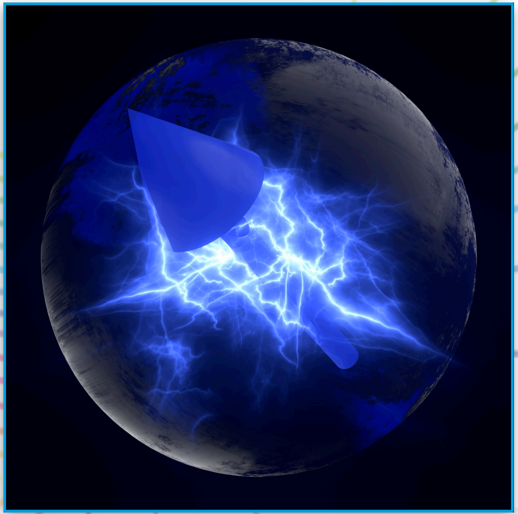
Introduction to

Spherical Neutron Polarimetry

and its application to condensed matter physics



Navid Qureshi

Outline

Content of the presentation

- Historical context
- The **Cryo**genic **P**olarisation **A**nalysis **D**evice
- Key contributions to magnetoelectric and multiferroic systems
- Conclusions

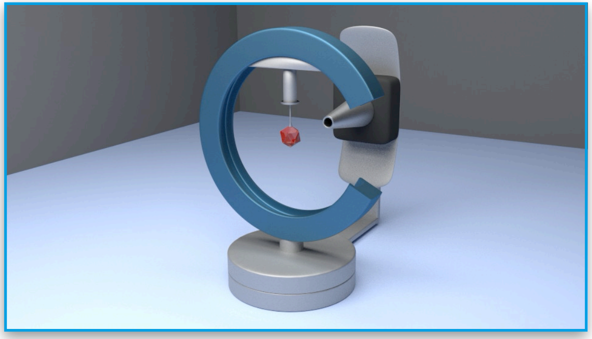
Historical context

Motivation

THE PROBLEM

Unpolarized neutrons only give access to the modulus of the magnetic interaction vector $\mathbf{M}_\perp$ via the observed intensity $I \sim \mathbf{M}_\perp \mathbf{M}_\perp^* = |M_\perp|^2$.

$$\mathbf{M}_\perp(\mathbf{Q}) = \hat{\mathbf{Q}} \times (\mathbf{M} \times \hat{\mathbf{Q}}) \qquad \mathbf{M}(\mathbf{Q}) = \sum_j \mathbf{S}_j f_j(\mathbf{Q}) \exp(i\mathbf{Q}\mathbf{r}_j)$$



THE RAY OF HOPE

In 1963 Blume and Maleev independently publish their equations relating the final neutron polarization to generalized cross sections.

Blume (1963) *Phys. Rev.* **130** 1670

Maleev et al. (1963) *Sov. Phys. Solid State.* **4** 2533

THE DREAM

Be able to measure all the terms in these equations precisely.

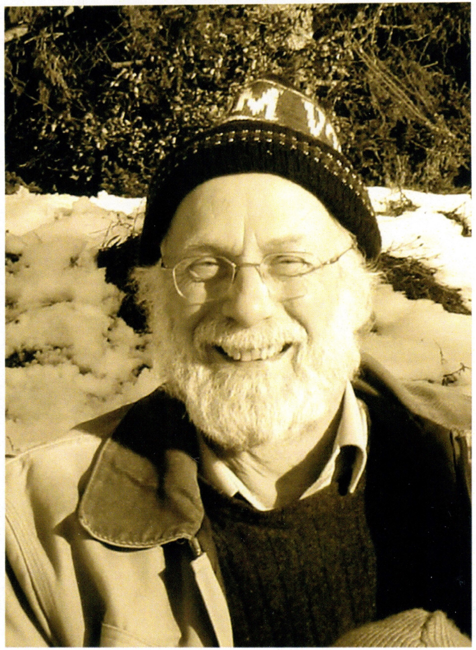
Historical context

The dream come true

THANK YOU FRANCIS !!!

- Polarizing neutron beams with Heusler alloy
- Cryoflipper (no wavelength and stray field dependency)
- **Cryo**genic **P**olarization **A**nalysis **D**evice for SNP

  (with major contributions by S. Pujol and P. J. Brown)
- ^{3}He spin filter cell and pumping station



Francis Tasset (1944 - 2023)

Historical context

Milestones (by Francis Tasset)

- **1963** Blume and Maleev independently publish the equations
- **1969** Riste, Moon and Koehler analyze polarization
- **1973** Alperin demonstrates that 3D polarization analysis is possible
- **1975** FT's sabbatical at ORNL working on superconducting Nb
- **1976** FT's cryoflipper based on superconducting screens
- **1986** First Cryopad taking advantage of Meissner shields
- **1993** FT introduces the term "Spherical Neutron Polarimetry"
- **1995** Cryopad II
- **1998** SNP is finally accepted and adopted by the experts
- **2002** Cryopad III (first project of ILL Millennium Programme)

$$\begin{aligned} \mathbf{P}'I &= \mathbf{P}(N^2 - \mathbf{M}_\perp \mathbf{M}_\perp^*) \\ &+ 2\Re[\mathbf{M}_\perp(\mathbf{P}\cdot\mathbf{M}_\perp^*)] \\ &+ 2\Re(N^*\mathbf{M}_\perp) \\ &+ \mathbf{P}\times 2\Im(N^*\mathbf{M}_\perp) \\ &- \Im(\mathbf{M}_\perp \times \mathbf{M}_\perp^*) \end{aligned}$$

$$\begin{aligned} I &= N^2 + \mathbf{M}_\perp \mathbf{M}_\perp^* \\ &+ 2\Re(\mathbf{P}\cdot N^*\mathbf{M}_\perp) \\ &+ \mathbf{P}\cdot\Im(\mathbf{M}_\perp \times \mathbf{M}_\perp^*) \end{aligned}$$

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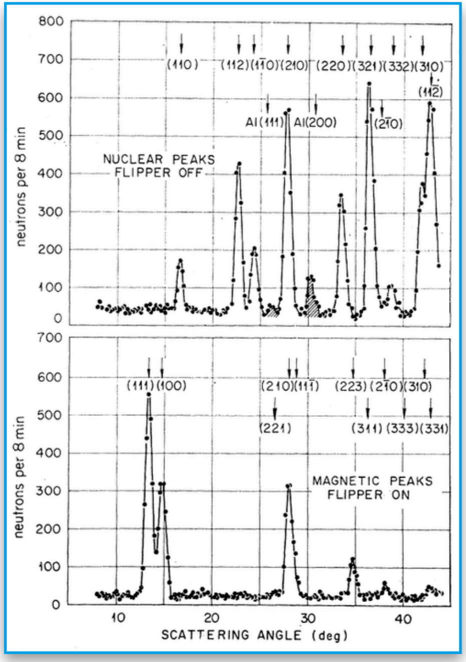

Moon et al. (1969) *Phys. Rev.* **181** 920

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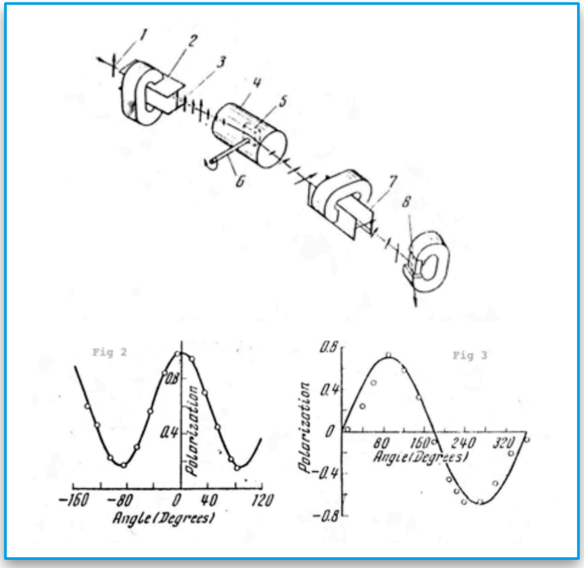

Alperin (1973) International Conference on Magnetism, Moscow, ed., Proc. ICM-73

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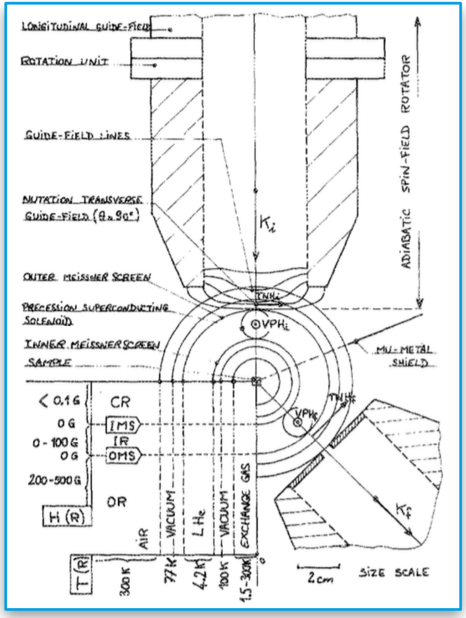
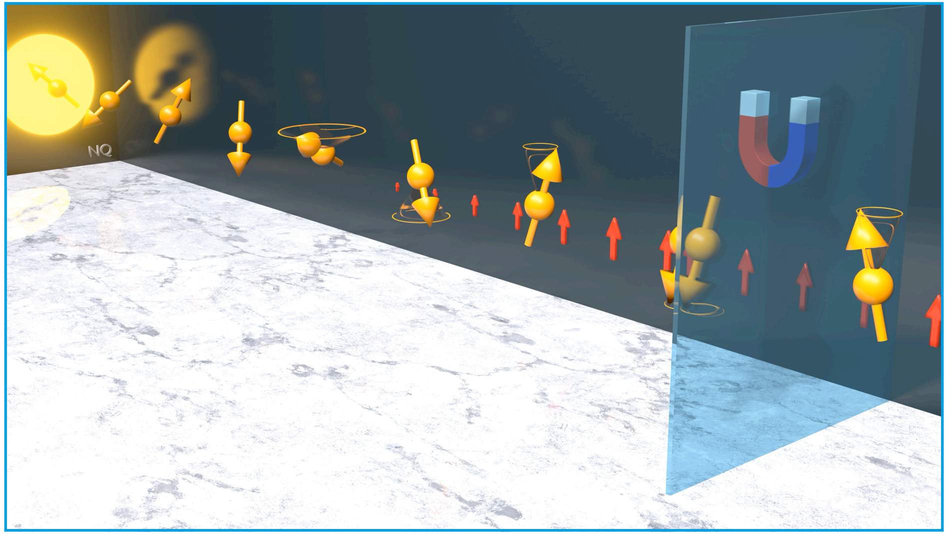


