Interrogating Nuclear Reactor Materials with Positrons

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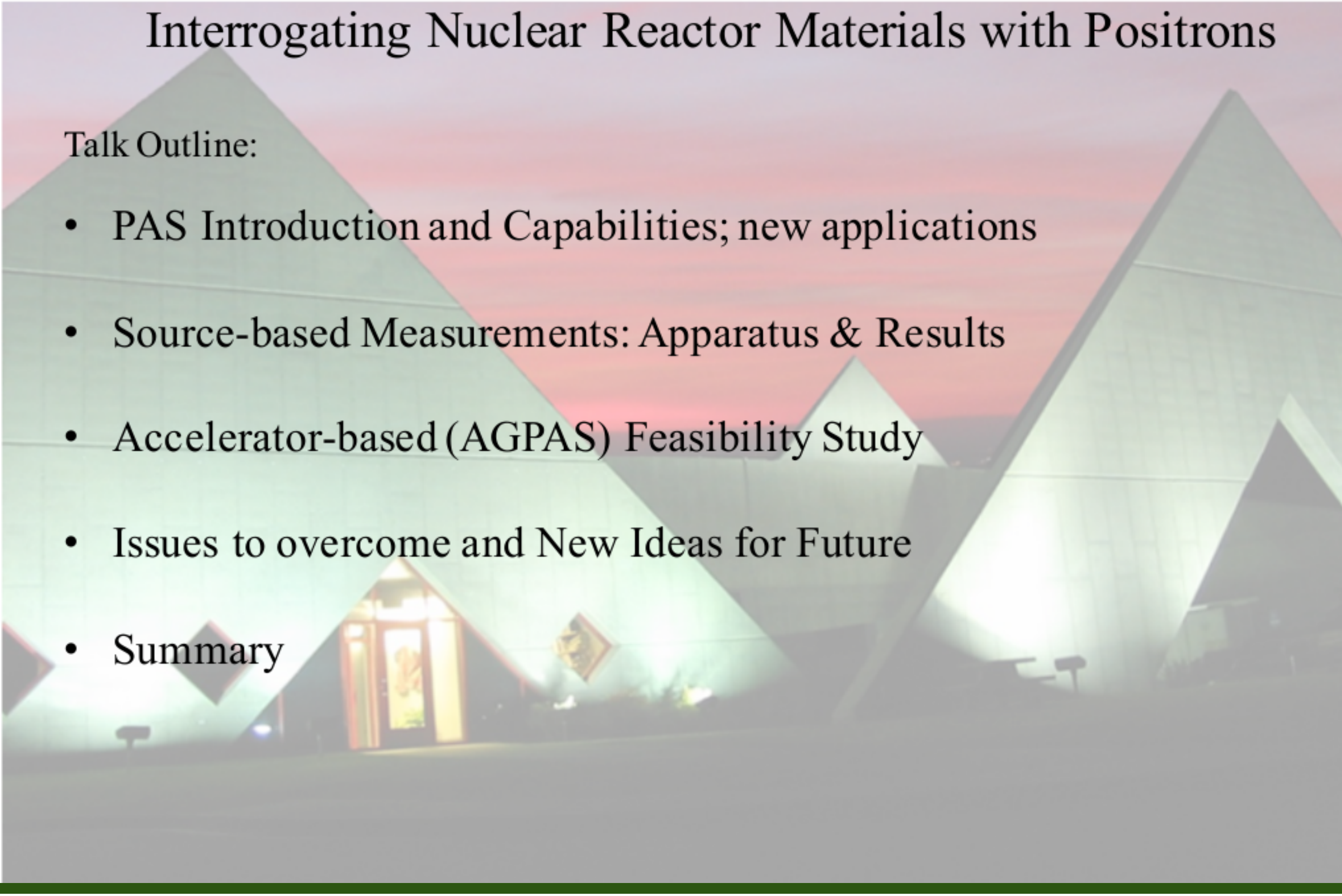
PHYSICS

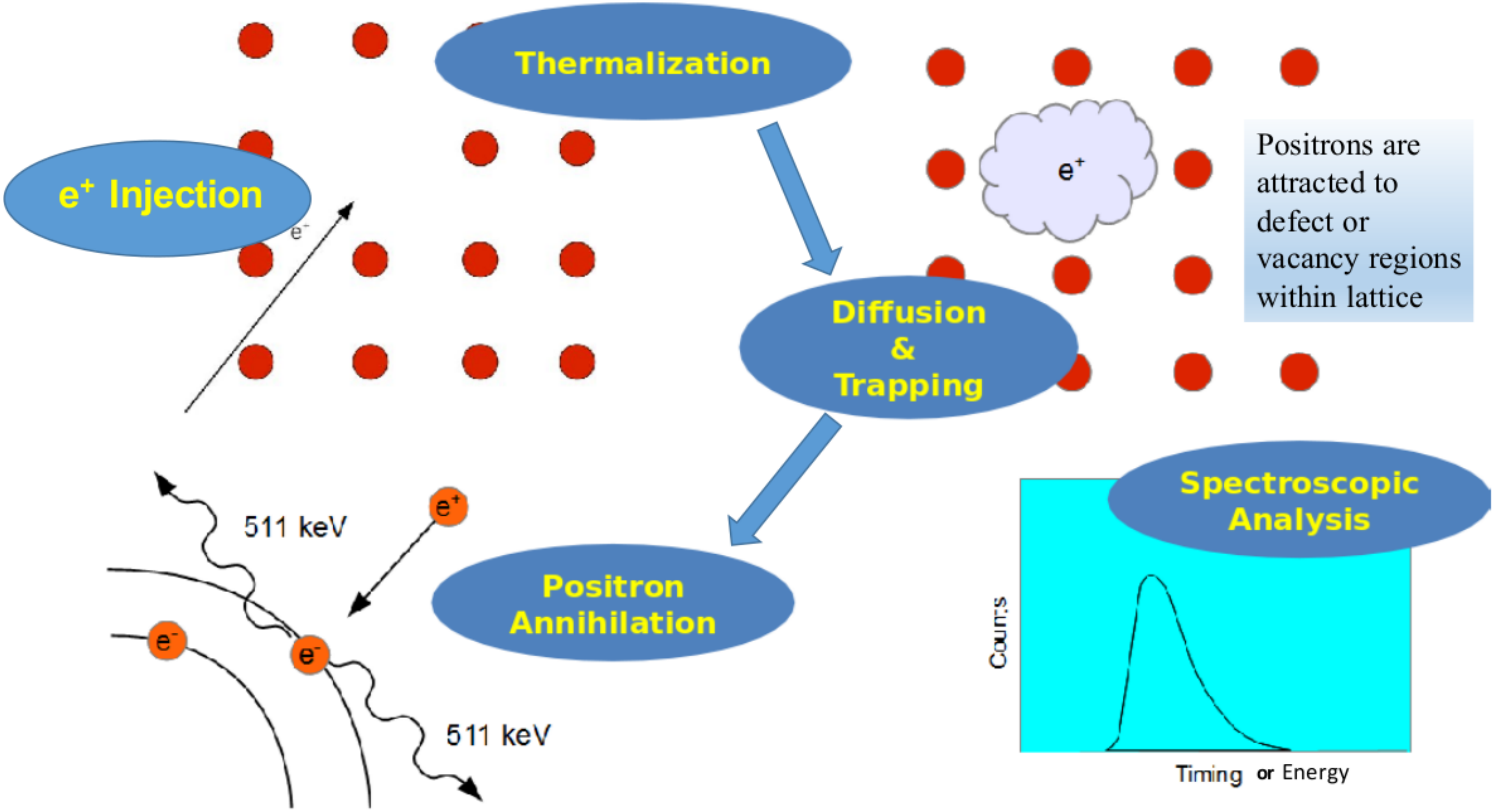
Interrogating Nuclear Reactor Materials with Positrons



Talk Outline:

