Structural Fingerprints of Plasticity: Linking the Shear Transformation Zone Size to Pair Distribution Functions in Metallic Glasses

Valeria Lemkova

E-Mail: v.lemkova@matsci.uni-saarland.de

Materials Science and Methods & Metallic Materials
Saarland University
Campus D2 3, 66123 Saarbruecken, Germany

What does a (material)scientist do in research?

What does a (material)scientist do in research?

Create a material

What does a (material)scientist do in research?

Create a material

analyse

What does a (material)scientist do in research?

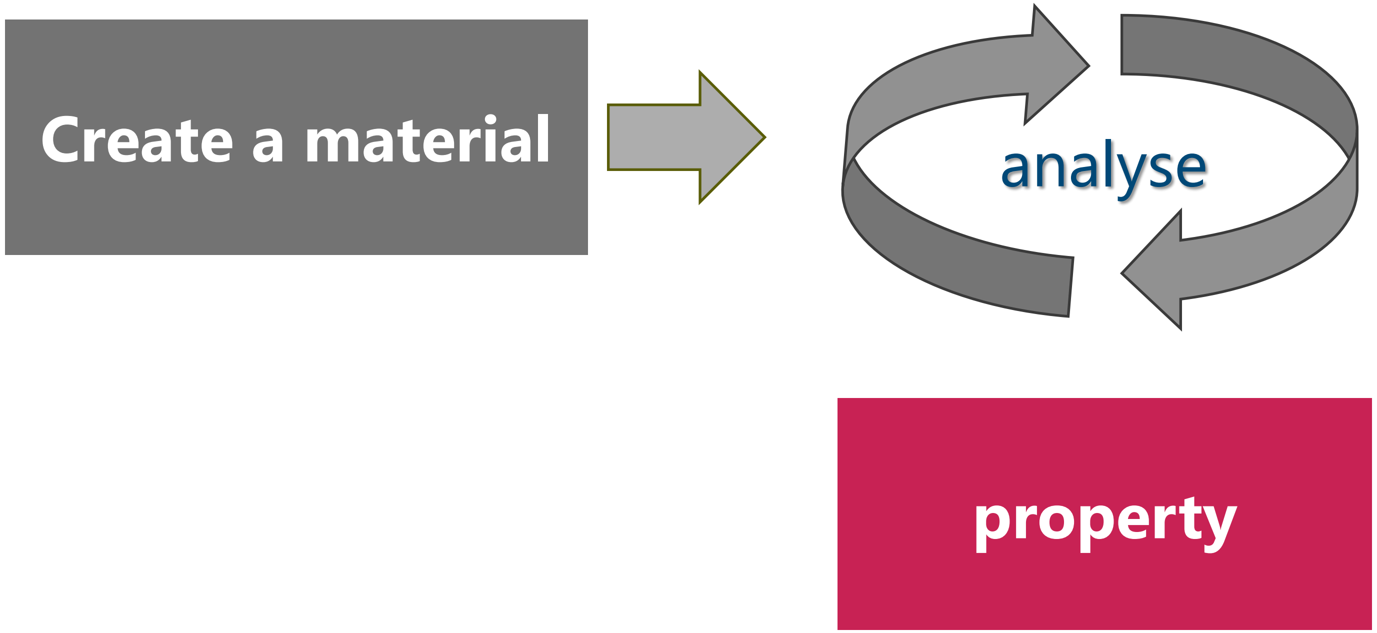
structure

Create a material →

analyse

property

What does a (material)scientist do in research?



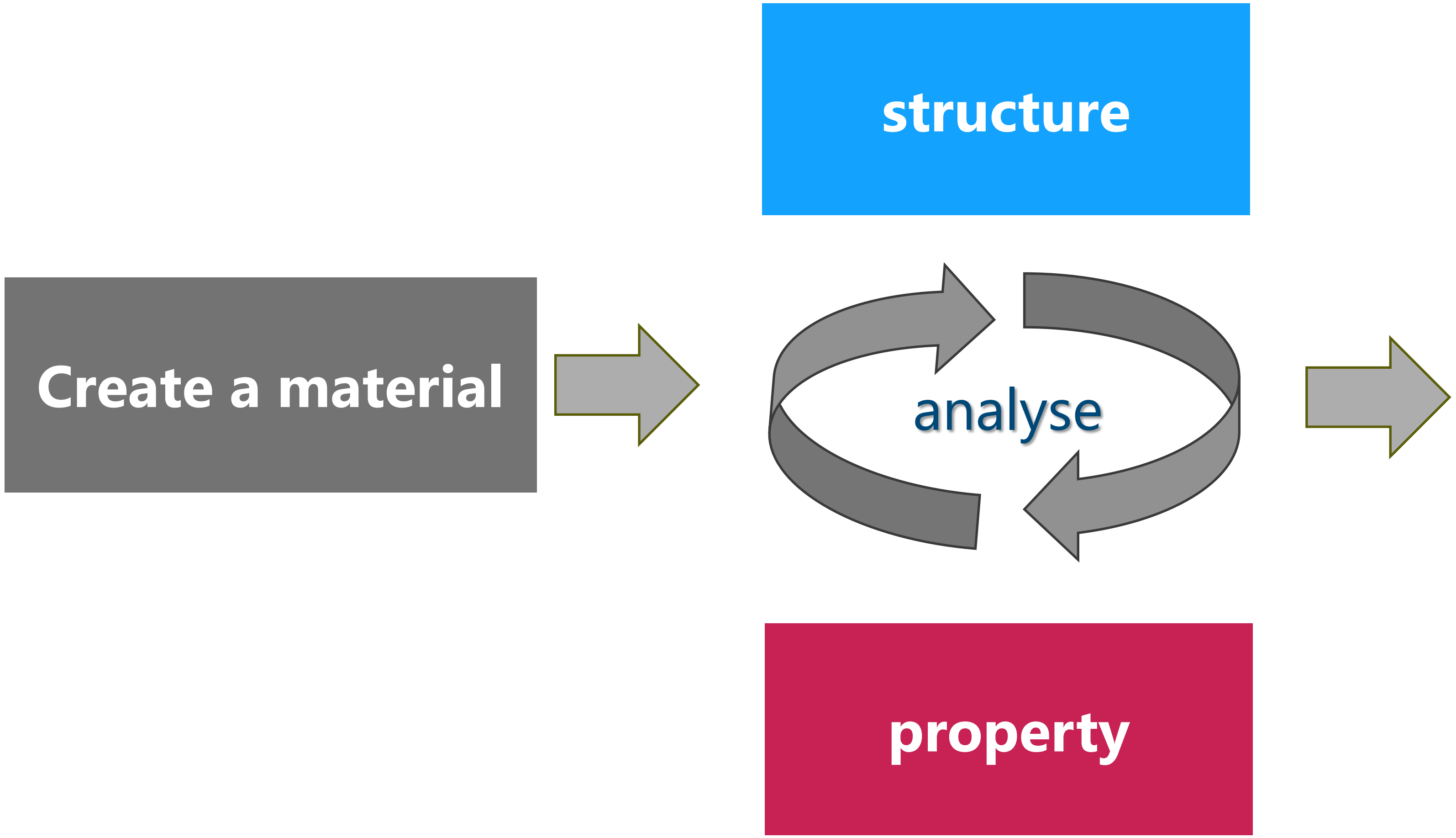
Create a material

structure

analyse

property

What does a (material)scientist do in research?



Create a material

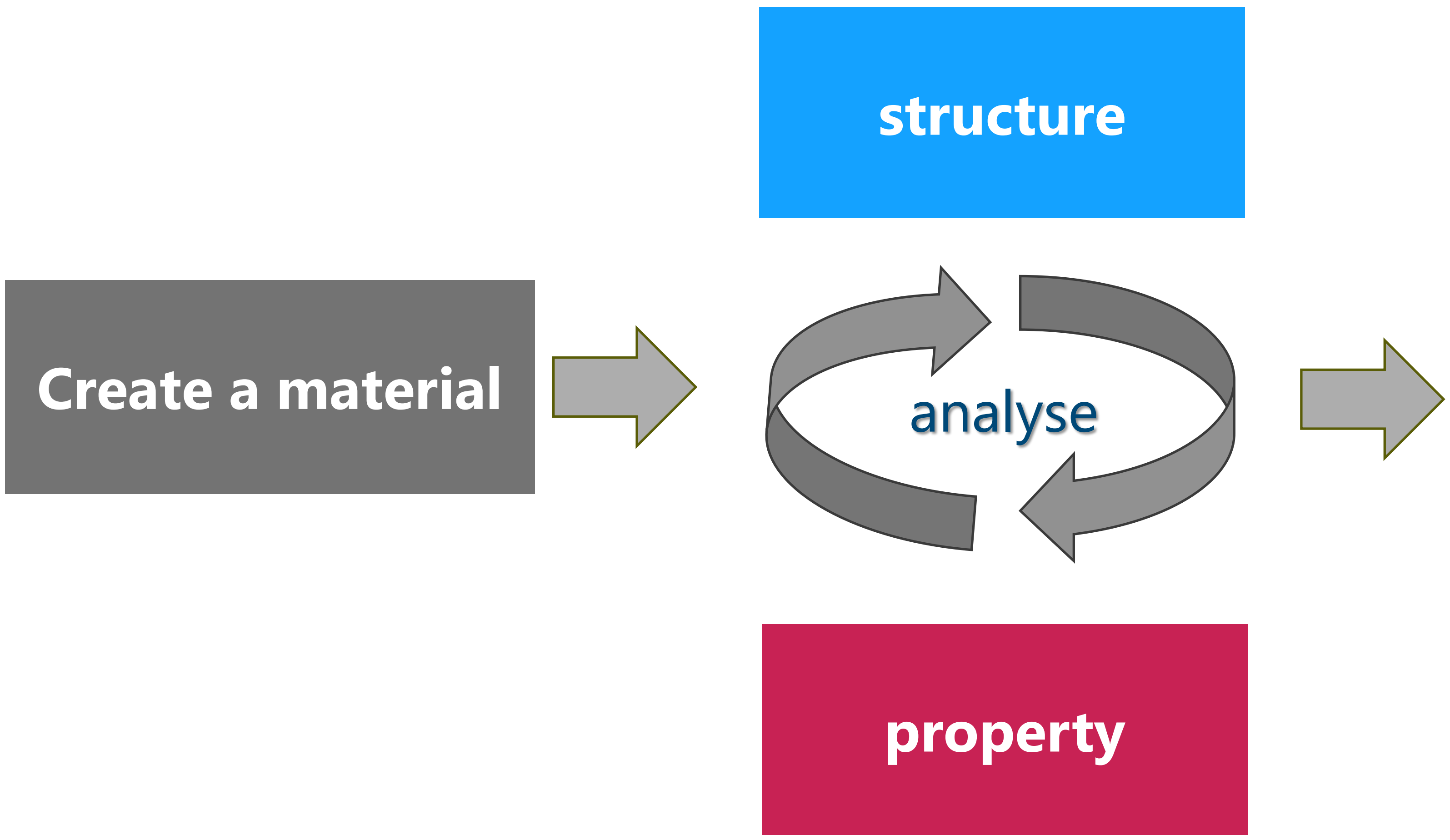
structure

analyse

property

Is my material fit for my application?

What does a (material)scientist do in research?



Create a material

structure

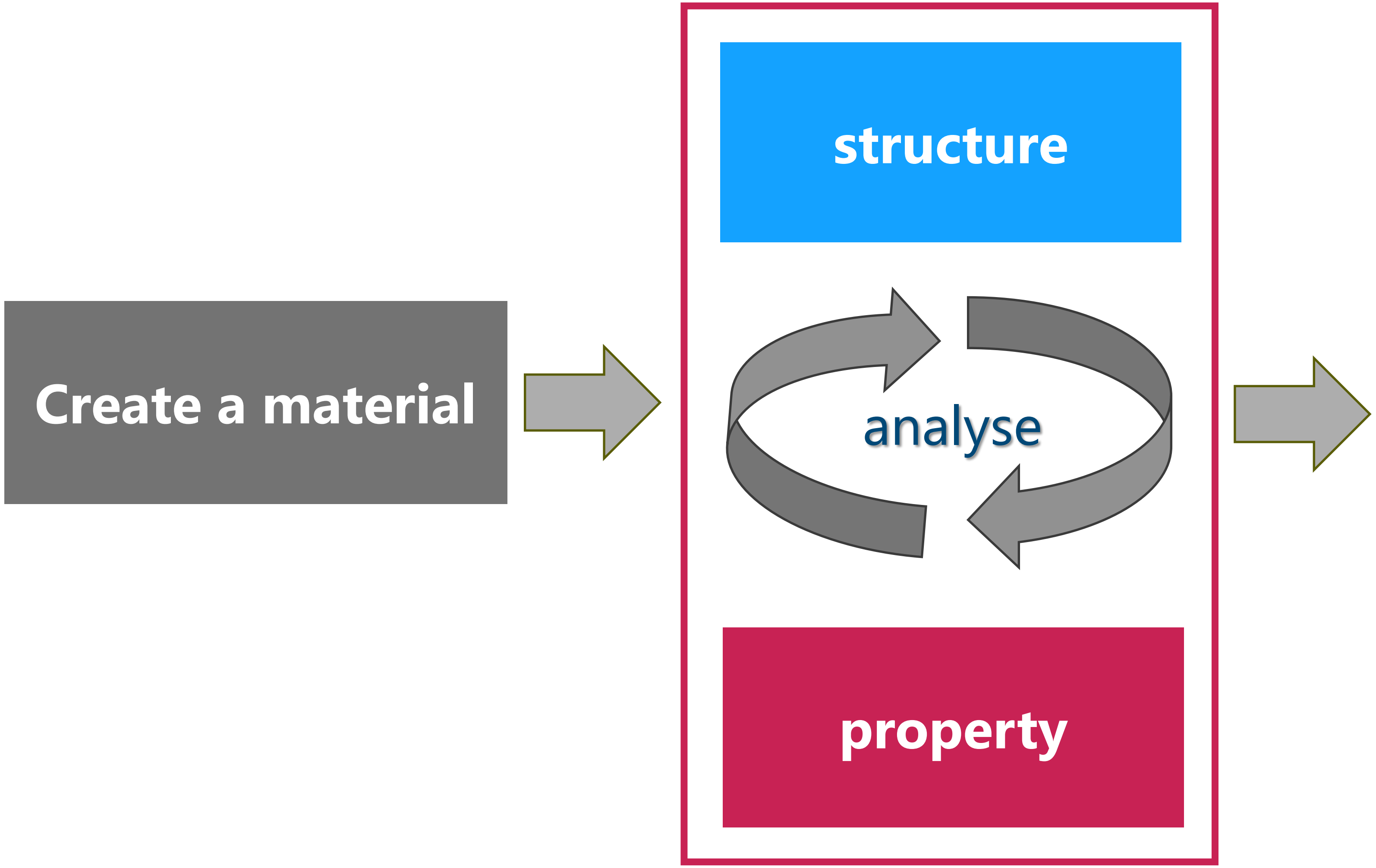
analyse

property

Is my material fit for my application?

@https://www.reuters.com/lifestyle/two-giant-rubber-ducks-debut-hong-kong-bid-drive-double-happiness-2023-06-09/

What does a (material)scientist do in research?



Create a material

structure

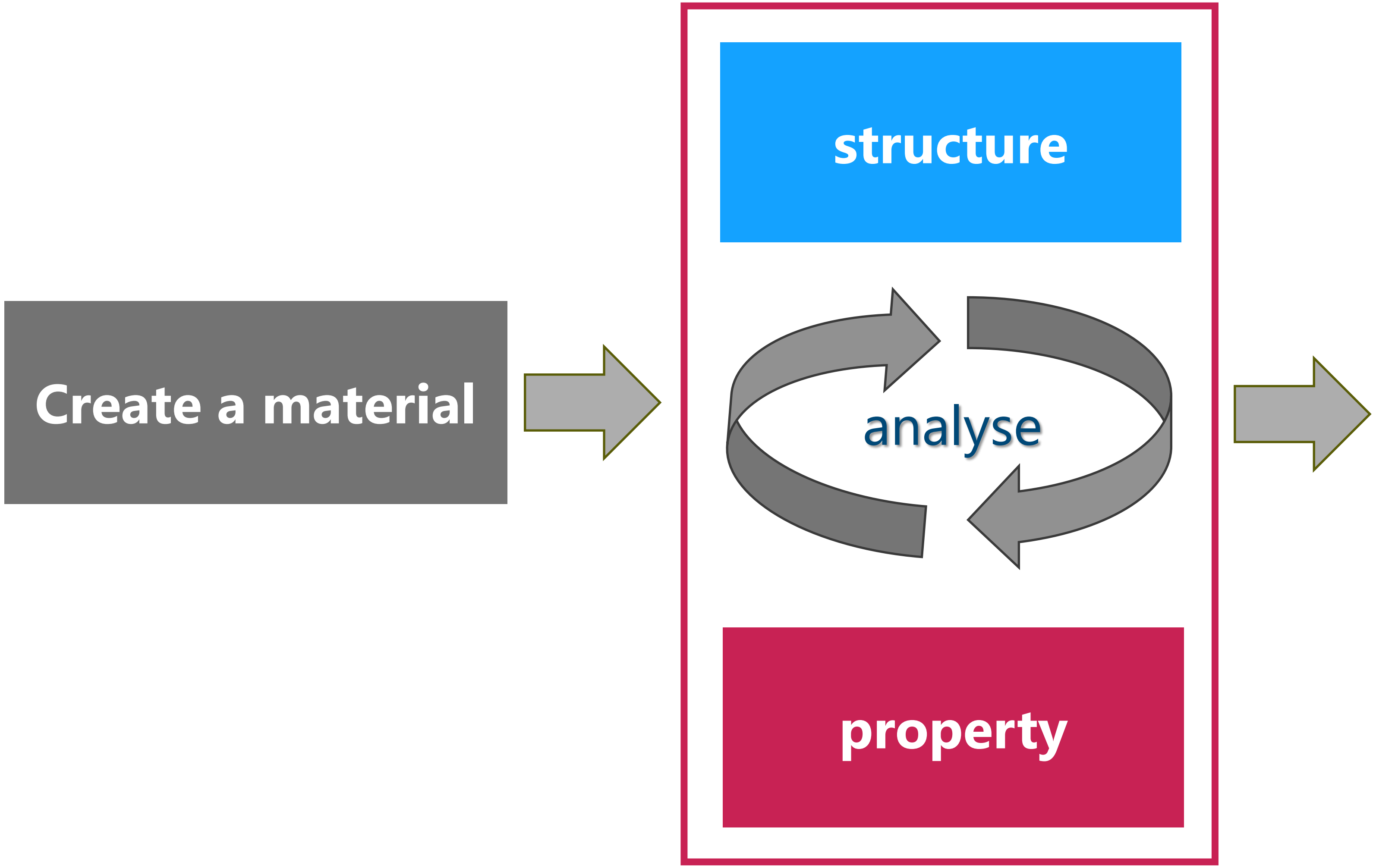
analyse

property

Is my material fit for my application?

@https://www.reuters.com/lifestyle/two-giant-rubber-ducks-debut-hong-kong-bid-drive-double-happiness-2023-06-09/

What does a (material)scientist do in research?



Create a material

structure

analyse

property

Is my material fit for my application?

@https://www.reuters.com/lifestyle/two-giant-rubber-ducks-debut-hong-kong-bid-drive-double-happiness-2023-06-09/

Can I predict **mechanical properties** from the **structural fingerprint**?

Motivation & Objectives

Why is understanding structure–property relationships in metallic glasses important?

Motivation & Objectives

Why is understanding structure–property relationships in metallic glasses important?

BMG = Bulk Metallic Glass

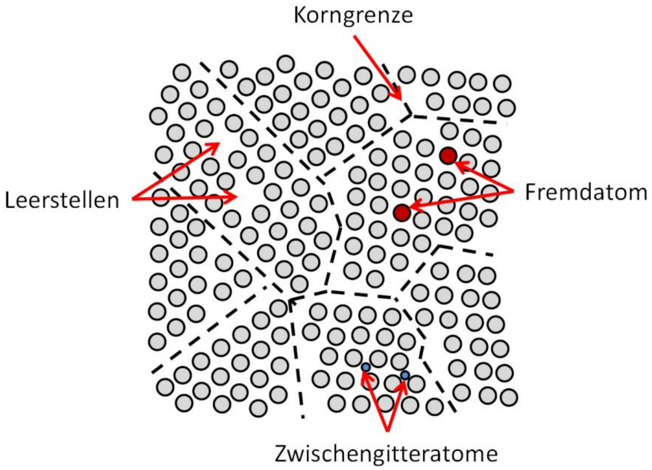
amorphous metal that can be cast to at least 2 mm

Motivation & Objectives

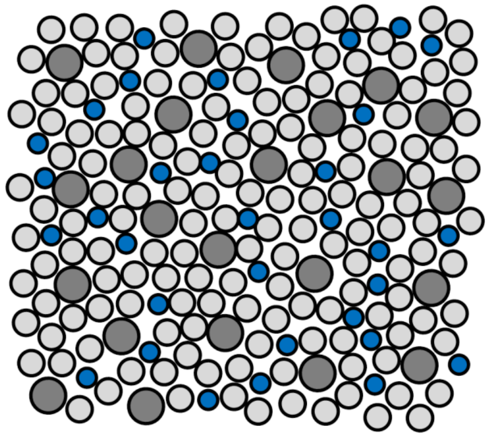
Why is understanding structure–property relationships in metallic glasses important?

