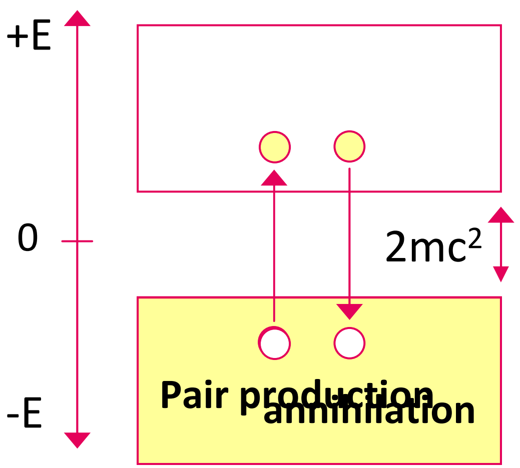
Positron Annihilation in Material Research

- Introduction
- Positron sources, positron beams
- Interaction of positrons with matter
- Annihilation channels: Emission of 1, 2 or 3 γ-quanta
- Annihilation spectroscopies: Lifetime, angular correlations, Doppler broadening
- Study of solid state properties by annihilation
- Medical application: PET (Positron emission tomography) – three-dimensional images of metabolic activity within the human body

Links:
http://www.positronannihilation.net/
http://positron.physik.uni-halle.de/

Diracs prediction of the positron

The positron (e+) as the antiparticle of the electron (e-) with electric charge of +1e, spin of 1⁄2, and the same mass as an electron was predicted by P.A.M. Dirac in 1930 as an interpretation of the negative energy solutions of his relavistic equation of motion for the wavefunction of the electron



$$E = \pm\sqrt{p^2c^2 + m^2c^4}$$

For each quantum state possessing a positive energy *E*, there is a corresponding state with energy *-E*.

Dirac hypothesized that the "vacuum“ is the state in which *all* the negative-energy states are filled (Dirac sea), and all the positive-energy states are empty.

A hole in the sea of negative-energy electrons would respond to electric fields as if it were a positively-charged particle = POSITRON (named by C. D. Andersen)

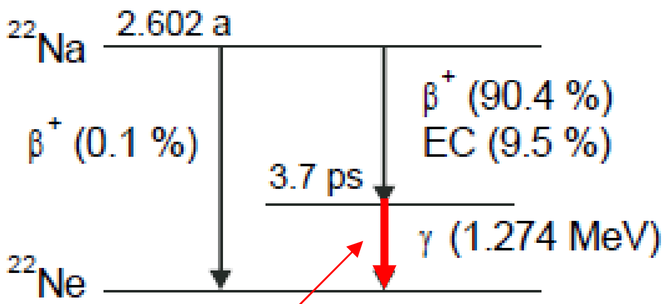
Positron sources

I. Positrons from ß+ decay

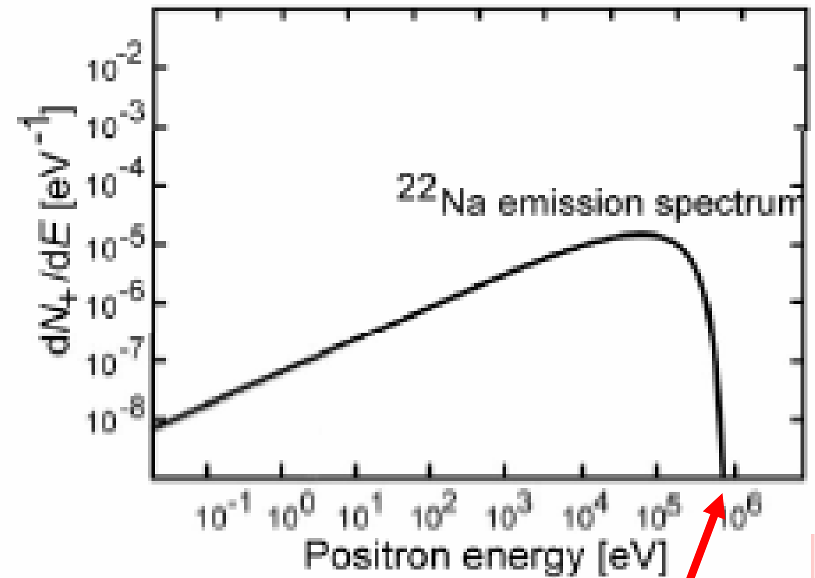
$$p \Rightarrow n + e^{+} + \nu$$

Continuous positron energy distribution

For material research mostly ^{22}Na



marks the birth of e+



E_{max} = 570 keV

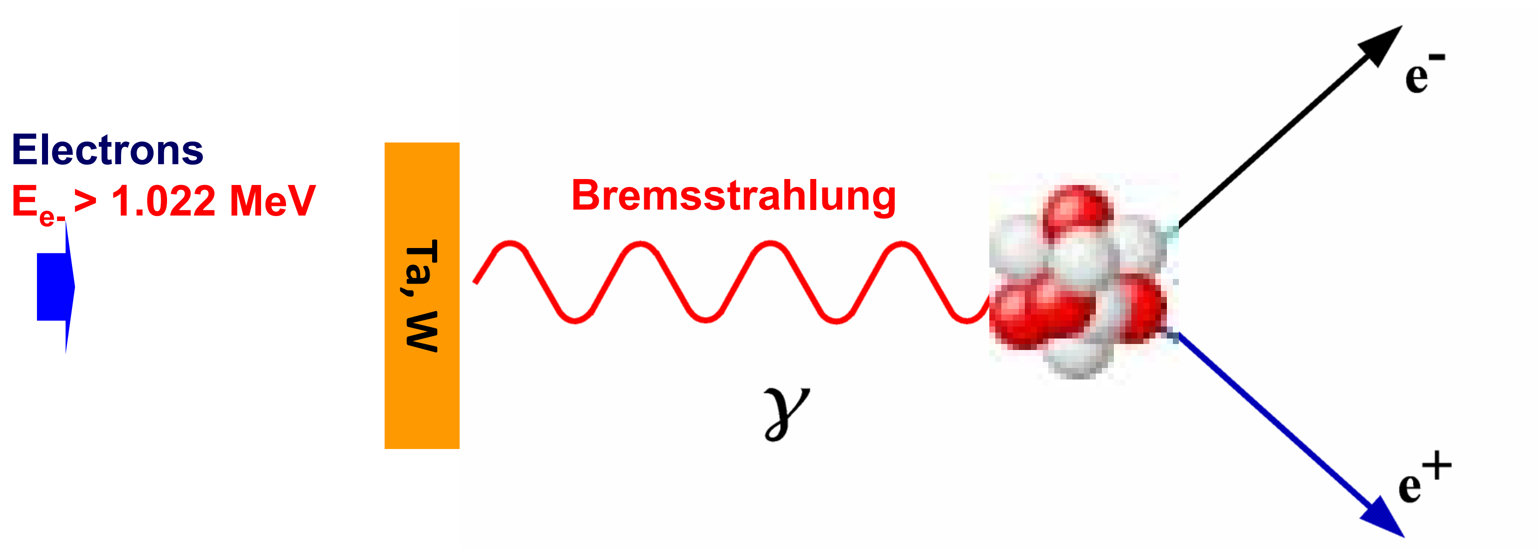
Production

- **Accelerator**
  Mg^{24} (d,α) Na^{22} ; σ = 19 mb @ 32 MeV

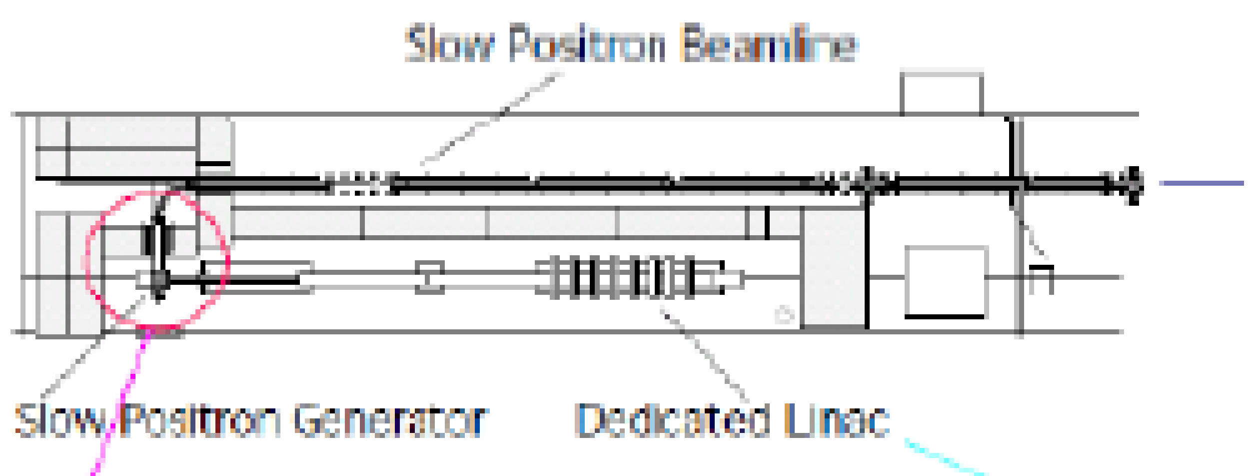
- **Nuclear reactor**
  double reaction: $Li^{6}(n, \alpha)H^{3}$, $Ne^{20}(H^{3}, n)Na^{22}$
  Commercially available up to 4GB (100 mCi)

Positron sources

II. Positrons produced by pair production



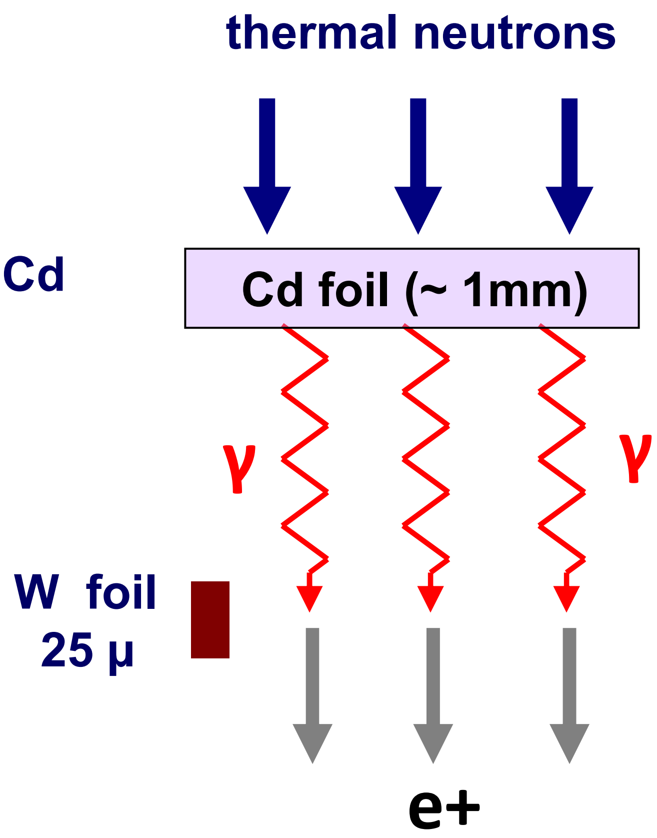
Dedicated system at KEK/Japan

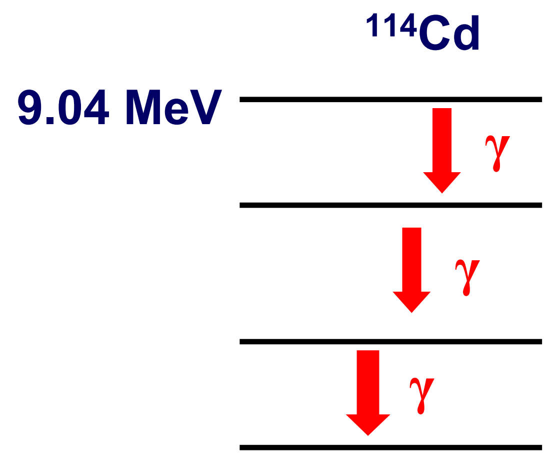


Positron sources

III Positrons from neutron induced pair production
@ FRM II, Munich University, Gemany