- PAS Introduction and Capabilities; new applications
- Source-based Measurements: Apparatus & Results
- Accelerator-based (AGPAS) Feasibility Study
- Issues to overcome and New Ideas for Future
- Summary

Positron Annihilation Spectroscopy Overview



e+ Injection

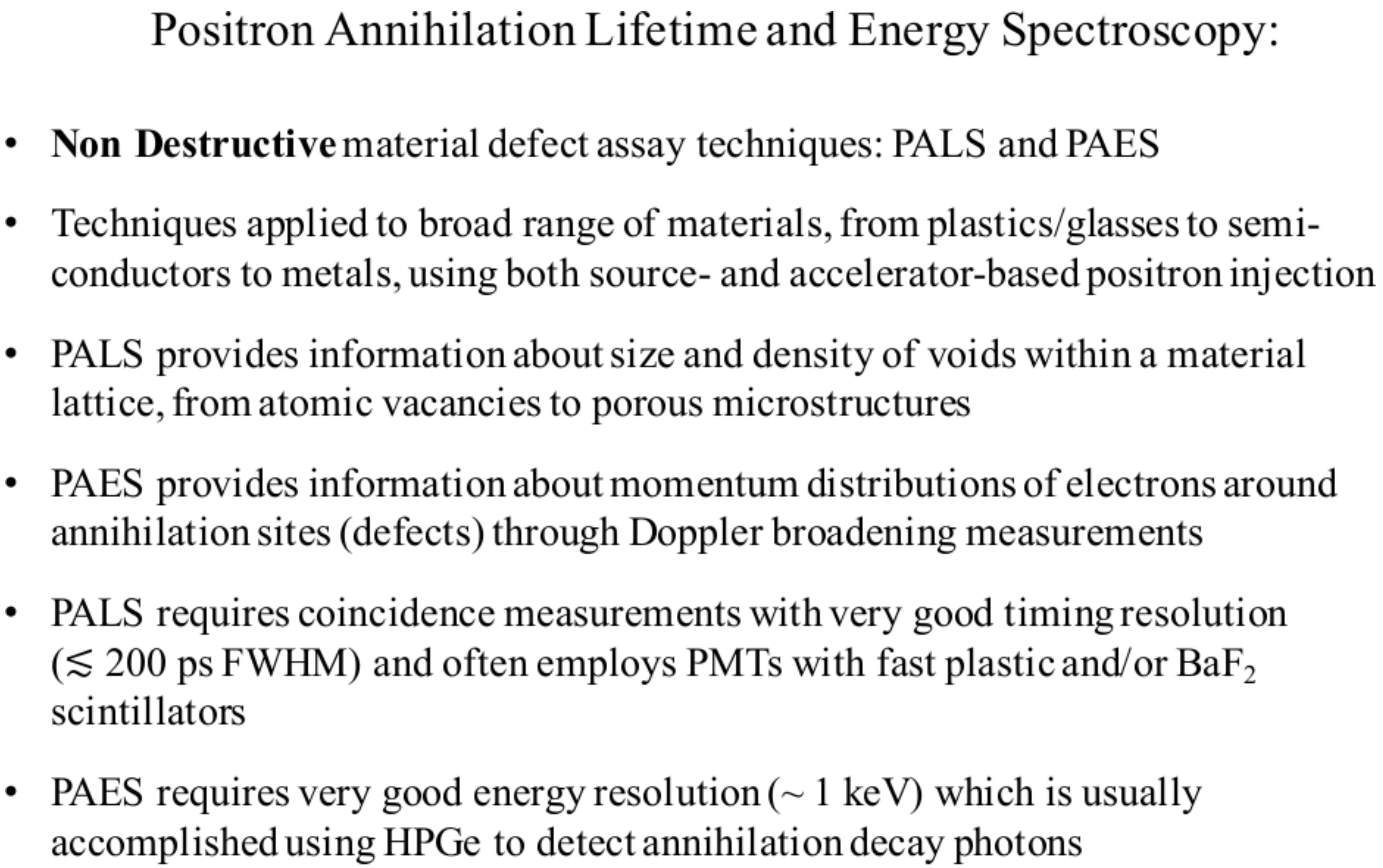
Thermalization

Diffusion & Trapping

Positrons are attracted to defect or vacancy regions within lattice

Positron Annihilation

Spectroscopic Analysis



Positron Annihilation Lifetime and Energy Spectroscopy:

- **Non Destructive** material defect assay techniques: PALS and PAES
- Techniques applied to broad range of materials, from plastics/glasses to semi-conductors to metals, using both source- and accelerator-based positron injection
- PALS provides information about size and density of voids within a material lattice, from atomic vacancies to porous microstructures
- PAES provides information about momentum distributions of electrons around annihilation sites (defects) through Doppler broadening measurements
- PALS requires coincidence measurements with very good timing resolution ($\lesssim 200$ ps FWHM) and often employs PMTs with fast plastic and/or BaF_2 scintillators
- PAES requires very good energy resolution (~ 1 keV) which is usually accomplished using HPGe to detect annihilation decay photons

Original Application Concept for AG-PAS

Past DOD-sponsored projects with private sector partners:

- Wyle Inc.
- Axter Inc.
- TechSource Inc.
- Positron Systems Inc.

- Positron lifetime measurements by proton capture
  F.A. Selim, D.P. Wells and J.F. Harmon
  Review of Scientific Instruments 76, 33905 (2005)
- Development of accelerator-based γ-ray-induced positron annihilation spectroscopy technique
  F.A.Selim, D.P.Wells, J.F. Harmon and J. Williams
  Journal of Applied Physics 97, 113539 (2005)
- Defect imaging of structural objects using positron annihilation spectroscopy
  A.W. Hunt, R. Spaulding, J. Urban-Klaehn, J.F. Harmon and D.P. Wells
  Nuclear Instruments & Methods in Physics Research B 241, 362 (2005)
- Gamma-induced positron annihilation spectroscopy and application to radiation-damaged alloys
  D.P. Wells, et.al
  Nuclear Instruments & Methods in Physics Research A 562, 688 (2006)

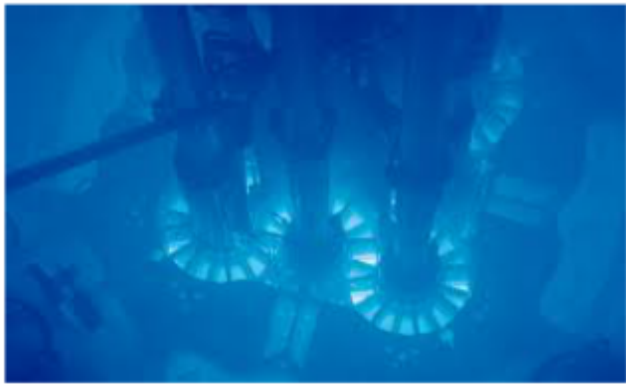
Non-destructive Bulk Defect Assay
(*The Dream: 3D imaging*)

Structural Materials failure cost the U.S. economy approximately $50 Billion/year. Non-destructive testing for thick structural materials is a multi-billion dollar industry, yet current techniques are not capable of reliably detecting the “approach to failure” of thick composites and many alloys.