What does amorphouse mean?

no long-range order (**LRO**) **but** some middle-range order (**MRO**) and short-range order (**SRO**)



Polykristallines Metall



Amorphes Metall

BMG = Bulk Metallic Glass

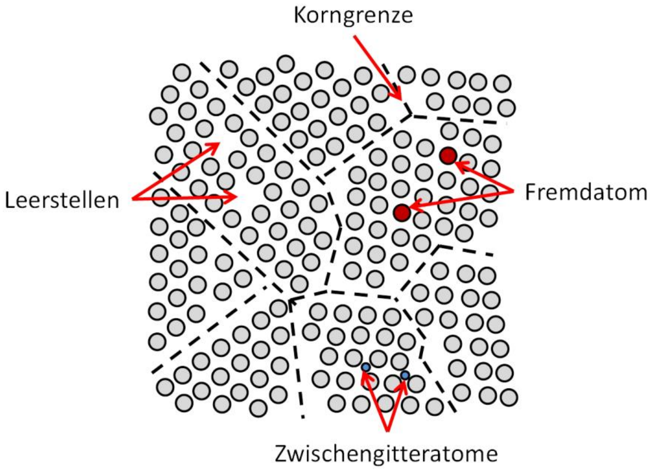
amorphous metal that can be cast to at least 2 mm

Motivation & Objectives

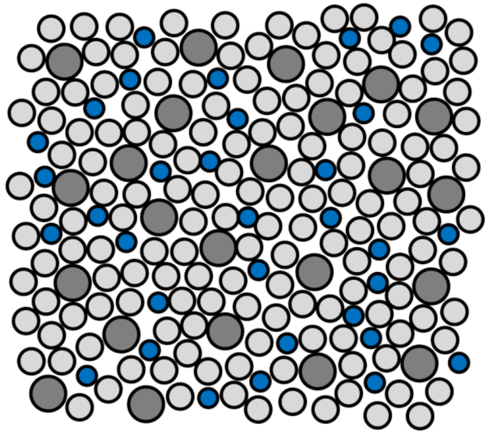
Why is understanding structure–property relationships in metallic glasses important?

What does amorphouse mean?

no long-range order (**LRO**) **but** some middle-range order (**MRO**) and short-range order (**SRO**)



Polykristallines Metall



Amorphes Metall

BMG = Bulk Metallic Glass

amorphous metal that can be cast to at least 2 mm

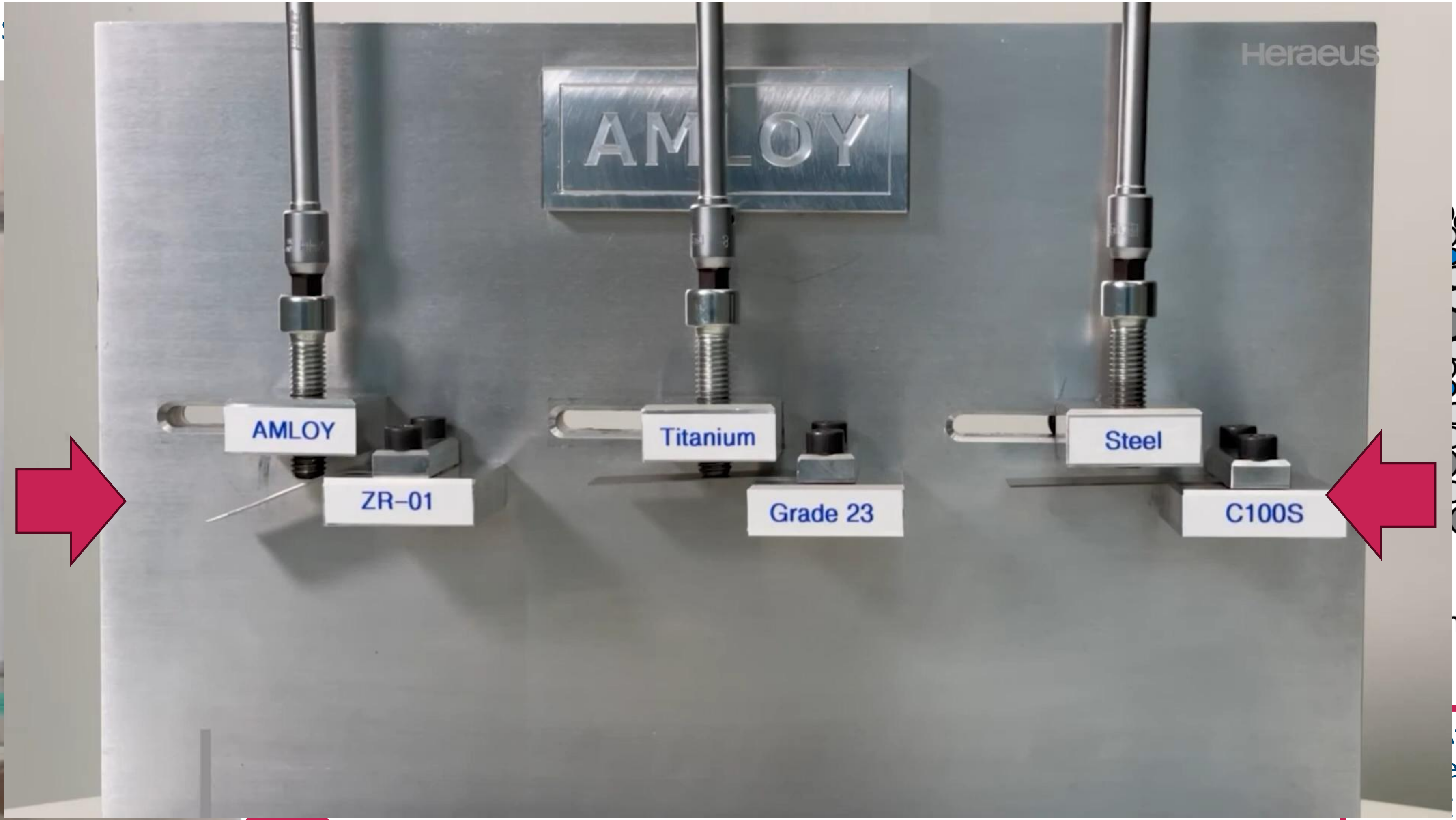
Nucleation must bei suppressed

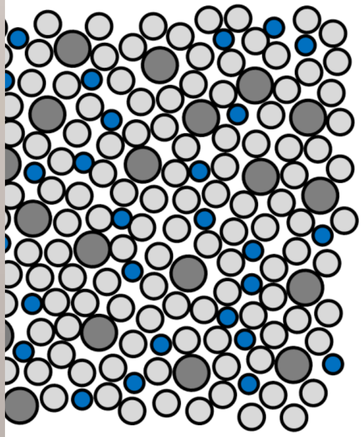
1. Atoms of significantly different sizes
2. Crystallization is energetically unfavorable

Motivation & Objectives

@https://www.heraeus-amloy.com

Why i





morphes Metall

toms of significantly
ent sizes
rystallization is
energetically unfavorable

be cast to at least 2 mm

bei suppressed

Motivation & Objectives

Why is understanding structure–property relationships in metallic glasses important?

Properties

Härter als Stahl – Elastisch wie Kunststoff

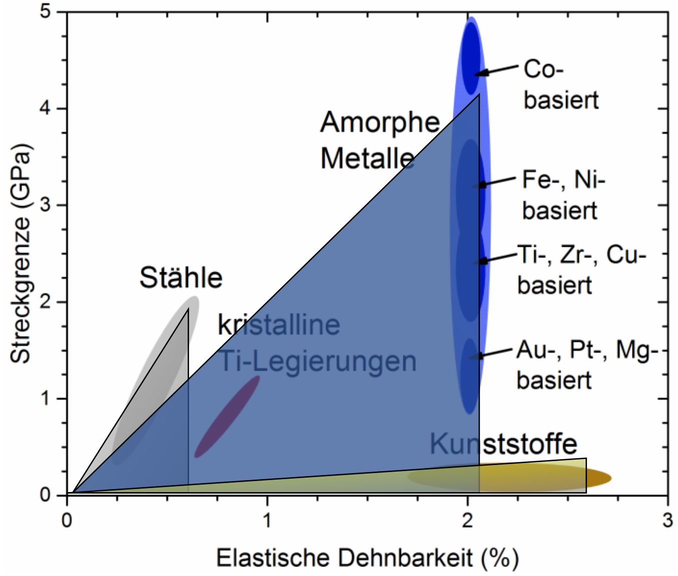
Harder than steel – As elastic as plastic

Motivation & Objectives

Why is understanding structure–property relationships in metallic glasses important?

Properties

- **High elastic limit**: up to **2%** (vs. ~0.2% in crystalline metals)
- **High strength**: up to **4.5 GPa**, approaching theoretical limits
- **High hardness & wear resistance**: up to **900 HV5**
- **Low energy dissipation**: excellent damping behavior
- **High elastic energy storage**: large area under the stress–strain curve



Härter als Stahl – Elastisch wie Kunststoff

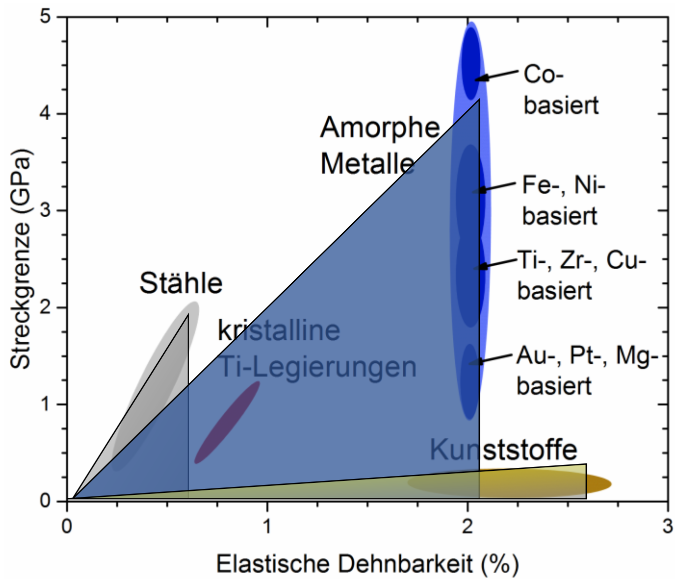
Harder than steel – As elastic as plastic

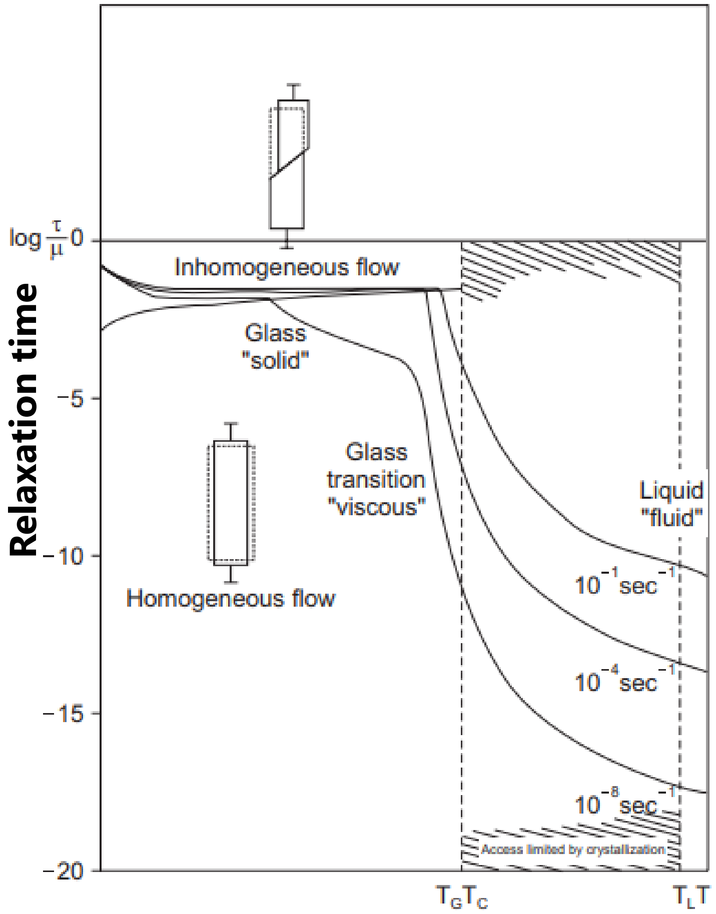
Motivation & Objectives

Why is understanding structure–property relationships in metallic glasses important?

Properties

- **High elastic limit**: up to **2%** (vs. ~0.2% in crystalline metals)
- **High strength**: up to **4.5 GPa**, approaching theoretical limits
- **High hardness & wear resistance**: up to **900 HV5**
- **Low energy dissipation**: excellent damping behavior
- **High elastic energy storage**: large area under the stress–strain curve





@Spaepen (1977) – A microscopic mechanism for steady state inhomogeneous flow in metallic glasses

> *Härter als Stahl – Elastisch wie Kunststoff*
>
> *Harder than steel – As elastic as plastic*

Motivation & Objectives

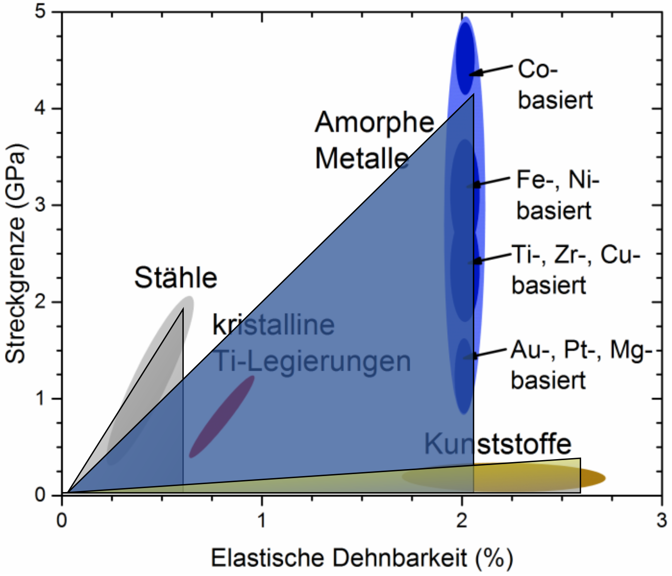
Why is understanding structure–property relationships in metallic glasses important?

Properties

- **High elastic limit**: up to **2%** (vs. ~0.2% in crystalline metals)
- **High strength**: up to **4.5 GPa**, approaching theoretical limits
- **High hardness & wear resistance**: up to **900 HV5**
- **Low energy dissipation**: excellent damping behavior
- **High elastic energy storage**: large area under the stress–strain curve

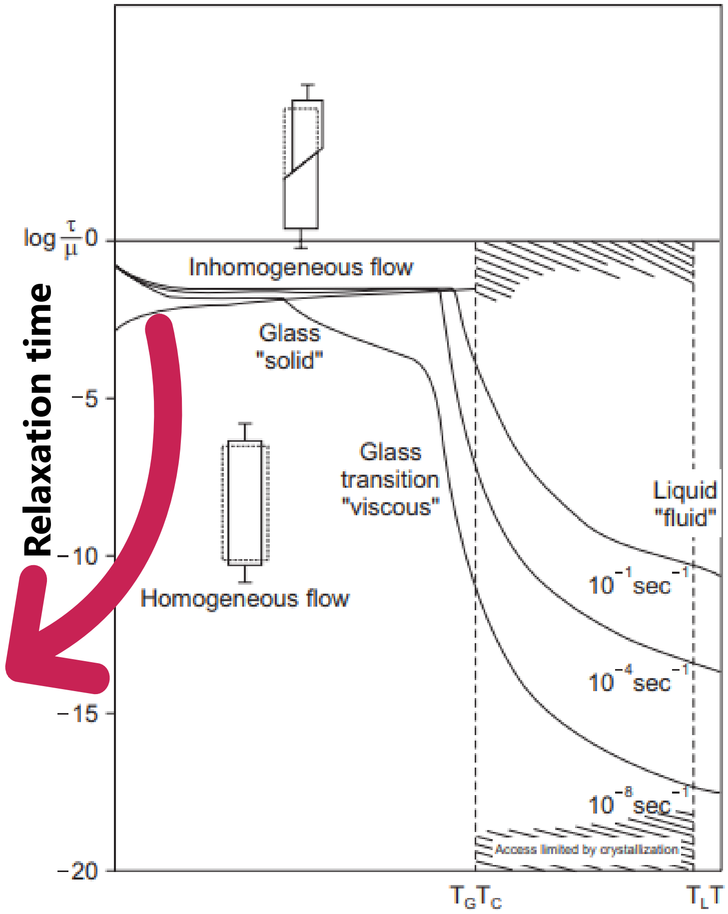
Härter als Stahl – Elastisch wie Kunststoff

Harder than steel – As elastic as plastic



Inhomogeneous flow:

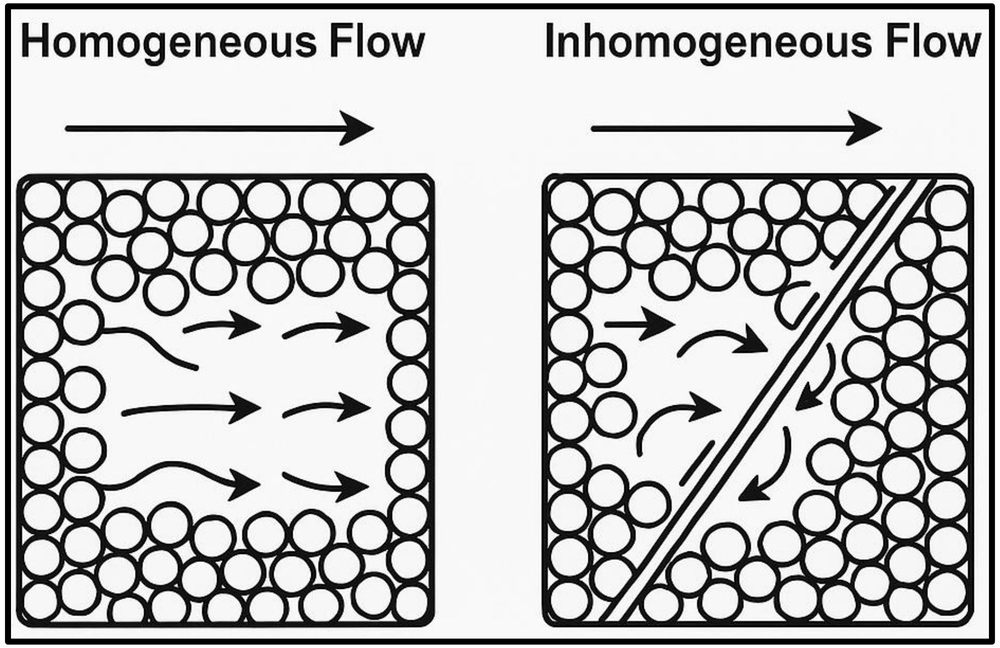
plastic deformation is **not** evenly distributed throughout the material



@Spaepen (1977) – A microscopic mechanism for steady state inhomogeneous flow in metallic glasses

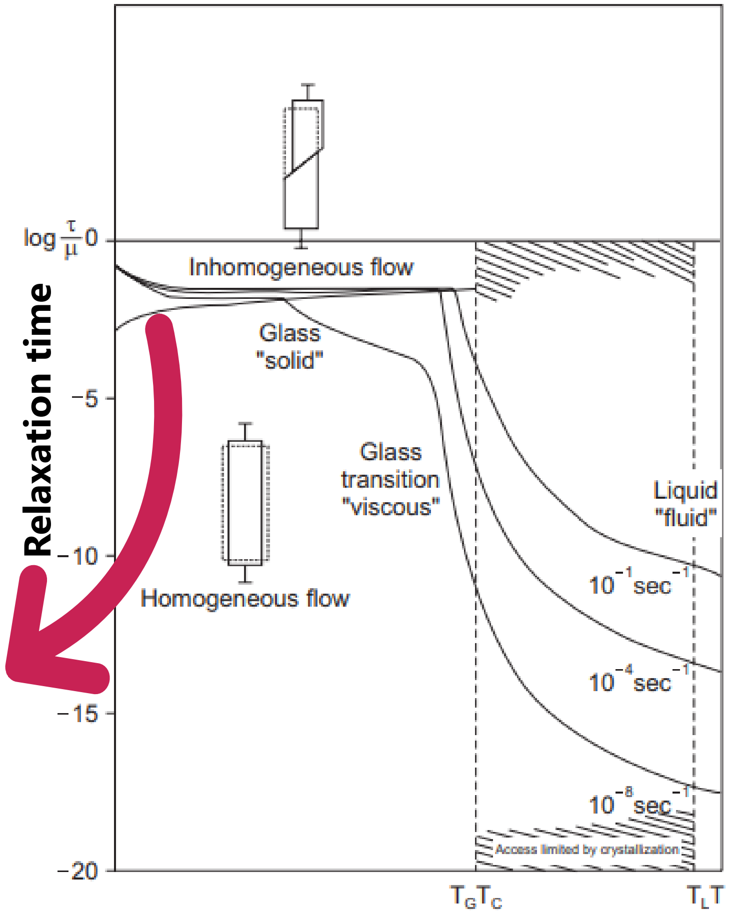
Motivation & Objectives

Why is understanding structure–property relationships in metallic glasses important?



