Defects in solids

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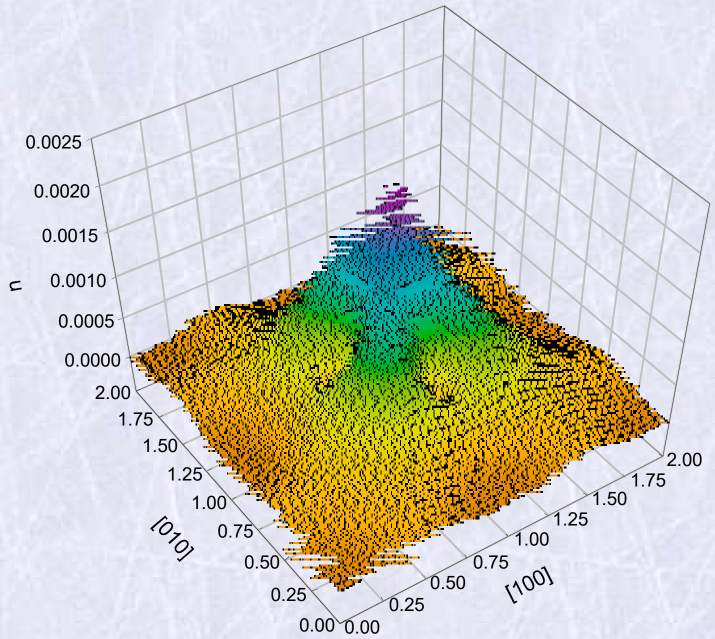
Outline

- lattice defects
- PAS experiment
- trapping model
- vacancies
- vacancy clusters
- dislocations



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Characterization of lattice defects in metallic materials by positron annihilation spectroscopy: A review

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A B S T R A C T

Positron is an excellent probe of lattice defects in solids. A thermallized positron delocalized in lattice can be trapped at open volume defects, e.g. vacancies, dislocations, grain boundaries etc. Positron annihilation spectroscopy is a non-destructive technique which enables characterization of open volume lattice defects in solids on the atomic scale. Positron lifetime and Doppler broadening of annihilation photo-peak are the most common observables related to positron annihilation process. Positron lifetime spectroscopy enables to identify defects in solids and to determine their concentrations while coincidence measurement of Doppler broadening provides information about local chemical environment of defects. This article provides a review of the state-of-art of defect characterization in bulk metallic materials by positron annihilation spectroscopy. Advanced analysis of positron annihilation data and recent developments of positron annihilation methodology are described and discussed on examples of defect studies of metallic materials. Future development in the field is proposed as well.

1. Introduction

Open volume defects of crystallite lattice, e.g. vacancies, dislocations, grain boundaries etc., have a strong influence on many physical properties of metallic materials. For example vacancies play the key role in diffusion processes [1] and phase transforma-

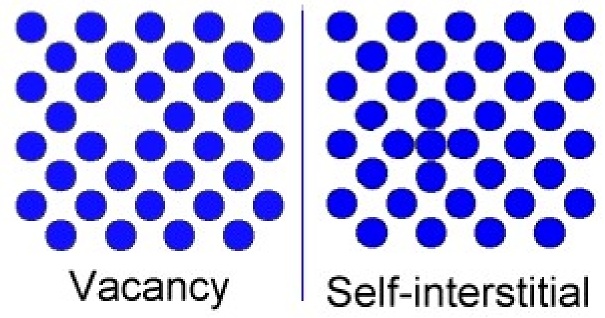
(ii) The design of hardenable Al-Si-Mg based (6xxx series) alloys by micro-alloying with elements having a high binding energy with vacancies [8–10]. This way one can control natural ageing of Al-Si-Mg alloys and also the strengthening effect during subsequent artificial ageing.

(iii) The enhancement of the equilibrium concentration of vacan-

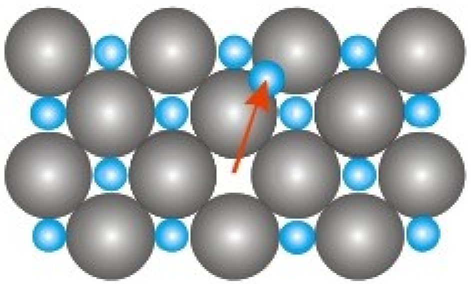
Lattice defects in solids

- defect = any irregularity of crystalline lattice
- point defects (e.g. vacancies)

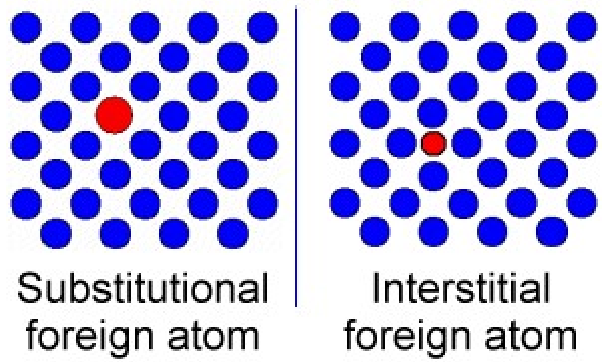
intrinsic point defects



Frenkel pair



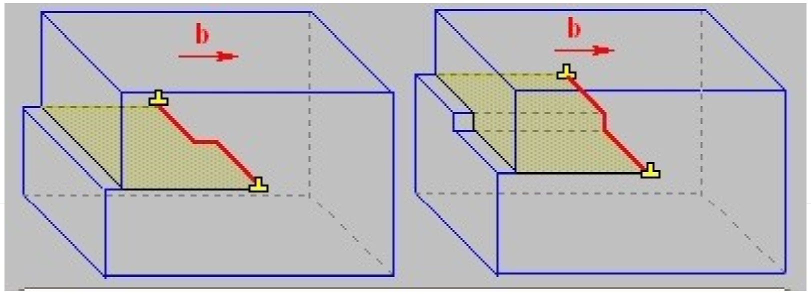
extrinsic point defects



Lattice defects in solids

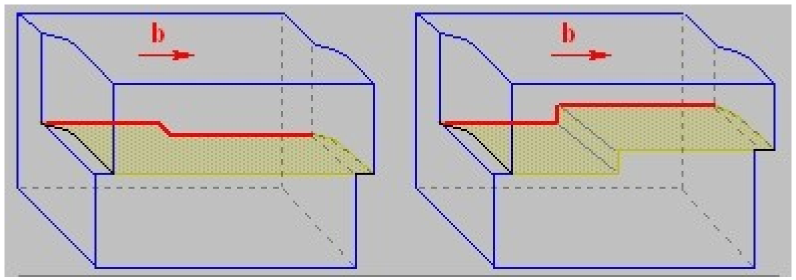
- defect = any irregularity of crystalline lattice
- line defects (e.g. dislocations)

edge dislocation

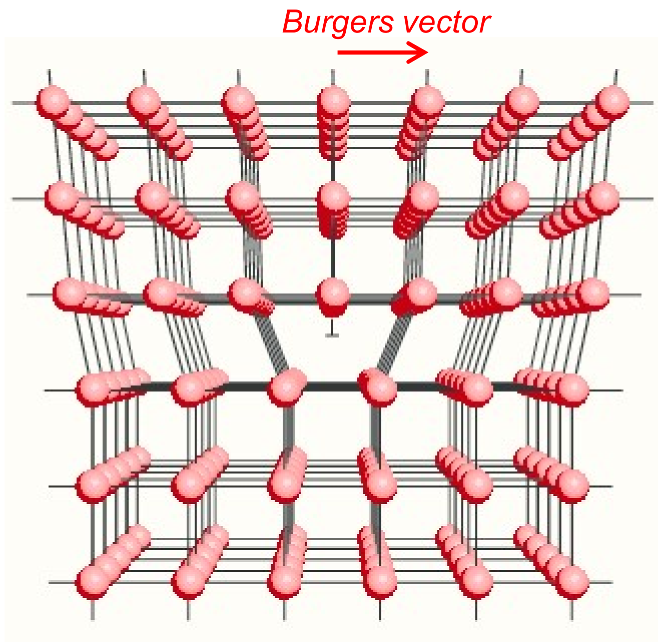


kink *jog*

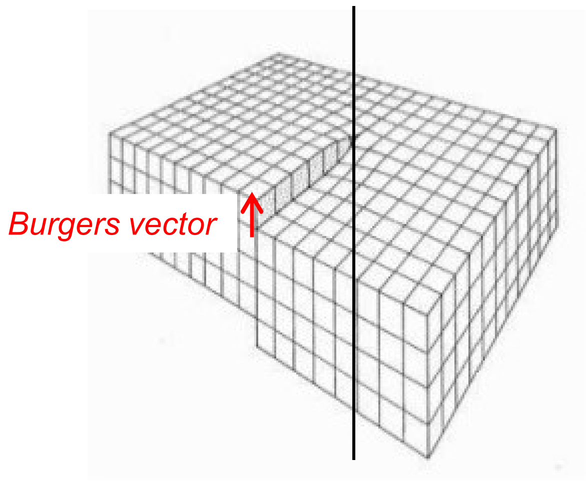
screw dislocation



kink *jog*



edge dislocation

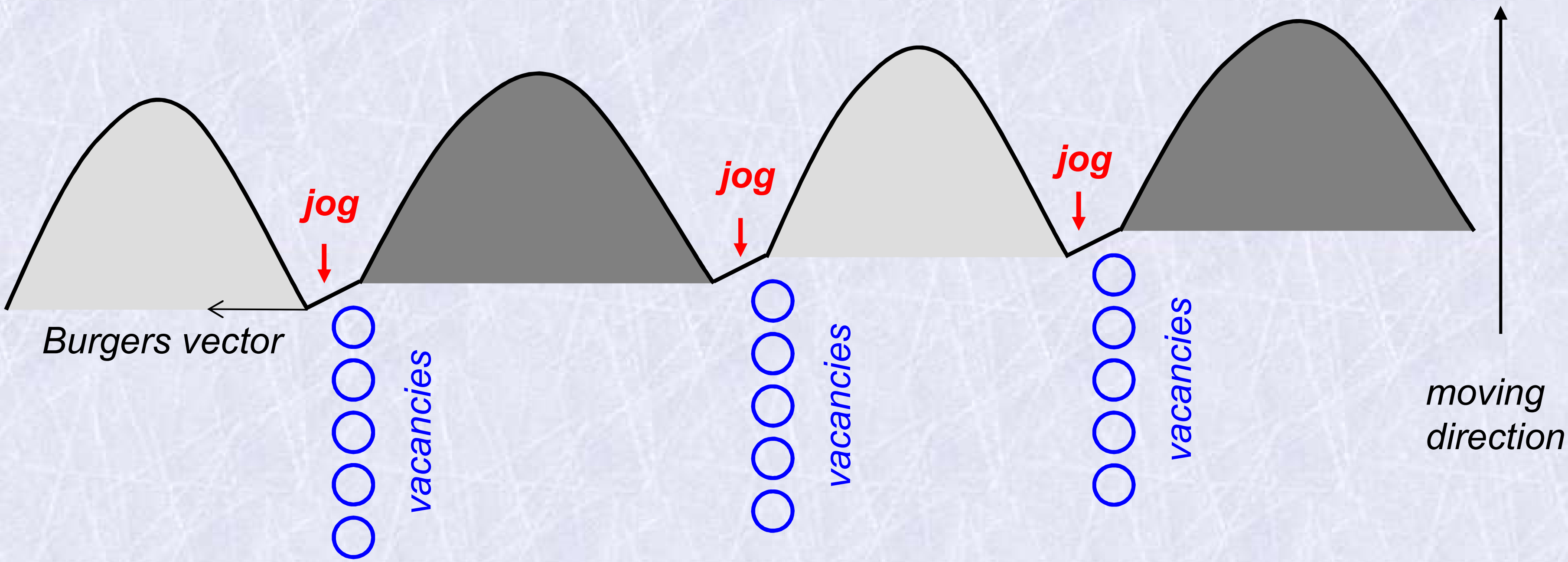


screw dislocation

Lattice defects in solids

• defect = any irregularity of crystalline lattice

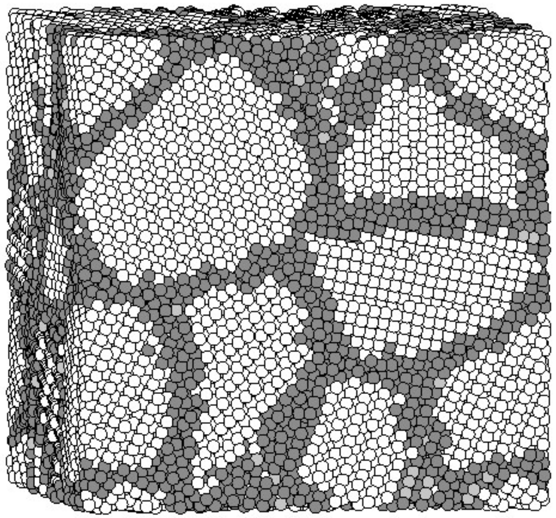
• line defects (e.g. dislocations)



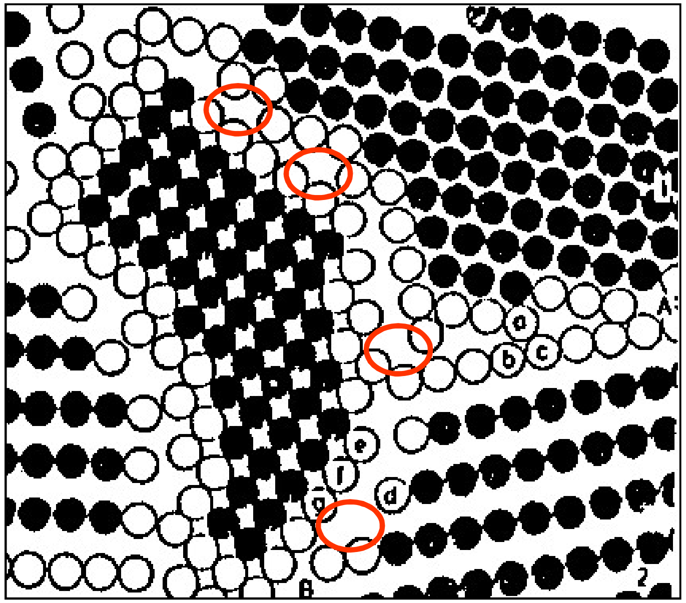
• slip of a screw dislocation with a jogs

Lattice defects in solids

- defect = any irregularity of crystalline lattice
- areal defects (e.g. grain boundaries)



grain boundaries

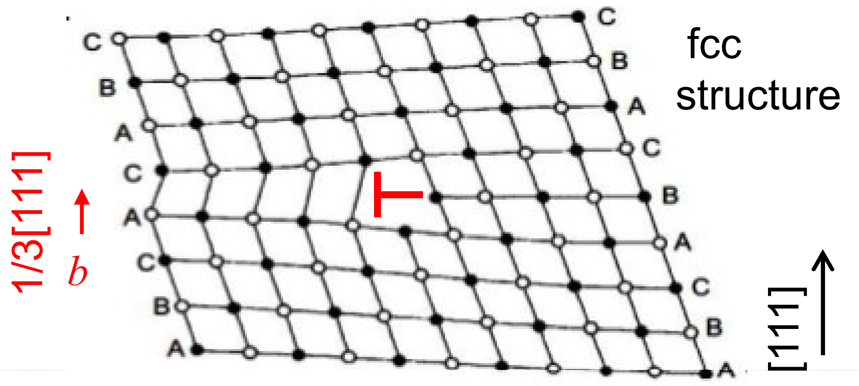


open volume misfit defects at grain boundaries

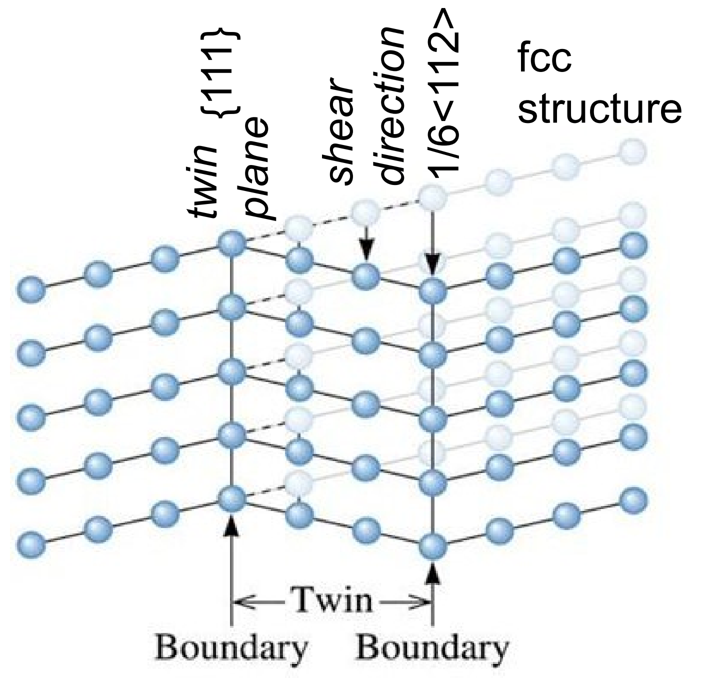
Lattice defects in solids

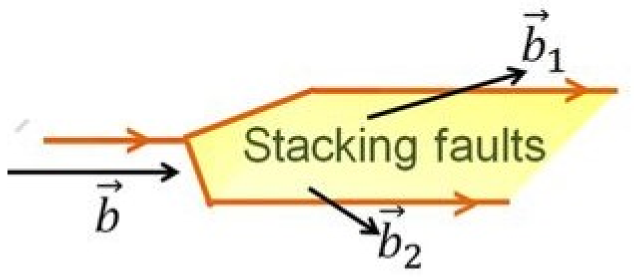
- defect = any irregularity of crystalline lattice

- areal defects (e.g. grain boundaries)

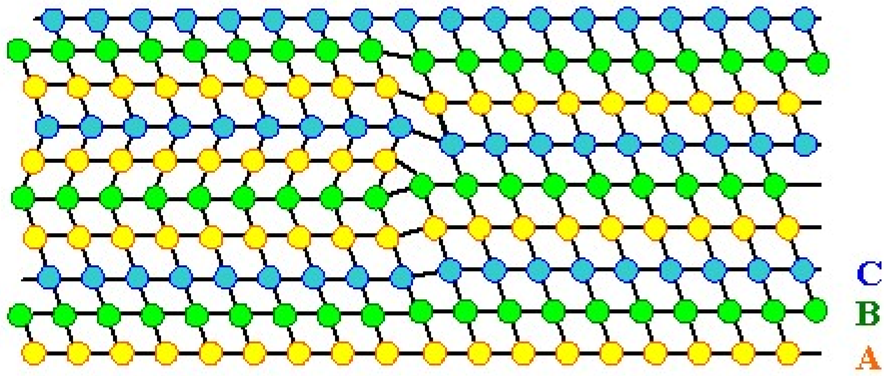


Frank partial dislocation and stacking fault





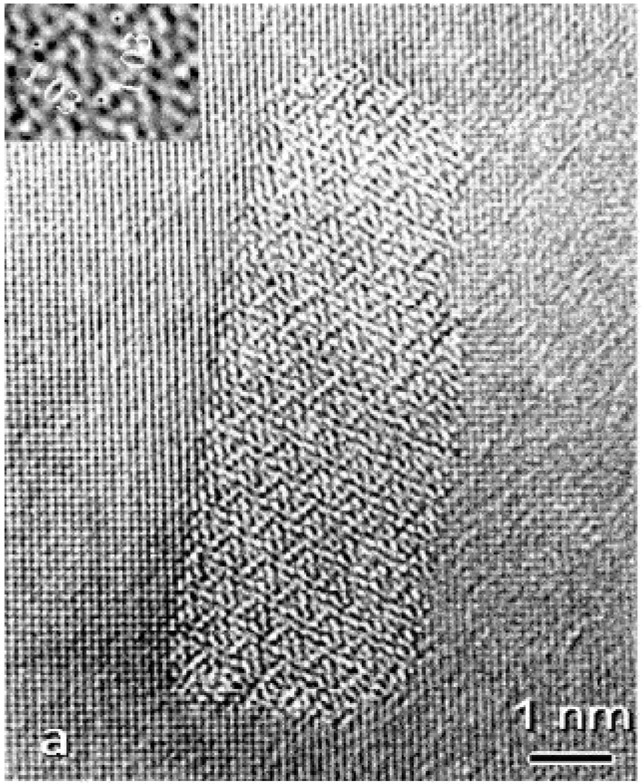
screw dislocation split into partials



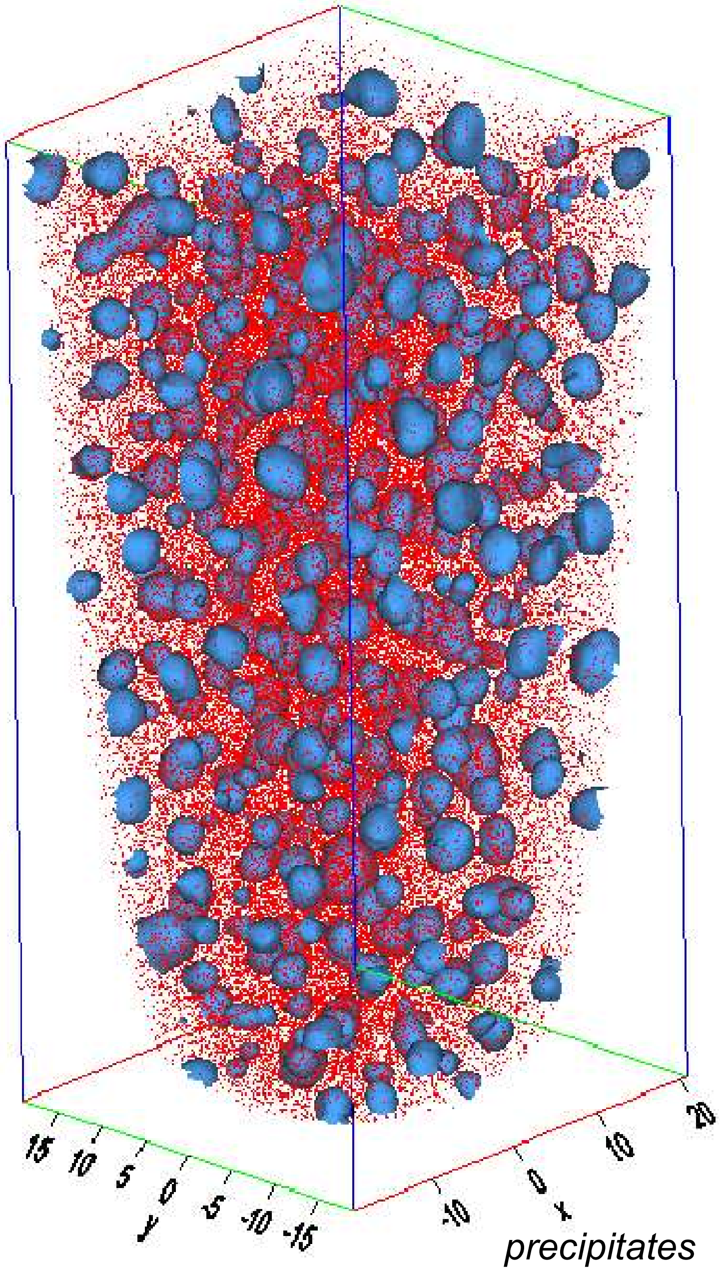
stacking fault

Lattice defects in solids

- defect = any irregularity of crystalline lattice
- volume defects (e.g. precipitates)



precipitate



precipitates

PAS techniques for defect studies

- **positron lifetime (LT) spectroscopy**
  - identification of type of defects
  - determination of defect concentrations
- **coincidence Doppler broadening (CDB)**
  - local chemical environment of defects
- **variable energy positron annihilation spectroscopy (VEPAS)**
  - defect depth profile
  - positron diffusion length

PAS techniques for defect studies

- **digital spectrometers**

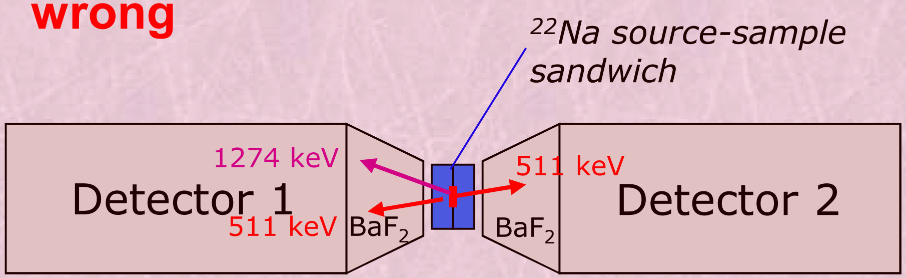
  - real time sampling of detector pulses and analysis by dedicated software

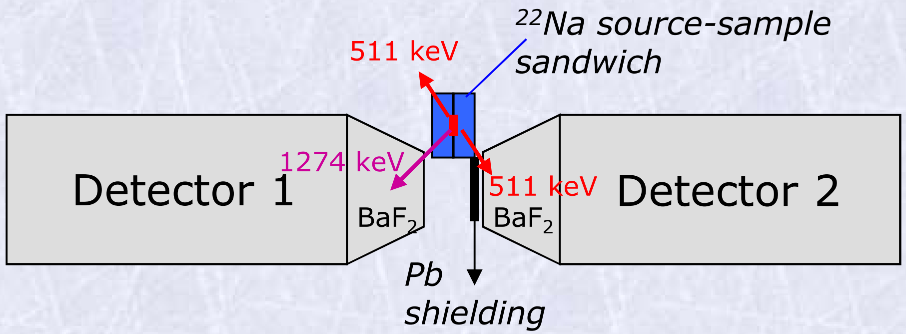
  - analysis can be repeated to find optimum strategy for determination of measured quantity

  - perfect control over shape of sampled waveforms $\rightarrow$ excellent clarity of spectrum

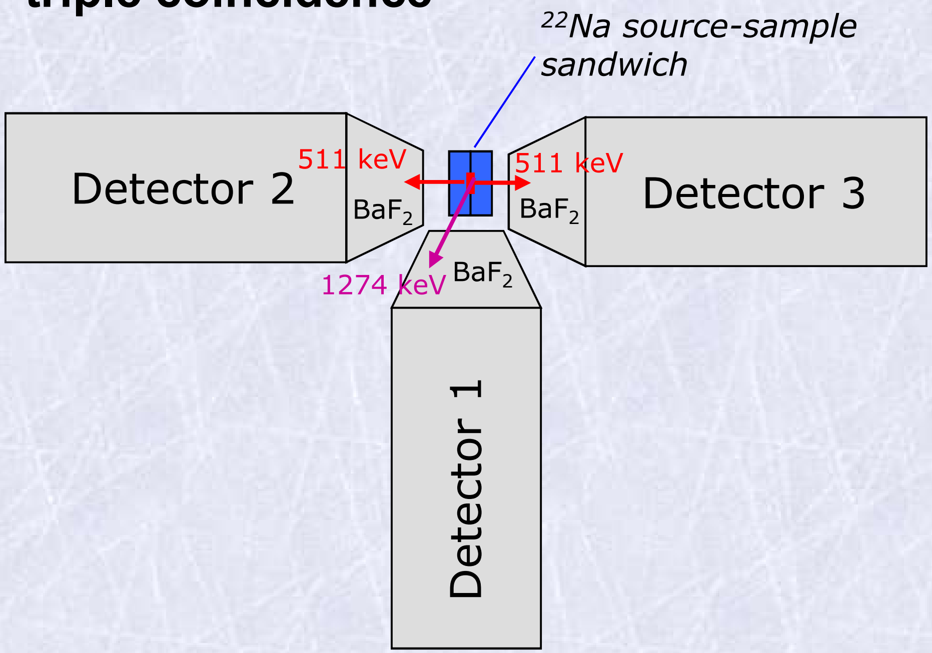
Geometry of positron lifetime spectrometer

