Positron-Annihilation Lifetime Spectroscopy using Electron Bremsstrahlung
Outline

• Motivation

• Accelerator-based positron production and annihilation studies at a superconducting electron LINAC: What marks the difference to reactors and radio-isotope sources?

• Applying pulsed beams: positron annihilation lifetime spectroscopy at thin films, bulk materials, and fluids

• Development of a pixelated detection system for position-sensitive positron annihilation lifetime measurements and experiments with structured targets and tomographic image reconstruction
Isotopes, reactors, accelerators

Production of positrons in weak ($W^+$) or electromagnetic interactions ($\gamma$)

Free proton decay is forbidden by energy conservation
$\rightarrow$ we need the proton inside a nucleus where it undergoes $\beta^+$-decay
Isotopes, reactors, accelerators

Production of positrons in weak interactions (mediated by W’s)

\[ ^{27}\text{Al}(p,n)^{27}\text{Si}(\beta^+\nu_e, 4.2 \text{ s})^{27}\text{Al} \]

neutron

(u d d)

\[ \nu_e \]

(e+)

proton

(u d u)

Sumitomi Heavy Industries Cyclotron
18 MeV protons, 50 \( \mu \)A beam current
Isotopes, reactors, accelerators

Production of positrons through electromagnetic interactions (photons)

Use intense source of photons for pair production

→ Capture-neutron gamma-rays from reactor $^{113}\text{Cd}(n,\gamma)^{114}\text{Cd}$

→ Bremsstrahlung from electron accelerators

FRMII Munich

AIST, Tsukuba, Japan

ELBE, Dresden
Positrons from accelerators

Accelerators can produce intense and pulsed slow positron beams. LINear ACcelerators are favored due to their high beam power and time structure.

A) normal conducting LINAC (AIST)
- $E \sim 50$ MeV
- $I_{\text{peak}} \sim 100$ mA
- $t_{\text{bunch}} \sim 1$ µs
- $f_{\text{rep}} \sim 100$ Hz
- Beam power $500$ W

B) superconducting LINAC (HZDR)
- $E \sim 50$ MeV
- $I_{\text{average}} \sim 1$ mA
- $f_{\text{rep}} \sim 10$ MHz
- Beam power $50$ kW

Sophisticated converter designs and heavy shielding needed

Stack of 50 100 µm thick W foils

EPOS water-cooled converter
Positrons from accelerators

SC-LINAC in CW mode

EPOS facility

NC-LINAC in bunched mode

Positrons from accelerators

- **Converter**
- **Moderator**
- **Linear Storage**
- **Chopper**
- **Sample**

**NC-LINAC in bunched mode**
- **Subh. Buncher**
- **Buncher**
- **Sample**

**SC-LINAC in CW mode**
- **Converter**
- **Moderator**
- **Magnetic Transport**
- **Chopper**
- **Sample**

**EPOS facility**
- **Buncher**
- **Sample**

- **38 ns / 26 MHz**
- **3 ms**
- **5 ns**
- **2 ns**
- **250 ps**

- **~ 2.8 GHz / 1 µs / 100 Hz / 10 µA**

**Positron and Positronium Chemistry, Goa 2014**
Andreas Wagner | Institute of Radiation Physics | www.hzdr.de

- **Converter**
- **Moderator**
- **E-**
- **E+**
- **1 µs**
- **10 eV**
- **3 ms**
- **5 ns**
- **10 ps**
- **2 keV**
- **10 eV**
- **3 ms**
- **5 ns**
- **10 ps**
- **2 keV**
Positrons from accelerators

- 1.6 mA, 40 MeV (64 kW) CW electron accelerator
- Coherent IR-radiation: 3 – 230 µm
- THz radiation: 100 µm – 3 mm
- Neutron time of flight: $E_n$ 0 – 10 MeV

- Bremsstrahlung: 16 MeV
- Gamma-induced Positrons
- Mono-energetic positrons: 0.2 – 20 keV

- Positrons from accelerators
- Mono-energetic positrons: 0.2 – 30 keV from $^{22}$Na

- Pulsed, mono-energetic positrons: 0.2 – 20 keV

- Electrons: 34 MeV radiation biology detector tests
What about bulk materials, fluids, gases …?

38 ns / 26 MHz

SC-LINAC in CW mode

Converter

Moderator

Magnetic transport

Chopper

Buncher

Sample

E-

10 ps

E+

e-

10 ps

Radiator

Gamma-induced Positron annihilation Spectroscopy (GiPS)

Sample

E+

10 ps

E-

10 ps
Positron production using electron-bremsstrahlung

\[ E_e = 16 \text{ MeV} \]
\[ I_e = 900 \mu\text{A} \]
\[ f = 26 \text{ MHz} \]
\[ \sigma_t < 10 \text{ ps} \]

Annihilation Lifetime Spectroscopy (Coincidence) Doppler Broadening Age-momentum Correlation

studies done so far:
- water, glycerol from 10°C to 100°C
- animal tissue
- metals and alloys
- neutron-activated reactor materials
Positrons: backgnd for nuclear physics exp’ts

Hard bremsstrahlung produces a huge amount of positrons via pair production inside the target material. High-energy photons act as a **volume source of positrons throughout the entire volume**.
Gamma-induced Positron Spectroscopy

Conventional LINAC mode
- Pulsed RF, highest energy
- Typically pile-up problems

SC-LINAC in CW mode
- Highest average power – high yield and low pile-up

High resolution lifetime spectrum with signal to noise ratios of better than $10^5:1$ using gamma-gamma coincidence techniques for background reduction. Lifetime spectra are free from artefacts.