Inhomogeneous flow:

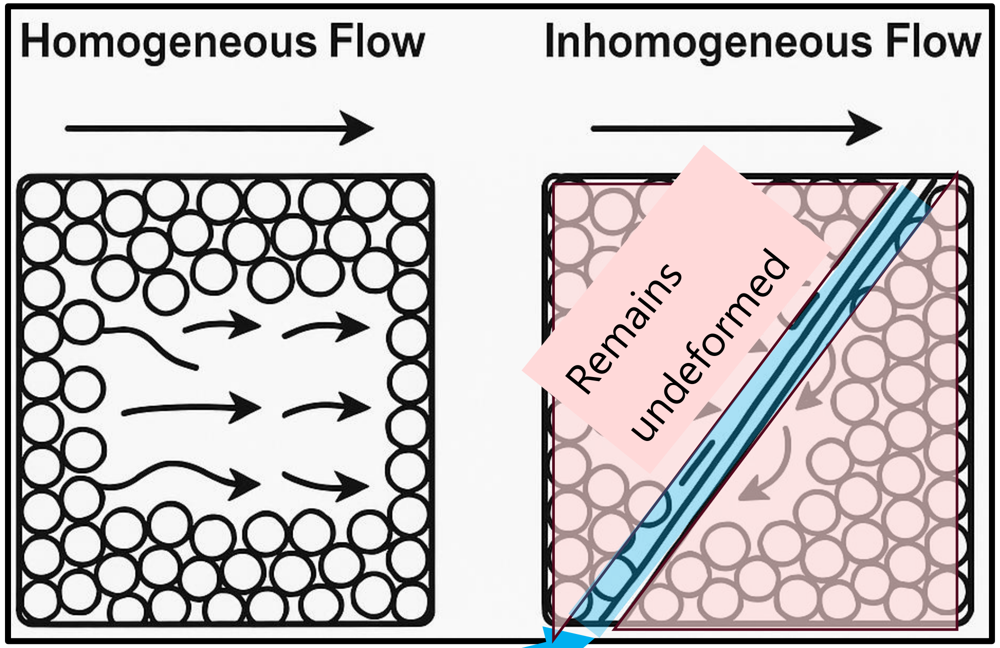
plastic deformation is **not** evenly distributed throughout the material



@Spaepen (1977) – A microscopic mechanism for steady state inhomogeneous flow in metallic glasses

Motivation & Objectives

Why is understanding structure–property relationships in metallic glasses important?



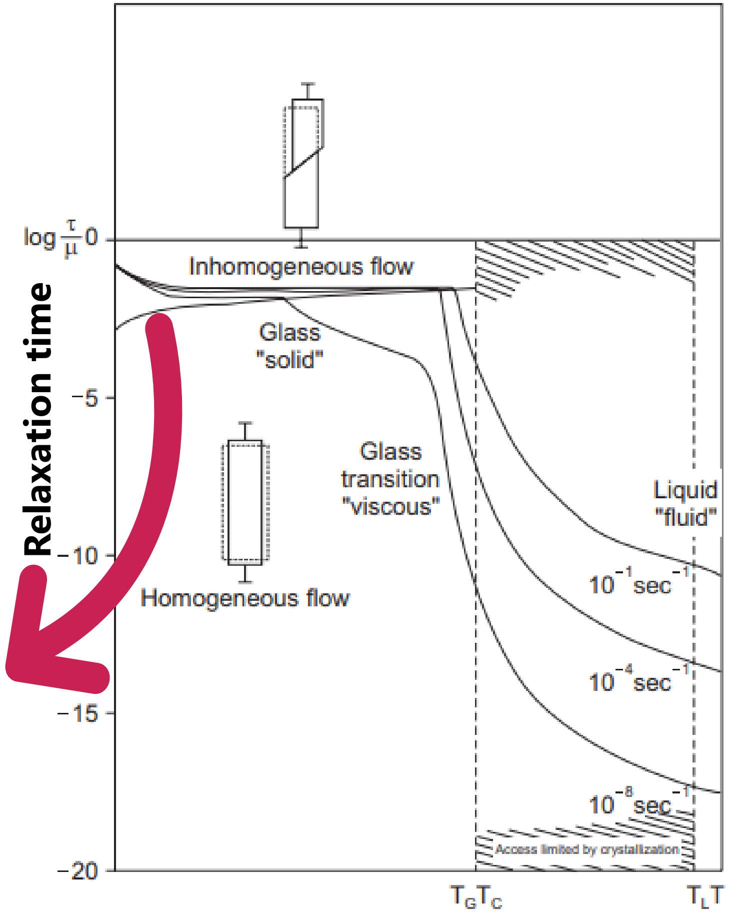
Localized region:

Shearbands form suddenly and locally

Brittle-like failure

Inhomogeneous flow:

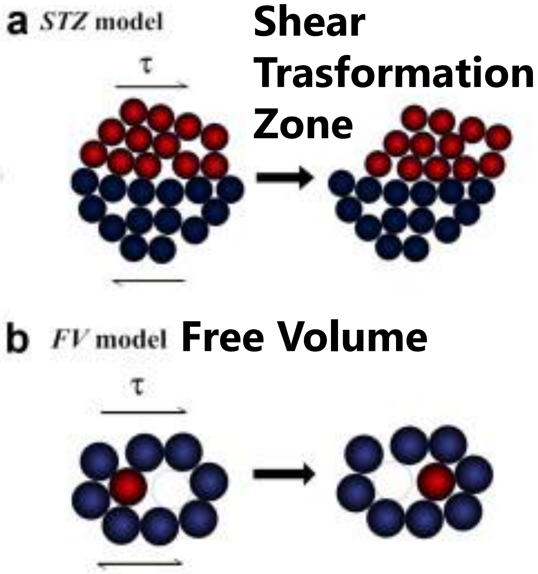
plastic deformation is **not** evenly distributed throughout the material



@Spaepen (1977) – A microscopic mechanism for steady state inhomogeneous flow in metallic glasses

Motivation & Objectives

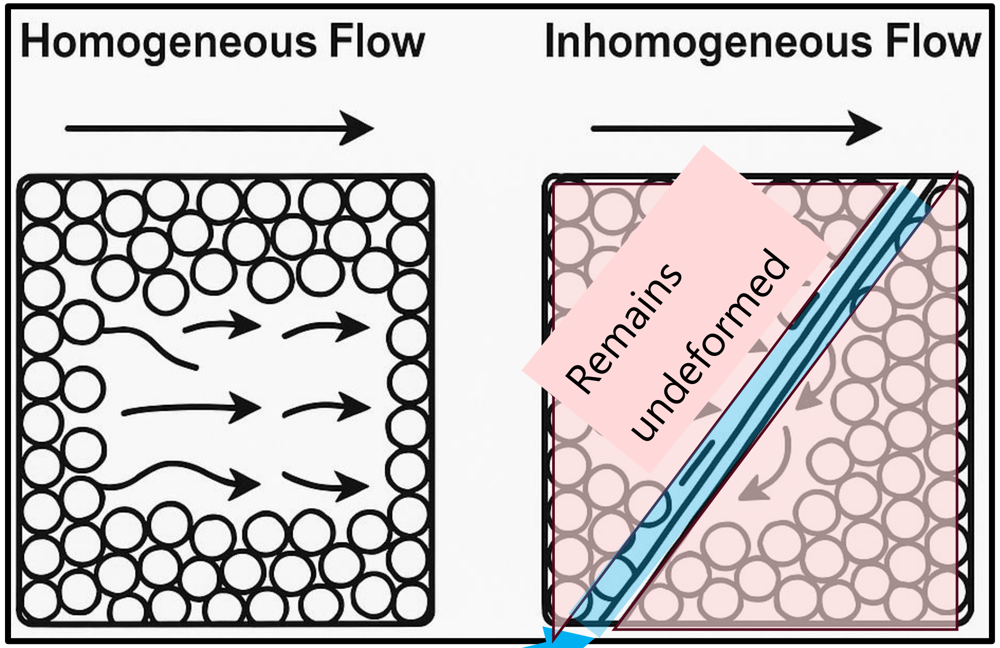
Why is understanding structure–property relationships in metallic glasses important?



Shear Trasformation Zone

Free Volume

@Ke H.B., Zeng J.F., Liu C.T., Yang Y (2014)
Structure Heterogeneity in Metallic Glass: Modeling and Experiment



Remains undeformed

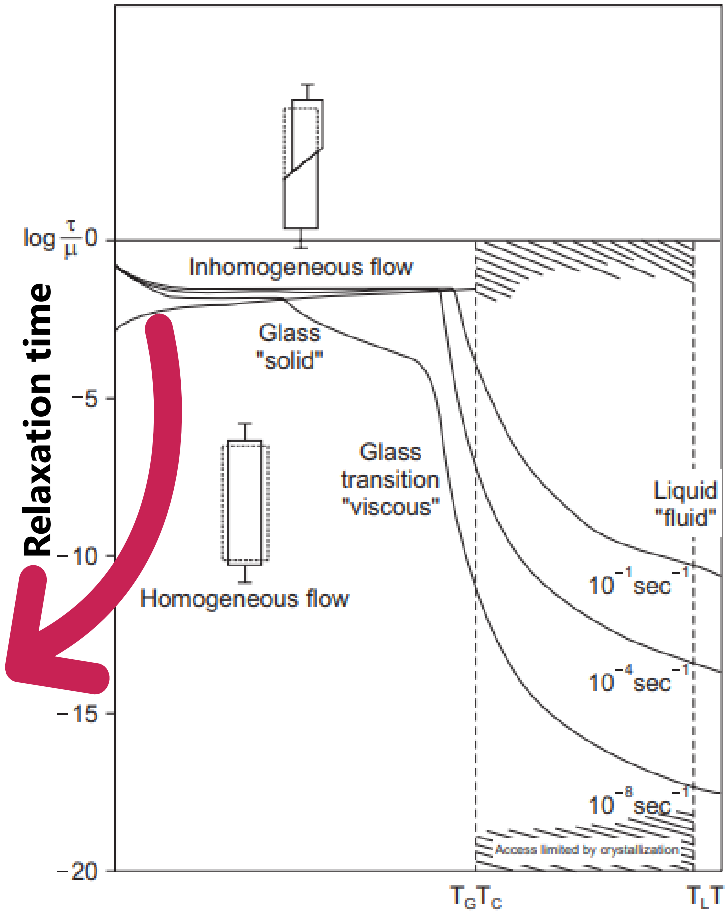
Localized region:

Shearbands form suddenly and locally

Brittle-like failure

Inhomogeneous flow:

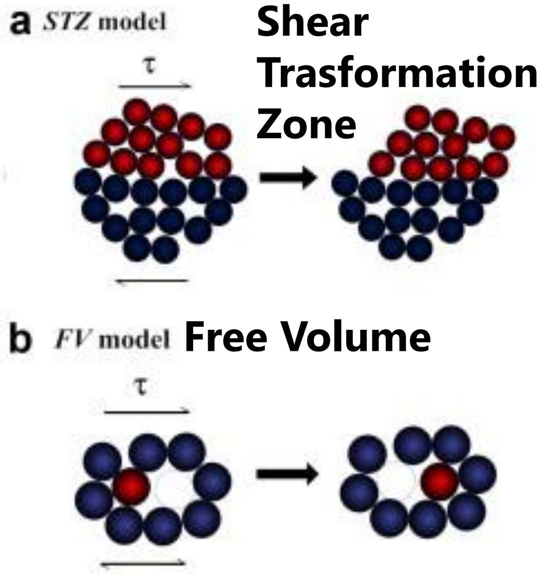
plastic deformation is **not** evenly distributed throughout the material



@Spaepen (1977) – A microscopic mechanism for steady state inhomogeneous flow in metallic glasses

Motivation & Objectives

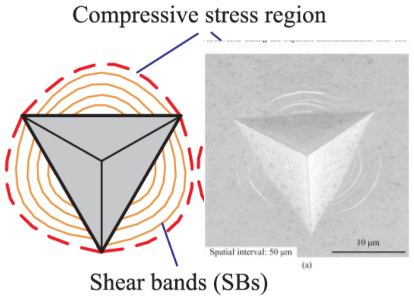
Why is understanding structure–property relationships in metallic glasses important?



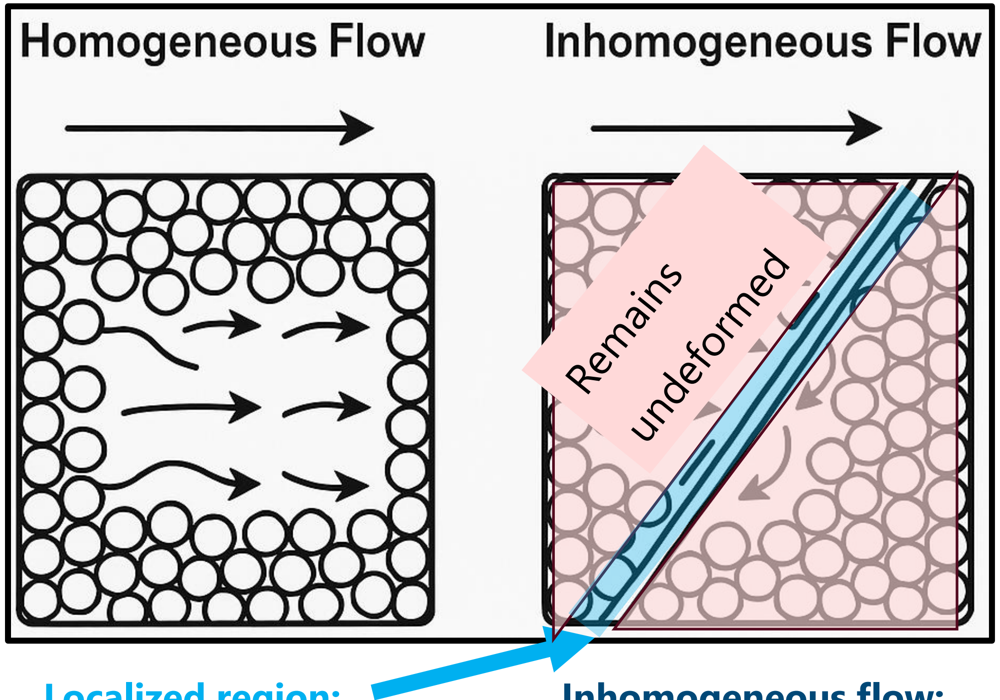
Shear Trasformation Zone

Free Volume

@Ke H.B., Zeng J.F., Liu C.T., Yang Y (2014) Structure Heterogeneity in Metallic Glass: Modeling and Experiment



@Huang H., Yan Jiwang (2017) Investigating shear band interaction in metallic glasses by adjacent nanoindentation



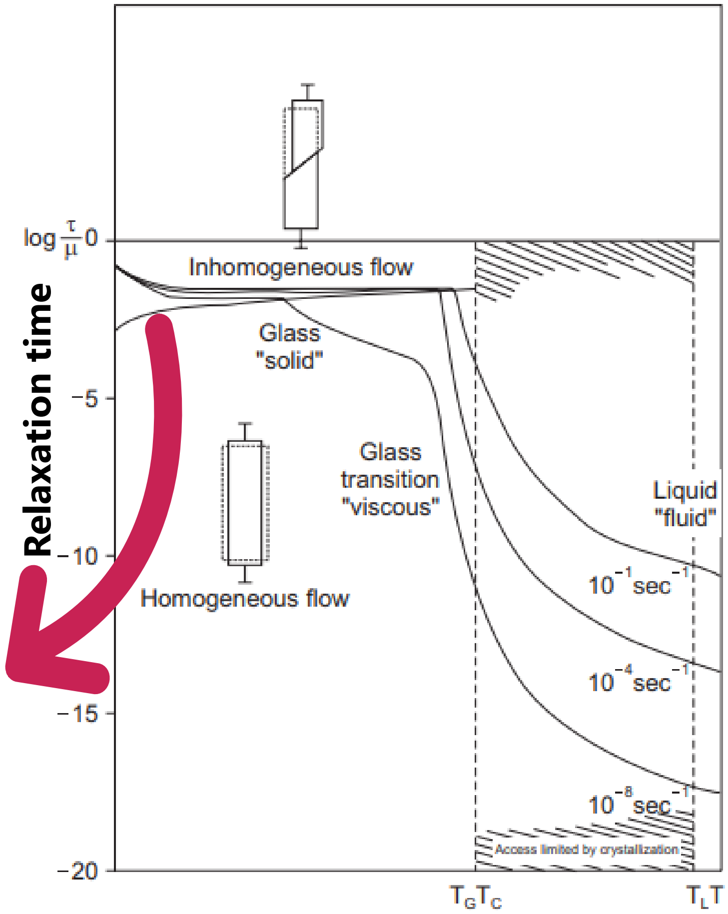
Localized region:

Shearbands form suddenly and locally

Brittle-like failure

Inhomogeneous flow:

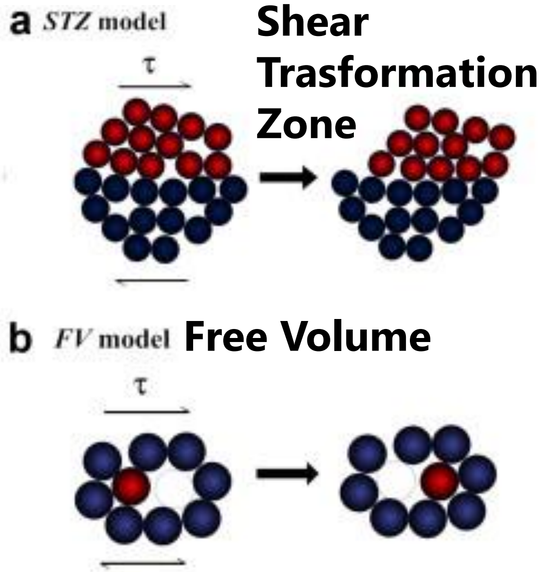
plastic deformation is **not** evenly distributed throughout the material



@Spaepen (1977) – A microscopic mechanism for steady state inhomogeneous flow in metallic glasses

Motivation & Objectives

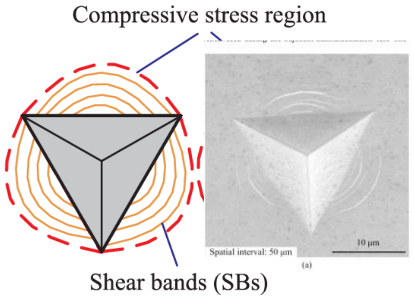
Why is understanding structure–property relationships in metallic glasses important?