New Application Concept: Nuclear Reactor Materials

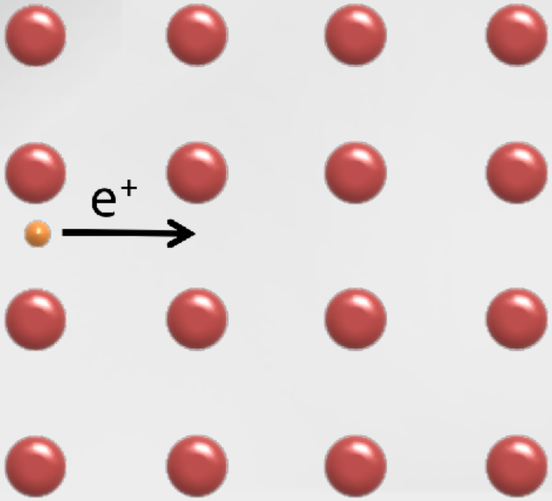
Possible Niche: Non-destructive, post fabrication bulk materials and sensor characterization

- Complimentary to MaCS/IMCL/IRC measurements – help connect characterizations of 100's of atoms to that of the bulk
- Probing depth dependent on positron energy and potentially tunable
- Non-destructive nature could allow for repeated measurements after successive neutron dose exposures and or thermal cycles – sensor characterization and "drift", strain creep, corrosion damage, …
- In general, non-destructive bulk characterizations seem to have vast potential applications:
    - Next generation reactor technologies
        - Claddings: metallic-glass, ODS, high-entropy, and coatings
    - Small modular reactors
    - Maintaining/sustaining present reactors
    -



Positron interactions: Thermalization

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



$E_p < 20\ MeV$, Ionization

$E_p < 5\ eV$, Plasmon excitation

$E_p < 0.1\ eV$, Positron-Phonon Interactions

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

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

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

Positron Intensity

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

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

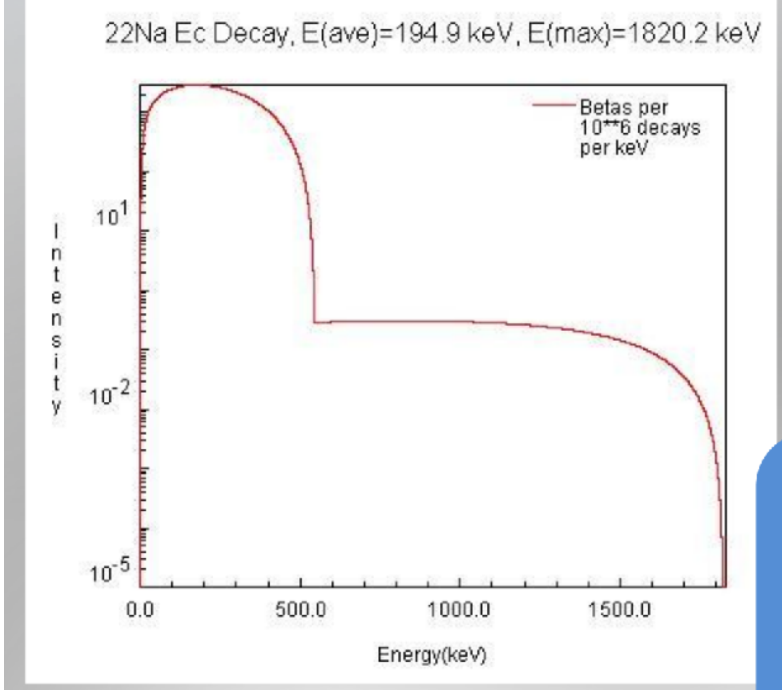
x: thickness of sample

α_+: positron absorption coefficient

E_{max}: maximum positron energy in MeV

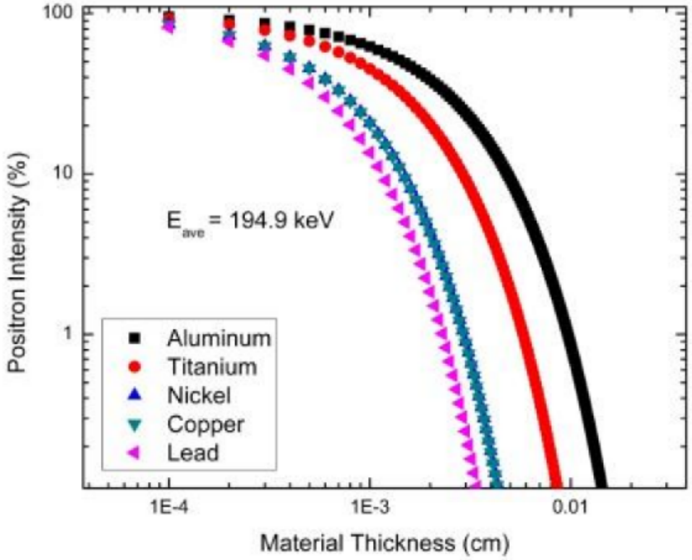
ρ: sample mass density in g/cm^3

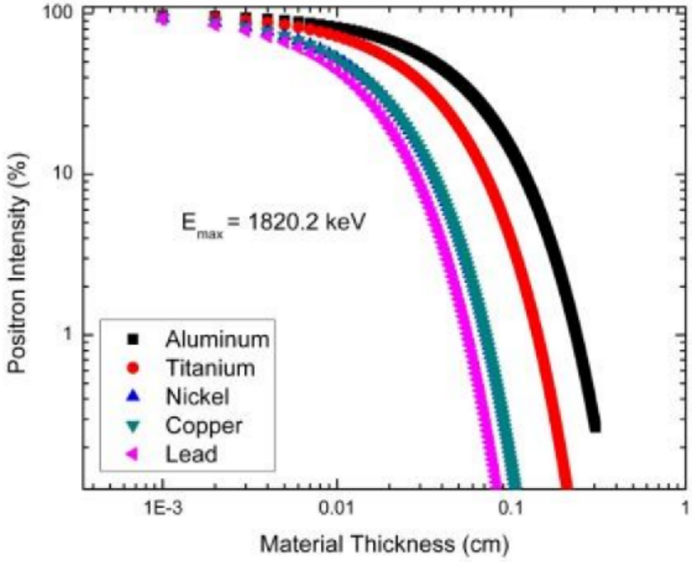
Positron interactions: Penetration Depth



Source positrons only penetrate 50 µm into materials

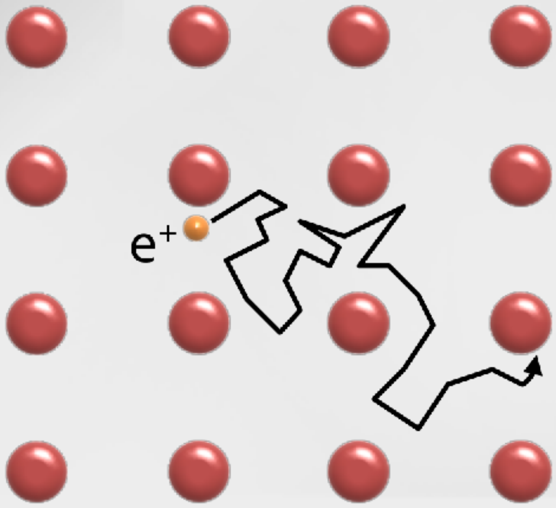
Material	Density (g/cm³)
Aluminum	2.70
Nickel	8.908
Copper	8.94
Lead	11.34





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

$$D_+ = \frac{v_{rms}\, l}{3} = \frac{{v_{rms}}^2\, \tau}{3} \qquad v_{rms} = \sqrt{\frac{3kT}{m^*}}$$

