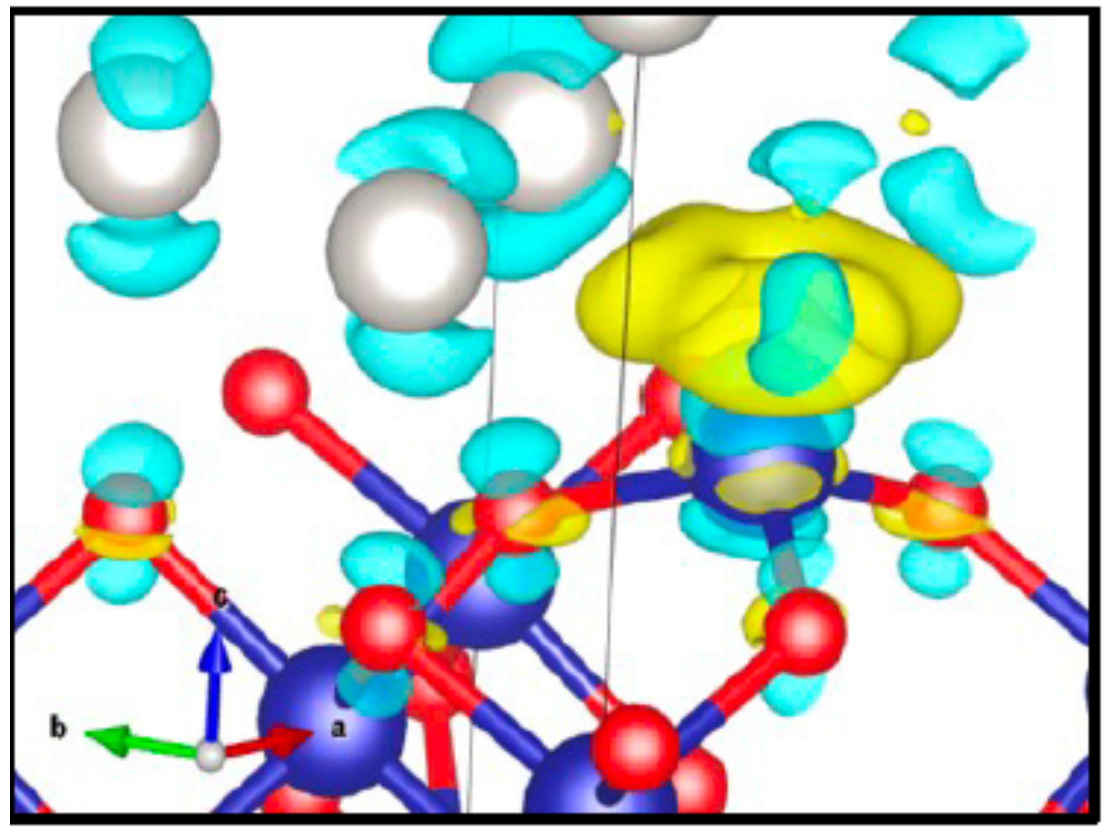
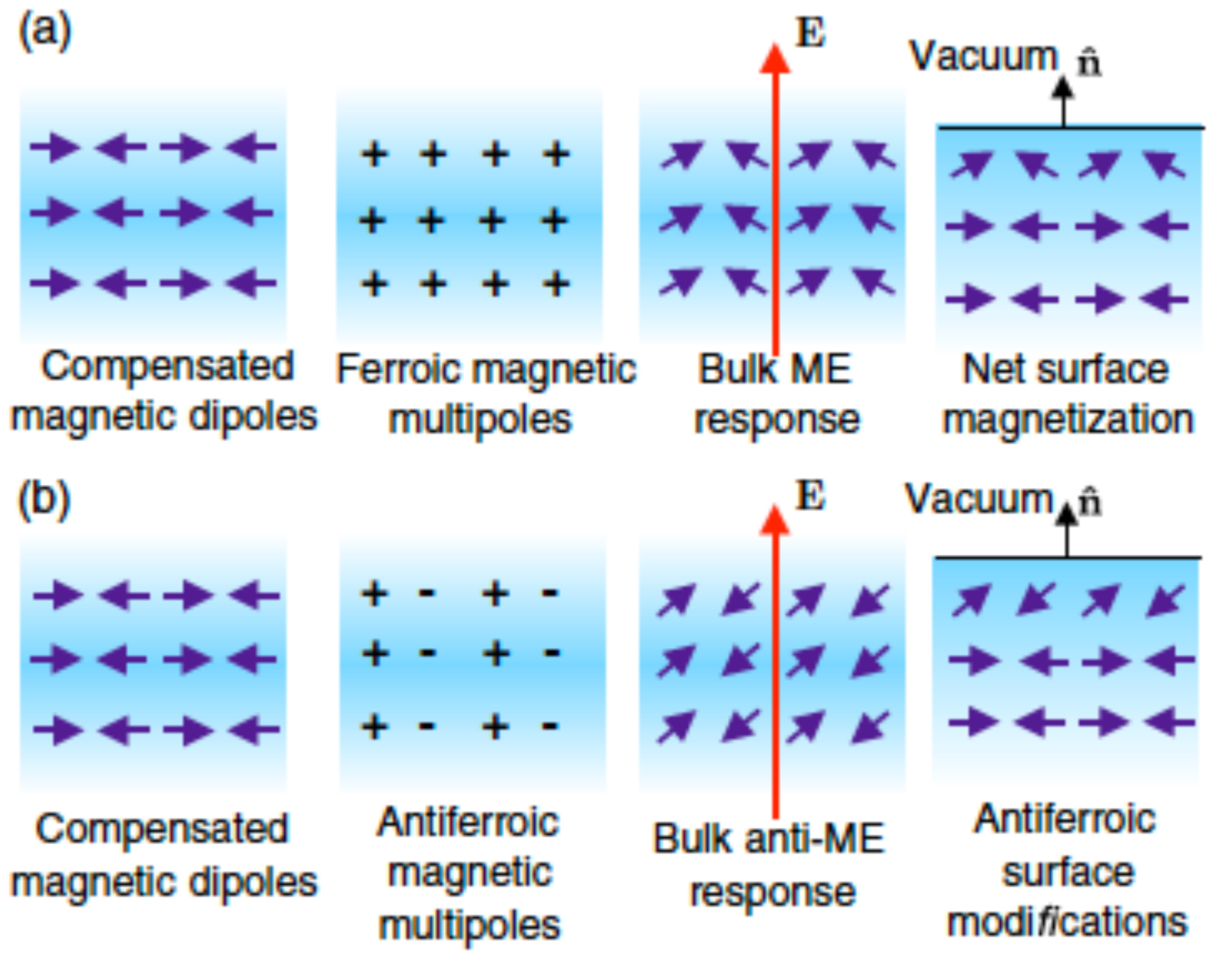
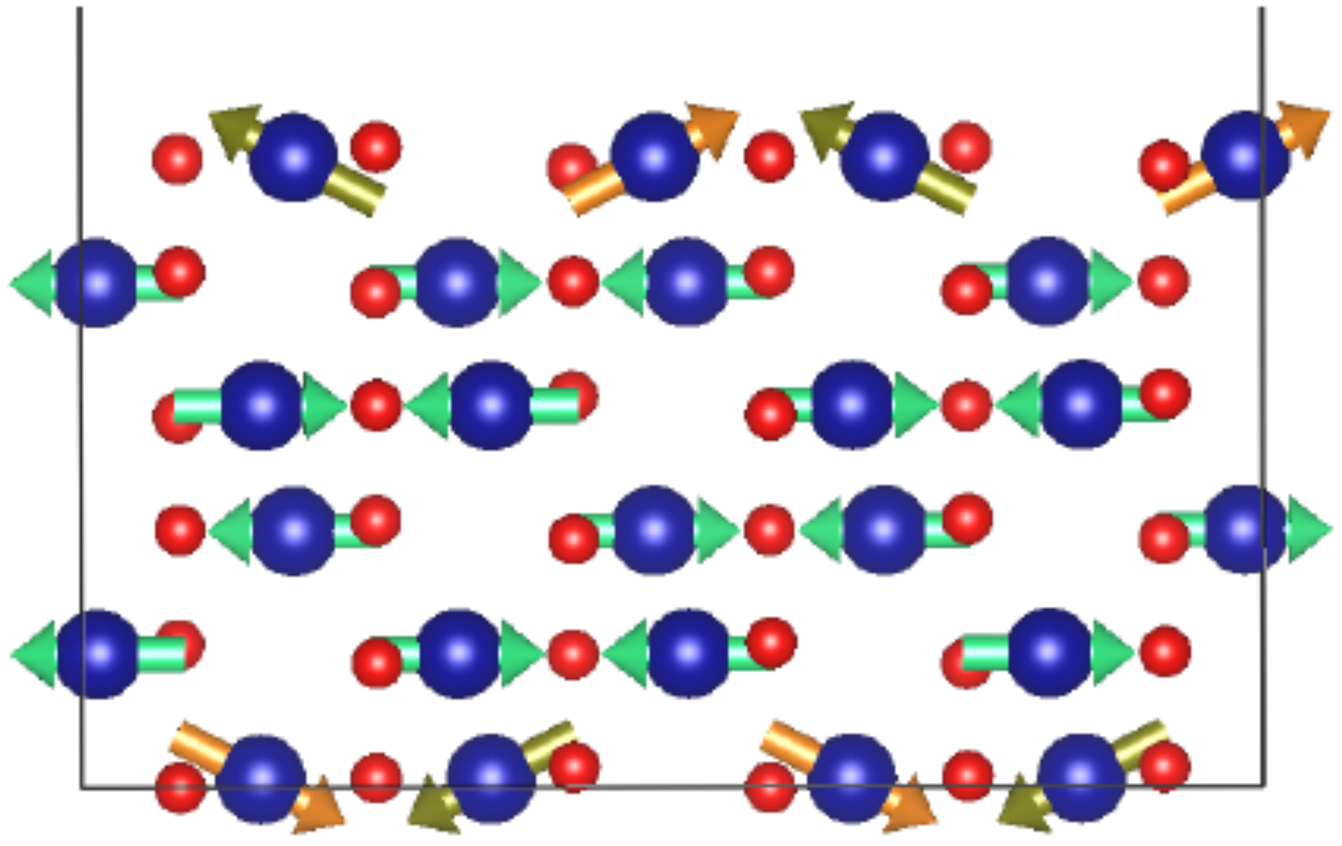
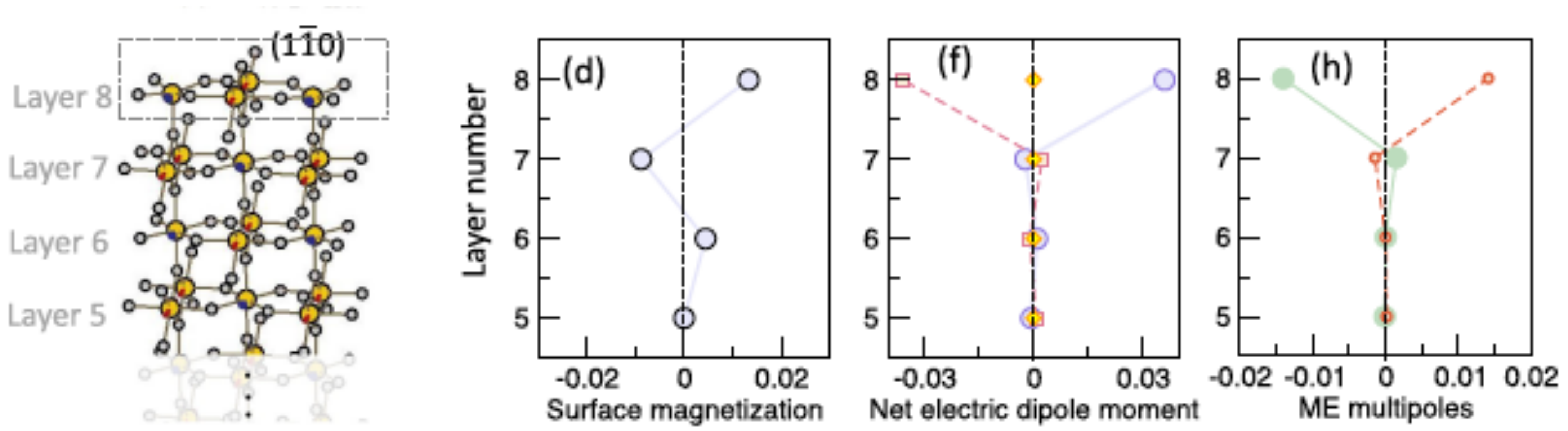
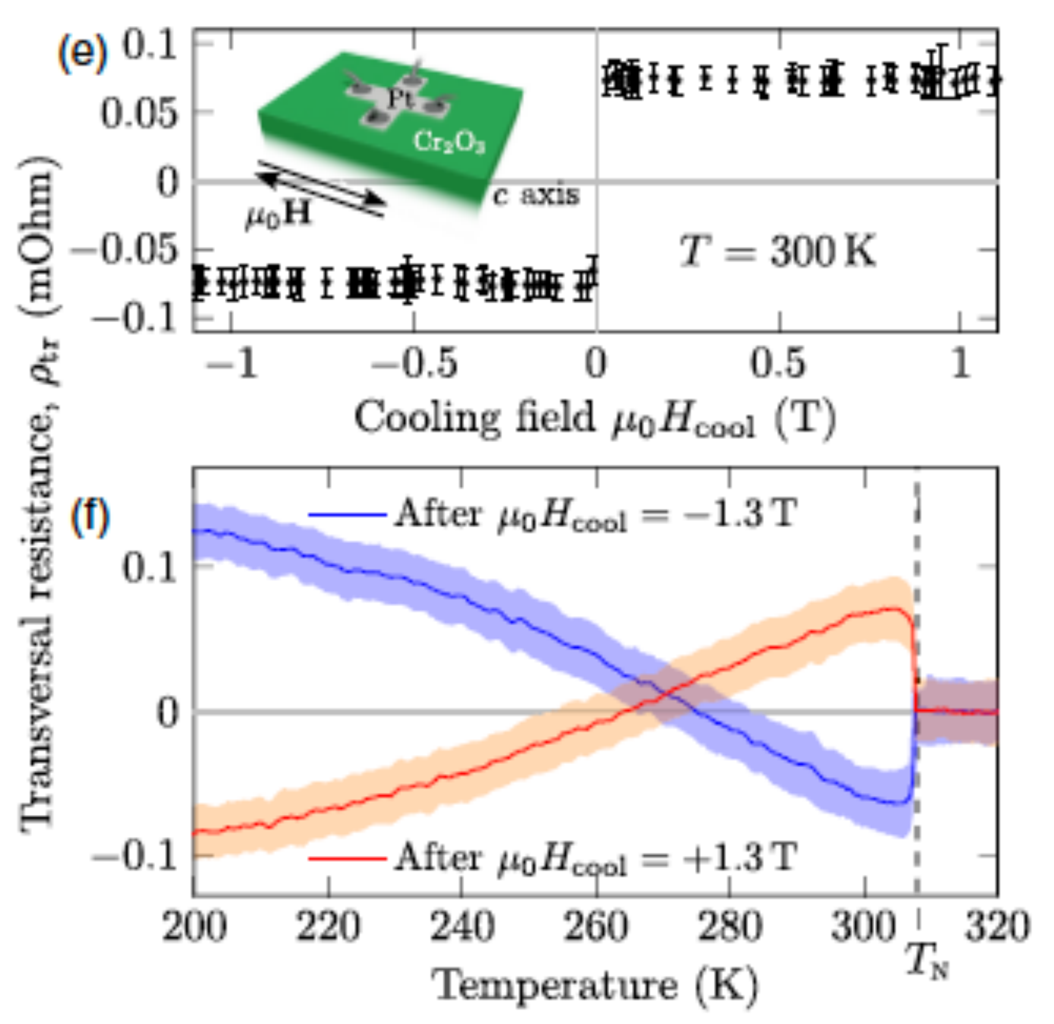






Leveraging the intrinsic electric fields at surfaces for spintronics applications





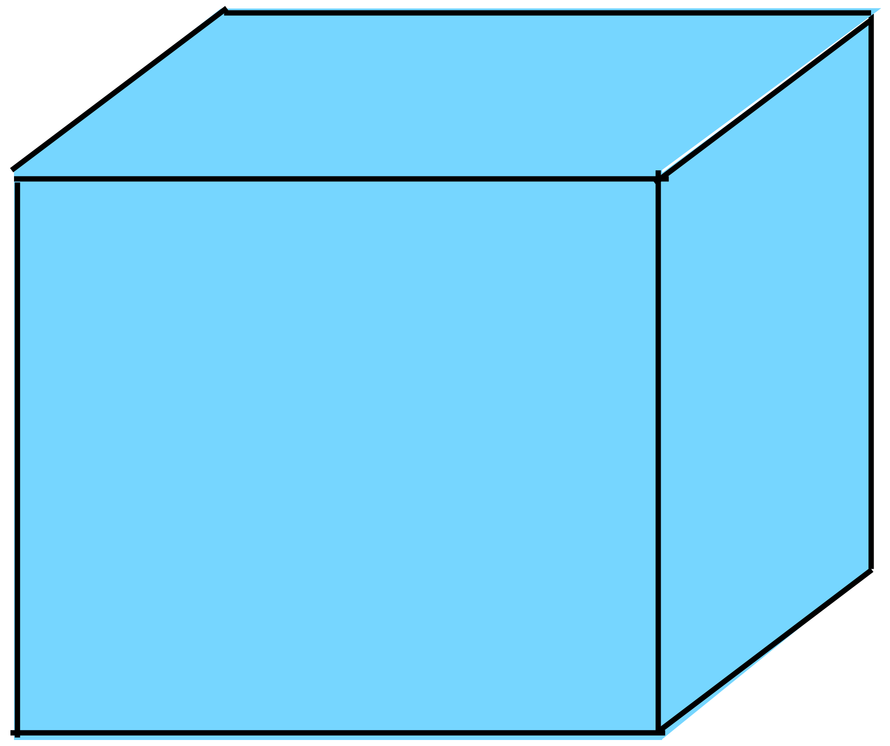
Sophie Frances Liss Weber

Chalmers University of Technology, Göteborg, Sweden

YRLGW: Magnetism in van der Waals materials: current challenges and future directions

08.07.2025

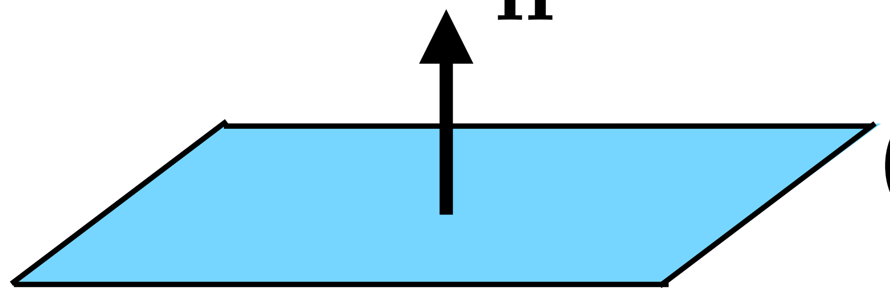
Surfaces of quantum materials are a platform for emergent, functional phenomena



Bulk magnetic point group (MPG)

$$\mathbf{g} = (\mathbf{R})$$

$\hat{\mathbf{n}}$



$(hkl) \perp \hat{\mathbf{n}}$

Surface magnetic point group

$\mathbf{g}^{\hat{\mathbf{n}}} = \left(\mathbf{R}^{\hat{\mathbf{n}}}\right) < \mathbf{g}$ for which $\mathbf{R}^{\hat{\mathbf{n}}}\hat{\mathbf{n}} = \hat{\mathbf{n}}$

$\hat{\mathbf{n}}$ $\quad$ $\mathbf{R}^{\hat{\mathbf{n}}}\hat{\mathbf{n}}$

- The magnetic space group characterizing a surface is a *lower-symmetry subgroup* of the infinitely periodic bulk
- Thus, low-symmetry properties (ferromagnetism, ferroelectricity…) more readily emerge at surfaces and interfaces

Outline

I. An unconventional bulk-boundary correspondence

(a) Bulk materials subjected to electric fields versus surfaces in equilibrium

(b) Specific case: surface properties of bulk magnetoelectrics (MEs)

II. Bulk ME responses as predictors of surface magnetic properties

(a) Surface ferromagnetism in antiferromagnets[1,2]

(b) Surface antiferromagnetism (rotation or magnitude changes of bulk Néel vector)[3]

