Neutron Diffraction Studies of Micromechanics of Material Deformation

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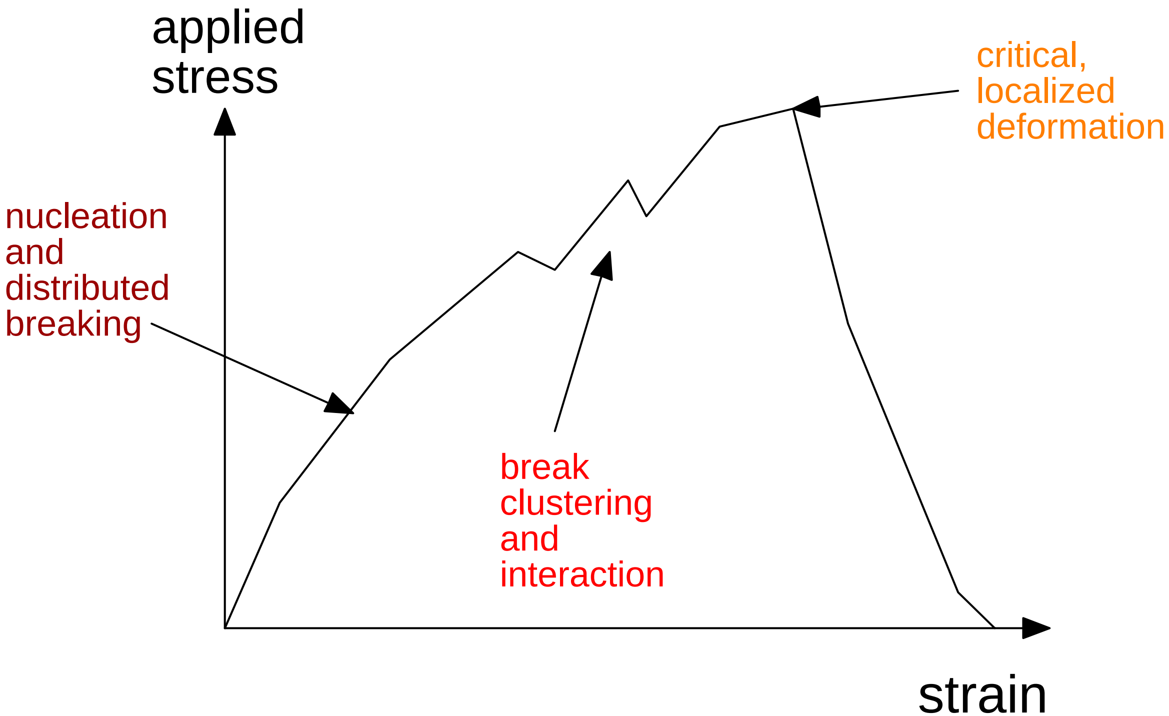
Outline

- **Introduction and motivation**
- **New engineering diffractometers: SMARTS and ENGIN-X**
- **Metallic glass composites**
- ***In-situ*-reinforced Si_3N_4**
- **Polycrystalline ferroelectrics**
- **Future directions: *DANSE***

Fracture of a Fiber Composite under Tension



- **Aim: prediction of strength and lifetime**
- **Need: "realistic" constitutive laws**

Complications

- **Fabrication processes**
- **Inhomogeneous dislocation densities**
- **Changes in grain size**
- **Geometrical constraints**
- **Interface introduced with different properties**
- **Residual stresses**

Motivation and Approach

- Little information about deformation and *in-situ* constitutive behavior of materials.
- Need to link experimental data with rigorous micromechanics modeling.
- Approach: Use neutron diffraction to investigate deformation in materials and complement it with modeling.
- Critical issues:
  - Need for model specimens
  - "High selectivity" of diffraction
  - Only elastic lattice strains are measured with diffraction
  - Lack of "realistic" constitutive laws to calculate stress and interpret diffraction data

Advantages of ND

- Non-destructive.
- Ability to distinguish different phases.
- Can measure elastic strain and texture.
- Multi-scale: *nm* to *cm*.
- Deep penetration.
- *In-situ* experiment capability.

$\Rightarrow$ Determination of *in-situ* constitutive behavior

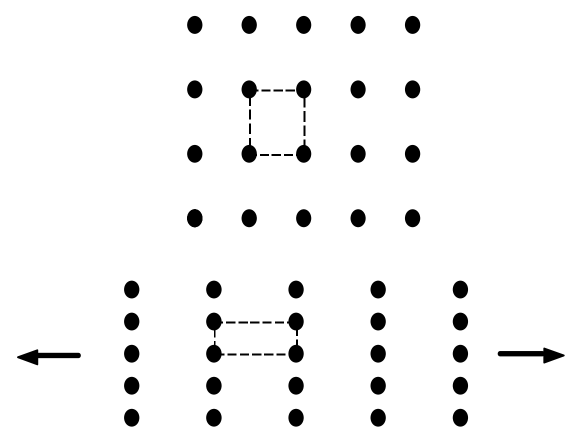
Bragg's law:

$$\lambda = 2d\sin\theta$$

Differences in lattice spacing

$\Rightarrow$ Elastic lattice strain

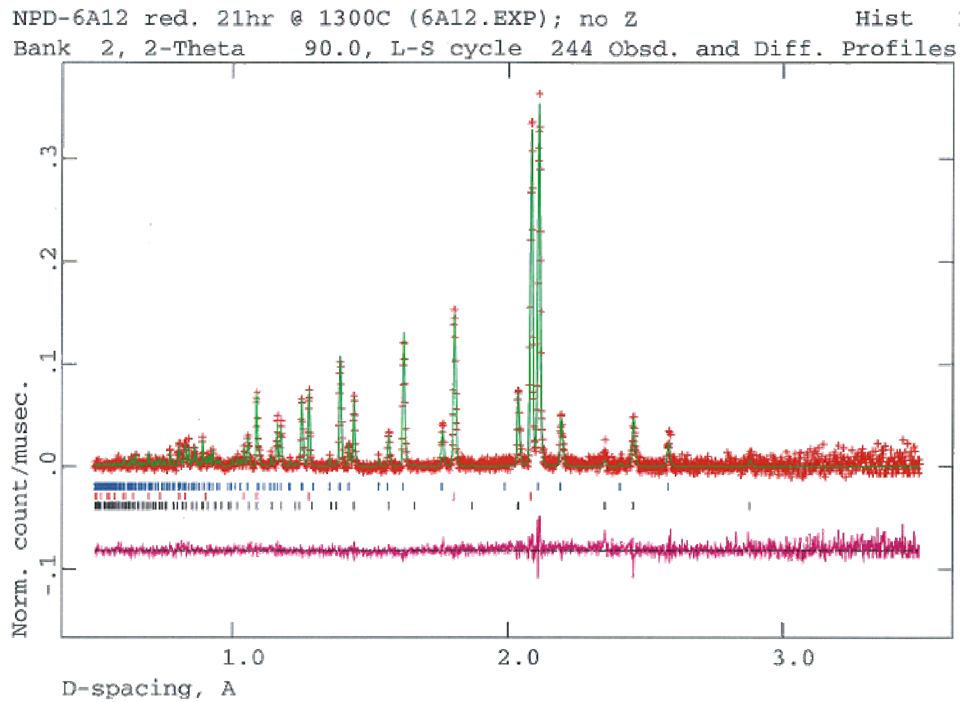
$$\varepsilon_{hkl}^{el} = \frac{d_{hkl} - d_{hkl}^{0}}{d_{hkl}^{0}} = \frac{d_{hkl}}{d_{hkl}^{0}} - 1$$



Neutron Powder Diffraction: *Data Analysis*

Rietveld Method*

- **Least-squares-based fitting method.**
- **Requires *a priori* phase information.**
- **Fits the whole diffraction pattern.**
- **Phase discrete.**
- **Can distinguish superimposed reflections.**
- **Yields detailed crystallographic information: lattice constants, texture, site occupancies, phase fractions, thermal parameters, etc.**



Fitting Parameter

"Weighted Pattern" Residual:

$$R_{wp} = \left[\frac{\sum_i w_i (I_{io} - I_{ic})^2}{\sum_i w_i (I_{io})^2} \right]^{1/2}$$

* **H. M. Rietveld, *Acta Cryst*., 22, 151 (1967); *J. Appl. Cryst*., 2, 65 (1969).**

SMARTS

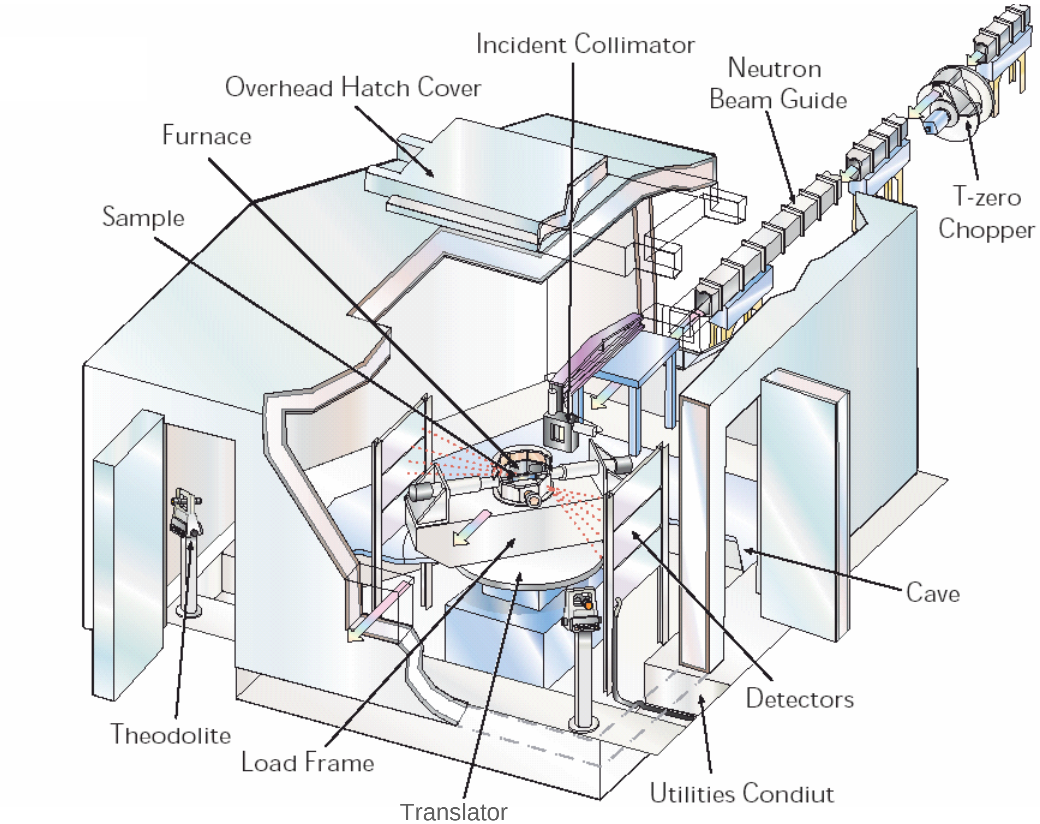
Spectrometer (for) MAterials Research (at) Temperature (&) Stress

- Third generation neutron powder diffractometer.
- 10-30 fold performance improvement over NPD.
- First dedicated engineering diffractometer.
- Optimized for engineering stress/strain studies.
- State-of-the-art ancillary equipment:
  - 250 kN (60,000 lb) load frame.
  - Controlled atmosphere furnace (T_{max}=1500°C under load, 2000°C stand alone).
- Radial collimators for 1 mm^3 spatial resolution.
- Rapid and accurate specimen handling capability.
- "Expert System" for experiment design and real time monitoring.

