Reconstruction of ferroelastic domain patterns using single crystal diffraction: potential for DFXM

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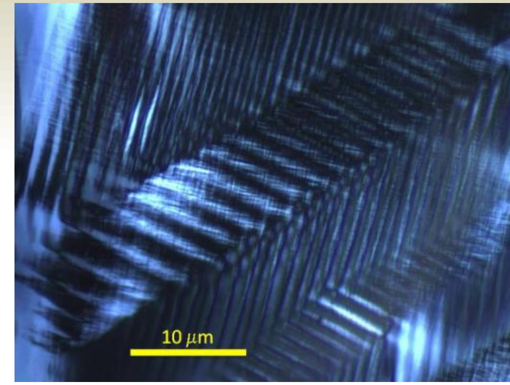
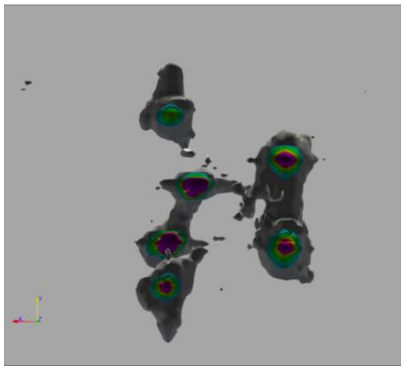
The content of the talk

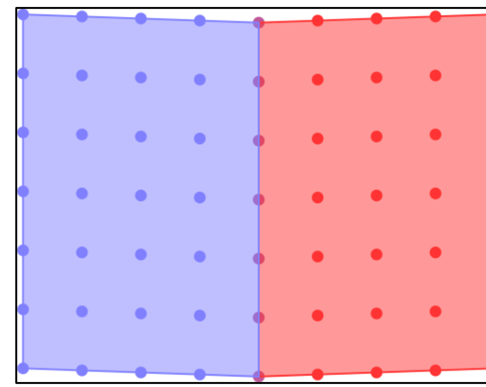


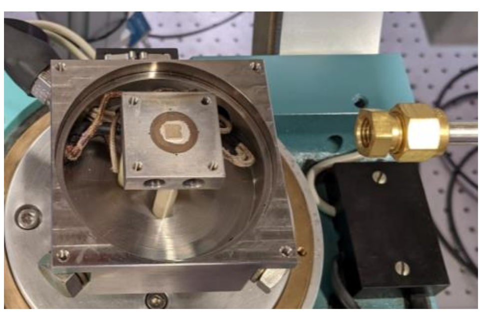
1. Ferroelastic domains. What and why?



2. Single crystal X-ray diffraction: how and if it can help in characterizing domain patterns

3. Mechanical compatibility of ferroelastic domains and prediction of diffraction patterns from them

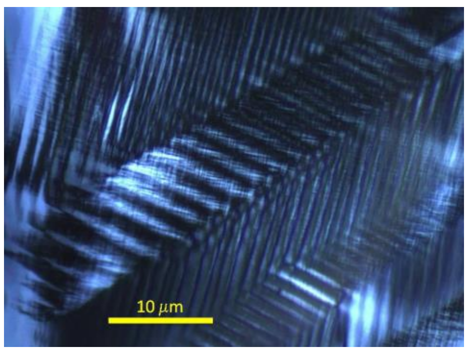


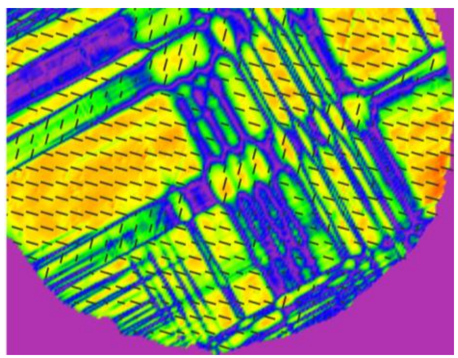
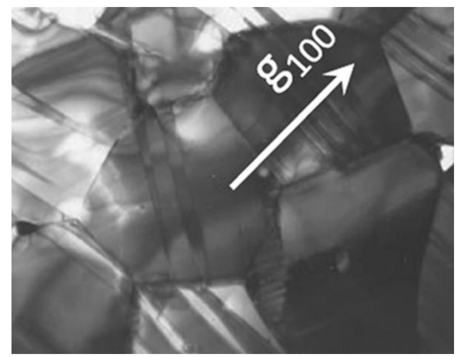


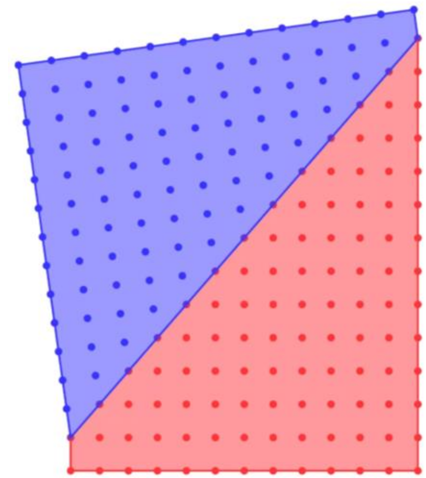
4. Diffraction-based recognition of ferroelastic domains in $BaTiO_3$.

Ferroelastic domains: how and why?

- *Ferroelastic domains result from a* symmetry-lowering phase transitions (e.g. from ***cubic*** to ***tetragonal*** / ***rhombohedral*** / ***monoclinic***). Their formation pursues the purpose of minimizing electrostatic / mechanical energy during the phase transition.





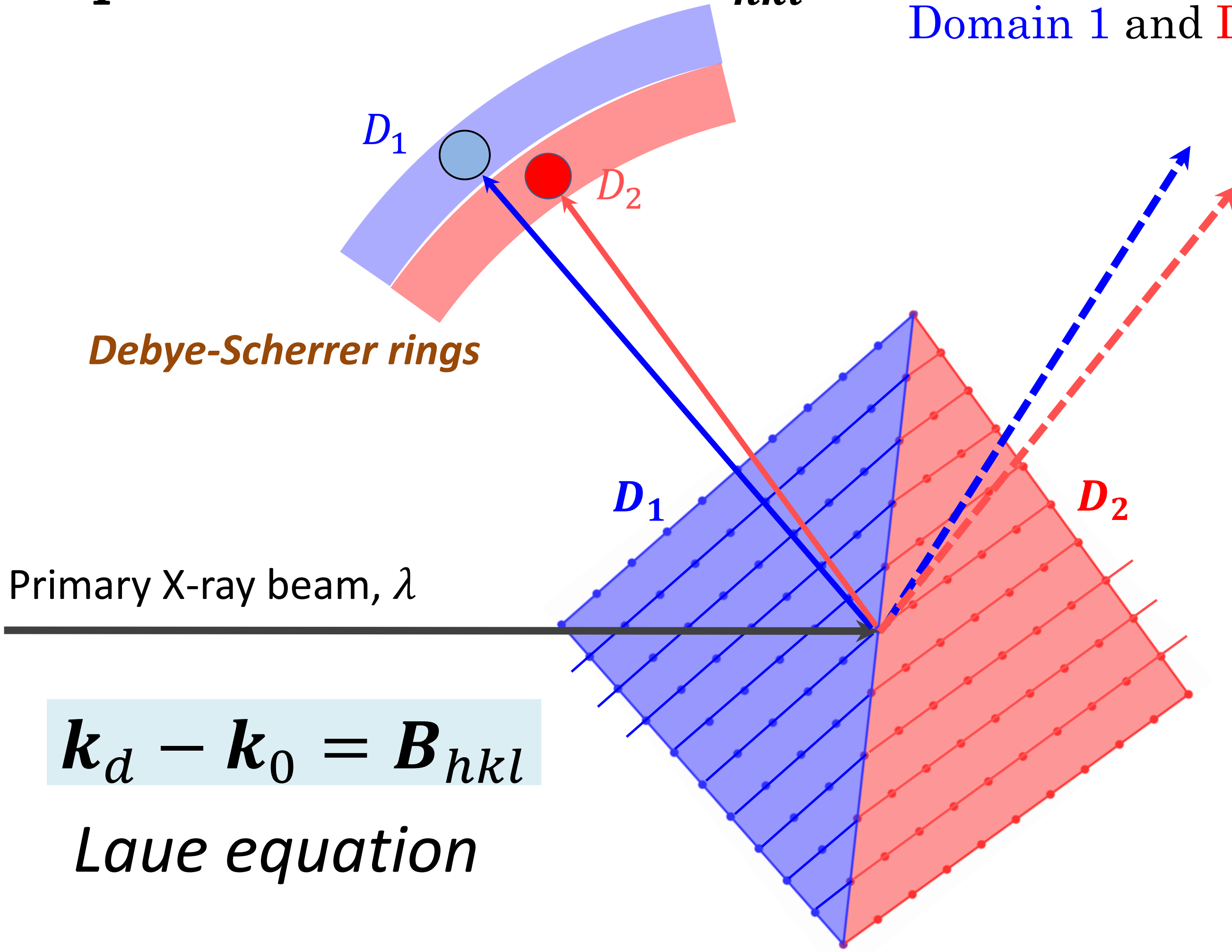
- Ferroelastic domains play major role in the enhancement of physical properties through domain wall motion.

