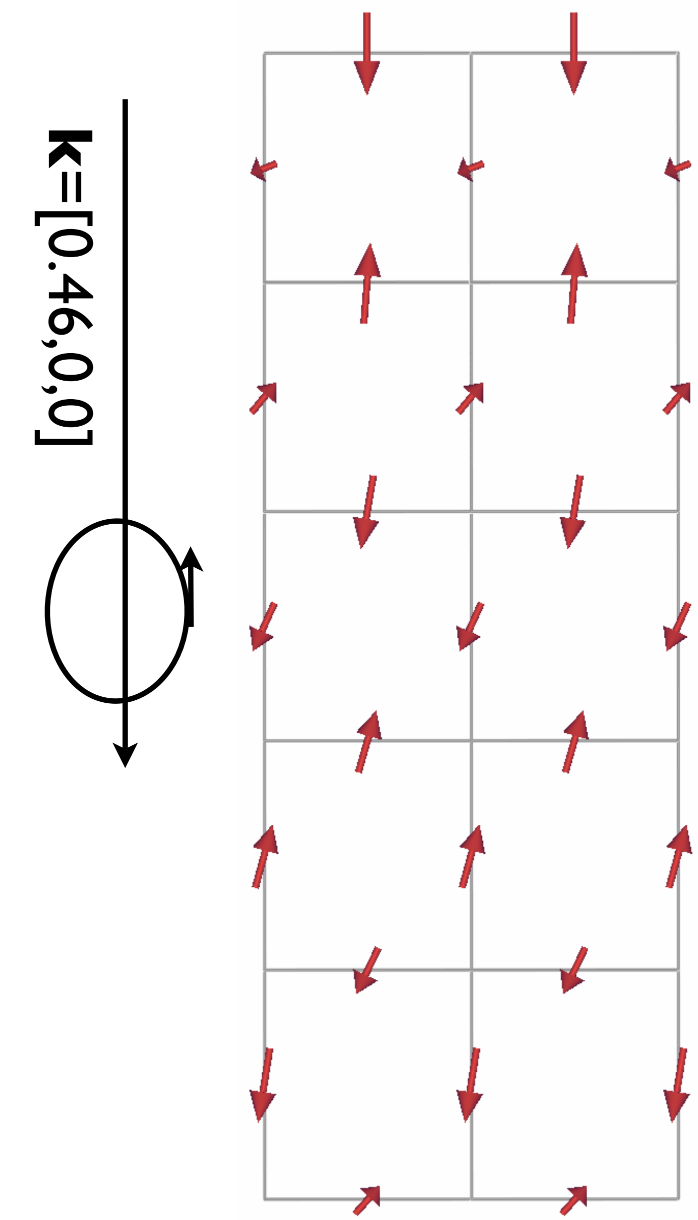
Complex amplitude of spin modulation perpendicular to **q**

position of spin in the zeroth cell

Example of modulated incommensurate structure and diffraction pattern

propagation vector **k**=[0.45,0,0]





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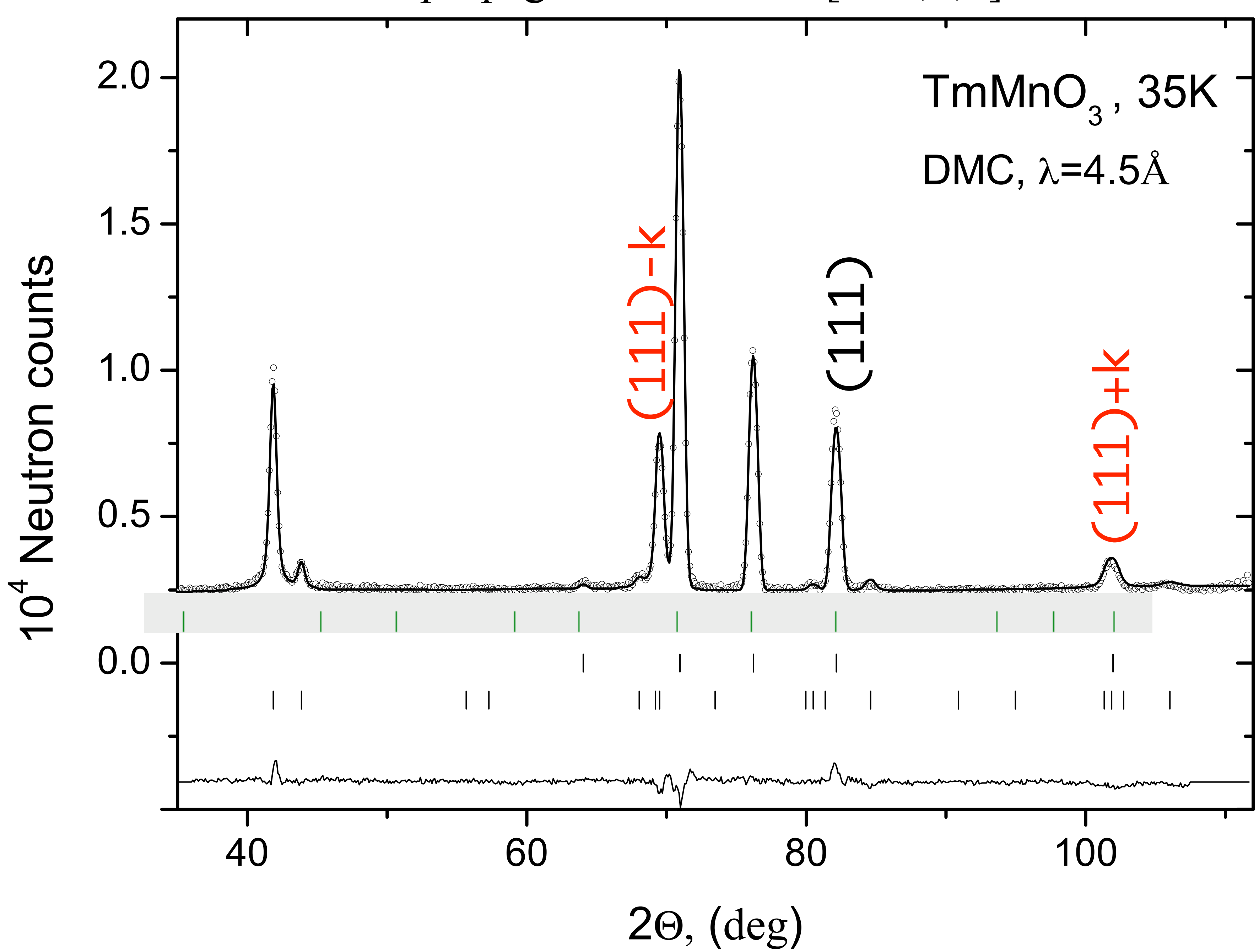

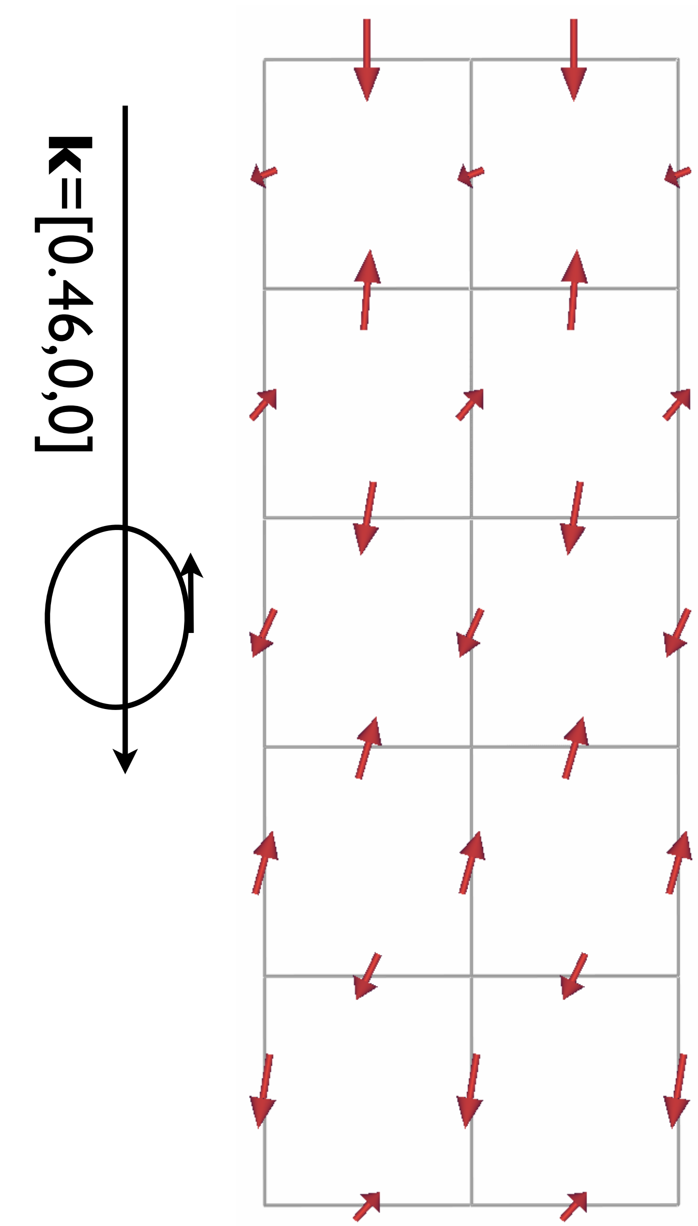

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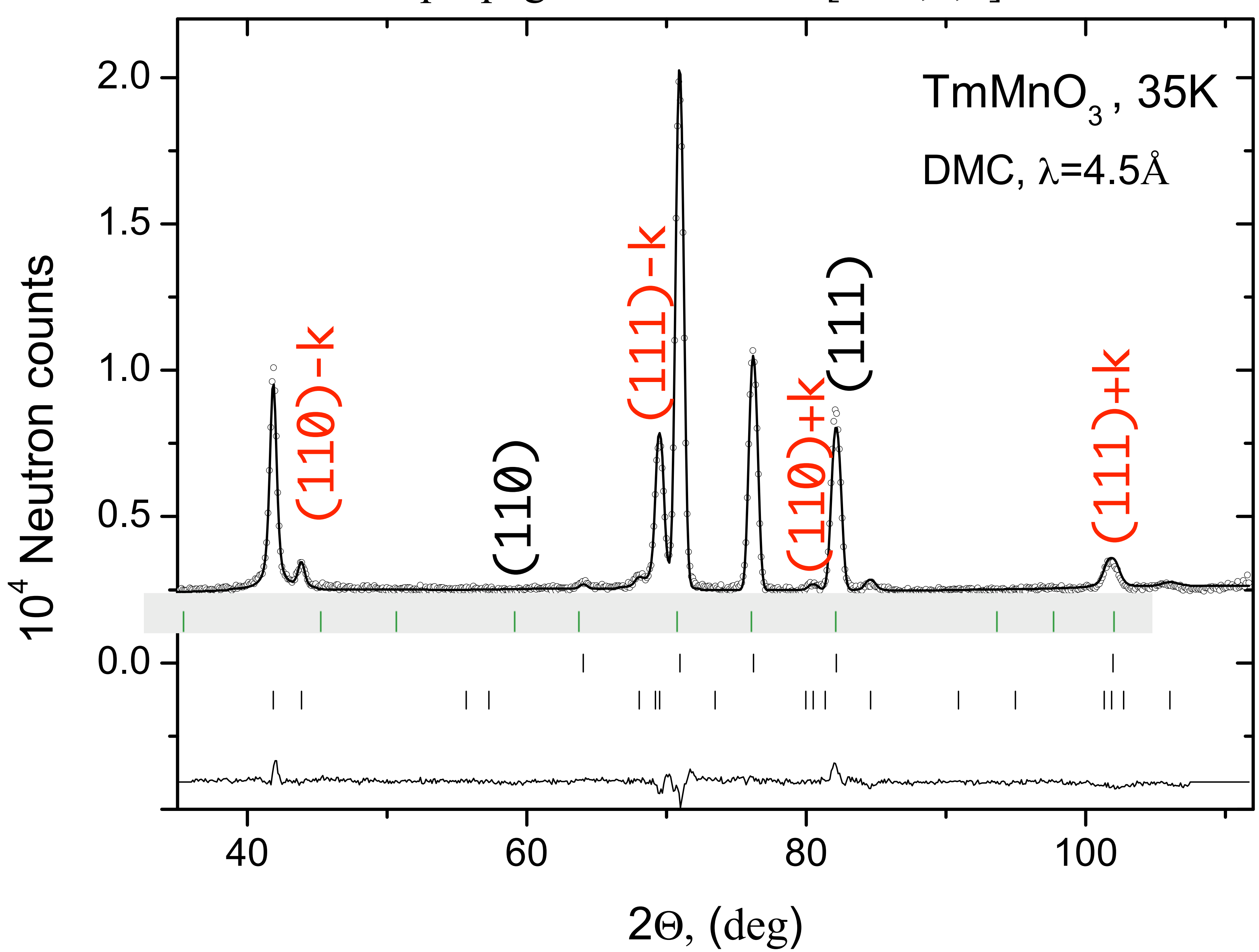
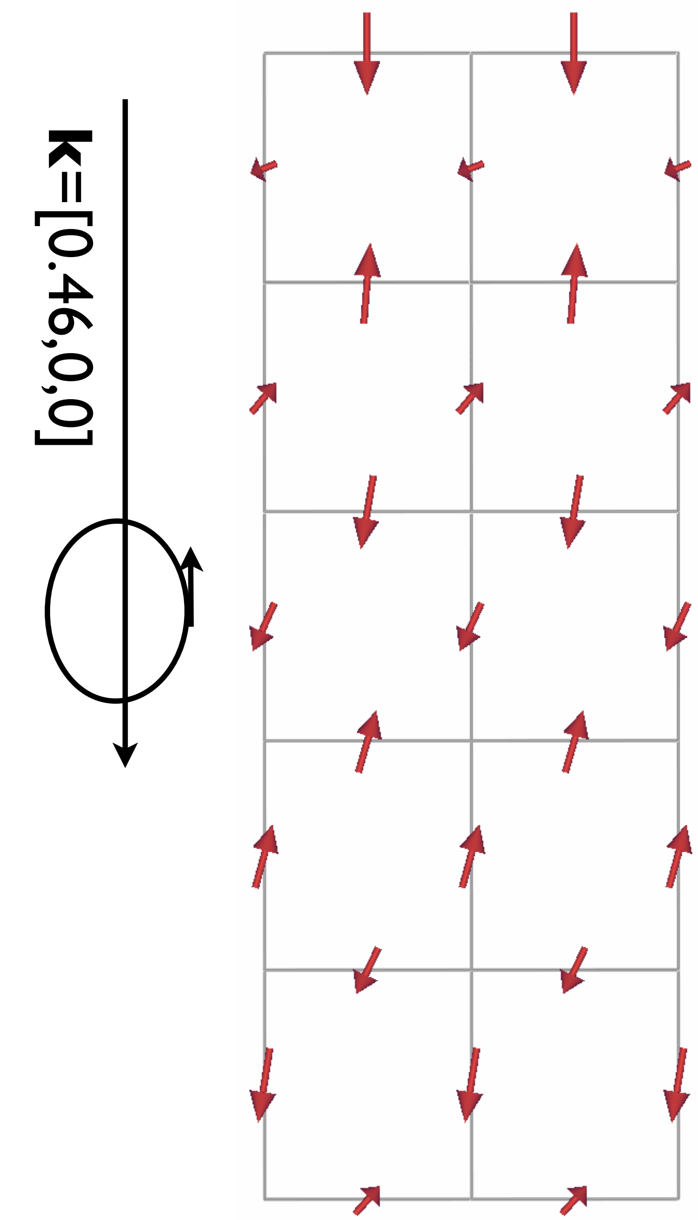




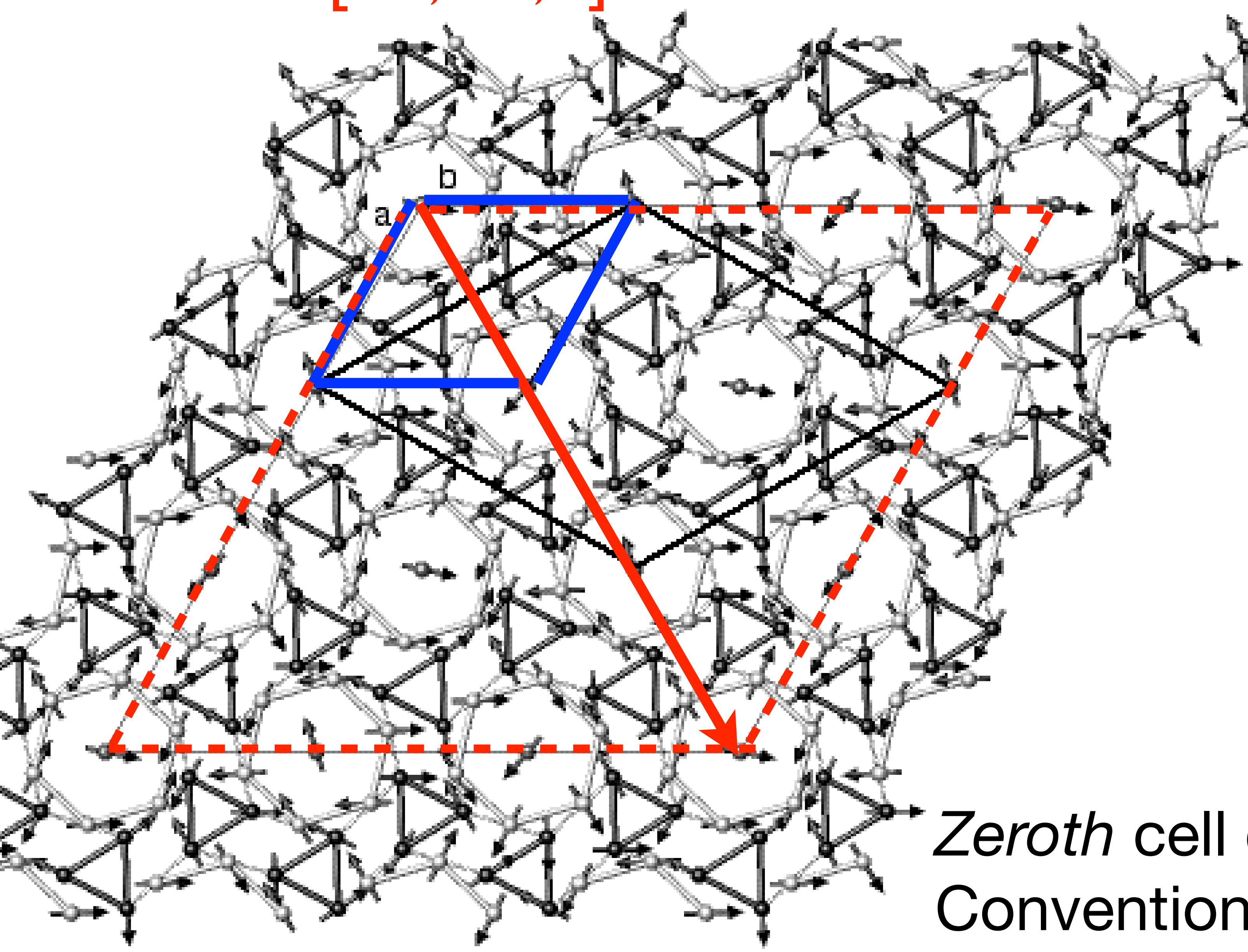
Example of <u>commensurate</u> magnetic structure

Antiferromagnetic (à la cycloidal spiral) three sub-lattice ordering in $Tb_{14}Au_{51}$

commensurate: k=m/n, m,n: integers

P6/m

k-vector=[1/3, 1/3, 0]



Zeroth cell: only 5 magnetic modes, i.e. 5 mixing coefficients C to find from experiment.

Zeroth cell contains 14 spins of Tb^{3+}
Conventional magnetic unit cell contains 126 spins of Tb^{3+}!!

PHYSICAL REVIEW B 72, 134413 (2005)

Short note on non-polarized neutron diffraction

magnetic

nuclear

$$I^{++} \propto \left\langle |\mathbf{Q}_\perp \boldsymbol{\sigma}_n + F|^2 \right\rangle_{\sigma_n}$$

average over neutron polarization

Short note on non-polarized neutron diffraction

magnetic nuclear

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average over neutron polarization

$$I \propto \langle (\mathbf{Q}_\perp \boldsymbol{\sigma}_n)(\mathbf{Q}^*_\perp \boldsymbol{\sigma}_n) + FF^* + \underbrace{\boldsymbol{\sigma}_n (F\mathbf{Q}^*_\perp + F^*\mathbf{Q}_\perp)}_{0} \rangle_{\boldsymbol{\sigma}_n}$$

no magnetic/nuclear interference

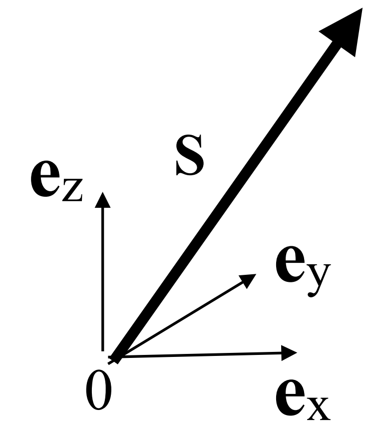
$$I \propto |\mathbf{Q}_\perp|^2 + |F|^2$$

Magnetic and nuclear scattering are completely independent and can be treated as two independent phases in the data analysis (Rietveld refinement)

Introduction to irreducible representations irreps and magnetic Shubnikov groups

Point groups. Magnetic moment rotations in 3D space. Notation of the group representation. Improper rotations.

3-dimensional vector space of classical spin $\mathbf{s} = \sum_{j=x,y,z} s_j \mathbf{e}_j$



Rotation matrices can be used to construct **3-dimensional representation matrices** of proper rotations

$$\varphi_z \begin{pmatrix} \cos\varphi & -\sin\varphi & 0 \\ \sin\varphi & \cos\varphi & 0 \\ 0 & 0 & 1 \end{pmatrix}$$

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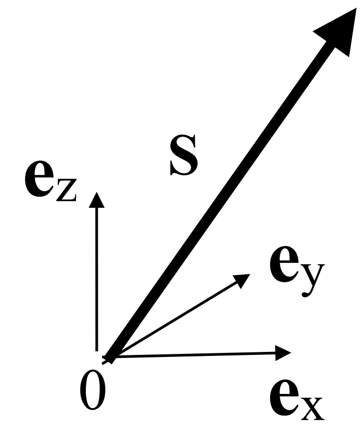

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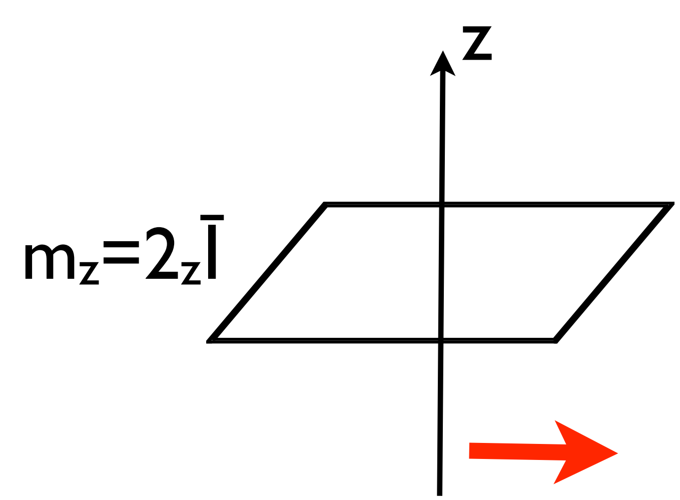
Note: For improper rotations such as inversion (I) or mirror plane we should remember that spin is an axial vector.

$$\mathbf{S} = \text{"}[\mathbf{v} \times \mathbf{r}]\text{"}$$

$$\bar{1}\mathbf{s} = \mathbf{s}$$



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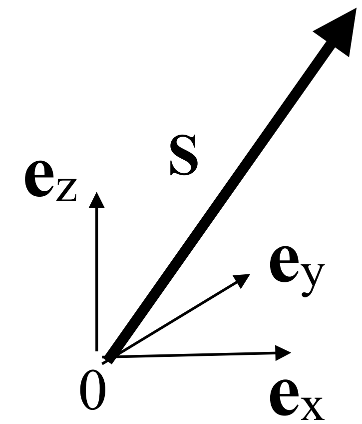

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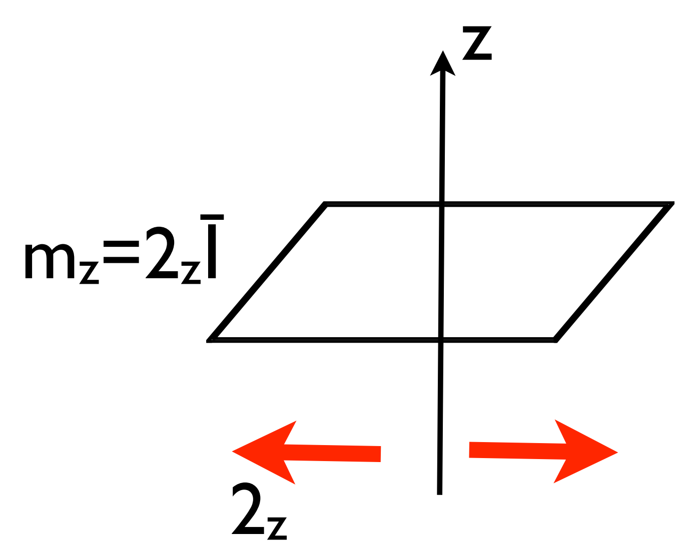
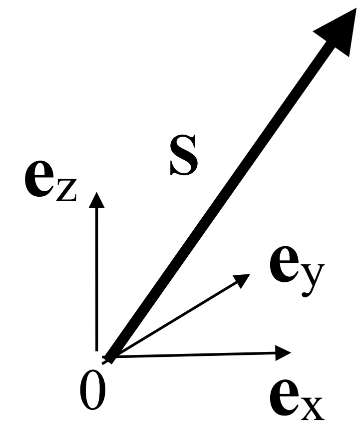


Point groups. Magnetic moment rotations in 3D space. Notation of the group representation. Improper rotations.

3-dimensional vector space of classical spin $\mathbf{s} = \sum_{j=x,y,z} s_j \mathbf{e}_j$



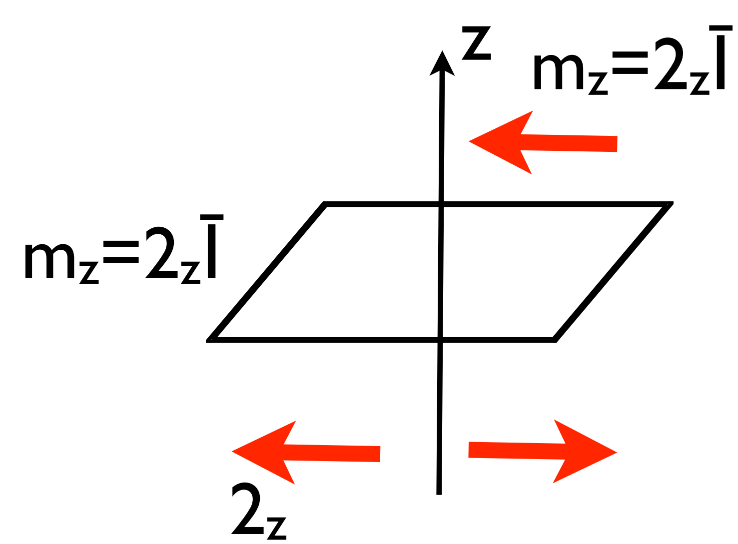
Rotation matrices can be used to construct **3-dimensional representation matrices** of proper rotations

$$\varphi_z \begin{pmatrix} \cos\varphi & -\sin\varphi & 0 \\ \sin\varphi & \cos\varphi & 0 \\ 0 & 0 & 1 \end{pmatrix}$$

Note: For improper rotations such as inversion (I) or mirror plane we should remember that spin is an axial vector.

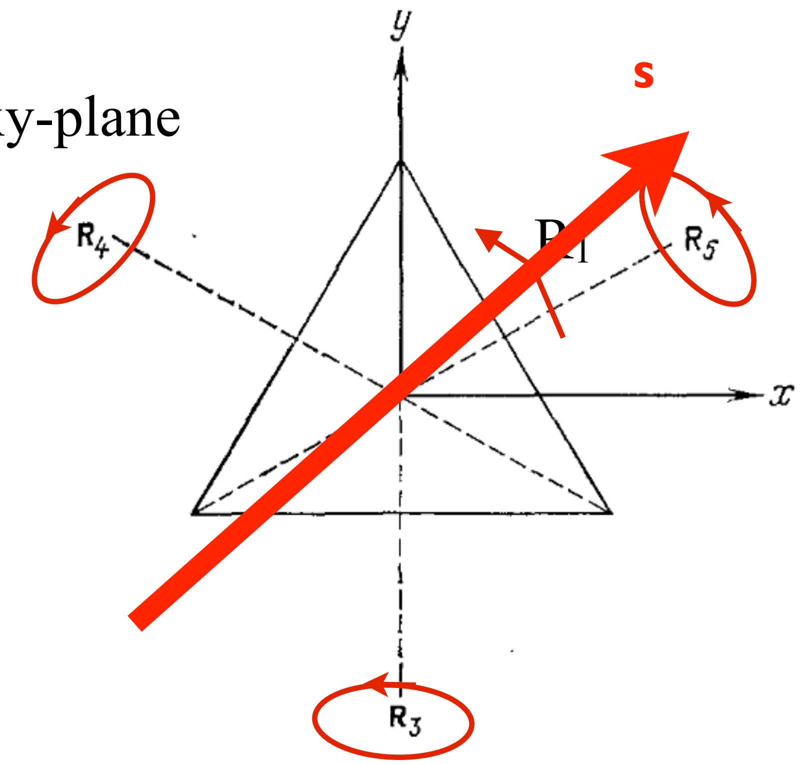
$$\mathbf{S} = \text{"}[\mathbf{v} \times \mathbf{r}]\text{"}$$

$$\bar{1}\mathbf{s} = \mathbf{s}$$



Representation of point group 32 in 3D rotation space of spin S
Example

6 symmetry elements (rotations):
R0=E, $R_1=2\pi/3$, $R_2=4\pi/3$ around z, R_3, R_4, R_5, = π around resp. axes in xy-plane



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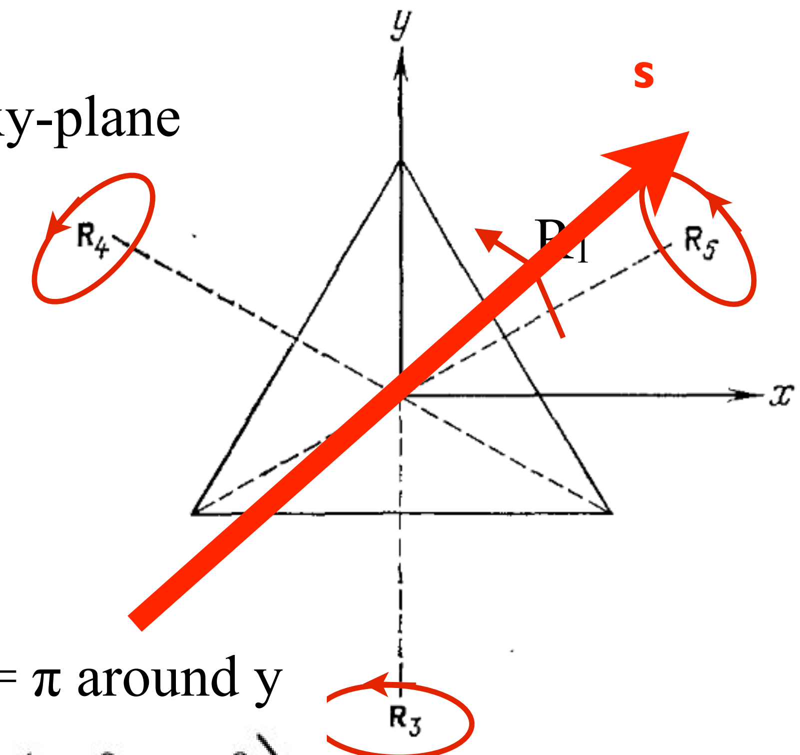

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1. **<u>3-</u>**dimensional representation

$R_1=2\pi/3$ | $R_2=4\pi/3$ around z | R_3 = π around y

$$T(R_1) = \begin{pmatrix} -\frac{1}{2} & -\sqrt{\frac{3}{4}} & 0 \\ \sqrt{\frac{3}{4}} & -\frac{1}{2} & 0 \\ 0 & 0 & 1 \end{pmatrix} \quad T(R_2) = \begin{pmatrix} -\frac{1}{2} & \sqrt{\frac{3}{4}} & 0 \\ -\sqrt{\frac{3}{4}} & -\frac{1}{2} & 0 \\ 0 & 0 & 1 \end{pmatrix} \quad T(R_3) = \begin{pmatrix} -1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -1 \end{pmatrix} \quad \ldots \text{ etc}$$