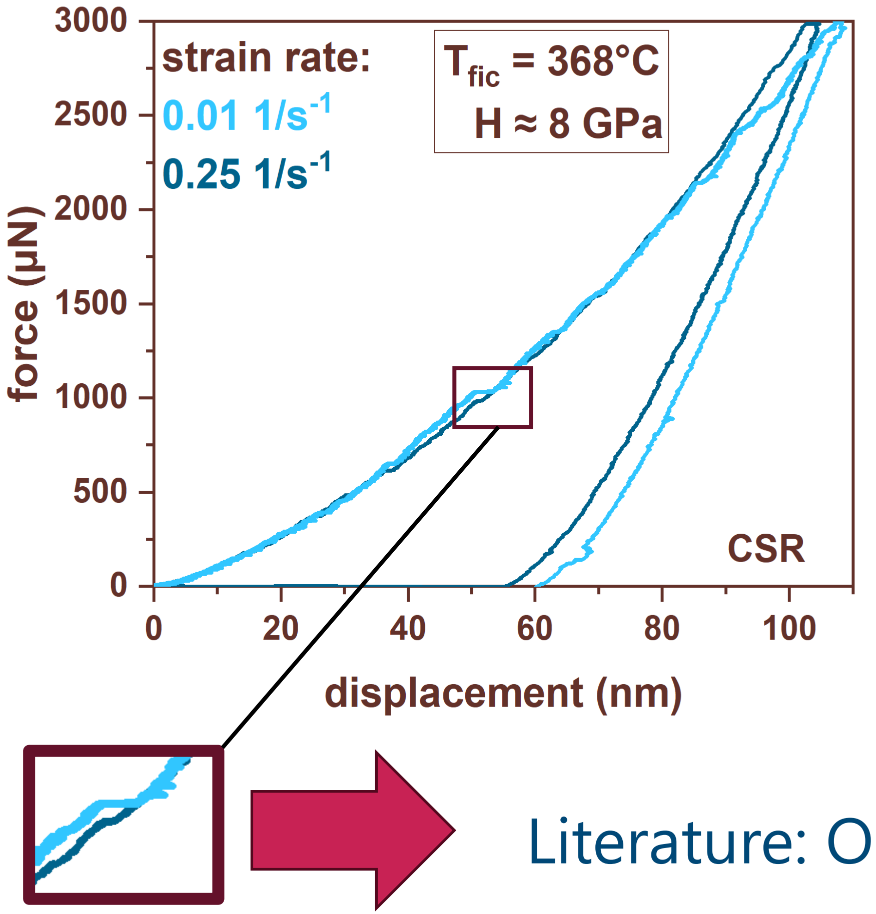
Shear Trasformation Zone

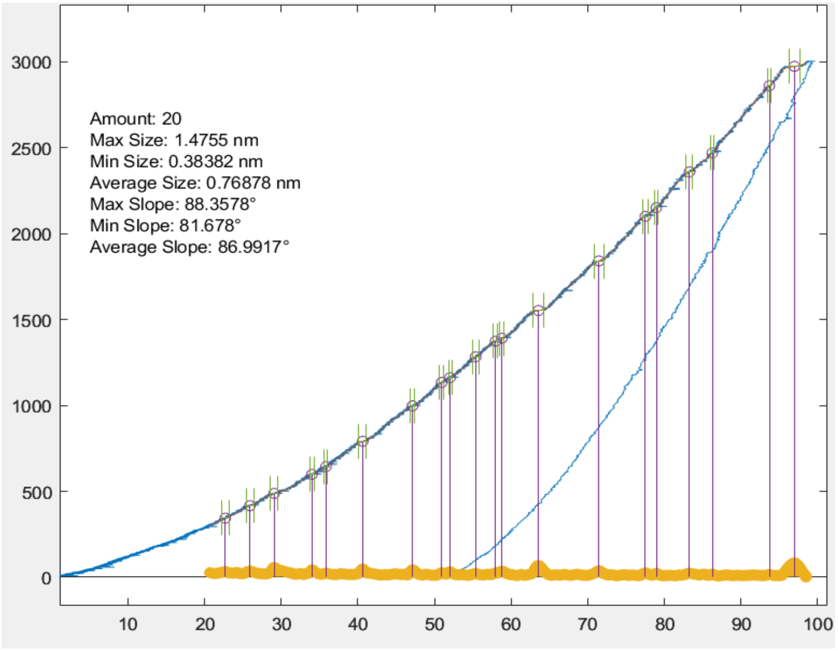
Free Volume

@Ke H.B., Zeng J.F., Liu C.T., Yang Y (2014)
Structure Heterogeneity in Metallic Glass: Modeling and Experiment



@Huang H., Yan Jiwang (2017) Investigating shear band interaction in metallic glasses by adjacent nanoindentation





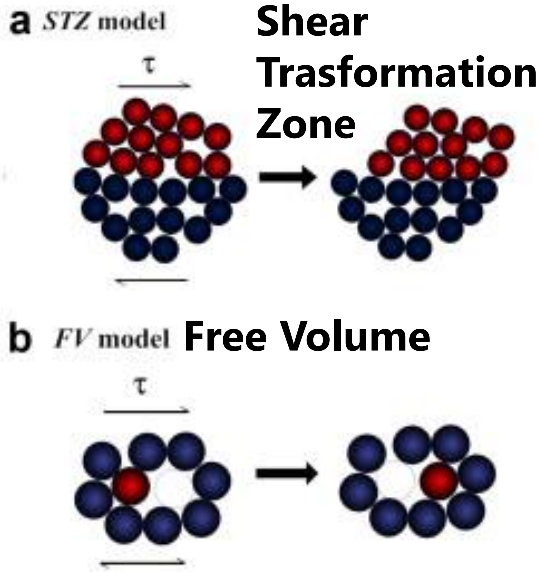
Amount: 20
Max Size: 1.4755 nm
Min Size: 0.38382 nm
Average Size: 0.76878 nm
Max Slope: 88.3578°
Min Slope: 81.678°
Average Slope: 86.9917°

@MATLAB

Literature: One Pop-In = One Shearband

Motivation & Objectives

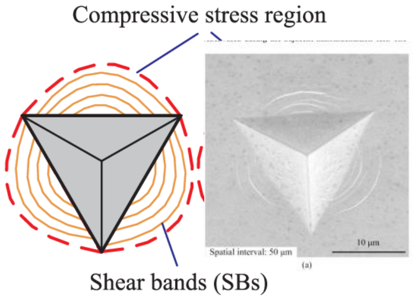
Why is understanding structure–property relationships in metallic glasses important?



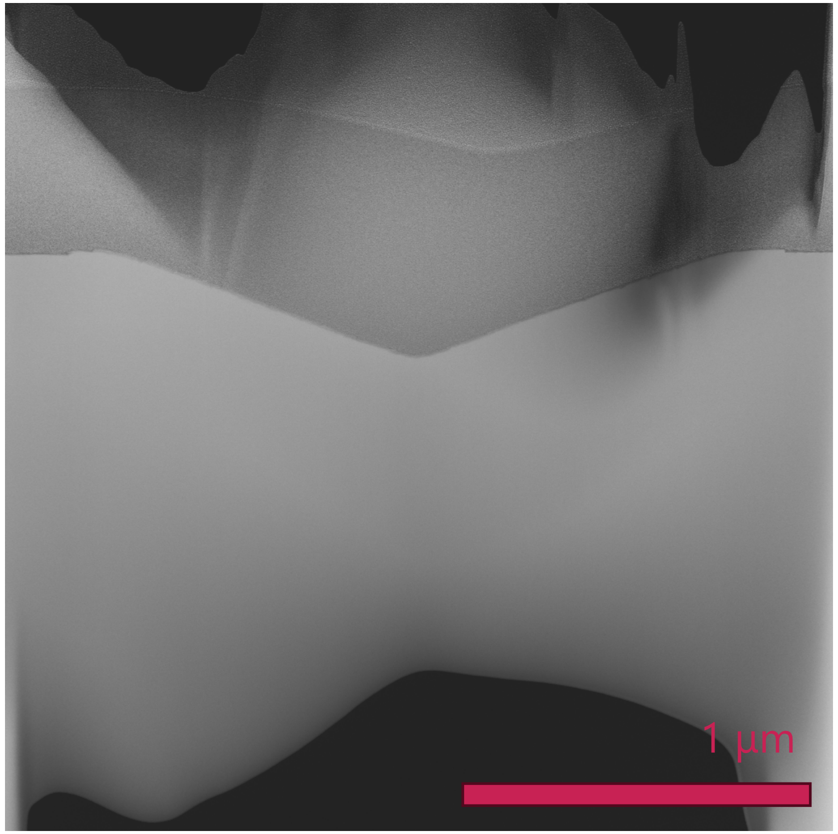
Shear Trasformation Zone

Free Volume

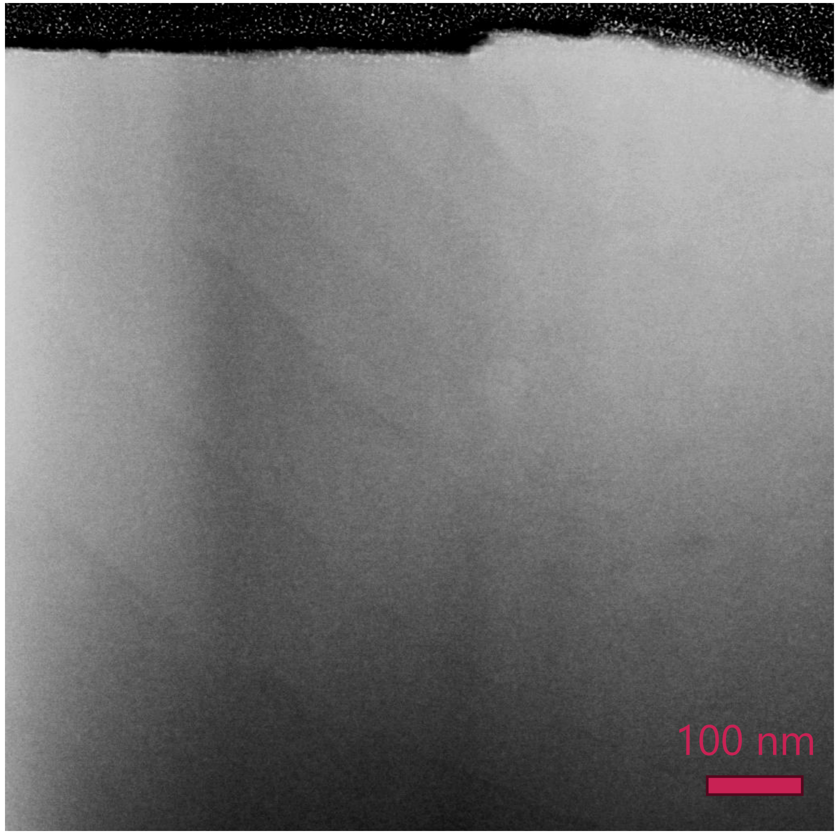
@Ke H.B., Zeng J.F., Liu C.T., Yang Y (2014) Structure Heterogeneity in Metallic Glass: Modeling and Experiment



@Huang H., Yan Jiwang (2017) Investigating shear band interaction in metallic glasses by adjacent nanoindentation



Jeol TEM JEM-F200

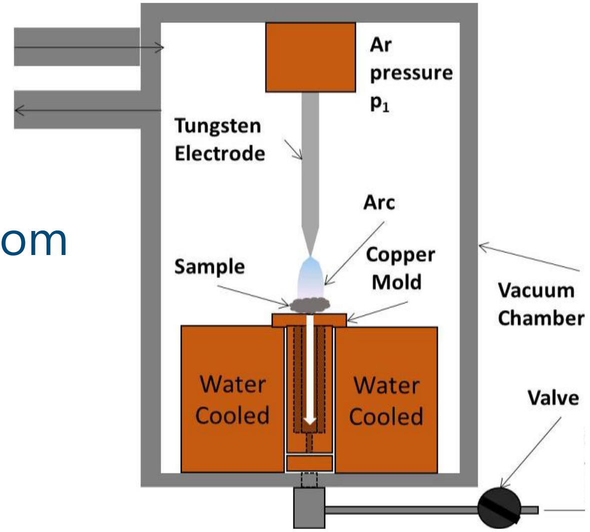


STEM HAADF

Material Preparation

Alloy casting

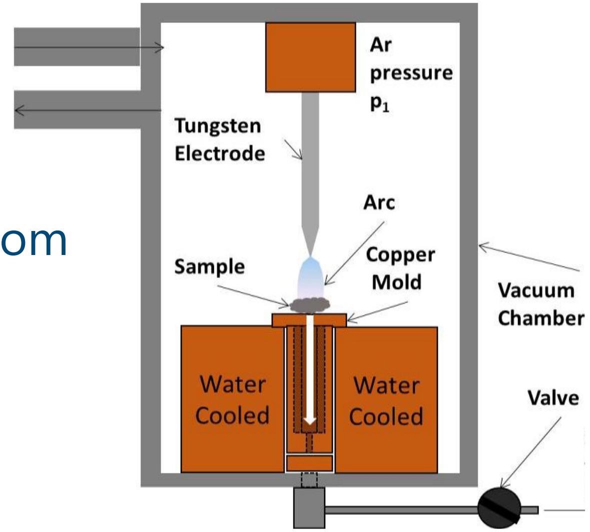
1. Weighing
2. Semi-finished products from the electric arc furnace
3. Suction casting system



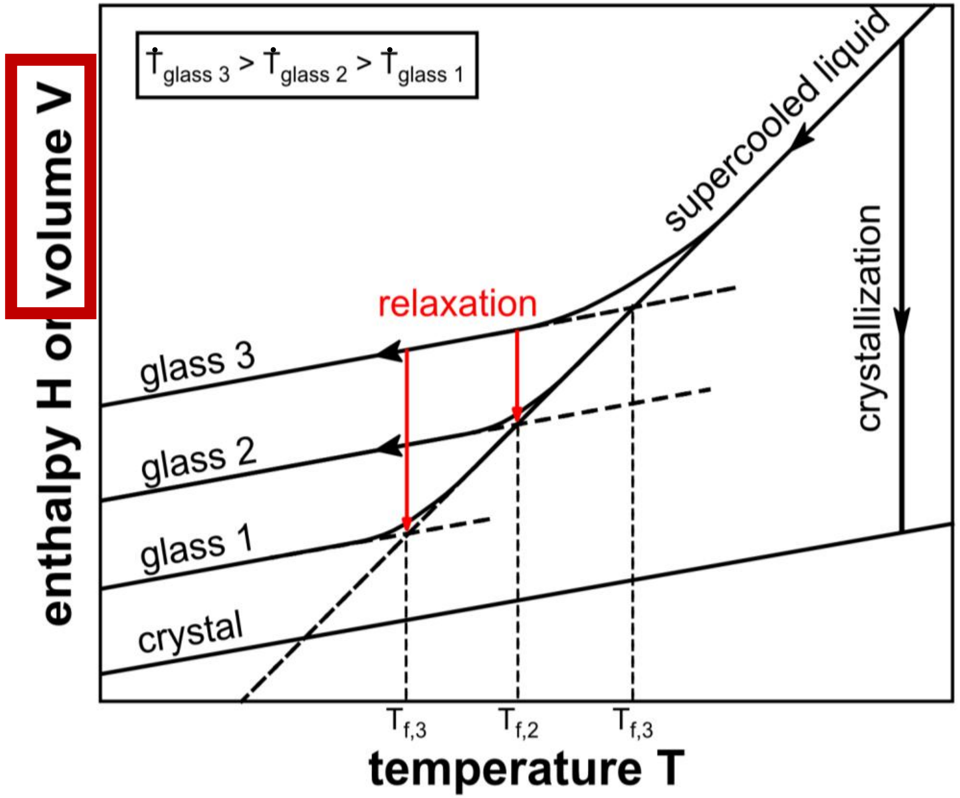
Material Preparation

Alloy casting

1. Weighing
2. Semi-finished products from the electric arc furnace
3. Suction casting system



Relaxation in sub-Tg



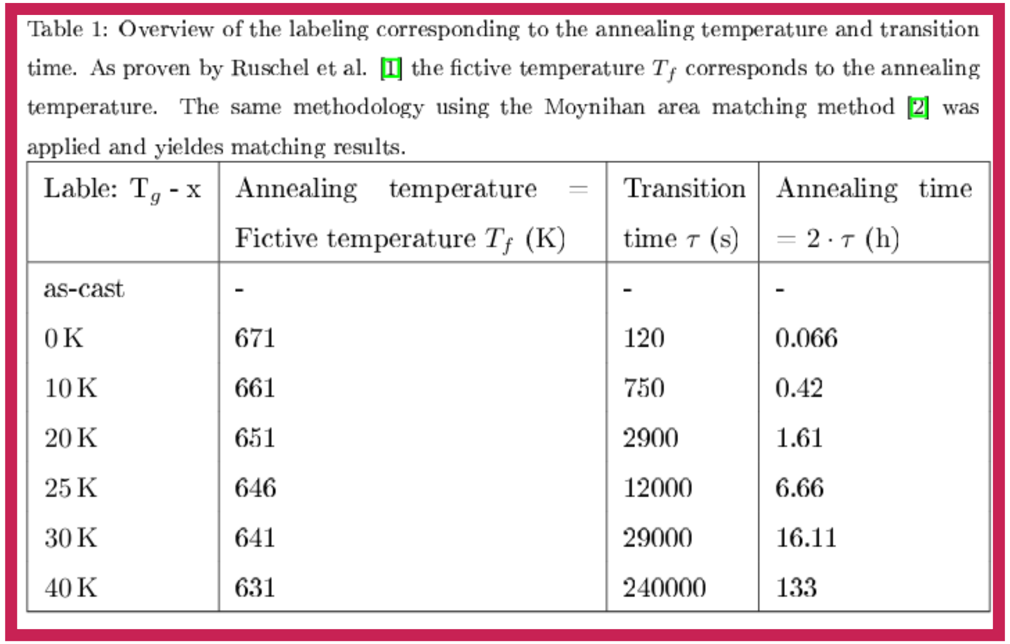


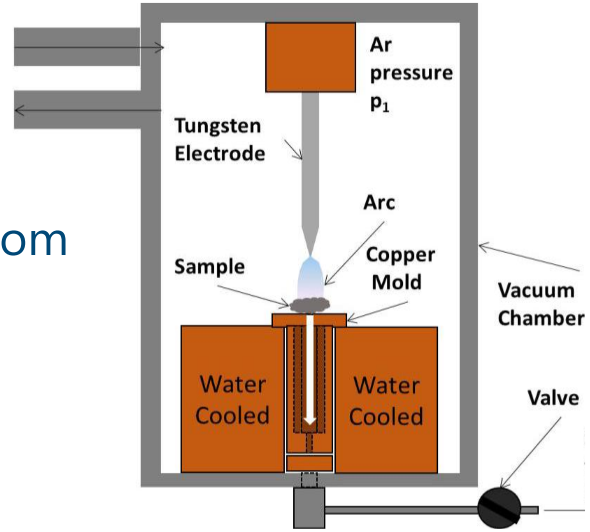
Table 1: Overview of the labeling corresponding to the annealing temperature and transition time. As proven by Ruschel et al. [1] the fictive temperature T_f corresponds to the annealing temperature. The same methodology using the Moynihan area matching method [2] was applied and yieldes matching results.

Lable: T_g - x	Annealing temperature = Fictive temperature T_f (K)	Transition time τ (s)	Annealing time $= 2 \cdot \tau$ (h)
as-cast	-	-	-
0 K	671	120	0.066
10 K	661	750	0.42
20 K	651	2900	1.61
25 K	646	12000	6.66
30 K	641	29000	16.11
40 K	631	240000	133

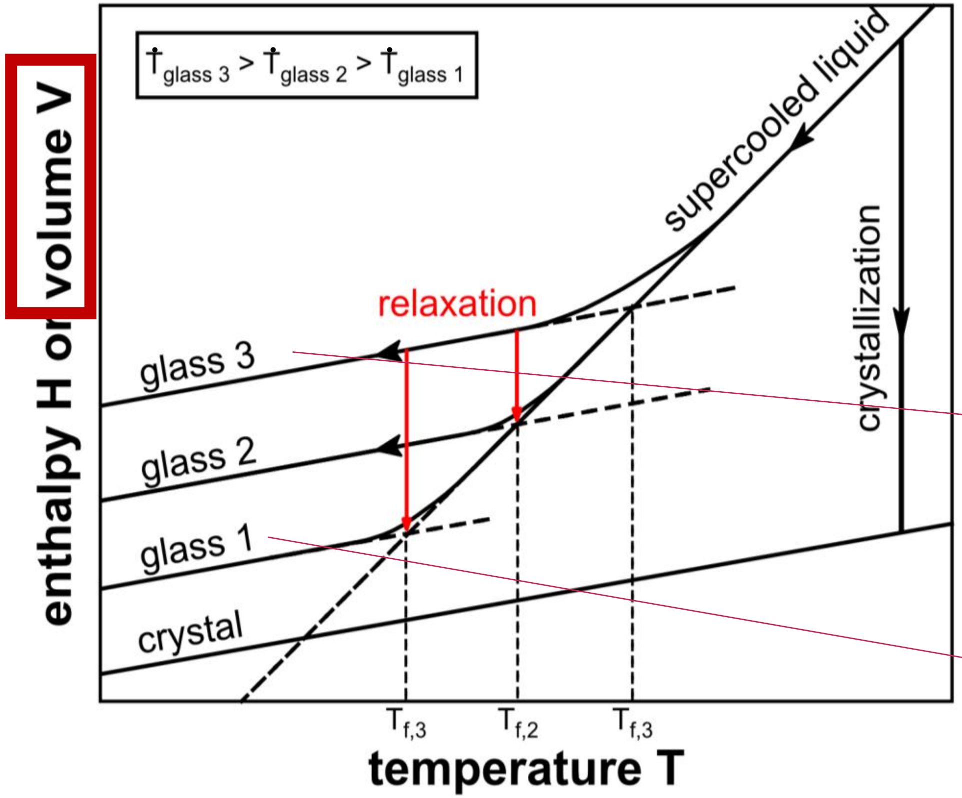
Material Preparation

Alloy casting

1. Weighing
2. Semi-finished products from the electric arc furnace
3. Suction casting system



Relaxation in sub-Tg



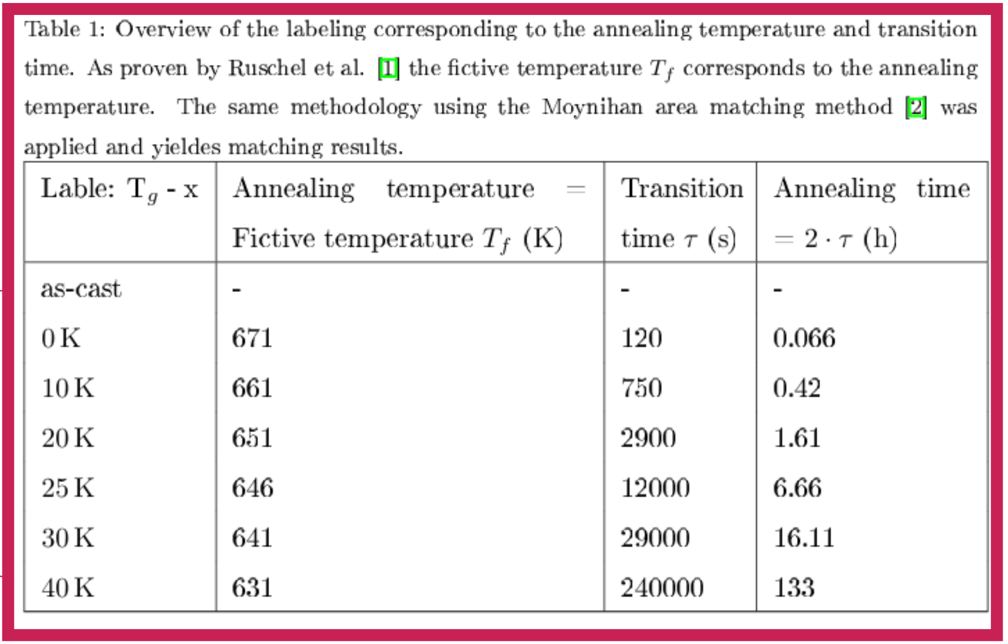


Table 1: Overview of the labeling corresponding to the annealing temperature and transition time. As proven by Ruschel et al. [1] the fictive temperature T_f corresponds to the annealing temperature. The same methodology using the Moynihan area matching method [2] was applied and yieldes matching results.