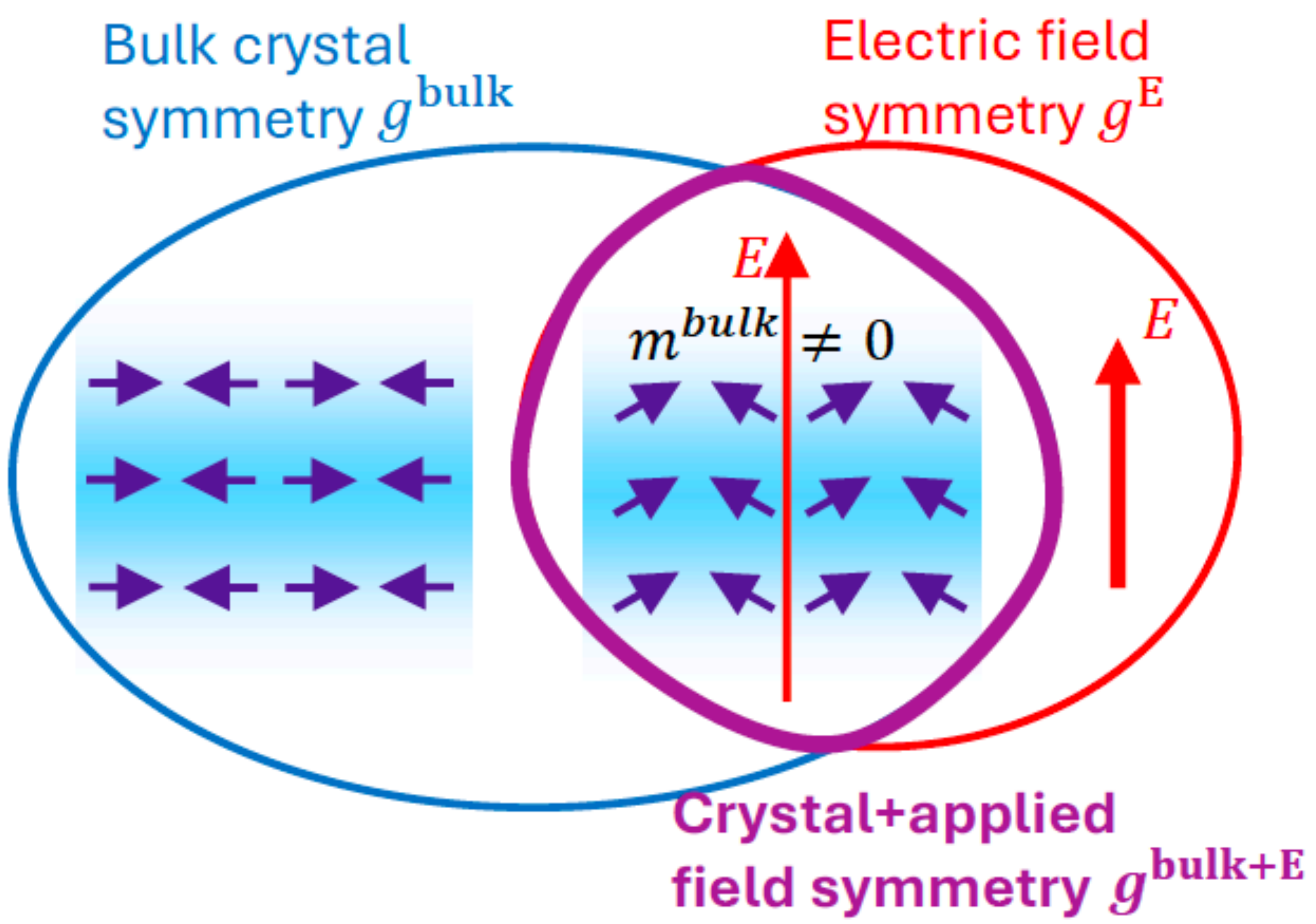
(c) Surface linear ME effects in bulk higher-order magnetoelectrics[4]

III. Outlook

(a) Relevance for van der Waals materials

1. SFW*, A. Urru* et al., *Phys. Rev. X* **14**, 021033 (2024)
2. O. V. Pylypovskyi*, SFW* et al., *Phys. Rev. Lett.* **132**, 226702 (2024)
3. SFW et al., *Phys. Rev. X* **15**, 021094 (2025)
4. S. Bhowal…SFW et al., *Phys. Rev. Lett.* **134**, 146703 (2025)

Bulk materials under applied electric fields versus surfaces in equilibrium

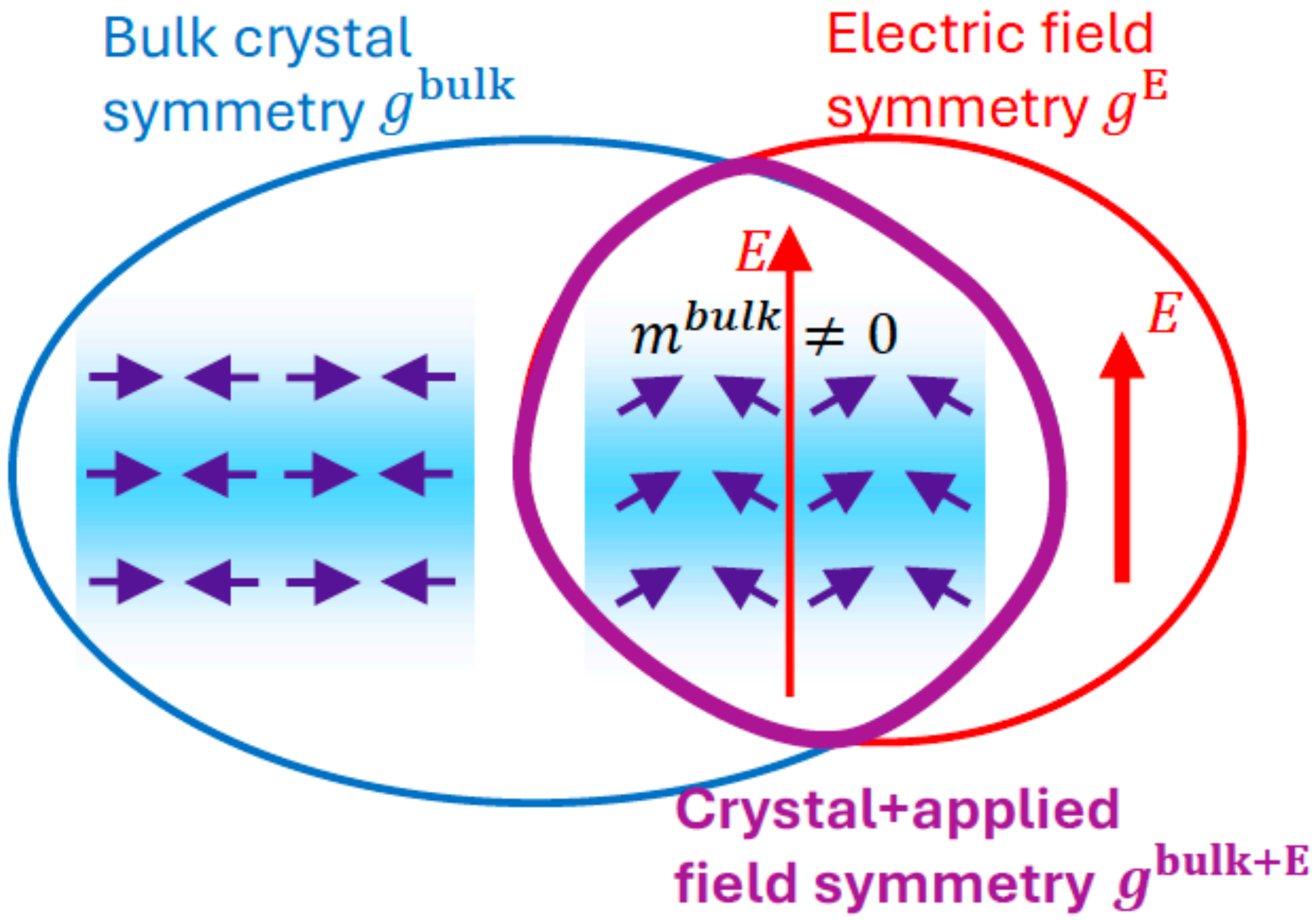


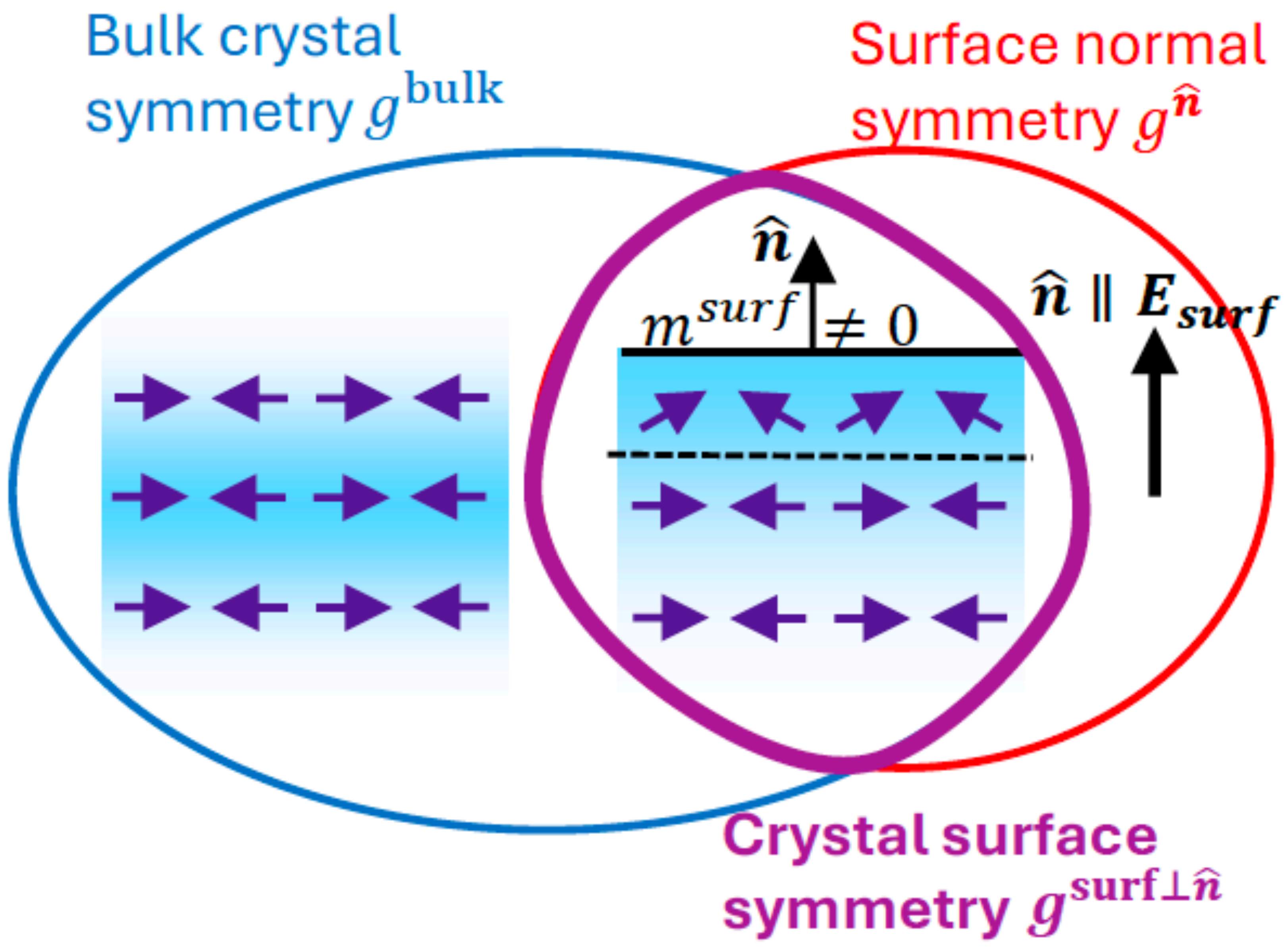
g^{bulk} : *set of operations leaving crystal's atomic positions and their spins invariant*

$g^{\mathbf{E}}$: *set of operations leaving electric field invariant (rotations about* $\mathbf{E}$ *axis and mirror planes containing* $\mathbf{E}$ *)*

- The point group of a bulk crystal subjected to an external electric field is the intersection of their isolated point groups
- The operations in $g^{\mathrm{bulk+\mathbf{E}}}$ determine the properties (magnetization, polarization, strain…) that the electric field can induce

Bulk materials under applied electric fields versus surfaces in equilibrium



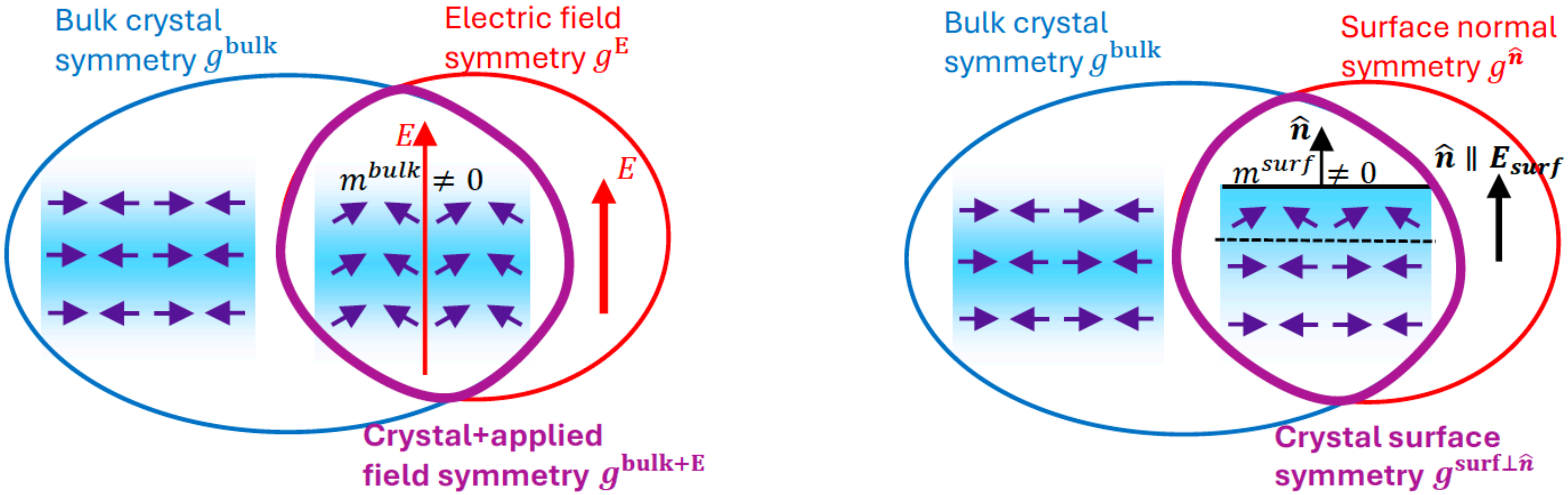


g^{bulk} : *set of operations leaving crystal's atomic positions and their spins invariant*

$g^{\hat{\mathbf{n}}}$: *set of operations leaving surface normal invariant (rotations about* $\hat{\mathbf{n}}$ *axis and mirror planes containing* $\hat{\mathbf{n}}$*)*

- $g^{\text{bulk}+\mathbf{E}}$ is identical to the point group describing a surface whose normal is parallel to $\mathbf{E}$: $g^{\text{bulk}+\mathbf{E}} = g^{\text{surf}\perp\hat{\mathbf{n}}\|\mathbf{E}}$

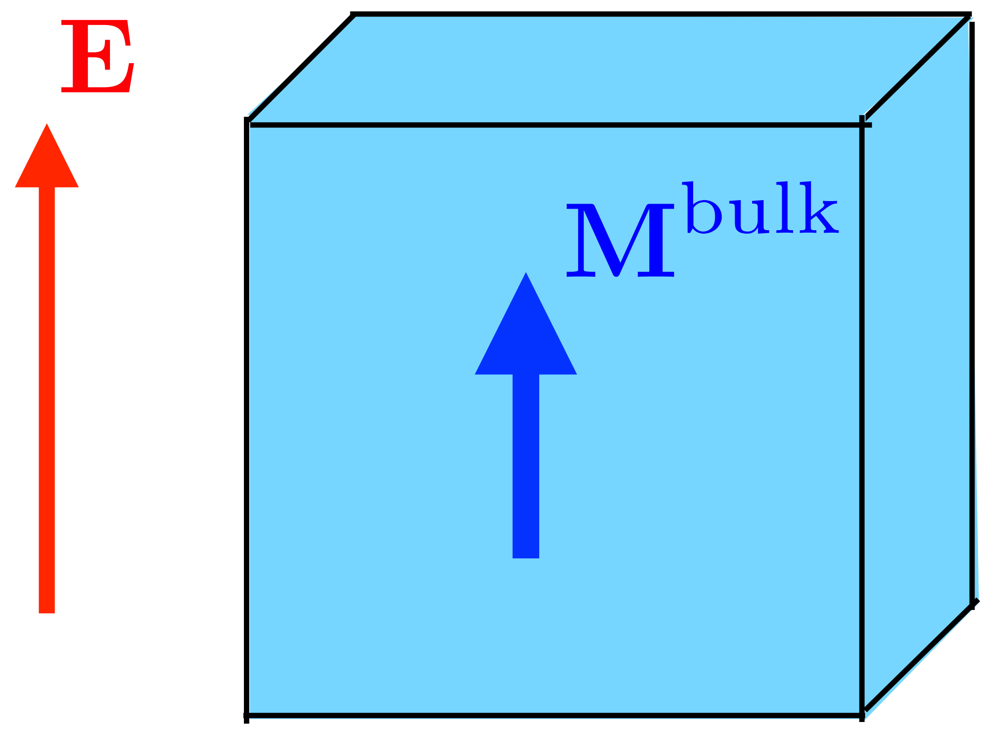
Bulk materials under applied electric fields versus surfaces in equilibrium

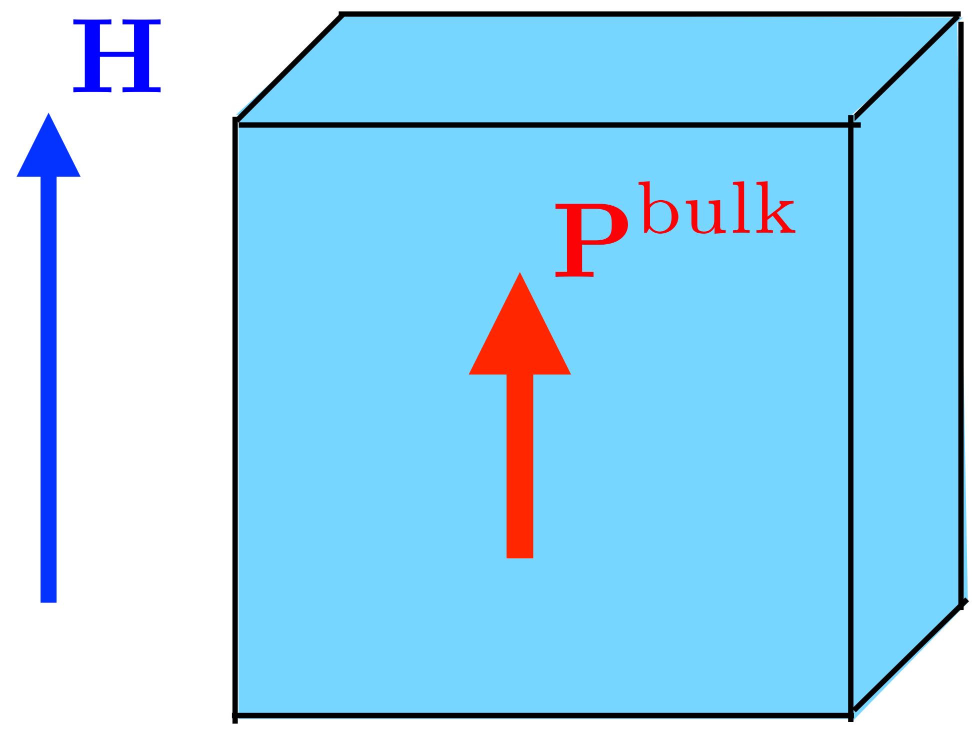


$$g^{\mathrm{bulk}+\mathbf{E}} = g^{\mathrm{surf}\perp\hat{\mathbf{n}}\|\mathbf{E}}$$