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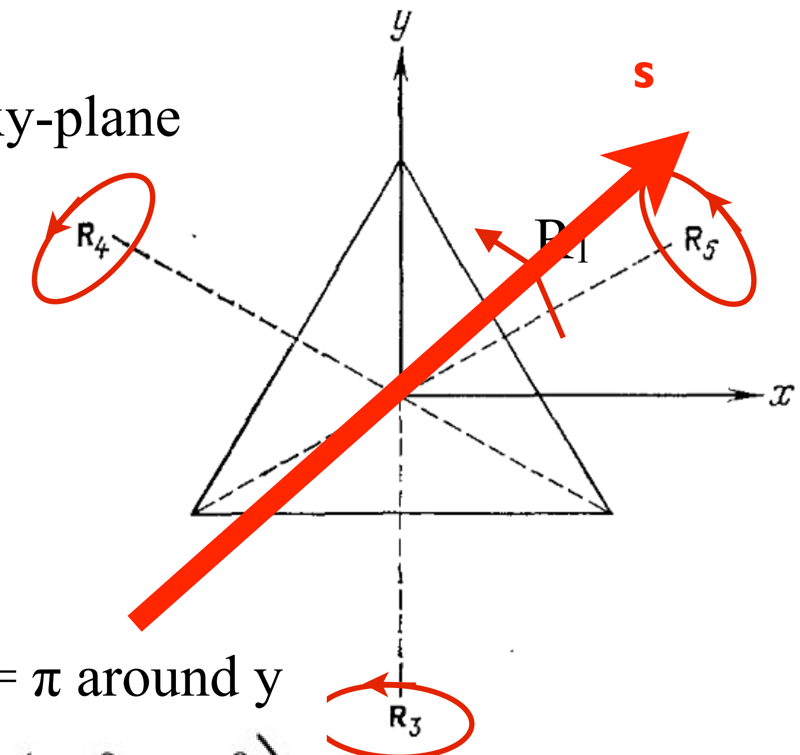

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2. By taking the one dimensional space of vector $\mathbf{e}_z$ alone we may generate very simple **one-**dimensional representation

$$T^{(2)}(R_1) = 1,\ T^{(2)}(R_2) = 1,\ T^{(2)}(R_3) = -1,\ T^{(2)}(R_4) = -1,\ T^{(2)}(R_5) = -1,\ T^{(2)}(E) = 1$$

Space group irreps, examples
dimensions up to 6 (cf. 3 for point groups)

Example 1

***Pnma* at X-point [1/2,0,0] of BZ, two 2D-irreps, e.g. mX1**
g: Group elements, *G*: matrices or irreducible representation *irrep*

$$g = \quad 1 \quad 2_x \quad 2_y \quad 2_z \quad -1 \quad n \quad m \quad a$$

$$G = \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \begin{bmatrix} 0 & -1 \\ 1 & 0 \end{bmatrix} \begin{bmatrix} -1 & 0 \\ 0 & 1 \end{bmatrix} \begin{bmatrix} 0 & -1 \\ -1 & 0 \end{bmatrix} \begin{bmatrix} -1 & 0 \\ 0 & 1 \end{bmatrix} \begin{bmatrix} 0 & -1 \\ -1 & 0 \end{bmatrix} \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \begin{bmatrix} 0 & -1 \\ 1 & 0 \end{bmatrix}$$

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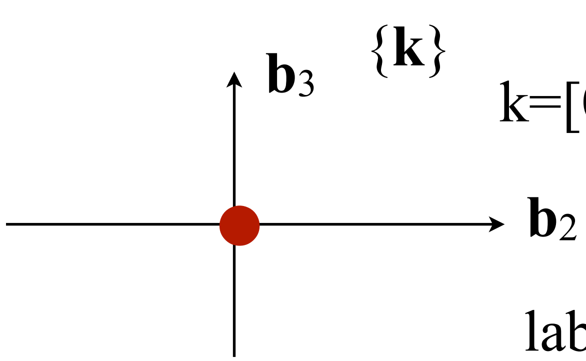
Example 2 *Pnma* k=[0,0,0], k19

irreps: eight 1D $\tau_1, \tau_2, \tau_3, \tau_4, \tau_5, \tau_6, \tau_7, \tau_8$



k=[0,0,0]

label Γ

$G_k = G$

$d^{\mathbf{k}\nu}(g)$

g	/2	/3	/4	/25	/26	/27	/28
$\hat{\tau}1$	1	1	1	1	1	1	1
$\hat{\tau}2$	1	1	1	−1	−1	−1	−1
$\hat{\tau}3$	1	−1	−1	1	1	−1	−1
$\hat{\tau}5$	−1	1	−1	1	−1	1	−1
$\hat{\tau}7$	−1	−1	1	1	−1	−1	1

$\hat{\tau}4 = \hat{\tau}3 \times \hat{\tau}2,\ \hat{\tau}6 = \hat{\tau}5 \times \hat{\tau}2,\ \hat{\tau}8 = \hat{\tau}7 \times \hat{\tau}2$

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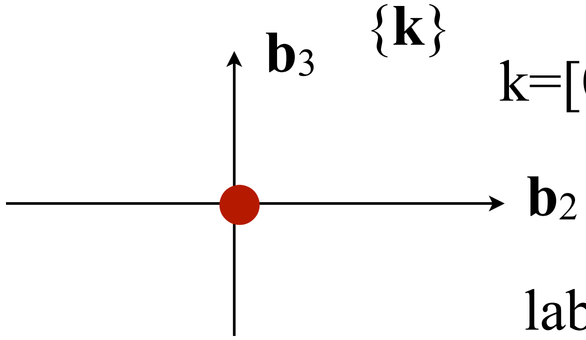

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Example 3

Higher dimensions: Ia3d (#230) k=[1,0,0]: 1(6D) ⊕ 3(2D)
k=[1/2,1/2,1/2]: 1(4D) ⊕ 2(2D)

Representation* Analysis (RA). Propagation vector k formalism. Magnetic mode S_0 is specified in zeroth block of the cell == parent cell without centering translations

Magnetic moment below a phase transition

$$\mathbf{S}(\mathbf{t}_n) = Re\left(C\mathbf{S}_0 e^{2\pi i \mathbf{t}_n \mathbf{k}}\right) \quad \sim \cos(2\pi \mathbf{t}_n \mathbf{k} + \varphi)$$

amplitude or mixing coefficients

magnetic mode

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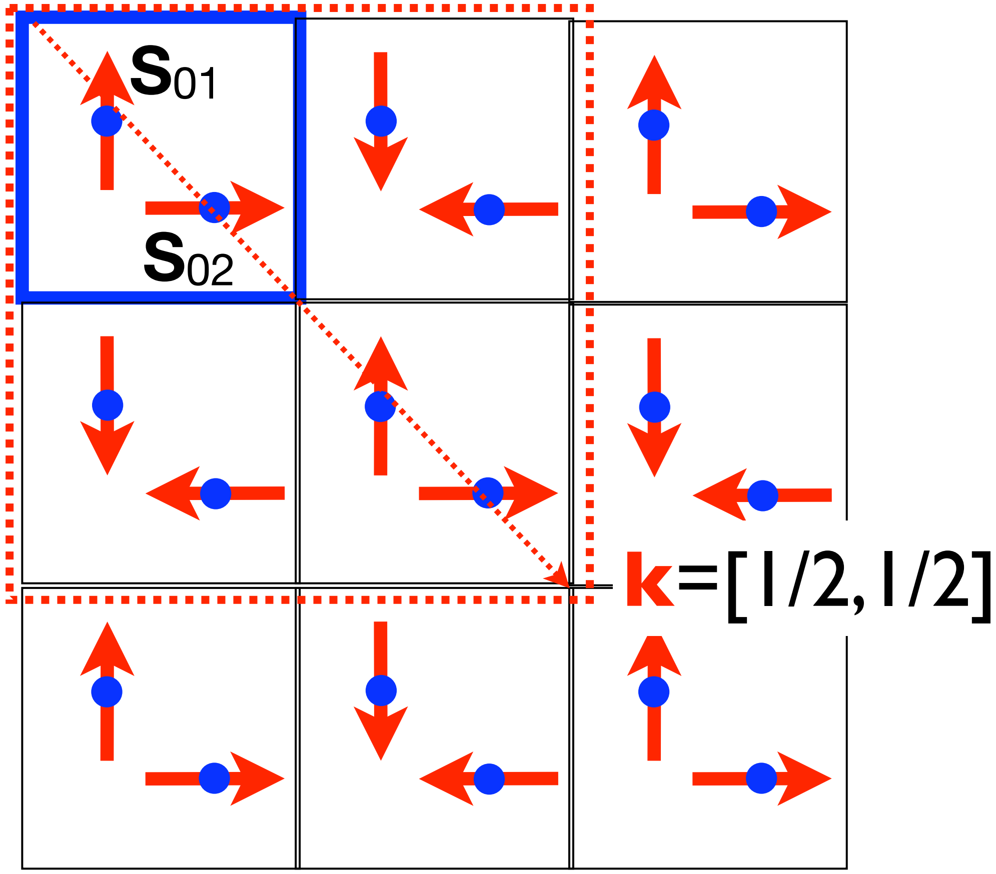
0th cell with many atoms in general



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each group element g --> *matrix* $\tau(g)$ that specifies the spin transformation under element g

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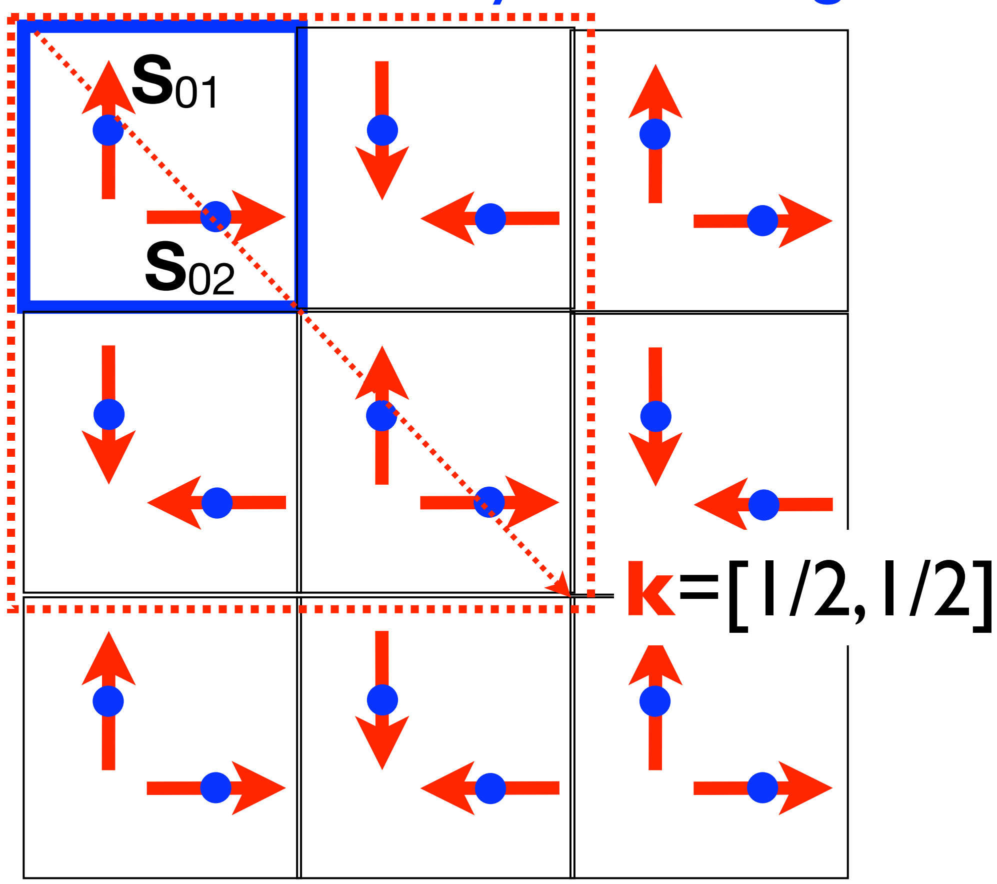

magnetic mode S_0 for chosen irrep* specifies magnetic configuration of all spins in zeroth cell $\longrightarrow$ $\mathbf{S}_0 = \begin{pmatrix} s_{x1} \\ s_{y1} \\ s_{z1} \\ s_{x2} \\ s_{y2} \\ s_{z2} \\ \cdots \\ \cdots \\ \cdots \\ s_{xN} \\ s_{yN} \\ s_{zN} \end{pmatrix}$

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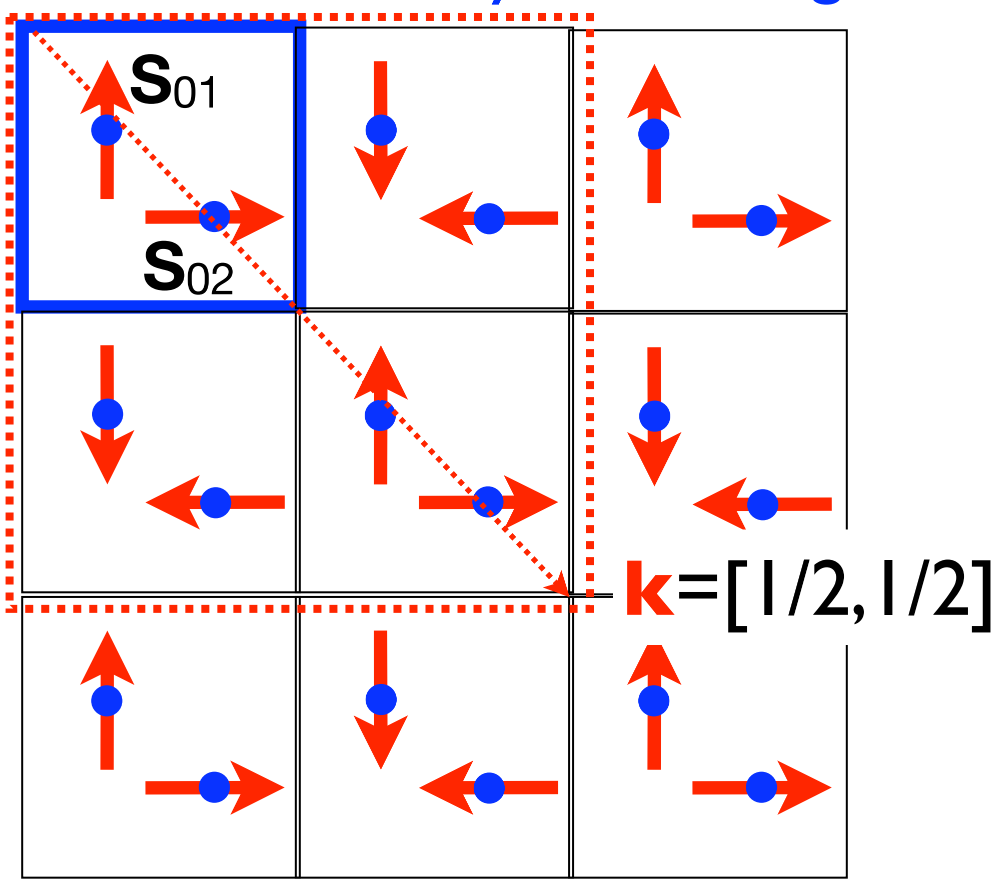

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E.g., atom1 $\mathbf{S}_{01} = \mathbf{e}_y$

atom2 $\mathbf{S}_{02} = \mathbf{e}_x$

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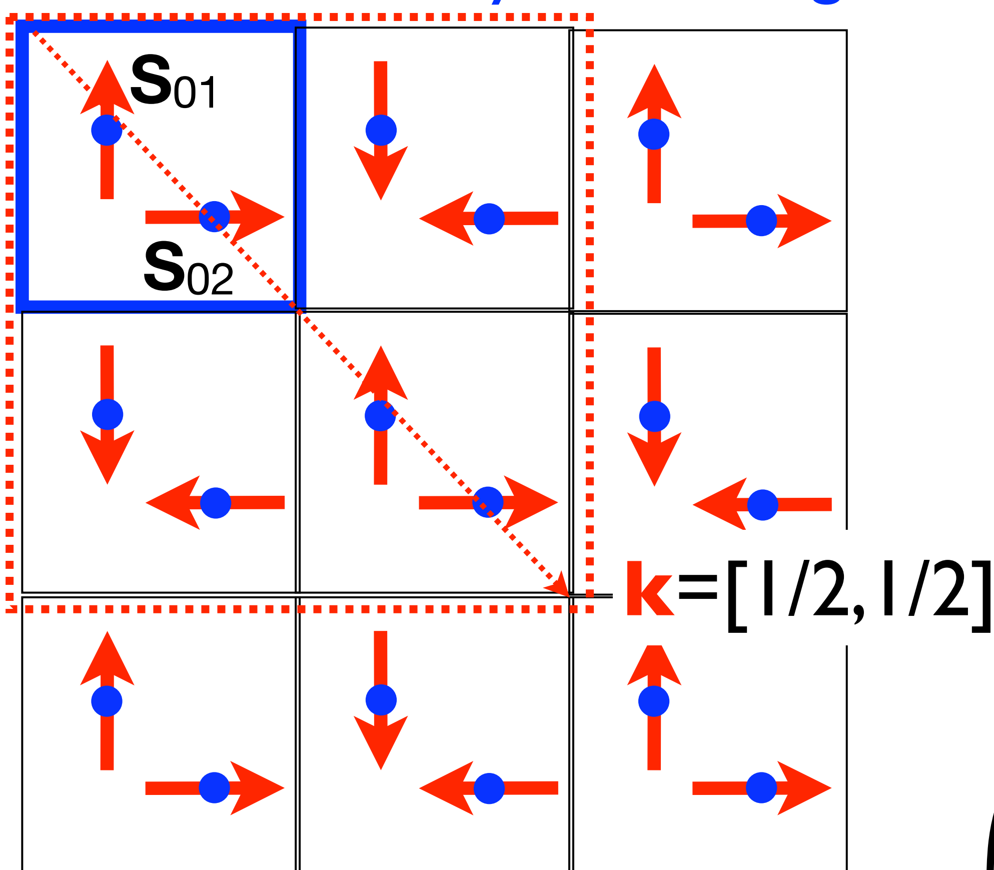

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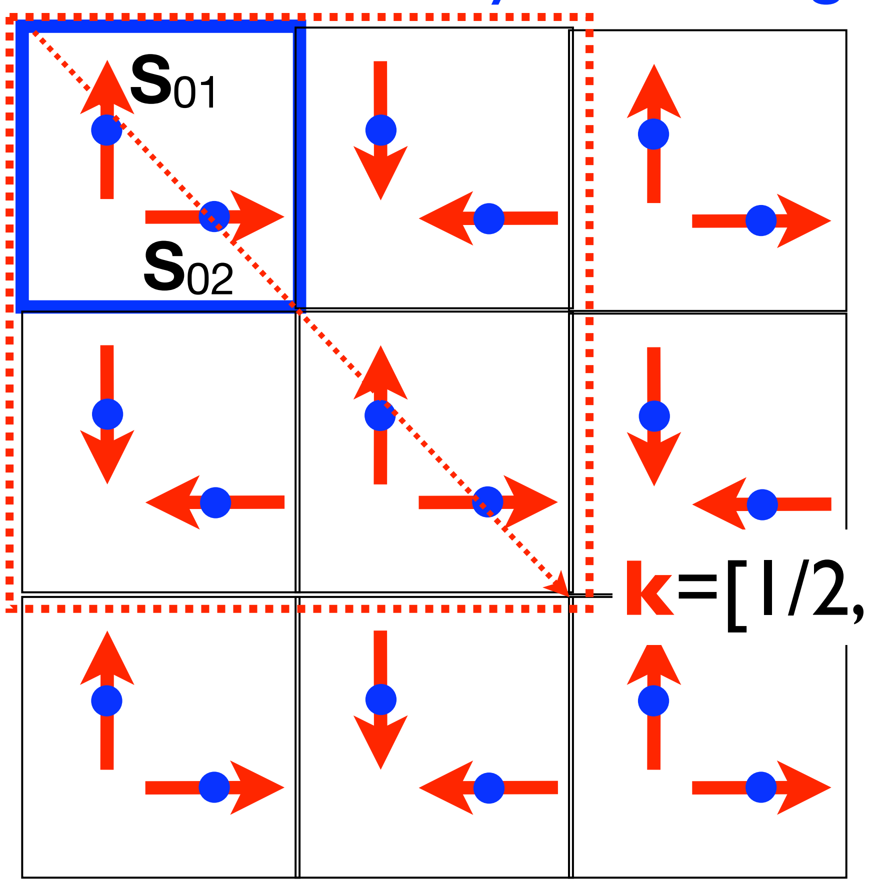

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S_0 and $C = |C|e^{i\varphi}$ are complex quantities

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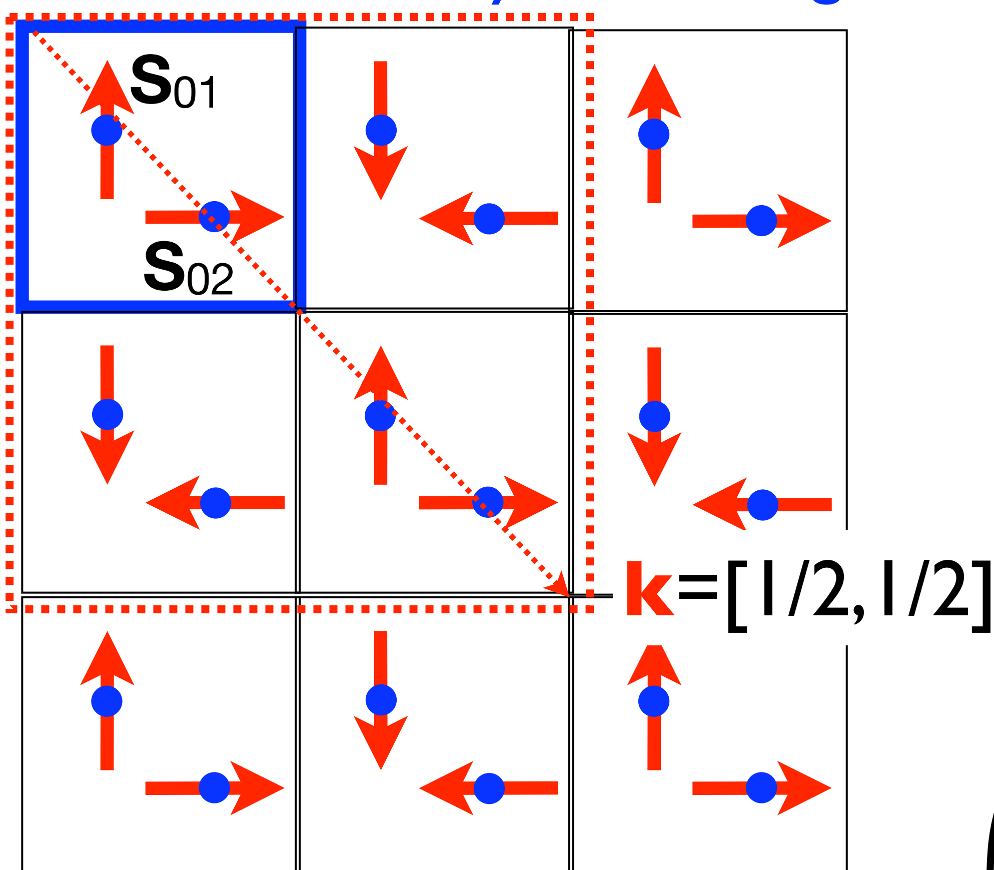

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...

Magnetic symmetry. 1651 3D-Shubnikov (Sh or Ш) space groups

14 Bravias lattice

32 point groups

230 space groups (SG)

antisymmetry: Heesh (1929), Shubnikov (1945).
groups: Zamorzaev (1953, 1957); Belov, Neronova, Smirnova (1955)
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spin reversal operator *R* or color change.
R-group (1,R)

⇒

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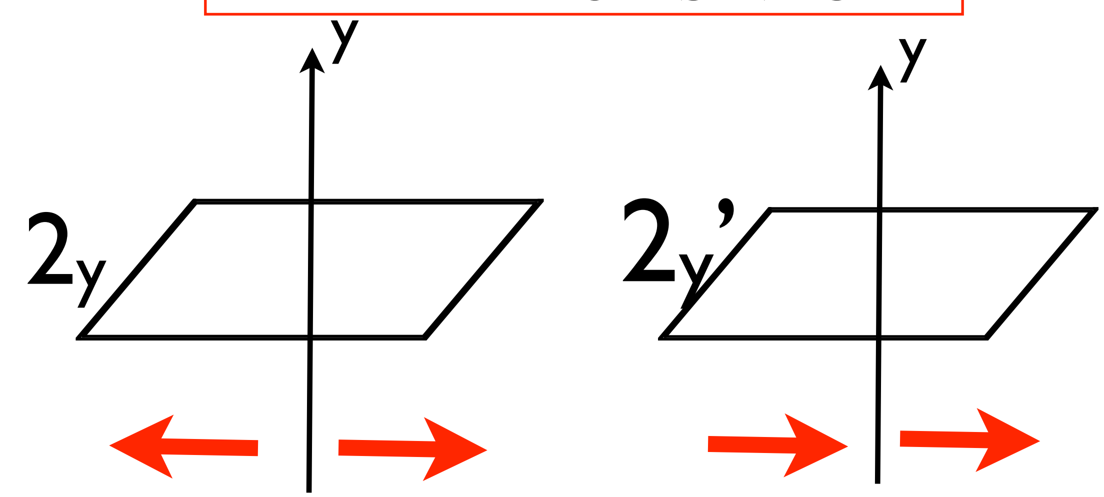

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230 (gray) paramagnetic groups Sh_p
<u>1,1’ ∈ Sh_p</u> ⇒ **S**=0 , e.g. *Pnma1’*

additional elements:
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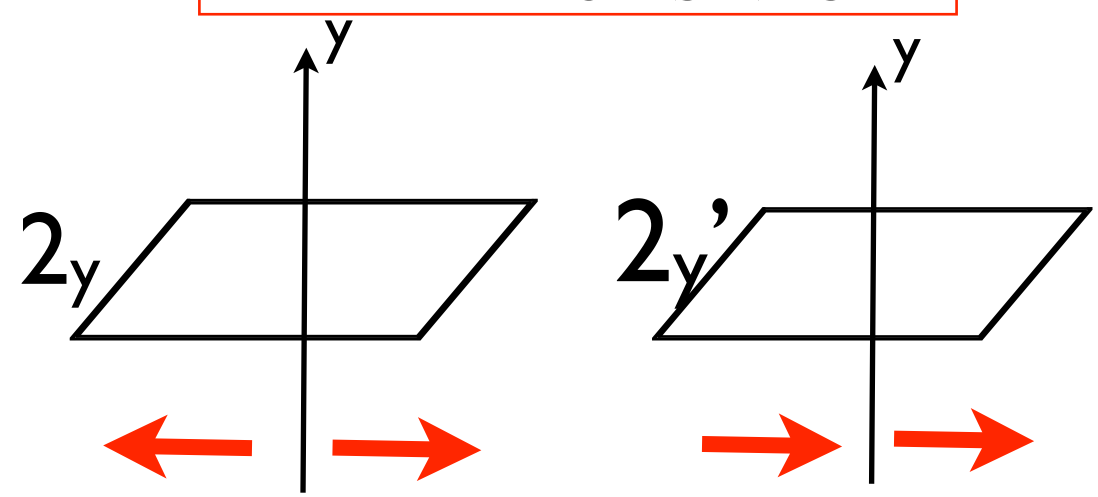

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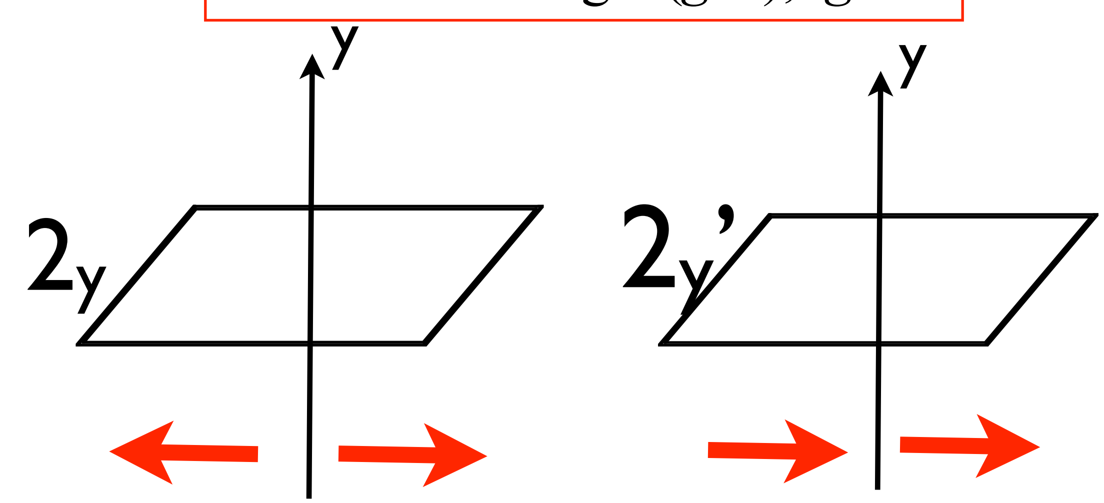

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230 (gray) paramagnetic groups Sh_p
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230 Single-color magnetic groups
<u>no antielements</u>

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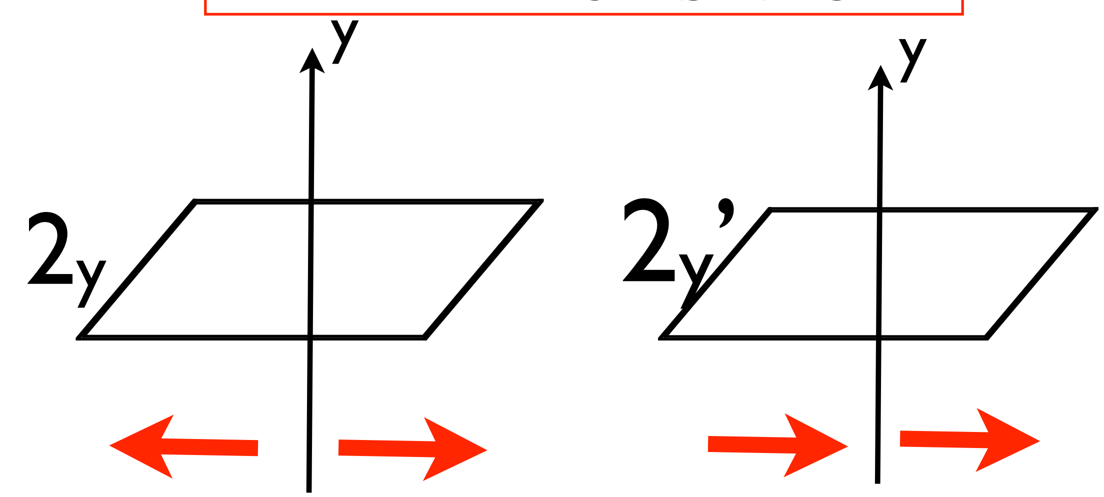

$\mathbf{S} = $ "$[\mathbf{v} \times \mathbf{r}]$"

230 (gray) paramagnetic groups Sh_p
<u>1,1' ∈ Sh_p</u> ⇒ **S**=0 , e.g. *Pnma1'*

230 Single-color magnetic groups
<u>no antielements</u>

1191 black/white magnetic groups that contain additional 'anti-elements' g'=($g \cdot R$) except g=1 (identity).
<u>No primed 1'</u>

antisymmetry: Heesh (1929), Shubnikov (1945).
groups: Zamorzaev (1953, 1957); Belov, Neronova, Smirnova (1955)
spin reversal: Landau and Lifschitz (1957)

Examples of Sh groups

59 $\underline{Pmmn}$	62 $\underline{Pnma}$
$Pm'mn$	$Pn'ma$
$Pmmn'$	$Pnm'a$
* $Pm'm'n$	$Pnma'$
* $Pmm'n'$	* $Pn'm'a$
$Pm'm'n'$	* $Pnm'a'$
$P_{2c}mmn$	* $Pn'ma'$
$P_{2c}m'mn$	$Pn'm'a'$
$P_{2c}m'm'n$	

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Ferromagnetic groups: point symmetry allows FM orientation of spins
Only 275 FM groups out of 1651...