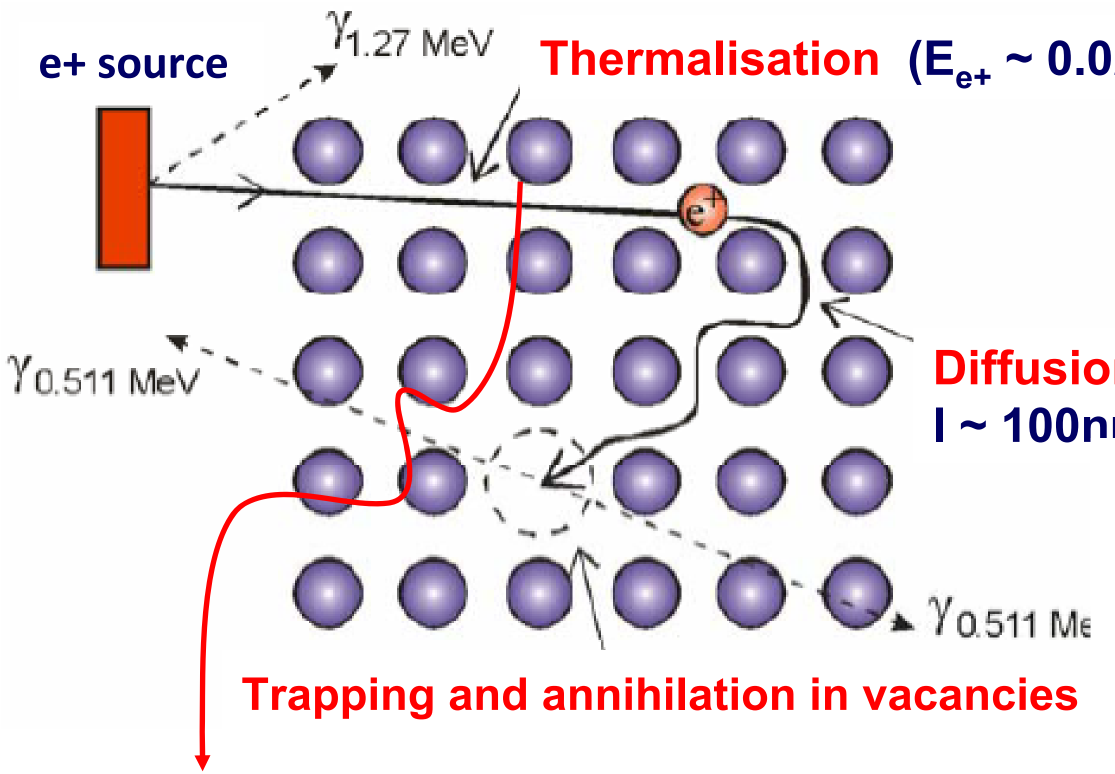
$^{113}Cd\ (n, \gamma)\ ^{114}Cd$





Interaction of positrons with matter

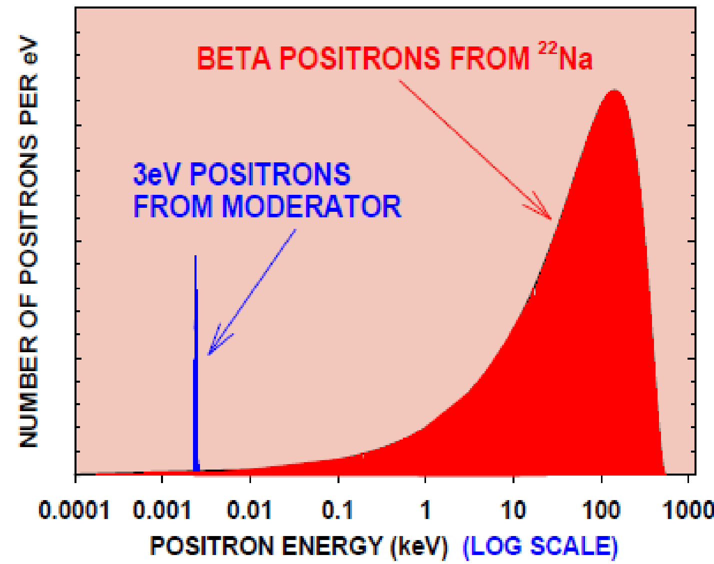
^{22}Na



Thermalisation (E_{e+} ~ 0.025 eV)

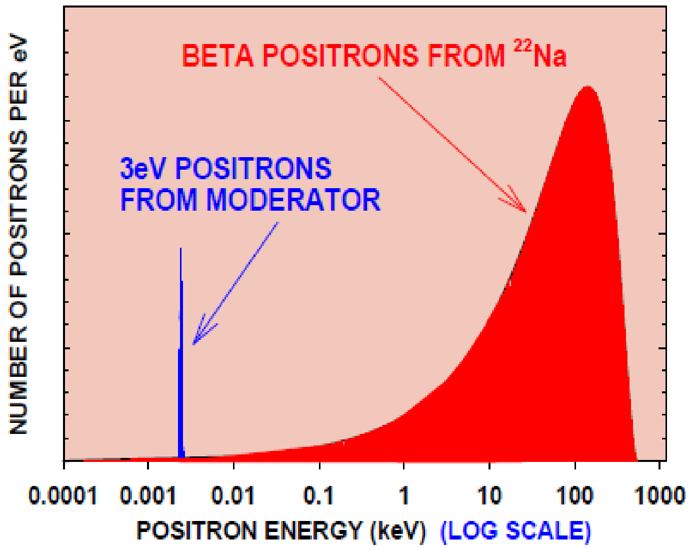
Diffusion over interstial lattice sites
l ~ 100nm

Trapping and annihilation in vacancies



Moderation of high energy positrons:
In metals with negative work function, e.g. W: emission of a few monoenergetic positrons, E_{e+} ~ 3 eV, fraction 0.05 %

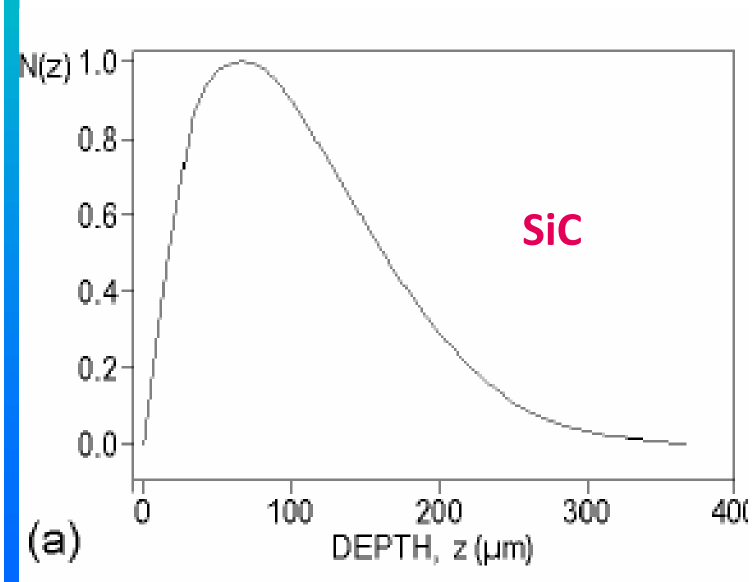
Moderation and variable-energy positron beams



The continuous ß+ spectrum of ^{22}Na

The high ß+ energies allow deep implantation of positrons into solids,

But: The continuous ß+ spectrum results in broad positron depth distributions

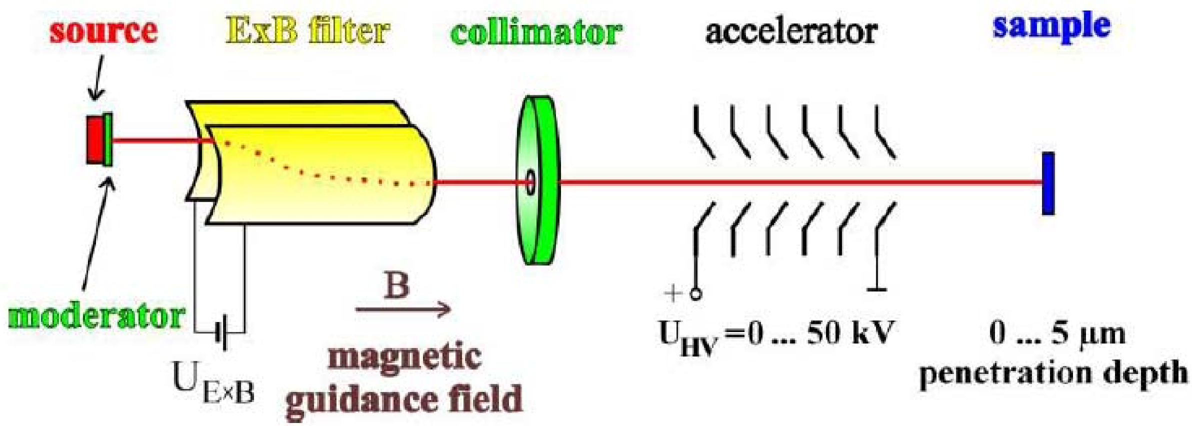


Therefore:

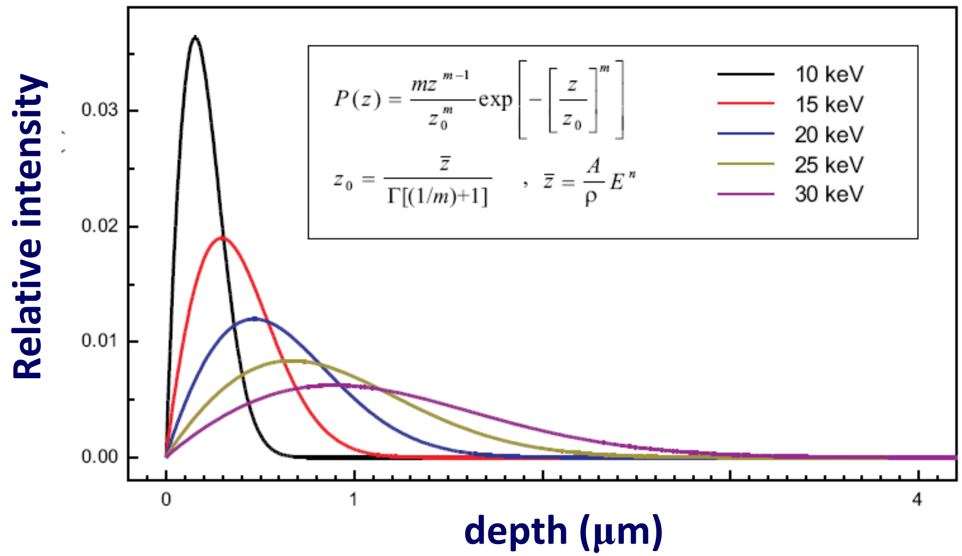
Non-moderated positrons are unsuited for studies of thin layers and near-surface regions

Solution:
Acceleration of moderated (monoenergetic) positrons

Accelerator of moderated positrons at Halle University



Implantation profiles at different energies



Electron-Positron Annihilation

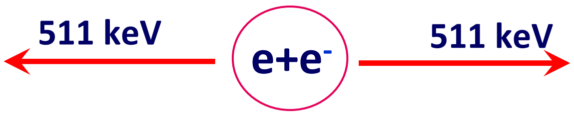
Since the positron is the antiparticle of the electron, it annihilates with the electron by gamma-ray emission liberating an energy of 2x511 = (1024) keV

The annihilation process follows the laws of quantum electrodynamics, conserving **energy, charge, parity, momentum, and angular momentum** of the $e^+ e^-$ pair

Annihilation channels

Emission of 2 photons

is the most probable process. Parity and angular conservation require antiparallel photon spins. In the center of mass system momentum and energy conservation leads to the emission of the 2 antiparallel photons with energy of 511 keV each:



Emission of 1 photon

requires the participation of a third particles, e.g. a nucleus. Compared to 2γ-emission, probability reduced by α =1/137 (fine structure costant).