- *Properties induced by* $\mathbf{E}$ *in the bulk should arise in equilibrium at a surface, in the absence of any applied field!!*

Review/interlude: bulk magnetoelectric (ME) effect





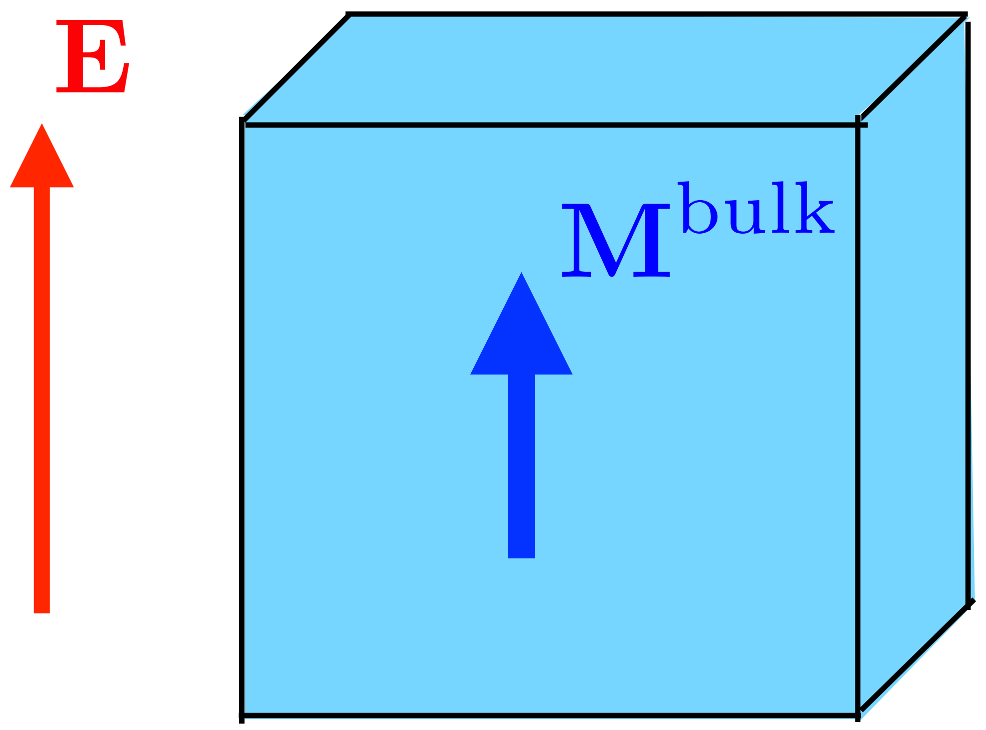
$$M_i = \boxed{\alpha_{ij}} E_j + \boxed{\beta_{ijk}} E_j E_k + \boxed{\gamma_{ijkl}} E_j E_k E_l + \ldots$$

Linear ME response tensor — Quadratic — Cubic

$$P_i = \alpha_{ij} H_i + \Omega_{ijk} H_i H_j + \kappa_{ijkl} H_i H_j H_k + \ldots$$

- Cross coupling of electric and magnetic fields: $\mathbf{E}$ induces $\mathbf{M}$ and $\mathbf{H}$ induces $\mathbf{P}$ in the bulk
- $\mathbf{M}$ and $\mathbf{P}$ can be induced at linear as well as higher orders of $\mathbf{E}$ and $\mathbf{H}$ (nonzero responses determined by bulk symmetry)

Review/interlude: bulk magnetoelectric (ME) effect



$$M_i = \alpha_{ij} E_j + \beta_{ijk} E_j E_k + \gamma_{ijkl} E_j E_k E_l + \ldots$$

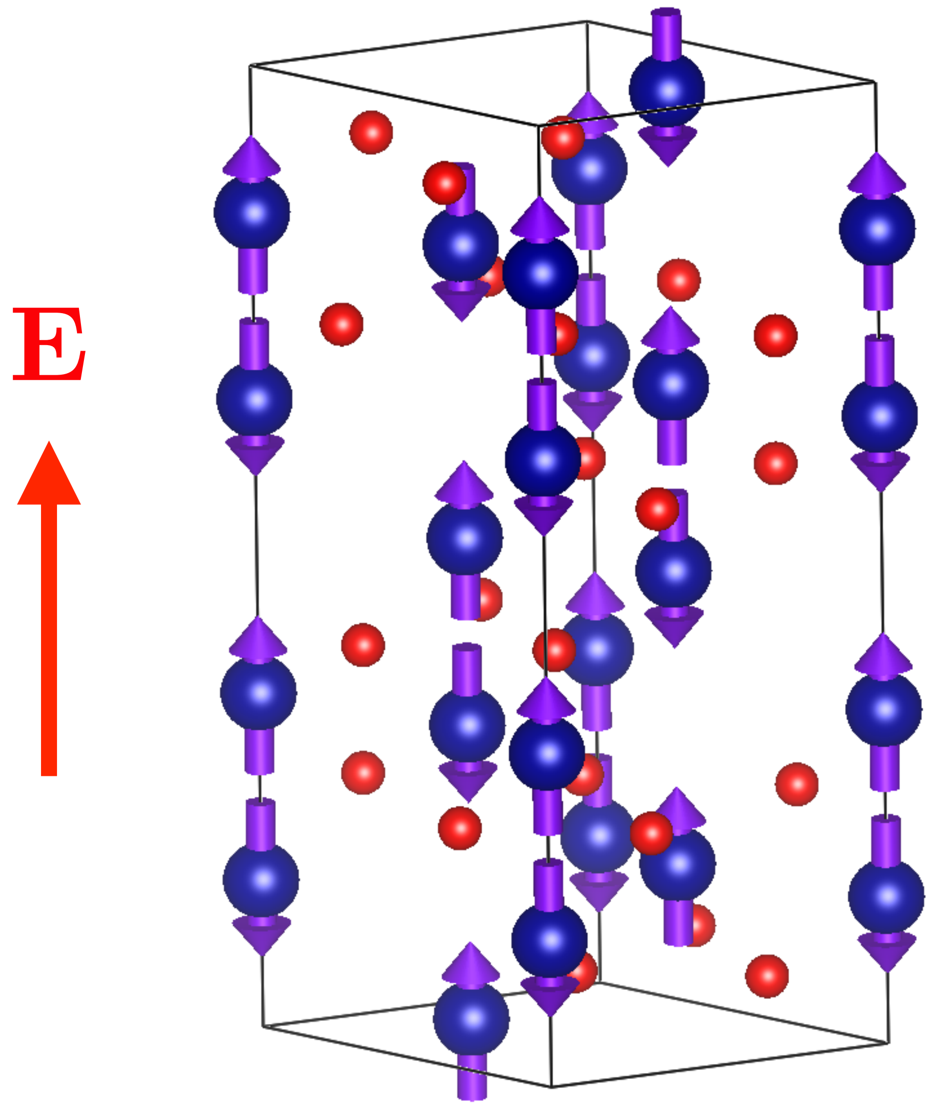
$$P_i = \alpha_{ij} H_i + \Omega_{ijk} H_i H_j + \kappa_{ijkl} H_i H_j H_k + \ldots$$

- We'll focus just on $\mathbf{M}$ induced by *polar vector* $\mathbf{E}$ for connection to surface magnetization

There is a one-to-one symmetry mapping for bulk ME responses *with* applied electric field, and surface magnetization *without* electric field!

$$M_i^{\mathrm{bulk}}(\mathbf{E}) = \alpha_{ij}E_j + \frac{1}{2}\beta_{ijk}E_jE_k + \frac{1}{2}\gamma_{ijkl}E_jE_kE_l + ...$$

$\mathbf{g}^{\mathbf{E}} = (\mathbf{R}^{\mathbf{E}}) < \mathbf{g}^{\mathrm{bulk}}$ for which $\mathbf{R}^{\mathbf{E}}\mathbf{E} = \mathbf{E}$



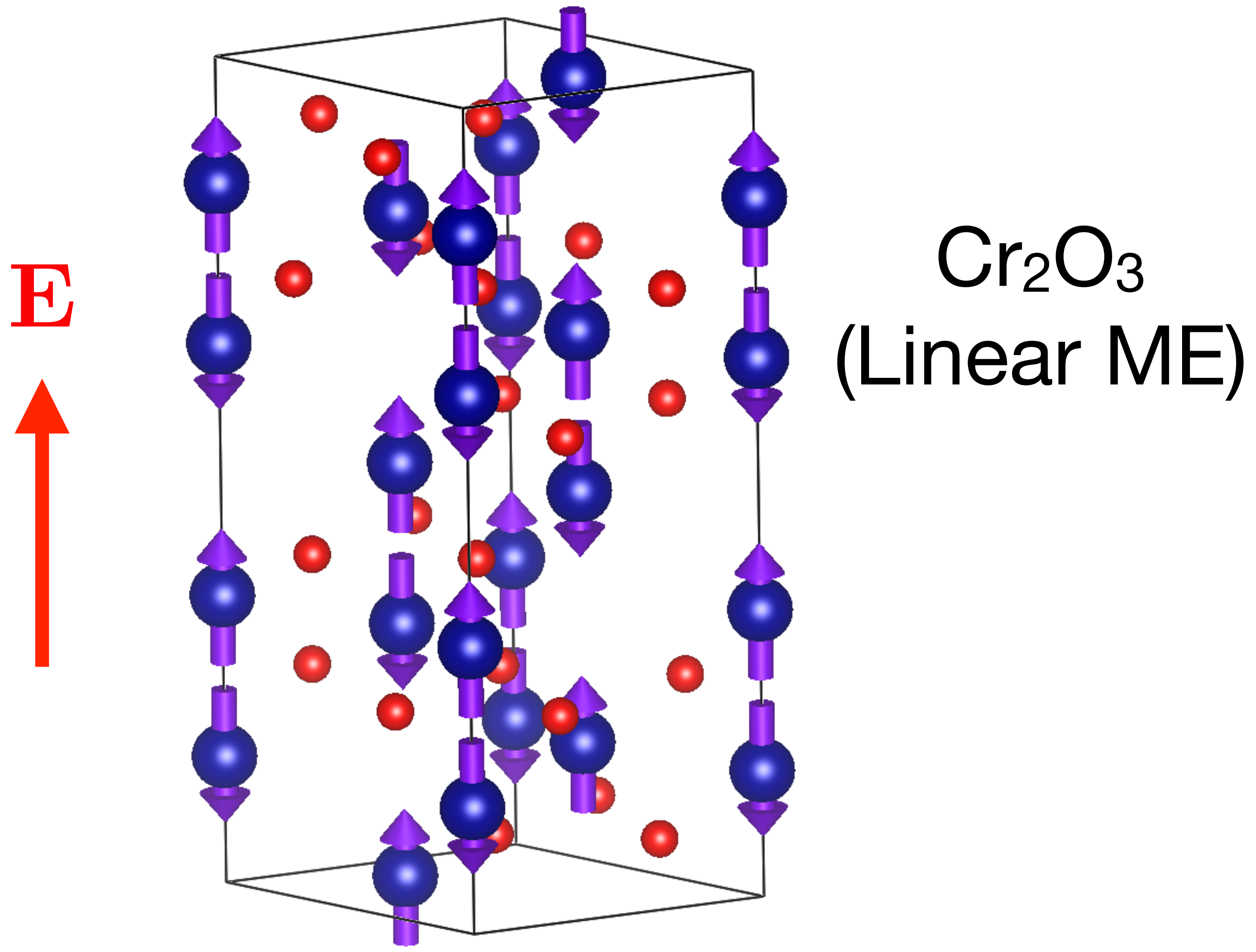
Cr_2O_3
(Linear ME)

- Induced magnetization determined by subgroup of bulk operations retained in presence of $\mathbf{E} \parallel \hat{\mathbf{r}}$

There is a one-to-one symmetry mapping for bulk ME responses *with* applied electric field, and surface magnetization *without* electric field!

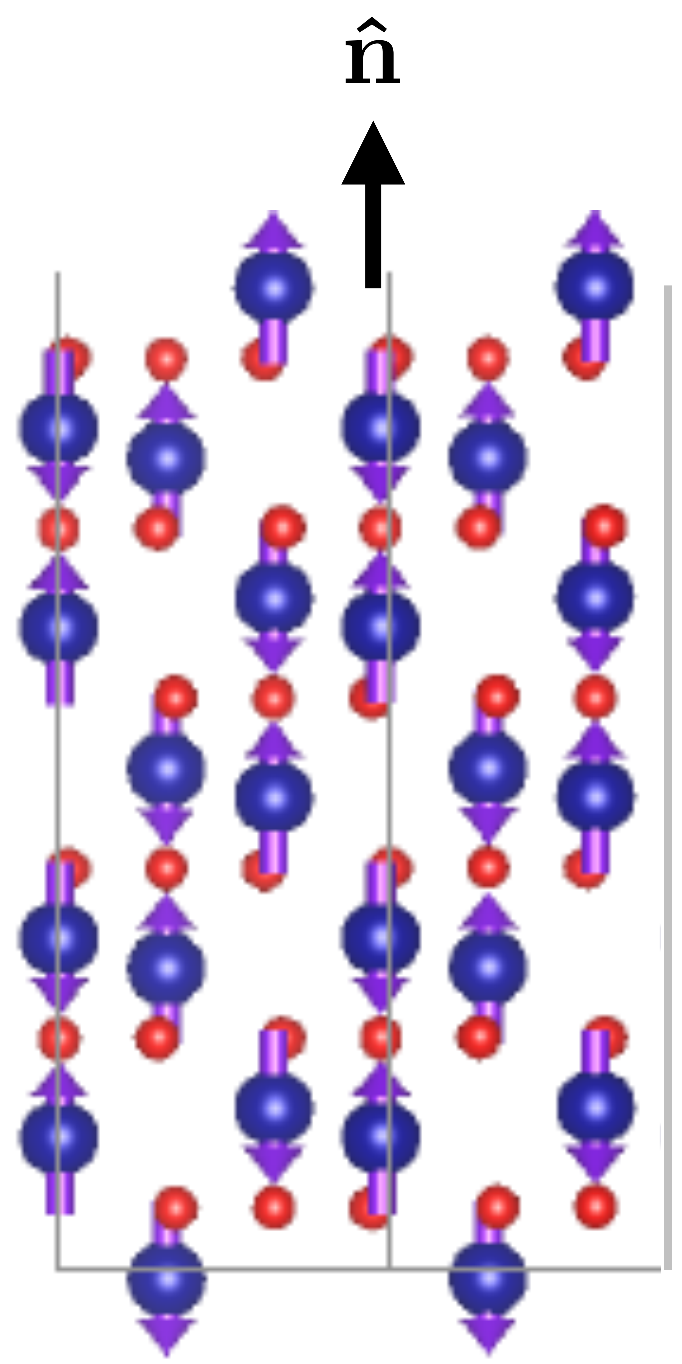
$$M_i^{\mathrm{bulk}}(\mathbf{E}) = \alpha_{ij} E_j + \frac{1}{2}\beta_{ijk} E_j E_k + \frac{1}{2}\gamma_{ijkl} E_j E_k E_l + \ldots$$

$\mathbf{g^E} = (\mathbf{R^E}) < \mathbf{g}^{\mathrm{bulk}}$ for which $\mathbf{R^E E} = \mathbf{E}$



$$M_i^{\mathrm{surf}}(\hat{\mathbf{n}}) \propto \alpha_{ij} n_j + \beta_{ijk} n_j n_k + \gamma_{ijkl} n_j n_k n_l + \ldots$$

$\mathbf{g}^{\hat{\mathbf{n}}} = (\mathbf{R}^{\hat{\mathbf{n}}}) < \mathbf{g}^{\mathrm{bulk}}$ for which $\mathbf{R}^{\hat{\mathbf{n}}}\hat{\mathbf{n}} = \hat{\mathbf{n}}$



- Induced magnetization determined by subgroup of bulk operations retained in presence of $\mathbf{E} \parallel \hat{\mathbf{r}}$
- Surface with normal $\hat{\mathbf{n}}$ acquires same magnetization components as bulk for $\mathbf{E} \parallel \hat{\mathbf{n}}$

II. Bulk ME responses as predictors of surface magnetic properties

(a) Surface ferromagnetism in antiferromagnets

(b) Surface antiferromagnetism (rotation or magnitude changes of bulk Néel vector)

