Characterization of defects in ultrafine-grained interstitial-free steel prepared by severe plastic deformation

J. Čížek, M. Janeček, T. Krajňák, J. Stráská, P. Hruška

Faculty of Mathematics and Physics, Charles University Prague, Czech Republic

J. Gubicza

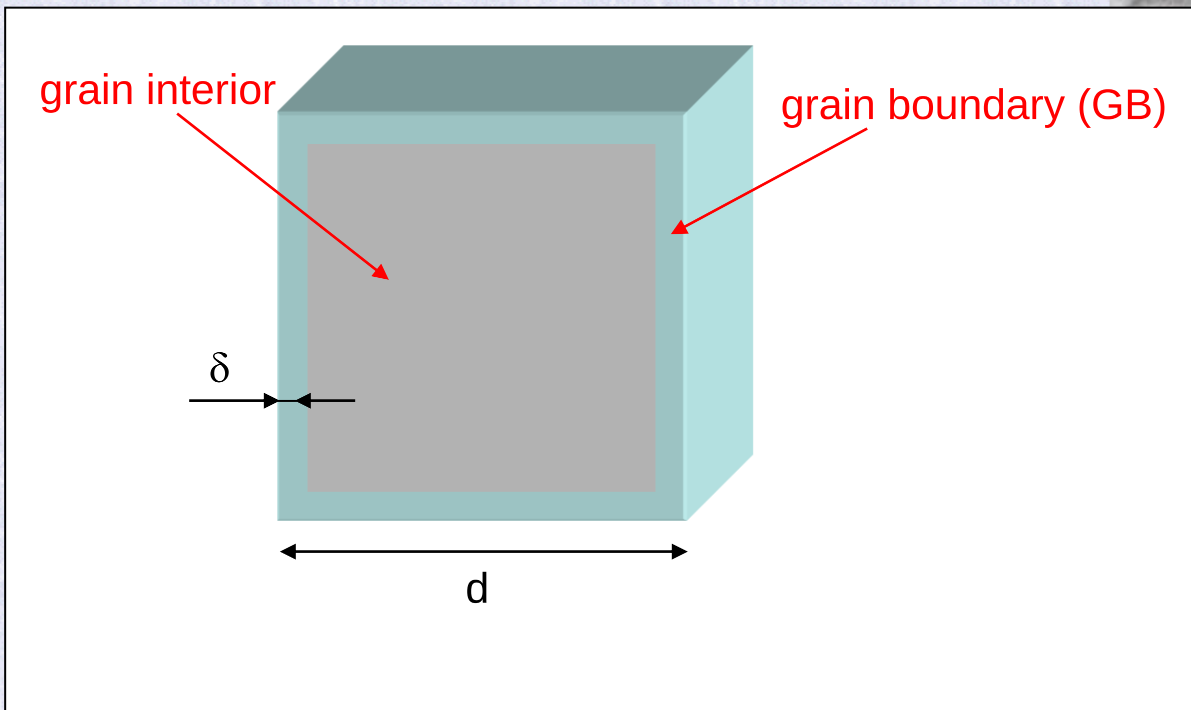
Department of Materials Physics, Eötvös Loránd University, Budapest, Hungary

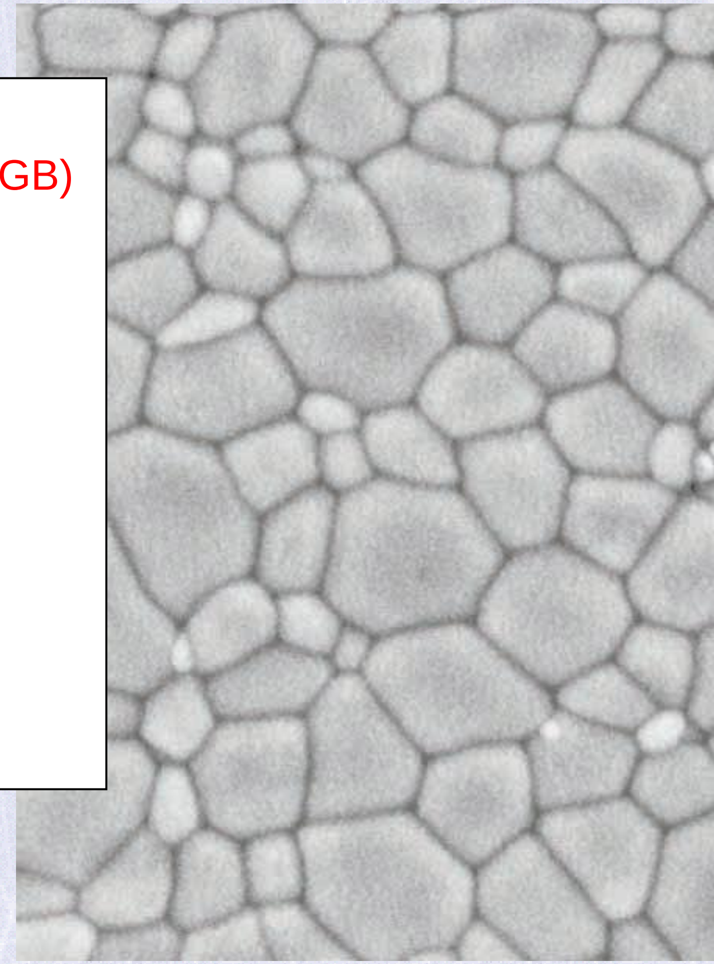
H.S. Kim

Department of Materials Science and Engineering, POSTECH, Pohang, South Korea

Ultra fine grained (UFG) materials

polycrystalline material

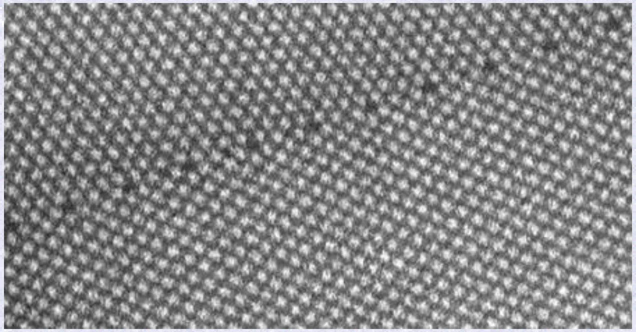




volume fraction of GB's

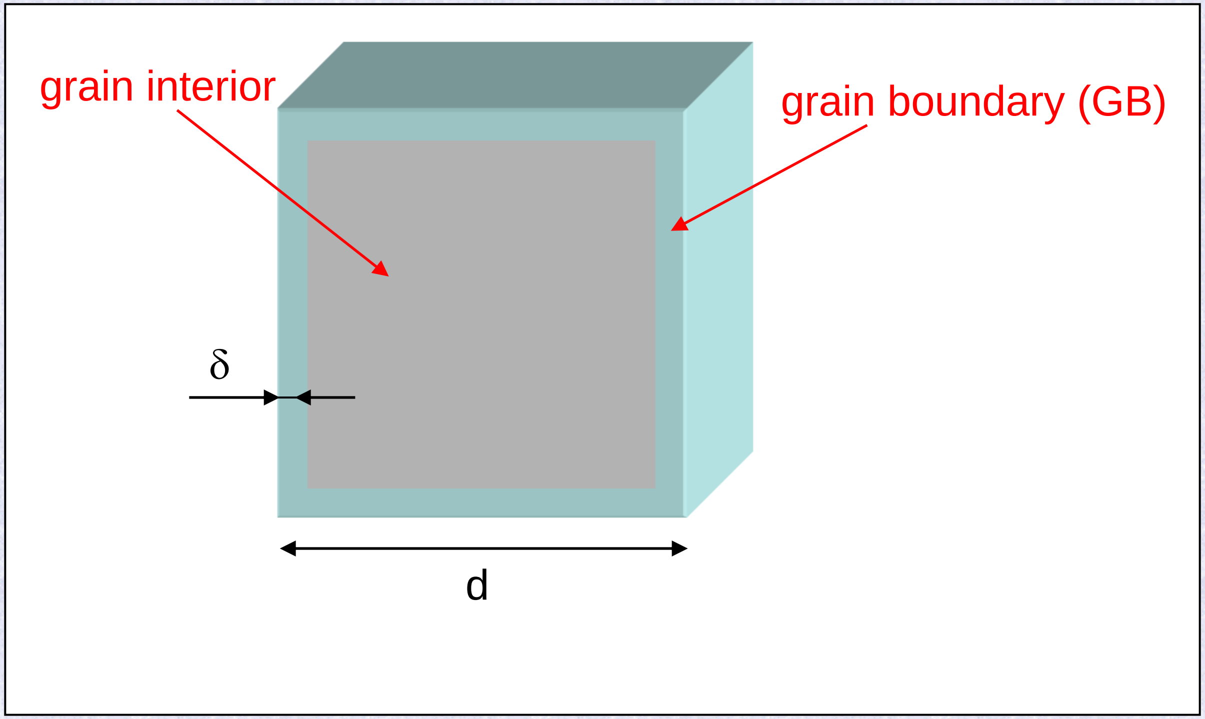
$$f_{GB} = 1 - \left(\frac{d - \delta}{d} \right)^3$$

Ultra fine grained (UFG) materials



crystallographic width of GB's: $\delta \approx 1$ nm

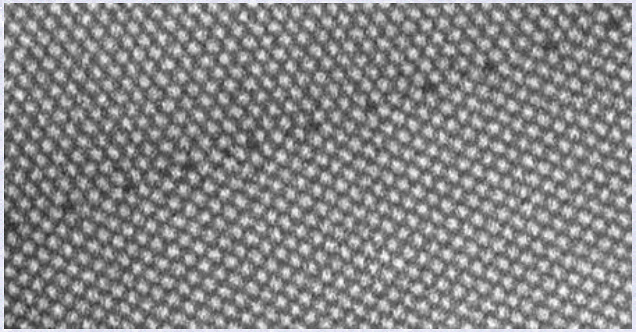
polycrystalline material



volume fraction of GB's

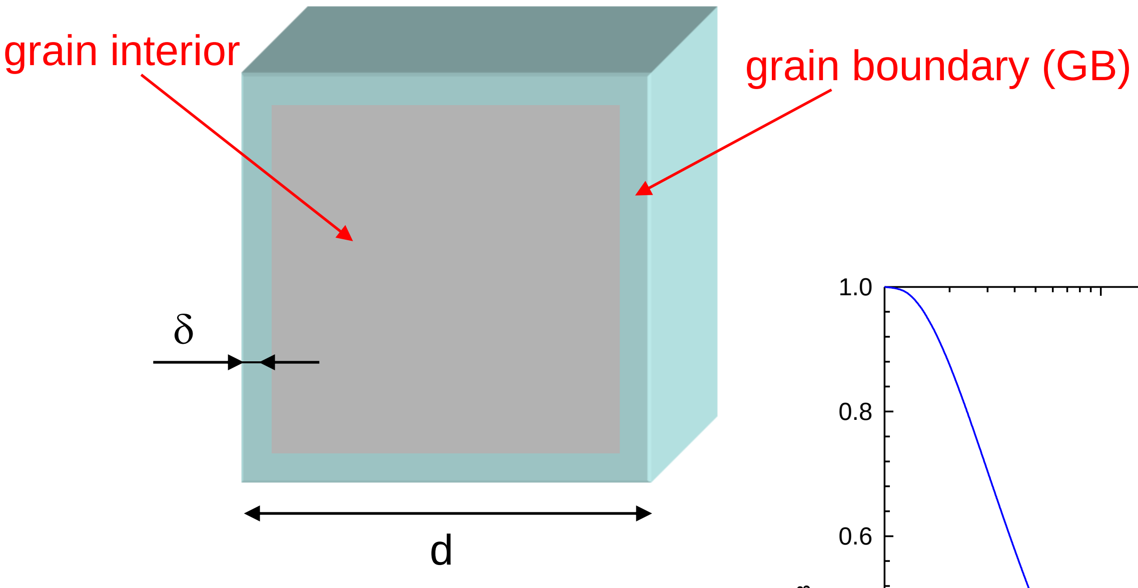
$$f_{GB} = 1 - \left(\frac{d-\delta}{d} \right)^3$$

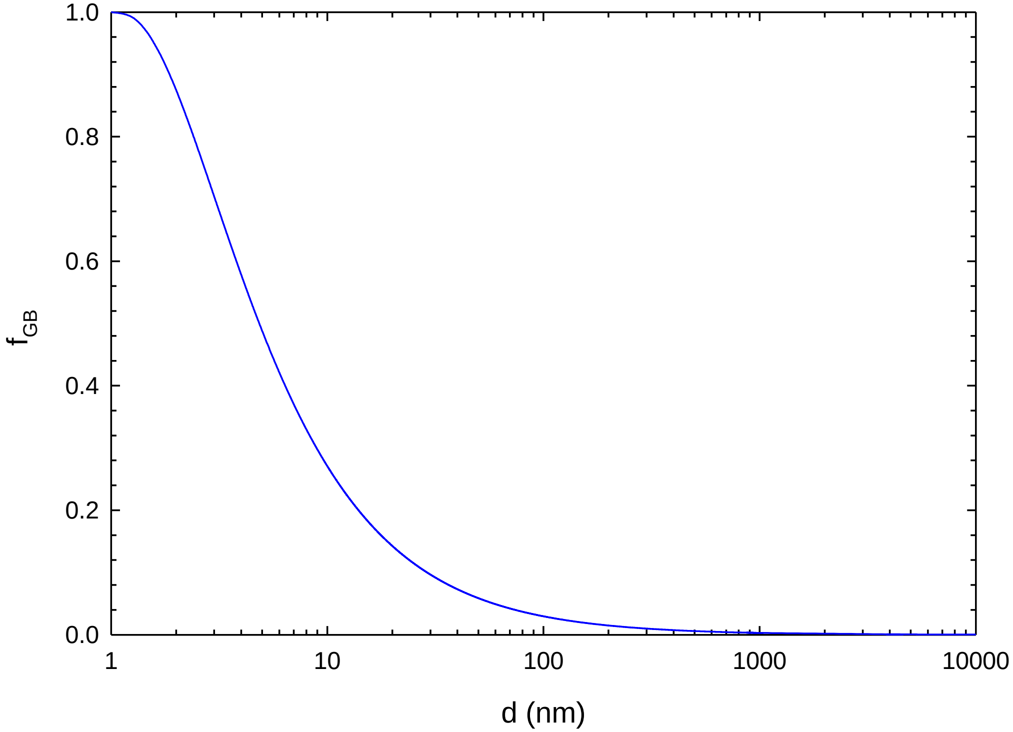
Ultra fine grained (UFG) materials



crystallographic width of GB's: $\delta \approx 1$ nm

polycrystalline material



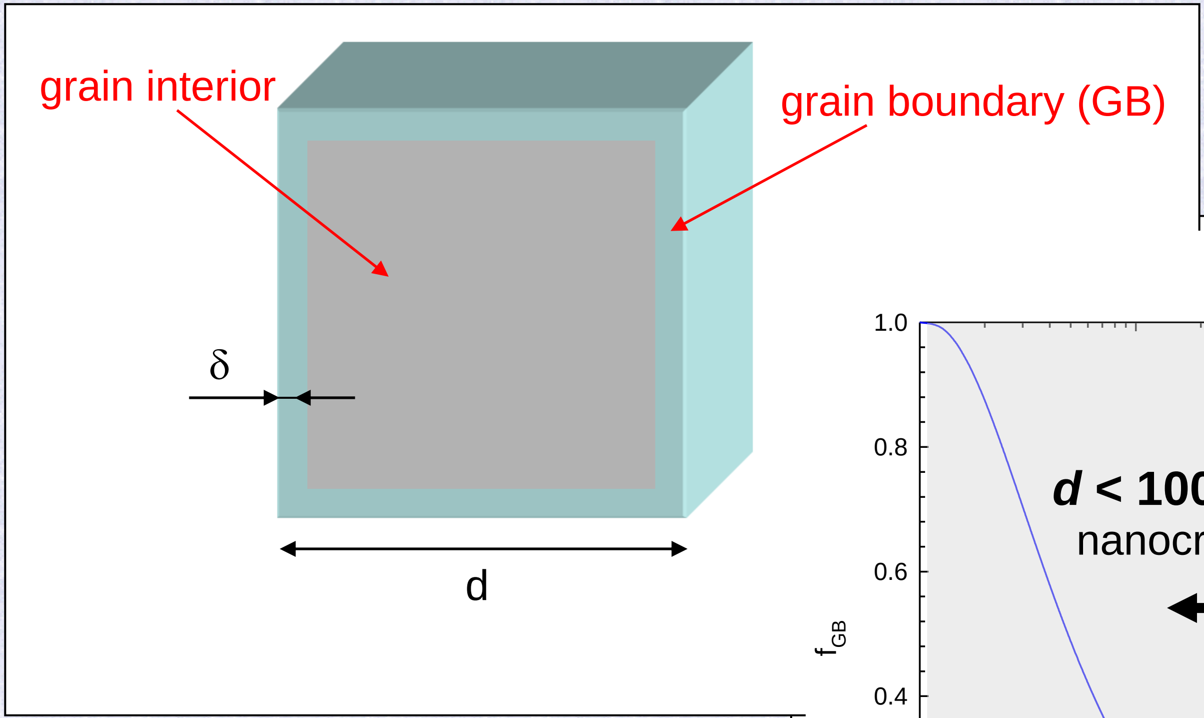


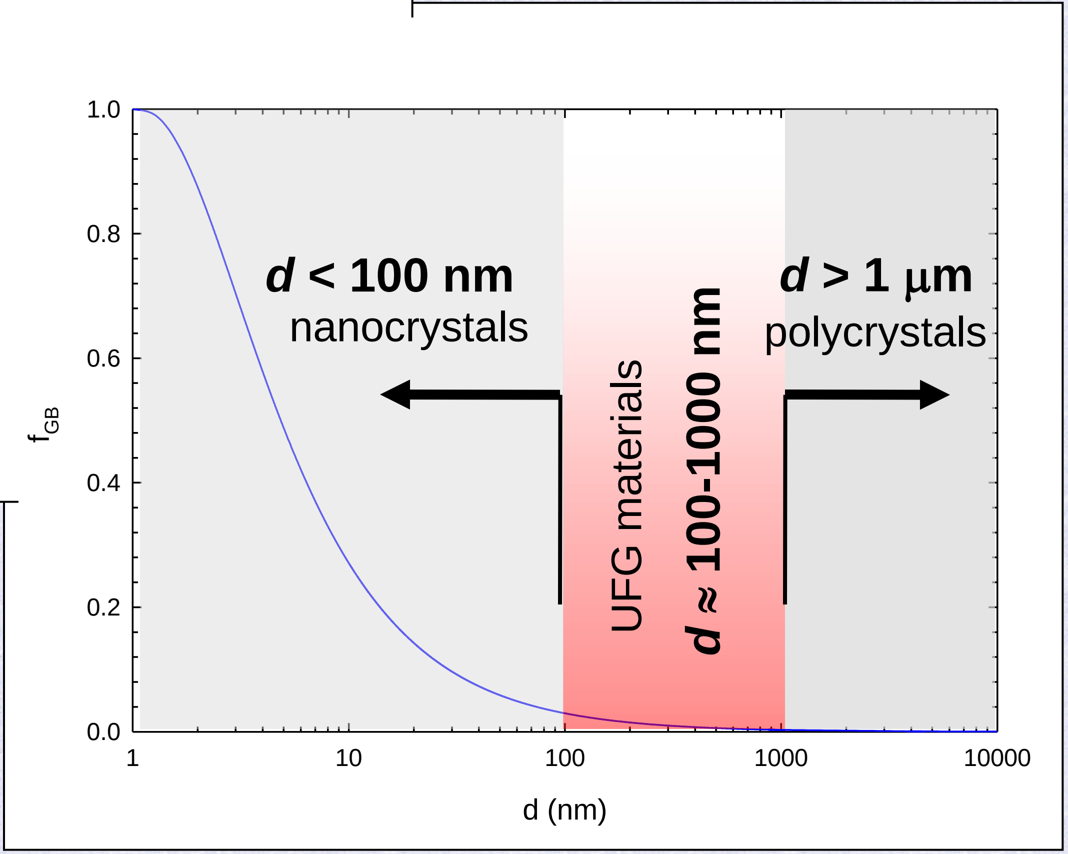
volume fraction of GB's

$$f_{GB} = 1 - \left(\frac{d - \delta}{d} \right)^3$$

Ultra fine grained (UFG) materials

polycrystalline material





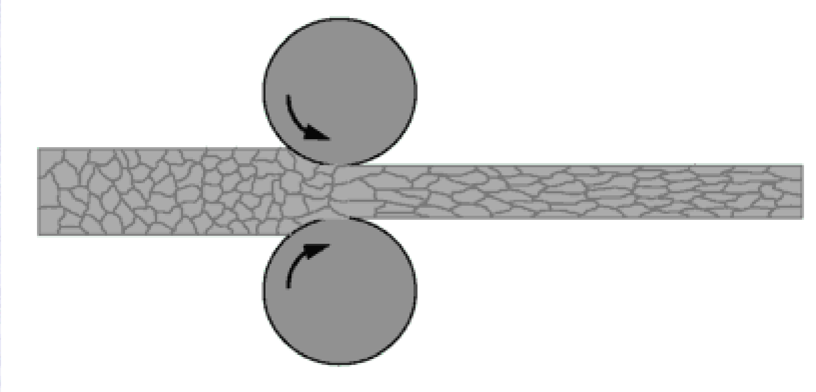
volume fraction of GB's

$$f_{GB} = 1 - \left(\frac{d - \delta}{d} \right)^3$$

Severe plastic deformation

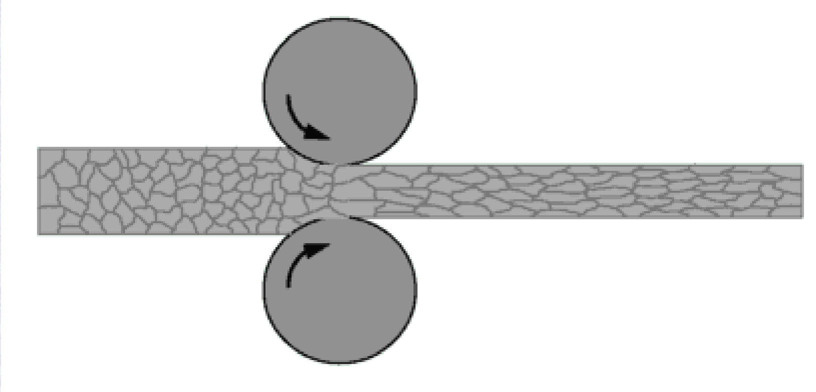
- plastic deformation $\rightarrow$ grain refinement

