Technical Status Update on PA Lifetime Spectroscopy Experiments and Results

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Technical Status Update on PA Lifetime Spectroscopy
Experiments and Results

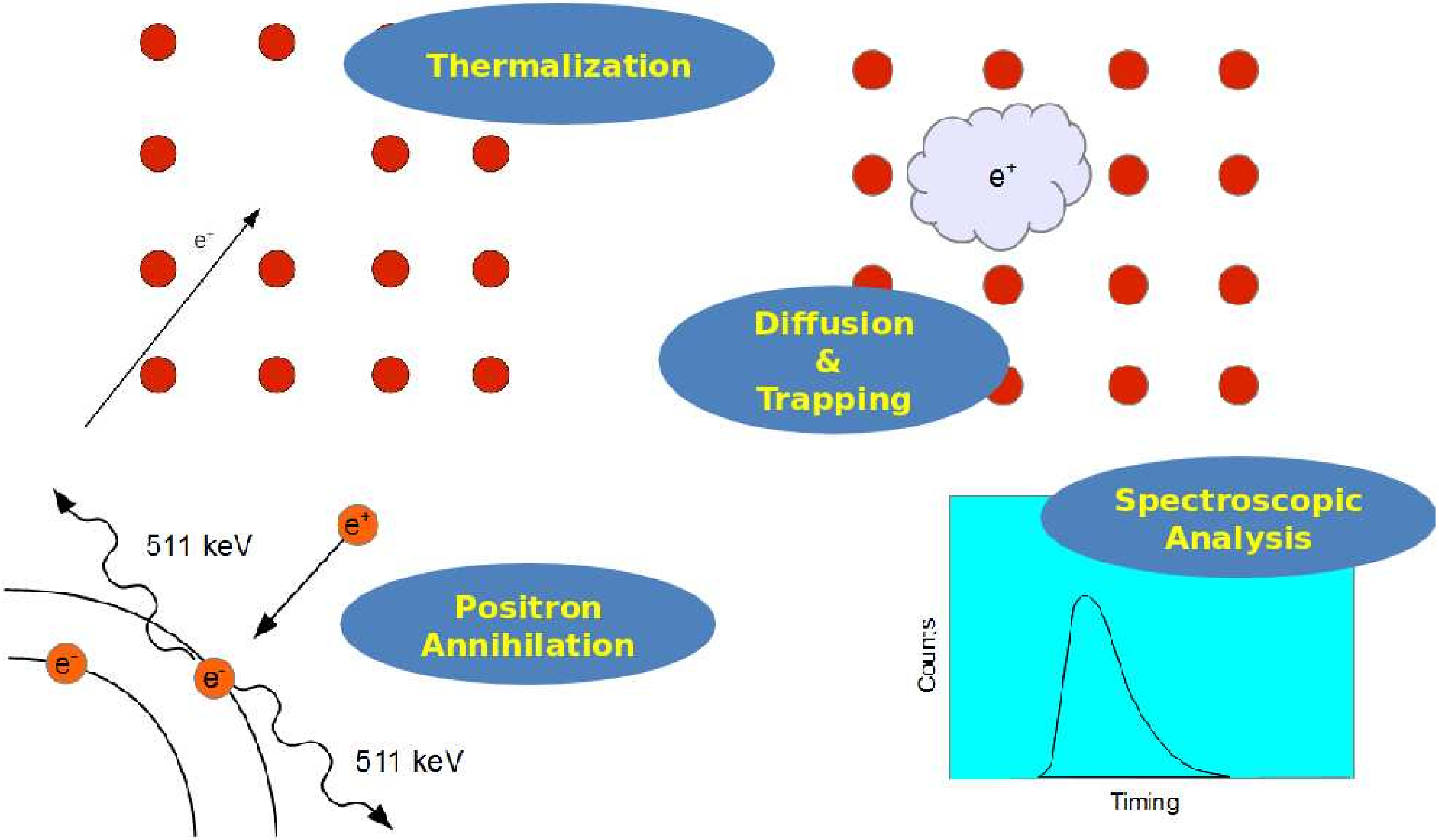
Outline

- Introduction
- Theory
- Methods and Materials
- Spectrometer Optimization/Benchmarking
- Analysis and Results
- Future Work

Goals of Study

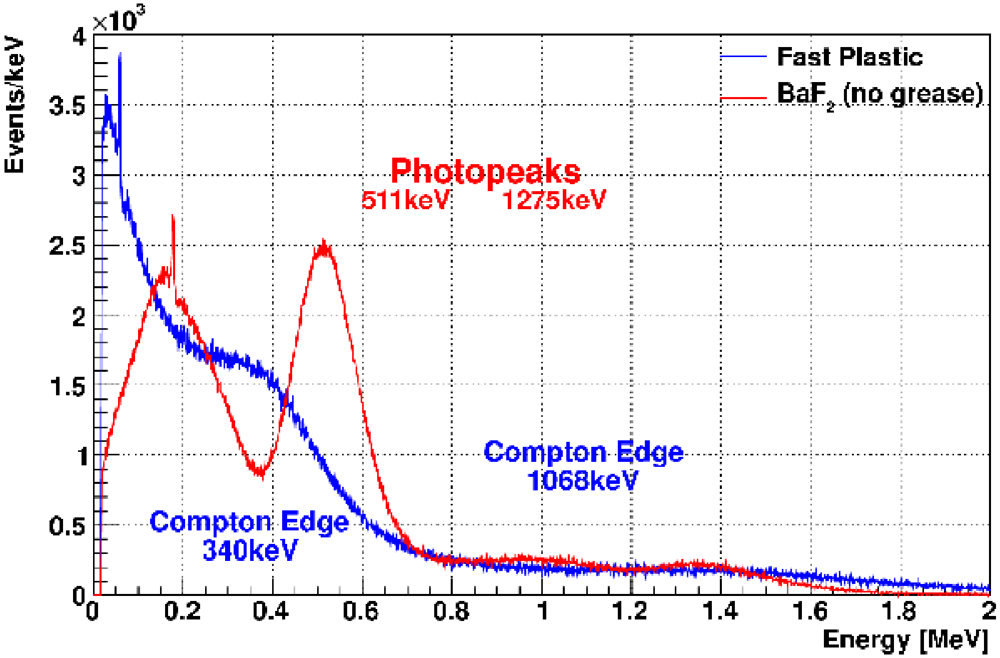
- Construction and optimization of a Positron Annihilation Lifetime Spectrometer
  - Capable of distinguishing between defect and defect-free material samples
- Source-based experiments: Benchmarking to literature values
  - Annealed samples (bulk/non-defect)
  - Unannealed (defect)
- Accelerator-based experiments: Feasibility studies and reproducing source-based measurements

PALS Overview

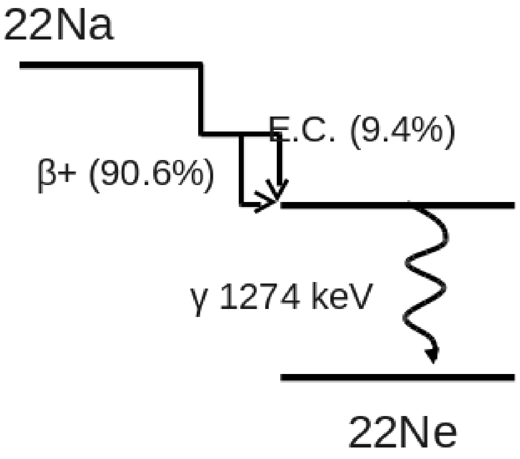


^{22}Na Positron Source

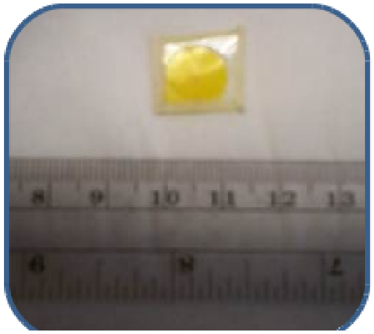
- 22Na is a well known positron source



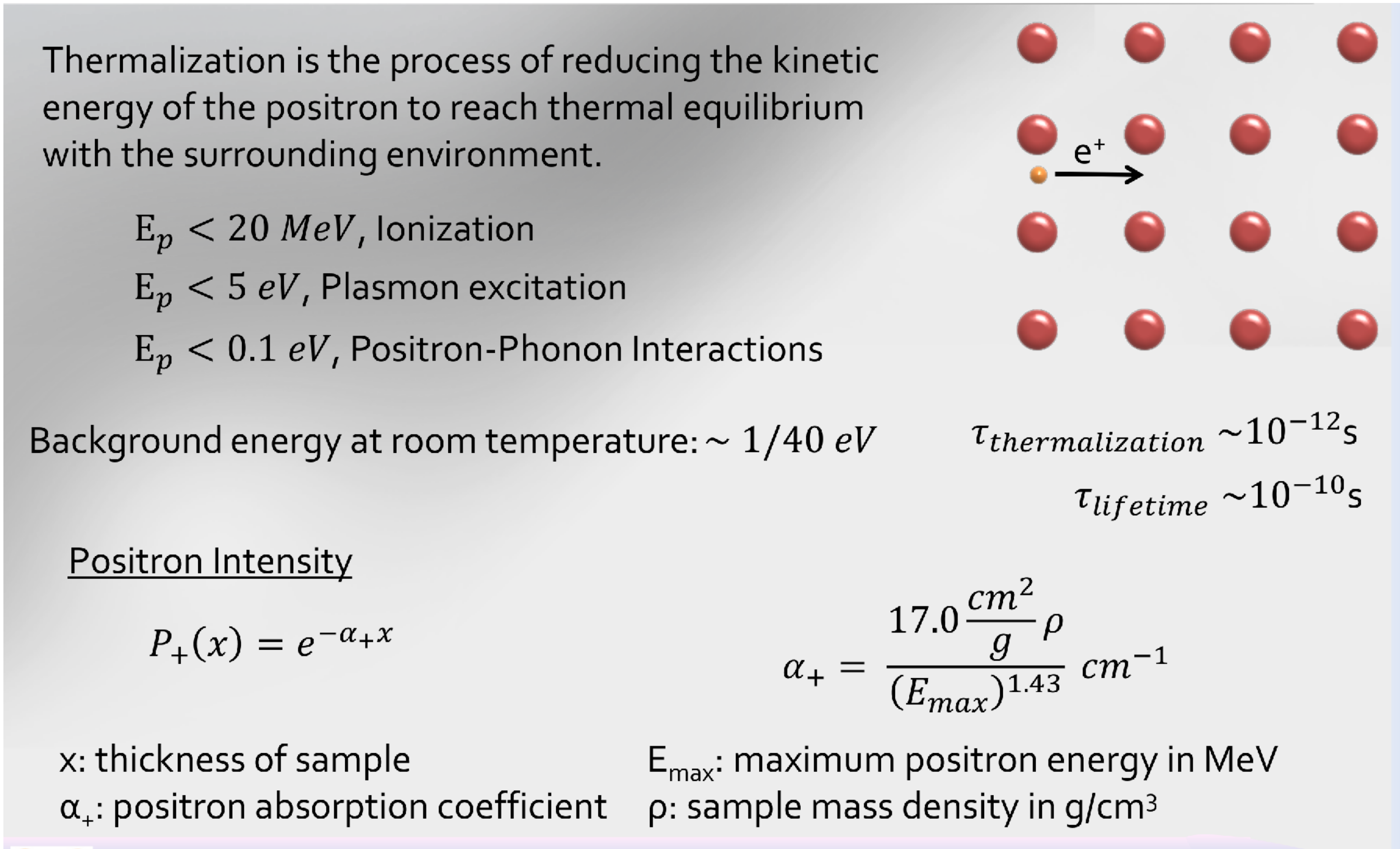
NaCl solution placed between two thin Kapton tape films



Source used:
50 µCi created in 2007



Positron Interactions: Thermalization



Thermalization is the process of reducing the kinetic energy of the positron to reach thermal equilibrium with the surrounding environment.

$\mathrm{E}_p < 20\ MeV$, Ionization
$\mathrm{E}_p < 5\ eV$, Plasmon excitation
$\mathrm{E}_p < 0.1\ eV$, Positron-Phonon Interactions

Background energy at room temperature: $\sim 1/40\ eV$

$\tau_{thermalization} \sim 10^{-12}\mathrm{s}$

$\tau_{lifetime} \sim 10^{-10}\mathrm{s}$

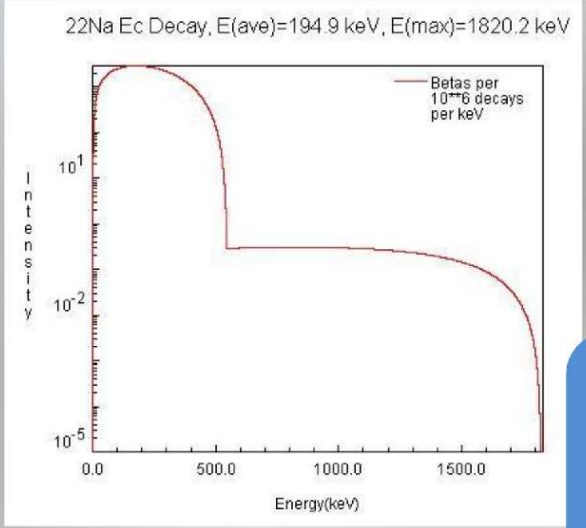
Positron Intensity

$$P_+(x) = e^{-\alpha_+ x}$$

$$\alpha_+ = \frac{17.0\,\frac{cm^2}{g}\,\rho}{(E_{max})^{1.43}}\ cm^{-1}$$

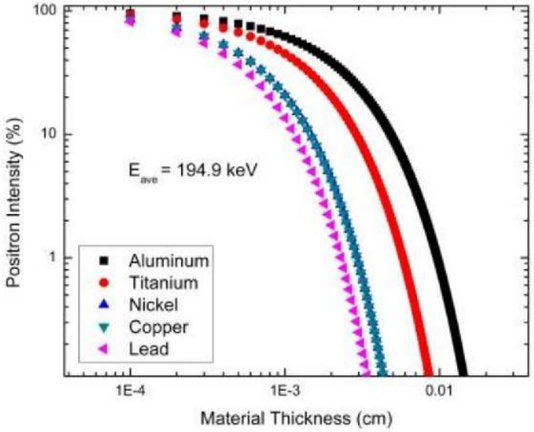
x: thickness of sample
α_+: positron absorption coefficient
E_{max}: maximum positron energy in MeV
ρ: sample mass density in g/cm^3