LANSCE Los Alamos Neutron Science Center

California Institute of Technology 1891

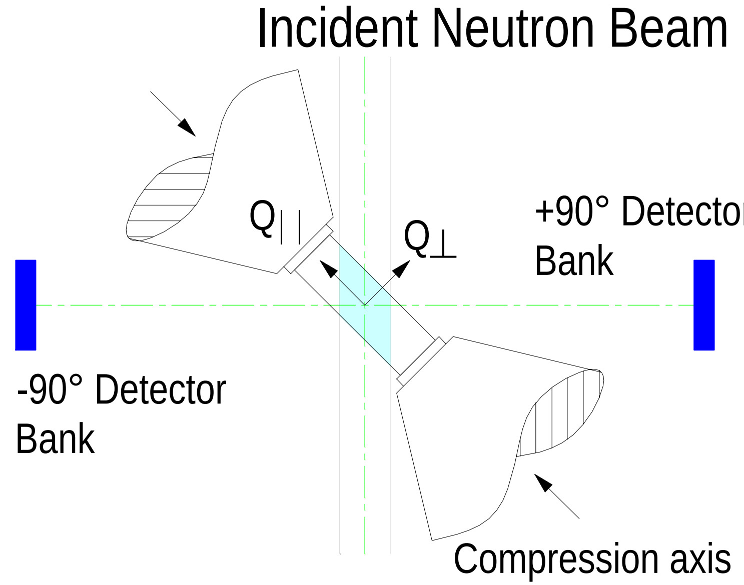
SMARTS: Spectrometer (for) MAterials Research (at) Temperature (and) Stress



Cave cutaway schematic

Cave with load frame & furnace installed on translator

SMARTS Load Frame

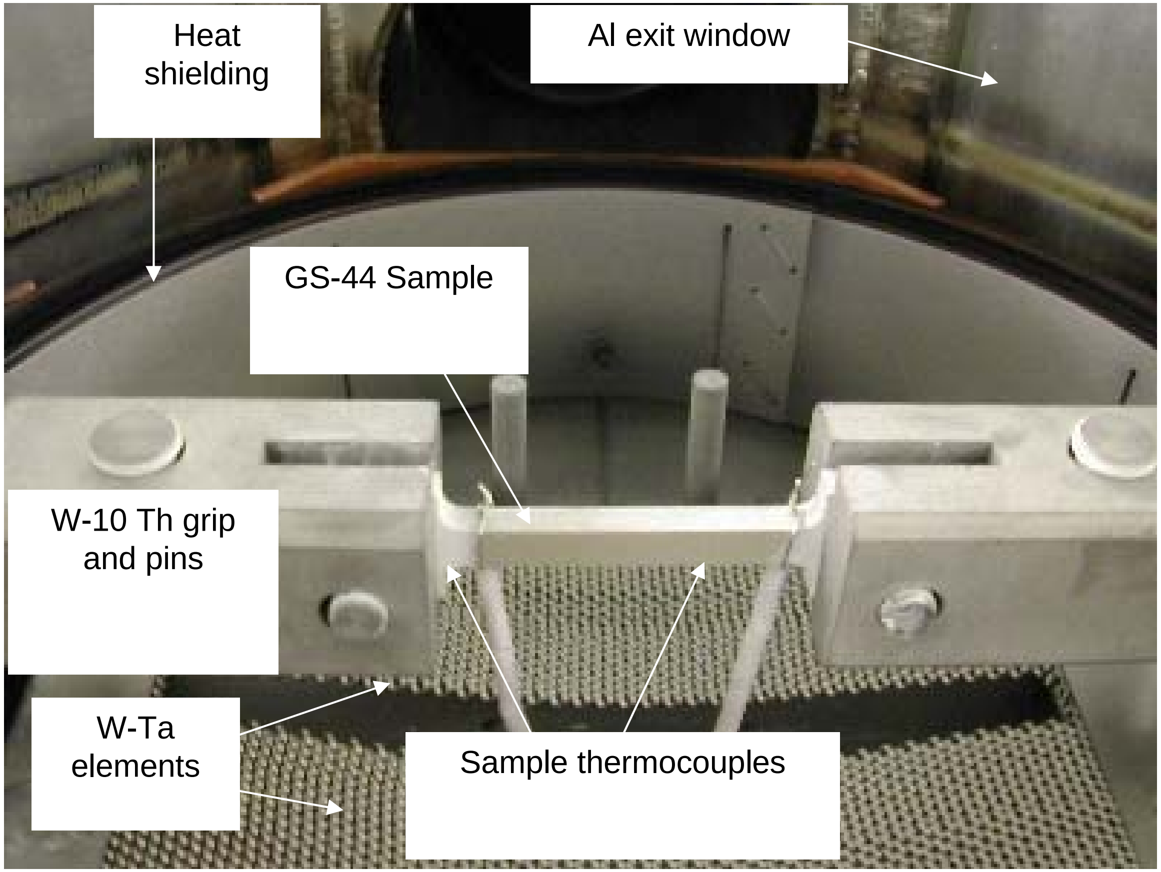


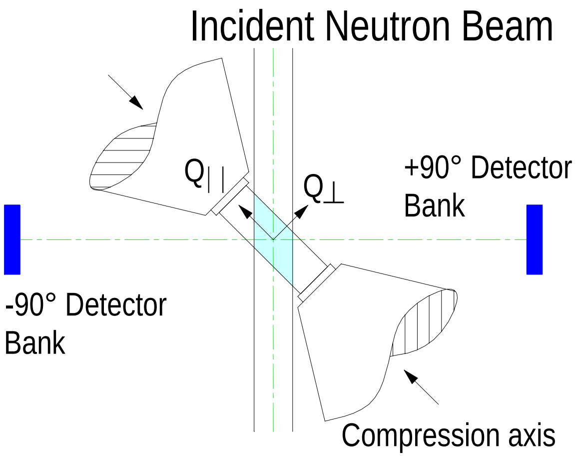
Bragg's law: $\lambda = 2d\sin\theta$

- Spectrometer for MAterials Research at Temperature and Stress (SMARTS)
- Schematic setup for *in-situ* compression loading
- Measurement time is about 10-20 minutes per load level
- Measure elastic strains in two directions simultaneously
- Bulk measurement contrary to conventional X-ray measurements

LANSCE Los Alamos Neutron Science Center

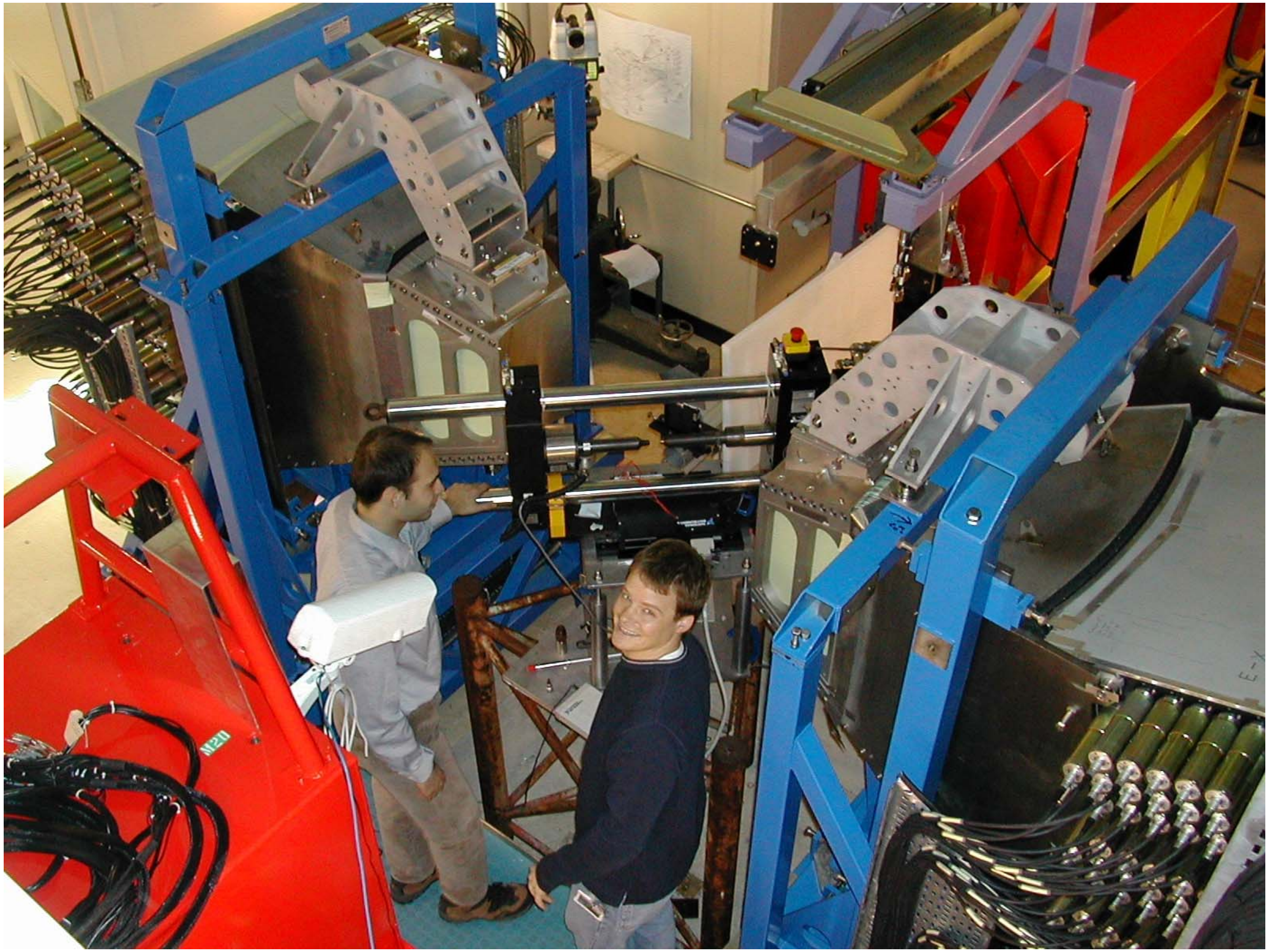
SMARTS Furnace





- **T_{max} = 1500°C under load**
- **Tmax = 2000°C stand alone**
- **Vacuum or inert atmosphere**

