Positron Annihilation techniques for material defect studies

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The positron

Prediction by Dirac (1930)

theory of relativity and quantum mechanics

$$E = \pm (p^2c^2 + m_o^2c^4)^{1/2}$$

P.A.M.
Dirac

"Hole Theory" these solutions are interpreted as electrons with negative energy

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Positron discovery: the first pictures

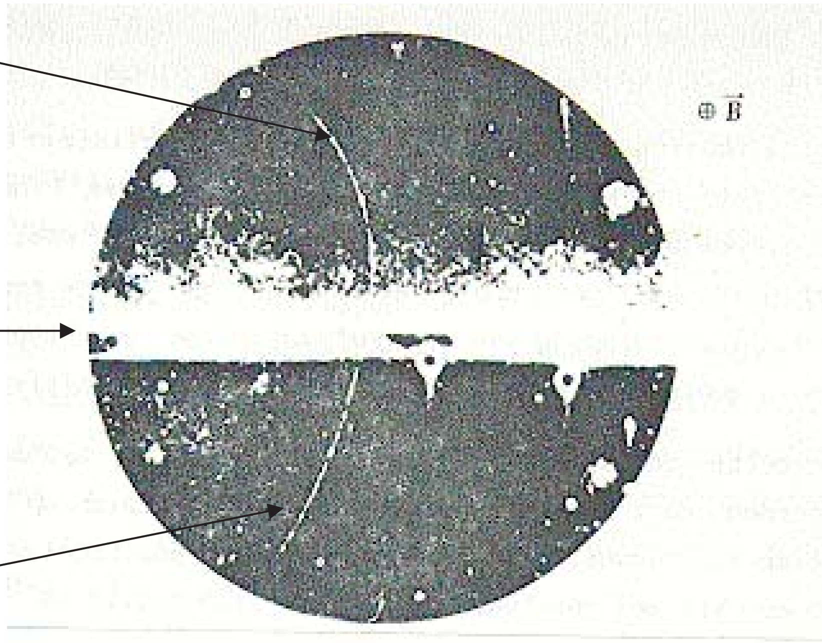
"Picture" of a positron in the cloud chamber (1932)

Decreased curvature

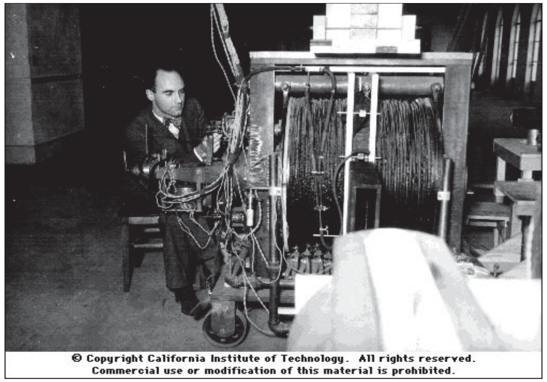
(23 MeV)

Lead foil

Begin of track (63 MeV)

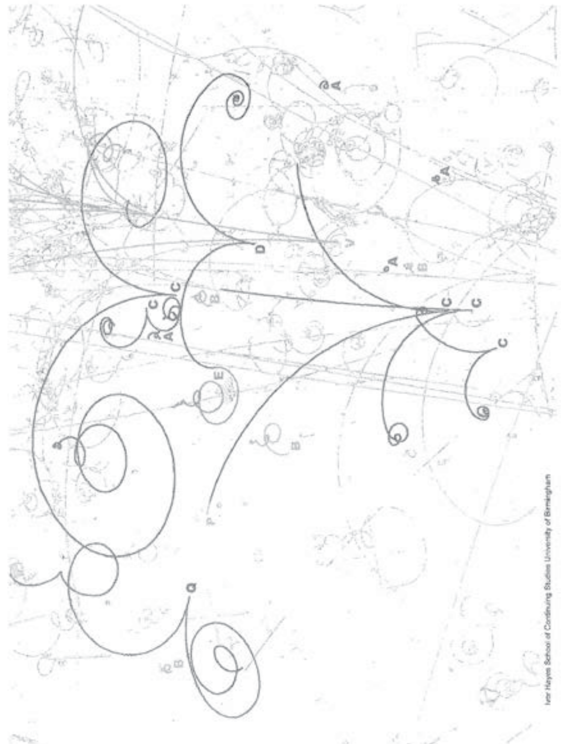


C.D. Anderson



More recent positron-electron tracks

Figure 1. Part of a bubble chamber picture. The dark lines are trails of tiny bubbles created as charged particles force their way through a tank of transparent liquid enclosed in a powerful magnet. At P, the main point of interest, a positron (antielectron) in flight meets an electron and annihilation takes place. One of the photons from the annihilation *materializes* at Q. At E, a different positron seems to change



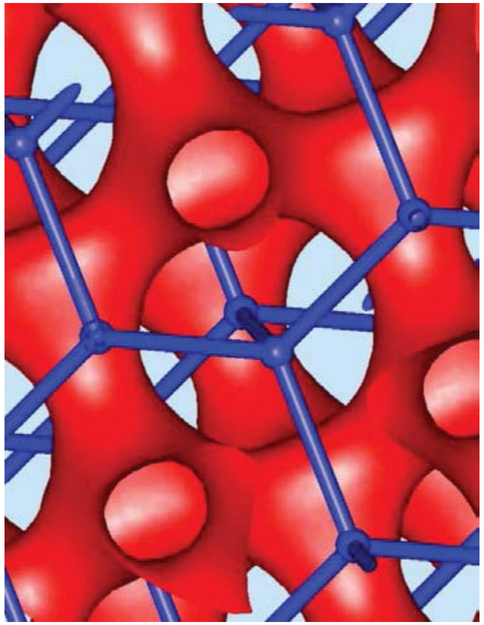
its sign. What has happened is that it has collided head-on with a stationary electron and has transferred all (within errors) its momentum to the electron. This shows that (within errors) the positron has the same mass as the electron. (This picture, from experiment E632 performed at the Fermilab 15 ft (4.6 m) bubble chamber, was found at the University of Birmingham. Graphics by Ivor Hayes.)

From: Phys. Edu 34(5) september 1995, G.T. Jones

Recent "picture" of a positron as iso-surface of the positron wave function

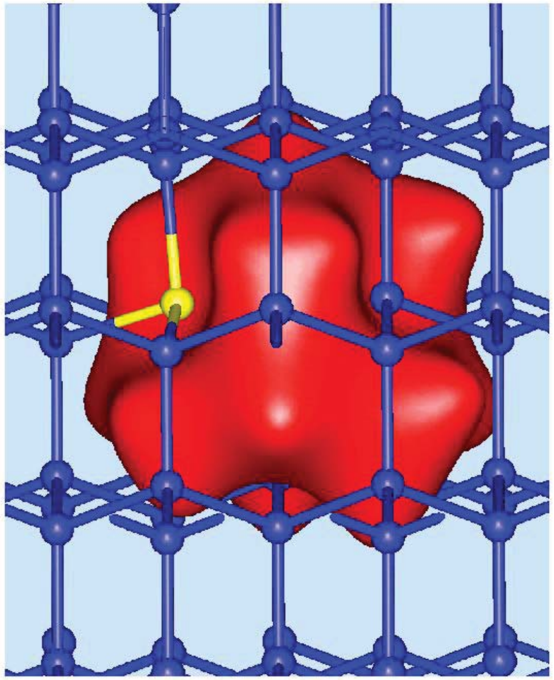
Seitsonen, Puska and Nieminen (Helsinki University of Technology, 1995)

Si



Free positron

Si vacancy + Sb neighboring atom



Trapped positron

Positron annihilation history

Prediction Ps

discovery Ps (Deutch)

1928

1932

1934

1951

60 ´s

70 ´s

80 ´s

90´s

2000

Electronic structure

studies

Bulk defect studies, polymers

Near surface defects

thin layers, interfaces, nano precipitates

New applications

New techniques (PEAS, AMOC, ...

Theory, QM calculations

Development exp. techniques

ACAR, Doppler, lifetime

Fast e+

Isotopes (weak sources)

Slow e+ beams

Isotopes (strong sources)

accelerators

reactors

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Positron annihilation (the Science Fiction approach)

The comic " Barbarella"

Jean Claude Forest 1964

Positron gun !

The movie "Barbarella"

Jane Fonda; Roger Vadim 1968

Antimatter + Matter → complete destruction

More details

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Characteristics of the positron as the anti-particle of the electron

	positron		electron
Rest Mass		$9.109\ 10^{-31}$ kg	
Charge	+	$1.60219\ 10^{-19}$ C	-
Spin	½	lepton	½
Magnetic moment	+	g q/2m S (g = 2.002319)	-
Classic radius		$2.81794\ 10^{-15}$ m	
Lifetime	stable (in vacuum) $t > 2 \times 10^{21}$ year		stable

$E = mc^2$ → rest mass = 511.0034 keV/c^2 (1 Joule = 1/q eV)

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Positron Annihilation

Process of transforming mass into photons

$e^+ + e^- \rightarrow$ gamma quanta (photons)

General Conservation laws:

- *energy*
- *linear momentum*
- *electric charge*
- *angular momentum*
- *parity*

most probable: two photon

Hence the study of the annihilation quanta will give information about the state of the annihilation positron electron pair *immediately before* the transition !

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Annihilation cross section for two photon annihilation

in the low velocity limit (free particles)

$$\sigma_{2y} = \pi\, r_o^{\,2}\, c \,/\, v$$

With r_o the classical Borh radius of the electron (positron)

~10^{-15} m

The other annihilation rates (1 , 3 photons) scale with the fine structure constant α^4 and α , respectively

$\alpha = e^2 / \hbar c\, 4\pi\varepsilon_o = 1/137$

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Case 1) zero momentum

both particles at rest (in lab system) or observation in the center of mass system

Before annihilation: | *After Annihilation*

Total energy

$E = E_{electron} + E_{positron} = 2\, m_o c^2$ ⟶ $E_{y1} + E_{y2}$ (1)

Total momentum

$p = 0$ ⟶ $E_{y1}/c + E_{y2}/c$ (2)

(as components of p vector)

- *<u>two</u> photons emitted in <u>opposite</u> directions*
- *energy of each photon* $= \underline{m_o c^2} = 511$ *keV*

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Case 2 : electron momentum $p = p_x$

Before annihilation: *After Annihilation*

Total energy $E = E_{electron} + E_{positron}$

$= (m_o^2 c^4 + p_x^2 c^2)^{1/2} + m_o c^2 \longrightarrow = E_{\gamma 1} + E_{\gamma 2}$ (1)

Total momentum $p = p_x \longrightarrow = E_{\gamma 1}/c + E_{\gamma 2}/c$

$p_y = p_z = 0$ $= p_y = p_z$ (2)

in case gamma´s emitted in the direcion of p

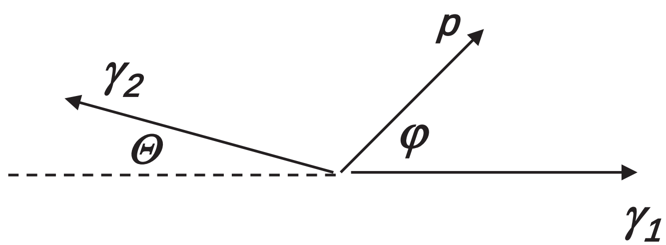
- *<u>two</u> photons emitted in <u>opposite</u> directions*
- *energy of the photons* $= m_o c^2 \pm c\, p_x/2$ *(doppler broadening)*

General solution

$$\Theta_{\gamma 1-\gamma 2} = \frac{2\,pc\,\sin(\phi)}{pc\,\cos(\phi) + m_0 c^2 + \sqrt{m_0^2 c^4 + p^2 c^2}}$$

$$E_{\gamma 1} = \frac{m_0 c^2 + \sqrt{m_0^2 c^4 + p^2 c^2} + pc\,\cos(\phi)}{2}$$

$$E_{\gamma 2} = \frac{m_0 c^2 + \sqrt{m_0^2 c^4 + p^2 c^2} - pc\,\cos(\phi)}{2}$$



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Approximation:

$$E = m_o c^2 \pm \tfrac{1}{2}\ p_{//} c \quad keV$$
$$\Theta_{y\text{-}y} = \pi - p_{\perp} / m\, c \quad radians$$

Some values :