Emission of 3 photons

is possible, but a factor α^3 less probable than 2-photon emission

2-photon annihilation

I. Life time

The annihilation probability:

$$\lambda = \pi\, c\, r_0^2\, n_e$$

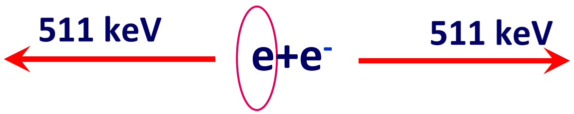
r_0 = classical electron radius

n_e = electron density

The positron life time provides information on the electron density

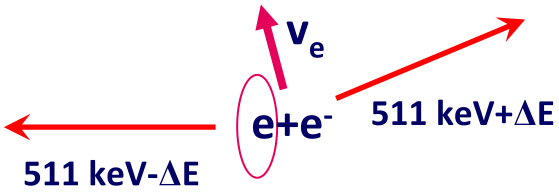
II. Angular distribution

Center-of-mass system



Laboratory system

The e+e- pair has a kinetic energy of the order of 10 eV and a momentum of about p ~10^{-2} m_0c. These are are mainly provided by the electron since the positron is thermalized (E(e+) ~ 1/40 eV) . This leads to changes in the energy and the emission direction of the two photons

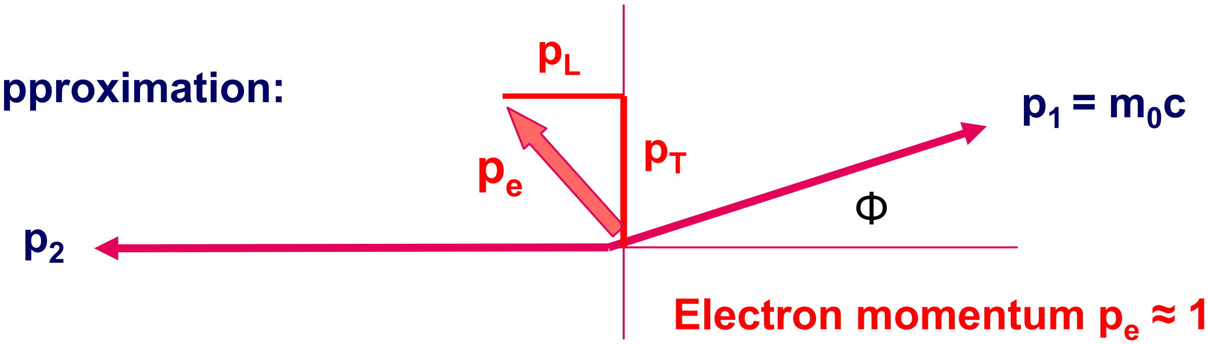


γγ-angular distribution of 2-photon annihilation:

A finite electron momentum leads to deviation from 180-degree emission.

Angular distribution calculated by Rindler (1960) using special relativity

Classical approximation:



Electron momentum $p_e \approx 10^{-2}\, m_0c$

Momentum conservation $\vec{p}_e = \vec{p}_1 - \vec{p}_2$ ⟹ $\sin\Phi \approx p_T / p_{\gamma 1} = p_T / m_0 c \approx 10^{-2}$

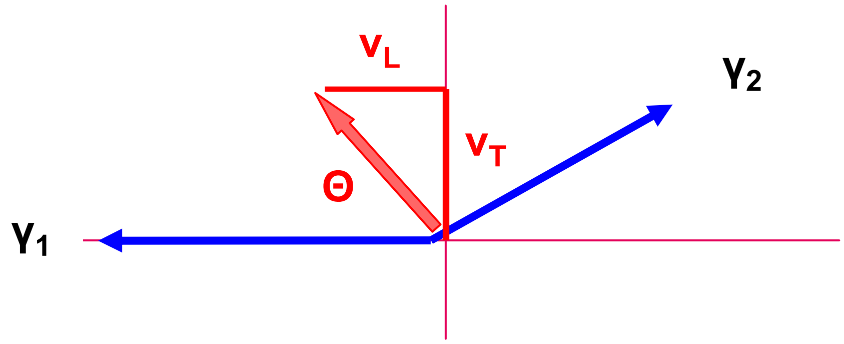
Order of magnitude: $\sin\Phi \approx \Phi = 10^{-2} \approx$ 3/10 degrees

Measurements of the angular distribution provides information on the transversal component of the electron momentum

Doppler shift in 2-photon annihilation

The e+e- pair moves when annihilating, resulting in an **energy shift ΔE (Doppler effect)** of the annihilation radiation

$$E_{1,2} = \frac{E_T}{2}\left(\frac{1 \pm (v/c)\cos\Theta}{(1-(v/c)^2)^{1/2}}\right)$$

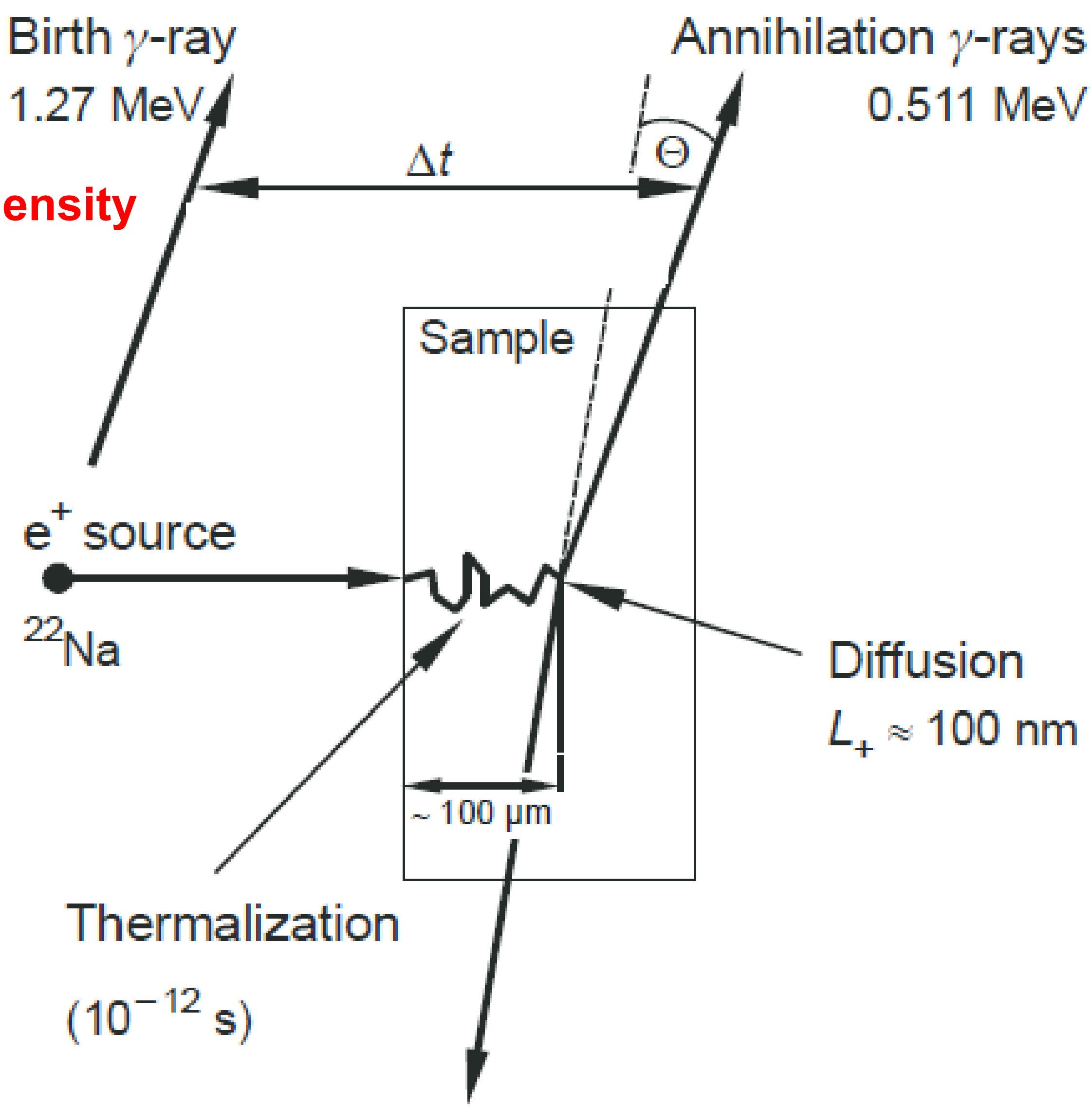


$E_T = 2x\ m_0c^2$; $v \ll c$ ⇒ $$E_{1,2} = m_0c^2 \pm \frac{cp_L}{2}$$

The energy shift $\Delta E = \pm\, cp_L/2$ provides information on the longitudinal momentum component of the the annihilating e+e- pair