Lable: T_g - x	Annealing temperature = Fictive temperature T_f (K)	Transition time τ (s)	Annealing time = $2 \cdot \tau$ (h)
as-cast	-	-	-
0 K	671	120	0.066
10 K	661	750	0.42
20 K	651	2900	1.61
25 K	646	12000	6.66
30 K	641	29000	16.11
40 K	631	240000	133

Materials Today Advances 23 (2024) 100522

Contents lists available at ScienceDirect

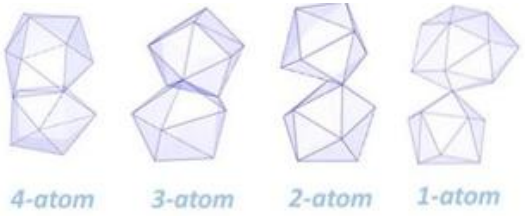
Materials Today Advances

journal homepage: www.journals.elsevier.com/materials-today-advances/

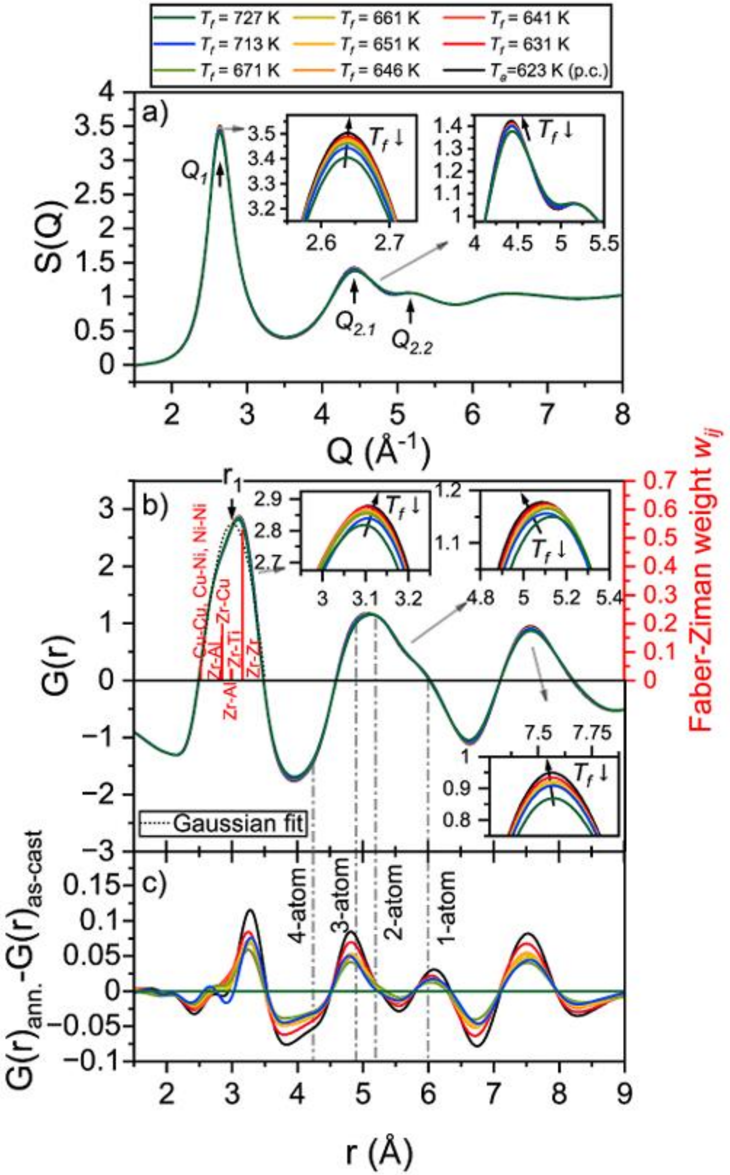
Unraveling the role of relaxation and rejuvenation on the structure and deformation behavior of the Zr-based bulk metallic glass Vit105

Lucas M. Ruschel [a,*], Sergej Jakovlev [a], Oliver Gross [b], Nico Neuber [a], Bastian Adam [a], Maximilian Frey [a], Benedikt Schmidt [a], Benedikt Bochtler [b], Ralf Busch [a]

[a] Chair of Metallic Materials, Saarland University, 66123, Saarbrücken, Germany
[b] Amorphous Metal Solutions GmbH, 66424, Homburg, Germany



@Ding, J., *et al.* *Sci Rep* **5**, 17429 (2015).



Materials Today Advances 23 (2024) 100522

Contents lists available at ScienceDirect

Materials Today Advances

journal homepage: www.journals.elsevier.com/materials-today-advances/

Unraveling the role of relaxation and rejuvenation on the structure and deformation behavior of the Zr-based bulk metallic glass Vit105

Lucas M. Ruschel [a,*], Sergej Jakovlev [a], Oliver Gross [b], Nico Neuber [a], Bastian Adam [a], Maximilian Frey [a], Benedikt Schmidt [a], Benedikt Bochtler [b], Ralf Busch [a]

[a] Chair of Metallic Materials, Saarland University, 66123, Saarbrücken, Germany
[b] Amorphous Metal Solutions GmbH, 66424, Homburg, Germany

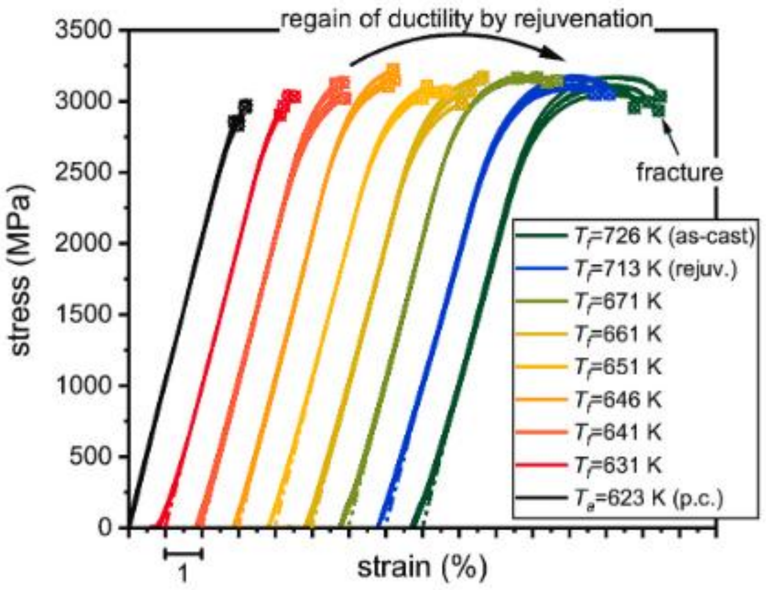
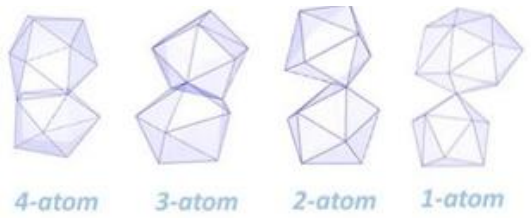
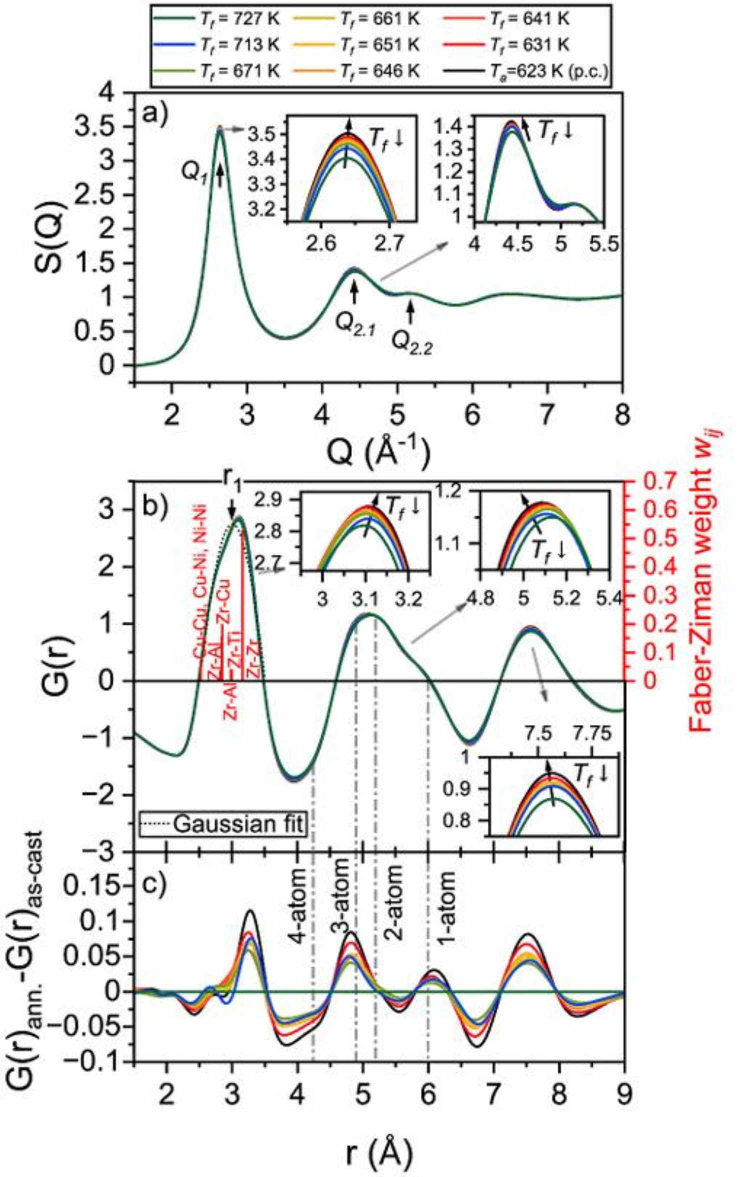


Fig. 4. Engineering stress-strain curves for different fictive temperatures T_f, revealing significant embrittlement with decreasing T_f. The deepest annealing temperature (T_a = 623 K, black curves) is affected by partial crystallization (p. c.) as revealed in DSC experiments.



@Ding, J., *et al.* *Sci Rep* **5**, 17429 (2015).



Materials Today Advances 23 (2024) 100522

Contents lists available at ScienceDirect

Materials Today Advances

journal homepage: www.journals.elsevier.com/materials-today-advances/

Unraveling the role of relaxation and rejuvenation on the structure and deformation behavior of the Zr-based bulk metallic glass Vit105

Lucas M. Ruschel [a,*], Sergej Jakovlev [a], Oliver Gross [b], Nico Neuber [a], Bastian Adam Maximilian Frey [a], Benedikt Schmidt [a], Benedikt Bochtler [b], Ralf Busch [a]

[a] Chair of Metallic Materials, Saarland University, 66123, Saarbrücken, Germany

[b] Amorphous Metal Solutions GmbH, 66424, Homburg, Germany

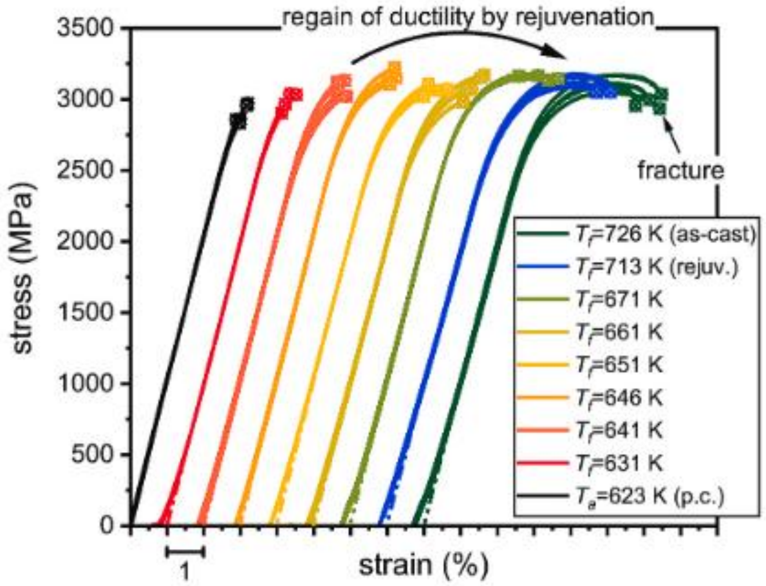
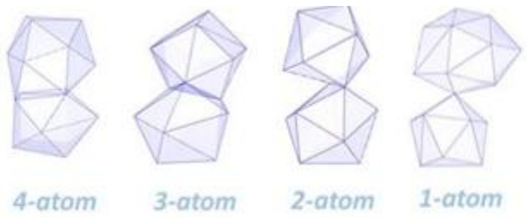
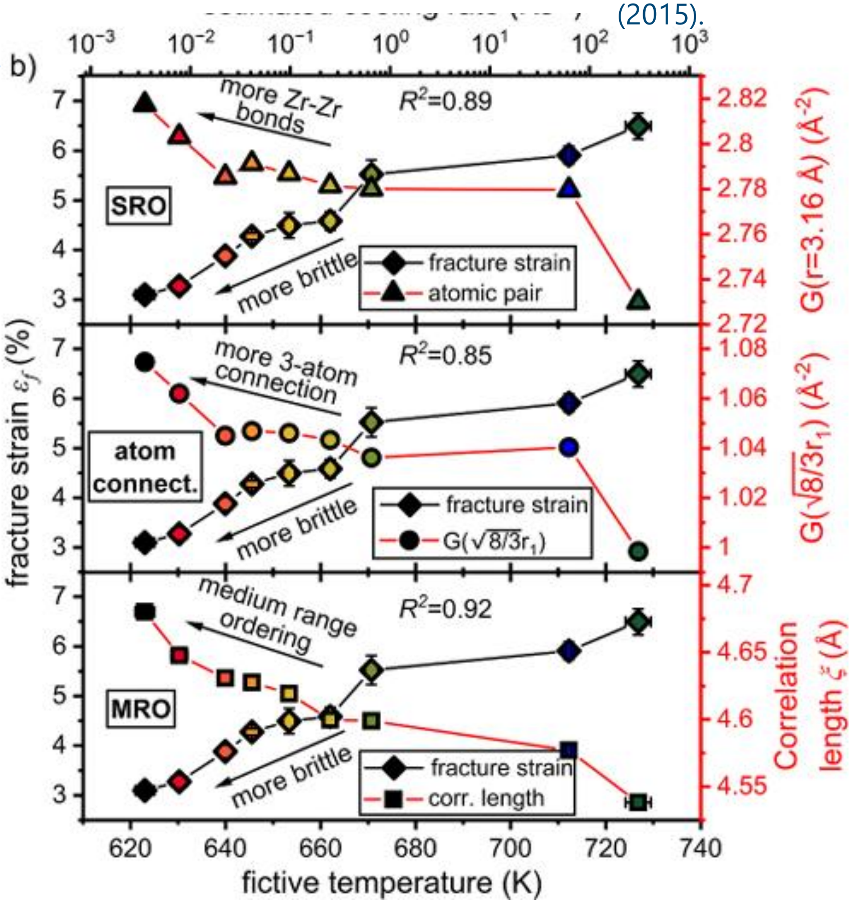
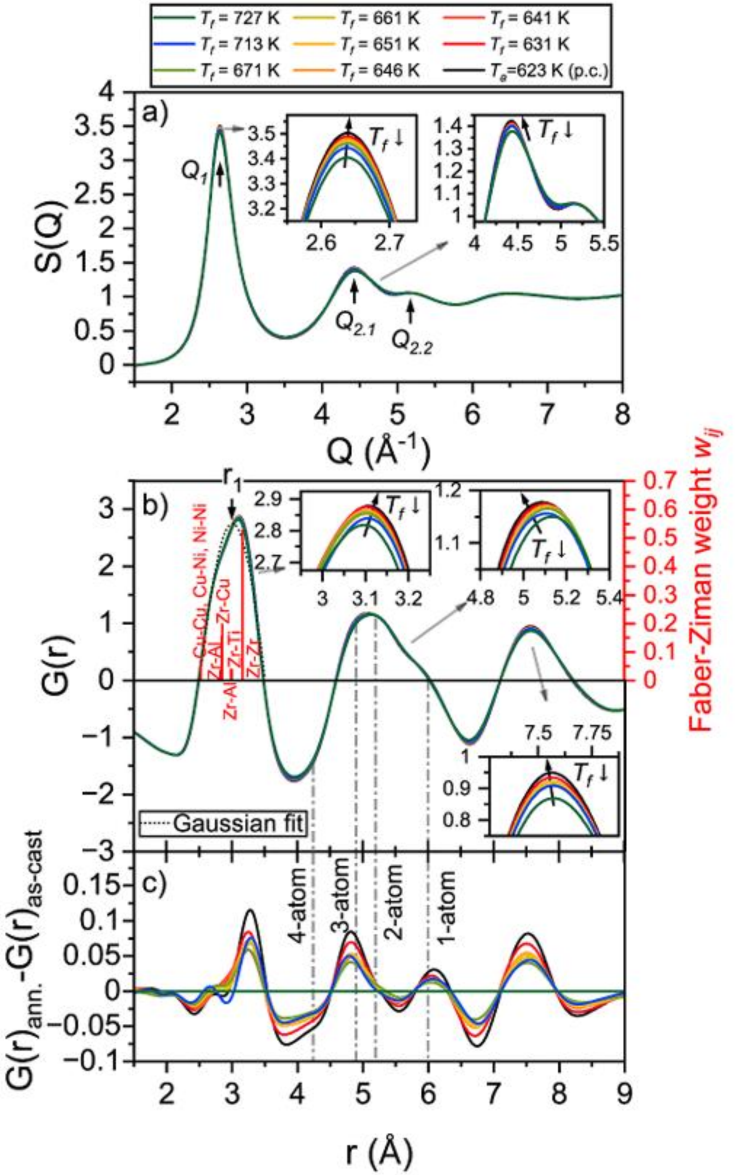


Fig. 4. Engineering stress-strain curves for different fictive temperatures T_f, revealing significant embrittlement with decreasing T_f. The deepest annealing temperature (T_a = 623 K, black curves) is affected by partial crystallization (p.c.) as revealed in DSC experiments.



@Ding, J., *et al.* *Sci Rep* **5**, 17429 (2015).





Materials Today Advances 23 (2024) 100522

Contents lists available at ScienceDirect

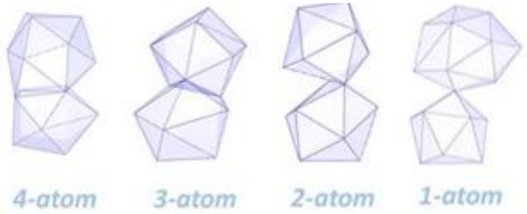
Materials Today Advances

journal homepage: www.journals.elsevier.com/materials-today-advances/

Unraveling the role of relaxation and rejuvenation on the structure and deformation behavior of the Zr-based bulk metallic glass Vit105

Lucas M. Ruschel [a,*], Sergej Jakovlev [a], Oliver Gross [b], Nico Neuber [a], Bastian Adam Maximilian Frey [a], Benedikt Schmidt [a], Benedikt Bochtler [b], Ralf Busch [a]

[a] Chair of Metallic Materials, Saarland University, 66123, Saarbrücken, Germany
[b] Amorphous Metal Solutions GmbH, 66424, Homburg, Germany



@Ding, J., *et al.* *Sci Rep* **5**, 17429 (2015).