(c) Surface linear ME effects in bulk higher-order magnetoelectrics

Net surface magnetization in magnetoelectric (ME) antiferromagnets

quadratic response tensor

$$M_i = \alpha_{ij} E_j$$

linear ME response tensor

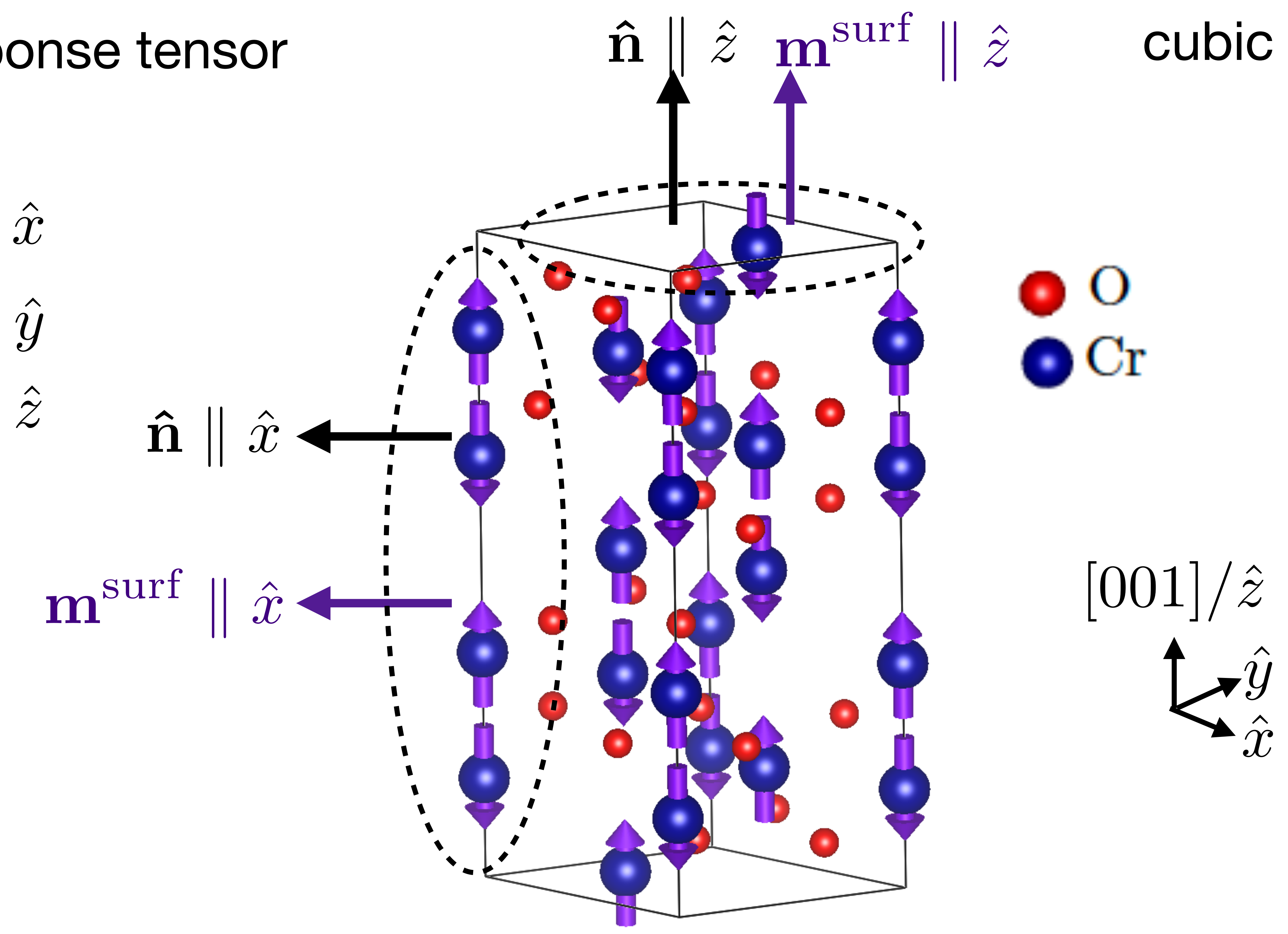
$$M_i = \beta_{ijk} E_j E_k$$

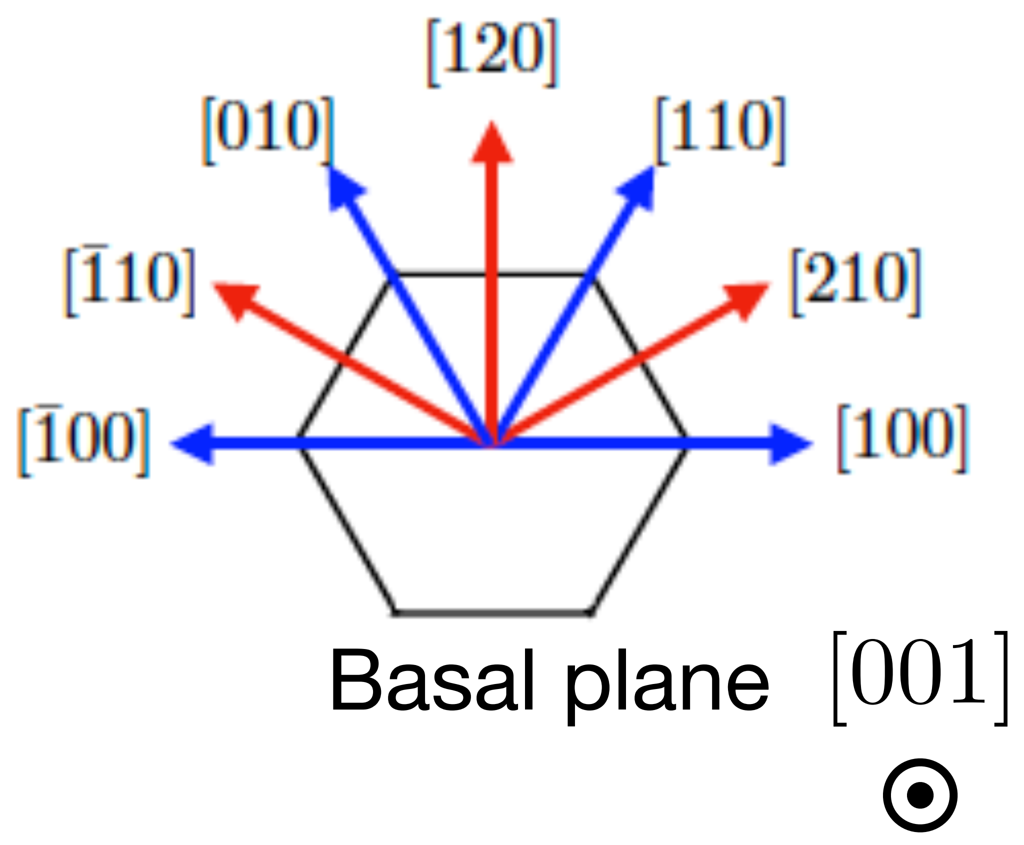
$$M_i = \gamma_{ijkl} E_j E_k E_l$$

cubic response tensor

$$\alpha^{\mathrm{Cr_2O_3}} = \begin{pmatrix} \alpha_\perp & 0 & 0 \\ 0 & \alpha_\perp & 0 \\ 0 & 0 & \alpha_\parallel \end{pmatrix} \begin{matrix} \hat{x} \\ \hat{y} \\ \hat{z} \end{matrix}$$

$$\begin{matrix} \hat{x} & \hat{y} & \hat{z} \end{matrix}$$

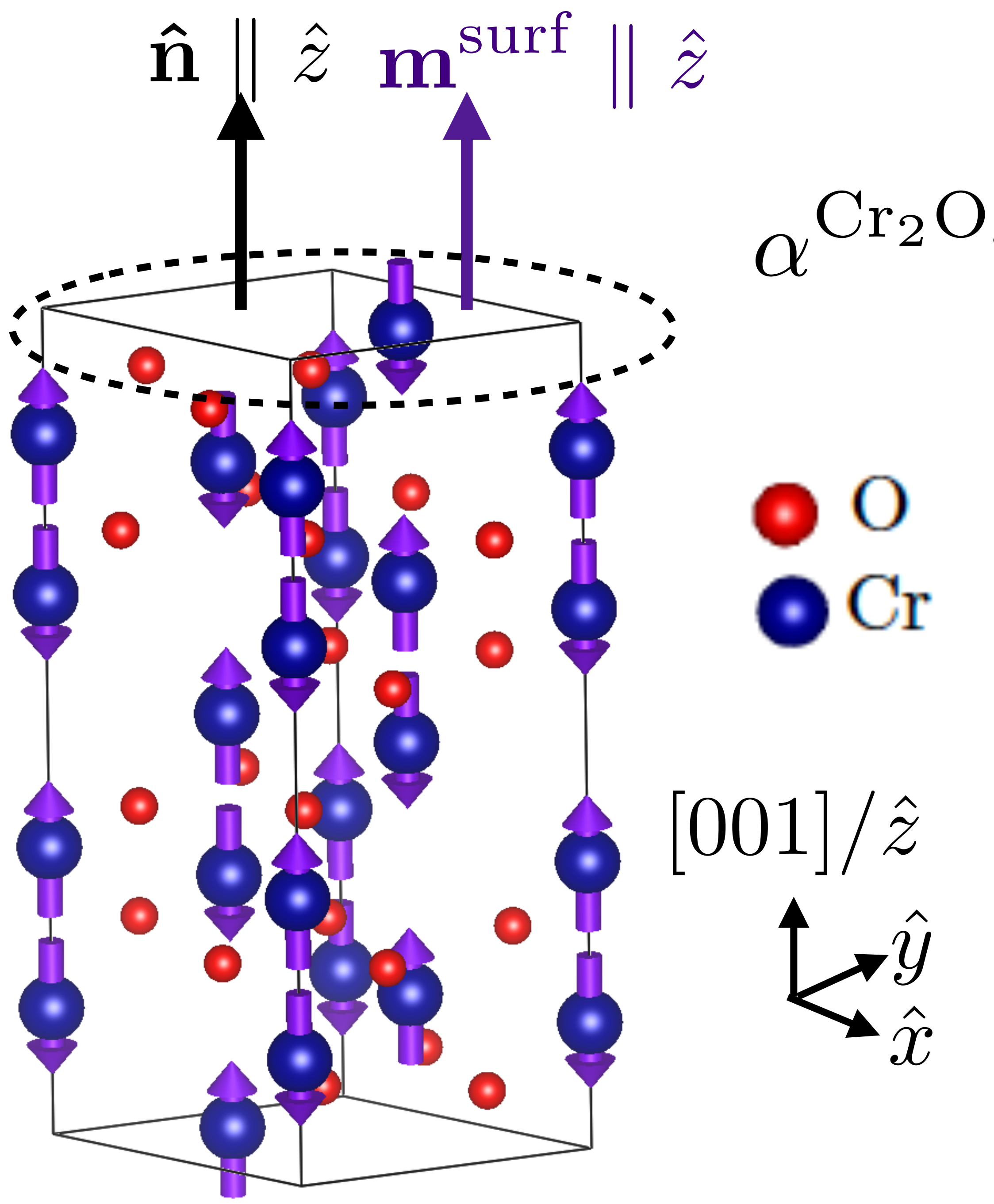




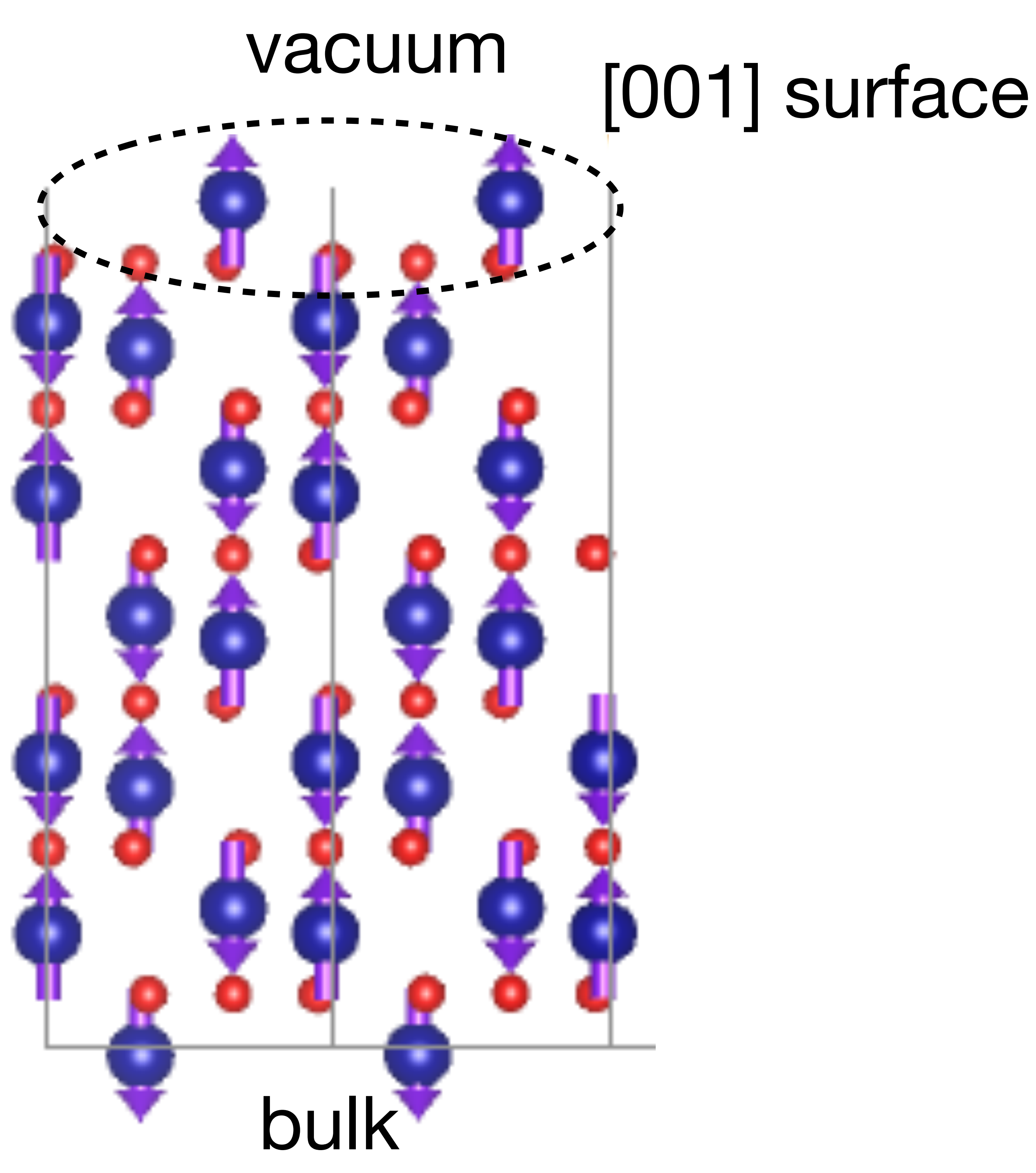
- Given a nonzero element α_{ij} of ME tensor, we expect a 2D equilibrium magnetization along $\hat{i}$ for a surface $\perp \hat{j}$

- Higher-order bulk ME materials also have symmetry-mandated surface magnetization!
  (e.g. for nonzero β_{ijj} we also expect $\hat{i}$-oriented magnetization on surface $\perp \hat{j}$)

Surface magnetization arises even for planes that are magnetically compensated in the bulk!



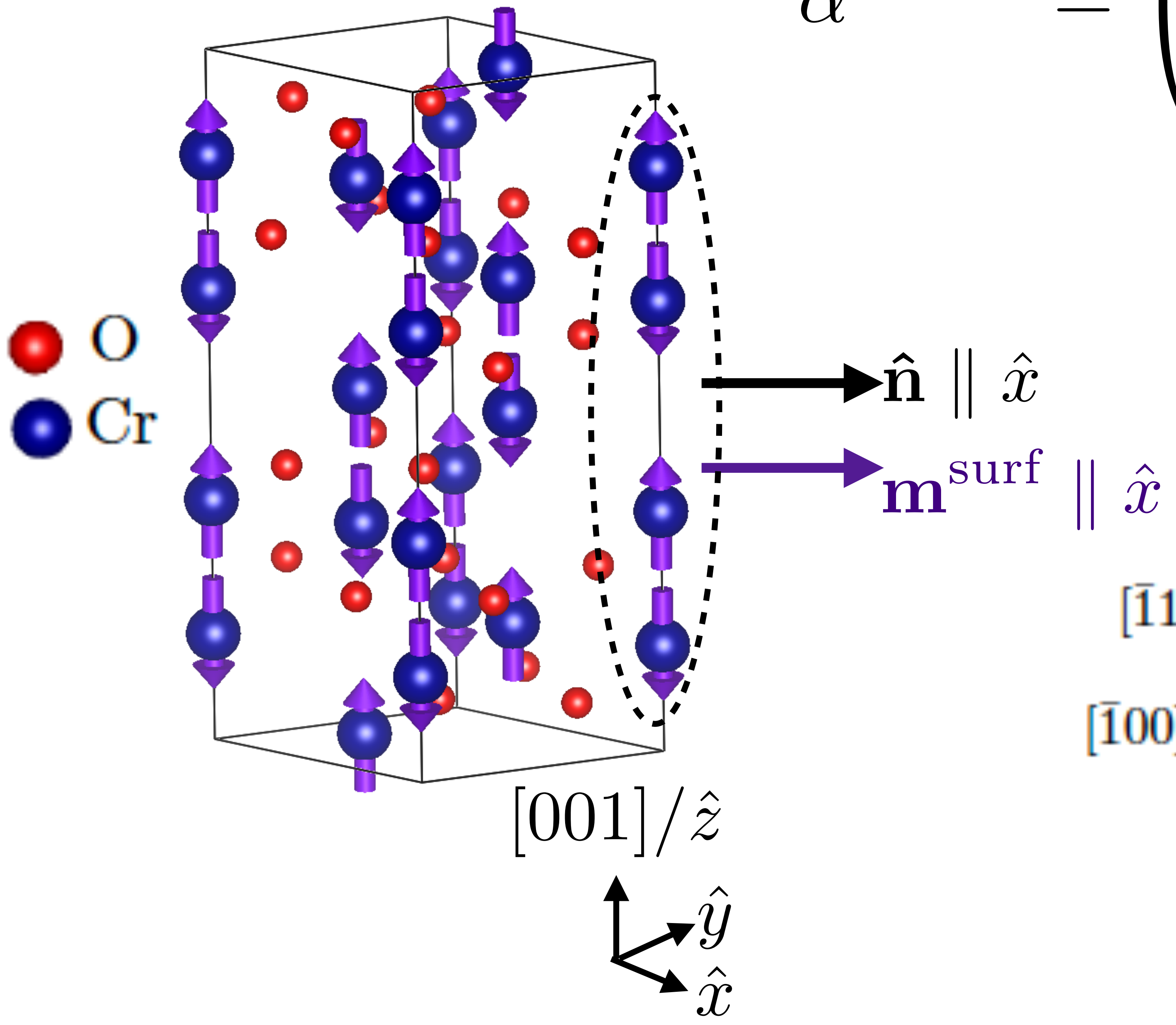
$$\alpha^{\mathrm{Cr_2O_3}} = \begin{pmatrix} \alpha_\perp & 0 & 0 \\ 0 & \alpha_\perp & 0 \\ 0 & 0 & \alpha_\parallel \end{pmatrix}$$

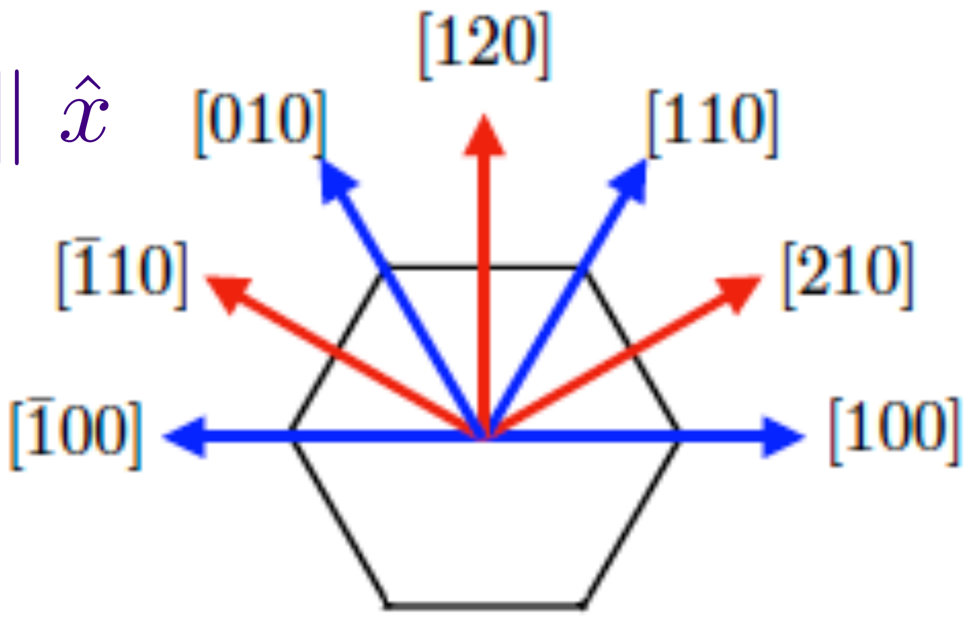


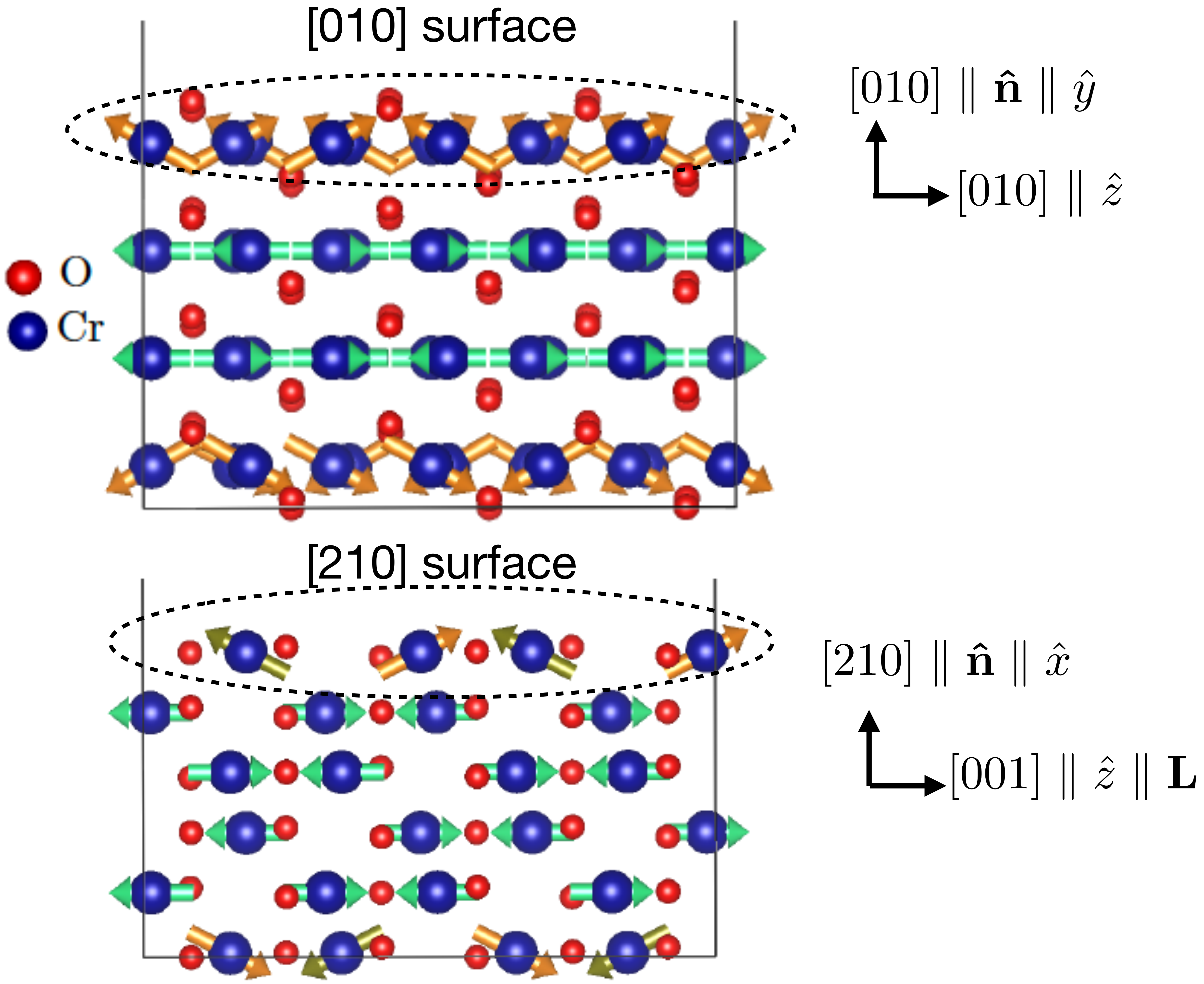
- Surface magnetization is unsurprising when 2D planes are already ferromagnetic in bulk (“uncompensated” surface magnetization)