- conventional plastic deformation



Severe plastic deformation

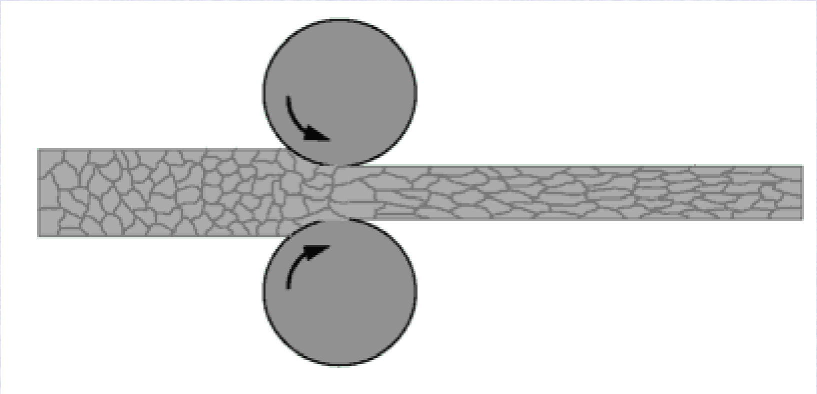
- plastic deformation $\rightarrow$ grain refinement
- conventional plastic deformation $\rightarrow$ formation of cracks

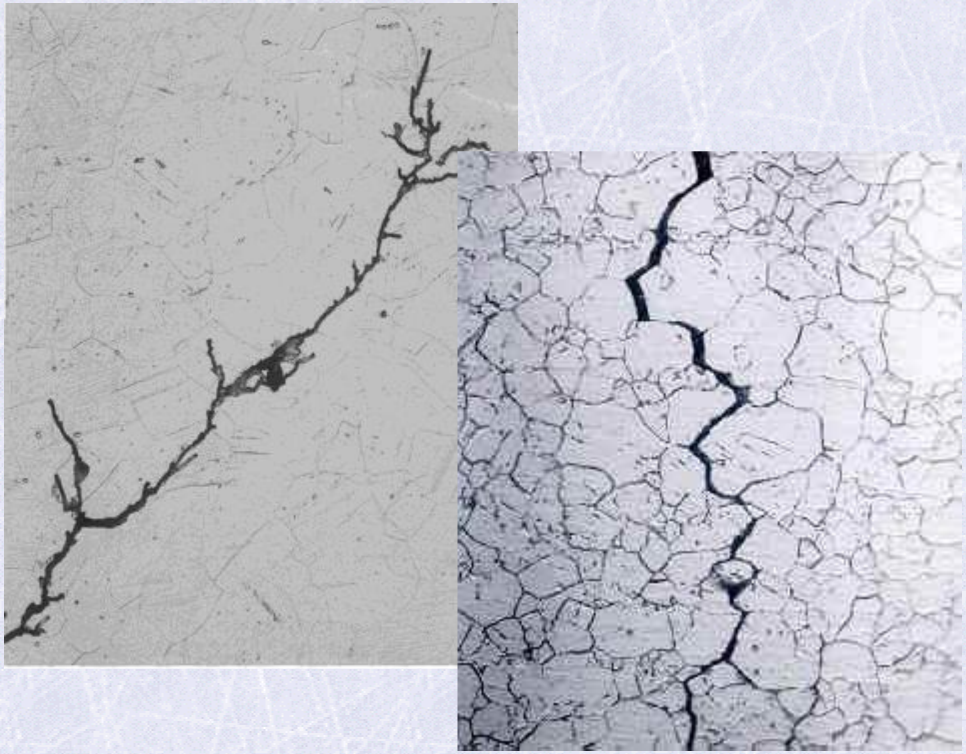


Severe plastic deformation

- plastic deformation $\rightarrow$ grain refinement

- conventional plastic deformation $\rightarrow$ formation of cracks $\rightarrow$ material failure





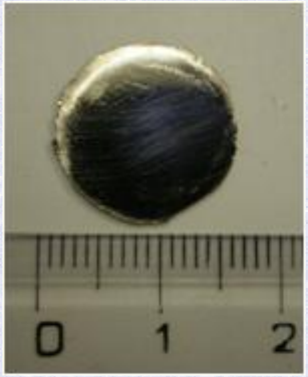
- deformation under **high pressure** $\rightarrow$ crack formation suppressed

High pressure torsion (HPT)

• the strongest grain refinement

• grain size ≈ 100 nm

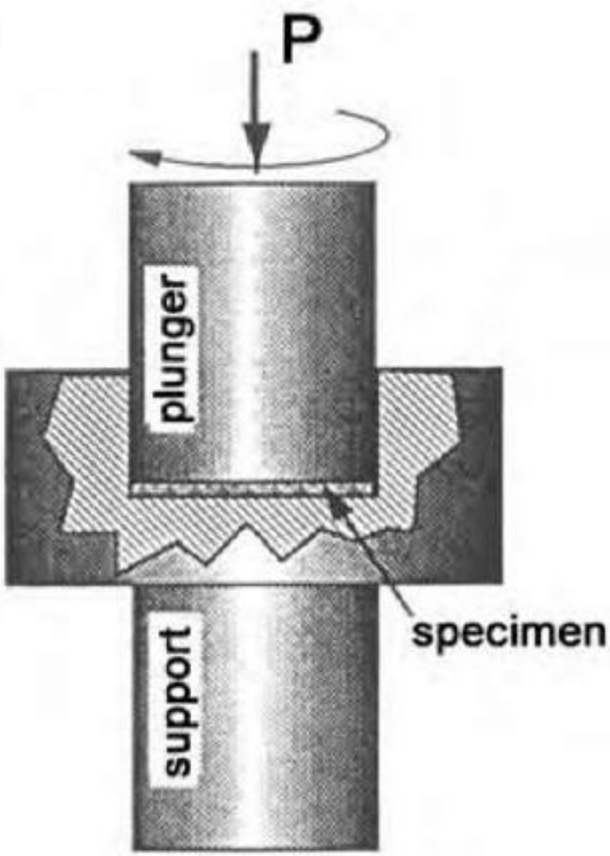
• disk shaped samples
diameter 10 – 20 mm, thickness 0.3 - 0.5 mm



UFG IF steel sample prepared by HPT

• dislocation density is an important parameter of UFG materials

high pressure 1 - 10 GPa



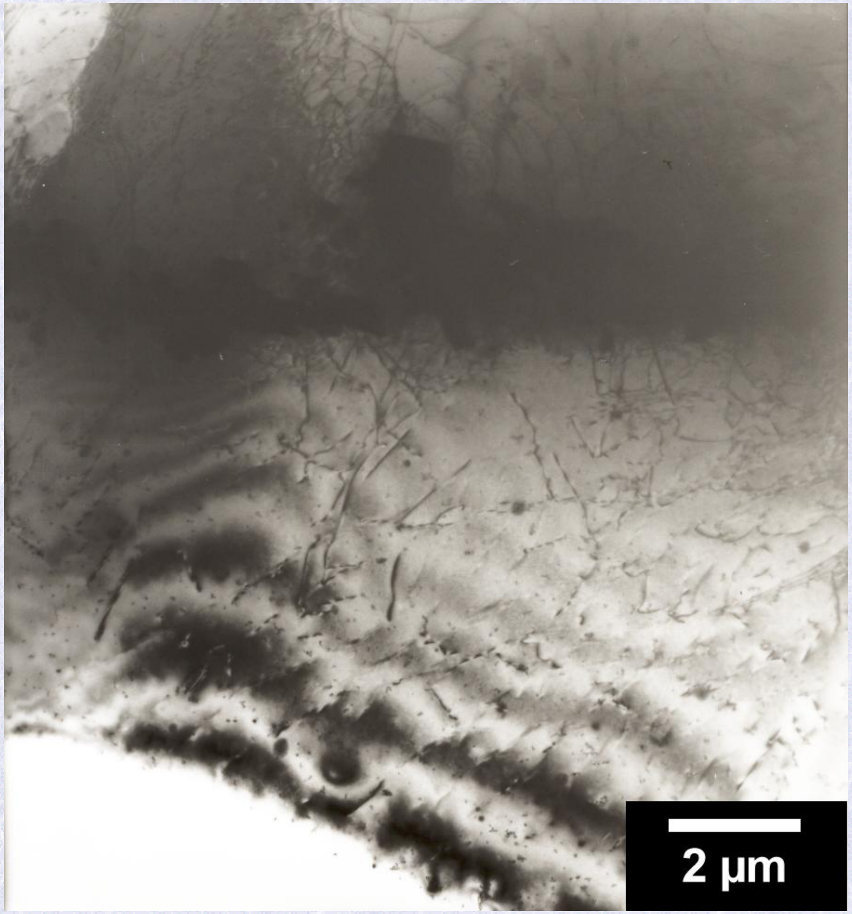
torsion deformation

R.Z. Valiev, Nature Materials 3, 511-516 (2004)

Methods for determination of dislocation density in UFG materials

- **TEM**

  - direct observation of dislocations

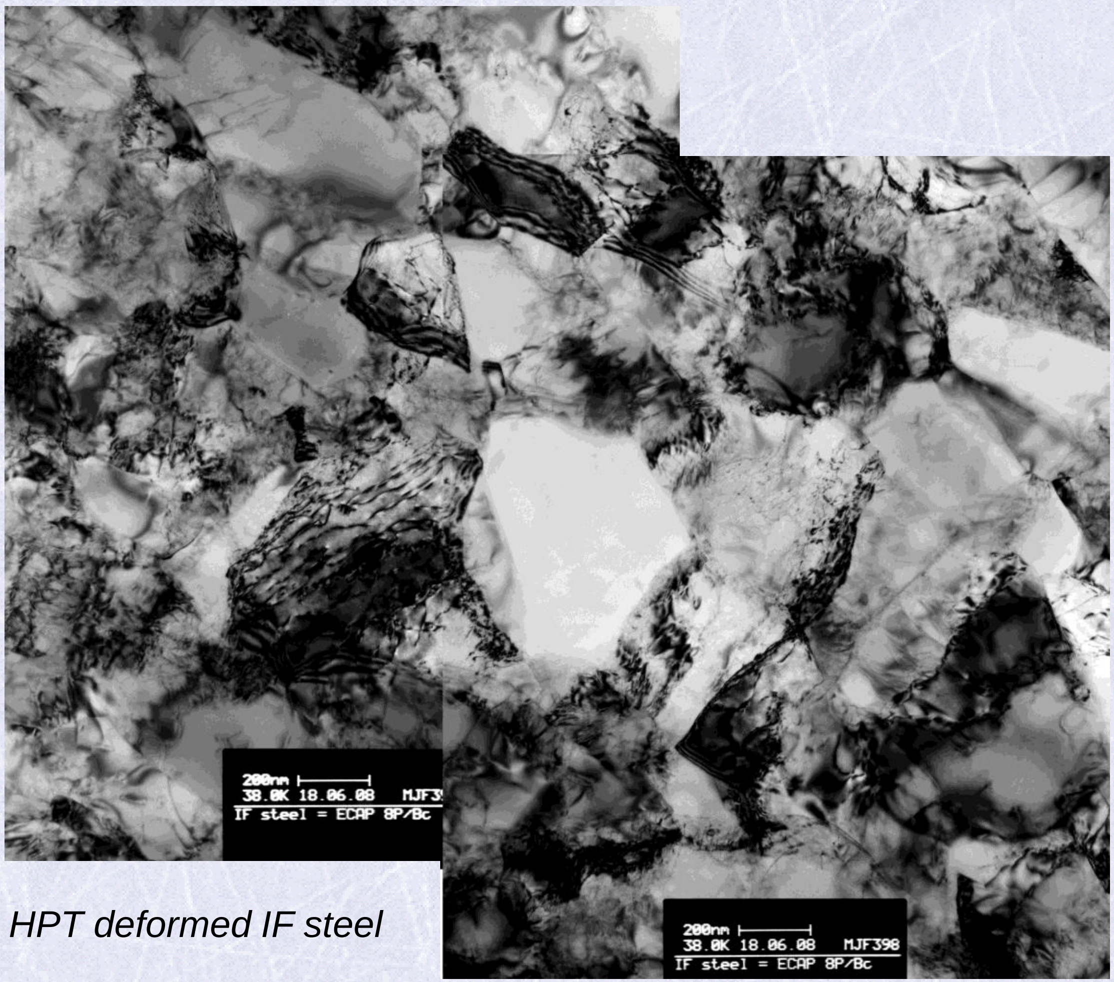


Methods for determination of dislocation density in UFG materials

- **TEM**

  - direct observation of dislocations

  - hard to resolve individual dislocations due to high dislocation density



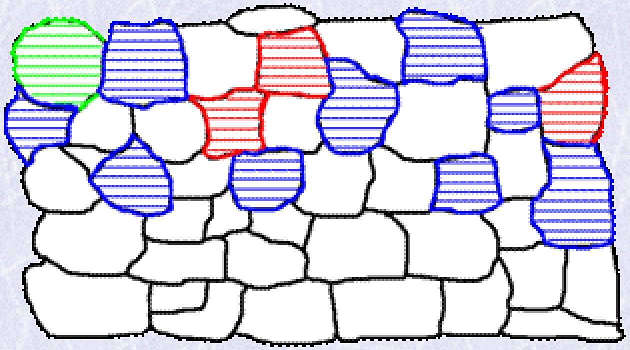
HPT deformed IF steel

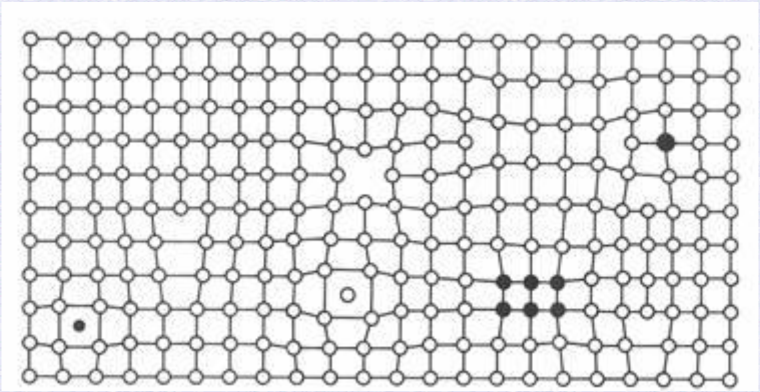
Methods for determination of dislocation density in UFG materials

• **X-ray diffraction (XRD)**

- broadening of XRD profiles

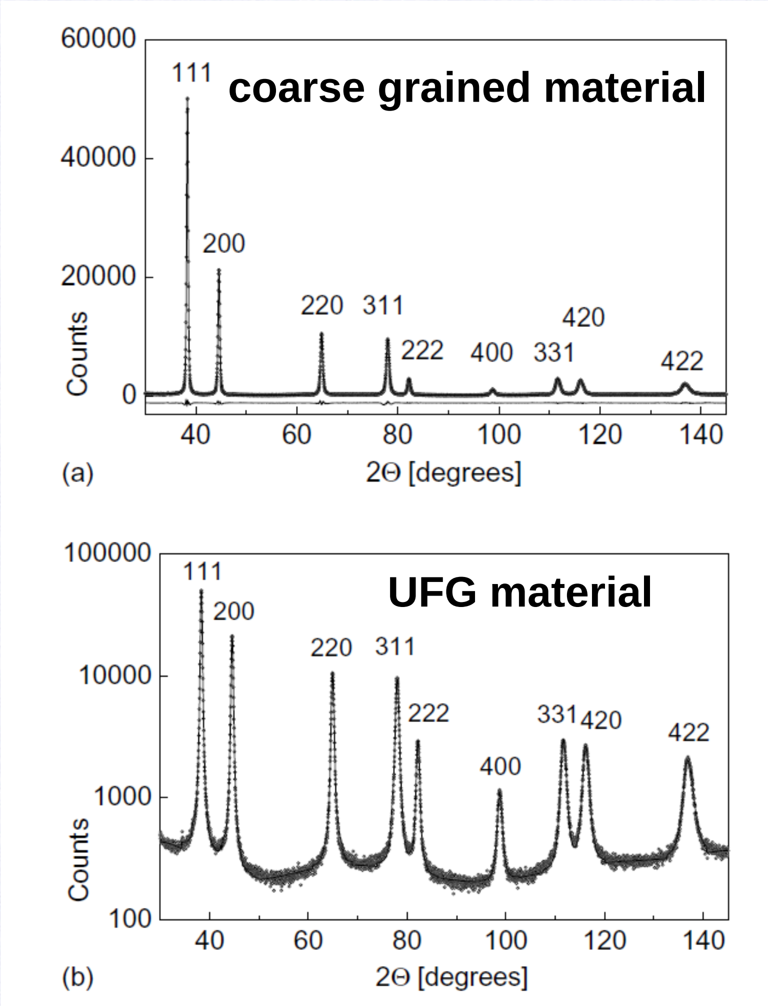
- crystallite size & microstrain broadening





Methods for determination of dislocation density in UFG materials

- **X-ray diffraction (XRD)**
  - broadening of XRD profiles
  - crystallite size & microstrain broadening
  - different dependence on scattering angle



G. Ribárik et al. Mater. Sci. Eng. A 387-389, 343 (2004)

XRD line profile analysis

- **Williamson-Hall (W-H) method**

G.K. Williamson, W.H. Hall , Acta Metall. 1, 22 (1953)

\- size broadening $\rightarrow$ independent on θ

\- strain broadening $\rightarrow$ proportional to sin θ

XRD line profile analysis

- **Williamson-Hall (W-H) method**

G.K. Williamson, W.H. Hall , Acta Metall. 1, 22 (1953)

- *hkl* peak width: $\Delta K = 2\Delta\theta_{hkl}\cos\theta_{hkl}/\lambda$

- magnitude of diffraction vector: $K = 2\sin\theta_{hkl}/\lambda$

$$\Delta K = \frac{0.9}{D} + \xi b\sqrt{\rho}K$$

crystallite size

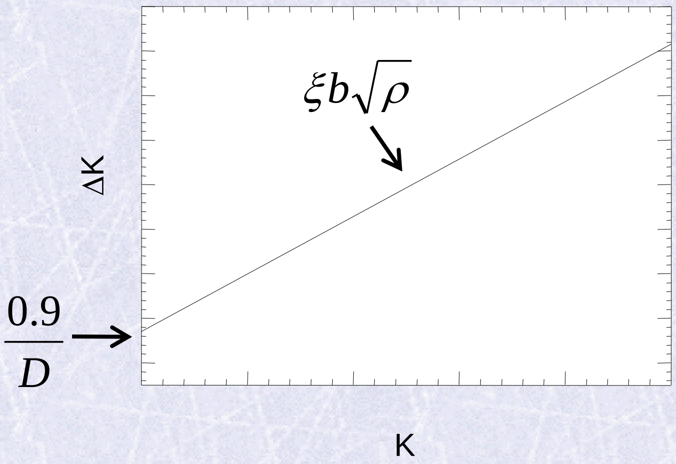
Burgers vector

dislocation density

strain broadening

size broadening

W-H plot



XRD line profile analysis

- **Williamson-Hall (W-H) method**

G.K. Williamson, W.H. Hall , Acta Metall. 1, 22 (1953)