F.Tasset et al. (1988) *J. Appl. Phys.* **63** 3606

The Cryopad

Manipulation of polarised neutron beams

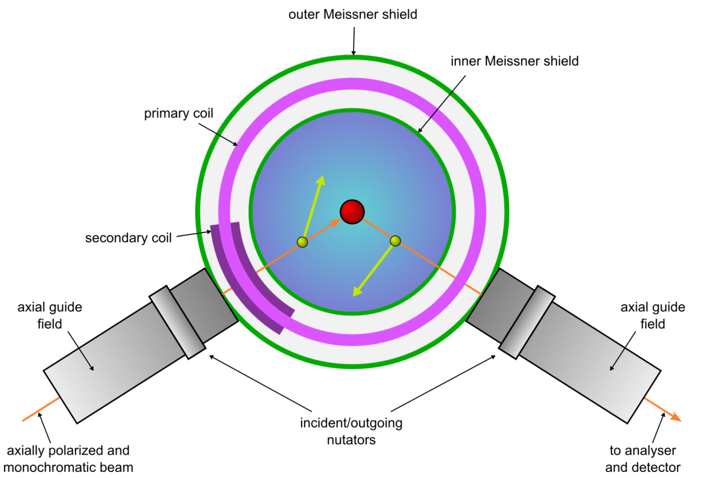


Francis Tasset's polarization lab

The Cryopad

Manipulation of polarised neutron beams

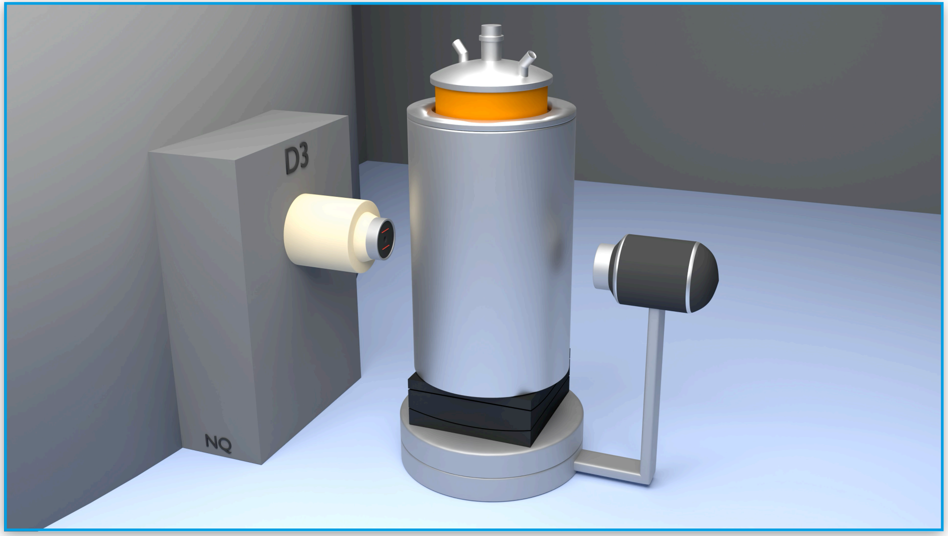
top view



mounted on D3

The Cryopad

Where the magic happens



Local coordination system:

x || **Q**

z vertical

y completes right-handed set

$$\mathbf{M}_{\perp,x} = 0$$

$$\mathbf{P}_i \parallel x \qquad \mathbf{P}_f \parallel y$$

$$P_{f,i} = P_{yx} = \frac{n^+ - n^-}{n^+ + n^-}$$

The Cryopad

The polarization matrix

Final neutron polarization is related to initial one by $\mathbf{P}_f = \mathcal{P}\mathbf{P}_i + \mathbf{P}'$

rotational part ($\mathcal{P}$)

created/annihilated polarisation ($\mathbf{P}'$)

$$\mathbf{P}_{f,i} = \begin{pmatrix} p_{fx}\frac{p_{ix}(N^2-M_\perp^2)-J_{yz}}{I_x} & p_{fx}\frac{-p_{iy}J_{nz}-J_{yz}}{I_y} & p_{fx}\frac{p_{iz}J_{ny}-J_{yz}}{I_z} \\ p_{fy}\frac{p_{ix}J_{nz}+R_{ny}}{I_x} & p_{fy}\frac{p_{iy}(N^2+M_{\perp y}^2-M_{\perp z}^2)+R_{ny}}{I_y} & p_{fy}\frac{p_{iz}R_{yz}+R_{ny}}{I_z} \\ p_{fz}\frac{-p_{ix}J_{ny}+R_{nz}}{I_x} & p_{fz}\frac{p_{iy}R_{yz}+R_{nz}}{I_y} & p_{fz}\frac{p_{iz}(N^2-M_{\perp y}^2+M_{\perp z}^2)+R_{nz}}{I_z} \end{pmatrix}$$

Easily distinguish nuclear, magnetic, chiral and interference terms.

$$I_x = N^2 + M_\perp^2 + p_{ix}J_{yz} \quad R_{yz} = 2\Re(M_{\perp y}M_{\perp z}^*) \quad R_{nz} = 2\Re(NM_{\perp z}^*)$$

$$I_y = N^2 + M_\perp^2 + p_{iy}R_{ny} \quad J_{yz} = 2\Im(M_{\perp y}M_{\perp z}^*) \quad J_{ny} = 2\Im(NM_{\perp y}^*)$$

$$I_z = N^2 + M_\perp^2 + p_{iz}R_{nz}$$

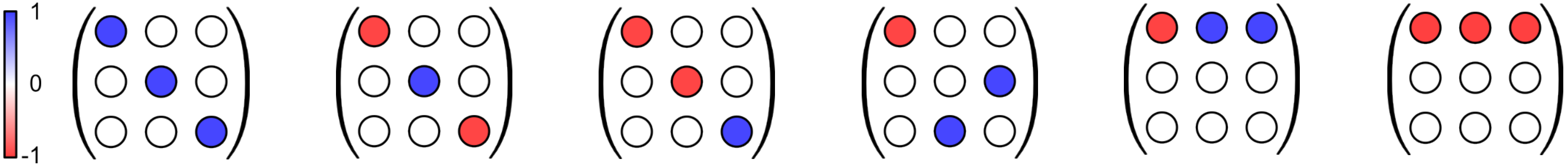
The Cryopad

The polarization matrix

Final neutron polarization is related to initial one by $\mathbf{P}_f = \mathcal{P}\mathbf{P}_i + \mathbf{P}'$

- $\mathcal{P}$: rotational part
- $\mathbf{P}'$: created/annihilated polarisation

$$\mathbf{P}_{f,i} = \begin{pmatrix} p_{fx}\frac{p_{ix}(N^2 - M_\perp^2) - J_{yz}}{I_x} & p_{fx}\frac{-p_{iy}J_{nz} - J_{yz}}{I_y} & p_{fx}\frac{p_{iz}J_{ny} - J_{yz}}{I_z} \\ p_{fy}\frac{p_{ix}J_{nz} + R_{ny}}{I_x} & p_{fy}\frac{p_{iy}(N^2 + M_{\perp y}^2 - M_{\perp z}^2) + R_{ny}}{I_y} & p_{fy}\frac{p_{iz}R_{yz} + R_{ny}}{I_z} \\ p_{fz}\frac{-p_{ix}J_{ny} + R_{nz}}{I_x} & p_{fz}\frac{p_{iy}R_{yz} + R_{nz}}{I_y} & p_{fz}\frac{p_{iz}(N^2 - M_{\perp y}^2 + M_{\perp z}^2) + R_{nz}}{I_z} \end{pmatrix}$$



nuclear | purely magnetic $M_{\perp z} = 0$ | purely magnetic $M_{\perp y} = 0$ | purely magnetic $M_{\perp y} = M_{\perp z}$ | left-handed helix $iM_{\perp y} = M_{\perp z}$ | right-handed helix $iM_{\perp y} = -M_{\perp z}$