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Ferromagnetic groups: point symmetry allows FM orientation of spins
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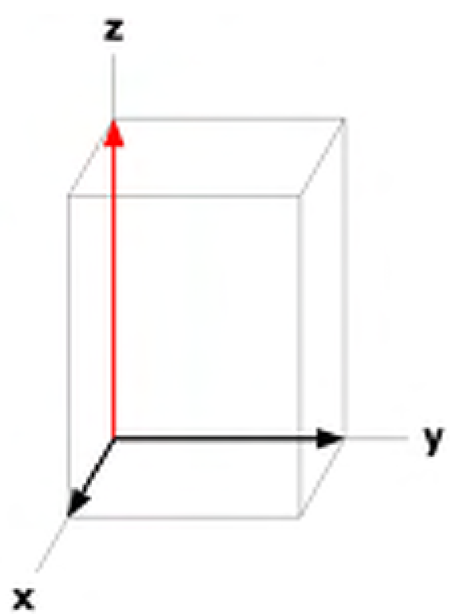
recap:
for 'anti-elements' g'=($g \cdot R$), $g \in G$
g can be a pure translation t, so t' gives centering/doubling

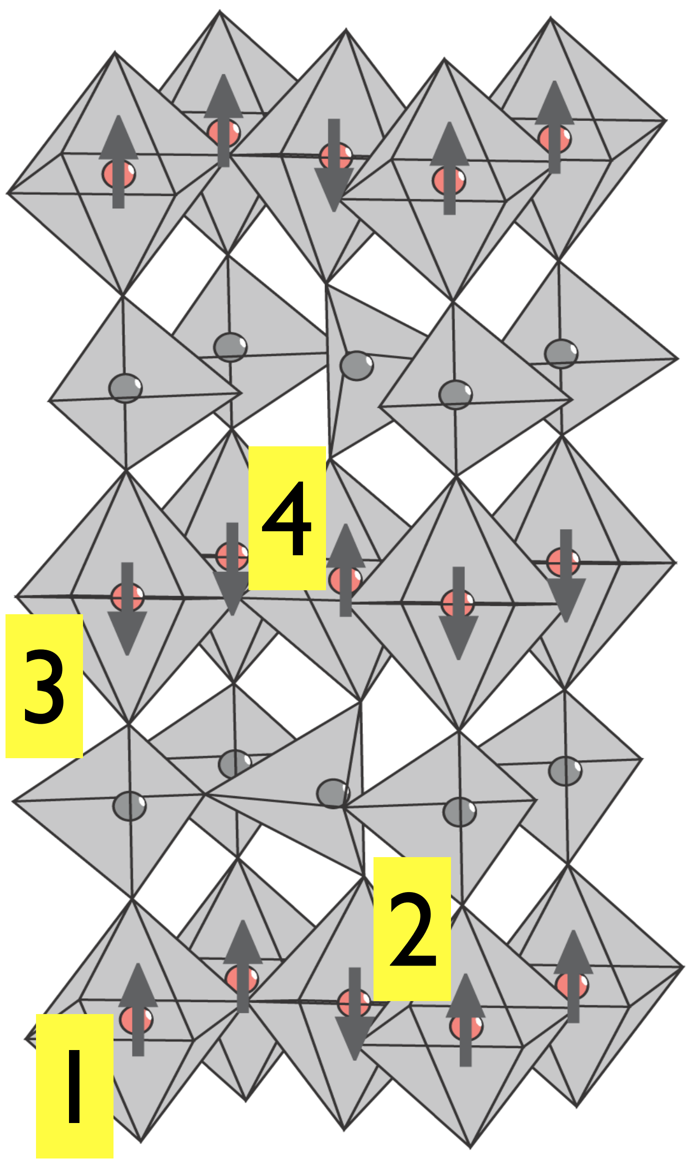


$P_{2c} = P_{a,b,2c}$
$t_\alpha = c = (0,0,1)$

Two ways of description of magnetic structures

Magnetic structure is an axial vector function $\mathbf{S}(\mathbf{r})$ defined on the discreet system of points (atoms), e.g. $\mathbf{S}(\mathbf{r}) = \mathbf{s}(\mathbf{r}_1) \oplus \mathbf{s}(\mathbf{r}_2) \oplus \mathbf{s}(\mathbf{r}_3) \oplus \mathbf{s}(\mathbf{r}_4)$

Crystal with space group *G*



1. How to make $\mathbf{S}(\mathbf{r})$ invariant? Find (new) symmetry elements.
   $g_{new}\,\mathbf{S}(\mathbf{r}) = \mathbf{S}(\mathbf{r})$ to itself, where $g_{new} \in G_{sh}$ subgroup of PG paramagnetic space group: PG=$G\otimes1'$, where 1'=spin/time reversal, G (parent space group).

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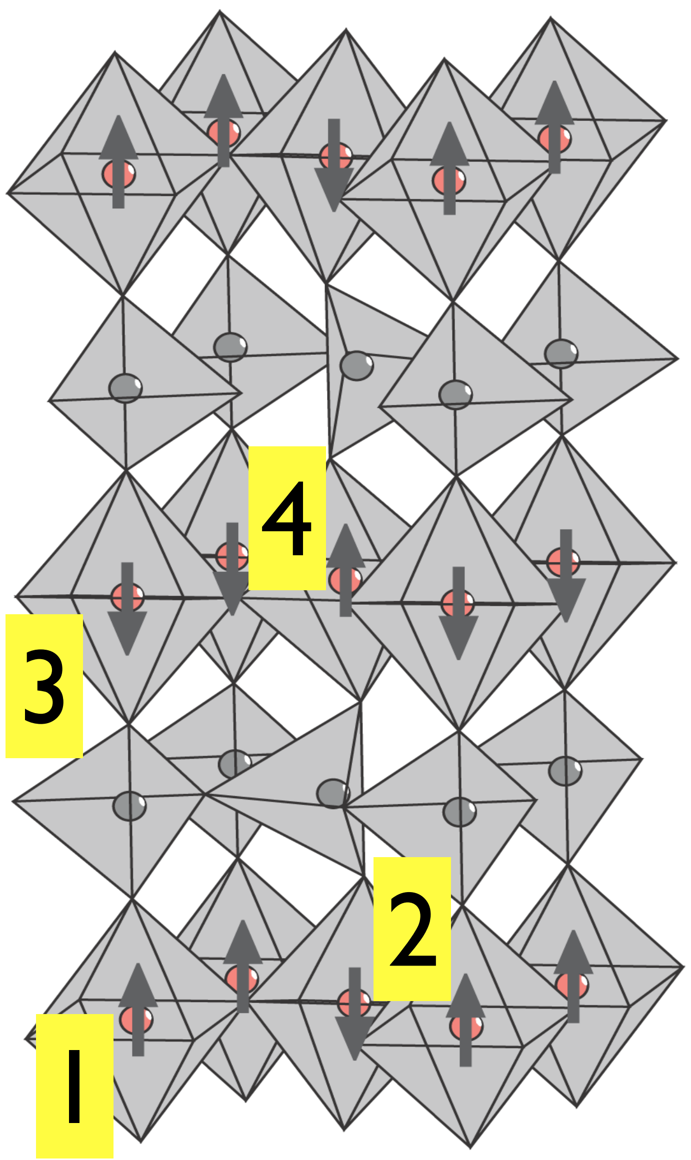

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2. How should $\mathbf{S}(\mathbf{r})$ be transformed under elements of G ?
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87.2.734	I4/m1'
87.3.735	I4'/m
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τ_5	1	-1	1	-1	1	-1	1	-1
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Example of Shubnikov group. Magnetic structure of Iron based superconductor KFeSe

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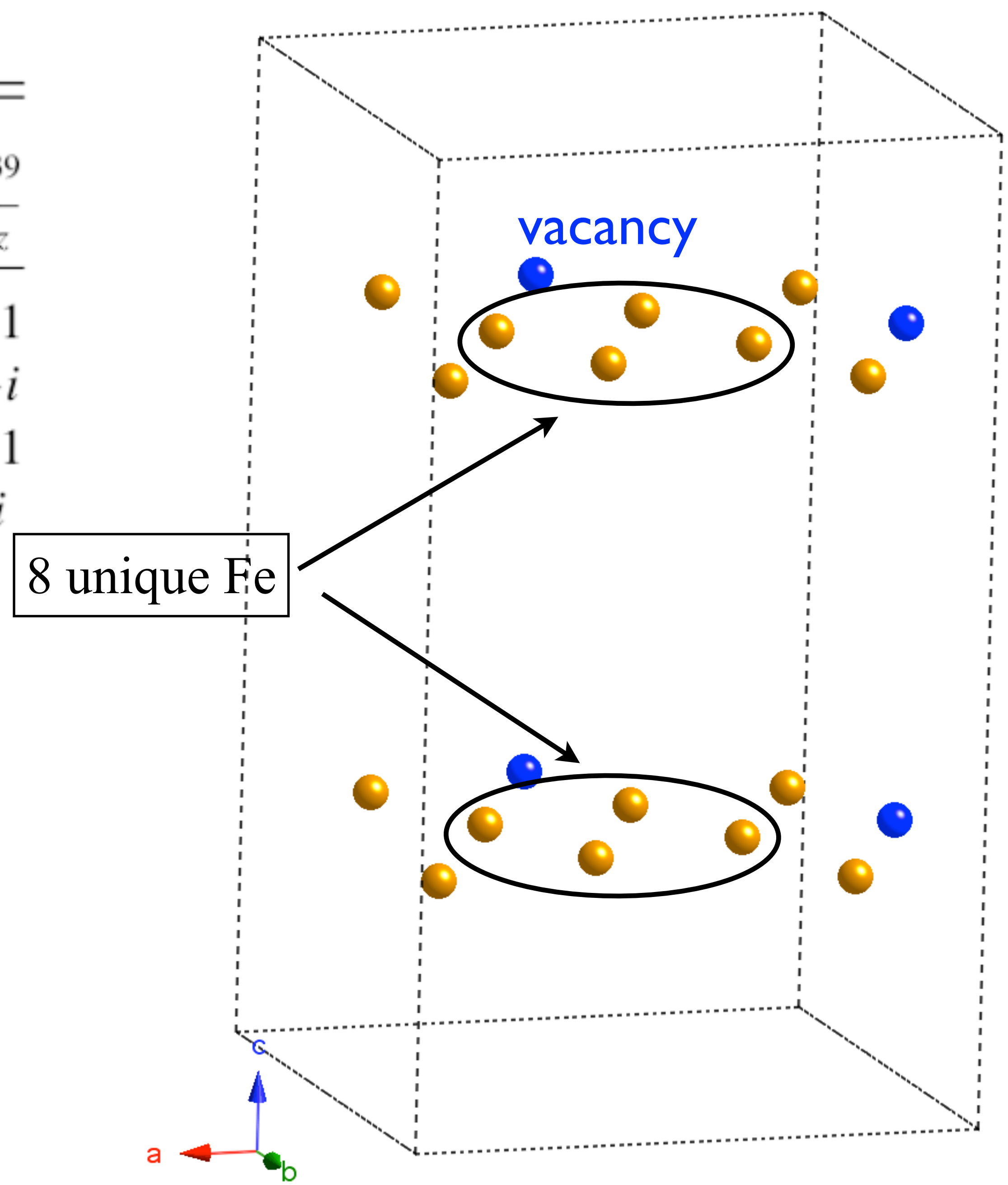
One unit cell with 16 Fe



Phys. Rev. B 83, 144410 (2011)

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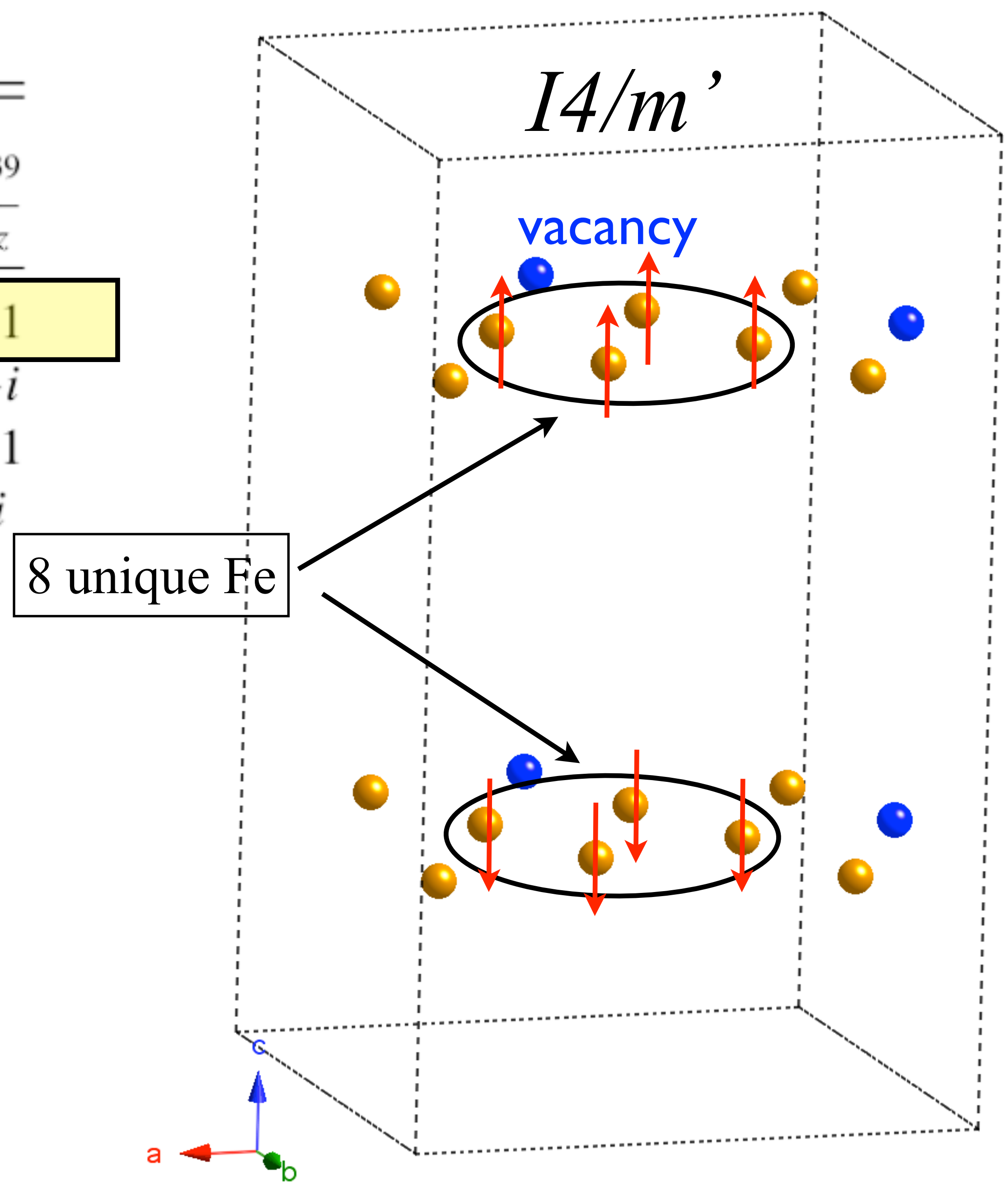

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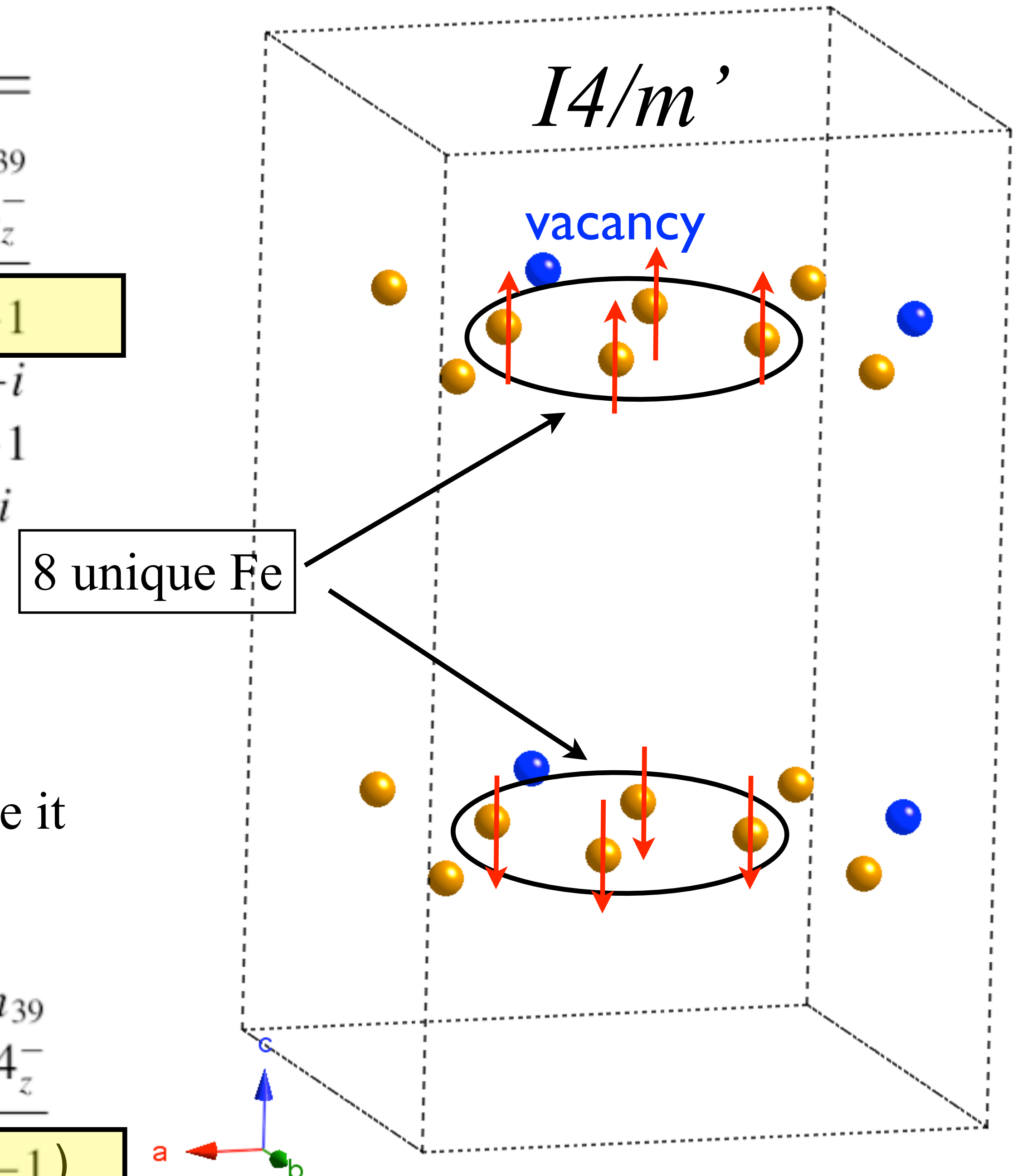

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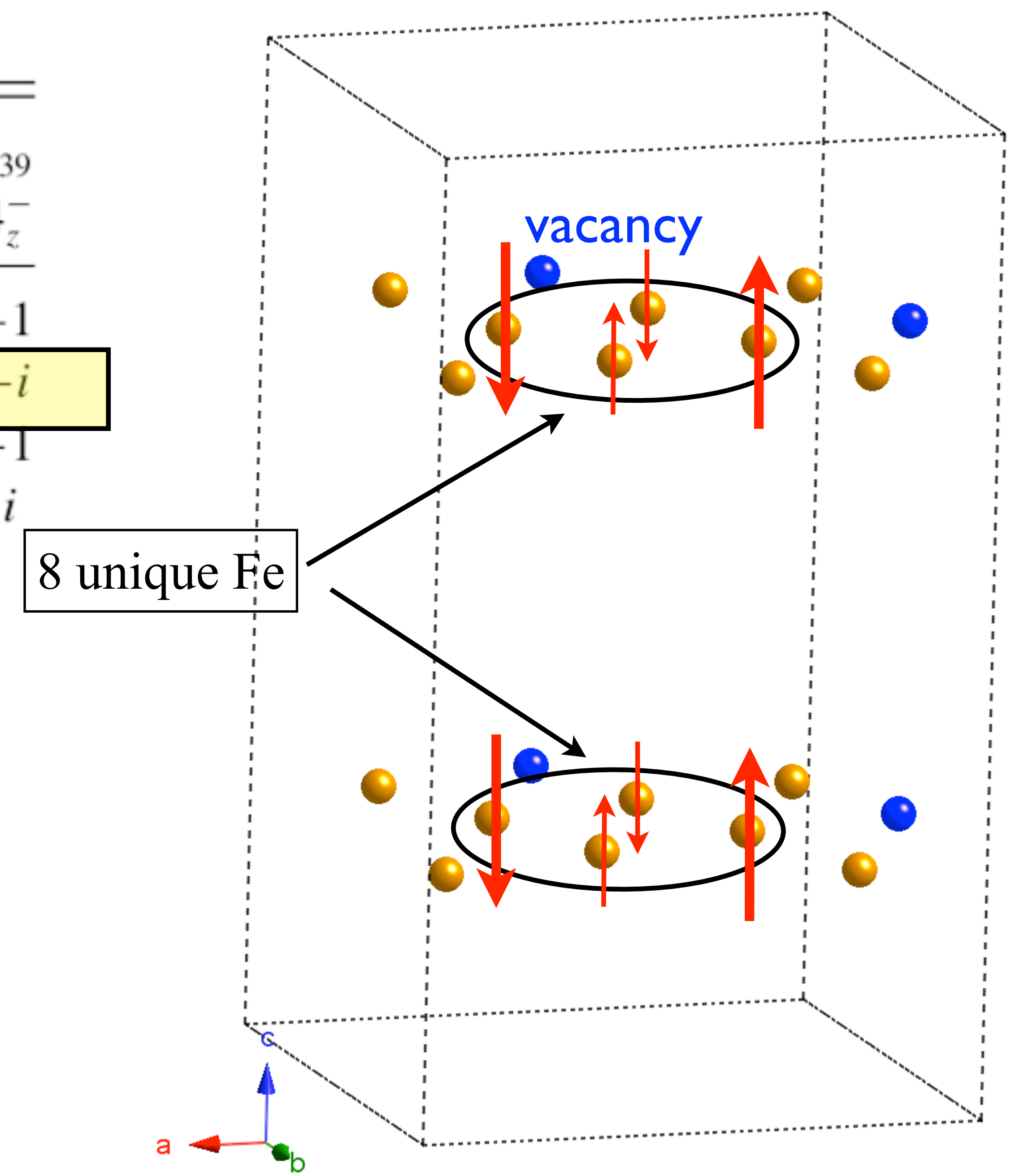

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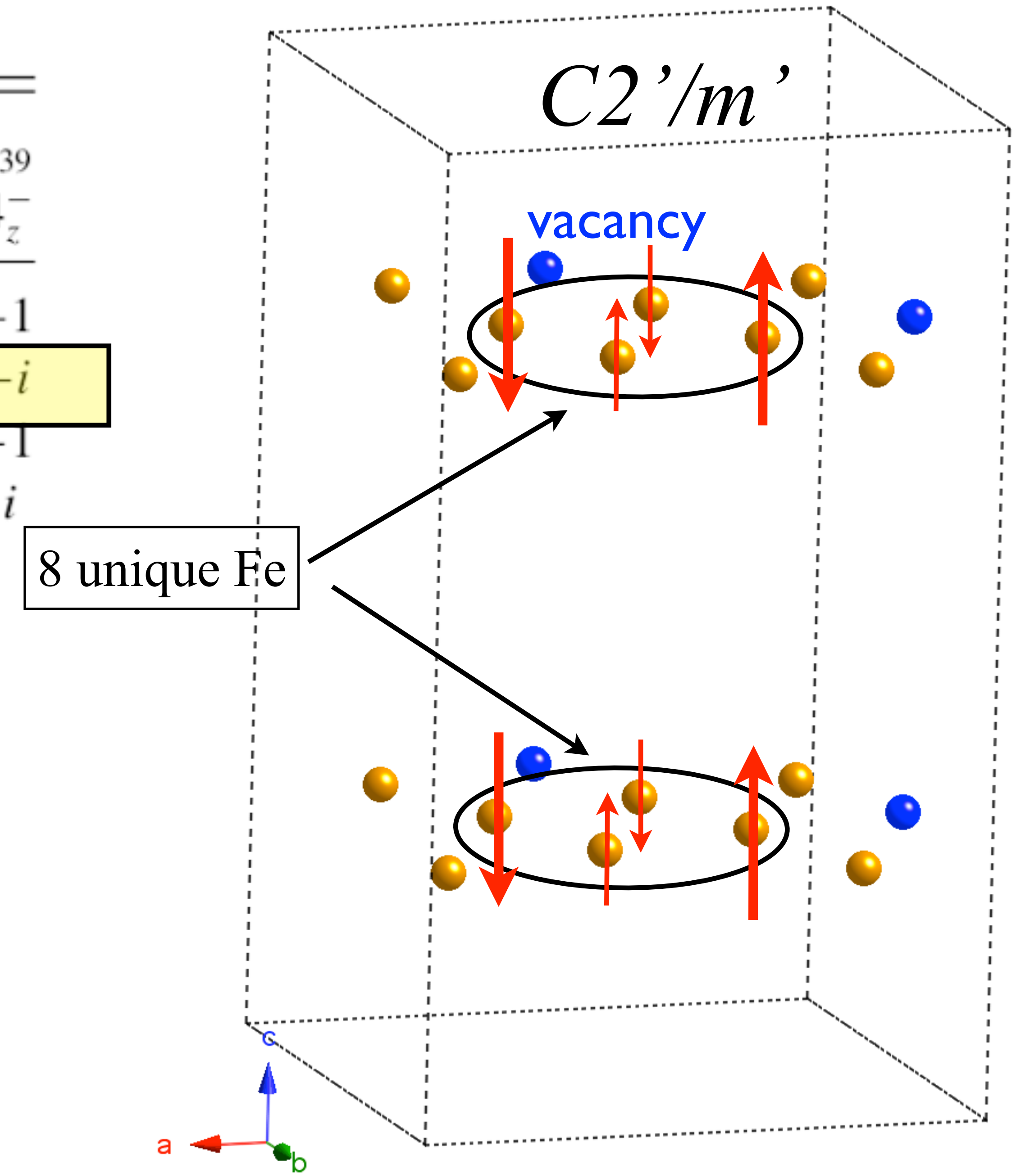

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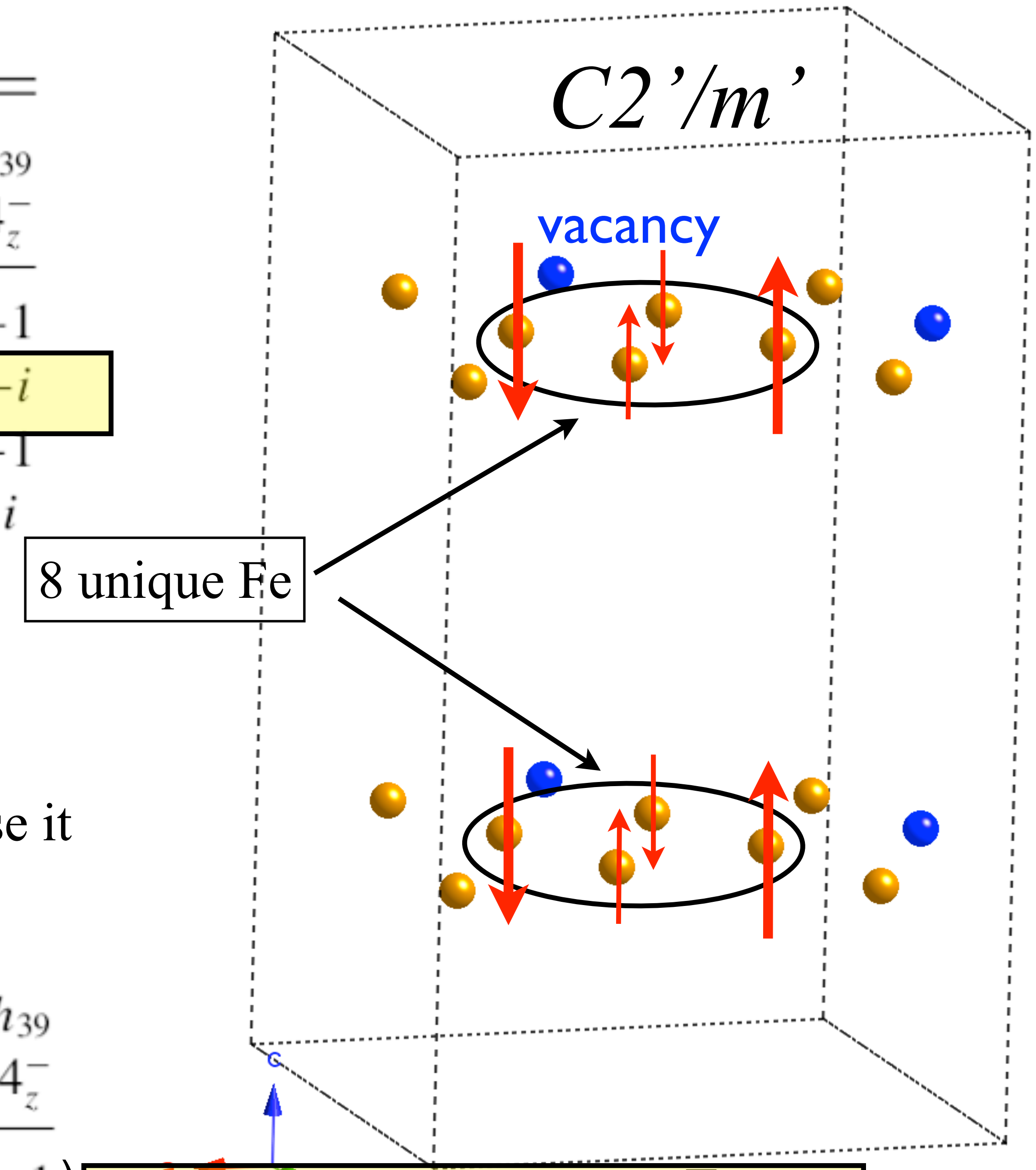

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$$\times|C|\exp(i\varphi)=\frac{1}{2}+i\frac{\sqrt{3}}{2}$$

Magnetic space groups and representation analysis: competing or friendly concepts?

In 1960th-70th opposed

E. F. Bertaut, CNRS, Grenoble
Representation Analysis (RA)*

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Shubnikov magnetic space groups

even until recent times RA was considered to be more powerful in neutron scattering community.*

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Currently > 2010-…

(Representation Analysis) and (Magnetic space groups) are complementary and **must** be used together to fully identify the magnetic symmetry.

---

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"Old new" trends in magnetic structure determination from ND. **Currently there is solid understanding that both RA and Shubnikov magnetic symmetry should be used together.** Big progress in software tools during last years in this way of analysis …

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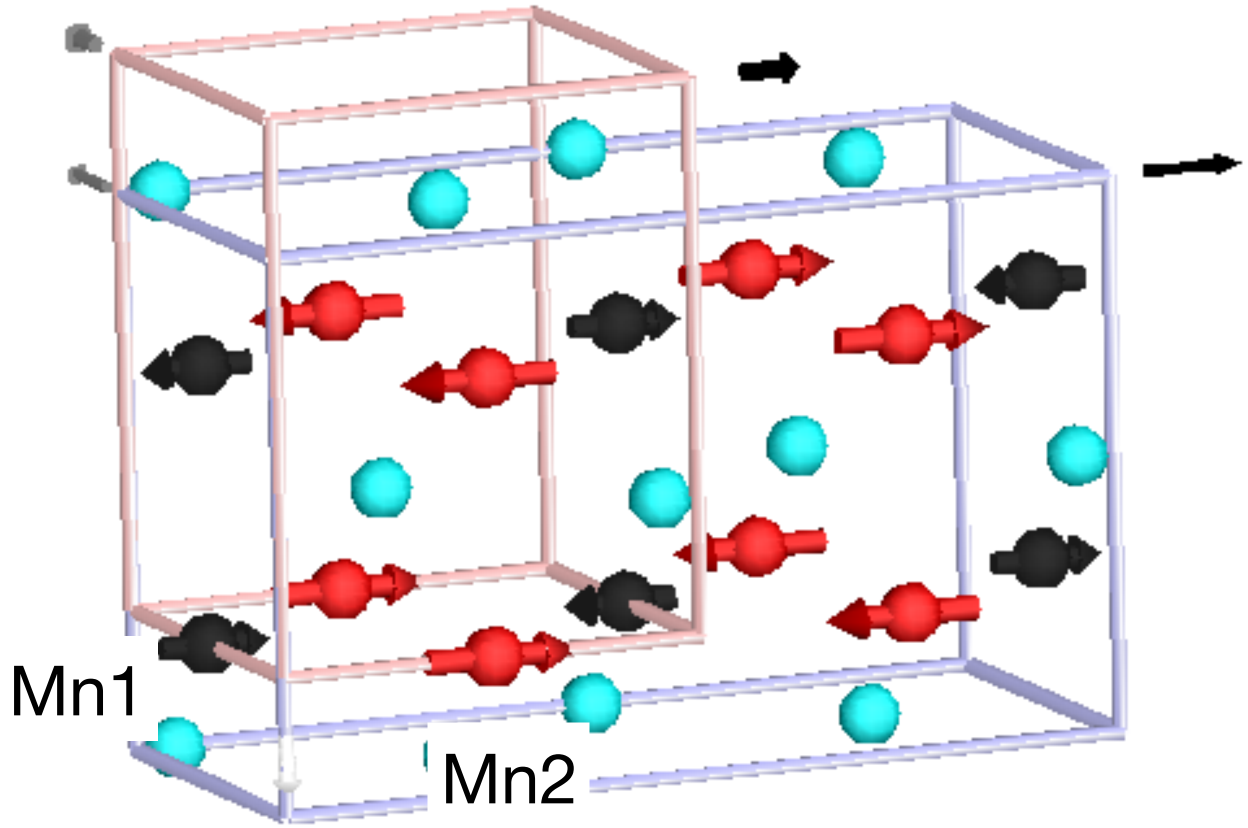
- In many (most) cases this allows one **to find a hidden symmetry**, which is not evident from the representation analysis alone.
- Regular practice for crystal structure transitions: This approach is routinely used by crystallographers in the analysis of crystal phase transition,
- Magnetic transitions: Usually, representation approach with a single arm and general direction of order parameter of propagation vector star. Possible high symmetry Shubnikov subgroups are lost.