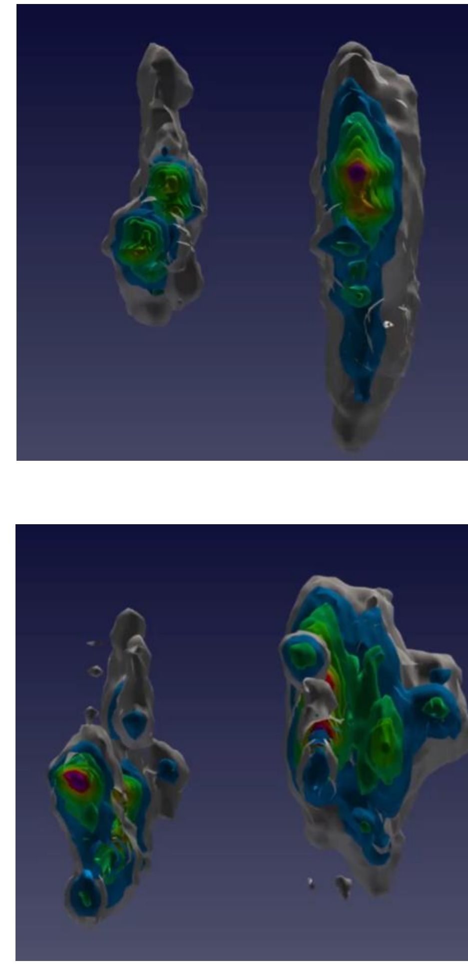
- Observation of ferroelastic domains and their dynamics is still a challenging task for which a handful of techniques exist only.

X-ray diffraction from multi-domain crystal

Reciprocal lattice vectors $\boldsymbol{B}_{hkl}$

X-ray beams diffracted from Domain 1 and Domain 2

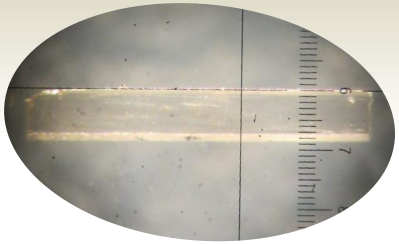


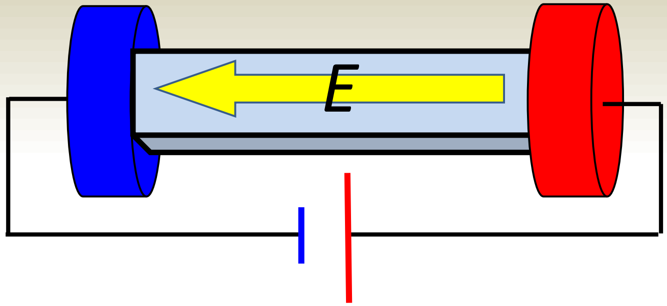


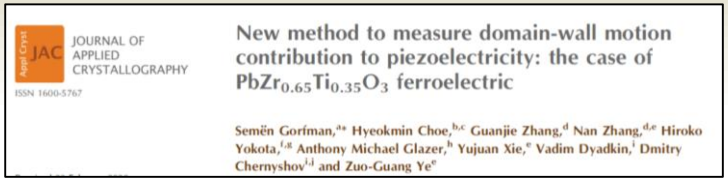
$$\boldsymbol{k}_d - \boldsymbol{k}_0 = \boldsymbol{B}_{hkl}$$

Laue equation

Reconstruction of reciprocal space



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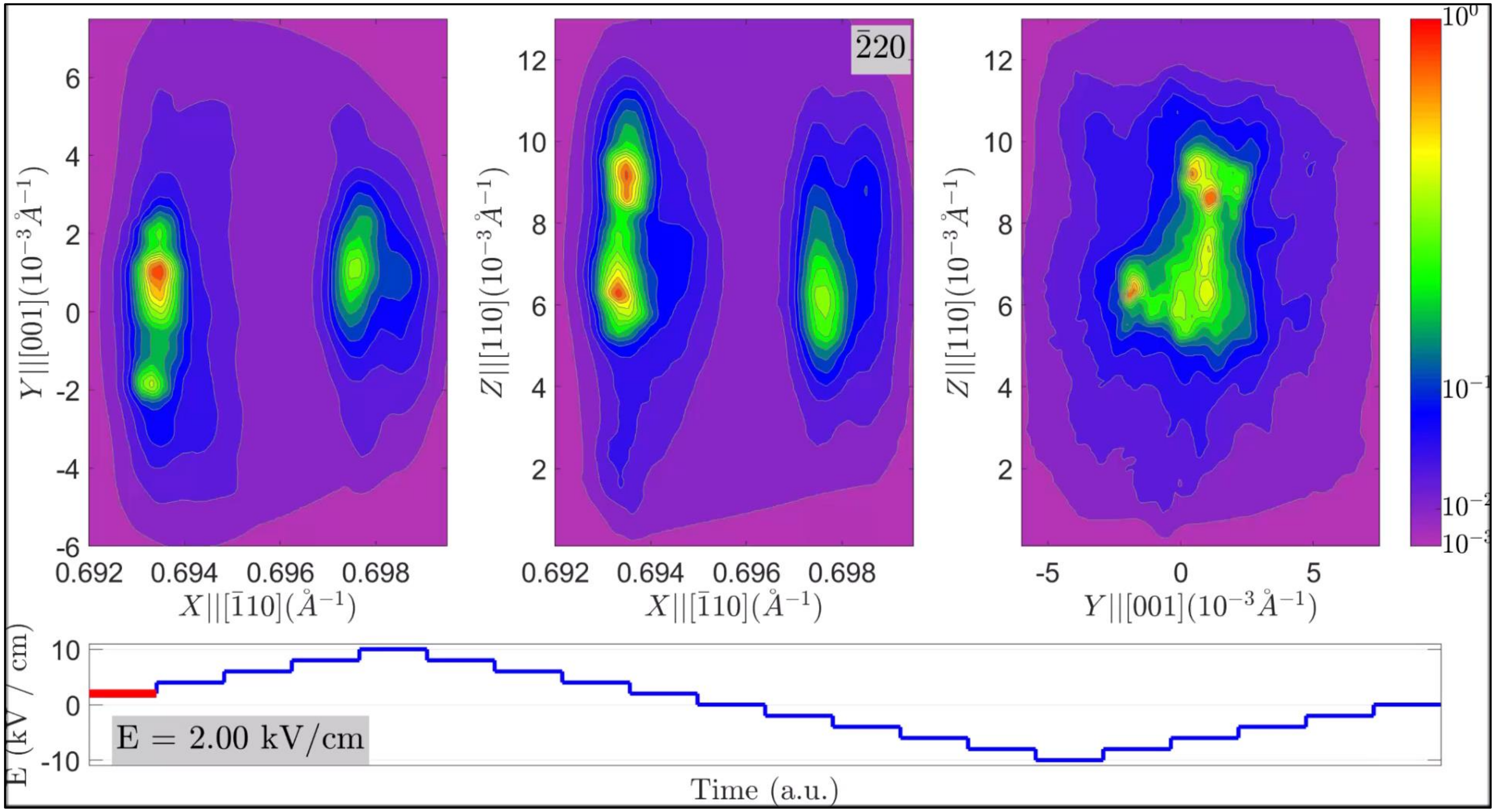
New method to measure domain-wall motion contribution to piezoelectricity: the case of $PbZr_{0.65}Ti_{0.35}O_3$ ferroelectric