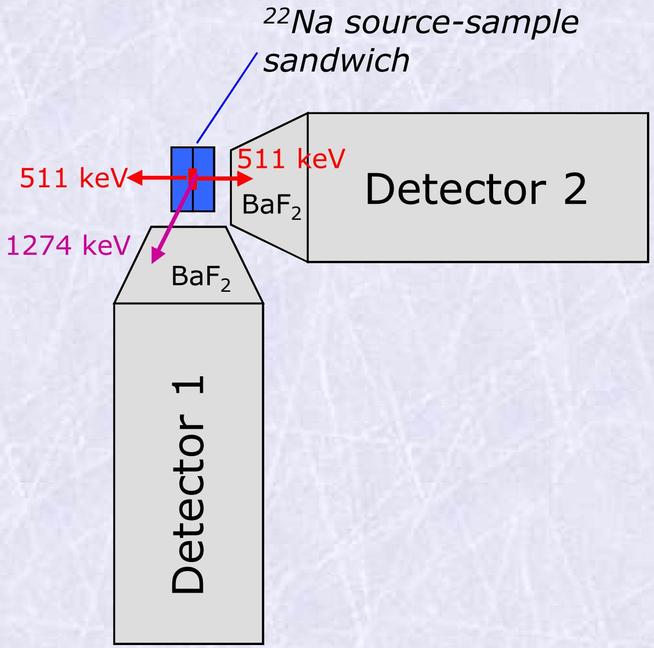
wrong



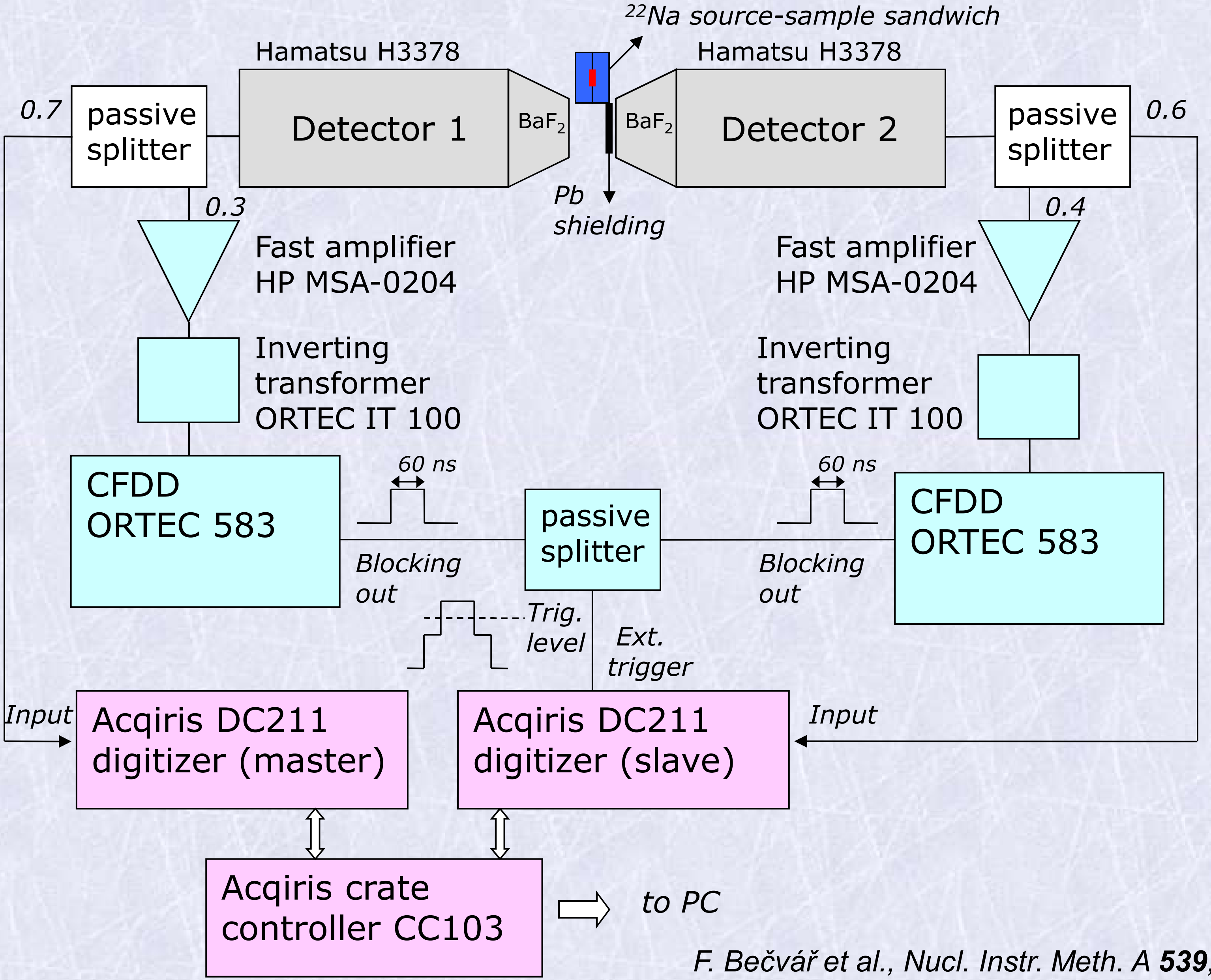


triple coincidence





Digital positron lifetime spectrometer



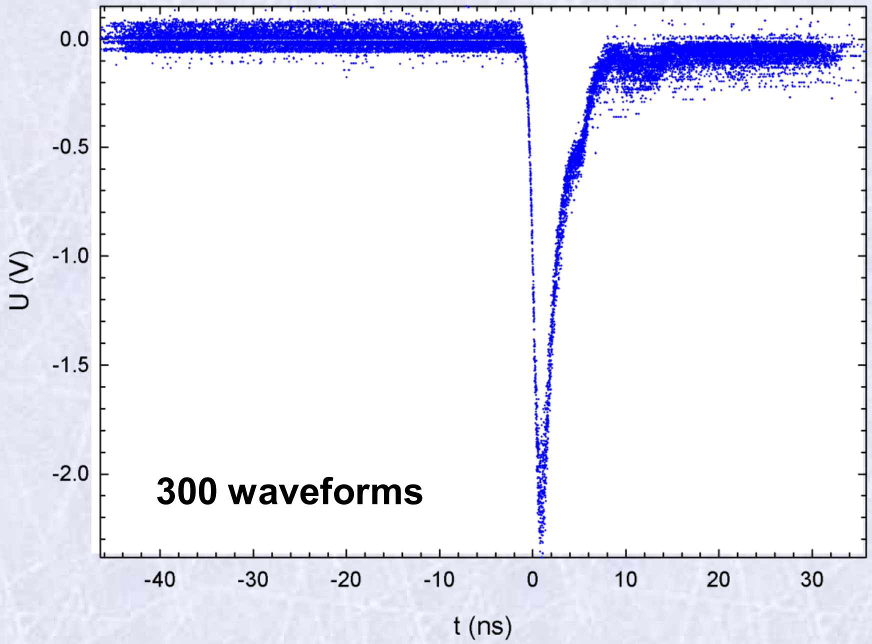
F. Bečvář et al., Nucl. Instr. Meth. A ***539****, 372 (2005)*

Digital positron lifetime spectrometer

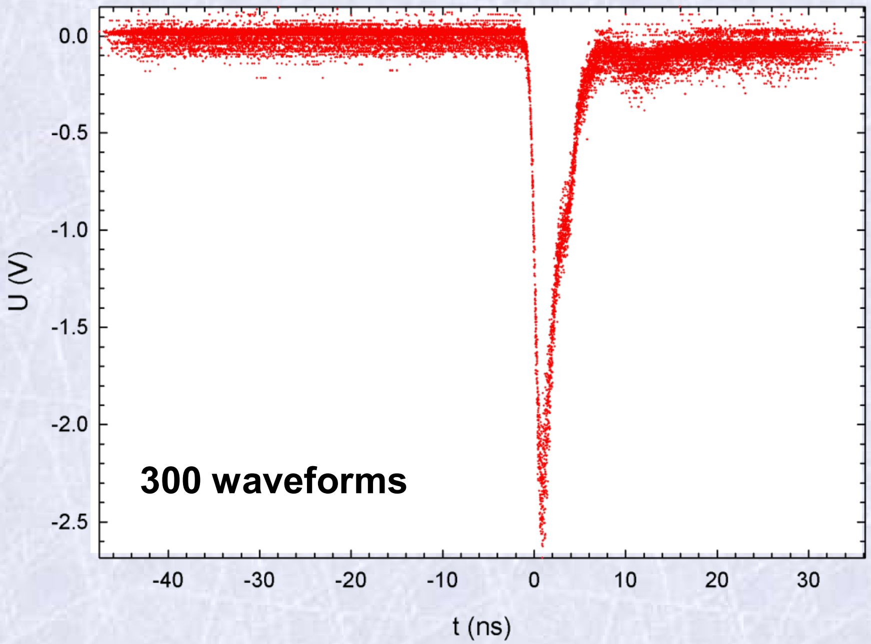
- **waveform** - single digitized detector pulse (300 points, 75 ns)

- normalized waveforms

detector 1



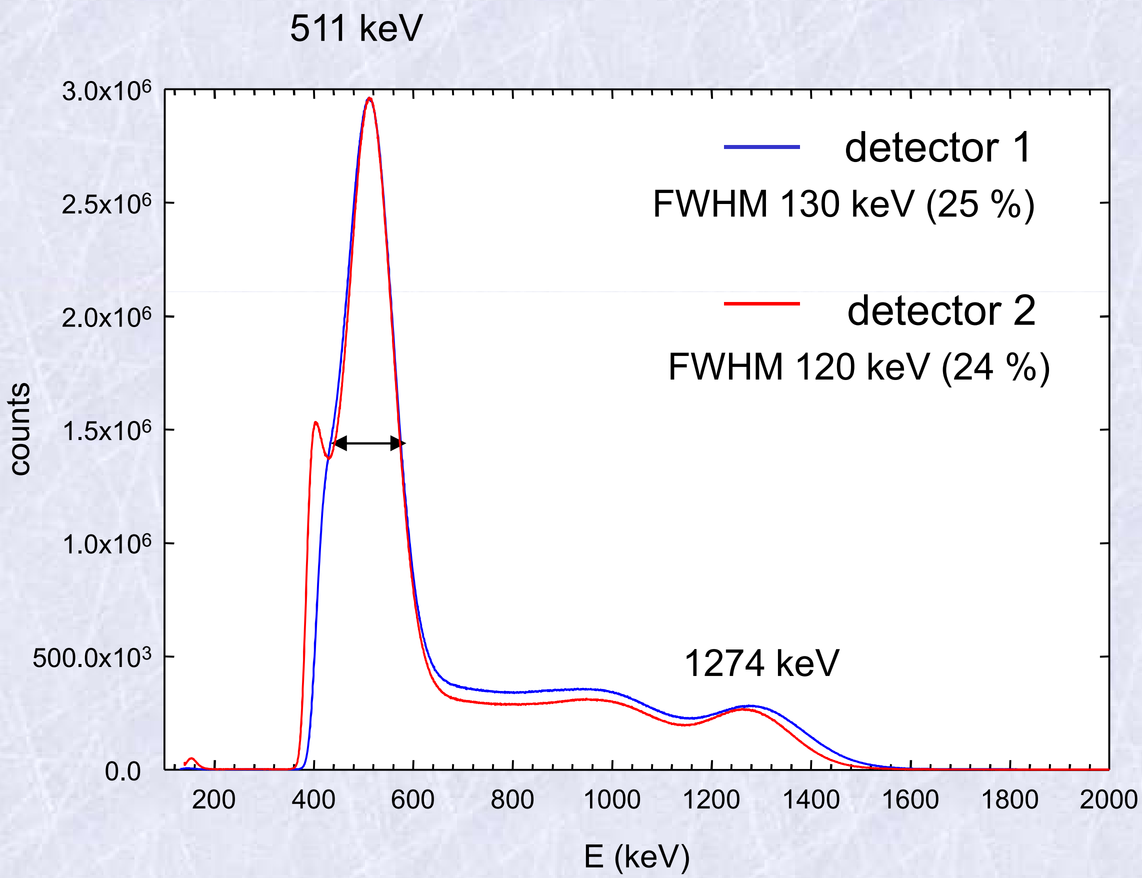
detector 2



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

Digital positron lifetime spectrometer

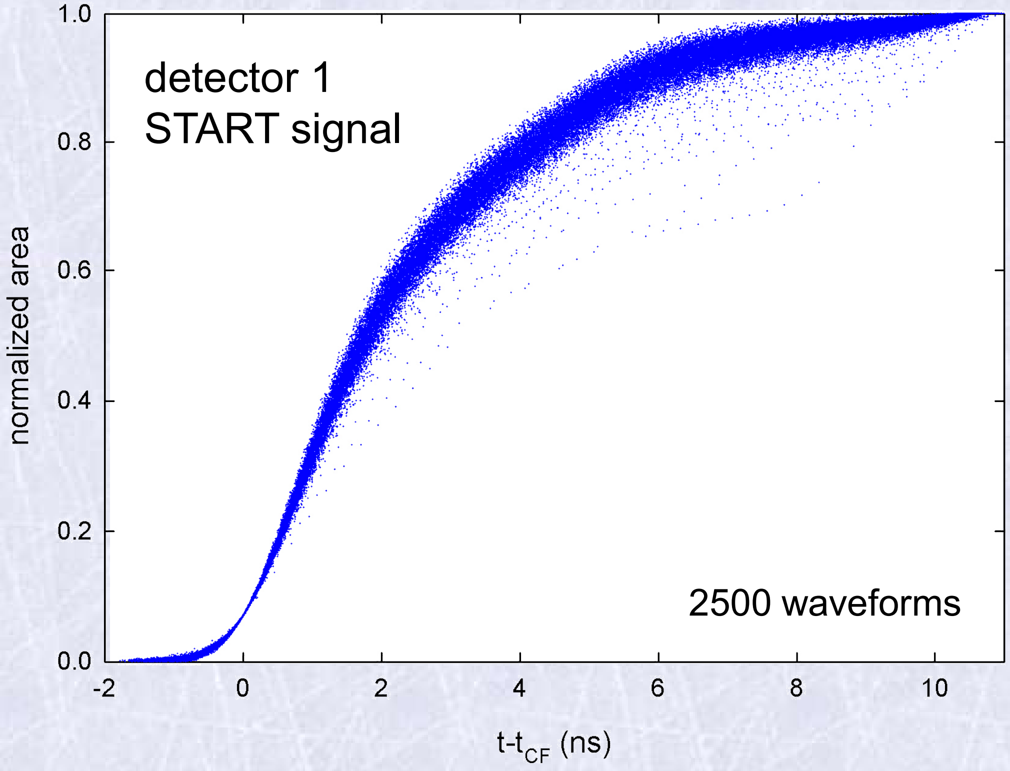
• γ-ray energy spectrum obtained by integration of waveforms



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

Digital positron lifetime spectrometer

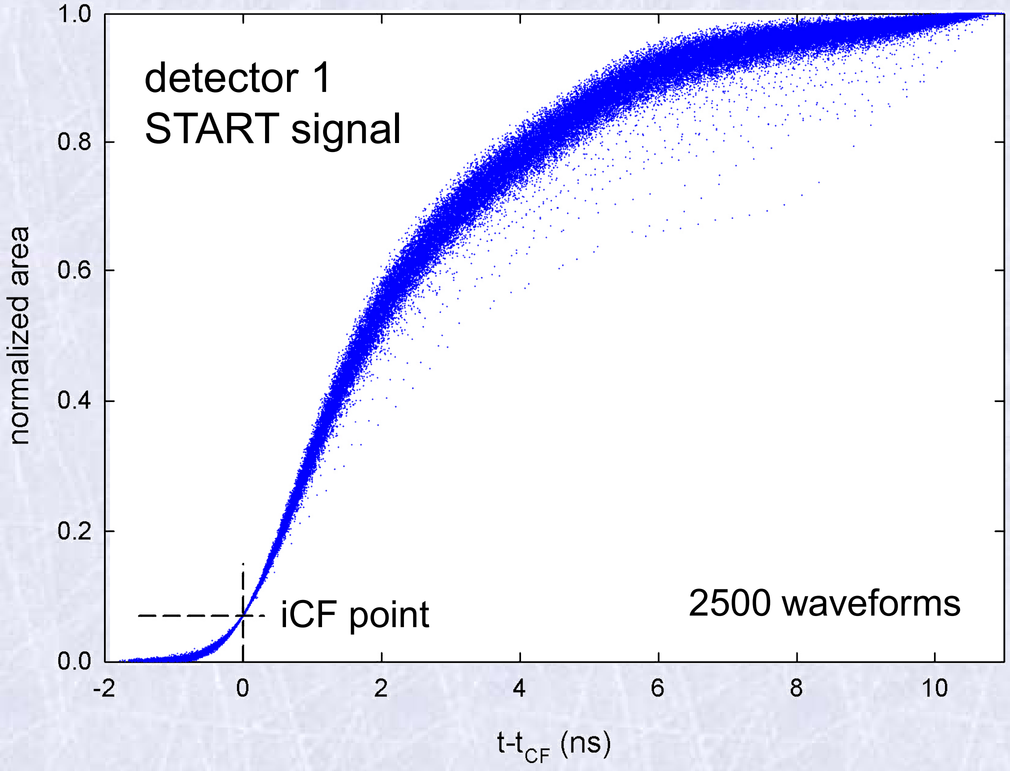
• *integral* constant fraction timing iCF



F. Bečvář , Nucl. Instr. Meth. B ***261****, 871 (2007)*

Digital positron lifetime spectrometer

- *integral* constant fraction timing iCF

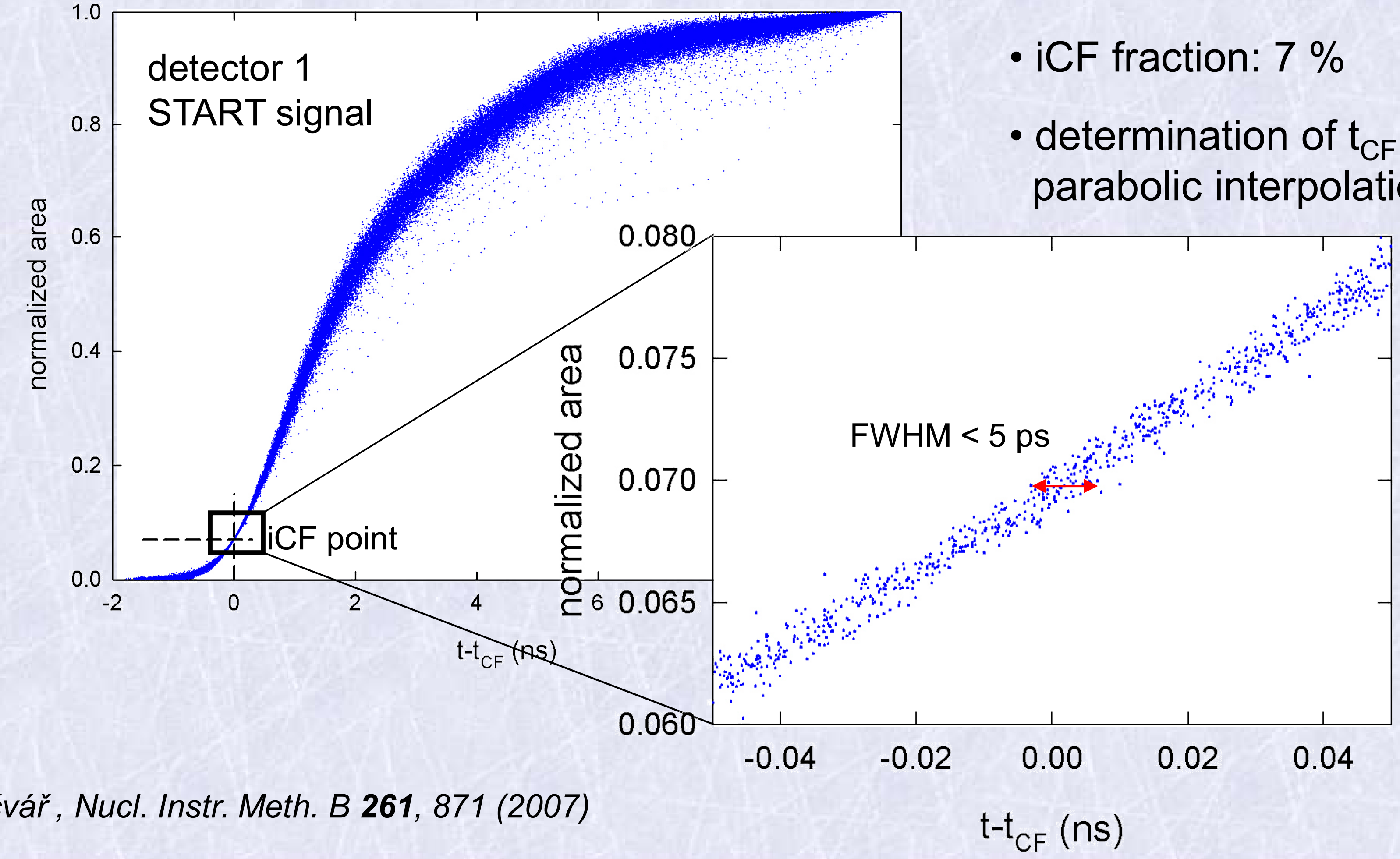


- iCF fraction: 7 %
- determination of t_{CF}: parabolic interpolation

F. Bečvář , Nucl. Instr. Meth. B ***261****, 871 (2007)*

Digital positron lifetime spectrometer

- *integral* constant fraction timing iCF



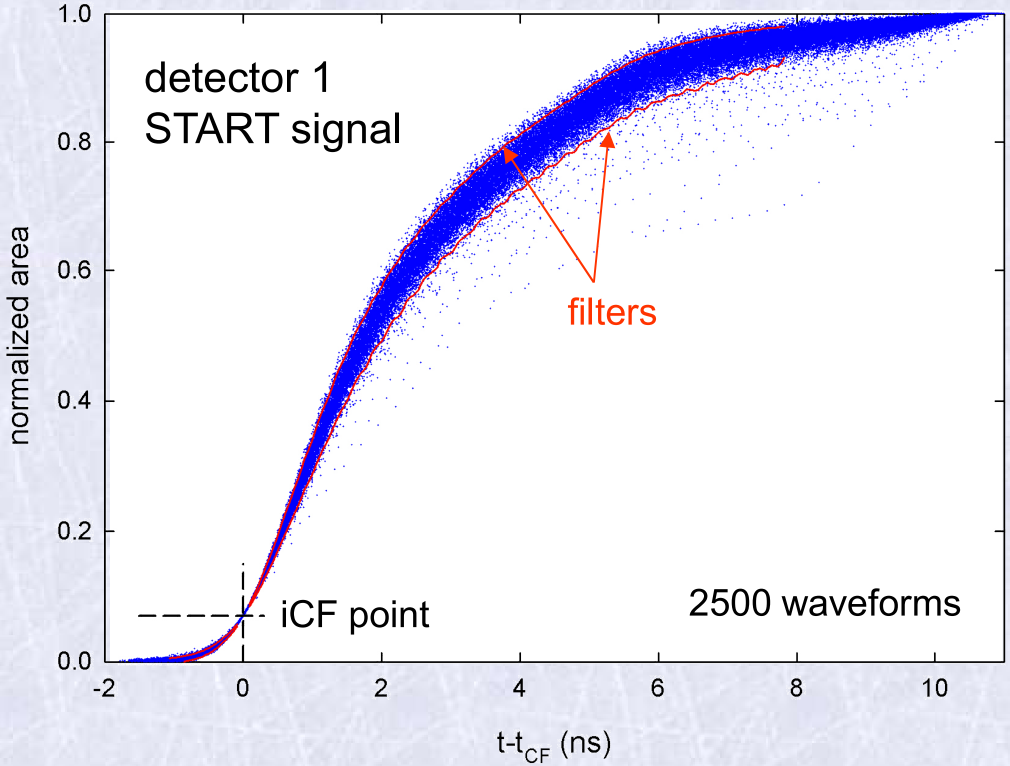
- iCF fraction: 7 %
- determination of t_{CF}: parabolic interpolation

F. Bečvář , Nucl. Instr. Meth. B ***261****, 871 (2007)*

Digital positron lifetime spectrometer

- *integral* constant fraction timing iCF

- shape of pulses controlled by **digital filters** – majorizing and minorizing function



- iCF fraction: 7 %

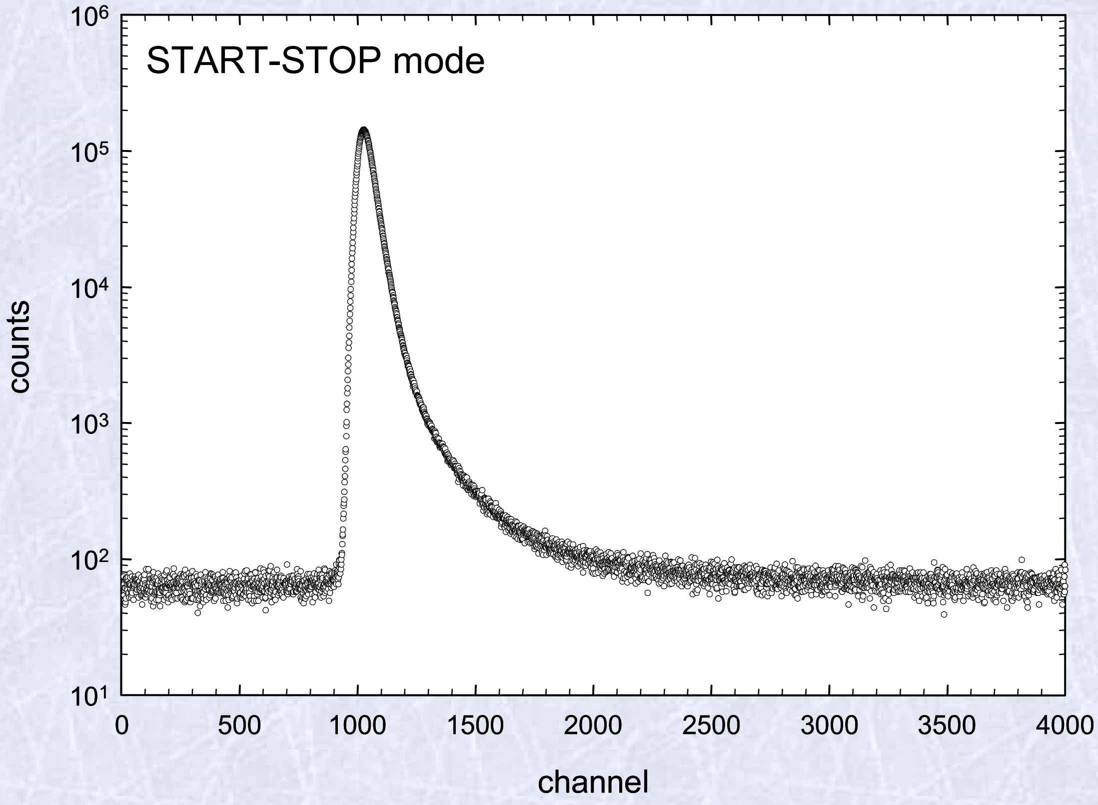
- determination of t_{CF}: parabolic interpolation

F. Bečvář , Nucl. Instr. Meth. B ***261****, 871 (2007)*

Digital positron lifetime spectrometer

• standard source (1.2 MBq), α-Fe reference specimen

• statistics: 8×10^6



Digital positron lifetime spectrometer

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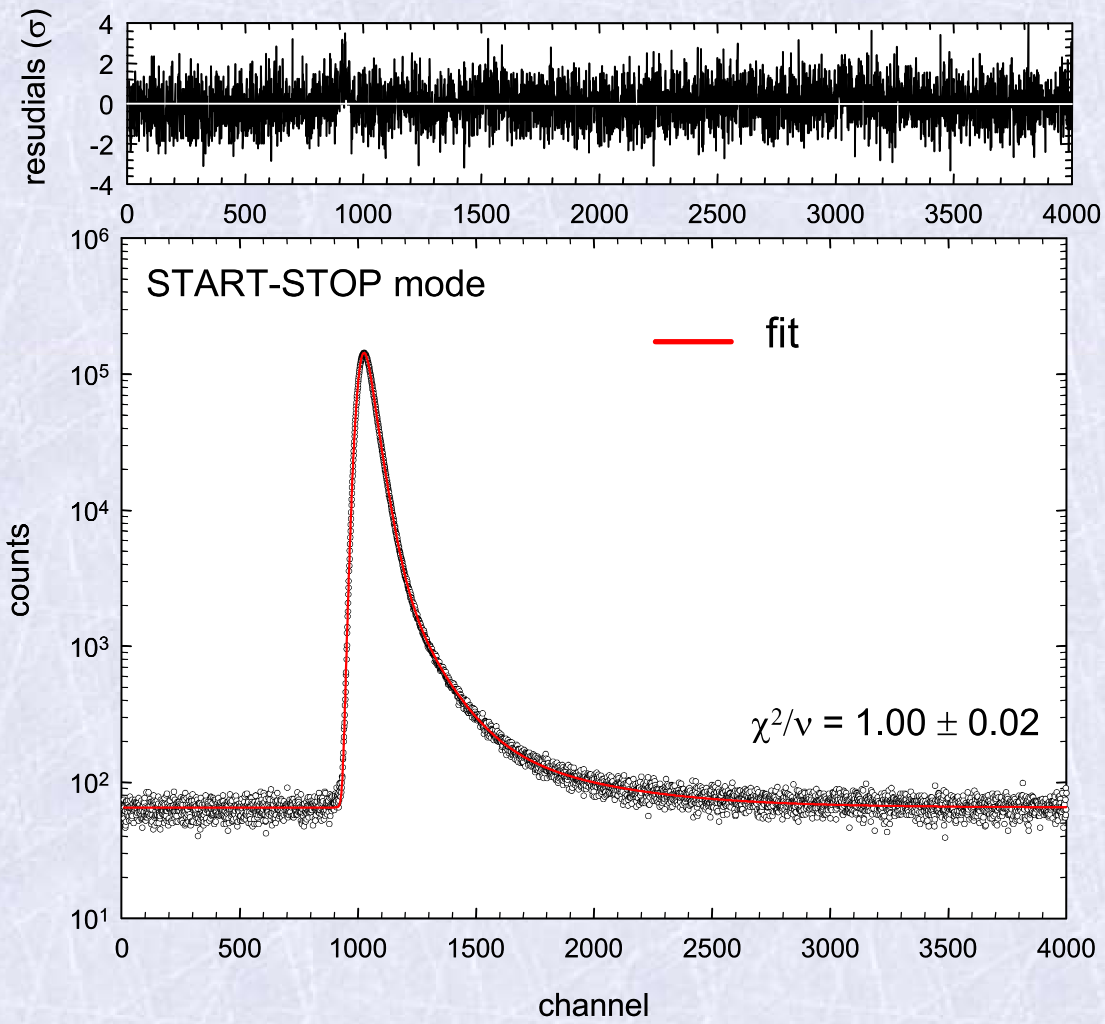