Two examples of magnetic structures

multiferroic $TmMnO_3$

one-arm two dimensional irrep **k**=[1/2,0,0].
Ferro-electric phase polar magnetic group P_bmn2_1

Moments along *x*



V. Yu. Pomjakushin, et al New Journal of Physics vol. 11, 043019 (2009)

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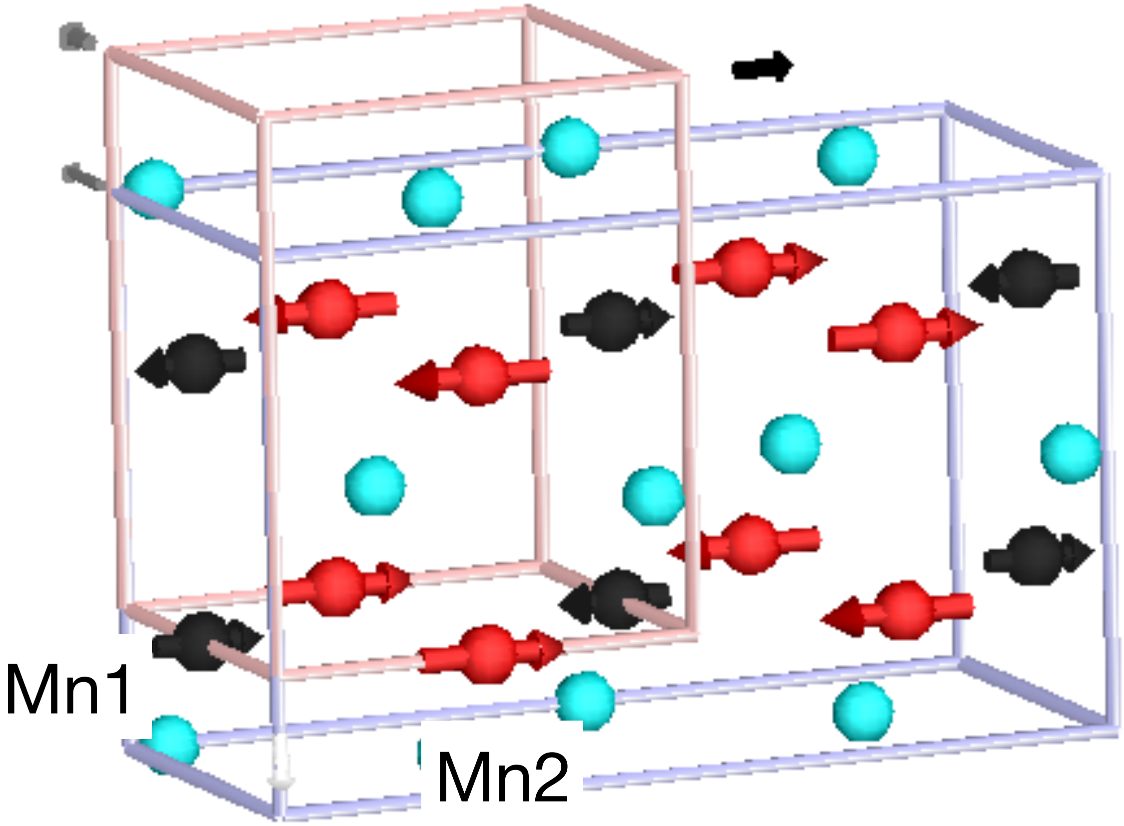
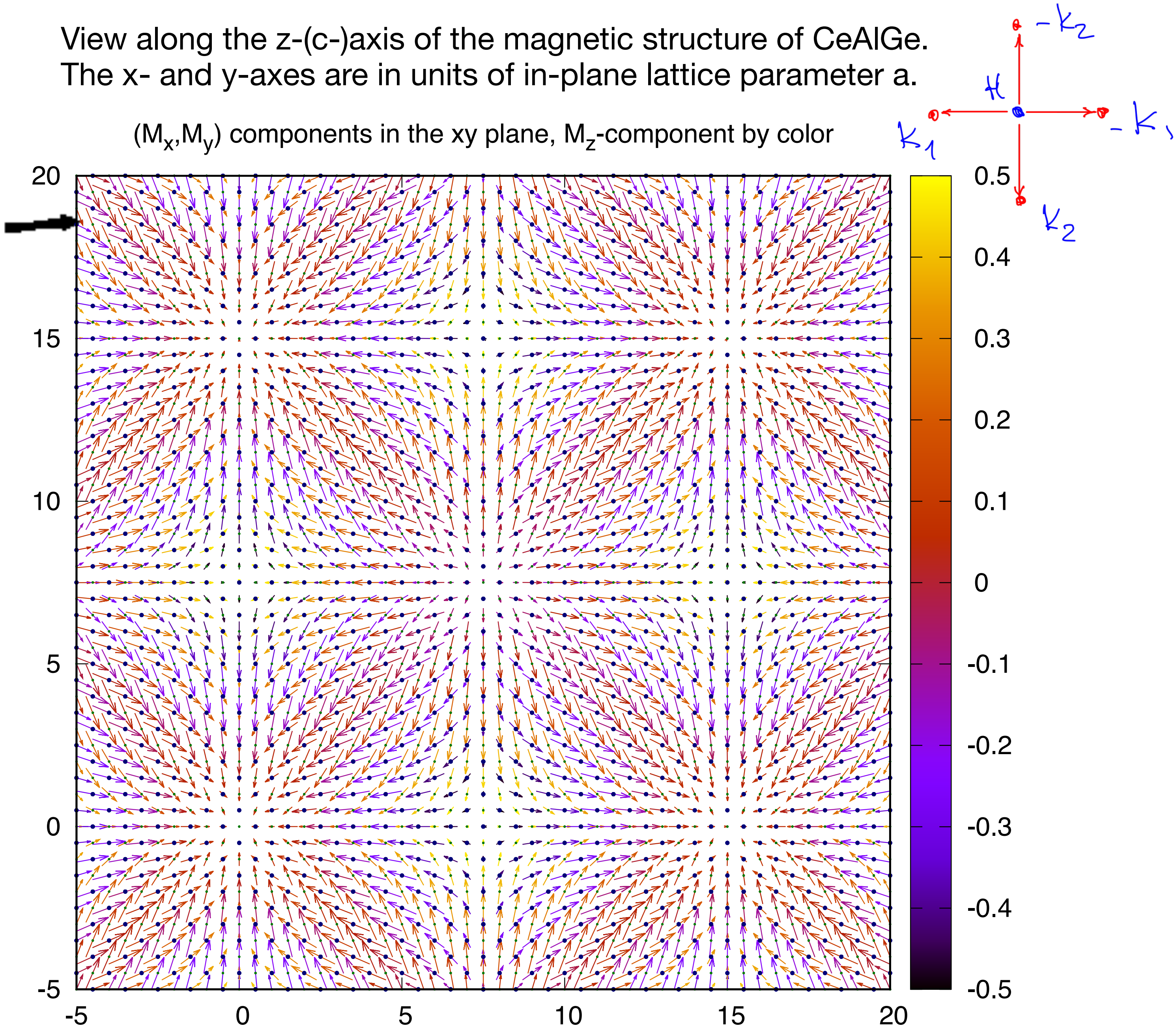


V. Yu. Pomjakushin, et al New Journal of Physics vol. 11, 043019 (2009)

magnetic Weyl semimetal CeAlGe

Topologically nontrivial magnetisation textures in real-space ==> topological Hall effect (THE). Full star superspace 3D+2 group I4_1md1'(a00)000s(0a0)0s0s

View along the z-(c-)axis of the magnetic structure of CeAlGe. The x- and y-axes are in units of in-plane lattice parameter a.



P. Puphal, et al, Physical Review Letters, 124, 017202 (2020)

Antiferromagnetic order in orthorhombic multiferroic $TmMnO_3$

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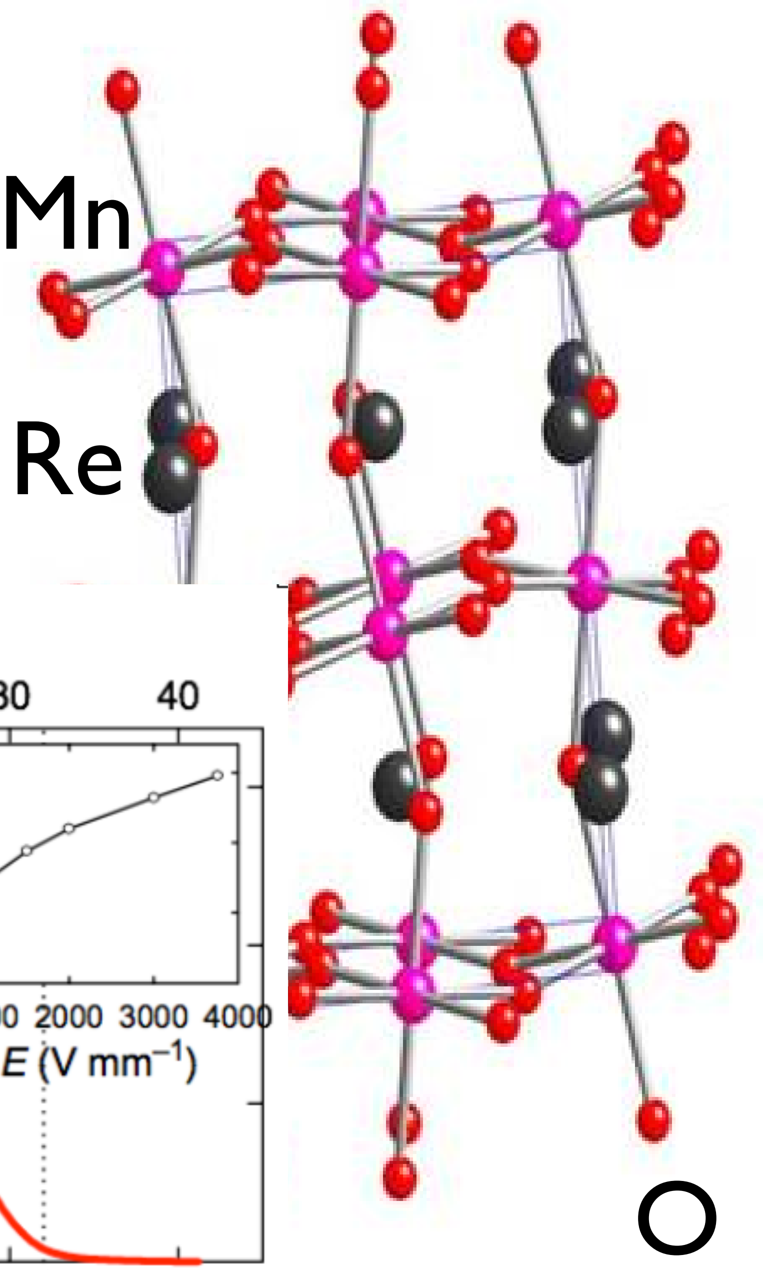
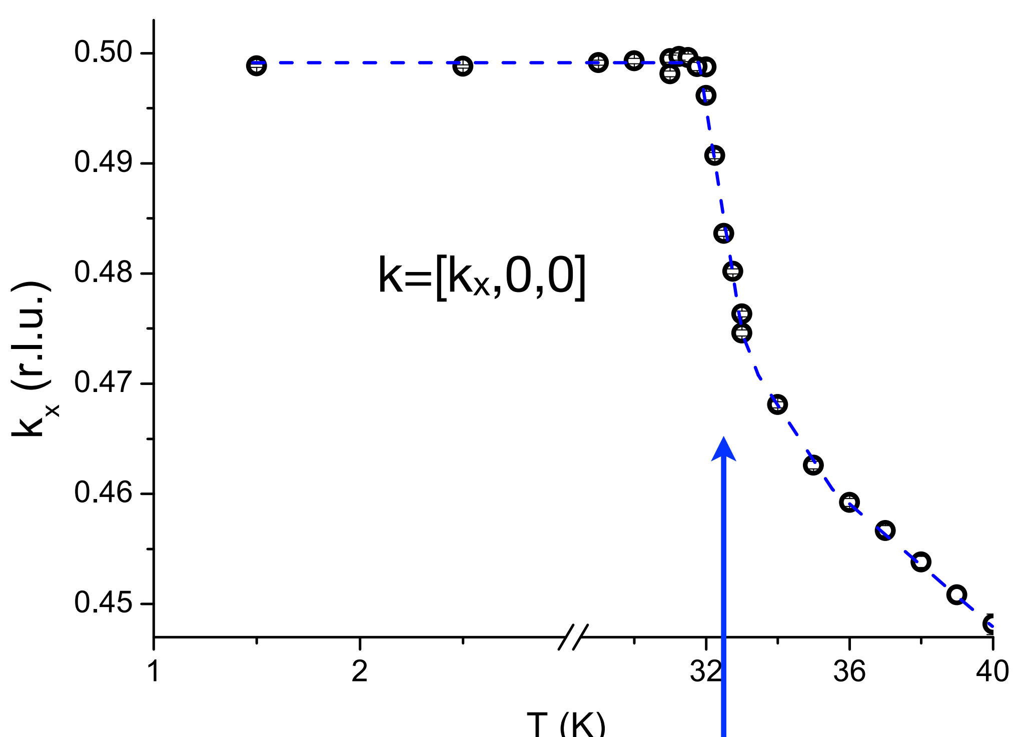
Pnma

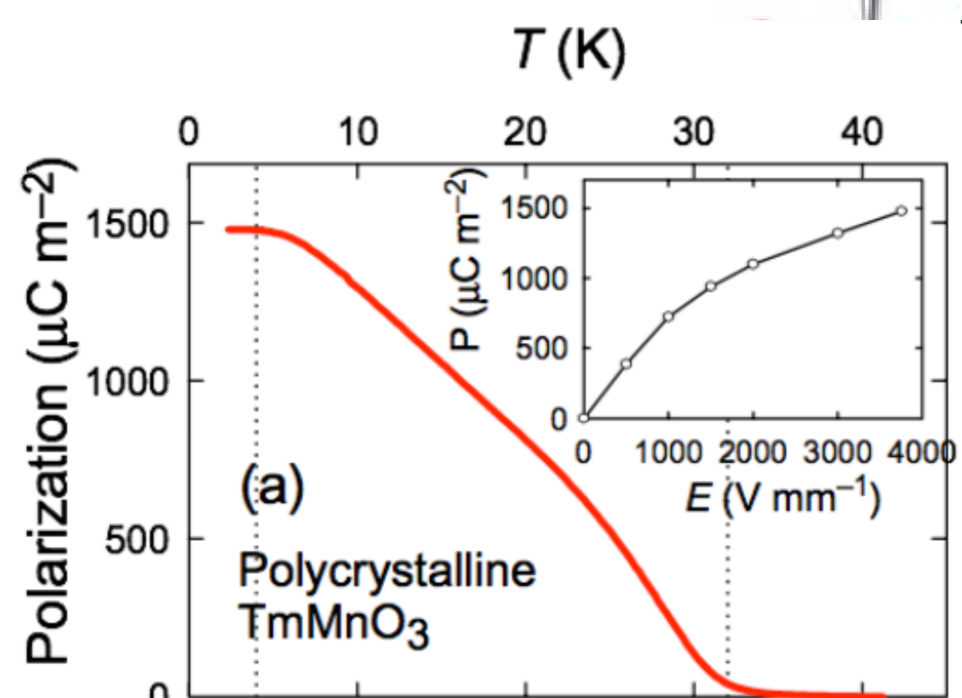
Mn

Re

O







magnetic $T_{Néel}$ & electric: T_C

New Journal of Physics 11, 043019 (2009)

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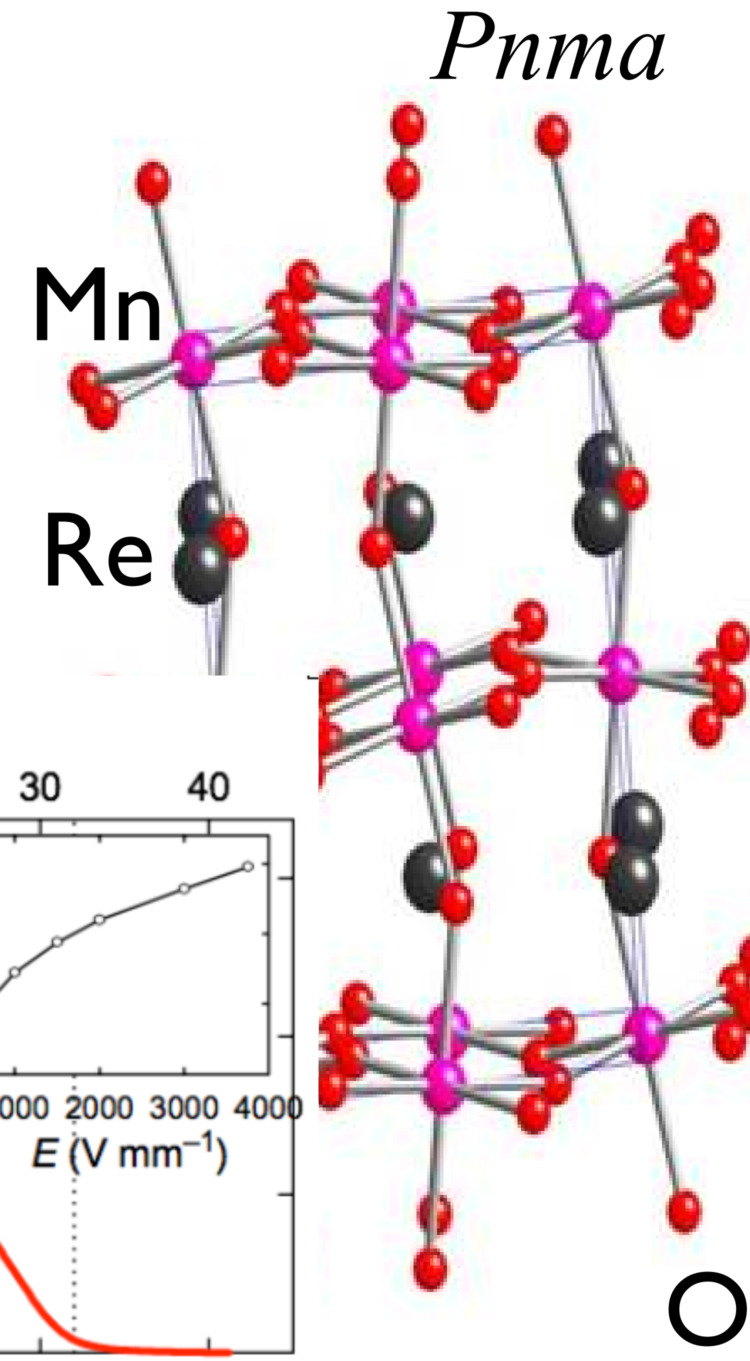
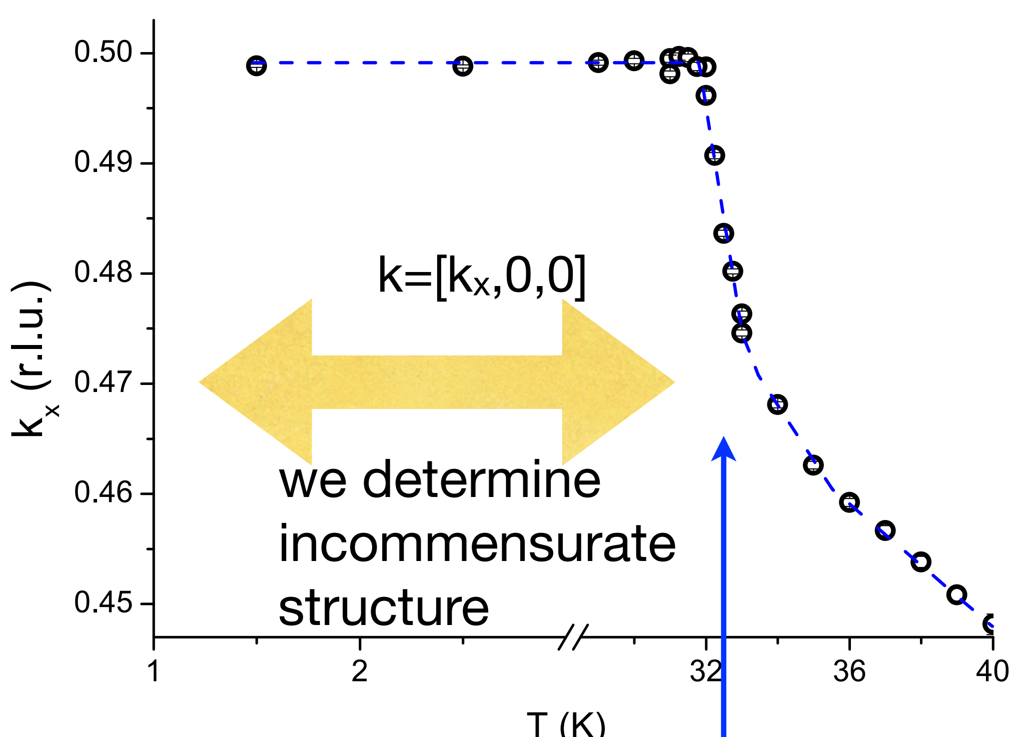
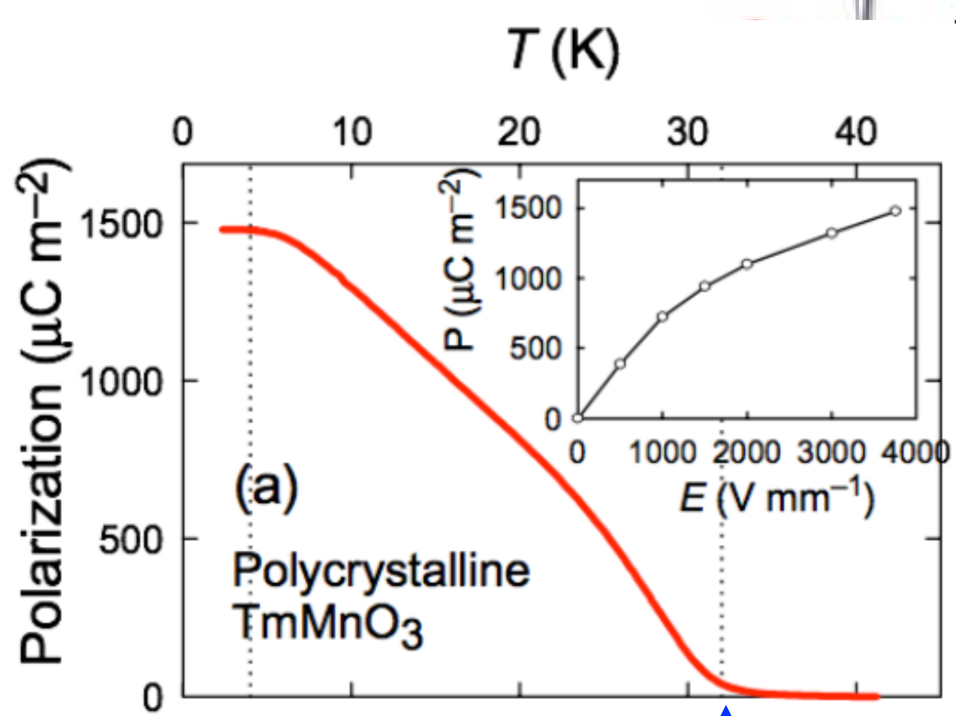






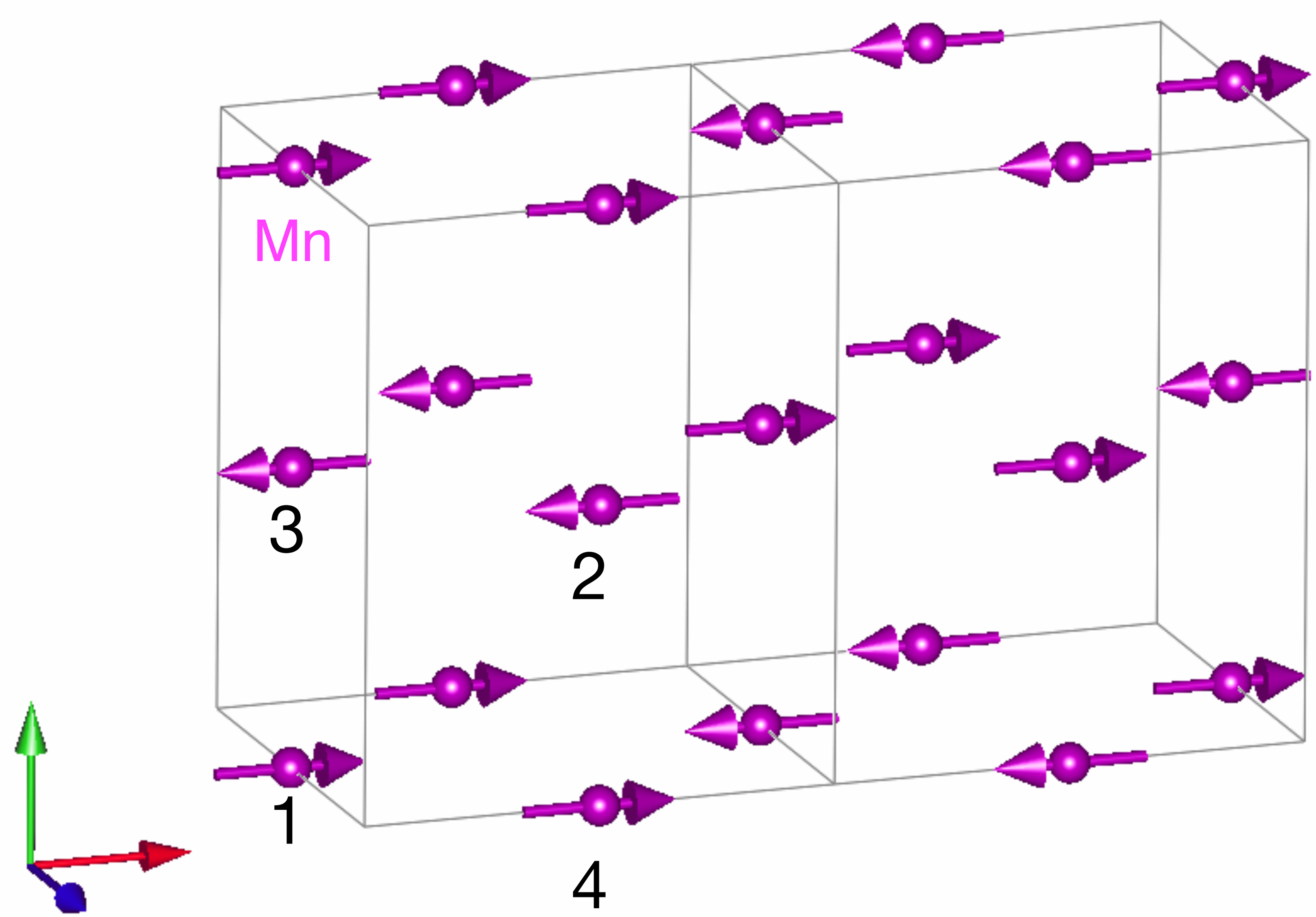
magnetic $T_{Néel}$ & electric: T_C

New Journal of Physics 11, 043019 (2009)

$TmMnO_3$
Two magnetic modes E_1 and E_2 along x.

Mn-position	(1) $0,0,\frac{1}{2}$	(2) $\frac{1}{2},\frac{1}{2},0$	(3) $0,\frac{1}{2},\frac{1}{2}$	(4) $\frac{1}{2},0,0$
$S_0^1 == E_1 =$	+1	+1	-1	-1
$S_0^2 == E_2 =$	+1	-1	-1	+1

Pnma k=[1/2,0,0], k20, *X*
irreps: two **2D** τ_1, τ_2
Mn mΓ: $\underline{3\tau_1} \oplus 3\tau_2$



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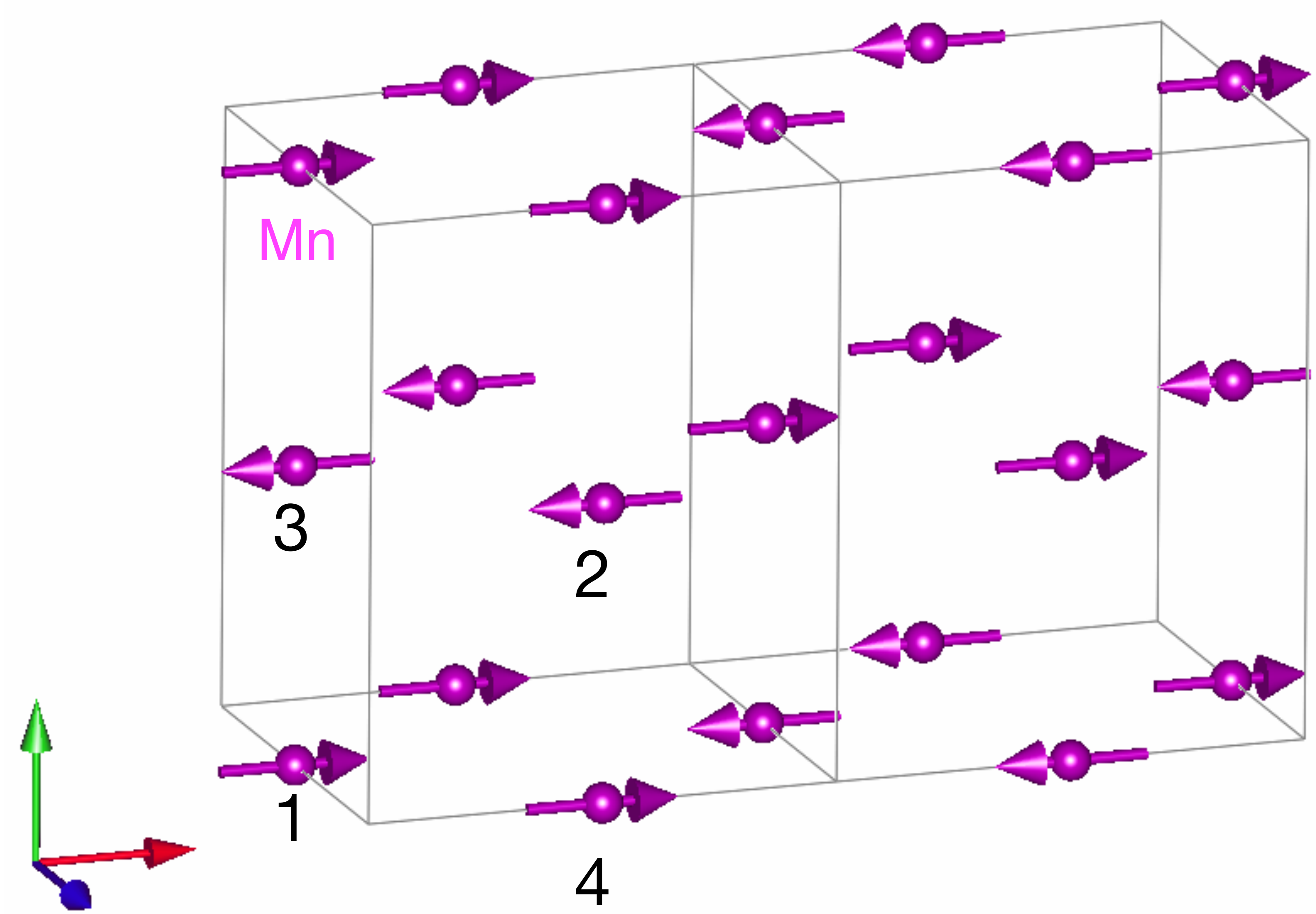
$S_0^1 == E_1 =$	+1	+1	-1	-1
$S_0^2 == E_2 =$	+1	-1	-1	+1

Any linear combination, in general

$C_1E_1+C_2E_2 =$	C_1+C_2	C_1-C_2	$-C_1-C_2$	C_1+C_2
$(E_1+E_2)/2 =$	+1	0	-1	0
$(E_1-E_2)/2 =$	0	+1	0	-1

Pnma k=[1/2,0,0], k20, *X*
irreps: two **2D** τ_1, τ_2
Mn mΓ: $\underline{3\tau_1} \oplus 3\tau_2$



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Any linear combination, in general