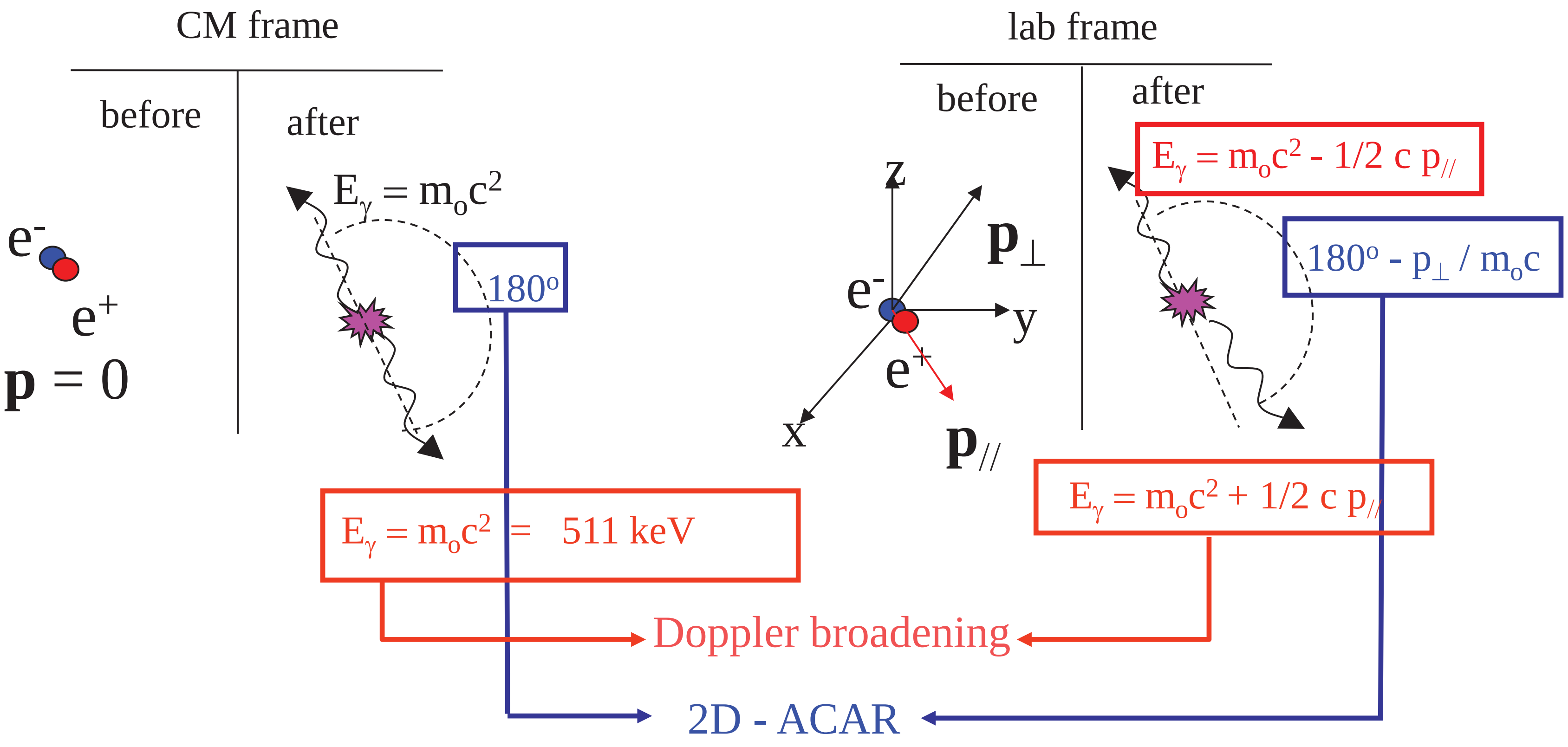
Energy of electrons (in solids) 10 eV → $\tfrac{1}{2}\ p_{//}\, c = 1.5 \times 10^3$ *eV*

(compare to 511×10^3 *eV)*

p of the order of 1×10^{-24} *m kg/s* → $p_{\perp} / mc = 3$ *mrad* *(3 mm at 1 m)*

The effect of conservation of momentum and energy

2γ $e^+ + e^-$ annihilation process : conservation of energy and momentum



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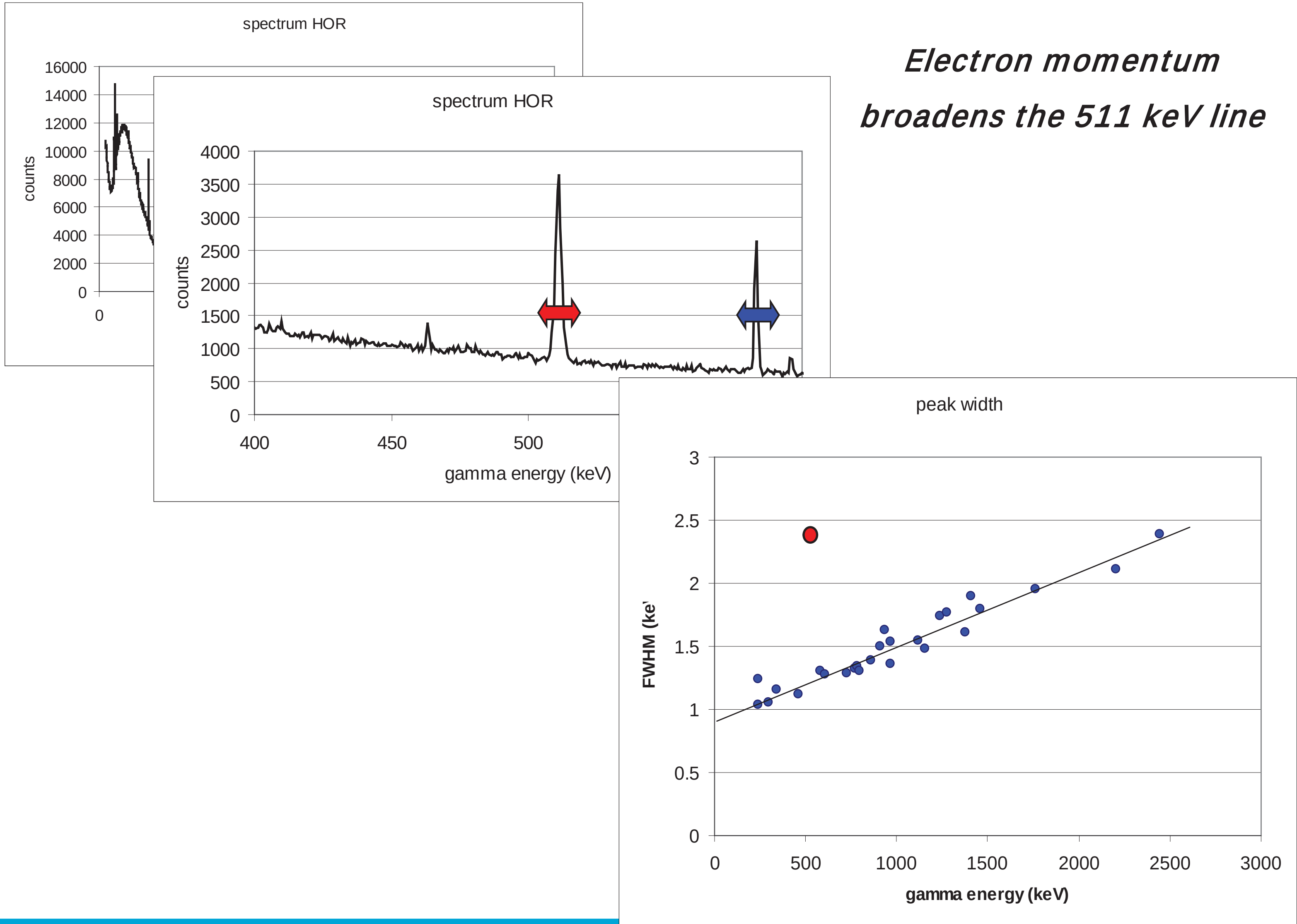
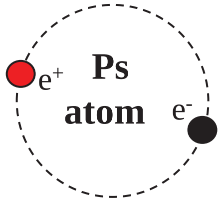
Electron momentum broadens the 511 keV line



Positronium:

An electron - positron bound state



H atom	*physical property*	*Ps atom*
1.0080	**atomic mass (amu)**	**0.00110**
0.99956	**reduced mass**	**½**
1 a_0 (radius)	**size**	**2 a_0 (diameter)**
13.598 eV	**ionization energy**	**6.803 eV**
J =0 (para)	**spin states**	**S = 0 (para) ↑↓**
J=1 (ortho)		**S = 1 (ortho) ↑↑**
∞	**life time**	**125 ps (125 x 10^{-12} s, para)**
		142 ns (142 x 10^{-9} s , ortho)

a_0 = Bohr radius = 0.528 A

Positronium annihilation modes:

Para Ps ↑↓ **“self” annihilates by two-gamma emission (125 ps)**

Ortho Ps ↑↑ “self” annihilates by *three-gamma* emission (142 ns)

or in media:

o- Ps (↑↑) + e^- (↓) → p-Ps (↑↓) + e^- (↑) (spin exchange)

o- Ps (↑↑) + e^- (↓) → 2 gamma´s + e^- (↑) (pick off)

Para -Ps has zero internal momentum therefore Eγ ~ 511 keV
(only translational momentum of the atom)

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Positron sources

- ***Isotopes***
  - *nuclear reactions*
  - *neutron absorption*

- *Pair production*
  - *Electron accelerators*
  - *Fission reactors*
    - *neutron → gamma conversion*
    - *prompt gammas*

Examples of positron emitters

Isotope	half life	% e+	production	application	
^{11}C	20 min	100	$^{11}B(p,n)^{11}C$	medical diagnostics (PET)	
			$^{10}B(d,n)^{11}C$		
			$^{14}N(p,\alpha)^{11}C$		
^{13}N	10 min	100	$^{12}C(d,n)^{13}N$	"	
			$^{16}O(p,\alpha)^{13}N$		
^{15}O	2 min	99	$^{14}N(d,n)^{15}O$	"	
^{18}F	110 min	97	$^{18}O(p,n)^{18}F$	"	
^{22}Na	2.6 y	90	$^{24}Mg(d,\alpha)^{22}Na$	PA	accelerators
^{58}Co	71.3 d	15.5	$^{58}Ni(n,p)^{58}Co$	"	nucl reactors
^{64}Cu	12.9 h	19.3	$^{63}Cu(n,y)^{64}Cu$	"	nucl reactors

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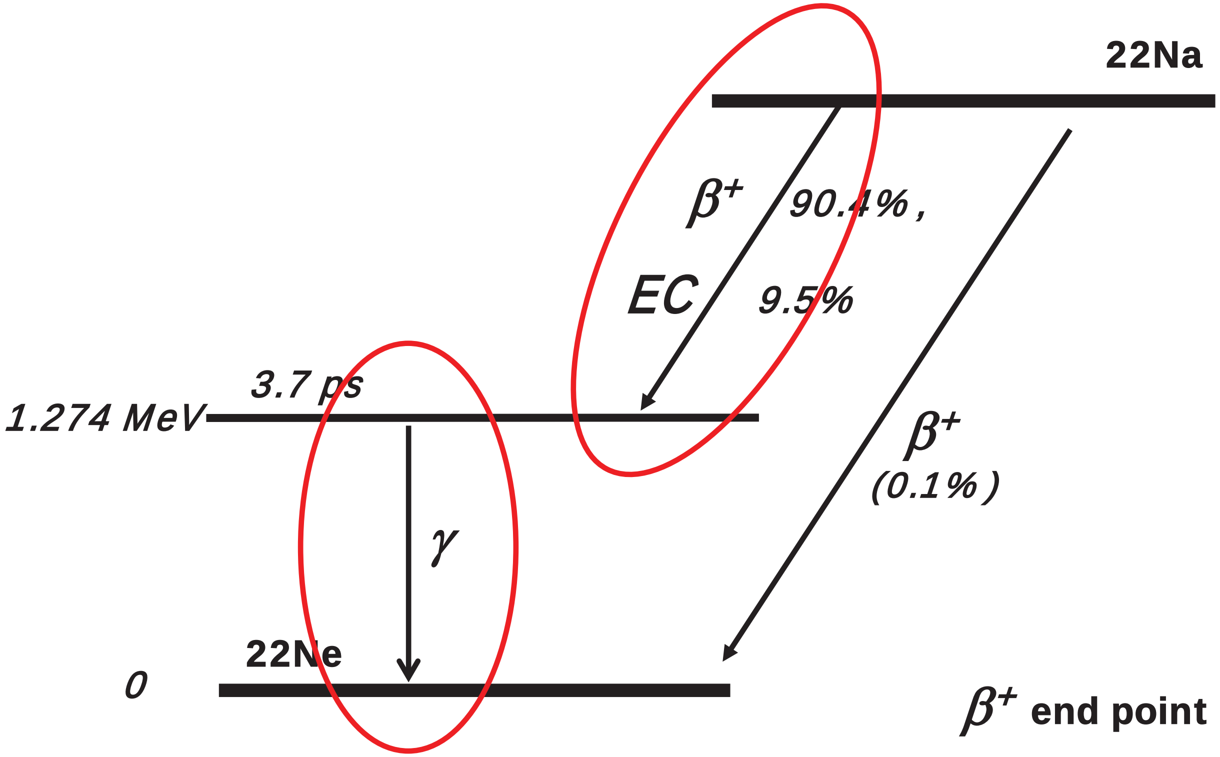
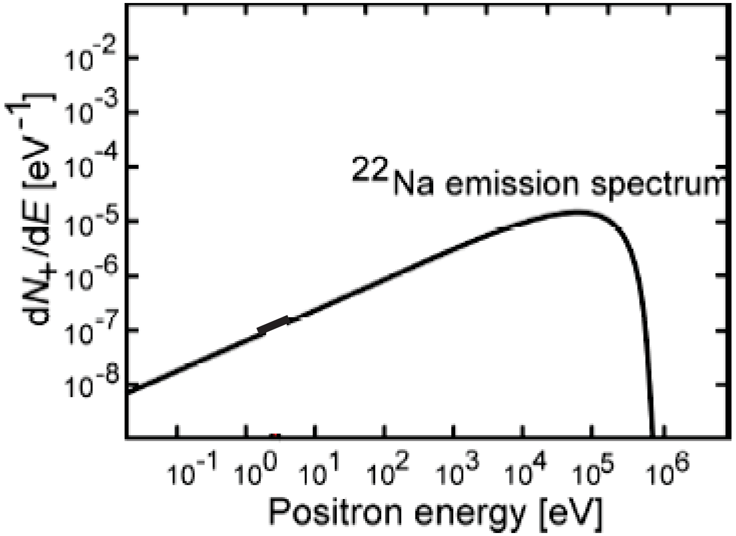
^{22}Na decay scheme