Semën Gorfman,[a*] Hyeokmin Choe,[b,c] Guanjie Zhang,[d] Nan Zhang,[d,e] Hiroko Yokota,[f,g] Anthony Michael Glazer,[h] Yujuan Xie,[e] Vadim Dyadkin,[i] Dmitry Chernyshov[i,j] and Zuo-Guang Ye[e]



The aim of this work: formalism / program for recognition of domains in high-resolution reciprocal space maps

Key hypothesis: mismatch-free connection of domains

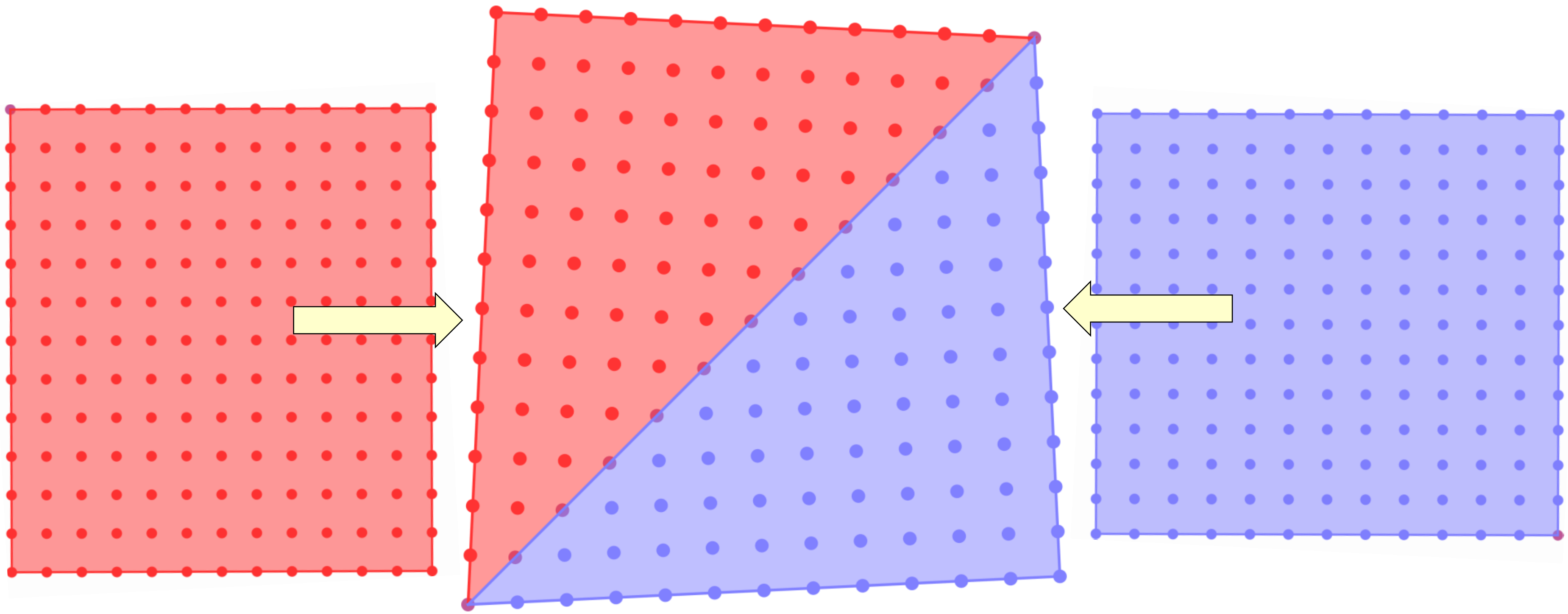
The Orientation of Domain Walls in Twinned Ferroelectric Crystals*

Jan Fousek†
Materials Research Laboratory, The Pennsylvania State University, Uni
and
Václav Janovec
Institute of Physics, Czechoslovak Academy of Sciences, Pr
(Received 1 August 1968)

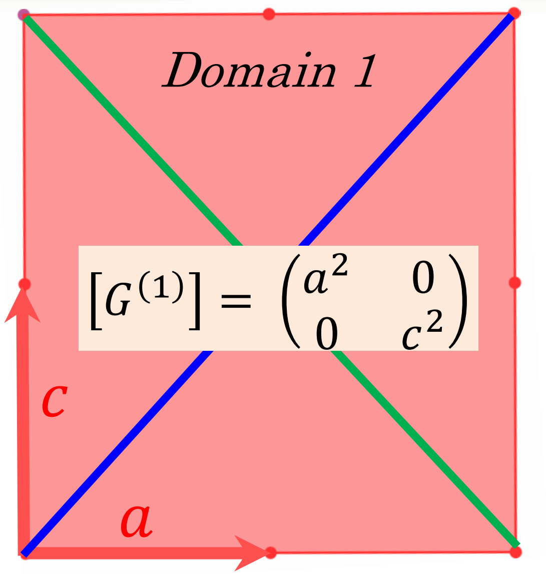
Domain-wall orientations in ferroelastics

J. Sapriel
Centre National d'Etudes des Télécommunications, 196 rue de Paris, 92220 Bagneux, France
(Received 12 March 1975)

Domains meet at the domain walls: such walls should be parallel to two-dimensional lattice planes, which allow mismatch-free connection.



On the theory of mismatch-free connection

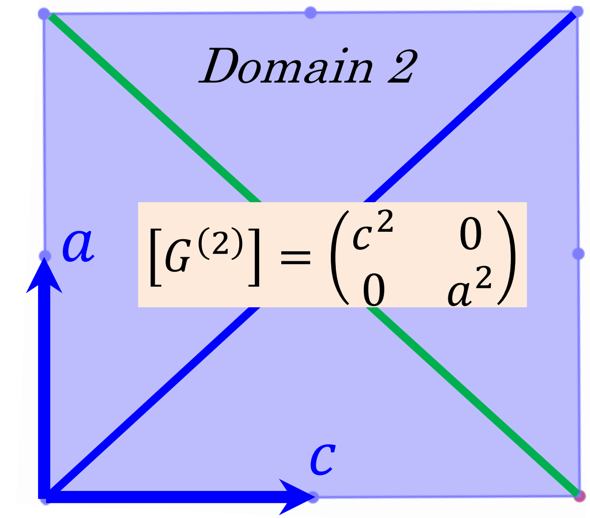


The matrix of dot product for each domain

$$G_{ij} = \boldsymbol{a}_i \boldsymbol{a}_j$$

$$[G^{(2)}] = [S][G^{(1)}][S]^T$$

$[S]$ is symmetry operation lost during the phase transition



$$(G_{ij}^{(2)} - G_{ij}^{(1)}) x_i x_j = 0$$

The equation for the mismatch-free boundary

For the 2D example above $(a^2 - c^2)(x_1^2 - x_2^2) = 0$ and two possible strain free boundaries

$(1\bar{1})$ plane $x_1 - x_2 = 0$

$x_1 + x_2 = 0$ (11) plane

Mismatch-free connection: more general aproach

Aims to reduce the equation $\Delta G_{ij}x_ix_j = 0$ to the equation of plane with the Miller indices (hkl): $hx_1 + kx_2 + lx_3 = 0$.

$$\begin{bmatrix} \Delta G_{11} & \Delta G_{12} & \Delta G_{13} \\ \Delta G_{12} & \Delta G_{22} & \Delta G_{23} \\ \Delta G_{13} & \Delta G_{23} & \Delta G_{33} \end{bmatrix} \xrightarrow[x_i = V_{ij}v_j]{\text{Transformation to the principal axes}} \begin{bmatrix} G_1 & 0 & 0 \\ 0 & G_2 & 0 \\ 0 & 0 & G_3 \end{bmatrix}$$