The Cryopad

The polarization matrix

Polarization matrix elements are elegantly calculated using the **density matrix formalism**:

$$P_f = \frac{\mathrm{Tr}(\boldsymbol{\sigma} \cdot \mathcal{M} \cdot \boldsymbol{\rho} \cdot \mathcal{M}^\dagger)}{\mathrm{Tr}(\mathcal{M} \cdot \boldsymbol{\rho} \cdot \mathcal{M}^\dagger)}$$

$$\boldsymbol{\rho} = \frac{1}{2}\begin{pmatrix} 1 + P_{i,z} & P_{i,x} - iP_{i,y} \\ P_{i,x} + iP_{i,y} & 1 - P_{i,z} \end{pmatrix}$$

density matrix of the incoming neutron beam

$$\boldsymbol{\sigma} = \begin{pmatrix} P_{f,z} & P_{f,x} - iP_{f,y} \\ P_{f,x} + iP_{f,y} & -P_{f,z} \end{pmatrix}$$

density matrix of the scattered neutron beam

$$\mathcal{M} = \begin{pmatrix} N + M_{\perp,z} & -iM_{\perp,y} \\ iM_{\perp,y} & N - M_{\perp,z} \end{pmatrix}$$

density matrix of the scattering system

Allows for **any** direction of $\mathbf{P}_i$ and $\mathbf{P}_f$

The Cryopad

The polarization matrix

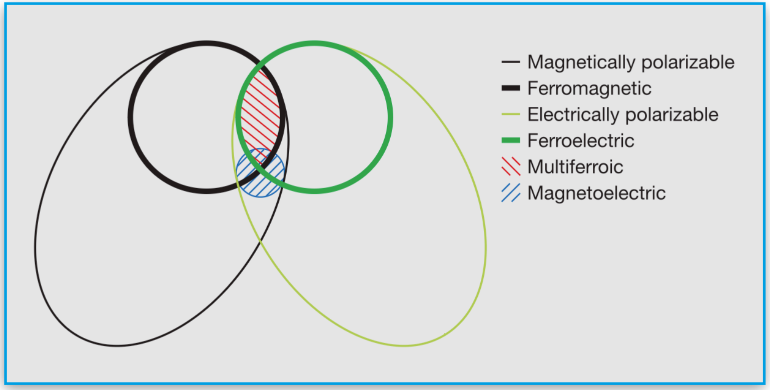
SNP at a glance

Purely nuclear scattering:	$\mathbf{P}_i$ is not affected
Purely magnetic scattering, $Re(M_\perp)$ || $Im(M_\perp)$:	$\mathbf{P}_i$ is rotated by 180° around $\mathbf{M}_\perp$
Chiral magnetic scattering, $Re(M_\perp) \perp Im(M_\perp)$:	$\mathbf{P}_i$ is rotated towards $\pm\mathbf{Q}$ (scattered intensity depends on $\mathbf{P}_i$, polarization created)
Nuclear-magnetic interference (in or out of phase):	$\mathbf{P}_i$ is rotated within the plane containing $\mathbf{P}_i$ and $\mathbf{M}_\perp$ (scattered intensity depends on $\mathbf{P}_i$, polarization created)
Nuclear-magnetic interference (in phase quadrature)	$\mathbf{P}_i$ is rotated towards $\mathbf{P}_i \times \mathbf{M}_\perp$

Examples

Magnetoelectric and multiferroic materials

- Magnetoelectric effect predicted by P. Curie in 1894
- Crosstalk between magnetization and electric polarization
- Multiferroics possess at least two (anti)ferroic orders
- Used as magnetic field sensors and transducers
- Immense potential for non-volatile data storage applications (faster and consumes less energy)
- spin-transfer torque: ~ 10 fJ
- capacitive ME device: ~ 1 aJ (10^4 less)



Eerenstein et al. (2006) *Nature* **442** 759

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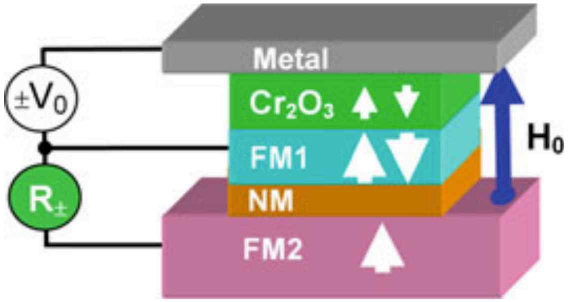
Magnetic Random Access Memory



Kleemann and Binek (2013)
Multiferroic and magnetoelectric materials

Examples

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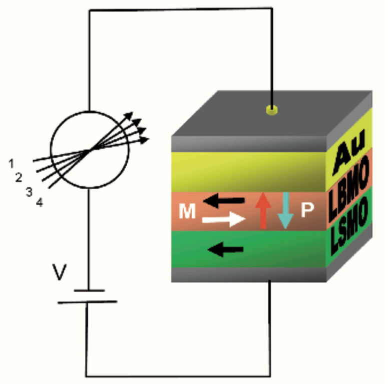
4-bit logic



Kleemann and Borisov (2013)
Multiferroic and magnetoelectric materials for spintronics

Examples

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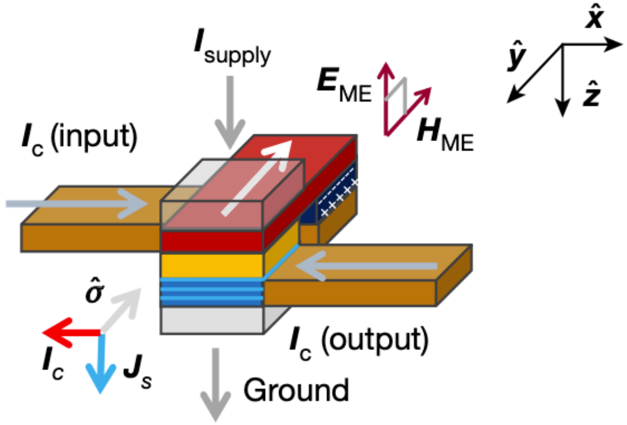
MESO logic device, Intel (MagnetoElectricSpinOrbit)



Manipatruni et al. (2019) *Nature* **565** 35

Examples

Cr_2O_3 - the prototype magnetoelectric

- Predicted by Dzyaloshinskii in 1960
- First observation of linear magnetoelectric effect
- $P_i = \alpha_{ij} H_j$ or $M_i = \alpha_{ji} E_j$
- Corundum structure, space group $R\bar{3}c$, T_N = 307 K
- Magnetic symmetry ($\bar{3}'m'$) breaks spatial and time-inversion

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

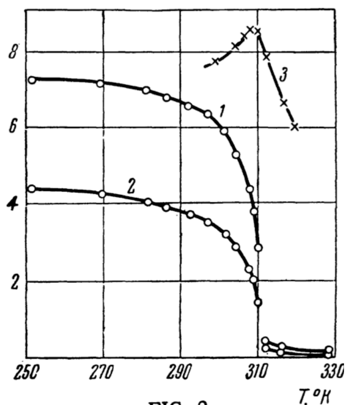
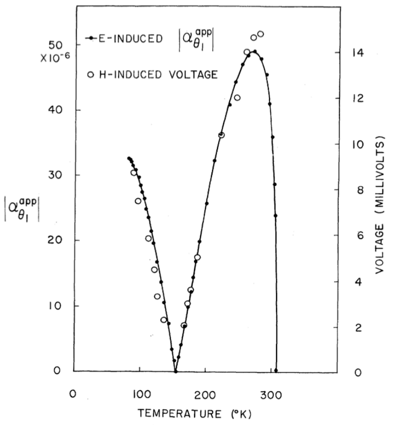


FIG. 2



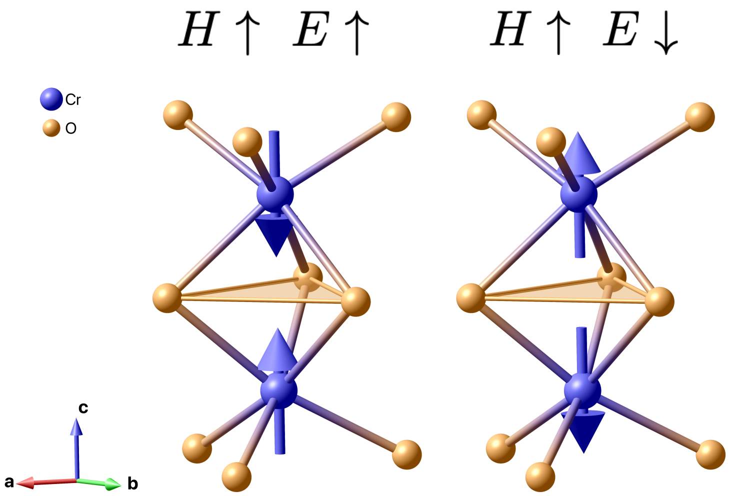
Rado and Folen (1961) *Phys. Rev. Lett.* **7** 310

Examples

Cr_2O_3 - sign of magnetoelectric coefficient

- Problem: presence of 180° magnetic domains
- Annealing under field achieves domain selection
- Domains indistinguishable with unpolarized neutrons
- SNP can tell the difference!
- *b* vertical, (-1 0 2): P_i || *z* rotates towards +/- *x*
- information about sign of magnetoelectric coefficient