ENGIN-X Diffractometer

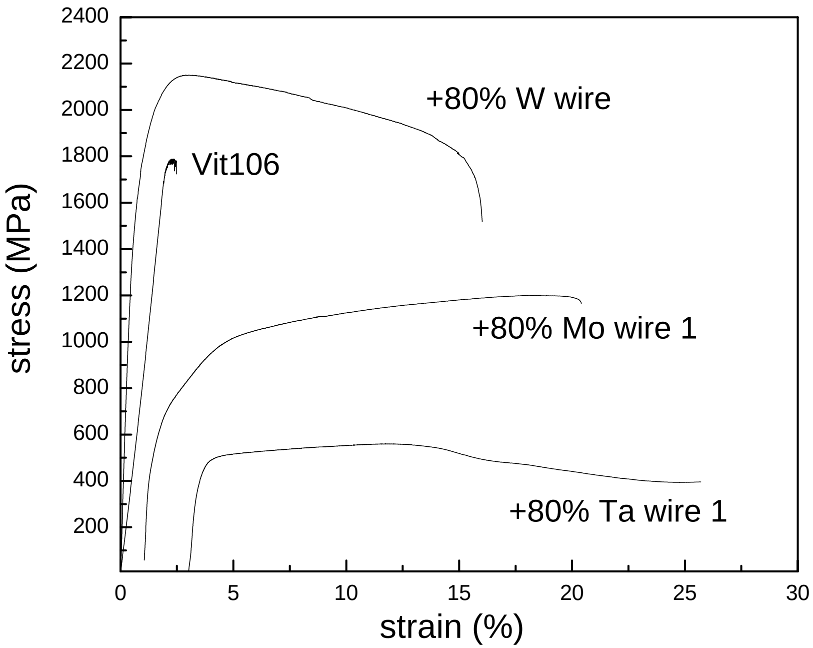


ISIS Neutron Scattering Facility
Rutherford Appleton Laboratory (Didcot, UK)

Bulk Metallic Glass Matrix Composites

- Main problem with BMGs: **catastrophic failure** under unconstrained loading.
- Main **deformation mechanism** is via **shear bands** (at room T).
- Addition of reinforcements has been shown to increase damage tolerance and toughness.
- Critical questions:
  - » What is the *in-situ* mechanical behavior of reinforcements?
  - » How do reinforcements interact with shear bands?

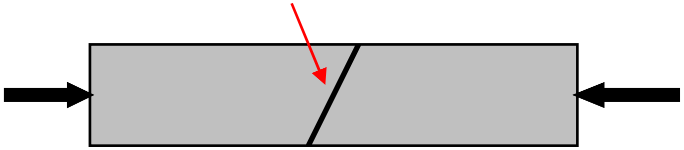
Compressive loading of fiber-reinforced BMG composites

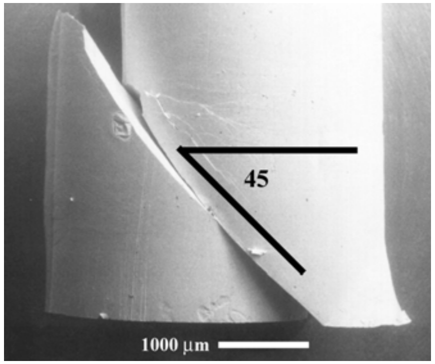


Shear band

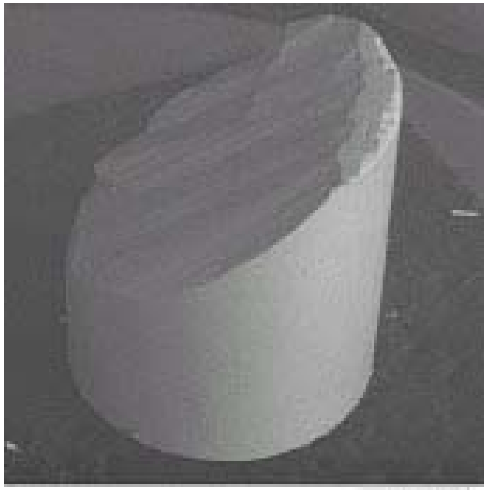
Monolithic

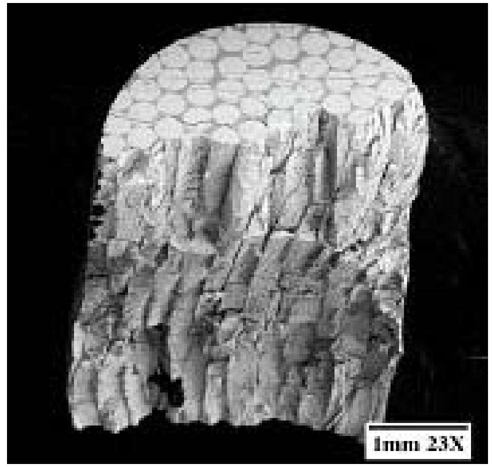
BMG

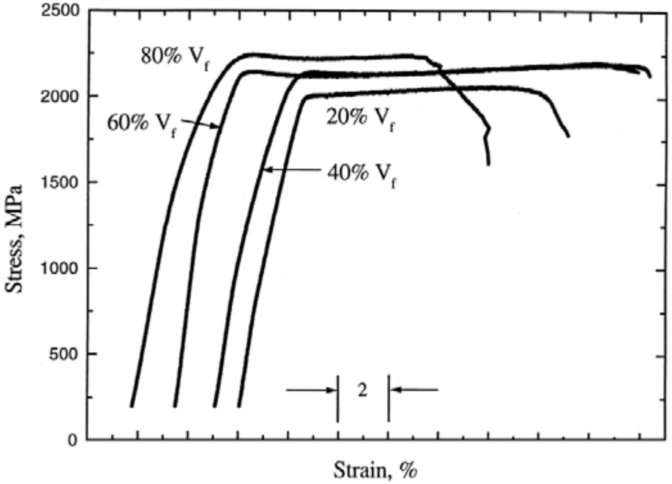




W-Fiber / BMG-Matrix Composites:
Compressive Loading Behavior





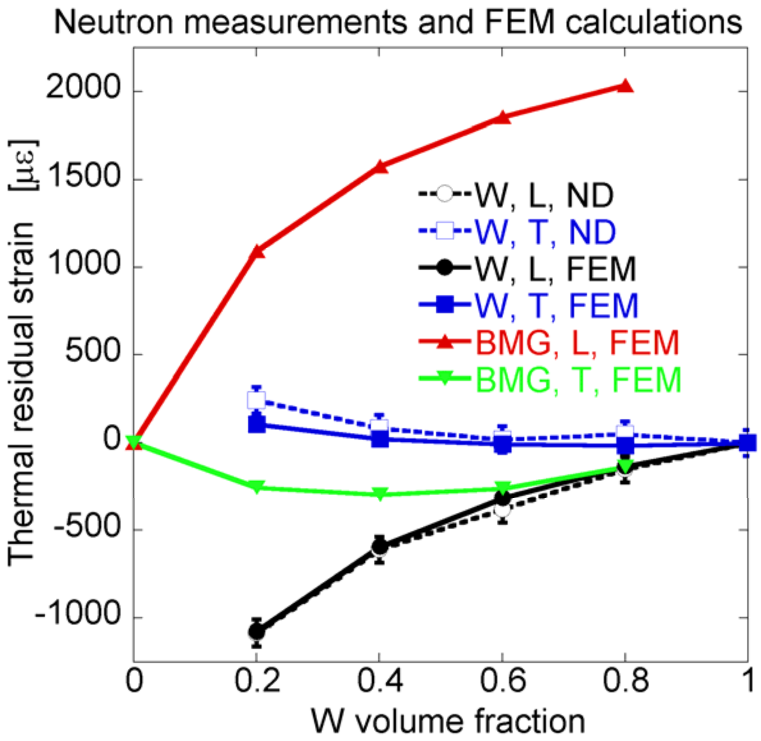


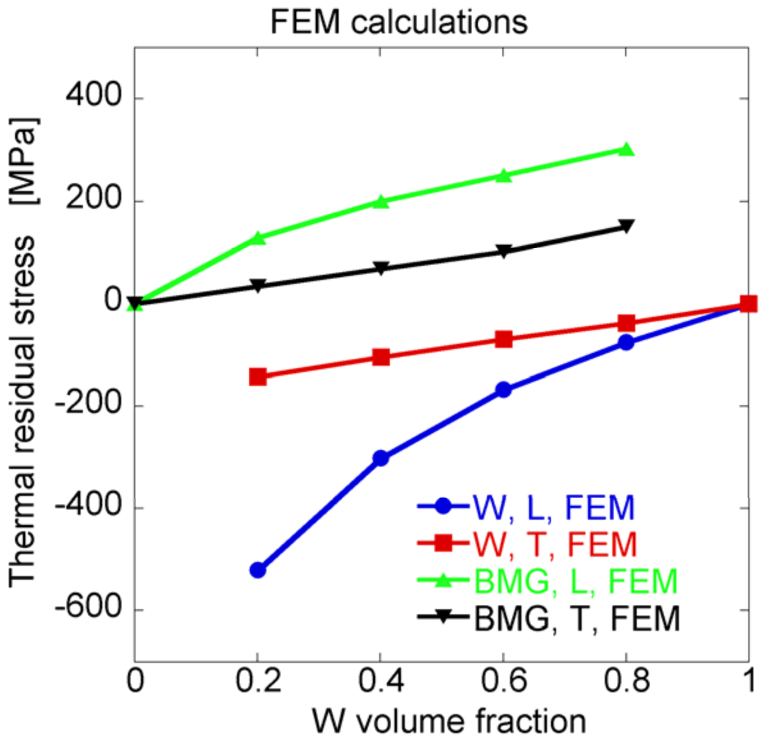
- Vitreloy 1 matrix: $Zr_{41.2}$ $Ti_{13.8}$ $Cu_{12.5}$ Ni_{10} $Be_{22.5}$
- Tungsten fiber composites:
  - Same ultimate stress as monolithic Vit.1
  - Large increase in ductility
  - Knee in stress strain curve as tungsten fibers yield

