



Crystallographic texture changes during martensitic transformations: X-ray and neutron diffraction studies and modeling

Roman Vasin

Frank Laboratory of Neutron Physics Joint Institute for Nuclear Research



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The layout

- 1. (Brief) introduction to crystallographic textures
- 2. A "good" example of transformation texture case
- 3. A "bad" example (or merely unfinished)?
- 4. Plans for experiments at the IBR-2 (FLNP JINR, Dubna)

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Crystallographic texture

Many materials (metals, alloys, ceramics, rocks,...) are polycrystals, i.e. they consist of grains (crystallites) of different shape, size and orientation.

Crystallographic texture is the lattice (or crystallographic) preferred orientation of crystallites of one crystalline phase (e.g., one or other mineral in case of the polymineral rock) in the chosen (macroscopic) coordinate system.

Common abbreviation: LPO (or CPO).



Random/chaotic orientation: NO crystallographic texture



Aligned grains: One-component crystallographic texture (≈ single crystal)



Multi-component crystallographic texture (different colors = different orientations, not different phases!)

Orientation – Euler angles

It is necessary to describe the transformation (rotation) $K_A \rightarrow K_B$ 1) Three Eulerian angles:

Roe-Matthies variant:

 $K_A \rightarrow K_A$ ' (rotation of K_A around Z_A : $0 \le \alpha < 2\pi$ to bring Y_A to $X_B Y_B$ plane) $K_A' \rightarrow K_A''$ (rotation of K_A ' around Y_A' : $0 \le \beta \le \pi$ to bring $Z_A \parallel Z_B$) $K_A'' \rightarrow K_B$ (rotation of K_A'' around Z_A'' : $0 \le \gamma < 2\pi$)





Roe R.-J. // J. Appl. Phys. 1965. V. 36. P. 2024.

Matthies S., Wenk H.-R., Vinel G.W.// J. Appl. Crystallogr. 1988. V. 21. P. 285.

Roe's definition: $\psi \equiv \alpha, \theta \equiv \beta, \phi \equiv \gamma$

 K_B possesses the orientation $g \equiv g^{B \leftarrow A} = \{\alpha, \beta, \gamma\}.$

Inverse operation $(K_B \rightarrow K_A) g^{-1} \equiv g^{A \leftarrow B} = \{\alpha, \beta, \gamma\}^{-1} = \{-\gamma, -\beta, -\alpha\} = \{\pi - \gamma, \beta, \pi - \alpha\}.$

Orientation distribution function (ODF)

Each crystallite in polycrystal possesses its own orientation g.

Let dV be the total volume of crystallites with orientation g within dg interval, and let V be the total volume of the polycrystal.

The 3D ODF f(g) is defined as

s defined as
$$\frac{dV}{V} = \frac{1}{8\pi^2} f(g) dg$$
$$\frac{1}{8\pi^2} \int_G f(g) dg = \frac{1}{8\pi^2} \int_0^{2\pi} d\alpha \int_0^{\pi} \sin\beta d\beta \int_0^{2\pi} f(\{\alpha, \beta, \gamma\}) d\gamma$$

ODF is a probability density for a polycrystal to contain a volume with a certain orientation within the range from g to g + dg.

Extreme cases:

random distribution (chaotic/random texture, no texture, "perfect powder")

$$f(g) = 1$$

«single-crystal-like» (the same orientation g_0 of all crystallites)

$$f(g) = 8\pi^2 \delta(g - g_0)$$



= 1

Pole figures (PFs) and inverse pole figures (IPFs)

Pole figures and inverse pole figures are introduced when it is necessary to know the orientations of crystallographic direction \mathbf{h}_i – usually, the normal to some crystallographic plane (hkl) – relative to K_A or some "unique" sample direction \mathbf{y} relative to K_B .

- \checkmark satisfy rotation symmetries of sample and crystallite
- ✓ non-negative
- ✓ 0 on PF/IPF means all ODF values along the integration path are equal to 0
- \checkmark normalized to unity





Austenite steel (surface coating on ferrite VVER-1000 reactor vessel), FCC, space group Fm $\overline{3}$ m. Z_A is normal to the vessel surface and ferrite/austenite boundary. Equal area projections.

Сумин В.В., Васин Р.Н., Папушкин И.В. и др. // Атомная энергия. 2011. Т. 110. №2. С. 78.