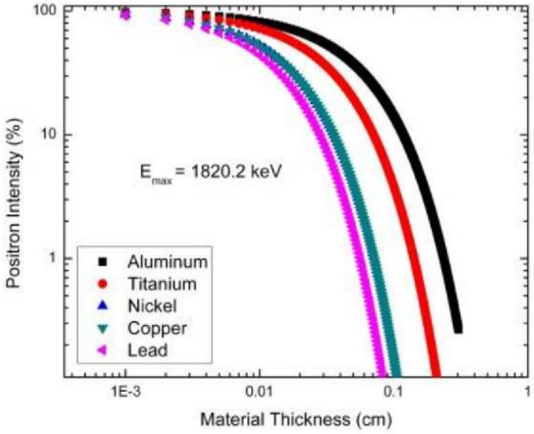
Positron Interactions: Penetration Depth



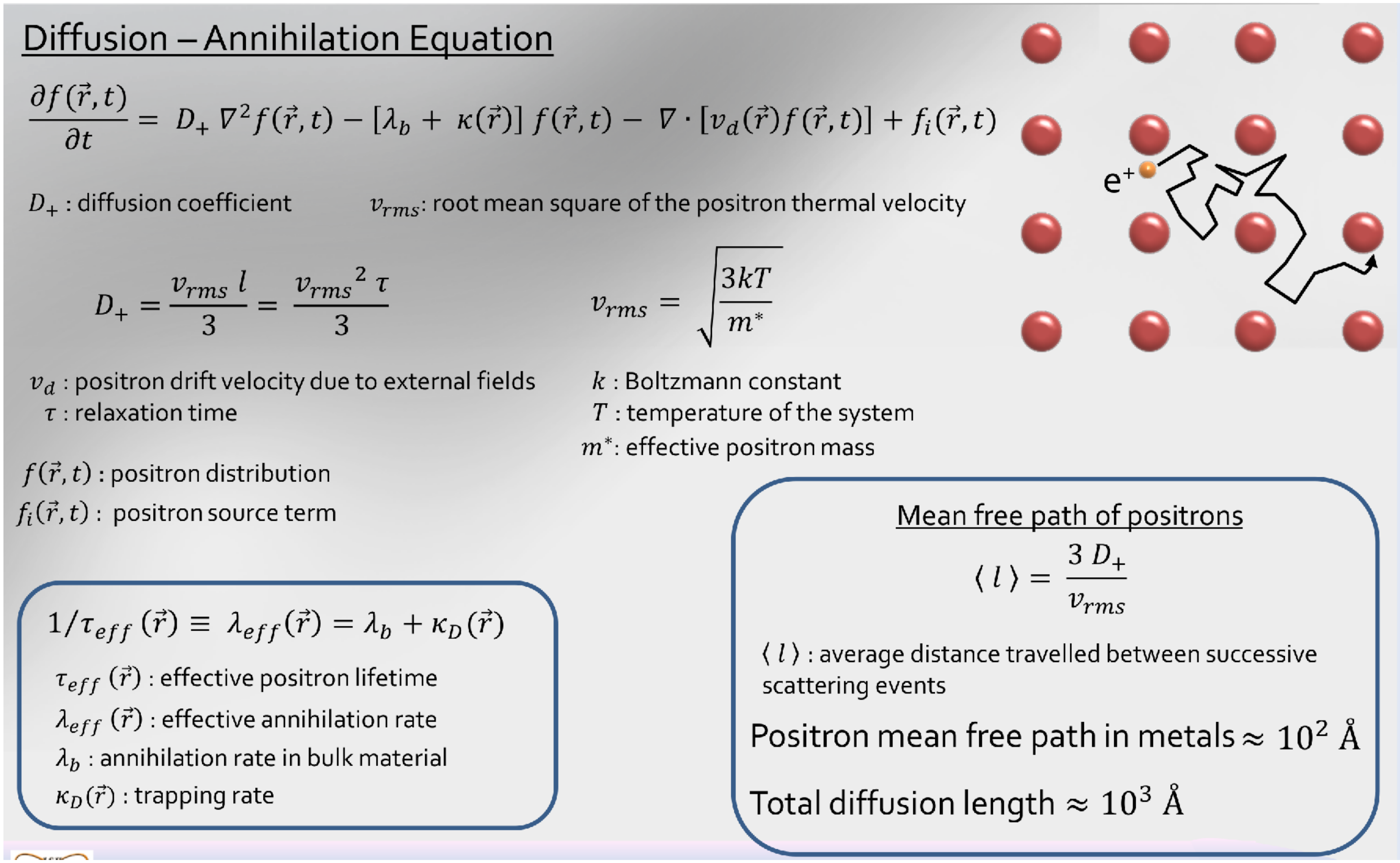
Source positrons only penetrate 50 μm into materials

Material	Density (g/cm³)
Aluminum	2.70
Nickel	8.908
Copper	8.94
Lead	11.34





Positron Interactions: Diffusion



Diffusion – Annihilation Equation

$$\frac{\partial f(\vec{r},t)}{\partial t} = D_+ \nabla^2 f(\vec{r},t) - [\lambda_b + \kappa(\vec{r})]\, f(\vec{r},t) - \nabla \cdot [v_d(\vec{r}) f(\vec{r},t)] + f_i(\vec{r},t)$$

D_+ : diffusion coefficient

v_{rms}: root mean square of the positron thermal velocity

$$D_+ = \frac{v_{rms}\, l}{3} = \frac{{v_{rms}}^2\, \tau}{3}$$

$$v_{rms} = \sqrt{\frac{3kT}{m^*}}$$

v_d : positron drift velocity due to external fields
τ : relaxation time

k : Boltzmann constant
T : temperature of the system
m^*: effective positron mass

$f(\vec{r},t)$: positron distribution
$f_i(\vec{r},t)$: positron source term

$$1/\tau_{eff}(\vec{r}) \equiv \lambda_{eff}(\vec{r}) = \lambda_b + \kappa_D(\vec{r})$$

$\tau_{eff}(\vec{r})$: effective positron lifetime
$\lambda_{eff}(\vec{r})$: effective annihilation rate
λ_b : annihilation rate in bulk material
$\kappa_D(\vec{r})$: trapping rate

Mean free path of positrons

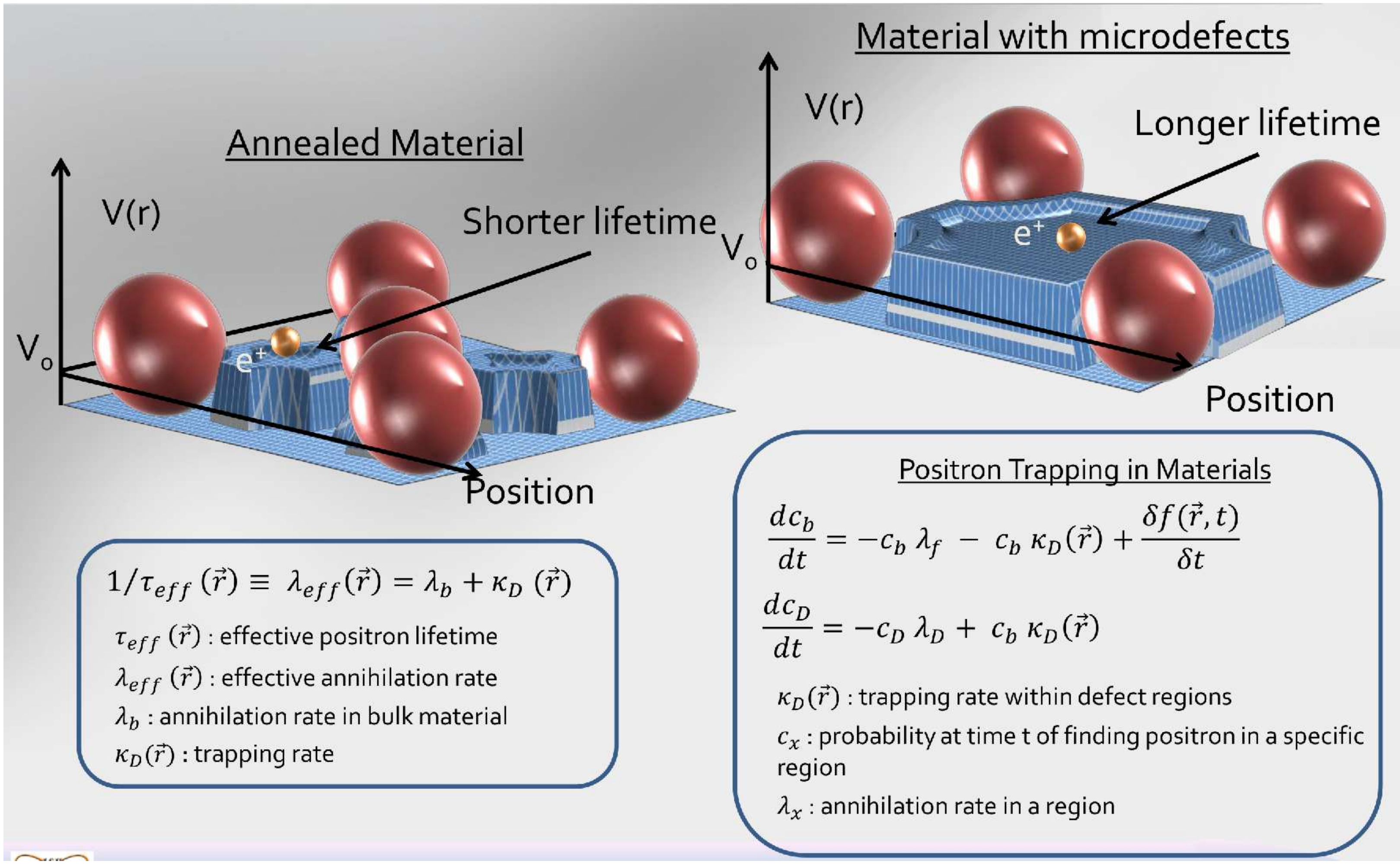
$$\langle\, l \,\rangle = \frac{3\, D_+}{v_{rms}}$$

$\langle\, l \,\rangle$: average distance travelled between successive scattering events

Positron mean free path in metals $\approx 10^2$ Å

Total diffusion length $\approx 10^3$ Å

Positron Interactions: Trapping



$$1/\tau_{eff}(\vec{r}) \equiv \lambda_{eff}(\vec{r}) = \lambda_b + \kappa_D(\vec{r})$$

$\tau_{eff}(\vec{r})$: effective positron lifetime

$\lambda_{eff}(\vec{r})$: effective annihilation rate

λ_b : annihilation rate in bulk material

$\kappa_D(\vec{r})$: trapping rate

Positron Trapping in Materials

$$\frac{dc_b}{dt} = -c_b\,\lambda_f - c_b\,\kappa_D(\vec{r}) + \frac{\delta f(\vec{r},t)}{\delta t}$$

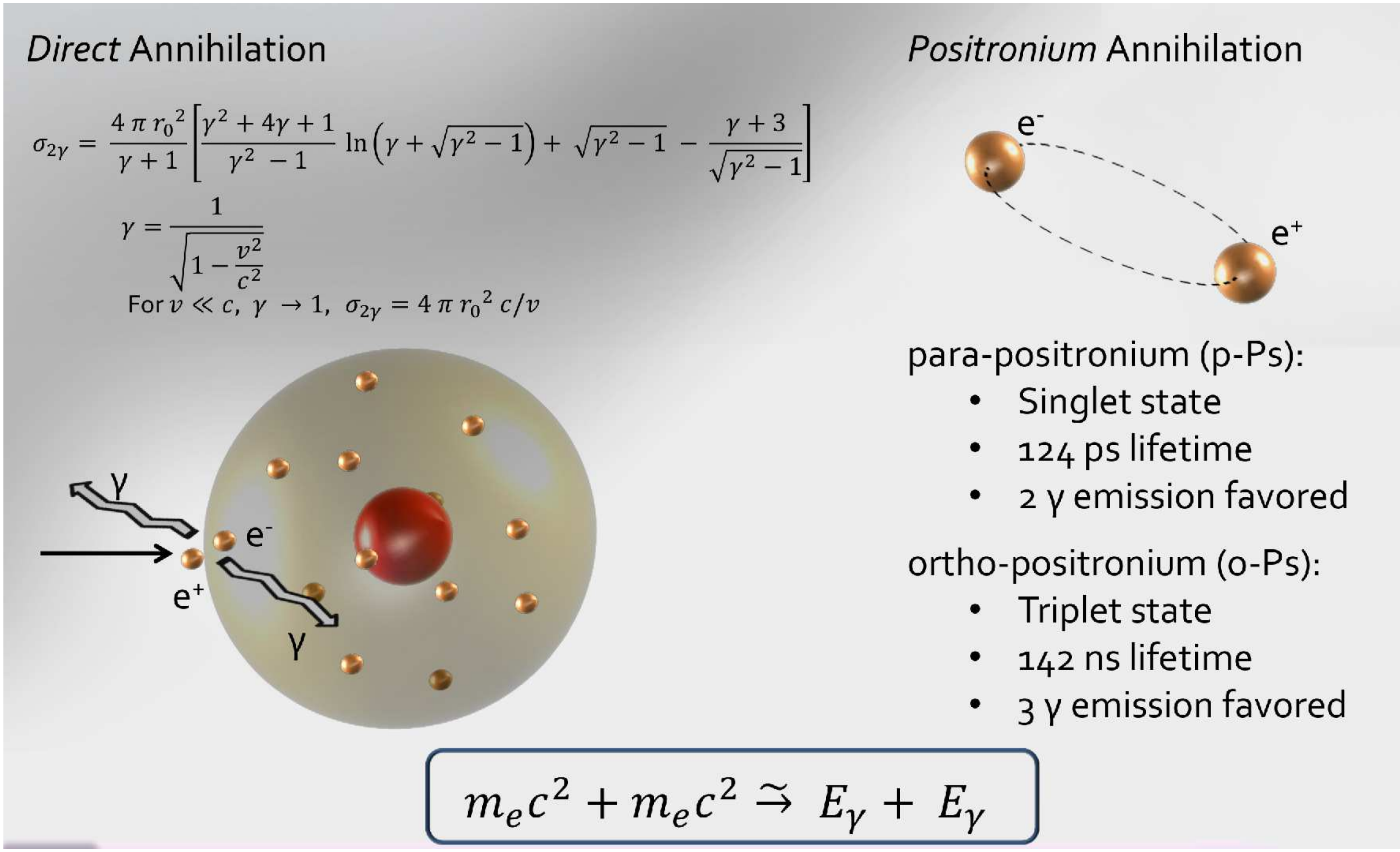
$$\frac{dc_D}{dt} = -c_D\,\lambda_D + c_b\,\kappa_D(\vec{r})$$