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Digital positron lifetime spectrometer

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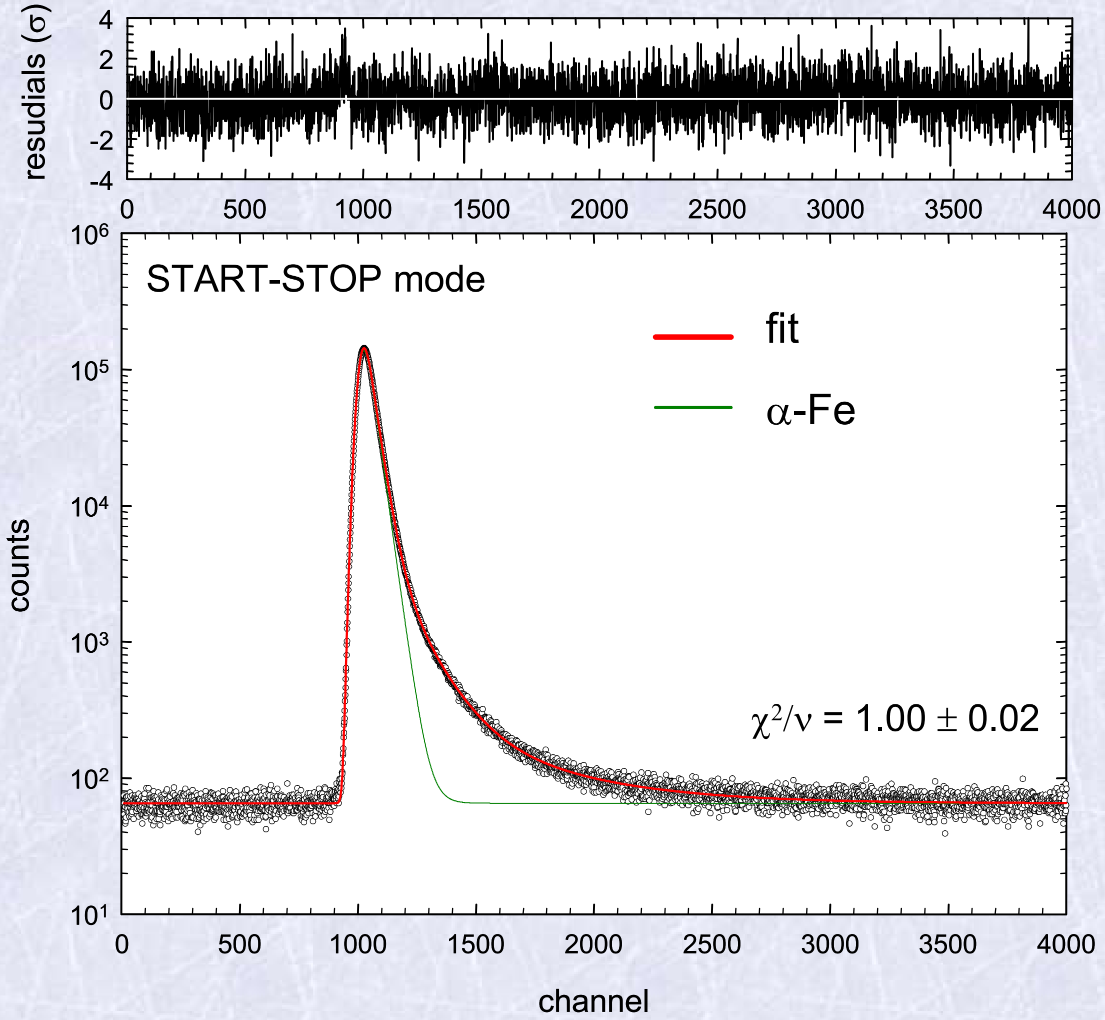
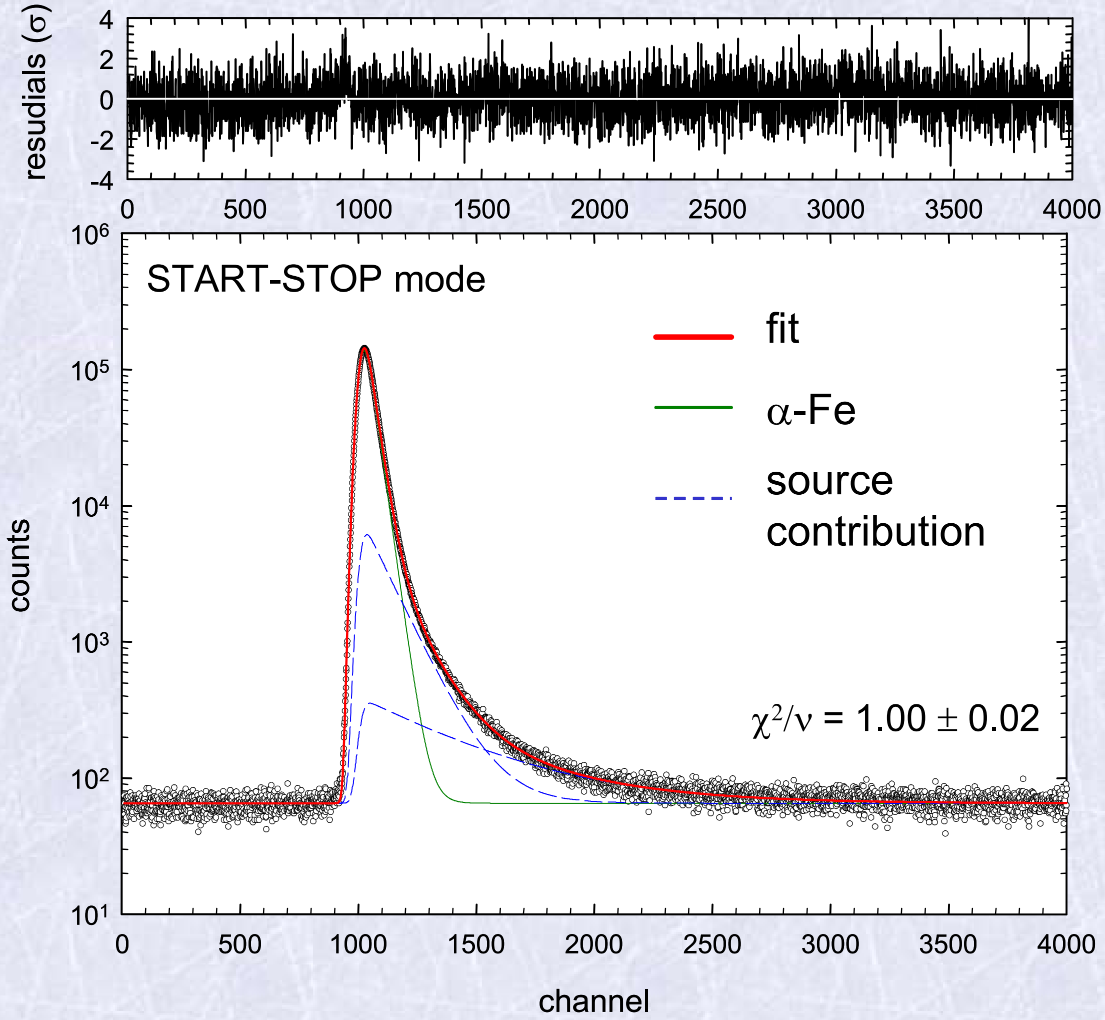


• statistics: 8×10^6

• α-Fe: τ = (107.0 $\pm$ 0.3) ps

Digital positron lifetime spectrometer

- standard source (1.2 MBq), α-Fe reference specimen



- statistics: 8×10^6
- α-Fe: τ = (107.0 ± 0.3) ps
- **time resolution: 143 ps (FWHM)**
- resolution function: two Gaussians

Digital positron lifetime spectrometer

- standard source (1.2 MBq), α-Fe reference specimen
- START-STOP mode
- count rate: 940 s^{-1}
- accumulated coincidences: 200 × 10^6 (2.5 day)

- waveforms outside energy windows: 156 × 10^6 (77.9 %)
- waveforms rejected by digital filters: 34 × 10^6 (17.1 %)
- waveforms rejected due to too noisy baseline: 1.6 × 10^6 (0.8 %)

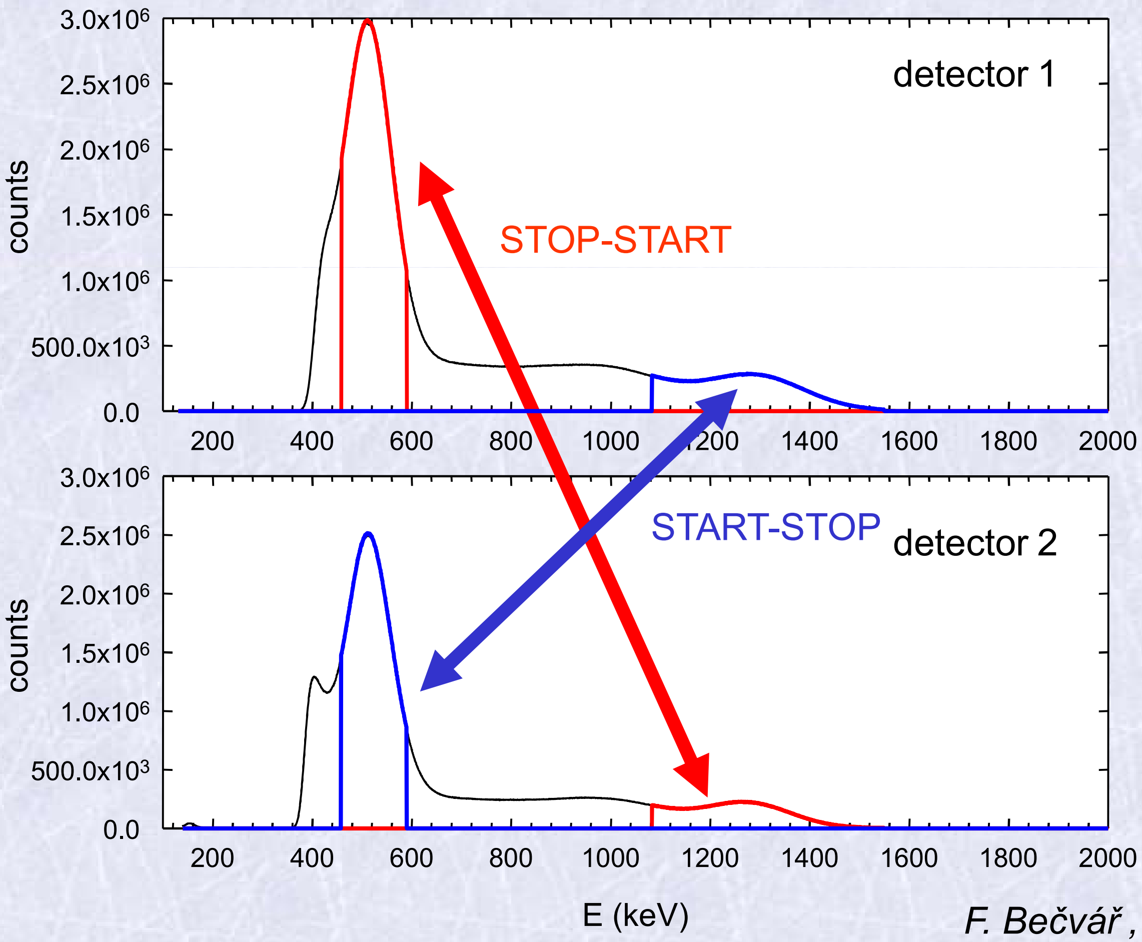
---

accepted waveforms: 8.4 × 10^6 (4.2 %)

(effective coincidence count rate: 40 s^{-1})

Digital positron lifetime spectrometer

- standard source (1.2 MBq), α-Fe reference specimen
- **energy windows**: START: (1080-1550) keV, STOP (460-590)
- **double count rate at no cost**



START-STOP mode:
detector 1: START
detector 2: STOP

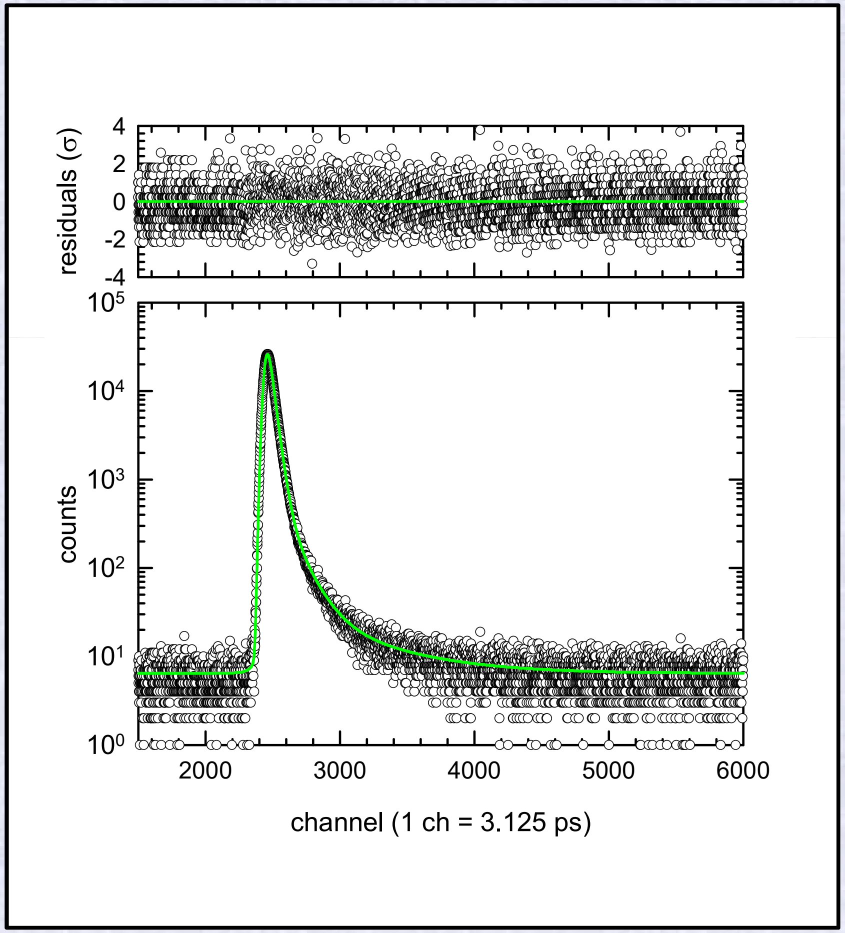
STOP-START mode:
detector 1: STOP
detector 2: START

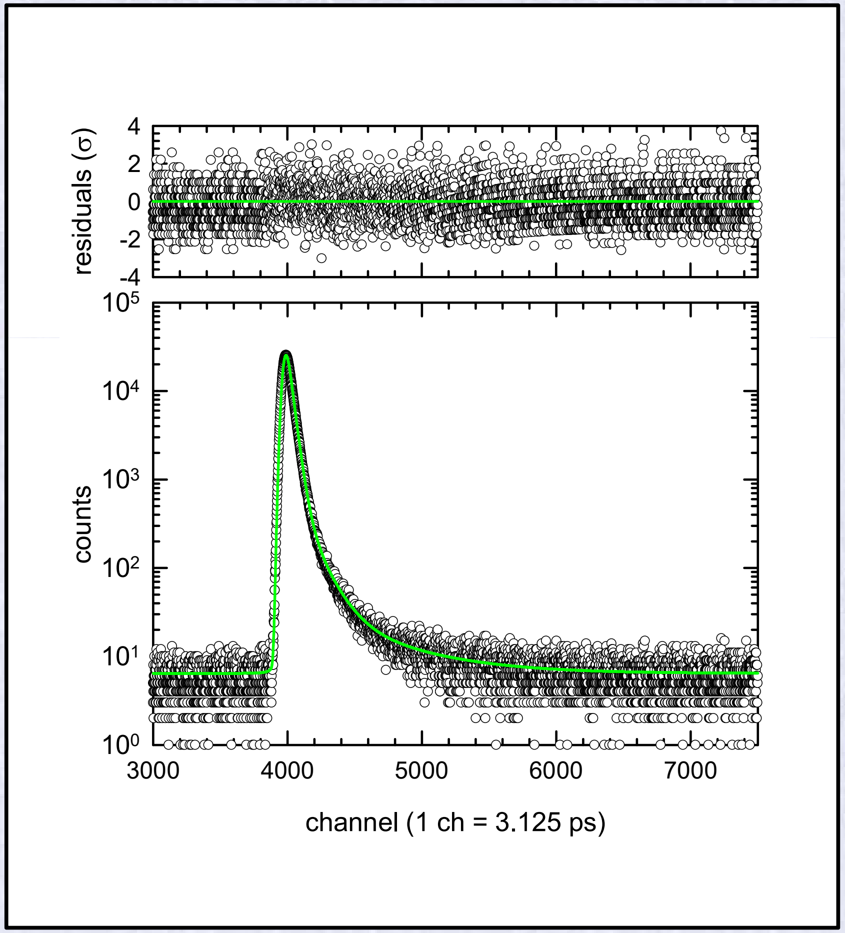
*F. Bečvář , Nucl. Instr. Meth. B **261**, 871 (2007)*

Digital positron lifetime spectrometer

START-STOP mode:
detector 1: START
detector 2: STOP

STOP-START mode:
detector 1: STOP
detector 2: START





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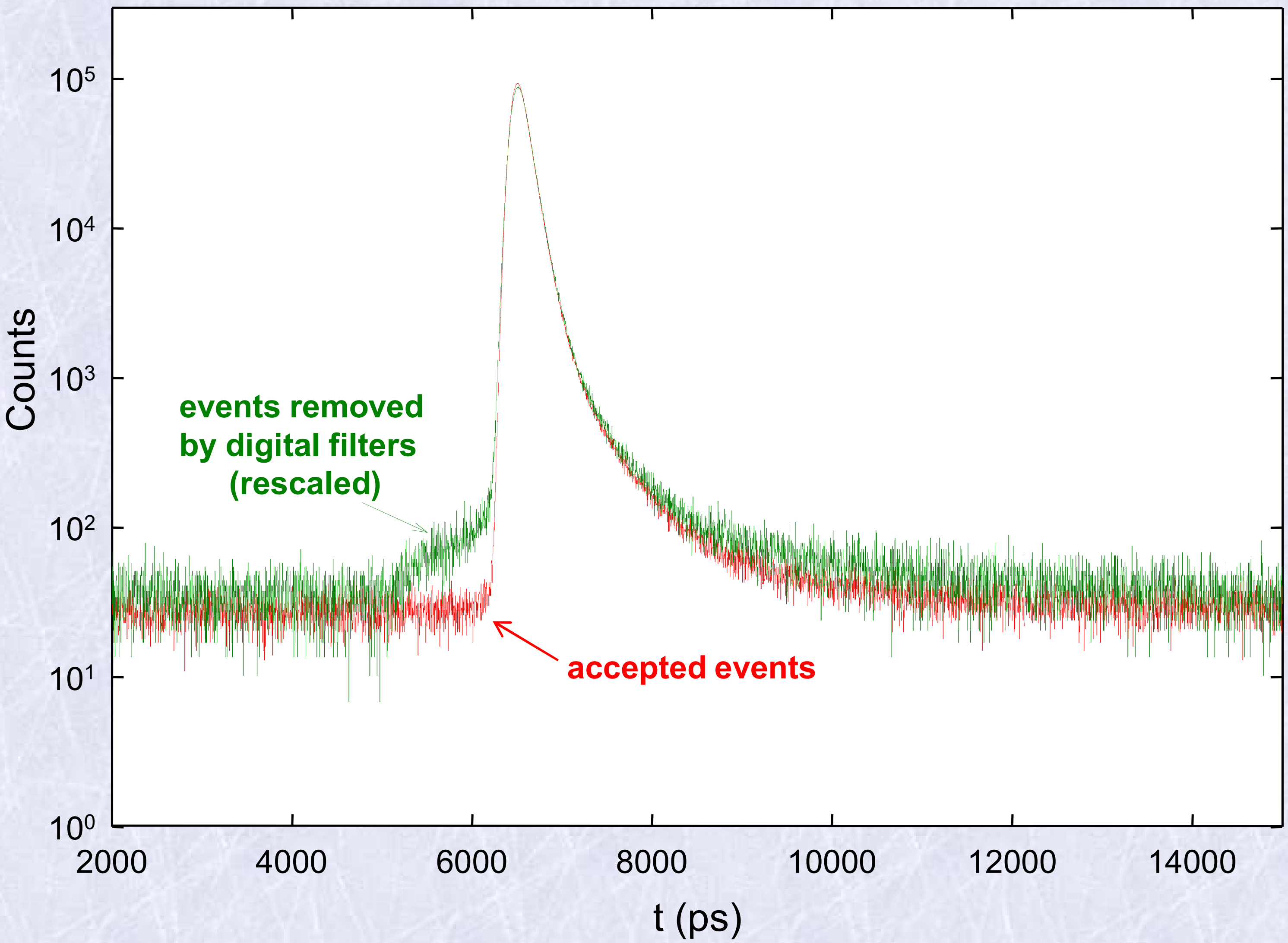
Digital positron lifetime spectrometer



LT spectroscopy

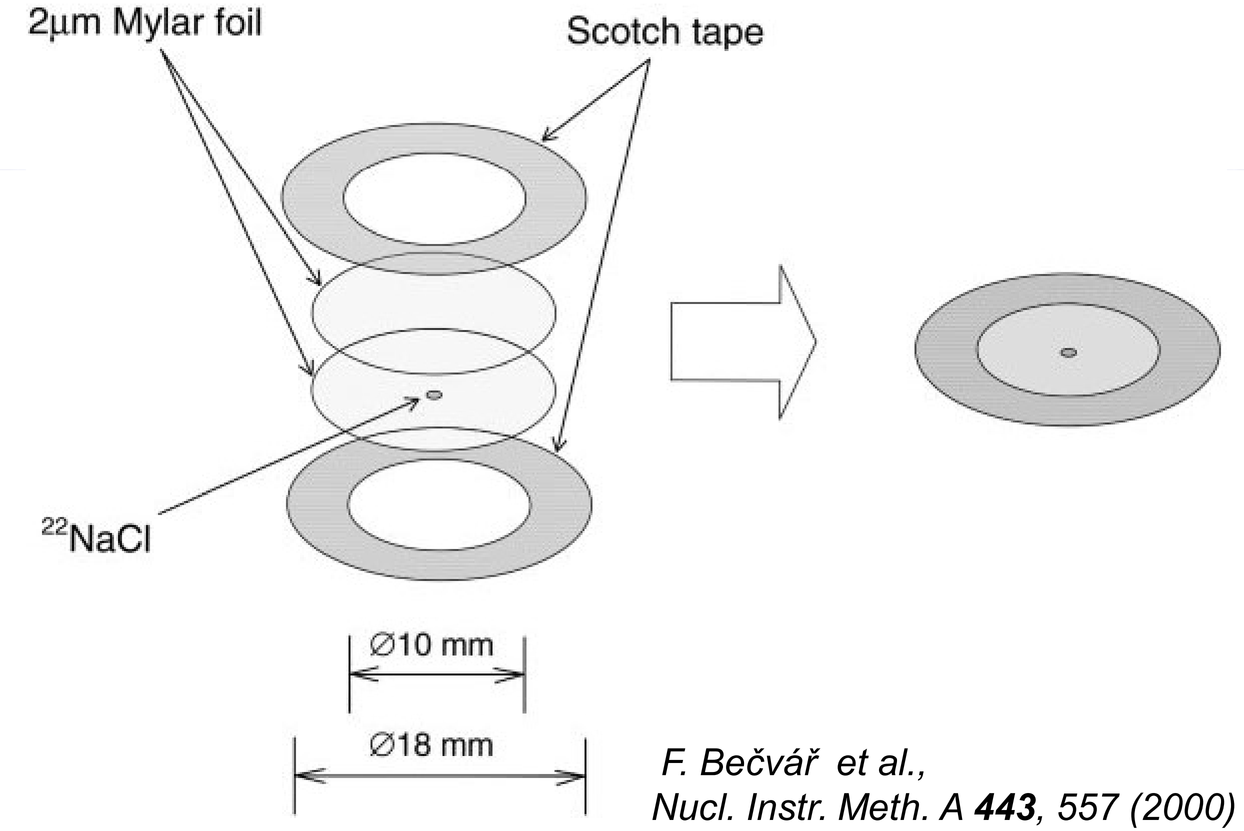
- what is necessary to know for meaningful fitting of LT spectra
- time calibration *F. Bečvář et al., Mater. Sci. Forum* ***363-365****, 695 (2001)*
- source contribution }
- resolution function } measurement of well defined reference sample

Source contribution

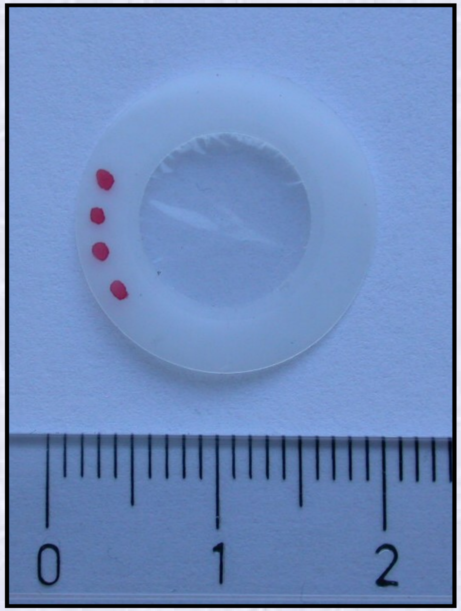
- positron source (1-3) μl $^{22}NaCl$ (1-2 MBq)

e⁺ annihilation → Ps annihilation

- $^{22}NaCl$ deposited on 2 μm mylar foil: τ_{s1} = 368(2) ps, $\tau_{s2,\ p\text{-}Ps}$ = 125 ps, $\tau_{s2,\ o\text{-}Ps}$ = 1.5(3) ns,
- $^{22}NaCl$ deposited on 7.6 μm kapton foil: τ_{s1} = 403(1) ps
- $^{22}NaCl$ deposited on 5 μm Ti foil: τ_{s1} = 380(5) ps, τ_{s2} = 174(4) ps