- ***In-situ deformation of W?***
- ***What happens to BMG?***

R. D. Conner, R. B. Dandliker and W. L. Johnson, Acta Mater., vol. 46(17), pp. 6089-6102, 1998

W-Fiber / BMG-Matrix Composites:
Thermal Residual Stresses





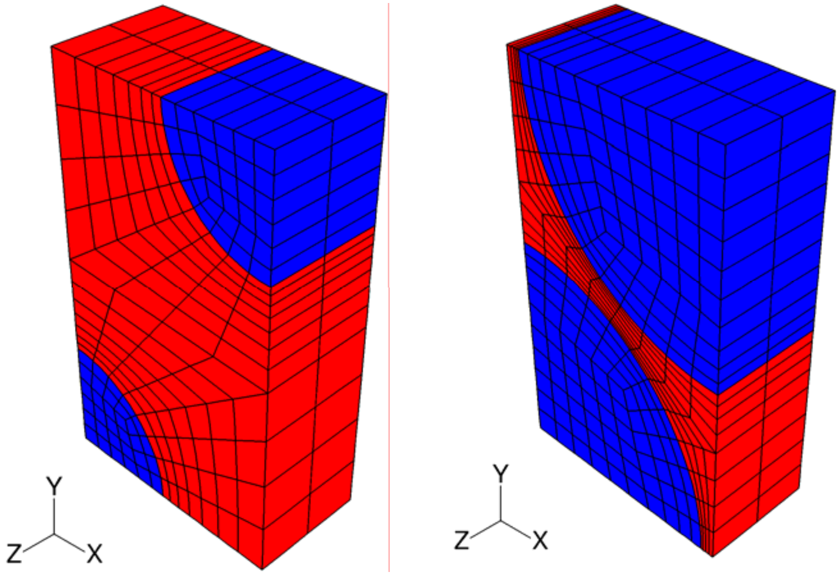
- **CTE mismatch: α_W (4.5x10^{-6} 1/K) < α_{BMG} (10x10^{-6} 1/K)**
- **Measured residual strain in W fibers using neutron diffraction**
- **Calculated thermal residual stresses in both phases using FEM**
- **Residual stresses are generated just below T_g**

D. Dragoi, E. Üstündag, B. Clausen and M. A. M. Bourke, Scripta Mater., vol. 45, pp. 245-252, 2001

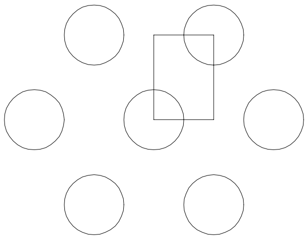
W-Fiber / BMG-Matrix Composites:
Finite Element Model

- Full 3-D model due to loading along fibers
  - Unit cell model
  - Plane strain along *z*
- Hexagonal stacking in all models to accommodate high volume fractions
- Thermal residual stresses: no relaxation below T_g*
- Constitutive laws:
  - W: deduced *in-situ* behavior
  - BMG: von Mises or Mohr-Coulomb**

$$\tau_c = 946 - 0.04\sigma_n \quad \text{[MPa]}$$



20% W/BMG   80% W/BMG

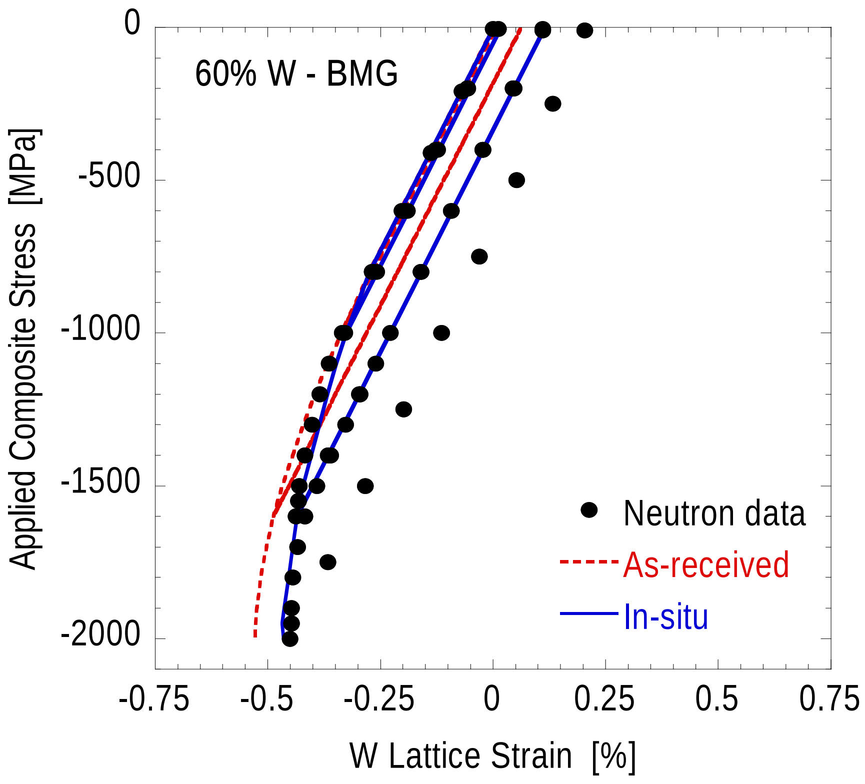


* D. Dragoi, E. Ustundag, B. Clausen and M.A.M. Bourke, *Scripta Mater.*, 45 (2), 245-252 (2001).

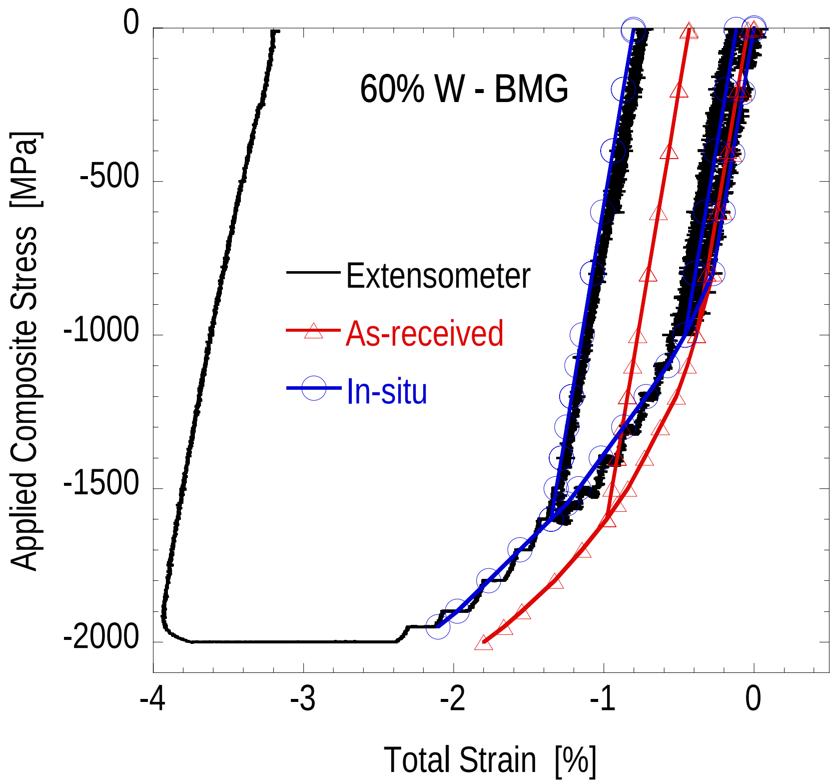
** J.J. Lewandowski *et al.*, in print: *Phil. Mag. A* (2002).

W-Fiber / BMG-Matrix Composites:
Compressive Loading Behavior

Neutron data vs. model



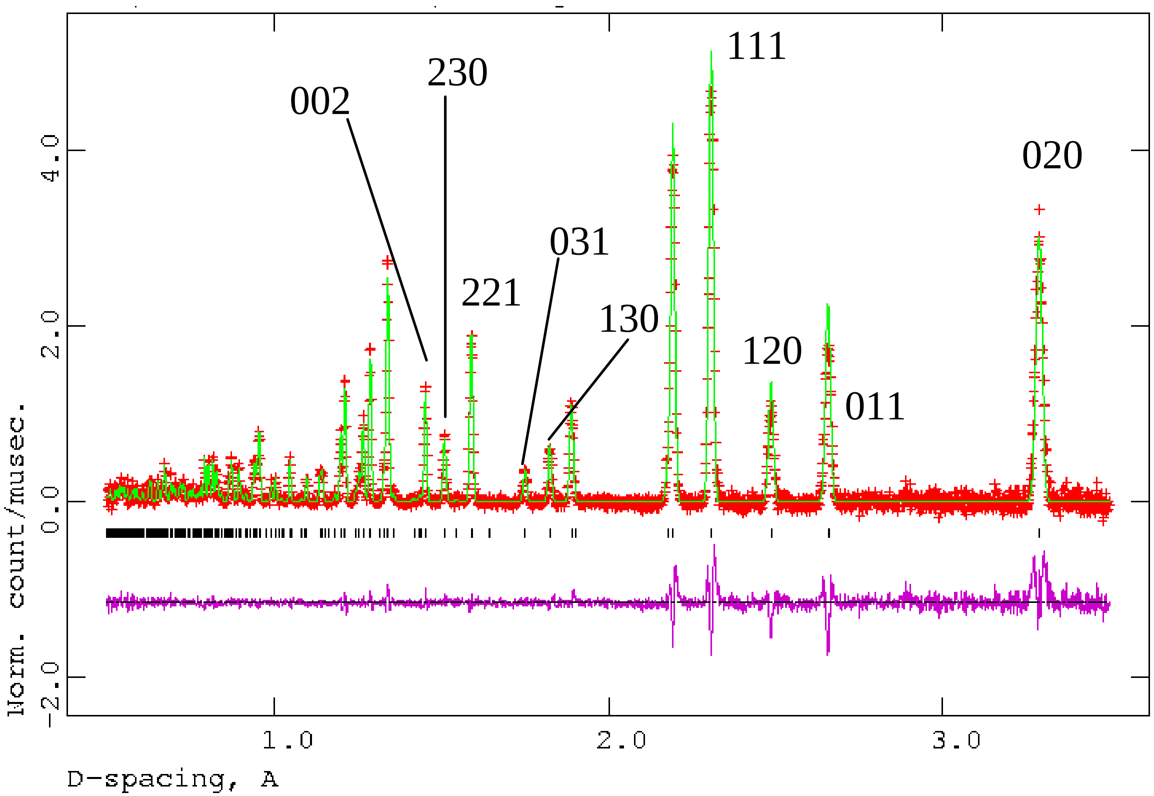
Macro data vs. model



- **Important to account for *in-situ* deformation and residual stresses**
- **W yields at -1300 MPa, BMG yields at -1900 MPa**
- **Composite yielding at -360 MPa (20% W-BMG), -1060 MPa (80% W-BMG)**
- **Model struggles at high stresses (multiple shear bands in BMG?)**

