Positron Annihilation in Material Research

• Introduction
• Positron sources, positron beams
• Interaction of positrons with matter
• Annihilation channels: Emission of 1, 2 or 3 $\gamma$-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 relativistic 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 β+ decay

\[ p \rightarrow n + e^+ + \nu \]

For material research mostly \(^{22}\text{Na}\)

Production

- **Accelerator**
  \(^{24}\text{Mg} (d,\alpha) \text{Na}^{22}\); \(\sigma = 19\ \text{mb} @ 32\ \text{MeV}\)

- **Nuclear reactor**
  double reaction: \(\text{Li}^6(n, \alpha)\text{H}^3\), \(\text{Ne}^{20}(\text{H}^3, n)\text{Na}^{22}\)
  Commercially available up to 4GB (100 mCi)
II. Positrons produced by pair production

Electrons $E_{e^-} > 1.022$ MeV

Bremsstrahlung

Dedicated system at KEK/Japan
III Positrons from neutron induced pair production
@ FRM II, Munich University, Germany

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

$\gamma$-foil (~ 1mm)

$\gamma$

$\gamma$

$\gamma$

W foil
25 $\mu$

e+$

$^{114}\text{Cd}$

9.04 MeV
Interaction of positrons with matter

\[ ^{22}\text{Na} \]

- **e\(^+\) source**
- **Thermalisation** \((E_{e^+} \sim 0.025 \text{ eV})\)
- **Diffusion** over interstitial lattice sites \(l \sim 100\text{nm}\)
- **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, \(E_{e^+} \sim 3 \text{ eV}\), fraction 0.05%
Moderation and variable-energy positron beams

The continuous $\beta^+$ spectrum of $^{22}\text{Na}$

The high $\beta^+$ energies allow deep implantation of positrons into solids,

But: The continuous $\beta^+$ 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

\[ P(z) = \frac{m_2^{m-1}}{z_0^m} \exp \left( -\frac{z}{z_0} \right)^m \]

\[ z_0 = \frac{\bar{z}}{\Gamma(1/m+1)} \]

Relative intensity

depth (\(\mu m\))
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 $2 \times 511 = (1024) \text{ keV}$

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 $2\gamma$-emission, probability reduced by $\alpha = 1/137$ (fine structure constant).

Emission of 3 photons
is possible, but a factor $\alpha^3$ less probable than 2-photon emission.
2-photon annihilation

I. Life time
The annihilating probability:
\[ \lambda = \pi c r_0^2 n_e \]
\[ r_0 = \text{classical electron radius} \]
\[ n_e = \text{electron density} \]

The positron life time provides information on the electron density

II. Angular distribution

Center-of-mass system
\[ e^+e^- \]
\[ 511 \text{ keV} \]

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

\[ e^+e^- \]
\[ 511 \text{ keV} \]
\[ 511 \text{ keV} + \Delta E \]
\[ 511 \text{ keV} - \Delta E \]

M. Forker, Nuclear Techniques in Solid State Research, CBPF 2012
**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:

\[
\begin{align*}
\mathbf{p}_L & \quad \quad \quad \quad \mathbf{p}_T \\
\mathbf{p}_e & \quad \quad \quad \quad \mathbf{p}_1 = m_0c
\end{align*}
\]

Electron momentum \( p_e \approx 10^{-2} m_0 c \)

Momentum conservation \( \mathbf{p}_e = \mathbf{p}_1 - \mathbf{p}_2 \)

\[
\sin \Phi \approx \frac{p_T}{p_{\gamma 1}} = \frac{p_T}{m_0c} \approx 10^{-2}
\]

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 $\Delta E$ (Doppler effect) of the annihilation radiation.