- *hkl* peak width: $\Delta K = 2\Delta\theta_{hkl}\cos\theta_{hkl}/\lambda$

- magnitude of diffraction vector: $K = 2\sin\theta_{hkl}/\lambda$

$$\Delta K = \frac{0.9}{D} + \xi b\sqrt{\rho}K$$

crystallite size

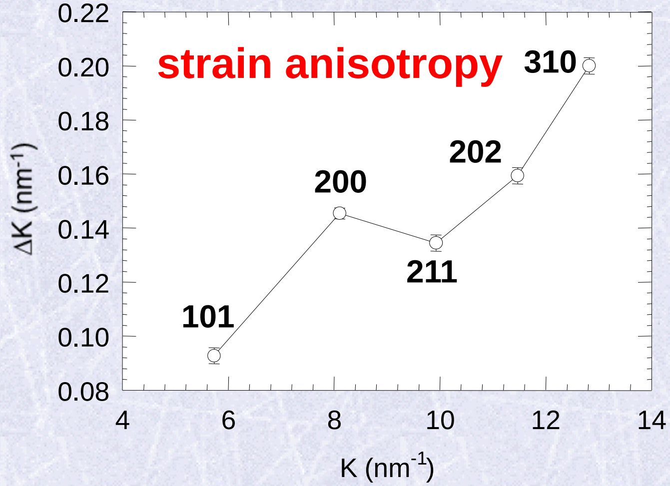
Burgers vector

dislocation density

strain broadening

size broadening

W-H plot



HPT deformed IF steel

Methods for determination of dislocation density in UFG materials

- **modified Williamson-Hall (W-H) method**

T. Ungár, A. Borbély, Appl. Phys. Lett. 69, 3173, (1996)

- hkl peak width: $\Delta K = 2\Delta\theta_{hkl}\cos\theta_{hkl}/\lambda$

- magnitude of diffraction vector: $K = 2\sin\theta_{hkl}/\lambda$

$$\Delta K = \frac{0.9}{D} + bM\sqrt{\frac{\pi}{2}\rho}\left(K\overline{C}^{1/2}\right)$$

dislocation distribution parameter

dislocation contrast factor

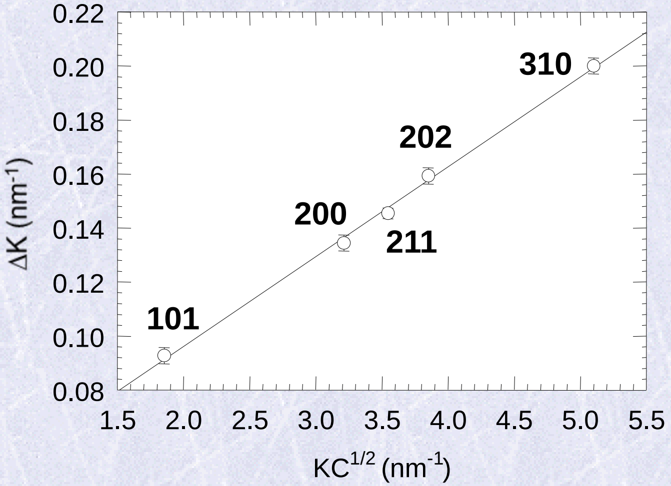
$$\overline{C} = \overline{C}_{h00}\left(1 - qH^2\right)$$

$$H^2 = \frac{h^2l^2 + h^2k^2 + l^2k^2}{\left(h^2 + k^2 + l^2\right)^2}$$

edge / screw character of dislocations

dislocation contrast factor for $\{h00\}$ reflections

Modified W-H plot



HPT deformed IF steel

fraction of screw dislocations:

$$f_{screw} = \frac{q - q_{edge}}{q_{screw} - q_{edge}}$$

for common slip systems in fcc and bcc structures calculated in T. Ungár et al. J. Appl. Cryst 32, 992 (1999)

XRD line profile analysis

- **modified Williamson-Hall (W-H) method**

linearization of relation

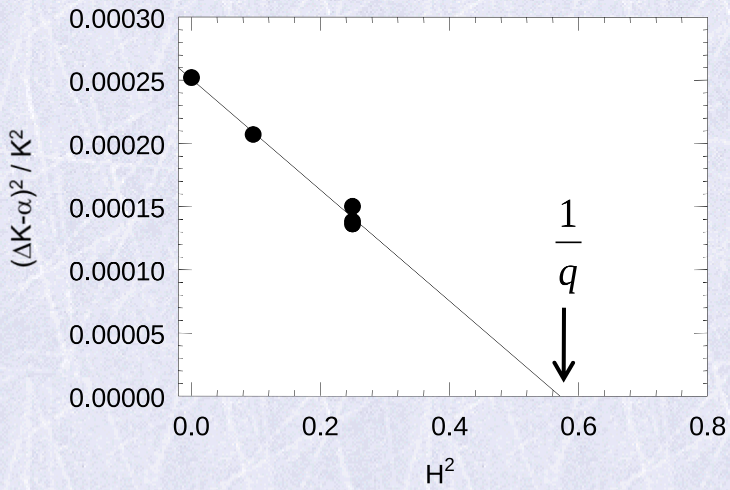
$$\frac{(\Delta K-\alpha)^2}{K^2}=\beta^2\overline{C}_{h00}\left(1-qH^2\right)$$

gives parameters:

$$\alpha=\frac{0.9}{D}$$ (crystallite size)

$$\beta=bM\sqrt{\frac{\pi}{2}\rho}$$ (dislocation density)

q (screw / edge character of dislocations)

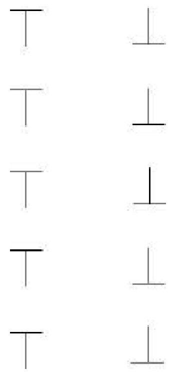


HPT deformed IF steel

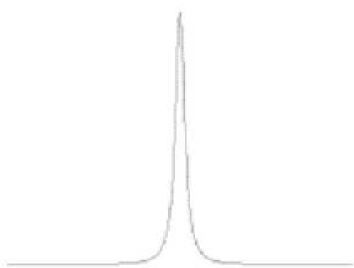
XRD line profile analysis

dislocation distribution parameter M

$M \ll 1$



dislocations form dipoles

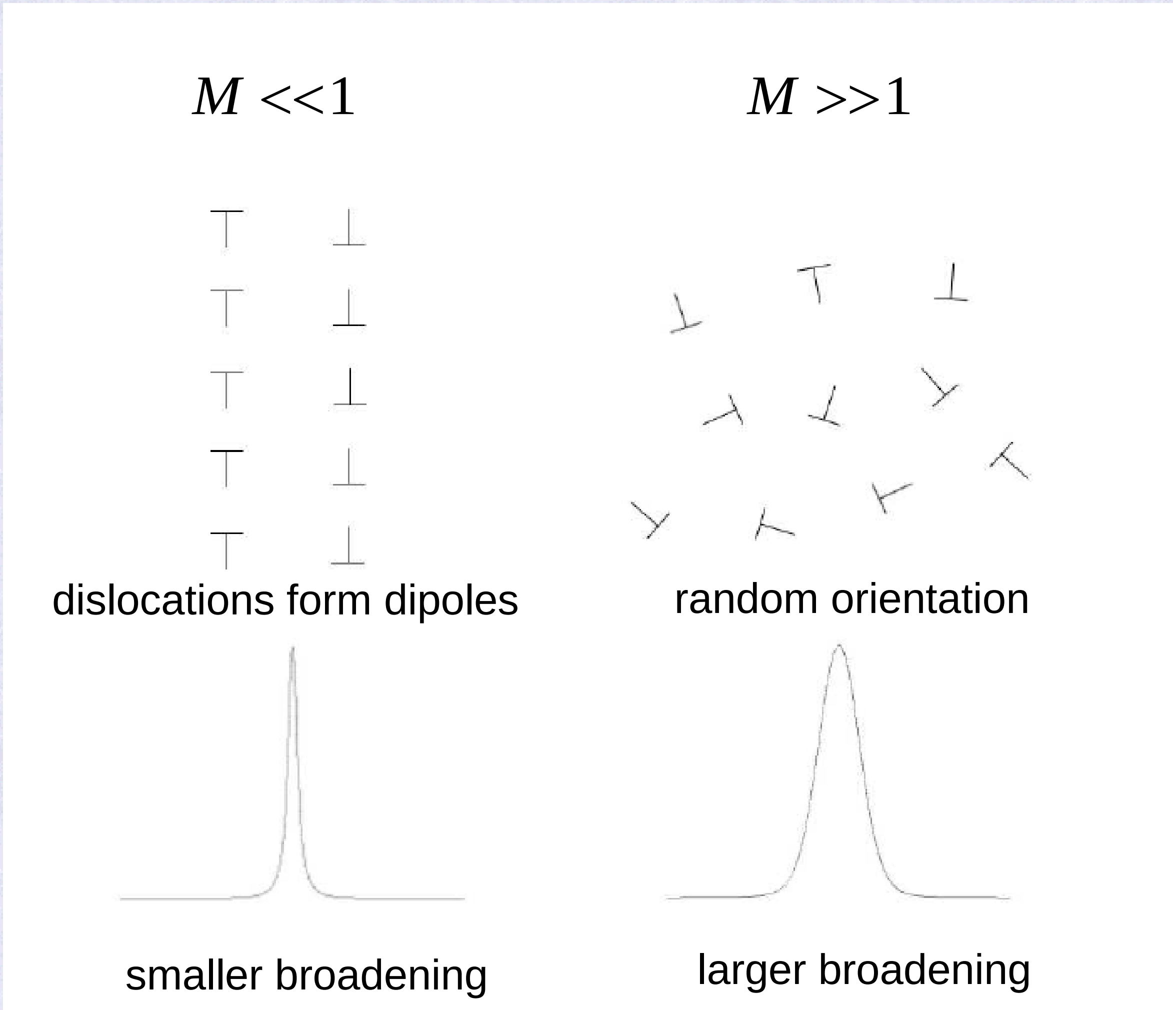


smaller broadening

G. Ribárik, PhD. Thesis (2008)

XRD line profile analysis

dislocation distribution parameter M



G. Ribárik, PhD. Thesis (2008)

XRD line profile analysis

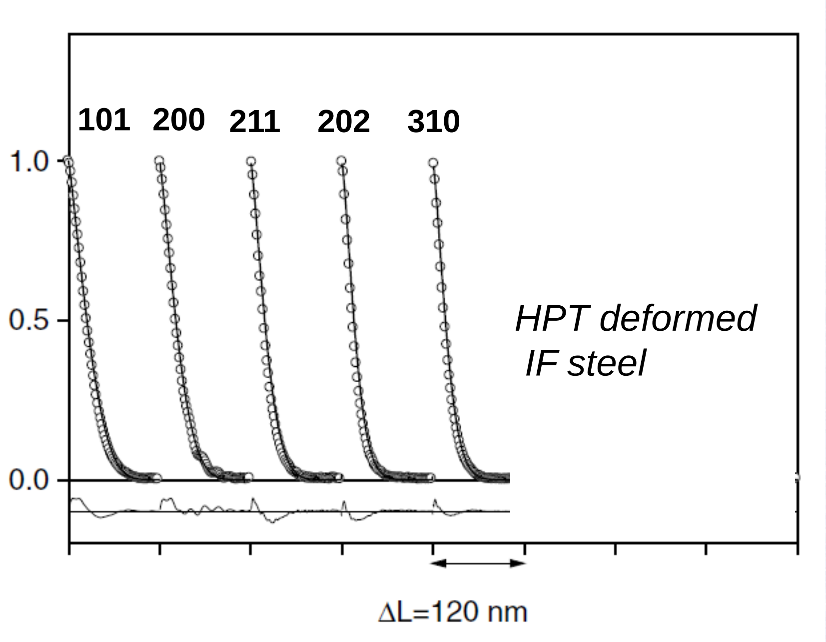
- **modified Warren-Averbach (MWA) method**

T. Ungár, A. Borbély, Appl. Phys. Lett. 69, 3173, (1996)

Fourier transform of XRD profiles

$$A(L) = A_S(L) A_D(L)$$

size coefficient distortion coefficient

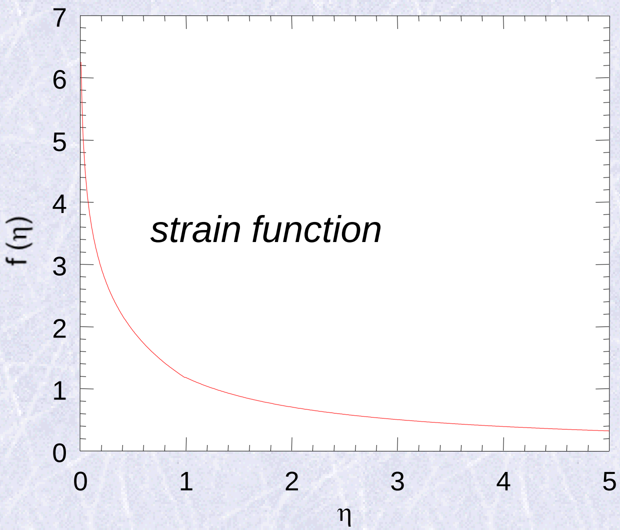


$$A_D(L) = \exp\left(-\frac{\pi}{2} b^2 K^2 \overline{C} \rho L^2 f(\eta) \right)$$

$$\eta = \frac{1}{2} \exp\left(\frac{7}{4}\right) \frac{L}{M} \sqrt{\rho}$$

strain (Wilkens) function

describes dislocation-dislocation correlation



XRD line profile analysis

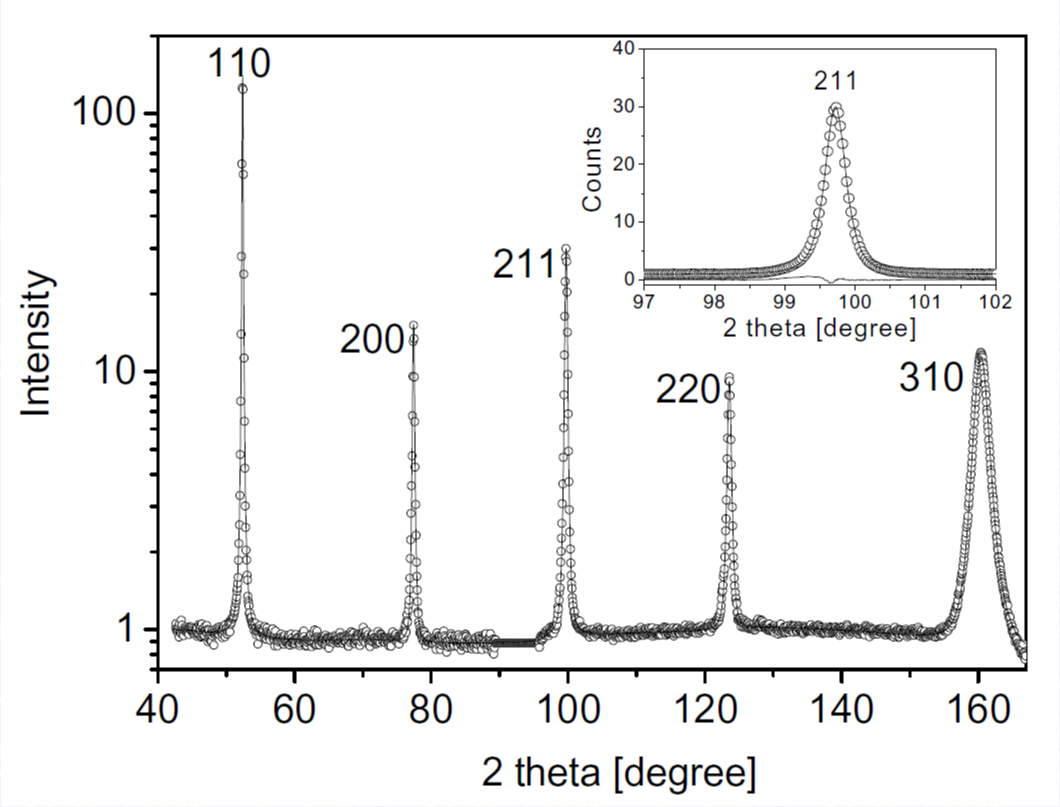
- **whole profile fitting**

G. Ribárik, T. Ungár, J. Gubicza, J. Appl. Cryst. 34, 669, (2001)

by self consistent MWH + MWA approach one gets:

1. mean crystallite size D
2. mean dislocation density ρ
3. screw / edge character of dislocations q
4. dislocation distribution parameter M

HPT deformed IF steel

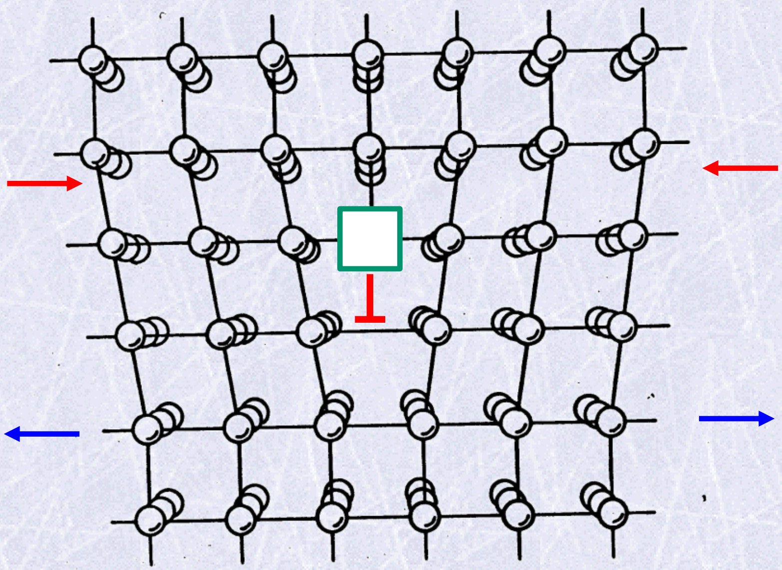


G. Ribárik, T. Ungár, J. Gubicza: Multiple Whole Profile fitting program MWP-fit, http://csendes.elte.hu/mwp/

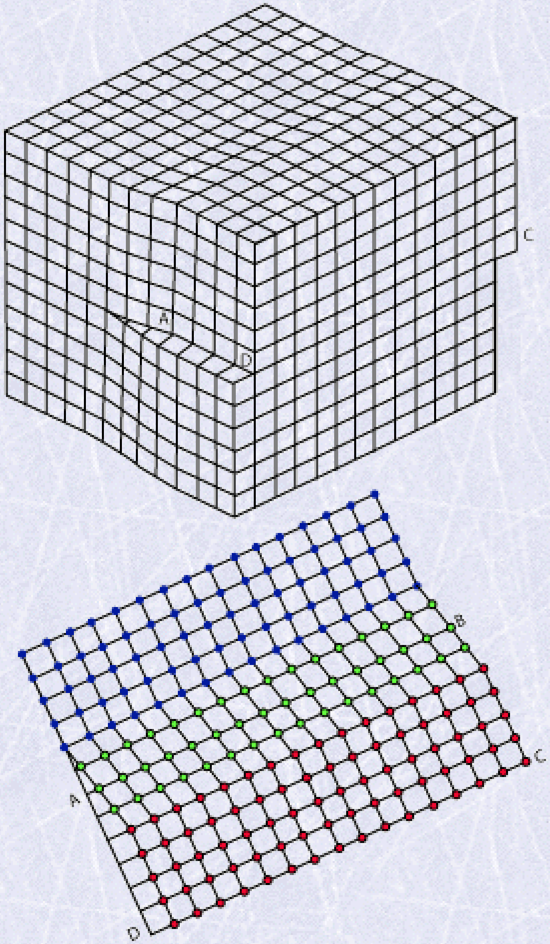
Positron trapping at dislocations

- dislocation line is a shallow positron trap
- weak positron localization at dislocation → diffusion along dislocation line
- final trapping at vacancy bound to dislocation or open volume at jog

edge dislocation



screw dislocation

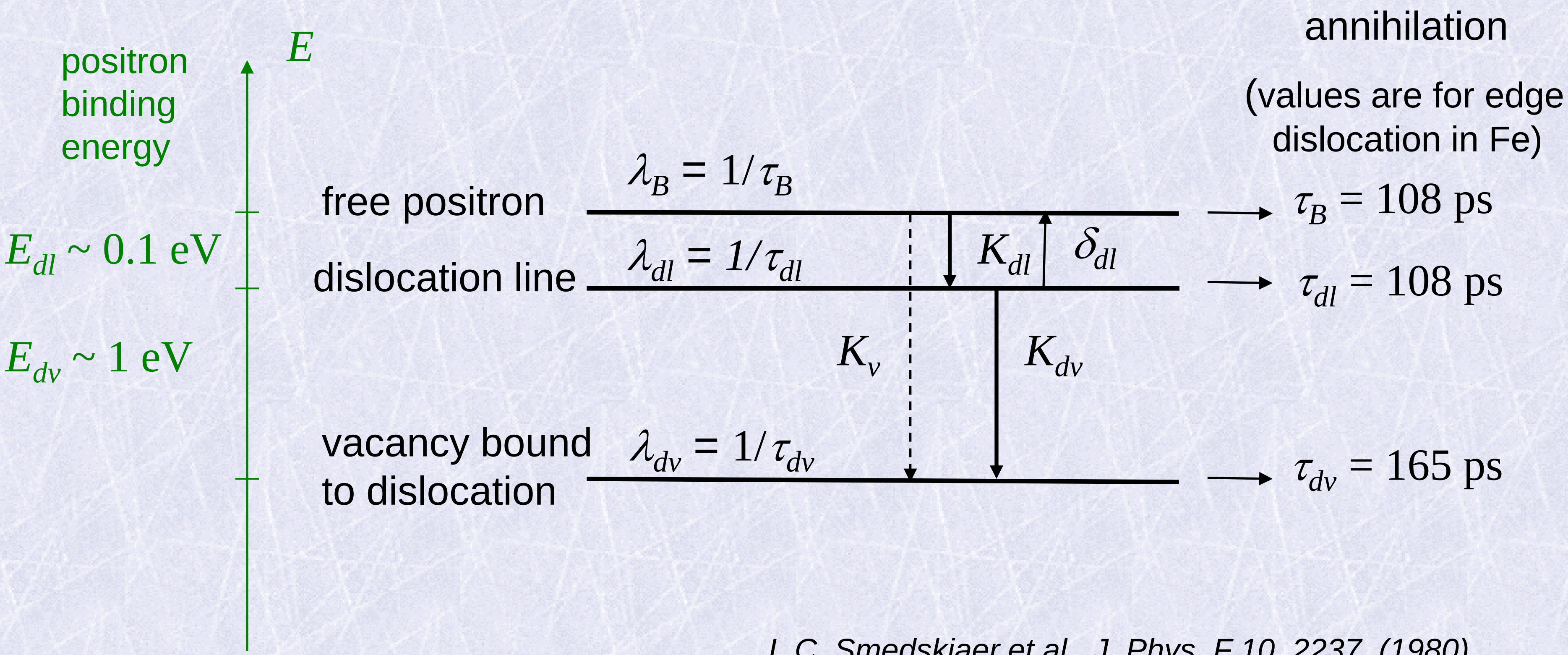


Positron trapping at dislocations

• two-step positron trapping at dislocation

$K_v << K_{dl}$ (vacancy is a point defect but dislocation is a line defect)