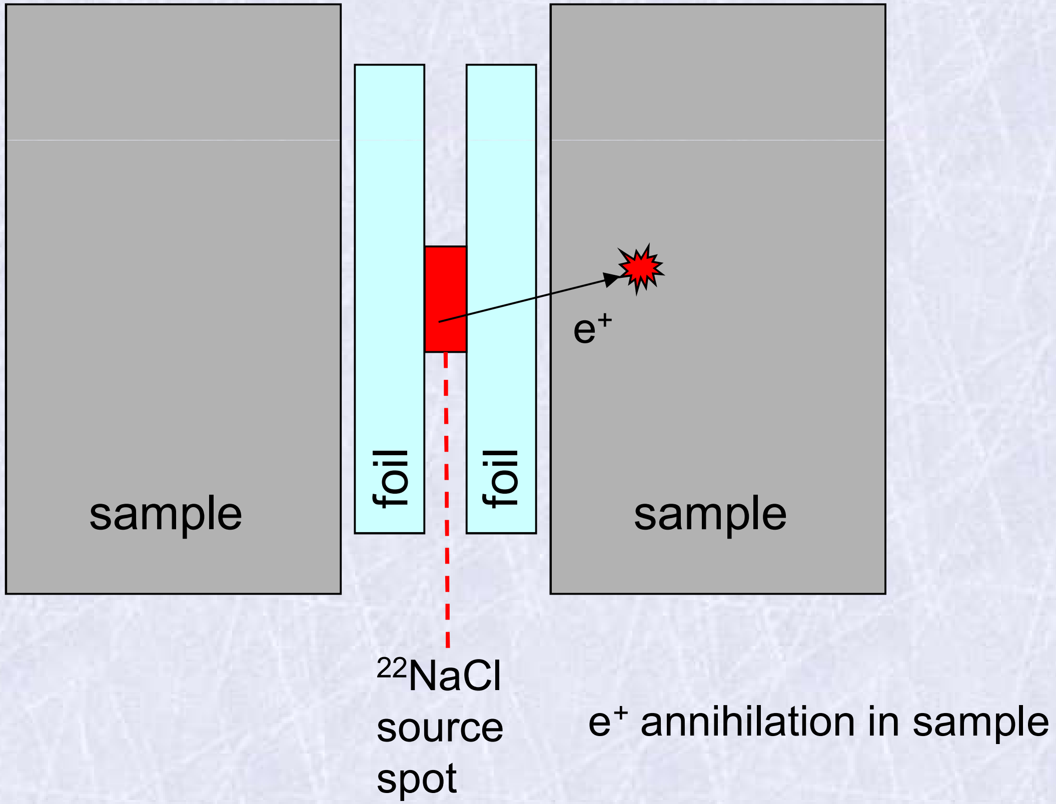


*F. Bečvář et al., Nucl. Instr. Meth. A **443**, 557 (2000)*



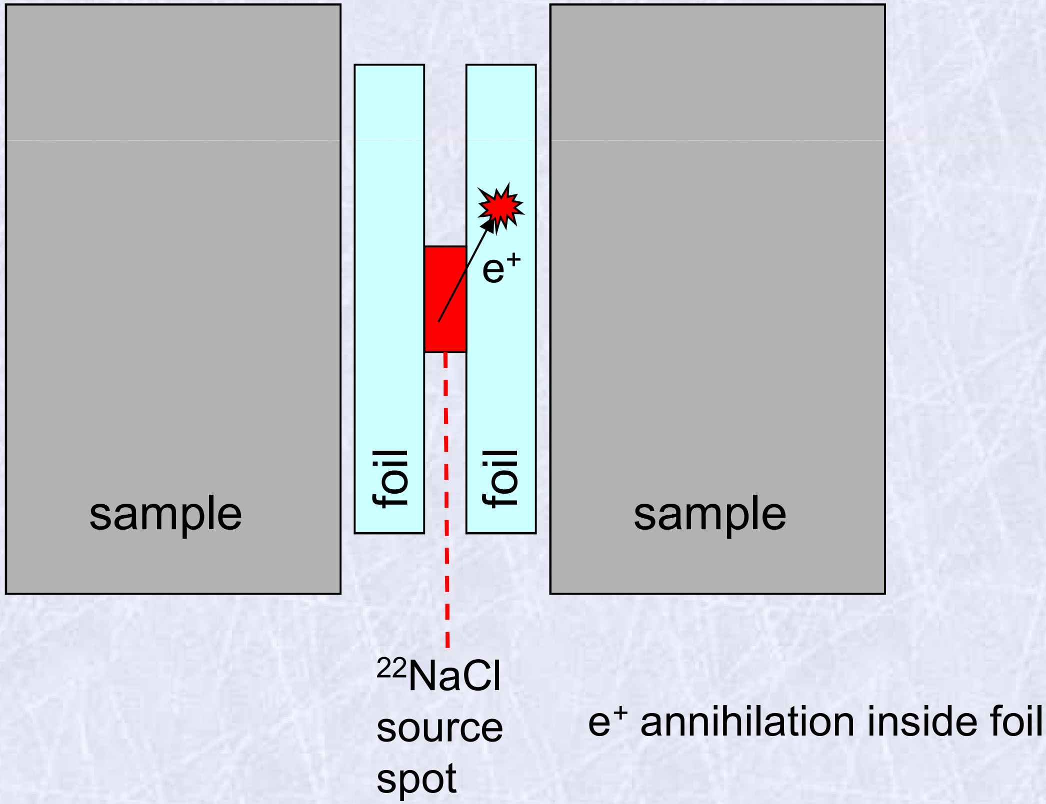
Source contribution

- intensity of source contribution



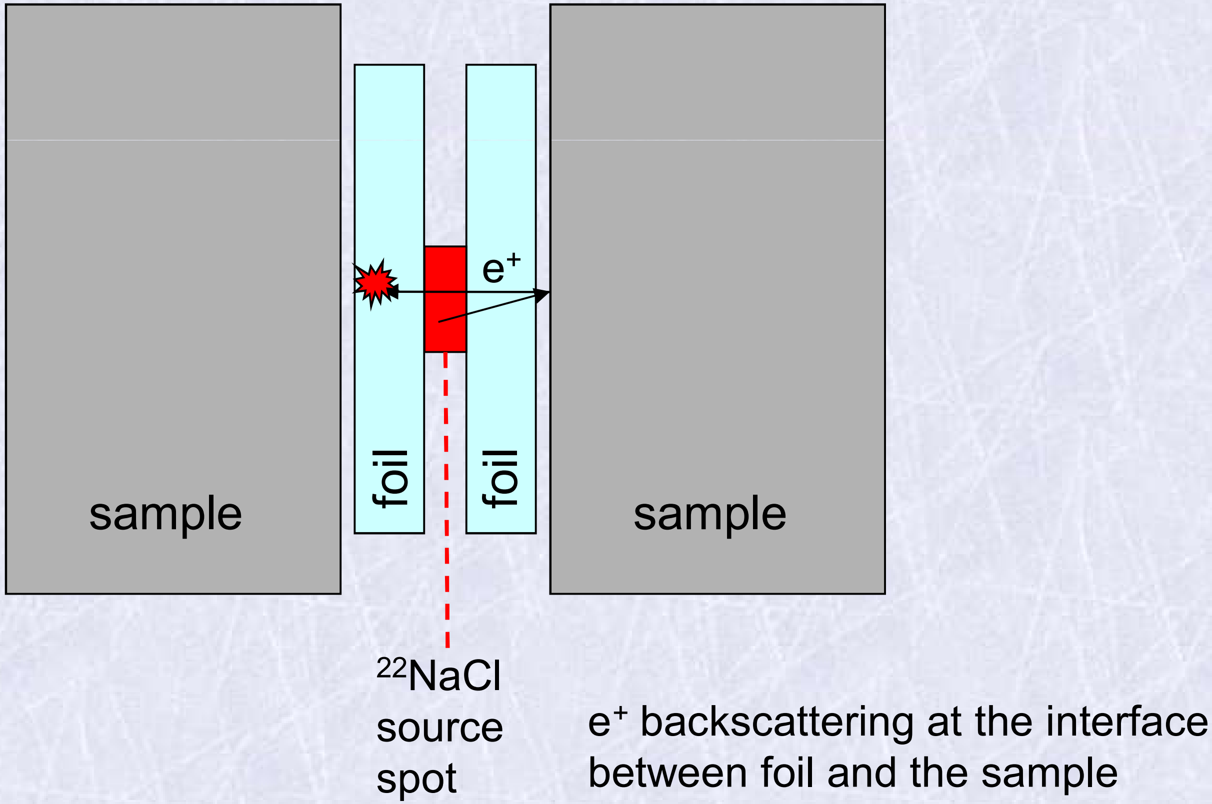
Source contribution

- intensity of source contribution



Source contribution

- positron reflection on the interface between materials with various Z
- positron reflectance increases with Z
- This makes intensity of the source contribution increasing with Z of the sample



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- Intensity of the source contribution: $I_S = I_{S0} + I(Z)$

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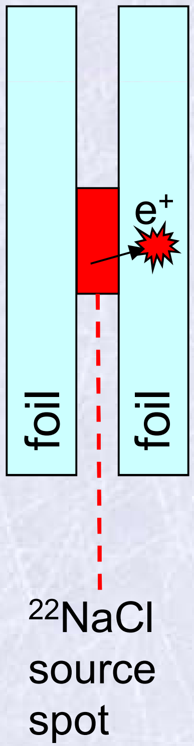
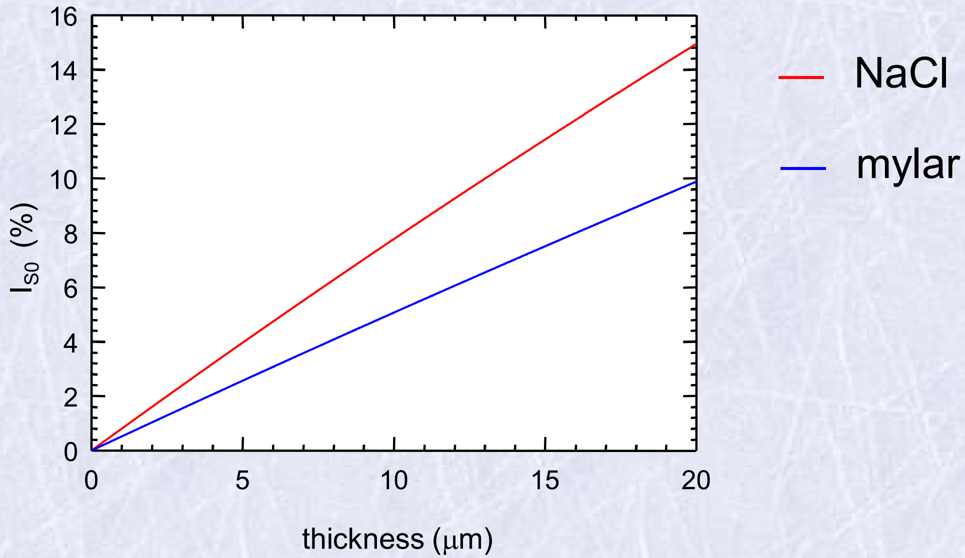
- This makes intensity of the source contribution increasing with Z of the sample
- Intensity of the source contribution: $I_S = I_{S0} + I(Z)$

constant





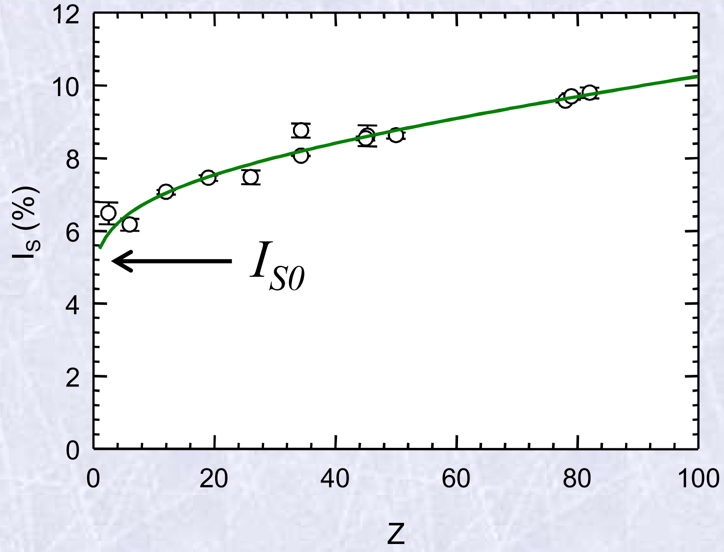
Source contribution

- positron reflection on the interface between materials with various Z
- positron reflectance increases with Z
- This makes intensity of the source contribution increasing with Z of the sample
- Intensity of the source contribution: $I_S = I_{S0} + I(Z)$

$$I(Z) = a\ln(Z+1) + dZ$$

$a = 0.70(1)$

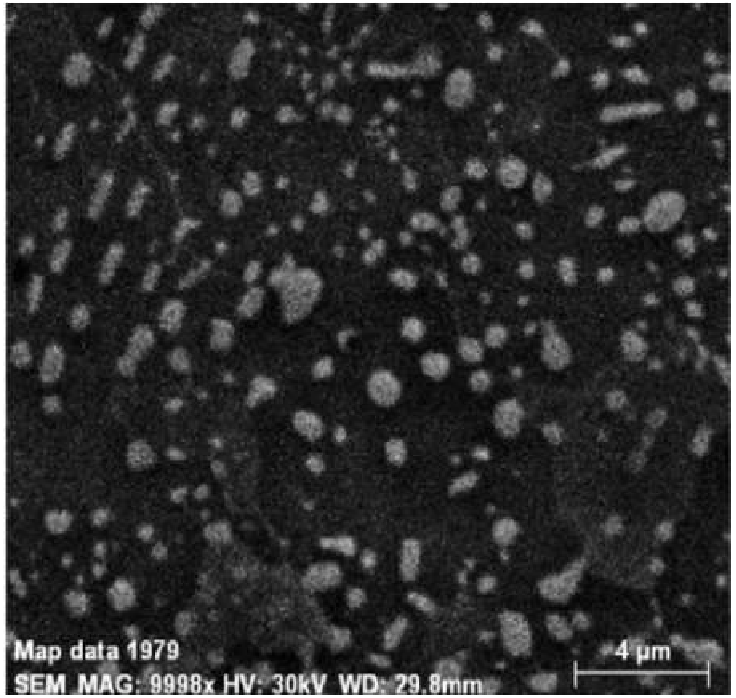
$b = 0.020(2)$



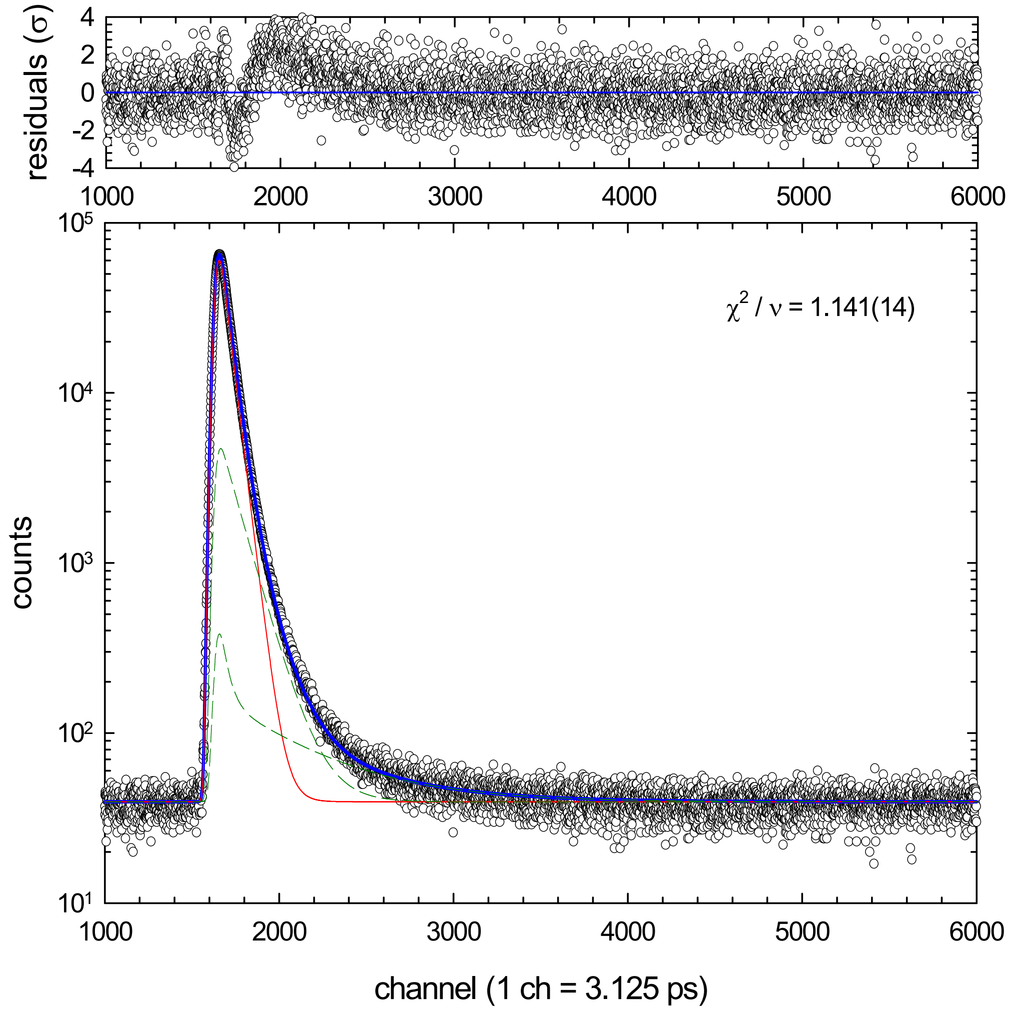
Fitting of LT spectrum

- 100Cr6 roll bearing steel

C	Cr	Mn	Si	S	P	Fe
1.04(3)	1.45(9)	0.35(6)	0.25(6)	< 0.025	< 0.025	balance



- single component fit
  τ_1 = 155.7(1) ps



Fitting of LT spectrum

- 100Cr6 roll bearing steel

- χ^2 **test**

$$\chi^2 = \frac{\sum_{i=1}^{m}\left(y_i - h(x_i|\boldsymbol{\theta})\right)^2}{h(x_i|\boldsymbol{\theta})}$$

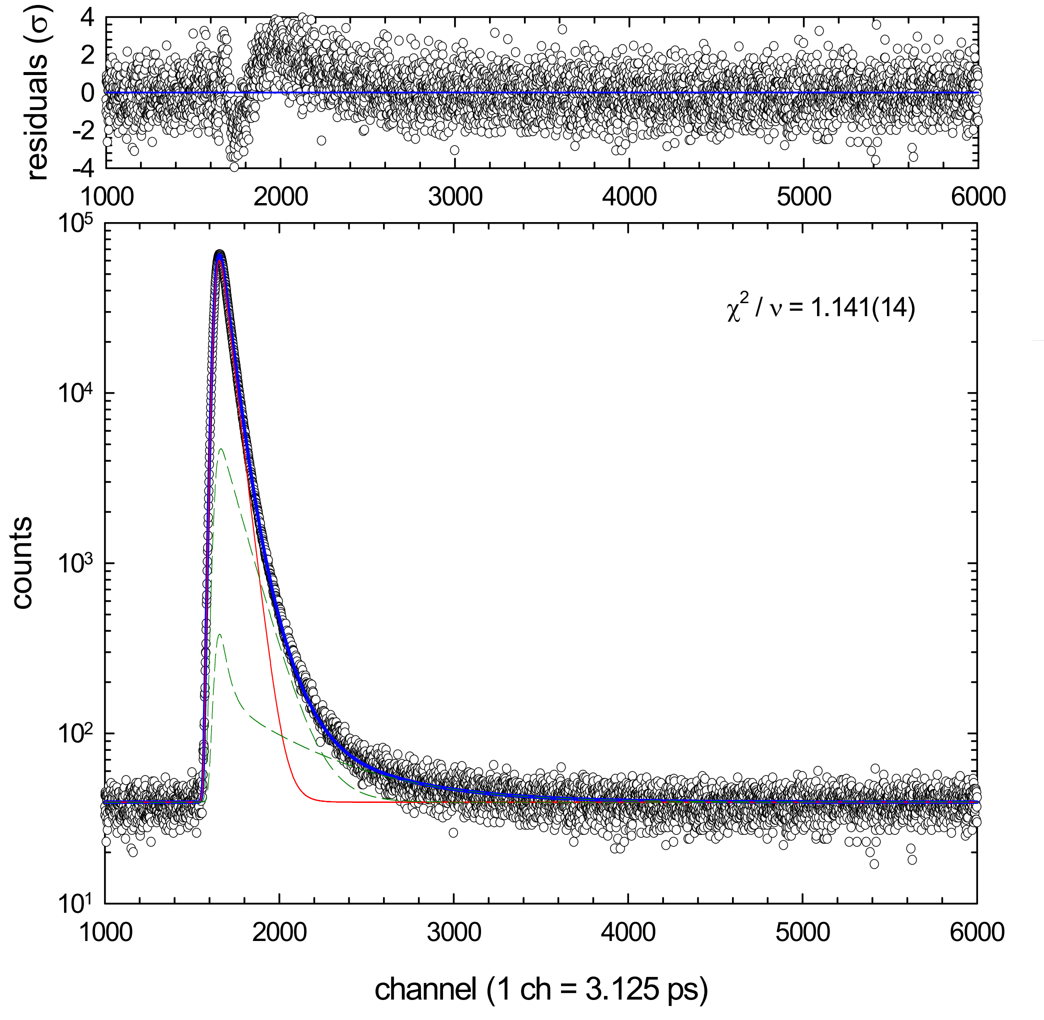
- χ^2 per degree of freedom: χ^2 / ν
- expectation value: 1
- variance: $\sigma^2 = 2/\nu$
- number of degrees of freedom

$$\nu = m - p$$

number of channels (m); number of fitting parameters (p)

- single component fit

τ_1 = 155.7(1) ps



Fitting of LT spectrum

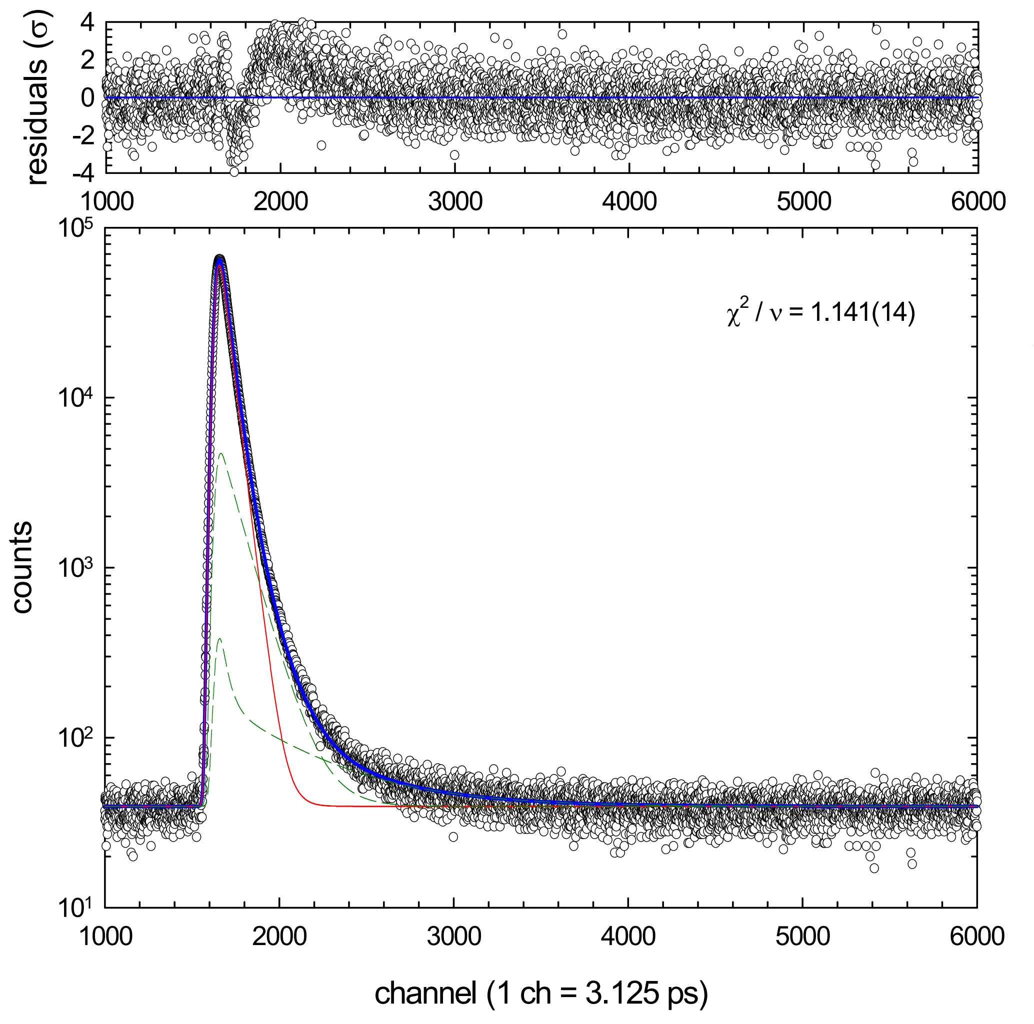
- 100Cr6 roll bearing steel

- single component fit
  τ_1 = 155.7(1) ps

- **residuals**

$$r = \frac{\sum_{i=1}^{m}\left(y_i - h(x_i|\boldsymbol{\theta})\right)}{\sqrt{h(x_i|\boldsymbol{\theta})}}$$

- r has normal distribution
- expectation value: 0
- variance: $\sigma^2 = 1$



Fitting of LT spectrum

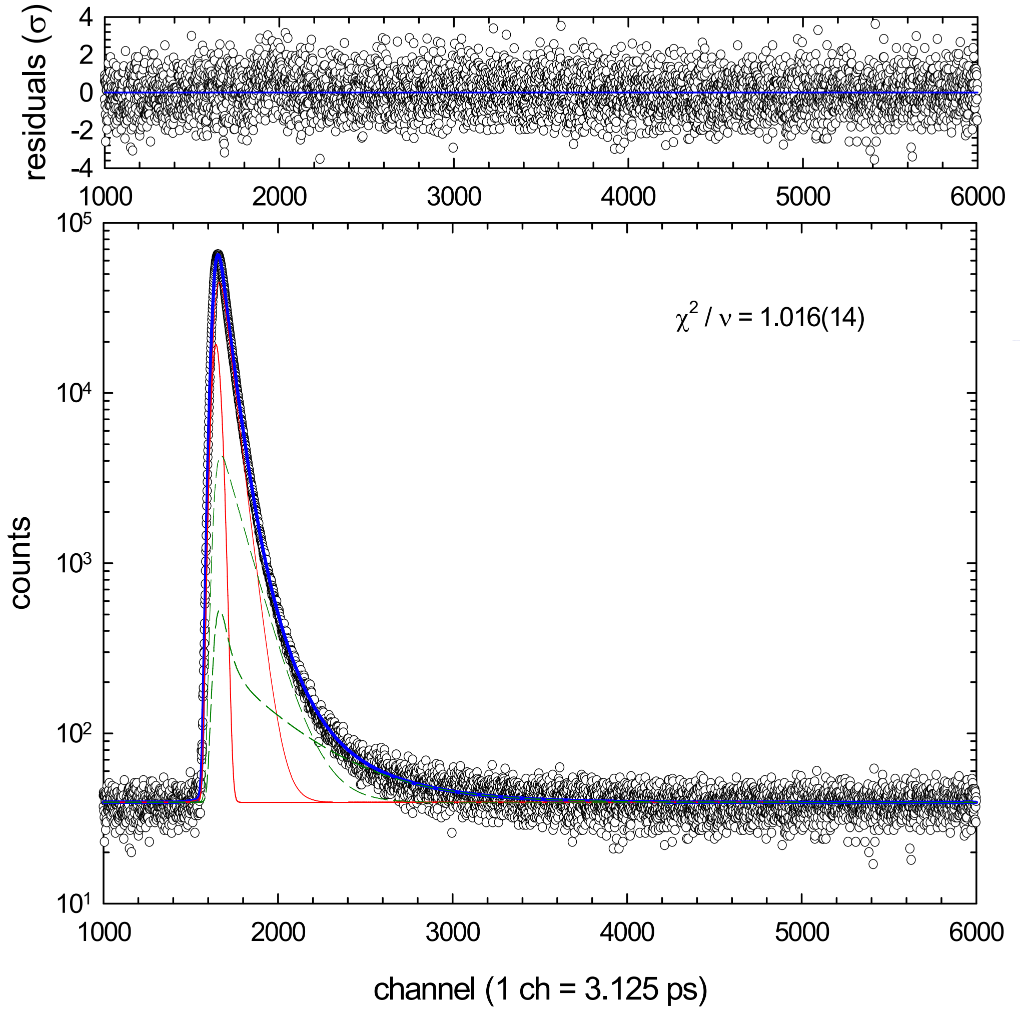
- 100Cr6 roll bearing steel

- two-component fit
  τ_1 = 20(5) ps, I_1 = 19(6)%,
  τ_2 = 156(3) ps, I_2 = 81(5)%

- **residuals**

$$r = \frac{\sum_{i=1}^{m}\left(y_i - h(x_i|\boldsymbol{\theta})\right)}{\sqrt{h(x_i|\boldsymbol{\theta})}}$$