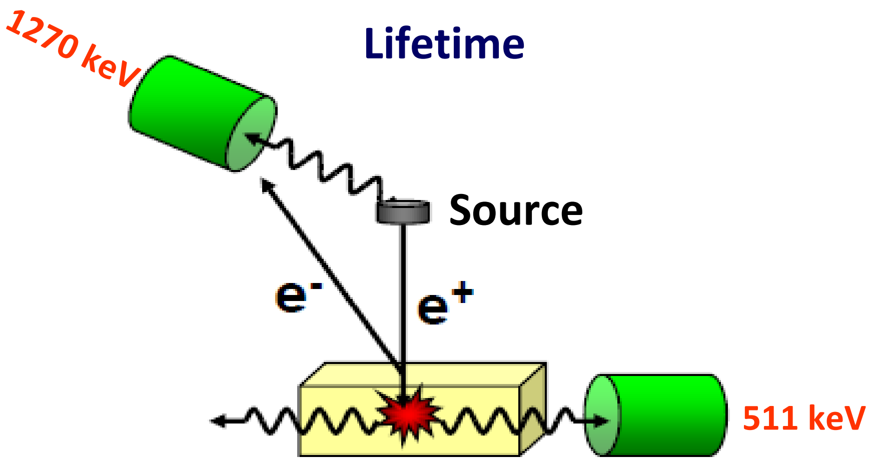
Positron Annihilation Spectroscopies

Electron density

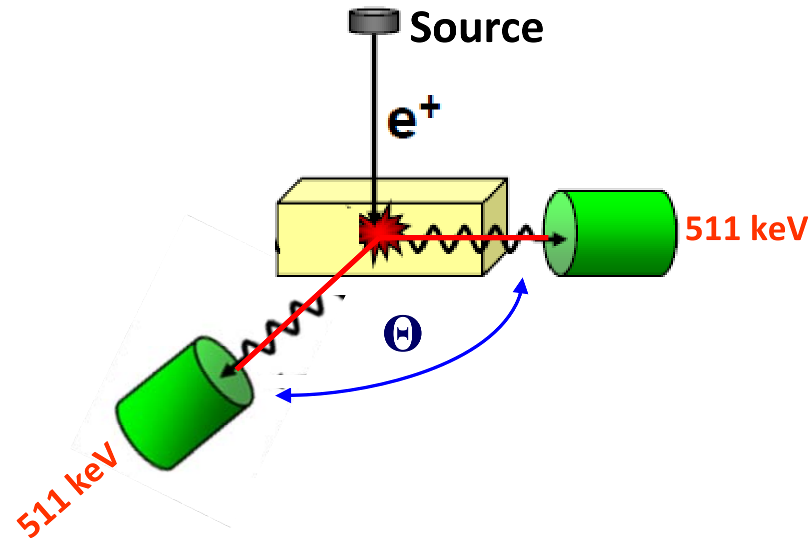


Positron Annihilation Spectroscopies

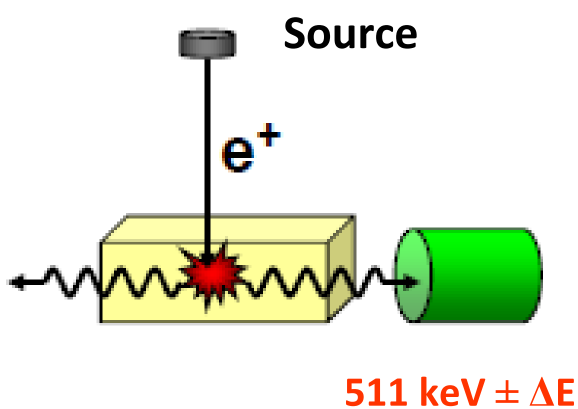
Lifetime



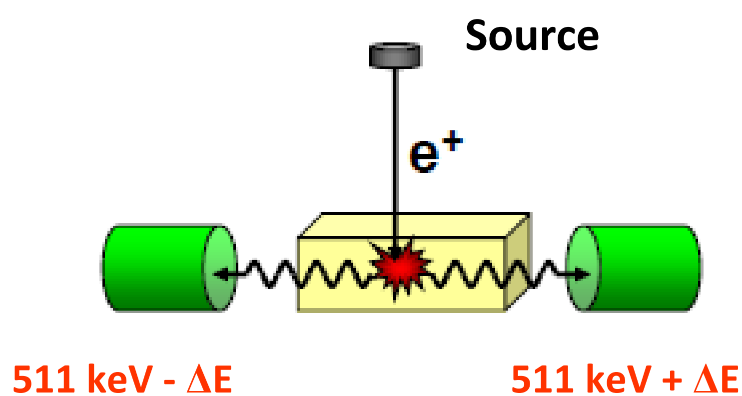
γγ-Angular Distribution



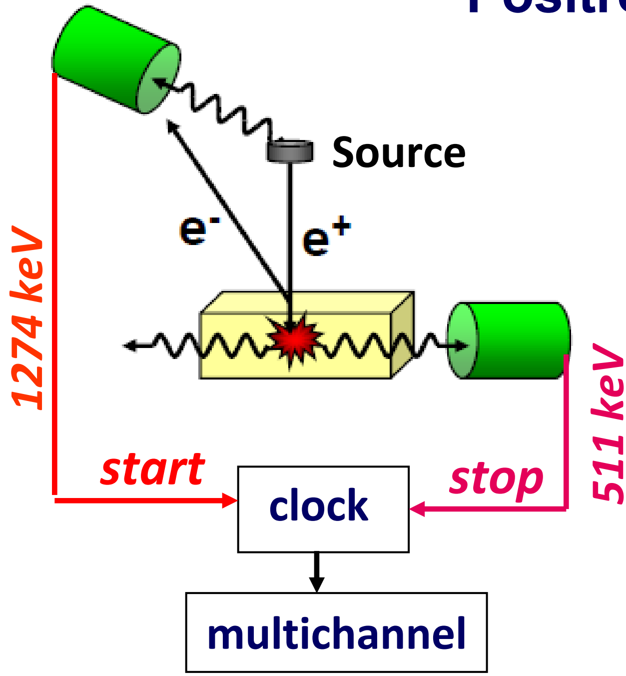
Single Detector Doppler



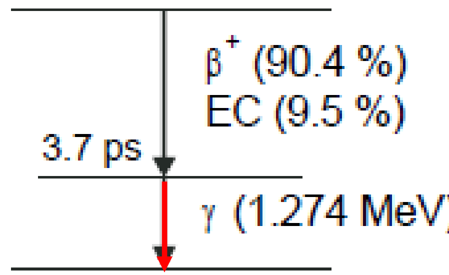
Coincidence Doppler

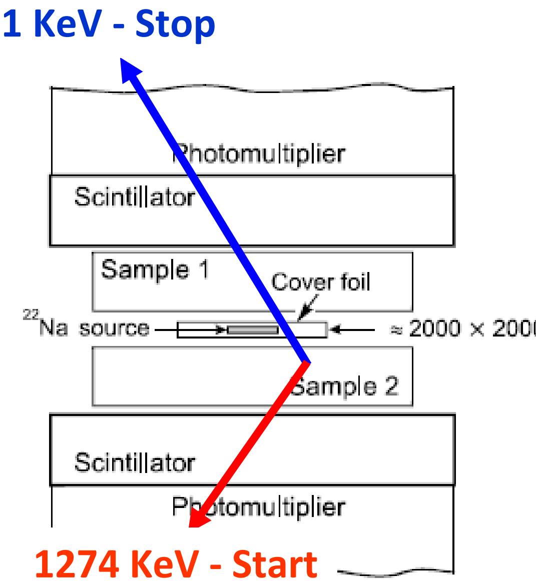


Positron life time spectrometer

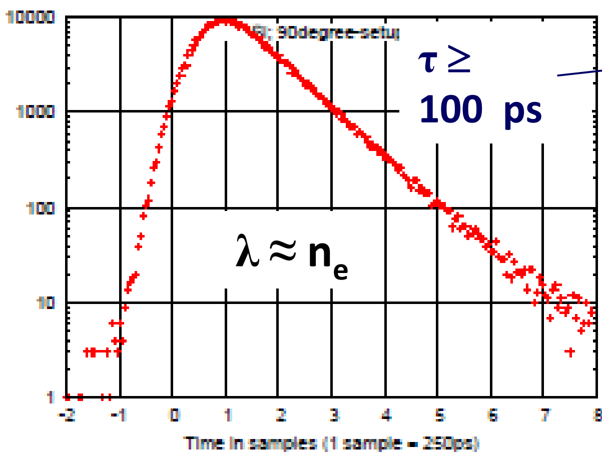


^{22}Na decay

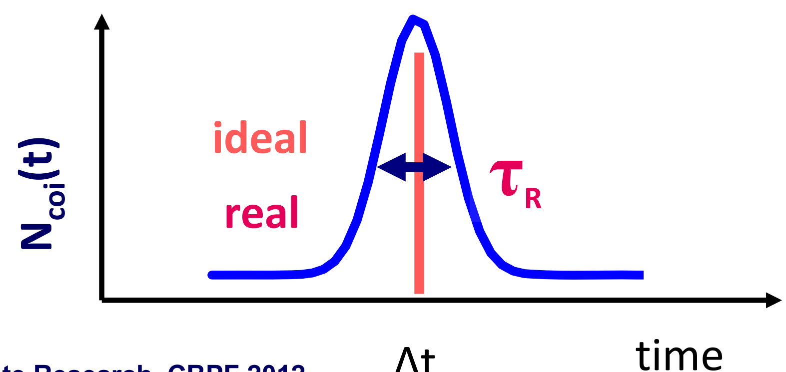




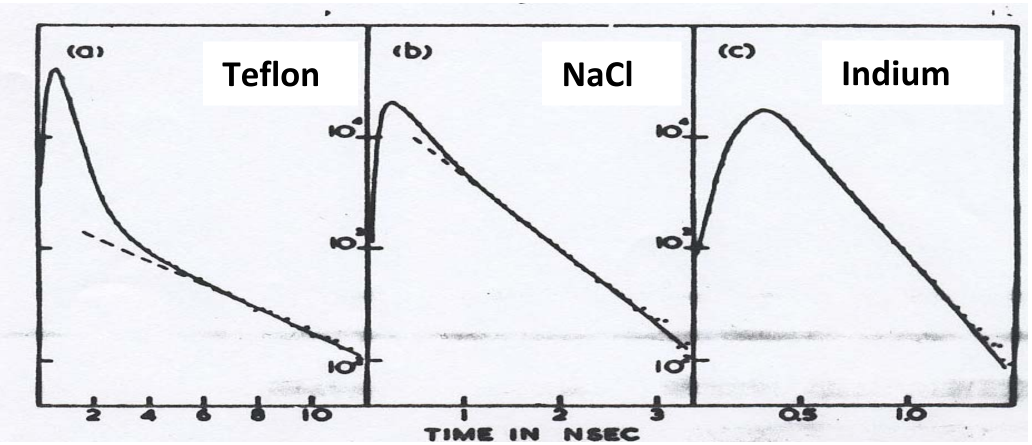
Time spectrum



High time resolution τ_R required

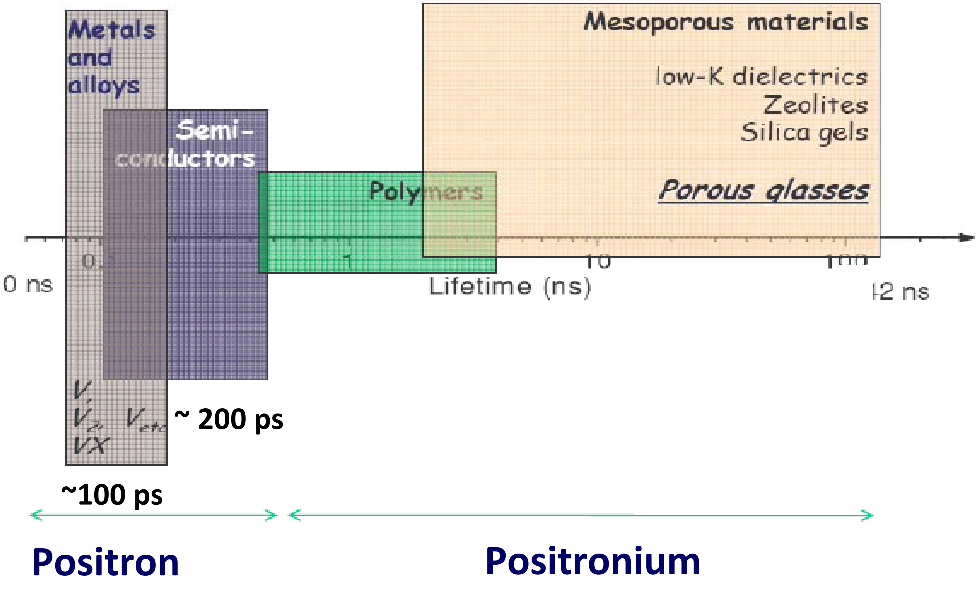


Life time spectra in different materials



Typical Lifetimes

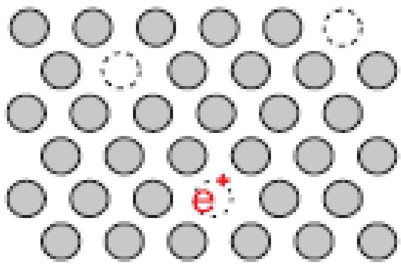
Typical life times



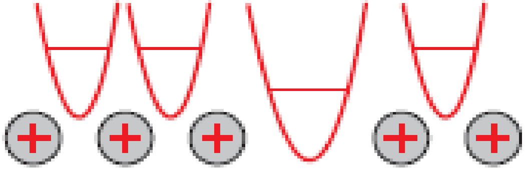
Open-volume defects studied by positron-lifetime measurements

Positrons are sensitive probes for open-volume defects, such as
vacancies and their agglomerates, nanoprecipitates,
nano-porosity, grain boundaries of nano-grains, acceptors