$\delta_{dl} << K_{dv}$ (there is always enough vacancies attached to dislocation)



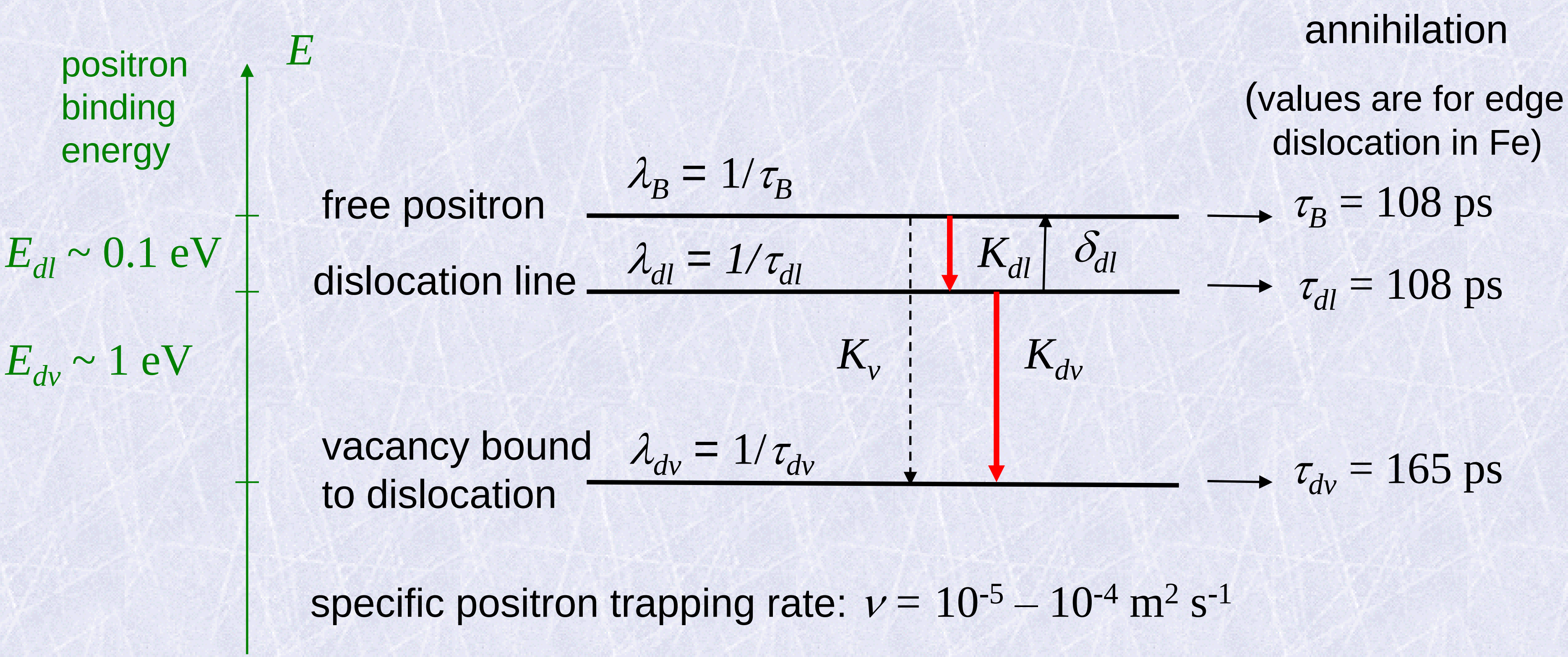
L.C. Smedskjaer et al., J. Phys. F 10, 2237, (1980)

Positron trapping at dislocations

- two-state positron trapping at dislocation

$K_v << K_{dl}$ (vacancy is a point defect but dislocation is a line defect)

$\delta_{dl} << K_{dv}$ (there is always enough vacancies attached to dislocation)



specific positron trapping rate: $\nu = 10^{-5} - 10^{-4}$ m^2 s^{-1}

dislocation density: $\rho = 10^{12} - 10^{16}$ m^{-2}

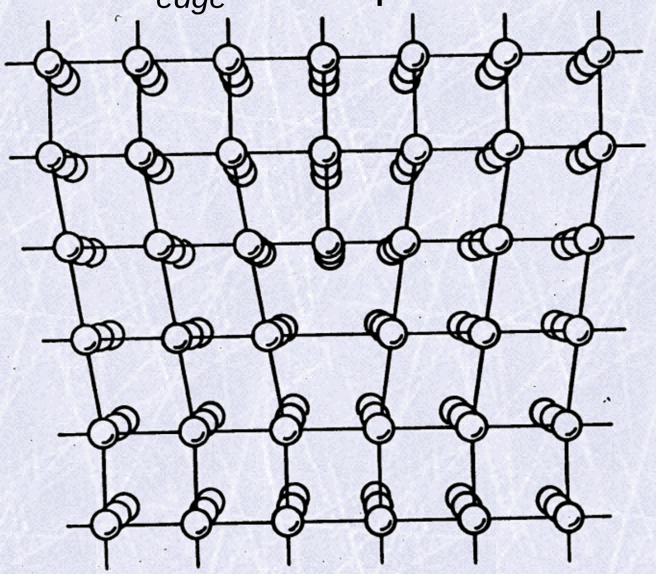
Positron trapping at dislocations in Fe (or steels)

• open volumes attached to edge dislocations are larger *Y.K. Park et al. , PRB 34, 823 (1986)*

• screw and edge dislocations can be distinguished

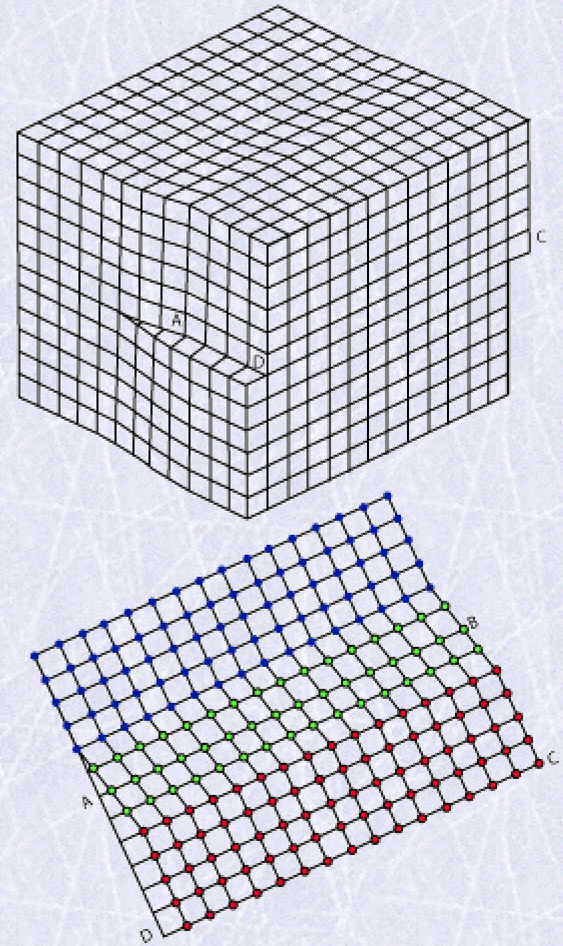
edge dislocation

τ_{edge} = 165 ps



screw dislocation

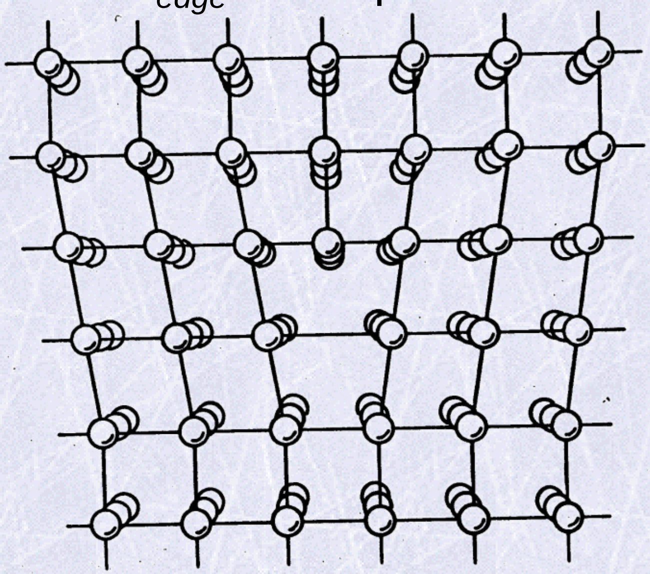
τ_{screw} = 142 ps



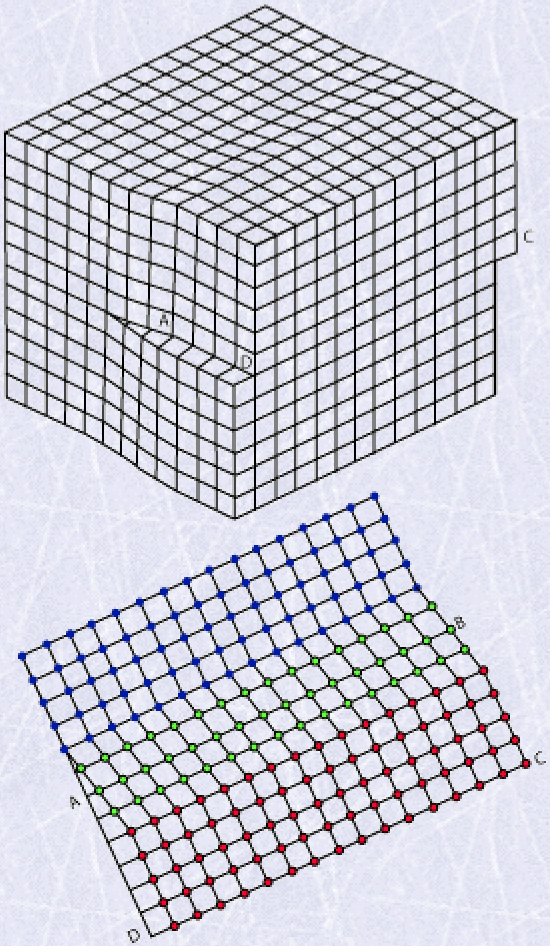
Positron trapping at dislocations in Fe (or steels)

- open volumes attached to edge dislocations are larger *Y.K. Park et al. , PRB 34, 823 (1986)*
- screw and edge dislocations can be distinguished
- fraction of screw dislocations: $f_{screw} = \dfrac{\tau - \tau_{edge}}{\tau_{screw} - \tau_{edge}}$

edge dislocation
τ_{edge} = 165 ps



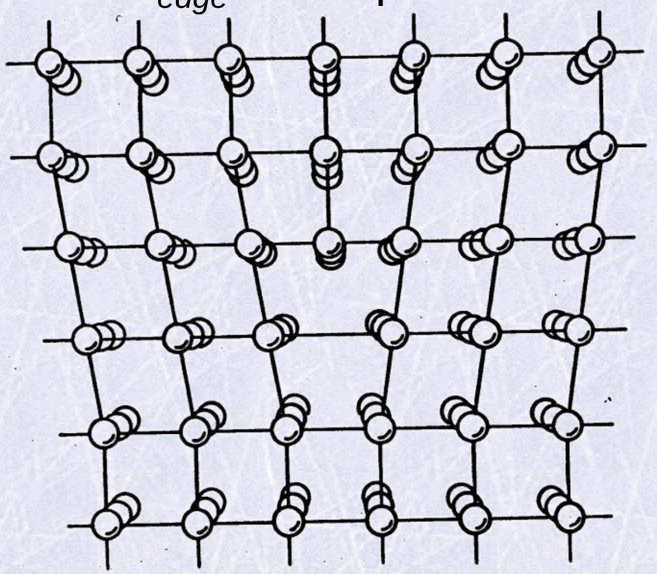
screw dislocation
τ_{screw} = 142 ps



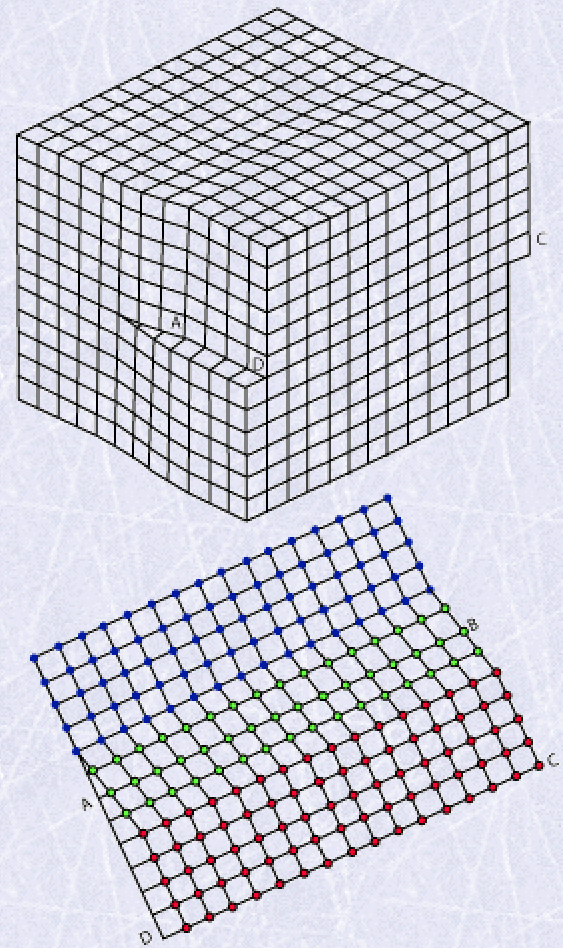
Positron trapping at dislocations in Fe (or steels)

- open volumes attached to edge dislocations are larger *Y.K. Park et al. , PRB 34, 823 (1986)*
- screw and edge dislocations can be distinguished
- typical lifetimes of trapped positrons in deformed steels: 150 – 155 ps

edge dislocation
τ_{edge} = 165 ps



screw dislocation
τ_{screw} = 142 ps



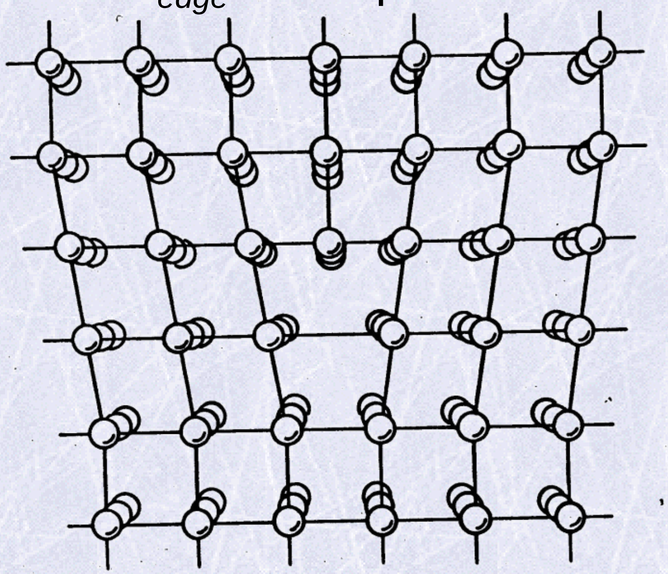
Positron trapping at dislocations in Fe (or steels)

- open volumes attached to edge dislocations are larger *Y.K. Park et al. , PRB 34, 823 (1986)*

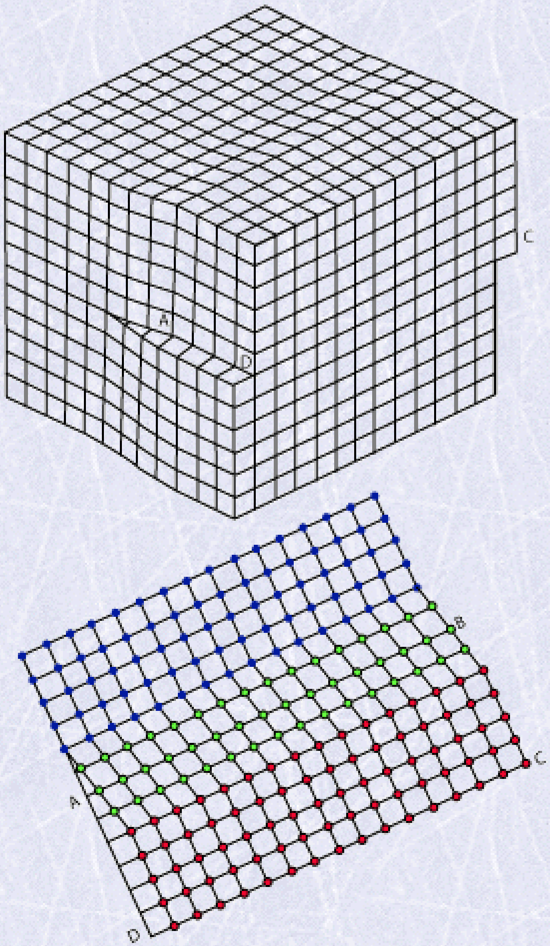
- screw and edge dislocations can be distinguished

- unfortunately the lifetimes for edge and screw dislocations have been determined only for Fe so far

edge dislocation
τ_{edge} = 165 ps



screw dislocation
τ_{screw} = 142 ps



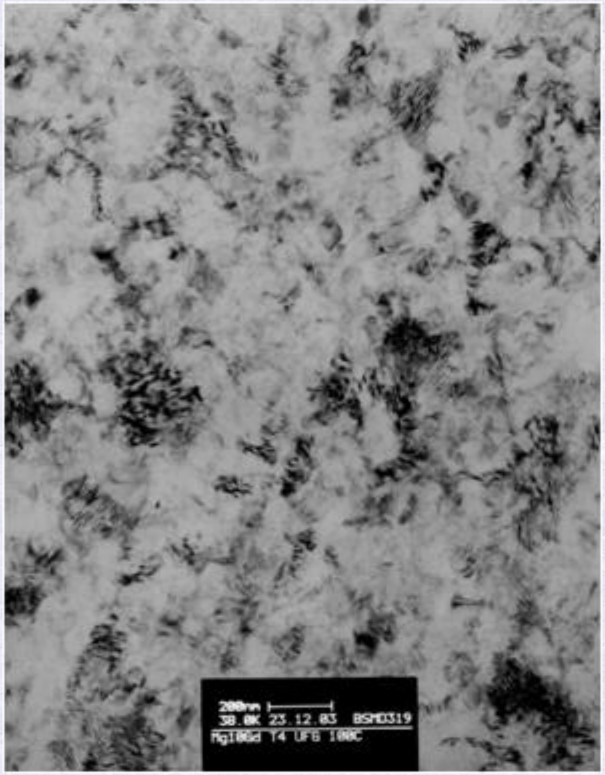
Positron trapping at dislocations – distribution of dislocations

- uniform distribution of dislocations → simple trapping model
  (hcp metals, metals with low SFE)

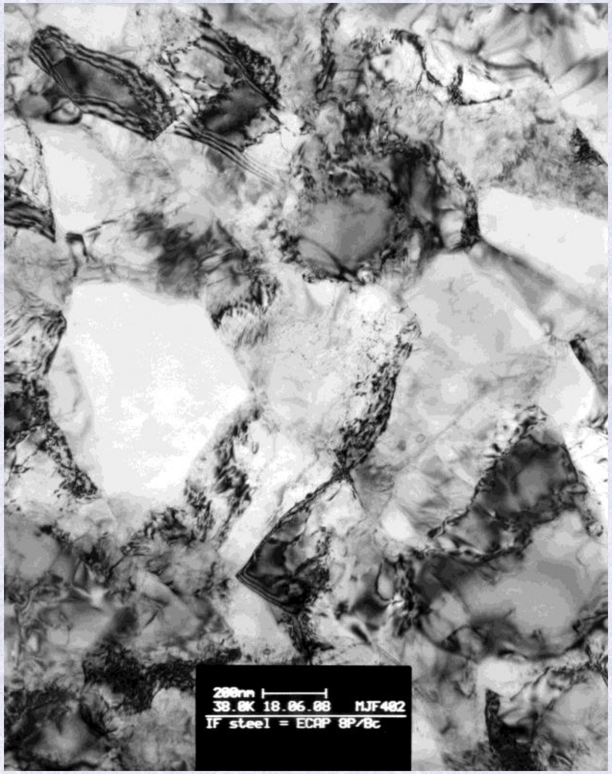
$$\rho = \frac{1}{\nu} \frac{I_2}{I_1} \left(\frac{1}{\tau_B} - \frac{1}{\tau_D} \right)$$

specific positron trapping rate to dislocations

- dislocation cell structure → diffusion trapping model
  (cubic metals with medium and high SFE)