β^+ end point energy 540 keV

half life 2.602 year

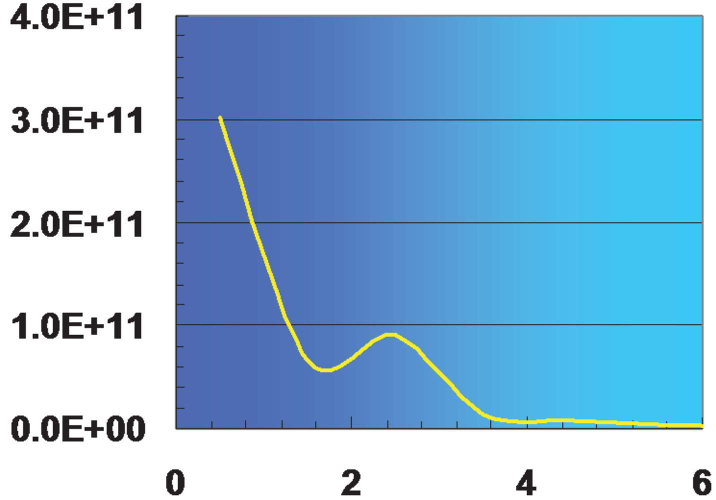
Positron kinetic energy distribution



$^{22}Na \rightarrow {}^{22}Ne + e^{+} + \nu_e$

$(1274\ keV\ {}^{22}Ne^{*})$

HOR gamma flux distribution



Pair production:

positron kinetic energy depends on the initial energy of the photon

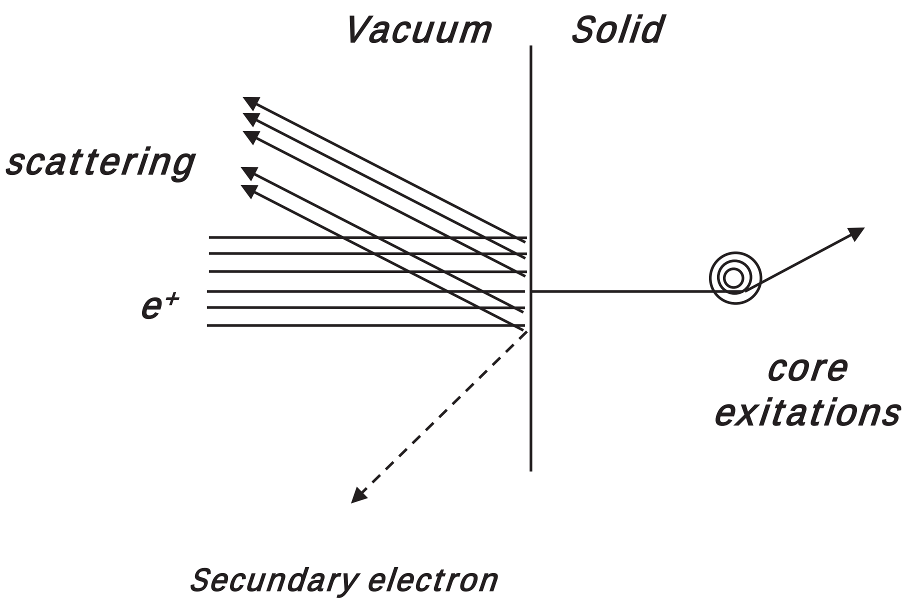
(Bremsstrahlung or prompt fission gamma energy - 1.022 MeV)

Positron - solid interaction

First encounter

Fast positrons entering a solid loose their kinetic energy by different ineleastic energy loss mechanisms

Timescale < 10 $^{-15}$ seconds



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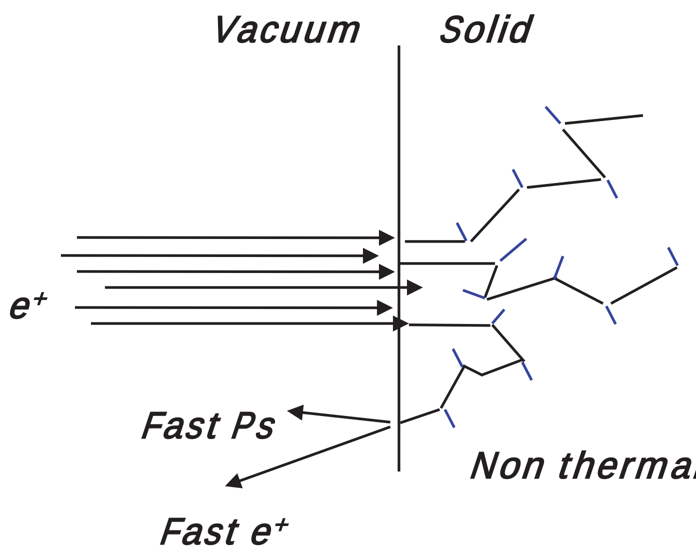
Positron - solid interaction

Thermalisation

Timescale < 10^{-12} seconds



- Plasmons
- Electron-hole pairs
- phonons

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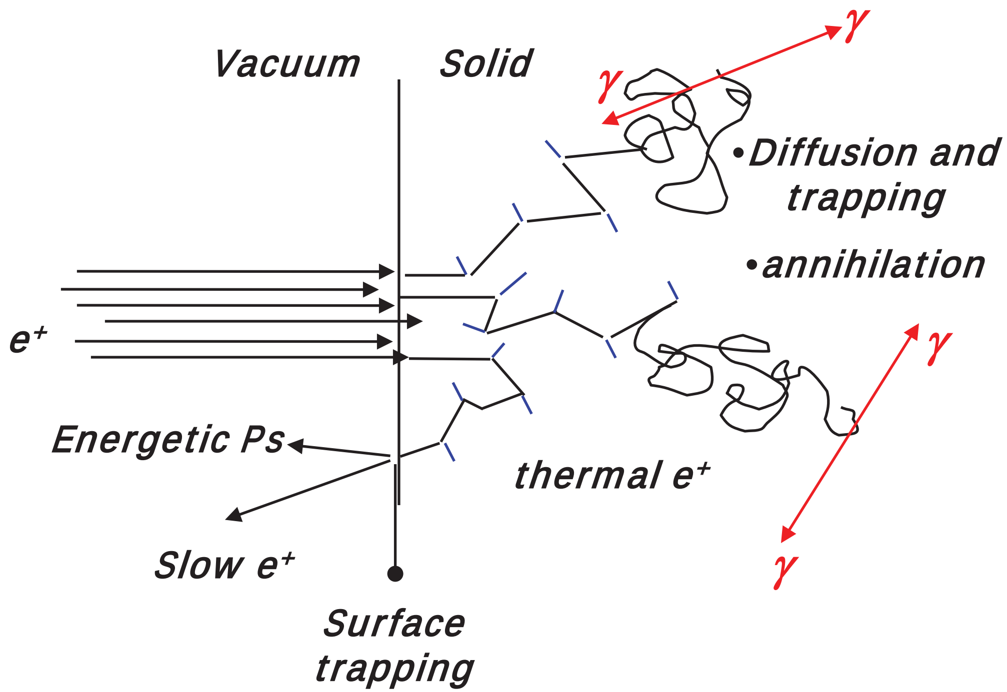
Positron - solid interaction

equilibrium

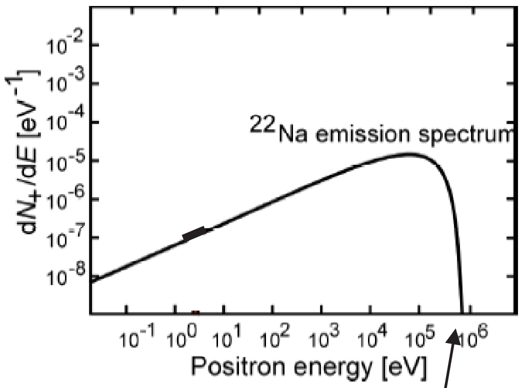
Timescale < 10^{-10} seconds



Positron implantation profile

Fast positrons

(β^+ energy distribution)



$$I(x) = I(0) \exp(-a_+ x)$$

$$a_+ = (16 \pm 1)\ \rho E_m^{-1.43} \quad (cm^{-1})$$

(E in MeV)

No depth resolution

Implantation depth range

Element	density [g/cm³]	a_+ [cm⁻¹]	r (99%) [μm]
Al	2.7	104	442
C	2.25	87	530
Li	.53	20	2250
Mo	10.2	394	117
Si	2.33	90	500
W	19.35	747	62

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Positron implantation profile

"Slow" positrons

Mono energetic beams (~ 0.1 - 25 keV)