Brown, Forsyth and Tasset (1998) J. *Phys.: Condens. Matter.* **10** 663



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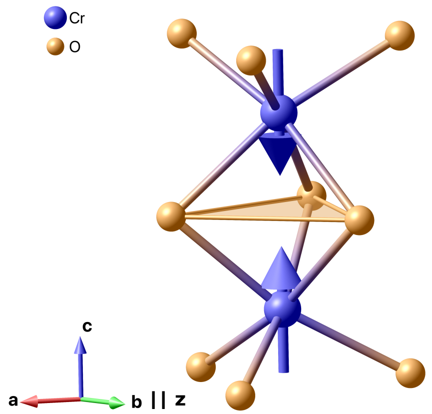
Cr_2O_3 - sign of magnetoelectric coefficient

$$\mathbf{P}_{f,i} = \begin{pmatrix} p_{fx}\frac{\cancel{p_{ix}(N^2 - M_\perp^2) - J_{yz}}}{I_x} & p_{fx}\frac{\cancel{-p_{iy}J_{nz} - J_{yz}}}{I_y} & p_{fx}\frac{p_{iz}J_{ny}\cancel{-J_{yz}}}{I_z} \\ p_{fy}\frac{\cancel{p_{ix}J_{nz} + R_{ny}}}{I_x} & p_{fy}\frac{p_{iy}(N^2 + M_{\perp y}^2 \cancel{- M_{\perp z}^2}) \cancel{+ R_{ny}}}{I_y} & p_{fy}\frac{\cancel{p_{iz}R_{yz} + R_{ny}}}{I_z} \\ p_{fz}\frac{-p_{ix}J_{ny}\cancel{+R_{nz}}}{I_x} & p_{fz}\frac{\cancel{p_{iy}R_{yz} + R_{nz}}}{I_y} & p_{fz}\frac{\cancel{p_{iz}(N^2 - M_{\perp y}^2 + M_{\perp z}^2) + R_{nz}}}{I_z} \end{pmatrix}$$

- *b* || *z*, moments along *c* —> no *z* component
- (-1 0 2) at room temperature: $N \approx M_{\perp y}$
- $\Im(N) = 0 \quad \Re(M_\perp) = 0$ —> No real interference terms

$$R_{ny} = 2\Re(NM_{\perp y}^*) \quad J_{ny} = 2\Im(NM_{\perp y}^*)$$



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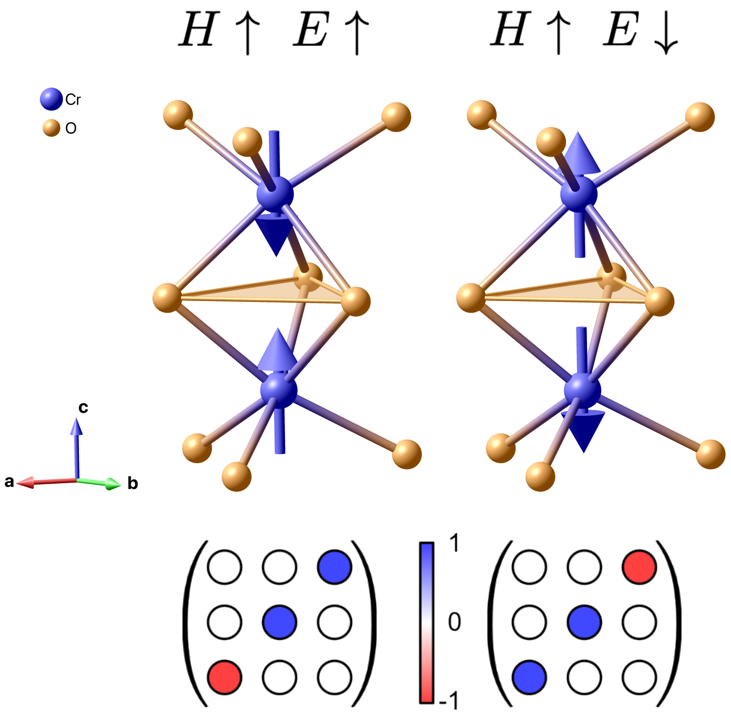
  (reported sign was wrong… reanalyzed and rectified)

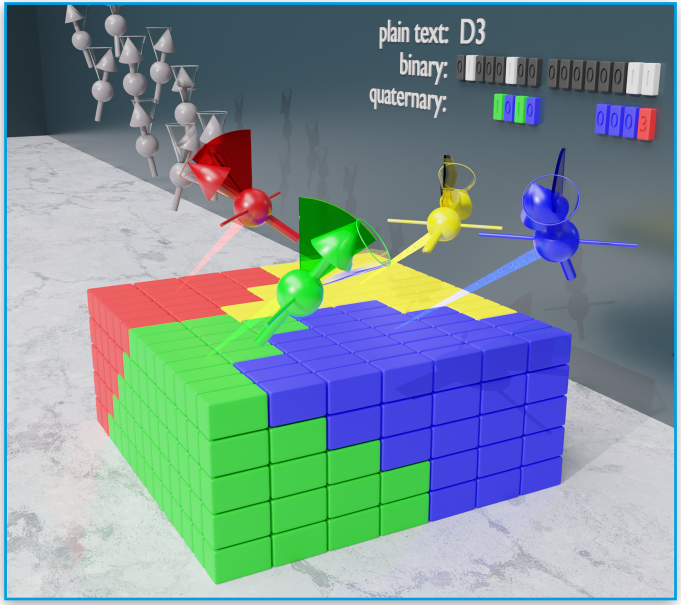
  Bousquet et al. (2024) *J. Phys.: Condens. Matter.* **36** 155701



Examples

$LiNi_{0.8}Fe_{0.2}PO_4$ - bulk AF quaternary memory

- Slowing down of Moore's law requires paradigm change
- Literature: quaternary memory only with ferro component
- $LiNi_{0.8}Fe_{0.2}PO_4$ reveals four magnetic domains
- **E** x **H** field cooling can achieve domain selection
- SNP can tell the difference between the domains



Qureshi et al. (2026) *Nat. Commun.*

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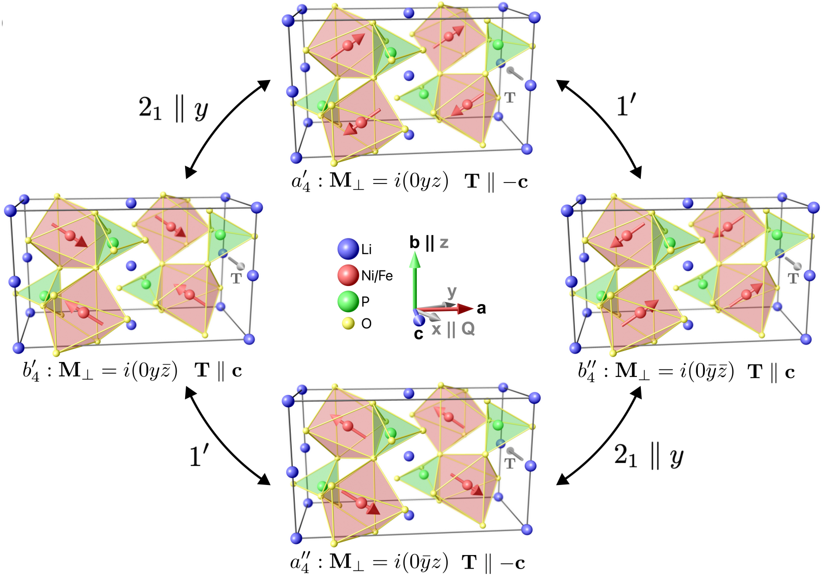
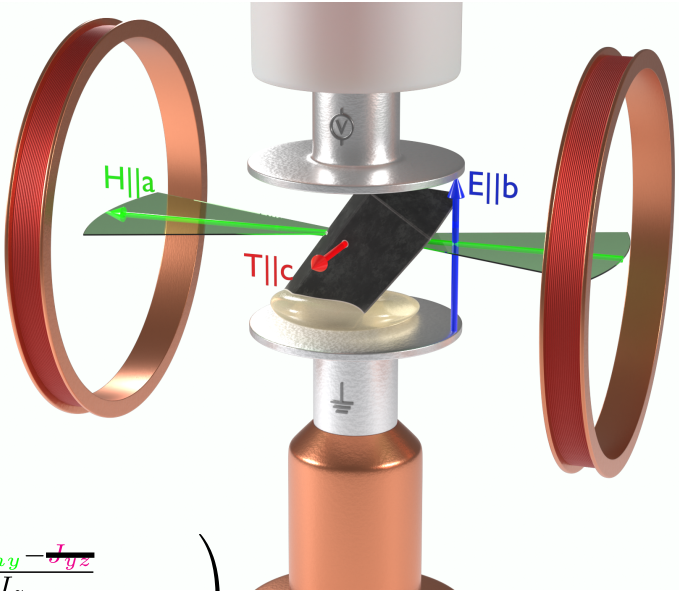


Qureshi et al. (2026) *Nat. Commun.*

Examples

$LiNi_{0.8}Fe_{0.2}PO_4$ - bulk AF quaternary memory

- *b* || *z*, **E** x **H** in *a*-*c* plane
- (101) has nuclear and magnetic contribution
- Antiparallel spins related by 1′ —> $\Re(M) = 0$
- *b* component —> $J_{nz} = 2\Im(NM^*_{\perp z})$
- *a* component —> $J_{ny} = 2\Im(NM^*_{\perp y})$



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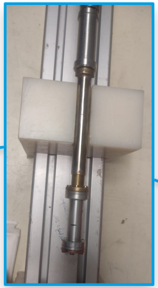
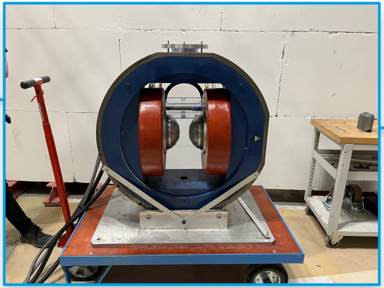
Examples