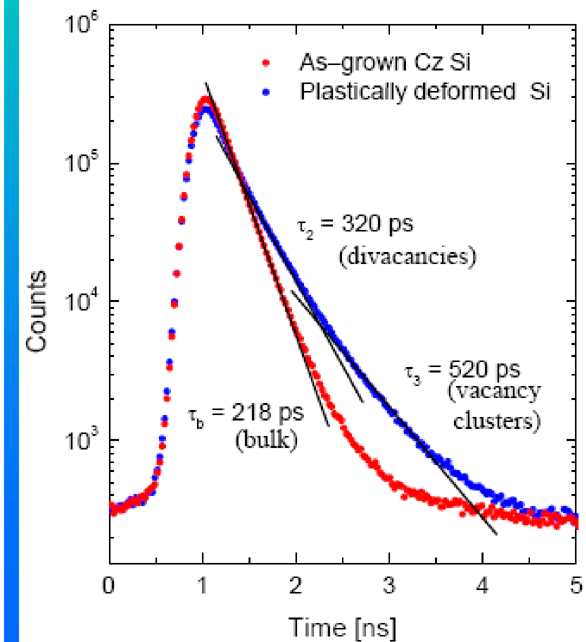
Lattice with vacancies



Potential



Trapped in such defects, positrons experience a smaller electron density and the positron lifetime therefore increases with respect to the defect-free sample.



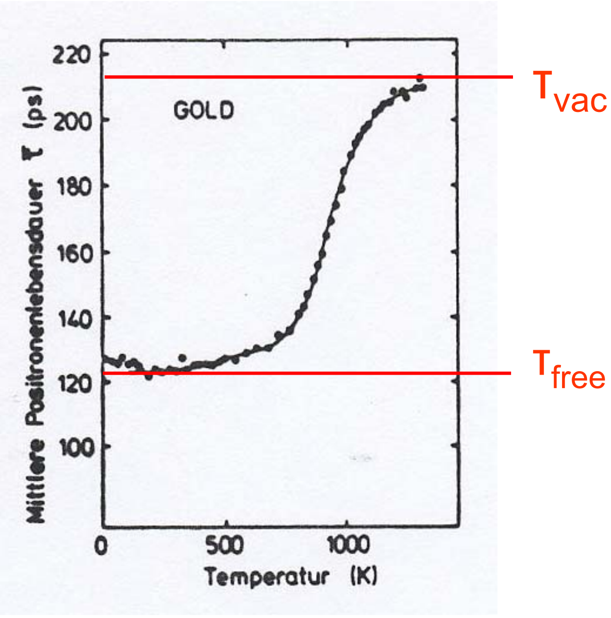
Several exponential decay components in the positron lifetime spectra reflect different defect configurations

Analysis by non-linear fitting: life times τ_i and intensities I_i

$$N(t) = \sum_i \frac{I_i}{\tau_i} \exp(-\frac{t}{\tau_i})$$

Positron-lifetime studies of DEFECTS

(i) Equilibrium Defects – vacancies in gold



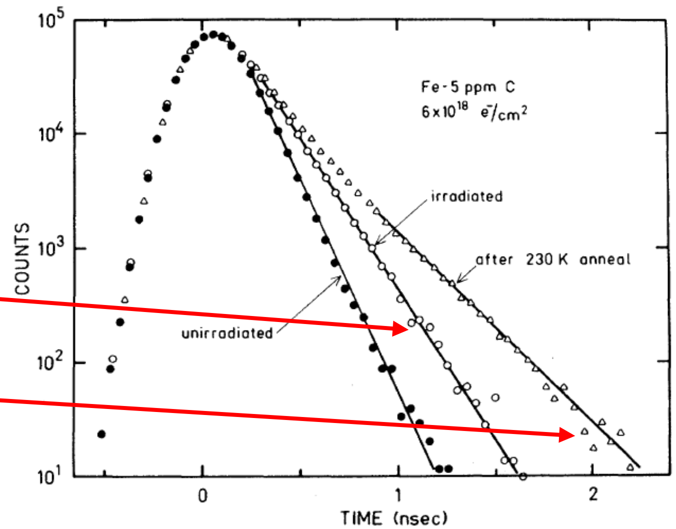
- T< 500 K: Annihilation of free e+ - T_{free}
- Vacancy concentration in equilibrium increases with T:

$$C(T) = C_0 \exp(-\frac{E_V}{k_B T})$$

- T > 1000 K: Annihilation of vacancy-trapped e+ - T_{vac}

→ Vacancy formation energy E_V

(ii) Non-equilibrium defects in Fe

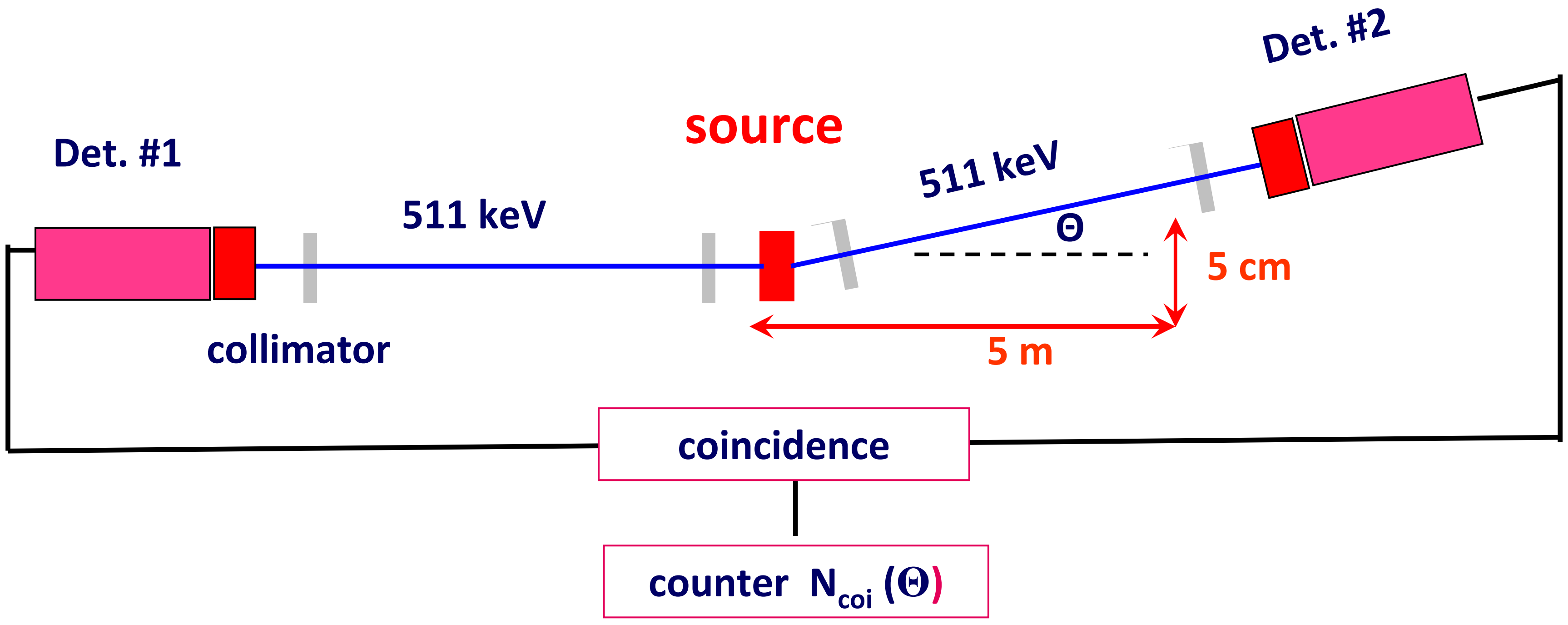


- Electron-irradiation produces vacancies
- e+ life time therefore increases after irradiation
- annealing leads to vacancy clustering
  and further life time increase