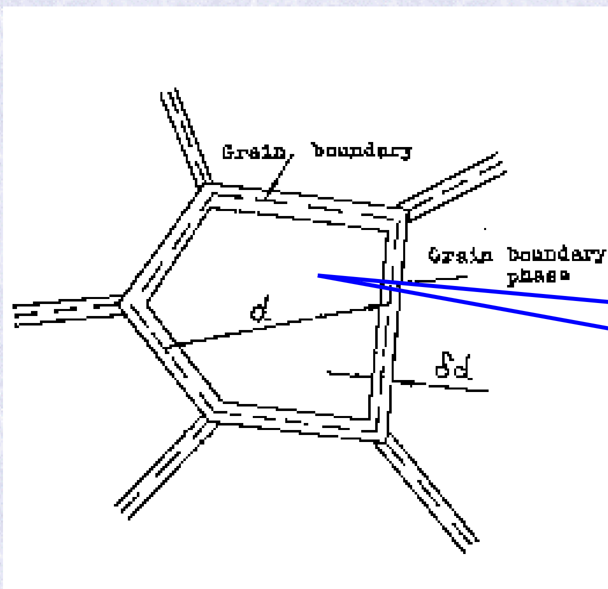
HPT-deformed Mg-10wt.%Gd alloy

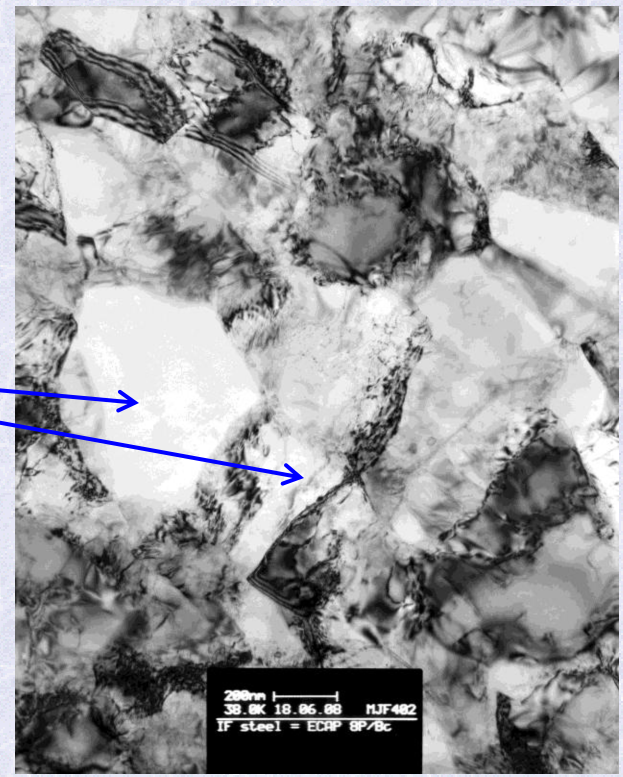


HPT-deformed IF steel

Positron trapping at dislocations – dislocation cell structure

- dislocation cell structure
- dislocation-free cell interiors

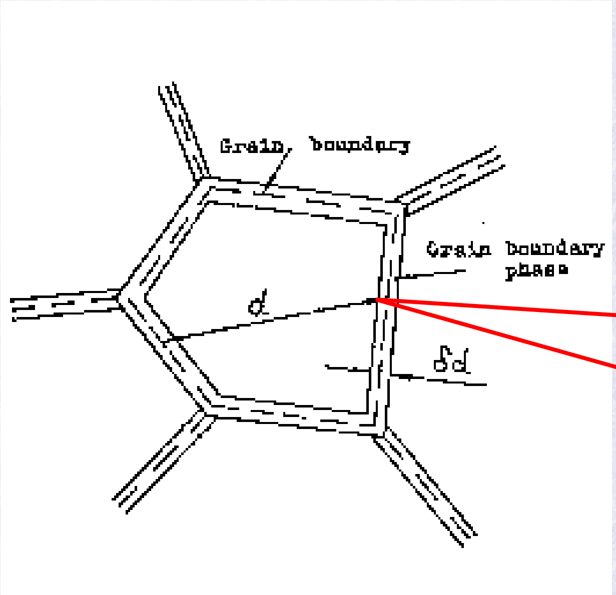


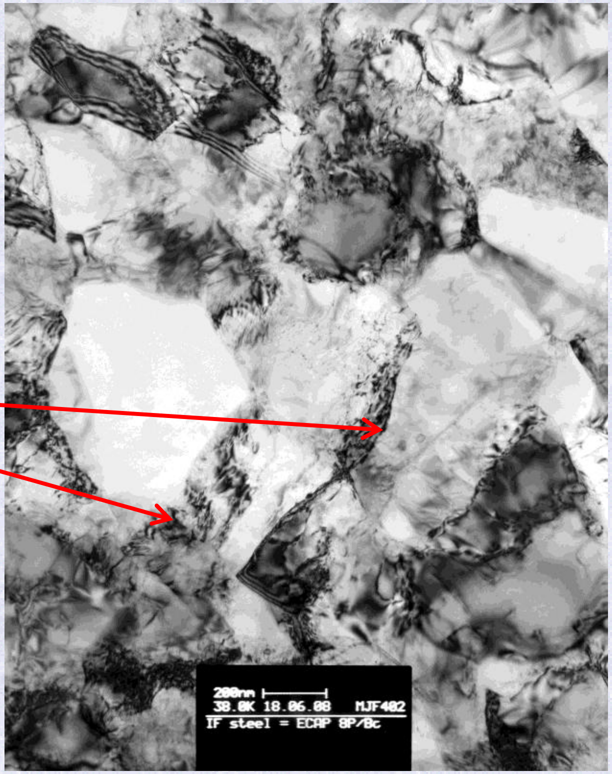


HPT-deformed IF steel

Positron trapping at dislocations – dislocation cell structure

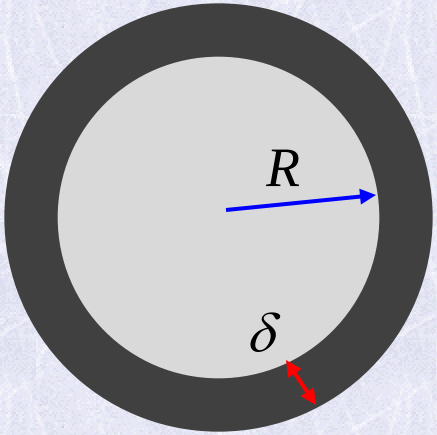
- dislocation cell structure
- dislocation-free cell interiors
- distorted regions with high density of dislocations (dislocation walls)





HPT-deformed IF steel

Diffusion trapping model (DTM)



• dislocation-free spherical cells with radius R

• surrounded by dislocation walls with thickness δ

thermalization

1. positrons stopped at distorted regions

trapping at dislocations

$$\frac{\partial n}{\partial t} = D_+ \left(\frac{\partial^2 n}{\partial r^2} + \frac{2}{r}\frac{\partial n}{\partial r} \right) - \lambda_B n$$

diffusion to distorted regions

annihilation from free state

$$\left(\frac{\partial n}{\partial t} \right)_{r=R} = -\frac{\nu \rho \delta}{\eta D_+} n(R,t)$$

boundary condition

2. positrons stopped inside cells

D_+ - positron diffusion coefficient

$$n(r,0) = \frac{1-\eta}{4/3\pi R^3}$$

initial condition

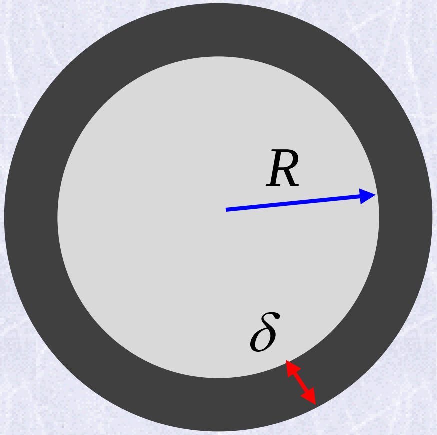
$$\eta = \frac{(R+\delta)^3 - R^3}{(R+\delta)^3}$$

volume fraction of distorted regions

A. Dupasquier et al. PRB 48, 9235 (1993)

J. Čížek et al. PRB 65, 094106 (2002)

Diffusion trapping model (DTM)



- spherical grains with radius R
- distorted regions along grain boundaries with thickness δ
- positron lifetime spectrum

$$S(t) = \sum_{k}^{\infty} t_k^{-1} i_k e^{-t/t_k} + \tau_d^{-1} I_d e^{-t/\tau_d}$$

τ_d – lifetime of positrons trapped at dislocations

$I_d = 1 - \sum_{k}^{\infty} i_k$ – intensity of dislocation component

$t_k = \left(\tau_B^{-1} + \frac{\beta_k^2 D_+}{R^2} \right)^{-1}$ – infinite number of ”free positron” components

$$i_k = 3(1-\eta) \frac{\nu \rho \delta}{\eta R} \alpha_k \left(\frac{1}{t_k^{-1} - \tau_B^{-1}} - \frac{1}{t_k^{-1} - \tau_d^{-1}} \right)$$

$$\beta_k \cot \beta_k + \xi - 1 = 0$$

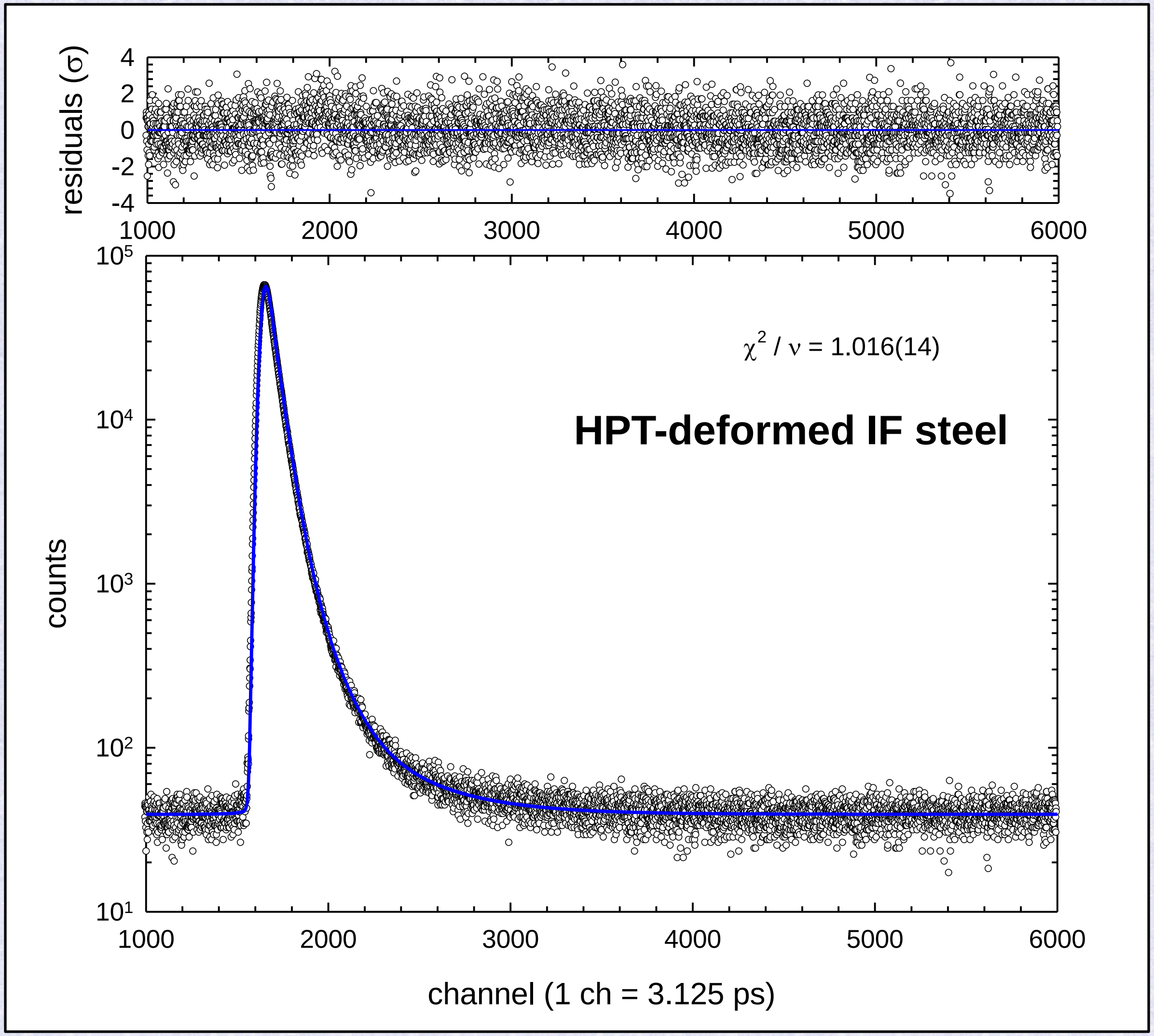
$$\alpha_k = \frac{2\xi}{\beta_k^2 + \xi(\xi - 1)} \qquad \xi = \frac{\nu R \rho \delta}{\eta D_+}$$

A. Dupasquier et al. PRB 48, 9235 (1993)

J. Čížek et al. PRB 65, 094106 (2002)

Diffusion trapping model (DTM)

- direct fitting of positron lifetime spectra by DTM
- from fitting we obtain the following structural parameters:
- size of cells $2R$
- mean dislocation density ρ
- volume fraction of distorted regions η
- lifetime of positrons trapped at dislocations τ_d
- fraction of screw dislocations f_{screw}



Diffusion trapping model (DTM)

- direct fitting of positron lifetime spectra by DTM
- fixed parameters

HPT-deformed IF steel

- width of distorted regions δ = 10 nm
- specific positron trapping rate to dislocations ν = 0.36 $\times$ 10^{-4} m^{2}s^{-1}

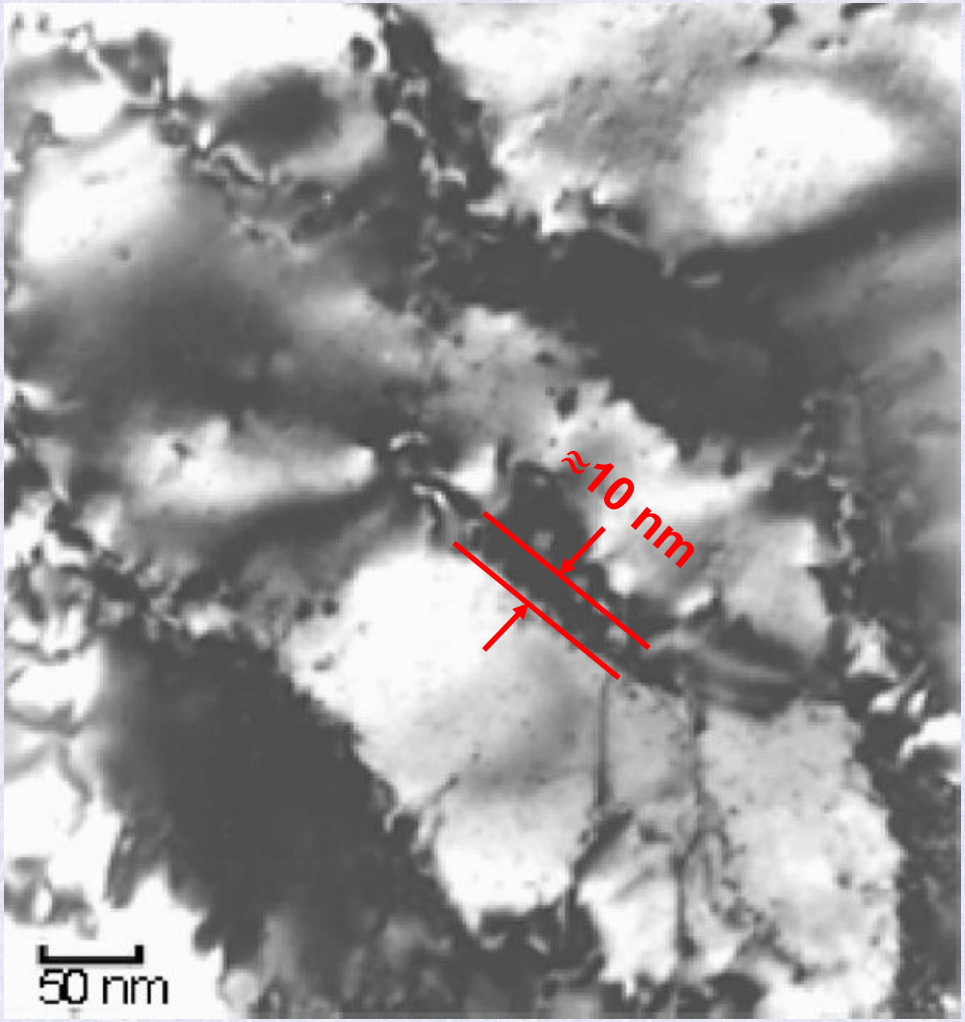
J. Čížek et al., Phys. Stat. Sol. A 178, 651 (2000)

- bulk positron lifetime for Fe
  τ_B = 108 ps

F. Bečvář et al., Appl. Surf. Sci. 255, 111 (2008)

- positron diffusion coefficient for Fe
  D_+ = 1.87 cm^2 s^{-1}

F. Lukáč et al., J. Phys. Conf. Ser. 443, 012025 (2013)



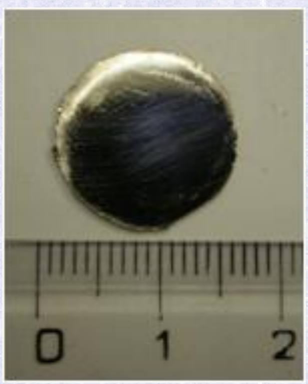
PAS and XRD

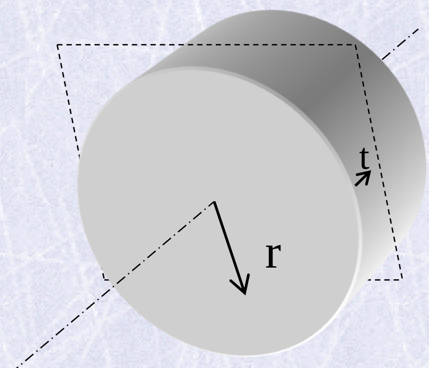
- **PAS**
- size of dislocation-free cells $2R$
- mean dislocation density ρ
- fraction of screw dislocations f_{screw}
- open volume point defects (vacancies, vacancy clusters)

- **XRD line profile analysis**
- size of crystallites D (coherently diffracting domains)
- mean dislocation density ρ
- fraction of screw dislocations f_{screw}
- dislocation distribution parameter M
- lattice parameters

High pressure torsion (HPT)

- Interstitial-free steel
  Pohang Steel Company (POSCO), Korea
- composition (wt.%):
  0.0026 C, 0.096 Mn, 0.045 Al, 0.041 Ti
- number of HPT revolutions *N* = ¼, ½, 1, 3, 5
- disk shaped samples
  diameter 14 mm, thickness 0.3 mm