$\kappa_D(\vec{r})$: trapping rate within defect regions

c_x : probability at time t of finding positron in a specific region

λ_x : annihilation rate in a region

Positron Interactions: Annihilation



Direct Annihilation

$$\sigma_{2\gamma} = \frac{4\,\pi\,{r_0}^2}{\gamma+1}\left[\frac{\gamma^2+4\gamma+1}{\gamma^2\ -1}\ln\left(\gamma+\sqrt{\gamma^2-1}\right)+\sqrt{\gamma^2-1}-\frac{\gamma+3}{\sqrt{\gamma^2-1}}\right]$$

$$\gamma = \frac{1}{\sqrt{1-\frac{v^2}{c^2}}}$$

For $v \ll c,\ \gamma \rightarrow 1,\ \sigma_{2\gamma} = 4\,\pi\,{r_0}^2\,c/v$

Positronium Annihilation

para-positronium (p-Ps):

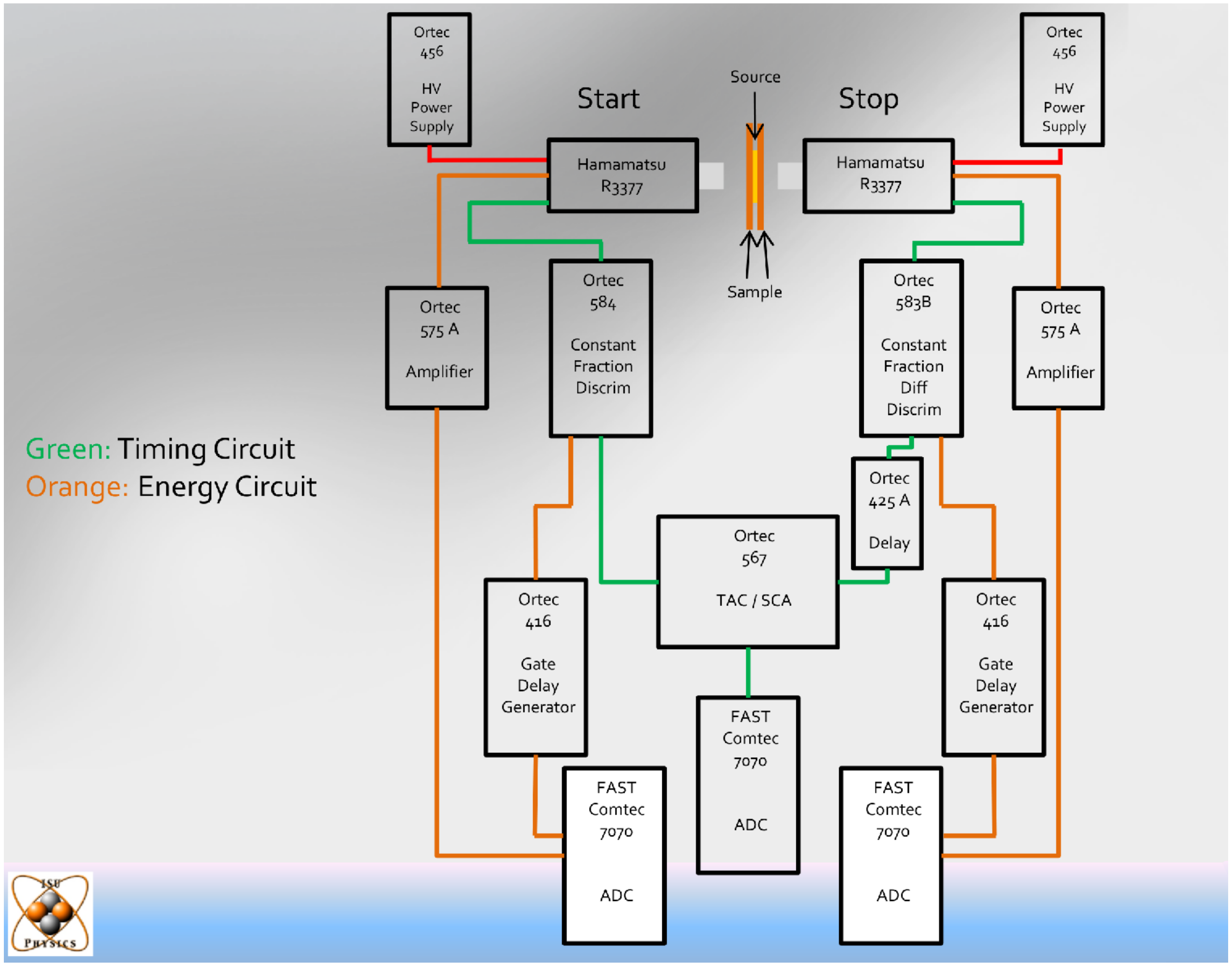
- Singlet state
- 124 ps lifetime
- 2 γ emission favored

ortho-positronium (o-Ps):

- Triplet state
- 142 ns lifetime
- 3 γ emission favored

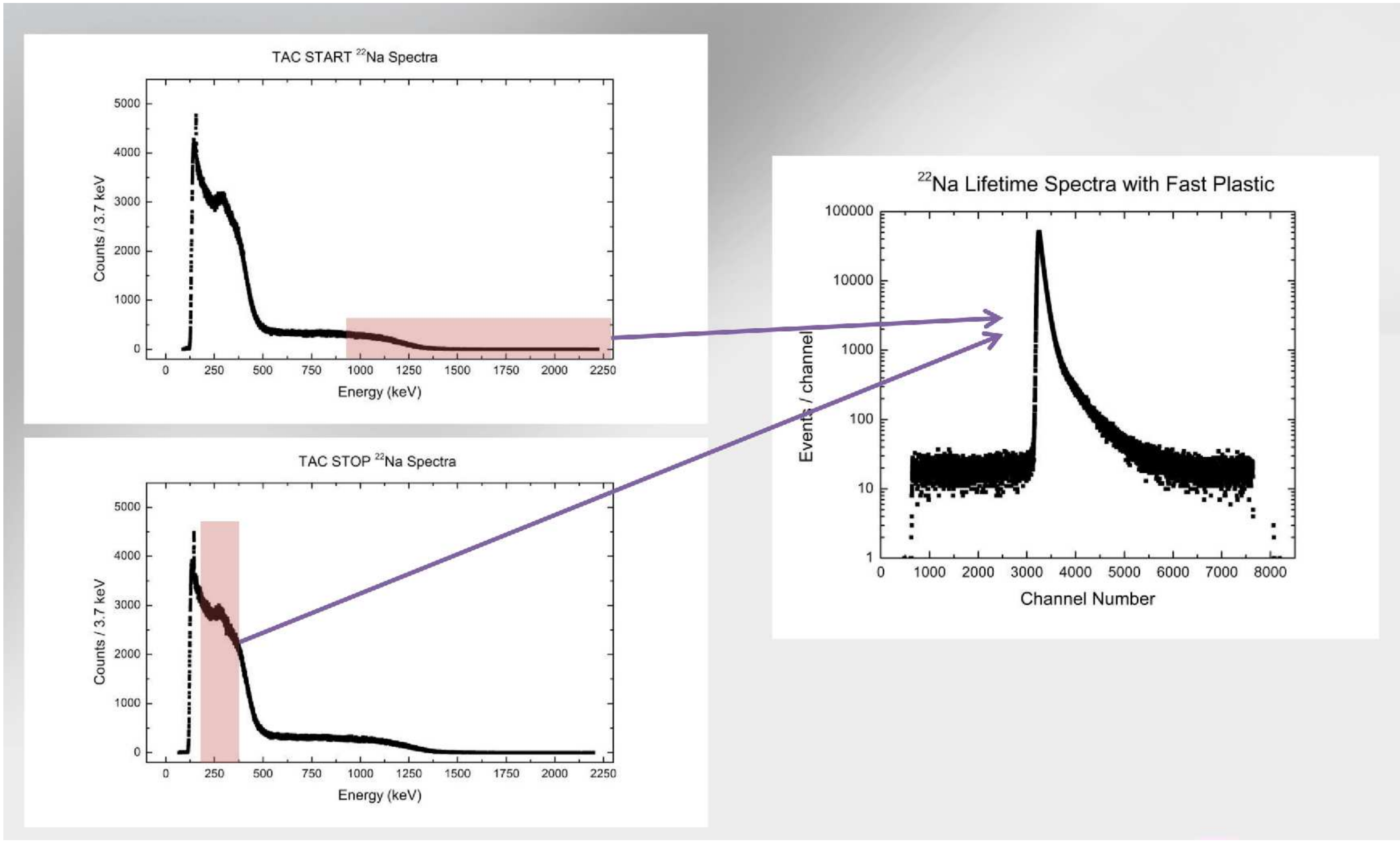
$$\boxed{m_e c^2 + m_e c^2 \overset{\sim}{\rightarrow} E_\gamma + E_\gamma}$$

PALS Spectrometer

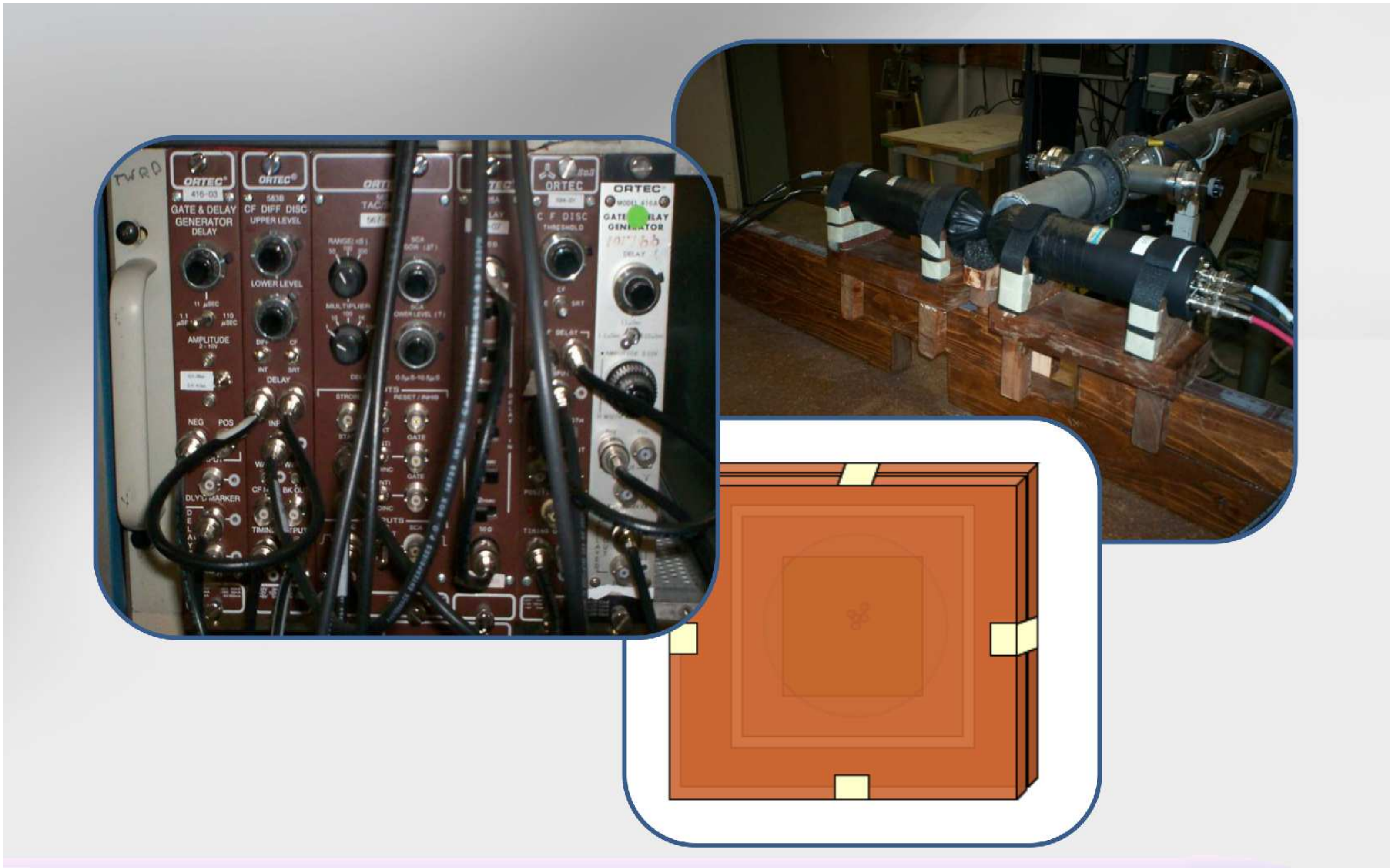


Positron Annihilation Lifetime Spectroscopy

Energy Discrimination & Lifetime Spectrum



Experimental Setup

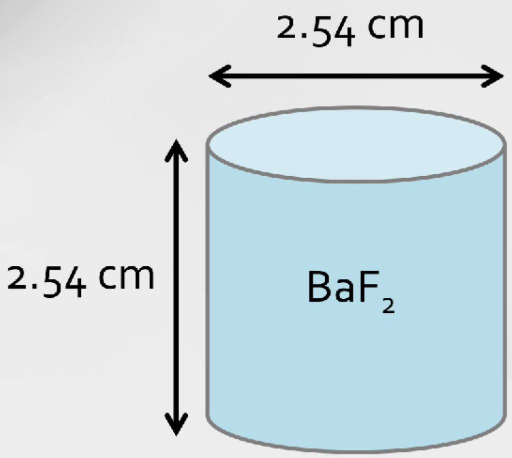


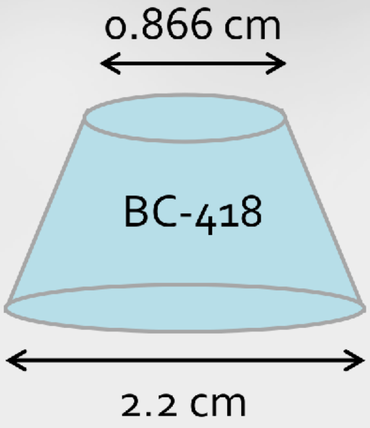
Positron Annihilation Lifetime Spectroscopy

Scintillator Properties

Barium Fluoride

- Has a fast and slow component
- Fast scintillation light is emitted in UV bands centered at 220 and 195 nm.
- Decay time of fast component varies between 0.6 – 0.8 ns
- Quartz window on PMT required to detect fast component
- Slow scintillation light is emitted in band centered at 310 nm.
- Decay time of slow component varies has an average value of 630 ns
- 15 % of produced photoelectrons are produced by the fast component
- 85% of produced photoelectrons are produced by the slow component
- Has index of refraction of 1.54 (220 nm) and 1.5 (310 nm)