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

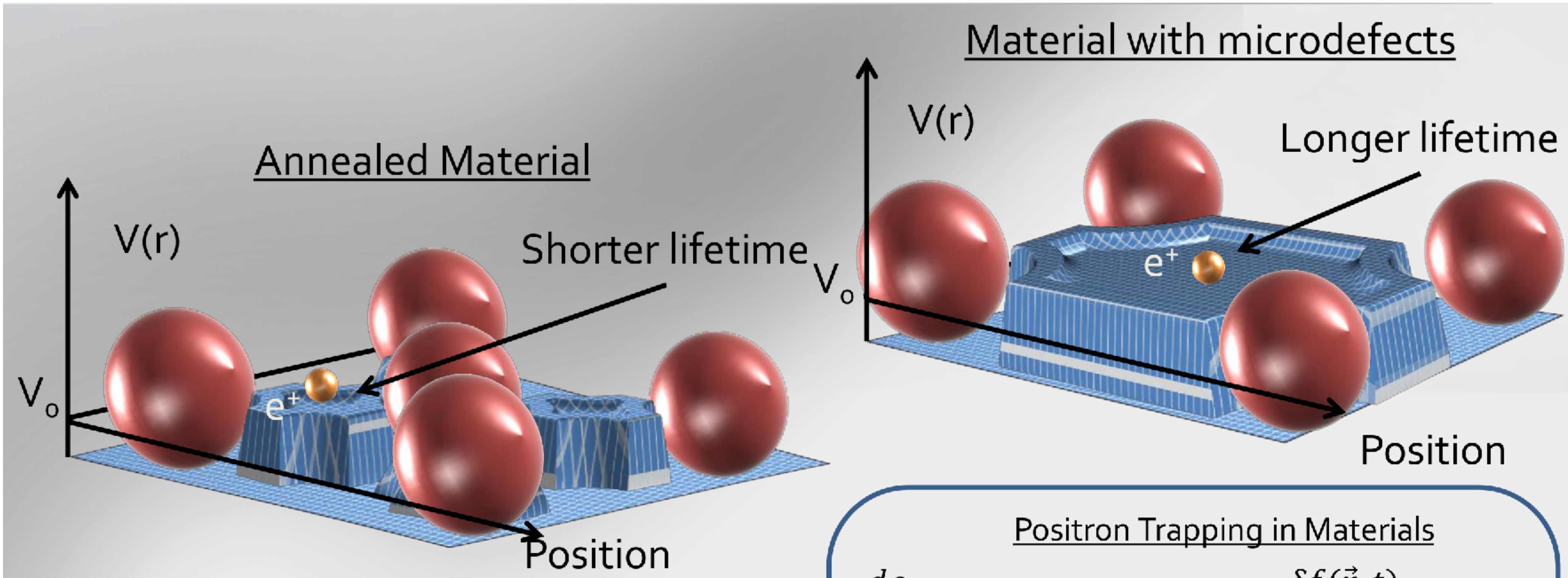
$$\langle l \rangle = \frac{3\, D_+}{v_{rms}}$$

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

Positron mean free path in metals ≈ 10^2 Å

Total diffusion length ≈ 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

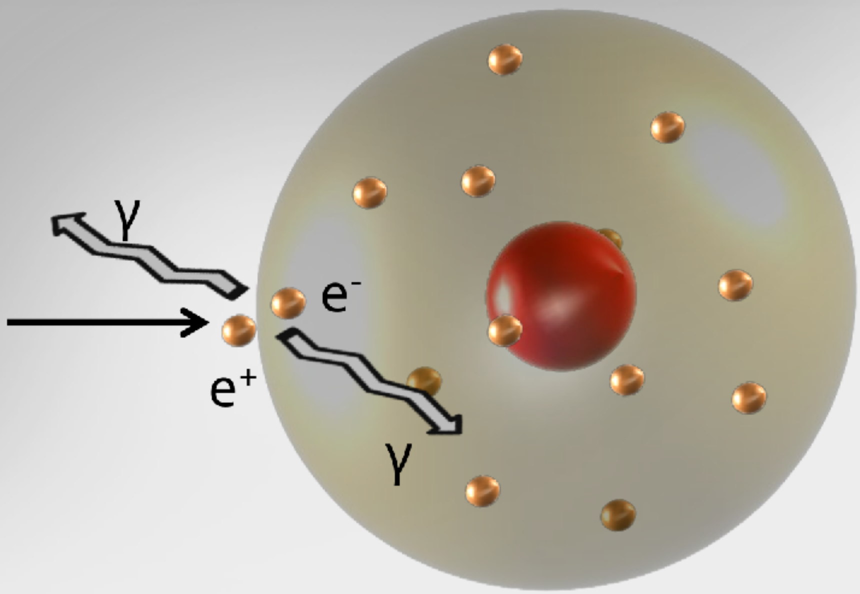
Positron interactions: Annihilation

Direct Annihilation

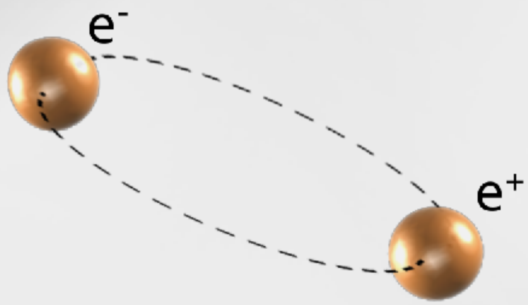
$$\sigma_{2\gamma} = \frac{4\,\pi\,r_0{}^2}{\gamma+1}\left[\frac{\gamma^2+4\gamma+1}{\gamma^2\;-1}\ln\left(\gamma+\sqrt{\gamma^2-1}\right)+\sqrt{\gamma^2-1}\;-\;\frac{\gamma+3}{\sqrt{\gamma^2-1}}\right]$$

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

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

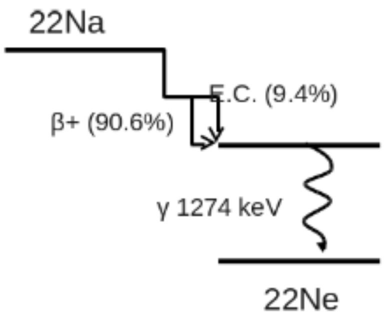
ortho-positronium (o-Ps):

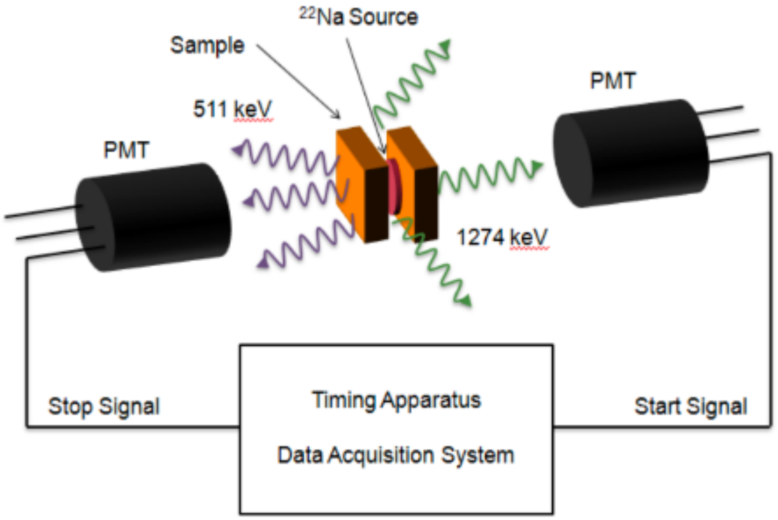
- Triplet state
- 142 ns lifetime
- 3 γ emission favored

$$\boxed{m_e c^2 + m_e c^2 \rightleftarrows E_\gamma + E_\gamma}$$

ISU/IAC PALS Apparatus (Source-based)