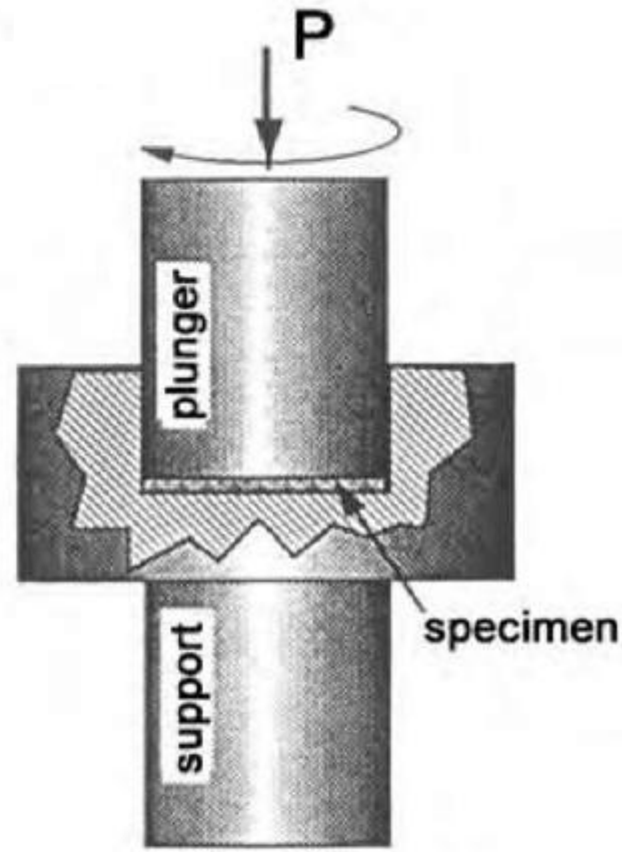




$$e = \frac{1}{\sqrt{3}} \frac{2\pi r N}{l}$$

- e - true strain
- N – number of rotations
- r - distance from center
- l - sample thickness

high pressure 2.5 GPa

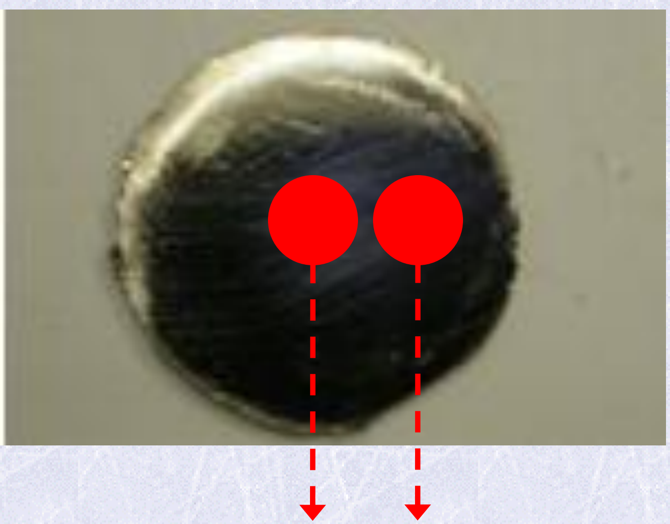


torsion deformation

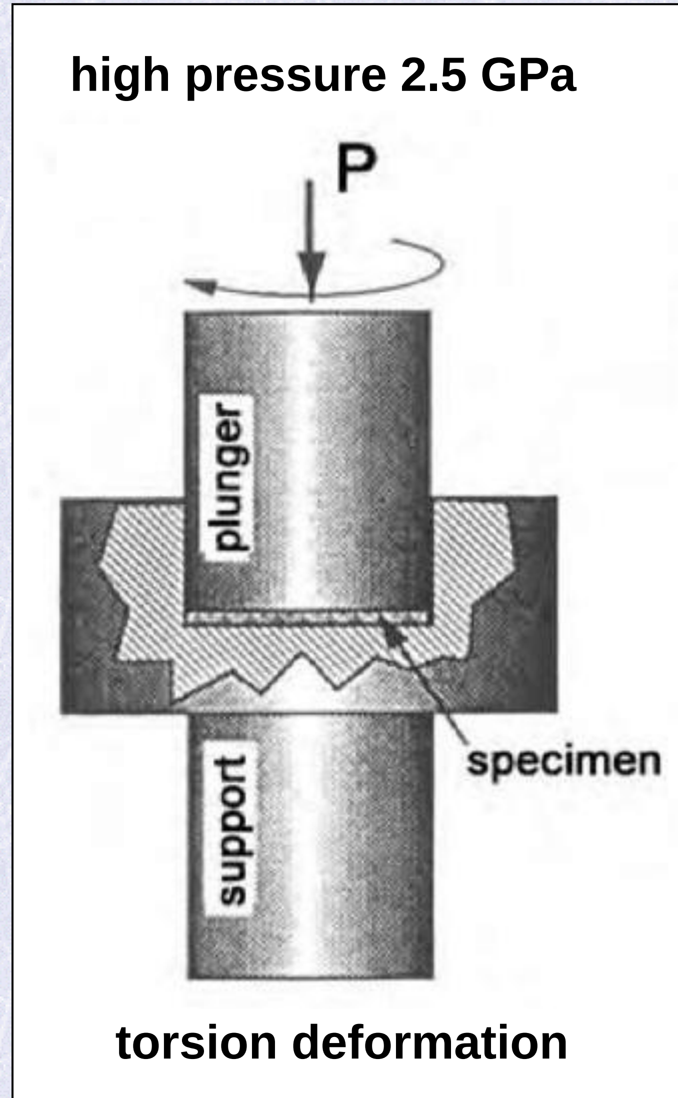
R.Z. Valiev, Nature Materials 3, 511-516 (2004)

High pressure torsion (HPT)

- Interstitial-free steel
  Pohang Steel Company (POSCO), Korea
- composition (wt.%):
  0.0026 C, 0.096 Mn, 0.045 Al, 0.041 Ti
- number of HPT revolutions *N* = ¼, ½, 1, 3, 5
- disk shaped samples
  diameter 14 mm, thickness 0.3 mm



center   *half-radius*
r = 0   *r* = 3.5 mm

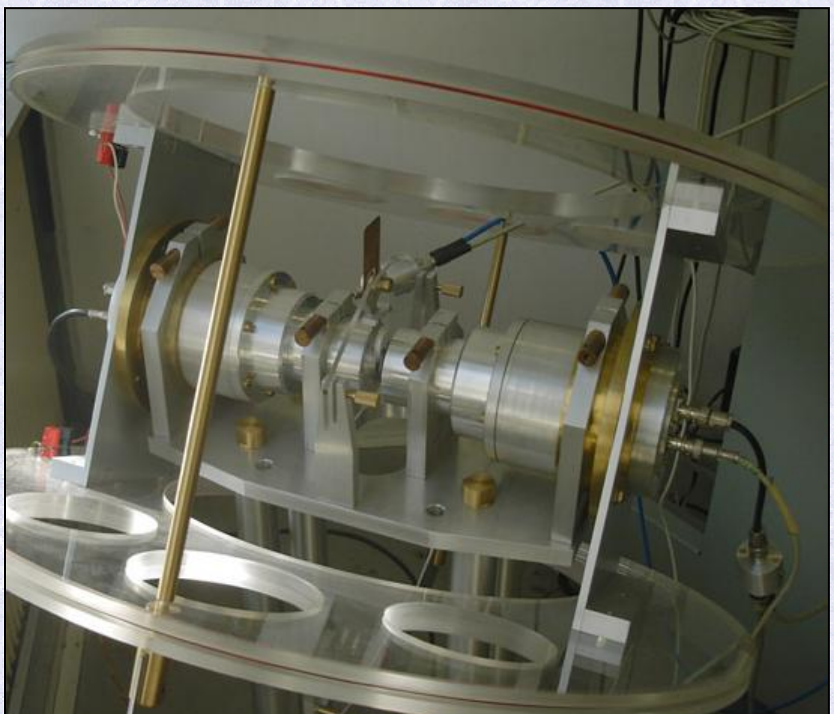


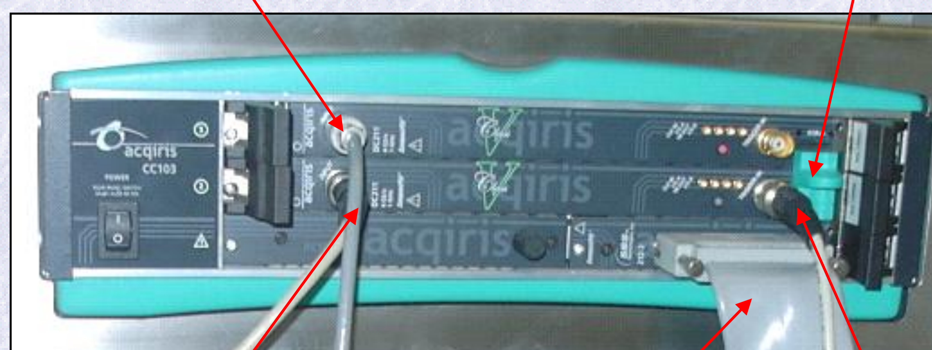
R.Z. Valiev, Nature Materials 3, 511-516 (2004)

LT spectroscopy

Digital LT spectrometer: *F. Bečvář et al., Nucl. Instr. Meth. A* ***539****, 372 (2005)*

- Two photomultipliers Hamamatsu H3378 & BaF_2 scintillators
- two 8-bit digitizers Acqiris DC211, sampling rate 4 GHz
- time resolution 145 ps (FWHM ^{22}Na)
- at least 10^7 annihilation events collected in each LT spectrum
- 1 MBq ^{22}Na source deposited on 2 μm Mylar foil
- source contribution determined using well annealed Fe: $\tau_{1s} \approx$ 368 ps, $I_{1s} \approx$ 9 %
  $\tau_{2s} \approx$ 1.5 ns, $I_{2s} \approx$ 1 %





X-ray diffraction

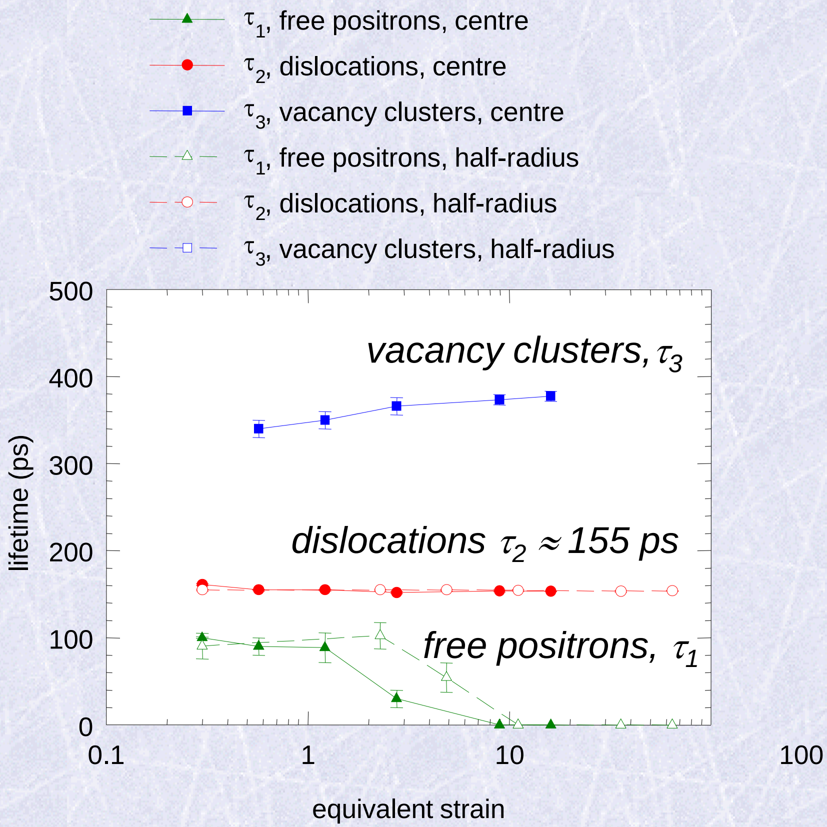
XRD diffractometer:

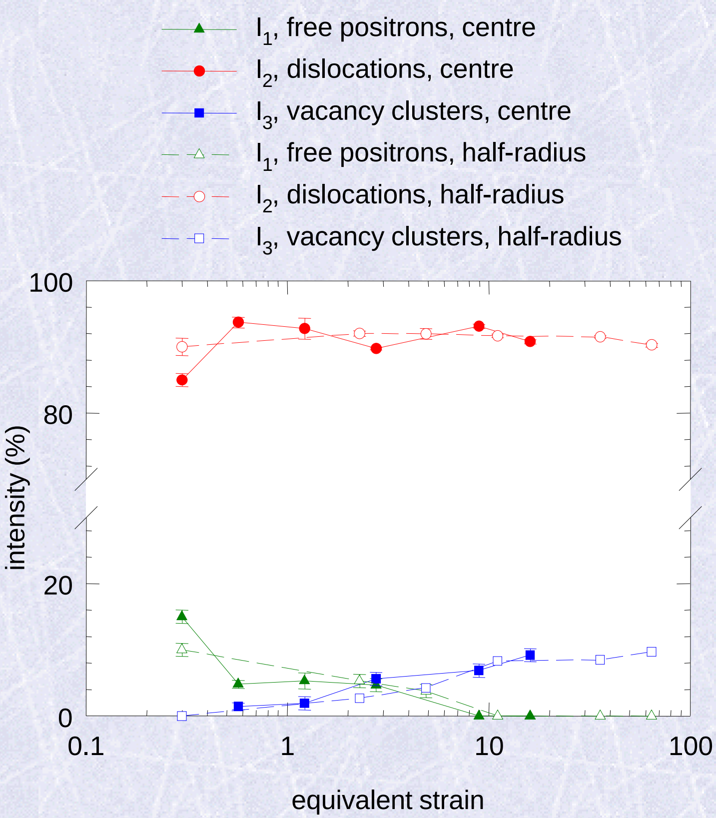
- Double crystal diffractometer with negligible instrumental broadening
- Ge monochromator
- Co $K_{\alpha 1}$ radiation (λ =0.1789 nm)
- size of X-ray beam spot: 2×0.2 mm^2
- linear position sensitive detector

Results of positron lifetime spectroscopy

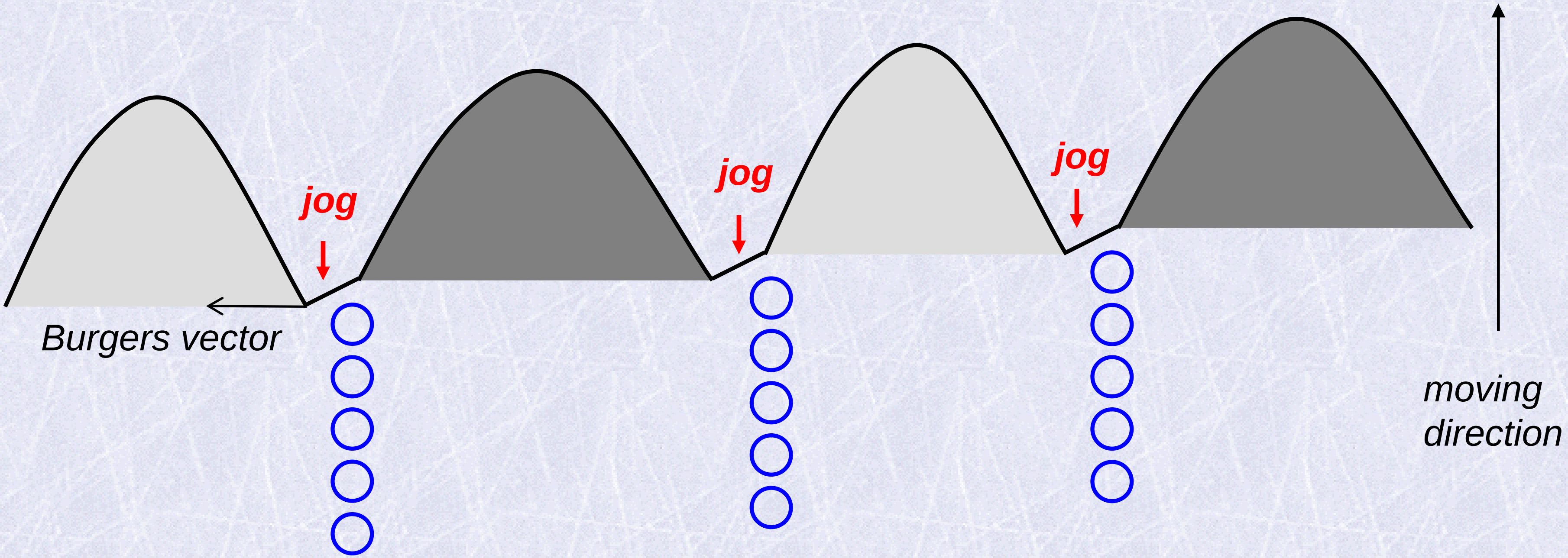
HPT deformed IF steels

- Decomposition of LT spectra into independent exponential components





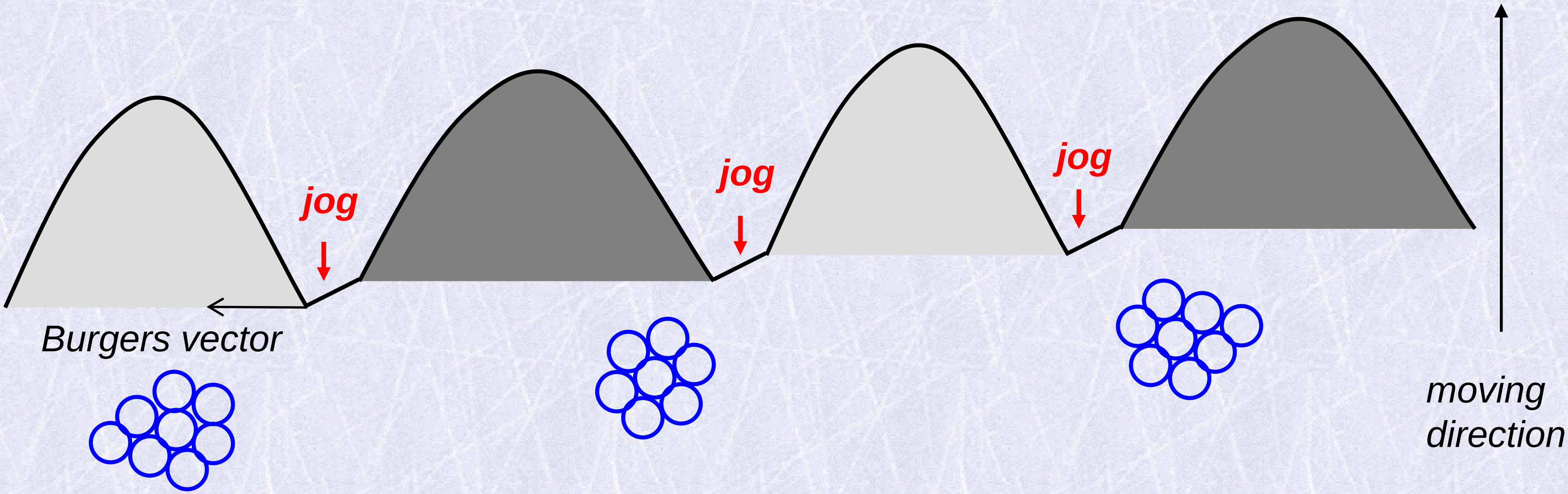
Generation of vacacies during HPT



• movement of dislocation with a jog

J. Čížek et al. J. Phys.: Conf. Series 443, 012008 (2013)

Generation of vacacies during SPD



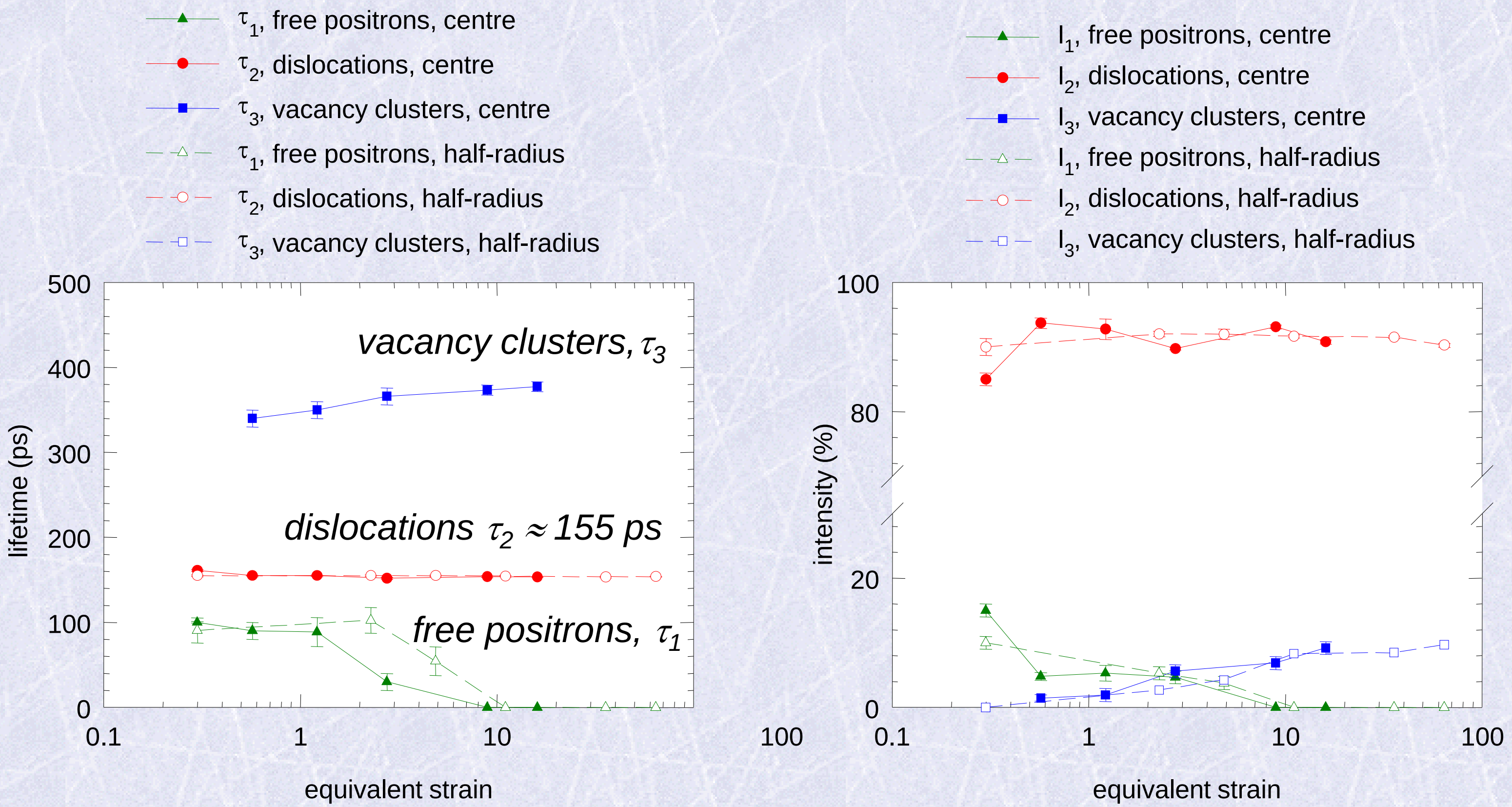
• vacancies agglomerate to small vacancy clusters