B. Clausen, S.Y. Lee, E. Ustundag, C.C. Aydiner, R.D. Conner, M.A.M. Bourke, **49**, 123-128, *Scripta Mater.* (2003)

β-Si_3N_4: Neutron Diffraction Experiments



- Single phase sample (β-Si_3N_4) – AS800 or GS44 from Honeywell
- Multiple reflections used in elastic constant and CTE determination
- Si_3N_4 fitting parameters:
  - Space group $P6_3/m$ – hexagonal; a = 7.608Å, c = 2.911Å
  - 6-term background function, absorption, Debye-Waller (thermal) parameter

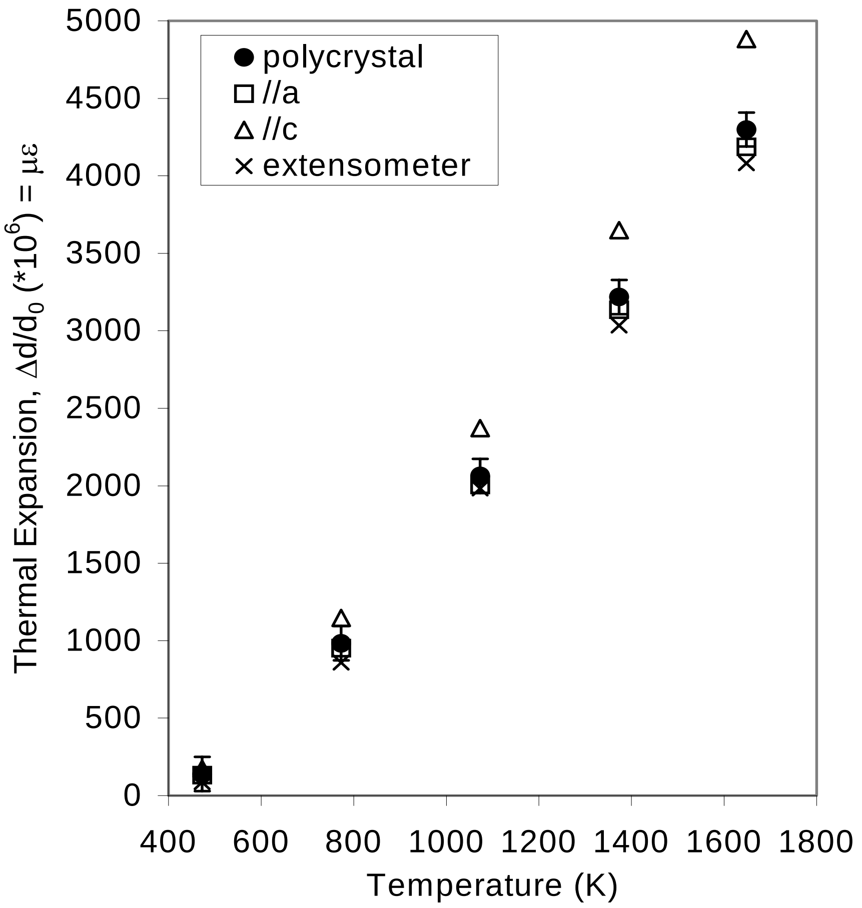
β-Si_3N_4: Coefficient of Thermal Expansion

- Diffraction data used directly in CTE calculation*.
- Multiple reflections employed; higher precision.
- Least squares analysis of redundant data.
- Result for CTE tensor of AS800 β-Si_3N_4:

$$\alpha_{ij} = \begin{bmatrix} 3.50 & 0 & 0 \\ 0 & 3.50 & 0 \\ 0 & 0 & 4.06 \end{bmatrix} (x10^{-6}\, 1/K)$$

- Polycrystalline value:

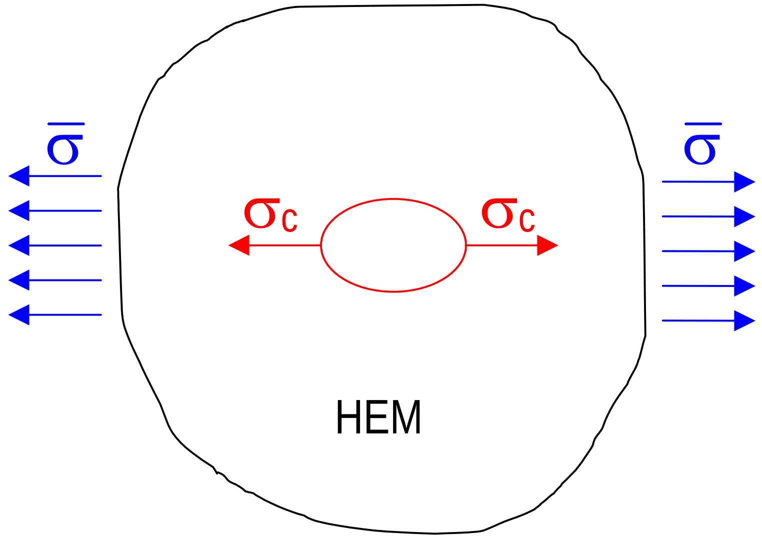
  α = 3.69 (x10^{-6} 1/K)



* S.M. Jessen and H. Kuppers, *J. Appl. Cryst.*, 24, 239-242 (1991).

Self-Consistent Model (SCM)

- **Model Assumptions:**
  - **Eshelby inclusion theory**
  - **Stresses and strains within an ellipsoidal inclusion are uniform**
  - **Homogeneous equivalent medium (HEM)**



- **Output:**
  - **Direct comparison with neutron diffraction measurements**
  - **Averages over grain sets representing reflections**
  - **Information about material behavior on a microscopic scale**
  - ***hkl* dependent behavior**
  - **Accurate description of texture**

Elastic Constants of AS800 β-Si_3N_4 at 1375°C*

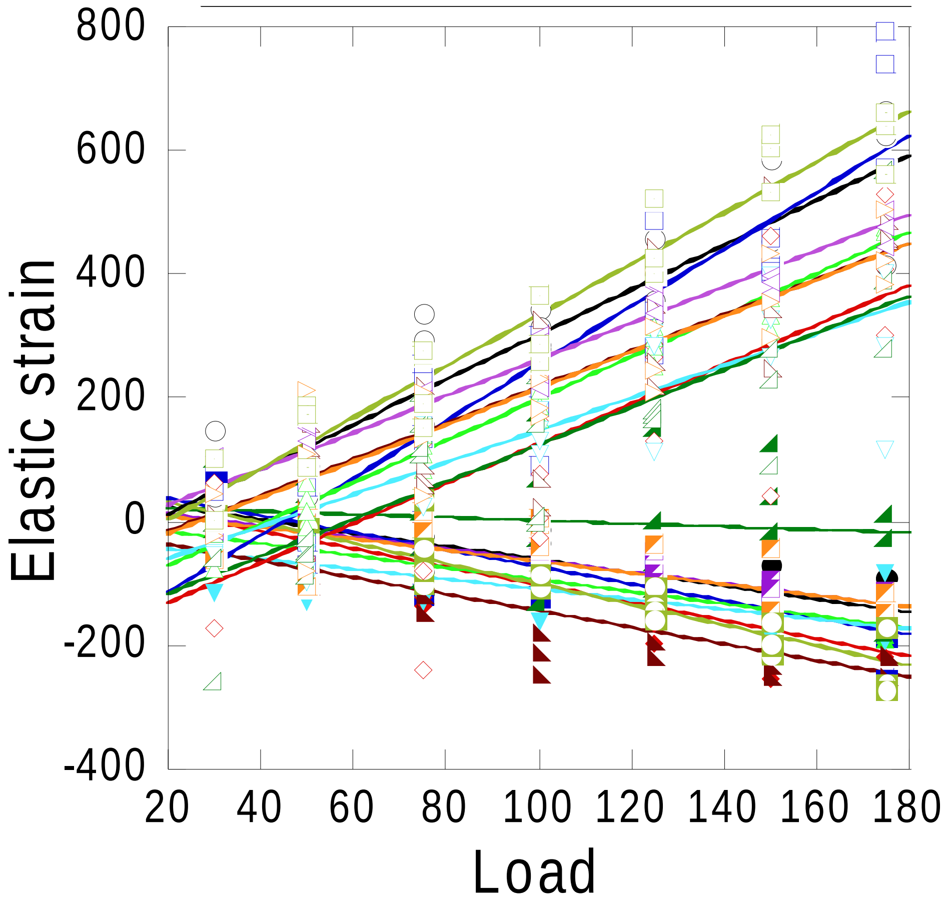
- Employed self-consistent modeling (EPSC)
- Least square fitting of *hkl*-dependent elastic strains in both longitudinal and transverse directions
- Polycrystalline average:

  E **= 310 GPa,** ν **= 0.31**

manufac. values at 1200°C:

E **= 293 GPa,** ν **= 0.28**

$$C_{ij} = \begin{bmatrix} 460 & 160 & 240 & 0 & 0 & 0 \\ 160 & 460 & 240 & 0 & 0 & 0 \\ 240 & 240 & 310 & 0 & 0 & 0 \\ 0 & 0 & 0 & 140 & 0 & 0 \\ 0 & 0 & 0 & 0 & 140 & 0 \\ 0 & 0 & 0 & 0 & 0 & 150 \end{bmatrix} \quad (GPa)$$