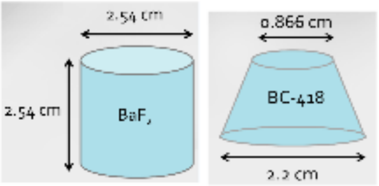
50 μC ^{22}Na source/Kapton film

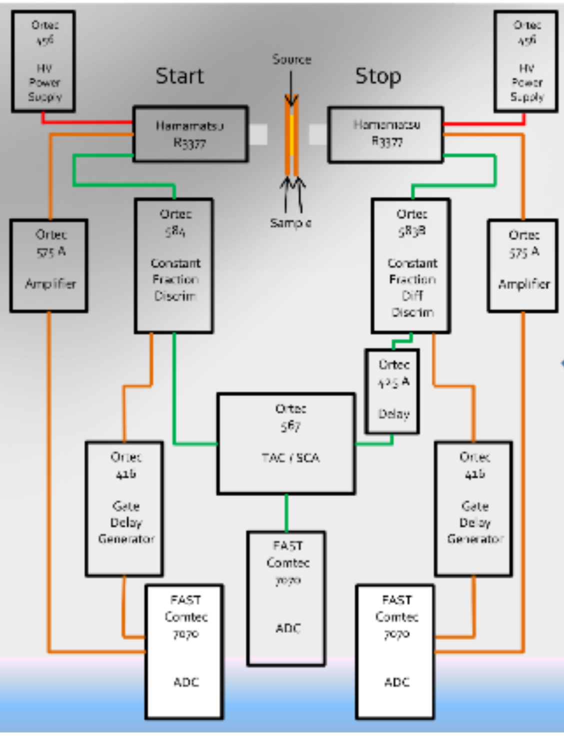




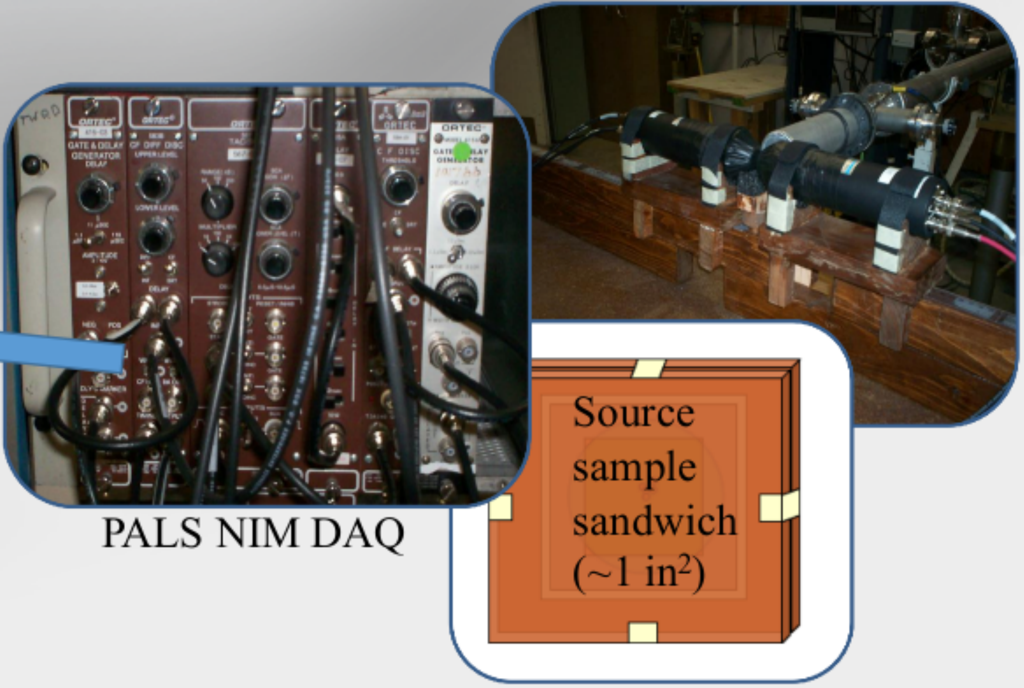
Hamamatsu R3377 PMTs

PMT scintillators





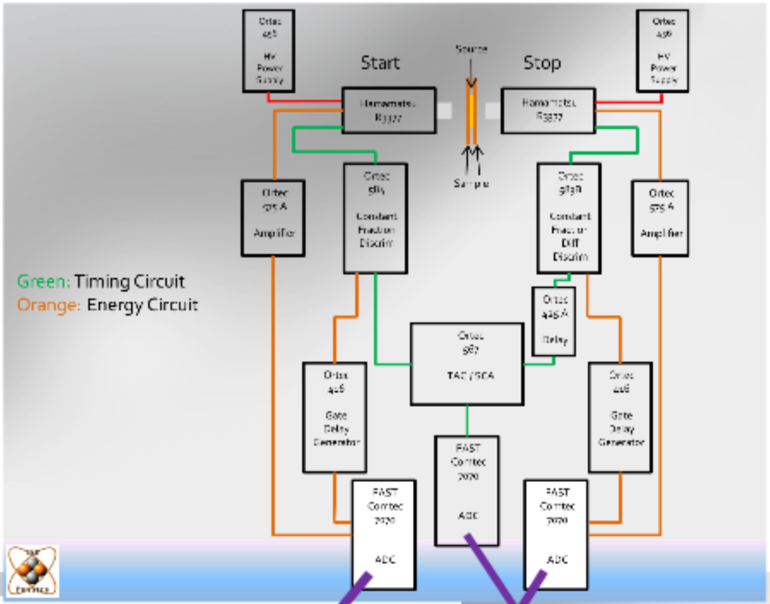
Green: Timing Circuit
Orange: Energy Circuit



PALS NIM DAQ

Source sample sandwich (~1 in^2)

Lifetime Extraction



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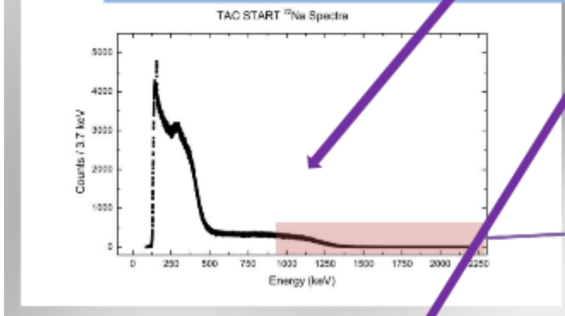
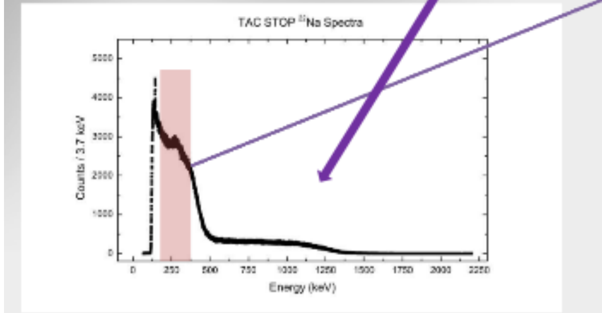
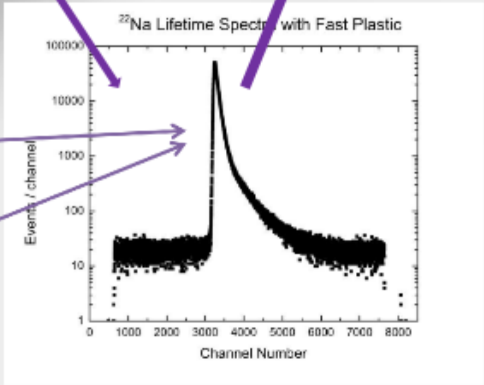






Positron lifetimes are extracted from the spectrum given by C(t)

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

Timing resolution function, R(t)

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

Multiexponential Model

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

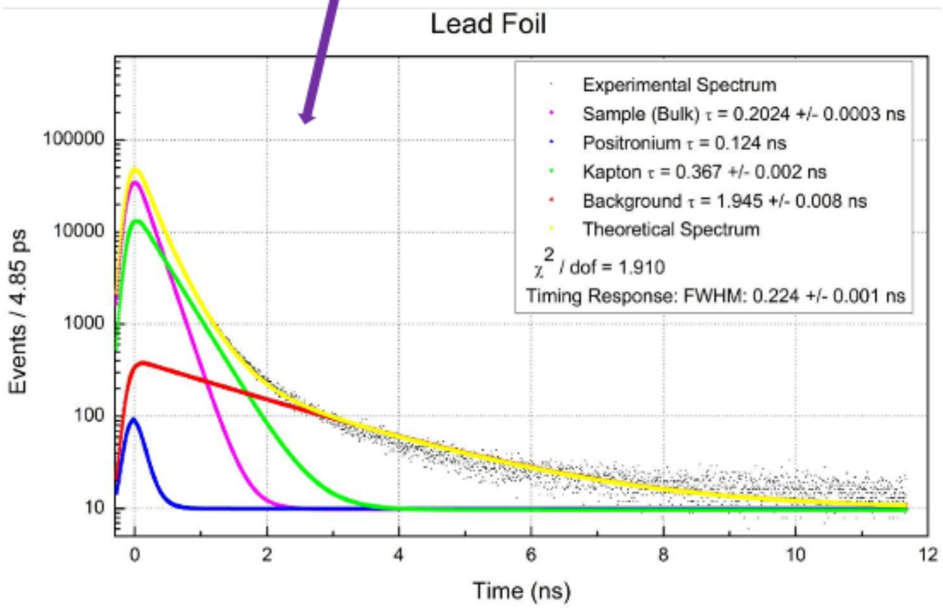
Lifetime intensity: I
Positron lifetime: τ
Annihilation rate: λ
Trapping rate: κ