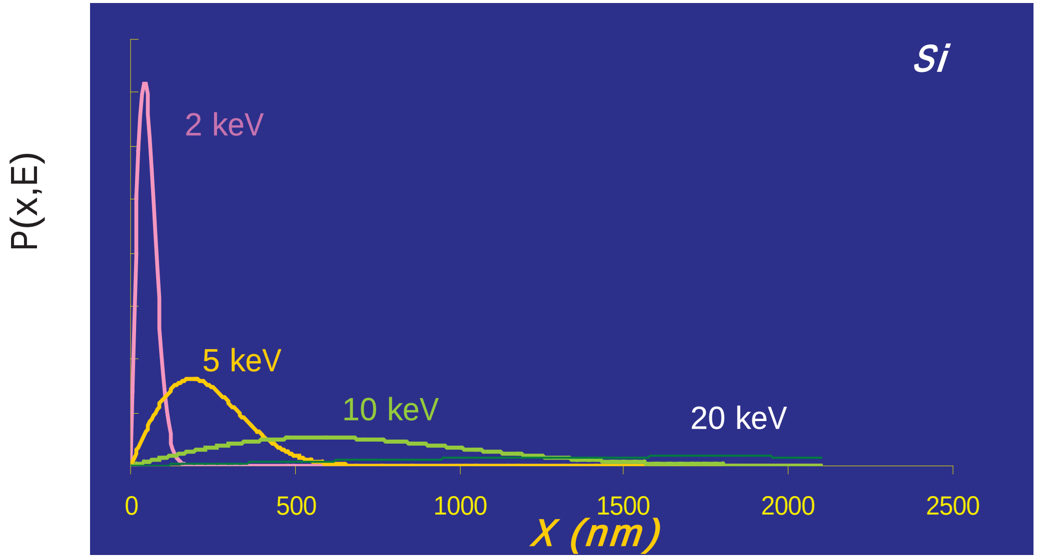
with m = 2

$$P(x,E) = \frac{2x}{x^2_o} exp - \left[\frac{x}{x_o}\right]^2$$

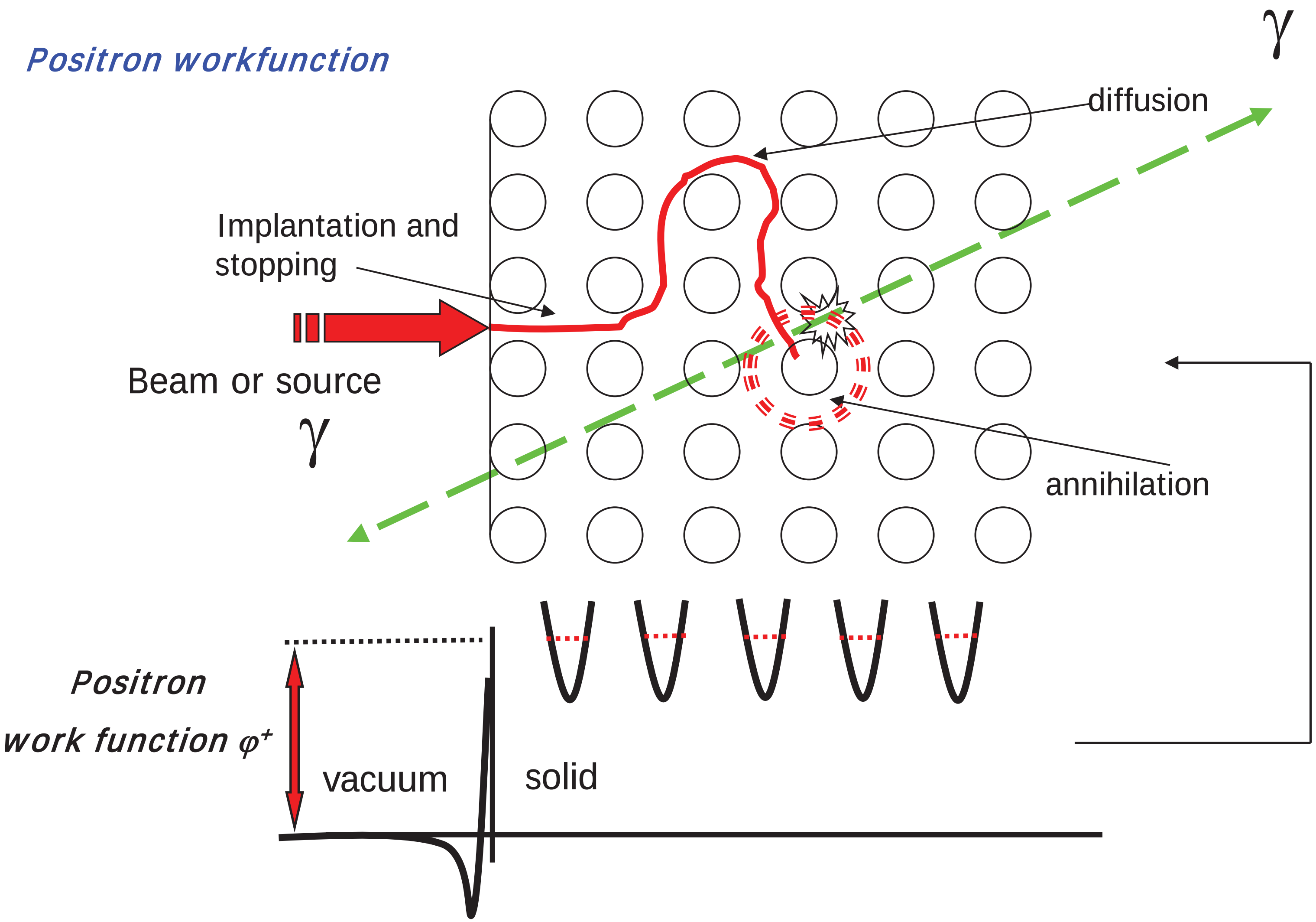
$x_o = 1.3 <x>$

$<x> = (\alpha / \rho) E^n$

$n = 1.6 – 1.8, \ \alpha \sim 4.0$

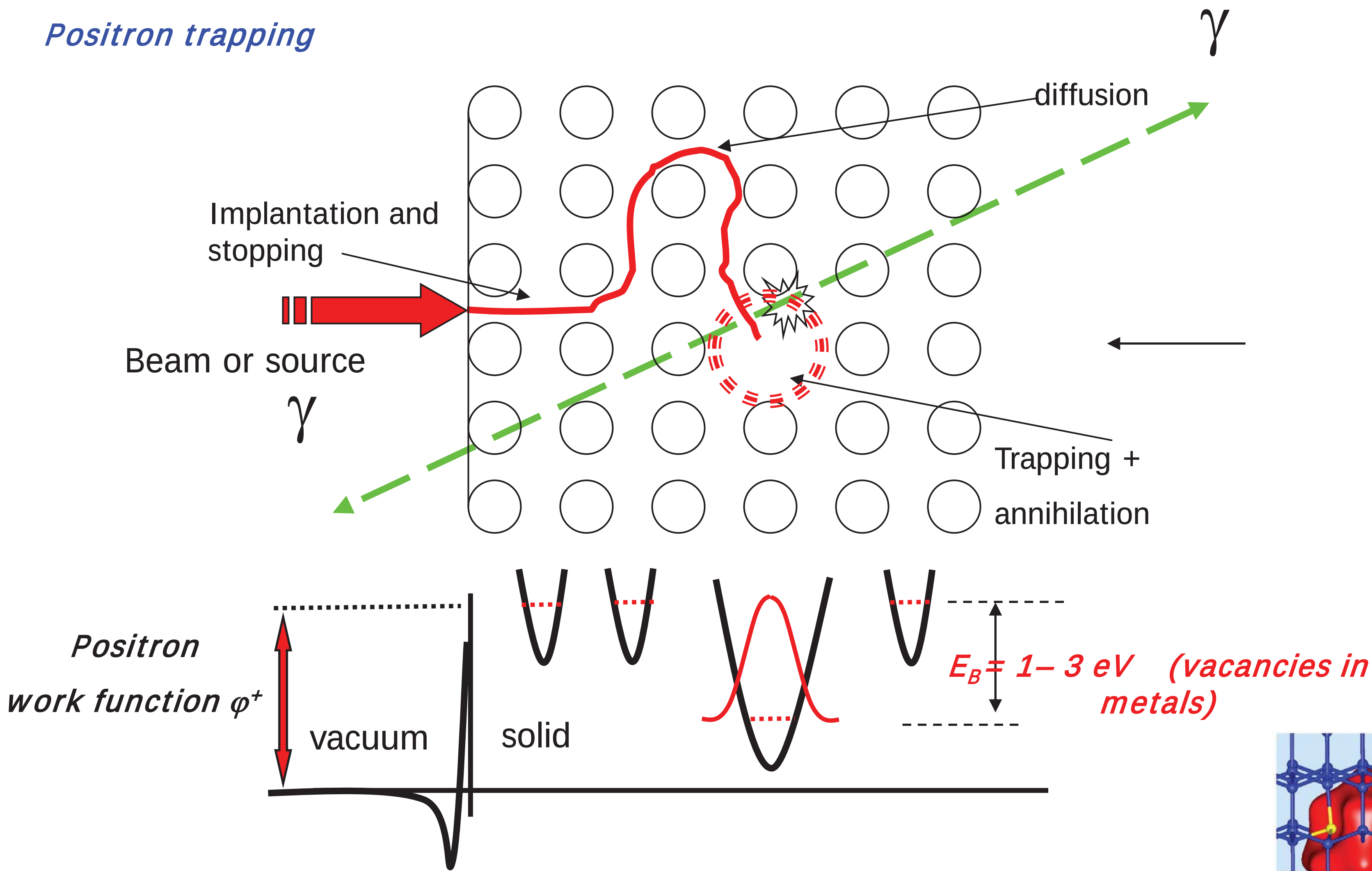


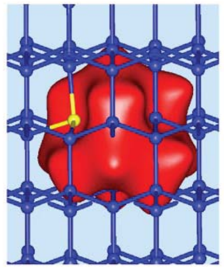
Positron workfunction



$\varphi^{+} < 0$ spontanous emission from the solid into vacuum (W, Mo, Ni, SiC, . . .)

Positron trapping

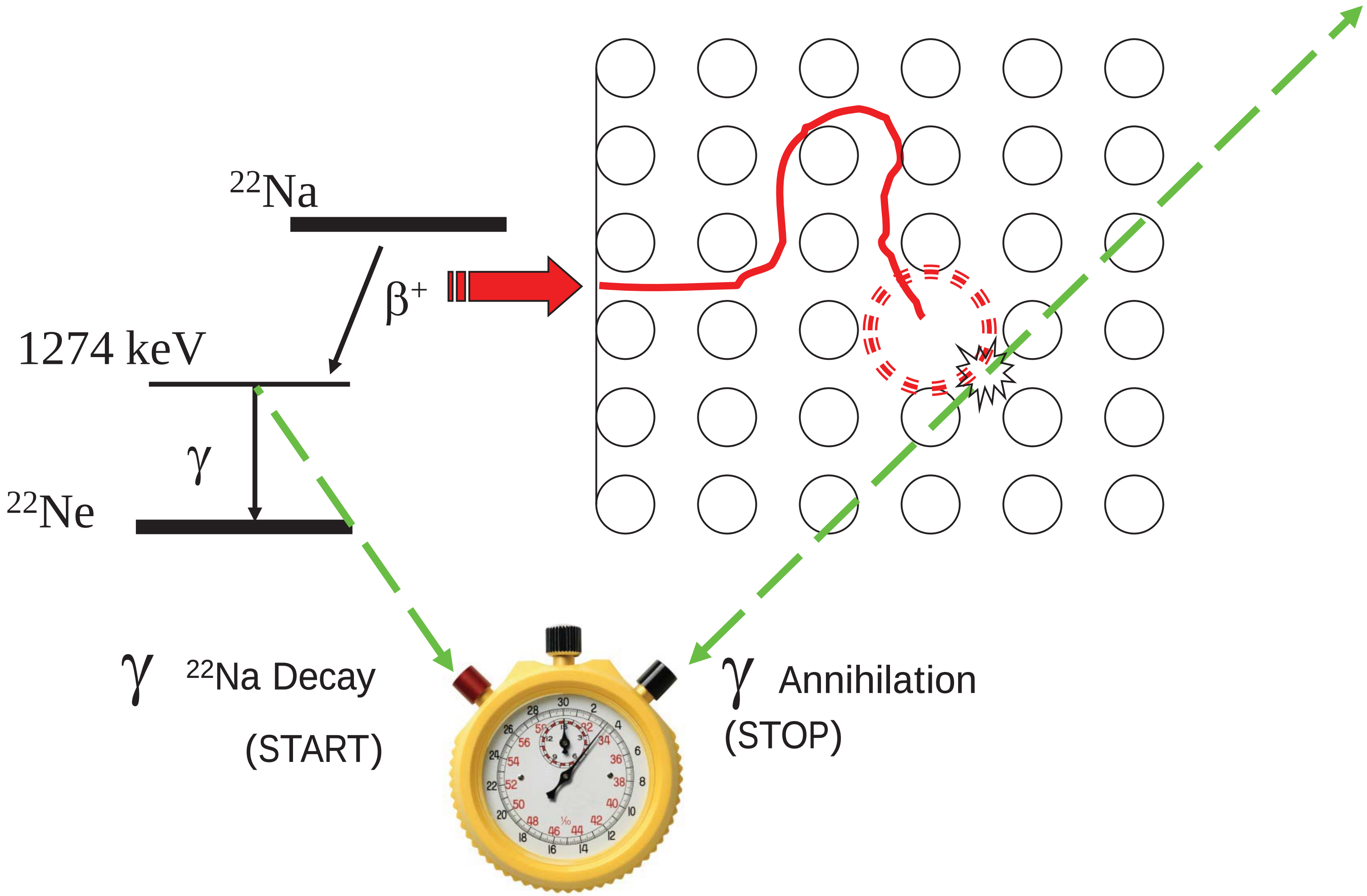




The attractive potential well (a defect) results in a confinement of the positron (collapse of the wave function)