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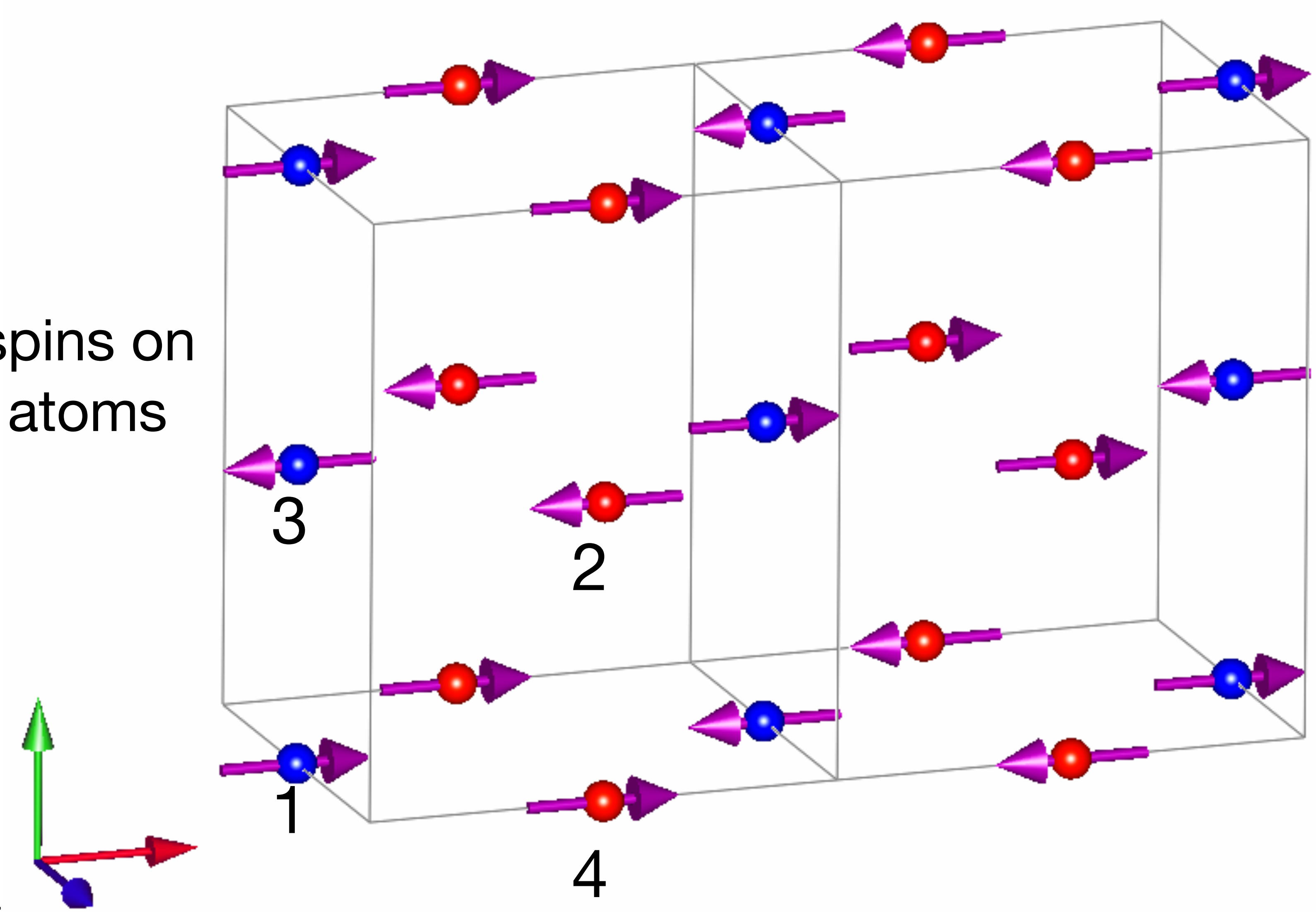
Pnma k=[1/2,0,0], k20, *X*
irreps: two **2D** τ_1, τ_2
Mn mΓ: $\underline{3\tau_1} \oplus 3\tau_2$

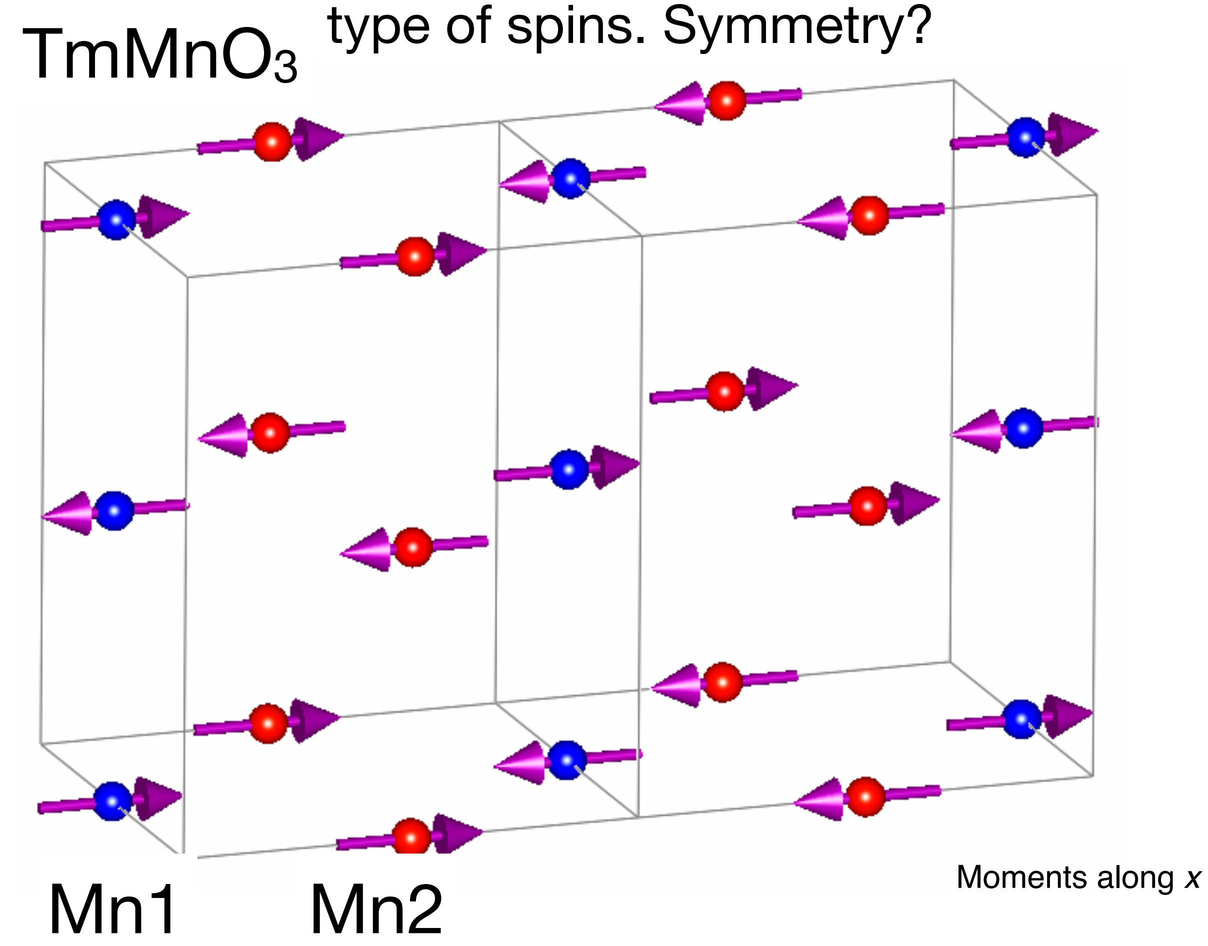
independent spins on
red and blue atoms



Symmetry analysis using both RA and magnetic subgroups

Pnma k=[1/2,0,0], *irrep*: **2D** mX1(τ_1)

RA with arbitrary mixing coefficients gives different spin sizes for the same type of spins. Symmetry?



Symmetry analysis using both RA and magnetic subgroups

Pnma k=[1/2,0,0], *irrep*: **2D** mX1(τ_1)

http://stokes.byu.edu/iso/

ISOTROPY Software Suite

Harold T. Stokes, Dorian M. Hatch, and Branton J. Campbell, Department of Physics and Astronomy, Brigham Young University, Provo, Utah 84606, USA,

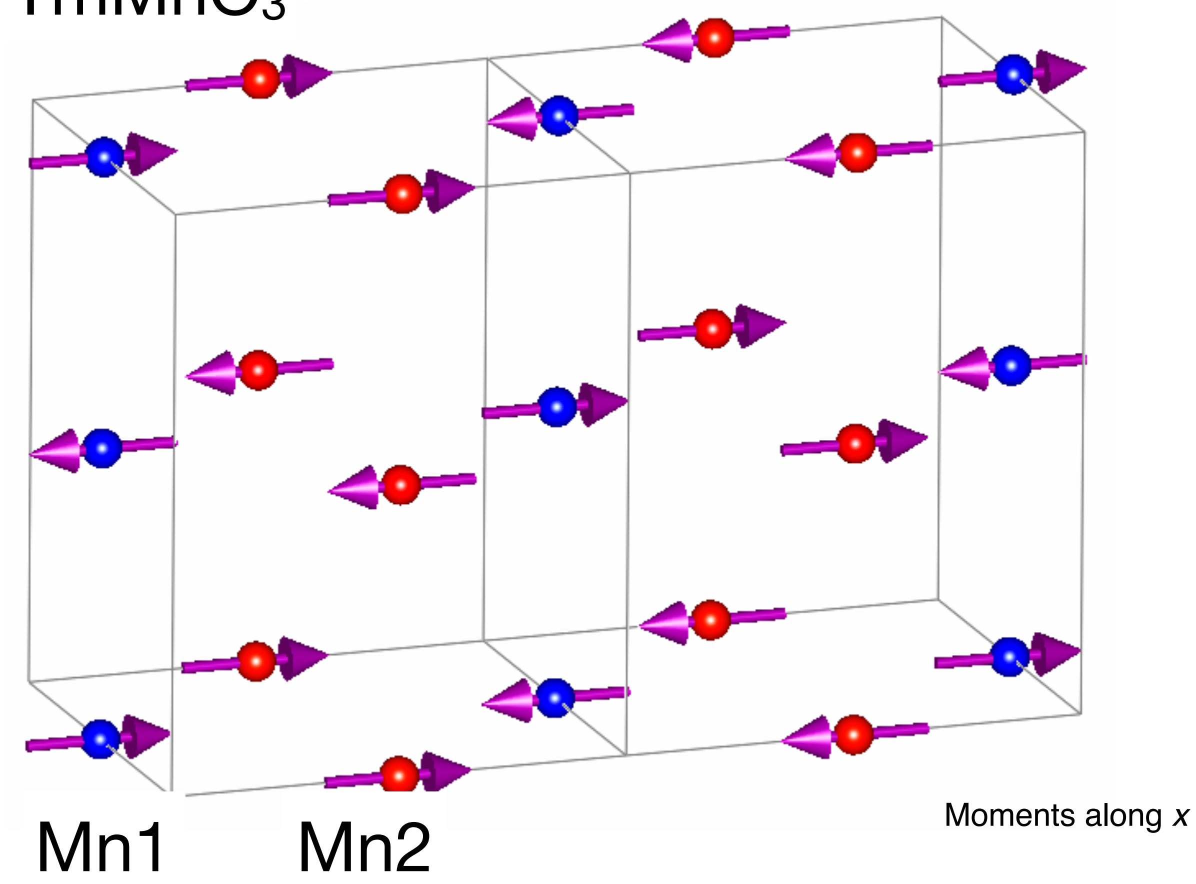
ISODISTORT

Version 6.1.8, November 2014

Harold T. Stokes. Branton J. Campbell, and Dorian M. Hatch,

RA with arbitrary mixing coefficients gives different spin sizes for the same type of spins. Symmetry?

$TmMnO_3$



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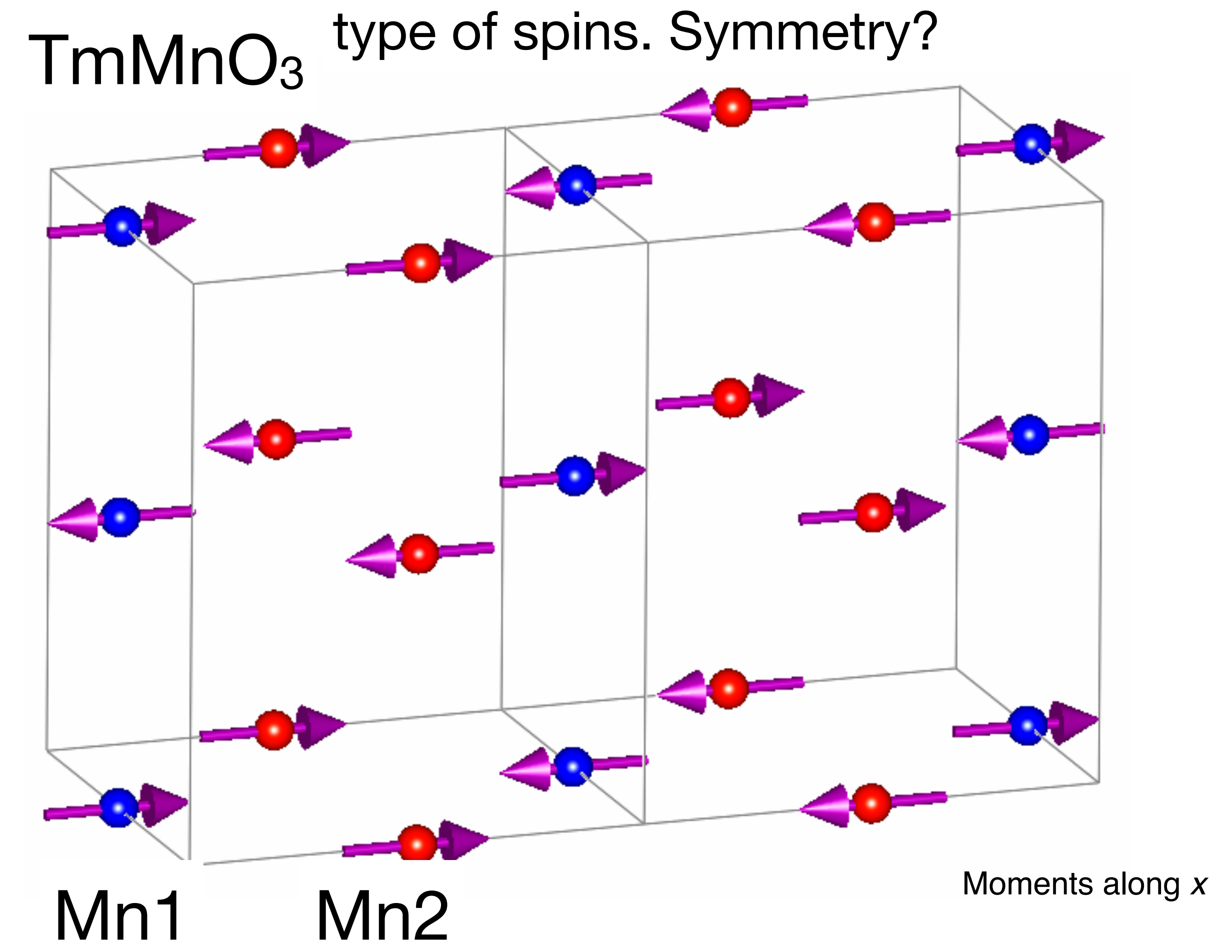
P1 (a,0) 11.55 P_a2_1/m, basis={(2,0,0),(0,1,0),(0,0,1)}, origin=(1/2,0,0), s=2, i=4, k-active= (1/2,0,0)
P3 (a,a) 31.129 P_bmn2_1, basis={(0,1,0),(2,0,0),(0,0,-1)}, origin=(3/4,1/4,0), s=2, i=4, k-active= (1/2,0,0)
C1 (a,b) 6.21 P_am, basis={(2,0,0),(0,1,0),(0,0,1)}, origin=(0,1/4,0), s=2, i=8, k-active= (1/2,0,0)

Order parameter direction

Magnetic Shubnikov Space group

RA with arbitrary mixing coefficients gives different spin sizes for the same type of spins. Symmetry?



Symmetry analysis using both RA and magnetic subgroups

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P3 (a,a)	31.129	P_bmn2_1,	basis={(0,1,0),(2,0,0),(0,0,-1)}, origin=(3/4,1/4,0), s=2, i=4, k-active= (1/2,0,0)
C1 (a,b)	6.21	P_am,	basis={(2,0,0),(0,1,0),(0,0,1)}, origin=(0,1/4,0), s=2, i=8, k-active= (1/2,0,0)

Order parameter direction

Magnetic Shubnikov Space group

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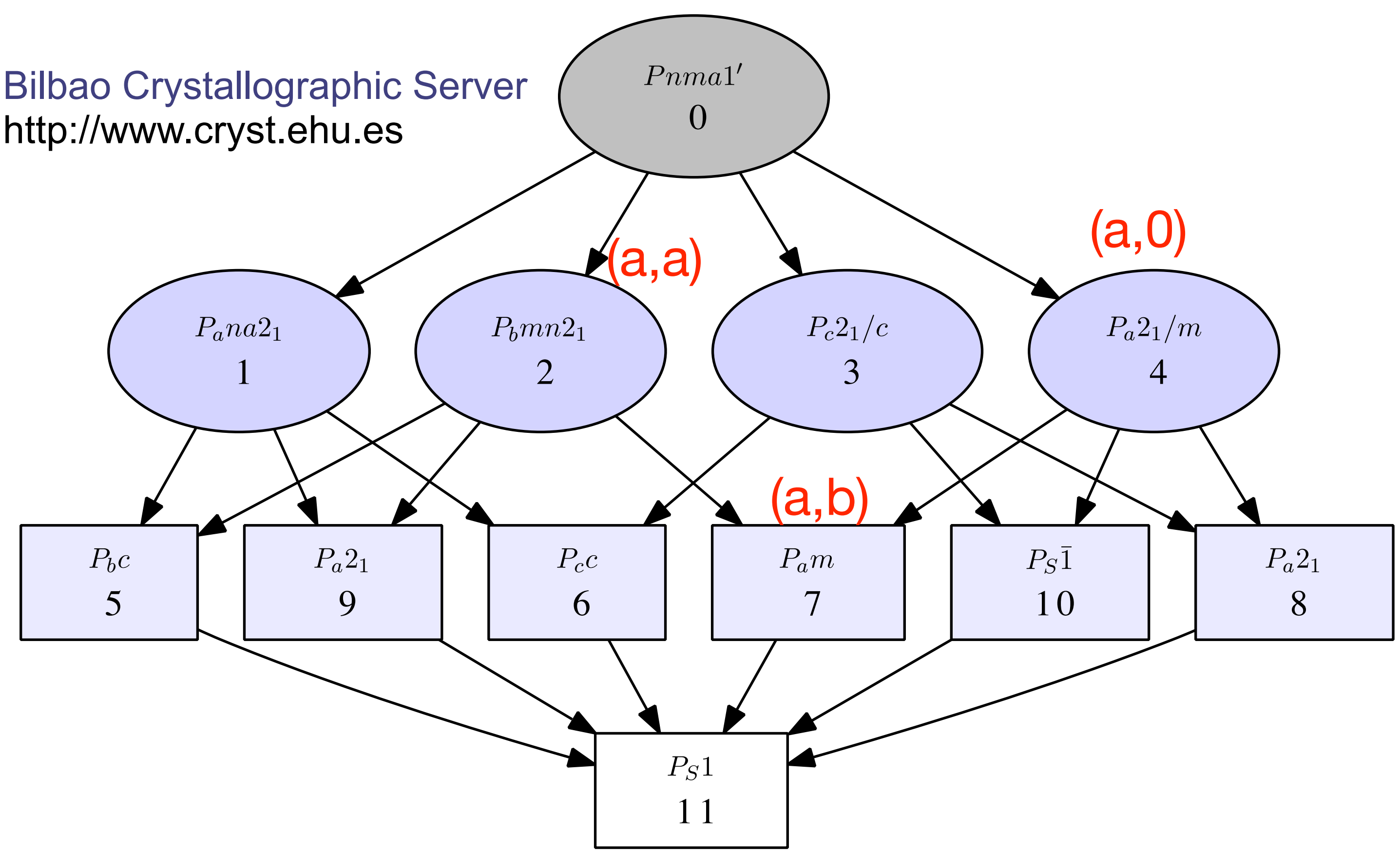
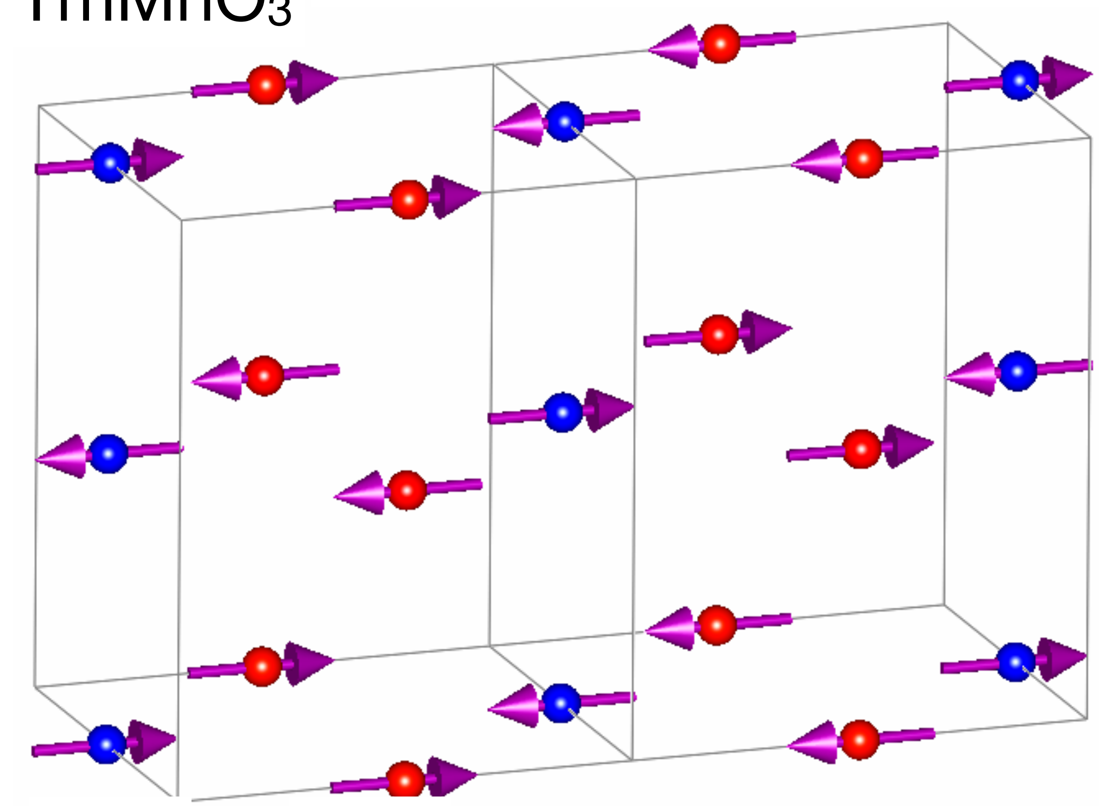
Bilbao Crystallographic Server
http://www.cryst.ehu.es



$TmMnO_3$



Mn1 Mn2

Moments along *x*

Case 1: magnetic mode E1 -> most symmetric maximal subgroup of Pnma1'

Order parameter direction

Magnetic Shubnikov Space group

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ISODISTORT

Version 6.1.8, November 2014

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Solution!

P1 (a,0) 11.55 P_a2_1/m, basis={(2,0,0),(0,1,0),(0,0,1)}, origin=(1/2,0,0), s=2, i=4, k-active= (1/2,0,0)

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Bilbao Crystallographic Server

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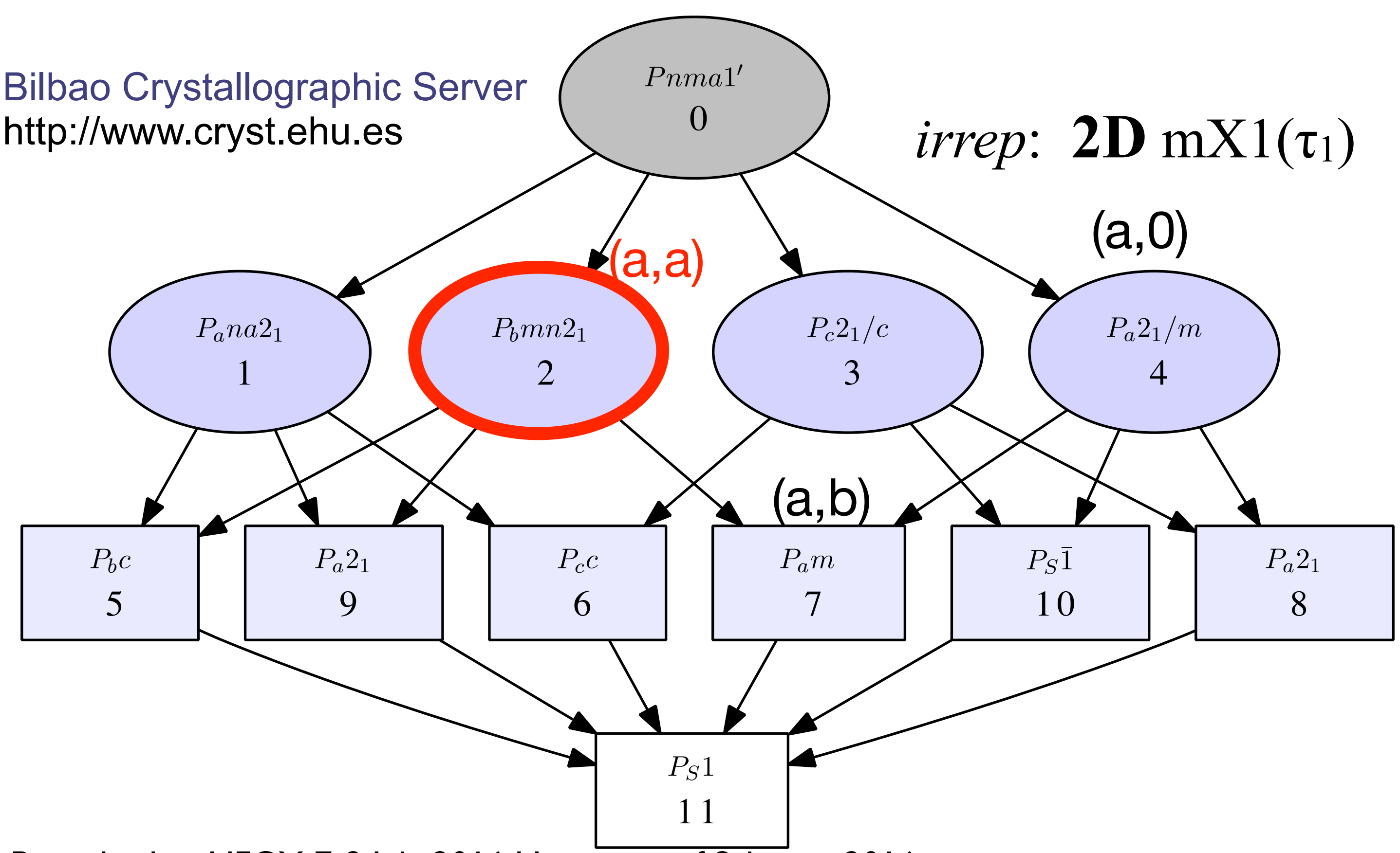
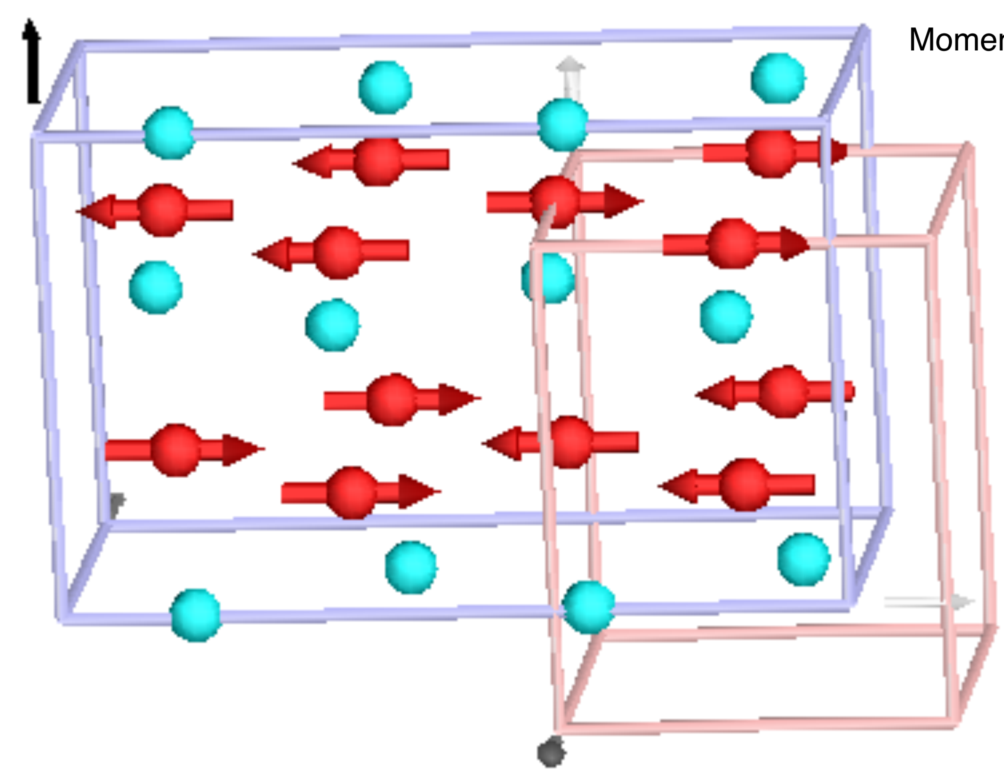


TmMnO$_3$



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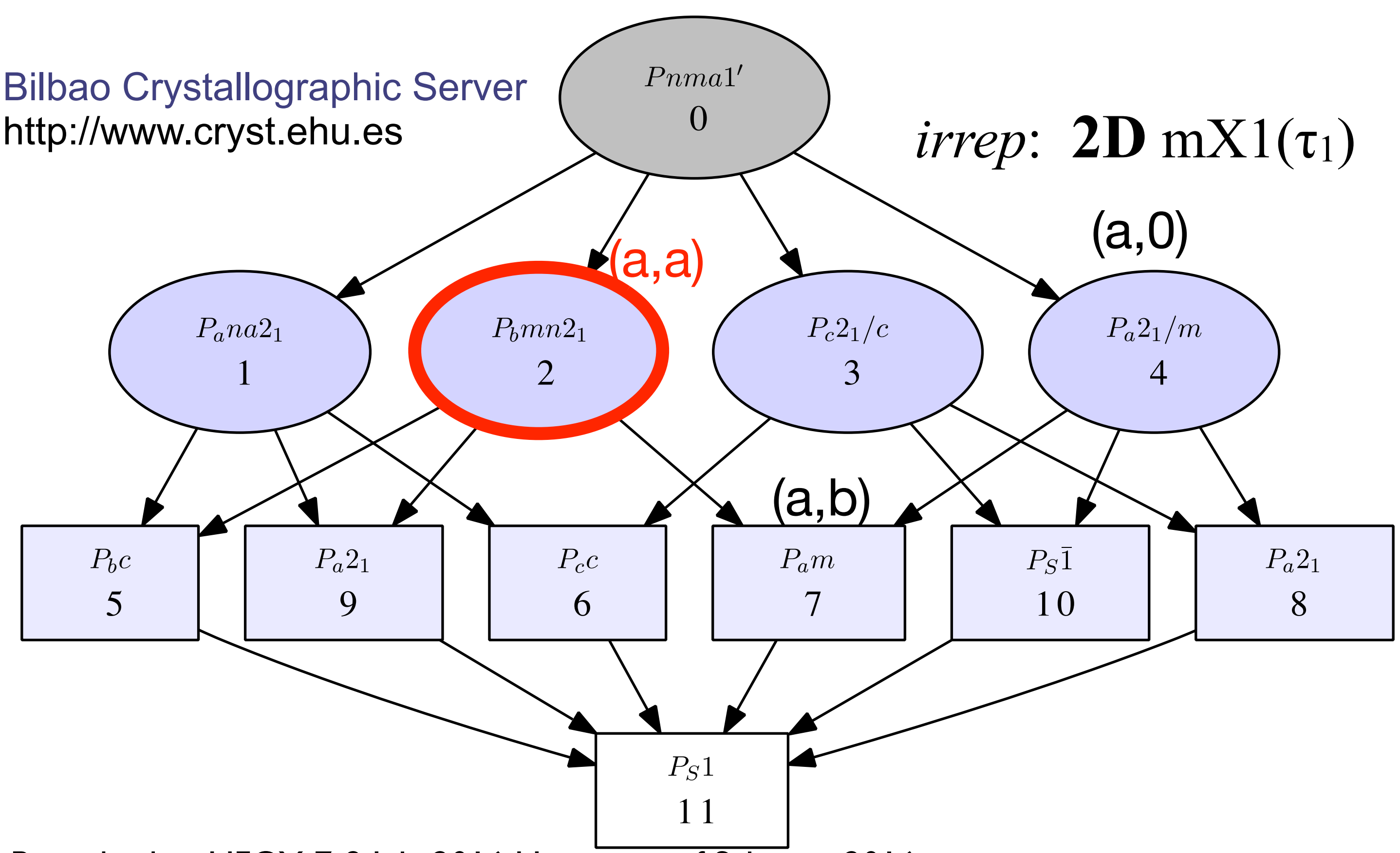
orthorhombic *Pmn2₁*

(1) 1 (2) $2(0,0,\frac{1}{2})$ $\frac{1}{4},0,z$ (3) $2(0,\frac{1}{2},0)$ $0,y,0$ (4) $2(\frac{1}{2},0,0)$ $x,\frac{1}{4},\frac{1}{4}$