Two-State Trapping Model

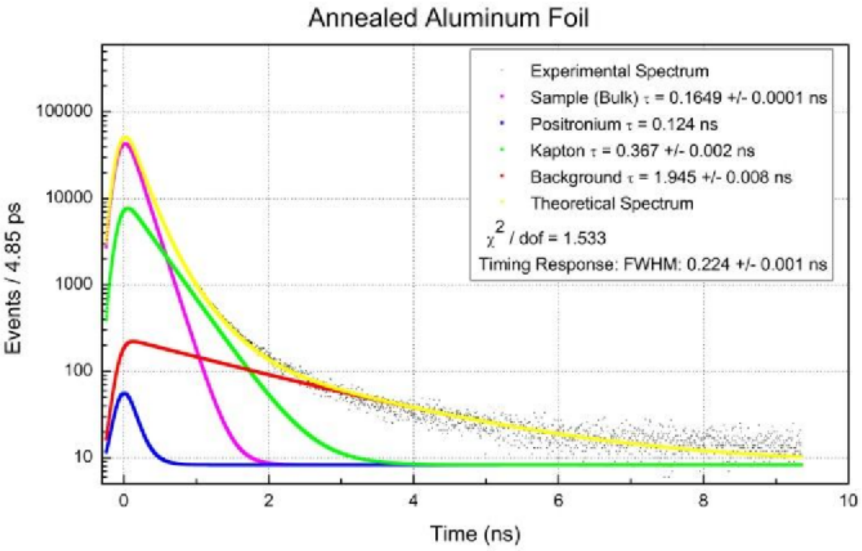
$$S(t_0) = c_b\lambda_b + \sum_{i=1}^{n} c_i\lambda_i$$

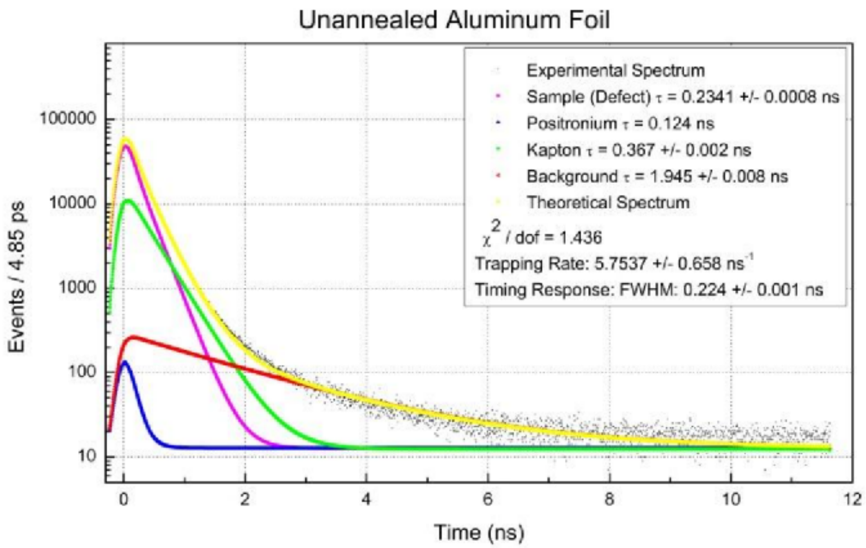
$$c_b = exp(-\lambda_b\, t)$$

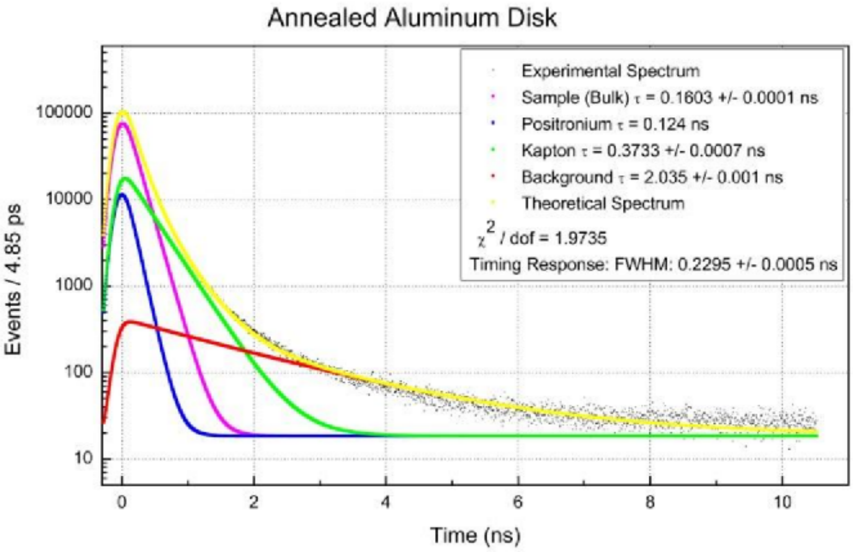
$$c_i = \frac{\kappa_i}{\lambda_b - \lambda_i}\left[exp(-\lambda_i\, t) - exp(-\lambda_b\, t)\right]$$