Trapping rate (ν_t) of the order of 10^{14} s^{-1}

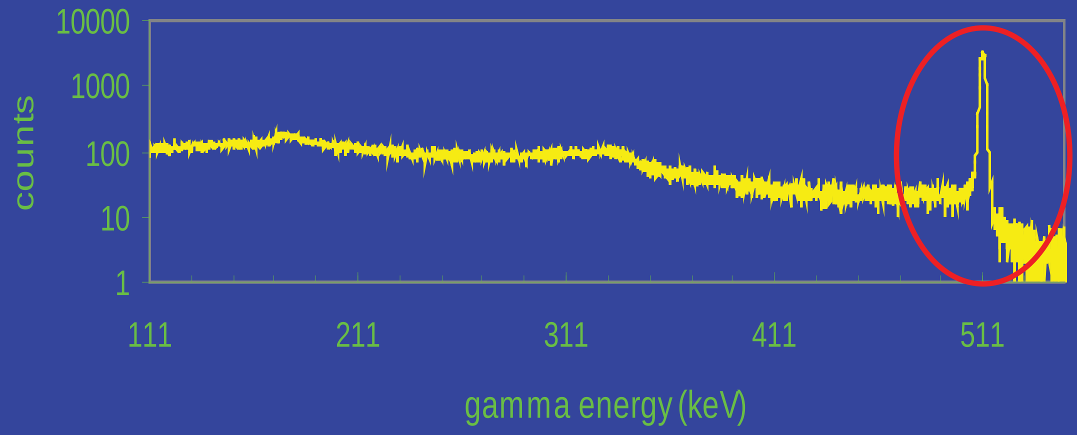
Probing Local electron density with positrons



Observables (1)

Doppler broadening
S(hape) and W(ing)

detecting one 511 keV gamma and determing the width of the photo peak



- S parameter

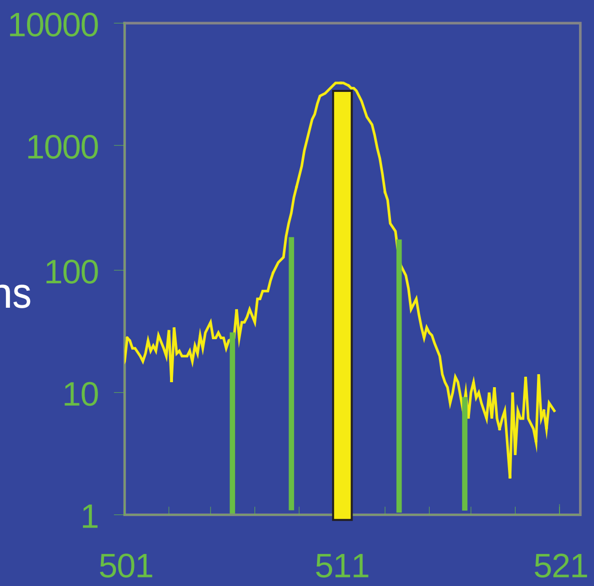
Low momentum

Valence, conduction electrons

P(ara) Positronium

Open volume

(vacancies, voids ...)



W parameter

High momentum

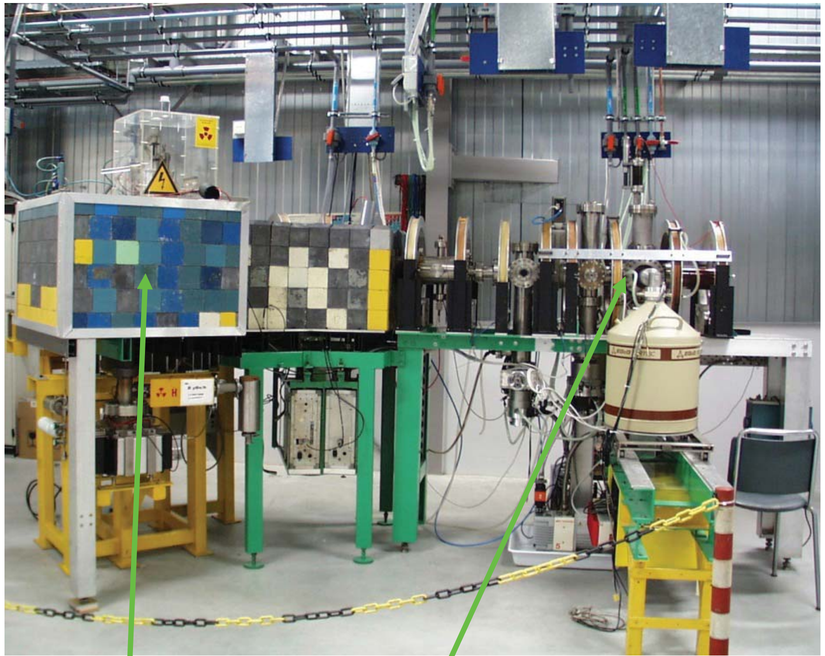
Core electrons

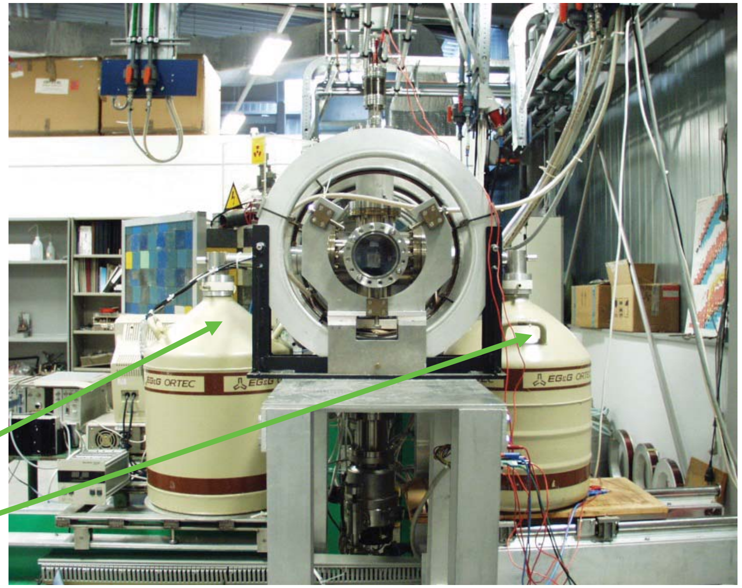
Local chemical surroundnig

Nano precipitates, sub-latice defects

Doppler broadening setup

The Variable Energy Positron beam (VEP)





^{22}Na source

sample chamber

Coincidence setup (2 detectors)

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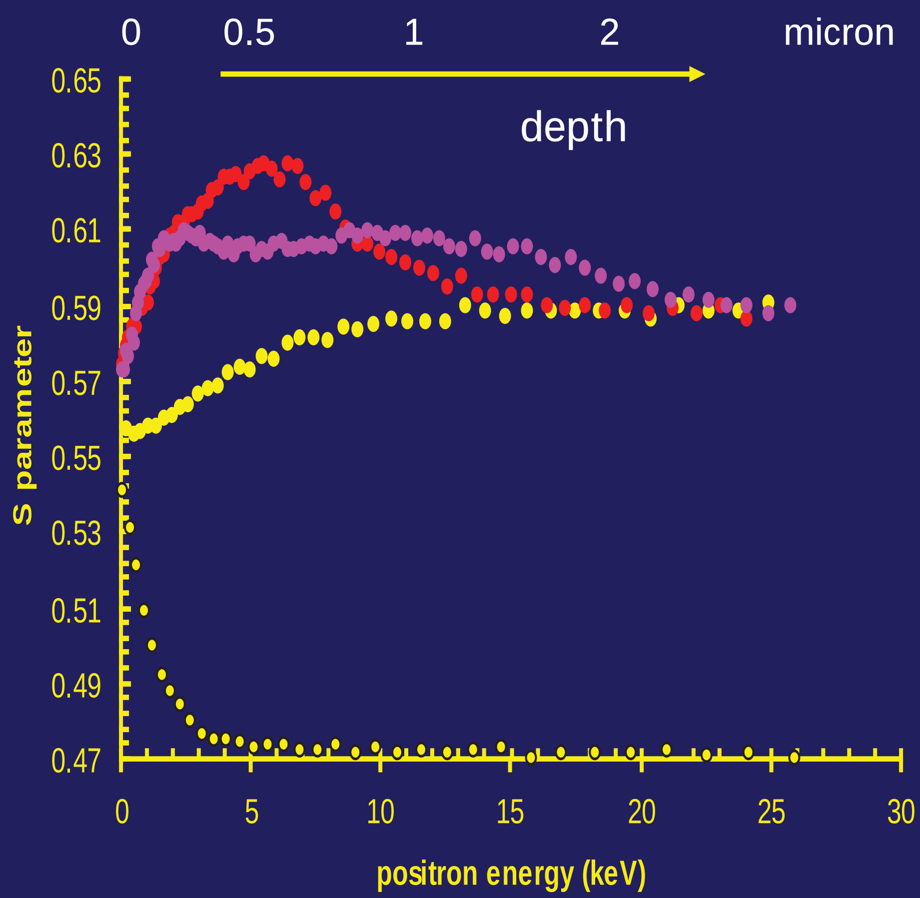
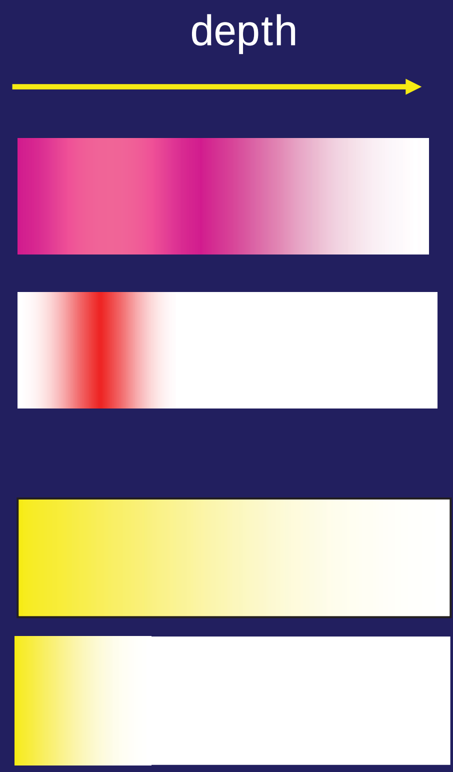
Some examples (beam exp´t)

? Model ?





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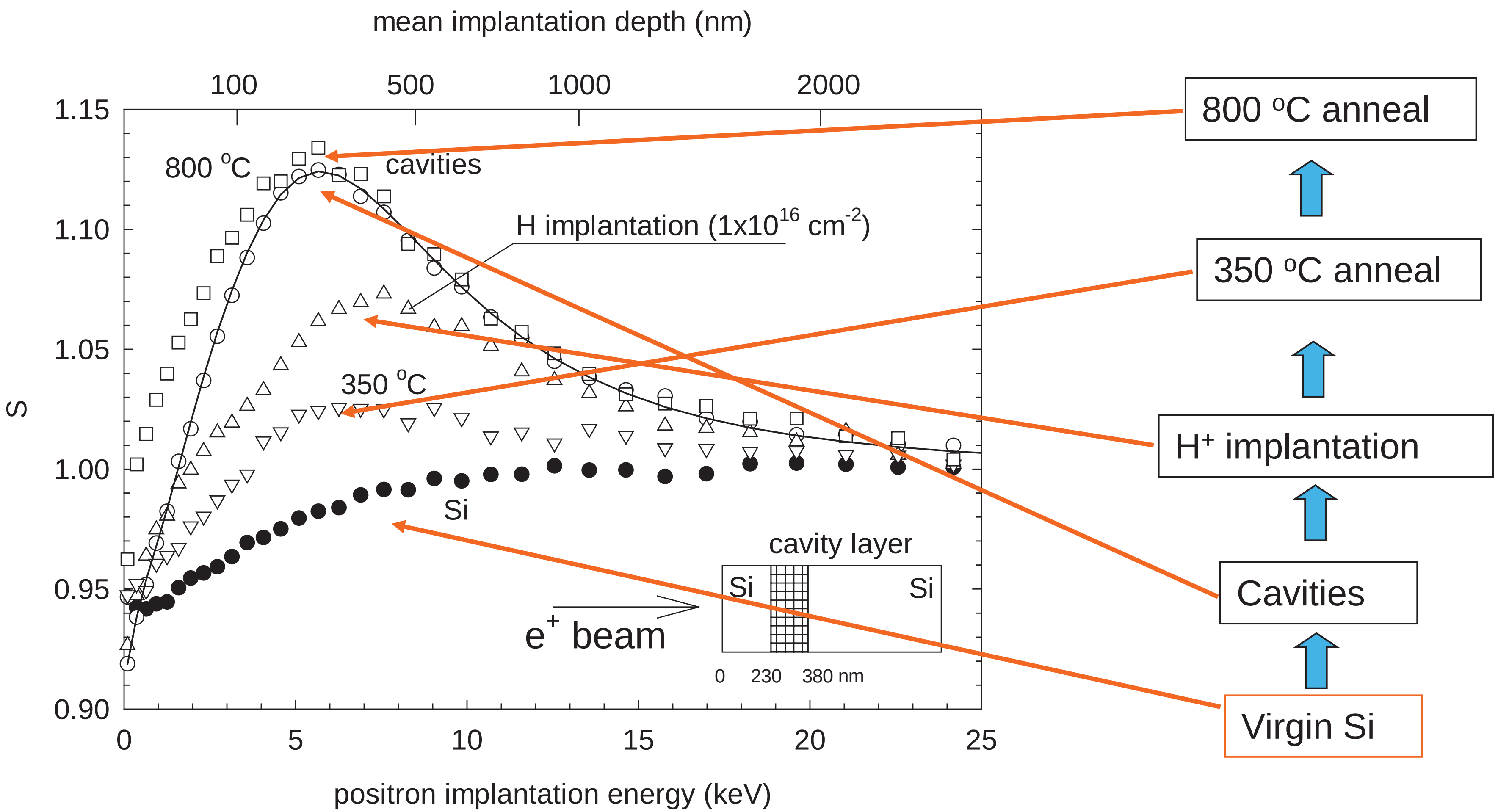
1 x 10^{16} H^+ cm^{-2}