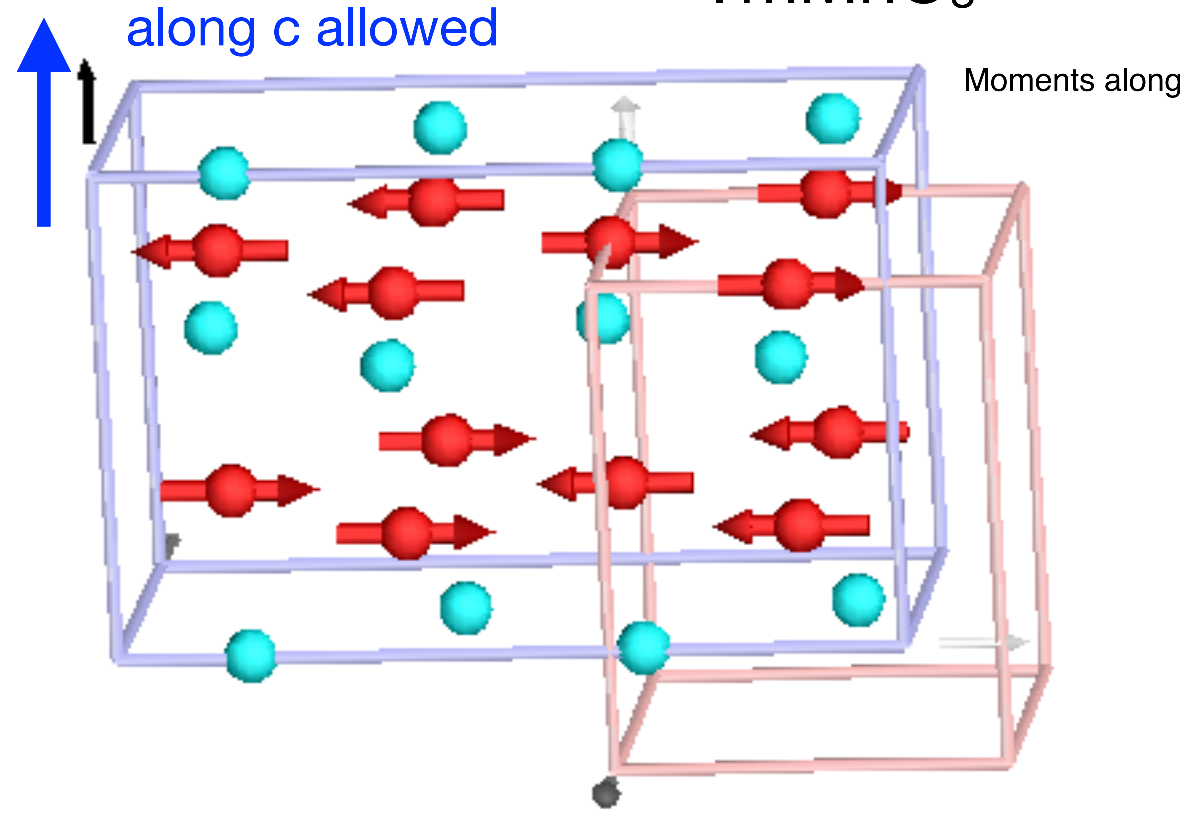
(5) $\bar{1}$ $0,0,0$ (6) a $x,y,\frac{1}{4}$ (7) m $x,\frac{1}{4},z$ (8) $n(0,\frac{1}{2},\frac{1}{2})$ $\frac{1}{4},y,z$

Bilbao Crystallographic Server
http://www.cryst.ehu.es



Electric polarisation along c allowed

$TmMnO_3$



Case 2: General solution in RA -> low symmetry non-maximal subgroup

Order parameter direction

Magnetic Shubnikov Space group

Harold T. Stokes, Dorian M. Hatch, and Branton J. Campbell, Department of Physics and Astronomy, Brigham Young University, Provo, Utah 84606, USA,

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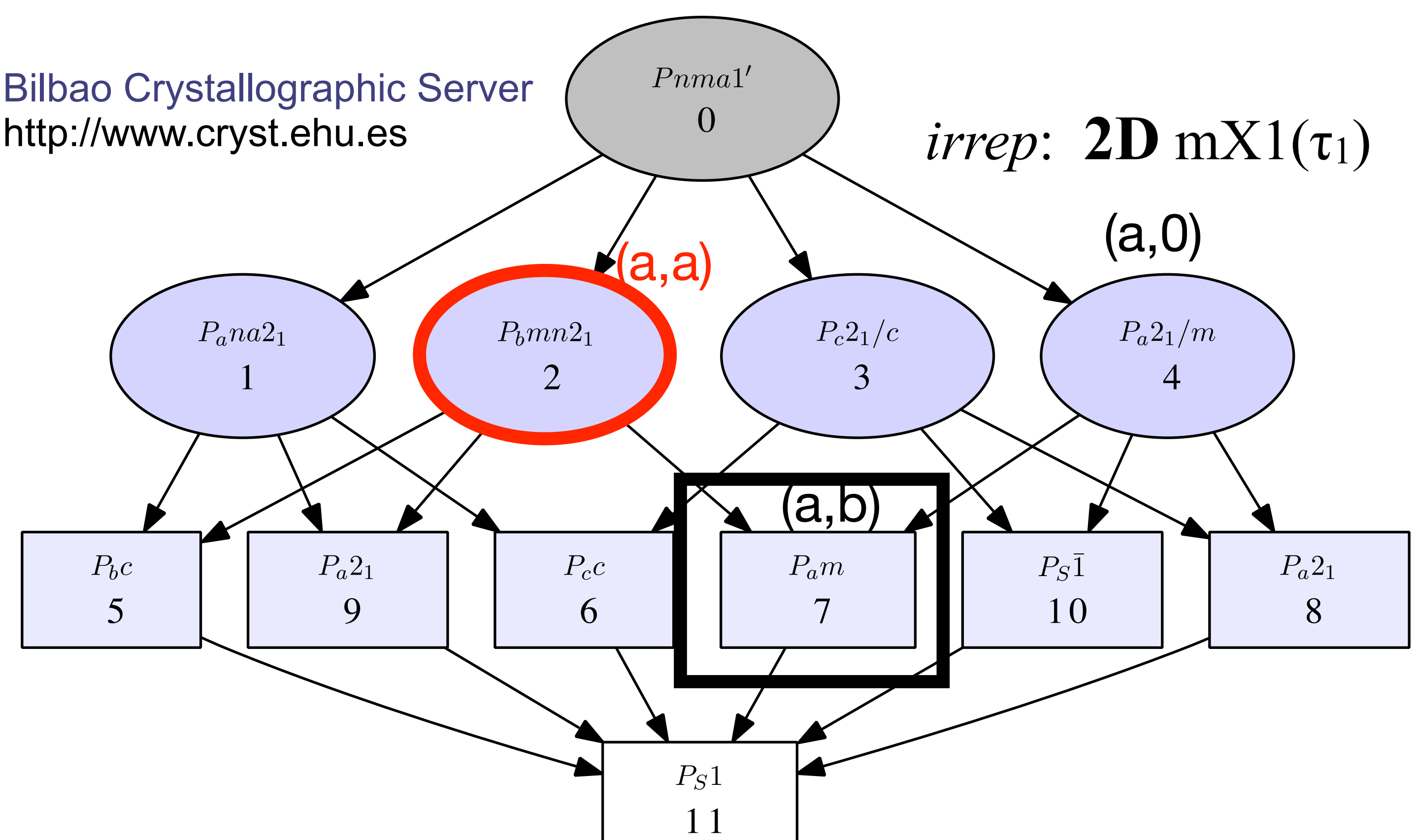
C1 (a,b) 6.21 P_am, basis={(2,0,0),(0,1,0),(0,0,1)}, origin=(0,1/4,0), s=2, i=8, k-active= (1/2,0,0)

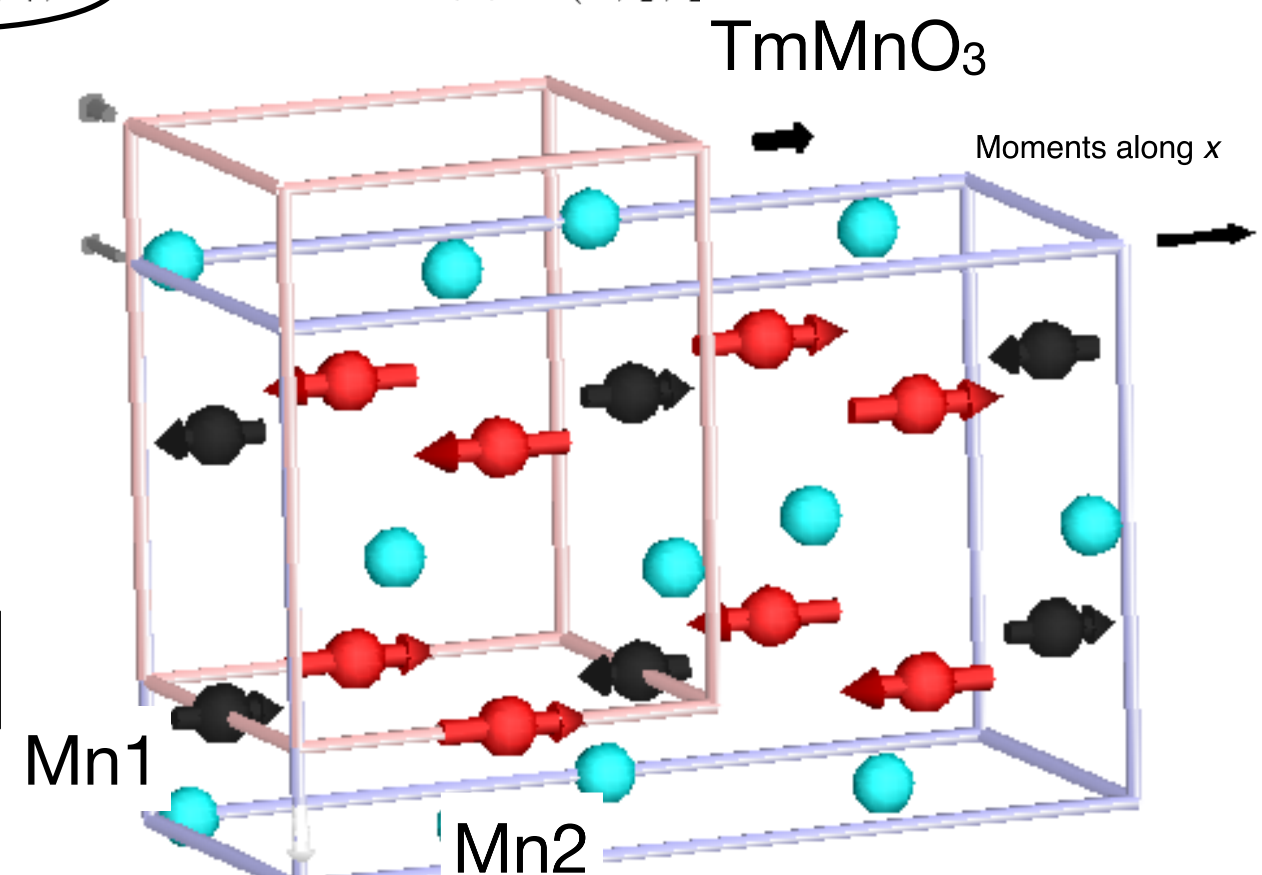
conventional general solution in RA: lowest symmetry for the given irrep

monoclinic Pm

(1) 1	(2) $2(0,0,\frac{1}{2})$ $\frac{1}{4},0,z$	(3) $2(0,\frac{1}{2},0)$ $0,y,0$	(4) $2(\frac{1}{2},0,0)$ $x,\frac{1}{4},\frac{1}{4}$
(5) $\bar{1}$ $0,0,0$	(6) a $x,y,\frac{1}{4}$	(7) m $x,\frac{1}{4},z$	(8) $n(0,\frac{1}{2},\frac{1}{2})$ $\frac{1}{4},y,z$

Bilbao Crystallographic Server
http://www.cryst.ehu.es





Superspace magnetic structure in Weyl semimetal CeAlGe. Multi arm antiferromagnetic order. Ref: P. Puphal et al, accepted PRL (2019) arxiv/nnn

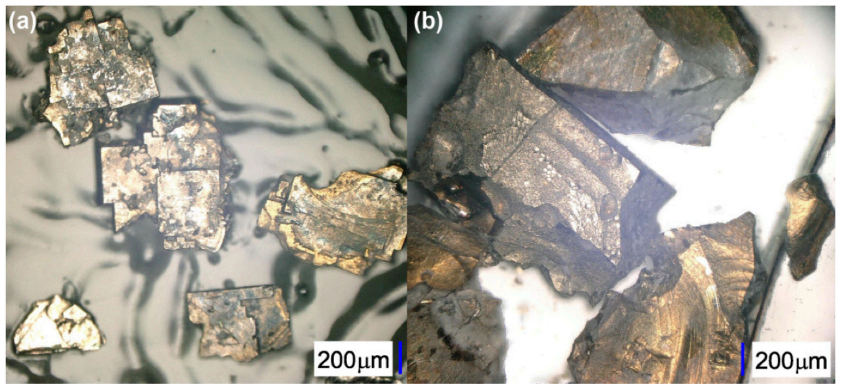


FIG. 2. Pictures of the flux-grown crystals of (a) CeAlGe and (b) PrAlGe right after flux removal using NaOH-H_2O, and before subsequent annealing

FIG. 3. Photos of (a) the cast CeAlGe rod, and the floating-zone-grown crystals of (b) CeAlGe and (c) PrAlGe.

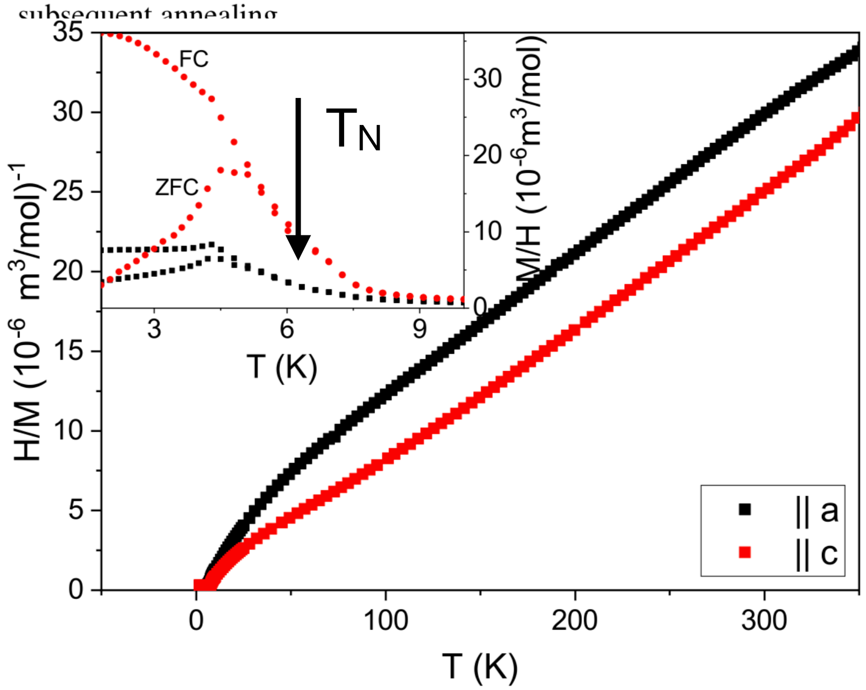
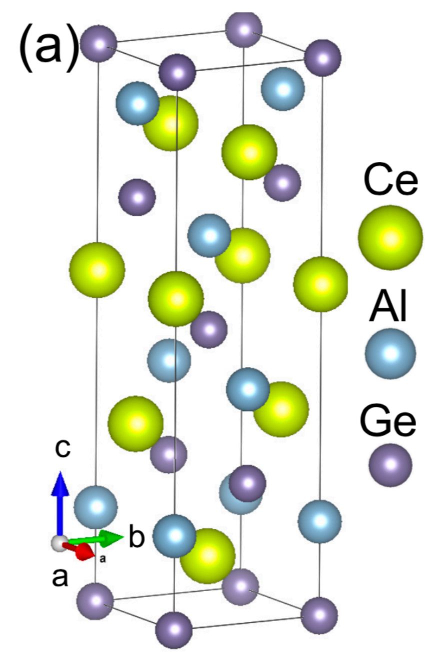


FIG. 8. Magnetic data obtained on a floating-zone-grown CeAlGe single crystal with a mass of 125.4 mg. The magnetic



Space Group: 109 I4_1md C4v-11

non-centrosymmetric

Lattice parameters:

a=4.25717, c=14.64520

<u>Ce1 4a (0,0,z), z=-0.41000 single magnetic Ce site</u>

Neutron diffraction experiments: HRPT and DMC, SANS at PSI Switzerland, D33, at ILL France

Resistivity: Topologicall Hall Effect in University of Tokyo

Samples: both powder and single crystals of CeAlGe grown at PSI in Solid State Chemistry group

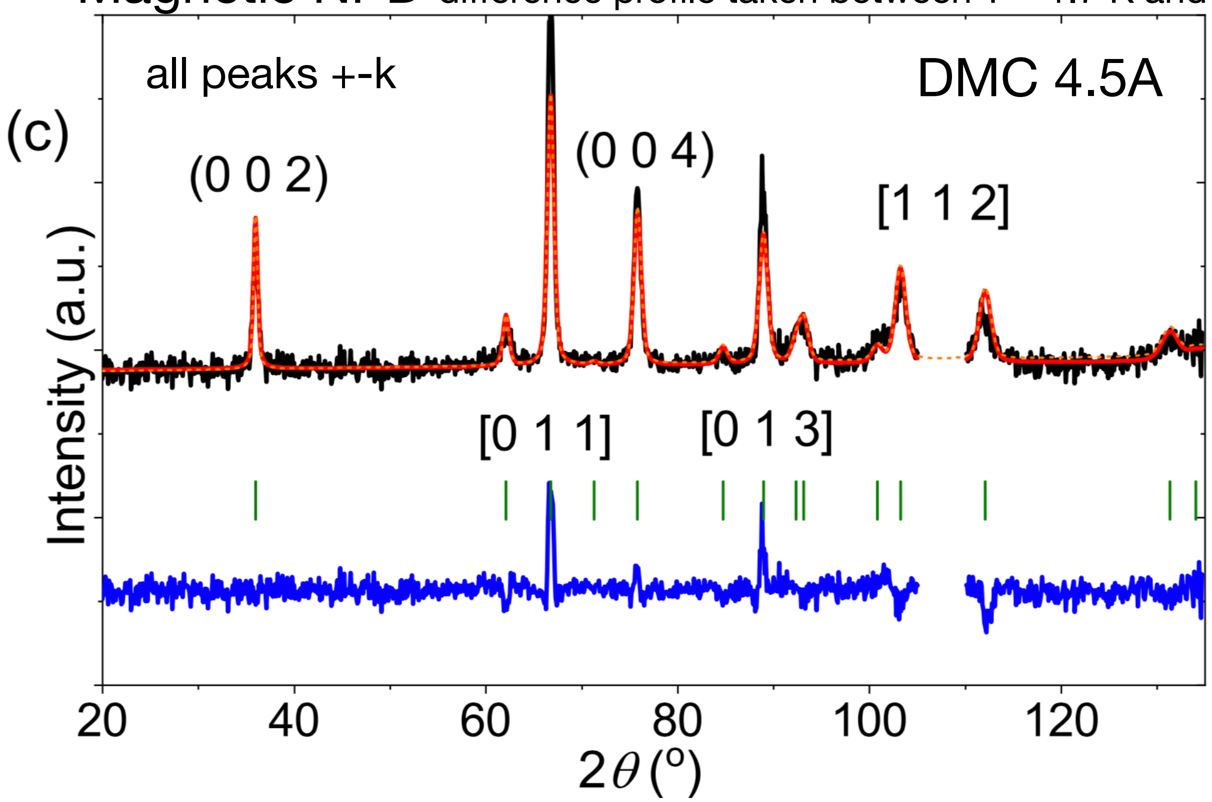
Magnetic peaks well seen from both powder and s.c. neutron diffraction

CeAlGe

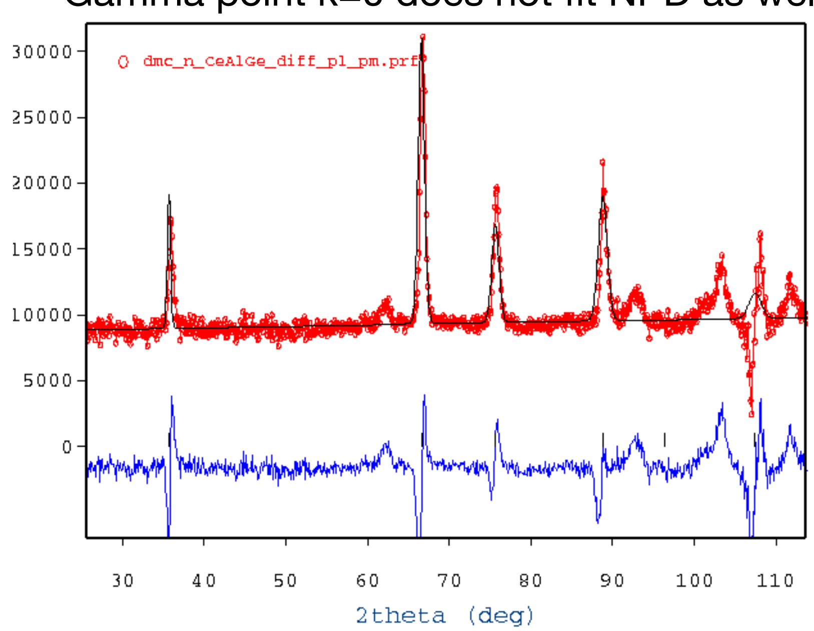
k1=[g,0,0], SM point of BZ, g=0.06503(22) ~65Å

Single crystal

Magnetic NPD difference profile taken between T = 1.7 K and 10 K



Gamma point k=0 does not fit NPD as well



P. Puphal, et al, Physical Review Letters, 124, 017202 (2020)

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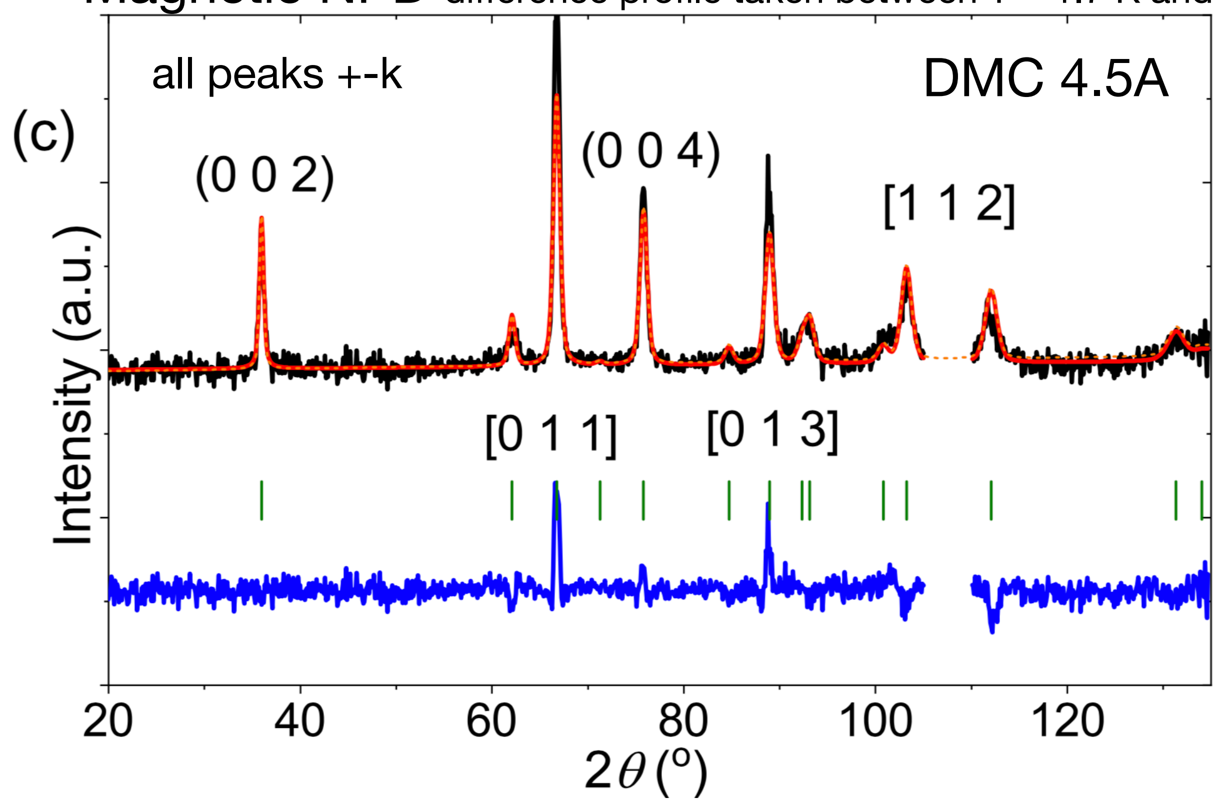
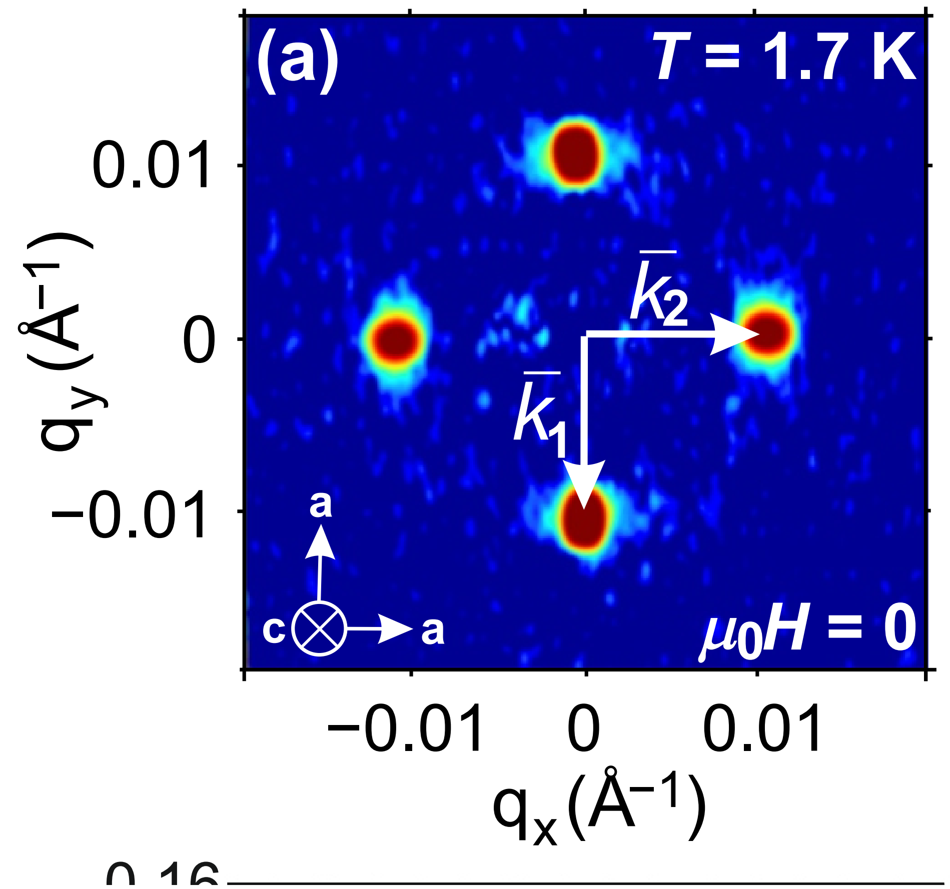
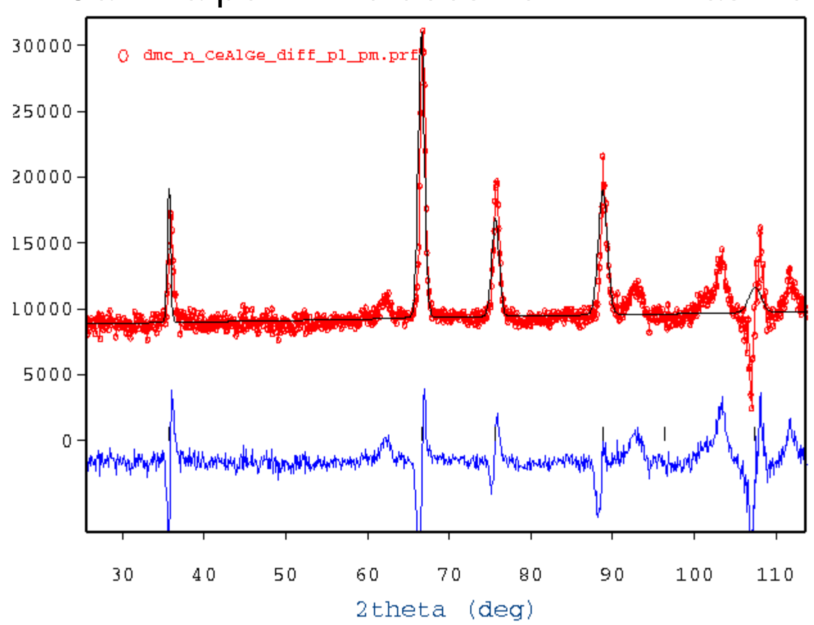


k1=[g,0,0], k2=[0,g,0]

Single crystal



Gamma point k=0 does not fit NPD as well



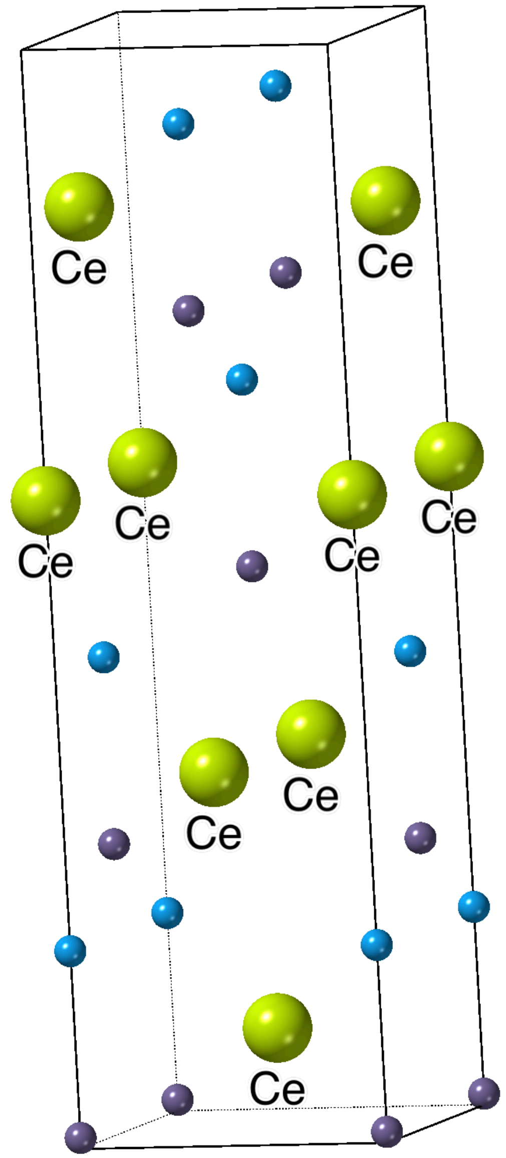
P. Puphal, et al, Physical Review Letters, 124, 017202 (2020)

Analysis of magnetic symmetry in CeAlGe

- one propagation vector 1k (±k) magnetic structure
- 2k (full propagation vector star) magnetic structure: actual solution supported by magnetisation, topological hall effect and calculation of topological charges
- both 1k and 2k-structures give similar good description of neutron diffraction intensities

One k-case, standard representation analysis without magnetic group symmetry arguments.

Space group $I4_1md$:
8 symops & I-centering,
Ce 4a (0,0,z) single magnetic Ce site: 4 atoms per cell



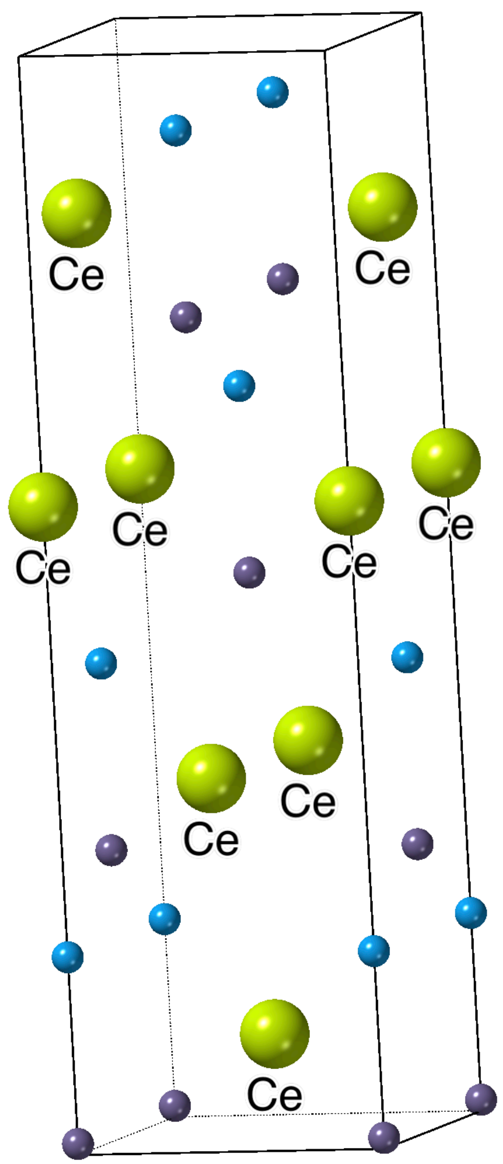
One k-case, standard representation analysis without magnetic group symmetry arguments.