→ Long lifetimes reveal atomic defects caused by neutron-induced damage.
→ Can (and how) defects be removed by thermal annealing?
Reactors vessel steel becomes brittle due to neutron-induced defects like open-volume defects. The atomic defects act as seeds for cracks.

- Preferential formation of double vacancies
- Thermal annealing (290°C) not sufficient to remove defects!
Physics with GiPS: Kapton

Annihilation lifetime in Kapton has been under debate for quite some time. Here, we try to get a measurement without source correction.

→ consistent single positron lifetime of \((381 \pm 1)\) ps

two components show larger \(\chi^2\)

applied cuts on Germanium and BaF\(_2\) detector energy signal reduce background from interactions outside the sample
Physics with GiPS: Fluids

Conventional lifetime measurements:
→ dissolve $^{22}\text{Na}$ and dispose it afterwards

Positrons from bremsstrahlung
→ homogeneously distributed, sharp time stamp

Target is temperature-stabilized, continuously circulated, degassed, dry-nitrogen flushed.

Positron Physics
Ortho-Positronium (o-Ps) in a fluid forms a bubble given by its zero-point energy and the surface tension.

We know estimate the change of the o-Ps pick-off annihilation lifetime with temperature in a bubble created by the o-Ps itself.

Physics with GiPS: Fluids

\[-\frac{\hbar^2}{2m_{Ps}} \Delta \Psi + U(r) \Psi = E \Psi\]  
stationary Schrödinger eqn.

\[\Psi = R(r) \cdot \Theta(\theta) \cdot \Phi(\phi)\]

\[R(r) = R_0 j_1(kr)\]

\[j_0(kr) = \frac{\sin kr}{kr}\]

\[E_0 = \frac{\hbar^2}{8m_{Ps} r_0^2} = \frac{\pi^2 \hbar^2}{4m_e r_0^2}\]

Ansatz: spherical Bessel fct.

1st non-trivial solution

zero-point energy

\[E_{surf} = 4\pi r_0^2 \sigma\]

\[\frac{\partial}{\partial r_0} \left( E_0 + E_{surf} \right) = 0\]

\[-\frac{\pi^2 \hbar^2}{2m_e r_0^3} + 8\pi r_0 \sigma = 0\]

\[r_0 = 4 \sqrt{\frac{\pi \hbar^2}{16m_e \sigma}} = 4.3 \text{ Å}\]

\[a_0 = \frac{4\pi e_0 \hbar^2}{m_{\mu} e^2} = \frac{\hbar c}{\alpha m_{\mu} c^2} = 1.06 \text{ Å}\]
Experiments with water are in variance with a simple bubble-type model. Extension: chemical reactions between radiolysis products of the slowing-down of the positron \( \rightarrow \) Ps chemistry.

- Radicals are positron scavengers which reduce annihilation lifetimes.
- Extended bubble model including chemistry [S.V. Stepanov et al., Mat. Sci. Forum 607] describes data well.
- Relevance for PET diagnostics since \( 2\gamma / 3\gamma \) ratio is affected.
- Chemistry of radiolysis directly accessible since the probe creates the ionization itself.

"preliminary"
Towards imaging of defects

Material failures impose a significant threat to the integrity and the safety of technical systems. A thorough understanding of the microscopic origin and the development of defects requires advanced methods.

... and the quests of today
Motivation

Establish a **non-destructive** and **non-intrusive** method which allows for **spatially resolved** positron-lifetime spectroscopy. Reconstruct PET-like images plus positron annihilation lifetime.

Possible Applications (list not complete):

- **Porosimetry**
- **Medicine** in-beam positron lifetime spectroscopy during hard x-ray tumor therapy
- **Engineering** pre-failure diagnostics of micro fractures fuel rod inspection

APS Physics & Society Newsletter
2011. R. Hargraves, R. Moir
Prerequisites

- Intense source of positrons with deep penetration (cm)
- Accurate time-stamping of positron creation (<10 ps)
- Position-sensitive positron detectors (mm)
- Time-resolution for lifetime spectroscopy (~100 ps)
- Efficient data acquisition
- 3-D image reconstruction

Gamma-induced Positron Spectroscopy
Towards 2/3-D positron lifetime tomography

- Two position-sensitive photon detectors with 169 elements each

**LSO-based commercial 13x13 PET pixel detector**

- Two position-sensitive photon detectors with 169 elements each
- Each crystal array read out using 4 PMT
- Summed PMT signal -> gamma energy
- Correlation of individual PMT signals -> position
- Positron annihilation time given by sum over all 8 PMT involved