Surface magnetization arises even for planes that are magnetically compensated in the bulk!

$$\alpha^{\mathrm{Cr_2O_3}} = \begin{pmatrix} \boxed{\alpha_\perp} & 0 & 0 \\ 0 & \boxed{\alpha_\perp} & 0 \\ 0 & 0 & \alpha_\parallel \end{pmatrix}$$

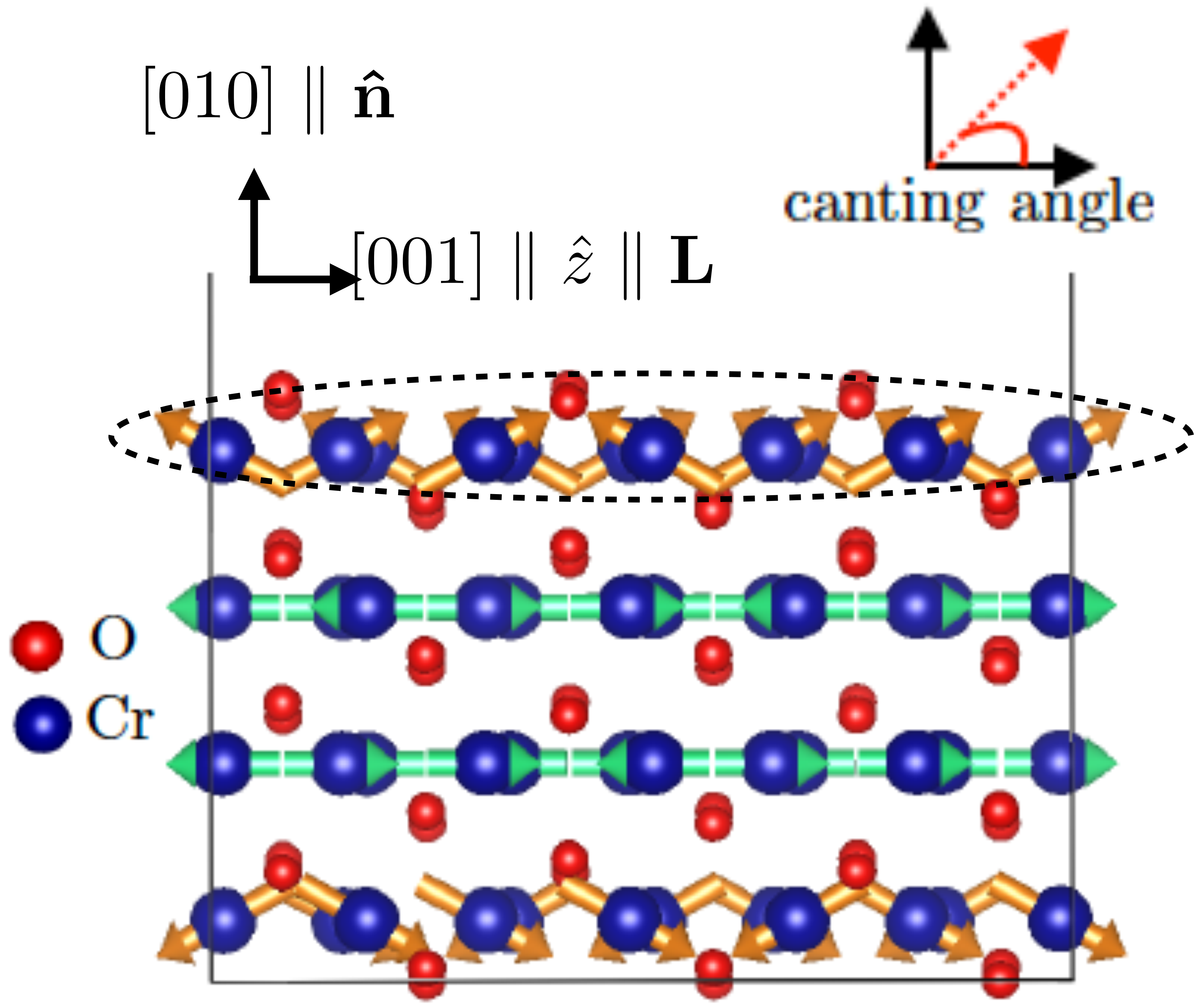




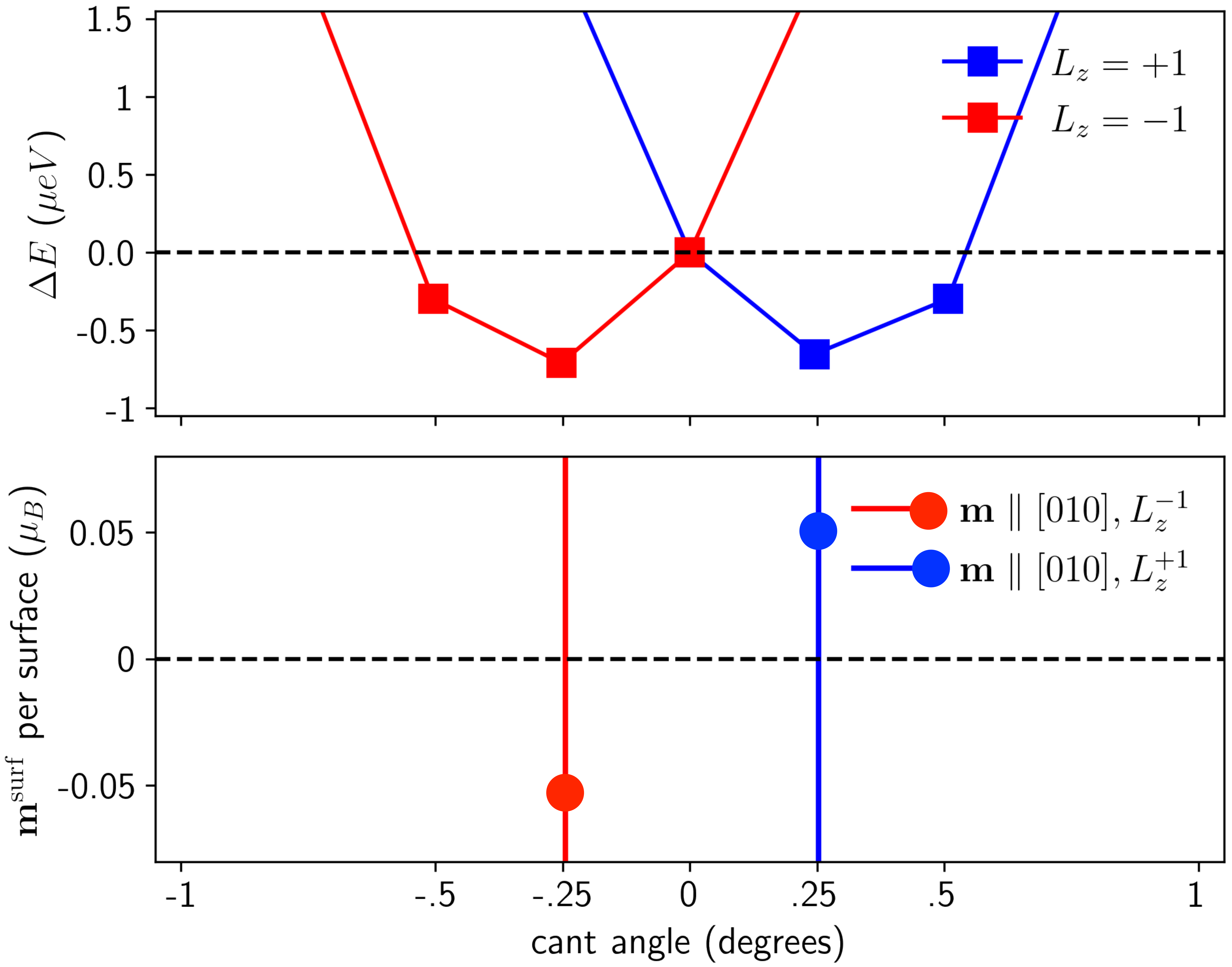


- More surprising for surface planes that are antiferromagnetic in bulk! (“induced surface magnetization”)
- Modification of magnetic moments from bulk order due to *symmetry lowering at surface*

Induced surface magnetization in Cr_2O_3 confirmed by density functional theory (DFT) and magnetotransport



SFW*, A. Urru* et al., *Phys. Rev. X* **14**, 021033 (2024)
(Featured in Physics)



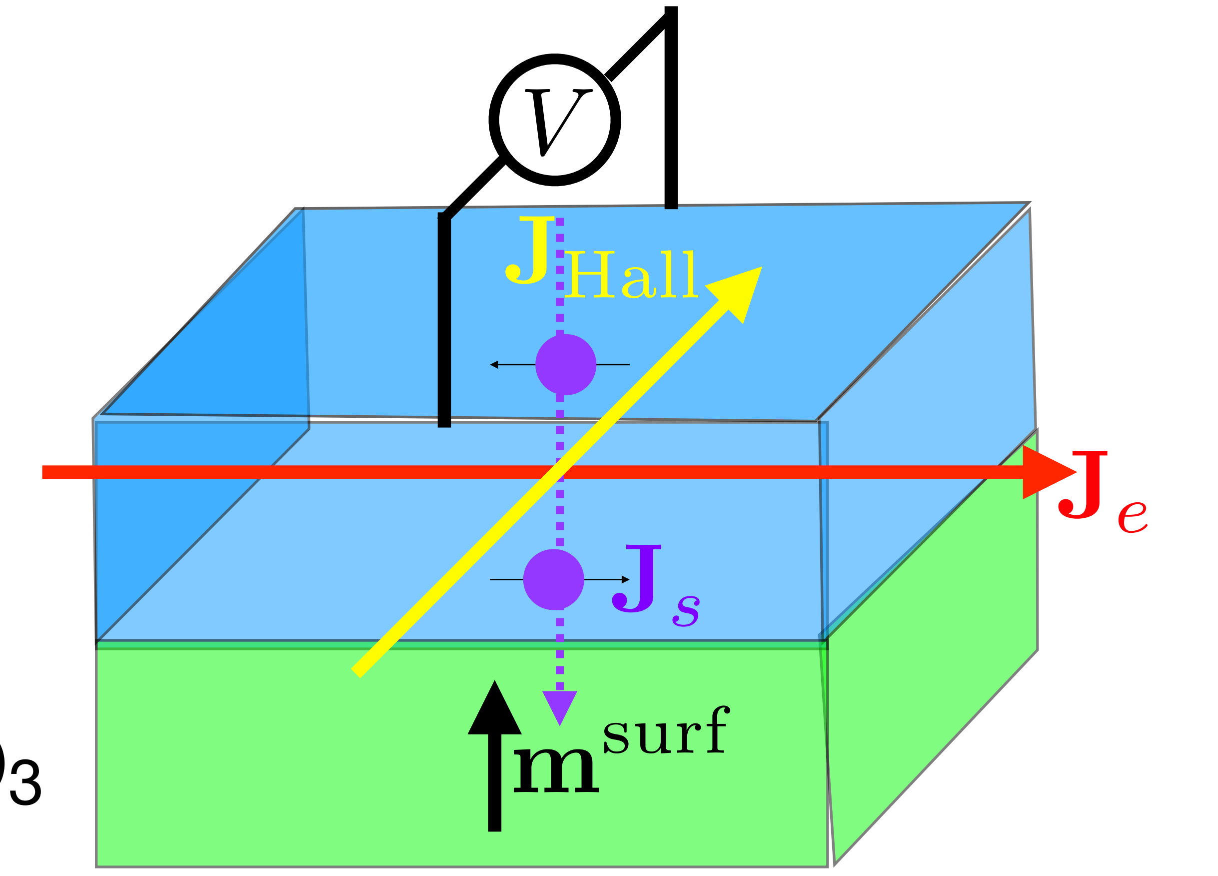
- Energetic preference for finite surface magnetization
- Sign of surface M switches for opposite bulk domains! (can be used for domain readout in spintronics)

Induced surface magnetization in Cr_2O_3 confirmed by DFT and magnetotransport

Spin-Hall magnetoresistance $\mathbf{J}_{\mathrm{Hall}} \propto \mathbf{m}_{\perp}^{\mathrm{surf}}$

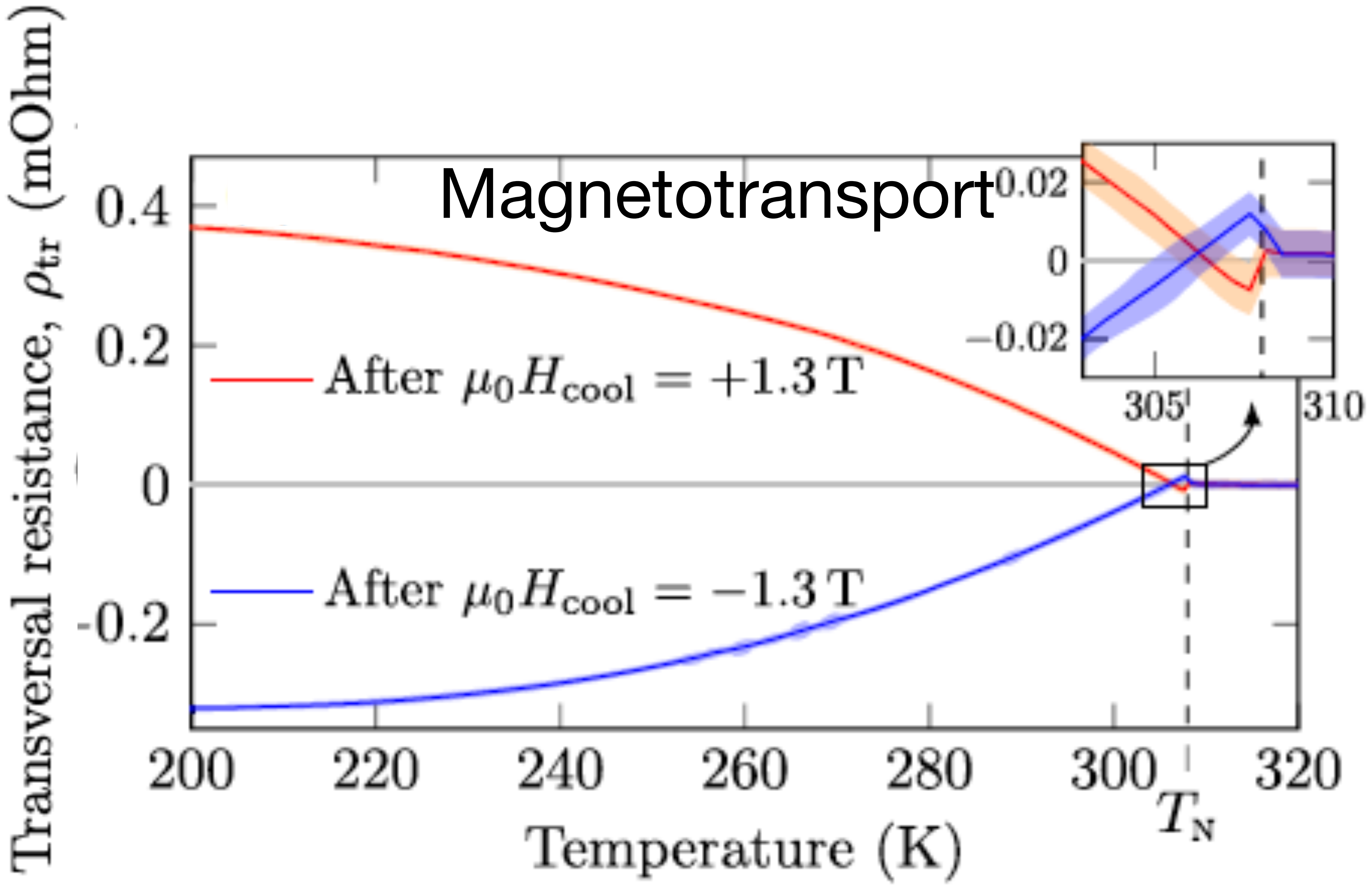
Pt

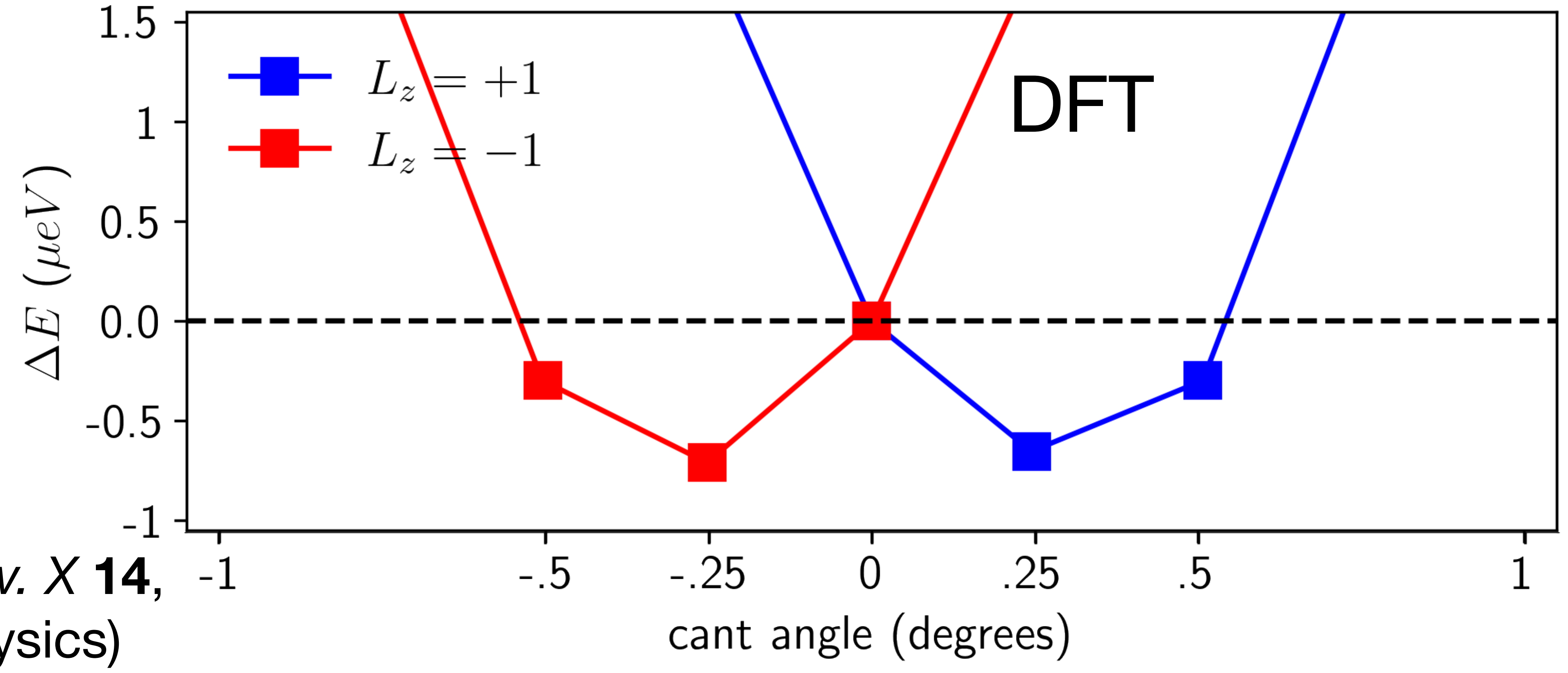
Cr_2O_3



O. V. Pylypovskyi*, SFW* et al., *Phys. Rev. Lett.* **132**, 226702 (2024) (Editor's suggestion)

Denys Makarov
(Helmholtz-Zentrum, Dresden)