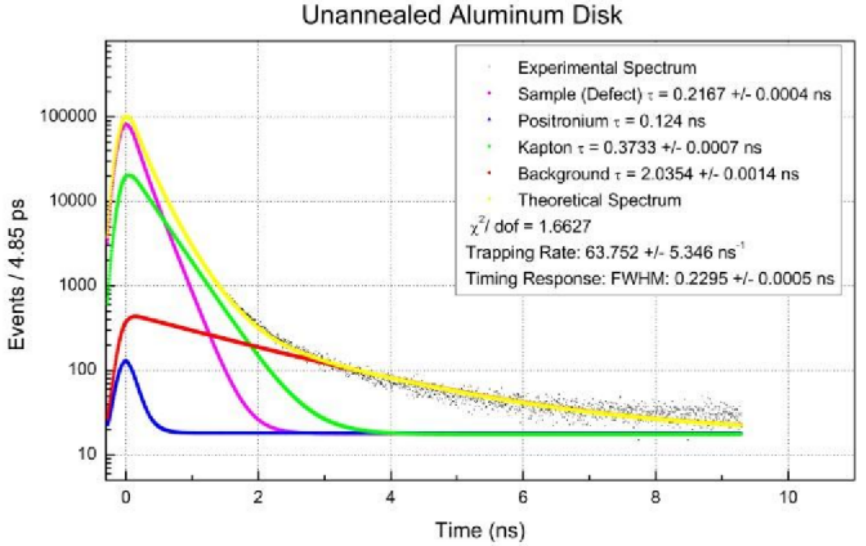


Aluminum Sample PALS Measurements







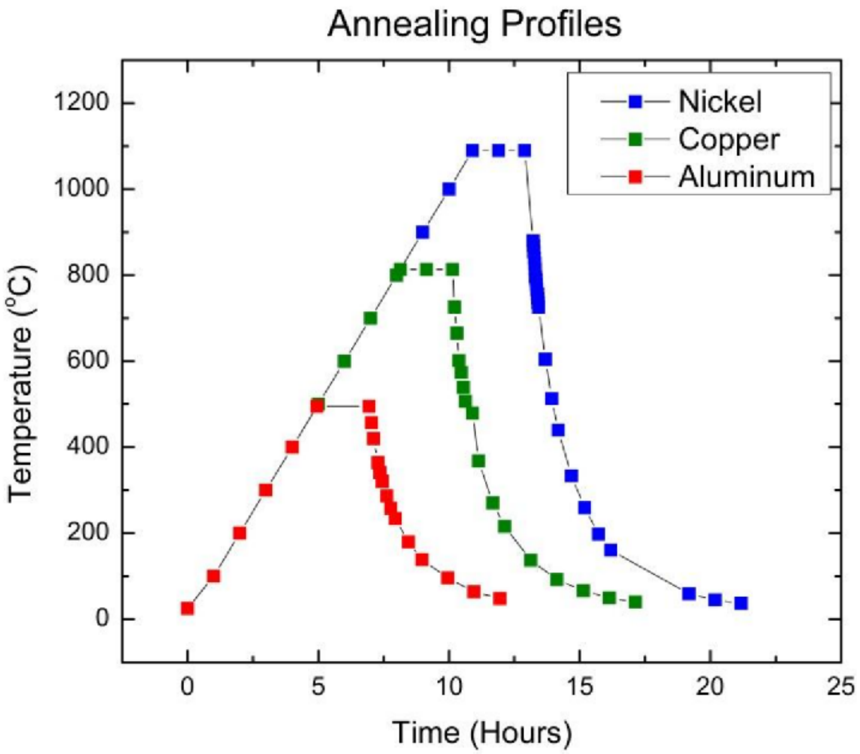


Process of Annealing Samples

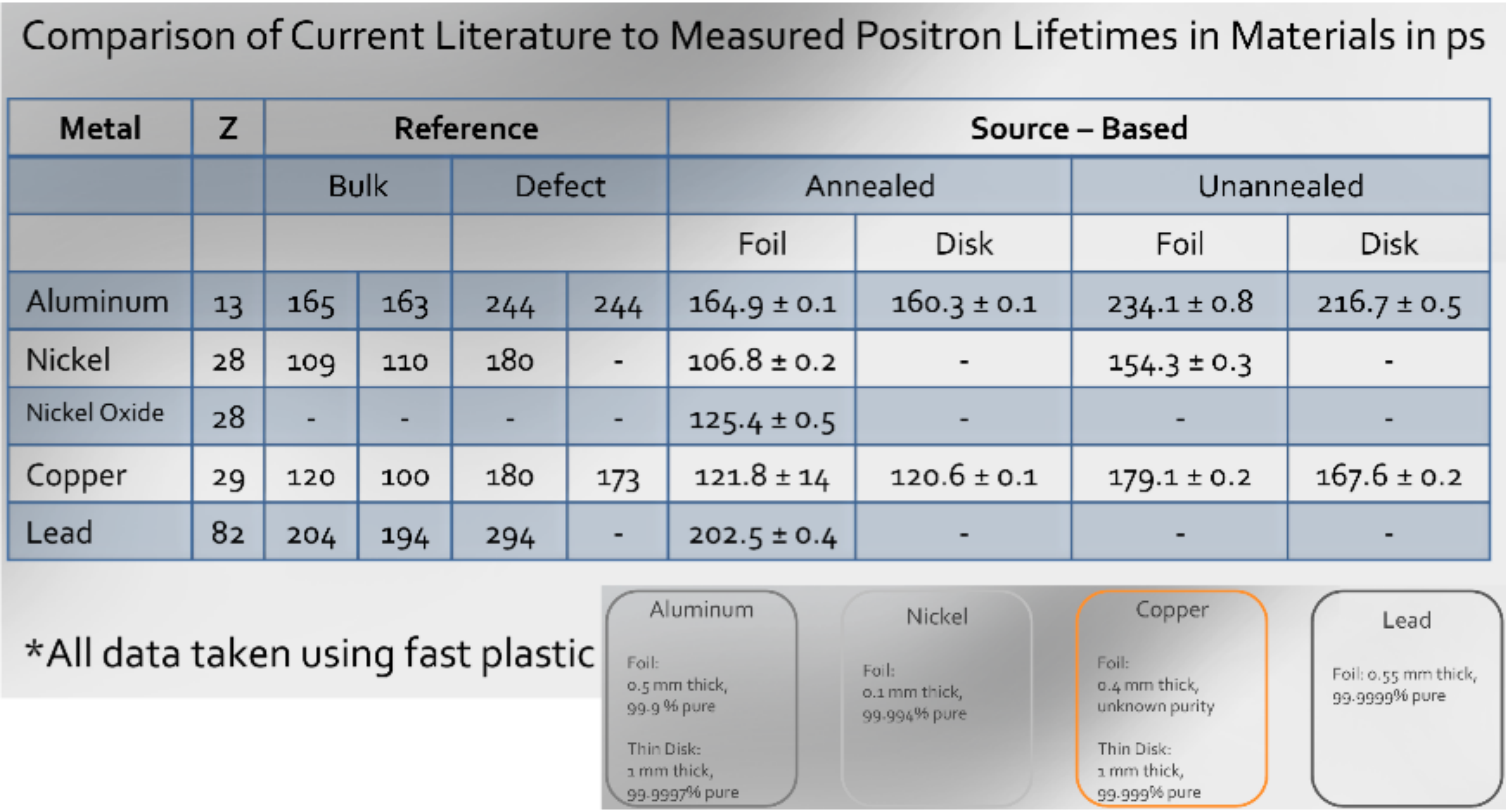
Material	Melting Point (°C)	Anneal Point (°C)
Aluminum	660	495
Nickel	1453	1090
Copper	1085	813

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

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



Summary of PALS Source-based Measurements: Apparatus benchmarking



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

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

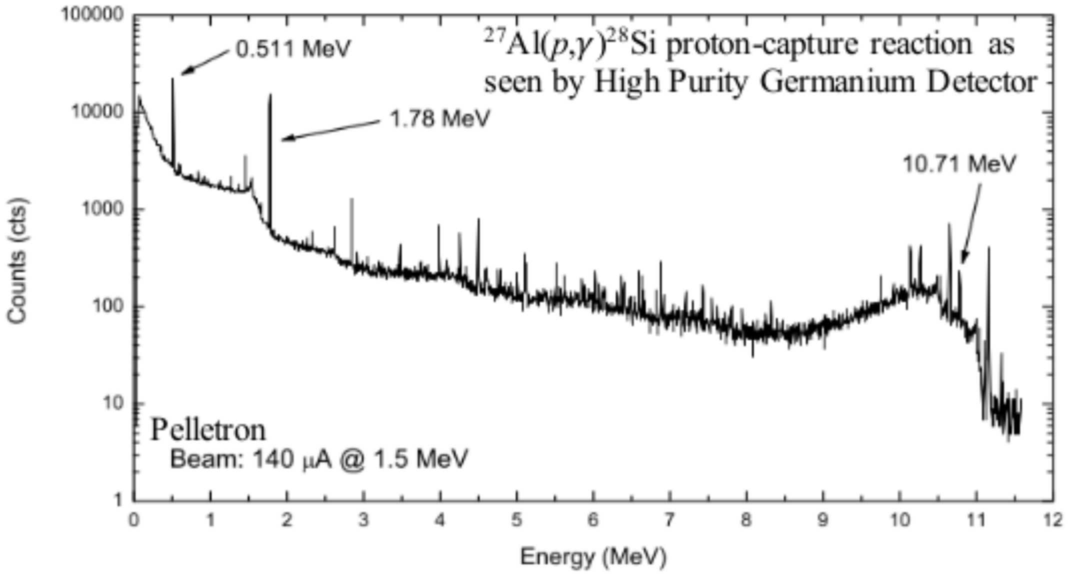
Lead

Foil: 0.55 mm thick,
99.9999% pure

Accelerator Based PALS

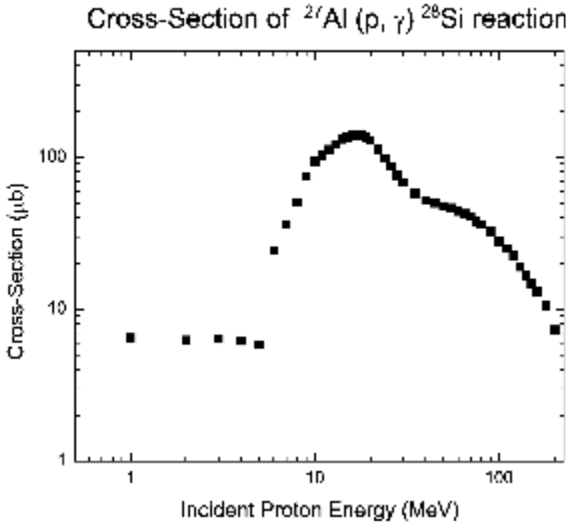
Initial Goal: Reproduce source-based lifetime measurements using the ^{27}Al(p,γ)^{28}Si proton-capture reaction: Accelerator-based Gamma-induced Positron Annihilation Spectroscopy (AG-PAS)