Measuring the texture (ODF) in diffraction experiment Integrated diffraction peak intensity is proportional to the volume of crystallites in Bragg condition:



A peculiar tomography problem: recalculate the 3D ODF with the best possible angular resolution based on finite number of (incomplete) PFs.

Bulk physical properties of polycrystals

Single crystal elasticity \rightarrow polycrystal elasticity



https://doi.org/10.1016/j.carbon.2010.12.003

Texture formation

Experimental pole figures (111) of the oxygen-free high-conductivity (OFHC) copper, $T = 25^{\circ}$ C, strain rate = 0.001 s⁻¹. Equal area projections.

Texture forms or changes due to:

- Crystallization
- Recrystallization
- Plastic deformation
- Twinning
- Sedimentation
- Structural phase transitions
- In particular, diffusionless martensitic transformations with definite orientation relationships between phases



C.A. Bronkhorst et al. (1991). Textures and microstructures. https://doi.org/10.1155/TSM.14-18.1031

$\alpha \leftrightarrow \beta$ quartz transformation



H.-R. Wenk et al. (2009). Phys. Chem. Minerals. https://doi.org/10.1007/s00269-009-0302-6

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S. Tateno et al. Science, 330, 359-361, 2010.



Plastic deformation of iron



rDAC experiment: Fe-9wt%Si

Sample in DAC loaded under nitrogen atmosphere



ALS 12.2.2



Fe-7.9wt%Si (*J.-F. Lin et al. Science, 295(5553), 313-315, 2002*)

Fe-9wt%Si (*R.A. Fischer et al. Earth Planet.Sci.Lett.*, *373*, *54-64*, *2013*)



Burger's orientation relationship





The calculation is straightforward!

- 1) For each $f(g^{BCC})$ value change $g^{BCC} \rightarrow g^{HCP}$ (all g^{HCPsym} should be considered) to get $f(g^{HCP})$
- 2) Interpolate $f(g^{\text{HCP}})$ into the same grid as $f(g^{\text{BCC}})$
- 3) Normalize $f(g^{\text{HCP}})$ to unity.

Quantifying the transformation texture



Redistribution of BCC texture components in experiment + absence of HCP texture component. Variant selection \leftrightarrow 24 rotations g^{BCCsym} are not equivalent?

It is easier to study using analytical model of texture! Model = 2 standard Gaussians with FWHM = $20^{\circ} + 0.2$ phon:



Quantifying the transformation texture

Using analytical model, it is easy to calculate BCC \rightarrow HCP transformation texture separately: \checkmark for texture components (100) or (111)

✓ for every g^{BCCsym} out of 24 possibilities



Some valuable information

Transformation texture modeling with Burger's orientation relationship reveals that:

✓ Typical BCC compression texture cannot yield HCP transformation texture with (0001) or (01 $\overline{1}0$) components in compression IPF; if observed, the experimental data should be reevaluated.



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Texture memory in U-0.7 Ti alloy



C. Mo et al. (2021). J. Nuclear Mater. https://doi.org/10.1016/j.jnucmat.2021.153317

Some physical properties of U phases



J.R. Bridge et al. (1956). JOM. https://doi.org/10.1007/BF03377866



Young's moduli of U phases in GPa, equal area projections, linear scale. View along C-axis.

E. S. Fisher & H. J. McSkimin. (1958). J. Appl. Phys. B. Beeler et al. (2013). J. Nucl. Mater.

4 main slip systems and 4 twinning regimes in α -U \rightarrow it is quite challenging to assess deformation regime

HIPPO diffractometer (LANSCE, Los-Alamos)



Flux at the sample: $\approx 2 \cdot 10^7 \text{ n/cm}^2/\text{s}$ *d*-resolution: $\approx 0.4\%$ for 144° banks, $\approx 0.7\%$ for 90° banks Total flight path ~ 10 m Sample dimensions ~ 2 cm Range of *d*-spacings: 0.12-47.5 Å

> *M.M. Schmitt et al. (2023). J. Appl. Crystallogr. https://doi.org/10.1107/S1600576723009275*