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

$E_T = 2x m_0c^2 ; v \ll c$

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

Electron density
Positron Annihilation Spectroscopies

**Lifetime**

- 1270 keV

**γγ-Angular Distribution**

- Source
- $e^+$
- 511 keV

**Single Detector Doppler**

- Source
- $e^+$
- 511 keV $\pm \Delta E$

**Coincidence Doppler**

- Source
- $e^+$
- 511 keV
- 511 keV $- \Delta E$
- 511 keV $+ \Delta E$
Positron life time spectrometer

Source → clock → start → stop → multichannel

1274 keV

$\beta^+ (90.4\%)$

$EC (9.5\%)$

$\gamma (1.274 \text{ MeV})$

$\tau \geq 100 \text{ ps}$

$\lambda \approx n_e$

$511 \text{ keV - Stop}$

$22\text{Na}$ decay

$22\text{Na}$ source → Sample 1 → Cover foil → Sample 2 → Scintillator → Photomultiplier

1274 keV - Start

Time spectrum

High time resolution $\tau_R$ required

$N_{coll}(t)$

ideal

real

$\tau_R$

$\Delta t$

time
Life time spectra in different materials

Teflon, NaCl, Indium

Typical lifetimes

~100 ps

~200 ps

Positron, Positronium
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 $\tau_i$ and intensities $I_i$

$$N(t) = \sum_i \frac{I_i}{\tau_i} \exp\left(-\frac{t}{\tau_i}\right)$$
(i) Equilibrium Defects – vacancies in gold

- \( T < 500 \text{ K} \): Annihilation of free e+ - \( T_{\text{free}} \)
- Vacancy concentration in equilibrium increases with T:
  \[ C (T) = C_0 \exp\left(-\frac{E_V}{k_B T}\right) \]
- \( T > 1000 \text{ K} \): Annihilation of vacancy-trapped e+ - \( T_{\text{vac}} \)

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

\[
\sin \Phi \approx \frac{p_T}{p_{\gamma1}} = \frac{p_T}{m_0c} \approx 10^{-2}
\]
1D-ACAR study of the electronic structure of simple metals

In the free-electron approximation:

$$\Theta_{\text{max}} = \frac{\hbar k_F}{m_e c}$$
2-dimensional ACAR

Measurements of both transversal momentum components

\[
\sin \Phi_{x,y} \approx \frac{p_{x,y}}{m_0 c}
\]

(position sensitive (Pixel) detector)
2D-ACAR study of electron moment distributions in solids

Defect-free GaAs

Quartz SiO₂

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.

Tanigawa et al., 1995

M. Biasini (1995)
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

\[ C_{1V}(T) = C_0 \exp\left(-\frac{H_F}{k_B T}\right) \]

\[ H_F = \text{formation enthalpy of one vacancy} \]

\[ C_{1V}(T_m) \approx 10^{-4\ldots-3} \text{/atom} \]

\( H_F = 4.0(3) \text{ eV} \)

\text{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}

\text{t to trapping model}
Coincidence Doppler Broadening Spectroscopy

Background Reduction
higher momentum resolution

\[ E_{\gamma_1} + E_{\gamma_2} = 2m_0 c^2 = 1022 \, \text{keV} \]
Positron-Emission-Tomography (PET)

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

18F- 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

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Half life</th>
<th>Nuclear reaction</th>
</tr>
</thead>
<tbody>
<tr>
<td>Oxygen-15</td>
<td>2.073 min</td>
<td>$^{14}\text{N}(d,n)^{15}\text{O}$</td>
</tr>
<tr>
<td>Nitrogen-13</td>
<td>9.95 min</td>
<td>$^{16}\text{O}(p,a)^{13}\text{N}$</td>
</tr>
<tr>
<td>Carbon-11</td>
<td>20.3 min</td>
<td>$^{14}\text{N}(p,a)^{11}\text{C}$</td>
</tr>
<tr>
<td>Fluorine-18</td>
<td>109.7 min</td>
<td>$^{18}\text{O}(p,n)^{18}\text{F}$</td>
</tr>
</tbody>
</table>

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 block

Scintillator Crystal

Photo Multiplier

Detector module

Detector Rings
50-80 cm

15-20 cm

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“: Bi4Ge3O12
„LYSO“: LuYSiO5:Ce3+
„LSO“, Lu2SiO5:Ce3+

TOTAL: up to 10.000 scintillators, 1000 photomultipliers

Costs (equipment, operation, personnel, etc): ~ 1000 US $/scan
Break even: ~ 60 scans/month