Literature values (at room T):

C_{11} = 430, C_{33} = 570, C_{12} = 190,

C_{13} = 130, C_{44} = 110 (GPa)

*** G.A. Swift, E. Üstündag, B. Clausen, M.A.M. Bourke and H.T. Lin, *Appl. Phys. Lett.* 82 [7], 1039-1042 (2003).**

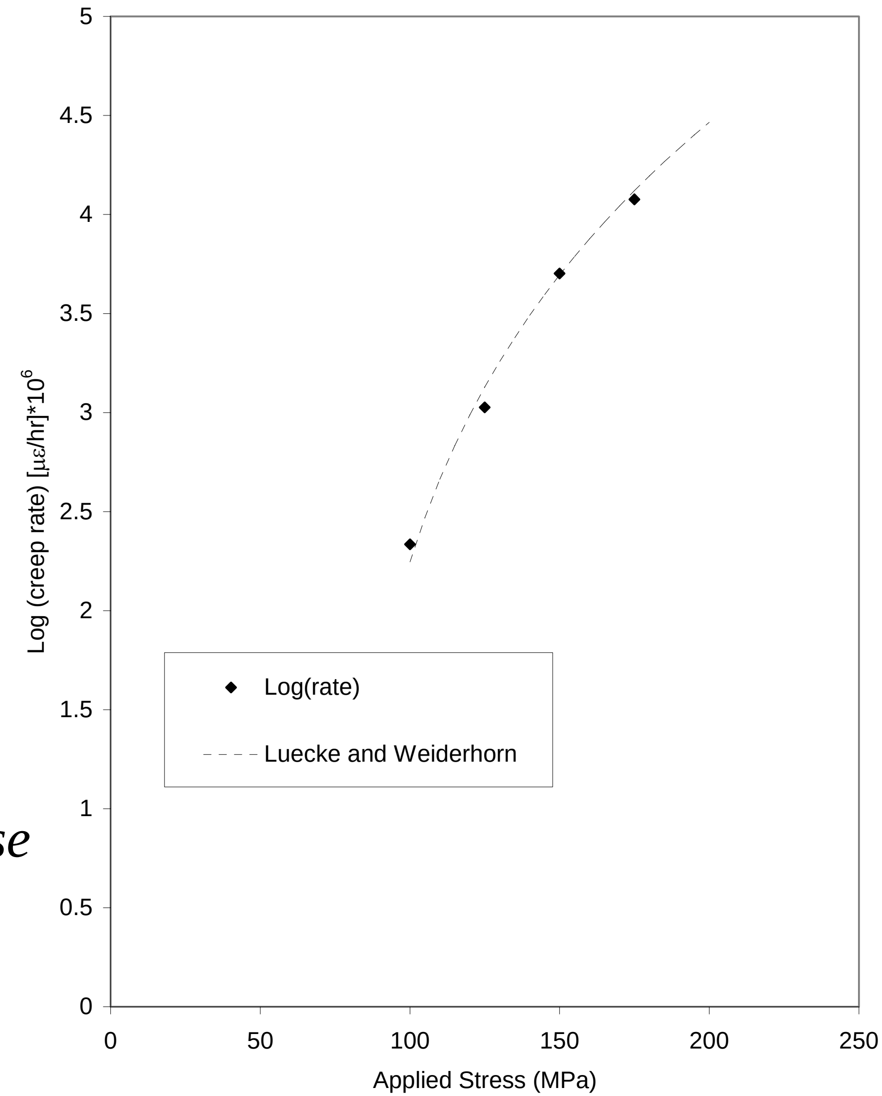
Creep Mechanisms of ISR Si_3N_4

- **Creep of ISR Si_3N_4 described by formation and growth of cavities in grain boundary phase**.**
- **Si_3N_4 grains remain elastic.**
- **Results in curvature of semi-log plot of creep rate vs. stress.**
- **Leads to the following creep equation:**

$$\dot{\varepsilon}_s = B\sigma \exp\left(\frac{-\Delta H}{RT}\right)\frac{f^3}{(1-f)^2}\exp(\alpha\sigma)$$

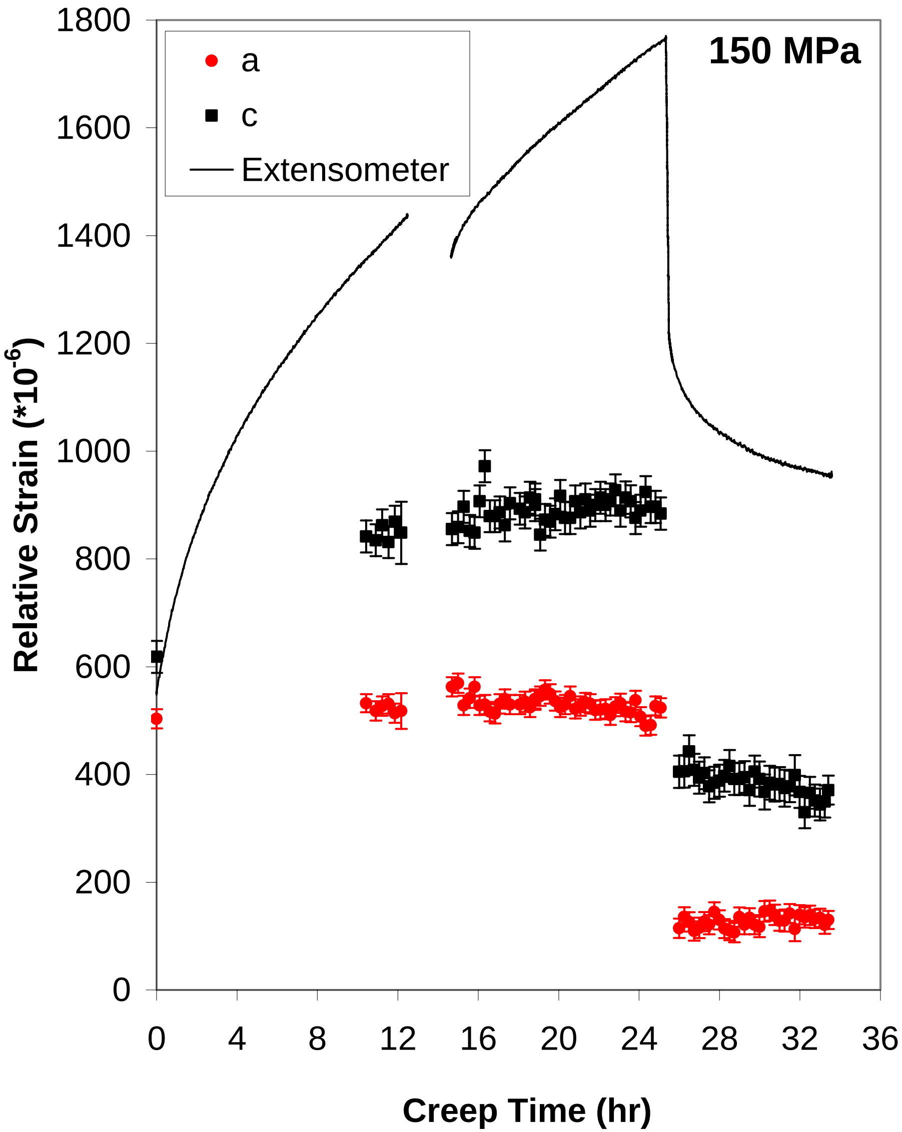
$$where\ \ \alpha \approx \frac{2\sigma_c}{9\sigma^2}\ \ and\ \ f : vol.\ frac. of\ g.b.\ phase$$

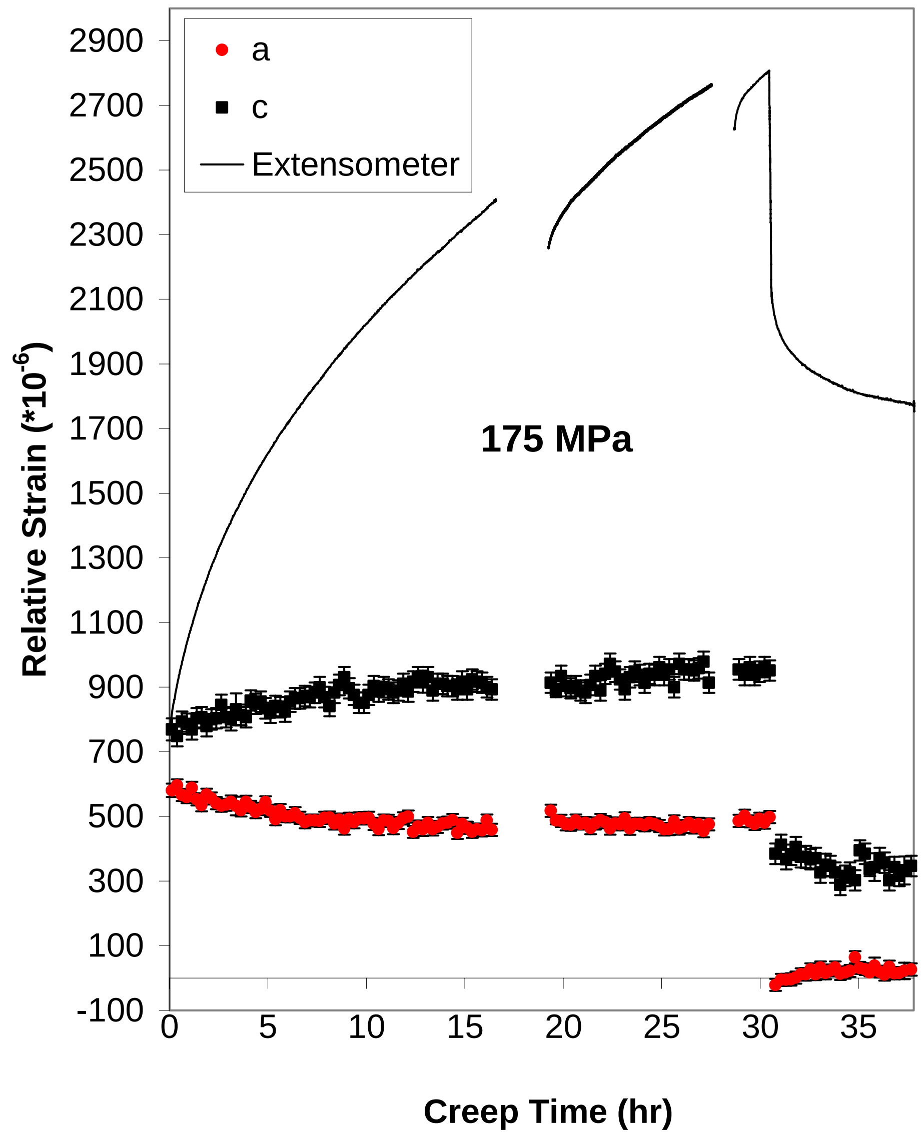
$$\dot{\varepsilon}_s = A'\sigma\exp(\alpha\sigma) \qquad A'(\sigma) = 0.08\cdot\left(\frac{\sigma-\sigma_c}{25}\right)$$