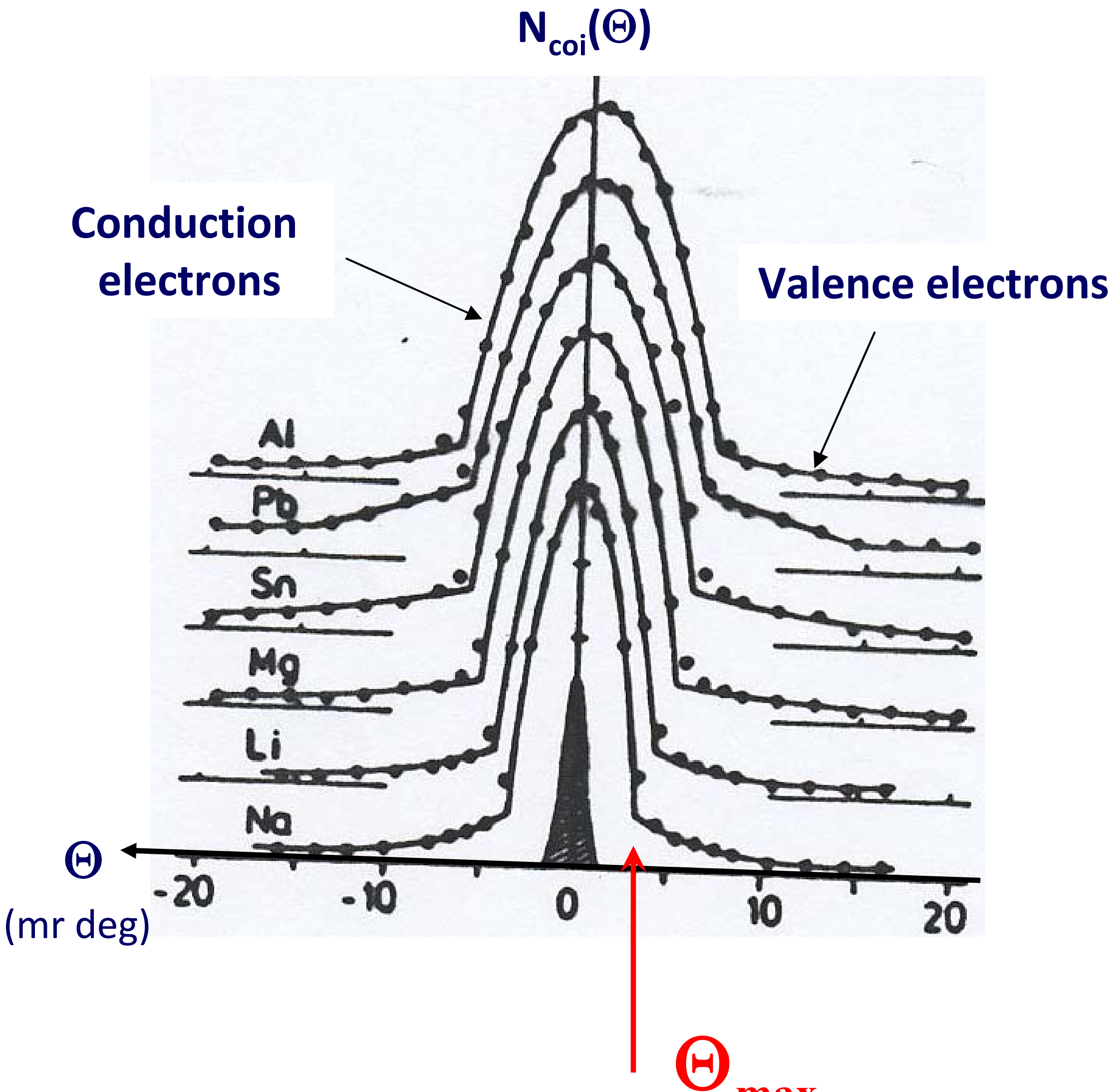
Angular correlation of annihilation radiation-ACAR (1-dimensional)

Determination of transverse electron momentum

$$\sin \Phi \approx p_T / p_{\gamma 1} = p_T / m_0 c \approx 10^{-2}$$



1D-ACAR study of the electronic structure of simple metals



In the free-electron approximation:

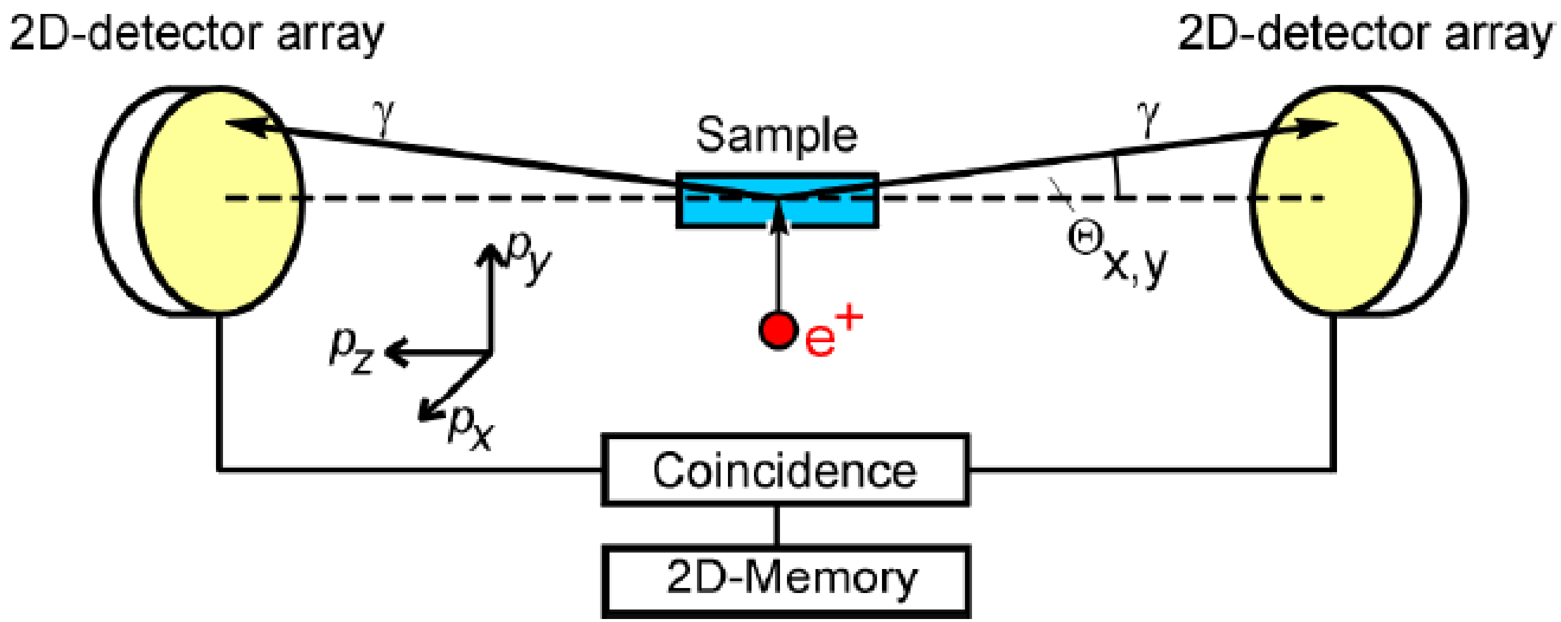
$$\Theta_{max} = \frac{\hbar k_F}{m_e c}$$

2-dimensional ACAR

Measurements of *both* transversal momentum components

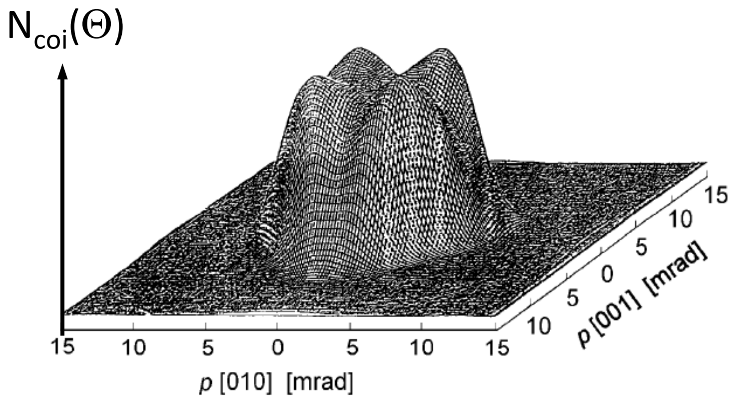
$$\sin \Phi_{x,y} \approx p_{x,y} / m_0 c$$

position sensitive (Pixel) detector



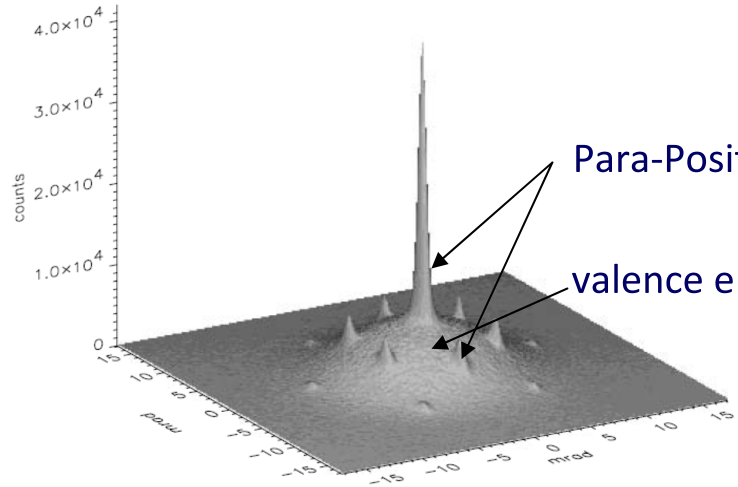
2D-ACAR study of electron moment distributions in solids

Defect-free GaAs



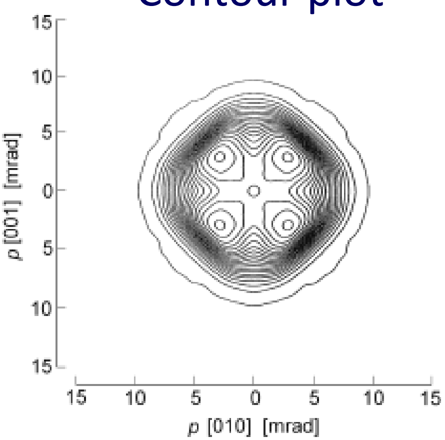
Tanigawa et al., 1995

Quartz SiO_2



M. Biasini (1995)

Contour plot

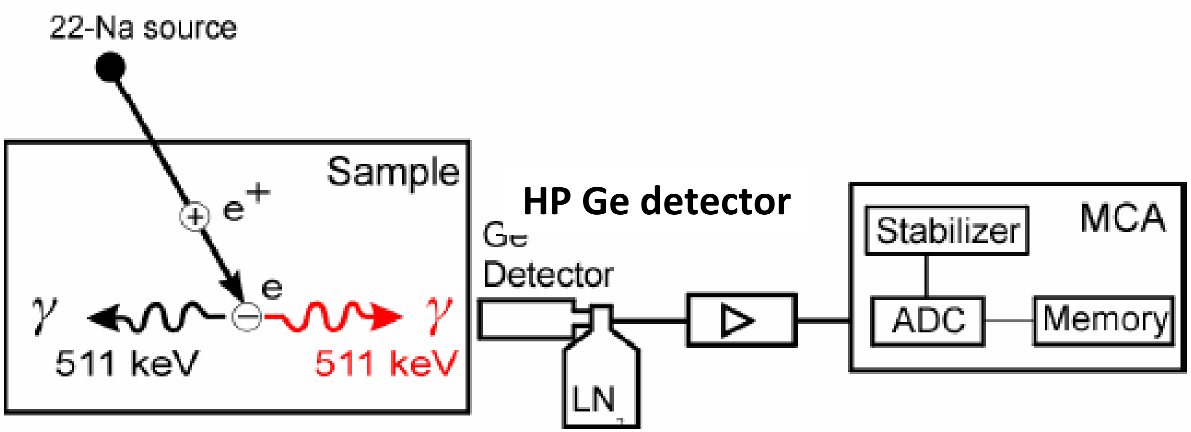


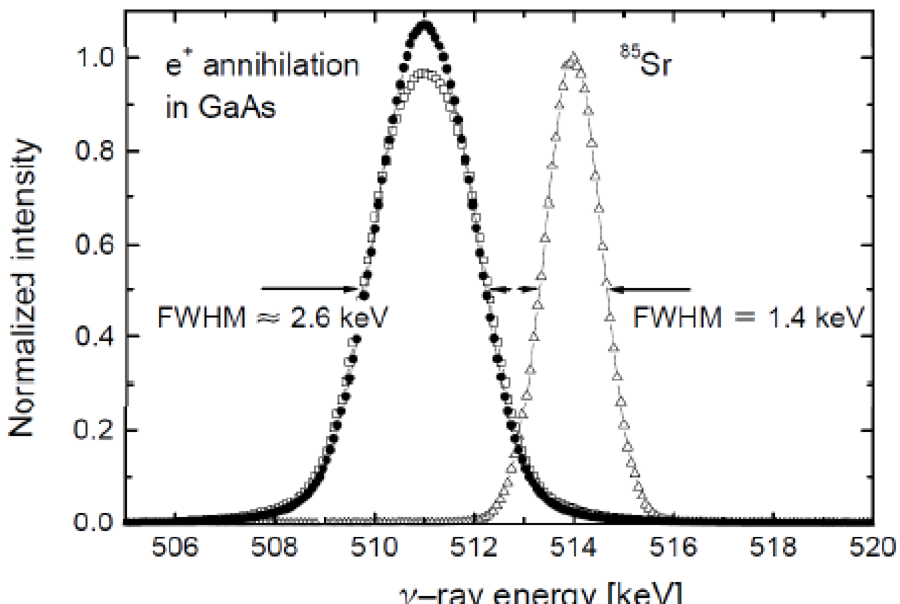
By taking measurements in several directions of a single crystal, the 3-dimensional Fermi surface can be reconstructed

A typical 2D-ACAR measurement may take **several weeks** and contain several hundred million counts !!!!