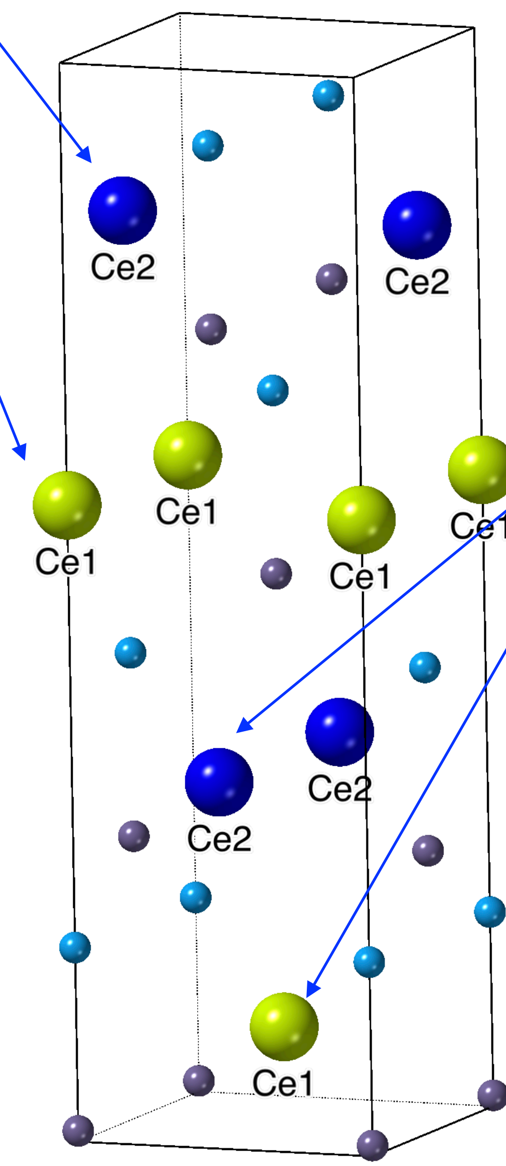
Space group $I4_1md$:
8 symops & I-centering,
Ce 4a (0,0,z) single magnetic Ce site: 4 atoms per cell

4(a) $\mathrm{Ce1}(0,0,z)$

$\mathrm{Ce2}(0,\frac{1}{2},z+\frac{1}{4})$

$\bar{y}, x+\frac{1}{2}, z+\frac{1}{4}$

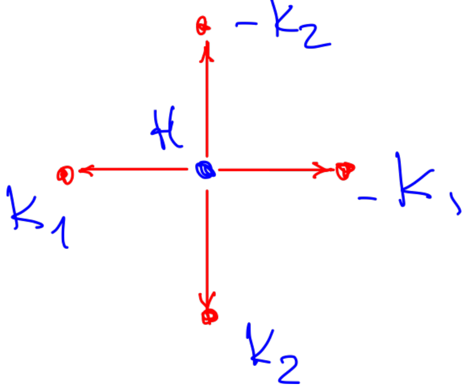




Two other Ce’s are generated by I-centering translations (½, ½, ½)+

One k-case, standard representation analysis without magnetic group symmetry arguments: Space group $I4_1md$, Ce 4a (0,0,z)

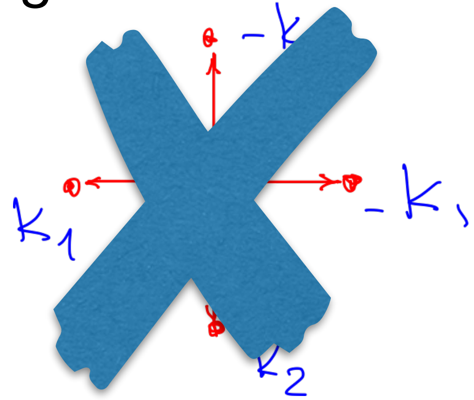
Propagation vector star $\{[g, 0, 0], [0, g, 0]\}$



$$\boldsymbol{k}_1 \xrightarrow[\bar{y}, x + \frac{1}{2}, z + \frac{1}{4}]{} \boldsymbol{k}_2$$

One k-case, standard representation analysis without magnetic group symmetry arguments: Space group $I4_1md$, Ce 4a (0,0,z)

Propagation vector star $\{[g,0,0],[0,g,0]\}$

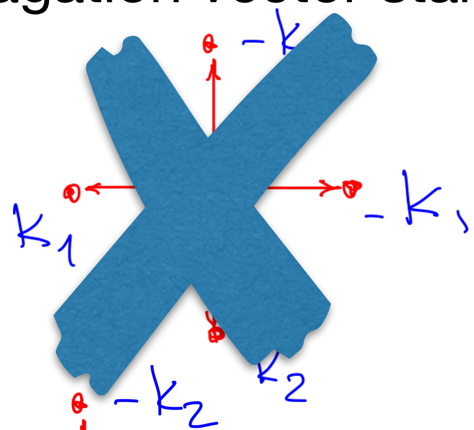


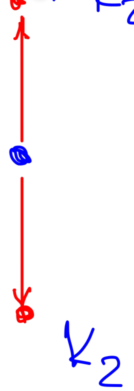
$$\boldsymbol{k}_1 \xrightarrow{\;\bar{y},\, x+\frac{1}{2},\, z+\frac{1}{4}\;} \boldsymbol{k}_2$$

One k-case, standard representation analysis without magnetic group symmetry arguments: Space group $I4_1md$, Ce 4a (0,0,z)

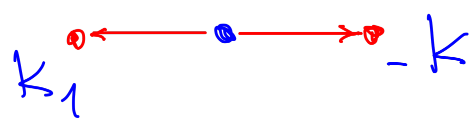
Propagation vector star $\{[g,0,0],[0,g,0]\}$

$$\mathbf{k}_1 \xrightarrow{\bar{y},\, x+\frac{1}{2},\, z+\frac{1}{4}} \mathbf{k}_2$$





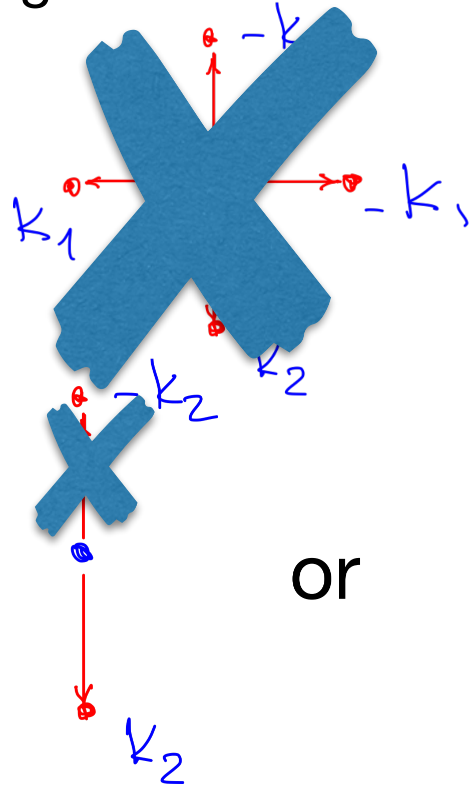
or



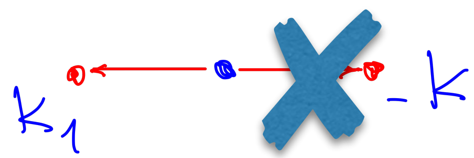
One k-case, standard representation analysis without magnetic group symmetry arguments: Space group $I4_1md$, Ce 4a (0,0,z)

Propagation vector star $\{[g,0,0],[0,g,0]\}$

$$\boldsymbol{k}_1 \xrightarrow{\bar{y}, x+\frac{1}{2}, z+\frac{1}{4}} \boldsymbol{k}_2$$



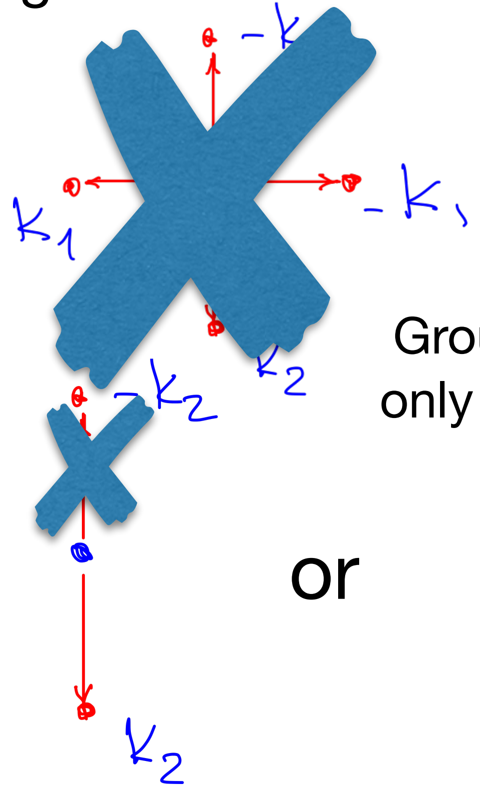
or



One k-case, standard representation analysis without magnetic group symmetry arguments: Space group $I4_1md$, Ce 4a (0,0,z)

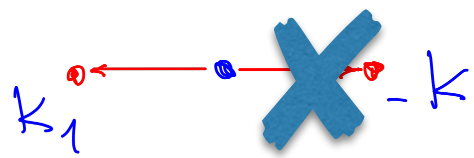
Propagation vector star $\{[g,0,0],[0,g,0]\}$

$$\mathbf{k}_1 \xrightarrow{\bar{y},\, x+\frac{1}{2},\, z+\frac{1}{4}} \mathbf{k}_2$$



Group G_k has x,y,z
only 2 symops $x,\bar{y},z$ out of 8!

or



$$\mathrm{Ce1}(0,0,z)$$
$$\mathrm{Ce2}(0,\frac{1}{2},z+\frac{1}{4})$$

Two independent sites. No symmetry relations between Ce1 and Ce2

One k-case, standard representation analysis without magnetic group symmetry arguments: Space group $I4_1md$, Ce 4a (0,0,z)

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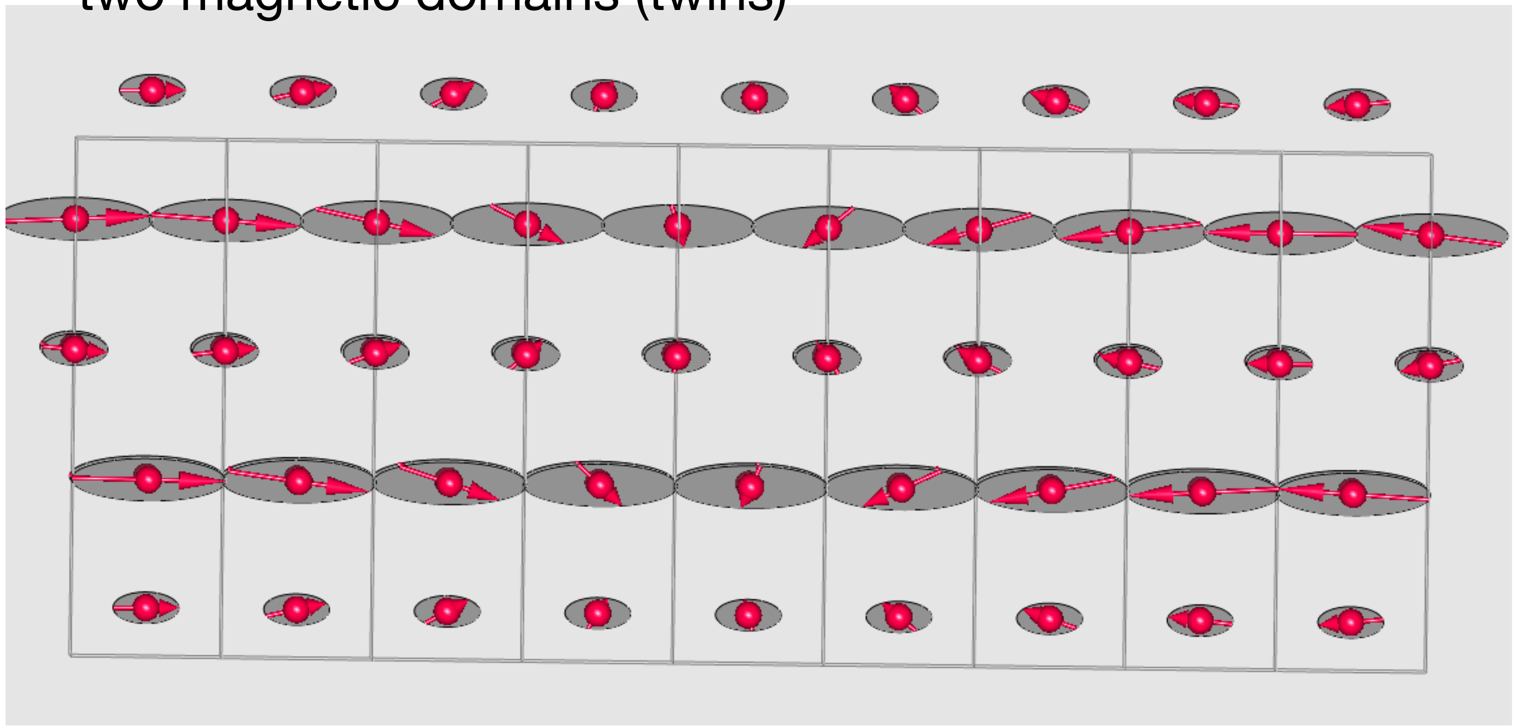
$k=|\boldsymbol{k}_1|=|\boldsymbol{k}_2|=g$

$$\mathbf{M}_{Ce(i)} = m_{ix}\sin(2\pi kx)\mathbf{e}_{\boldsymbol{x}} + m_{iz}\sin(2\pi kx+\varphi_i)\mathbf{e}_{\boldsymbol{z}}, \quad i=1,2$$

Experimental values:
Ce1: m_{1x}= -0.64(1), m_{1z} =-0.30(6)
Ce2: m_{2x}= -1.50(2), m2z = 0.46(8)

$\varphi_1=\varphi_2\approx 90°$

Solution: tau2/SM2 irreducible representation
Cycloid in ac-plane for k1=[g,0,0], in bc=lane for k2=[0,g,0]
two magnetic domains (twins)



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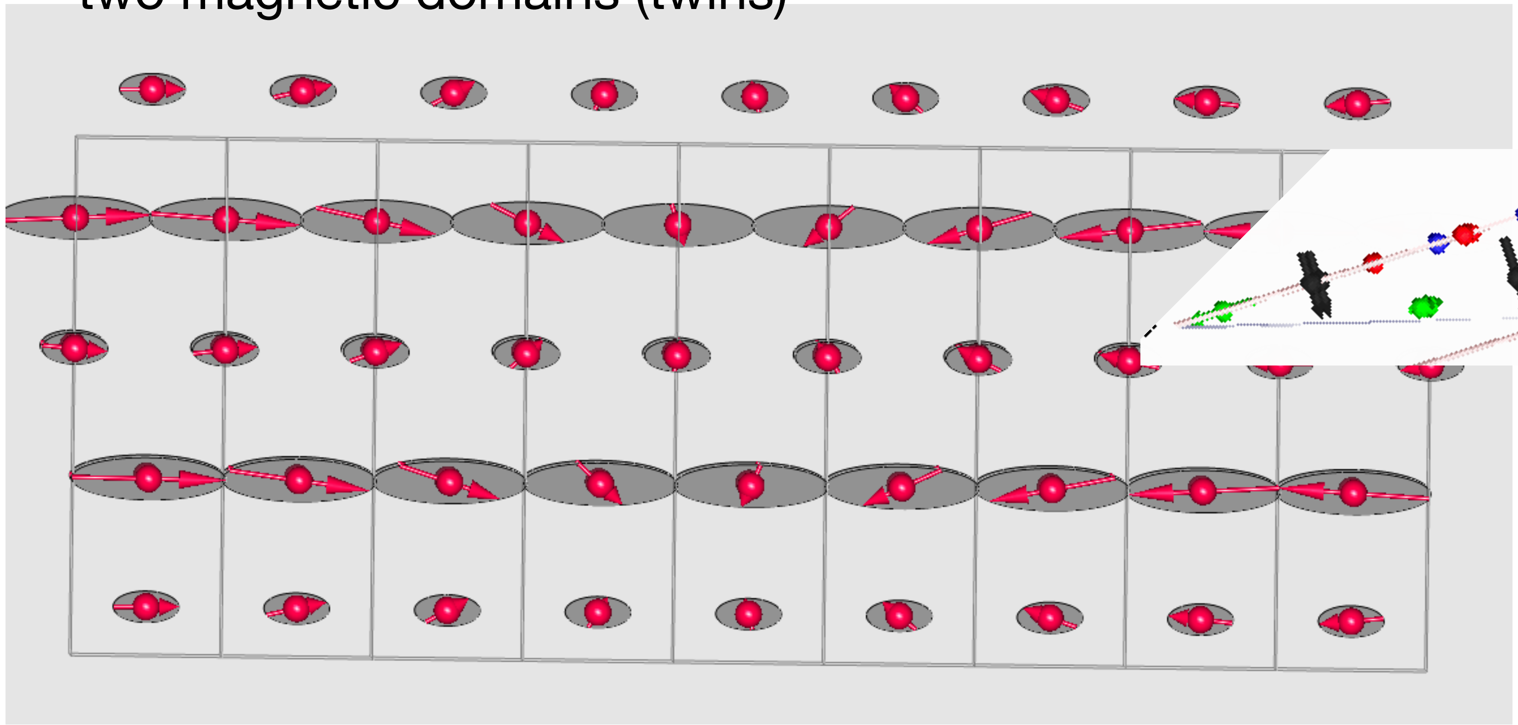
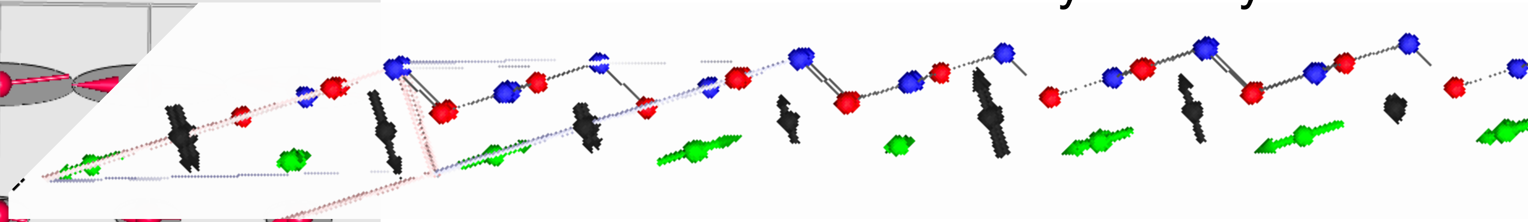


Note: if $\varphi_1=\varphi_2=0$ → amplitude modulation, different symmetry



Symmetry of cycloid. 3D+1 superspace group for SM2 irrep

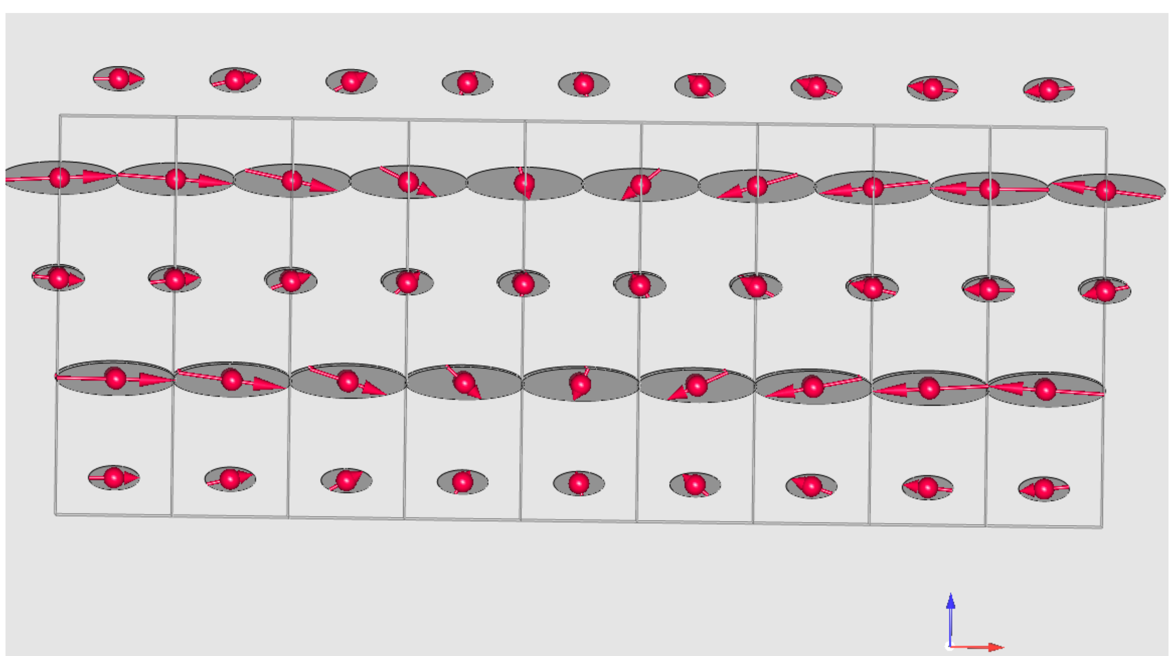
ISOTROPY Software Suite http://iso.byu.edu
Harold T. Stokes, Dorian M. Hatch, and Branton J. Campbell, Department of Physics and Astronomy, Brigham Young University, Provo, Utah 84606, USA,

$I4_1md1'$

Advantage of magnetic symmetry even for 1k-case

I2mm1'(0,0,g)0s0s, basis={(0,0,-1,0),(0,1,0,0),(1,0,0,0),(0,0,0,1)}, k-active= (g,0,0)

atom	site	x	y	z	occ	mx	my	mz	
Ce1_1	2a	0.41000	0.00000	0.00000	1.00000				
						mx1	0	mz1	k1 amplitude
						0	0.00000	90	k1 phase, degrees
Ce1_2	2b	0.66000	0.00000	0.50000	1.00000				
						mx2	0	mz2	k1 amplitude
						0	0.00000	90	k1 phase, degrees



$$\mathbf{M}_{Ce(i)} = m_{ix}\sin(2\pi kx)\mathbf{e}_{\boldsymbol{x}} + m_{iz}\cos(2\pi kx)\mathbf{e}_{\boldsymbol{z}}, \quad i=1,2$$

phase shift 90 degrees is fixed by symmetry!

Experimental values:
Ce1: m_{1x}= -0.64(1), m_{1z} =-0.30(6)
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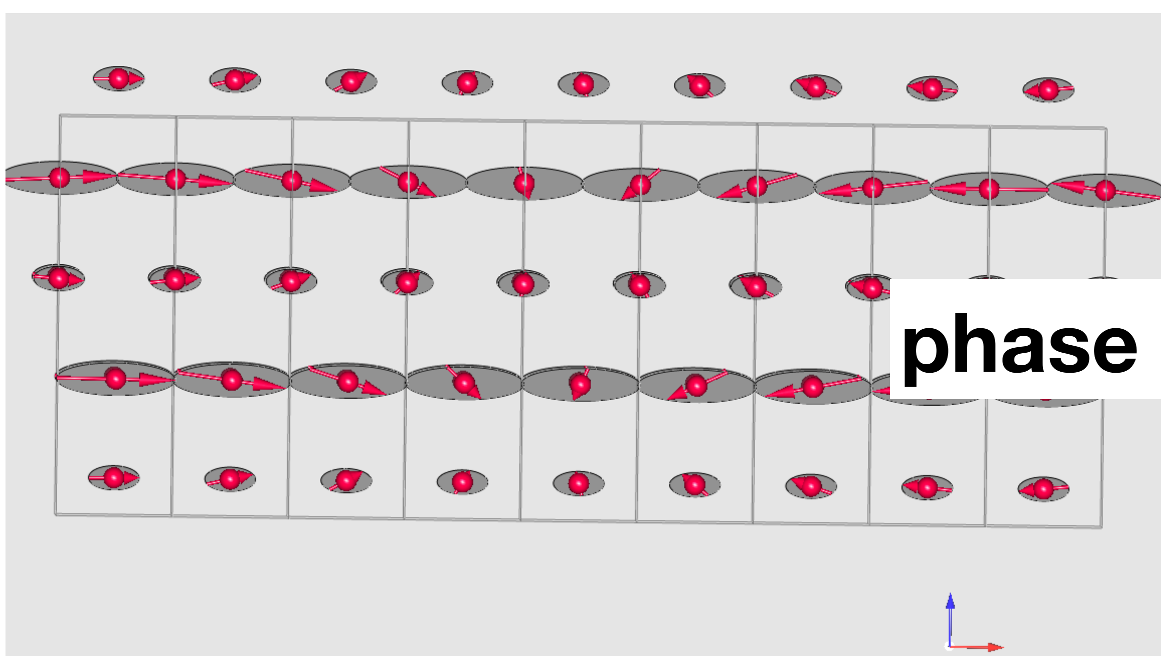

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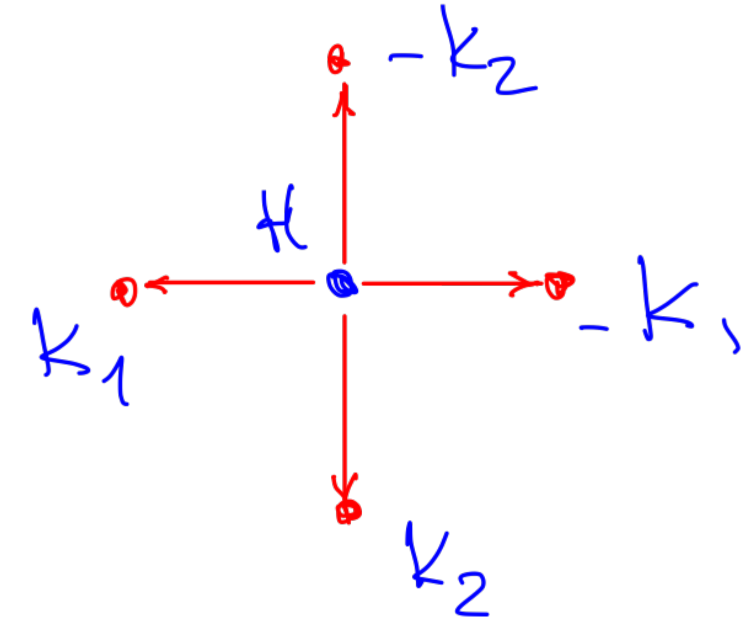
CeAlGe: Maximal symmetry full star superspace 3D+2 magnetic group I4_1md1'(a00)000s(0a0)0s0s

Parent Space Group: 109 I4_1md C4v-11,
Ce1 4a (0,0,z), z=-0.41000 single Ce site

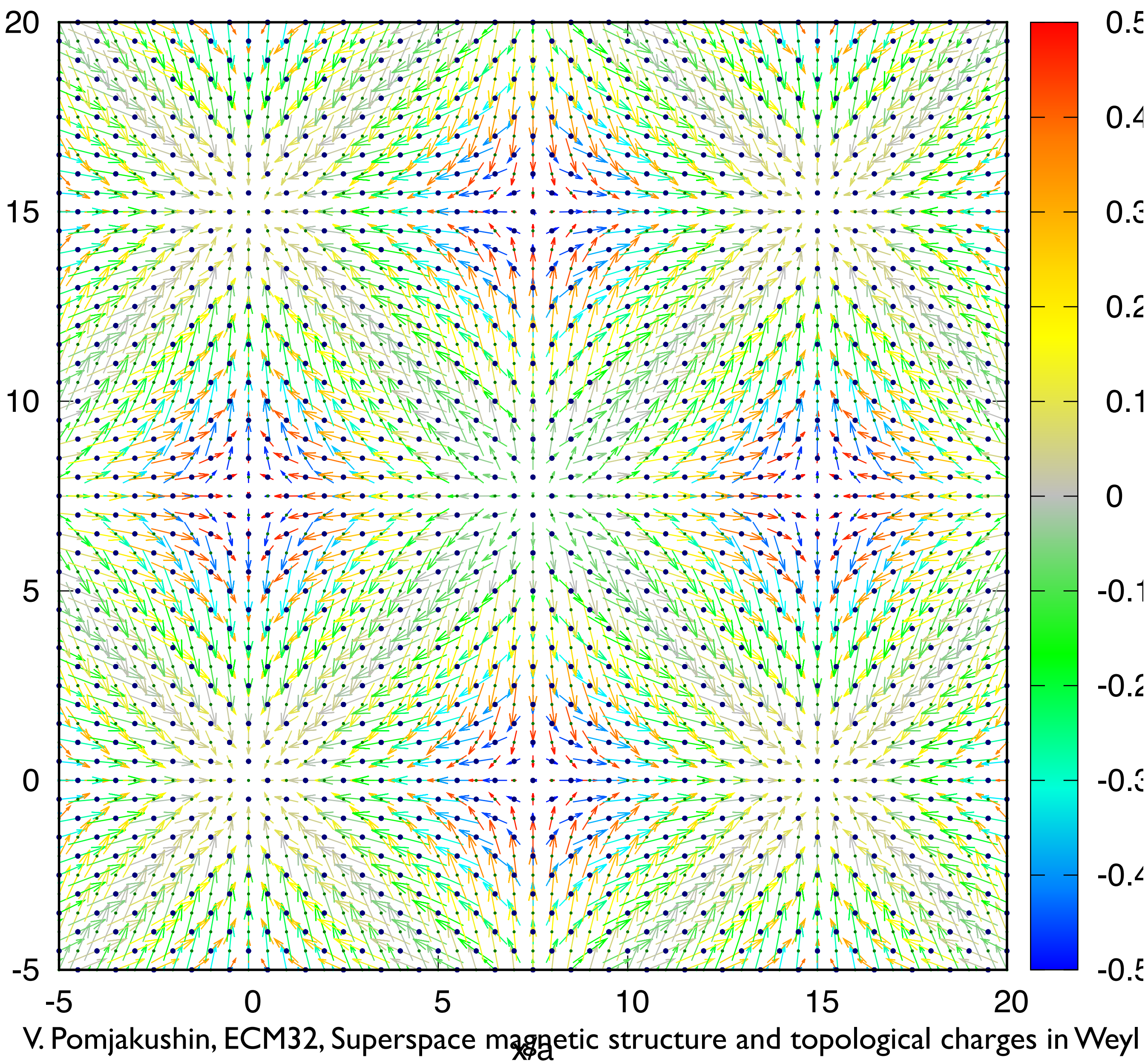
IR: mSM2 , k-active= (g,0,0),(0,g,0)
P (g,0;g,0) 109.2.67.4.m240.? I4_1md1'(a,0,0)000s(0,a,0)0s0s

k1=[g,0,0], SM point of BZ,
g=0.06503(22): four arms



View along the z-(c-)axis of the magnetic structure of CeAlGe.
The x- and y-axes are in units of in-plane lattice parameter a.

(M_x,M_y) components in the xy plane, M_z-component by color



ISOTROPY Software Suite
Harold T. Stokes, Dorian M. Hatch, and Branton J. Campbell, Department of Physics and Astronomy, Brigham Young University, Provo, Utah 84606, USA,

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Parent Space Group: 109 I4_1md C4v-11,

Ce1 4a (0,0,z), z=-0.41000 single Ce site

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P (g,0;g,0) 109.2.67.4.m240.? I4_1md1'(a,0,0)000s(0,a,0)0s0s

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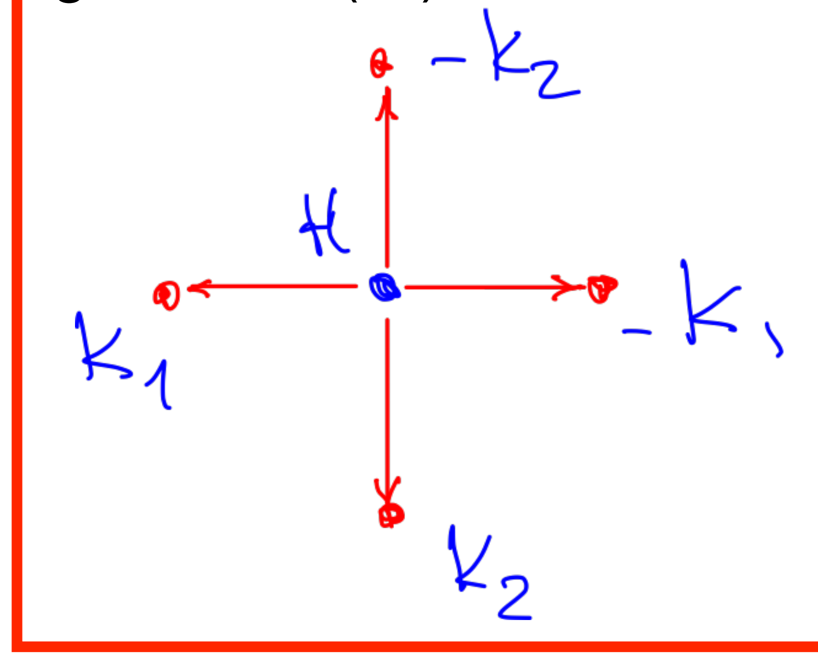

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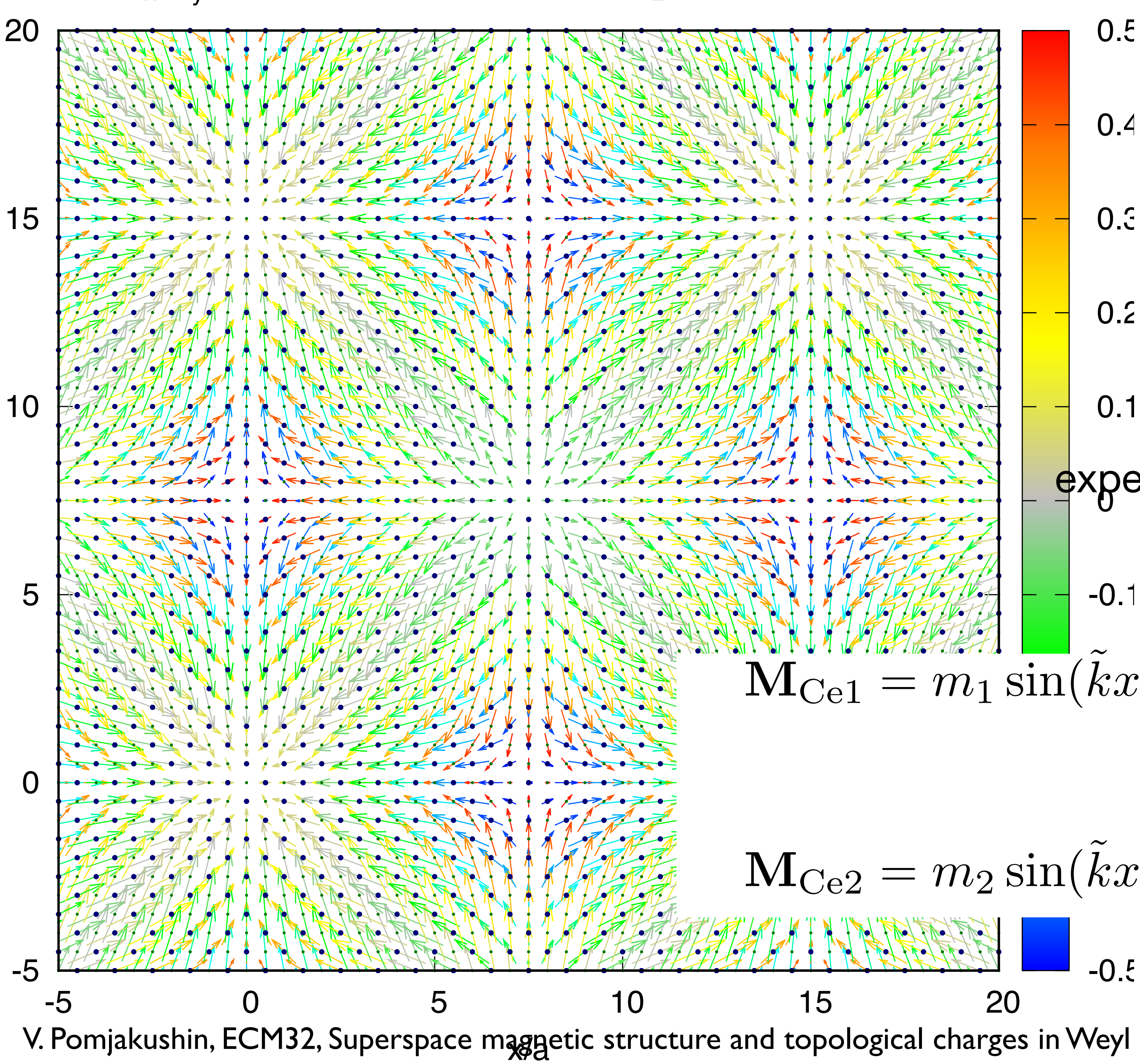

All Ce are equivalent and their moments are given symmetrically by 4 parameters

experiment: (m1,m2,m3,m4) = (0.44(1), 1.02(1), −0.21(5), 0.29(7)) μ_B.