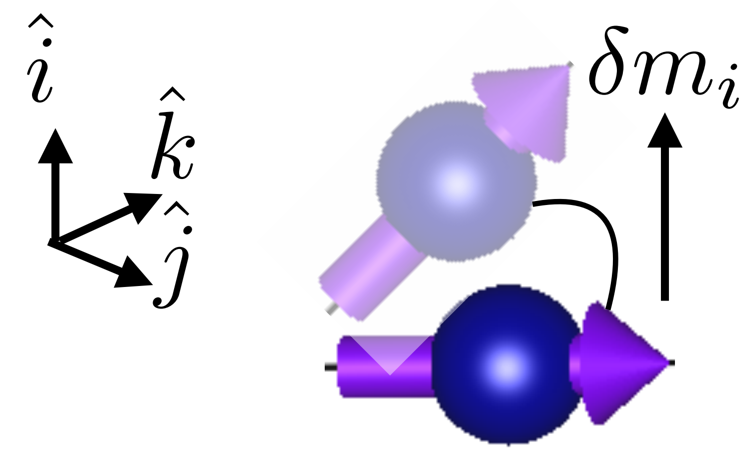




SFW*, A. Urru* et al., *Phys. Rev. X* **14**, 021033 (2024) (Featured in Physics)

- Out-of-plane surface magnetization also detected in magnetotransport measurements of collaborators

Beyond bulk magnetoelectrics



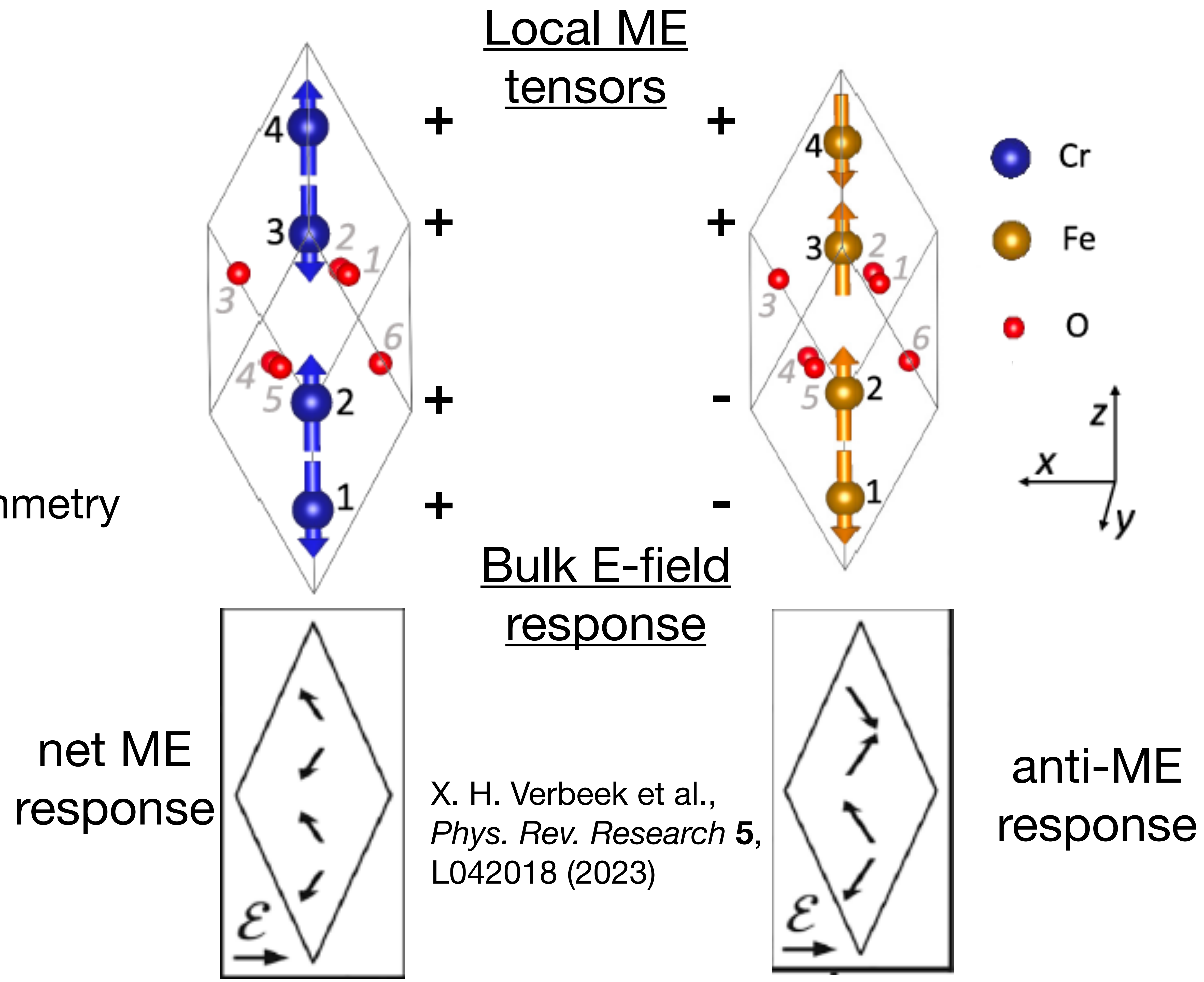
$$\delta m_i = \sum_j \beta^{loc}_{ijk} E_j E_k$$

$$\delta m_i = \sum_j \alpha^{loc}_{ij} E_j$$

$$\delta m_i = \sum_j \gamma^{loc}_{ijkl} E_j E_k E_l$$

determined by Wyckoff site symmetry

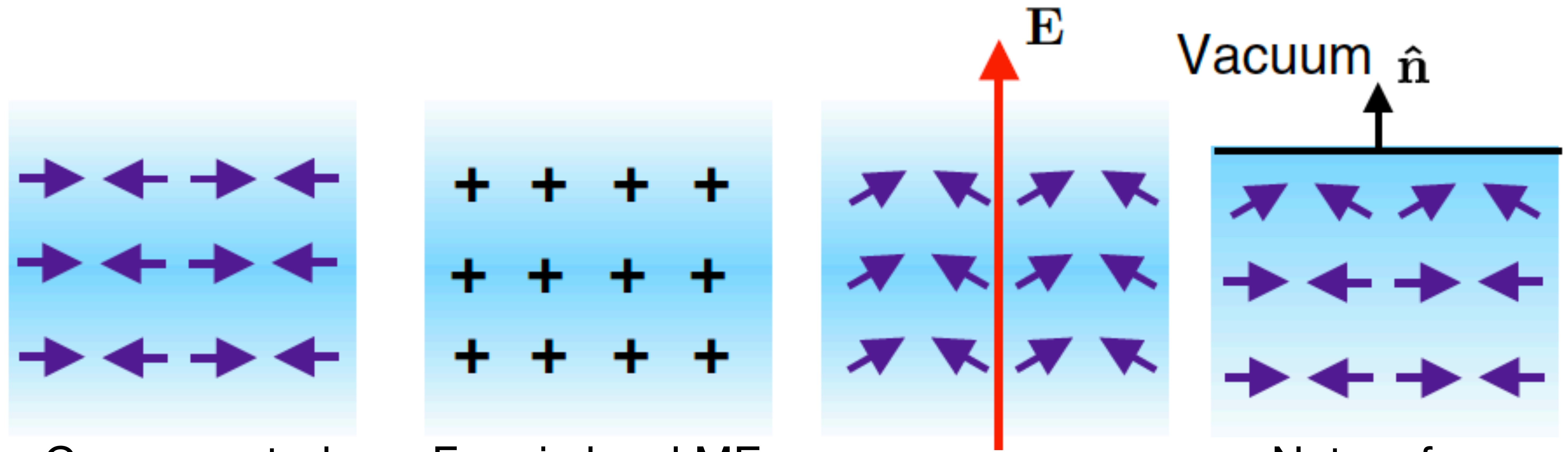
$$\mathbf{g}^{W,\mathrm{bulk}} < \mathbf{g}^{\mathrm{bulk}}$$



X. H. Verbeek et al., *Phys. Rev. Research* **5**, L042018 (2023)

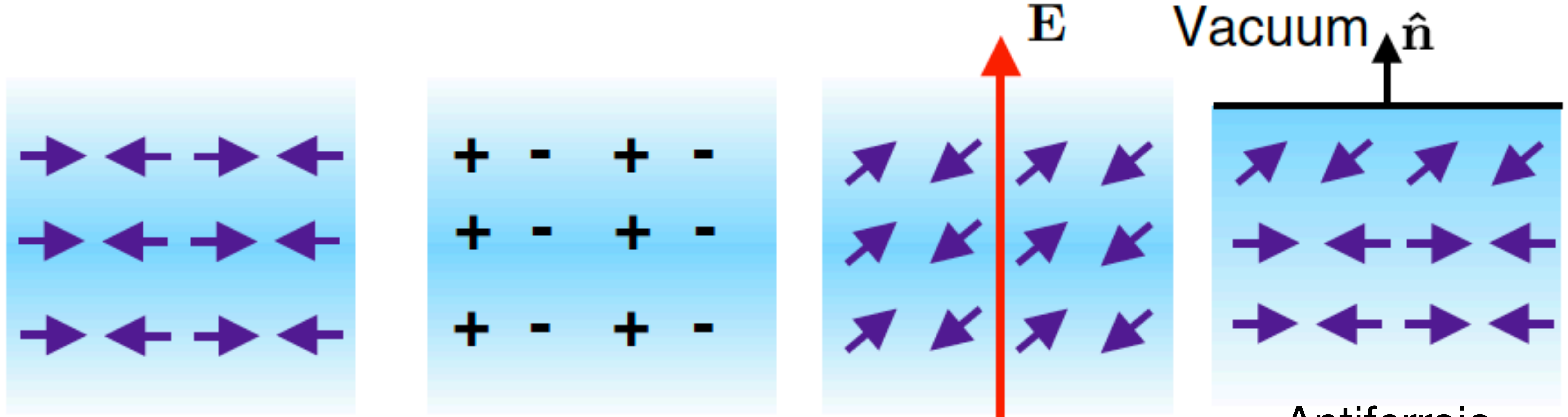
- Even if bulk crystal symmetry disallows net ME response, *local*, atomic-site ME responses still occur
- If local ME responses have same sign on all atoms: net ME response.
- If local tensors are antiferroically ordered: an antiferroic ME response!!

Bulk “anti-MEs” have analogous symmetry-dictated surface magnetic order



Compensated magnetic dipoles | Ferroic local ME responses | Bulk ME response | Net surface magnetization

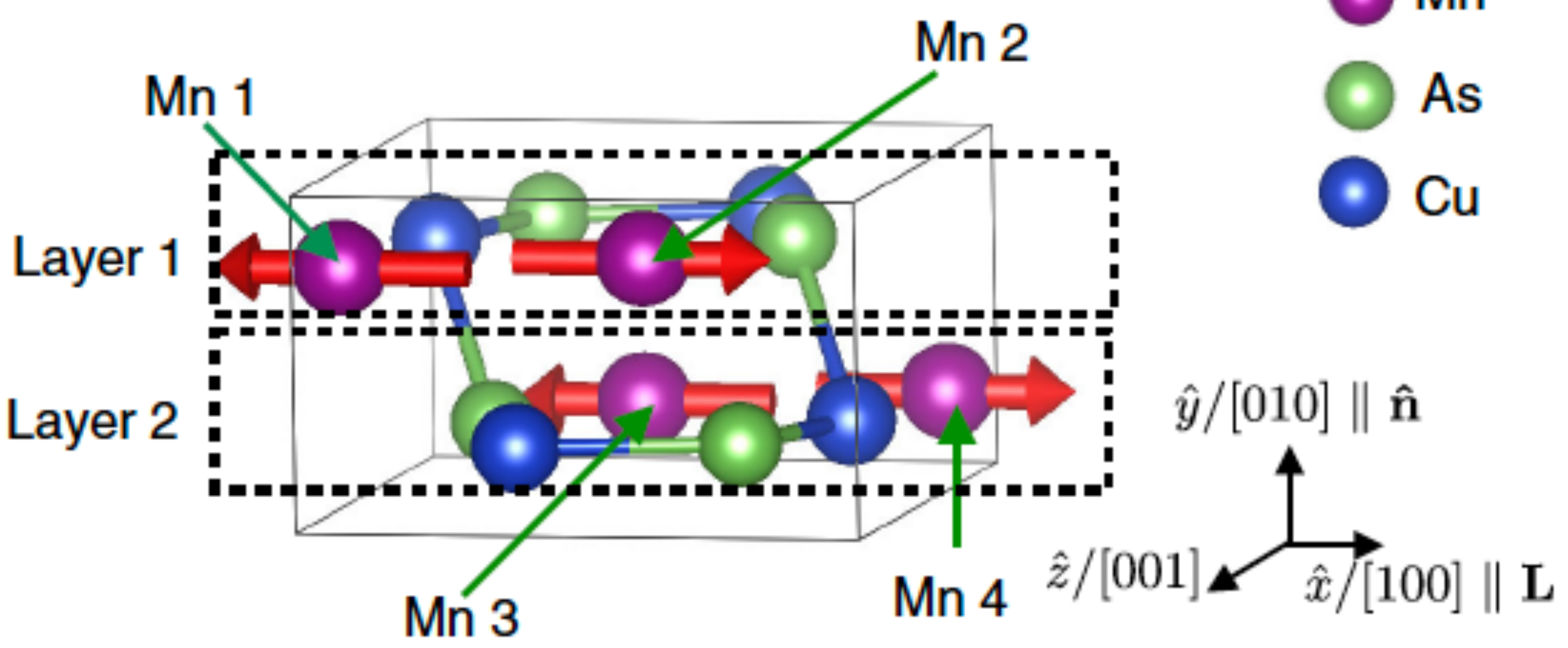
SFW et al., *Phys. Rev. X* **15**, 021094 (2025)



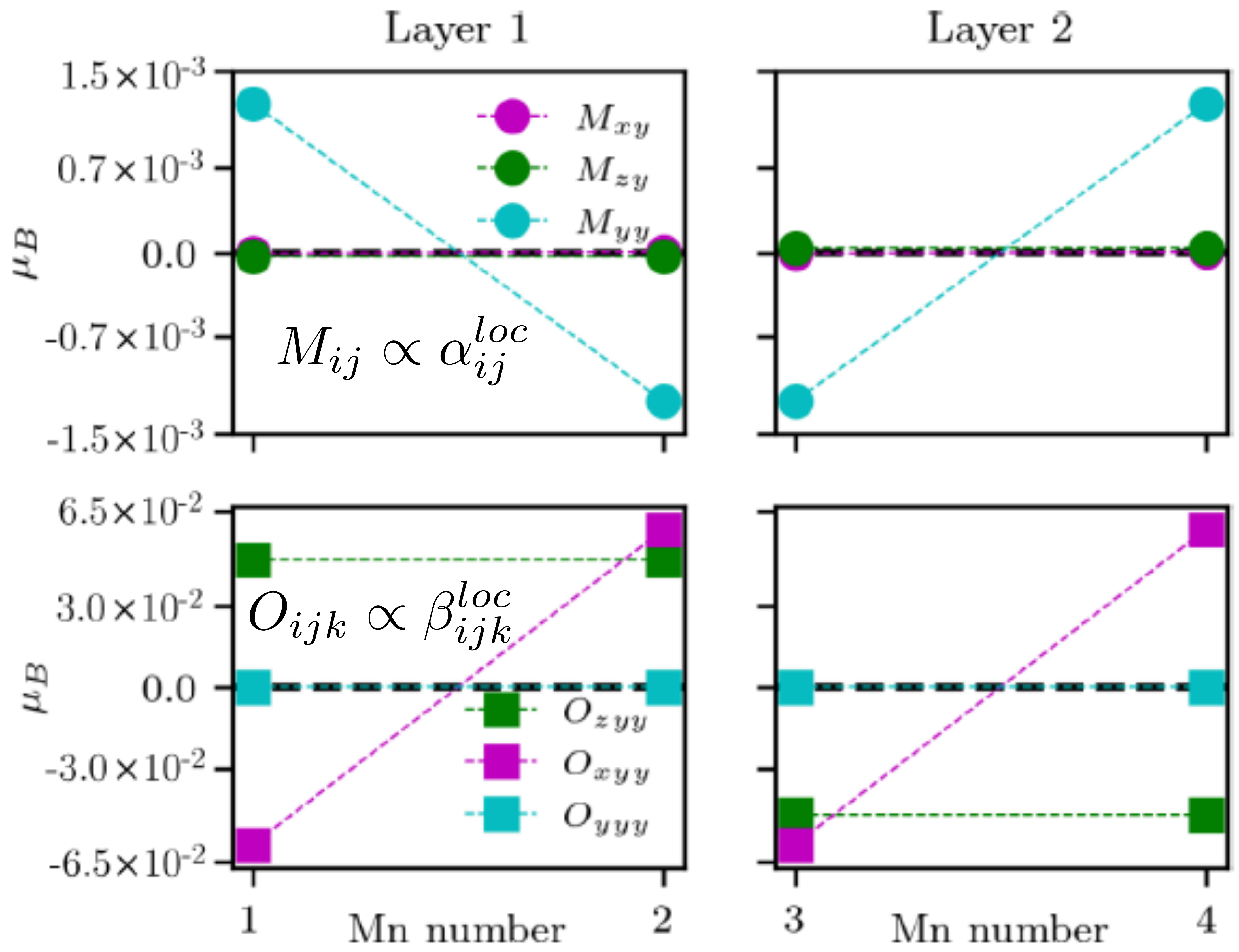
Compensated magnetic dipoles | Antiferroic local ME responses | Bulk anti-ME response | Antiferroic surface modifications

- An AFM surface stays AFM, but surface Néel vector gets modified (rotations and length changes)

DFT-based validation: surface magnetic order of CuMnAs



SFW et al., *Phys. Rev. X* **15**, 021094 (2025)



Local linear ME tensors

Local quadratic ME tensors

Based on relative signs of local bulk ME tensors, for the (010) surface we expect:

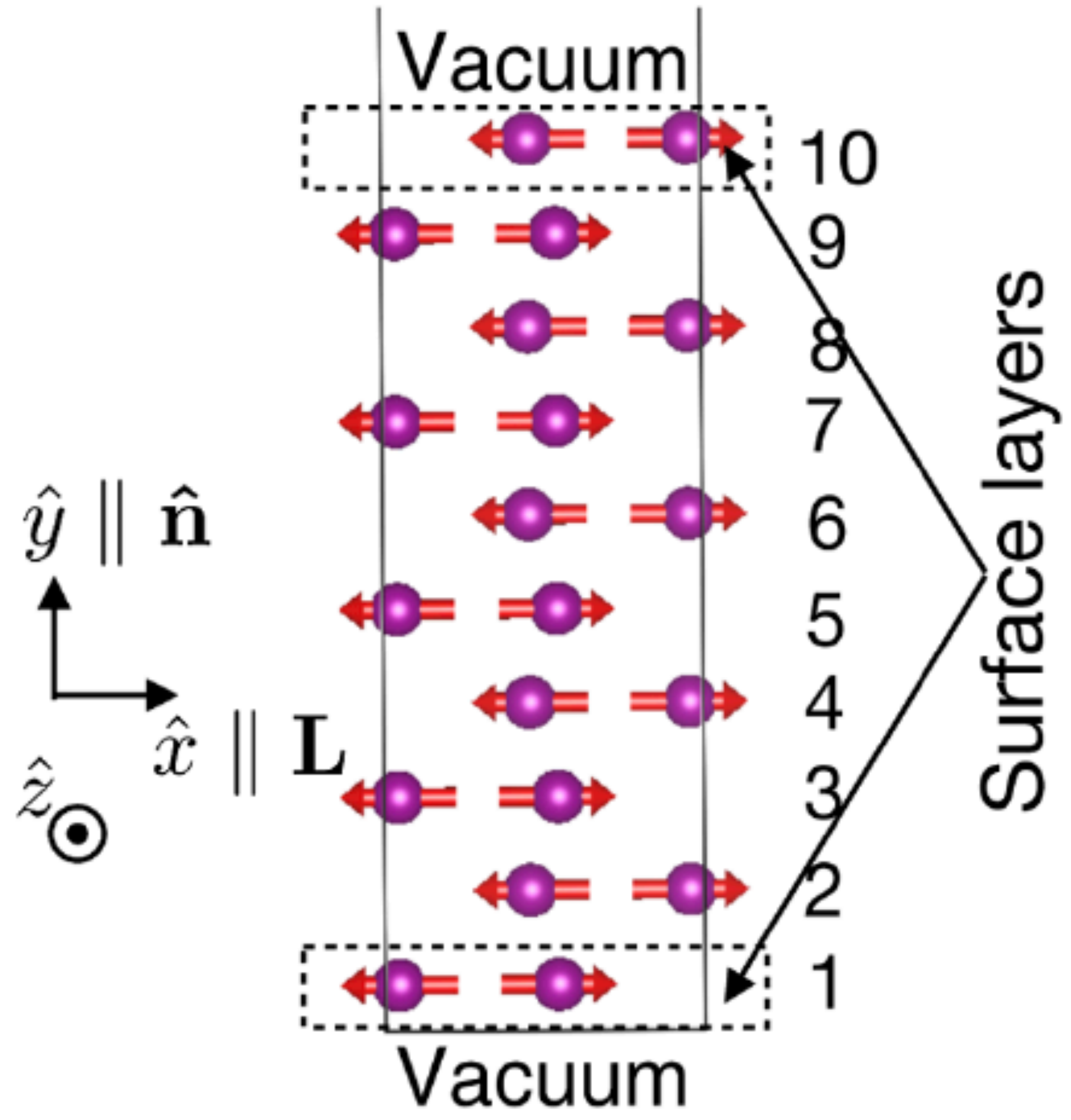
1. Out-of-plane canting $\parallel \hat{y}/[010]$ which is antiferroic in-plane (surface Néel vector rotation)