$LiNi_{0.8}Fe_{0.2}PO_4$ - bulk AF quaternary memory

single crystal glued on capacitor plate

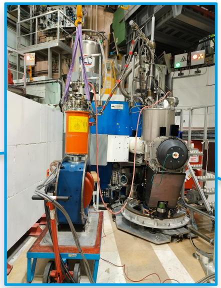
closed electric-field stick

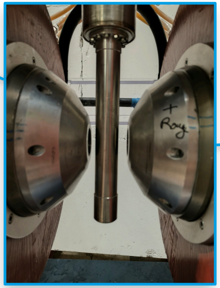




D3’s electromagnet

D3’s zone





thin-tail inside magnet

cryostat inside Cryopad

Examples

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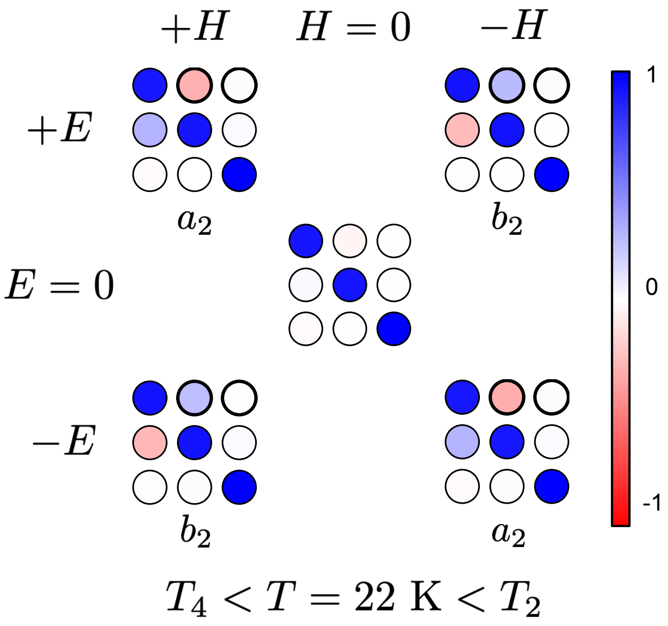
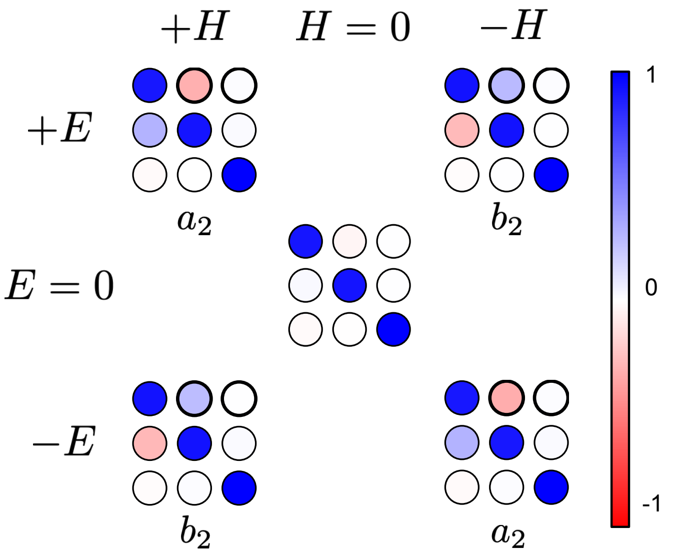


Majority domain: up to 91(2)%

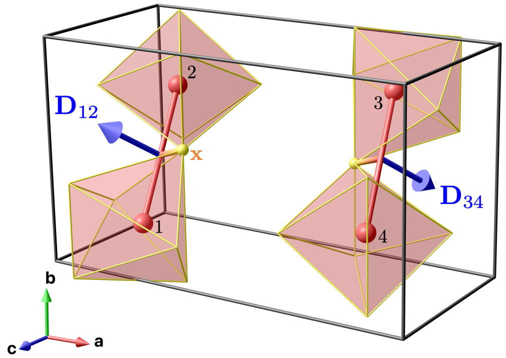
Examples

$LiNi_{0.8}Fe_{0.2}PO_4$ - bulk AF quaternary memory



$T_4 < T = 22\ \mathrm{K} < T_2$ $\quad$ $T = 2\ \mathrm{K} < T_4$

- **E** x **H**$_a$ selects toroidal domain
- **H**$_c$ selects rotation domain (DM)



$$\mathcal{H}_{1b}^{\mathrm{DM}} = -D_{12}^{b}(S_1^a S_2^c - S_1^c S_2^a - S_3^a S_4^c + S_3^c S_4^a)$$

Majority domain: up to 91(2)% $\quad$ up to 66(2)%

Examples

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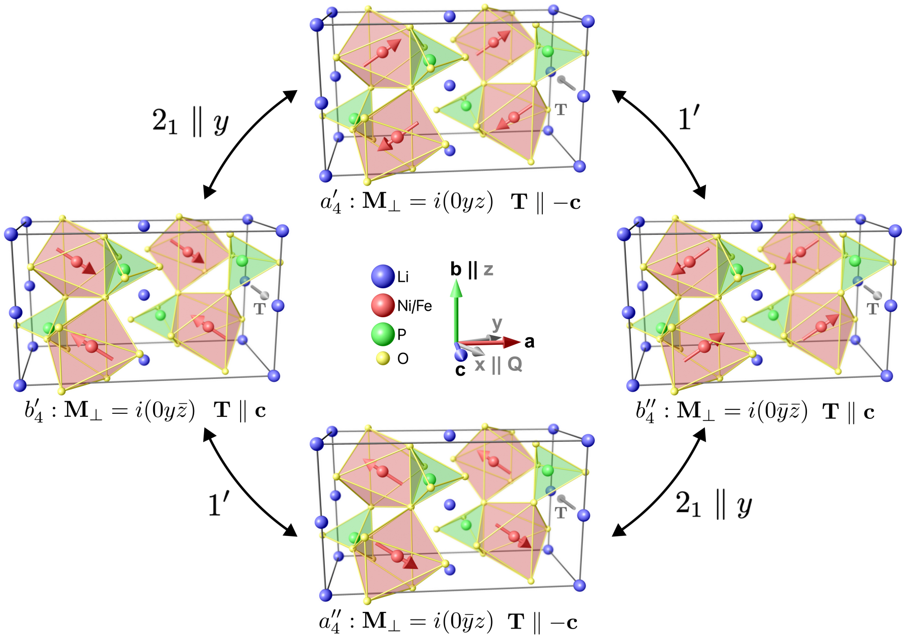

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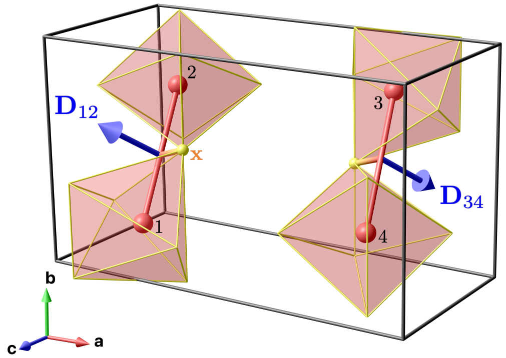

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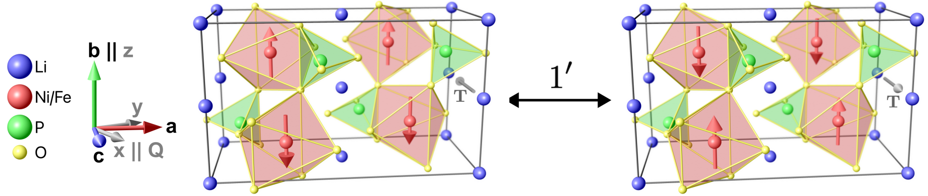
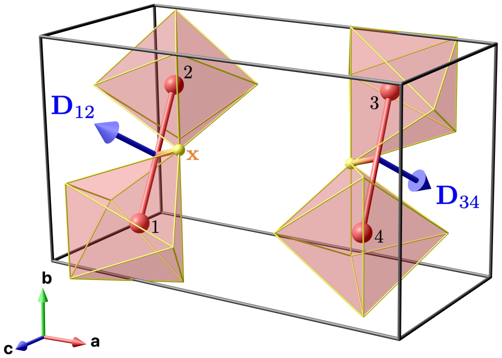


$a_2 : \mathbf{M}_\perp = i(00z) \quad \mathbf{T} \parallel -\mathbf{c}$

$b_2 : \mathbf{M}_\perp = i(00\bar{z}) \quad \mathbf{T} \parallel \mathbf{c}$

H-only cooling: 33(2)% : 67(2)%

- **E** x $\mathbf{H}_a$ selects toroidal domain
- $\mathbf{H}_c$ selects rotation domain (DM)



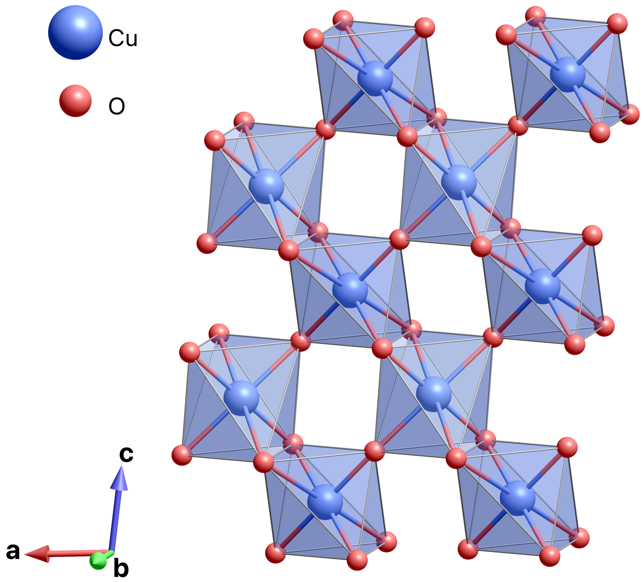
$$\mathcal{H}^{\mathrm{DM}}_{1b} = -D^b_{12}(S^a_1 S^c_2 - S^c_1 S^a_2 - S^a_3 S^c_4 + S^c_3 S^a_4)$$

$$\mathcal{H}^{\mathrm{DM}}_{1c} = D^c_{12}(S^a_1 S^b_2 - S^b_1 S^a_2 - S^a_3 S^b_4 + S^b_3 S^a_4)$$