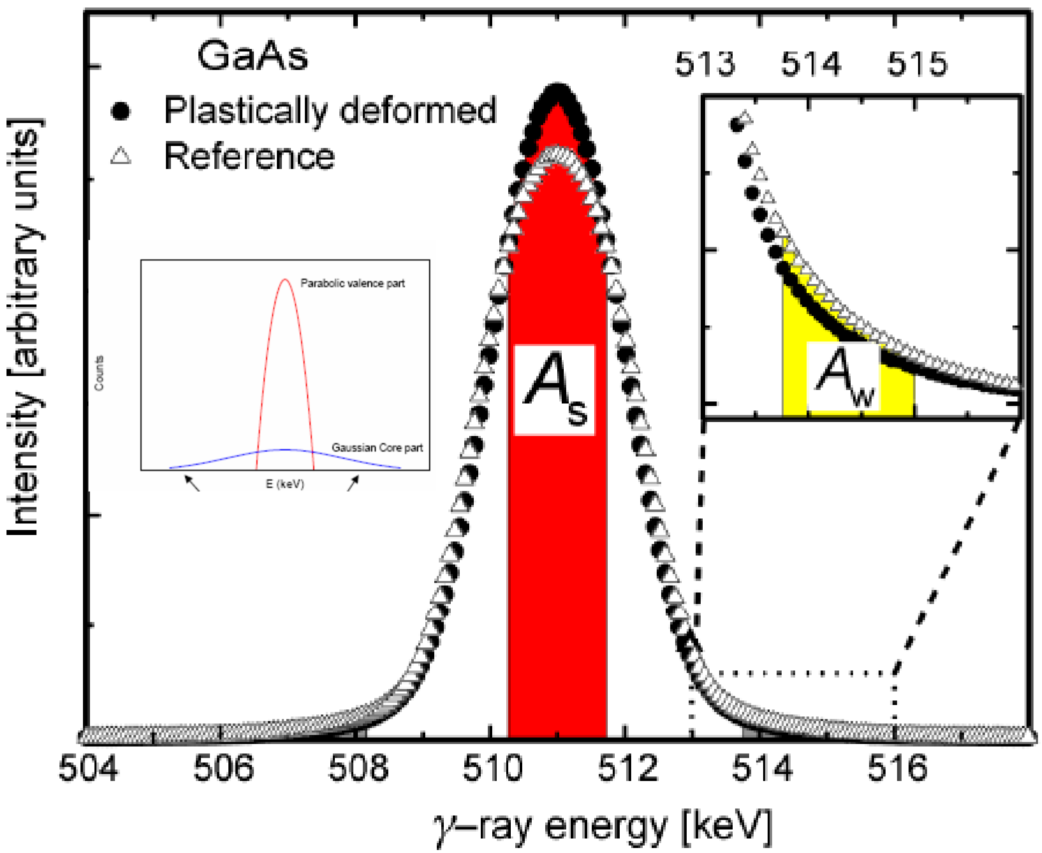
Doppler Broadening Spectroscopy

Measurement of the width of the Doppler-broadened 511 keV annihilation line





The Shape Parameters S, W of the Doppler-broadened Annihilation Line



S parameter

$S = A_S/A_0$

Valence electron (low momentum) annihilation sensitive to open volume defects

W parameter

$W = A_W/A_0$

Core electron (high momentum) annihilation sensitive to the chemical (element) surrounding at the annihilation site

Vacancies in thermal Equilibrium

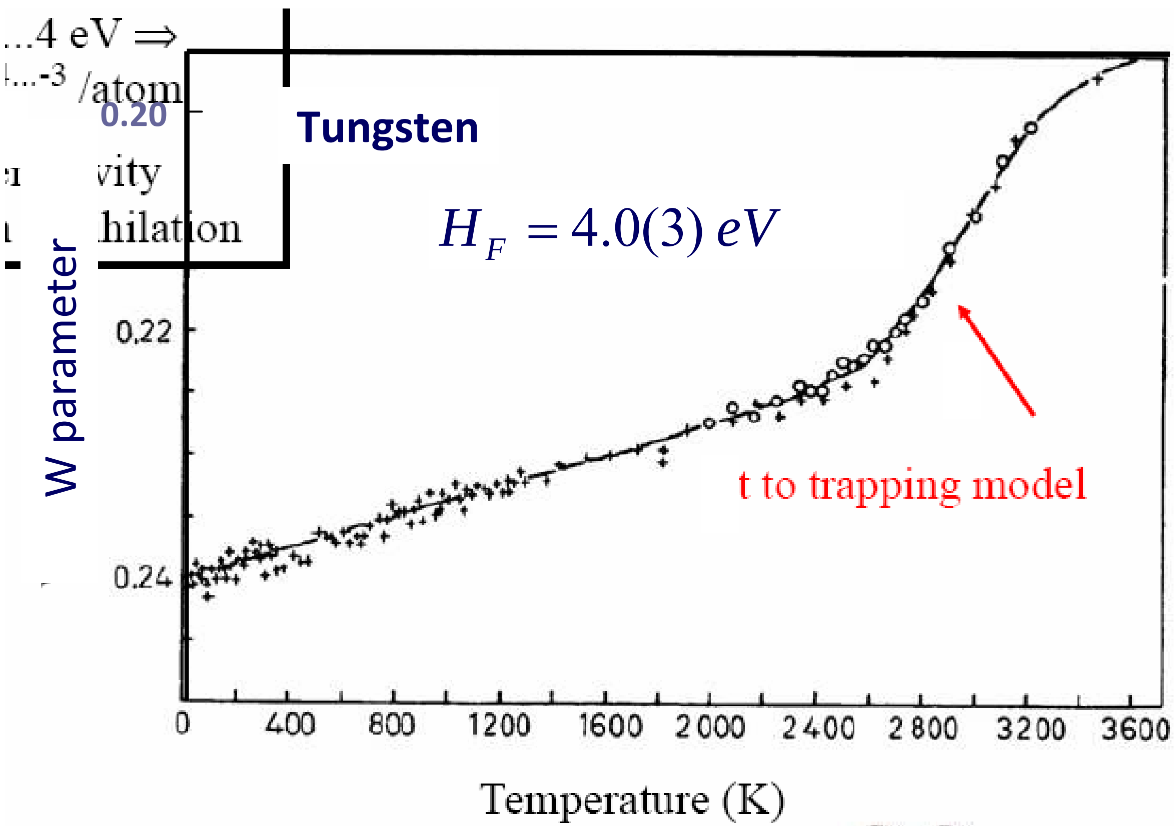
$$C_{1V}(T) = C_0 \exp(-\frac{H_F}{k_B T})$$

H

H_F = **formation enthalpy** of *one* vacancy

$C_{1V}(T_m) \approx 10^{-4...-3}$ /atom

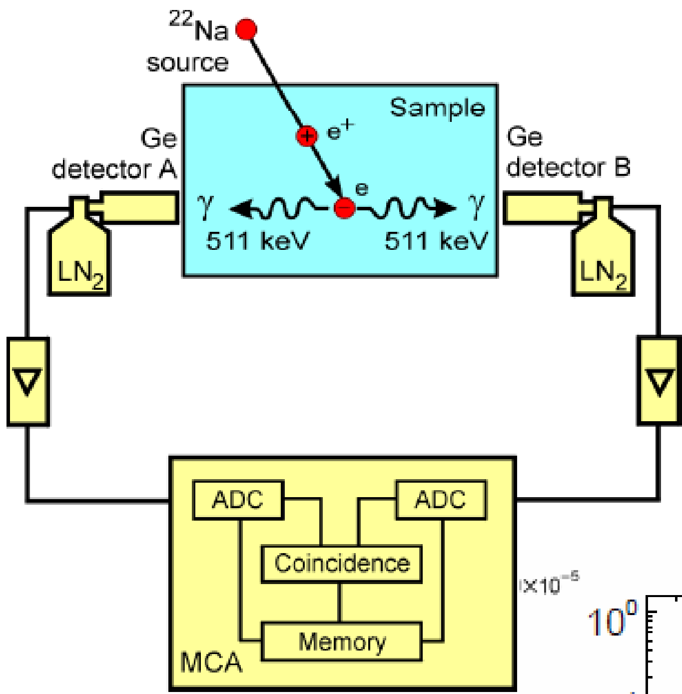
e+ annihilation in vacancies (lower electron momentum)
results in a narrowing of the annihilation line
= increase of the S parameter, decrease of the W parameter