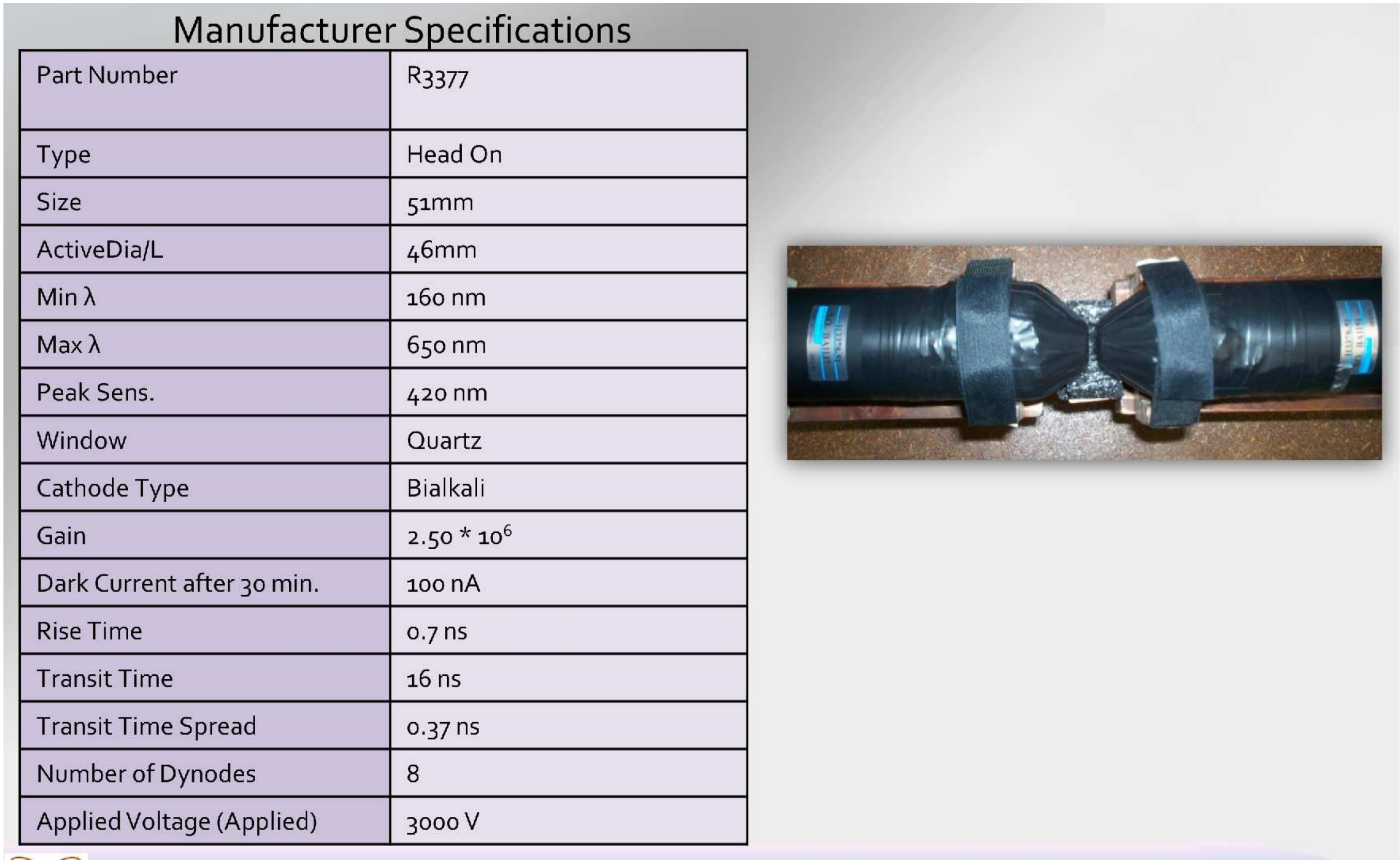




BC-418 Fast Plastic

- Light output, % Anthracene: 67
- Rise time of 0.5 ns
- Decay time of 1.4 ns
- Pulse Width, FWHM of 1.2 ns
- Scintillation light is emitted at max 391 nm
- Has index of refraction of 1.58

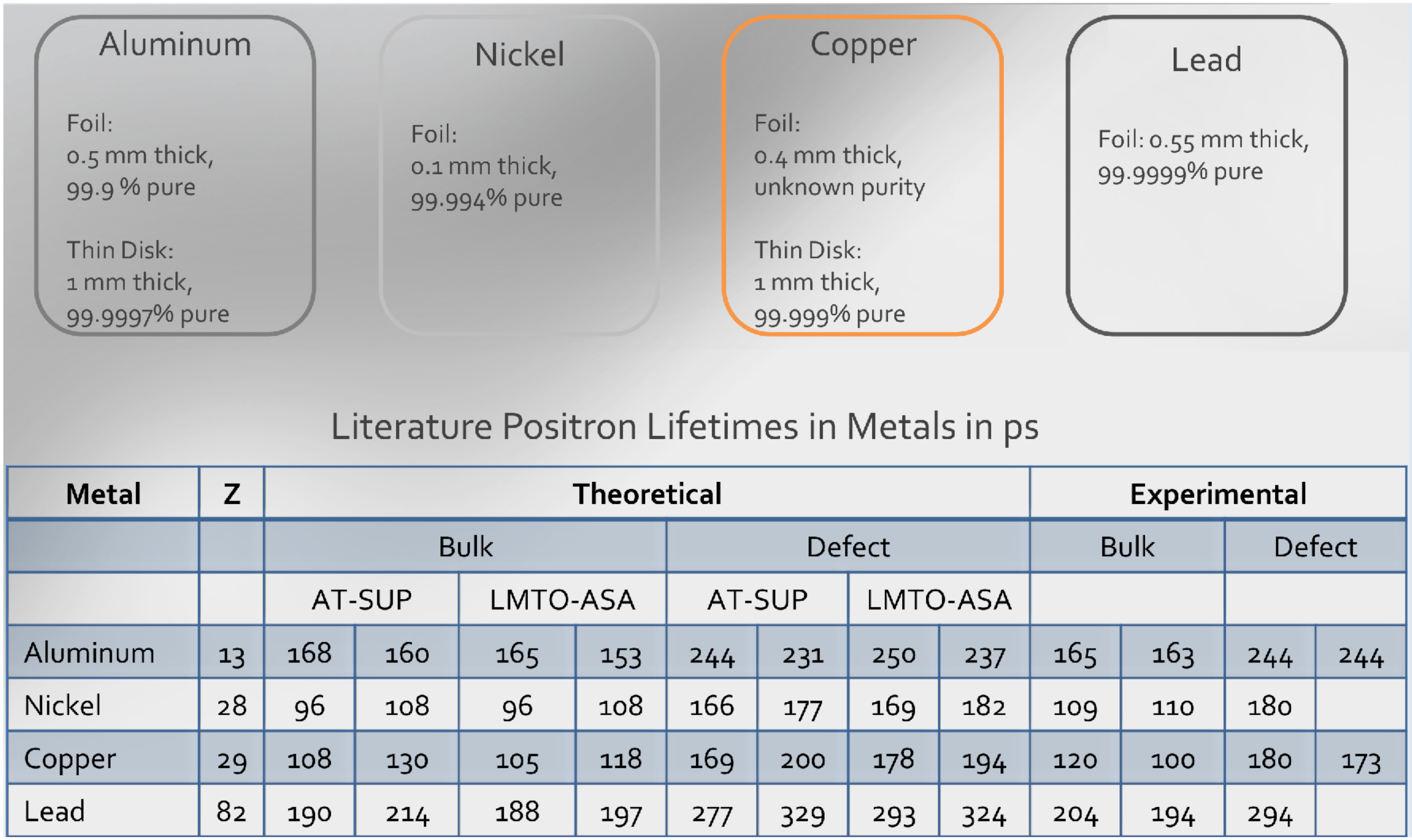
PMT Specifications (Hamamatsu R3377)



Manufacturer Specifications

Part Number	R3377
Type	Head On
Size	51mm
ActiveDia/L	46mm
Min λ	160 nm
Max λ	650 nm
Peak Sens.	420 nm
Window	Quartz
Cathode Type	Bialkali
Gain	$2.50 * 10^6$
Dark Current after 30 min.	100 nA
Rise Time	0.7 ns
Transit Time	16 ns
Transit Time Spread	0.37 ns
Number of Dynodes	8
Applied Voltage (Applied)	3000 V

Physical Properties of Test Samples



Aluminum

Foil:
0.5 mm thick,
99.9 % pure

Thin Disk:
1 mm thick,
99.9997% pure

Nickel

Foil:
0.1 mm thick,
99.994% pure

Copper

Foil:
0.4 mm thick,
unknown purity

Thin Disk:
1 mm thick,
99.999% pure

Lead

Foil: 0.55 mm thick,
99.9999% pure

Literature Positron Lifetimes in Metals in ps

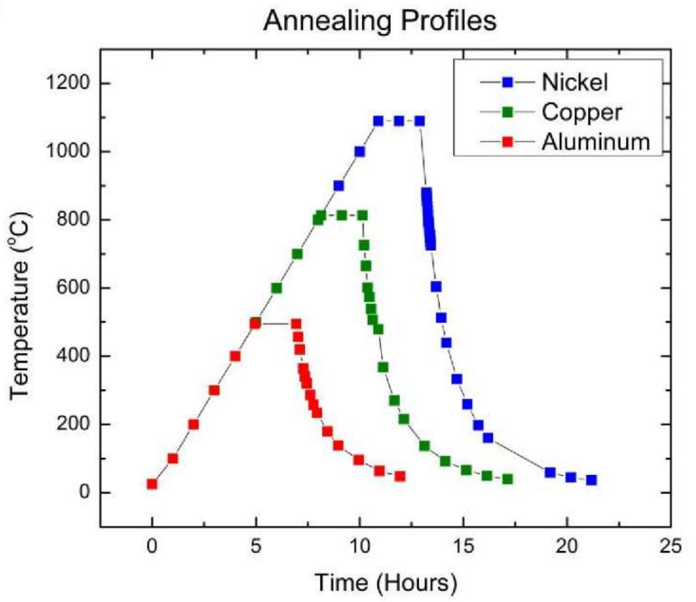
Metal	**Z**	**Theoretical**								**Experimental**			
		Bulk				Defect				Bulk		Defect	
		AT-SUP		LMTO-ASA		AT-SUP		LMTO-ASA					
Aluminum	13	168	160	165	153	244	231	250	237	165	163	244	244
Nickel	28	96	108	96	108	166	177	169	182	109	110	180	
Copper	29	108	130	105	118	169	200	178	194	120	100	180	173
Lead	82	190	214	188	197	277	329	293	324	204	194	294	

Process of Annealing Samples

Material	Melting Point (°C)	Anneal Point (°C)
Aluminum	660	495
Nickel	1453	1090
Copper	1085	813

Annealing realigns atomic lattice to original configuration, thus removing any material defects

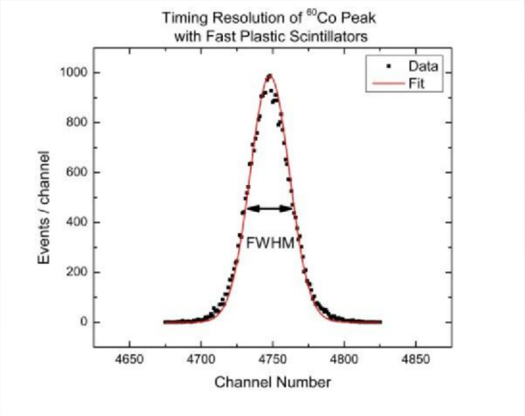
$$\frac{dT}{dt} = 100\ ^{\circ}\mathrm{C/hr}$$

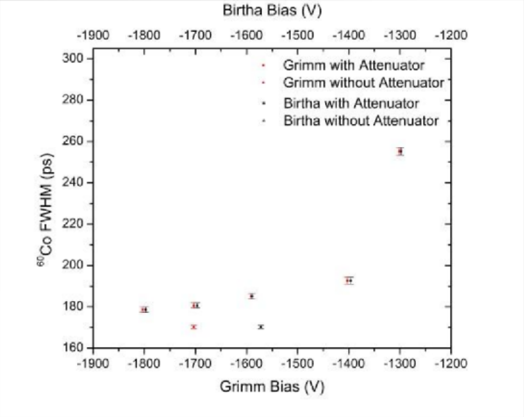


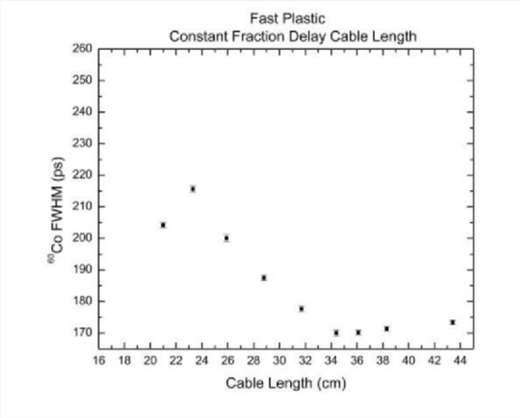
Timing Optimization (Fast Plastic)

- Energy calibration completed using Compton edges

$$E - E' = E - \frac{E}{1 + \frac{(1-\cos\theta)\,E}{m_e c^2}} = \frac{2\,E^2}{2\,E + m_e c^2}$$







Timing Resolution FWHM
170.17 ± 0.73 ps

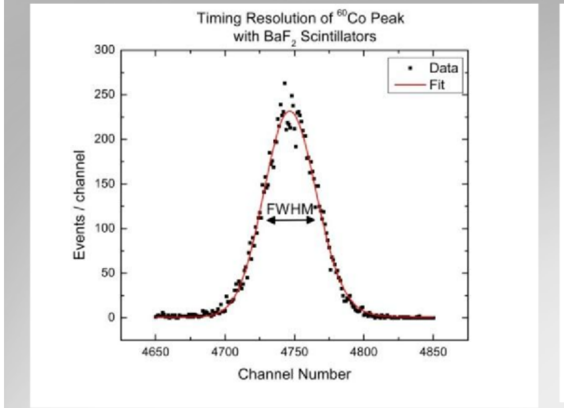
V_{START} = - 1703 V
V_{STOP} = - 1572 V

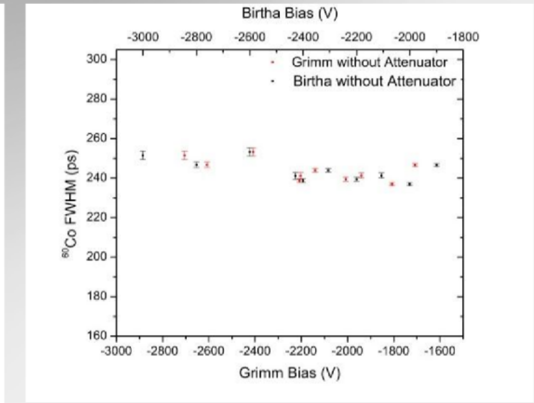
CF Delay Cable Length:
36.1 cm

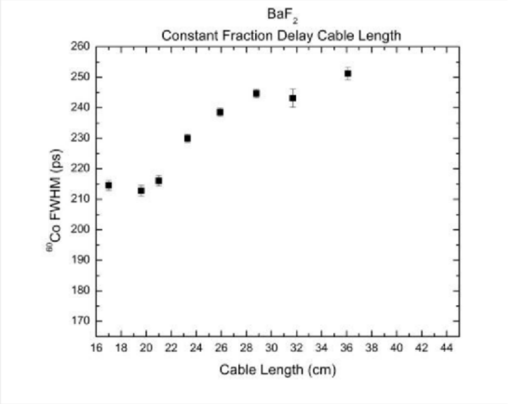
Timing Optimization (BaF_2)