$\tilde{k}=2\pi|\boldsymbol{k}_1|=2\pi|\boldsymbol{k}_2|=2\pi g$

$$\mathbf{M}_{\mathrm{Ce1}} = m_1 \sin(\tilde{k}x)\mathbf{e}_{\boldsymbol{x}} + m_2 \sin(\tilde{k}y)\mathbf{e}_{\boldsymbol{y}} + \Big(m_3 \cos(\tilde{k}x) + m_4 \cos(\tilde{k}y)\Big)\,\mathbf{e}_{\boldsymbol{z}}$$

$$\mathbf{M}_{\mathrm{Ce2}} = m_2 \sin(\tilde{k}x)\mathbf{e}_{\boldsymbol{x}} + m_1 \sin(\tilde{k}y)\mathbf{e}_{\boldsymbol{y}} + \Big(m_4 \cos(\tilde{k}x) + m_3 \cos(\tilde{k}y)\Big)\,\mathbf{e}_{\boldsymbol{z}}$$

ISOTROPY Software Suite

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CeAlGe: Maximal symmetry full star superspace 3D+2 magnetic group I4_1md1'(a00)000s(0a0)0s0s

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IR: mSM2 , k-active= (g,0,0),(0,g,0)
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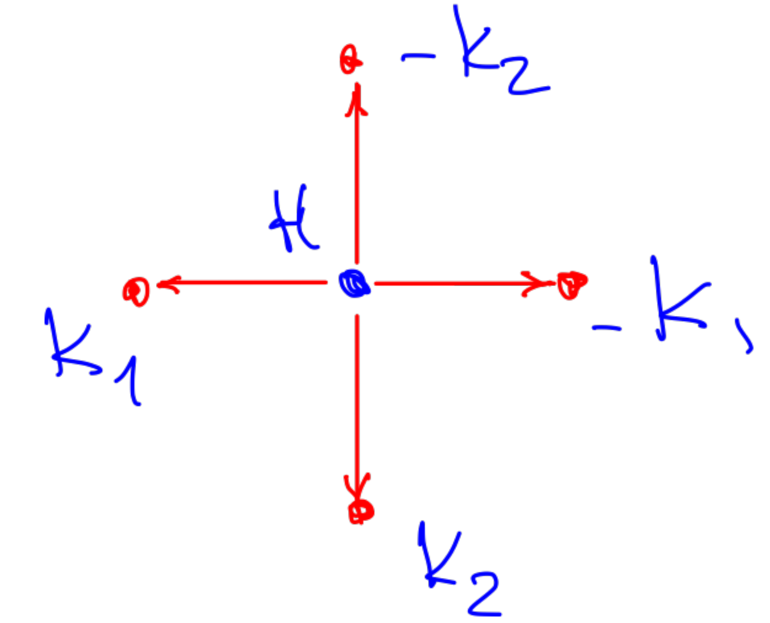

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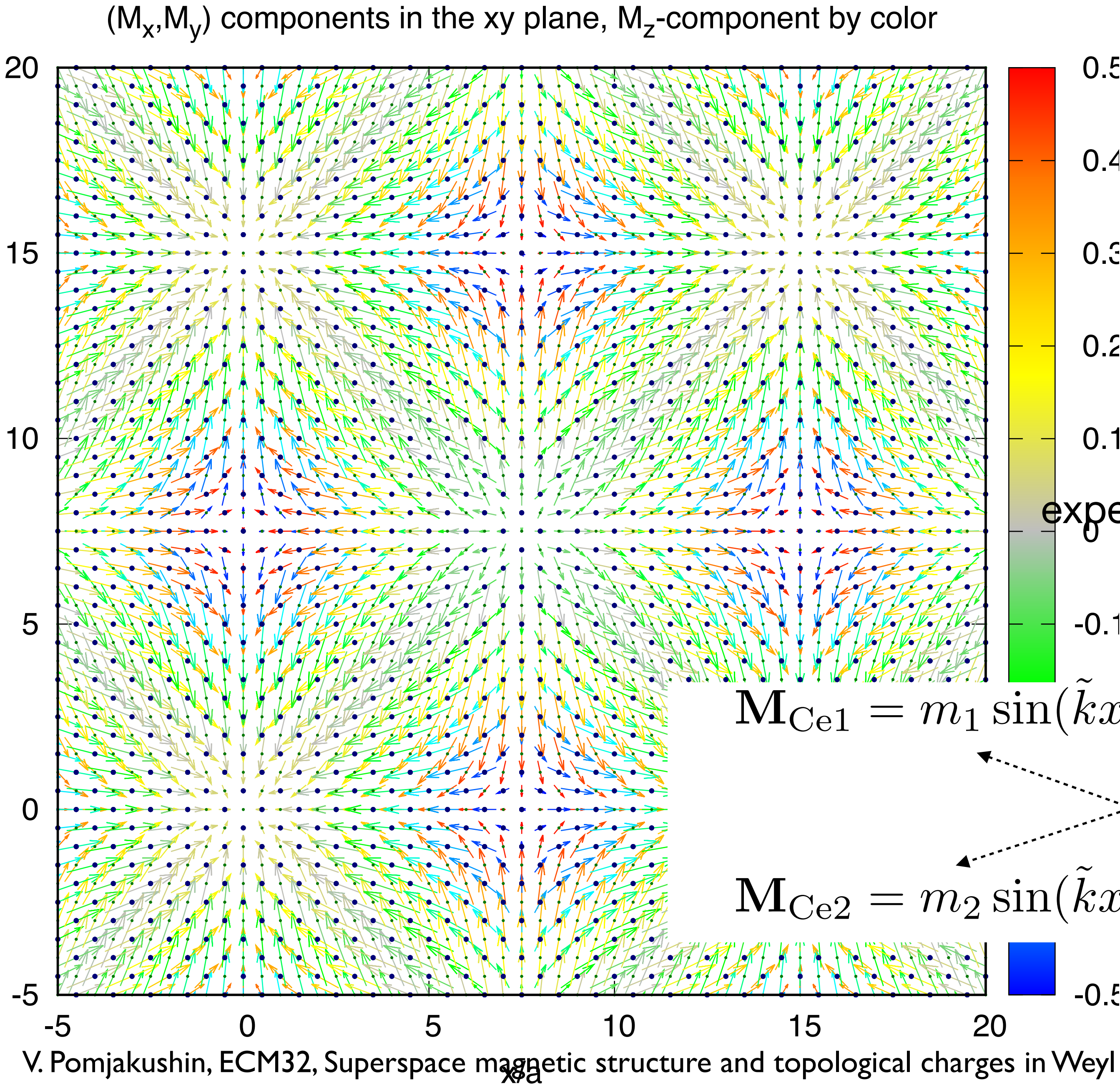

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$$\mathbf{M}_{\mathrm{Ce2}} = m_2 \sin(\tilde{k}x)\mathbf{e}_{\boldsymbol{x}} + m_1 \sin(\tilde{k}y)\mathbf{e}_{\boldsymbol{y}} + \Big(m_4 \cos(\tilde{k}x) + m_3 \cos(\tilde{k}y)\Big)\,\mathbf{e}_{\boldsymbol{z}}$$

ISOTROPY Software Suite

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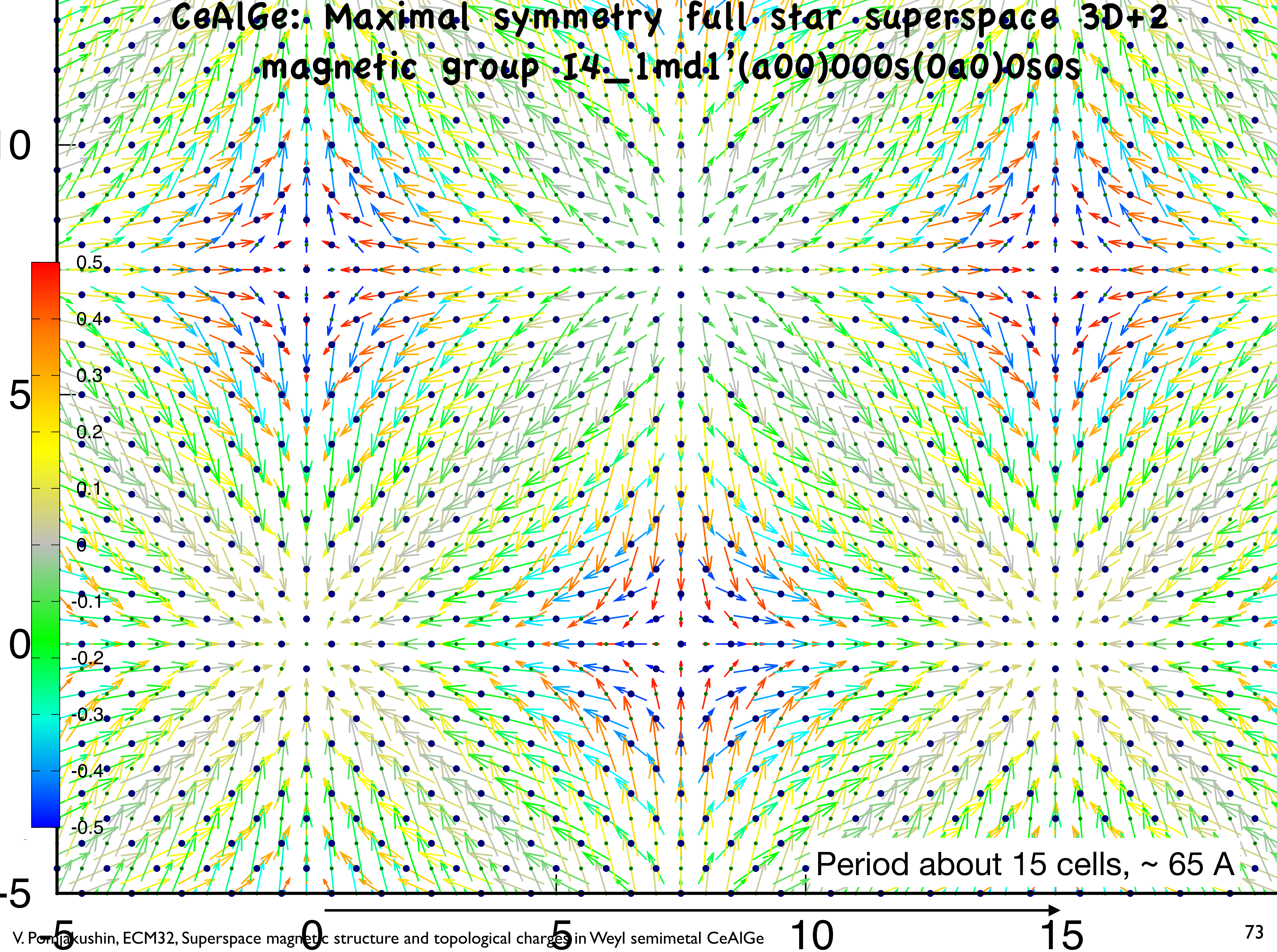


CeAlGe: Maximal symmetry full star superspace 3D+2
magnetic group I4_1md1'(a00)000s(0a0)0s0s
Period about 15 cells, ~ 65 A
0.5
0.4
0.3
0.2
0.1
0
-0.1
-0.2
-0.3
-0.4
-0.5
10
5
0
-5
-5
0
5
10
15

Topological density and charge. H=0

15:40 Tuesday, 20. August 2019 talk MS24-05 "Superspace Magnetic Structure and Topological Charges in Weyl Semimetal CeAlGe" at MS24: Magnetic Order: Methods and Properties

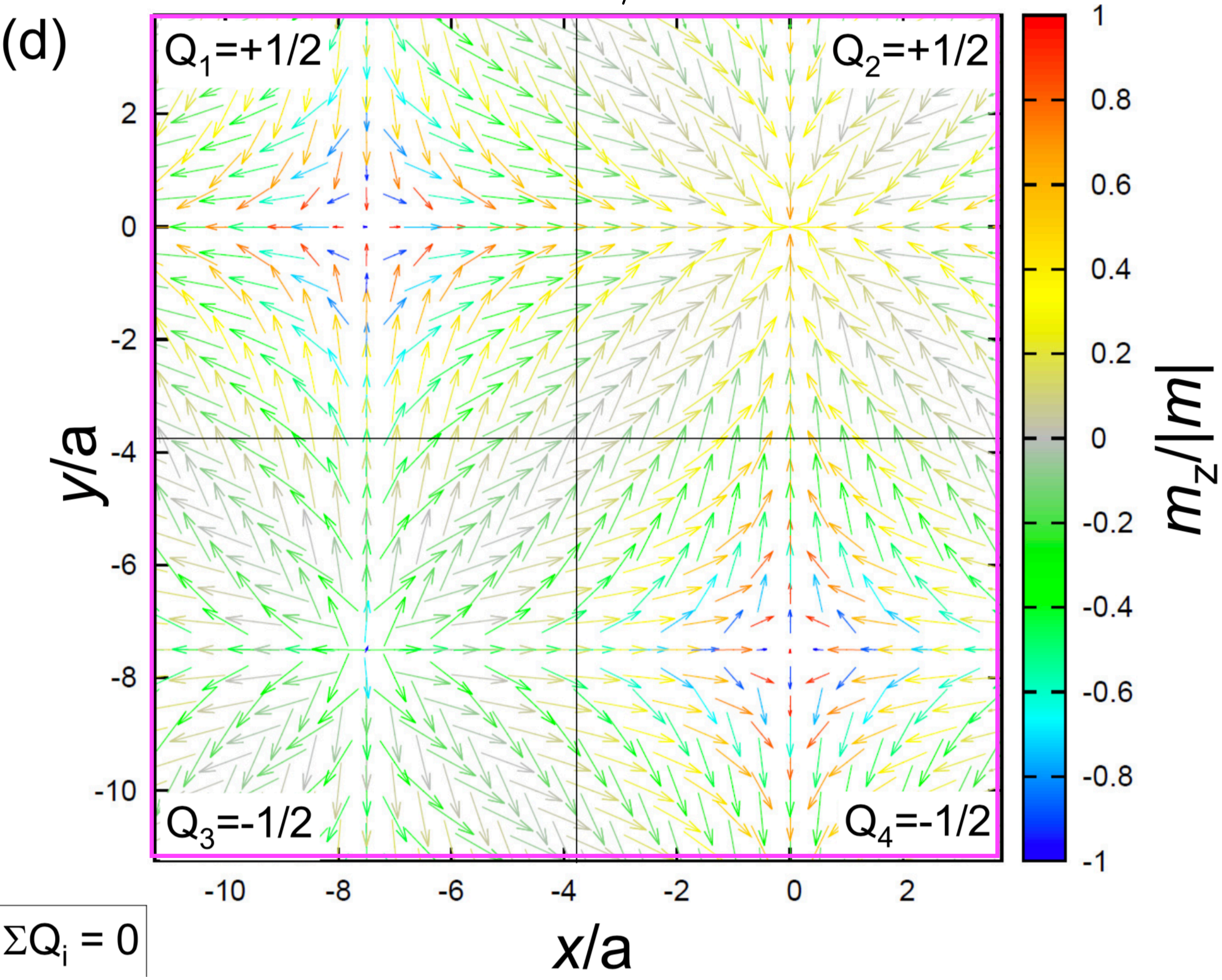
experiment: (m1,m2,m3,m4) = (0.44(1), 1.02(1), −0.21(5), 0.29(7)) μ_B.

$$\mathbf{n} = \mathbf{M}/M$$

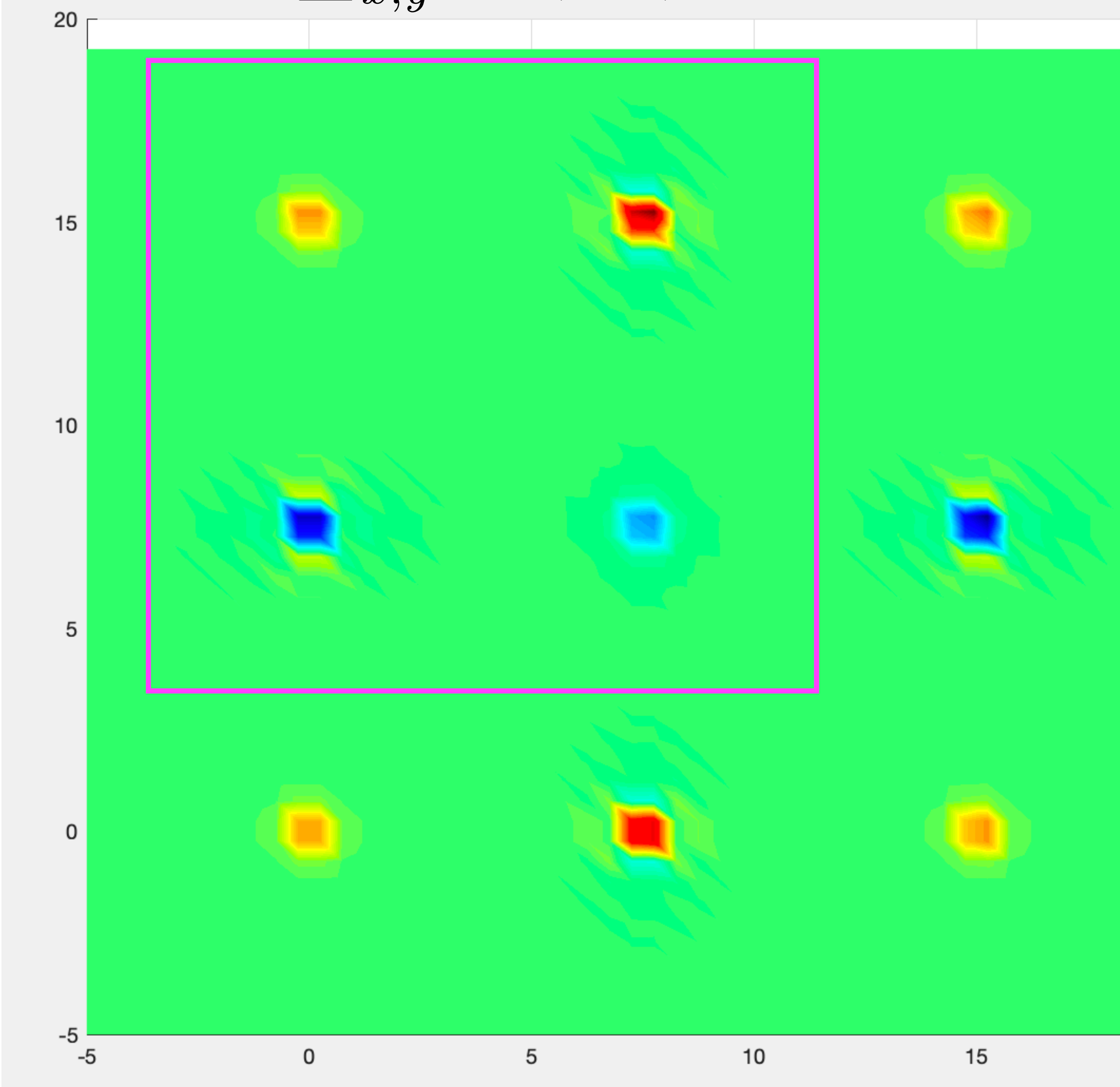
$$\Delta w(x,y) = \frac{1}{4\pi}\left(\mathbf{n}\cdot[\Delta\mathbf{n}_x \times \Delta\mathbf{n}_y]\right)$$

solid angle per square placket

$$Q = \sum_{x,y} \Delta w(x,y)$$



$\Sigma Q_i = 0$



$$\mathbf{M}_{\mathrm{Ce2}} = m_2\sin(\tilde{k}x)\mathbf{e}_{\boldsymbol{x}} + m_1\sin(\tilde{k}y)\mathbf{e}_{\boldsymbol{y}} + \left(m_4\cos(\tilde{k}x) + m_3\cos(\tilde{k}y)\right)\mathbf{e}_{\boldsymbol{z}}$$

$$\mathbf{M}_{\mathrm{Ce1}} = m_1\sin(\tilde{k}x)\mathbf{e}_{\boldsymbol{x}} + m_2\sin(\tilde{k}y)\mathbf{e}_{\boldsymbol{y}} + \left(m_3\cos(\tilde{k}x) + m_4\cos(\tilde{k}y)\right)\mathbf{e}_{\boldsymbol{z}}$$

$\tilde{k}=2\pi|\boldsymbol{k}_1|=2\pi|\boldsymbol{k}_2|=2\pi g$

Relation of magnetic Shubnikov/superspace symmetry and representation analysis RA

Paramagnetic crystallographic space group (*PSG*)

Propagation vector of magnetic structure **k**

choose one irreducible representation (*irrep*) of *PSG*

magnetic symmetry

representation

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representation

Construction of basis functions (normal modes)

Constraints on the mixing coefficients of basis function for >1D *irrep* and/or multi-arm star of **k**,

{-**k**,**k**} star for incommensurate.

Magnetic structure made from linear combination of normal modes.

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magnetic symmetry

representation

is *irrep* real and 1D, and single arm {**k**}-star?

Yes

Shubnikov from *PSG*
Symop g that have *irrep*(*g*)=-1
are primed in Sh-group

Construction of basis functions (normal modes)

Constraints ? the mixing coefficients of basis function for >1D *irrep* and/or multi-arm star of **k**, {-**k**,**k**} star for incommensurate.

Magnetic structure made from admissible spin directions in Sh-group

equivalent

Magnetic structure made from linear combination of normal modes.

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choose one irreducible representation (*irrep*) of *PSG*

magnetic symmetry

representation

is *irrep* real and 1D, and single arm {**k**}-star?

No

is **k** commensurate?

Yes

Yes

Construction of basis functions (normal modes)

Shubnikov from *PSG* *Symop g* that have *irrep*(*g*)=-1 are primed in Sh-group

combining nD *irrep* based on full star {**k**} & c.c into real 2nD

Constraints ? the mixing coefficients of basis function for >1D *irrep* and/or multi-arm star of **k**, {-**k**,**k**} star for incommensurate.

choice of direction of order parameter for *irrep*

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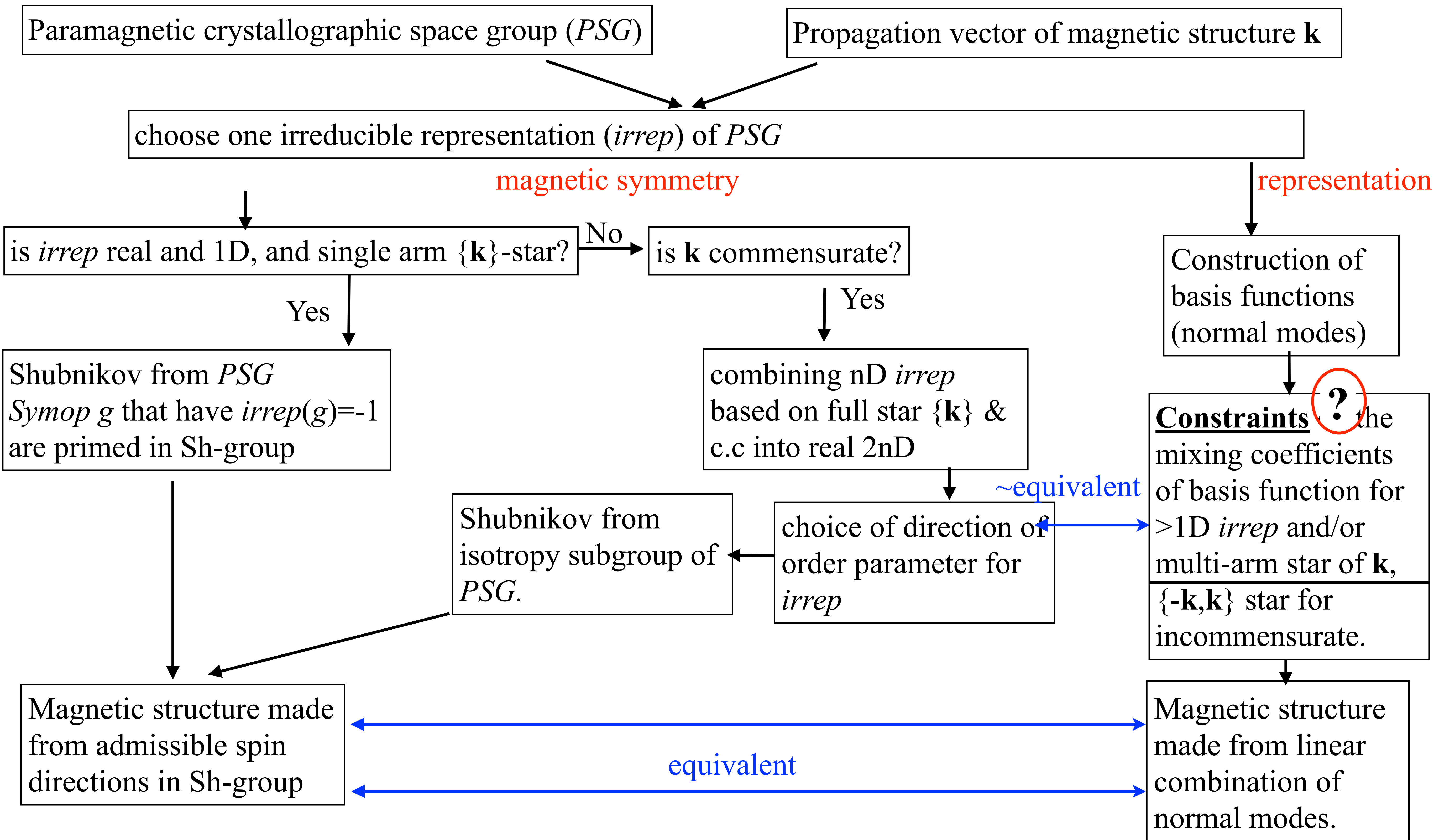
Relation of magnetic Shubnikov/superspace symmetry and representation analysis RA



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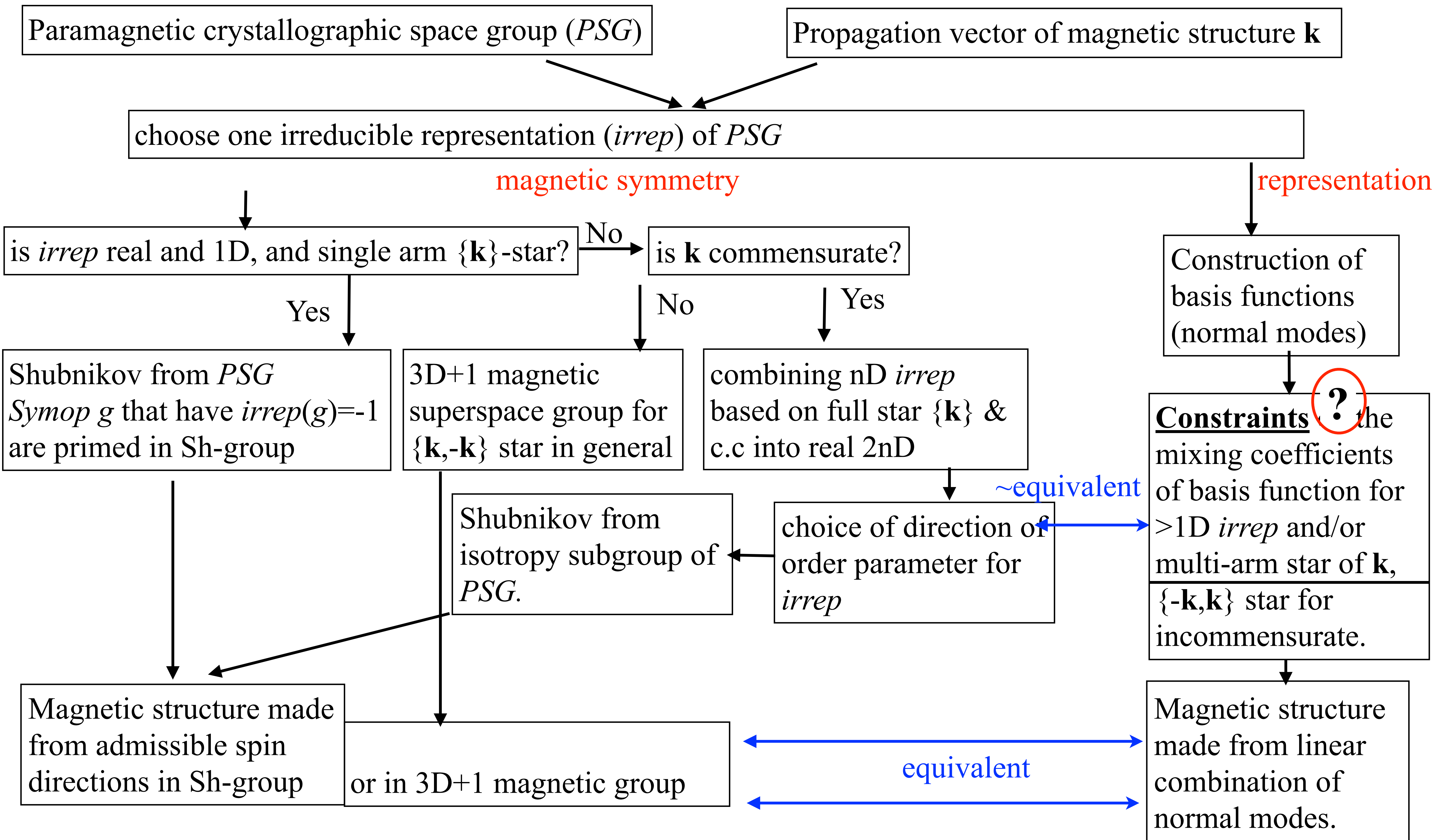

Relation of magnetic Shubnikov/superspace symmetry and representation analysis RA

Paramagnetic crystallographic space group (*PSG*)

Propagation vector of magnetic structure **k**

The disadvantage of using only RA:

In general: there are no rules to make **constraints**, ? (except ones based on physical grounds)

but the **constraints** appear in a natural way from magnetic group symmetry arguments

Shubnikov from *PSG*
Symop g that have *irrep*(*g*)=-1 are primed in Sh-group

3D+1 magnetic superspace group for {**k**,-**k**} star in general

combining nD *irrep* based on full star {**k**} & c.c into real 2nD

Constraints ? the mixing coefficients of basis function for >1D *irrep* and/or multi-arm star of **k**, {-**k**,**k**} star for incommensurate.

equivalent

choice of direction of order parameter for *irrep*

Shubnikov from isotropy subgroup of *PSG*.

Magnetic structure made from admissible spin directions in Sh-group

or in 3D+1 magnetic group

equivalent

Magnetic structure made from linear combination of normal modes.

Thank you!