Experiment & 90° detectors bank patterns



Phases and their unit cell parameters

#	T, °C	Phase	Vol.%	a, Å	b, Å	c, Å	b/a	c/a
1	100	α-U	100	2.93078(3)	5.96290(7)	5.07716(6)	2.035	1.732
2	670	β-U	92.4	10.883(6)	10.883(6)	5.719(3)	1	0.525
		U ₂ Ti	7.6	4.939(3)	4.939(3)	2.9223(2)	1	0.592
3	300	α-U	91.2	2.87258(3)	5.83670(5)	4.98232(5)	2.032	1.734
		U ₂ Ti	8.8	4.8369(5)	4.8369(5)	2.8544(3)	1	0.59
4	790	γ-U	100	3.546(6)	3.546(6)	3.546(6)	1	1
5	200	α-U	92.0	2.8823(8)	5.867(2)	5.004(1)	2.036	1.736
		U ₂ Ti	8.0	4.868(1)	4.868(1)	2.8675(8)	1	0.589

Pole figures during $\alpha \rightarrow \beta \rightarrow \alpha$



 α -U, 100°C, initial texture.

β-U, 670°C.







 U_2 Ti, 300°C, after $\alpha \rightarrow \beta \rightarrow \alpha$.

α -U, 300°C, after $\alpha \rightarrow \beta \rightarrow \alpha$.



Correspondences for $\alpha \leftrightarrow \beta$ U transformation

$_{\alpha}C_{\beta}11$	$_{\alpha}C_{\beta}12$	$_{\alpha}C_{\beta}13$	
$(hkl)_{\beta} = (hkl)_{\alpha} _{\alpha}C_{\beta}21$	$_{\alpha}C_{\beta}22$	$_{\alpha}C_{\beta}23$	CORRESPONDENCE MATRIX #4
$_{\alpha}C_{\beta}31$	$_{\alpha}C_{\beta}32$	$_{\alpha}C_{\beta}33$	$_{\alpha}C_{\beta} = 0 1/2 1,$
			1 -2 0

The transformation from β to α uranium in dilute uranium alloys is believed to be martensitic in nature but, in view of the complex crystal structure, only one atom in 30 (at best) can be carried to its correct position by the lattice deformation; the others must also shuffle. In view of the complex shuffles, it is not surprising that the growth process appears to be thermally activated. The transformation differs from all others described in several ways. It is the only martensitic transformation known in which the principal axes of the lattice deformation are not parallel to simple low index directions of the parent and also the only transformation in which there is not a unique correspondence.

J.W. Christian. The theory of transformations in metals and alloys. Pergamon, 2002.

CORRESP	ONDENC	E MATR	RIX #3
11/	4 -1/2	1	
$_{\alpha}C_{\beta} = 3/4$	4 -1/2	-1,	
· 1/2	2 2	0	

conference on the Peaceful Uses of Atomic Energy. **CORRESPONDENCE #6** $(100)_{\alpha} \rightarrow (001)_{\beta}, [012]_{\alpha} \rightarrow [110]_{\beta}.$

Correspondences \rightarrow Orientation relationships

 $ODF \leftrightarrow rotations$

But correspondence matrices transform orthogonal directions into nonorthogonal! Angle between $(\overline{174})_{\alpha}$ and $(4\overline{21})_{\alpha}$ is $\approx 96.32^{\circ}$ Angle between $(\overline{174})_{\alpha}$ and $(17.11.)_{\alpha}$ is $\approx 86.63^{\circ}$ Angle between $(\overline{171})_{\beta}$ and $(101)_{\beta}$ is $\approx 80.41^{\circ}$.

Use **polar decomposition**! A real square matrix ${}_{\alpha}C_{\beta} \rightarrow$ product of unitary orthogonal matrix (**rotation**!) and positive symmetric matrix (deformation!).