- Energy calibration completed using Photopeaks

$$E = h\upsilon$$







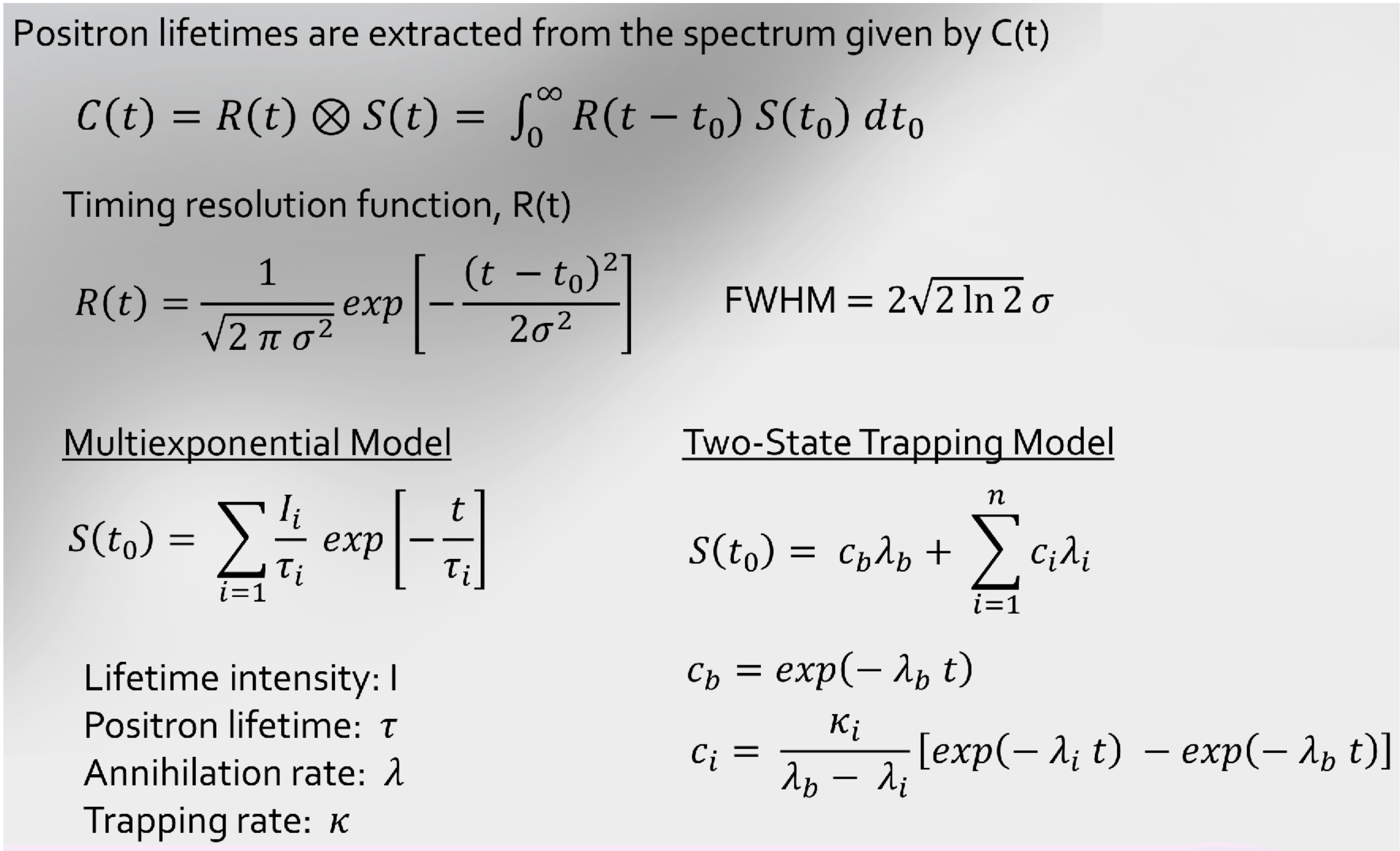
Timing Resolution FWHM
213.34 ± 1.79 ps

V_{START} = -2705 V
V_{STOP} = - 3001 V

CF Delay Cable Length:
19.6 cm

START ^{137}Cs Energy Resolution = 17.89 ± 0.96 %
STOP ^{137}Cs Energy Resolution = 16.54 ± 1.20 %

Lifetime Extraction



Positron lifetimes are extracted from the spectrum given by C(t)

$$C(t) = R(t) \otimes S(t) = \int_0^\infty R(t - t_0)\, S(t_0)\, dt_0$$

Timing resolution function, R(t)

$$R(t) = \frac{1}{\sqrt{2\,\pi\,\sigma^2}}\, exp\left[-\frac{(t\ - t_0)^2}{2\sigma^2}\right]$$

$$\text{FWHM} = 2\sqrt{2\ln 2}\,\sigma$$

Multiexponential Model

$$S(t_0) = \sum_{i=1} \frac{I_i}{\tau_i}\, exp\left[-\frac{t}{\tau_i}\right]$$

Lifetime intensity: I
Positron lifetime: τ
Annihilation rate: λ
Trapping rate: κ

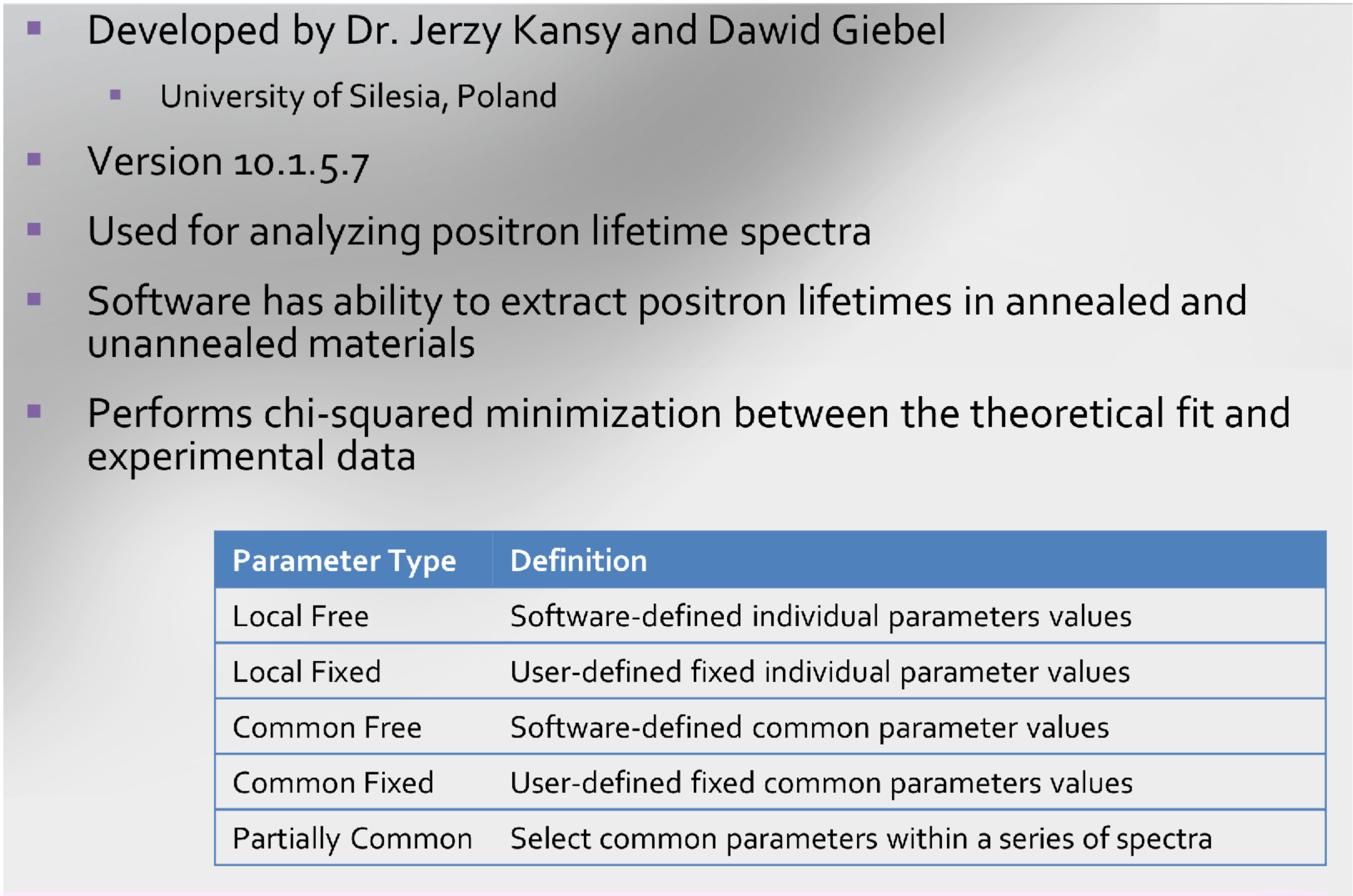
Two-State Trapping Model

$$S(t_0) = c_b\lambda_b + \sum_{i=1}^{n} c_i\lambda_i$$

$$c_b = exp(-\,\lambda_b\ t)$$

$$c_i = \frac{\kappa_i}{\lambda_b -\ \lambda_i}[exp(-\,\lambda_i\ t)\ - exp(-\,\lambda_b\ t)]$$

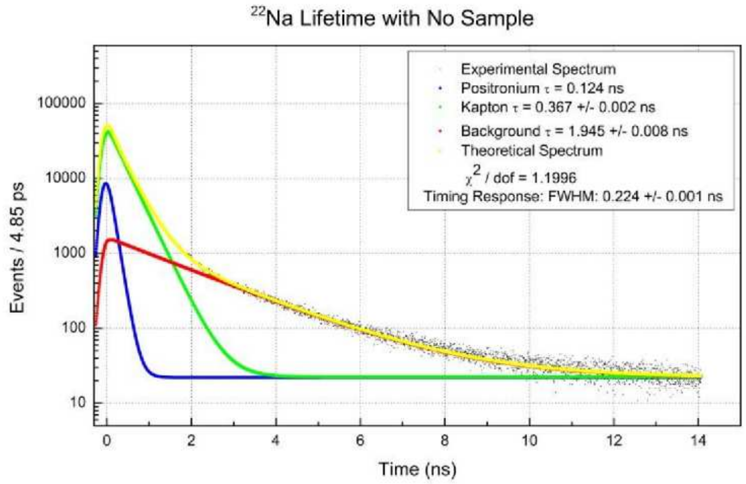
Kansy LT10 Software



- Developed by Dr. Jerzy Kansy and Dawid Giebel
  - University of Silesia, Poland
- Version 10.1.5.7
- Used for analyzing positron lifetime spectra
- Software has ability to extract positron lifetimes in annealed and unannealed materials
- Performs chi-squared minimization between the theoretical fit and experimental data

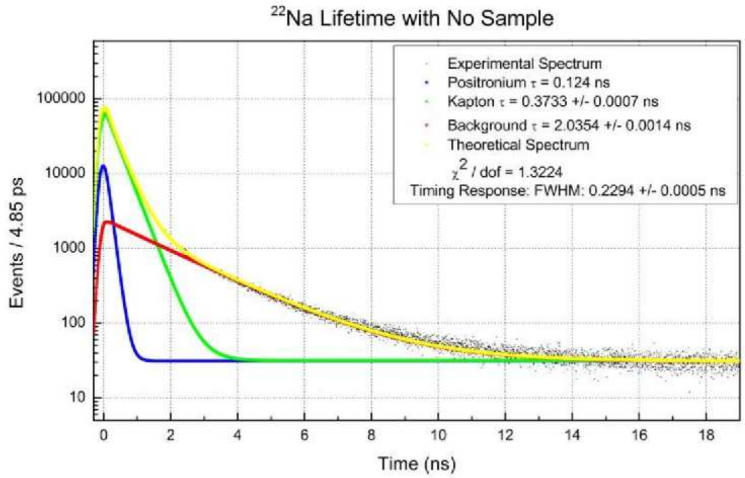
Parameter Type	Definition
Local Free	Software-defined individual parameters values
Local Fixed	User-defined fixed individual parameter values
Common Free	Software-defined common parameter values
Common Fixed	User-defined fixed common parameters values
Partially Common	Select common parameters within a series of spectra