- r has normal distribution
- expectation value: 0
- variance: $\sigma^2 = 1$



LT spectrum

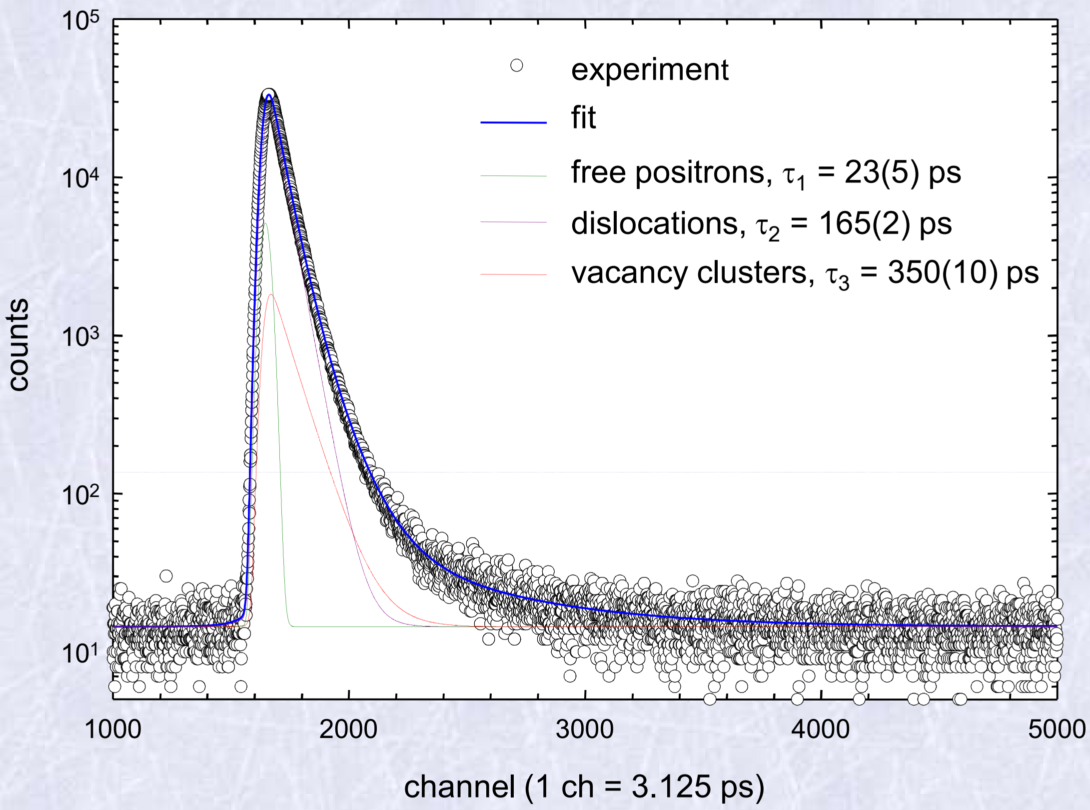
N type of defects

↓

$N+1$ components

$$S_{id} = \sum_{i=1}^{N+1} \frac{1}{\tau_i} I_i e^{-\frac{t}{\tau_i}}$$

LT spectrum of heavily deformed Nb



decomposition of LT spectrum

lifetimes τ_i → type of defects

intensities I_i → defect concentrations

LT spectroscopy

• fitting of LT spectrum

↓

• lifetimes τ_i and intensities I_i of exponential components

↓

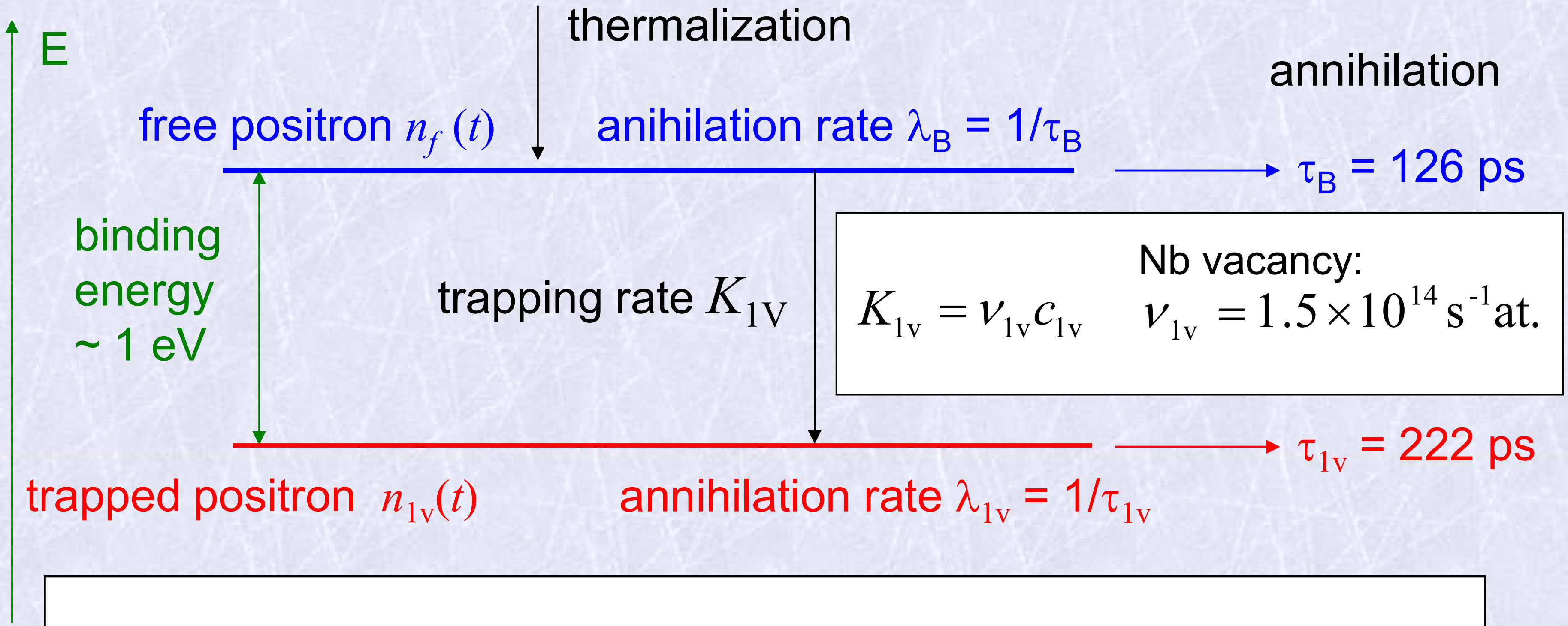
• (comparison with *ab-initio* theoretical calculations) ⟶ identification of defects

↓

• application of positron trapping model ⟶ determination of defect concentrations

Simple trapping model (STM)

Nb with vacancies



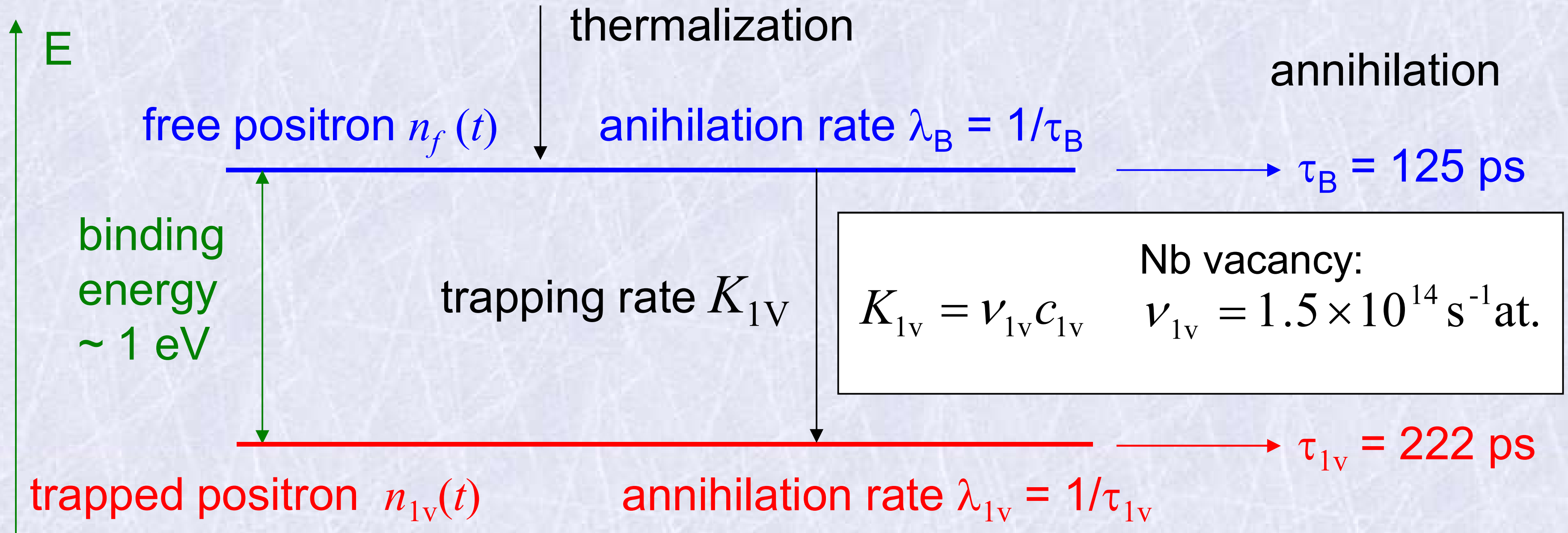
assumptions:

1. single type of defects
2. uniform distribution of defects
3. only thermalized e^+ are trapped
4. no detrapping

R.N. West Adv. Phys. 22, 263 (1973)

Simple trapping model (STM)

Nb with vacancies



$$\frac{dn_f}{dt} = -\lambda_B n_f - K_{1v} n_f$$

$n_f(t)$ ..... probability that e^+ is in the free state at time t

$$\frac{dn_{1v}}{dt} = -\lambda_{1v} n_{1v} + K_{1v} n_f$$

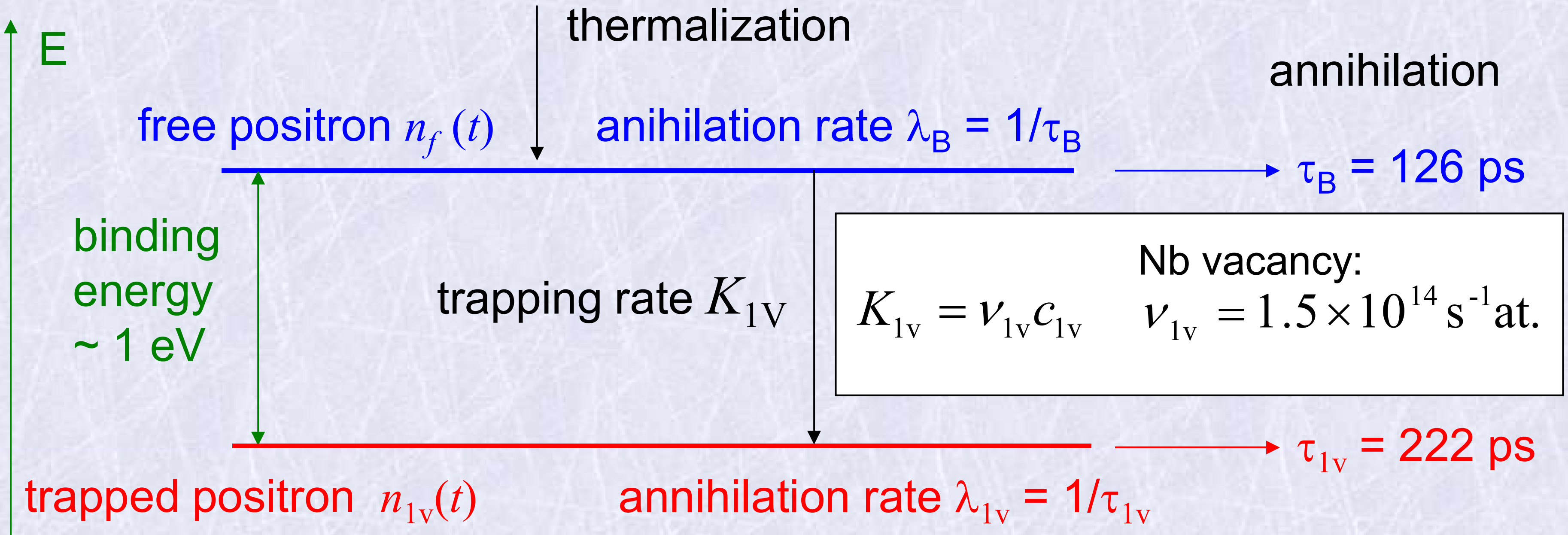
$n_{1v}(t)$ ..... probability that e^+ is in trapped state at time t

$$n_{1v}(t=0) = 0 \qquad n_f(t=0) = 1$$

R.N. West Adv. Phys. 22, 263 (1973)

Simple trapping model (STM)

Nb with vacancies



two-component spectrum: $$S_{id} = \frac{1}{\tau_1} I_1 e^{-\frac{t}{\tau_1}} + \frac{1}{\tau_2} I_2 e^{-\frac{t}{\tau_2}}$$

$$\tau_1 = \frac{1}{\lambda_B + K_{1v}} \qquad I_1 = 1 - I_2$$

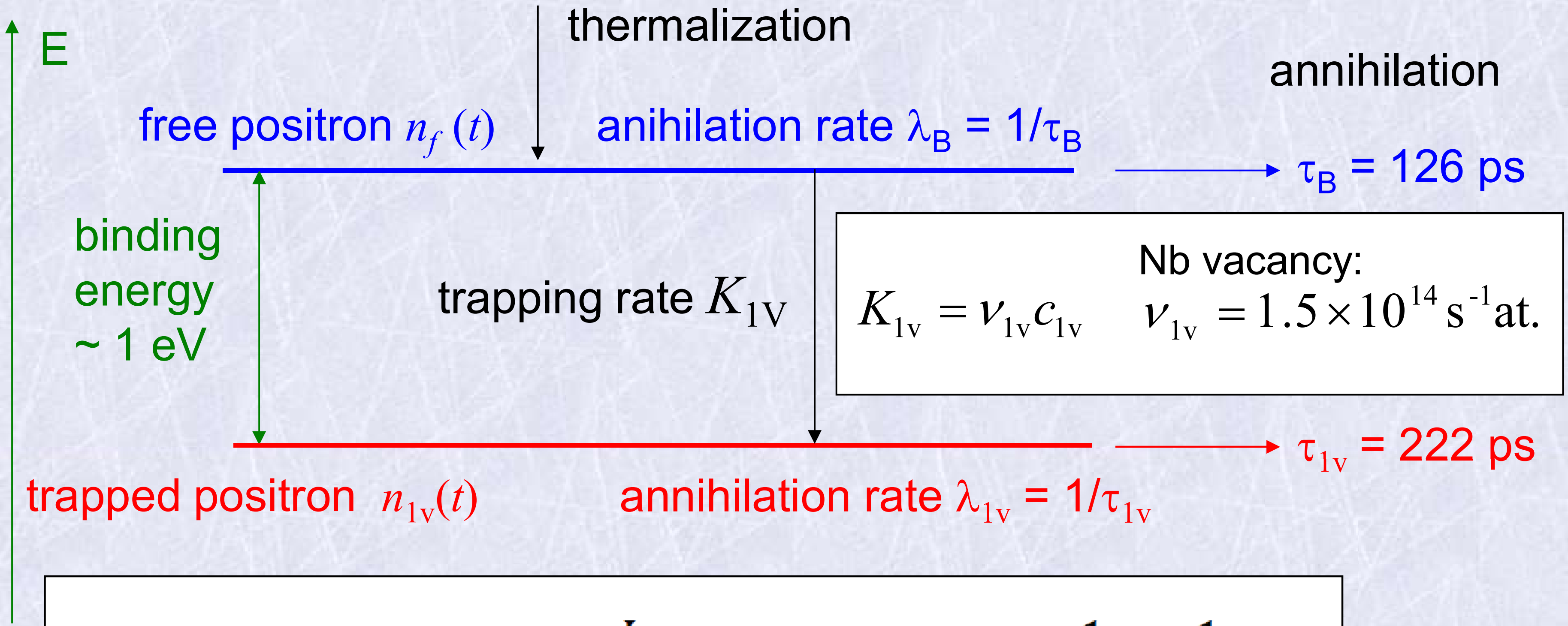
free positrons

$$\tau_2 = \frac{1}{\lambda_{1v}} \qquad I_2 = \frac{K_{1v}}{\lambda_B + K_{1v} - \lambda_{1v}}$$

positrons trapped
in vacancies

Simple trapping model (STM)

Nb with vacancies



trapping rate: $$K_{1V} = \frac{I_2}{I_1}(\lambda_B - \lambda_{1V}) = I_2\left(\frac{1}{\tau_1} - \frac{1}{\tau_2}\right)$$

concentration of vacancies: $$c_{1V} = \frac{K_{1V}}{\nu_{1V}}$$

Simple trapping model (STM)

Nb with vacancies

• check whether STM assumptions are fulfilled

$$\tau_1 = \frac{1}{\lambda_B + K_{1v}} \qquad I_1 = 1 - I_2$$

free positrons

$$\tau_2 = \frac{1}{\lambda_{1v}} \qquad I_2 = \frac{K_{1v}}{\lambda_B + K_{1v} - \lambda_{1v}}$$

positrons trapped in vacancies

$$\tau_B = \left(\frac{I_1}{\tau_1} + \frac{I_2}{\tau_2}\right)^{-1}$$

• for example: electron irradiated Nb

τ_1 = 78(3) ps, I_1 = 46(2) %

τ_2 = 217(5) ps, I_2 = 64(2) %

$\tau_f = (I_1/\tau_1 + I_2/\tau_2)^{-1}$ = 124(4) ps

vacancy concentration: c_{1V} = 32(5) ppm

• defect-free Nb reference: τ_B = 125(1) ps

LT spectroscopy

fitting LT spectrum directly by the trapping model

- fitting of LT spectrum ←

fitting parameters: $\tau_B = 1/\lambda_B,\ \tau_{1v} = 1/\lambda_{1v},\ K_{1v}$

↓

- lifetimes τ_i and intensities I_i of exponential components

$$\frac{1}{\tau_B} = \left(\frac{I_1}{\tau_1} + \frac{I_2}{\tau_2}\right)$$

↓

- (comparison with *ab-initio* theoretical calculations) ⟶ identification of defects

↓

- application of positron trapping model ⟶ determination of defect concentrations

$$\tau_1 = \frac{1}{\lambda_B + K_{1v}} \qquad I_1 = 1 - I_2$$

free positrons

$$\tau_2 = \frac{1}{\lambda_{1v}} \qquad I_2 = \frac{K_{1v}}{\lambda_B + K_{1v} - \lambda_{1v}}$$

positrons trapped in vacancies

$$S_{id} = \frac{1}{\tau_1} I_1 e^{-\frac{t}{\tau_1}} + \frac{1}{\tau_2} I_2 e^{-\frac{t}{\tau_2}}$$

model LT spectrum

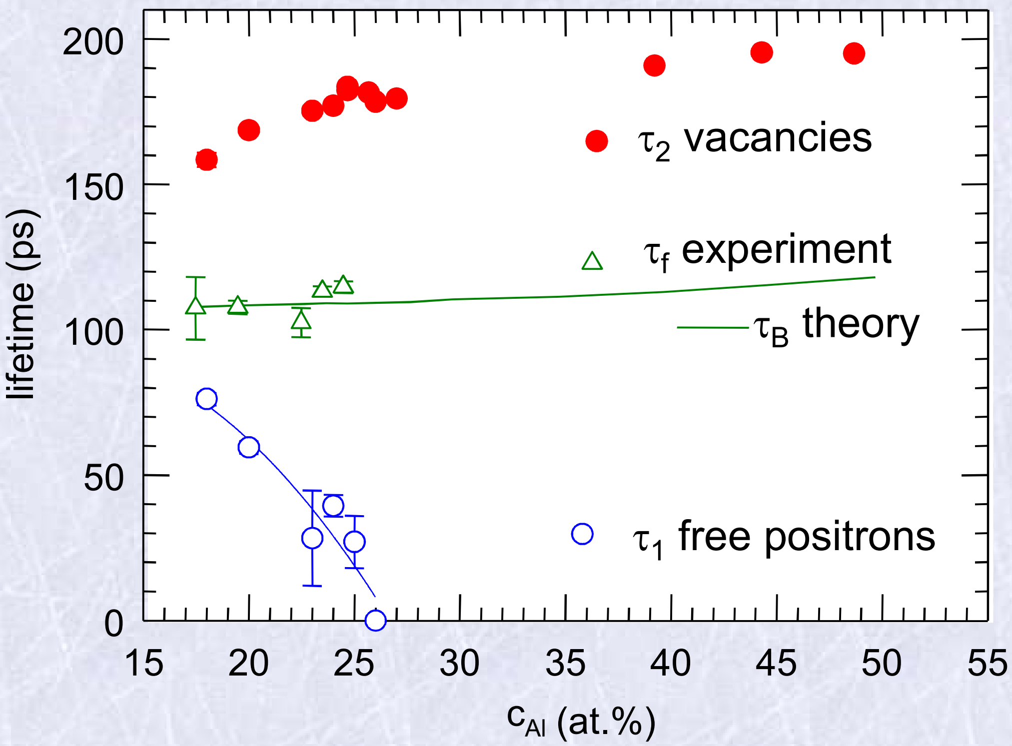
Simple trapping model (STM)

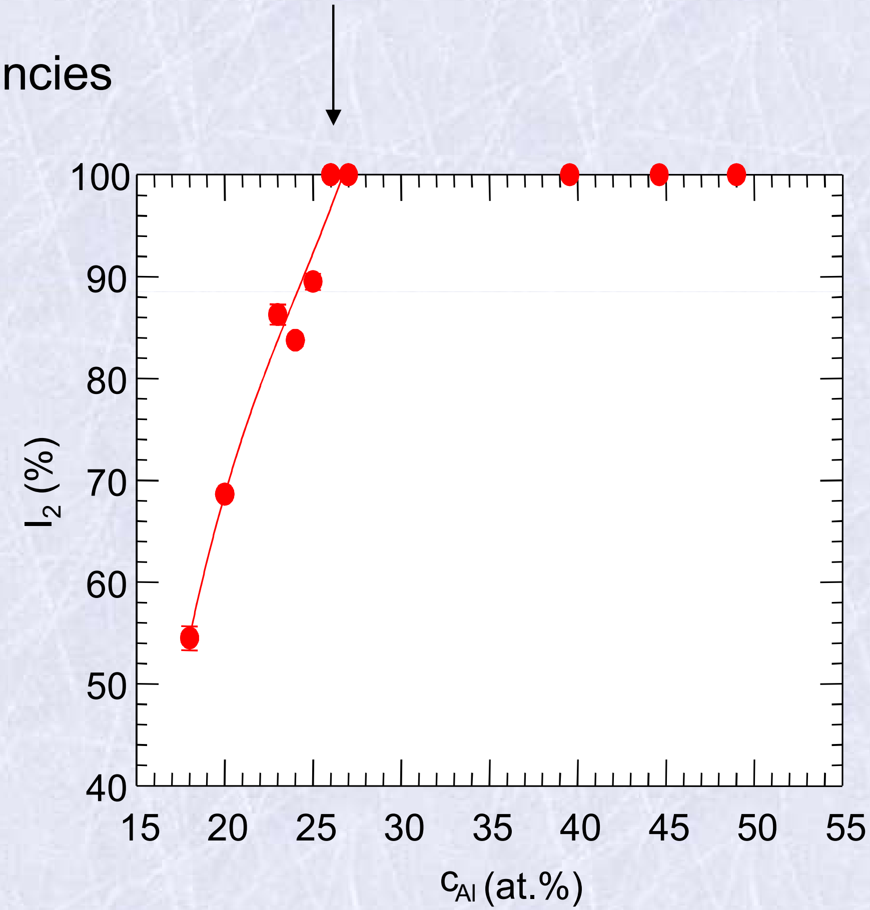
Fe-Al alloys

- quenched from 1000°C
- two component LT spectrum:
  τ_1 – free positrons
  τ_2 – positrons trapped at quenched-in vacancies

$$\tau_f = \left(\frac{I_1}{\tau_1} + \frac{I_2}{\tau_2} \right)^{-1}$$

$c_{Al} \geq 26$ at.% → saturated trapping

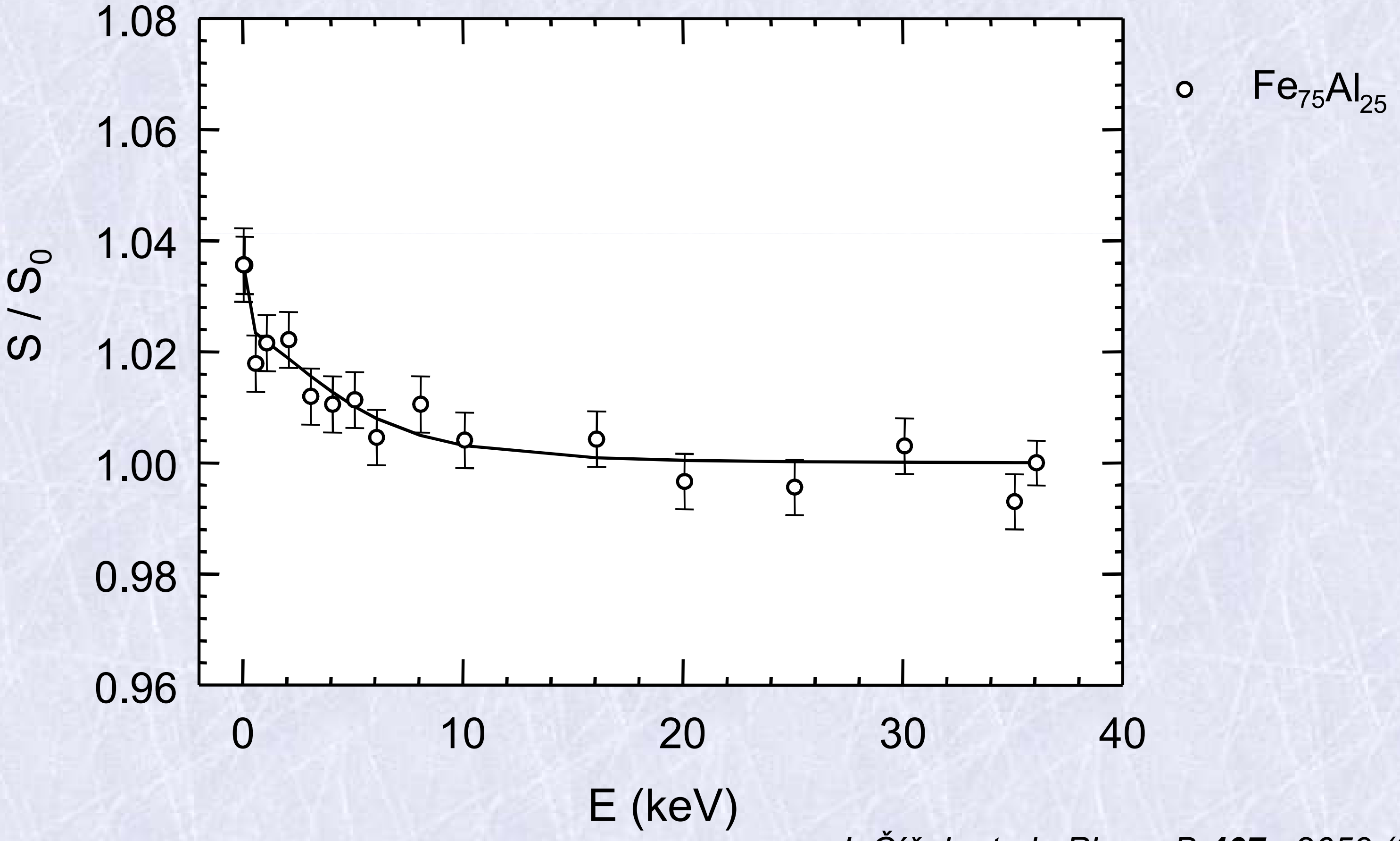




*J. Čížek et al., Phys. B **407** , 2659 (2012)*

Positron back-diffusion measurement

Fe-Al alloys quenched from 1000°C – dependence of S parameter on e⁺ energy



J. Čížek et al., Phys. B ***407*** *, 2659 (2012)*

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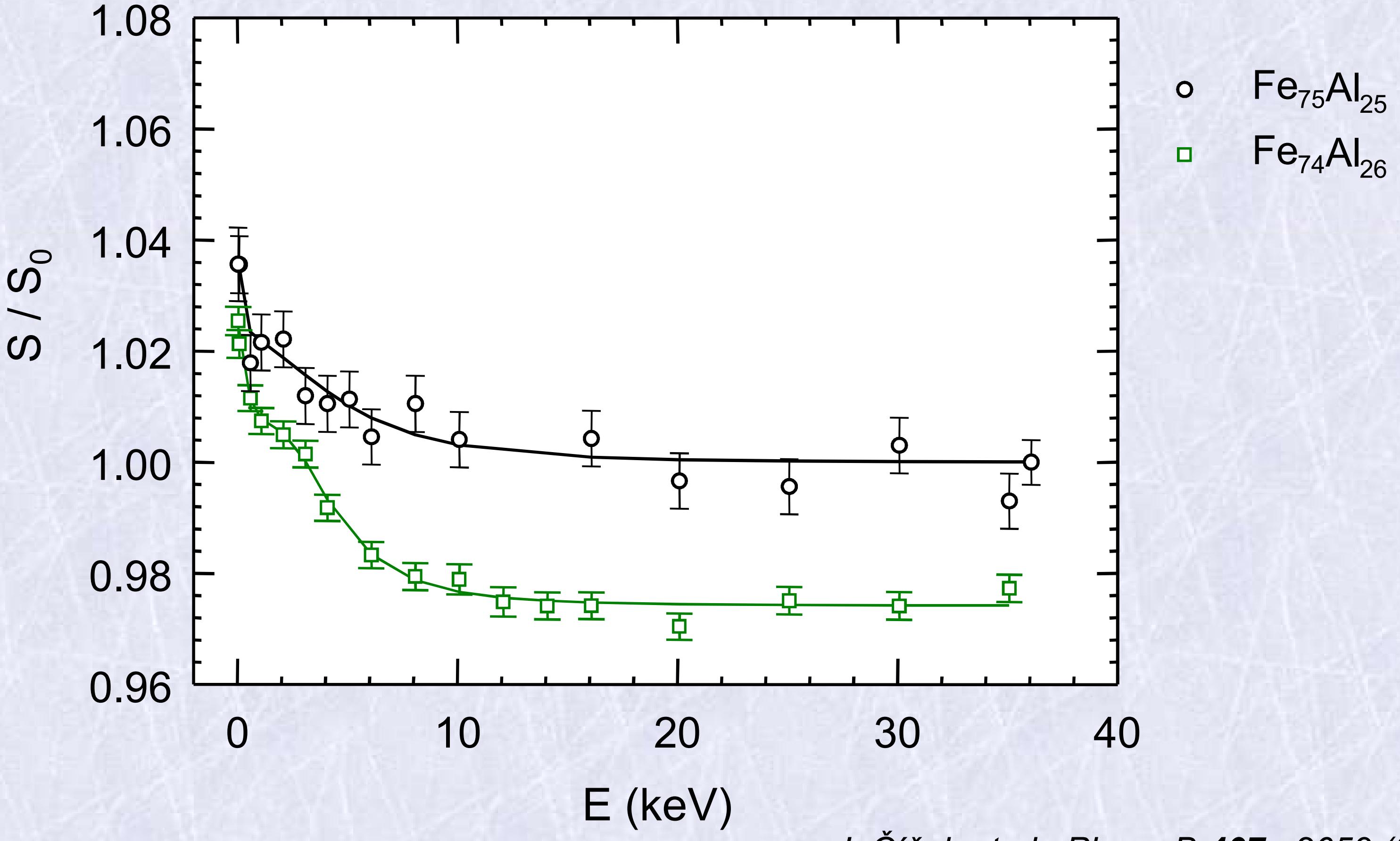