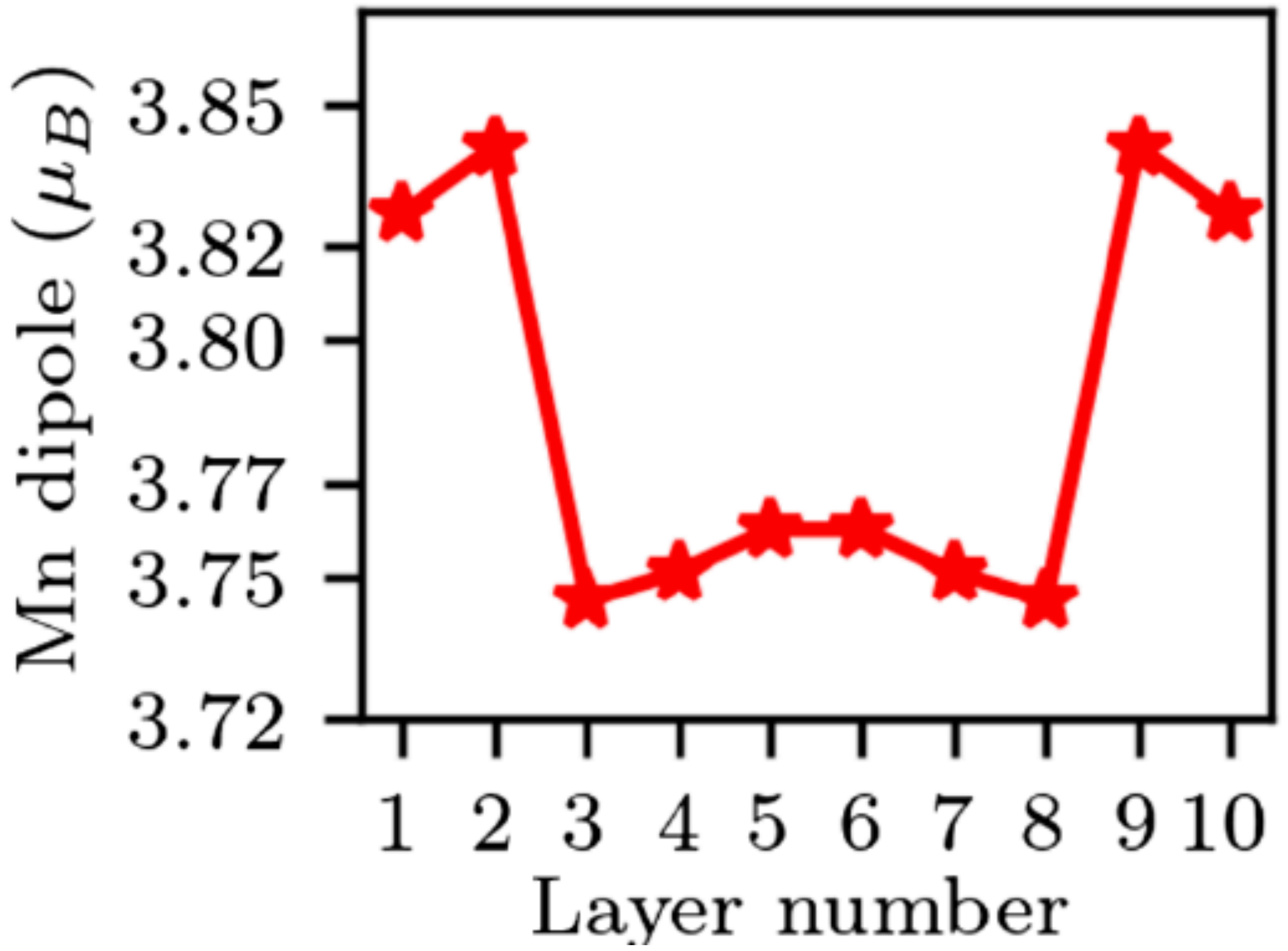
2. Equal and opposite changes in length of dipole moments $\parallel \hat{x}/[100]$ (surface Néel vector length change)

3. In-plane canting $\parallel \hat{z}/[001]$ which is ferroic in-plane, but switches sign between layers ("roughness sensitive" surface ferromagnetism)

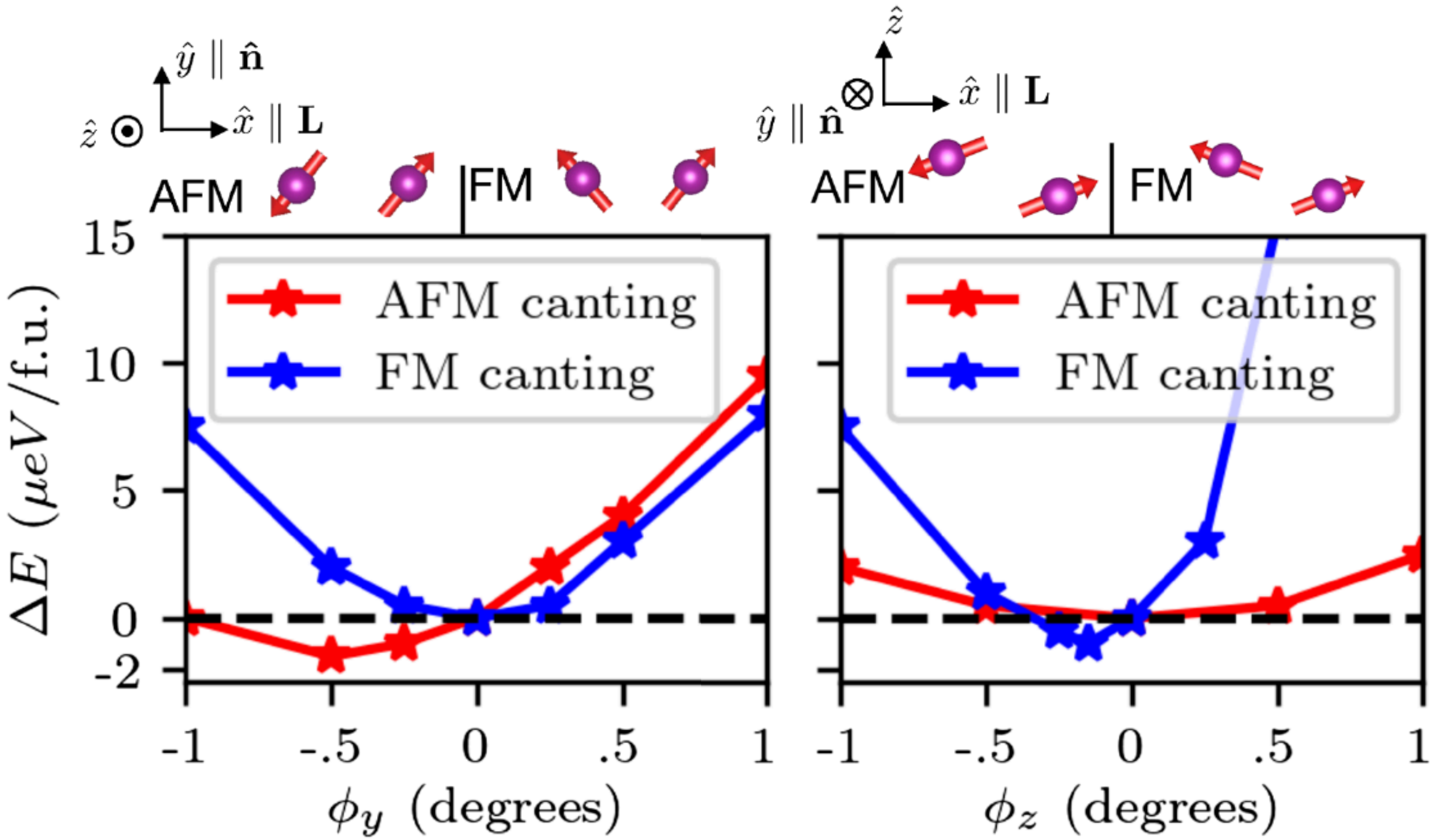
Energetics validate ME-based predications

SFW et al., *Phys. Rev. X* **15**, 021094 (2025)





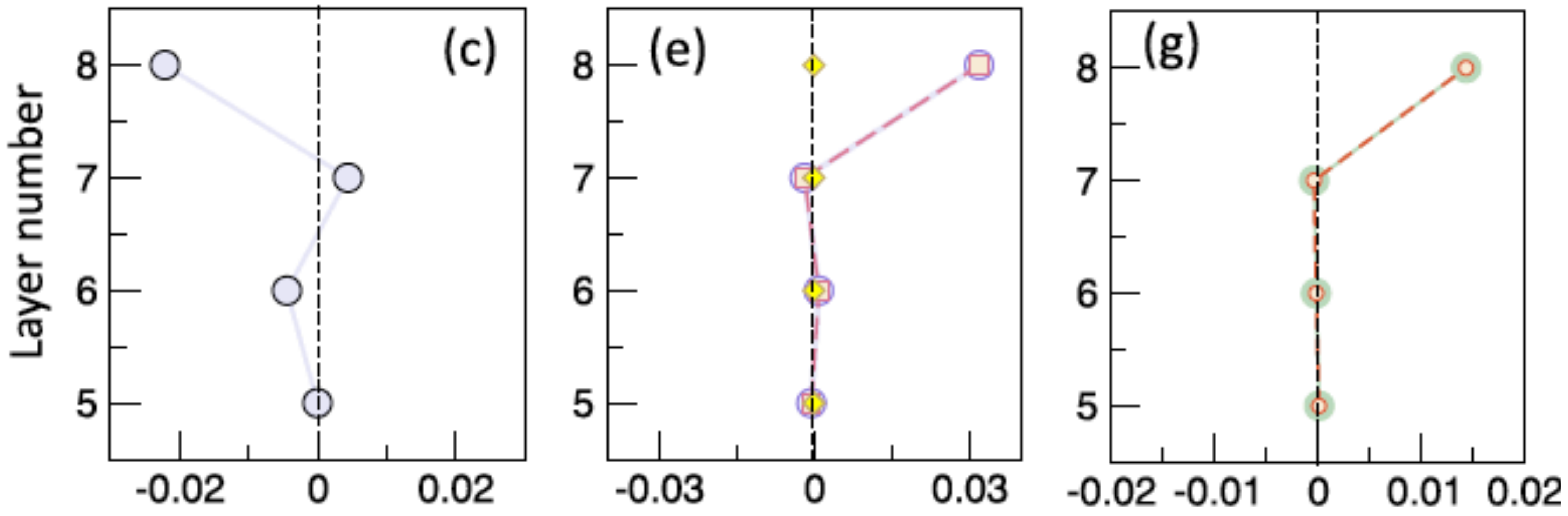
surface Néel vector length changes



out-of plane Néel vector rotation

“roughness-sensitive” surface ferromagnetism

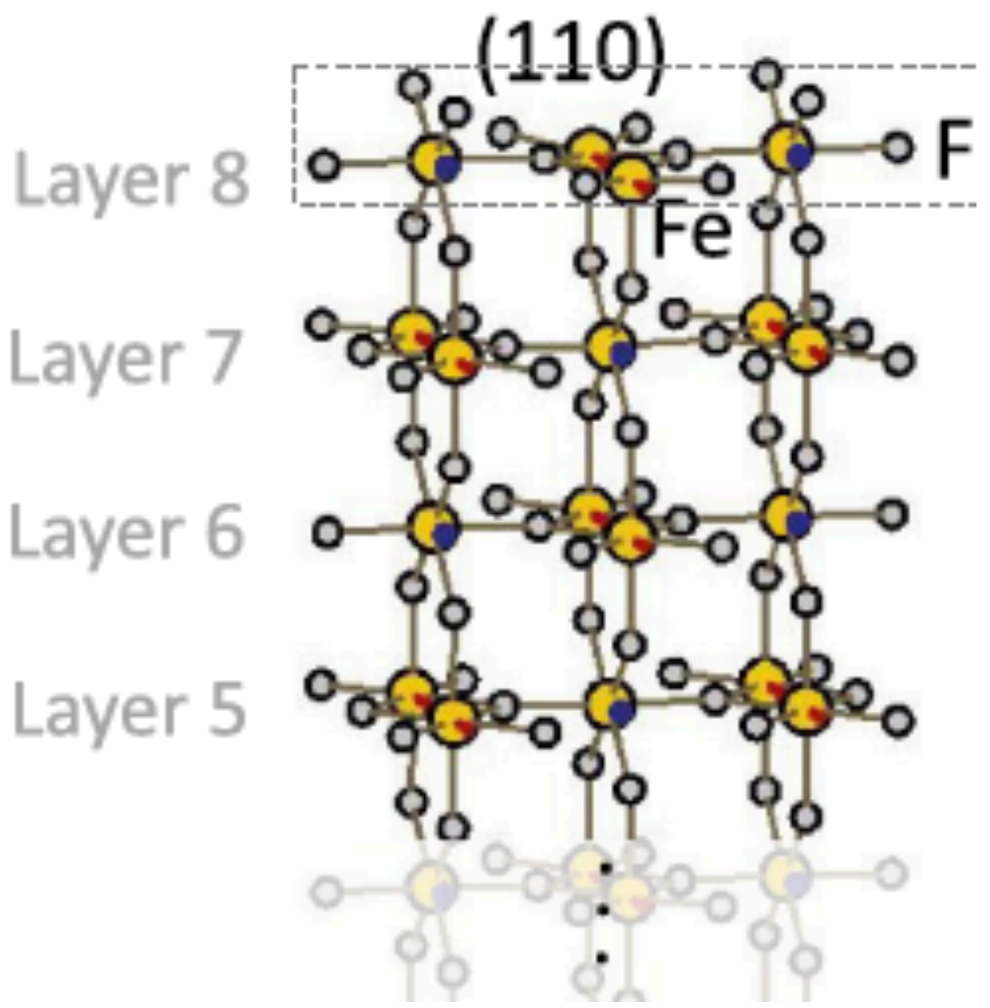
Additional surface property of higher-order bulk MEs:
surface linear ME effect

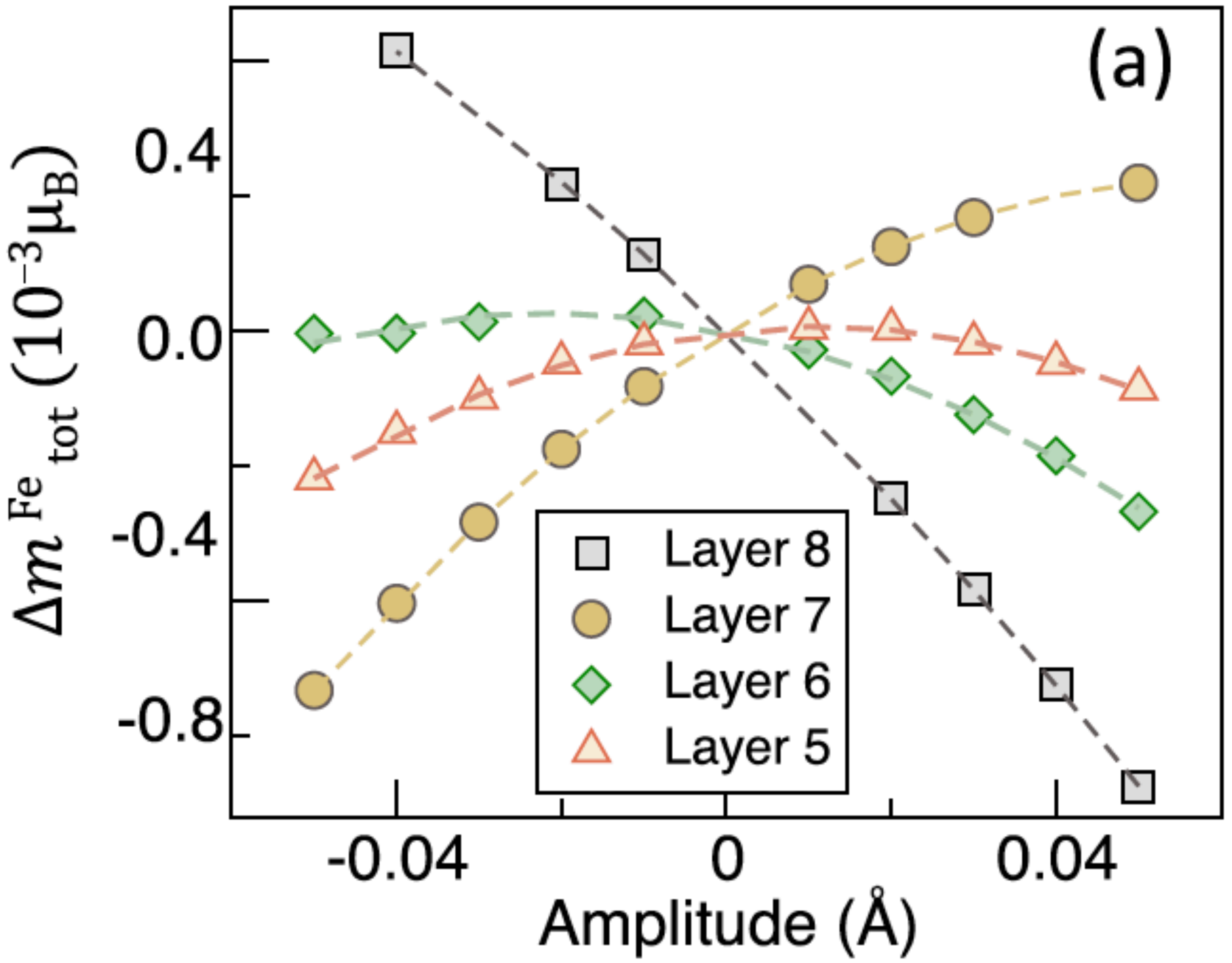


layer magnetization | electric dipole | local linear ME tensors

S. Bhowal, A. Urru, SFW and N. A. Spaldin,
Phys. Rev. Lett. **134**, 146703 (2025)

FeF$_2$





$\propto E^{applied}$

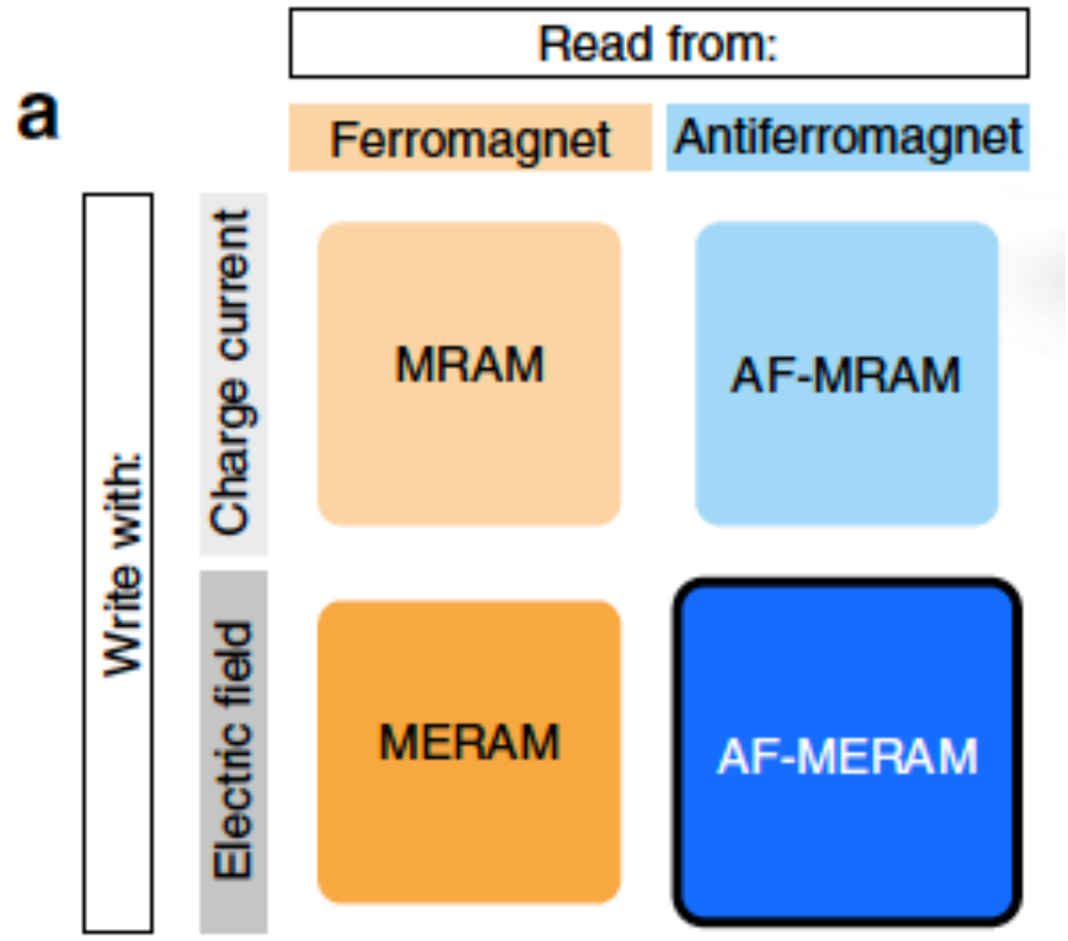
- E.g. quadratic ME FeF2; (110) surface has equilibrium surface magnetization, and a change in surface magnetization linear in an external electric field! (Surface *linear* ME effect)