** W.E. Luecke and S.M. Wiederhorn, *J. Am. Ceram. Soc.* 82 (10), 2769-2778 (1999).

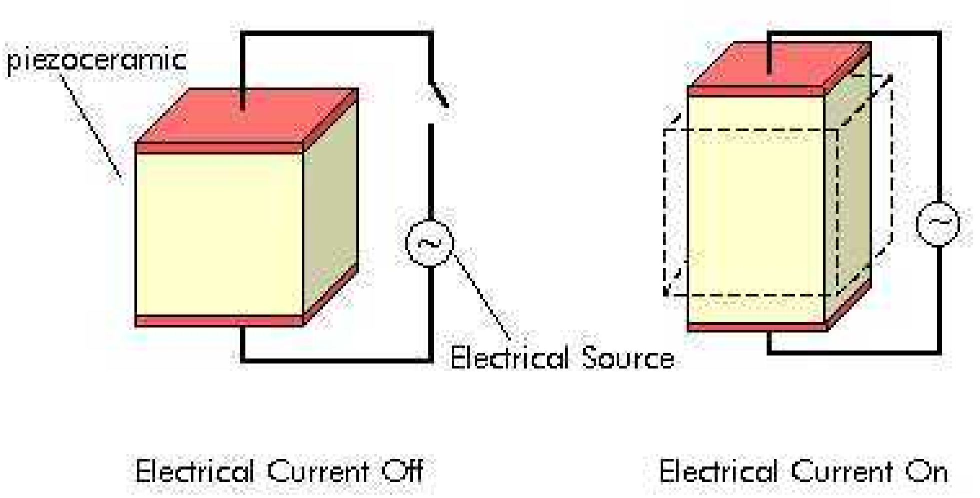
Creep of GS-44 at 1200°C: *Constant Stress Test*





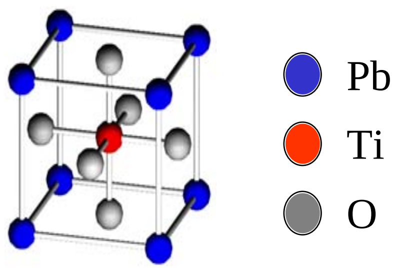
Constitutive Behavior of Ferroelectric Materials

- Ferroelectric and piezoelectric materials couple electrical signals to mechanical displacements.
- Ideal for applications in vibration control, sensors, transducers, and micromechanical devices.

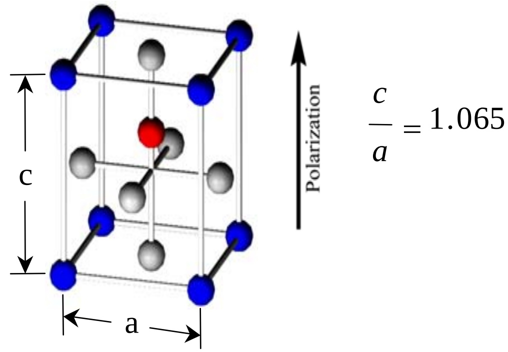


How Does Ferroelectricity Work?

$PbTiO_3$

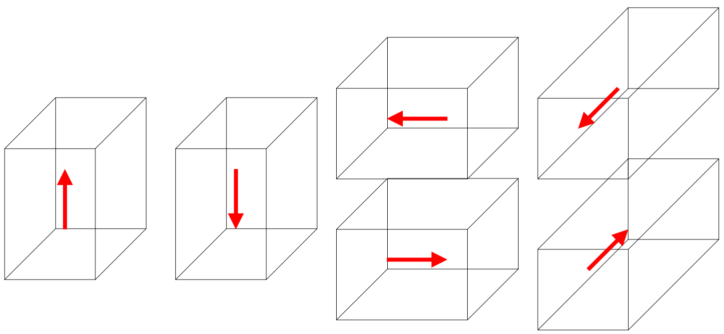


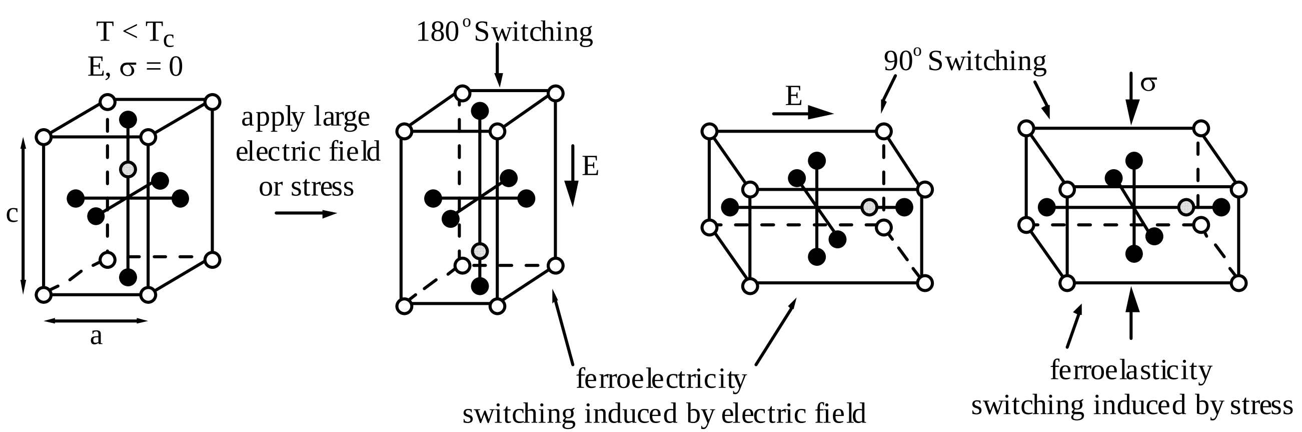
High temperature
(non-polar cubic)



Room temperature
(<001> polarized tetragonal)

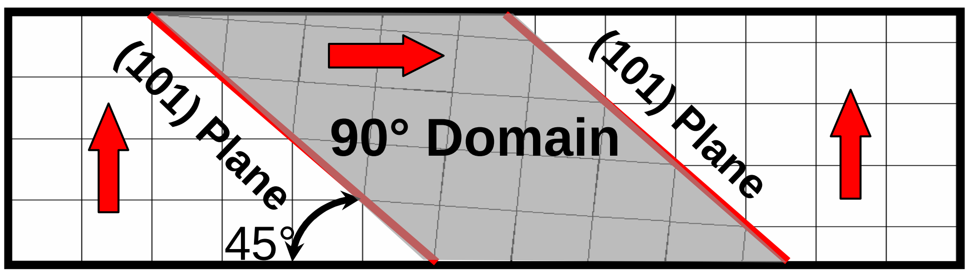
Six equivalent <001>$_{cubic}$ directions give six equivalent states at room temperature



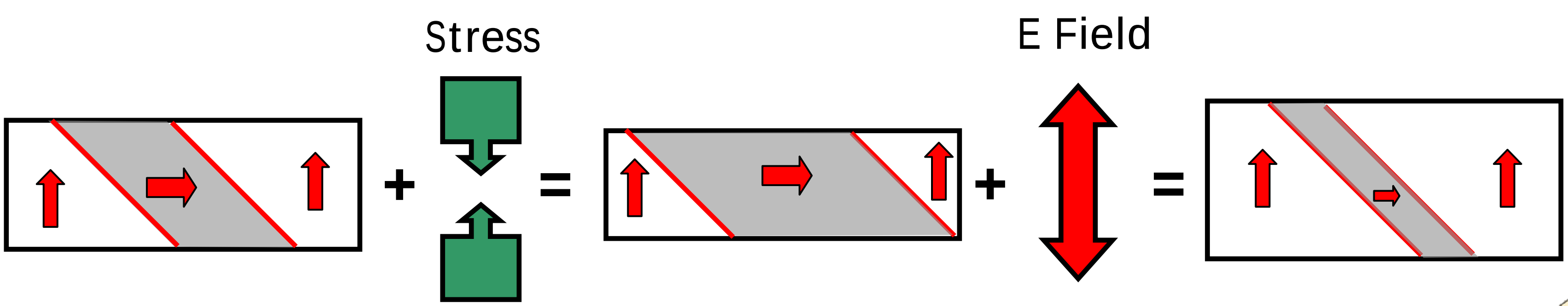


Microscopic Effects

- Regions of organized unit cell polarizations are separated by twin boundaries called domain walls