And for the case when e.g. $G_2 = 0,\ G_1G_3 < 0 \quad (*)$

$$G_1v_1^2 + G_3v_3^2 = 0$$

SOLUTION 1

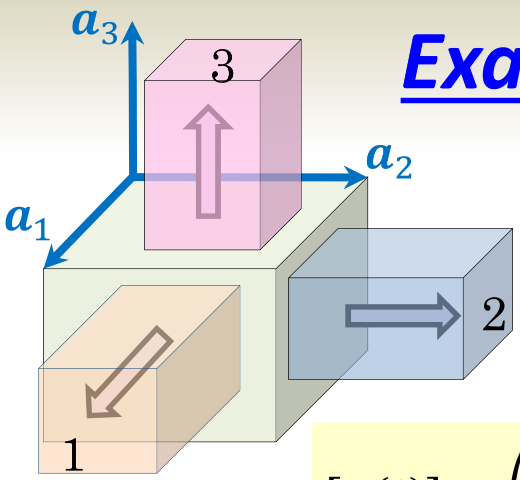
$$\sqrt{|G_1|}v_1 - \sqrt{|G_3|}v_3 = 0$$

SOLUTION 2

$$\sqrt{|G_1|}v_1 + \sqrt{|G_3|}v_3 = 0$$

Conclusion: any pairs of domains for which (*) is fulfilled can connect mismatch-free with each other via two possible interfaces

Examples: tetragonal domains

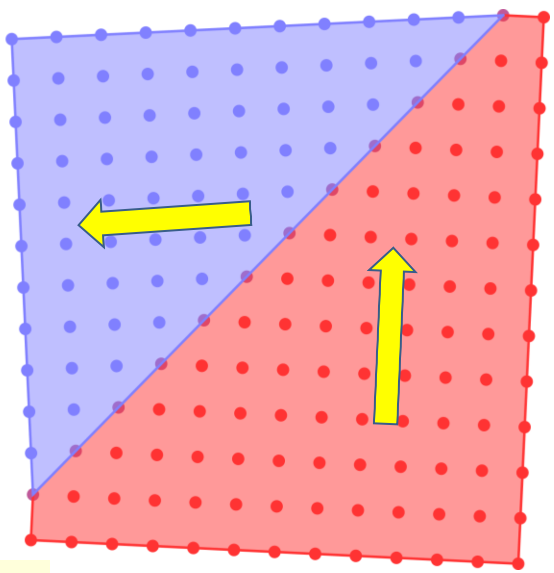


$$[G^{(2)}] = \begin{pmatrix} a^2 & 0 & 0 \\ 0 & c^2 & 0 \\ 0 & 0 & a^2 \end{pmatrix}$$

$$[G^{(1)}] = \begin{pmatrix} c^2 & 0 & 0 \\ 0 & a^2 & 0 \\ 0 & 0 & a^2 \end{pmatrix}$$

$$[G^{(3)}] = \begin{pmatrix} a^2 & 0 & 0 \\ 0 & a^2 & 0 \\ 0 & 0 & c^2 \end{pmatrix}$$

DW	1	2	(hkl)	$\angle(P_1P_2)$
1	1	2	(110)	~ 90
2	1	2	$(1\bar{1}0)$	~ 90
3	1	3	(101)	~ 90
4	1	3	$(10\bar{1})$	~ 90
5	2	3	(011)	~ 90
6	2	3	$(01\bar{1})$	~ 90



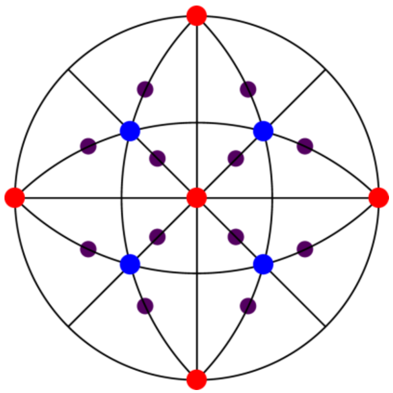
6 orientations of domain walls (90° DW)

More general case / domains of arbitrary symmetry

MATLAB-based script

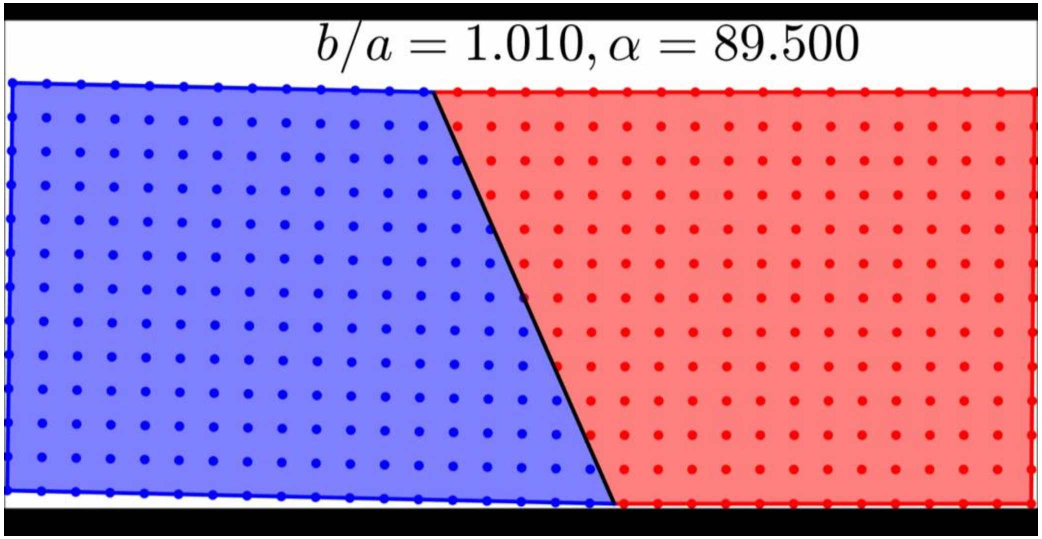
- ***Input**: free lattice parameters, symmetry operations of the parent phase*
- ***Output:** list of mismatch-free domain walls, twinning matrices, angles between polarization directions*

Example: (monoclinic domains, MA / MB)

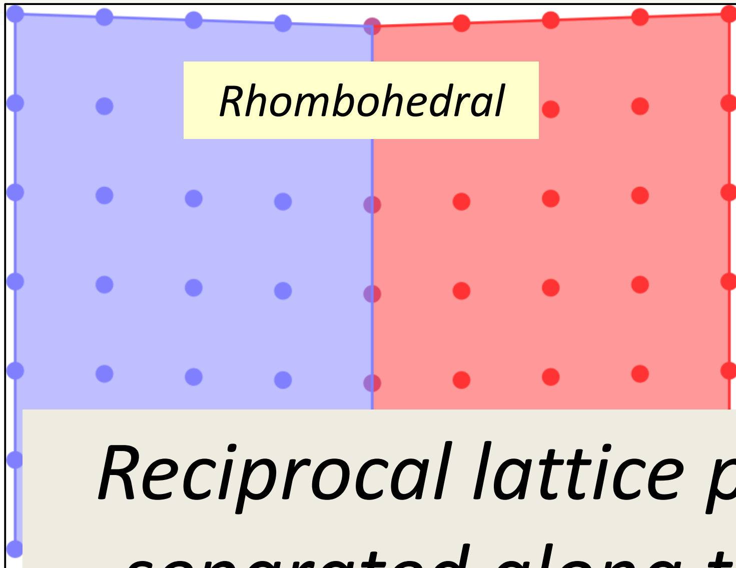


- ***12 domain variants***
- ***84 orientations of domain walls***
- ***48 Prominent domain walls (with fixed Miller-indices)***
- ***36 rotatable domain walls (The orientation depends on the lattice parameters)***

- ***Illustration of rotatable domain wall***
- ***The orientation changes with a lattice parameters***