Source-based Fast Plastic Results: ^{22}Na without Sample

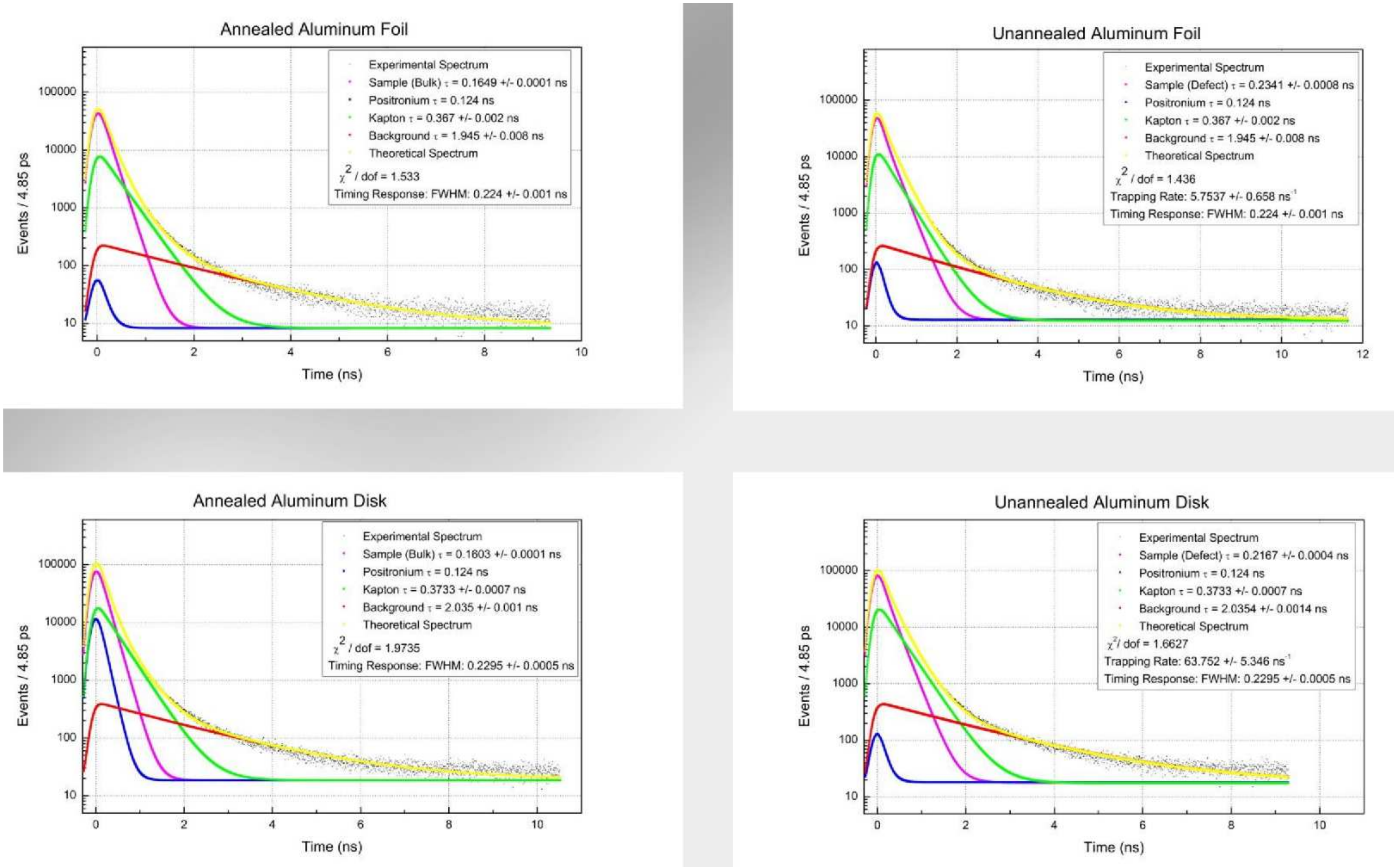


Thin Foils Background

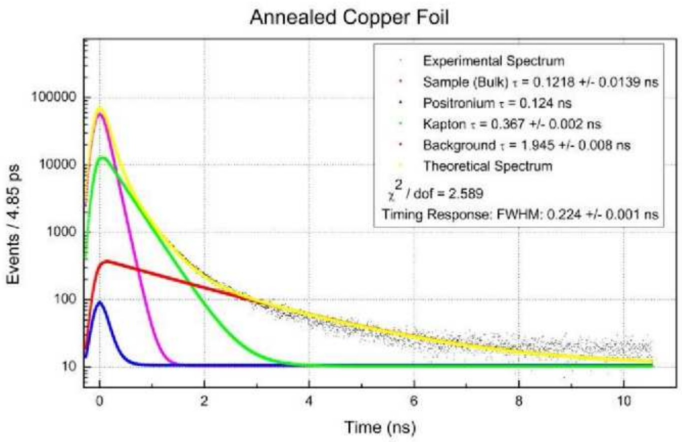
Thin Disks Background

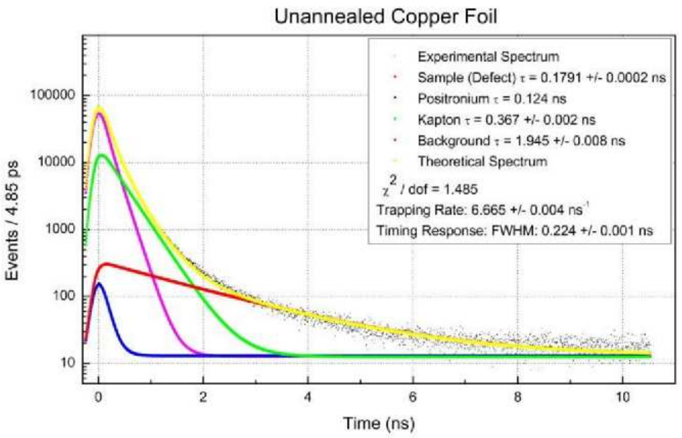


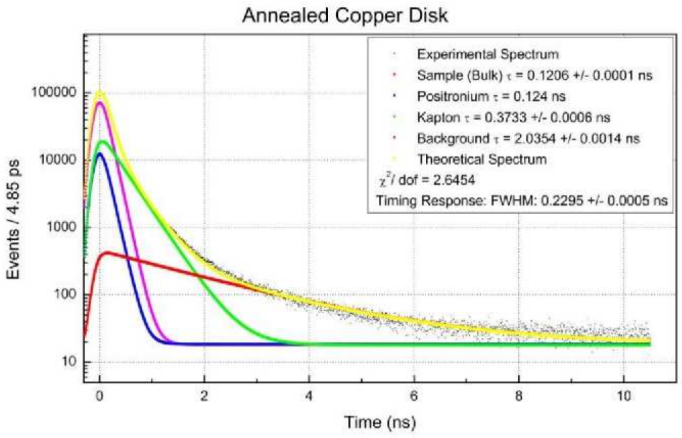
Source-based Fast Plastic Results: Aluminum

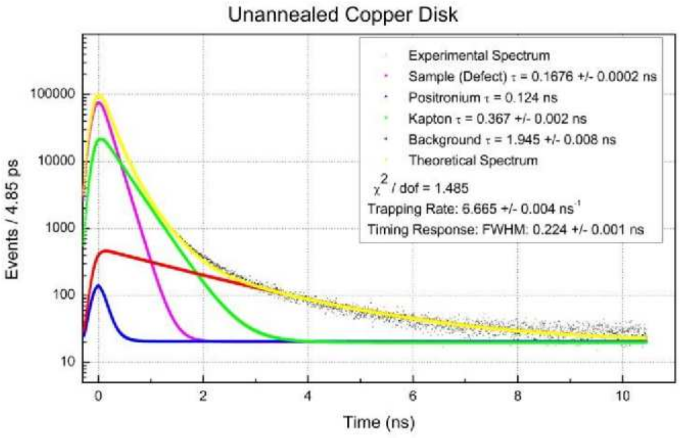


Source-based Fast Plastic Results: Copper

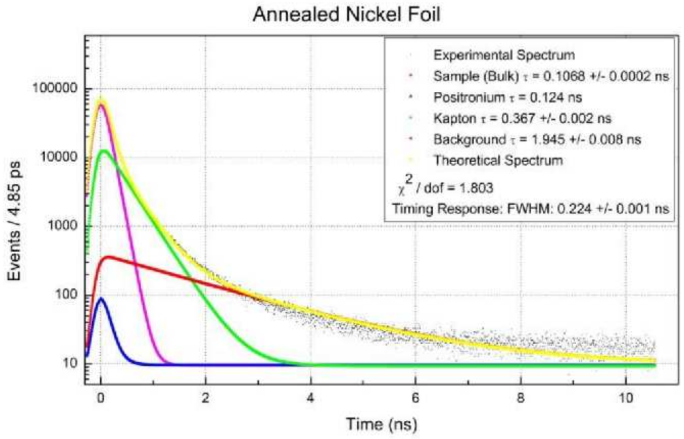

Annealed Copper Foil

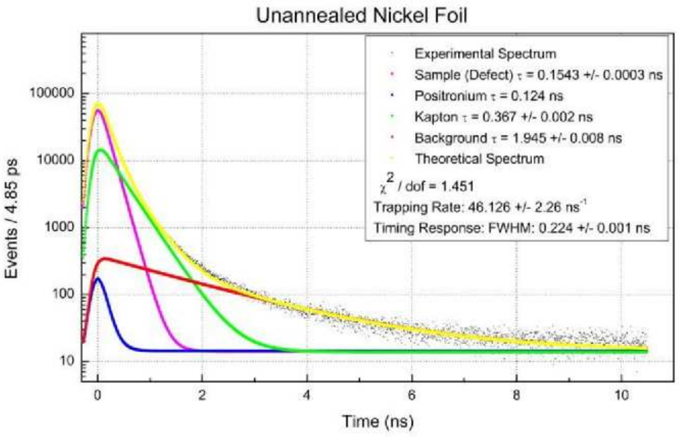

Unannealed Copper Foil

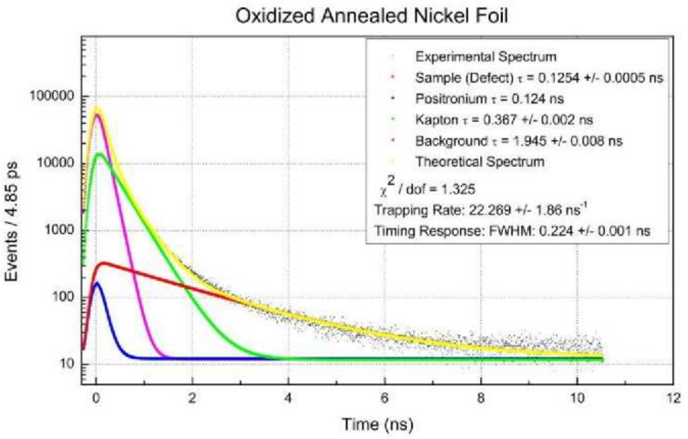

Annealed Copper Disk


Unannealed Copper Disk

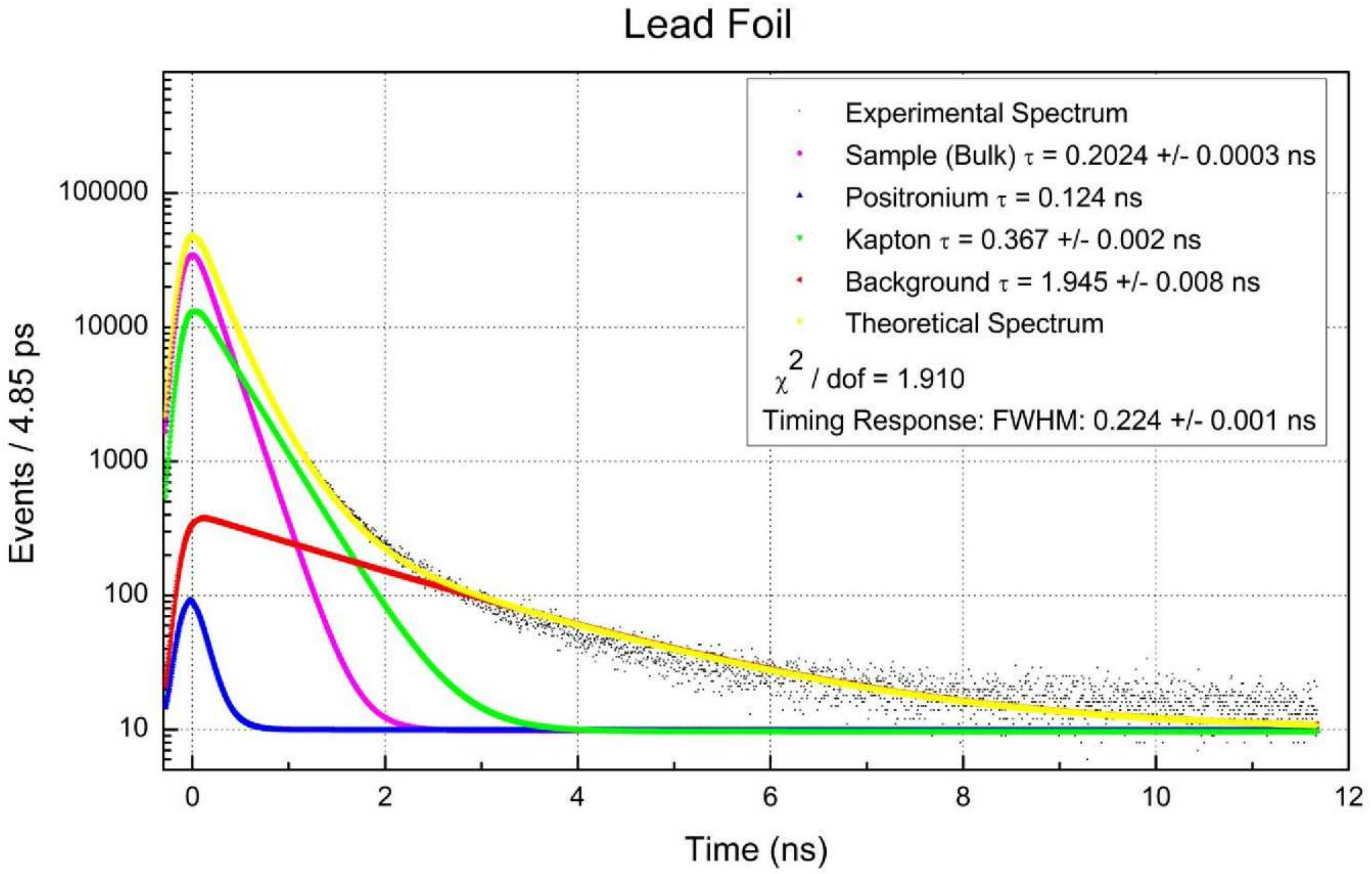
Source-based Fast Plastic Results: Nickel


Annealed Nickel Foil


Unannealed Nickel Foil

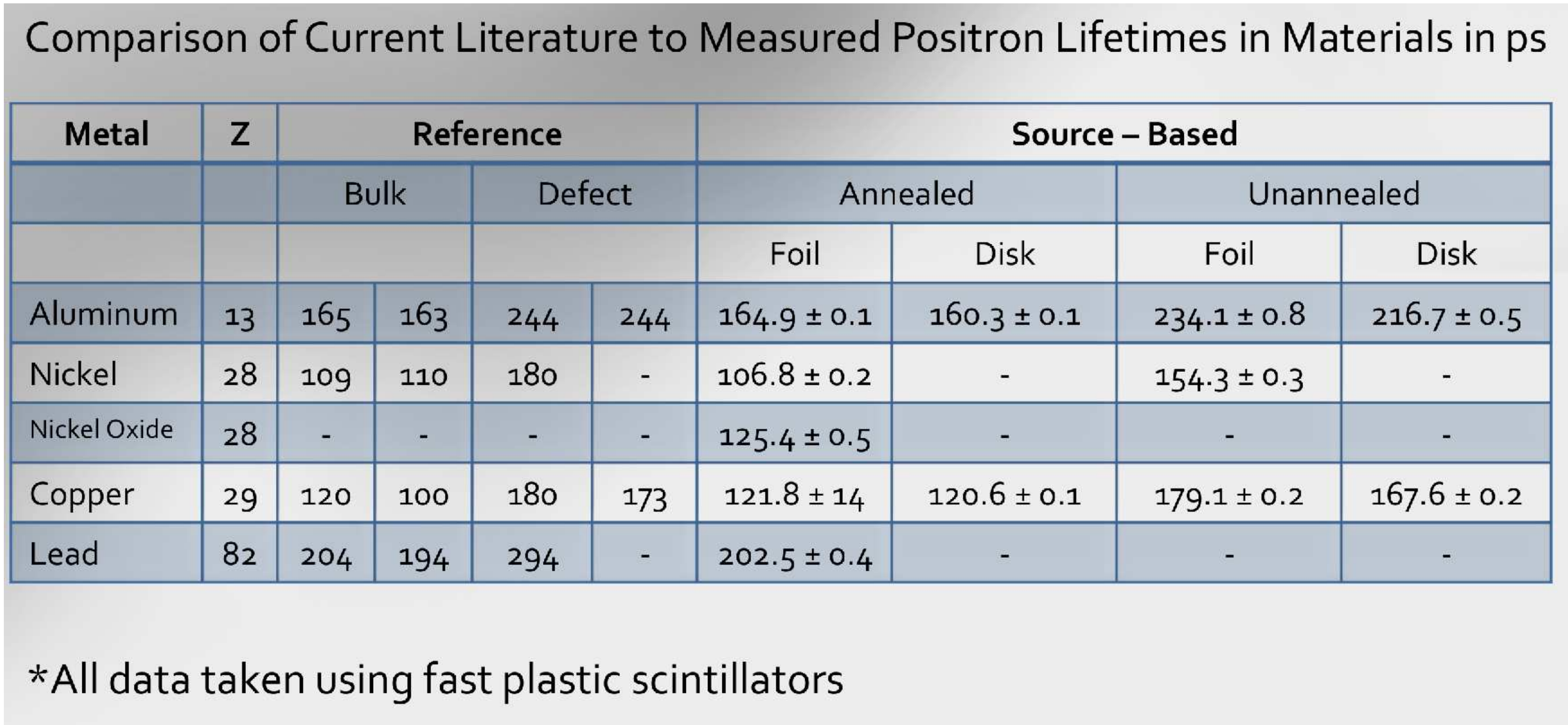

Oxidized Annealed Nickel Foil

Source-based Fast Plastic Results: Lead



Positron Annihilation Lifetime Spectroscopy

Source-based Fast Plastic Results: Summary



Comparison of Current Literature to Measured Positron Lifetimes in Materials in ps