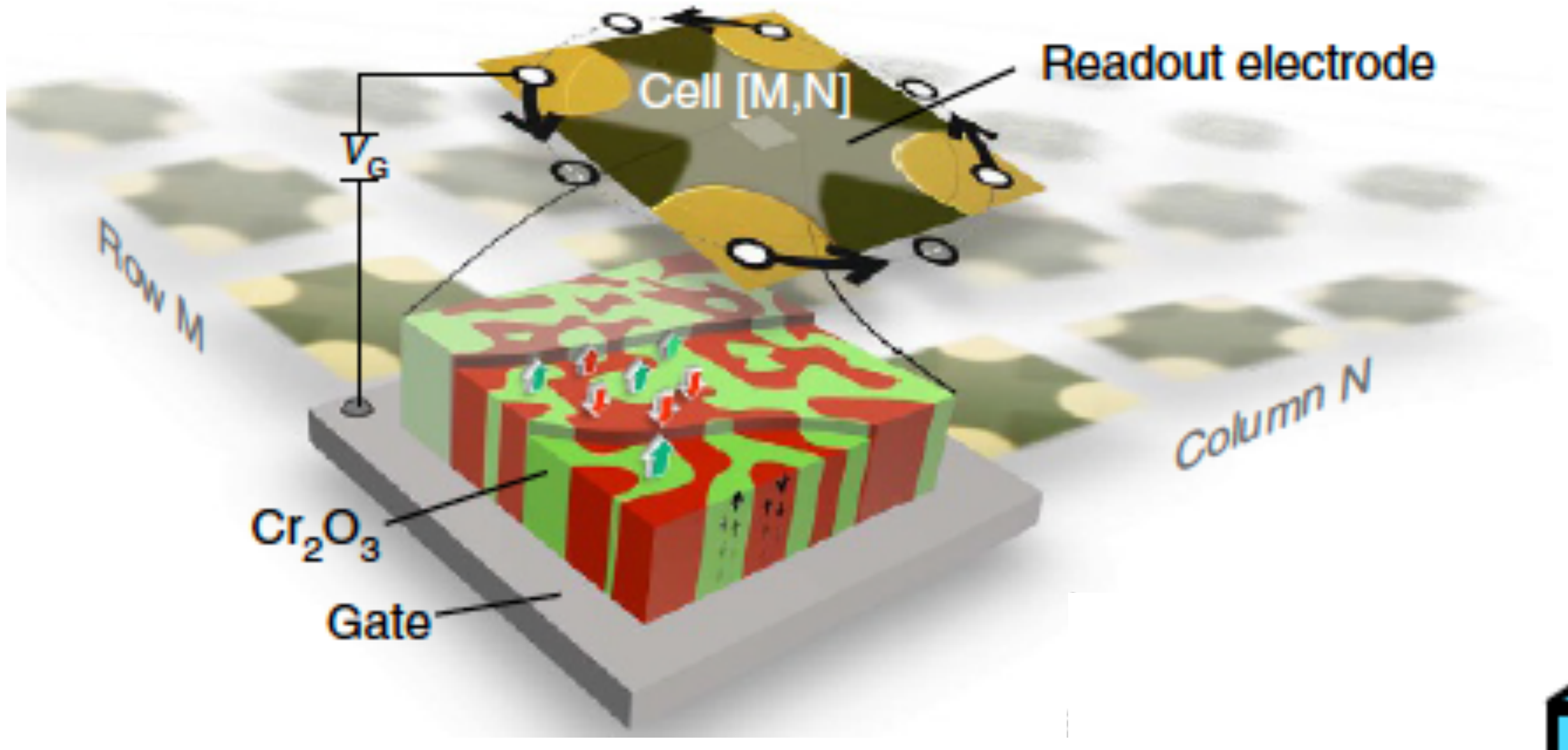
- Surface linear ME tensor α_{ij}^{surf} is proportional to effective electric field E^{surf}, $\alpha_{ij}^{surf} \propto \beta_{ijj}^{bulk} E_j^{surf}$

III. Outlook

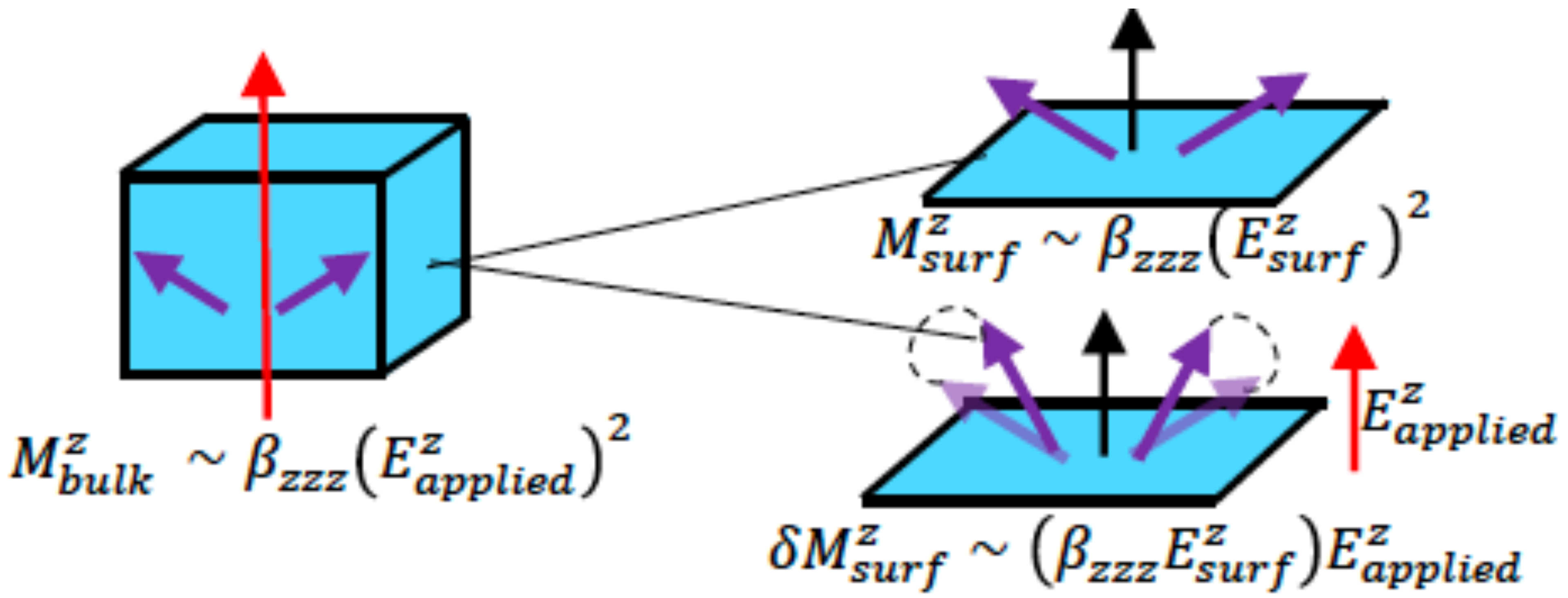
(a) Relevance for van der Waals materials

Some general spintronics utilities for surface magnetic properties



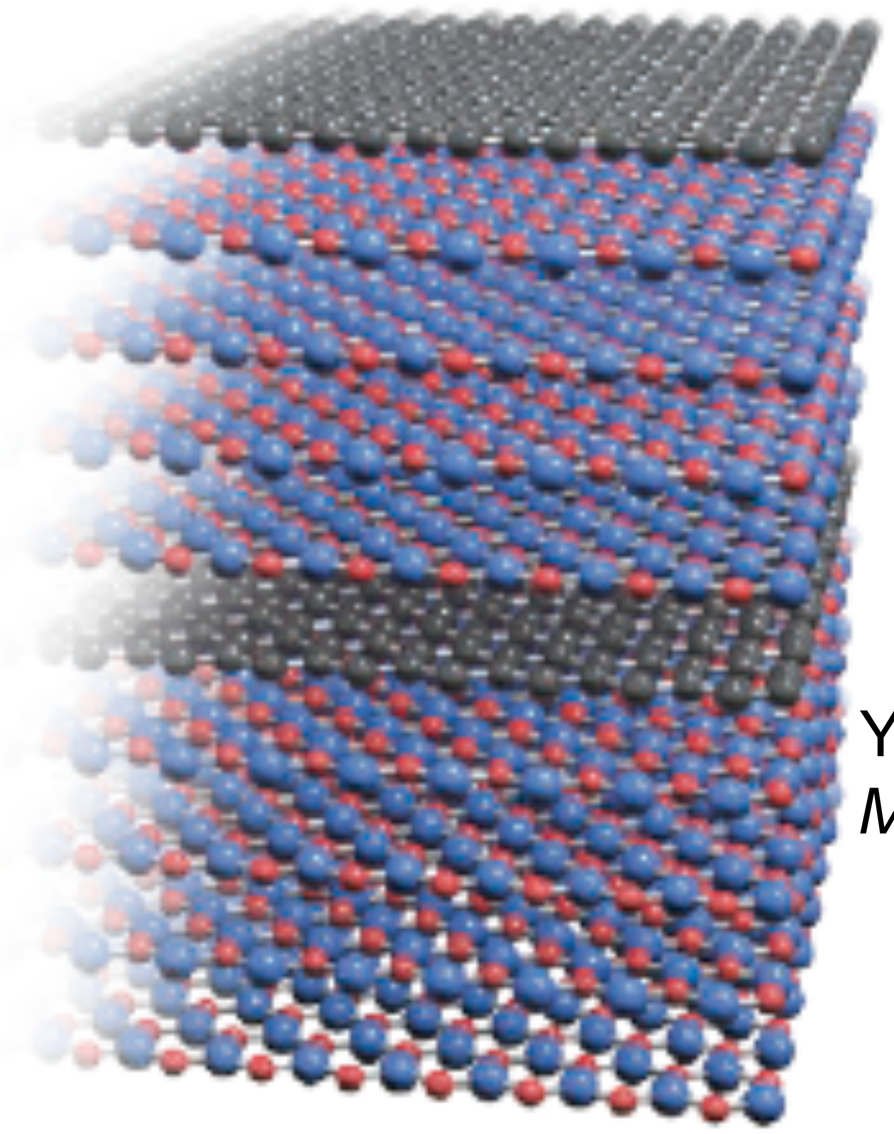


T. Kosub et al., *Nature Comm.* **8**, 13985 (2017)

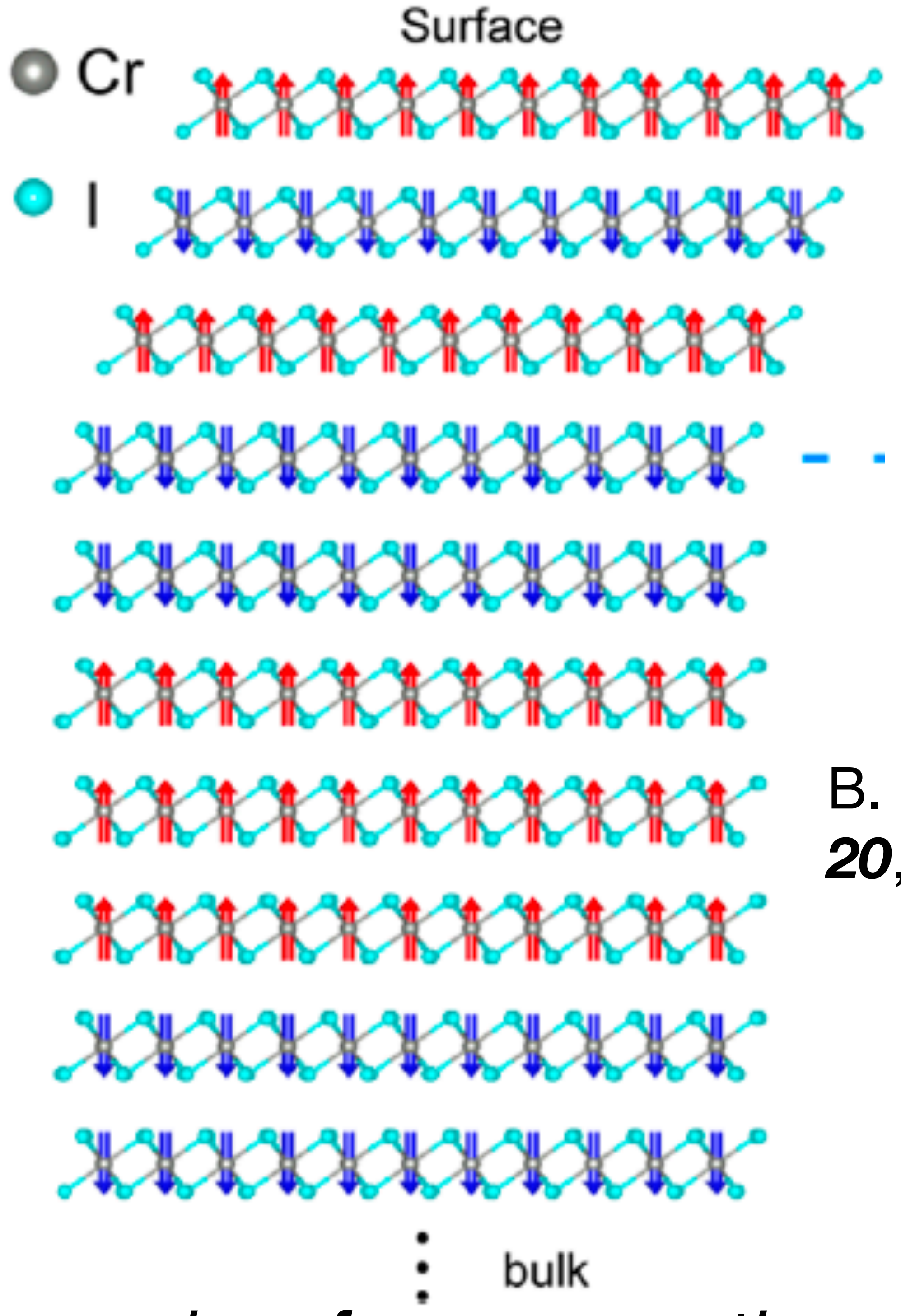


- Surface magnetization: domain readout in antiferromagnets, exchange bias…
- Surface Néel vector modifications: possible efficiency increases in spin-orbit torque (?!)
- Surface linear ME effects; enhanced magnitudes of magnetoelectric coupling

Symmetry guided search for layered ("bulk") van der Waals materials with surface magnetic properties



Y. Liu et al., *Nature Reviews Materials* **1**, 16042 (2016)



B. Niu et al., *Nano Lett. Materials* ***20***, 553-558 (2020)

- *Layered vdWs with space groups allowing for bulk ME responses will have discussed surface magnetic properties, analogously to covalent magnetic materials*

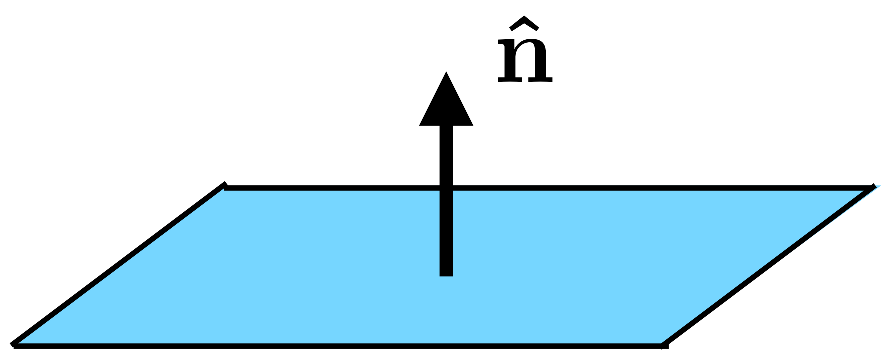
Strengths of vdW motif:

1. High tunability and control of synthesis can enable tailored design of desired bulk symmetries

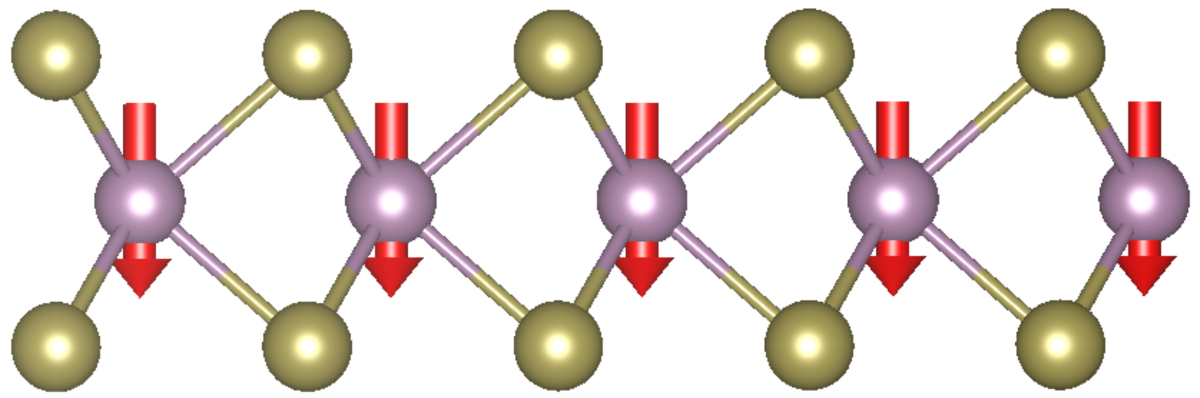
2. Ultra-clean, unreconstructed surfaces: enhanced responses

3. Quasi-2D nature; potential to decouple, and separately control, surface and "bulk" properties

Application to monolayer limit



$$g^{surf} \rightarrow g^{monolayer}$$



- Monolayer symmetries ("layer" point groups) are not equivalent to surface symmetries (e.g., a surface can never preserve inversion)
- But any surface point group can also be a monolayer point group (converse not true!):
- Provides strategy for symmetry-based search/design of "surface symmetry" van der Waals monolayers

Thank you!

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Currently hiring postdoc! (application deadline July 15th)

Postdoctoral position in prediction of low-dimensional magnetoelectrics

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The group of Sophie Weber (Condensed Matter and Materials Theory Division of Physics, Chalmers University of Technology) invites applications for a postdoctoral position on the first-principles-based prediction and design of new, low-dimensional magnetoelectric materials. Low-dimensional materials are highly desirable for efficient device integration, and the intrinsic coupling between electric polarization and magnetism in magnetoelectrics could revolutionize storage and information technologies. In this project, the applicant will combine symmetry analysis, electronic structure calculations, and high-throughput database screening to predict promising new material candidates. The position is for two years, with a start date of January 2026.