J. Čížek et al., Phys. B ***407*** *, 2659 (2012)*

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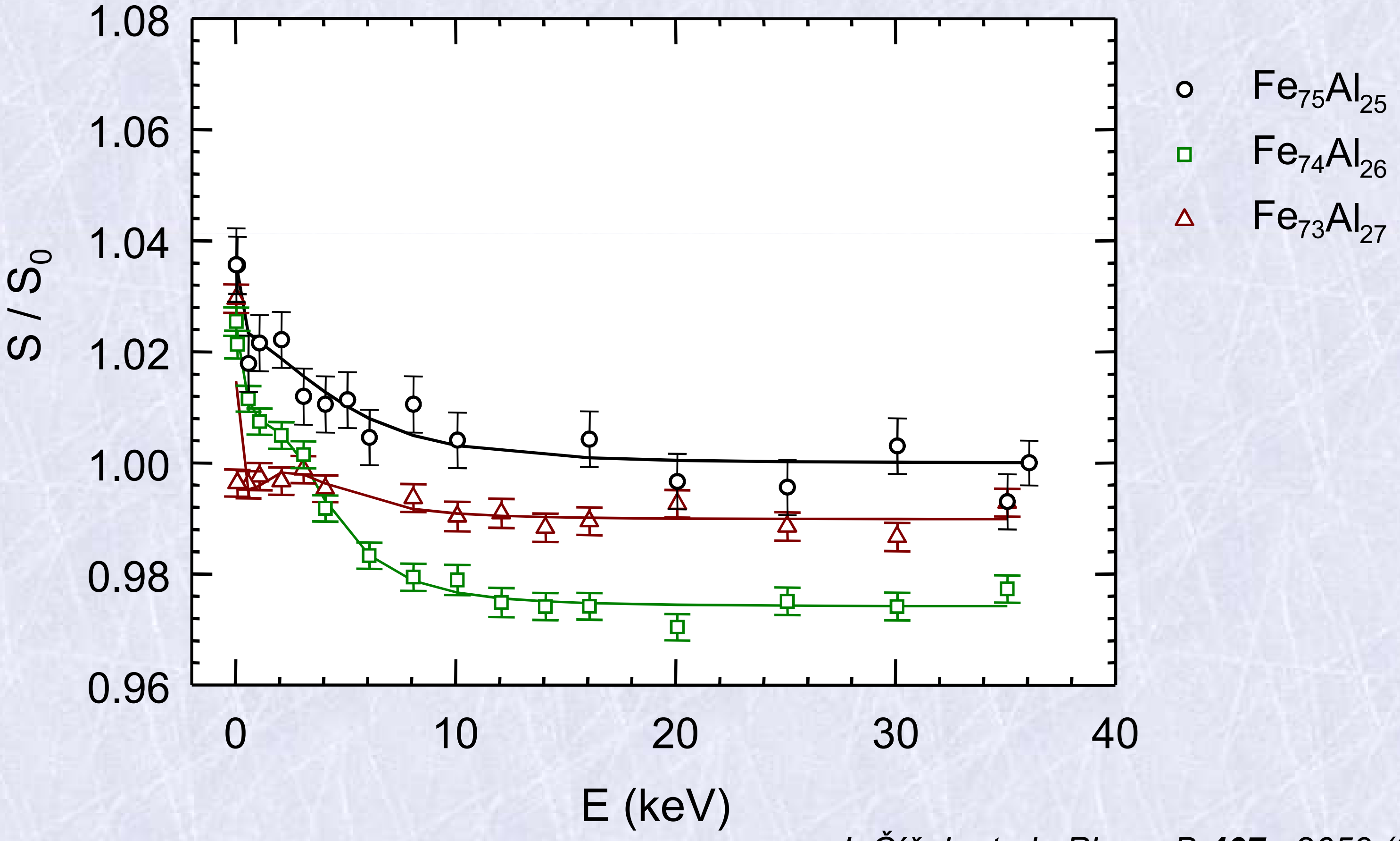

*J. Čížek et al., Phys. B **407** , 2659 (2012)*

Positron back-diffusion measurement

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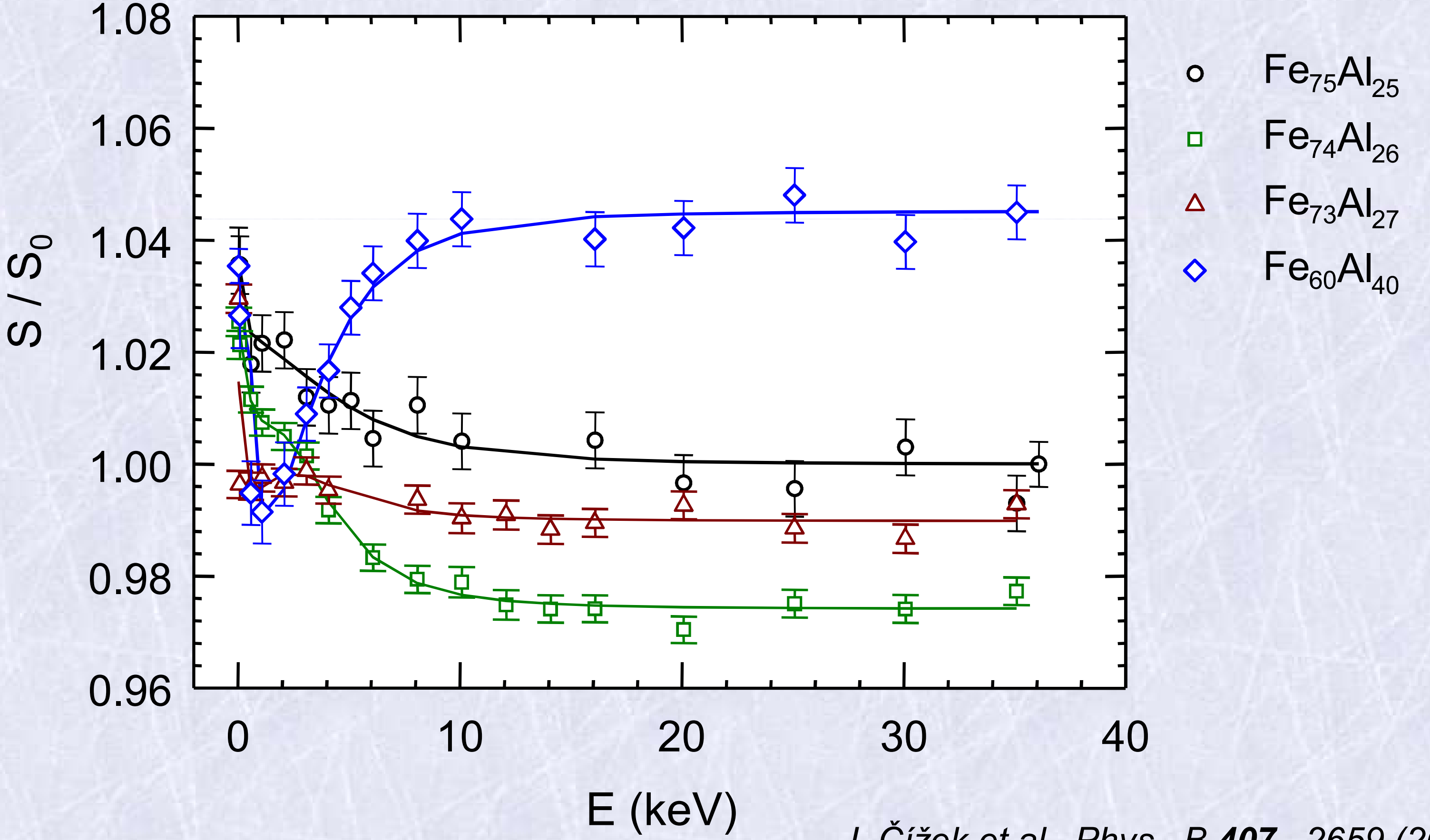

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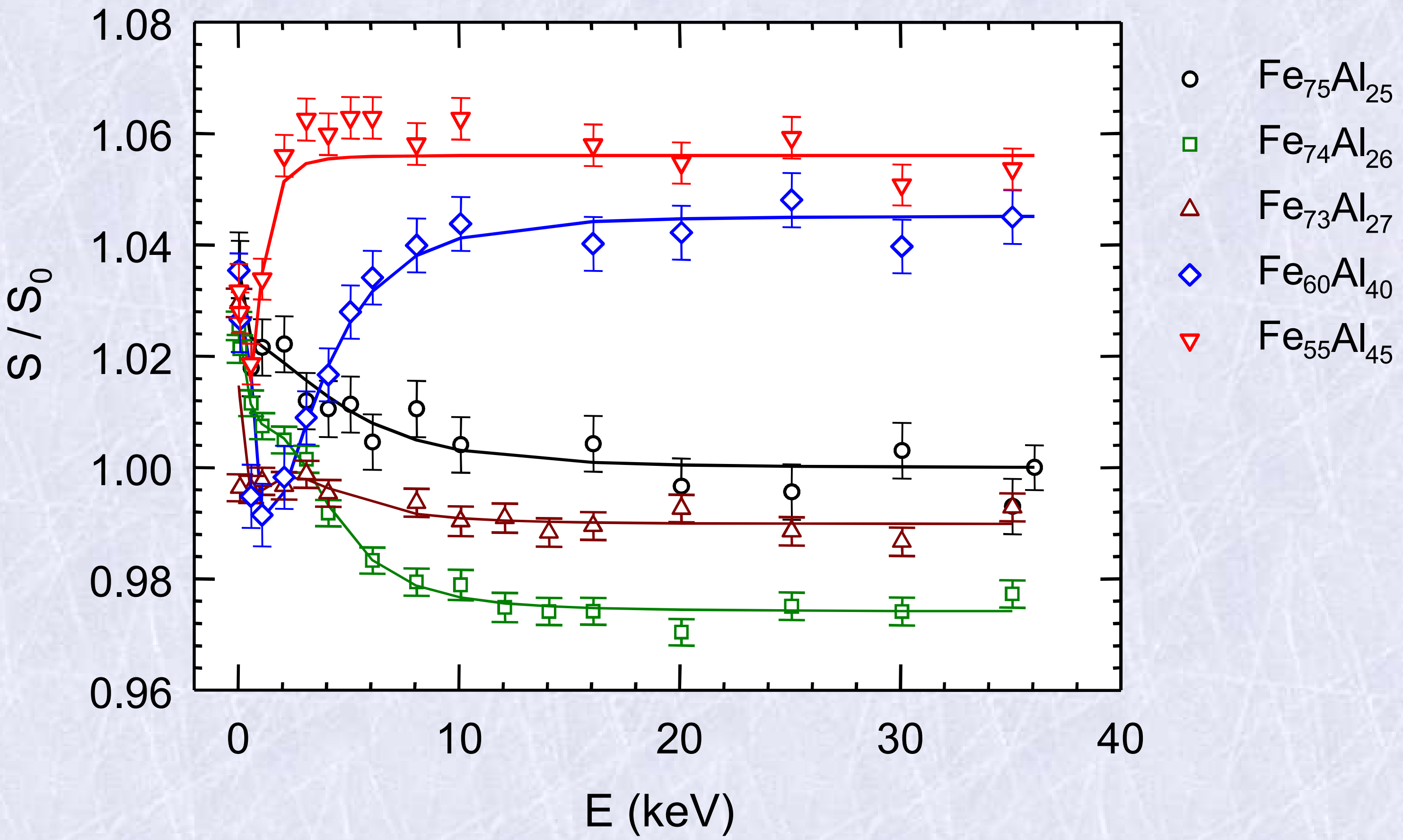
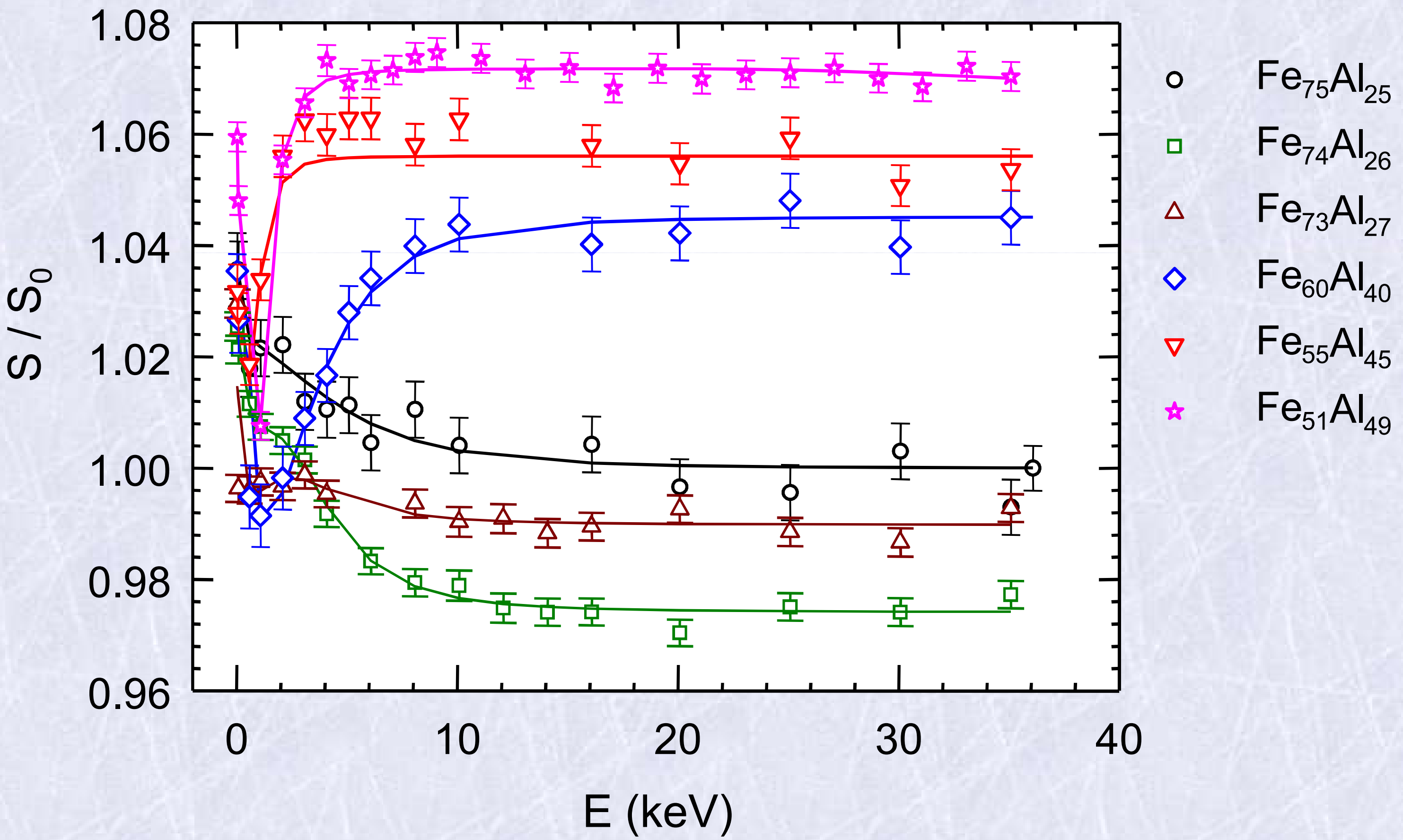


J. Čížek et al., Phys. B ***407*** *, 2659 (2012)*

Positron back-diffusion measurement

Fe-Al alloys quenched from 1000°C – dependence of S parameter on e⁺ energy

- solid lines – model curves calculated by VEPFIT
- two layered model: (i) surface oxide layer 15-20 nm (ii) bulk Fe-Al alloy



*J. Čížek et al., Phys. B **407** , 2659 (2012)*

Positron back-diffusion measurement

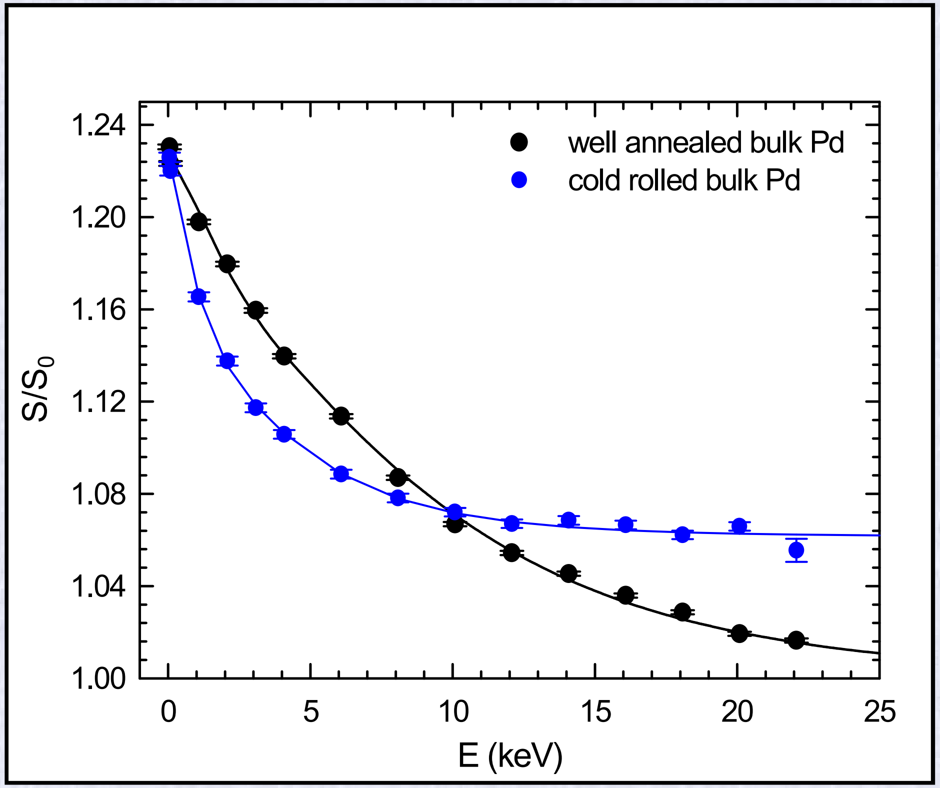
- positron diffusion length L_+
- perfect lattice: $L_{+,B} = \sqrt{D_+ \tau_B}$

D_+ - positron diffusion coefficient

τ_B – bulk positron lifetime

- presence of defects → shortening of L_+ $\quad L_+ = \sqrt{D_+ \tau_1}$

τ_1 – lifetime of the free positron component ($\tau_1 < \tau_B$)



Positron back-diffusion measurement

- positron diffusion length L_+

- perfect lattice: $L_{+,B} = \sqrt{D_+ \tau_B}$

D_+ - positron diffusion coefficient

τ_B – bulk positron lifetime

- presence of defects $\rightarrow$ shortening of L_+ $\quad L_+ = \sqrt{D_+ \tau_1}$

τ_1 – lifetime of the free positron component

- simple trapping model: $\tau_1 = \dfrac{1}{\dfrac{1}{\tau_B} + K}$

- positron trapping rate to defects: $K = \dfrac{1}{\tau_B}\left(\dfrac{L_{+,B}^2}{L_+^2} - 1\right)$

- net concentration of defects: $$c = \frac{K}{\nu} = \frac{1}{\nu \tau_B}\left(\frac{L_{+,B}^2}{L_+^2} - 1\right)$$

Positron back-diffusion measurement

- **positron lifetime measurement**

$$c = \frac{1}{\nu}\frac{I_2}{I_1}\left(\frac{1}{\tau_B} - \frac{1}{\tau_2}\right) = \frac{1}{\nu} I_2 \left(\frac{1}{\tau_1} - \frac{1}{\tau_2}\right)$$

- It is hard to resolve free positron component when $I_1 < 5\%$ (saturated trapping)

- in FeAl alloys it corresponds $c > 2 \times 10^{-4} = 200$ ppm

- **positron back-diffusion measurement**

$$c = \frac{K}{\nu} = \frac{1}{\nu \tau_B}\left(\frac{L_{+,B}^2}{L_+^2} - 1\right)$$

- It is hard to determine positron diffusion length when $L_+ < 1$ nm

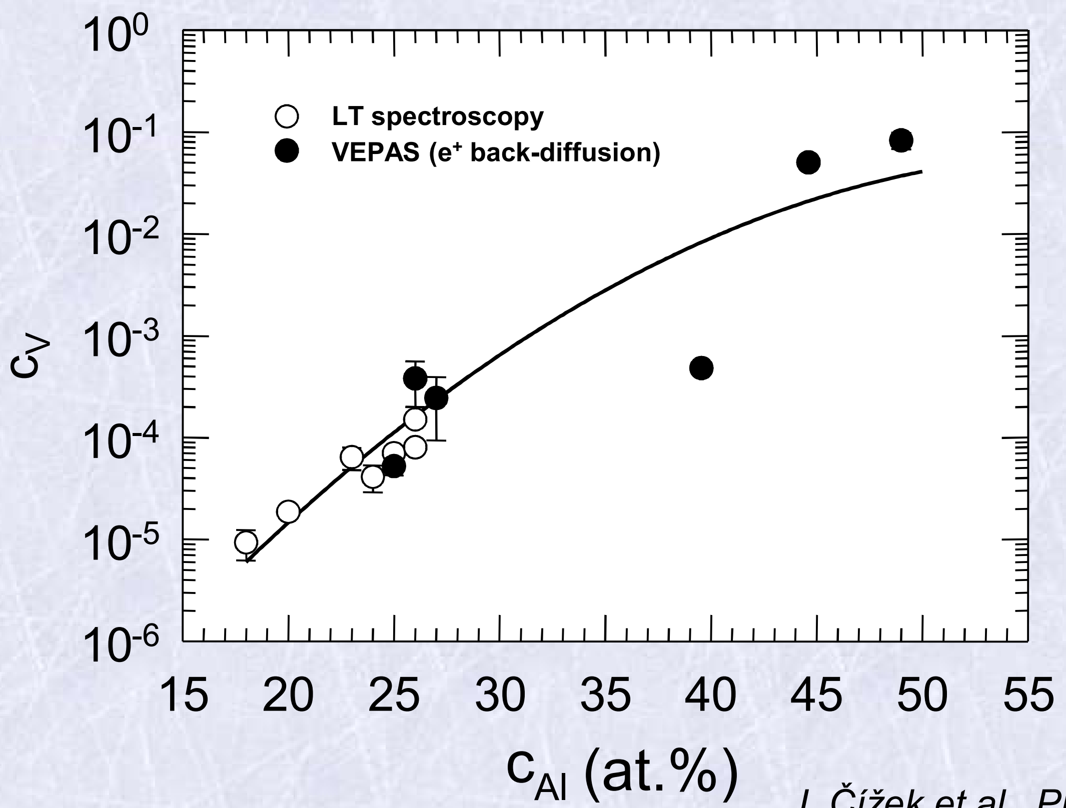
- in FeAl alloys it corresponds $c > 1 \times 10^{-3} = 10$ at.% $= 10^5$ ppm

- positron back-diffusion measurement can be used for determination of defect concentration when LT spectroscopy cannot be used because of saturated trapping

Concentration of quenched-in vacancies

Fe-Al alloys quenched from 1000°C

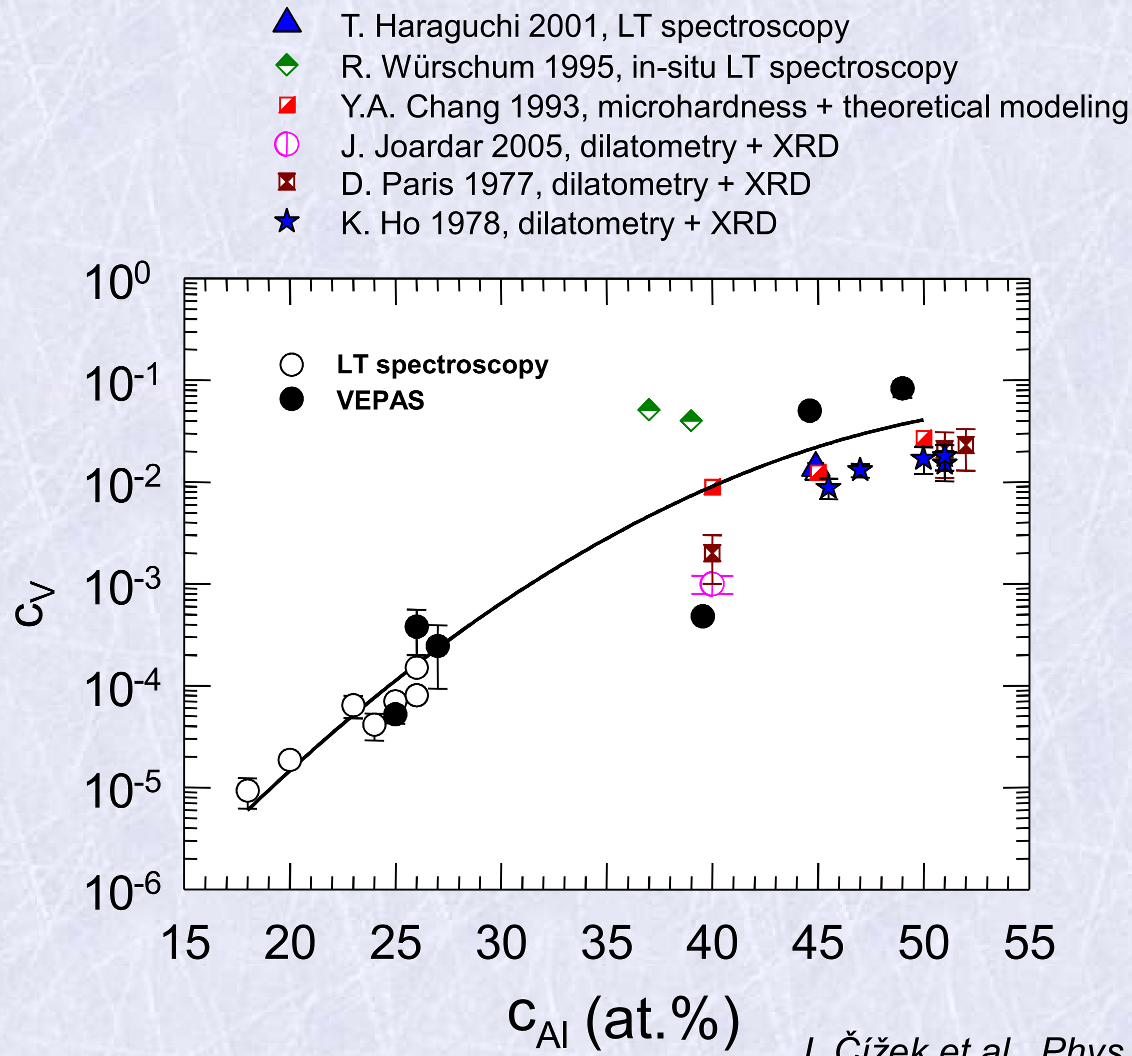
- LT spectroscopy and VEPAS gave mutually consistent results
- $Fe_{75}Al_{25}$ alloy: LT spectroscopy: $c_V = (7.0 \pm 0.5) \times 10^{-5}$
  VEPAS (e^+ back-diffusion): $c_V = (5 \pm 1) \times 10^{-5}$



*J. Čížek et al., Phys. B **407** , 2659 (2012)*

Concentration of quenched-in vacancies

Fe-Al alloys quenched from 1000°C



J. Čížek et al., Phys. B ***407*** *, 2659 (2012)*

Vacancies

$$c_{1V} = \frac{K_{1V}}{\nu_{1V}}$$

- calculated specific e^+ trapping rates for vacancies

Z — $_3$Li
304 — bulk positron lifetime τ_B (ps)
341 — lifetime τ_V of positron trapped at vacancy (ps)
-0.45 — positron binding energy to vacancy (eV)
1.217 — specific positron trapping rate to vacancy (10^{14} at.s^{-1})

$E_B = E_0 - E_V$

E_0 – ground state energy of delocalized positron

E_V – ground state energy of positron trapped at vacancy

Fermi golden rule

$$\nu \approx T_{0\to 0} = \frac{2\pi}{\eta} \left| \left\langle \psi_{0,0} \left| H' \right| \psi_{V,0} \right\rangle \right|^2$$