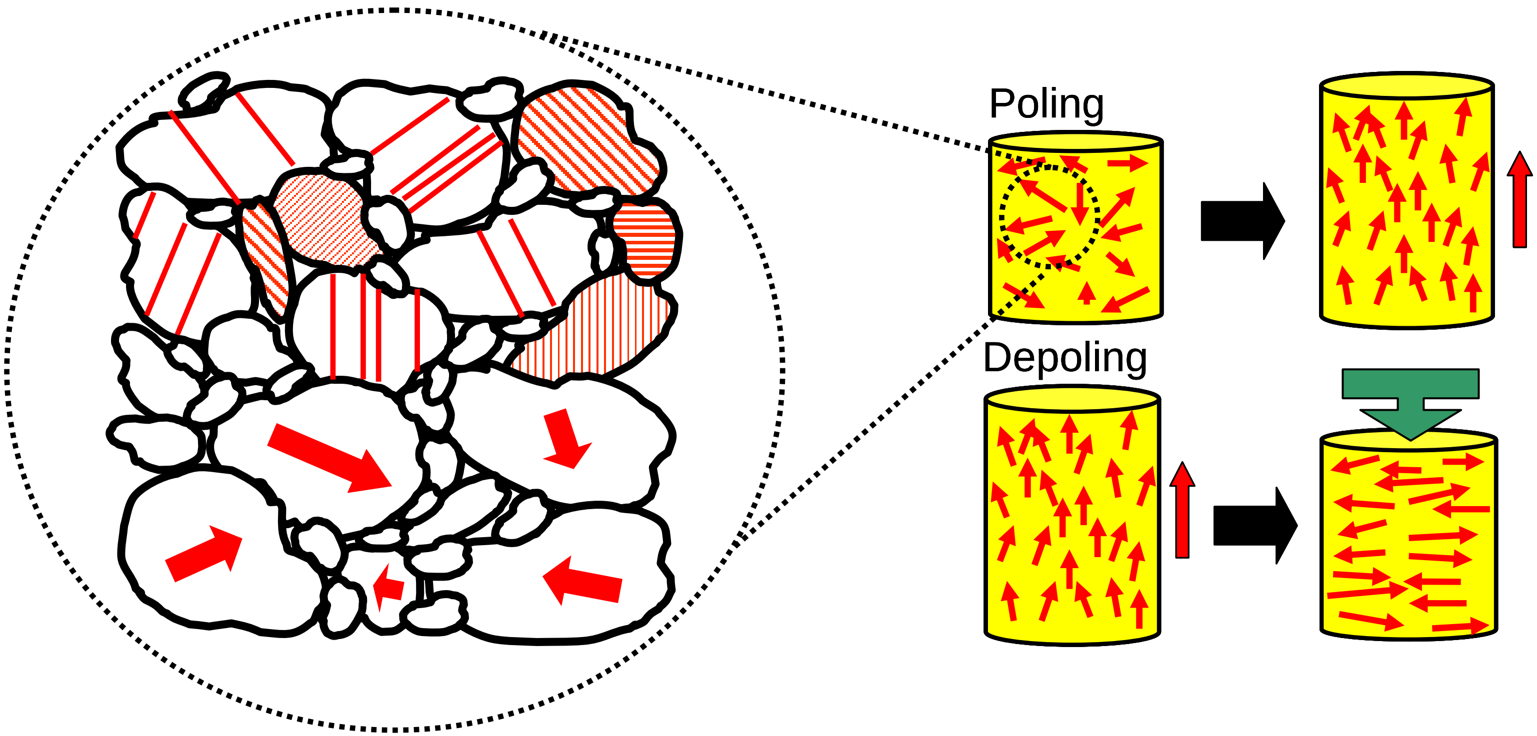


- Application of stress or electric field induces motion of domain walls, changing polarization and strain in the crystal

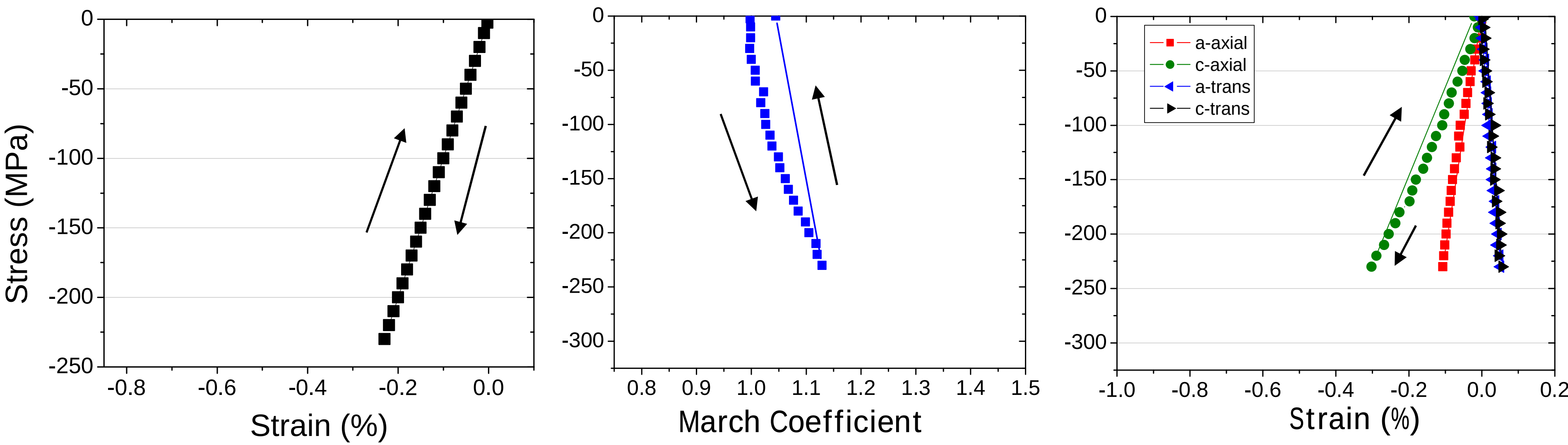


Meso-/Macroscopic Effects

- Grains within a polycrystal possess randomly oriented domains
- Electrical poling is used to align a significant number of domains and produce a technologically viable ceramic material
- Domain motion may be constrained by grain orientation and local boundary conditions



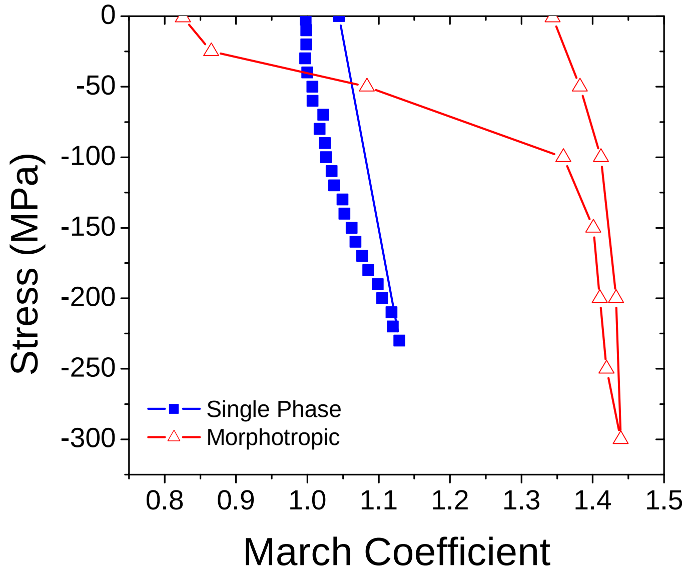
Compression of Single Phase *Tetragonal* PZT

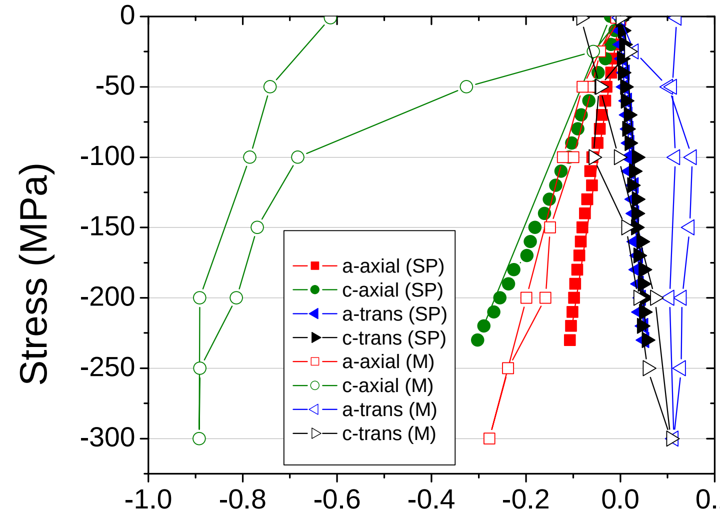


- Strain gauge data indicate linear elastic behavior
- March coefficient results suggest minor 90°domain switching
- Lattice strains are approximately linear

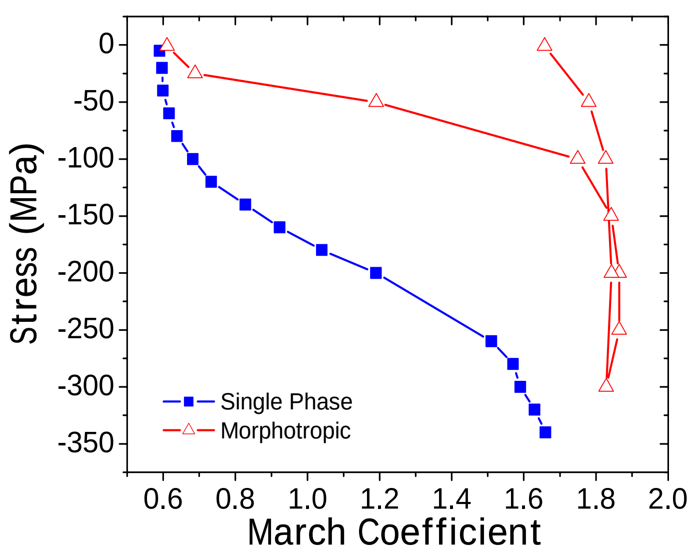
Comparison of Various PZTs

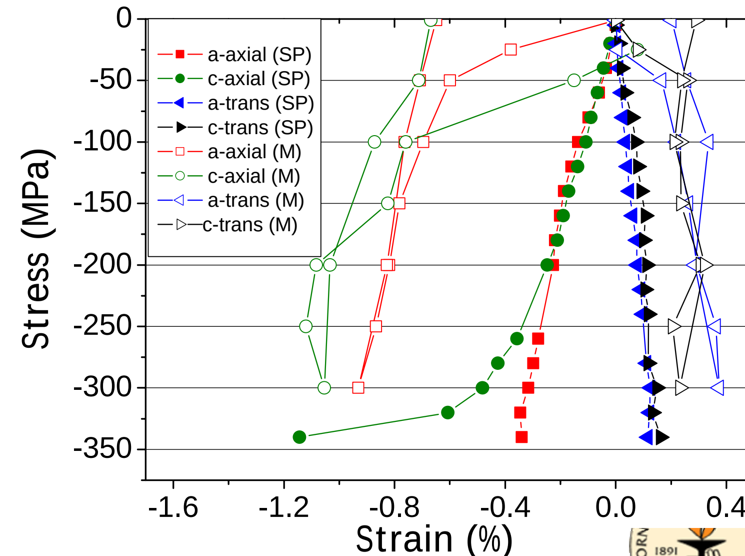
Tetragonal





Rhombohedral





R.C. Rogan, E. Üstündag, B. Clausen, M.R. Daymond, *J. Appl. Phys.* 93[7], 4104-4111 (2003).