Examples

CuO - a high-T multiferroic

- so far the only known binary multiferroic compound
- monoclinic space group *C2/c*
- spiral magnetic ordering induces electric polarisation at 230 K
- DFT + MC: applying pressure drives multiferroic state towards RT
- recently confirmed by neutron diffraction under pressure



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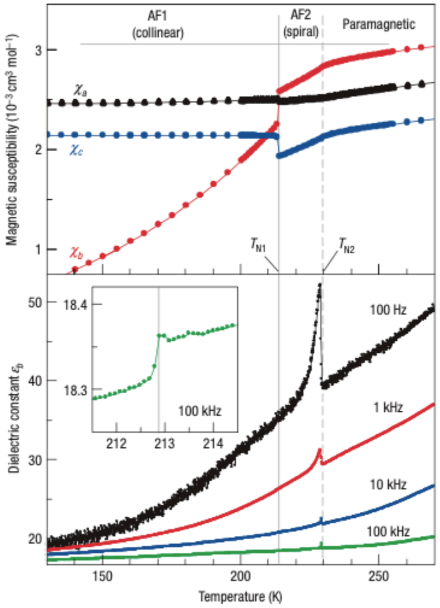

Kimura et al. (2008) *Nat. Mater.* **7** 291

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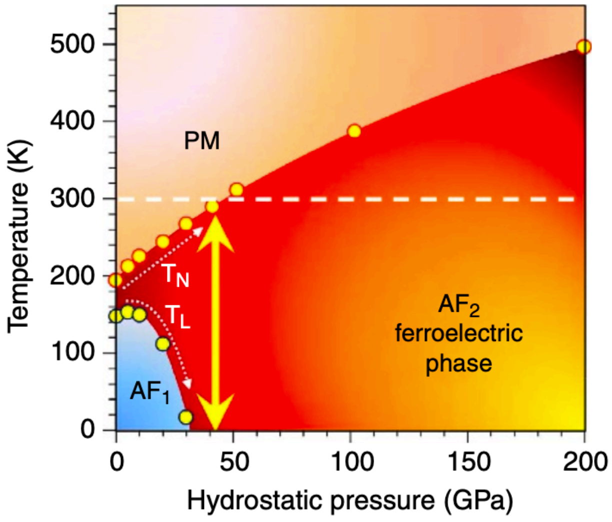

Rocquefelte et al. (2013) *Nat. Mater.* **4** 2511

Examples

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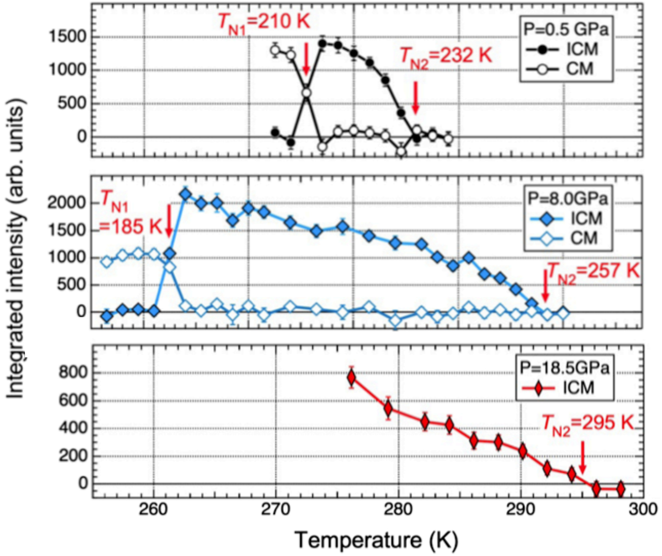
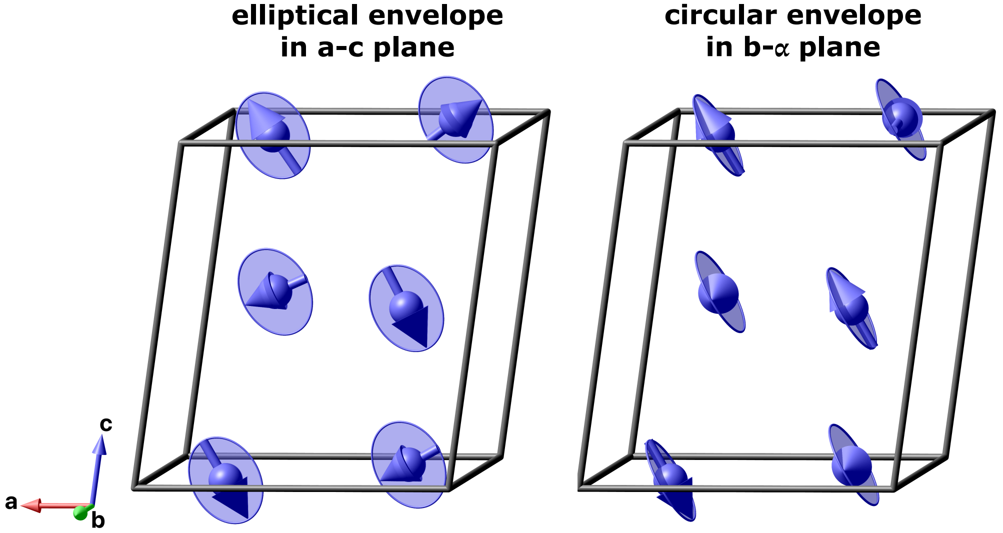


Terada et al. (2022) *Phys. Rev. Lett.* **129** 217601

Examples

CuO - a high-T multiferroic

- CuO is a building block of high-T_C cuprates
- magnetism in CuO —> hole pairing mechanism?
- unpolarized neutrons: model ambiguity



$\chi^2 = 76 \quad R_\mathrm{w} = 17.0\%$ $\chi^2 = 64 \quad R_\mathrm{w} = 15.5\%$

Forsyth, Brown et al. (1988) *J. Phys. C: Solid State Phys.* **21** 2917

Brown, …, Forsyth, … and Tasset (1991) *J. Phys.: Condens. Matter.* **3** 4281

Examples

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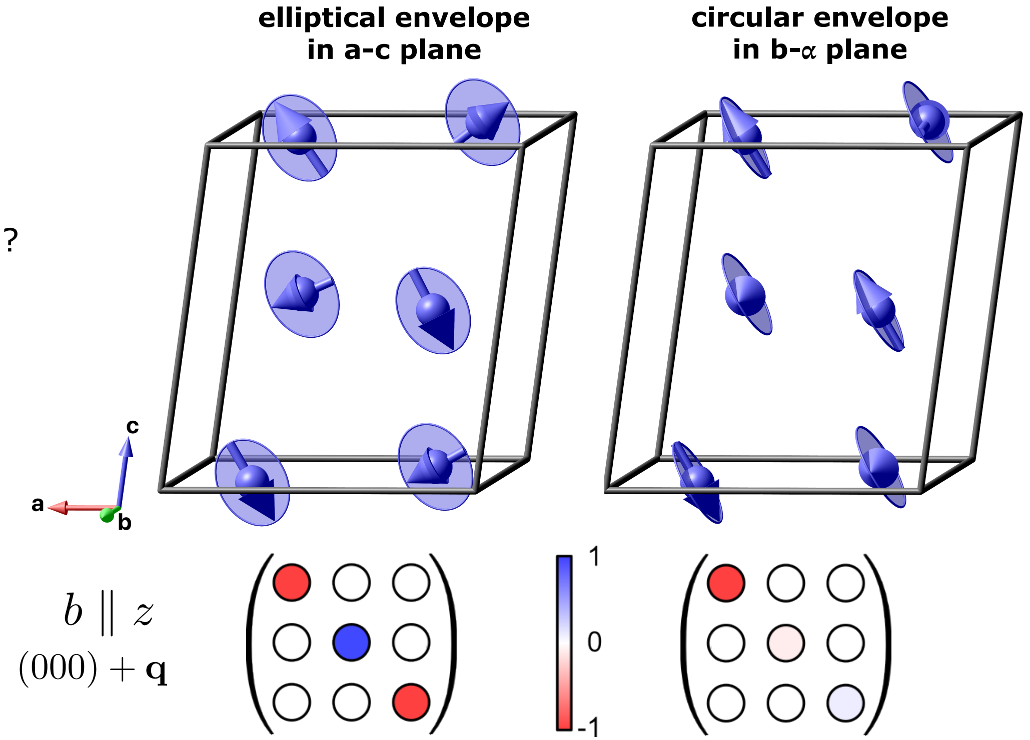
- correct model determined by SNP
- superior to unpolarized neutrons especially for weak reflections and non-collinear structures