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1	$_{1}$H																	$_{2}$He
2	$_{3}$Li 304 341 -0.45 1.217	$_{4}$Be											$_{5}$B	$_{6}$C	$_{7}$N	$_{8}$O	$_{9}$F	$_{10}$Ne
3	$_{11}$Na 328 365 -0.20 0.362	$_{12}$Mg											$_{13}$Al 168 244 -2.13 3.139	$_{14}$Si	$_{15}$P	$_{16}$S	$_{17}$Cl	$_{18}$Ar
4	$_{19}$K 374 414 -0.31 0.177	$_{20}$Ca	$_{21}$Sc	$_{22}$Ti	$_{23}$V 117 200 -4.01 6.077	$_{24}$Cr 107 184 -3.11 6.247	$_{25}$Mn	$_{26}$Fe 102 177 -3.29 6.779	$_{27}$Co	$_{28}$Ni 97 166 -2.74 6.156	$_{29}$Cu 109 170 -1.24 3.817	$_{30}$Zn	$_{31}$Ga	$_{32}$Ge	$_{33}$As	$_{34}$Se	$_{35}$Br	$_{36}$Kr
5	$_{37}$Rb 376 419 -0.35 0.155	$_{38}$Sr 310 387 -0.97 0.407	$_{39}$Y	$_{40}$Zr	$_{41}$Nb 126 222 -3.65 4.103	$_{42}$Mo 108 199 -3.94 5.150	$_{43}$Tc	$_{44}$Ru	$_{45}$Rh 97 179 -2.95 4.677	$_{46}$Pd 107 173 -1.11 2.634	$_{47}$Ag 125 200 -1.16 2.215	$_{48}$Cd	$_{49}$In	$_{50}$Sn	$_{51}$Sb	$_{52}$Te	$_{53}$I	$_{54}$Xe
6	$_{55}$Cs 392 431 -0.42 0.113	$_{56}$Ba 305 393 -1.39 0.461	$_{71}$Lu	$_{72}$Hf	$_{73}$Ta 118 217 -4.19 4.392	$_{74}$W	$_{75}$Re	$_{76}$Os	$_{77}$Ir 90 175 -4.03 5.301	$_{78}$Pt 98 181 -2.48 3.738	$_{79}$Au 112 191 -1.51 2.496	$_{80}$Hg	$_{81}$Tl	$_{82}$Pb 191 277 -0.79 0.841	$_{83}$Bi	$_{84}$Po	$_{85}$At	$_{86}$Rn
7	$_{87}$Fr	$_{88}$Ra	$_{103}$Lr	$_{104}$Rf	$_{105}$Db	$_{107}$Sg	$_{107}$Bh											
			$_{57}$La	$_{58}$Ce	$_{59}$Pr	$_{60}$Nd	$_{61}$Pm	$_{62}$Sm	$_{63}$Eu 270 364 -1.28 0.638	$_{64}$Gd	$_{65}$Tb	$_{66}$Dy	$_{67}$Ho	$_{68}$Er	$_{69}$Tm	$_{70}$Yb		
			$_{89}$Ac	$_{90}$Th	$_{91}$Pa	$_{92}$U	$_{93}$Np	$_{94}$Pu	$_{95}$Am	$_{96}$Cm	$_{97}$Bk	$_{98}$Cf	$_{99}$Es	$_{100}$Fm	$_{101}$Md	$_{102}$No		

Vacancies

• calculated specific positron trapping rates: comparison with experiment

element	calculated (LDA) ν (10^{14}) at.s^{-1}	experimental ν (10^{14}) at.s^{-1}	method	Reference
Al	3.139	2.5 ± 1.5 2.5 ± 1.0 3 ± 2	thermal vac. thermal vac. thermal vac.	E. Gramsch, K.G. Lynn , *Phys. Rev. B* **40** (1989) 2537 J. A. Jackman, G. M. Hood and R. J. Schultz, *J. Phys. F* **17** (1987) 1817 T.M. Hall, A.N. Goland, C.N. Snead Jr., *Phys. Rev. B* **10** (1974) 3062.
Fe	6.779	11 ± 2	e^- irrad.	A. Vehanen, P. Hautojärvi, J. Johansson, J. Yli-Kauppila, P. Moser, *Phys. Rev. B* **25** (1982) 762
Cu	3.817	1.4 ± 0.2	$\Delta l/l - \Delta a/a$	J.-E. Kluin, Th. Hehenkamp, *Phys. Rev. B* **44** (1991) 11597
Ag	2.215	2.3 ± 0.2	$\Delta l/l - \Delta a/a$	J. Wolff, J.-E. Kluin, Th. Hehenkamp, *Mater. Sci. Forum* **105-110** (1992) 1329
Au	2.496	3.5 ± 1.0 2.9 ± 0.8	thermal vac. thermal vac.	T.M. Hall, A.N. Goland, C.N. Snead Jr., *Phys. Rev. B* **10** (1974) 3062 T.M. Hall, A.N. Goland, K.C. Jain, R.W. Siegel, *Phys. Rev. B* **12** (1975) 1613

• Thermal vacancies

$$c_V = \exp\left(\frac{S_f}{k}\right)\exp\left(-\frac{H_f}{kT}\right)$$

H_f - vacancy formation enthalpy

S_f - vacancy formation entropy $S_f = (2\text{-}3)\,k$

k - Boltzmann constant

Vacancies

• calculated specific positron trapping rates: comparison with experiment

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• simultaneous measurement of length change $\Delta l \,/\, l$ (differential dilatometry) and change of the lattice parameter $\Delta a \,/\, a$ (X-ray diffraction)

$$c_v = 3\left(\frac{\Delta l}{l} - \frac{\Delta a}{a}\right)$$

Vacancies

• calculated specific positron trapping rates: comparison with experiment

element	calculated (LDA) ν (10^{14}) at.s^{-1}	experimental ν (10^{14}) at.s^{-1}	method	Reference
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		2.5 ± 1.0	thermal vac.	J. A. Jackman, G. M. Hood and R. J. Schultz, *J. Phys. F* **17** (1987) 1817
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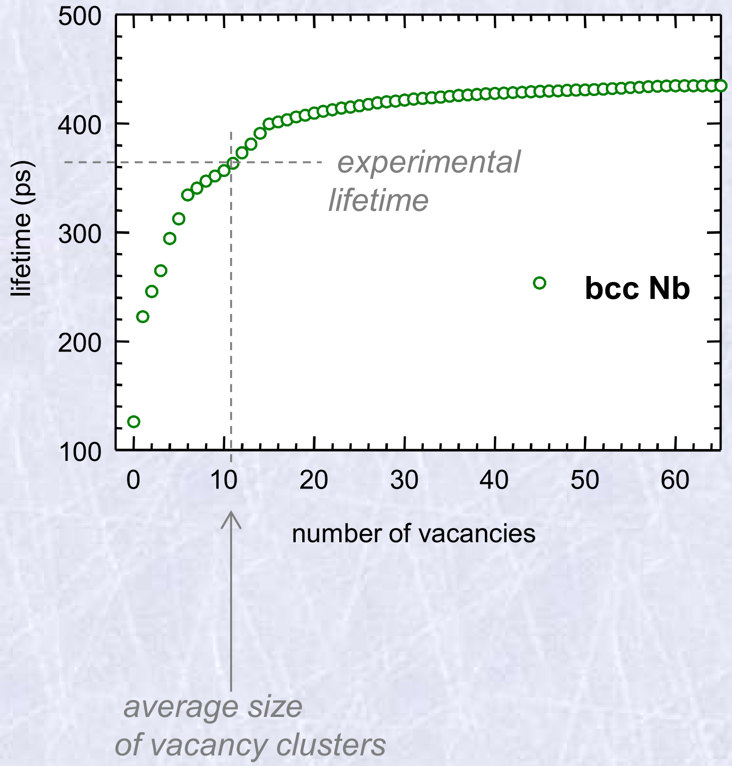
• **specific trapping rate for vacancies in metals: ν_V = 10^{14}-10^{15} at.s^{-1}**

vacancy clusters

- vacancy clusters
- *ab-inito* calculations of positron lifetimes

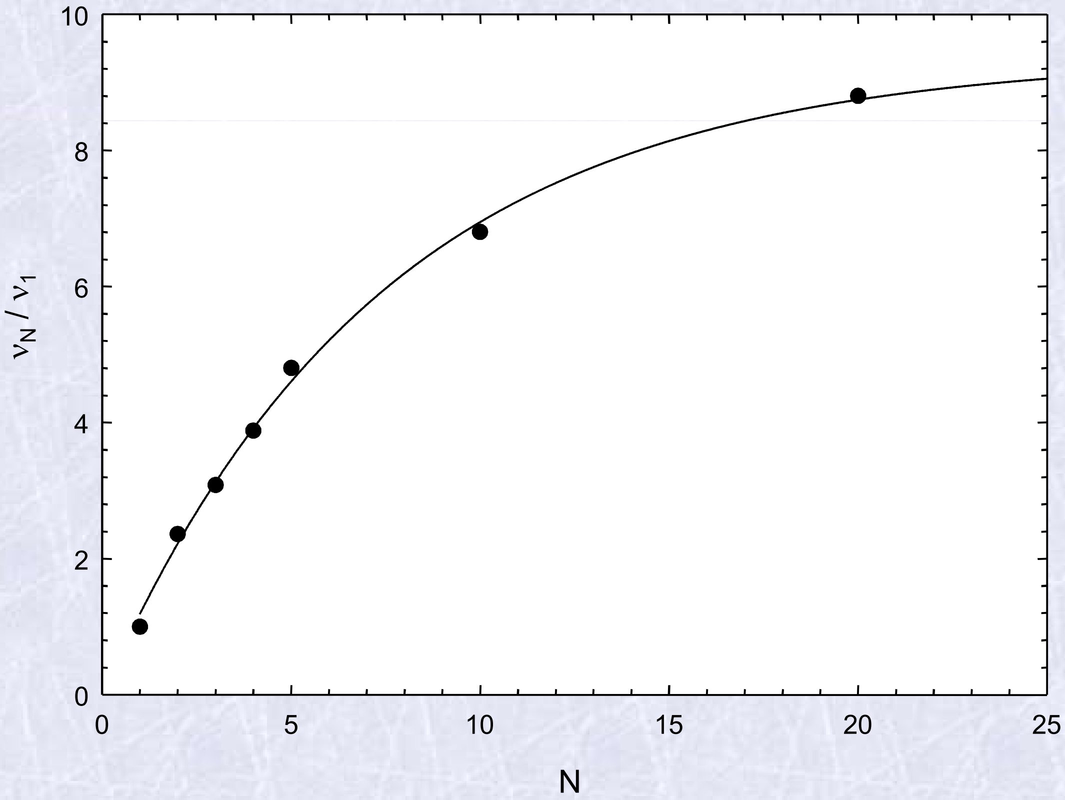


J. Čížek et al., J. Phys, Conf. Ser. ***443*** *, 012008 (2013)*

vacancy clusters

- specific positron trapping rates for vacancy clusters
- small clusters ($N \leq 10$): $\nu_N \sim N$
- larger clusters ($N > 10$): ν_N gradually saturates

R. M. Nieminen, J. Laakkonen, Appl. Phys.20, 181 (1979)



$$\nu_N / \nu_1 \approx a\left(1 - e^{-bN}\right)$$

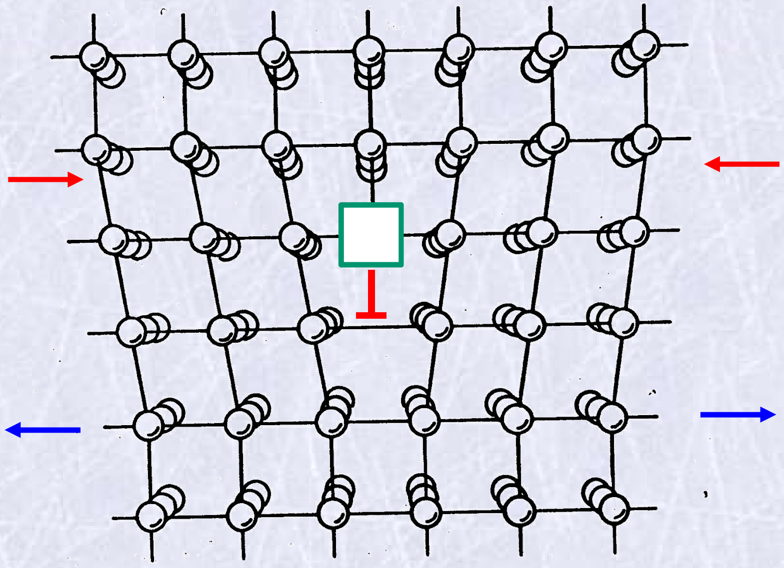
$$a = 9.4$$

$$b = 0.13$$

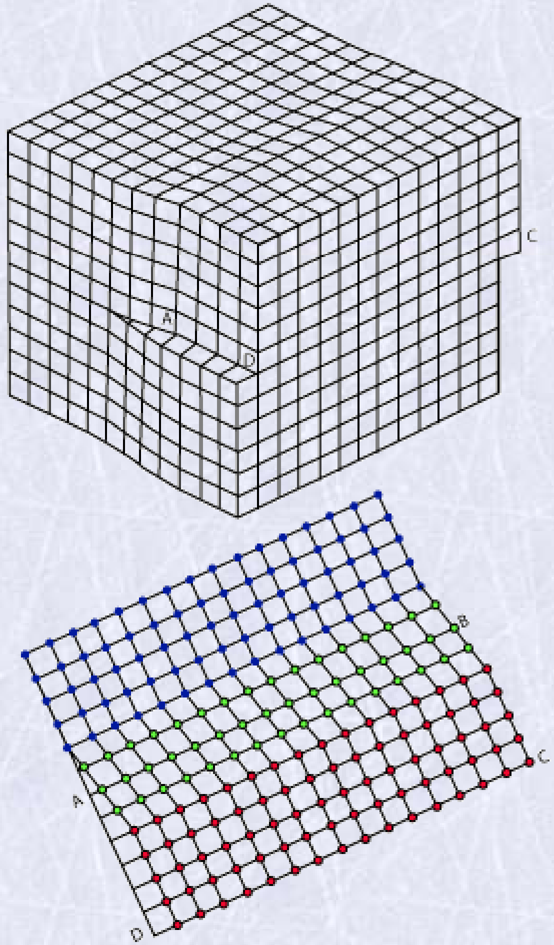
Dislocations

- 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

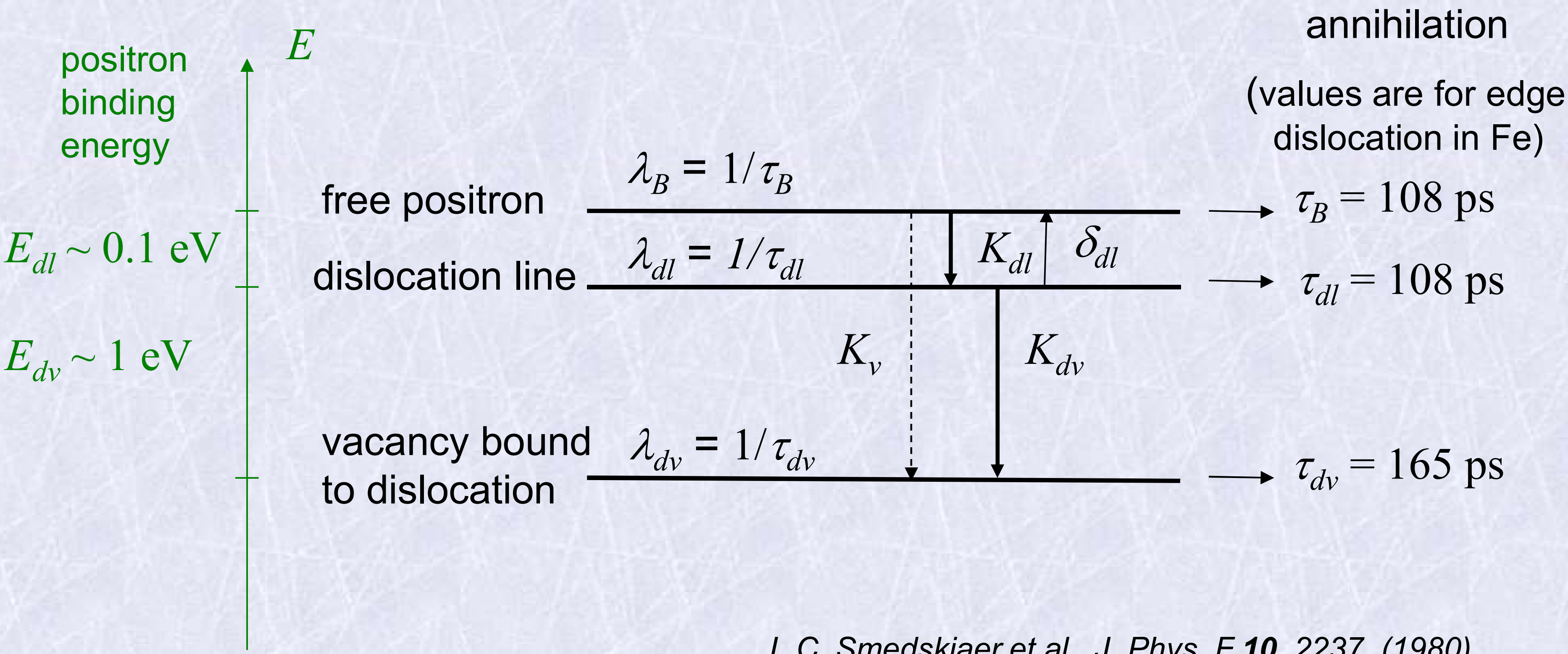


Positron trapping at dislocations

• 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)



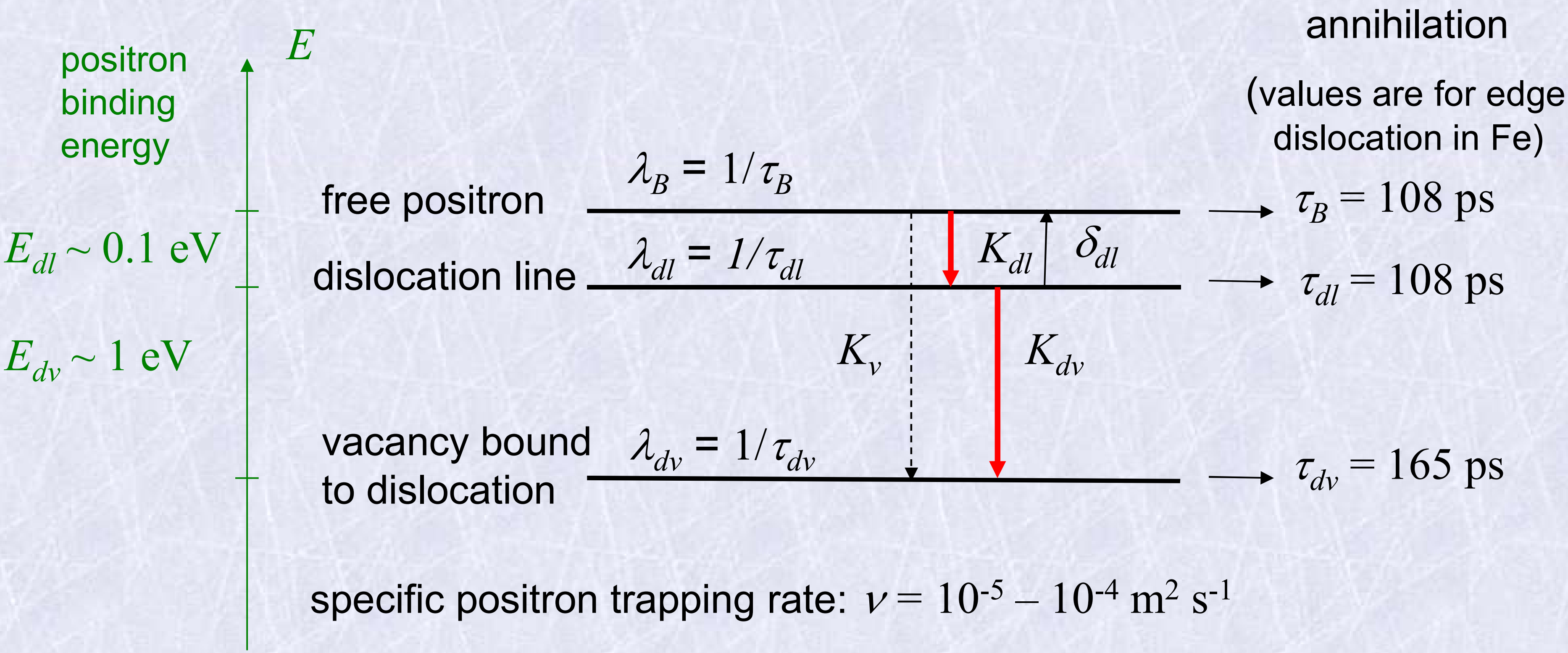
L.C. Smedskjaer et al., J. Phys. F ***10****, 2237, (1980)*

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)



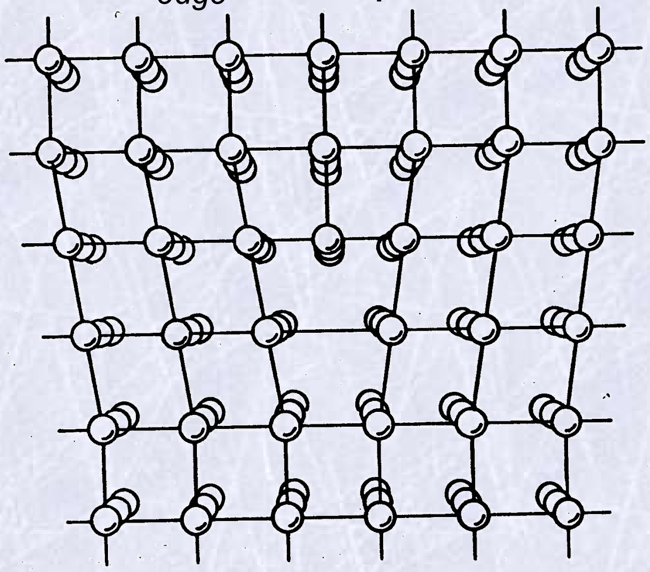
specific positron trapping rate: $\nu = 10^{-5} - 10^{-4}$ m^2 s^{-1}

dislocation density: $\rho = 10^{12} - 10^{15}$ m^{-2}

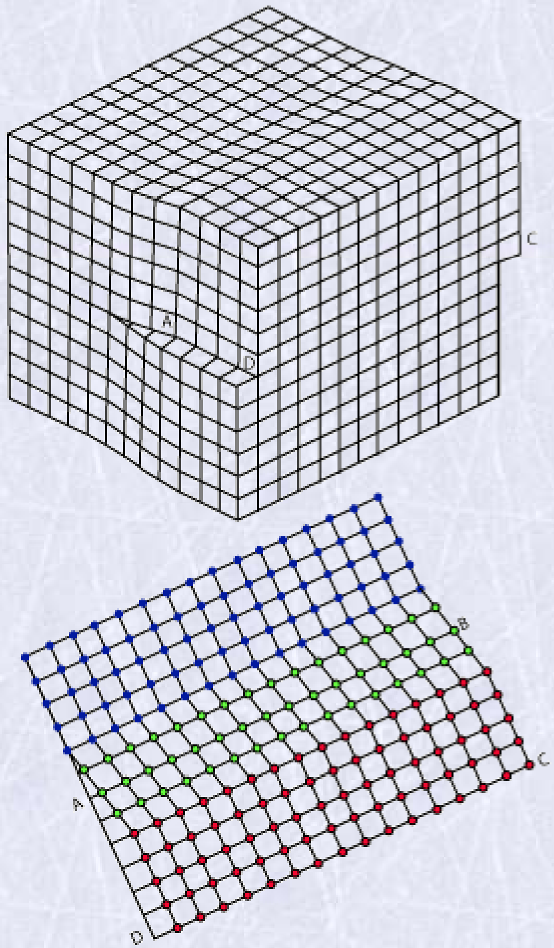
Positron trapping at dislocations in Fe (or steels)

- 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



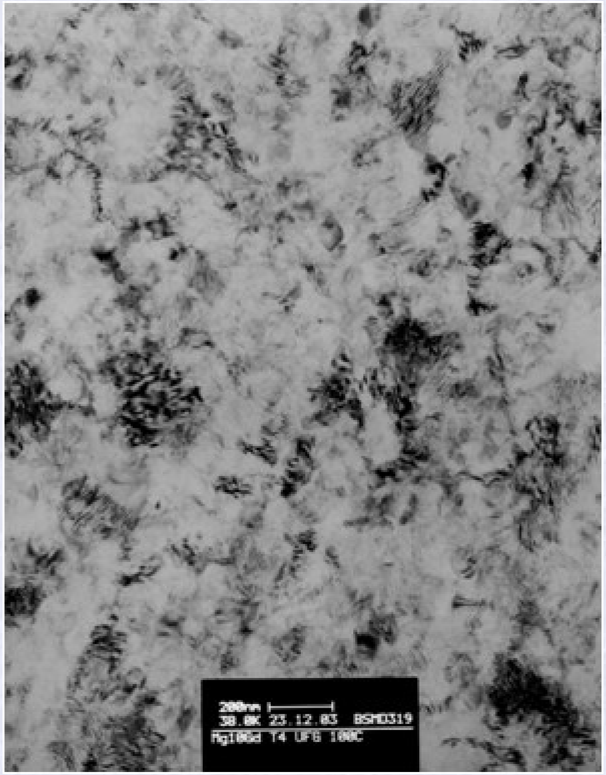
Positron trapping at dislocations – distribution of dislocations

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

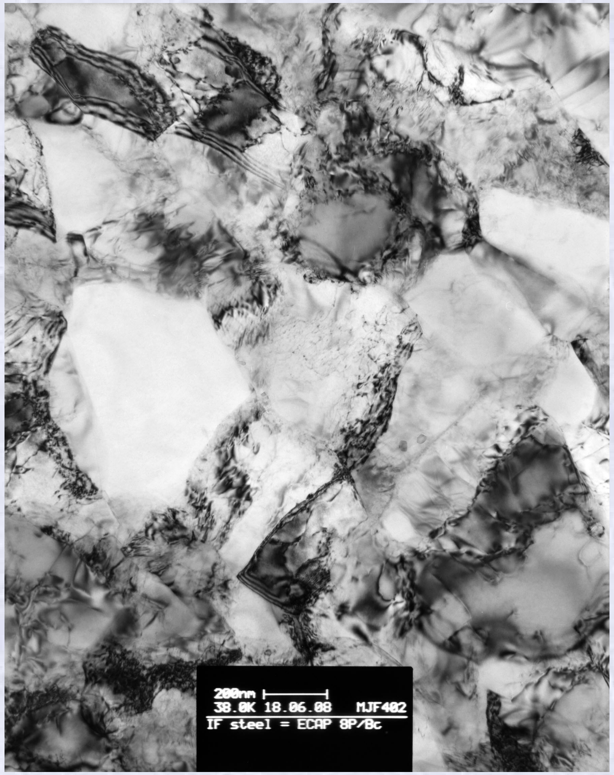
$$\rho = \frac{1}{\nu}\frac{I_2}{I_1}\left(\frac{1}{\tau_B} - \frac{1}{\tau_D}\right)$$

ν – 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



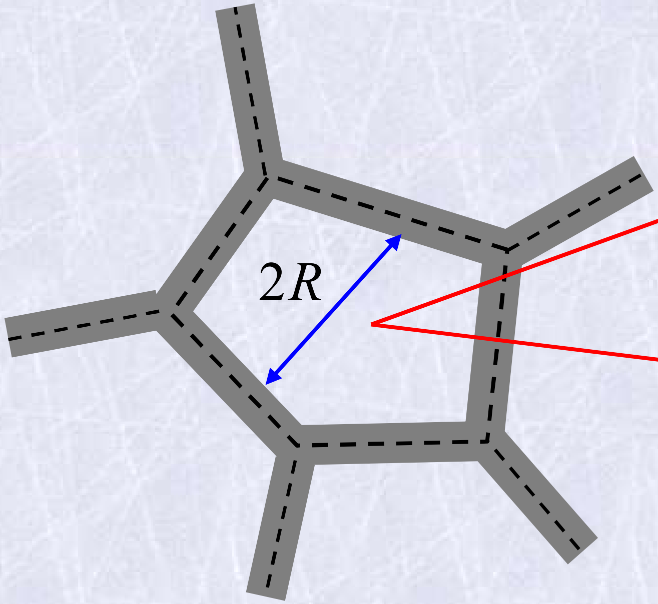
HPT-deformed IF steel

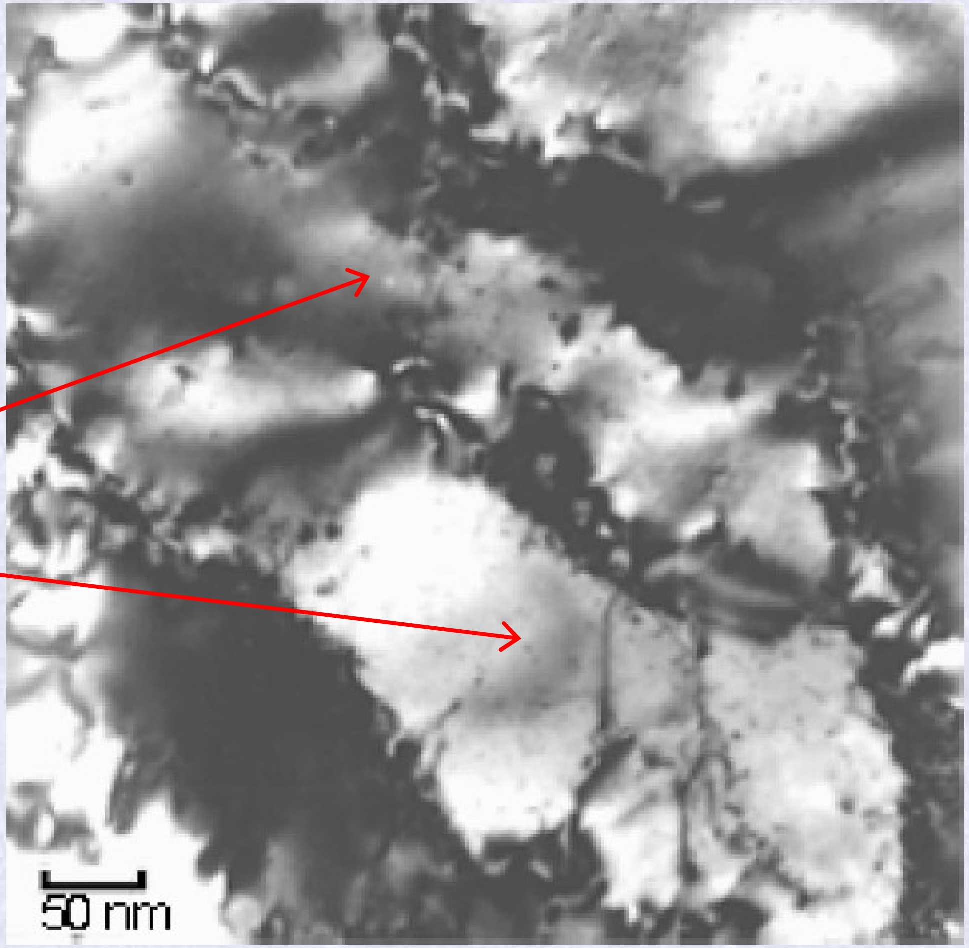
Positron trapping at dislocations – dislocation cell structure