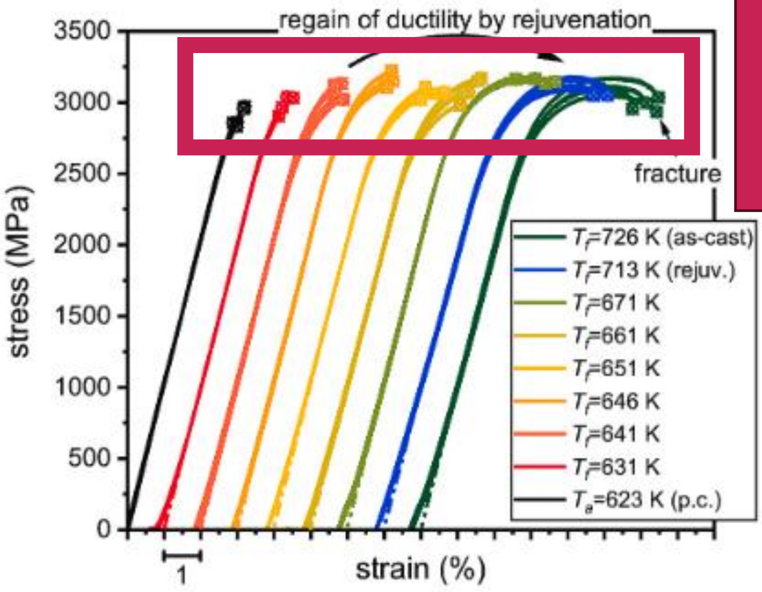
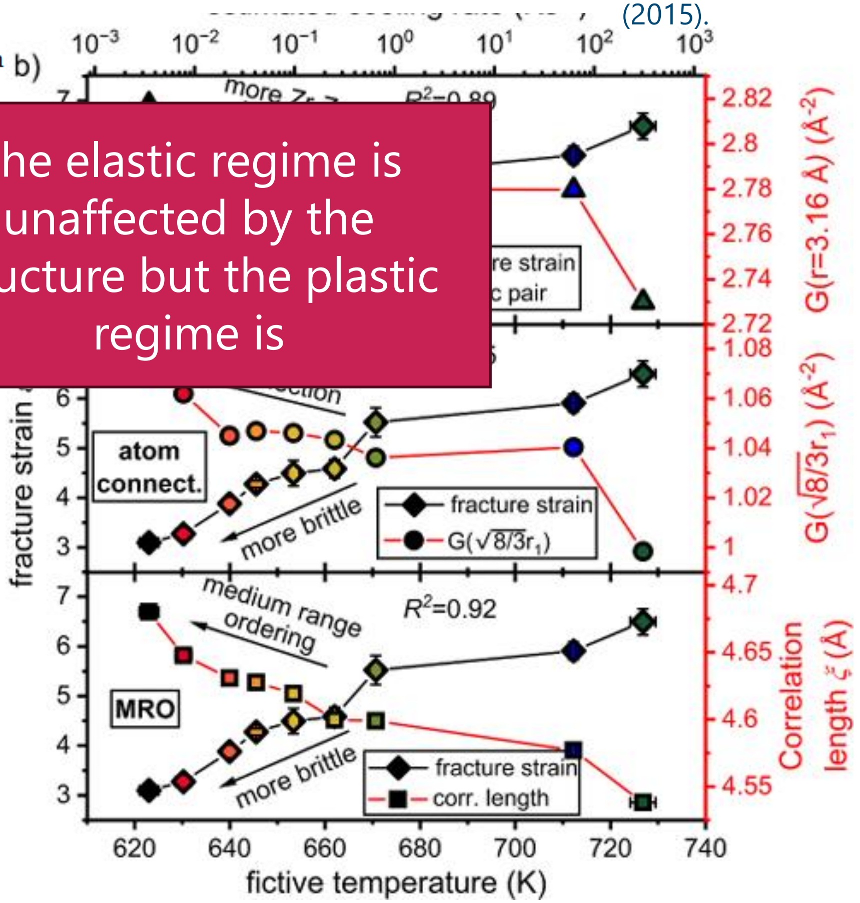
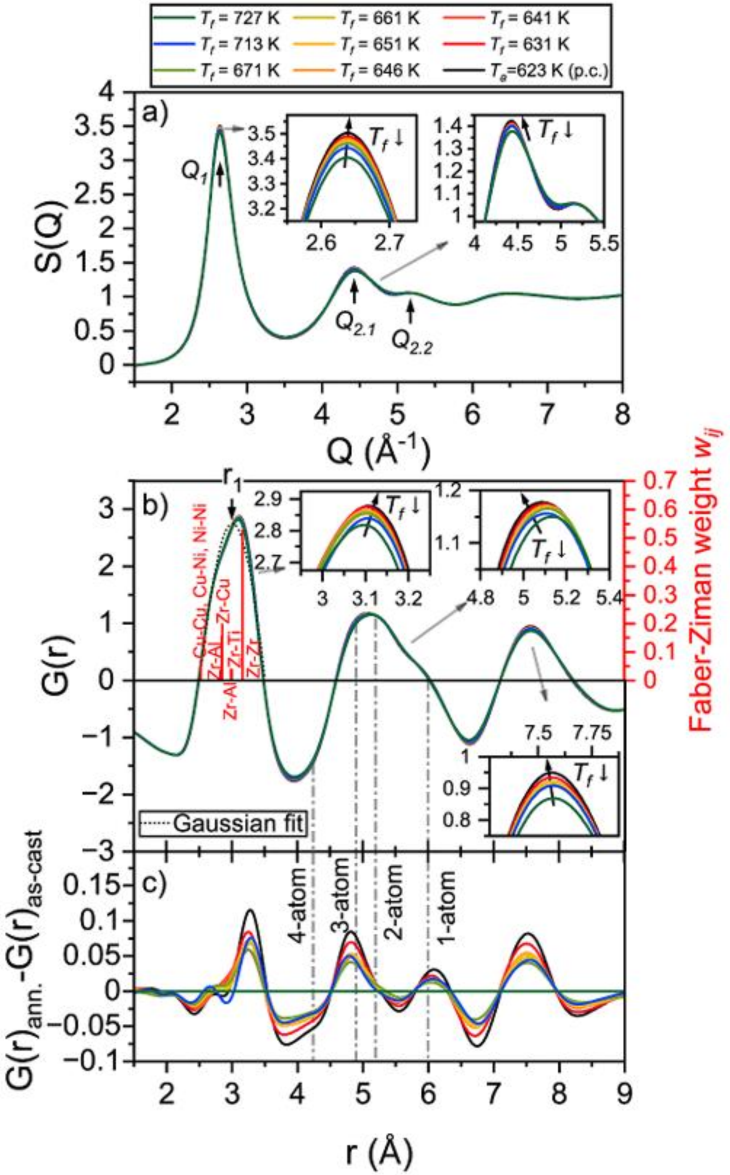


Fig. 4. Engineering stress-strain curves for different fictive temperatures T_f, revealing significant embrittlement with decreasing T_f. The deepest annealing temperature (T_a = 623 K, black curves) is affected by partial crystallization (p.c.) as revealed in DSC experiments.

The elastic regime is unaffected by the structure but the plastic regime is





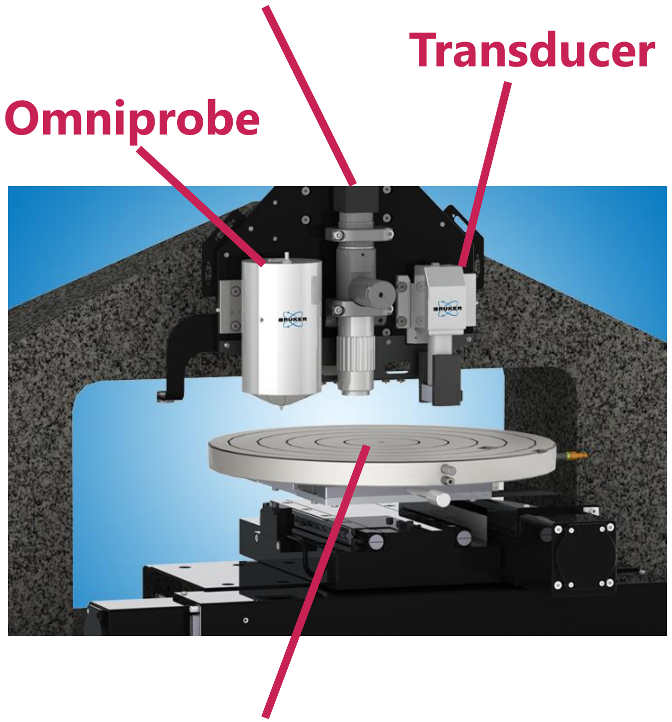
Nanoindentation

Constant Strain Rate
$\dot{\varepsilon}$ = 0.01; 0.025; 0.1; 0.25; 1
100-200 nm Depth

Bruker TI990 Triboindenter:

Low Load Transducer

@www.bruker.com

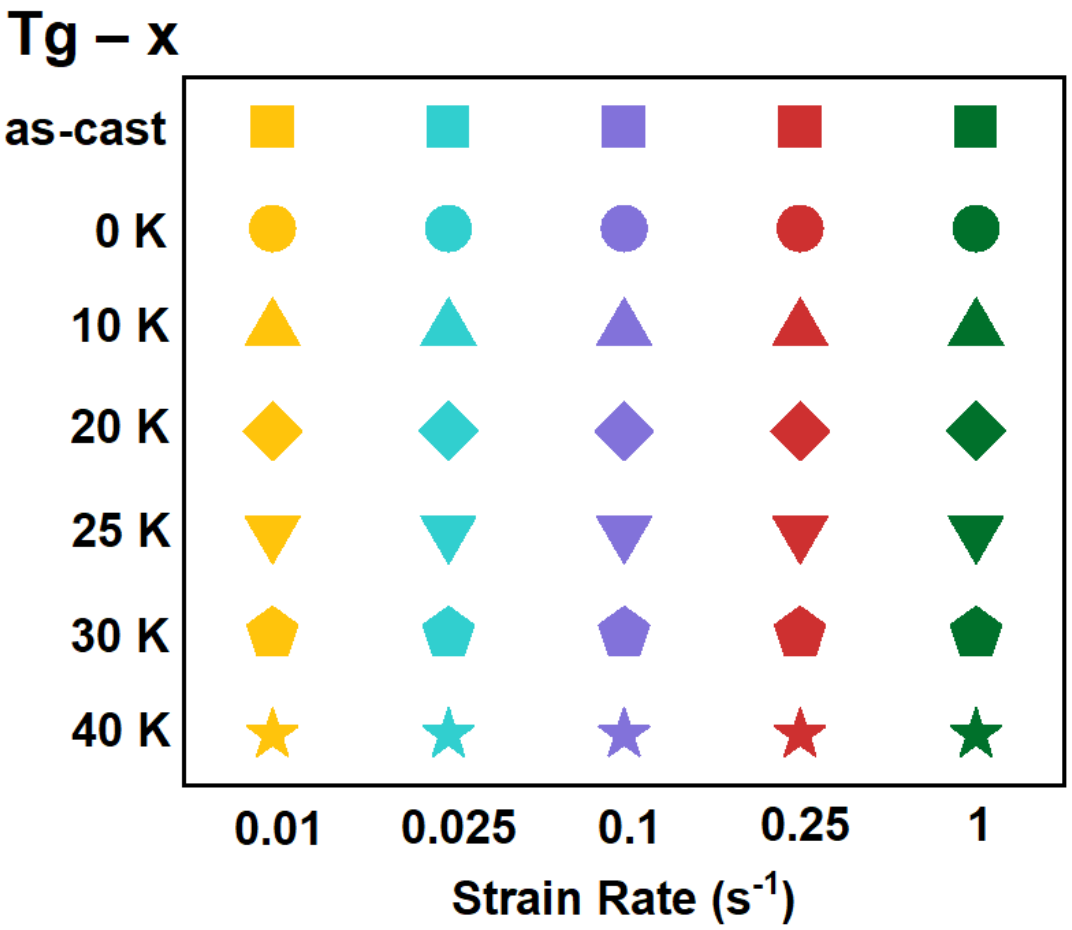


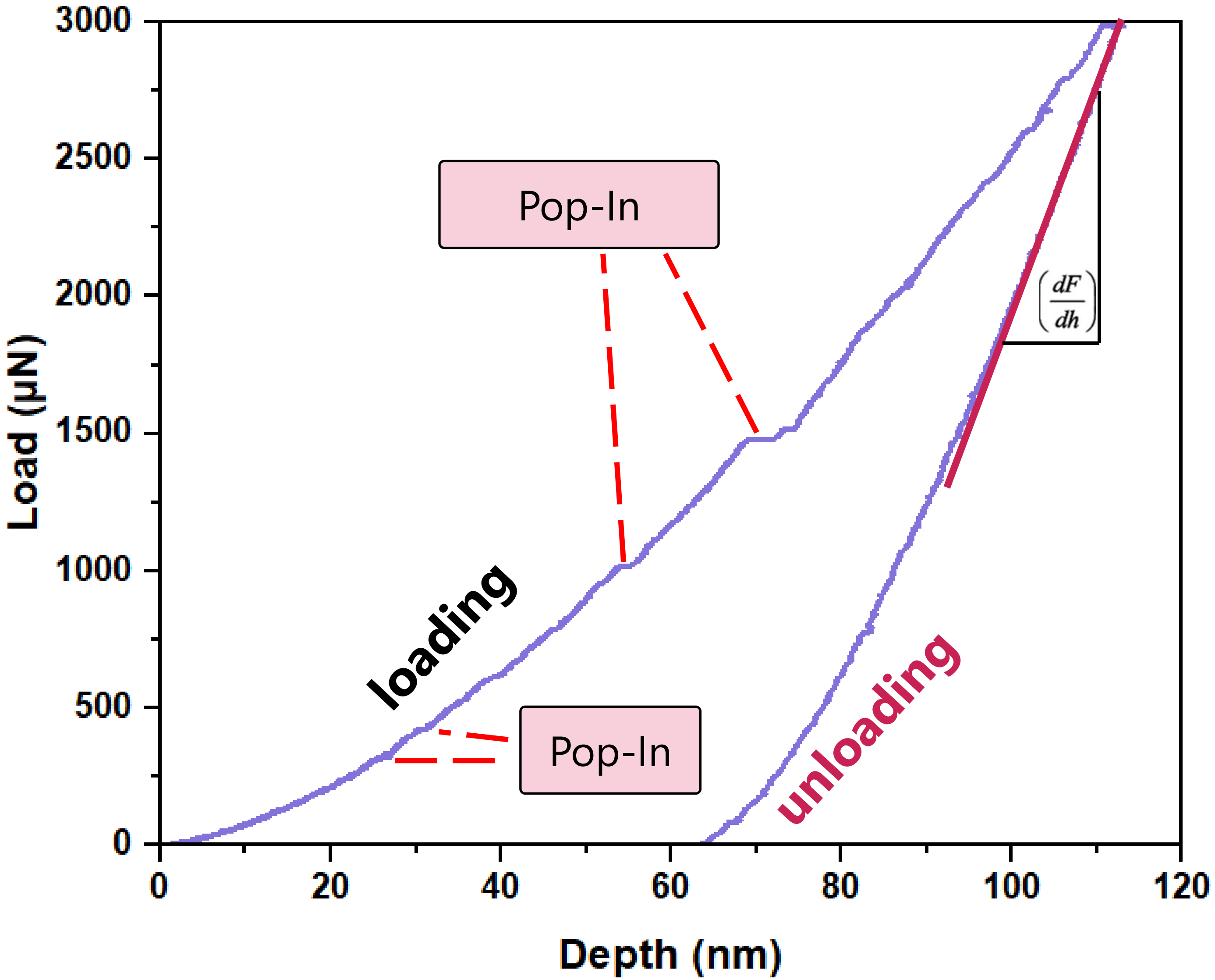
Optics

Tiboscanner and Transducer

Omniprobe

Sample table





UNIVERSITÄT DES SAARLANDES

Unload Segment

Using Oliver-Pharr Method

E-modul

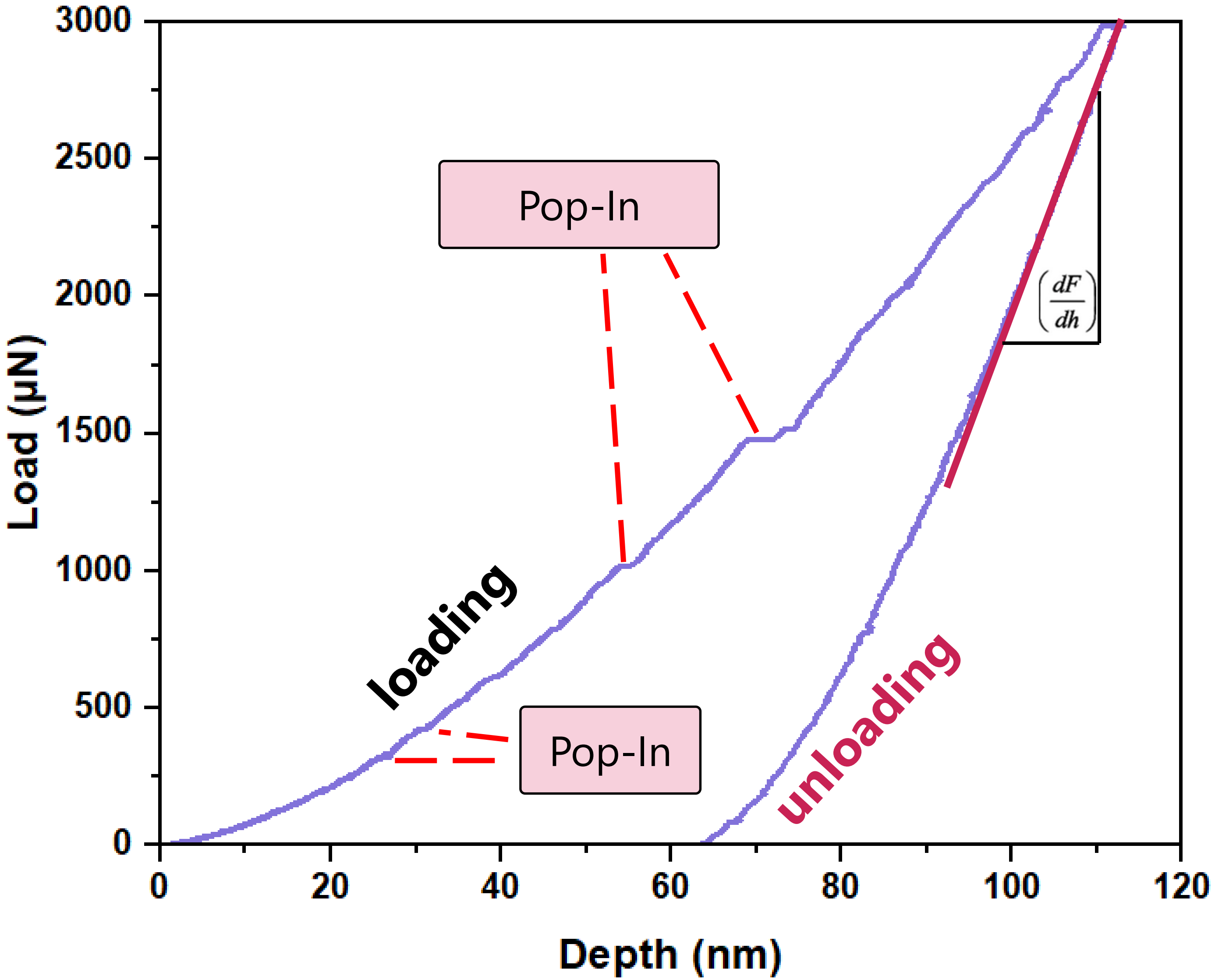
$$E^* = \frac{1}{2}\left(\frac{dF}{dh}\right)_{h_m}\sqrt{\frac{\pi}{A}}$$