J. Čížek et al. J. Phys.: Conf. Series 443, 012008 (2013)

Results of positron lifetime spectroscopy

HPT deformed IF steels

- Decomposition of LT spectra into independent exponential components

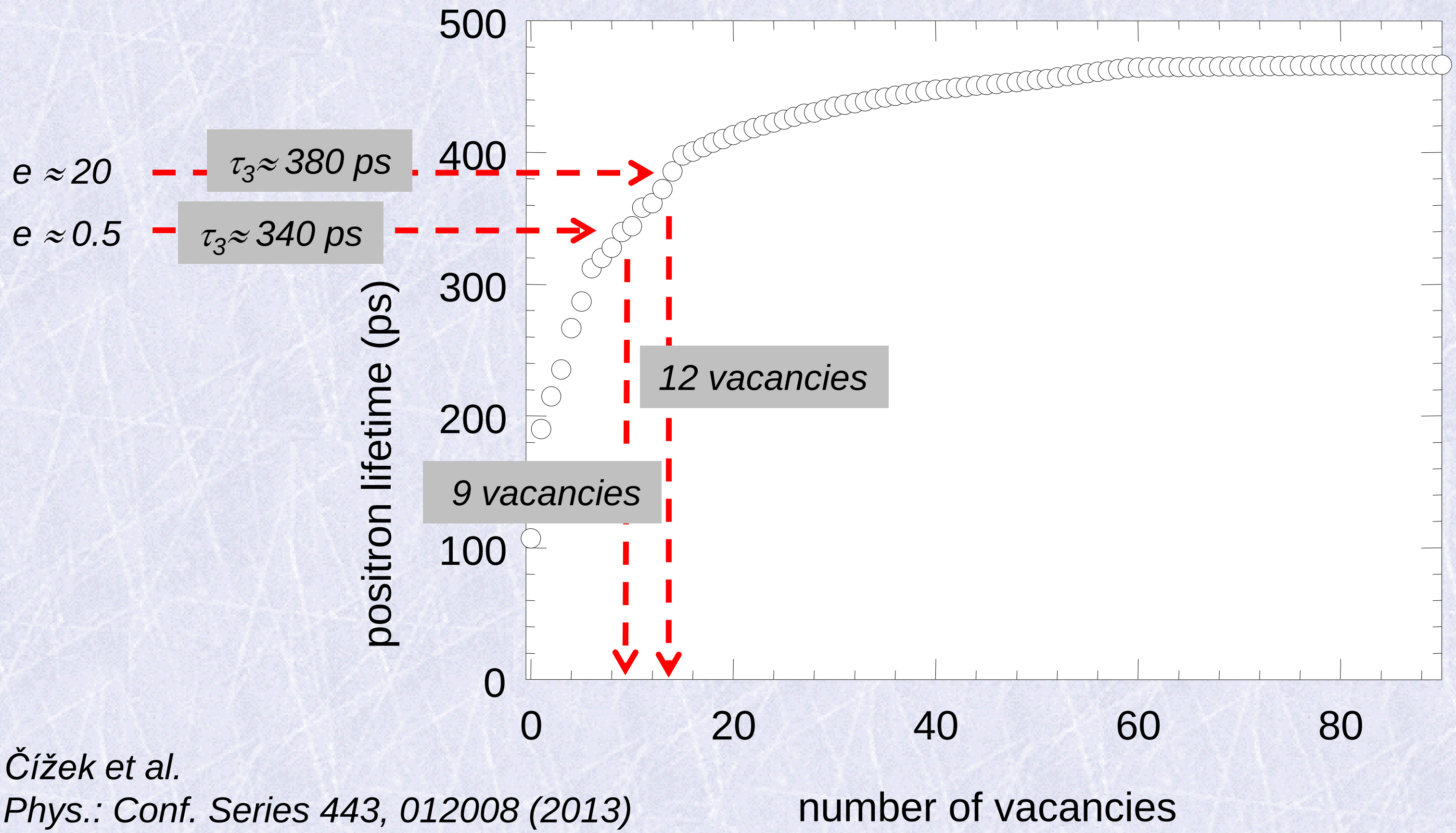


Results of positron lifetime spectroscopy

Ab-initio theoretical calculations

- bcc Fe

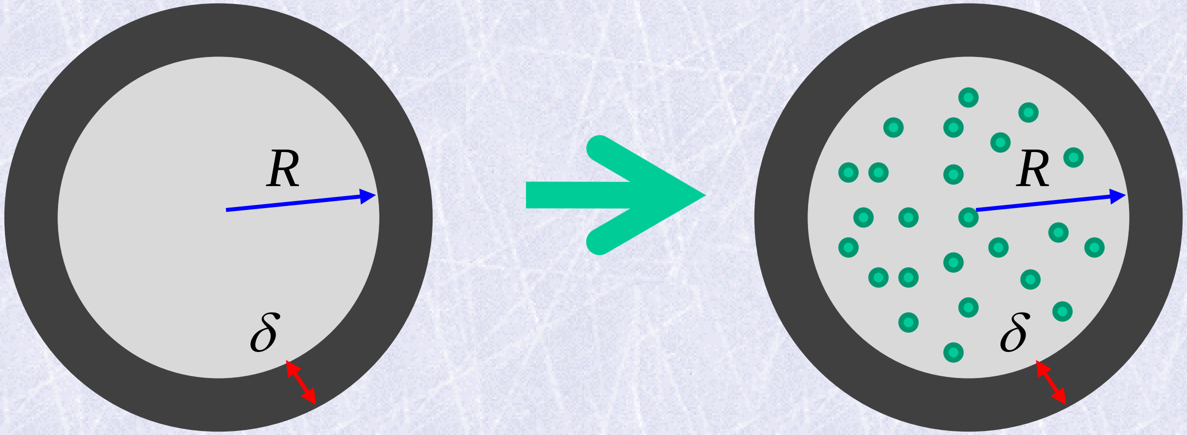
- dependence of positron lifetime on the size of vacancy clusters



J. Čížek et al.
J. Phys.: Conf. Series 443, 012008 (2013)

Diffusion trapping model with vacancy clusters

- cells contain uniformly distributed vacancy clusters



- positron lifetime spectrum

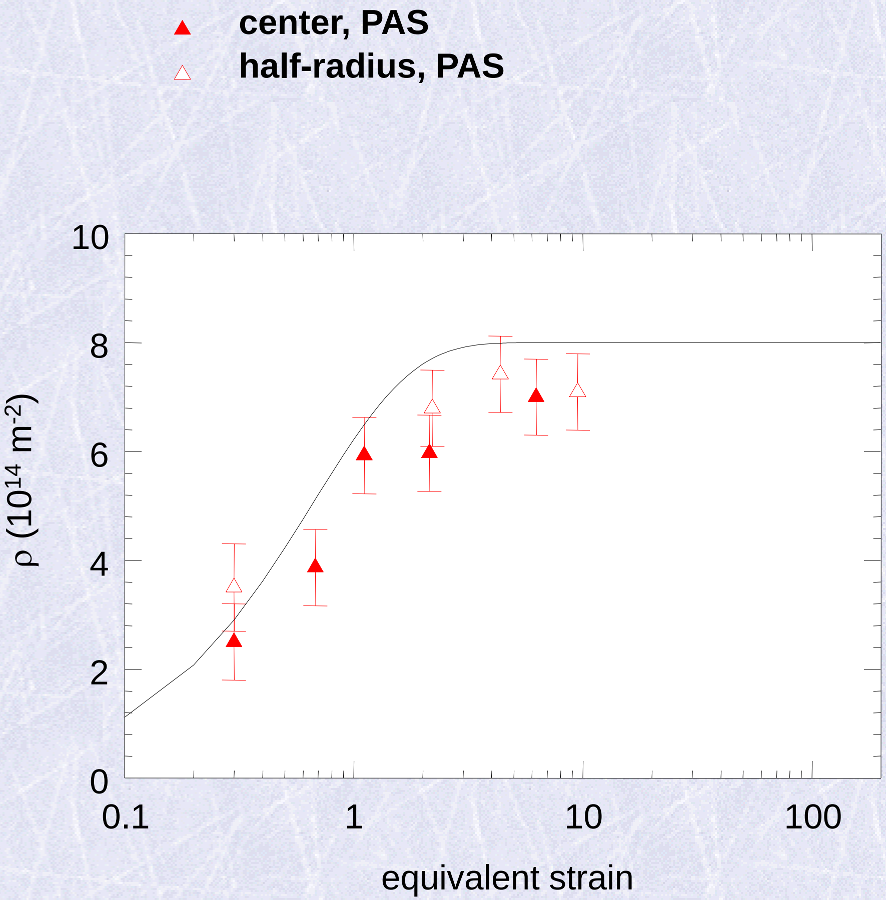
$$S(t) = \sum_k^{\infty} t_k^{-1} i_k e^{-t/t_k} + \tau_d^{-1} I_d e^{-t/\tau_d} + \tau_{cl}^{-1} I_{cl} e^{-t/\tau_{cl}}$$

positrons trapped at vacancy clusters

J. Čížek et al. PRB 65, 094106 (2002)

Dislocation density

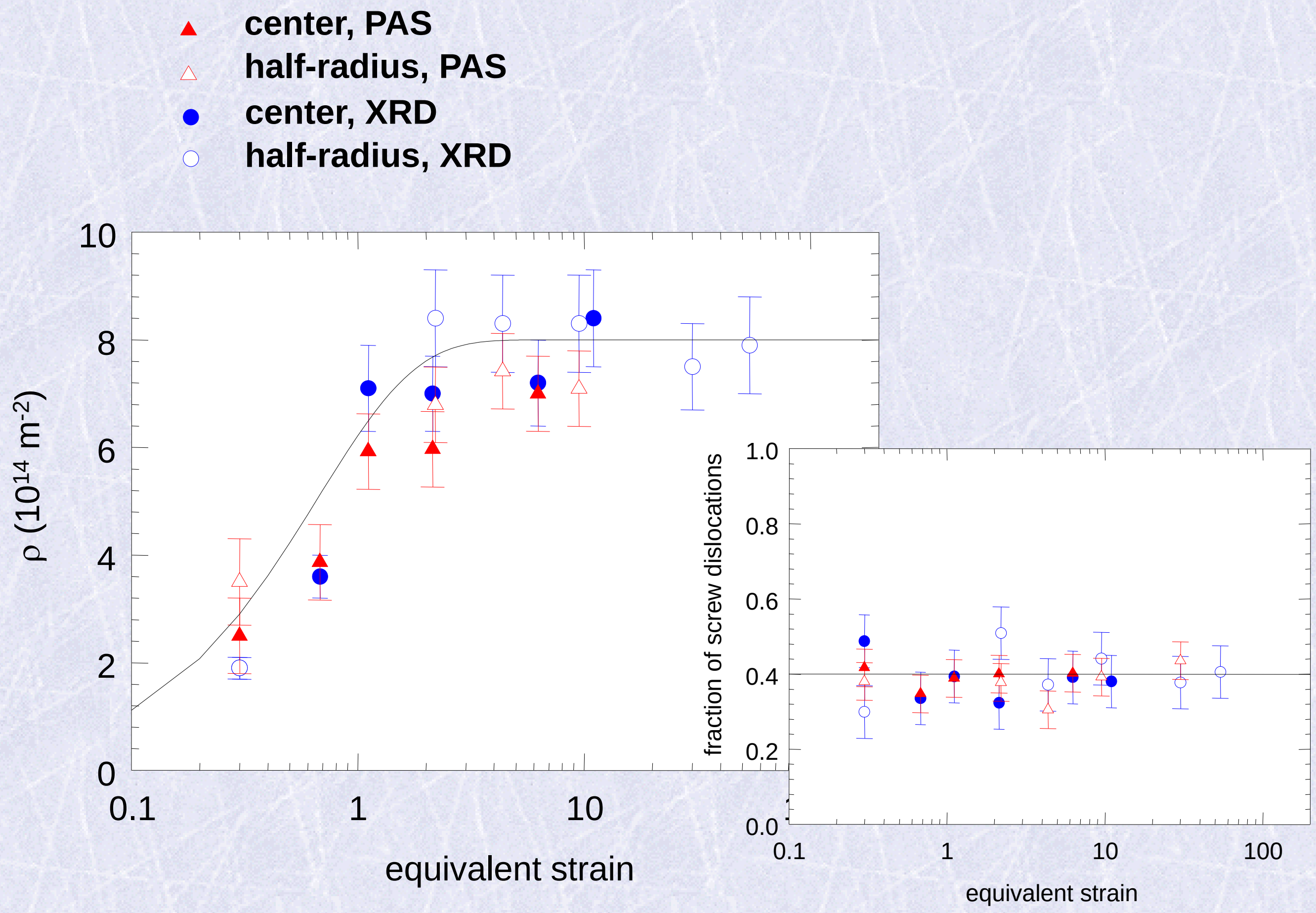
HPT deformed IF steel



Dislocation density

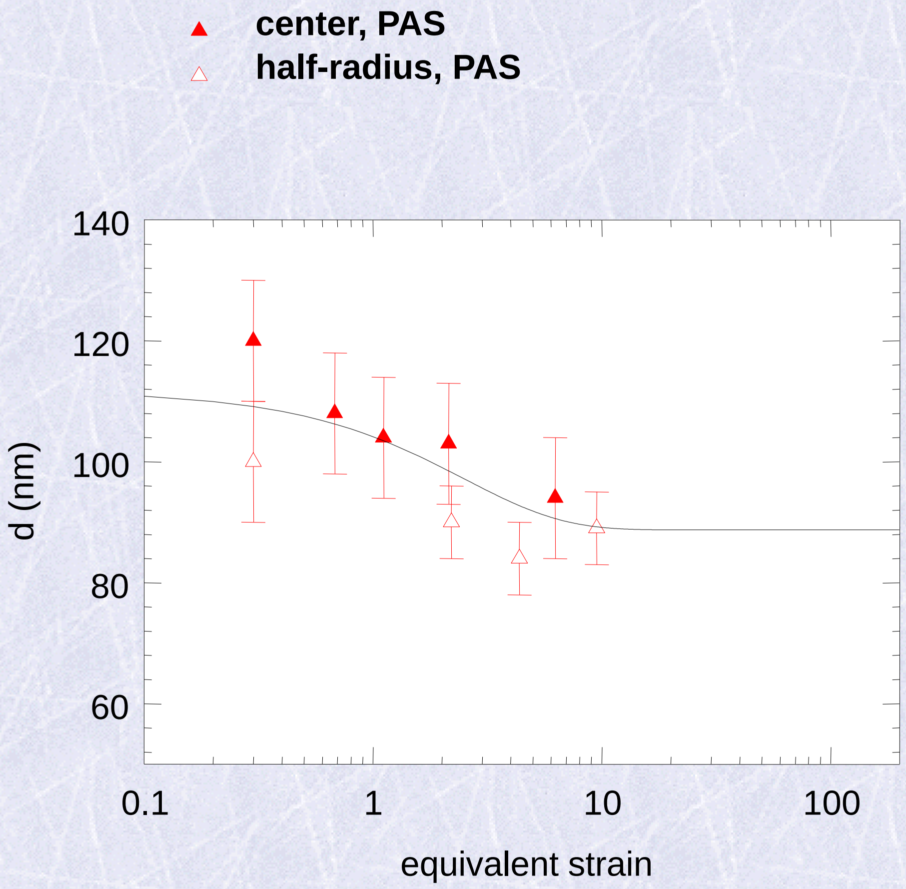
HPT deformed IF steel

- good agreement between PAS and XRD
- dislocation density increases with strain and saturates at $e \geq 3$
- edge dislocations prevail



Dislocation cell size

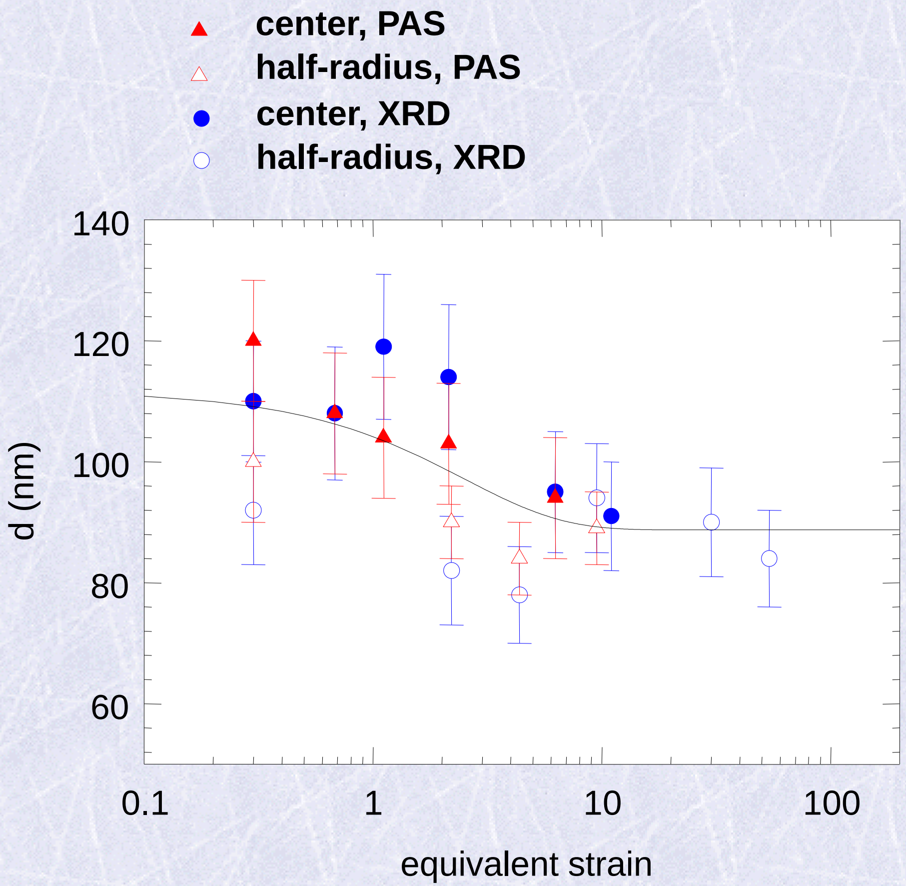
HPT deformed IF steel



Dislocation cell size

HPT deformed IF steel

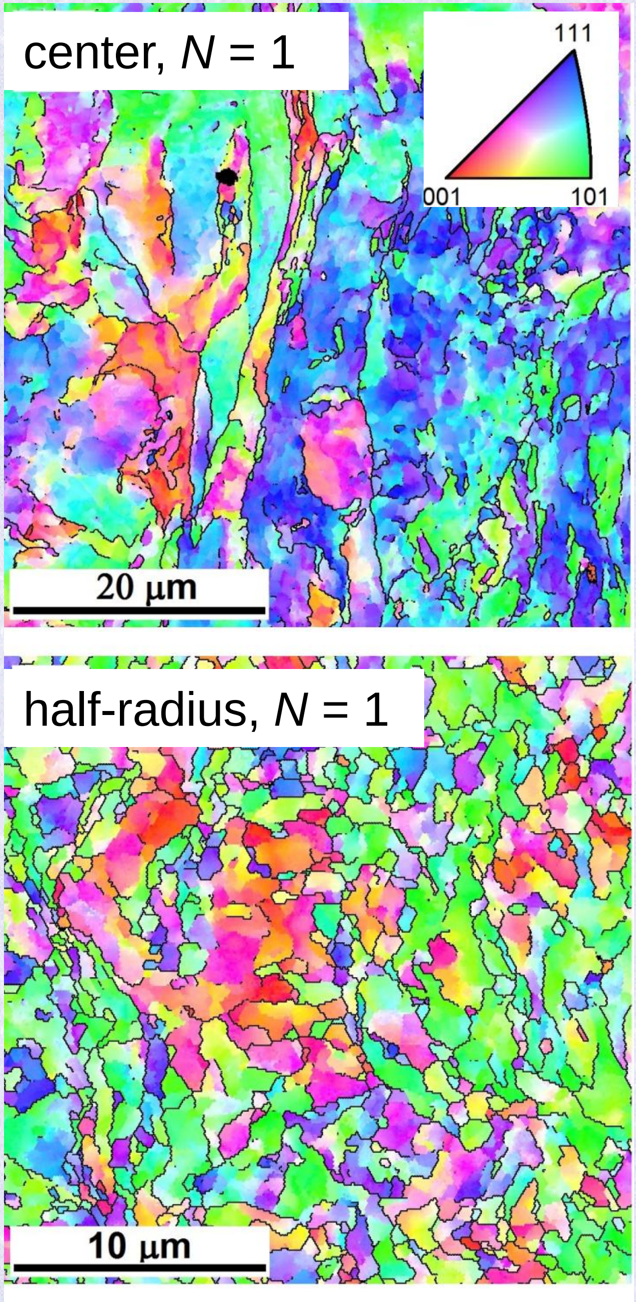
- cell size is close to 100 nm already at N = ¼
- saturation at $d \approx 90$ nm
- good agreement between PAS and XRD