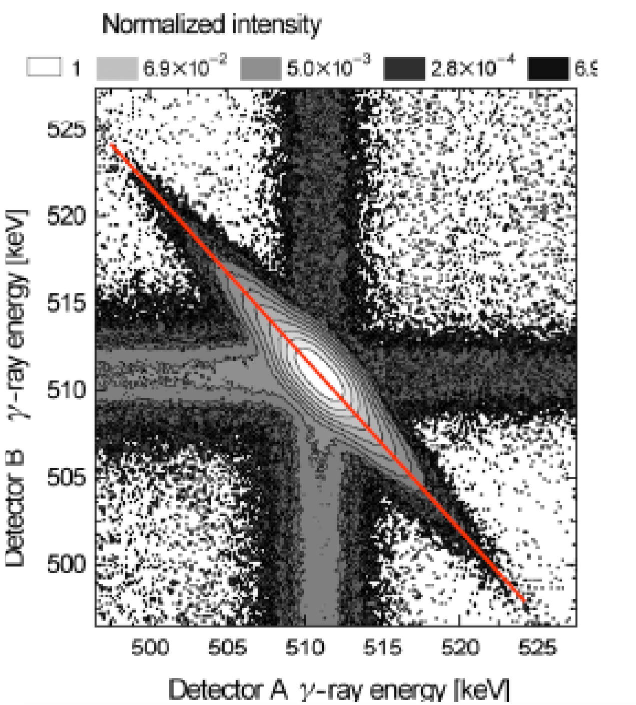


Coincidence Doppler Broadening Spectroscopy

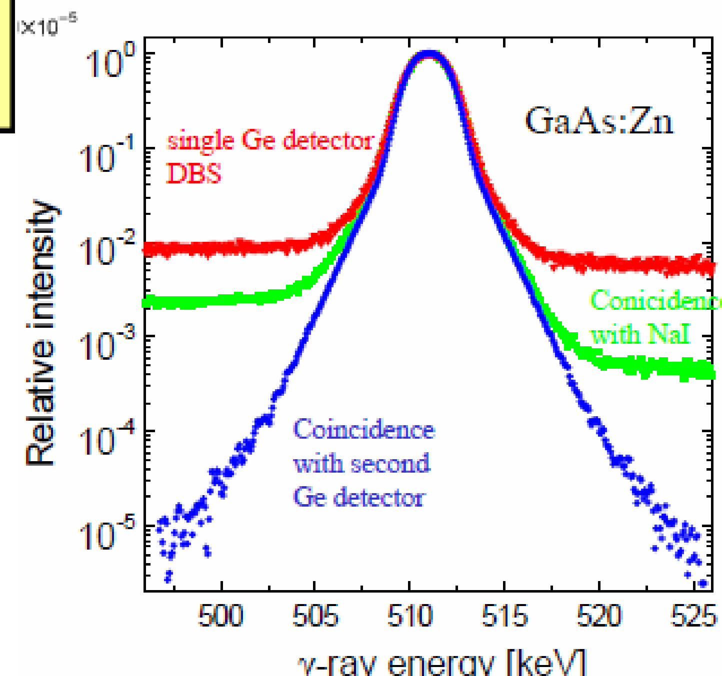
Background Reduction

higher momentum resolution



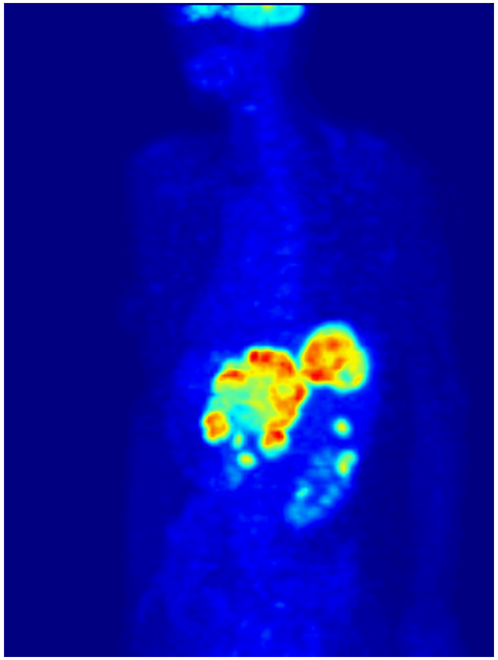


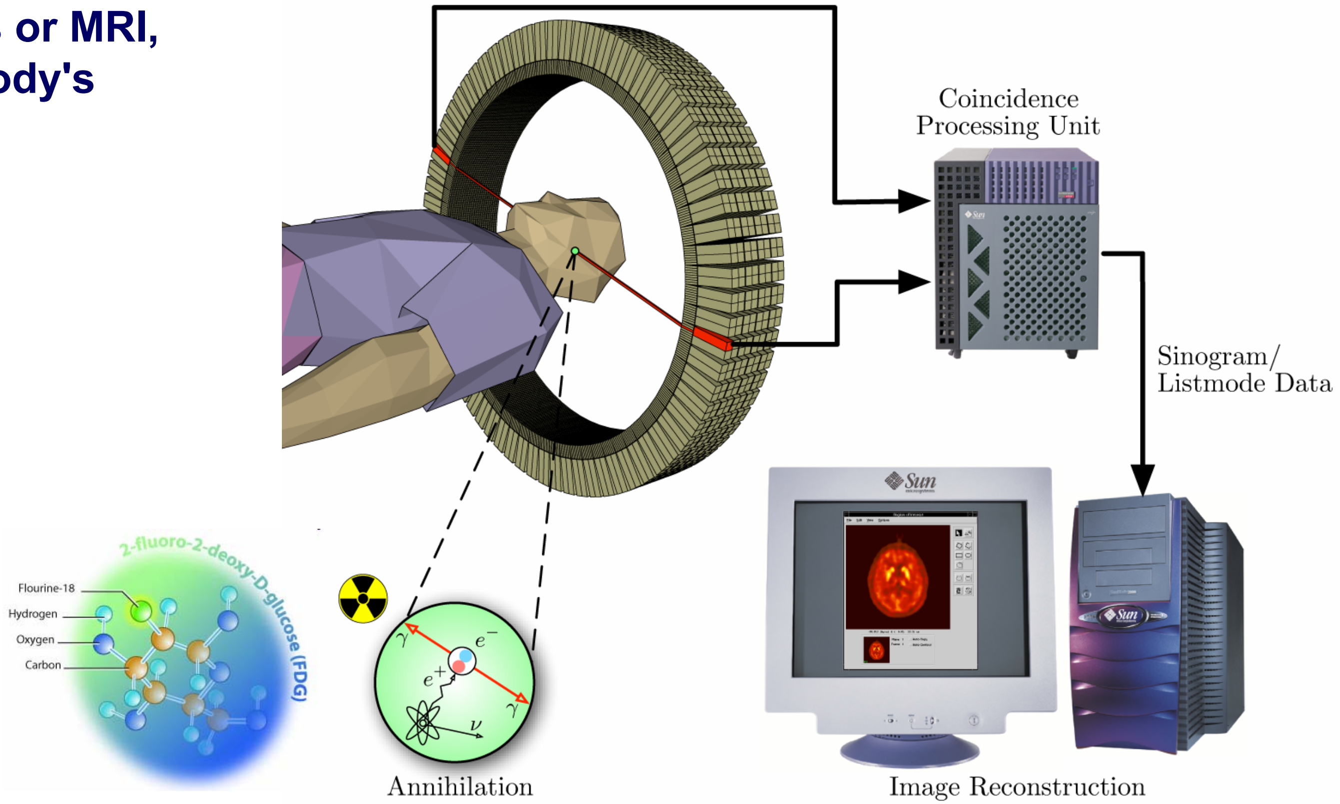
$$E_{\gamma 1}+E_{\gamma 2}= 2\, m_0\, c^2 =1022 \text{ keV}$$



Positron-Emission-Tomography (PET)

Traditional diagnostic techniques, such as x-rays, CT scans or MRI, produce images of the body's anatomy or structure.





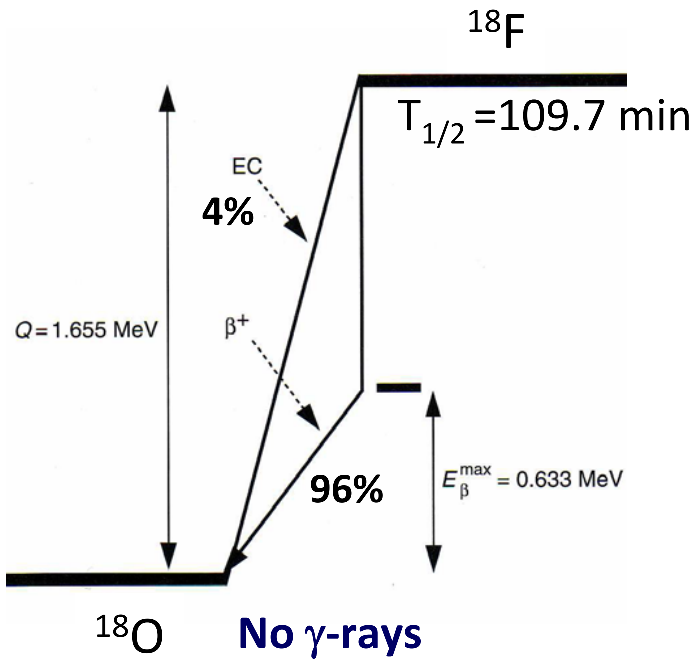
Source: Wikipedia

^{18}F- 2-Fluor-2-desoxy-D-glucose (2-FDG) PET scan

Image of the local glucose consumption

PET produces images of the body's basic biochemistry or function

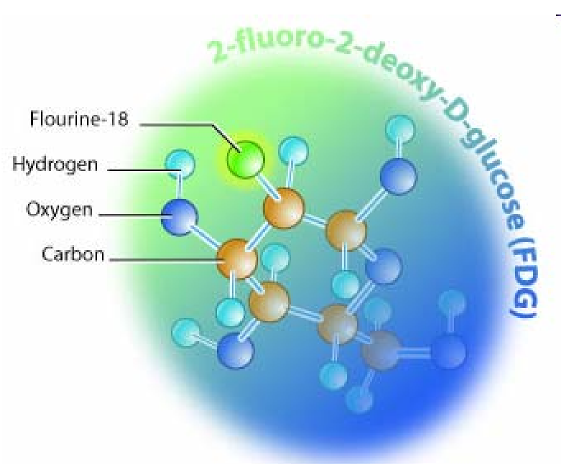
Positron emitters used in PET



Radionuclide	*Half life*	*Nuclear reaction*
Oxygen-15	2.073 min	$^{14}N(d,n)^{15}O$
Nitrogen-13	9.95 min	$^{16}O(p,a)^{13}N$
Carbon-11	20.3 min	$^{14}N(p,a)^{11}C$
Fluorine-18	109.7 min	$^{18}O(p,n)^{18}F$

Short-lived, nearby cyclotron required

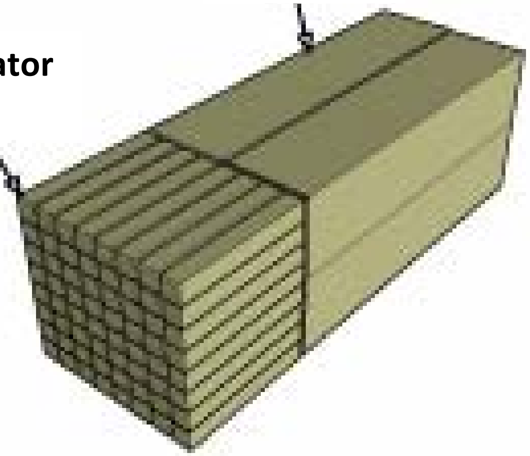
^{18}F- 2-fluoro-2-deoxy-D-glucose



PET's most important clinical role is in oncology, with fluorine-18 as the tracer, since it has proven to be the most accurate non-invasive method of detecting and evaluating most cancers. It is also well used in cardiac and brain imaging.

PET detector system

Up to 6 detector rings
30-40 **detector modules** each



Detector module:

4-8 Detector blocks per module
1 Photomultiplier and 4x4 to 6x6 scintillators per block
Scintillator dimensions: ´(6-8)x(6-8x)(20x30) mm

Scintillators used for PET

„BGO“ : $Bi_4Ge_3O_{12}$
„LYSO“: $LuYSiO_5:Ce^{3+}$
„LSO“, $Lu_2SiO_5:Ce^{3+}$

TOTAL : *up to 10.000 scintillators , 1000 photomultipliers*

Costs (equipment, operation, personel, etc): ~ 1000 US $/scan

Break even: ~ 60 scans/month