Hardness

$$H = \left(\frac{F}{A}\right)_{hm}$$

Strain Rate Sensitivity

$$m = \frac{lgH}{lg\dot{\varepsilon}}$$

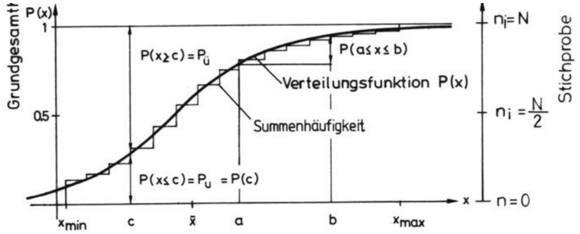


Load Segment

Pop-In Position from MatLab Script

P and hc from Pop-Ins

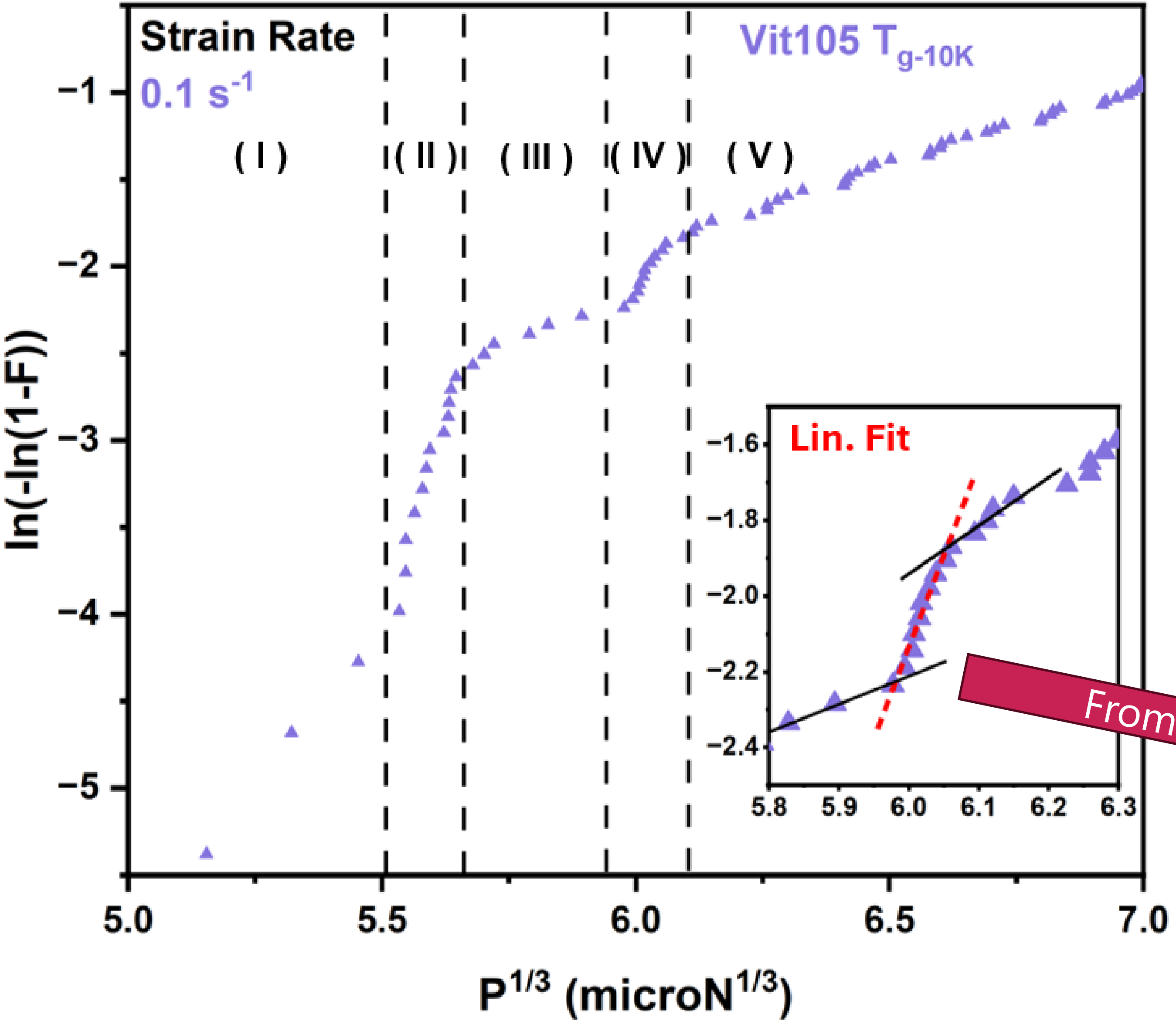
Cumulative Distribution Function



Activation Volume

STZ Volume and Number of Atoms in STZs

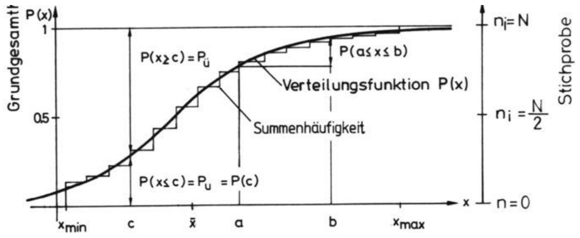
Load Segment



Pop-In Position from MatLab Script

P and hc from Pop-Ins

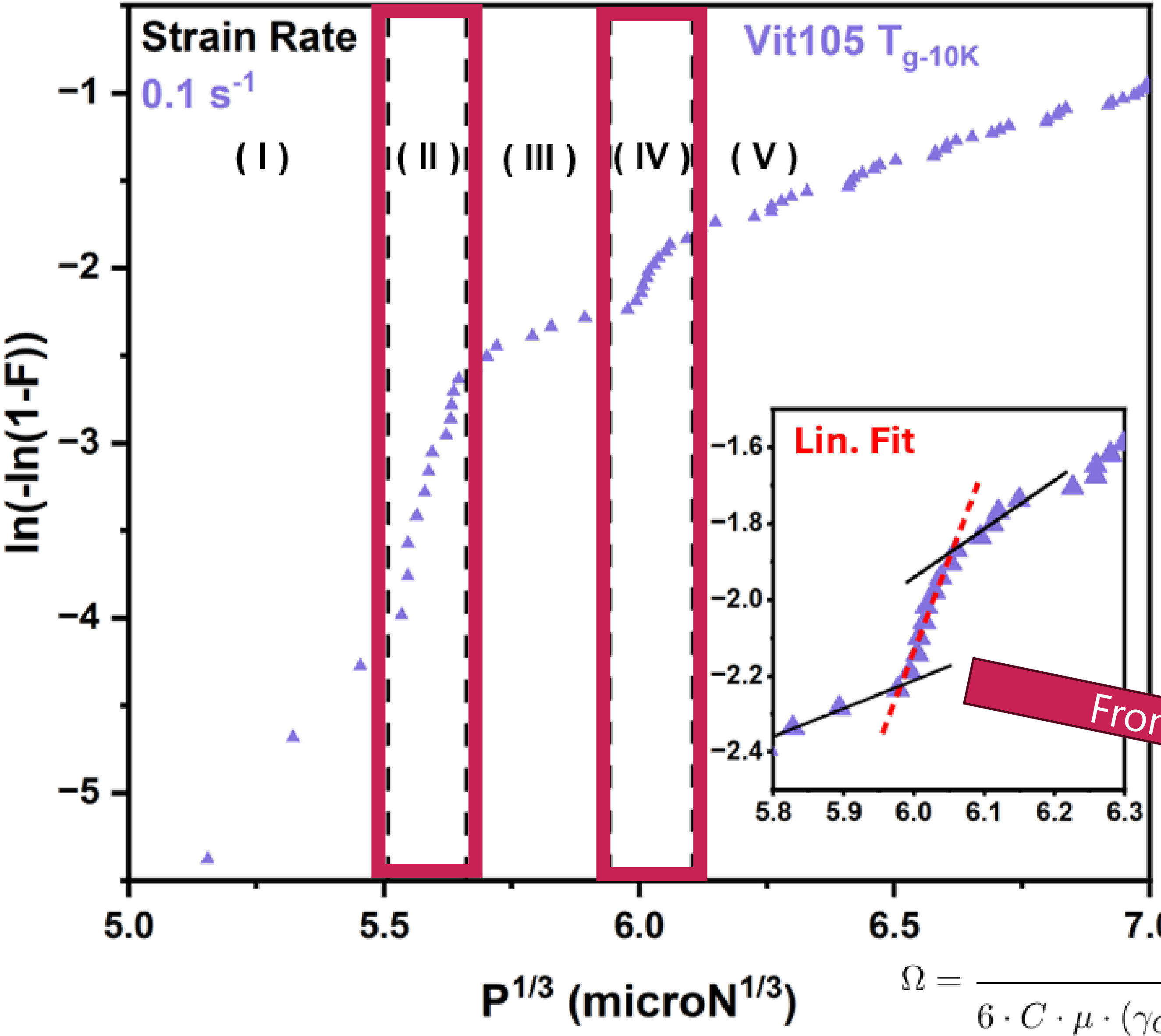
Cumulative Distribution Function

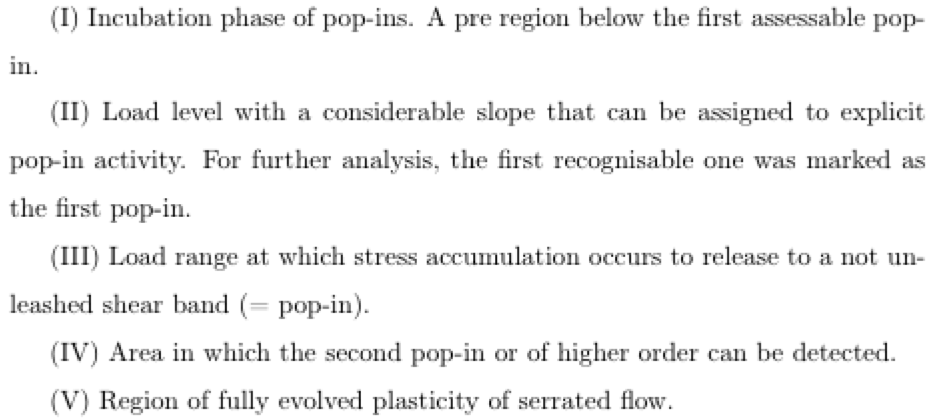


From Slope

Activation Volume

STZ Volume and Number of Atoms in STZs



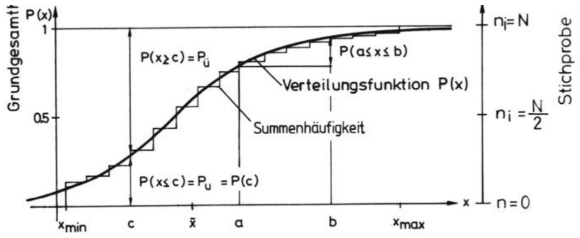

(I) Incubation phase of pop-ins. A pre region below the first assessable pop-in.

(II) Load level with a considerable slope that can be assigned to explicit pop-in activity. For further analysis, the first recognisable one was marked as the first pop-in.

(III) Load range at which stress accumulation occurs to release to a not unleashed shear band (= pop-in).

(IV) Area in which the second pop-in or of higher order can be detected.

(V) Region of fully evolved plasticity of serrated flow.



From Slope

Activation Volume

$$V = \alpha \cdot kT \cdot \frac{\pi}{0.47} \cdot \left(\frac{3\rho}{4E_R}\right)^{2/3}$$

STZ Volume

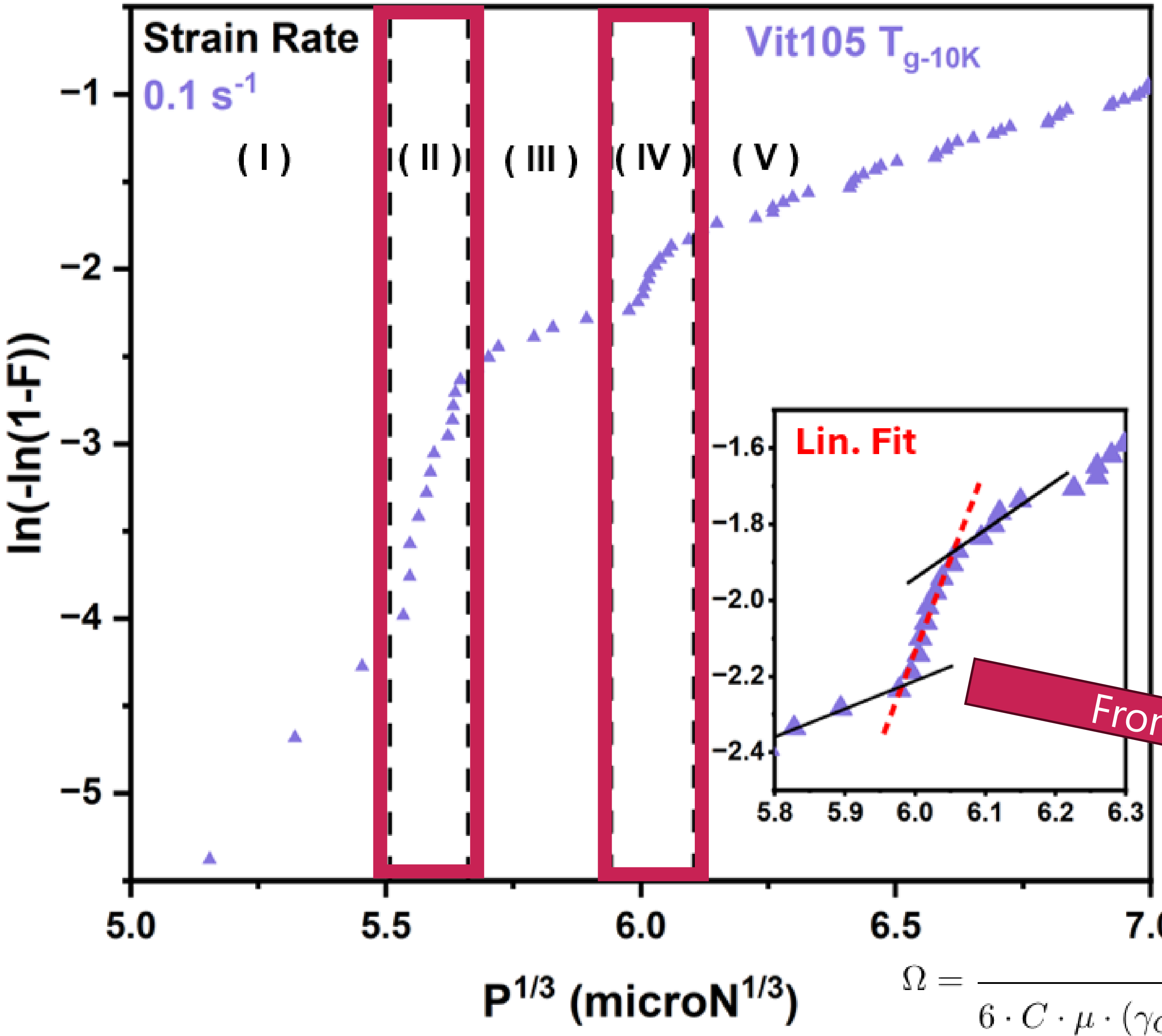
Shear strength

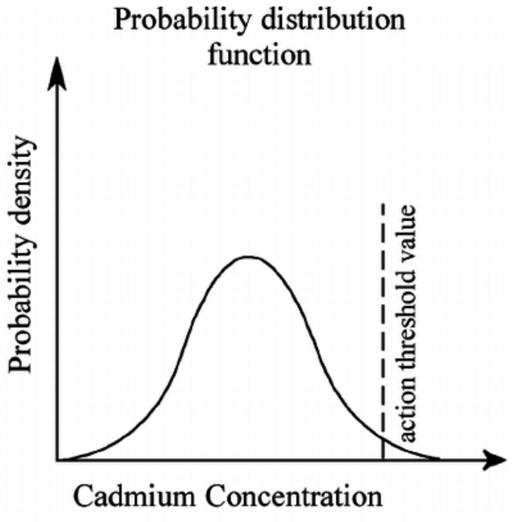
$$\Omega = \frac{\tau_{C0}}{6 \cdot C \cdot \mu \cdot (\gamma_C)^2 \cdot \xi \sqrt{\left(1 - \frac{\tau_C}{\tau_{C0}}\right)}} \cdot V$$

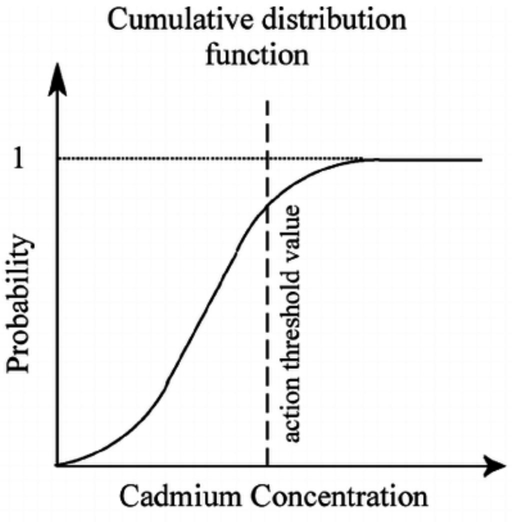
Shear modulus

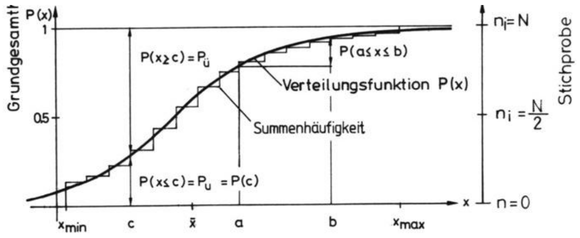
Flow stress

Estimation through Density
Number of Atoms in STZs









From Slope

Activation Volume

$$V = \alpha \cdot kT \cdot \frac{\pi}{0.47} \cdot \left(\frac{3\rho}{4E_R}\right)^{2/3}$$

STZ Volume

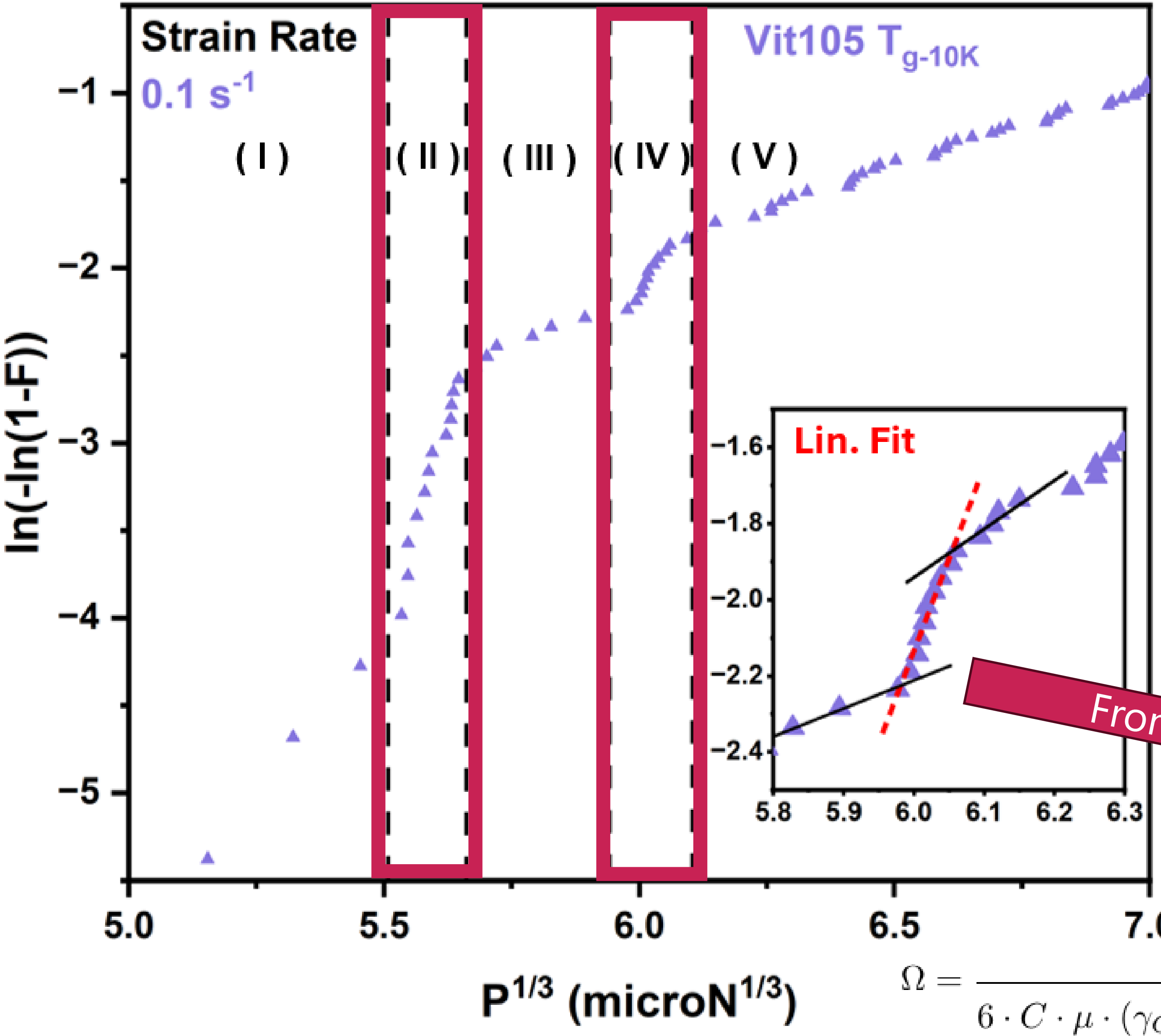
Shear strength

$$\Omega = \frac{\tau_{C0}}{6 \cdot C \cdot \mu \cdot (\gamma_C)^2 \cdot \xi \sqrt{\left(1 - \frac{\tau_C}{\tau_{C0}}\right)}} \cdot V$$

Shear modulus

Flow stress

Estimation through Density
Number of Atoms in STZs



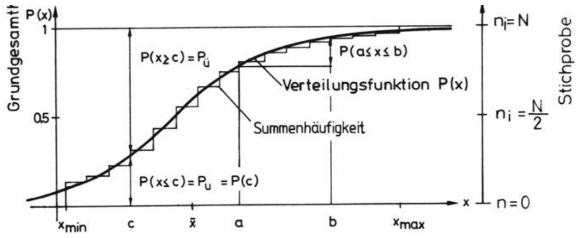
(I) Incubation phase of pop-ins. A pre region below the first assessable pop-in.

(II) Load level with a considerable slope that can be assigned to explicit pop-in activity. For further analysis, the first recognisable one was marked as the first pop-in.

(III) Load range at which stress accumulation occurs to release to a not unleashed shear band (= pop-in).

(IV) Area in which the second pop-in or of higher order can be detected.

(V) Region of fully evolved plasticity of serrated flow.