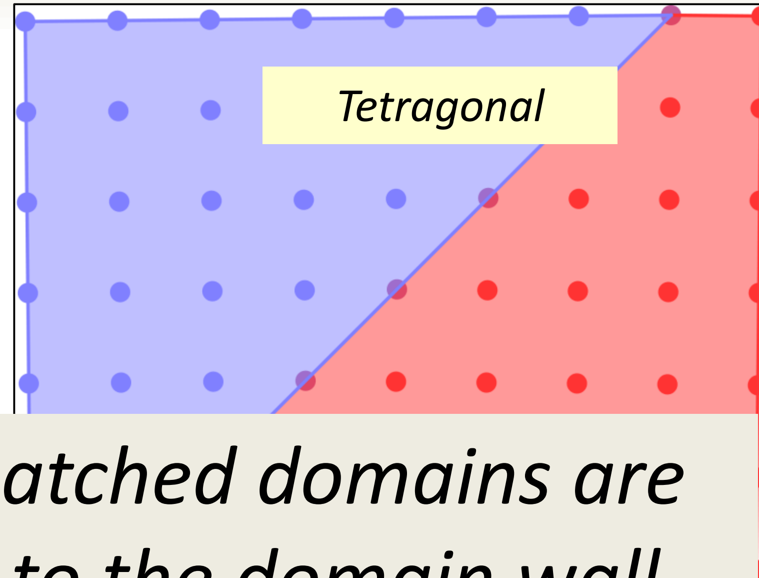


But what about reciprocal lattices? Examples first…

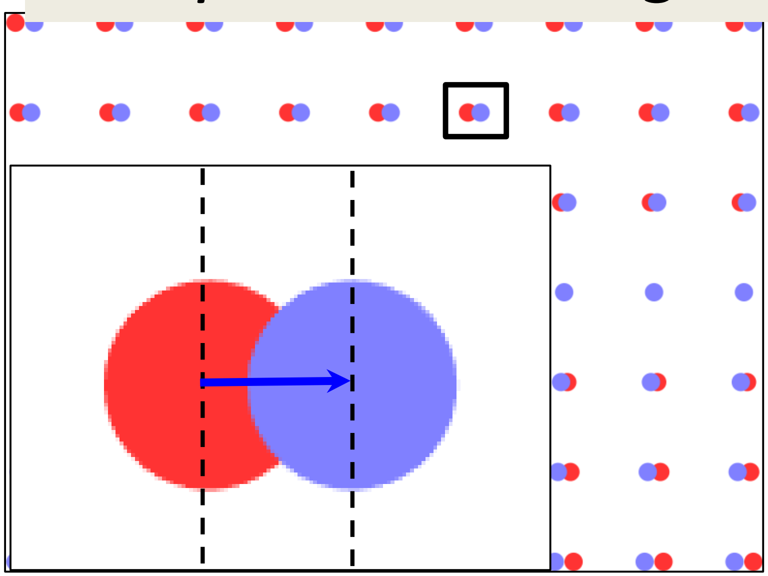


Real lattice

$\boldsymbol{a}_1\ \boldsymbol{a}_2\ \boldsymbol{a}_3$



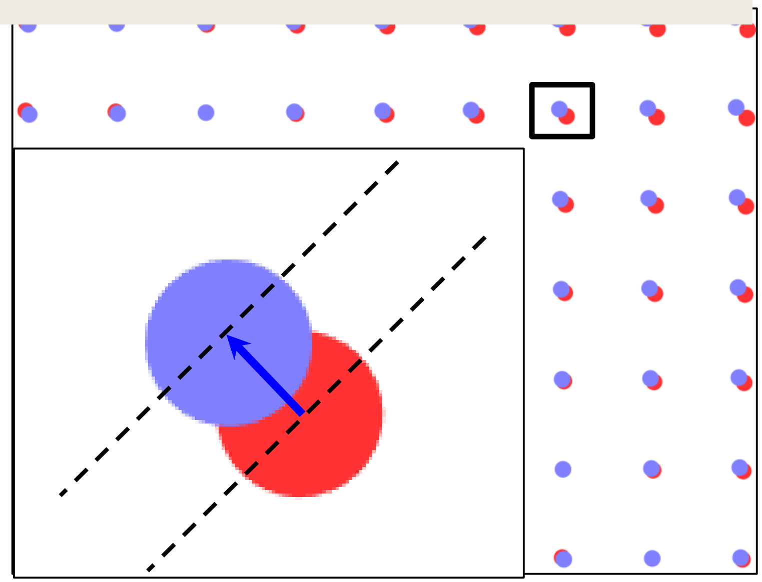
Reciprocal lattice points of matched domains are separated along the normal to the domain wall



Reciprocal lattice

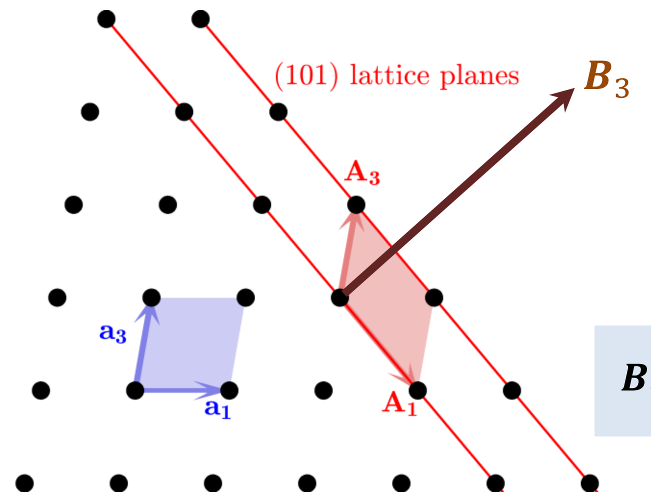
$\boldsymbol{b}_1\ \boldsymbol{b}_2\ \boldsymbol{b}_3$

$\boldsymbol{a}_i \boldsymbol{b}_j = \delta_{ij}$



Twinning relationship between reciprocal lattice vectors

- Assume that domains match along (hkl) plane
- The lattice basis vectors $\boldsymbol{a}_i^{(m)}$ $(i = 1 \dots 3,\ m = 1,2)$
- Transform them to $\boldsymbol{A}_i^{(m)}$ (so that $\boldsymbol{A}_{1,2}^{(m)} \parallel (hkl)$)



Twinning matrix

$$\left(\boldsymbol{A}_1^{(2)} \quad \boldsymbol{A}_2^{(2)} \quad \boldsymbol{A}_3^{(2)}\right) = \left(\boldsymbol{A}_1^{(1)} \quad \boldsymbol{A}_2^{(1)} \quad \boldsymbol{A}_3^{(1)}\right)\begin{pmatrix} 1 & 0 & y_1 \\ 0 & 1 & y_2 \\ 0 & 0 & y_3 \end{pmatrix}$$

Using relationship $\boldsymbol{A}_i \boldsymbol{B}_j = \delta_{ij}$ we get

$$\boldsymbol{B}_1^{(2)} = \boldsymbol{B}_1^{(1)} - \frac{y_1}{y_3}\boldsymbol{B}_3^{(1)} \qquad \boldsymbol{B}_2^{(2)} = \boldsymbol{B}_2^{(1)} - \frac{y_2}{y_3}\boldsymbol{B}_3^{(1)} \qquad \boldsymbol{B}_3^{(2)} = \frac{1}{y_3}\boldsymbol{B}_3^{(1)}$$

research papers

Algorithms for target transformations of lattice basis vectors

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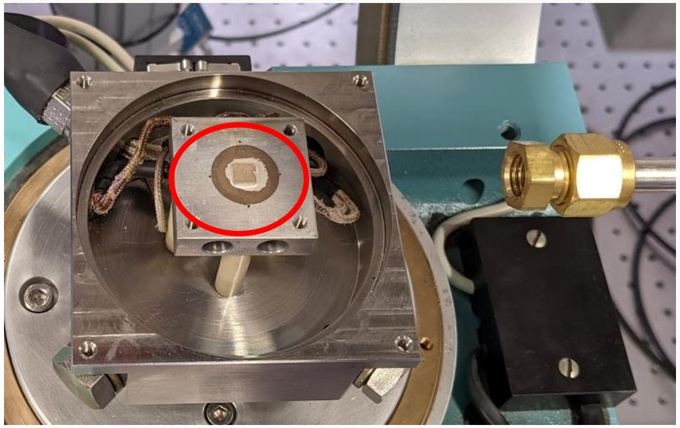
The splitting of reflection with the indices H, K, L (relative $\boldsymbol{B}_i^*$)

$$\Delta \boldsymbol{H} = H\Delta\boldsymbol{B}_1 + K\Delta\boldsymbol{B}_1 + L\Delta\boldsymbol{B}_3 = \left(-H\frac{y_1}{y_3} - K\frac{y_2}{y_3} + L\frac{1-y_3}{y_3}\right)\boldsymbol{B}_3^{(1)}$$

Conclusion: *Peaks are separated along the line, that is perpendicular* to the domain wall.