	β→α U transformatio	n	$\alpha \rightarrow \beta$ U transformation		
OR	$_{lpha}C_{eta}$	$\{\alpha,\beta,\gamma\}$	$_{\beta}C_{\alpha} = (_{\alpha}C_{\beta})^{-1}$	$\{\alpha,\beta,\gamma\}^{-1}$	Ref.
#1	$\begin{pmatrix} -1/2 & 7/2 & 1/2 \\ -3/2 & -1/2 & 1/2 \\ 1 & 0 & 1 \end{pmatrix}$	{354.6, 31.7, 108.0}	$\begin{pmatrix} -1/15 & -7/15 & 4/15 \\ 4/15 & -2/15 & -1/15 \\ 1/15 & 7/15 & 11/15 \end{pmatrix}$	{72.0, 31.7, 185.4}	[Lomer, 1956]
#2	$-\begin{pmatrix} 3/2 & -13/4 & 1/2 \\ -3/2 & -1/4 & 1/2 \\ 1 & 1/2 & 1 \end{pmatrix}$	{214.8, 145.3, 286.3}	$-\begin{pmatrix} 1/15 & -7/15 & 1/5 \\ -4/15 & -2/15 & 1/5 \\ 1/15 & 8/15 & 7/10 \end{pmatrix}$	{253.7, 145.3, 325.2}	[Lomer, 1956]*
#3	$\begin{pmatrix} 11/4 & -1/2 & 1 \\ 3/4 & -1/2 & -1 \\ 1/2 & 2 & 0 \end{pmatrix}$	{75.8, 95.9, 244.0}	$\begin{pmatrix} 4/15 & 4/15 & 2/15 \\ -1/15 & -1/15 & 7/15 \\ 7/30 & -23/30 & -2/15 \end{pmatrix}$	{296.0, 95.9, 104.2}	[Lomer, 1956]
#4	$\begin{pmatrix} 3 & 3/2 & 0 \\ 0 & 1/2 & 1 \\ 1 & -2 & 0 \end{pmatrix}$	{296.7, 82.1, 87.1}	$\begin{pmatrix} 4/15 & 0 & 1/5 \\ 2/15 & 0 & -2/5 \\ -1/15 & 1 & 1/5 \end{pmatrix}$	{92.9, 82.1, 243.3}	[Lomer, 1956]
#5	$\begin{pmatrix} -15 & 70 & 2 \\ -12 & -3 & 4 \\ 8 & 1 & 7 \end{pmatrix}$	{8.8, 31.9, 93.8}	$\frac{1}{8519} \begin{pmatrix} -25 & -488 & 286\\ 116 & -121 & 36\\ 12 & 575 & 885 \end{pmatrix}$	{86.2, 31.9, 171.2}	[Butcher & Rowe, 1953]**
#6	$\begin{pmatrix} 0 & 0 & 1 \\ 27 & -7 & 0 \\ 7 & 27 & 0 \end{pmatrix}$	{75.4, 90.0, 180.0}	$\begin{pmatrix} 0 & 27/778 & 7/778 \\ 0 & -7/778 & 27/778 \\ 1 & 0 & 0 \end{pmatrix}$	{0.0, 90.0, 104.6}	[Konobeevs ky et al., 1958]***

Transformation texture calculations





Transformation texture calculations



Several ORs?



Best fit: 0.35 OR #1 + 0.32 OR #2 II + 0.11 OR #3 + 0.22 OR #4

Successfully decreases the texture sharpness and is much closer in terms of symmetry. Bot not quite...

 $\beta \rightarrow \alpha$ transformation does not work too...



Some valuable information

Experiments and transformation texture modeling using available orientation relationships reveal that:

- ✓ There is definitely the texture memory effect in α -U after α → β → α transformation, but some variant selection may be active due to increased texture sharpness and absence of some subsidiary components.
- ✓ Experimental α-U texture is relatively strong and symmetric. Experimental β-U texture is close to random, but some features exist, e.g., in (001) pole figure.
- ✓ All the published orientation relationships cannot reproduce observed $\alpha \rightarrow \beta$ transformation textures. The combination of several ORs also does not work. Nevertheless, all ORs decrease the texture strength. Most likely, there is no variant selection (which is expected to increase texture strength).
- ✓ All the published orientation relationships cannot reproduce observed $\beta \rightarrow \alpha$ transformation textures. There <u>should be a variant selection</u> to significantly increase the texture strength.
- ✓ Variant selection related to U_2 Ti (which is initially absent)?
- ✓ Is there a "true" (unpublished) orientation relationship? It is in fact possible to search for it based on experimental textures!

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HRFD @ IBR-2 with the new BS detector

November 2024



Platform for the PSD

BS + **PSD** cover large solid angle of \vec{q} in K_A. Expectations: texture measurements will be possible within ~ 1 hour (including *in situ*!).



Conclusions

- 1. It is possible to quantitatively work with bulk transformation textures including variant selections or simultaneously occurring ORs (and in the course of this work some software has been developed for this purpose)!
- 2. This seems highly relevant in studies of inelastic deformation regimes in materials undergoing martensitic transformations, or fine-tuning anisotropy of physical properties of such materials.
- 3. *In situ* neutron diffraction texture measurements are feasible only if measurement time is no more than few hours; now there is a possibility to organize such experiments at the HRFD @ IBR-2.

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