From Slope

Activation Volume

$$V = \alpha \cdot kT \cdot \frac{\pi}{0.47} \cdot (\frac{3\rho}{4E_R})^{2/3}$$

STZ Volume

Shear strength

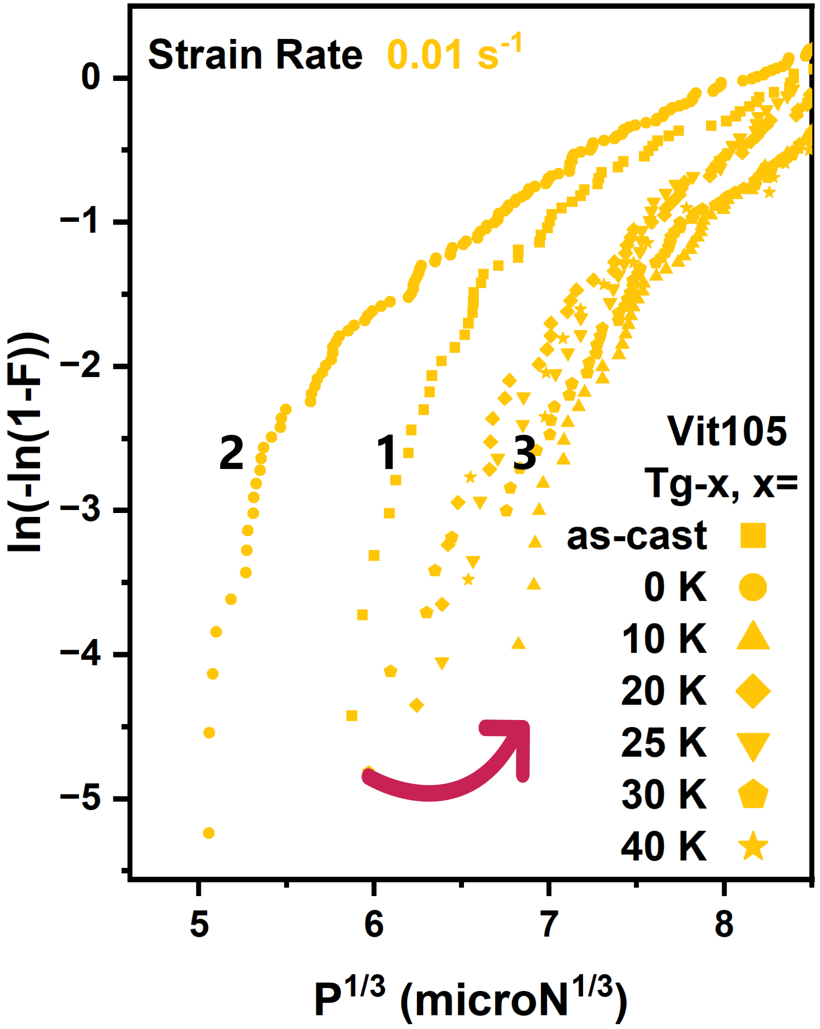
$$\Omega = \frac{\tau_{C0}}{6 \cdot C \cdot \mu \cdot (\gamma_C)^2 \cdot \xi \sqrt{(1 - \frac{\tau_C}{\tau_{C0}})}} \cdot V$$

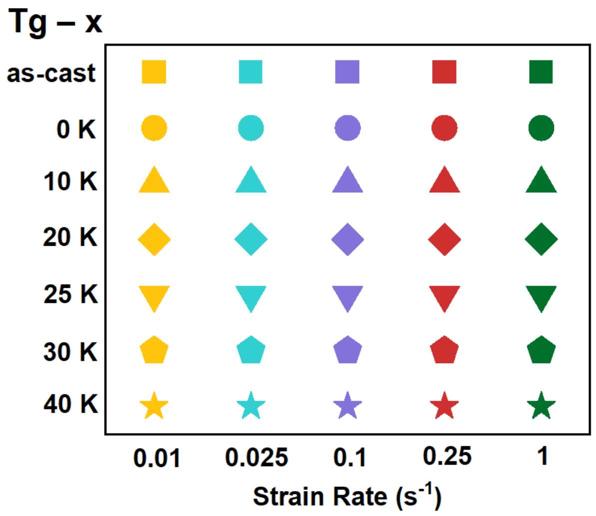
Shear modulus

Flow stress

Estimation through Density
Number of Atoms in STZs

Empirical Cumulative Distribution





Representative for all other Strain Rates

1. As-cast
2. 0K
3. 10K, 20K, 25K, 30K and 40K -> Clustering (no difference)

Evidence for internal stresses (Zug) after casting

Onset of Plasticity shifts to higher critical loads through structural relaxation

STZ Volume and Number of Atoms in STZs

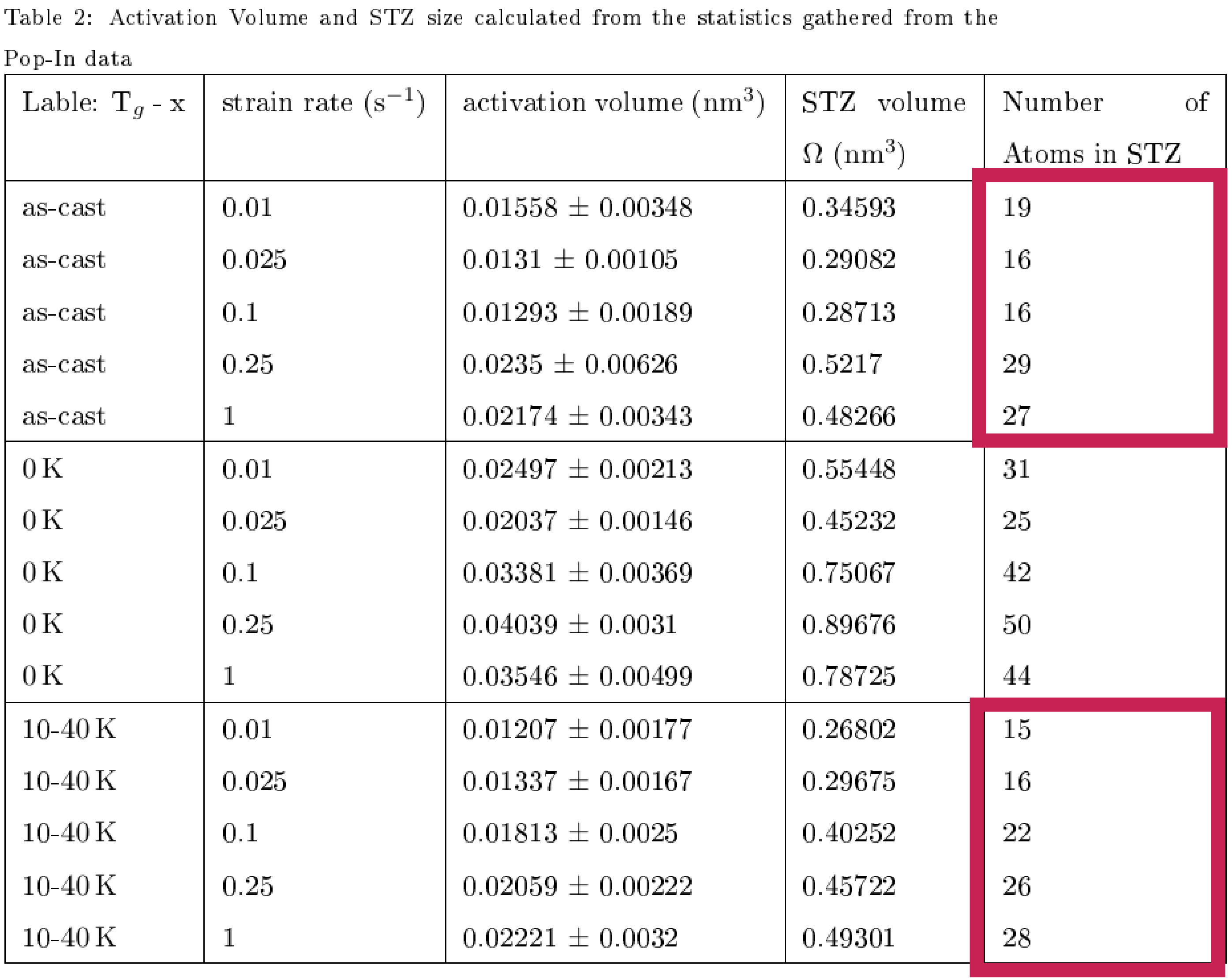
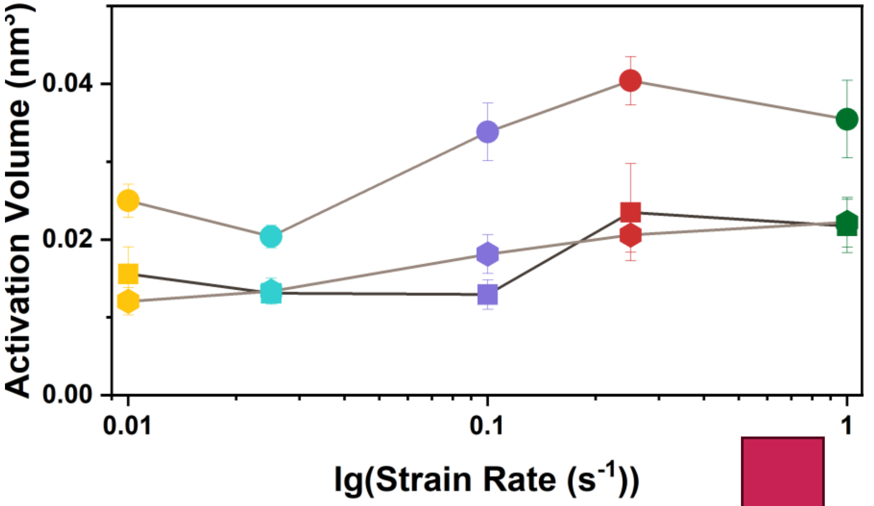


Table 2: Activation Volume and STZ size calculated from the statistics gathered from the Pop-In data

Lable: T_g - x	strain rate (s^{-1})	activation volume (nm^3)	STZ volume Ω (nm^3)	Number of Atoms in STZ
as-cast	0.01	0.01558 ± 0.00348	0.34593	19
as-cast	0.025	0.0131 ± 0.00105	0.29082	16
as-cast	0.1	0.01293 ± 0.00189	0.28713	16
as-cast	0.25	0.0235 ± 0.00626	0.5217	29
as-cast	1	0.02174 ± 0.00343	0.48266	27
0 K	0.01	0.02497 ± 0.00213	0.55448	31
0 K	0.025	0.02037 ± 0.00146	0.45232	25
0 K	0.1	0.03381 ± 0.00369	0.75067	42
0 K	0.25	0.04039 ± 0.0031	0.89676	50
0 K	1	0.03546 ± 0.00499	0.78725	44
10-40 K	0.01	0.01207 ± 0.00177	0.26802	15
10-40 K	0.025	0.01337 ± 0.00167	0.29675	16
10-40 K	0.1	0.01813 ± 0.0025	0.40252	22
10-40 K	0.25	0.02059 ± 0.00222	0.45722	26
10-40 K	1	0.02221 ± 0.0032	0.49301	28

Plastic deformation is scale-dependent



Activation Volume is unaffected by the structural relaxation (3-atom connection)

STZ Volume and Number of Atoms in STZs

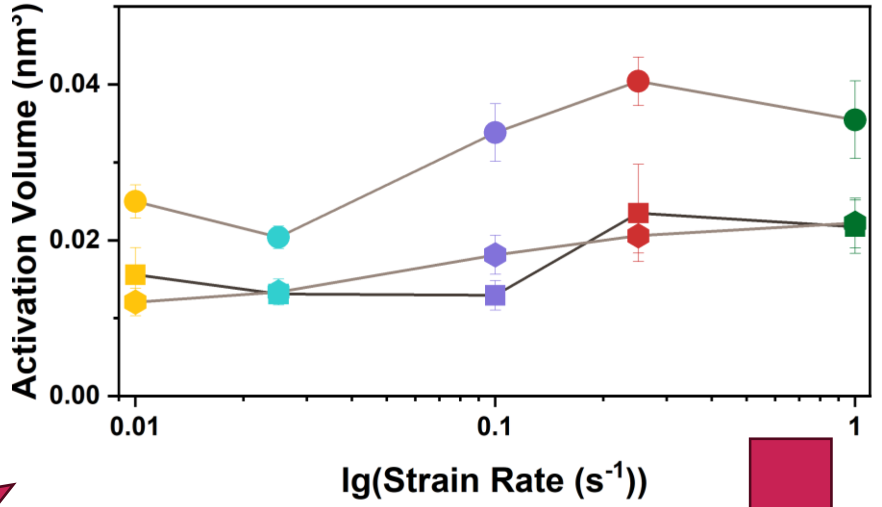
Key Findings:

- Elastic regime: Unaffected by structural relaxation
- Plastic regime: Strongly influenced by structural relaxation

But

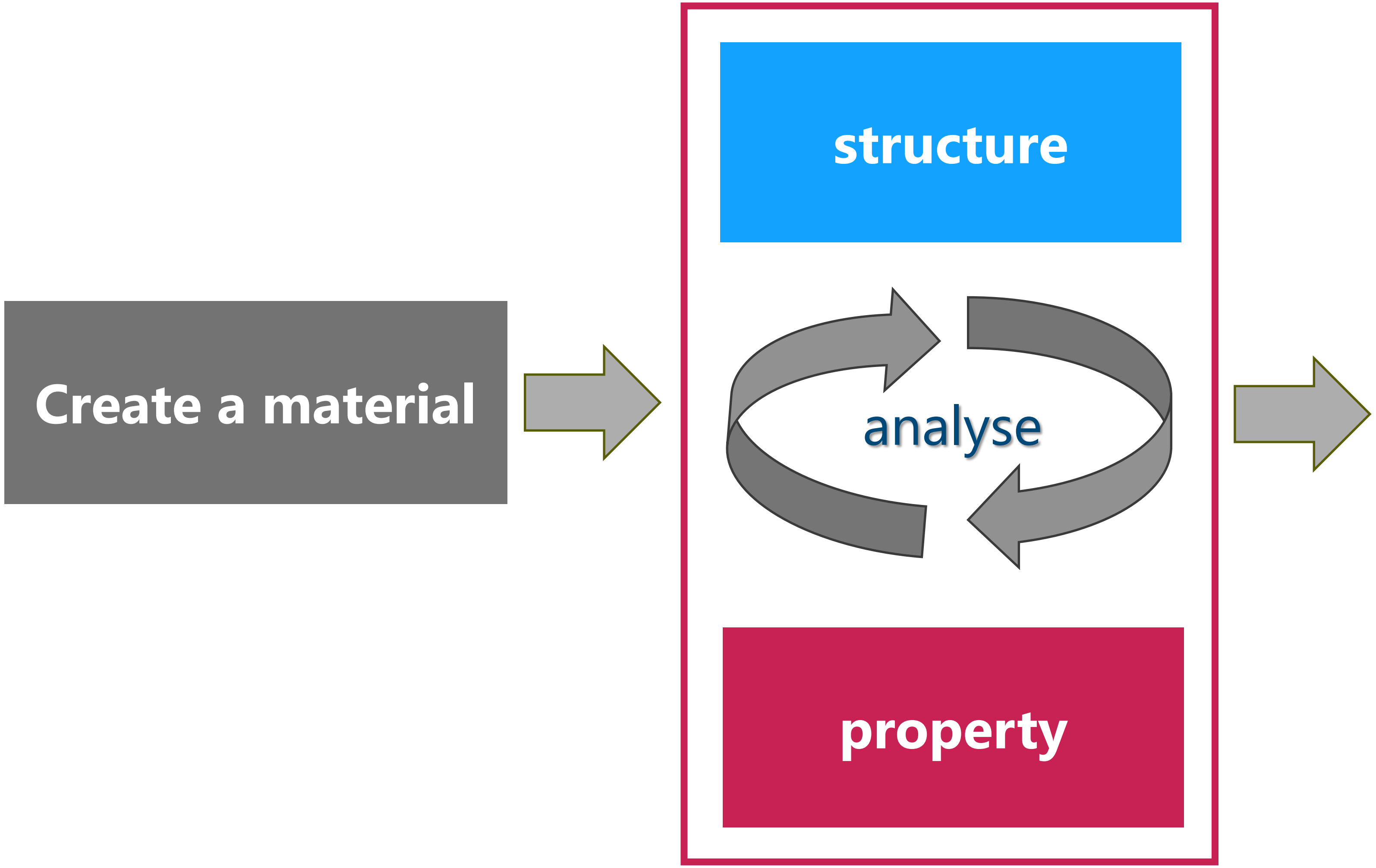
Activation Volume remains **constant**

Plastic deformation is scale-dependent



Activation Volume is unaffected by the structural relaxation (3-atom connection)

What does a (material)scientist in research?


Create a material

structure

analyse

property

Is my material fit for my application?


@https://www.reuters.com/lifestyle/two-giant-rubber-ducks-debut-hong-kong-bid-drive-double-happiness-2023-06-09/

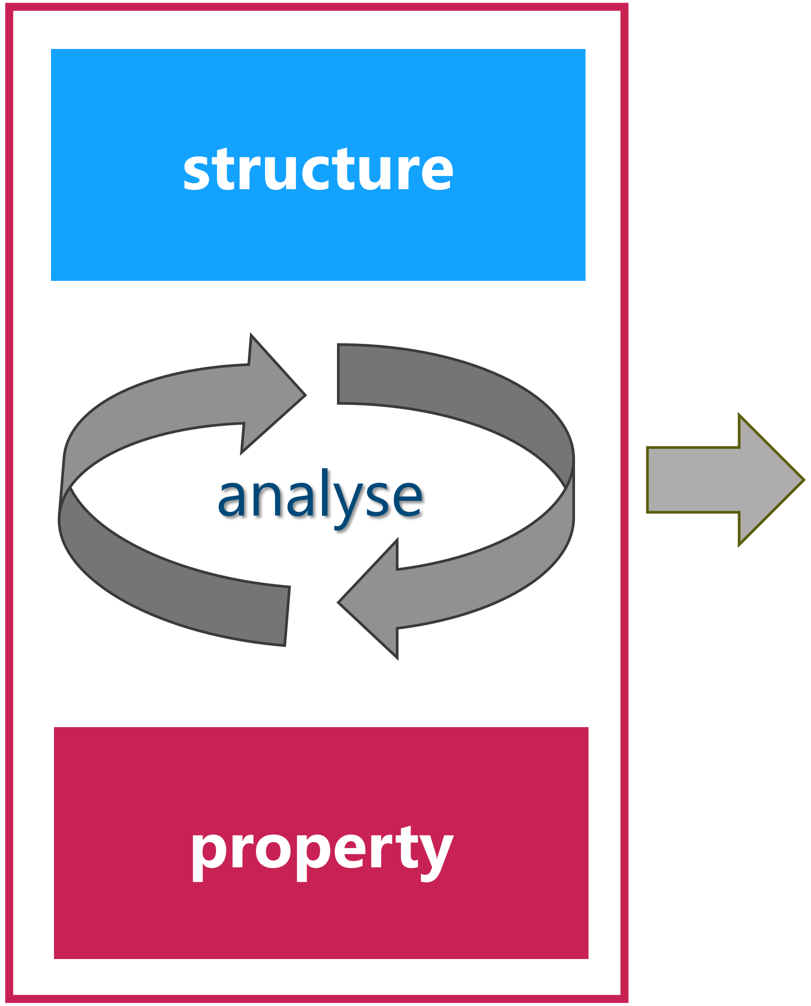
Can I predict **mechanical properties** from the **structural fingerprint?**

What does a (material)scientist in research?

Confirm: structural fingerprints can predict aspects of plasticity

Link: nano-scale events with atomic-scale mechanisms

Provide: Pathway to tailor mechanical properties via structural relaxation

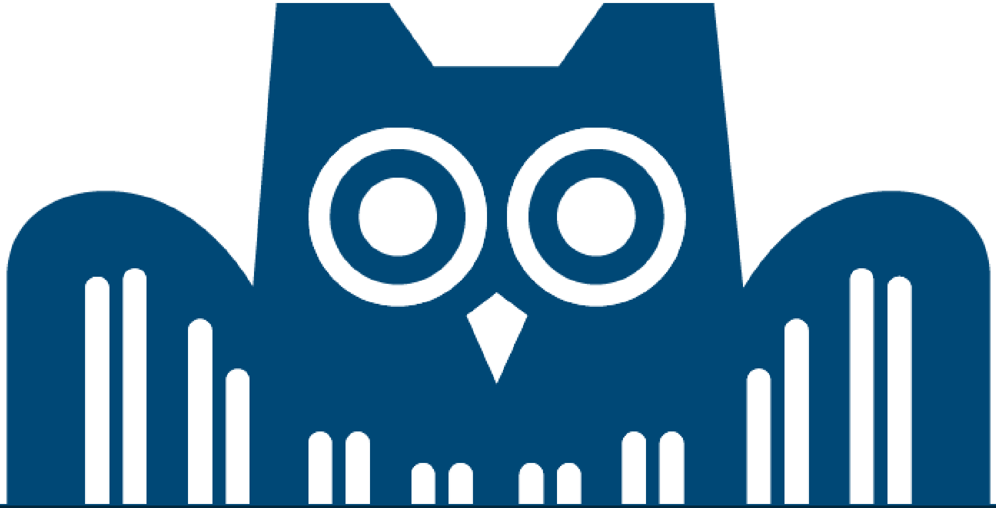


Is my material fit for my application?

@https://www.reuters.com/lifestyle/two-giant-rubber-ducks-debut-hong-kong-bid-drive-double-happiness-2023-06-09/

Can I predict **mechanical properties** from the **structural fingerprint?**

Thank You! Questions?



https://doi.org/10.1016/j.jallcom.2026.188302