• dislocation cell structure

• dislocation-free cell interiors

• distorted regions with high density of dislocations (dislocation walls)





*J. Čížek et al. Phys. Rev. B **65**, 094106 (2002)*

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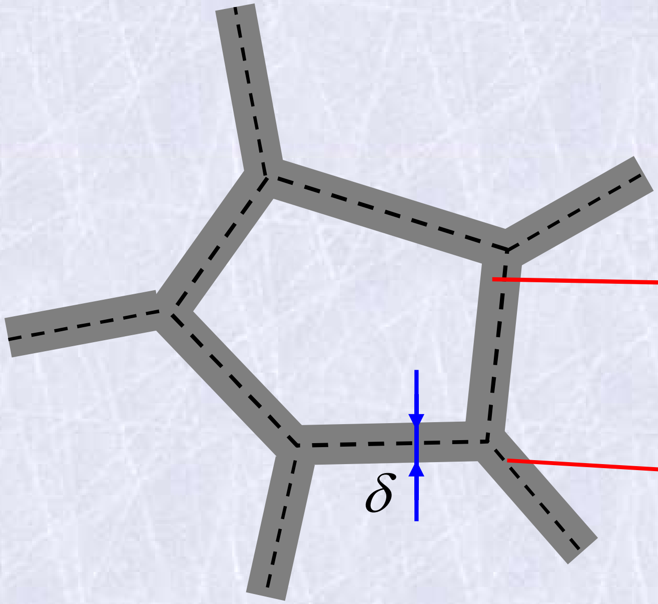
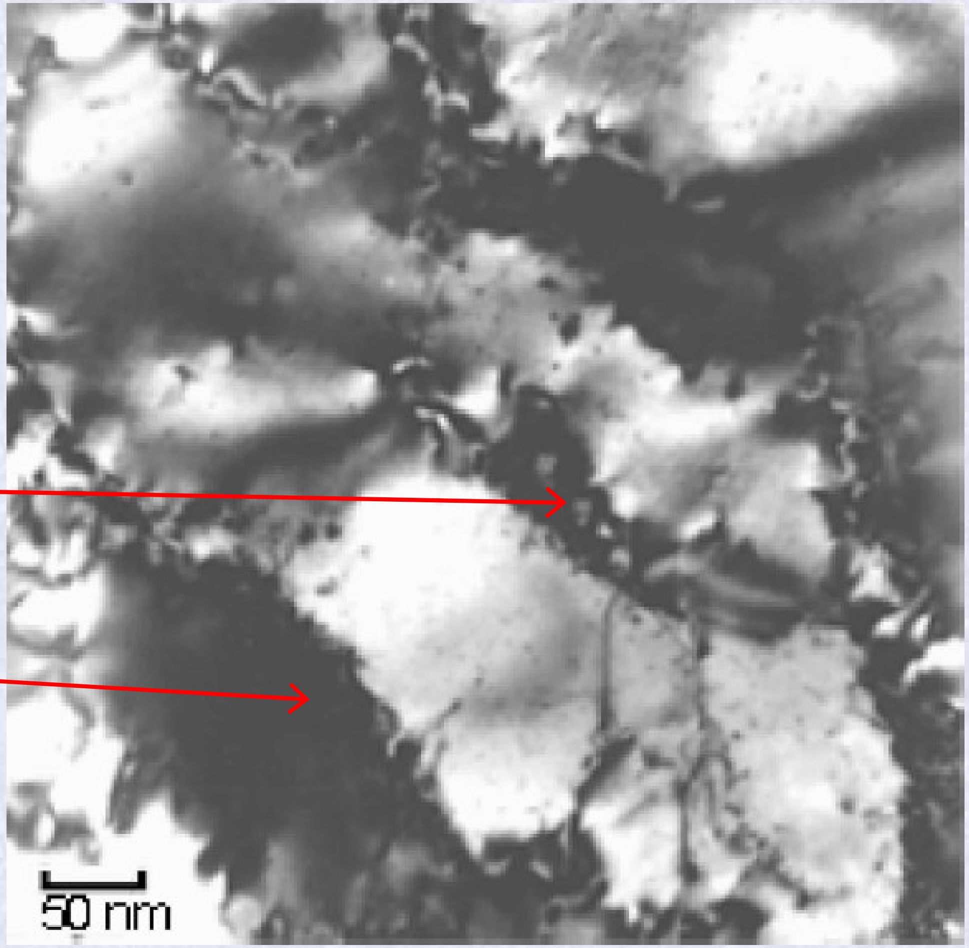
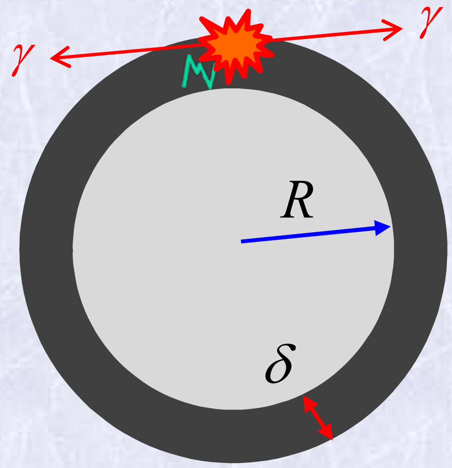




J. Čížek et al. Phys. Rev. B ***65****, 094106 (2002)*

Diffusion trapping model (DTM)



- dislocation-free spherical with radius R
- surrounded by dislocation walls with thickness δ

thermalization

↓

1. e^+ stopped in dislocation walls ⟶ trapping at dislocations

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

J. Čížek et al. PRB ***65****, 094106 (2002)*

Diffusion trapping model (DTM)

- dislocation-free spherical with radius R
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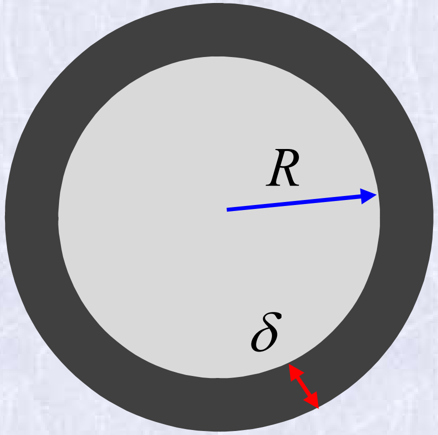


thermalization

↓

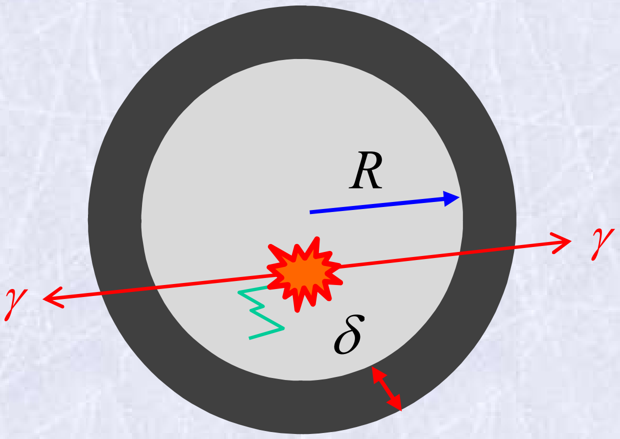
1. e^+ stopped in dislocation walls ⟶ trapping at dislocations

2. e^+ stopped inside cells

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

J. Čížek et al. PRB ***65****, 094106 (2002)*

Diffusion trapping model (DTM)



- dislocation-free spherical with radius R
- surrounded by dislocation walls with thickness δ

thermalization

↓

1. e^+ stopped in dislocation walls ⟶ trapping at dislocations

annihilation in
delocalized
state

↖

2. e^+ stopped inside cells

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

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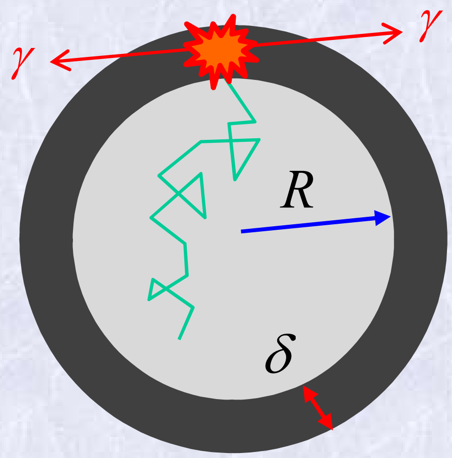
Diffusion trapping model (DTM)



- dislocation-free spherical with radius R
- surrounded by dislocation walls with thickness δ

thermalization

↓

1. e^+ stopped in dislocation walls → 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 dislocation walls

annihilation in delocalized state

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

boundary condition

2. e^+ stopped inside cells

D_+ - positron diffusion coefficient

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

initial condition

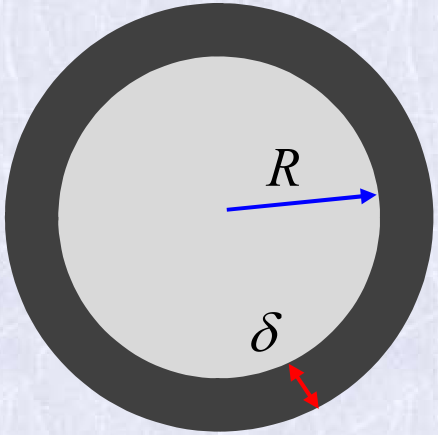
$$\eta = \frac{(R+\delta)^3 - R^3}{(R+\delta)^3}$$

volume fraction of dislocation walls

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

J. Čížek et al. PRB ***65****, 094106 (2002)*

Diffusion trapping model (DTM)



- dislocation-free spherical with radius R
- surrounded by dislocation walls 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

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

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

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

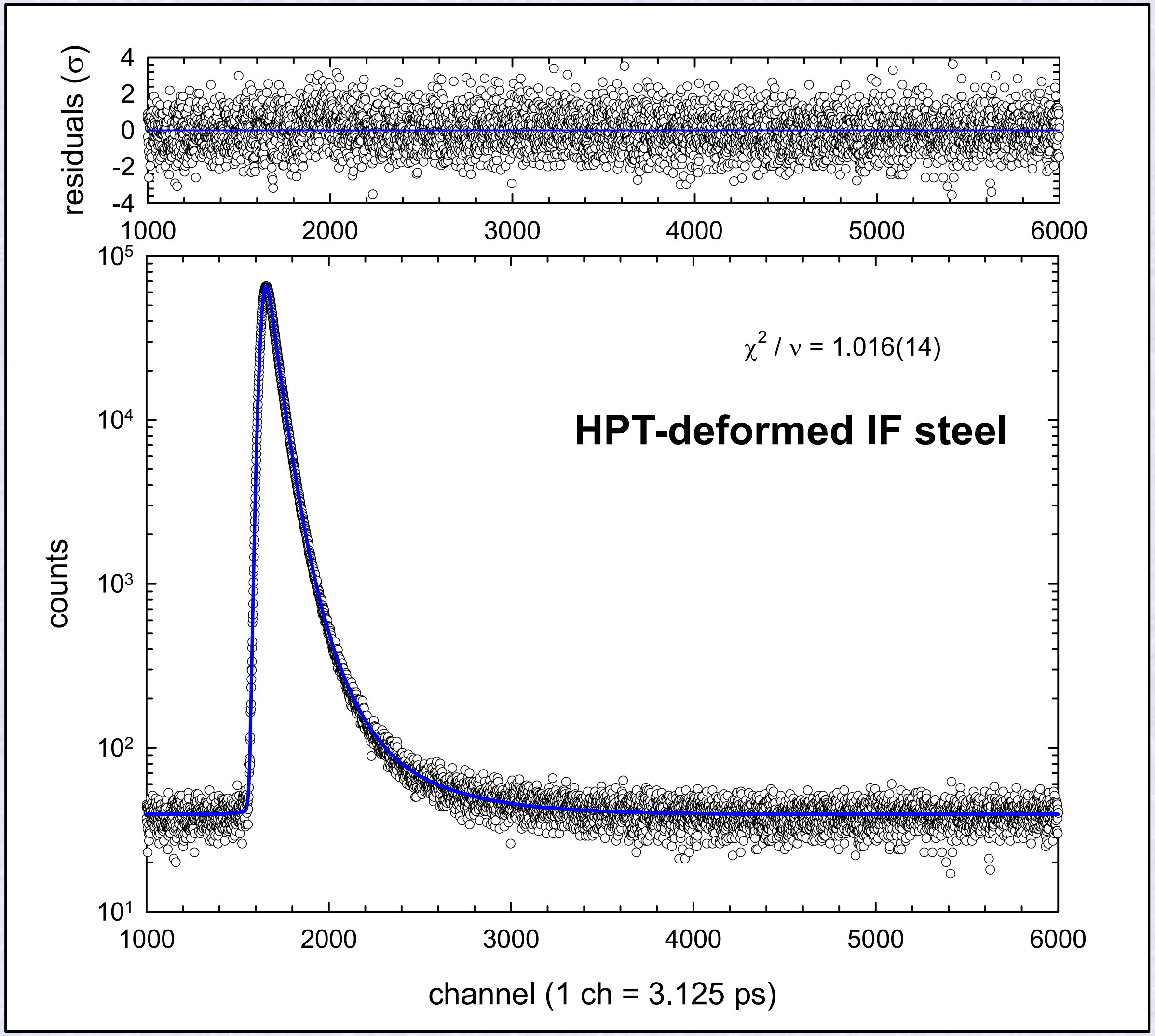
*A. Dupasquier et al. PRB **48**, 9235 (1993)*

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

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

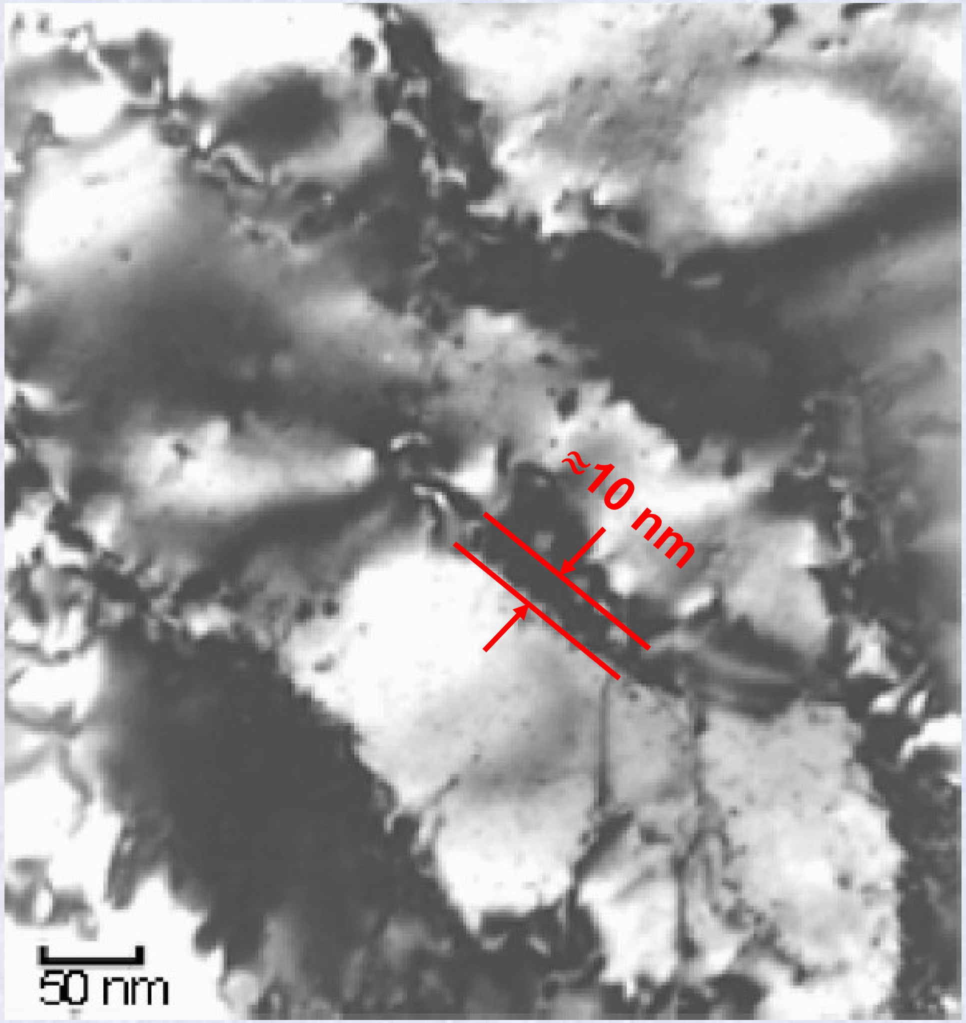
- bulk positron lifetime for Fe τ_B = 108 ps

*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)*

HPT-deformed IF steel



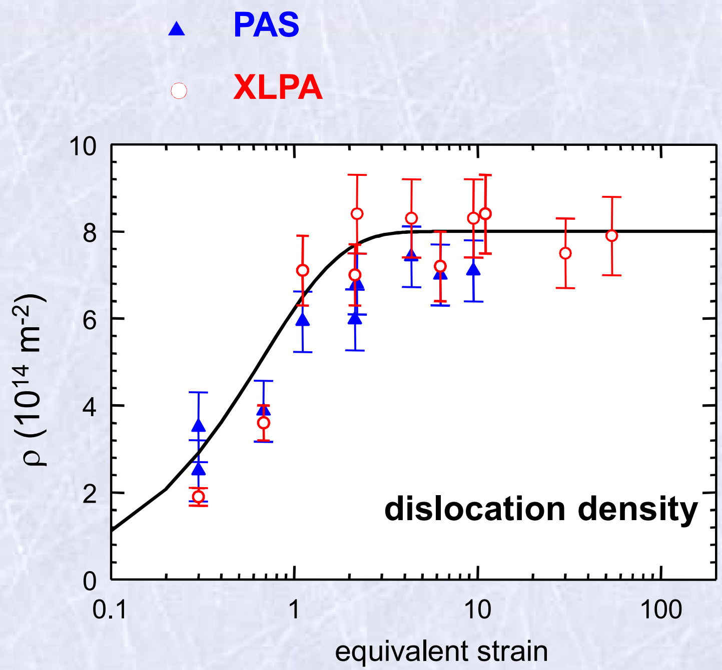
Diffusion trapping model (DTM)

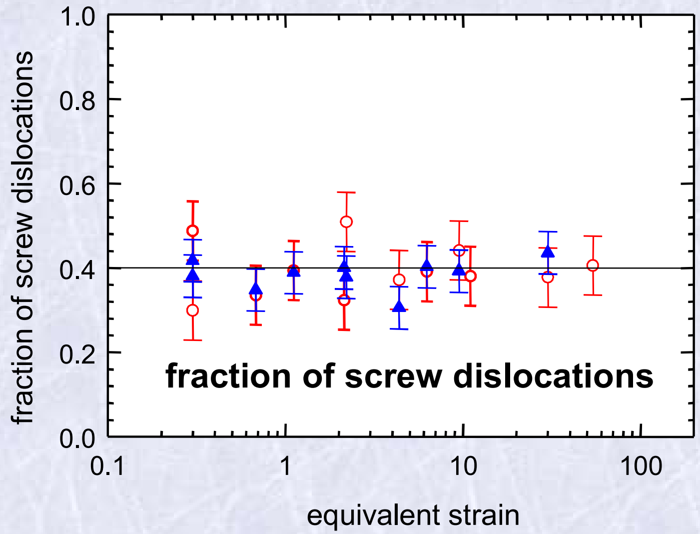
HPT-deformed steel

- good agreement of PAS and (XLPA = X-ray line profile analysis)

- dislocation density increases with deformation and saturates at strain $e \geq 3$

- edge character of dislocations prevails



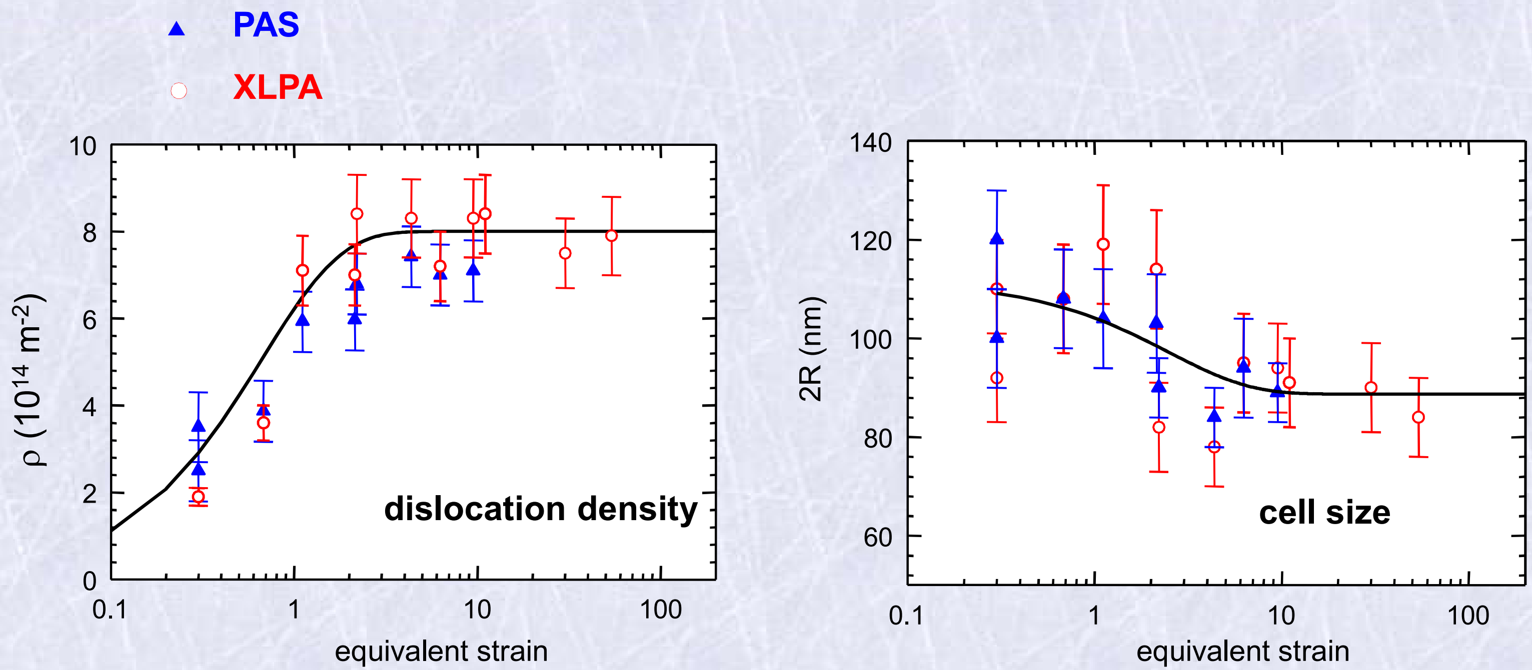


J. Čížek et al. Acta Mater. ***107****, 83 (2016)*

Diffusion trapping model (DTM)

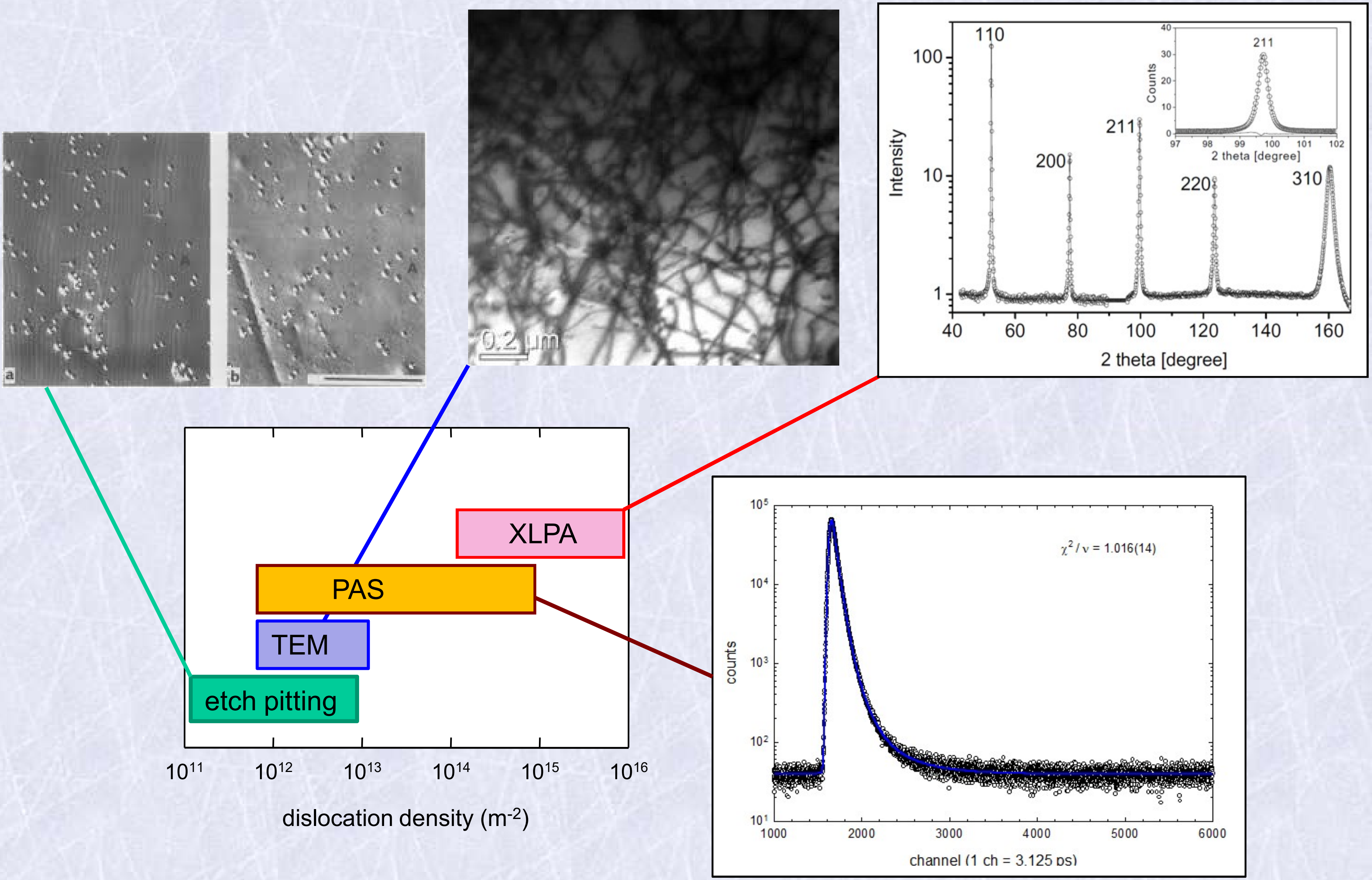
HPT-deformed steel

- good agreement of PAS and (XLPA = X-ray line profile analysis)
- dislocation density increases with deformation and saturates at strain $e \geq 3$
- edge character of dislocations prevails
- cell size saturates at $e > 10$



J. Čížek et al. Acta Mater. ***107****, 83 (2016)*

Dislocations – determination of dislocation density



dislocation density: total length of dislocation lines per unit volume $\rho = \sum l_{disl} / V$

SLOPOS -15

Prague, Czech Republic

September 2-6, 2019

See you in Prague