Metal	**Z**	**Reference**				**Source – Based**			
		Bulk		Defect		Annealed		Unannealed	
						Foil	Disk	Foil	Disk
Aluminum	13	165	163	244	244	164.9 ± 0.1	160.3 ± 0.1	234.1 ± 0.8	216.7 ± 0.5
Nickel	28	109	110	180	-	106.8 ± 0.2	-	154.3 ± 0.3	-
Nickel Oxide	28	-	-	-	-	125.4 ± 0.5	-	-	-
Copper	29	120	100	180	173	121.8 ± 14	120.6 ± 0.1	179.1 ± 0.2	167.6 ± 0.2
Lead	82	204	194	294	-	202.5 ± 0.4	-	-	-

*All data taken using fast plastic scintillators

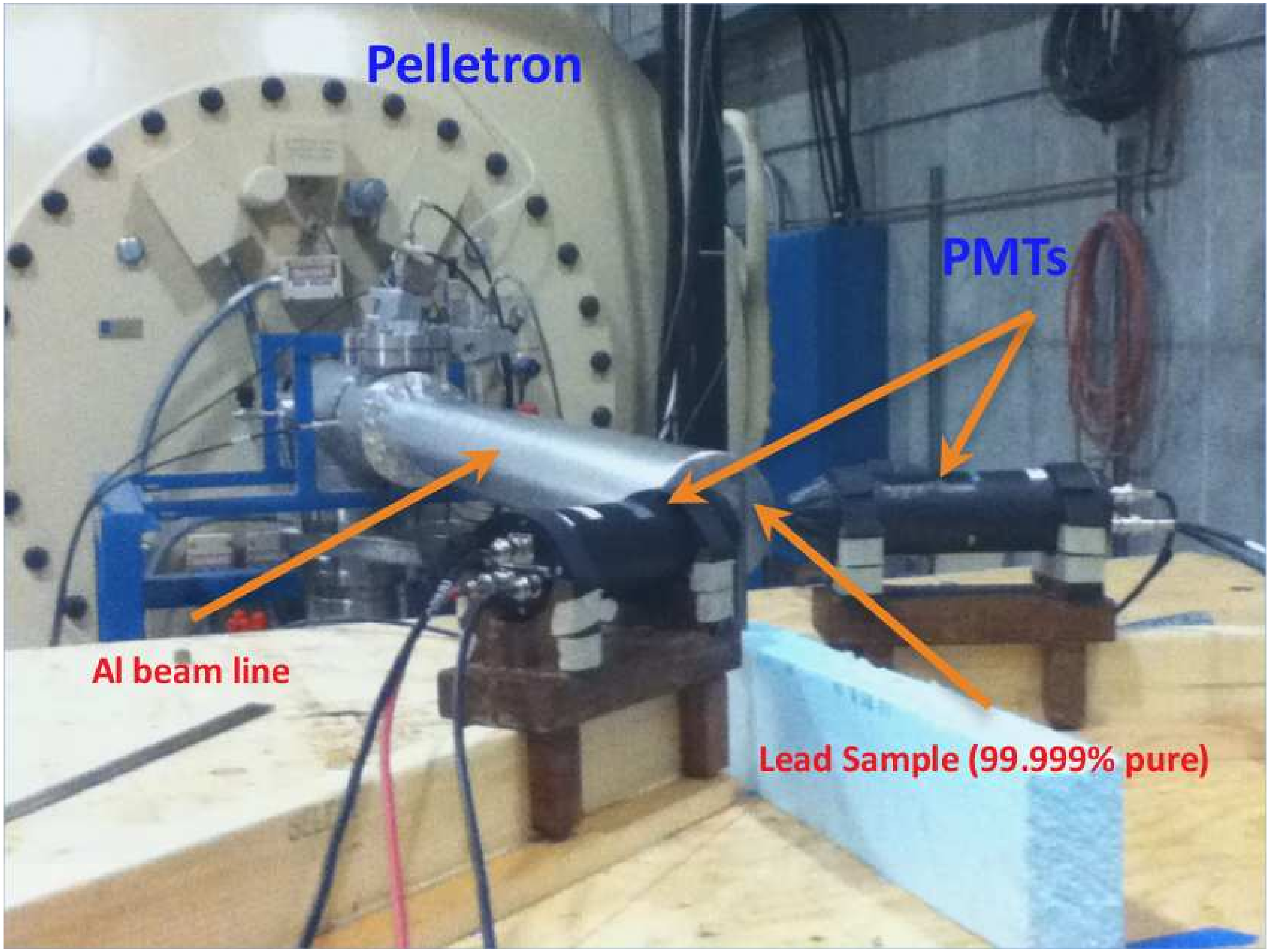
Source-based Conclusions

- A high precision lifetime spectrometer has been constructed and successfully benchmarked; Brian Wieland's Masters degree
- Bulk positron lifetimes in high-purity aluminum, copper, nickel, and lead are in excellent agreement with current literature values
- Measured defect lifetime values may differ from literature values as a result of different types of defects within the unannealed samples
- Spectrometer is capable of resolving difference between annealed and unannealed samples
- Spectrometer is capable of detecting surface defects caused by oxidation of materials
- Additional studies will need to be completed for BaF_2 scintillators
- This study lays the foundation for future accelerator-based positron annihilation lifetime spectroscopy to access volume defect densities

Accelerator-based PALS

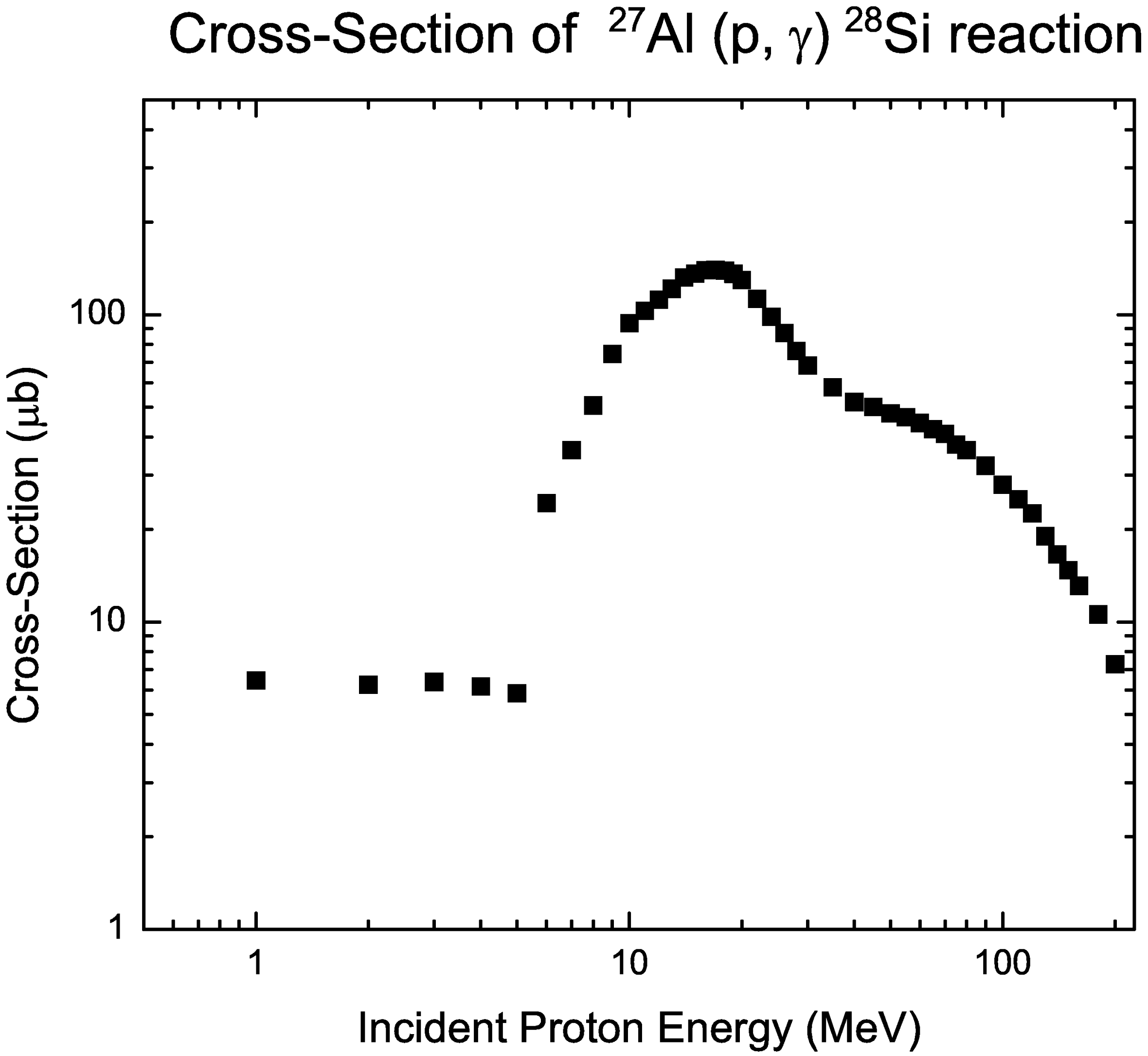
- Initial Goal: Reproduce source-based lifetime measurements using the $^{27}\mathrm{Al}(p,\gamma)^{28}\mathrm{Si}$ proton-capture reaction
- Decay of excited Si nucleus often produces two or more photons: One is used as the positron “birth” signal while the other produces $e^{+}e^{-}$ pairs in the sample leading to the 511keV “death” signal
- Initial measurements used 2MeV Van de Graaf, however 10μA maximum beam current produced extremely limited event-rates ($\sim$0.2Hz)
- Pelletron capable of 200μA and up to 8MeV; fabricated custom Al beamline; first PALS experiments conducted May 2012

Accelerator-based Experimental Setup



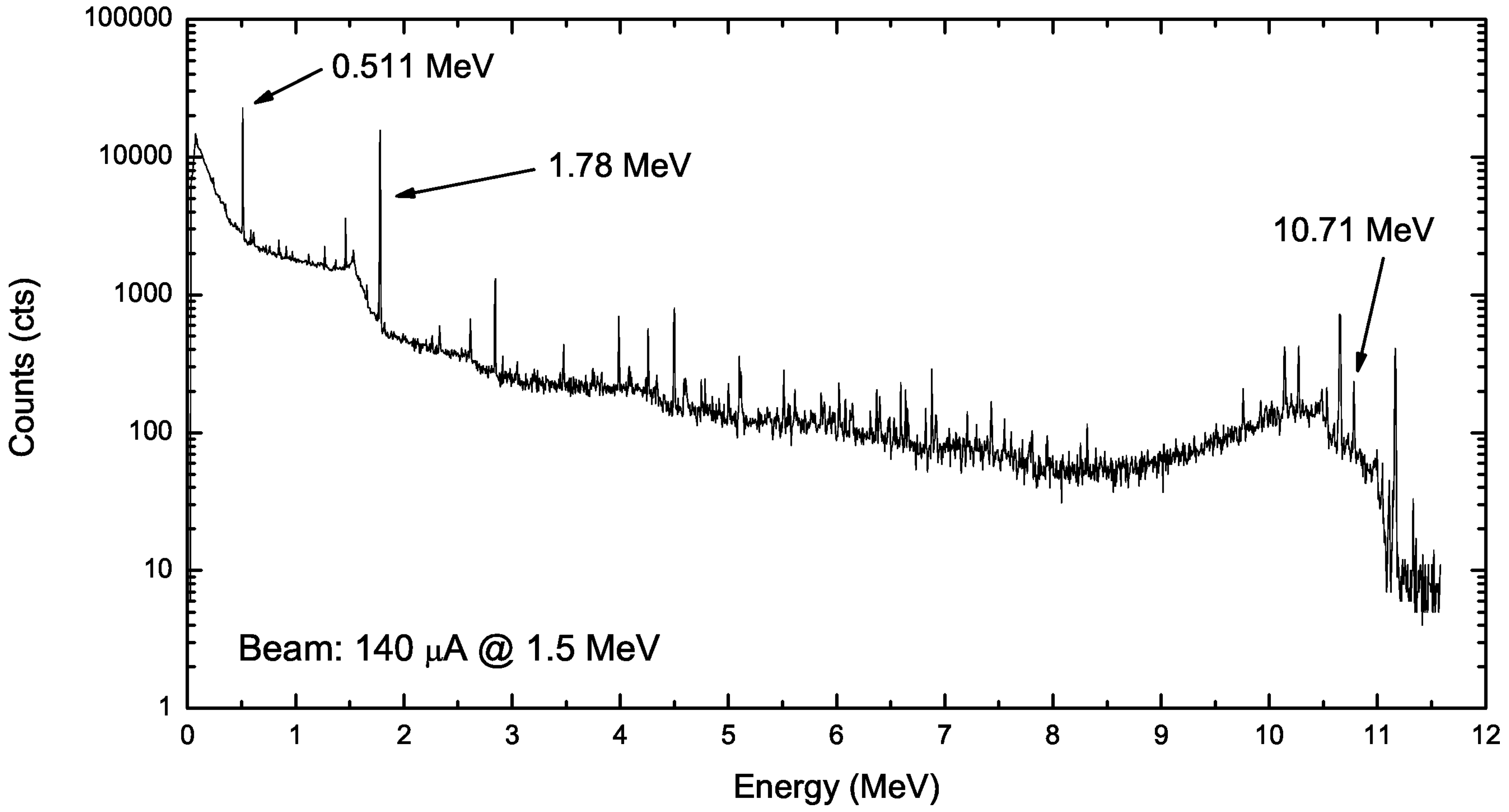
Positron Annihilation Lifetime Spectroscopy