Electron backscatter diffraction (EBSD)

HPT deformed IF steel

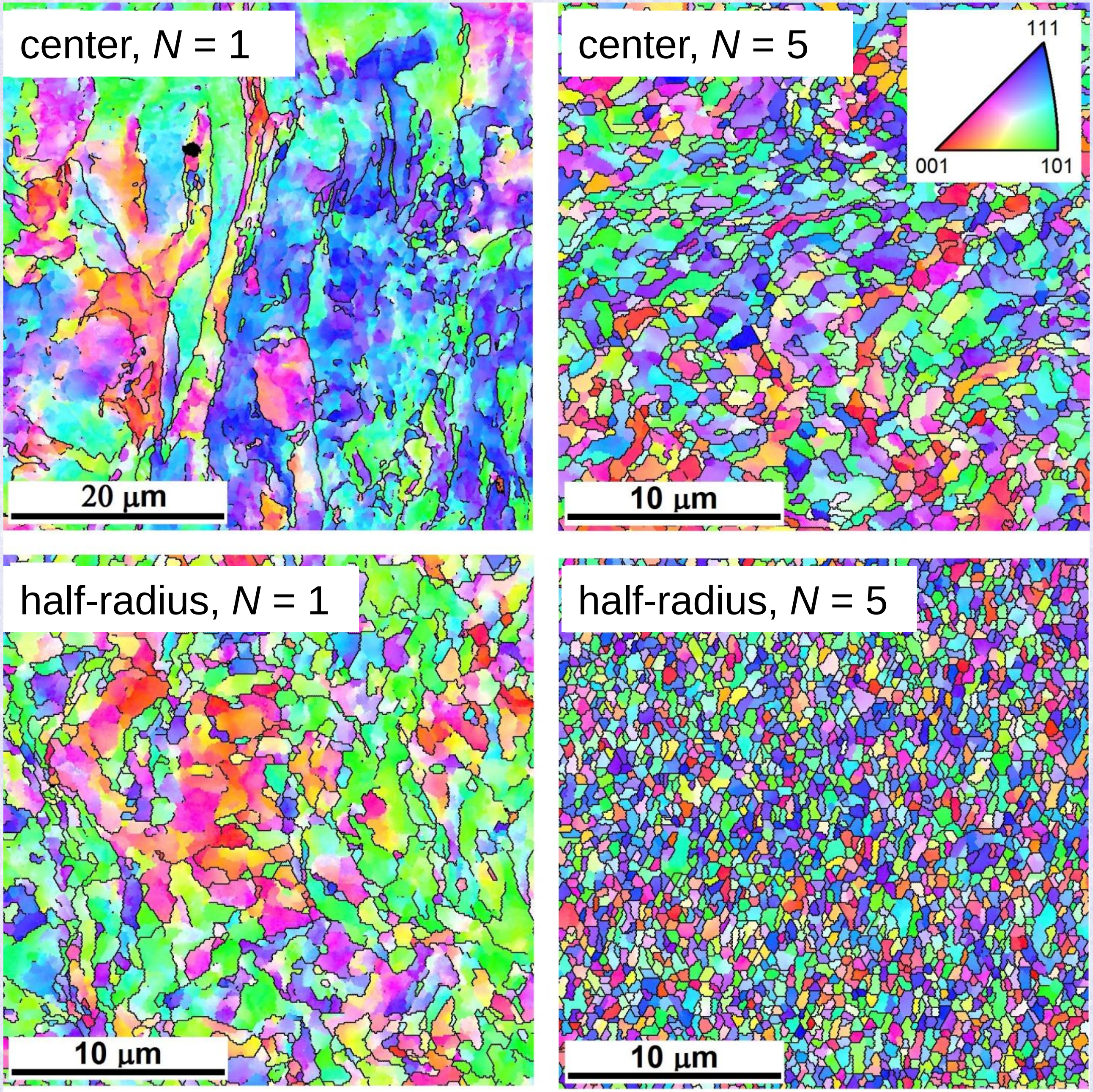
- early stage of HPT processing: bimodal structure



Electron backscatter diffraction (EBSD)

HPT deformed IF steel

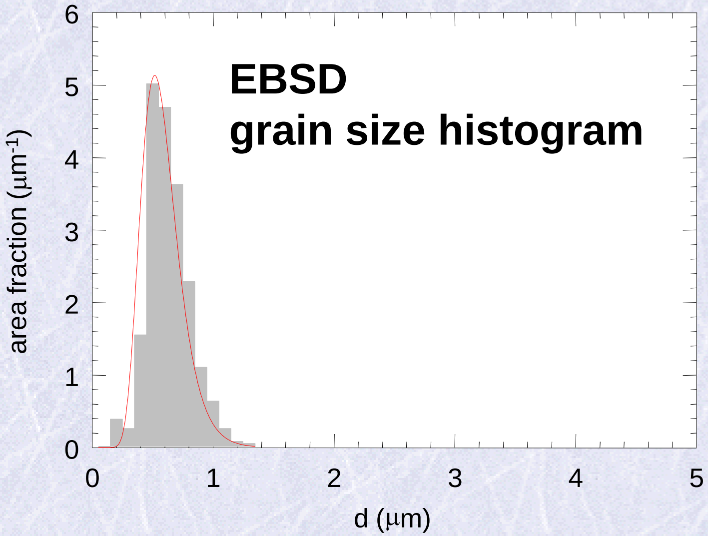
- early stage of HPT processing: bimodal structure
- N = 5: equiaxed fine grains
- grain refinement is faster at half-radius than in the centre

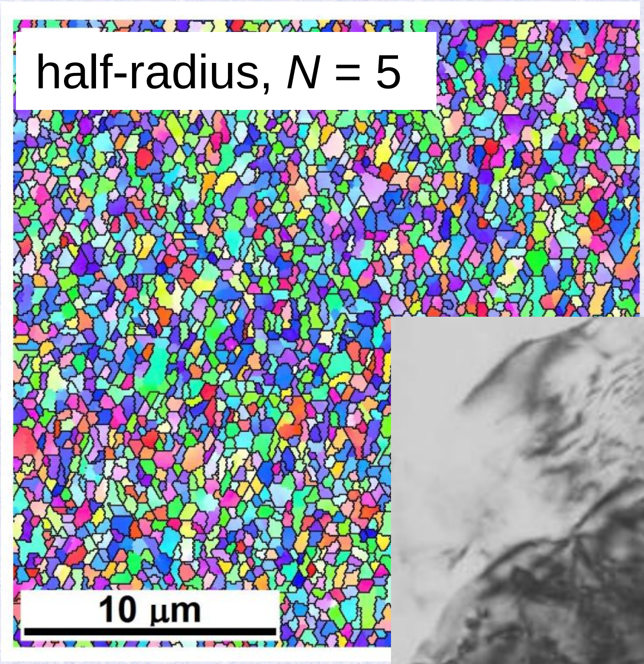


Electron backscatter diffraction (EBSD)

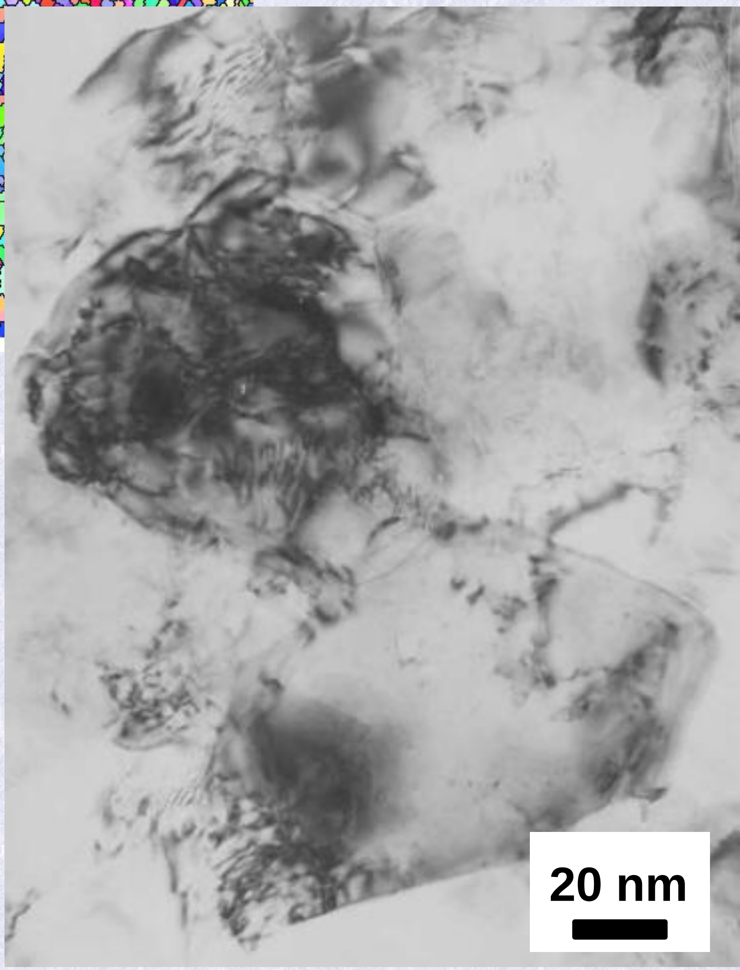
HPT deformed IF steel

- half radius, $N = 5$ ($e = 54$)
- mean grain size ≈ 600 nm

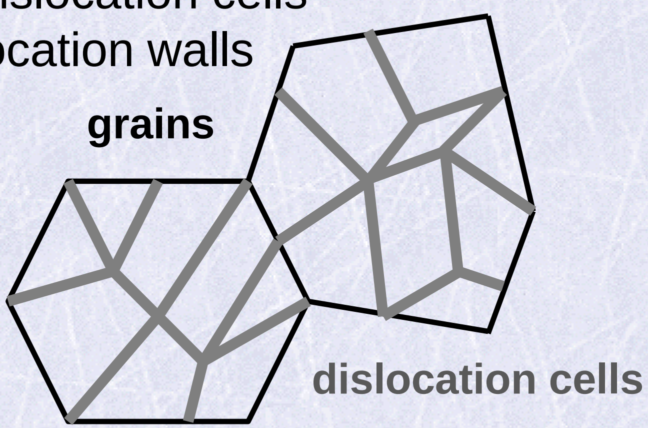




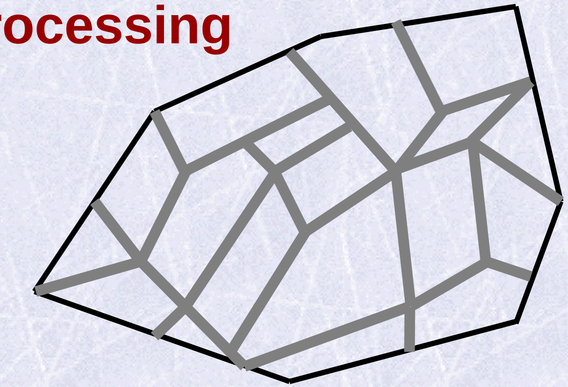
TEM bright field image



- grains consist of dislocation cells separated by dislocation walls

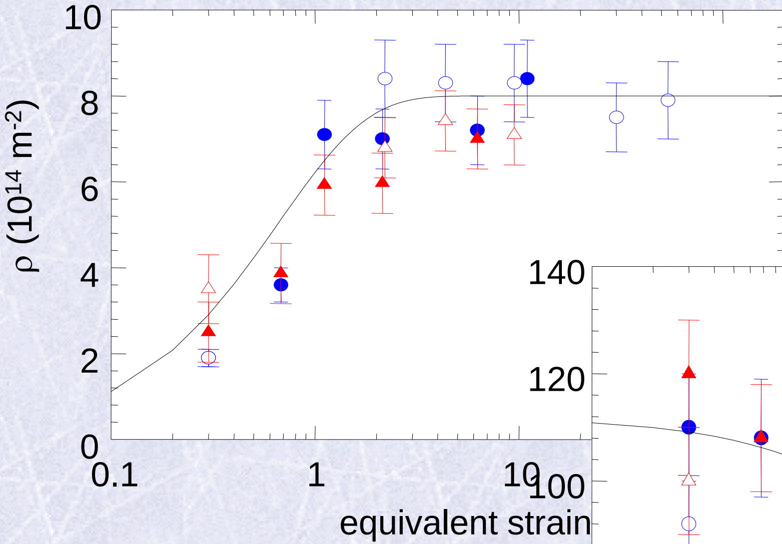


Development of mictrostructure during HPT processing

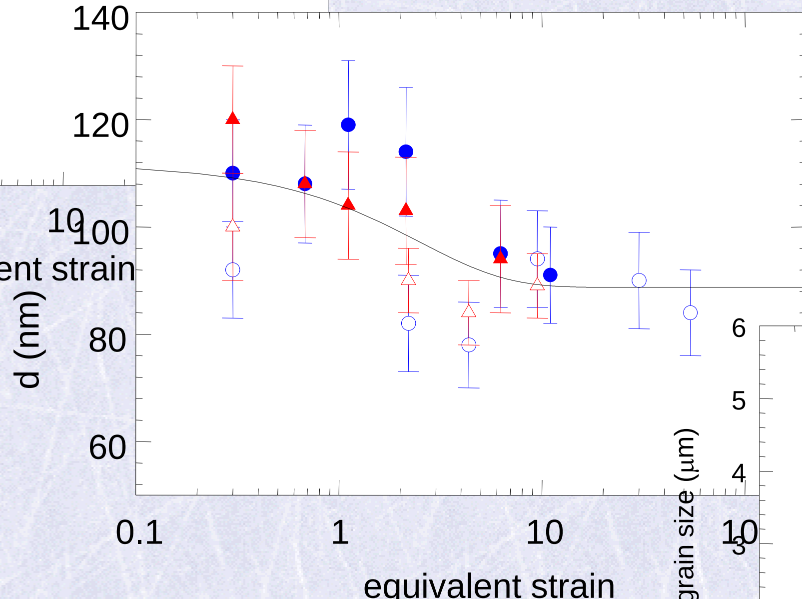


HPT deformed IF steel

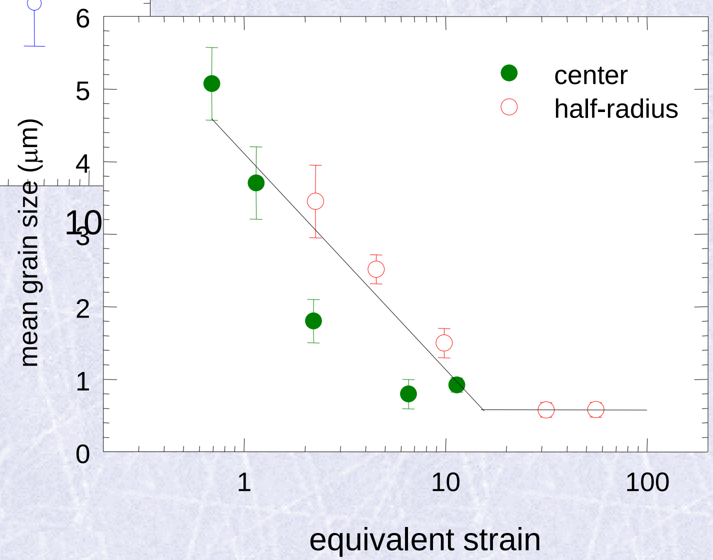
dislocation density saturated at $e \approx 3$



cell size saturated at $e \approx 8$

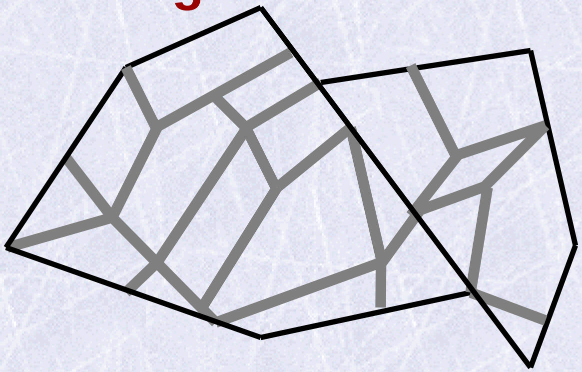


grain size saturated at $e \approx 15$

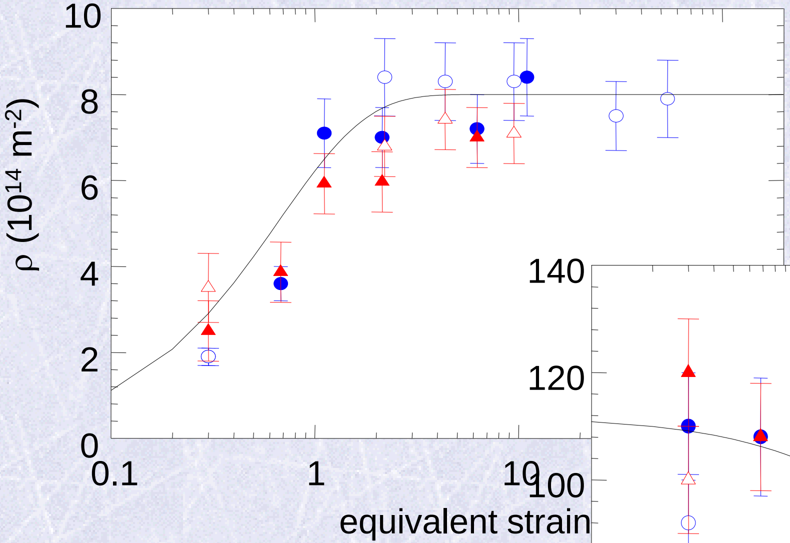


Development of mictrostructure during HPT processing

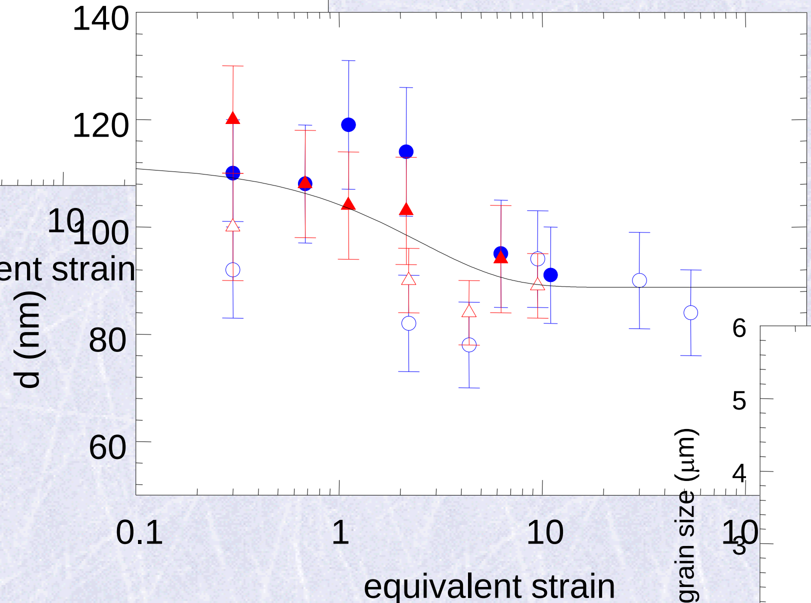
HPT deformed IF steel



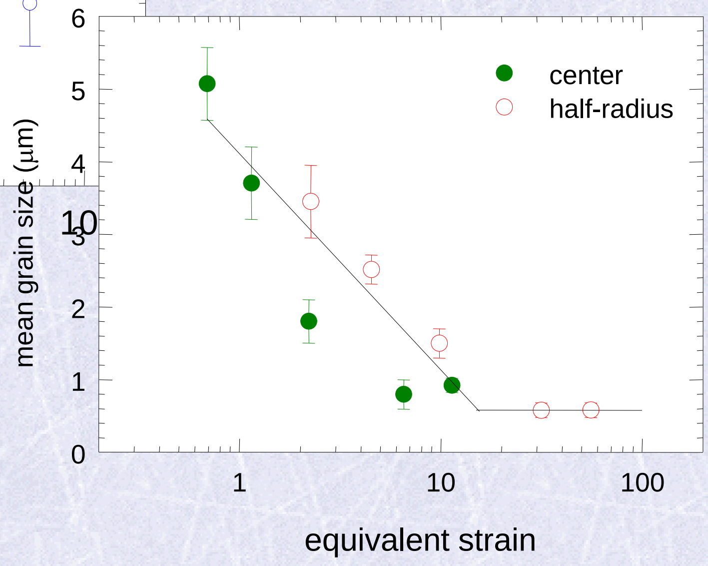
dislocation density saturated at $e \approx 3$



cell size saturated at $e \approx 8$



grain size saturated at $e \approx 15$



Conclusions

- Positron lifetime spectroscopy enables to determine
  - size of dislocation cells
  - mean dislocation density
  - edge or screw character of dislocations
- PAS results are consistent with results of X-ray line profile analysis
- use of PAS combined with XRD is beneficial for characterization of UFG materials
- PAS is more sensitive to dislocations than XRD
- XRD enables to determine ρ in materials containing very high number of dislocations
- PAS provides information not only about dislocations but also about open volume point defects (vacancies, vacancy clusters …)