High-resolution reciprocal space mapping of $BaTiO_3$

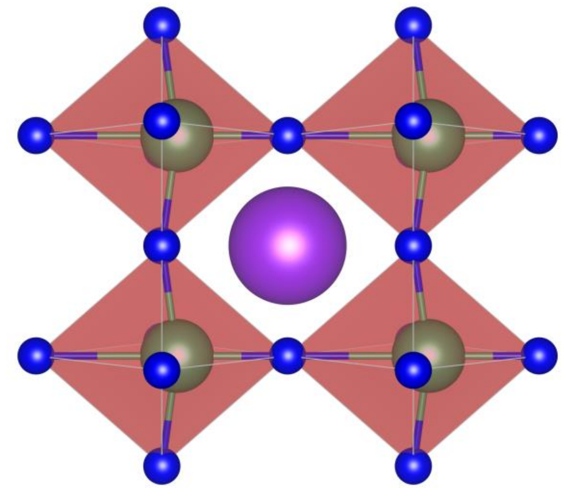


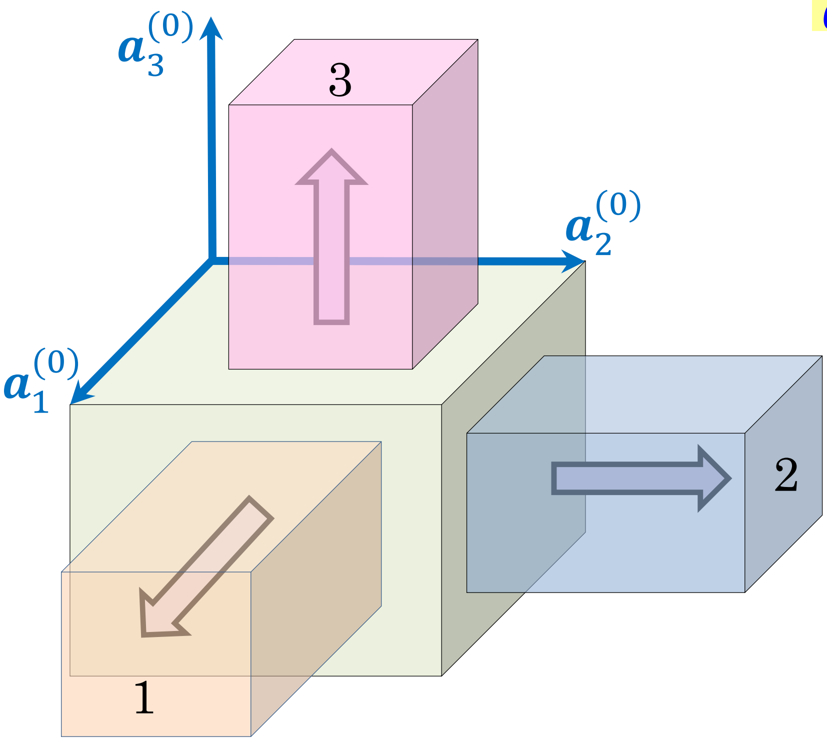
Space group

$P4mm$

Tetragonality:

$\frac{c}{a} = 1.011$





1 (a domain): $\boldsymbol{P}$ ‖ [100]

2 (b domain): $\boldsymbol{P}$ ‖ [010]

3 (c domain): $\boldsymbol{P}$ ‖ [001]