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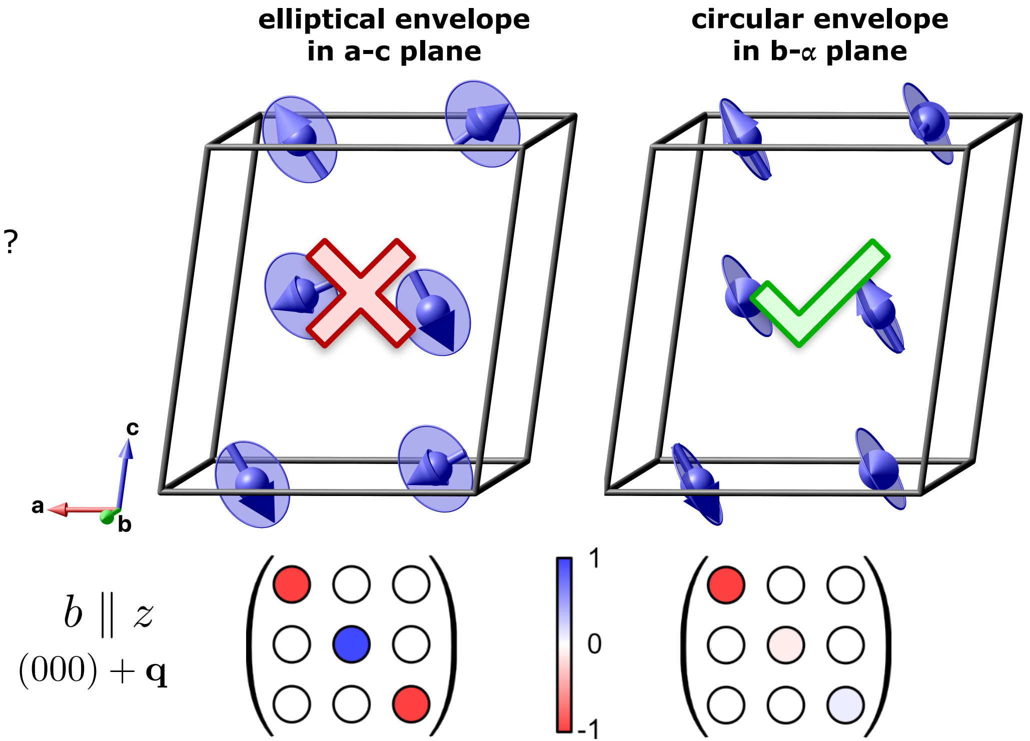
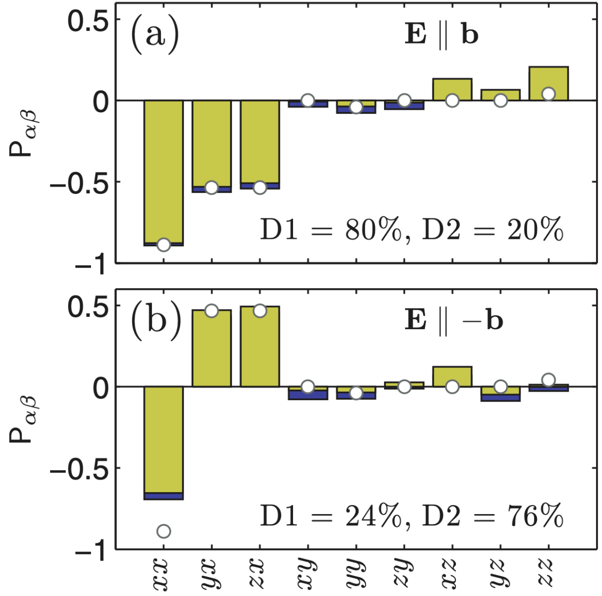


Examples

CuO - switching behavior

- strong coupling between magnetic order and electric polarization
- electric field control of chiral domains
- magnetic chirality is inaccessible with unpolarized neutrons
- SNP can tell the difference



Babkevich et al. (2012) *Phys. Rev. B* **85** 134428

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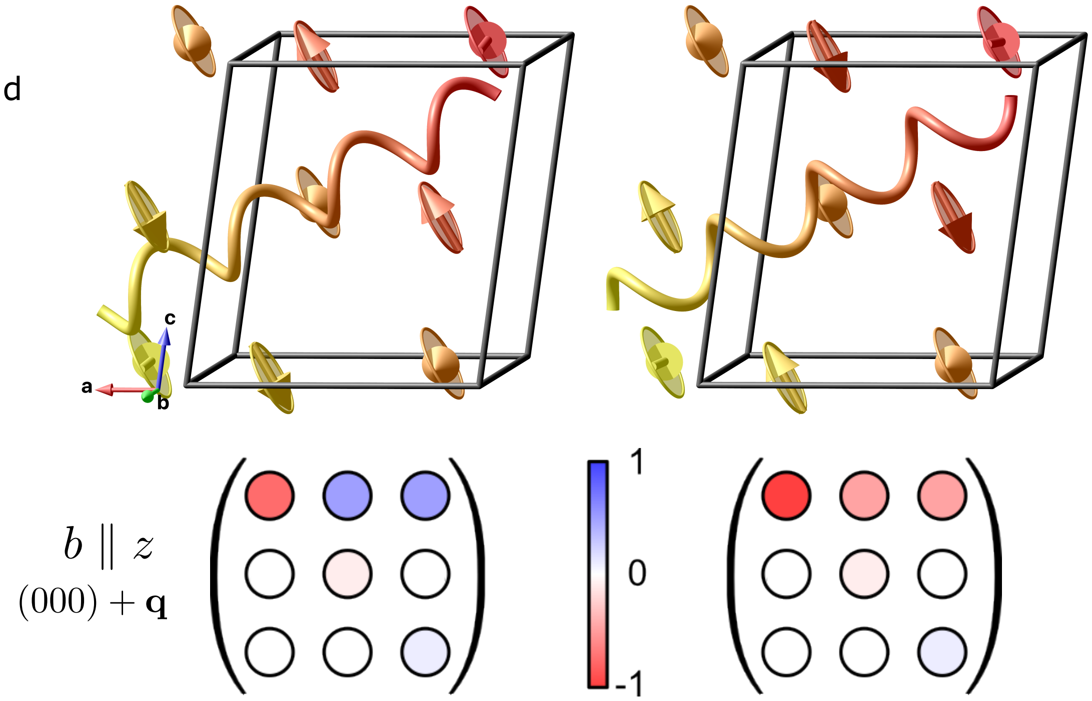
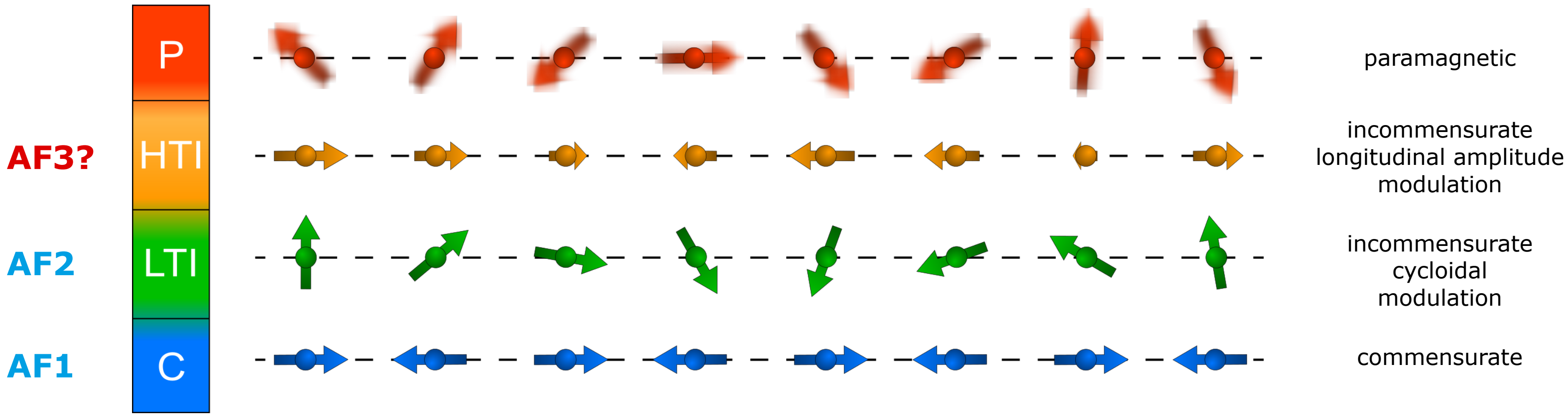


Examples

CuO - search for the AF3 phase

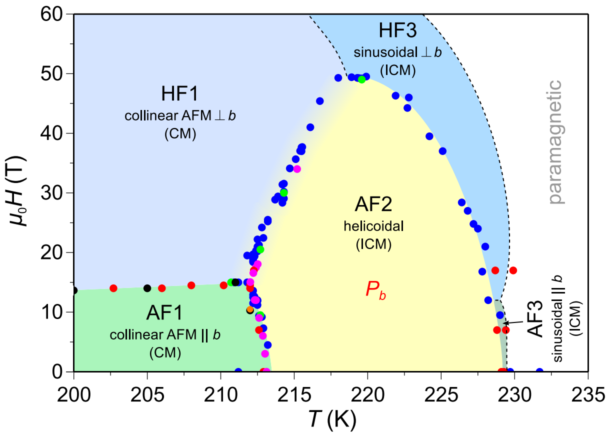
usual sequence of magnetic phases in multiferroics (e.g. $TbMnO_3$, $Ni_3V_2O_8$):