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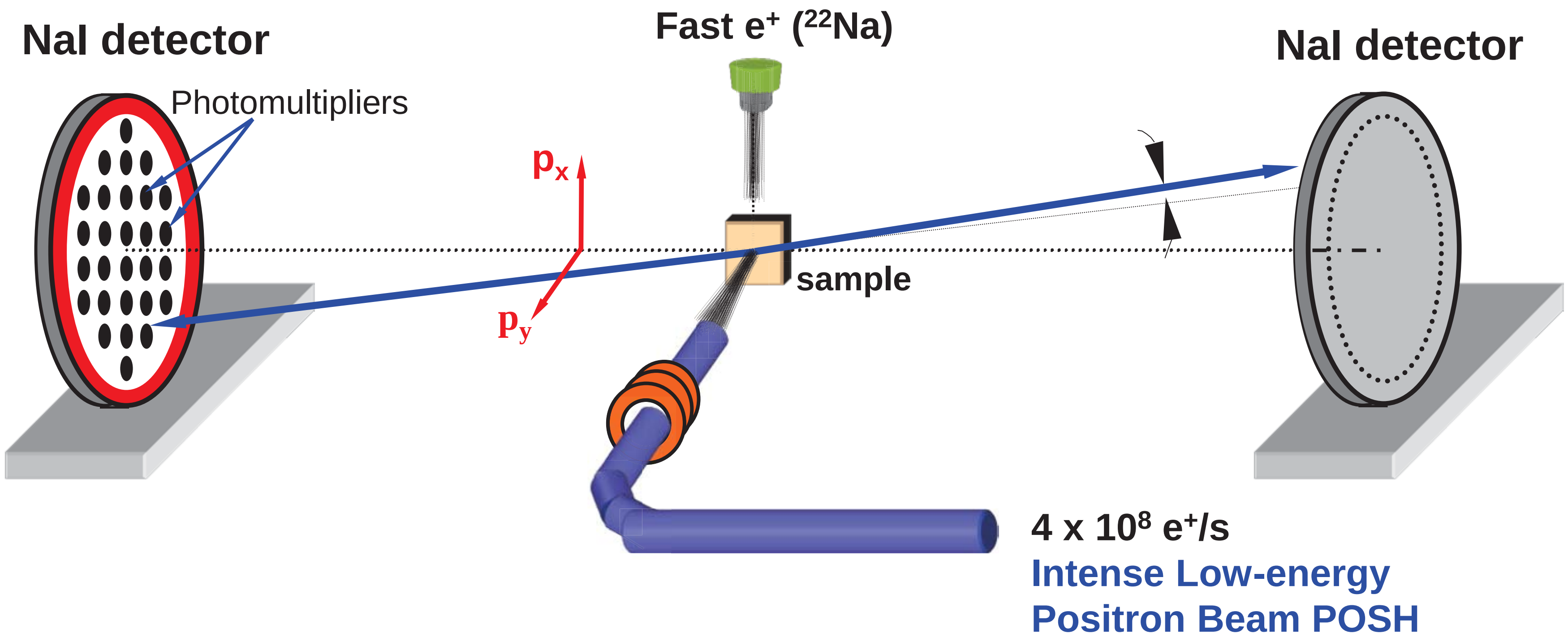
Observables (2)

Angular Correlation **measument of the angle between the two 511 keV gammas**



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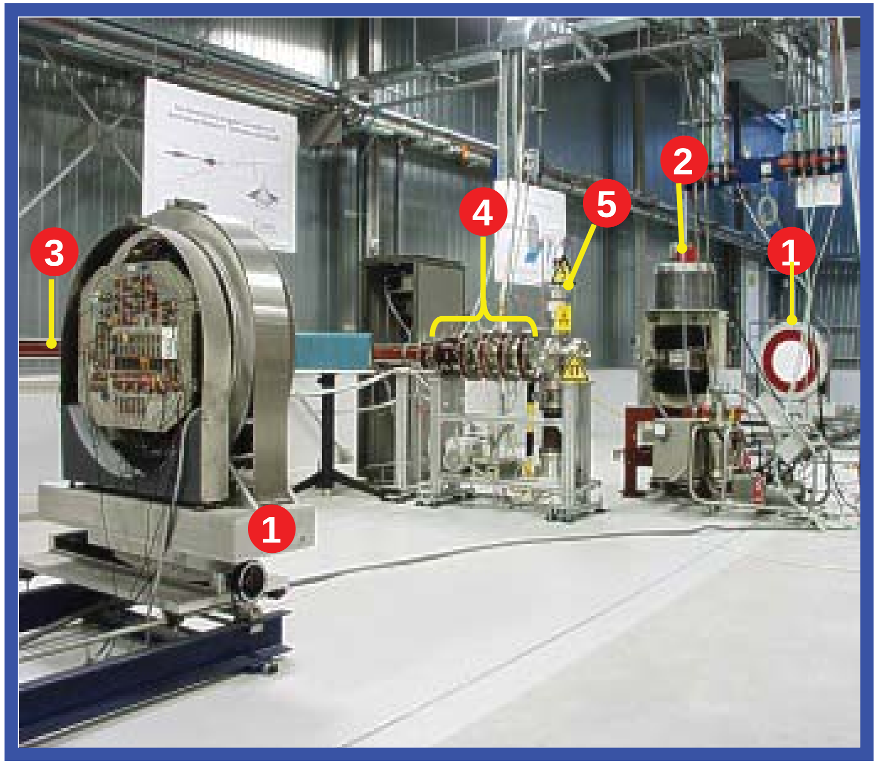
The Delft POSH 2D-ACAR setup



1 Anger cameras

2 source (^{22}Na) chamber

3 POSH e^+ beam line

4 Accelerator

5 Target chamber

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Example of 1 D ACAR

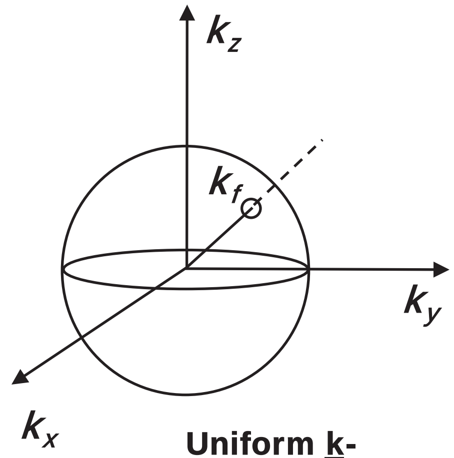
Measurement of the Fermi level of electrons in Al

free electron gas model

Highest energy level occupied E_f

→ Maximum momentum $p_f = (2 m E_f)^{1/2}$

→ Maximum angle angular correlation (α_f)



Uniform k-distribution

show

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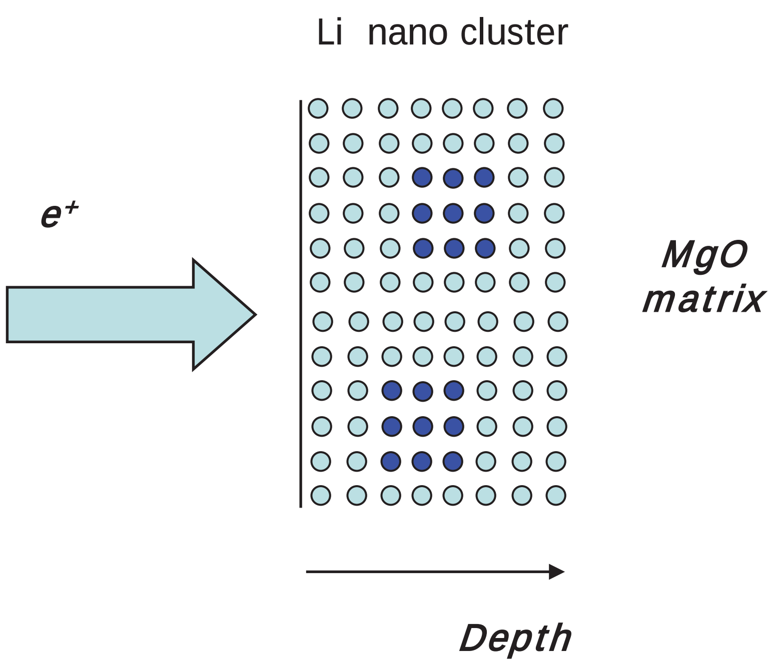
Li nanoclusters are *FCC* and in simple epitaxy with the MgO host matrix.



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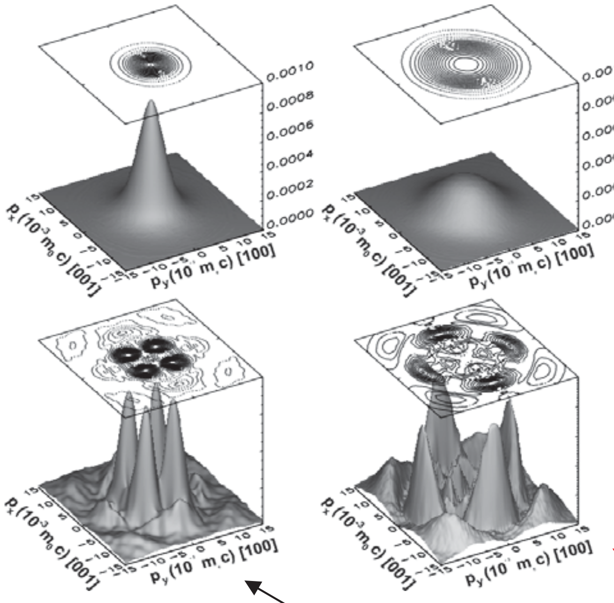
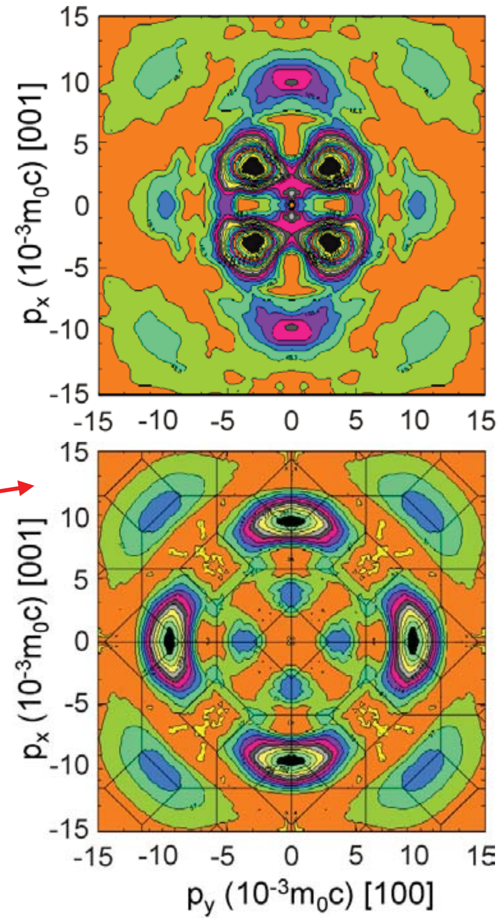
Li nanoclusters are *FCC* and in simple epitaxy with the MgO host matrix.