High-resolution reciprocal space mapping of $BaTiO_3$

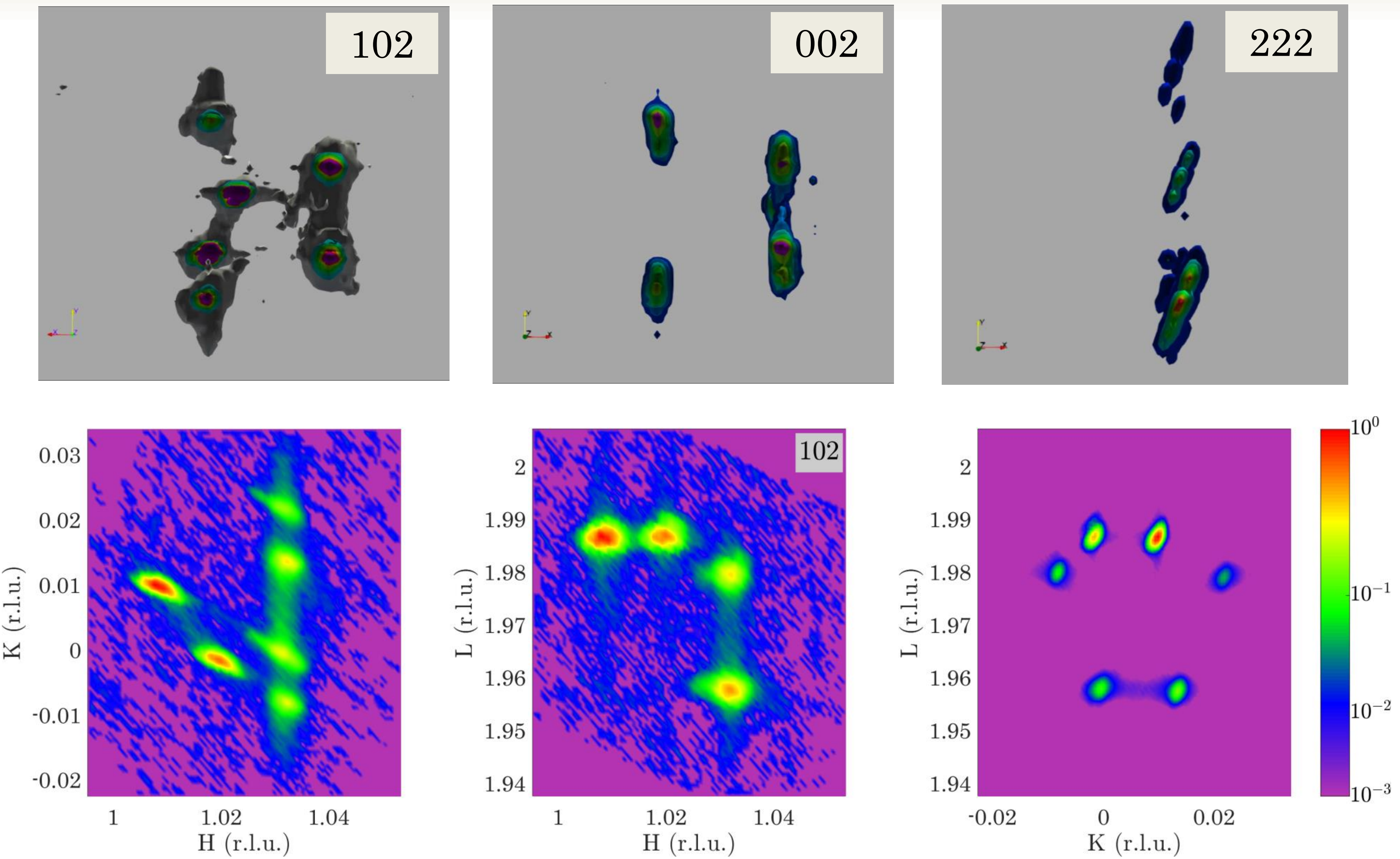
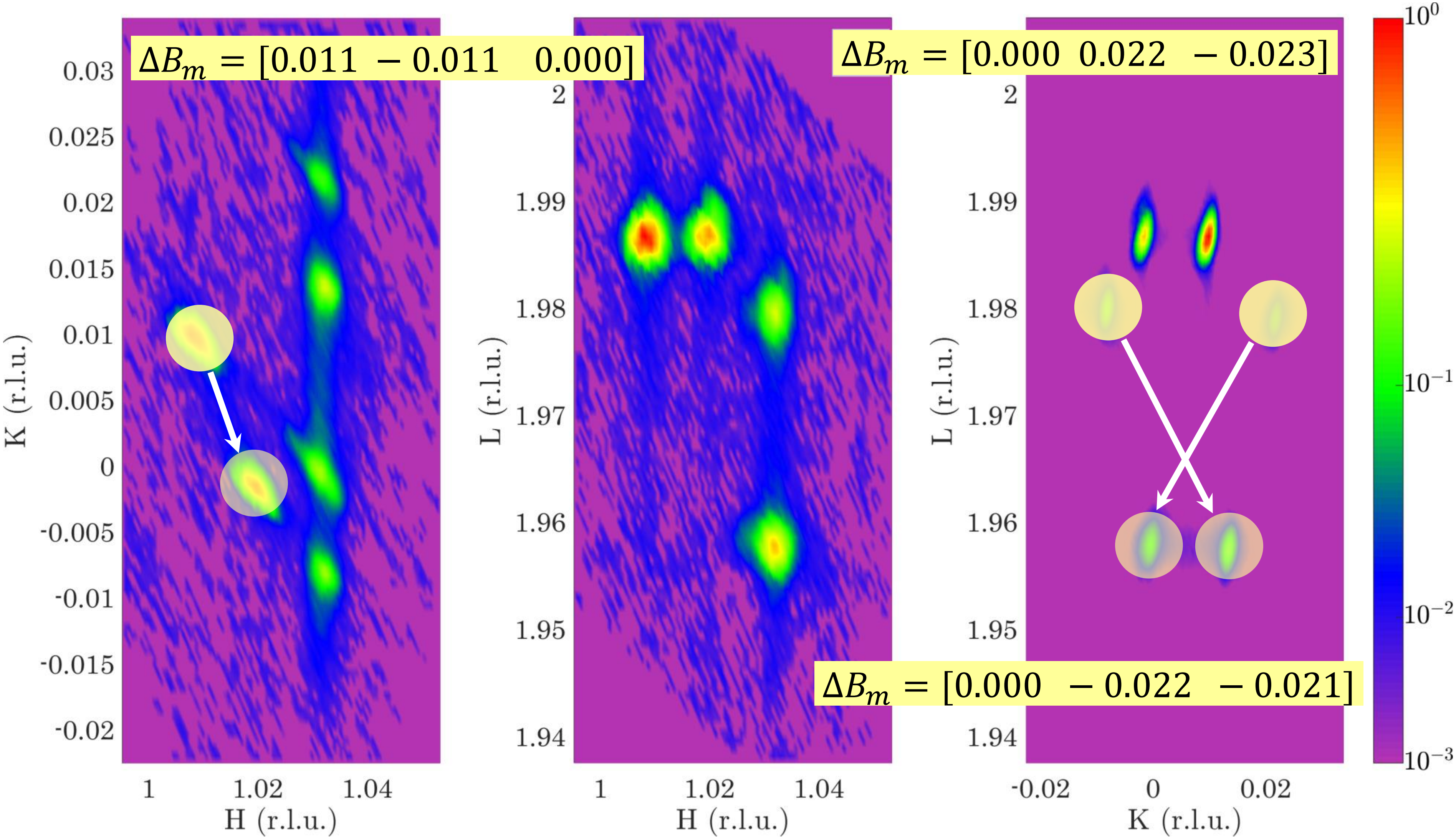
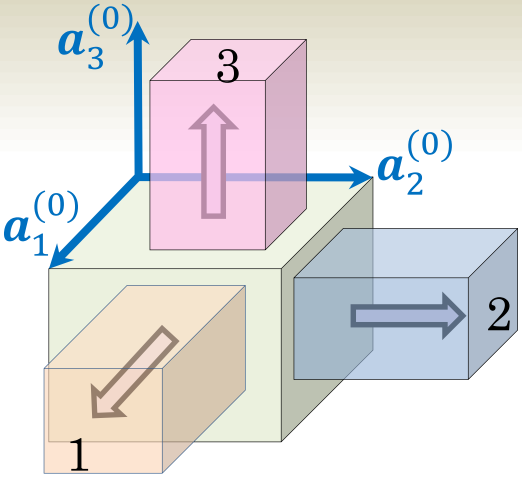
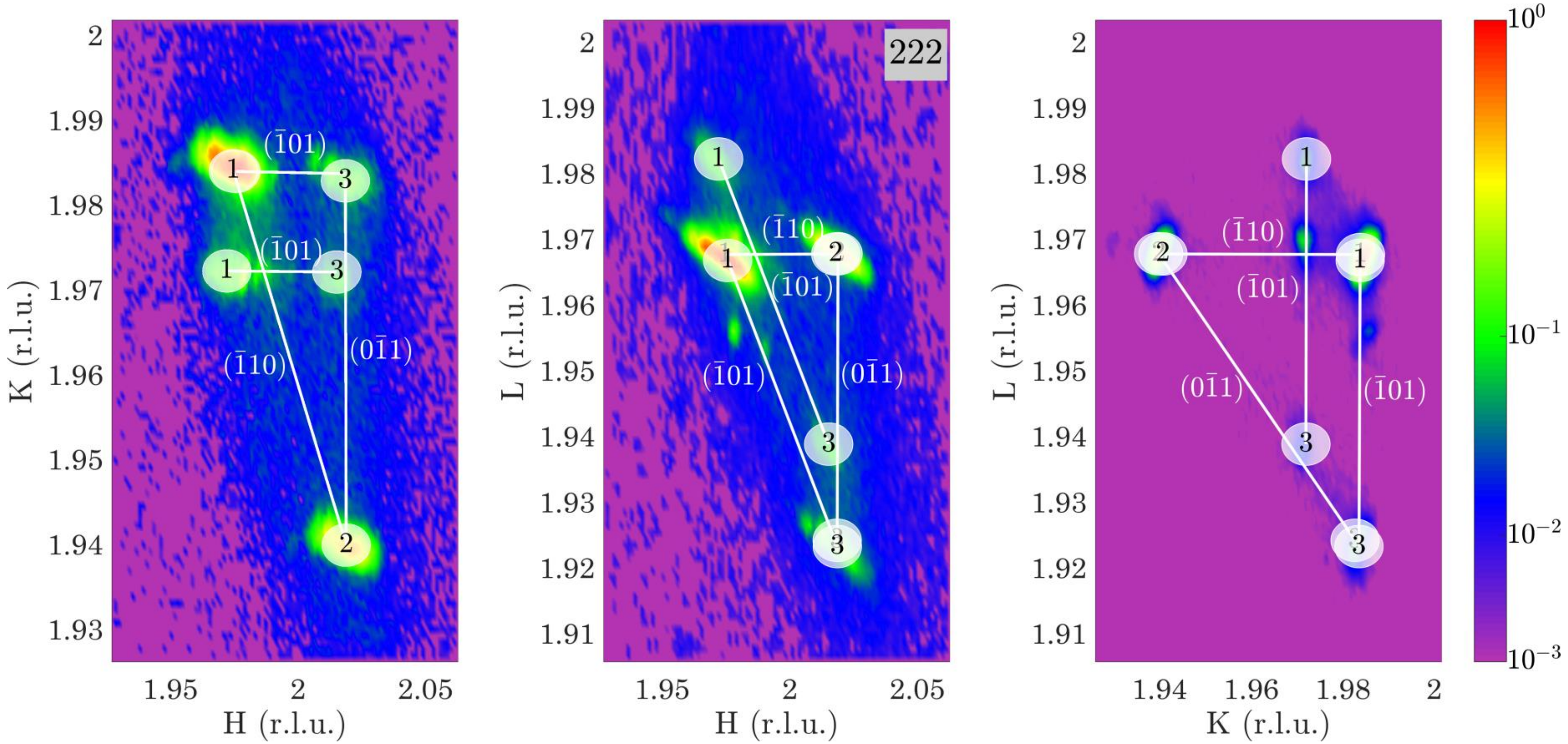