^{27}Al(p,γ)^{28}Si Cross Section



Positron Annihilation Lifetime Spectroscopy

^{27}Al(p,γ)^{28}Si Photon Spectrum



Pelletron-based PALS: Studies

- Event rates for various sample-detector geometries – sample size and distance from Al endcap very important
- Event rates for various shielding configurations – need to block line of sight between two detectors, helped to block stop detector from line of sight to Al endcap, although in general shielding is problematic for obvious reasons
- Event rates and energy spectra for different proton energies – found that beam energy needs to be 10-20% above 0.992MeV resonance; beyond 2 - 2.5MeV, individual pmt event rates/system deadtime saturated

Positron Annihilation Lifetime Spectroscopy

Shielding-Sample-Detector Configurations


MPANT File: 20120717-003


MPANT File: 20120717-008


MPANT File: 20120717-014


MPANT File: 20120717-006


MPANT File: 20120717-009


MPANT File: 20120717-015


MPANT File: 20120717-007


MPANT File: 20120717-013


MPANT File: 20120717-016

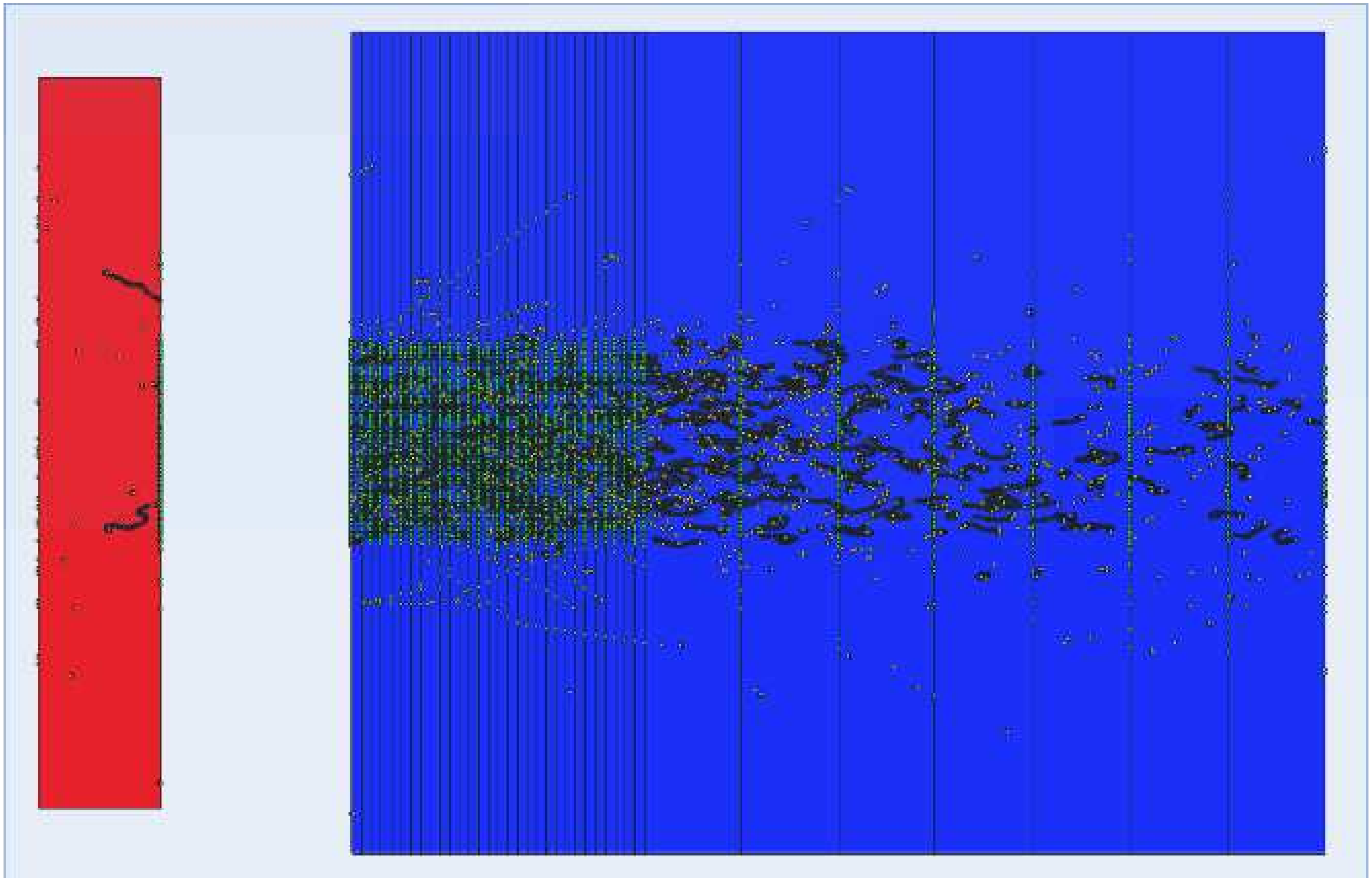
Pelletron-based PALS: Issues

- Given the current spectrometer design, the PALS coincidence event rates are only 2 - 5Hz at best depending on beam current, sample thickness and shielding; $\Longrightarrow$ 60 hour data-runs to reach 1M events
- Experiment suffers from isotropic photon beam and small solid angle acceptance of detectors in conjunction with coincidence-style measurement
- Background events difficult to separate from sample events
- BaF_2 scintillator spectra from Pelletron not yet fully understood–this should provide better background rejection
- Detector signals often exceeded max specs for processing electronics – needed to operate pmts at much lower voltages than optimal
-

Accelerator-based PALS: Ideas and Future Work

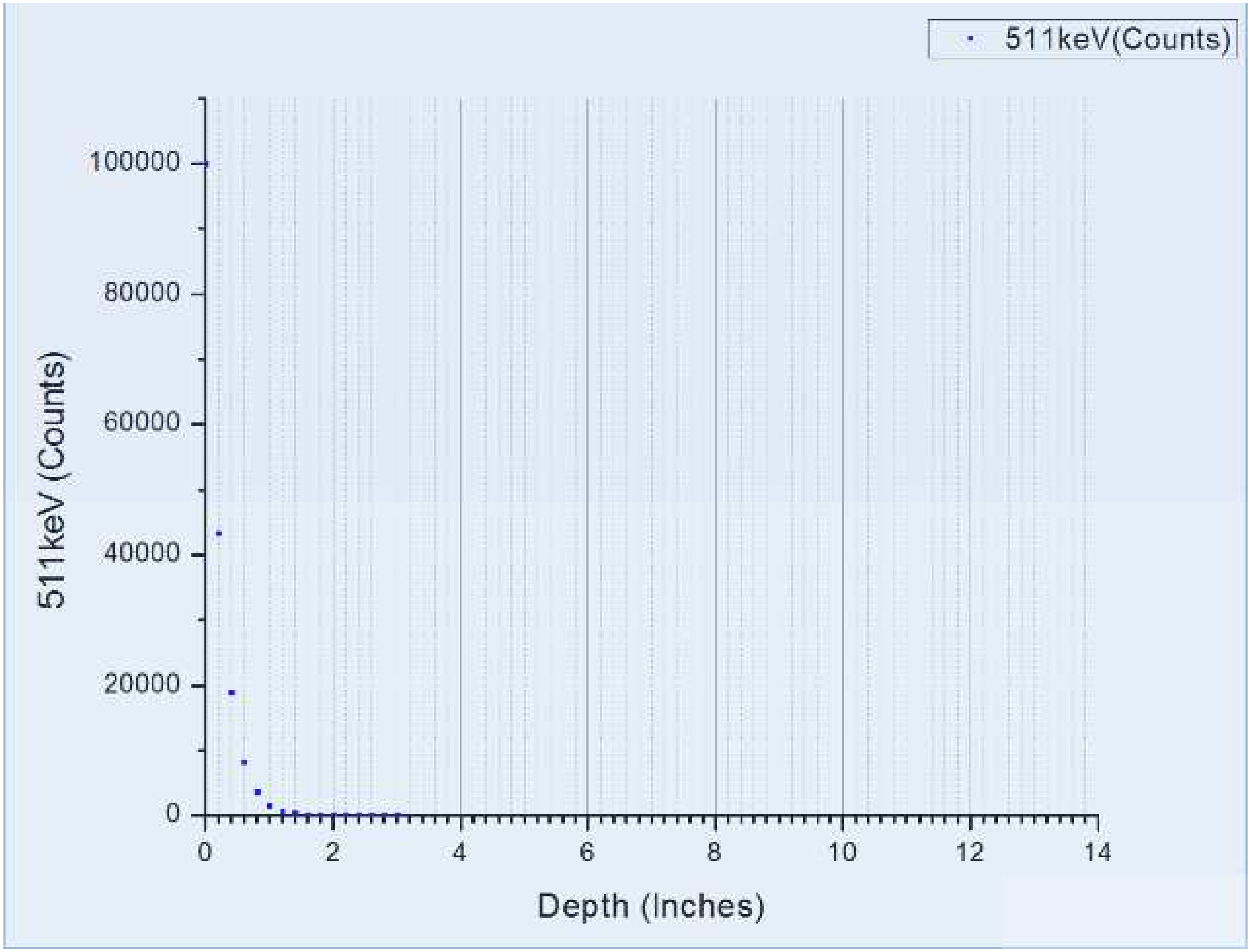
- Use fluorine reaction instead of aluminum reaction – can get 4 times more rate with higher photon energies – but still isotropic “beam”
- A very high rep-rate or CW electron to bremsstrahlung beam would be great; ideally ... energy tunable CW positron beam ...
- Increase numbers of detectors to improve solid angle coverage
- Employ back-to-back 511keV (stop) detectors for greatly improved background rejection
- Use two CFDD – currently we have only one CFDD and one CFD – accepts more background
- Simulation work is ongoing to understand issues related to shielding, sample thickness limitations for 511keV photon escape, photon multiplicities from excited Si decays,...

Simulation Studies: MC-NPX Input Deck

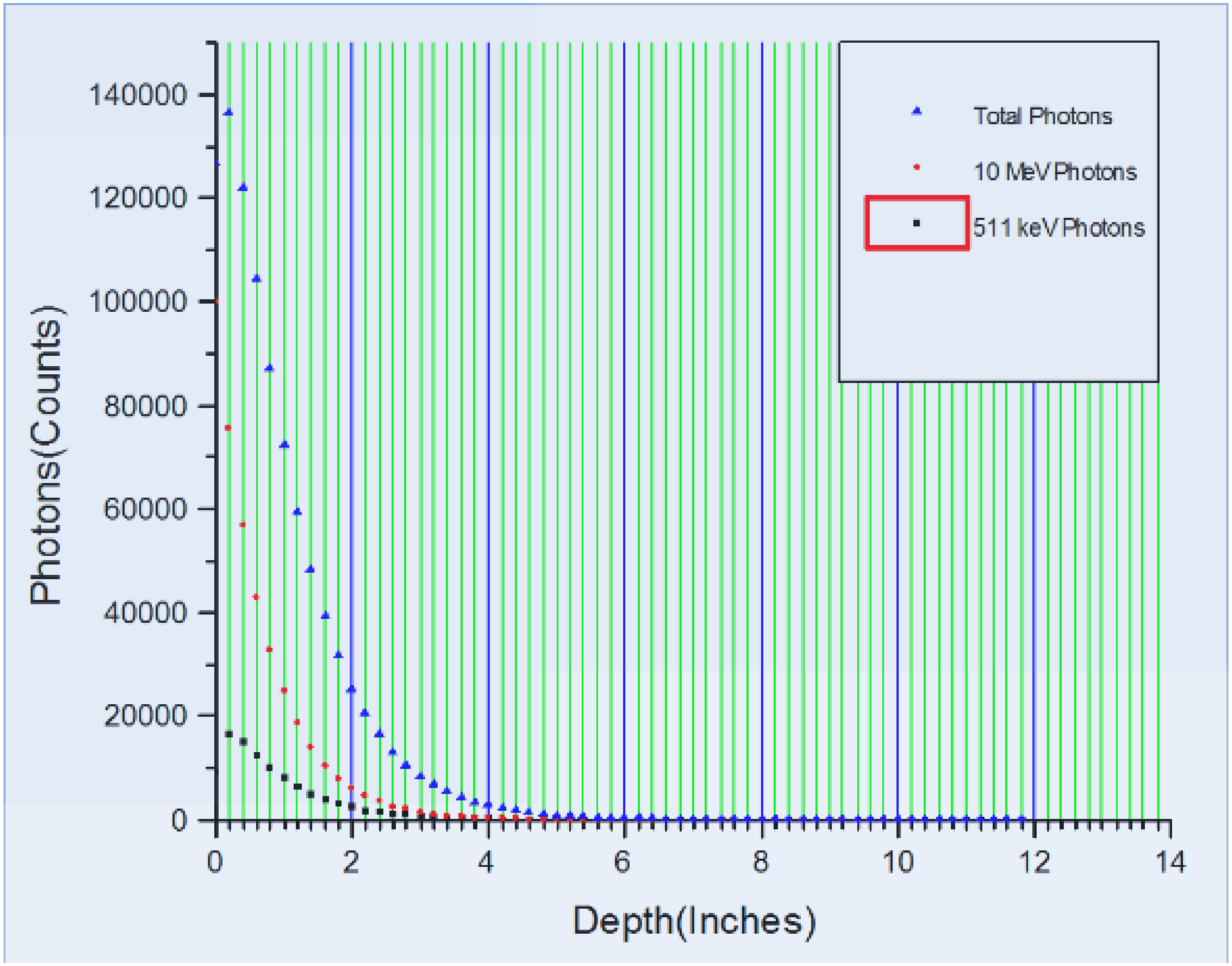


Positron Annihilation Lifetime Spectroscopy

Simulation Studies: 511 γ Survival



Simulation Studies: All γ Survival



Accelerator-based PALS: Conclusions

- R & D for AG-PALS (volume defect essay) is still in early stages as there are many technical hurdles to overcome
- Obviously event rate is a big problem with the current measurements; if we reduce our measurement statistics requirements to 200k events–could achieve in 12 hours with Pelletron
- The PALS data is rich with information as compared with PAES but technically much more challenging
- Investment in both accelerator and spectrometer design is needed for viable AG-PALS and even more so for if one is to achieve “imaging” measurements
- There is currently one PhD candidate working on this and potentially more in the queue