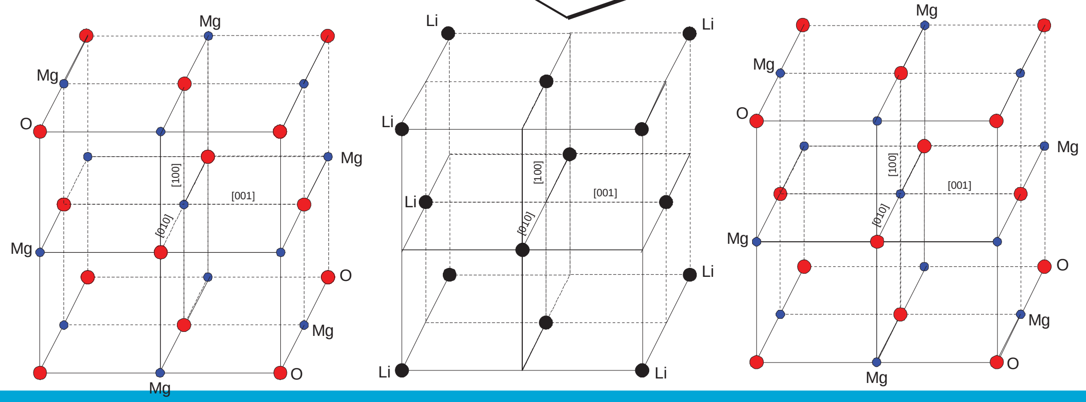
MgO(100):Li POSH (4 keV)



MgO(100) fast e^+ (^{22}Na)

MgO(100):Li nanocrystals





In MgO Li nano clusters have FCC structure !

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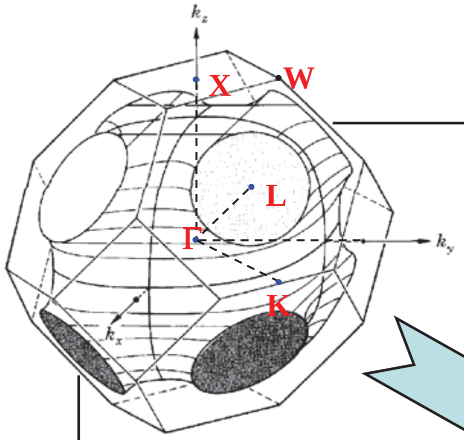
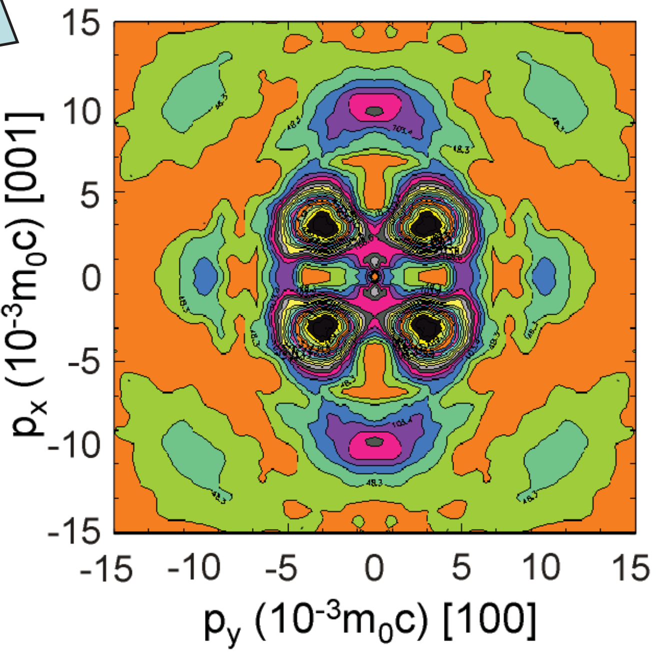
Typical FS surface of an fcc alkali like metal



Projection on

p_x-p_y plane



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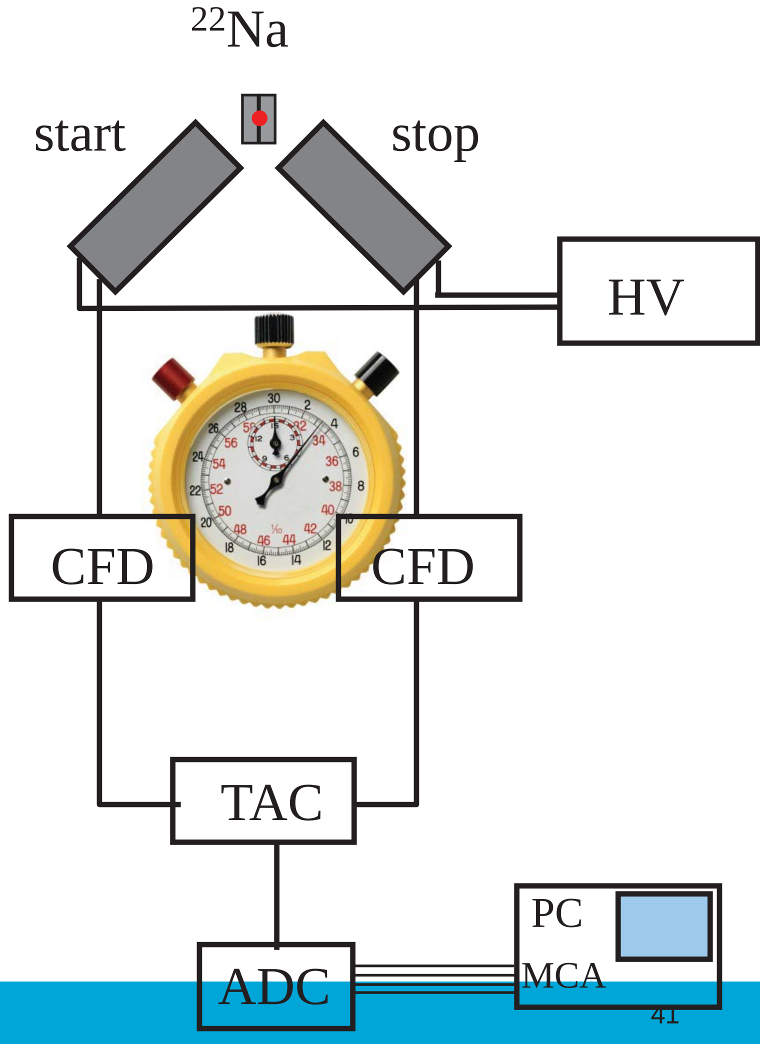
Observables (3)

positron lifetime (PALS)

measurement of the time elapsed between positron injection and annihilation

resolution 260 ps



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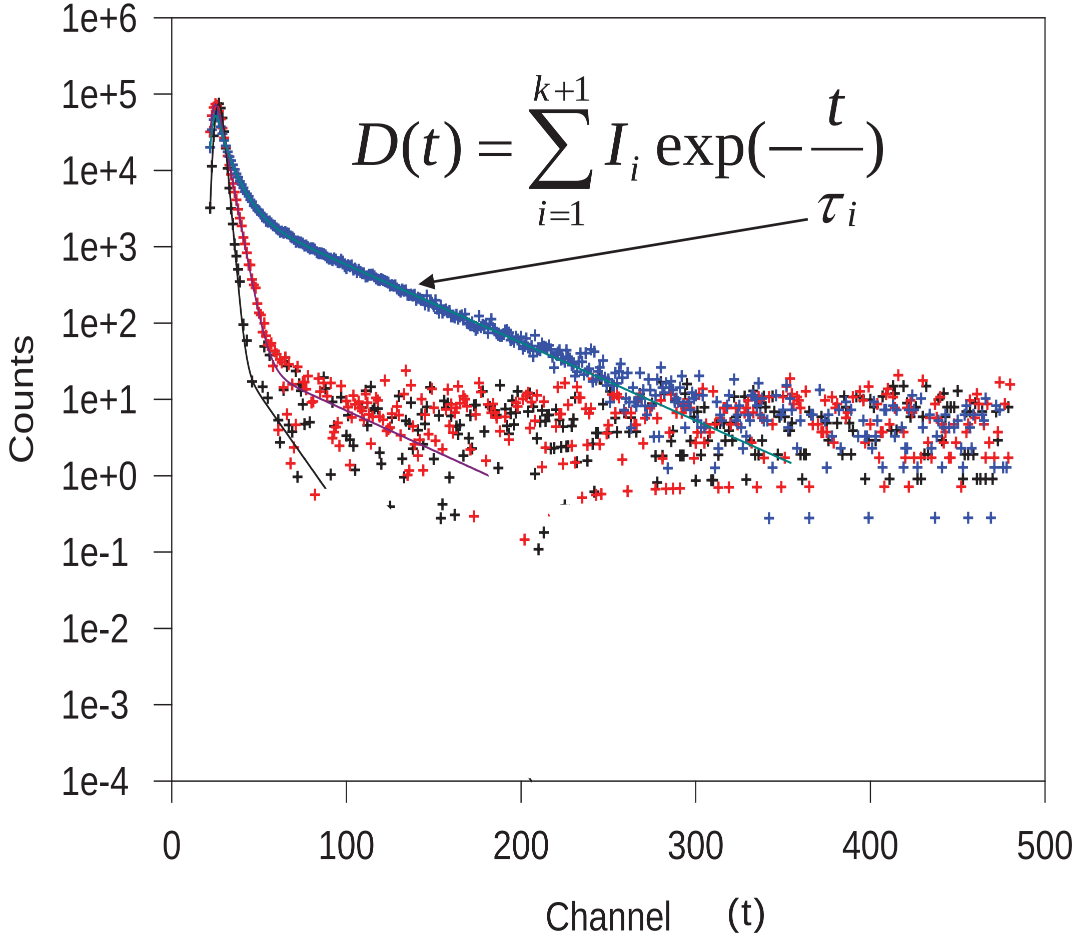
Typical PALS spectra



$$D(t) = \sum_{i=1}^{k+1} I_i \exp(-\frac{t}{\tau_i})$$

Positron Lifetimes in dense solids (metals and semiconductors)

$e^+ + e^- \rightarrow 2\ \gamma$

- *Tungsten (100 ps)*
- *Silicon (220 ps)*

Positron lifetimes in polymer

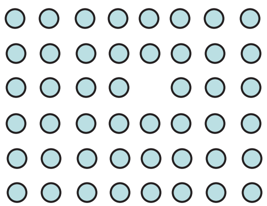
ortho - Ps formation (140 ns)

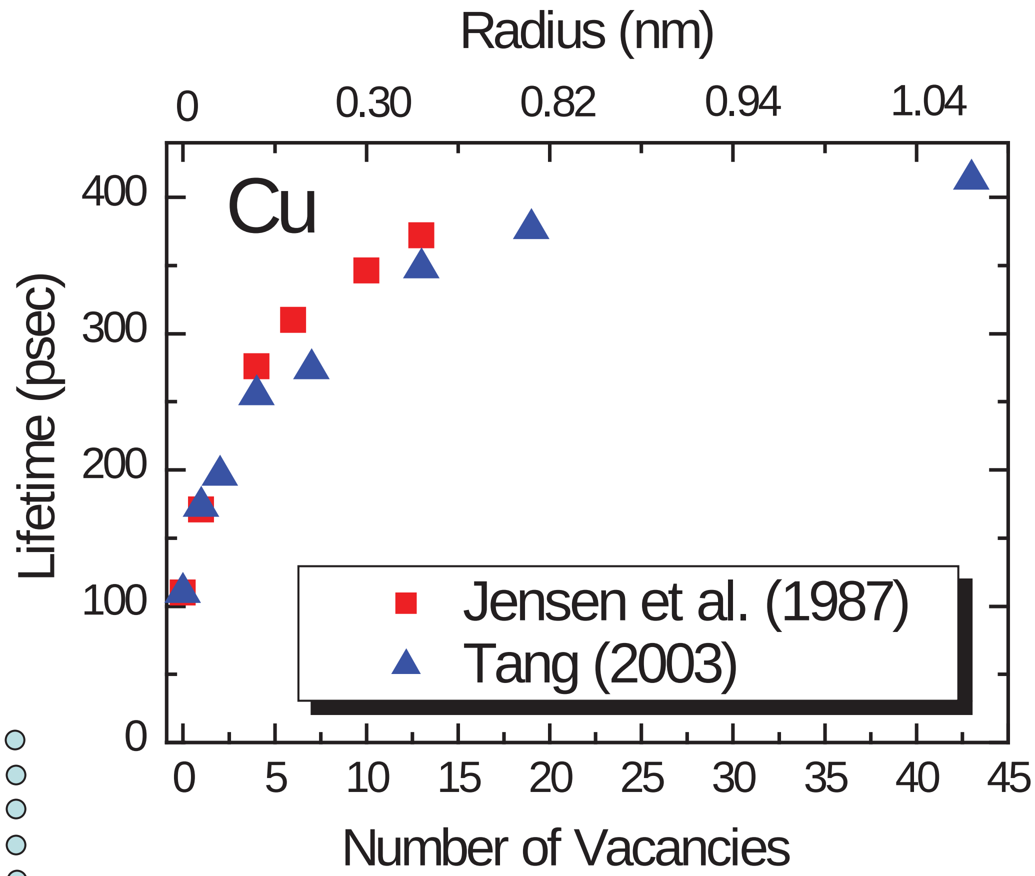
- *PolyEthyleen (PE) (2-3 ns)*

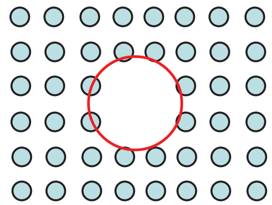
Calculated positron lifetimes in vacancy clusters