- 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 511 keV "death" signal



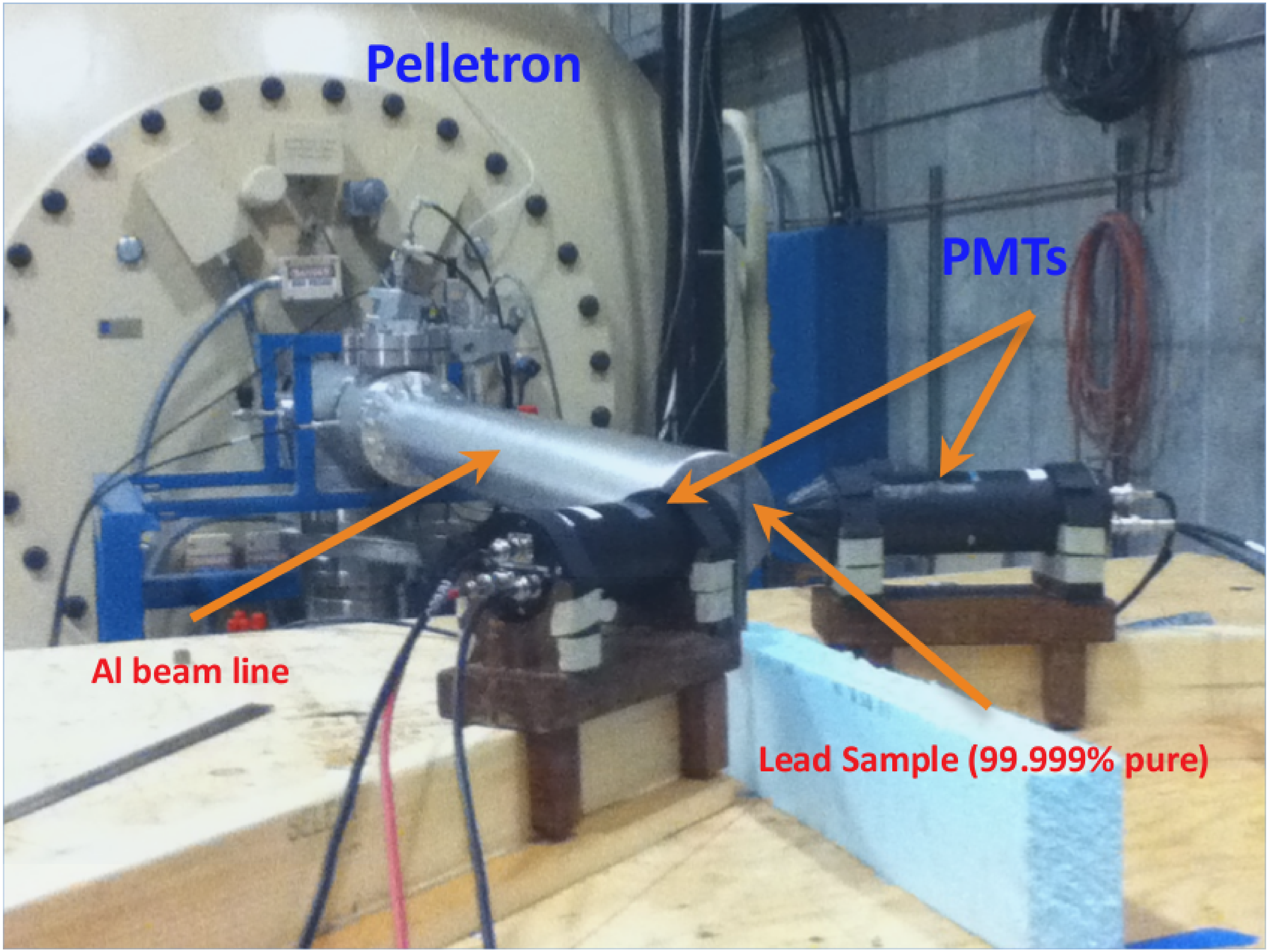
$$p + {}^{27}\mathrm{Al} \rightarrow {}^{28}\mathrm{Si}^{*} \rightarrow {}^{28}\mathrm{Si} + \gamma$$

❖ The idea is that the 1.78 MeV gamma provides the Start signal while a higher energy gamma penetrates sample (volume-wide) and pair produces—injecting a positron…



- Initial measurements used 2 MeV Van de Graaf, however 10 μA max beam current produced extremely limited event-rates (~0.2Hz)
- Pelletron capable of 200 μA and up to 8 MeV; fabricated custom Al. beamline; first AG-PALS experiments conducted May 2012

Accelerator Based Experimental Setup

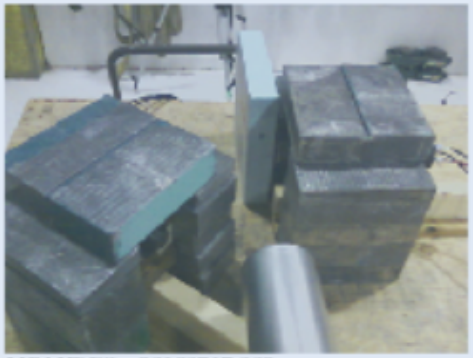


Accelerator Based PALS Feasibility Study

- Sample size and distance from Al end-cap are very important for event rate considerations

- Shielding configurations – need to block line of sight between two detectors, helped to block stop detector from line of sight to Al end-cap, although in general shielding is problematic for obvious reasons

- Event rates and proton beam energies – found that beam energy needs to be 10 - 20% above 0.992 MeV resonance; beyond 2 - 2.5 MeV, individual pmt event rates/system deadtime saturated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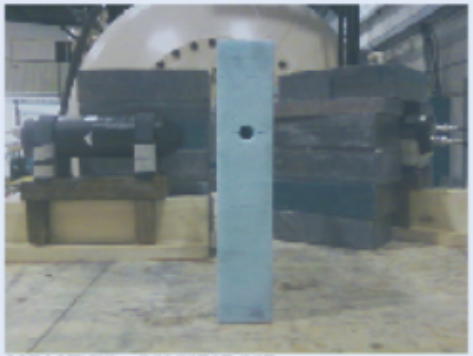
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Accelerator Based PALS : Issues to overcome

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

Accelerator Based PALS : Ideas for Future

- 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 511 keV (stop) detectors for greatly improved background rejection
- Use two CFDD – currently we have only one CFDD and one CFD – accepts more background
- Indirect connection: Use electron linac to produce high energy β^+ emitting isotopes (with additional photon, ideally); then use these to do source-based measurements

Some Summary Remarks

- Non-destructive possibilities on bulk materials are enticing and complementary to MaCS/IMCL/IRC and other assay tools available at INL
- Support next generation reactor technologies: Most notably (what I've been hearing here) in the area of cladding-related and sensor materials
- Standard source-based PALS system can readily achieve 50 μm sampling depth (practically bulk) and is compact (could be glove-box installed)
- IAC accelerator-based PAS systems need R & D, but have grand potential for volume defect assay (3D imaging)
-