Examples

CuO - search for the AF3 phase

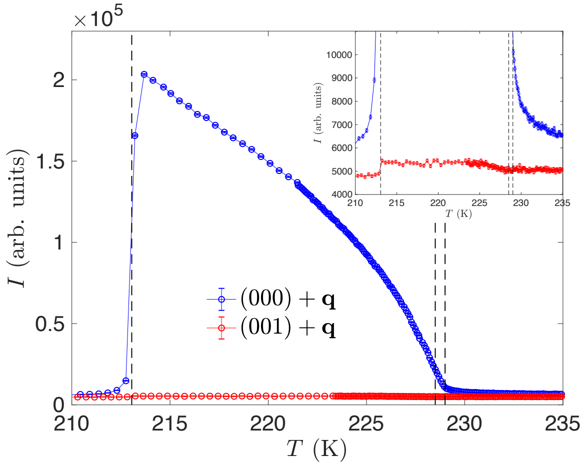
macroscopic methods



0.5 K stability range!

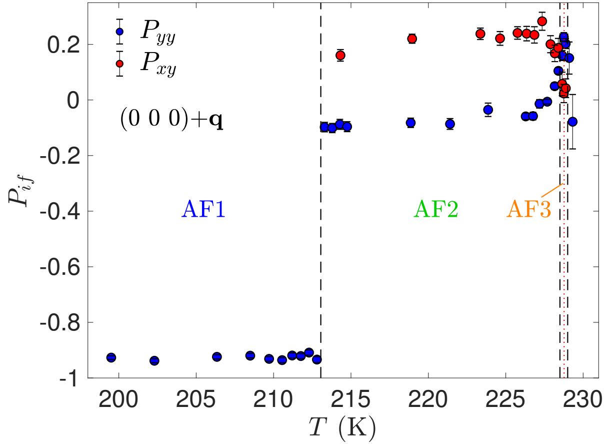
Wang, Qureshi et al. (2016) *Nat. Commun.* **7** 10295

unpolarized neutrons



No anomalies in *I*(*T*)

SNP on P_{yy} and P_{xy} elements



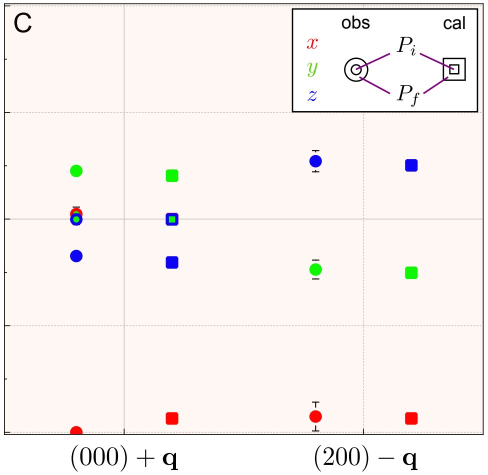
Microscopic proof of AF3!

Qureshi et al. (2020) *Sci. Adv.* **6** eaay7661

Examples

CuO - search for the AF3 phase

observed vs. calculated

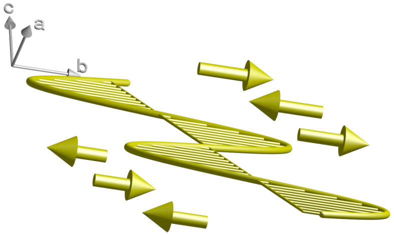


analyzed with

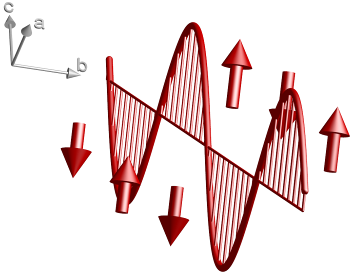
MAG2POL
Navid Qureshi, ILL 2016 - 2023

Qureshi (2019) *J. Appl. Cryst.* **52** 175

phase coexistence



SDW μ || b



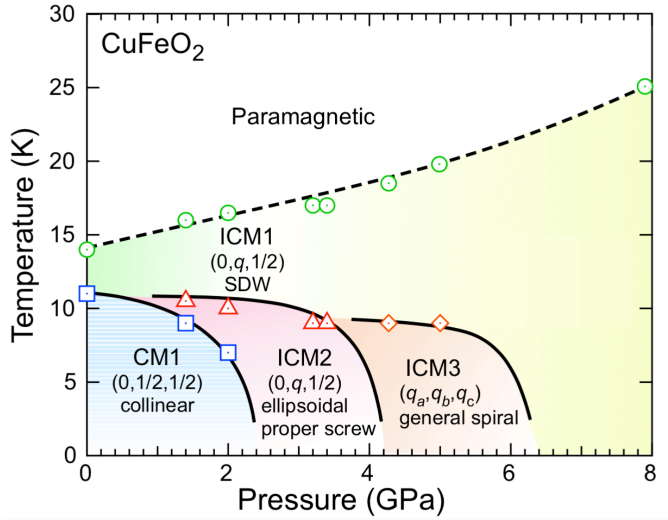
SDW μ || α

Examples

$CuFeO_2$ - pressure-induced multiferroicity

Complex p-T phase diagram

including collinear spin-density waves and non-collinear spiral structures

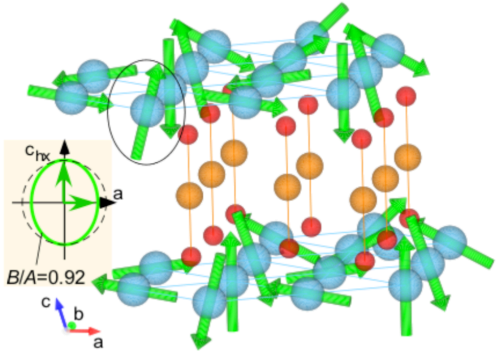


Lack of information

Limited number of observations (integrated intensities) did not allow to reveal:

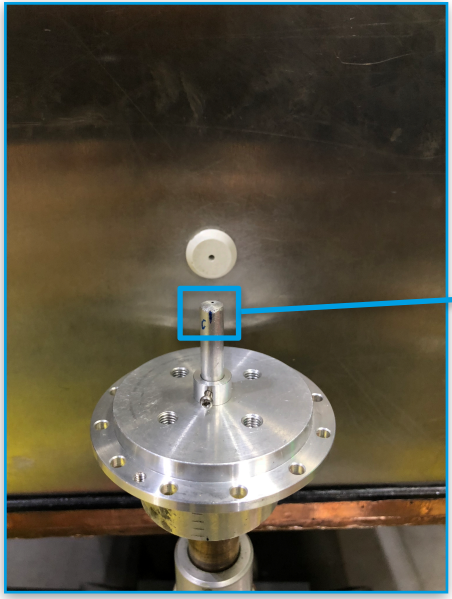
Terada et al. (2014) *Phys. Rev. B* **83** 220403(R)

- stability region of ICM1
- ellipsoidicity ratio of ICM2
- magnetic structure of ICM3



Examples

$CuFeO_2$ - pressure-induced multiferroicity



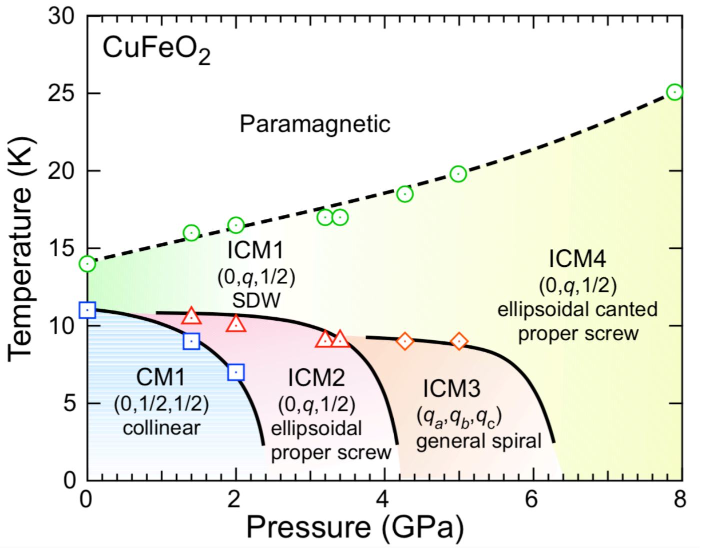
0.5 x 0.5 x 0.2 mm^3

non-magnetic pressure cell (up to 10 GPa)

Examples

$CuFeO_2$ - pressure-induced multiferroicity

- SNP under pressure: It's possible!!!
- Determined 5 complex magnetic structures at 3 different pressures in 7 days with a 0.05 mm³ sample
- SNP is an excellent approach to study novel pressure-induced phenomena associated with complex magnetic order



Terada, Qureshi et al. (2018) *Nat. Commun.* **9** 4368

Conclusions

SNP - strengths and limitations

SNP is a very powerful technique for the determination of complex magnetic structures and magnetic domain populations, but there are limitations…

- SNP yields the precise direction of the magnetic interaction vector
- A few reflections are sufficient to determine the magnetic structure uniquely
- Scattering from a multi-domain state —> depolarization occurs (strength: distinguish between depolarization and rotation away from polarization/analysis axis)
- Ferro- and ferrimagnetic samples depolarize the beam
- If purely magnetic scattering —> no information about the magnetic moment amplitude or magnetic moment configuration (e.g. *A*, *G* or *C* modes)
- Nuclear and magnetic scattering —> precise knowledge of the nuclear structure is beneficial

Conclusions

SNP and its application to condensed matter physics

- SNP is a powerful technique without alternatives for certain scientific challenges
- Magnetic domain sensitivity makes it the ideal probe for switching behavior of smart materials
- Weak signals (small moments, small samples, thin films) are no problem as long as different from background
- Importance for future spintronic candidates

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