$$\lambda_0(n_e) = \pi r_o^2 c n_e$$

vacancy







Vacancy cluster

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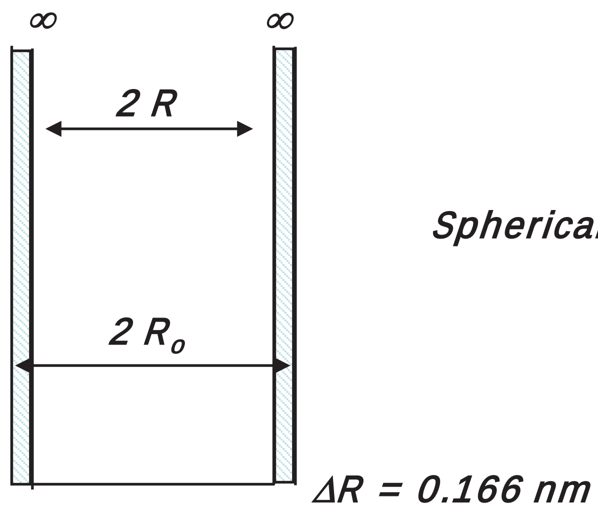
Positron lifetimes in polymers

Semi empirical relation between volume size (R) and o-Ps decay rate

Usually the third long lifetime component

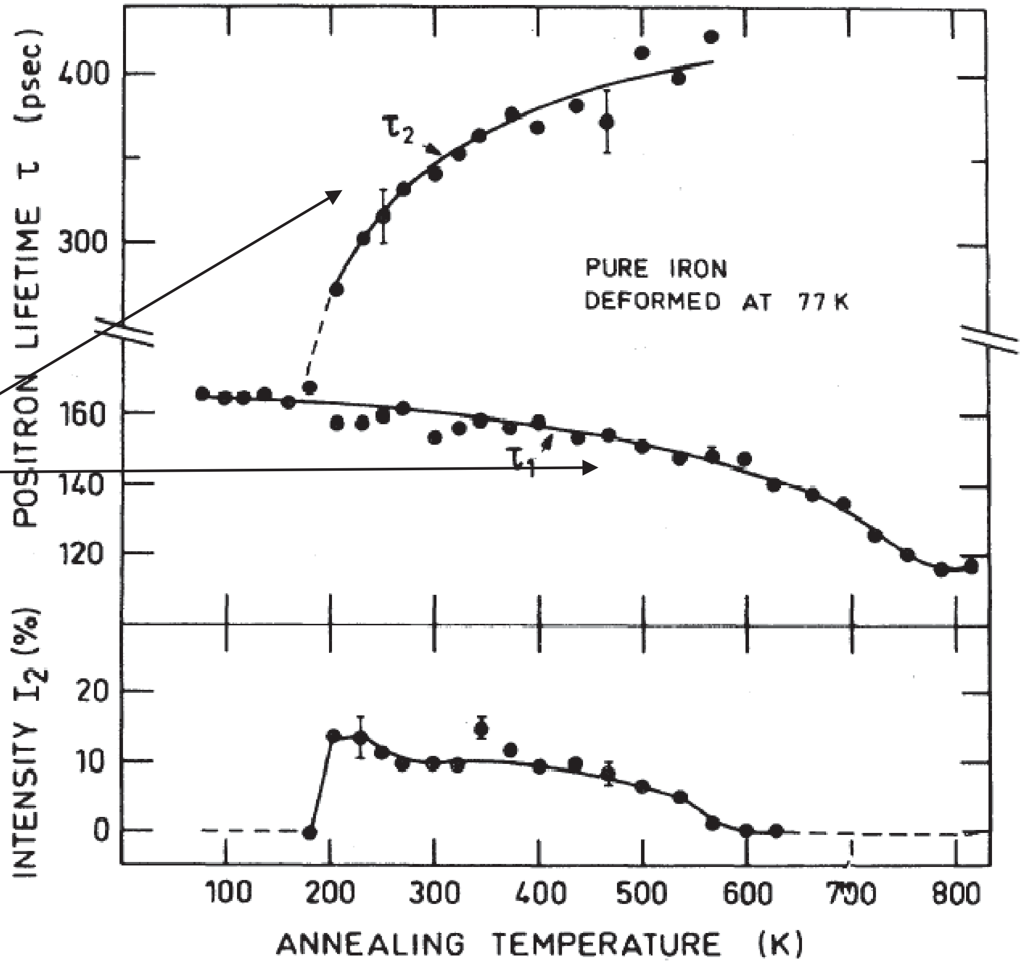
$$\tau_3 = \left[2\left(1 - \frac{R}{R_o} + \frac{1}{2\pi}\sin(2\pi R / R_0) \right) \right]^{-1}$$



Spherical potental well

Annealing of Fe deformed at low temperature

$$D(t) = \sum_{i=1}^{k+1} I_i \exp(-\frac{t}{\tau_i})$$



Vacancies migrate and cluster above ~200 K

Vehanen et al. (1982)

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sensitivity range for measuring defect concentration (Al and Mo)

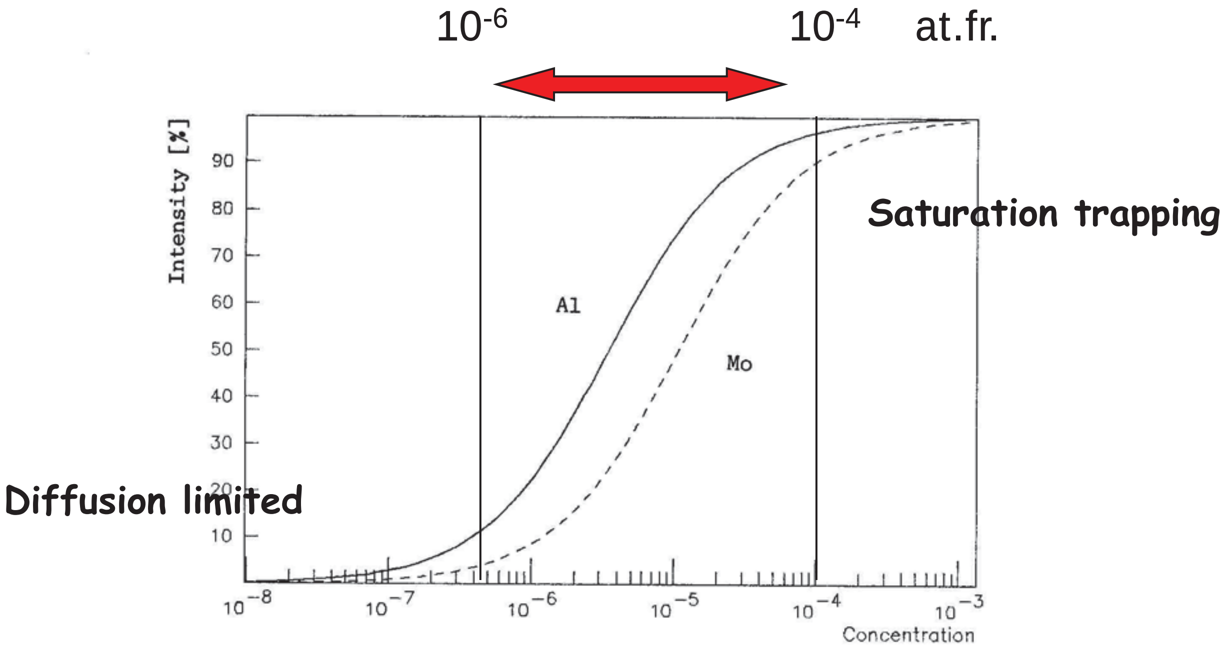

Fig. 1.2 Intensity of the defect lifetime for monovacancies in Al and Mo as a function of the defect concentration (in defects per atom)

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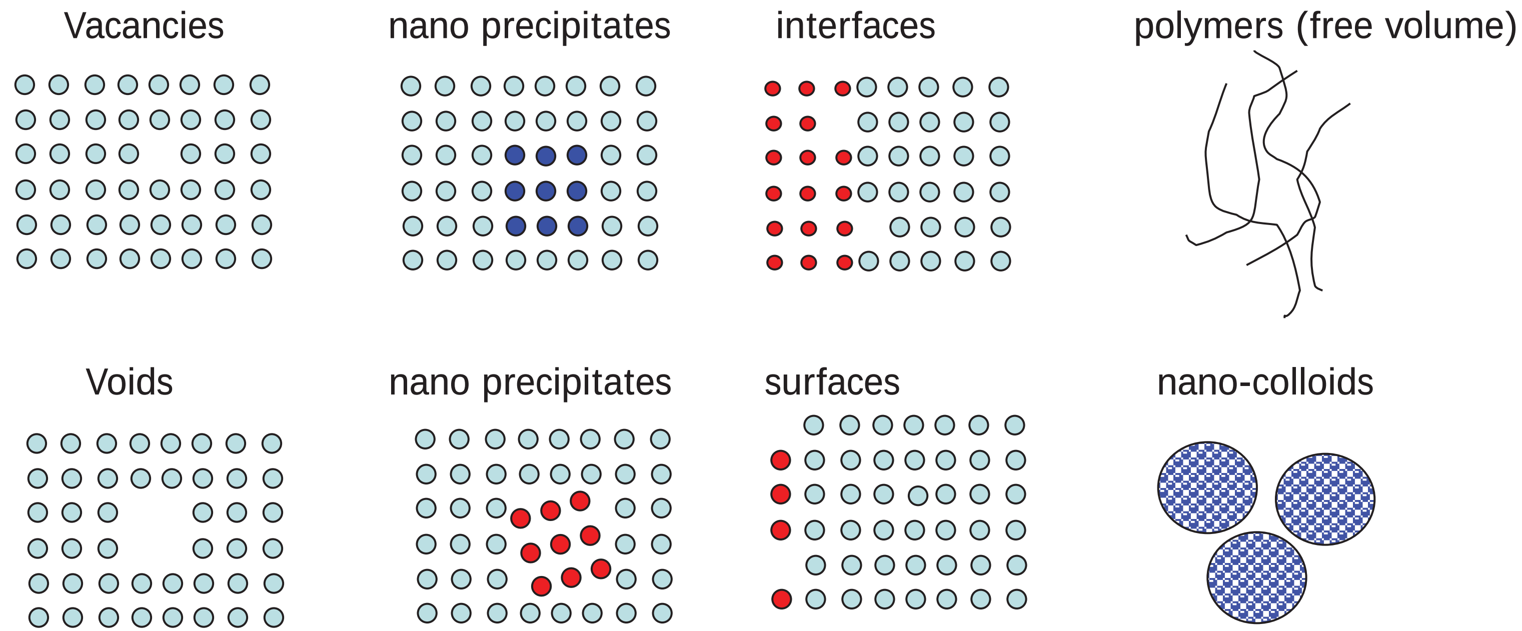
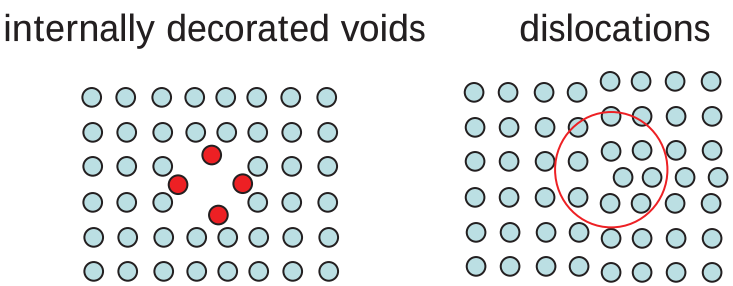
examples of systems studied with positrons





Metals, Alloys

Semiconductors (Si, GaAs, ...

Polymers

Colloids, Ceramics, Metaloxides

.....

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