Lutetium oxyorthosilicate

Lu$_2$SiO$_5$:Ce

©Siemens
courtesy: university hospital Dresden
Electronics (VME)

- Block detector
- Transimpedance amplifier
- NIM
- VME
  - CFD LE
  - CAEN V1495 FPGA
  - CAEN V1290 TDC
  - CAEN V965 QDC
  - CES RIO4 ROC

Multi-hit and multi-event buffered readout in VME block mode and readout with 10 µs dead time for 36 channels (QDC & TDC) per event. Throughput is about 10 MB/s sustained. Data acquisition and analysis framework using Multiple-Branch System MBS by Helmholtz-Center for Heavy Ion Research (GSI).
Calibrations

\[ E = E_1 + E_2 + E_3 + E_4 \]

\[ t = \frac{1}{4}(t_1+t_2+t_3+t_4) - t_{\text{accel}} \]

\[ x = \frac{(E_1+E_2)-(E_3+E_4)}{E_1+E_2+E_3+E_4} ; \quad y= \frac{(E_1+E_3)-(E_2+E_4)}{E_1+E_2+E_3+E_4} \]

Calibration done using 7 cm x 7 cm aqueous $^{18}$F source w/ 200 MBq ($T_{1/2} \approx 2$ h) produced in-house.
Sample cases

Proof of principle, first test
Simple 2D target
→ proof of principle
→ simple back-projection method

3D target
→ Reconstruction of data as a function of life time

Real world sample (cutout from 91.4 T magnet coil)
→ What we can learn from our method
Sample selected to give balanced positron yield.
Lifetime-gated 2D reconstructed image by back-projection.
3D tomography applied for the **first time** using bulk volume positron production. Target is rotated in 2 deg. steps and the image is reconstructed using a cubical \((30 \text{ mm})^3\) voxel space and back-projection algorithm.
Maximum Likelihood Expectation Maximization

Iterative method for image reconstruction based on a algorithm developed in PET

Solves the inversion problem numerically where one has a system matrix $M$, an a-priori unknown source
distribution $s$ and a measured distribution $r$.

$$\hat{M} \cdot s = r$$

The system matrix has a size of $13^2 \times 13^2 \times 180 \times 30^3 = 138 \times 10^9$.

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**step 1**

**all**

**prompt**

**long**
MLEM

2\textsuperscript{nd} Iteration

After the first iteration, the data was corrected using MLEM. Here are the results for the different gates:

- **Step 2**
  - **All**
  - **Prompt**
  - **Long**

Each image shows the distribution of counts in different regions of the sample, with the x-length and z-length in pixels. The long gate shows a more uniform distribution, while the prompt gate highlights specific areas with higher counts.
MLEM

5th Iteration

Copper
Aluminium
Steel

step 5

all
prompt
long
MLEM

step 10

10th Iteration

Copper
Aluminium
Steel

long gate
short gate

ALL
prompt
long
MLEM

Copper
Aluminium
Steel

20th Iteration

step 20

all

prompt

long

long gate

short gate

counts

positron lifetime / ns

0 5 10 15 20 25
0 5 10 15 20 25
0 5 10 15 20 25

x-length/px

z-length/px

x-length/px

z-length/px

x-length/px

z-length/px
Gating on positron lifetimes with 225 ps timing resolution.

Now the Al is clearly discriminated against the surrounding Teflon.
Cut through the record coil which reached 91.4 T peak field. Coil is fed by the world’s largest capacitor bank w/ 50 MJ stored energy.
Tomography: B-field coil

48 h measurement time, 316 GB, 1.6 G events
324 M filtered coincidences
Lifetime-sensitive analysis: B-field coil
Now, we select specific voxels and determine the annihilation lifetimes for spatially separated regions. Since the voxel is identified as an ensemble over all possible lines-of-response between two detector crystals, the lifetime distribution is a convolution as well. Some real physics questions needed...
Extensions

Digital Silicon Photomultiplier (dSiPM) Module
DPC3200-22-44
(819200 pixel each)

Digitally counting the number and the time of arrival of scintillation photons (here LYSO)

#Courtesy: Philips Digital Photon Counting
Extensions

digital Silicon Photomultiplier (dSiPM)

Employ the scaled accelerator radio frequency (13 MHz) via a phase-locked loop (PLL) as dSiPM system clock.

- Intrinsic synchronization for optimal timing resolution.
- 170 ps FWHM seem possible

Scintillation materials

Collaborative effort within gamma-ray imaging group at particle-therapy center Oncoray.
(Courtesy: J. Petzoldt, K. Römer, G. Pausch, et al.)
Summary

Summary:
- Accelerator-driven positron production
- Annihilation lifetime spectroscopy for fluids, reactor materials...
- First results for 3D tomography
The team

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and all the collaborators
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- S.V. Stepanov, D.S. Zvezhinskiy (ITEP, MEPhI)

Apply for beam time: deadlines 1st weeks in May and November

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Thank you for attention