Illustration of the recognition procedure

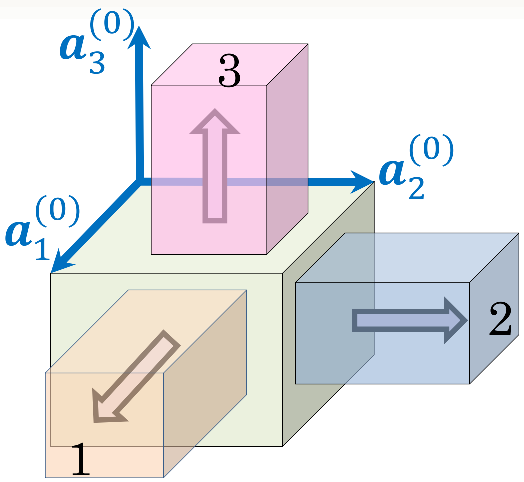


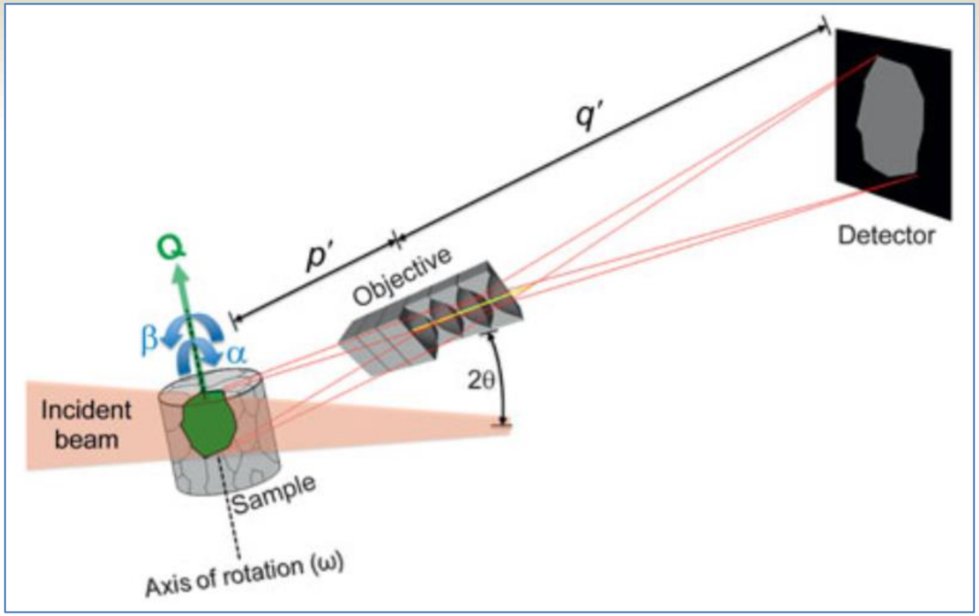


Results: recognition of domains in $BaTiO_3$



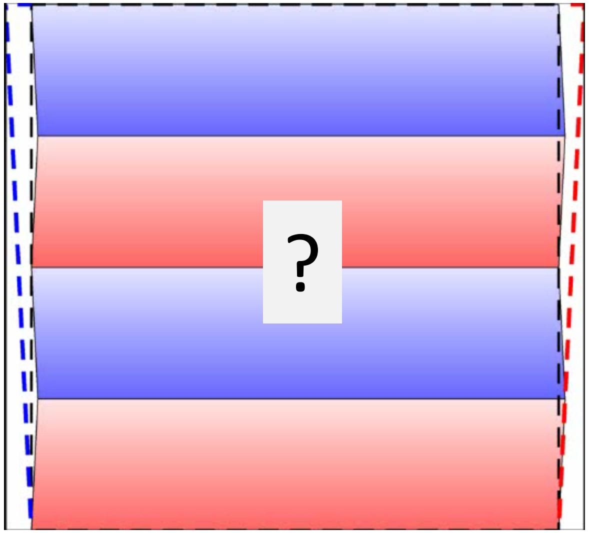
Potential for DFXM



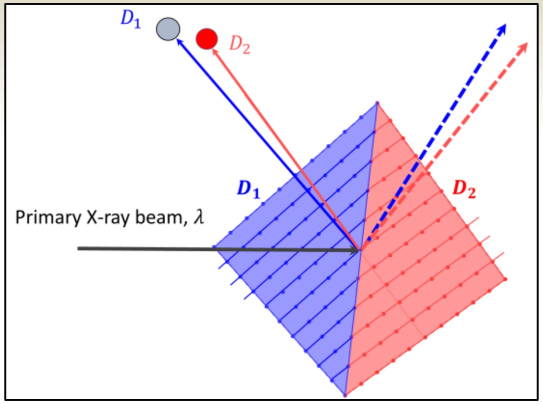


The combination of reciprocal and real space imaging is mutually beneficial because

- *Is condition of mechanical compatibility really fulfilled?*
- *What is the real 3D topology of domain structures?*
- *What is the shape and size of domain walls?*

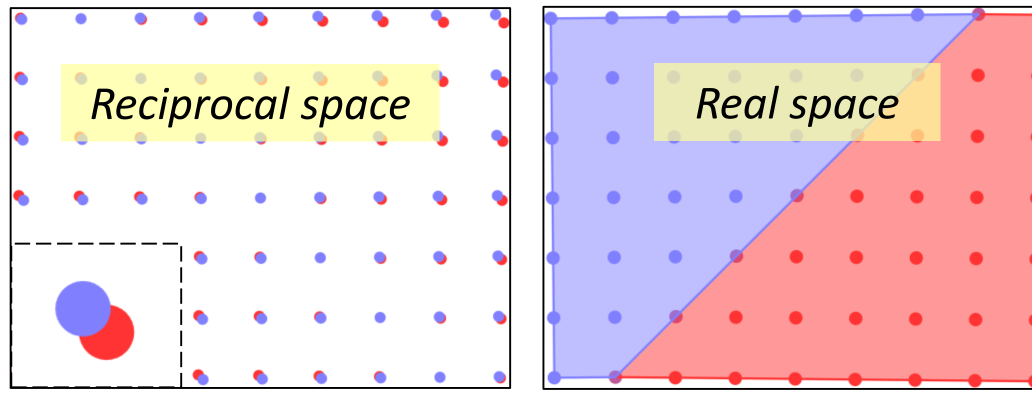


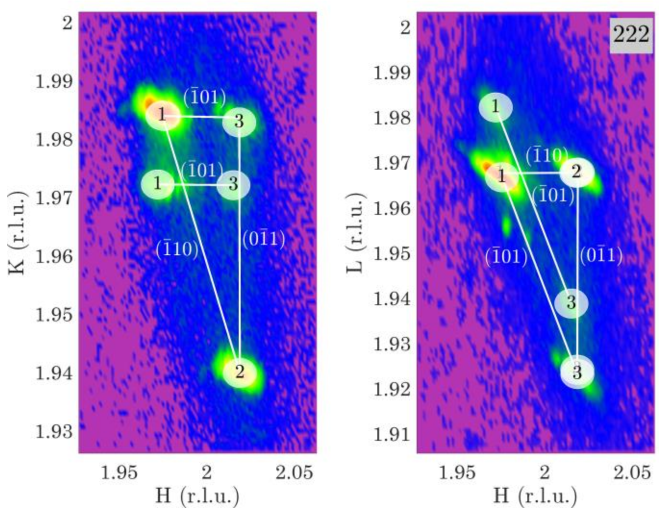
Conclusions



- *Single-crystal X-ray diffraction offers several advantages for observation of ferroelastic domains (penetration depth, sensitivity to the lattice parameters and domains orientation).*

- *We applied the theory of permissible domain walls to calculate possible separation of Bragg peaks.*





- *We tested the algorithm for the identification of peak pairs in single crystal $BaTiO_3$.*

- *The method will be applied to follow the response of different domains under external perturbation (e.g. electric field and temperature)… To be continued…*

הקרן הלאומית למדע
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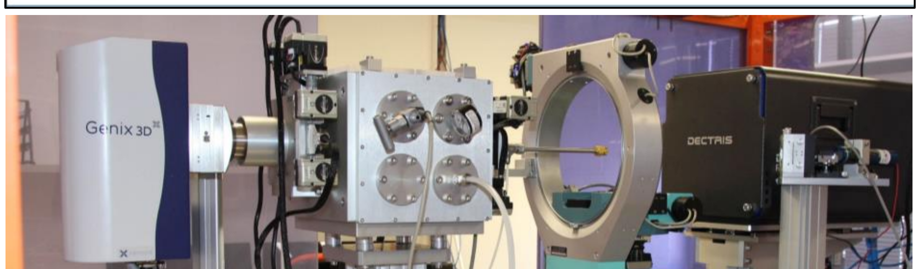

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***Technion* (Israel)**

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